Facile approach to multifunctionalized 5-alkylidene-3-pyrrolin-2-ones via regioselective oxidative cyclization of 2,4-pentanediones with primary amines and sodium sulfinates

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Supporting Information

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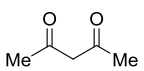
1. General information.

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.

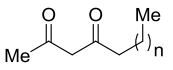
All chemicals and solvents were used as received without further purification unless otherwise stated. Column chromatography was performed on silica gel (200–300 mesh).

2,4-Pentanediones **1i**-**1o** were prepared according to the literature[1]. *N*-((2Z,4E)-4-(phenylimino)pent-2-en-2-yl)aniline **13** were prepared according to the literature[2]. 2,4-Pentanediones **1a**-**1h** and **1p**, primary amines **2**, sodium sulfinates **3**, oxidants, iodine reagents and acids were purchased from Energy Chemical and Adamas-beta®. The specific substrate structure is as follows:

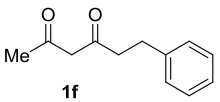
Substrate of 2,4-pentanediones **1**:



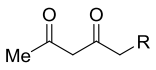
1a



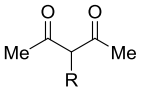
n = 0, **1b**; n = 1, **1c**;
n = 9, **1d**; n = 13, **1e**;



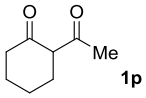
1f



R = Ph, **1g**;
R = 4-ClC_6H_4, **1h**;

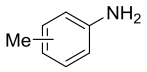


R = Ph, **1i**; R = 4-MeAr, **1j**; R = 3-MeAr, **1k**;
R = 4-FAr, **1l**; R = 4-ClAr, **1m**; R = 4-PhAr, **1n**;
R = 4-CO_2MeAr, **1o**;

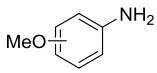


1p

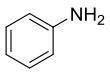
Substrate of primary amines **2**:



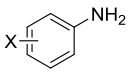
4-Me, **2a**;
3-Me, **2b**;
2-Me, **2c**;
2,3-Me, **2d**;



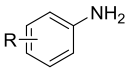
4-OMe, **2e**;
3-OMe, **2f**;
3,4,5-OMe, **2g**;



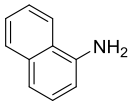
2h



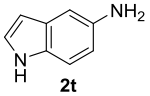
4-F, **2i**; 3-F, **2j**;
4-Cl, **2k**; 3-Cl, **2l**;
3-Br, **2m**;



4-CF_3, **2n**; 4-SO_2Me, **2o**;
4-OH, **2p**; 3-OH, **2q**;
4-CO_2H, **2r**;

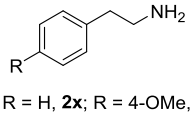


2s

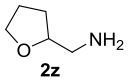


2t

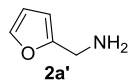
Et—NH_2 **2u**
^{n}Bu—NH_2 **2v**
Bn—NH_2 **2w**



R = H, **2x**; R = 4-OMe, **2y**;



2z



2a'

Substrate of sodium sulfinates **3**:

3b

3c

Ar = Ph, **3a**;
Ar = 4-MeC_6H_4, **3d**;
Ar = 4-ClC_6H_4, **3e**;
Ar = 4-FC_6H_4, **3f**;

Abbreviations:

TBHP: *tert*-butyl hydroperoxide; BPO: dibenzoyl peroxide; TBPB: *tert*-butyl peroxybenzoate;
CHP: cumene hydroperoxide; DMSO: dimethyl sulfoxide; DMF: *N*,*N*-dimethylformamide;
THF: tetrahydrofuran; PE: petroleum ether; EA: ethyl acetate;
DCM: dichloromethane; MTBE: methyl *tert*-butyl ether;

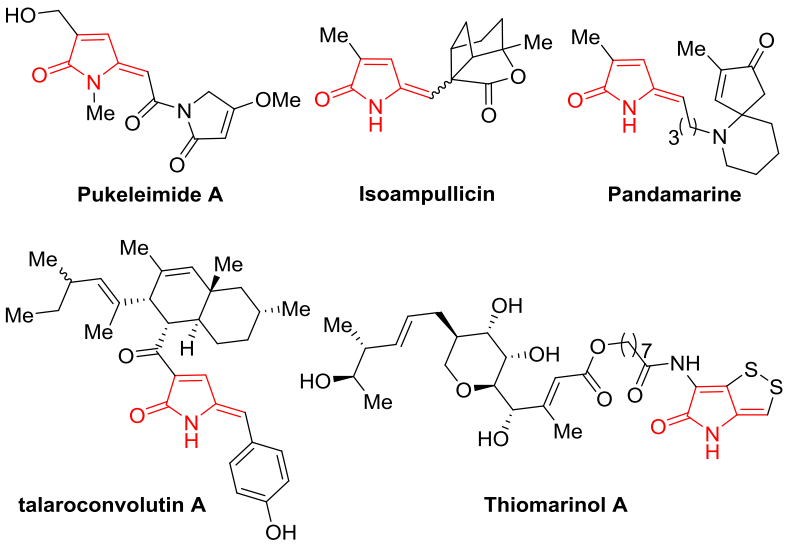
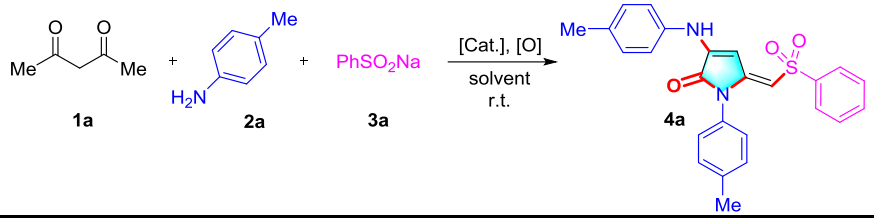


Figure S1 Representative examples of biologically active molecules with a 5-alkylidene-3-pyrrolin-2-one core

2. Optimization of reaction conditions.

Table S1 Optimization of reaction conditions

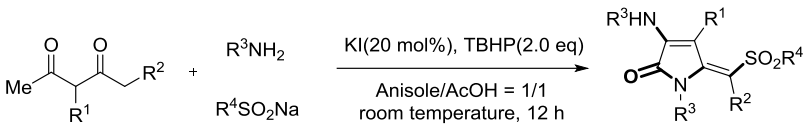


Entry	Oxidant (eq.)	Catalyst (mol%)	Solvent (*v*/*v* = 1/1)	Time (h)	Yield (%)
1	TBHP (2.0)	KI (40)	DMSO/HCO_2H	12	57
2	TBHP (2.0)	KI (40)	DMSO/TFA	18	n.d.[g]
3	TBHP (2.0)	KI (40)	DMSO/HCl	18	n.d.
4	TBHP (2.0)	KI (40)	DMSO/TSOH	18	trace
5	TBHP (2.0)	KI (40)	DMSO/$EtCO_2H$	15	37
6	TBHP (2.0)	KI (40)	DMSO/H_2SO_4	12	n.d.
7	TBHP (2.0)	KI (40)	DMSO/HNO_3	12	n.d.
8	TBHP (2.0)	KI (40)	DMSO/AcOH	12	62
9[d]	TBHP (2.0)	KI (40)	DMSO/AcOH	12	59
10[e]	TBHP (2.0)	KI (40)	DMSO/AcOH	12	60
11	TBHP (2.0)	I_2 (40)	DMSO/AcOH	12	60
12	TBHP (2.0)	LiI (40)	DMSO/AcOH	12	59
13	TBHP (2.0)	NH_4I (40)	DMSO/AcOH	12	50
14	TBHP (2.0)	CuI (40)	DMSO/AcOH	12	37
15	TBHP (2.0)	I_2O_5 (40)	DMSO/AcOH	12	34
16	TBHP (2.0)	NaI (40)	DMSO/AcOH	12	53
17[f]	TBHP (2.0)	KI (40)	DMSO/AcOH	12	60
18	DTBP (2.0)	KI (40)	DMSO/AcOH	23	trace
19	BPO (2.0)	KI (40)	DMSO/AcOH	12	58
20	TBPB (2.0)	KI (40)	DMSO/AcOH	12	33
21	CHP (2.0)	KI (40)	DMSO/AcOH	12	45
22	$K_2S_2O_8$ (2.0)	KI (40)	DMSO/AcOH	18	trace
23	Oxone (2.0)	KI (40)	DMSO/AcOH	18	trace
24	TBHP (2.0)	KI (40)	Cyclohexane /AcOH	12	49
25	TBHP (2.0)	KI (40)	1,4-dioxane/AcOH	13	50
26	TBHP (2.0)	KI (40)	THF/AcOH	15	52
27	TBHP (2.0)	KI (40)	Toluene/AcOH	12	55
28	TBHP (2.0)	KI (40)	PE/AcOH	12	36
29	TBHP (2.0)	KI (40)	EA/AcOH	12	58
30	TBHP (2.0)	KI (40)	DCM/AcOH	12	60
31	TBHP (2.0)	KI (40)	Water/AcOH	12	n.r.[h]
32	TBHP (2.0)	KI (40)	MeCN/AcOH	12	40
33	TBHP (2.0)	KI (40)	DMF/AcOH	12	58
34	TBHP (2.0)	KI (40)	MTBE/AcOH	12	62
35	TBHP (2.0)	KI (40)	Et_2O/AcOH	12	64
36	TBHP (2.0)	KI (40)	Anisole/AcOH	12	78
37	TBHP (3.0)	KI (40)	Anisole/AcOH	9	62
38	TBHP (1.0)	KI (40)	Anisole/AcOH	12	46
39	TBHP (2.0)	KI (30)	Anisole/AcOH	10	82
40	**TBHP (2.0)**	**KI (20)**	**Anisole/AcOH**	12	**95**
41	TBHP (2.0)	KI (10)	Anisole/AcOH	12	60
42	TBHP (2.0)	KI (20)	Anisole	15	n.r.
43	TBHP (2.0)	KI (20)	AcOH	15	trace
44	TBHP (2.0)	-	Anisole/AcOH	15	n.r.
45	-	KI (20)	Anisole/AcOH	15	trace

[a]Reaction conditions: **1** (0.5 mmol), **2** (1.25 mmol), **3** (1 mmol), oxidant, catalyst in 4 mL of solvent. [b]Isolated yield. [c]Determined by 1H NMR of the crude products *E*/*Z* > 99:1; [d]*V*(DMSO)/*V*(AcOH) = 1/2; [e]*V*(DMSO)/*V*(AcOH) = 2/1; [f]TBHP in decane; [g]n.d. = not detected;[h]n.r. = no reaction.

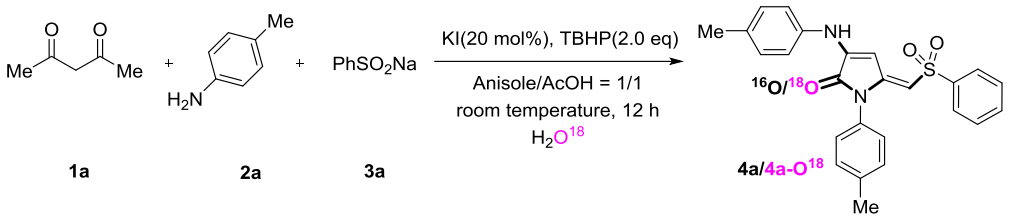
3. General procedure.

3.1 Synthesis of 5-alkylidene-pyrrolin-2-ones 4-8.



2,4-Pentanediones **1** (0.5 mmol), primary amines **2** (1.25 mmol), sodium sulfinates (1.0 mmol), KI (20 mol%), TBHP (70 wt.% in water, 2.0 eq.) and anisole/acetic acid (2.0 mL, v/v = 1/1) were charged into a 10 mL Ace Glass tubes, and the mixture was stirred at room temperature 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 5-alkylidene-3-pyrrolin-2-ones **4-8**.

3.2 ^{18}O labeling reaction.



2,4-Pentanediones **1a** (0.25 mmol), primary amines **2a** (0.62 mmol), sodium sulfinates (0.5 mmol), KI (20 mol%), TBHP (in decene, 2.0 eq.), anisole/acetic acid (0.5 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 room temperature 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 5-alkylidene-3-pyrrolin-2-ones **4a/4a-O^{18}**.

4. Confirmation of *Z*/*E* configuration of compound 7c.

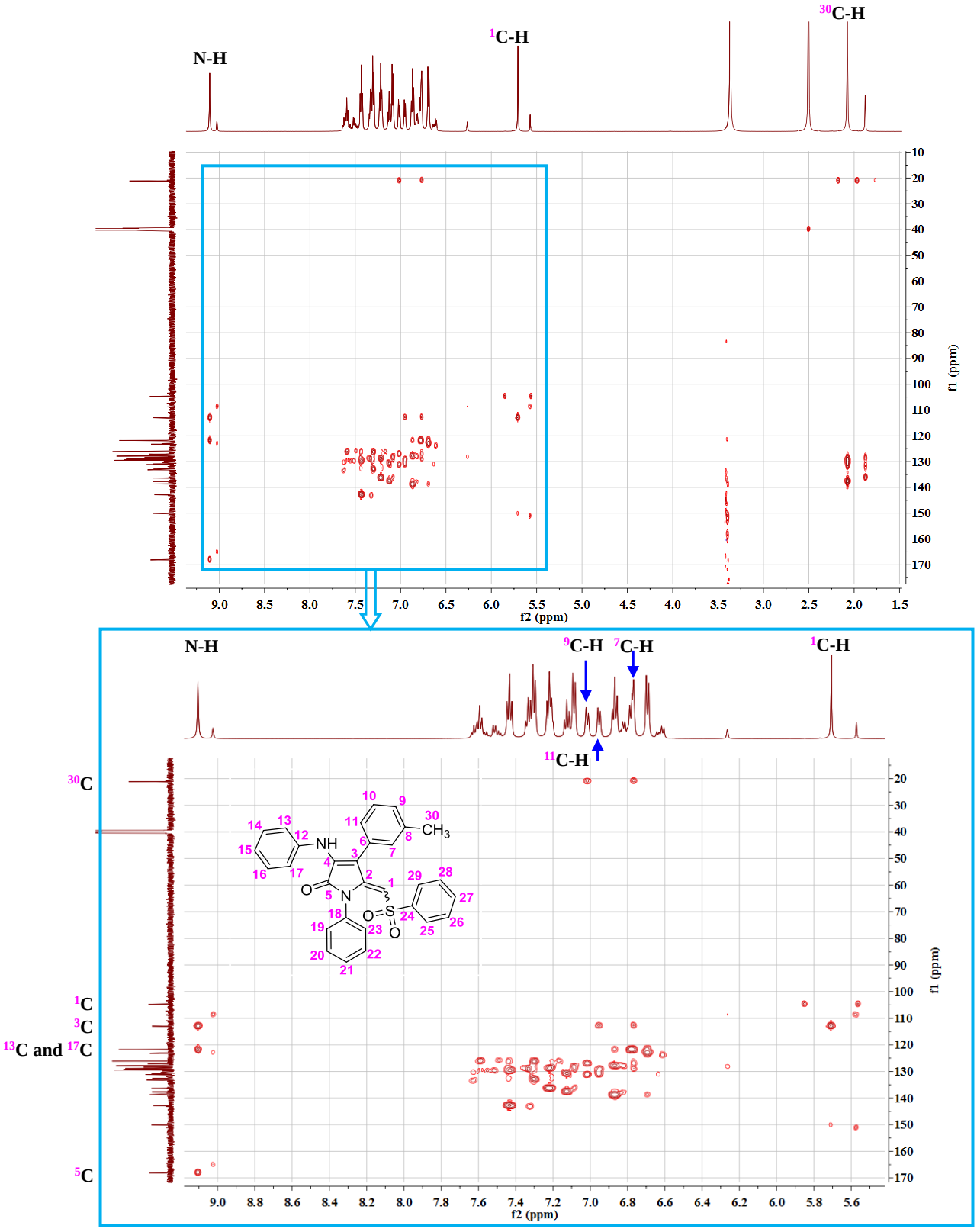


Figure S2. **HMBC** (600 MHz, DMSO-d_6) spectra of compound **7c**

After the analysis of HMBC, the positions of ^{1}C-H, ^{7}C-H, ^{9}C-H and ^{11}C-H in the structure of compound **7c** in ^{1}H NMR spectrum is confirmed.

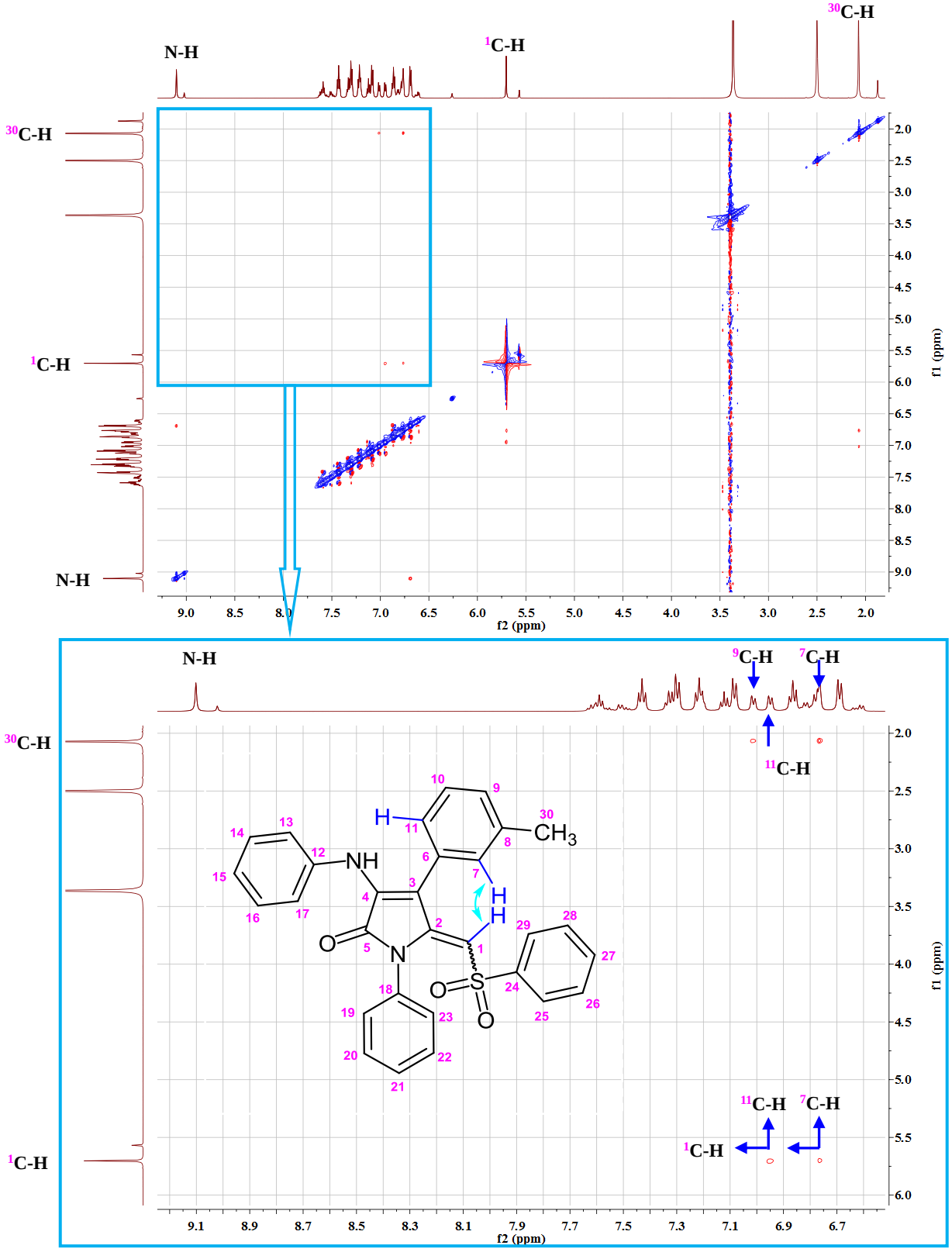
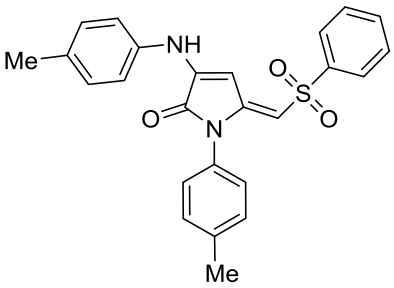


Figure S3. **ROSEY** (600 MHz, DMSO-d_6) spectra of compound **7c**

After the analysis of ROSEY, it is confirmed that ^{1}C-H in the structure of compound **7c** is only associated with ^{7}C-H and ^{11}C-H, not ^{19}C-H and ^{23}C-H. Therefore, it is proved that the structure configuration of major product in compound **7c** is the *Z*-type.

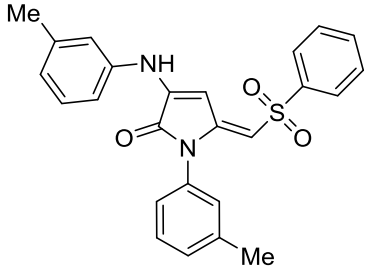
5. Spectroscopic data.

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



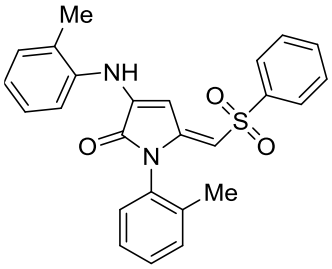
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 18:1, R_f = 0.2; Yellow solid: 204 mg (95%), mp = 211–213 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.89 (d, *J* = 7.6 Hz, 2H, ArH), 7.59–7.56 (m, 1H, A rH), 7.52–7.49 (m, 2H, ArH), 7.28 (s, 2H, ArH), 7.22 (d, *J* = 8.0 Hz, 2H, ArH), 7.16–7. 13 (m, 3H, ArH+NH), 7.09 (s, 2H, ArH), 7.08 (s, 1H, C=CH), 5.63 (s, 1H, C=CH), 2.40 (s, 3H, $ArCH_3$), 2.37 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.9, 152.1, 142.8, 139.3, 136.5, 136.1, 133.6, 133.0, 130.4, 130.4, 130.3, 130.3, 129.7, 129.3, 129.3, 1 27.9, 127.9, 126.9, 126.9, 118.4, 118.4, 105.3, 92.0, 21.3, 20.9; **HRMS** (TOF ES+): m/z c alcd for $C_{25}H_{23}N_2O_3S$ $[(M+H)^+]$, 431.1424, found, 431.1430.

(*E*)-5-((Phenylsulfonyl)methylene)-1-(*m*-tolyl)-3-(*m*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one (4b)



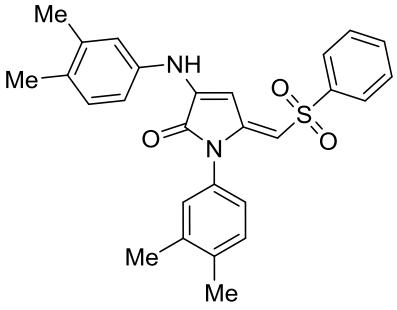
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 200 mg (93%), mp = 147–149 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.90 (d, *J* = 7.4 Hz, 2H, ArH), 7.60–7.57 (m, 1H, ArH), 7.53–7.51 (m, 2H, ArH), 7.38–7.30 (m, 2H, ArH), 7.24–7.21 (m, 1H, ArH), 7.21 (s, 1H, C=CH), 7.09 (d, *J* = 5.8 Hz, 1H, ArH), 7.04–7.02 (m, 3H, ArH+NH), 6.99 (d, *J* = 7.9 Hz, 1H, ArH), 6.97 (d, *J* = 7.6 Hz, 1H, ArH), 5.66 (s, 1H, C=CH), 2.41 (s, 3H, $ArCH_3$), 2.38 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.7, 151.9, 142.7, 140.0, 139.8, 138.9, 135.8, 133.0, 132.2, 130.0, 129.7, 129.5, 129.3, 129.3, 128.8, 126.9, 126.9, 125.1, 124.7, 119.1, 115.2, 105.6, 92.5, 21.6, 21.3; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{23}N_2O_3S$ $[(M+H)^+]$, 431.1424, found, 431.1430.

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



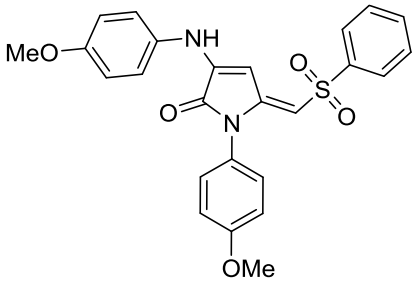
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 18:1, R_f = 0.2; Yellow oli: 196 mg (91%); **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.87 (d, *J* = 7.4 Hz, 2H, ArH), 7.59–7.57 (m, 1H, ArH), 7.52–7.50 (m, 2H, ArH), 7.46 (d, *J* = 7.9 Hz, 1H, ArH), 7.38–7.36 (m, 2H, ArH), 7.33–7.30 (m, 1H, ArH), 7.28–7.27 (m, 2H, ArH), 7.12 (s, 1H, C=CH), 7.11–7.10 (m, 2H, ArH), 6.87 (s, 1H, NH), 5.41 (s, 1H, C=CH), 2.36 (s, 3H, $ArCH_3$), 2.06 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.5, 151.6, 142.7, 137.3, 137.1, 136.5, 133.0, 131.5, 131.2, 131.2, 129.9, 129.3, 129.3, 129.1, 127.9, 127.5, 127.3, 126.8, 126.8, 124.2, 118.3, 105.4, 92.2, 17.6, 17.5; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{23}N_2O_3S$ [$(M+H)^+$], 431.1424, found, 431.1430.

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



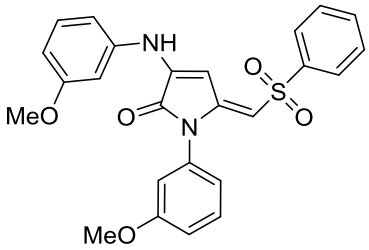
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 18:1 , R_f = 0.2 ; Yellow solid: 203 mg (89%), mp = 177–179 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.90–7.88 (m, 2H, ArH), 7.59–7.56 (m, 1H, ArH), 7.52–7.50 (m, 2H, ArH), 7.22–7.18 (dd, 13.7, 8.0 Hz, 2H, ArH), 7.12 (s, 1H, C=CH), 7.04–7.02 (dd, *J* = 8.0, 2.3 Hz, 1H, ArH), 6.99–6.98 (m, 3H, ArH+NH), 6.93–6.91 (dd, *J* = 7.9, 2.0 Hz, 1H, ArH), 5.63 (s, 1H, C=CH), 2.31 (s, 3H, $ArCH_3$), 2.29 (s, 3H, $ArCH_3$), 2.28 (s, 3H, $ArCH_3$), 2.27 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.9, 152.4, 142.9, 138.4, 138.2, 138.1, 136.7, 136.2, 132.9, 132.4, 130.8, 130.8, 129.9, 129.3, 129.3, 129.1, 126.9, 126.9, 125.5, 119.9, 115.6, 105.2, 91.8, 20.1, 19.9, 19.6, 19.3; **HRMS** (TOF ES+): m/z calcd for $C_{27}H_{27}N_2O_3S$ [$(M+H)^+$], 459.1737, found, 459.1745.

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



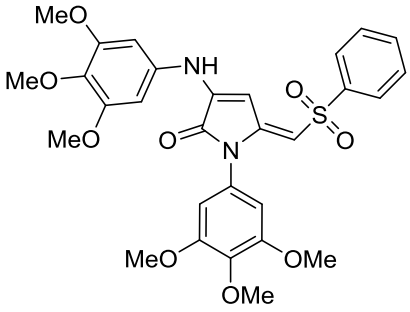
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 25:1 , R_f = 0.2; Yellow solid: 221 mg (96%) , mp = 217–218 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.89–7.88 (m, 2H, ArH), 7.59–7.56 (m, 1H, ArH), 7.52–7.50 (m, 2H, ArH), 7.19 (d, *J* = 8.9 Hz, 2H, ArH), 7.12 (d, *J* = 8.9 Hz, 2H, ArH), 7.07 (s, 1H, C=CH), 7.01 (s, 1H, NH), 6.97 (d, *J* = 2.2 Hz, 2H, ArH), 6.96 (d, *J* = 2.2 Hz, 2H, ArH), 5.59 (s, 1H, C=CH), 3.85 (s, 3H, $ArOCH_3$), 3.84 (s, 3H, $ArOCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.9, 159.9, 156.2, 152.5, 142.8, 136.6, 132.9, 132.2, 129.5, 129.5, 129.3, 129.3, 126.8, 126.8, 124.8, 120.1, 120.1, 115.0, 115.0, 115.0, 115.0, 104.9, 90.9, 55.6, 55.6; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{23}N_2O_5S$ [$(M+H)^+$], 463.1322, found, 463.1328.

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



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 8:1, R_f = 0.2; Yellow solid: 207 mg (90%), mp = 171–172 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.91–7.89 (m, 2H, ArH), 7.60–7.57 (m, 1H, ArH), 7.52 (dd, *J* = 8.4 Hz, 7.0 Hz, 2H, ArH), 7.39–7.36 (m, 1H, ArH), 7.35–7.32 (m, 1H, ArH), 7.25 (s, 1H, C=CH), 7.07 (s, 1H, NH), 6.98–6.97 (dd, *J* = 8.4 Hz, 1.9Hz, 1H, ArH), 6.86–6.84 (dd, *J* = 8.0 Hz, 1.7Hz, 1H, ArH), 6.79–6.77 (m, 2H, ArH), 6.73 (t, 1H, *J* = 2.1 Hz, ArH), 6.70–6.69 (dd, *J* = 8.3, 2.0 Hz, 1H, ArH), 5.72 (s, 1H, C=CH), 3.86 (s, 3H, $ArOCH_3$), 3.79 (s, 3H, $ArOCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.5, 160.7, 160.4, 151.5, 142.6, 140.1, 135.7, 133.3, 133.0, 130.7, 130.5, 129.3, 129.3, 126.9, 126.9, 120.2, 114.9, 113.8, 110.7, 109.4, 106.0, 104.3, 93.1, 55.5, 55.4; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{23}N_2O_5S$ [$(M+H)^+$], 463.1322, found, 463.1329.

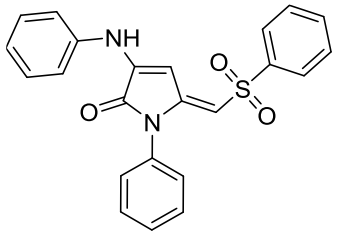
(*E*)-5-((Phenylsulfonyl)methylene)-1-(3,4,5-trimethoxyphenyl)-3-((3,4,5-trimethoxyphenyl)amino)-1,5-dihydro-*2H*-pyrrol-2-one.(4g)



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 8:1 , R_f = 0.2; Yellow solid: 244 mg (84%), mp = 198–199 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.89 (d, *J* = 7.6 Hz, 2H, ArH), 7.61–7.59 (m, 1H, ArH), 7.54–7.51 (m, 2H, ArH), 7.25 (s, 1H, C=CH), 6.96 (s, 1H, NH), 6.50 (s, 2H, ArH), 6.40 (s, 2H, ArH), 5.71 (s, 1H, C=CH), 3.93 (s, 6H, $ArOCH_3$), 3.88 (s, 6H, $ArOCH_3$), 3.81 (s, 6H, $ArOCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.6, 154.0, 154.0, 153.8, 153.8, 151.7, 142.6, 138.5, 135.9, 135.0, 134.4, 133.1, 129.3, 129.3, 127.7, 126.8, 126.8, 106.0, 105.5, 105.5, 96.2, 96.2, 92.7, 61.1, 60.9, 56.2,

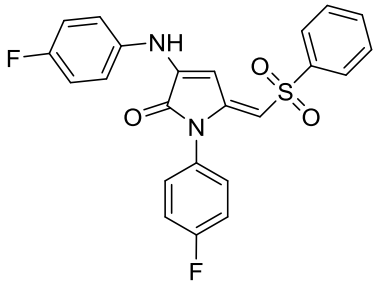
56.2, 56.2, 56.2; **HRMS** (TOF ES+): m/z calcd for $C_{29}H_{31}N_2O_9S$ [$(M+H)^+$], 583.1745, found, 583.1750.

(*E*)-1-Phenyl-3-(phenylamino)-5-((phenylsulfonyl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one (4h)



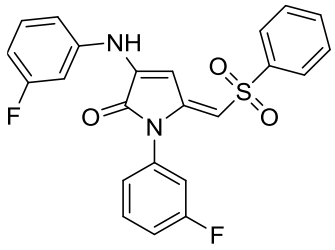
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 18:1, R_f = 0.2; Yellow solid: 177 mg (88%), mp = 204–205 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.90 (d, J = 7.6 Hz, 2H, ArH), 7.59–7.57 (m, 1H, A rH), 7.53–7.42 (m, 8H, ArH), 7.25–7.21 (m, 4H, ArH+C=CH), 7.16–7.13 (m, 2H, ArH+N H), 5.68 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.7, 151.7, 142.6, 139.0, 1 35.8, 133.1, 132.4, 129.8, 129.8, 129.8, 129.8, 129.3, 129.3, 129.1, 128.2, 128.2, 126.9, 12 6.9, 123.8, 118.3, 118.3, 105.8, 92.7; **HRMS** (TOF ES+): m/z calcd for $C_{23}H_{19}N_2O_5S$ [(M +H)$^+$], 403.1111, found, 403.1115.

(*E*)-1-(4-Fluorophenyl)-3-((4-fluorophenyl)amino)-5-((phenylsulfonyl)methylene)-1,5-dihydr o-*2H*-pyrrol-2-one. (4i)



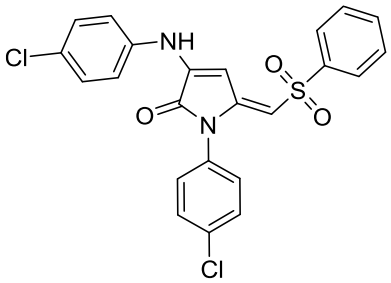
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 206 mg (94%), mp = 245–246 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.90–7.87 (m, 2H, ArH), 7.62–7.58 (m, 1H, ArH), 7.53 (dd, J = 8.4, 7.1 Hz, 2H, ArH), 7.23–7.19 (m, 4H, ArH+NH), 7.19–7.17 (m, 3H, ArH), 7.15–7.12 (m, 2H, ArH), 7.06 (s, 1H, C=CH), 5.61 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.6, 162.60 (d, J = 250.0 Hz), 159.07 (d, J = 244.4 Hz), 151.4, 142.4, 136.1, 135.0 (d, J = 2.8 Hz), 133.2, 130.1 (d, J = 8.9 Hz), 130.1 (d, J = 8.9 Hz), 129.4, 129.4, 128.2 (d, J = 3.2 Hz), 126.9, 126.9, 120.1 (d, J = 7.9 Hz), 120.1 (d, J = 7.9 Hz), 116.9 (d, J = 22.8 Hz), 116.9 (d, J = 22.8 Hz), 116.7 (d, J = 23.0 Hz), 116.7 (d, J = 23.0 Hz), 105.9, 92.3; **HRMS** (TOF ES+): m/z calcd for $C_{23}H_{17}N_2F_2O_3S$ [$(M+H)^+$], 439.0922, found, 439.0928.

(*E*)-1-(3-Fluorophenyl)-3-((3-fluorophenyl)amino)-5-((phenylsulfonyl)methylene)-1,5-dihydr o-*2H*-pyrrol-2-one. (4j).



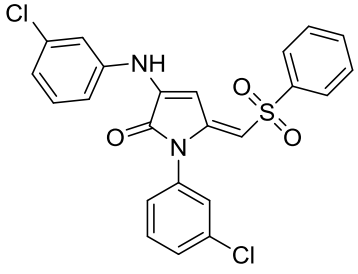
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 15:1 , R_f = 0.2; Yellow solid: 201 mg (92%), mp = 218–219 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.91 (d, *J* = 7.6 Hz, 2H, ArH), 7.62–7.60 (m, 1H, ArH), 7.56–7.53 (m, 2H, ArH), 7.49–7.45 (m, 1H, ArH), 7.41–7.37 (m, 1H, ArH), 7.29 (s, 1H, C=CH), 7.19–7.16 (m, 2H, ArH+NH), 7.04–7.02 (m, 2H, ArH), 6.97 (d, *J* = 8.9 Hz, 1H, ArH), 6.94 (d, *J* = 10.0 Hz, 1H, ArH), 5.74 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.2, 163.4 (*J* = 244.5 Hz), 162.8 (*J* = 247.5 Hz), 150.5, 142.2, 140.4 (*J* = 10.3 Hz), 135.3, 133.5 (*J* = 10.0 Hz), 133.3, 131.2 (*J* = 9.5 Hz), 131.0 (*J* = 9.0 Hz), 129.4, 129.4, 127.0, 127.0, 123.9, 116.5 (*J* = 20.8 Hz), 115.7 (*J* = 23.2 Hz), 113.9, 110.7 (*J* = 21.4 Hz), 106.8, 105.7 (*J* = 25.4 Hz), 94.0; **HRMS** (TOF ES+): m/z calcd for $C_{23}H_{17}N_2F_2O_3S$ [$(M+H)^+$], 439.0922, found, 439.0927.

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



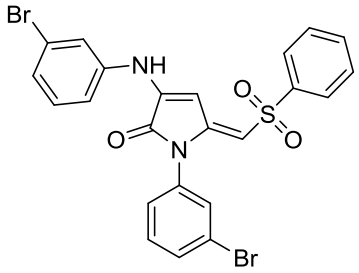
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 20:1 , R_f = 0.2; Yellow solid: 171mg (73%), mp = 245–246 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.90–7.89 (m, 2H, ArH), 7.62–7.59 (m, 1H, ArH), 7.54–7.52 (m, 2H, ArH), 7.47–7.45 (m, 2H, ArH), 7.39–7.38 (m, 2H, ArH), 7.25 (s, 1H, C=CH), 7.18–7.15 (m, 4H, ArH), 7.12 (s, 1H, NH), 5.66 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.4, 150.8, 142.2, 137.5, 135.5, 135.3, 133.3, 130.7, 130.1, 130.1, 129.9, 129.9, 129.4, 129.4, 129.4, 129.4, 129.0, 126.9, 126.9, 119.5, 119.5, 106.4, 93.3; **HRMS** (TOF ES+): m/z calcd for $C_{23}H_{17}Cl_2N_2O_3S$ [$(M+H)^+$], 471.0331, found, 471.0334.

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



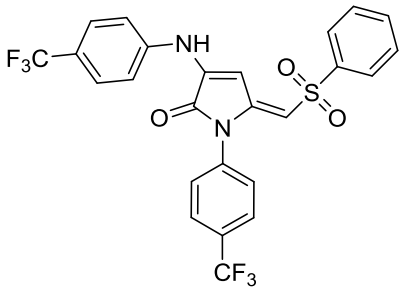
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1 , R_f = 0.2; Yellow solid: 162 mg (69%), mp = 213–214 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.92 (d, *J* = 7.6 Hz, 2H, ArH), 7.63–7.60 (m, 1H, ArH), 7.56–7.54 (m, 2H, ArH), 7.44–7.43 (m, 2H, ArH), 7.38–7.36 (m, 1H, ArH), 7.27 (s, 1H, C=CH), 7.24 (s, 1H, ArH), 7.20 (s, 1H, ArH), 7.16 (d, *J* = 8.1 Hz, 1H), 7.13–7.11 (m, 3H, ArH+NH), 5.72 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.2, 150.6, 142.2, 140.0, 135.4, 135.4, 135.3, 133.3, 133.3, 130.9, 130.7, 129.6, 129.4, 129.4, 128.4, 127.0, 127.0, 126.4, 124.0, 118.6, 116.2, 106.9, 94.1; **HRMS** (TOF ES+): m/z calcd for $C_{23}H_{17}Cl_2N_2O_3S$ [$(M+H)^+$], 471.0331, found, 471.0331.

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



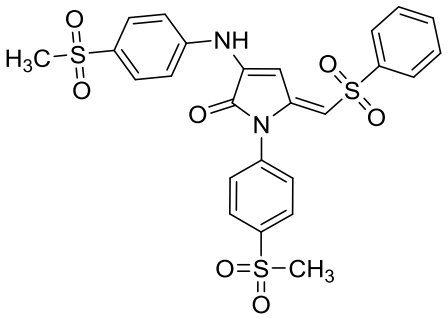
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 251 mg (90%), mp = 212–214 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.92 (d, *J* = 7.3 Hz, 2H, ArH), 7.62–7.54 (m, 4H, ArH+NH), 7.39–7.36 (m, 3H, ArH), 7.32–7.29 (m, *J* = 7.8 Hz, 3H, ArH), 7.21 (d, *J* = 7.0 Hz, 1H, ArH), 7.17 (d, *J* = 7.4 Hz, 1H, ArH), 7.05 (s, 1H, C=CH), 5.72 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.1, 150.6, 142.2, 140.1, 135.2, 133.4, 133.3, 132.5, 131.3, 131.2, 131.0, 129.4, 129.4, 127.0, 127.0, 126.9, 126.8, 123.4, 123.1, 121.5, 116.7, 106.9, 94.1; **HRMS** (TOF ES+): m/z calcd for $C_{23}H_{17}Br_2N_2O_3S$ [$(M+H)^+$], 558.9321, found, 558.9323.

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



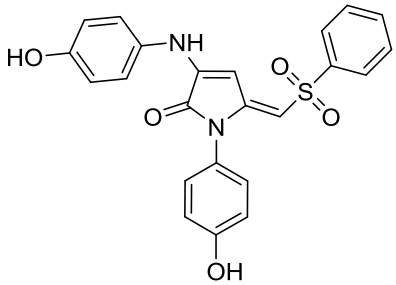
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 11:1, R_f = 0.2; Yellow solid: 207 mg (77%), mp = 271–273 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.91 (d, *J* = 7.7 Hz, 1H, ArH), 7.77 (d, *J* = 8.1 Hz, 1H, ArH), 7.69 (d, *J* = 8.2 Hz, 1H, ArH), 7.62 (s, 1H, NH), 7.56–7.53 (m, 1H, ArH), 7.45 (s, 1H, C=CH), 7.38 (d, *J* = 8.1 Hz, 1H, ArH), 7.33 (d, *J* = 8.2 Hz, 1H, ArH), 5.75 (s, 1H, C=CH); **^{13}C NMR** (600 MHz, $CDCl_3$): δ = 165.1, 149.8, 141.9, 141.9, 135.4, 134.8, 133.4, 131.2 (d, *J* = 34.1 Hz), 129.4, 129.4, 128.4, 128.4, 127.1, 127.1, 127.0, 127.0, 127.0, 127.0, 125.52 (d, *J* = 33.0 Hz), 123.9 (d, *J* = 268.5 Hz), 123.4 (d, *J* = 274.5 Hz), 117.8, 117.8, 107.4, 95.1; **HRMS** (TOF ES+): m/z calcd for $C_{27}H_{17}N_2F_6O_3S$ [$(M+H)^+$], 539.0859, found, 539.0858.

(*E*)-1-(4-(Methylsulfonyl)phenyl)-3-((4-(methylsulfonyl)phenyl)amino)-5-((phenylsulfonyl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one. (4o).



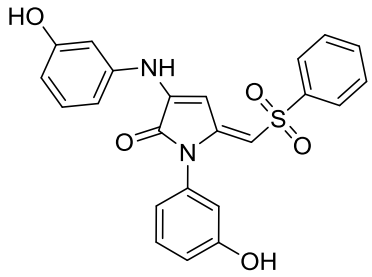
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 5:1, R_f = 0.2; Yellow solid: 131 mg (47%), mp = 304–306 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 10.04 (s, 1H, NH), 8.09 (d, J = 8.4 Hz, 2H, ArH), 8.01 (d, J = 8.7 Hz, 2H, ArH), 7.97 (d, J = 7.5 Hz, 2H, ArH), 7.71–7.68 (m, 1H, ArH), 7.66–7.64 (m, 3H, ArH), 7.62 (d, J = 8.4 Hz, 3H, ArH), 7.29 (s, 1H, C=CH), 5.94 (s, 1H, C=CH), 3.31 (s, 3H, CH_3), 3.24 (s, 3H, CH_3); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.0, 150.8, 145.2, 142.8, 141.2, 137.5, 136.9, 134.7, 133.9, 130.1, 130.1, 129.9, 129.9, 129.4, 129.4, 129.0, 129.0, 127.2, 127.2, 119.1, 119.1, 105.9, 95.4, 44.2, 43.8; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{23}N_2O_7S_3$ [$(M+H)^+$], 559.0662, found, 559.0662.

(*E*)-1-(4-Hydroxyphenyl)-3-((4-hydroxyphenyl)amino)-5-((phenylsulfonyl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one.(4p)



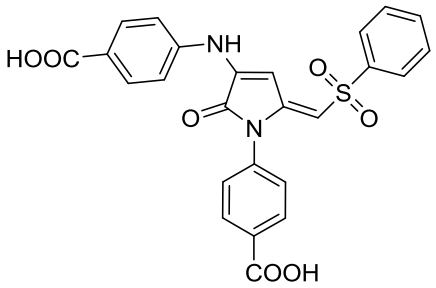
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 2:1 , R_f = 0.2; Orange solid: 169 mg (78%), mp = 241–243 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 9.91 (s, 1H, NH), 9.49 (s, 1H, ArOH), 9.35 (s, 1H, ArOH), 7.86 (d, J = 7.7 Hz, 2H, ArH), 7.68–7.65 (m, 1H, ArH), 7.61–7.59 (m, 2H, ArH), 7.21 (d, J = 8.4 Hz, 2H, ArH), 7.09 (d, J = 8.2 Hz, 2H, ArH), 6.87–6.84 (m, 4H, ArH), 6.71 (s, 1H, C=CH), 5.35 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.6, 158.3, 154.3, 153.6, 143.5, 138.9, 133.5, 131.9, 130.3, 130.3, 130.0, 130.0, 126.8, 126.8, 123.8, 121.7, 121.7, 116.5, 116.5, 116.4, 116.4, 102.3, 88.9; **HRMS** (TOF ES+): m/z calcd for $C_{23}H_{19}N_2O_5S$ [$(M+H)^+$], 435.1009, found, 435.1010.

(*E*)-1-(3-hydroxyphenyl)-3-((3-hydroxyphenyl)amino)-5-((phenylsulfonyl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one. (4q)



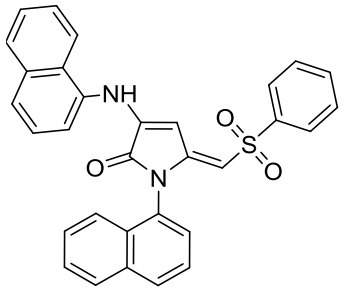
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 1:1, R_f = 0.2; Yellow solid: 163 mg (75%), mp =251–252 °C; **^{1}H NMR** (500 MHz, DMSO-d_6): δ = 9.91 (s, 1H, NH), 9.74 (s, 1H, ArOH), 9.44 (s, 1H, ArOH), 7.89 (d, *J* = 7.2 Hz, 2H, ArH), 7.67–7.66 (m, 1H, ArH), 7.62–7.61 (m, 2H, ArH), 7.31 (t, *J* = 7.7 Hz, 1H, ArH), 7.22 (t, *J* = 7.6 Hz, 1H, ArH), 6.95 (s, 1H, C=CH), 6.88–6.84 (m, 3H, ArH), 6.71–6.70 (m, 1H, ArH), 6.67 (s, 1H, ArH), 6.53 (d, *J* = 8.2 Hz, 1H, ArH), 5.53 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.2, 158.6, 158.6, 152.5, 143.2, 141.6, 137.9, 133.9, 133.7, 130.8, 130.6, 130.1, 130.1, 126.9, 126.9, 119.3, 116.5, 115.8, 111.1, 110.5, 106.6, 103.5, 91.6; **HRMS** (TOF ES+): m/z calcd for $C_{23}H_{19}N_2O_5S$ [$(M+H)^+$], 435.1009, found,435.1010.

(*E*)-4-((1-(4-Carboxyphenyl)-2-oxo-5-((phenylsulfonyl)methylene)-2,5-dihydro-*1H*-pyrrol-3-yl)amino)benzoic acid. (4r)



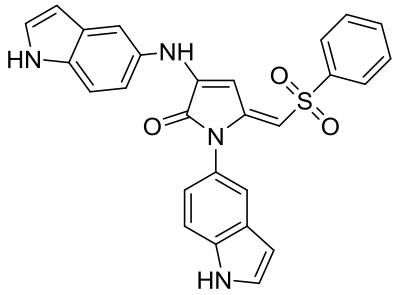
$V_{Methanol}/V_{Ethyl\ acetate}$ = 2:1, R_f = 0.2; Yellow solid: 125 mg (51%), mp = 343–345 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 9.89 (s, 1H, NH), 8.08 (d, *J* = 8.2 Hz, 2H, ArH), 8.02 (d, *J* = 8.4 Hz, 2H, ArH), 7.94 (d, *J* = 7.8 Hz, 2H, ArH), 7.69–7.67 (m, 1H, ArH), 7.62–7.60 (m, 2H, ArH), 7.48 (dd, *J* = 13.9, 8.3 Hz, 5H, ArH), 7.20 (s, 1H, C=CH), 5.81 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 167.3, 167.1, 165.0, 151.2, 144.6, 142.9, 137.0, 136.9, 133.8, 131.5, 131.5, 131.1, 131.1, 130.1, 130.1, 129.1, 129.1, 127.2, 127.2, 125.3, 118.7, 118.7, 118.7, 105.3, 94.5; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{19}N_2O_7S$ [$(M+H)^+$], 491.0907, found, 491.0908.

(*E*)-1-(Naphthalen-1-yl)-3-(naphthalen-1-ylamino)-5-((phenylsulfonyl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one. (4s)



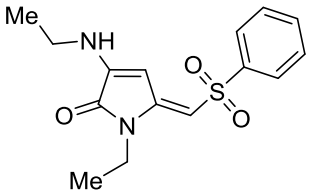
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1 , R_f = 0.2; Yellow solid: 93mg (37%), mp = 219–220 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 8.03–8.02 (m, 1H, ArH), 7.99 (d, *J* = 8.3 Hz, 1H, ArH), 7.96–7.93 (m, 2H, ArH), 7.81–7.79 (m, 2H, ArH), 7.75 (d, *J* = 8.1 Hz, 1H, ArH), 7.68–7.67 (m, 1H, ArH), 7.63–7.59 (m, 3H, ArH), 7.58–7.54 (m, 4H, ArH), 7.48–7.42 (m, 5H, ArH+NH), 7.27 (s, 1H, C=CH), 5.41 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 166.2, 152.1, 142.6, 137.2, 134.6, 134.4, 134.1, 133.0, 130.5, 130.4, 129.2, 129.2, 129.0, 128.8, 128.7, 127.8, 127.6, 127.0, 126.8, 126.8, 126.7, 126.7, 126.1, 126.0, 125.6, 125.1, 122.3, 120.4, 116.1, 106.5, 93.2; **HRMS** (TOF ES+): m/z calcd for $C_{31}H_{23}N_2O_3S$ [$(M+H)^+$], 503.1424, found, 503.1429.

(*E*)-3-((1*H*-Indol-5-yl)amino)-1-(1*H*-indol-5-yl)-5-((phenylsulfonyl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one. (4t)



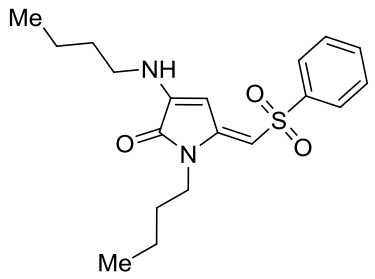
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 13:1 , R_f = 0.2; Orange yellow solid: 120 mg (50%), mp = 267–268 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 11.42 (s, 1H, NH), 11.22 (s, 1H, NH), 9.45 (s, 1H, NH), 7.84 (d, *J* = 7.8 Hz, 2H, ArH), 7.67–7.74 (m, 1H, ArH), 7.60–7.58 (m, 2H, ArH), 7.54–7.52 (m, 1H, ArH), 7.49–7.47 (m, 4H, ArH), 7.42–7.41 (m, 1H, ArH), 7.23 (d, *J* = 8.6 Hz, 1H, C=CH), 6.96 (d, *J* = 8.4 Hz, 1H, C=CH), 6.82 (s, 1H, C=CH), 6.50 (d, *J* = 12.0 Hz, 2H, C=CH), 5.33 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 166.0, 154.4, 143.6, 139.4, 136.0, 133.5, 133.4, 132.3, 130.0, 130.0, 128.3, 128.3, 127.6, 127.1, 126.7, 126.7, 124.3, 121.6, 121.0, 115.6, 112.7, 112.6, 111.3, 102.1, 102.1, 101.7, 88.6; **HRMS** (TOF ES+): m/z calcd for $C_{27}H_{21}N_4O_3S$ [$(M+H)^+$], 481.1329, found, 481.1334.

(*E*)-1-Ethyl-3-(ethylamino)-5-((phenylsulfonyl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one. (4u)



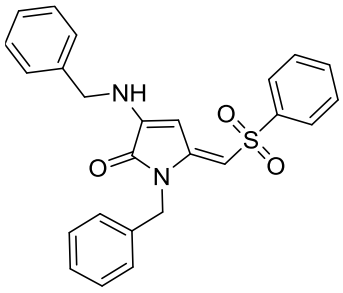
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 8:1, R_f = 0.2; Light yellow solid: 61 mg (40%), mp = 152–154 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 7.92 (d, *J* = 7.6 Hz, 2H, ArH), 7.67–7.65 (m, 1H, ArH), 7.63–7.61 (m, 2H, ArH), 7.31 (t, *J* = 5.5 Hz, 1H, NH), 6.19 (s, 1H, C=CH), 5.99 (s, 1H, C=CH), 3.55(q, *J* = 6.9 Hz, 2H, CH_2), 3.16–3.11 (m, 2H, CH_2), 1.15–1.12 (t, *J* = 7.2 Hz, 3H, CH_3), 0.98–0.96 (t, *J* = 7.0 Hz, 3H, CH_3); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.3, 151.5, 144.1, 144.0, 133.2, 129.9, 129.9, 126.6, 126.6, 100.6, 85.8, 38.8, 34.2, 14.3, 13.8; **HRMS** (TOF ES+): m/z calcd for $C_{15}H_{19}N_2O_3S$ [$(M+H)^+$], 307.1111, found, 307.1111.

(*E*)-1-Butyl-3-(butylamino)-5-((phenylsulfonyl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one. (4v)



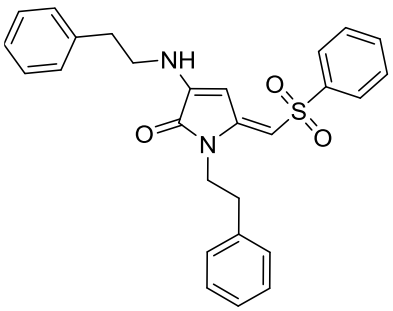
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 83 mg (46%), mp = 152–154 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 7.90 (d, J = 7.3 Hz, 2H, ArH), 7.67–7.65 (m, 1H, ArH), 7.62–7.60 (m, 2H, ArH), 7.32 (t, J = 5.8 Hz, 1H, NH), 6.15 (s, 1H, C=CH), 5.97 (s, 1H, C=CH), 3.51–3.48 (t, J = 7.1 Hz, 2H, CH_2), 3.11–3.07 (q, J = 6.8 Hz, 2H, CH_2), 1.54–1.49 (m, 2H, CH_2), 1.37–1.35 (m, 2H, CH_2), 1.33–1.29 (m, 2H, CH_2), 1.19–1.15 (m, 2H, CH_2), 0.89 (t, J = 7.1 Hz, 3H, CH_3), 0.82 (t, J = 7.1 Hz, 3H, CH_3); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.5, 152.0, 144.1, 144.0, 133.2, 129.8, 129.8, 126.6, 126.6, 100.7, 85.6, 43.7, 39.0, 30.8, 30.3, 20.1, 19.7, 14.2, 14.1; **HRMS** (TOF ES+): m/z calcd for $C_{19}H_{27}N_2O_3S$ [$(M+H)^+$], 363.1737, found, 363.1738.

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



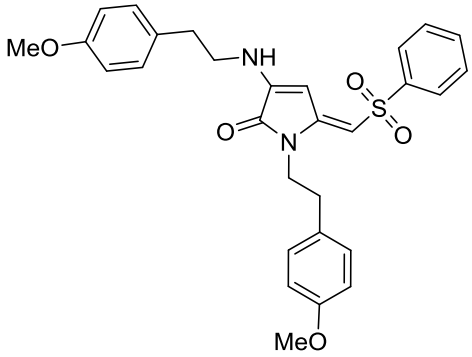
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 6:1, R_f = 0.2; Yellow solid: 77 mg (36%), mp = 178–179 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 8.02 (t, J = 5.9 Hz, 1H, NH), 7.66 (d, J = 7.7 Hz, 2H, ArH), 7.63–7.60 (m, 1H, ArH), 7.53–7.50 (m, 2H, ArH), 7.40–7.37 (m, 2H, ArH), 7.34–7.32 (m, 3H, ArH), 7.29–7.27 (m, 2H, ArH), 7.24–7.22(m, 1H, ArH), 7.11 (d, J = 7.4 Hz, 2H, ArH), 6.08 (s, 1H, C=CH), 5.97 (s, 1H, C=CH), 4.74 (s, 2H, CH_2), 4.36 (d, J = 5.9 Hz, 2H, CH_2); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.8, 151.5, 143.8, 143.7, 138.2, 137.2, 133.2, 129.8, 129.8, 129.0, 129.0, 129.0, 129.0, 127.9, 127.9, 127.8, 127.7, 127.2, 127.2, 126.5, 126.5, 102.1, 87.8, 47.4, 42.4; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{23}N_2O_3S$ [$(M+H)^+$], 431.1424, found, 431.1428.

(*E*)-1-Phenethyl-3-(phenethylamino)-5-((phenylsulfonyl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one. (4x)



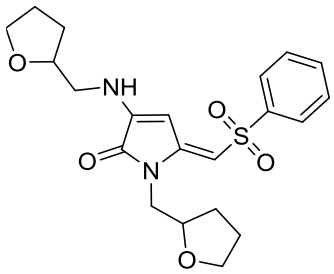
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 4:1, R_f = 0.2; Yellow solid: 105 mg (46%), mp = 148–150 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 7.87 (d, J = 7.8 Hz, 2H, ArH), 7.68–7.66 (m, 1H, ArH), 7.64–7.61 (m , 2H, ArH), 7.32–7.30 (m, 2H, ArH), 7.26 (t, J = 5.7 Hz, 1H, NH), 7.23 (d, J = 7.3 Hz, 3H, ArH), 7.18–7.15 (m, 2H, ArH), 7.12 (d, J = 7.1 Hz, 1H, ArH), 7.07 (d, J = 7.3 Hz, 2H, ArH), 6.15 (s, 1H, C=CH), 6.01 (s, 1H, C=CH), 3.75 (t, J = 7.0 Hz, 2H, CH_2), 3.35–3.31 (m, 2H, CH_2), 2.83 (t, J = 7.2 Hz, 2H, CH_2), 2.70 (t, J = 7.0 Hz, 2H, CH_2); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.3, 151.7, 144.0, 143.6, 139.5, 138.5, 133.2, 129.8, 129.8, 129.2, 129.2, 129.1, 129.1, 128.8, 128.8, 128.6, 128.6, 126.8, 126.7, 126.7, 126.7, 101.2, 86.3, 45.5, 40.6, 34.4, 34.0; **HRMS** (TOF ES+): m/z calcd for $C_{27}H_{27}N_2O_3S$ [$(M+H)^+$], 459.1737, found, 459.1738.

(*E*)-1-(4-Methoxyphenethyl)-3-((4-methoxyphenethyl)amino)-5-((phenylsulfonyl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one. (4y)



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Orange solid: 130 mg (50%), mp = 139–141 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 7.88 (d, J = 7.3 Hz, 2H, ArH), 7.68–7.65 (m, J = 7.3 Hz, 1H, ArH), 7.63–7.60 (m, 2H, ArH), 7.24 (t, J = 5.8 Hz, 1H, NH), 7.14–7.10 (m, 2H, ArH), 6.99 (d, J = 8.2 Hz, 2H, ArH), 6.87 (d, J = 8.5 Hz, 2H, ArH), 6.74 (d, J = 8.4 Hz, 2H, ArH), 6.18 (s, 1H, C=CH), 5.99 (s, 1H, C=CH), 3.72 (s, 3H, $ArOCH_3$), 3.70 (m, 2H, CH_2), 3.68 (s, 3H, $ArOCH_3$), 3.29–3.26 (m, 2H, CH_2), 2.76 (t, J = 7.3 Hz, 2H, CH_2), 2.63 (t, J = 7.2 Hz, 2H, CH_2); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.3, 158.2, 158.2, 151.7, 144.1, 143.6, 133.2, 131.3, 130.3, 130.2, 130.2, 130.1, 130.1, 129.8, 129.8, 126.6, 126.6, 114.2, 114.2, 114.0, 114.0, 101.1, 86.2, 55.4, 55.4, 45.8, 40.8, 33.6, 33.1; **HRMS** (TOF ES+): m/z calcd for $C_{29}H_{31}N_2O_5S$ [$(M+H)^+$], 519.1948, found, 519.1948.

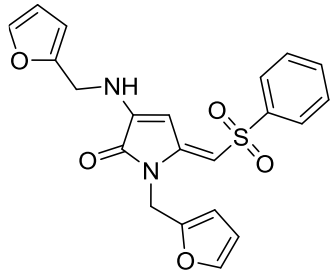
(*E*)-5-((Phenylsulfonyl)methylene)-1-((tetrahydrofuran-2-yl)methyl)-3-(((tetrahydrofuran-2-yl)methyl)amino)-1,5-dihydro-*2H*-pyrrol-2-one. (4z)



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 3:1, R_f = 0.2; Yellow solid: 107 mg (51%), mp = 151–153 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 7.89 (d, J = 7.4 Hz, 2H, ArH), 7.66–7.60 (m, 3H, ArH), 7.20 (t, J = 6.2 Hz, 1H, NH), 6.20 (s, 1H, C=CH), 6.09 (s, 1H, C=CH), 4.02–3.98 (m, 1H, CH), 3.93–3.89 (m, 1H,

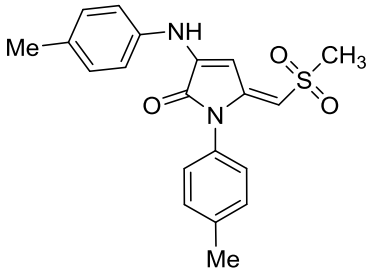
CH), 3.77–3.73 (m, 1H, CH_2), 3.65–3.61 (m, 1H, CH_2), 3.60–3.55 (m, 2H, CH_2), 3.53–3.48 (m, 2H, CH_2), 3.20–3.11 (m, 2H, CH_2), 1.93–1.89 (m, 1H, CH_2), 1.87–1.81 (m, 3H, CH_2), 1.77–1.74 (m, 1H, CH_2), 1.73–1.69 (m, 1H, CH_2), 1.60–1.56 (m, 1H, CH_2), 1.47–1.43 (m, 1H, CH_2); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.8, 152.2, 144.1, 144.0, 133.2, 129.8, 129.8, 126.6, 126.6, 101.7, 86.7, 77.1, 76.4, 67.6, 67.4, 48.3, 43.32, 29.1, 28.7, 25.6, 25.3; **HRMS** (TOF ES+): m/z calcd for $C_{21}H_{27}N_2O_5S$ [$(M+H)^+$], 419.1635, found, 419.1635.

(*E*)-1-(Furan-2-ylmethyl)-3-((furan-2-ylmethyl)amino)-5-((phenylsulfonyl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one. (4a')



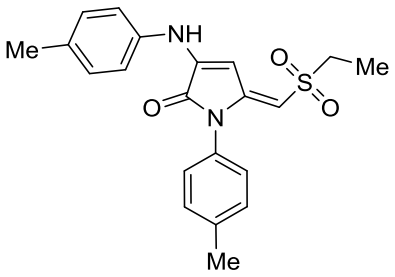
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 3:1, R_f = 0.2; Light yellow solid: 84 mg (41%), mp = 150–152 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 7.82–7.80 (m, 3H, ArH+NH), 7.66–7.64 (m, 1H, ArH), 7.63–7.57 (m, 3H, ArH+C=CH), 7.53–7.51 (m, 1H, C=CH), 6.46–6.45 (m, 1H, C=CH), 6.36–6.34 (m, 2H, C=CH), 6.31 (d, *J* = 2.9 Hz, 1H, C=CH), 6.27 (s, 1H, C=CH), 6.12 (s, 1H, C=CH), 4.75 (s, 2H, CH_2), 4.33 (m, 2H, CH_2); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.2, 151.1, 151.0, 149.9, 143.8, 143.2, 143.2, 143.1, 133.3, 129.9, 129.9, 126.6, 126.6, 110.9, 110.9, 108.8, 108.7, 102.4, 88.3, 40.5, 36.0; **HRMS** (TOF ES+): m/z calcd for $C_{21}H_{19}N_2O_5S$ [$(M+H)^+$], 411.1009, found, 411.1009.

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



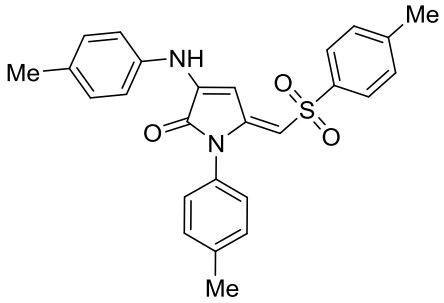
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 18:1, R_f = 0.2; Yellow solid: 153 mg (83%), mp = 242–244 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.32 (d, *J* = 7.9 Hz, 2H, ArH), 7.20 (s, 1H, NH), 7.19 (s, 1H, ArH), 7.15 (d, *J* = 8.1 Hz, 2H, ArH), 7.13–7.10 (m, 3H, ArH), 6.96 (s, 1H, C=CH), 5.62 (s, 1H, C=CH), 3.04 (s, 3H, CH_3), 2.43 (s, 3H, $ArCH_3$), 2.35 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.9, 153.1, 139.5, 136.3, 136.2, 133.6, 130.5, 130.5, 130.3, 130.3, 129.7, 128.0, 128.0, 118.4, 118.4, 104.1, 91.4, 45.4, 21.3, 20.9; **HRMS** (TOF ES+): m/z calcd for $C_{20}H_{21}N_2O_3S$ [$(M+H)^+$], 369.1267, found, 369.1268.

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



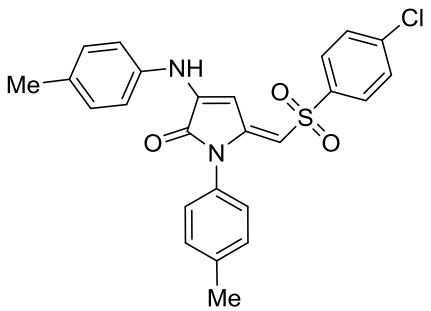
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 4:1, R_f = 0.2; Yellow solid: 31 mg (0.1 mmol, 80%), mp = 221–223 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 9.39 (s, 1H, NH), 7.37 (d, J = 8.0 Hz, 2H, ArH), 7.28–7.25 (m, 4H, ArH), 7.22 (d, J = 8.2 Hz, 2H, ArH), 6.80 (s, 1H, C=CH), 5.32 (s, 1H, C=CH), 3.15 (q, J = 7.4 Hz, 2H, CH_2), 2.38 (s, 3H, $ArCH_3$), 2.28 (s, 3H, $ArCH_3$), 1.16 (t, J = 7.3 Hz, 3H, CH_3); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 165.6, 153.1, 139.0, 138.0, 137.8, 132.9, 130.6, 130.6, 130.3, 130.3, 128.8, 128.8, 119.7, 119.7, 119.7, 101.2, 91.4, 50.7, 21.2, 20.9, 7.8; **HRMS** (TOF ES+): m/z calcd for $C_{21}H_{23}N_2O_3S$ [$(M+H)^+$], 383.1424, found, 383.1423.

(*E*)-1-(*p*-Tolyl)-3-(*p*-tolylamino)-5-(tosylmethylene)-1,5-dihydro-*2H*-pyrrol-2-one. (5c)



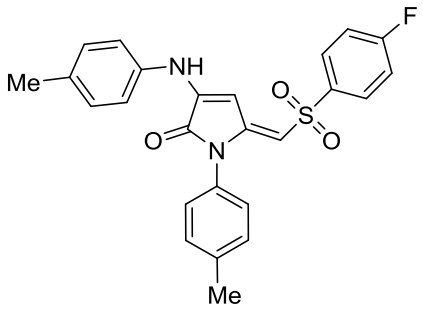
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 18:1 , R_f = 0.2; Yellow solid: 200mg (90%), mp = 239–241 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.77 (d, J = 8.3 Hz, 2H, ArH), 7.30 (d, J = 8.0 Hz, 2H, ArH), 7.27–7.26 (m, 2H, ArH), 7.23 (d, J = 8.1 Hz, 2H, ArH), 7.16–7.15 (m, 2H, ArH), 7.13 (s, 1H, C=CH), 7.08 (d, J = 8.2 Hz, 2H, ArH), 7.03 (s, 1H, NH), 5.63 (s, 1H, C=CH), 2.41 (s, 3H, CH_3), 2.40 (s, 3H, CH_3), 2.37 (s, 3H, CH_3); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 165.8, 151.6, 143.9, 139.8, 139.2, 136.5, 135.9, 133.5, 130.3, 130.3, 130.3, 130.3, 129.9, 129.9, 129.7, 127.9, 127.9, 126.9, 126.9, 118.3, 118.3, 105.8, 92.0, 21.6, 21.2, 20.9; **HRMS** (TOF ES+): m/z calcd for $C_{26}H_{25}N_2O_3S$ [$(M+H)^+$], 445.1580, found, 445.1580.

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



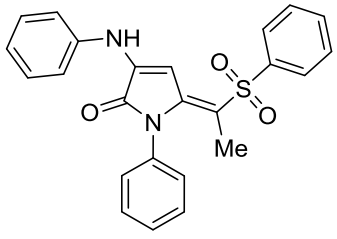
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 18:1, R_f = 0.2; Yellow solid: 218 mg (94%), mp = 264–266 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.82 (d, *J* = 8.6 Hz, 2H, ArH), 7.47 (d, *J* = 8.6 Hz, 2H, ArH), 7.28 (s, 1H, NH), 7.27 (s, 1H, ArH), 7.23 (d, *J* = 8.1 Hz, 2H, ArH), 7.14 (d, *J* = 8.3 Hz, 2H, ArH), 7.11 (s, 1H, C=CH), 7.08 (d, *J* = 8.0 Hz, 3H, ArH), 5.58 (s, 1H, C=CH), 2.40 (s, 3H, $ArCH_3$), 2.37 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 163.3, 150.1, 138.9, 137.1, 137.0, 133.9, 133.9, 131.3, 128.0, 128.0, 127.9, 127.9, 127.9, 127.1, 127.1, 125.9, 125.9, 125.5, 125.5, 116.0, 116.0, 102.2, 89.2, 18.8, 18.5; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{22}ClN_2O_3S$ [$(M+H)^+$], 465.1034, found, 465.1033.

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



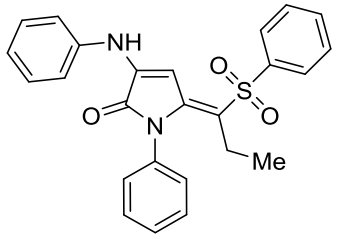
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Brown yellow solid: 190 mg (85%), mp = 245–247 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 7.92–7.85 (m, 2H, ArH), 7.28 (s, 1H, NH), 7.27 (s, 1H, ArH), 7.23 (d, *J* = 8.1 Hz, 2H, ArH), 7.19–7.16 (m, 2H, ArH), 7.15–7.12 (m, 3H, ArH), 7.11–7.07 (m, 2H, ArH), 7.05 (s, 1H, C=CH), 5.59 (s, 1H, C=CH), 2.40 (s, 3H, $ArCH_3$), 2.38 (s, 3H, $ArCH_3$); **^{13}C NMR** (600 MHz, $CDCl_3$): δ = 163.7, 163.2 (*J* = 253.5 Hz), 150.1, 137.3, 136.8 (d, *J* = 3.1 Hz), 134.3, 134.1, 131.6, 128.3, 128.3, 128.2, 128.2, 127.5 (d, *J* = 9.2 Hz), 127.5 (d, *J* = 9.2 Hz), 127.5, 125.9, 125.9, 116.3, 116.3, 114.4 (d, *J* = 22.6 Hz), 114.4 (d, *J* = 22.6 Hz), 102.9, 89.6, 19.2, 18.8; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{22}N_2FO_3S$ [$(M+H)^+$], 449.1330, found, 449.1334.

(*E*)-1-Phenyl-3-(phenylamino)-5-(1-(phenylsulfonyl)ethylidene)-1,5-dihydro-*2H*-pyrrol-2-one (6a)



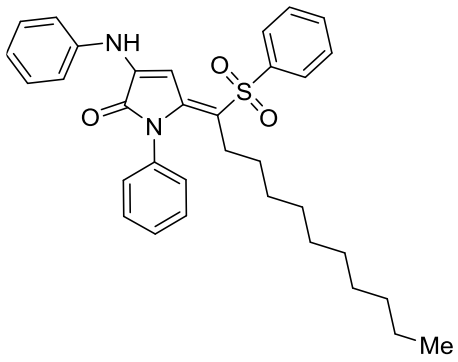
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 13:1, R_f = 0.2; Yellow solid: 158 mg (76%), mp = 216–217 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 9.24 (s, 1H, NH), 7.89 (d, *J* = 7.5 Hz, 2H, ArH), 7.73–7.71 (m, 1H, ArH), 7.68–7.65 (m, 2H, ArH), 7.51–7.48 (m, 2H, ArH), 7.46–7.42 (m, 3H, ArH), 7.40 (s, 1H, C=CH), 7.34 (d, *J* = 7.8 Hz, 2H, ArH), 7.30 (d, *J* = 7.8 Hz, 2H, ArH), 7.07 (t, *J* = 7.3 Hz, 1H, ArH), 1.42 (s, 3H, C-CH_3).**^{13}C NMR** (150 MHz, DMSO-d_6): δ = 167.4, 147.3, 141.7, 141.0, 136.8, 135.5, 133.9, 130.2, 130.2, 129.9, 129.9, 129.9, 129.9, 129.1, 129.0, 129.0, 127.1, 127.1, 123.1, 119.0, 119.0, 114.0, 97.4, 15.7; **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-(1-(phenylsulfonyl)propylidene)-1,5-dihydro-*2H*-pyrrol-2-one. (6b)



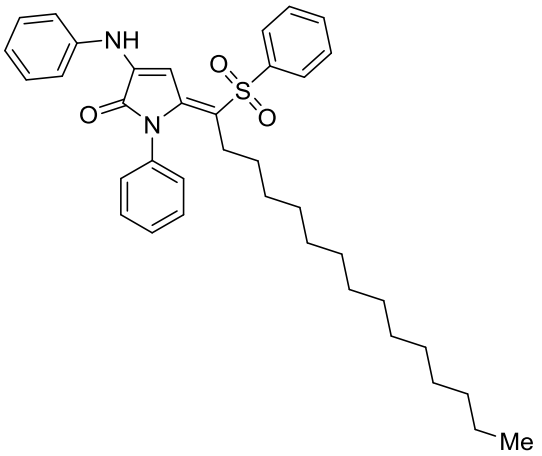
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 146 mg (68%), mp = 183–184 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 9.24 (s, 1H, NH), 7.90 (d, J = 7.3 Hz, 2H, ArH), 7.73–7.67 (m, 3H, ArH), 7.52–7.47 (m, 3H, ArH), 7.43–7.40 (m, 4H, ArH+C=CH), 7.28–7.26 (m, 3H, ArH), 7.07 (t, J = 7.3 Hz, 1H, ArH), 1.94 (q, J = 7.2 Hz, 2H, CH_2), 0.61 (t, J = 7.2 Hz, 3H, CH_3); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 167.5, 147.7, 142.9, 140.9, 136.8, 135.3, 133.7, 130.1, 130.1, 129.9, 129.9, 129.9, 129.5, 129.5, 129.5, 129.5, 127.0, 127.0, 123.1, 120.7, 119.0, 119.0, 97.6, 21.3, 15.2; **HRMS** (TOF ES+): m/z calcd for $C_{25}H_{23}N_2O_3S$ [$(M+H)^+$], 431.1424, found, 431.1434.

(*E*)-1-Phenyl-3-(phenylamino)-5-(1-(phenylsulfonyl)undecylidene)-1,5-dihydro-*2H*-pyrrol-2-one. (6c)



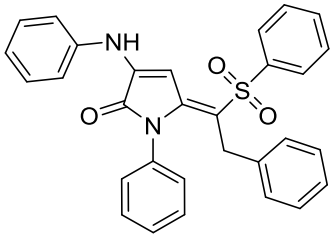
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 15:1, R_f = 0.2; Light yellow solid: 217 mg (80%), mp = 154–156 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 9.21 (s, 1H, NH), 7.89 (d, J = 7.4 Hz, 2H, ArH), 7.74–7.68 (m, 1H, ArH), 7.68–7.66 (m, 2H, ArH), 7.51–7.46 (m, 3H, ArH), 7.47–7.46 (m, 4H, ArH), 7.26 (s, 2H, ArH), 7.24 (s, 1H, C=CH), 7.06 (t, J = 7.3 Hz, 1H, ArH), 1.88–1.85 (m, 2H, CH_2), 1.27–1.18 (m, 6H, CH_2), 1.14–1.10 (m, 2H, CH_2), 1.07–1.01 (m, 4H, CH_2), 0.92–0.89 (m, 2H, CH_2), 0.72–0.84 (t, J = 7.1 Hz, 3H, CH_3), 0.55–0.51 (m, 2H, CH_2); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 167.5, 147.5, 142.9, 140.9, 136.6, 135.2, 133.7, 130.1, 130.1, 129.8, 129.8, 129.8, 129.5, 129.5, 129.4, 127.0, 123.1, 119.8, 118.9, 118.9, 97.6, 31.7, 31.7, 30.4, 29.3, 29.2, 29.1, 28.7, 28.2, 22.6, 22.6, 14.5, 14.5; **HRMS** (TOF ES+): m/z calcd for $C_{33}H_{39}N_2O_3S$ [$(M+H)^+$], 543.2676, found, 543.2677.

(*E*)-1-Phenyl-3-(phenylamino)-5-(1-(phenylsulfonyl)pentadecylidene)-1,5-dihydro-*2H*-pyrrol-2-one. (6d)



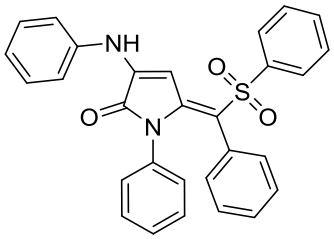
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 15:1, R_f = 0.2; Light yellow solid: 188 mg (63%), mp = 158–160 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 9.21 (s, 1H, NH), 7.88 (d, J = 7.4 Hz, 2H, ArH), 7.72–7.69 (m, 1H, ArH), 7.72–7.69 (m, 2H, ArH), 7.49 (d, J = 7.4 Hz, 3H, ArH), 7.42–7.37 (m, 4H, ArH), 7.22 (s, 2H, ArH), 7.24 (s, 1H, ArH, C=CH), 7.06 (t, J = 7.3 Hz, 1H, ArH), 1.87–1.85 (m, 2H, CH_2), 1.27–1.18 (m, 15H, CH_3+CH_2), 1.14–1.10 (m, 2H, CH_2), 1.06–1.03 (m, 4H, CH_2), 0.92–0.88 (m, 2H, CH_2), 0.86–0.83 (m, 2H, CH_2), 0.56–0.51 (m, 2H, CH_2); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 167.5, 147.58, 142.9, 140.9, 136.6, 135.2, 133.7, 130.0, 130.0, 129.8, 129.8, 129.8, 129.8, 129.5, 129.5, 129.3, 126.9, 126.9, 123.1, 119.8, 118.9, 118.9, 97.6, 31.8, 30.4, 29.5, 29.5, 29.5, 29.5, 29.4, 29.3, 29.2, 29.2, 28.7, 28.2, 22.6, 14.5; **HRMS** (TOF ES+): m/z calcd for $C_{37}H_{47}N_2O_3S$ [$(M+H)^+$], 599.3302, found, 599.3301.

(*E*)-1-Phenyl-5-(2-phenyl-1-(phenylsulfonyl)ethylidene)-3-(phenylamino)-1,5-dihydro-*2H*-pyrrol-2-one. (6e)



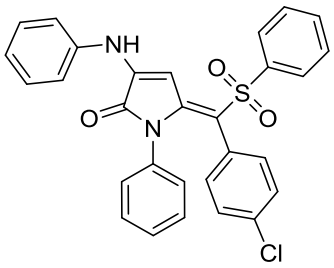
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 13:1, R_f = 0.2; Yellow solid: 123 mg (50%), mp = 205–207 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 9.34 (s, 1H, NH), 7.75 (d, J = 7.7 Hz, 2H, ArH), 7.66–7.63 (m, 1H, ArH), 7.59–7.56 (m, 2H, ArH), 7.46–7.44 (m, 2H, ArH), 7.36–7.32 (m, 2H, ArH), 7.31 (s, 1H, C=CH), 7.30–7.24 (m, 3H, ArH), 7.11 (d, J = 7.6 Hz, 6H, ArH), 6.78 (d, J = 6.5 Hz, 2H, ArH), 3.45 (s, 2H, CH_2); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 167.4, 149.2, 142.9, 140.8, 138.7, 135.9, 135.7, 133.7, 129.9, 129.9, 129.9, 129.5, 129.5, 129.3, 129.3, 129.2, 129.2, 128.4, 128.4, 128.1, 128.1, 127.1, 127.1, 126.4, 123.3, 119.2, 119.2, 117.4, 97.3, 33.2; **HRMS** (TOF ES+): m/z calcd for $C_{30}H_{25}N_2O_3S$ [$(M+H)^+$], 493.1580, found 493.1580.

(*E*)-1-Phenyl-5-(phenyl(phenylsulfonyl)methylene)-3-(phenylamino)-1,5-dihydro-*2H*-pyrrol-2-one. (6f)



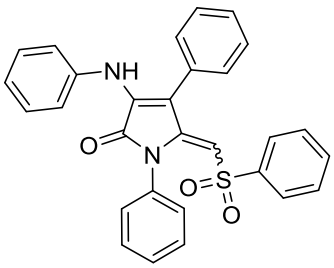
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 13:1, R_f = 0.2; Yellow solid: 155 mg (65%), mp = 218–219 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 9.40 (s, 1H, NH), 7.64–7.61 (m 1H, ArH), 7.59 (d, J = 7.6 Hz, 3H, ArH), 7.53 (t, J = 7.7 Hz, 2H, ArH), 7.49–7.46 (m, 2H, ArH), 7.41 (d, J = 7.8 Hz, 2H, ArH), 7.12 (t, J = 7.2 Hz, 1H, ArH), 6.99–6.96 (m, 3H, ArH+C=CH), 6.95–6.92 (m, 1H, ArH), 6.86–6.79 (m, 4H, ArH), 6.72 (d, J = 7.4 Hz, 2H, ArH); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 167.6, 148.7, 141.7, 140.9, 136.3, 135.5, 133.7, 133.5, 113.5, 131.0, 129.9, 129.9, 129.7, 129.7, 129.5, 129.5, 128.6, 128.6, 128.4, 127.7, 127.4, 127.4, 127.4, 127.4, 123.4, 119.5, 119.3, 119.3, 96.8; **HRMS** (TOF ES+): m/z calcd for $C_{29}H_{23}N_2O_3S$ [$(M+H)^+$], 479.1424, found, 479.1434.

(*E*)-5-((4-Chlorophenyl)(phenylsulfonyl)methylene)-1-phenyl-3-(phenylamino)-1,5-dihydro-*2H*-pyrrol-2-one. (6g)



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 13:1, R_f = 0.2; Yellow solid: 159 mg (62%), mp = 248–250 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 9.45 (s, 1H, NH), 7.66–7.63 (m, 1H, ArH), 7.62–7.60 (m, 2H, ArH), 7.57–7.54 (m, 3H, ArH), 7.49–7.46 (m, 2H, ArH), 7.42 (s, 1H, C=CH), 7.40 (s, 1H, ArH), 7.13–7.11 (m, 1H, ArH), 7.08–7.05 (m 1H, ArH), 7.03–7.01 (m, 2H, ArH), 6.87 (dd, J = 7.8, 3.4 Hz, 4H, ArH), 6.72 (d, J = 8.4 Hz, 2H, ArH); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 167.5, 149.2, 141.5, 140.8, 136.5, 135.5, 135.2, 135.2, 133.8, 133.5, 130.1, 129.9, 129.9, 129.9, 129.9, 129.9, 129.8, 129.7, 128.7, 128.7, 127.8, 127.4, 127.4, 127.4, 123.5, 119.4, 119.4, 117.8, 96.5; **HRMS** (TOF ES+): m/z calcd for $C_{29}H_{22}ClN_2O_3S$ [$(M+H)^+$], 513.1034, found, 513.1039.

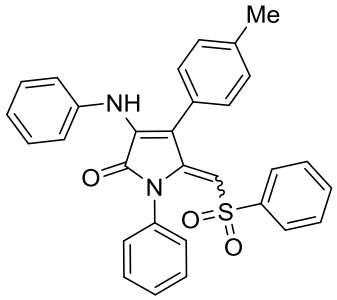
1,4-Diphenyl-3-(phenylamino)-5-((phenylsulfonyl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one. (7a)



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 13:1, R_f = 0.2; Yellow solid: 162 mg (68%), *Z*/*E* = 11/1, mp = 234–235 °C; **Major: ^{1}H NMR** (600 MHz, DMSO-d_6) δ = 9.12 (s, 1H, NH), 7.60–7.58 (m, 1H, ArH),

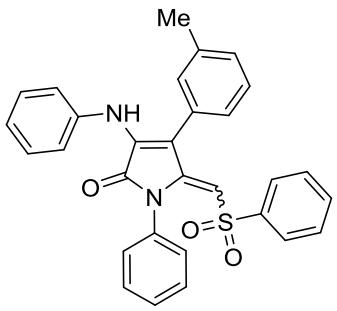
7.44–7.41 (m, 2H, ArH), 7.36–7.32 (m, 1H, ArH), 7.29 (d, *J* = 7.4 Hz, 2H, ArH), 7.25–7.17 (m, 5H, ArH),7.10–7.08 (m, 4H, ArH), 6.87–6.84 (m, 2H, ArH), 6.78–6.75 (m, 1H, ArH), 6.70 (d, *J* = 7.5 Hz, 2H, ArH), 5.68 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 168.2, 151.2 (minor), 150.2, 143.4 (minor), 142.8, 138.6, 138.1 (minor), 137.6 (minor), 136.4, 135.1 (minor), 133.6 (minor), 133.2, 132.8 (minor), 132.6, 131.6 (minor), 131.2, 131.1 (minor), 130.3 (minor), 130.2, 130.2, 129.8 (minor), 129.5, 129.5, 129.3 (minor), 129.2, 129.2, 128.8, 128.8, 128.5, 128.5, 128.5, 128.4, 128.0, 128.0, 127.9 (minor), 127.4 (minor), 126.1, 126.1, 123.8 (minor), 123.2, 121.7, 121.7, 113.0, 104.7; **HRMS** (TOF ES+): m/z calcd for $C_{29}H_{23}N_2O_3S$ [(M+H)$^+$], 479.1424, found, 479.1425.

(*E*)-1-Phenyl-3-(phenylamino)-5-((phenylsulfonyl)methylene)-4-(*p*-tolyl)-1,5-dihydro-*2H*-pyrrol-2-one.(7b)



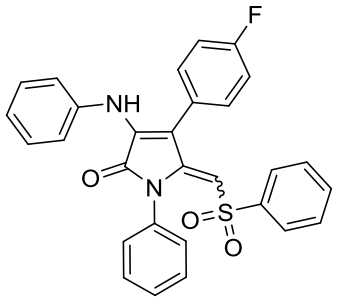
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 13:1, R_f = 0.2; Yellow solid: 133 mg (54%), *Z*/*E* = 9/1, mp = 223–225 ^{o}C; **Major: ^{1}H NMR** (600 MHz, DMSO-d_6) δ = 9.05 (s, 1H, NH), 7.62–7.58 (m, 1H, ArH), 7.44–7.42 (m, 2H, ArH), 7.34–7.32 (m, 1H, ArH), 7.29 (d, *J* = 7.8 Hz, 2H, ArH), 7.23–7.20 (m, 2H, ArH), 7.08 (d, *J* = 7.7 Hz, 2H, ArH), 7.03 (d, *J* = 7.9 Hz, 2H, ArH), 6.99 (d, *J* = 7.9 Hz, 2H, ArH), 6.89–6.86 (m, 2H, ArH), 6.79–6.77 (m, 1H, ArH), 6.71 (d, *J* = 7.8 Hz, 2H, ArH), 5.69 (s, 1H, C=CH), 2.24 (s, 3H, $ArCH_3$).**^{13}C NMR** (150 MHz, DMSO-d_6): δ = 168.2, 165.1 (minor), 151.2 (minor), 150.2, 143.3 (minor), 142.8, 138.8, 138.8 (minor), 137.8, 137.6 (minor), 136.4, 133.6 (minor), 133.2, 132.4, 130.9 (minor), 130.3 (minor), 130.0, 130.0, 129.8 (minor), 129.5, 129.5, 129.2, 129.2, 129.2, 129.2, 128.8, 128.8, 128.5, 128.2, 128.0, 128.0, 127.8 (minor), 126.1, 126.1, 126.0 (minor), 123.5 (minor), 123.0, 122.9 (minor), 109.6 (minor), 107.6 (minor), 121.4, 121.4, 113.8, 104.9, 21.3; **HRMS** (TOF ES+): m/z calcd for $C_{30}H_{25}N_2O_3S$ [(M+H)$^+$], 493.1580, found, 493.1579.

(*Z*)-1-Phenyl-3-(phenylamino)-5-((phenylsulfonyl)methylene)-4-(*m*-tolyl)-1,5-dihydro-*2H*-pyrrol-2-one. (7c)



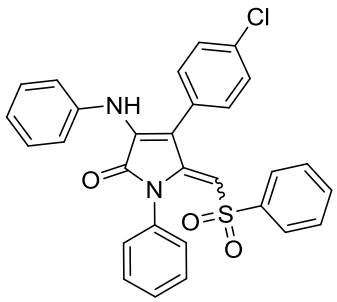
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 10:1, R_f = 0.2; Yellow solid: 128 mg (52%), *Z*/*E* = 6/1, mp = 209–211 °C; **Major: ^{1}H NMR** (600 MHz, DMSO-d_6) δ = 9.11 (s, 1H, NH), 7.59–7.58 (m, 1H, ArH), 7.43 (t, *J* = 7.8 Hz, 2H, ArH), 7.33–7.29 (m, 3H, ArH), 7.23–7.21 (m, 2H, ArH), 7.13 (t, *J* = 7.6 Hz, 1H, ArH), 7.09 (d, *J* = 7.5 Hz, 2H, ArH), 7.02 (d, *J* = 7.5 Hz, 1H, ArH), 6.95 (d, *J* = 7.5 Hz, 1H, ArH), 6.88–6.86 (m, 2H, ArH), 6.79–6.77 (m, 2H, ArH), 6.69 (d, *J* = 7.7 Hz, 2H, ArH), 5.71 (s, 1H, C=CH), 2.08 (s, 3H, $ArCH_3$); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 168.1, 150.1, 142.9, 138.7, 137.7, 136.4, 133.2, 132.6, 131.2, 130.9, 130.3 (minor), 129.8 (minor), 129.5, 129.5, 129.2, 129.2, 128.9, 128.8, 128.8, 128.5, 128.4, 127.9, 127.9, 127.7 (minor), 127.2 (minor), 127.1, 126.1, 126.1, 125.9 (minor), 123.3, 123.0 (minor), 121.8, 121.8, 113.0, 104.8, 21.2; **HRMS** (TOF ES+): m/z calcd for $C_{30}H_{25}N_2O_3S$ [$(M+H)^+$], 493.1580, found, 493.1579.

(*Z*)-4-(4-Fluorophenyl)-1-phenyl-3-(phenylamino)-5-((phenylsulfonyl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one. (7d)



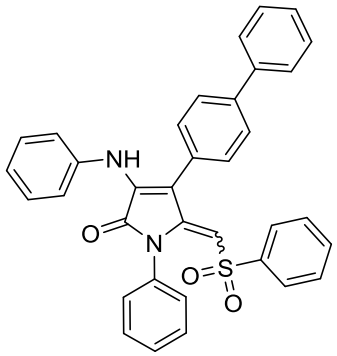
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 9:1, R_f = 0.2; Yellow solid: 82 mg (33%), *Z*/*E* = 3/1, mp = 221–223 °C; **Major: ^{1}H NMR** (600 MHz, DMSO-d_6): δ = 9.16 (s, 1H, NH), 7.63–7.56 (m, 2H, ArH), 7.44–7.41 (m, 2H, ArH), 7.30–7.26 (m, 2H, ArH), 7.22–7.20 (m, 2H, ArH), 7.14–7.10 (m, 2H, ArH), 7.08 (d, *J* = 7.4 Hz, 2H, ArH), 7.04–7.01 (m, 2H, ArH), 6.92–6.89 (m, 2H, ArH), 6.71 (d, *J* = 7.5 Hz, 3H, ArH), 5.64 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 168.1, 165.0, 162.1 (d, *J* = 245.9 Hz), 150.2, 143.5 (minor), 142.9, 138.5, 138.0 (minor), 136.4, 133.5 (minor), 133.2 (minor), 133.2, 133.1, 133.0 (minor), 132.4 (d, *J* = 8.5 Hz), 132.4 (d, *J* = 8.5 Hz), 130.3 (minor), 129.8, (minor), 129.5, 129.5, 129.3 (minor), 129.2, 129.2, 128.8, 128.8, 128.5 (minor), 128.0, 128.0, 127.9 (minor), 127.6 (d, *J* = 2.7 Hz), 126.1, 126.1, 125.8 (minor), 124.1 (minor), 123.6, 123.4, 122.1, 122.1, 115.5 (d, *J* = 21.5 Hz), 114.1 (minor), 111.8, 107.0 (minor), 104.5; **HRMS** (TOF ES+): m/z calcd for $C_{29}H_{22}N_2FO_3S$ [$(M+H)^+$], 497.1330, found, 497.1342.

(*Z*)-4-(4-Chlorophenyl)-1-phenyl-3-(phenylamino)-5-((phenylsulfonyl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one. (7e)



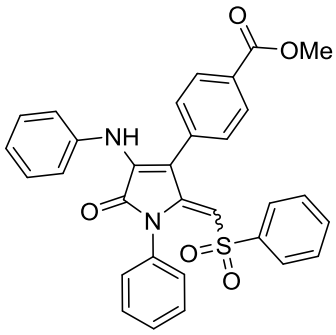
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 13:1, R_f = 0.2; Yellow solid: 76 mg (30%), *Z*/*E* = 3/1, mp = 210–212 °C; **Major: ^{1}H NMR** (600 MHz, DMSO-d_6): δ = 9.20 (s, 1H, NH), 7.61–7.58 (m, 1H, ArH), 7.44–7.41 (m, 2H ArH), 7.29 (d, *J* = 7.8 Hz, 2H, ArH), 7.25 (d, *J* = 8.4 Hz, 2H, ArH), 7.22 (d, *J* = 7.7 Hz, 2H, ArH), 7.07 (d, *J* = 7.6 Hz, 4H, ArH), 6.93–6.90 (m, 2H, ArH), 6.84–6.82 (m, 2H, ArH), 6.75–6.70 (m, 2H, ArH), 5.65 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 167.9, 164.9 (minor), 151.1 (minor), 149.9, 143.3 (minor), 142.8, 138.5, 138.2 (minor), 138.0 (minor), 136.4, 133.5 (minor), 133.2, 133.2, 133.1, 132.9 (minor), 132.4 (minor), 132.0, 132.0, 130.3 (minor), 130.1 (minor), 129.8 (minor), 129.4, 129.4, 129.3, 129.2, 129.2, 128.9, 128.9, 128.5, 128.5, 128.1, 128.1, 128.0 (minor), 127.7, 127.2 (minor), 126.2, 126.2, 125.9, 124.2 (minor), 123.7 (minor), 123.5 (minor), 122.2, 122.2, 111.5, 106.9 (minor), 104.2; **HRMS** (TOF ES+): m/z calcd for $C_{29}H_{22}ClN_2O_3S$ [$(M+H)^+$], 513.1034, found, 513.1043.

(*Z*)-4-([1,1'-Biphenyl]-4-yl)-1-phenyl-3-(phenylamino)-5-((phenylsulfonyl)methylene)-1,5-dihydro-*2H*-pyrrol-2-one. (7f)



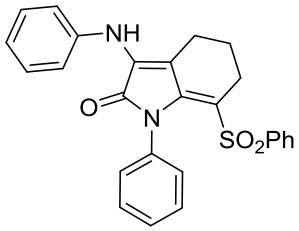
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 9:1, R_f = 0.2; Yellow solid: 130 mg (47%), *Z*/*E* = 5/1, mp = 224–226 °C; **Major: ^{1}H NMR** (600 MHz, DMSO-d_6): δ = 9.17 (s, 1H, NH), 7.62–7.61 (m, 3H, ArH), 7.50–7.46 (m, 4H, ArH), 7.43–7.42 (m, 2H, ArH), 7.39–7.37 (m, 1H, ArH), 7.30 (d, *J* = 7.6 Hz, 2H, ArH), 7.23–7.21 (m, 2H, ArH), 7.17 (d, *J* = 7.8 Hz, 2H, ArH), 7.10 (d, *J* = 7.7 Hz, 2H, ArH), 6.88–6.86 (m, 2H, ArH), 6.74 (d, *J* = 8.1 Hz, 2H, ArH), 5.78 (s, 1H, C=CH); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 168.1, 165.0 (minor), 151.2 (minor), 150.1, 143.3 (minor), 142.8, 140.0, 139.9, 138.7, 138.2 (minor), 137.9 (minor), 136.4, 133.6 (minor), 133.2, 132.9, 132.8 (minor), 131.6 (minor), 130.7, 130.7, 130.3 (minor), 130.3, 129.8 (minor), 129.5, 129.5, 129.4 (minor), 129.5, 129.5, 129.2, 129.4, 128.9, 128.9, 128.5, 128.1 (minor), 128.0, 128.0, 127.8 (minor), 127.1, 127.1, 127.0, 126.7, 126.7. 126.1, 126.1, 125.9 (minor), 125.6 (minor), 123.8 (minor), 123.4 (minor), 123.2, 121.9, 121.9, 112.8, 108.1 (minor), 104.7; **HRMS** (TOF ES+): m/z calcd for $C_{35}H_{27}N_2O_3S$ [$(M+H)^+$], 555.1737, found, 555.1737.

Methyl(*Z*)-4-(5-oxo-1-phenyl-4-(phenylamino)-2-((phenylsulfonyl)methylene)-2,5-dihydro-*1H*-pyrrol-3-yl)benzoate. (7g)



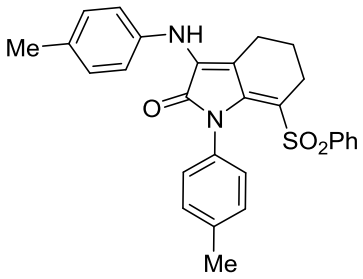
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 8:1, R_f = 0.2; Yellow solid: 93 mg (37%), *Z*/*E* = 3/1, mp = 230–232 °C; **Major: ^{1}H NMR** (600 MHz, DMSO-d_6) δ = 9.30 (s, 1H, NH), 7.74 (d, *J* = 7.0 Hz, 2H, ArH), 7.60–7.58 (m, 2H, ArH), 7.44–7.41 (m, 2H, ArH), 7.30–7.29 (m, 3H, ArH), 7.22 (d, *J* = 6.2 Hz, 4H, ArH), 7.09 (d, *J* = 6.8 Hz, 2H, ArH), 6.87–6.85 (m, 2H, ArH), 6.72 (d, *J* = 7.2 Hz, 2H, ArH), 5.67 (s, 1H, C=CH), 3.83 (s, 3H, OCH_3); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 167.9, 166.3, 149.7, 142.8, 142.2 (minor), 138.5, 138.1 (minor), 137.9 (minor), 137.2 (minor), 136.4, 136.4, 133.5, 133.2, 133.1 (minor), 131.3, 130.6, 130.6, 130.3 (minor), 129.7 (minor), 129.5, 129.5, 129.4 (minor), 129.2, 129.2, 129.2, 129.2, 128.9, 128.9, 128.9, 128.9, 128.5, 128.1, 128.1, 127.9 (minor), 126.2, 126.2, 126.0 (minor), 123.7 (minor), 123.6 (minor), 122.2, 111.1, 104.5, 52.7; **HRMS** (TOF ES+): m/z calcd for $C_{31}H_{25}N_2O_5S$ [$(M+H)^+$], 537.1479, found, 537.1480.

1-Phenyl-3-(phenylamino)-7-(phenylsulfonyl)-1,4,5,6-tetrahydro-*2H*-indol-2-one. (8a)



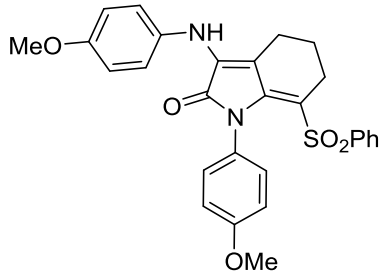
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 8:1, R_f = 0.2; Yellow solid: 106 mg (48%), mp = 199–201 °C; **^{1}H NMR** (600 MHz, DMSO-d_6) δ = 8.60 (s, 1H, NH), 7.63–7.61 (t, *J* = 7.4 Hz, 1H, ArH), 7.47–7.44 (m 2H, ArH), 7.30–7.26 (m, 5H, ArH), 7.23–7.21 (m, 2H, ArH), 7.03 (dd, *J* = 12.8, 7.7 Hz, 4H, ArH), 6.98–6.96 (t, *J* = 7.4 Hz, 1H, ArH), 2.51 (d, *J* = 2.0 Hz, 2H, CH_2), 2.18–2.16 (t, *J* = 6.2 Hz, 2H, CH_2), 1.81–1.62 (m, 2H, CH_2); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 169.2, 145.5, 141.3, 141.1, 138.7, 133.5, 129.6, 129.6, 129.2, 129.1, 129.1, 128.6, 128.6, 128.5, 128.5, 127.5, 126.8, 126.8, 122.5, 120.6, 120.6, 117.8, 115.4, 28.1, 24.8, 22.5; **HRMS** (TOF ES^+): m/z calcd for $C_{26}H_{23}N_2O_3S$ [$(M+H)^+$], 443.1424, found, 443.1429.

7-(Phenylsulfonyl)-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,4,5,6-tetrahydro-*2H*-indol-2-one.(8b)



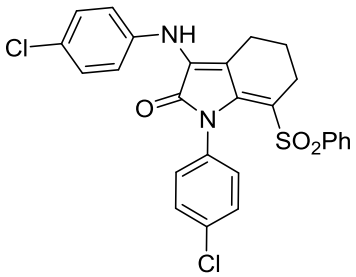
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 8:1, R_f = 0.2; Yellow solid: 102 mg (46%), mp = 206–208 °C; **^{1}H NMR** (600 MHz, DMSO-d_6) δ = 8.49 (s, 1H, NH), 7.61–7.58 (m, 1H, ArH), 7.44–7.41 (m, 2H, ArH), 7.22 (d, *J* = 7.6 Hz, 2H, ArH), 7.09 (d, *J* = 8.1 Hz, 2H, ArH), 6.95–6.93 (m, 4H, ArH), 6.86 (d, *J* = 8.1 Hz, 2H, ArH), 2.55 (t, *J* = 5.9 Hz, 2H, CH_2), 2.31 (s, 3H, CH_3), 2.25 (s, 3H, CH_3), 2.12 (t, *J* = 6.2 Hz, 2H, CH_2), 1.76–1.65 (m, 2H, CH_2); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 169.5, 146.4, 141.8, 138.2, 136.8, 135.8, 133.2, 131.8, 129.6, 129.5, 129.5, 129.4, 129.4, 129.0, 129.0, 128.4, 128.4, 126.5, 126.5, 121.3, 121.3, 115.7, 114.7, 28.2, 24.7, 22.5, 21.2, 20.8; **HRMS** (TOF ES$^+$): m/z calcd for $C_{28}H_{27}N_2O_3S$ [(M+H)$^+$], 471.1737, found, 417.1737.

1-(4-Methoxyphenyl)-3-((4-methoxyphenyl)amino)-7-(phenylsulfonyl)-1,4,5,6-tetrahydro-*2H*-indol-2-one.(8c)



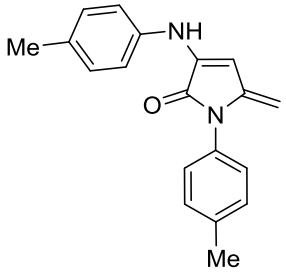
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 8:1, R_f = 0.2; Yellow solid: 80 mg (32%), mp = 195–197 °C; **^{1}H NMR** (600 MHz, DMSO-d_6): δ = 8.39 (s, 1H, NH), 7.60–7.57 (m, 1H, ArH), 7.42–7.40 (m, 2H, ArH), 7.27–7.19 (m, 2H, ArH), 7.05–6.98 (m, 2H, ArH), 6.90–6.81 (m, 4H, ArH), 6.66 (d, *J* = 8.9 Hz, 1H, ArH), 3.75 (s, 3H, ArOMe), 3.73 (s, 3H, ArOMe), 2.56 (t, *J* = 5.8 Hz, 2H, CH_2), 2.07 (t, *J* = 6.3 Hz, 2H, CH_2), 1.75–1.62 (m, 2H, CH_2); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 169.6, 158.6, 155.8, 146.9, 142.2, 133.6, 133.2, 130.9, 130.3, 130.0, 130.0, 129.4, 129.4, 126.1, 126.1, 123.8, 123.8, 114.2, 114.2, 113.8, 113.6, 113.6. 113.2, 55.7, 55.6, 28.4, 24.4, 22.7; **HRMS** (TOF ES$^+$): m/z calcd for $C_{28}H_{27}N_2O_5S$ [(M+H)$^+$], 503.1635, found, 503.1636.

1-(4-Chlorophenyl)-3-((4-chlorophenyl)amino)-7-(phenylsulfonyl)-1,4,5,6-tetrahydro-*2H*-indol-2-one.(8d)



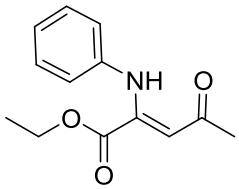
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 8:1, R_f = 0.2; Yellow solid: 76 mg (30%), mp = 227–229 °C; **^{1}H NMR** (600 MHz, DMSO-d_6) δ = 8.75 (s, 1H, NH), 7.67–7.65 (m, 1H, ArH), 7.51–7.49 (m, 2H, ArH), 7.39 (d, *J* = 7.9 Hz, 2H, ArH), 7.31 (d, *J* = 8.6 Hz, 2H, ArH), 7.24 (d, *J* = 8.5 Hz, 2H, ArH), 7.08 (d, *J* = 8.5 Hz, 2H, ArH), 7.00 (d, *J* = 8.6 Hz, 2H, ArH), 2.48 (d, *J* = 6.4 Hz, 2H, CH_2), 2.18 (t, *J* = 6.2 Hz, 2H, CH_2), 1.95 – 1.67 (m, 2H, CH_2); **^{13}C NMR** (150 MHz, DMSO-d_6): δ = 168.8, 144.9, 141.4, 140.2, 137.3, 133.6, 132.1, 130.3, 130.3, 129.6, 129.6, 128.9, 128.9, 128.8, 128.5, 128.5, 126.6, 126.6, 125.8, 121.7, 121.7, 119.5, 116.0, 28.2, 24.7, 22.4; **HRMS** (TOF ES$^+$): m/z calcd for $C_{26}H_{21}Cl_2N_2O_3S$ [(M+H)$^+$], 511.0644, found, 511.0649.

5-Methylene-1-(*p*-tolyl)-3-(*p*-tolylamino)-1,5-dihydro-*2H*-pyrrol-2-one.(10)



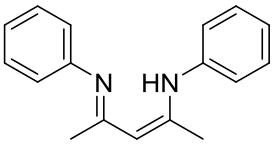
$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 25:1, R_f = 0.2; Gray-yellow solid: 77 mg (53%), mp = 153–155 °C; **^{1}H NMR** (600 MHz, DMSO-d_6) δ = 8.52 (s, 1H, NH), 7.33 (d, J = 8.1 Hz, 2H, ArH), 7.26 (d, J = 8.4 Hz, 2H, ArH), 7.20 (d, J = 8.2 Hz, 2H, ArH), 7.13 (d, J = 8.3 Hz, 2H, ArH), 6.48 (s, 1H, 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.

Ethyl (*Z*)-2-oxo-4-(phenylamino)pent-3-enoate.(12)



$V_{Petroleum\ ether}/V_{Ethyl\ acetate}$ = 25:1, R_f = 0.2; Yellow oli: 89 mg (76%); **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 11.44 (s, 1H, NH), 7.29 (dd, J = 15.1, 7.0 Hz, 2H, ArH), 7.14 (t, J = 7.4 Hz, 1H, ArH), 6.97 (d, J = 7.9 Hz, 2H, ArH), 5.69 (s, 1H, CH=CH), 4.14 (q, J = 7.1 Hz, 2H, CH_2), 2.22 (s, 3H, Me), 1.07 (s, 1H, Me); **^{13}C NMR** (150 MHz, $CDCl_3$): δ = 199.3, 164.5, 148.3, 139.6, 129.1, 129.1, 128.8, 125.0, 121.8, 100.0, 62.1, 30.2, 13.6; **HRMS** (TOF ES$^+$): m/z calcd for $C_{13}H_{17}NO_3$ [(M+H)$^+$], 234.1125, found, 234.1133.

***N*-((2*Z*,4*E*)-4-(Phenylimino)pent-2-en-2-yl)aniline.(13)**



Light yellow solid: 1.38 g (86%); **^{1}H NMR** (600 MHz, $CDCl_3$): δ = 12.71 (s, 1H, NH), 7.33–7.22 (m, 4H, ArH), 7.05 (t, J = 7.3 Hz, 2H, ArH), 6.96 (d, J = 8.0 Hz, 4H, ArH), 4.88 (s, 1H, CH=CH), 2.00 (s, 6H, $2CH_3$); **^{13}C NMR** (600 MHz, $CDCl_3$): δ = 158.3, 158.3, 144.5, 144.5, 127.6, 127.6, 127.6, 127.6, 122.0, 122.0, 121.4, 121.4, 121.4, 121.4, 96.1, 19.7, 19.7; **HRMS** (TOF ES$^+$): m/z calcd for $C_{17}H_{19}N_2$ [(M+H)$^+$], 251.1543, found, 251.1536.

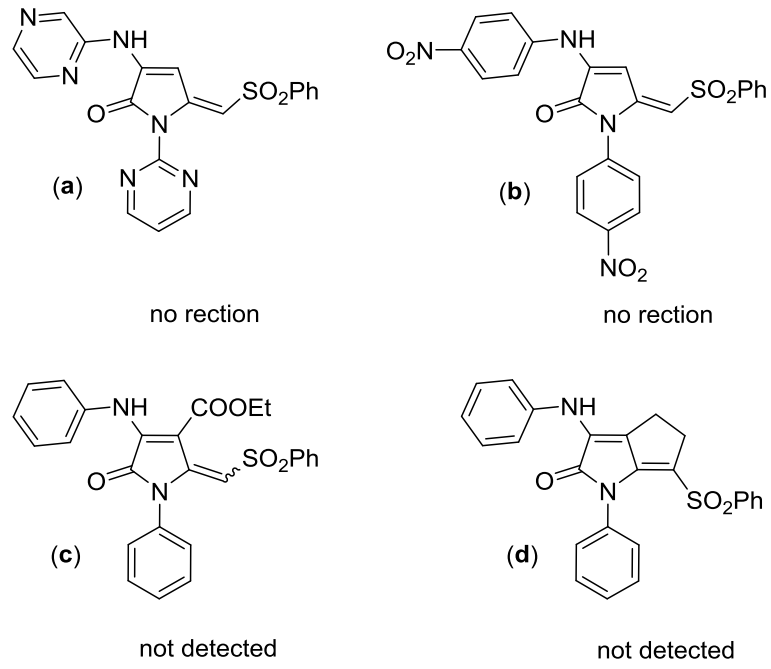


Figure S4. Unsuccessful examples.

6. X-ray Structure and Data[3] of 4a (CCDC 2145010).

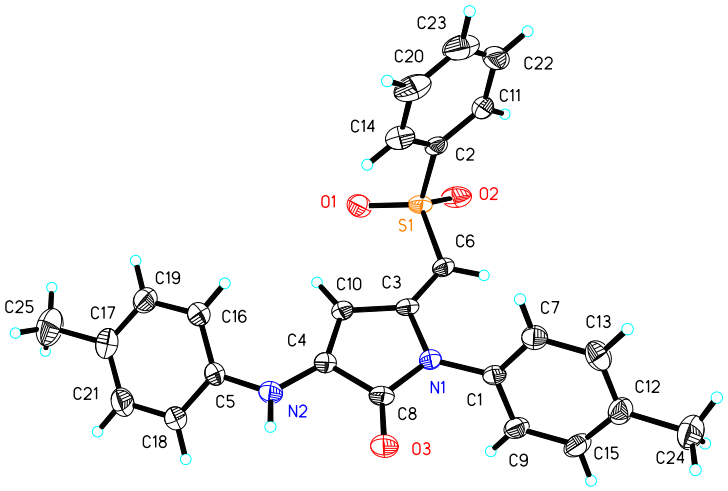


Figure S5 X-Ray crystal structure of 4a.

Table S2 Crystal data and structure refinement for **4a.**

Empirical formula	$C_{25}H_{22}N_2O_3S$
Formula weight	430.50
Temperature	296.15 K
Wavelength	0.71073 Å
Crystal system, space group	monoclinic, P 1 21/c 1
Unit cell dimensions	a = 8.1561(9) A alpha = 90 deg.
	b = 20.032(2) A beta = 95.626(2) deg.
	c = 13.5195(15) A gamma = 90 deg.
Volume	2198.2(4) A^3
Z, Calculated density	4, 1.301 Mg/m^3
Absorption coefficient	0.176 mm^-1
F(000)	904
Theta range for data collection	2.51 to 27.27 deg.
Limiting indices	-10<=h<=10, -25<=k<=26, -16<=l<=17
Completeness to theta = 25.242	99.8%
Absorption correction	Multi-scan
Refinement method	SHELXL-2018/3 (Sheldrick 2015)
Data/restraints/parameters	4873 / 0 / 282
Goodness-of-fit on F^2	1.025
Final R indices [I>2sigma(I)]	R1 = 0.1292, wR2 = 0.1252
R indices (all data)	R1 = 0.0731, wR2 = 0.1406
Largest diff. peak and hole	0.453 and -0.281 e.A^-3

7. ^{1}H NMR, ^{13}C NMR and HRMS spectra for spectroscopic data.

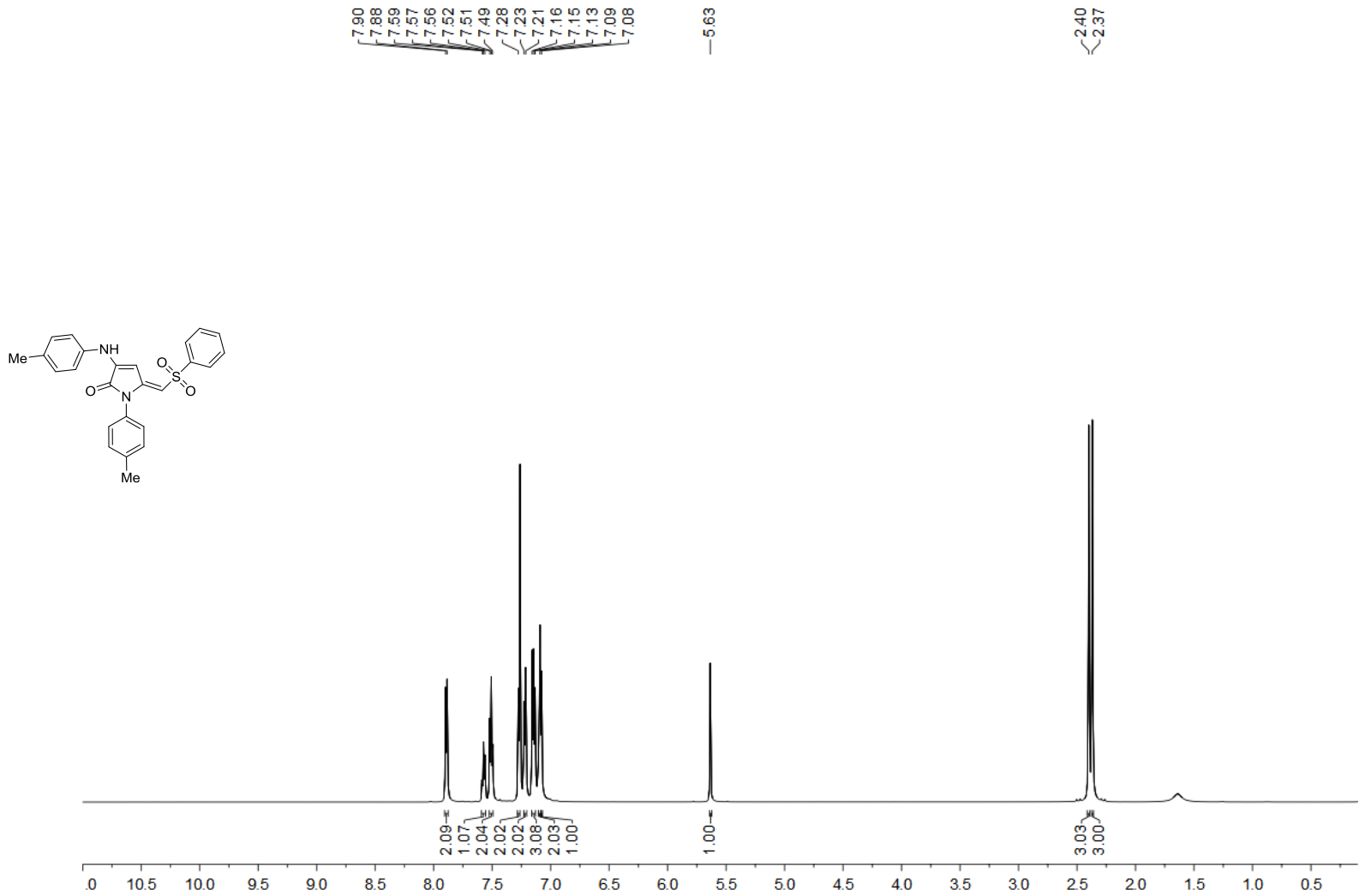


Figure S6. **^{1}H NMR** (600 MHz, $CDCl_3$) spectra of compound **4a**

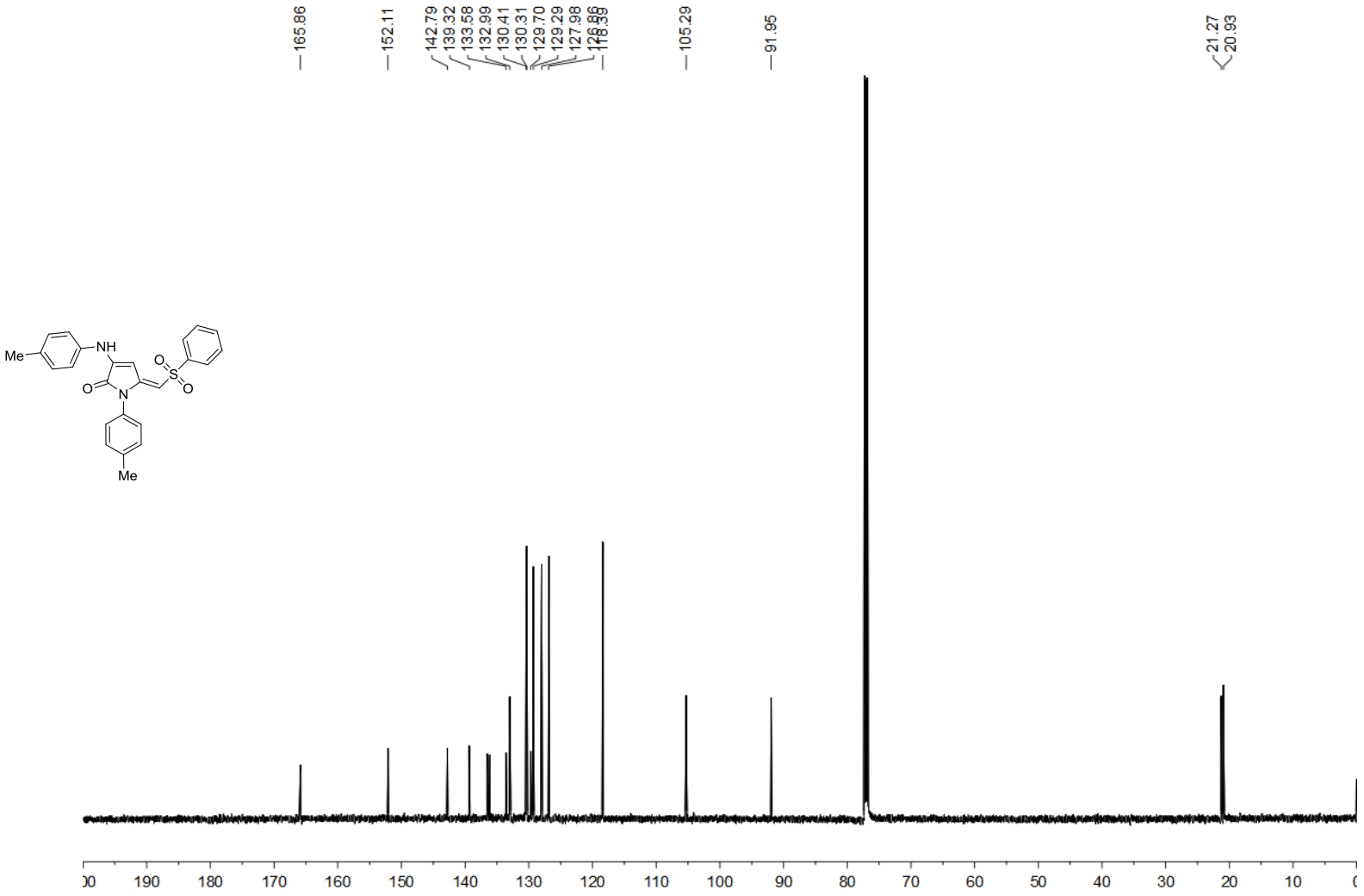


Figure S7. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4a**

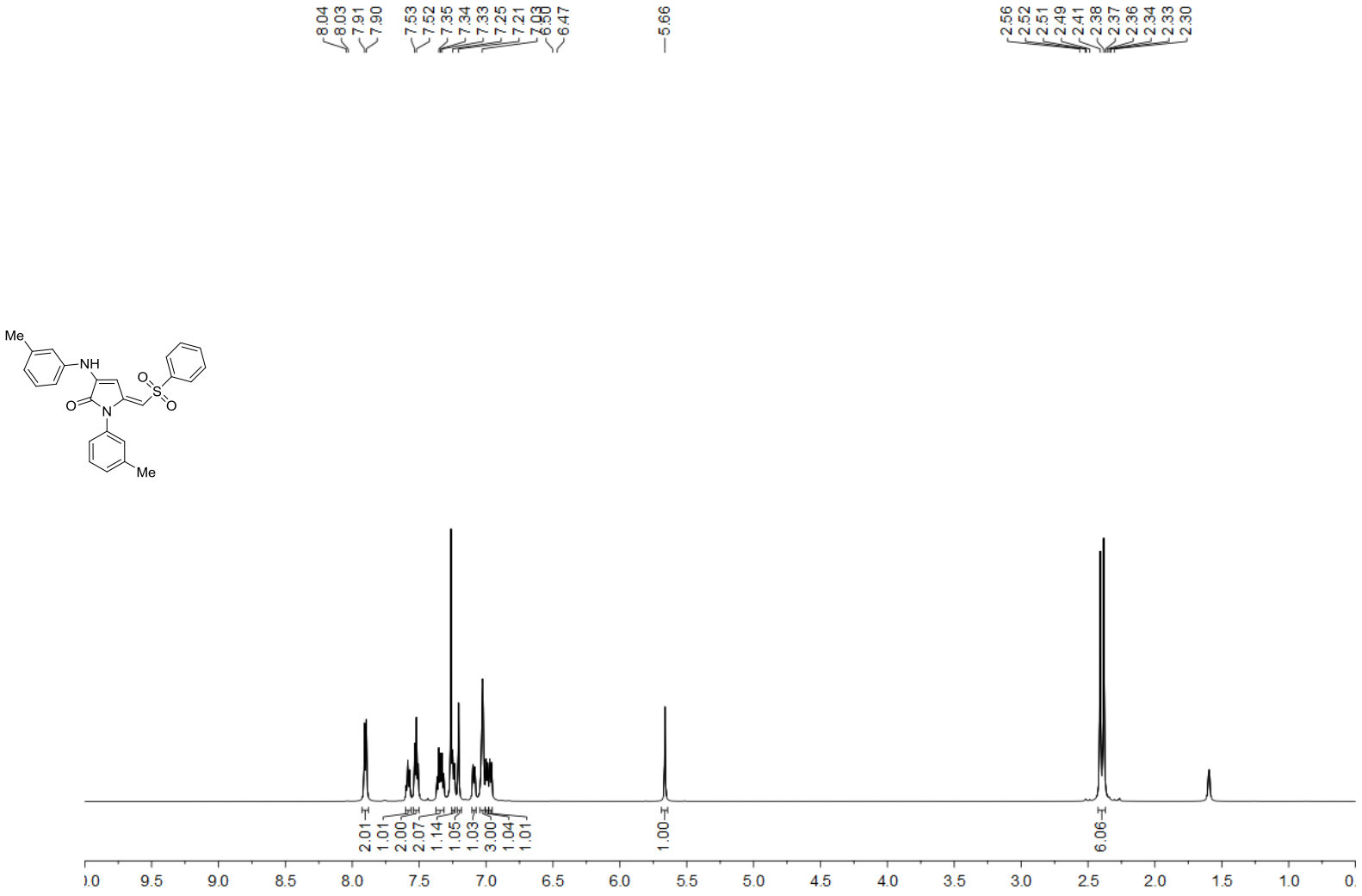


Figure S8. **^{1}H NMR** (600 MHz, $CDCl_3$) spectra of compound **4b**

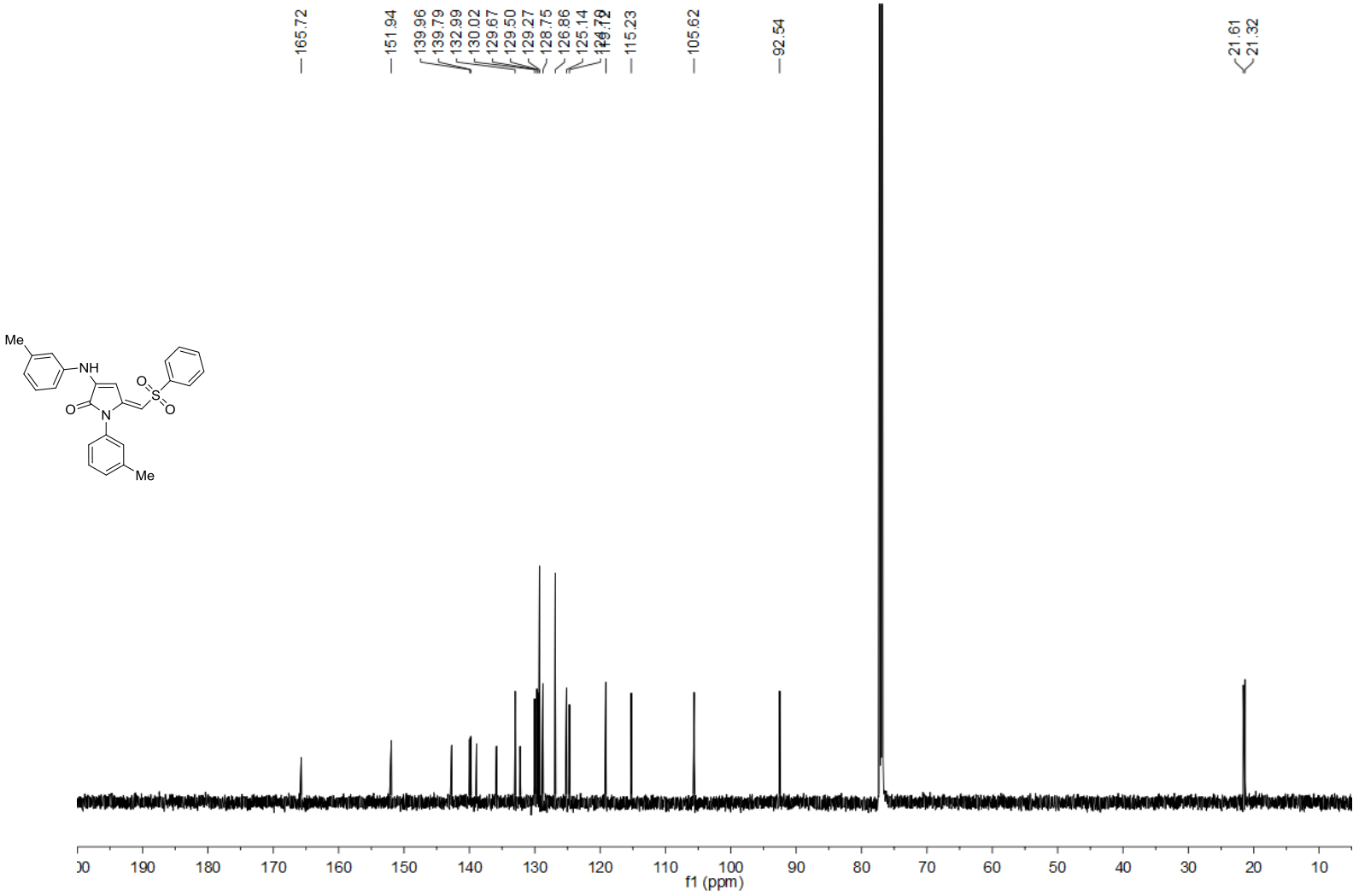


Figure S9. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4b**

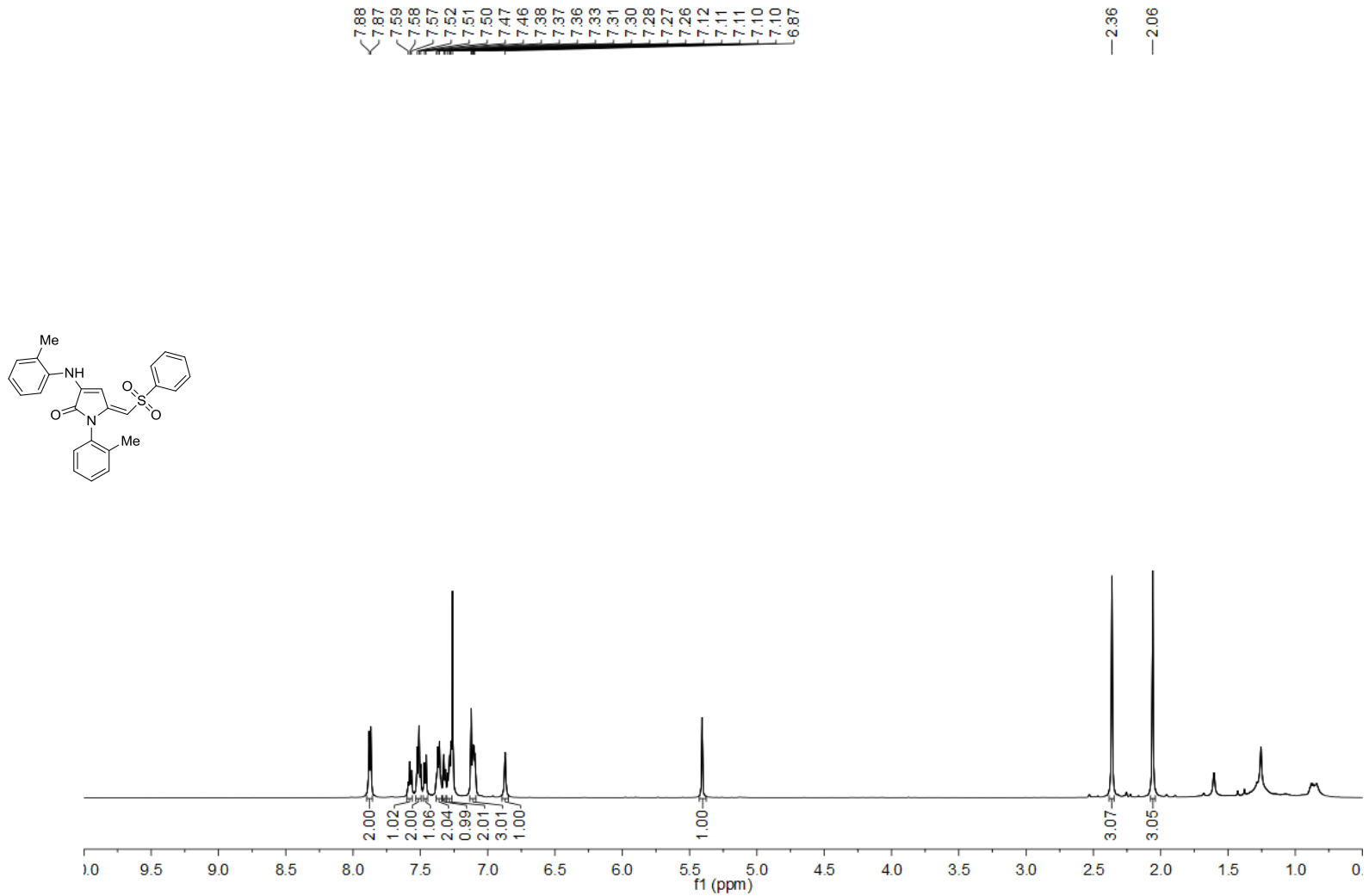


Figure S10. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4c**

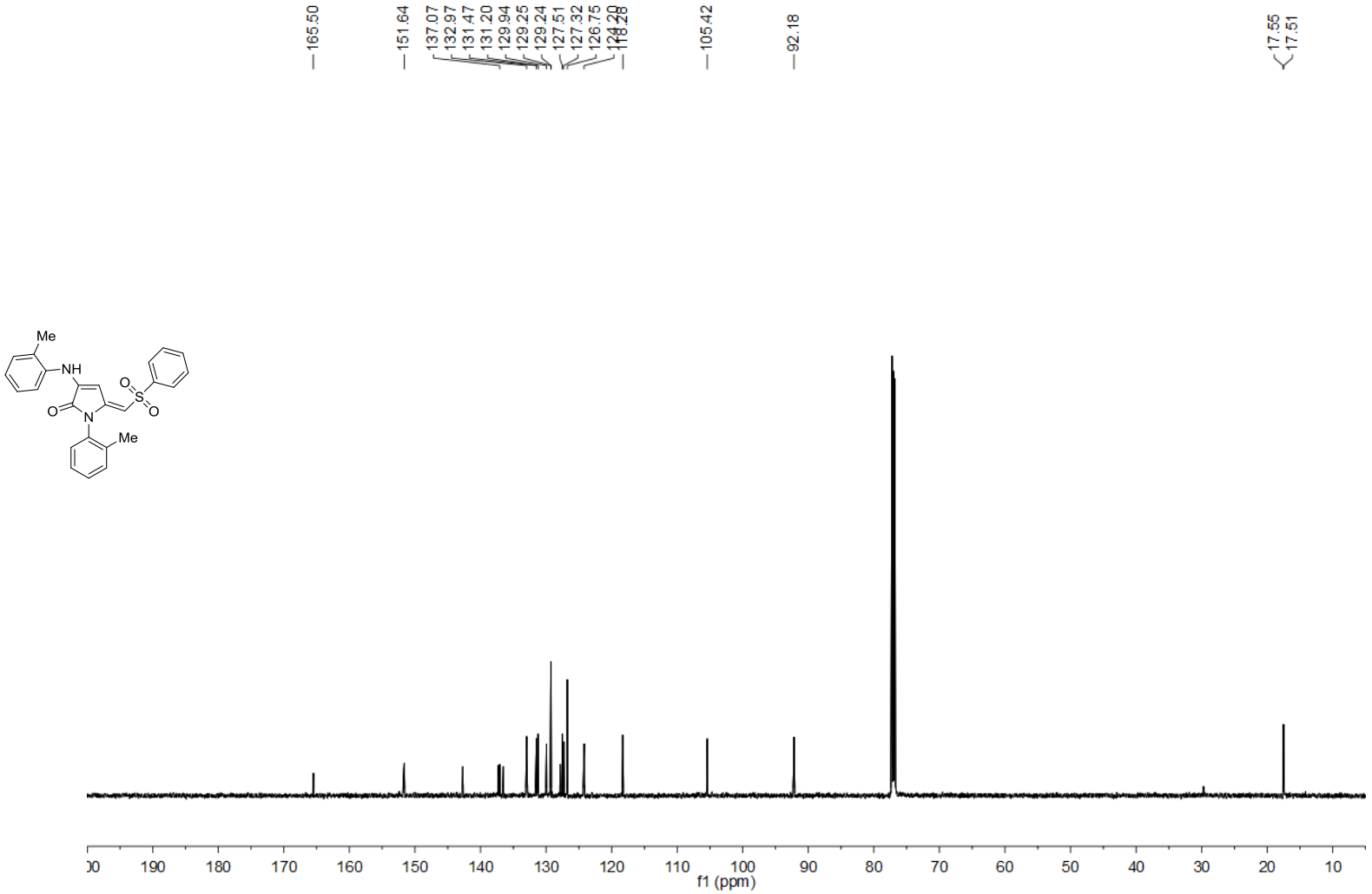


Figure S11. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4c**

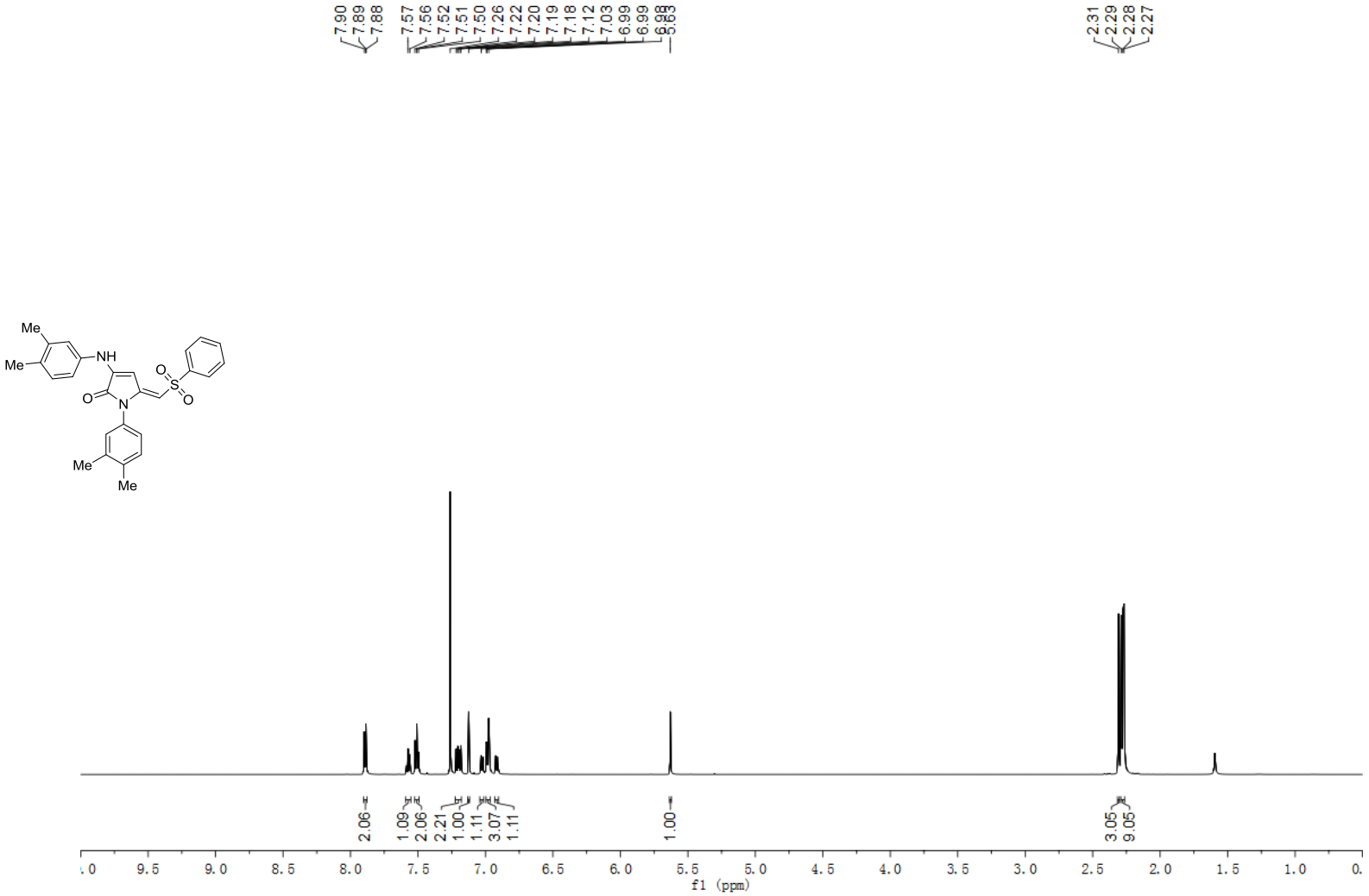


Figure S12. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4d**

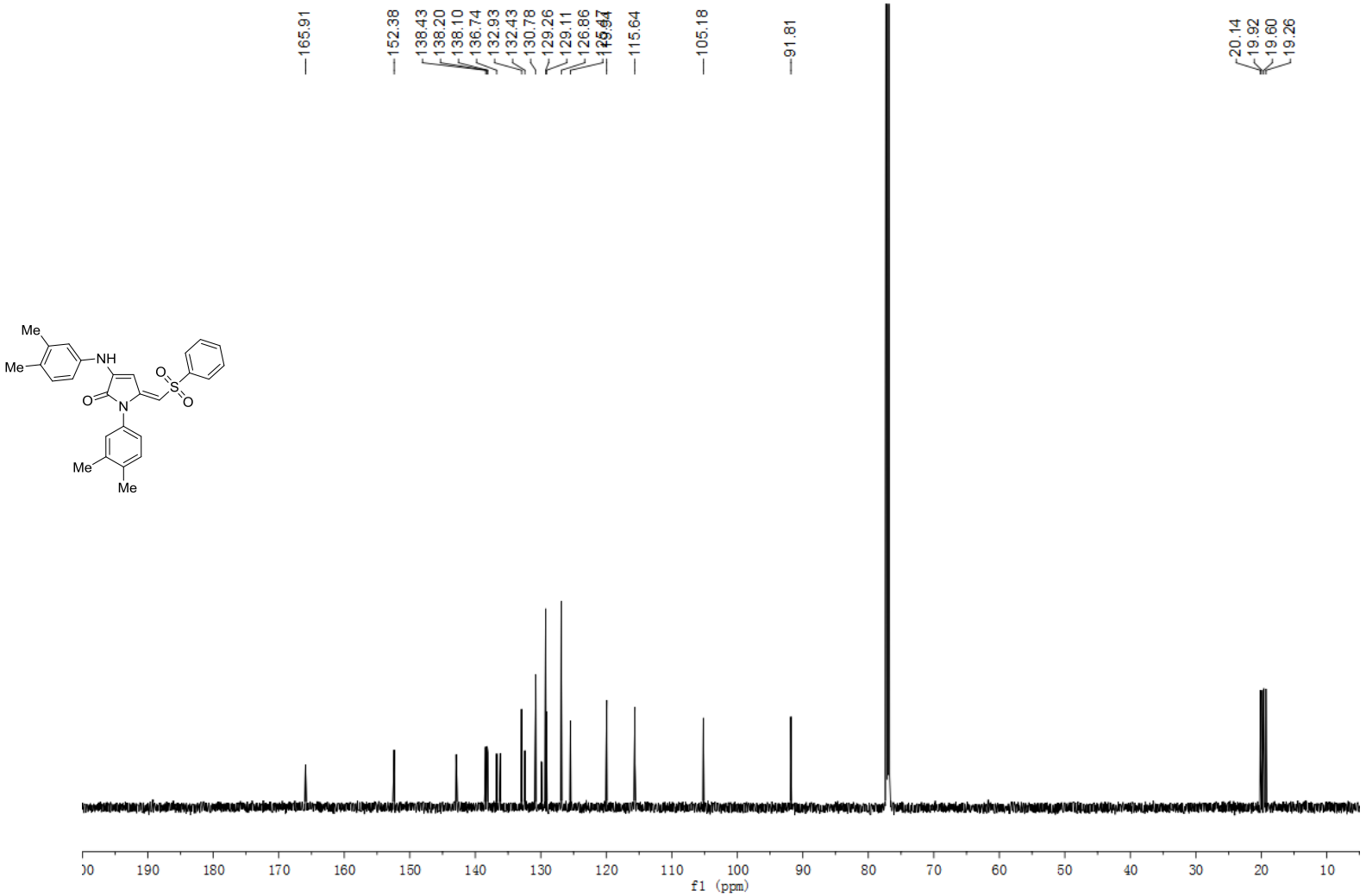


Figure S13. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4d**

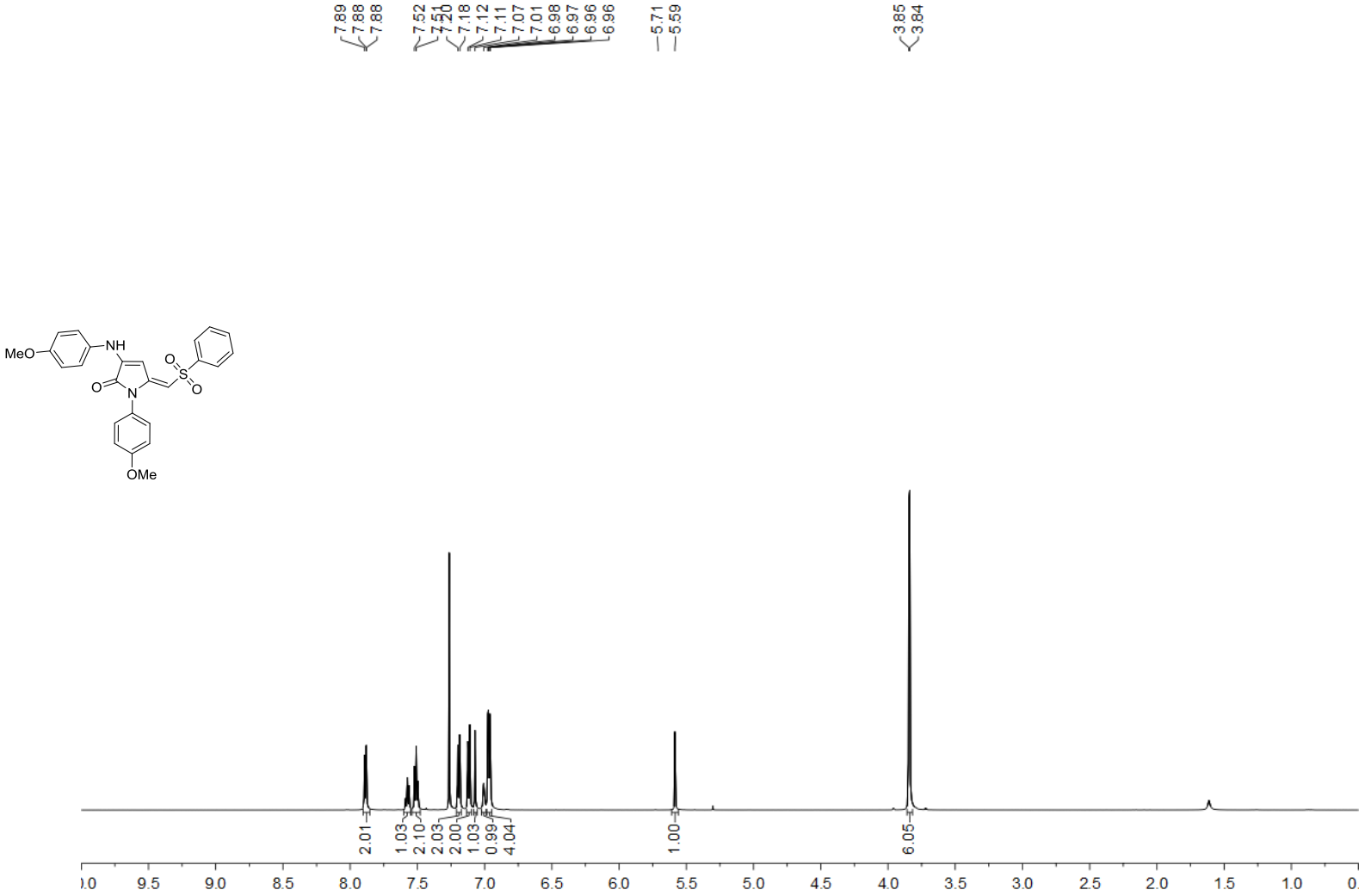


Figure S14. **^{1}H NMR** (600 MHz, $CDCl_3$) spectra of compound **4e**

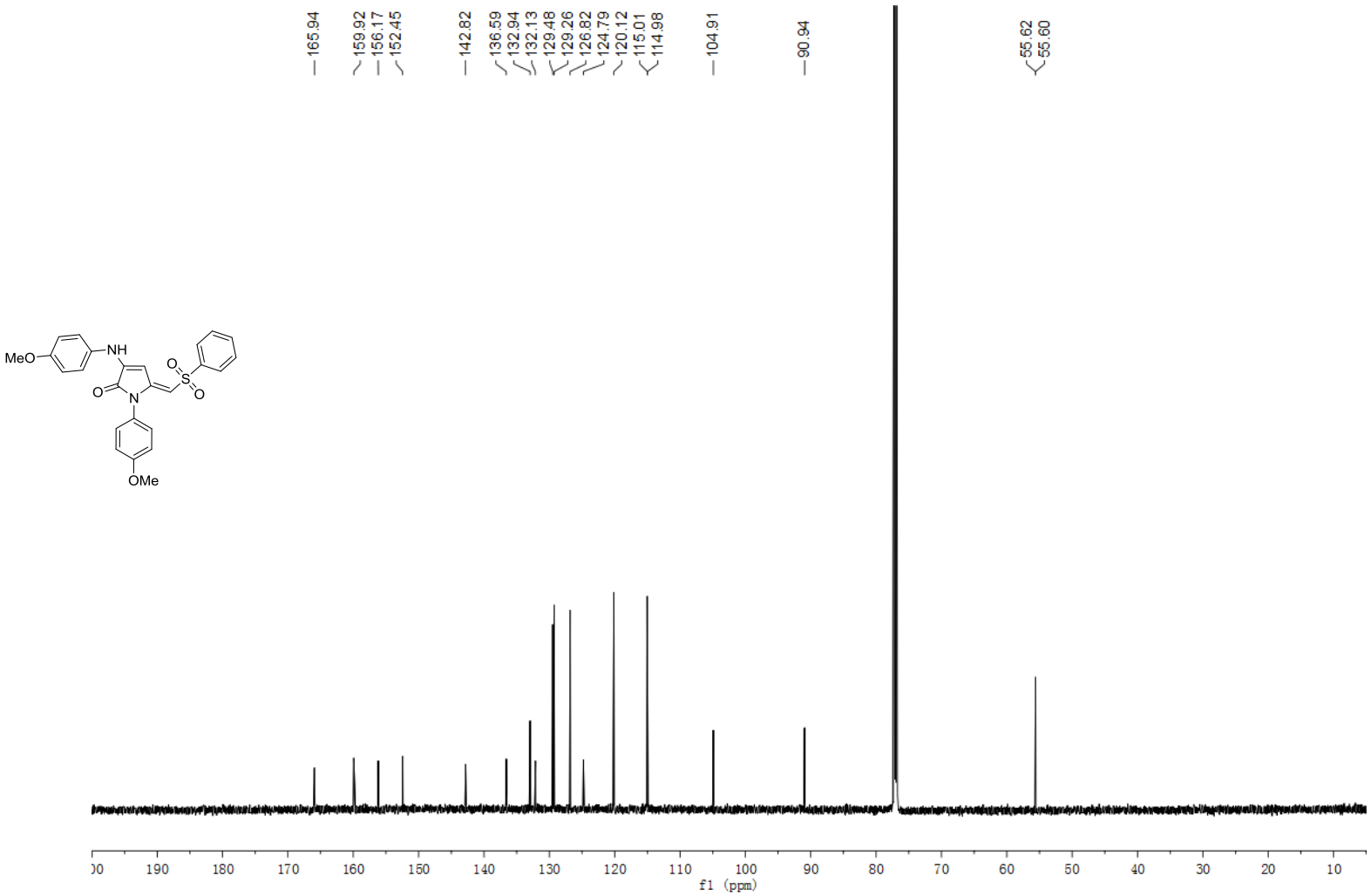


Figure S15. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4e**

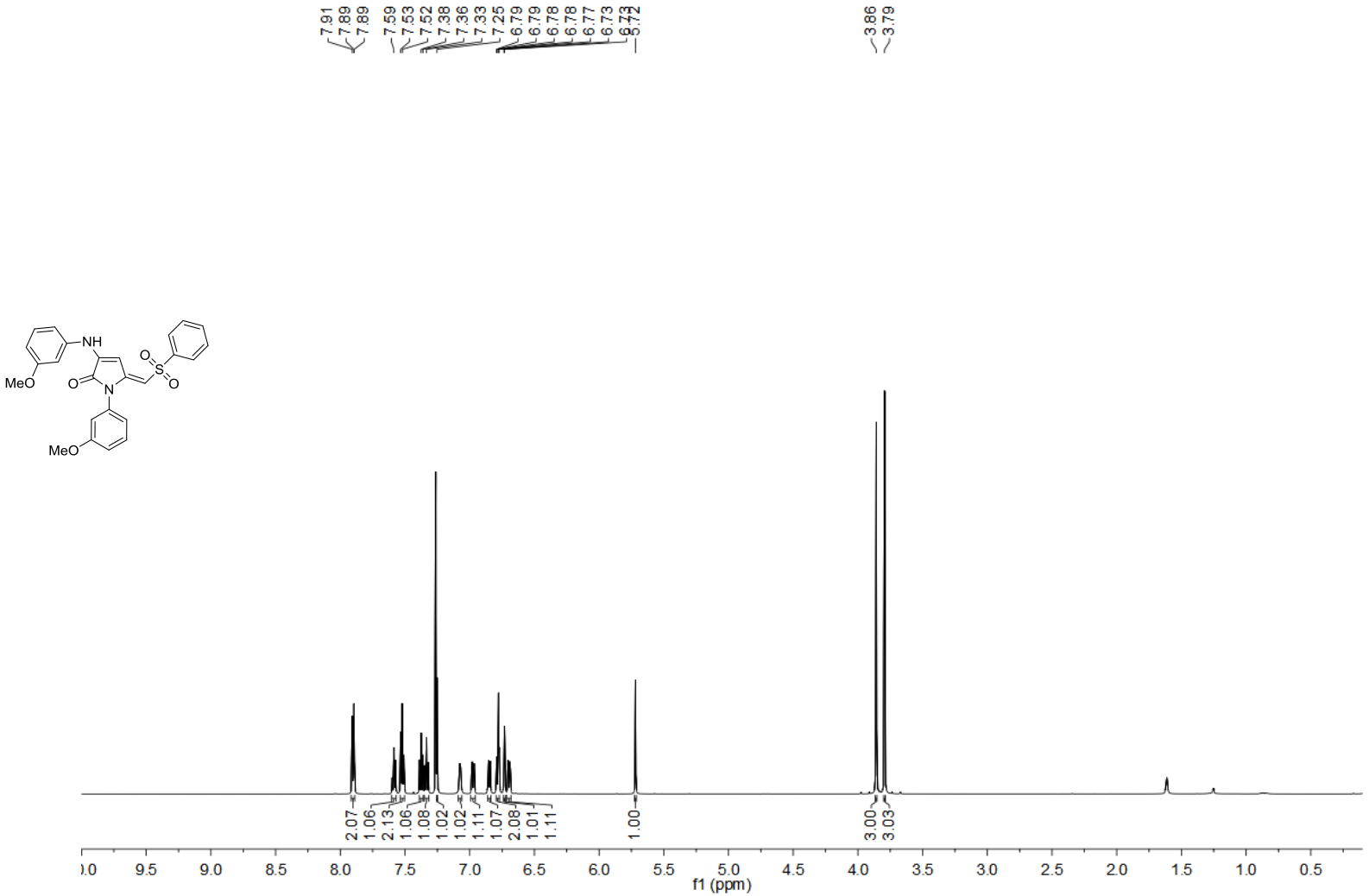


Figure S16. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4f**

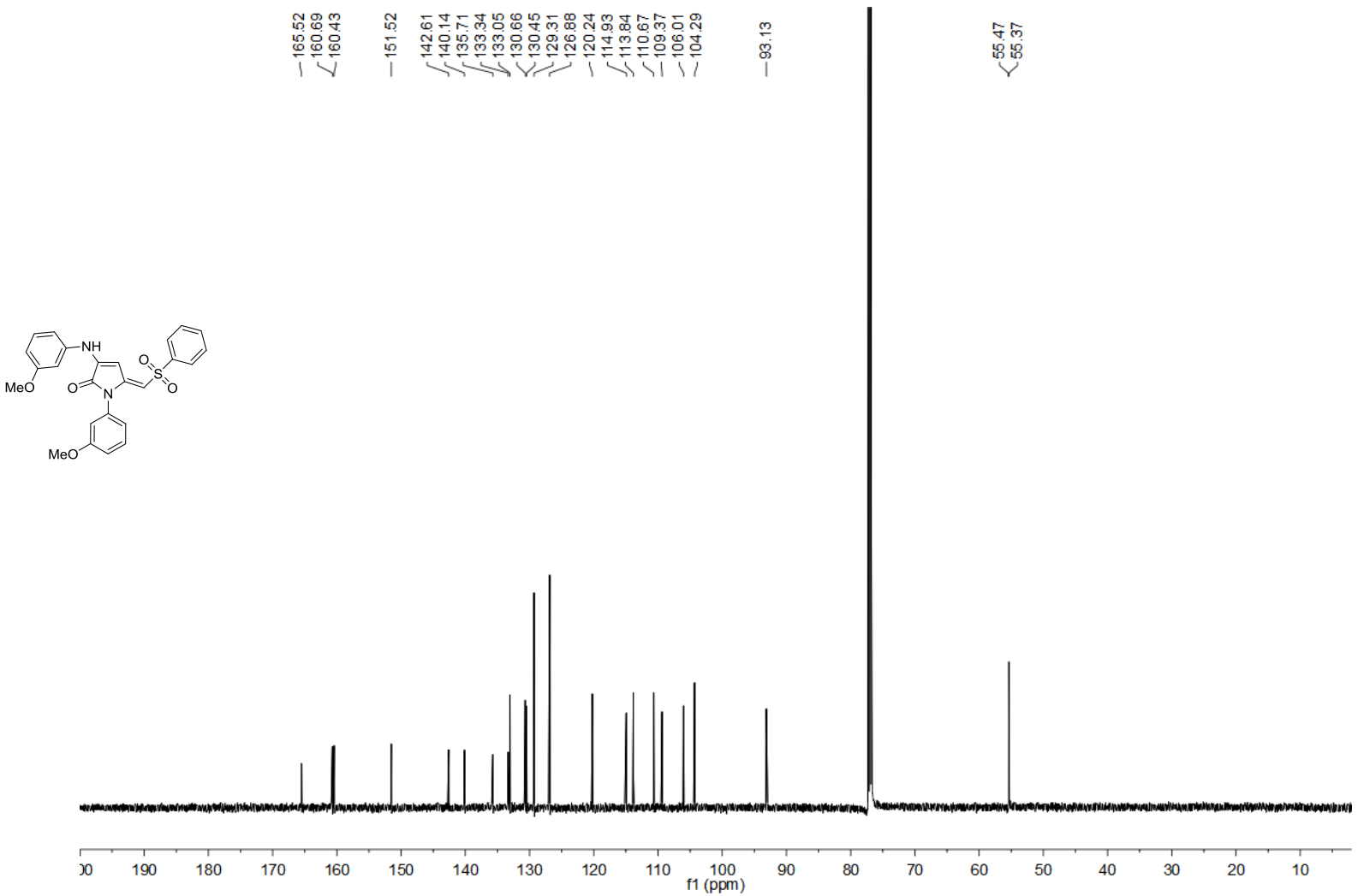


Figure S17. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4f**

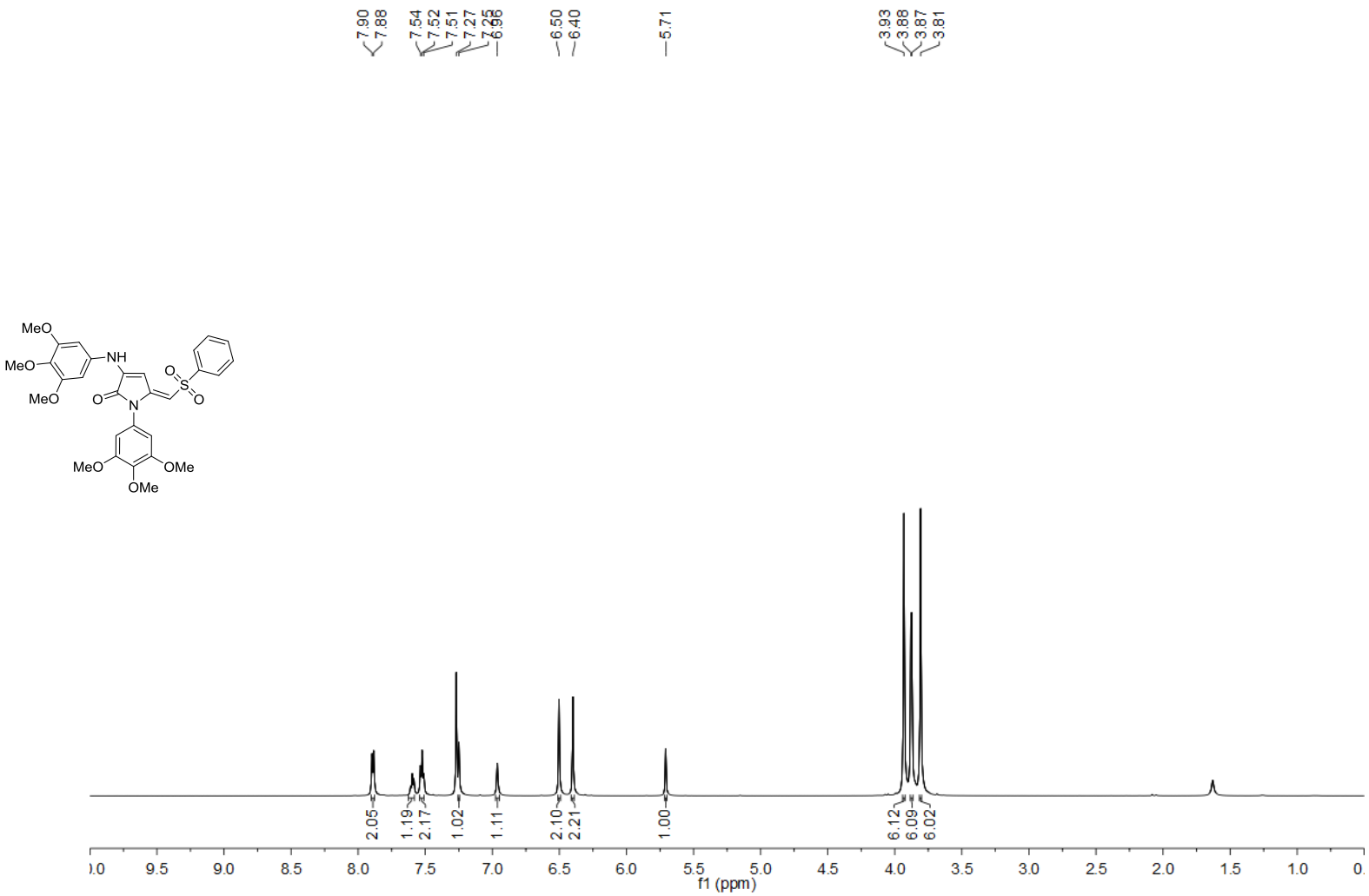


Figure S18. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4g**

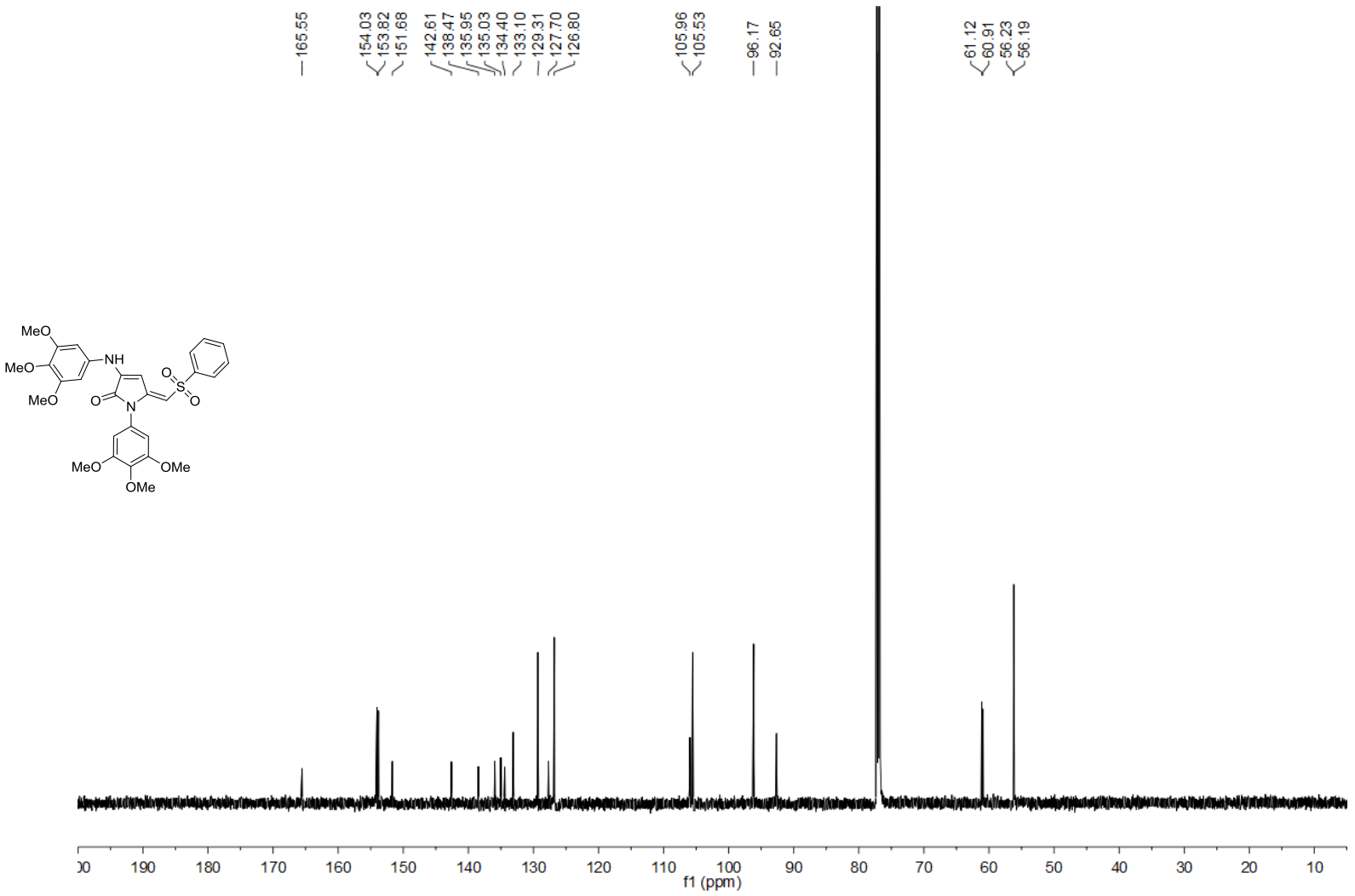


Figure S19. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4g**

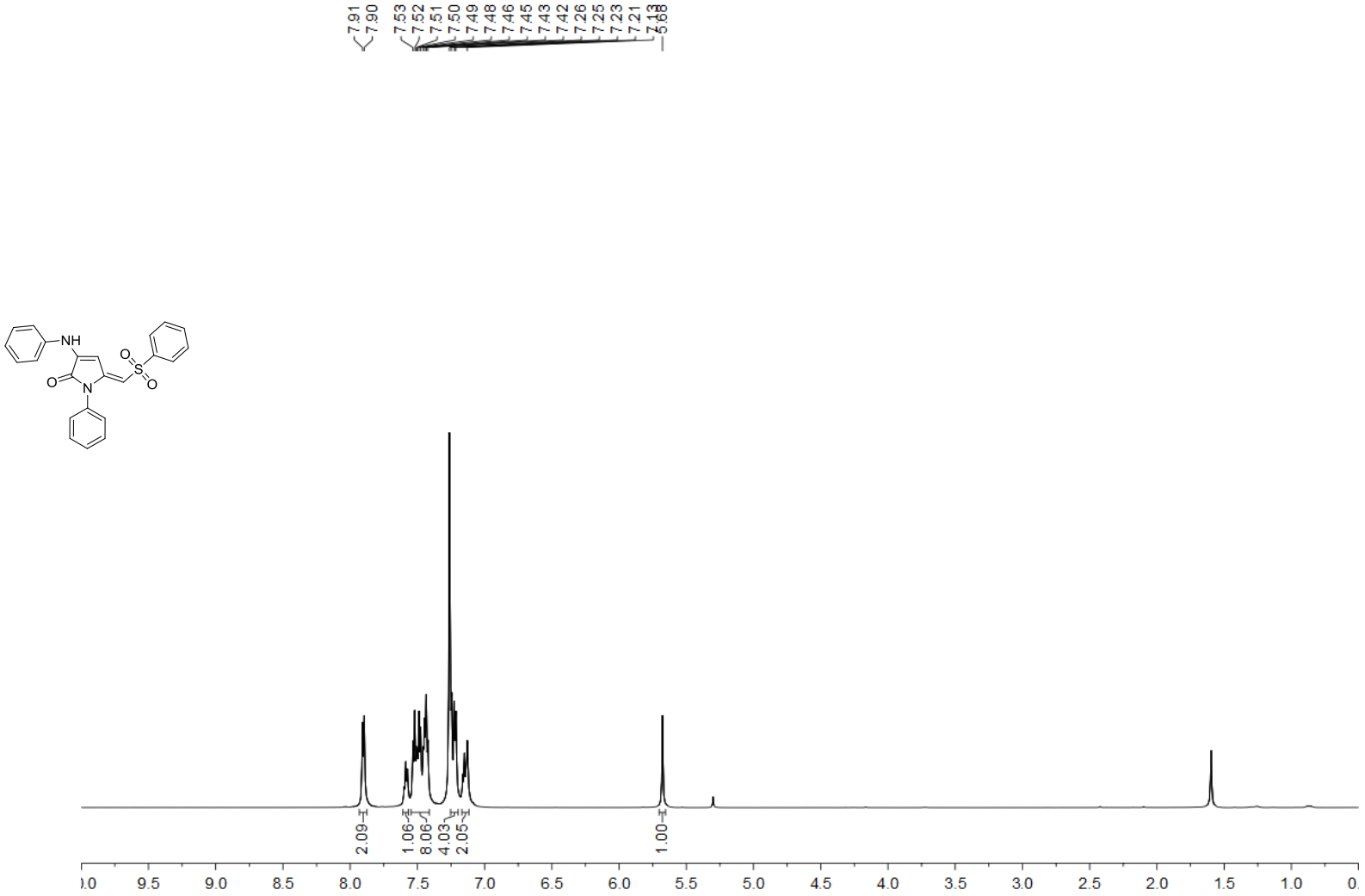


Figure S20. **^{1}H NMR** (600 MHz, $CDCl_3$) spectra of compound **4h**

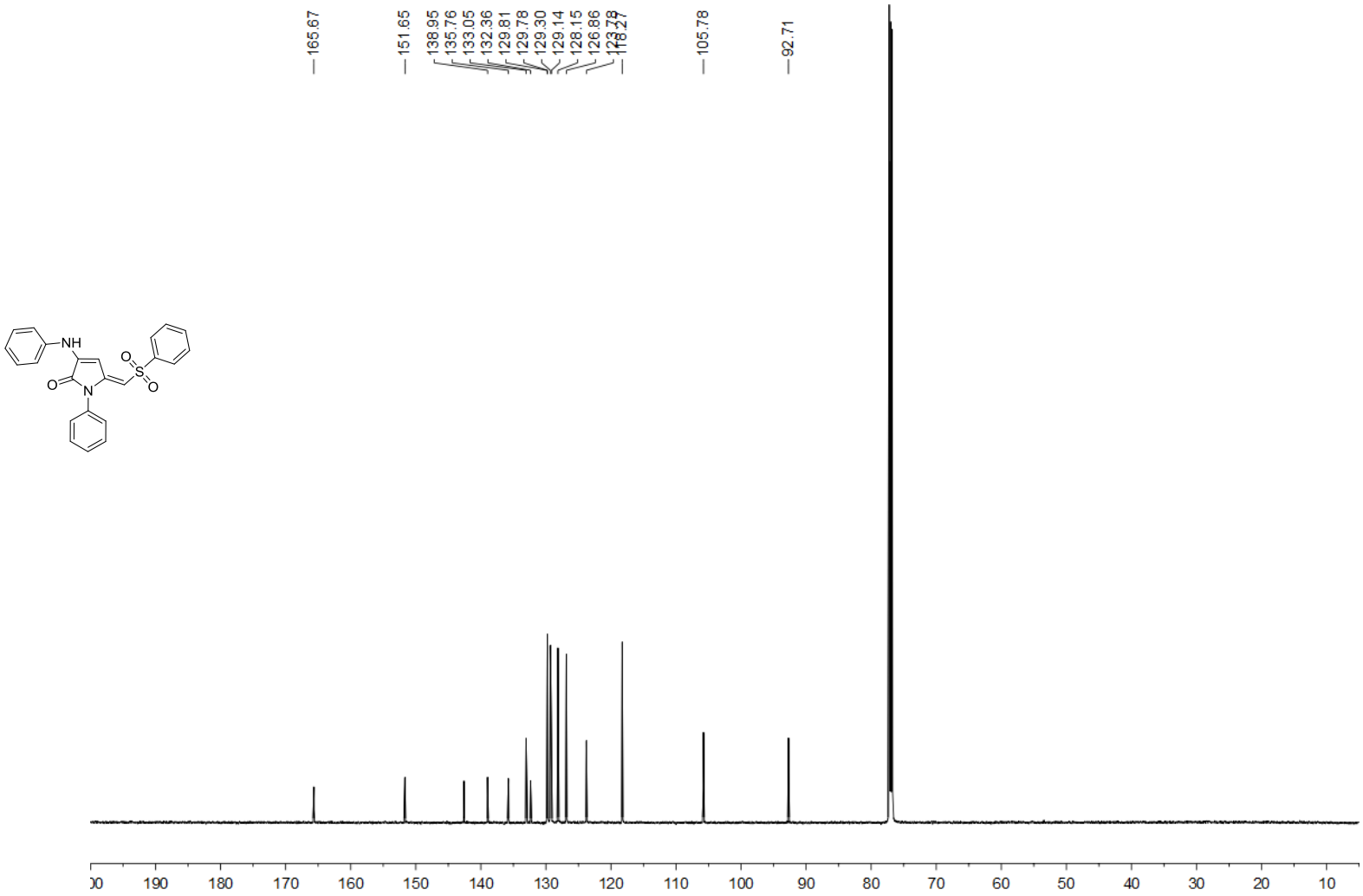


Figure S21. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4h**

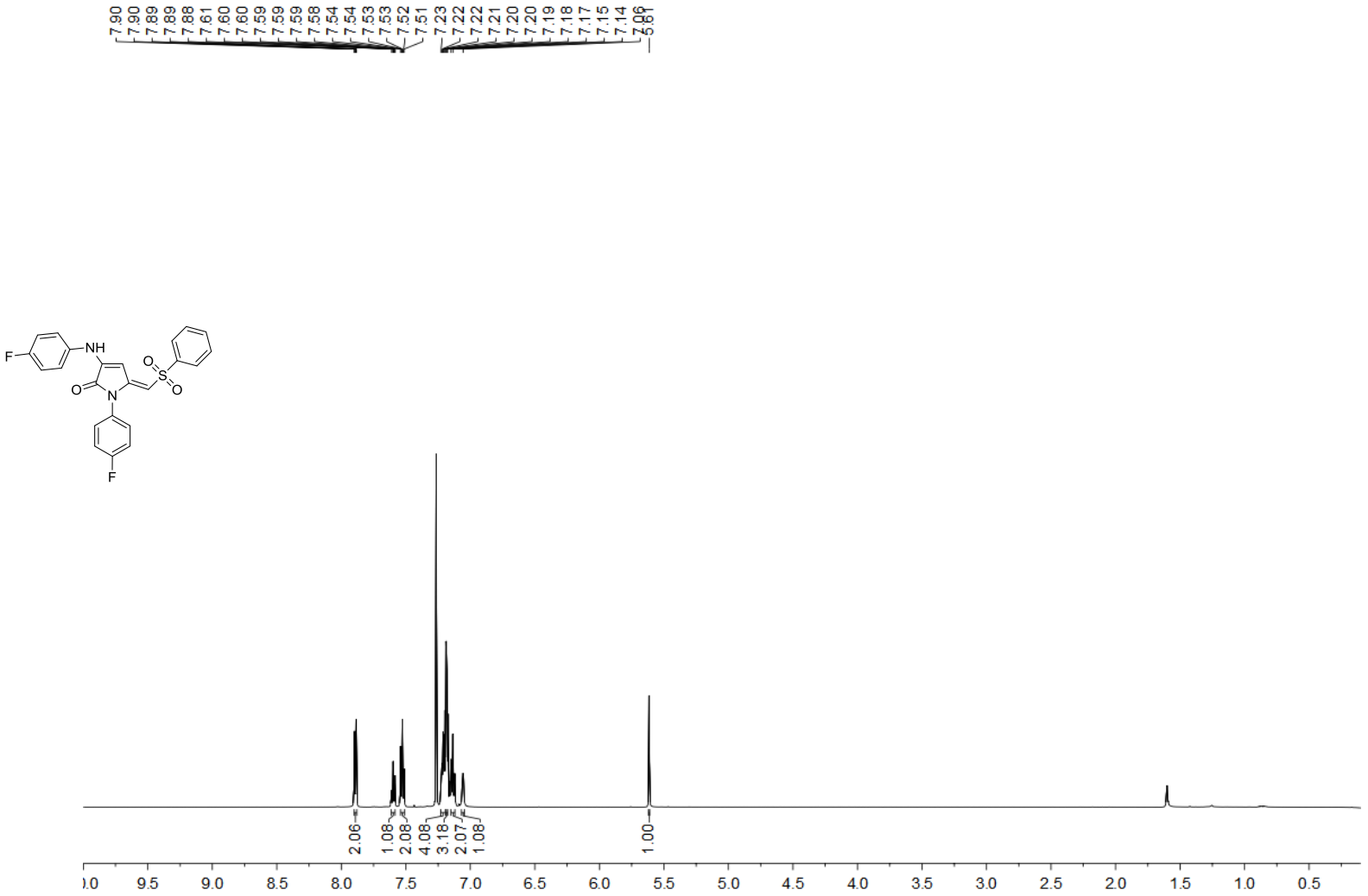


Figure S22. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4i**

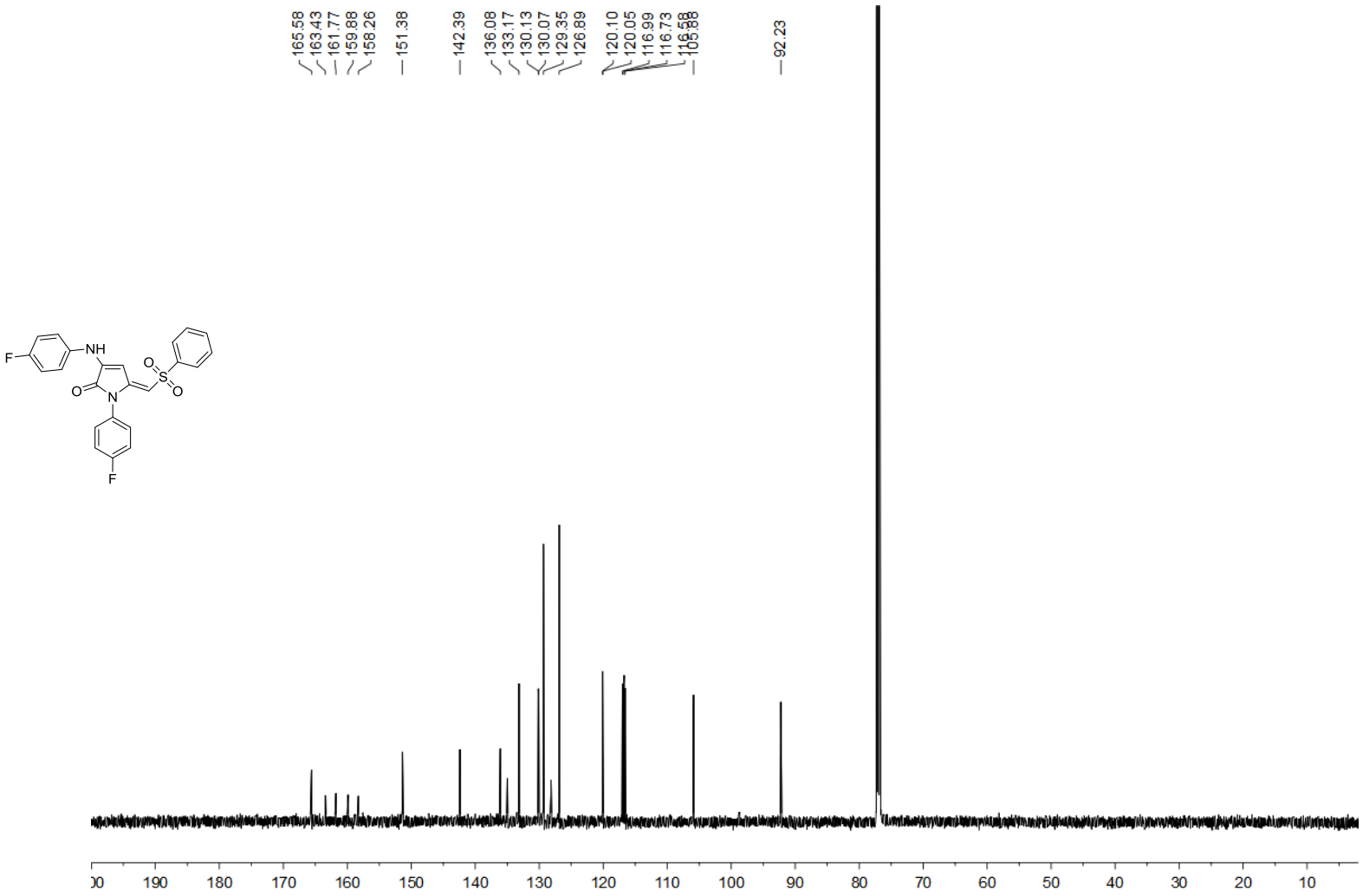


Figure S23. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4i**

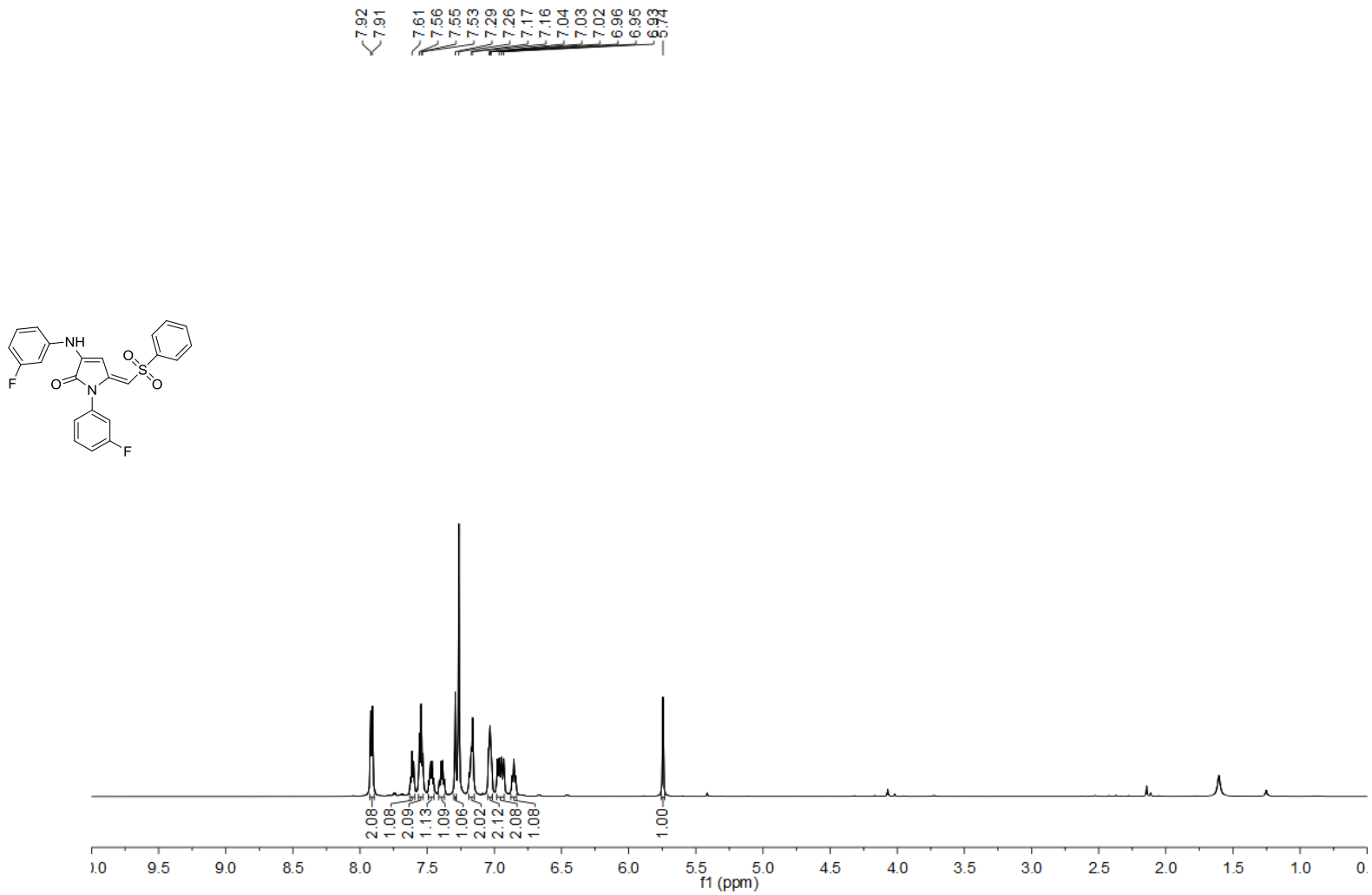


Figure S24. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4j**

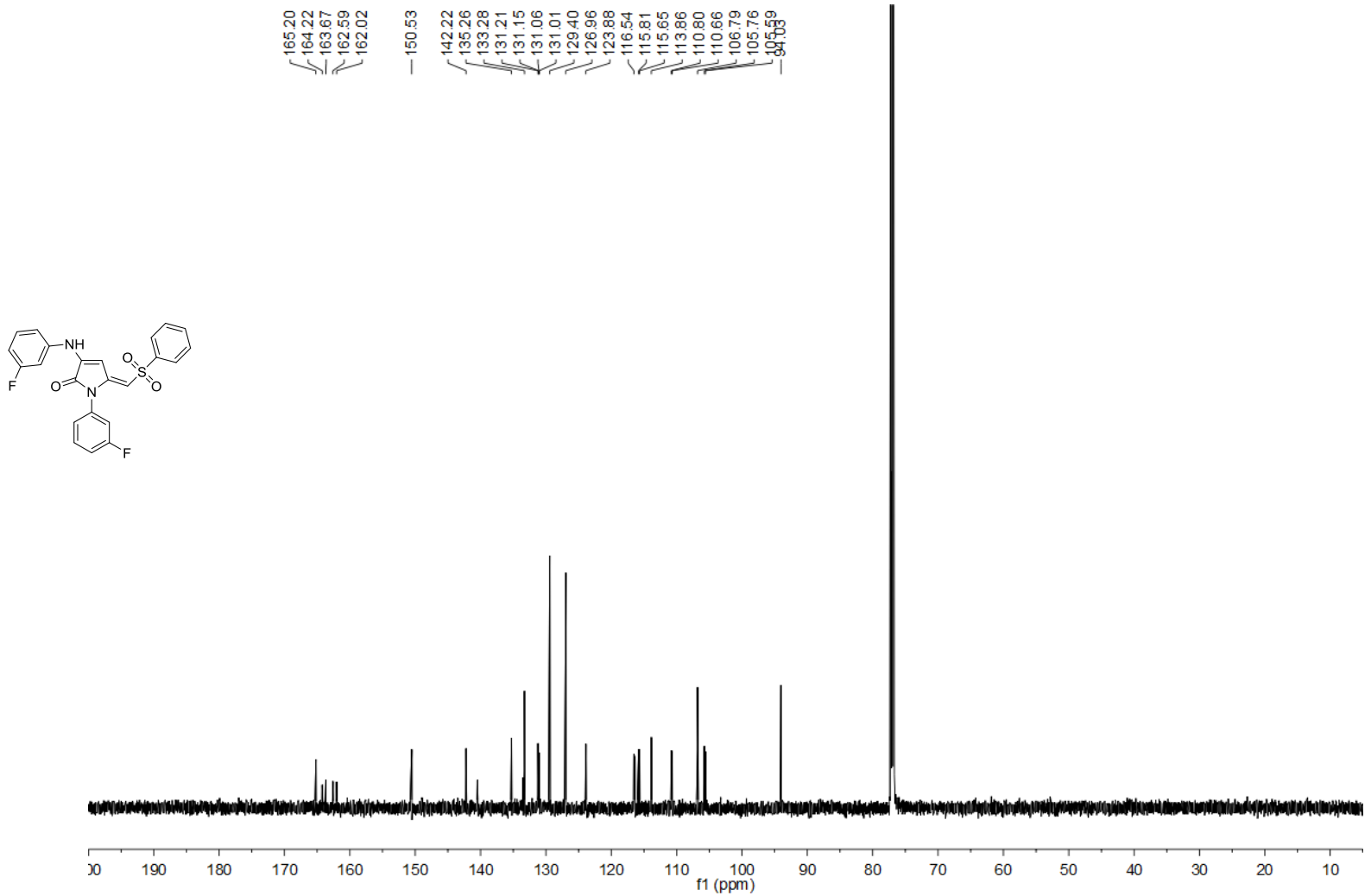


Figure S25. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4j**

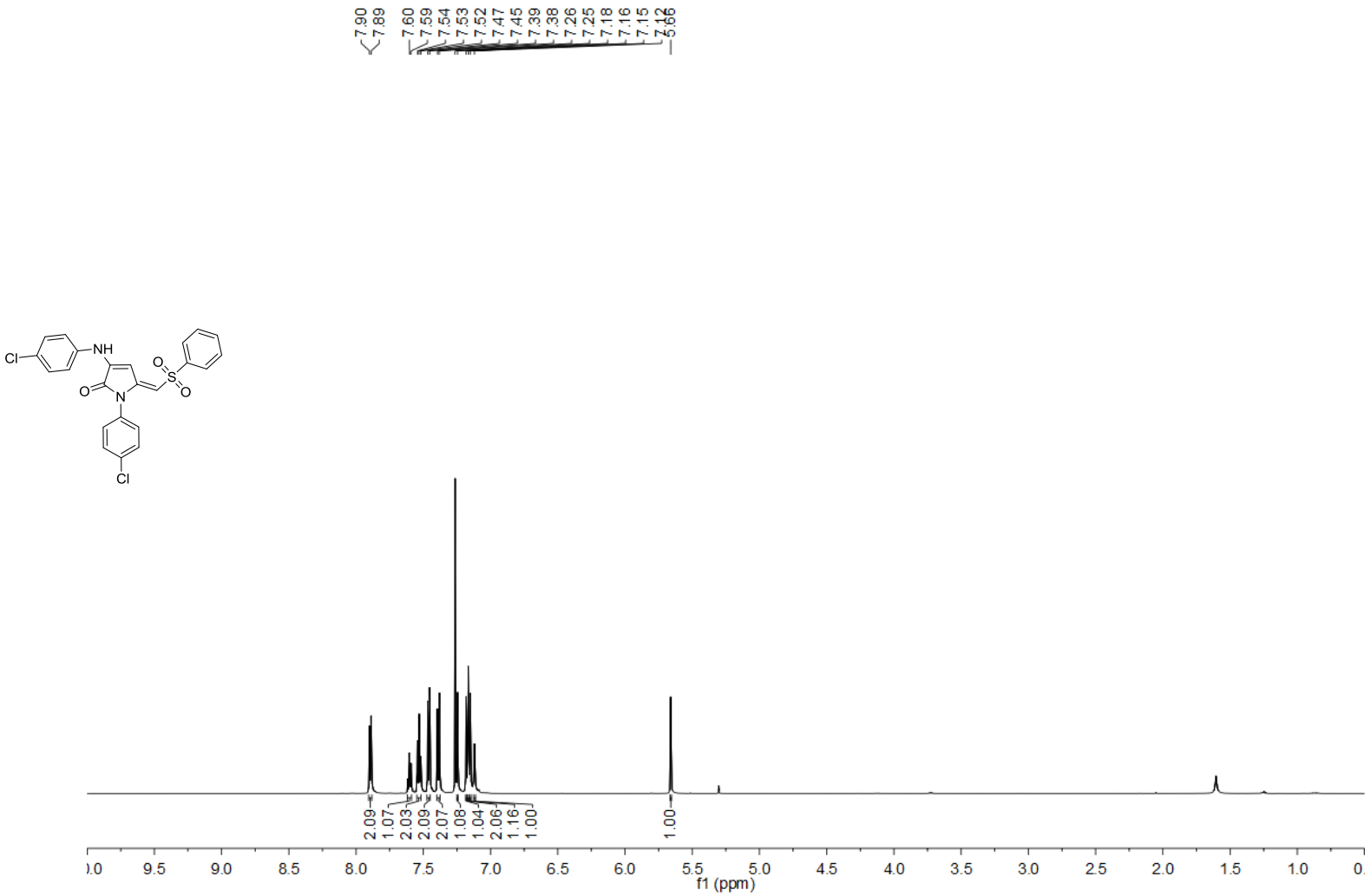


Figure S26. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4k**

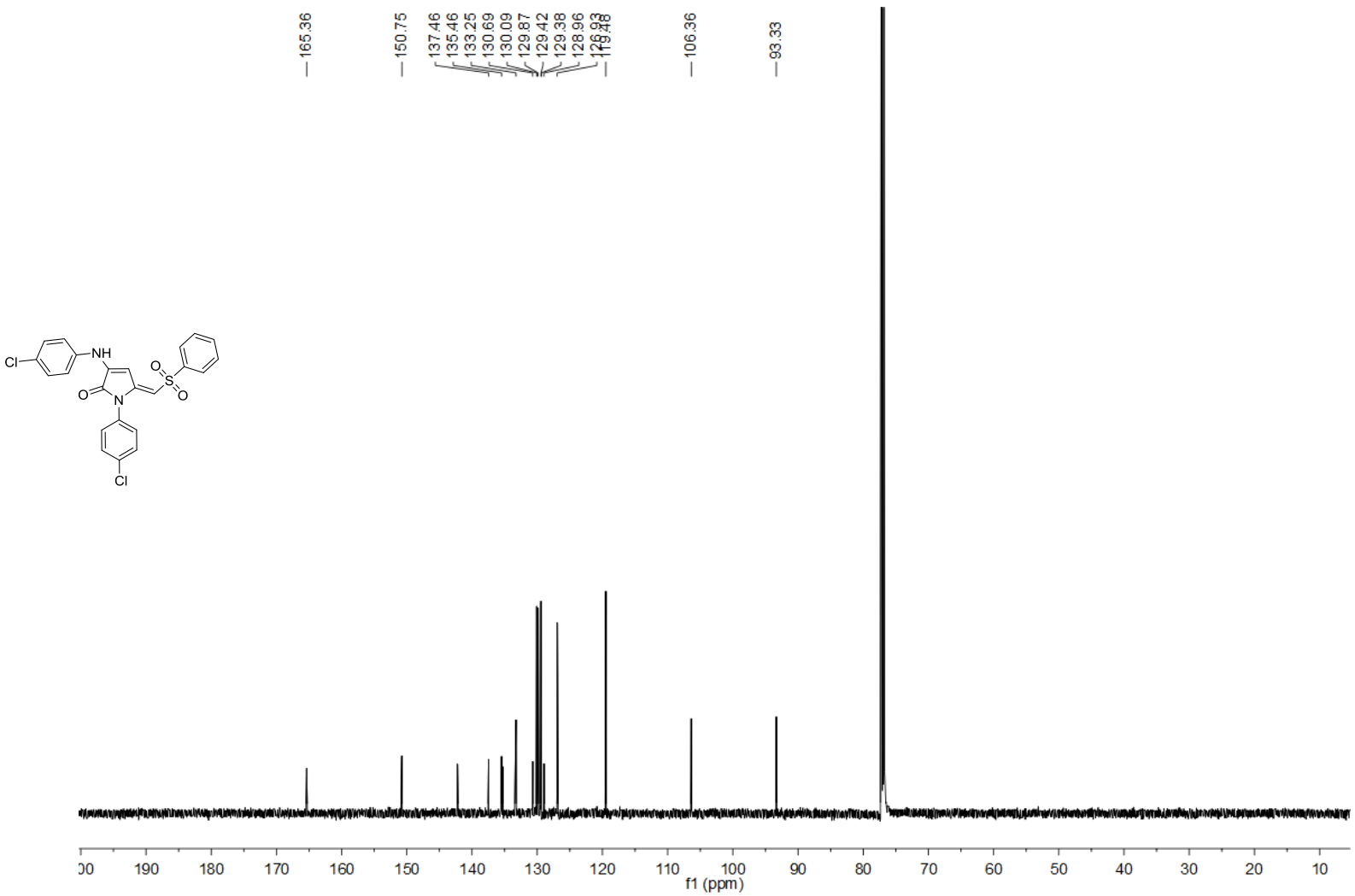


Figure S27. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4k**

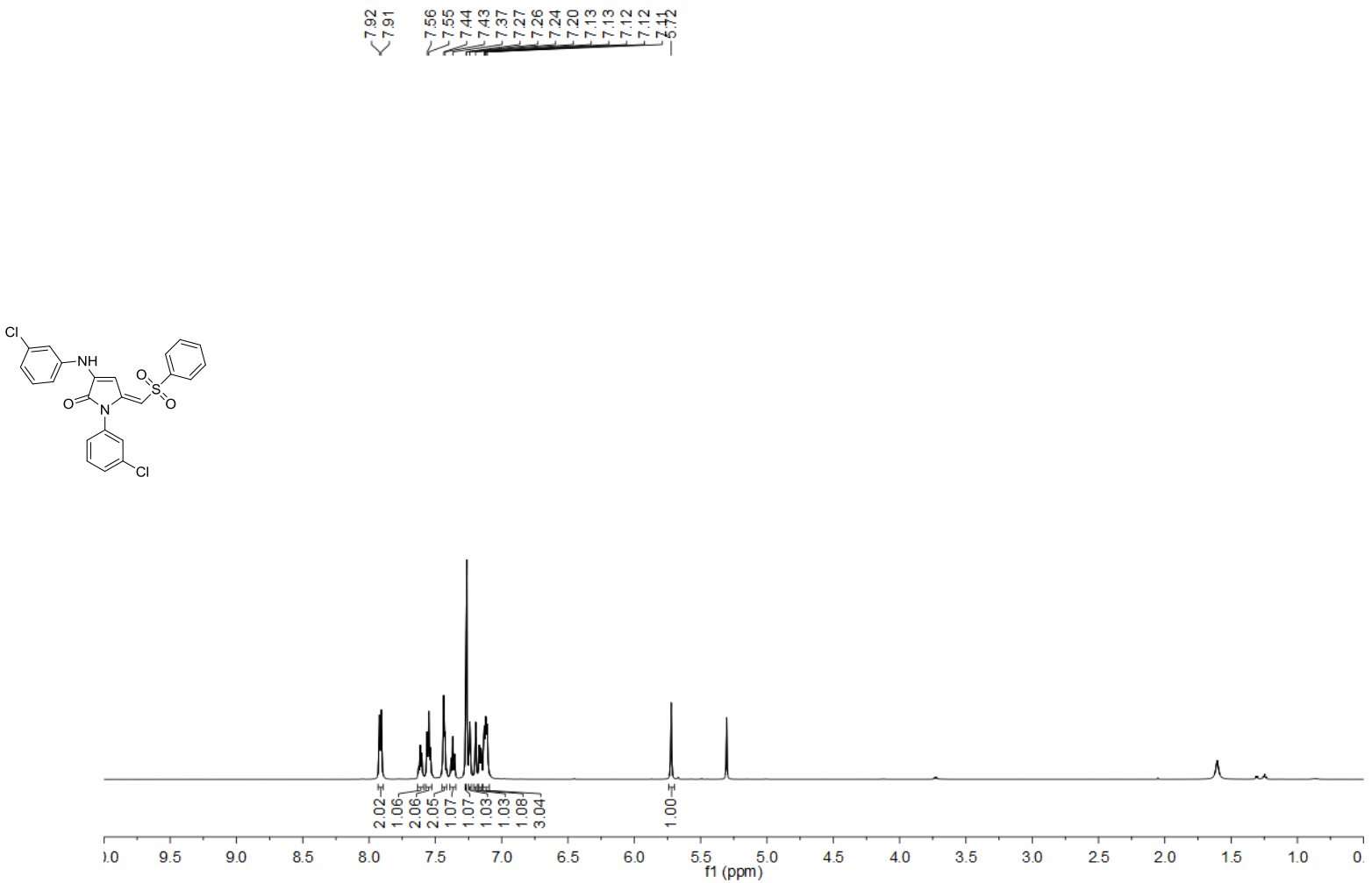


Figure S28. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4l**

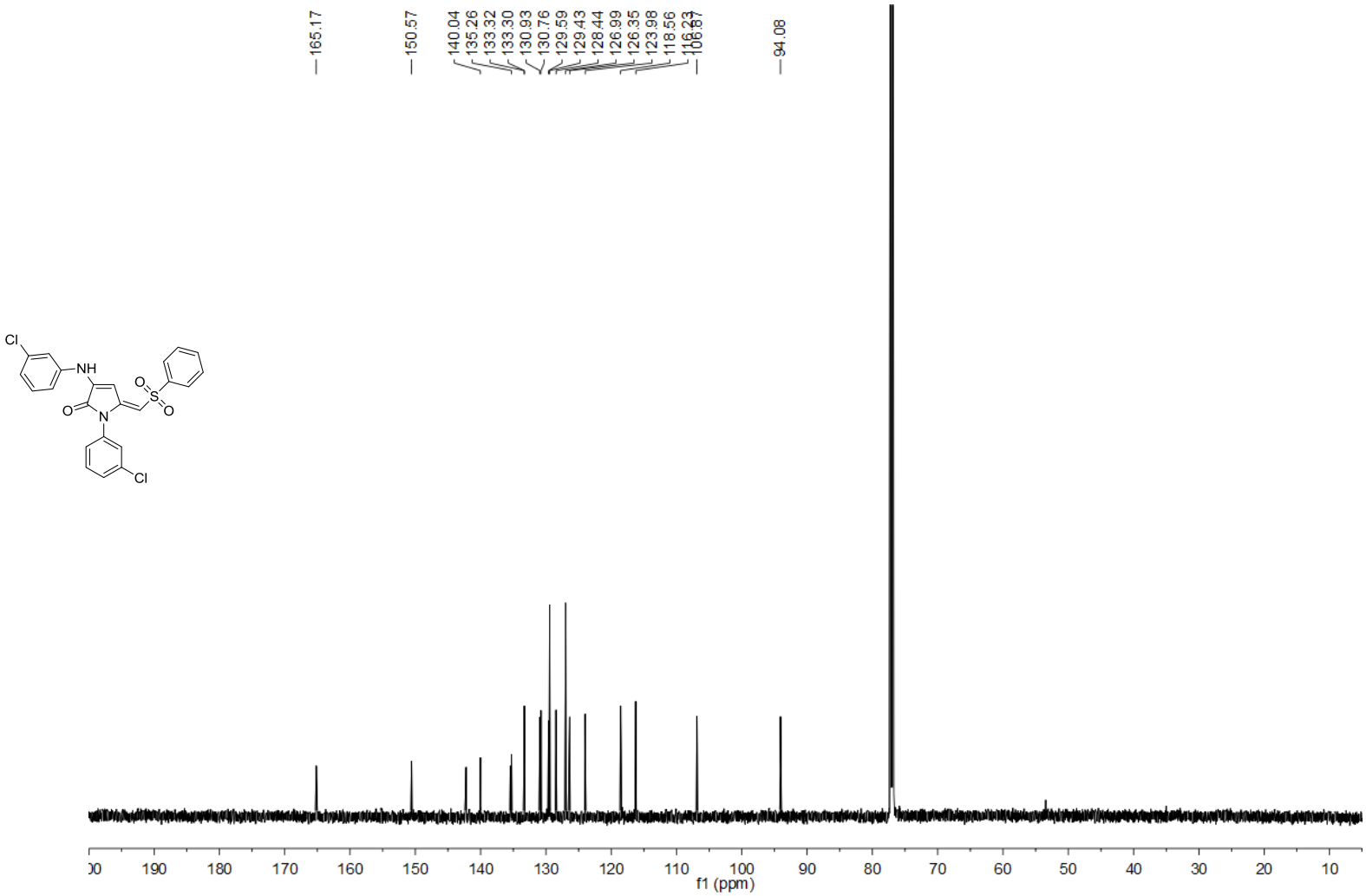


Figure S29. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4l**

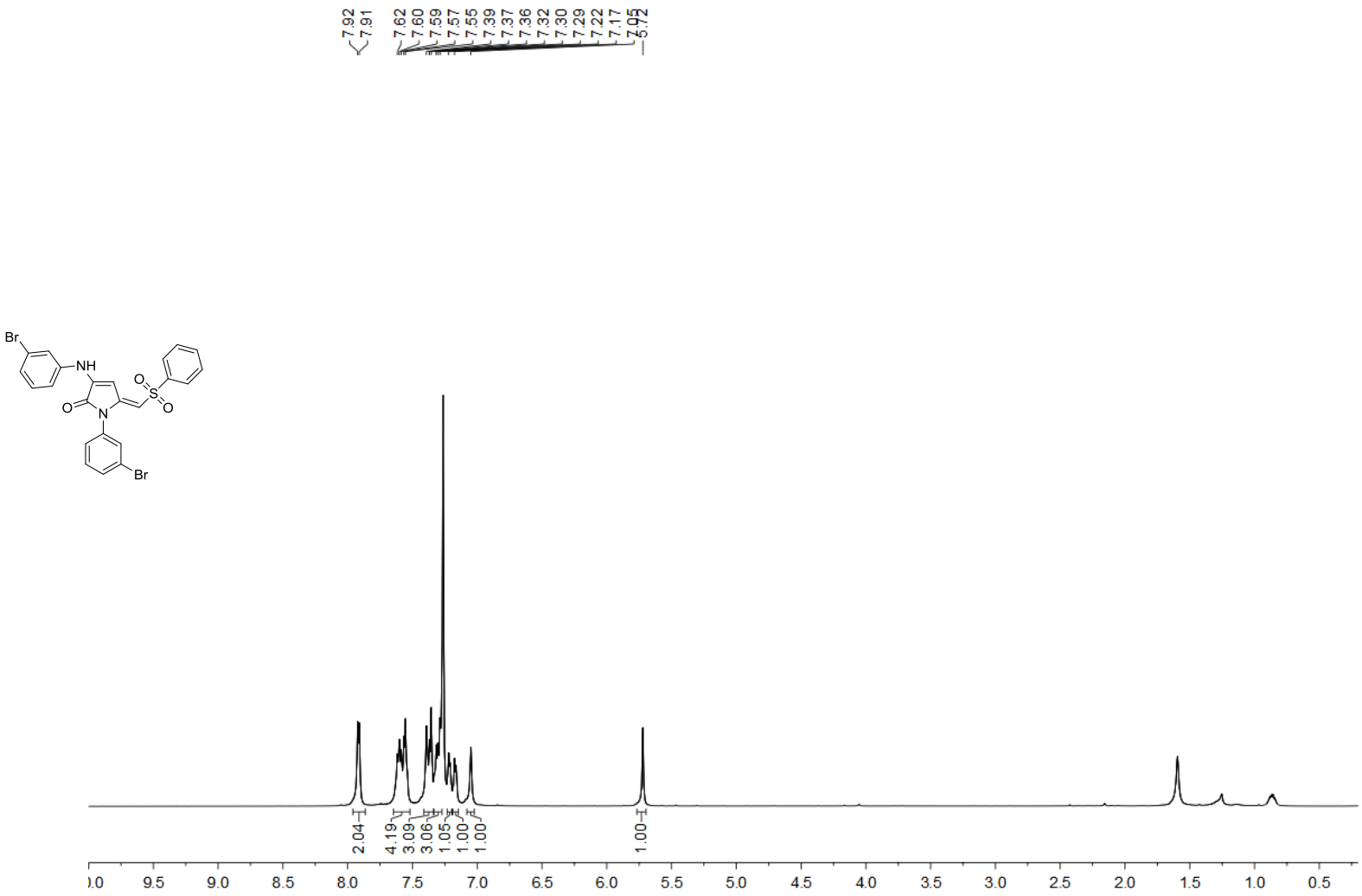


Figure S30 **^{1}H NMR** (600 MHz, $CDCl_3$) spectra of compound **4m**

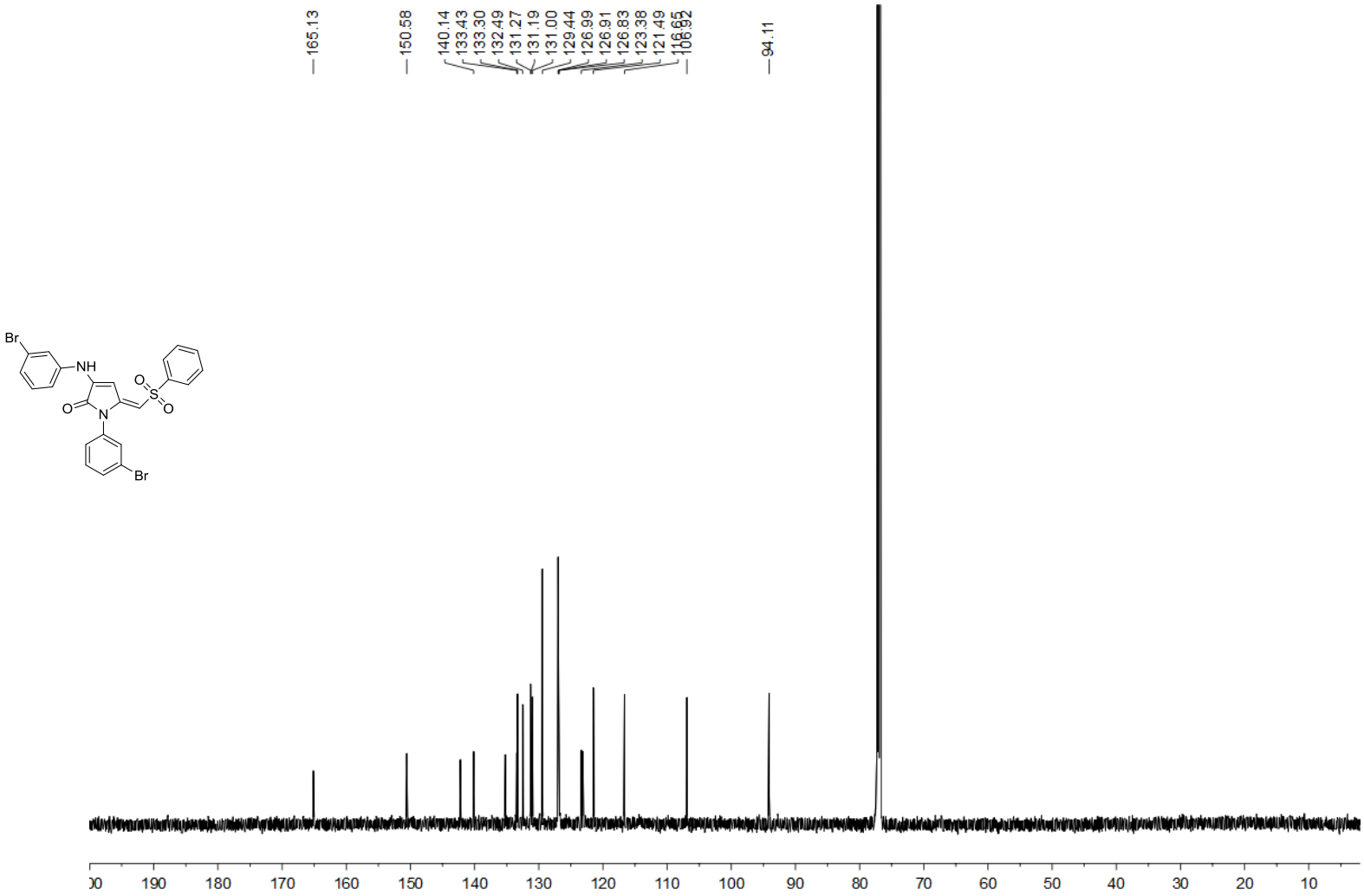


Figure S31. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4m**

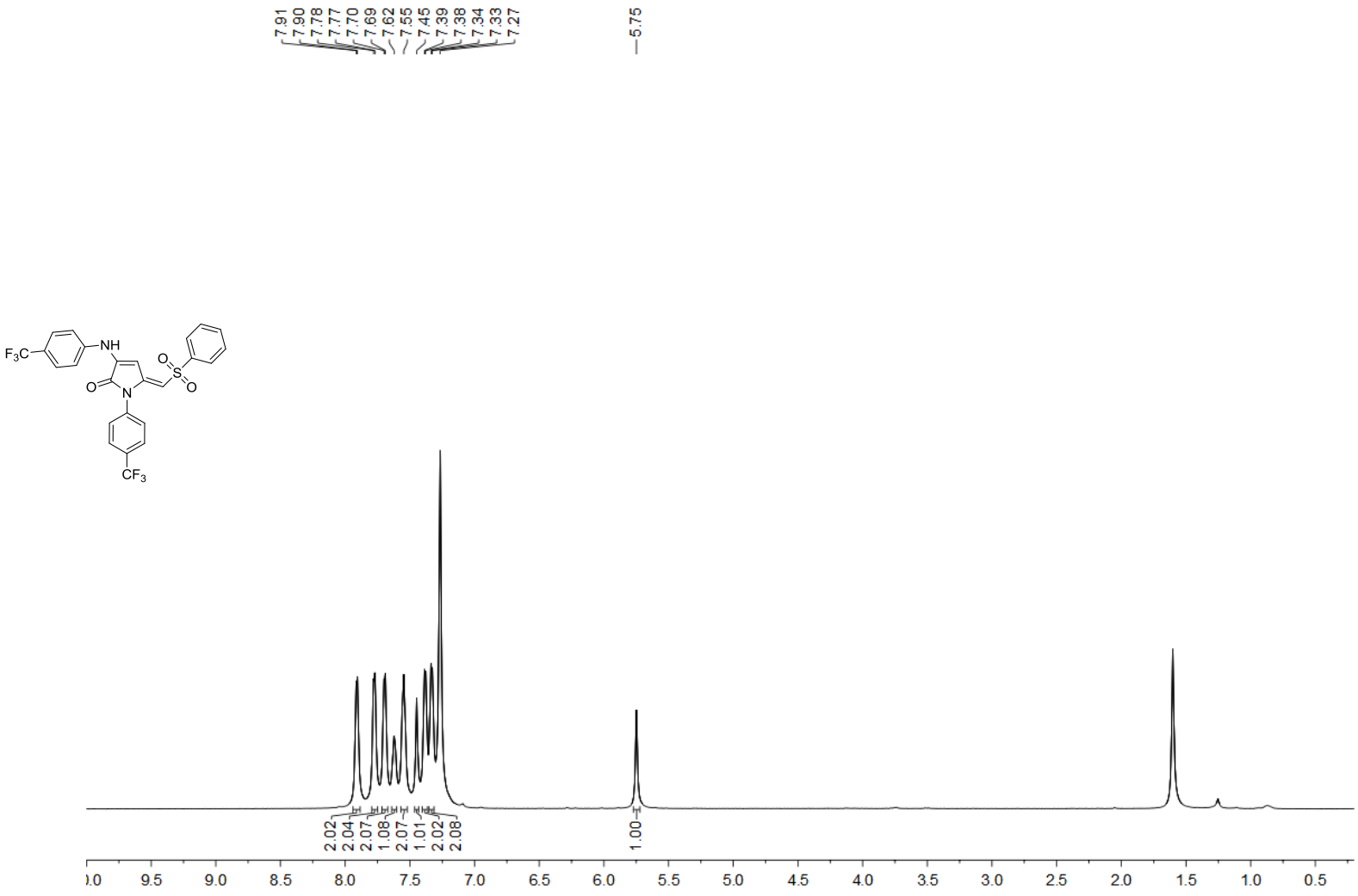


Figure S32. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4n**

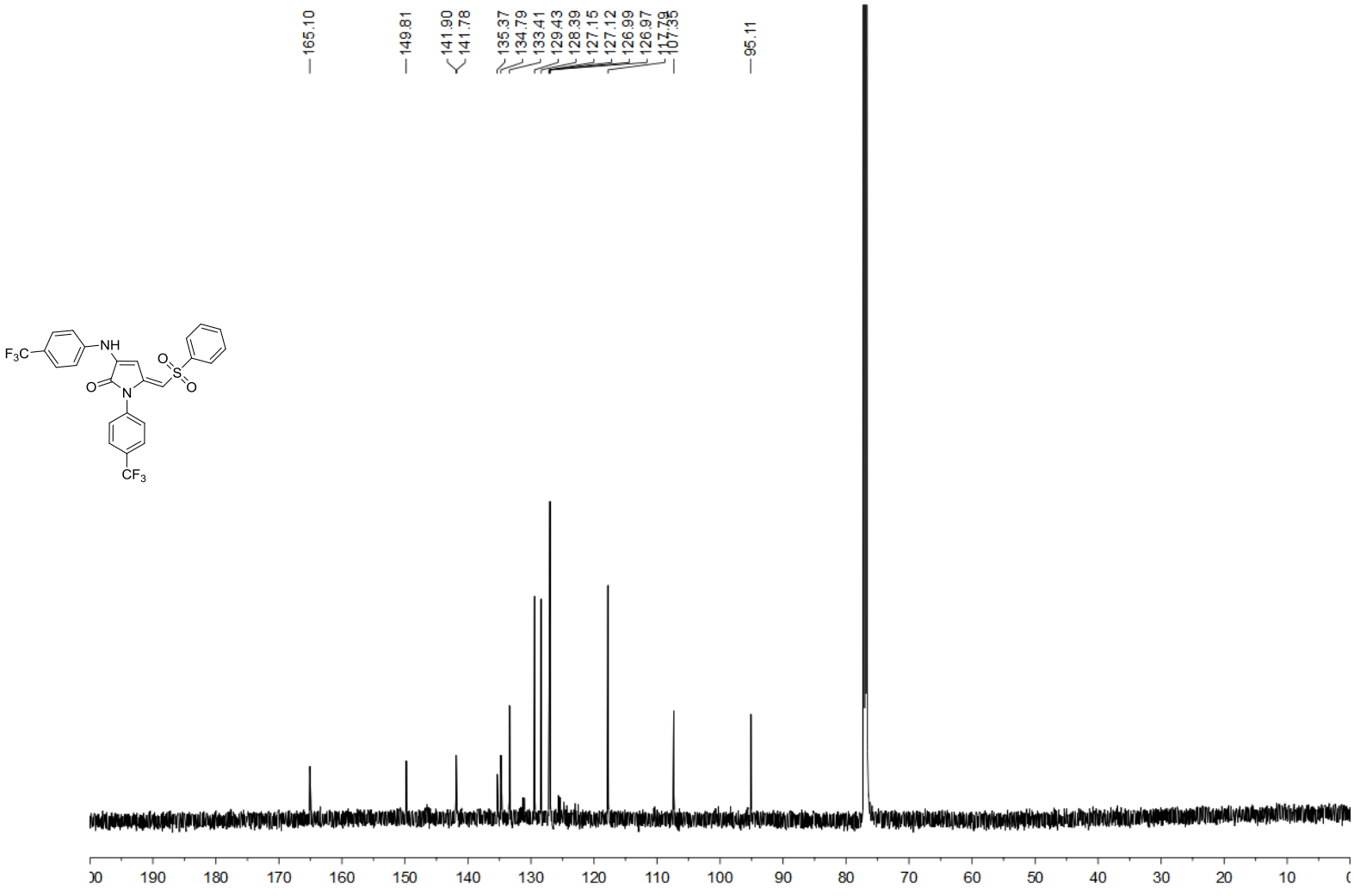


Figure S33. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4n**

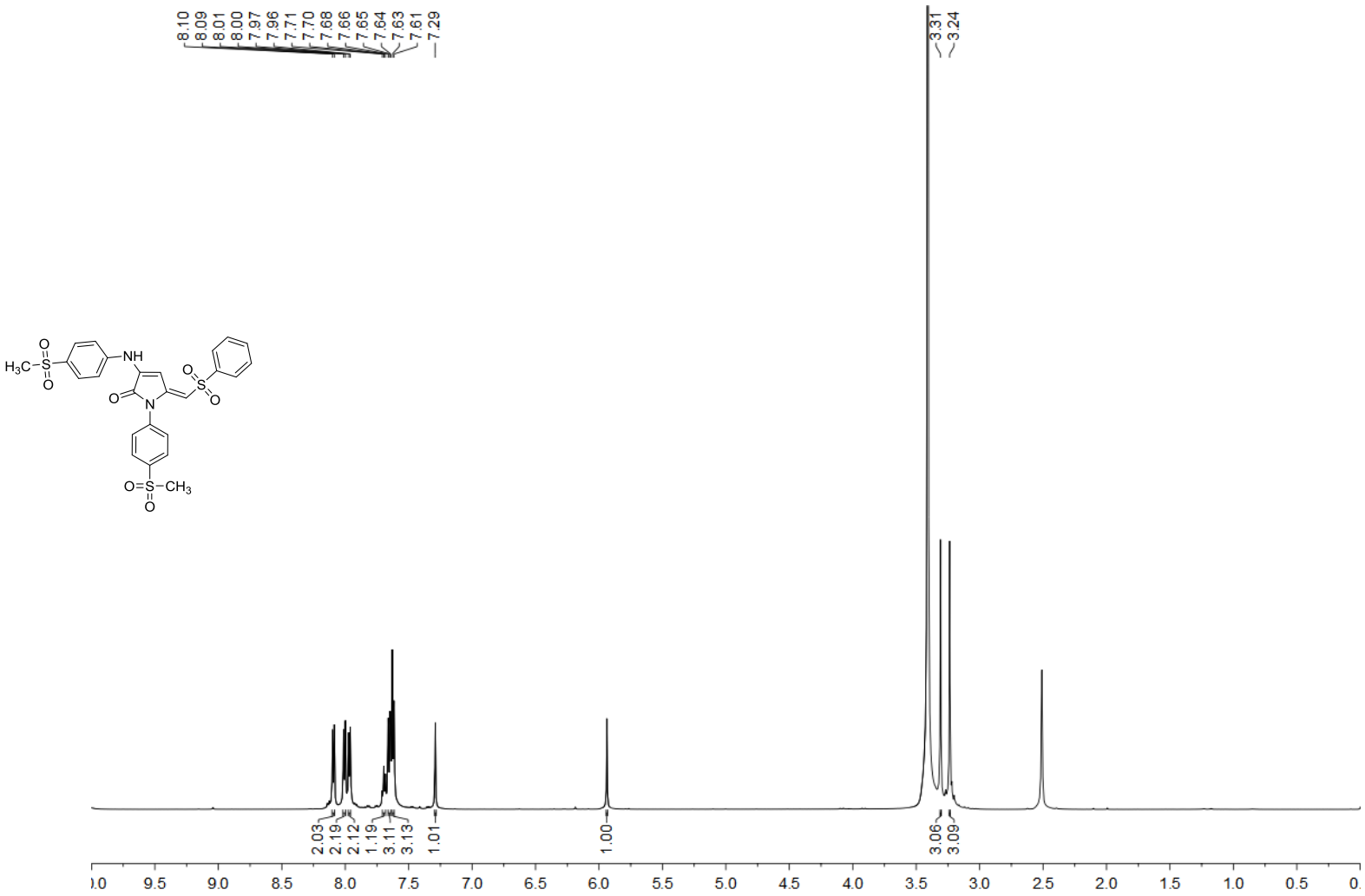


Figure S34. [1]**H NMR** (600 MHz, DMSO-d_6) spectra of compound **4o**

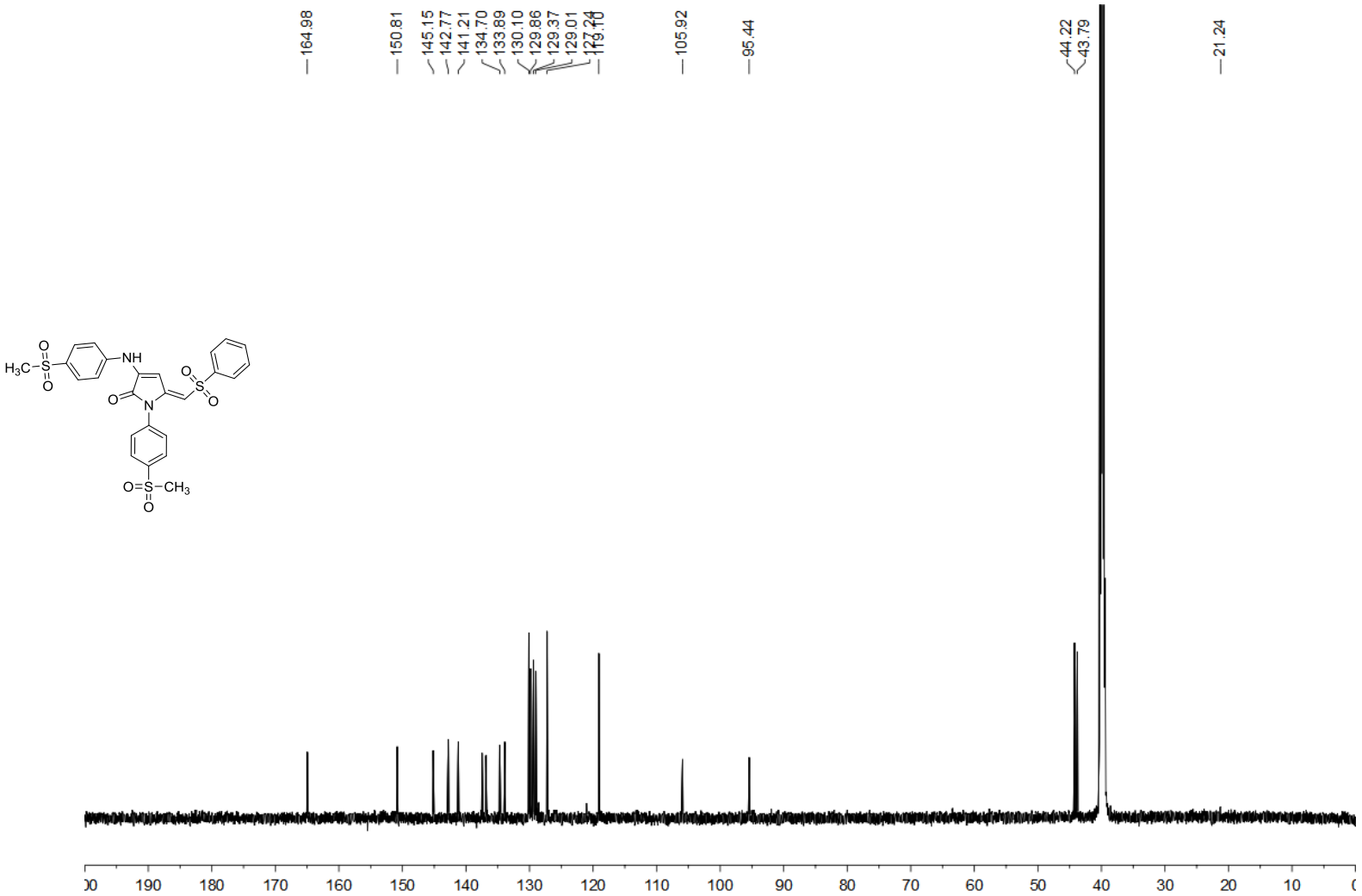


Figure S35. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **4o**

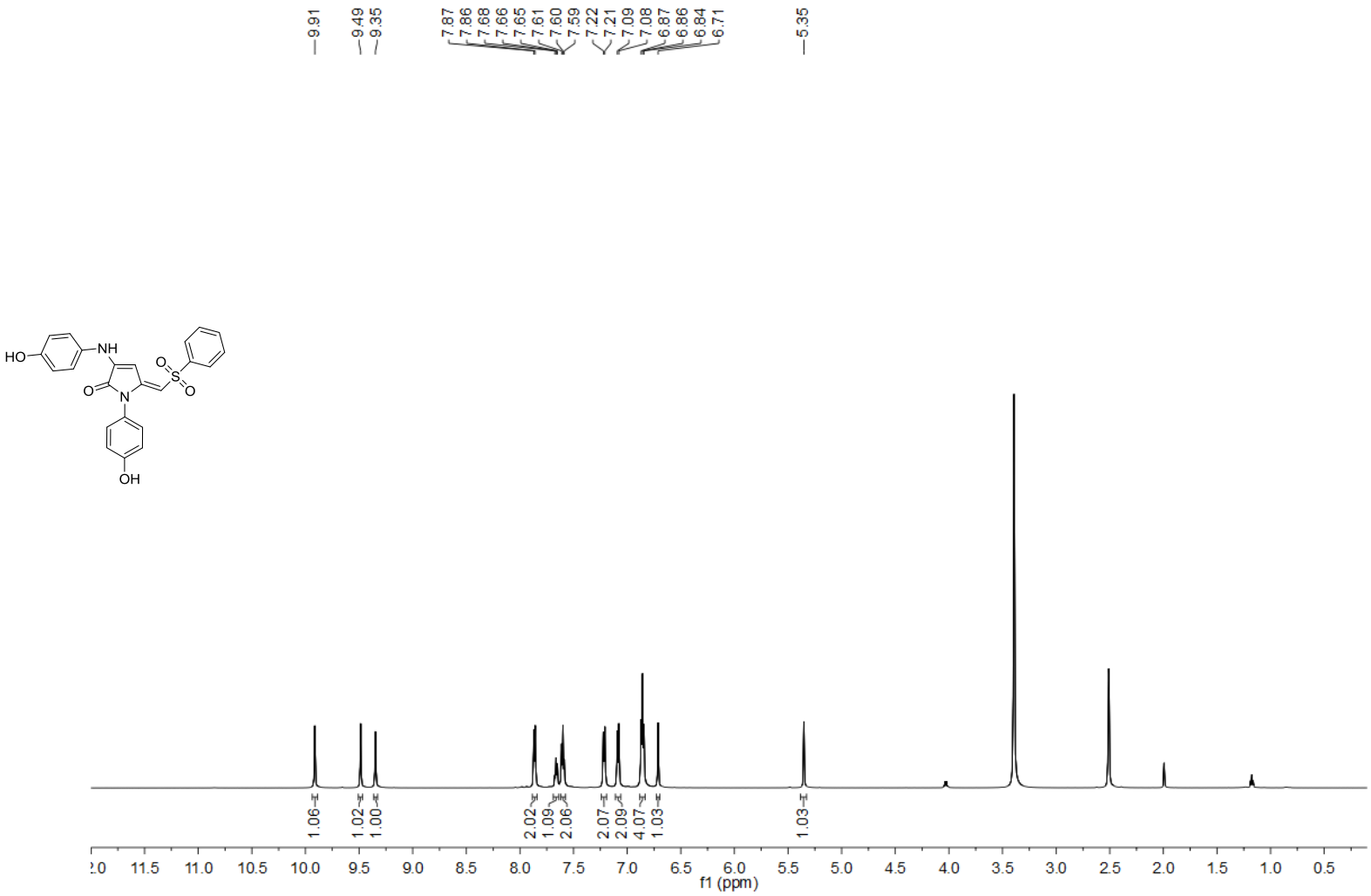


Figure S36. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **4p**

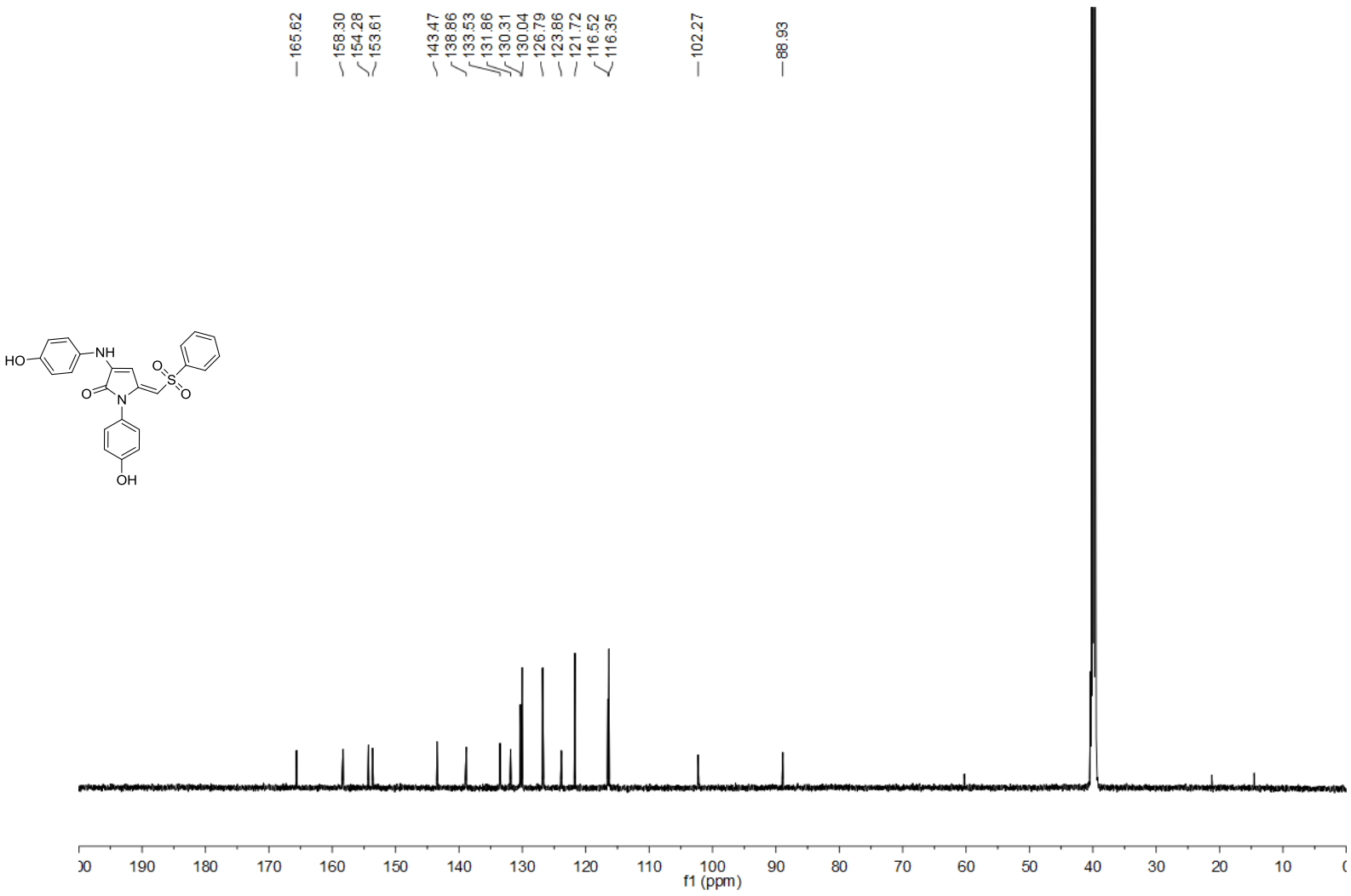


Figure S37. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **4p**

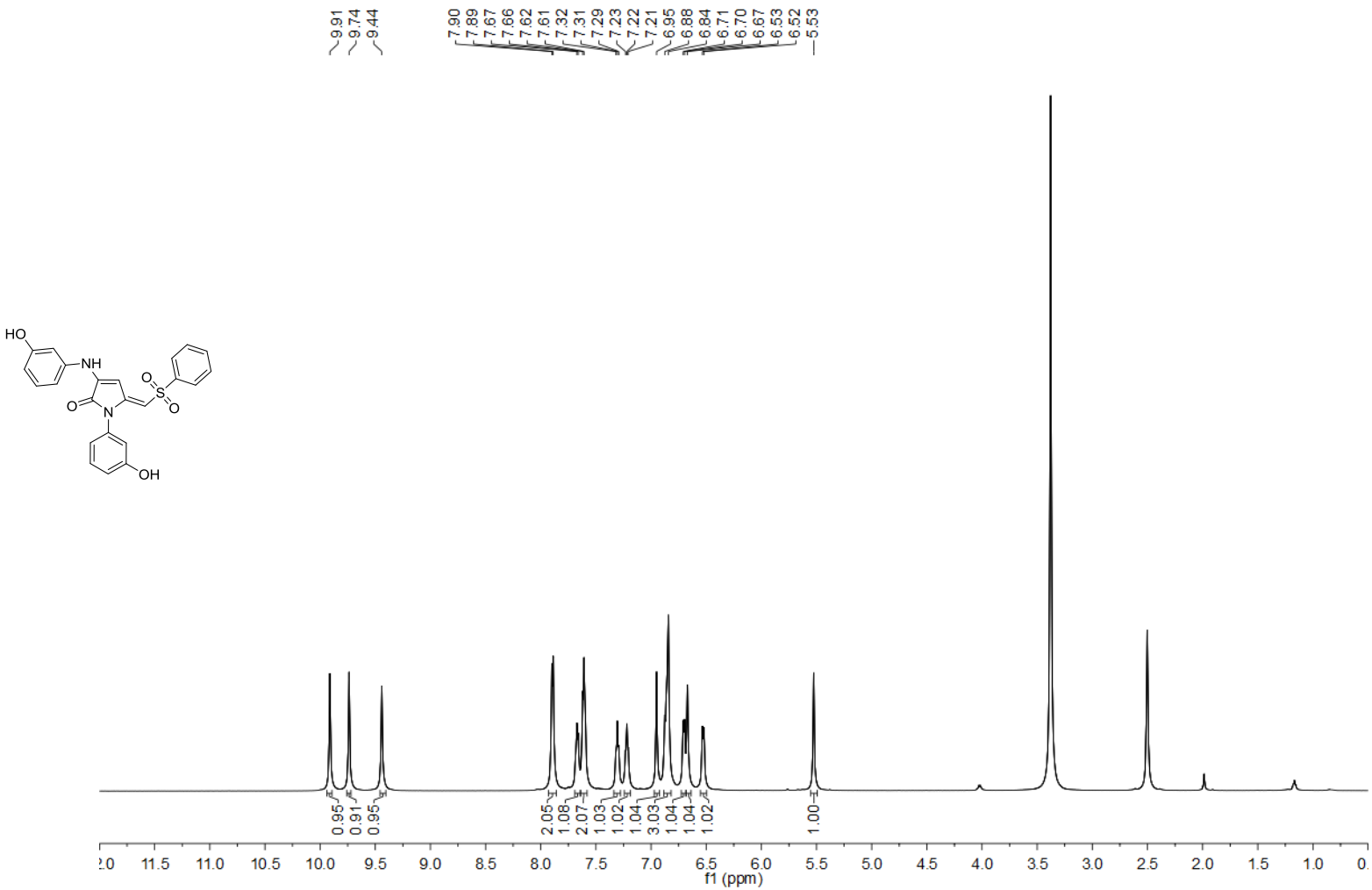


Figure S38. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **4q**

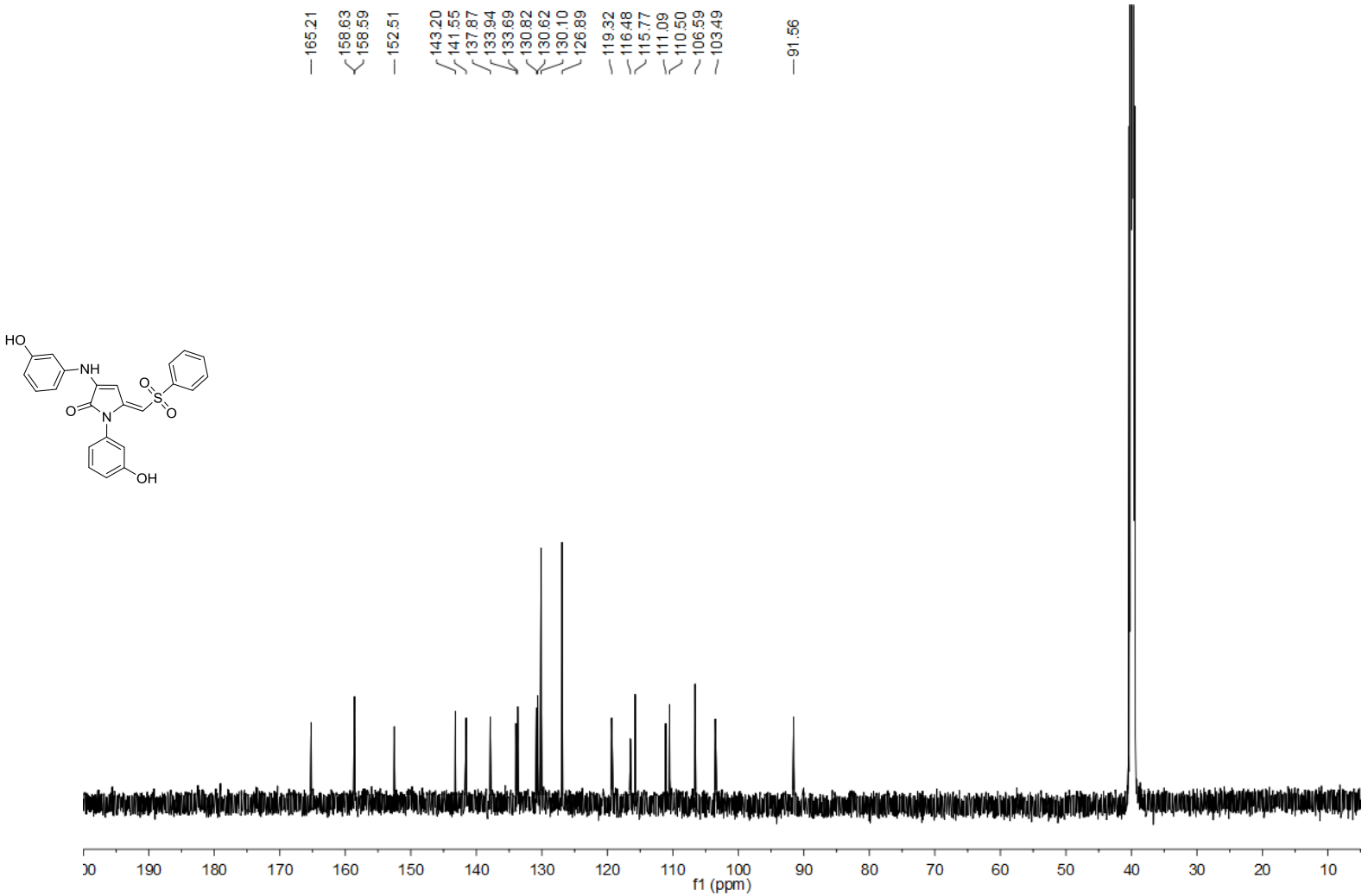


Figure S39. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **4q**

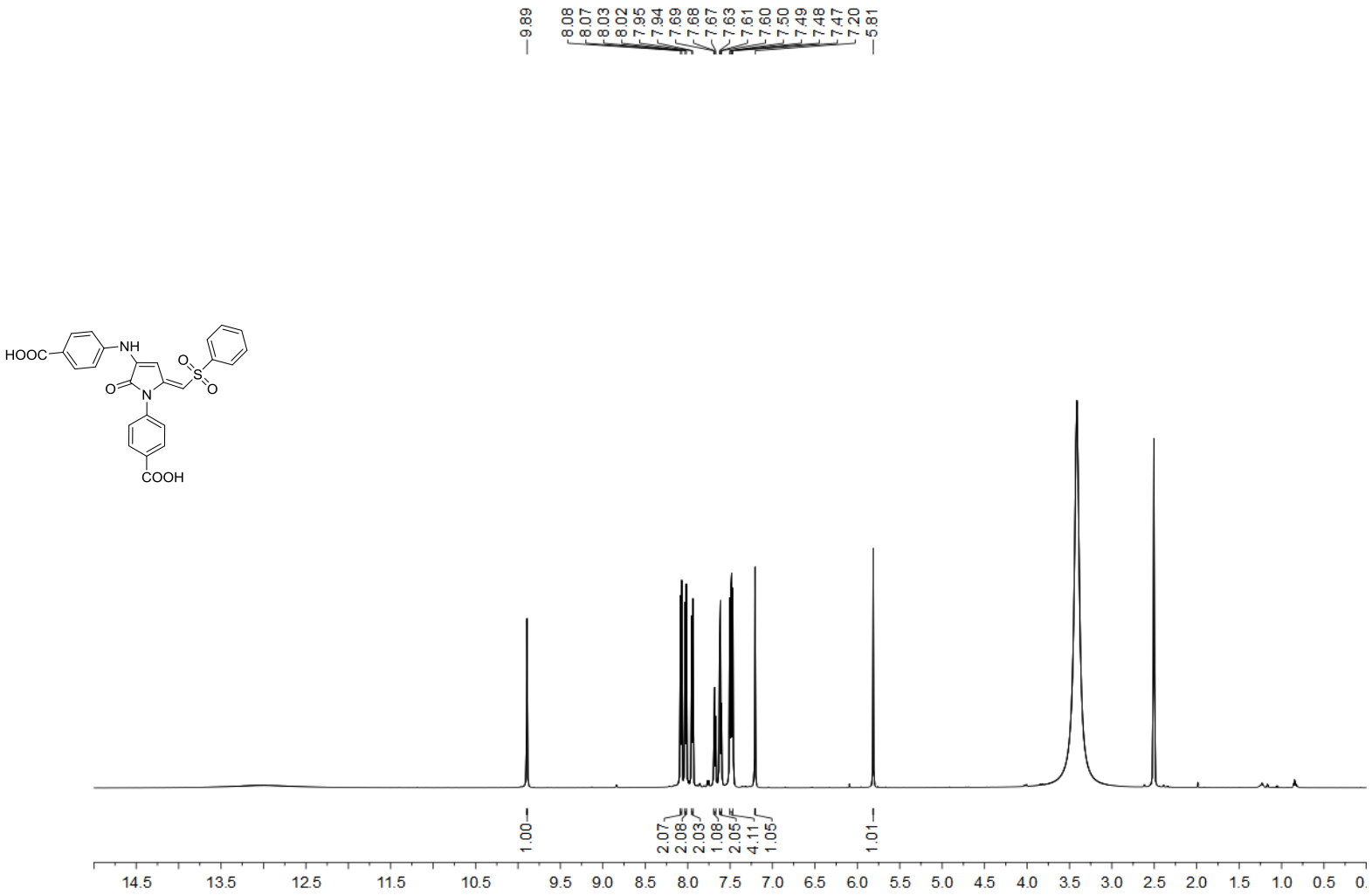


Figure S40. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **4r**

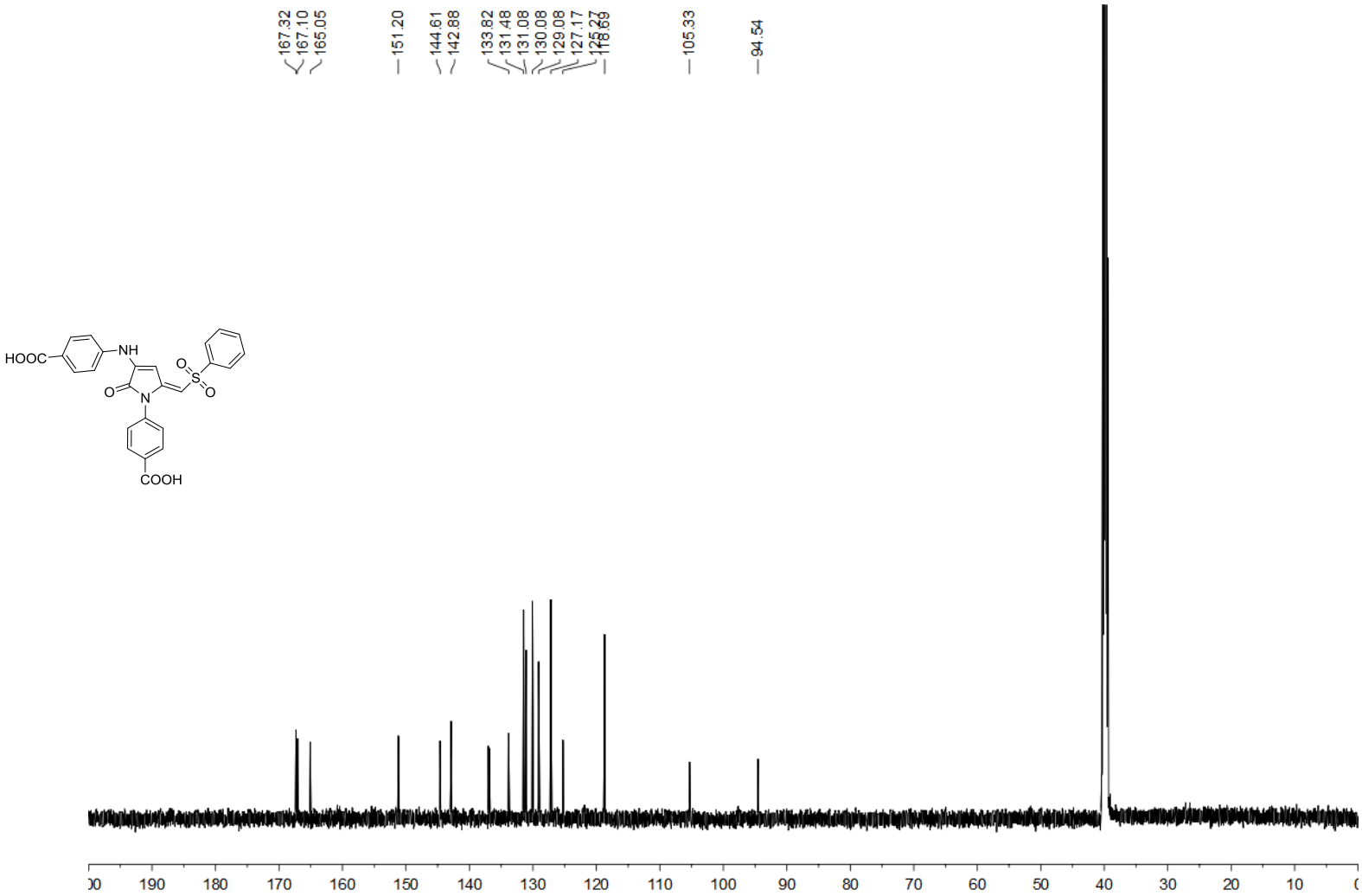


Figure S41. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **4r**

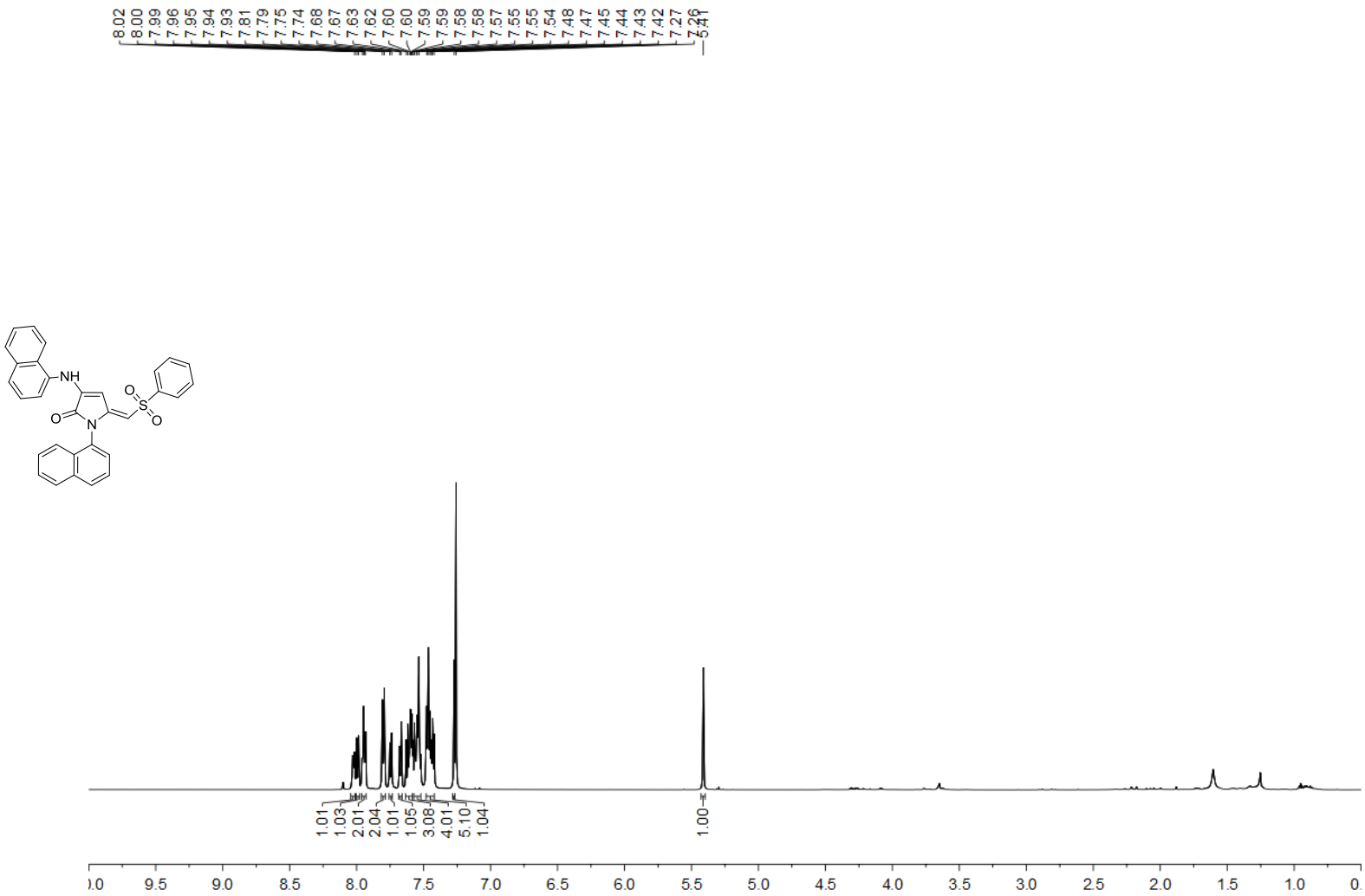


Figure S42. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **4s**

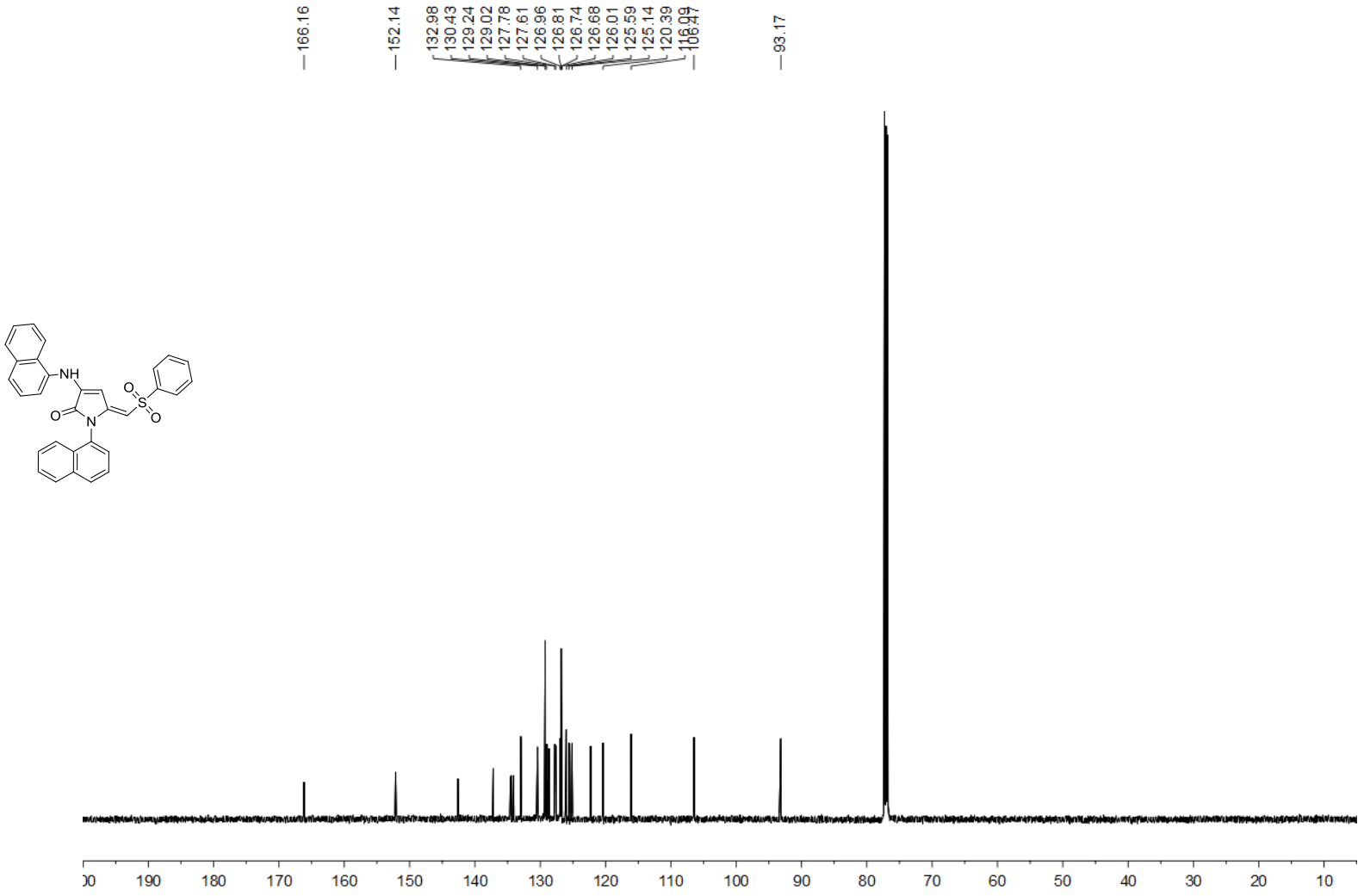


Figure S43. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **4s**

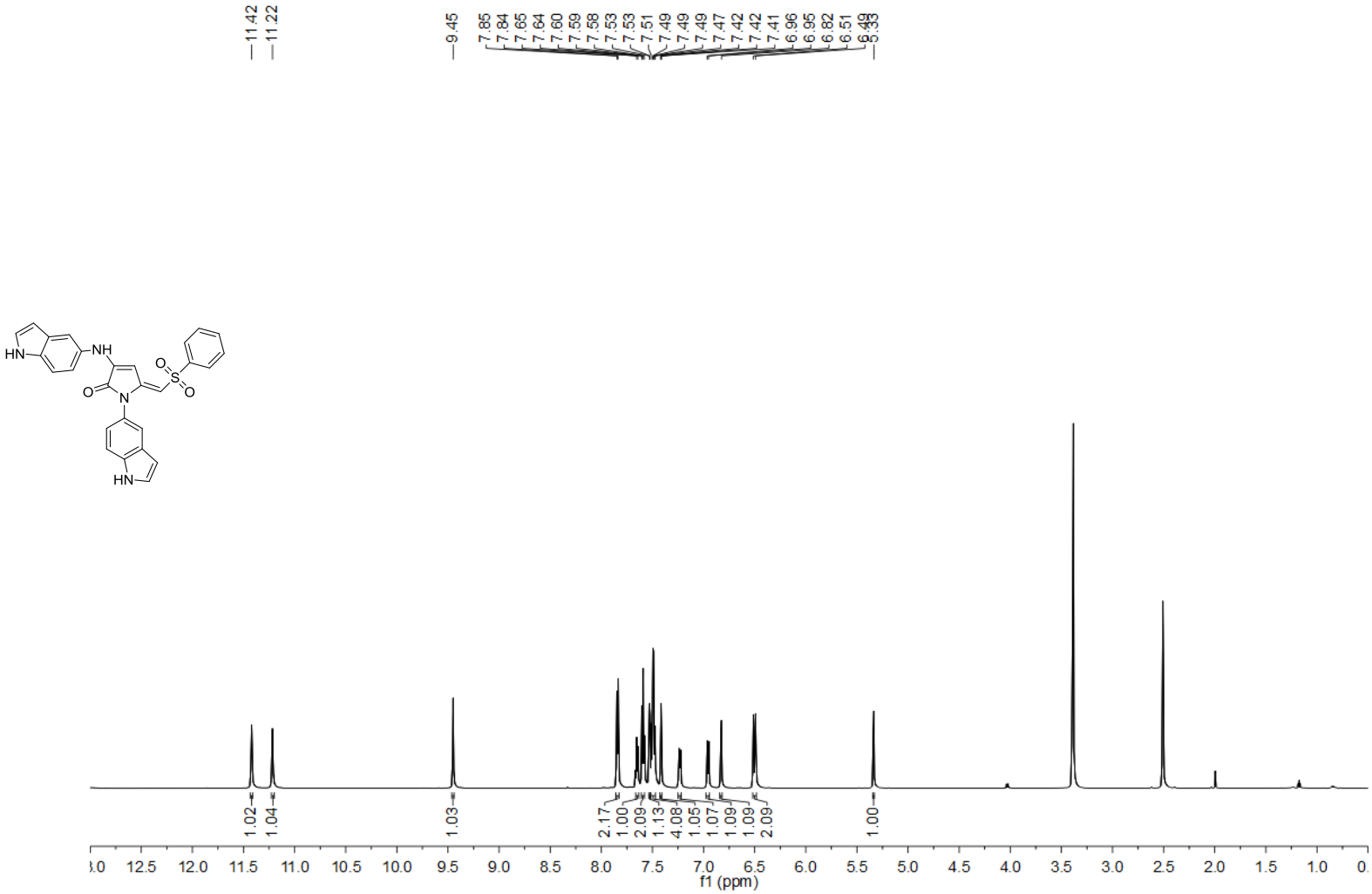


Figure S44. [1]**H NMR** (600 MHz, DMSO-d_6) spectra of compound **4t**

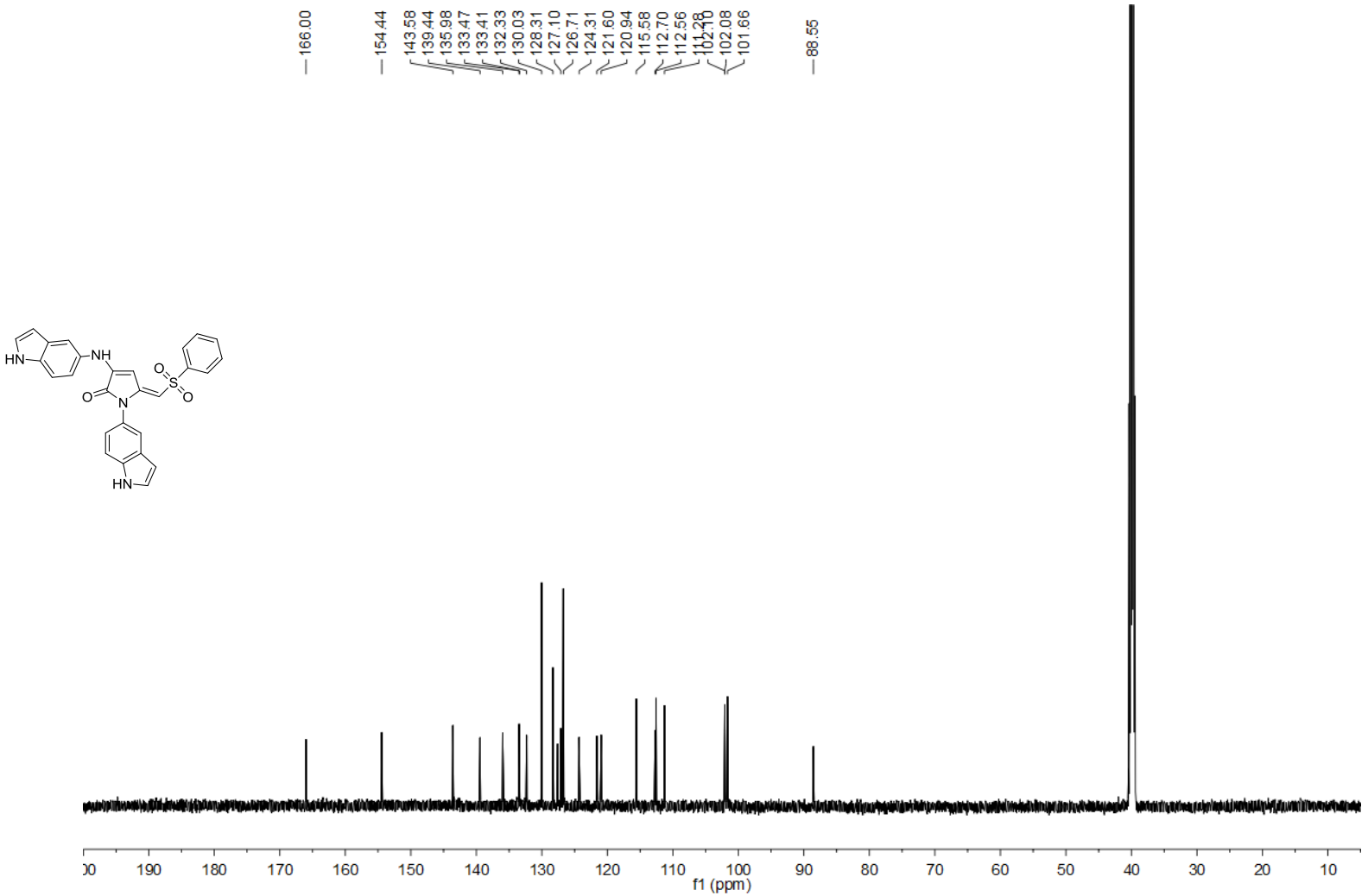


Figure S45. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **4t**

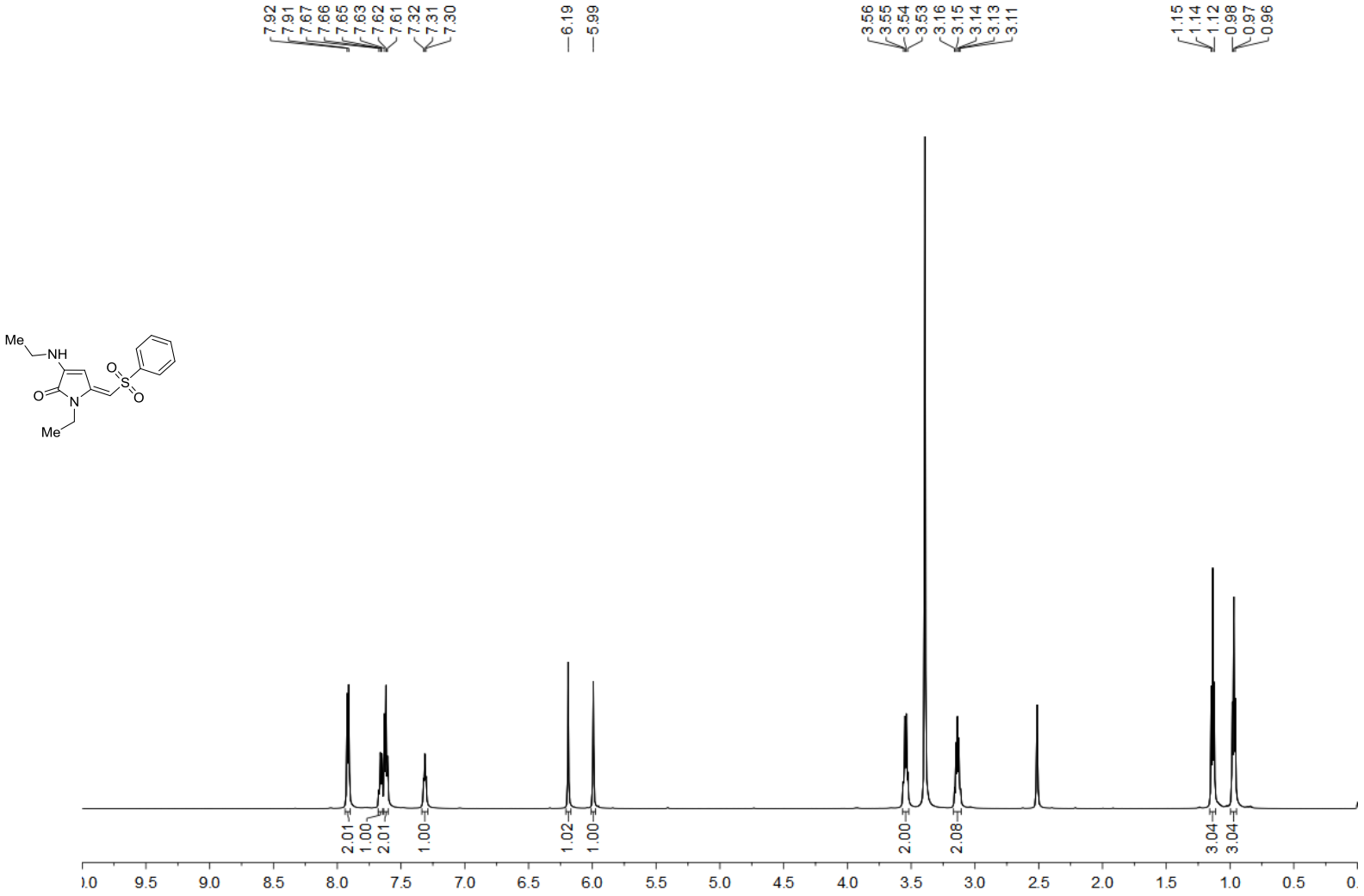


Figure S46. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **4u**

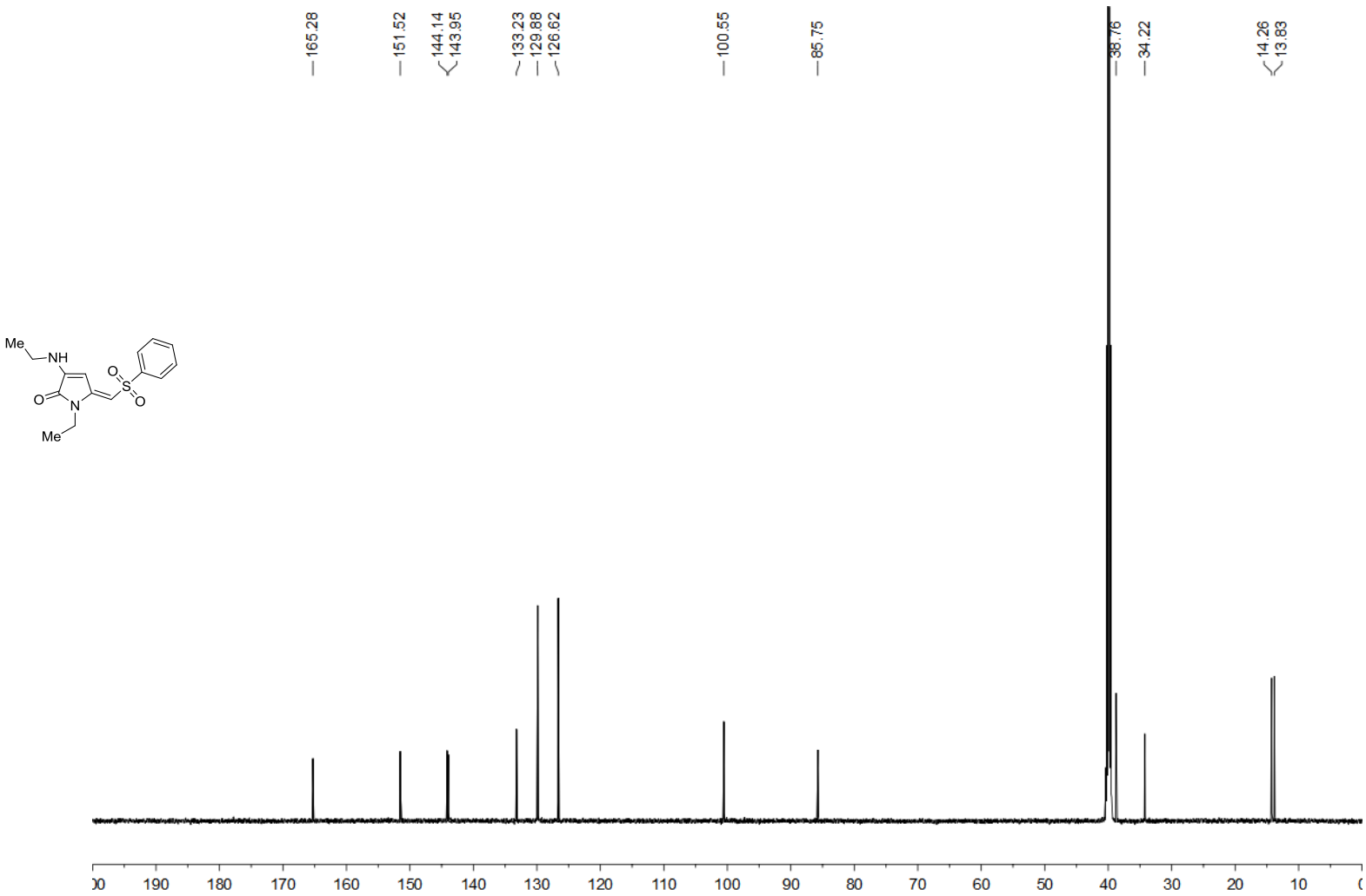


Figure S47. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **4u**

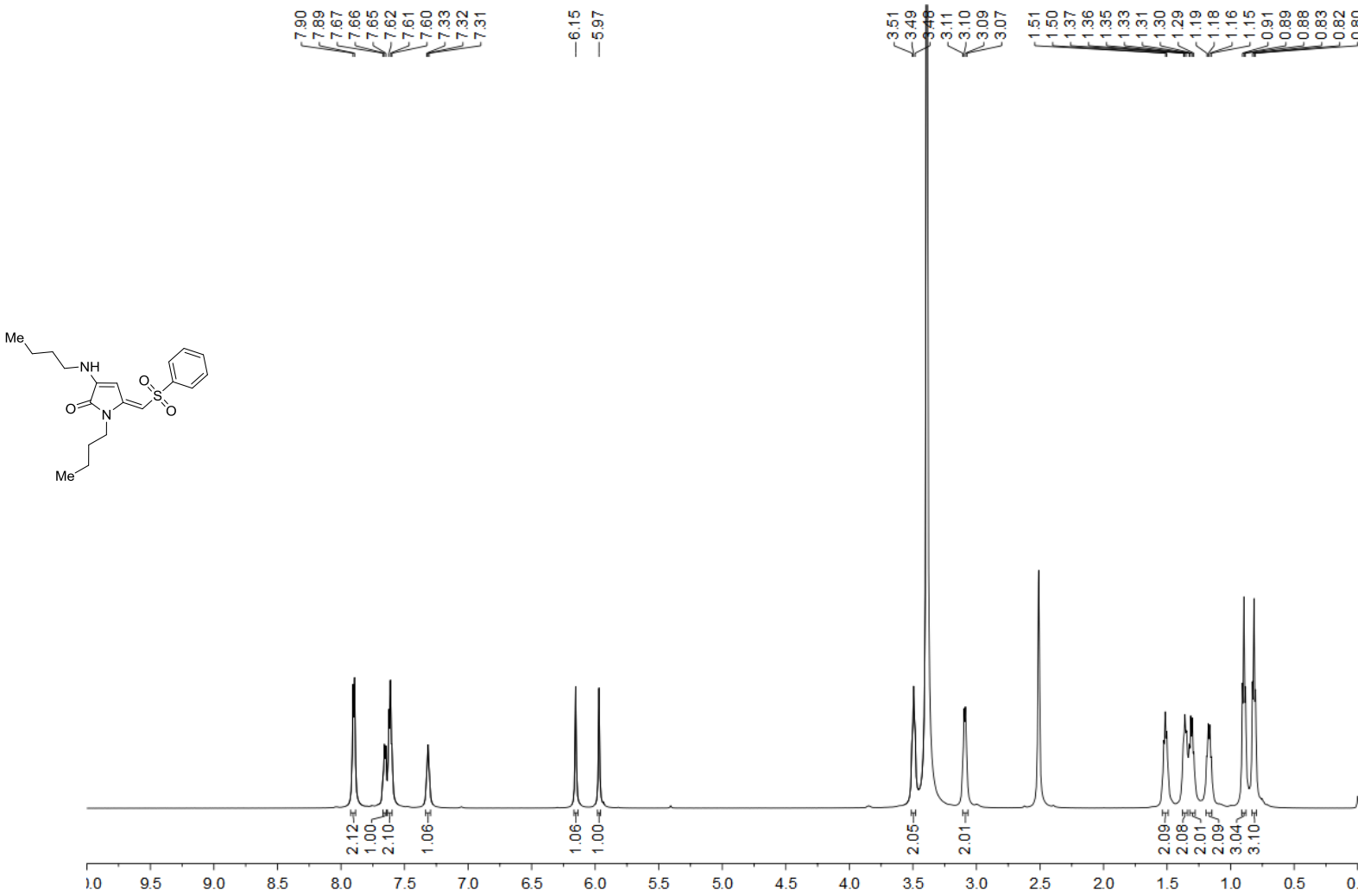


Figure S48. [1]**H NMR** (600 MHz, DMSO-d_6) spectra of compound **4v**

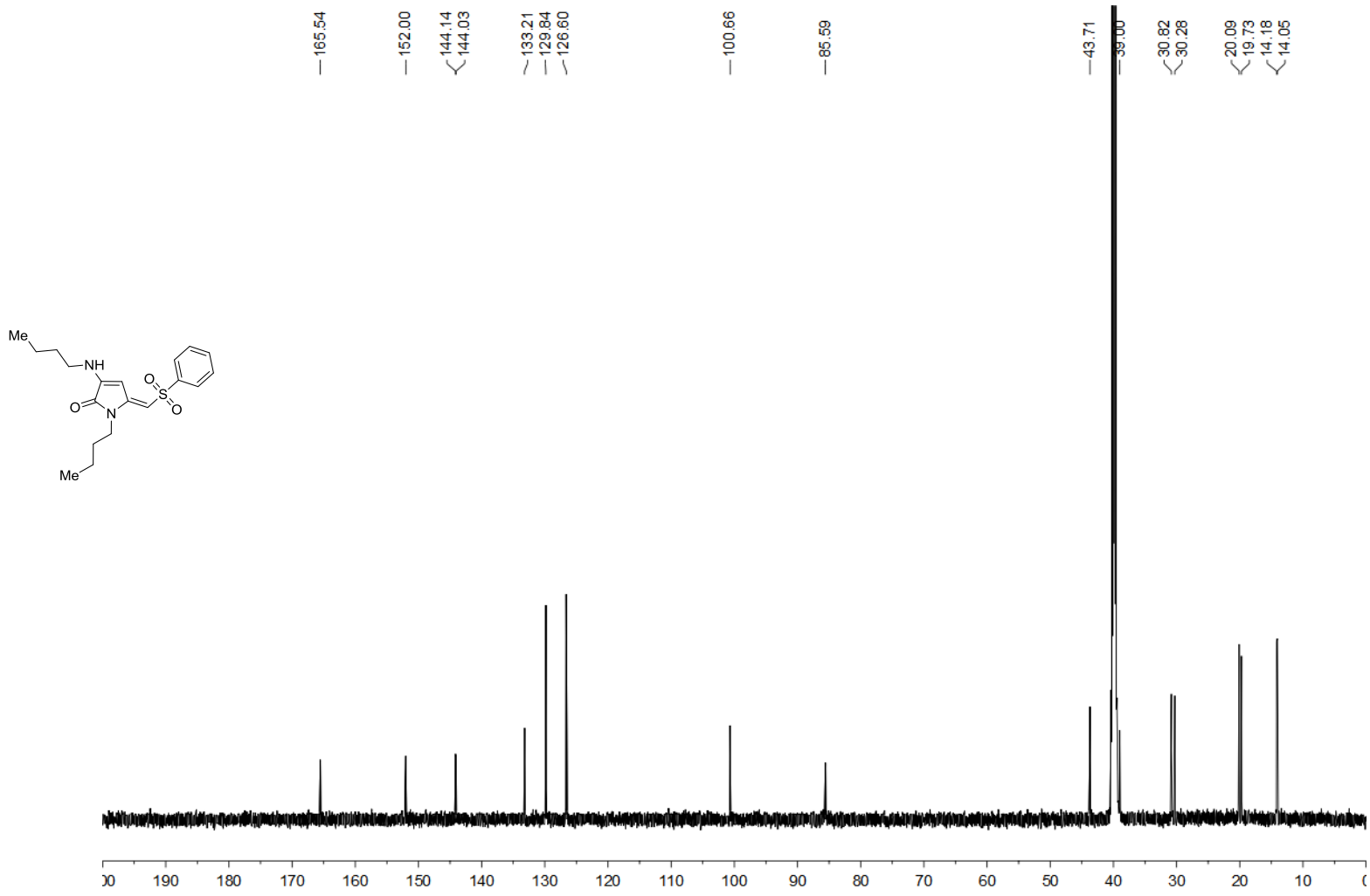


Figure S49. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **4v**

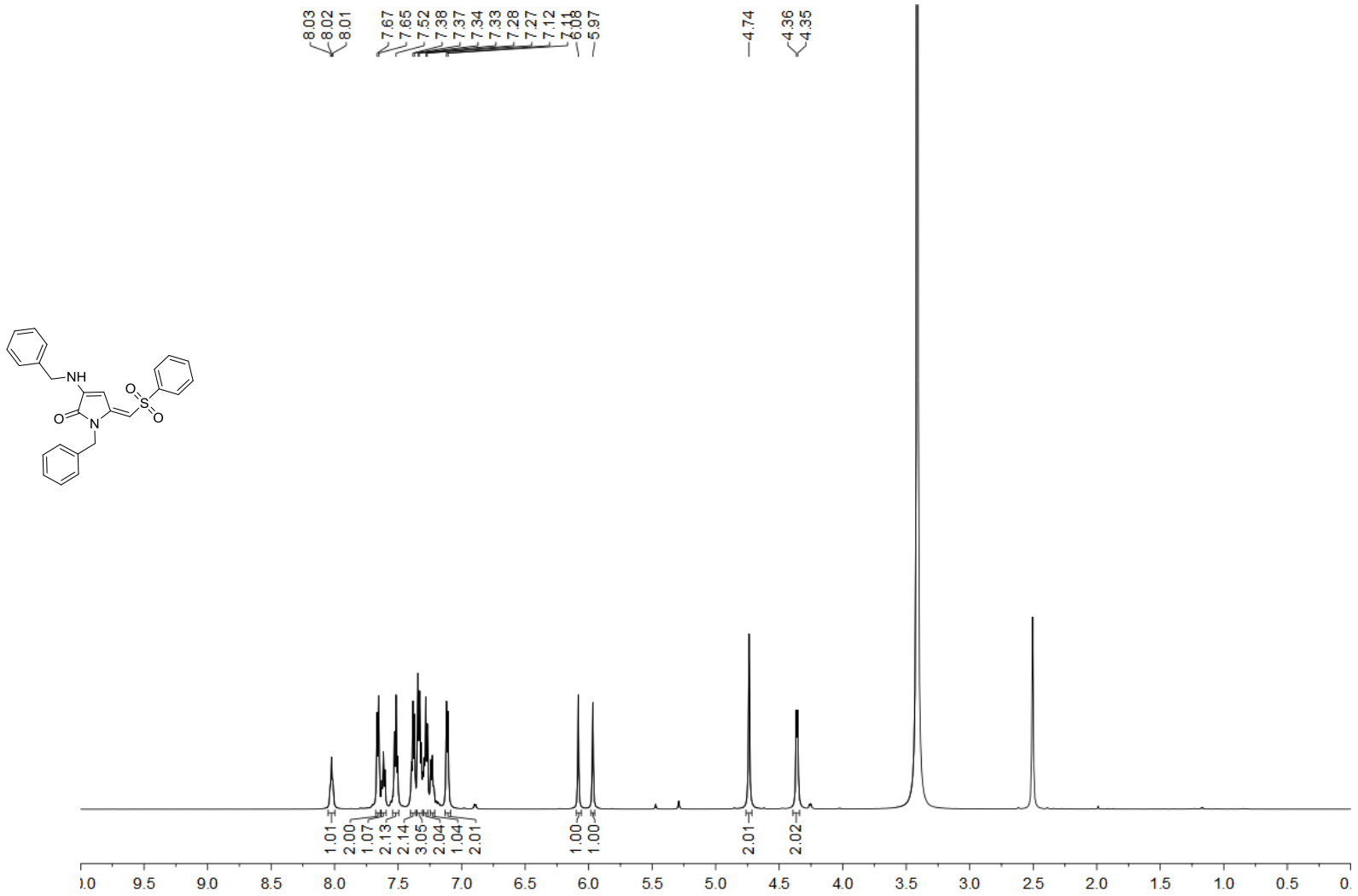


Figure S50. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **4w**

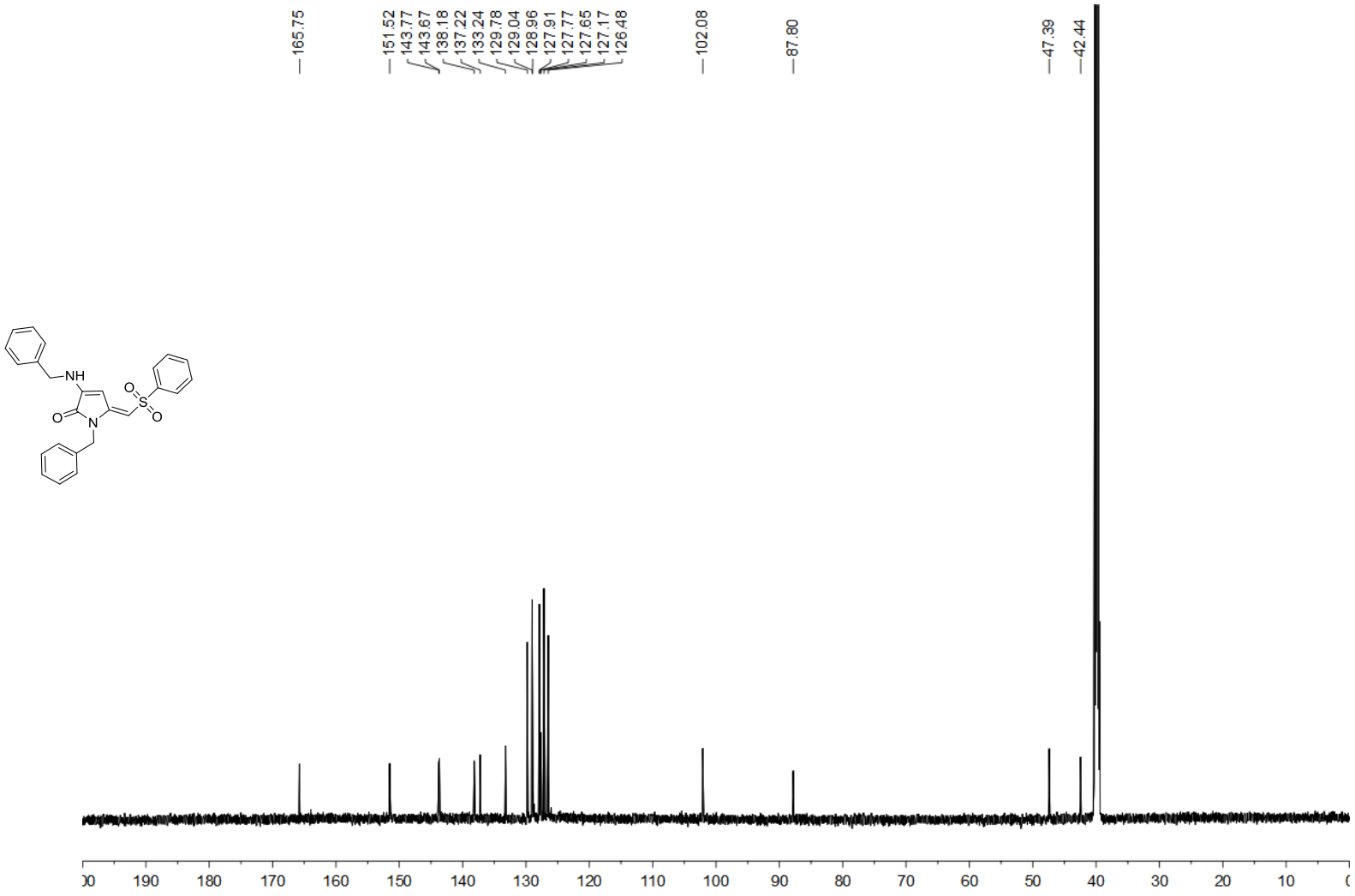


Figure S51. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **4w**

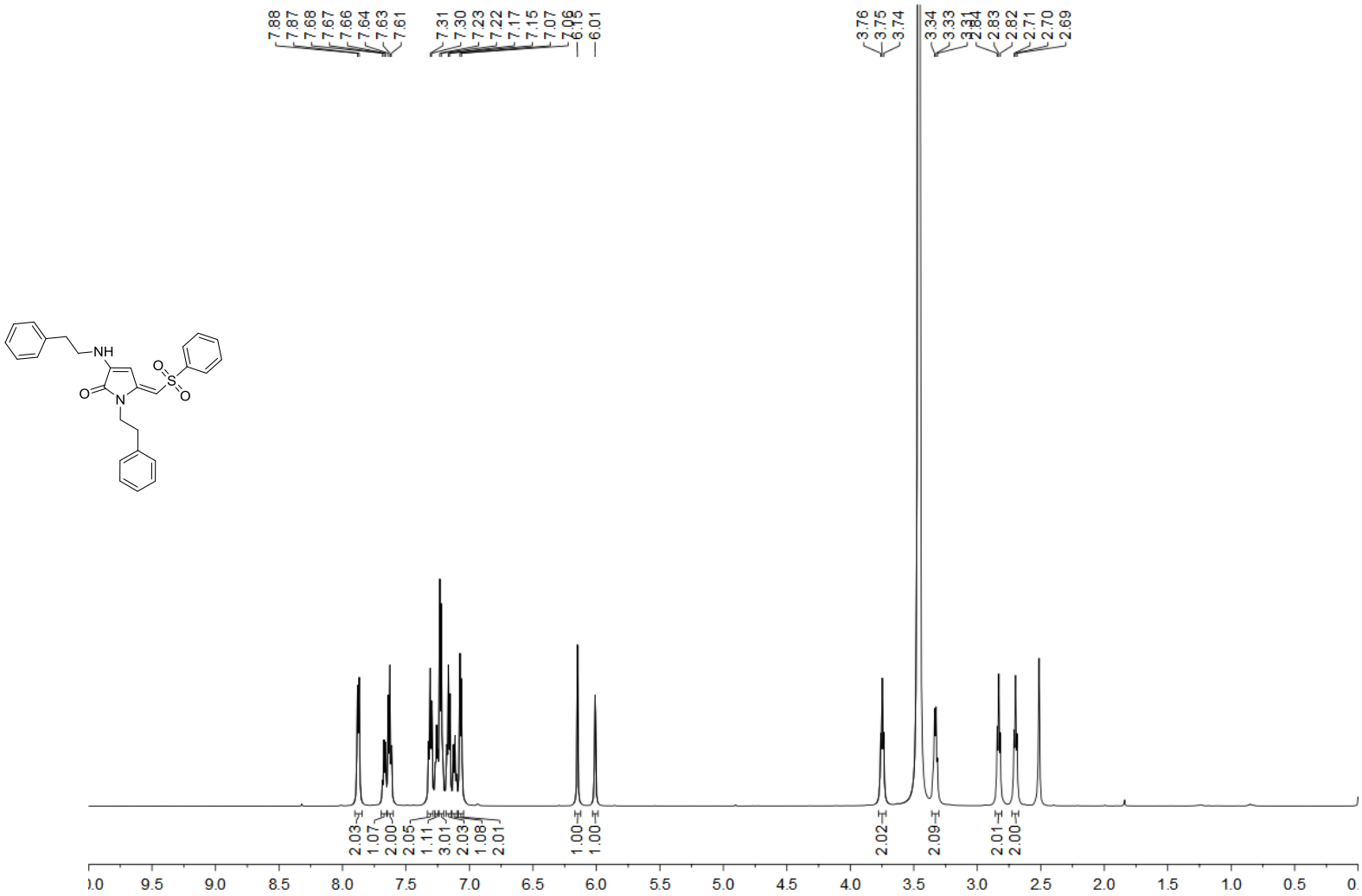


Figure S52. [1]**H NMR** (600 MHz, DMSO-d_6) spectra of compound **4x**

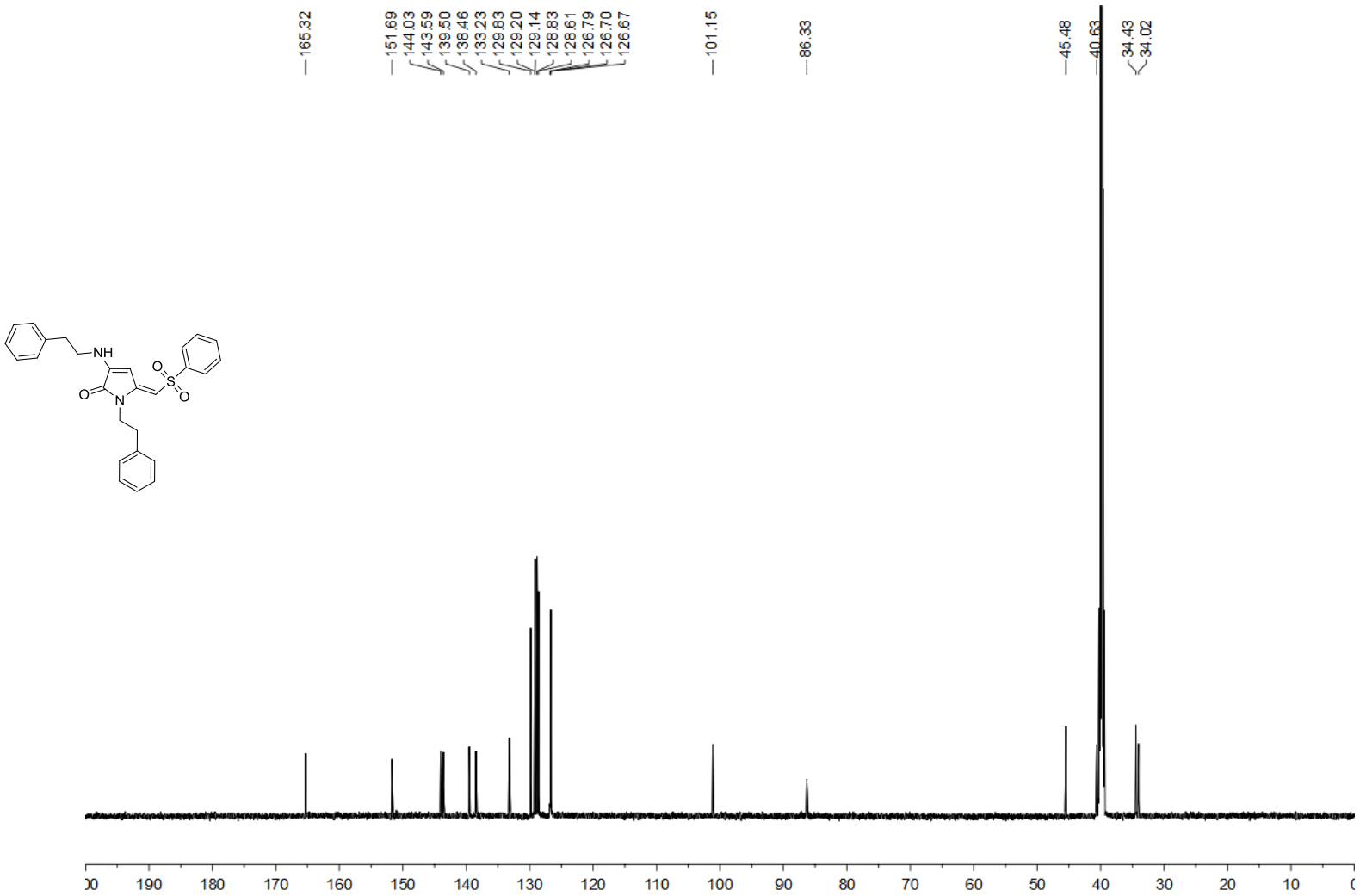


Figure S53. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **4x**

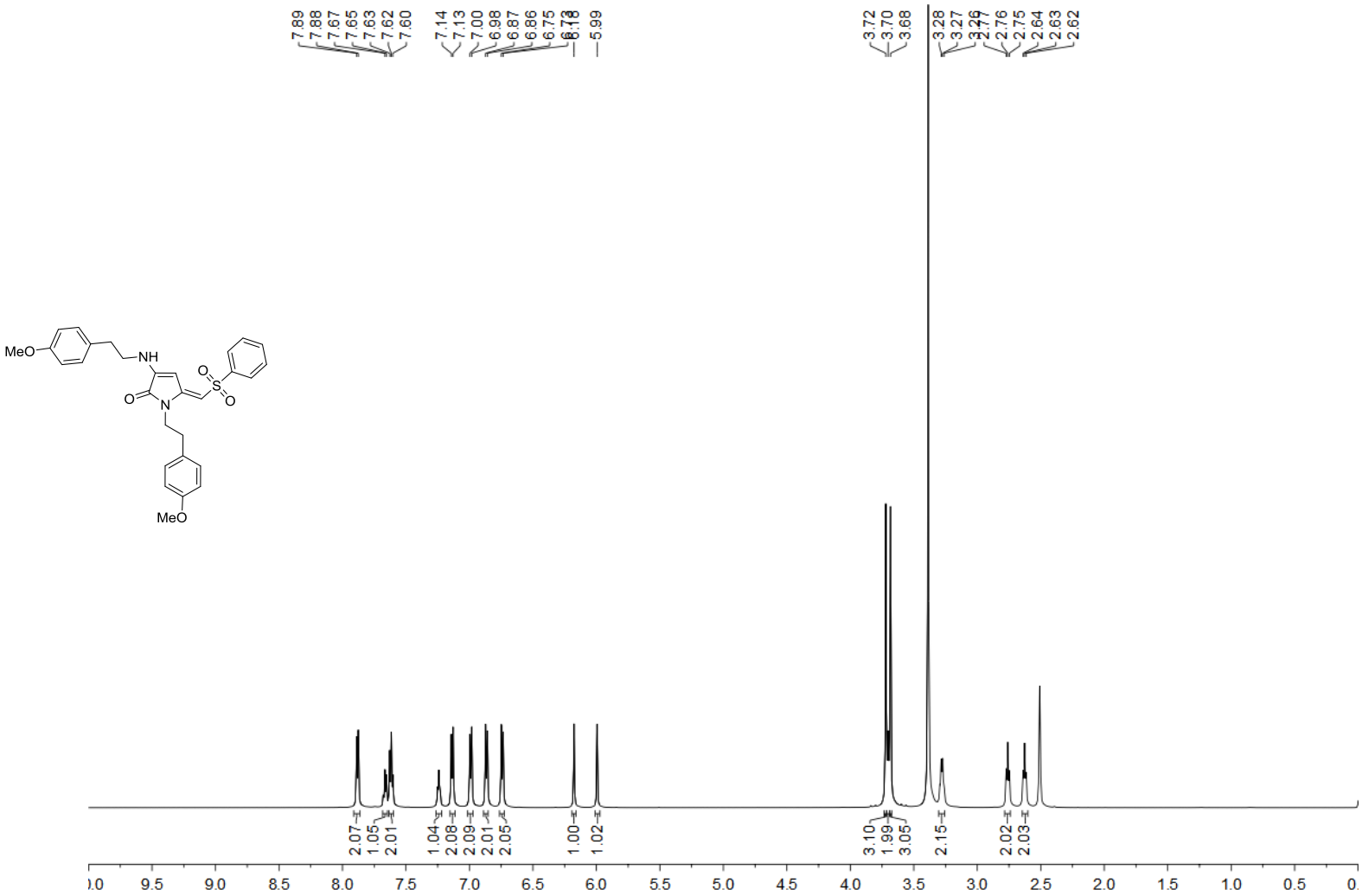


Figure S54. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **4y**

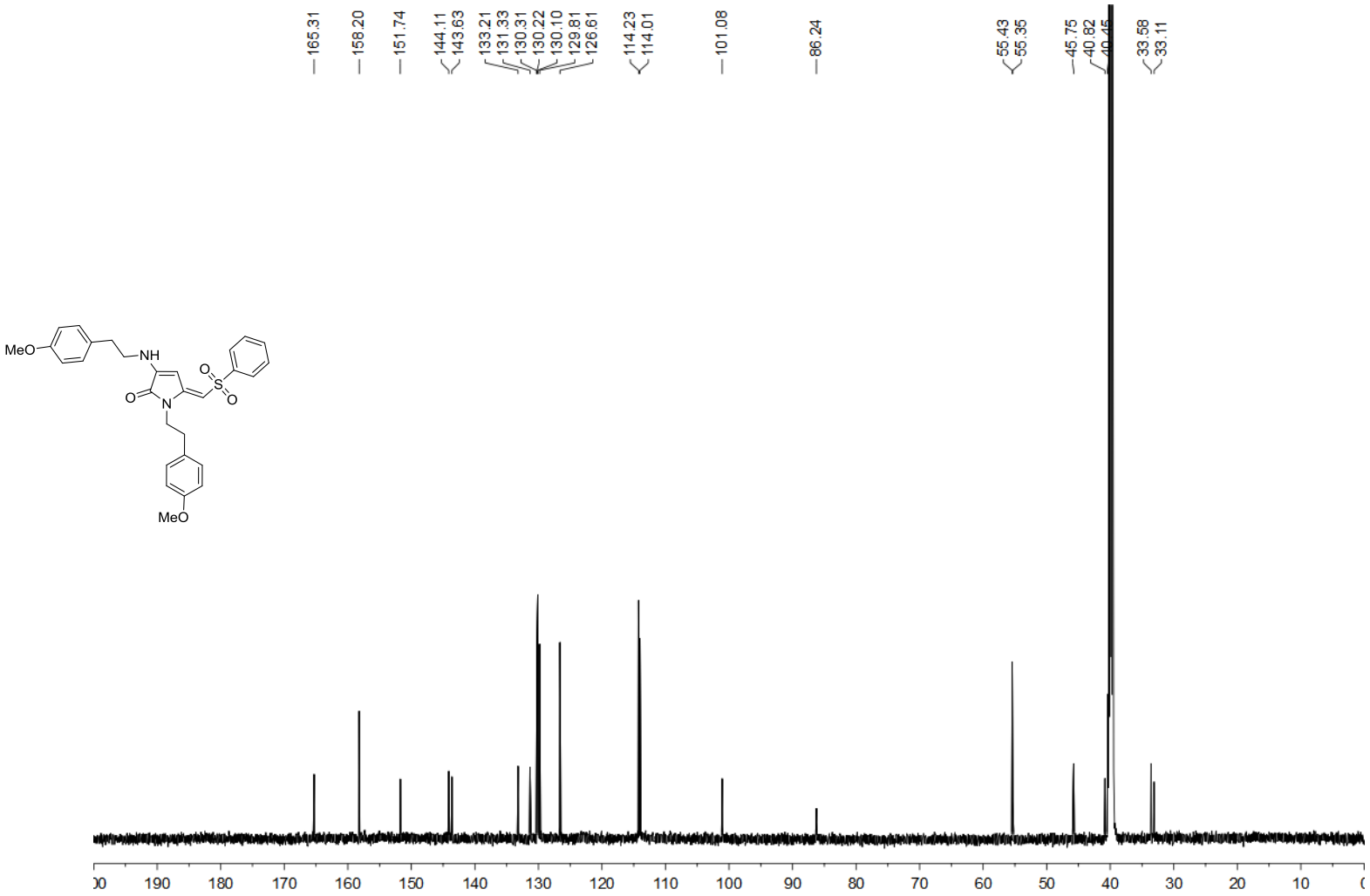


Figure S55. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **4y**

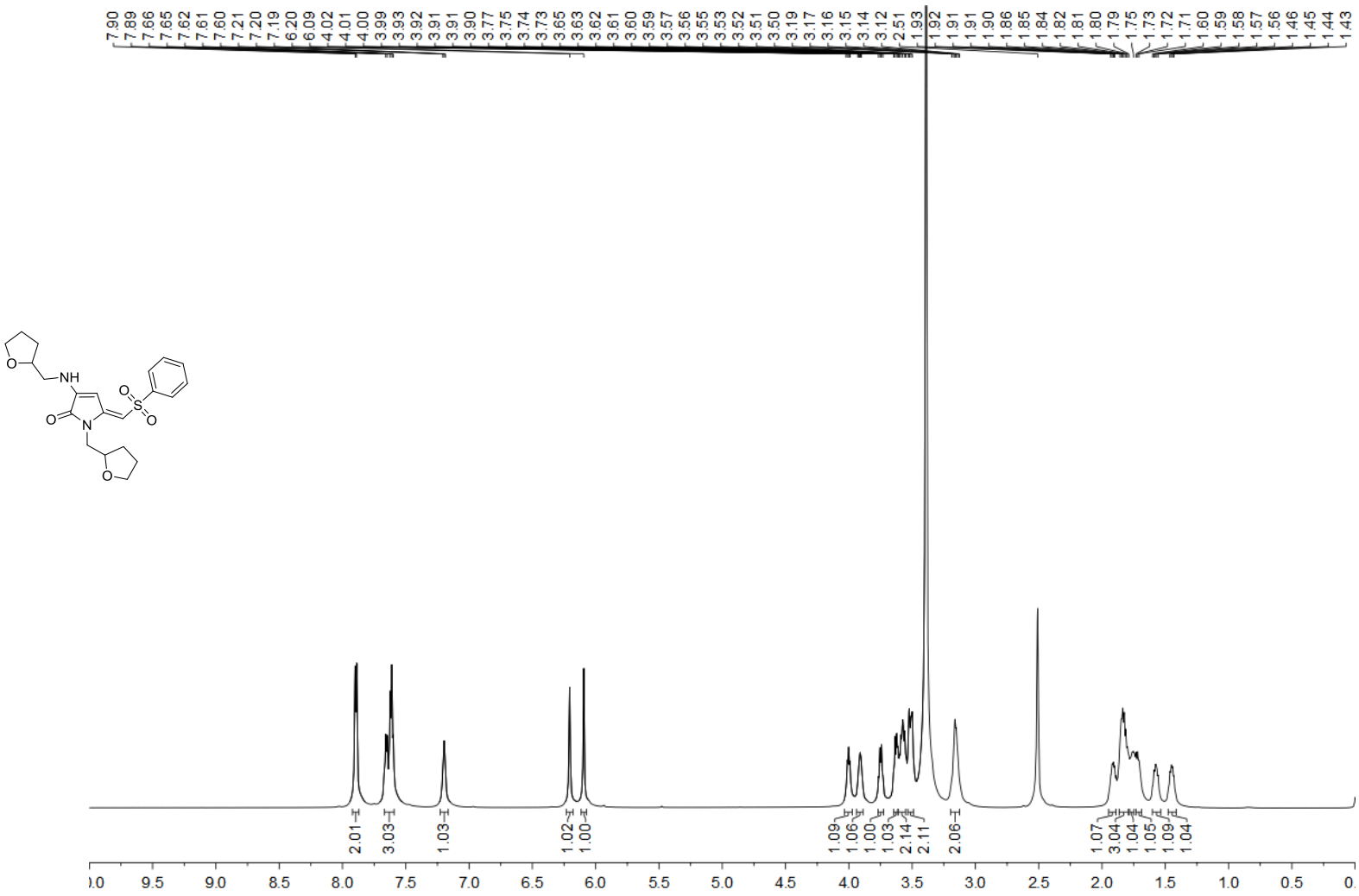


Figure S56. **^{1}H NMR** (600 MHz, DMSO-d_6) spectra of compound **4z**

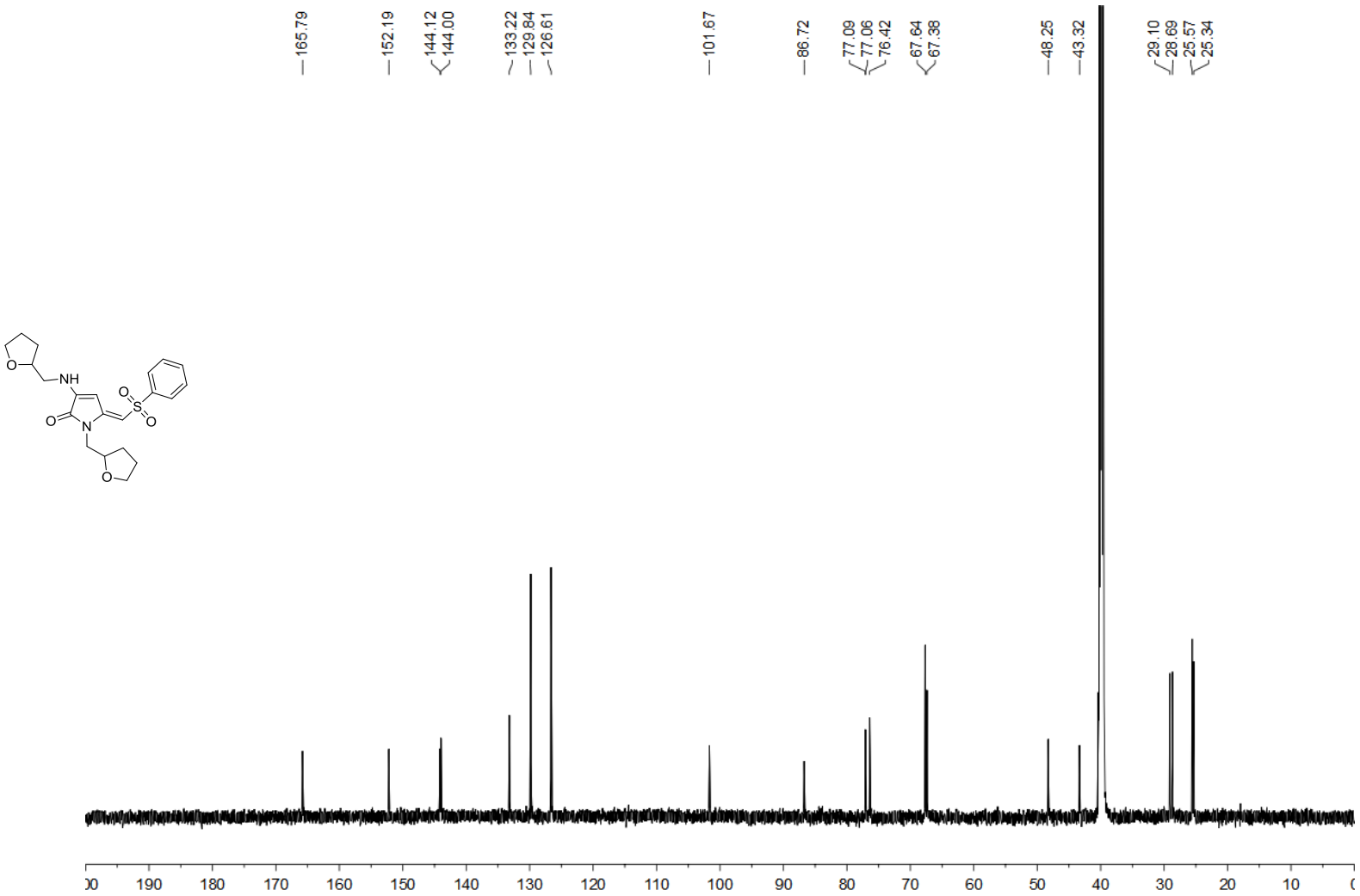


Figure S57. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **4z**

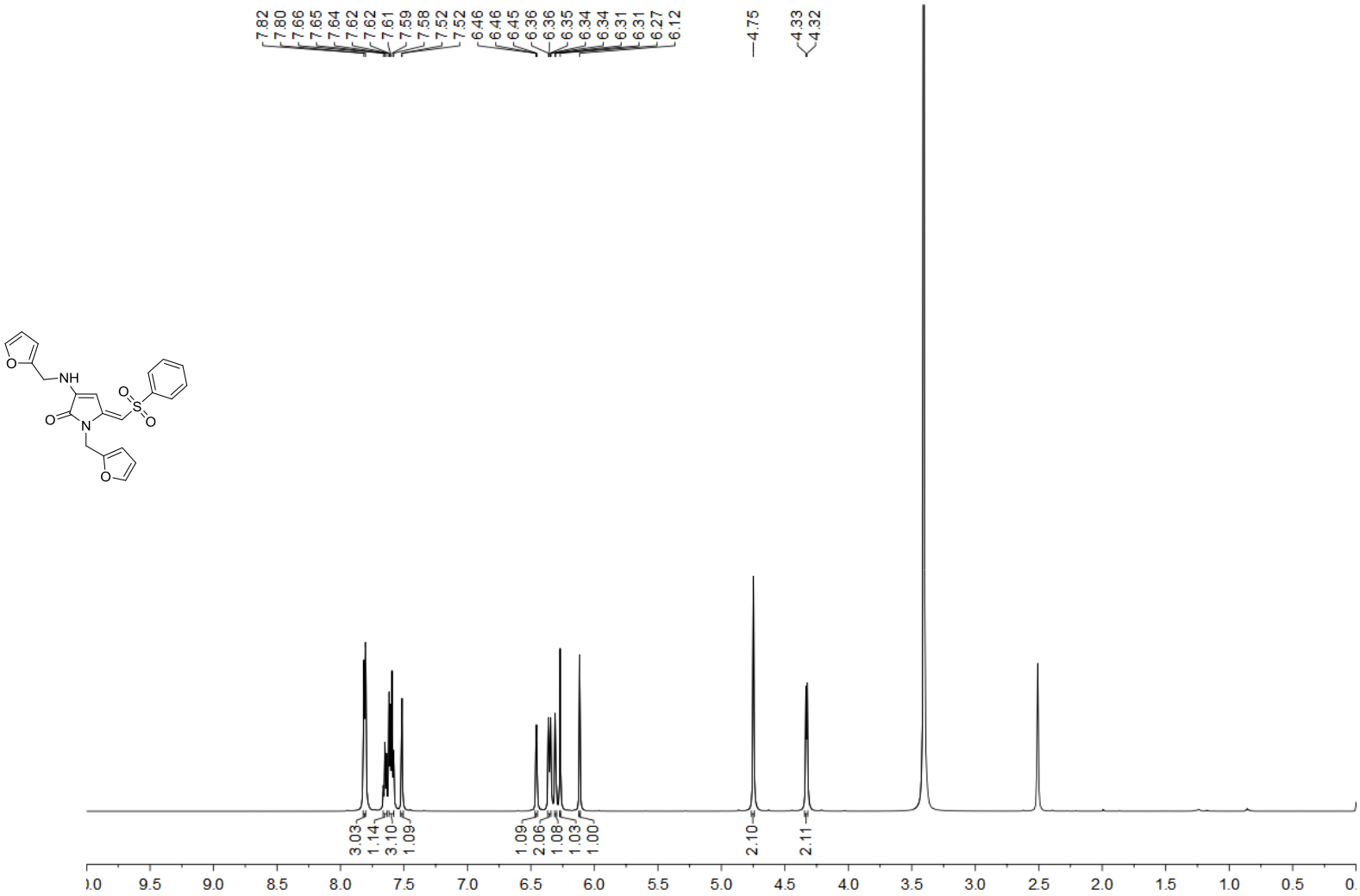


Figure S58. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **4a'**

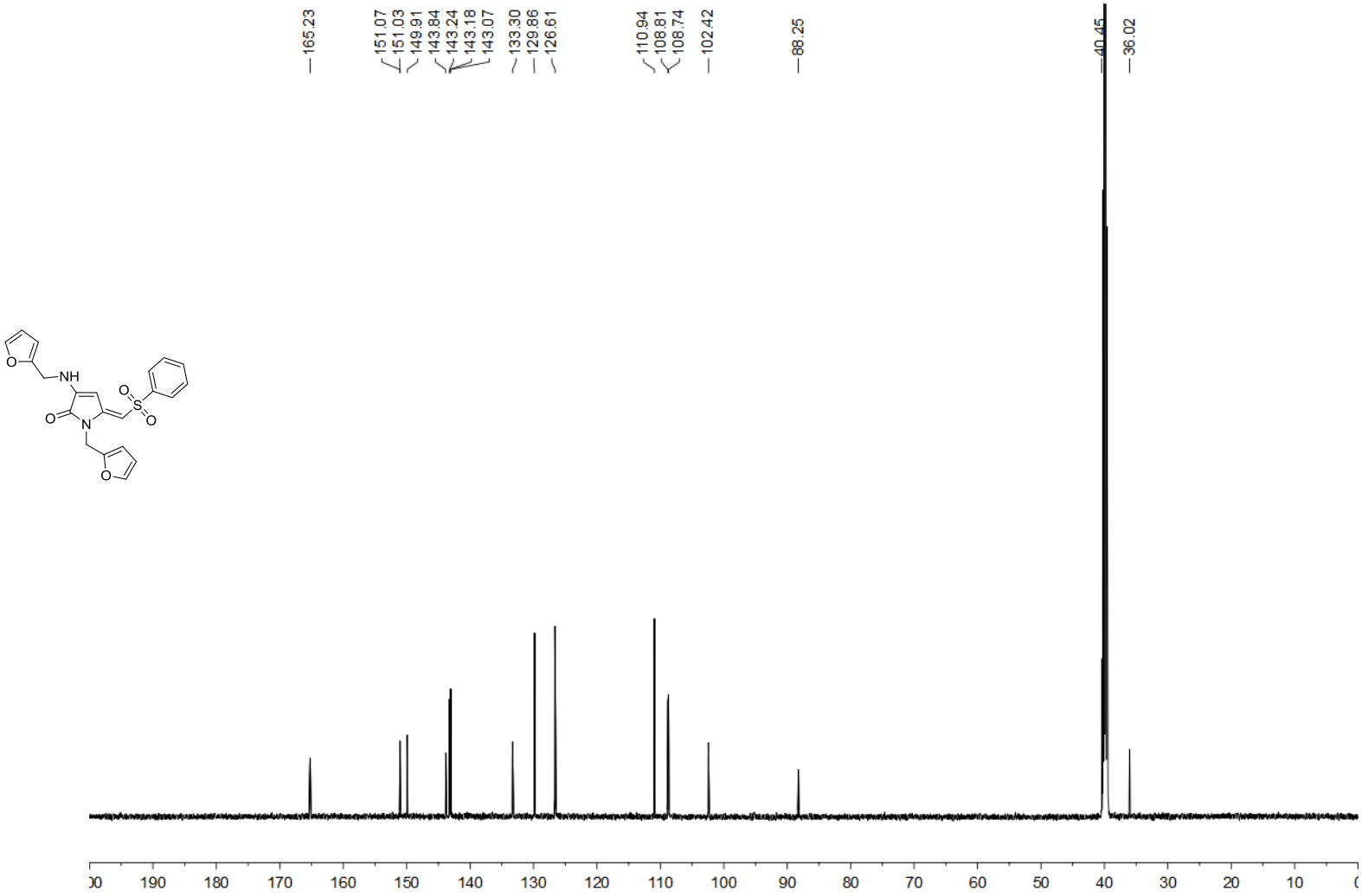


Figure S59. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **4a'**

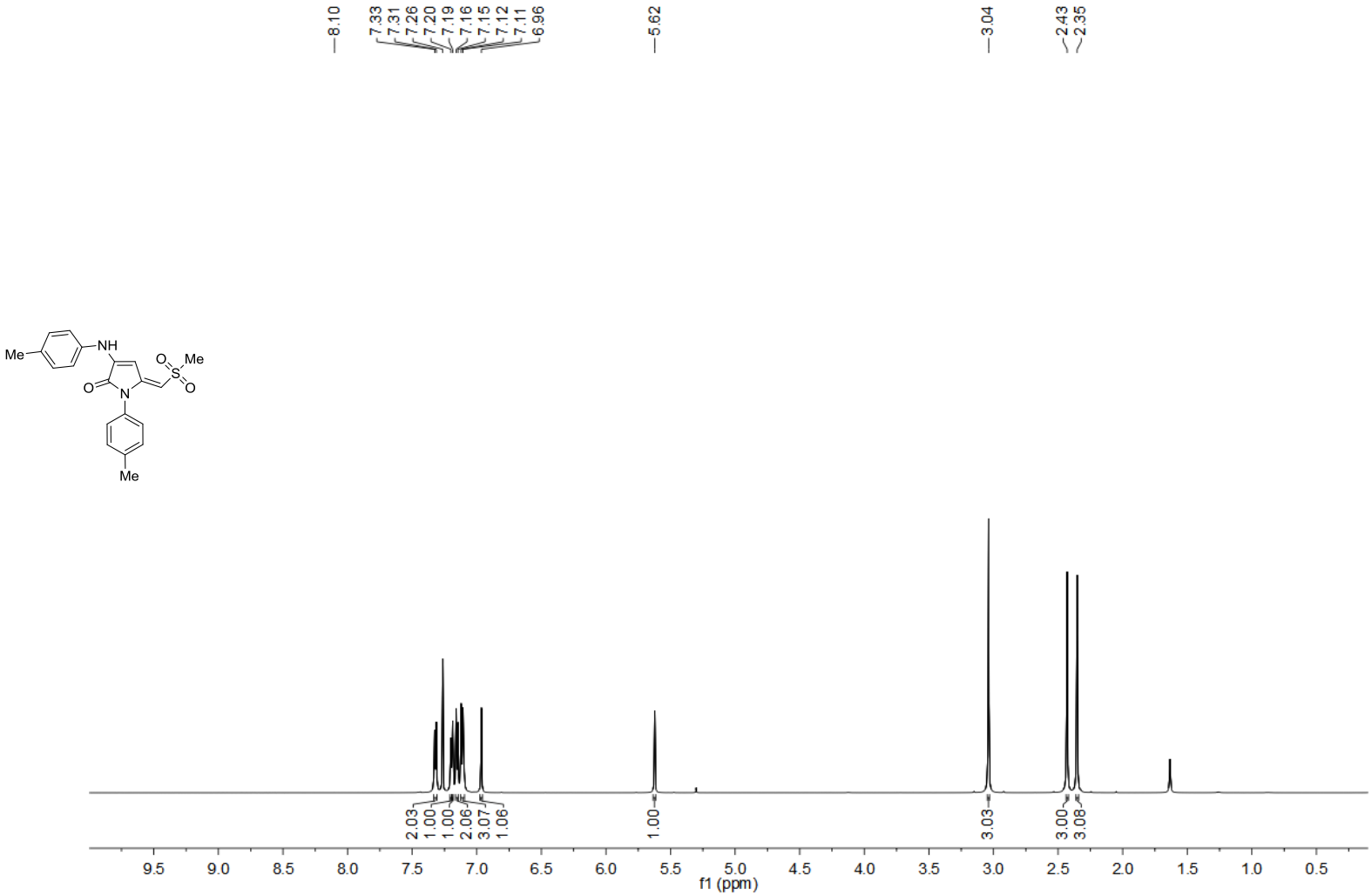


Figure S60. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **5a**

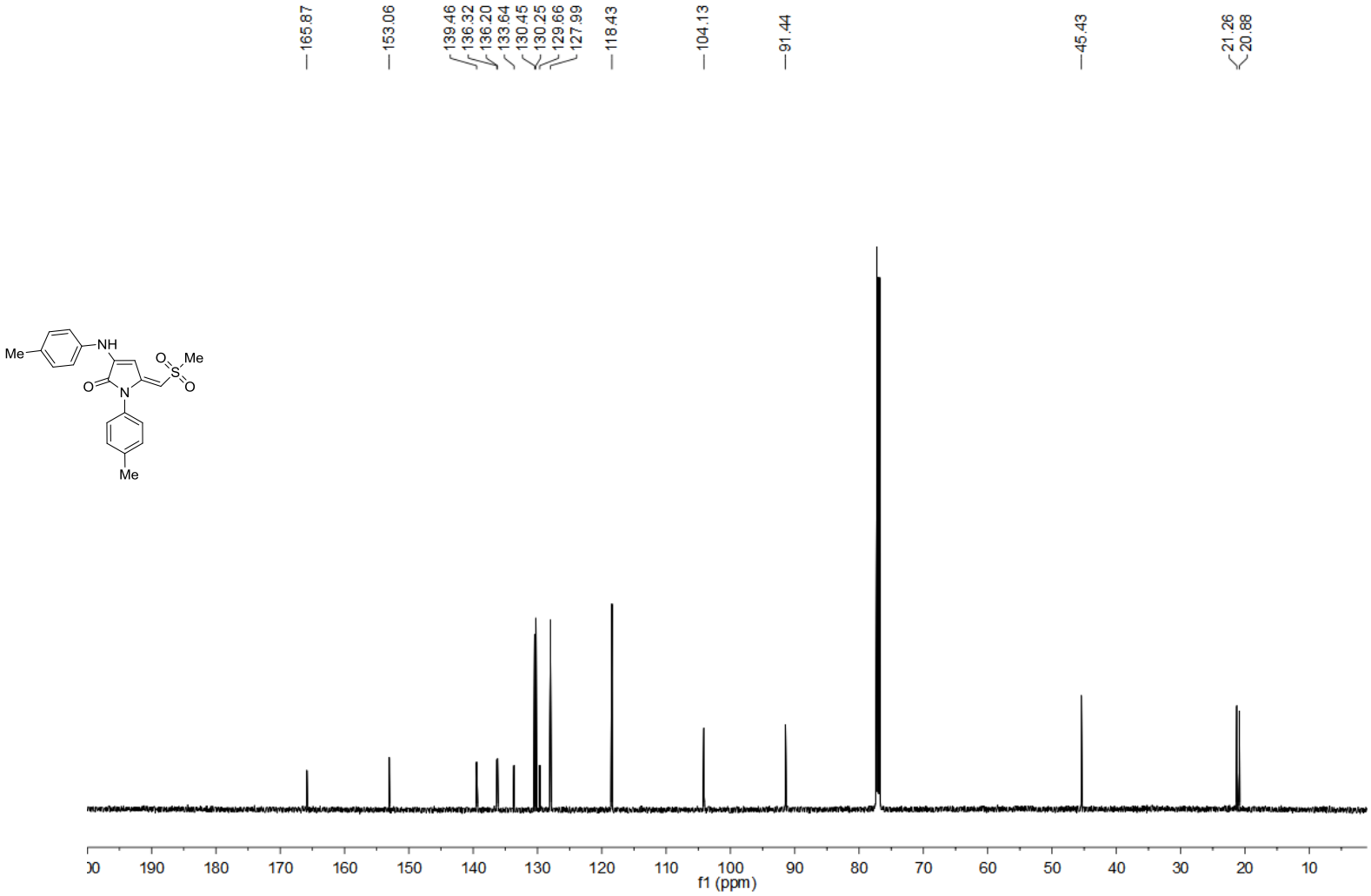


Figure S61. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **5a**

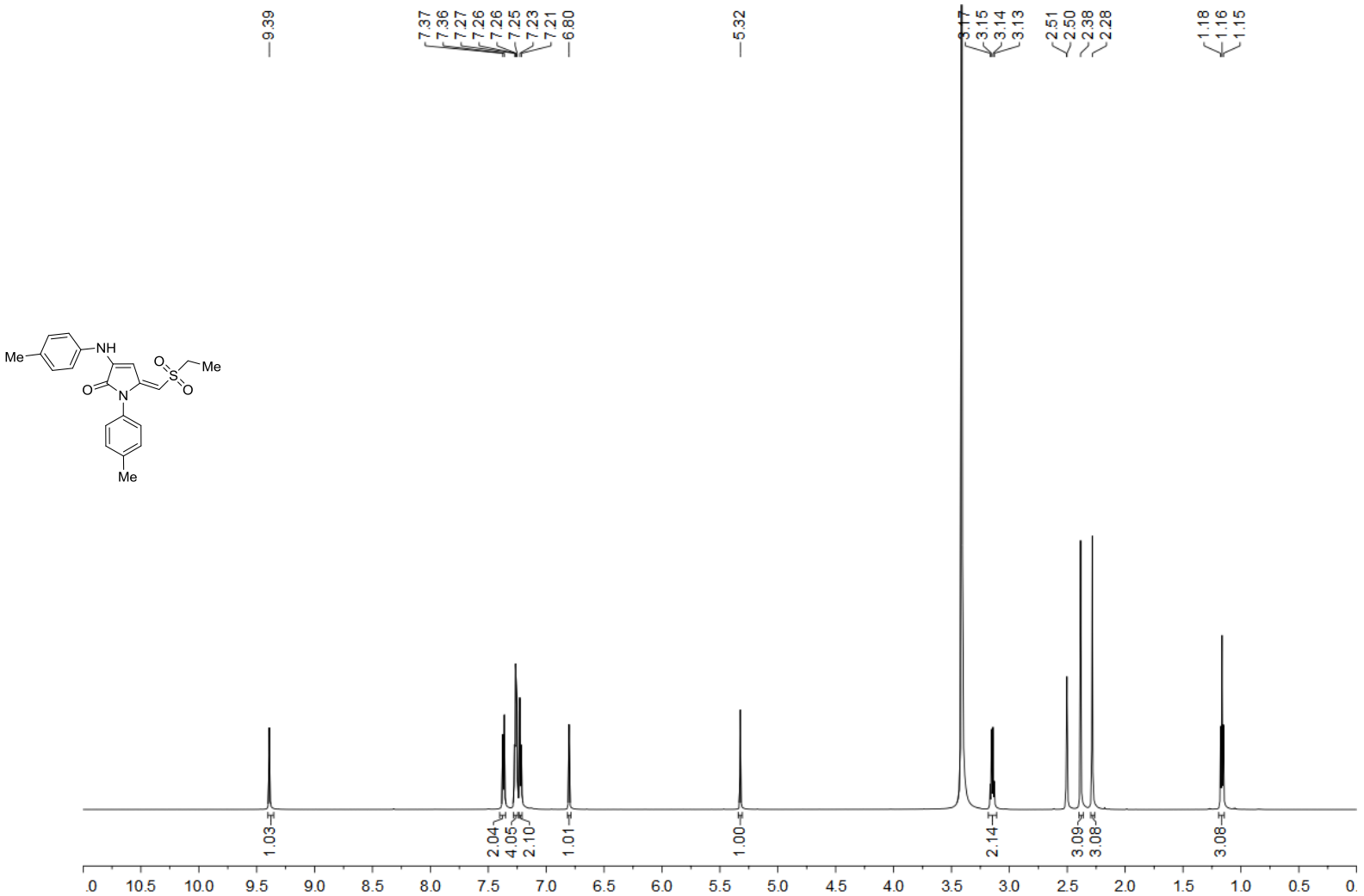


Figure S62. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **5b**

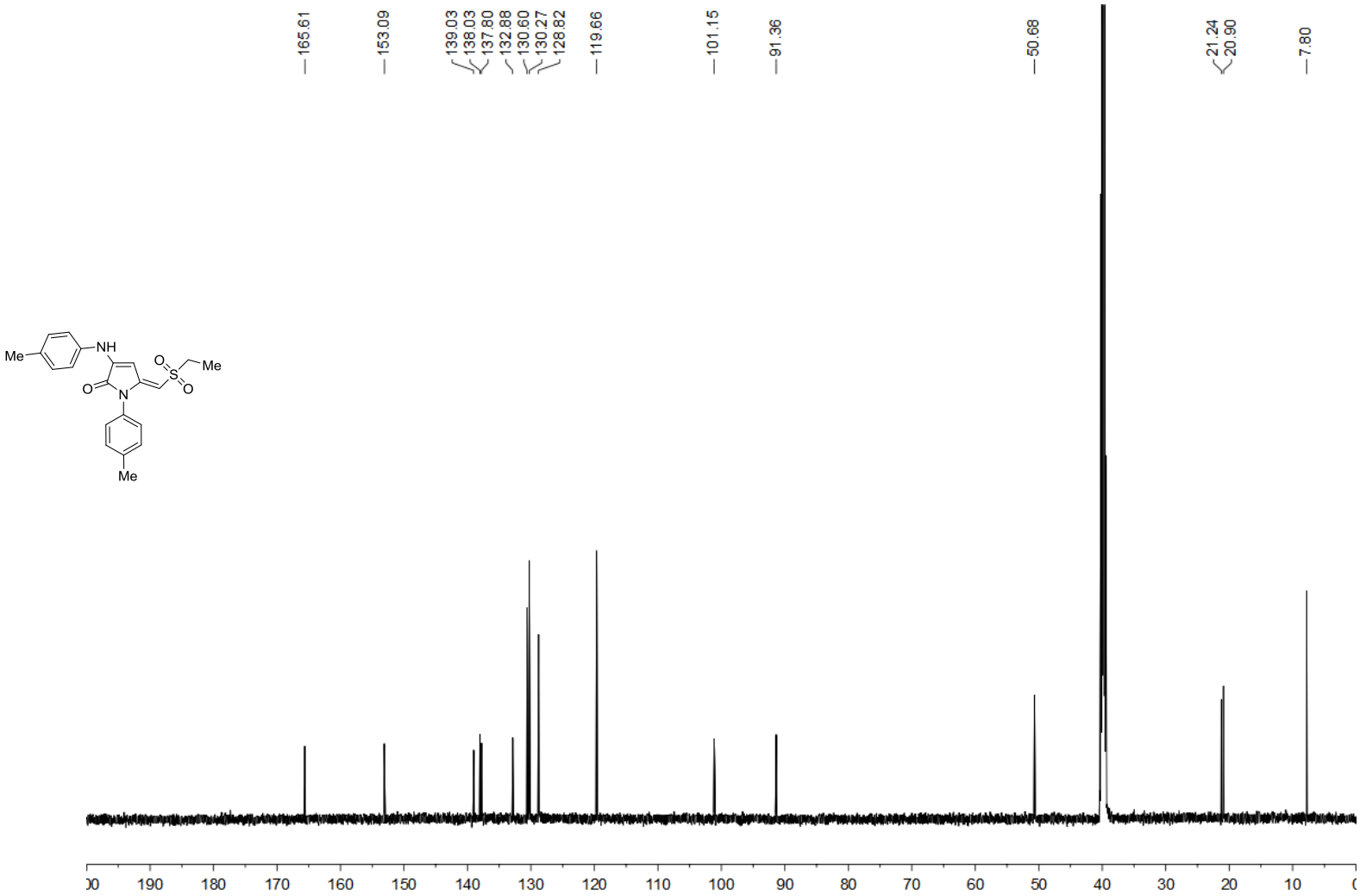


Figure S63. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **5b**

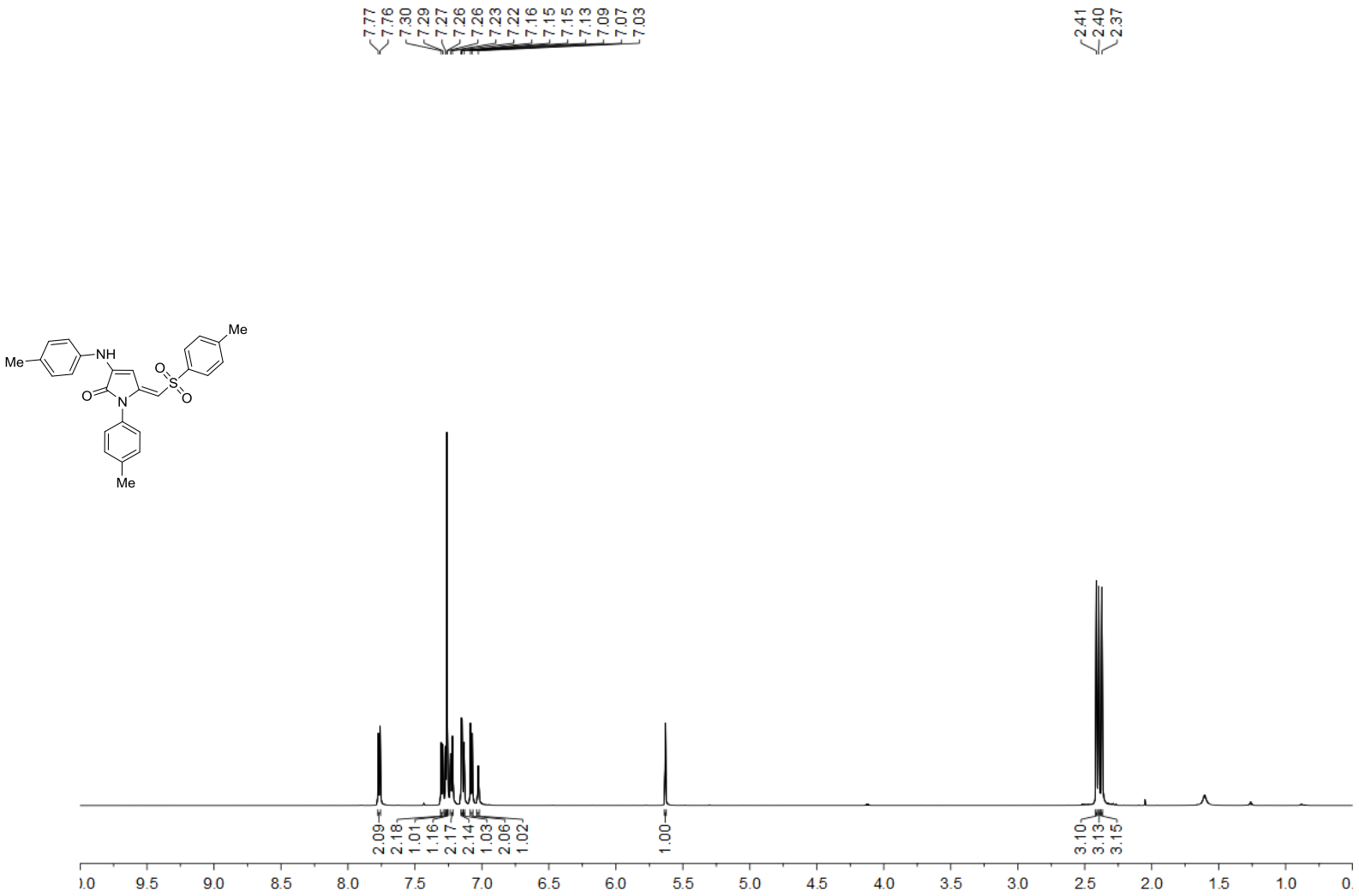


Figure S64. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **5c**

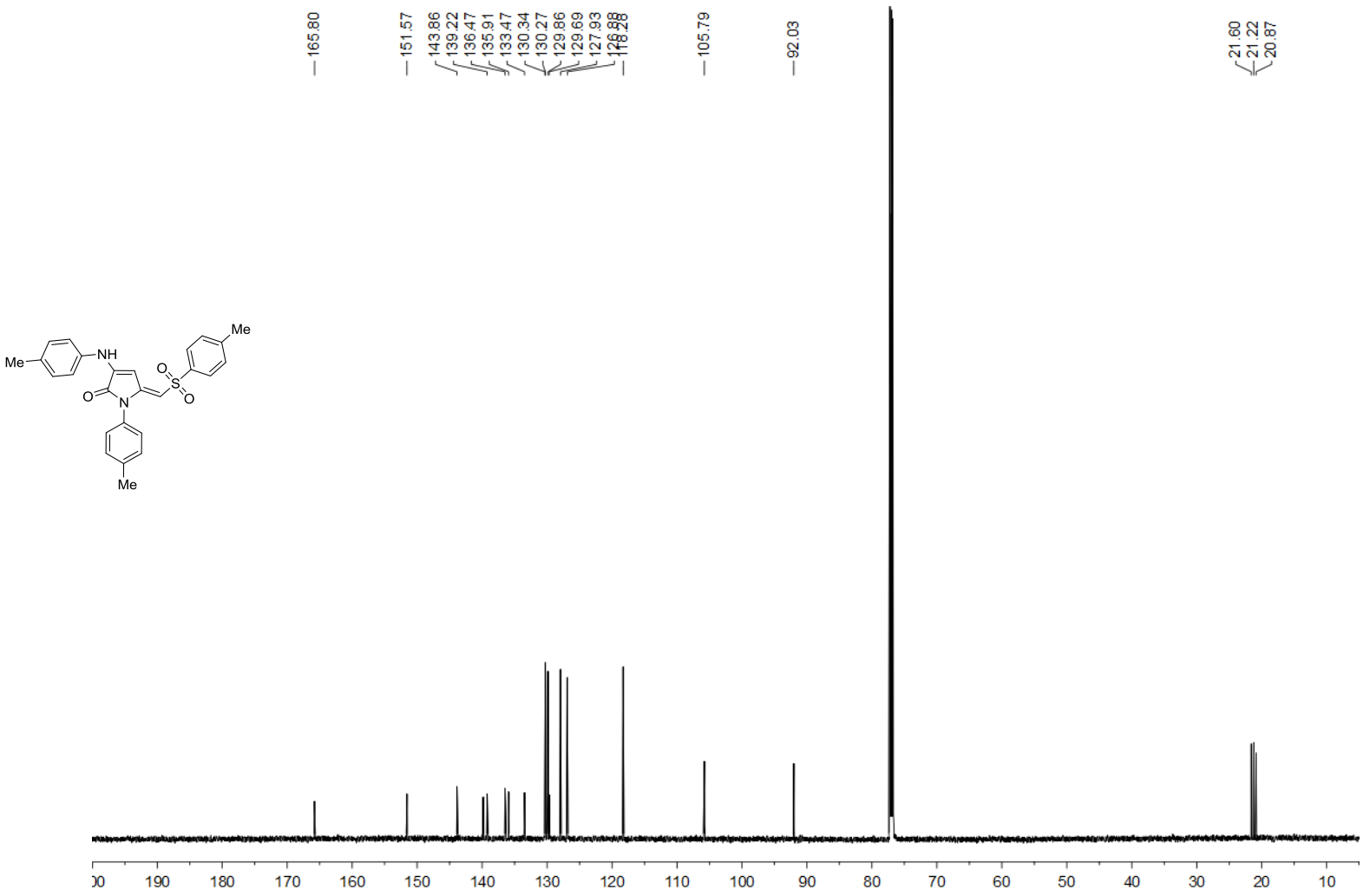


Figure S65. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **5c**

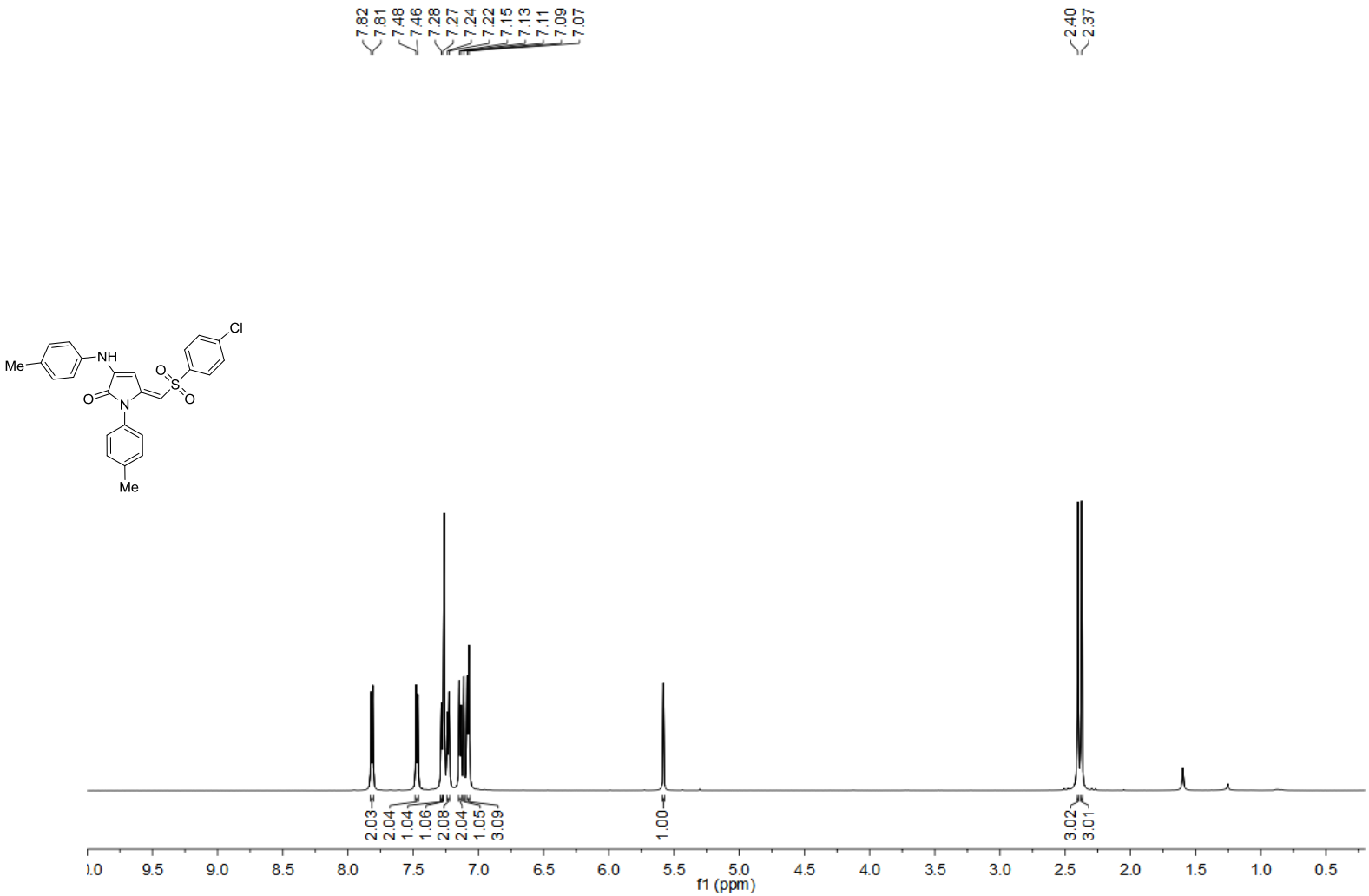


Figure S66. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **5d**

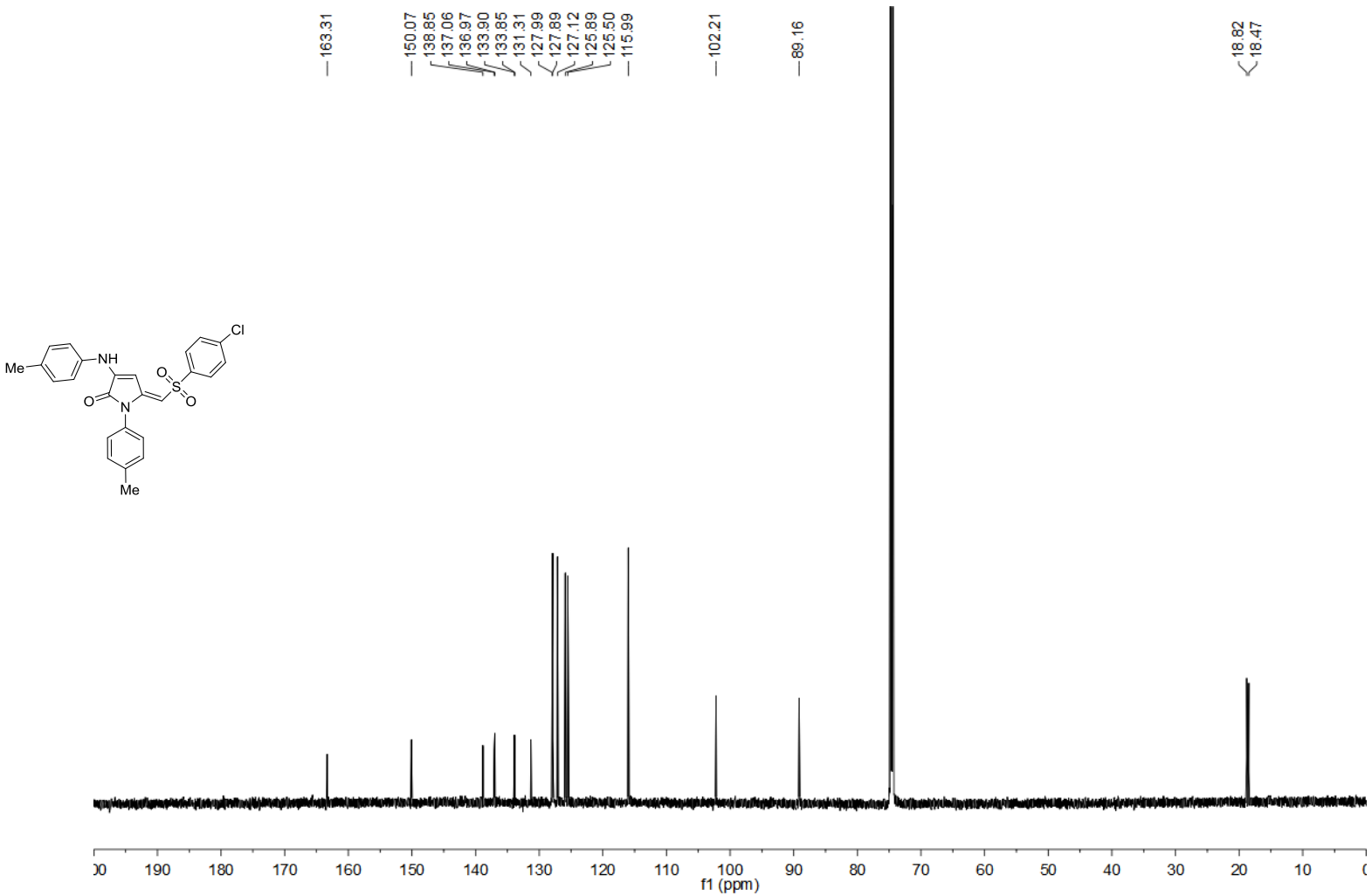


Figure S67. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **5d**

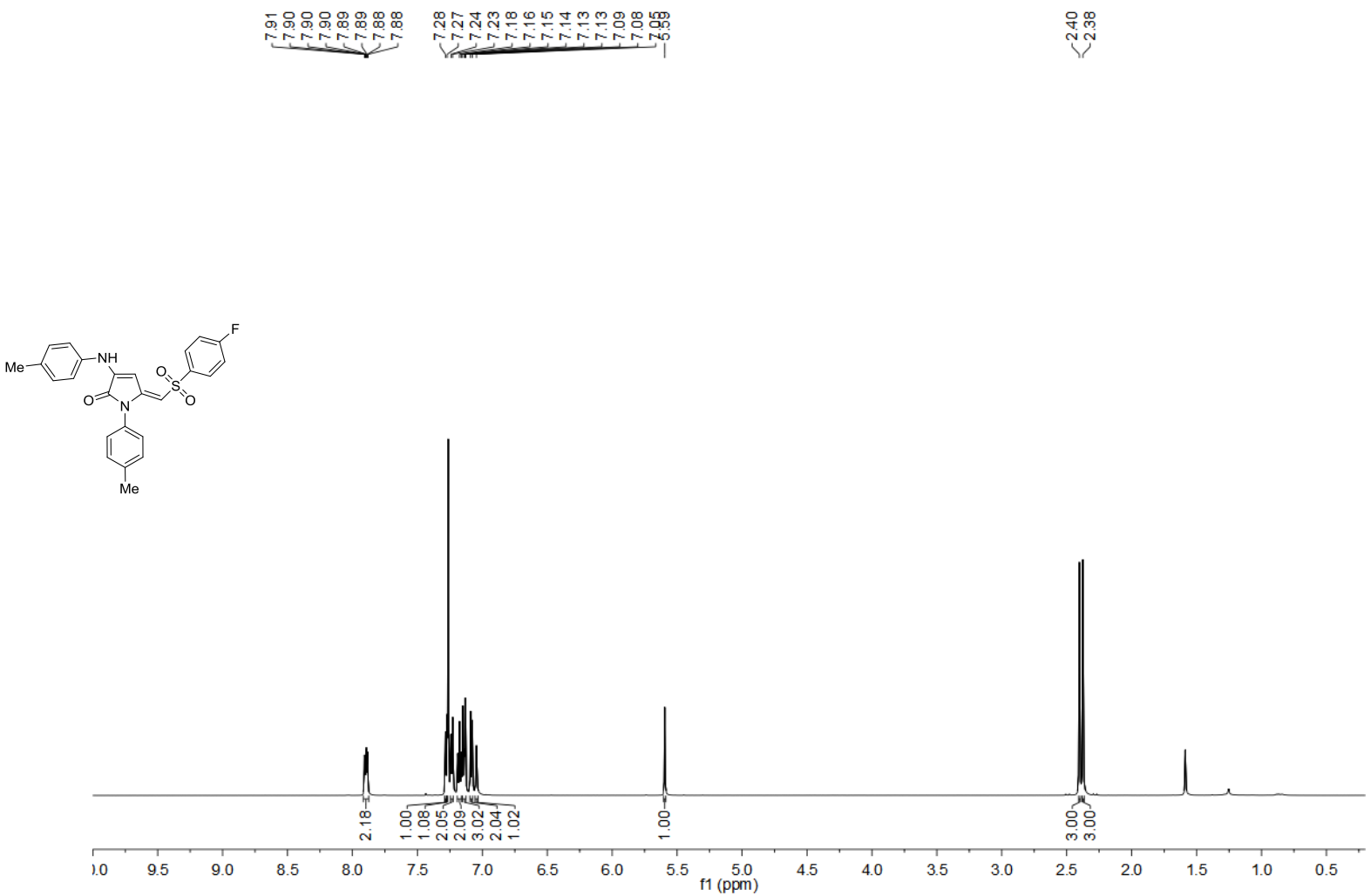


Figure S68. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **5e**

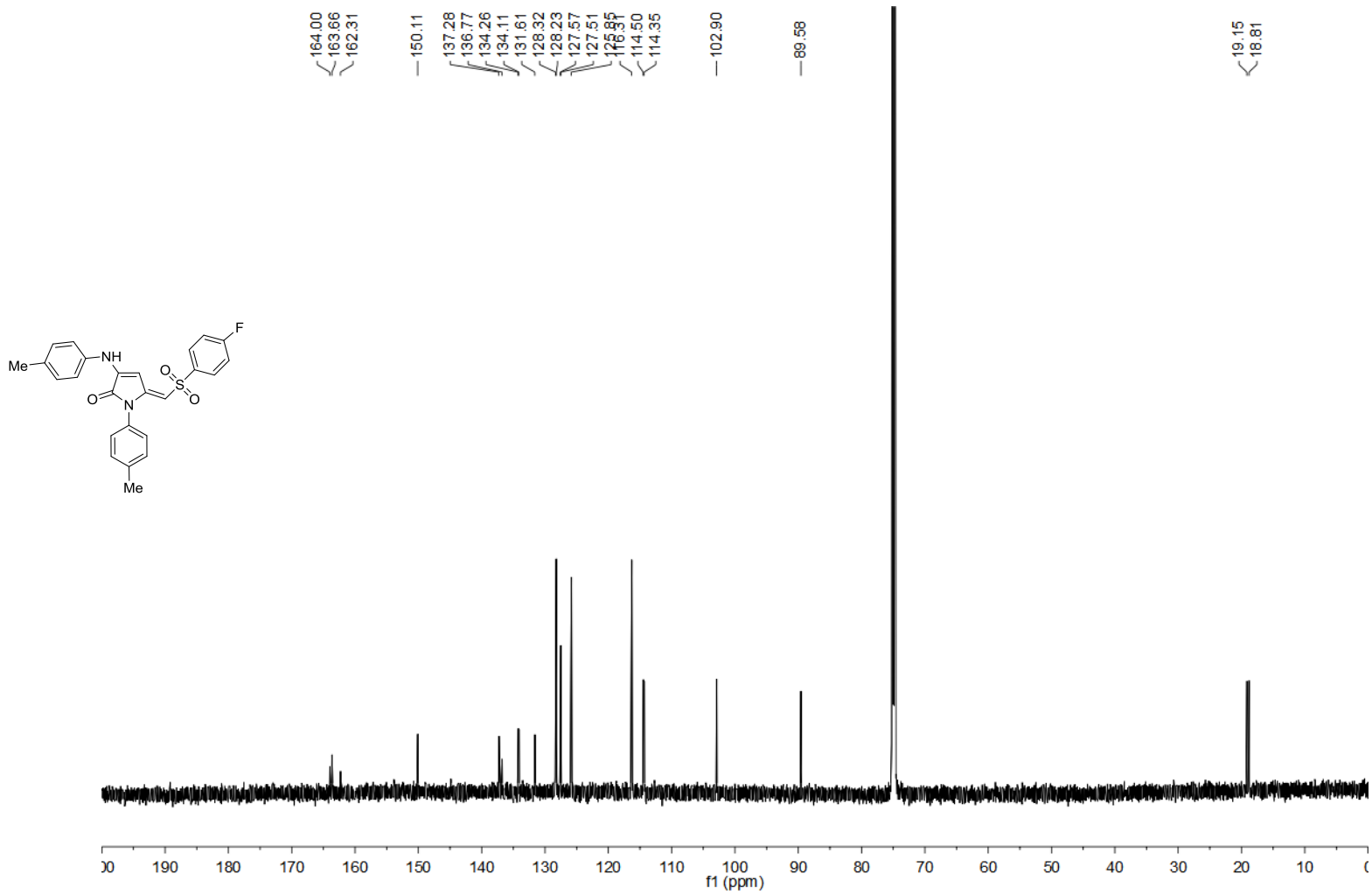


Figure S69. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **5e**

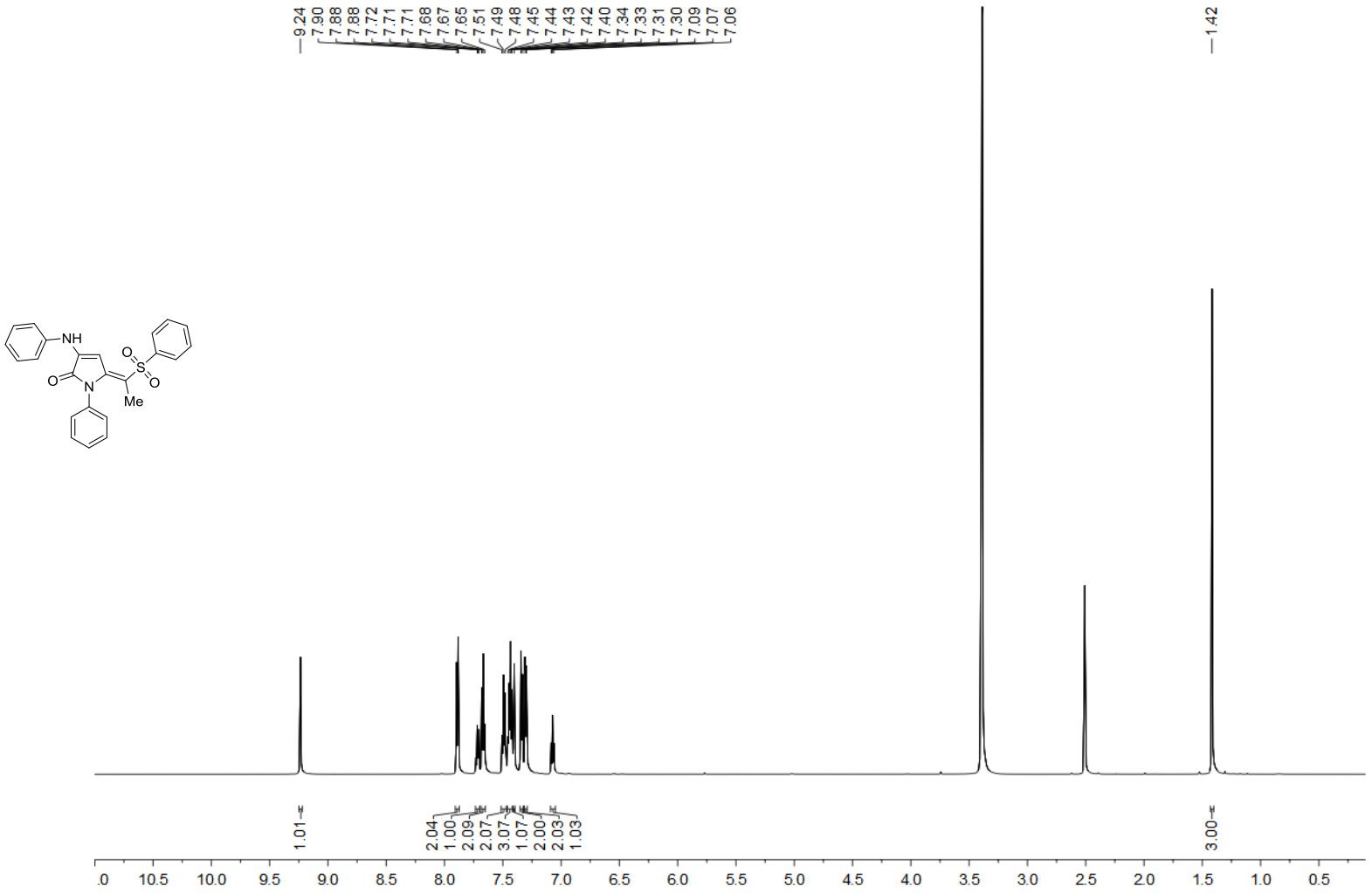


Figure S70. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **6a**

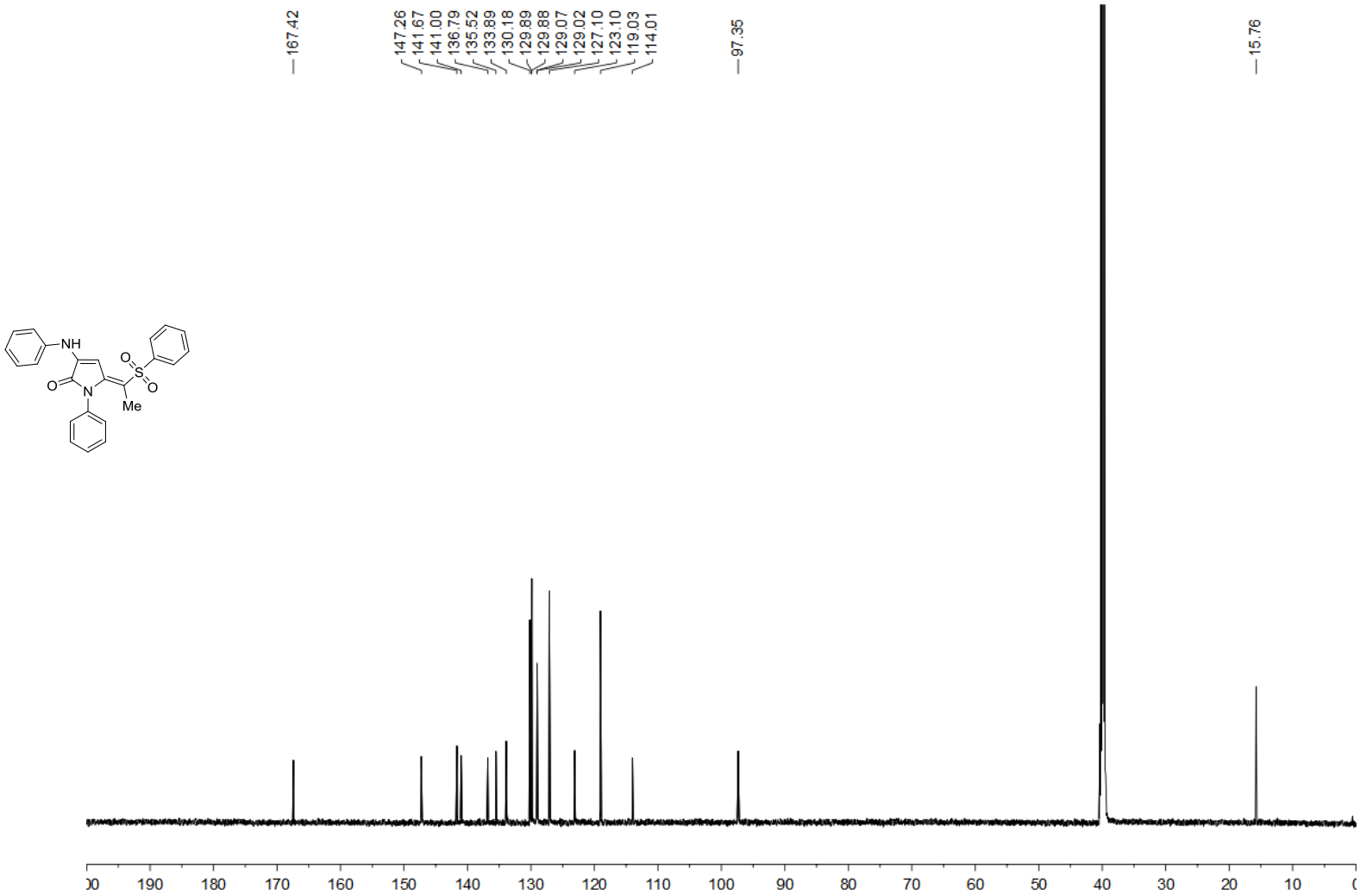


Figure S71. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **6a**

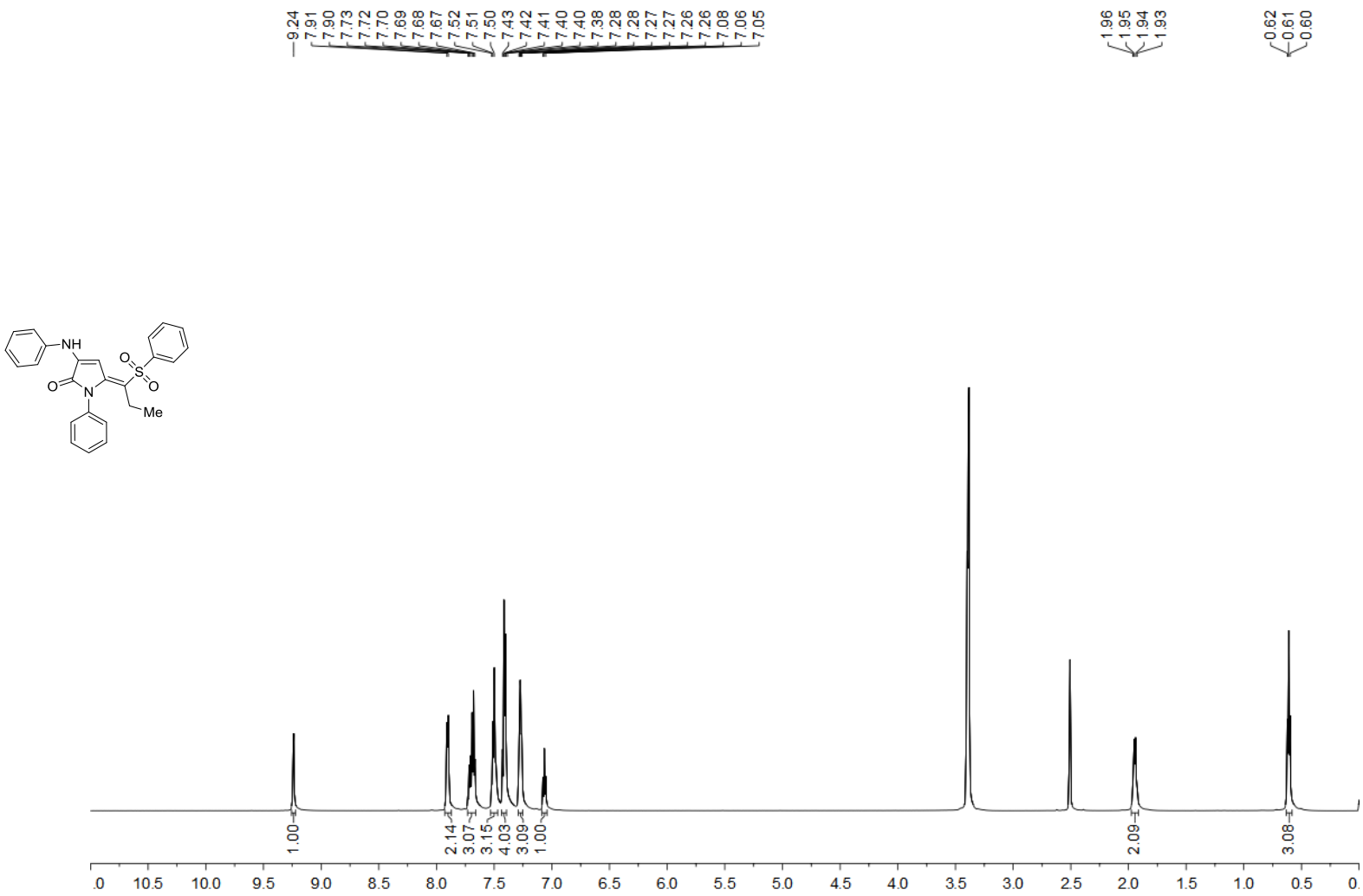


Figure S72. [1]**H NMR** (600 MHz, DMSO-d_6) spectra of compound **6b**

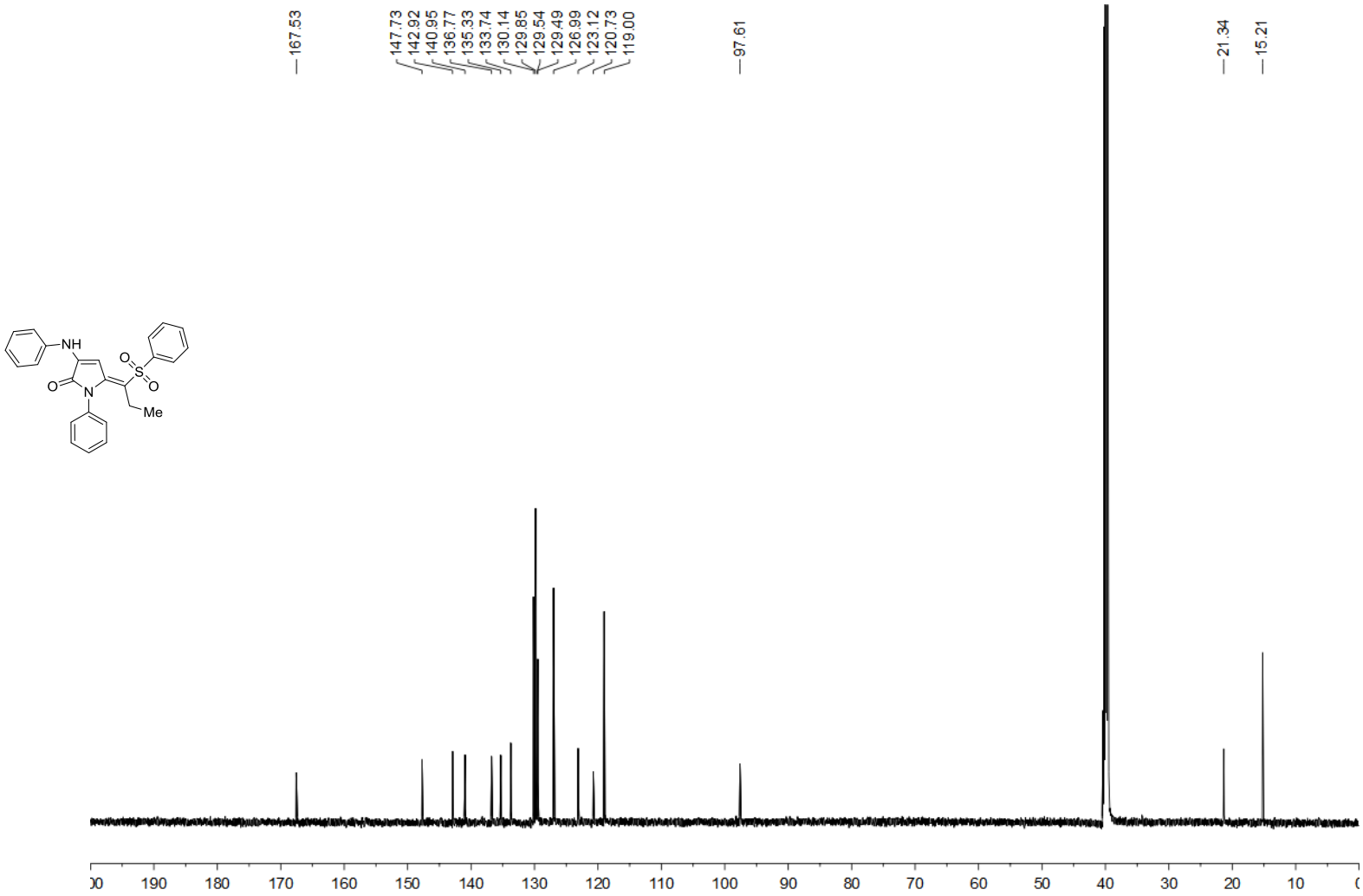


Figure S73. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **6b**

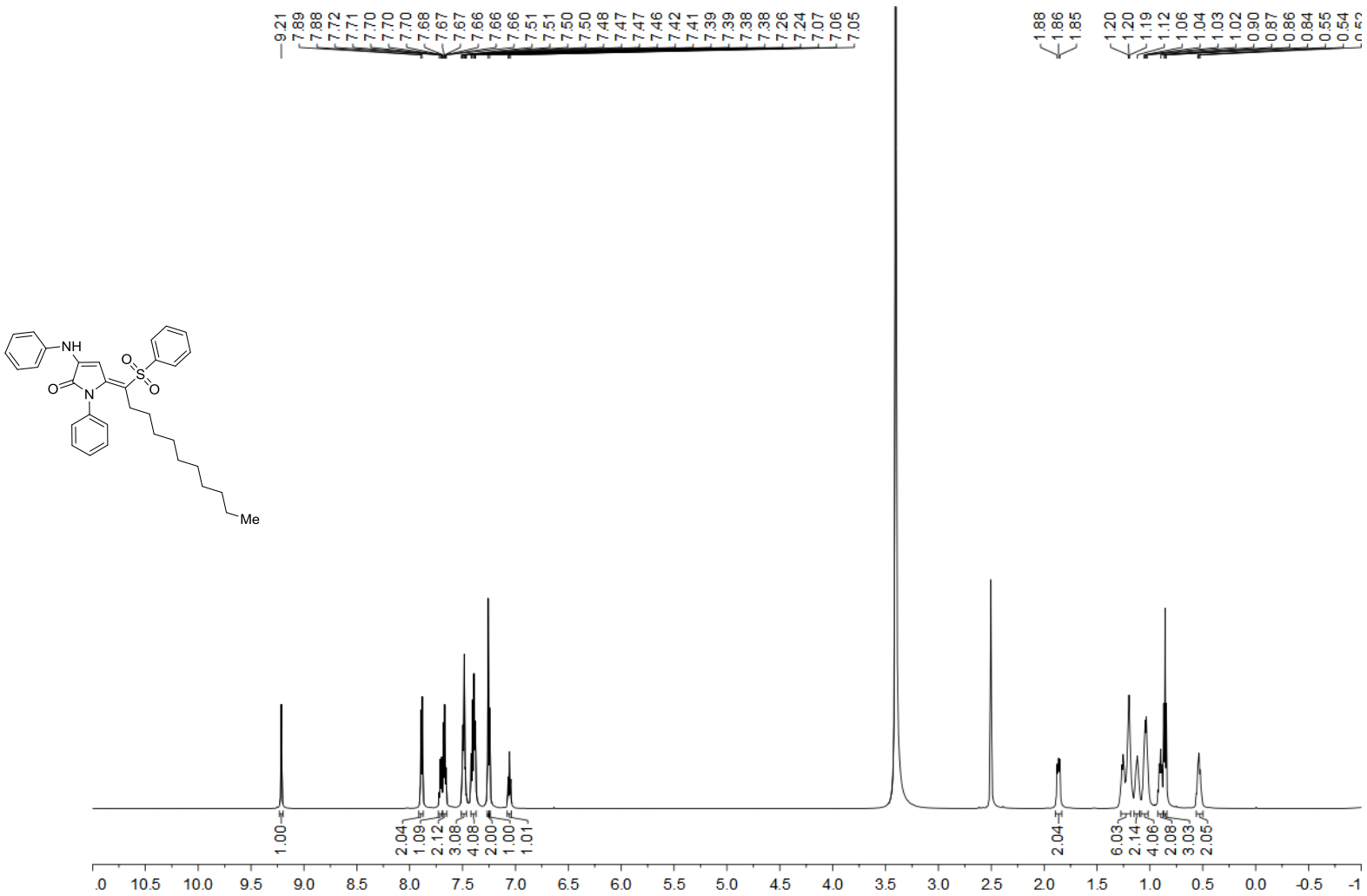


Figure S74. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **6c**

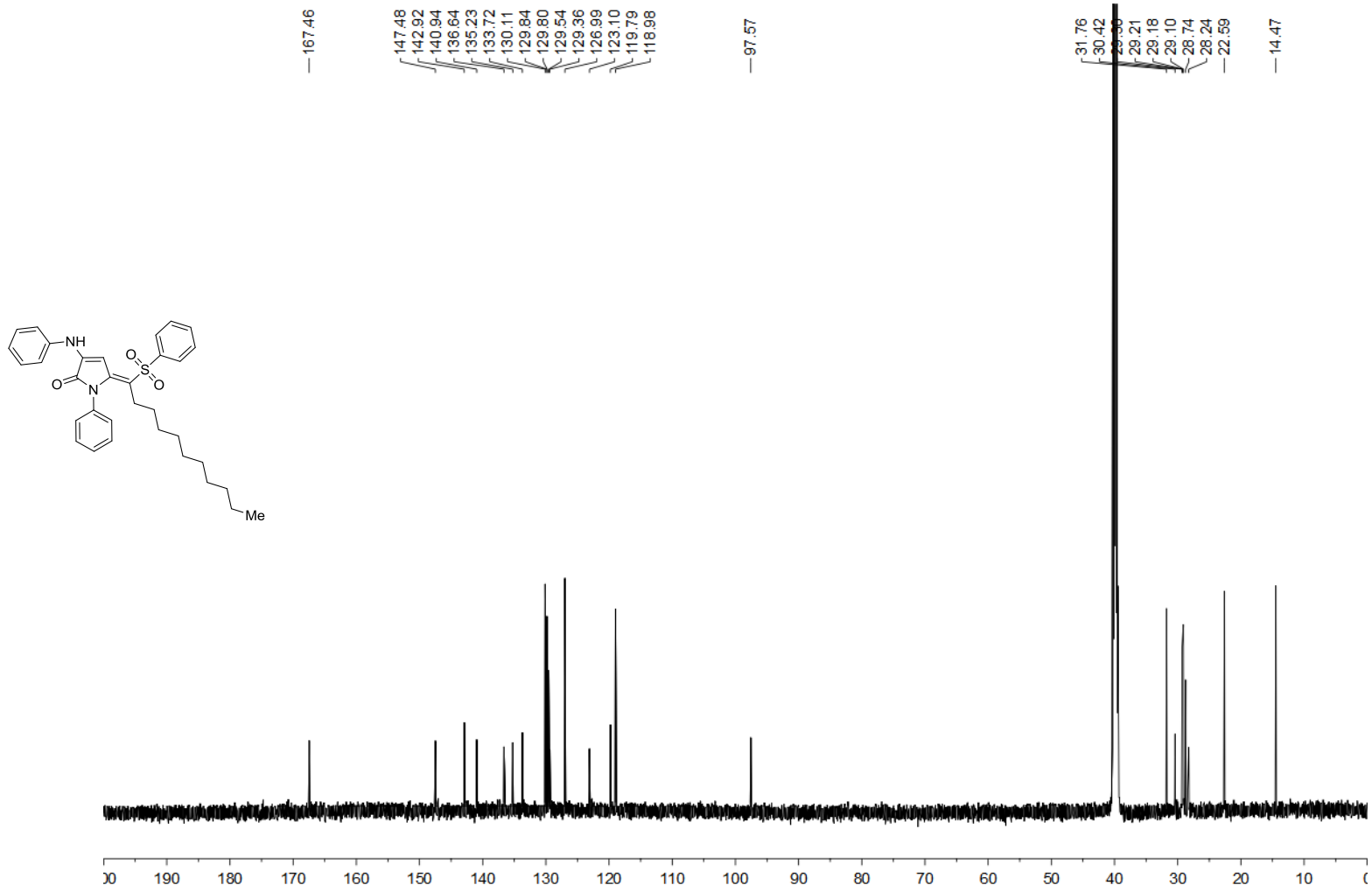


Figure S75. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **6c**

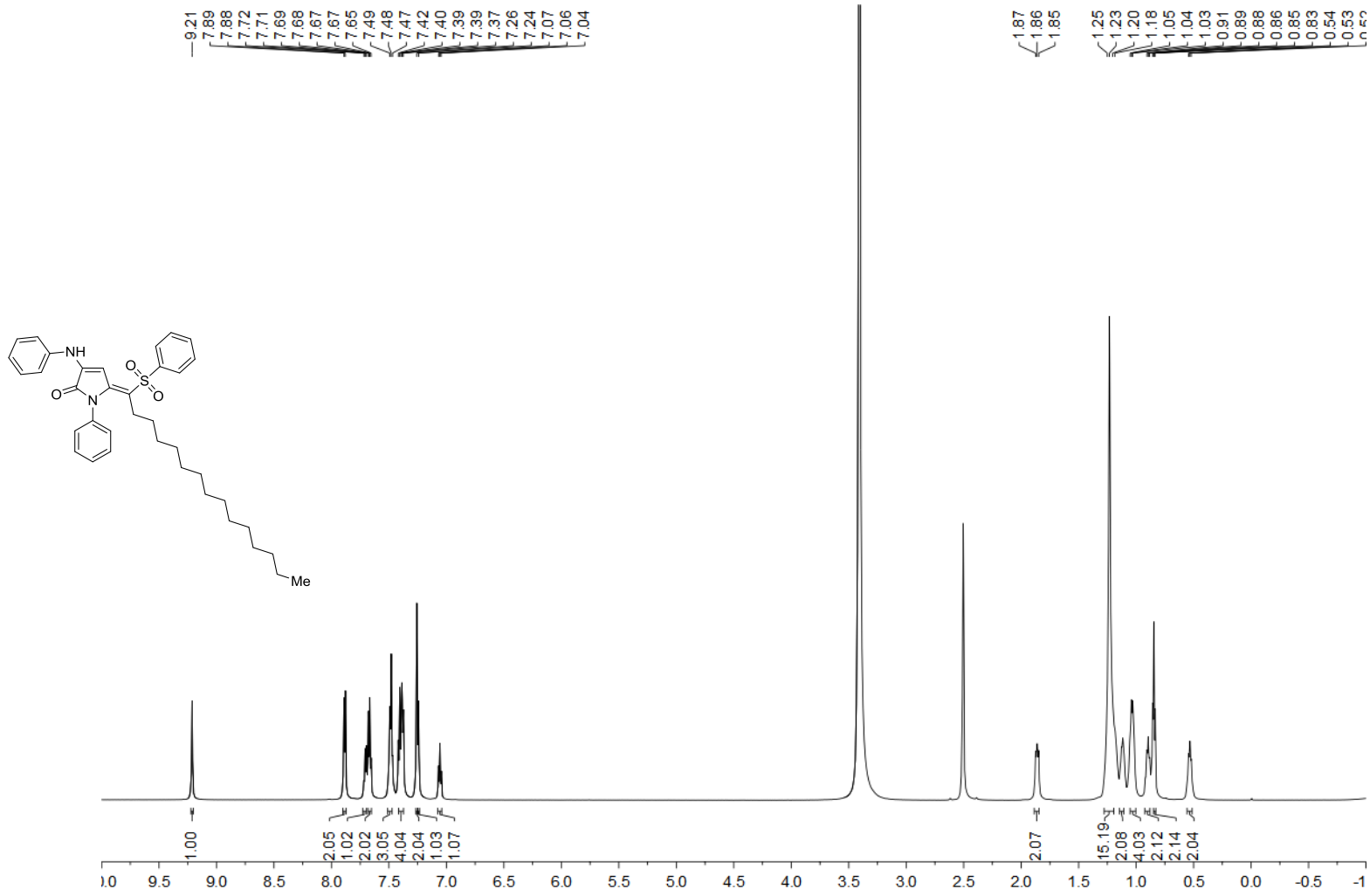


Figure S76. [1]**H NMR** (600 MHz, DMSO-d_6) spectra of compound **6d**

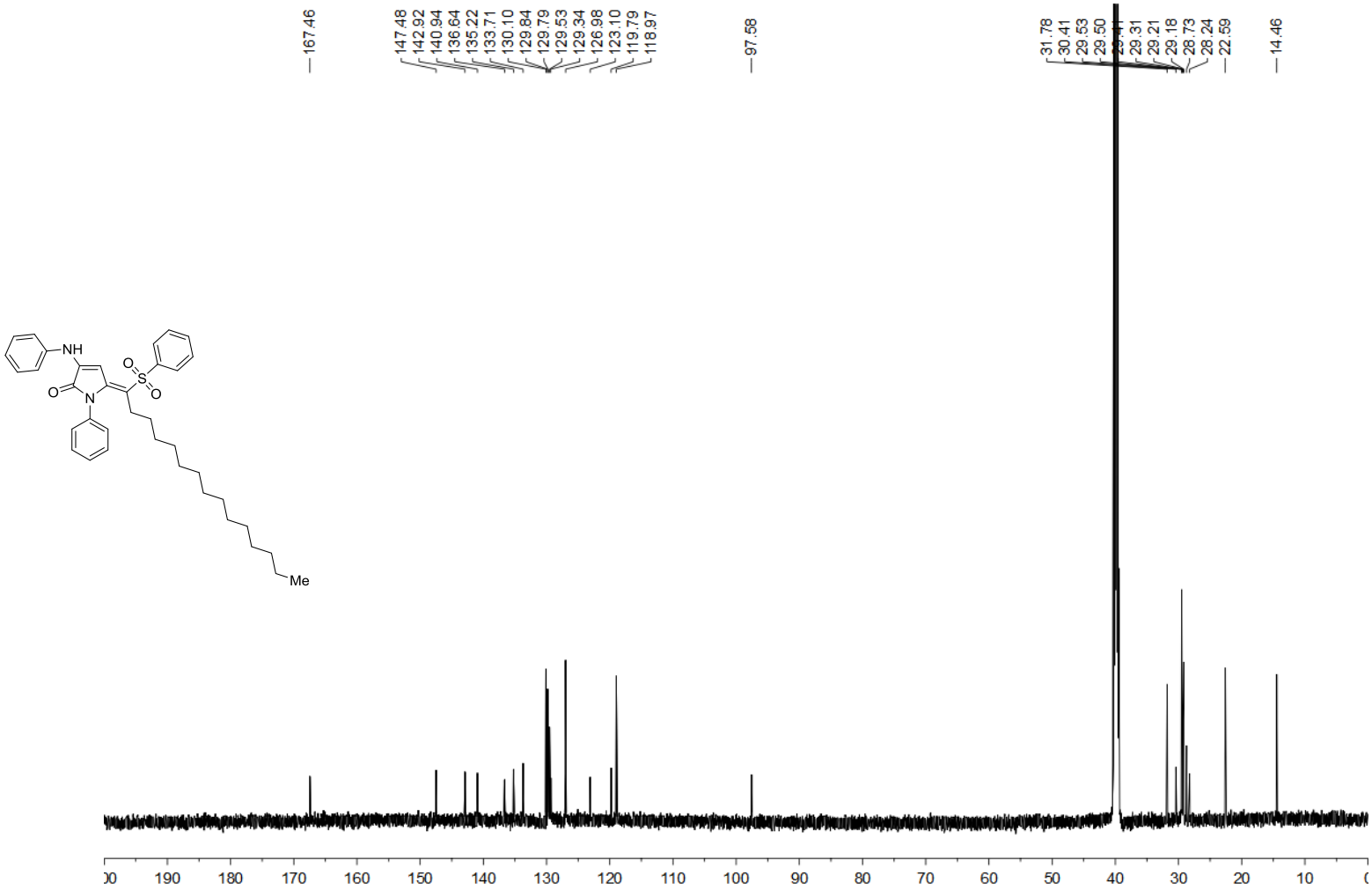


Figure S77. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **6d**

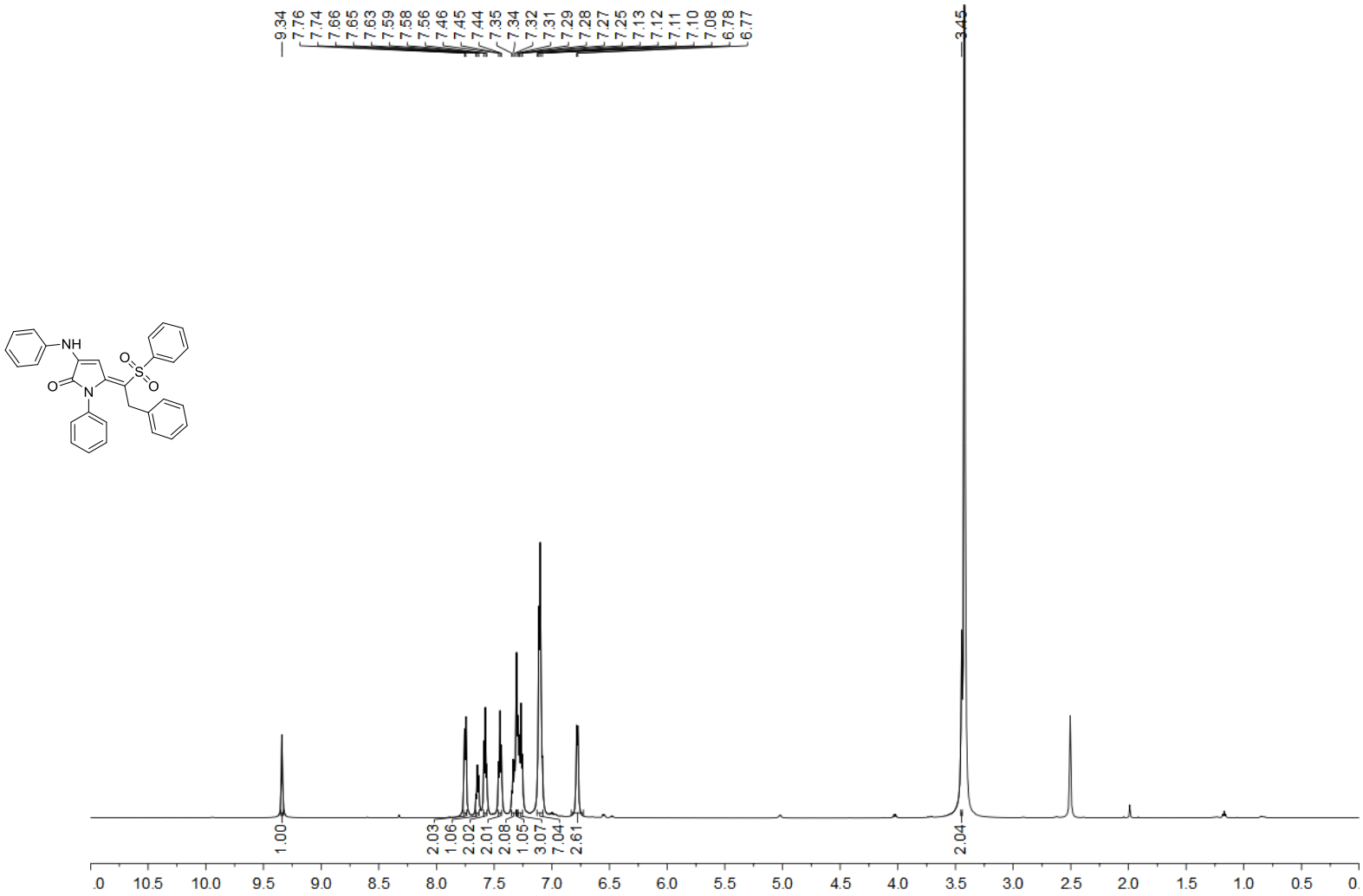


Figure S78. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **6e**

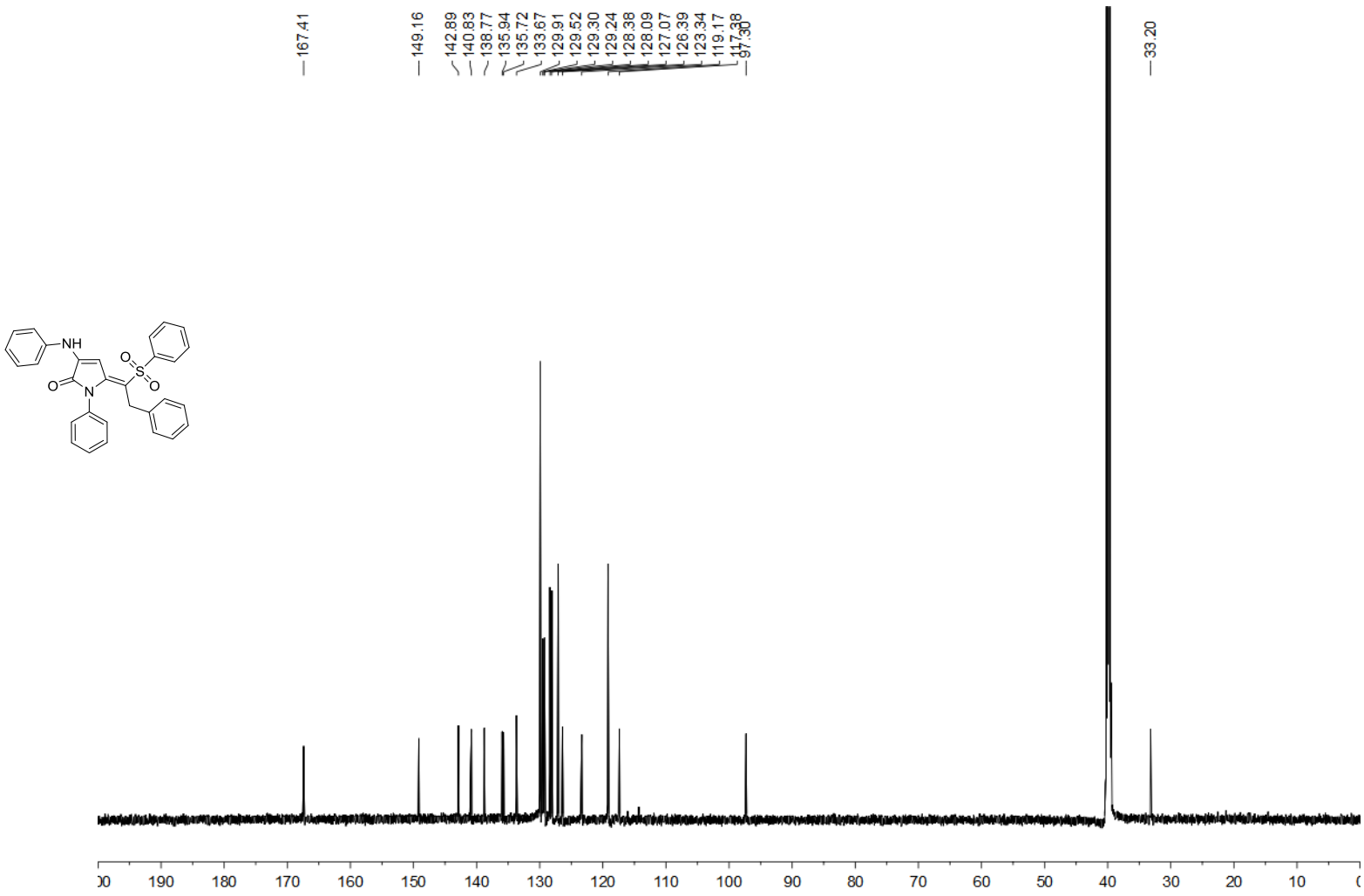


Figure S79. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **6e**

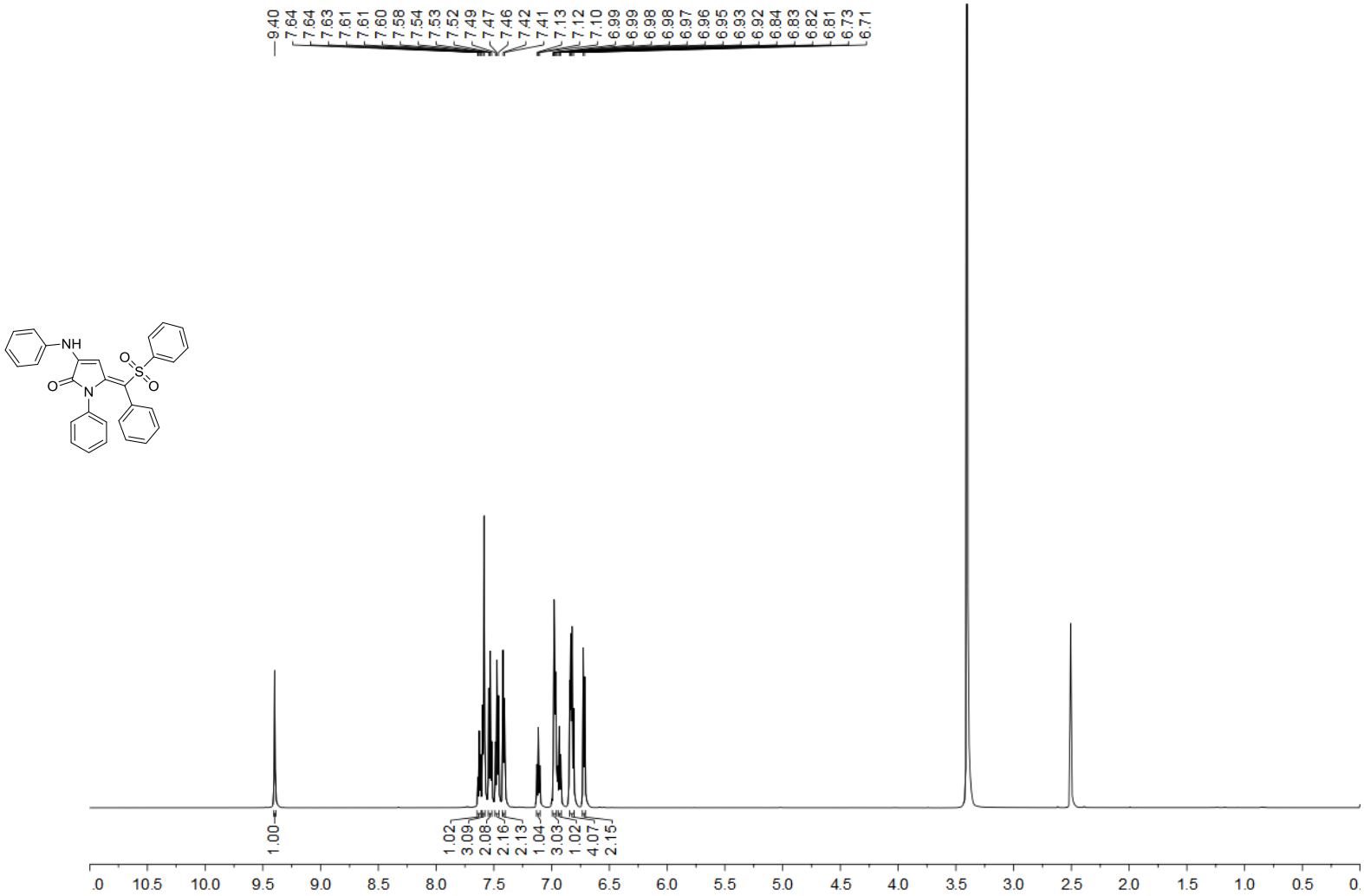


Figure S80. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **6f**

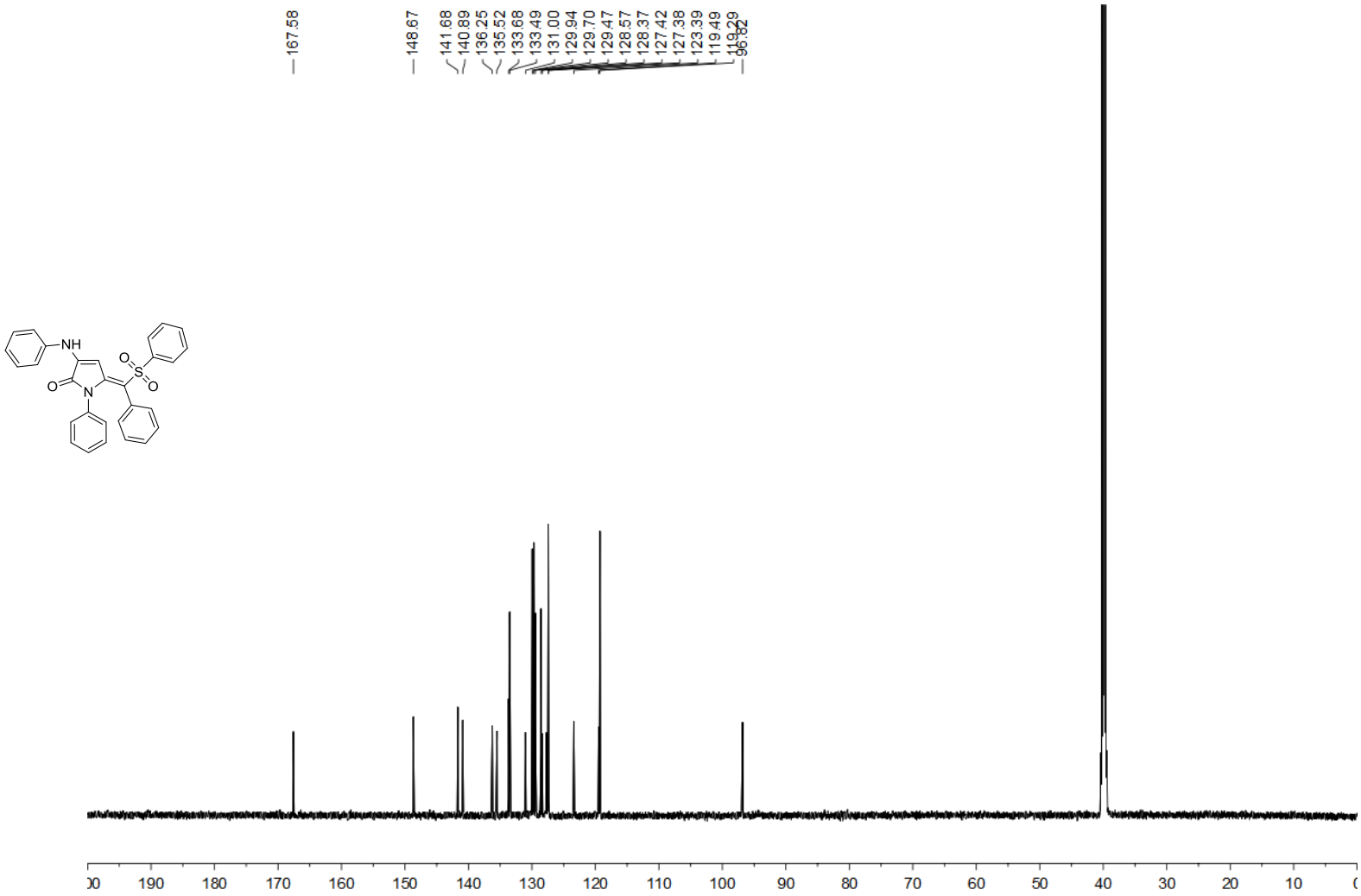


Figure S81. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **6f**

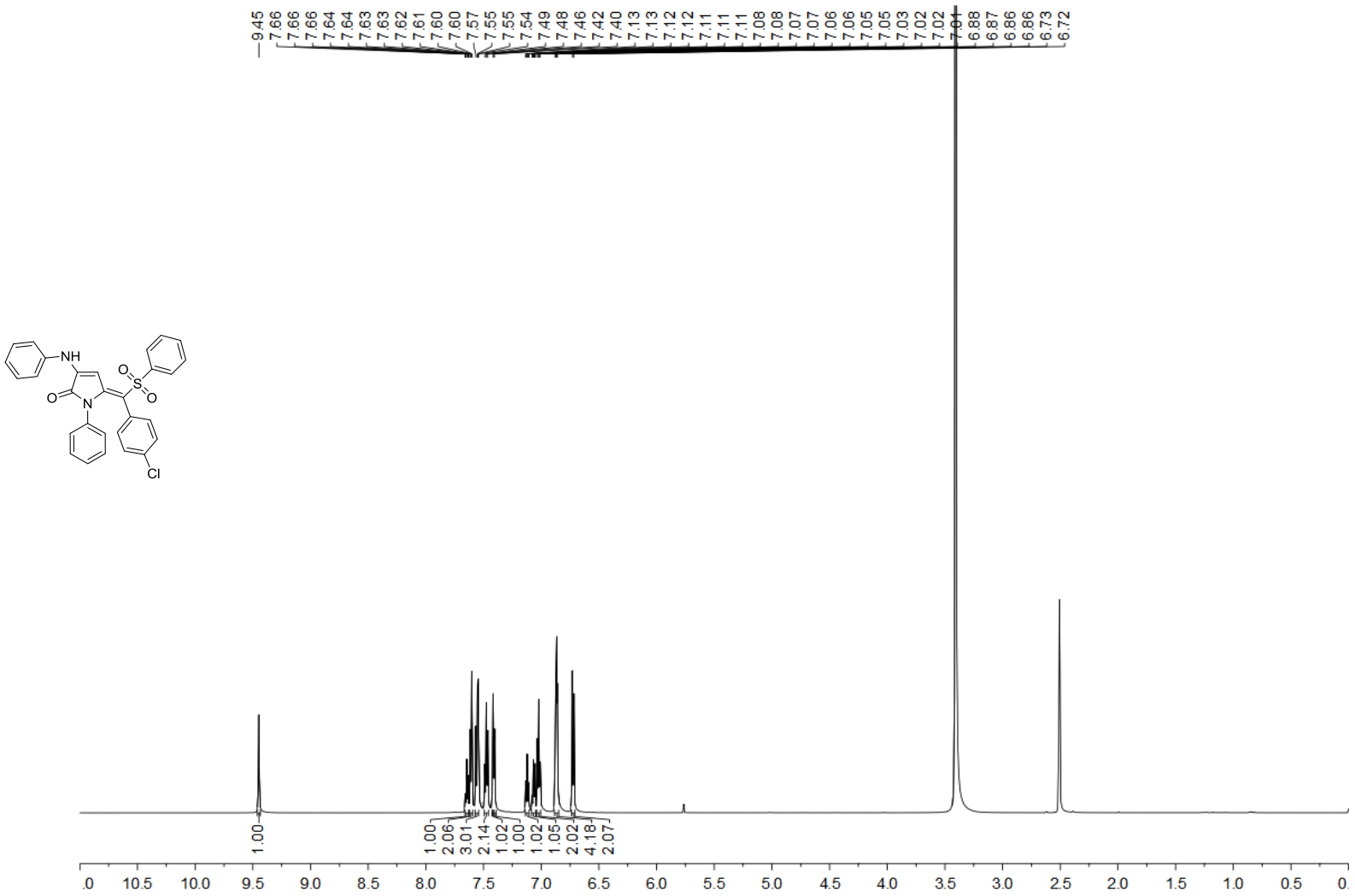


Figure S82. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **6g**

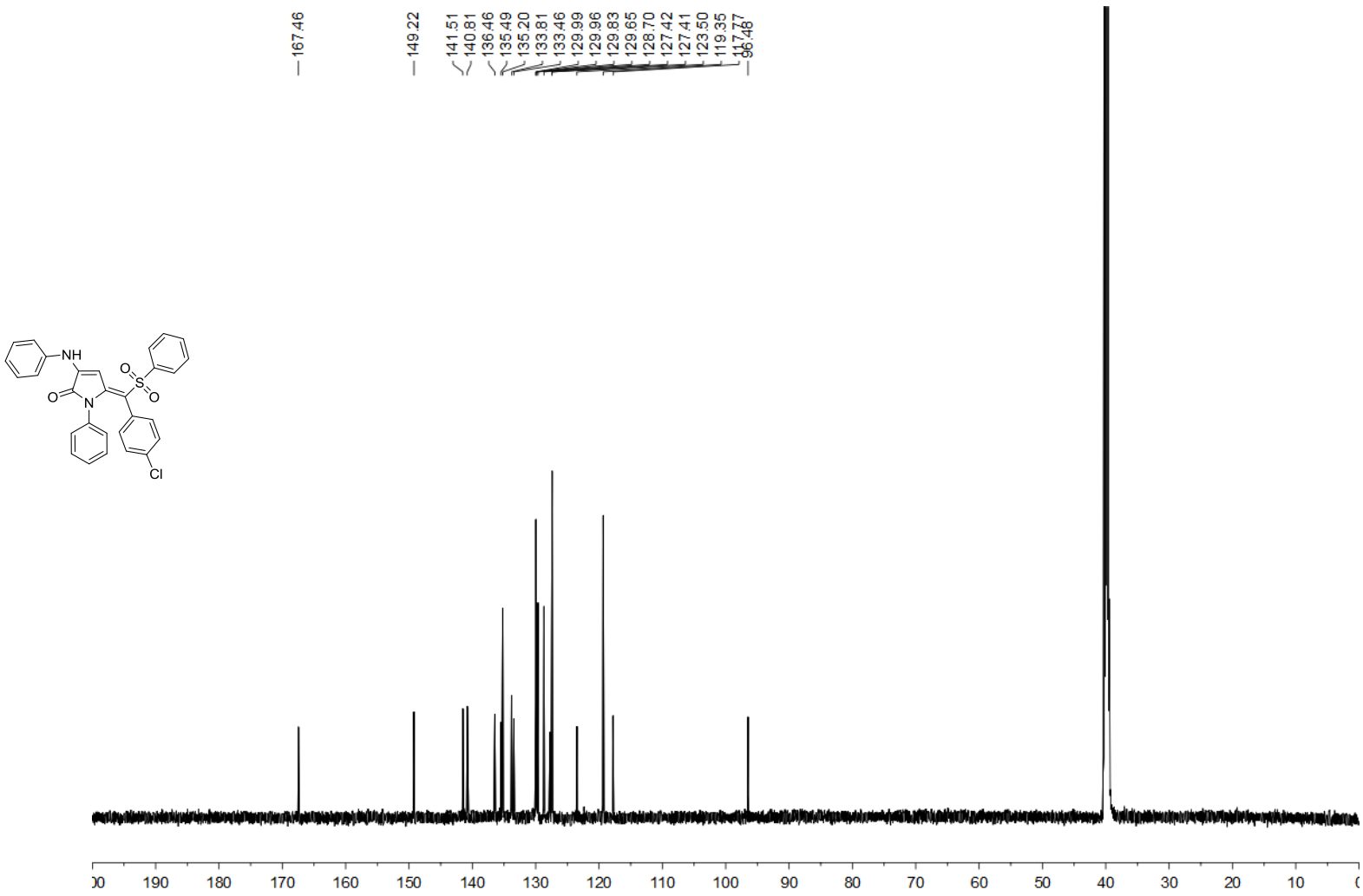


Figure S83. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **6g**

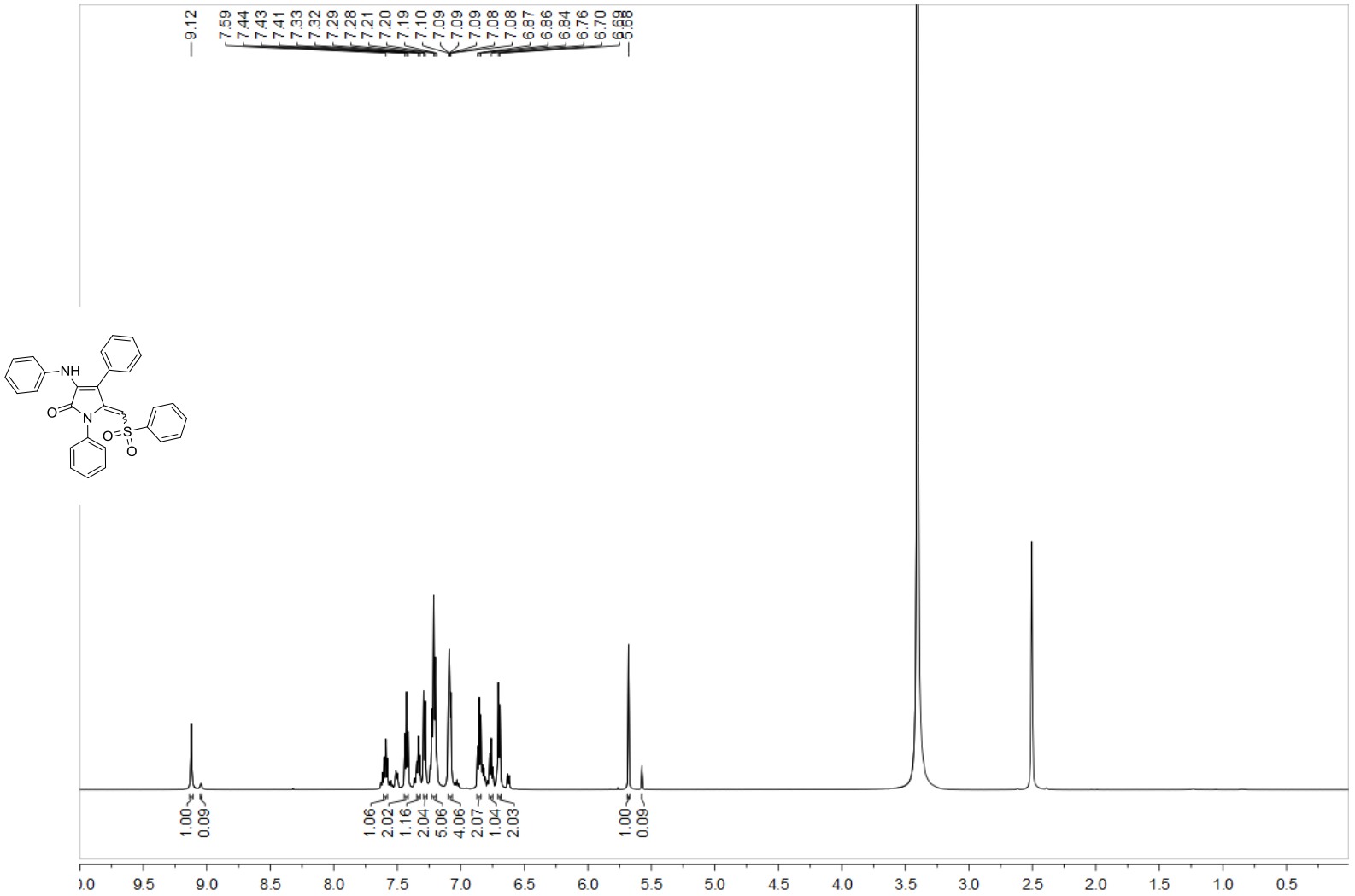


Figure S84. **^{1}H NMR** (600 MHz, DMSO-d_6) spectra of compound **7a**

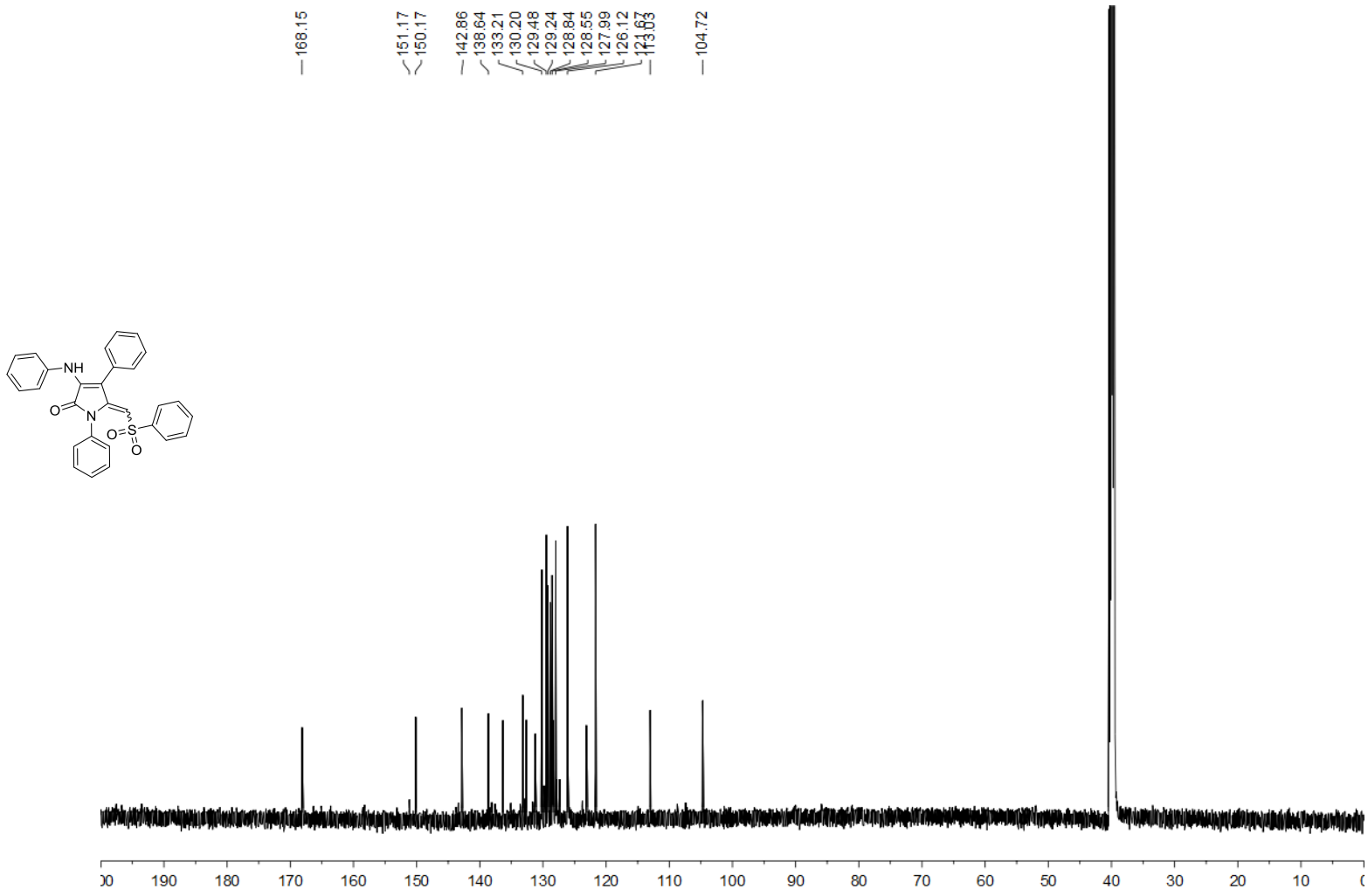


Figure S85. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **7a**

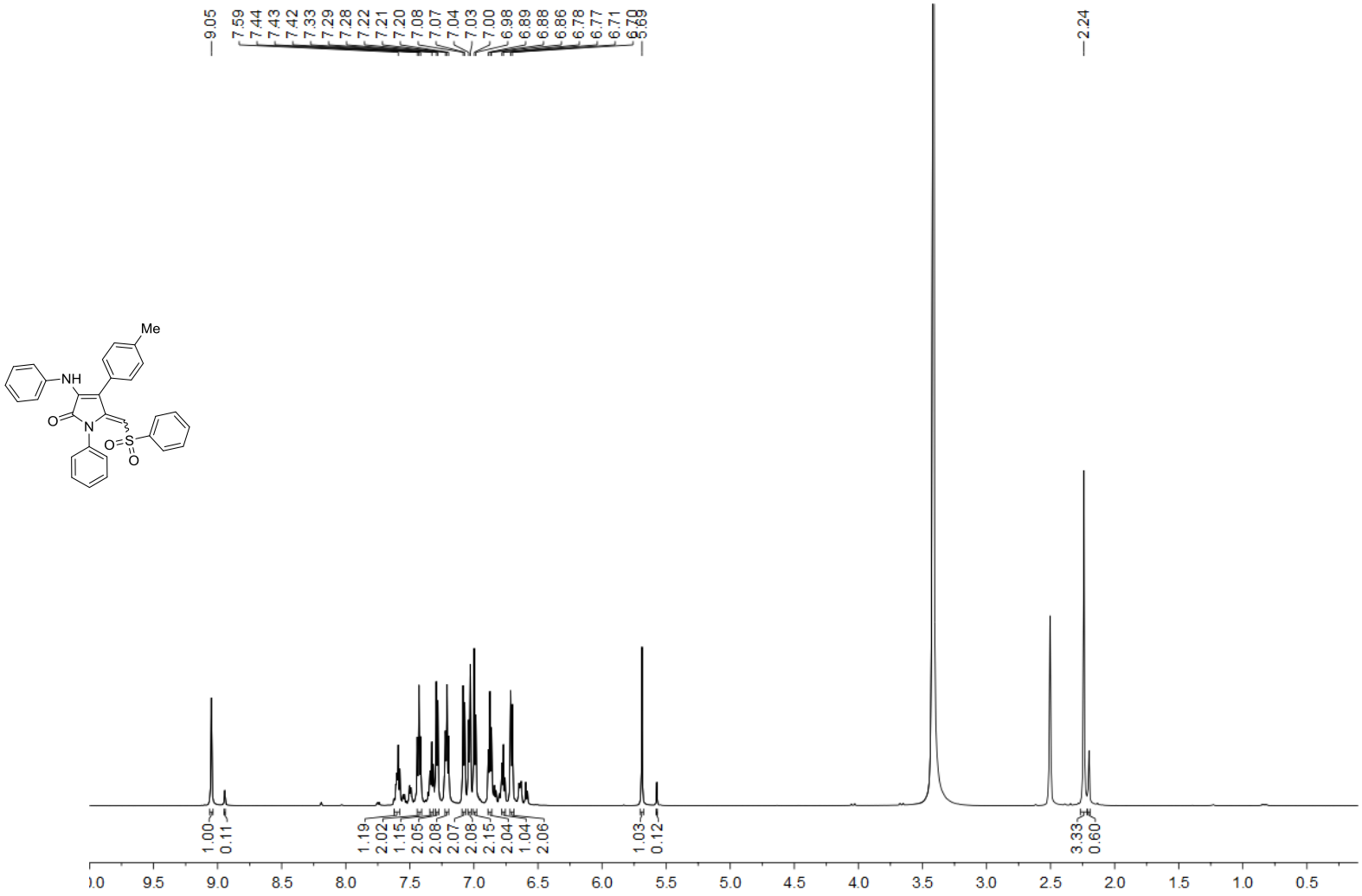


Figure S86. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **7b**

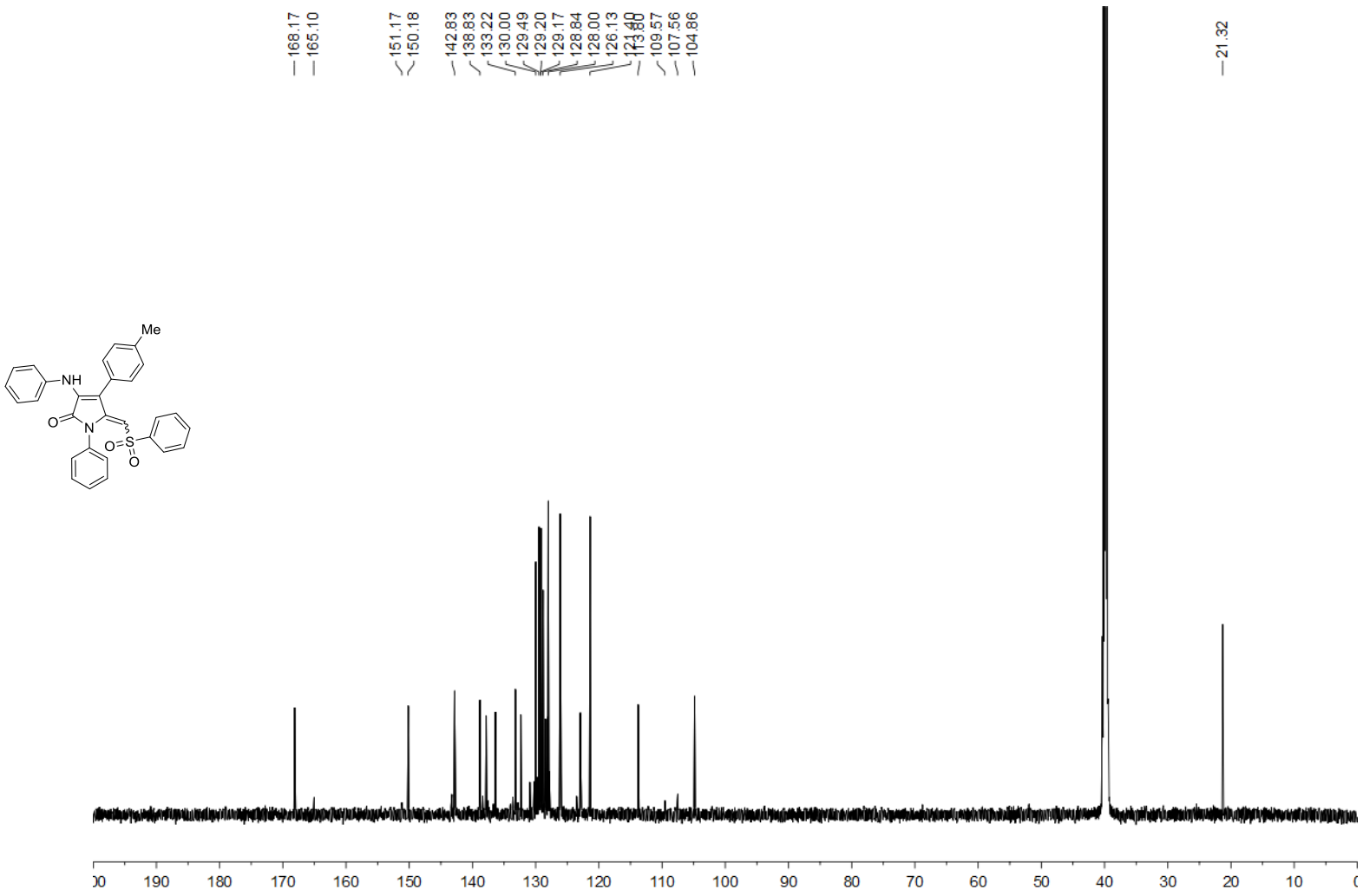


Figure S87. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **7b**

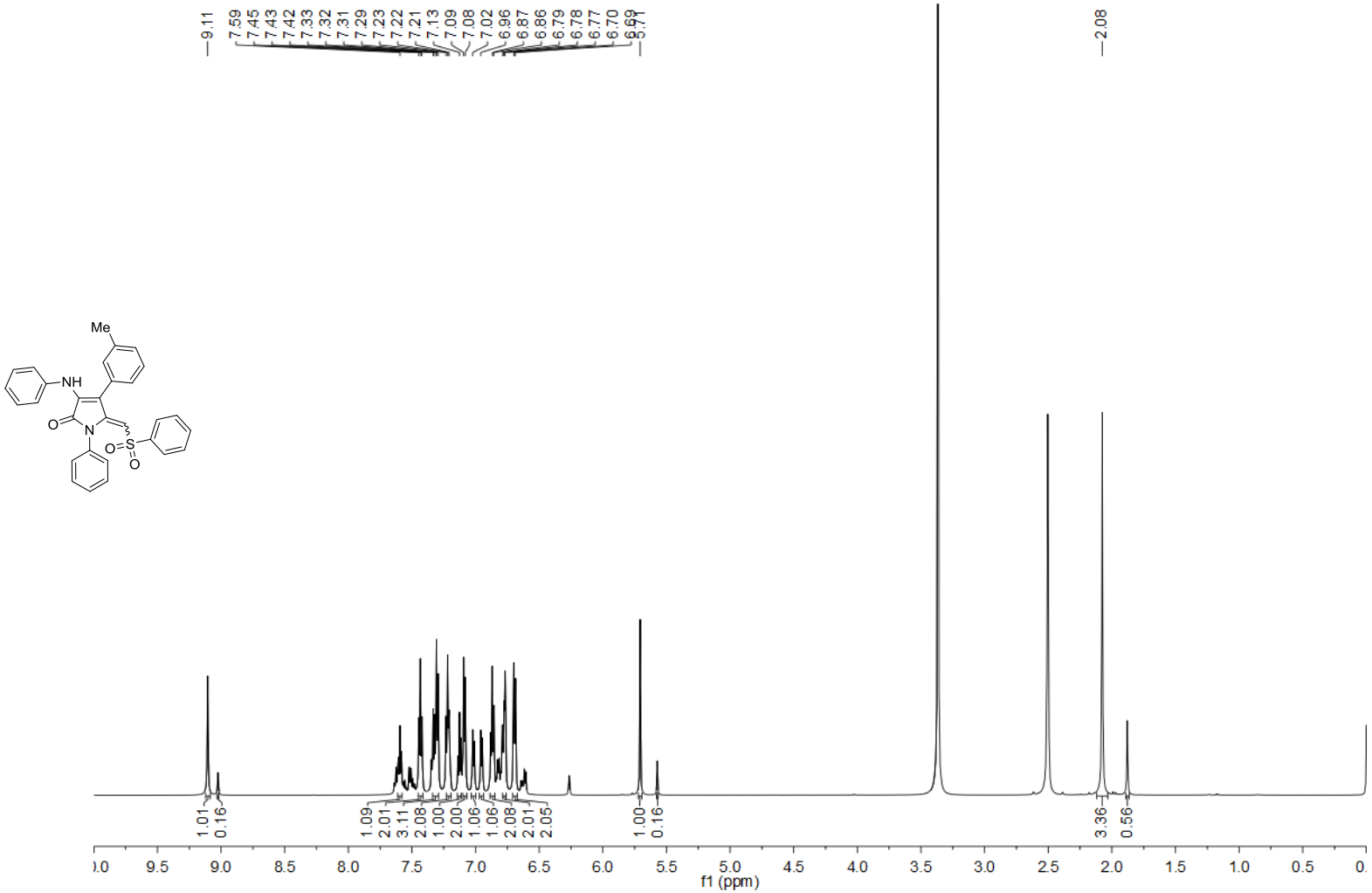


Figure S88. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **7c**

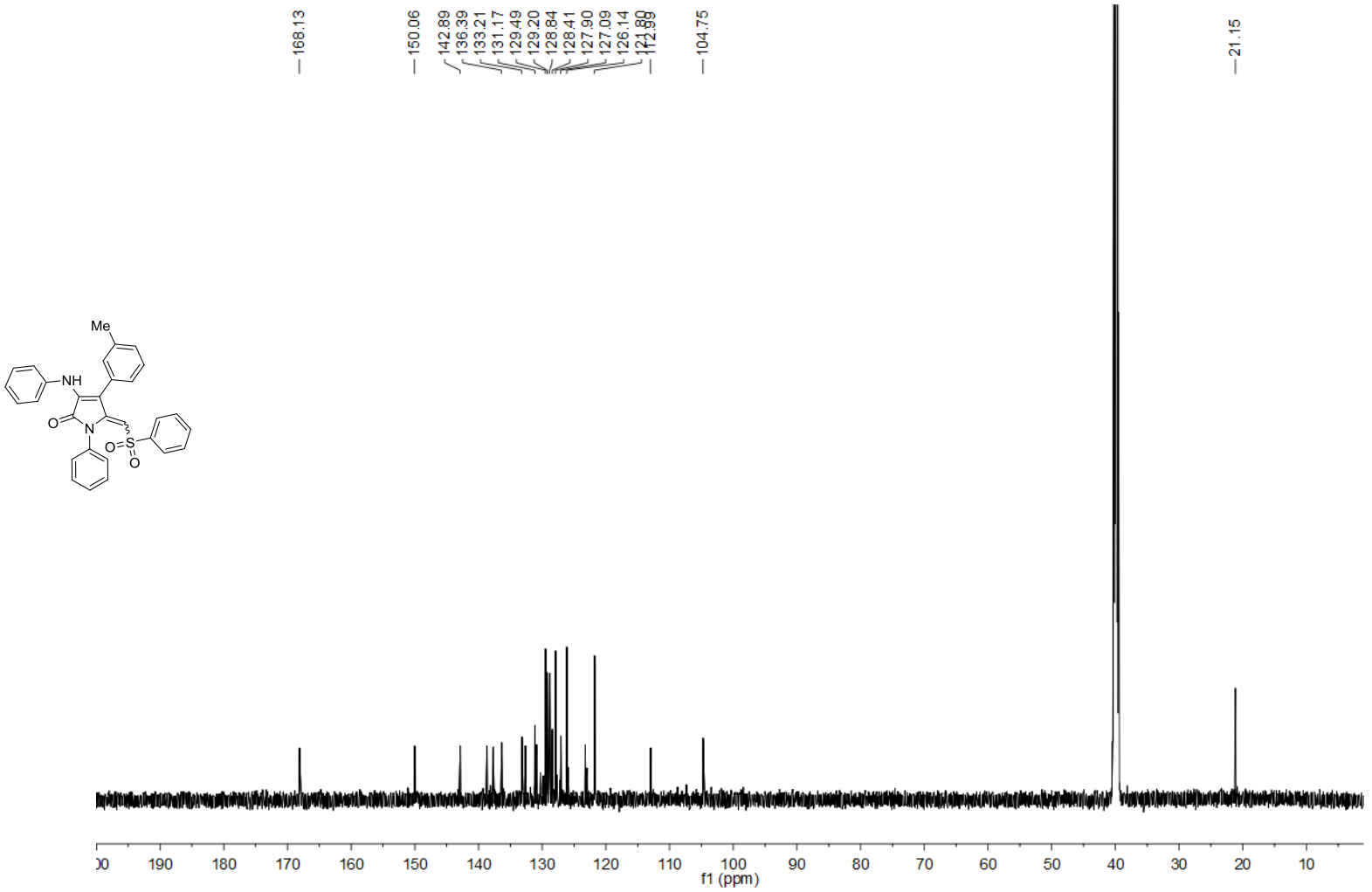


Figure S89. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **7c**

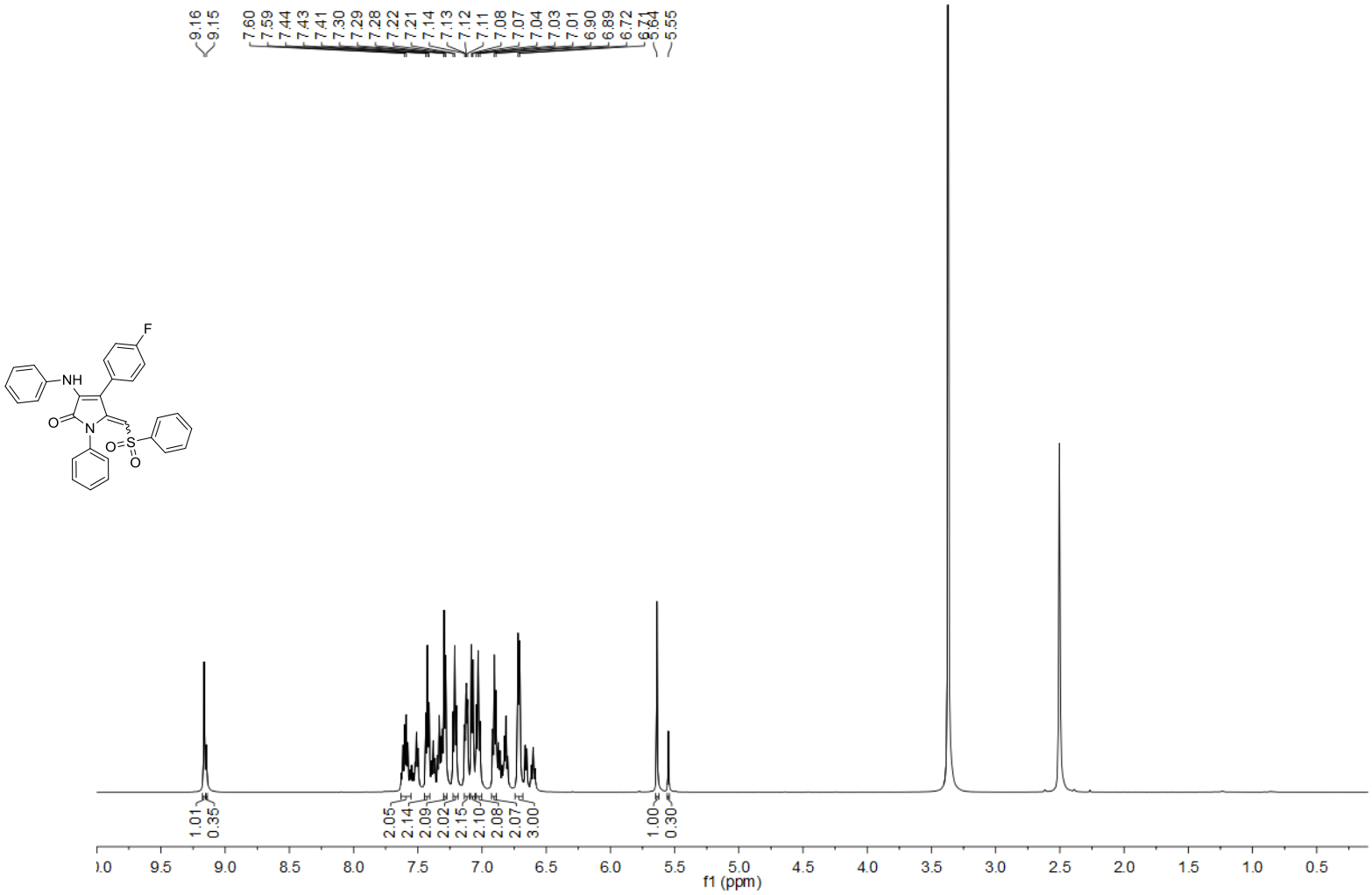


Figure S90. [1]**H NMR** (600 MHz, DMSO-d_6) spectra of compound **7d**

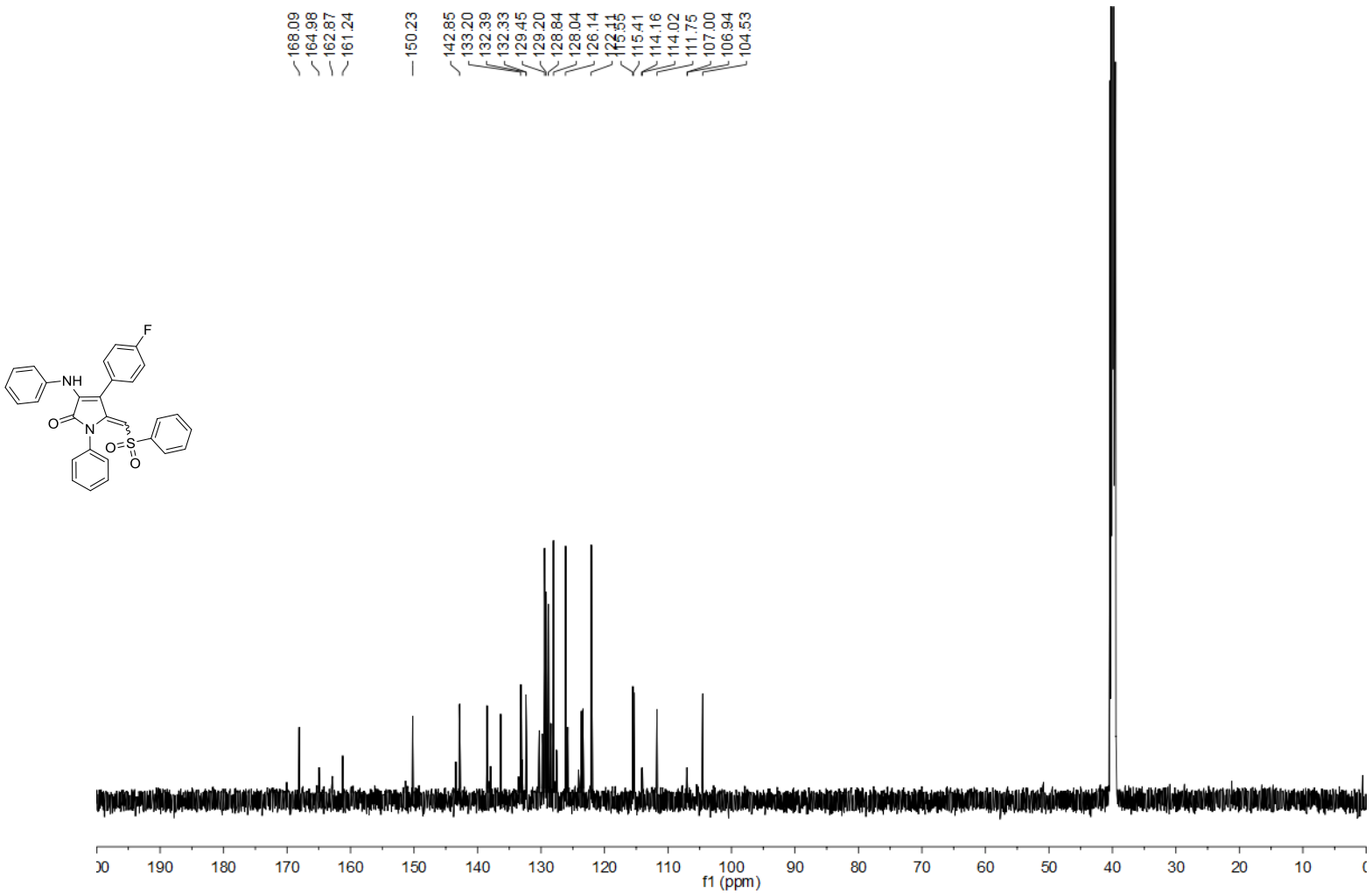


Figure S91. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **7d**

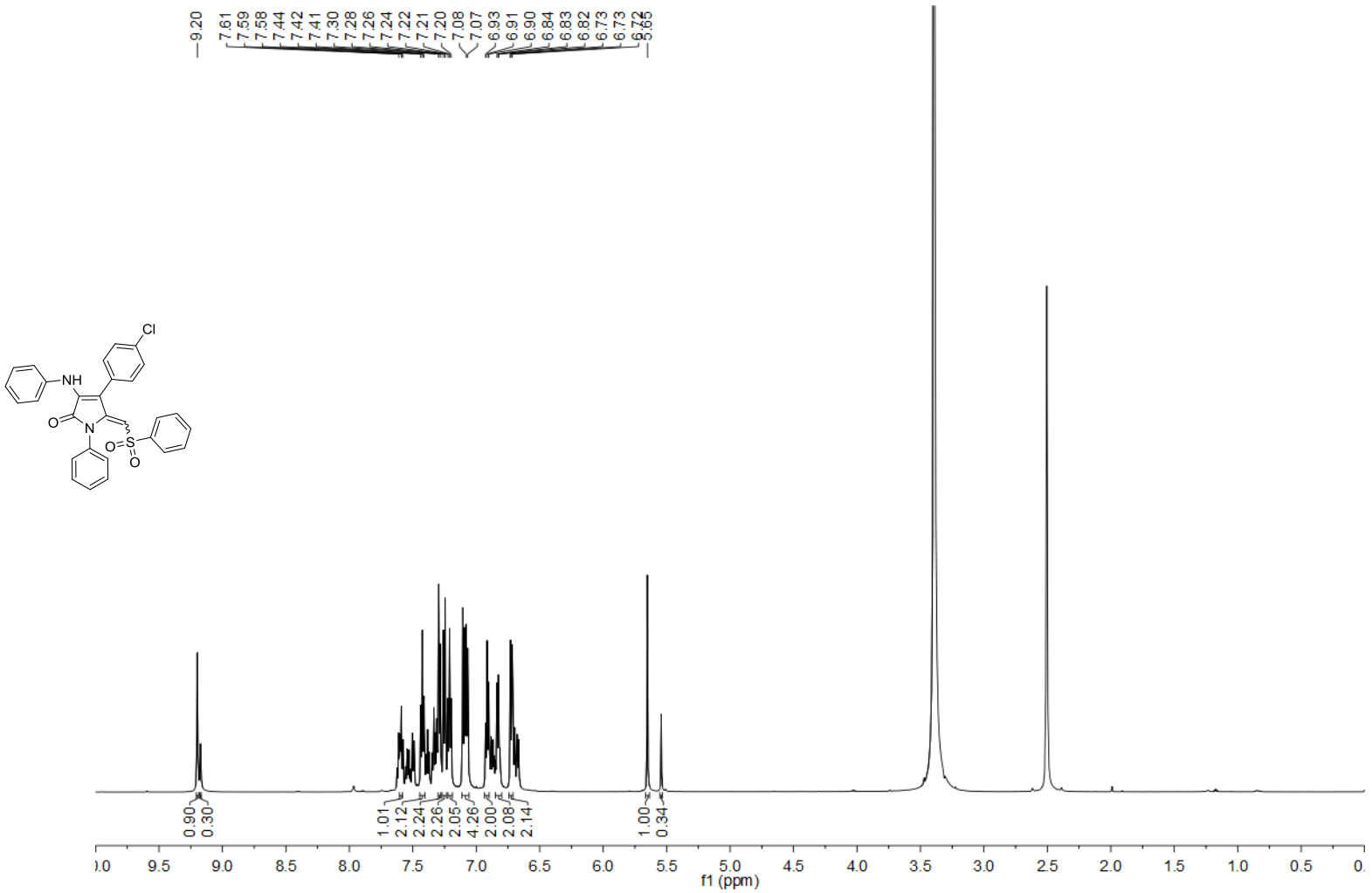


Figure S92. **^{1}H NMR** (600 MHz, DMSO-d_6) spectra of compound **7e**

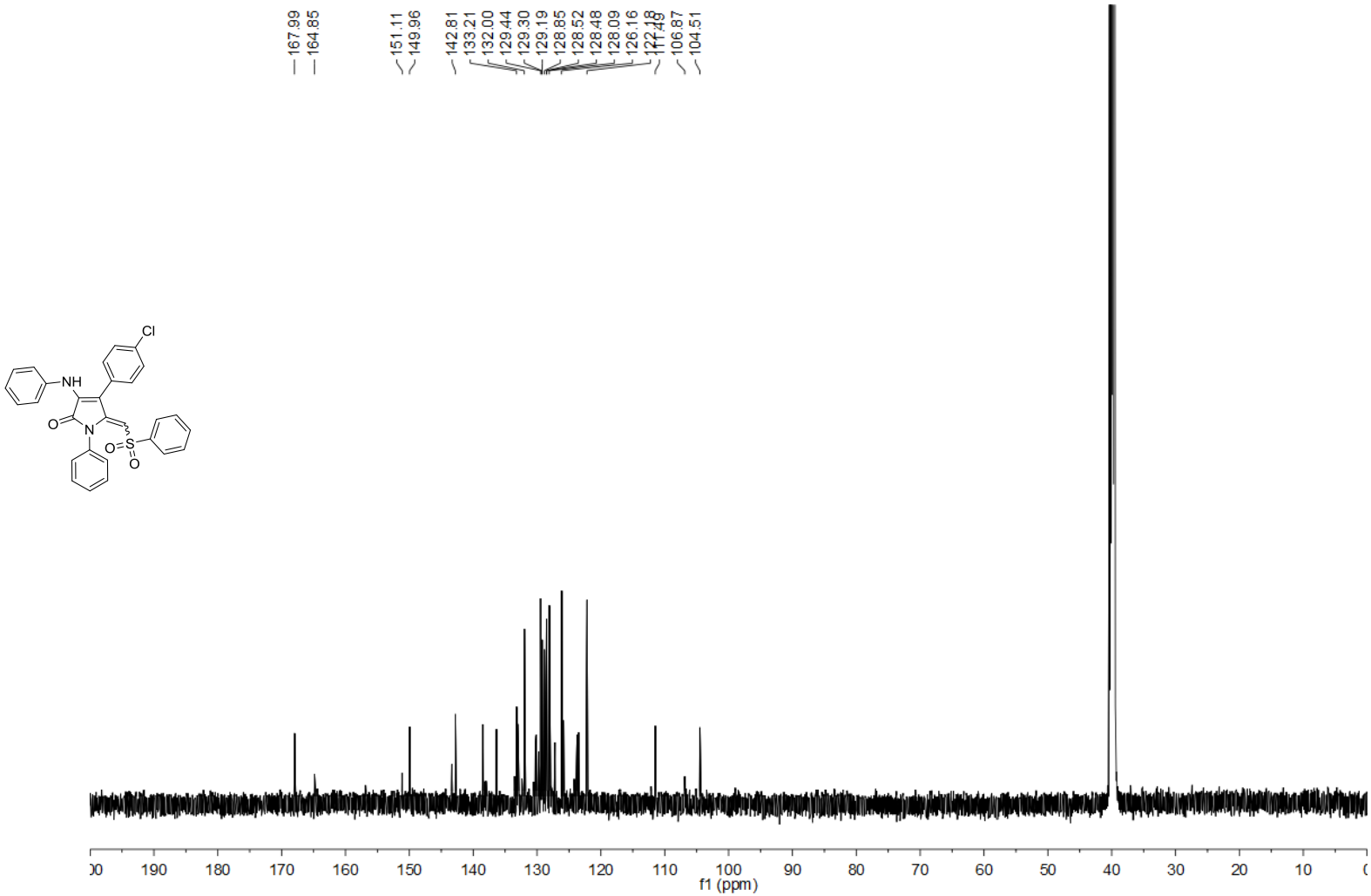


Figure S93. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **7e**

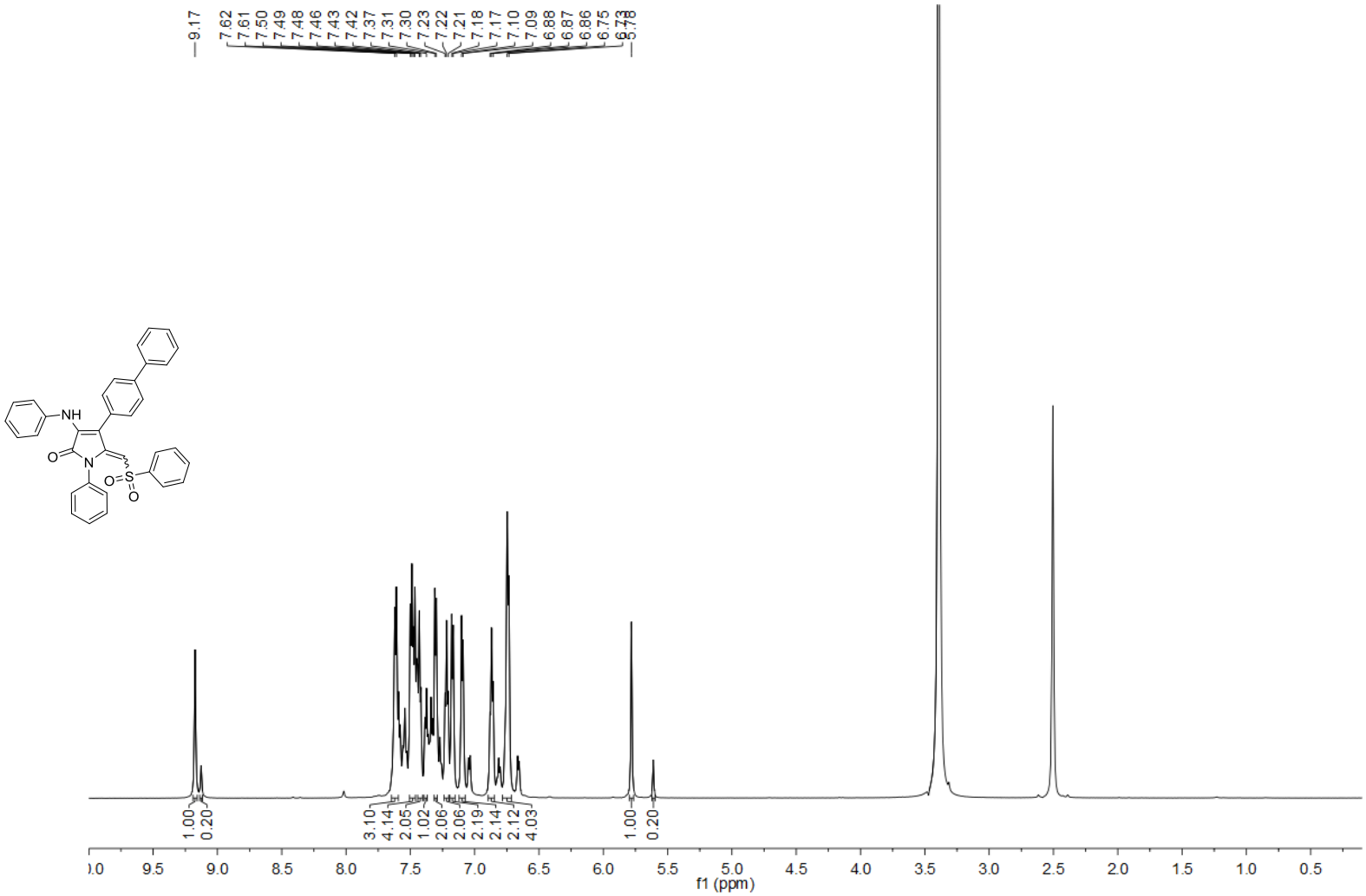


Figure S94. **^{1}H NMR** (600 MHz, DMSO-d_6) spectra of compound **7f**

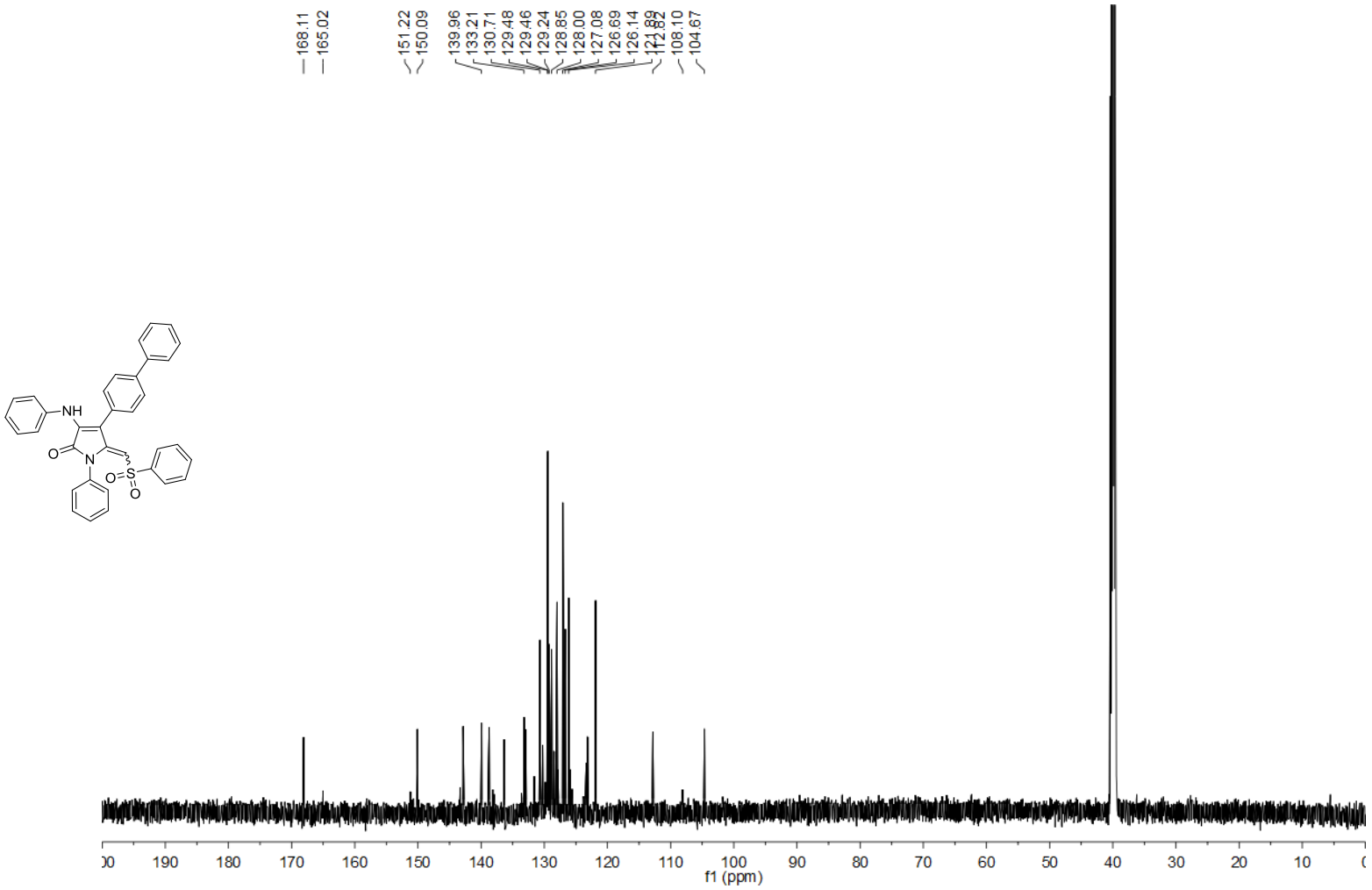


Figure S95. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **7f**

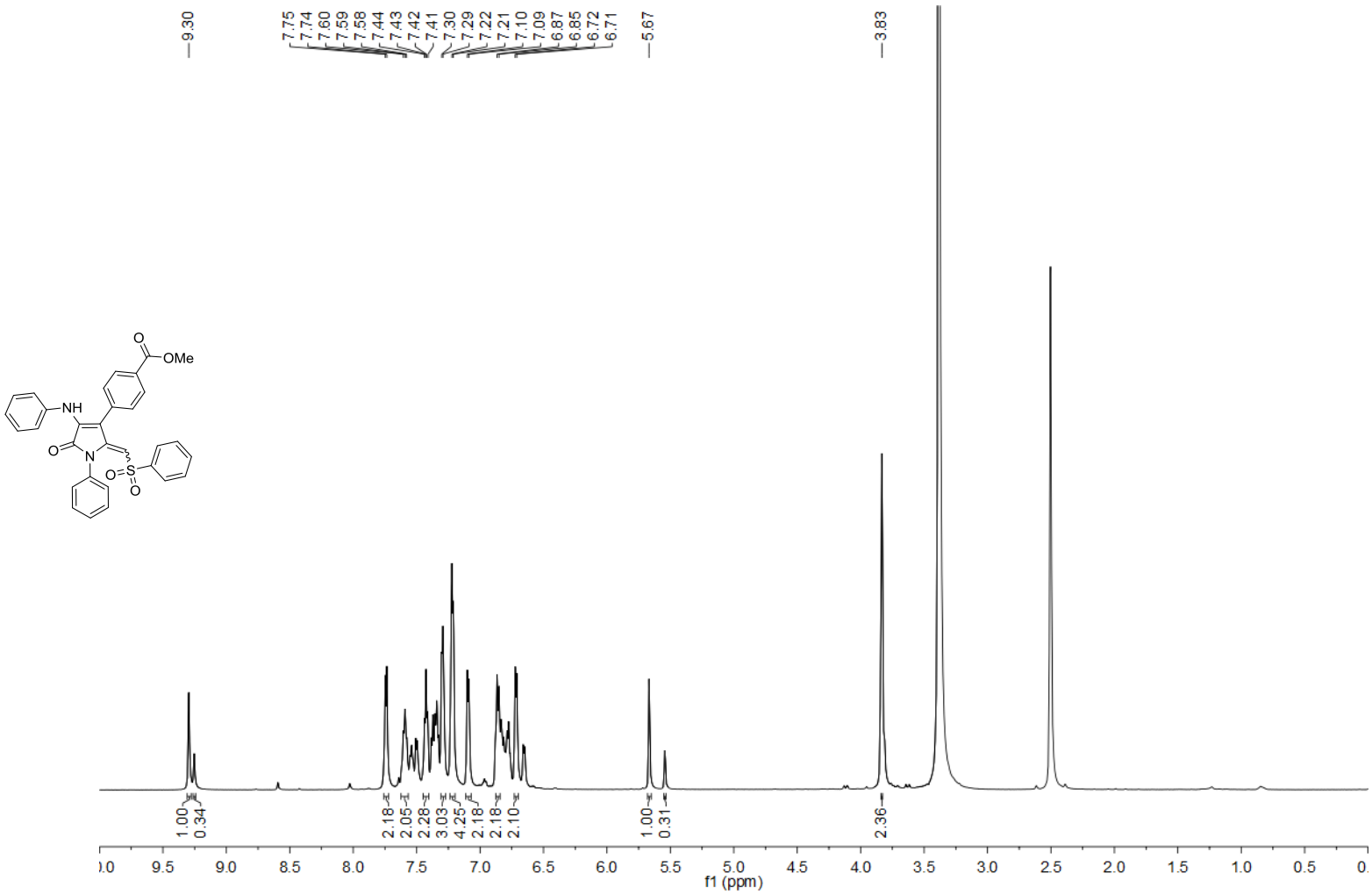


Figure S96. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **7g**

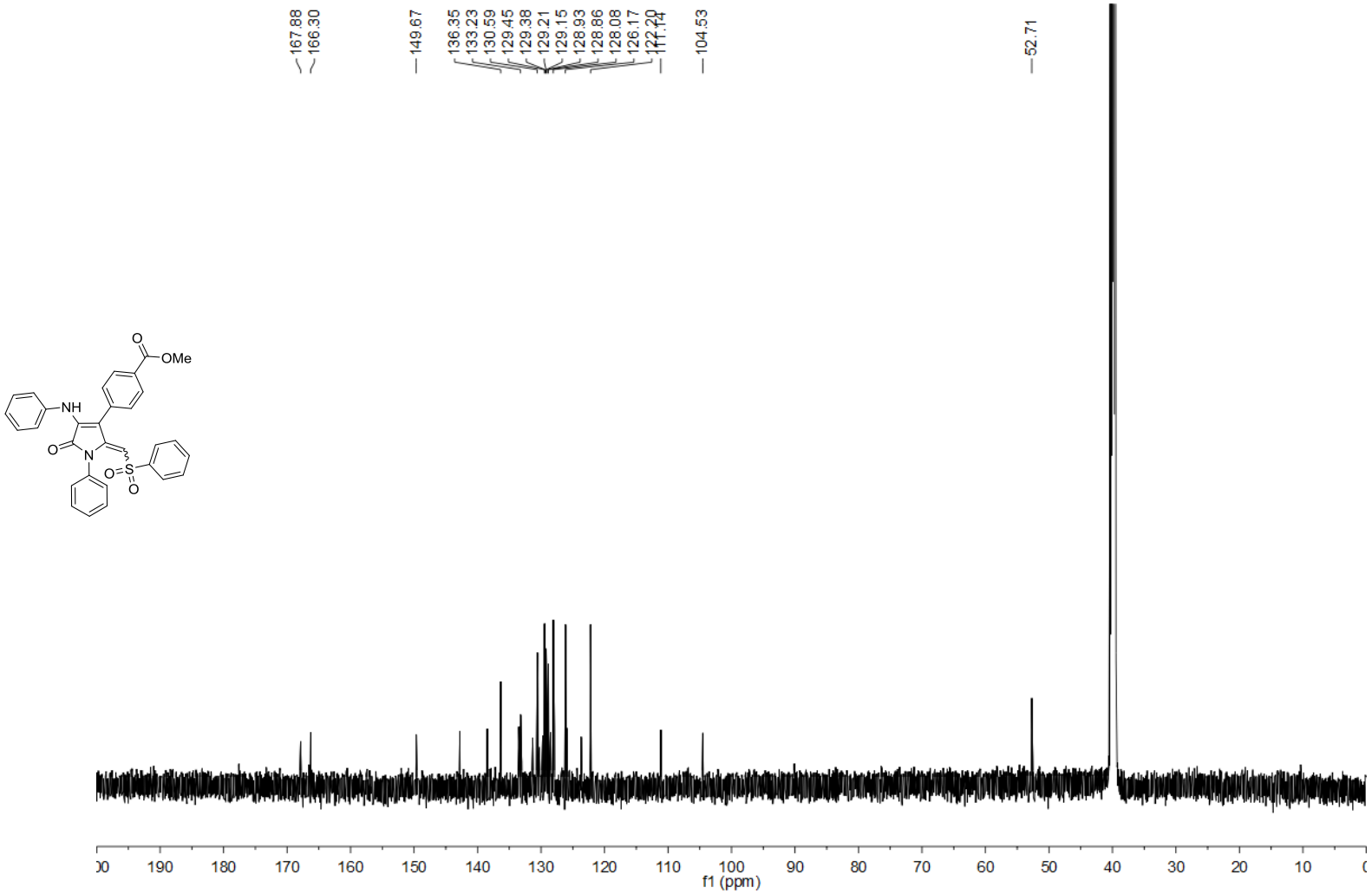


Figure S97. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **7g**

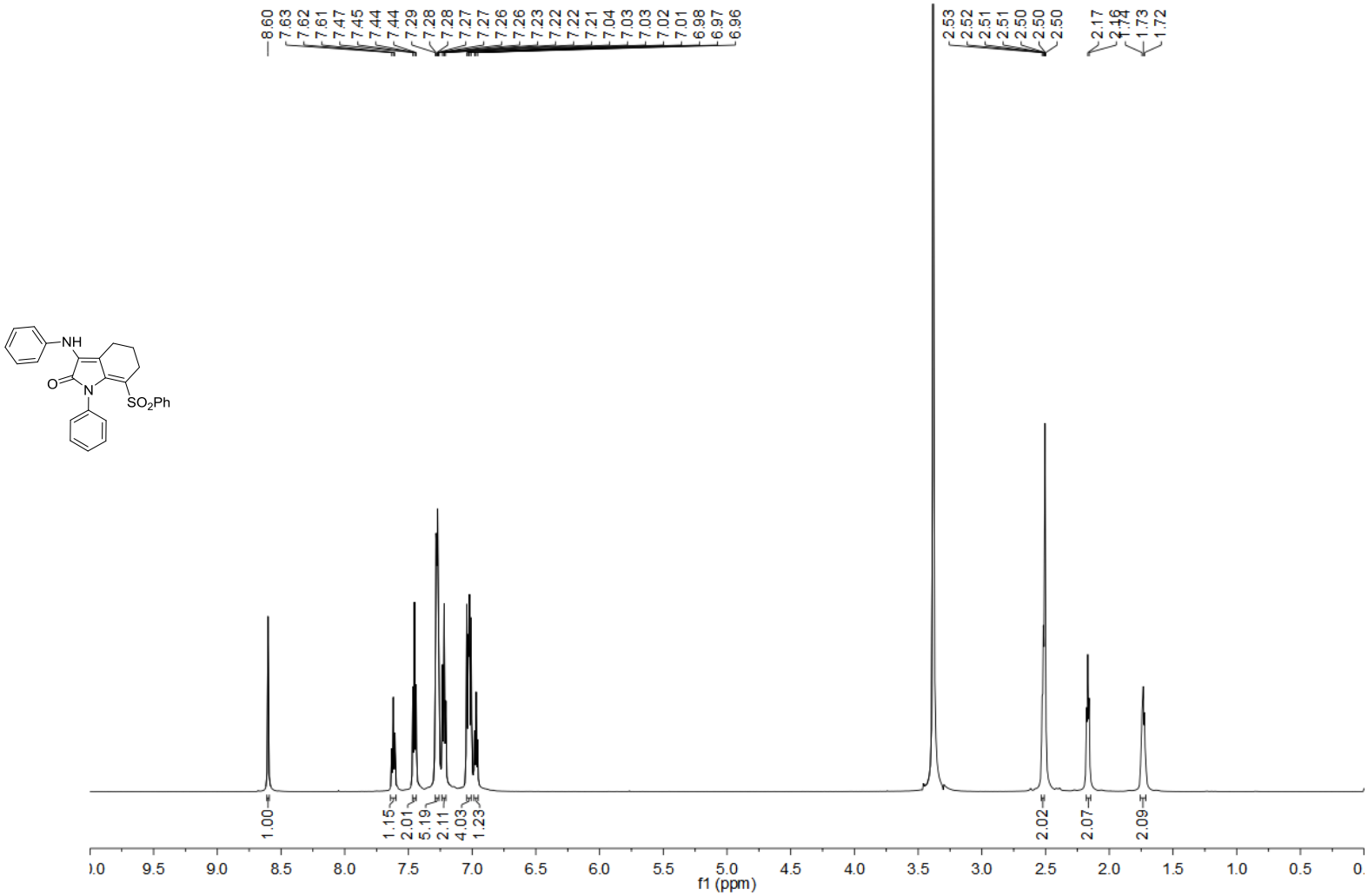


Figure S98. ^{1}**H NMR** (600 MHz, DMSO-d_6) spectra of compound **8a**

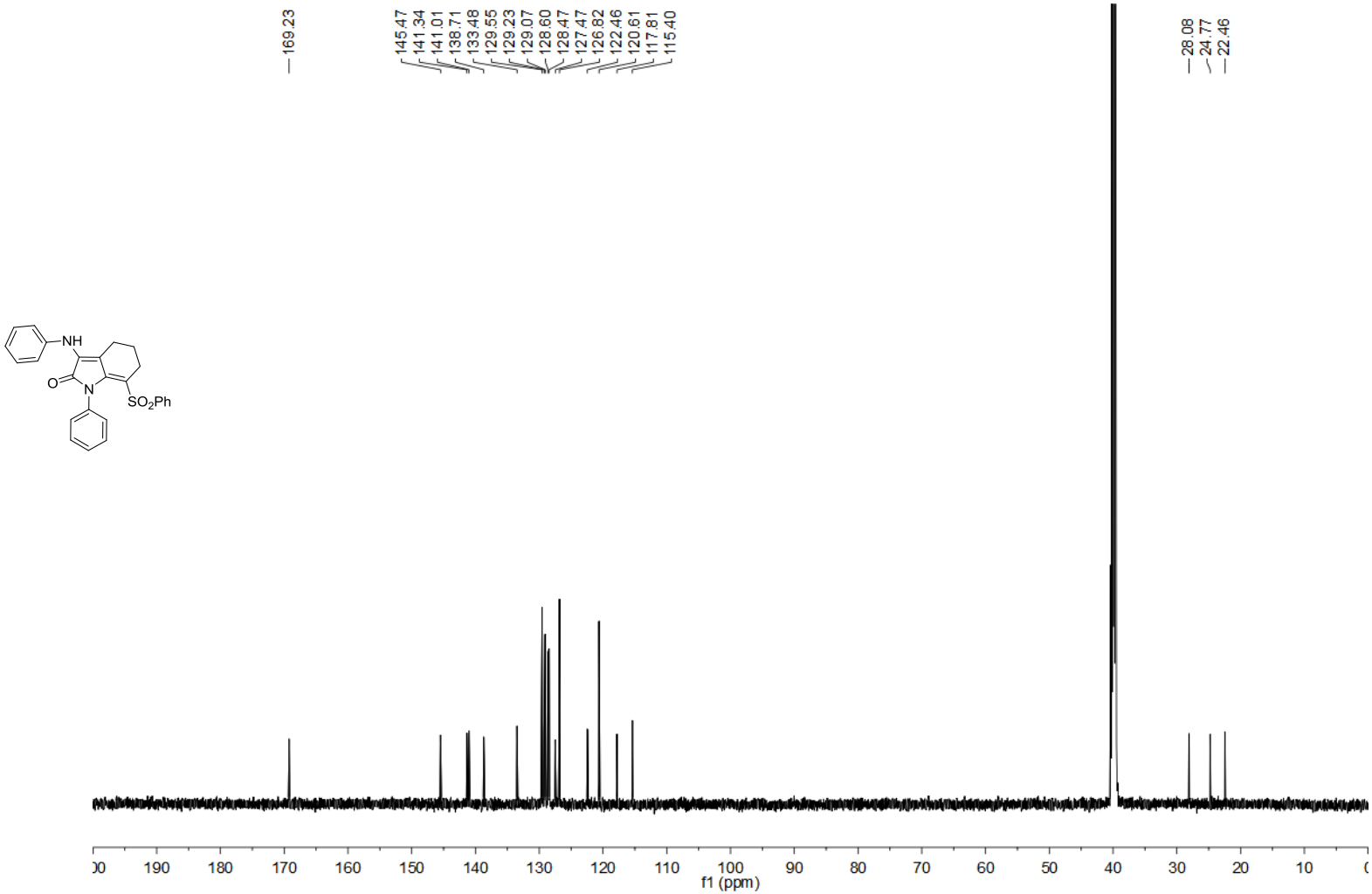


Figure S99. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **8a**

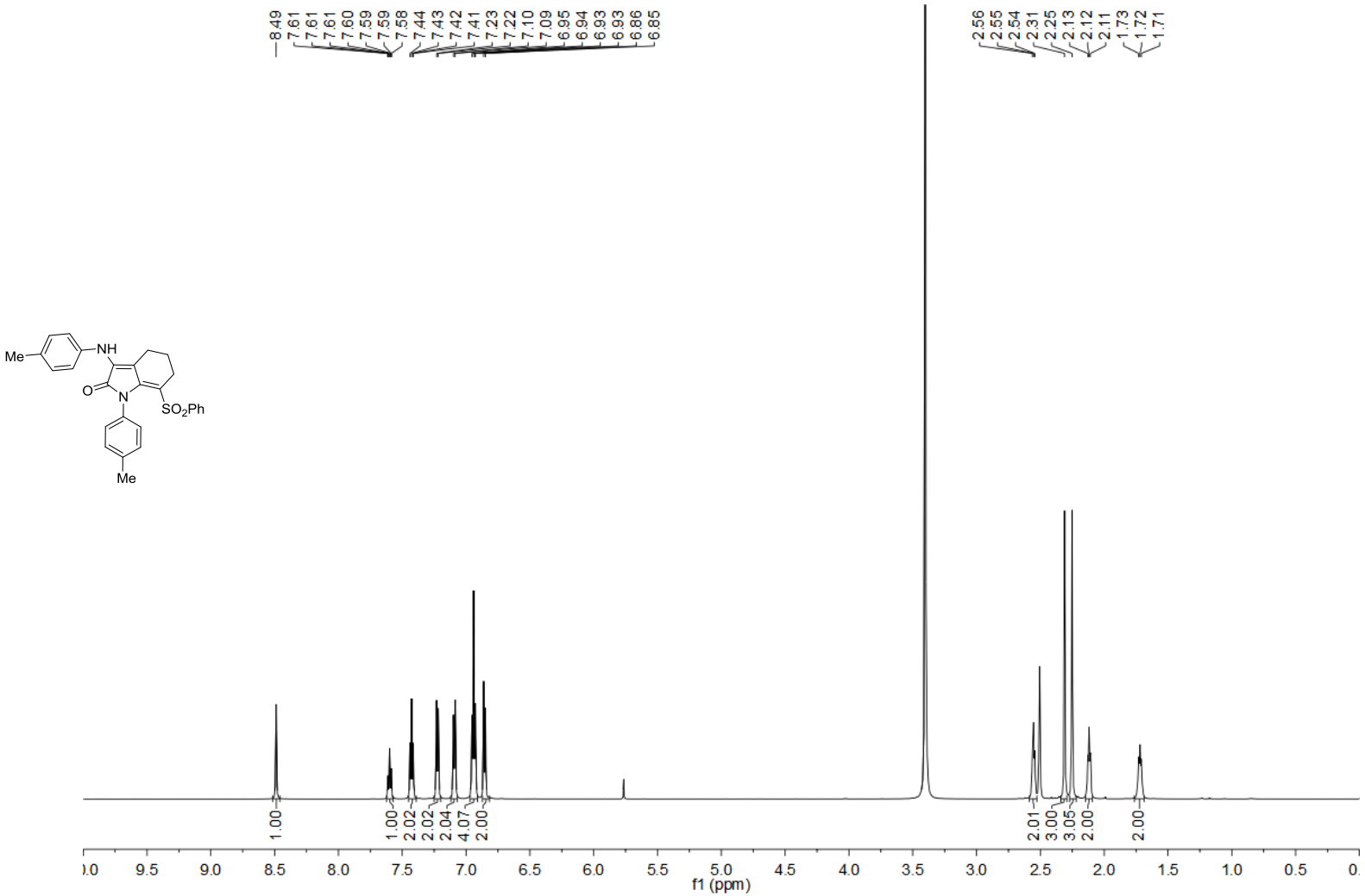


Figure S100. [1]**H NMR** (600 MHz, DMSO-d_6) spectra of compound **8b**

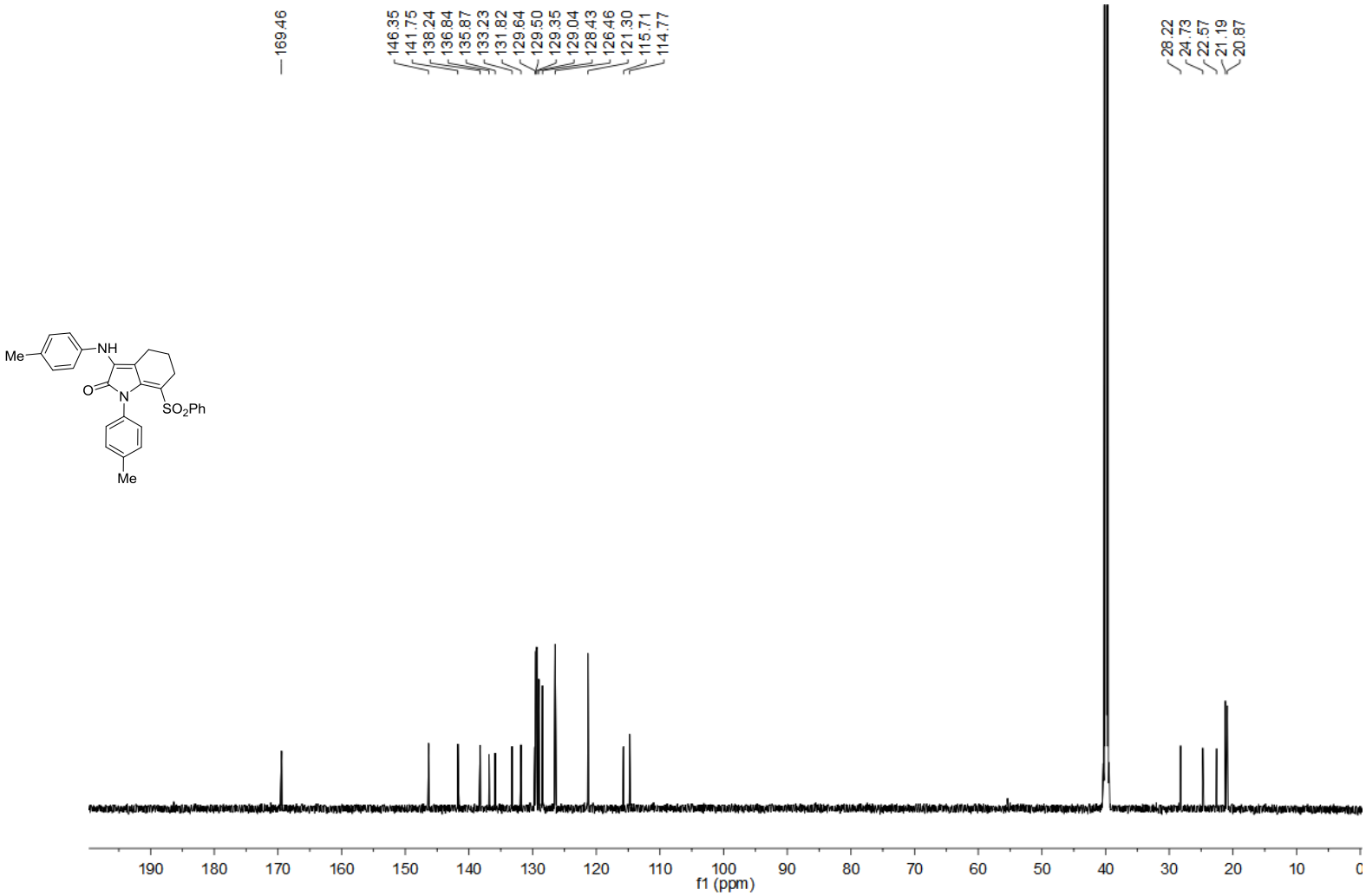


Figure S101. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **8b**

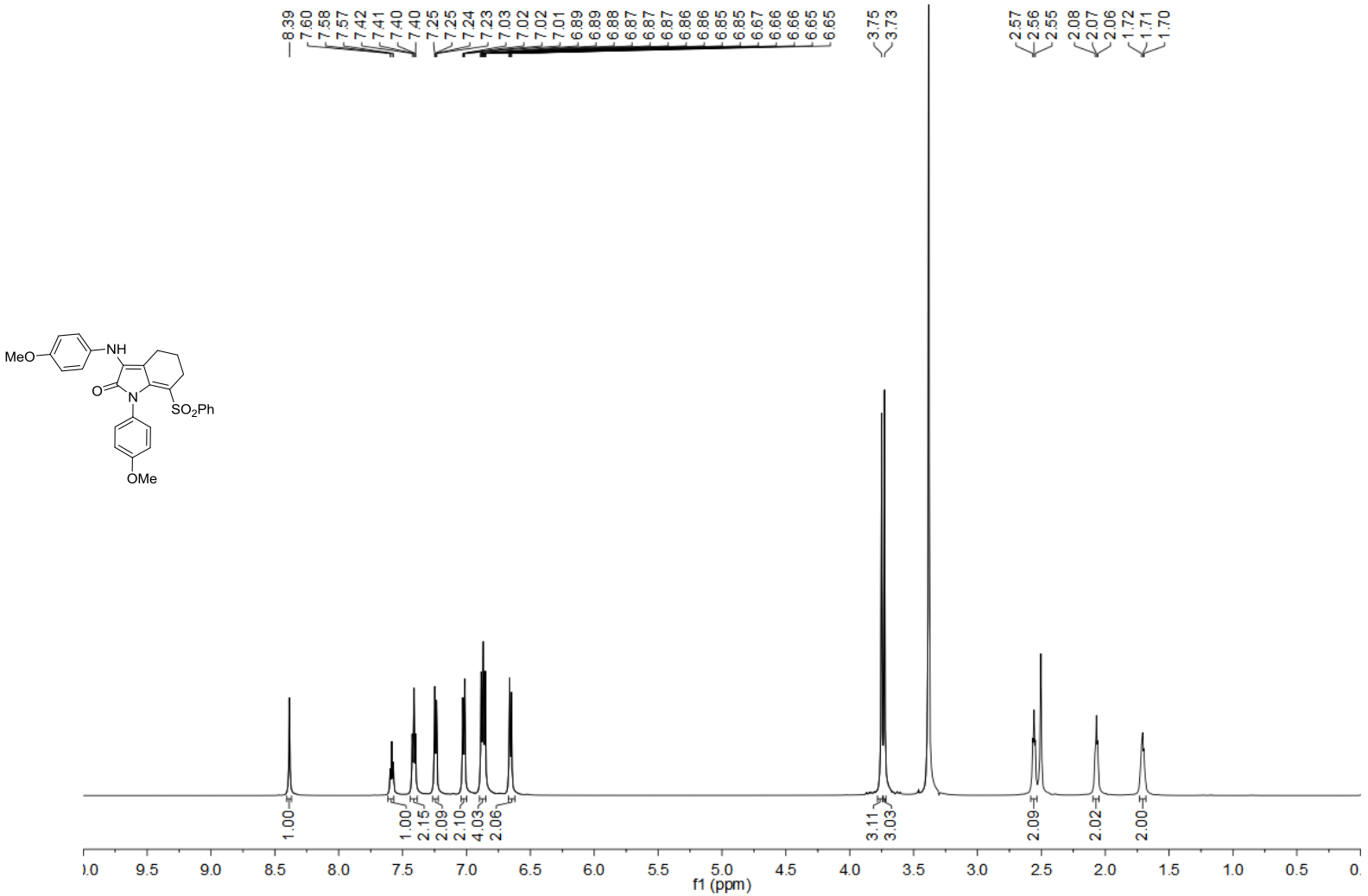


Figure S102. [1]**H NMR** (600 MHz, DMSO-d_6) spectra of compound **8c**

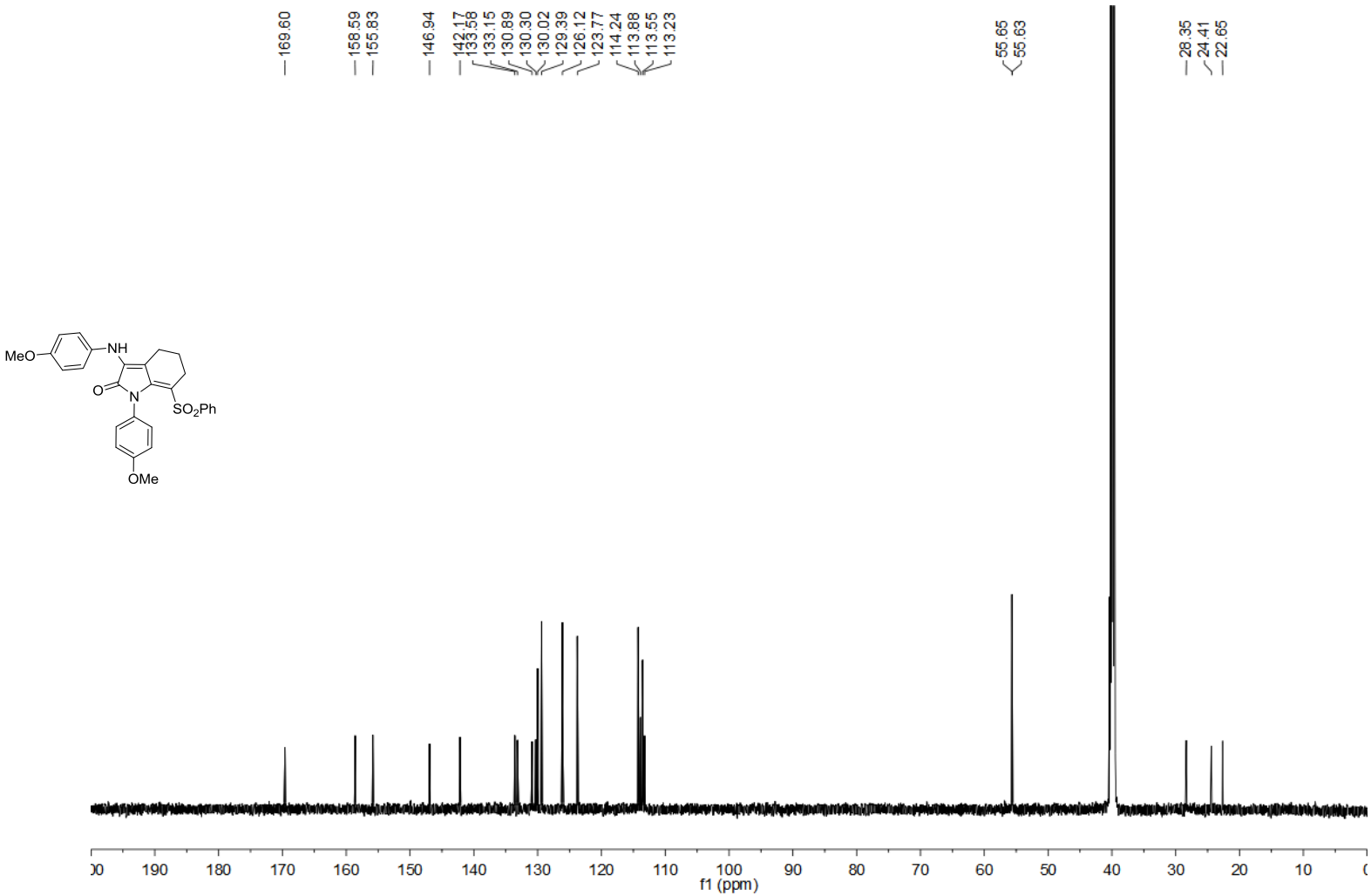


Figure S103. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **8c**

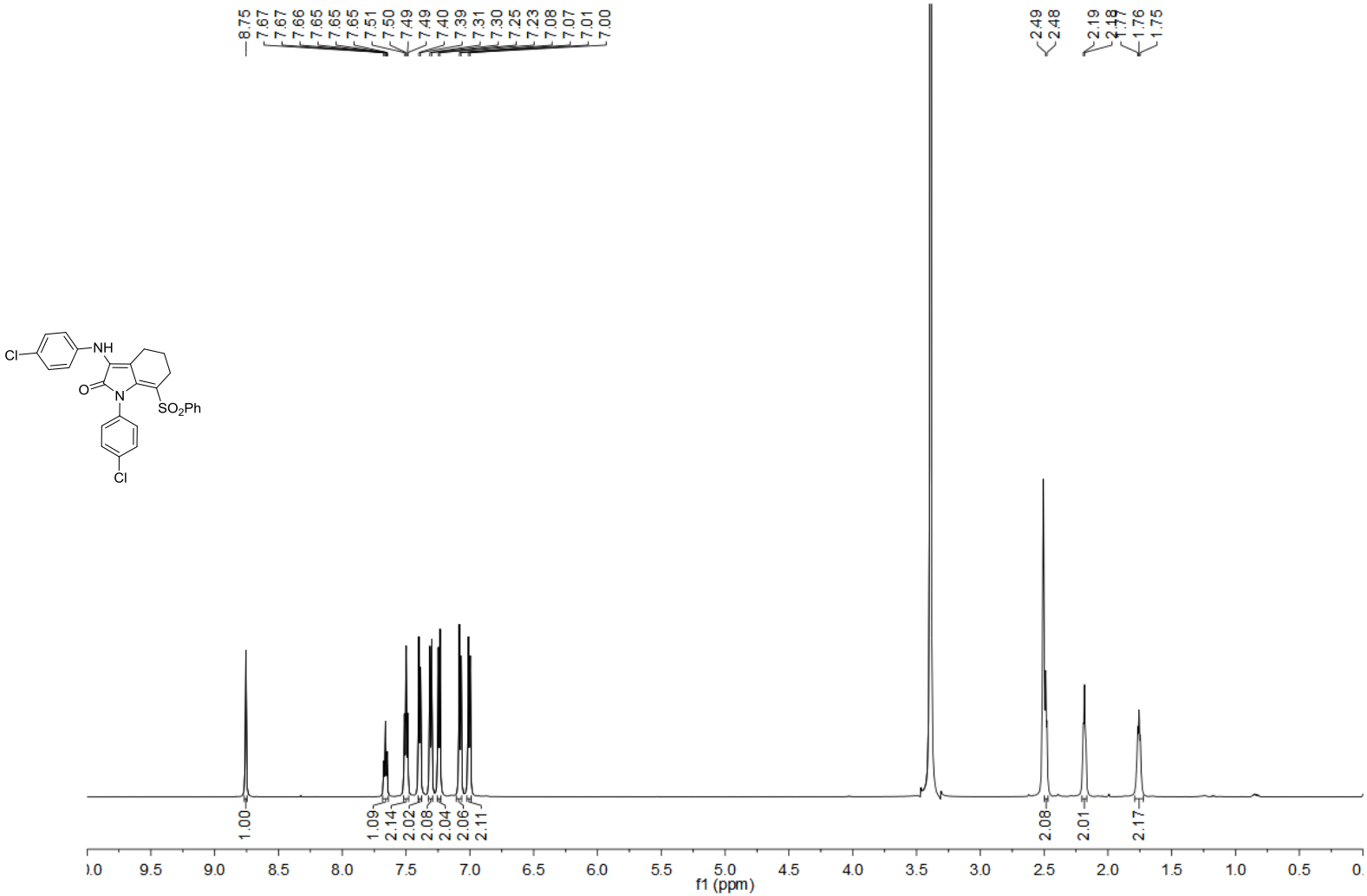


Figure S104. [1]**H NMR** (600 MHz, DMSO-d_6) spectra of compound **8d**

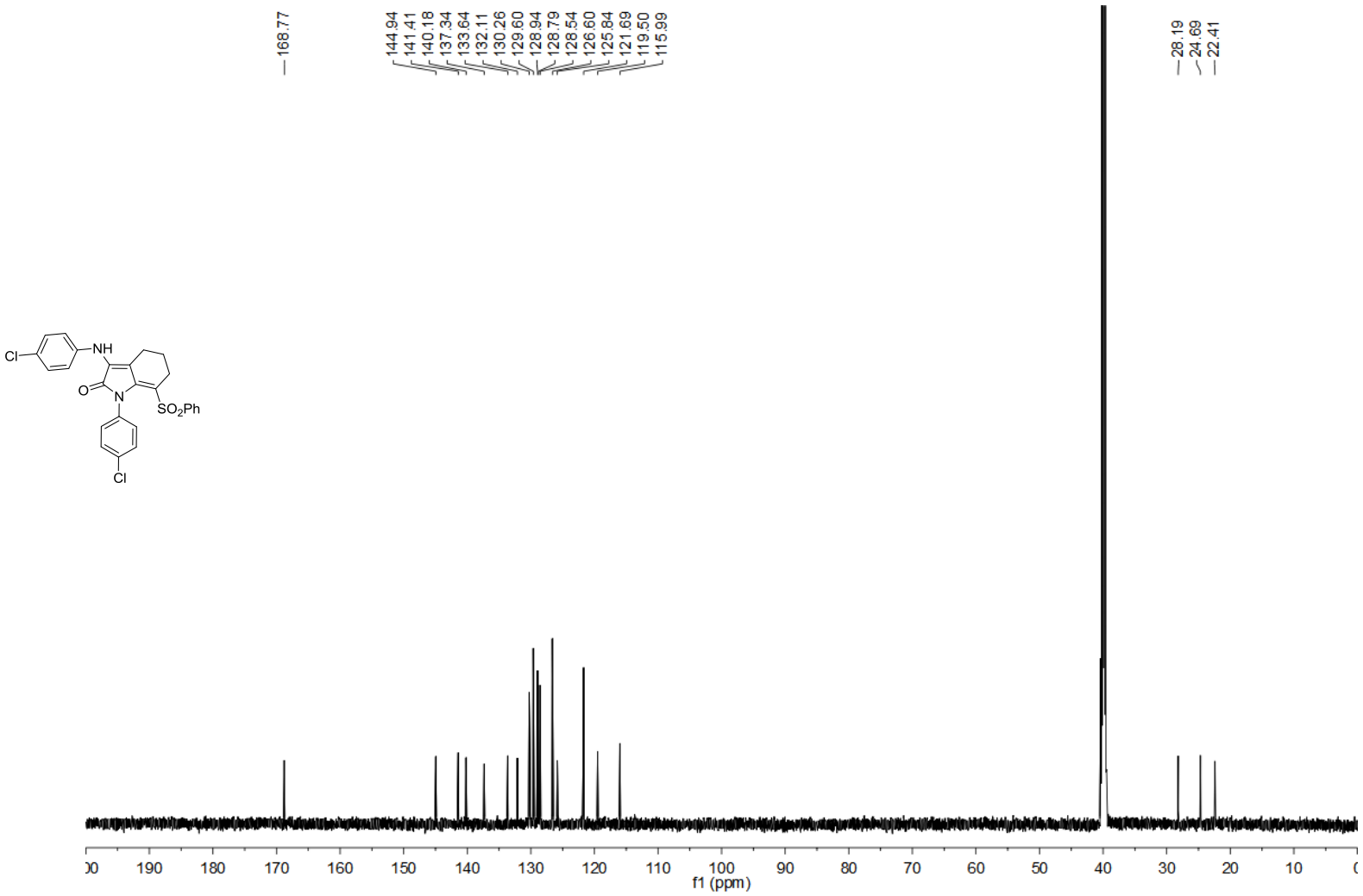


Figure S105. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **8d**

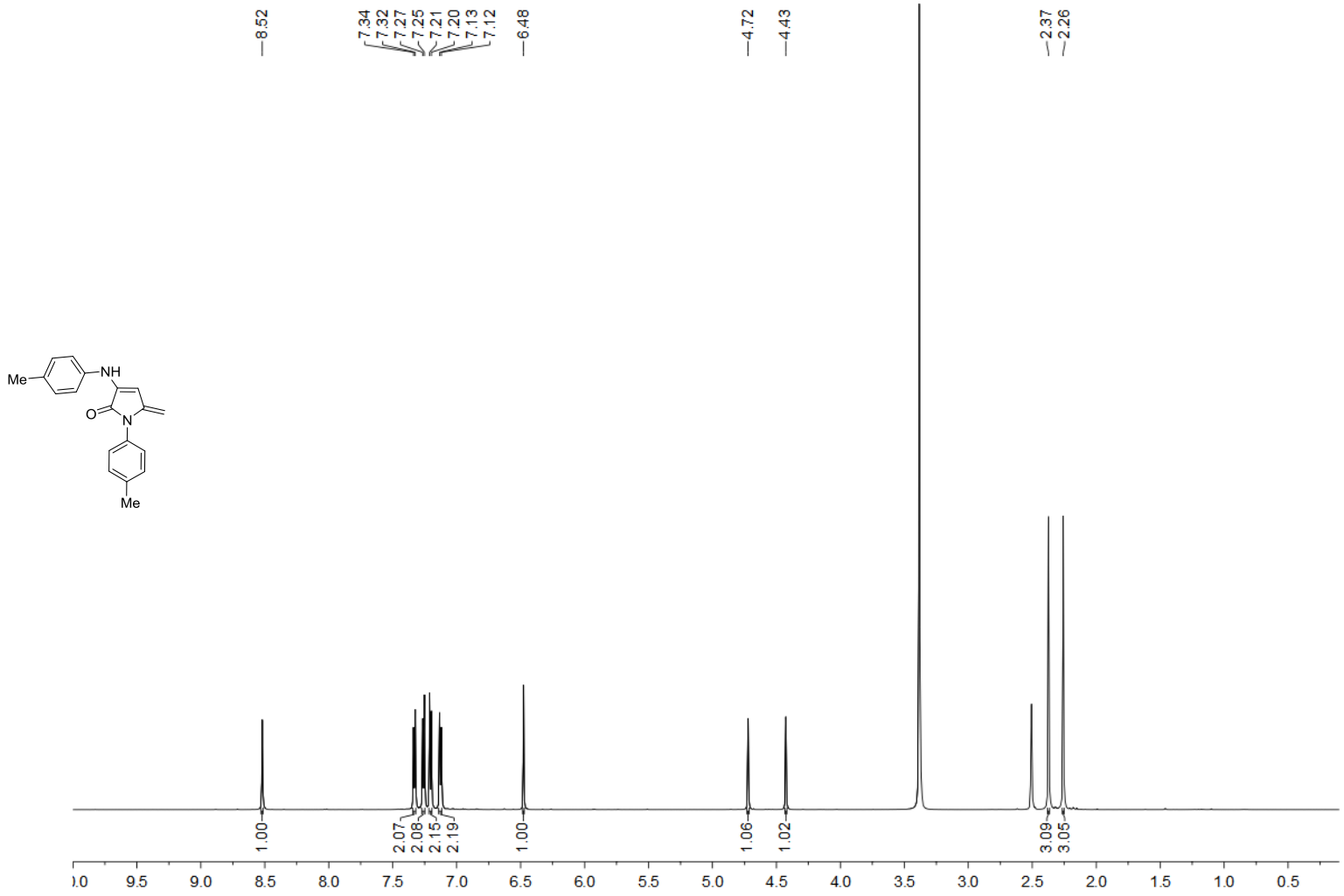


Figure S106. **^{1}H NMR** (600 MHz, DMSO-d_6) spectra of compound **10**

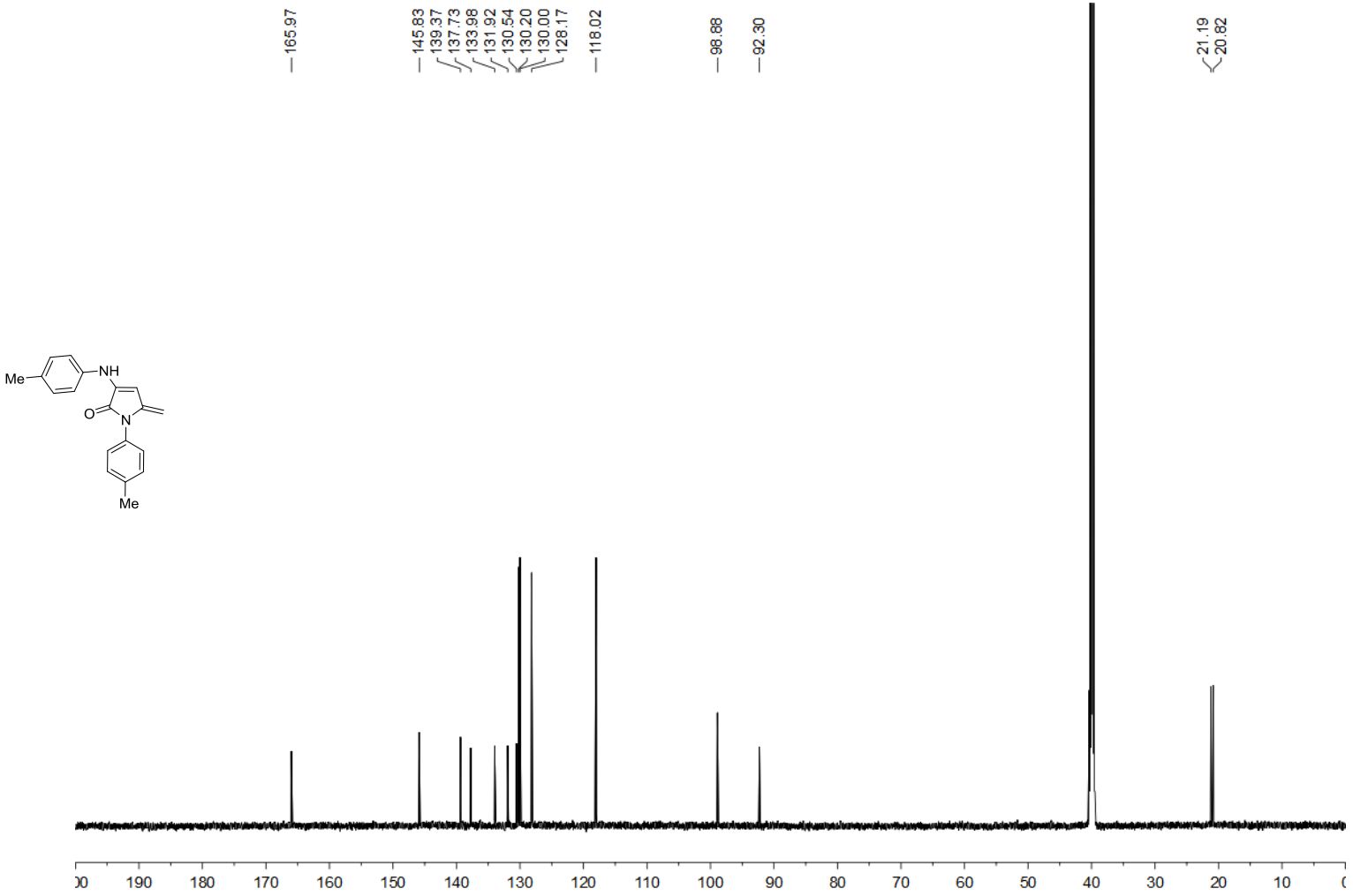


Figure S107. ^{13}C NMR (150 MHz, DMSO-d_6) spectra of compound **10**

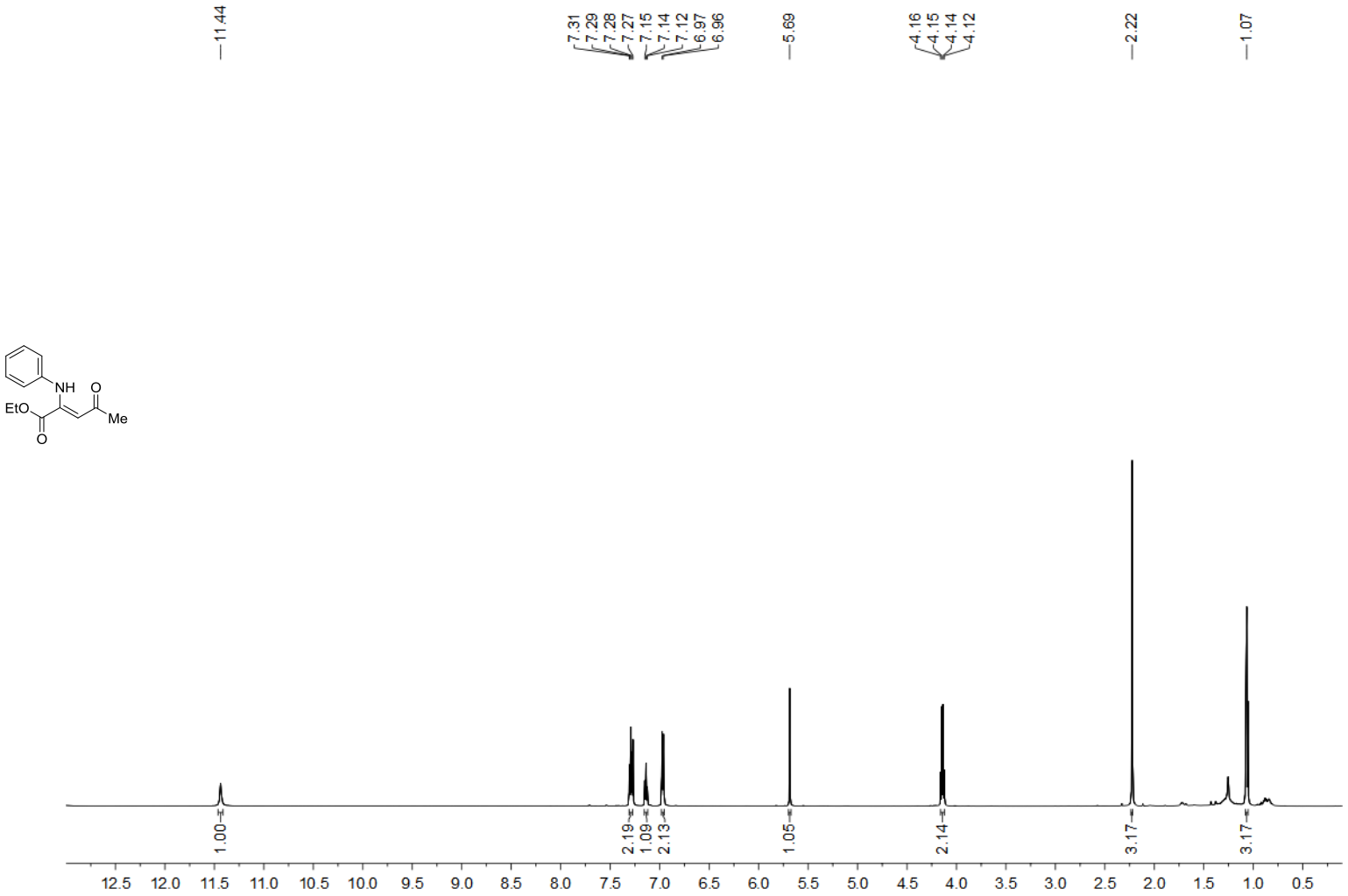


Figure S108. ^{1}**H NMR** (600 MHz, $CDCl_3$) spectra of compound **12**

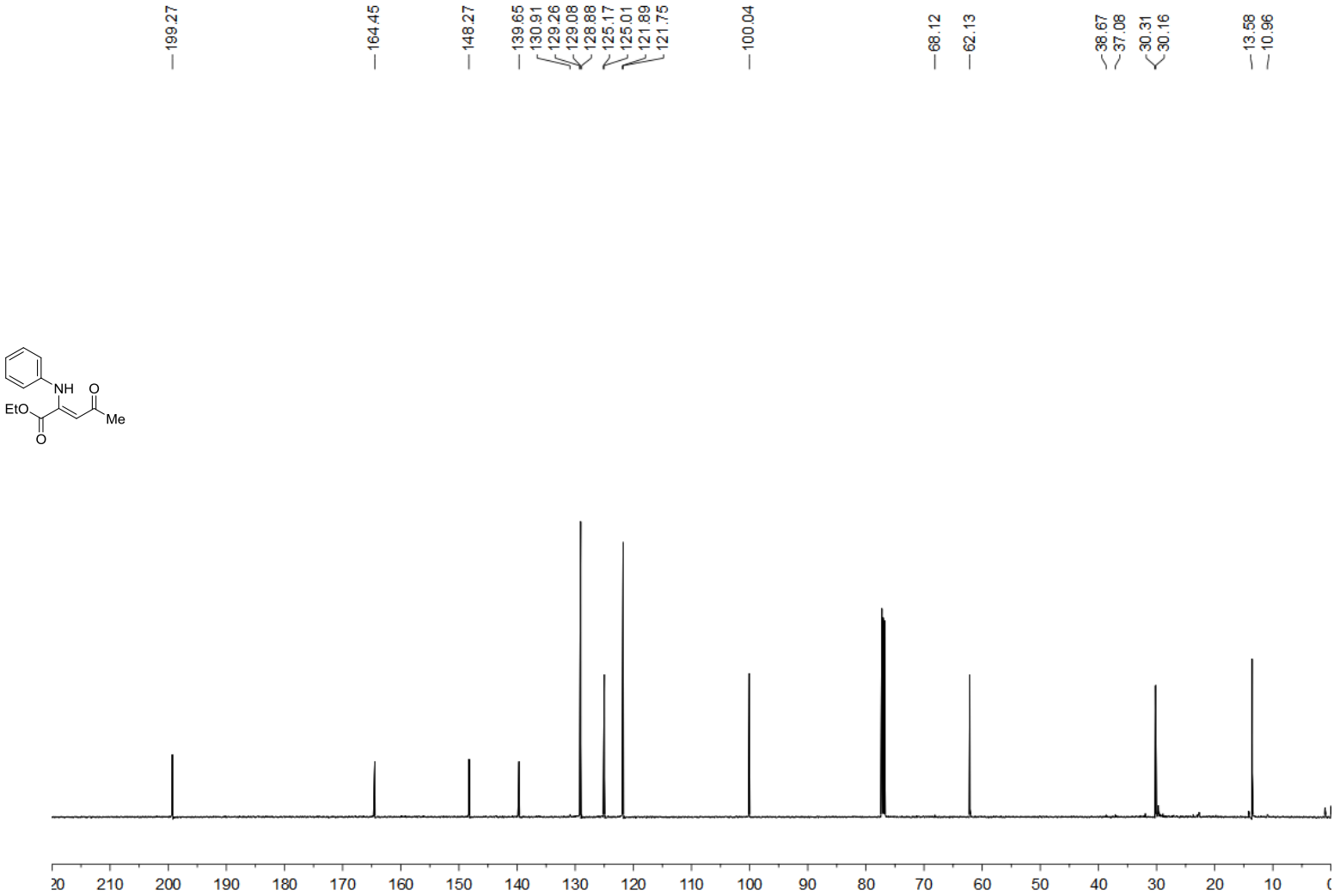


Figure S109. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **12**

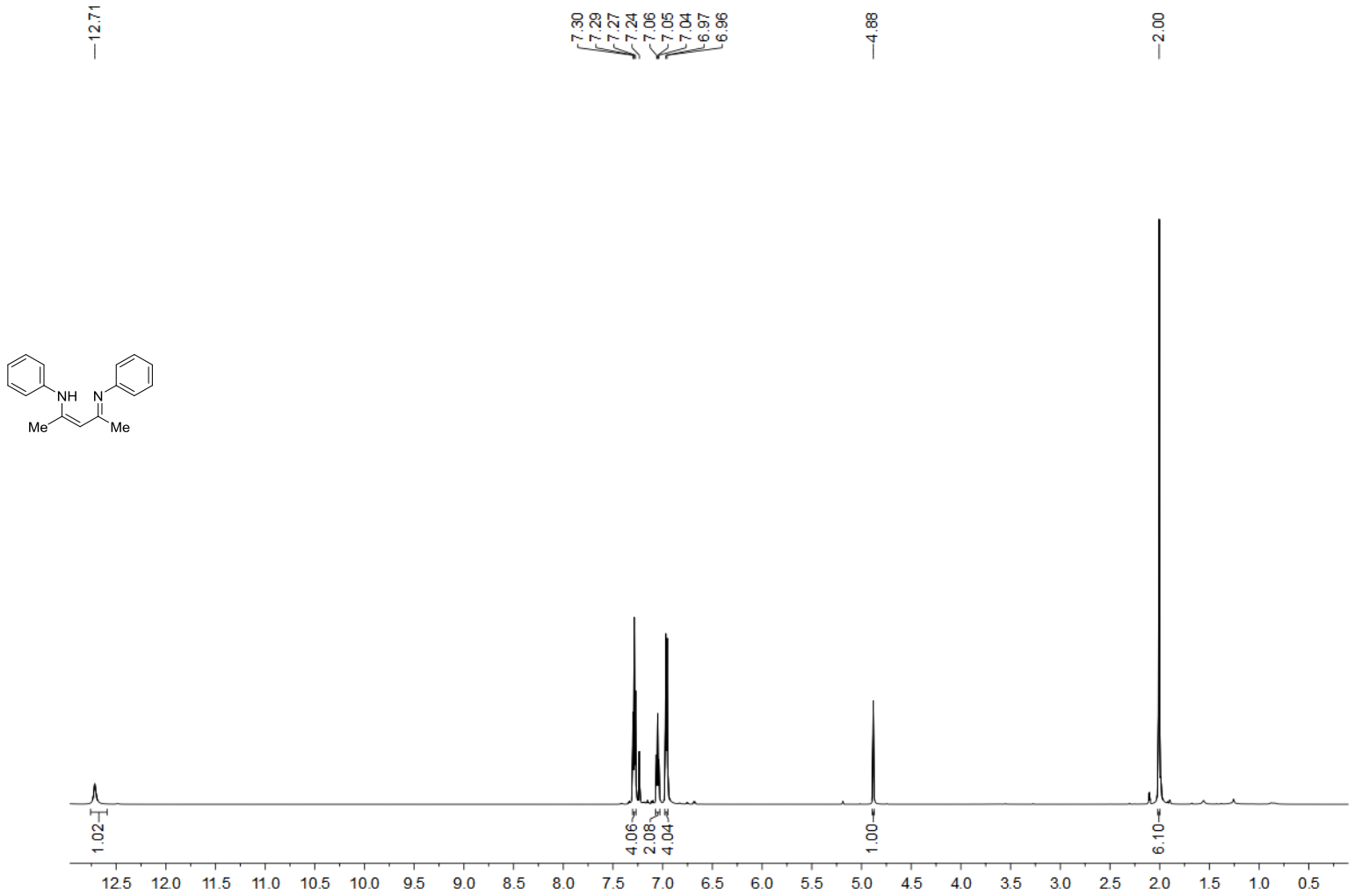


Figure S110. **^{1}H NMR** (600 MHz, $CDCl_3$) spectra of compound **13**

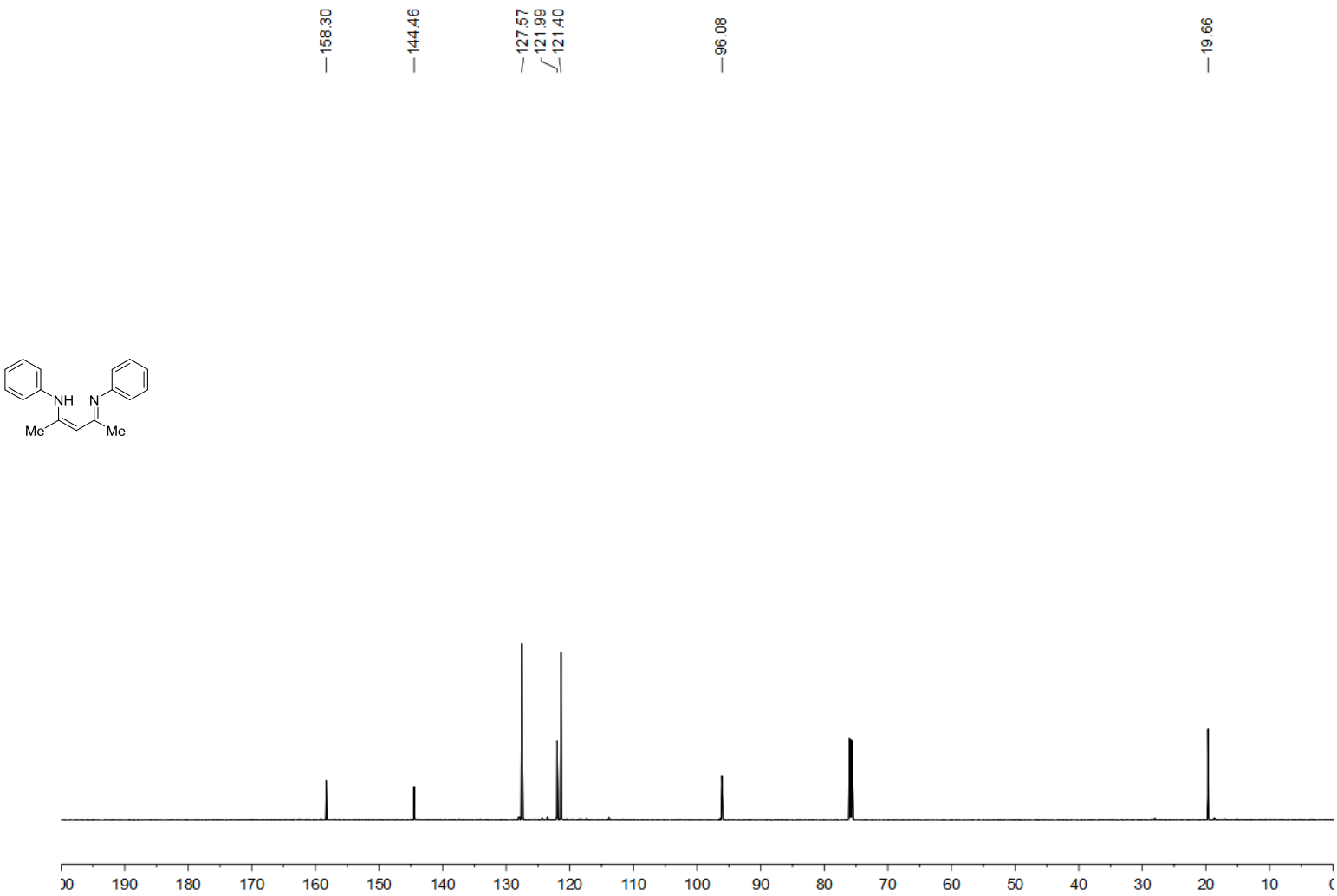


Figure S111. ^{13}C NMR (150 MHz, $CDCl_3$) spectra of compound **13**

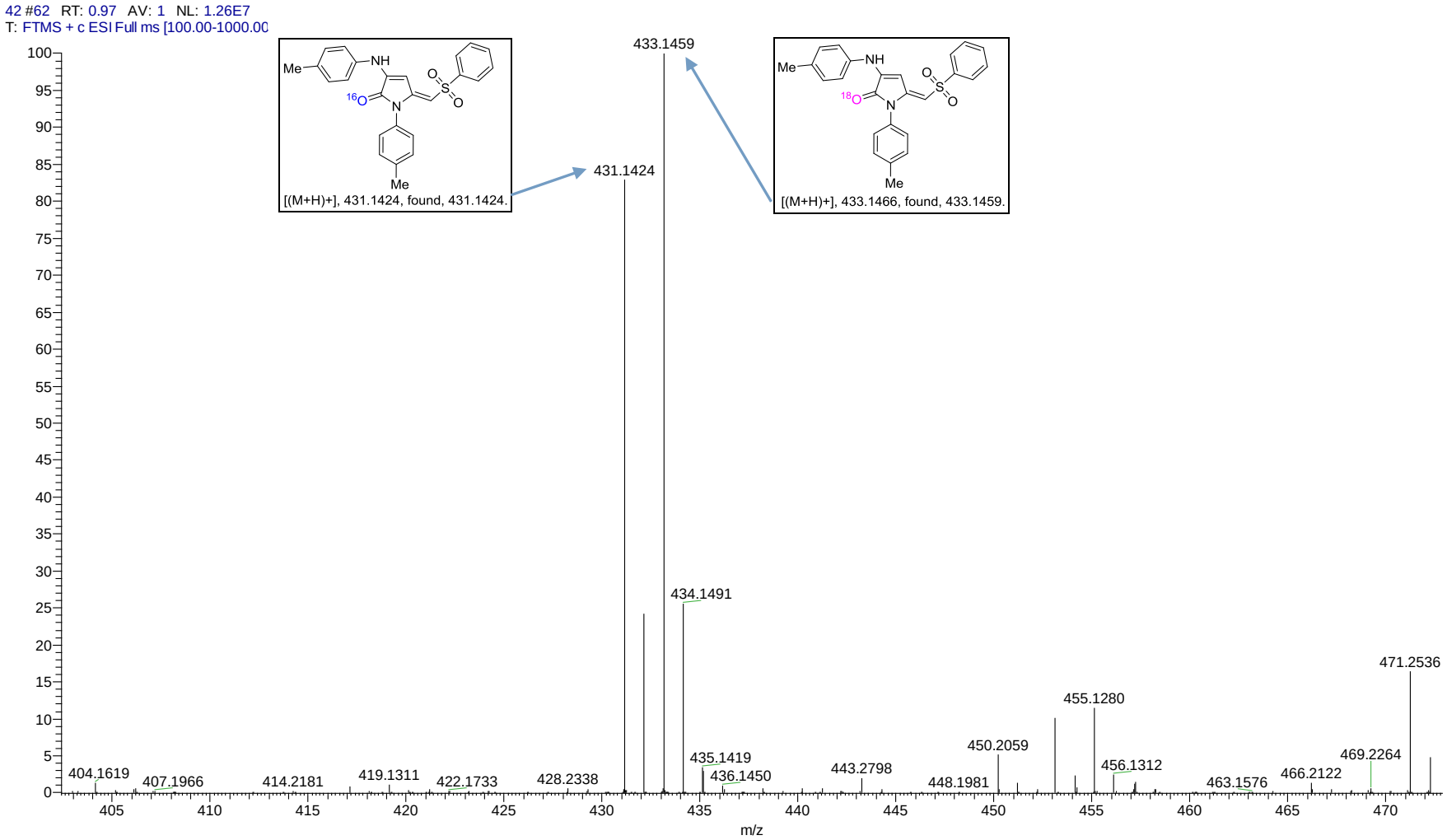


Figure S112. HRMS spectrum of compound **4a**/**4a-O^{18}**

8. References and notes.

1. Puente, Á.; Ofial, A. R.; Mayr, H. Nucleophilic reactivities of bis-acceptor-substituted benzyl anions. *Eur. J. Org. Chem.* **2017**, 1196.
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 2145010 contain the supplementary crystallographic data for compound **4a**. These data can be obtained free of charge from The Cambridge Crystallographic Data Center *via* www.ccdc.cam.ac.uk/data_request/cif.