Access to thiionized-, selenolized-, and alkylated 5-alkylidene 3-pyrrolin-2-one derivatives *via* regioselective oxidative cascade annulation reaction

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Supporting Information

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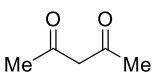
1. General information.

All compounds were fully characterized by spectroscopic data. The NMR spectra were recorded on a DRX600 (^{1}H: 600 MHz, ^{13}C: 150 MHz), chemical shifts (δ) are expressed in ppm, and J values are given in Hz, and deuterated $CDCl_3$ and DMSO-d_6 were used as solvent. The reactions were monitored by thin layer chromatography (TLC) using silica gel GF_{254}. The melting points were determined on XT-4A melting point apparatus and are uncorrected. HRMS were performed on an Agilent LC/MS TOF instrument.

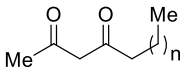
All chemicals and solvents were used as received without further purification unless otherwise stated. Column chromatography was performed on silica gel (200–300 mesh).

2,4-Pentanediones **1w** and **1z** were prepared according to the literature[1]. *N*-((2Z,4E)-4-(phenylimino)pent-2-en-2-yl)aniline **10** were prepared according to the literature[2]. 2,4-Pentanediones **1a-1n**, primary amines **2**, disulfide **3**, diselenide **4**, ether, oxidants, iodine reagents and acids were purchased from Energy Chemical and Adamas-beta®. The specific substrate structure is as follows:

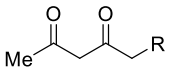
Substrate of 2,4-pentanediones **1**:



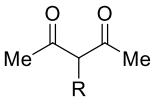
1a



n = 1, **1j**;
n = 13, **1l**;

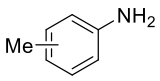


R = Ph, **1m**;
R = 4-ClC_6H_4, **1n**;

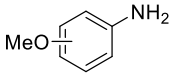


R = Me, **1w**;
R = 4-ClAr, **1z**;

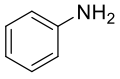
Substrate of primary amines **2**:



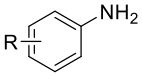
4-Me, **2a**;
3,4-Me, **2b**;
2-Me, **2c**



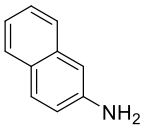
4-OMe, **2d**;



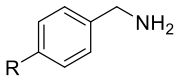
2e



3-Cl, **2f**; 4-Cl, **2g**;
3-Br, **2h**; 3-CF_3, **2i**;



2j

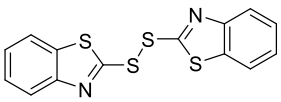


R = H, **2k**
R = Cl, **2l**

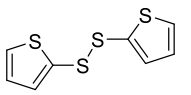
Substrate of disulfides **3**:

ArS−SAr

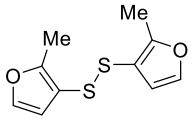
Ar = Ph, **3a**;
Ar = 4-$OMeC_6H_4$, **3l**;
Ar = 4-MeC_6H_4, **3m**;
Ar = 4-FC_6H_4, **3n**;
Ar = 4-ClC_6H_4, **3o**;
Ar = 3-ClC_6H_4, **3p**;
Ar = 4-BrC_6H_4, **3q**;
Ar = 4-$NO_2C_6H_4$, **3r**;
Ar = Bn, **3s**;



3v

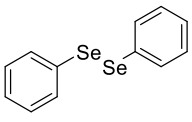


3t

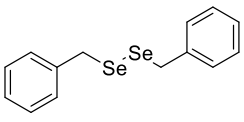


3u

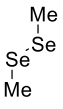
Substrate of diselenides **4**:



4a



4g



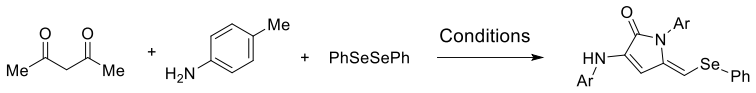
4h

Abbreviations:

TBHP: *tert*-butyl hydroperoxide; BPO: dibenzoyl peroxide; TBPB: *tert*-butyl peroxybenzoate;
CHP: cumene hydroperoxide; DTBP: di-tert-butyl peroxide; DCP: dicumyl peroxide
m-CPBA: *m*-chloroperbenzoic acid DMSO: dimethyl sulfoxide; DMF: *N*,*N*-dimethylformamide;
THF: tetrahydrofuran; DCM: dichloromethane; DCE: 1,2-dichloroethane
PE: petroleum ether; EA: ethyl acetate;

2. Optimization of reaction conditions.

Table S1. Optimization of reaction conditions.[a,b,c]

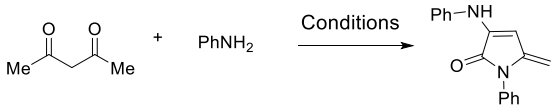


1a **2a** **5a** Ar = *4*-MePh **7a**

Entry	Oxidant (eq.)	Catalyst (mol%)	Solvent	T (°C)	Time (h)	Yield (%)
1	TBHP (2.0)	NH_4I (20)	DCE/AcOH	r.t.	14	49
2	CHP (2.0)	NH_4I (20)	DCE/AcOH	r.t.	14	56
3	DTBP (2.0)	NH_4I (20)	DCE/AcOH	r.t.	18	n.d.
4	BPO (2.0)	NH_4I (20)	DCE/AcOH	r.t.	10	32
5	TBPB (2.0)	NH_4I (20)	DCE/AcOH	r.t.	14	21
6	CHP (2.0)	NH_4I (20)	DCM/AcOH	r.t.	14	63
7	CHP (2.0)	NH_4I (20)	1.4-Dioxane/AcOH	r.t.	14	31
8	CHP (2.0)	NH_4I (20)	THF/AcOH	r.t.	14	58
9	CHP (2.0)	NH_4I (20)	DMSO/AcOH	r.t.	14	27
10	CHP (2.0)	NH_4I (20)	DMF/AcOH	r.t.	18	trace.
11	CHP (2.0)	NH_4I (20)	Toluene/AcOH	r.t.	14	19
12	CHP (2.0)	NH_4I (20)	*o*-Xylene/AcOH	r.t.	14	60
13	CHP (2.0)	NaI (20)	DCM/AcOH	r.t.	14	37
14	CHP (2.0)	KI (20)	DCM/AcOH	r.t.	15	39
15	CHP (2.0)	LiI (20)	DCM/AcOH	r.t.	13	42
16	CHP (2.0)	NH_4I (20)	DCM/AcOH	60	10	trace.
17	CHP (2.0)	NH_4I (20)	DCM/AcOH	80	10	trace.
18	CHP (1.0)	NH_4I (20)	DCM/AcOH	r.t.	14	38
19	CHP (3.0)	NH_4I (20)	DCM/AcOH	r.t.	14	14
21	CHP (2.0)	NH_4I (10)	DCM/AcOH	r.t.	14	trace
22	CHP (2.0)	NH_4I (30)	DCM/AcOH	r.t.	14	55

[a]Reaction conditions: **1a** (0.1 mmol), **2a** (0.25 mmol) **5a** (0.06 mmol), catalyst and oxidant in 2 mL of solvent (v/v = 1/1). [b]Isolated yields. [c]Determined by ^{1}H NMR of the crude product E/Z>99:1.

Table S2. Optimization of reaction conditions.[a,b]

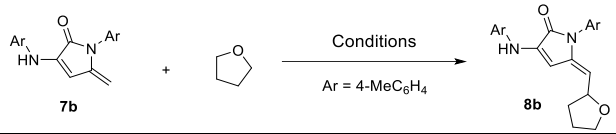


1a **2d** **7a**

Entry	Oxidant (eq.)	Catalyst (mol%)	Solvent (v/v)	T (°C)	Time (h)	Yield (%)
1	CHP (2.0)	NaI (20)	Toluene /AcOH	80	14	53
2	Oxone (2.0)	NaI (20)	Toluene/AcOH	60	15	trace.
3	$K_2S_2O_8$ (2.0)	NaI (20)	Toluene/AcOH	60	13	trace.
4	DTBP (2.0)	NaI (20)	Toluene/AcOH	60	10	32
5	TBPB (2.0)	NaI (20)	Toluene/AcOH	60	14	21
6	BPO (2.0)	NaI (20)	Toluene/AcOH	60	14	43
7	TBHP (2.0)	NaI (20)	Toluene/AcOH	60	14	55
8	TBHP (2.0)	NH_4I (20)	Toluene/AcOH	60	14	57
9	TBHP (2.0)	KI (20)	Toluene/AcOH	60	14	42
10	TBHP (2.0)	LiI (20)	Toluene/AcOH	60	18	trace.
11	TBHP (2.0)	I_2O_5 (20)	Toluene/AcOH	60	12	46
12	TBHP (2.0)	NH_4I (20)	*o*-Xylene/AcOH	60	11	56
13	TBHP (2.0)	NH_4I (20)	DCM/AcOH	60	10	37
14	TBHP (2.0)	NH_4I (20)	1.4-Dioxane/AcOH	60	12	39
15	TBHP (2.0)	NH_4I (20)	Anisose/AcOH	60	12	42
16	TBHP (2.0)	NH_4I (20)	DCE/AcOH	60	12	60
17	TBHP (2.0)	NH_4I (20)	THF/AcOH	60	12	trace.
19	TBHP (2.0)	NH_4I (20)	DMSO/AcOH	60	12	54
20	TBHP (2.0)	NH_4I (20)	MeCN/AcOH	60	12	44
21	TBHP (2.0)	NH_4I (20)	DMF/AcOH	60	12	trace
22	TBHP (2.0)	NH_4I (20)	DCE/AcOH	r.t.	12	66
23	TBHP (2.0)	NH_4I (20)	DCE/AcOH	80	12	59
24	TBHP (2.0)	NH_4I (10)	DCE/AcOH	r.t.	12	23
25	TBHP (2.0)	NH_4I (30)	DCE/AcOH	r.t.	12	58
26	TBHP (1.0)	NH_4I (20)	DCE/AcOH	r.t.	10	trace
27	TBHP (3.0)	NH_4I (20)	DCE/AcOH	r.t.	8	57

[a]Reaction conditions: **1a** (0.2 mmol), **2d** (0.5 mmol), catalyst and oxidant in 2 mL of solvent (v/v = 1/1); [b]Isolated yields.

Table S3. Optimization of reaction conditions.[a,b]

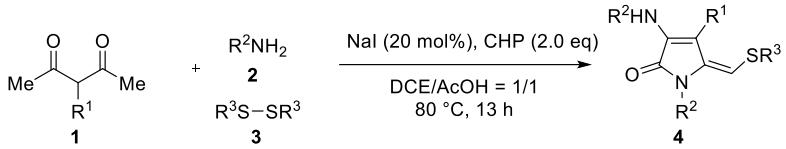


Entry	Oxidant (eq.)	Catalyst (eq.)	Atmosphere	T (°C)	Time (h)	Yield (%)
1	DTBP (2.0)	CuBr (0.4)	N_2	110	36	n.d.
2	DTBP (2.0)	Cu_2O (0.4)	N_2	110	24	51
3	DTBP (2.0)	CuCl (0.4)	N_2	110	24	trace.
4	DTBP (2.0)	$CuCl_2$ (0.4)	N_2	110	35	23
5	DTBP (2.0)	$CuSO_4$ (0.4)	N_2	110	30	n.d.
6	DTBP (2.0)	CuI (0.4)	N_2	110	36	56
7	DTBP (2.0)	Cu_2O (0.4)	N_2	110	38	51
8	DTBP (2.0)	Cu_2O (1.0.)	N_2	110	30	61
9	CHP (2.0)	Cu_2O (1.0.)	N_2	110	30	58
10	TBHP (2.0)	Cu_2O (1.0.)	N_2	110	28	53
11	DCP (2.0)	Cu_2O (1.0.)	N_2	110	26	47
12	BPO (2.0)	Cu_2O (1.0.)	N_2	110	27	trace.
13	TBPB (2.0)	Cu_2O (1.0.)	N_2	110	29	trace.
14	DTBP (2.0)	Cu_2O (1.0.)	O_2	110	36	trace.
15	DTBP (2.0)	Cu_2O (1.0.)	Air	110	34	trace.
16	DTBP (2.0)	Cu_2O (1.0.)	N_2	100	36	47
17	DTBP (2.0)	Cu_2O (1.0.)	N_2	120	9	51
18	DTBP (2.0)	\	N_2	120	6	58
19	DTBP (2.0)	H_2O	N_2	120	7	68
20	DTBP (1.0)	H_2O	N_2	120	7	35
21	DTBP (3.0)	H_2O	N_2	120	7	64
22	DTBP (5.0)	H_2O	N_2	120	7	62

[a]Reaction conditions: **7a** (0.2 mmol), catalyst in 3 mL of THF. [b]Isolated yield. [c]Determined by ^{1}H NMR of the crude product E/Z>99/1;

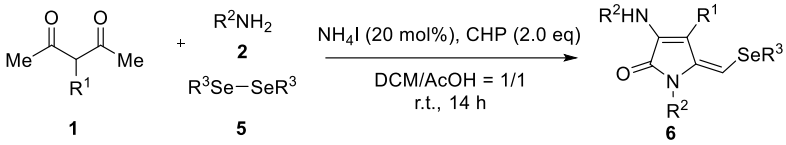
3. General procedure.

3.1 Synthesis of thiionized 5-alkylidene 3-pyrrolin-2-ones 4.



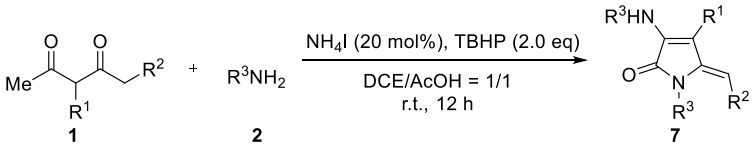
2,4-Pentanediones **1** (0.3 mmol), primary amines **2** (0.75 mmol), disulfides **3** (0.2 mmol), NaI (20 mol%), CHP (2.0 eq.) and DCE/acetic acid (3.0 mL, v/v = 1/1) were charged into a 10 mL Ace Glass tubes, and the mixture was stirred at 80℃ for 13.0 h until 2,4-pentanediones were completely consumed. EtOAc (15 mL × 2) were added. The organic phase was washed with water (10 mL), dried over Na_2SO_4, concentrated and purified by flash column chromatography to afford 5-sulfydryl-3-pyrrolin-2-ones **4**.

3.2 Synthesis of selenolized 5-alkylidene 3-pyrrolin-2-ones 6.



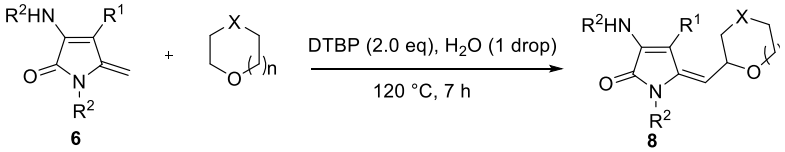
2,4-Pentanediones **1** (0.1 mmol), primary amines **2** (0.25 mmol), diselenides **5** (0.06 mmol), NH_4I (20 mol%), CHP (2.0 eq.) and DCM/acetic acid (2.0 mL, v/v = 1/1) were charged into a 10 mL Ace Glass tubes, and the mixture was stirred at r.t. for 14.0 h until 2,4-pentanediones were completely consumed. EtOAc (15 mL × 2) were added. The organic phase was washed with water (10 mL), dried over Na_2SO_4, concentrated and purified by flash column chromatography to afford 5-selenyl-3-pyrroline-2-one **6**.

3.3 Synthesis of 5-alkylidene 3-pyrrolin-2-ones 7.



2,4-Pentanediones **1** (0.2 mmol), primary amines **2** (0.5 mmol), NH_4I (20 mol%), TBHP (2.0 eq.) and DCE/acetic acid (4.0 mL, v/v = 1/1) were charged into a 10 mL Ace Glass tubes, and the mixture was stirred at r.t. for 12.0 h until 2,4-pentanediones were completely consumed. EtOAc (15 mL × 2) were added. The organic phase was washed with water (10 mL), dried over Na_2SO_4, concentrated and purified by flash column chromatography to afford 3-pyrrolin-2-ones **7**.

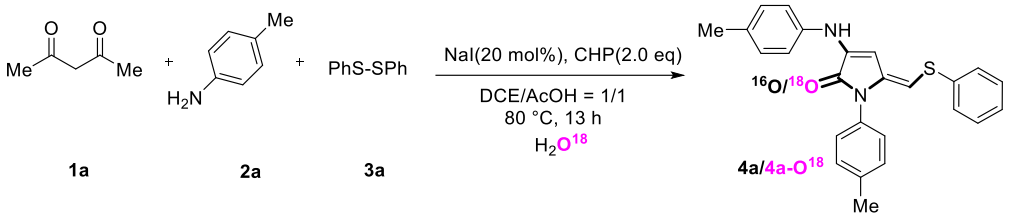
3.4 Synthesis of alkylated 5-alkylidene 3-pyrrolin-2-ones 8.



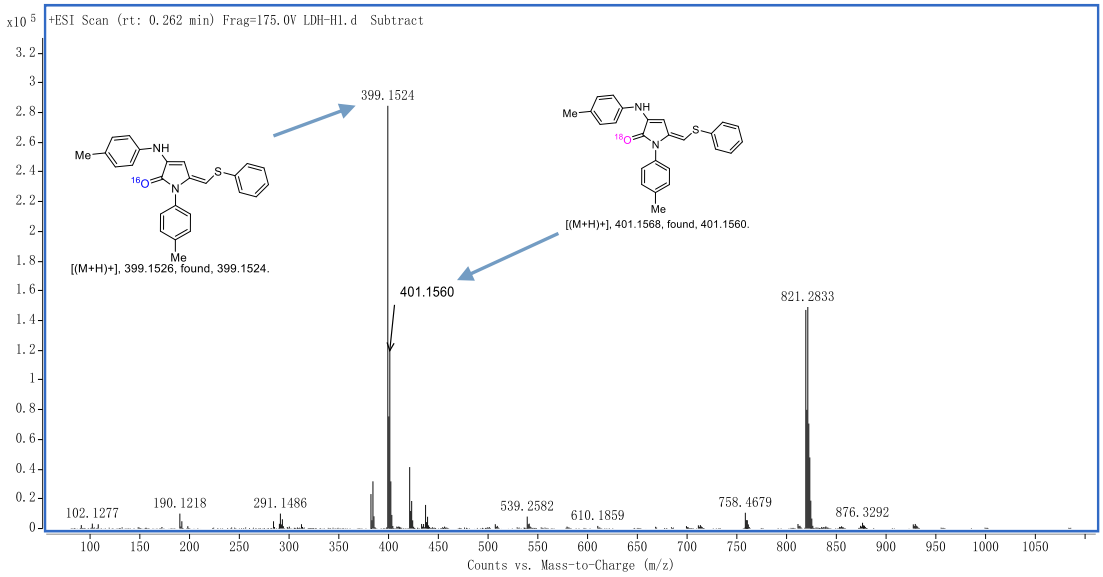
3-Pyrrolin-2-ones **6** (0.2 mmol), DTBP (2.0 eq.), H_2O (1 drop) and ether (3mL) were charged into a 10 mL Ace Glass tubes, and the mixture was stirred at 120 ℃ for 7.0 h until 3-pyrrolin-2-ones were completely consumed. EtOAc (15 mL × 2) were added. The organic phase was washed with

water (10 mL), dried over Na_2SO_4, concentrated and purified by flash column chromatography to afford 5-etheryl-3-pyrrolin-2-ones **8**.

3.5 ^{18}O labeling reaction.



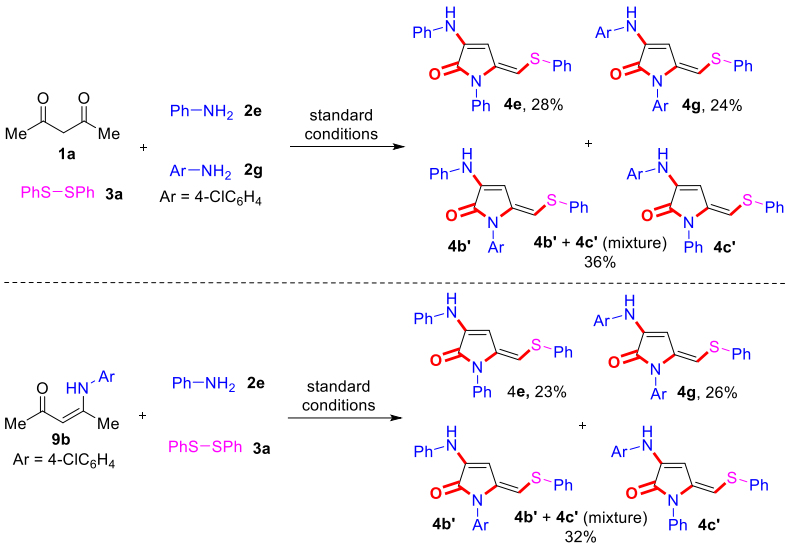
Acetylacetone **1** (0.3 mmol), primary amines **2a** (0.75 mmol), disulfide **3a** (0.2 mmol), NaI (20 mol%), CHP (2.0 eq.) and DCE/acetic acid (3.0 mL, v/v = 1/1), and H_2O^{18} (0.5 mL) were charged into a 10 mL Ace Glass tubes, and the mixture was stirred at 80℃ for 13.0 h until acetylacetone were completely consumed. EtOAc (15 mL × 2) were added. The organic phase was washed with water (10 mL), dried over Na_2SO_4, concentrated and purified by flash column chromatography to afford 5-alkylidene-3-pyrrolin-2-ones **4a/4a-O^{18}**.

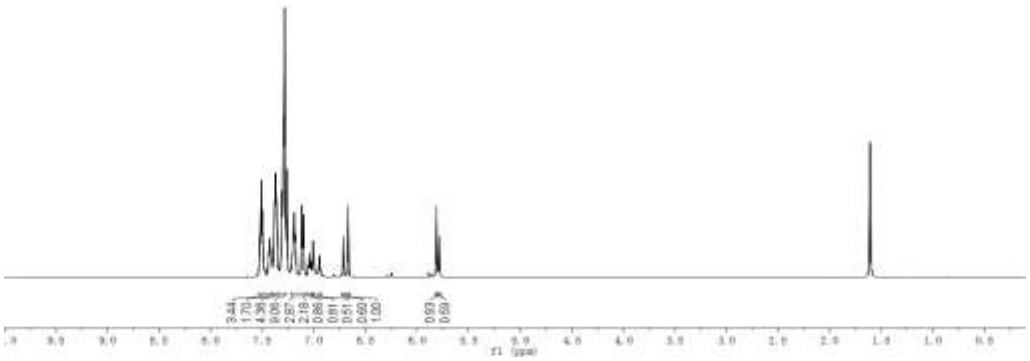


3.6 Competitive experiments.

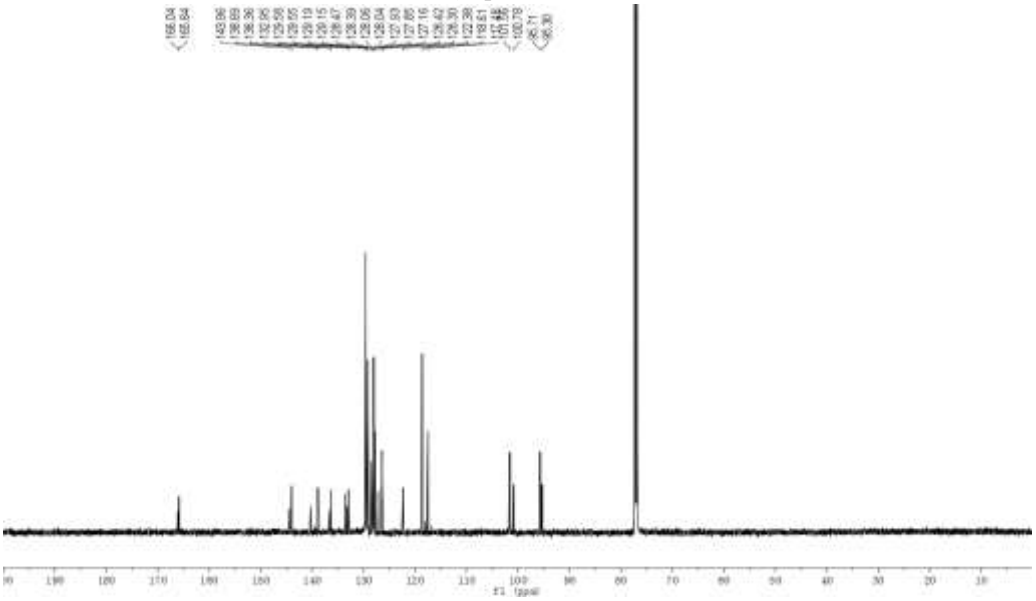
Path a: Acetylacetone **1a** (0.6 mmol), aniline **2e** (0.75 mmol), 4-chlorofenamide **2g** (0.75 mmol), disulfide **3a** (0.4 mmol), NaI (20 mol%), CHP (2.0 eq.) and DCE/acetic acid (6.0 mL, v/v = 1/1) were charged into a 20 mL Ace Glass tubes, and the mixture was stirred at 80℃ for 13.0 h until acetylacetone were completely consumed. EtOAc (15 mL × 2) were added. The organic phase was washed with water (10 mL), dried over Na_2SO_4, concentrated and purified by flash column chromatography to afford 5-alkylidene-3-pyrrolin-2-ones **4e**, **4g**, and a mixture of **4b'** and **4c'**.

Path b: *β*-Enaminone **9b** (0.6 mmol), aniline **2e** (0.75 mmol), disulfide **3a** (0.4 mmol), NaI (20 mol%), CHP (2.0 eq.) and DCE/acetic acid (6.0 mL, v/v = 1/1) were charged into a 20 mL Ace Glass tubes, and the mixture was stirred at 80℃ for 13.0 h until acetylacetone were completely consumed. EtOAc (15 mL × 2) were added. The organic phase was washed with water (10 mL), dried over Na_2SO_4, concentrated and purified by flash column chromatography to afford 5-alkylidene-3-pyrrolin-2-ones **4e**, **4g**, and a mixture of **4b'** and **4c'**.

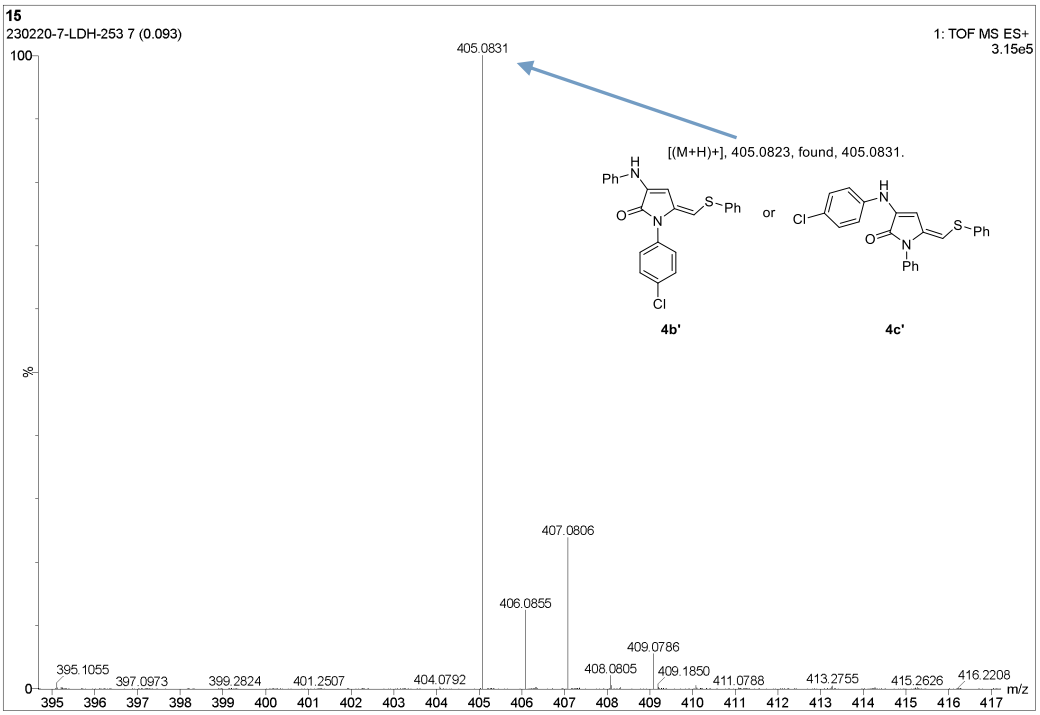




^{1}H NMR (600 MHz, $CDCl_3$) spectra of the mixture of **4b'** and **4c'**.



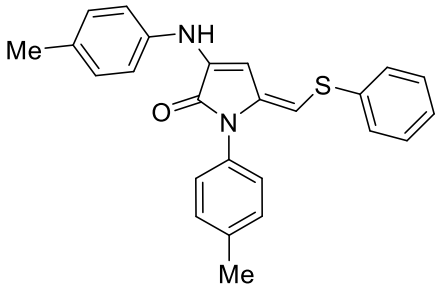
^{13}C NMR (150 MHz, $CDCl_3$) spectra of the mixture of **4b'** and **4c'**.



H RMS spectra of the mixture of **4b'** and **4c'**.

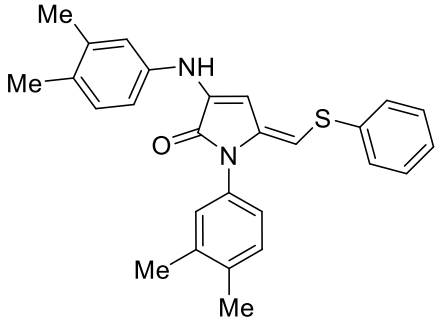
4. Spectroscopic data.

(*E*)-5-((Phenylthio)methylene)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (4 a).



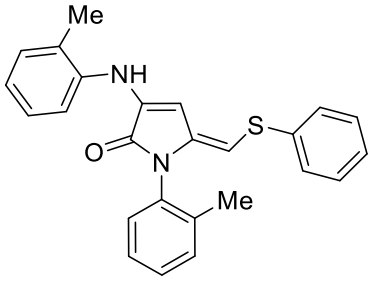
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 78 mg (66%), mp = 191–192 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.30 (d, *J* = 8.2 Hz, 2H, ArH), 7.28–7.27 (m, 4H, ArH), 7.24 (d, *J* = 8.2 Hz, 2H, ArH), 7.17 (d, *J* = 8.1 Hz, 3H, ArH), 7.08 (d, *J* = 8.3 Hz, 2H, ArH), 6.81 (s, 1H, NH), 6.62 (s, 1H, C=CH), 5.73 (s, 1H, C=CH), 2.40 (s, 3H, $ArCH_3$), 2.33 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 166.1, 144.9, 138.3, 137.7, 136.7, 133.5, 131.9, 130.9, 130.1, 130.1, 130.0, 130.0, 129.0, 129.0, 127.8, 127.8, 127.7, 127.7, 126.1, 117.5, 117.5, 99.9, 94.4, 21.2, 20.8; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{23}N_2SO$: $[(M+H)^+]$, 399.1526, found, 399.1530.

(*E*)-1-(3,4-Dimethylphenyl)-3-((3,4-dimethylphenyl)amino)-5-((phenylthio)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (4b).



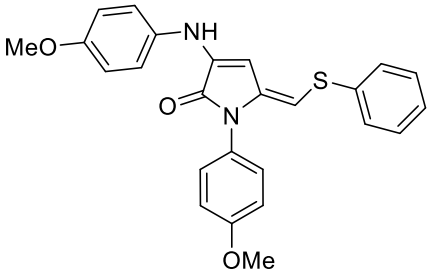
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 85 mg (67%), mp = 190–191 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.28 (d, *J* = 8.0 Hz, 2H, ArH), 7.25–7.22 (m, 3H, ArH), 7.17 (s, 1H, ArH), 7.11 (d, *J* = 8.5, 2H, ArH), 7.08 (s, 1H, ArH), 6.95 (d, *J* = 7.4, 2H, ArH), 6.78 (s, 1H, NH), 6.60 (s, 1H, C=CH), 5.71 (s, 1H, C=CH), 7.29 (s, 3H, $ArCH_3$), 2.38 (s, 3H, $ArCH_3$), 2.37 (s, 3H, $ArCH_3$), 2.33 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 166.2, 145.2, 138.1, 138.0, 137.9, 137.2, 136.9, 133.7, 131.2, 130.8, 130.6, 130.5, 129.1, 129.1, 129.1, 127.7, 127.7, 126.1, 125.4, 119.1, 114.9, 99.8, 94.3, 20.1, 19.9, 19.6, 19.2; **HRMS** (TOF ES+): m/z calcd for $C_{27}H_{27}N_2SO$: $[(M+H)^+]$, 427.1839, found, 427.1843.

(*E*)-5-((Phenylthio)methylene)-1-(*o*-tolyl)-3-(*o*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (4c).



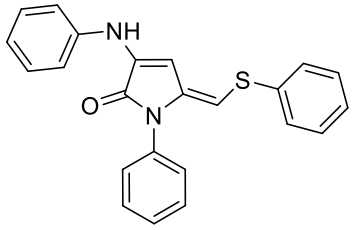
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 70 mg (59%), mp = 123–125 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.40 (d, *J* = 7.8 Hz, 1H, ArH), 7.35 (d, *J* = 3.9 Hz, 2H, ArH), 7.33 – 7.30 (m, 2H, ArH), 7.29 (d, *J* = 7.4, 2H, ArH), 7.23 (m, 4H, ArH), 7.17 (s, 1H, ArH), 6.99 (d, *J* = 7.3 Hz, 1H, ArH), 6.66 (s, 1H, NH), 6.63 (s, 1H, C=CH), 5.47 (s, 1H, C=CH), 2.35 (s, 3H, $ArCH_3$), 2.23 (s, *J* = 2.3 Hz, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 163.9, 142.6, 136.3, 135.4, 134.7, 131.8, 130.5, 129.2, 128.9, 127.3, 127.1, 127.1, 125.7, 125.7, 125.7, 125.2, 125.0, 124.9, 124.2, 120.6, 114.9, 98.2, 92.8, 15.8, 15.6; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{23}N_2SO$: [$(M+H)^+$], 399.1526, found, 399.1525.

(*E*)-1-(4-Methoxyphenyl)-3-((4-methoxyphenyl)amino)-5-((phenylthio)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (4d).



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 8:1, R_f = 0.2; Yellow solid: 62 mg (48%), mp = 212–213 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.28 (d, *J* = 8.4 Hz, 2H, ArH), 7.27–7.25 (m, 4H, ArH), 7.17 (t, *J* = 6.6 Hz, 1H, ArH), 7.13 (d, *J* = 8.8 Hz, 2H, ArH), 7.00 (d, *J* = 8.6 Hz, 2H, ArH), 6.91 (d, *J* = 8.6 Hz, 2H, ArH), 6.79 (s, 1H, NH), 6.54 (s, 1H, C=CH), 5.67 (s, 1H, C=CH), 3.84 (s, 3H, ArOMe), 3.81 (s, 3H, ArOMe); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 166.3, 159.4, 155.2, 145.4, 136.9, 134.2, 133.6, 129.4, 129.4, 129.1, 129.1, 127.7, 127.7, 126.3, 126.1, 119.3, 119.3, 114.8, 114.8, 114.8, 114.8, 99.5, 93.4, 55.6, 55.6; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{23}N_2SO$: [$(M+H)^+$], 431.1424, found, 431.1412.

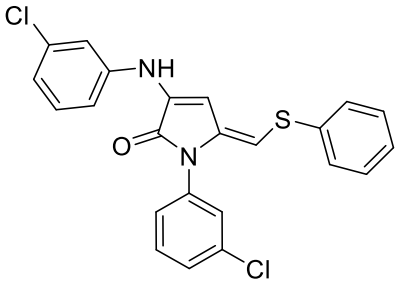
(*E*)-1-Phenyl-3-(phenylamino)-5-((phenylthio)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (4e).



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 77 mg (70%), mp = 172–173 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 8.98 (s, 1H, NH), 7.57 (d, *J* = 7.7 Hz, 2H, ArH), 7.46 (t, *J* = 6.4 Hz, 3H, ArH), 7.39 (d, *J* = 7.9 Hz, 2H, ArH), 7.36 (d, *J* = 7.4 Hz, 2H, ArH), 7.32 (d, *J* = 7.5 Hz, 2H,

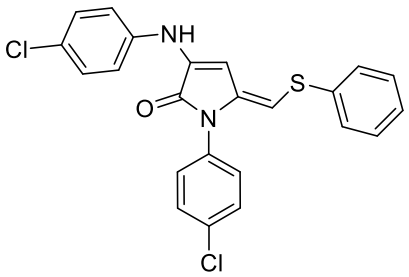
ArH), 7.29–7.27 (m, 2H, ArH), 7.20 (t, *J* = 7.3 Hz, 1H, ArH), 6.99 (t, *J* = 7.2 Hz, 1H, ArH), 6.64 (s, 1H, C=CH), 5.55 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.6, 145.5, 141.4, 136.9, 134.9, 134.1, 129.9, 129.9, 129.9, 129.9, 129.7, 129.7, 128.8, 128.7, 128.8, 127.4, 127.4, 126.6, 122.4, 118.6, 118.6, 97.6, 94.6; **HRMS** (TOF ES+): m/z calcd for $C_{23}H_{19}N_2SO$: $[(M+H)^+]$, 371.1213, found, 371.1219.

(*E*)-1-(3-Chlorophenyl)-3-((3-chlorophenyl)amino)-5-((phenylthio)methylene)-1,5-dihydro-2*H*-pyrrol-2-one (4f).



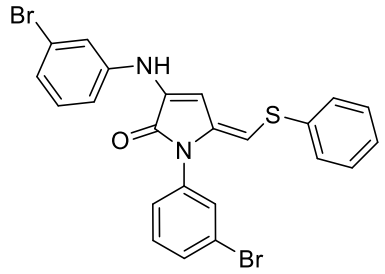
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 76 mg (63%), mp = 167–168 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 9.20 (s, 1H, NH), 7.61 (d, *J* = 5.8 Hz, 1H, ArH), 7.59 (d, *J* = 7.8 Hz, 1H, ArH), 7.55 (d, *J* = 7.8 Hz, 1H, ArH), 7.46 (d, *J* = 7.4 Hz, 1H, ArH), 7.44 (s, 1H, ArH), 7.38 (d, *J* = 6.8 Hz, 2H, ArH), 7.35 (s, 1H, ArH), 7.32 (t, *J* = 6.7 Hz, 3H, ArH), 7.23 (t, *J* = 6.7 Hz, 1H, ArH), 7.04 (d, *J* = 6.4 Hz, 1H, ArH), 6.71 (s, 1H, C=CH), 5.70 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, DMSO-d_6): δ =165.3, 144.3, 142.9, 136.5, 135.4, 134.1, 134.0, 133.9, 131.5, 131.5, 129.8, 129.8, 128.9, 128.8, 127.7, 127.7, 127.6, 126.7, 121.9, 118.3, 116.5, 99.3, 96.6; **HRMS** (TOF ES+): m/z calcd for $C_{23}H_{17}Cl_2N_2SO$: $[(M+H)^+]$, 439.0433, found, 439.0438.

(*E*)-1-(4-Chlorophenyl)-3-((4-chlorophenyl)amino)-5-((phenylthio)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (4g).



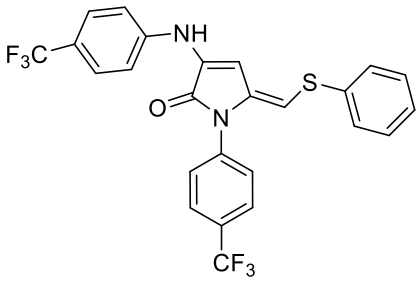
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 85mg (62%), mp = 213–214 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.48 (d, *J* = 8.3 Hz, 2H, ArH), 7.31 (d, *J* = 3.1 Hz, 2H, ArH), 7.30–7.29 (m, 3H, ArH), 7.28 (d, *J* = 7.3 Hz, 3H, ArH), 7.22 (t, *J* = 7.1 Hz, 1H, ArH), 7.09 (d, *J* = 8.4 Hz, 2H, ArH), 6.99 (s, 1H, NH), 6.66 (s, 1H, C=CH), 5.80 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.7, 143.2, 138.8, 136.0, 134.3, 132.9, 132.1, 129.8, 129.8, 129.6, 129.6, 129.3, 129.3, 129.3, 129.3, 128.1, 128.1, 127.3, 126.6, 118.7, 118.7, 102.0, 95.9; **HRMS** (TOF ES+): m/z calcd for $C_{23}H_{16}Cl_2N_2SONa$: $[(M+Na)^+]$, 461.0253, found, 461.0256.

(*E*)-1-(3-Bromophenyl)-3-((3-bromophenyl)amino)-5-((phenylthio)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (4h).



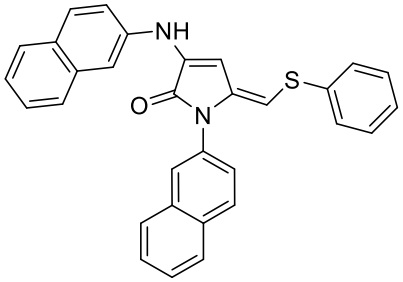
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 108 mg (69%), mp = 171–172 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.56–7.55 (m, 2H, ArH), 7.38 (t, *J* = 8.2 Hz, 2H, ArH), 7.33 (s, 1H, ArH), 7.31 (s, 1H, ArH), 7.31–7.30 (m, 3H, ArH+NH), 7.23–7.21 (m, 2H, ArH), 7.16 (d, *J* = 7.5 Hz, 1H, ArH), 7.10 (d, *J* = 7.0 Hz, 1H, ArH), 7.00–6.97 (m, 1H, ArH), 6.70 (s, 1H, C=CH), 5.84 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.5, 142.8, 141.5, 135.9, 134.8, 132.5, 131.6, 131.2, 130.9, 130.8, 129.3, 129.3, 128.2, 128.2, 126.7, 126.7, 125.4, 123.3, 122.9, 120.3, 116.0, 102.7, 96.6; **HRMS** (TOF ES+): m/z calcd for $C_{23}H_{17}Br_2N_2SO$: [$(M+H)^+$], 526.9423, found, 526.9427.

(*E*)-5-((Phenylthio)methylene)-1-(4-(trifluoromethyl)phenyl)-3-((4-(trifluoromethyl)phenyl)amino)-1,5-dihydro-*2H*-pyrrol-2-one (4i).



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 109 mg (72%), *E/Z* = 20/1, mp = 239–240 °C; **Major: ^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.79 (d, *J* = 8.2 Hz, 2H, ArH), 7.59 (d, *J* = 8.3 Hz, 2H, ArH), 7.53 (d, *J* = 8.4 Hz, 2H, ArH), 7.34–7.30 (m, 4H, ArH), 7.24 (d, *J* = 8.0 Hz, 3H, ArH), 7.18 (s, 1H, NH), 6.83 (s, 1H, C=CH), 5.92 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.5, 143.1, 141.9, 136.8, 135.6, 132.2, 130.4 (*J* = 33.1 Hz), 129.6 (minor), 129.4 (minor), 129.3, 129.3, 129.2 (minor), 128.5, 128.5, 128.2, 128.2, 127.7 (minor), 126.9, 126.9, 126.9, 126.8, 126.8, 126.8, 124.2 (d, *J* = 270.0 Hz), 124.1 (d, *J* = 33.1 Hz), 123.7 (d, *J* = 270.0 Hz), 119.8 (minor), 116.9, 116.9, 108.8 (minor), 103.8, 97.7; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{17}F_6N_2SO$: [$(M+H)^+$], 507.0960, found, 507.0958.

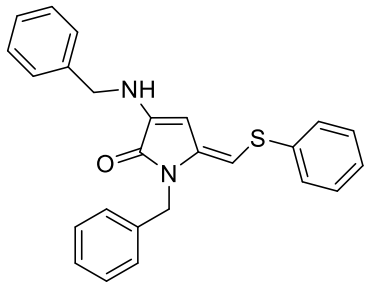
(*E*)-1-Benzyl-3-(benzylamino)-5-((phenylthio)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (4j).



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 7:1, R_f = 0.2; Yellow solid: 49 mg (35%), mp = 192–193 °C; **^{1}H NMR**

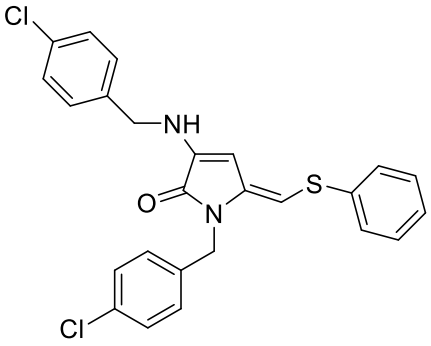
(600 MHz, $CDCl_3$): δ = 7.98 (d, *J* = 8.6 Hz, 1H , ArH), 7.92–7.88 (m, 3H, ArH), 7.81 (d, *J* = 8.1 Hz, 1H, ArH), 7.78 (dd, *J* = 8.2, 4.0 Hz, 3H, ArH), 7.59 (s, 1H, ArH), 7.56–7.54 (m, 3H, ArH), 7.49–7.46 (m, 2H, ArH), 7.40–7.37 (m, 2H, ArH), 7.31–7.28 (m, 2H, ArH), 7.19–7.17 (m, 1H, ArH), 7.15 (s, 1H, NH), 6.90 (s, 1H, C=CH), 5.88 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 166.2, 144.2, 137.8, 136.4, 134.3, 133.5, 133.0, 132.8, 131.1, 129.6, 129.6, 129.2, 129.2, 128.1, 127.9, 127.9, 127.9, 127.8, 127.1, 127.1, 126.9, 126.9, 126.9, 126.8, 126.4, 125.6, 124.5, 119.2, 112.3, 101.6, 95.9; **HRMS** (TOF ES+): m/z calcd for $C_{31}H_{23}N_2SO$: $[(M+H)^+]$, 471.1526, found, 471.1528.

(*E*)-1-Benzyl-3-(benzylamino)-5-((phenylthio)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (4k).



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 9:1, R_f = 0.2; Yellow solid: 44 mg (37%), mp = 213–214 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.37 (d, *J* = 7.3 Hz, 1H, ArH), 7.34 (t, *J* = 7.2 Hz, 5H, ArH), 7.31–7.28 (m, 2H, ArH), 7.22 (d, *J* = 7.5 Hz, 2H, ArH), 7.17 (d, *J* = 7.4 Hz, 2H, ArH), 7.11–7.09 (m, 1H, ArH), 6.97 (dd, *J* = 7.6, 1.8 Hz, 2H, ArH), 5.81 (s, 1H, C=CH), 5.59 (s, 1H, C=CH), 5.00 (t, *J* = 5.9 Hz, 1H, NH), 4.91 (s, 2H, CH_2), 4.32 (d, *J* = 5.7 Hz, 2H, CH_2); ^{13}C NMR (150 MHz, $CDCl_3$): δ = 166.2, 144.5, 140.2, 137.5, 137.3, 137.1, 128.9, 128.9, 128.8, 128.8, 128.8, 128.8, 127.8, 127.7, 127.7, 127.5, 126.9, 126.9, 126.8, 126.8, 125.6, 96.9, 91.6, 48.5, 43.3; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{23}N_2SO$: $[(M+H)^+]$, 399.1526, found, 399.1511.

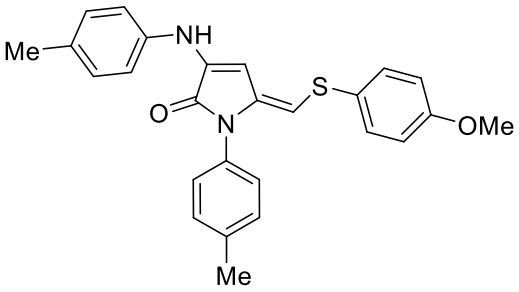
(*E*)-1-(4-Chlorobenzyl)-3-((4-chlorobenzyl)amino)-5-((phenylthio)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (4l).



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 7:1, R_f = 0.2; Light yellow solid: 35 mg (25%), mp = 135–136 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.32 (t, *J* = 8.7 Hz, 4H, ArH), 7.26 (d, *J* = 3.9 Hz, 2H, ArH), 7.20 (t, *J* = 7.6 Hz, 2H, ArH), 7.15 (d, *J* = 8.0 Hz, 2H, ArH), 7.13 (t, *J* = 8.0 Hz, 1H, ArH), 6.98 (d, *J* = 7.7 Hz, 2H, ArH), 5.77 (s, 1H, C=CH), 5.56 (s, 1H, C=CH), 5.02 (t, *J* = 5.9 Hz, 1H, NH), 4.87 (s, 2H, CH_2), 4.30 (d, *J* = 5.9 Hz, 2H, CH_2); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 166.0, 143.7, 139.9, 137.1, 135.7, 135.5, 133.5, 133.3, 128.9, 128.9, 128.9, 128.9, 128.9, 128.9, 128.9, 128.4, 128.4, 126.9, 126.9, 125.8, 125.8, 97.6, 92.0, 47.7, 42.6; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{21}Cl_2N_2SO$:

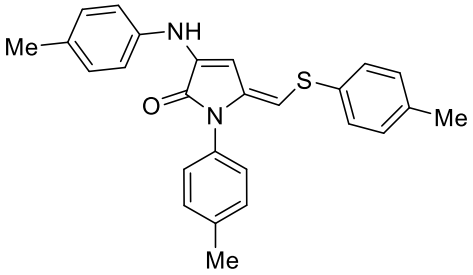
[(M+H)$^+$], 467.0746, found, 467.0751.

(*E*)-5-(((4-Bromophenyl)thio)methylene)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (4m).



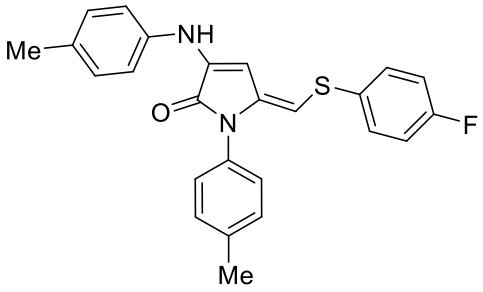
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 7:1, R_f = 0.2; Yellow solid: 74 mg (58%), mp = 165–167 ^{o}C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.28–7.25 (m, 4H, ArH), 7.21 (d, J = 8.1 Hz, 2H, ArH), 7.17 (d, J = 8.0 Hz, 2H, ArH), 7.08 (d, J = 8.3 Hz, 2H, ArH), 6.84 (d, J = 8.8 Hz, 2H, ArH), 6.80 (s, 1H, NH), 6.63 (s, 1H, C=CH), 5.69 (s, 1H, C=CH), 3.78 (s, 3H, ArOMe), 2.38 (s, 3H, $ArCH_3$), 2.33 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 166.1, 158.9, 142.9, 138.3, 137.9, 133.3, 131.9, 131.1, 131.1, 131.0, 130.1, 130.1, 130.1, 130.1, 127.9, 127.9, 126.6, 117.5, 117.5, 114.8, 114.8, 102.9, 94.6, 55.4, 21.2, 20.8; **HRMS** (TOF ES+): m/z calcd for $C_{26}H_{25}N_2SO_2$: [(M+H)$^+$], 429.1631, found, 429.1639.

(*E*)-1-(*p*-Tolyl)-3-(*p*-tolylamino)-5-((*p*-tolylthio)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (4n).



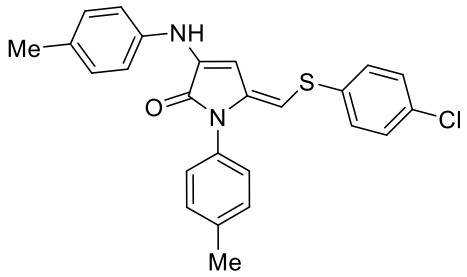
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 84 mg (68%), mp = 202–203 ^{o}C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 8.84 (s, 1H, NH), 7.33 (d, J = 7.9 Hz, 2H, ArH), 7.29–7.25 (m, 4H, ArH), 7.17 (t, J = 6.9 Hz, 3H, ArH), 7.14 (d, J = 8.1 Hz, 3H, ArH), 6.52 (s, 1H, C=CH), 5.45 (s, 1H, C=CH), 2.35 (s, 3H, $ArCH_3$), 2.25 (s, 3H, $ArCH_3$), 2.24 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.6, 144.9, 138.9, 138.3, 136.3, 135.0, 133.1, 131.5, 131.3, 130.5, 130.5, 130.4, 130.4, 130.1, 130.1, 128.5, 128.5, 128.0, 128.0, 118.7, 118.7, 98.2, 93.8, 21.2, 20.9, 20.9; **HRMS** (TOF ES+): m/z calcd for $C_{26}H_{25}N_2SO$: [(M+H)$^+$], 413.1693, found, 413.1685.

(*E*)-5-(((4-Fluorophenyl)thio)methylene)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (4o).



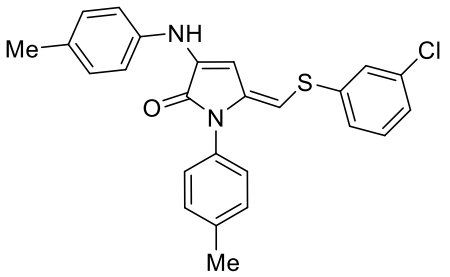
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 76 mg (61%), mp = 169–170 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.29 (d, *J* = 7.9 Hz, 2H, ArH), 7.25 (d, *J* = 5.1 Hz, 2H, ArH), 7.22 (d, *J* = 8.1 Hz, 2H, ArH), 7.17 (d, *J* = 8.1 Hz, 2H, ArH), 7.08 (d, *J* = 8.3 Hz, 2H, ArH), 6.99 (t, *J* = 8.6 Hz, 2H, ArH), 6.83 (s, 1H, NH), 6.61 (s, 1H, C=CH), 5.67 (s, 1H, C=CH), 2.40 (s, 3H, $ArCH_3$), 2.33 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 166.2, 161.7 (d, *J* = 246.5 Hz), 144.8, 138.4, 137.7, 133.7, 132.1, 131.7 (d, *J* = 3.1 Hz), 130.2 (d, *J* = 6.1 Hz), 130.2 (d, *J* = 6.1 Hz), 130.1, 130.1, 130.1, 130.1, 130.1, 129.9, 127.9, 127.9, 117.6, 116.2 (d, *J* = 22.1 Hz), 116.2 (d, *J* = 22.1 Hz), 100.4, 94.3, 21.3, 20.8; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{22}FN_2SO$: $[(M+H)^+]$, 417.1431, found, 417.1439.

(*E*)-5-(((4-Chlorophenyl)thio)methylene)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (4p).



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 8:1, R_f = 0.2; Light yellow solid: 84 mg (65%), mp = 181–182 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 8.92 (s, 1H, NH), 7.38 (d, *J* = 8.5 Hz, 2H, ArH), 7.35 (d, *J* = 8.0 Hz, 2H, ArH), 7.31 (d, *J* = 8.0 Hz, 2H, ArH), 7.29–7.26 (m, 4H, ArH), 7.16 (d, *J* = 8.1 Hz, 2H, ArH), 6.52 (s, 1H, C=CH), 5.47 (s, 1H, C=CH), 2.37 (s, 3H, $ArCH_3$), 2.26 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.8, 146.9, 138.8, 138.3, 136.4, 135.4, 131.5, 131.4, 131.0, 130.4, 130.4, 130.1, 130.1, 129.7, 129.7, 128.9, 128.9, 128.5, 128.5, 118.8, 118.8, 95.7, 93.6, 21.2, 20.9; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{22}ClN_2SO$: $[(M+H)^+]$, 433.1136, found, 433.1134.

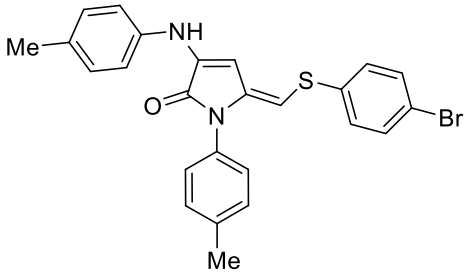
(*E*)-5-(((3-Chlorophenyl)thio)methylene)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (4q).



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 8:1, R_f = 0.2; Light yellow solid: 84 mg (65%), mp = 181–182 °C; **^{1}H**

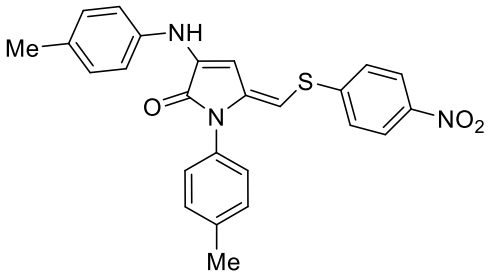
NMR (600 MHz, $CDCl_3$): δ = 7.31 (d, J = 8.1 Hz, 2H, ArH), 7.25 (d, J = 8.2 Hz, 2H, ArH), 7.22–7.19 (m, 2H, ArH), 7.18 (d, J = 7.9 Hz, 1H, ArH), 7.16 (s, 1H, ArH), 7.14–7.11 (m, 2H, ArH), 7.08 (d, J = 8.4 Hz, 2H, ArH), 6.84 (s, 1H, NH), 6.59 (s, 1H, C=CH), 5.65 (s, 1H, C=CH), 2.41 (s, 3H, $ArCH_3$), 2.33 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 166.2, 146.6, 139.3, 138.6, 137.6, 134.9, 133.9, 132.2, 130.9, 130.2, 130.2, 130.1, 130.1, 130.1, 127.8, 127.8, 126.9, 126.1, 125.3, 117.7, 117.7, 97.8, 94.1, 21.3, 20.8; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{22}ClN_2SO$: $[(M+H)^+]$, 433.1136, found, 433.1139.

(*E*)-5-(((4-Bromophenyl)thio)methylene)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (4r).



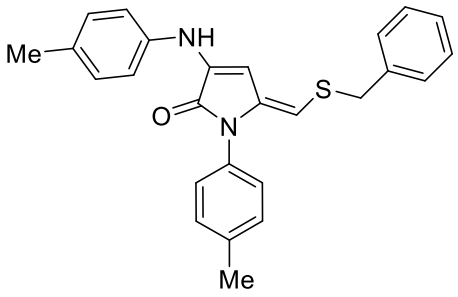
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 81 mg (57%), mp = 216–217 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.38 (d, J = 8.2 Hz, 2H, ArH), 7.30 (d, J = 7.9 Hz, 2H, ArH), 7.23 (d, J = 8.2 Hz, 2H, ArH), 7.16 (d, J = 8.0 Hz, 2H, ArH), 7.12 (d, J = 8.5 Hz, 2H, ArH), 7.07 (d, J = 8.1 Hz, 2H, ArH), 6.85 (s, 1H, NH), 6.58 (s, 1H, C=CH), 5.64 (s, 1H, C=CH), 2.40 (s, 3H, $ArCH_3$), 2.33 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 166.2, 145.9, 138.5, 137.6, 136.2, 133.9, 132.2, 132.1, 132.1, 130.9, 130.2, 130.2, 130.1, 130.1, 129.1, 129.1, 127.9, 127.9, 119.9, 117.7, 117.7, 98.6, 94.2, 21.3, 20.8; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{22}BrN_2SO$: $[(M+H)^+]$, 477.0631, found, 477.0637.

(*E*)-5-(((4-Nitrophenyl)thio)methylene)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (4s).



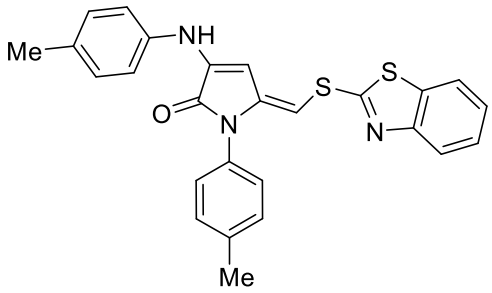
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 7:1, R_f = 0.2; Yellow solid: 60 mg (43%), mp = 242–243 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 9.00 (s, 1H, NH), 8.14 (d, J = 8.5 Hz, 2H, ArH), 7.46 (d, J = 8.4 Hz, 2H, ArH), 7.38–7.34 (m, 4H, ArH), 7.25 (d, J = 8.0 Hz, 2H, ArH), 7.14 (d, J = 8.0 Hz, 2H, ArH), 6.48 (s, 1H, C=CH), 5.50 (s, 1H, C=CH), 2.38 (s, 3H, $ArCH_3$), 2.24 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.9, 149.4, 148.0, 145.3, 138.5, 138.5, 135.9, 131.8, 131.2, 130.5, 130.5, 130.2, 130.2, 128.6, 128.6, 126.5, 126.5, 124.7, 124.7, 118.9, 118.9, 93.0, 92.3, 21.2, 20.9; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{21}N_3SO_3Na$: $[(M+Na)^+]$, 466.1196, found, 466.1193.

(*E*)-5-((Benzylthio)methylene)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (4t).



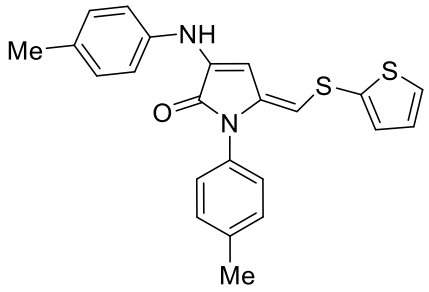
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 64 mg (52%), mp = 152–153 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.29 (t, *J* = 7.2 Hz, 2H, ArH), 7.25 (d, *J* = 6.8 Hz, 3H, ArH), 7.23 (d, *J* = 2.9 Hz, 2H, ArH), 7.15 (d, *J* = 8.0 Hz, 2H, ArH), 7.09 (d, *J* = 7.9 Hz, 2H, ArH), 7.02 (d, *J* = 8.2 Hz, 2H, ArH), 6.72 (s, 1H, NH), 6.41 (s, 1H, C=CH), 5.48 (s, 1H, C=CH), 3.84 (s, 2H, CH_2), 2.41 (s, 3H, $ArCH_3$), 2.33 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.9, 142.1, 138.1, 137.9, 137.5, 132.9, 131.7, 131.1, 130.0, 130.0, 129.9, 129.9, 128.9, 128.9, 128.6, 128.6, 127.8, 127.8, 127.3, 117.4, 117.4, 102.9, 94.8, 39.5, 21.3, 20.8; **HRMS** (TOF ES+): m/z calcd for $C_{26}H_{25}N_2SO$: $[(M+H)^+]$, 413.1682, found, 413.1688.

(*E*)-5-((Benzo[*d*]thiazol-2-ylthio)methylene)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (4u).



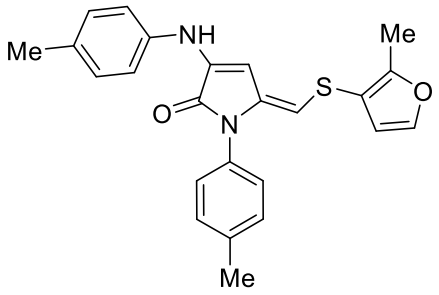
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 52 mg (38%), mp = 221–222 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 9.14 (s, 1H, NH), 8.00 (d, *J* = 5.8 Hz, 1H, ArH), 7.83 (d, *J* = 7.6 Hz, 1H, ArH), 7.46 (t, *J* = 7.0 Hz, 1H, ArH), 7.42 (d, *J* = 5.9 Hz, 2H, ArH), 7.36 (d, *J* = 7.5 Hz, 3H, ArH), 7.31–7.28 (m, 2H, ArH), 7.15 (d, *J* = 5.8 Hz, 2H, ArH), 6.57 (s, 1H, C=CH), 5.69 (s, 1H, C=CH), 2.41 (s, 3H, $ArCH_3$), 2.25 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 170.3, 166.0, 154.3, 151.3, 138.7, 138.4, 136.4, 135.3, 132.0, 131.1, 130.6, 130.6, 130.2, 130.2, 128.5, 128.5, 126.9, 124.8, 122.3, 121.7, 119.2, 119.2, 92.7, 90.8, 21.3, 20.9; **HRMS** (TOF ES+): m/z calcd for $C_{26}H_{22}N_3S_2O$: $[(M+H)^+]$, 456.1199, found, 456.1200.

(*E*)-5-((Thiophen-2-ylthio)methylene)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (4v).



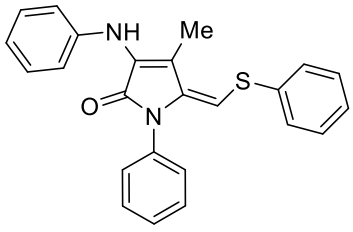
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 80 mg (66%), mp = 202–203 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 8.88 (s, 1H, NH), 7.61 (d, J = 5.2 Hz, 1H, C=CH), 7.32 (d, 2H, J = 7.8 Hz, ArH), 7.30 (d, J = 8.3 Hz, 2H, ArH), 7.21 (d, J = 8.0 Hz, 2H, ArH), 7.19 (d, J = 5.2 Hz, 1H, C=CH), 7.18 (d, J = 8.5 Hz, 2H, ArH), 7.04 (t, J = 3.3 Hz, 1H, C=CH), 6.56 (s, 1H, C=CH), 5.43 (s, 1H, C=CH), 2.35 (s, 3H, $ArCH_3$), 2.27 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.6, 143.8, 138.8, 138.4, 135.1, 134.2, 131.8, 131.4, 131.3, 130.4, 130.4, 130.2, 130.2, 130.2, 128.6, 128.5, 128.5, 118.8, 118.8, 100.7, 93.5, 21.2, 20.9; **HRMS** (TOF ES+): m/z calcd for $C_{23}H_{21}N_2S_2O$: [$(M+H)^+$], 405.1090, found, 405.1092

(*E*)-5-(((2-Methylfuran-3-yl)thio)methylene)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (4w).



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 83 mg (69%), mp = 195–196 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.26 (d, J = 6.5 Hz, 2H, ArH), 7.25 (d, J = 2.0 Hz, 1H, C=CH), 7.18 (d, J = 8.4 Hz, 2H, ArH), 7.16 (d, J = 8.5 Hz, 2H, ArH), 7.09 (d, J = 8.2 Hz, 2H, ArH), 6.81 (s, 1H, NH), 6.62 (s, 1H, C=CH), 6.31 (d, J = 2.0 Hz, 1H, C=CH), 5.48 (s, 1H, C=CH), 2.39 (s, 3H, CH_3), 2.34 (s, 3H, $ArCH_3$), 2.31 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.8, 154.1, 140.8, 140.7, 138.2, 137.9, 133.1, 131.8, 131.1, 130.1, 130.1, 130.1, 130.1, 127.8, 127.8, 117.5, 117.5, 114.3, 109.9, 104.4, 94.3, 21.3, 20.8, 11.9; **HRMS** (TOF ES+): m/z calcd for $C_{24}H_{23}N_2SO_2$: [$(M+H)^+$], 403.1475, found, 403.1483.

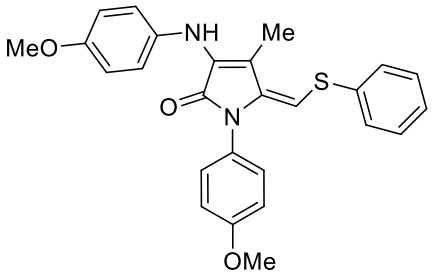
(*E*)-4-Methyl-1-phenyl-3-(phenylamino)-5-((phenylthio)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (4x).



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 7:1, R_f = 0.2; Yellow solid: 85 mg (74%), *E*/*Z* = 1/1, mp = 192–193 °C;

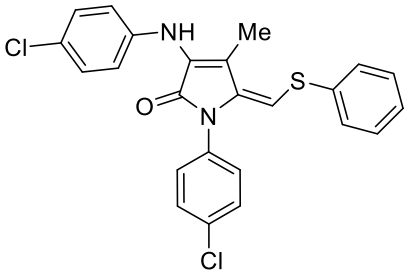
Major: ^{1}**H NMR** (600 MHz, DMSO-d_6): δ = 8.21 (s, 1H, NH) (minor), 7.93 (s, 1H, NH), 7.53 (d, J = 7.7 Hz, 2H, ArH), 7.47–7.41 (m, 3H, ArH) (minor), 7.51 (d, J = 8.3 Hz, 3H, ArH), 7.39 (d, J = 7.5 Hz, 1H, ArH), 7.35–7.34 (m, 2H, ArH), 7.22 (t, J = 7.8 Hz, 2H, ArH), 7.19 (t, J = 7.8 Hz, 2H, ArH) (minor), 6.89 (d, J = 7.8 Hz, 2H, ArH), 6.84–6.82 (m, 3H, ArH), 6.78 (t, J = 7.3 Hz, 1H, ArH) (minor), 6.12 (s, 1H, C=CH), 5.72 (s, 1H, C=CH) (minor), 2.14 (s, 3H, CH_3), 1.96 (s, 3H, CH_3) (minor); ^{13}**C NMR** (150 MHz, DMSO-d_6): δ = 167.1, 164.7 (minor), 143.8, 142.9 (minor), 140.2, 136.0, 134.6 (minor), 137.4, 130.1, 130.1, 129.9, 129.9, 129.9, 129.9, 129.7, 129.5, 129.3, 129.1, 129.1, 129.0, 129.0, 128.9 (minor), 128.8 (minor), 128.6 (minor), 128.6, 128.6, 127.6, 123.8 (minor), 120.4 (minor), 119.7 (minor), 118.4 (minor), 116.9, 105.2 (minor), 104.3, 14.6 (minor), 10.9; **HRMS** (TOF ES+): m/z calcd for $C_{24}H_{21}N_2OS$ [$(M+H)^+$], 385.1369, found, 385.1340.

(*E*)-1-(4-Methoxyphenyl)-3-((4-methoxyphenyl)amino)-4-methyl-5-((phenylthio)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (4y).



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 5:1, R_f = 0.2; Yellow solid: 103 mg (78%), *E/Z* = 1/1, mp = 142–143 °C; **Major:** ^{1}**H NMR** (600 MHz, DMSO-d_6): δ = 7.79 (s, 1H, NH) (minor), 7.67 (s, 1H, NH), 7.36 (d, J = 7.7 Hz, 2H, ArH) (minor), 7.33–7.31 (m, 3H, ArH), 7.29 (d, J = 9.3 Hz, 2H, ArH), 7.24 (d, J = 8.9 Hz, 2H, ArH), 7.05 (d, J = 8.8 Hz, 2H, ArH), 7.03 (d, J = 8.6 Hz, 2H, ArH) (minor), 6.91 (d, J = 8.7 Hz, 2H, ArH), 6.82 (d, J = 8.8 Hz, 2H, ArH), 5.95 (s, 1H, C=CH), 5.57 (s, 1H, C=CH) (minor), 3.81 (s, 3H, ArOMe), 3.78 (s, 3H, ArOMe) (minor), 3.71 (s, 3H, ArOMe), 3.70 (s, 3H, ArOMe) (minor), 2.06 (s, 3H, CH_3), 1.86 (s, 3H, CH_3) (minor); ^{13}**C NMR** (150 MHz, DMSO-d_6): δ = 167.3, 164.9 (minor), 159.6, 159.3 (minor), 154.4, 153.4 (minor), 141.8, 138.3, 136.4, 135.6, 133.4 (minor), 131.0, 130.6 (minor), 129.9, 129.9 (minor), 128.9, 128.9, 128.9, 128.9, 128.7 (minor), 128.6, 128.3, 128.3, 127.4, 127.1 (minor), 120.7 (minor), 119.9, 117.7 (minor), 115.0, 114.4, 114.4, 114.2 (minor), 112.7, 102.9 (minor), 102.2, 55.8, 55.6, 14.3, 10.9 (minor); **HRMS** (TOF ES+): m/z calcd for $C_{26}H_{25}N_2O_3S$ [$(M+H)^+$], 445.1580, found, 445.1590.

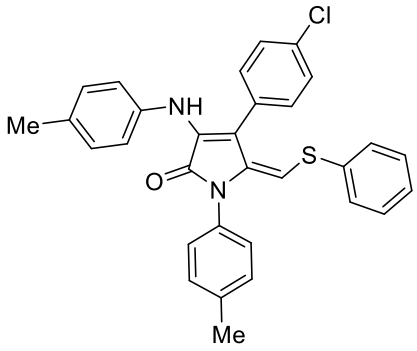
(*E*)-1-(4-Chlorophenyl)-3-((4-chlorophenyl)amino)-4-methyl-5-((phenylthio)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (4z).



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 7:1, R_f = 0.2; Yellow solid: 70 mg (52%), *E/Z* = 1/1, mp = 127–128 °C; **Major:** ^{1}**H NMR** (600 MHz, DMSO-d_6): δ = 8.13 (s, 1H, NH, minor), 8.13 (s, 1H, NH), 7.58 (d,

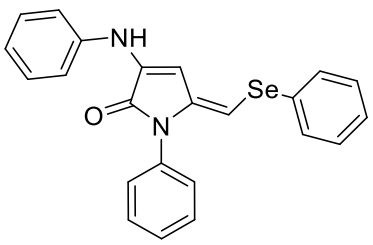
8.6 Hz, 2H, ArH), 7.58 (d, 8.6 Hz, 2H, ArH), 7.44 (d, *J* = 8.5 Hz, 2H, ArH), 7.38 (d, *J* = 8.4 Hz, 3H, ArH), 7.30–7.27 (m, 2H, ArH) (minor), 7.24 (d, *J* = 8.7 Hz, 2H, ArH), 7.21 (d, *J* = 8.7 Hz, 2H, ArH) (minor), 6.87 (d, *J* = 8.7 Hz, 2H, ArH), 6.81 (s, 2H, ArH) (minor), 6.20 (s, 1H, C=CH), 6.20 (s, 1H, C=CH) (minor), 2.16 (s, 3H, CH_3), 1.97 (s, 3H, CH_3) (minor); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 166.8, 164.4 (minor), 142.8 (minor), 142.2, 139.4, 136.9 (minor), 135.8, 135.6, 134.8 (minor), 133.4, 133.3 (minor), 133.1, 132.0 (minor), 131.4, 131.1, 130.1, 130.1, 129.9, 129.9, 129.9 (minor), 129.5(minor), 129.2, 129.2, 128.9, 128.7, 128.7, 128.6 (minor), 127.7 (minor), 127.6, 125.6, 123.6, 122.9 (minor), 120.5 (minor), 118.9, 118.1 (minor), 106.4 (minor), 105.1, 14.5, 10.8 (minor); **HRMS** (TOF ES+): m/z calcd for $C_{24}H_{19}Cl_2N_2OS$ [$(M+H)^+$], 453.0590, found, 453.0588.

(*E*)-4-(4-Chlorophenyl)-5-((phenylthio)methylene)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (4a').



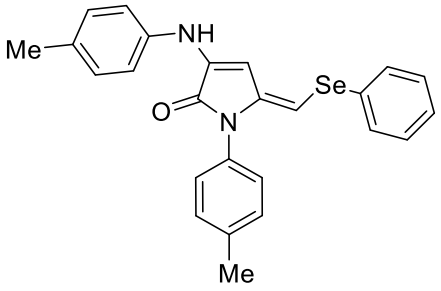
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 7:1, R_f = 0.2; Yellow solid: 94 mg (62%), *E/Z* = 5/1, mp = 205–206 °C; **Major: ^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.32 (d, *J* = 8.8 Hz, 2H, ArH), 7.28 (d, *J* = 8.3 Hz, 1H, ArH), 7.22 (d, *J* = 7.4 Hz, 1H, ArH), 7.17–7.15 (m, 3H, ArH), 7.13 (d, *J* = 6.4 Hz, 2H, ArH), 7.06 (d, *J* = 8.2 Hz, 2H, ArH), 6.73 (d, *J* = 7.9 Hz, 3H, ArH), 6.52 (d, *J* = 7.7 Hz, 1H, ArH), 6.48 (d, *J* = 8.0 Hz, 2H, ArH), 6.42 (s, 1H, NH), 5.82 (s, 1H, C=CH), 2.44 (s, 3H, $ArCH_3$), 2.18 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 166.8, 164.6 (minor), 139.0, 138.5 (minor), 137.6 (minor), 136.9 (minor), 136.6, 136.2, 136.0, 135.6 (minor), 133.6 (minor), 133.4, 133.2 (minor), 132.5, 132.4 (minor), 132.0 (minor), 131.3 (minor), 131.2 (minor), 131.9, 130.7 (minor), 130.9, 130.9, 130.2 (minor), 130.0, 130.0, 129.2, 129.2, 129.1, 129.1, 129.1, 129.1, 129.0, 129.0, 128.7, 128.7, 128.6 (minor), 128.6 (minor), 128.5 (minor), 128.4 (minor), 128.1, 128.1, 127.8 (minor), 127.0, 126.6 (minor), 121.9, 120.2, 120.2, 112.9 (minor), 109.2 (minor), 106.8 (minor), 106.5, 21.5, 21.3 (minor), 20.8 (minor), 20.7; **HRMS** (TOF ES+): m/z calcd for $C_{31}H_{26}ClN_2OS$ [$(M+H)^+$], 509.1449, found, 509.1453.

(*E*)-1-Phenyl-3-(phenylamino)-5-((phenylselanyl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (6 a).



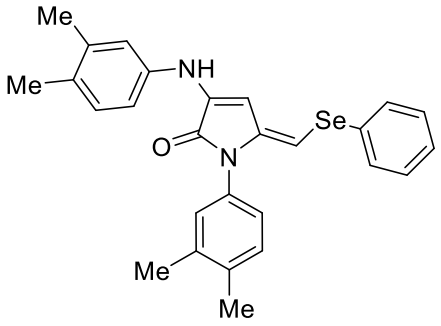
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 25 mg (61%), mp = 154–155 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.50 (t, *J* = 7.6 Hz, 2H, ArH), 7.41 (d, *J* = 7.1 Hz, 3H, ArH), 7.26 (t, *J* = 7.1 Hz, 4H, ArH), 7.27 (d, *J* = 7.2 Hz, 2H, ArH), 7.24 (t, *J* = 7.2 Hz, 1H, ArH), 7.17 (d, *J* = 7.9 Hz, 2H, ArH), 7.04 (d, *J* = 7.3 Hz, 1H, ArH), 6.91 (s, 1H, NH), 6.66 (s, 1H, C=CH), 6.07 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 166.3, 145.3, 140.2, 133.7, 133.6, 131.7, 130.6, 130.6, 129.6, 129.6, 129.5, 129.5, 129.4, 129.4, 128.3, 128.1, 128.1, 126.9, 122.4, 117.5, 117.5, 97.3, 96.6; **HRMS** (TOF ES+): m/z calcd for $C_{23}H_{19}N_2SeO$: [$(M+H)^+$], 419.0657, found, 419.0661.

(*E*)-5-((Phenylselanyl)methylene)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (6b).



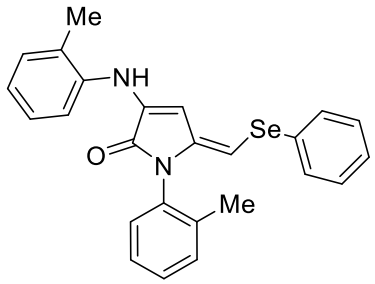
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 28 mg (63%), mp = 177–178 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.40 (d, *J* = 7.1 Hz, 2H, ArH), 7.28 (d, *J* = 7.8 Hz, 2H, ArH), 7.25 (d, *J* = 6.6 Hz, 2H, ArH), 7.22 (d, *J* = 8.1 Hz, 3H, ArH), 7.16 (d, *J* = 7.9 Hz, 2H, ArH), 7.06 (d, *J* = 8.1 Hz, 2H, ArH), 6.83 (s, 1H, NH), 6.58 (s, 1H, C=CH), 6.01 (s, 1H, C=CH), 2.39 (s, 3H, $ArCH_3$), 2.33 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 166.4, 145.8, 138.3, 137.7, 134.0, 131.9, 131.8, 131.0, 130.6, 130.6, 130.1, 130.1, 130.1, 130.1, 129.3, 129.3, 127.9, 127.9, 126.8, 117.6, 117.6, 96.5, 95.8, 21.3, 20.8; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{23}N_2SeO$: [$(M+H)^+$], 447.0970, found, 447.0973.

(*E*)-1-(3,4-Dimethylphenyl)-3-((3,4-dimethylphenyl)amino)-5-((phenylselanyl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (6c).



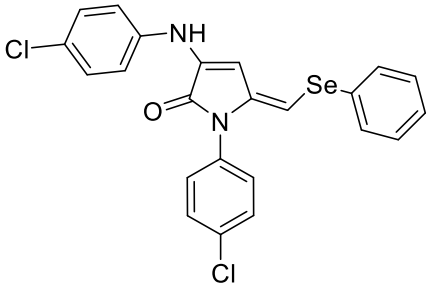
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 30 mg (64%), mp = 176–178 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.40 (d, *J* = 7.3 Hz, 2H, ArH), 7.25 (s, 2H, ArH), 7.22 (d, *J* = 7.3 Hz, 2H, ArH), 7.12 (d, *J* = 8.1 Hz, 2H, ArH), 7.06 (d, *J* = 7.7 Hz, 1H, ArH), 6.93 (d, *J* = 7.5 Hz, 2H, ArH), 6.77 (s, 1H, NH), 6.56 (s, 1H, C=CH), 5.99 (s, 1H, C=CH), 2.29 (s, 6H, $ArCH_3$), 2.27 (s, 3H, $ArCH_3$), 2.24 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 166.5, 146.1, 138.0, 137.9, 137.9, 137.1, 134.1, 131.9, 131.2, 130.8, 130.5, 130.5, 129.3, 129.3, 129.1, 126.8, 125.5, 119.2, 114.9, 96.3, 95.6, 20.1, 19.9, 19.6, 19.2; **HRMS** (TOF ES+): m/z calcd for $C_{27}H_{27}N_2SeO$: [$(M+H)^+$], 475.1283, found, 475.1287.

(*E*)-5-((Phenylselanyl)methylene)-1-(*o*-tolyl)-3-(*o*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (6d).



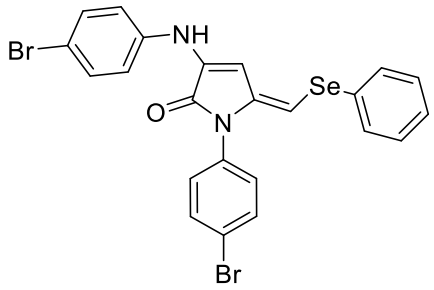
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow oli: 25 mg (57%); **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.37 (d, *J* = 7.9 Hz, 3H, ArH), 7.34 (d, *J* = 4.1 Hz, 2H, ArH), 7.30–7.28 (m, 2H, ArH), 7.25 (s, 1H, ArH), 7.24–7.22 (m, 2H, ArH), 7.21 (d, *J* = 6.9 Hz, 2H, ArH), 7.00 (t, *J* = 7.4 Hz, 1H, ArH), 6.66 (s, 1H, NH), 6.58 (s, 1H, C=CH), 5.76 (s, 1H, C=CH), 2.35 (s, 3H, $ArCH_3$), 2.21 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): 166.2, 145.6, 138.4, 137.4, 134.3, 132.6, 131.8, 131.3, 131.0, 130.5, 130.5, 129.4, 129.3, 129.3, 129.3, 127.2, 127.0, 126.8, 126.9, 122.7, 117.0, 96.6, 96.1, 17.8, 17.6; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{23}N_2SeO$: $[(M+H)^+]$, 447.0970, found, 447.0977.

(*E*)-1-(4-Chlorophenyl)-3-((4-chlorophenyl)amino)-5-((phenylselanyl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (6e).



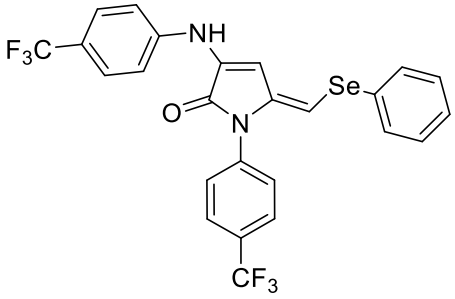
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 9:1, R_f = 0.2; Yellow solid: 25 mg (52%), *E/Z* = 33/1, mp = 200–201 ^{o}C; **Major: ^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.47 (d, *J* = 8.5 Hz, 2H, ArH), 7.41 (d, *J* = 6.9 Hz, 2H, ArH), 7.32 (d, *J* = 8.7 Hz, 2H, ArH), 7.29 (d, *J* = 8.6 Hz, 4H, ArH), 7.25 (d, *J* = 7.1 Hz, 1H, ArH), 7.09 (d, *J* = 8.7 Hz, 2H, ArH), 6.91 (s, 1H, NH), 6.60 (s, 1H, C=CH), 6.08 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.9, 144.1, 138.7, 134.3, 133.3, 132.1, 132.1 (minor), 131.2, 130.9, 130.9, 129.8, 129.8, 129.8 (minor), 129.6, 129.6, 129.5, 129.5, 129.3, 129.3, 127.4, 127.2, 118.7, 118.7, 118.0 (minor), 98.5, 97.2; **HRMS** (TOF ES+): m/z calcd for $C_{23}H_{17}N_2Cl_2SeO$: $[(M+H)^+]$, 486.9878, found, 486.9876.

(*E*)-1-(4-Bromophenyl)-3-((4-bromophenyl)amino)-5-((phenylselanyl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (6f).



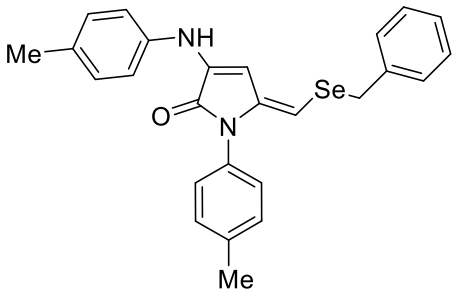
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 27 mg (47%), *E/Z* = 14/1, mp = 183–184 ^oC; **Major: ^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.62 (d, *J* = 8.5 Hz, 2H, ArH), 7.47 (d, *J* = 8.6 Hz, 2H, ArH), 7.41 (d, *J* = 7.2 Hz, 2H, ArH), 7.29 (d, *J* = 6.4 Hz, 2H, ArH), 7.23 (d, *J* = 8.5 Hz, 3H, ArH), 7.04 (d, *J* = 8.7 Hz, 2H, ArH), 6.88 (s, 1H, NH), 6.61 (s, 1H, C=CH), 6.09 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.8, 143.9, 139.2, 133.1, 132.7, 132.7, 132.6, 132.5, 132.5, 132.3 (minor), 132.3 (minor), 130.1, 130.9, 130.9, 129.6, 129.6, 129.4, 129.4, 127.2, 122.3, 118.9, 118.9, 118.4 (minor), 114.7, 104.7 (minor), 102.9 (minor), 98.6, 97.3; **HRMS** (TOF ES+): m/z calcd for $C_{23}H_{17}N_2Br_2SeO$: $[(M+H)^+]$, 574.8867, found, 574.8867.

(*E*)-5-((Phenylselanyl)methylene)-1-(4-(trifluoromethyl)phenyl)-3-((4-(trifluoromethyl)phenyl)amino)-1,5-dihydro-2*H*-pyrrol-2-one (6g).



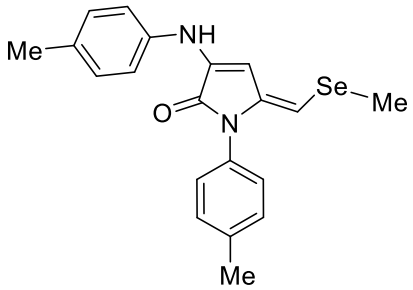
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 12 mg (21%), *E/Z* = 5/1, mp = 217–218 ^oC; **Major: ^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.78 (d, *J* = 8.1 Hz, 2H, ArH), 7.61 (d, *J* = 8.3 Hz, 2H, ArH), 7.55 (dd, *J* = 8.4, 4.3 Hz, 1H, ArH), 7.51 (d, *J* = 8.1 Hz, 2H, ArH), 7.44 (d, *J* = 7.1 Hz, 2H, ArH), 7.29 (d, *J* = 7.4 Hz, 2H, ArH), 7.22 (d, *J* = 8.3 Hz, 2H, ArH), 7.13 (s, 1H, NH), 6.77 (s, 1H, C=CH), 6.21 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 167.1 (minor), 165.7, 143.0, 142.9, 136.8, 132.6 (minor), 132.5, 131.5 (minor), 131.3, 131.3, 130.8 (minor), 130.3 (d, *J* = 33.4 Hz), 130.0 (minor), 129.6, 129.5, 129.5, 129.4, 128.2, 128.2, 127.5, 127.0 (minor), 126.9(minor), 126.8 (minor), 126.8 (minor), 126.8, 126.7, 126.6 (minor), 126.6 (minor), 124.2 (d, *J* = 268.5 Hz), 124.1 (d, *J* = 32.9 Hz), 123.7 (d, *J* = 270.0 Hz), 123.4 (minor), 123.2 (minor), 116.9, 116.9, 116.2, 106.1 (minor), 103.8 (minor), 100.2, 99.0; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{16}F_6N_2NaOSe$: $[(M+Na)^+]$, 577.0224, found, 577.0223.

(*E*)-5-((Benzylselanyl)methylene)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (6h).



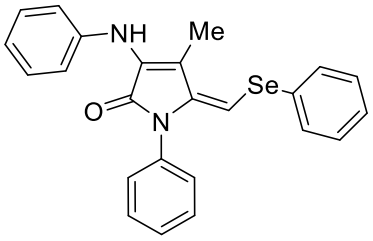
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 8:1, R_f = 0.2; Yellow solid: 19 mg (42%), mp = 158–159 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.27 (d, *J* = 7.7 Hz, 2H, ArH), 7.25 (d, *J* = 7.0 Hz, 3H, ArH), 7.20 (d, *J* = 7.6 Hz, 2H, ArH), 7.16 (d, *J* = 8.1 Hz, 2H, ArH), 7.10 (d, *J* = 8.0 Hz, 2H, ArH), 7.01 (d, *J* = 8.1 Hz, 2H, ArH), 6.69 (s, 1H, NH), 6.34 (s, 1H, C=CH), 5.75 (s, 1H, C=CH), 3.89 (s, 2H, CH_2), 2.42 (s, 3H, $ArCH_3$), 2.34 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 166.2, 144.5, 138.3, 138.1, 137.8, 133.4, 131.8, 131.1, 130.0, 130.0, 129.9, 129.9, 128.9, 128.9, 128.6, 128.6, 127.8, 127.8, 127.0, 117.5, 117.5, 97.1, 96.0, 31.5, 21.3, 20.8; **HRMS** (TOF ES+): m/z calcd for $C_{26}H_{25}N_2SeO$: $[(M+H)^+]$, 461.1127, found, 461.1134.

(*E*)-5-((Methylselanyl)methylene)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (6i).



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 8:1, R_f = 0.2; Yellow solid: 23 mg (61%), *E/Z* = 50/1, mp = 182–183 °C; **Major: ^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.29 (d, *J* = 8.0 Hz, 2H, ArH), 7.19 (d, *J* = 8.1 Hz, 2H, ArH), 7.17 (d, *J* = 8.2 Hz, 2H, ArH), 7.07 (d, *J* = 8.3 Hz, 2H, ArH), 6.78 (s, 1H, NH), 6.46 (s, 1H, C=CH), 5.80 (s, 1H, C=CH), 2.41 (s, 3H, $ArCH_3$), 2.33 (s, 3H, $ArCH_3$), 2.15 (s, 3H, CH_3); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 166.1, 141.8, 138.2, 137.9, 133.3, 131.7, 131.2, 130.0, 130.0, 130.0, 130.0, 129.2 (minor), 127.9, 127.9, 117.5, 117.5, 116.9 (minor), 99.6, 95.7, 21.3, 20.8, 8.2; **HRMS** (TOF ES+): m/z calcd for $C_{20}H_{21}N_2SeO$: $[(M+H)^+]$, 385.0814, found, 385.0819.

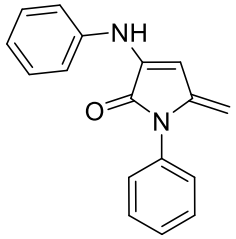
(*E*)-4-Methyl-1-phenyl-3-(phenylamino)-5-((phenylselanyl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (6j).



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 15 mg (36%), *E/Z* = 2/1, mp = 166–167 °C; **Major: ^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.46 (t, *J* = 7.6 Hz, 2H, ArH), 7.42 (d, *J* = 7.3 Hz, 2H,

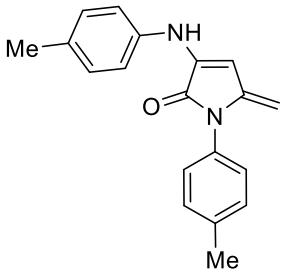
ArH), 7.37 (t, *J* = 7.4 Hz, 1H, ArH), 7.31 (d, *J* = 7.7 Hz, 3H, ArH), 7.29–7.26 (m, 5H, ArH+NH), 6.99 (t, *J* = 7.4 Hz, 1H, ArH), 6.95 (d, *J* = 7.8 Hz, 2H, ArH), 6.16 (s, 1H, C=CH), 2.16 (s, 3H, CH_3); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.4, 141.7 (minor), 141.3, 140.3, 134.8 (minor), 134.2, 138.8 (minor), 132.5, 131.6, 131.5, 131.5, 129.5, 129.5, 129.5, 129.5, 129.5, 129.5, 129.0, 129.0, 128.6, 128.6, 128.2, 127.9 (minor), 127.8 (minor), 127.6, 121.8, 121.5 (minor), 119.5 (minor), 118.8, 118.6 (minor), 117.2 (minor), 103.9, 102.6 (minor), 14.7, 11.5 (minor); **HRMS** (TOF ES+): m/z calcd for $C_{24}H_{21}N_2SeO$: [$(M+H)^+$], 433.0814, found, 433.0818.

5-Methylene-1-phenyl-3-(phenylamino)-1,5-dihydro-*2H*-pyrrol-2-one (7a).



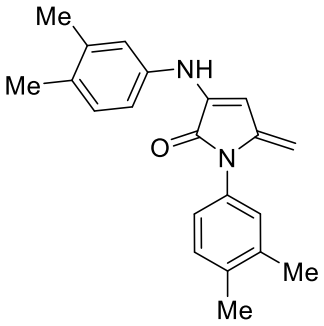
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 20:1, R_f = 0.2; Yellow solid: 34 mg (66%), mp = 179–180 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 8.64 (s, 1H, NH), 7.55 (t, *J* = 7.8 Hz, 2H, ArH), 7.45 (t, *J* = 7.4 Hz, 1H, ArH), 7.38 (d, *J* = 8.0 Hz, 3H, ArH), 7.32 (d, *J* = 7.9 Hz, 3H, ArH), 6.96 (t, *J* = 7.2 Hz, 1H, ArH), 6.59 (s, 1H, C=CH), 4.78 (s, 1H, C=CH_2), 4.48 (s, 1H, C=CH_2); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.9, 145.6, 141.8, 134.5, 133.7, 129.8, 129.8, 129.6, 129.6, 128.3, 128.3, 128.3, 121.7, 117.9, 117.9, 99.8, 92.8; **HRMS** (TOF ES+): m/z calcd for $C_{17}H_{15}N_2O$ [$(M+H)^+$], 263.1179, found, 263.1182.

5-Methylene-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (7b).



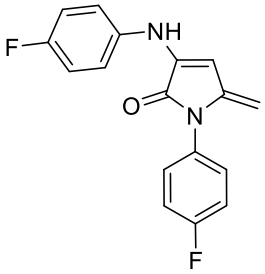
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 25:1, R_f = 0.2; Gray yellow solid: 33 mg (57%), mp = 153–155 °C; **^{1}H NMR** (600 MHz, DMSO-d_6) δ = 8.52 (s, 1H, NH), 7.33 (d, *J* = 8.1 Hz, 2H, ArH), 7.26 (d, *J* = 8.4 Hz, 2H, ArH), 7.20 (d, *J* = 8.2 Hz, 2H, ArH), 7.13 (d, *J* = 8.3 Hz, 2H, ArH), 6.48 (s, 1H, C=CH), 4.72 (s, 1H, C=CH_2), 4.43 (s, 1H, C=CH_2), 2.37 (s, 3H, $ArCH_3$), 2.26 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 166.0, 145.8, 139.4, 137.7, 133.9, 131.9, 130.5, 130.2, 130.2, 130.0, 130.0, 128.2, 128.2, 118.0, 118.0, 98.9, 92.3, 21.2, 20.8; **HRMS** (TOF ES^+): m/z calcd for $C_{19}H_{19}N_2O$ [$(M+H)^+$], 291.1492, found, 291.1491.

1-(3,4-Dimethylphenyl)-3-((3,4-dimethylphenyl)amino)-5-methylene-1,5-dihydro-*2H*-pyrrol-2-one (7c).



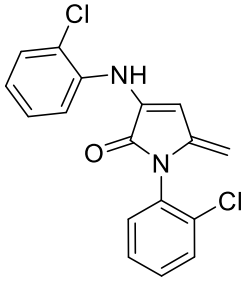
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 15:1, R_f = 0.2; Yellow solid: 19 mg (31%), mp = 159–160 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 8.37 (s, 1H, NH), 7.27 (d, J = 7.7 Hz, 1H, ArH), 7.14 (s, 1H, ArH), 7.08 (d, J = 9.6 Hz, 3H, ArH), 7.02 (d, J = 7.4 Hz, 1H, ArH), 6.47 (s, 1H, C=CH), 4.71 (s, 1H, C=CH_2), 4.40 (s, 1H, C=CH_2), 2.27 (d, J = 4.3 Hz, 6H, ArCH_3), 2.21 (s, 3H, ArCH_3), 2.16 (s, 3H, ArCH_3); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 166.0, 145.9, 139.6, 137.8, 137.3, 136.6, 133.9, 132.1, 130.5, 130.5, 129.5, 129.2, 125.7, 119.2, 115.5, 98.7, 92.3, 20.2, 19.8, 19.5, 19.2; **HRMS** (TOF ES+): m/z calcd for $C_{21}H_{23}N_2O$ [$(M+H)^+$], 319.1805, found, 319.1805.

1-(4-Fluorophenyl)-3-((4-fluorophenyl)amino)-5-methylene-1,5-dihydro-*2H*-pyrrol-2-one (7d).



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 18:1, R_f = 0.2; Yellow solid: 35 mg (59%), mp = 181–182 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 8.86 (s, 1H, NH), 7.41–7.38 (m, 4H, ArH), 7.37–7.35 (m, 2H, ArH), 7.16 (t, J = 8.2, 2H, ArH), 6.63 (s, 1H, C=CH), 4.82 (s, 1H, C=CH_2), 4.55 (s, 1H, C=CH_2); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.9, 161.7 (d, J = 245.2 Hz), 157.4 (d, J = 238.3 Hz), 145.6, 138.3, 133.9, 130.7 (d, J = 2.7 Hz), 130.6 (d, J = 8.8 Hz), 130.6 (d, J = 8.8 Hz), 119.6 (d, J = 7.8 Hz), 119.6 (d, J = 7.8 Hz), 119.6 (d, J = 7.8 Hz), 116.6 (d, J = 22.8 Hz), 116.6 (d, J = 22.8 Hz), 116.2 (d, J = 22.2 Hz), 99.2, 92.7; **HRMS** (TOF ES+): m/z calcd for $C_{17}H_{13}F_2N_2O$ [$(M+H)^+$], 299.0990, found, 299.0990.

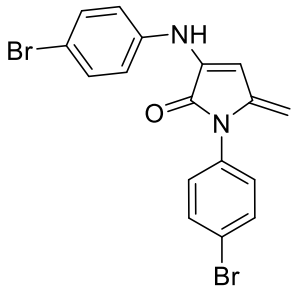
1-(2-Chlorophenyl)-3-((2-chlorophenyl)amino)-5-methylene-1,5-dihydro-*2H*-pyrrol-2-one (7e).



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 15:1, R_f = 0.2; Yellow oli: 22 mg (34%), **^{1}H NMR** (600 MHz, $CDCl_3$): δ

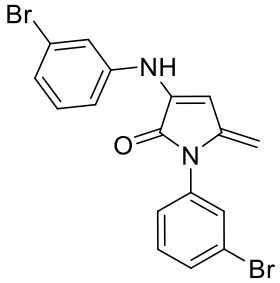
= 7.53–7.45 (m, 1H, ArH), 7.34 (t, *J* = 7.6 Hz, 4H, ArH), 7.28 (dd, *J* = 7.2, 2.4 Hz, 1H, ArH), 7.24 (t, *J* = 7.7 Hz, 1H, ArH), 7.20 (s, 1H, NH), 6.89 (t, *J* = 7.6 Hz, 1H, ArH), 6.31 (s, 1H, C=CH), 4.67 (s, 1H, C=CH_2), 4.32 (s, 1H, C=CH_2); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 164.5, 143.3, 136.1, 132.9, 131.5, 130.7, 130.1, 129.6, 129.3, 128.9, 126.7, 126.7, 121.6, 121.2, 115.4, 99.4, 92.8; **HRMS** (TOF ES+): m/z calcd for $C_{17}H_{13}Cl_2N_2O$ [$(M+H)^+$], 331.0399, found, 331.0403.

1-(4-Bromophenyl)-3-((4-bromophenyl)amino)-5-methylene-1,5-dihydro-*2H*-pyrrol-2-one (7f).



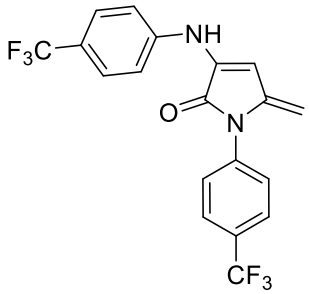
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 15:1, R_f = 0.2; Yellow solid: 25 mg (30%), mp = 161–162 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 8.86 (s, 1H, NH), 7.74 (d, *J* = 8.5 Hz, 2H, ArH), 7.48 (d, *J* = 8.7 Hz, 2H, ArH), 7.33 (dd, *J* = 8.2, 5.6 Hz, 4H, ArH), 6.63 (s, 1H, C=CH), 4.82 (s, 1H, C=CH_2), 4.55 (s, 1H, C=CH_2); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.6, 144.9, 141.2, 133.7, 133.3, 132.8, 132.8, 132.3, 132.3, 130.4, 130.4, 121.2, 119.9, 119.9, 113.0, 101.0, 93.5; **HRMS** (TOF ES+): m/z calcd for $C_{17}H_{13}Br_2N_2O$ [$(M+H)^+$], 418.9389, found, 418.9392.

1-(3-Bromophenyl)-3-((3-bromophenyl)amino)-5-methylene-1,5-dihydro-*2H*-pyrrol-2-one (7g).



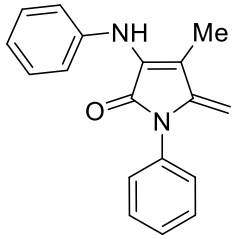
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 15:1, R_f = 0.2; Yellow solid: 25 mg (30%), mp = 151–152 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 8.88 (s, 1H, NH), 7.67 (d, *J* = 7.9 Hz, 1H, ArH), 7.61 (s, 1H, ArH), 7.55 (s, 1H, ArH), 7.51 (t, *J* = 8.0 Hz, 1H, ArH), 7.41–7.37 (m, 2H, ArH), 7.29 (t, *J* = 8.0 Hz, 1H, ArH), 7.13 (d, *J* = 7.7 Hz, 1H, ArH), 6.69 (s, 1H, C=CH), 4.88 (s, 1H, C=CH_2), 4.56 (s, 1H, C=CH_2); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.6, 144.9, 143.5, 135.9, 133.0, 131.6, 131.6, 131.3, 131.1, 127.5, 124.2, 122.6, 122.1, 120.4, 116.4, 101.8, 93.9; **HRMS** (TOF ES+): m/z calcd for $C_{17}H_{12}Br_2N_2ONa$ [$(M+Na)^+$], 440.9209, found, 440.9207.

5-Methylene-1-(4-(trifluoromethyl)phenyl)-3-((4-(trifluoromethyl)phenyl)amino)-1,5-dihydro-*2H*-pyrrol-2-one (7h).



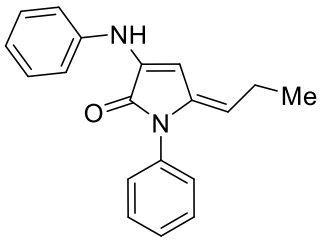
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 15:1, R_f = 0.2; Yellow solid: 43 mg (54%), mp = 209–210 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 9.18 (s, 1H, NH), 7.92 (d, J = 7.7 Hz, 2H, ArH), 7.64 (dd, J = 13.1, 8.4 Hz, 4H, ArH), 7.55 (d, J = 8.0 Hz, 2H, ArH), 6.85 (s, 1H, C=CH), 4.92 (s, 1H, C=CH_2), 4.69 (s, 1H, C=CH_2); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.5, 145.3, 144.4, 138.1, 132.8, 128.8, 128.8, 128.4 (d, J = 32.2 Hz), 126.9, 126.9, 126.8, 126.8, 125.1 (d, J = 270.0 Hz), 124.5 (d, J = 270.0 Hz), 121.4 (d, J = 31.8 Hz), 117.6, 117.6, 103.3, 94.6; **HRMS** (TOF ES+): m/z calcd for $C_{19}H_{13}F_6N_2O$ [$(M+H)^+$], 399.0927, found, 399.0927.

4-Methyl-5-methylene-1-phenyl-3-(phenylamino)-1,5-dihydro-*2H*-pyrrol-2-one (7i).



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 15:1, R_f = 0.2; Yellow solid: 17 mg (31%), mp = 178–179 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 8.09 (s, 1H, NH), 7.53 (d, J = 15.5 Hz, 2H, ArH), 7.42 (t, J = 7.4 Hz, 1H, ArH), 7.32 (d, J = 7.4 Hz, 2H, ArH), 7.22 (t, J = 7.8 Hz, 2H, ArH), 6.90 (d, J = 7.7 Hz, 2H, ArH), 6.84 (t, J = 7.3 Hz, 1H, ArH), 4.89 (d, J = 1.7 Hz, 1H, C=CH_2), 4.55 (d, J = 1.6 Hz, 1H, C=CH_2), 1.88 (s, 3H, CH_3); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.9, 146.7, 142.9, 134.9, 130.9, 129.7, 129.7, 129.1, 129.1, 128.4, 128.4, 128.1, 120.5, 118.2, 118.1, 118.1, 91.7, 11.1; **HRMS** (TOF ES+): m/z calcd for $C_{18}H_{17}N_2O$ [$(M+H)^+$], 277.1335, found, 277.1338.

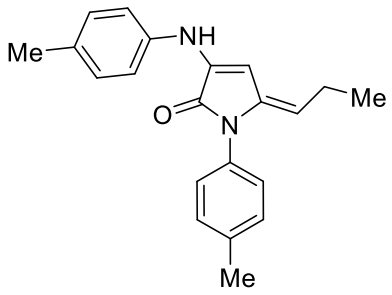
(*E*)-1-Phenyl-3-(phenylamino)-5-propylidene-1,5-dihydro-*2H*-pyrrol-2-one (7j).



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 15:1, R_f = 0.2; Yellow solid: 21 mg (36%), mp = 180–181 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 8.50 (s, 1H, NH), 7.53 (t, J = 7.6 Hz, 2H, ArH), 7.45 (d, J = 7.5 Hz, 1H, ArH), 7.41 (d, J = 8.0 Hz, 2H, ArH), 7.33–7.30 (m, 4H, ArH), 6.95 (t, J = 7.3 Hz, 1H, ArH), 6.74 (s, 1H, C=CH), 4.93 (t, J = 8.0 Hz, 1H, C=CH), 2.34 (p, J = 7.5 Hz, 2H, ArH), 0.97 (t, J = 7.4 Hz, 3H, CH_3); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.5, 141.9, 139.2, 134.8, 133.1, 129.7, 129.7, 129.6, 129.6, 128.8, 128.8, 128.3, 121.5, 117.9, 117.9, 111.9, 96.4, 20.5, 15.5; **HRMS**

(TOF ES+): m/z calcd for $C_{19}H_{19}N_2O$ [$(M+H)^+$], 291.1492, found, 291.1493.

(*E*)-5-Propylidene-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-2*H*-pyrrol-2-one (7k).

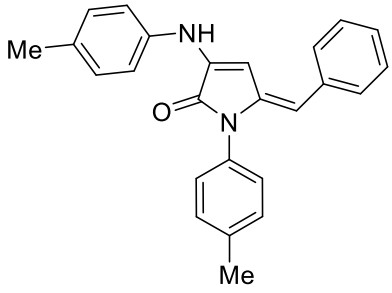


$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 27 mg (43%), mp = 194–195 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.29 (d, *J* = 8.0 Hz, 2H, ArH), 7.19 (d, *J* = 8.1 Hz, 2H, ArH), 7.16 (d, *J* = 8.1 Hz, 2H, ArH), 7.06 (d, *J* = 8.1 Hz, 2H, ArH), 6.69 (s, 1H, NH), 6.38 (s, 1H, C=CH), 5.05 (t, *J* = 8.0 Hz, 1H, C=CH), 2.41 (s, 3H, $ArCH_3$), 2.33 (s, 3H, $ArCH_3$), 2.33–2.28 (m, *J* = 7.6 Hz, 2H, CH_2), 1.04 (t, *J* = 7.5 Hz, 3H, CH_3); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.9, 139.1, 138.4, 137.9, 132.8, 131.7, 131.3, 129.9, 129.9, 129.9, 129.9, 128.1, 128.1, 117.3, 117.3, 112.6, 94.5, 21.3, 20.9, 20.8, 15.0; **HRMS** (TOF ES+): m/z calcd for $C_{21}H_{23}N_2O$ [$(M+H)^+$], 319.1805, found, 319.1805.

(*E*)-5-Pentadecylidene-1-phenyl-3-(phenylamino)-1,5-dihydro-2*H*-pyrrol-2-one (7l).

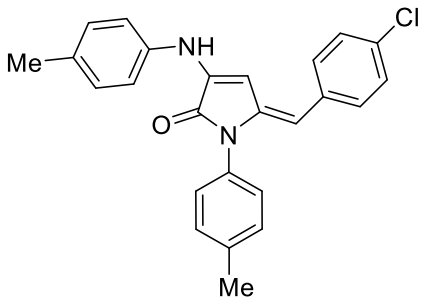
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 20:1, R_f = 0.2; Light yellow solid: 50 mg (55%), mp = 66–67 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.42 (t, *J* = 7.7 Hz, 2H, ArH), 7.33 (d, *J* = 7.4 Hz, 1H, ArH), 7.31 (d, *J* = 8.1 Hz, 2H, ArH), 7.24 (d, *J* = 7.5 Hz, 2H, ArH), 7.11 (d, *J* = 8.1 Hz, 2H, ArH), 6.94 (t, *J* = 7.3 Hz, 1H, ArH), 6.87 (s, 1H, NH), 6.39 (s, 1H, C=CH), 5.03 (t, *J* = 8.1 Hz, 1H, C=CH), 2.28 (t, *J* = 7.5 Hz, 6H, CH_2), 2.24–2.21 (m, 2H, CH_2), 1.58–1.53 (m, 7H, CH_2), 1.33 (t, *J* = 7.4, 3H, CH_3), 0.82–0.80 (m, 12H, CH_2); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 178.1, 164.2, 139.3, 137.5, 132.6, 130.8, 127.8, 127.6, 126.6, 126.3, 120.1, 115.5, 110.3, 93.9, 32.4, 30.3, 28.5, 28.1, 28.1, 28.0, 28.0, 28.0, 27.9, 27.9, 27.8, 27.8, 27.7, 25.9, 23.0, 21.1, 12.5; **HRMS** (TOF ES+): m/z calcd for $C_{31}H_{43}N_2O$ [$(M+H)^+$], 459.3370, found, 459.3369.

(*E*)-5-Benzylidene-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (7m).



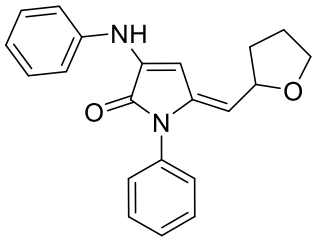
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 45 mg (61%), mp = 222–223 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.37–7.35 (m, 3H, ArH), 7.33 (d, *J* = 8.1 Hz, 3H, ArH), 7.27–7.24 (m, 3H, ArH), 7.16 (d, *J* = 8.1 Hz, 2H, ArH), 7.06 (d, *J* = 8.2 Hz, 2H, ArH), 6.82 (s, 1H, NH), 6.66 (s, 1H, C=CH), 6.03 (s, 1H, C=CH), 2.44 (s, 3H, $ArCH_3$), 2.33 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.7, 140.8, 138.3, 137.9, 136.2, 134.3, 131.7, 131.4, 130.0, 130.0, 128.9, 128.9, 128.7, 128.4, 126.7, 117.5, 117.5, 110.1, 95.1, 21.3, 20.8; **HRMS** (TOF ES+): m/z calcd for $C_{21}H_{23}N_2O$: [$(M+H)^+$], 367.1805, found, 367.1805.

(*E*)-5-(4-Chlorobenzylidene)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (7n).



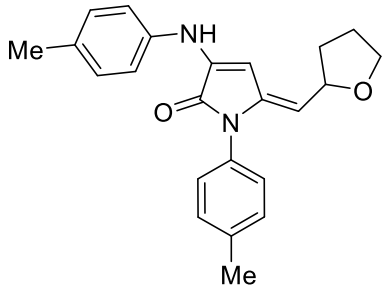
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 47 mg (59%), mp = 236–237 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.32 (t, *J* = 7.5 Hz, 4H, ArH), 7.26 (t, *J* = 5.9 Hz, 4H, ArH), 7.17 (d, *J* = 7.9 Hz, 2H, ArH), 7.05 (d, *J* = 7.9 Hz, 2H, ArH), 6.84 (s, 1H, NH), 6.57 (s, 1H, C=CH), 5.94 (s, 1H, C=CH), 2.44 (s, 3H, $ArCH_3$), 2.34 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.6, 141.2, 138.4, 137.8, 134.7, 134.6, 132.4, 131.9, 131.2, 130.1, 130.1, 130.1, 130.1, 130.0, 130.0, 128.8, 128.8, 128.4, 128.4, 117.6, 117.6, 108.6, 94.4, 21.3, 20.8; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{22}ClN_2O$: [$(M+H)^+$], 401.1415, found, 401.1417.

(*E*)-1-Phenyl-3-(phenylamino)-5-((tetrahydrofuran-2-yl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (8a).



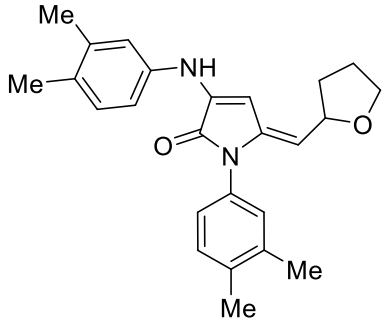
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 8:1, R_f = 0.2; Yellow solid: 46 mg (70%), mp = 191–192 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.49 (t, *J* = 7.7 Hz, 2H, ArH), 7.41 (t, *J* = 7.4 Hz, 1H, ArH), 7.36 (t, *J* = 7.7 Hz, 2H, ArH), 7.32 (d, *J* = 7.6 Hz, 2H, ArH), 7.17 (d, *J* = 7.9 Hz, 2H, ArH), 7.03 (t, *J* = 7.2 Hz, 1H, ArH), 6.85 (s, 1H, NH), 6.52 (s, 1H, C=CH), 5.07 (d, *J* = 8.9 Hz, 1H, C=CH), 4.77 (q, *J* = 8.2 Hz, 1H, CH), 3.90 (q, *J* = 7.3 Hz, 1H, CH_2), 3.82 (q, *J* = 7.5 Hz, 1H, CH_2), 2.17–2.11 (m, 1H, CH_2), 1.99–1.94 (m, 2H, CH_2), 1.63–1.58 (m, 1H, CH_2); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.9, 141.5, 140.5, 133.9, 133.1, 129.5, 129.5, 129.4, 129.4, 128.4, 128.4, 128.2, 122.1, 117.4, 117.4, 110.1, 94.9, 75.2, 68.2, 33.5, 26.4; **HRMS** (TOF ES+): m/z calcd for $C_{21}H_{21}N_2O_2$: [(M+H)+], 333.1598, found, 333.1601.

(*E*)-5-((Tetrahydrofuran-2-yl)methylene)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (8b).



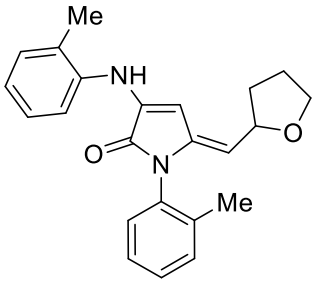
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 8:1, R_f = 0.2; Yellow solid: 48 mg (68%), mp = 194–195 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.28 (d, *J* = 8.1 Hz, 2H, ArH), 7.19 (d, *J* = 8.2 Hz, 2H, ArH), 7.15 (d, *J* = 8.2 Hz, 2H, ArH), 7.06 (d, *J* = 8.3 Hz, 2H, ArH), 6.76 (s, 1H, NH), 6.43 (s, 1H, C=CH), 5.03 (d, *J* = 9.0 Hz, 1H, C=CH), 4.75 (t, *J* = 8.5, 1H, CH), 3.89 (q, *J* = 7.4 Hz, 1H, CH), 3.81 (q, *J* = 8.0, 7.6 Hz, 1H, CH), 2.40 (s, 3H, $ArCH_3$), 2.33 (s, 3H, $ArCH_3$), 2.15–2.10 (m, 1H, CH), 1.99–1.91 (m, 2H, CH_2), 1.62–1.57 (m, 1H, CH_2); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 166.1, 141.8, 138.1, 138.0, 133.5, 131.7, 131.3, 129.9, 129.9, 129.9, 129.9, 128.2, 128.2, 117.6, 117.6, 109.6, 94.1, 75.3, 68.1, 33.4, 26.4, 21.3, 20.8; **HRMS** (TOF ES+): m/z calcd for $C_{23}H_{25}N_2O_2$: [$(M+H)^+$], 361.1911, found, 361.1913.

(*E*)-1-(3,4-Dimethylphenyl)-3-((3,4-dimethylphenyl)amino)-5-((tetrahydrofuran-2-yl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (8c).



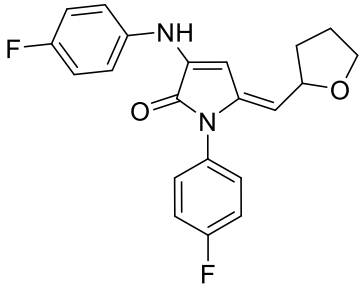
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 7:1, R_f = 0.2; Yellow solid: 28 mg (56%), mp = 174–175 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.23 (d, *J* = 7.8 Hz, 1H, ArH), 7.11 (d, *J* = 8.6 Hz, 1H, ArH), 7.08 (s, 1H, ArH), 7.03 (d, *J* = 7.7 Hz, 1H, ArH), 6.93 (d, *J* = 4.9 Hz, 2H, ArH), 6.67 (s, 1H, NH), 6.41 (s, 1H, C=CH), 5.00 (d, *J* = 9.0 Hz, 1H, C=CH), 4.75 (q, *J* = 8.2 Hz, 1H, CH), 3.91–3.87 (m, 1H, CH_2), 3.82 (q, *J* = 8.9, 8.0 Hz, 1H, CH_2), 2.30–2.28 (m, 9H, $ArCH_3$), 2.24 (s, 3H, $ArCH_3$), 2.12 (dq, *J* = 12.4, 6.4 Hz, 1H, CH_2), 1.96 (q, *J* = 7.4 Hz, 2H, CH_2), 1.61–1.57 (m, 1H, CH_2); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 166.1, 141.9, 138.3, 137.8, 137.8, 136.9, 133.5, 131.5, 130.5, 130.4, 130.3, 129.4, 125.7, 119.1, 114.8, 109.5, 93.8, 75.3, 68.1, 33.4, 26.3, 20.1, 19.9, 19.6, 19.1; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{29}N_2O_2$: [$(M+H)^+$], 389.2224, found, 389.2225.

(*E*)-5-((Tetrahydrofuran-2-yl)methylene)-1-(*o*-tolyl)-3-(*o*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (8d).



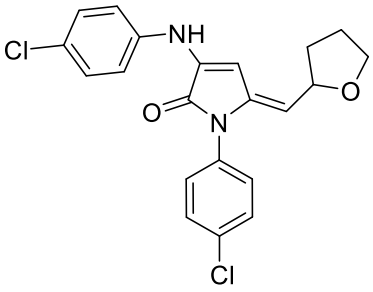
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 8:1, R_f = 0.2; Yellow oli: 37 mg (52%); **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.37 (d, *J* = 7.9 Hz, 2H, ArH), 7.34 (s, 2H, ArH), 7.24–7.21 (m, 2H, ArH), 7.17 (d, *J* = 7.6 Hz, 1H, ArH), 6.99 (t, *J* = 7.3 Hz, 1H, ArH), 6.57 (s, 1H, NH), 6.43 (s, 1H, C=CH), 4.79 (d, *J* = 9.0 Hz, 1H, C=CH), 4.74–4.69 (m, 1H, CH), 3.93–3.85 (m, 1H, CH), 3.79 (m, 1H, CH), 2.34 (d, 6H, $ArCH_3$), 2.19–2.14 (m, 2H, CH_2), 1.98–1.83 (m, 1H, CH), 1.67–1.43 (m, 1H, CH); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.7, 141.2, 138.7, 137.7, 133.7, 132.8, 131.2, 130.9, 129.7, 129.4, 129.0, 127.0, 122.4, 122.4, 116.9, 109.8, 94.5, 75.2, 68.1, 33.5, 26.2, 17.7, 17.5; **HRMS** (TOF ES+): m/z calcd for $C_{23}H_{25}N_2O_2$: $[(M+H)^+]$, 361.1911, found, 361.1917.

(*E*)-1-(4-Fluorophenyl)-3-((4-fluorophenyl)amino)-5-((tetrahydrofuran-2-yl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (8e).



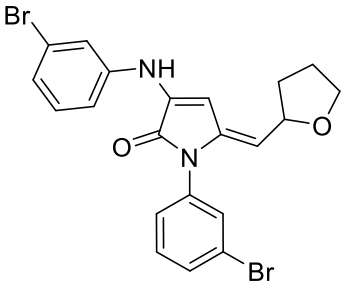
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 8:1, R_f = 0.2; Yellow solid: 49 mg (67%), mp = 204–205 ^{o}C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.29 (dd, *J* = 8.7, 4.9 Hz, 2H, ArH), 7.19 (d, *J* = 8.4 Hz, 2H, ArH), 7.12 (dd, *J* = 8.9, 4.5 Hz, 2H, ArH), 7.05 (t, *J* = 8.5 Hz, 2H, ArH), 6.80 (s, 1H, NH), 6.43 (s, 1H, C=CH), 5.01 (d, *J* = 8.9 Hz, 1H, C=CH), 4.73 (q, *J* = 8.3 Hz, 1H, CH), 3.90 (q, *J* = 7.2 Hz, 1H, CH_2), 3.82 (q, *J* = 7.8 Hz, 1H, CH_2), 2.14 (dq, *J* = 12.2, 6.8 Hz, 1H, CH_2), 1.97 (dt, *J* = 10.3, 7.0 Hz, 2H, CH_2), 1.64–1.59 (m, 1H, CH_2); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.9, 162.13 (d, *J* = 248.2 Hz), 158.19 (d, *J* = 242.1 Hz), 141.48, 136.59, 136.58, 133.53, 130.17 (d, *J* = 8.6 Hz), 130.17 (d, *J* = 8.6 Hz), 129.78 (d, *J* = 3.1 Hz), 119.13 (d, *J* = 7.8 Hz), 119.13 (d, *J* = 7.8 Hz), 116.30 (d, *J* = 47.1 Hz), 116.30 (d, *J* = 47.1 Hz), 116.30 (d, *J* = 47.1 Hz), 110.09, 94.27, 75.1, 68.21, 33.44, 26.34; **HRMS** (TOF ES+): m/z calcd for $C_{21}H_{19}F_2N_2O_2$: $[(M+H)^+]$, 369.1409, found, 369.1413.

(*E*)-1-(4-Chlorophenyl)-3-((4-chlorophenyl)amino)-5-((tetrahydrofuran-2-yl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (8f).



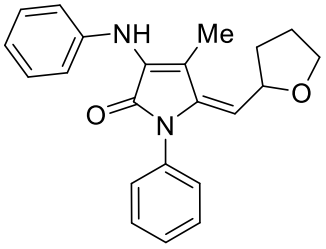
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 8:1, R_f = 0.2; Yellow solid: 51 mg (64%), mp = 226–227 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.47 (d, *J* = 8.3 Hz, 2H, ArH), 7.31 (d, *J* = 8.4 Hz, 2H, ArH), 7.26 (d, *J* = 7.3 Hz, 2H, ArH), 7.09 (d, *J* = 8.1 Hz, 2H, ArH), 6.82 (s, 1H, NH), 6.49 (s, 1H, C=CH), 5.07 (d, *J* = 8.8 Hz, 1H, , C=CH), 4.75 (q, *J* = 7.8 Hz, 1H, CH), 3.93–3.82 (m, 2H, CH_2), 2.15 (dt, *J* = 12.4, 6.3 Hz, 1H, CH_2), 1.98 (q, *J* = 8.3, 7.7 Hz, 2H, CH_2), 1.64 (d, *J* = 9.2 Hz, 1H, CH_2); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.6, 140.9, 138.9, 134.1, 132.8, 132.3, 129.7, 129.6, 129.6, 129.5, 129.5, 129.5, 127.1, 118.6, 118.6, 110.7, 95.5, 75.0, 68.2, 33.4, 26.3; **HRMS** (TOF ES+): m/z calcd for $C_{21}H_{19}Cl_2N_2O_2$: [$(M+H)^+$], 401.0818, found, 401.0820.

(*E*)-1-(3-Bromophenyl)-3-((3-bromophenyl)amino)-5-((tetrahydrofuran-2-yl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (8g).



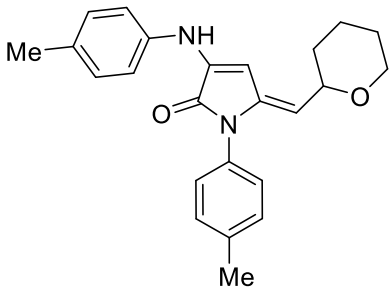
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 7:1, R_f = 0.2; Yellow solid: 37 mg (37%), mp = 156–157 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.56 (d, *J* = 7.9 Hz, 1H, ArH), 7.49 (s, 1H, ArH), 7.37 (t, *J* = 8.0 Hz, 1H, ArH), 7.31 (s, 1H, ArH), 7.21 (t, *J* = 8.0 Hz, 1H, ArH), 7.15 (d, *J* = 7.8 Hz, 1H, ArH), 7.07 (d, *J* = 7.9 Hz, 1H, ArH), 6.85 (s, 1H, NH), 6.53 (s, 1H, C=CH), 5.10 (d, *J* = 8.8 Hz, 1H, C=CH), 4.76 (q, *J* = 8.2 Hz, 1H, CH), 3.93 (q, *J* = 7.3 Hz, 1H, CH_2), 3.85 (q, *J* = 7.4 Hz, 1H, CH_2), 2.17 (dq, *J* = 12.4, 6.3 Hz, 1H, CH_2), 2.00 (q, *J* = 7.4 Hz, 2H, CH_2), 1.67–1.64 (m, 1H, CH_2); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.4, 141.7, 140.7, 135.1, 132.4, 131.5, 131.4, 130.8, 130.6, 127.1, 125.1, 123.2, 122.7, 120.0, 116.0, 111.1, 96.2, 75.0, 68.3, 33.4, 26.3; **HRMS** (TOF ES+): m/z calcd for $C_{21}H_{18}Br_2N_2O_2Na$: [$(M+Na)^+$], 510.9627, found, 510.9630.

(*E*)-4-Methyl-1-phenyl-3-(phenylamino)-5-((tetrahydrofuran-2-yl)methylene)-1,5-dihydro-2*H*-pyrrol-2-one (8h).



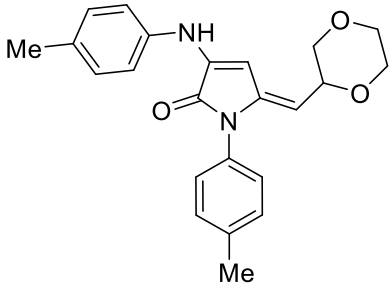
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 6:1, R_f = 0.2; Yellow solid: 42 mg (61%), *E/Z* = 1/1, mp = 189–190 °C; **Major: ^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.49 (d, *J* = 7.6 Hz, 3H, ArH), 7.42 (q, *J* = 7.5 Hz, 3H, ArH) (minor), 7.30 (d, *J* = 7.3 Hz, 4H, ArH), 7.03–6.98 (m, 2H, ArH) (minor), 6.95–6.93 (m, 3H, ArH), 6.06 (s, 1H, NH), 5.18 (d, *J* = 9.0 Hz, 1H, C=CH), 5.14 (d, *J* = 9.2 Hz, 1H, ArH) (minor), 5.01 (q, *J* = 7.9 Hz, 1H, CH), 3.90 (q, *J* = 7.0 Hz, 2H, CH_2), 3.85–3.78 (m, 2H, ArH) (minor), 3.50 (q, *J* = 7.4 Hz, 1H, CH_2) (minor), 2.20–2.17 (m, 1H, CH_2) (minor), 2.09 (s, 3H, CH_3) (minor), 1.99–1.91 (m, 2H, CH_2), 1.91 (s, 3H, CH_3), 1.88–1.82 (m, 1H, CH_2) (minor), 1.66–1.63 (m, 2H, CH_2), 1.50–1.45 (m 1H, CH_2) (minor); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 167.9 (minor), 165.8, 142.1 (minor), 141.4, 140.1 (minor), 136.9, 134.3 (minor), 132.3, 129.4, 129.4, 129.2 (minor), 128.4 (minor), 128.9, 128.9, 128.9, 128.9, 128.9, 128.1 (minor), 121.7, 118.9 (minor), 118.8, 118.8, 117.5 (minor), 114.1 (minor), 113.5, 110.9, 73.9 (minor), 73.6, 68.1 (minor), 67.8, 33.7 (minor), 33.0, 26.3 (minor), 26.1, 14.8 (minor), 11.5; **HRMS** (TOF ES+): m/z calcd for $C_{22}H_{23}N_2O_2$: $[(M+H)^+]$, 347.1754, found, 347.1758.

(*E*)-5-((Tetrahydro-*2H*-pyran-2-yl)methylene)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (8i).



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 7:1, R_f = 0.2; Yellow solid: 26 mg (36%), mp = 191–192 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.29–7.27 (m, 2H, ArH), 7.17 (dd, *J* = 8.2, 2.8 Hz, 4H, ArH), 7.07 (d, *J* = 8.3 Hz, 2H, ArH), 6.77 (s, 1H, NH), 6.38 (s, 1H, C=CH), 5.03 (d, *J* = 8.6 Hz, 1H, C=CH), 4.24 (td, *J* = 11.0, 8.5, 2.3 Hz, 1H, CH), 4.04–4.00 (m, 1H, CH_2), 3.56 (td, *J* = 11.2, 3.1 Hz, 1H, CH_2), 2.40 (s, 3H, $ArCH_3$), 2.34 (s, 3H $ArCH_3$), 1.86 (t, *J* = 9.3 Hz, 1H, CH_2), 1.70–1.68 (m, 1H, CH_2), 1.63 (d, *J* = 13.8 Hz, 1H, ArH), 1.60–1.53 (m, 3H, CH_2), 1.46–1.41 (m, 1H, CH_2); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 166.1, 141.4, 138.1, 133.6, 131.7, 131.3, 130.0, 130.0, 129.9, 129.9, 128.2, 128.2, 128.2, 117.6, 117.6, 110.2, 94.0, 74.3, 68.4, 33.0, 25.6, 23.4, 21.3, 20.8; **HRMS** (TOF ES+): m/z calcd for $C_{24}H_{28}N_2O_2$: $[(M+H)^+]$, 375.2067, found, 375.2073.

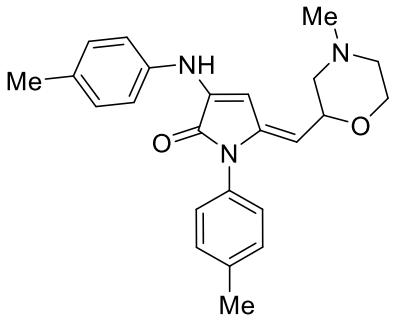
(*E*)-5-((1,4-dioxan-2-yl)methylene)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (8j).



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 30 mg (40%), mp = 181–182 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.28 (d, *J* = 8.2 Hz, 2H, ArH), 7.18 (dd, *J* = 14.1, 7.9 Hz, 4H, ArH), 7.07

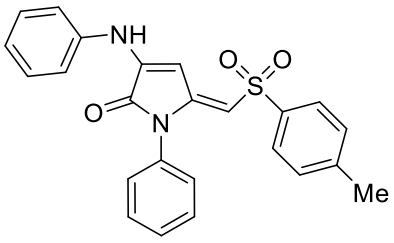
(d, *J* = 7.9 Hz, 2H, ArH), 6.78 (s, 1H, NH), 6.36 (s, 1H, C=CH), 4.86 (d, *J* = 8.8 Hz, 1H, C=CH), 4.51 (td, *J* = 9.4, 2.8 Hz, 1H, CH), 3.87–3.81 (m, 2H, CH_2), 3.72 (td, *J* = 9.6, 8.7, 5.2 Hz, 2H, CH_2), 3.61 (td, *J* = 11.8, 9.9, 4.0 Hz, 1H, CH_2), 3.38–3.34 (m, 1H, CH_2), 2.40 (s, 3H, $ArCH_3$), 2.35 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.9, 143.5, 138.3, 137.7, 133.9, 132.0, 130.9, 130.0, 130.0, 130.0, 130.0, 128.1, 128.1, 117.7, 117.7, 103.7, 93.4, 72.5, 70.9, 66.5, 66.1, 21.2, 20.8; **HRMS** (TOF ES+): m/z calcd for $C_{23}H_{25}N_2O_3$: $[(M+H)^+]$, 377.1860, found, 377.1863.

(*E*)-5-((4-methylmorpholin-2-yl)methylene)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (8k).



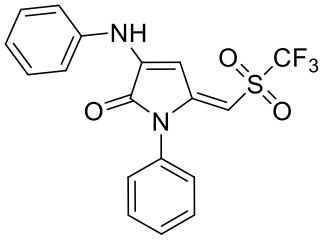
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 2:1, R_f = 0.2; Yellow solid: 30 mg (41%), mp = 211–213 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.27 (s, 2H, ArH), 7.19 (d, *J* = 7.3 Hz, 2H, ArH), 7.15 (d, *J* = 7.6 Hz, 2H, ArH), 7.06 (d, *J* = 7.4 Hz, 2H, ArH), 6.77 (s, 1H, NH), 6.37 (s, 1H, C=CH), 4.80 (d, *J* = 9.6 Hz, 1H, C=CH), 3.86 (d, *J* = 11.5 Hz, 1H, CH), 3.72 (s, 1H, CH), 3.63 (t, *J* = 11.4 Hz, 1H, CH), 3.26 – 3.23 (m, 1H, CH), 3.05 (t, *J* = 10.0 Hz, 1H, CH), 2.76 (d, *J* = 11.9 Hz, 1H, CH), 2.48 (s, 1H, CH), 2.40 (s, 3H, $ArCH_3$), 2.35 (s, 3H, $ArCH_3$), 2.25 (s, 3H, CH_3); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.4, 142.3, 137.7, 137.6 (minor), 137.5 (minor), 137.4, 133.2, 131.4, 130.6, 129.6, 129.5, 127.6 (minor),127.5, 127.5, 117.1, 117.1, 117.0 (minor), 106.2, 93.0, 70.6, 66.7, 66.5 (minor), 60.8, 55.1, 54.4, 53.2, 43.5, 20.7, 20.3; **HRMS** (TOF ES+): m/z calcd for $C_{24}H_{28}N_3O_3$: $[(M+H)^+]$, 390.2176, found, 390.2179.

(*E*)-1-Phenyl-3-(phenylamino)-5-(((trifluoromethyl)sulfonyl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (9).



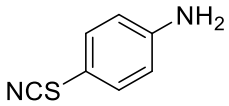
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 8:1, R_f = 0.2; Yellow solid: 39 mg (37%), mp = 234–235 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 9.57 (s, 1H, NH), 7.77 (d, *J* = 8.3 Hz, 2H, ArH), 7.54 (t, *J* = 7.5 Hz, 2H, ArH), 7.49 (d, *J* = 7.0 Hz, 1H, ArH), 7.46 (d, *J* = 7.6 Hz, 2H, ArH), 7.41 (t, *J* = 7.5 Hz, 4H, ArH), 7.32 (d, *J* = 7.2 Hz, 2H, ArH), 7.12 (t, *J* = 7.2 Hz, 1H, ArH), 7.00 (s, 1H, C=CH), 5.48 (s, 1H, C=CH), 2.37 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.4, 152.0, 144.2, 140.5, 140.3, 137.8, 133.1, 130.5, 130.5, 130.2, 130.2, 129.9, 129.9, 129.5, 129.0, 129.0, 127.0, 127.0, 123.8, 119.6, 119.6, 104.2, 91.7, 21.5; **HRMS** (TOF ES+): m/z calcd for $C_{24}H_{21}N_2O_3S$: $[(M+H)^+]$, 417.1267, found, 417.1270.

(*E*)-1-Phenyl-3-(phenylamino)-5-(((trifluoromethyl)sulfonyl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (10).



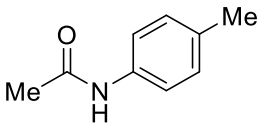
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 8:1, R_f = 0.2; Yellow solid: 59 mg (15%), mp = 225–226 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 10.19 (s, 1H, NH), 7.60 (t, J = 7.5 Hz, 2H, ArH), 7.55 (t, J = 7.3 Hz, 1H, ArH), 7.49 (d, J = 7.8 Hz, 1H, ArH), 7.46 (d, J = 6.8 Hz, 2H, ArH), 7.40 (d, J = 8.0 Hz, 2H, ArH), 7.19 (t, J = 7.4 Hz, 1H, ArH), 6.64 (s, 1H, C=CH), 5.16 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.4, 162.7, 140.6, 139.6, 132.5, 130.4, 130.4, 130.1, 130.1, 130.0, 129.1, 129.1, 125.0, 120.5, 120.5, 120.3 (d, J = 325.7 Hz), 89.5, 87.5; **HRMS** (TOF ES+): m/z calcd for $C_{18}H_{14}F_3N_2O_3S$: [$(M+H)^+$], 395.0672, found, 395.0672.

4-Thiocyanatoaniline (11)



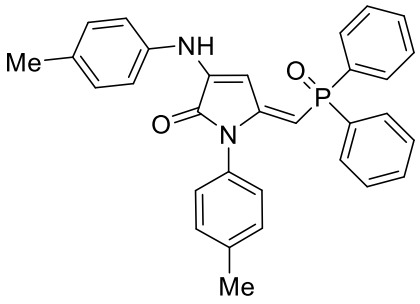
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 3:1, R_f = 0.2; Yellow oli: 40 mg (67%); **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.36 (d, J = 8.5 Hz, 2H, ArH), 6.68 (d, J = 8.5 Hz, 2H, ArH), 3.94 (s, 2H, NH_2); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 148.8, 134.6, 134.6, 116.1, 116.1, 112.4, 109.6; **HRMS** (TOF ES+): m/z calcd for $C_7H_7N_2$: [$(M+H)^+$], 151.0324, found, 151.0330.

***N*-(*p*-Tolyl)acetamide (12)**



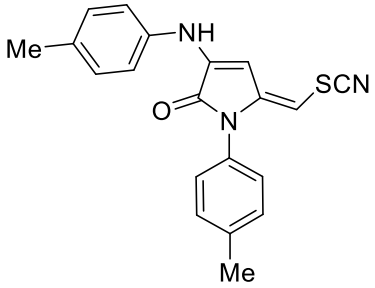
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 5:1, R_f = 0.2; White solid: 39 mg (60%), mp = 135–136 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 9.85 (s, 1H, NH), 7.45 (d, J = 8.5 Hz, 2H, ArH), 7.08 (d, J = 8.2 Hz, 2H, ArH), 2.24 (s, 3H, $ArCH_3$), 2.01 (s, 3H, CH_3); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 168.5, 137.3, 132.3, 129.5, 129.5, 119.4, 119.4, 24.4, 20.9; **HRMS** (TOF ES+): m/z calcd for $C_9H_{12}NO$: [$(M+H)^+$], 150.0913, found, 150.0908.

(*E*)-5-((Diphenylphosphoryl)methylene)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (13).



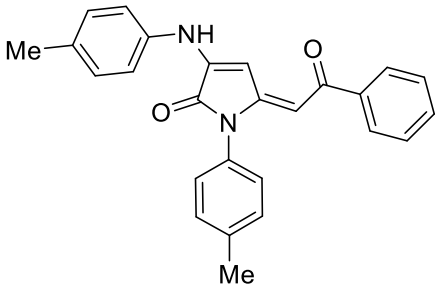
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 2:1, R_f = 0.2; Yellow solid: 70 mg (76%), mp = 117–118 °C; **^{1}H NMR** (600 MHz, Chloroform-*d*) δ = 7.69 (dd, *J* = 12.3, 7.7 Hz, 4H, ArH), 7.51 (t, *J* = 7.5 Hz, 2H, ArH), 7.46 (d, *J* = 7.1 Hz, 4H, ArH), 7.30 (d, *J* = 7.9 Hz, 2H, ArH), 7.19 (d, *J* = 8.4 Hz, 2H, ArH), 7.11 (d, *J* = 7.9 Hz, 2H, ArH), 6.98 (d, *J* = 8.5 Hz, 2H, ArH), 6.92 (s, 1H, NH), 6.87 (s, 1H, C=CH), 5.34 (d, *J* = 15.7 Hz, 1H, C=CH), 2.41 (s, 3H, $ArCH_3$), 2.31 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$) δ = 166.1, 154.7 (d, *J* = 10.9 Hz), 138.8, 136.9, 134.9 (d, *J* = 15.0 Hz), 134.2, 132.5, 131.7 (d, *J* = 2.8 Hz), 131.7 (d, *J* = 2.8 Hz), 131.7, 131.1 (d, *J* = 10.0 Hz), 131.1 (d, *J* = 10.0 Hz), 130.4 (d, *J* = 14.3 Hz), 130.3, 130.1, 130.1, 130.1, 128.7, 128.6, 128.1, 128.1, 117.8, 117.8, 95.4, 95.1 (d, *J* = 5.4 Hz), 95.1 (d, *J* = 111.8 Hz), 94.7, 21.3, 20.8; **HRMS** (TOF ES+): m/z calcd for $C_{29}H_{24}N_2O_2P$: [$(M+H)^+$], 491.1883, found, 491.1891.

(*E*)-5-(Thiocyanatomethylene)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (14).



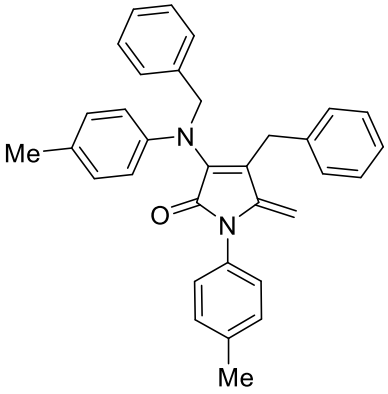
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 46 mg (67%), mp = 203–204 °C; **^{1}H NMR** (600 MHz, Chloroform-*d*) δ = 77.31 (d, *J* = 7.9 Hz, 2H, ArH), 7.21 (d, *J* = 8.1 Hz, 2H, ArH), 7.16 (d, *J* = 7.9 Hz, 2H, ArH), 7.10 (d, *J* = 8.1 Hz, 2H, ArH), 7.03 (s, 1H, NH), 6.48 (s, 1H, C=CH), 5.33 (s, 1H, C=CH), 2.43 (s, 3H, $ArCH_3$), 2.36 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$) δ = 166.2, 151.8, 139.1, 136.8, 135.5, 133.2, 133.2, 130.3, 130.3, 130.2, 129.9, 127.7, 127.7, 118.3, 118.3, 111.2, 92.0, 84.4, 21.2, 20.9; **HRMS** (TOF ES+): m/z calcd for $C_{20}H_{18}N_3OS$: [$(M+H)^+$], 348.1165, found, 348.1170.

(*E*)-5-(2-Oxo-2-phenylethylidene)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (15).



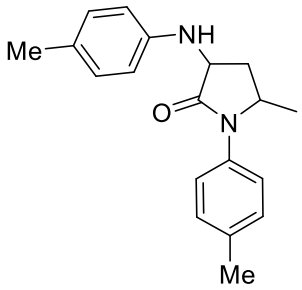
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 46 mg (21%), E/Z = 10/1, mp = 125–126 ^oC; **Major:^{1}H NMR** (600 MHz, $CDCl_3$) δ = 77.87 (d, *J* = 7.3 Hz, 2H, ArH), 7.80 (d, *J* = 7.5 Hz, 2H, ArH), 7.76 (s, 1H, NH), 7.50 (d, *J* = 7.3 Hz, 2H, ArH), 7.48 (s, 1H, ArH, minor), 7.42 (t, *J* = 7.4 Hz, 2H, ArH), 7.36 (d, 1H, *J* = 7.3 Hz, ArH), 7.20 (d, 2H, *J* = 7.1 Hz, ArH, minor), 7.19–7.18 (m, 2H, ArH), 7.04 (s, 1H, C=CH), 6.35 (s, 1H, C=CH), 2.46 (s, 3H, $ArCH_3$), 2.36 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$) δ = 190.1, 166.7, 165.6 (minor), 154.9, 139.6, 138.9, 136.8, 136.7, 135.3, 135.1 (minor), 134.3, 133.2, 132.3, 131.8, 130.5 (minor), 130.3, 130.2, 129.6, 128.8, 128.5, 128.0, 127.8, 126.9, 120.2, 118.2, 100.4, 95.5, 21.3, 20.9; **HRMS** (TOF ES+): m/z calcd for $C_{26}H_{23}N_2O$: $[(M+H)^+]$, 395.1754, found, 395.1760.

4-Benzyl-3-(benzyl(*p*-tolyl)amino)-5-methylene-1-(*p*-tolyl)-1,5-dihydro-*2H*-pyrrol-2-one (16).



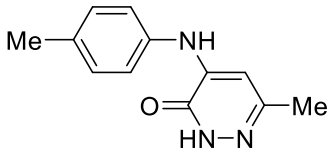
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 15:1, R_f = 0.2; Yellow oli: 47 mg (51%); **^{1}H NMR** (600 MHz, $CDCl_3$) δ = 7.25 (d, *J* = 6.6 Hz, 5H, ArH), 7.20 (d, *J* = 8.1 Hz, 3H, ArH), 7.18 (d, *J* = 6.4 Hz, 4H), 6.96 (dd, *J* = 11.4, 8.0 Hz, 4H, ArH), 6.77 (d, *J* = 8.3 Hz, 2H, ArH), 5.08 (s, 2H, CH_2), 4.71 (d, *J* = 3.6 Hz, 2H, C=CH), 3.63 (s, 2H, CH_2), 2.38 (s, 3H, $ArCH_3$), 2.24 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$) δ = 166.4, 145.3, 144.0, 139.1, 138.6, 137.5, 136.0, 131.7, 130.8, 129.9, 129.8, 129.7, 129.7, 129.5, 128.8, 128.4, 128.4, 128.4, 128.4, 127.9, 127.9, 127.7, 127.7, 126.9, 126.6, 126.2, 119.1, 117.9, 94.0, 54.3, 30.8, 21.27, 20.6; **HRMS** (TOF ES+): m/z calcd for $C_{33}H_{31}N_2O$: $[(M+H)^+]$, 471.2431, found, 471.2437.

5-Methyl-1-(*p*-tolyl)-3-(*p*-tolylamino)pyrrolidin-2-one (17).



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 8:1, R_f = 0.2; Yellow solid: 40 mg (68%) mp = 147–148 °C; **^{1}H NMR** (600 MHz, $CDCl_3$) δ = 7.21 (d, *J* = 4.8 Hz, 4H, ArH), 7.03 (d, *J* = 8.1 Hz, 2H, ArH), 6.64 (d, *J* = 8.2 Hz, 2H, ArH), 4.55 (s, 1H, NH), 4.23 – 4.20 (m, 1H, CH), 4.06 – 4.03 (m, 1H, CH), 3.05 – 3.01 (m, 1H, CH), 2.36 (s, 3H, $ArCH_3$), 2.26 (s, 3H, $ArCH_3$), 1.63 (q, *J* = 11.9 Hz, 1H, CH), 1.21 (s, 3H, CH_3); **^{13}C NMR** (150 MHz, $CDCl_3$) δ = 172.7, 145.3, 136.4, 134.1, 129.8, 129.8, 129.7, 129.7, 127.7, 124.7, 124.7, 113.9, 113.9, 55.8, 52.2, 38.6, 21.1, 20.5, 20.4; **HRMS** (TOF ES+): m/z calcd for $C_{19}H_{23}N_2O$: $[(M+H)^+]$, 295.1805, found, 295.1815.

6-Methyl-4-(*p*-tolylamino)pyridazin-3(*2H*)-one (18).



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 3:1, R_f = 0.2; White solid: 40 mg (78%) mp = 234–235 °C; **^{1}H NMR** ^{1}H NMR (600 MHz, CD_3OD) δ 7.57 (d, *J* = 8.2 Hz, 2H, ArH), 7.17 (d, *J* = 8.1 Hz, 2H, ArH), 6.58 (s, 1H, C=CH), 2.37 (s, 3H, $ArCH_3$), 2.33 (s, 3H, CH_3); **^{13}C NMR** (150 MHz, CD_3OD) δ = 161.6, 146.9, 140.8, 135.4, 133.7, 128.9, 128.9, 120.2, 120.2, 104.2, 19.53, 9.1; **HRMS** (TOF ES+): m/z calcd for $C_{12}H_{14}N_3O$: $[(M+H)^+]$, 216.1131, found, 216.1140.

5. (1) X-ray Structure and Data[3] of 4b (CCDC 2226645) .

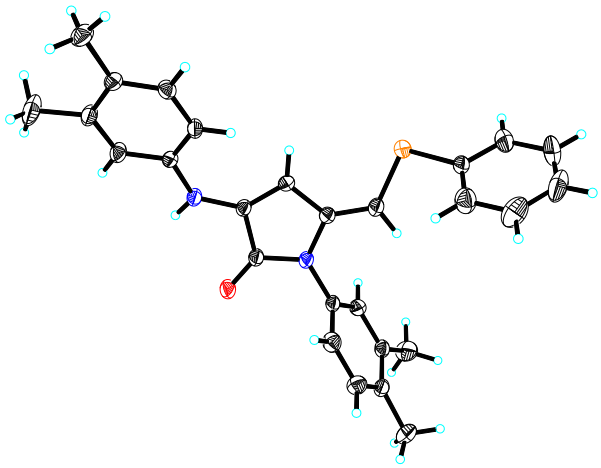


Figure S1 X-Ray crystal structure of **4b**.

Table S4 Crystal data and structure refinement for **4b**.

Empirical formula	$C_{27}H_{26}N_2OS$
Formula weight	426.56
Temperature	296 K
Wavelength	0.71073 Å
Crystal system, space group	monoclinic, P 2_1/c
Unit cell dimensions	a = 11.2145(17) A alpha = 90 deg.
	b = 22.855(4) A beta = 105.549(3) deg.
	c = 9.3203(14) A gamma = 90 deg.
Volume	2301.4(6) A^3
Z, Calculated density	4, 1.231 Mg/m^3
Absorption coefficient	0.162 mm^-1
F(000)	904
Theta range for data collection	5.188 to 55.192 deg.
Limiting indices	-14<=h<=14, -29<=k<=28, -12<=l<=9
Completeness to theta = 25.242	99.8%
Absorption correction	Multi-scan
Refinement method	SHELXL-2018/3 (Sheldrick 2015)
Data/restraints/parameters	5210 / 0 / 284
Goodness-of-fit on F^2	1.043
Final R indices [I>2sigma(I)]	R1 = 0.0552, wR2 = 0.1101
R indices (all data)	R1 = 0.1013, wR2 = 0.1301
Largest diff. peak and hole	0.27 and -0.35 e.A^-3

(2) X-ray Structure and Data[3] of 7b (CCDC 2226644) .

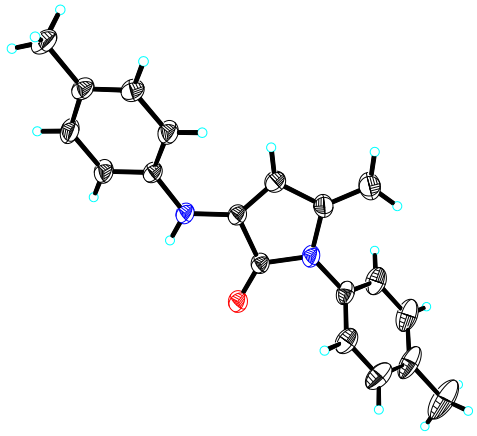


Figure S2 X-Ray crystal structure of **7b**.

Table S5 Crystal data and structure refinement for **7b**.

Empirical formula	$C_{19}H_{18}N_2O$
Formula weight	290.35
Temperature	296 K
Wavelength	0.71073 Å
Crystal system, space group	monoclinic, P 2_1/c
Unit cell dimensions	a = 11.578(6) A alpha = 90 deg.
	b = 10.373(5) A beta = 92.346(9) deg.
	c = 13.434(6) A gamma = 90 deg.
Volume	1612.0(13) A^3
Z, Calculated density	4, 1.196 Mg/m^3
Absorption coefficient	0.075 mm^-1
F(000)	616.0
Theta range for data collection	5.274 to 55.24 deg.
Limiting indices	-14<=h<=14, -9<=k<=13, -14<=l<=17
Completeness to theta = 25.242	99.8%
Absorption correction	Multi-scan
Refinement method	SHELXL-2018/3 (Sheldrick 2015)
Data/restraints/parameters	3638 / 0 / 201
Goodness-of-fit on F^2	0.972
Final R indices [I>2sigma(I)]	R1 = 0.0807, wR2 = 0.1832
R indices (all data)	R1 = 0.1652, wR2 = 0.2296
Largest diff. peak and hole	0.26 and -0.29 e.A^-3

(3) X-ray Structure and Data[3] of 8b (CCDC 2226646) .

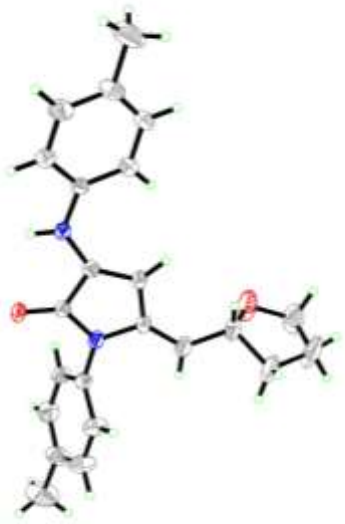


Figure S3 X-Ray crystal structure of **8b**.

Table S6 Crystal data and structure refinement for **8b**.

Empirical formula	$C_{23}H_{24}N_2O$
Formula weight	360.44
Temperature	296.15 K
Wavelength	0.71073 Å
Crystal system, space group	monoclinic, $P2_1/c$
Unit cell dimensions	a = 32.9740(7) A alpha = 90 deg.
	b = 9.2490(18) A beta = 91.554(4) deg.
	c = 15.640(3) A gamma = 90 deg.
Volume	4768.0(16) A^3
Z, Calculated density	8, 1.004 Mg/m^3
Absorption coefficient	0.064 mm^-1
F(000)	1536.0
Theta range for data collection	5.282 to 55.134 deg.
Limiting indices	-38<=h<=40, -11<=k<=11, -12<=l<=20
Completeness to theta = 25.242	99.8%
Absorption correction	Multi-scan
Refinement method	SHELXL-2018/3 (Sheldrick 2015)
Data/restraints/parameters	5358 / 0 / 246
Goodness-of-fit on F^2	1.030
Final R indices [I>2sigma(I)]	R1 = 0.0876, wR2 = 0.2455
R indices (all data)	R1 = 0.1619, wR2 = 0.2864
Largest diff. peak and hole	0.27 and -0.21 e.A^-3

6. ^{1}H NMR, ^{13}C NMR and HRMS spectra for spectroscopic data.

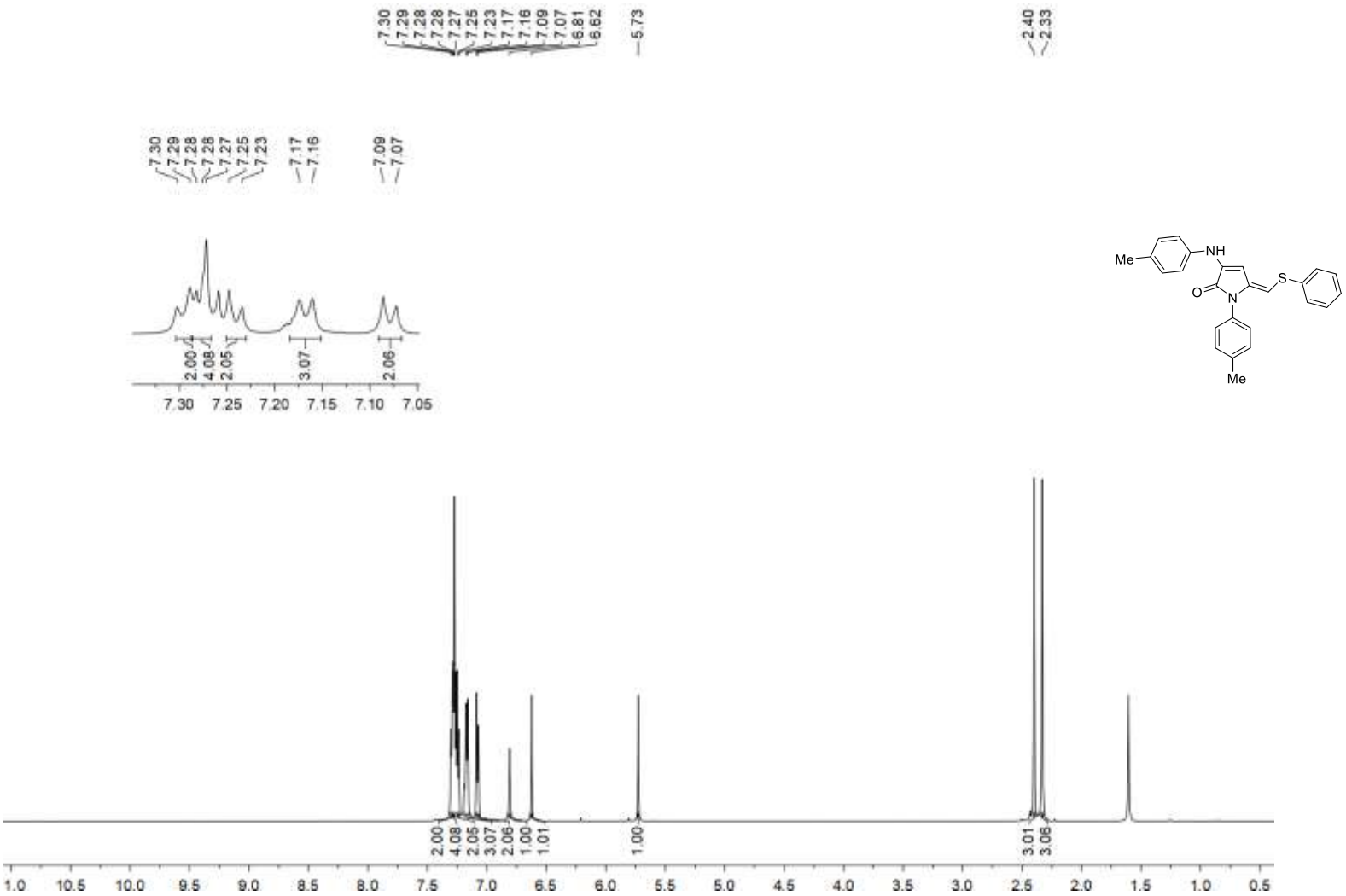


Figure S4. **^{1}H NMR** (600 MHz, $CDCl_3$) spectra of compound **4a**

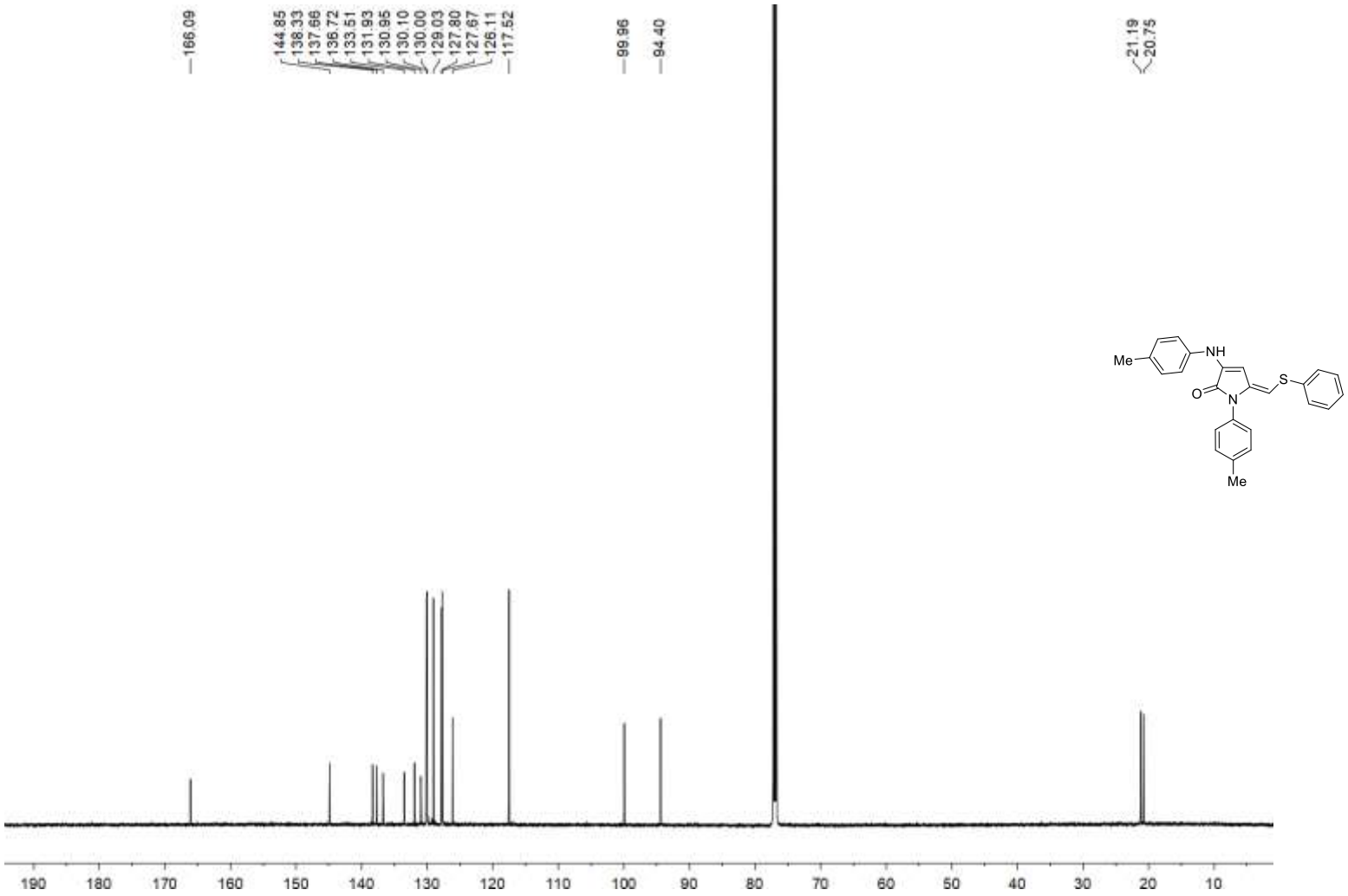


Figure S5. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4a**

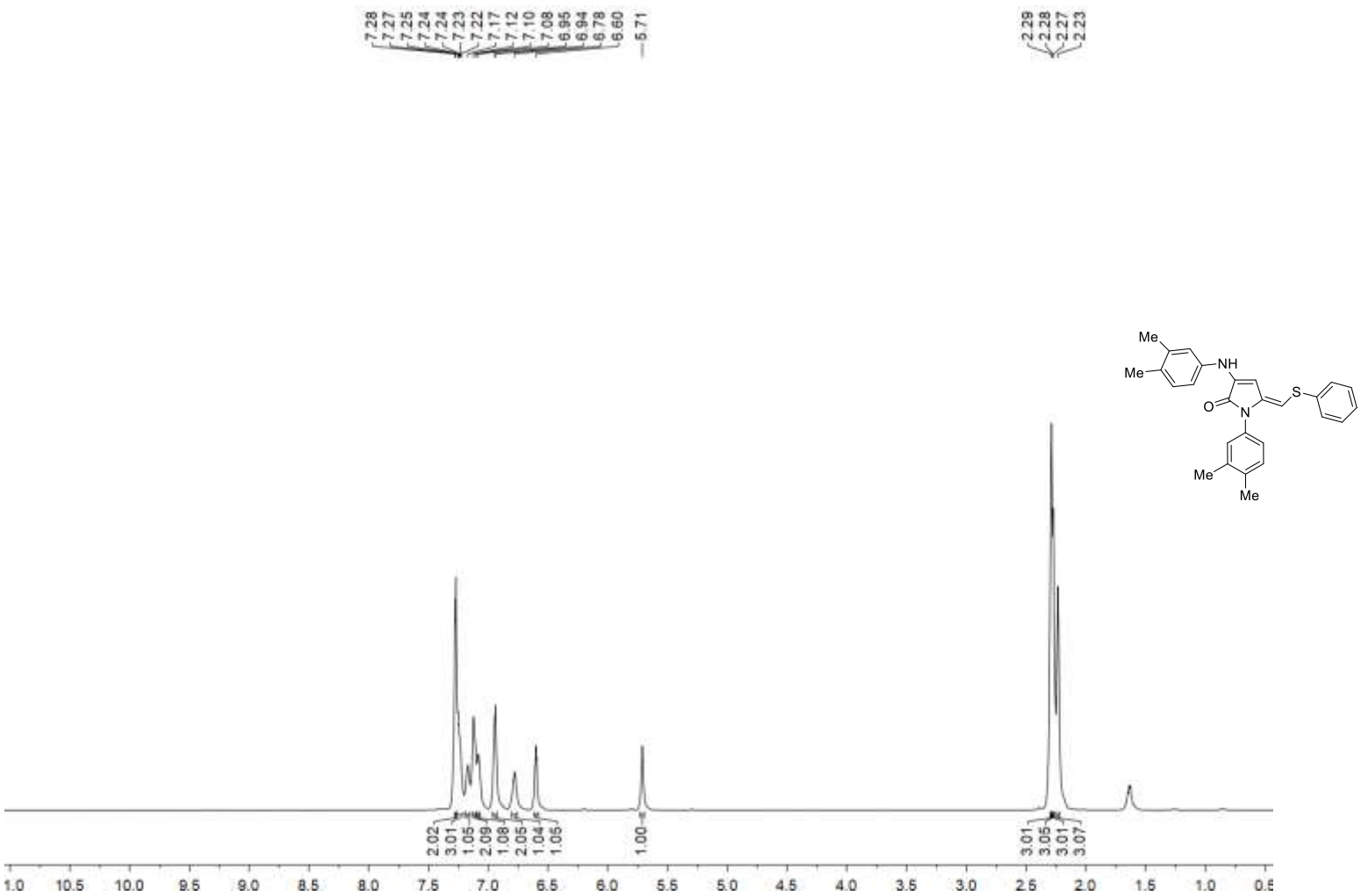


Figure S6. **^{1}H NMR** (600 MHz, $CDCl_3$) spectra of compound **4b**

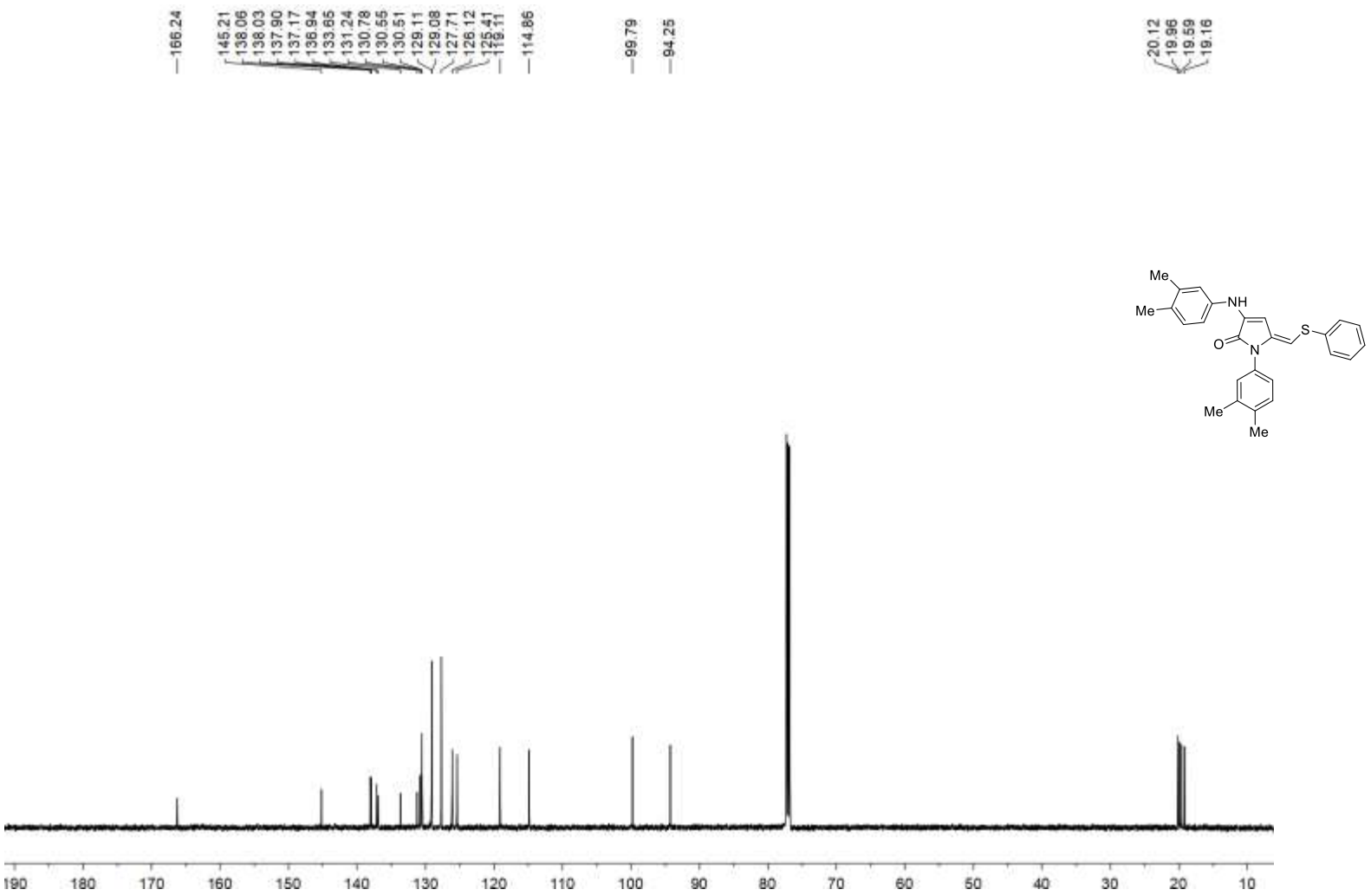


Figure S7. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4b**

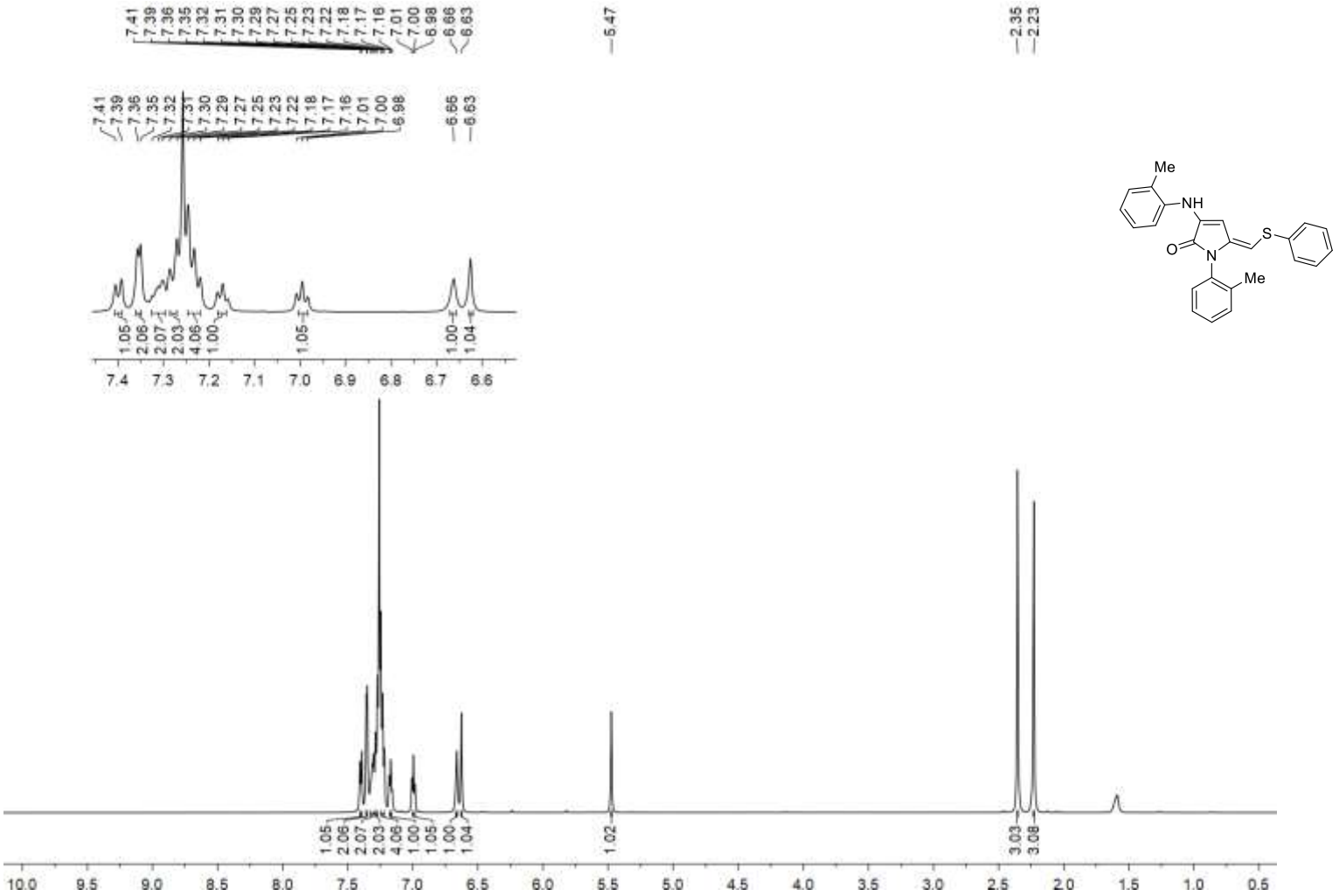


Figure S8. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4c**

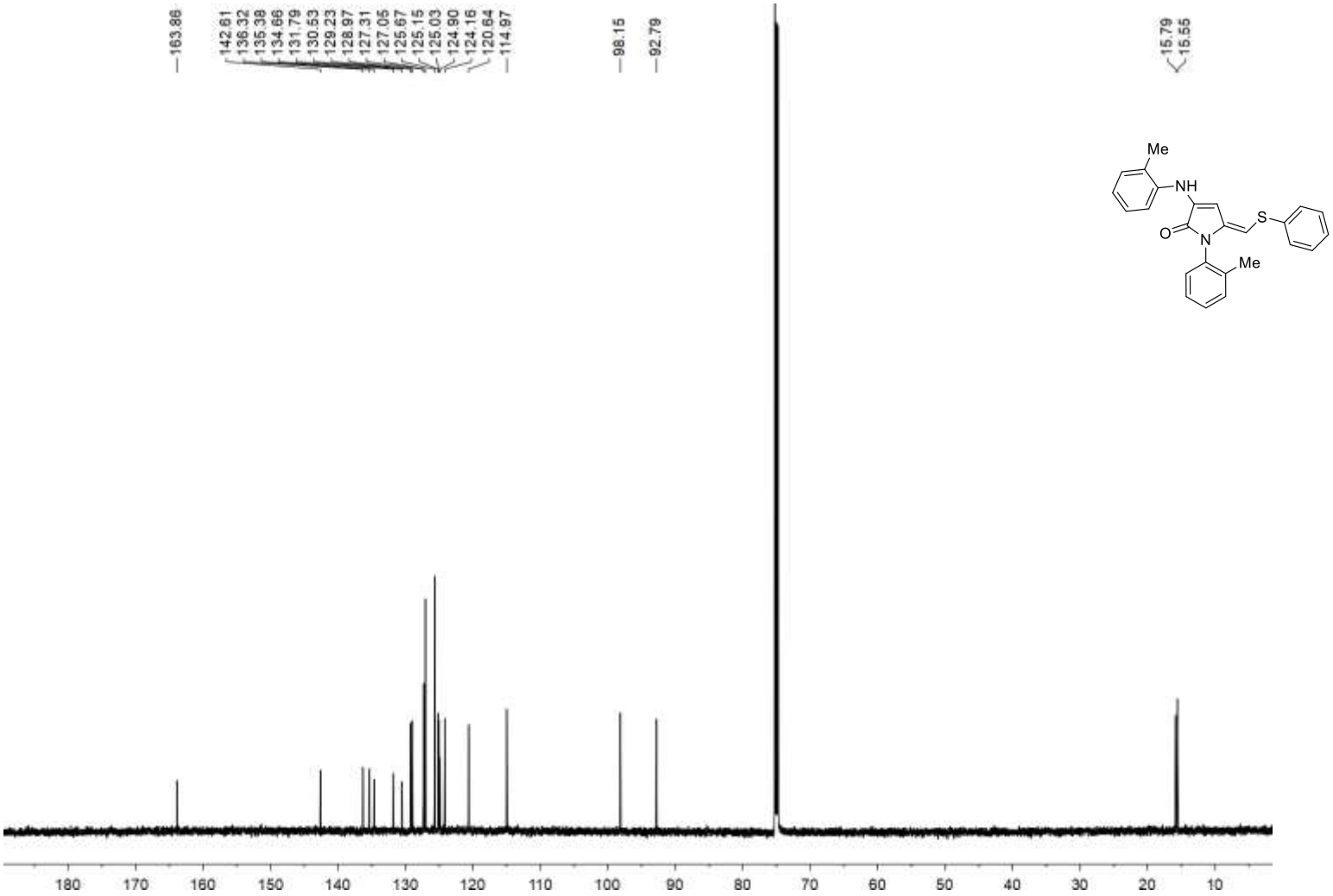


Figure S9. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4c**

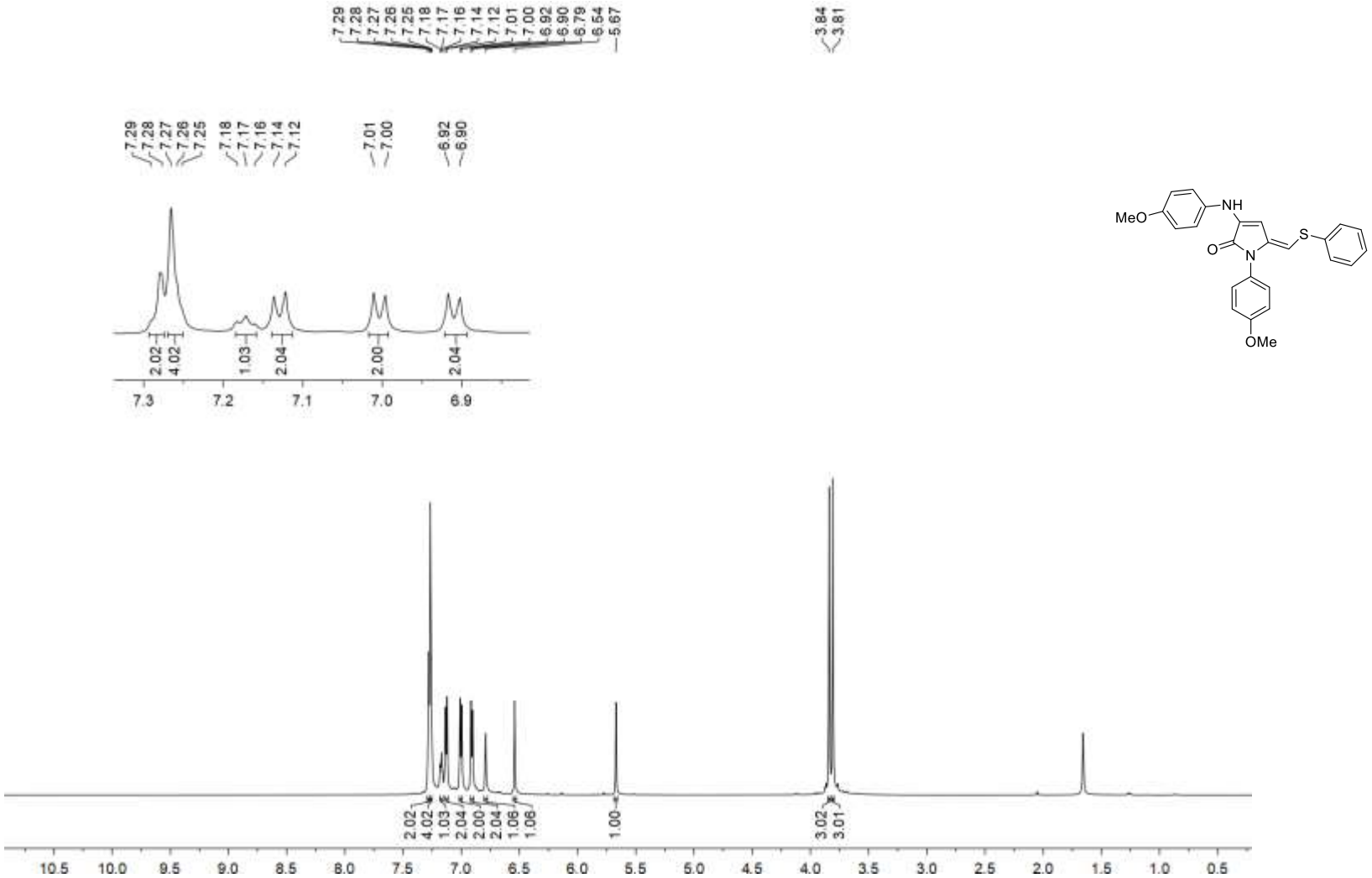


Figure S10. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4d**

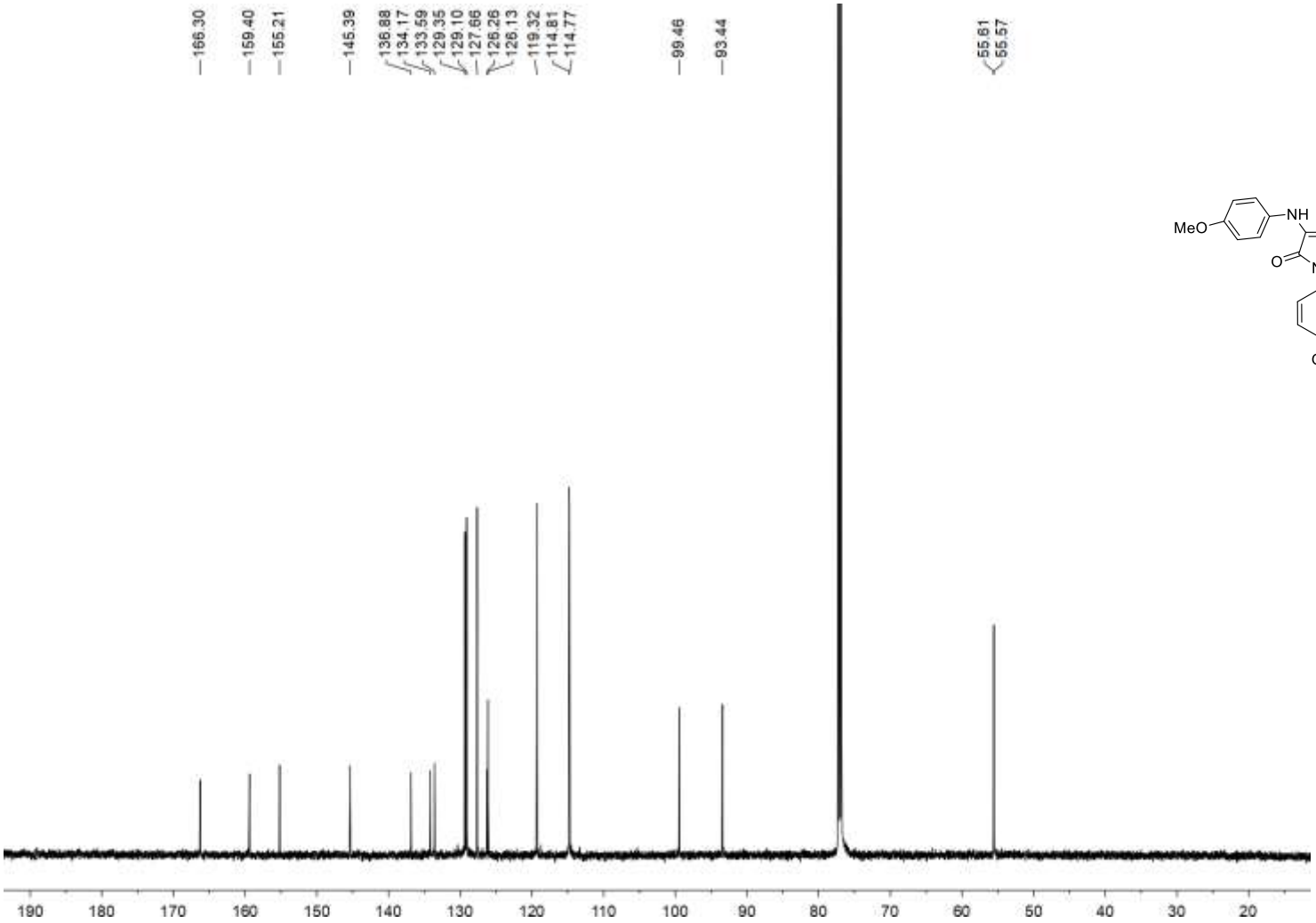


Figure S11. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4d**

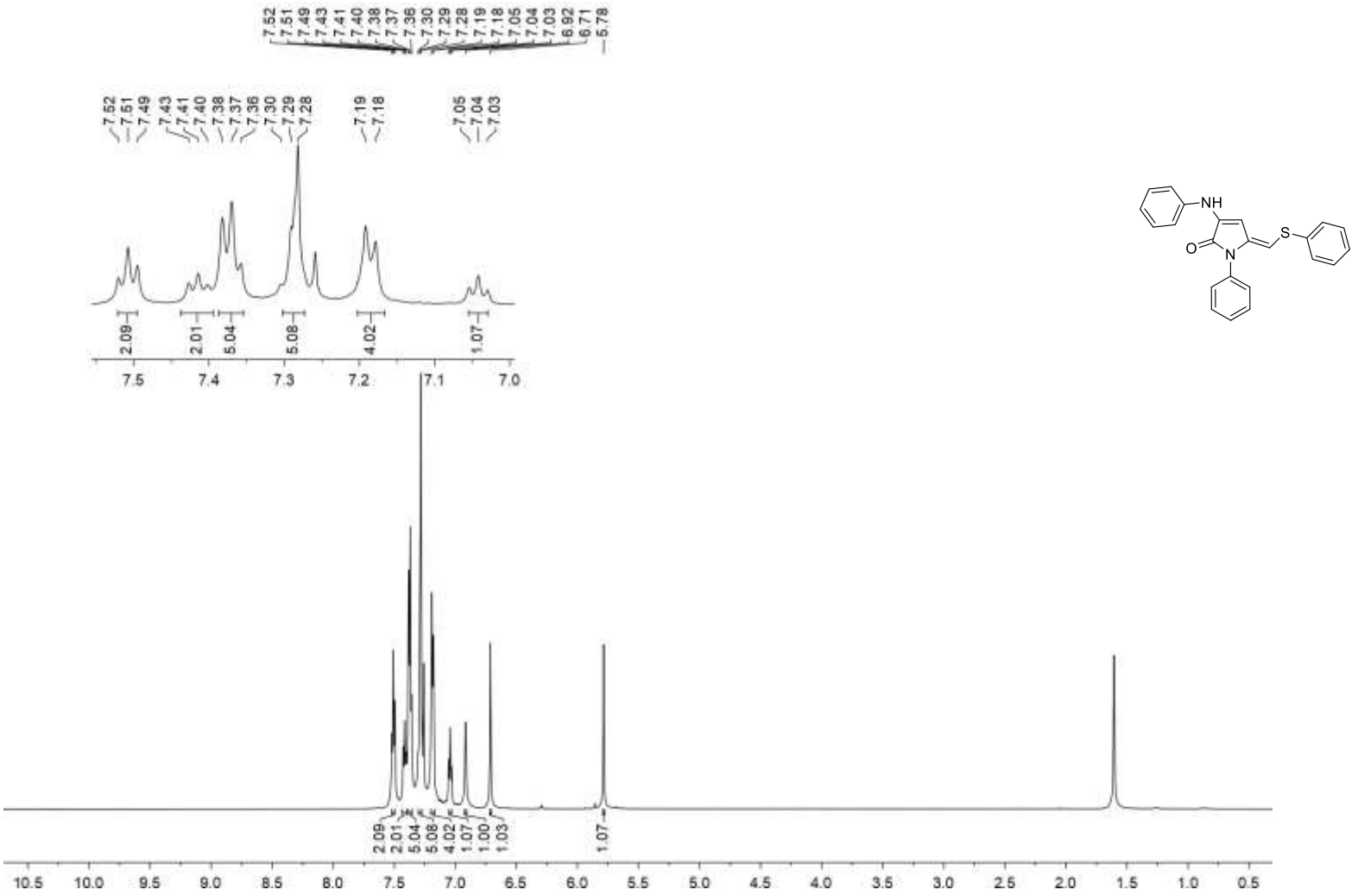


Figure S12. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4e**

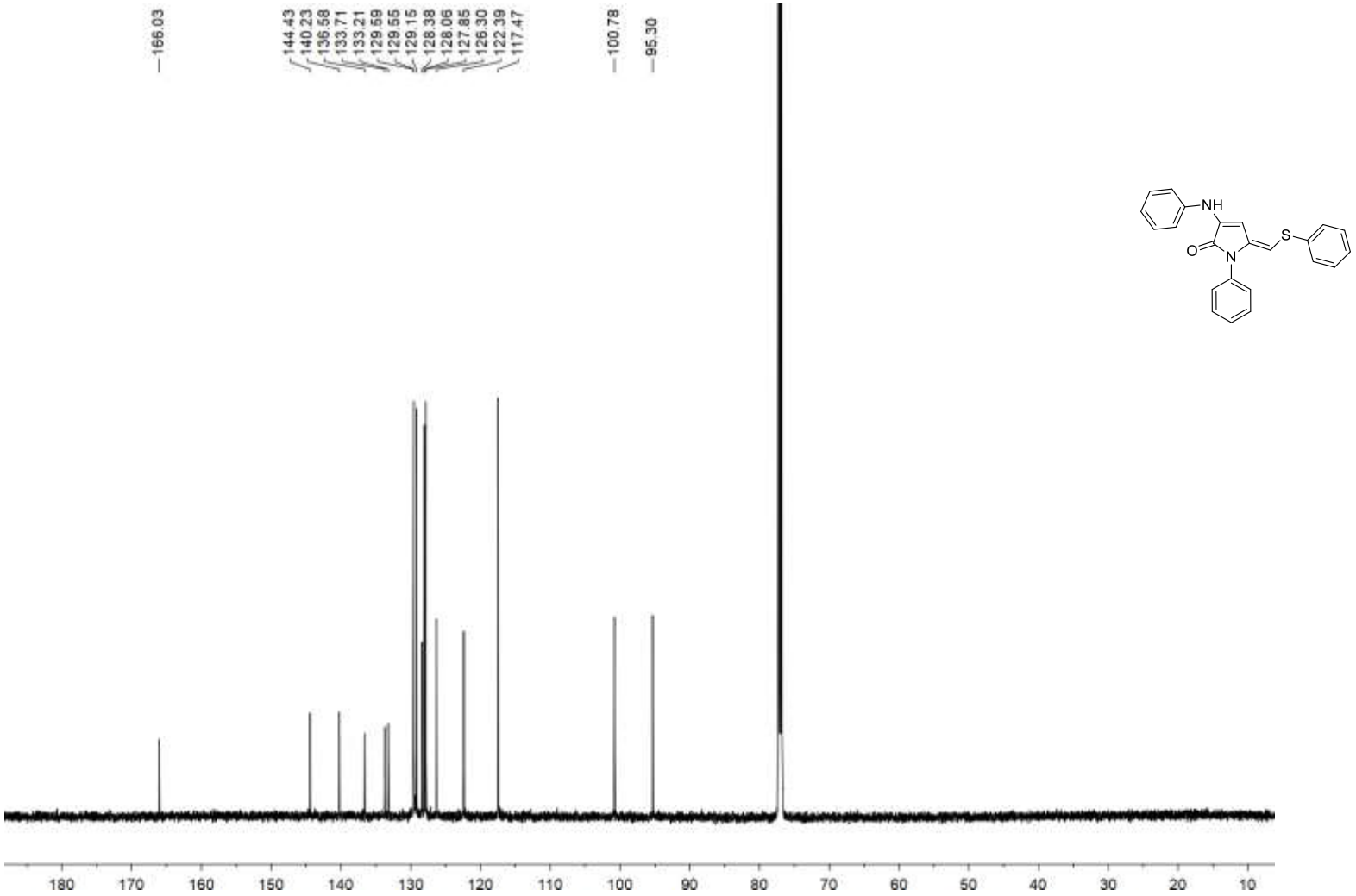


Figure S13. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4e**

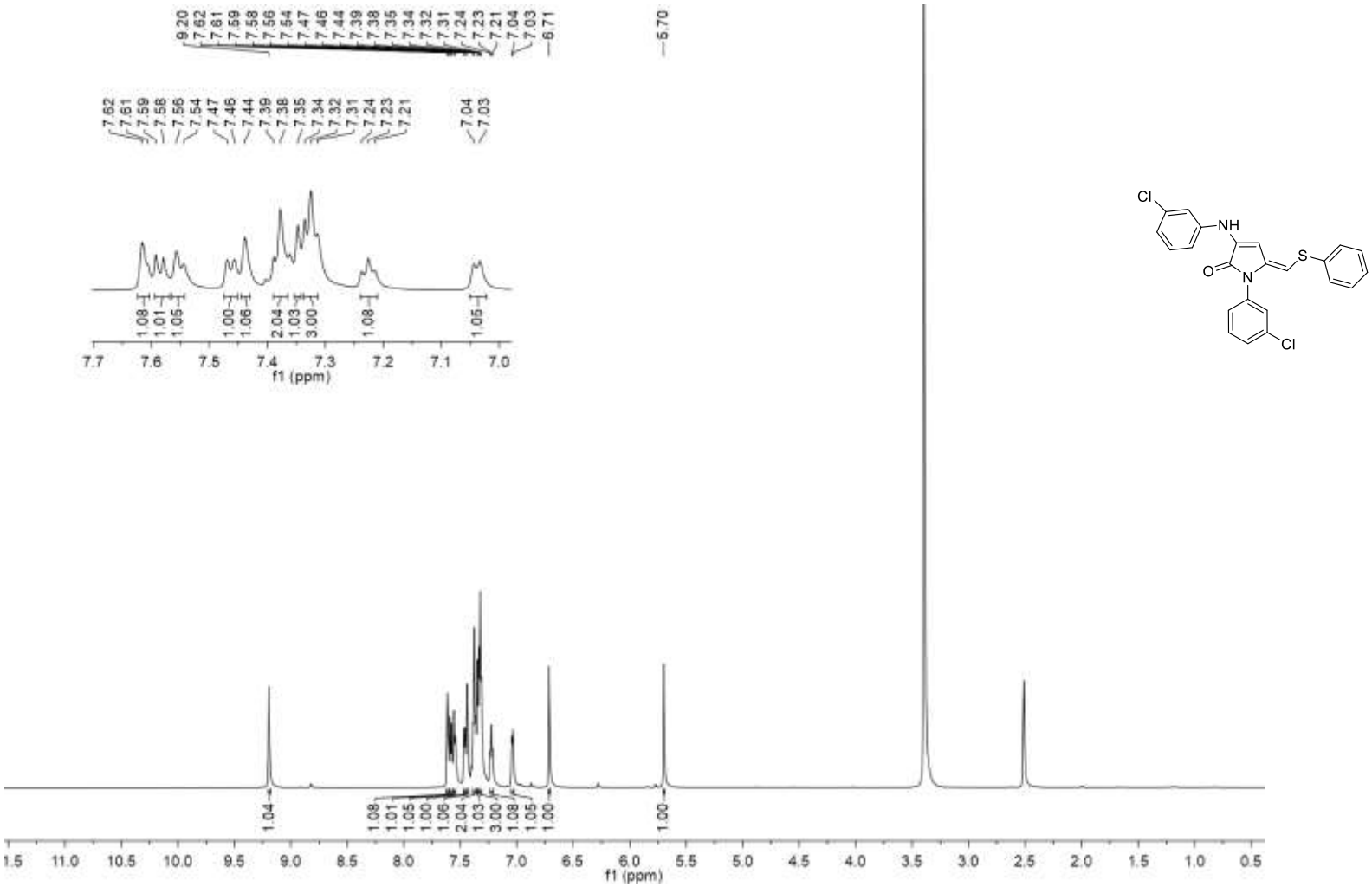


Figure S14. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **4f**

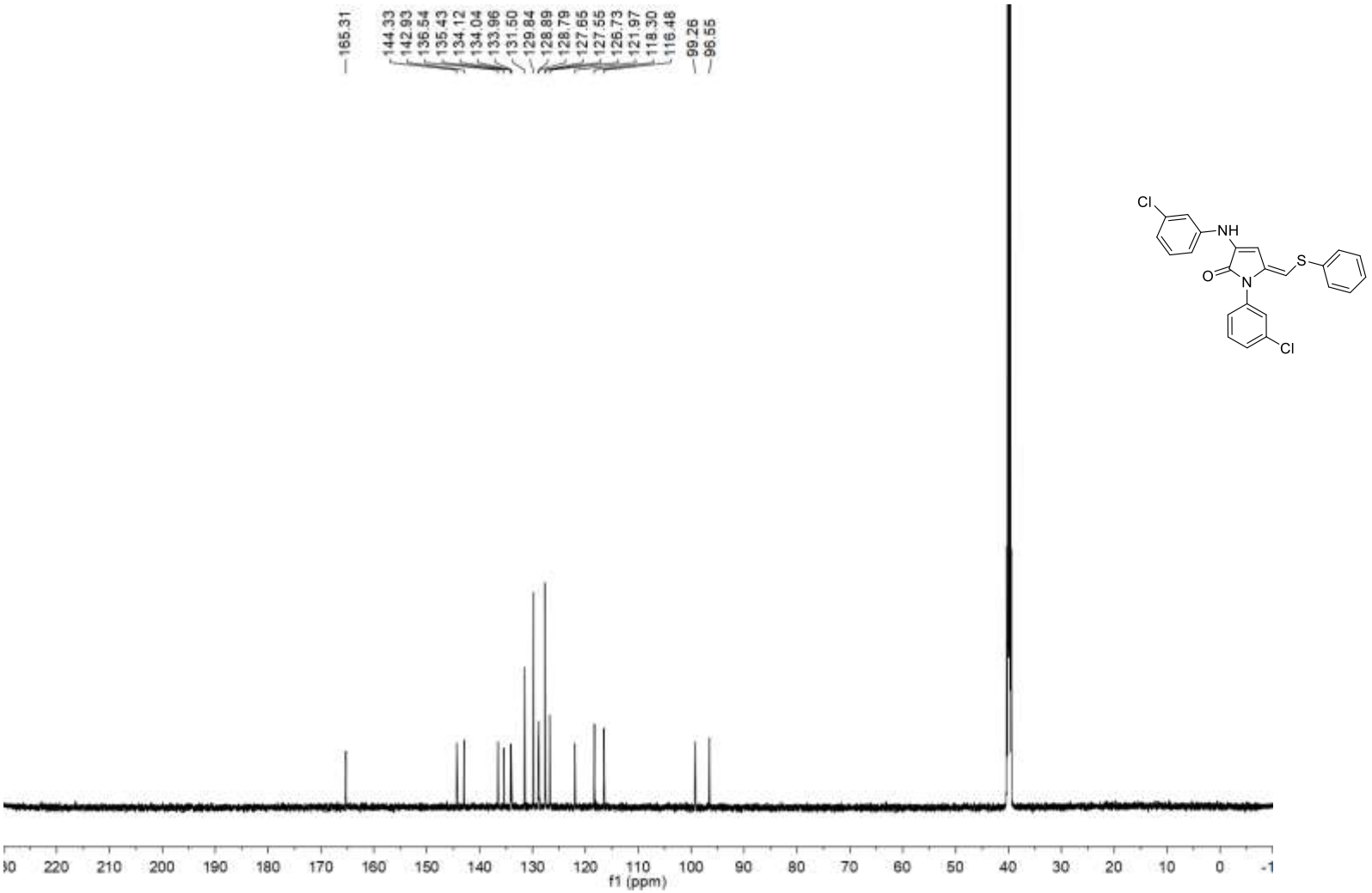


Figure S15. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **4f**

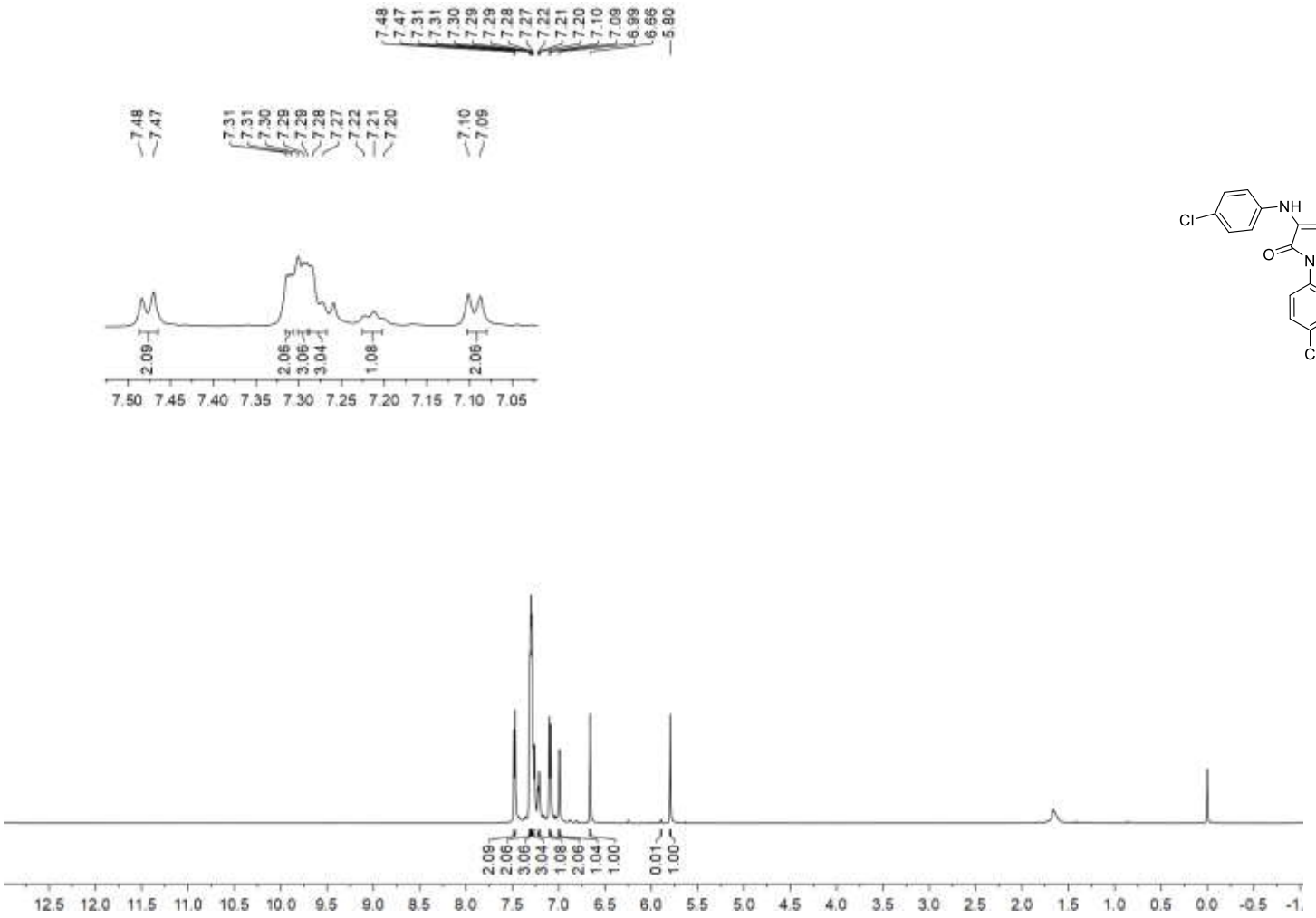


Figure S16. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4g**

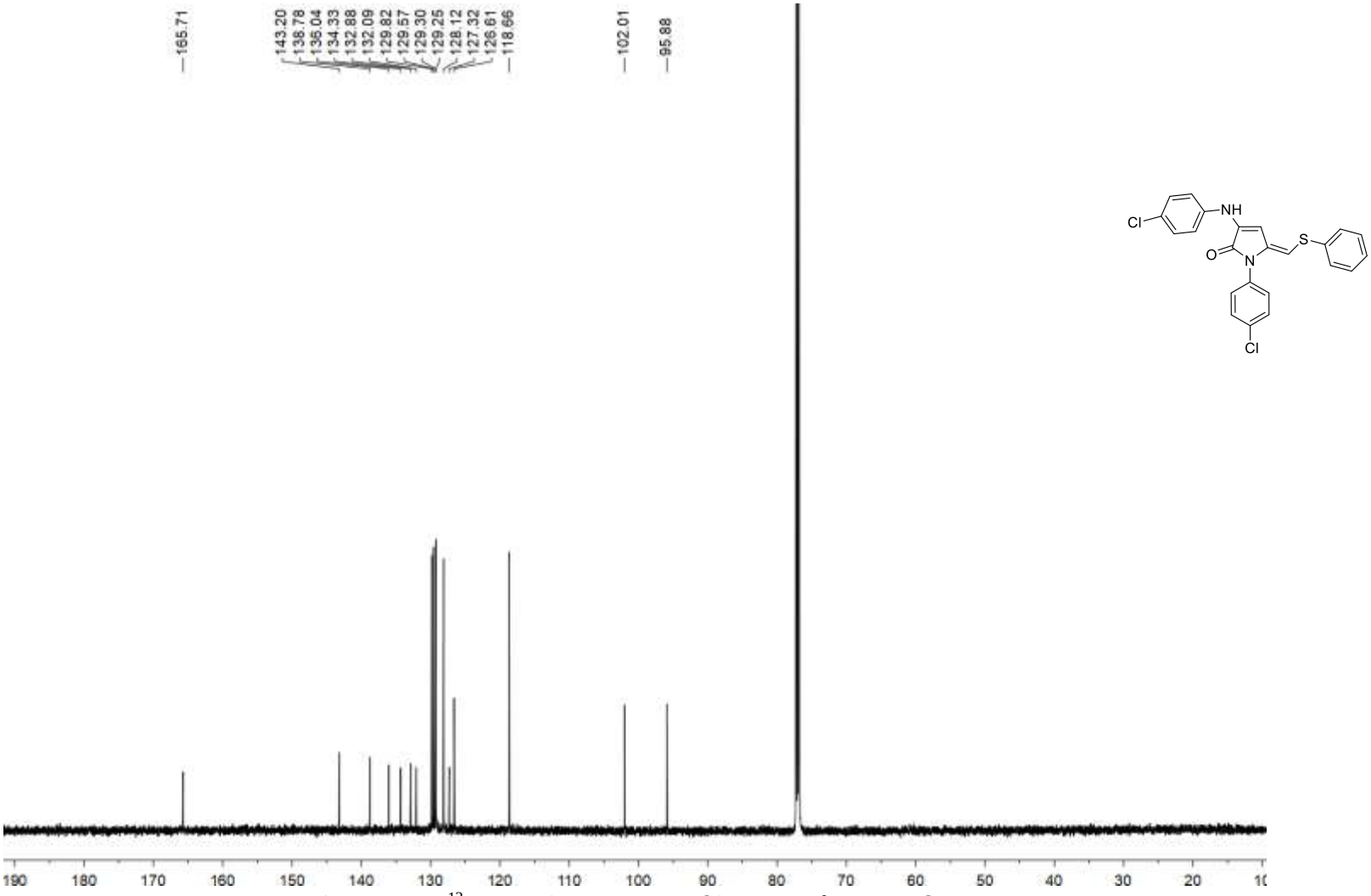


Figure S17. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4g**

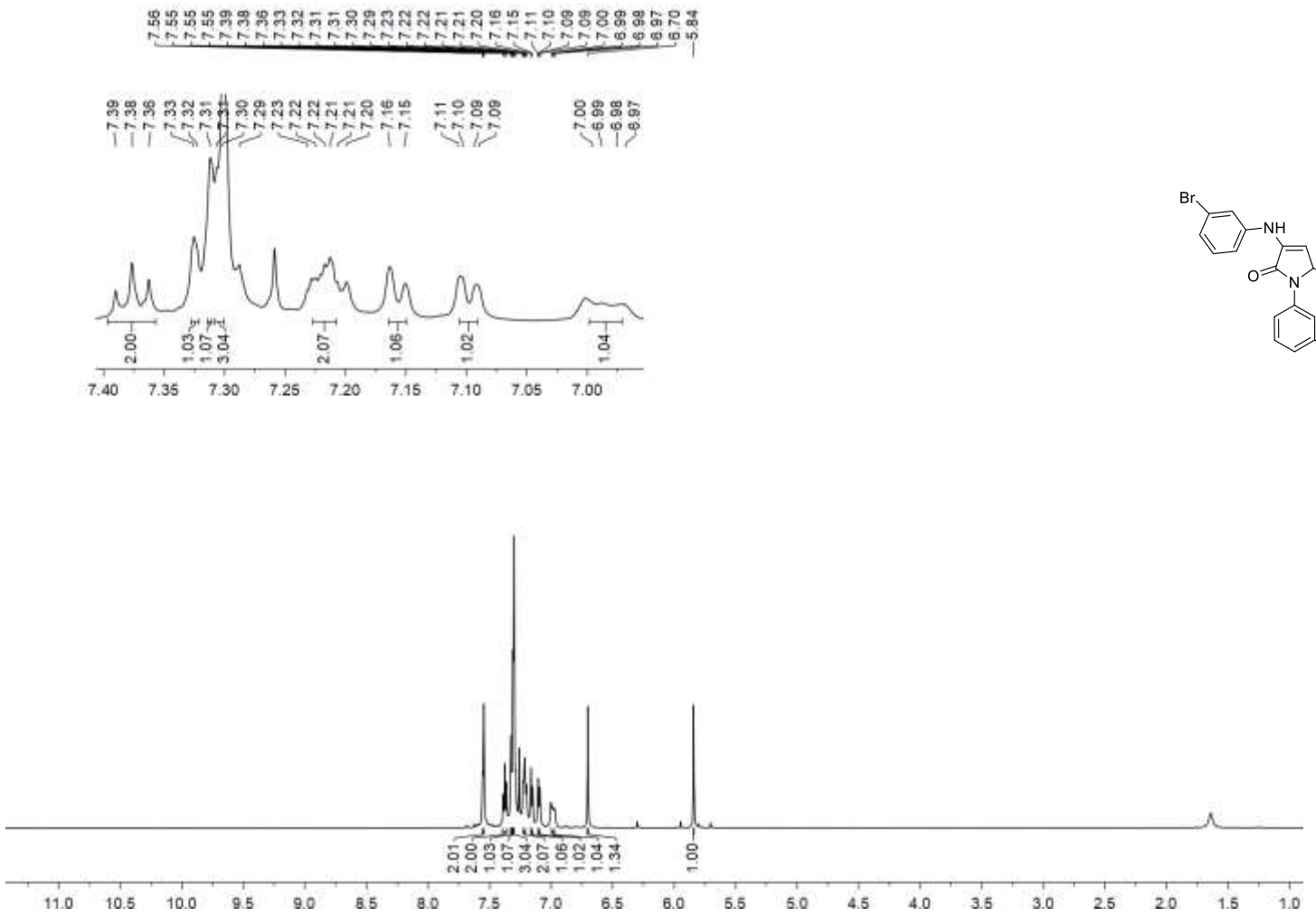


Figure S18. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4h**

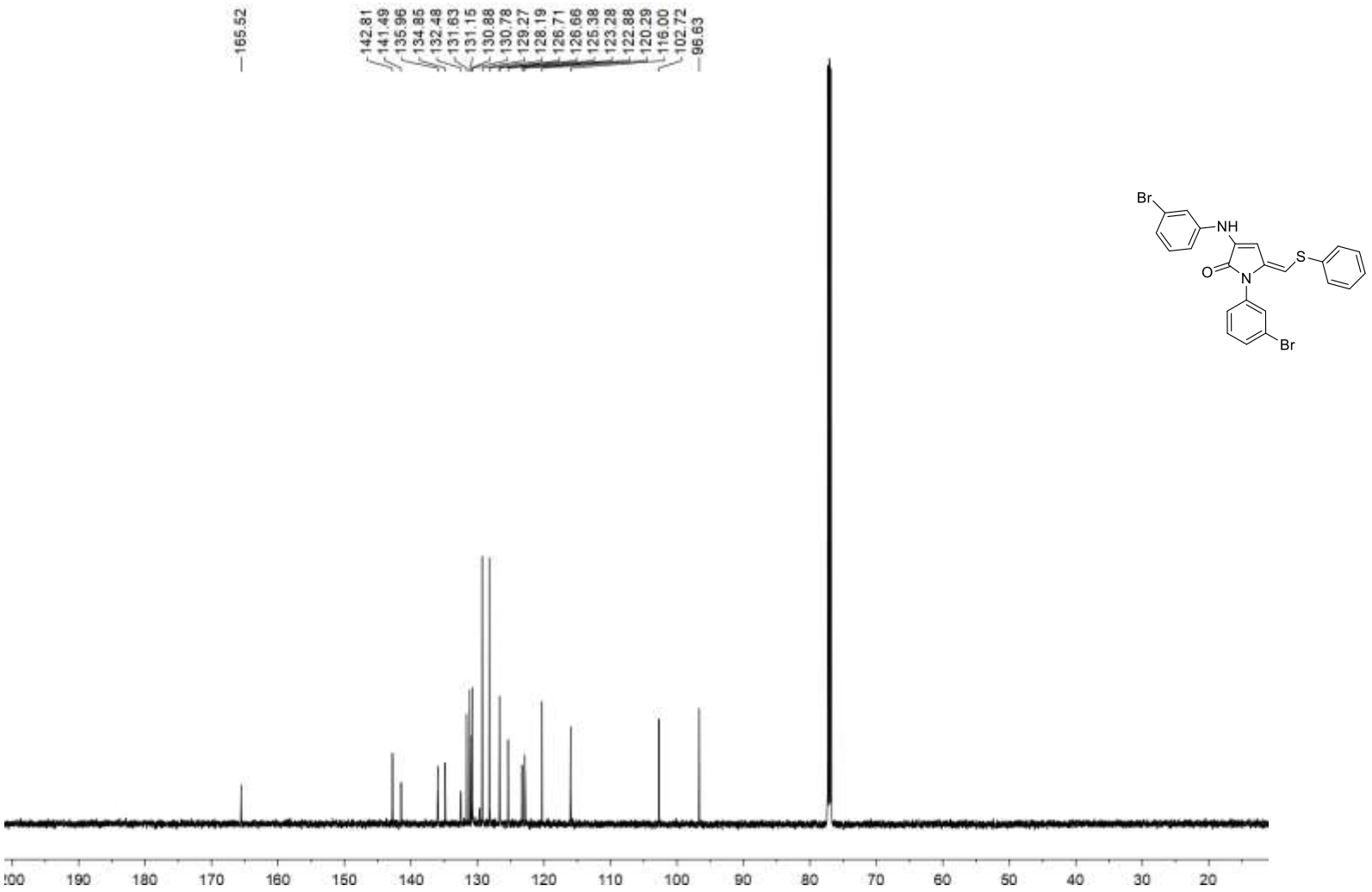


Figure S19. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4h**

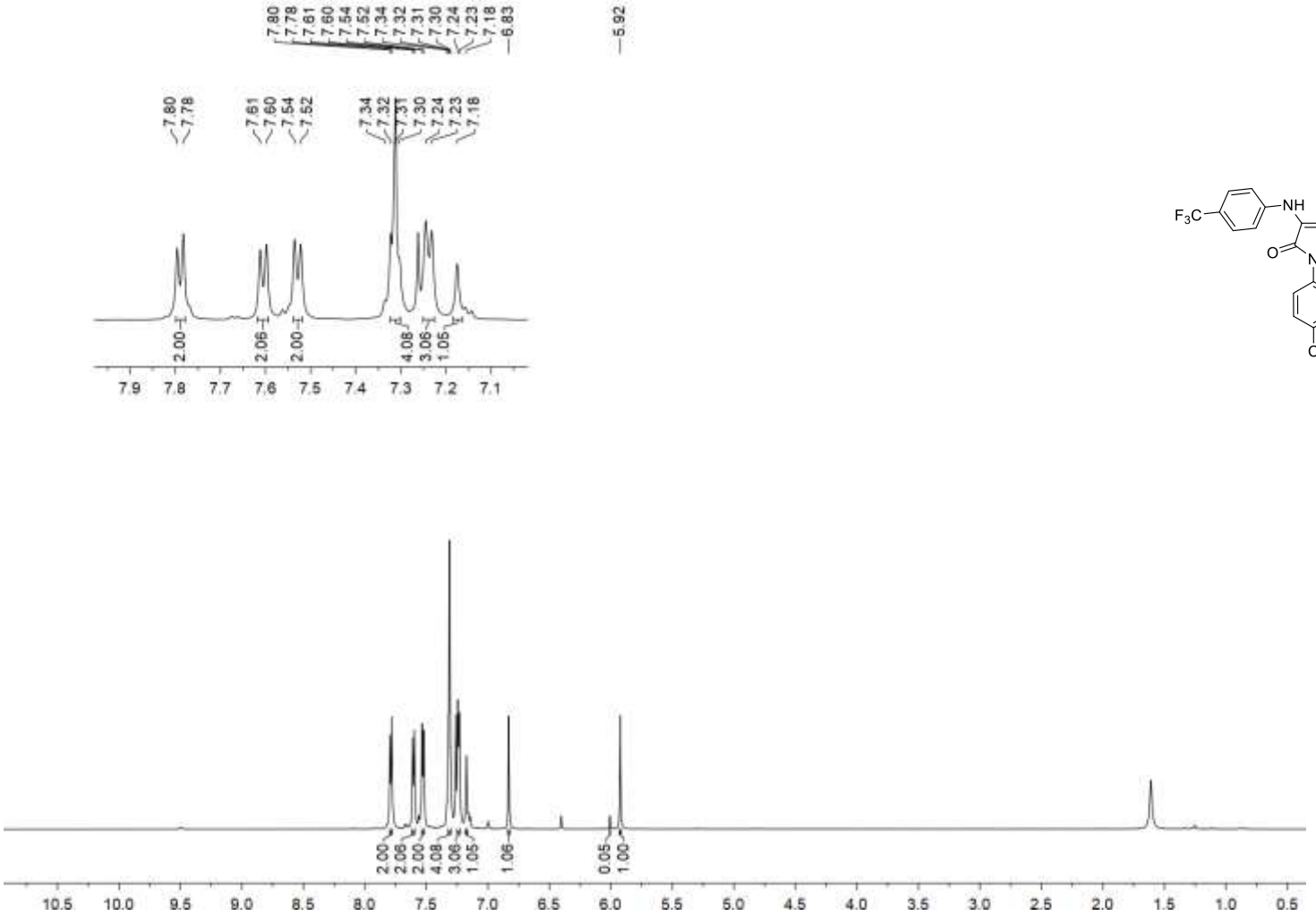


Figure S20. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4i**

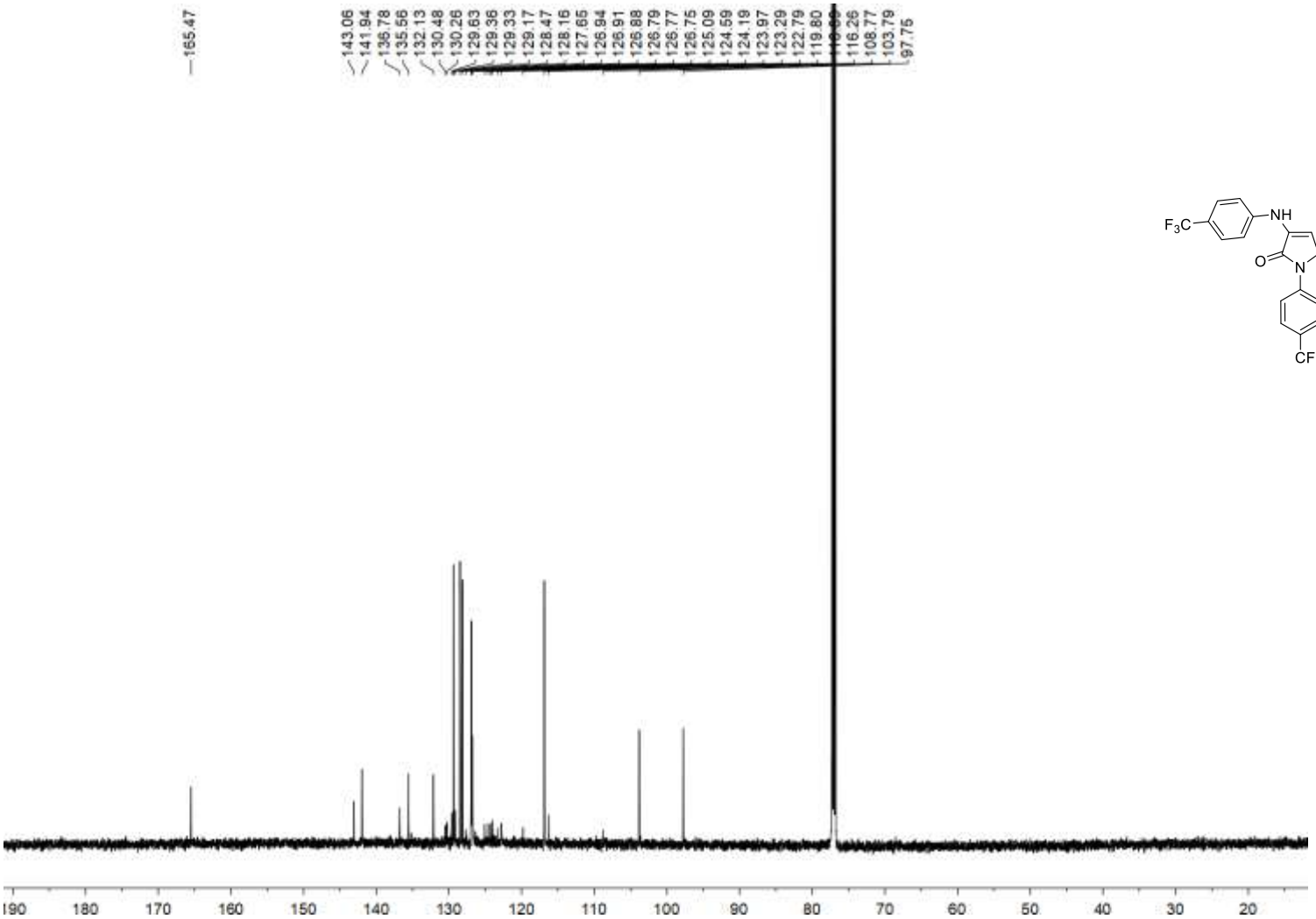


Figure S21. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4i**

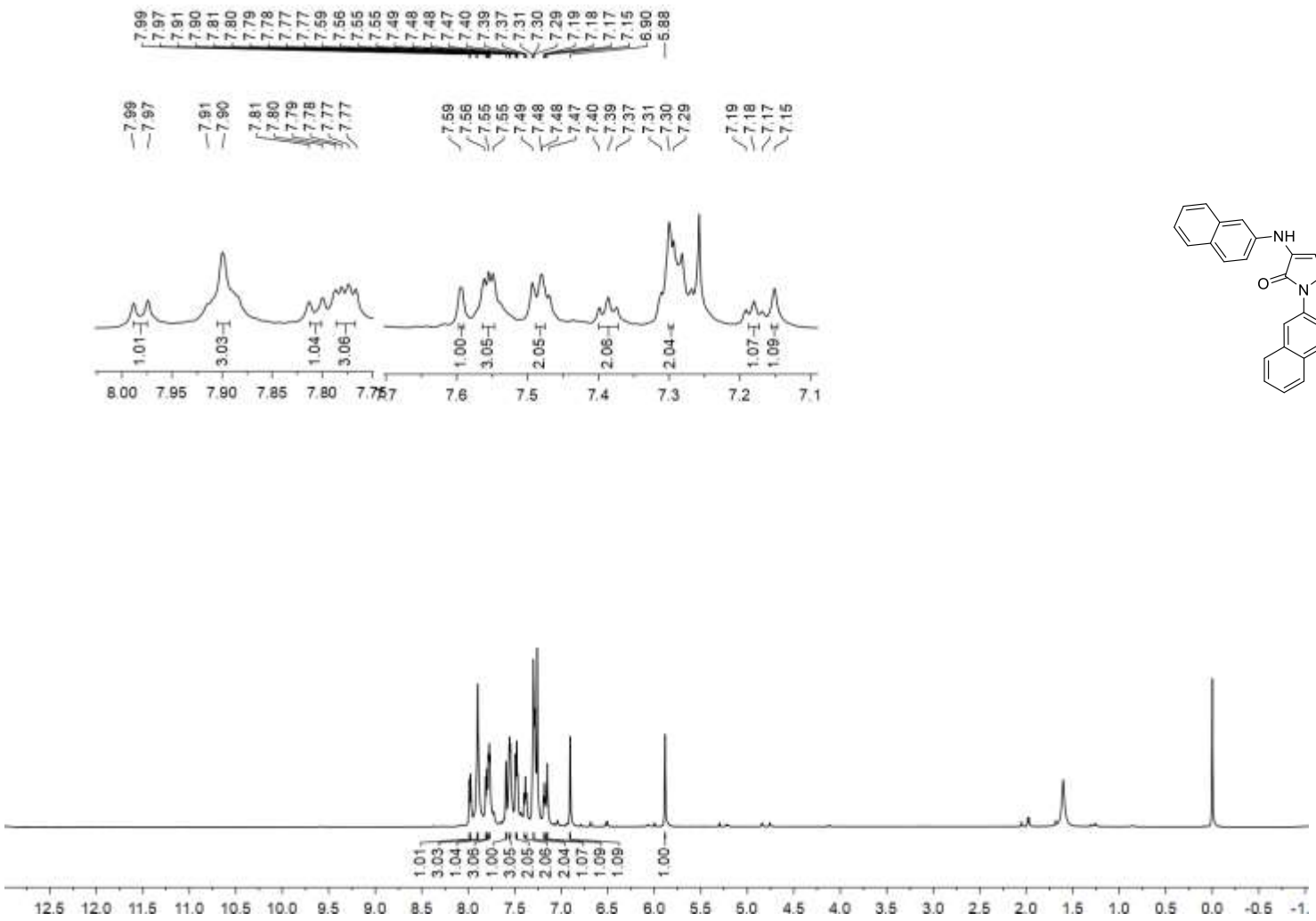


Figure S22. **^{1}H NMR** (600 MHz, $CDCl_3$) spectra of compound **4j**

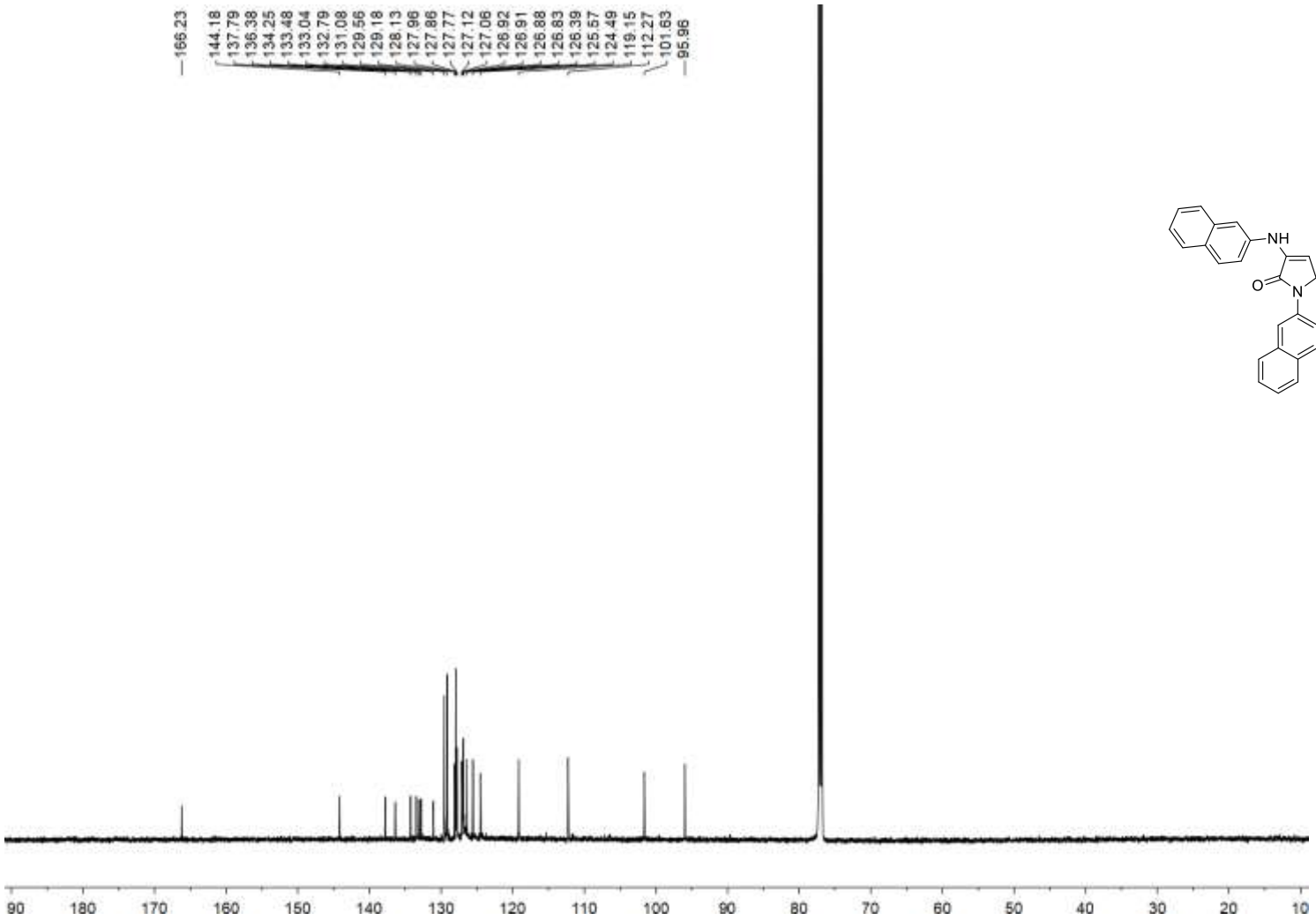


Figure S23. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4j**

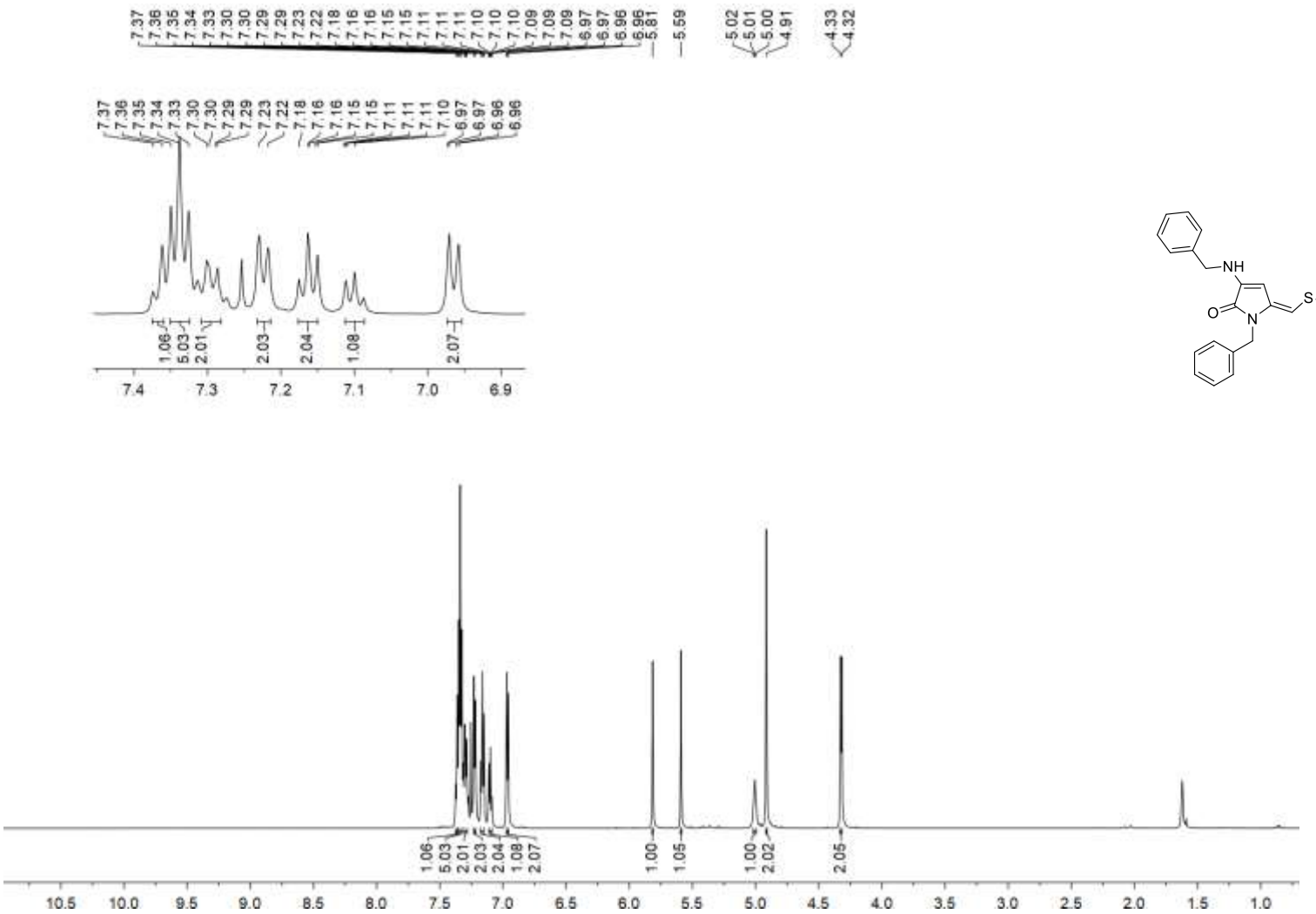


Figure S24. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4k**

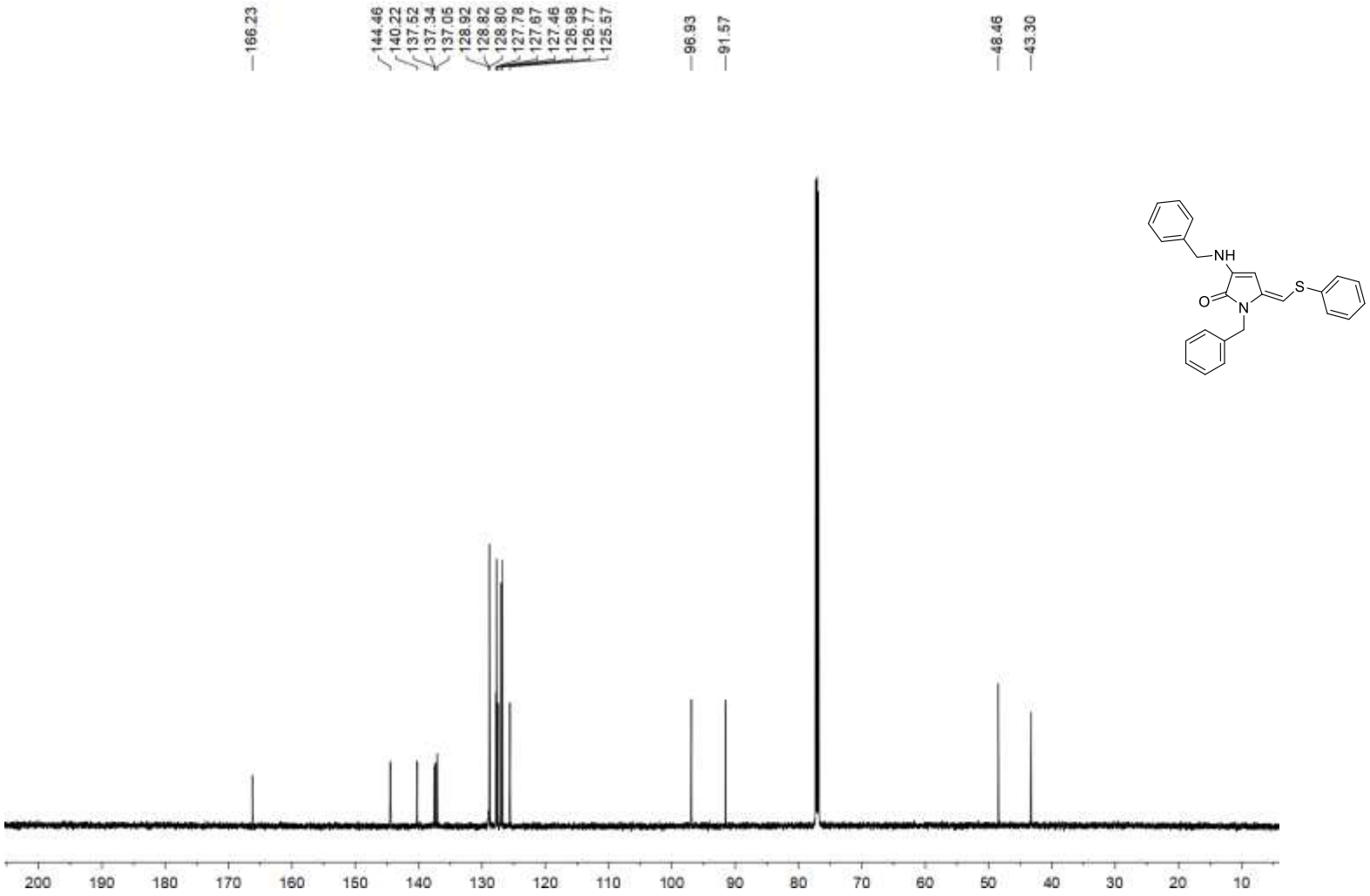


Figure S25. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4k**

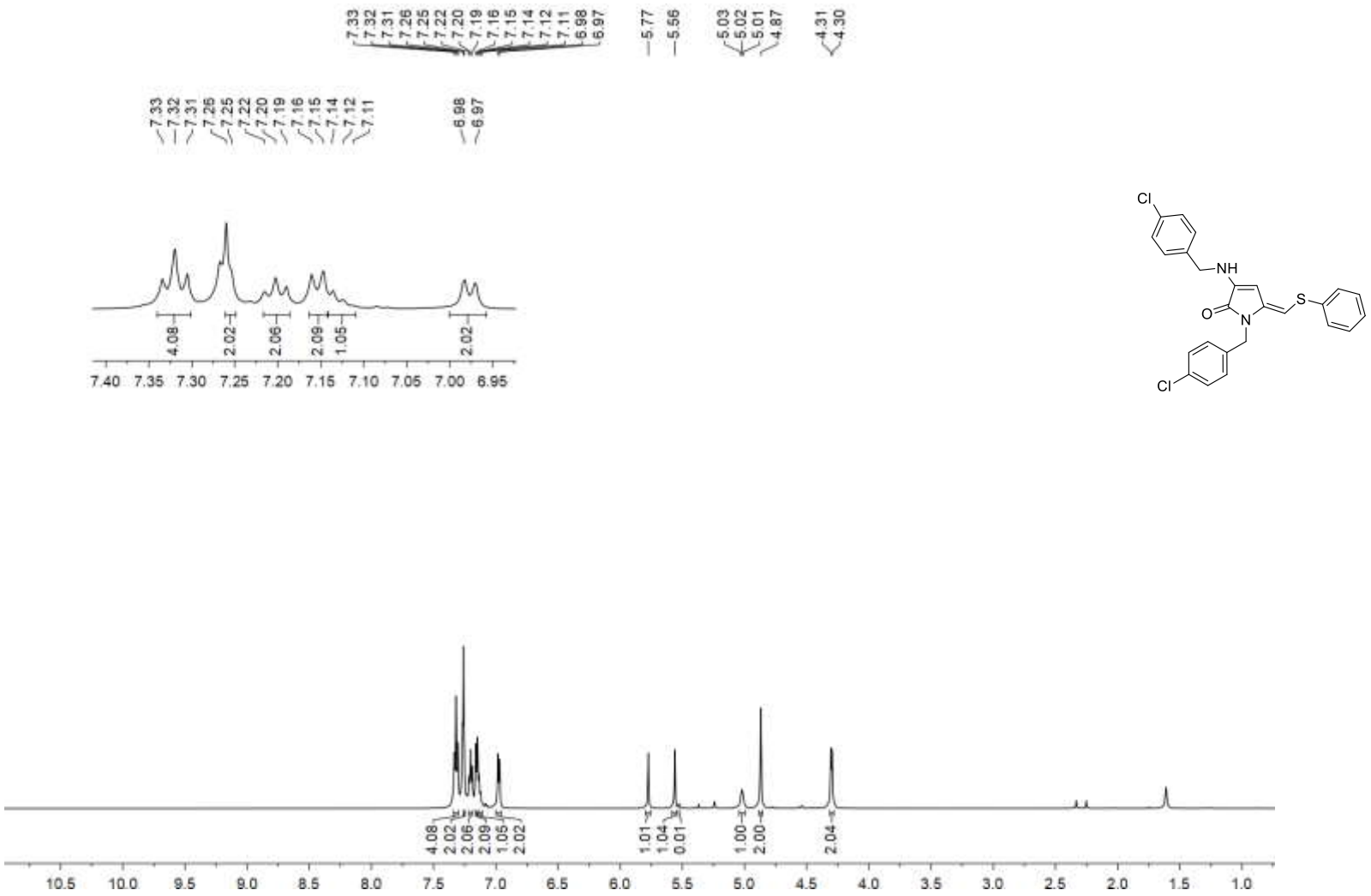


Figure S26. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4l**

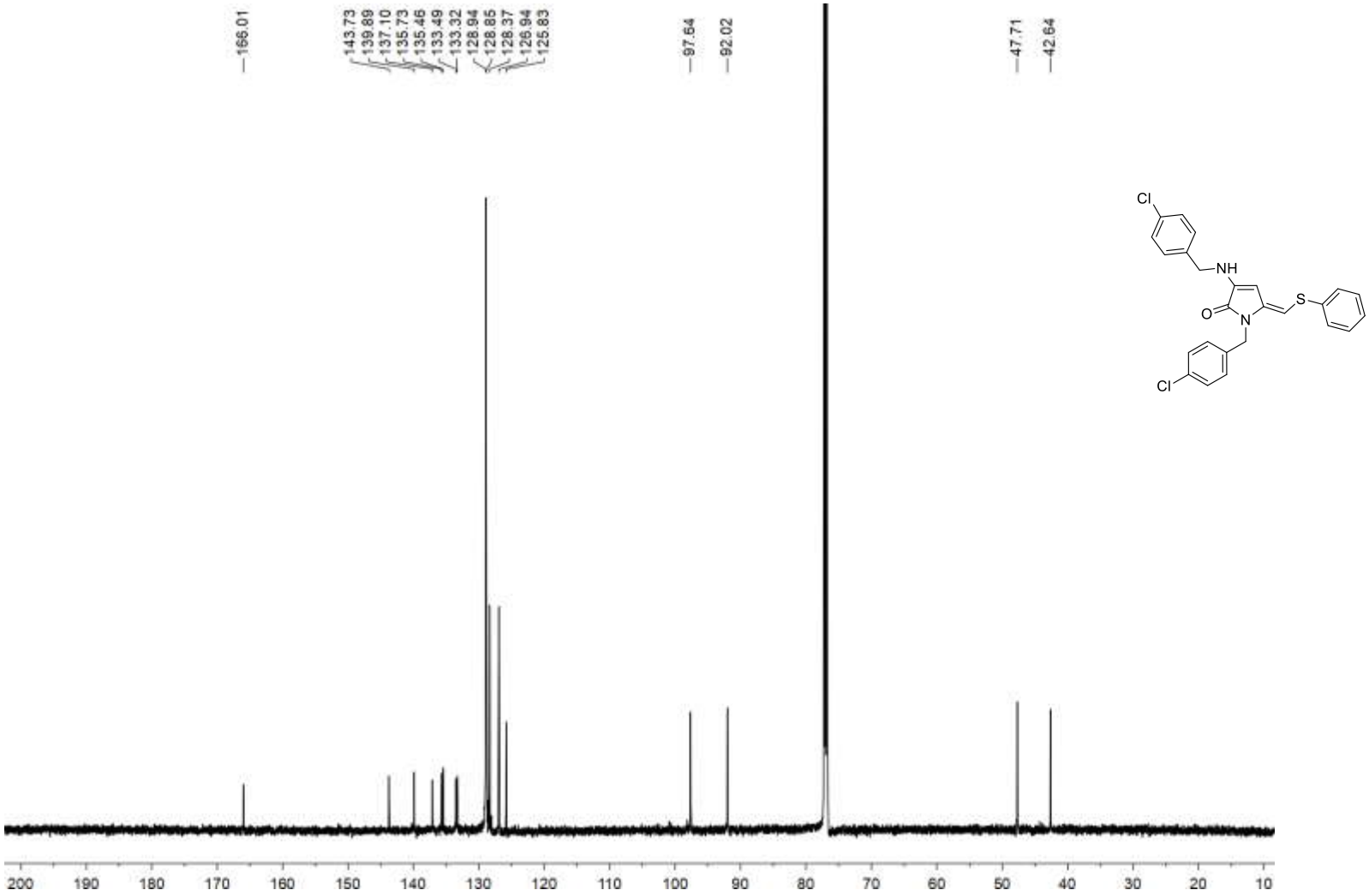


Figure S27. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4l**

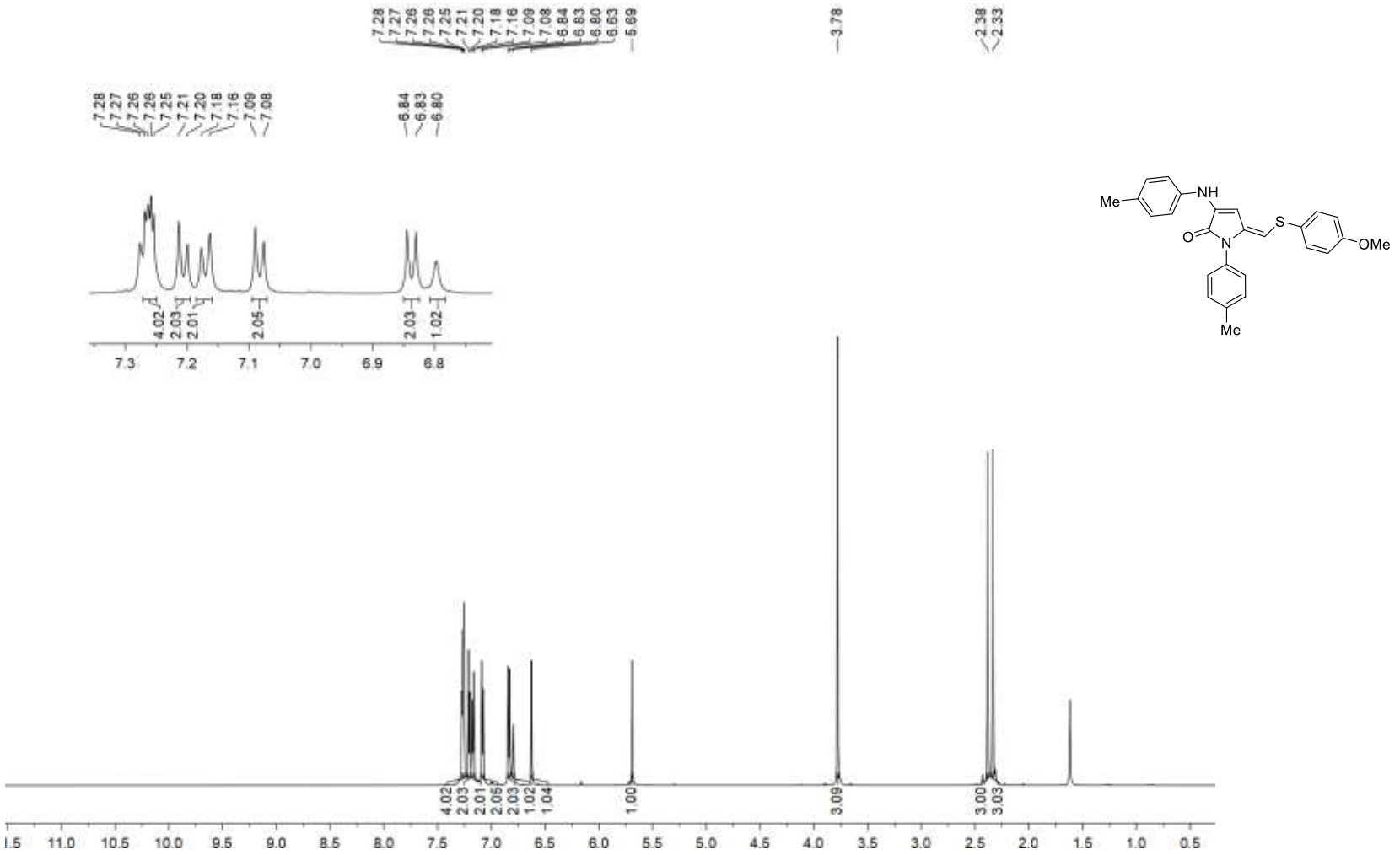


Figure S28. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4m**

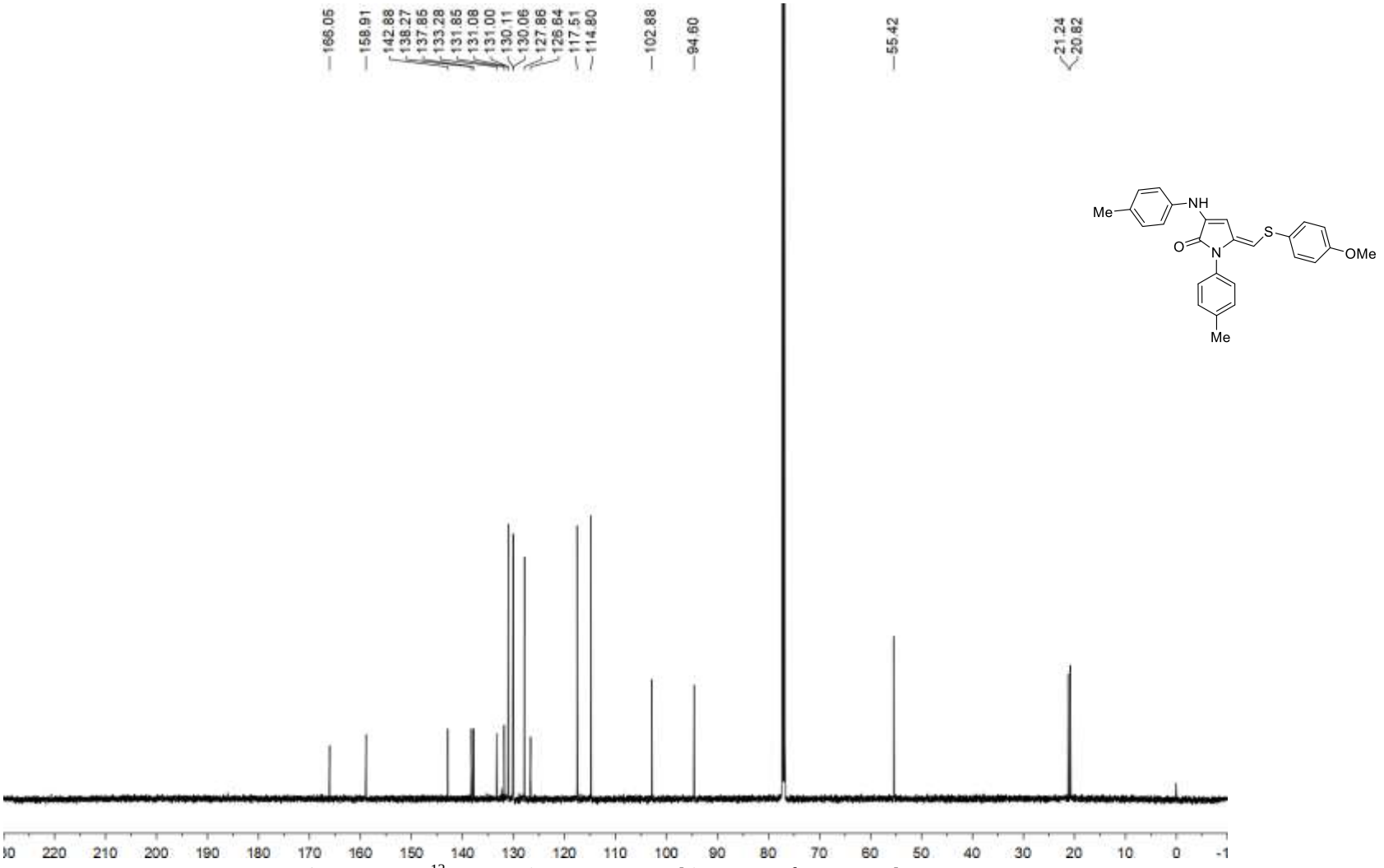


Figure S29. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4m**

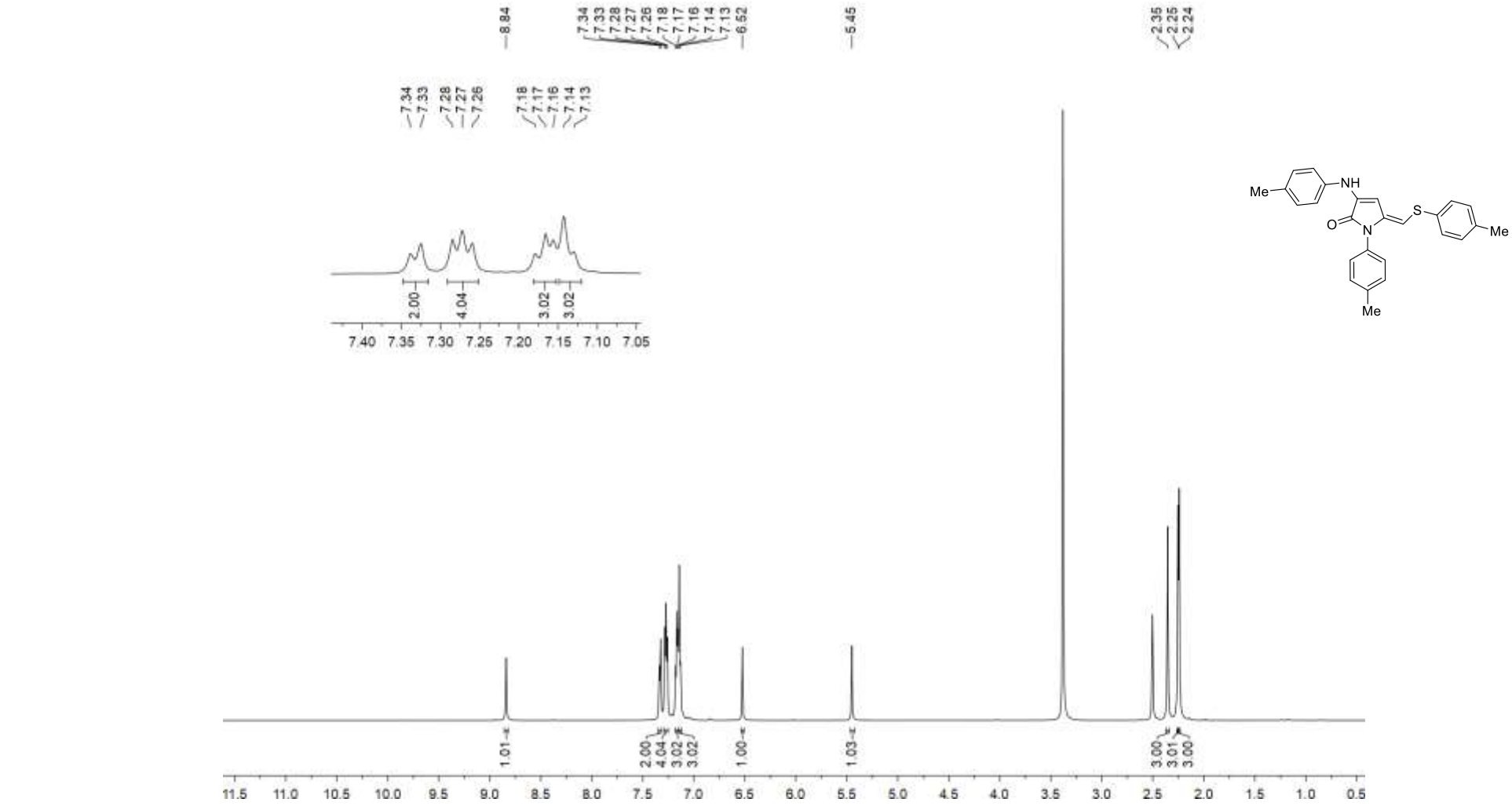


Figure S30. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **4n**

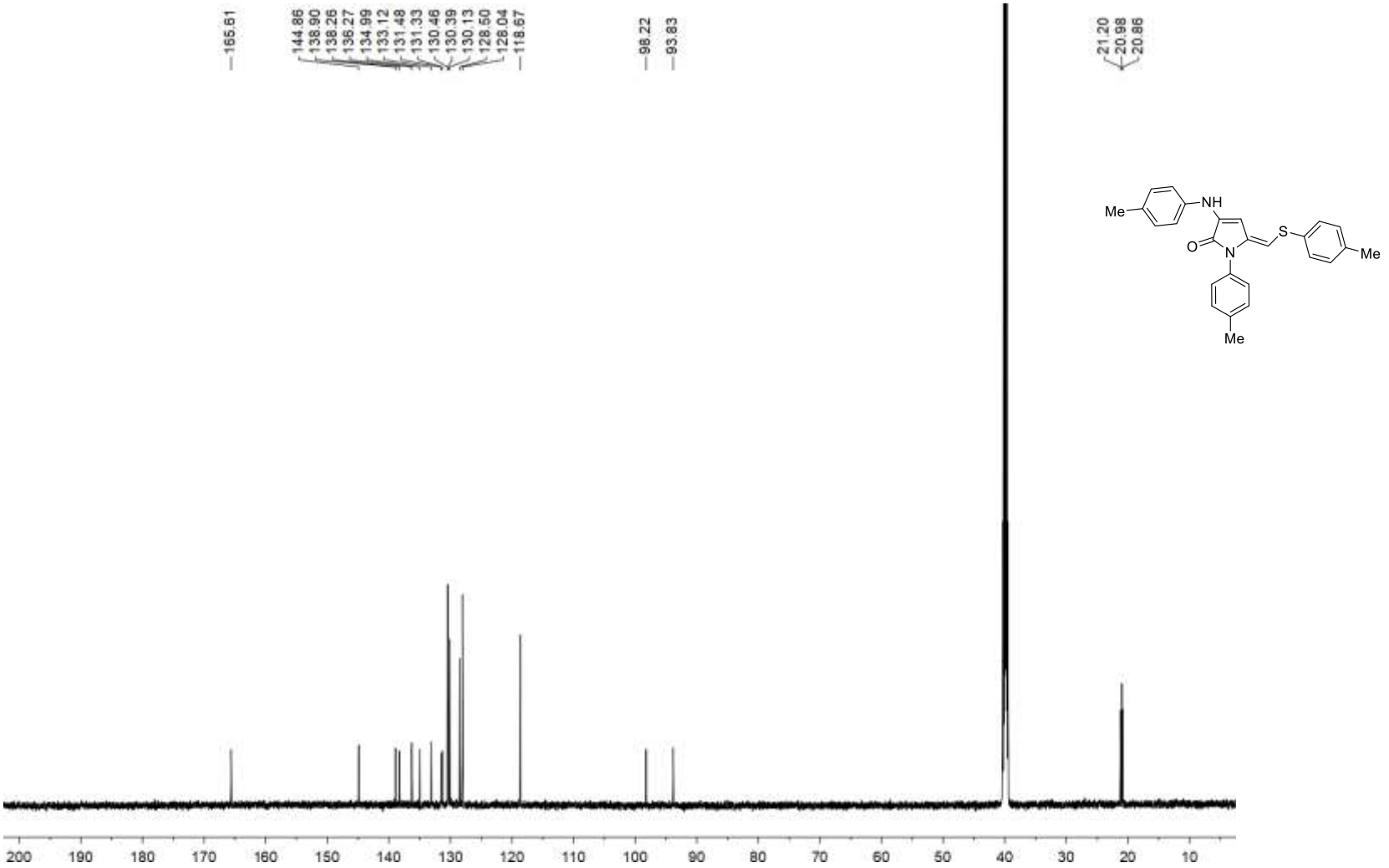


Figure S31. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound 4**n**

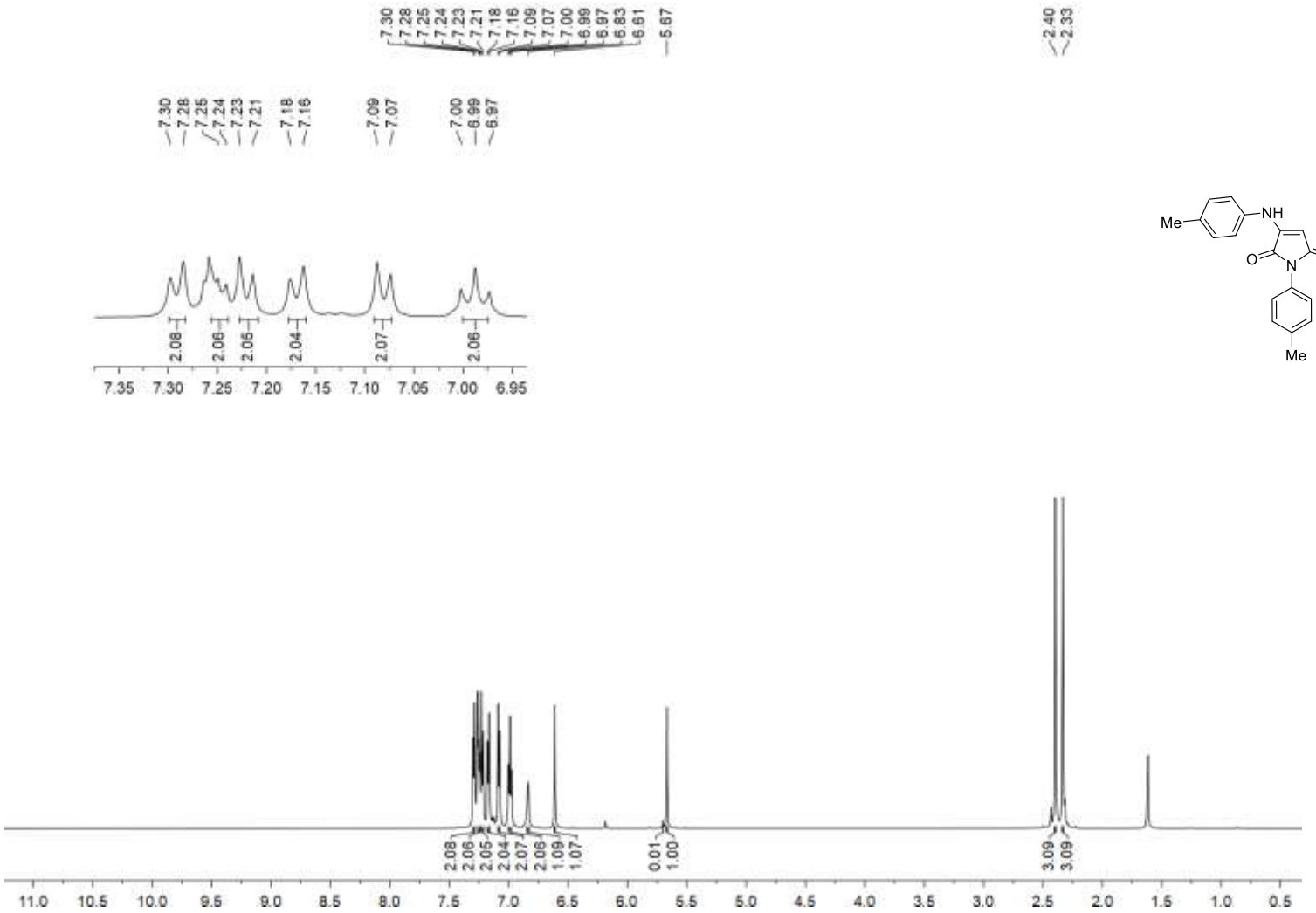


Figure S32. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4o**

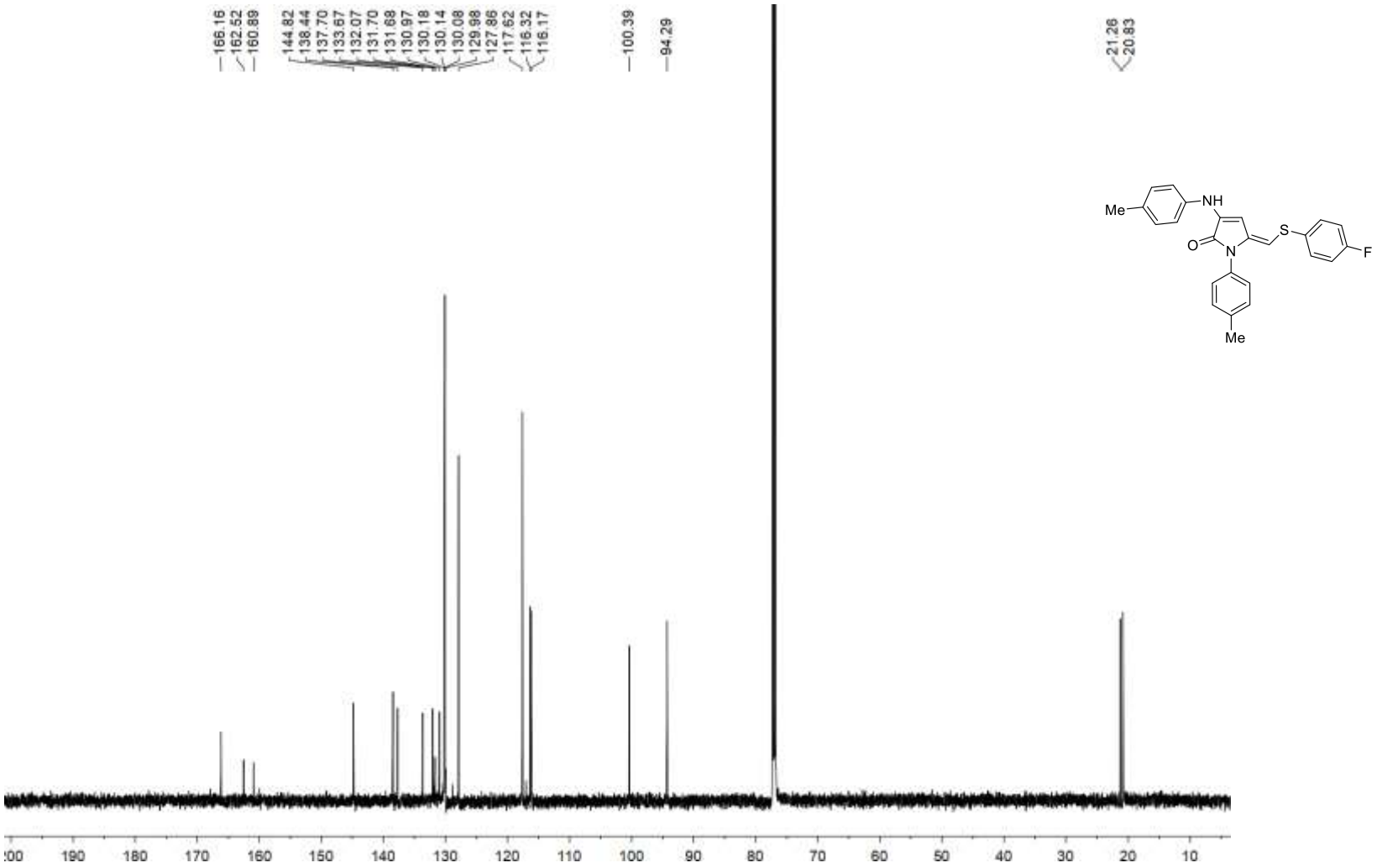


Figure S33. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4o**

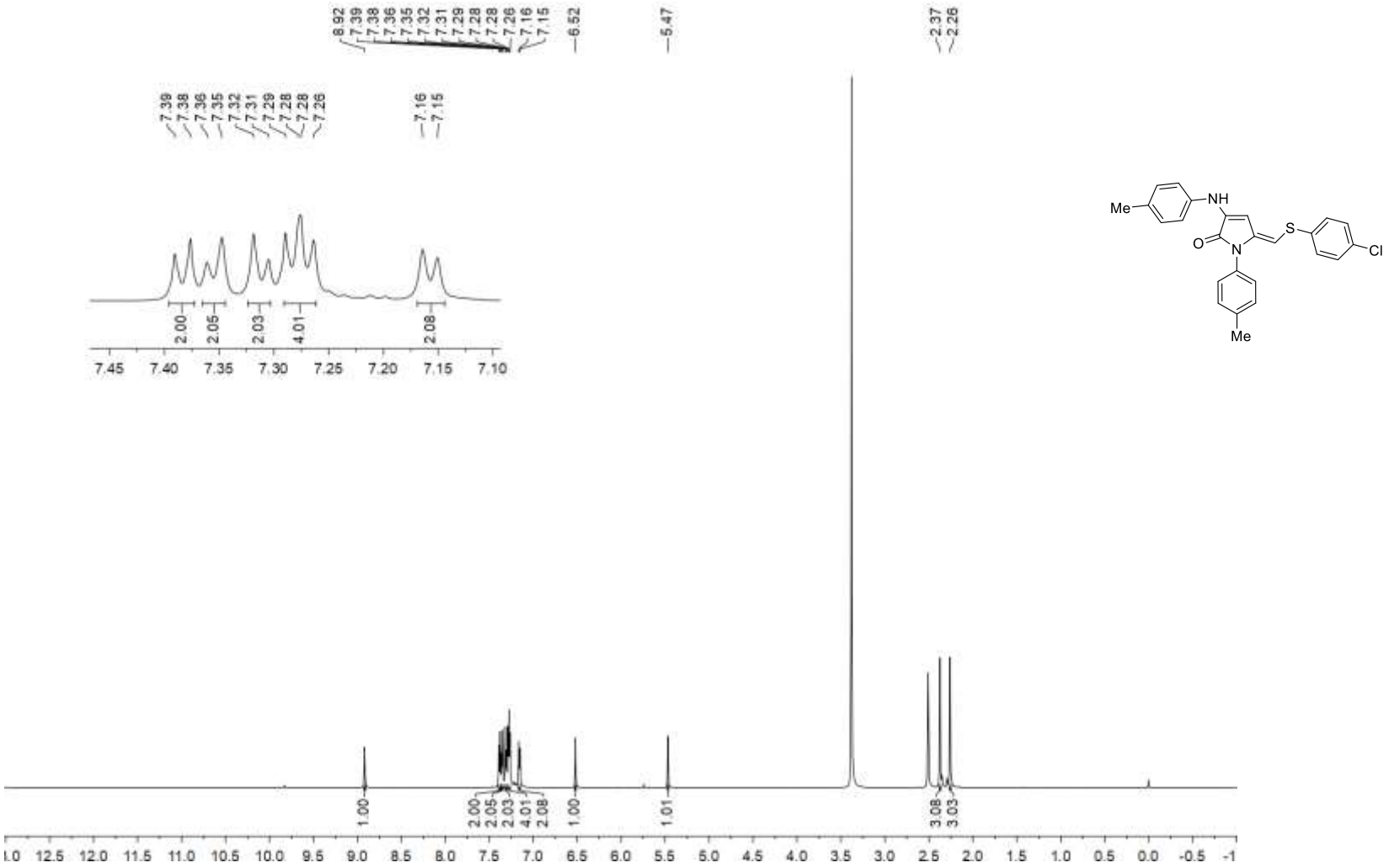


Figure S34. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **4p**

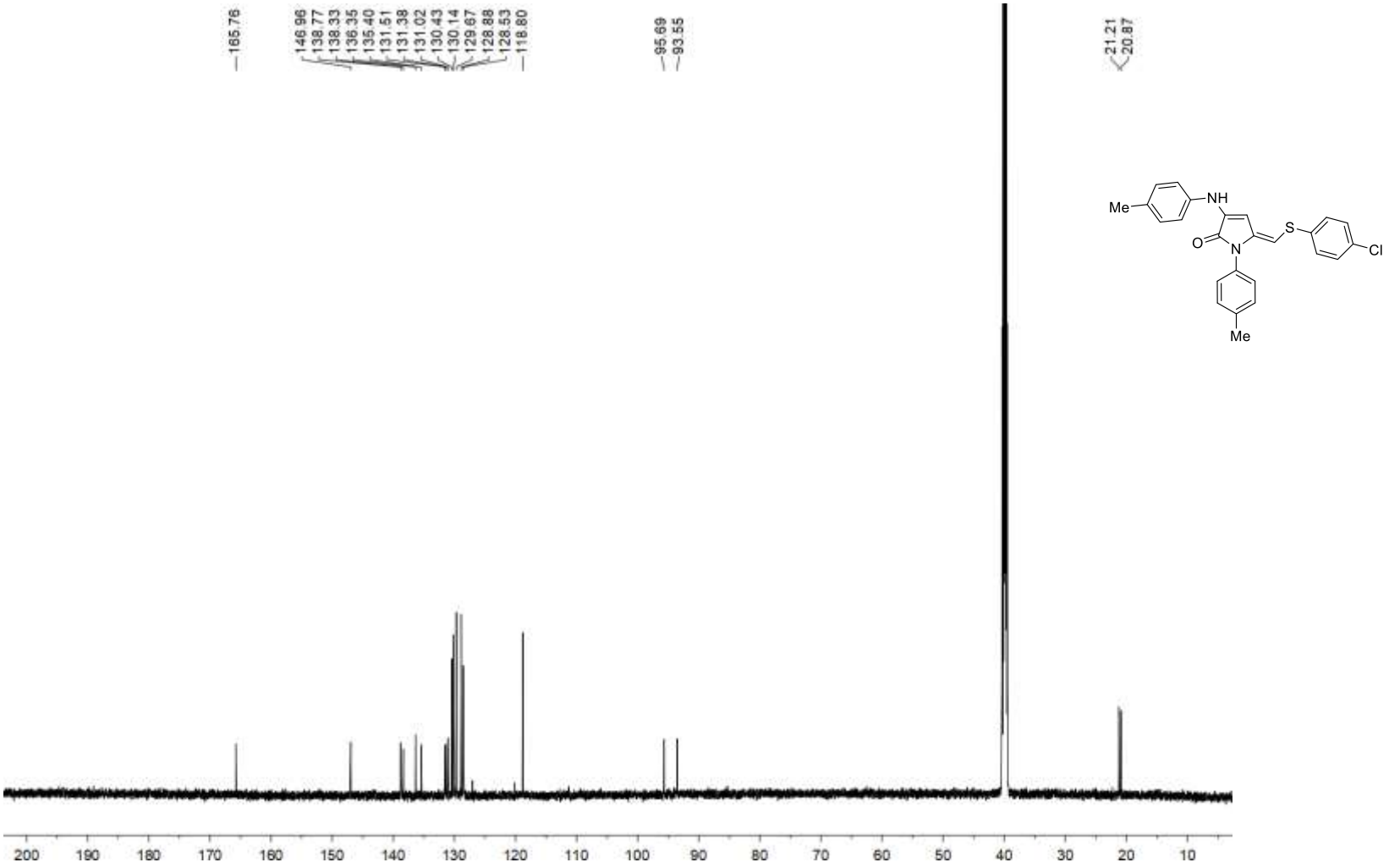


Figure S35. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **4p**

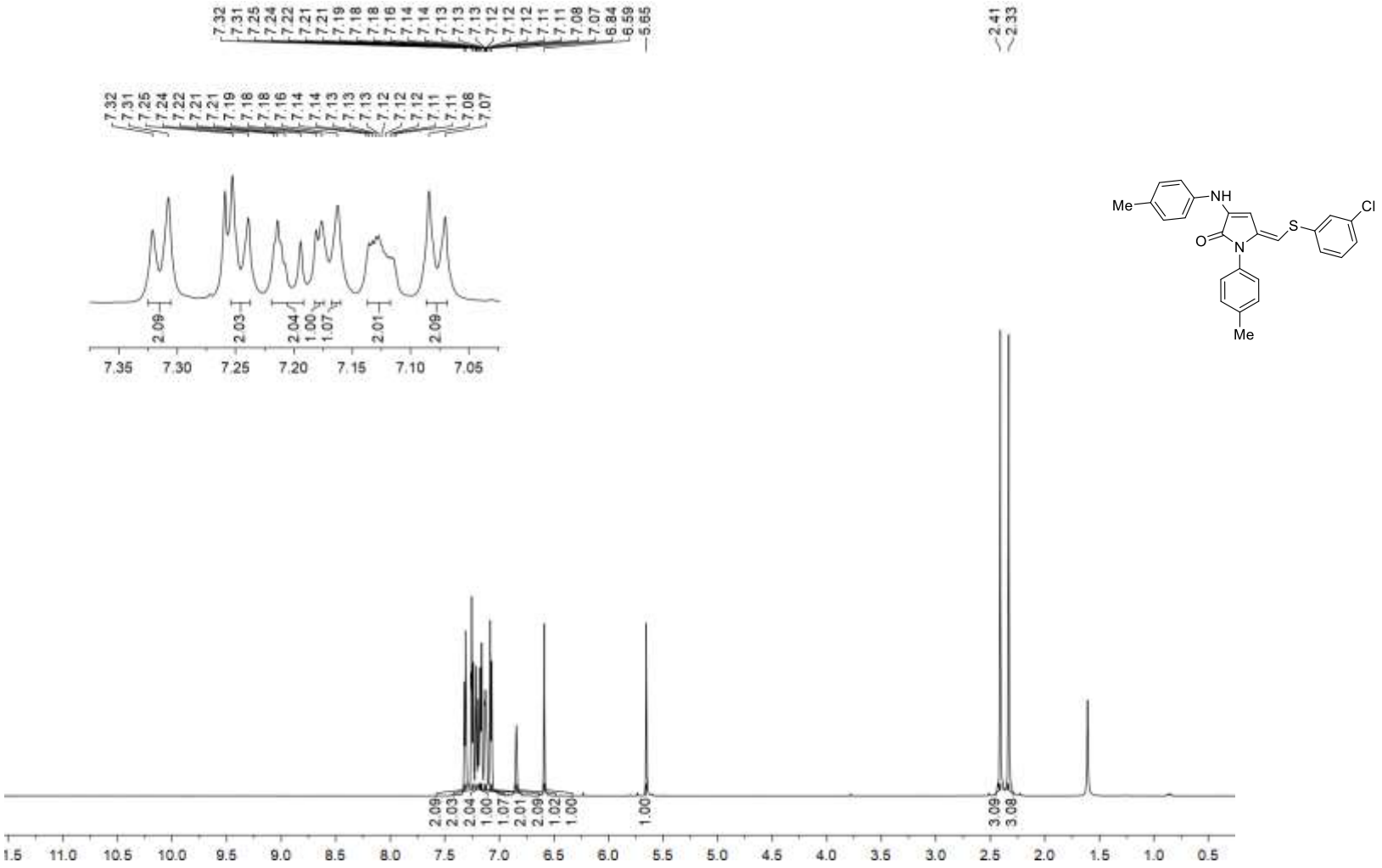


Figure S36. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4q**

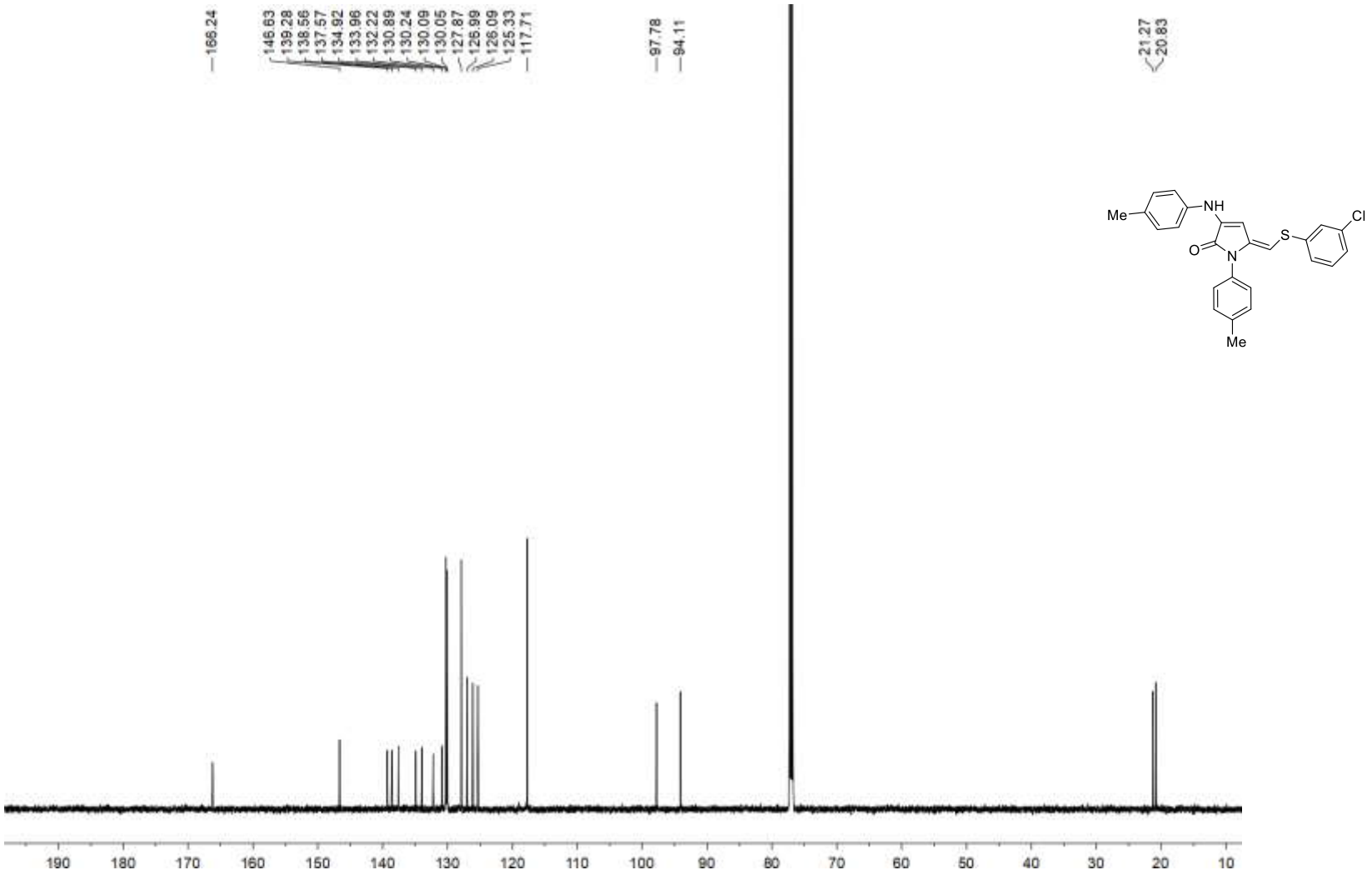


Figure S37. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4q**

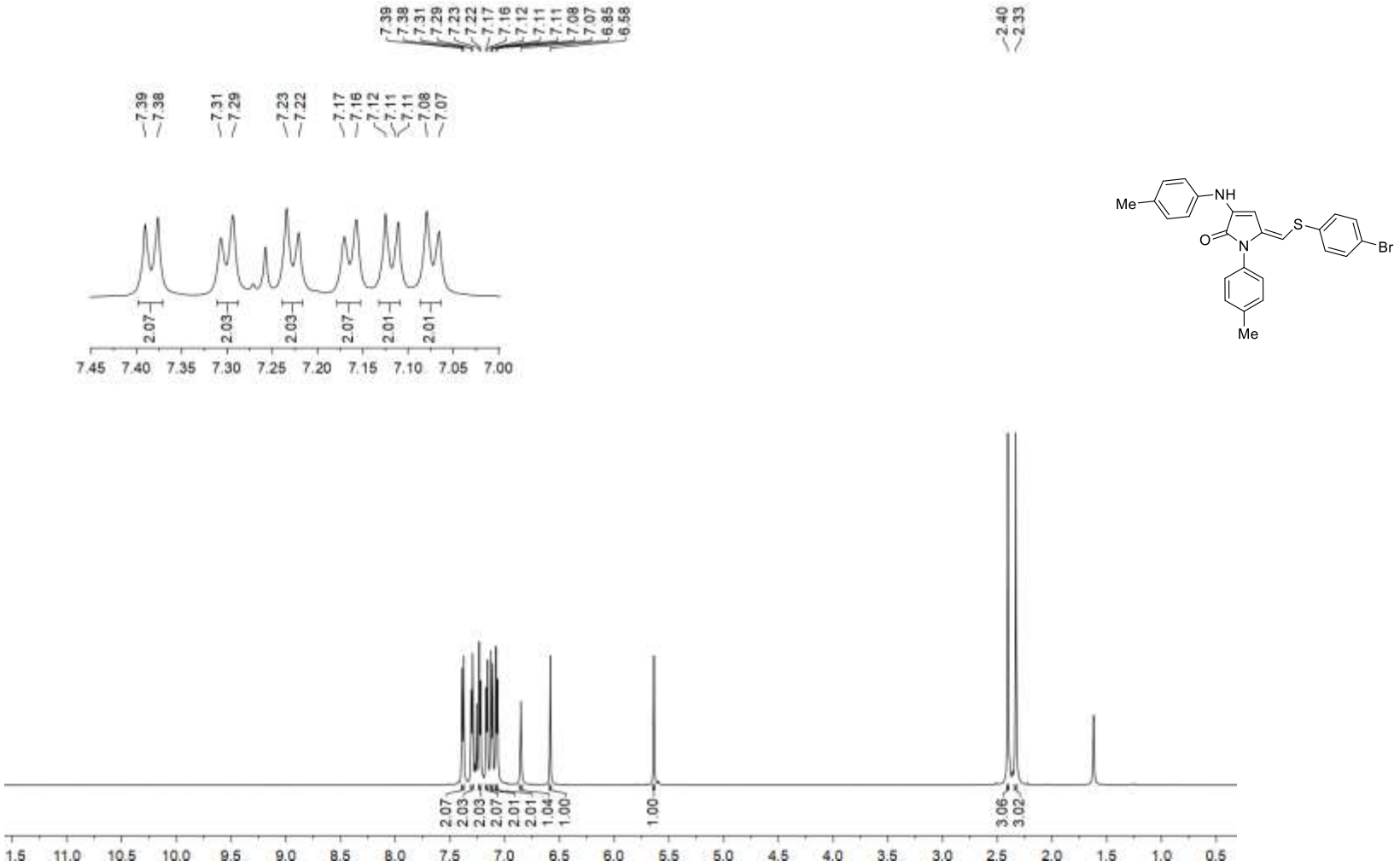


Figure S38. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4r**

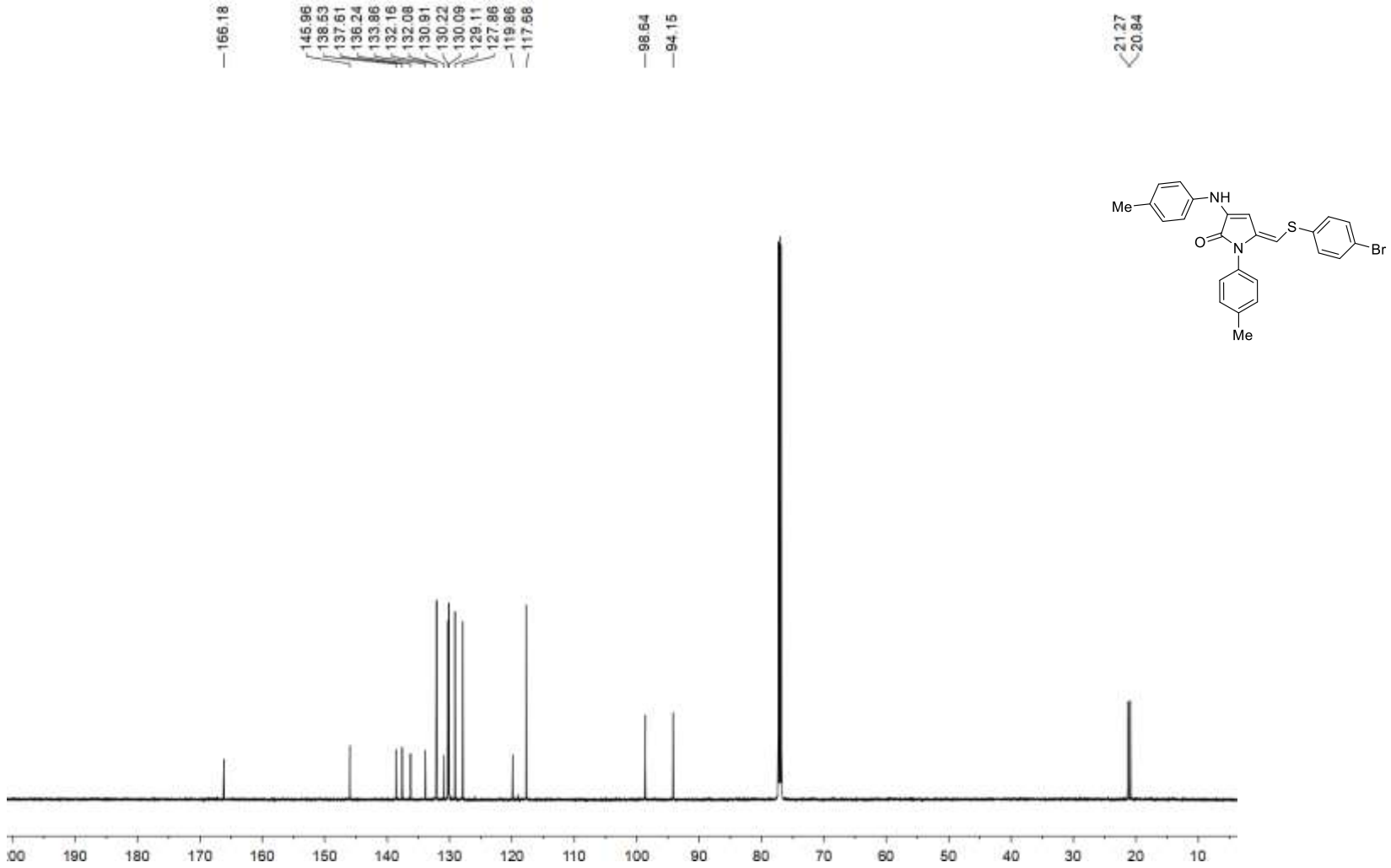


Figure S39. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4r**

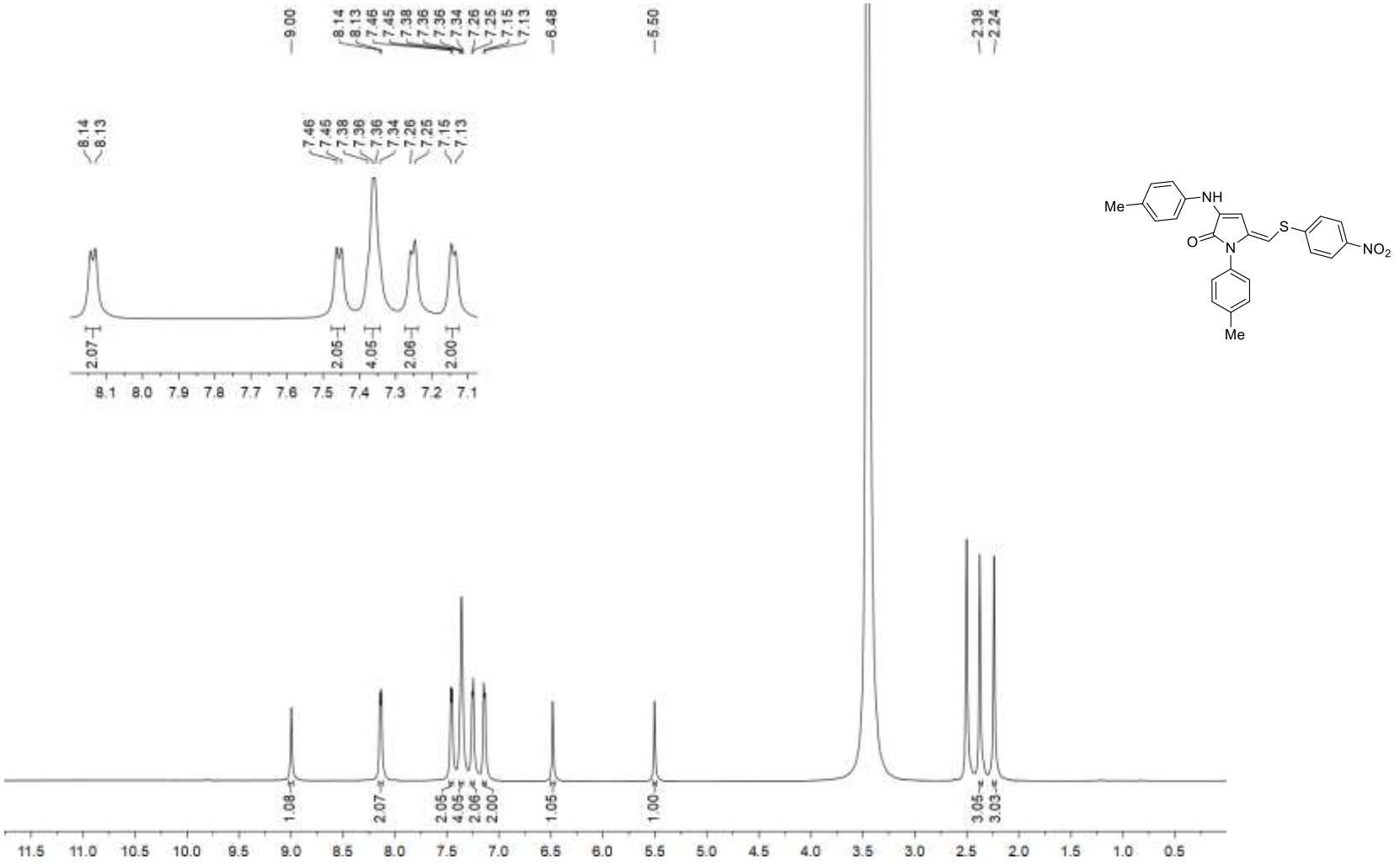


Figure S40. [1]**H NMR** (600 MHz, DMSO-d_6) spectra of compound **4s**

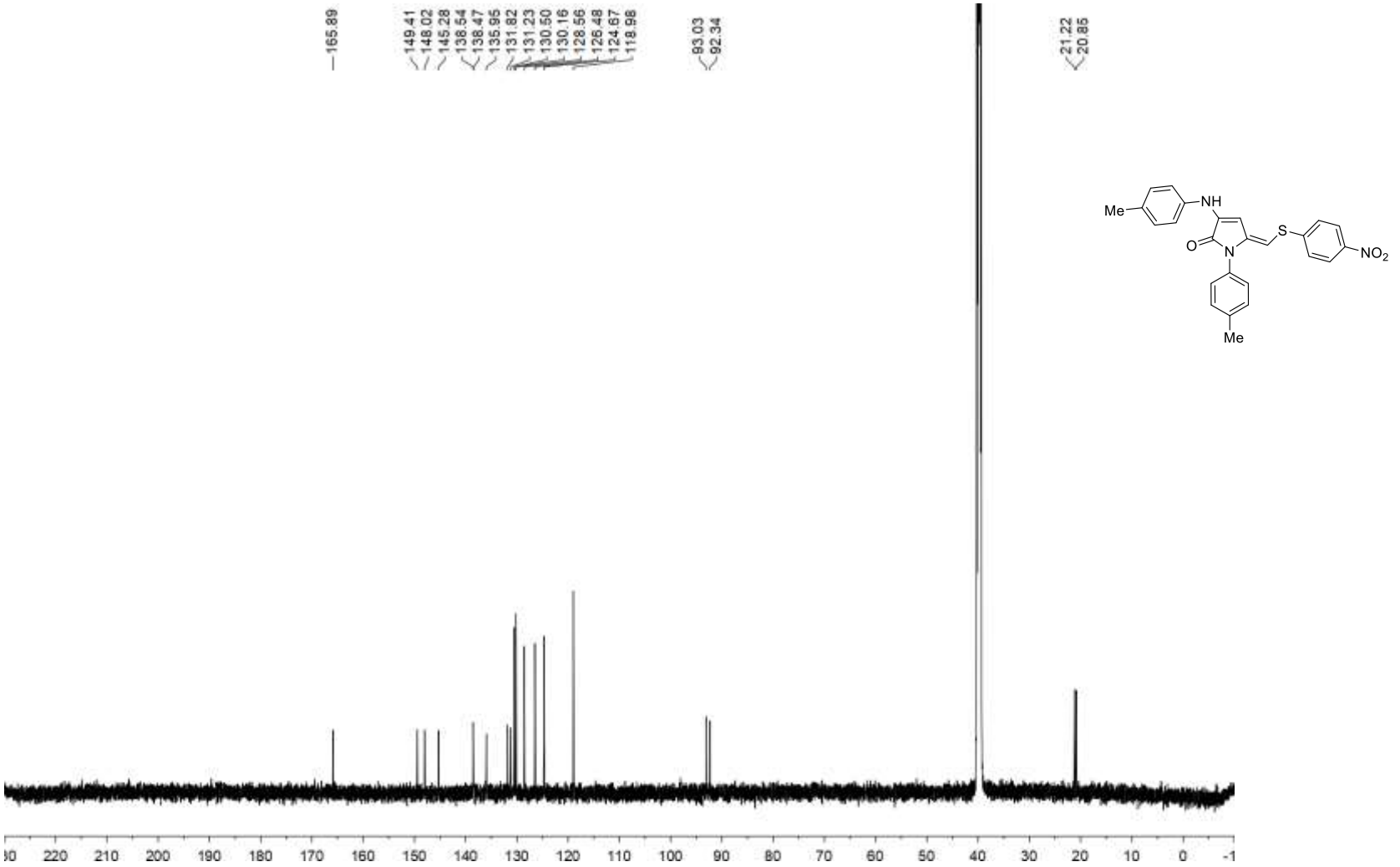


Figure S41. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **4s**

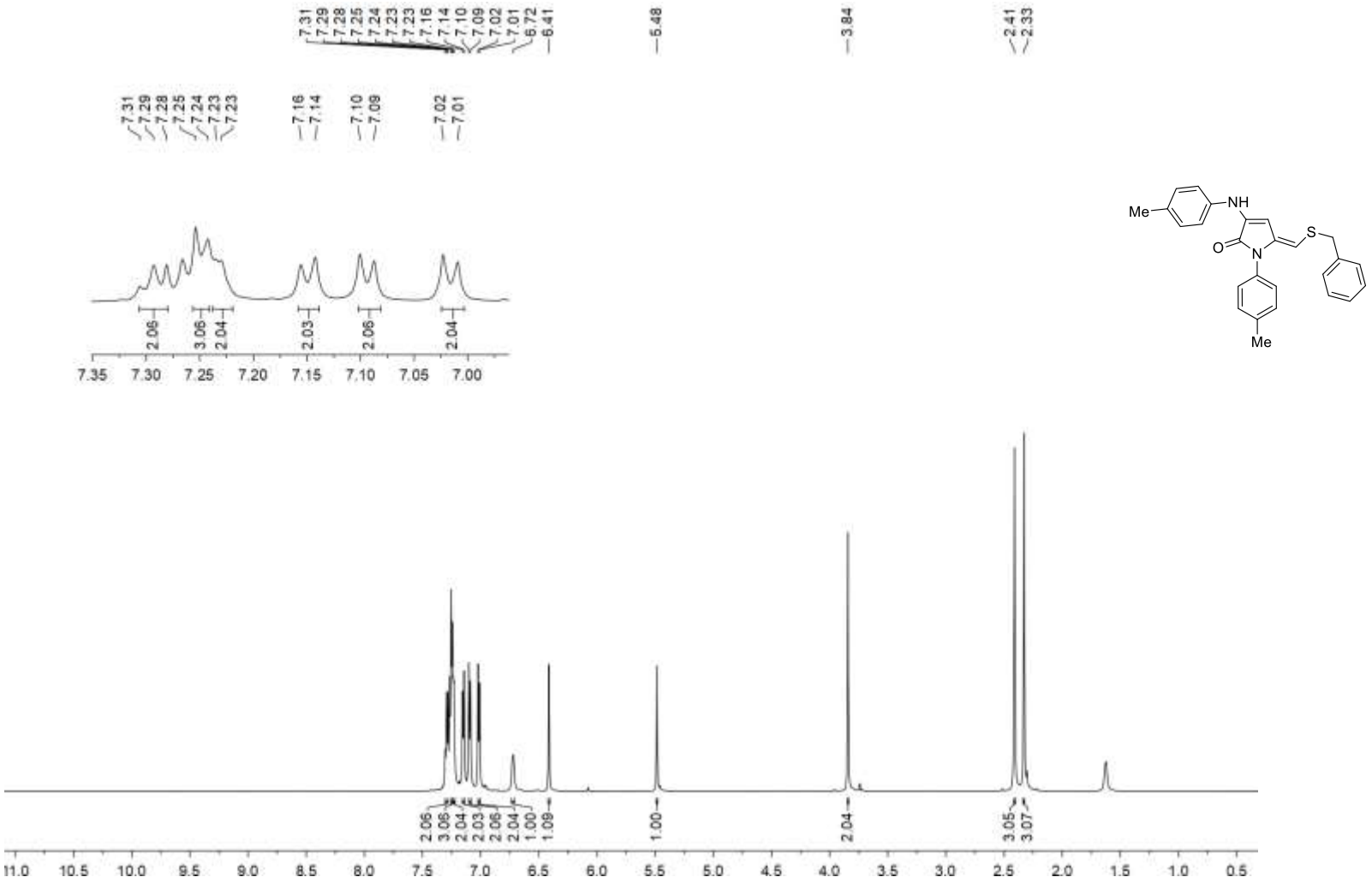


Figure S42. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4t**

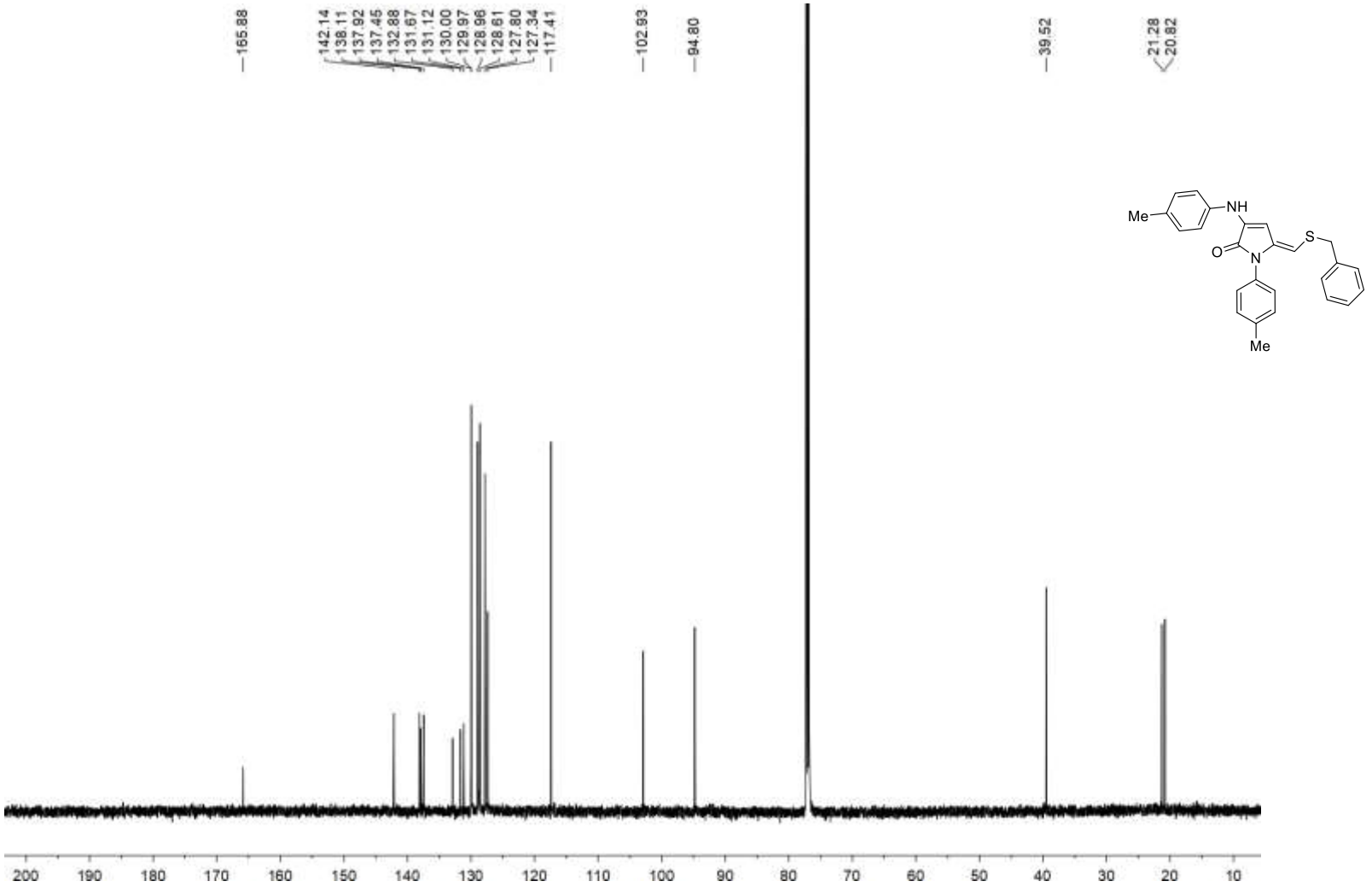


Figure S43. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4t**

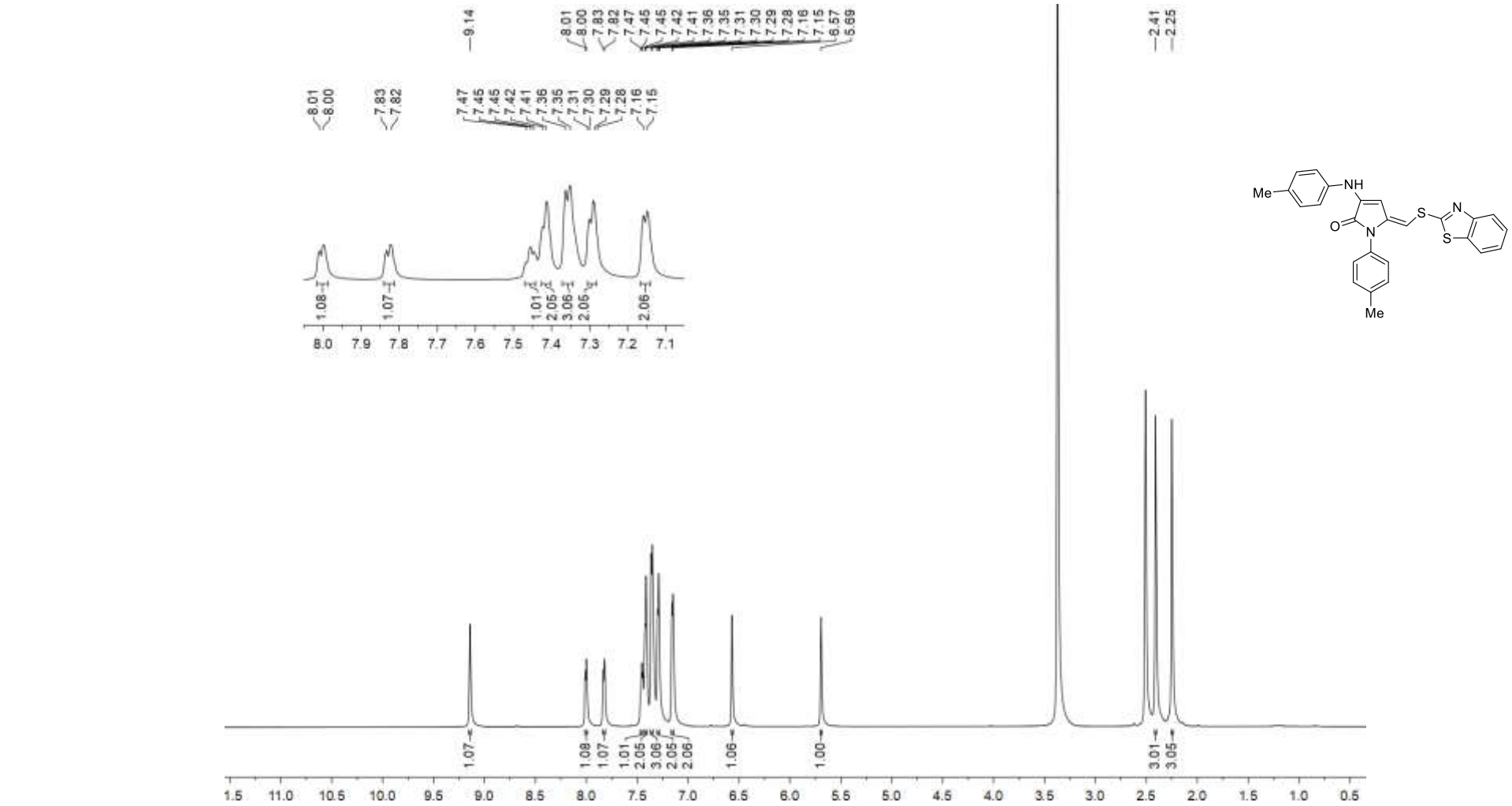


Figure S44. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **4u**

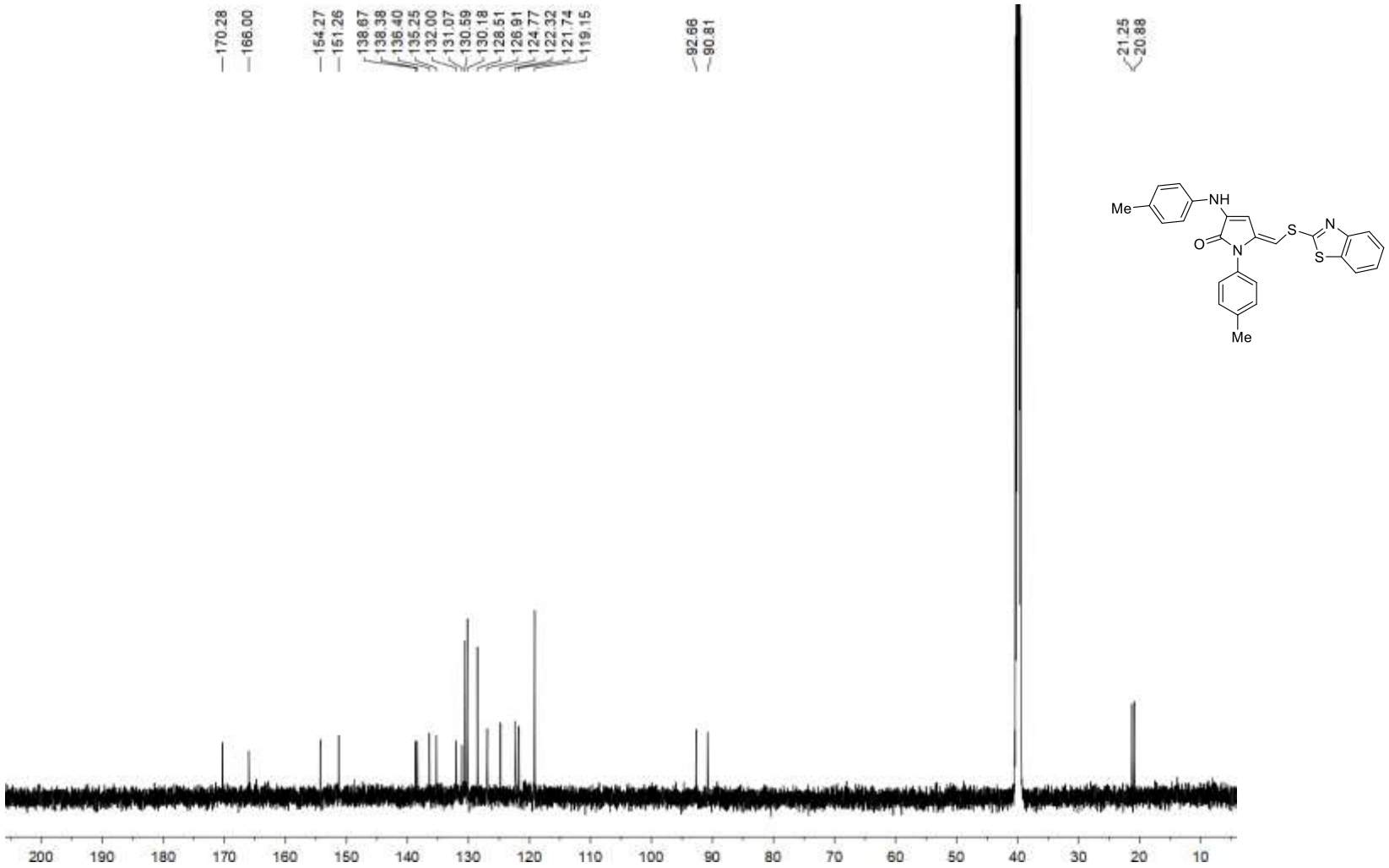


Figure S45. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **4u**

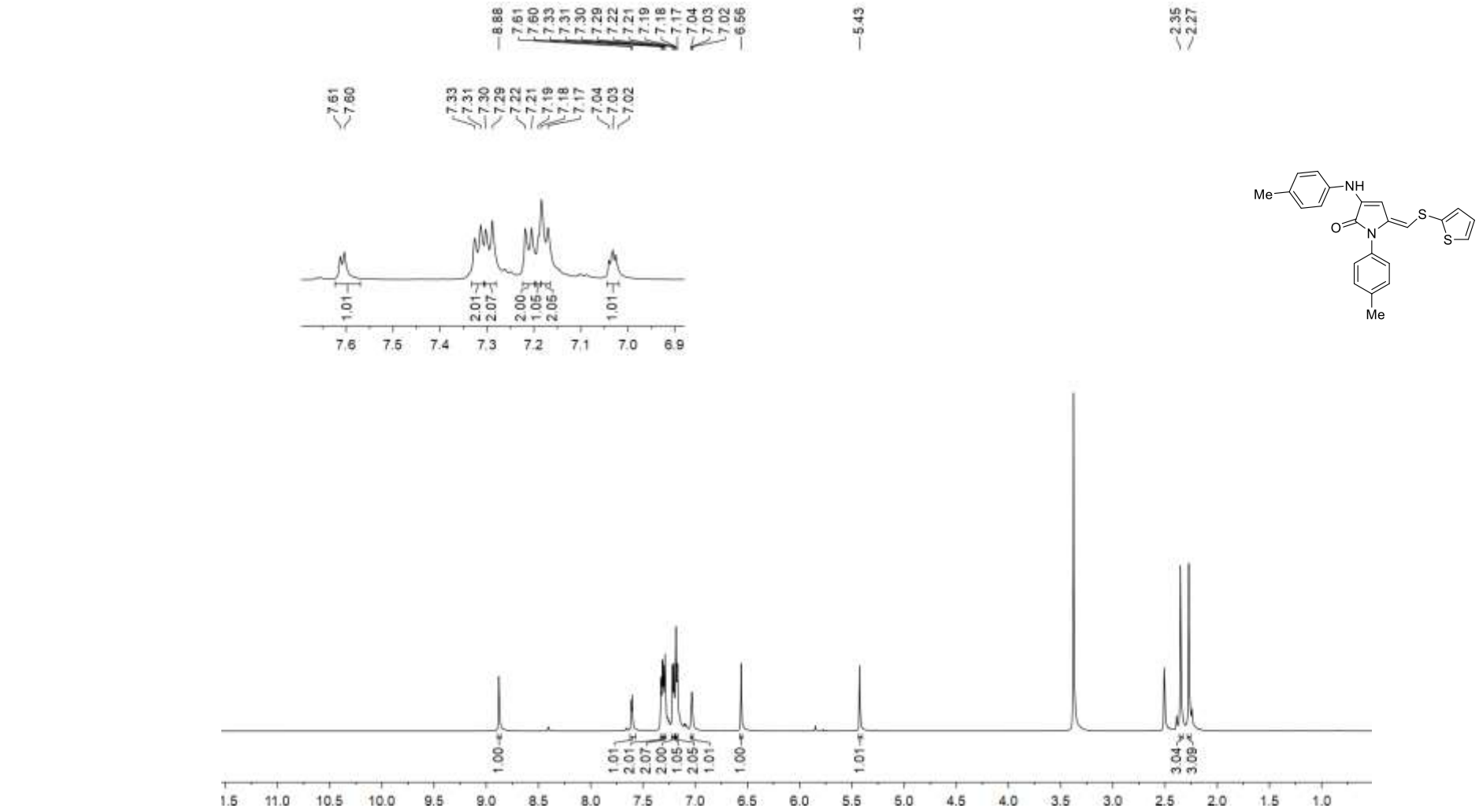


Figure S46. [1]**H NMR** (600 MHz, DMSO-d_6) spectra of compound **4v**

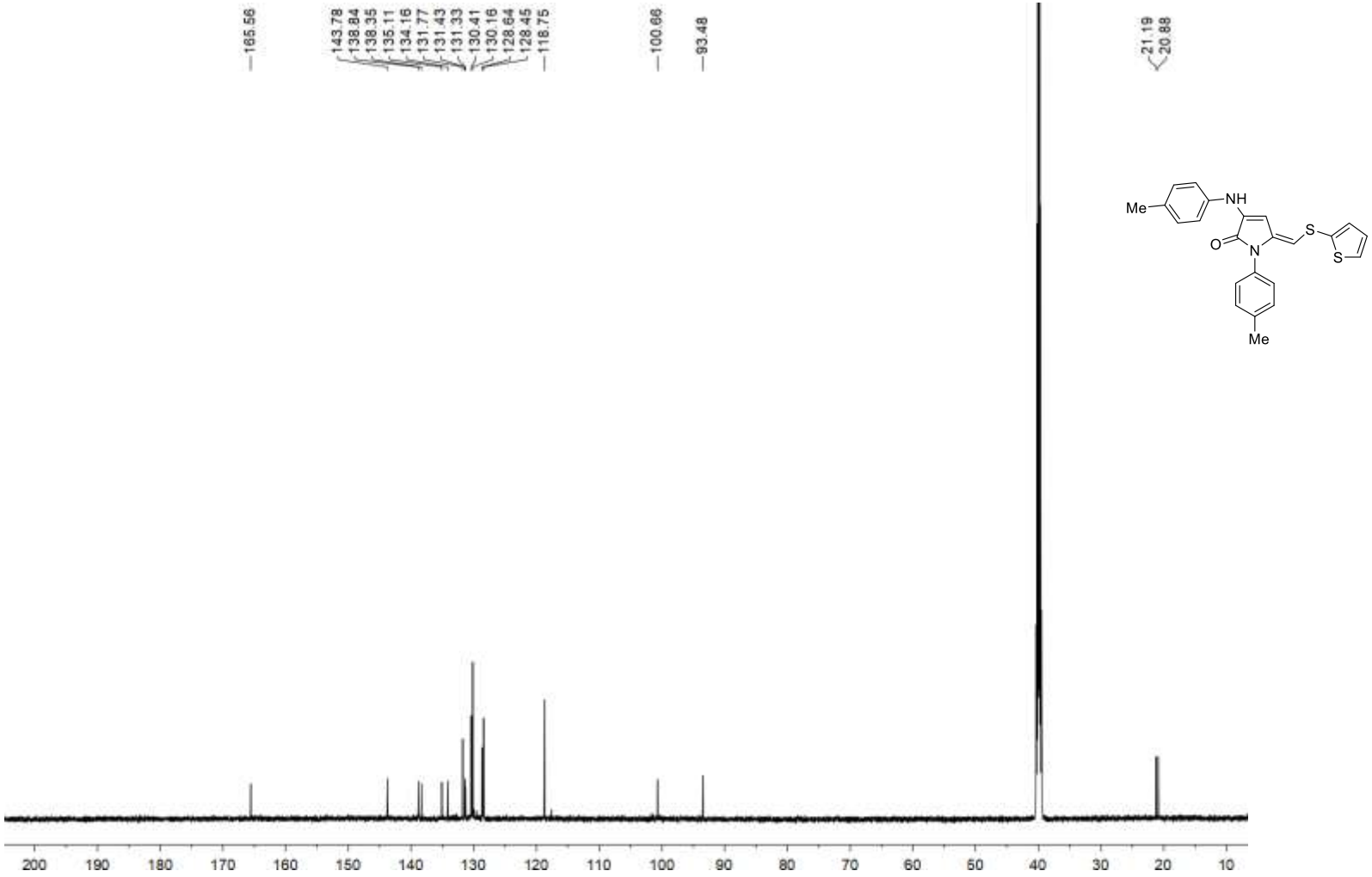


Figure S47. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **4v**

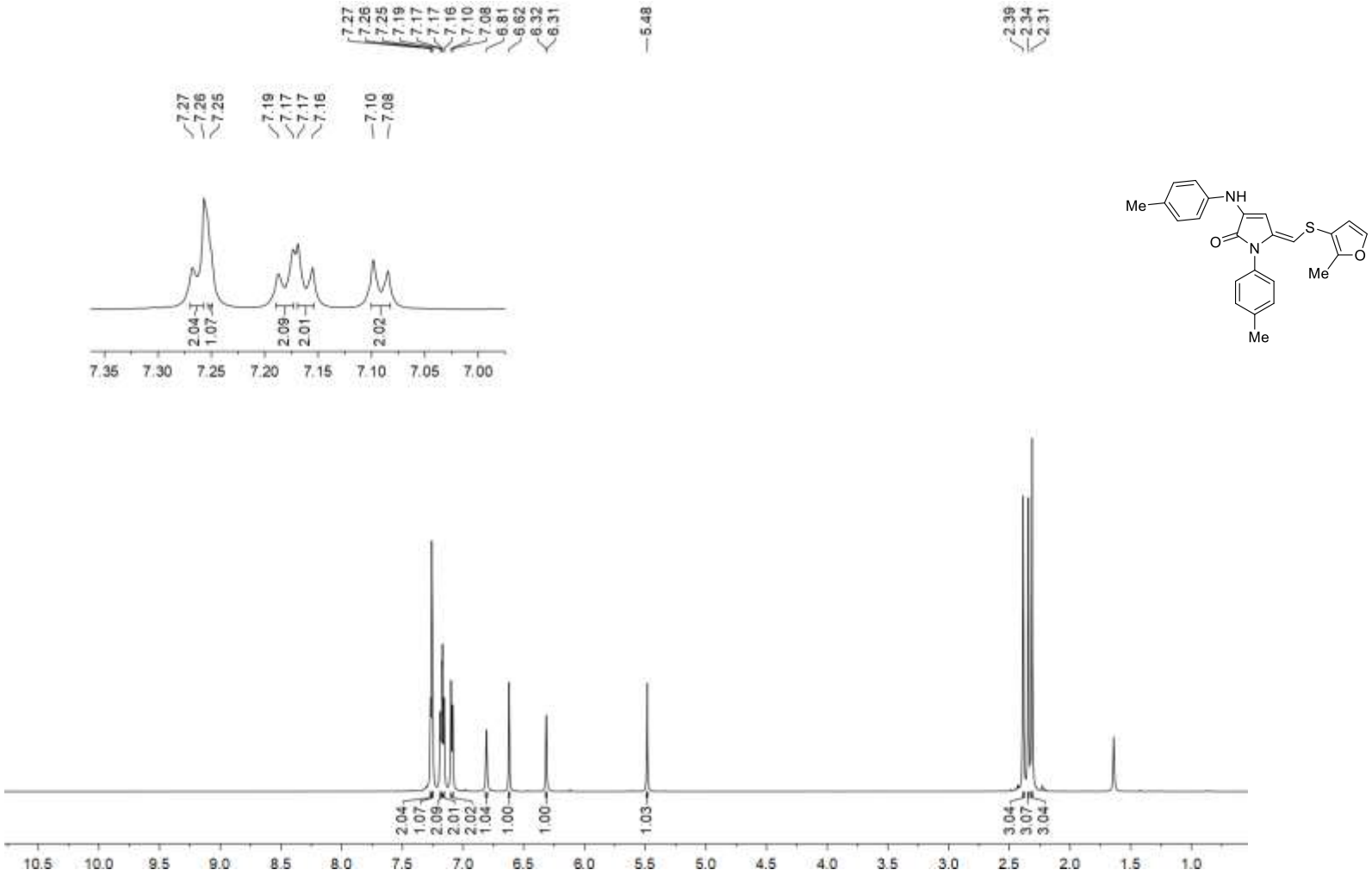


Figure S48. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4w**

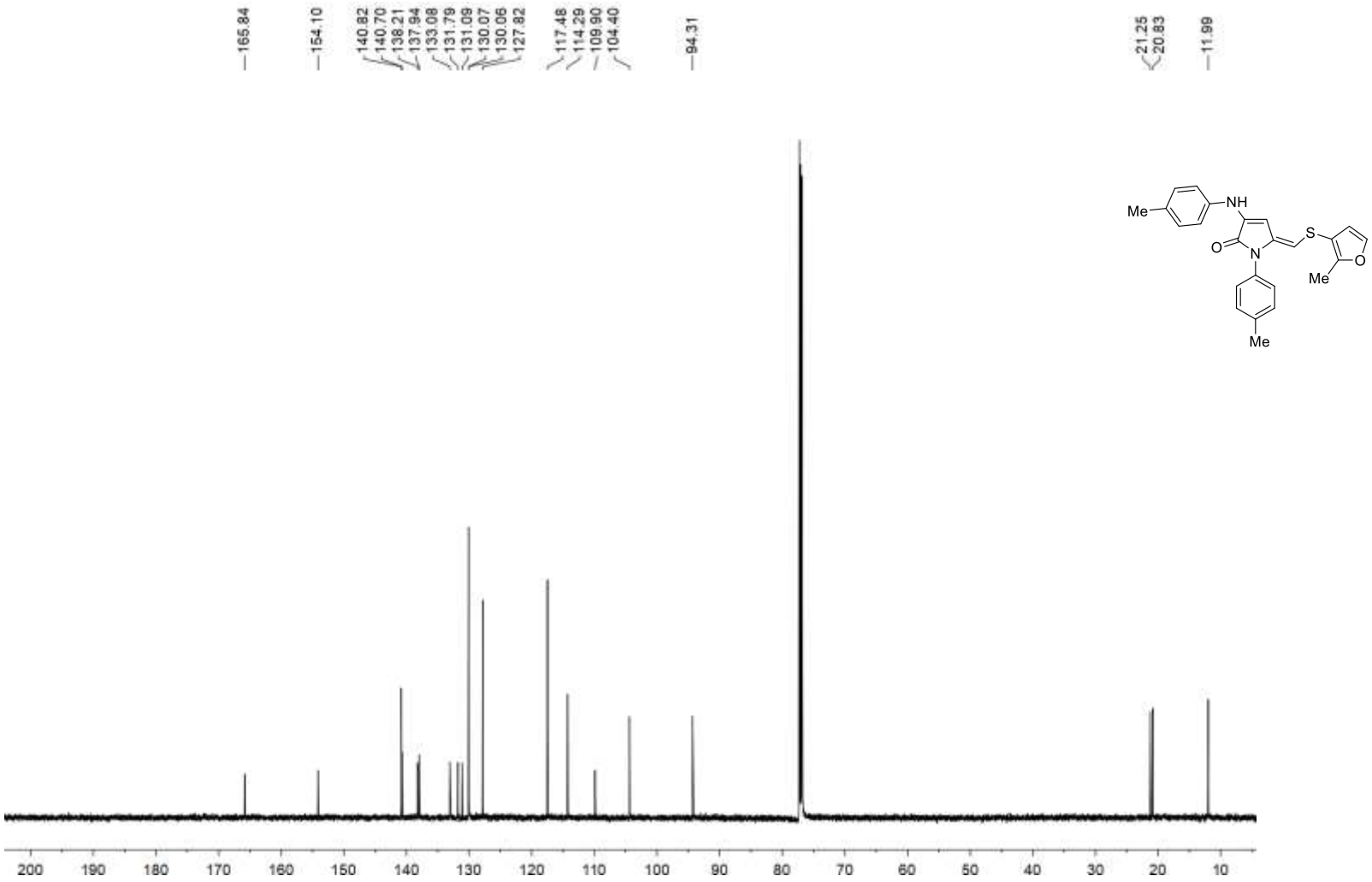


Figure S49. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4w**

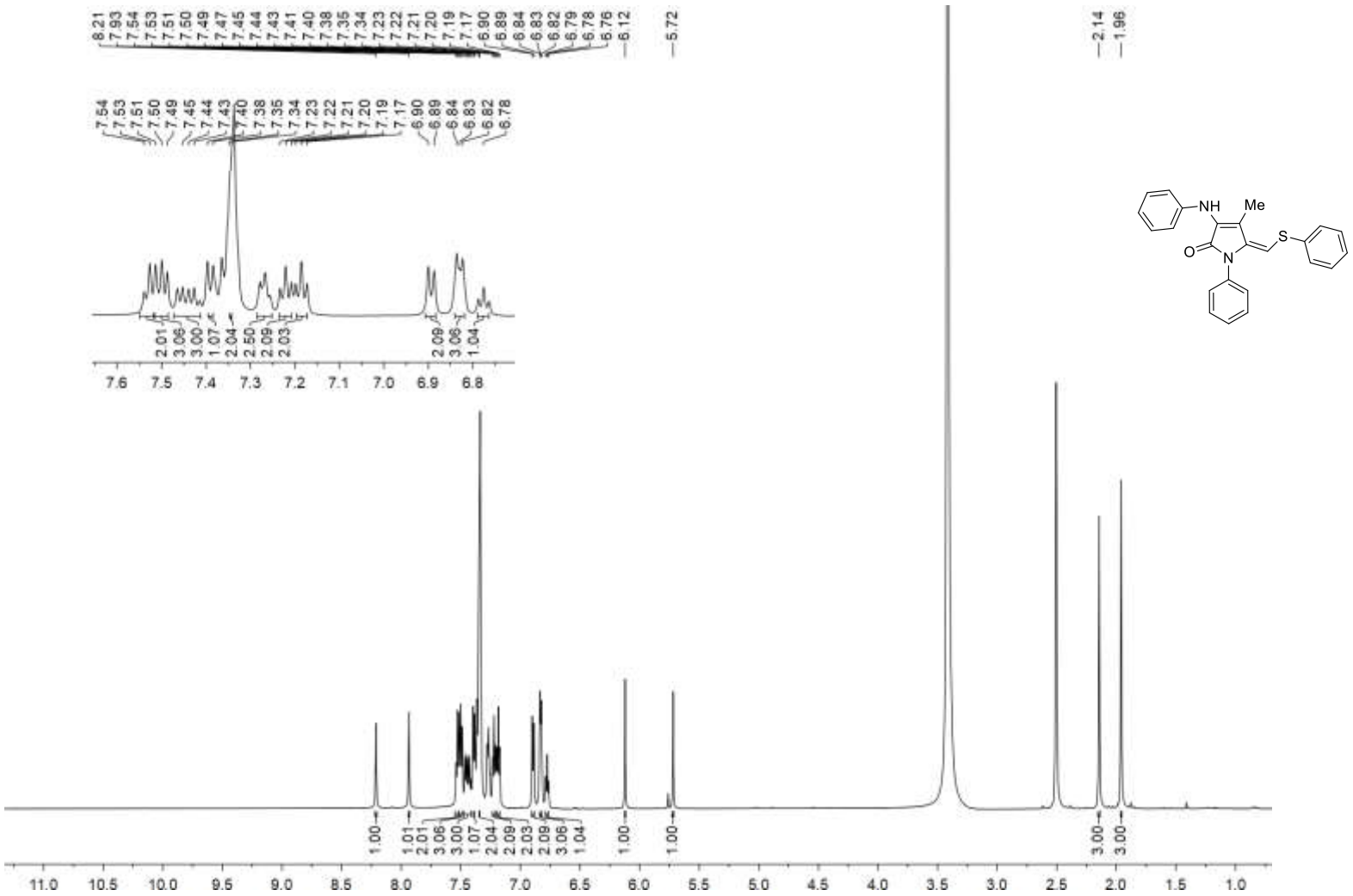


Figure S50. [1]**H NMR** (600 MHz, DMSO-d_6) spectra of compound **4x**

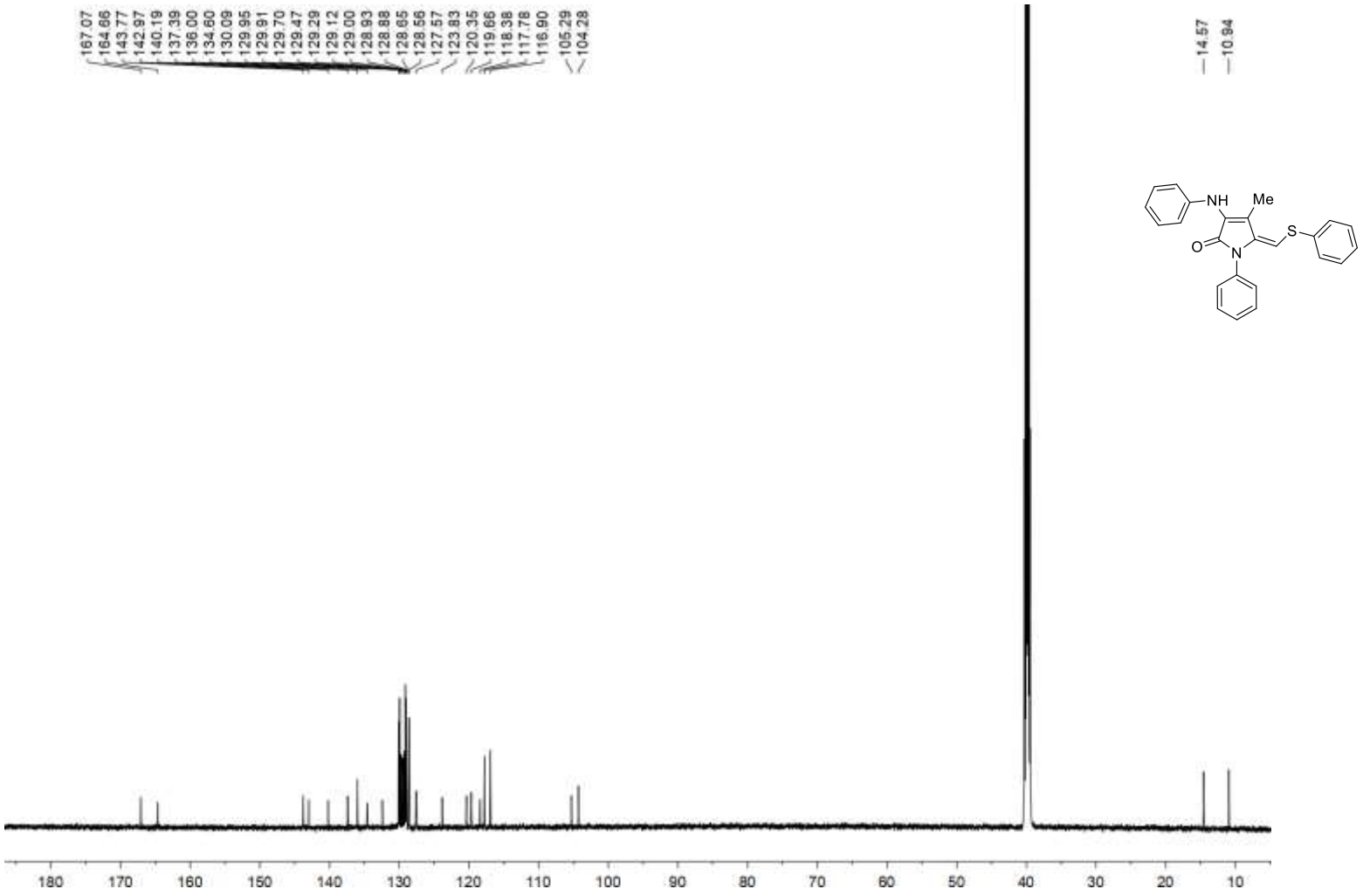


Figure S51. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **4x**

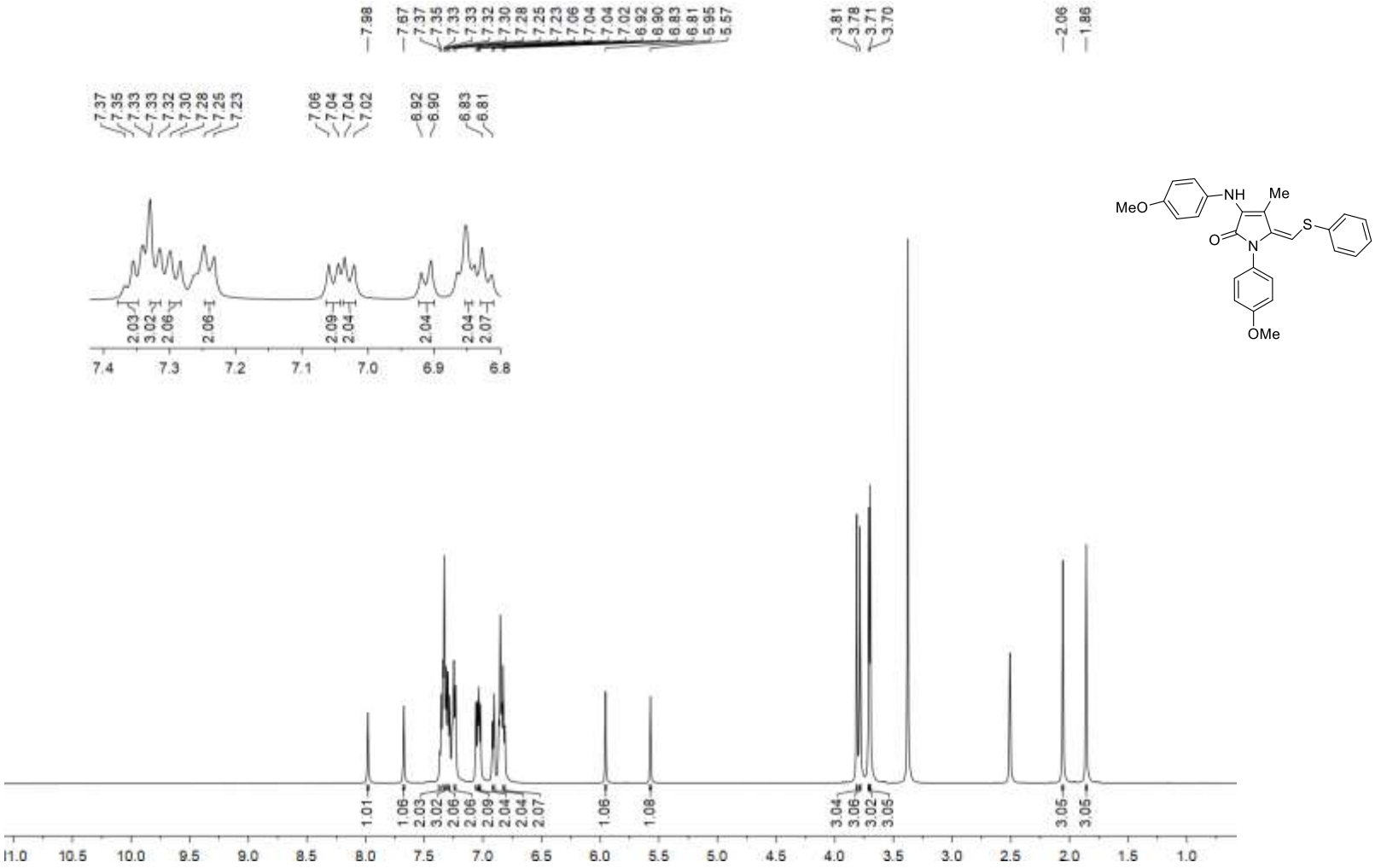


Figure S52. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **4y**

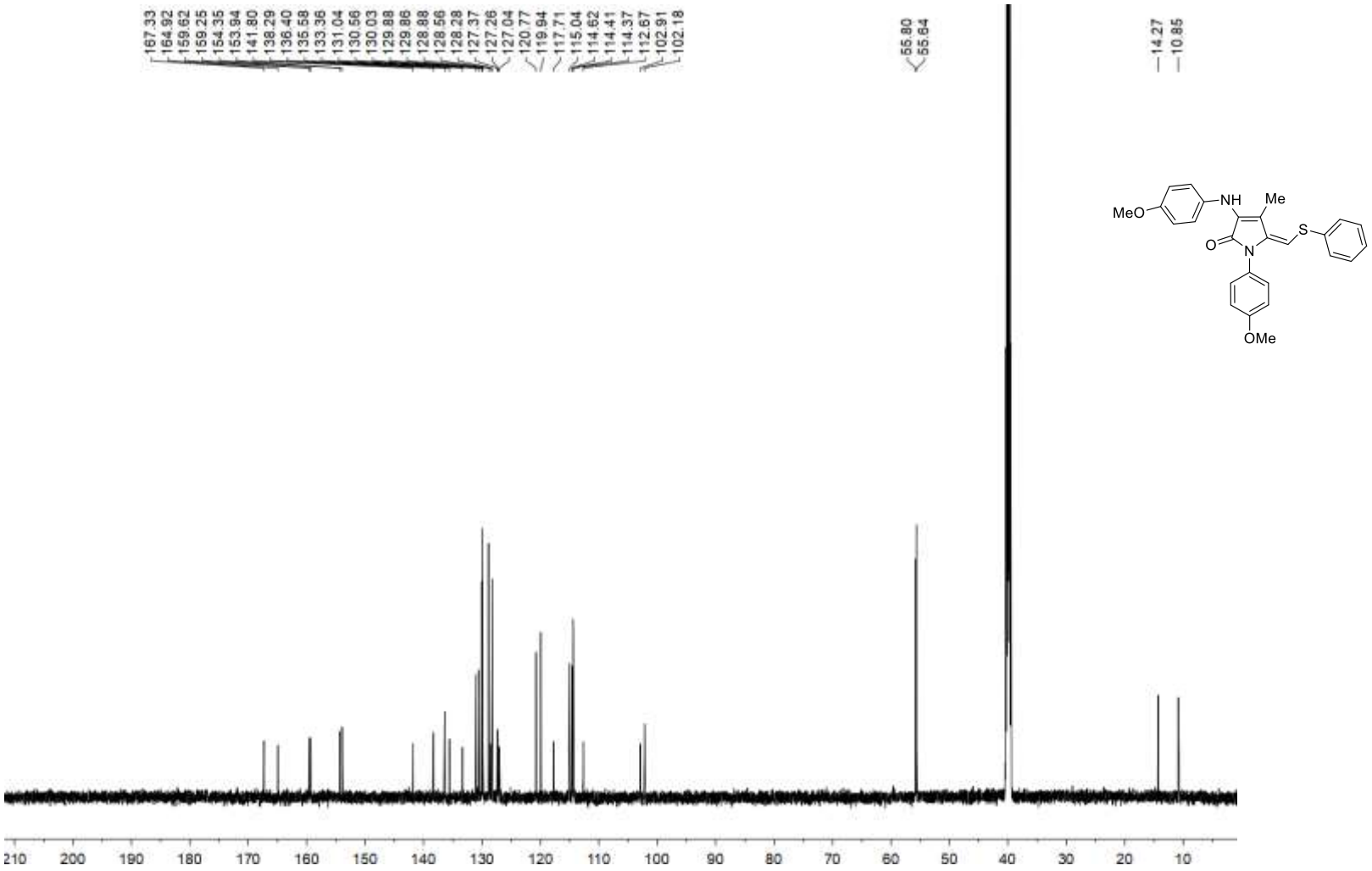


Figure S53. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **4y**

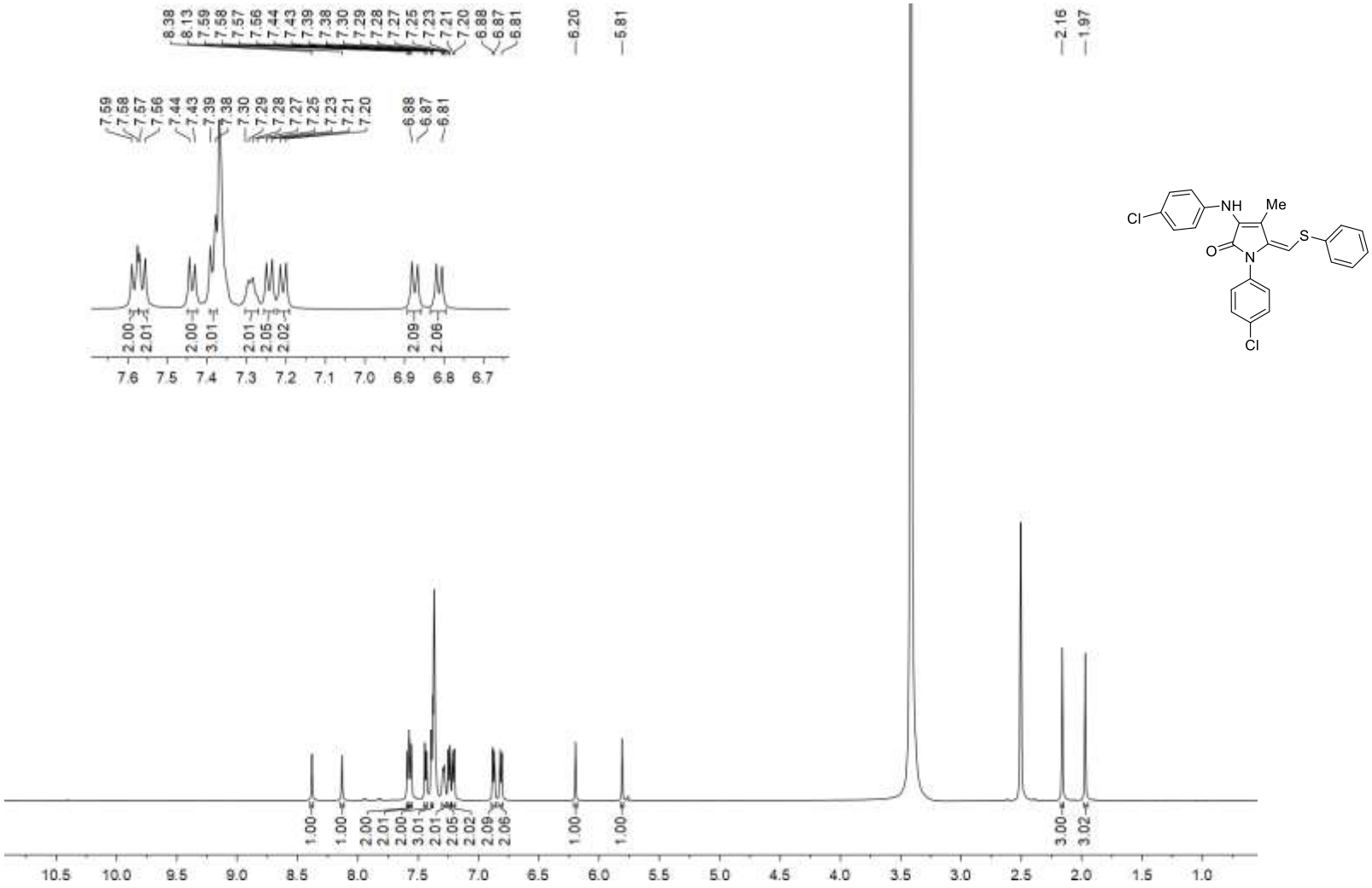


Figure S54. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **4z**

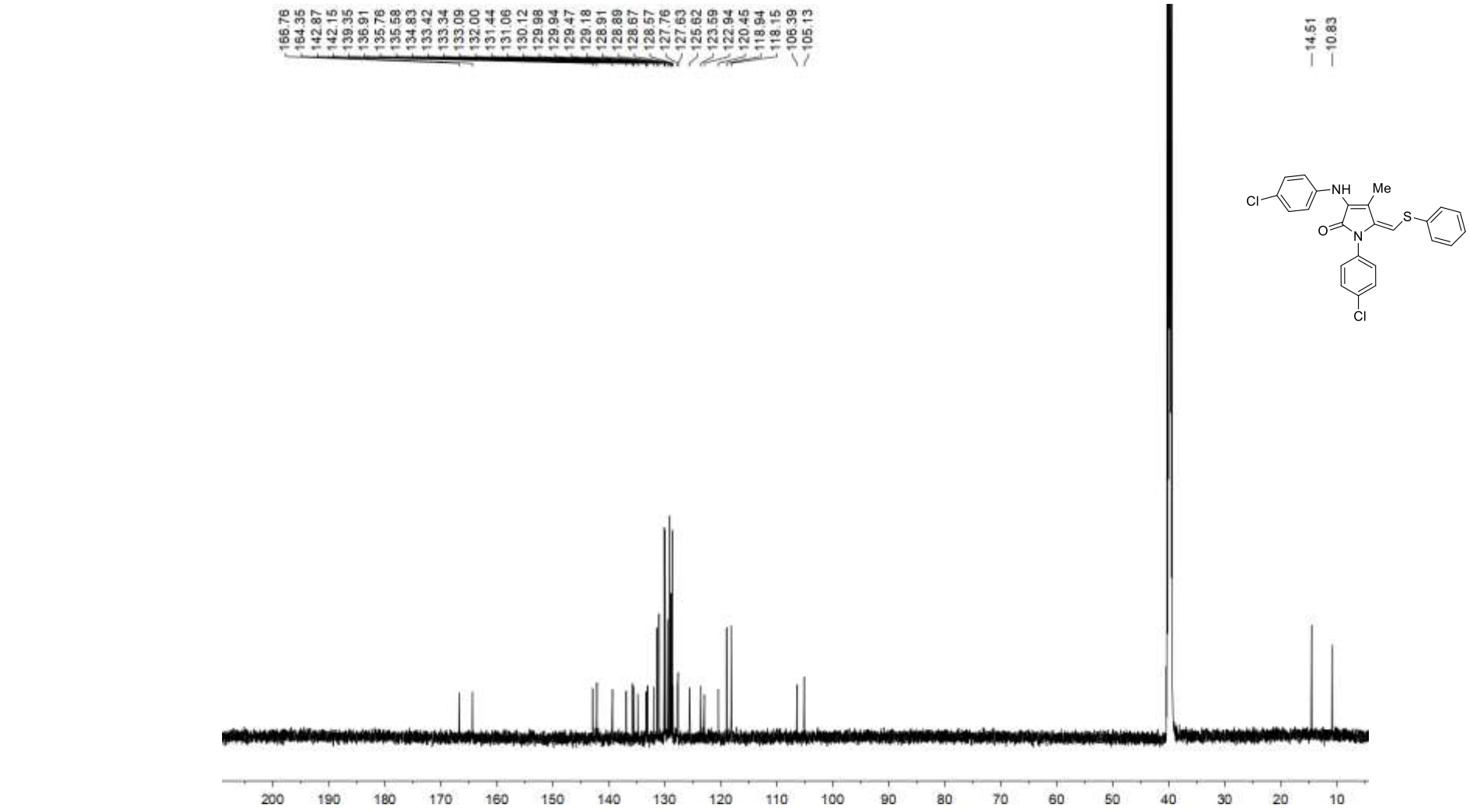


Figure S55. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **4z**

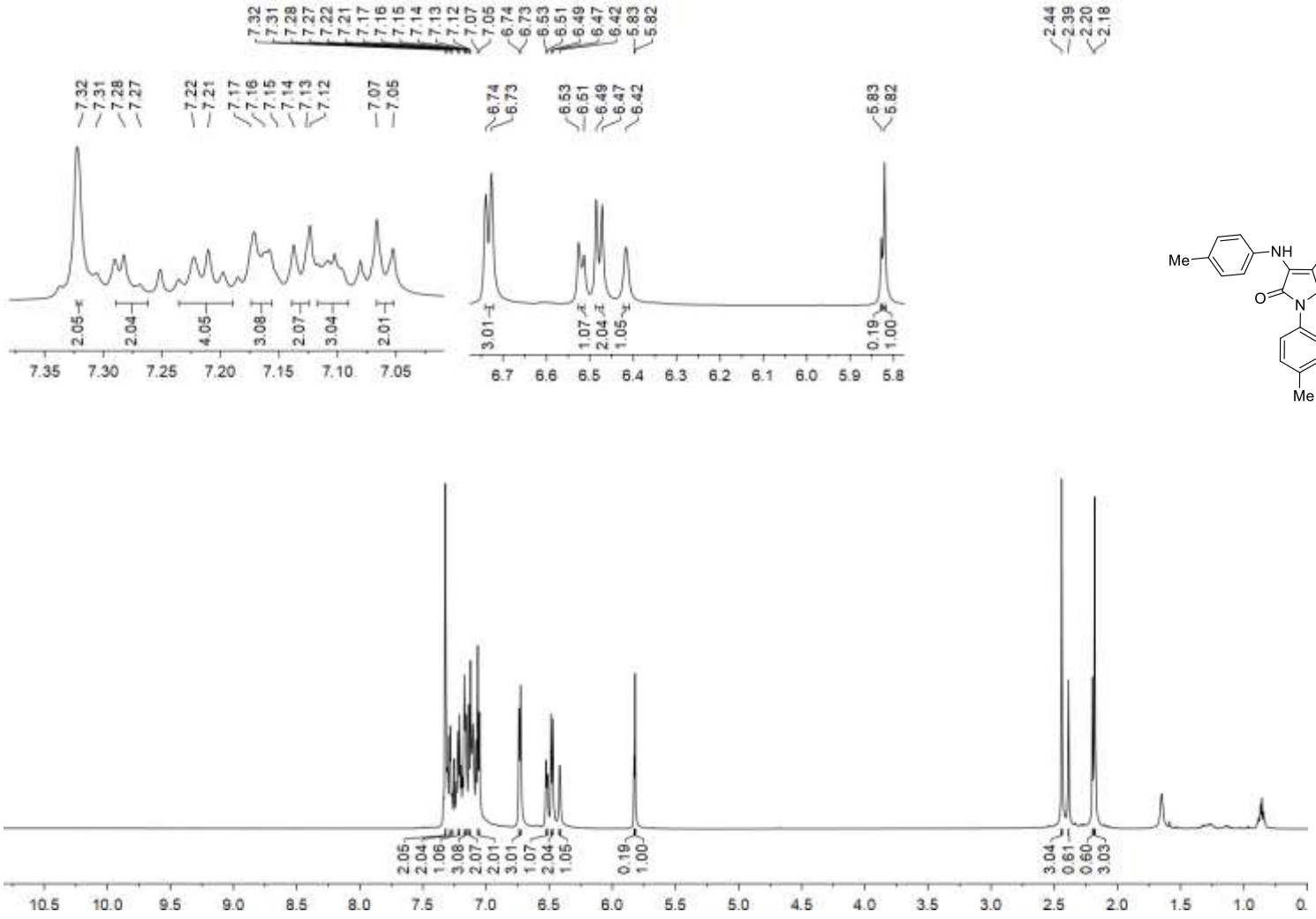


Figure S56. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4a'**

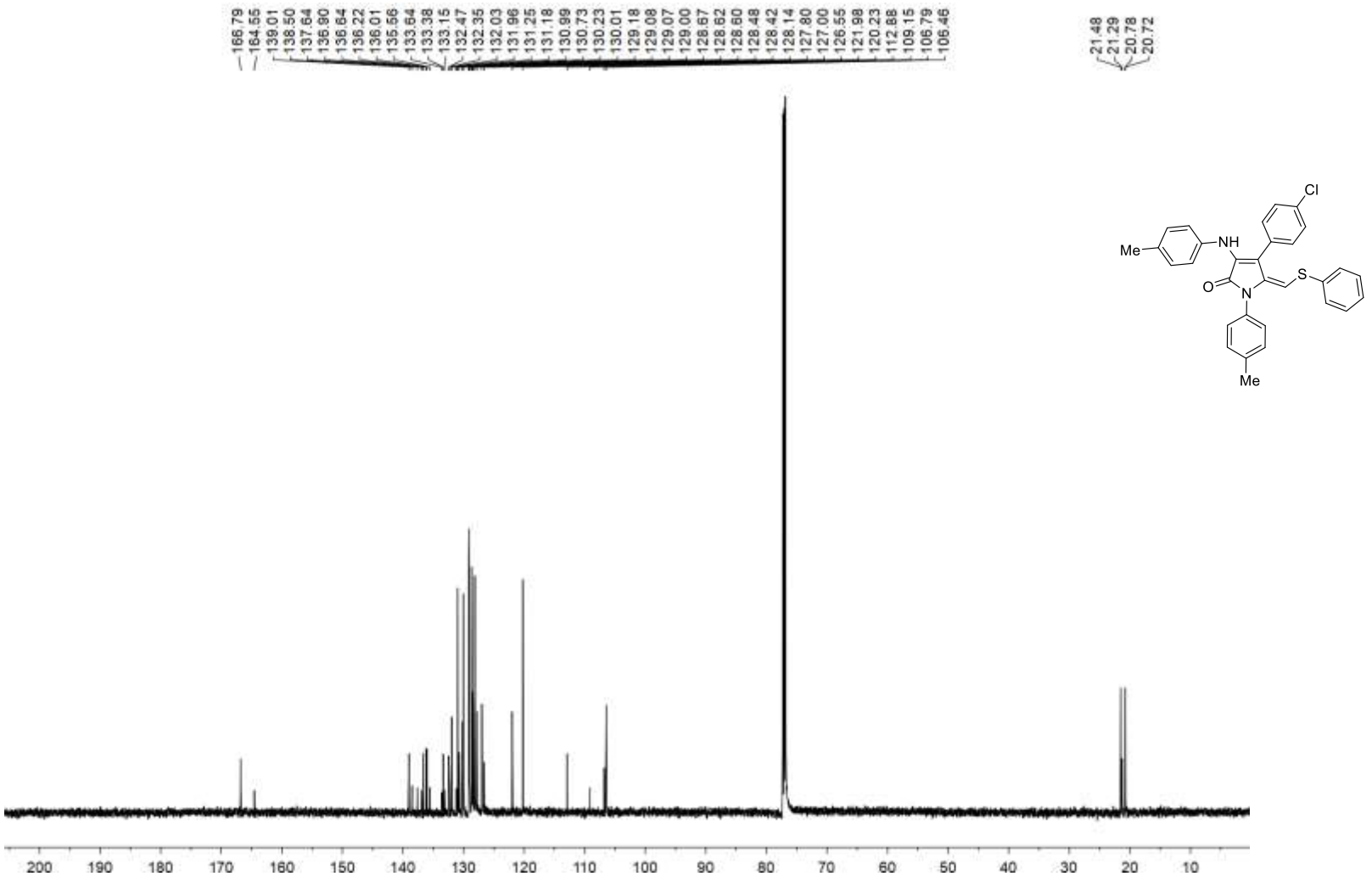


Figure S57. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4a'**

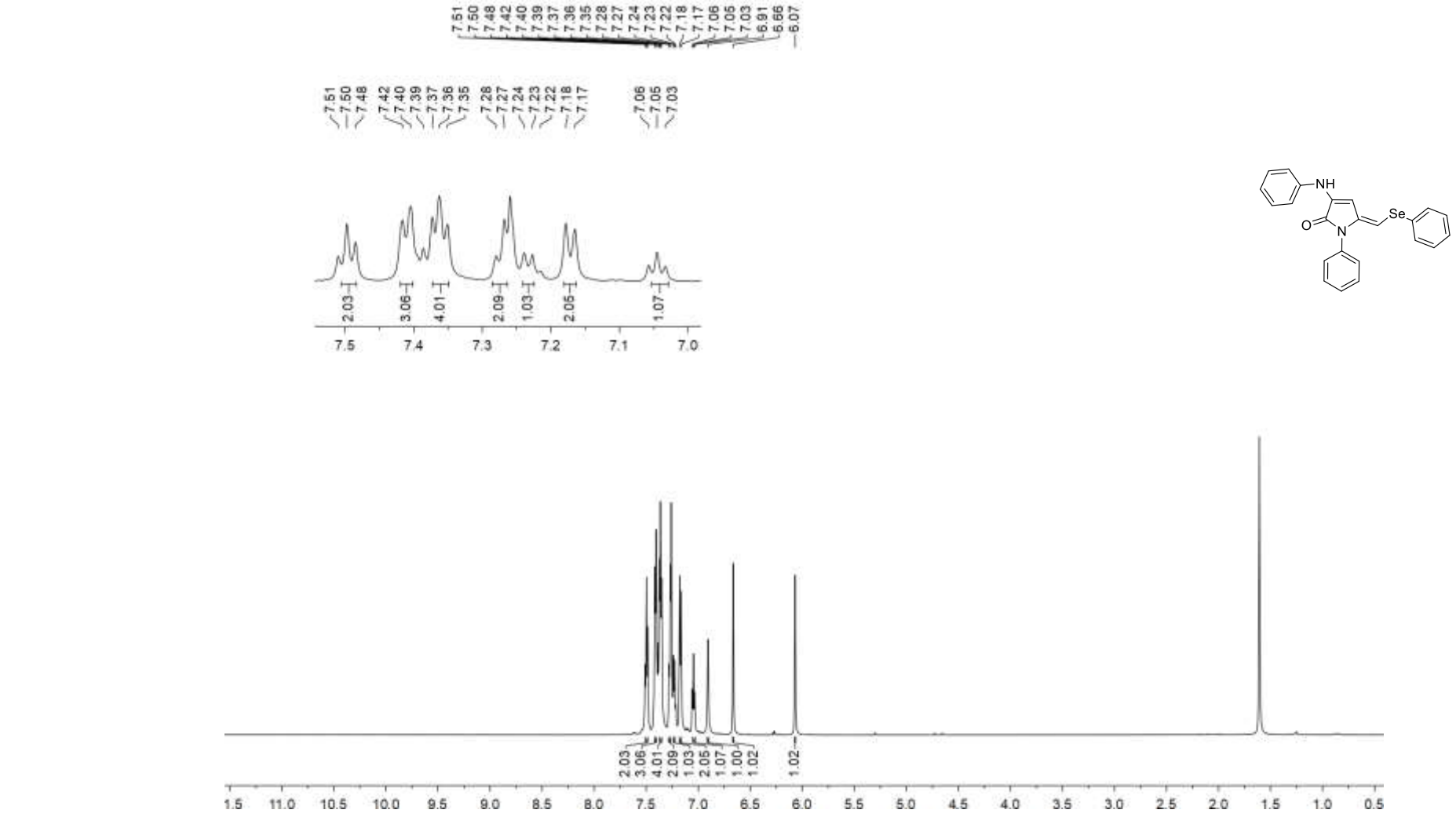


Figure S58. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **6a**

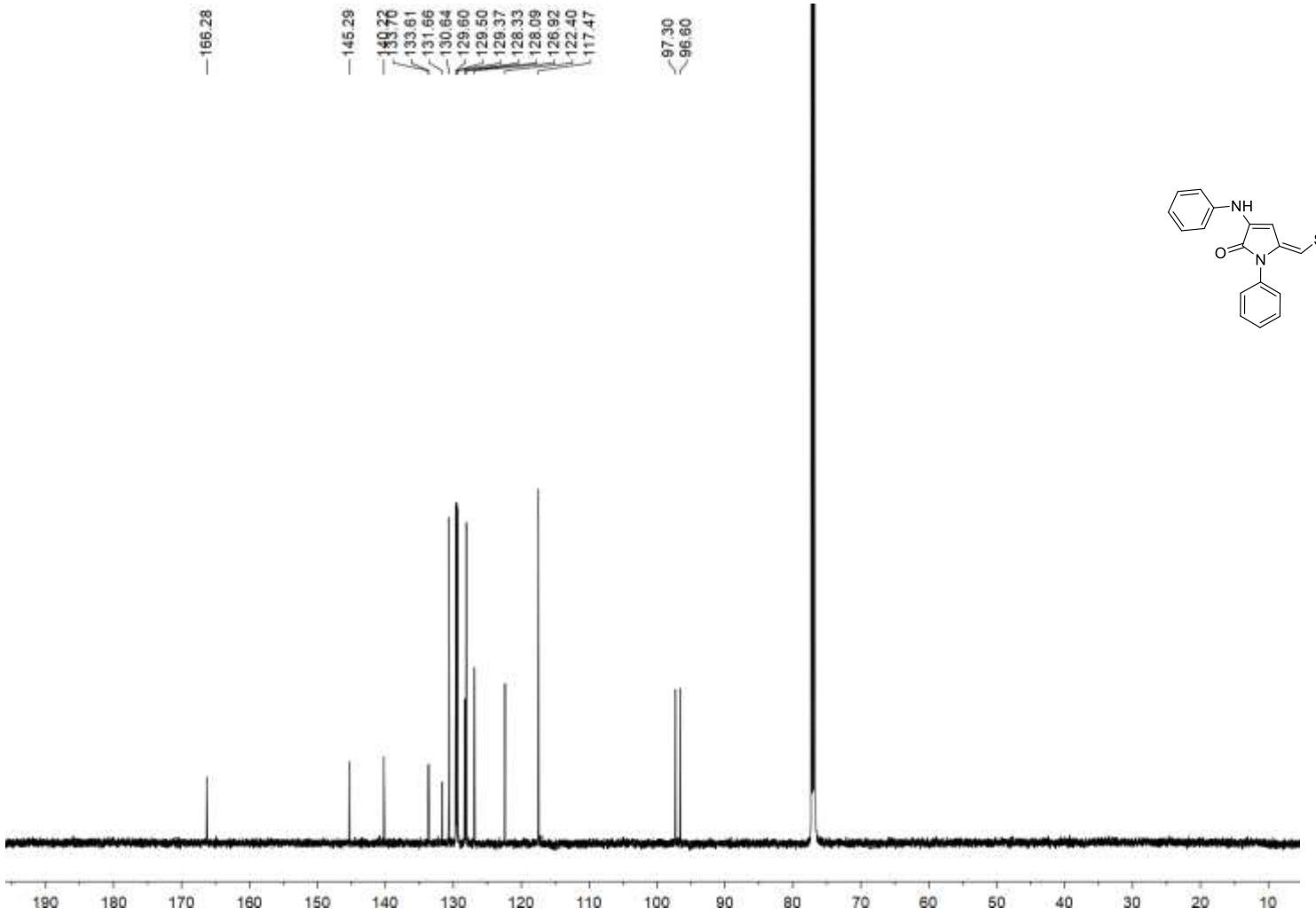


Figure S59. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **6a**

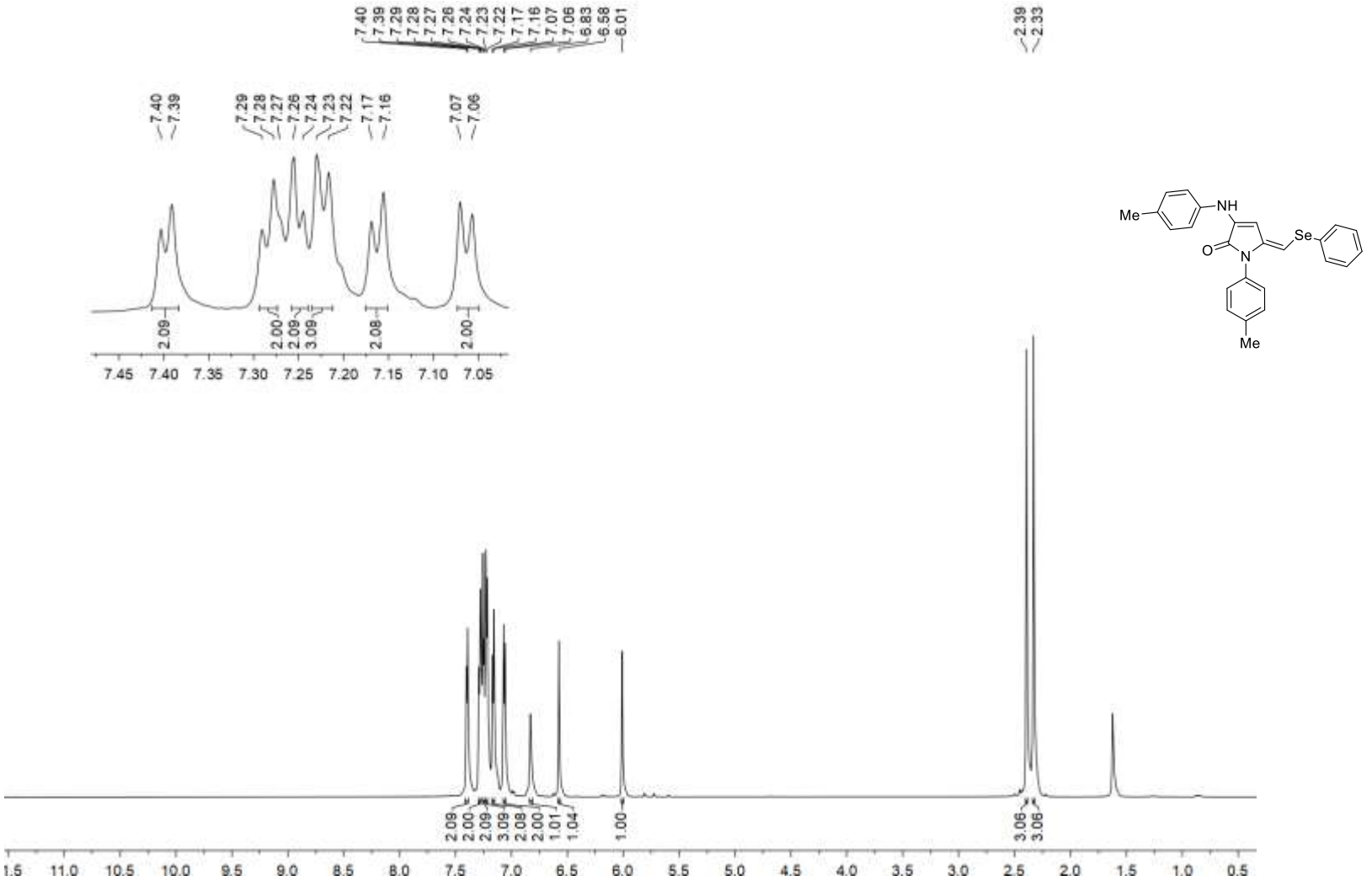


Figure S60. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **6b**

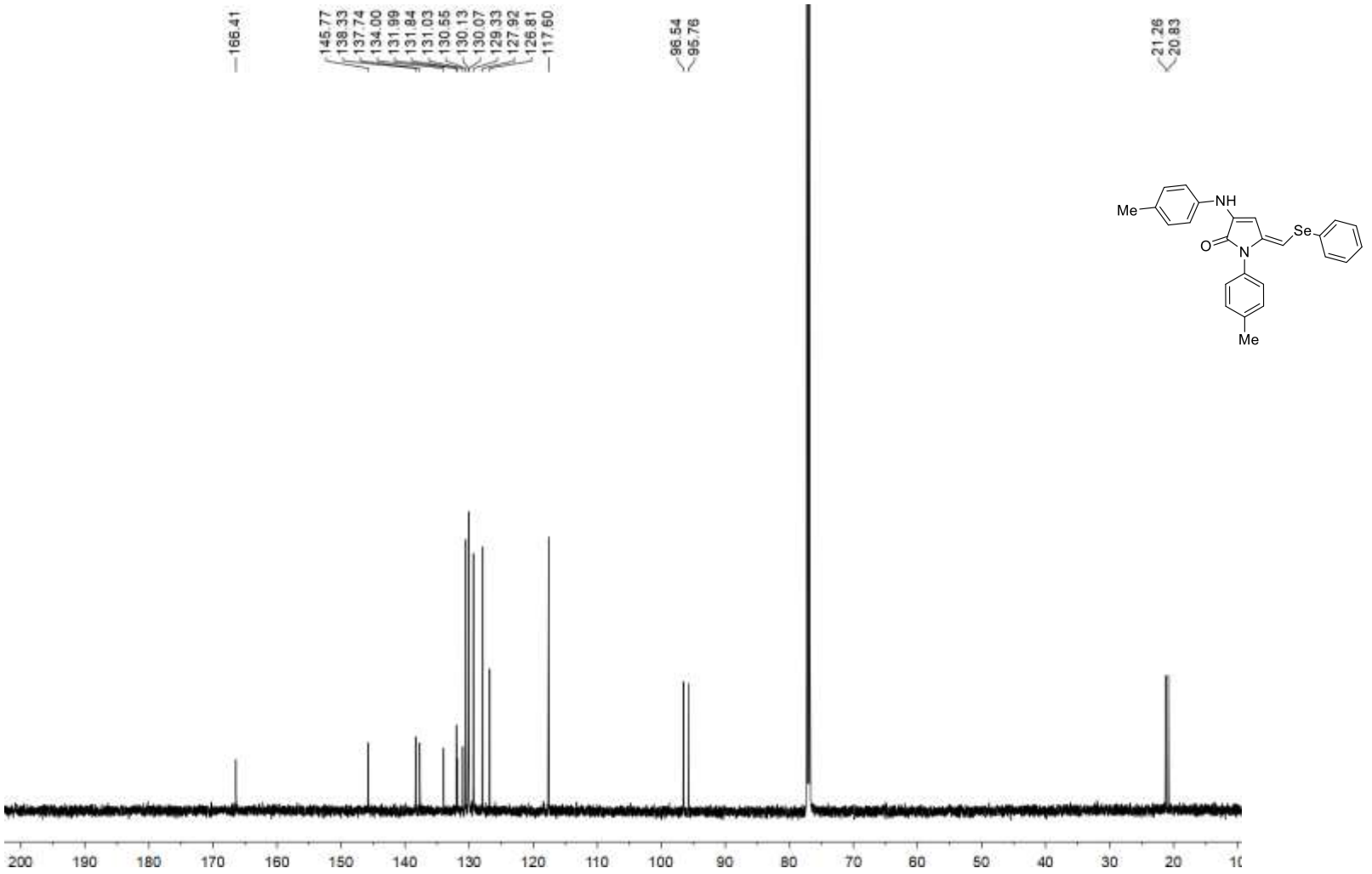


Figure S61. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **6b**

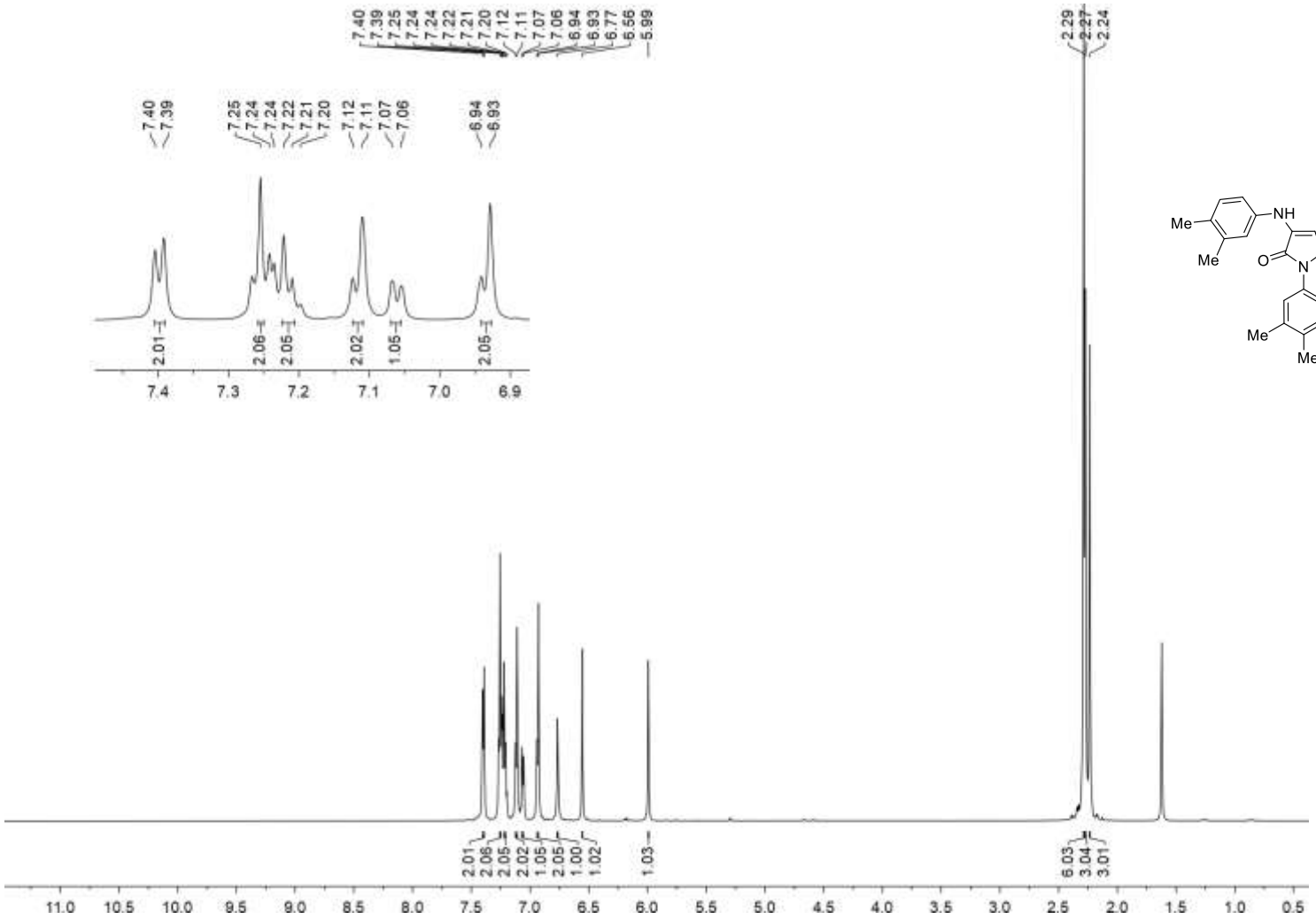


Figure S62. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **6c**

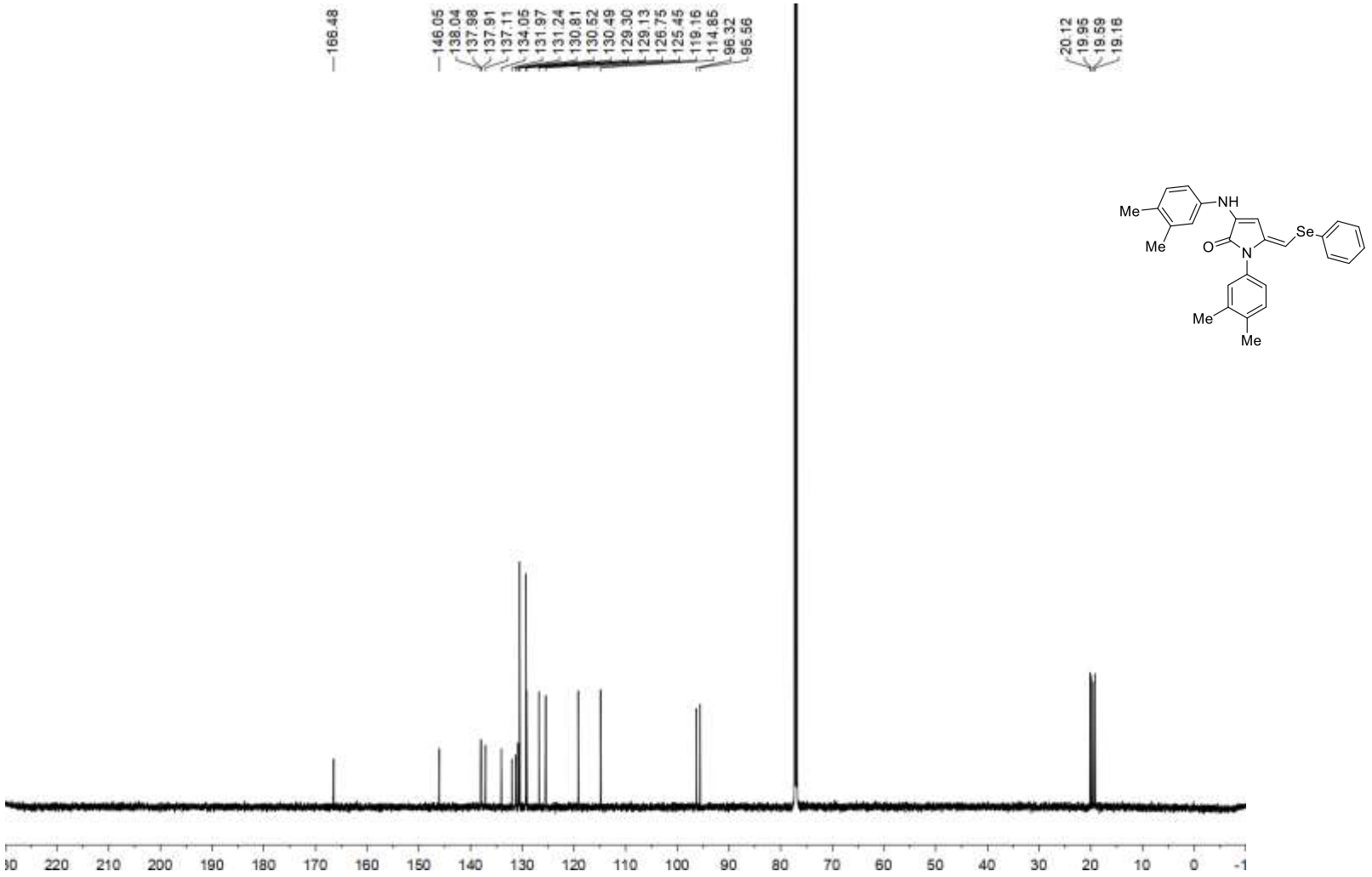


Figure S63. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **6c**

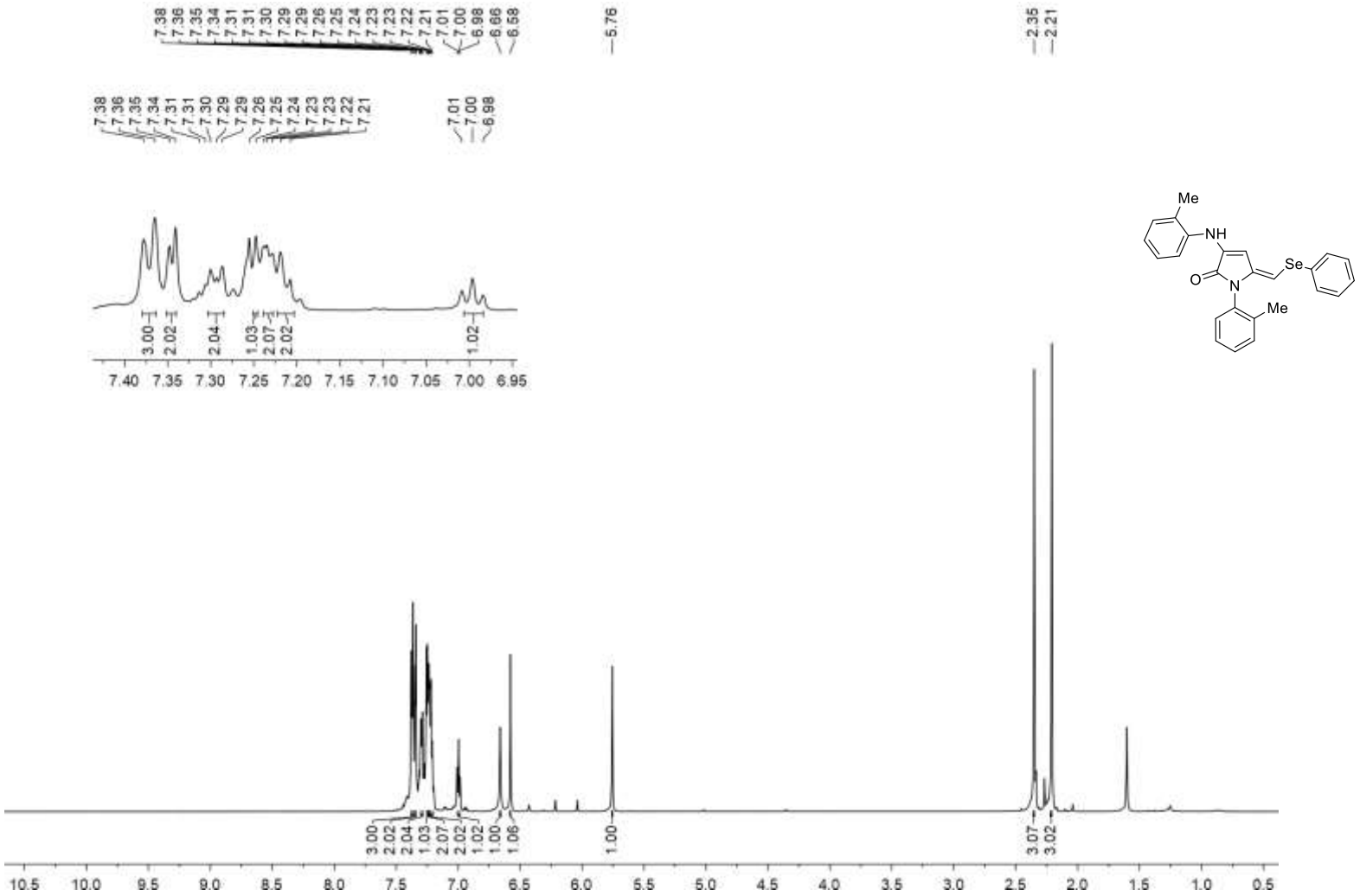


Figure S64. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **6d**

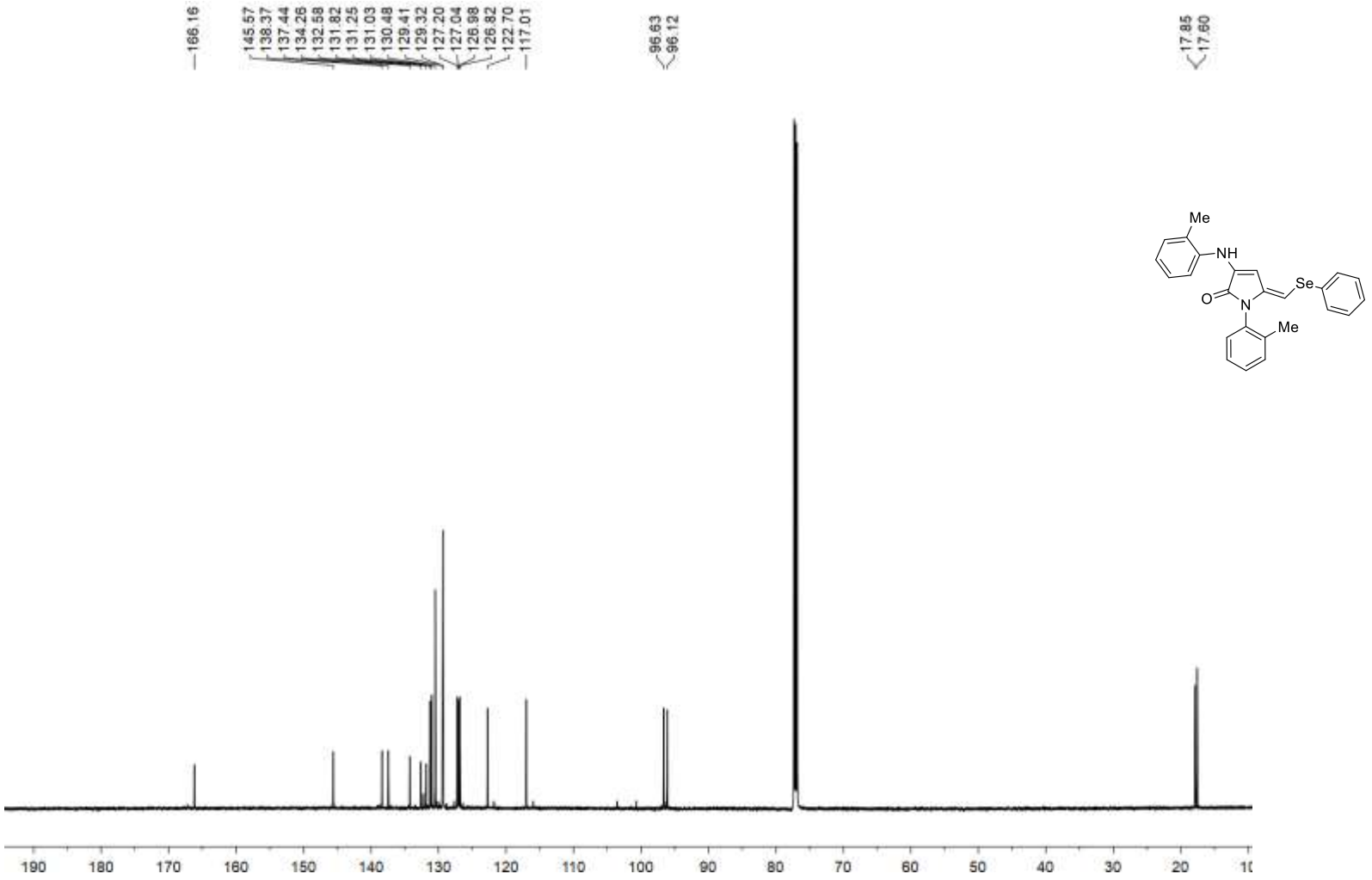


Figure S65. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **6d**

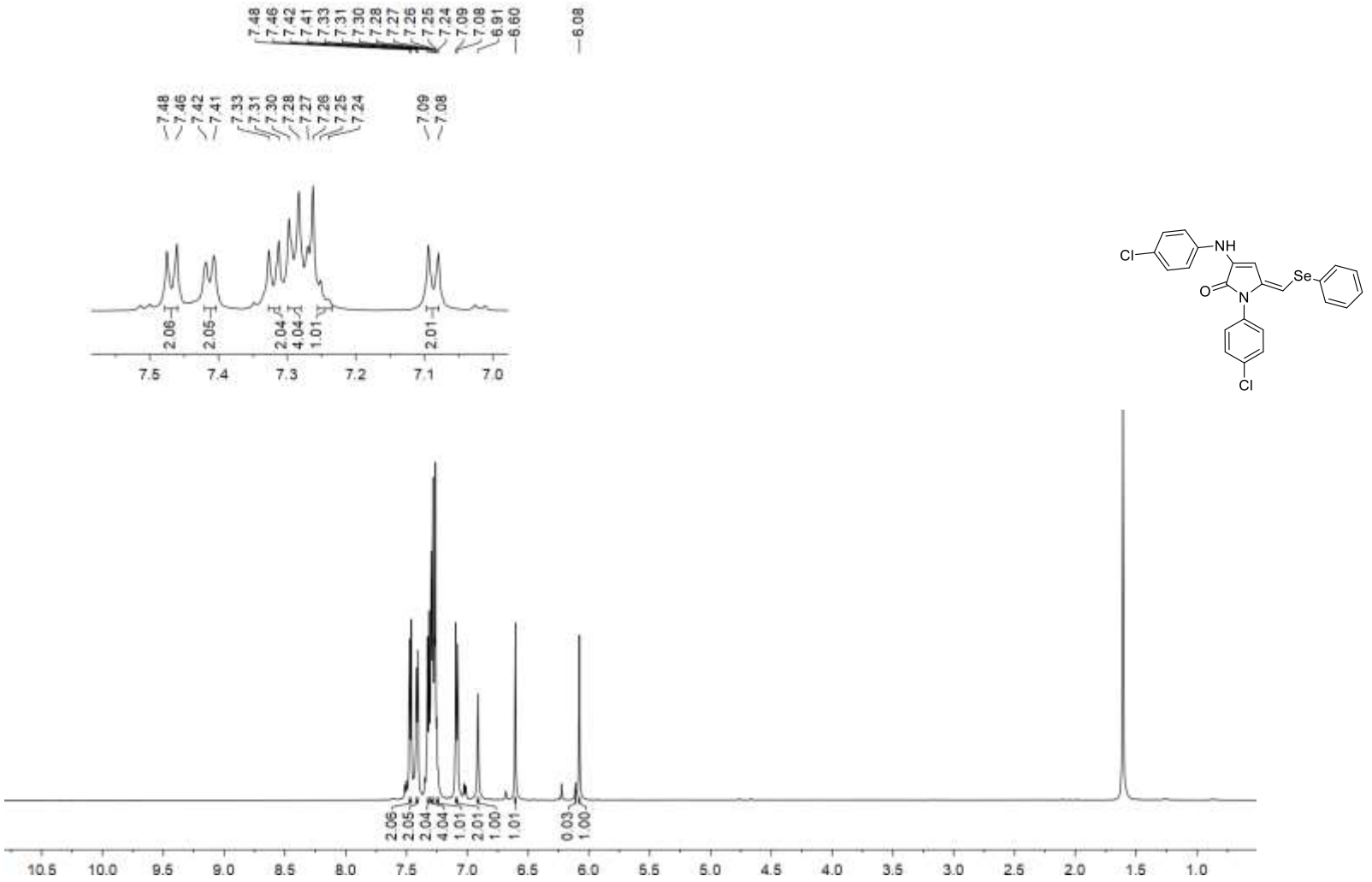


Figure S66. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **6e**

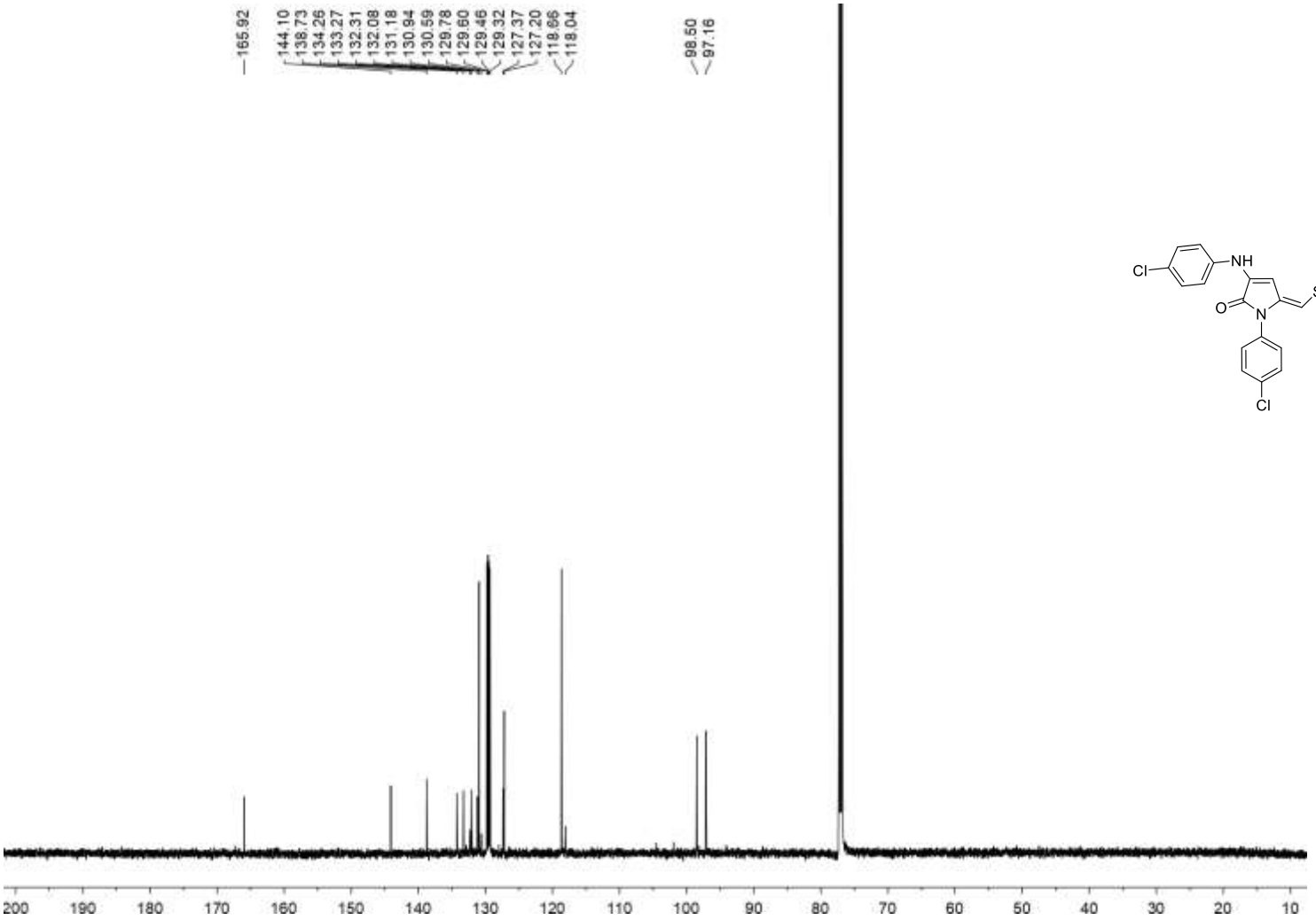


Figure S67. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **6e**

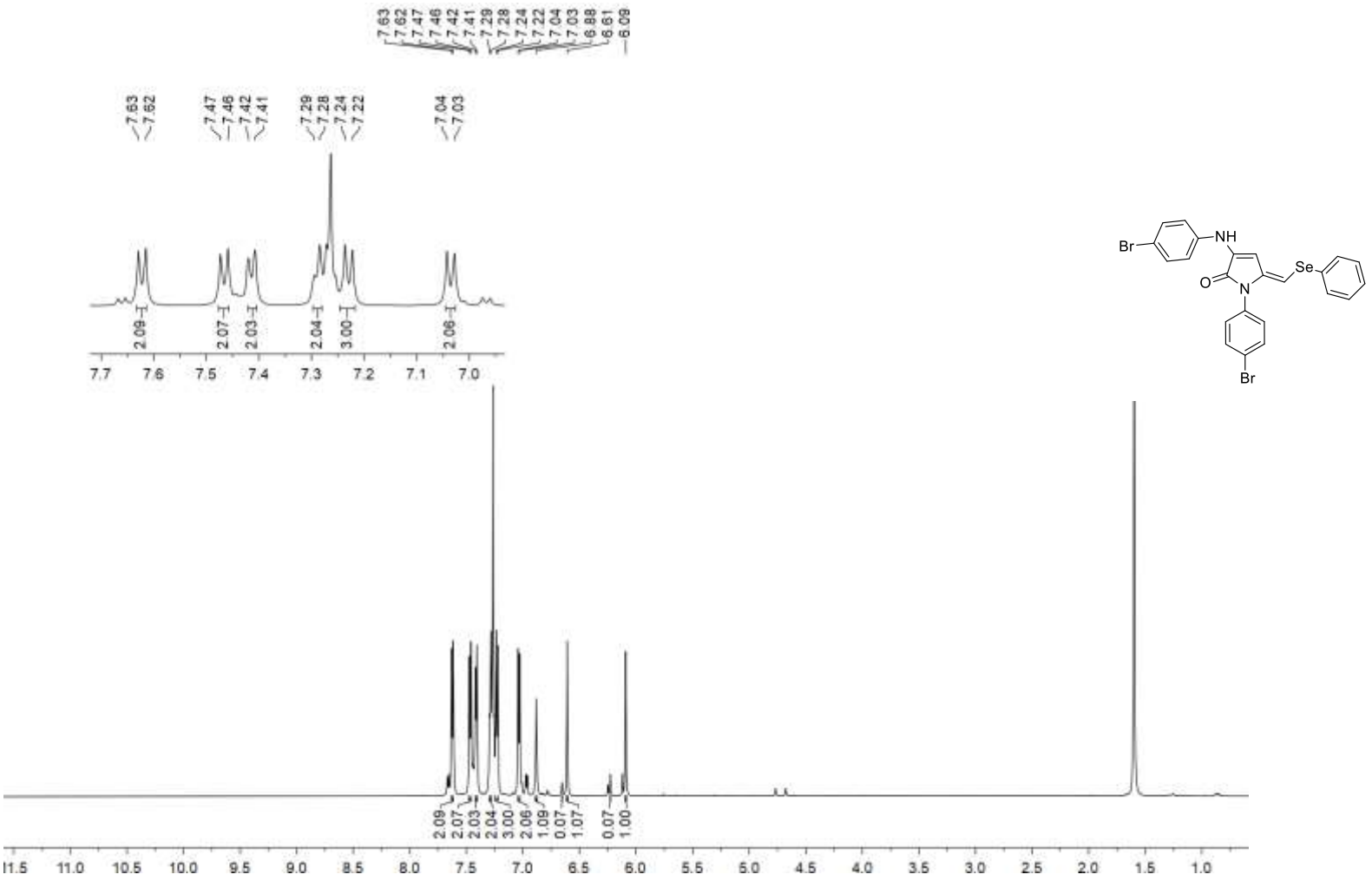


Figure S68. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **6f**

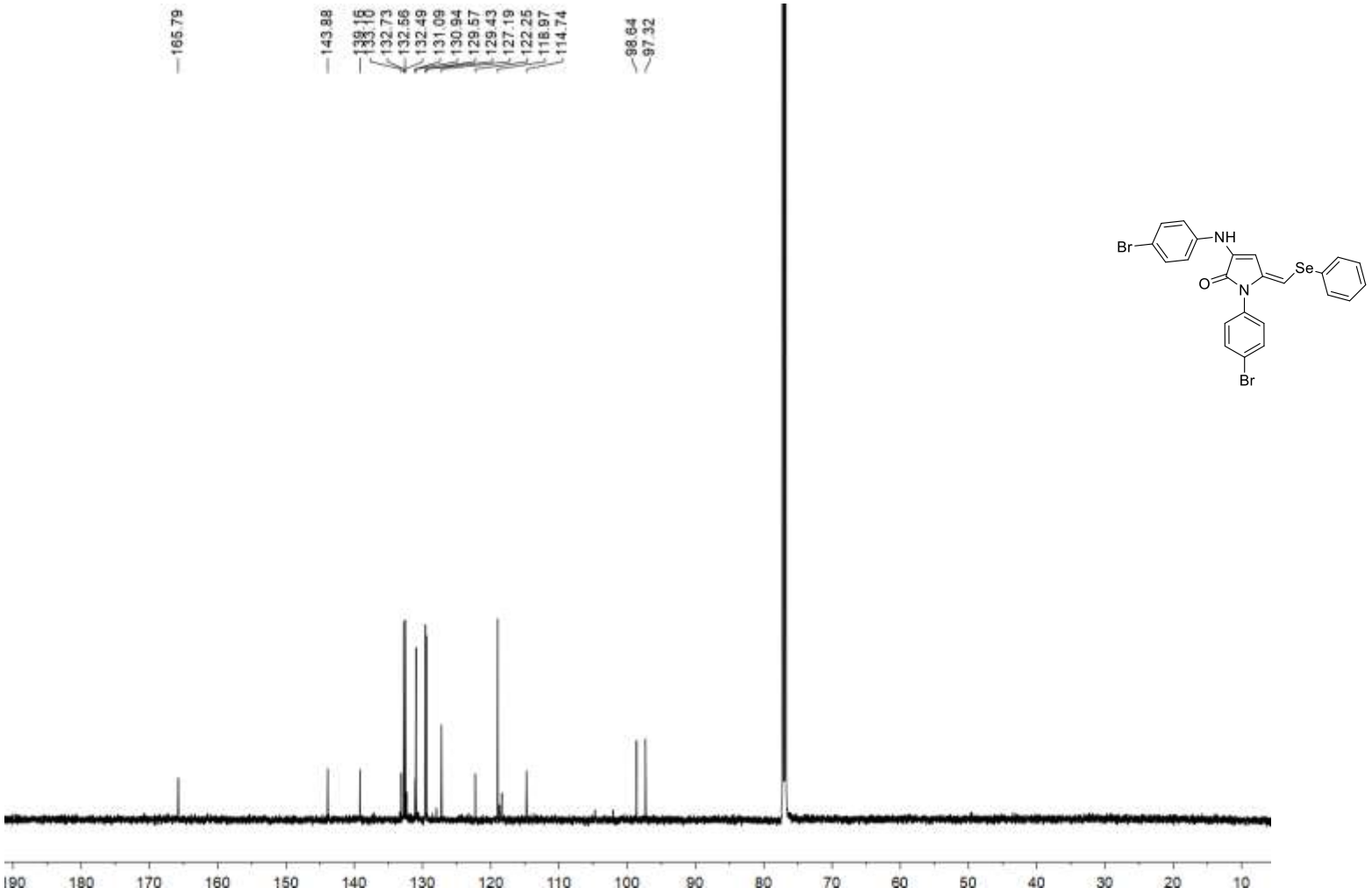


Figure S69. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **6f**

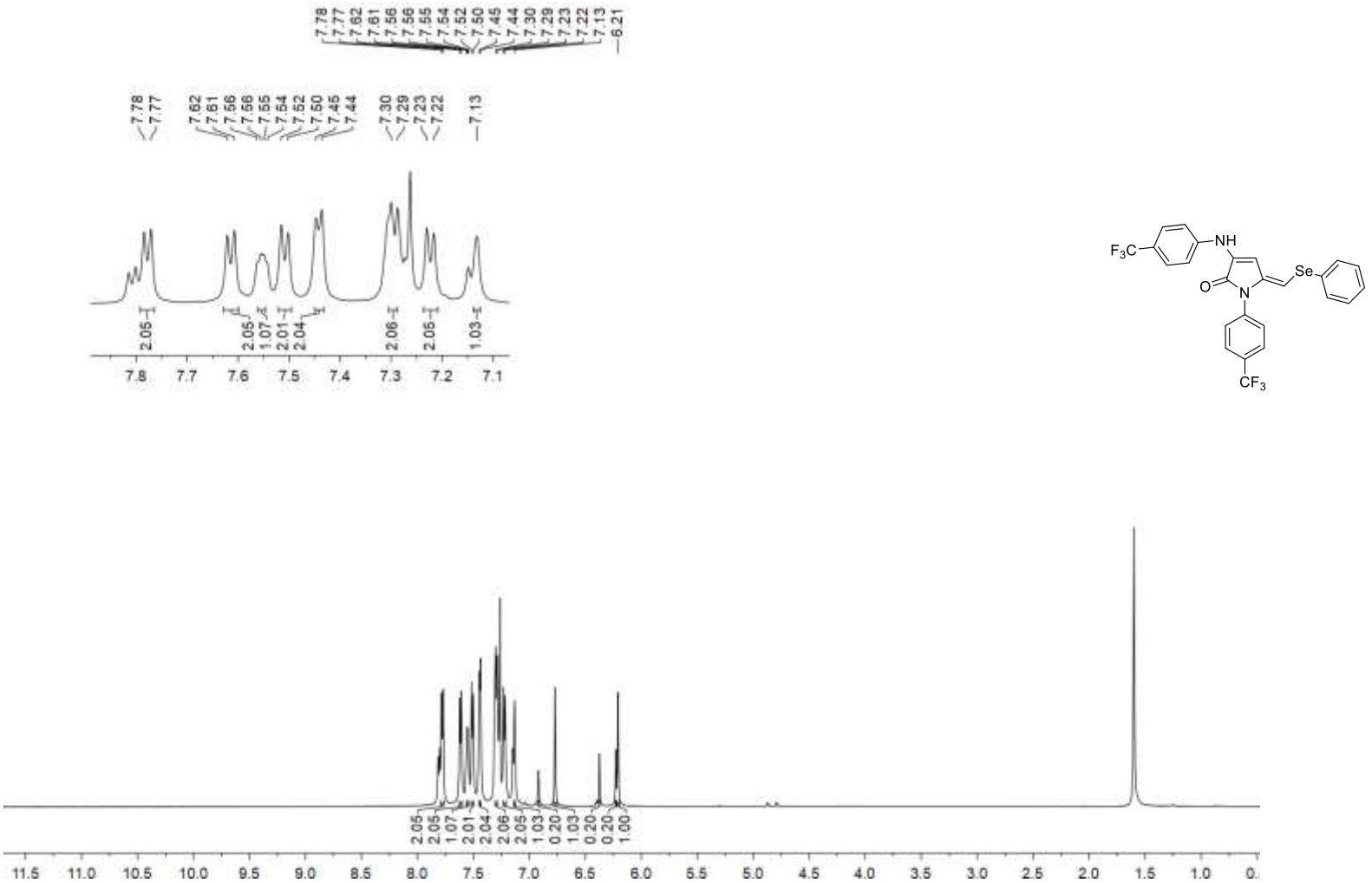


Figure S70. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **6g**

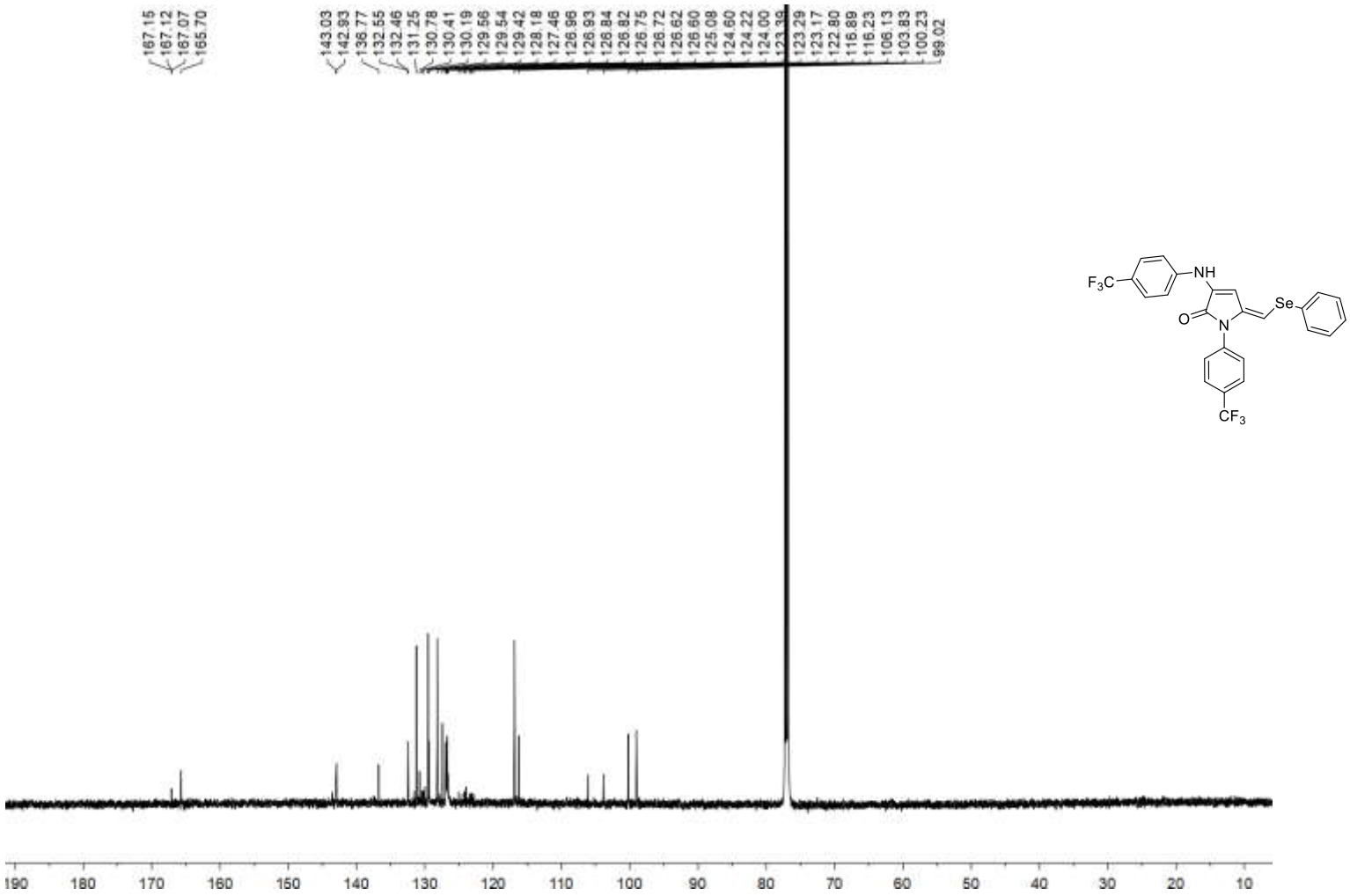


Figure S71. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **6g**

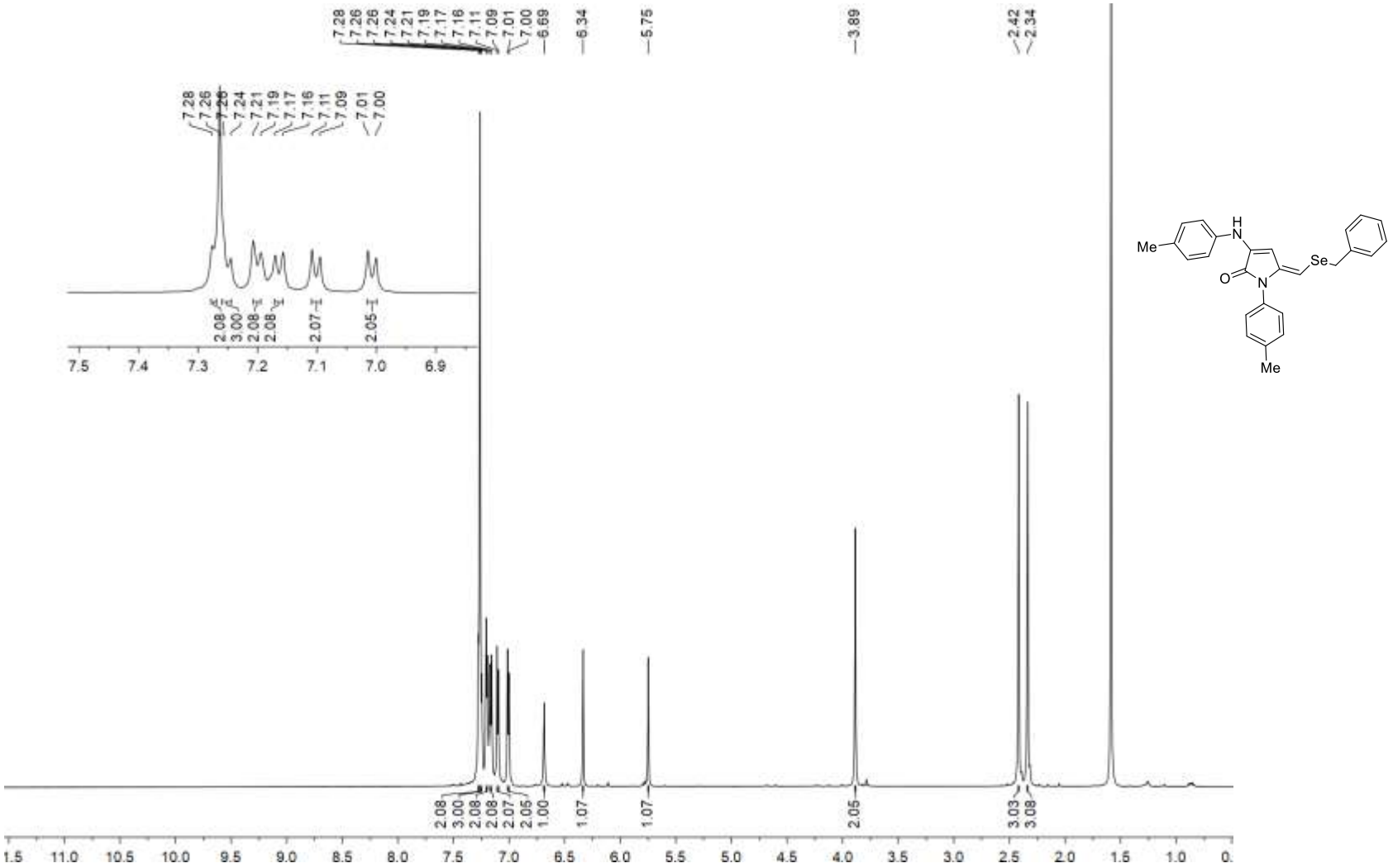


Figure S72. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **6h**

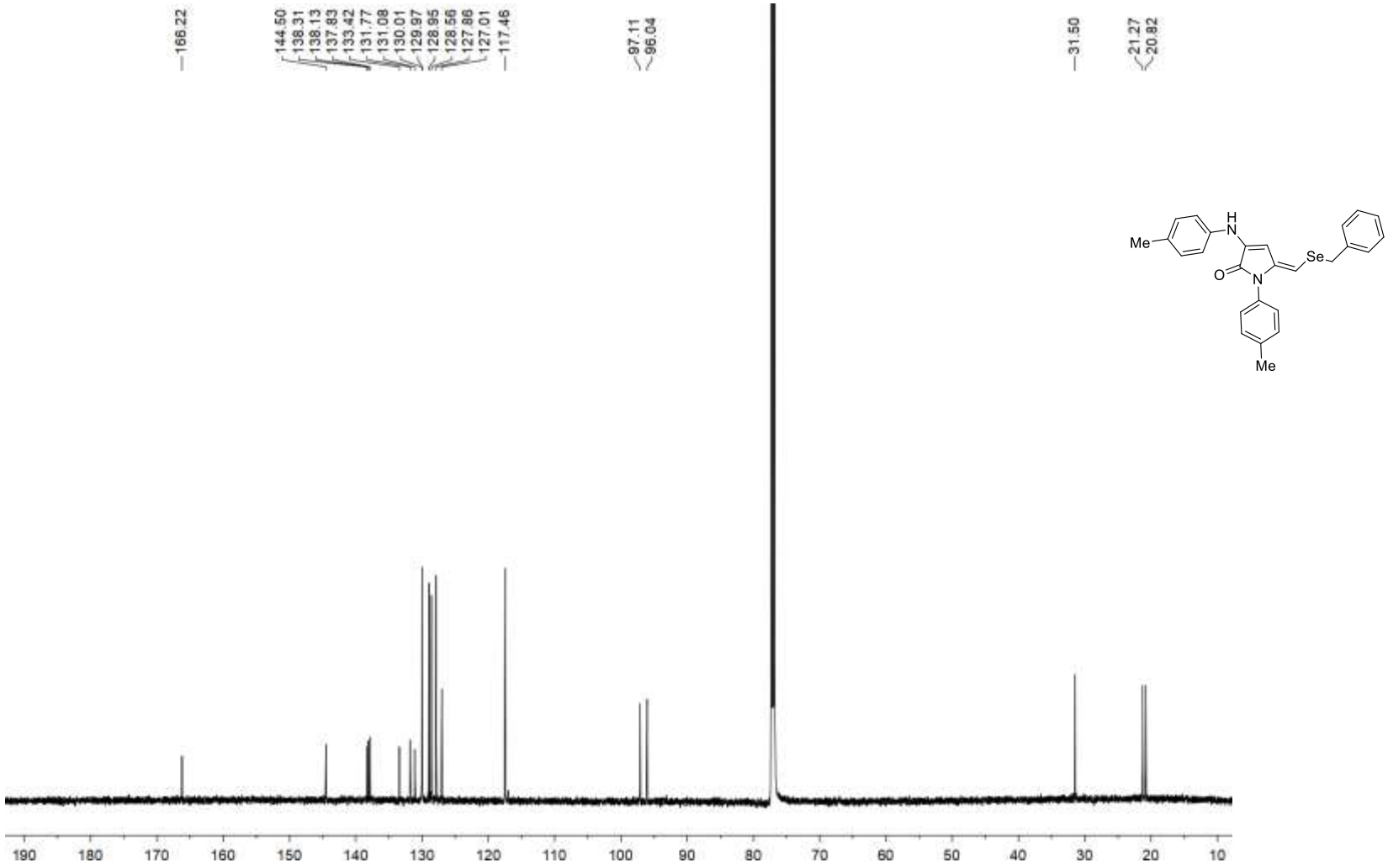


Figure S73. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **6h**

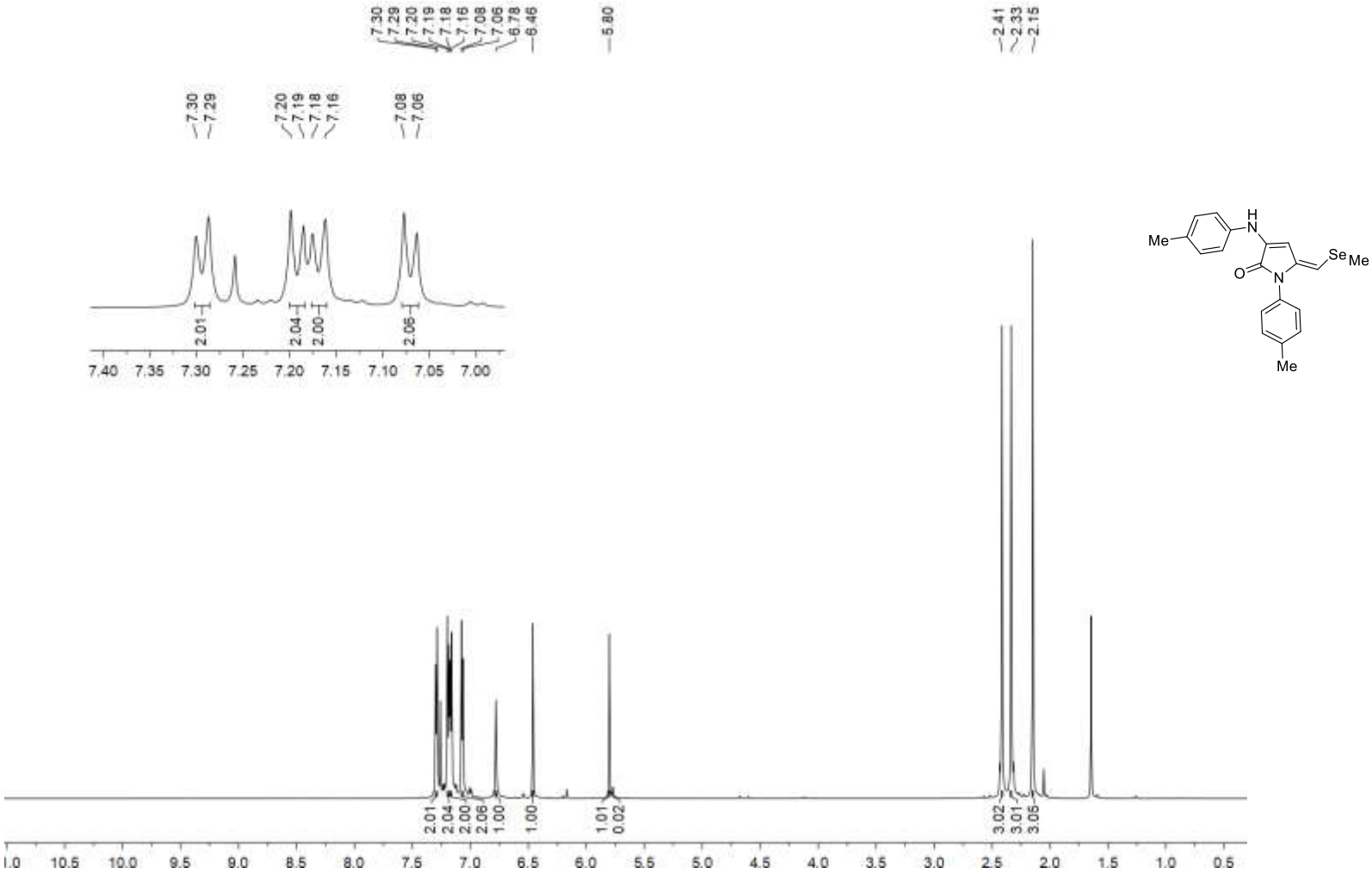


Figure S74. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **6i**

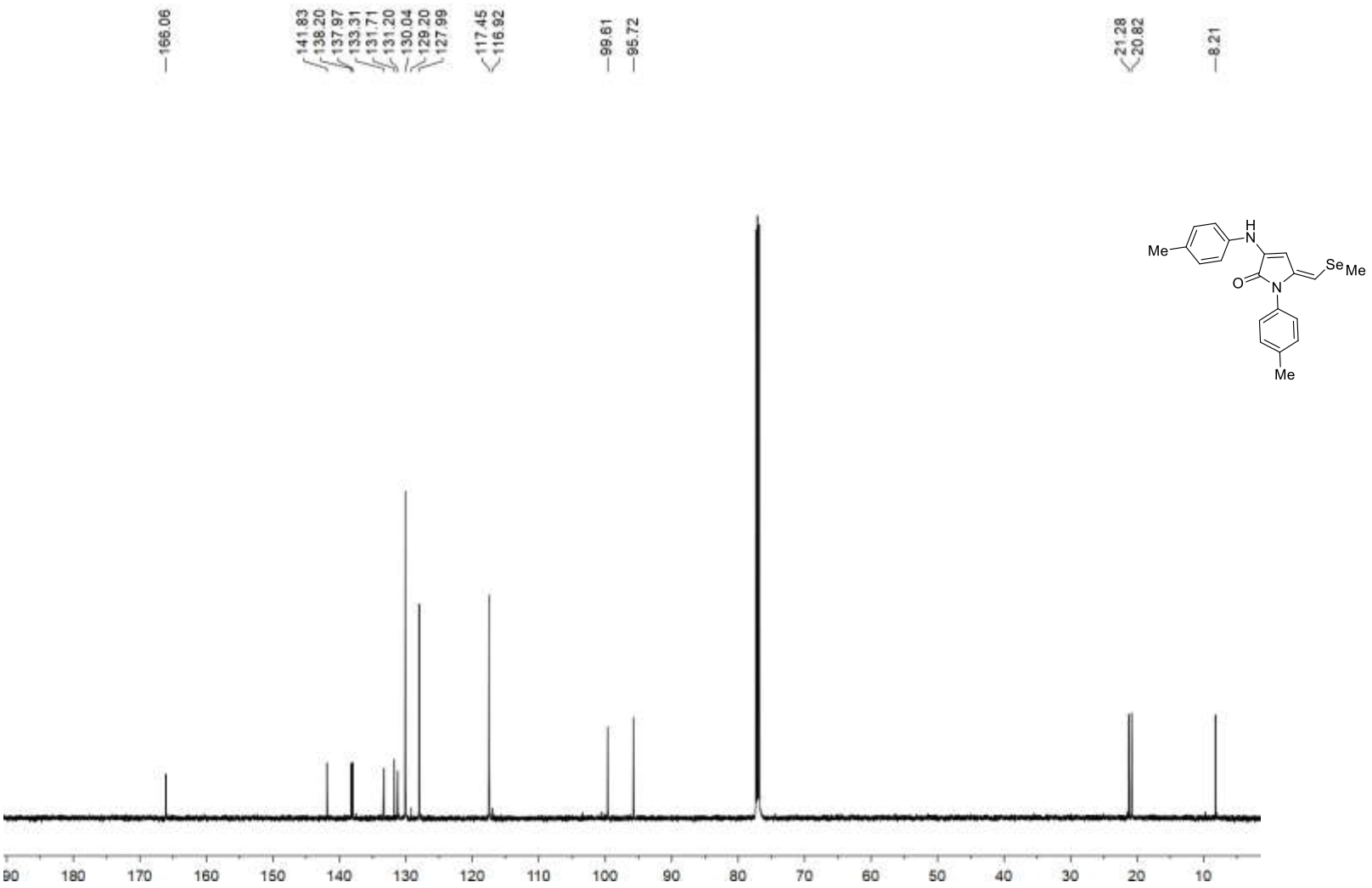


Figure S75. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **6i**

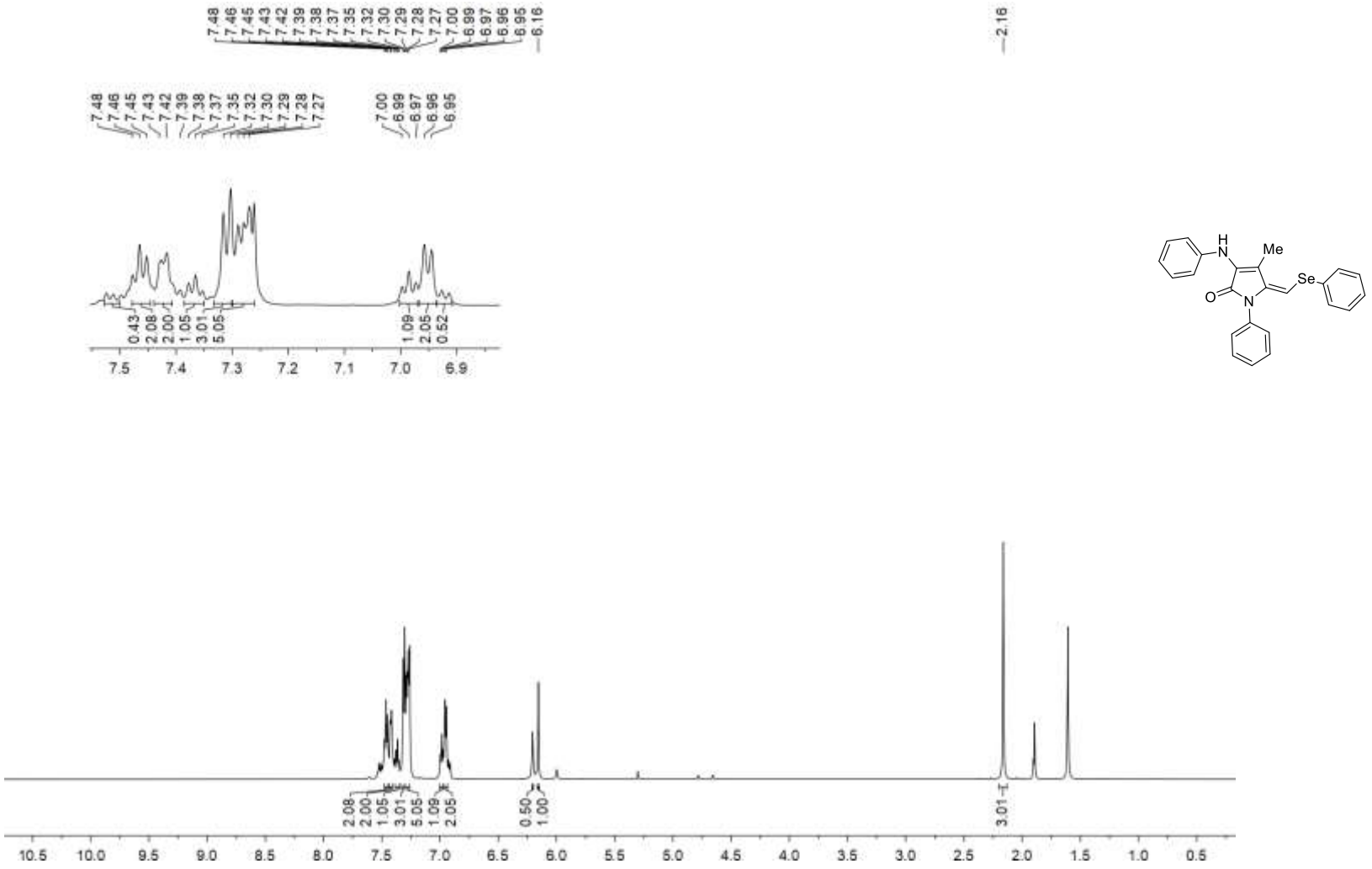


Figure S76. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **6j**

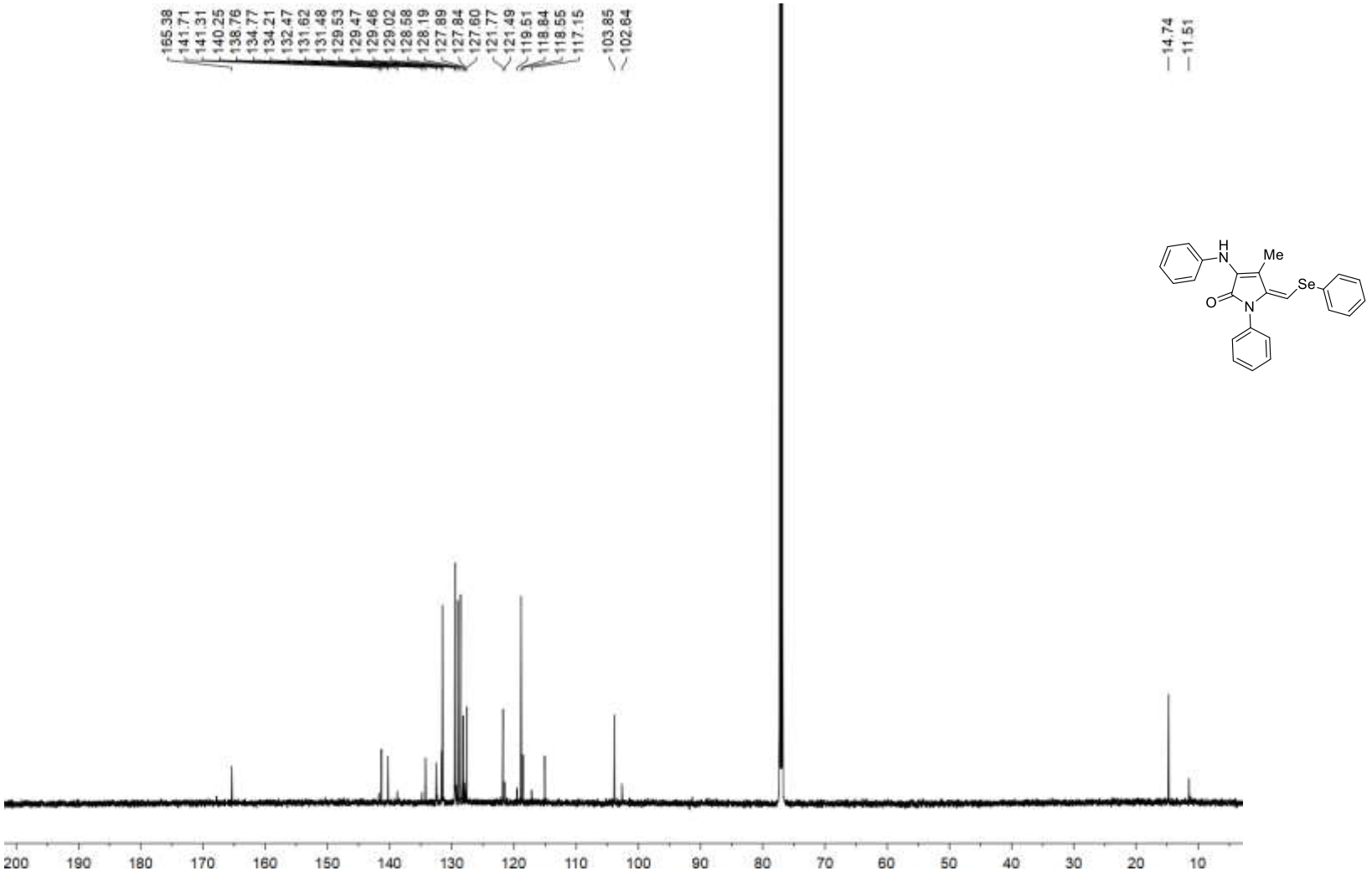


Figure S77. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **6j**

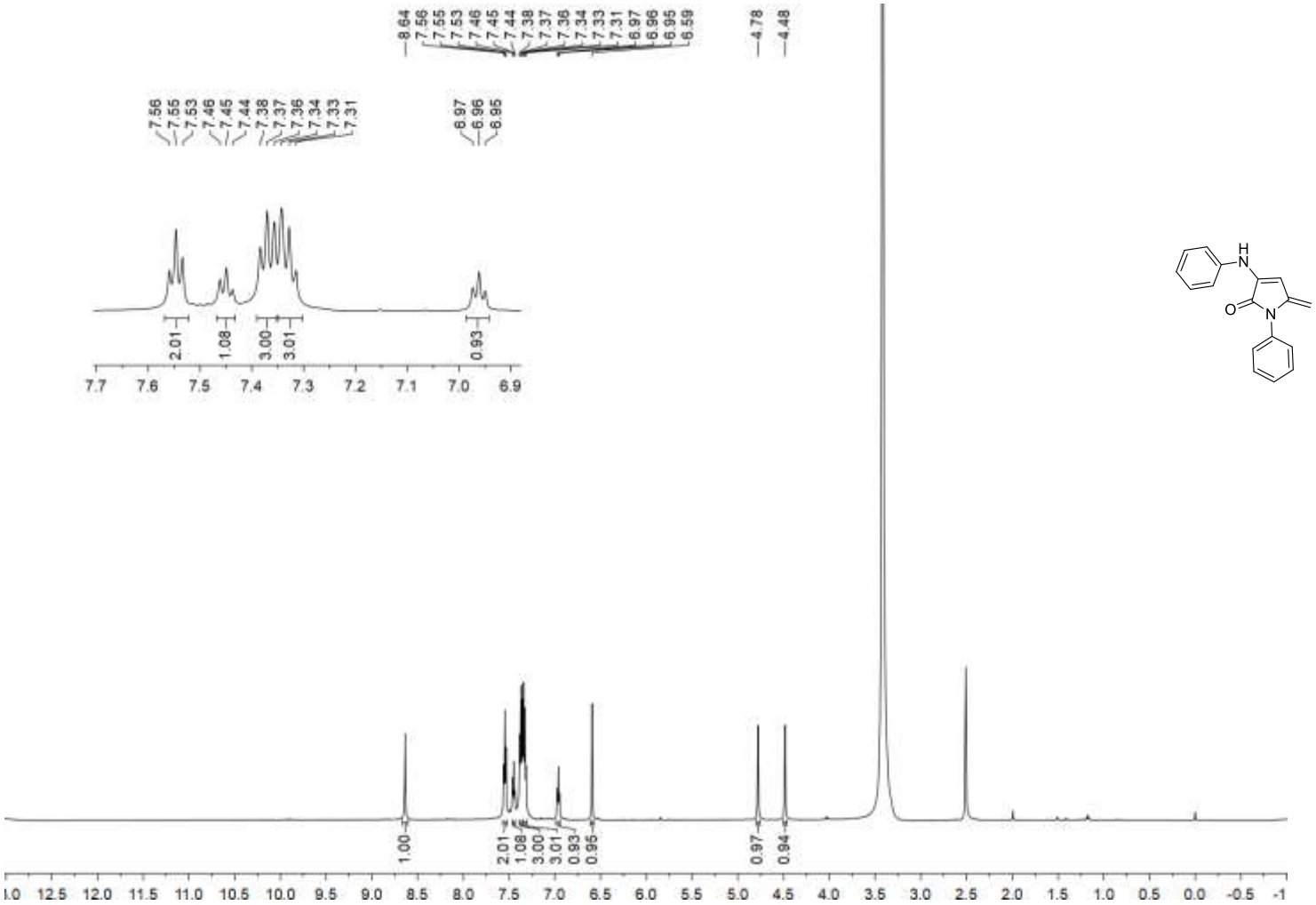


Figure S78. [1]**H NMR** (600 MHz, DMSO-d_6) spectra of compound **7a**

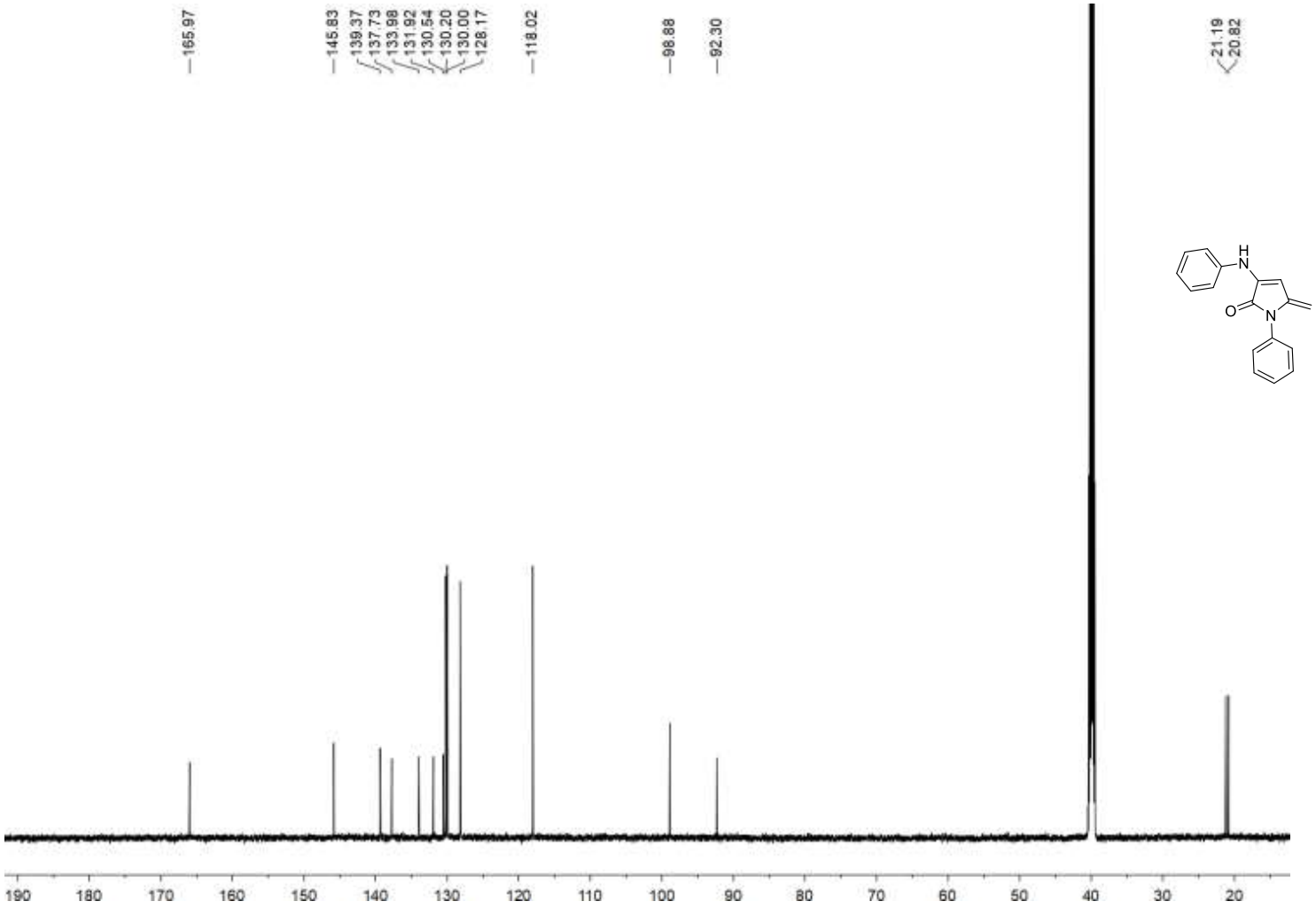


Figure S79. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **7a**

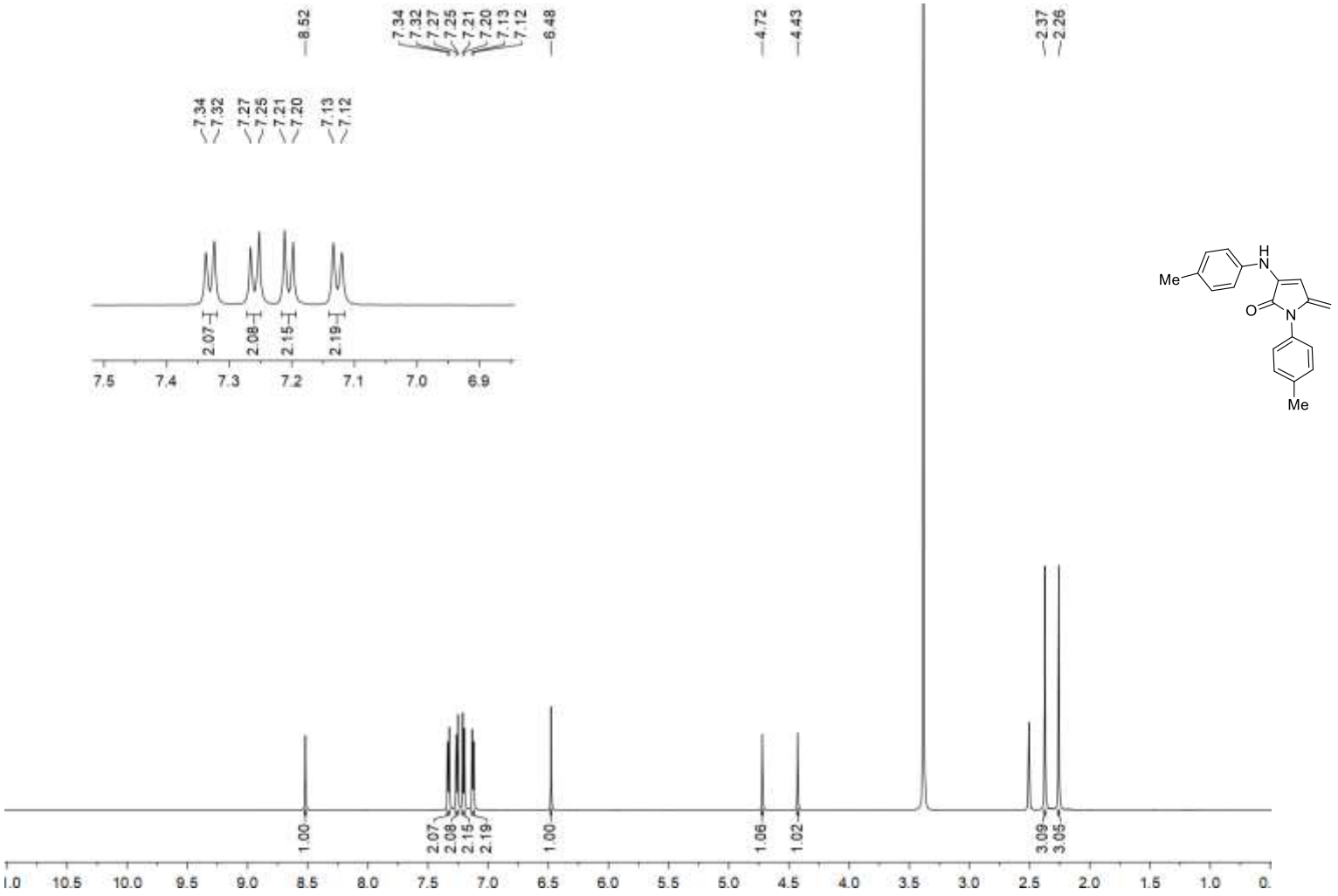


Figure S80. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **7b**

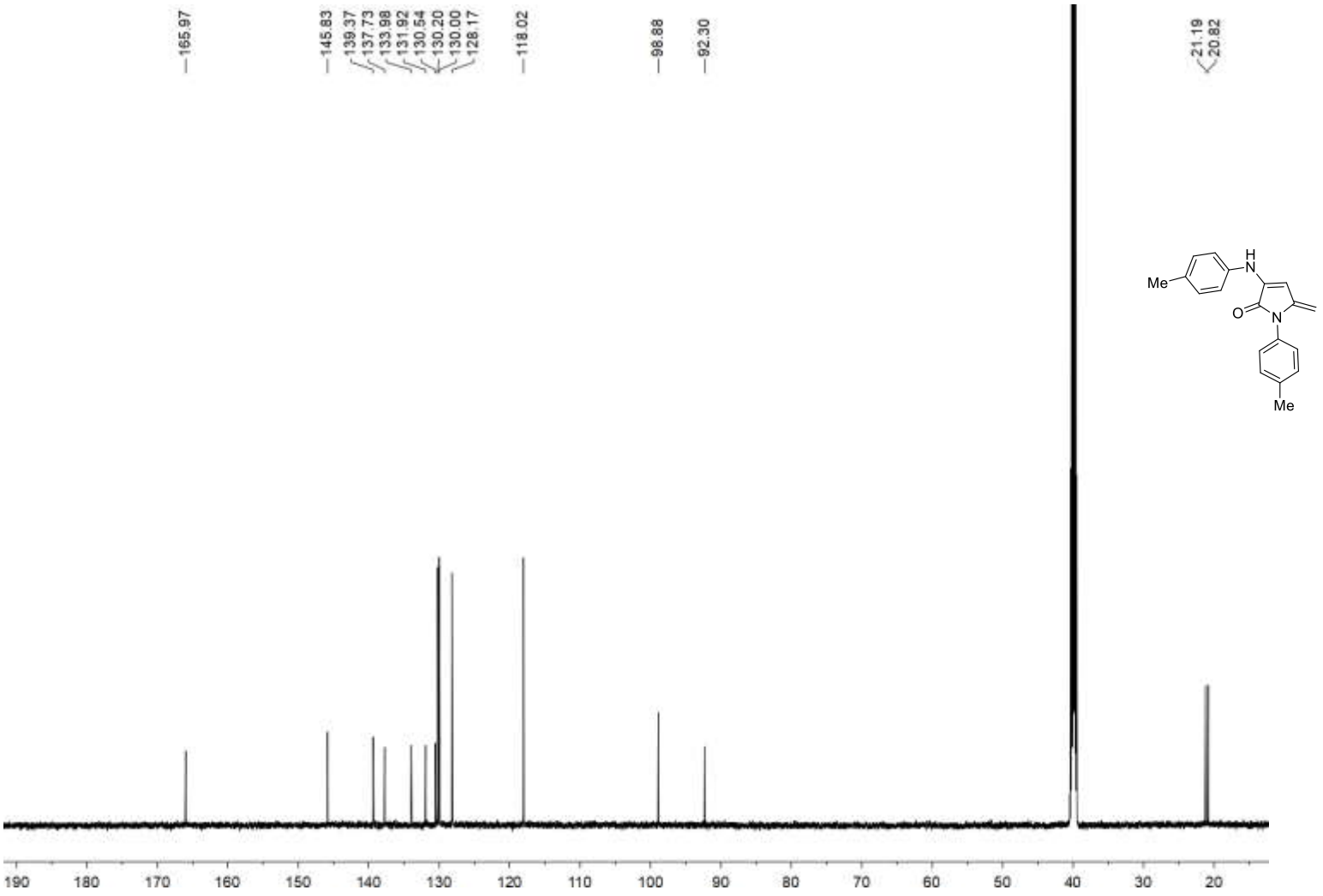


Figure S81. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **7b**

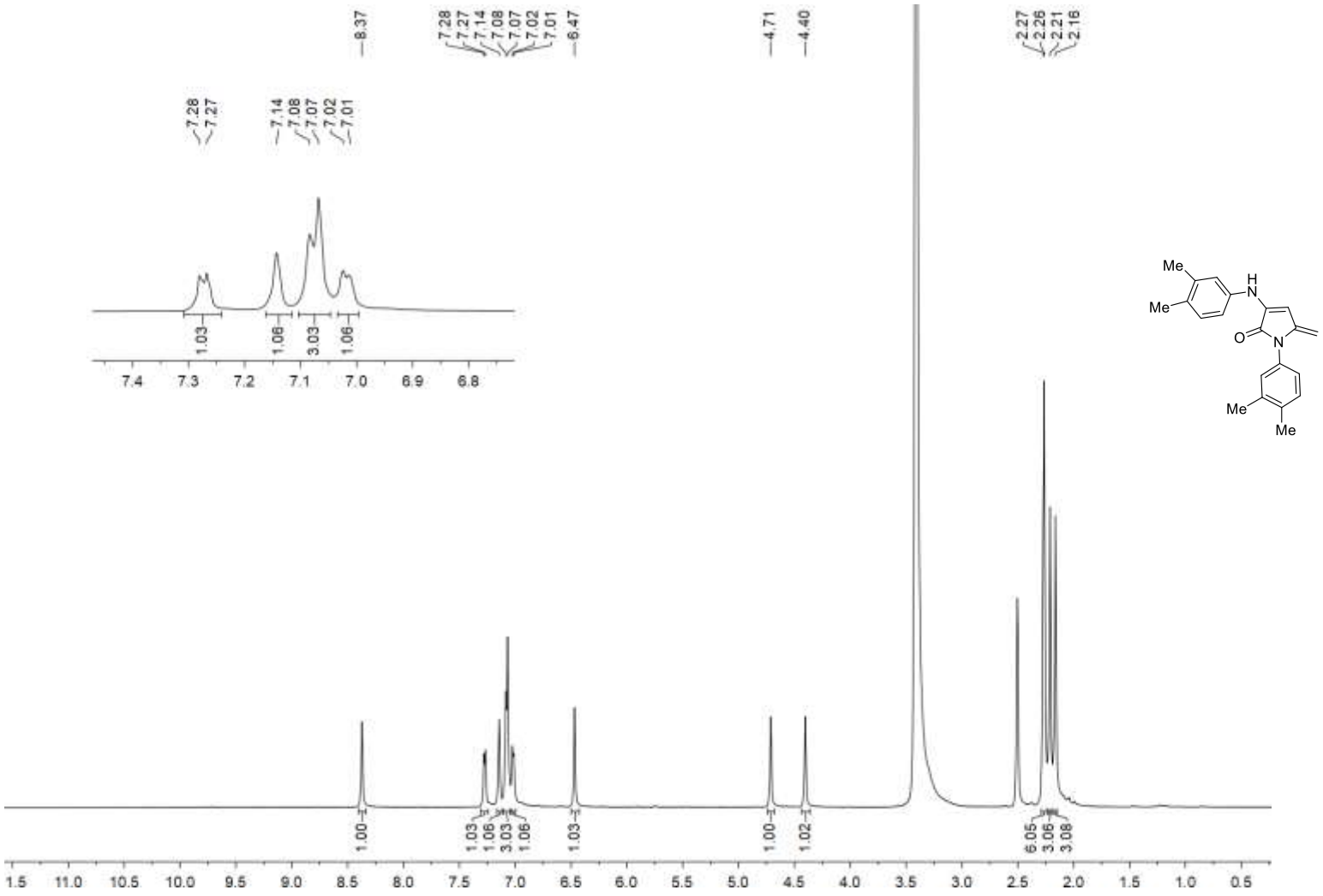


Figure S82. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **7c**

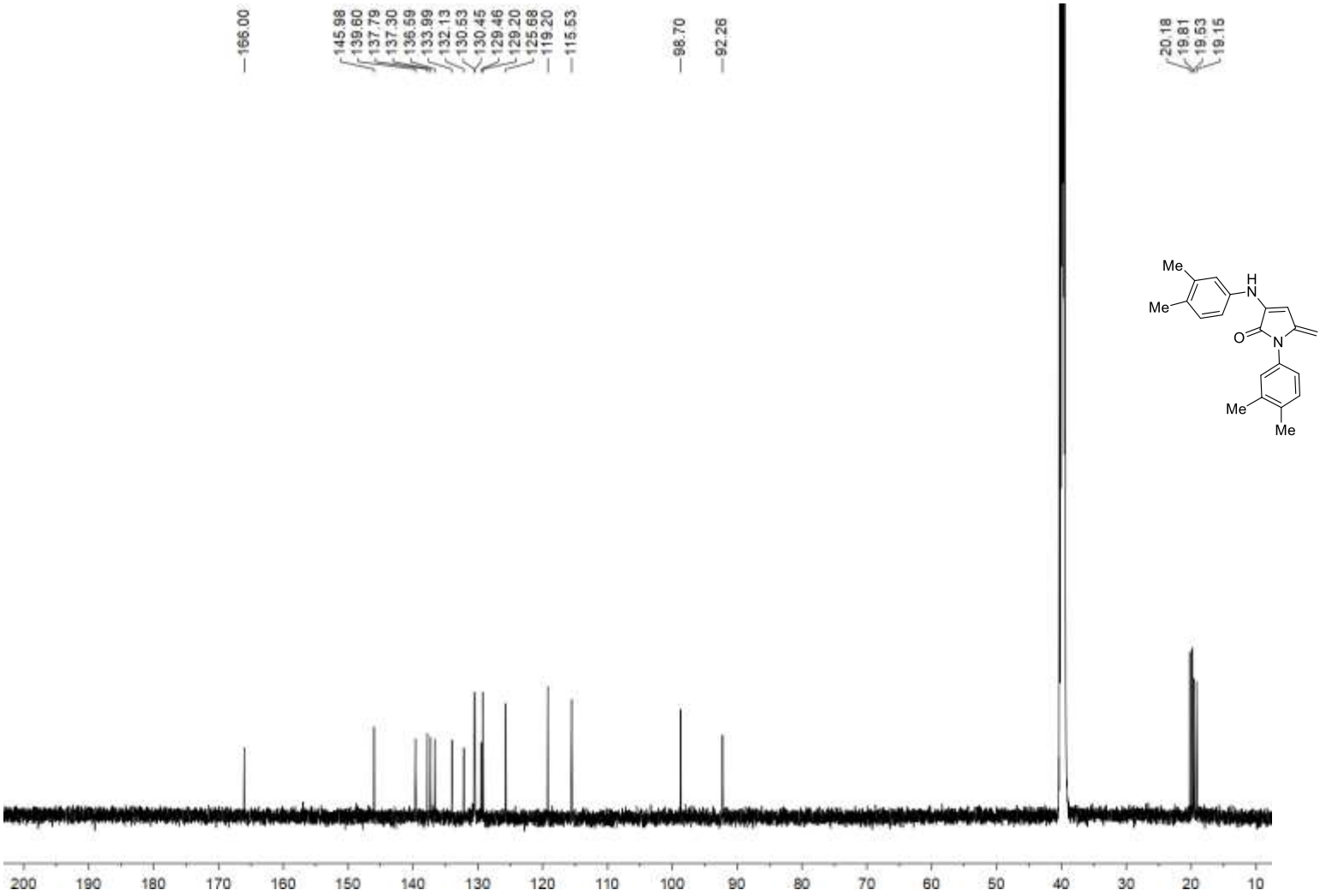


Figure S83. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **7c**

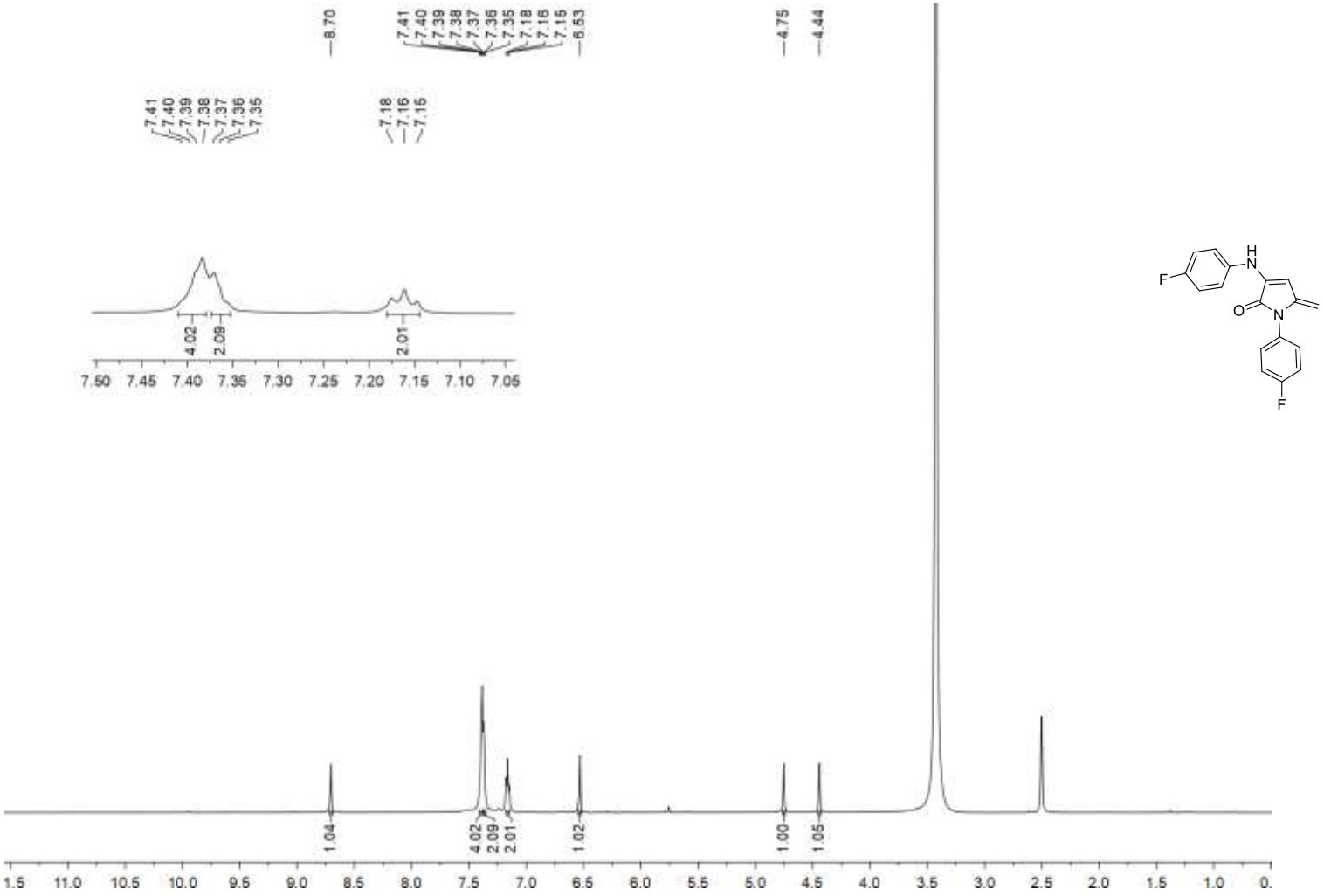


Figure S84. [1]**H NMR** (600 MHz, DMSO-d_6) spectra of compound **7d**

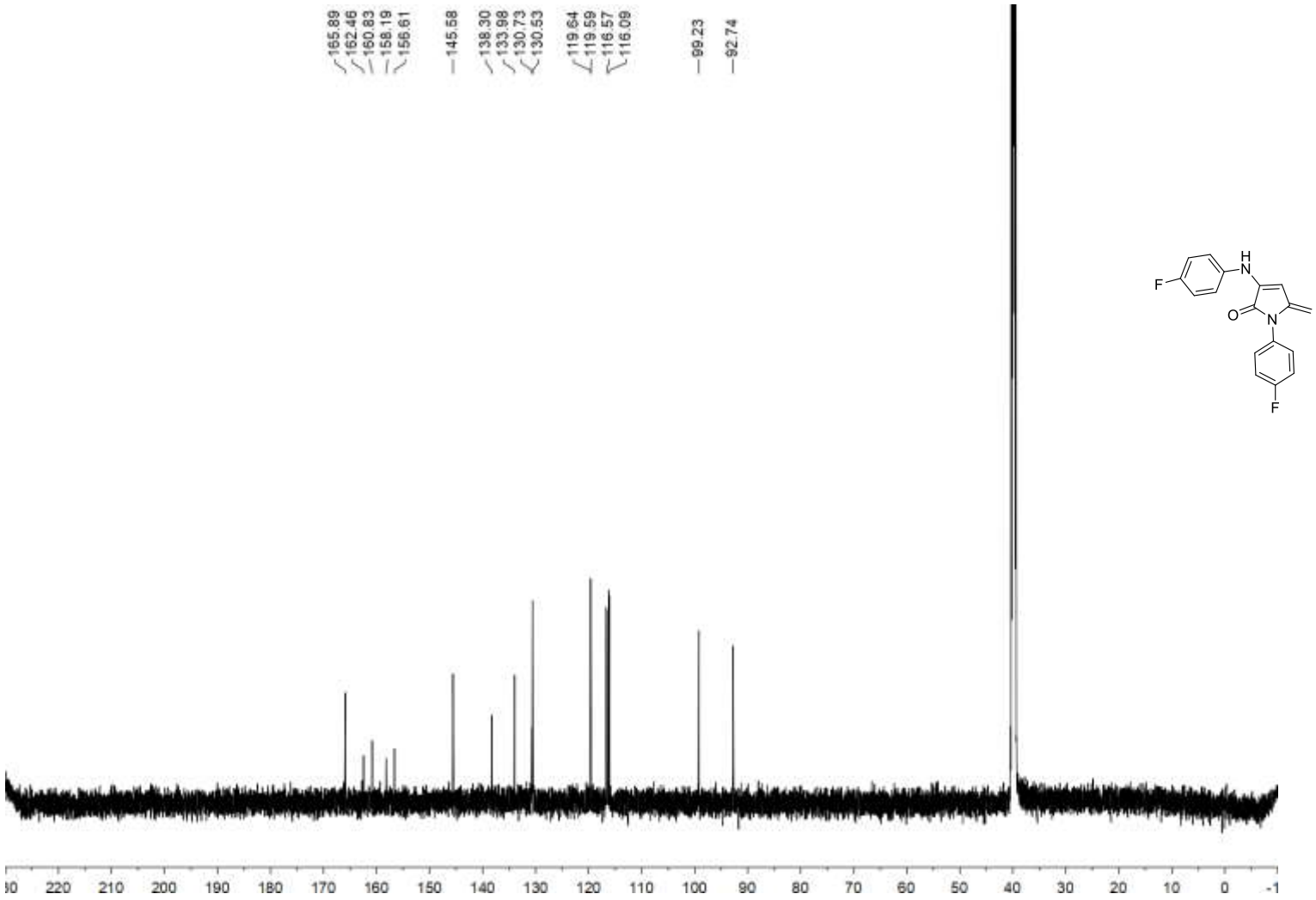


Figure S85. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **7d**

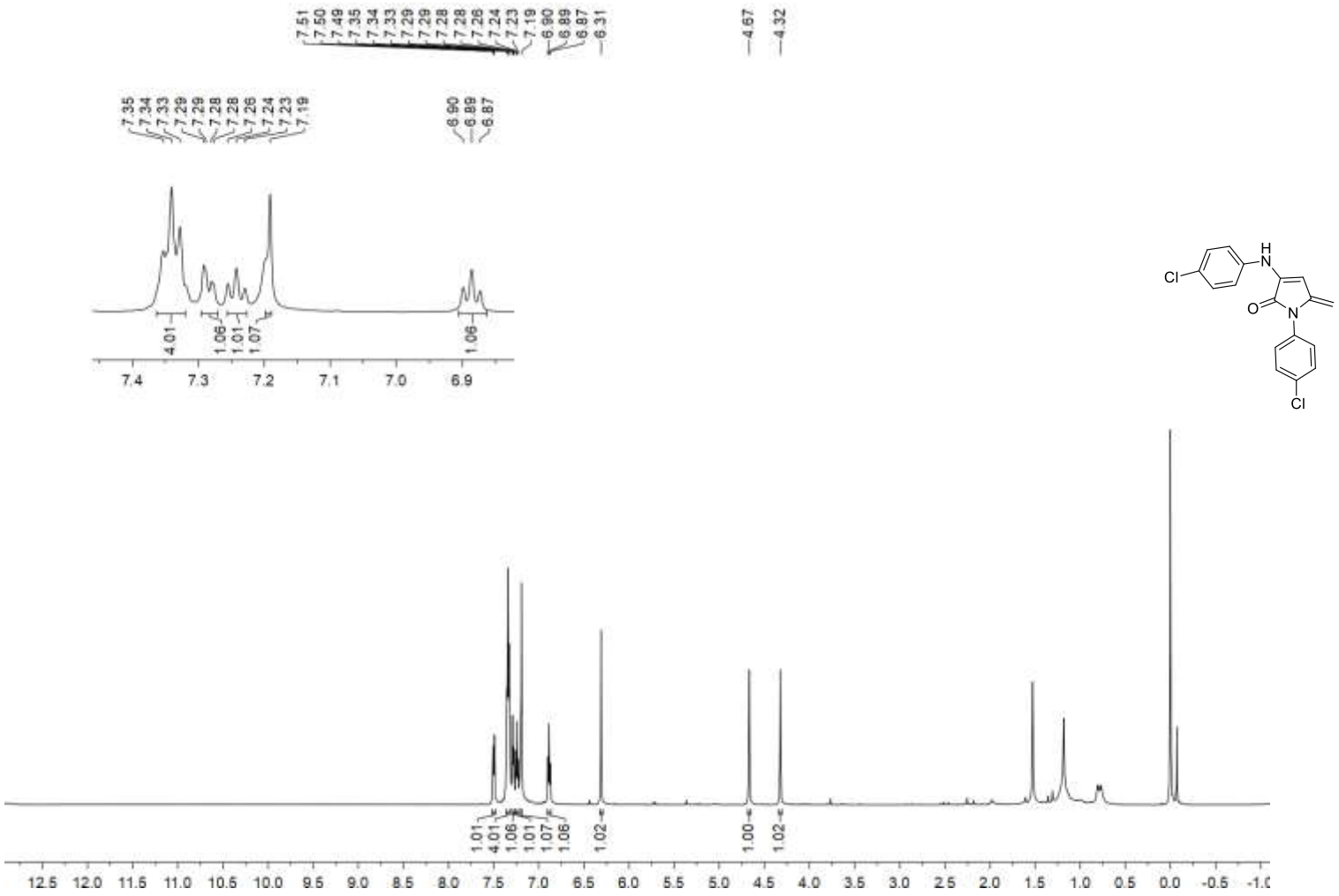


Figure S86. [1]**H NMR** (600 MHz, DMSO-d_6) spectra of compound **7e**

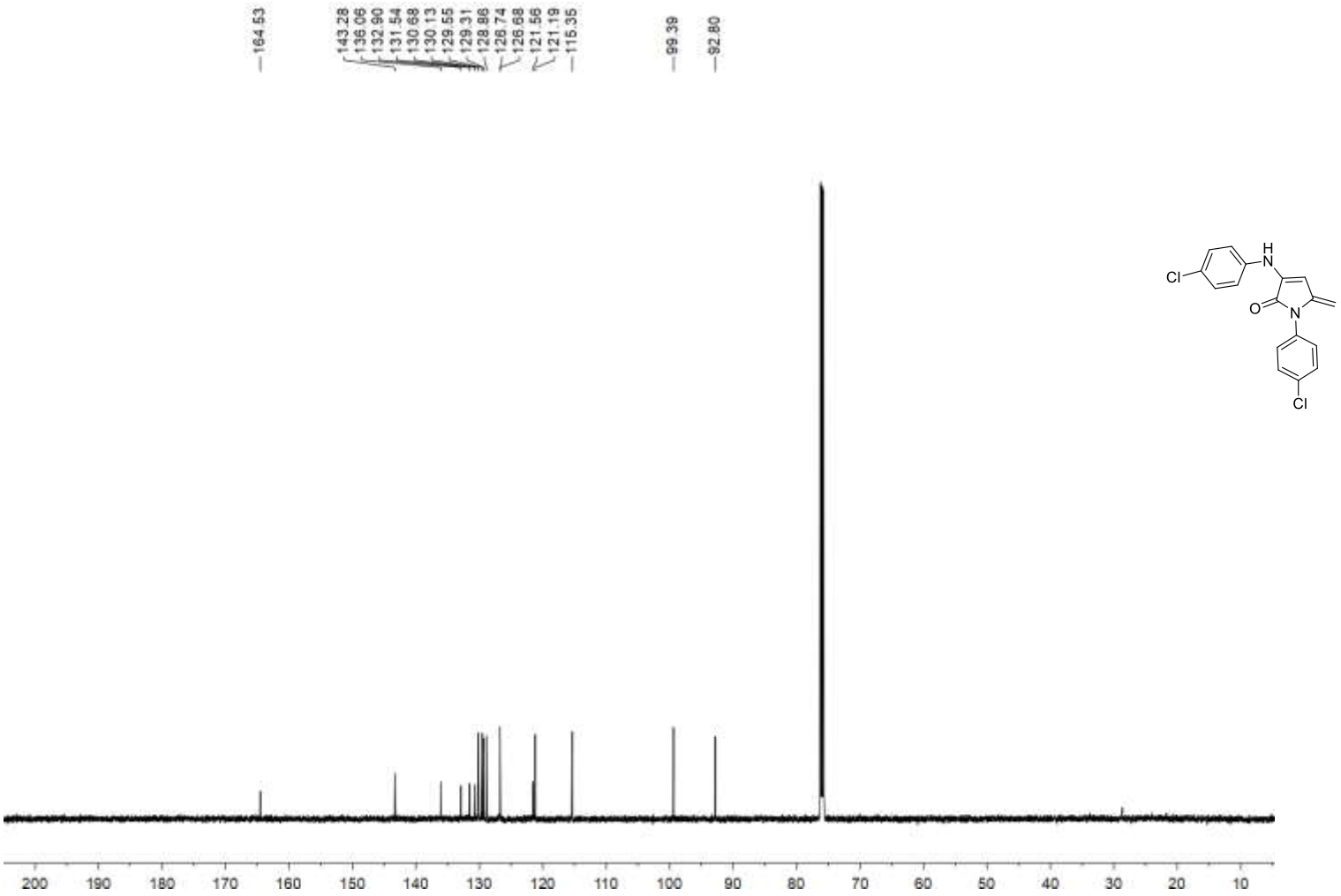


Figure S87. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **7e**

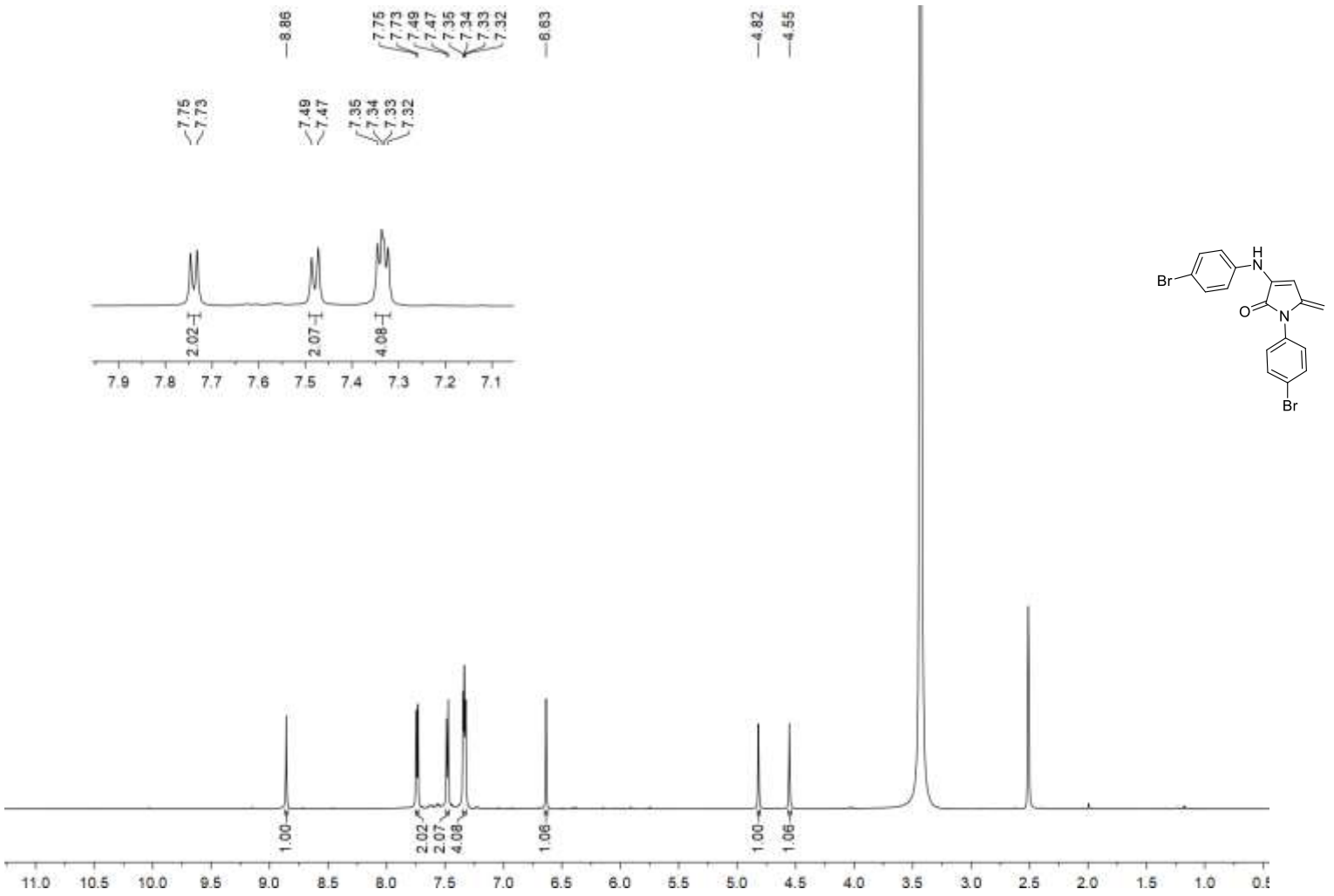


Figure S88. **^{1}H NMR** (600 MHz, DMSO-d_6) spectra of compound **7f**

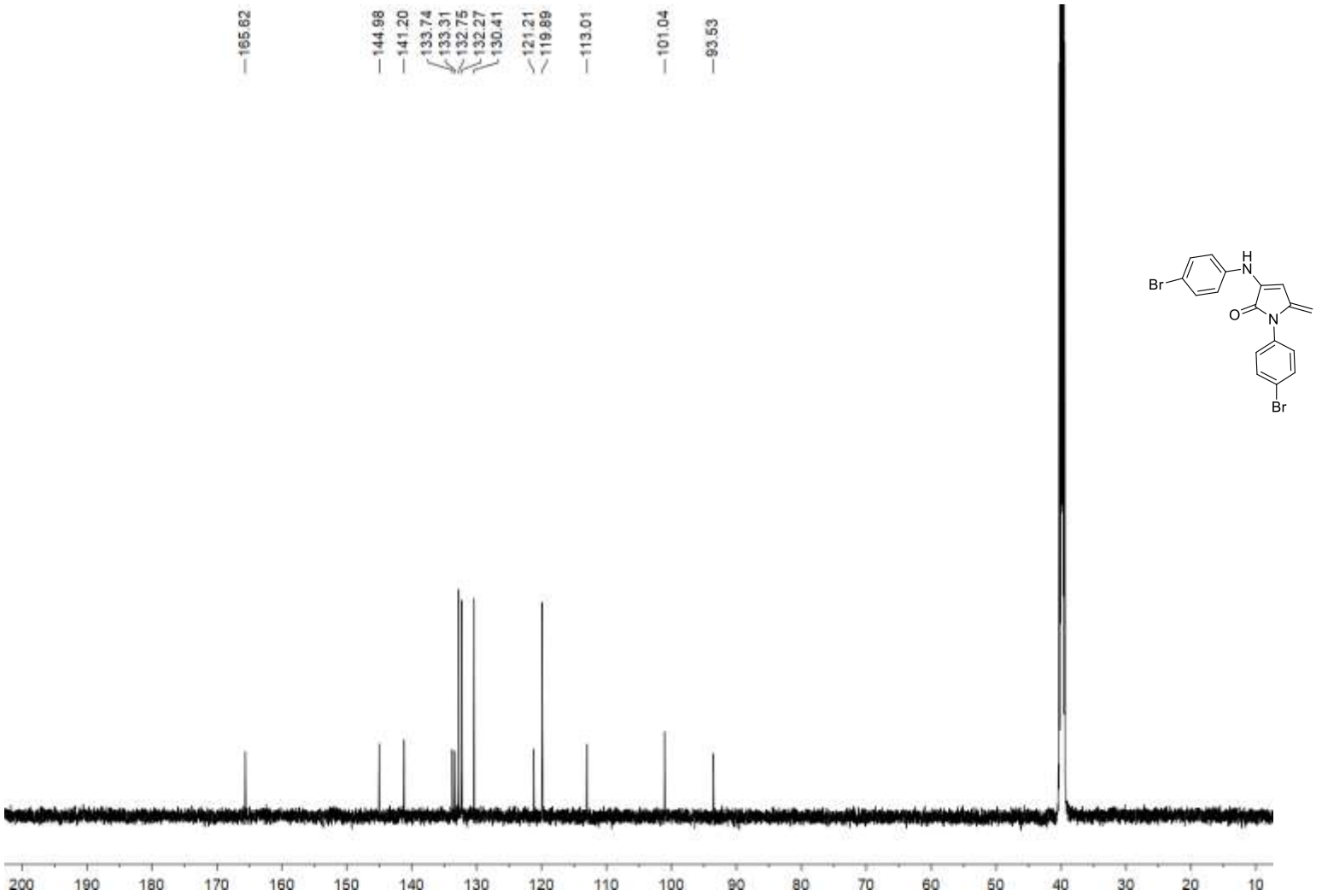


Figure S89. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **7f**

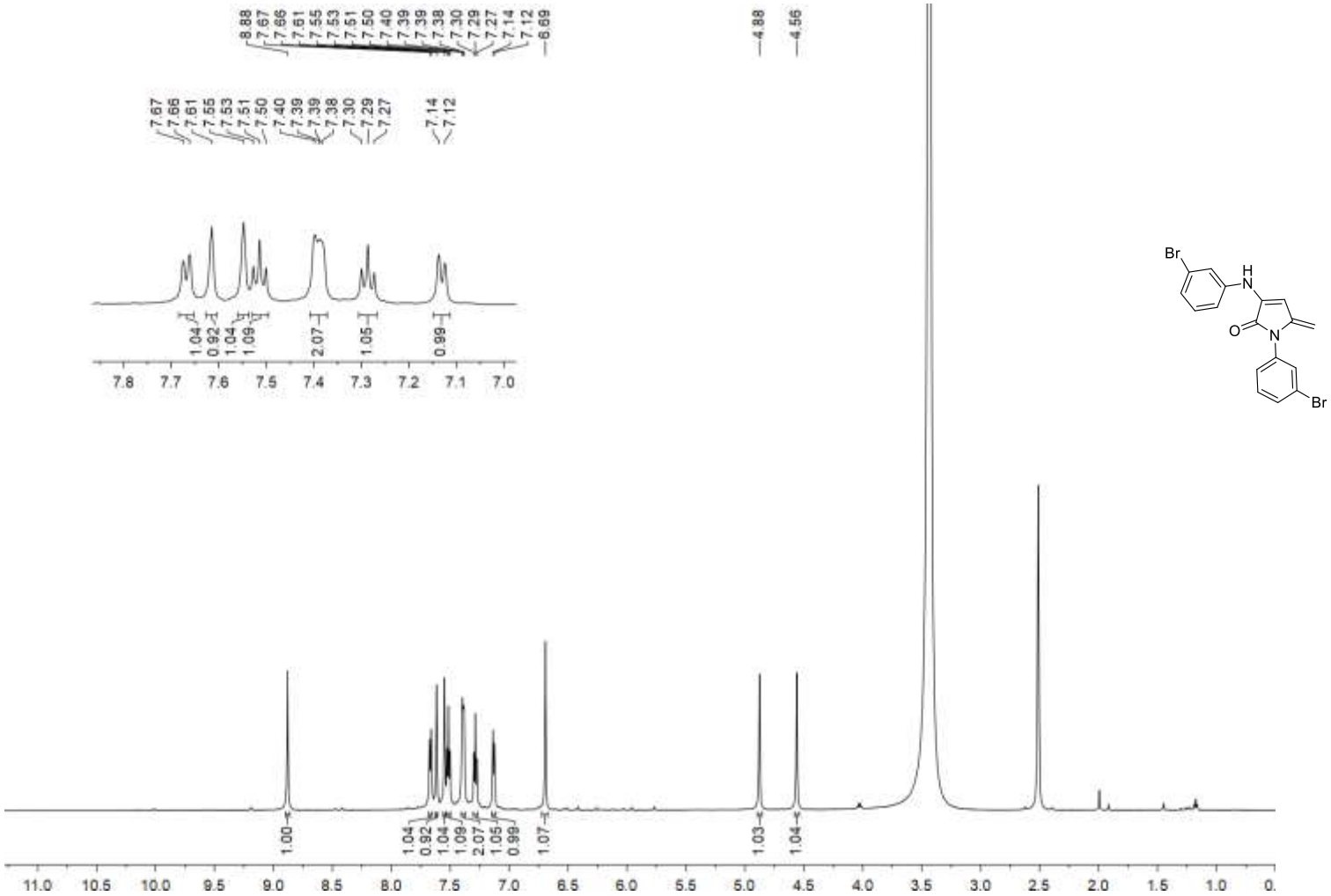


Figure S90. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **7g**

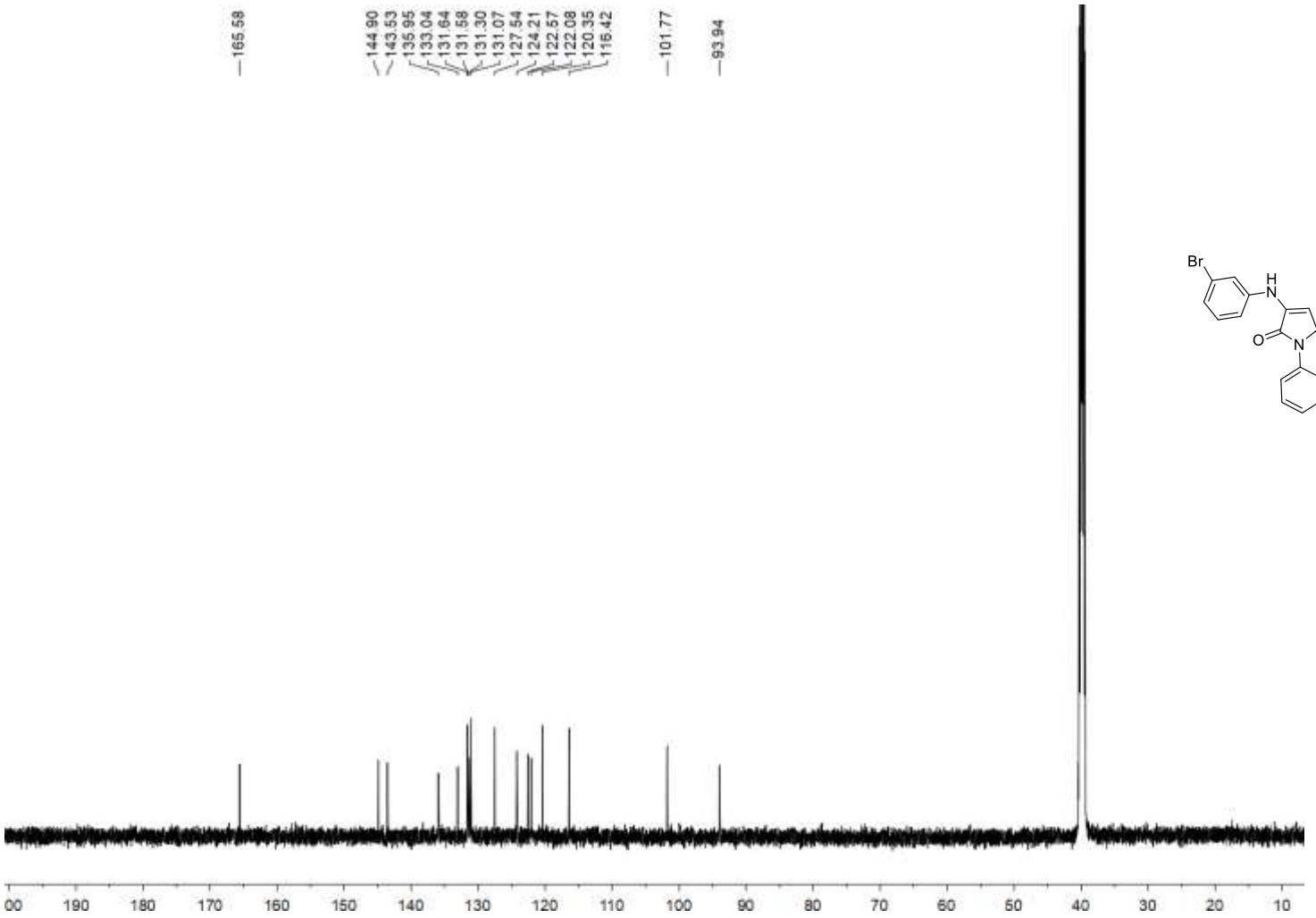


Figure S91. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **7g**

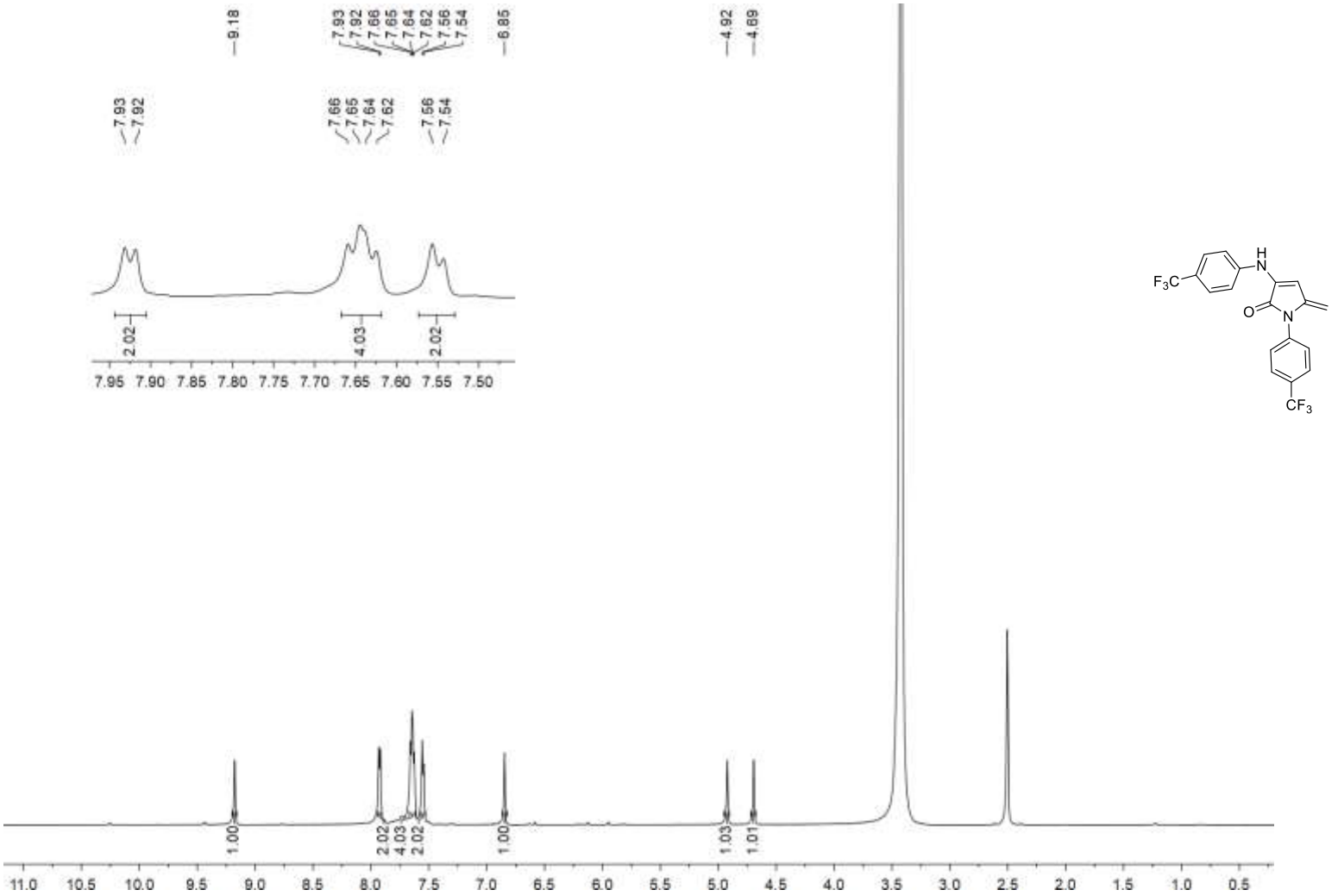


Figure S92. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **7h**

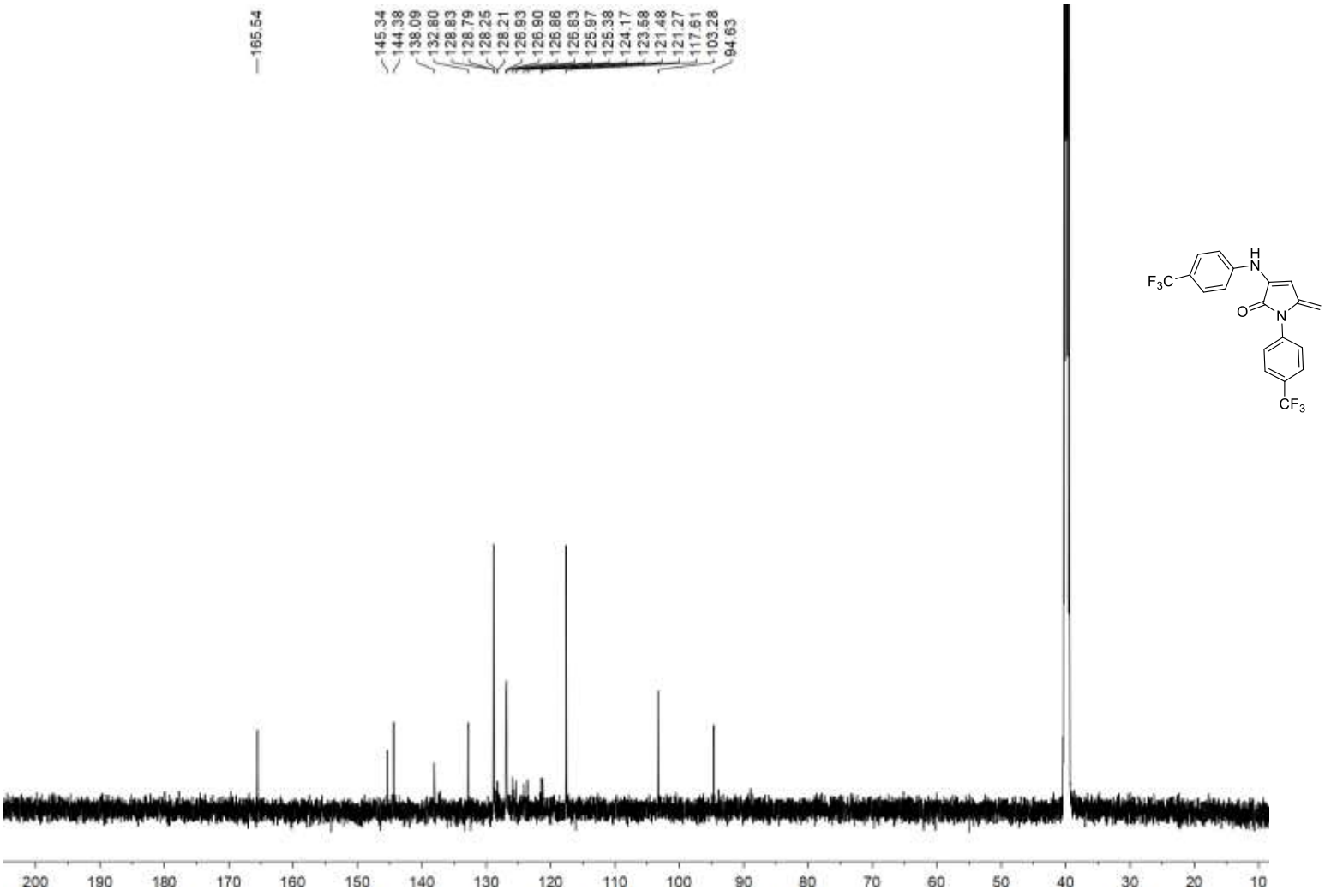


Figure S93. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **7h**

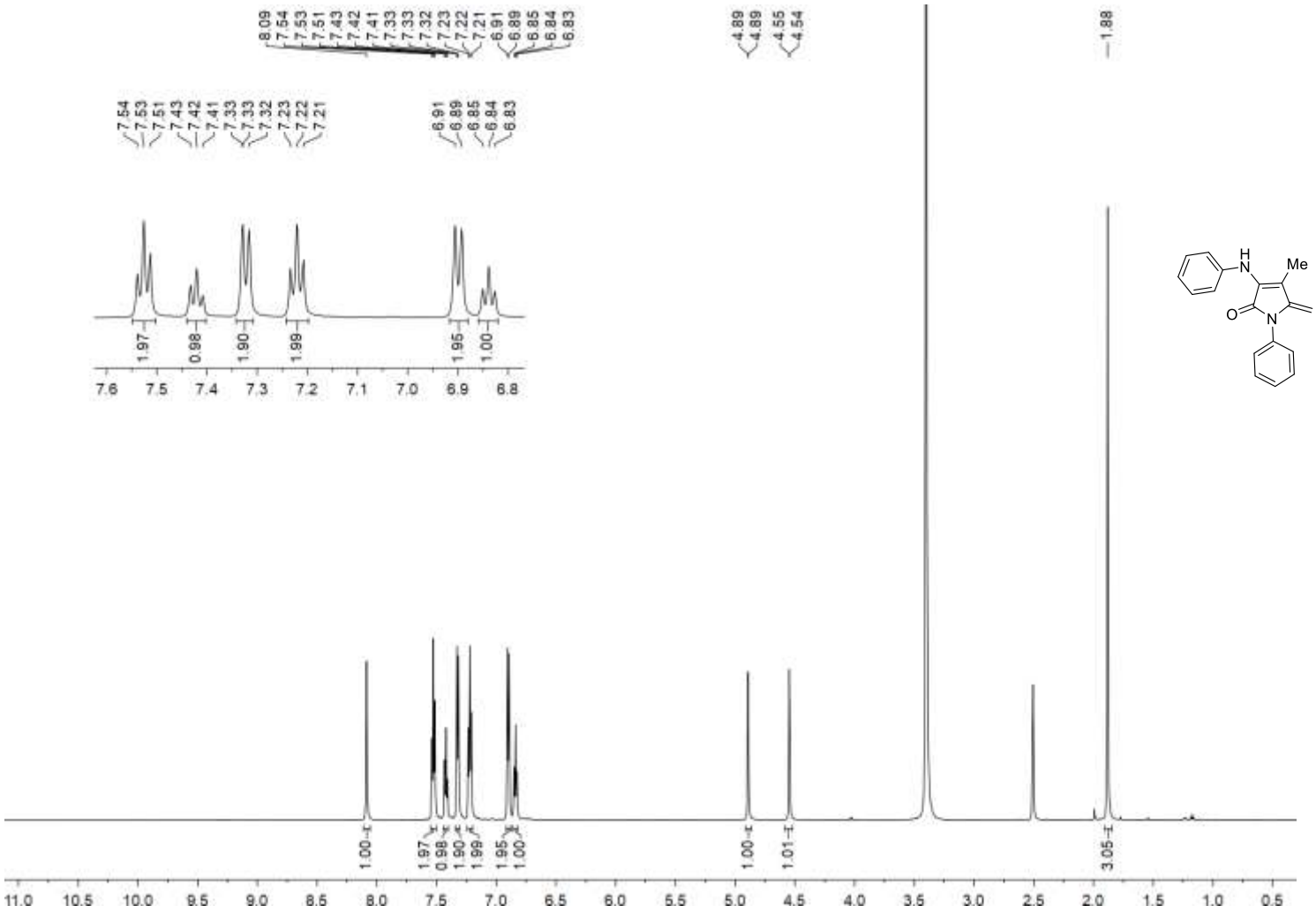


Figure S94. [1]**H NMR** (600 MHz, DMSO-d_6) spectra of compound **7i**

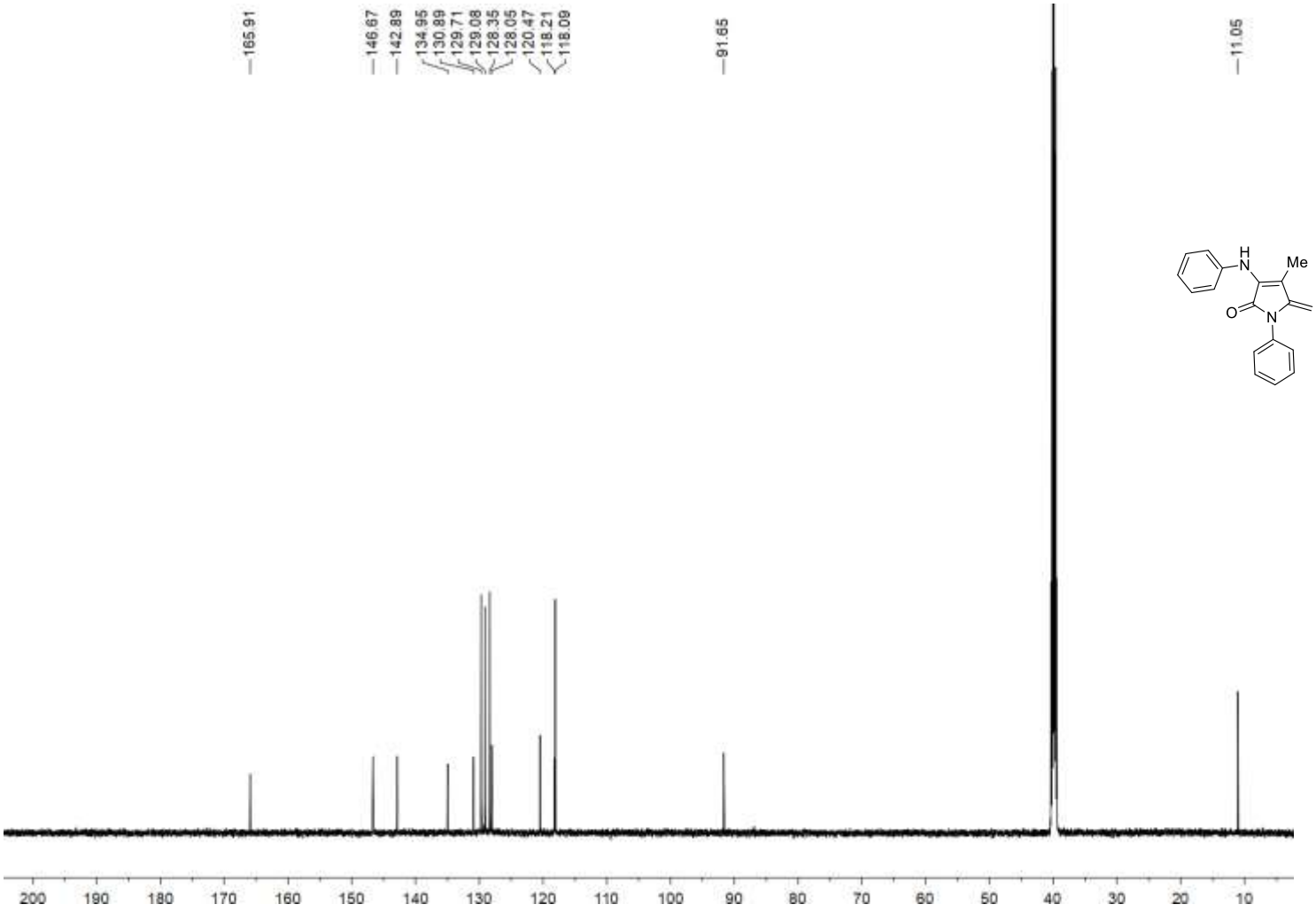


Figure S95. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **7i**

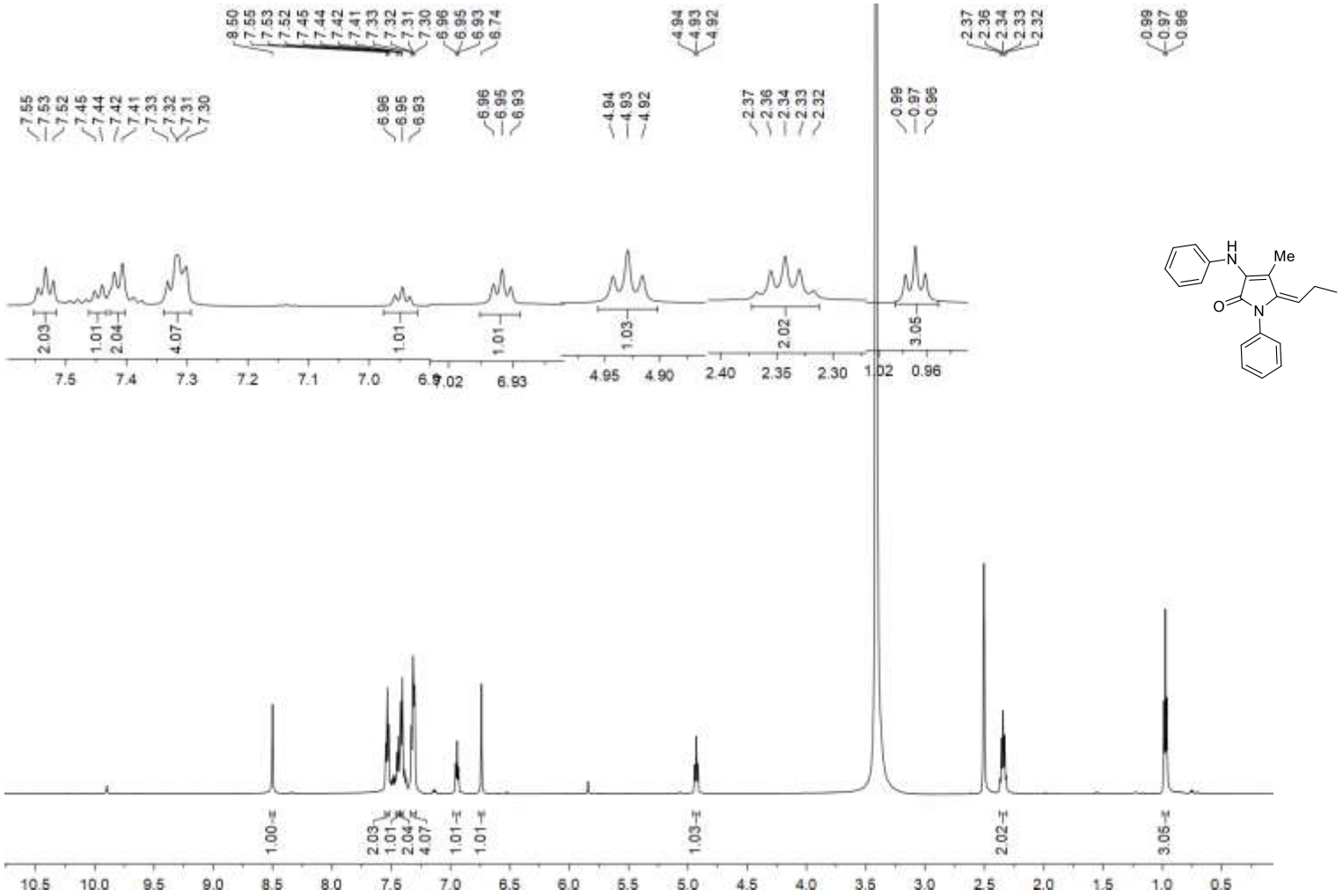


Figure S96. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **7j**

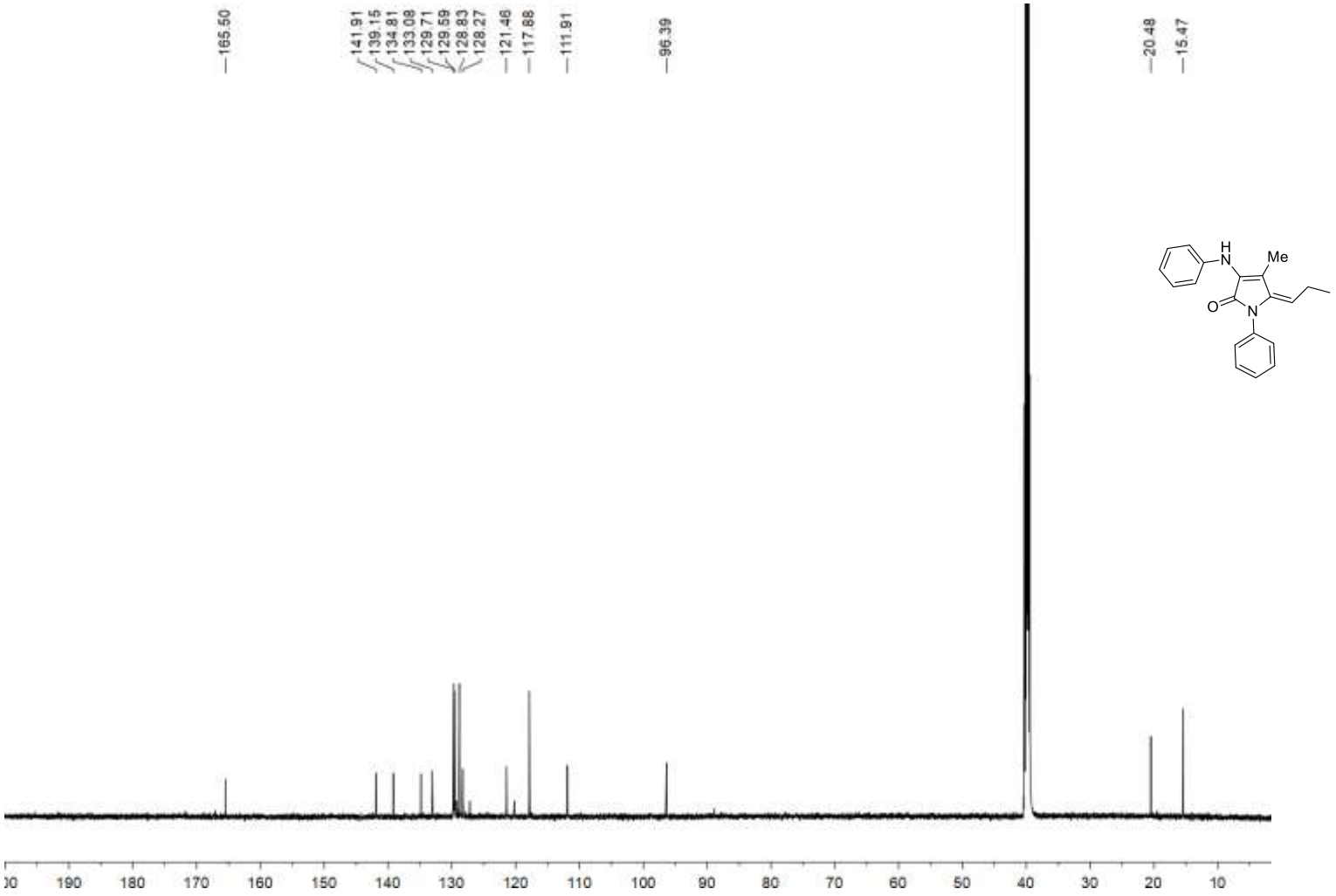


Figure S97. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **7j**

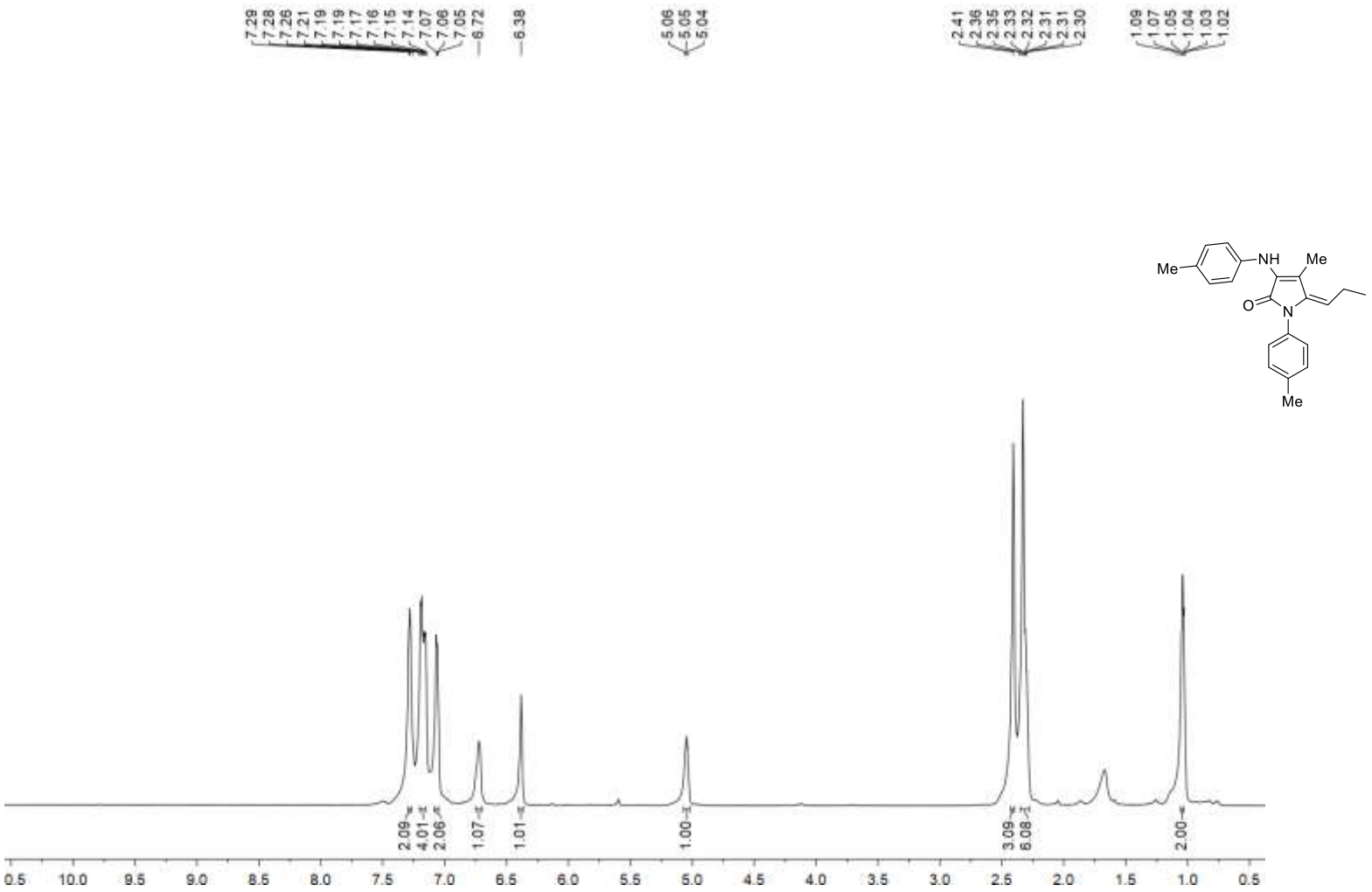


Figure S98. **^{1}H NMR** (600 MHz, $CDCl_3$) spectra of compound **7k**

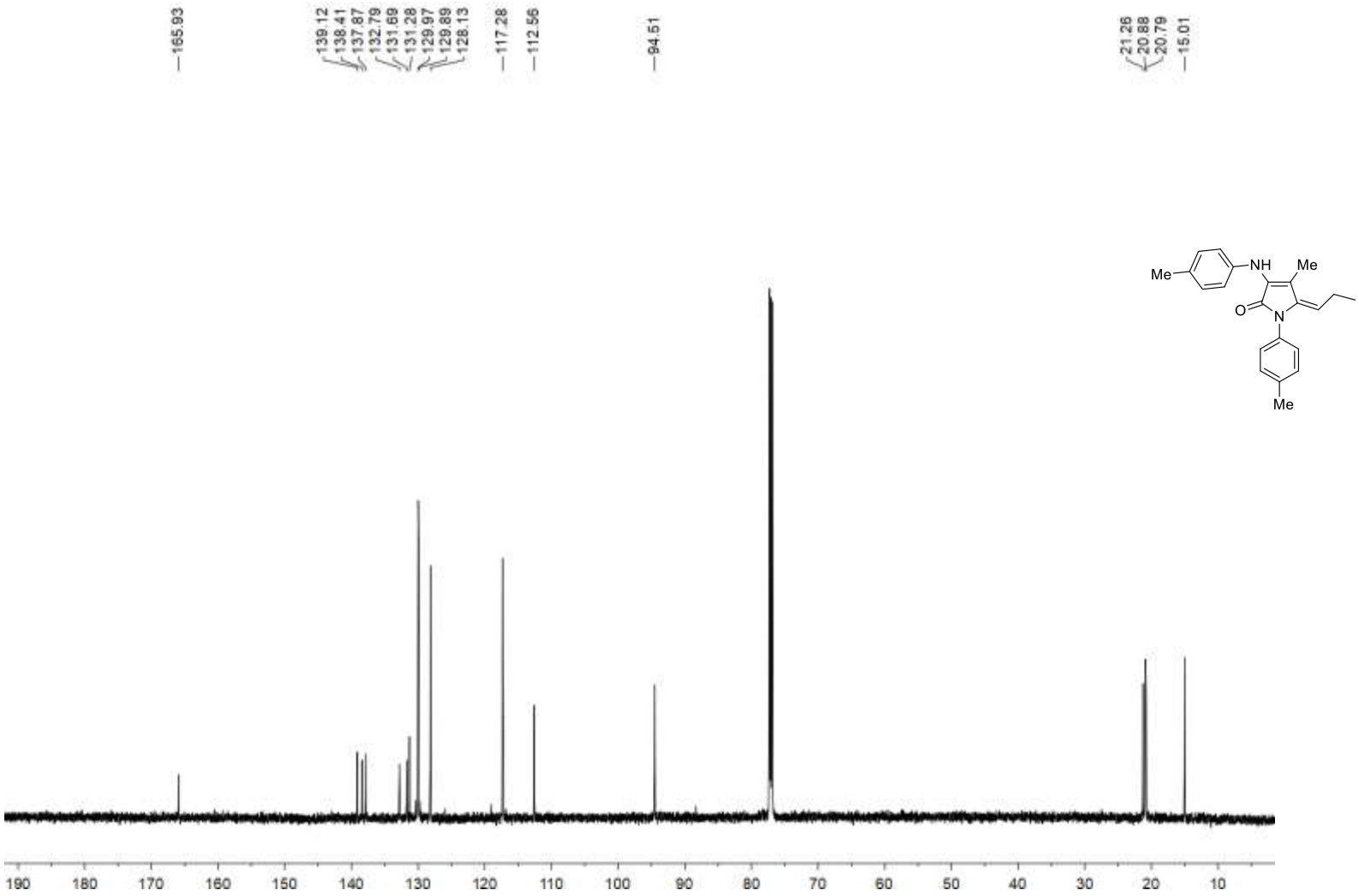


Figure S99. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **7k**

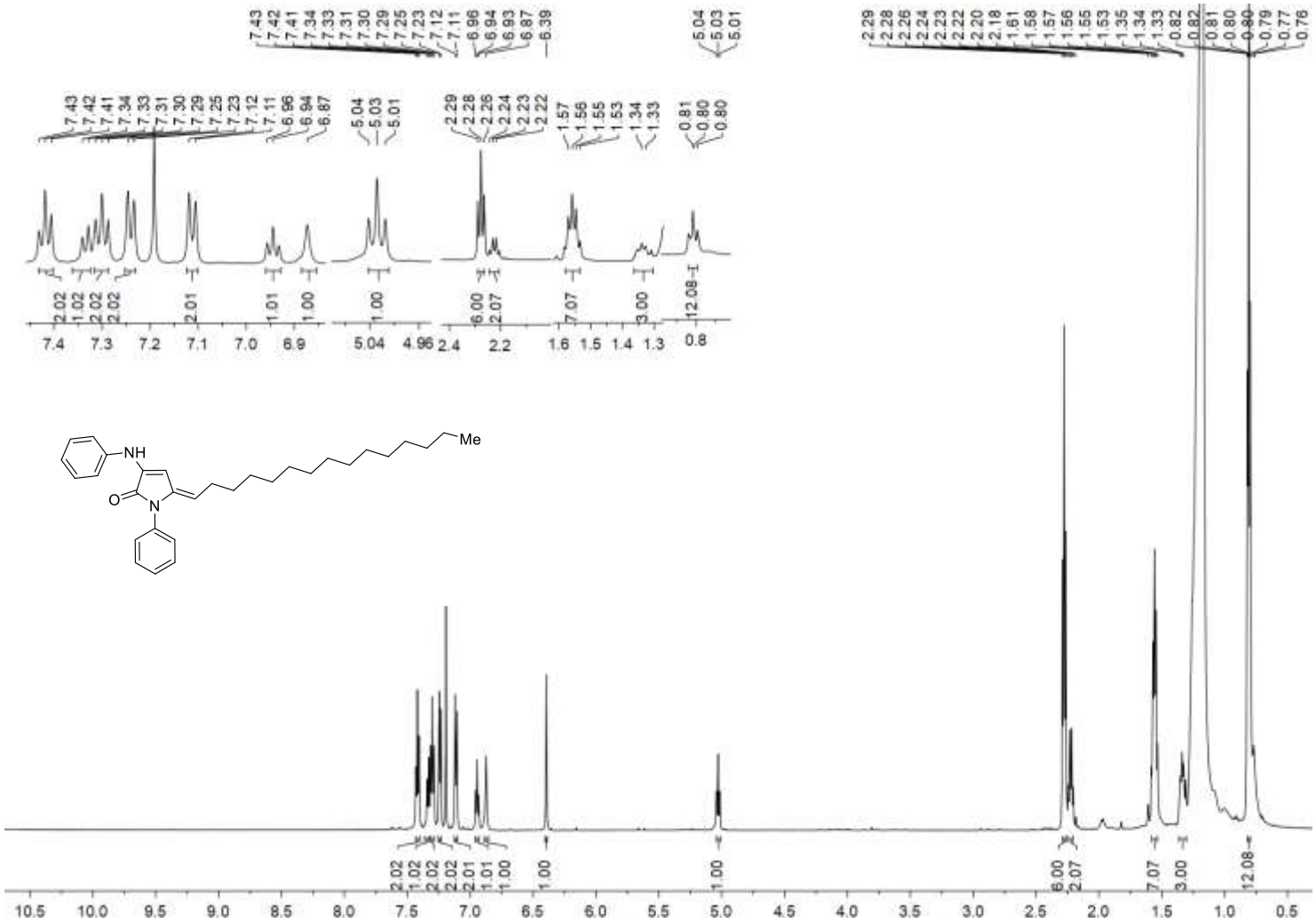


Figure S100. **^{1}H NMR** (600 MHz, $CDCl_3$) spectra of compound **7l**

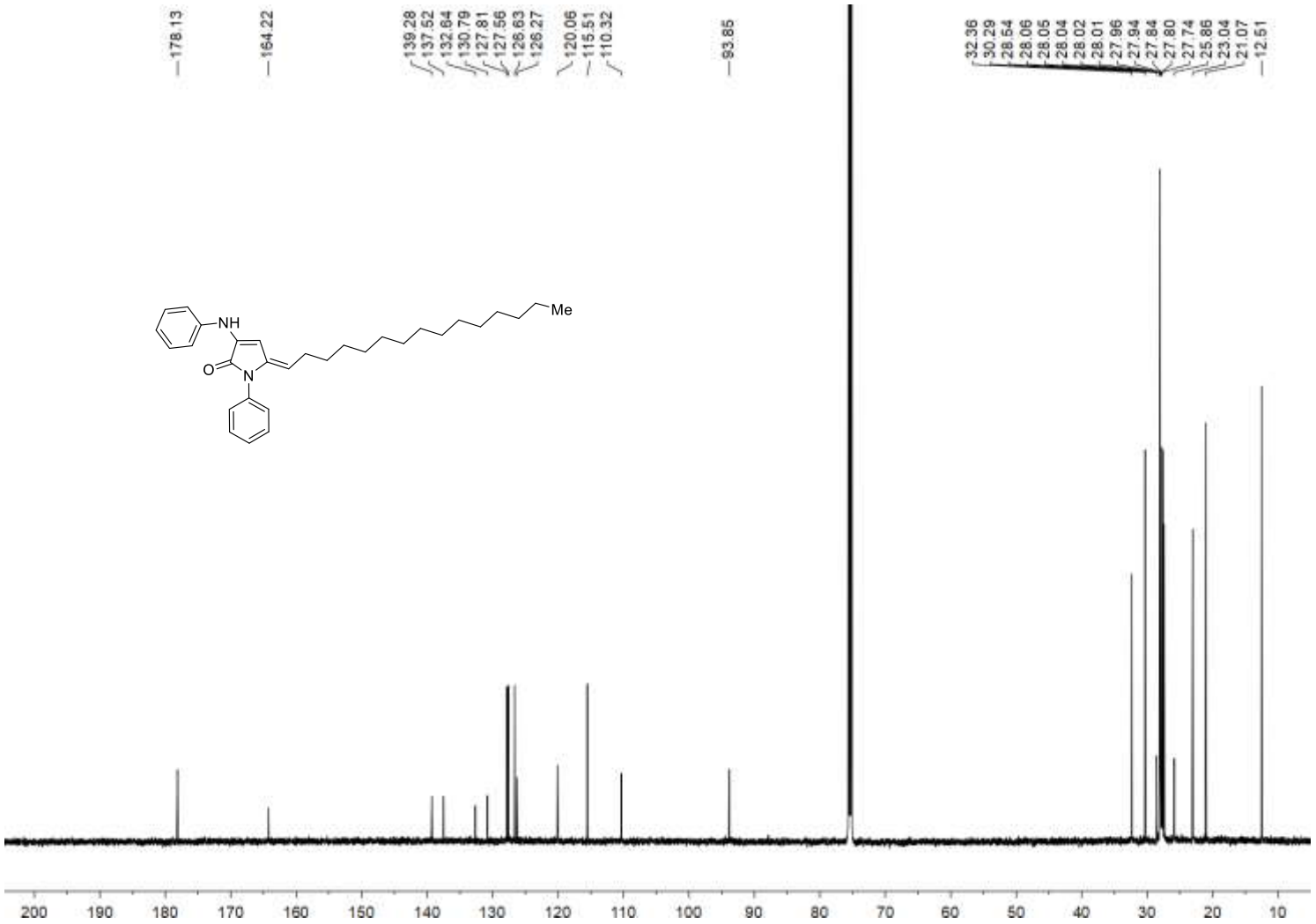


Figure S101. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **7l**

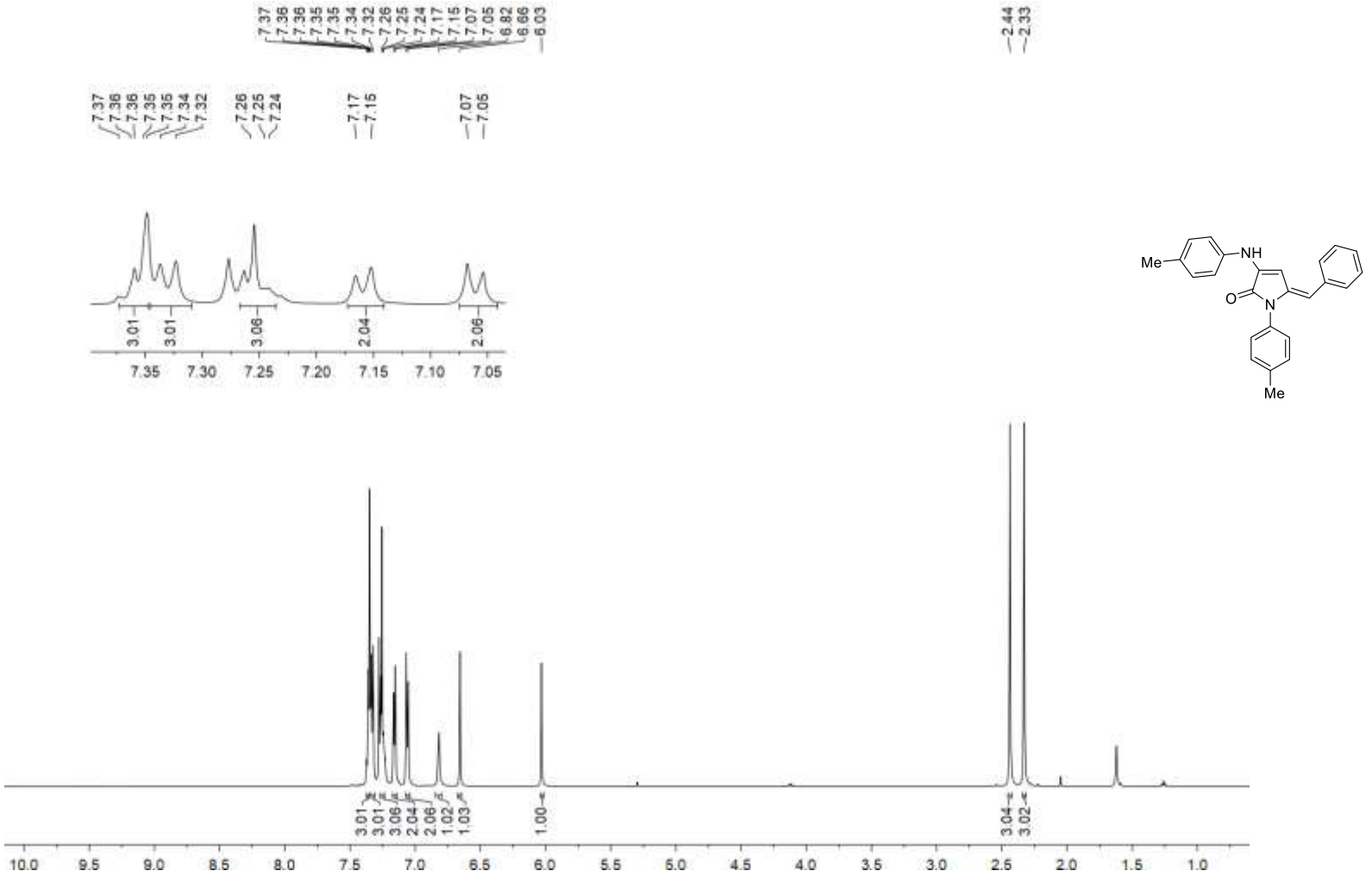


Figure S102. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **7m**

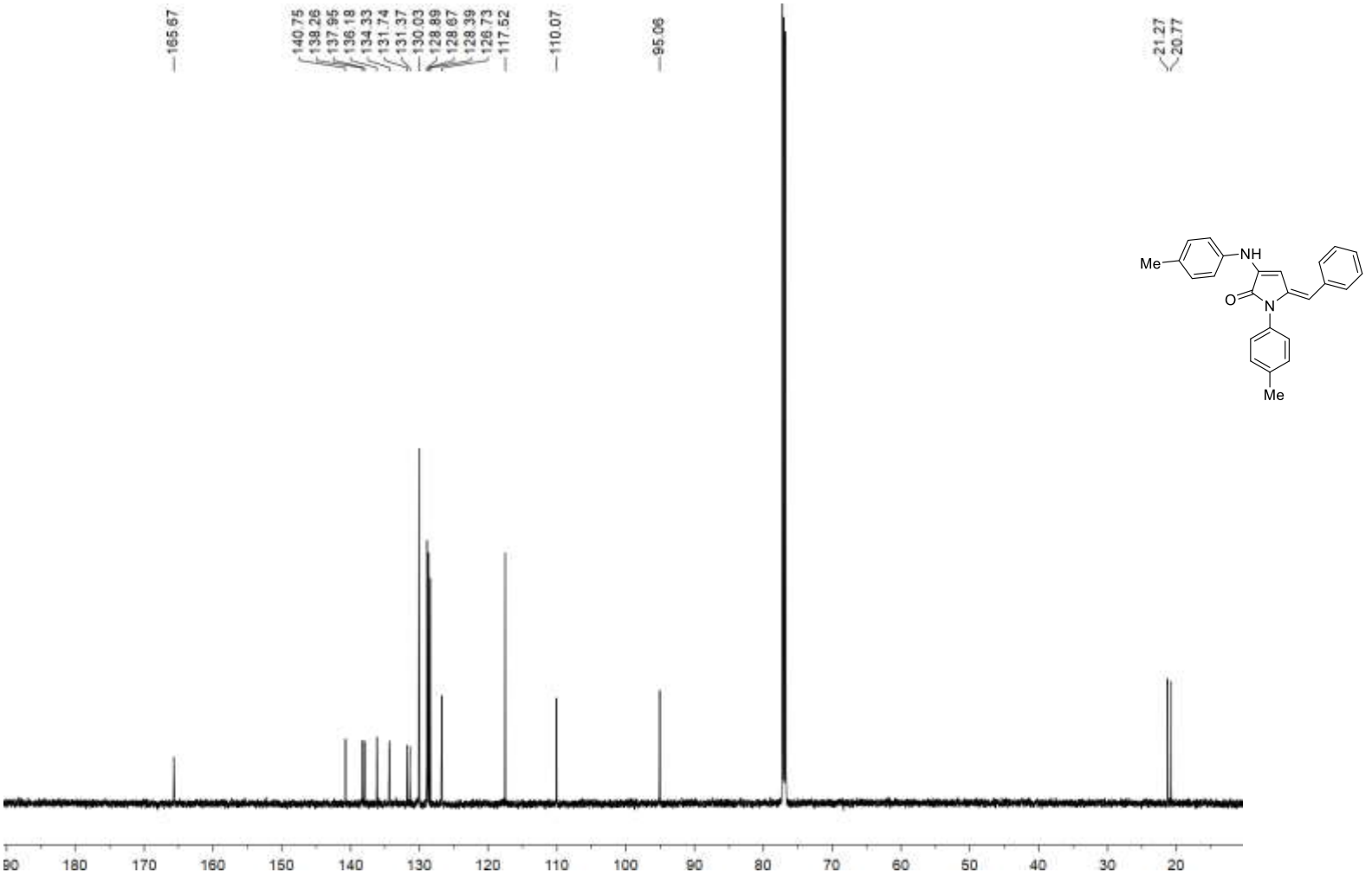


Figure S103. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **7m**

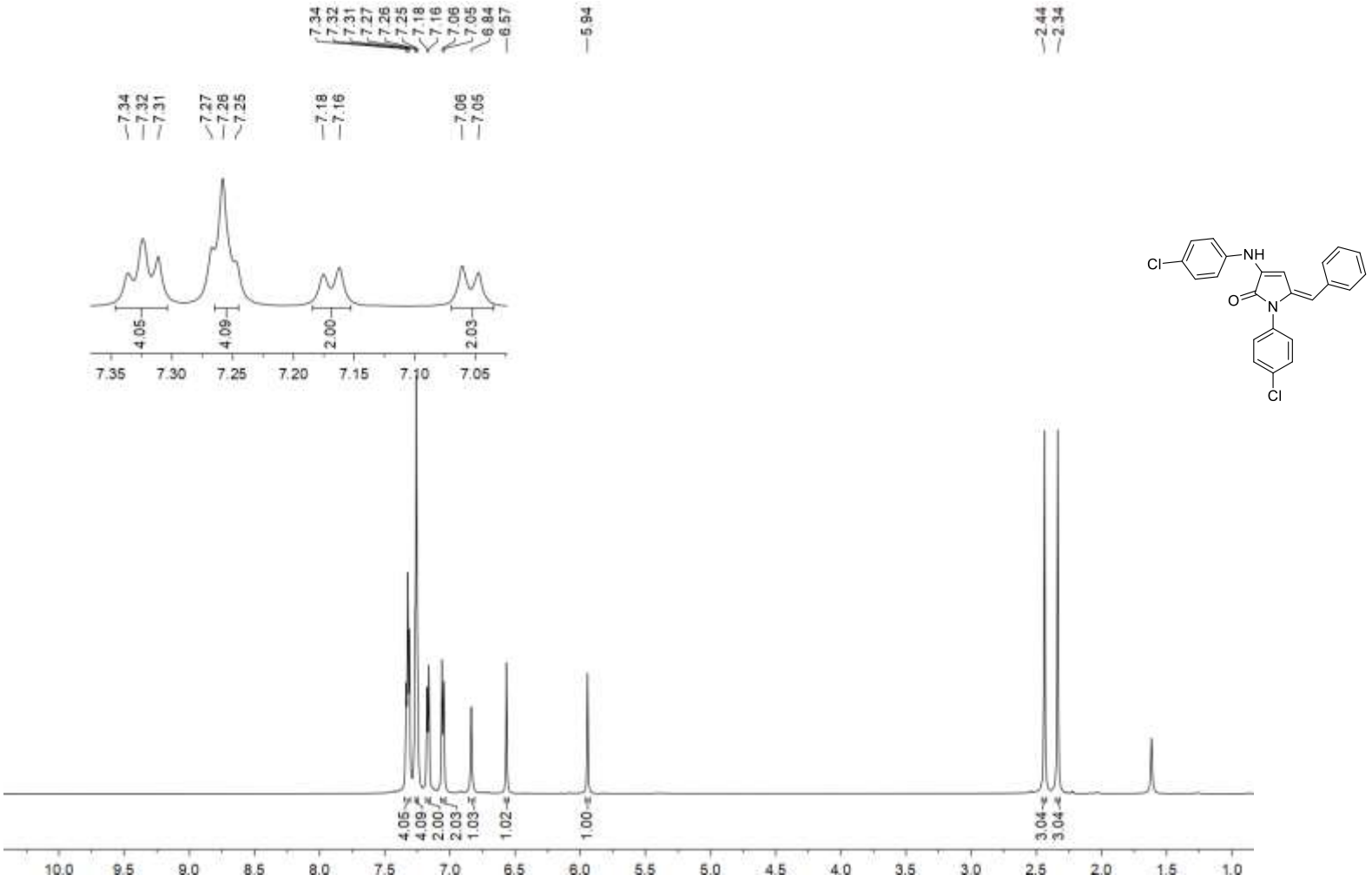


Figure S104. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **7n**

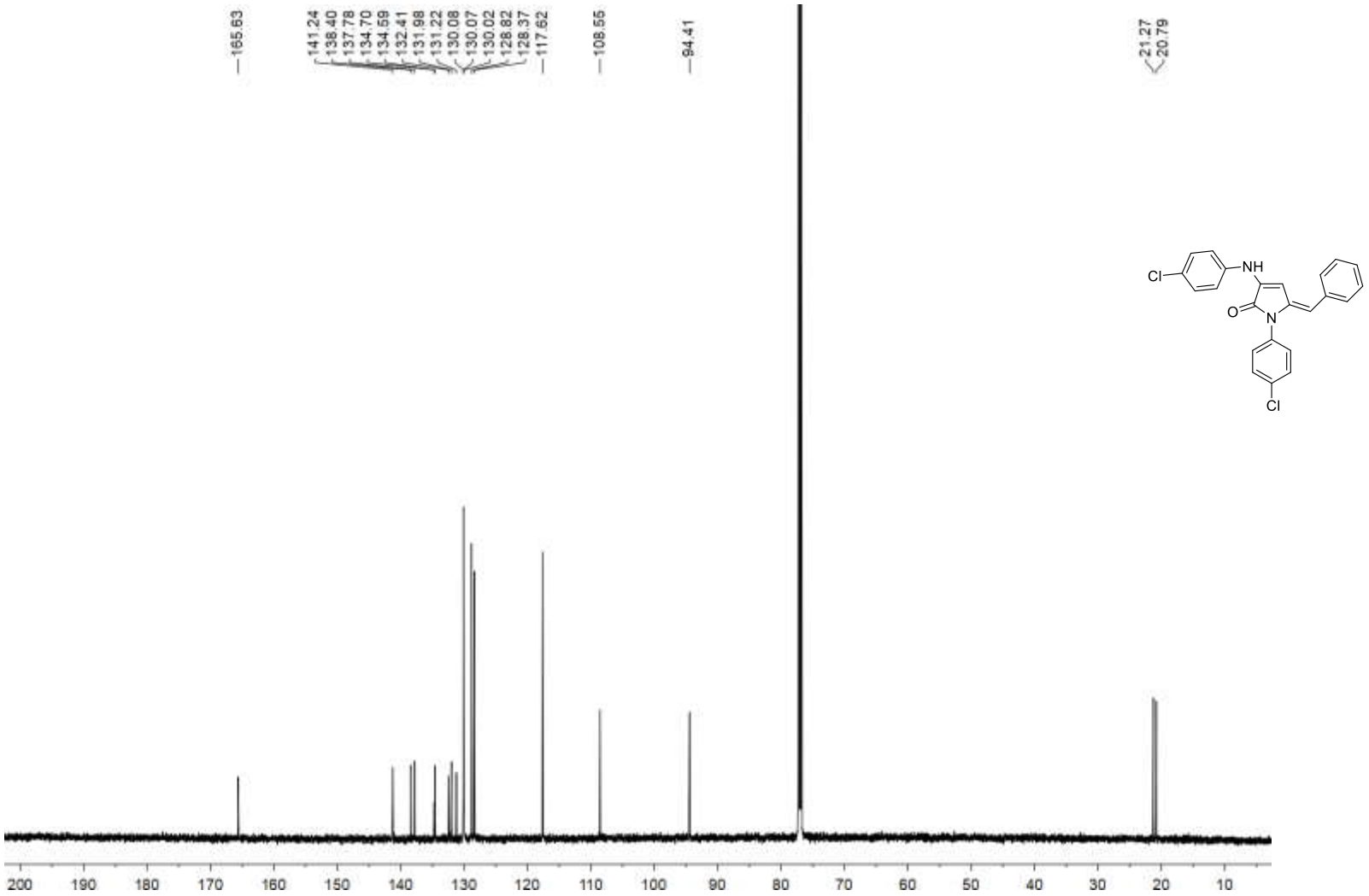


Figure S105. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **7n**

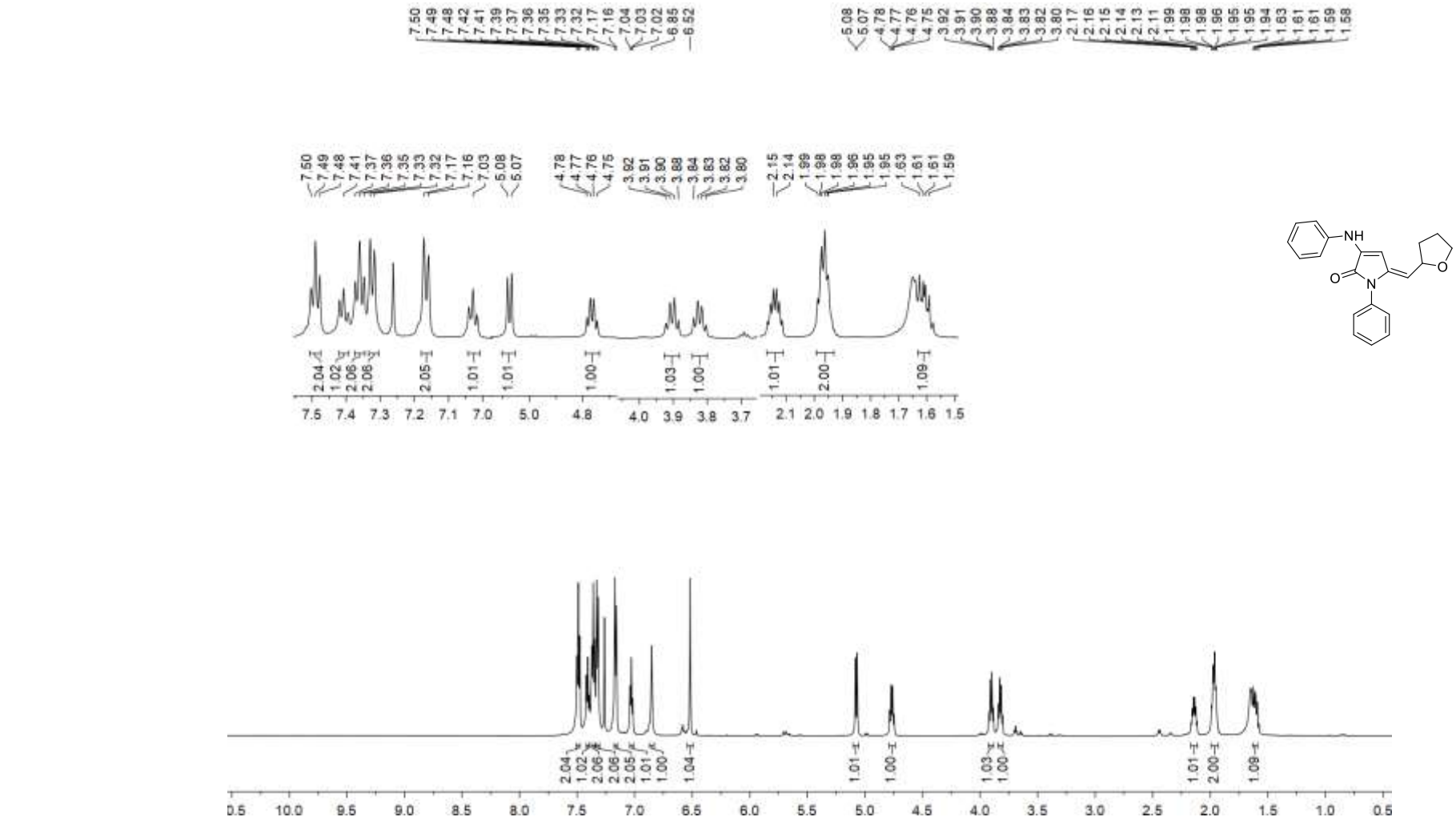


Figure S106. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **8a**

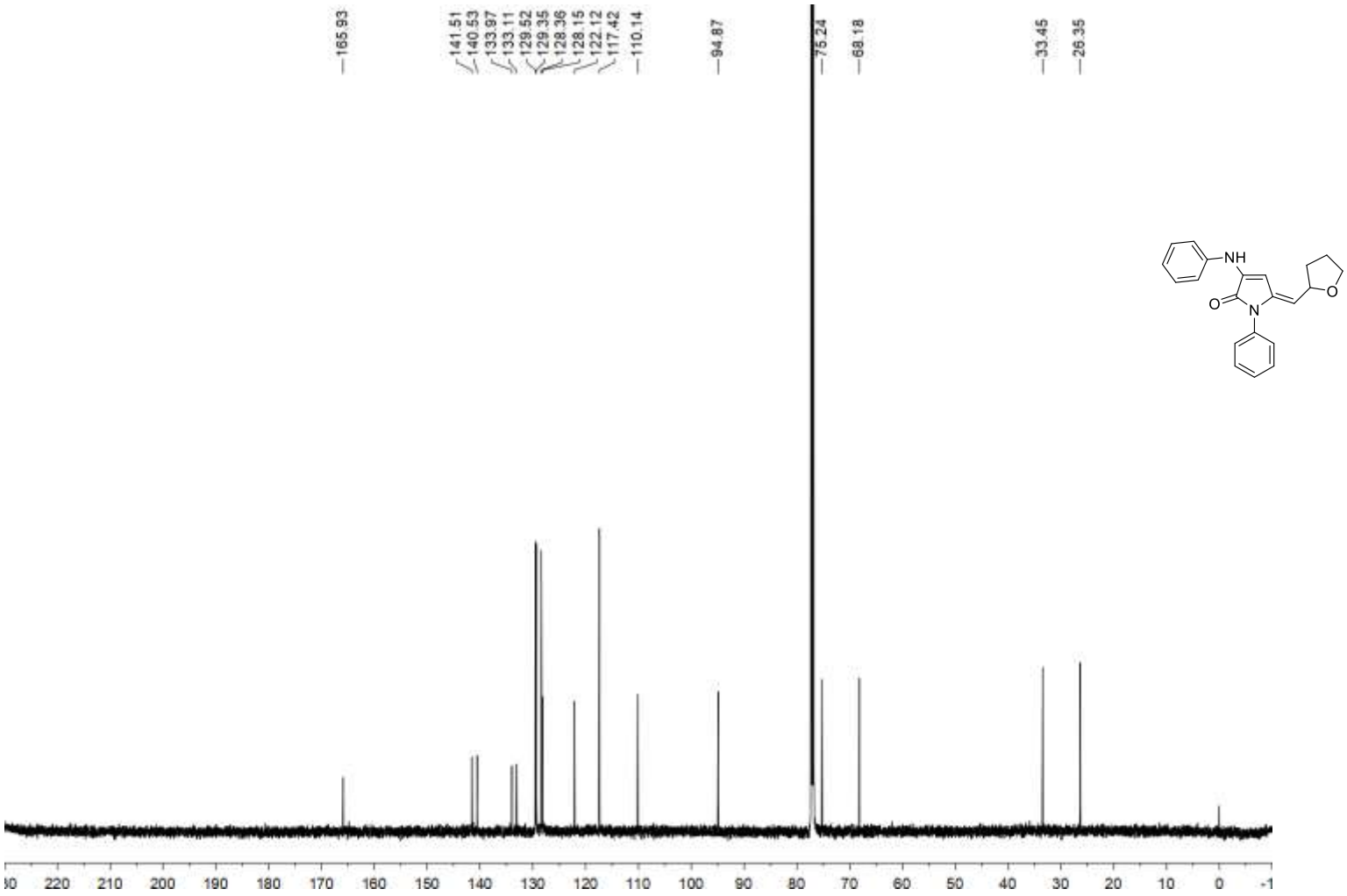


Figure S107. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **8a**

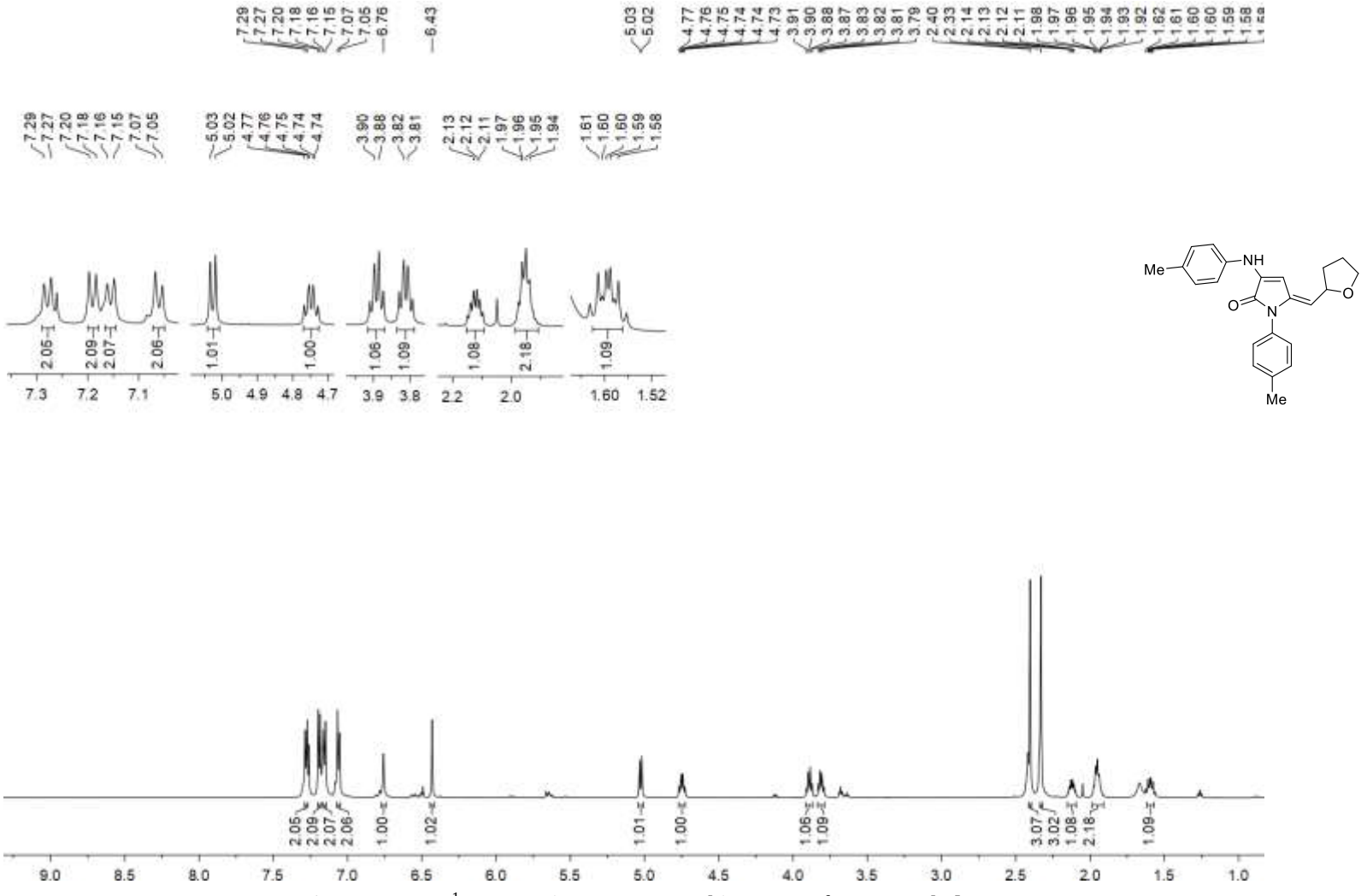


Figure S108. **^{1}H NMR** (600 MHz, $CDCl_3$) spectra of compound **8b**

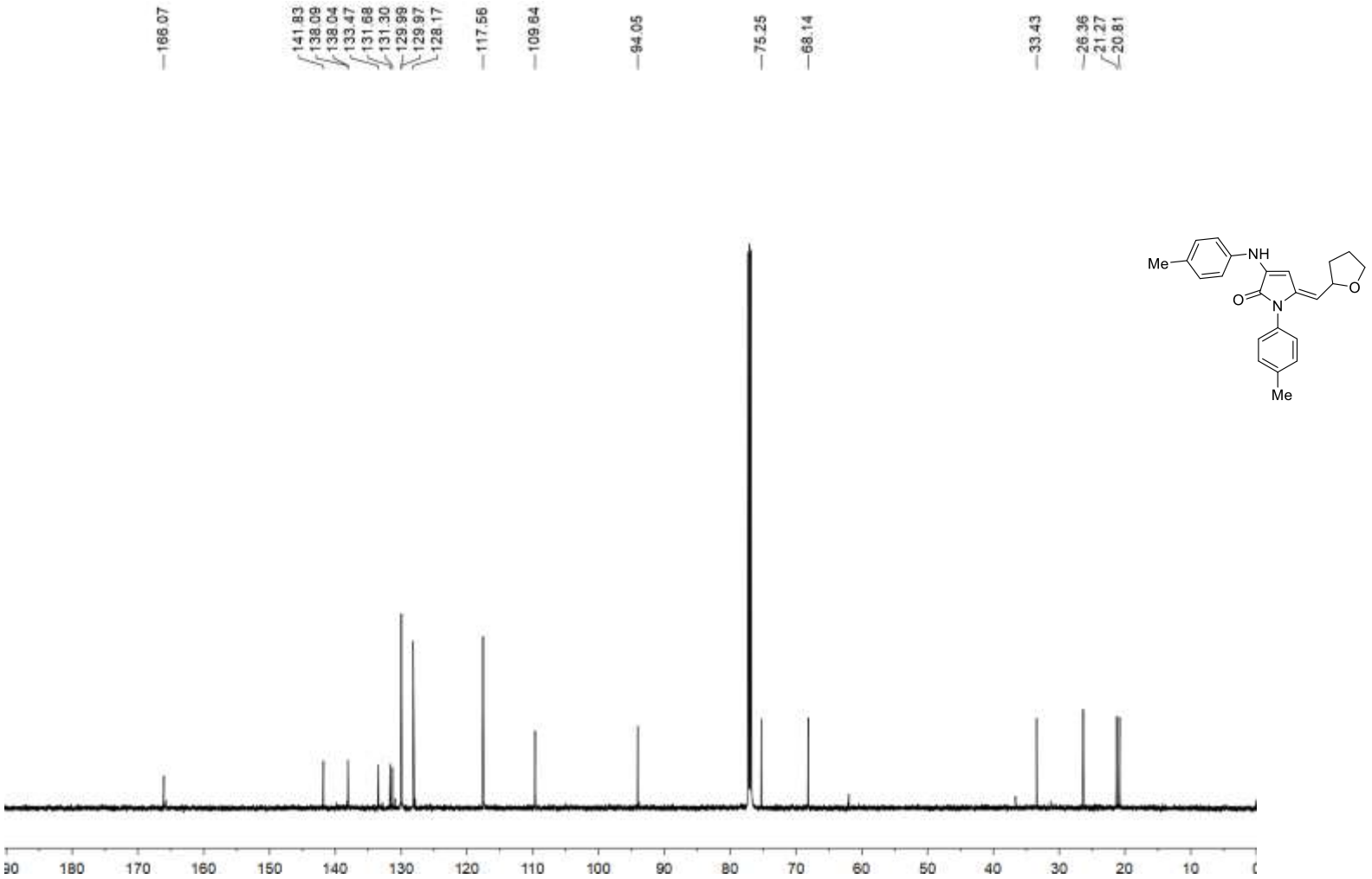


Figure S109. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **8b**

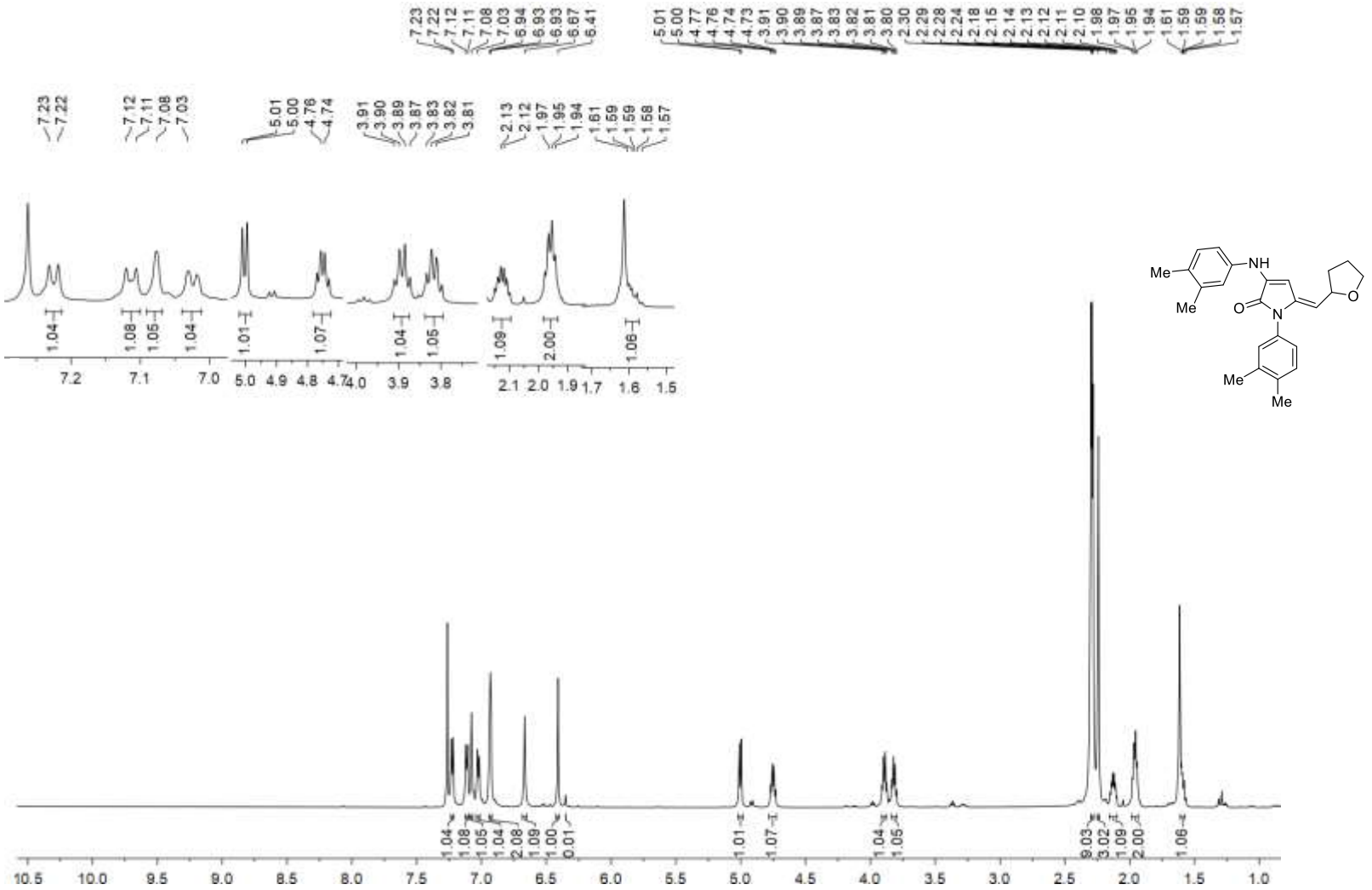


Figure S110. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **8c**

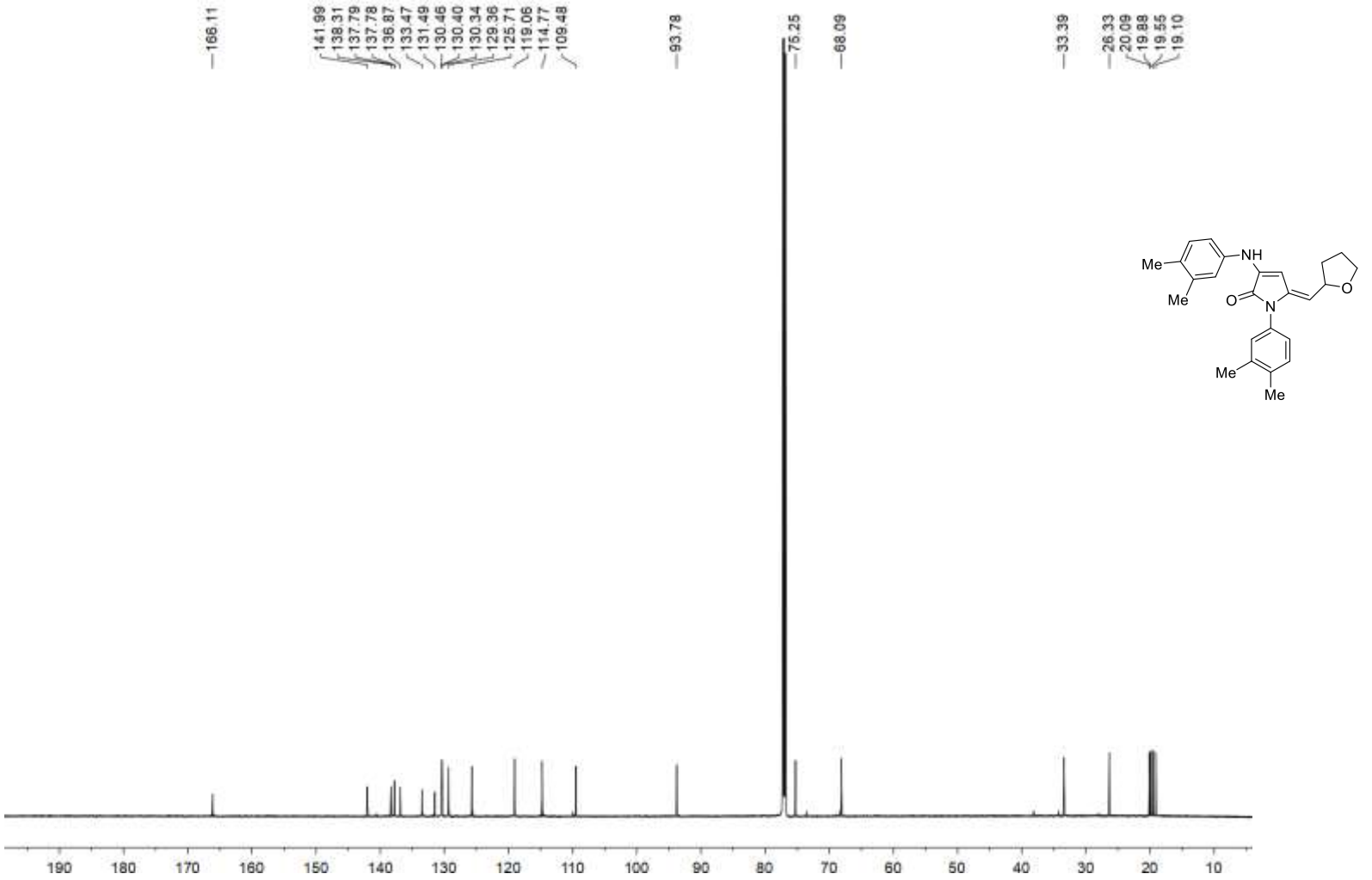


Figure S111. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **8c**

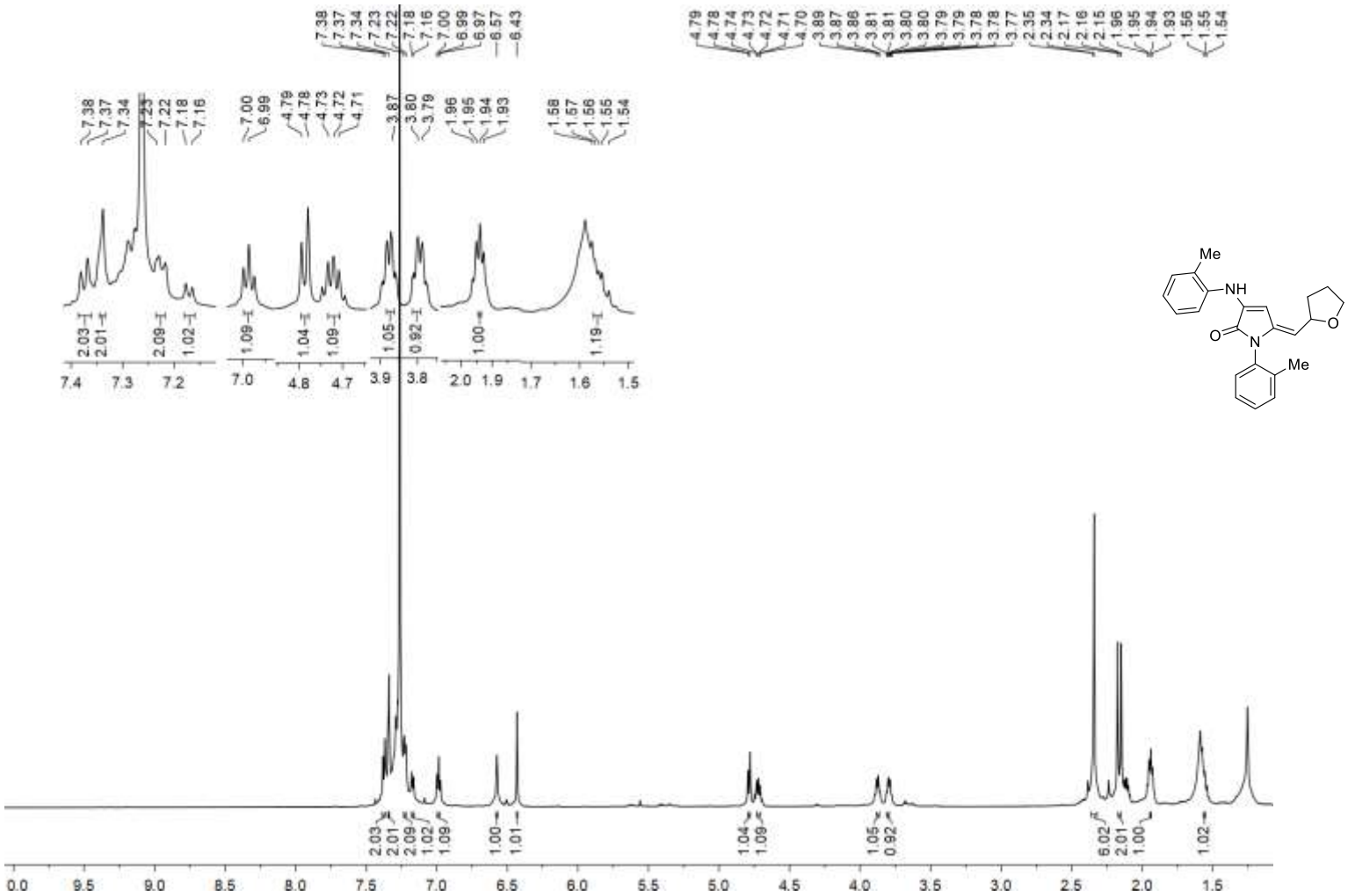


Figure S112. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **8d**

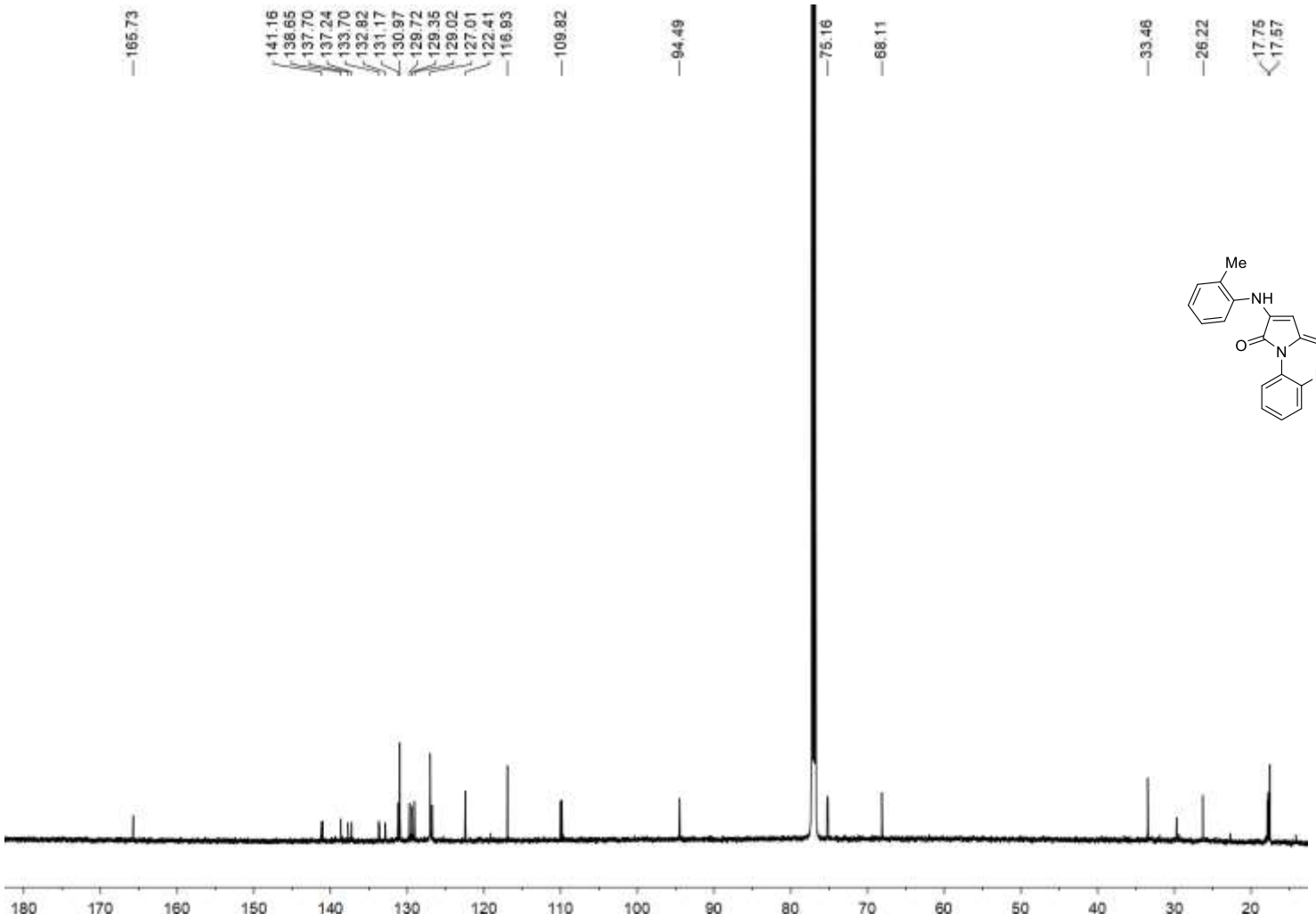


Figure S113. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **8d**

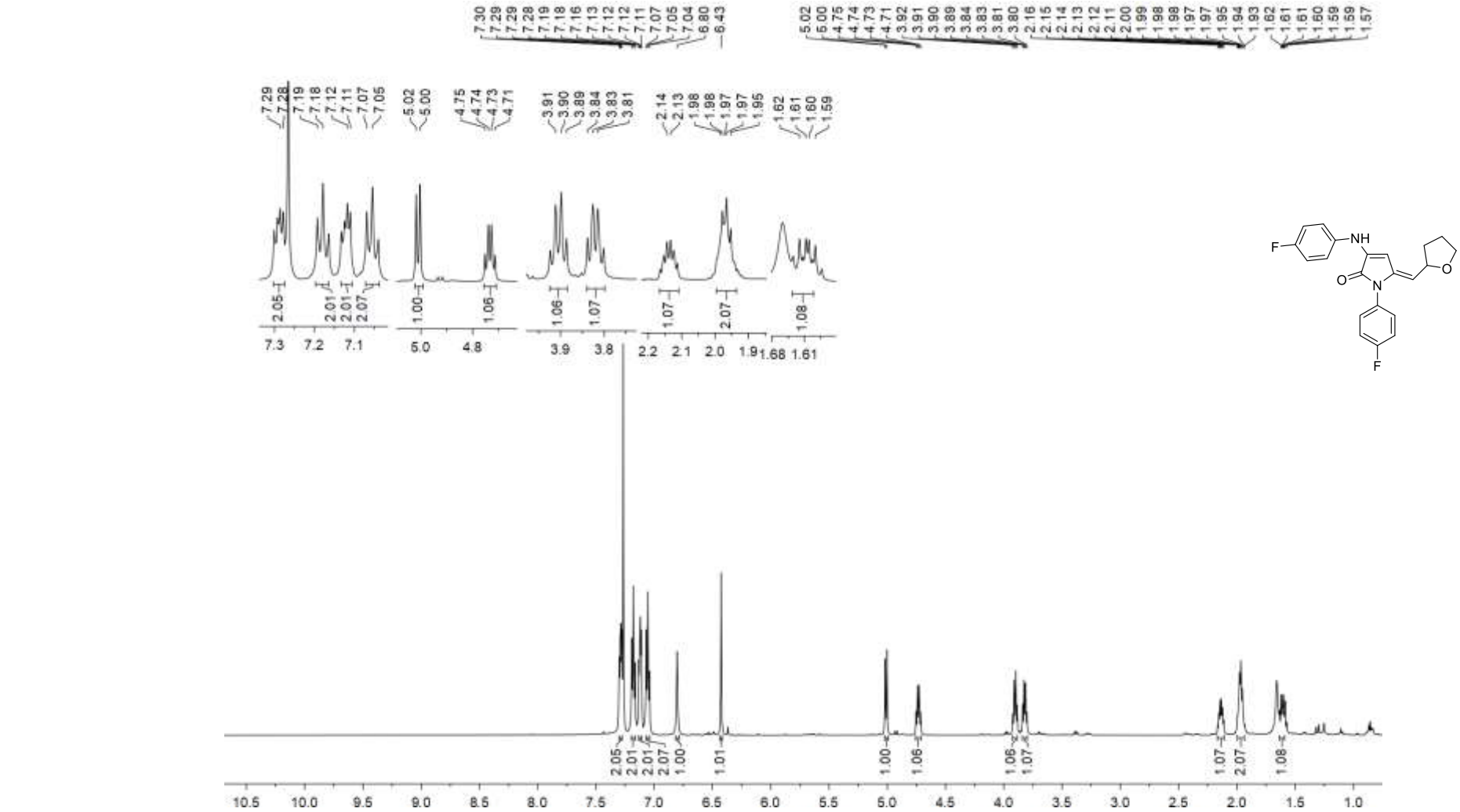


Figure S114. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **8e**

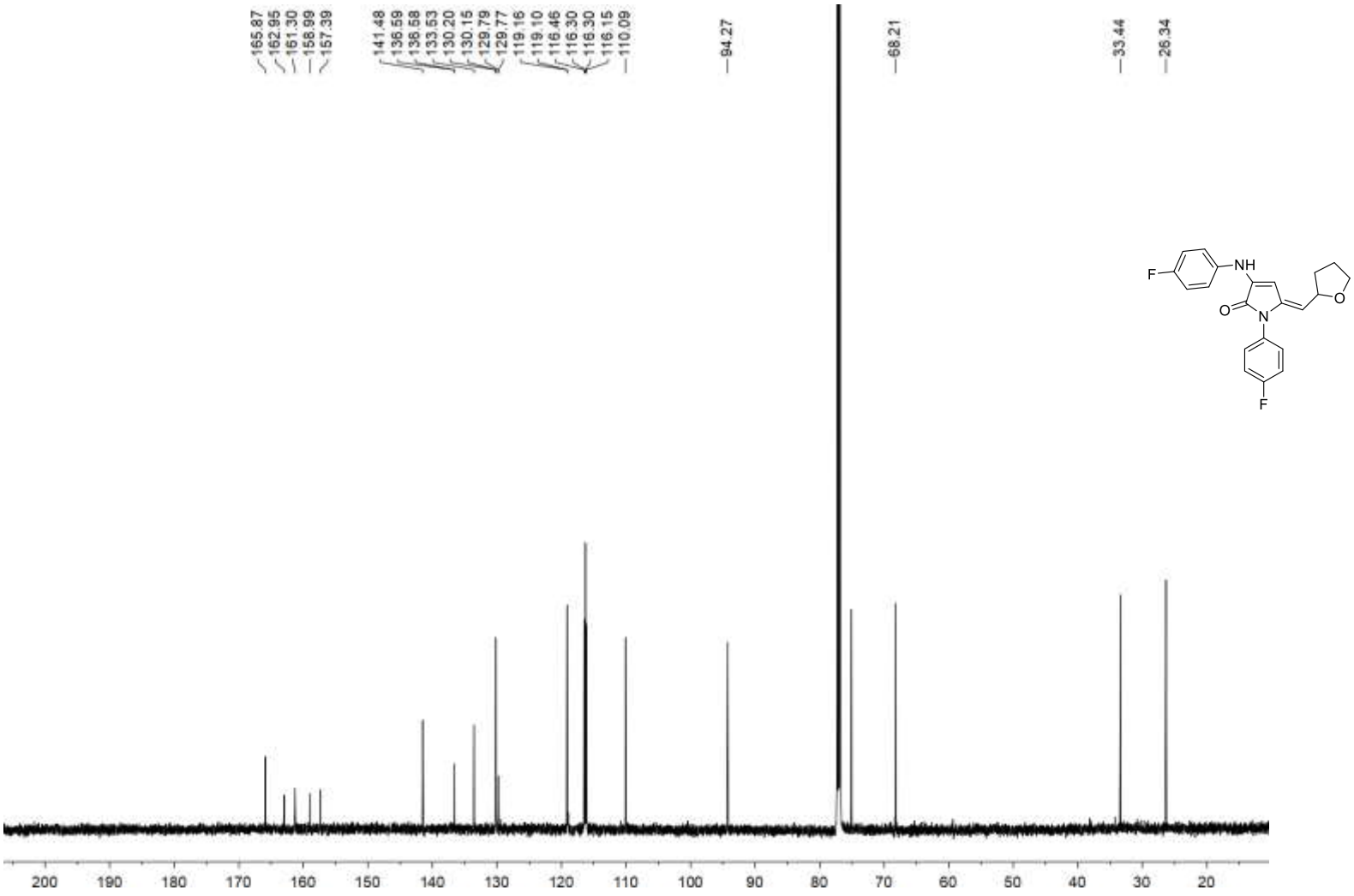


Figure S115. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **8e**

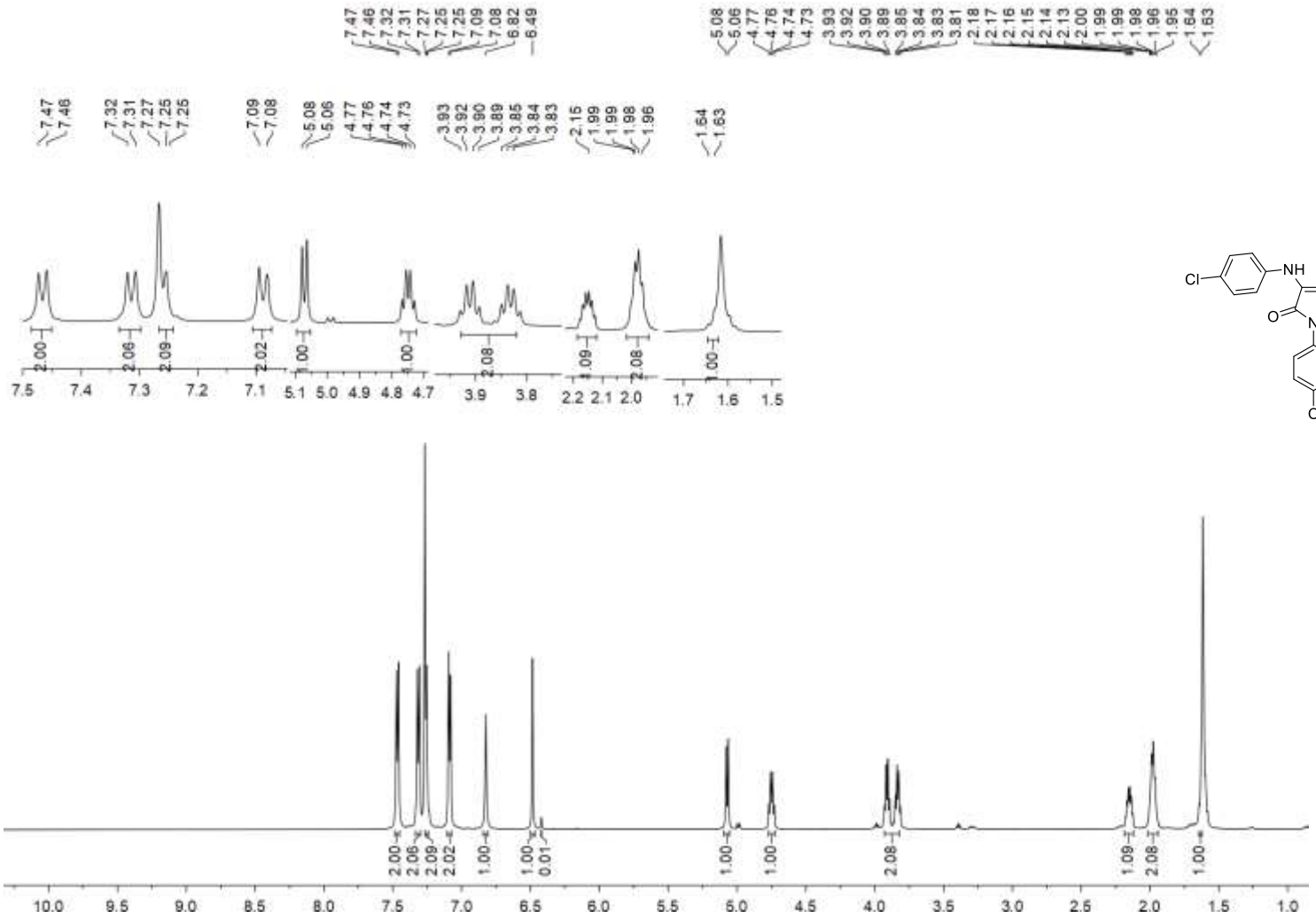


Figure S116. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **8f**

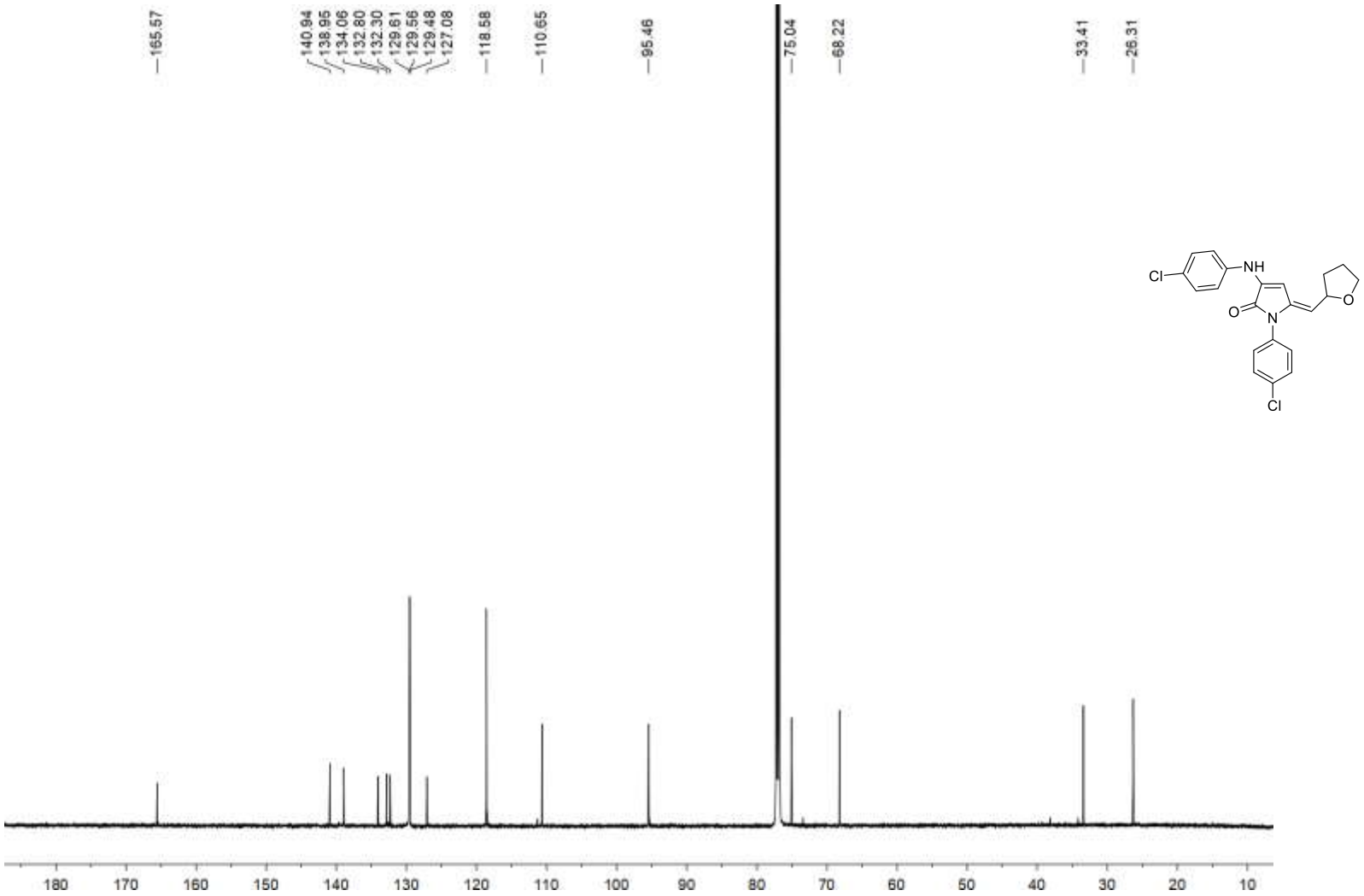


Figure S117. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **8f**

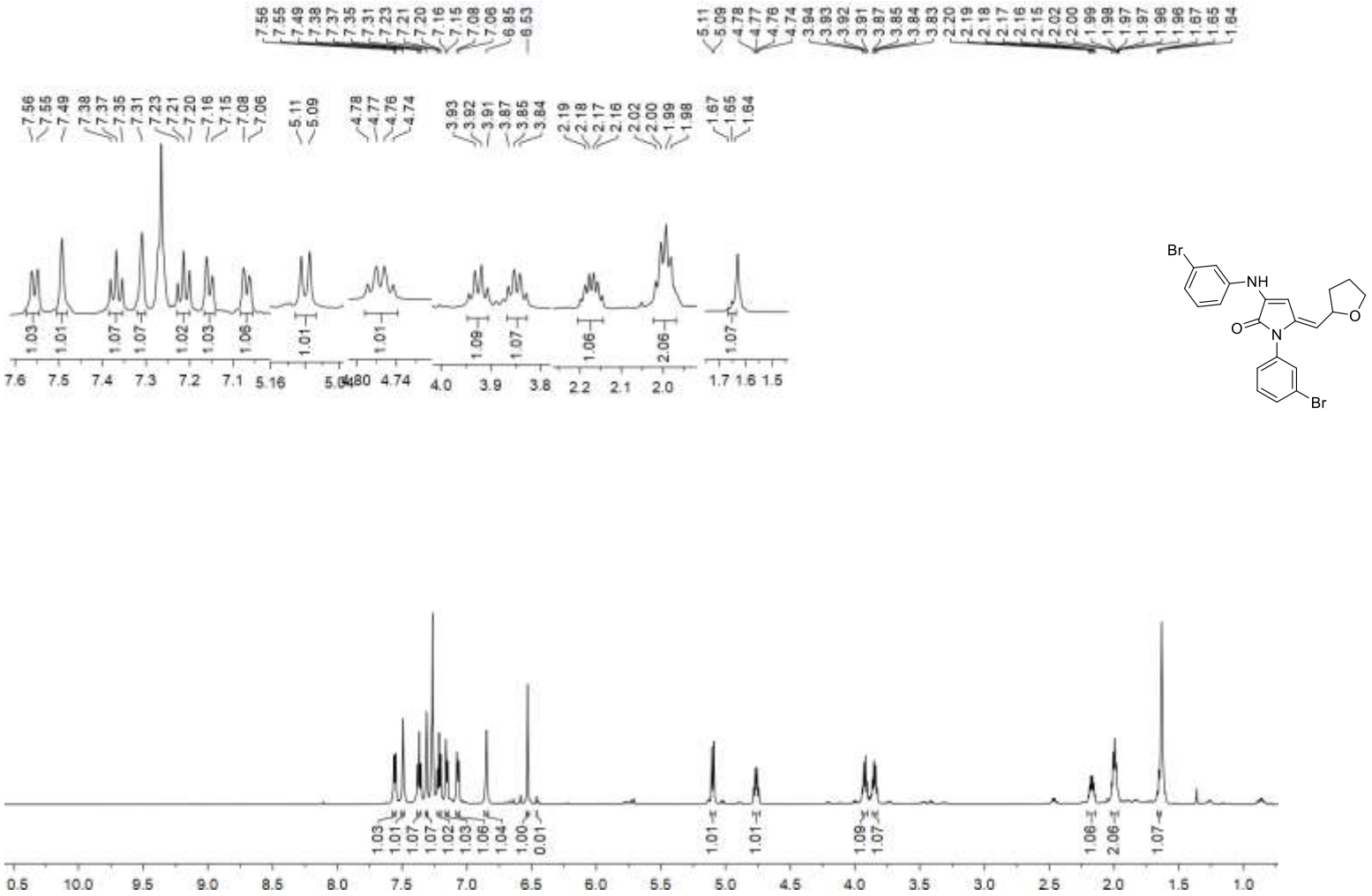


Figure S118. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **8g**

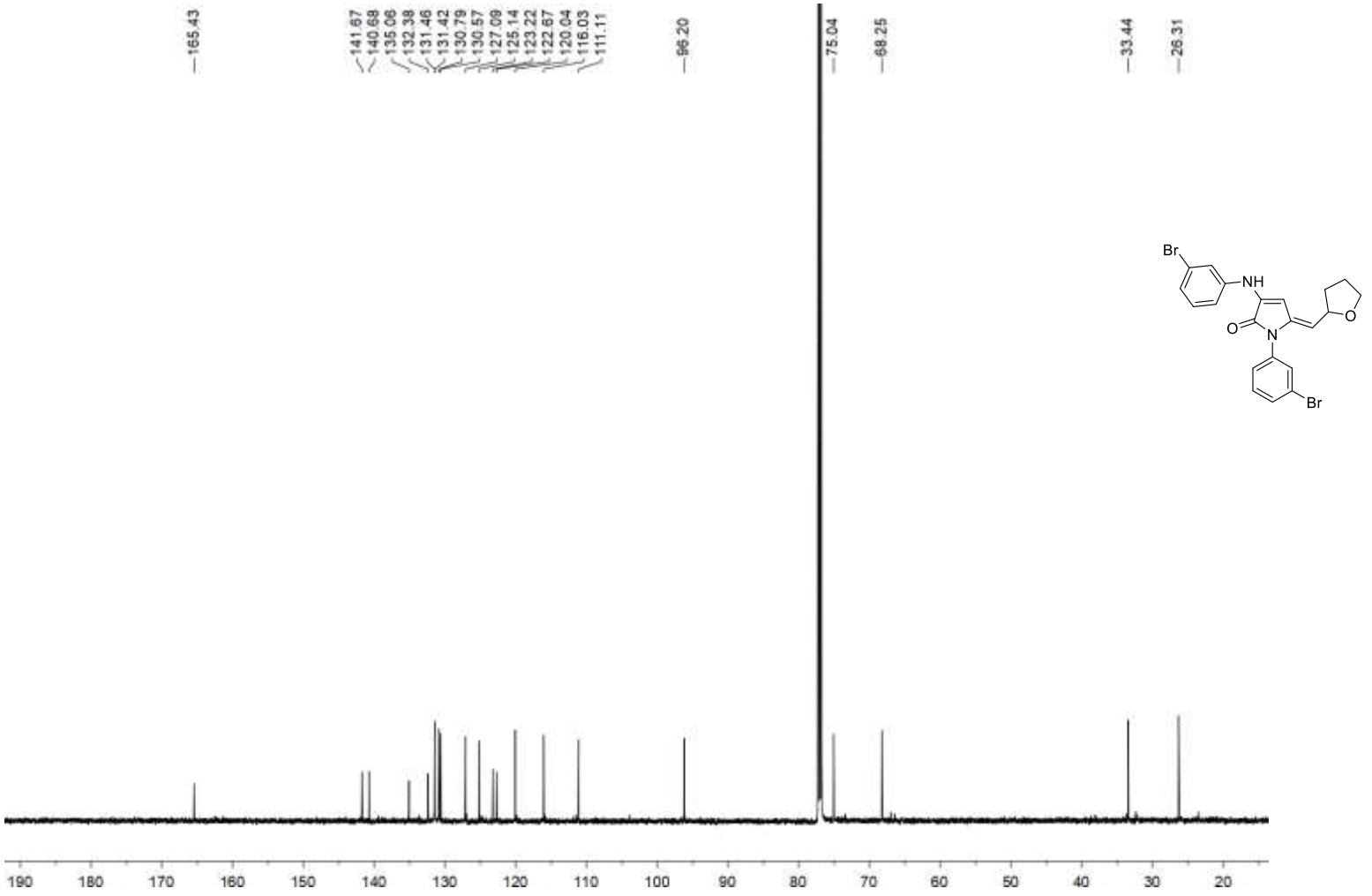


Figure S119. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **8g**

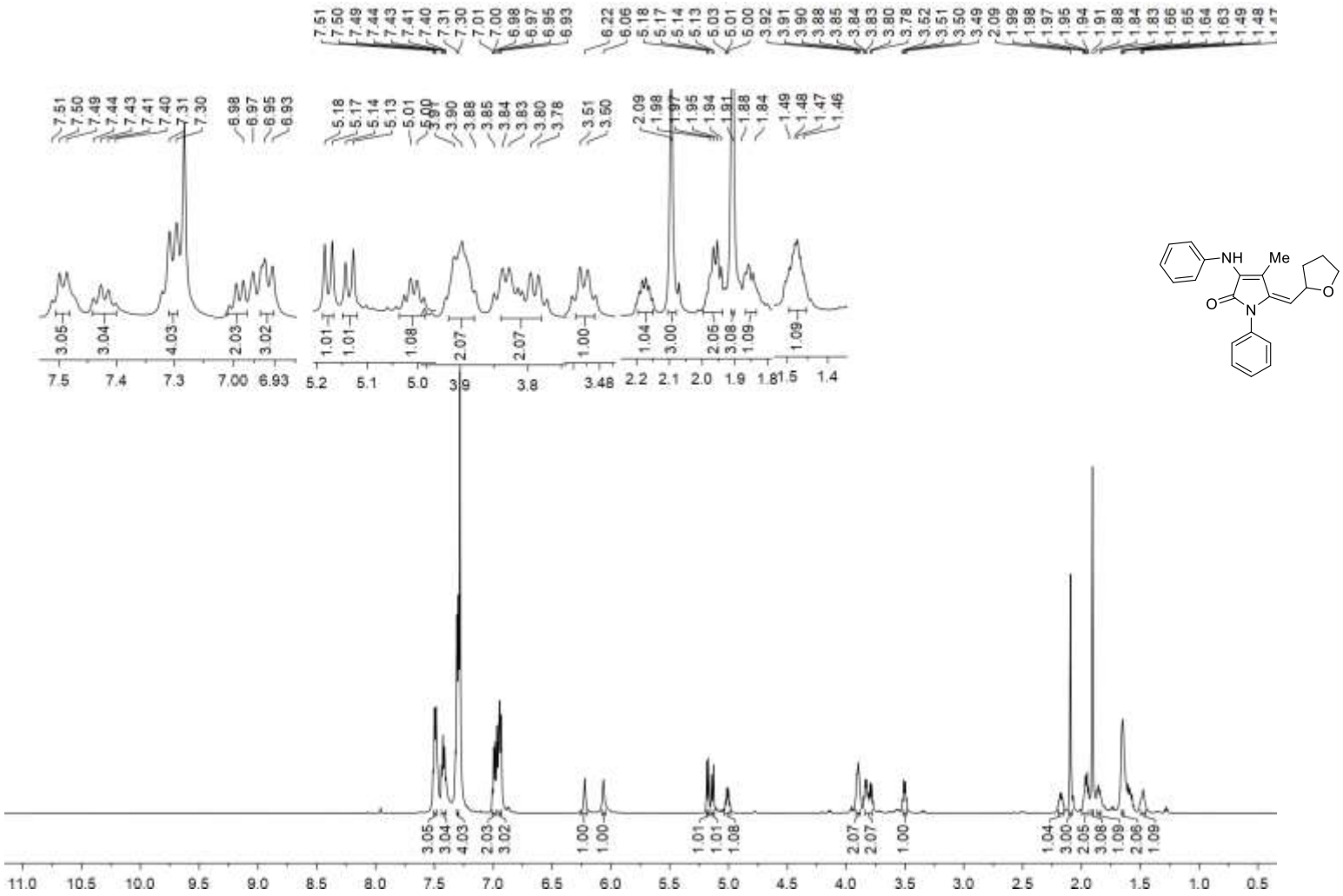


Figure S120. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **8h**

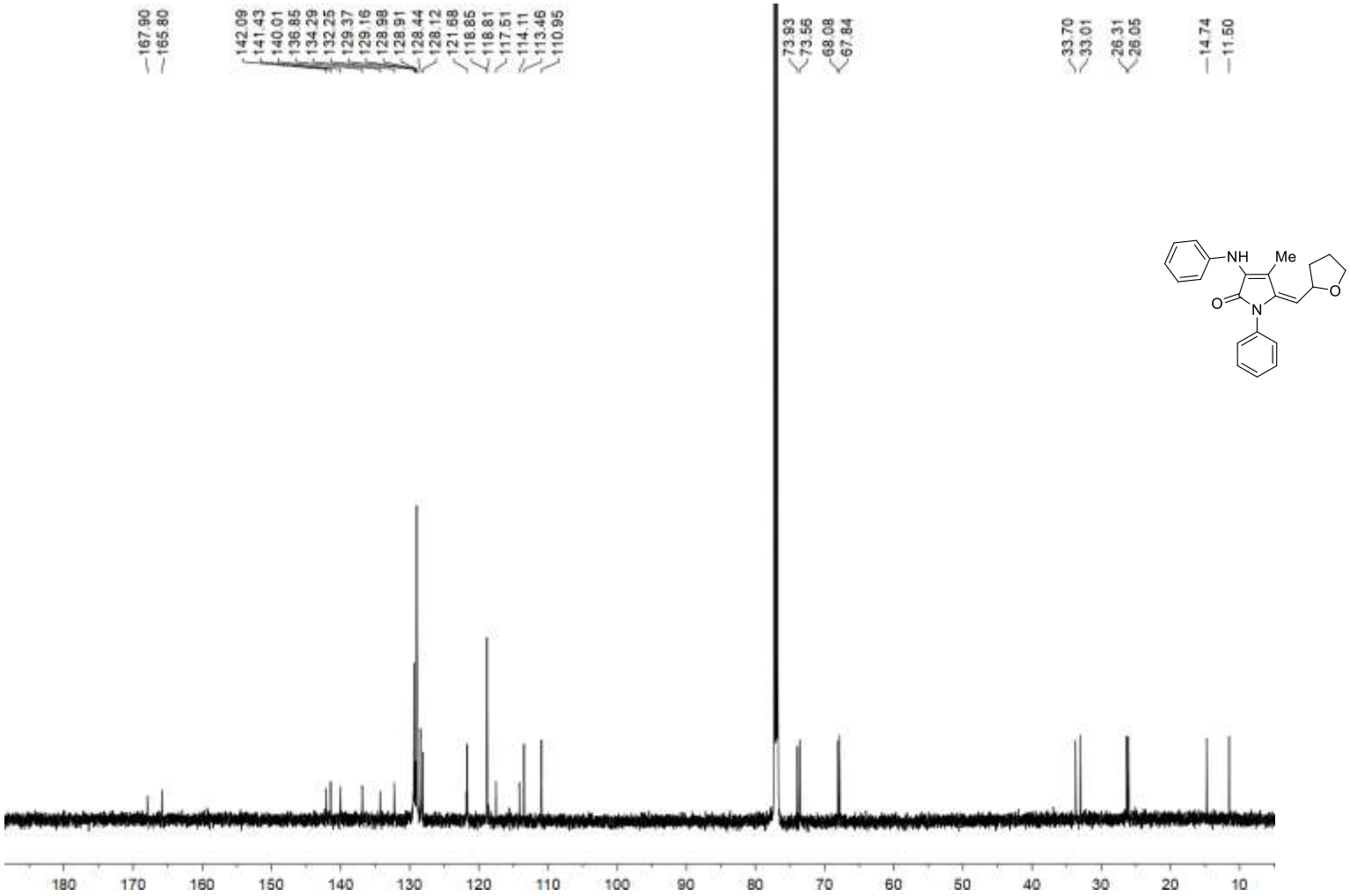


Figure S121. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **8h**

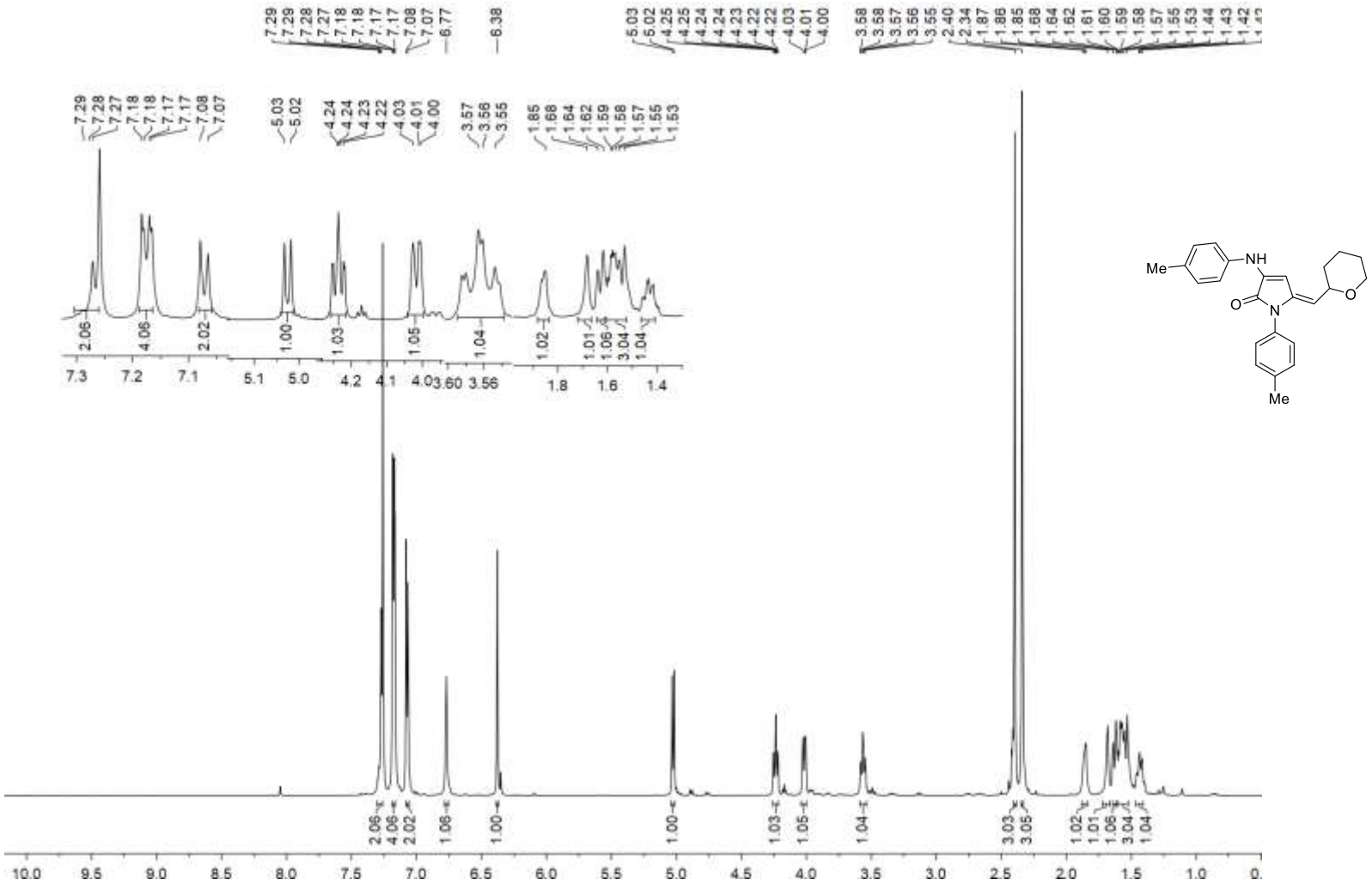


Figure S122. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **8i**

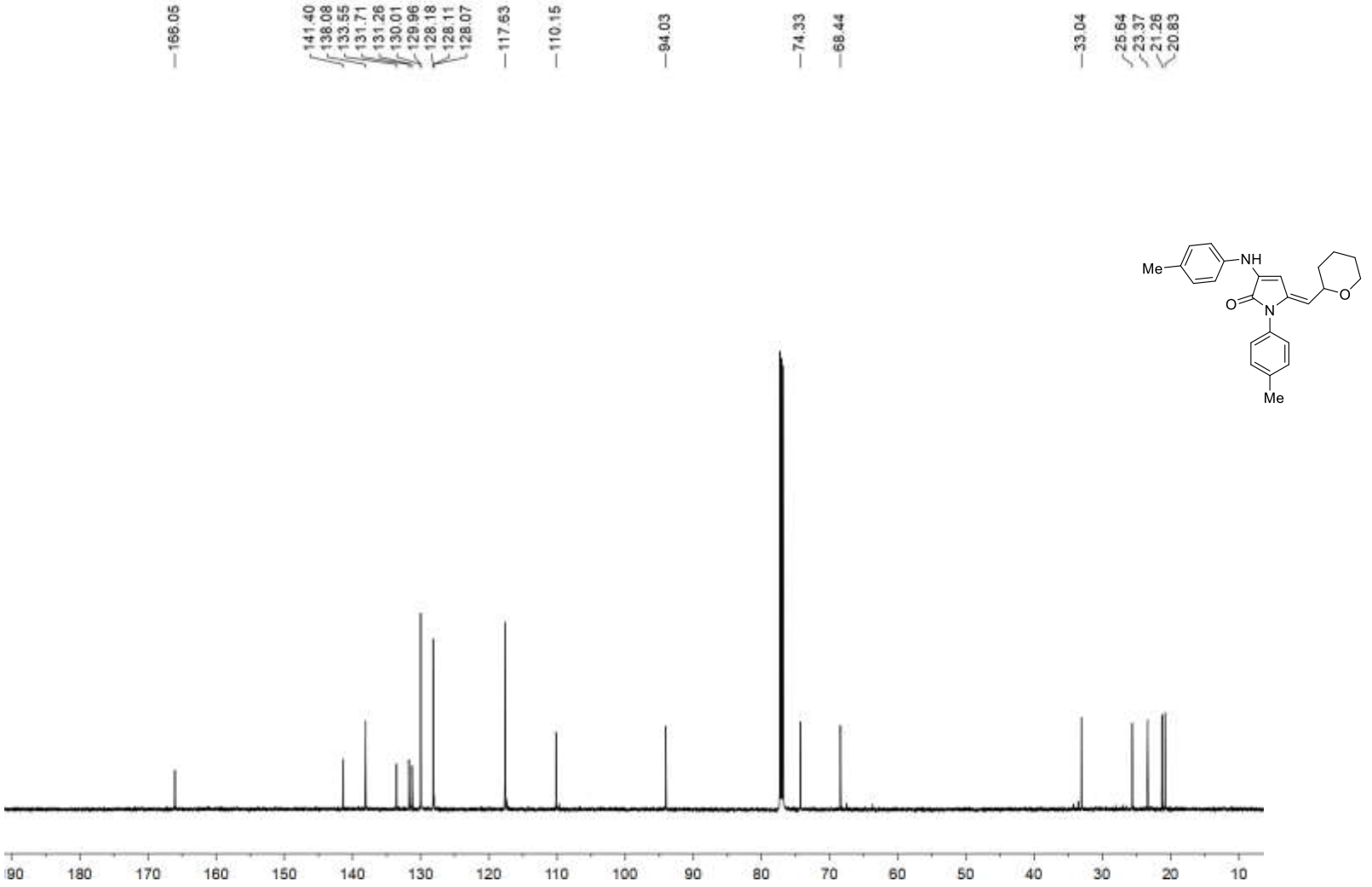


Figure S123. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **8i**

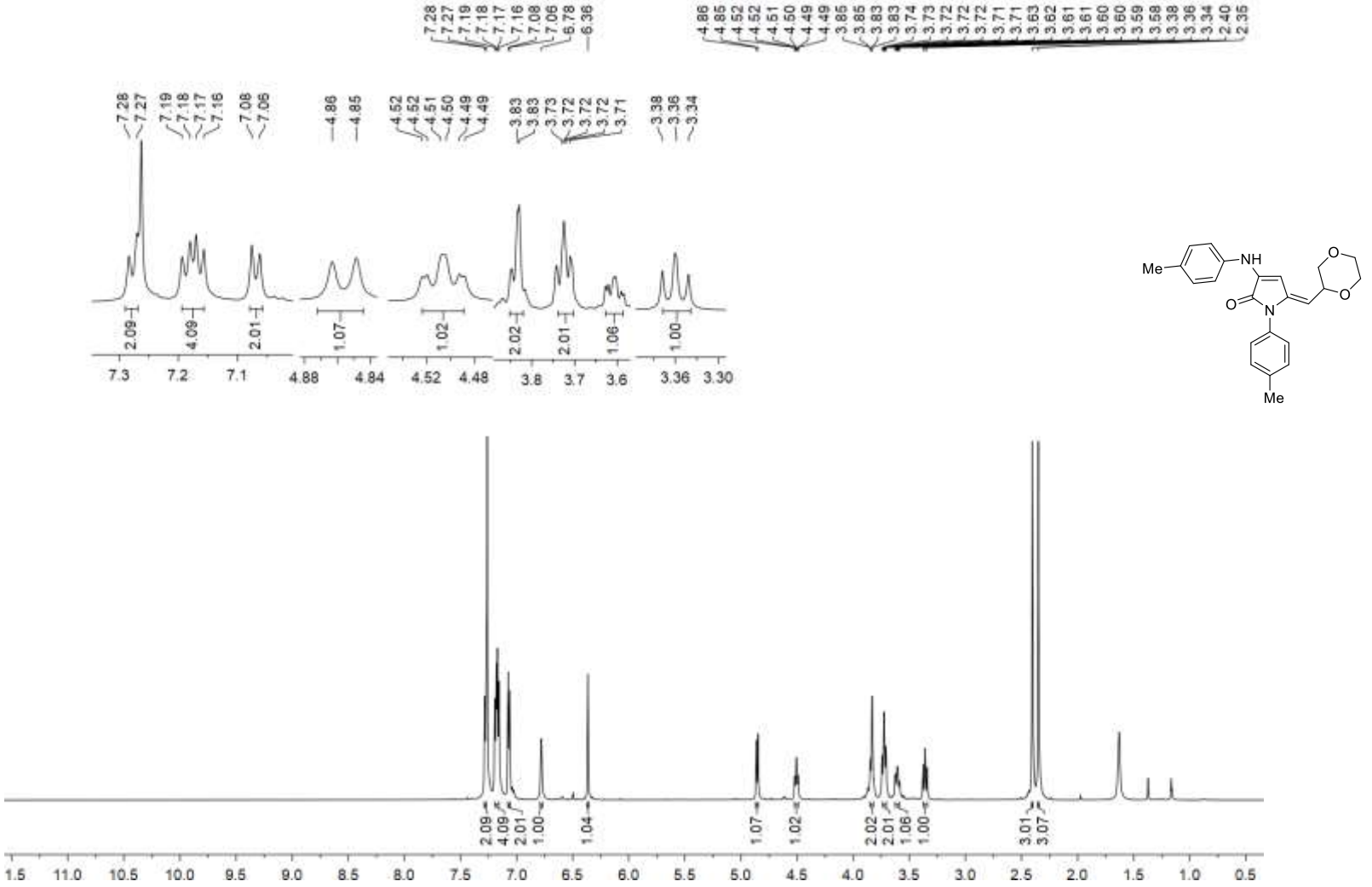


Figure S124. **^{1}H NMR** (600 MHz, $CDCl_3$) spectra of compound **8j**

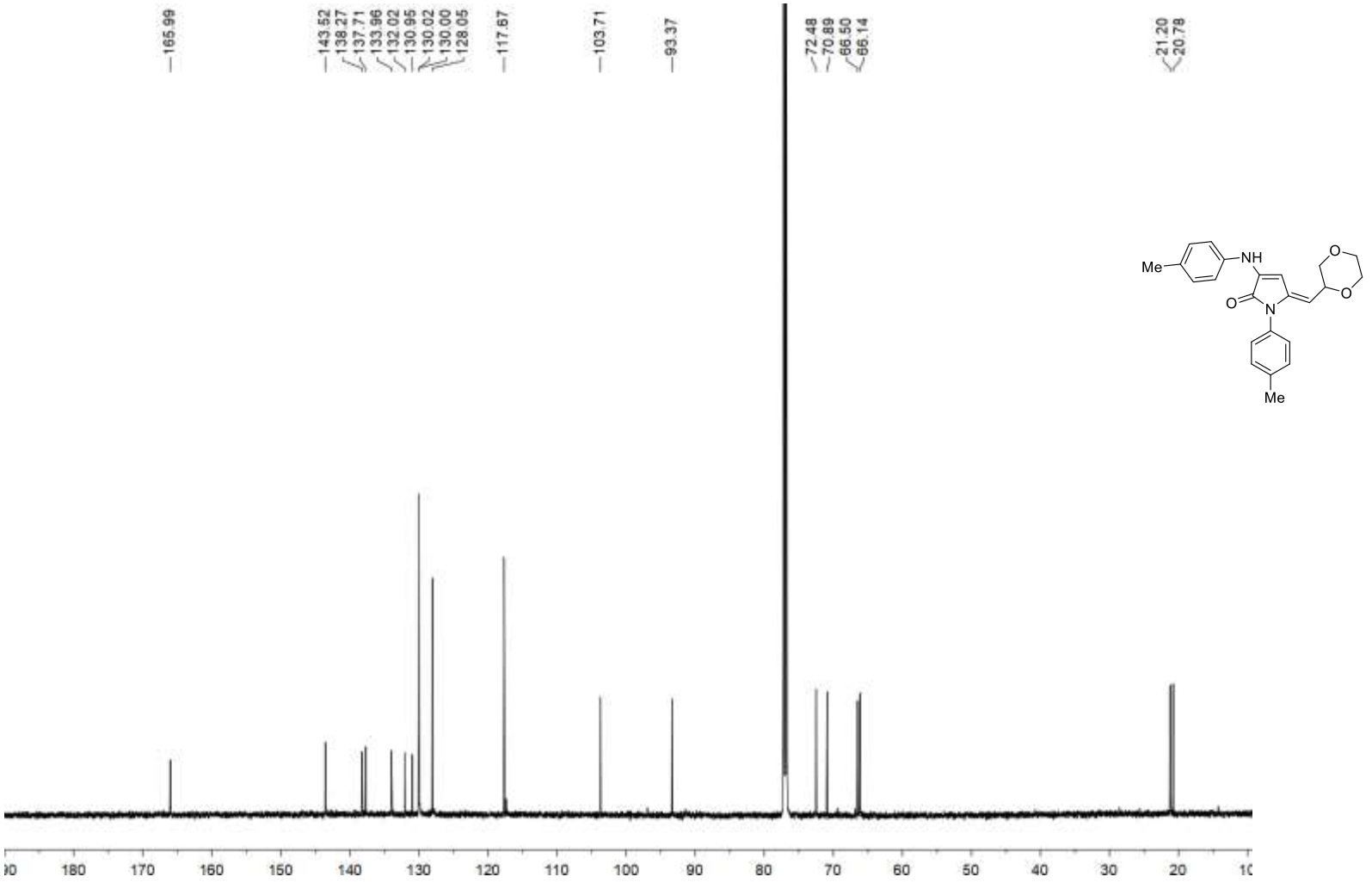


Figure S125. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **8j**

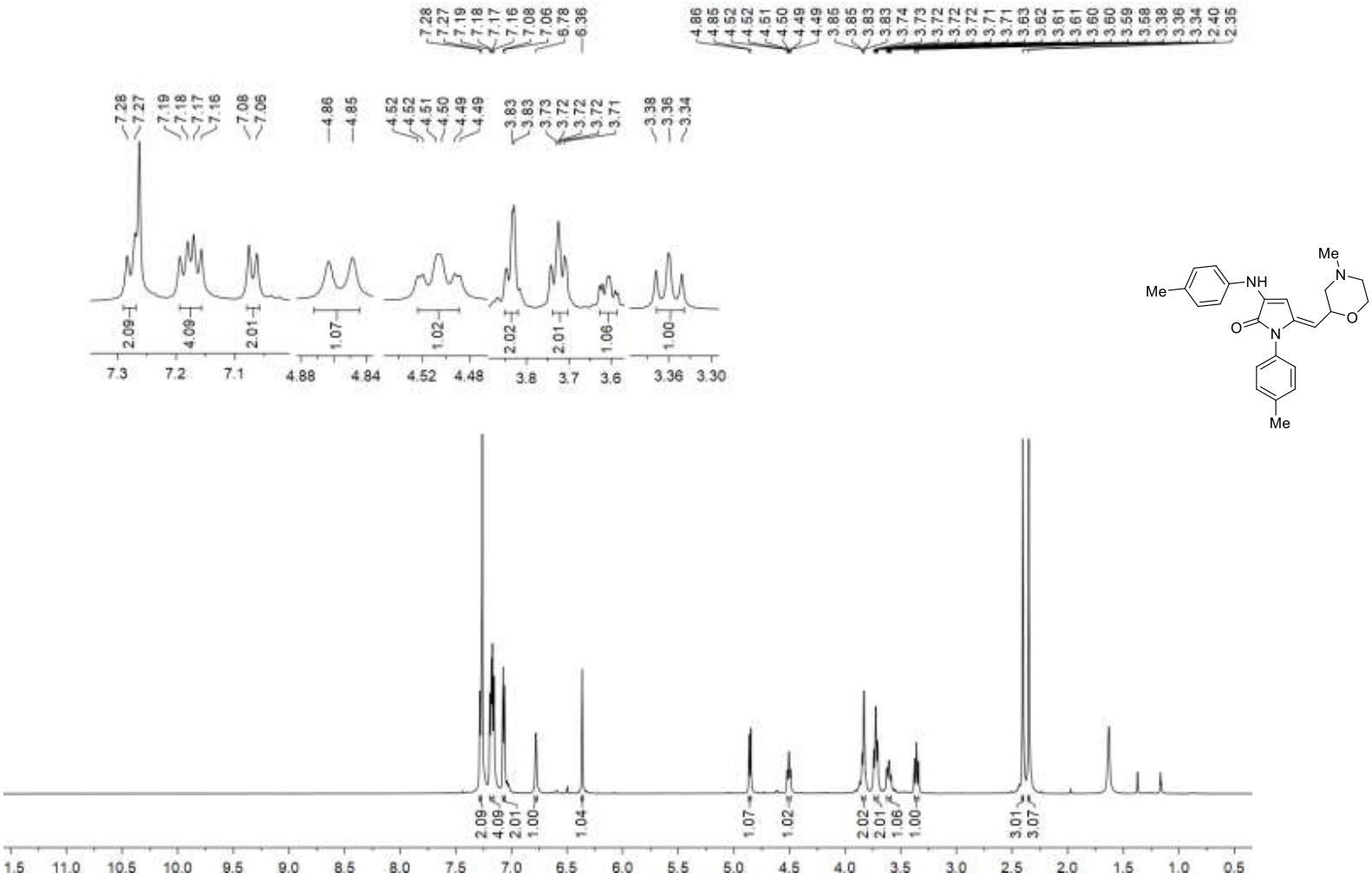


Figure S126. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **8k**

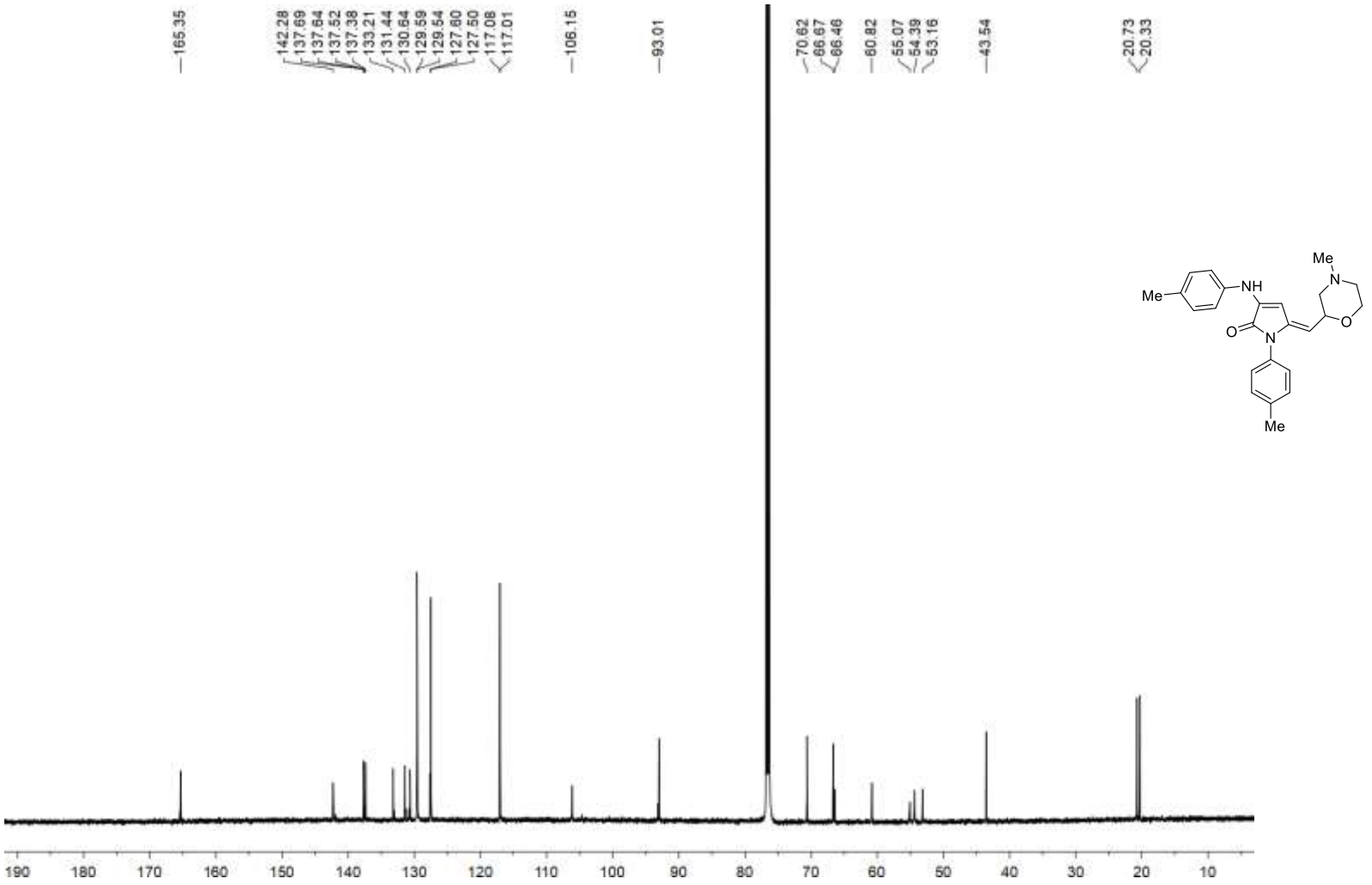


Figure S127. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **8k**

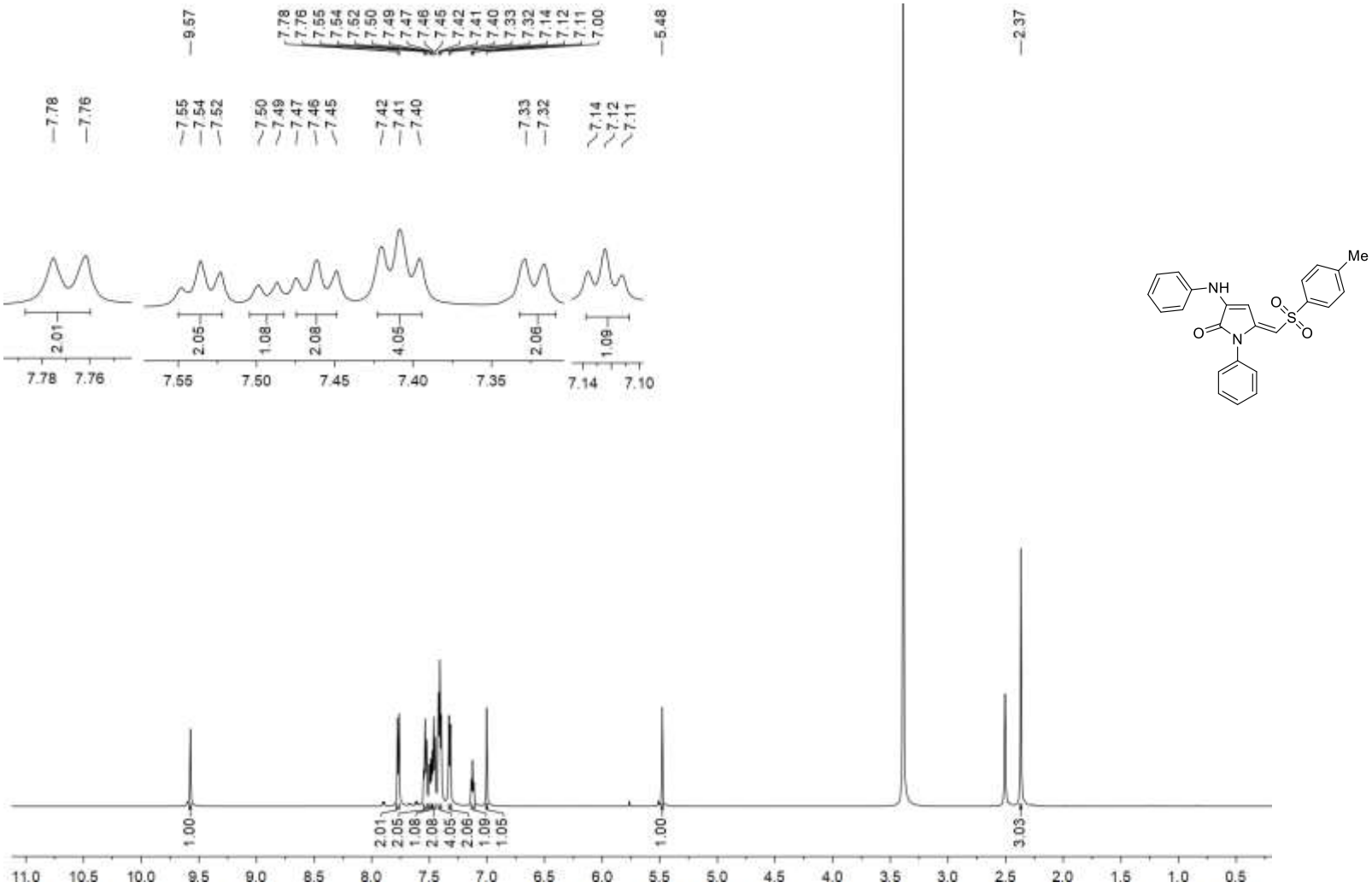


Figure S128. **^{1}H NMR** (600 MHz, DMSO-d_6) spectra of compound **9**

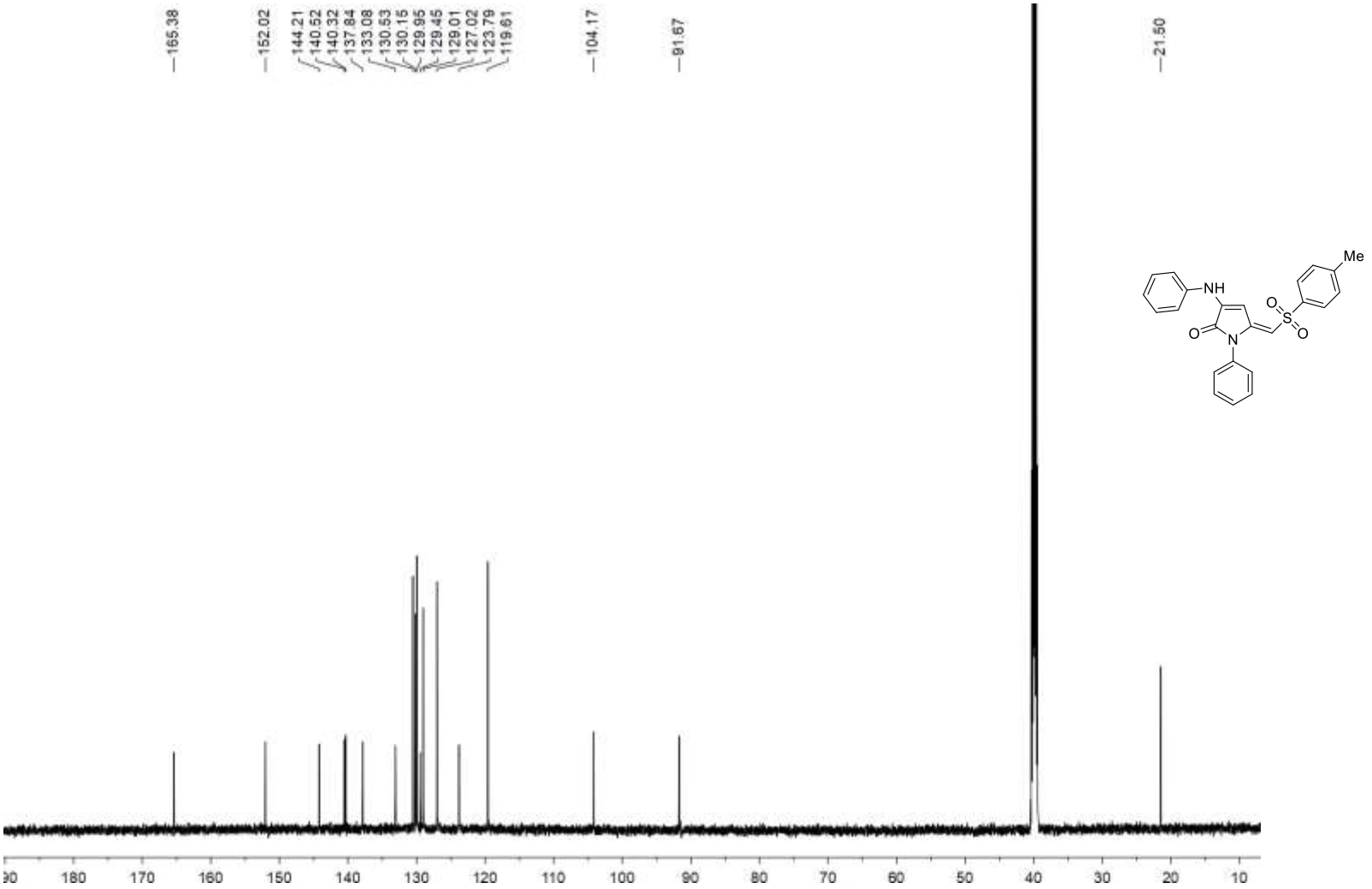


Figure S129. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **9**

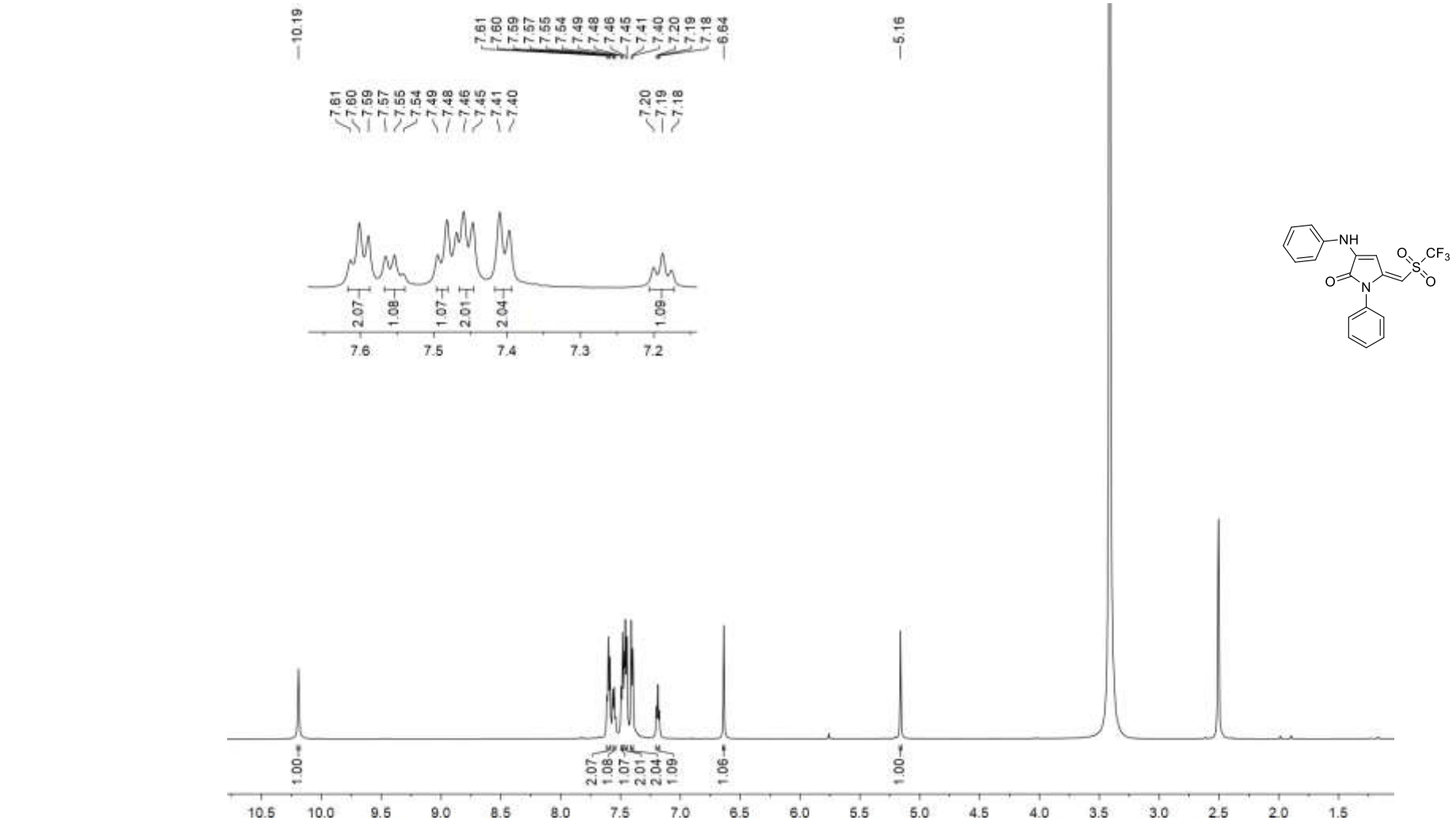


Figure S130. **^{1}H NMR** (600 MHz, DMSO-d_6) spectra of compound **10**

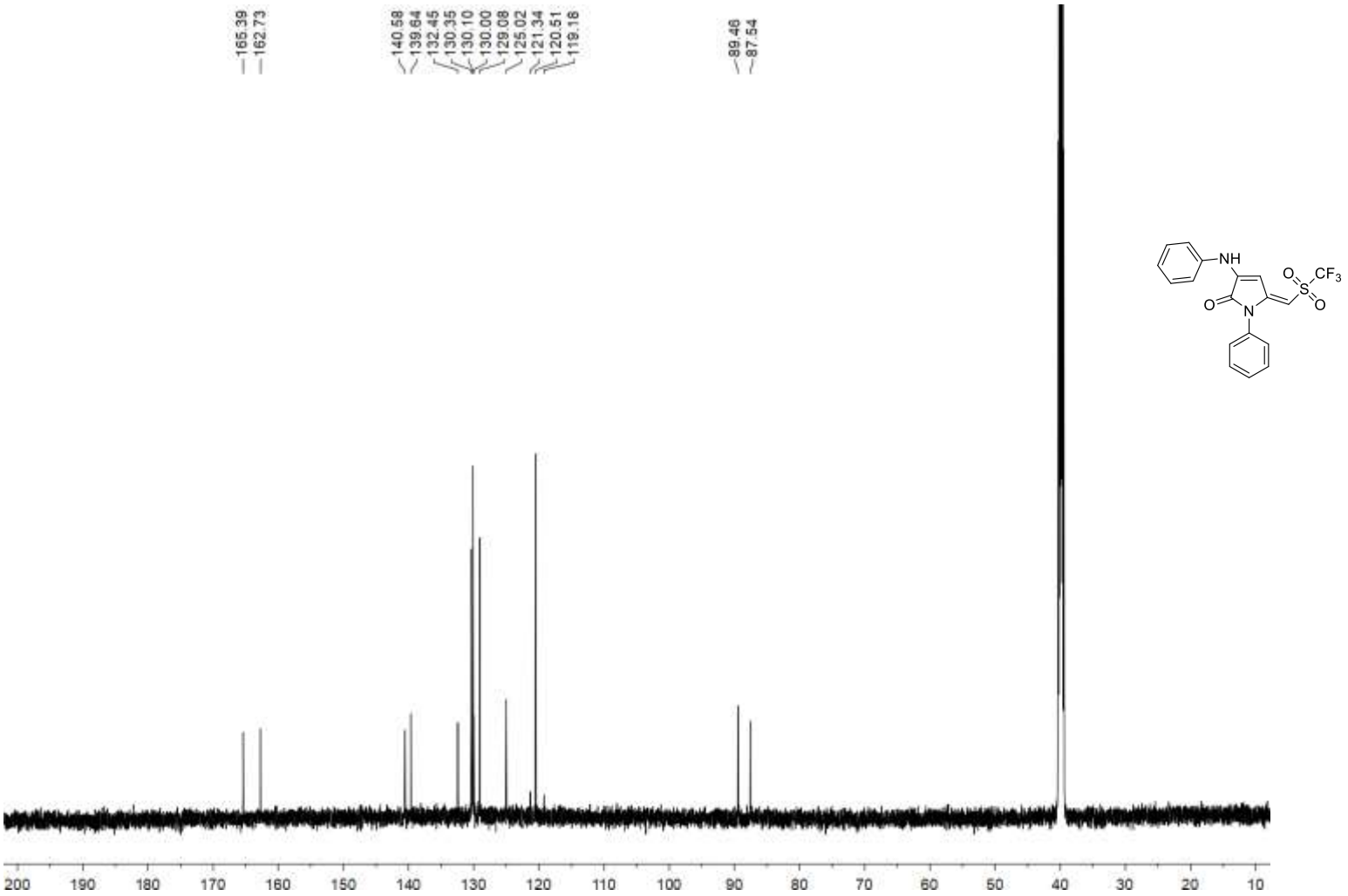


Figure S131. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **10**

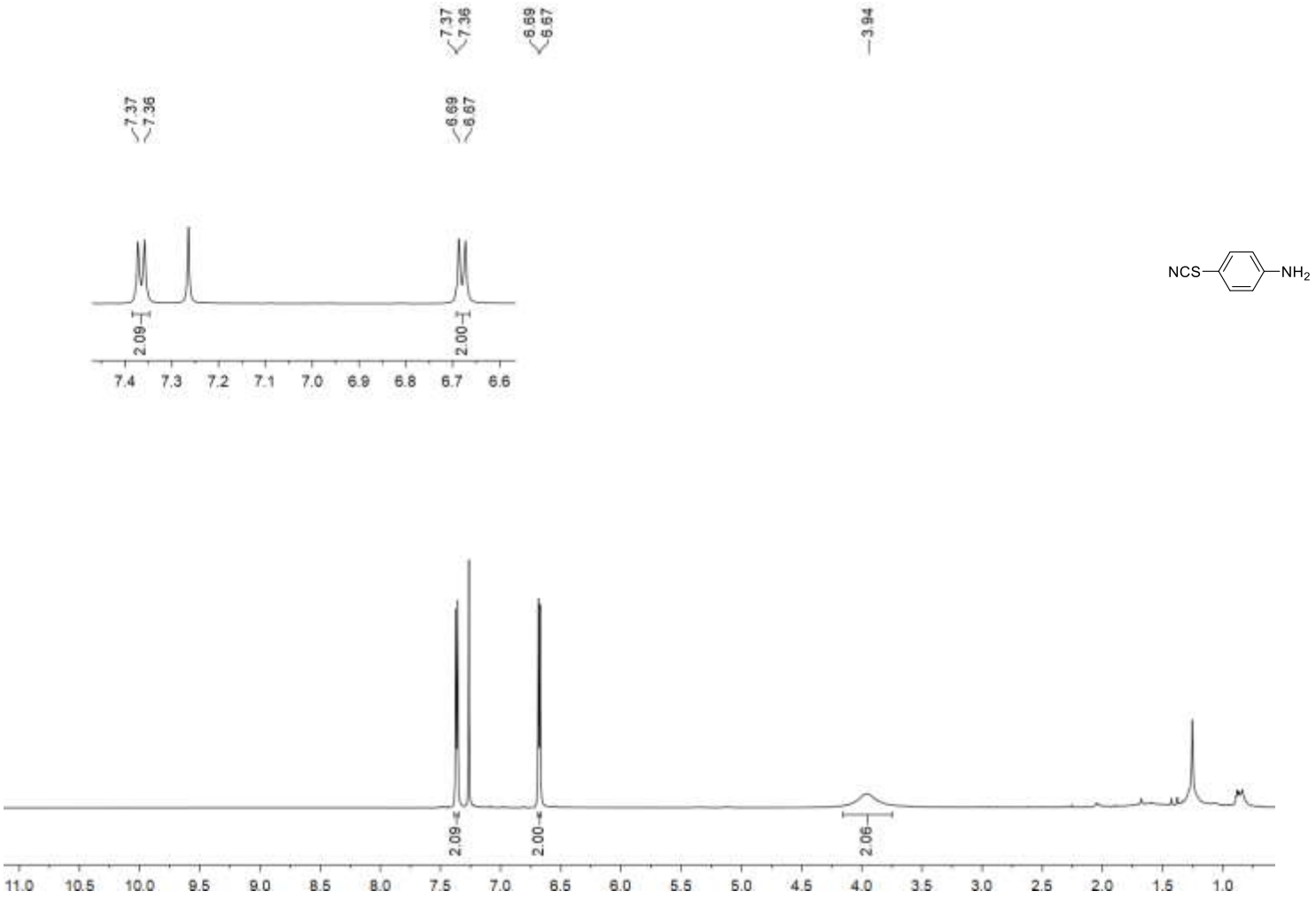


Figure S132. **^{1}H NMR** (600 MHz, $CDCl_3$) spectra of compound **11**

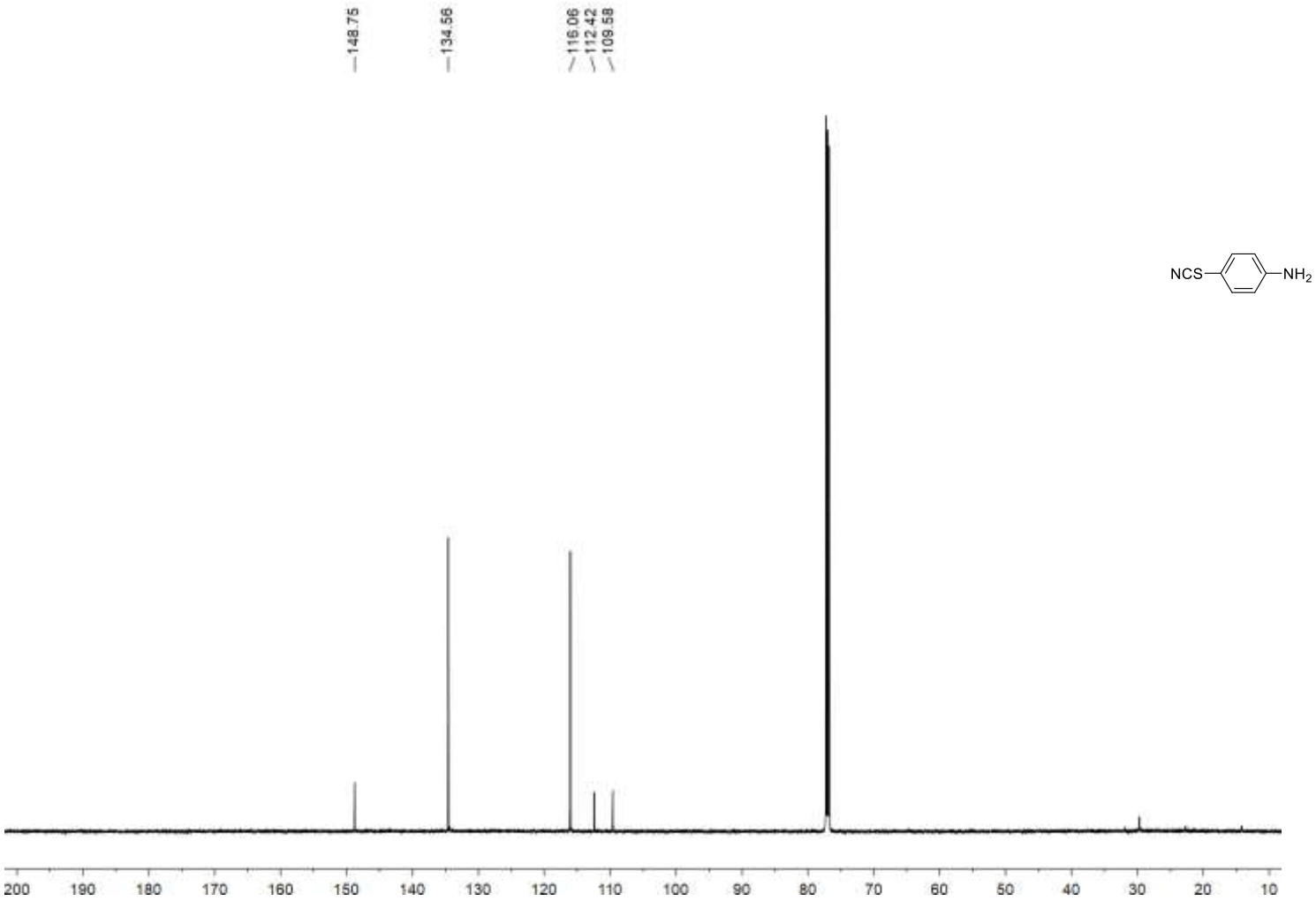


Figure S133. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **11**

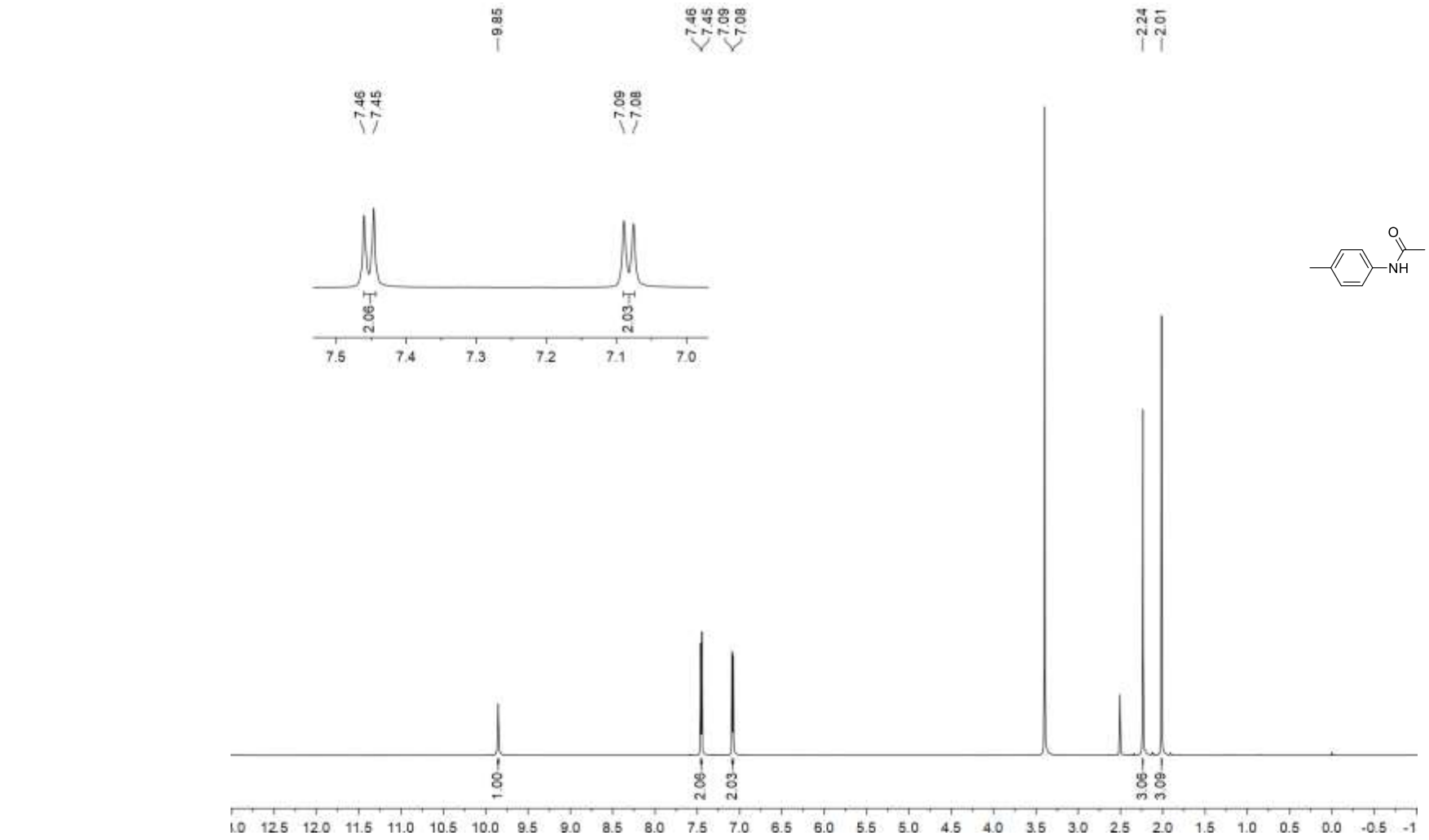


Figure S134. **^{1}H NMR** (600 MHz, DMSO-d_6) spectra of compound **12**

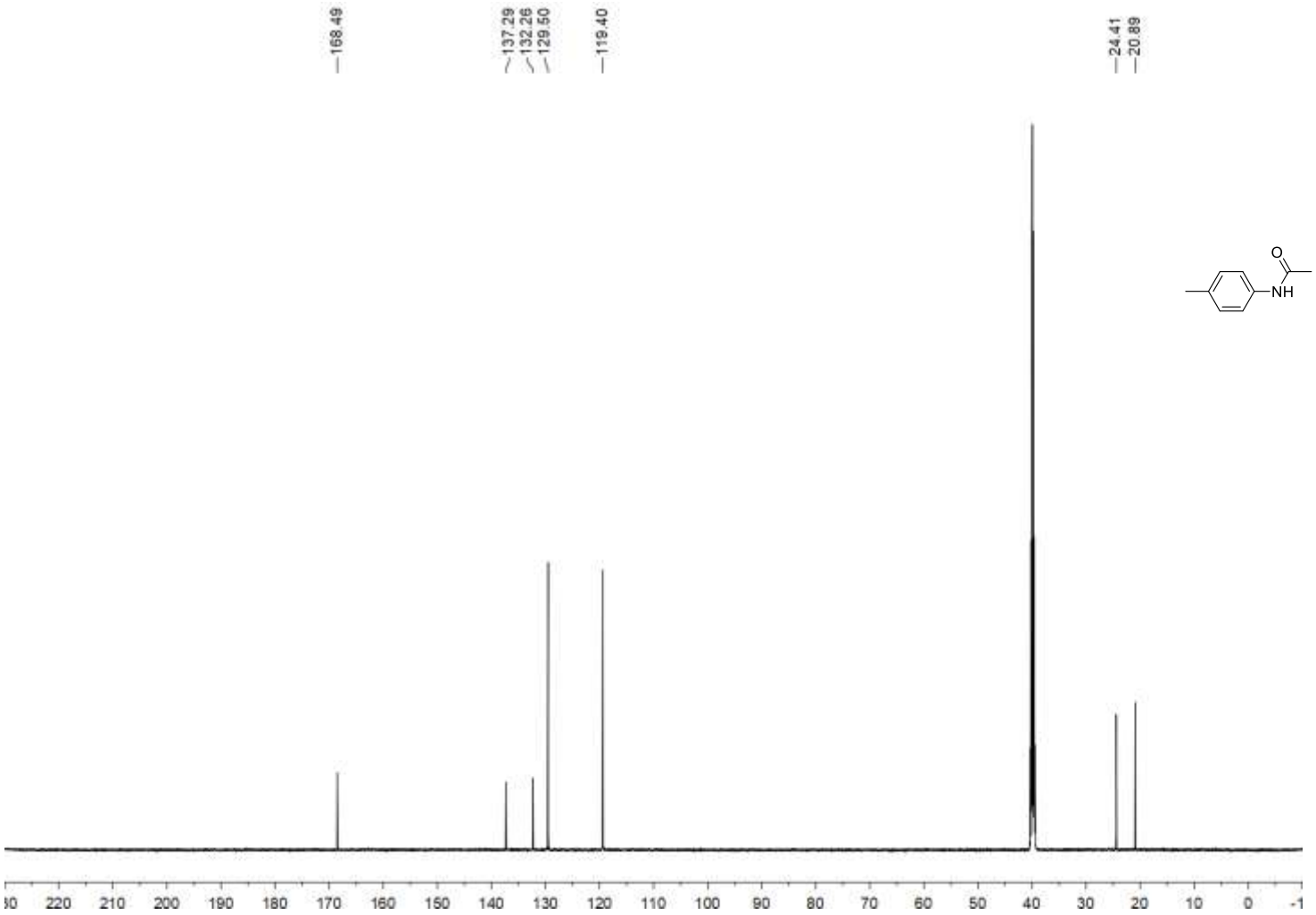


Figure S135. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **12**

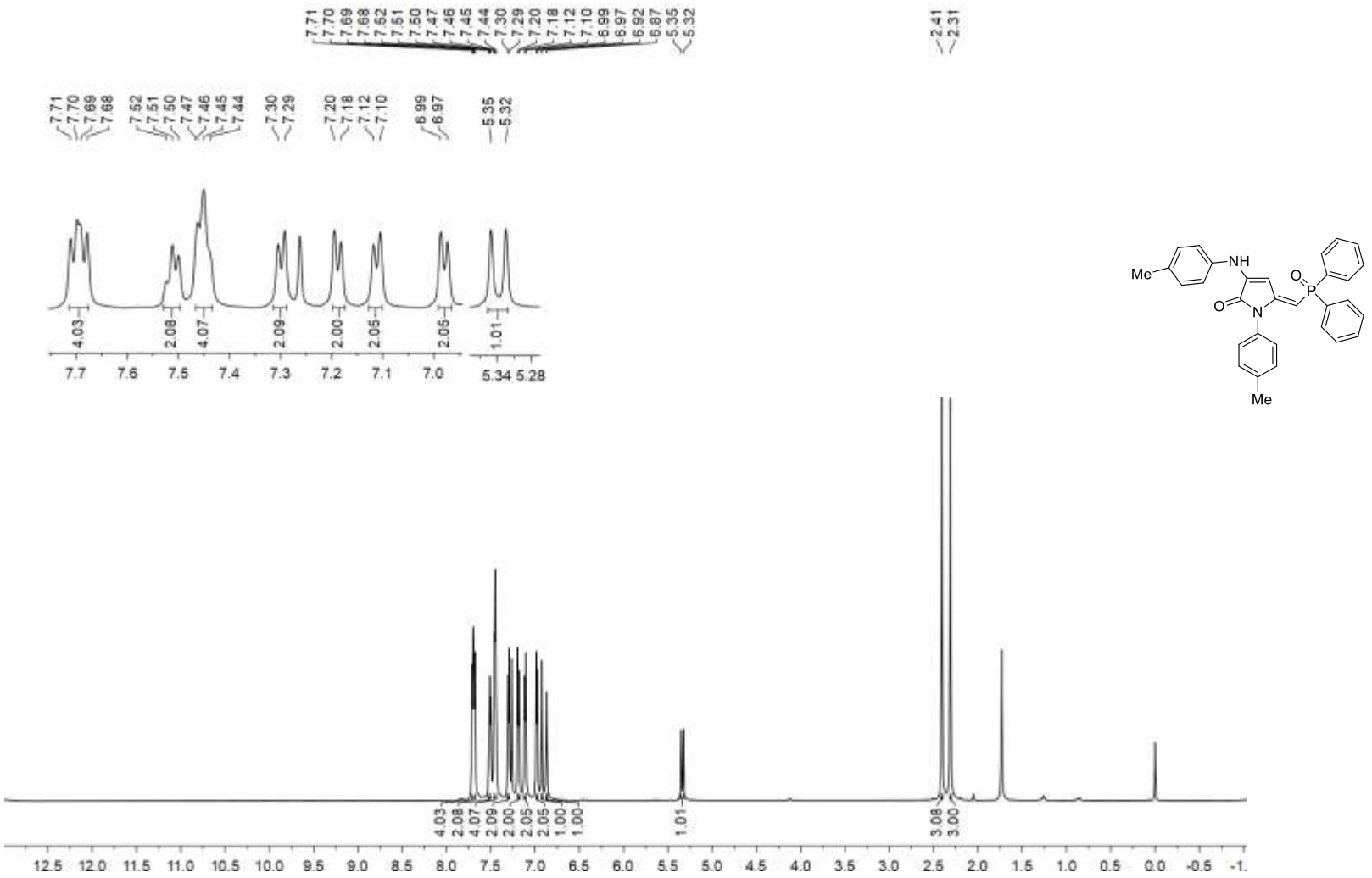


Figure S136. **^{1}H NMR** (600 MHz, $CDCl_3$) spectra of compound **13**

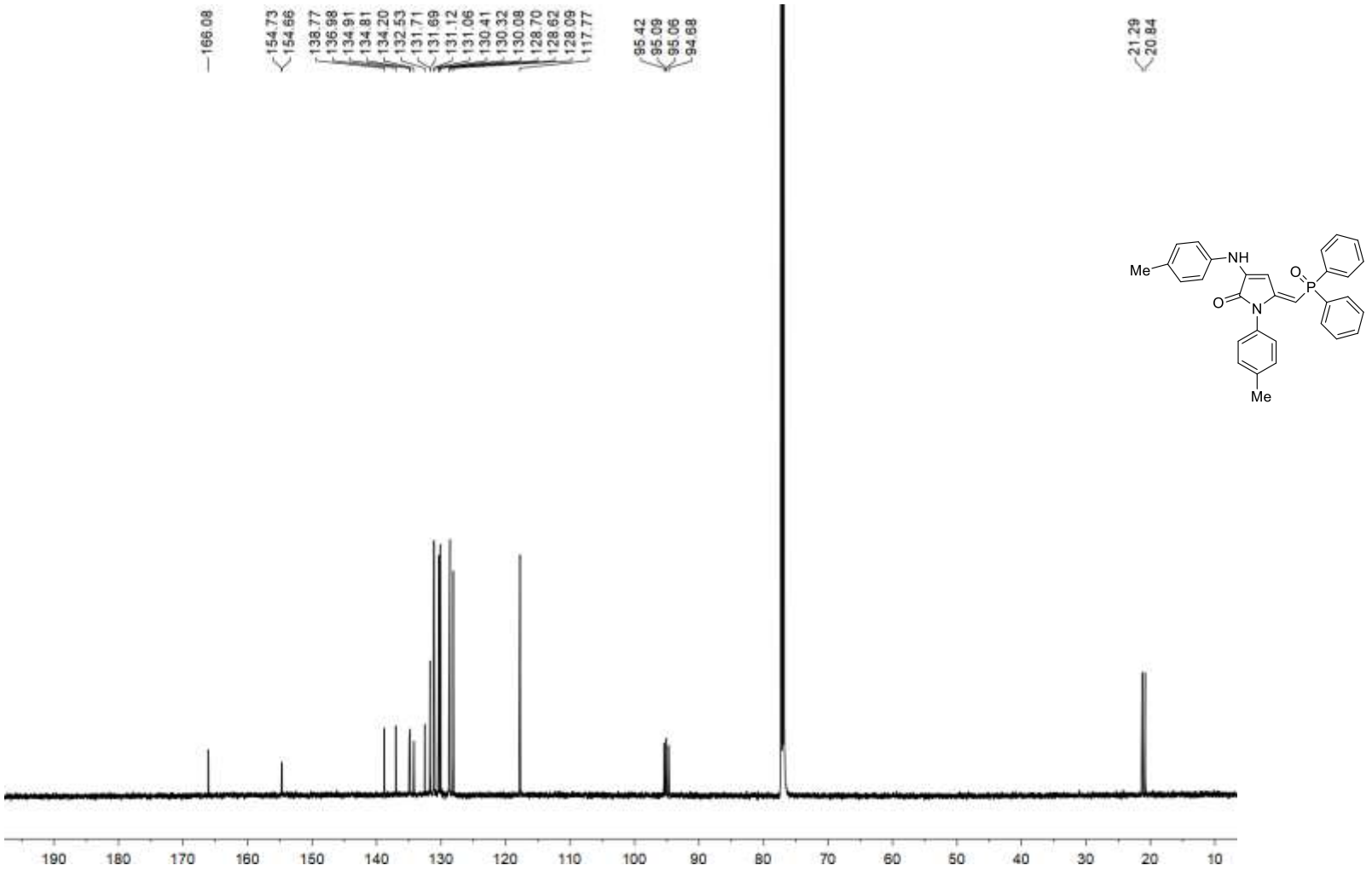


Figure S137. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **13**

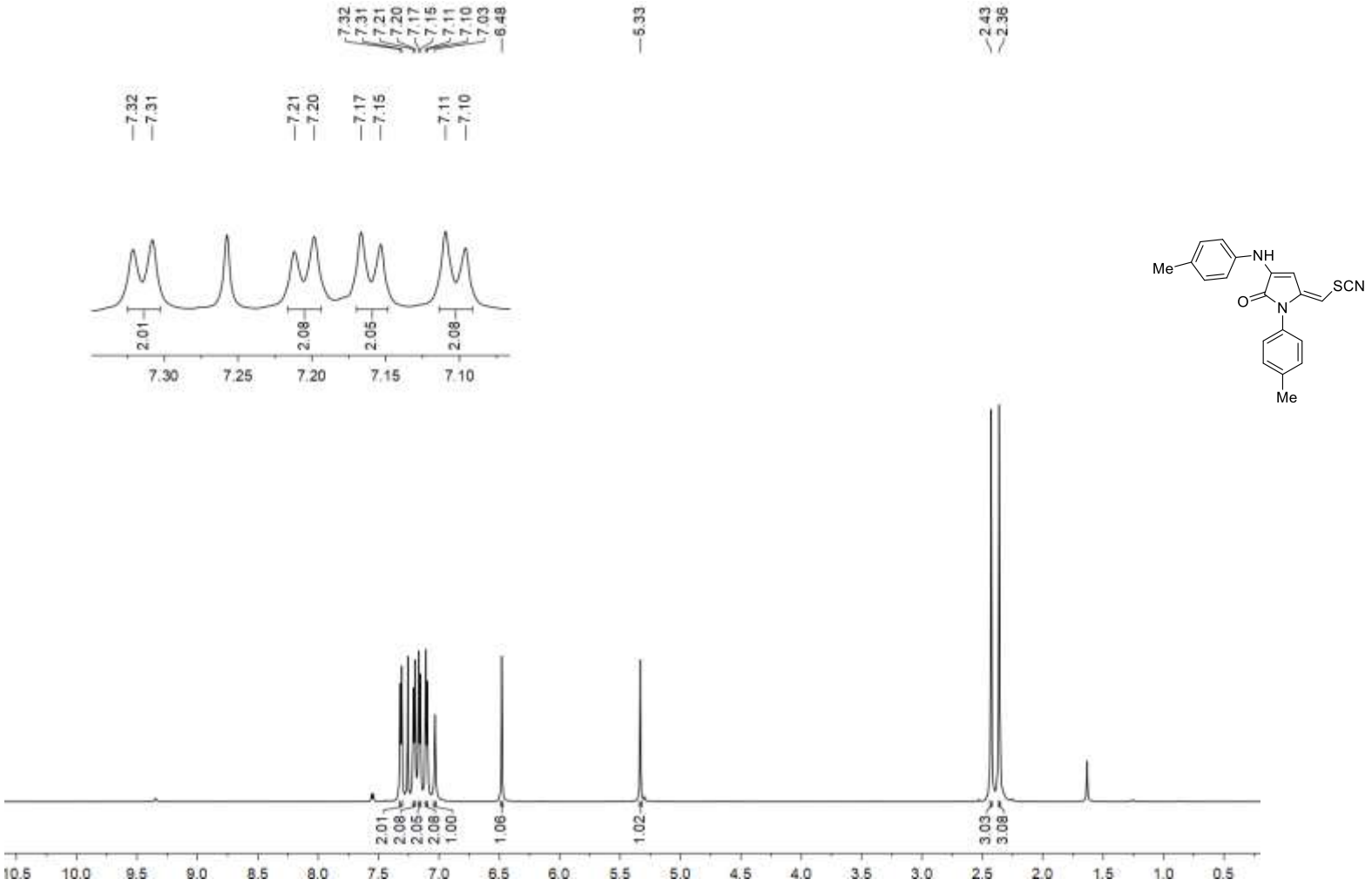


Figure S138. **^{1}H NMR** (600 MHz, $CDCl_3$) spectra of compound **14**

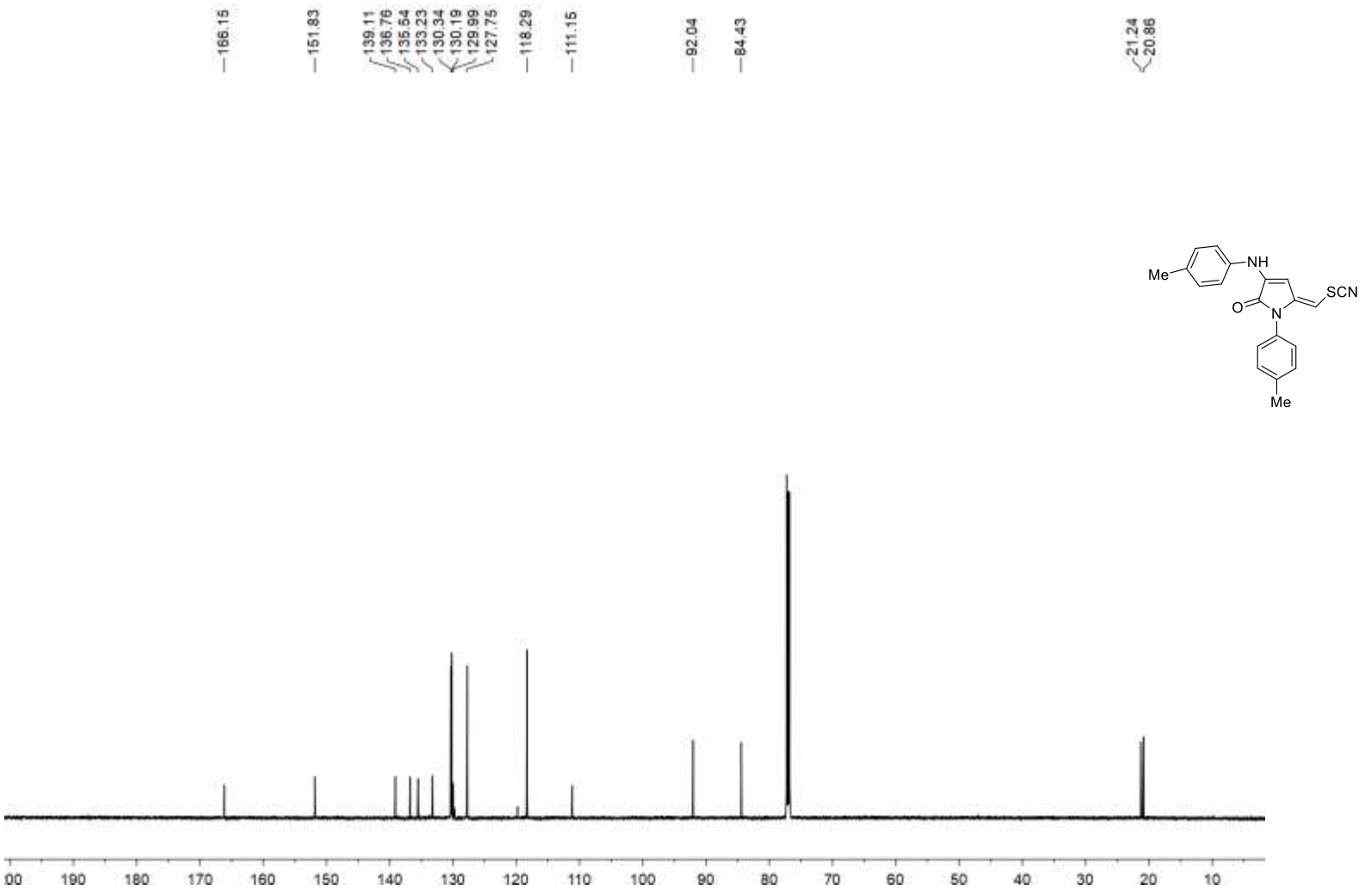


Figure S139. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **14**

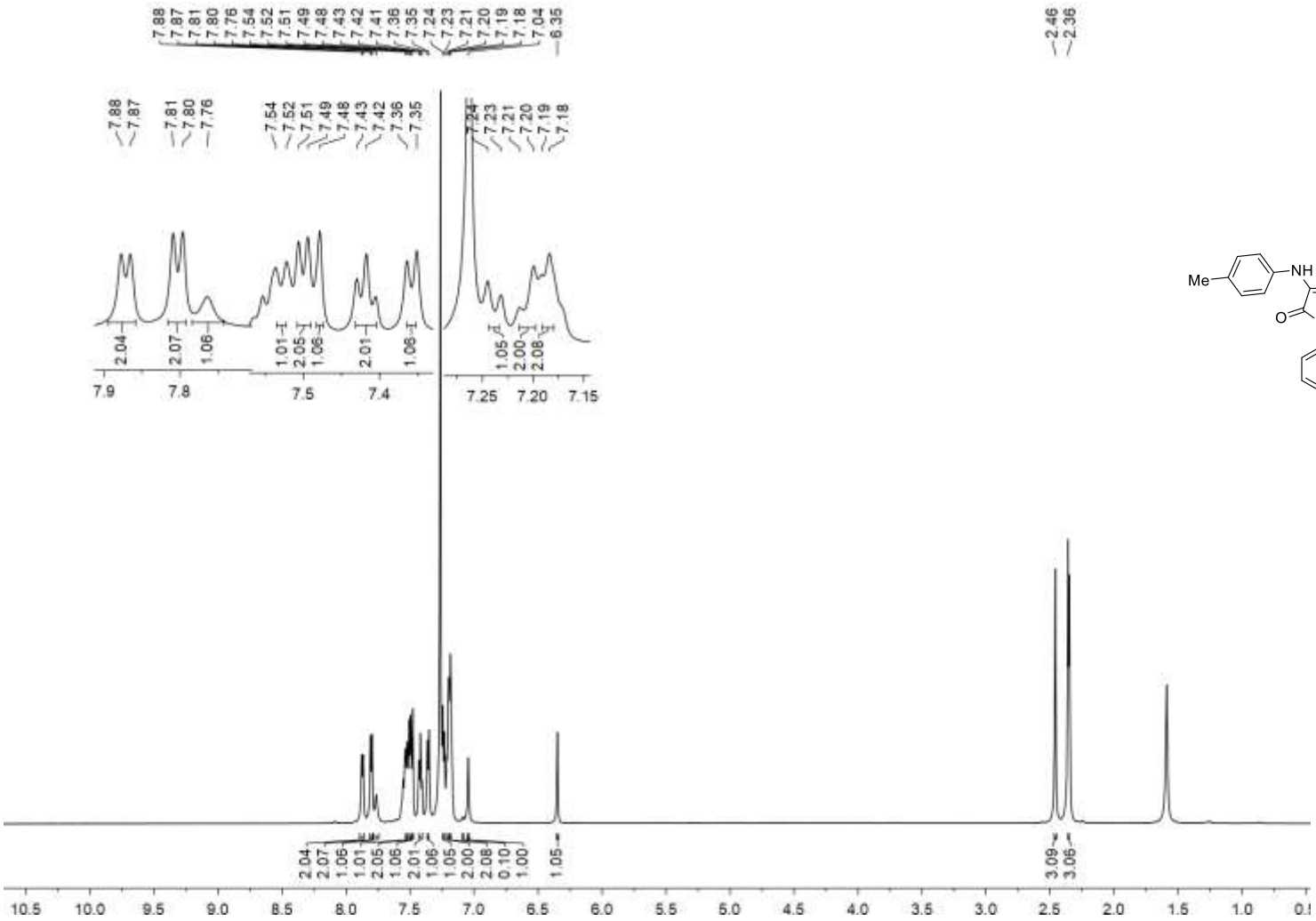


Figure S140. **^{1}H NMR** (600 MHz, $CDCl_3$) spectra of compound **15**

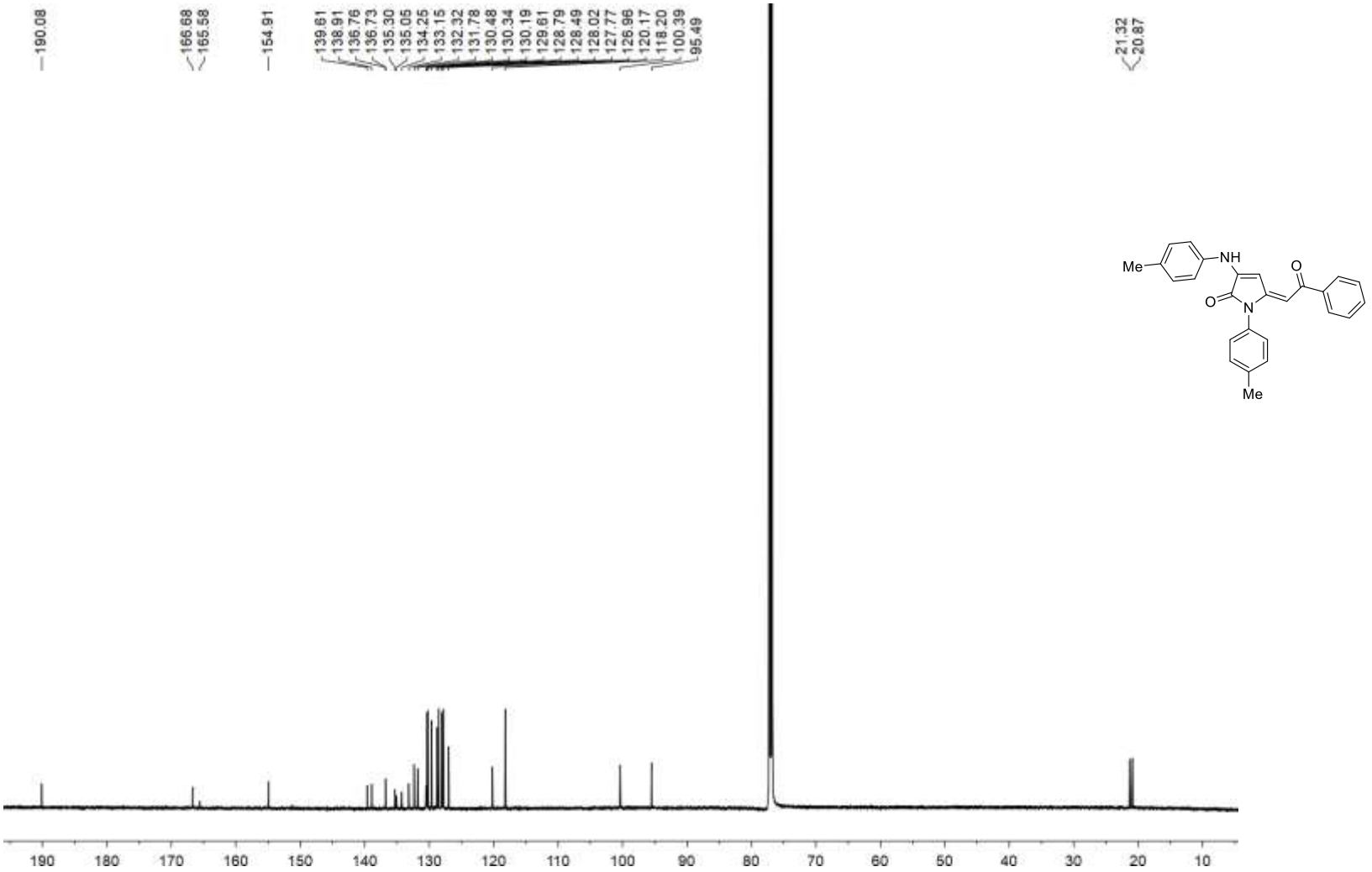


Figure S141. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **15**

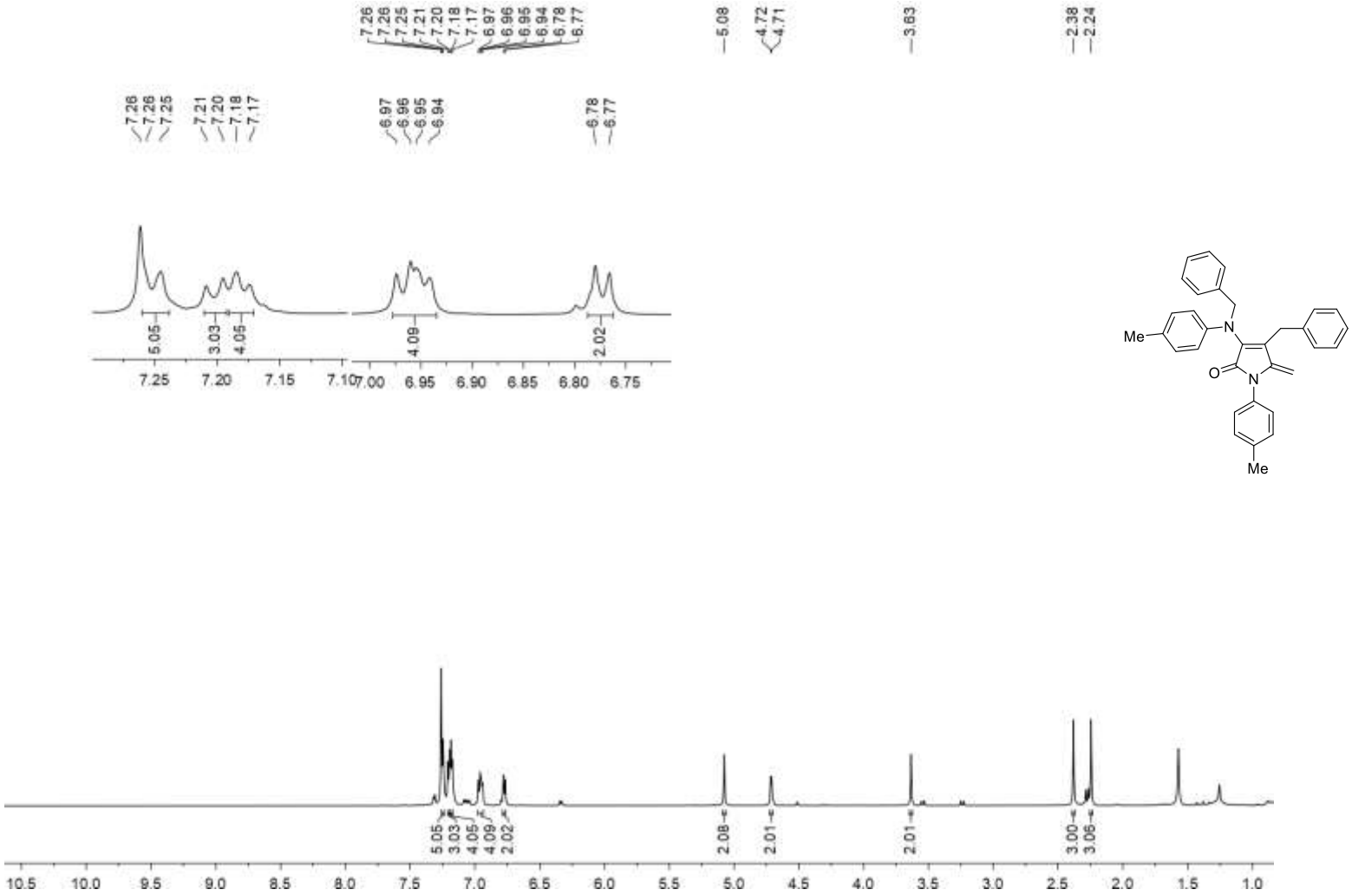


Figure S142. **^{1}H NMR** (600 MHz, $CDCl_3$) spectra of compound **16**

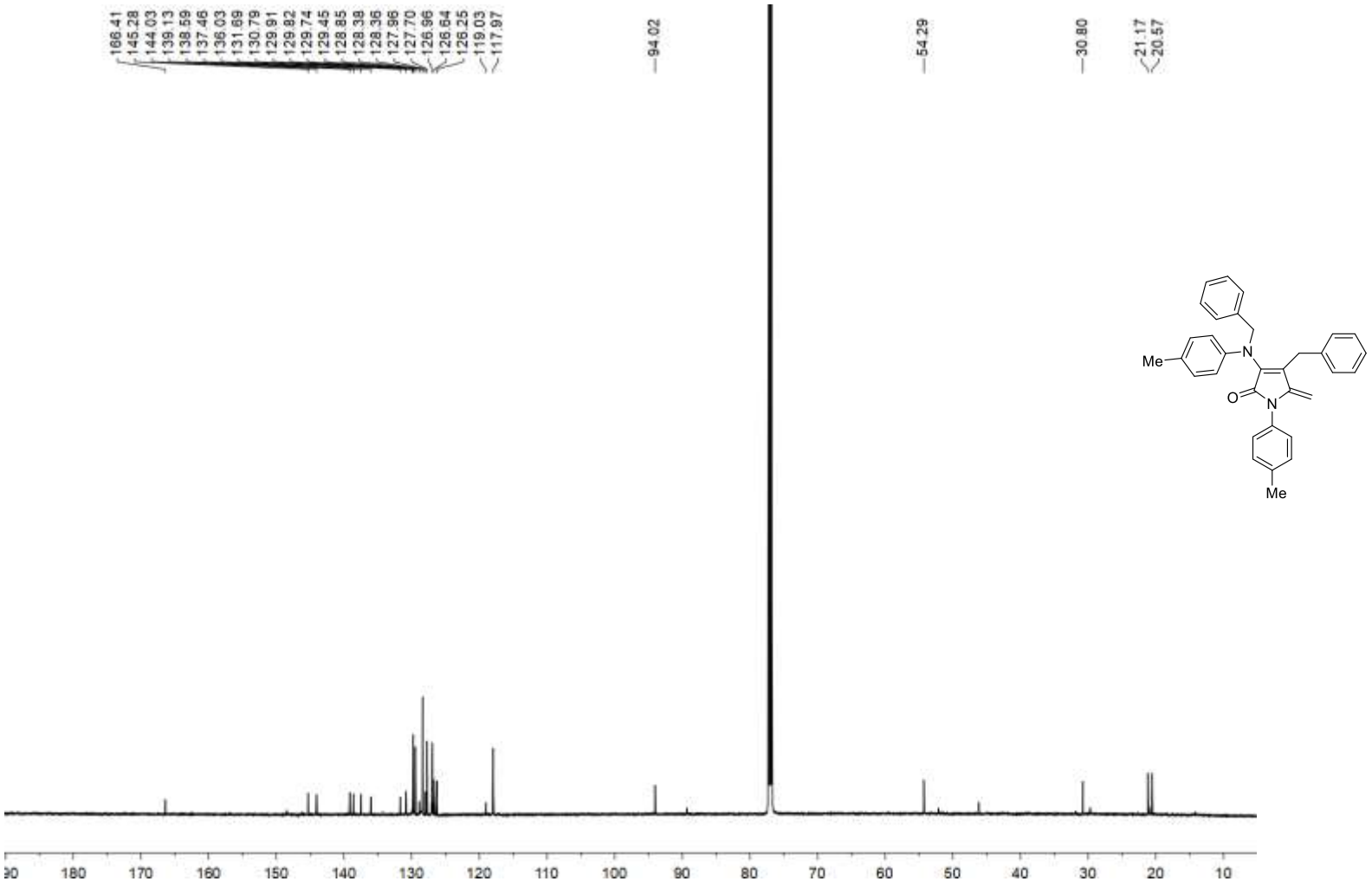


Figure S143. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **16**

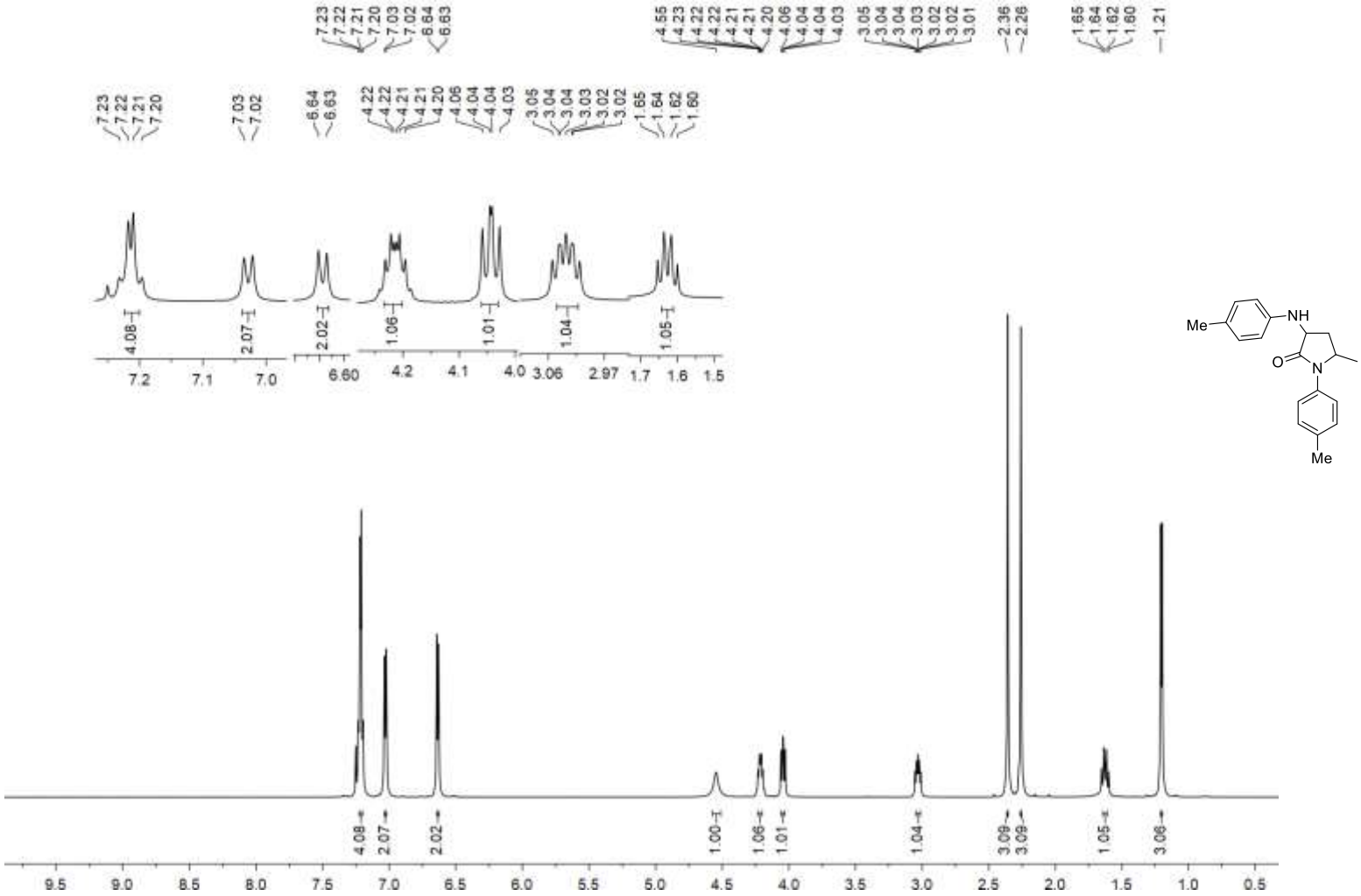


Figure S144. **^{1}H NMR** (600 MHz, $CDCl_3$) spectra of compound **17**

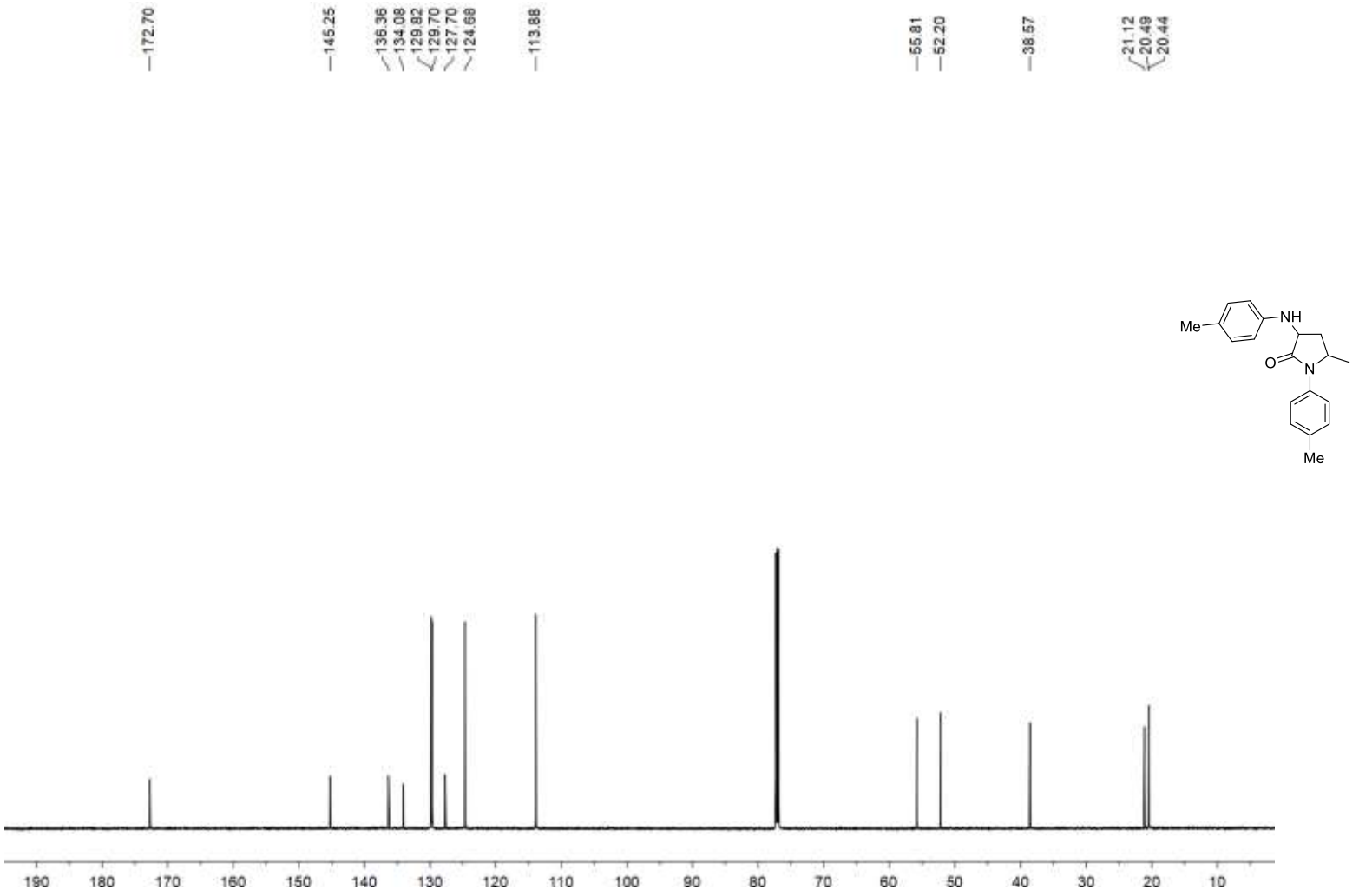


Figure S145. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **17**

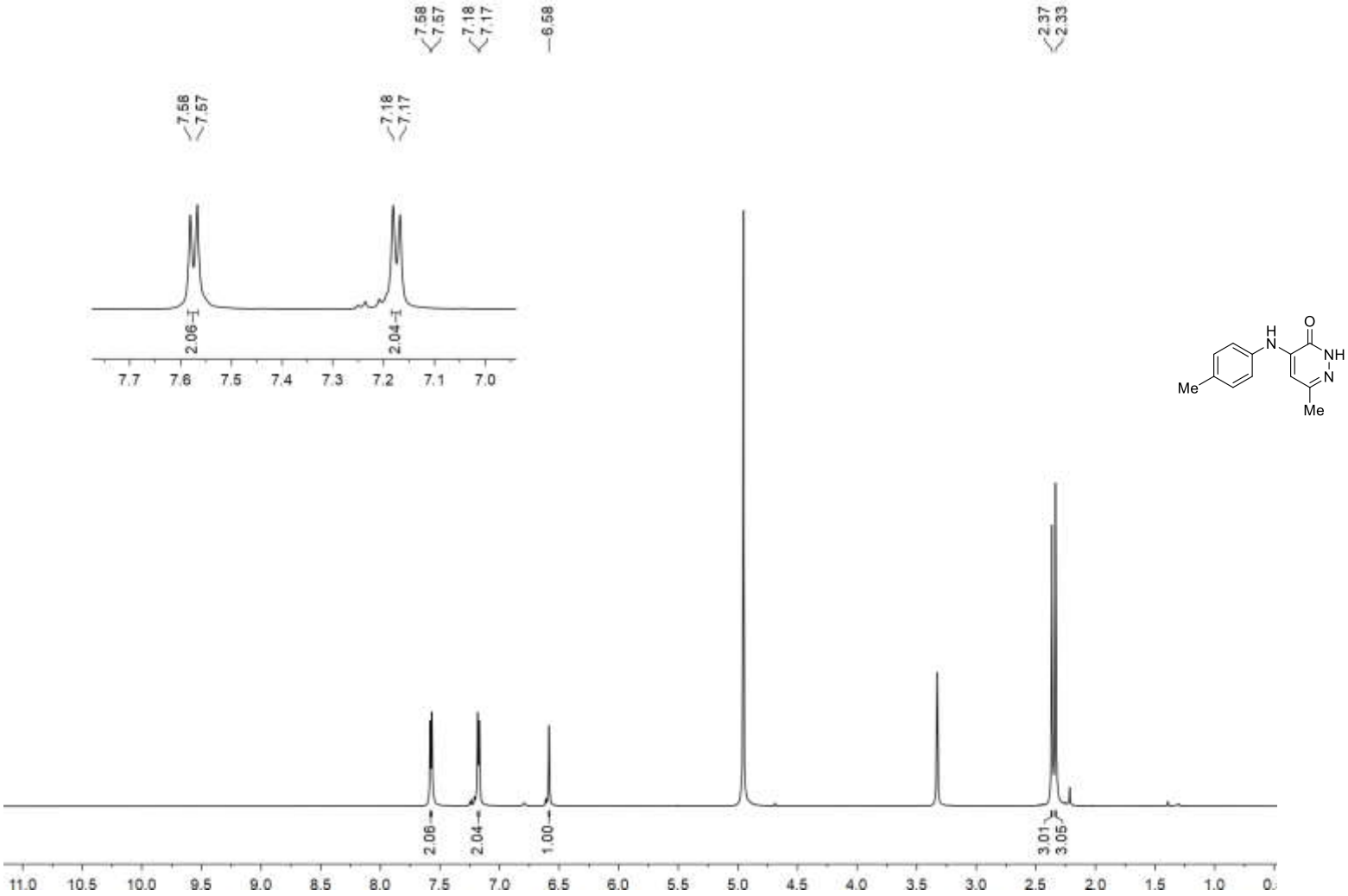


Figure S146. **^{1}H NMR** (600 MHz, CD_3OD) spectra of compound **18**

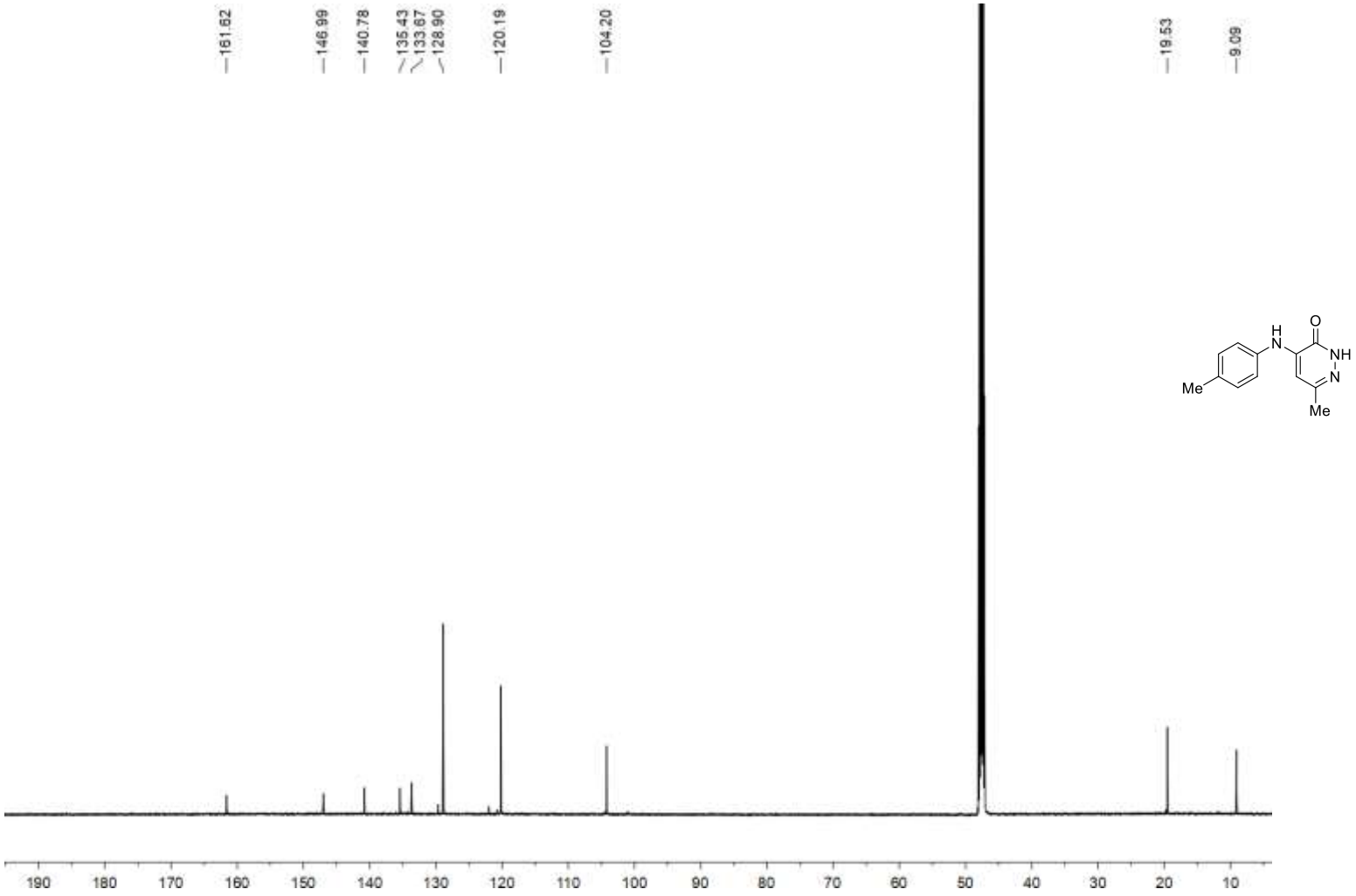


Figure S147. ^{13}C NMR (150 MHz, CD_3OD) spectra of compound **18**

7. References and notes.

1. Jian, L.; Run, L; Chun, Z.; Jian, Z. Copper(II) and 1,1'-trimethylene-2,2'-biimidazole-promoted arylation of acetylacetone with aryl iodides. *Chin. J. Chem.* **2011**, *29*, 309.
2. Lee, D.-H.; Taher, A.; Hossain, S.; Jin, M.-J. An efficient and general method for the Heck and Buchwald–Hartwig coupling reactions of aryl chlorides. *Org. Lett.*, **2011**, *13*, 5540.
3. CCDC 2226645, 2226644, and 2226646 contain the supplementary crystallographic data for compound **4b**, **7b** and **8b**. These data can be obtained free of charge from The Cambridge Crystallographic Data Center *via* www.ccdc.cam.ac.uk/data_request/cif.