Electronic Supplementary Material (ESI) for Organic Chemistry Frontiers.

Supporting Information

Photocatalytic Cross-Dehydrogenative Coupling Reaction toward the Synthesis of *N*,*N*-Disubstituted Hydrazides and Their Bromides

Lvyin Zheng,*† Xiaoya Zhuo,† Yihan Wang, Xiaoying Zou, Yumei Zhong, and Wei Guo*

Key Laboratory of Organo-Pharmaceutical Chemistry of Jiangxi Province, Gannan Normal University, Ganzhou 341000, China

Fax: (+86) 0797-8393536; *E-mail: zhenglvyin@126.com; guoweigw@126.com

†L.Z. and X.Z. contributed equally to this work.

List of Contents

1. General Methods

Melting points were tested using a digital melting point instrument and are uncorrected. IR spectra data were measured on an infrared spectrometer using potassium bromide pellets. ^{1}H and $^{13}C\{^{1}H\}$ NMR spectra were carried out on a 400 MHz NMR spectrometer with DMSO-d_6 or $CDCl_3$ as solutions. Gas chromatography-mass spectrometry (GC-MS) was collected using electron ionization. High-resolution mass spectrometry (HRMS) data were obtained on a high-resolution mass spectrometer (LCMS-ITTOF). The crystal data were recorded on a diffractometer Rigaku Oxford diffraction supernova dual source, Cu at Zero equipped with an AtlasS2 CCD using Cu Kα radiation (1.54178 Å) using a scan mode. The reaction was performed on the photoreaction instrument (WP-TEC-1020L, WATTCAS, China) with a heating mantle and a condenser system. The distance from the light source to the irradiation vessel is 5 mm. The thin layer chromatography (TLC) and column chromatography were prepared on commercially available 100-400 mesh silica gel. *N*,*N*-Disubstituted hydrazines was prepared according to our previous reports.[1] The other starting materials, reagents and solvents were purchased commercially. Unless otherwise noted, all purchased products were used without further purification.

2. General Procedure for the Synthesis of *N*,*N*-Disubstituted Hydrazide Bromides.

N,*N*-Disubstituted hydrazines (0.3 mmol), aldehydes (0.1 mmol), $Ir[dF(CF_3)ppy]_2(dtbpy)PF_6$ (1 mol%), TBAB (35 mol%), CCl_3Br (1.5 equiv.), and CH_3CN (1 mL, 0.1 M) were successively added into a quartz reaction tube with a stir bar. The reaction was performed under the irradiation of 10 W blue LEDs (410 nm) at room temperature for 12 h. After evaporating the solvent under reduced pressure, the crude residue was purified by chromatography on a silica gel for separation using petroleum ether/ethyl acetate (5:1, v/v) as eluent to provide the desired product.

3. General Procedure for the Synthesis of *N*,*N*-Disubstituted Hydrazides.

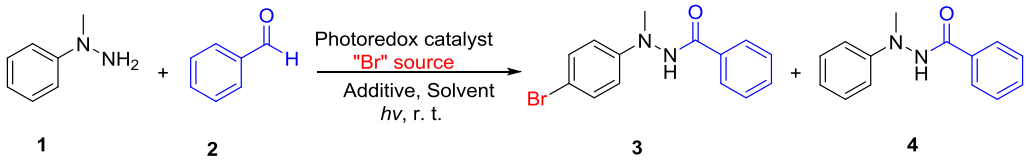
N,*N*-Disubstituted hydrazines (0.3 mmol), aldehydes (0.1 mmol), $Ir[dF(CF_3)ppy]_2(dtbpy)PF_6$ (1 mol%), CCl_3Br (1.5 equiv.), and DMF (1 mL, 0.1 M) were successively added into a quartz reaction tube with a stir bar. The reaction was performed under the irradiation of 10 W blue LEDs (410 nm) at room temperature for 6 h. After evaporating the solvent under reduced pressure, the crude residue was purified by chromatography on a silica gel for separation using petroleum ether/ethyl acetate (5:1, v/v) as eluent to provide the desired product.

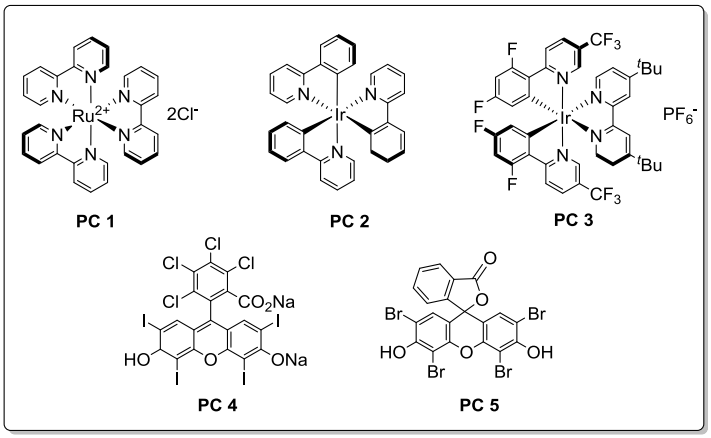
4. Optimization of the Reaction Conditions

In order to verify our idea and investigate the optimal conditions of this photocatalytic synthesis of *N*,*N*-disubstituted hydrazides from the model substrates, 1-methyl-1-phenylhydrazine **1** (0.3 mmol, 3.0 equiv.), benzaldehyde **2** (0.1 mmol, 1.0 equiv.), we initiated our study using 1 mol% tris(2,2'-bipyridine)ruthenium dichloride (**PC 1**) as photocatalyst in CH_3CN (1 mL, 0.1 M) with the addition of CCl_3Br (0.15 mol, 1.5 equiv.) and tetrabutyl ammonium bromide (TBAB) (35 mol%) under a 10 W blue LED irradiation (410 nm) at room temperature for 12 h (Table S1, entry 1). To our delight, the desired *N*,*N*-disubstituted hydrazide bromide **3** was obtained in 68% yield with high regioselectivity. The structure of product **3** was characterized by X-ray crystal diffraction measurement (CCDC 2111900) (see Figure S1, Supporting Information). Inspired by the original result, we further evaluated other photocatalysts, such as *fac*-$Ir(ppy)_3$ (**PC 2**), $Ir[dF(CF_3)ppy]_2(dtbpy)PF_6$ (**PC 3**), rose Bengal (**PC 4**) and Eosin Y (**PC 5**), suggested that **PC 3** was the best, affording product **3** in 85% yield (Table S1, entries 2-5). When varying the reaction solvent from CH_3CN to ethyl acetate (EA), dichloroethane (DCE), toluene and dimethyl sulfoxide (DMSO), product **3** was obtained in 43-85% yields, and a trace of product **4** was detected by GC-MS (Table S1, entries 6-9). Furthermore, screening of other common organic solvents such as tetrahydrofuran (THF), dichloromethane (DCM), *N*,*N*-dimethyl formamide (DMF), and dimethyl acetamide (DMA) (Table S1, entries 10-13),

demonstrated that the transformation in DMF provided **4** in the best yield (Table S1, entry 12). The replacement of TBAB with tetrabutylammonium chloride (TBAC) or tetrabutylammonium iodide (TBAI), led to a detrimental effect on the formation of product **3** or **4** (Table S1, entries 14-15). The result of entry 14 also showed that the Br atom of product **3** derives from CCl_3Br, rather than the Br^- in TBAB. Control experiments revealed that photocatalysts, CCl_3Br, and light sources are essential to generate the desired product **3** or **4** (Table S1, entries 16-19). After evaluating a series of variables, including photocatalysts, solvents, light sources, and the amount of CCl_3Br and TBAB (see Tables S2-S5, Supporting Information), we selected the optimal conditions in entry 3 as the standard reaction conditions for the synthesis of *N*,*N*-disubstituted hydrazide bromides. When the reaction was carried out in the case of DMF for 6 h without the addition of TBAB, no product **3** was detected, and product **4** was obtained in 55% (Table S1, entry 20).

Table S1 Screening of Reaction Conditions[a]



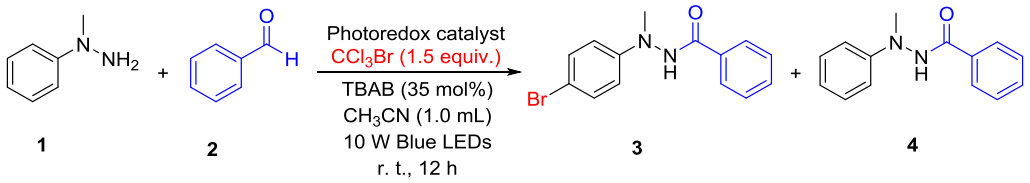


Entry	Photocatalyst	"Br" source	Additive	Solvent	Yield (3) (%)	Yield (4) (%)
1	**PC 1**	CCl_3Br	TBAB	CH_3CN	68	None
2	**PC 2**	CCl_3Br	TBAB	CH_3CN	70	None
3	**PC 3**	CCl_3Br	TBAB	CH_3CN	85	None
4	**PC 4**	CCl_3Br	TBAB	CH_3CN	43	None

5	**PC 5**	CCl_3Br	TBAB	CH_3CN	31	None
6	**PC 3**	CCl_3Br	TBAB	EA	85	Trace
7	**PC 3**	CCl_3Br	TBAB	DCE	76	Trace
8	**PC 3**	CCl_3Br	TBAB	toluene	43	Trace
9	**PC 3**	CCl_3Br	TBAB	DMSO	53	Trace
10	**PC 3**	CCl_3Br	TBAB	THF	56	40
11	**PC 3**	CCl_3Br	TBAB	DCM	14	43
12	**PC 3**	CCl_3Br	TBAB	DMF	7	48
13	**PC 3**	CCl_3Br	TBAB	DMA	17	30
14	**PC 3**	CCl_3Br	TBAC	CH_3CN	34	37
15	**PC 3**	CCl_3Br	TBAI	CH_3CN	Trace	40
16	/	CCl_3Br	TBAB	CH_3CN	Trace	None
17	**PC 3**	/	TBAB	CH_3CN	None	None
18[b]	**PC 3**	CCl_3Br	TBAB	CH_3CN	None	None
19[c]	**PC 3**	CCl_3Br	TBAB	CH_3CN	None	None
20[d]	**PC 3**	CCl_3Br	/	DMF	None	55

[a]Reaction conditions: 1-methyl-1-phenylhydrazine **1** (0.3 mmol), benzaldehyde **2** (0.1 mmol), photocatalyst (1 mol%), CCl_3Br (1.5 equiv.), additive (35 mol%) and solvent (1.0 mL), under irradiation with a 10 W blue LED (410 nm) at room temperature for 12 h in the open air. Isolated yields based on **2**. [b]Reaction was conducted in the dark at room temperature. [c]Reaction was conducted in the dark at 80 °C. [d]Reaction was conducted for 6 h.

Table S2 Optimization of Photocatalysts[a]

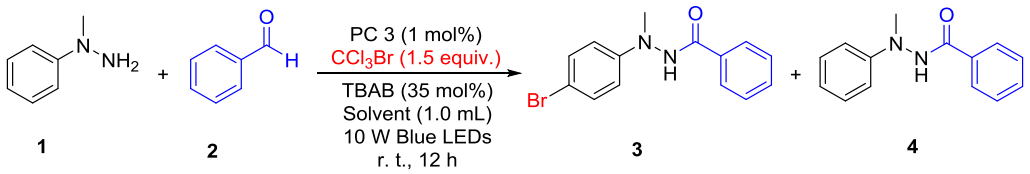


1 2 3 4

Entry	Photocatalyst	"Br" source	Additive	Solvent	Yield (3) (%)	Yield (4) (%)
1	**PC 1**	CCl_3Br	TBAB	CH_3CN	68	None
2	**PC 2**	CCl_3Br	TBAB	CH_3CN	70	None
3	**PC 3**	CCl_3Br	TBAB	CH_3CN	85	None
4	**PC 4**	CCl_3Br	TBAB	CH_3CN	43	None
5	**PC 5**	CCl_3Br	TBAB	CH_3CN	31	None
6	/	CCl_3Br	TBAB	CH_3CN	Trace	None

[a]Reaction conditions: 1-methyl-1-phenylhydrazine **1** (0.3 mmol), benzaldehyde **2** (0.1 mmol), photocatalyst (1 mol%), CCl_3Br (1.5 equiv.), TBAB (35 mol%) and CH_3CN (1.0 mL), under irradiation with a 10 W blue LED (410 nm) at room temperature for 12 h in the open air. Isolated yields based on **2**.

Table S3 Optimization of Solvents[a]

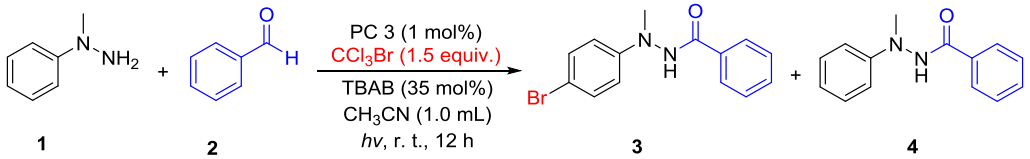


1 2 3 4

Entry	Photocatalyst	"Br" source	Additive	Solvent	Yield (3) (%)	Yield (4) (%)
1	**PC 3**	CCl_3Br	TBAB	CH_3CN	85	None
2	**PC 3**	CCl_3Br	TBAB	EA	85	Trace
3	**PC 3**	CCl_3Br	TBAB	DCE	76	Trace
4	**PC 3**	CCl_3Br	TBAB	toluene	43	Trace
5	**PC 3**	CCl_3Br	TBAB	DMSO	53	Trace
6	**PC 3**	CCl_3Br	TBAB	THF	56	40
7	**PC 3**	CCl_3Br	TBAB	DCM	14	43
8	**PC 3**	CCl_3Br	TBAB	DMF	7	48
9	**PC 3**	CCl_3Br	TBAB	DMA	17	30

[a]Reaction conditions: 1-methyl-1-phenylhydrazine **1** (0.3 mmol), benzaldehyde **2** (0.1 mmol), PC **3** (1 mol%), CCl_3Br (1.5 equiv.), TBAB (35 mol%) and solvent (1.0 mL), under irradiation with a 10 W blue LED (410 nm) at room temperature for 12 h in the open air. Isolated yields based on **2**.

Table S4 Optimization of Light Sources[a]

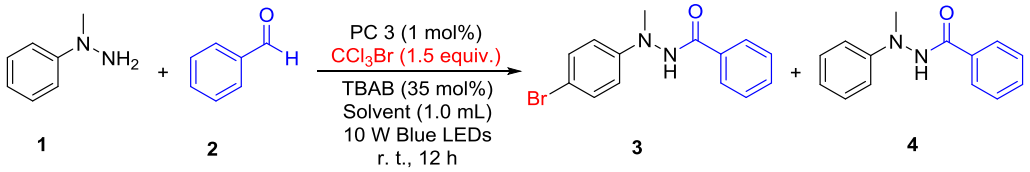


1 2 3 4

Entry	Photocatalyst	Light Sources (nm)	Yield (3) (%)	Yield (4) (%)
1	**PC 3**	365	85	None
2	**PC 3**	410	85	None
3	**PC 3**	440	76	None
4	**PC 3**	520	None	None
5	**PC 3**	565	None	None
6	**PC 3**	6000-6500k	72	None

[a]Reaction conditions: 1-methyl-1-phenylhydrazine **1** (0.3 mmol), benzaldehyde **2** (0.1 mmol), PC **3** (1 mol%), CCl_3Br (1.5 equiv.), TBAB (35 mol%) and CH_3CN (1.0 mL), irradiation with different light sources at room temperature for 12 h in the open air. Isolated yields based on **2**.

Table S5 Optimization of the Amount of CCl_3Br and TBAB



1 2 3 4

Entry	Photocatalyst	"Br" source (equiv.)	Additive	Yield (3) (%)	Yield (4) (%)
1	**PC 3**	CCl_3Br (1.5)	TBAB (15%)	52	None
2	**PC 3**	CCl_3Br (1.5)	TBAB (150%)	85	None
3	**PC 3**	CCl_3Br (0.8)	TBAB (35%)	48	None
4	**PC 3**	CCl_3Br (2.5)	TBAB (35%)	62	None
5	**PC 3**	CCl_3Br (3.5)	TBAB (35%)	49	None

[a]Reaction conditions: 1-methyl-1-phenylhydrazine **1** (0.3 mmol), benzaldehyde **2** (0.1 mmol), PC **3** (1 mol%), CCl_3Br, TBAB and CH_3CN (1.0 mL), under irradiation with a 10 W blue LED (410 nm) at room temperature for 12 h in the open air. Isolated yields based on **2**.

5. The Crystal Structure of 3

The crystal growth procedure: Compound **3** (20 mg) was dissolved into 1 mL of ethyl acetate, and then petroleum ether (2 mL) was added into the mixture. The mixture was evaporated slowly at room temperature to obtain crystal **3**.

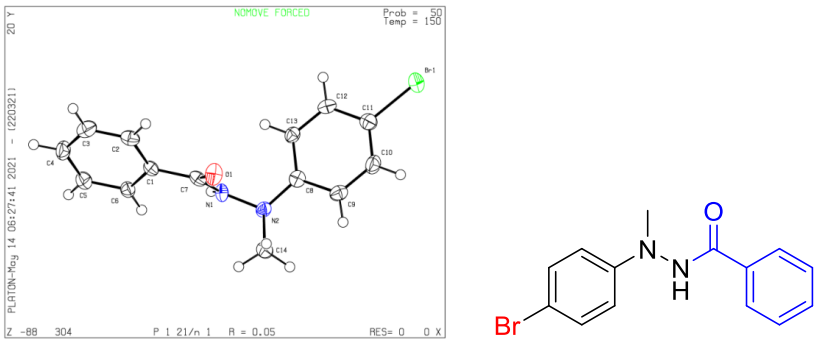


Figure S1 The Crystal Structure of **4a**

The CCDC number of **3** is 2111900, the detail information please see **3**.cif document.

CIF files of 3

checkCIF/PLATON report

Structure factors have been supplied for datablock(s) 304

THIS REPORT IS FOR GUIDANCE ONLY. IF USED AS PART OF A REVIEW PROCEDURE FOR PUBLICATION, IT SHOULD NOT REPLACE THE EXPERTISE OF AN EXPERIENCED CRYSTALLOGRAPHIC REFEREE.

No syntax errors found. CIF dictionary Interpreting this report

Datablock: 304

```
Bond precision:  C-C = 0.0067 A          Wavelength=0.71073

Cell:            a=5.7585(5)     b=26.542(2)          c=8.5839(9)
                 alpha=90        beta=94.793(10)      gamma=90
Temperature:     150 K

                Calculated                Reported
Volume          1307.4(2)                 1307.4(2)
Space group     P 21/n                    P 1 21/n 1
Hall group      -P 2yn                    -P 2yn
Moiety formula  C14 H13 Br N2 O           C14 H13 Br N2 O
Sum formula     C14 H13 Br N2 O           C14 H13 Br N2 O
Mr              305.16                    305.17
Dx,g cm-3       1.550                     1.550
Z               4                         4
Mu (mm-1)       3.133                     3.133
F000            616.0                     616.0
F000'           615.08
h,k,lmax        6,31,10                   6,31,10
Nref            2299                      2297
Tmin,Tmax       0.643,0.708               0.261,1.000
Tmin'           0.619

Correction method= # Reported T Limits: Tmin=0.261 Tmax=1.000
AbsCorr = MULTI-SCAN

Data completeness= 0.999          Theta(max)= 24.998

R(reflections)= 0.0534( 1638)     wR2(reflections)= 0.1219( 2297)

S = 1.047                 Npar= 164
```

Alert level C

PLAT341_ALERT_3_C	Low Bond Precision on C-C Bonds	0.00669	Ang.
PLAT906_ALERT_3_C	Large K Value in the Analysis of Variance	3.862	Check
PLAT911_ALERT_3_C	Missing FCF Refl Between Thmin & STh/L= 0.595	2	Report

Alert level G

PLAT007_ALERT_5_G	Number of Unrefined Donor-H Atoms	1	Report
PLAT909_ALERT_3_G	Percentage of I>2sig(I) Data at Theta(Max) Still	40%	Note
PLAT910_ALERT_3_G	Missing # of FCF Reflection(s) Below Theta(Min).	1	Note
PLAT941_ALERT_3_G	Average HKL Measurement Multiplicity	2.3	Low
PLAT978_ALERT_2_G	Number C-C Bonds with Positive Residual Density.	1	Info

0 **ALERT level A** = Most likely a serious problem - resolve or explain
0 **ALERT level B** = A potentially serious problem, consider carefully
3 **ALERT level C** = Check. Ensure it is not caused by an omission or oversight
5 **ALERT level G** = General information/check it is not something unexpected

0 ALERT type 1 CIF construction/syntax error, inconsistent or missing data
1 ALERT type 2 Indicator that the structure model may be wrong or deficient
6 ALERT type 3 Indicator that the structure quality may be low
0 ALERT type 4 Improvement, methodology, query or suggestion
1 ALERT type 5 Informative message, check

It is advisable to attempt to resolve as many as possible of the alerts in all categories. Often the minor alerts point to easily fixed oversights, errors and omissions in your CIF or refinement strategy, so attention to these fine details can be worthwhile. In order to resolve some of the more serious problems it may be necessary to carry out additional measurements or structure refinements. However, the purpose of your study may justify the reported deviations and the more serious of these should normally be commented upon in the discussion or experimental section of a paper or in the "special_details" fields of the CIF. checkCIF was carefully designed to identify outliers and unusual parameters, but every test has its limitations and alerts that are not important in a particular case may appear. Conversely, the absence of alerts does not guarantee there are no aspects of the results needing attention. It is up to the individual to critically assess their own results and, if necessary, seek expert advice.

Publication of your CIF in IUCr journals

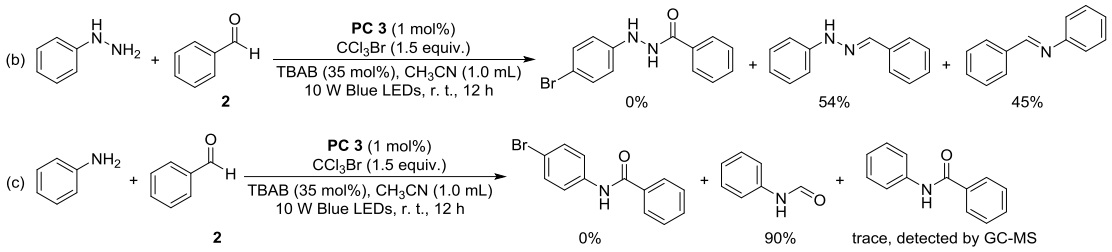
A basic structural check has been run on your CIF. These basic checks will be run on all CIFs submitted for publication in IUCr journals (*Acta Crystallographica*, *Journal of Applied Crystallography*, *Journal of Synchrotron Radiation*); however, if you intend to submit to *Acta Crystallographica Section C* or *E* or *IUCrData*, you should make sure that full publication checks are run on the final version of your CIF prior to submission.

Publication of your CIF in other journals

Please refer to the *Notes for Authors* of the relevant journal for any special instructions relating to CIF submission.

PLATON version of 22/03/2021; check.def file version of 19/03/2021

6. The Substrate Scope for Phenylhydrazine and Aniline



Scheme S1 The Substrate Scope for Phenylhydrazine and Aniline

When compound **1** was replaced with compound phenylhydrazine, no desired compound was detected. Moreover, the reaction of compound aniline with **2** failed to yield the desired product (Scheme S1, b). The above two experiments demonstrate that *N*-substituted hydrazine is an essential substrate for the transformation.

7. HRMS of 79

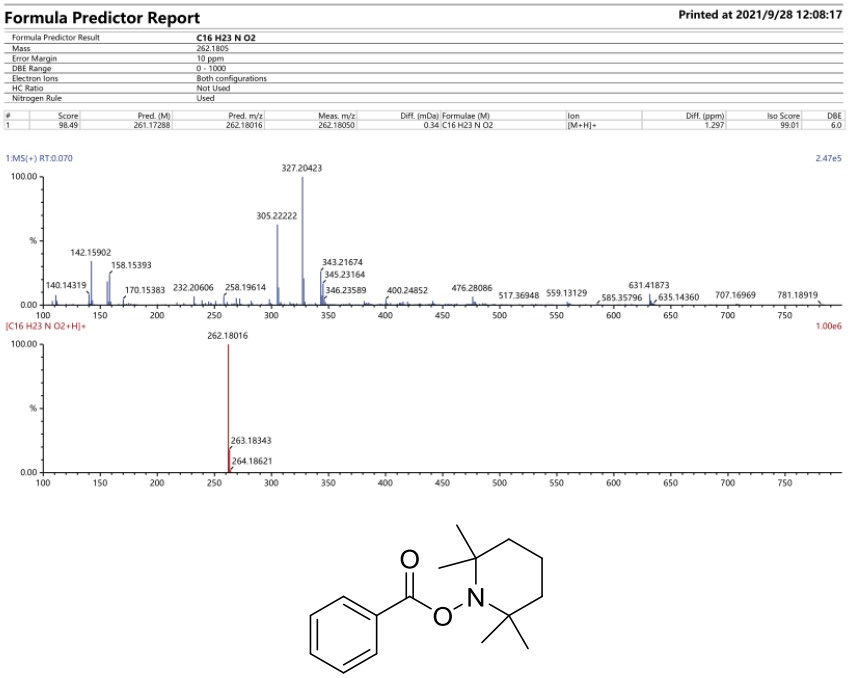


Figure S2 HRMS of **79**

HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{16}H_{24}NO_2$ 262.1802, found 262.1805.

8. GCMS of *N*-phenylbenzamide

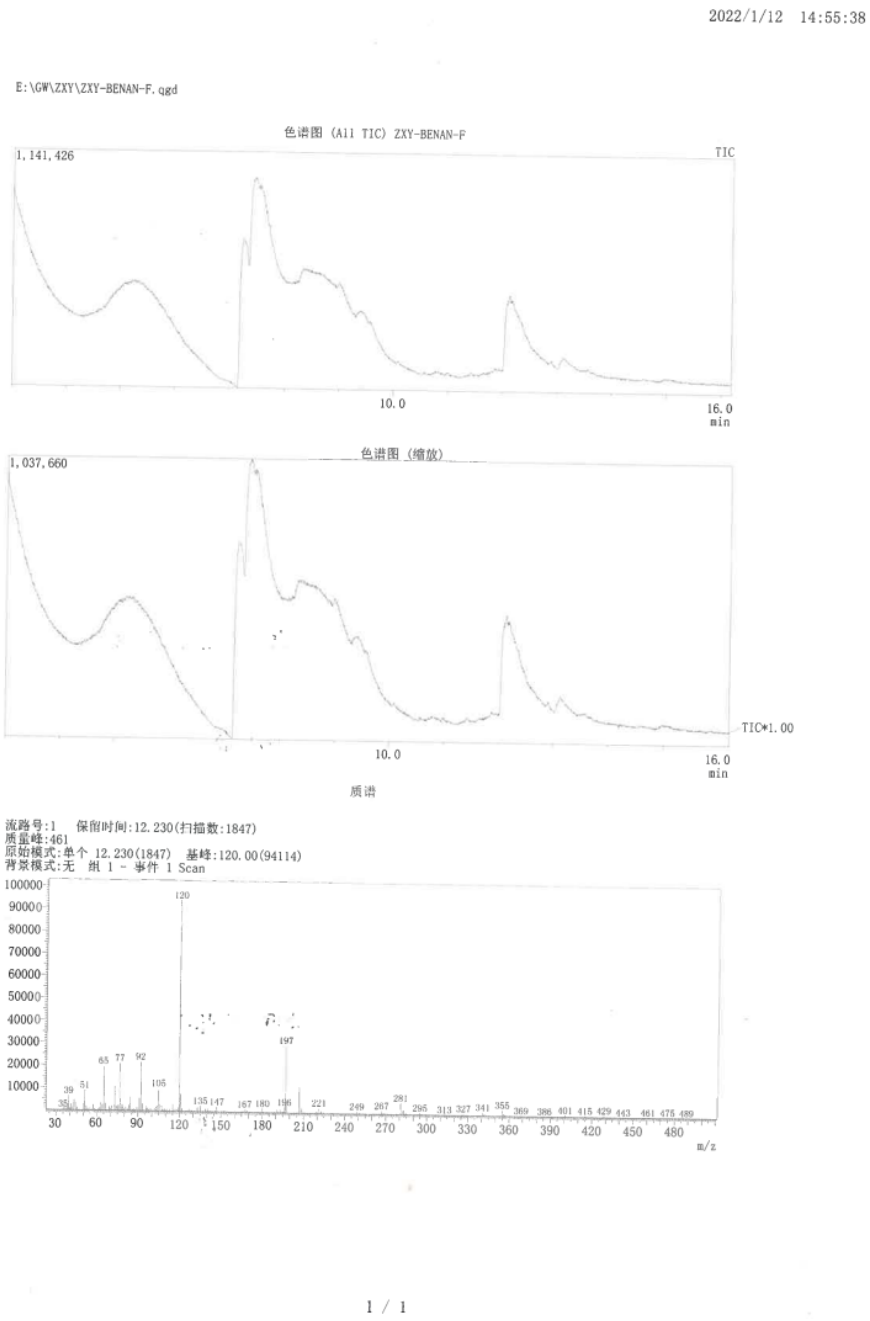
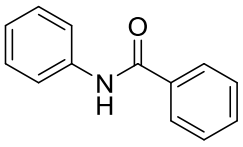


Figure S3 GCMS of *N*-Phenylbenzamide



N-phenylbenzamide: RT = 12.23 min, GC-MS: 197, 120, 105, 92, 77.

9. GCMS of 82

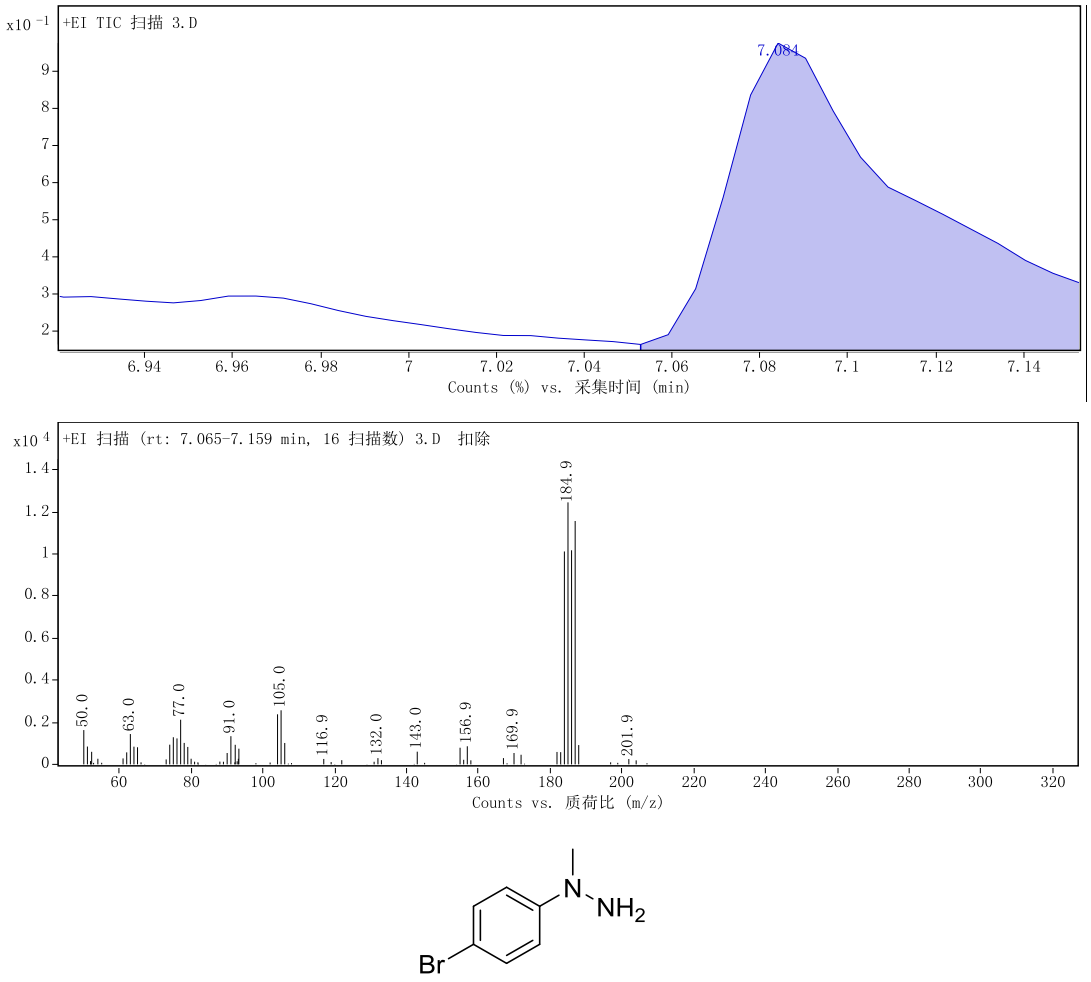


Figure S4 GCMS of the intermediate **82**

82: MS (EI, 70 eV) *m*/*z* 201, 185, 157, 105

10. Light On/off Experiments

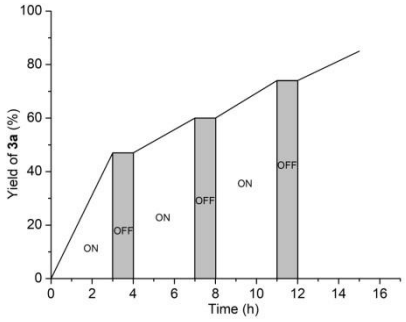


Figure S5 Light On/off Experiments

The procedures for Light On/off experiments: To a mixture of 1-methyl-1-phenylhydrazine (0.3 mmol), benzaldehyde (0.1 mmol), $Ir[dF(CF_3)ppy]_2(dtbpy)PF_6$ **PC 3** (1 mol%), TBAB (35 mol%), CCl_3Br (1.5 equiv.), and CH_3CN (1 mL, 0.1 M) were successively added into a quartz reaction tube with a stir bar. The reaction mixture was separately stirred and irradiated by 10 W blue LEDs (410 nm) at room temperature for 3 h, 6 h, and 9 h. The desired product **3** was isolated in 47%, 60%, and 74%, respectively. Additionally, the reaction mixture was stirred and irradiated by 10 W blue LEDs (410 nm) at room temperature for 3 h, then the reaction mixture was continuously stirred in the dark for 1 h, the corresponding product was also obtained in 47% yield. Additionally, when the reaction mixture was stirred and irradiated by 10 W blue LEDs (410 nm) at room temperature for 6 h, then the reaction mixture was continuously stirred in the dark for 1 h, the corresponding product was obtained in 60% yield. Additionally, when the reaction mixture was stirred and irradiated by 10 W blue LEDs (410 nm) at room temperature for 9 h, then the reaction mixture was continuously stirred in the dark for 1 h, the corresponding product **3** was still obtained in 74% yield.

11. Fluorescence Quenching Experiments

Quenched by 1-methyl-1-phenylhydrazine **1**: For each quenching experiment, the emission intensity of photocatalyst **PC 3** (1×10^{-5} M in CH_3CN, λ_{ex} = 298 nm, λ_{em} = 476 nm) with different concentration of quencher **1** (0, 1.0, 2.0, 3.0, 4.0, 5.0 mM) was collected. As shown in Figure S6, 1-methyl-1-phenylhydrazine **1** was capable of quenching the excited state of photocatalyst **PC 3**.

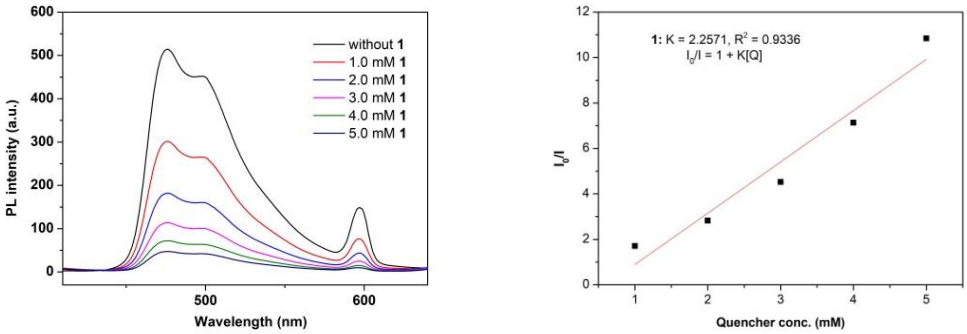


Figure S6 The Fluorescence Emission Spectra of a Solution of **PC 3** in CH_3CN Containing Different Concentration of Compound **1** and Stern-Volmer Graph

Quenched by compound **4**: For each quenching experiment, the emission intensity of photocatalyst **PC 3** (1×10^{-5} M in CH_3CN, λ_{ex} = 298 nm, λ_{em} = 476 nm) with different concentration of quencher **4** (0, 1.0, 2.0, 3.0, 4.0, 5.0 mM) was collected. As shown in Figure S7, 1-methyl-1-phenylhydrazine **4** was capable of quenching the excited state of photocatalyst **PC 3**.

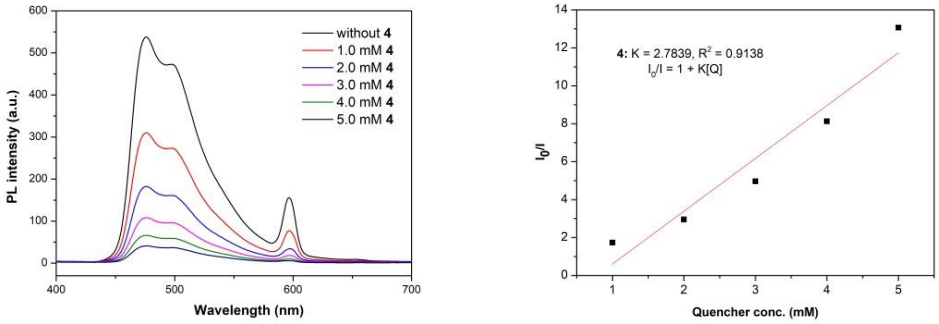


Figure S7 The Fluorescence Emission Spectra of a Solution of **PC 3** in CH_3CN Containing Different Concentration of Compound **4** and Stern-Volmer Graph

Quenched by intermediate **82**: For each quenching experiment, the emission intensity of photocatalyst PC 3 (1×10^{-5} M in CH_3CN, λ_{ex} = 298 nm, λ_{em} = 476 nm) with different concentration of quencher (0, 1.0, 2.0, 3.0, 4.0, 5.0 mM) was collected. As shown in Figure S8, 1-methyl-1-phenylhydrazine **82** was capable of quenching the excited state of photocatalyst **PC 3**.

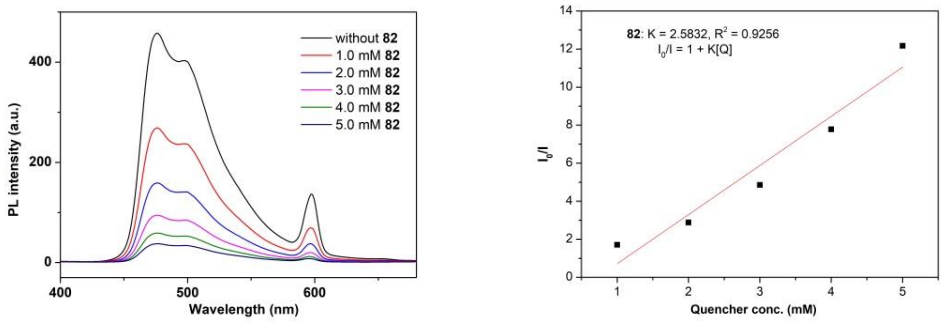


Figure S8 The Fluorescence Emission Spectra of a Solution of **PC 3** in CH_3CN Containing Different Concentration of Compound **82** and Stern-Volmer Graph

12. Cyclic Voltammetry (CV) Experiments

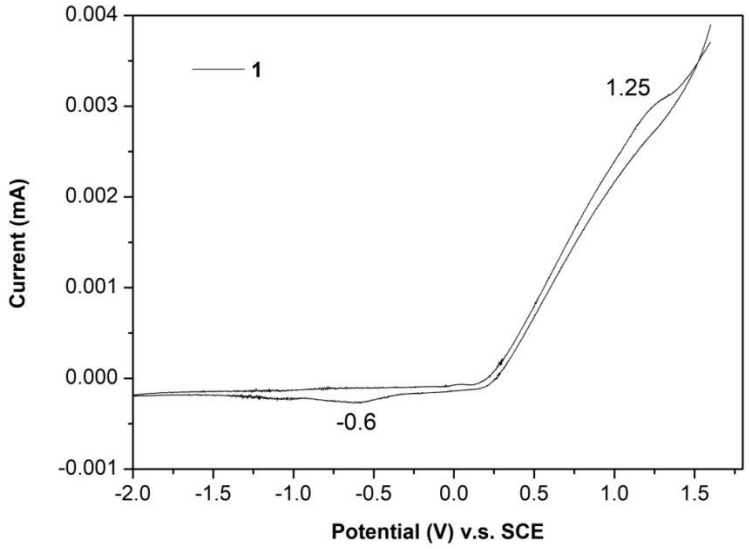
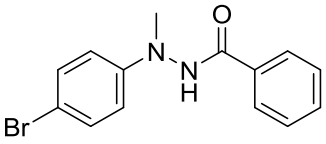
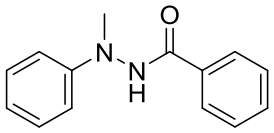


Figure S9 Cyclic Voltammogram of Compound **1** in CH_3CN at 100 mV/s (v.s. SCE)

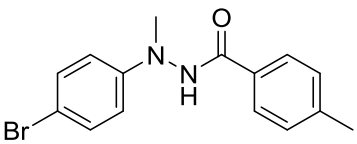
13. Characterization Data



N'-(4-Bromophenyl)-N'-methylbenzohydrazide (***3***). Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Brown solid: 85% yield (25.8 mg, 0.09 mmol); mp 176.5-177.8 ℃; IR (KBr, cm^{-1}) 3236, 3073, 2922, 2815, 1650, 1516, 1489, 1311, 1222, 1137, 1092, 902, 819, 734, 689, 506; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.74 (s, 1H), 7.90-7.92 (d, J = 8.0 Hz, 2H), 7.59 (t, J = 8.0 Hz, 1H), 7.51 (t, J = 8.0 Hz, 2H), 7.35 (d, J = 8.0 Hz, 2H), 6.77 (d, J = 8.0 Hz, 2H), 3.20 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.4, 149.2, 132.6, 131.9, 131.4, 128.5, 127.4, 114.3, 109.4, 40.2; MS (EI, 70 eV) *m/z* 304, 199, 185, 155, 105; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{14}BrN_2O$ 305.0284, found 305.0282.

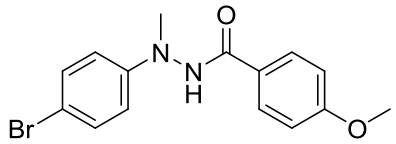


N'-Methyl-N'-phenylbenzohydrazide (***4***).[2] Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellowish solid: 55% yield (12.4 mg, 0.06 mmol); mp 151.1-152.9 ℃; IR (KBr, cm^{-1}) 3279, 3050, 2873, 1656, 1526, 1486, 1289, 1119, 1027, 994, 895, 750, 691, 539; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.65 (s, 1H), 7.91 (d, J = 8.0 Hz, 2H), 7.59 (t, J = 8.0 Hz, 1H), 7.51 (t, J = 8.0 Hz, 2H), 7.21 (t, J = 8.0 Hz, 2H), 6.82 (d, J = 8.0 Hz, 2H), 6.76 (t, J = 8.0 Hz, 1H), 3.21 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.5, 149.9, 132.9, 131.8, 128.9, 128.5, 127.3, 118.2, 112.3, 40.2; MS (EI, 70 eV) *m/z* 226, 121, 105, 77; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{15}N_2O$ 227.1179, found 227.1189.

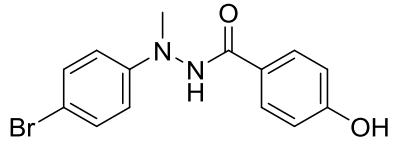


N'-(4-Bromophenyl)-N',4-dimethylbenzohydrazide (***5***). Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 79% yield (25.1 mg, 0.08mmol); mp >

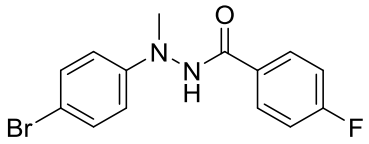
190 ℃; IR (KBr, cm^{-1}) 3253, 2920,2852, 1654, 1591, 1501, 1319, 1283, 1114, 896, 817, 755, 620, 542; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.65 (s, 1H), 7.80 (d, J = 8.0 Hz, 2H), 7.34 (d, J = 8.0 Hz, 2H), 7.31 (d, J = 8.0 Hz, 2H), 6.75 (d, J = 8.0 Hz, 2H), 3.18 (s, 3H), 2.37 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.3, 149.3, 141.9, 131.4, 129.8, 129.0, 127.4, 114.3, 109.3, 40.2, 21.0; MS (EI, 70 eV) *m/z* 318, 199, 185, 155, 119; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{15}H_{16}BrN_2O$ 319.0441, found 319.0457.



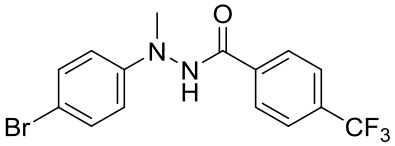
N'-(4-Bromophenyl)-4-methoxy-N'-methylbenzohydrazide ***(6)***. Eluent: petroleum ether/ethyl acetate (v/v = 3/1); Yellowish solid: 71% yield (23.7 mg, 0.07 mmol); mp > 190 ℃; IR (KBr, cm^{-1}) 3230, 2962, 2835, 1735, 1647, 1521, 1493, 1299, 1258, 1182, 1026, 903, 843, 780, 627, 509; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.60 (s, 1H), 7.88 (d, J = 8.0 Hz, 2H), 7.34 (d, J = 8.0 Hz, 2H), 7.04 (d, J = 8.0 Hz, 2H), 6.75 (d, J = 8.0 Hz, 2H), 3.82 (s, 3H), 3.18 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 164.9, 162.1, 149.4, 131.4, 129.3, 124.8, 114.3, 113.7, 109.2, 55.4, 40.2; MS (EI, 70 eV) *m/z* 334, 199, 185, 155, 135, 107; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{15}H_{16}BrN_2O_2$ 335.0390, found 335.0406.



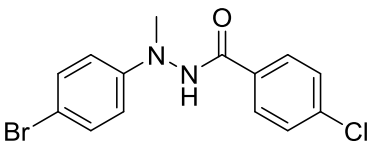
N'-(4-Bromophenyl)-4-hydroxy-N'-methylbenzohydrazide ***(7)***. Eluent: petroleum ether/ethyl acetate (v/v = 1/1); Yellow solid: 65% yield (20.8 mg, 0.07 mmol); mp 180.1-181.7 ℃; IR (KBr, cm^{-1}) 3241, 3131, 2923, 2870, 1666, 1637, 1505, 1424, 1351, 1281, 1173, 1003, 975, 898, 769, 623, 512; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.49 (s, 1H), 10.12 (s, 1H), 7.77 (d, J = 8.0 Hz, 2H), 7.33 (d, J = 8.0 Hz, 2H), 6.84 (d, J = 8.0 Hz, 2H), 6.73 (d, J = 8.0 Hz, 2H), 3.17 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.1, 160.7, 149.4, 131.4, 129.4, 123.2, 115.0, 114.2, 109.1, 40.2; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{14}BrN_2O_2$ 321.0233, found 321.0224.



N'-(4-Bromophenyl)-4-fluoro-N'-methylbenzohydrazide (***8***). Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Gray solid: 80% yield (25.8 mg, 0.08 mmol); mp 169.1-171.3 ℃; IR (KBr, cm^{-1}) 3262, 3071, 2922, 2812, 1660, 1527, 1462, 1499, 1301, 1232, 1116, 895, 816, 768, 614, 501; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.76 (s, 1H), 7.98 (dd, J = 8.0, 4.0 Hz, 2H), 7.33-7.37 (m, 4H), 6.77 (d, J = 8.0 Hz, 2H), 3.19 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 164.3 (d, J = 248.0 Hz), 164.5, 149.2, 131.4, 130.1 (d, J = 9.0 Hz), 129.1 (d, J = 3.0 Hz), 115.5 (d, J = 21.0 Hz), 114.4, 109.5, 40.2; MS (EI, 70 eV) *m/z* 322, 199, 183, 155, 123, 105; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{13}BrFN_2O$ 323.0190, found 323.0204.

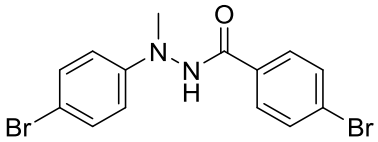


N'-(4-Bromophenyl)-N'-methyl-4-(trifluoromethyl)benzohydrazide (***9***). Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellowish solid: 85% yield (31.6 mg, 0.09 mmol); mp > 190 ℃; IR (KBr, cm^{-1}) 3252, 3060, 2922, 1663, 1536, 1457, 1324, 1289, 1134, 1067, 977, 821, 774, 645, 511; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.95 (s, 1H), 8.10 (d, J = 8.0 Hz, 2H), 7.90 (d, J = 8.0 Hz, 2H), 7.36 (d, J = 12.0 Hz, 2H), 6.80 (d, J = 8.0 Hz, 2H), 3.21 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 164.4, 149.0, 136.5, 131.4, 130.1, 128.4, 125.5 (q, J = 4.0 Hz), 123.8 (q, J = 271.0 Hz), 114.5, 109.7, 40.1; MS (EI, 70 eV) *m/z* 372, 199, 185, 172, 145, 120; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{15}H_{13}BrF_3N_2O$ 373.0158, found 373.0152.

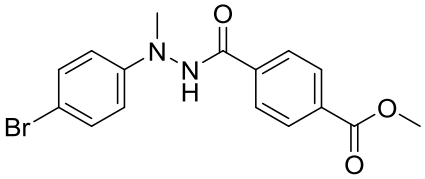


N'-(4-Bromophenyl)-4-chloro-N'-methylbenzohydrazide (***10***). Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 63% yield (21.3 mg, 0.06 mmol); mp > 190 ℃; IR (KBr, cm^{-1}) 3253, 3044, 2921, 1658, 1529, 1488, 1454, 1292, 1113, 1094,

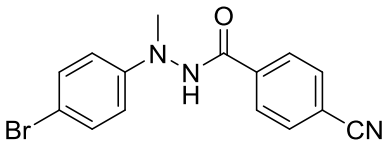
998, 894, 760, 607, 528; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.82 (s, 1H), 7.93 (d, J = 12.0 Hz, 2H), 7.59 (d, J = 8.0 Hz, 2H), 7.35 (d, J = 8.0 Hz, 2H), 6.77 (d, J = 8.0 Hz, 2H), 3.19 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 164.5, 149.1, 136.7, 131.4, 131.4, 129.3, 128.6, 114.4, 109.5, 40.1; MS (EI, 70 eV) *m/z* 340, 199, 183, 155, 139; HRMS (ESI) *m/z* [M + H]$^{+}$ Calcd for $C_{14}H_{13}BrClN_2O$ 338.9894, found 338.9887.



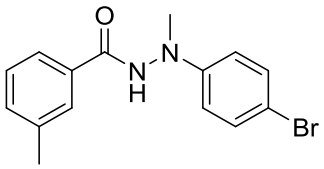
4-Bromo-N'-(4-bromophenyl)-N'-methylbenzohydrazide ***(11)***. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); White solid: 65% yield (25.0 mg, 0.07 mmol); mp > 190 ℃; IR (KBr, cm^{-1}) 3250, 3038, 2921, 1658, 1525, 1454, 1294, 1112, 1009, 976, 846, 756, 605, 539; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.81 (s, 1H), 7.84 (d, J = 8.0 Hz, 2H), 7.73 (d, J = 8.0 Hz, 2H), 7.35 (d, J = 12.0 Hz, 2H), 6.77 (d, J = 8.0 Hz, 2H), 3.18 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 164.6, 149.1, 131.8, 131.6, 131.4, 129.5, 125.7, 114.4, 109.5, 40.1; MS (EI, 70 eV) *m/z* 384, 199, 183, 155, 104; HRMS (ESI) *m/z* [M + H]$^{+}$ Calcd for $C_{14}H_{13}Br_2N_2O$ 382.9389, found 382.9381.



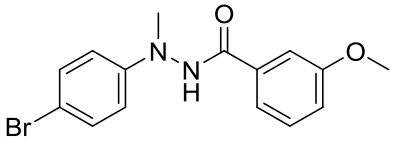
Methyl 4-(2-(4-Bromophenyl)-2-methylhydrazine-1-carbonyl)benzoate ***(12)***. Eluent: petroleum ether/ethyl acetate (v/v = 3/1); Yellow solid: 67% yield (24.4 mg, 0.07 mmol); mp 173.2-174.5 ℃; IR (KBr, cm^{-1}) 3256, 2996, 2945,2881, 1722, 1659,1525, 1434, 1282, 11110, 1017, 969, 895, 739, 606, 548; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.91 (s, 1H), 8.05 (q, J = 8.0 Hz, 4H), 7.36 (d, J = 8.0 Hz, 2H), 6.79 (d, J = 8.0 Hz, 2H), 3.89 (s, 3H), 3.20 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 165.6, 164.7, 149.0, 136.8, 132.3, 131.4, 129.3, 127.8, 114.4, 109.6, 52.4, 40.1; MS (EI, 70 eV) *m/z* 362, 333, 281, 199, 185, 163, 135; HRMS (ESI) *m/z* [M + H]$^{+}$ Calcd for $C_{16}H_{16}BrN_2O_3$ 363.0339, found 363.0334.



N'-(4-Bromophenyl)-4-cyano-N'-methylbenzohydrazide ***(13)***. Eluent: petroleum ether/ethyl acetate (v/v = 1/1); Yellow solid: 61% yield (20.1 mg, 0.06 mmol); mp 147.2-148.8 ℃; IR (KBr, cm^{-1}) 3235, 3048, 2922, 2232, 1658, 1591, 1492, 1303, 1289, 1115, 1005, 898, 817, 766, 613, 565; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.96 (s, 1H), 8.03 (q, J = 8.0 Hz, 4H), 7.35 (d, J = 8.0 Hz, 2H), 6.79 (d, J = 8.0 Hz, 2H), 3.19 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 164.2, 149.0, 136.7, 132.6, 131.5, 128.3, 118.2, 114.5, 114.2, 109.7, 40.1; MS (EI, 70 eV) *m/z* 329, 199, 185, 157, 130; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{15}H_{13}BrN_3O$ 330.0237, found 330.0245.

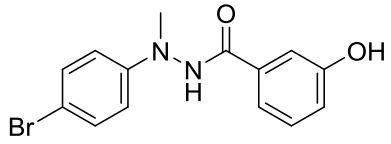


N'-(4-Bromophenyl)-N',3-dimethylbenzohydrazide ***(14)***. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 72% yield (22.9 mg, 0.07 mmol); mp 124.6-125.8 ℃; IR (KBr, cm^{-1}) 3264, 2921,2852, 1659, 1588, 1489, 1308, 1234, 1130, 1077, 933, 822, 756, 689, 509; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.67 (s, 1H), 7.73 (s, 1H), 7.71-7.68 (m, 1H), 7.40-7.39 (m, 2H), 7.35 (d, J = 8.0 Hz, 2H), 6.76 (d, J = 8.0 Hz, 2H), 3.18 (s, 3H), 2.37 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.6, 149.2, 137.8, 132.7, 132.4, 131.4, 128.4, 127.9, 124.5, 114.3, 109.3, 40.2, 20.9; MS (EI, 70 eV) *m/z* 318, 201, 183, 155, 119; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{15}H_{16}BrN_2O$ 319.0441, found 319.0455.

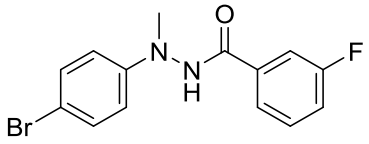


N'-(4-Bromophenyl)-3-methoxy-N'-methylbenzohydrazide ***(15)***. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Brown solid: 73% yield (24.4 mg, 0.07 mmol); mp

93.6-94.7 ℃; IR (KBr, cm^{-1}) 3243, 3003, 2961,2835, 1658, 1589, 1490, 1296, 1114, 1079, 998, 816, 757, 688, 541; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.73 (s, 1H), 7.49 (d, J = 8.0 Hz, 1H), 7.46-7.40 (m, 2H), 7.35 (d, J = 8.0 Hz, 2H), 7.16 (dd, J = 8.0 Hz, 4.0 Hz, 1H), 6.77 (d, J = 8.0 Hz, 2H), 3.82 (s, 3H), 3.19 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.2, 159.3, 149.2, 134.0, 131.4, 129.7, 119.6, 117.8, 114.3, 112.5, 109.4, 55.3, 40.2; MS (EI, 70 eV) *m/z* 334, 199, 185, 152, 135, 107; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{15}H_{16}BrN_2O_2$ 335.0390, found 335.0406.

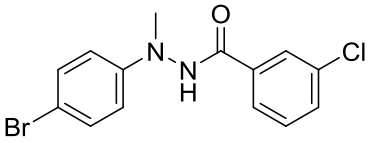


*N'-(4-Bromophenyl)-3-hydroxy-N'-methylbenzohydrazide (**16**).* Eluent: petroleum ether/ethyl acetate (v/v = 1/1); Brown solid: 73% yield (23.4 mg, 0.07 mmol); mp 114.1-115.6 ℃; IR (KBr, cm^{-1}) 3342, 3248, 3066, 2921, 1633, 1589, 1492, 1296, 1219, 1115, 1047, 997, 840, 747, 687, 528; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.62 (s, 1H), 9.76 (s, 1H), 7.35 (d, J = 12.0 Hz, 2H), 7.32-7.27 (m, 3H), 6.97 (dd, J = 8.0 Hz, 4.0 Hz, 1H), 6.75 (d, J = 8.0 Hz, 2H), 3.17 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.6, 157.5, 149.3, 134.1, 131.4, 129.6, 118.8, 117.8, 114.3, 114.3, 109.4, 40.2; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{14}BrN_2O_2$ 321.0233, found 321.0228.

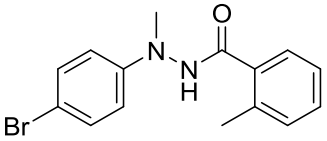


*N'-(4-Bromophenyl)-3-fluoro-N'-methylbenzohydrazide (**17**).* Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Brown solid: 84% yield (27.0 mg, 0.08 mmol); mp 111.5-112.7 ℃; IR (KBr, cm^{-1}) 3255, 3069, 2926, 1657, 1588, 1489, 1295, 1270, 1116, 1079, 998, 880, 755, 612, 522; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.84 (s, 1H), 7.77 (d, J = 8.0 Hz, 1H), 7.70 (d, J = 8.0 Hz, 1H), 7.60-7.55 (m, 1H), 7.45 (t, J = 8.0 Hz, 1H), 7.35 (d, J = 8.0 Hz, 2H), 6.79 (d, J = 12.0 Hz, 2H), 3.20 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 164.2 (d, J = 3.0 Hz), 162.0 (d, J = 244.0 Hz), 149.1, 135.0 (d, J = 7.0 Hz), 131.4, 130.8 (d, J = 8.0 Hz), 123.6 (d, J = 3.0 Hz), 118.8

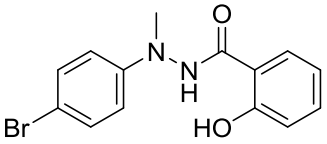
(d, *J* = 21.0 Hz), 114.4, 114.2 (d, *J* = 23.0 Hz), 109.6, 40.1; MS (EI, 70 eV) *m/z* 322, 199, 183, 155, 123, 105; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{13}BrFN_2O$ 323.0190, found 323.0205.



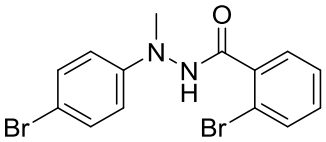
N'-(4-Bromophenyl)-3-chloro-N'-methylbenzohydrazide ***(18)***. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 80% yield (27.0 mg, 0.08 mmol); mp 123.5-124.8 ℃; IR (KBr, cm^{-1}) 3278, 3176, 2965, 1650, 1534, 1489, 1310, 1286, 1109, 1079, 916, 813, 731, 685, 506; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 10.85 (s, 1H), 7.95 (s, 1H), 7.86 (d, *J* = 8.0 Hz, 1H), 7.68 (d, *J* = 8.0 Hz, 1H), 7.56 (t, *J* = 8.0 Hz, 1H), 7.36 (d, *J* = 8.0 Hz, 2H), 6.79 (d, *J* = 8.0 Hz, 2H), 3.19 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 164.2, 149.1, 134.7, 133.4, 131.7, 131.4, 130.6, 127.2, 126.2, 114.5, 109.6, 40.1; MS (EI, 70 eV) *m/z* 340, 199, 185, 155, 139; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{13}BrClN_2O$ 338.9894, found 338.9911.



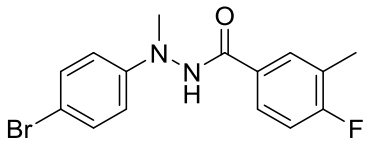
N'-(4-Bromophenyl)-N',2-dimethylbenzohydrazide ***(19)***. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Brown solid: 72% yield (22.9 mg, 0.07mmol); mp 146.7-147.8 ℃; IR (KBr, cm^{-1}) 3250, 2956, 2892, 1726, 1658, 1507, 1487, 1301, 1280, 1113, 1086, 902, 817, 748, 666, 565; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 10.42 (s, 1H), 7.48 (d, *J* = 8.0 Hz, 1H), 7.41-7.37 (m, 3H), 7.30 (d, *J* = 8.0 Hz, 2H), 6.82 (d, *J* = 8.0 Hz, 2H), 3.20 (s, 3H), 2.38 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 168.1, 149.2, 135.8, 134.6, 131.5, 130.6, 130.0, 127.3, 125.7, 114.4, 109.5, 40.2, 19.3; MS (EI, 70 eV) *m/z* 320, 199, 185, 155, 136, 119; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{15}H_{16}BrN_2O$ 319.0441, found 319.0454.



*N'-(4-Bromophenyl)-2-hydroxy-N'-methylbenzohydrazide (**20**)*. Eluent: petroleum ether/ethyl acetate (v/v = 1/1); Reddish brown solid: 77% yield (24.6 mg, 0.08 mmol); mp 115.3.-116.8 ℃; IR (KBr, cm^{-1}) 3268, 3147, 2920, 1644, 1535, 1490, 1310, 1240, 1104, 1037, 902, 812, 793, 625, 532; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 11.76 (s, 1H), 10.77 (s, 1H), 7.86 (d, J = 8.0 Hz, 1H), 7.44 (t, J = 8.0 Hz, 1H), 7.36 (d, J = 8.0 Hz, 2H), 6.98-6.91 (m, 2H), 6.79 (d, J = 8.0 Hz, 2H), 3.20 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 167.9, 159.1, 148.9, 134.0, 131.5, 128.2, 119.0, 117.3, 115.2, 114.4, 109.6, 40.1; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{14}BrN_2O_2$ 321.0233, found 321.0247.

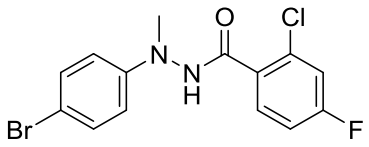


*2-Bromo-N'-(4-bromophenyl)-N'-methylbenzohydrazide (**21**)*. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Grey solid: 73% yield (28.0 mg, 0.07 mmol); mp 98.1-99.0 ℃; IR (KBr, cm^{-1}) 3268, 3002, 2960, 1660, 1505, 1488, 1303, 1215, 1103, 1024, 902, 819, 750, 696, 563; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.57 (s, 1H), 7.71 (d, J = 8.0 Hz, 1H), 7.57 (d, J = 8.0 Hz, 1H), 7.50 (t, J = 8.0 Hz, 1H), 7.44 (d, J = 8.0 Hz, 1H), 7.38 (d, J = 12.0 Hz, 2H), 6.88 (d, J = 8.0 Hz, 2H), 3.21 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 166.3, 148.9, 136.9, 132.8, 131.5, 131.4, 129.1, 127.7, 119.2, 114.6, 109.7, 40.0; MS (EI, 70 eV) *m/z* 384, 199, 185, 155, 105; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{13}Br_2N_2O$ 382.9389, found 382.9385.

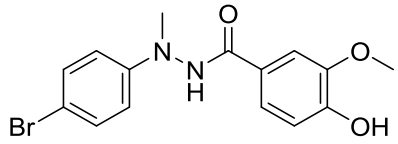


*N'-(4-Bromophenyl)-4-fluoro-N',3-dimethylbenzohydrazide (**22**)*. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Brown solid: 76% yield (25.5 mg, 0.08 mmol); mp 136.2-137.1 ℃; IR (KBr, cm^{-1}) 3256, 3064, 2921, 2882, 1881, 1657, 1590, 1492, 1317, 1260, 1122, 1076, 944, 820, 774, 652, 511; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.70 (s, 1H), 7.86 (d, J = 8.0 Hz, 1H), 7.80-7.76 (m, 1H), 7.34 (d, J = 8.0 Hz, 2H), 7.27 (t, J = 8.0 Hz, 1H), 6.76 (d, J = 8.0 Hz, 2H), 3.18 (s, 3H), 2.29 (s, 3H);

$^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 164.6, 162.8 (d, J = 247.0 Hz), 149.2, 131.4, 131.2 (d, J = 6.0 Hz), 128.8 (d, J = 4.0 Hz), 127.4 (d, J = 9.0 Hz), 124.6 (d, J = 18.0 Hz), 115.2 (d, J = 22.0 Hz), 114.4, 109.4, 40.2, 14.1 (d, J = 3.0 Hz); MS (EI, 70 eV) *m/z* 336, 199, 183, 155, 137, 109; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{15}H_{15}BrFN_2O$ 337.0346, found 337.0343.

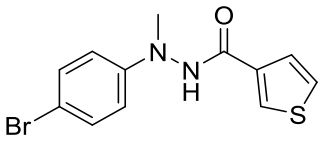


N'-(4-Bromophenyl)-2-chloro-4-fluoro-N'-methylbenzohydrazide ***(23)***. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 71% yield (25.3 mg, 0.07 mmol); mp 104.1-105.2 ℃; IR (KBr, cm^{-1}) 3305, 3218, 3026, 2993, 2809, 1867, 1665, 1599, 1487, 1346, 1258, 1101, 1040, 920, 808, 717, 620, 536; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.60 (s, 1H), 7.69 (t, J = 8.0 Hz, 1H), 7.58 (dd, J = 8.0 Hz, 4.0 Hz, 1H), 7.38 (d, J = 12.0 Hz, 2H), 7.34 (dd, J = 8.0 Hz, 4.0 Hz, 1H), 6.86 (d, J = 8.0 Hz, 2H), 3.19 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 164.7, 162.4 (d, J = 248.0 Hz), 148.8, 131.7 (d, J = 11.0 Hz), 131.4, 131.0 (d, J = 9.0 Hz), 117.2 (d, J = 25.0 Hz), 117.1, 114.6 (d, J = 12.0 Hz), 114.6, 109.8, 40.1; MS (EI, 70 eV) *m/z* 358, 199, 185, 157, 129; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{12}BrClFN_2O$ 356.9800, found 356.9815.

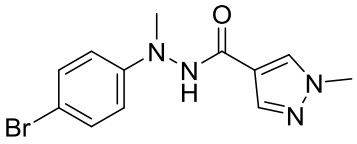


N'-(4-Bromophenyl)-4-hydroxy-3-methoxy-N'-methylbenzohydrazide ***(24)***. Eluent: petroleum ether/ethyl acetate (v/v = 1/1); Brown solid: 57% yield (20.0 mg, 0.06 mmol); mp 56.1-57.3 ℃; IR (KBr, cm^{-1}) 3511, 3236, 2923, 2851, 1649, 1592, 1491, 1379, 1288, 1118, 1027, 997, 846, 753, 648, 557; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.52 (s, 1H), 9.71 (s, 1H), 7.47 (d, J = 4.0 Hz, 1H), 7.41 (dd, J = 8.0 Hz, 4.0 Hz, 1H), 7.34 (d, J = 8.0 Hz, 2H), 6.85 (d, J = 8.0 Hz, 1H), 6.74 (d, J = 8.0 Hz, 2H), 3.82 (s, 3H), 3.17 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.0, 150.1, 149.4, 147.3, 131.4, 123.4, 121.0, 115.0, 114.3, 111.3, 109.2, 55.7, 40.2; HRMS (ESI)

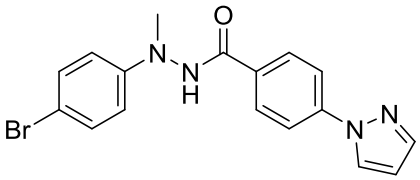
m/z $[M + H]^+$ Calcd for $C_{15}H_{16}BrN_2O_3$ 351.0339, found 351.0355.



N'-(4-Bromophenyl)-N'-methylthiophene-3-carbohydrazide (***25***). Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 58% yield (18.0 mg, 0.06 mmol); mp 149.1-150.2 ℃; IR (KBr, cm^{-1}) 3233, 3100, 2919, 2879, 1657, 1536, 1486, 1415, 1285, 1114, 1074, 951, 880, 749, 663, 573; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 10.59 (s, 1H), 8.25 (d, J = 4.0 Hz, 1H), 7.65 (dd, J = 8.0 Hz, 4.0 Hz, 1H), 7.55 (d, J = 4.0 Hz, 1H), 7.35 (d, J = 12.0 Hz, 2H), 6.76 (d, J = 12.0 Hz, 2H), 3.18 (s, 3H).; $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 161.2, 149.2, 135.4, 131.4, 129.7, 127.2, 126.6, 114.3, 109.4, 40.2; MS (EI, 70 eV) *m/z* 312, 199, 185, 155, 111; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{12}H_{12}BrN_2OS$ 310.9848, found 310.9844.

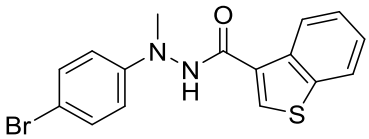


N'-(4-Bromophenyl)-N',1-dimethyl-1H-pyrazole-4-carbohydrazide (***26***). Eluent: petroleum ether/ethyl acetate (v/v = 1/1); Brown solid: 53% yield (16.3 mg, 0.05 mmol); mp 159.3-160.4 ℃; IR (KBr, cm^{-1}) 3234, 3110, 3001, 2925, 2803, 1666, 1559, 1487, 1393, 1260, 1106, 1007, 973, 819, 767, 660, 503; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 10.33 (s, 1H), 8.20 (s, 1H), 7.91 (s, 1H), 7.33 (d, J = 8.0 Hz, 2H), 6.73 (d, J = 12.0 Hz, 2H), 3.87 (s, 3H), 3.16 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 161.1, 149.4, 138.5, 132.4, 131.4, 116.0, 114.3, 109.3, 40.4, 38.8; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{12}H_{14}BrN_4O$ 309.0346, found 309.0342.

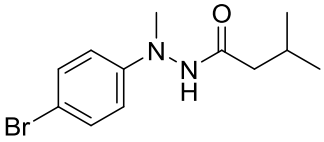


N'-(4-Bromophenyl)-N'-methyl-4-(1H-pyrazol-1-yl)benzohydrazide (***27***). Eluent: petroleum ether/ethyl acetate (v/v = 3/1); White solid: 68% yield (25.2 mg, 0.07

mmol); mp > 190 ℃; IR (KBr, cm^{-1}) 3247, 2921, 1657, 1503, 1434, 1390, 1291, 1111, 1044, 933, 894, 748, 652, 607; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.80 (s, 1H), 8.63 (d, J = 4.0 Hz, 1H), 8.01 (q, J = 8.0 Hz, 4H), 7.81 (d, J = 4.0 Hz, 1H), 7.36 (d, J = 8.0 Hz, 2H), 6.79 (d, J = 8.0 Hz, 2H), 6.60 (d, J = 4.0, Hz, 1H), 3.21 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 164.7, 149.2, 142.0, 141.8, 131.4, 129.9, 129.0, 128.2, 117.9, 114.4, 109.5, 108.5, 40.2; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{17}H_{16}BrN_4O$ 371.0502, found 371.0522.

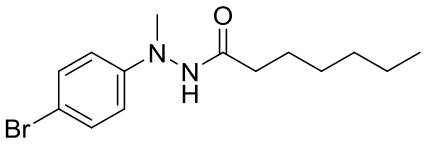


N'-(4-Bromophenyl)-N'-methylbenzo[b]thiophene-3-carbohydrazide ***(28)***. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellowish solid: 48% yield (17.3 mg, 0.05 mmol); mp 135.1-137.1 ℃; IR (KBr, cm^{-1}) 3260, 3066, 2930, 2854, 2797, 2685, 1650, 1592, 1491, 1314, 1281, 1195, 1029, 999, 813, 766, 616, 515; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.72 (s, 1H), 8.54 (s, 1H), 8.41-8.39 (m, 1H), 8.09-8.06 (m, 1H), 7.47-7.43 (m, 2H), 7.36 (d, J = 12.0 Hz, 2H), 6.83 (d, J = 8.0 Hz, 2H), 3.24 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 162.3, 149.3, 139.4, 137.1, 131.8, 131.4, 128.5, 125.1, 125.0, 124.2, 122.8, 114.4, 109.5, 40.3; MS (EI, 70 eV) *m/z* 362, 281, 207, 185, 161; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{16}H_{14}BrN_2OS$ 361.0005, found 361.0005.

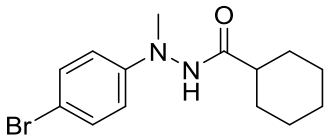


N'-(4-Bromophenyl)-N',3-dimethylbutanehydrazide ***(29)***. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 39% yield (11.1 mg, 0.04 mmol); mp 103.9-105.3 ℃; IR (KBr, cm^{-1}) 3206, 3266, 2954, 2869, 1881, 1660, 1517, 1491, 1368, 1221, 1110, 1001, 979, 823, 762, 692, 568; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.00 (s, 1H), 7.32 (d, J = 8.0 Hz, 2H), 6.68 (d, J = 12.0 Hz, 2H), 3.06 (s, 3H), 2.01 (s, 3H), 0.92 (d, J = 8.0 Hz, 6H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 170.6, 149.1, 131.3, 114.2, 109.2, 42.4, 40.1, 25.4, 22.3; MS (EI, 70 eV) *m/z* 286, 200,

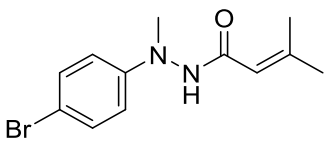
185, 155, 105; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{12}H_{18}BrN_2O$ 285.0597, found 285.0609.



*N'-(4-Bromophenyl)-N'-methylheptanehydrazide (**30**)*. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Brown solid: 36% yield (11.2 mg, 0.04 mmol); mp 57.3-58.4 ℃; IR (KBr, cm^{-1}) 3266, 3015, 2926, 2854, 1559, 1523, 1490, 1326, 1258, 1114, 1097, 968, 812, 753, 696, 539; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 9.98 (s, 1H), 7.31 (d, J = 12.0 Hz, 2H), 6.67 (d, J = 12.0 Hz, 2H), 3.05 (s, 3H), 2.12 (t, J = 8.0 Hz, 2H), 1.30-1.23 (m, 8H), 0.87 (t, J = 8.0 Hz, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 171.3, 149.2, 131.3, 114.2, 109.2, 40.2, 33.2, 30.9, 28.3, 24.9, 22.0, 13.9; MS (EI, 70 eV) *m/z* 314, 200, 185, 155, 105; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{22}BrN_2O$ 313.0910, found 313.0907.

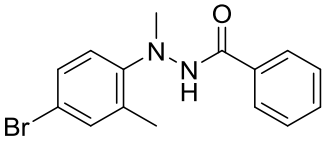


*N'-(4-Bromophenyl)-N'-methylcyclohexanecarbohydrazide (**31**)*. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 42% yield (12.9 mg, 0.04 mmol); mp 171.5-172.5 ℃; IR (KBr, cm^{-1}) 3261, 3075, 2933, 2852, 1665, 1520, 1490, 1319, 1262, 1108, 1039, 956, 821, 745, 604, 543; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 9.94 (s, 1H), 7.31 (d, J = 12.0 Hz, 2H), 6.66 (d, J = 8.0 Hz, 2H), 3.04 (s, 3H), 2.20-2.12 (m, 1H), 1.77-1.71 (m, 4H), 1.40-1.14 (m, 6H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 174.1, 149.2, 131.3, 114.1, 109.1, 42.0, 40.2, 28.9, 25.4, 25.1; MS (EI, 70 eV) *m/z* 310, 200, 185, 155, 105; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{20}BrN_2O$ 311.0754, found 311.0751.

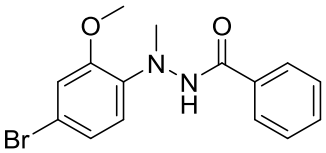


*N'-(4-Bromophenyl)-N',3-dimethylbut-2-enehydrazide (**32**)*. Eluent: petroleum

ether/ethyl acetate (v/v = 5/1); Brown liquid: 37% yield (10.4 mg, 0.04 mmol); IR (KBr, cm^{-1}) 3239, 2928, 1732, 1648, 1593, 1491, 1360, 1257, 1114, 1078, 998, 814, 754, 608, 549; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 9.92 (s, 1H), 7.32 (d, J = 8.0 Hz, 2H), 6.66 (d, J = 12.0 Hz, 2H), 5.69 (s, 1H), 3.08 (s, 3H), 2.08 (s, 3H), 1.83 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.1, 151.9, 149.2, 131.3, 116.1, 114.1, 109.0, 40.2, 27.0, 19.5; MS (EI, 70 eV) *m/z* 310, 200, 185, 155, 105; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{12}H_{16}BrN_2O$ 283.0441, found 283.0434.

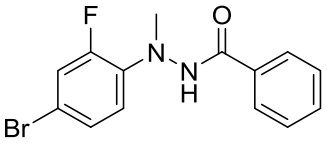


*N'-(4-Bromo-2-methylphenyl)-N'-methylbenzohydrazide (**33**)*. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellowish solid: 86% yield (27.3 mg, 0.09 mmol); mp 144.1-145.2 ℃; IR (KBr, cm^{-1}) 3218, 3027, 2953, 2864, 1651, 1519, 1488, 1302, 1290, 1121, 1099, 907, 807, 719, 680, 573; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.32 (s, 1H), 7.81 (d, J = 8.0 Hz, 2H), 7.54 (t, J = 8.0 Hz, 1H), 7.46 (t, J = 8.0 Hz, 2H), 7.34-7.29 (m, 2H), 7.12 (d, J = 8.0 Hz, 1H), 3.05 (s, 3H), 2.21 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 164.1, 147.9, 132.7, 132.5, 132.5, 131.0, 128.1, 127.9, 126.8, 119.3, 113.9, 41.2, 18.0; MS (EI, 70 eV) *m/z* 318, 215, 199, 171, 117, 105; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{15}H_{16}BrN_2O$ *m/z* 319.0441, found 319.0433.

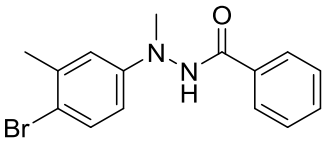


*N'-(4-Bromo-2-methoxyphenyl)-N'-methylbenzohydrazide (**34**)*. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellowish solid: 70% yield (23.4 mg, 0.07 mmol); mp 146.2-147.8 ℃; IR (KBr, cm^{-1}) 3225, 3063, 2960, 1650, 1547, 1496, 1306, 1244, 1116, 1029, 972, 846, 746, 693, 583; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.37 (s, 1H), 7.83 (d, J = 8.0 Hz, 2H), 7.54 (t, J = 8.0 Hz, 1H), 7.47 (t, J = 8.0 Hz, 2H), 7.09 (s, 1H), 7.04 (d, J = 8.0 Hz, 1H), 6.95 (d, J = 12.0 Hz, 1H), 3.79 (s, 3H), 3.14 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 164.8, 151.1, 139.2, 133.3, 131.4, 128.3,

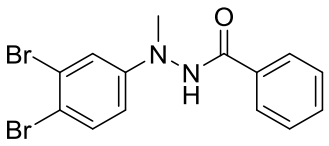
127.3, 122.9, 118.9, 115.2, 113.5, 56.0, 42.1; MS (EI, 70 eV) *m/z* 318, 215, 199, 171, 117, 105; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{15}H_{16}BrN_2O_2$ 335.0390, found 335.0385.



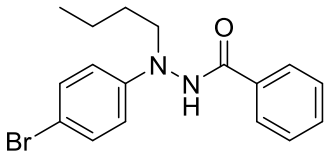
N'-(4-Bromo-2-fluorophenyl)-N'-methylbenzohydrazide ***(35)***. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); White solid: 70% yield (22.5 mg, 0.07 mmol); mp 130.9-131.7 ℃; IR (KBr, cm^{-1}) 3443, 3198, 3071, 2939, 1661, 1517, 1492, 1287, 1219, 1123, 1092, 910, 867, 721, 690, 584; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 10.74 (s, 1H), 7.84 (d, J = 8.0 Hz, 2H), 7.57 (t, J = 8.0 Hz, 1H), 7.48 (t, J = 8.0 Hz, 2H), 7.39 (dd, J = 8.0 Hz, 4.0 Hz, 1H), 7.27 (dd, J = 8.0 Hz, 4.0 Hz, 1H), 7.01 (t, J = 8.0 Hz, 1H), 3.20 (d, J = 4.0 Hz, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.1, 152.2 (d, J = 247.0 Hz), 137.9 (d, J = 7.0 Hz), 132.7, 131.8, 128.5, 127.3, 127.2 (d, J = 3.0 Hz), 119.4, 119.2 (d, J = 3.0 Hz), 111.2 (d, J = 9.0 Hz), 41.8 (d, J = 4.0 Hz); MS (EI, 70 eV) *m/z* 324, 217, 203, 190, 105; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{13}BrFN_2O$ 323.0190, found 323.0186.



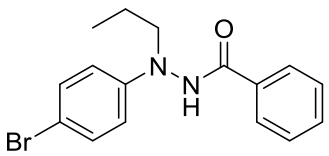
N'-(4-Bromo-3-methylphenyl)-N'-methylbenzohydrazide ***(36)***. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 63% yield (20.0 mg, 0.06 mmol); mp 132.3-133.2 ℃; IR (KBr, cm^{-1}) 3261, 3061, 2921, 2853, 1655, 1507, 1484, 1311, 1272, 1193, 1091, 1020, 917, 838, 726, 689, 561; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 10.71 (s, 1H), 7.91 (d, J = 8.0 Hz, 2H), 7.59 (t, J = 8.0 Hz, 1H), 7.51 (t, J = 8.0 Hz, 2H), 7.34 (d, J = 8.0 Hz, 1H), 6.80 (d, J = 4.0 Hz, 1H), 6.59 (dd, J = 8.0 Hz, 4.0 Hz, 1H), 3.19 (s, 3H), 2.28 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.5, 149.5, 137.2, 132.7, 132.1, 131.9, 128.5, 127.4, 114.7, 112.1, 112.0, 40.2, 22.8; MS (EI, 70 eV) *m/z* 318, 213, 187, 169, 105; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{15}H_{16}BrN_2O$ 319.0441, found 319.0436.



N'-(3,4-Dibromophenyl)-N'-methylbenzohydrazide ***(37)***. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 73% yield (28.0 mg, 0.07 mmol); mp 134.2-135.4 ℃; IR (KBr, cm^{-1}) 3260, 2922, 2811, 1653, 1583, 1475, 1308, 1224, 1113, 1000, 902, 805, 728, 689, 572; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.83 (s, 1H), 7.91 (d, J = 8.0 Hz, 2H), 7.60 (t, J = 8.0 Hz, 1H), 7.52 (t, J = 8.0 Hz, 3H), 7.11 (s, 1H), 6.77 (dd, J = 8.0 Hz, 4.0 Hz, 1H), 3.21 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.4, 150.3, 133.5, 132.4, 132.0, 128.6, 127.4, 124.2, 116.4, 113.5, 110.9, 40.1; MS (EI, 70 eV) *m/z* 384, 279, 263, 235, 155, 105; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{13}Br_2N_2O$ 382.9389, found 382.9384.

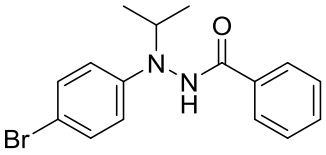


N'-(4-Bromophenyl)-N'-butylbenzohydrazide ***(38)***. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 74% yield (25.6 mg, 0.07mmol); mp 148.7-149.8 ℃; IR (KBr, cm^{-1}) 3258, 3057, 2924, 2867, 1657, 1525, 1489, 1307, 1285, 1121, 1027, 997, 803, 714, 690, 501; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.54 (s, 1H), 7.91 (d, J = 8.0 Hz, 2H), 7.60 (t, J = 8.0 Hz, 1H), 7.52 (t, J = 8.0 Hz, 2H), 7.33 (d, J = 8.0 Hz, 2H), 6.76 (d, J = 12.0 Hz, 2H), 3.49 (t, J = 8.0 Hz, 2H), 1.62-1.54 (m, 2H), 1.44-1.34 (m, 2H), 0.90 (t, J = 8.0 Hz, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.6, 148.6, 132.7, 131.8, 131.4, 128.5, 127.4, 114.2, 108.9, 51.0, 28.8, 19.6, 13.9; MS (EI, 70 eV) *m/z* 346, 303, 227, 199, 157, 105; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{17}H_{20}BrN_2O$ 347.0754, found 347.0750.

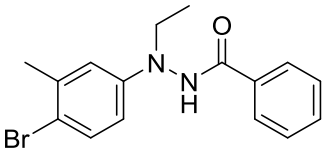


N'-(4-Bromophenyl)-N'-propylbenzohydrazide ***(39)***. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 74% yield (24.6 mg, 0.07 mmol); mp

126.0-127.8 ℃; IR (KBr, cm^{-1}) 3255, 3058, 2963, 2873, 1659, 1526, 1490, 1382, 1285, 1123, 1076, 998, 814, 718, 692, 504; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.55 (s, 1H), 7.92 (d, J = 4.0 Hz, 2H), 7.60 (t, J = 8.0 Hz, 1H), 7.51 (t, J = 8.0 Hz, 2H), 7.33 (d, J = 8.0 Hz, 2H), 6.77 (d, J = 8.0 Hz, 2H), 3.46 (t, J = 8.0 Hz, 2H), 1.64-1.59 (m, 2H), 0.95 (t, J = 8.0 Hz, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.6, 148.7, 132.7, 131.9, 131.4, 128.5, 127.4, 114.2, 109.0, 53.0, 20.0, 11.3; MS (EI, 70 eV) *m/z* 332, 303, 227, 211, 185, 155, 132, 105; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{16}H_{18}BrN_2O$ 333.0597, found 333.0613.

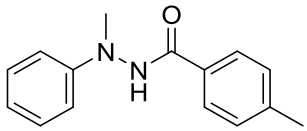


N'-(4-Bromophenyl)-N'-isopropylbenzohydrazide ***(40)***. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Brown solid: 65% yield (21.6 mg, 0.07 mmol); mp 122.6-123.6 ℃; IR (KBr, cm^{-1}) 3268, 3048, 2926, 2853, 1653, 1531, 1491, 1366, 1281, 1135, 1077, 996, 814, 717, 694, 545; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.10 (s, 1H), 7.93 (d, J = 8.0 Hz, 2H), 7.60 (t, J = 8.0 Hz, 1H), 7.52 (t, J = 8.0 Hz, 2H), 7.33 (d, J = 8.0 Hz, 2H), 6.74 (d, J = 8.0 Hz, 2H), 4.27-4.21 (m, 1H), 1.15 (d, J = 4.0 Hz, 6H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 166.9, 147.3, 132.9, 131.8, 131.5, 128.5, 127.6, 114.5, 108.6, 49.8, 18.4; MS (EI, 70 eV) *m/z* 334, 275, 229, 213, 199, 185, 155, 105; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{16}H_{18}BrN_2O$ 333.0597, found 333.0590.

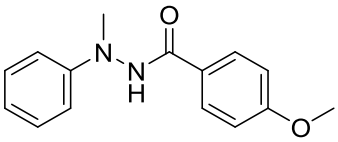


N'-(4-Bromo-3-methylphenyl)-N'-ethylbenzohydrazide ***(41)***. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellowish solid: 61% yield (20.3 mg, 0.06 mmol); mp 127.5-128.1 ℃; IR (KBr, cm^{-1}) 3272, 3058, 2984, 2923, 2873, 1651, 1523, 1484, 1349, 1288, 1190, 1021, 904, 849, 715, 694, 591; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.50 (s, 1H), 7.92 (d, J = 8.0 Hz, 2H), 7.60 (t, J = 8.0 Hz, 1H), 7.51 (t, J = 8.0 Hz, 2H), 7.33 (d, J = 8.0 Hz, 1H), 6.81 (d, J = 4.0 Hz, 1H), 6.60 (dd, J = 8.0 Hz,

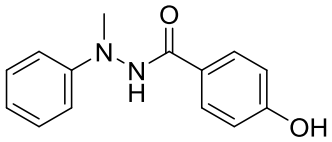
4.0 Hz, 1H), 3.55 (q, *J* = 8.0 Hz, 2H), 2.27 (s, 3H), 1.16 (t, *J* = 8.0 Hz, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.8, 148.6, 137.2, 132.8, 132.2, 131.8, 128.5, 127.4, 114.7, 112.1, 111.9, 45.4, 22.8, 11.9; MS (EI, 70 eV) *m/z* 346, 303, 227, 199, 157, 105; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{16}H_{18}BrN_2O$ 333.0597, found 333.0594.



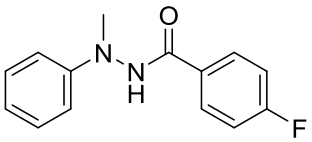
N',4-Dimethyl-N'-phenylbenzohydrazide (***42***).[3] Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 49% yield (11.8 mg, 0.05 mmol); mp 116.5-117.3 ℃; IR (KBr, cm^{-1}) 3253, 3028, 2920, 2869, 1652, 1599, 1454, 1301, 1228, 1110, 1028, 993, 900, 820, 748, 690, 510; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.56 (s, 1H), 7.82 (d, *J* = 8.0 Hz, 2H), 7.31 (d, *J* = 8.0 Hz, 2H), 7.20 (t, *J* = 8.0 Hz, 2H), 6.81 (d, *J* = 8.0 Hz, 2H), 6.75 (t, *J* = 8.0 Hz, 1H), 3.20 (s, 3H), 2.37 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.4, 149.9, 141.7, 130.1, 129.0, 128.8, 127.3, 118.1, 112.2, 40.1, 21.0; MS (EI, 70 eV) *m/z* 240, 136, 119, 105, 91; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{15}H_{17}N_2O$ 241.1335, found 241.1331.



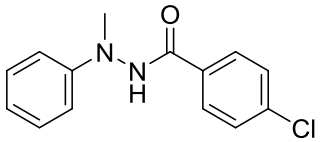
4-Methoxy-N'-methyl-N'-phenylbenzohydrazide (***43***).[3] Eluent: petroleum ether/ethyl acetate (v/v = 3/1); Yellow solid: 47% yield (12.0 mg, 0.05 mmol); mp 158.5-159.6 ℃; IR (KBr, cm^{-1}) 3262, 3004, 2931, 2839, 1649, 1503, 1454, 1255, 1184, 1029, 994, 891, 752, 691, 541; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.50 (s, 1H), 7.90 (d, *J* = 8.0 Hz, 2H), 7.20 (t, *J* = 8.0 Hz, 2H), 7.04 (d, *J* = 8.0 Hz, 2H), 6.80 (d, *J* = 8.0 Hz, 2H), 6.74 (t, *J* = 8.0 Hz, 1H), 3.83 (s, 3H), 3.19 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.0, 162.0, 150.0, 129.2, 128.8, 125.0, 118.0, 113.7, 112.2, 55.4, 40.2; MS (EI, 70 eV) *m/z* 256, 151, 135, 105, 92, 77; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{15}H_{17}N_2O_2$ 257.1285, found 257.1280.



*4-Hydroxy-N'-methyl-N'-phenylbenzohydrazide (**44**).* Eluent: petroleum ether/ethyl acetate (v/v = 1/1); Brown solid: 40% yield (9.7 mg, 0.04 mmol); mp 77.6-78.2 ℃; IR (KBr, cm^{-1}) 3228, 1659, 1606, 1503, 1452, 1376, 1280, 1174, 1025, 902, 851, 755, 693, 543; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.39 (s, 1H), 10.09 (s, 1H), 7.78 (d, J = 8.0 Hz, 2H), 7.19 (t, J = 8.0 Hz, 2H), 6.84 (d, J = 8.0 Hz, 2H), 6.79 (d, J = 8.0 Hz, 2H), 6.73 (t, J = 8.0 Hz, 1H), 3.18 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.2, 160.6, 150.1, 129.3, 128.8, 123.5, 117.9, 115.0, 112.2, 40.2; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{15}N_2O_2$ 243.1128, found 243.1124.

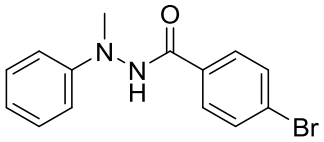


*4-Fluoro-N'-methyl-N'-phenylbenzohydrazide (**45**).* Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 55% yield (13.4 mg, 0.06 mmol); mp 149.8-150.4 ℃; IR (KBr, cm^{-1}) 3269, 3067, 2922, 2810, 1655, 1501, 1459, 1317, 1234, 1118, 1095, 995, 894, 751, 692, 537; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.67 (s, 1H), 7.98 (dd, J = 8.0 Hz, 4.0 Hz, 2H), 7.35 (t, J = 8.0 Hz, 2H), 7.21 (t, J = 8.0 Hz, 2H), 6.82 (d, J = 8.0 Hz, 2H), 6.76 (t, J = 8.0 Hz, 1H), 3.20 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 164.5, 165.2 (d, J = 248.0 Hz), 149.9, 130.1 (d, J = 9.0 Hz), 129.4 (d, J = 3.0 Hz), 128.9, 118.3, 115.5 (d, J = 22.0 Hz), 112.3, 40.1; MS (EI, 70 eV) *m/z* 244, 214, 123, 121, 105, 77; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{14}FN_2O$ 245.1085, found *m/z* 245.1080.

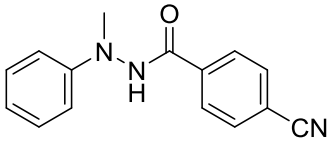


*4-Chloro-N'-methyl-N'-phenylbenzohydrazide (**46**).*[3] Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 46% yield (12.0 mg, 0.05 mmol); mp 164.1-165.7 ℃; IR (KBr, cm^{-1}) 3344, 3057, 2968, 2868, 1652, 1597, 1496, 1311,

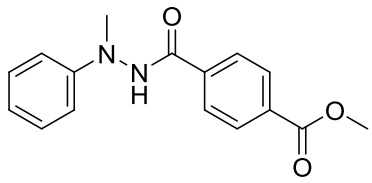
1294, 1139, 1094, 973, 892, 749, 690, 525; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.73 (s, 1H), 7.95 (d, J = 8.0 Hz, 2H), 7.59 (d, J = 8.0 Hz, 2H), 7.21 (t, J = 8.0 Hz, 2H), 6.83 (d, J = 8.0 Hz, 2H), 6.77 (t, J = 8.0 Hz, 1H), 3.21 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 164.5, 149.8, 136.6, 131.7, 129.3, 128.9, 128.6, 118.3, 112.3, 40.1; MS (EI, 70 eV) *m/z* 260, 139, 121, 105, 92, 77; HRMS (ESI) *m/z* [M + H]$^{+}$ Calcd for $C_{14}H_{14}ClN_2O$ 261.0789, found 261.0784.



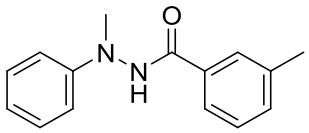
*4-Bromo-N'-methyl-N'-phenylbenzohydrazide (**47**)*. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 43% yield (13.1 mg, 0.04 mmol); mp 172.5-173.3 ℃; IR (KBr, cm^{-1}) 3254, 3039, 2876, 1655, 1527, 1480, 1301, 1102, 1057, 995, 892, 749, 689, 548; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.73 (s, 1H), 7.87 (d, J = 8.0 Hz, 2H), 7.73 (d, J = 8.0 Hz, 2H), 7.21 (t, J = 8.0 Hz, 2H), 6.82 (d, J = 4.0 Hz, 2H), 6.77 (t, J = 8.0 Hz, 1H), 3.20 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 164.6, 149.8, 132.0, 131.5, 129.5, 128.8, 125.5, 118.3, 112.3, 40.1; MS (EI, 70 eV) *m/z* 304, 183, 155, 121, 105, 77; HRMS (ESI) *m/z* [M + H]$^{+}$ Calcd for $C_{14}H_{14}BrN_2O$ 305.0284, found 305.0280.



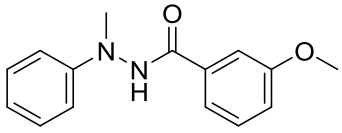
*4-Cyano-N'-methyl-N'-phenylbenzohydrazide (**48**)*. Eluent: petroleum ether/ethyl acetate (v/v = 1/1); Yellow solid: 37% yield (9.3 mg, 0.04 mmol); mp 149.5-150.1 ℃; IR (KBr, cm^{-1}) 3243, 3090, 2924, 2879, 2232, 1661, 1526, 1496, 1302, 1212, 1115, 1019, 900, 864, 769, 692, 527; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.87 (s, 1H), 8.06 (d, J = 8.0 Hz, 2H), 8.01 (d, J = 8.0 Hz, 2H), 7.22 (t, J = 8.0 Hz, 2H), 6.84 (d, J = 8.0 Hz, 2H), 6.78 (t, J = 8.0 Hz, 1H), 3.21 (s, 3H).; ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 164.2, 149.6, 136.9, 132.6, 128.9, 128.2, 118.5, 118.2, 114.1, 112.4, 40.1; MS (EI, 70 eV) *m/z* 251, 130, 121, 105, 92, 77; HRMS (ESI) *m/z* [M + H]$^{+}$ Calcd for $C_{15}H_{14}N_3O$ 252.1131, found 252.1128.



*Methyl 4-(2-methyl-2-phenylhydrazine-1-carbonyl)benzoate (**49**).* Eluent: petroleum ether/ethyl acetate (v/v = 3/1); Brown solid: 40% yield (11.3 mg, 0.04 mmol); mp 124.7-125.8 ℃; IR (KBr, cm^{-1}) 3250, 3058, 2951, 2867, 1723, 1658, 1537, 1436, 1284, 1112, 1018, 967, 895, 795, 691, 543; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.81 (s, 1H), 8.08 (d, J = 8.0 Hz, 2H), 8.02 (d, J = 12.0 Hz, 2H), 7.22 (t, J = 8.0 Hz, 2H), 6.83 (d, J = 8.0 Hz, 2H), 6.77 (t, J = 8.0 Hz, 1H), 3.90 (s, 3H), 3.21 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.6, 164.7, 149.7, 137.1, 132.2, 129.3, 128.9, 127.8, 118.4, 112.4, 52.4, 40.1; MS (EI, 70 eV) *m/z* 284, 253, 163, 135, 121, 105; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{16}H_{17}N_2O_3$ 285.1234, found 285.1229.

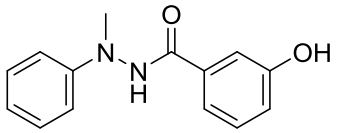


*N',3-Dimethyl-N'-phenylbenzohydrazide (**50**).* Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 37% yield (8.9 mg, 0.04 mmol); mp 114.5-114.3 ℃; IR (KBr, cm^{-1}) 3251, 3026, 2922, 2815, 1649, 1599, 1498, 1301, 1237, 1184, 1027, 929, 818, 746, 689, 548; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.58 (s, 1H), 7.74 (s, 1H), 7.72-7.69 (m, 1H), 7.40 (d, J = 8.0 Hz, 2H), 7.21 (t, J = 8.0 Hz, 2H), 6.82 (d, J = 8.0 Hz, 2H), 6.76 (t, J = 8.0 Hz, 1H), 3.20 (s, 3H), 2.38 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.6, 149.9, 137.8, 132.9, 132.3, 128.8, 128.4, 127.9, 124.4, 118.1, 112.3, 40.1, 20.9; MS (EI, 70 eV) *m/z* 240, 207, 133, 119, 105, 91; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{15}H_{17}N_2O$ 241.1335, found 241.1332.

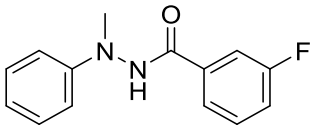


*3-Methoxy-N'-methyl-N'-phenylbenzohydrazide (**51**).* Eluent: petroleum ether/ethyl

acetate (v/v = 3/1); Yellow solid: 35% yield (9.0 mg, 0.04 mmol); mp 129.7-130.8 ℃; IR (KBr, cm^{-1}) 3200, 3011, 2970, 2868, 1652, 1599, 1460, 1325, 1297, 1183, 1032, 938, 873, 749, 693, 546; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.63 (s, 1H), 7.51 (d, J = 8.0 Hz, 1H), 7.47 (s, 1H), 7.42 (t, J = 8.0 Hz, 1H), 7.21 (t, J = 8.0 Hz, 2H), 7.15 (d, J = 8.0 Hz, Hz, 1H), 6.82 (d, J = 8.0 Hz, 2H), 6.76 (d, J = 8.0 Hz, 1H), 3.82 (s, 3H), 3.21 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 165.2, 159.2, 149.9, 134.2, 129.6, 128.8, 119.5, 118.2, 117.7, 112.4, 112.3, 55.3, 40.1; MS (EI, 70 eV) *m/z* 256, 152, 135, 121, 105, 92; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{15}H_{17}N_2O_2$ 257.1285, found 257.1283.

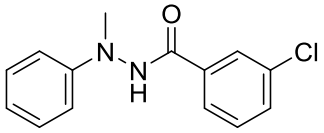


*3-Hydroxy-N'-methyl-N'-phenylbenzohydrazide (**52**)*. Eluent: petroleum ether/ethyl acetate (v/v = 1/1); Yellow solid: 44% yield (10.6 mg, 0.04 mmol); mp 97.1-98.2 ℃; IR (KBr, cm^{-1}) 3224, 2821, 1658, 1599, 1497, 1300, 1241, 1115, 1025, 998, 844, 752, 692, 525; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.53 (s, 1H), 9.74 (s, 1H), 7.35 (d, J = 8.0 Hz, 1H), 7.30 (t, J = 8.0 Hz, 2H), 7.21 (t, J = 8.0 Hz, 2H), 6.98 (d, J = 8.0 Hz, 1H), 6.80 (d, J = 8.0 Hz, 2H), 6.75 (t, J = 8.0 Hz, 1H), 3.19 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 165.6, 157.4, 149.9, 134.4, 129.5, 128.9, 118.7, 118.1, 117.8, 114.3, 112.2, 40.1; MS (EI, 70 eV) *m/z* 346, 303, 227, 199, 157, 105; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{15}N_2O_2$ 243.1128, found 243.1126.

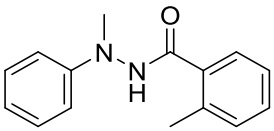


*3-Fluoro-N'-methyl-N'-phenylbenzohydrazide (**53**)*. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Gray solid: 40% of yield (9.8 mg, 0.04 mmol); mp 103.1-104.0 ℃; IR (KBr, cm^{-1}) 3267, 3069, 2921, 2874, 1657, 1588, 1483, 1317, 1295, 1124, 1031, 998, 837, 751, 635, 528; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.73 (s, 1H), 7.77 (d, J = 8.0 Hz, 1H), 7.71 (d, J = 8.0 Hz, 1H), 7.58 (q, J = 8.0 Hz, 1H), 7.45 (t, J = 8.0 Hz, 1H), 7.22 (t, J = 8.0 Hz, 2H), 6.83 (d, J = 8.0 Hz, 2H), 6.77 (t, J = 8.0 Hz, 1H),

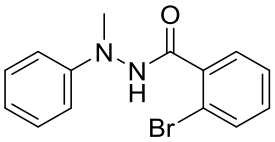
3.21 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 164.2 (d, J = 3.0 Hz), 161.9 (d, J = 243.0 Hz), 149.7, 135.2 (d, J = 7.0 Hz), 130.7 (d, J = 8.0 Hz), 128.8, 123.5 (d, J = 3.0 Hz), 118.6 (d, J = 21.0 Hz), 118.3, 114.2 (d, J = 23.0 Hz), 112.3, 40.1; MS (EI, 70 eV) *m/z* 244, 123, 121, 105, 92, 77; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{14}FN_2O$ 245.1085, found 245.1083.



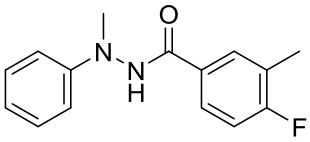
*3-Chloro-N'-methyl-N'-phenylbenzohydrazide (**54**)*. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 46% yield (12.0 mg, 0.05 mmol); mp 111.0-112.2 ℃; IR (KBr, cm^{-1}) 3249, 3028, 2926, 2819, 1649, 1599, 1465, 1339, 1297, 1184, 1088, 912, 810, 765, 680, 500; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.76 (s, 1H), 7.97 (s, 1H), 7.88 (d, J = 8.0 Hz, 1H), 7.66 (d, J = 8.0 Hz, 1H), 7.55 (t, J = 8.0 Hz, 1H), 7.22 (t, J = 8.0 Hz, 2H), 6.83 (d, J = 8.0 Hz, 2H), 6.77 (t, J = 8.0 Hz, 1H), 3.21 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 164.2, 149.7, 134.9, 133.4, 131.6, 130.5, 128.9, 127.2, 126.1, 118.4, 112.4, 40.1; MS (EI, 70 eV) *m/z* 260, 139, 121, 105, 92, 77; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{14}ClN_2O$ 261.0789, found 261.0785.



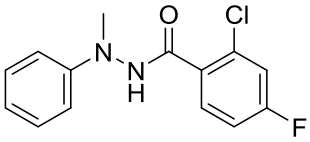
*N',2-Dimethyl-N'-phenylbenzohydrazide (**55**)*. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 30% yield (7.2 mg, 0.03 mmol); mp 106.8-107.9 ℃; IR (KBr, cm^{-1}) 3258, 3062, 2927, 2878, 1651, 1597, 1498, 1303, 1285, 1115, 1089, 993, 899, 748, 693, 510; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.31 (s, 1H), 7.48 (d, J = 8.0 Hz, 1H), 7.39 (t, J = 8.0 Hz, 1H), 7.30 (d, J = 8.0 Hz, 2H), 7.24 (t, J = 8.0 Hz, 2H), 6.87 (d, J = 8.0 Hz, 2H), 6.78 (t, J = 8.0 Hz, 1H), 3.21 (s, 3H), 2.40 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 168.1, 149.8, 135.7, 135.0, 130.6, 129.8, 128.8, 127.2, 125.6, 118.2, 112.3, 40.1, 19.2; MS (EI, 70 eV) *m/z* 240, 136, 199, 105, 91; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{15}H_{17}N_2O$ 241.1335, found 241.1333.



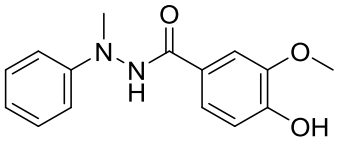
*2-Bromo-N'-methyl-N'-phenylbenzohydrazide (**56**)*. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 28% yield (8.5 mg, 0.03 mmol); mp 125.4-126.5 ℃; IR (KBr, cm^{-1}) 3246, 3016, 2956, 2807, 1654, 1521, 1493, 1303, 1261, 1103, 1030, 994, 899, 747, 694, 520; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.45 (s, 1H), 7.71 (d, J = 8.0 Hz, 1H), 7.57 (d, J = 8.0 Hz, 1H), 7.50 (t, J = 8.0 Hz, 1H), 7.42 (t, J = 8.0 Hz, 1H), 7.23 (t, J = 8.0 Hz, 2H), 6.93 (d, J = 8.0 Hz, 2H), 6.79 (t, J = 8.0 Hz, 1H), 3.21 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 166.2, 149.6, 137.2, 132.8, 131.4, 129.0, 128.8, 127.7, 119.2, 118.4, 112.5, 40.0; MS (EI, 70 eV) *m/z* 304, 183, 155, 121, 105, 92; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{14}BrN_2O$ 305.0284, found 305.0280.



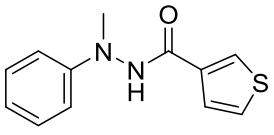
*4-Fluoro-N',3-dimethyl-N'-phenylbenzohydrazide (**57**)*. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Brown solid: 41% yield (10.6 mg, 0.04 mmol); mp 113.4-114.2 ℃; IR (KBr, cm^{-1}) 3264, 3078, 2924, 2875, 1658, 1598, 1497, 1313, 1263, 1120, 1030, 845, 750, 689, 518; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.60 (s, 1H), 7.88 (d, J = 8.0 Hz, 1H), 7.81-7.77 (m, 1H), 7.27 (t, J = 8.0 Hz, 1H), 7.21 (t, J = 8.0 Hz, 2H), 6.82 (d, J = 8.0 Hz, 2H), 6.76 (t, J = 8.0 Hz, 1H), 3.20 (s, 3H), 2.30 (d, J = 4.0 Hz, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 164.6, 162.7 (d, J = 246.0 Hz), 149.9, 131.2 (d, J = 6.0 Hz), 129.1 (d, J = 3.0 Hz), 128.8, 127.2 (d, J = 9.0 Hz), 124.5 (d, J = 18.0 Hz), 118.2, 115.0 (d, J = 23.0 Hz), 112.3, 40.1, 14.1 (d, J = 4.0 Hz); MS (EI, 70 eV) *m/z* 258, 154, 137, 121, 105; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{15}H_{16}FN_2O$ 259.1241, found 259.1237.



2-Chloro-4-fluoro-N'-methyl-N'-phenylbenzohydrazide ***(58)***. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Brown solid: 39% yield (10.8 mg, 0.04 mmol); mp 115.8-116.8 ℃; IR (KBr, cm^{-1}) 3176, 2996, 2809, 1657, 1599, 1490, 1308, 1262, 1113, 1099, 919, 878, 746, 690, 529; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.49 (s, 1H), 7.68 (dd, J = 8.0 Hz, 4.0 Hz, 1H), 7.58 (dd, J = 8.0 Hz, 4.0 Hz, 1H), 7.35 (t, J = 8.0 Hz, 1H), 7.25 (t, J = 8.0 Hz, 2H), 6.91 (d, J = 8.0 Hz, 2H), 6.80 (t, J = 8.0 Hz, 1H), 3.21 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 164.7, 162.3 (d, J = 249.0 Hz), 149.5, 131.7 (d, J = 6.0 Hz), 131.6 (d, J = 2.0 Hz), 130.9 (d, J = 10.0 Hz), 128.8, 118.5, 117.2 (d, J = 25.0 Hz), 114.5 (d, J = 21.0 Hz), 112.5, 40.0; MS (EI, 70 eV) *m/z* 278, 157, 129, 121, 105; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{13}ClFN_2O$ 279.0695, found 279.0692.

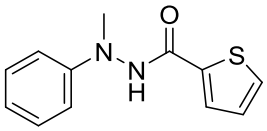


4-Hydroxy-3-methoxy-N'-methyl-N'-phenylbenzohydrazide ***(59)***. Eluent: petroleum ether/ethyl acetate (v/v = 1/1); Yellow solid: 25% yield (6.8 mg, 0.03 mmol); mp 118.0-119.1 ℃; IR (KBr, cm^{-1}) 3051, 2259, 2130, 1657, 1297, 1154, 1028, 824, 762, 626, 521; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.42 (s, 1H), 9.69 (s, 1H), 7.48 (s, 1H), 7.42 (d, J = 8.0 Hz, 1H), 7.20 (t, J = 8.0 Hz, 2H), 6.85 (d, J = 8.0 Hz, 1H), 6.79 (d, J = 8.0 Hz, 2H), 6.74 (t, J = 8.0 Hz, 1H), 3.82 (s, 3H), 3.18 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.1, 150.1, 150.1, 147.3, 128.8, 123.7, 121.0, 118.0, 115.0, 112.2, 111.3, 55.7, 40.2; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{15}H_{17}N_2O_3$ 273.1234, found 273.1231.

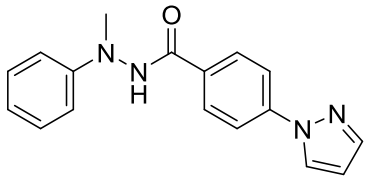


N'-Methyl-N'-phenylthiophene-3-carbohydrazide ***(60)***. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); White solid: 28% yield (6.5 mg, 0.03 mmol); mp 183.3-184.5 ℃; IR (KBr, cm^{-1}) 3235, 3070, 2971, 2872, 1649, 1538, 1453, 1287, 1114, 1029, 951, 848, 751, 690, 530; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.49 (s,

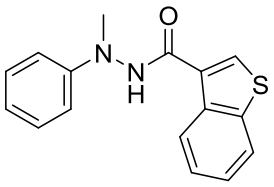
1H), 8.26 (d, *J* = 4.0 Hz, 1H), 7.65 (d, *J* = 8.0 Hz, 1H), 7.57 (d, *J* = 4.0 Hz, 1H), 7.21 (t, *J* = 8.0 Hz, 2H), 6.81 (d, *J* = 8.0 Hz, 2H), 6.76 (t, *J* = 8.0 Hz, 1H), 3.20 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 161.2, 149.9, 135.7, 129.4, 128.8, 127.1, 126.7, 118.2, 112.2, 40.2; MS (EI, 70 eV) *m/z* 232, 128, 121, 111, 105, 92; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{12}H_{13}N_2OS$ 233.0743, found 233.0742.



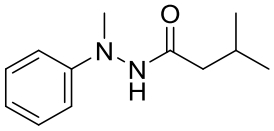
N'-Methyl-N'-phenylthiophene-2-carbohydrazide ***(61)***. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Brown solid: 29% yield (6.7 mg, 0.03 mmol); mp 134.5-135.6 ℃; IR (KBr, cm^{-1}) 3243, 3081, 2921, 2873, 1642, 1535, 1416, 1356, 1293, 1115, 1029, 974, 851, 747, 630, 521; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.67 (s, 1H), 7.88 (d, *J* = 4.0 Hz, 1H), 7.84 (d, *J* = 4.0 Hz, 1H), 7.24-7.19 (m, 3H), 6.82 (d, *J* = 8.0 Hz, 2H), 6.77 (t, *J* = 8.0 Hz, 1H), 3.20 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 160.5, 149.8, 137.6, 131.5, 128.9, 128.7, 128.1, 118.4, 112.3, 40.3; MS (EI, 70 eV) *m/z* 232, 216, 121, 111, 105, 92; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{12}H_{13}N_2OS$ 233.0743, found 233.0743.



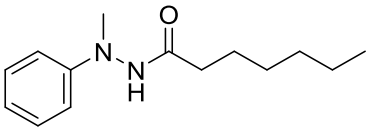
N'-Methyl-N'-phenyl-4-(1H-pyrazol-1-yl)benzohydrazide ***(62)***. Eluent: petroleum ether/ethyl acetate (v/v = 3/1); Yellow solid: 38% yield (11.1 mg, 0.04 mmol); mp 141.8-142.9 ℃; IR (KBr, cm^{-1}) 3262, 3148, 3070, 2919, 2815, 1653, 1501, 1446, 1336, 1204, 1122, 1047, 937, 850, 749, 651, 515; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.70 (s, 1H), 8.62 (d, *J* = 4.0 Hz, 1H), 8.03 (q, *J* = 8.0 Hz, 4H), 7.81 (d, *J* = 4.0 Hz, 1H), 7.22 (t, *J* = 8.0 Hz, 2H), 6.84 (d, *J* = 8.0 Hz, 2H) 6.80-6.75 (m, 1H), 6.60 (d, *J* = 4.0 Hz, 1H), 3.22 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 164.7, 149.9, 141.9, 141.7, 130.2, 128.9, 128.9, 128.1, 118.2, 117.8, 112.3, 108.5, 40.2; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{17}H_{17}N_4O$ 293.1397, found 293.1389.



N'-Methyl-N'-phenylbenzo[b]thiophene-3-carbohydrazide ***(63)***. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Brown solid: 24% yield (6.8 mg, 0.02 mmol); mp 129.8-130.7 ℃; IR (KBr, cm^{-1}) 3283, 3099, 2993, 2878, 1642, 1524, 1420, 1365, 1274, 1185, 1020, 977, 884, 756, 692, 520; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.60 (s, 1H), 8.48 (s, 1H), 8.37 (d, J = 8.0 Hz, 1H), 8.05 (d, J = 8.0 Hz, 1H), 7.48-7.41 (m, 2H), 7.22 (t, J = 8.0 Hz, 2H), 6.86 (d, J = 8.0 Hz, 2H), 6.77 (t, J = 8.0 Hz, 1H), 3.23 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 162.7, 150.1, 139.6, 137.3, 131.7, 129.2, 129.0, 125.4, 125.3, 124.4, 123.0, 118.6, 112.6, 40.5; MS (EI, 70 eV) *m/z* 282, 177, 161, 133, 121, 105; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{16}H_{15}N_2OS$ 283.0900, found 283.0897.

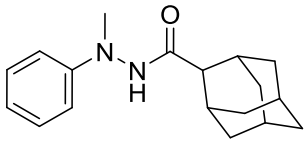


N',3-Dimethyl-N'-phenylbutanehydrazide ***(64)***. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow liquid: 23% yield (4.7 mg, 0.03 mmol); IR (KBr, cm^{-1}) 3236, 3028, 2959, 2871, 1665, 1500, 1465, 1303, 1259, 1115, 1031, 945, 877, 750, 691, 520; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 9.89 (s, 1H), 7.21-7.17 (m, 2H), 6.75-6.71 (m, 3H), 3.07 (s, 3H), 2.02 (d, J = 2.0 Hz, 3H), 0.93 (d, J = 4.0 Hz, 6H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 170.5, 149.8, 128.7, 118.0, 112.1, 42.5, 40.1, 25.4, 22.3; MS (EI, 70 eV) *m/z* 206, 123, 122, 107, 92, 77; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{12}H_{19}N_2O$ 207.1492, found 207.1492.

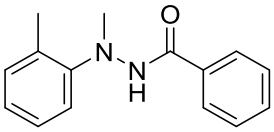


N'-Methyl-N'-phenylheptanehydrazide ***(65)***. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow liquid: 19% yield (4.4 mg, 0.02 mmol); IR (KBr, cm^{-1}) 3239, 3028,

2928, 2858, 1667, 1600, 1500, 1459, 1303, 1280, 1118, 1032, 995, 880, 749, 691, 512; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 9.88 (s, 1H), 7.18 (t, J = 8.0 Hz, 2H), 6.74-6.71 (m, 3H), 3.07 (s, 3H), 2.13 (t, J = 8.0 Hz, 2H), 1.59-1.51 (m, 2H), 1.30-1.28 (m, 6H), 0.90-0.86 (m, 3H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 171.2, 149.8, 128.7, 117.9, 112.1, 40.1, 33.2, 30.9, 28.2, 24.9, 22.0, 13.9; MS (EI, 70 eV) *m/z* 234, 123, 122, 105, 92, 77; HRMS (ESI) *m/z* [M + H]$^{+}$ Calcd for $C_{14}H_{23}N_2O$ 235.1805, found 235.1804.

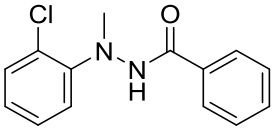


(1r,3r,5r,7r)-N'-Methyl-N'-phenyladamantane-2-carbohydrazide ***(66)***. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); White solid: 51% yield (14.5 mg, 0.05 mmol); mp 158.6-160.6 ℃; IR (KBr, cm^{-1}) 3283, 2903, 2849, 1656, 1599, 1499, 1342, 1272, 1106, 1084, 926, 829, 743, 689, 521; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 9.68 (s, 1H), 7.17 (t, J = 8.0 Hz, 2H), 6.72-6.69 (m, 3H), 3.04 (s, 3H), 1.98-2.00 (m, 3H), 1.87-1.86 (m, 6H), 1.69-1.68 (m, 6H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 175.6, 150.1, 128.7, 117.7, 111.9, 40.0, 39.4, 38.4, 36.1, 27.6; MS (EI, 70 eV) *m/z* 284, 180, 135, 121, 105, 93; HRMS (ESI) *m/z* [M + H]$^{+}$ Calcd for $C_{18}H_{25}N_2O$ 285.1961, found 285.1959.

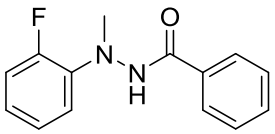


N'-Methyl-N'-(o-tolyl)benzohydrazide ***(67)***. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Reddish brown solid: 56% yield (13.4 mg, 0.06 mmol); mp 130.8-131.8 ℃; IR (KBr, cm^{-1}) 3212, 3058, 2922, 2804, 1651, 1544, 1492, 1312, 1145, 1100, 1027, 967, 898, 769, 696, 577; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.23 (s, 1H), 7.83 (d, J = 8.0 Hz, 2H), 7.53 (t, J = 8.0 Hz, 1H), 7.46 (t, J = 8.0 Hz, 2H), 7.21-7.11 (m, 3H), 6.94 (t, J = 8.0 Hz, 1H), 3.06 (s, 3H), 2.23 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 164.5, 149.0, 133.3, 131.4, 131.0, 130.4, 128.3, 127.3, 126.0, 122.7, 117.8, 41.8, 18.6; MS (EI, 70 eV) *m/z* 240, 135, 119, 105, 91, 77;

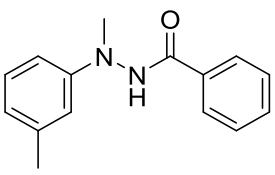
HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{15}H_{17}N_2O$ 241.1335, found 241.1335.



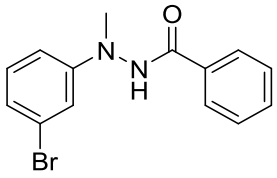
*N'-(2-Chlorophenyl)-N'-methylbenzohydrazide (**68**)*. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 46% yield (12.0 mg, 0.05 mmol); mp 145.0-146.5 ℃; IR (KBr, cm^{-1}) 3212, 3058, 2922, 1651, 1543, 1478, 1314, 1285, 1123, 1049, 966, 898, 764, 695, 575; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 10.52 (s, 1H), 7.83 (d, J = 8.0 Hz, 2H), 7.53 (d, J = 8.0 Hz, 1H), 7.46 (t, J = 8.0 Hz, 2H), 7.36-7.26 (m, 3H), 7.00 (t, J = 8.0 Hz, 1H), 3.17 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 164.7, 147.1, 133.2, 131.5, 130.5, 128.4, 127.4, 127.4, 125.0, 123.5, 120.0, 41.7; MS (EI, 70 eV) *m/z* 260, 224, 155, 139, 105, 77; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{14}ClN_2O$ 261.0789, found 261.0788.



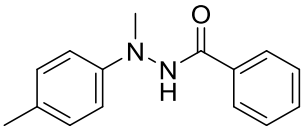
*N'-(2-Fluorophenyl)-N'-methylbenzohydrazide (**69**)*. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 60% yield (14.6 mg, 0.06 mmol); mp 117.6-118.8 ℃; IR (KBr, cm^{-1}) 3239, 3059, 2960, 2889, 1655, 1539, 1498, 1309, 1222, 1125, 1097, 972, 897, 752, 695, 587; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 10.65 (s, 1H), 7.87 (d, J = 8.0 Hz, 2H), 7.56 (t, J = 8.0 Hz, 1H), 7.48 (t, J = 8.0 Hz, 2H), 7.12-7.07 (m, 3H), 6.93-6.88 (m, 1H), 3.22 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.1, 152.5 (d, J = 242.0 Hz), 138.2 (d, J = 7.0 Hz), 133.0, 131.6, 128.4, 127.3, 124.3 (d, J = 3.0 Hz), 121.5 (d, J = 8.0 Hz), 117.7 (d, J = 3.0 Hz), 116.2 (d, J = 20.0 Hz), 42.0 (d, J = 3.0 Hz); MS (EI, 70 eV) *m/z* 244, 139, 123, 105, 95, 77; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{14}FN_2O$ 245.1085, found 245.1085.



*N'-Methyl-N'-(m-tolyl)benzohydrazide (**70**)*. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Reddish brown solid: 45% yield (10.8 mg, 0.05 mmol); mp 96.3-97.4 ℃; IR (KBr, cm^{-1}) 3323, 3057, 2962, 2878, 1650, 1518, 1484, 1292, 1117, 1012, 909, 839, 781, 690, 562; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 10.62 (s, 1H), 7.92 (d, J = 8.0 Hz, 2H), 7.59 (t, J = 8.0 Hz, 1H), 7.51 (t, J = 8.0 Hz, 2H), 7.09 (t, J = 8.0 Hz, 1H), 6.64-6.58 (m, 3H), 3.19 (s, 3H), 2.24 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.4, 150.0, 137.9, 133.0, 131.7, 128.7, 128.5, 127.3, 119.0, 112.8, 109.6, 40.2, 21.4; MS (EI, 70 eV) *m/z* 240, 135, 119, 105, 91, 77; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{15}H_{17}N_2O$ 241.1335, found 241.1335.

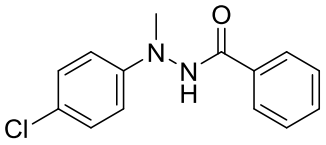


*N'-(3-Bromophenyl)-N'-methylbenzohydrazide (**71**)*. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Reddish brown solid: 39% yield (11.9 mg, 0.04 mmol); mp 95.9-96.6 ℃; IR (KBr, cm^{-1}) 3246, 3059, 2925, 2810, 1649, 1590, 1448, 1322, 1221, 1115, 1088, 989, 836, 767, 688, 561; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 10.76 (s, 1H), 7.92 (d, J = 8.0 Hz, 2H), 7.60 (t, J = 8.0 Hz, 1H), 7.52 (t, J = 8.0 Hz, 2H), 7.16 (t, J = 8.0 Hz, 1H), 6.94-6.90 (m, 2H), 6.81 (dd, J = 8.0 Hz, 4.0 Hz, 1H), 3.21 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.4, 151.4, 132.5, 131.9, 130.7, 128.5, 127.3, 122.3, 120.5, 114.4, 111.2, 40.1; MS (EI, 70 eV) *m/z* 304, 199, 183, 155, 105, 77; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{14}BrN_2O$ 305.0284, found 305.0283.

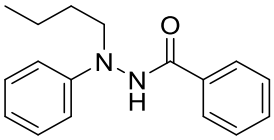


*N'-Methyl-N'-(p-tolyl)benzohydrazide (**72**)*. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 45% yield (10.8 mg, 0.05 mmol); mp 88.5-89.6 ℃; IR (KBr, cm^{-1}) 3263, 3029, 2923, 2853, 1666, 1514, 1494, 1341, 1289, 1117, 1072, 981, 896, 712, 694, 506; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 10.58 (s, 1H), 7.91 (d, J = 8.0 Hz, 2H), 7.58 (t, J = 8.0 Hz, 1H), 7.50 (t, J = 8.0 Hz, 2H), 7.02 (d, J = 8.0 Hz, 2H),

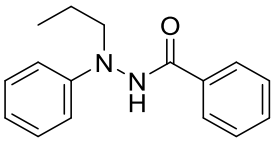
6.74 (d, *J* = 8.0 Hz, 2H), 3.18 (s, 3H), 2.20 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.4, 147.9, 133.0, 131.7, 129.3, 128.4, 127.3, 126.8, 112.5, 40.3, 20.0; MS (EI, 70 eV) *m/z* 240, 135, 119, 105, 91, 77; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{15}H_{17}N_2O$ 241.1335, found 241.1335.



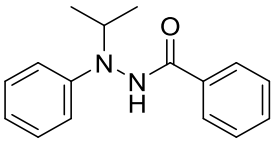
*N'-(4-Chlorophenyl)-N'-methylbenzohydrazide (**73**)*. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 40% yield (10.4 mg, 0.04 mmol); mp 168.4-169.5 ℃; IR (KBr, cm^{-1}) 3240, 3075, 2923, 2852, 1651, 1514, 1494, 1312, 1222, 1137, 1094, 902, 821, 733, 689, 510; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.71 (s, 1H), 7.91 (d, *J* = 8.0 Hz, 2H), 7.59 (t, *J* = 8.0 Hz, 1H), 7.51 (t, *J* = 8.0 Hz, 2H), 7.23 (d, *J* = 8.0 Hz, 2H), 6.82 (d, *J* = 8.0 Hz, 2H), 3.20 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.5, 148.8, 132.7, 131.8, 128.5, 128.5, 127.3, 121.8, 113.8, 40.2; MS (EI, 70 eV) *m/z* 260, 224, 155, 139, 105, 77; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{14}H_{14}ClN_2O$ 261.0789, found 261.0789.



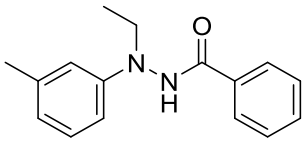
*N'-Butyl-N'-phenylbenzohydrazide (**74**)*. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 51% yield (13.7 mg, 0.05 mmol); mp 124.1-125.2 ℃; IR (KBr, cm^{-1}) 3262, 3056, 2958, 2868, 1658, 1598, 1496, 1377, 1284, 1115, 1028, 938, 897, 750, 691, 510; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.46 (s, 1H), 7.94 (d, *J* = 8.0 Hz, 2H), 7.59 (t, *J* = 8.0 Hz, 1H), 7.52 (t, *J* = 8.0 Hz, 2H), 7.20 (t, *J* = 8.0 Hz, 2H), 6.82 (d, *J* = 8.0 Hz, 2H), 6.73 (t, *J* = 8.0 Hz, 1H), 3.53 (t, *J* = 8.0 Hz, 2H), 1.65-1.57 (m, 2H), 1.46-1.37 (m, 2H), 0.92 (t, *J* = 8.0 Hz, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.7, 149.3, 133.0, 131.7, 128.9, 128.5, 127.4, 117.9, 112.2, 51.0, 29.0, 19.7, 13.9; MS (EI, 70 eV) *m/z* 268, 225, 163, 132, 119, 105; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{17}H_{21}N_2O$ 269.1648, found 269.1648.



*N'-Phenyl-N'-propylbenzohydrazide (**75**).* Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellowish solid: 46% yield (11.7 mg, 0.05mmol); mp 134.5-135.6 ℃; IR (KBr, cm^{-1}) 3259, 3056, 2961, 2873, 1657, 1526, 1496, 1304, 1239, 1138, 1026, 905, 805, 750, 691, 511; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.45 (s, 1H), 7.93 (d, J = 8.0 Hz, 2H), 7.59 (t, J = 8.0 Hz, 1H), 7.51 (t, J = 8.0 Hz, 2H), 7.19 (t, J = 8.0 Hz, 2H), 6.82 (d, J = 8.0 Hz, 2H), 6.73 (t, J = 8.0 Hz, 1H), 3.48 (t, J = 8.0 Hz, 2H), 1.68-1.59 (m, 2H), 0.96 (t, J = 8.0 Hz, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.7, 149.4, 133.0, 131.7, 128.9, 128.5, 127.4, 117.9, 112.2, 53.1, 20.1, 11.4; MS (EI, 70 eV) *m/z* 254, 149, 133, 121, 105, 77; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{16}H_{19}N_2O$ 255.1492, found 255.1492.

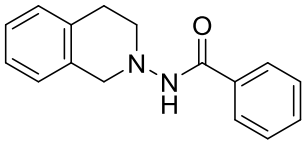


*N'-Isopropyl-N'-phenylbenzohydrazide (**76**).* Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellow solid: 52% yield (13.2 mg, 0.05 mmol); mp 162.8-163.8 ℃; IR (KBr, cm^{-1}) 3263, 3052, 2975, 2873, 1660, 1535, 1492, 1364, 1279, 1129, 1078, 994, 898, 748, 691, 522; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.00 (s, 1H), 7.95 (d, J = 8.0 Hz, 2H), 7.60 (t, J = 8.0 Hz, 1H), 7.52 (t, J = 8.0 Hz, 2H), 7.20 (t, J = 8.0 Hz, 2H), 6.80 (d, J = 8.0 Hz, 2H), 6.72 (t, J = 8.0 Hz, 1H), 4.30-4.24 (m, 1H), 1.17 (d, J = 4.0 Hz, 6H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 167.1, 148.0, 133.2, 131.6, 129.0, 128.4, 127.6, 117.7, 112.6, 49.6, 18.9; MS (EI, 70 eV) *m/z* 254, 239, 207, 149, 133, 107; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{16}H_{19}N_2O$ 255.1492, found 255.1491.

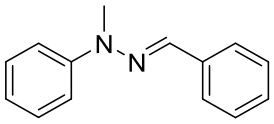


*N'-Ethyl-N'-(m-tolyl)benzohydrazide (**77**).* Eluent: petroleum ether/ethyl acetate

(v/v = 5/1); Yellow solid: 42% yield (10.7 mg, 0.04 mmol); mp 93.1-94.2 ℃; IR (KBr, cm^{-1}) 3263, 3048, 2973, 1657, 1531, 1466, 1344, 1299, 1173, 1071, 902, 846, 772, 692, 555; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.39 (s, 1H), 7.93 (d, J = 8.0 Hz, 2H), 7.59 (t, J = 8.0 Hz, 1H), 7.51 (t, J = 8.0 Hz, 2H), 7.08 (t, J = 8.0 Hz, 1H), 6.64-6.62 (m, 2H), 6.56 (d, J = 4.0 Hz, 1H), 3.56 (q, J = 8.0 Hz, 2H), 2.24 (s, 3H), 1.17 (t, J = 8.0 Hz, 3H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 165.8, 149.1, 137.9, 133.0, 131.7, 128.8, 128.5, 127.4, 118.9, 112.9, 109.7, 45.4, 21.5, 12.0; MS (EI, 70 eV) *m/z* 254, 149, 133, 121, 105, 91, 77; HRMS (ESI) *m/z* [M + H]$^+$ Calcd for $C_{16}H_{19}N_2O$ 255.1492, found 255.1491.

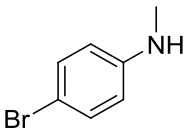


*N-(3,4-Dihydroisoquinolin-2(1H)-yl)benzamide (**78**)*. Eluent: petroleum ether/ethyl acetate (v/v = 5/1); Yellowish solid: 35% yield (8.8 mg, 0.04 mmol); mp 177.6-178.1 ℃; IR (KBr, cm^{-1}) 3213, 3058, 2928, 2853, 1647, 1546, 1491, 1303, 1246, 1183, 1026, 907, 881, 748, 694, 544; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.52 (s, 1H), 7.91 (d, J = 8.0 Hz, 2H), 7.59 (t, J = 8.0 Hz, 1H), 7.51 (t, J = 8.0 Hz, 2H), 6.99-6.95 (m, 2H), 6.66-6.61 (m, 2H), 3.50 (s, 2H), 2.75 (t, J = 8.0 Hz, 2H), 2.06-2.00 (m, 2H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 165.9, 146.4, 133.5, 132.2, 129.2, 129.0, 127.8, 127.2, 122.5, 118.4, 112.1, 50.9, 27.2, 22.4; MS (EI, 70 eV) *m/z* 252, 207, 147, 105, 77; HRMS (ESI) *m/z* [M + H]$^+$ Calcd for $C_{16}H_{17}NO$ 253.1335, found 253.1335.

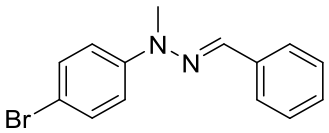


*(E)-2-Benzylidene-1-methyl-1-phenylhydrazine (**80**)*[4]. Eluent: petroleum ether/ethyl acetate (v/v = 20/1); Yellowish solid: 34% yield (7.1 mg, 0.03 mmol); mp 103.2-104.1 ℃; IR (KBr, cm^{-1}) 3096, 3027, 1587, 1502, 1423, 1382, 1240, 1113, 1030, 970, 879, 753, 692, 564; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.74 (d, J = 8.0 Hz, 2H), 7.52 (s, 1H), 7.43-7.34 (m, 6H), 7.30 (t, J = 8.0 Hz, 1H), 6.97 (t, J = 8.0 Hz,

1H), 3.43 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 147.9, 136.8, 131.8, 129.0, 128.5, 127.7, 126.0, 120.5, 115.2, 33.0; MS (EI, 70 eV) *m/z* 210, 195, 167, 133, 106, 77.



4-Bromo-N-methylaniline (***81***)[5]. Eluent: petroleum ether/ethyl acetate (v/v = 20/1); Brown liquid: 36% yield (6.7 mg, 0.04 mmol); IR (KBr, cm^{-1}) 3425, 2927, 2885, 1598, 1449, 1317, 1261, 1178, 1075, 998, 813, 504; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.28 (d, *J* = 8.0 Hz, 2H), 6.49 (d, *J* = 8.0 Hz, 2H), 2.81 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 148.2, 131.7, 113.8, 108.6, 30.6; MS (EI, 70 eV) *m/z* 185, 133, 106, 77.



2-Benzylidene-1-(4-bromophenyl)-1-methylhydrazine (***83***)[4]. Eluent: petroleum ether/ethyl acetate (v/v = 20/1); Yellowish solid: 34% yield (9.8 mg, 0.03 mmol); mp 105.4-106.1 ℃; IR (KBr, cm^{-1}) 3024, 2927, 1588, 1493, 1309, 1238, 1123, 1001, 970, 821, 758, 696, 517; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.73 (d, *J* = 8.0 Hz, 2H), 7.51 (s, 1H), 7.46-7.41 (m, 4H), 7.34 (t, *J* = 8.0 Hz, 1H), 7.28 (d, *J* = 8.0 Hz, 2H), 3.37 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 146.8, 136.3, 132.6, 131.7, 128.6, 127.9, 126.1, 116.5, 112.6, 32.7; MS (EI, 70 eV) *m/z* 288, 209, 184, 155, 144, 105, 77.

References

(1) Guo, W.; Cai, L.; Xie, Z.; Liu, G.; Deng, L.; Zhuo, X.; Zhong, Y.; Zou, X.; Zheng, L.; Fan, X. Photocatalyzed Intermolecular Amination for the Synthesis of Hydrazonamides. *Org. Chem. Front.* **2021**, *8*, 3838-3846.

(2) (a) Wang, H.; Jung, H.; Song, F.; Zhu,S.; Bai, Z.; Chen, D.; He, G.; Chang, S.; Chen, G. Nitrene-mediated intermolecular N-N coupling for efficient synthesis of hydrazides. *Nat. Chem.* **2021**, *13*, 378-385. (b) Audouin, C.; Mestdagh, N.; Lassoie, M.-A.; Houssin, R.; Hénichart, J.-P. N-Aminoindoline derivatives as inhibitors of

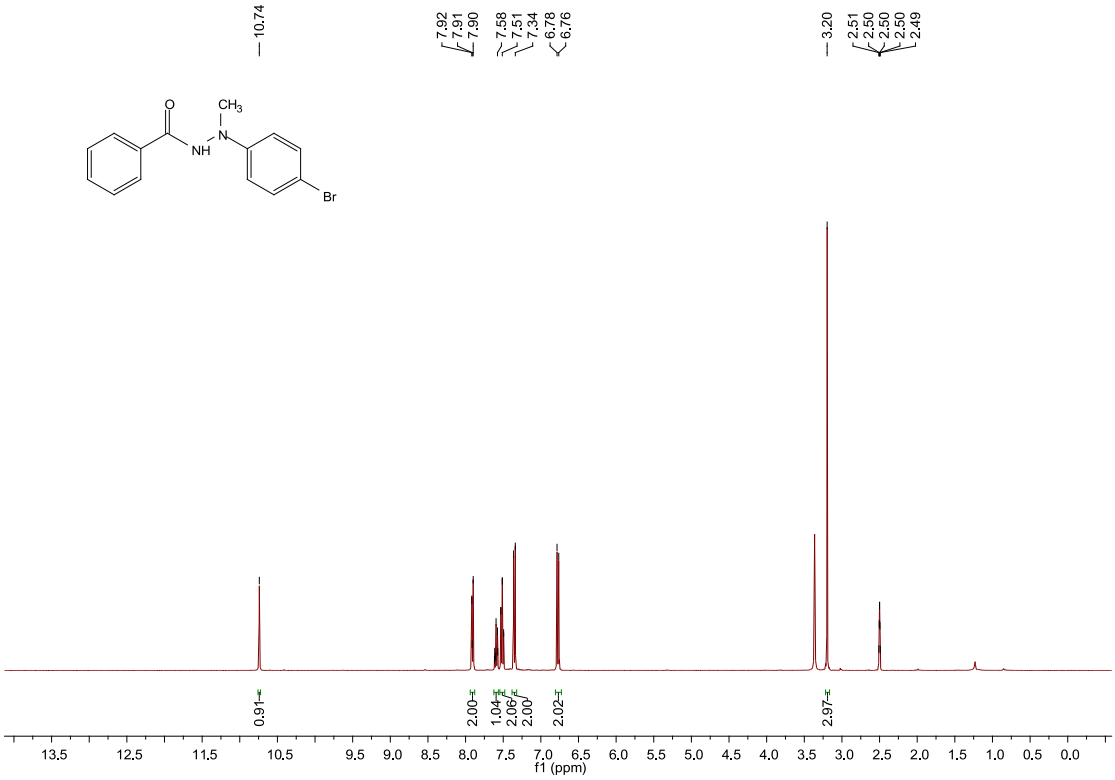
5-lipoxygenase. *Bioorg. & Med. Chem. Lett.* **2001**, *11*, 845-848.

(3) Ghosh, A. K.; Mondal, S.; Hajra, A. Dioxygen-Triggered Oxo-Sulfonylation of Hydrazones. *Org. Lett.* **2020**, *22*, 2771-2775.

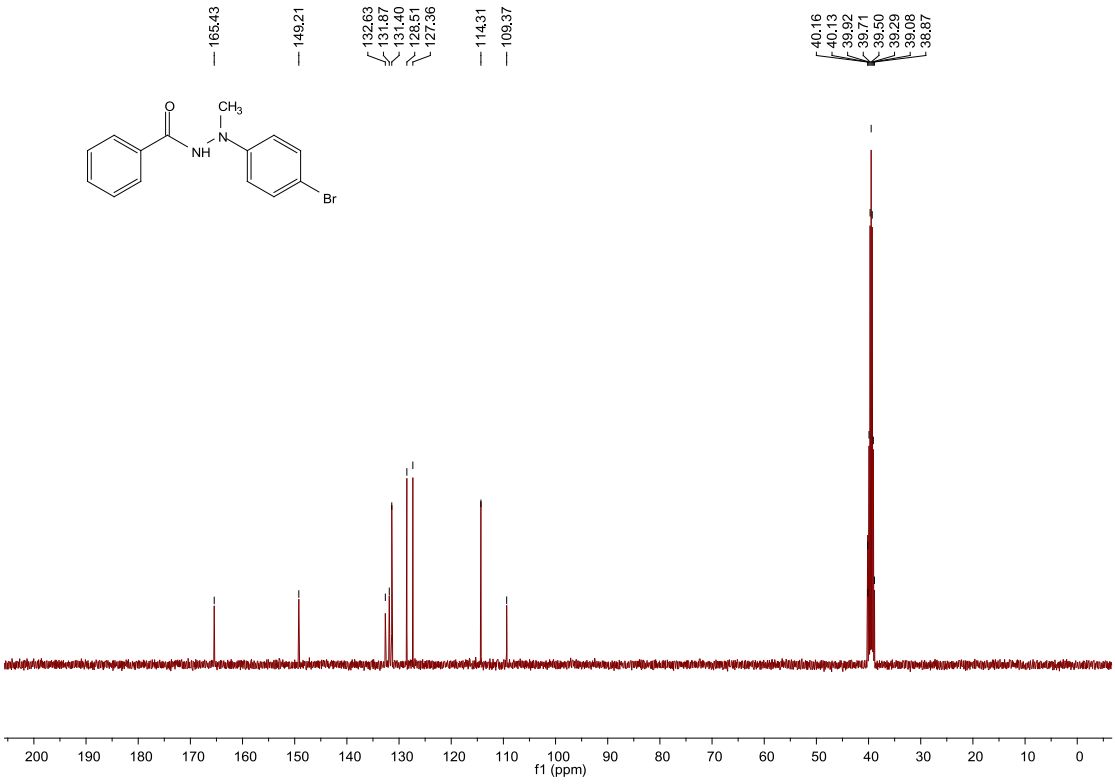
(4) Tan, Z.; Zhang, S.; Zhang, Y.; Li, Y.; Ni, M.; Feng, B. Transition-Metal-Free Trifluoromethylation of Aldehyde Derivatives with Sodium Trifluoromethanesulfinate. *J. Org. Chem.* **2017**, *82*, 9384-9399.

(5) Lator, A.; Gaillard, S.; Poater, A.; Renaud, J.-L. Well-Defined Phosphine-Free Iron-Catalyzed N-Ethylation and N-Methylation of Amines with Ethanol and Methanol. *Org. Lett.* **2018**, *20*, 5985-5990.

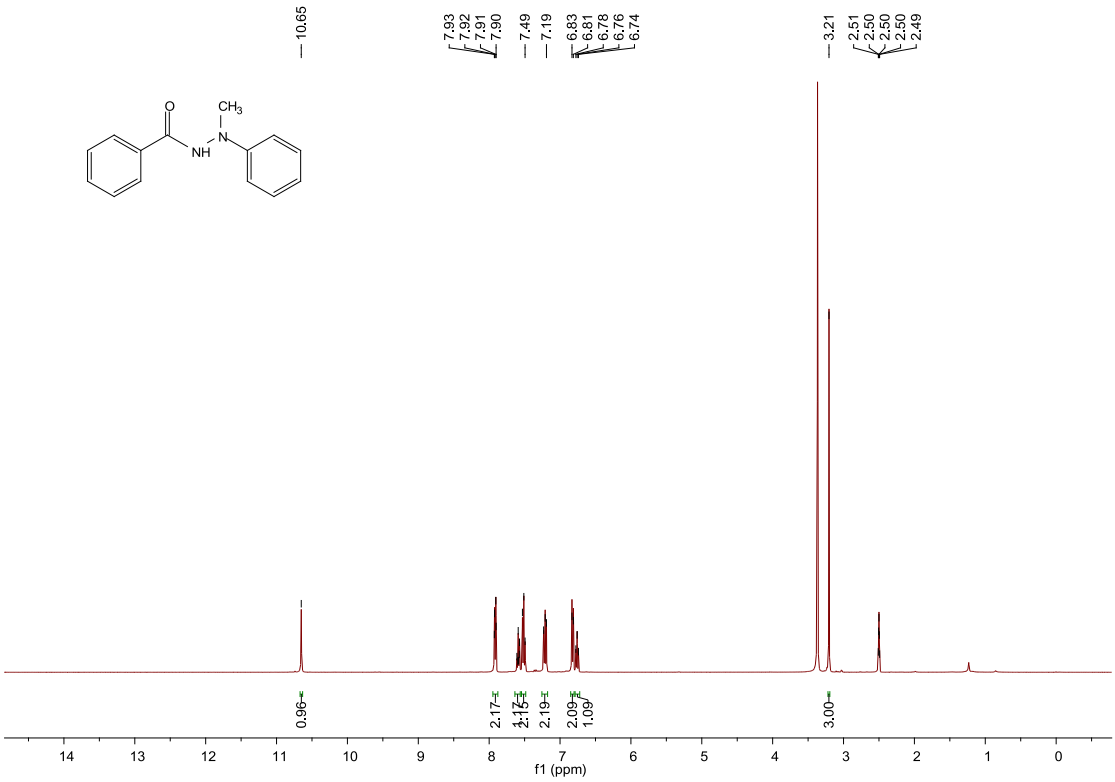
14. NMR Spectra



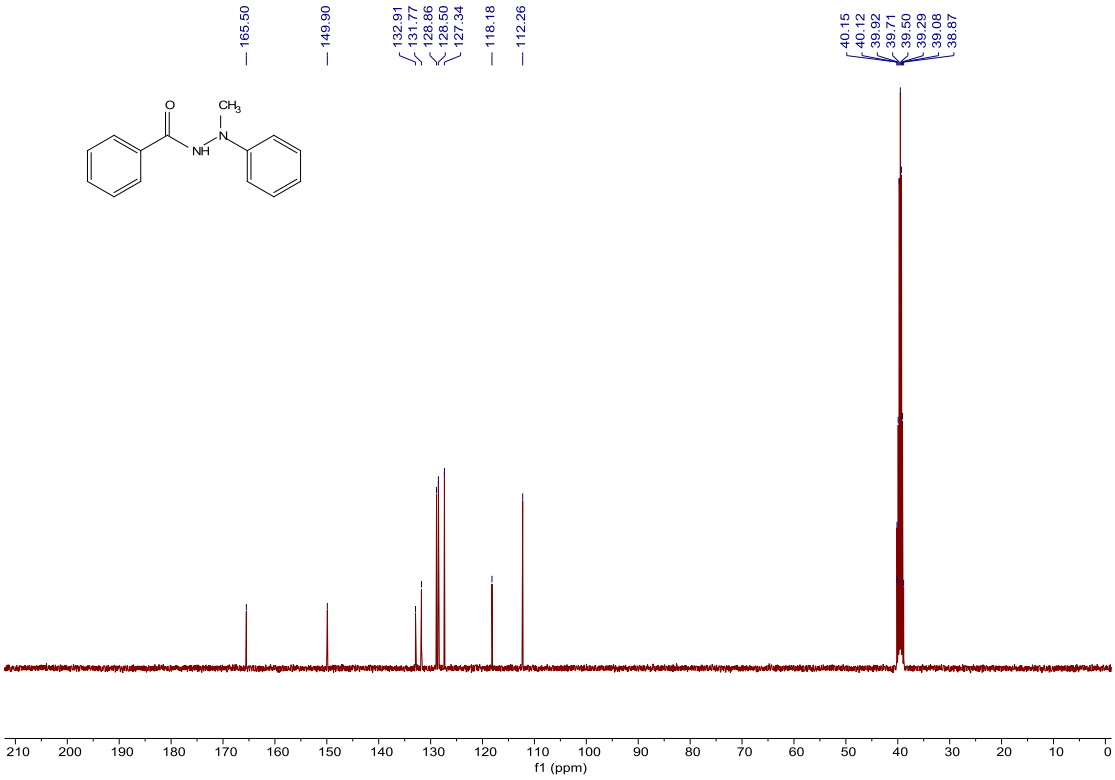
^{1}H NMR of 3 in DMSO-d_6 (400 MHz)



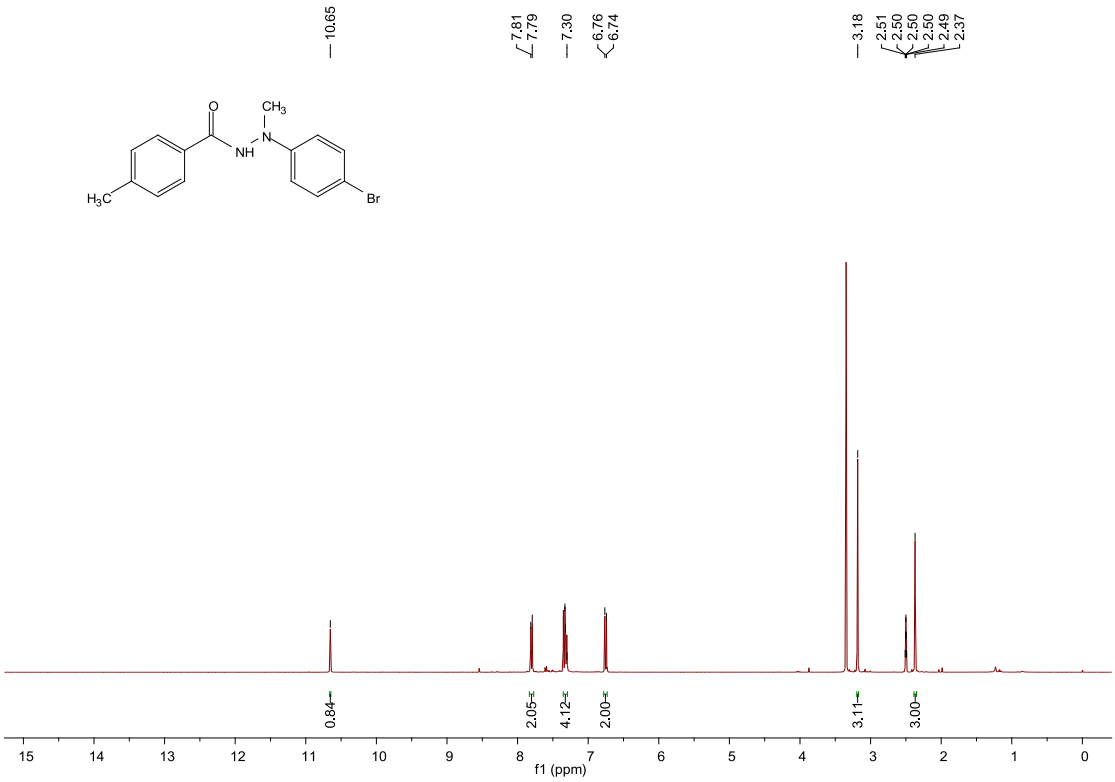
^{13}C{^{1}H} NMR of 3 in DMSO-d_6 (100 MHz)



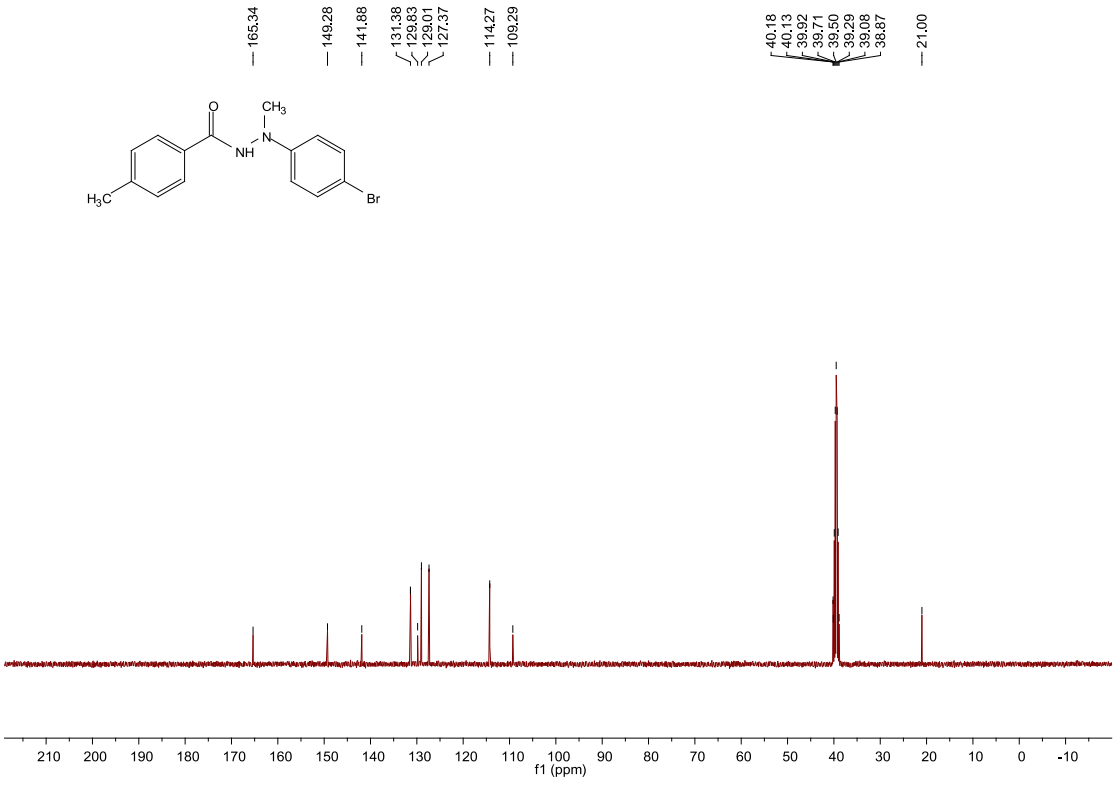
^{1}H NMR of 4 in DMSO-d_6 (400 MHz)



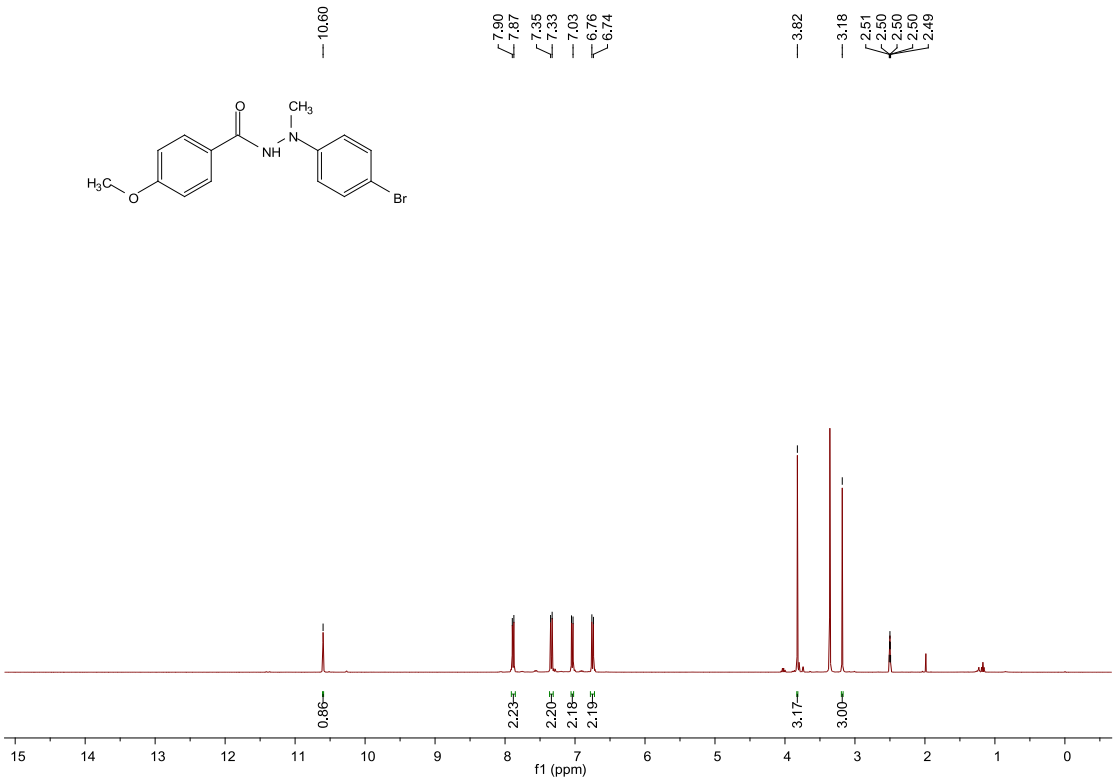
^{13}C{^{1}H} NMR of 4 in DMSO-d_6 (100 MHz)



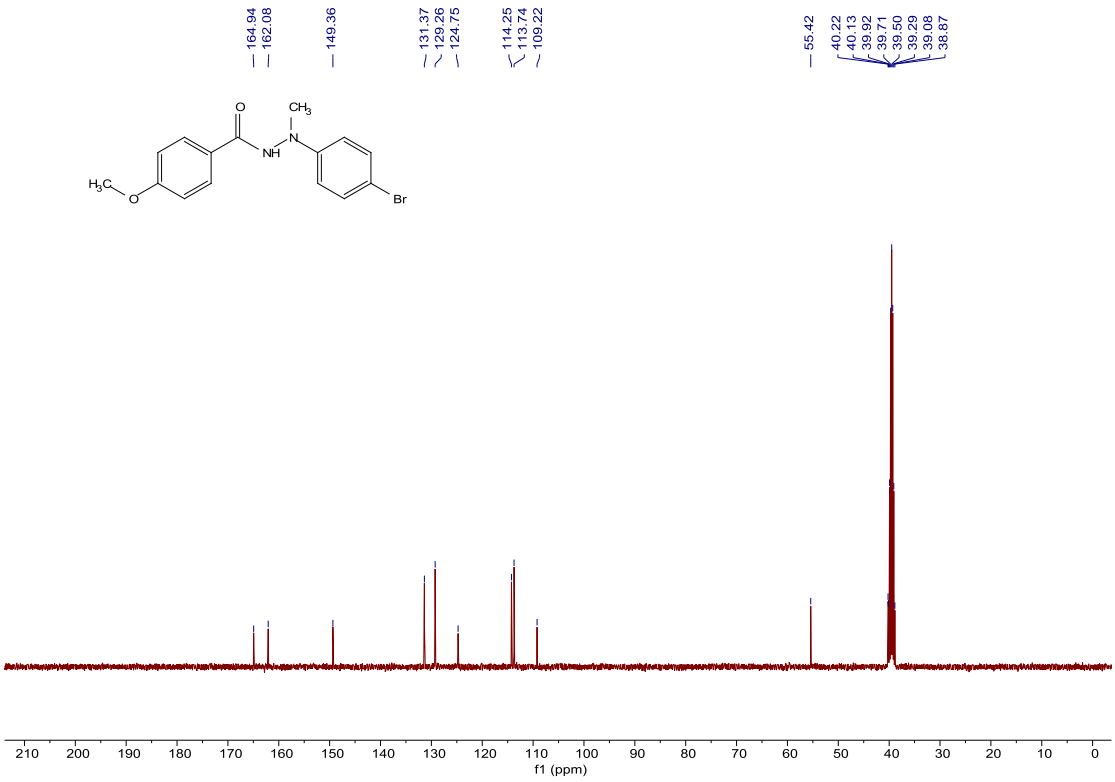
^{1}H NMR of 5 in DMSO-d_6 (400 MHz)



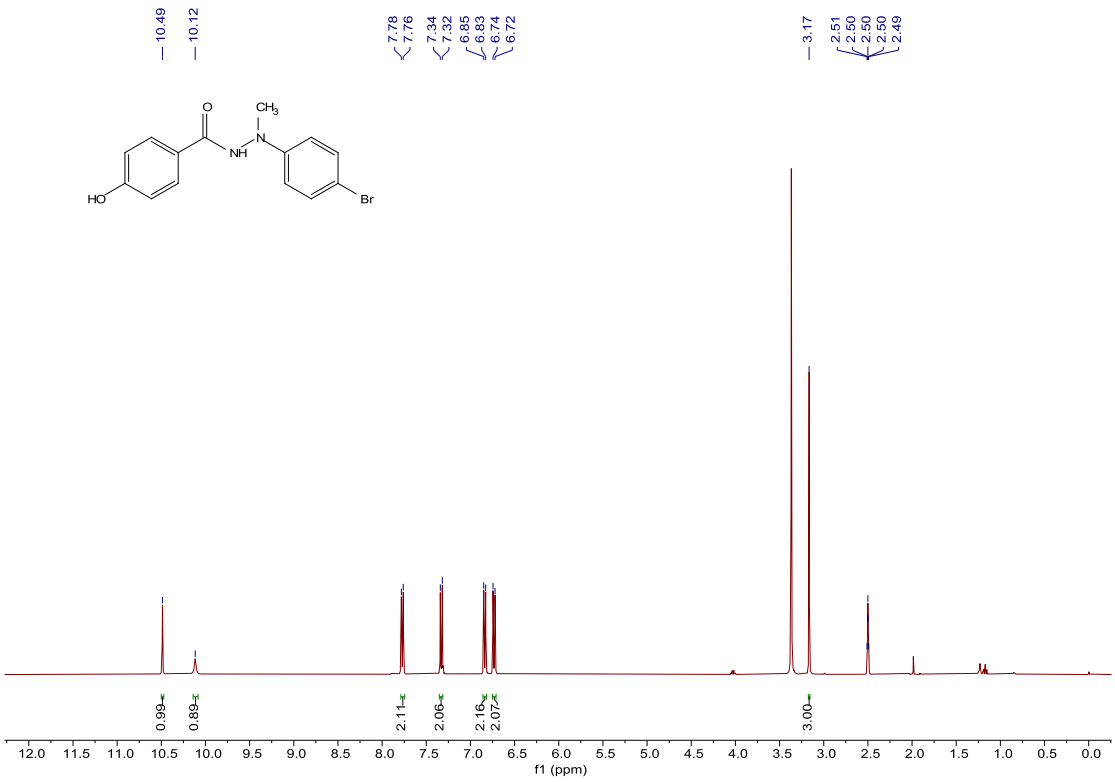
^{13}C{^{1}H} NMR of 5 in DMSO-d_6 (100 MHz)



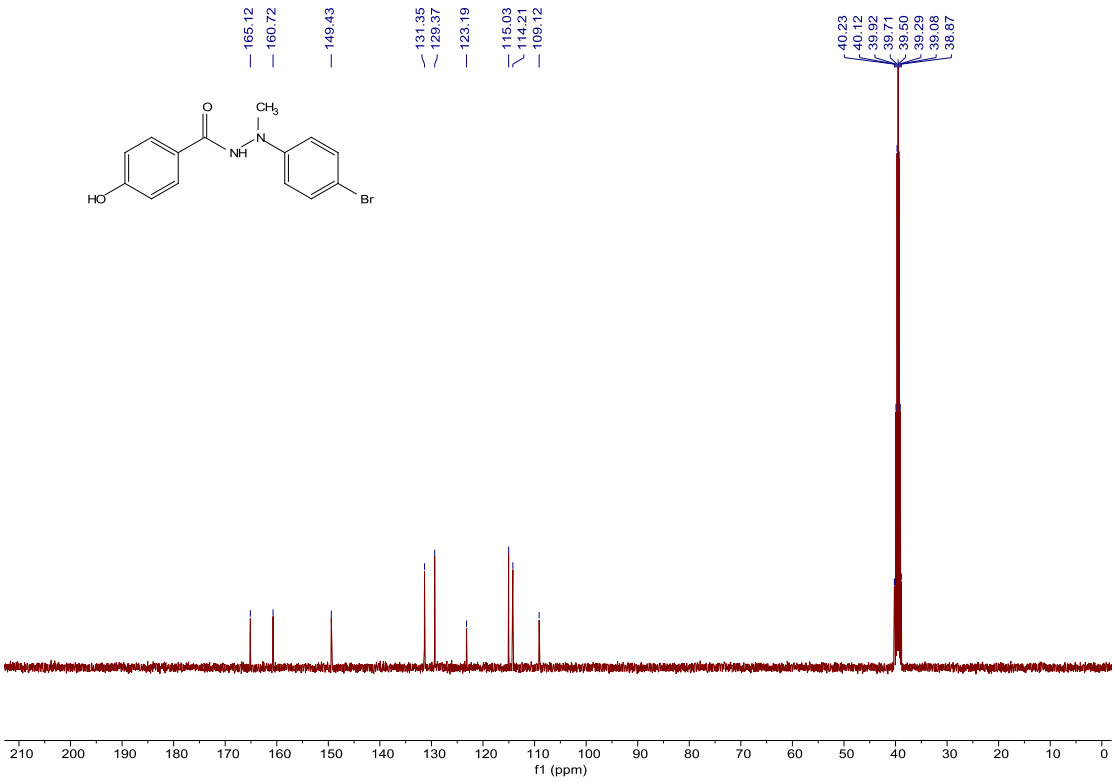
^{1}H NMR of 6 in DMSO-d_6 (400 MHz)



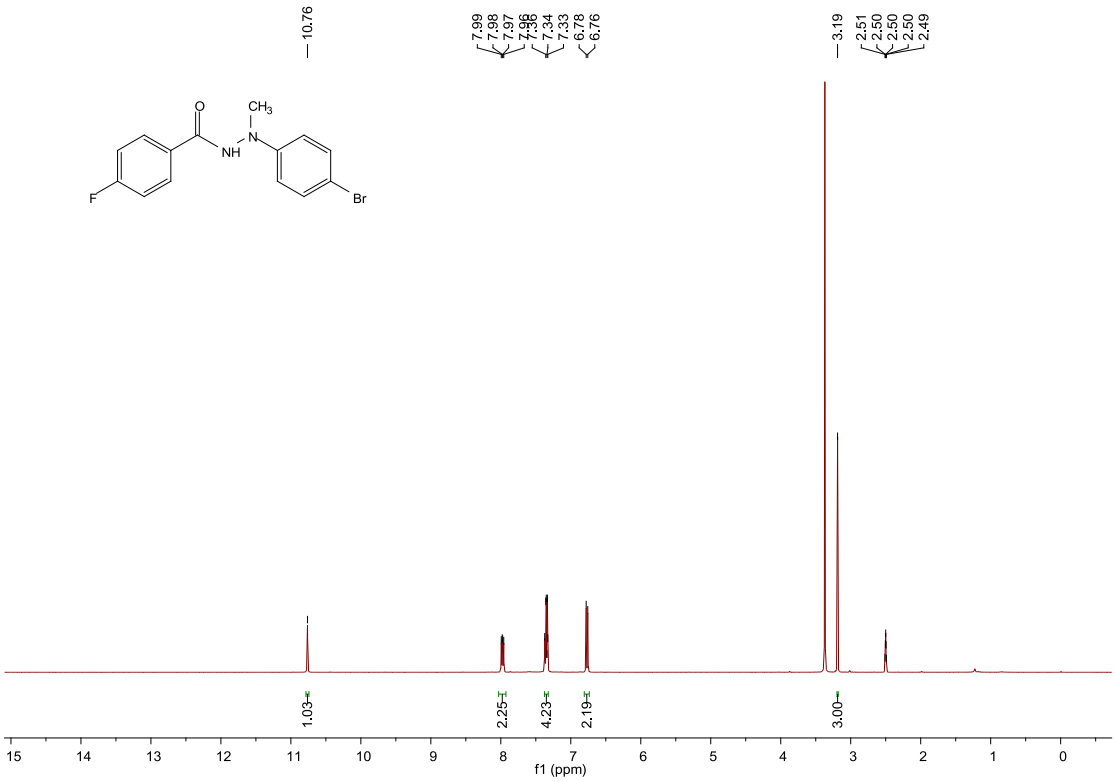
^{13}C{^{1}H} NMR of 6 in DMSO-d_6 (100 MHz)



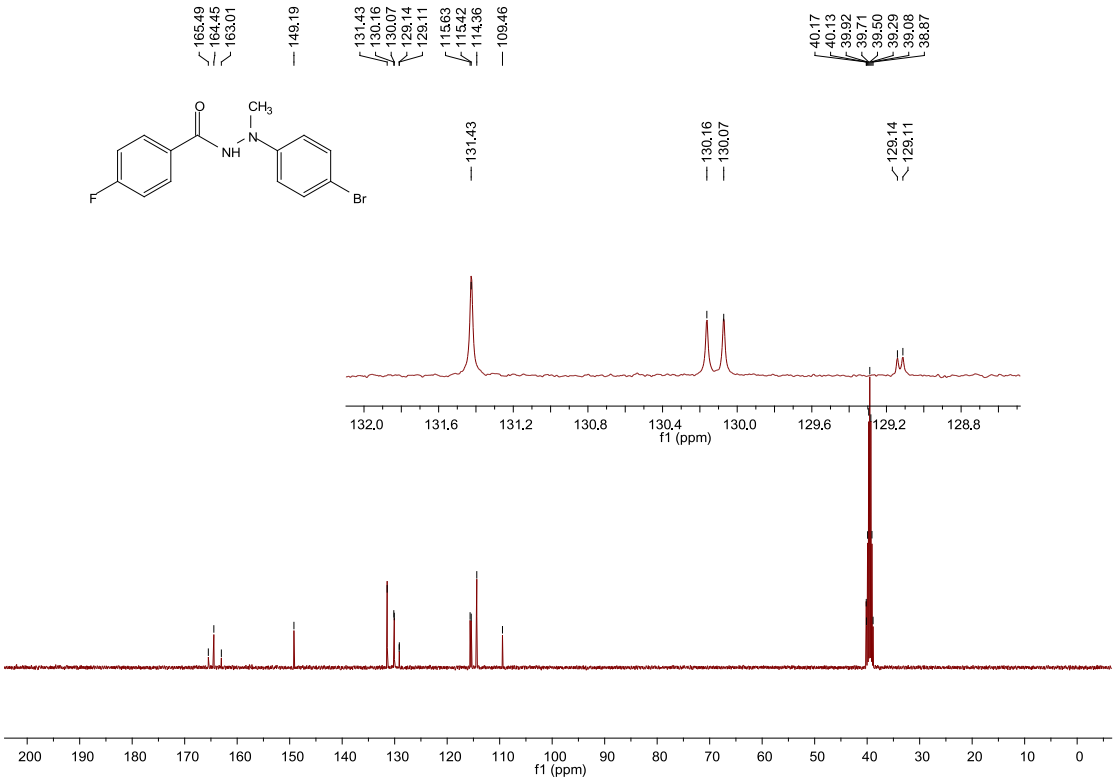
^{1}H NMR of 7 in DMSO-d_6 (400 MHz)



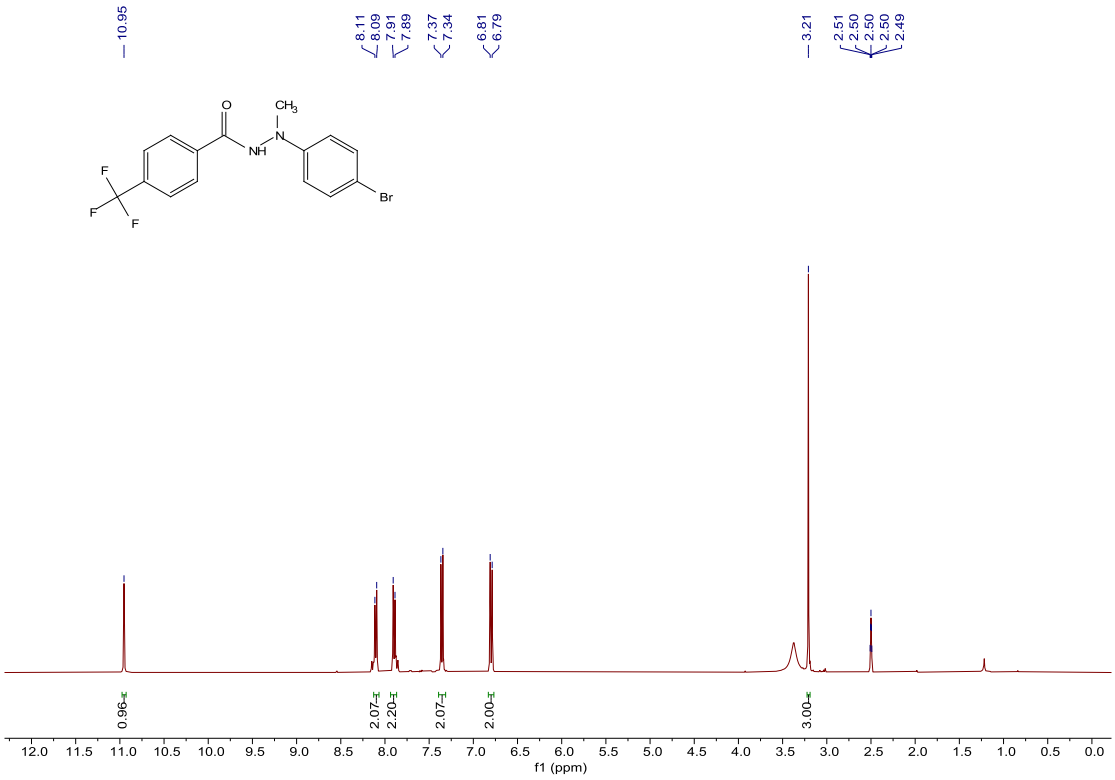
^{13}C{^{1}H} NMR of 7 in DMSO-d_6 (100 MHz)



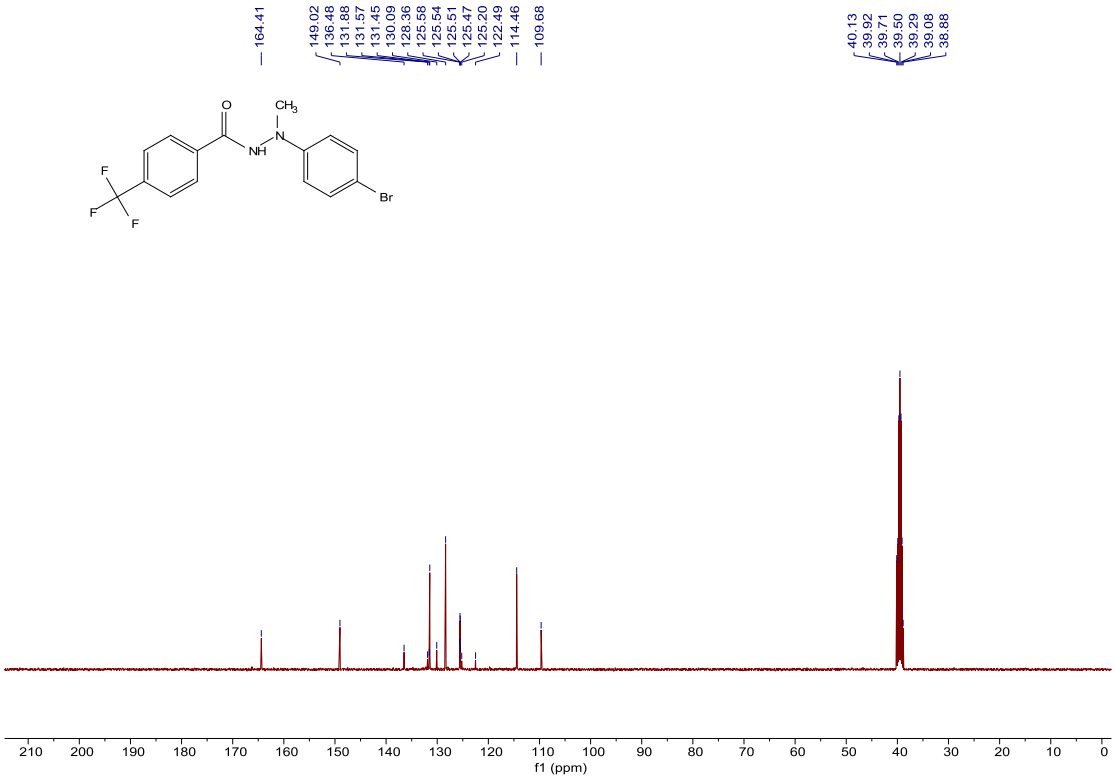
^{1}H NMR of 8 in DMSO-d_6 (400 MHz)



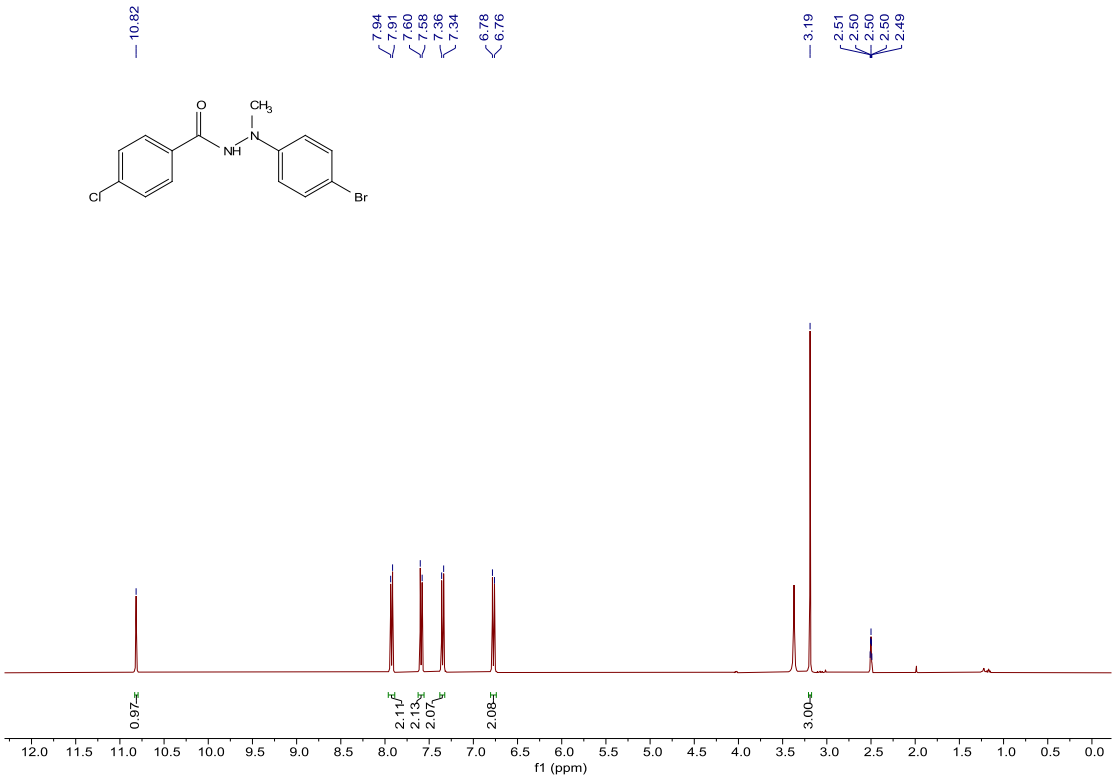
^{13}C{^{1}H} NMR of 8 in DMSO-d_6 (100 MHz)



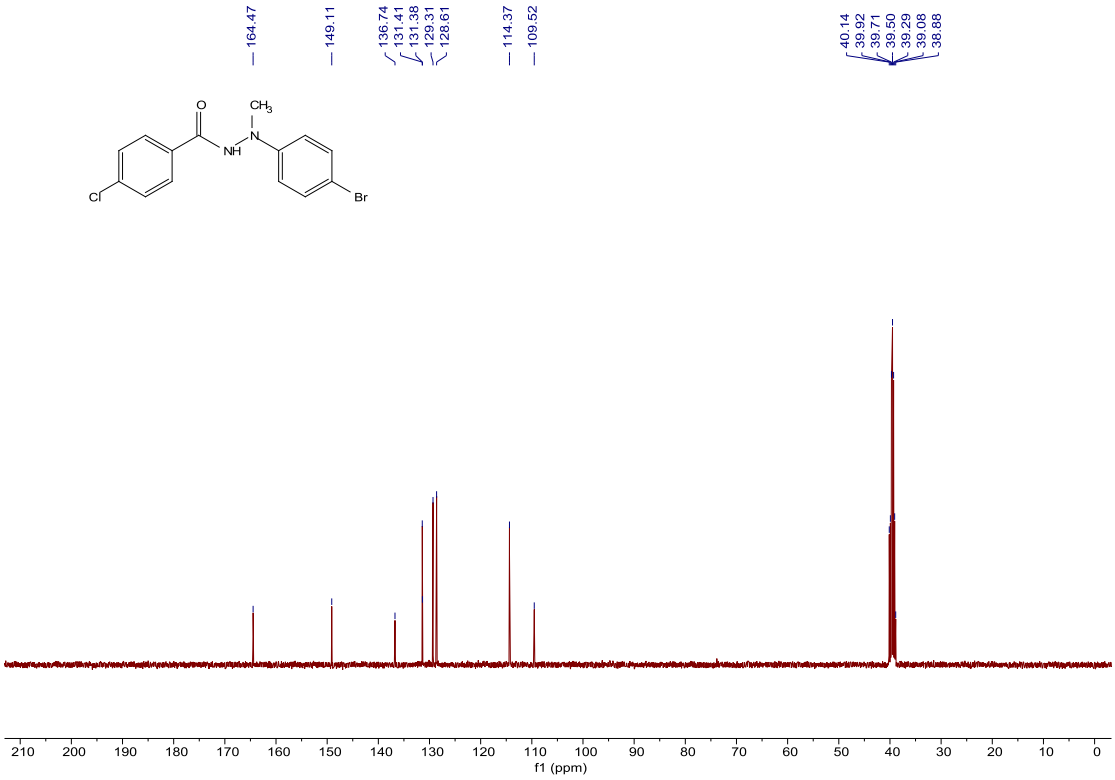
^{1}H NMR of 9 in DMSO-d_6 (400 MHz)



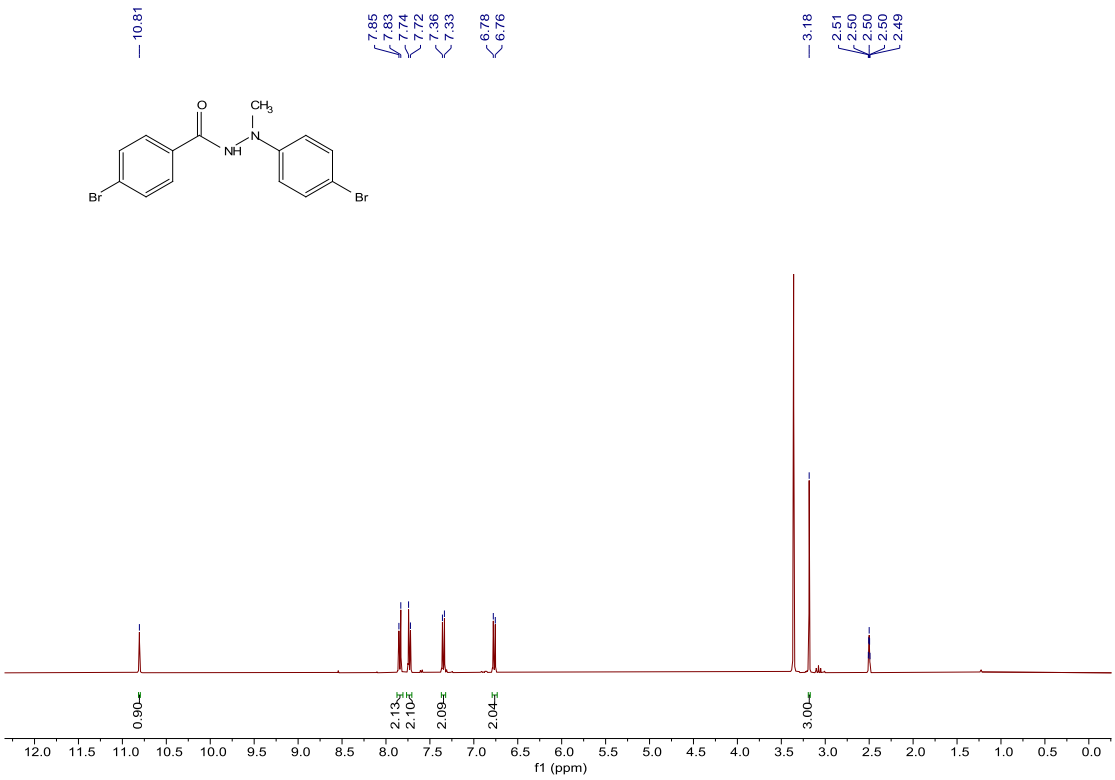
^{13}C{^{1}H} NMR of 9 in DMSO-d_6 (100 MHz)



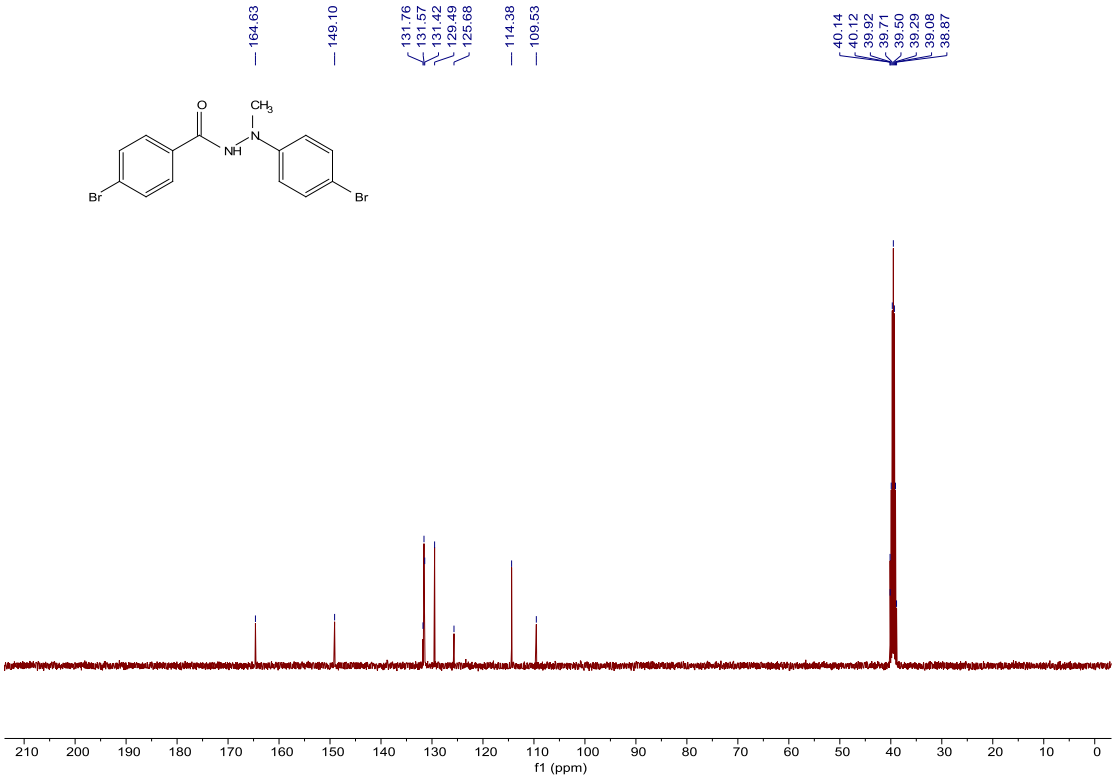
^{1}H NMR of 10 in DMSO-d_6 (400 MHz)



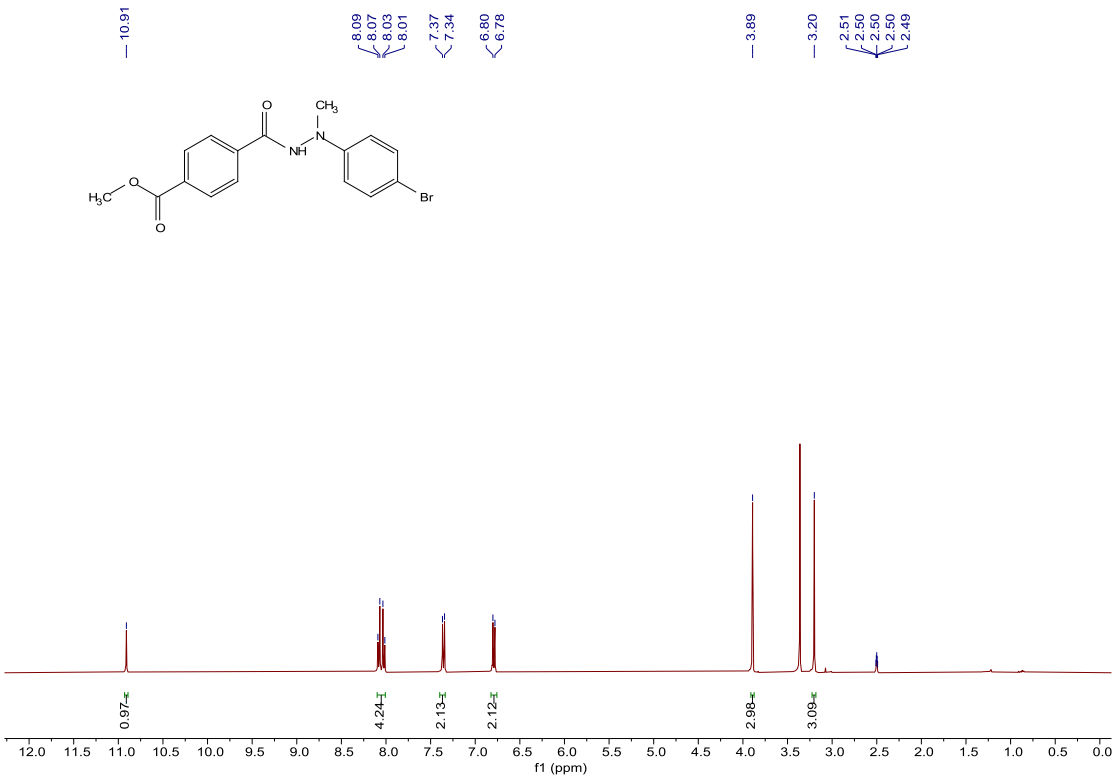
^{13}C{^{1}H} NMR of 10 in DMSO-d_6 (100 MHz)



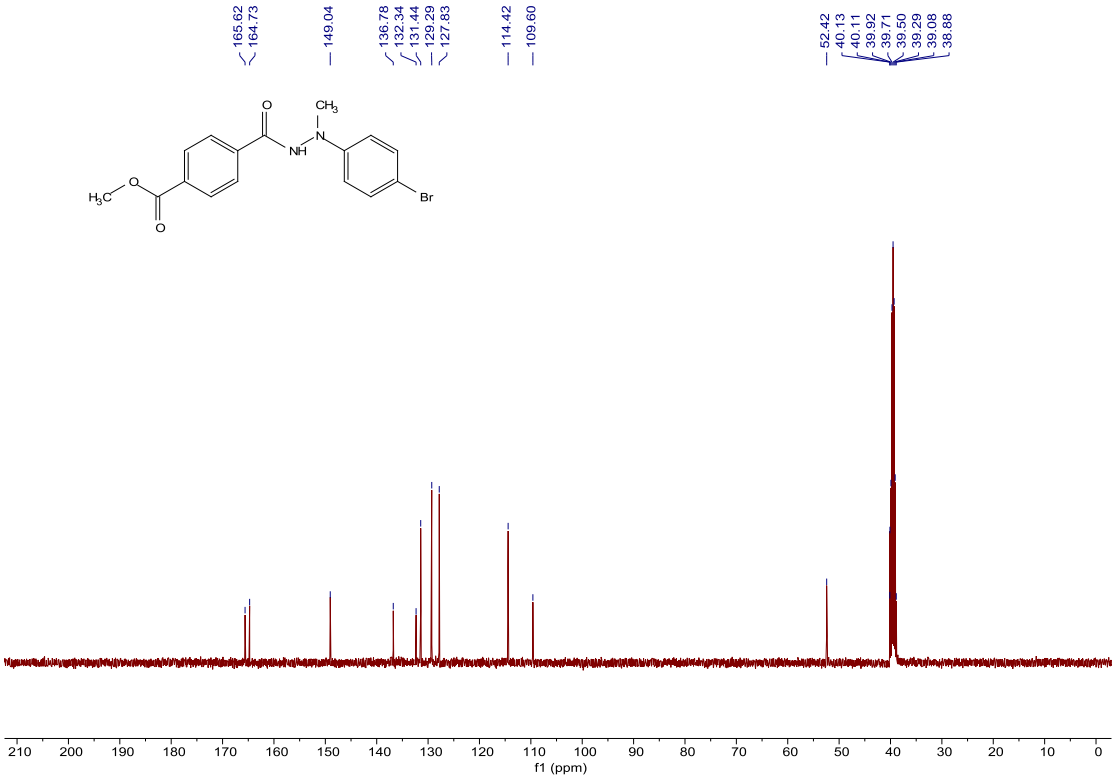
^{1}H NMR of 11 in DMSO-d_6 (400 MHz)



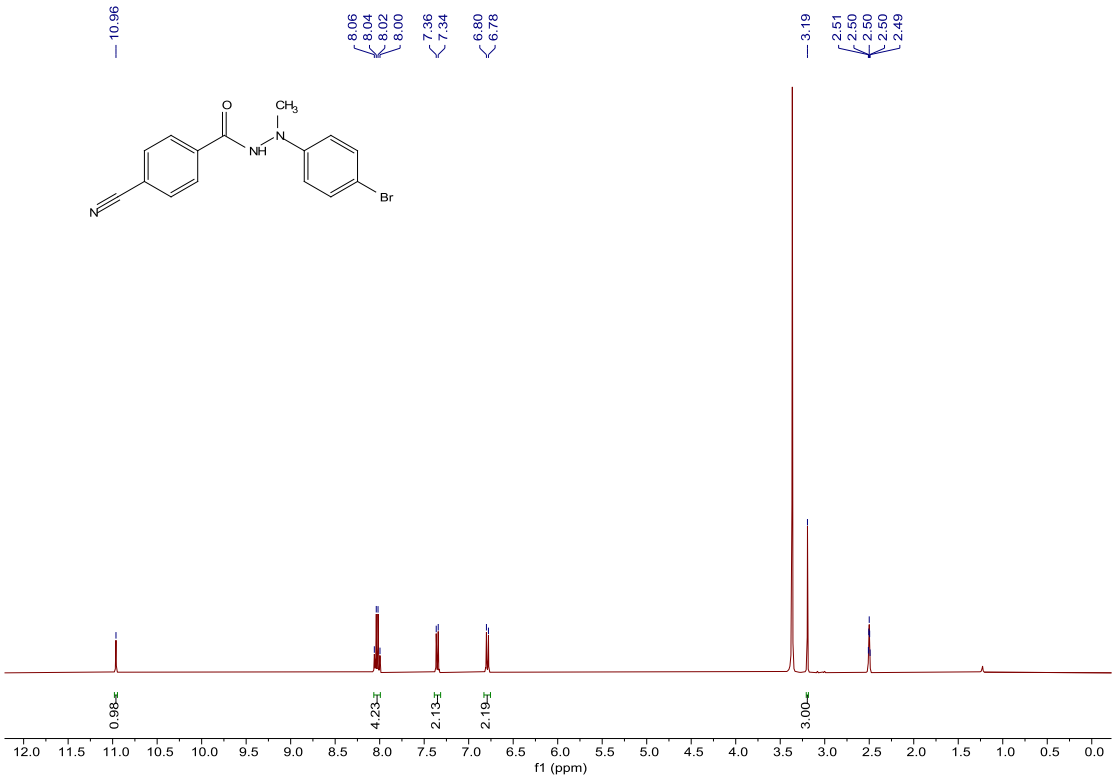
^{13}C{^{1}H} NMR of 11 in DMSO-d_6 (100 MHz)



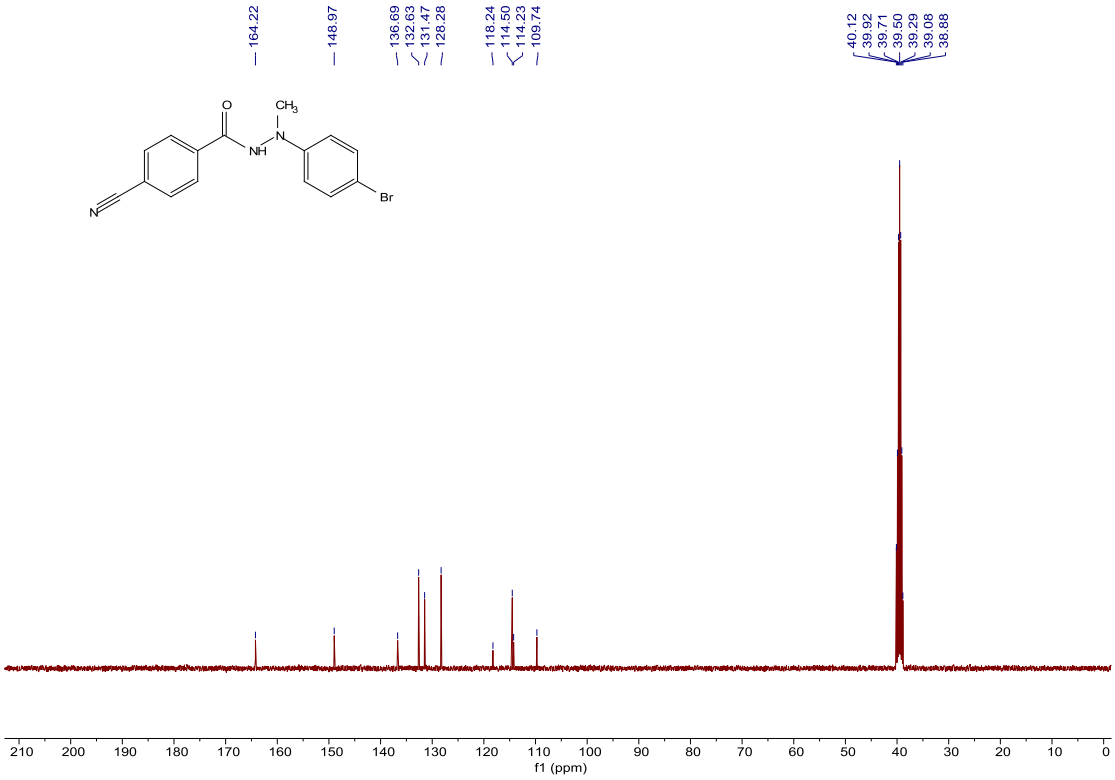
^{1}H NMR of 12 in DMSO-d_6 (400 MHz)



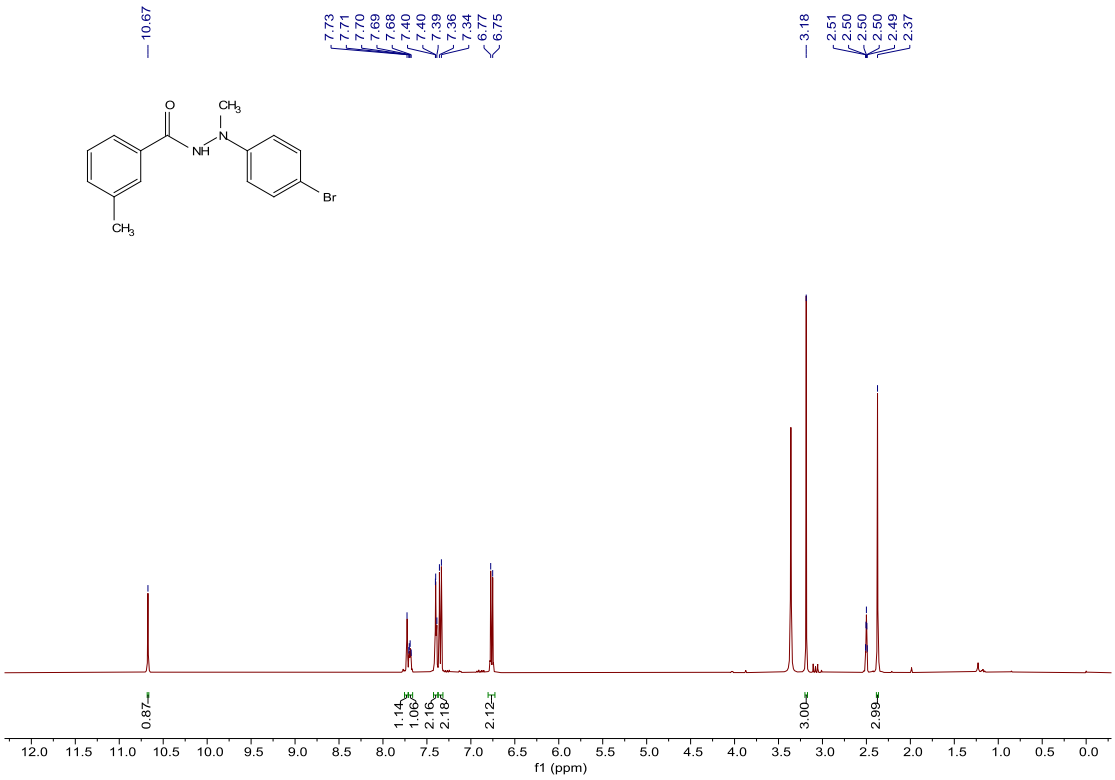
^{13}C{^{1}H} NMR of 12 in DMSO-d_6 (100 MHz)



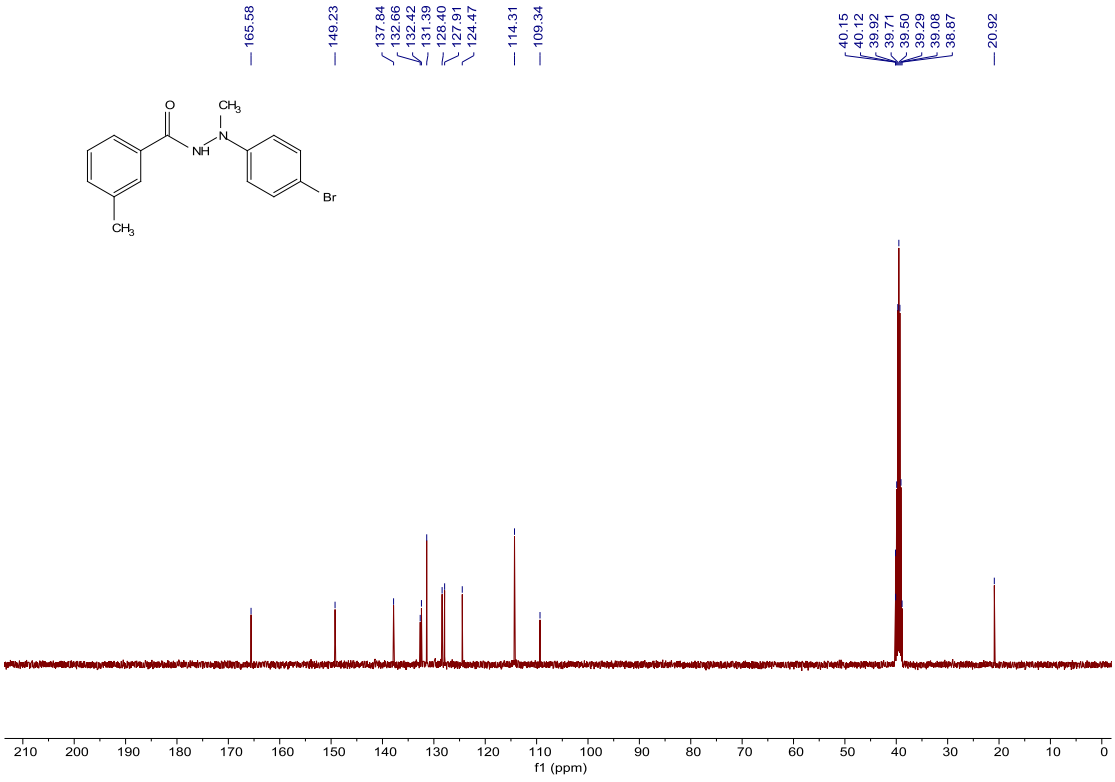
^{1}H NMR of 13 in DMSO-d_6 (400 MHz)



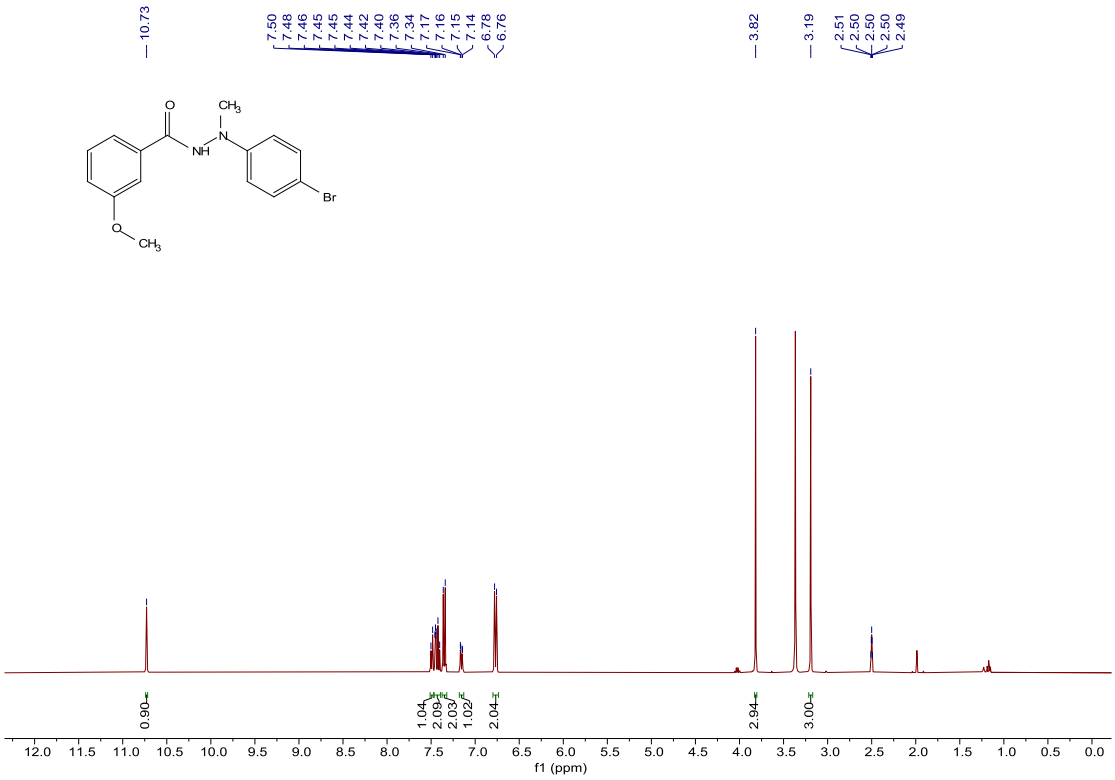
^{13}C{^{1}H} NMR of 13 in DMSO-d_6 (100 MHz)



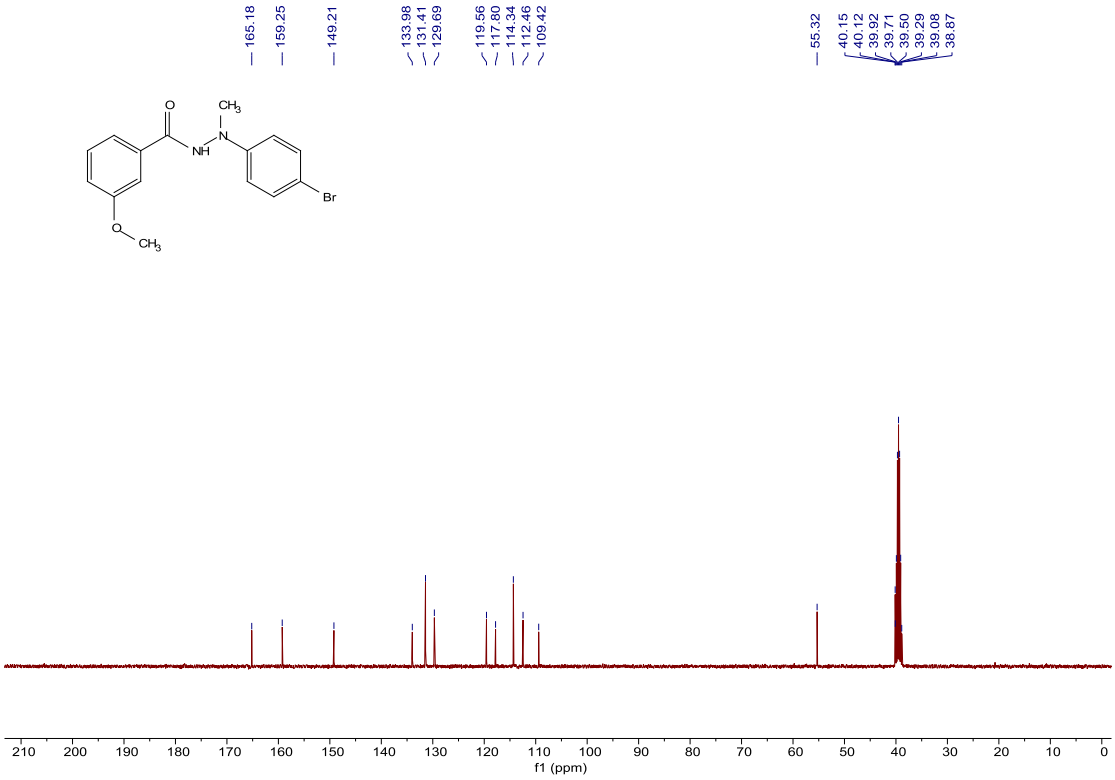
^{1}H NMR of 14 in DMSO-d_6 (400 MHz)



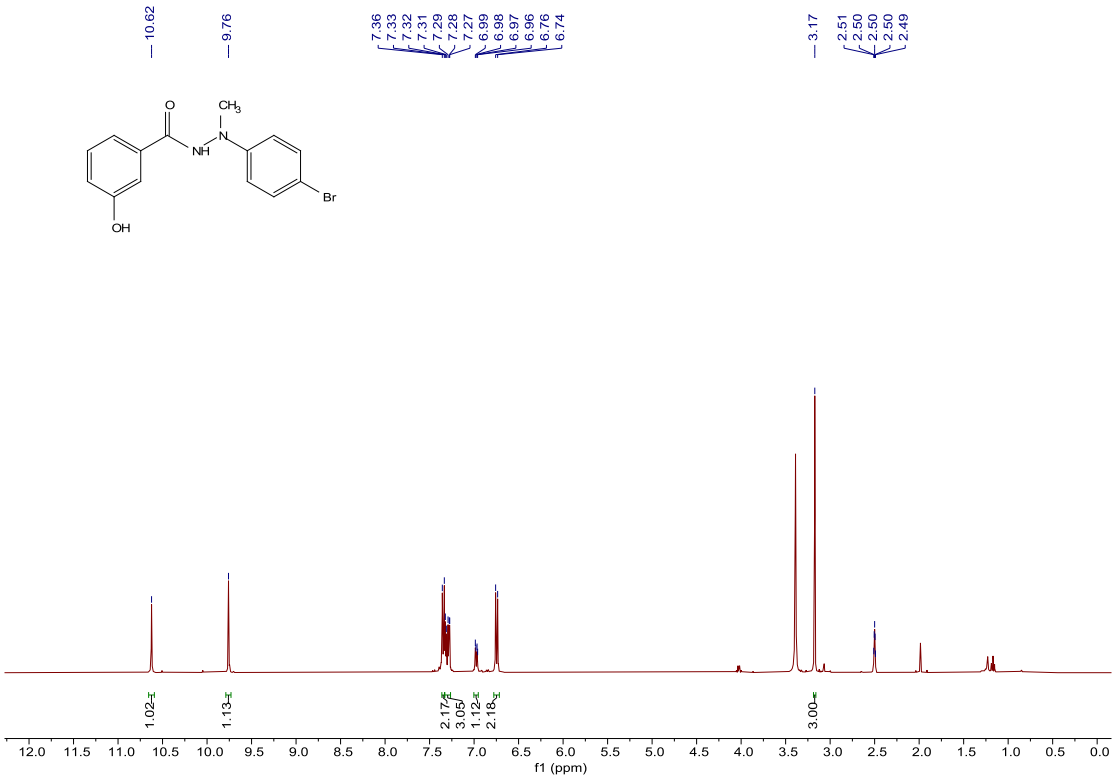
^{13}C{^{1}H} NMR of 14 in DMSO-d_6 (100 MHz)



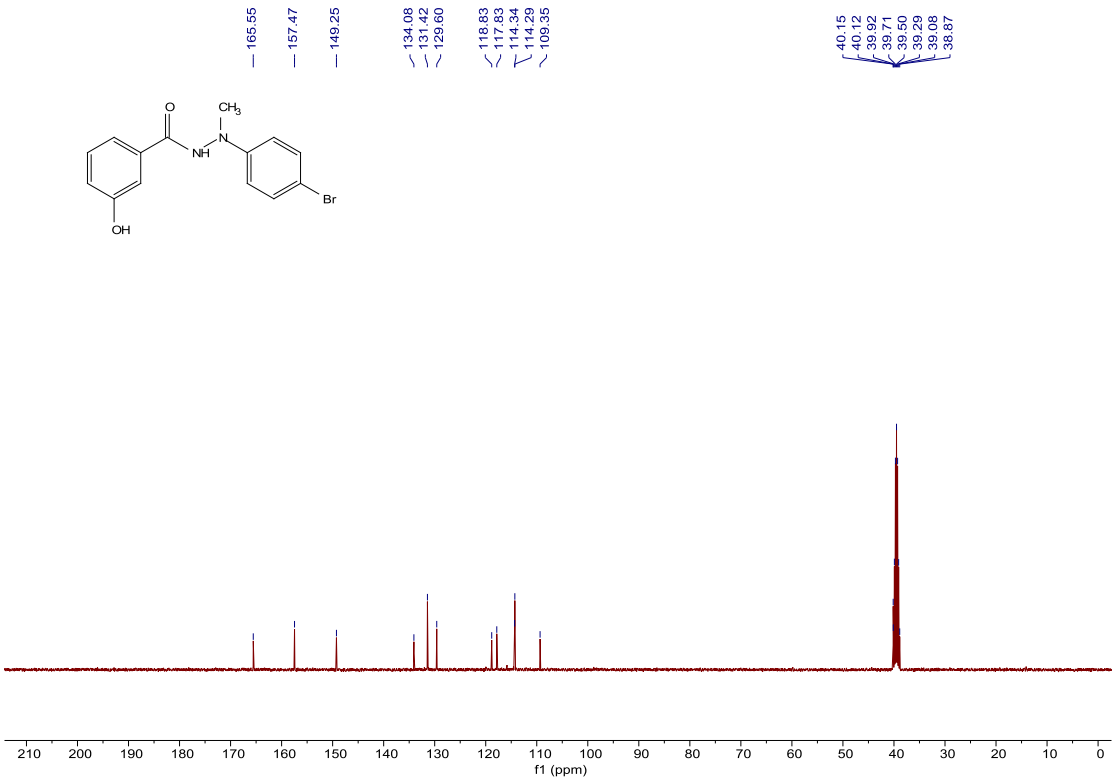
^{1}H NMR of 15 in DMSO-d_6 (400 MHz)



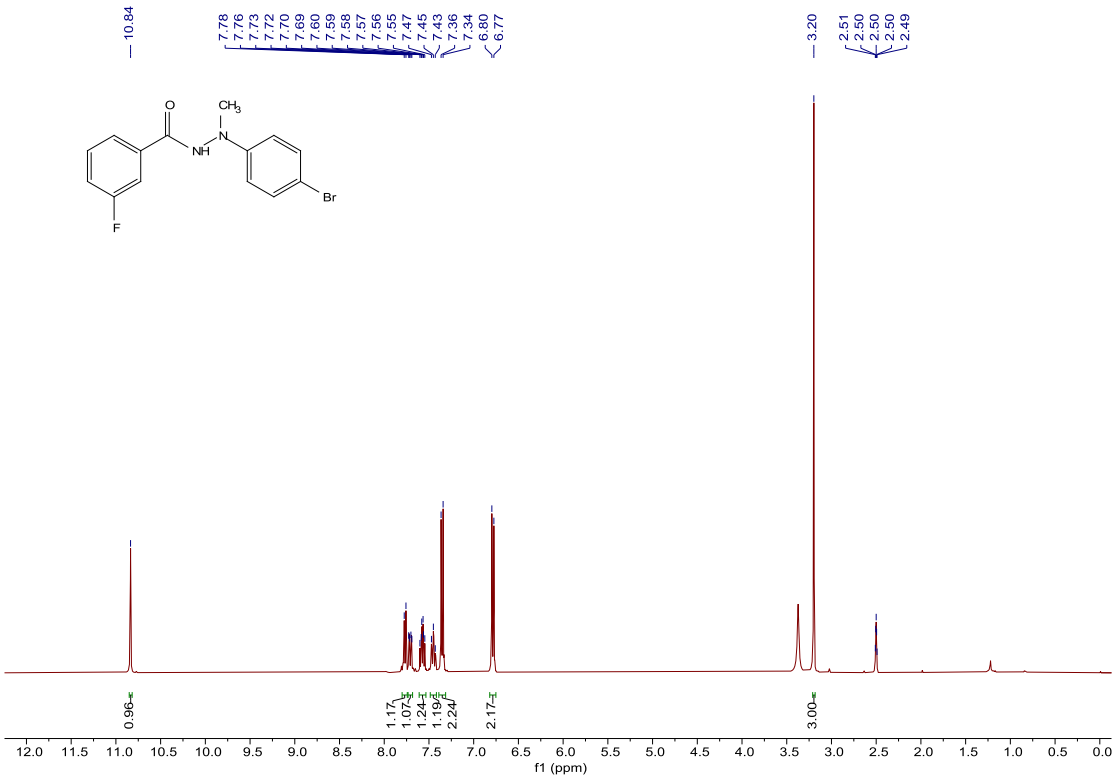
^{13}C{^{1}H} NMR of 15 in DMSO-d_6 (100 MHz)



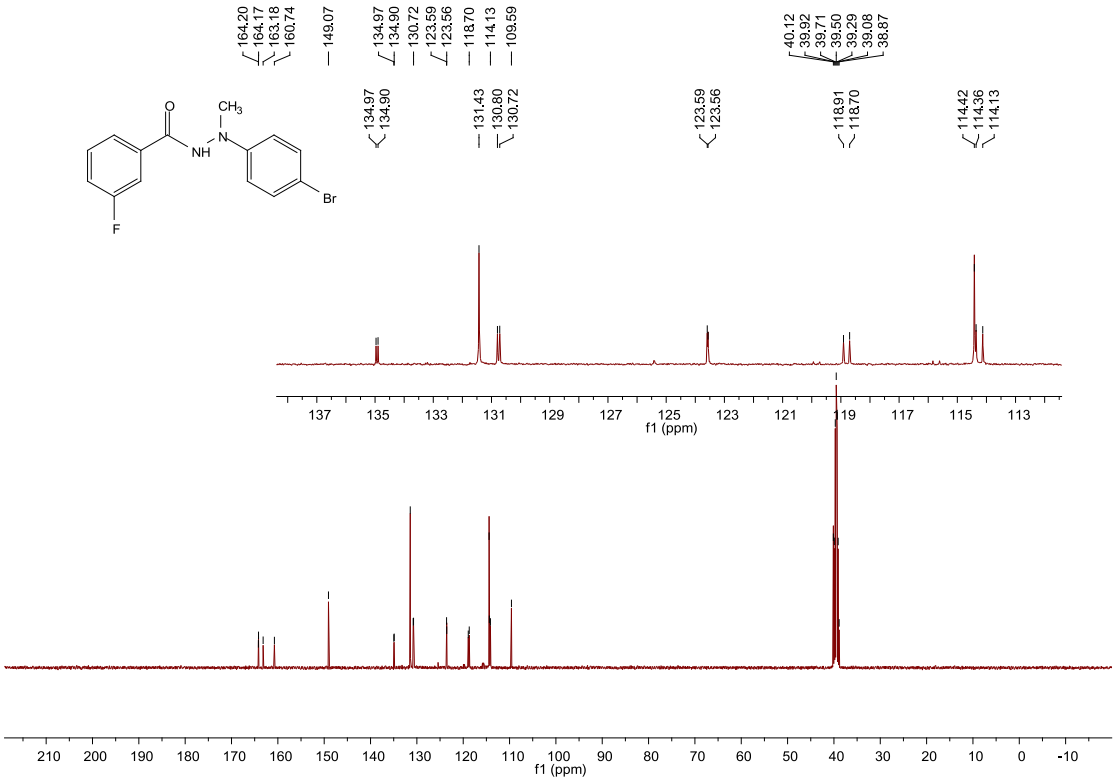
^{1}H NMR of 16 in DMSO-d_6 (400 MHz)



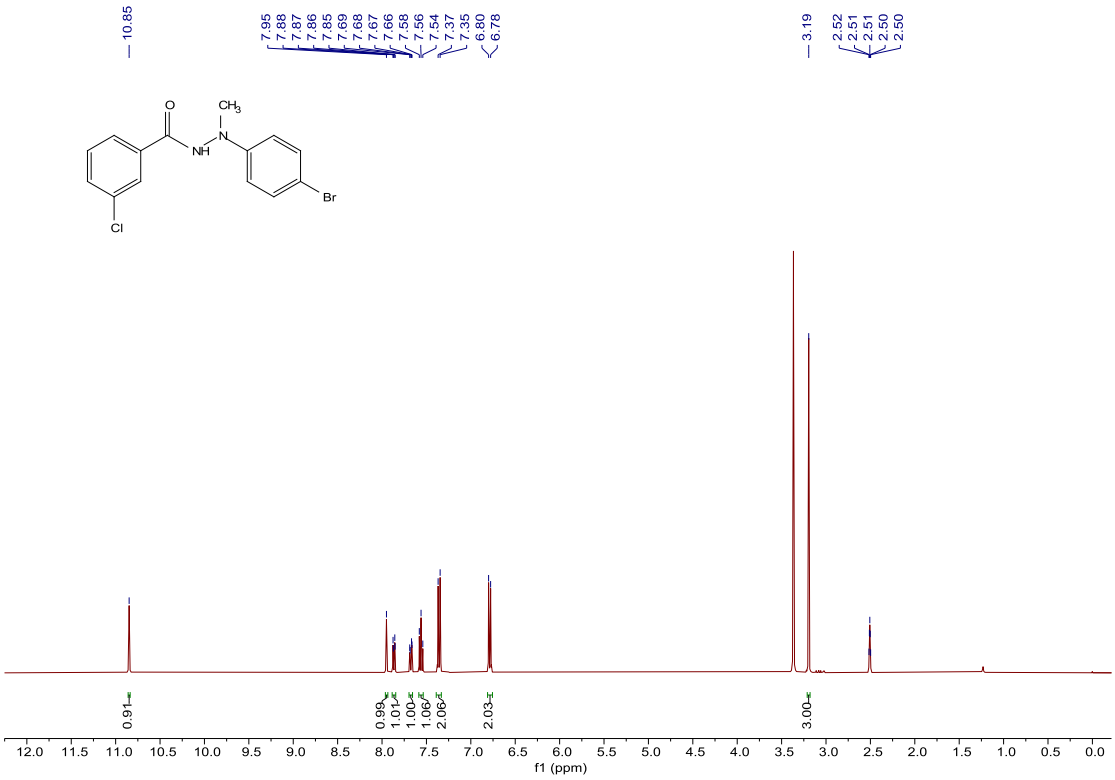
^{13}C{^{1}H} NMR of 16 in DMSO-d_6 (100 MHz)



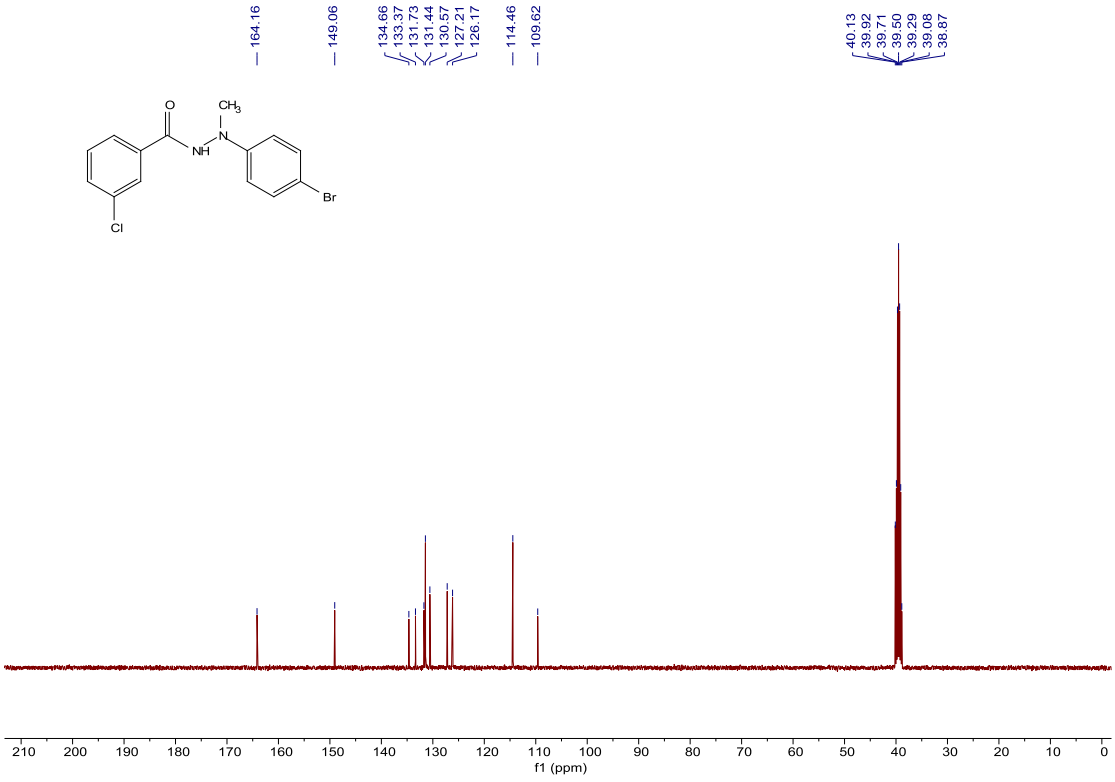
^{1}H NMR of 17 in DMSO-d_6 (400 MHz)



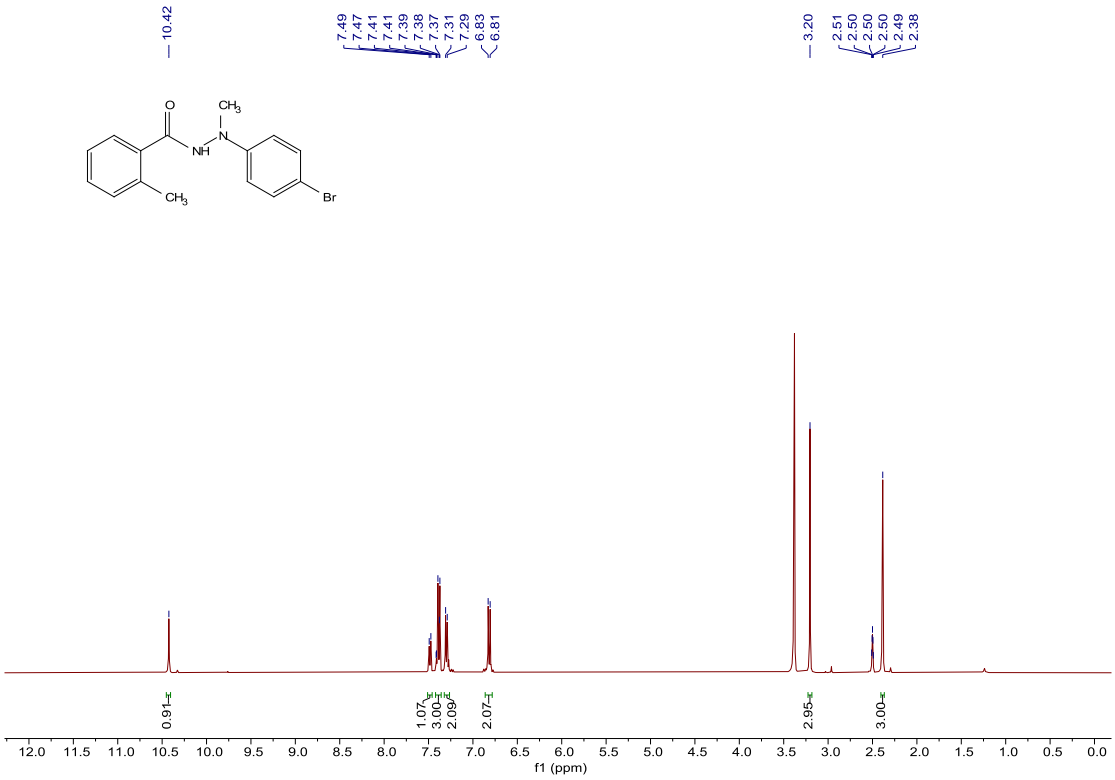
^{13}C{^{1}H} NMR of 17 in DMSO-d_6 (100 MHz)



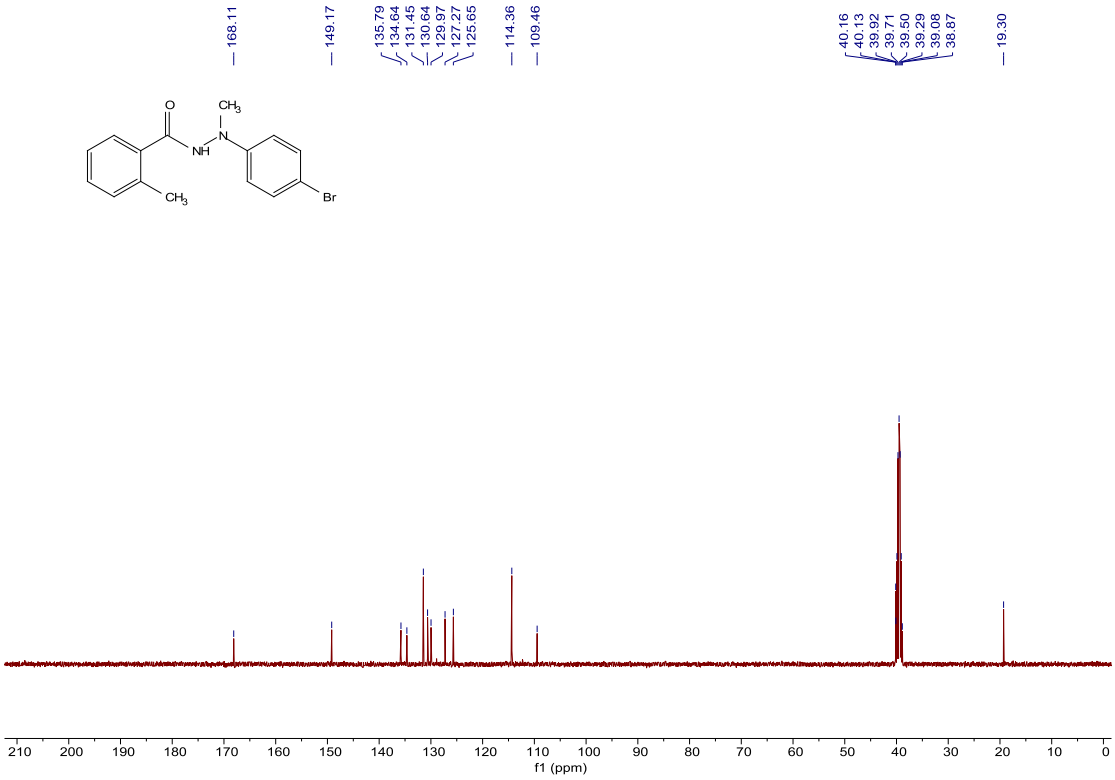
^{1}H NMR of 18 in DMSO-d_6 (400 MHz)



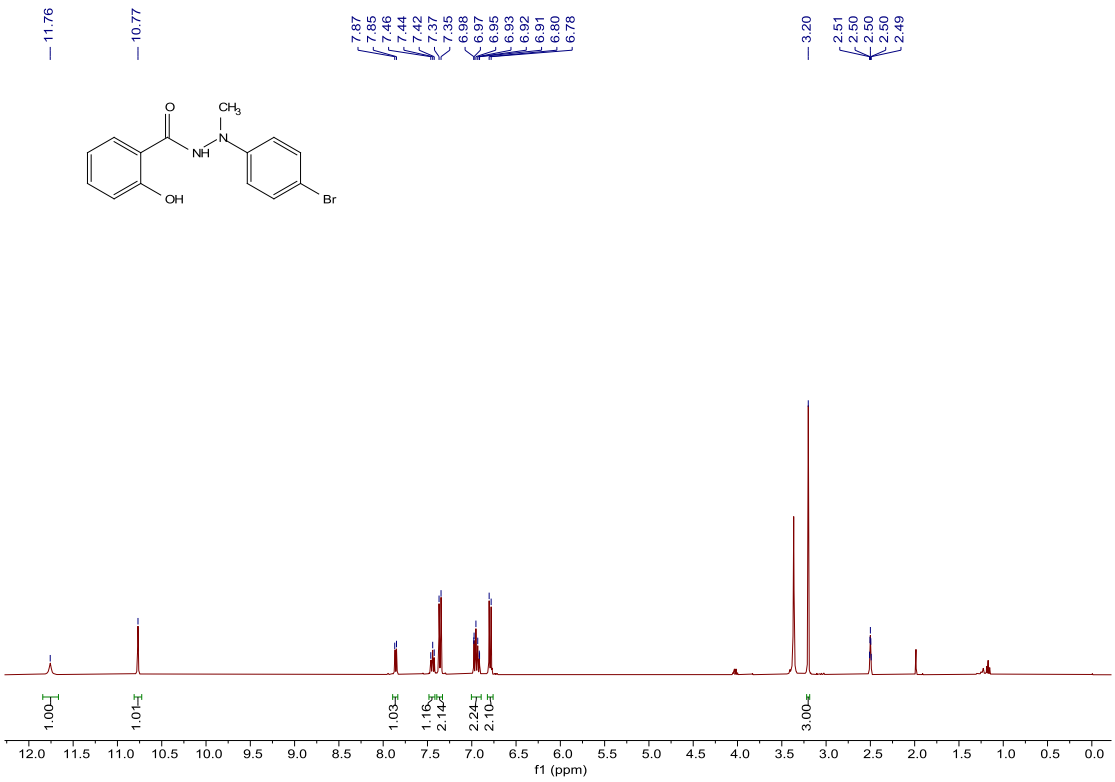
^{13}C{^{1}H} NMR of 18 in DMSO-d_6 (100 MHz)



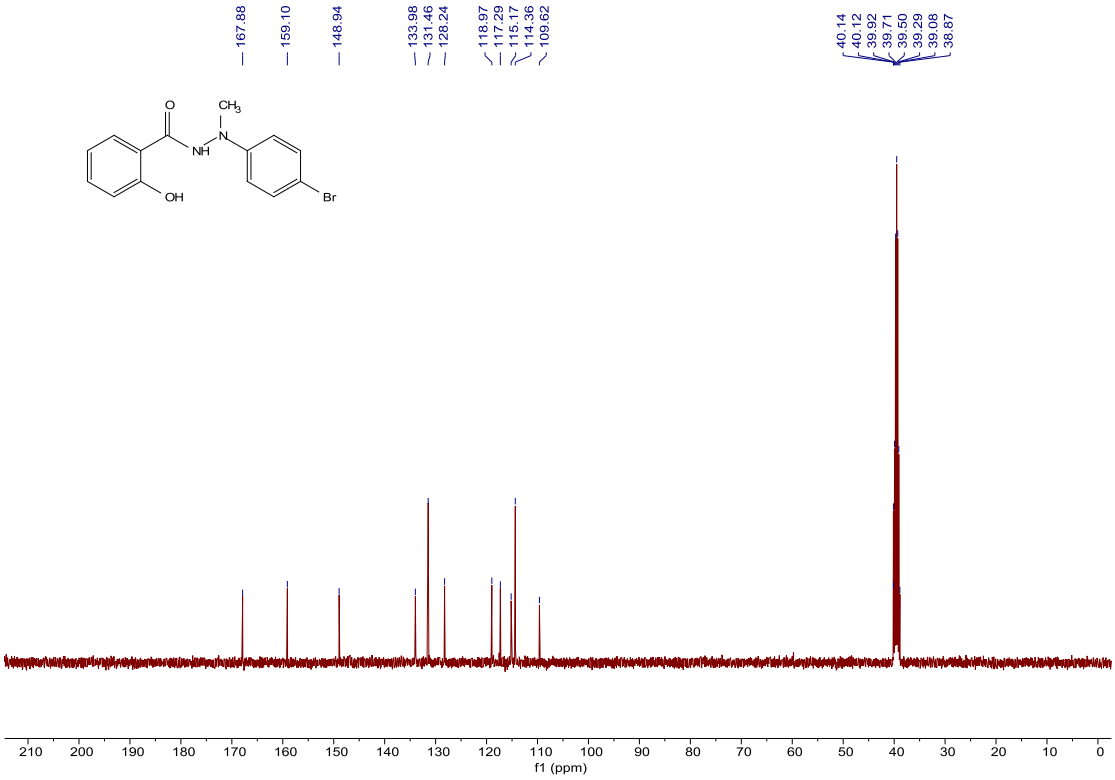
^{1}H NMR of 19 in DMSO-d_6 (400 MHz)



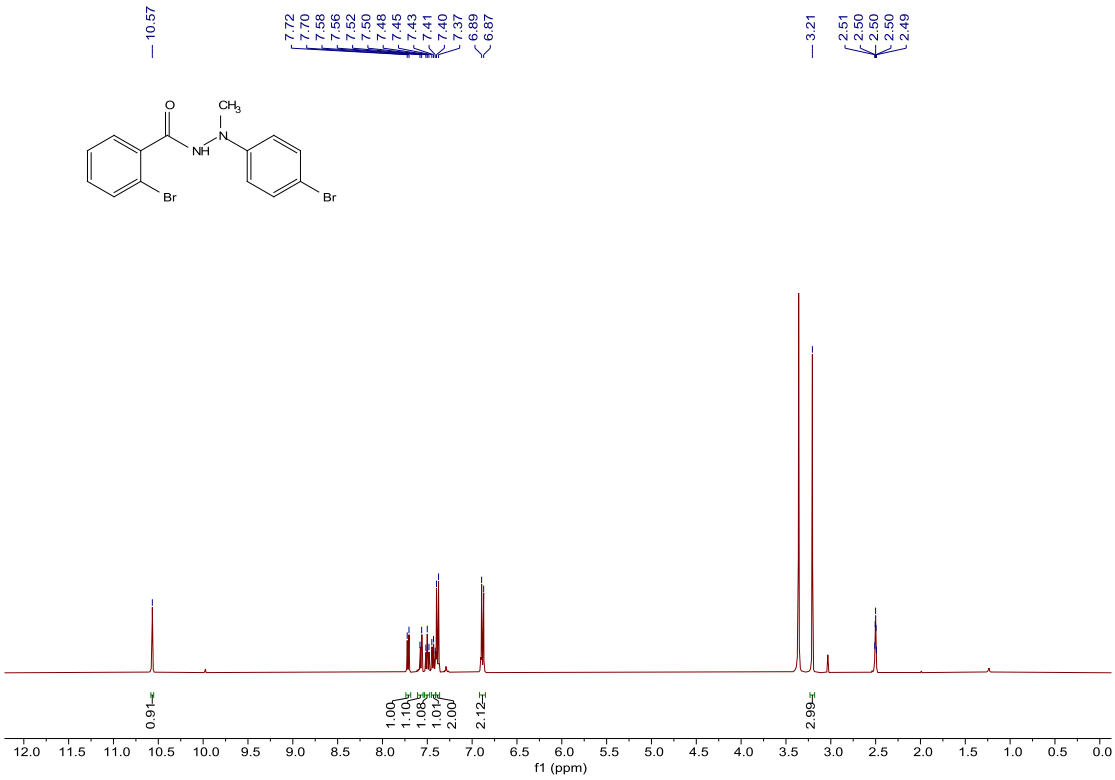
^{13}C{^{1}H} NMR of 19 in DMSO-d_6 (100 MHz)



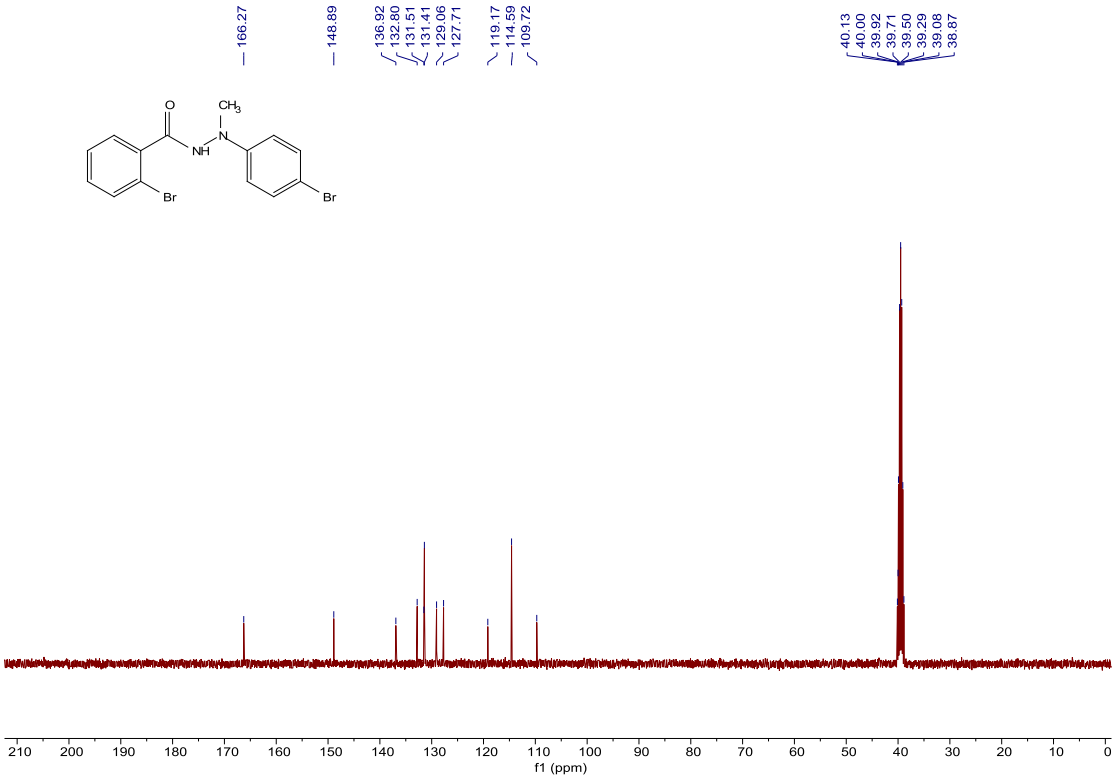
^{1}H NMR of 20 in DMSO-d_6 (400 MHz)



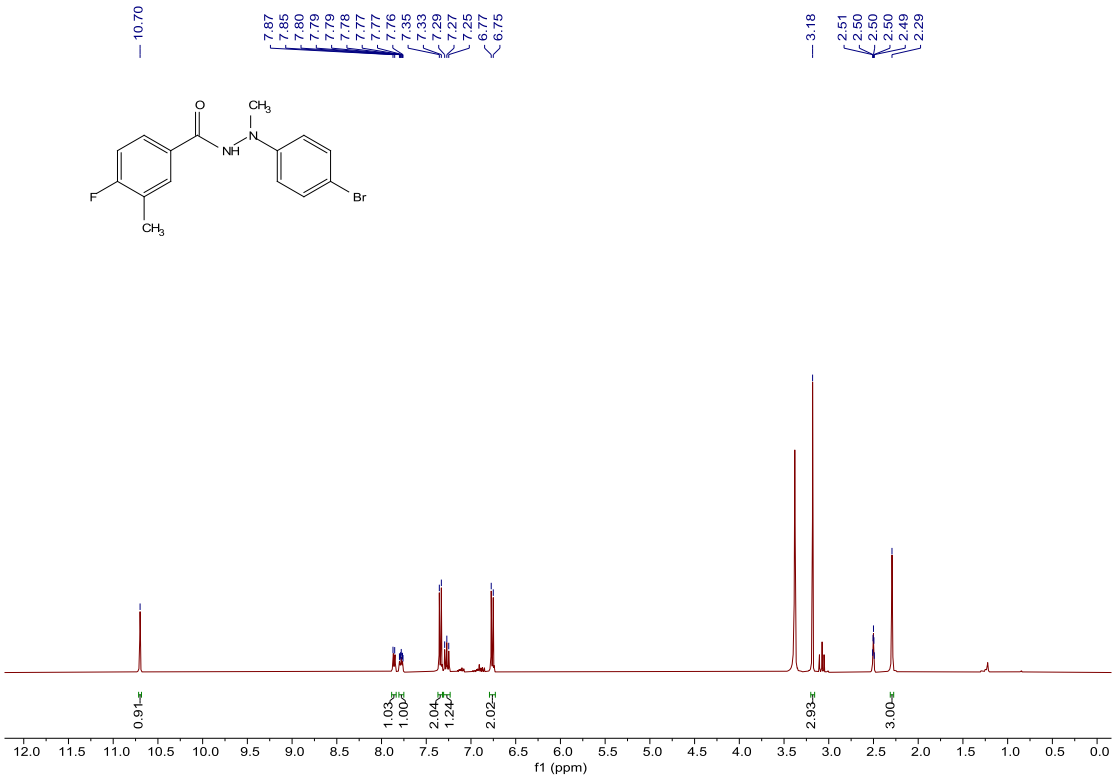
^{13}C{^{1}H} NMR of 20 in DMSO-d_6 (100 MHz)



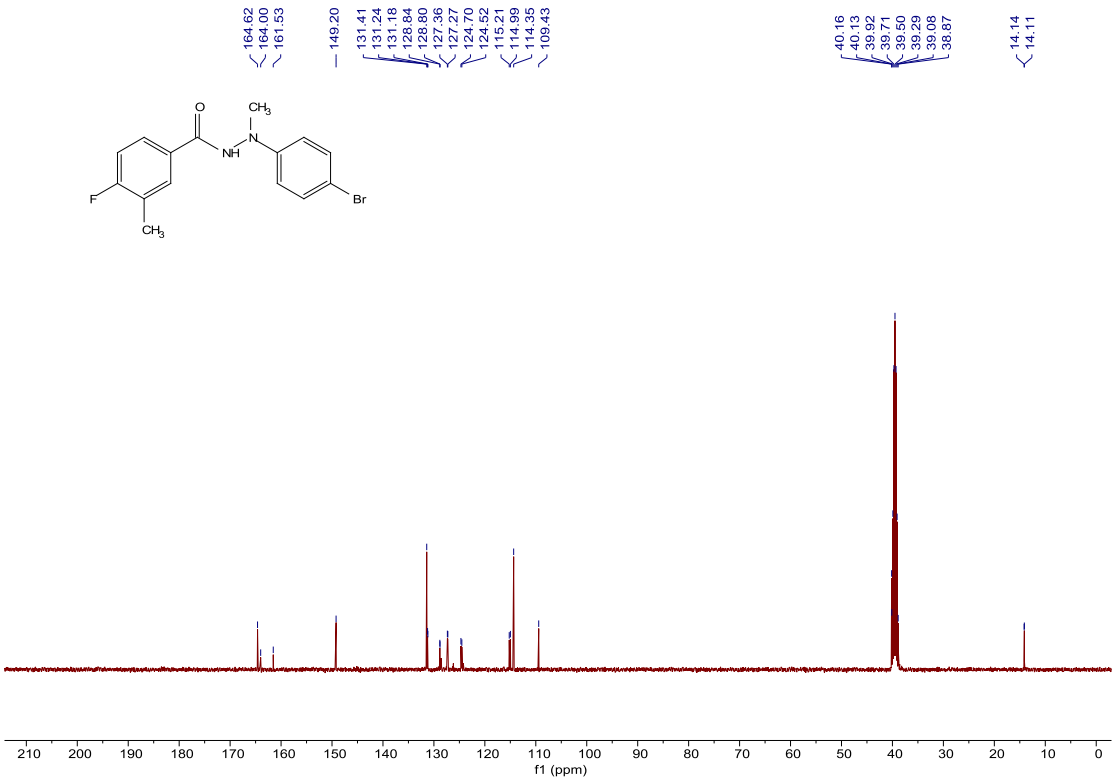
^{1}H NMR of 21 in DMSO-d_6 (400 MHz)



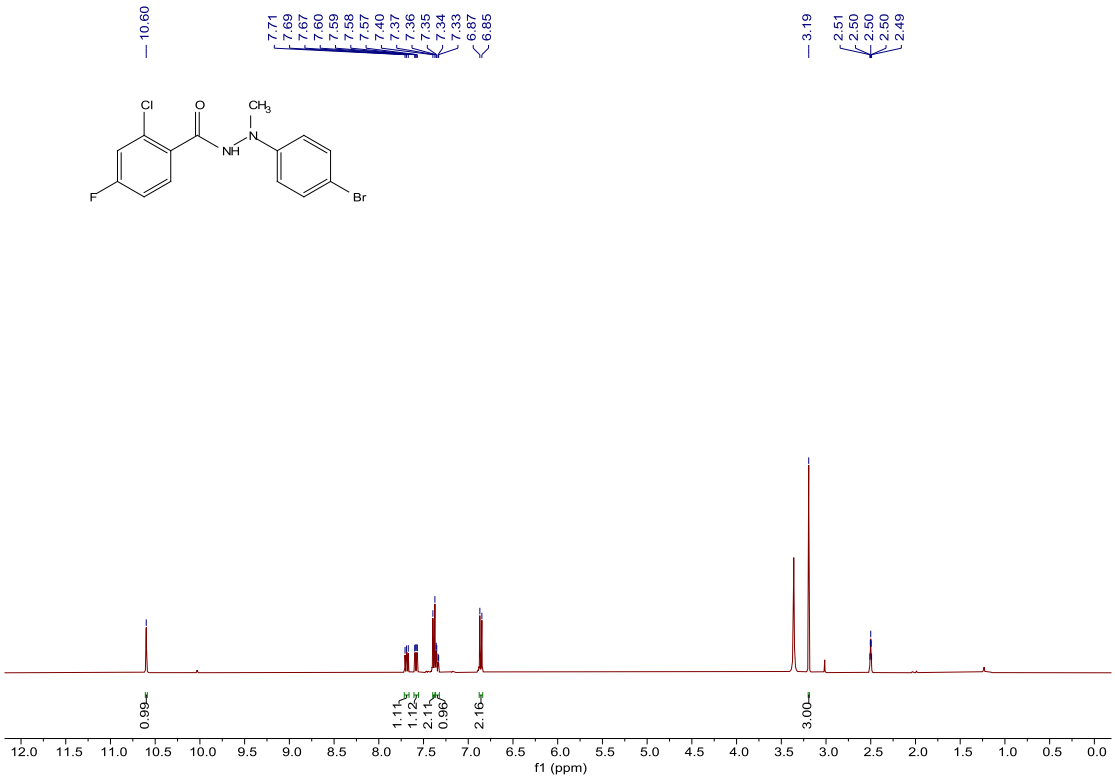
^{13}C{^{1}H} NMR of 21 in DMSO-d_6 (100 MHz)



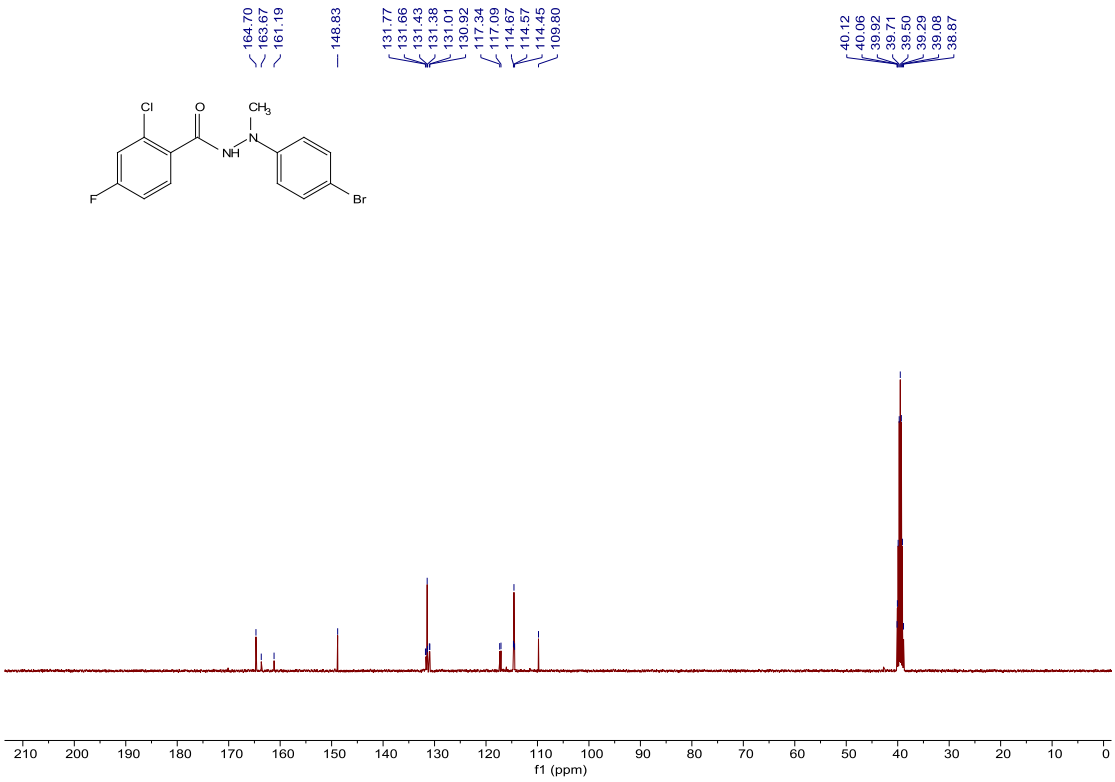
^{1}H NMR of 22 in DMSO-d_6 (400 MHz)



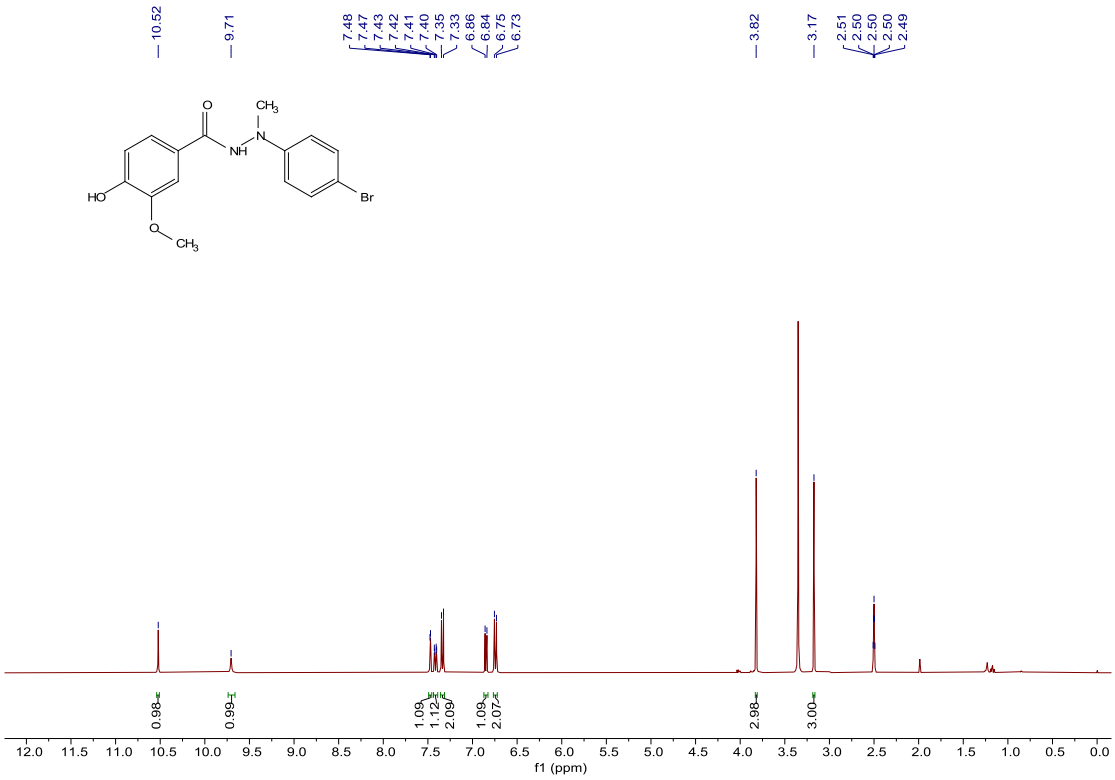
^{13}C{^{1}H} NMR of 22 in DMSO-d_6 (100 MHz)



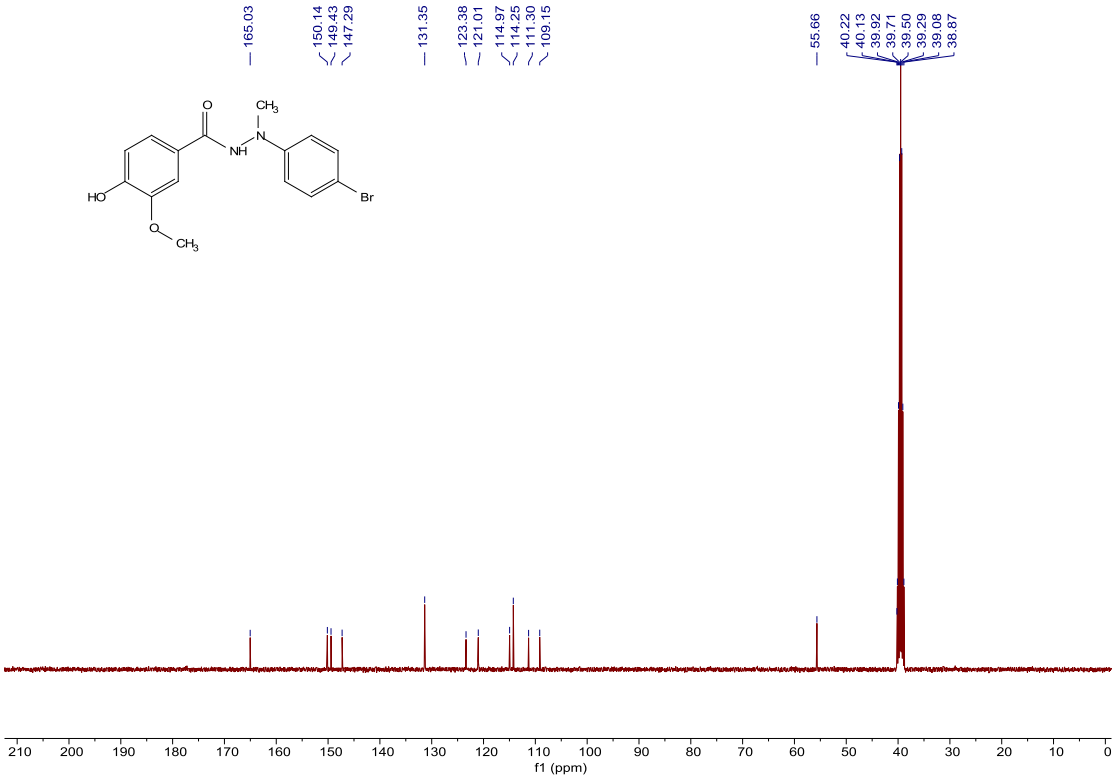
^{1}H NMR of 23 in DMSO-d_6 (400 MHz)



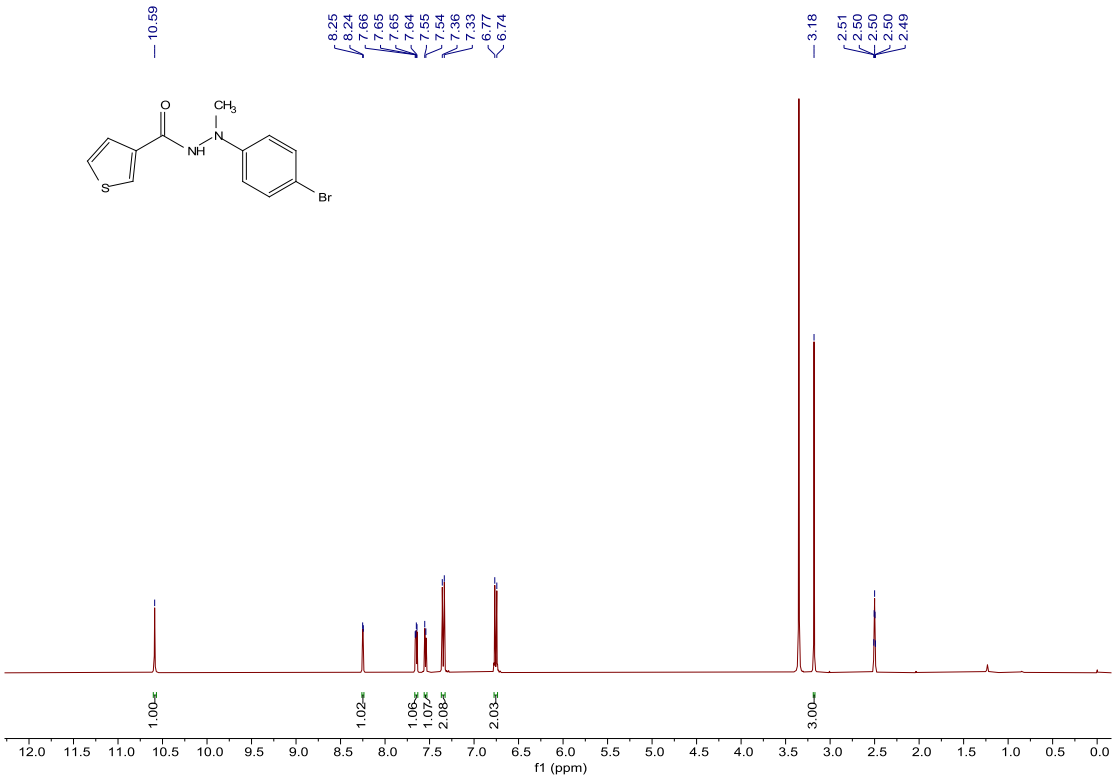
^{13}C{^{1}H} NMR of 23 in DMSO-d_6 (100 MHz)



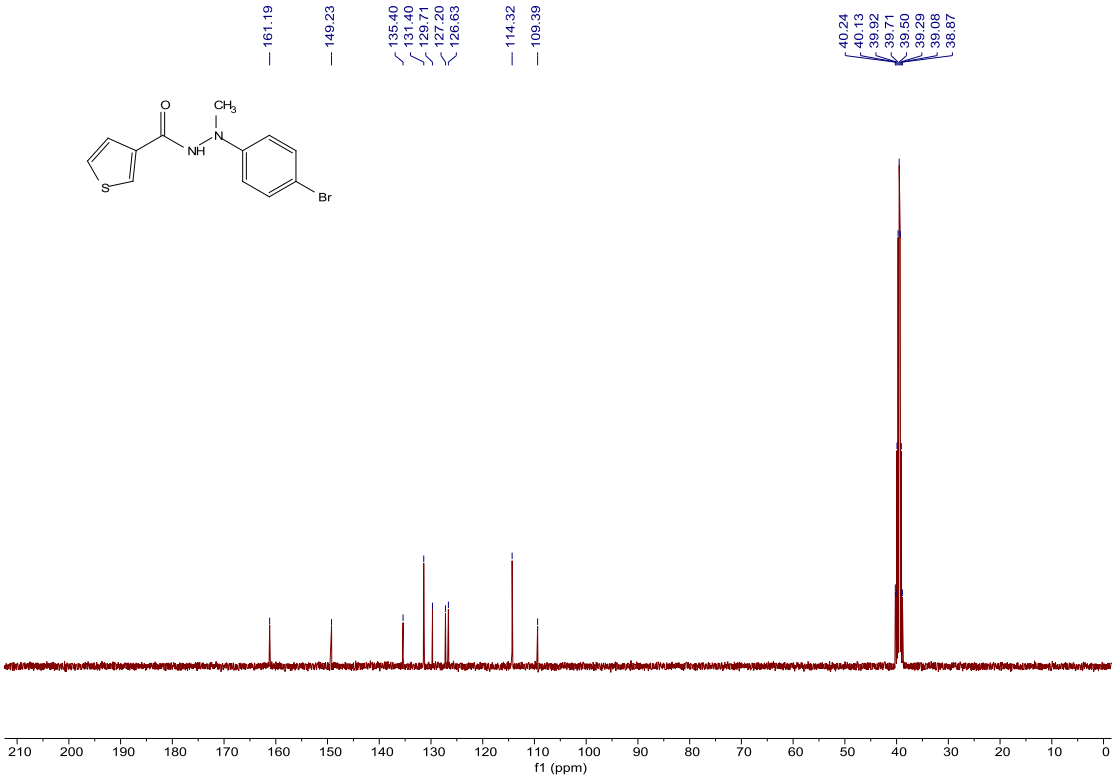
^{1}H NMR of 24 in DMSO-d_6 (400 MHz)



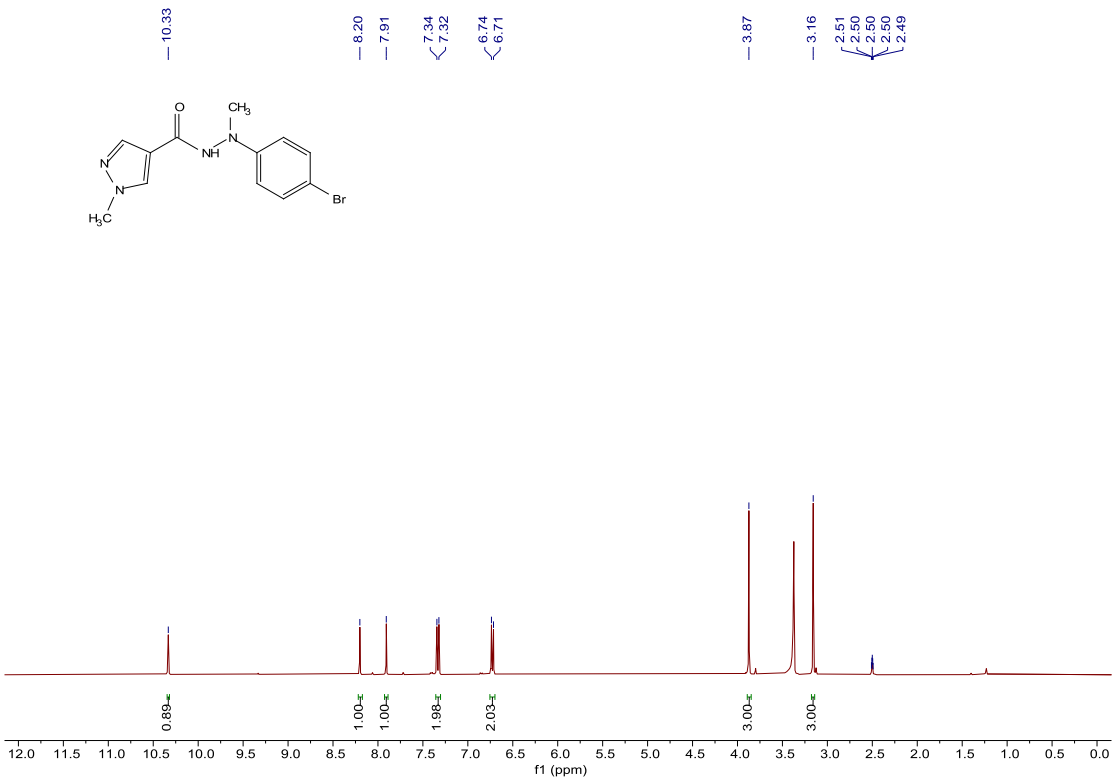
^{13}C{^{1}H} NMR of 24 in DMSO-d_6 (100 MHz)



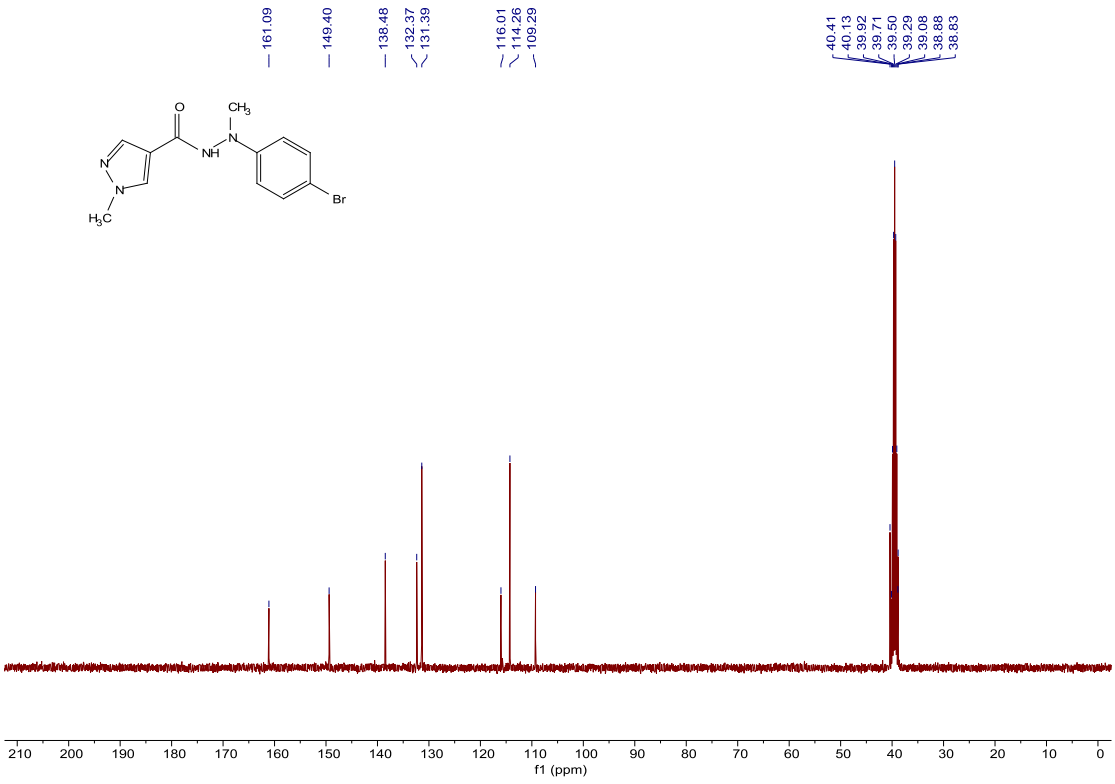
^{1}H NMR of 25 in DMSO-d_6 (400 MHz)



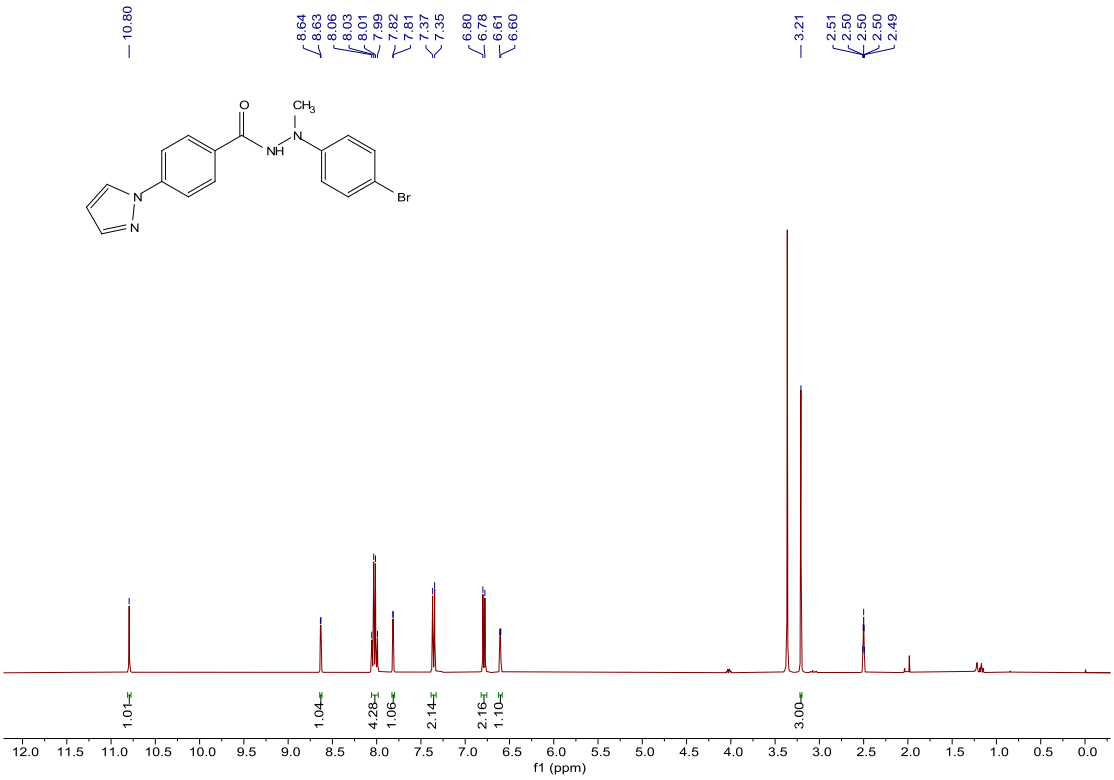
^{13}C{^{1}H} NMR of 25 in DMSO-d_6 (100 MHz)



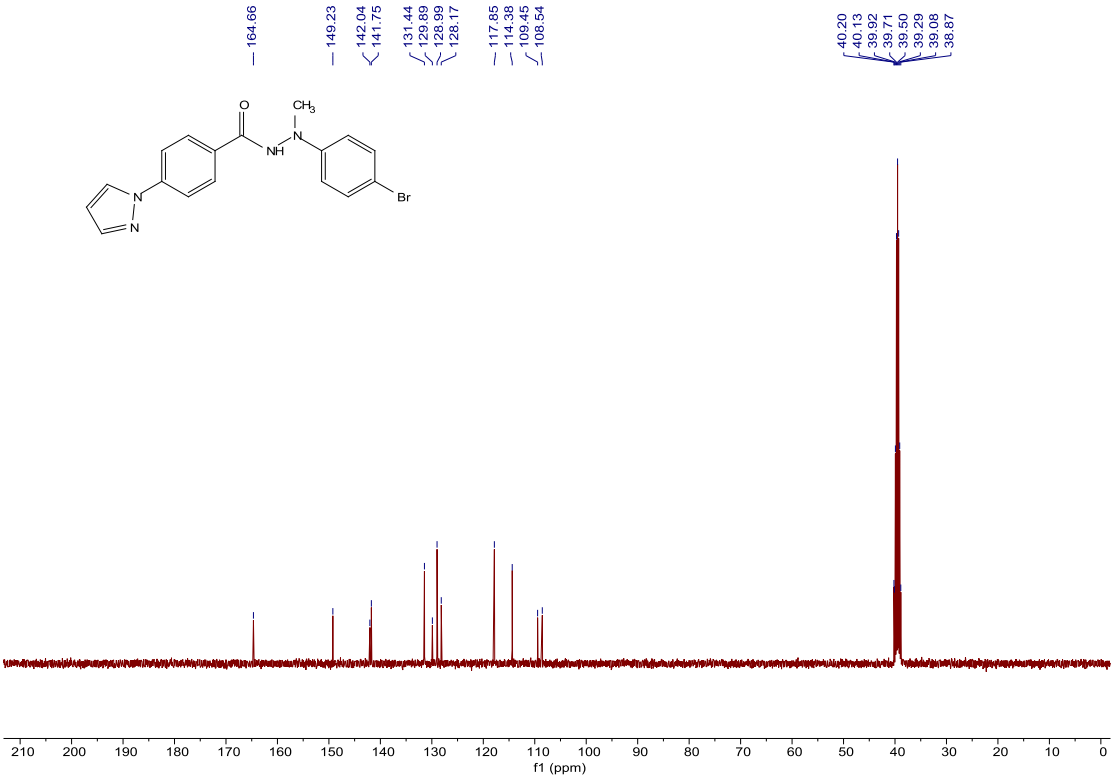
^{1}H NMR of 26 in DMSO-d_6 (400 MHz)



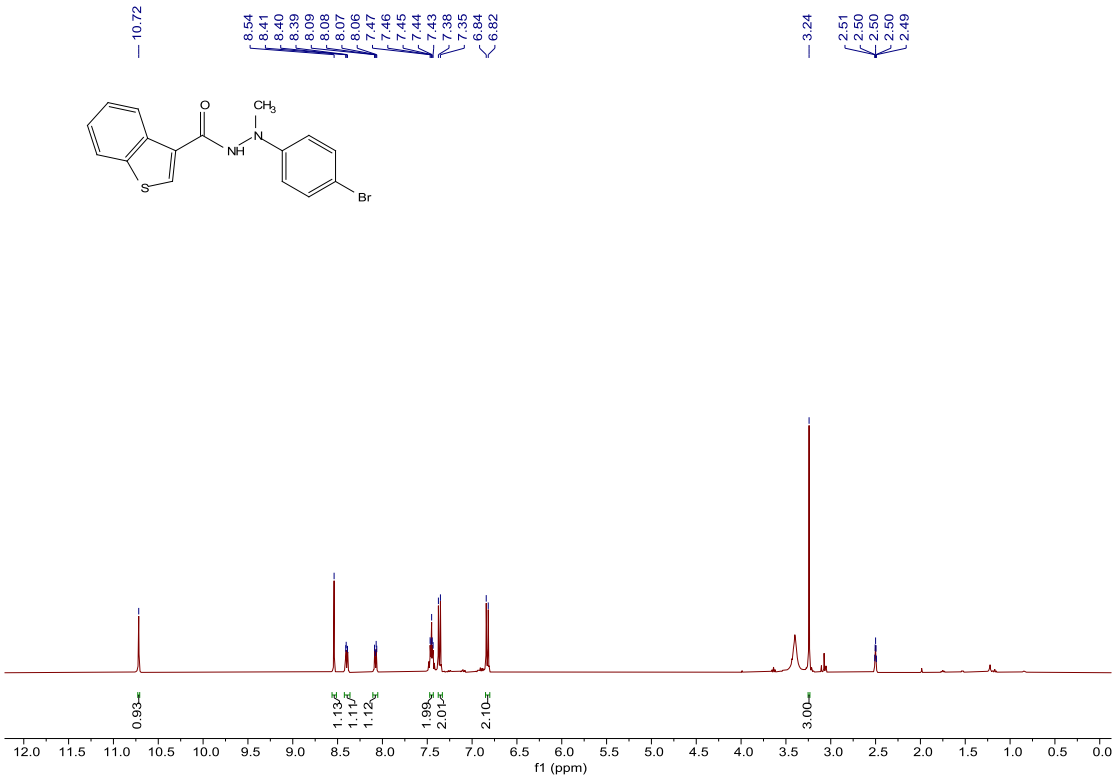
^{13}C{^{1}H} NMR of 26 in DMSO-d_6 (100 MHz)



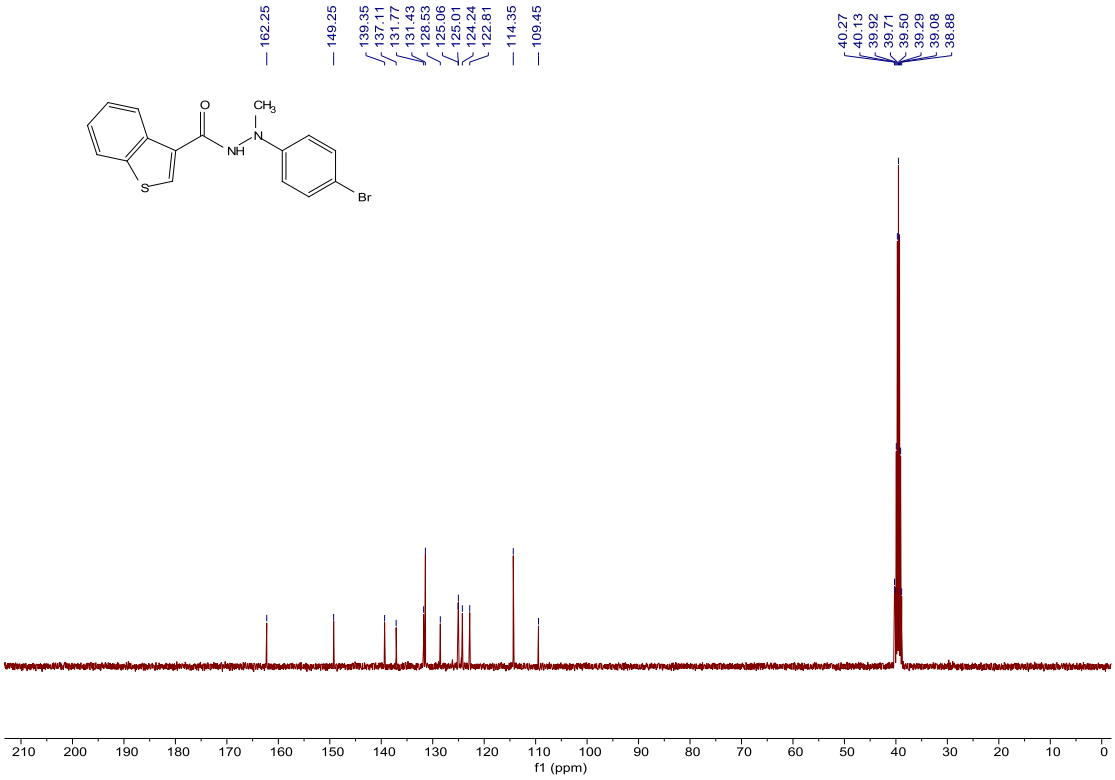
^{1}H NMR of 27 in DMSO-d_6 (400 MHz)



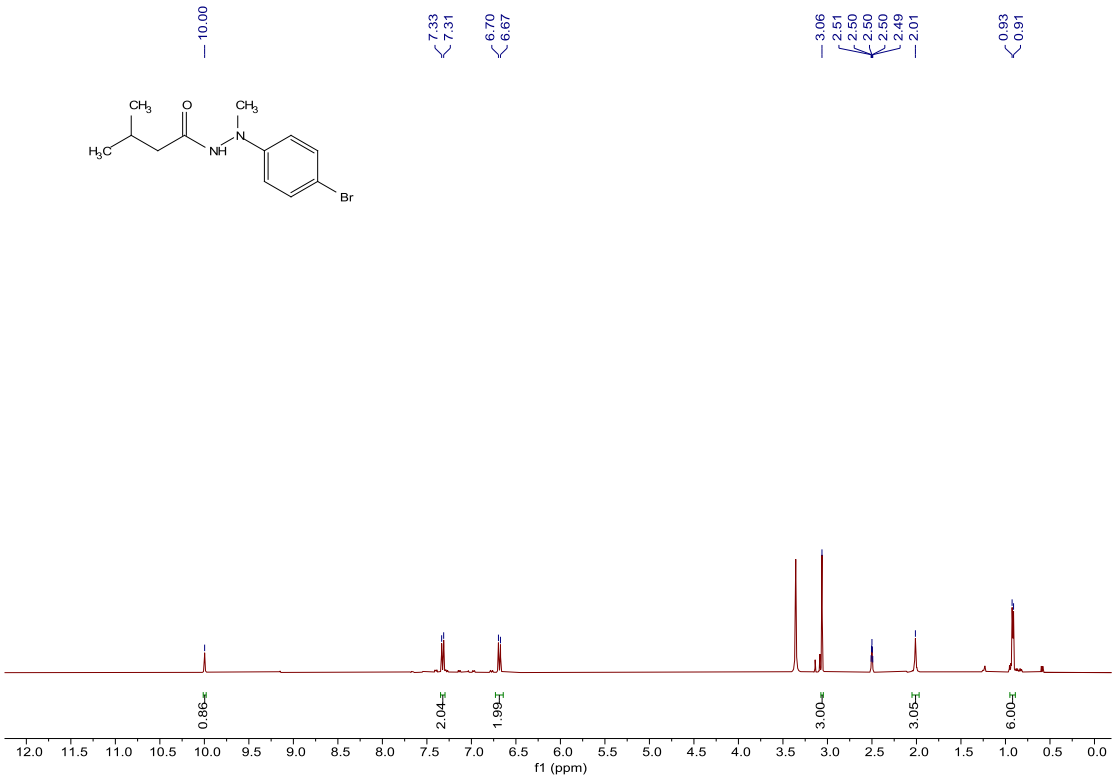
^{13}C{^{1}H} NMR of 27 in DMSO-d_6 (100 MHz)



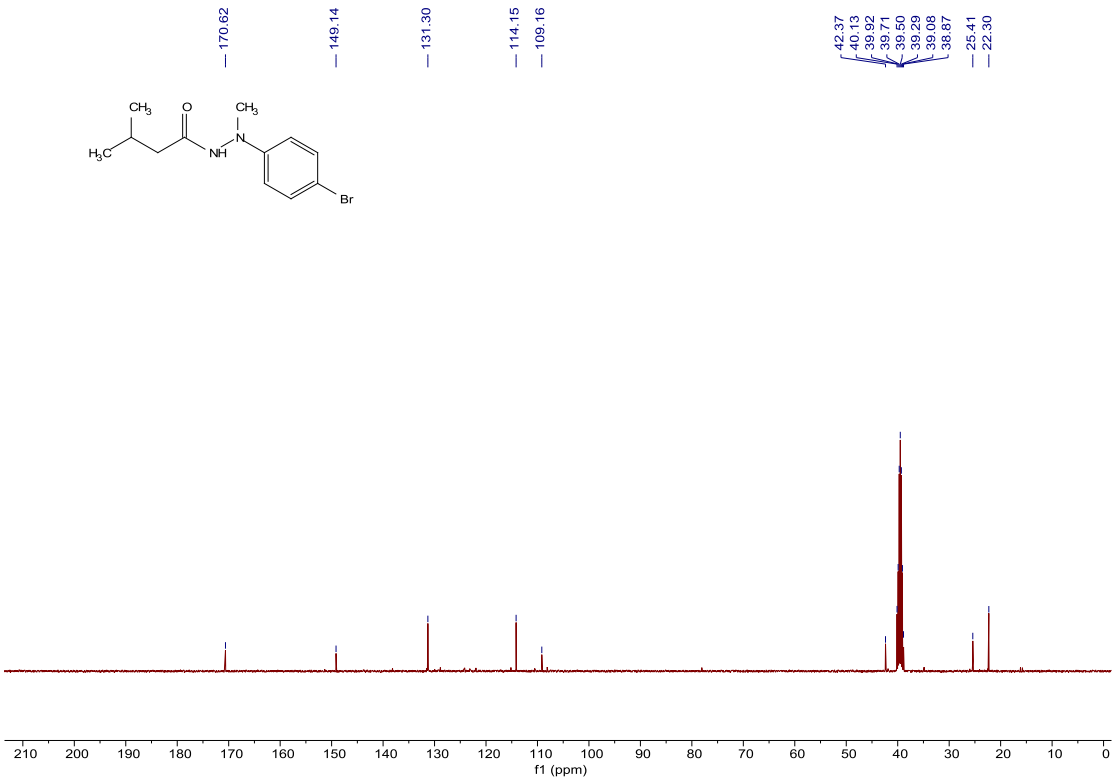
^{1}H NMR of 28 in DMSO-d_6 (400 MHz)



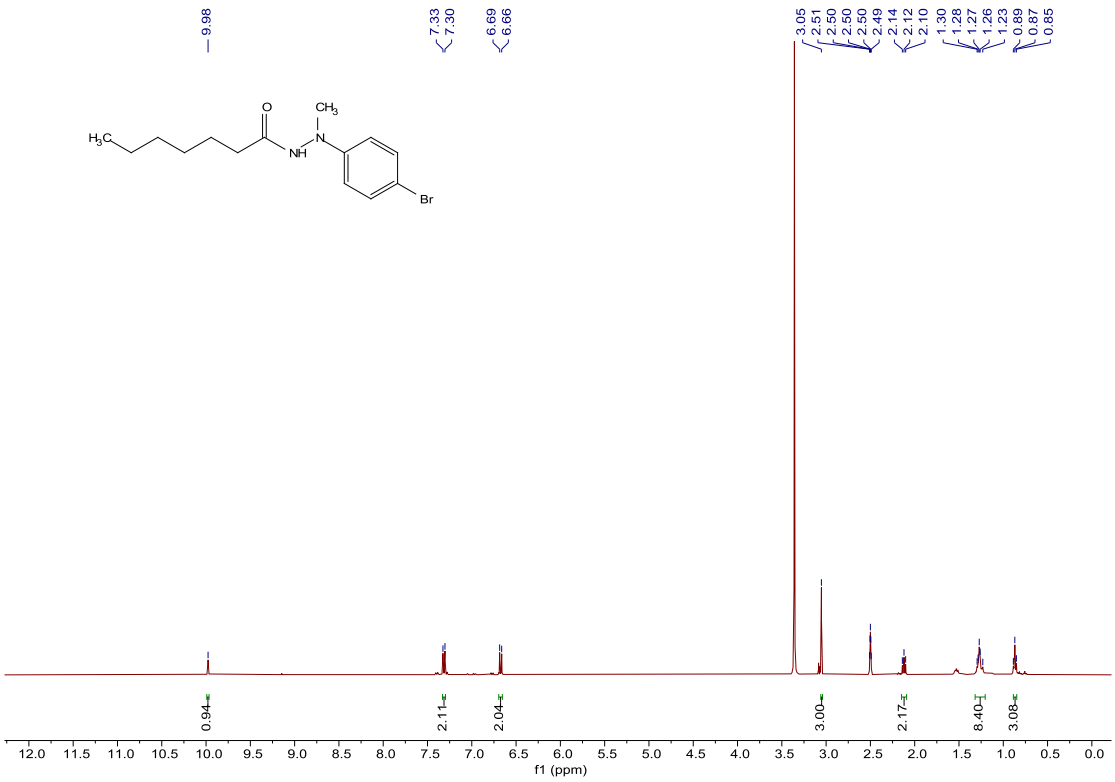
^{13}C{^{1}H} NMR of 28 in DMSO-d_6 (100 MHz)



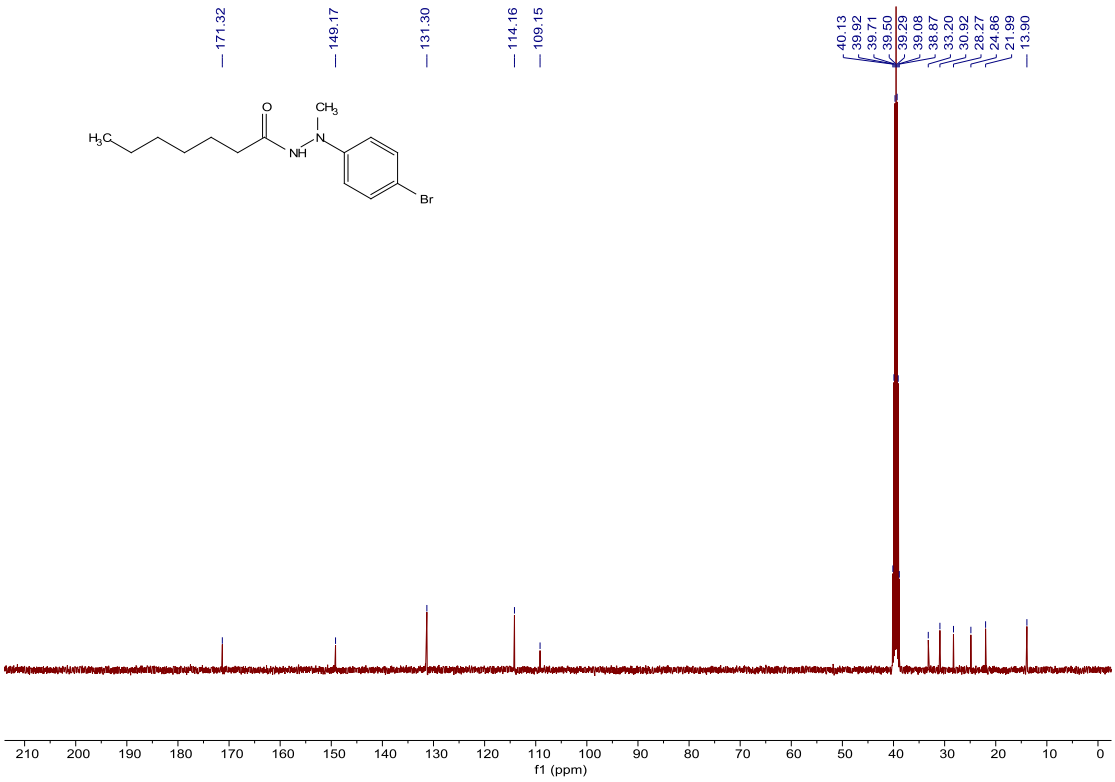
^{1}H NMR of 29 in DMSO-d_6 (400 MHz)



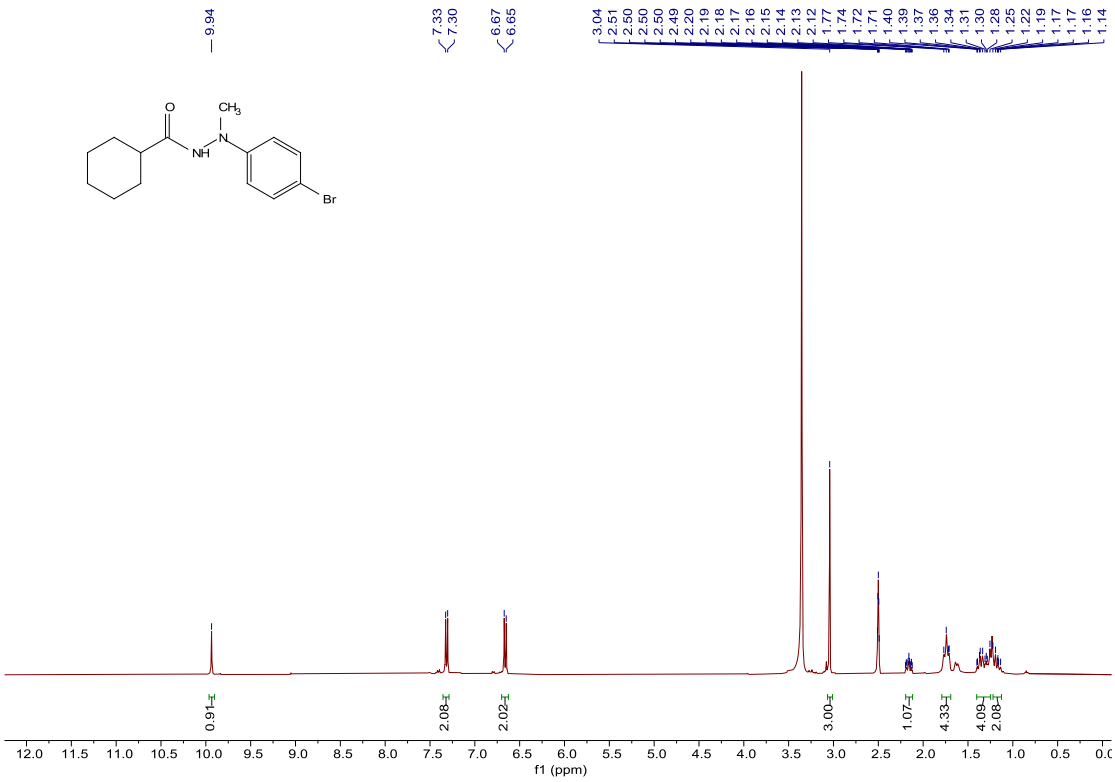
^{13}C{^{1}H} NMR of 29 in DMSO-d_6 (100 MHz)



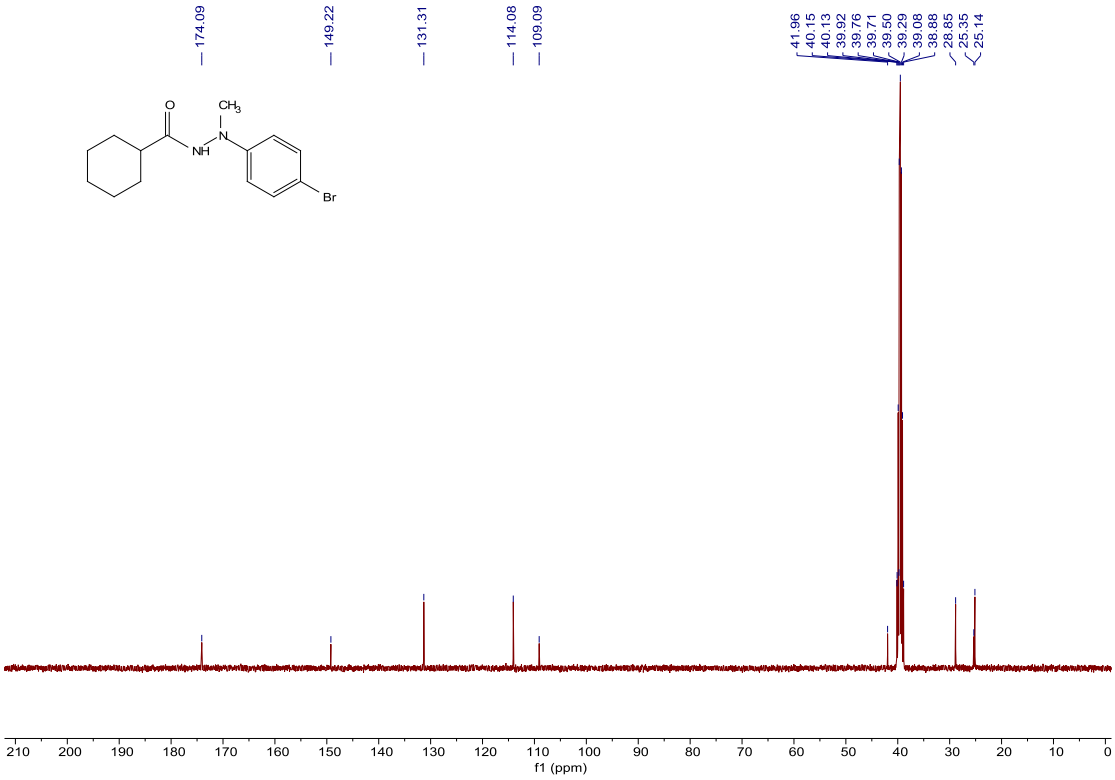
^{1}H NMR of 30 in DMSO-d_6 (400 MHz)



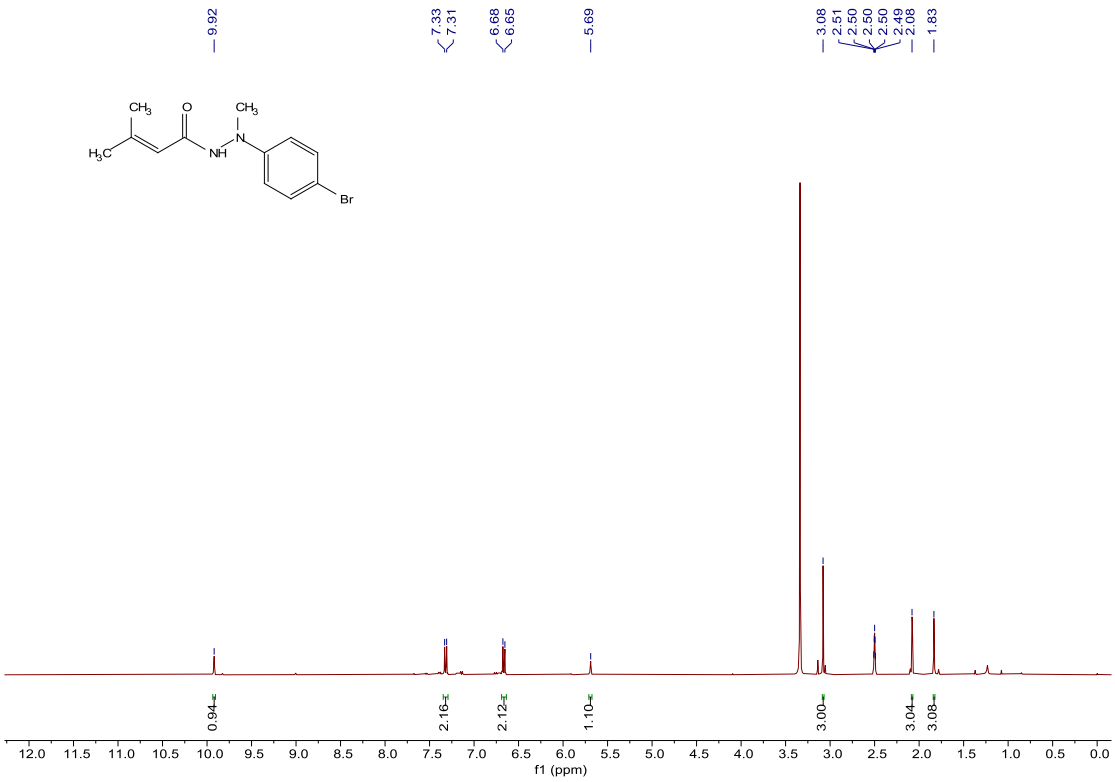
^{13}C{^{1}H} NMR of 30 in DMSO-d_6 (100 MHz)



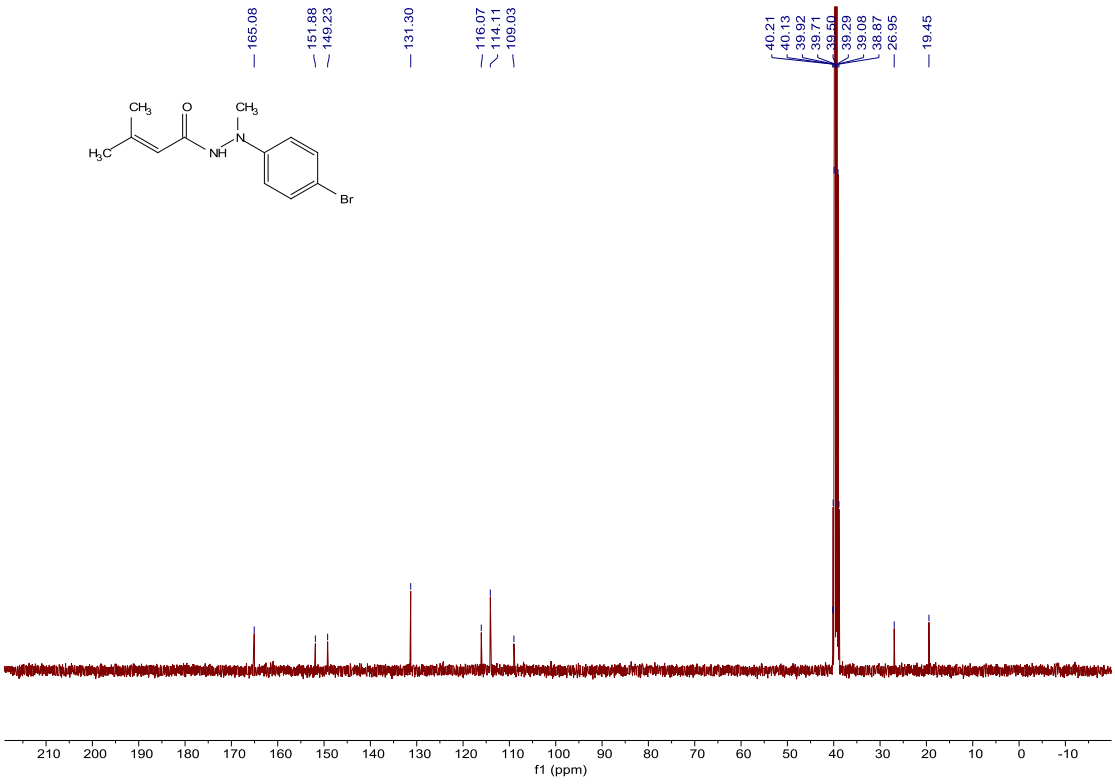
^{1}H NMR of 31 in DMSO-d_6 (400 MHz)



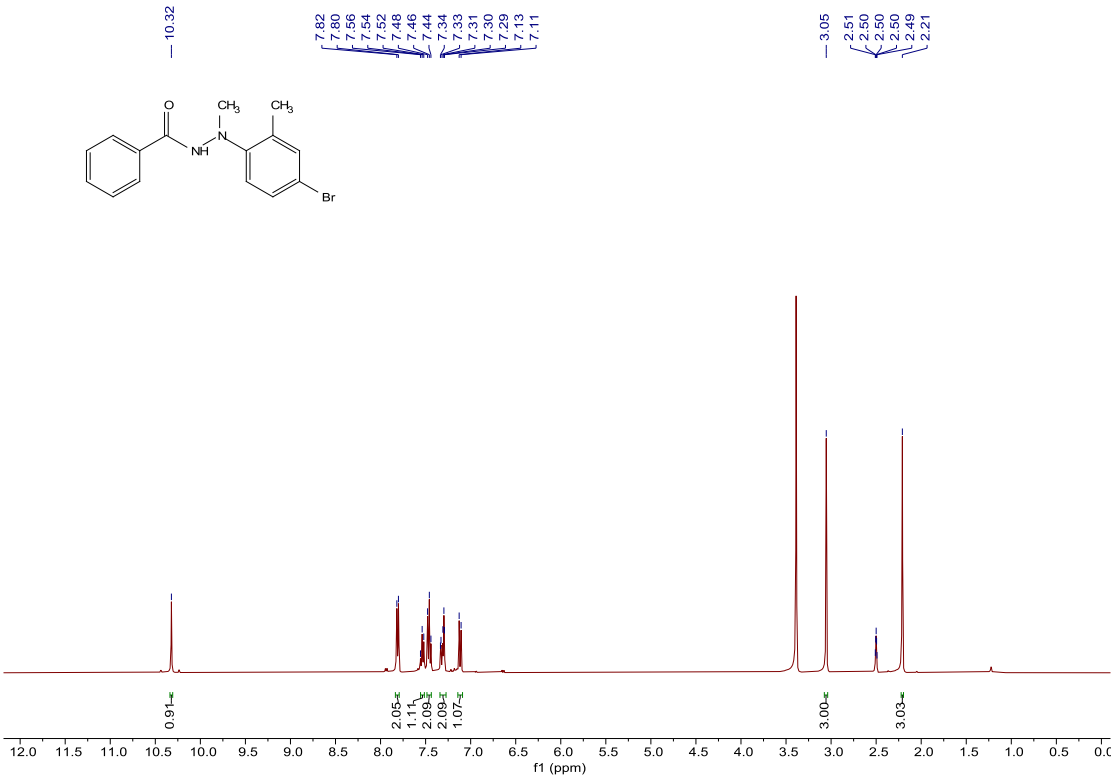
^{13}C{^{1}H} NMR of 31 in DMSO-d_6 (100 MHz)



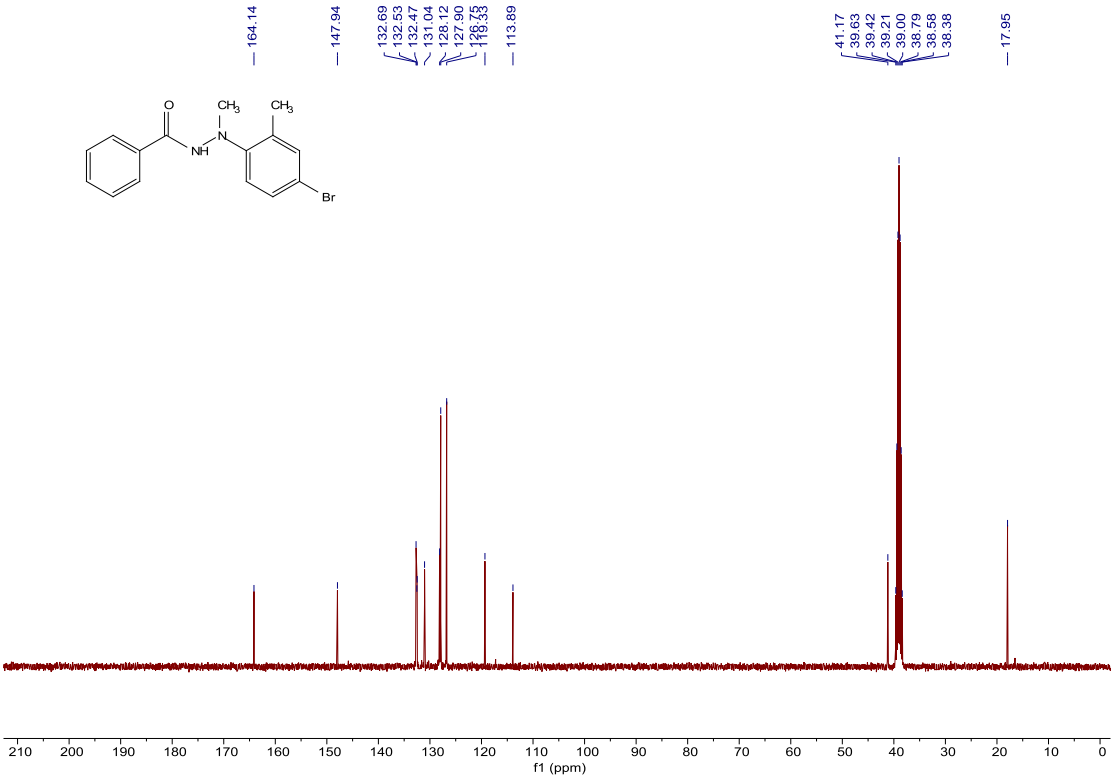
^{1}H NMR of 32 in DMSO-d_6 (400 MHz)



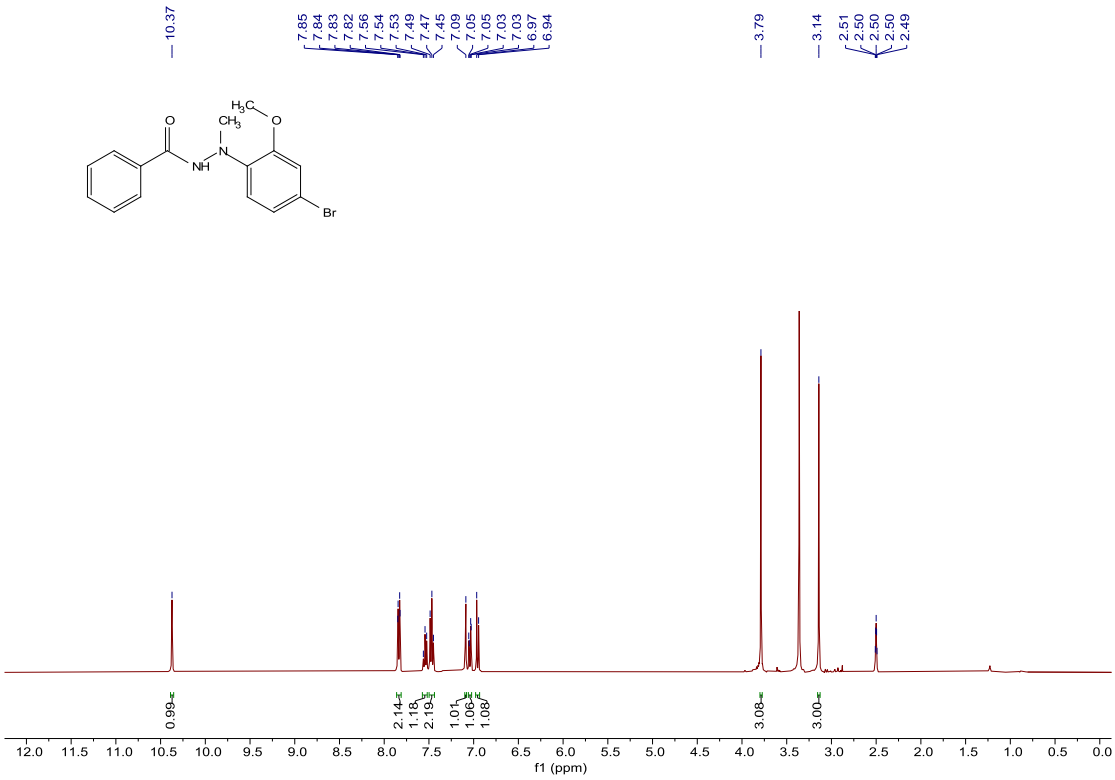
^{13}C{^{1}H} NMR of 32 in DMSO-d_6 (100 MHz)



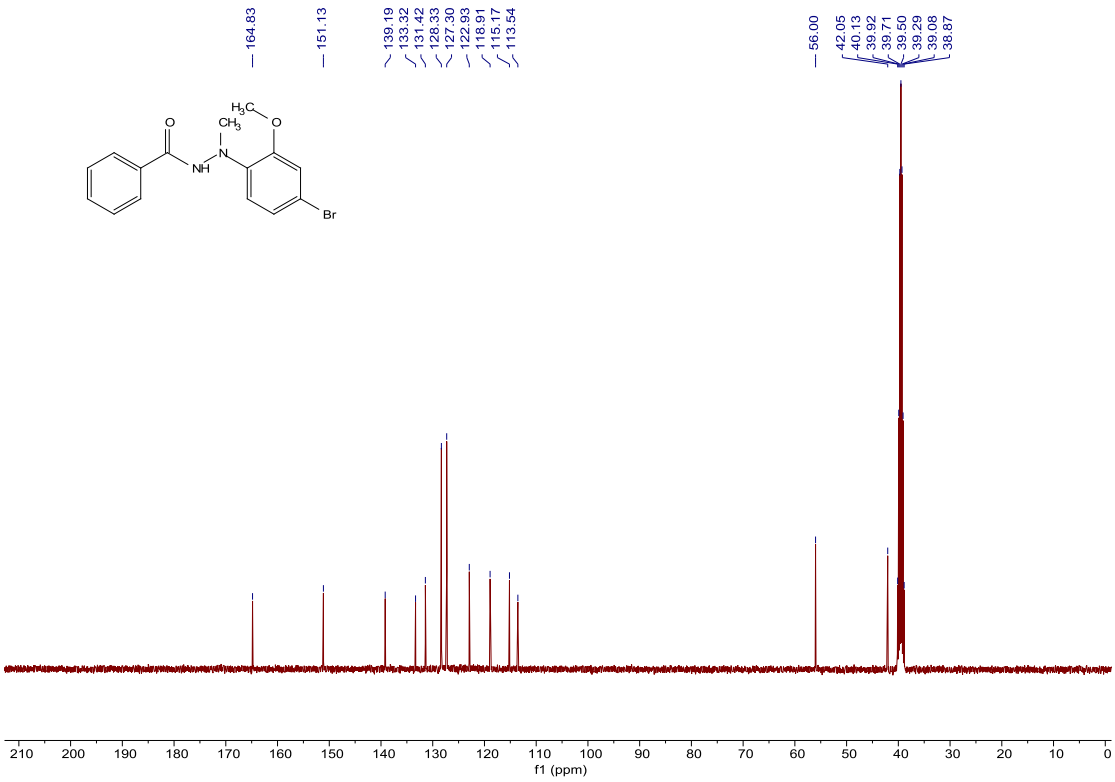
^{1}H NMR of 33 in DMSO-d_6 (400 MHz)



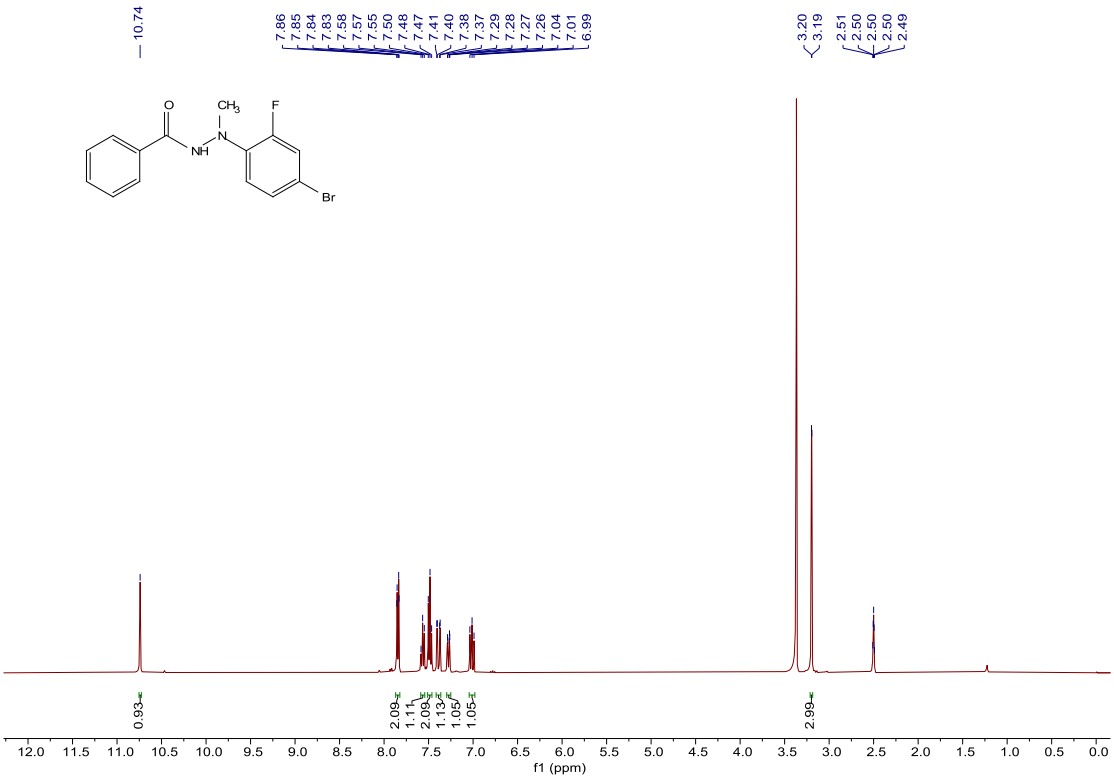
^{13}C{^{1}H} NMR of 33 in DMSO-d_6 (100 MHz)



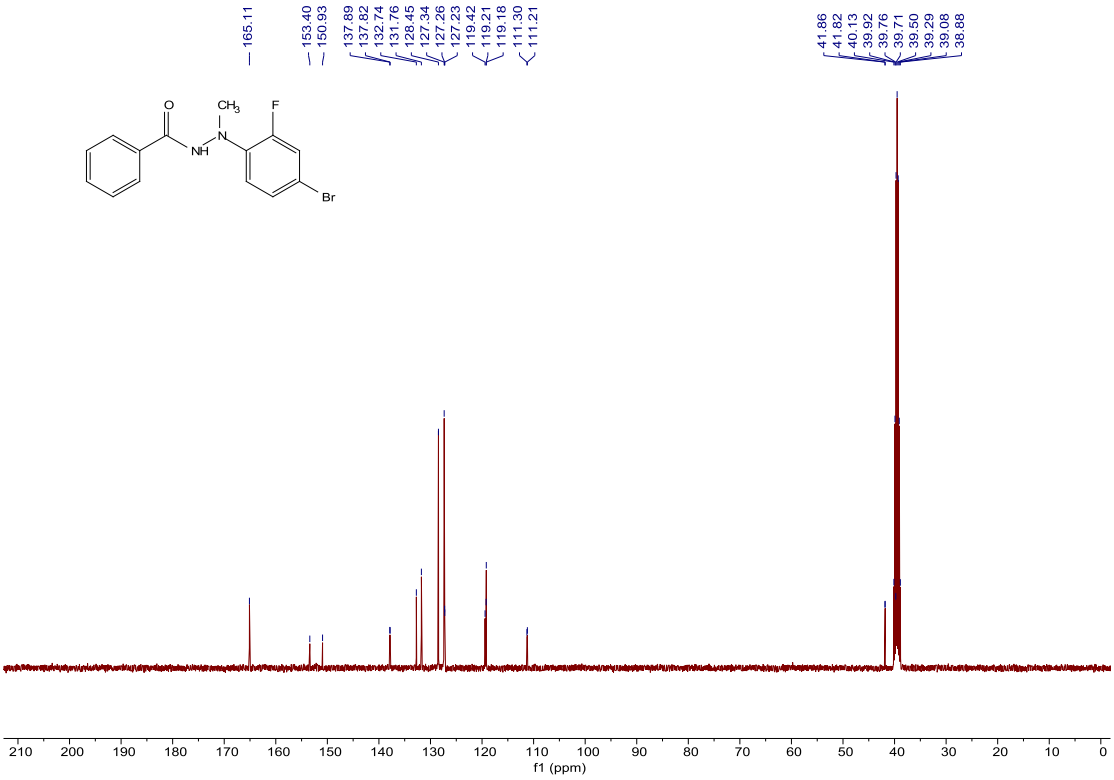
^{1}H NMR of 34 in DMSO-d_6 (400 MHz)



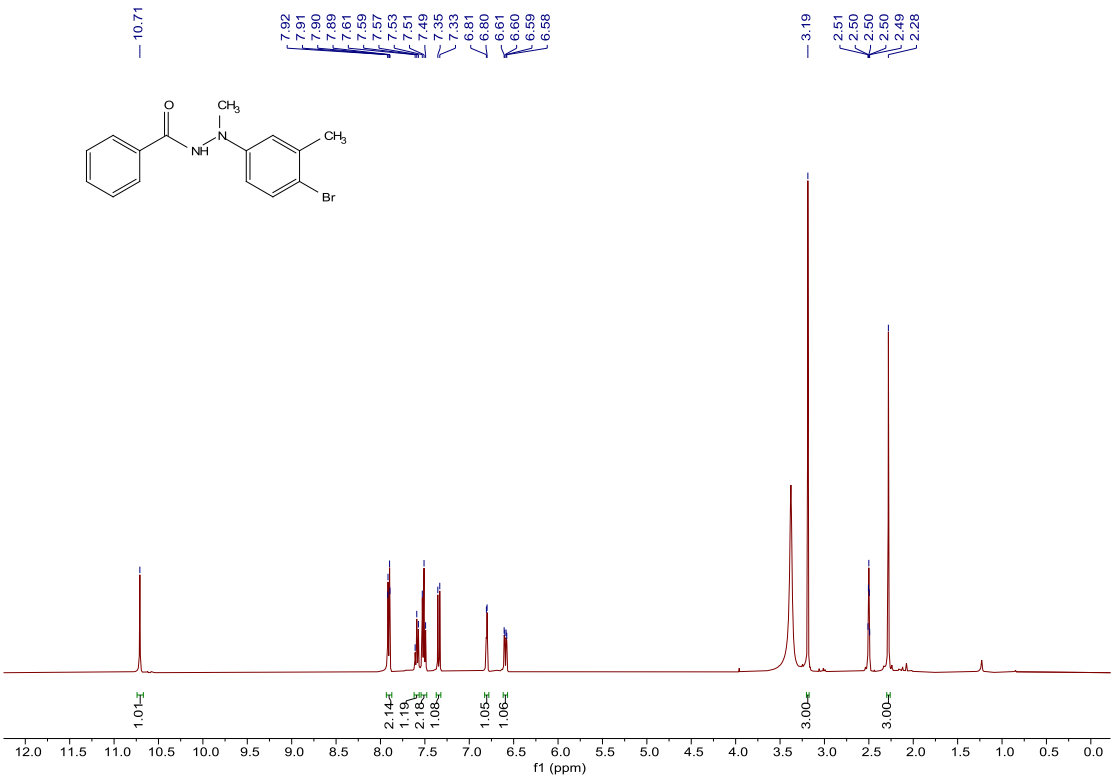
^{13}C{^{1}H} NMR of 34 in DMSO-d_6 (100 MHz)



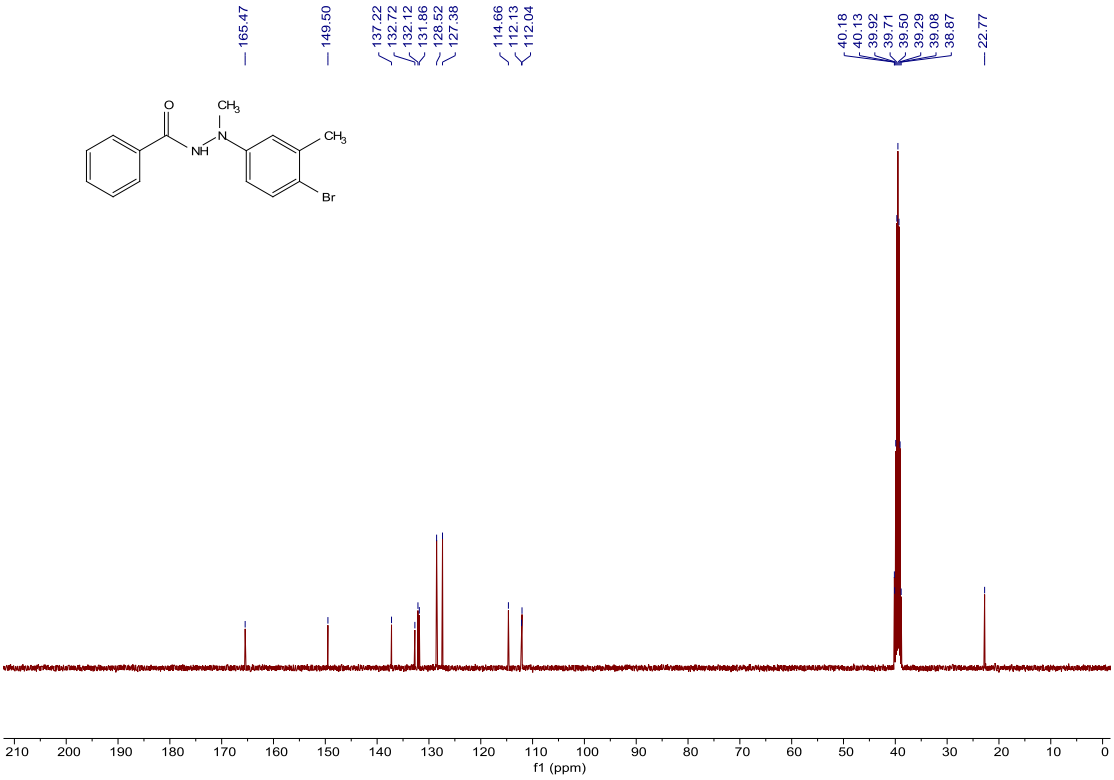
^{1}H NMR of 35 in DMSO-d_6 (400 MHz)



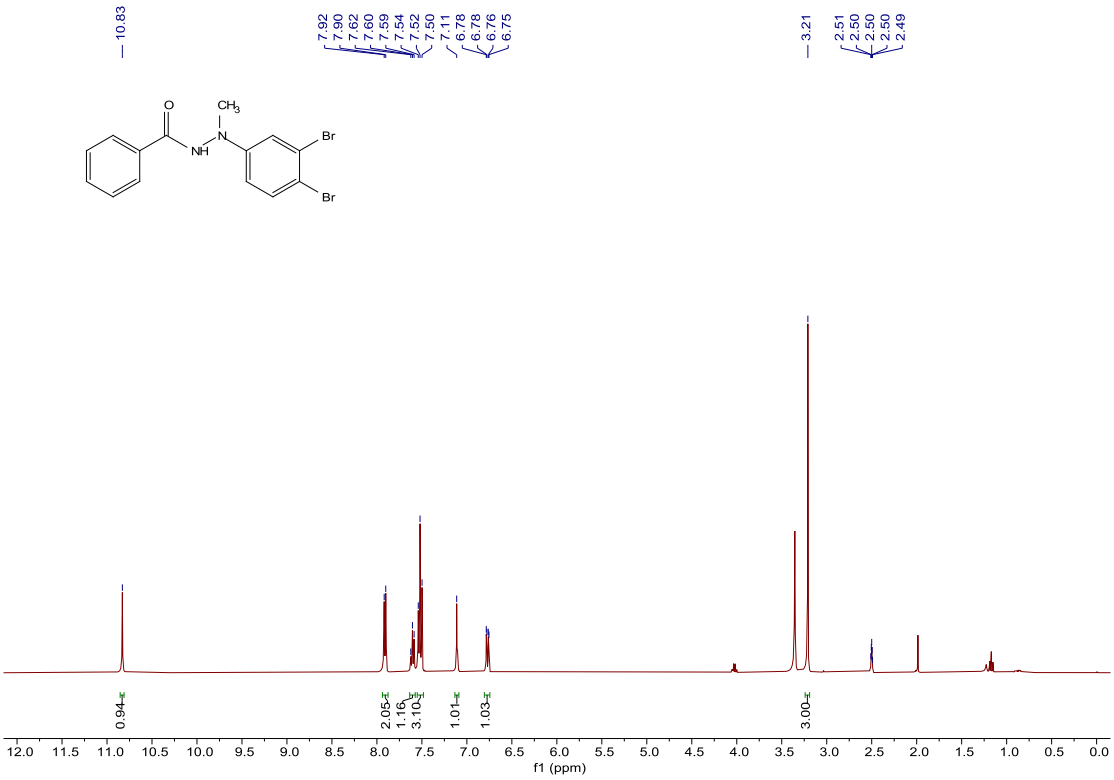
^{13}C{^{1}H} NMR of 35 in DMSO-d_6 (100 MHz)



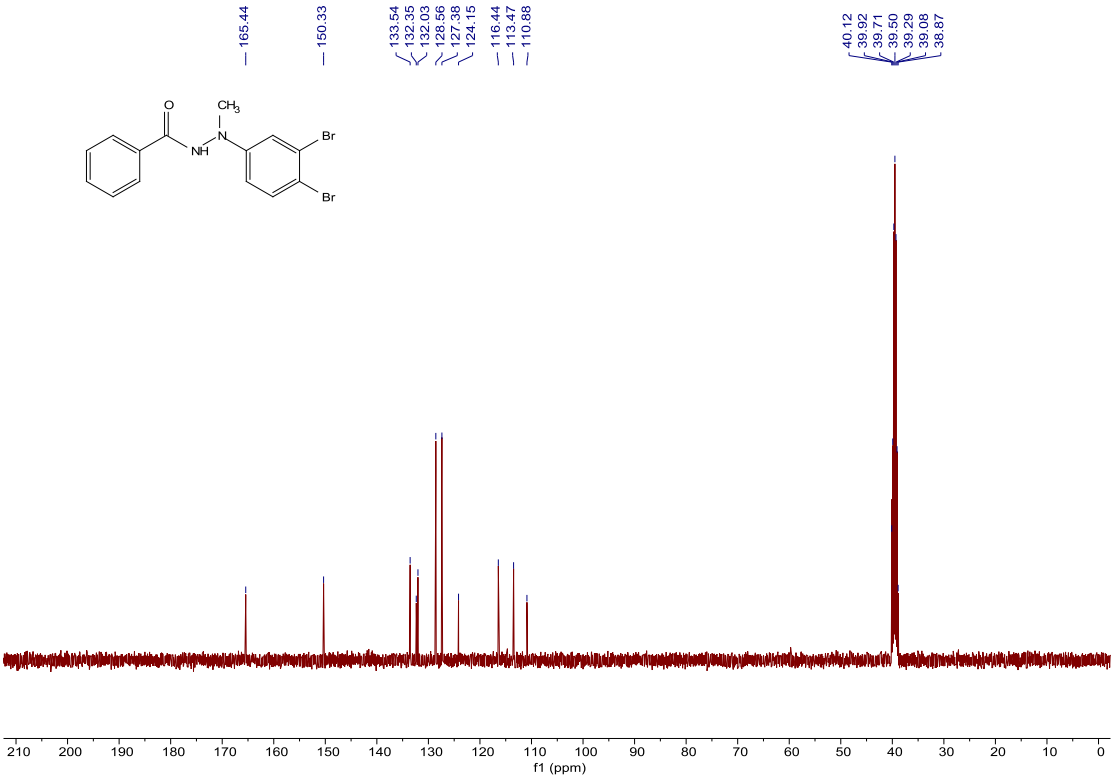
^{1}H NMR of 36 in DMSO-d_6 (400 MHz)



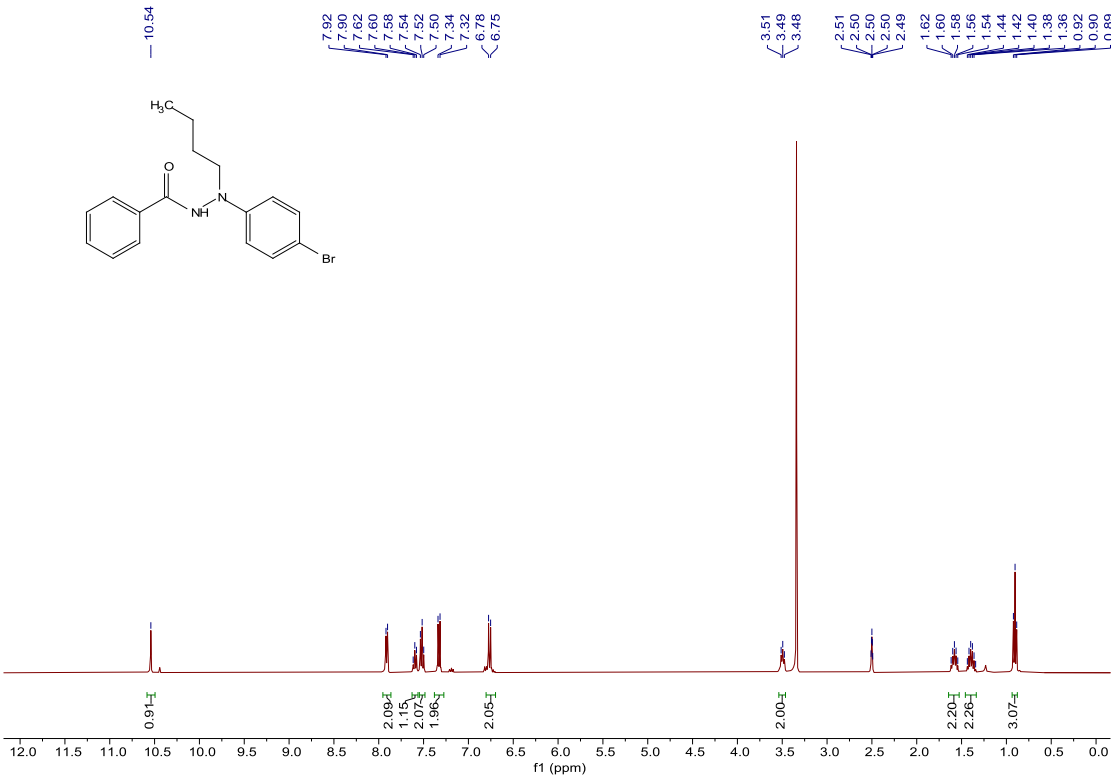
^{13}C{^{1}H} NMR of 36 in DMSO-d_6 (100 MHz)



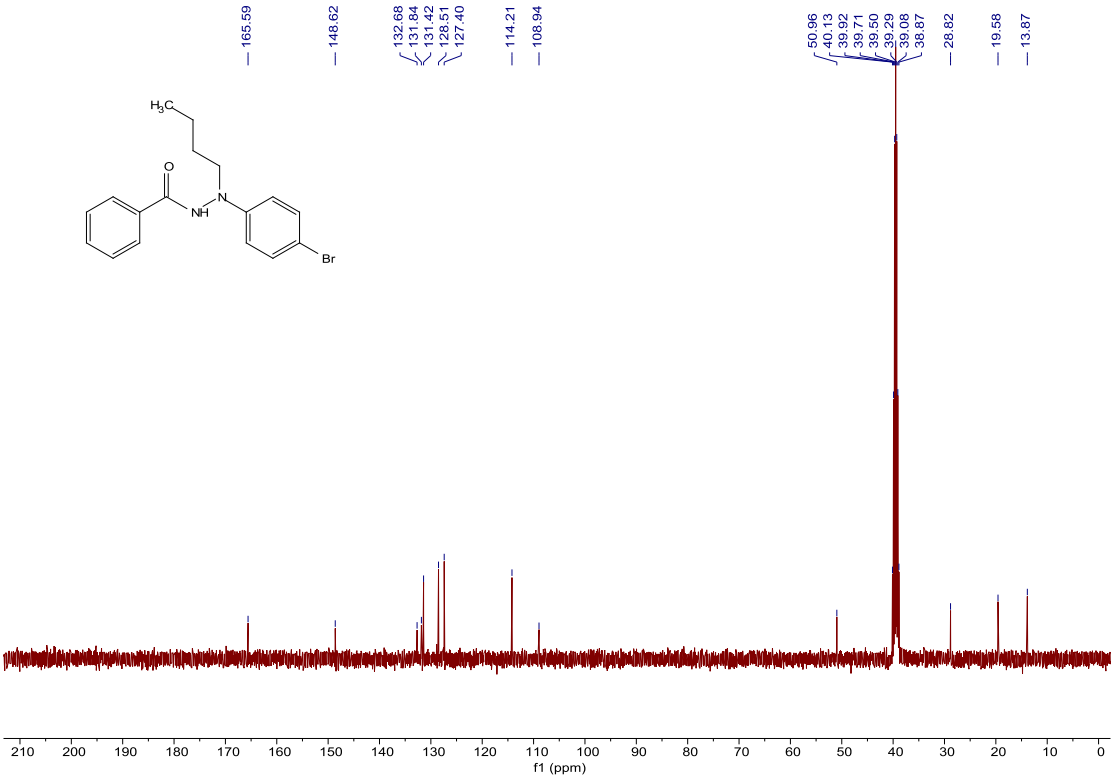
^{1}H NMR of 37 in DMSO-d_6 (400 MHz)



^{13}C{^{1}H} NMR of 37 in DMSO-d_6 (100 MHz)

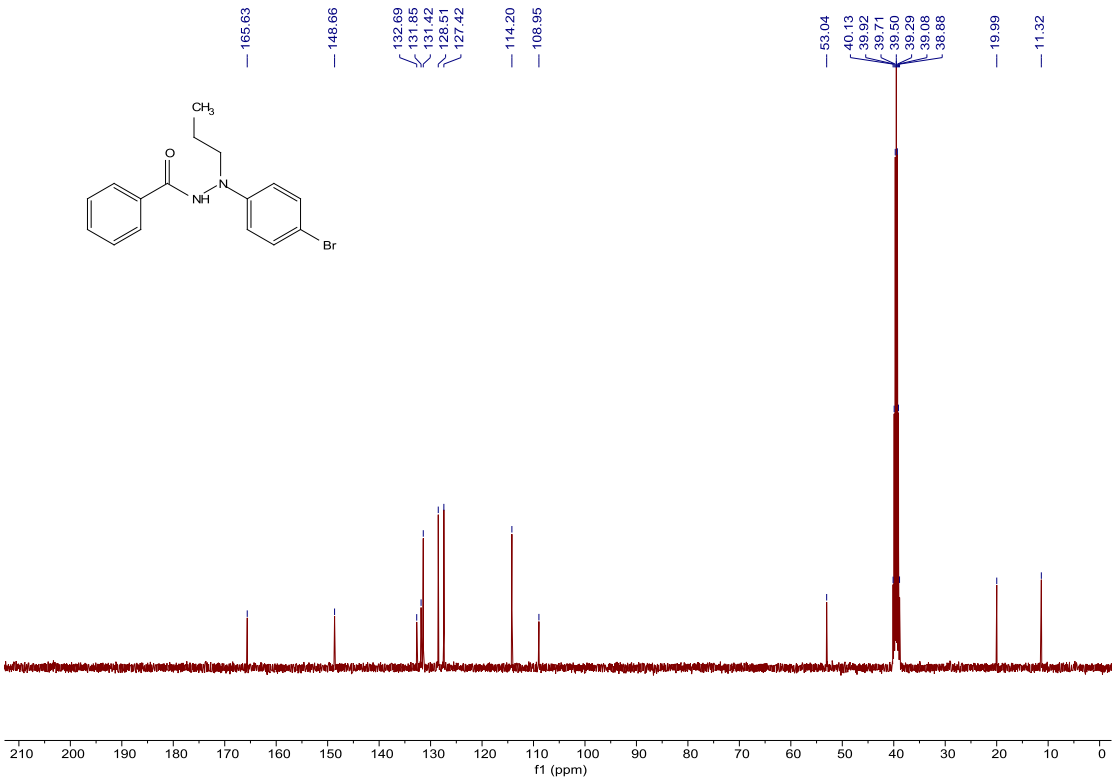


^{1}H NMR of 38 in DMSO-d_6 (400 MHz)

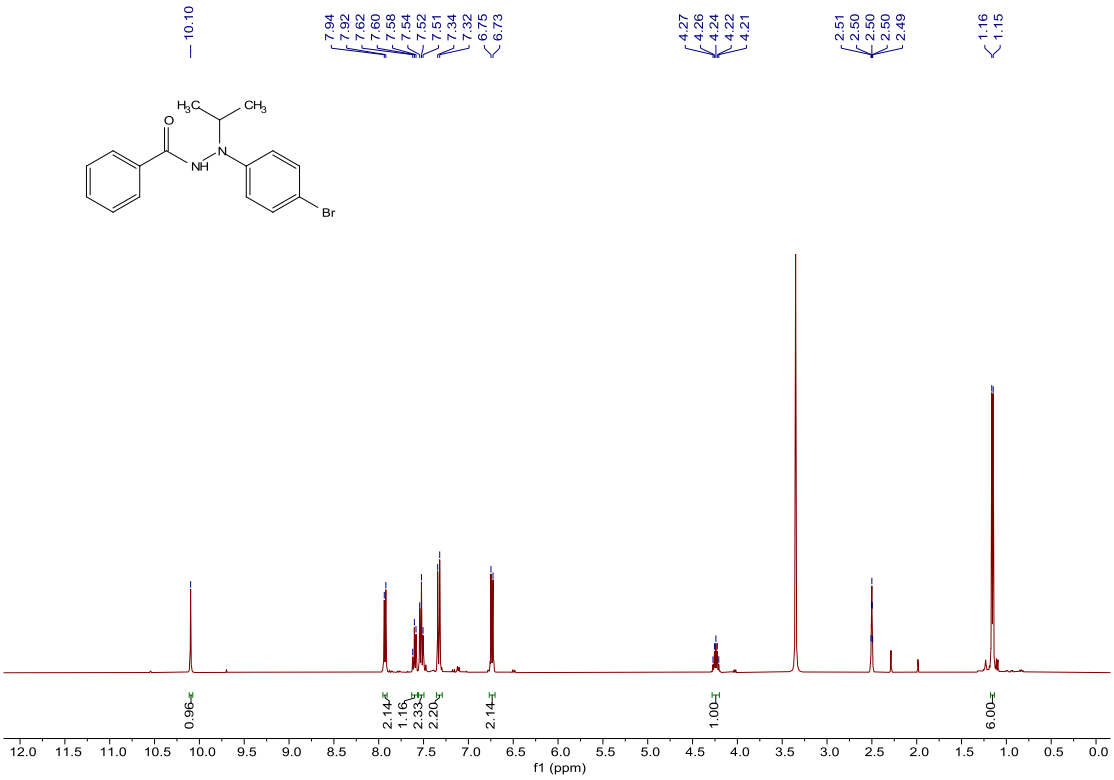


^{13}C{^{1}H} NMR of 38 in DMSO-d_6 (100 MHz)

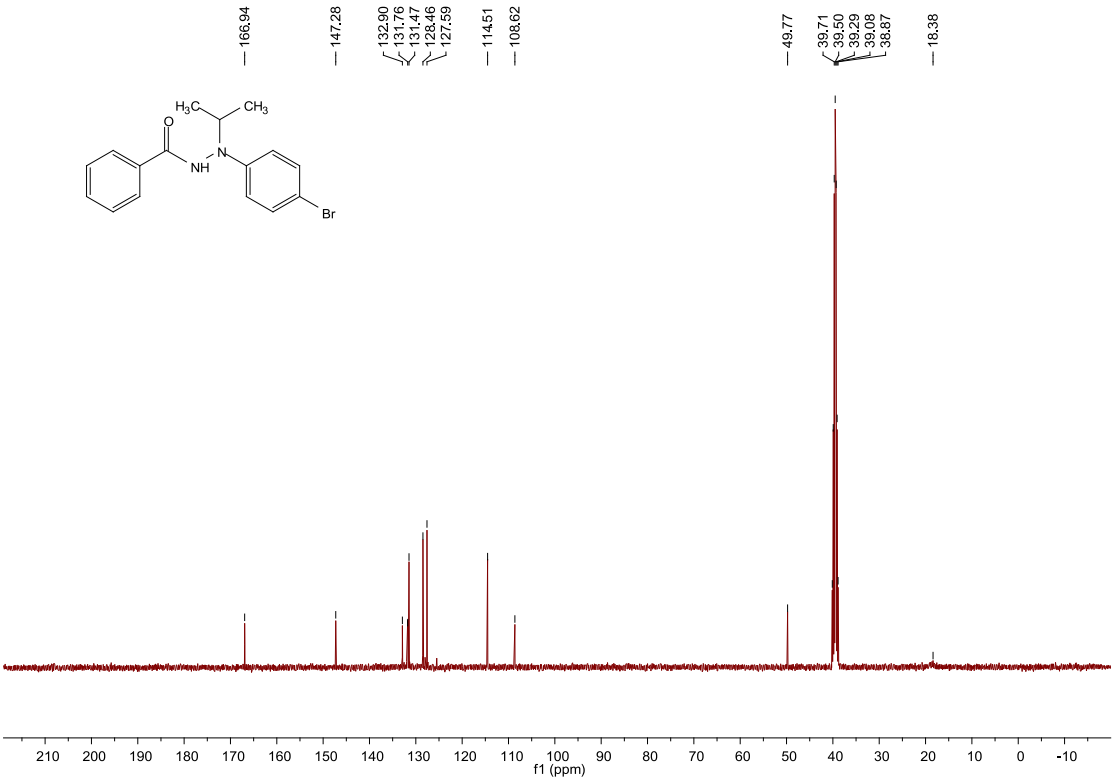
^{1}H NMR of 39 in DMSO-d_6 (400 MHz)



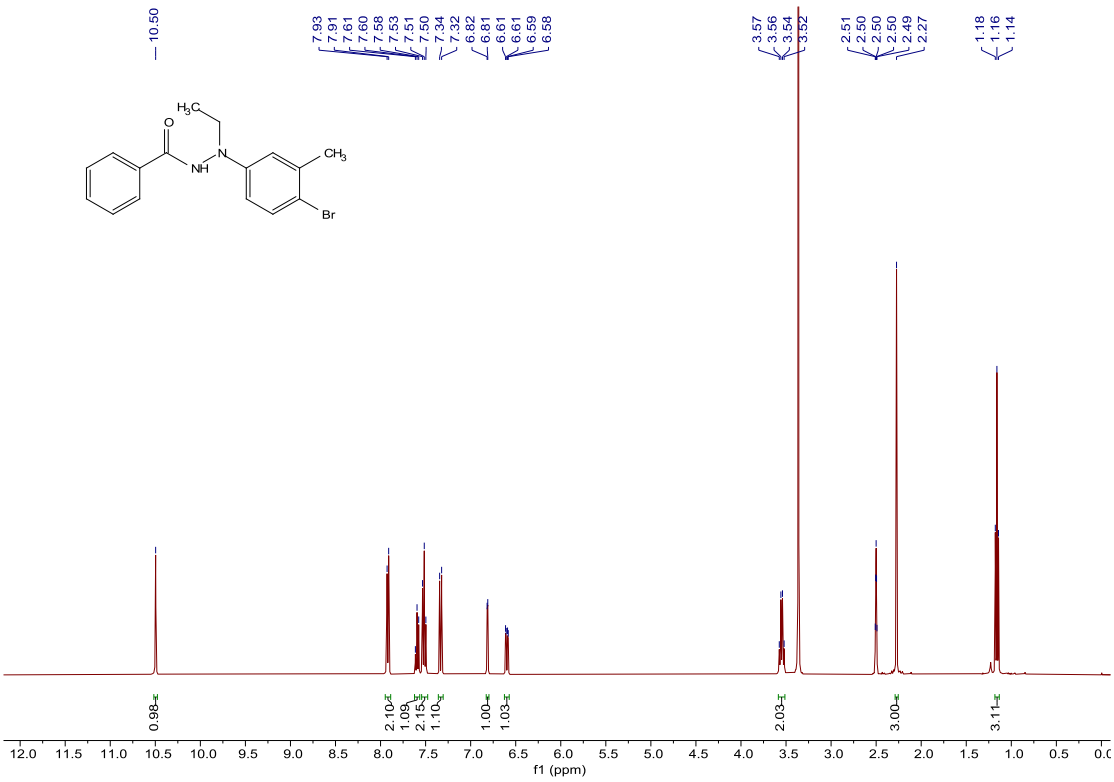
^{13}C{^{1}H} NMR of 39 in DMSO-d_6 (100 MHz)



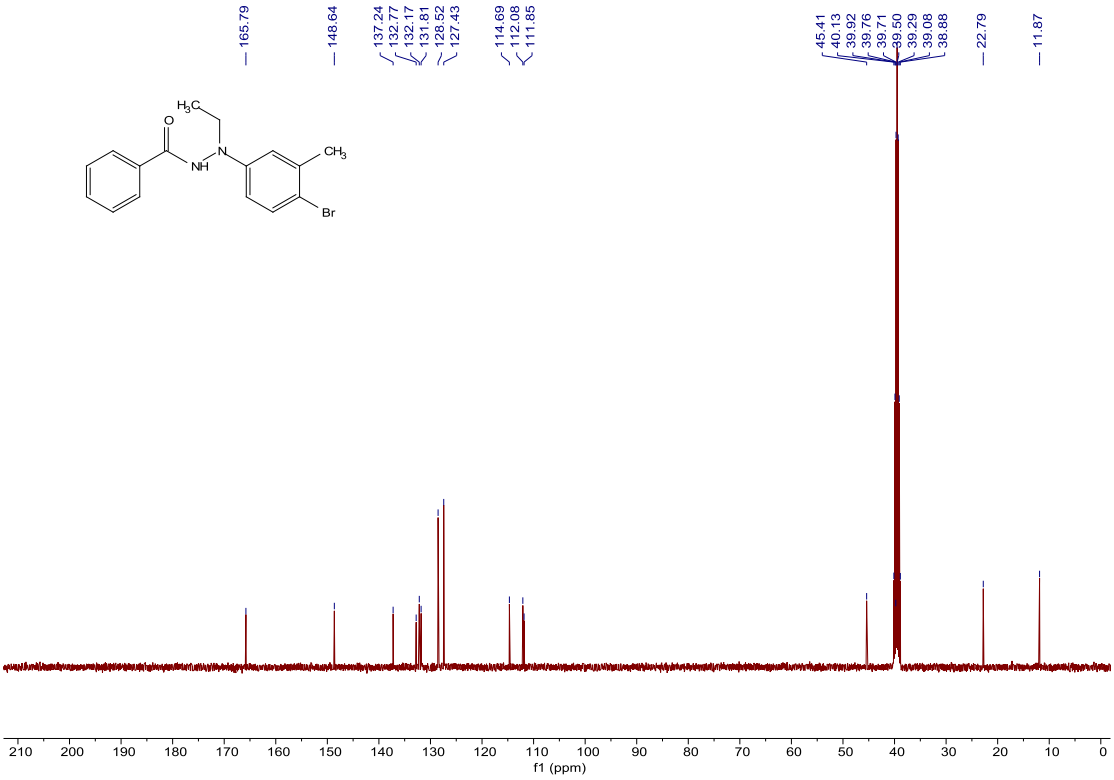
^{1}H NMR of 40 in DMSO-d_6 (400 MHz)



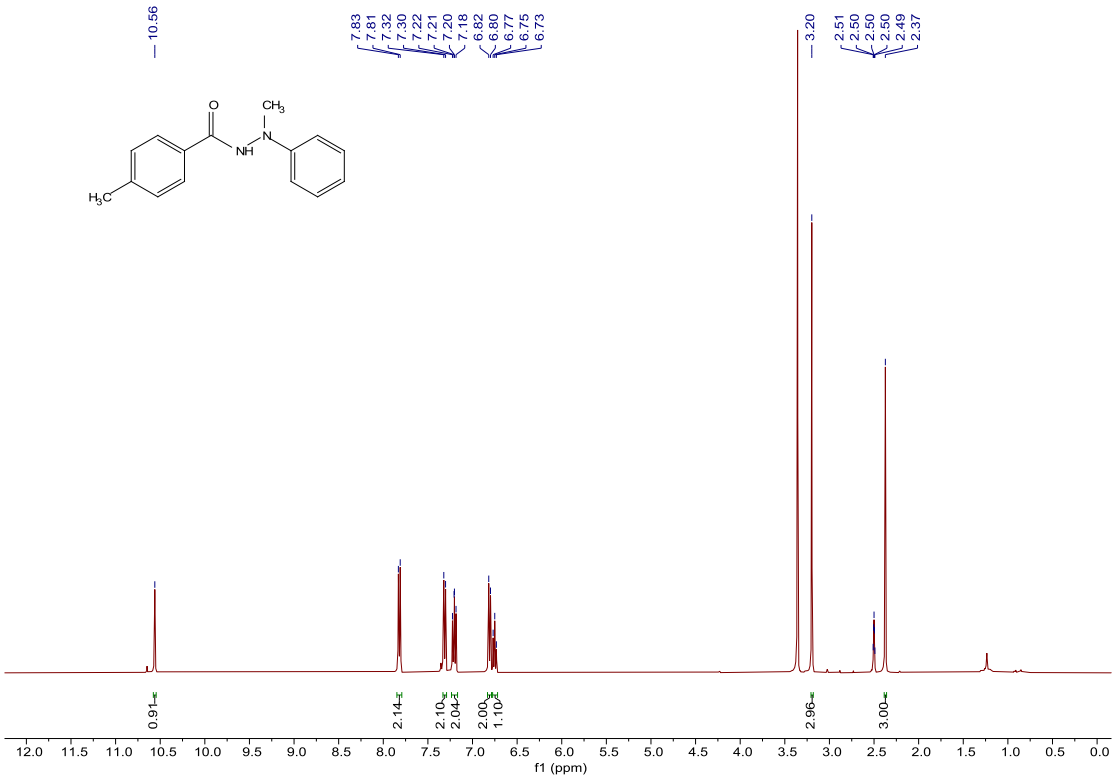
^{13}C{^{1}H} NMR of 40 in DMSO-d_6 (100 MHz)



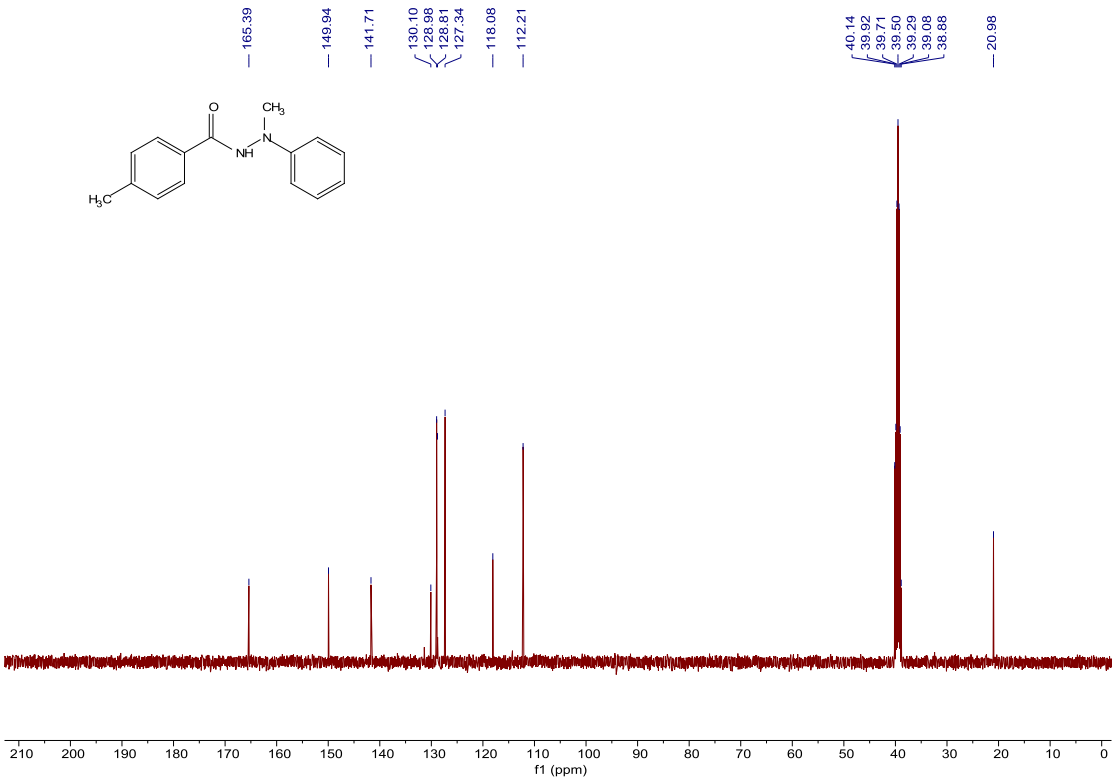
^{1}H NMR of 41 in DMSO-d_6 (400 MHz)



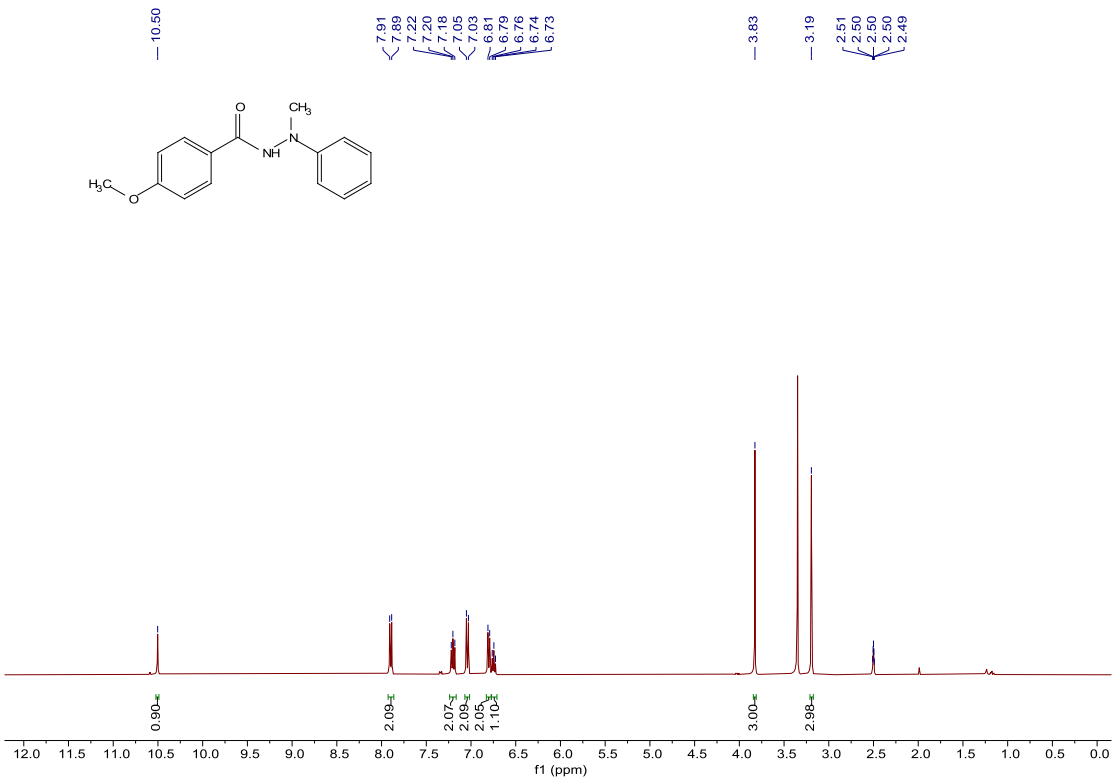
^{13}C{^{1}H} NMR of 41 in DMSO-d_6 (100 MHz)



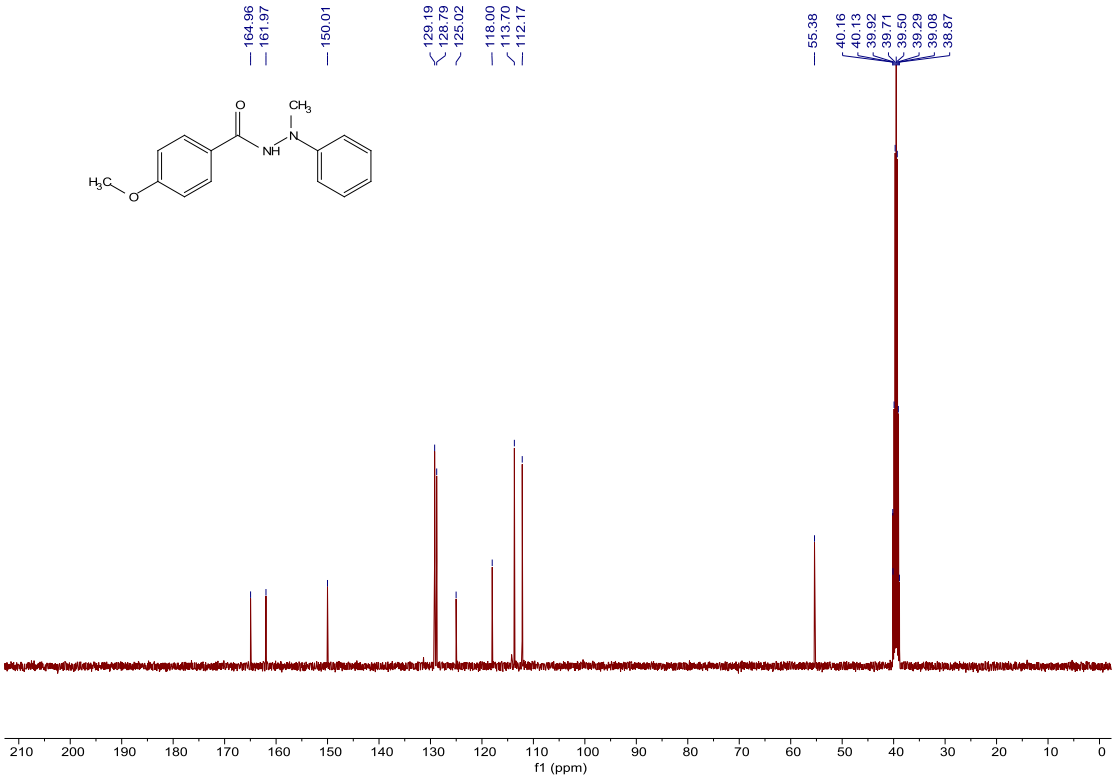
^{1}H NMR of 42 in DMSO-d_6 (400 MHz)



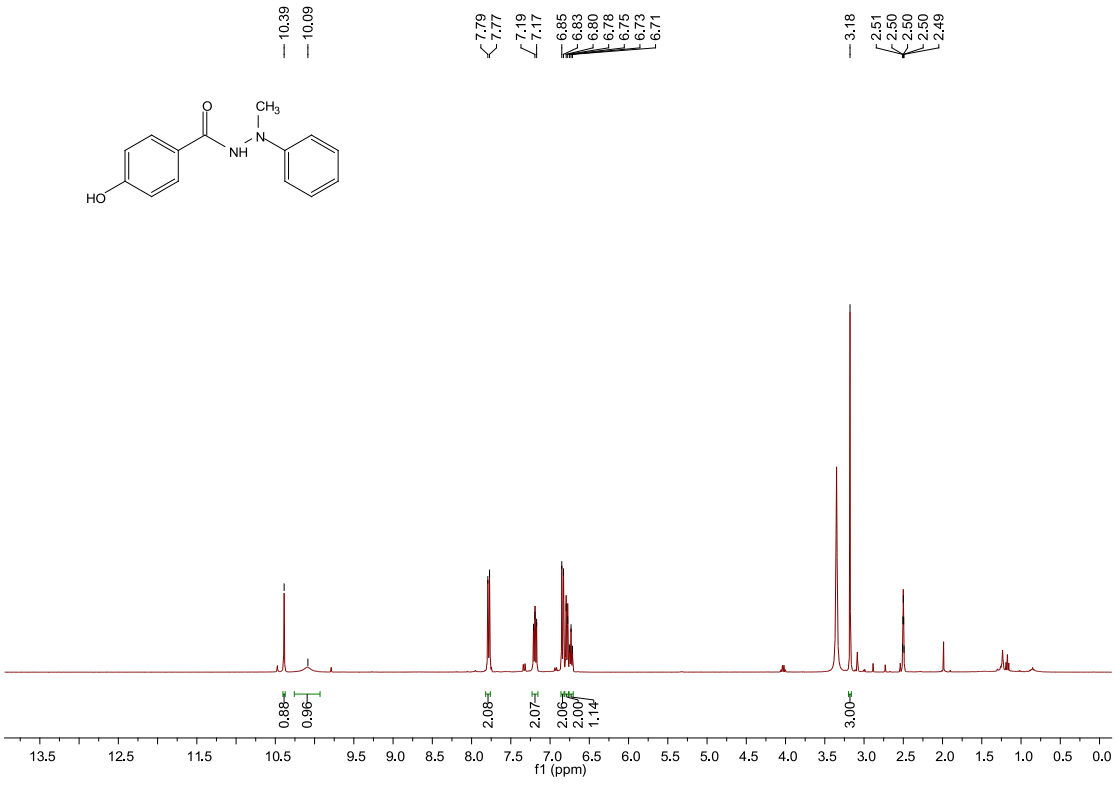
^{13}C{^{1}H} NMR of 42 in DMSO-d_6 (100 MHz)



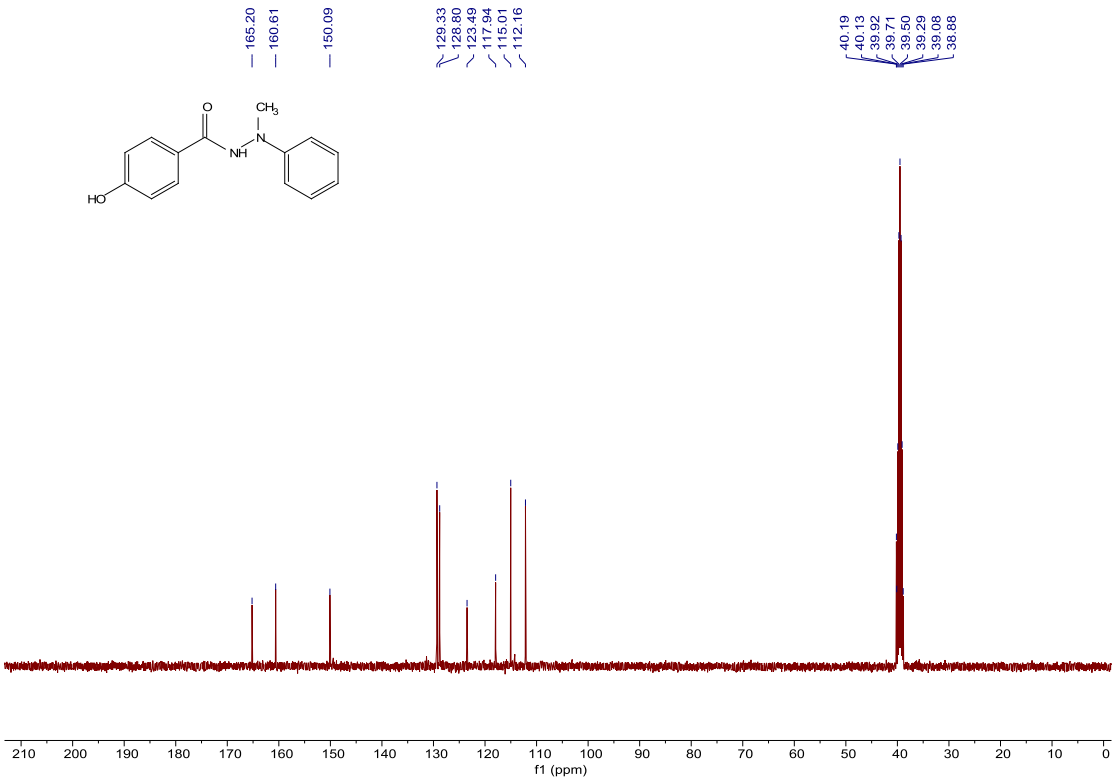
^{1}H NMR of 43 in DMSO-d_6 (400 MHz)



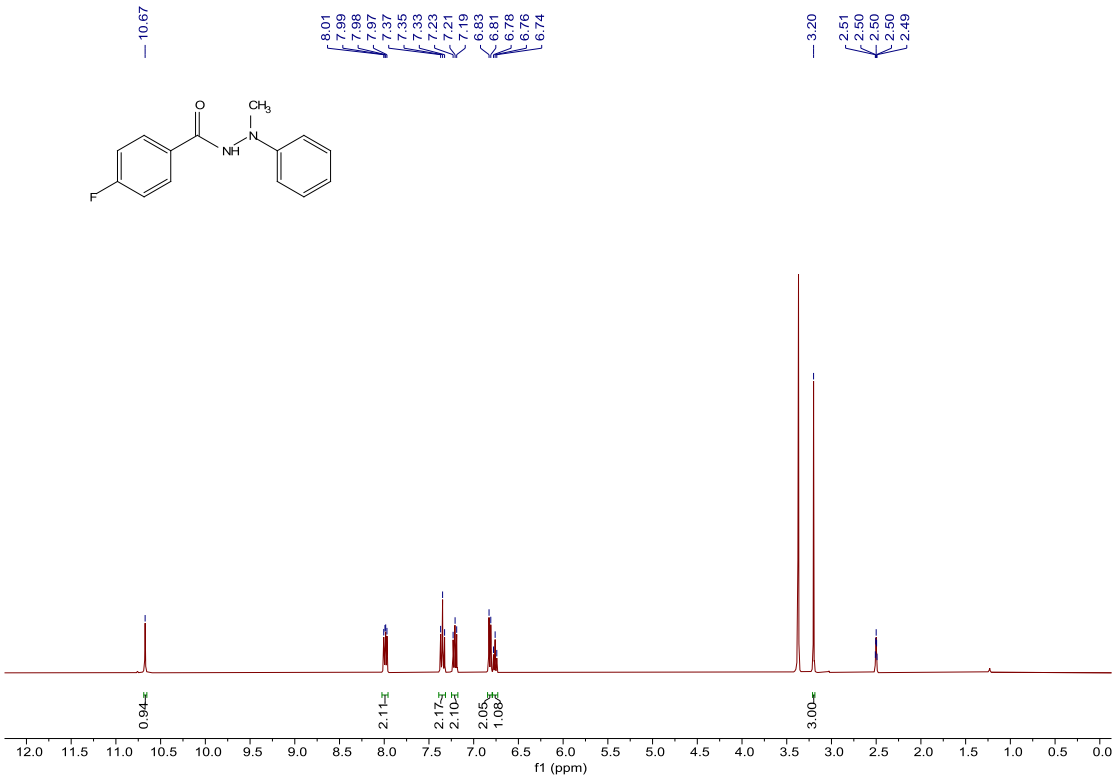
^{13}C{^{1}H} NMR of 43 in DMSO-d_6 (100 MHz)



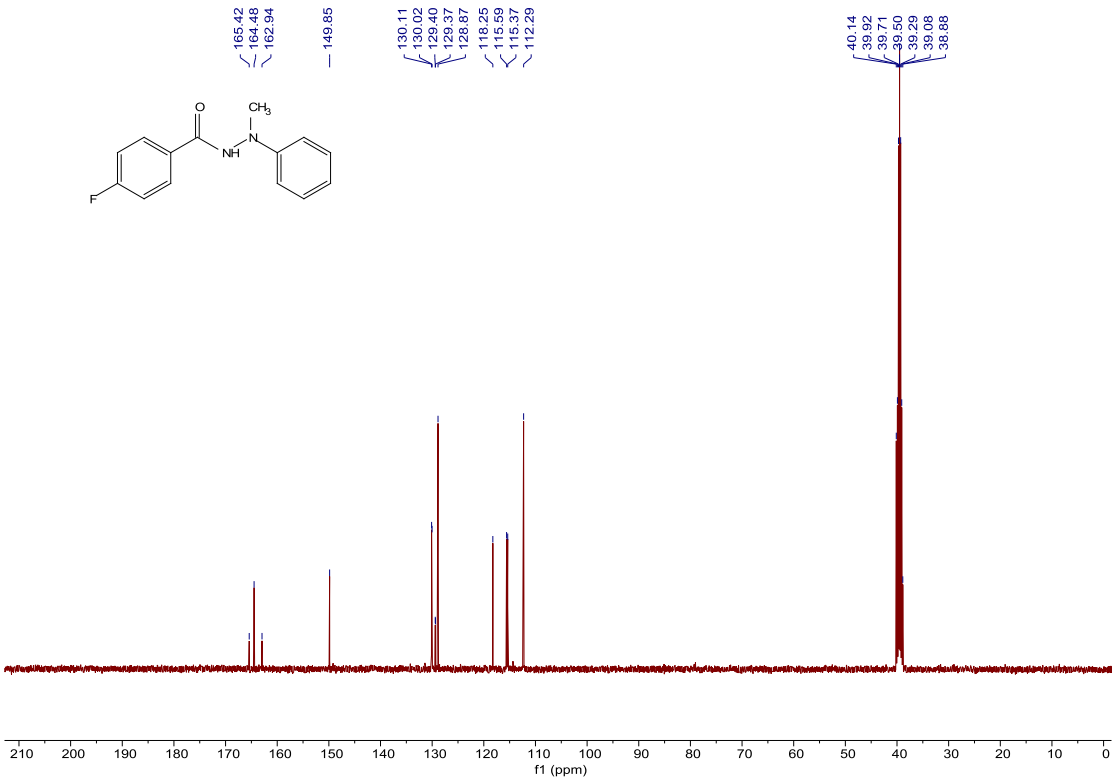
^{1}H NMR of 44 in DMSO-d_6 (400 MHz)



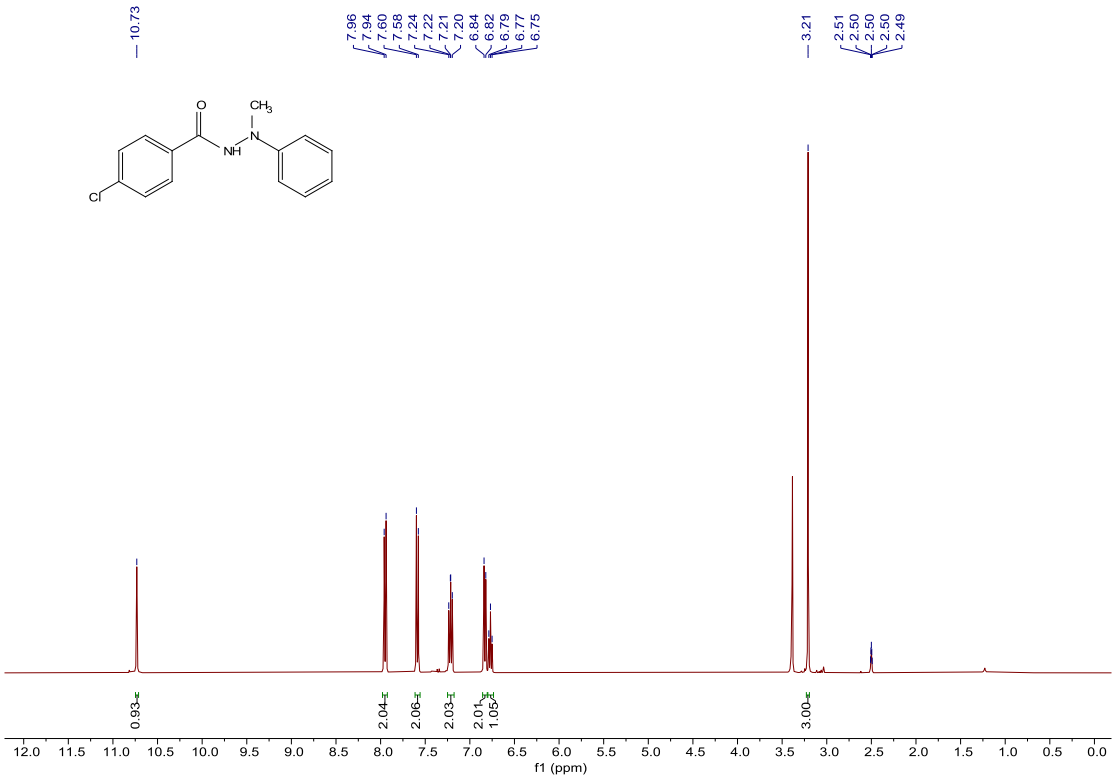
^{13}C{^{1}H} NMR of 44 in DMSO-d_6 (100 MHz)



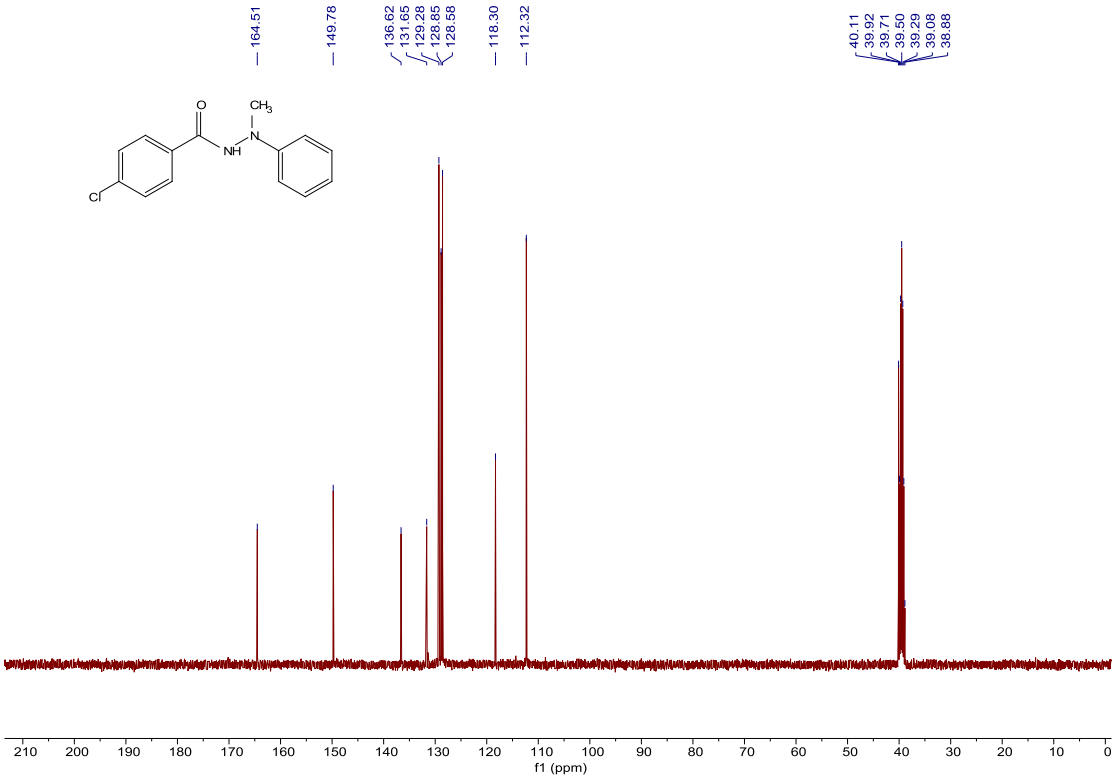
^{1}H NMR of 45 in DMSO-d_6 (400 MHz)



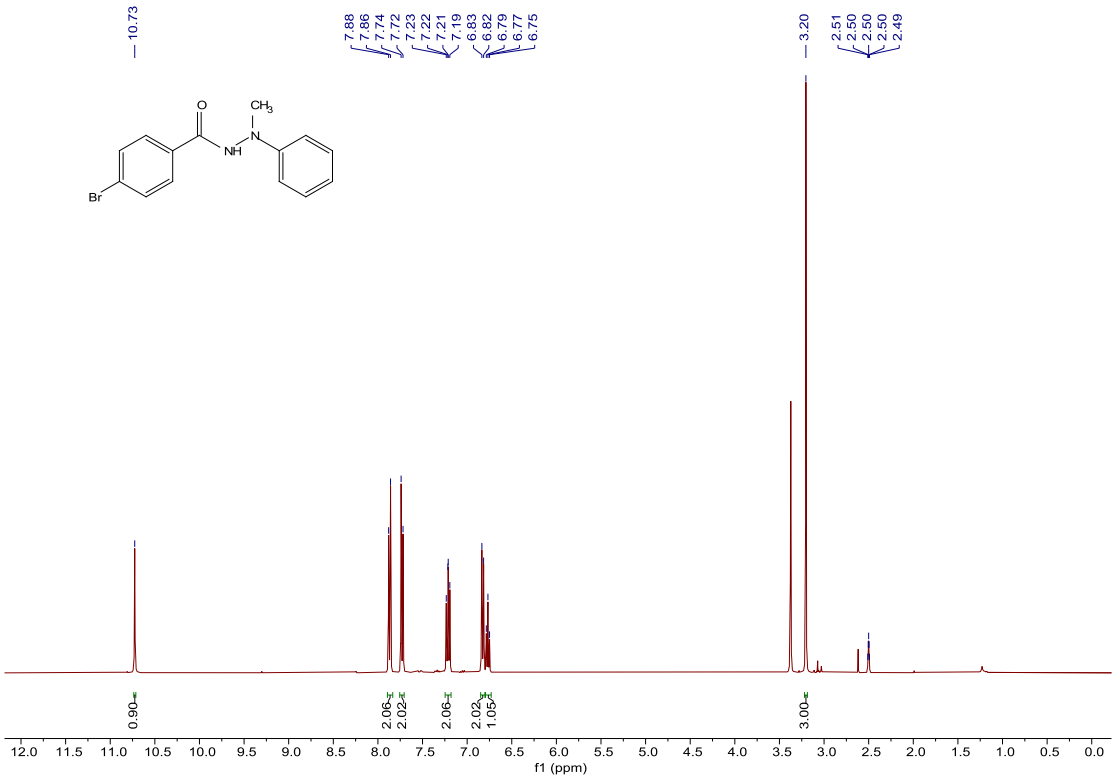
^{13}C{^{1}H} NMR of 45 in DMSO-d_6 (100 MHz)



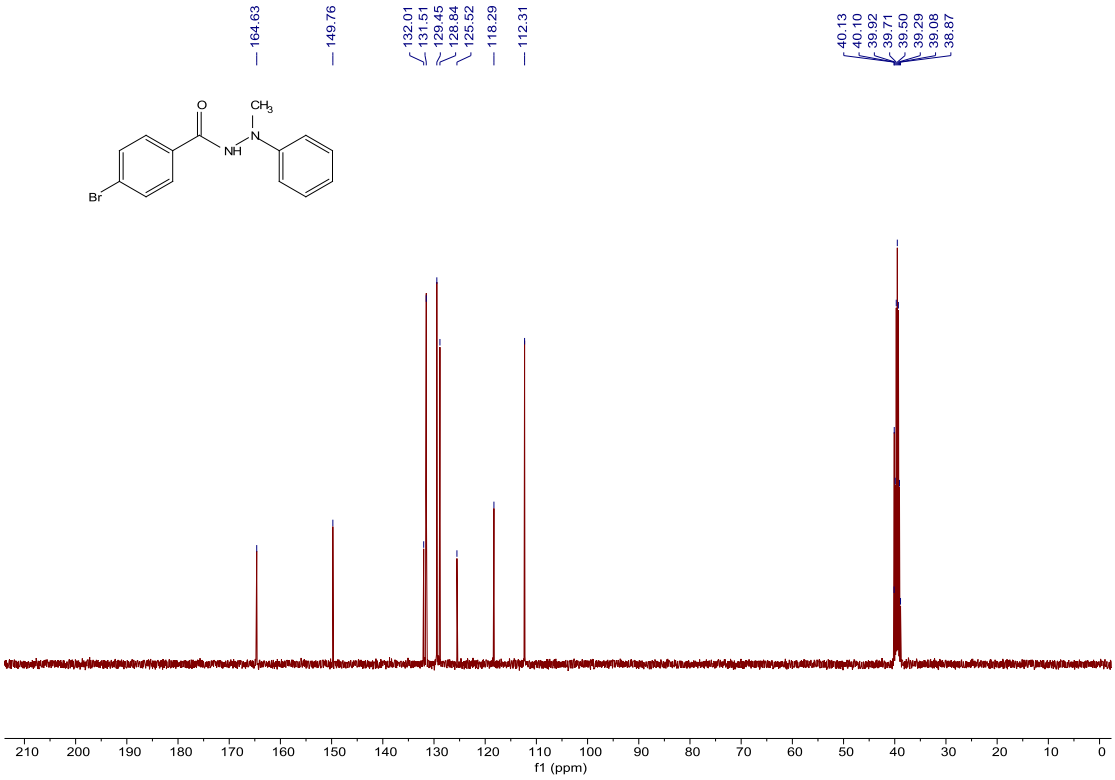
^{1}H NMR of 46 in DMSO-d_6 (400 MHz)



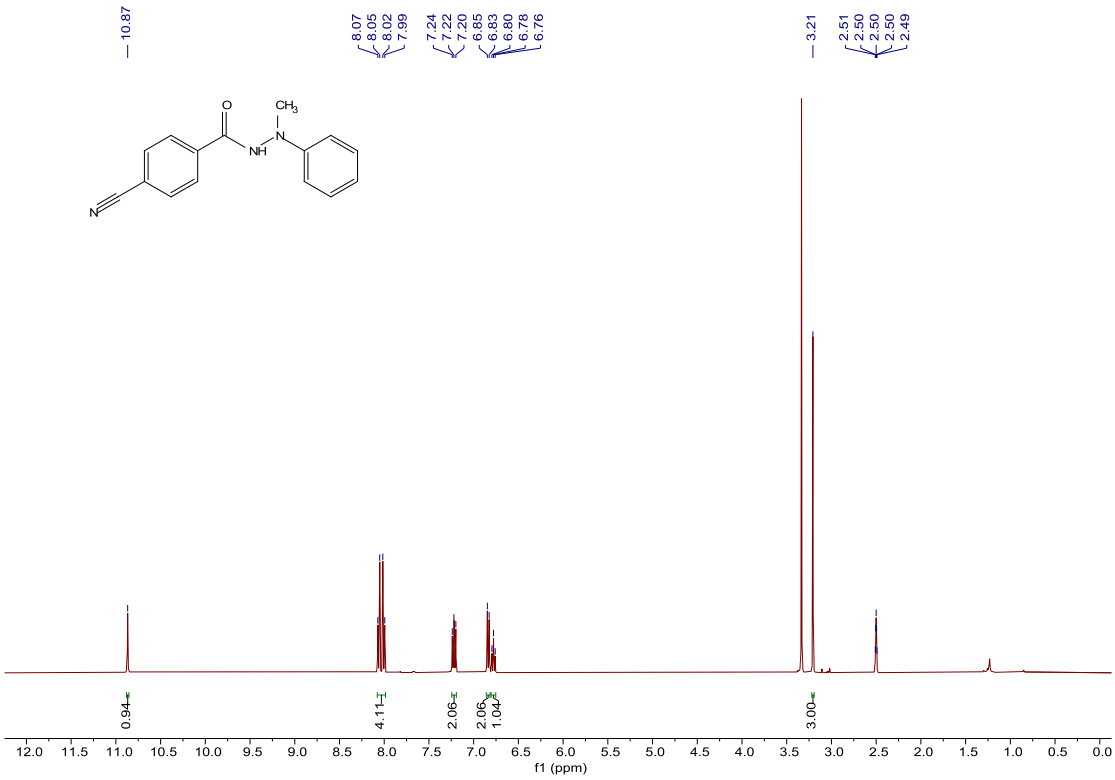
^{13}C{^{1}H} NMR of 46 in DMSO-d_6 (100 MHz)



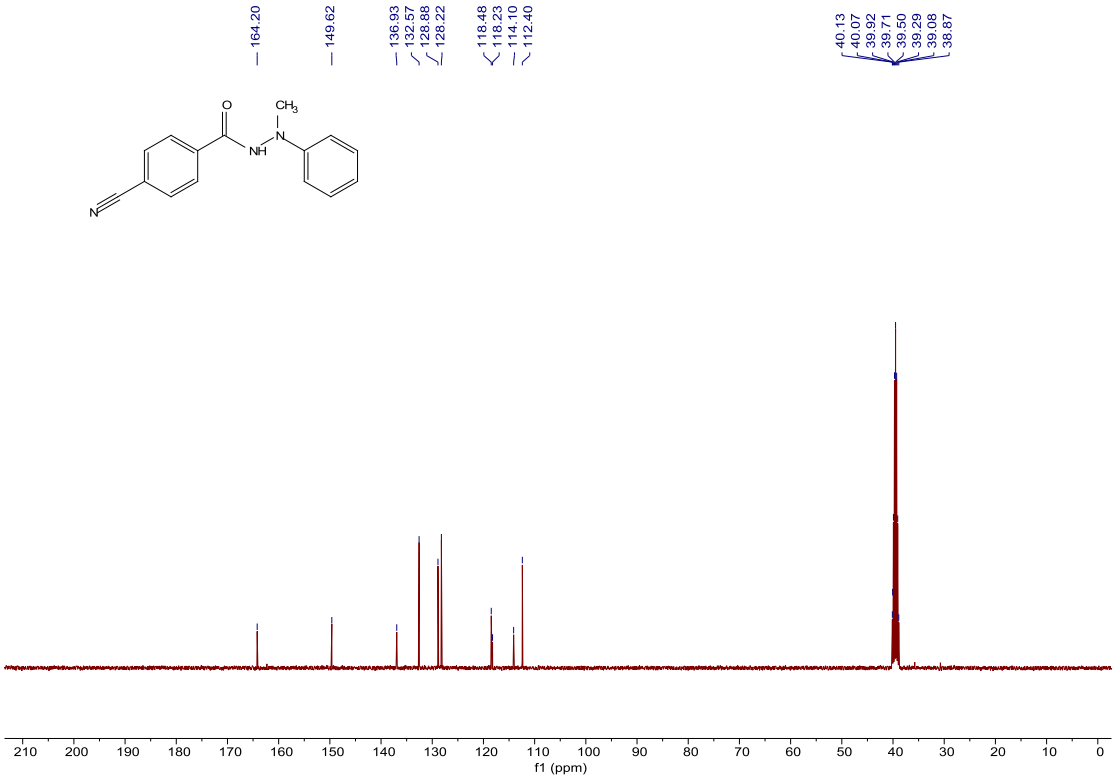
^{1}H NMR of 47 in DMSO-d_6 (400 MHz)



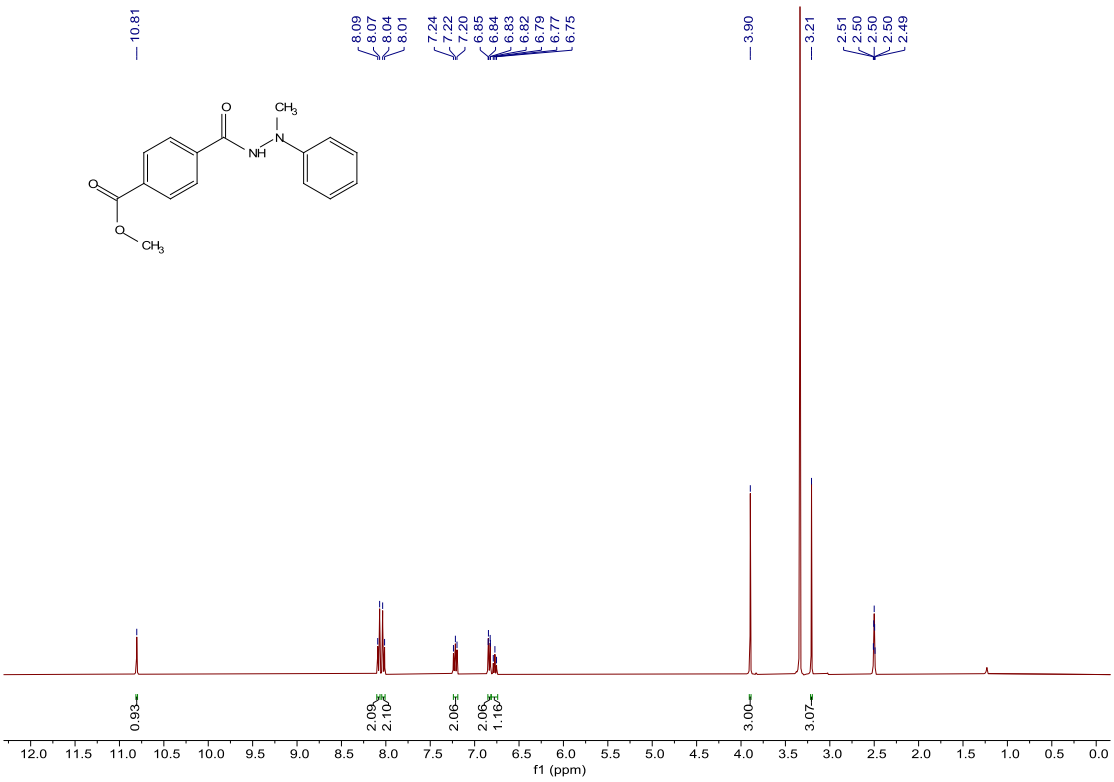
^{13}C{^{1}H} NMR of 47 in DMSO-d_6 (100 MHz)



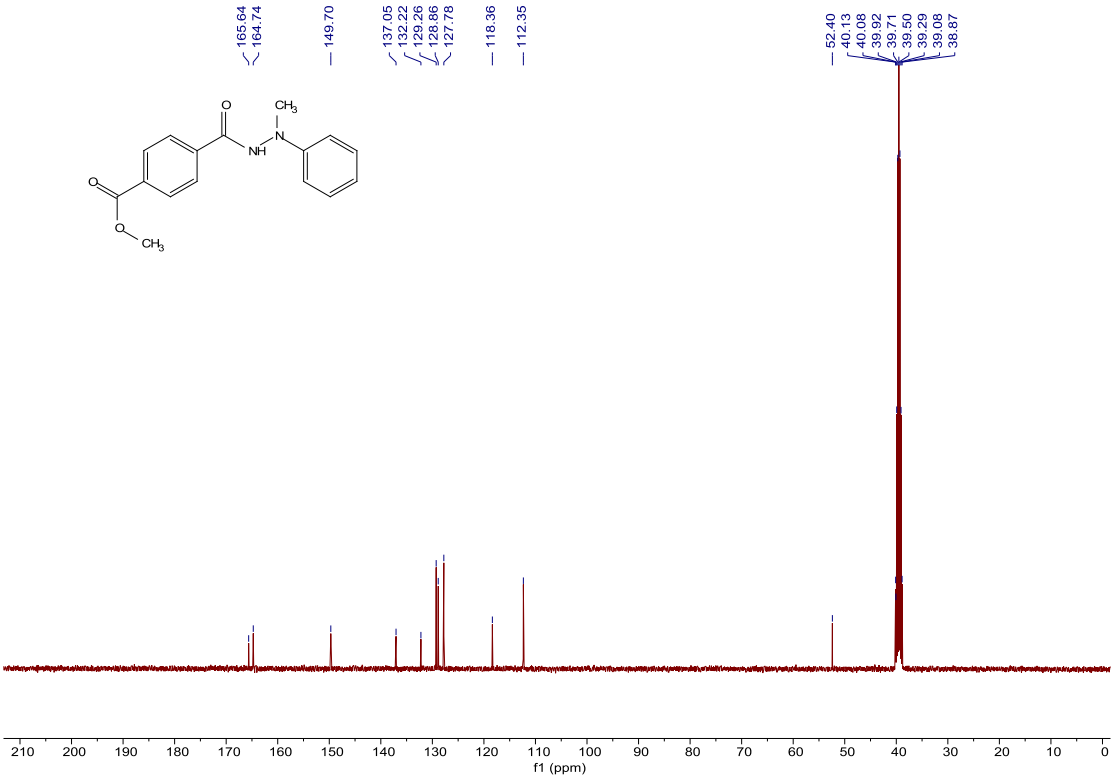
^{1}H NMR of 48 in DMSO-d_6 (400 MHz)



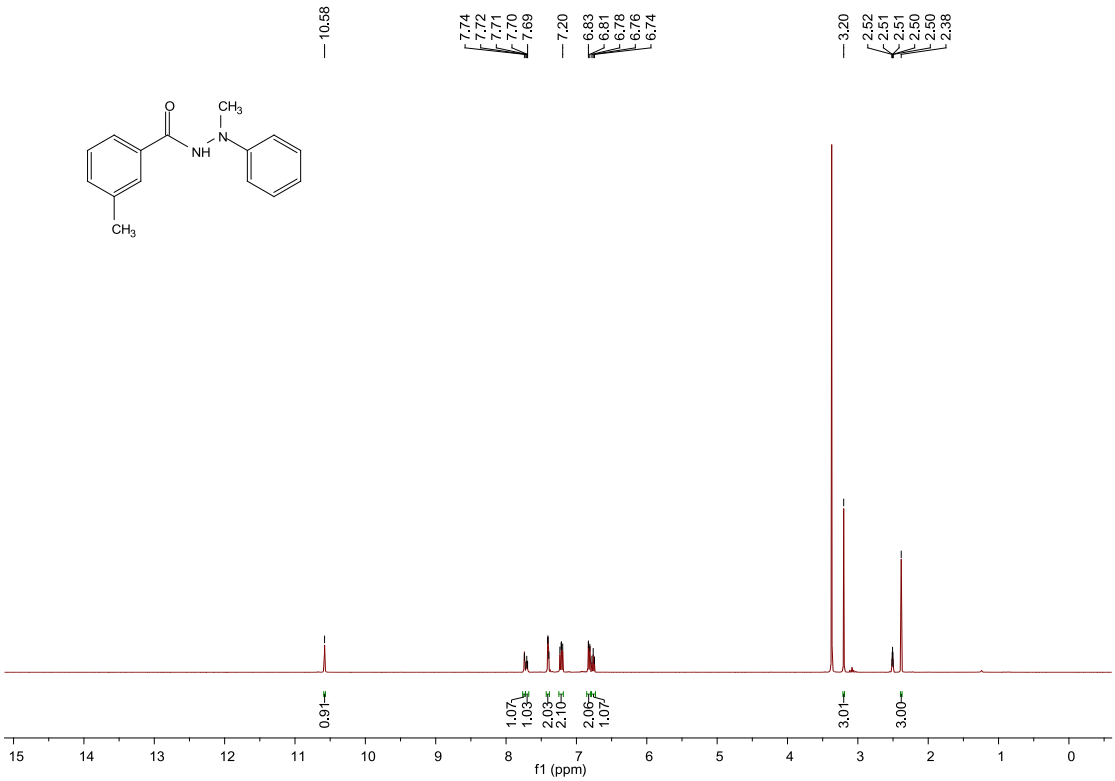
^{13}C{^{1}H} NMR of 48 in DMSO-d_6 (100 MHz)



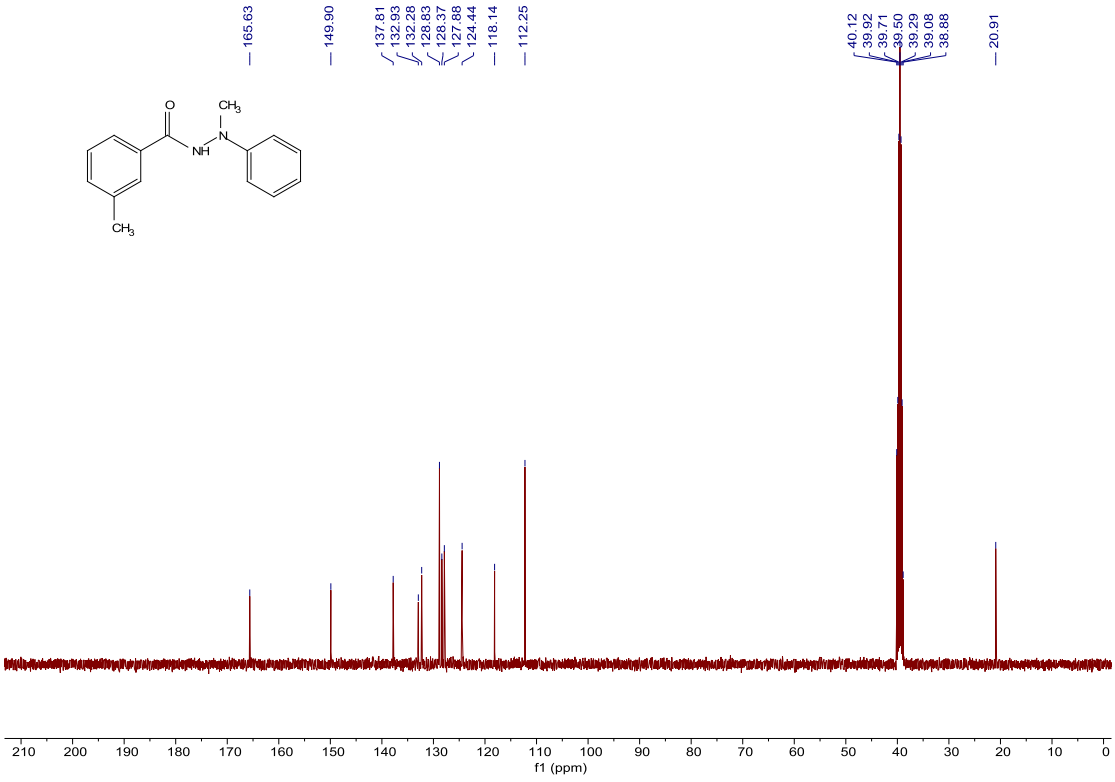
^{1}H NMR of 49 in DMSO-d_6 (400 MHz)



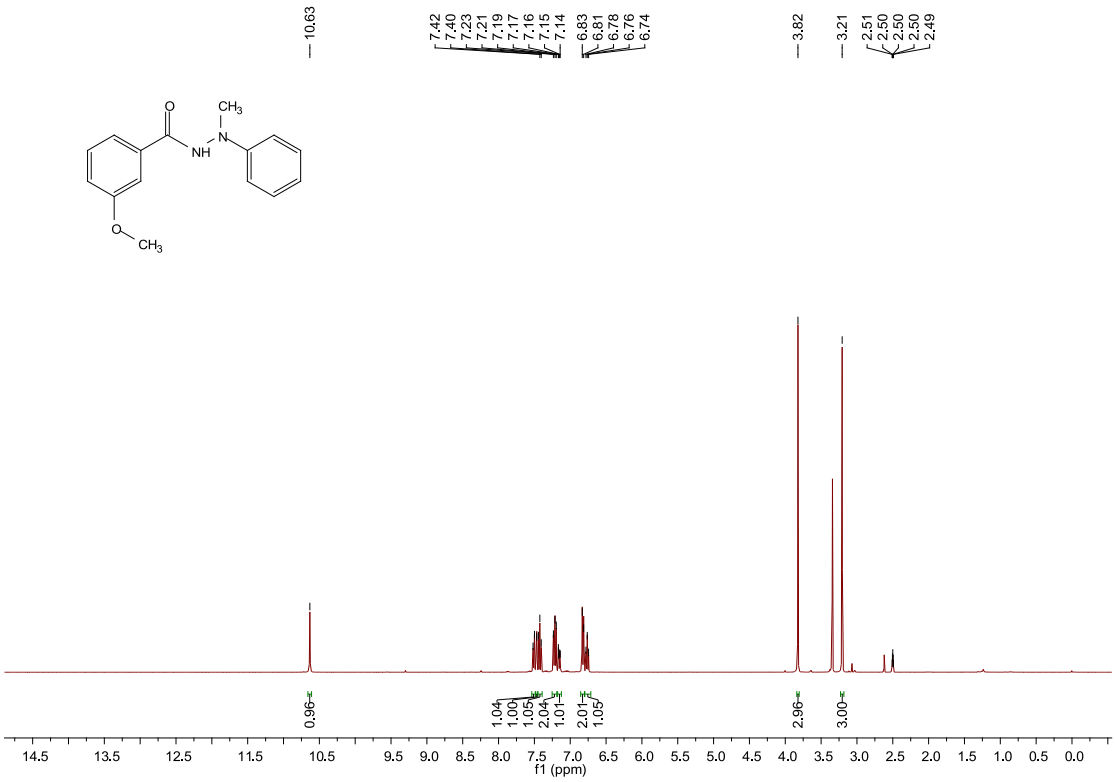
^{13}C{^{1}H} NMR of 49 in DMSO-d_6 (100 MHz)



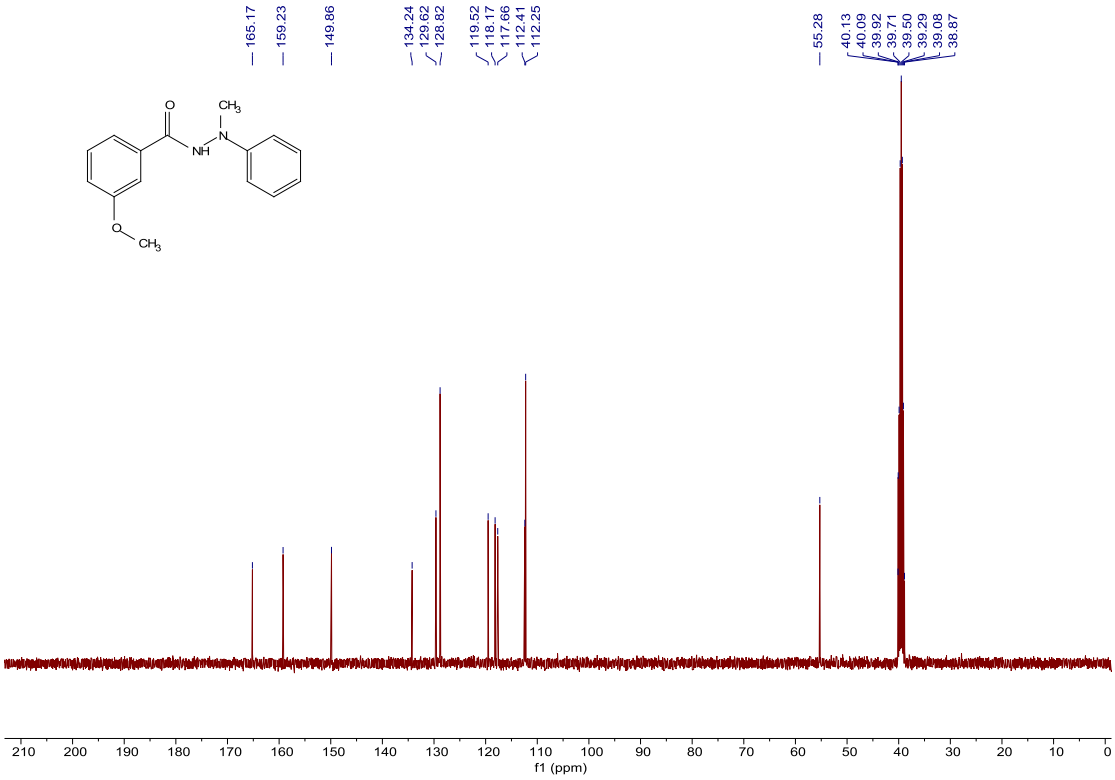
^{1}H NMR of 50 in DMSO-d_6 (400 MHz)



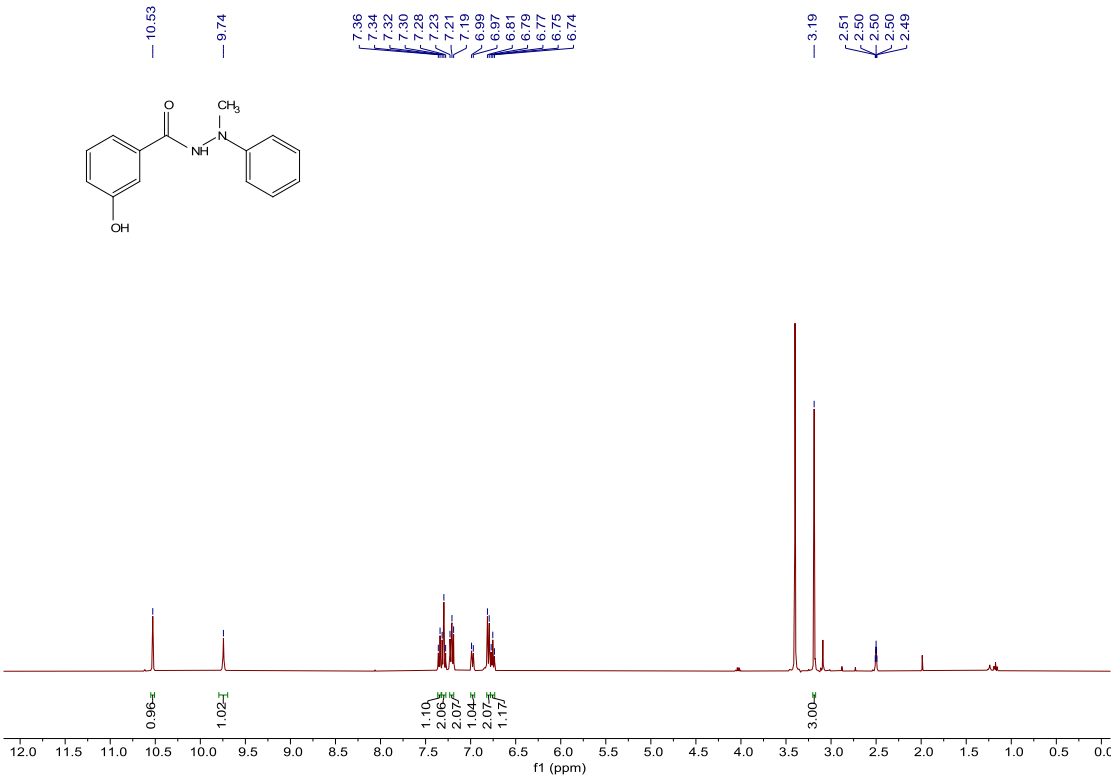
^{13}C{^{1}H} NMR of 50 in DMSO-d_6 (100 MHz)



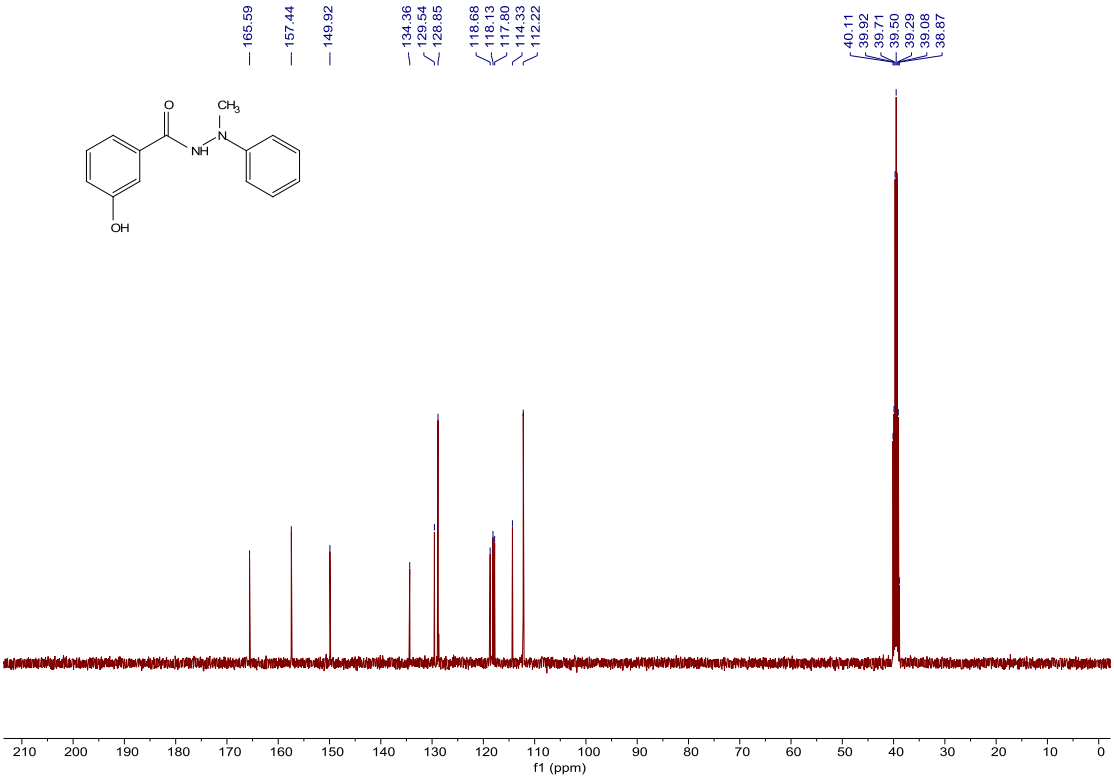
^{1}H NMR of 51 in DMSO-d_6 (400 MHz)



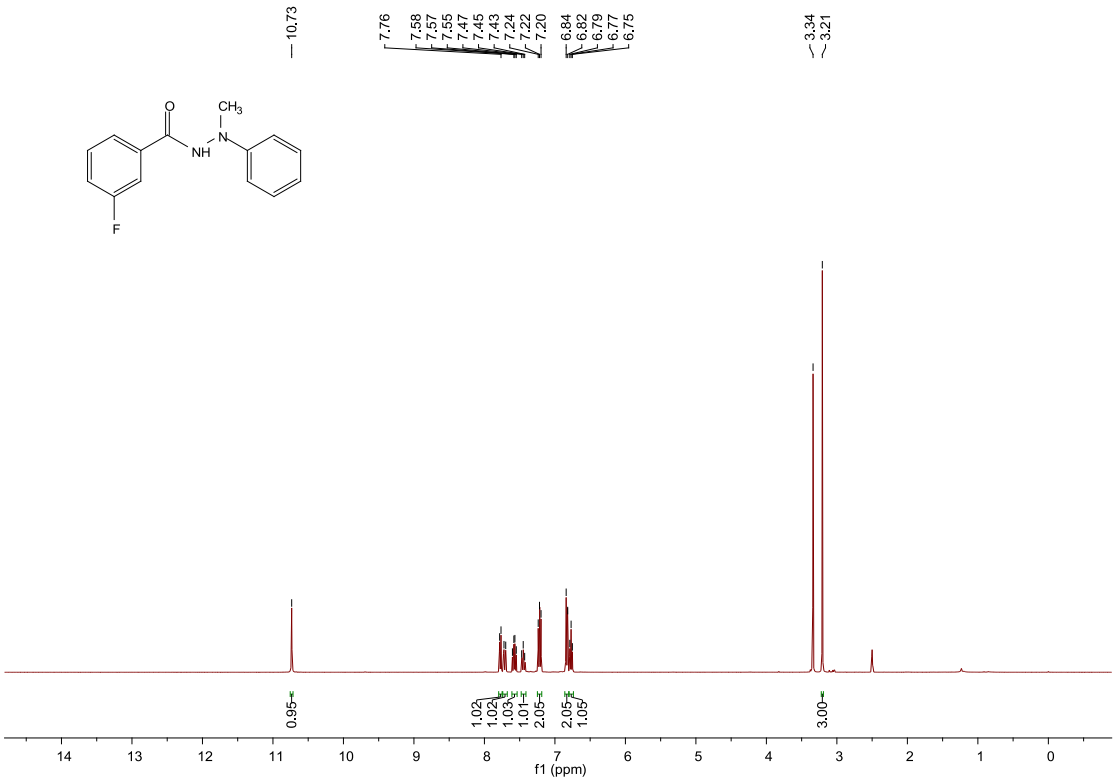
^{13}C{^{1}H} NMR of 51 in DMSO-d_6 (100 MHz)



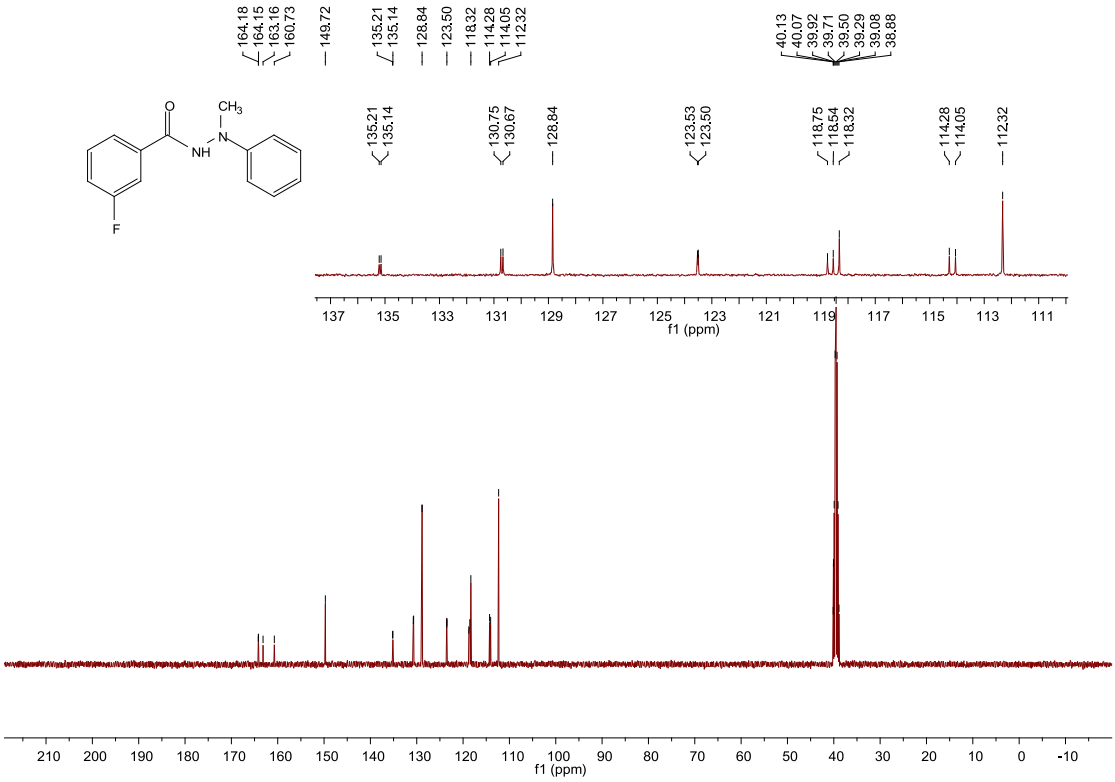
^{1}H NMR of 52 in DMSO-d_6 (400 MHz)



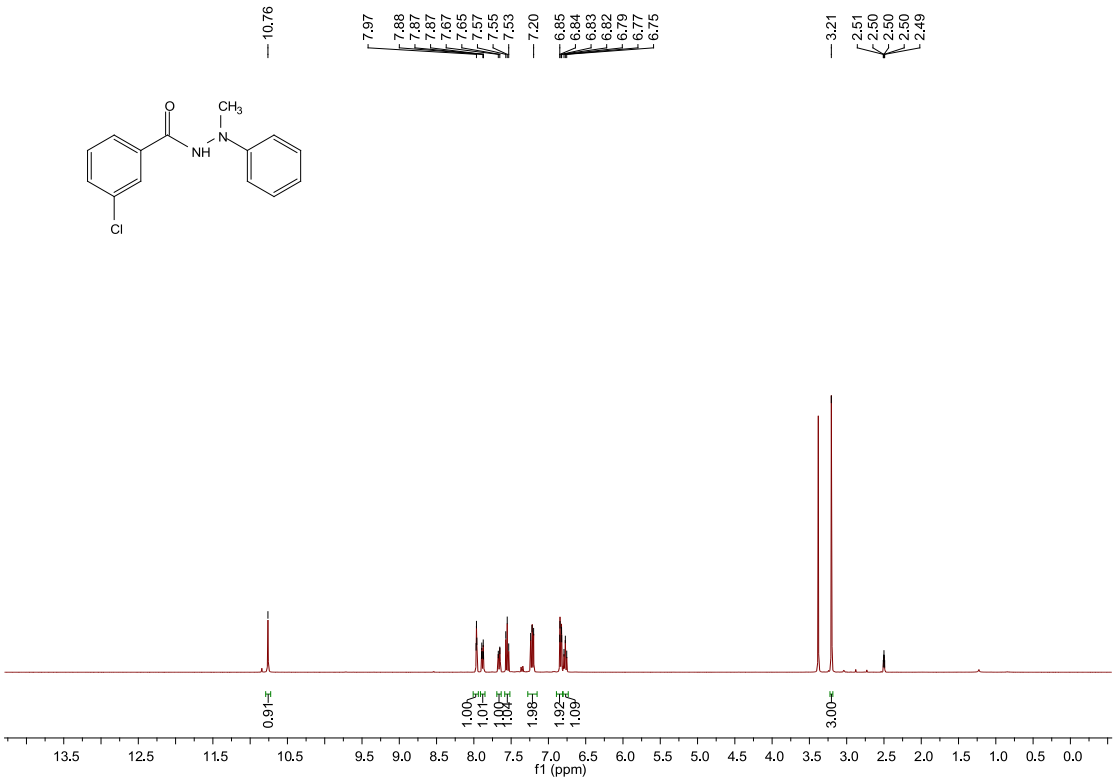
^{13}C{^{1}H} NMR of 52 in DMSO-d_6 (100 MHz)



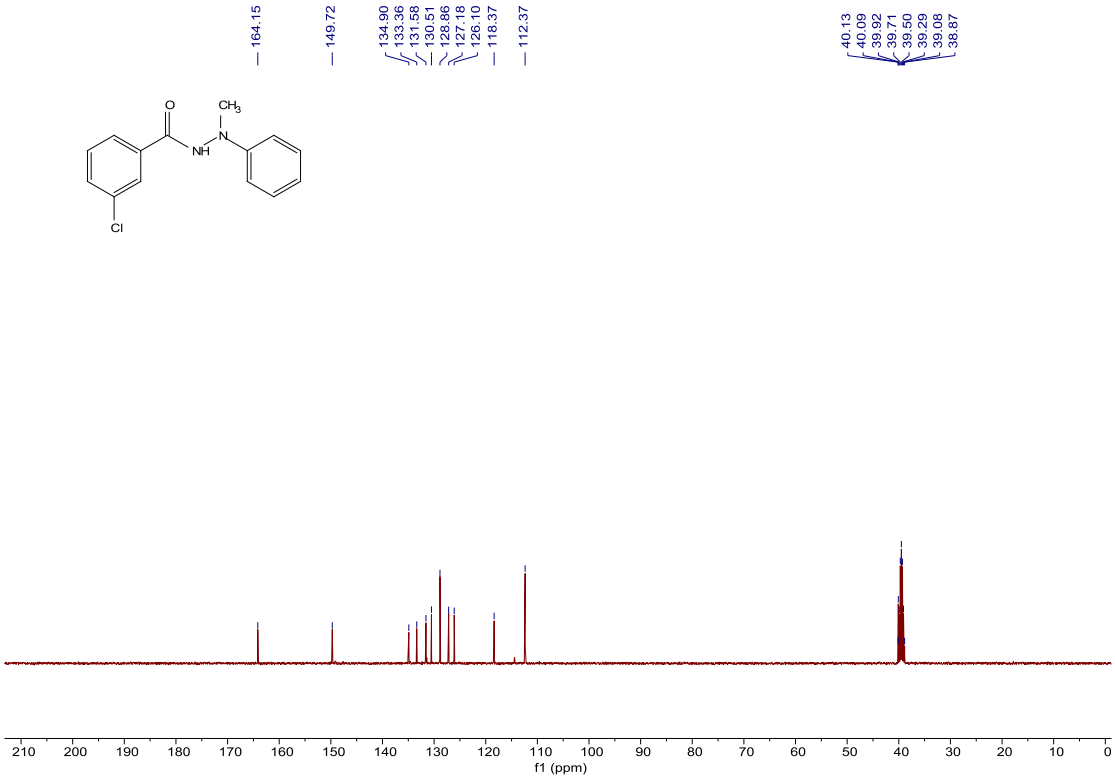
^{1}H NMR of 53 in DMSO-d_6 (400 MHz)



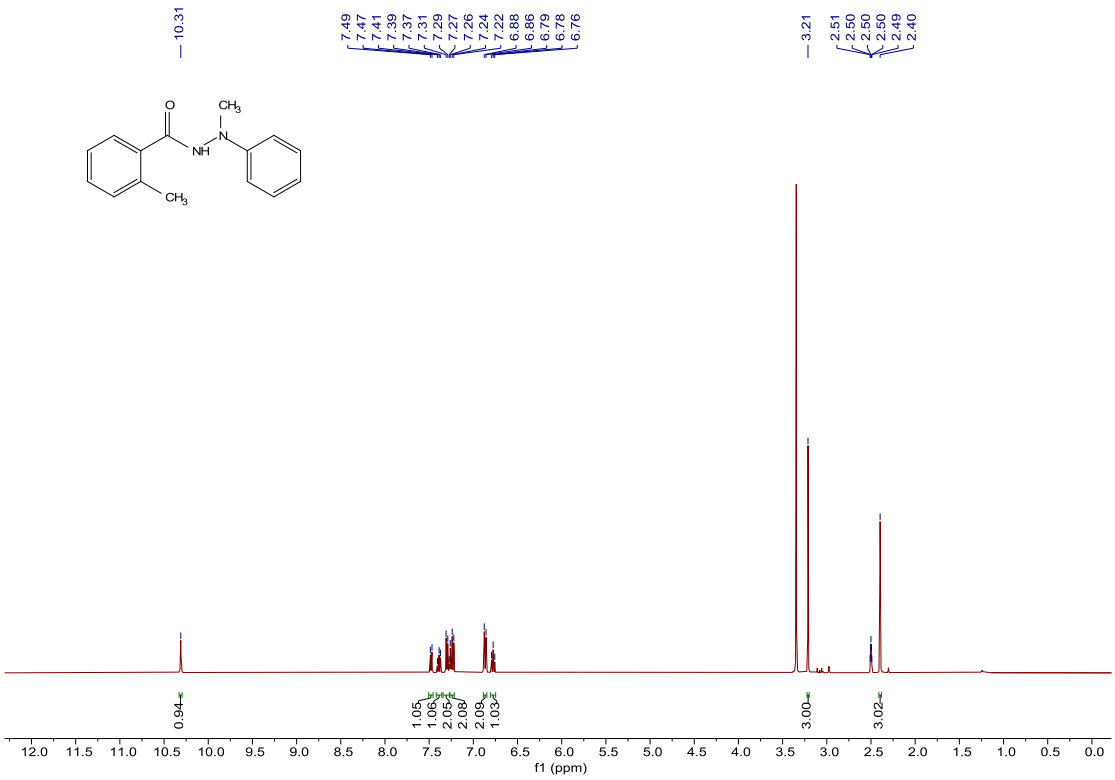
^{13}C{^{1}H} NMR of 53 in DMSO-d_6 (100 MHz)



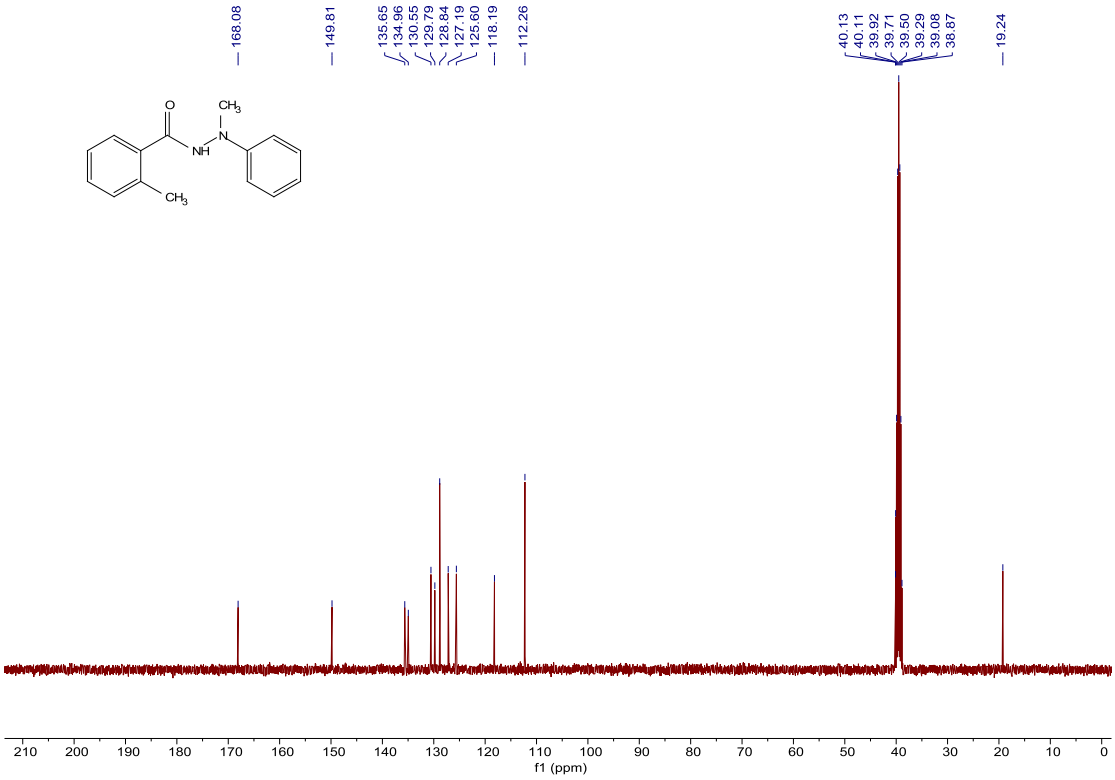
^{1}H NMR of 54 in DMSO-d_6 (400 MHz)



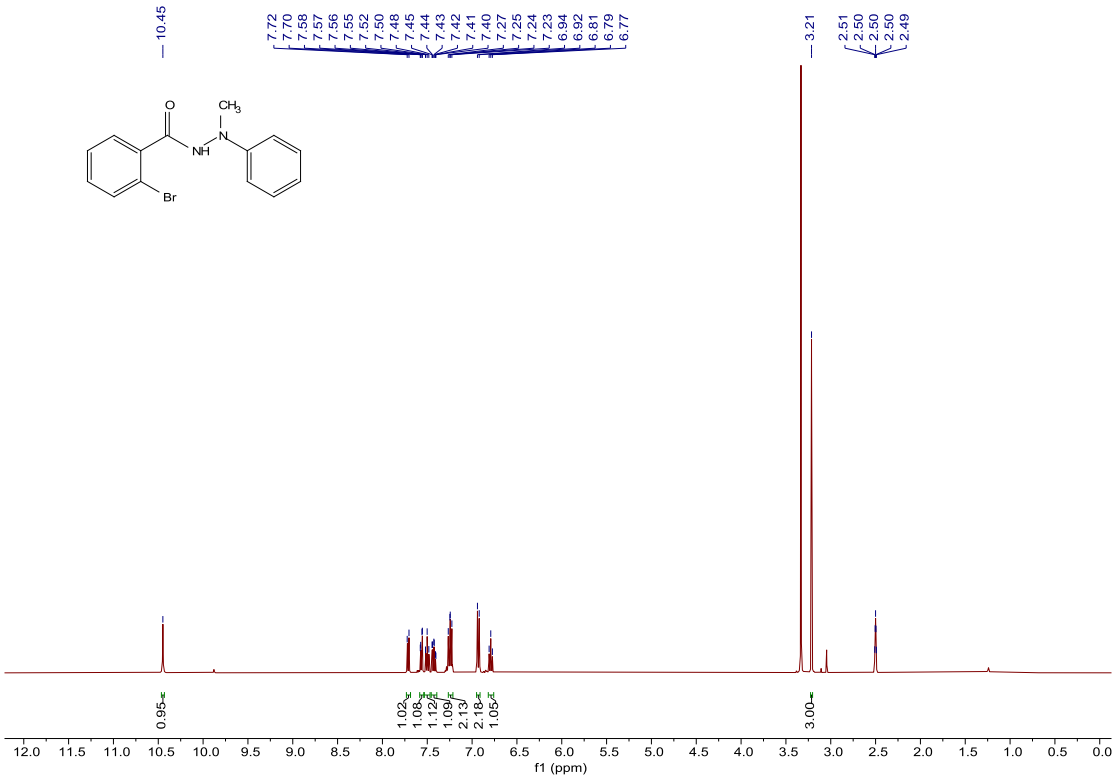
^{13}C{^{1}H} NMR of 54 in DMSO-d_6 (100 MHz)



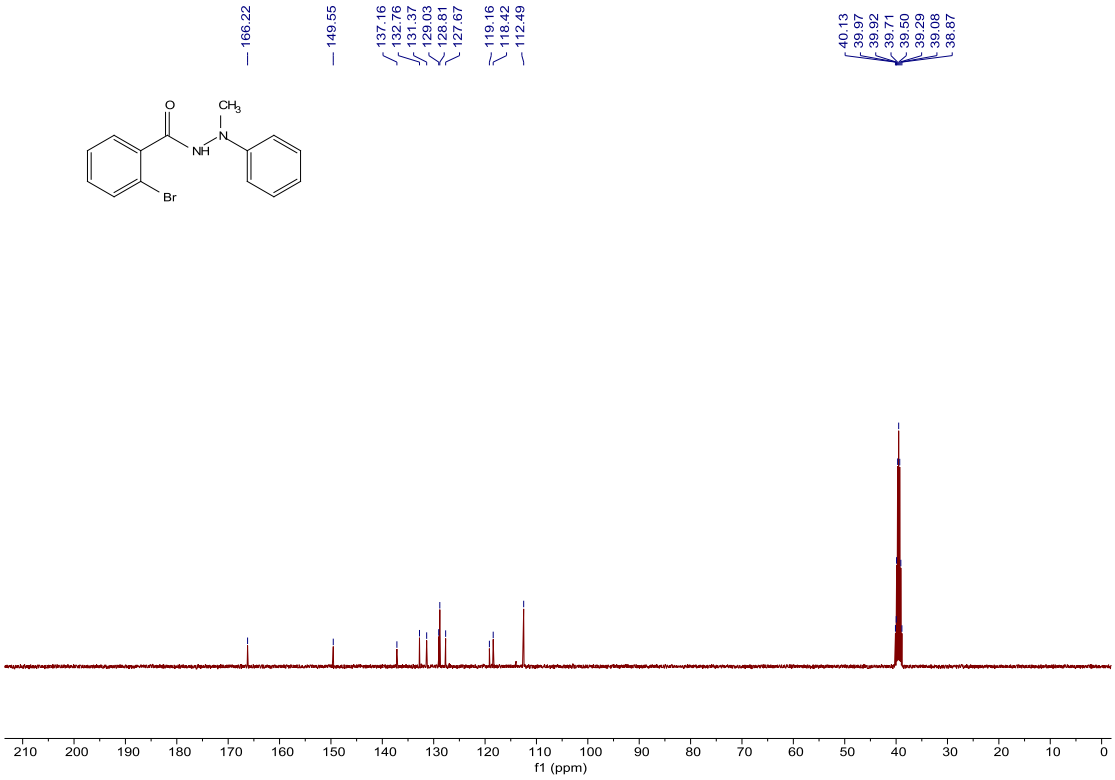
^{1}H NMR of 55 in DMSO-d_6 (400 MHz)



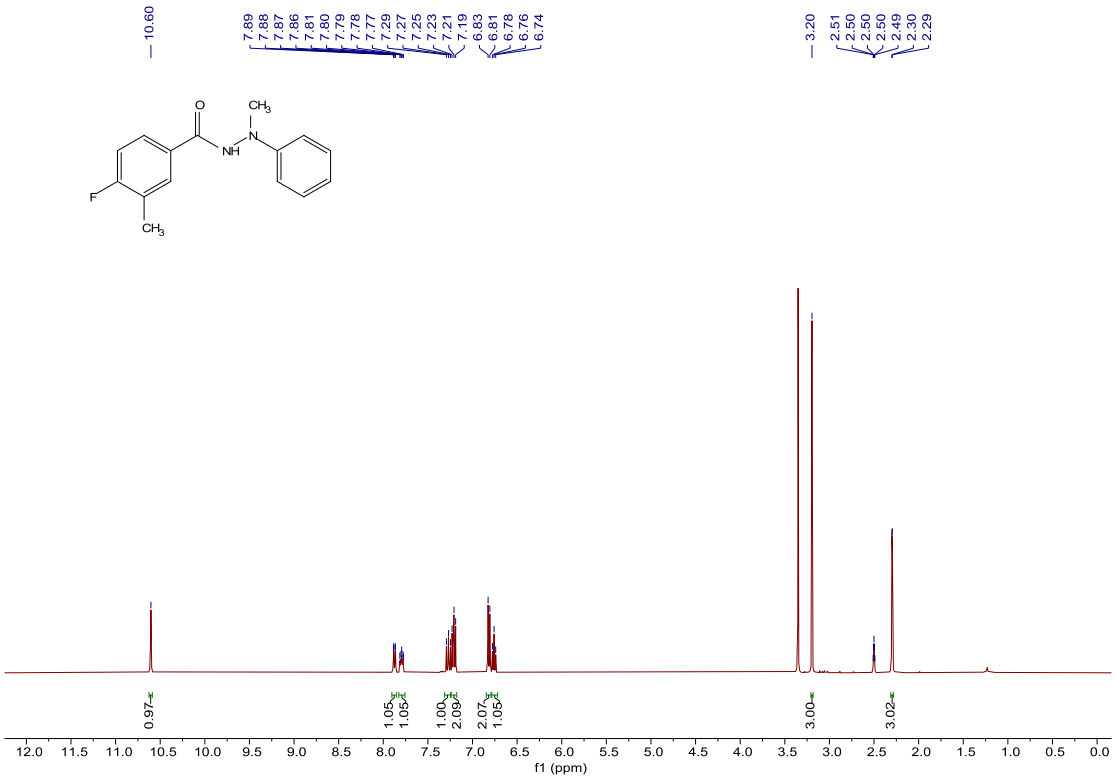
^{13}C{^{1}H} NMR of 55 in DMSO-d_6 (100 MHz)



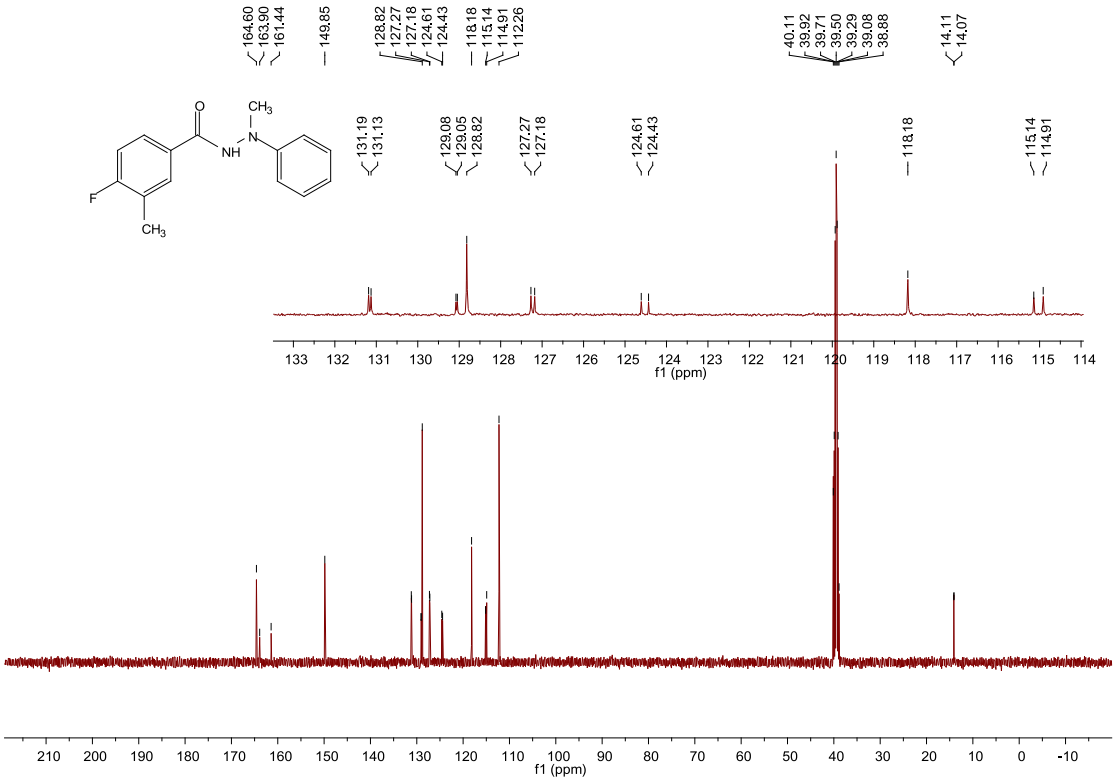
^{1}H NMR of 56 in DMSO-d_6 (400 MHz)



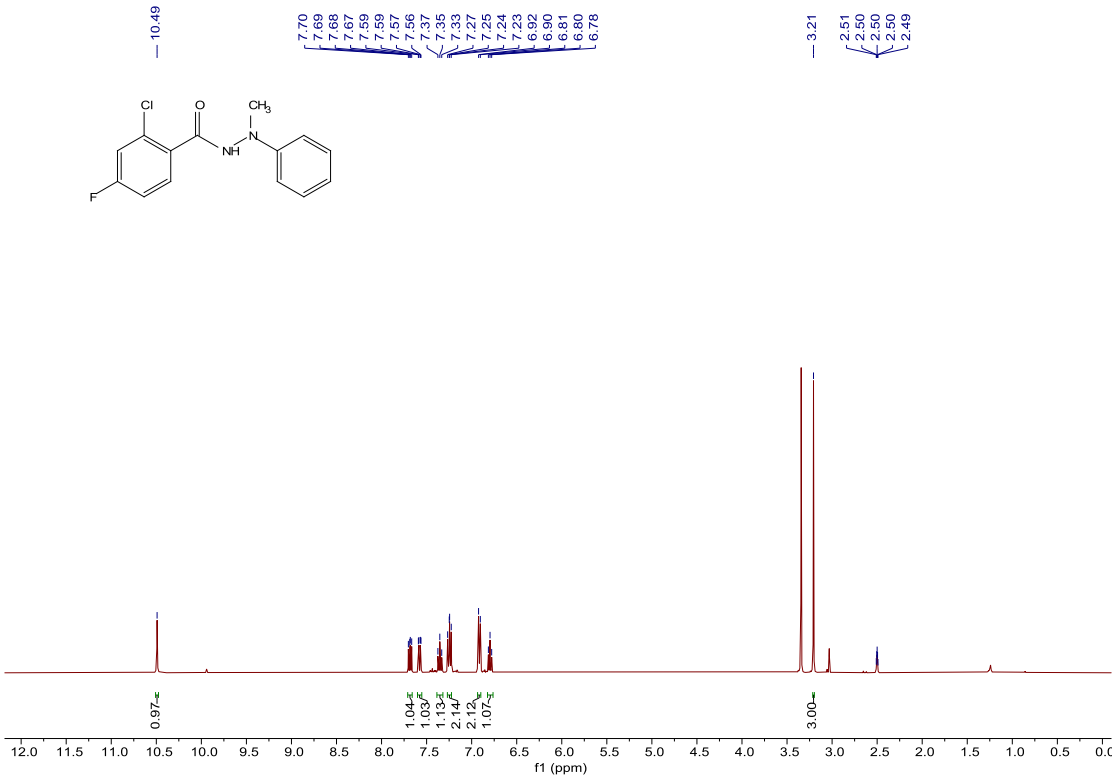
^{13}C{^{1}H} NMR of 56 in DMSO-d_6 (100 MHz)



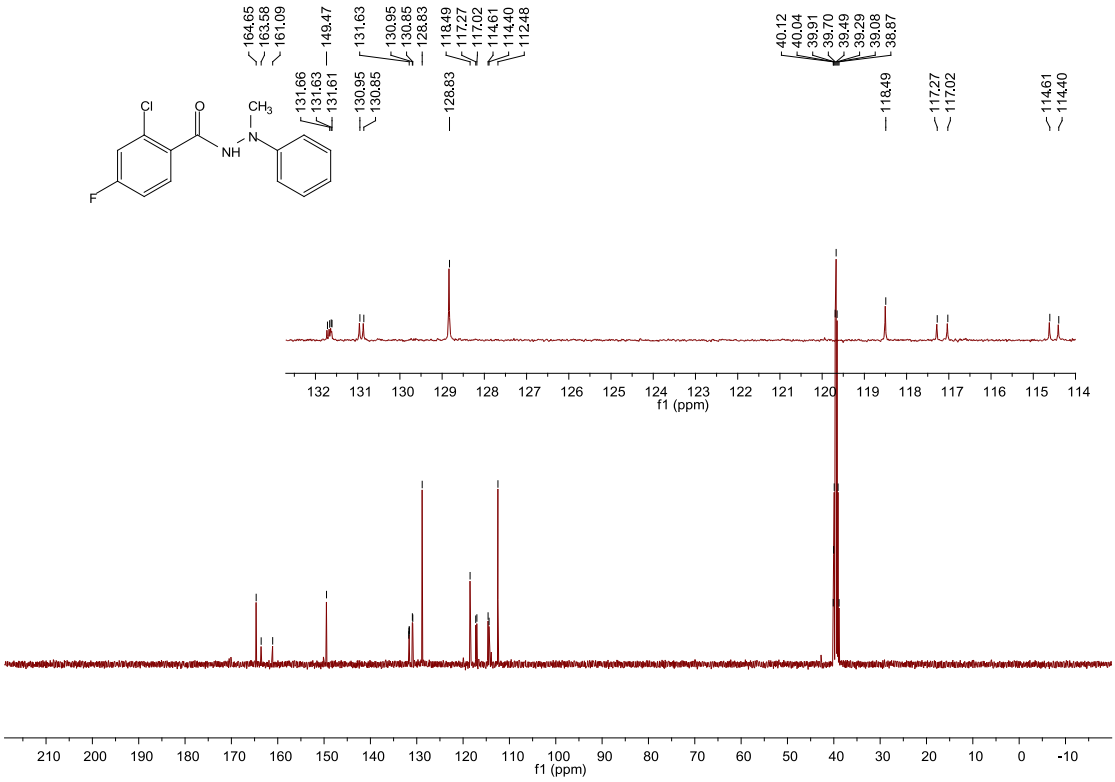
^{1}H NMR of 57 in DMSO-d_6 (400 MHz)



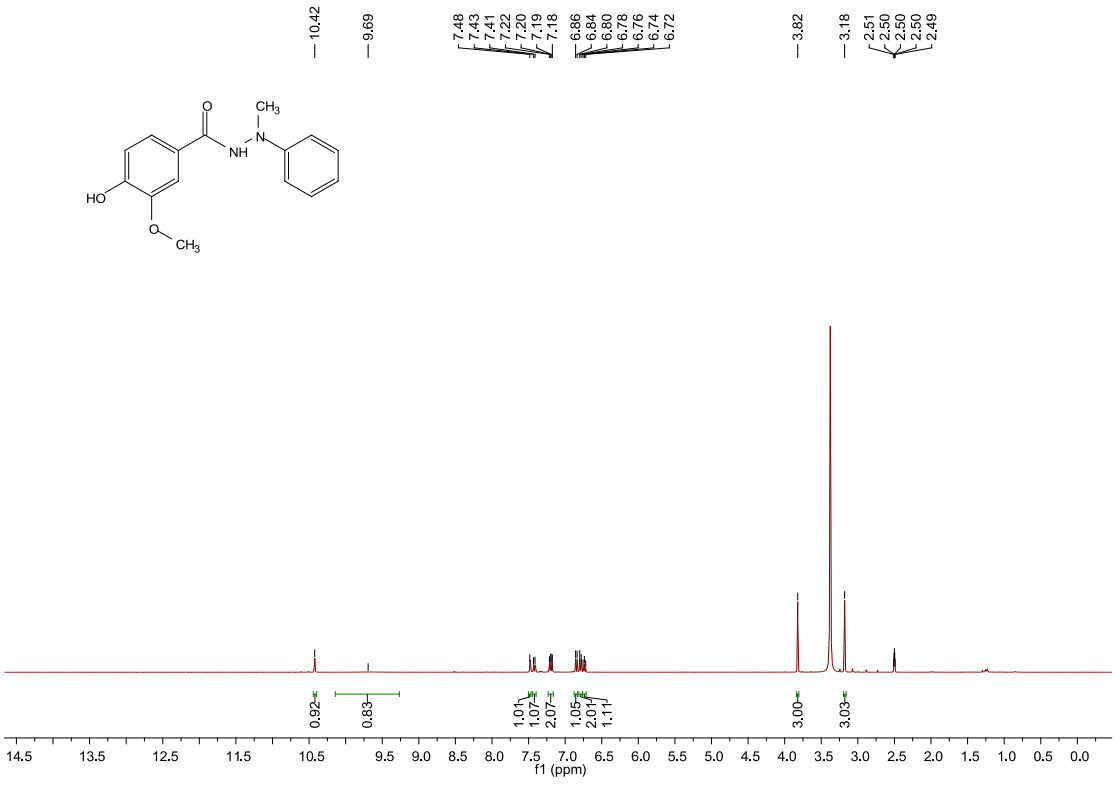
^{13}C{^{1}H} NMR of 57 in DMSO-d_6 (100 MHz)



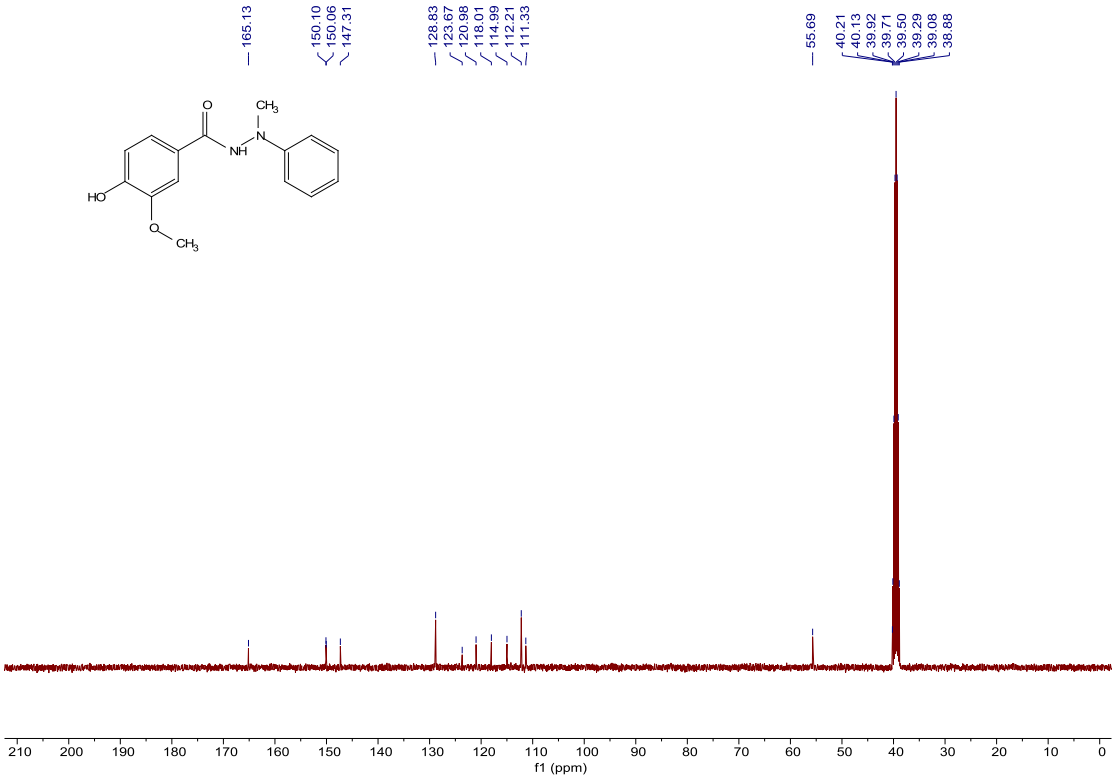
^{1}H NMR of 58 in DMSO-d_6 (400 MHz)



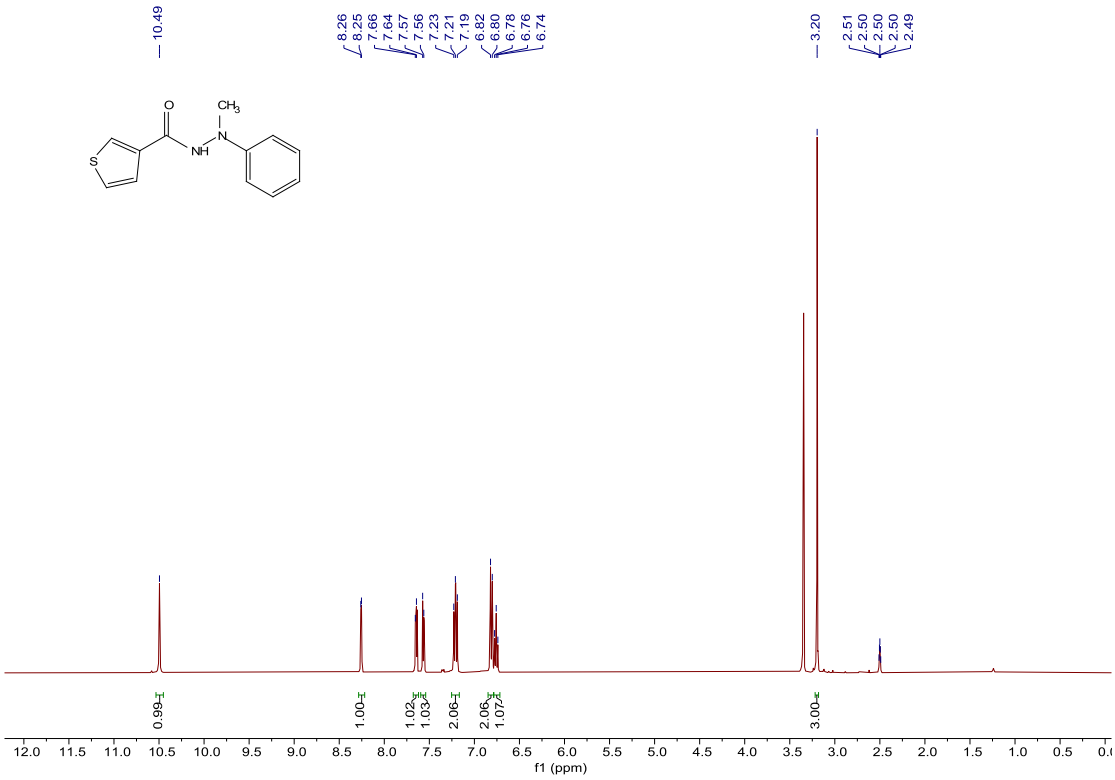
^{13}C{^{1}H} NMR of 58 in DMSO-d_6 (100 MHz)



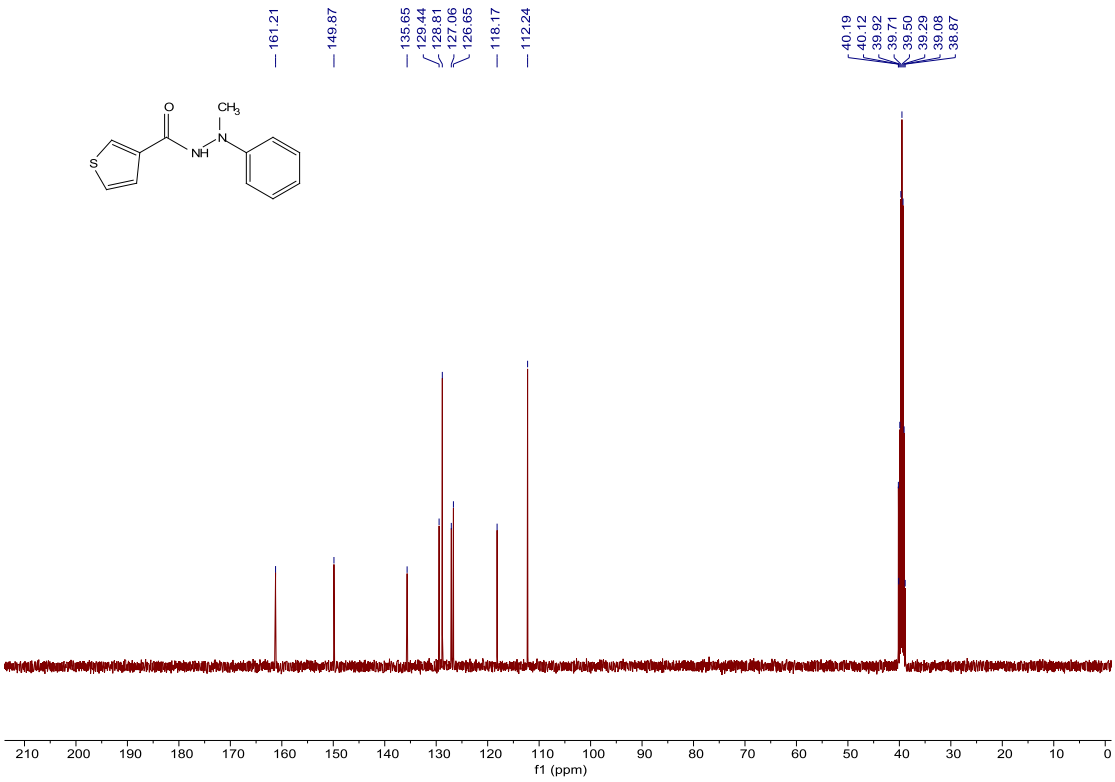
^{1}H NMR of 59 in DMSO-d_6 (400 MHz)



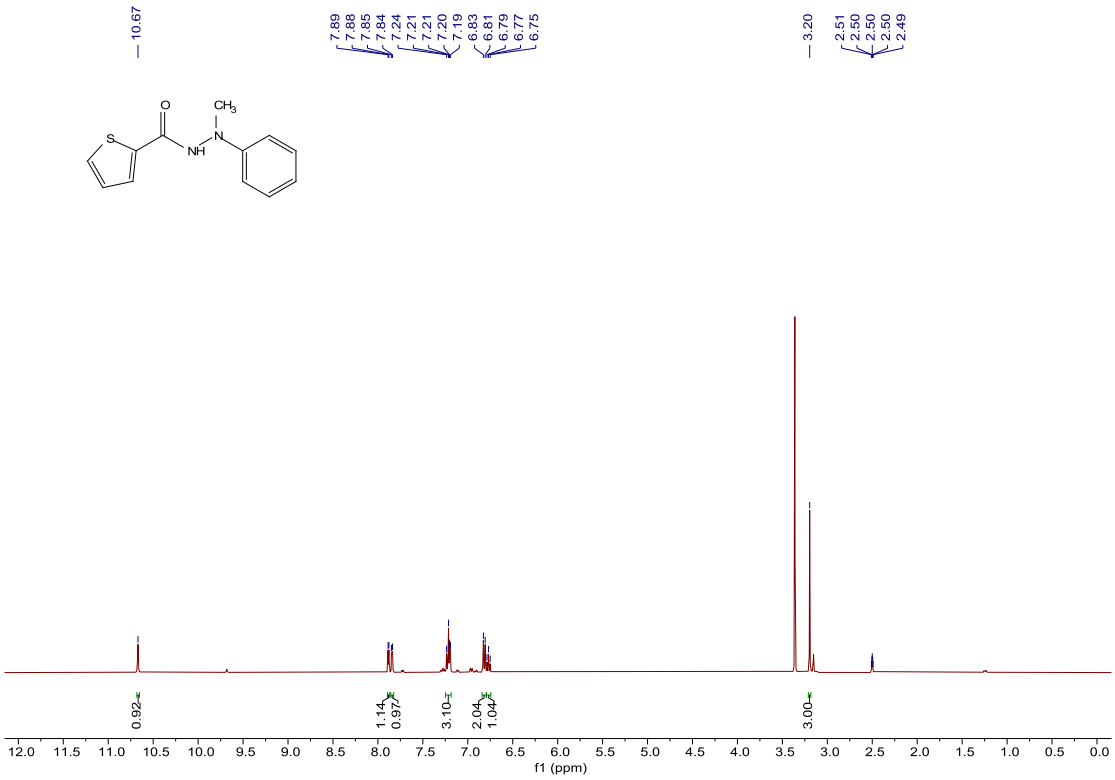
^{13}C{^{1}H} NMR of 59 in DMSO-d_6 (100 MHz)



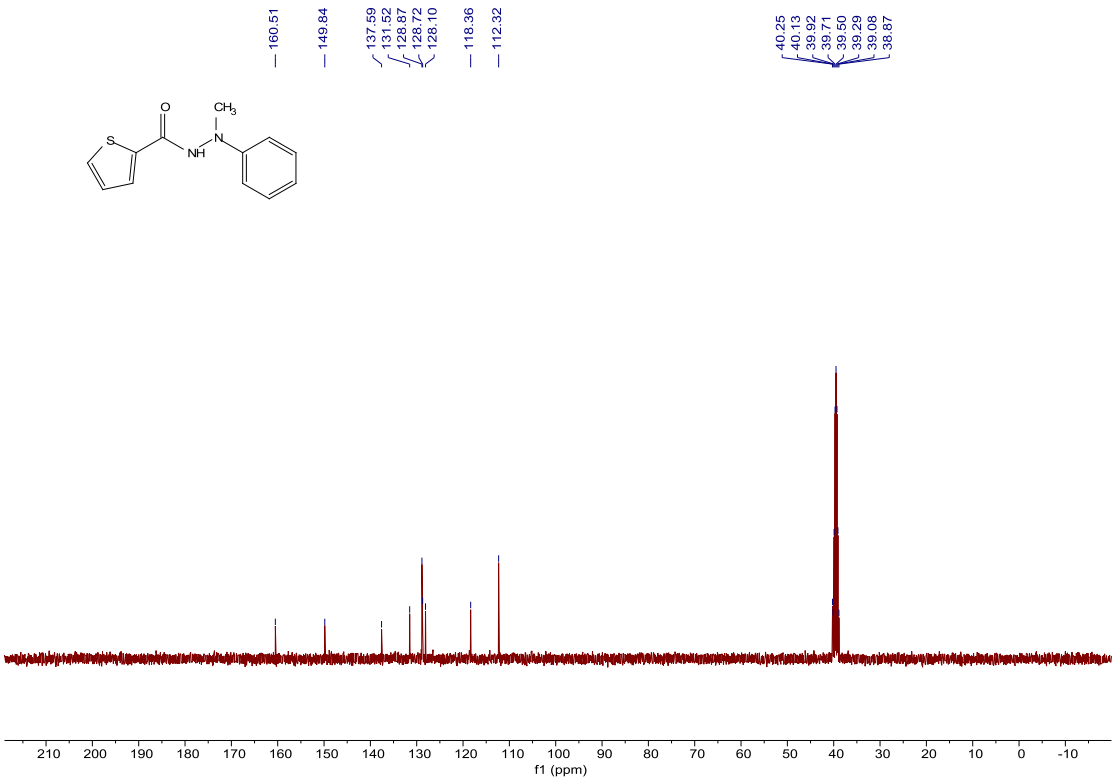
^{1}H NMR of 60 in DMSO-d_6 (400 MHz)



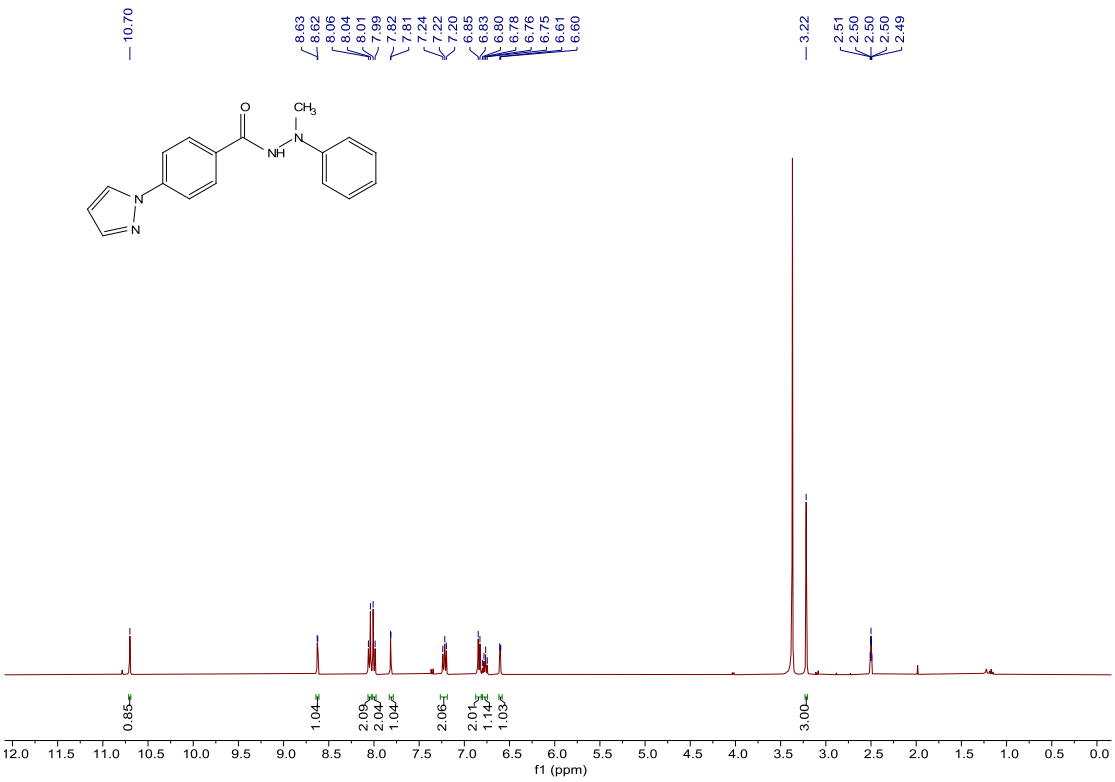
^{13}C{^{1}H} NMR of 60 in DMSO-d_6 (100 MHz)



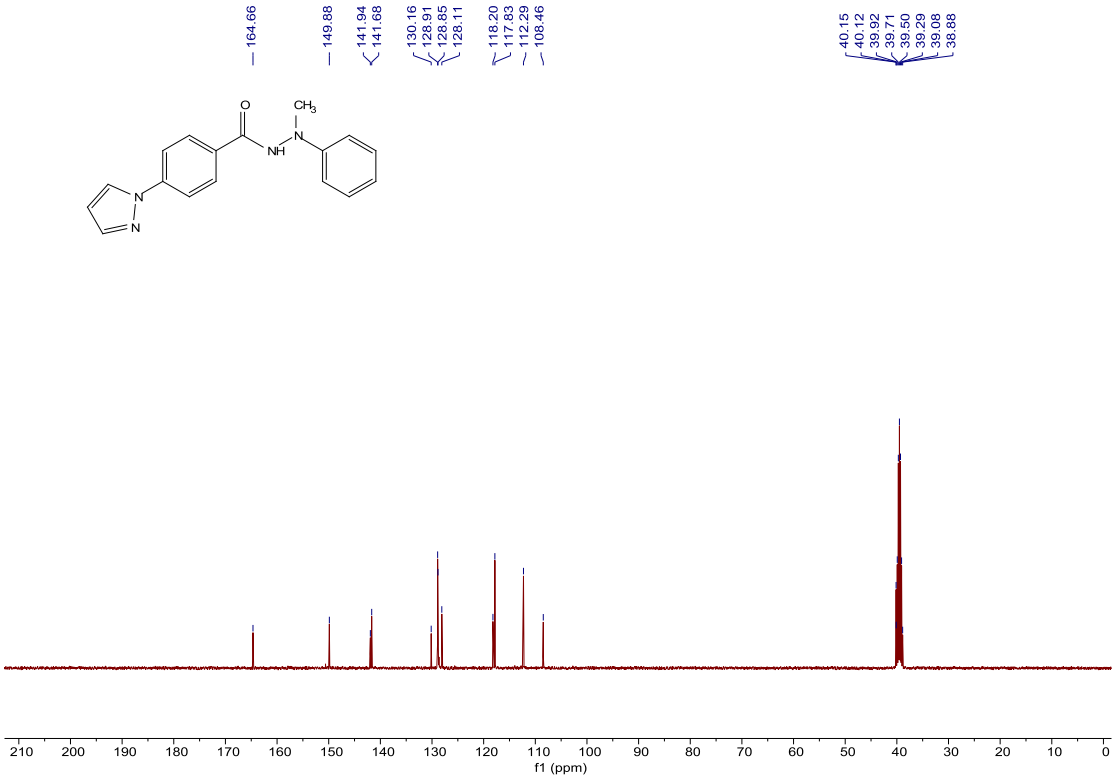
^{1}H NMR of 61 in DMSO-d_6 (400 MHz)



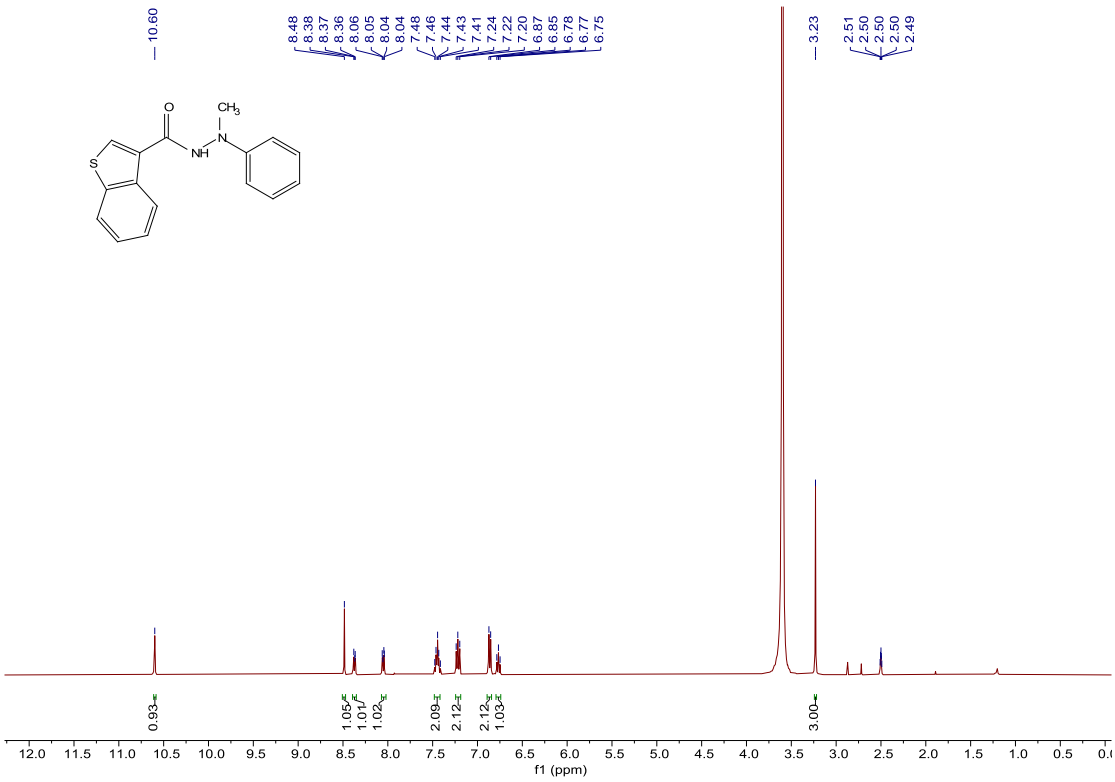
^{13}C{^{1}H} NMR of 61 in DMSO-d_6 (100 MHz)



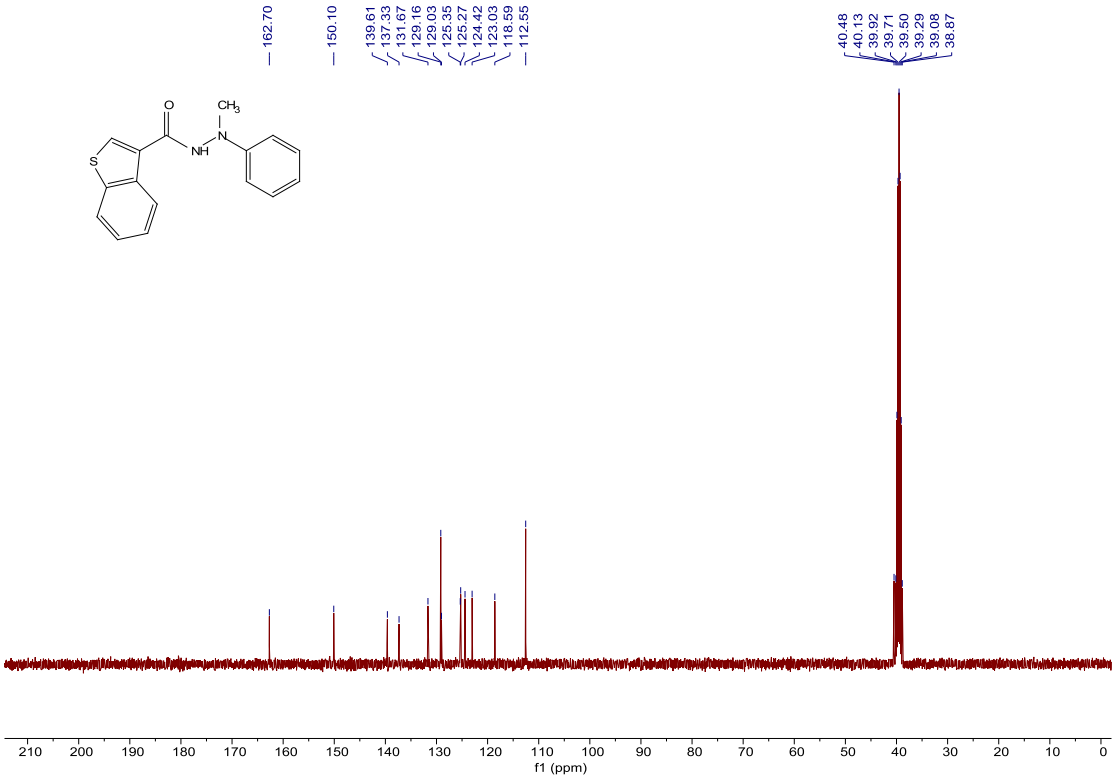
^{1}H NMR of 62 in DMSO-d_6 (400 MHz)



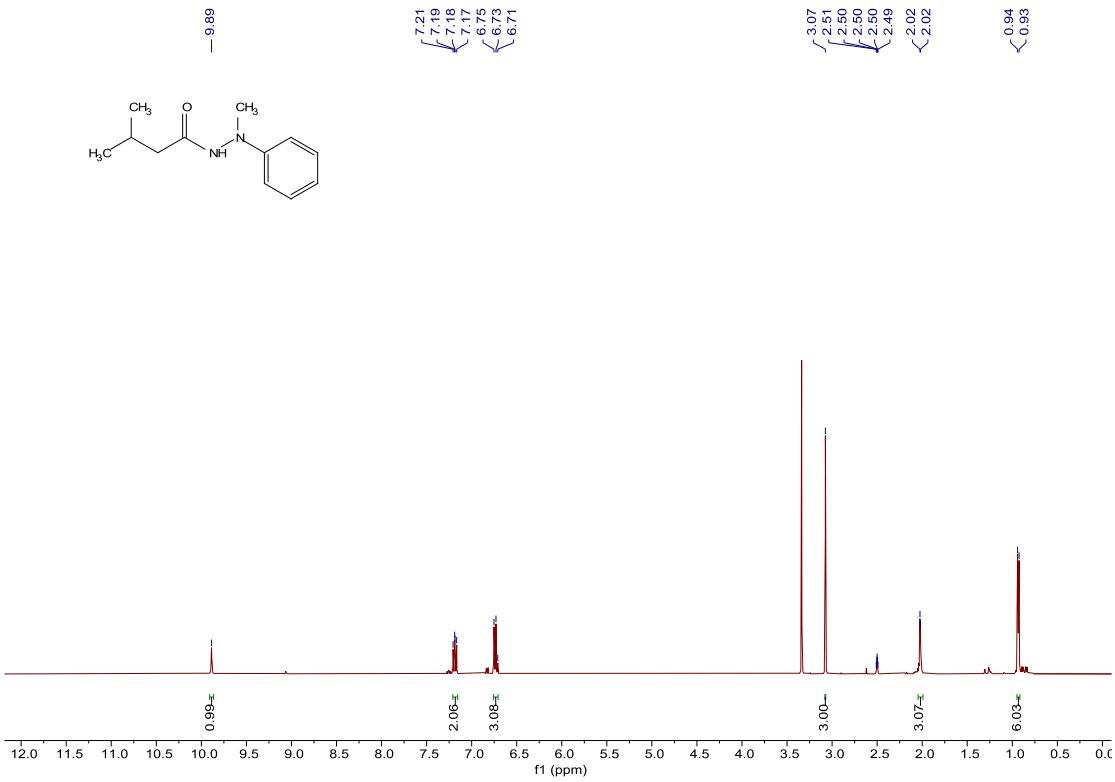
^{13}C{^{1}H} NMR of 62 in DMSO-d_6 (100 MHz)



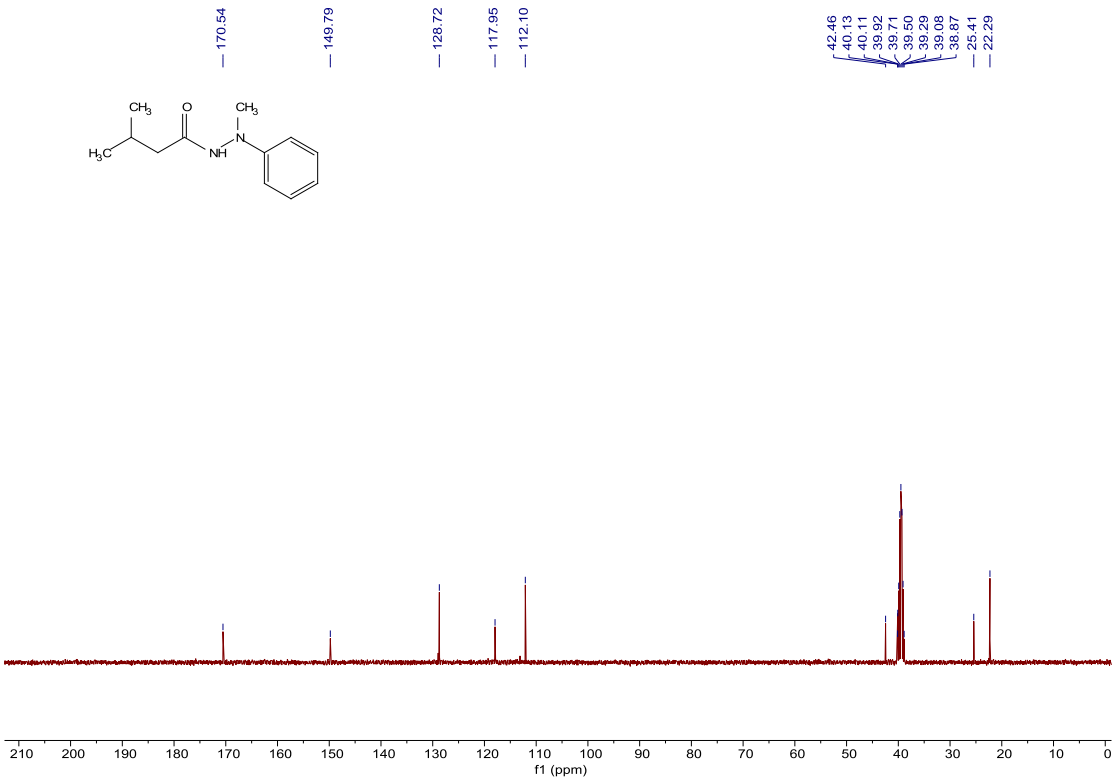
^{1}H NMR of 63 in DMSO-d_6 (400 MHz)



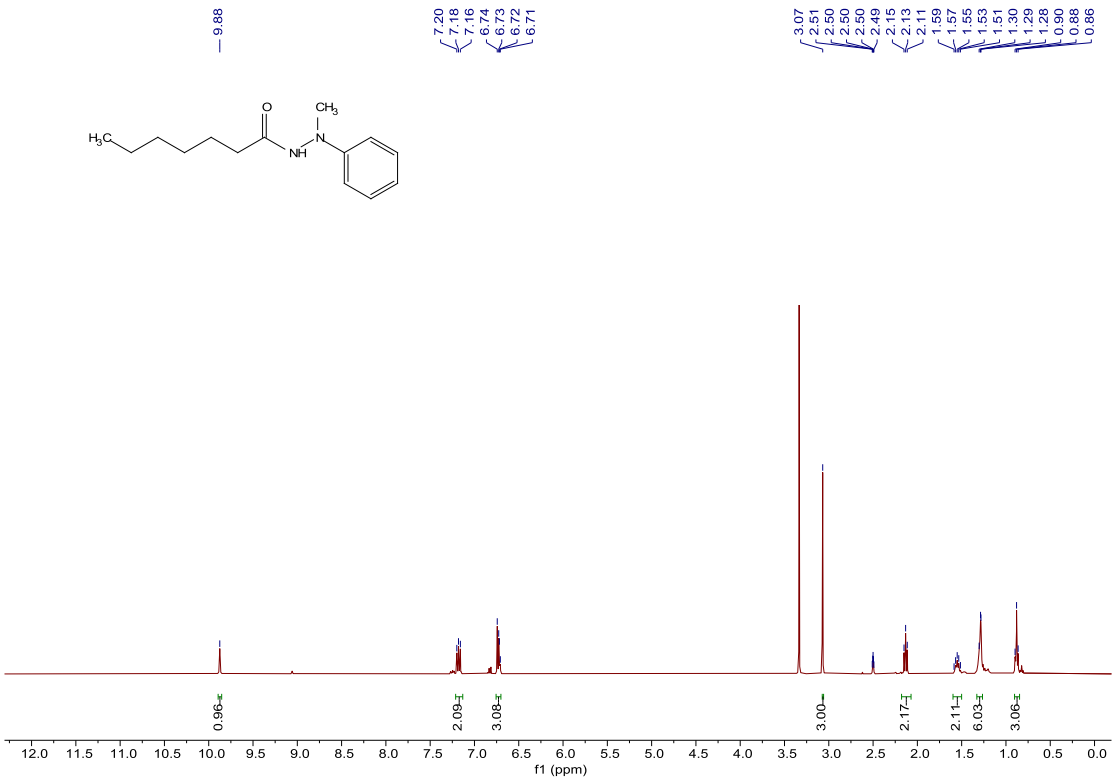
^{13}C{^{1}H} NMR of 63 in DMSO-d_6 (100 MHz)



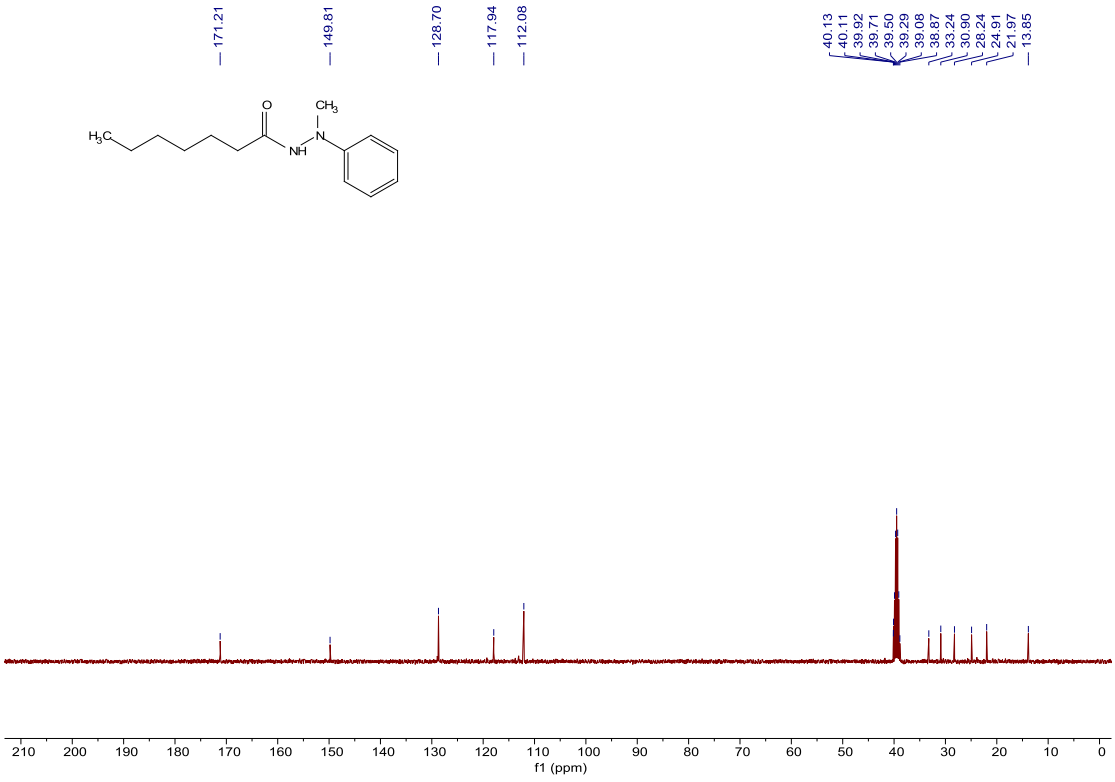
^{1}H NMR of 64 in DMSO-d_6 (400 MHz)



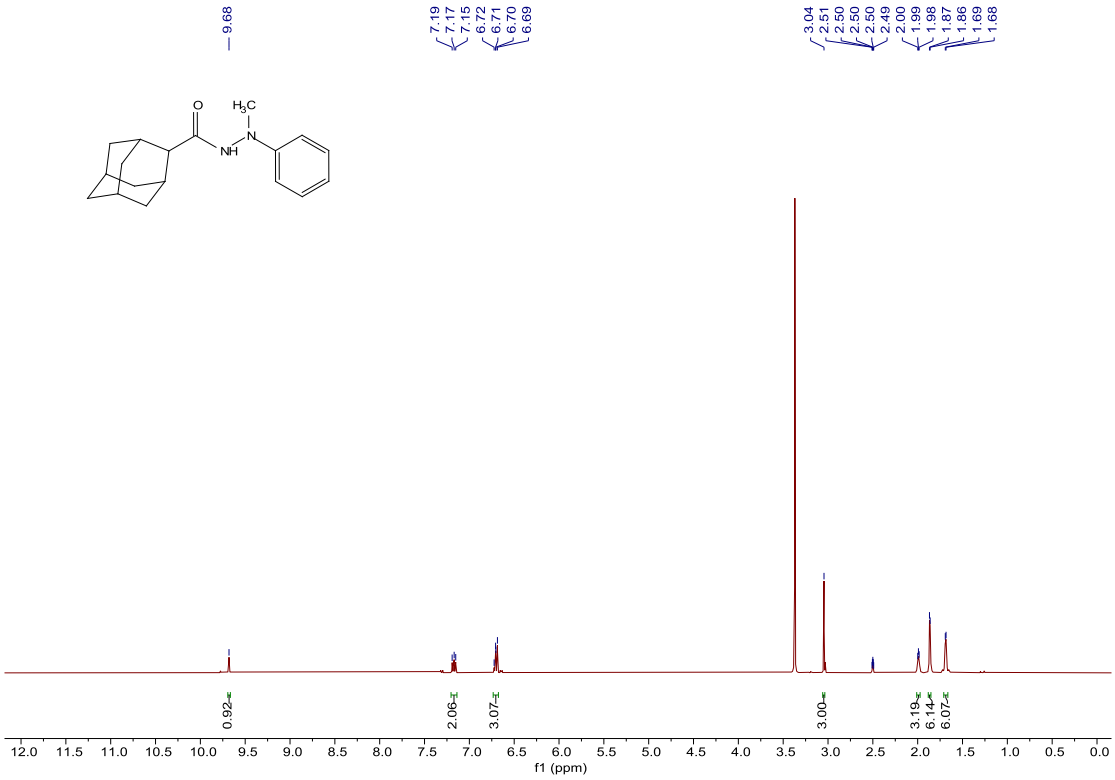
^{13}C{^{1}H} NMR of 64 in DMSO-d_6 (100 MHz)



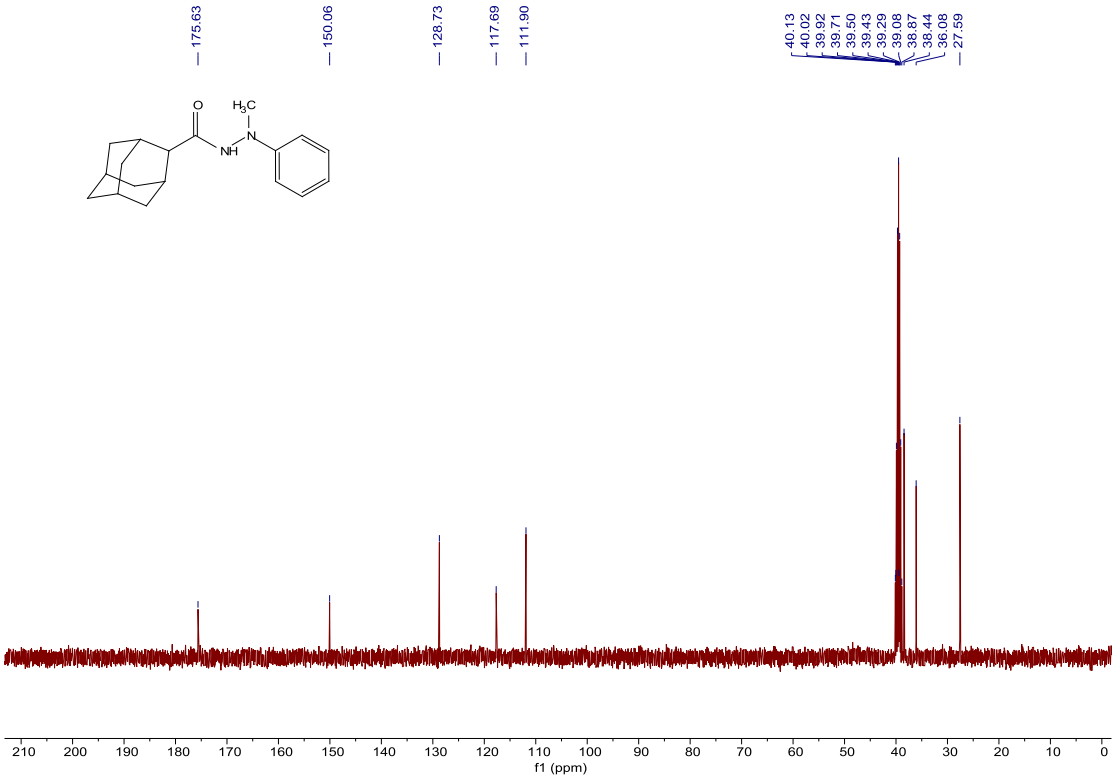
^{1}H NMR of 65 in DMSO-d_6 (400 MHz)



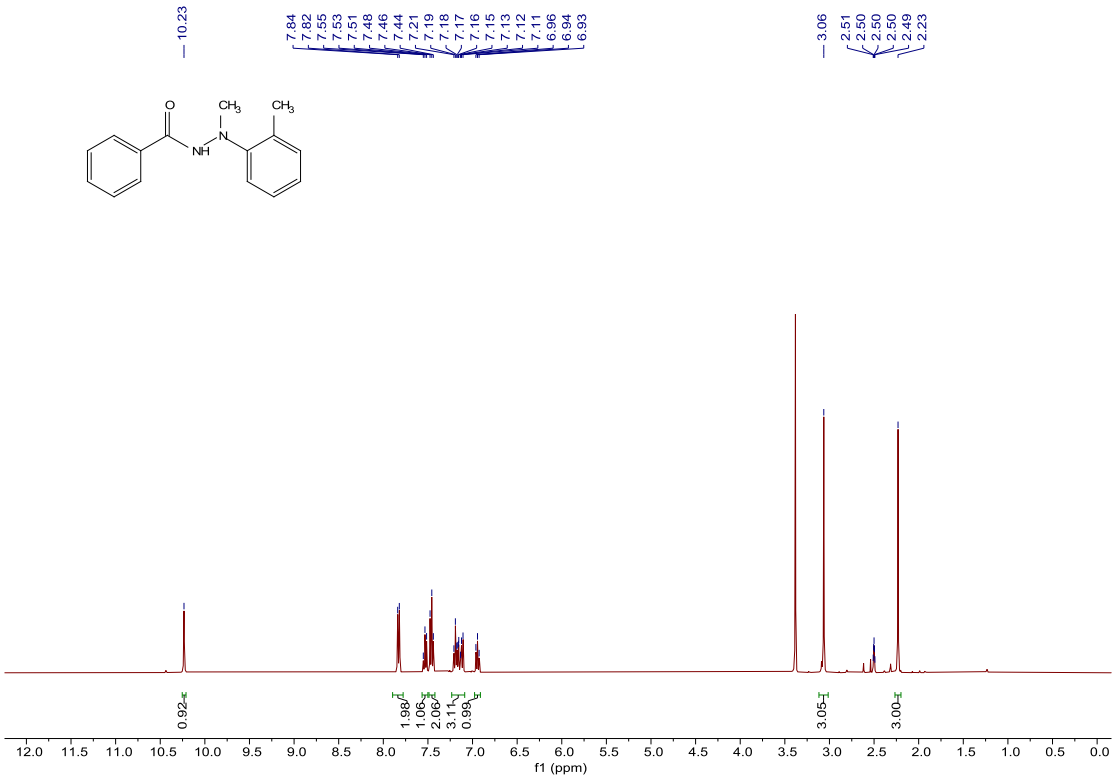
^{13}C{^{1}H} NMR of 65 in DMSO-d_6 (100 MHz)



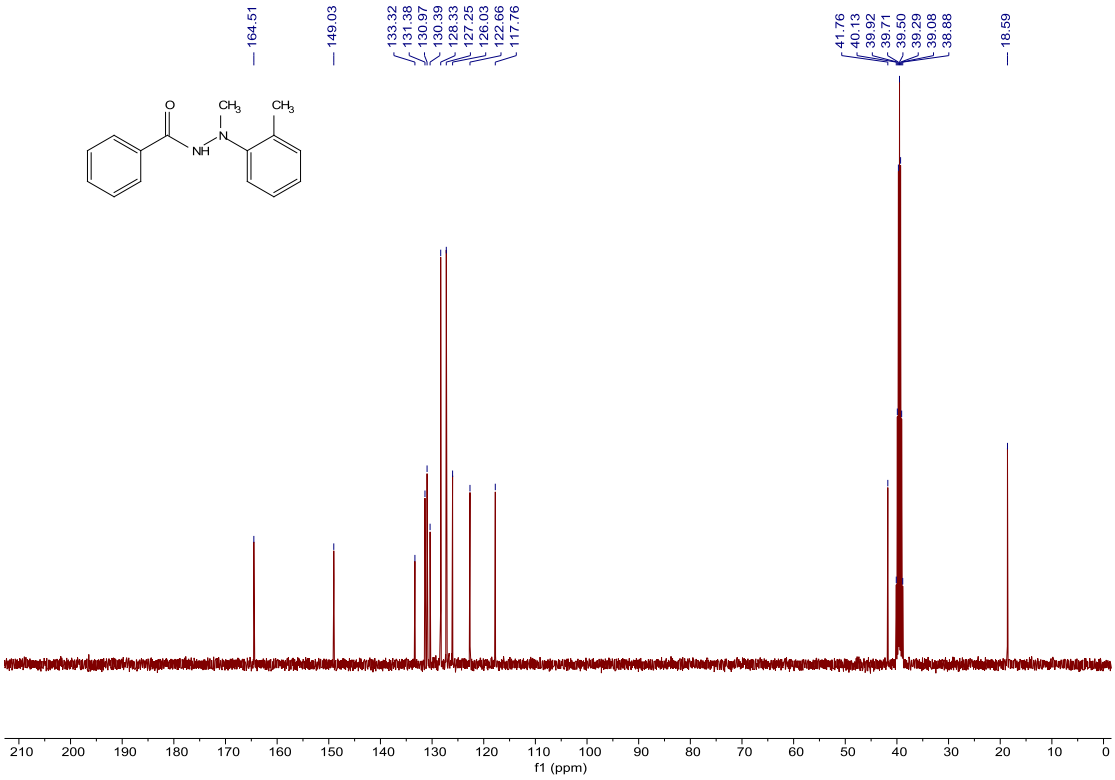
^{1}H NMR of 66 in DMSO-d_6 (400 MHz)



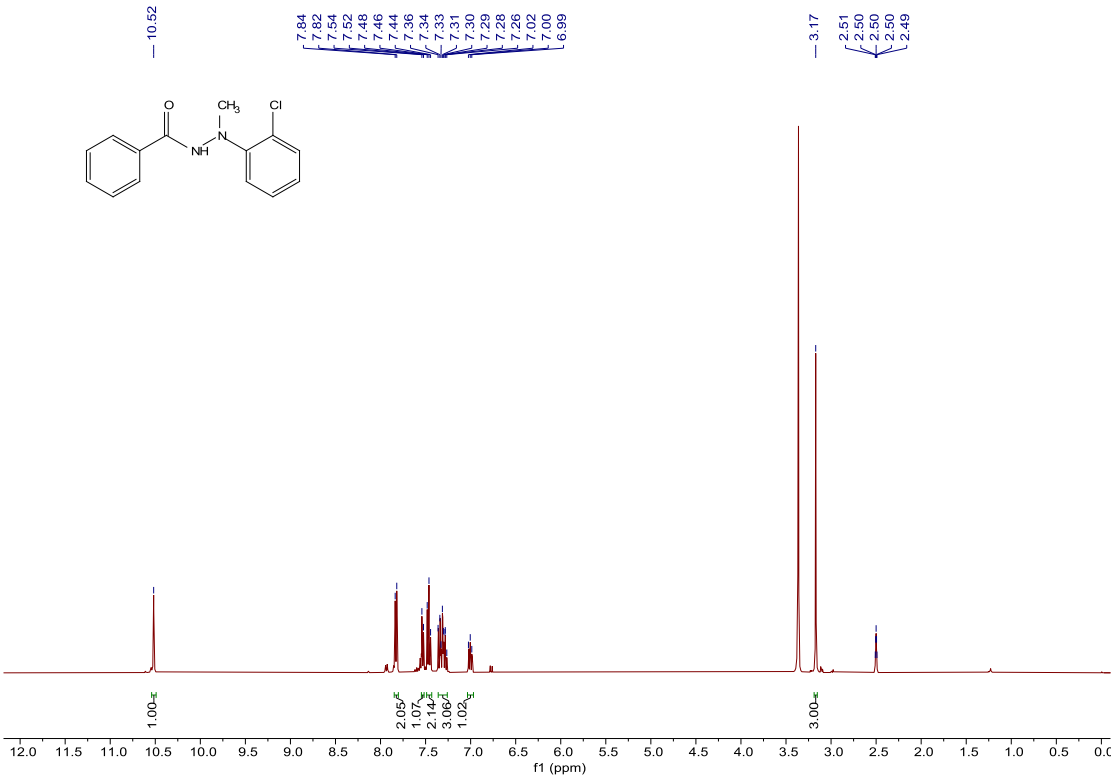
^{13}C{^{1}H} NMR of 66 in DMSO-d_6 (100 MHz)



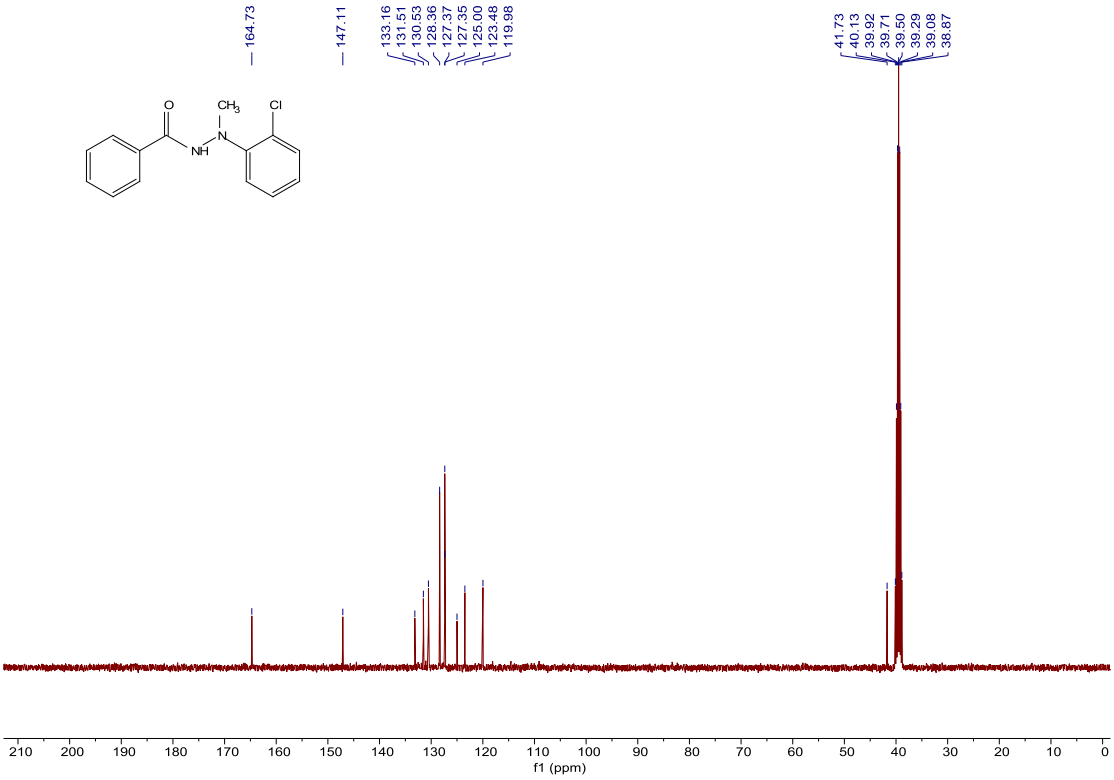
^{1}H NMR of 67 in DMSO-d_6 (400 MHz)



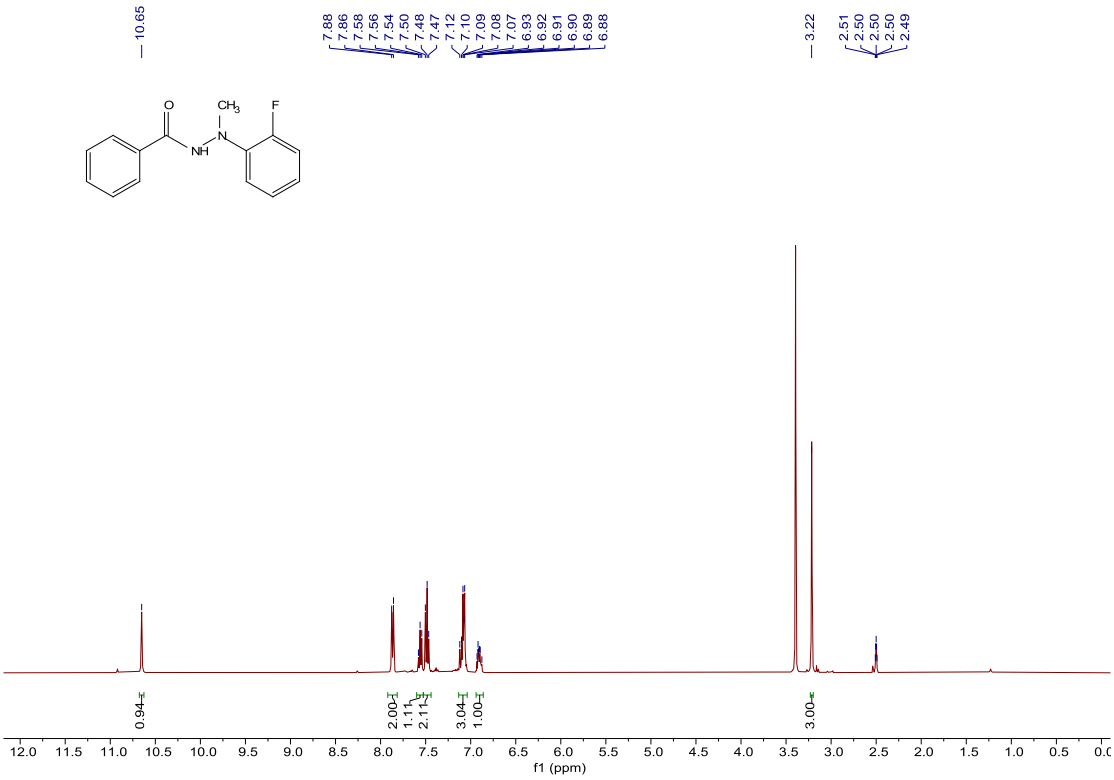
^{13}C{^{1}H} NMR of 67 in DMSO-d_6 (100 MHz)



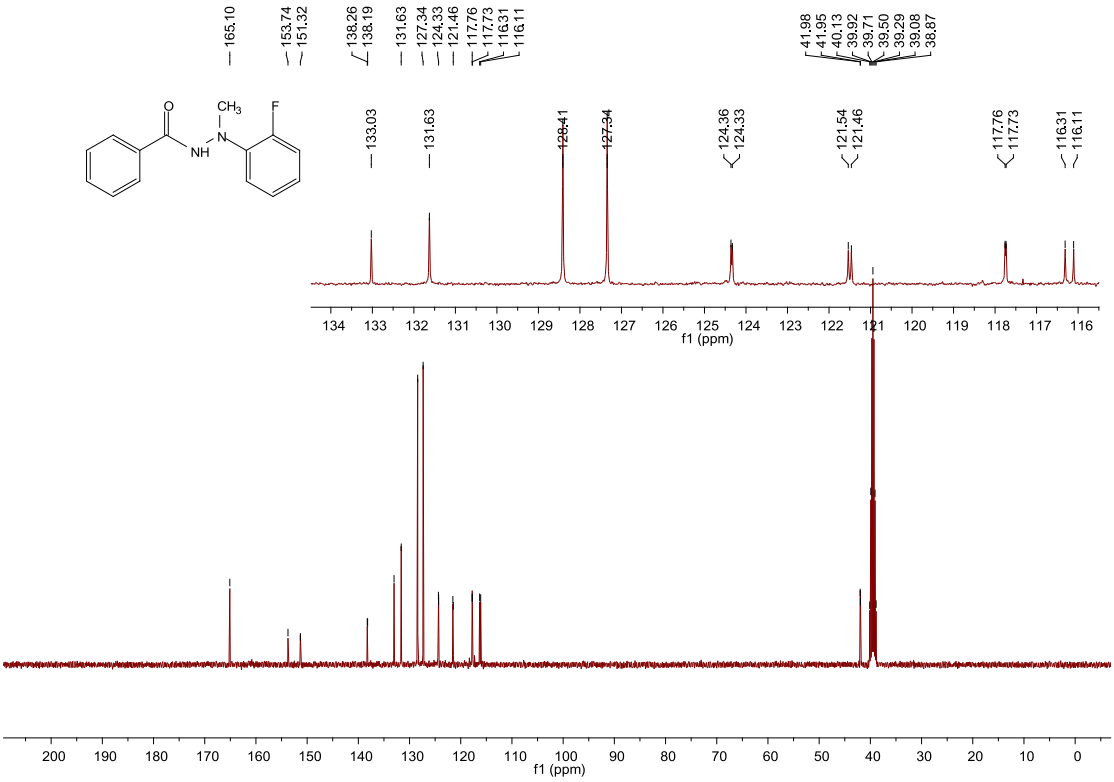
^{1}H NMR of 68 in DMSO-d_6 (400 MHz)



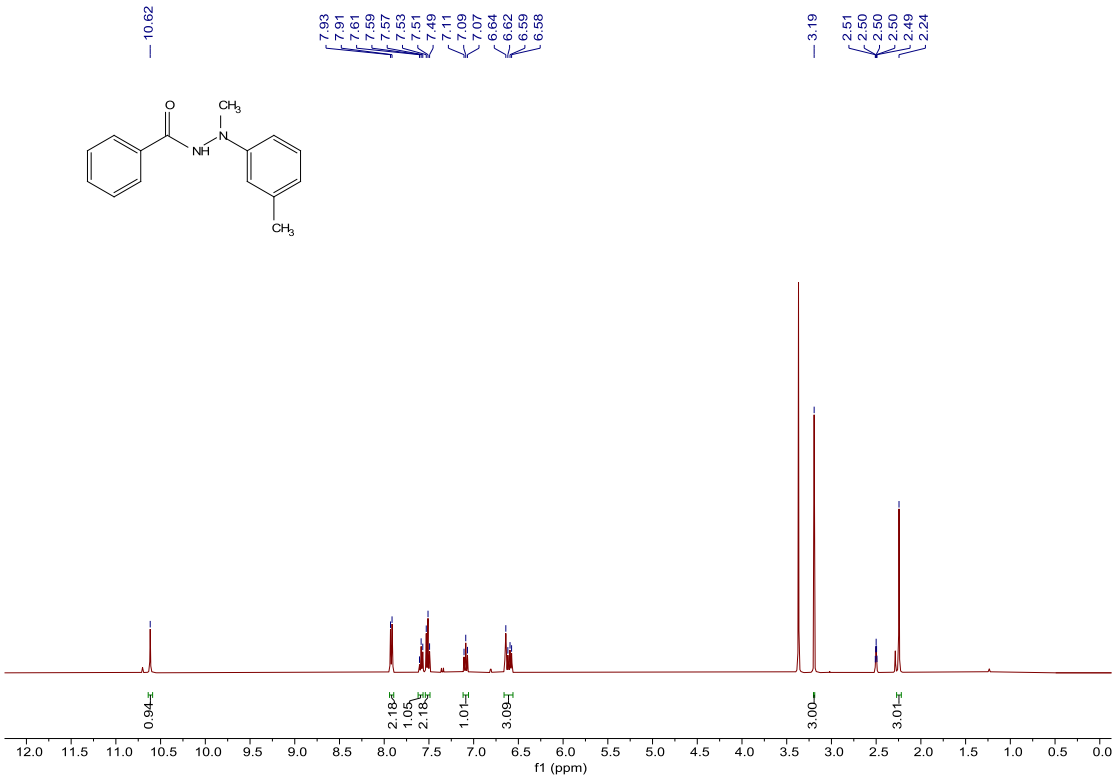
^{13}C{^{1}H} NMR of 68 in DMSO-d_6 (100 MHz)



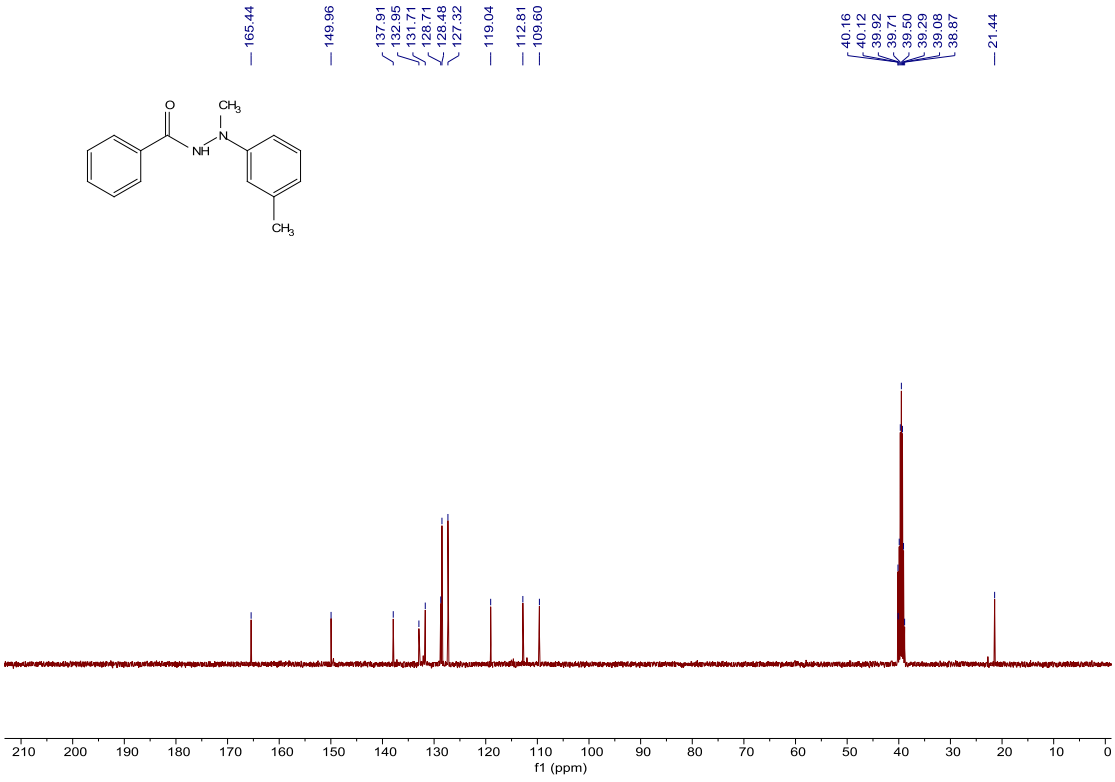
^{1}H NMR of 69 in DMSO-d_6 (400 MHz)



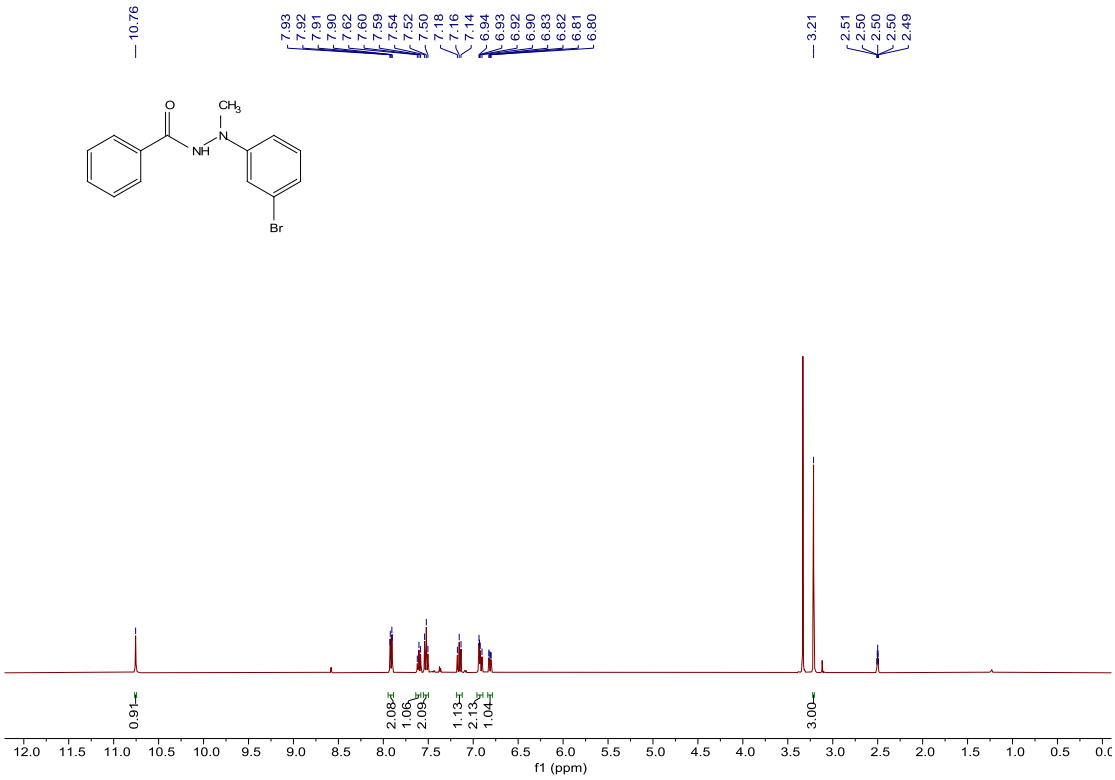
^{13}C{^{1}H} NMR of 69 in DMSO-d_6 (100 MHz)



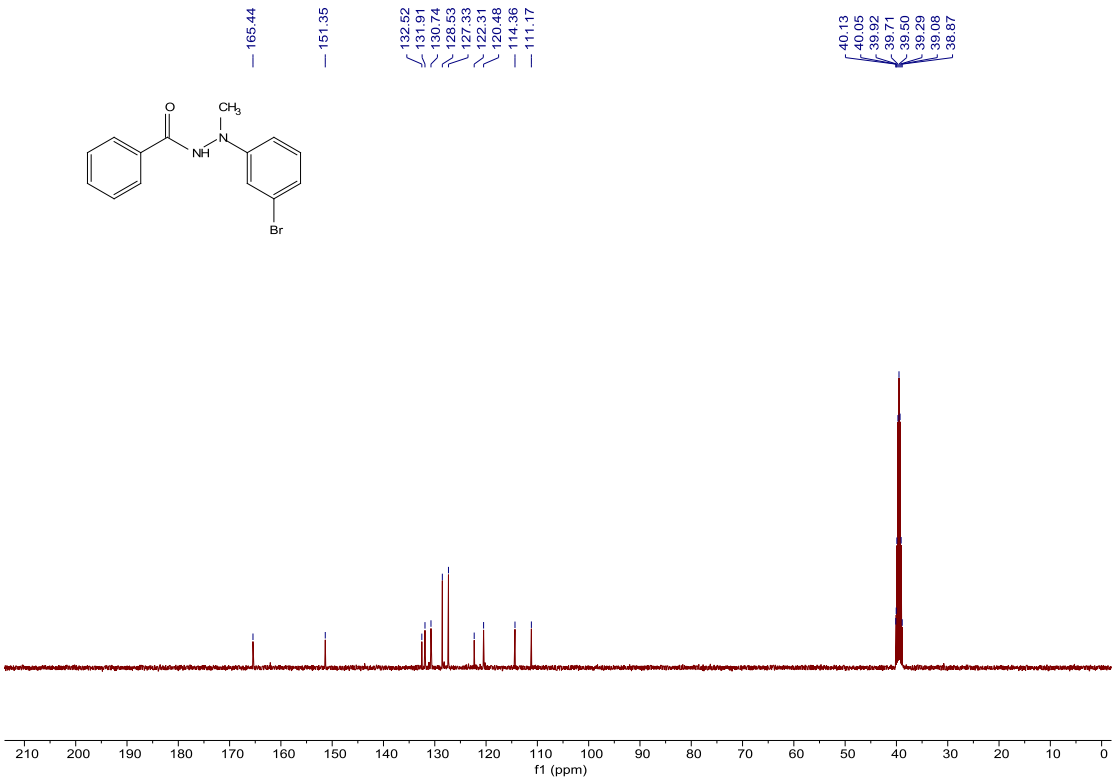
^{1}H NMR of 70 in DMSO-d_6 (400 MHz)



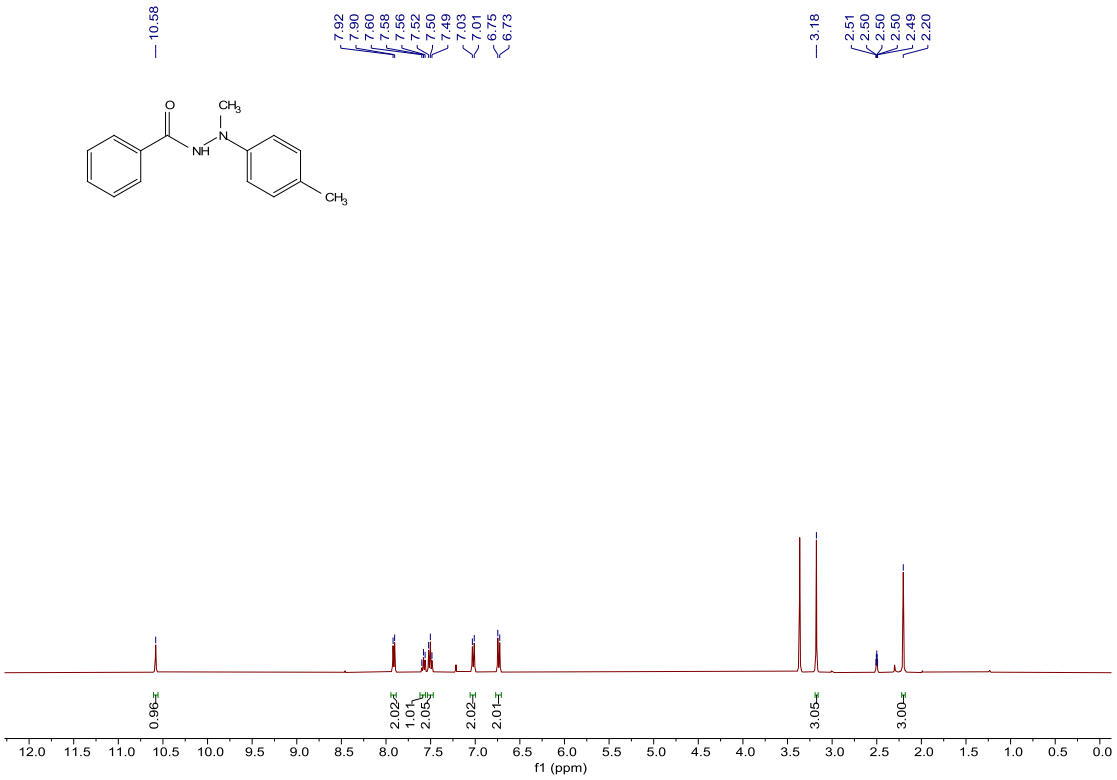
^{13}C{^{1}H} NMR of 70 in DMSO-d_6 (100 MHz)



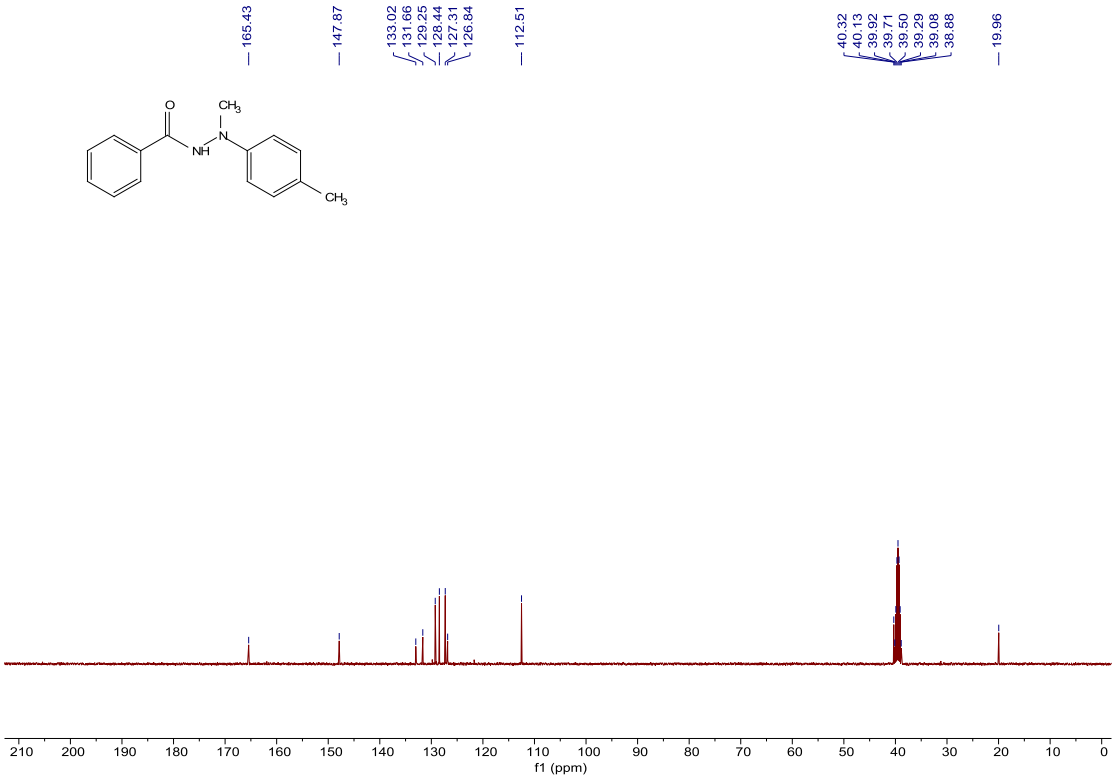
^{1}H NMR of 71 in DMSO-d_6 (400 MHz)



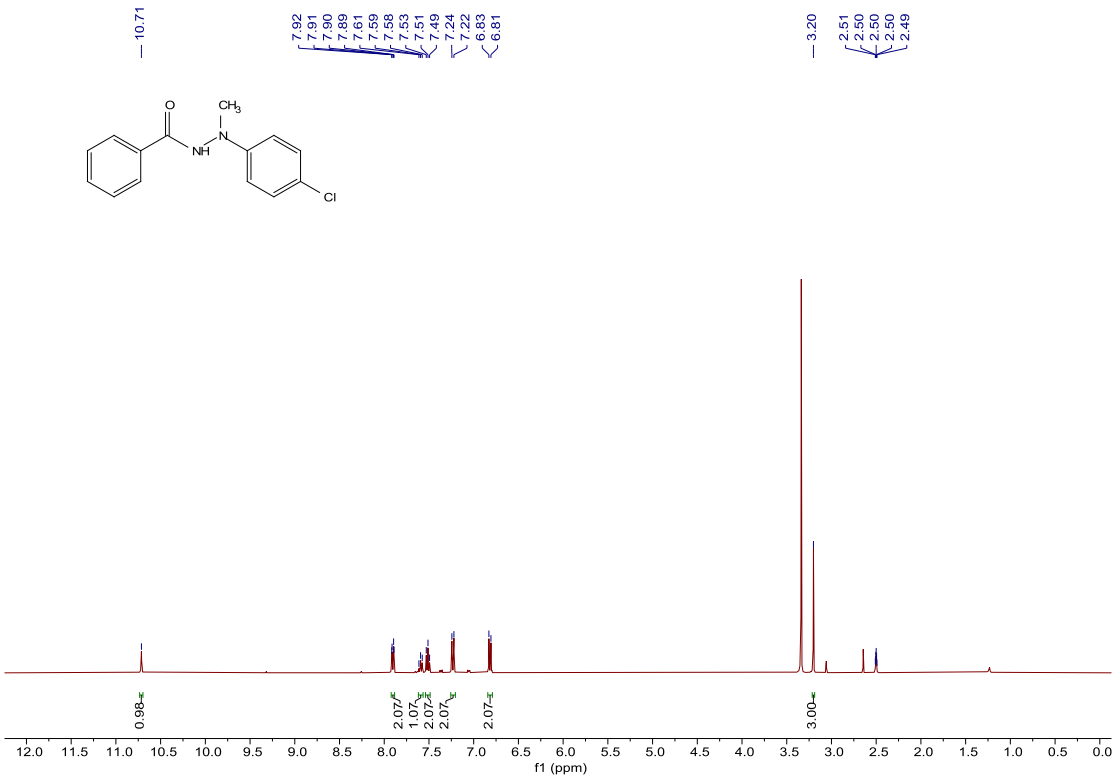
^{13}C{^{1}H} NMR of 71 in DMSO-d_6 (100 MHz)



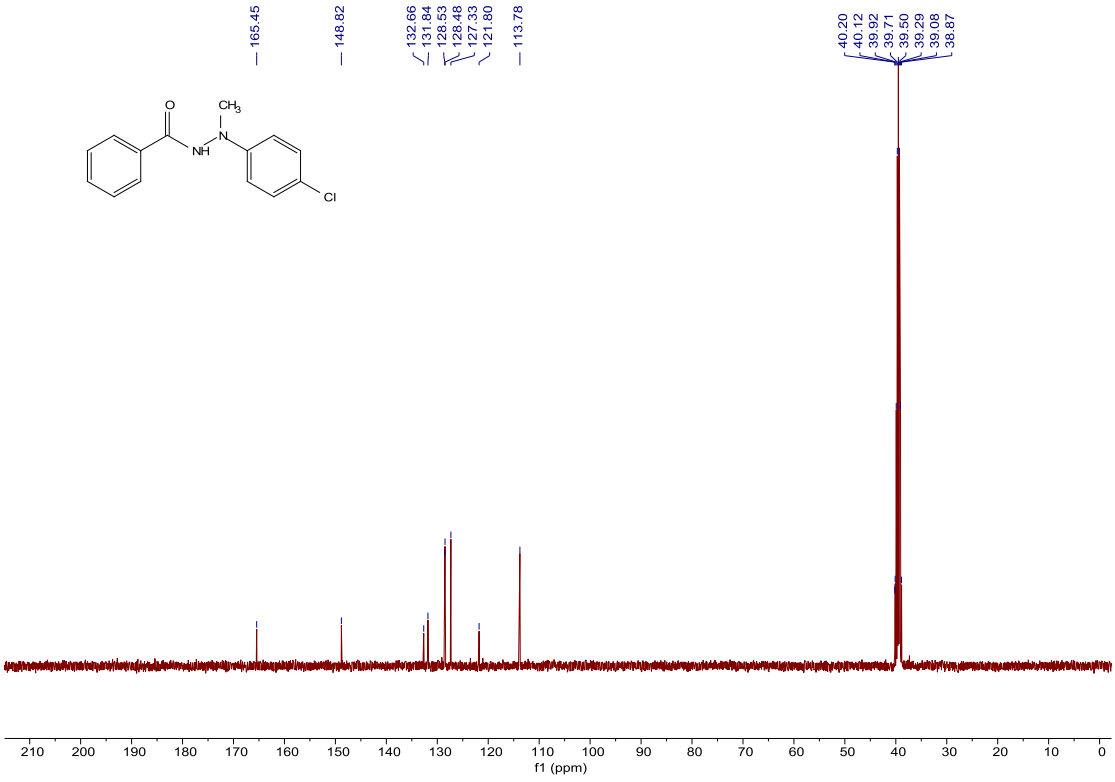
^{1}H NMR of 72 in DMSO-d_6 (400 MHz)



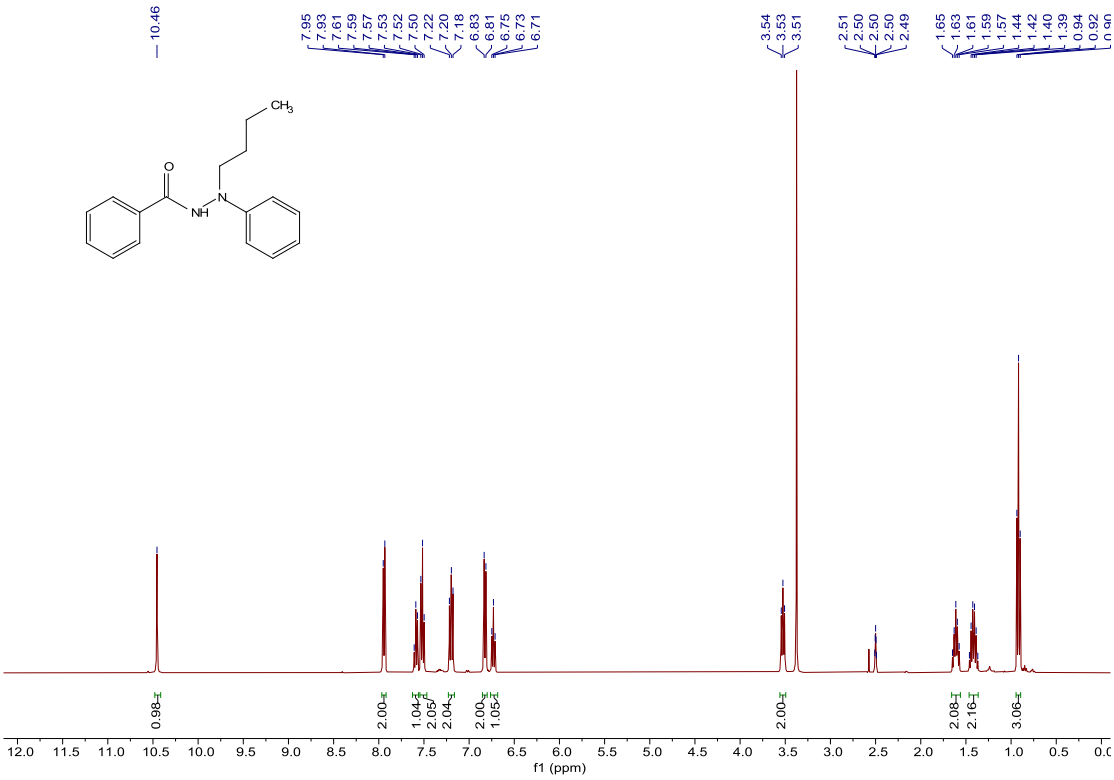
^{13}C{^{1}H} NMR of 72 in DMSO-d_6 (100 MHz)



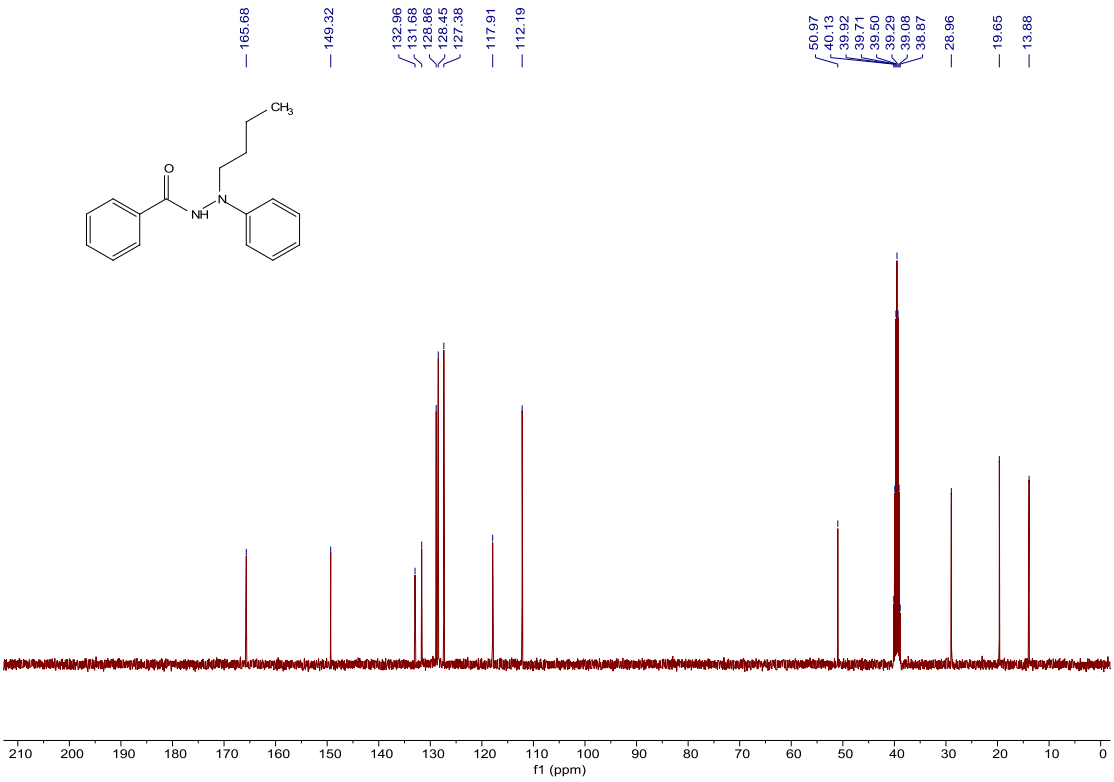
^{1}H NMR of 73 in DMSO-d_6 (400 MHz)



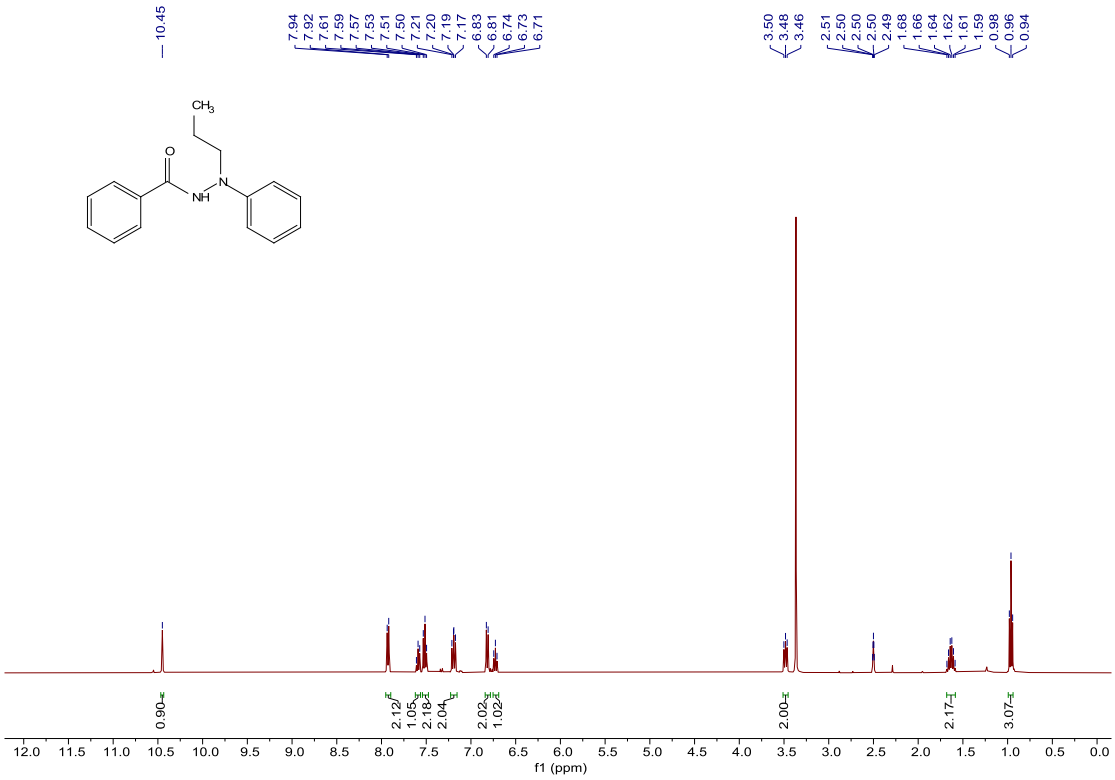
^{13}C{^{1}H} NMR of 73 in DMSO-d_6 (100 MHz)



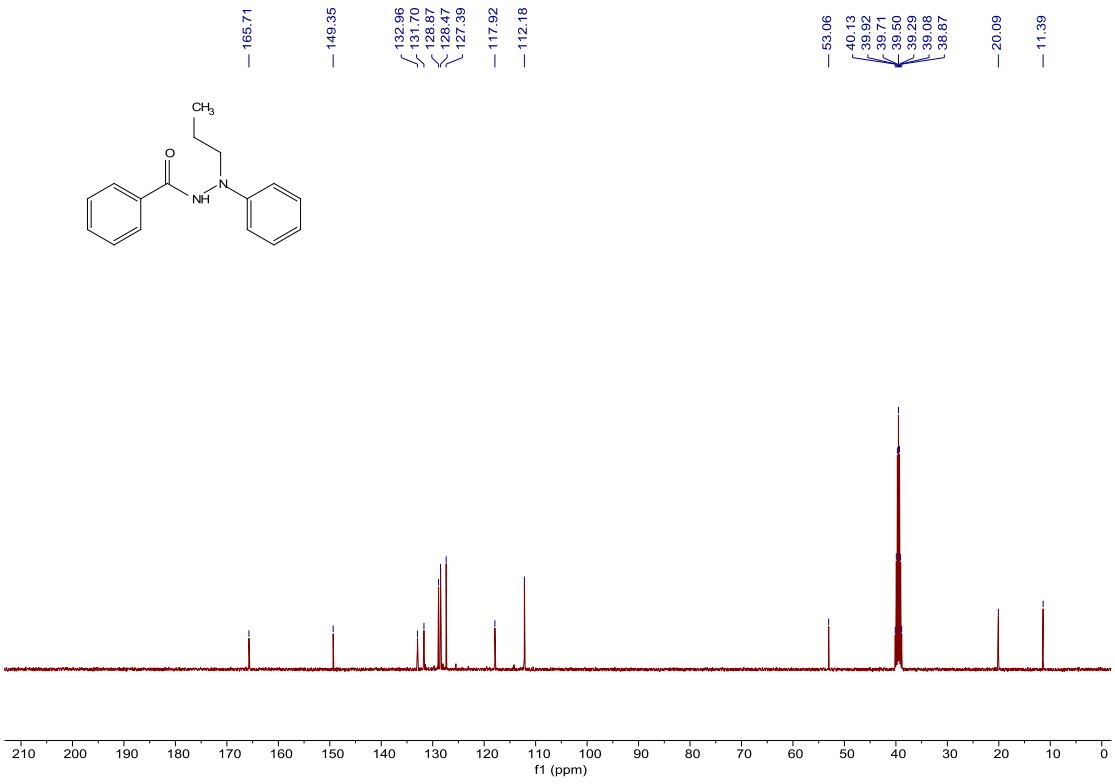
^{1}H NMR of 74 in DMSO-d_6 (400 MHz)



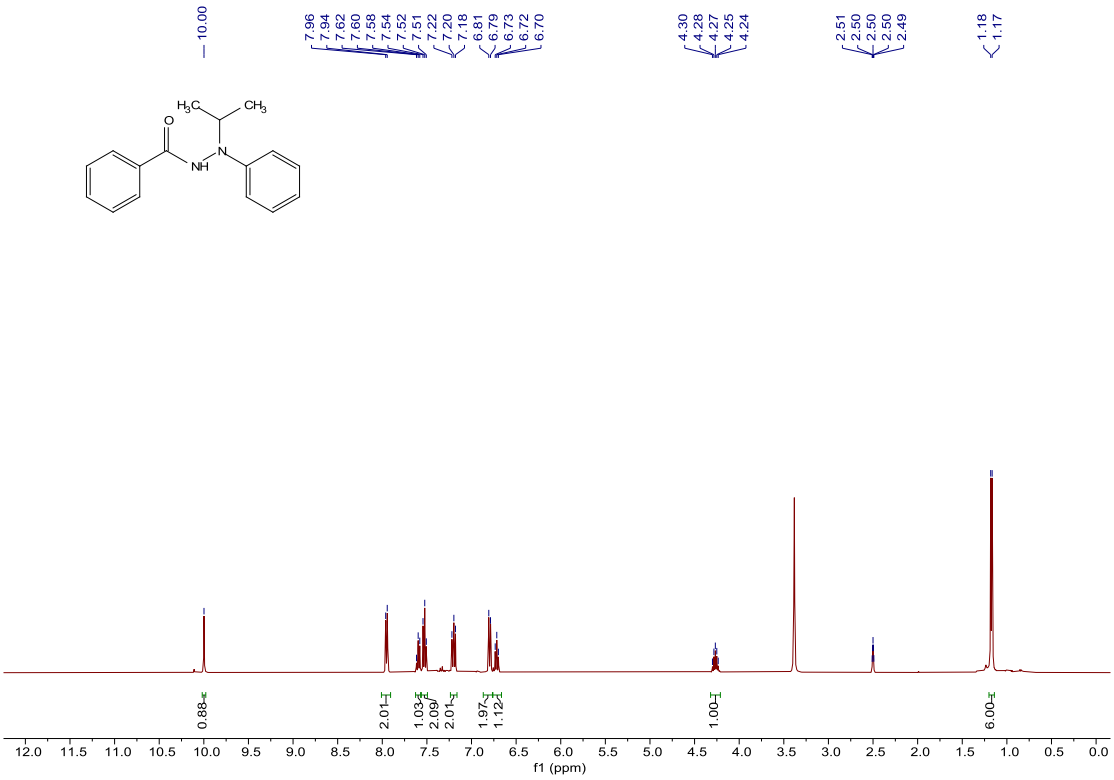
^{13}C{^{1}H} NMR of 74 in DMSO-d_6 (100 MHz)



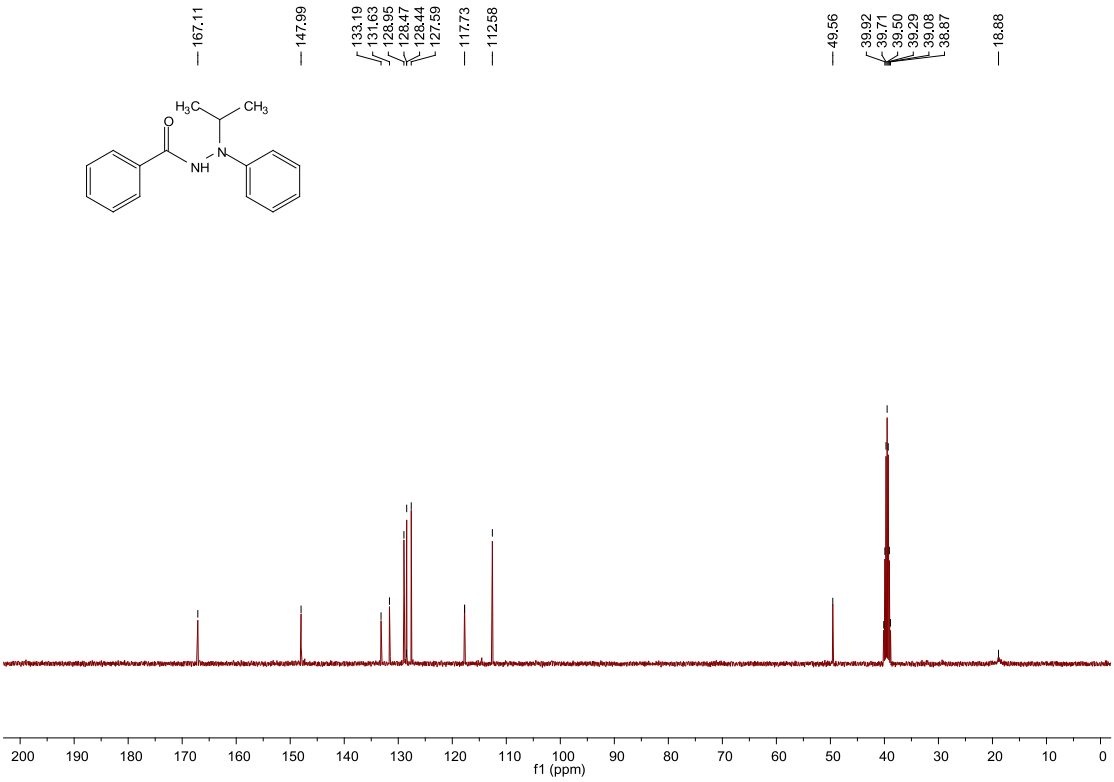
^{1}H NMR of 75 in DMSO-d_6 (400 MHz)



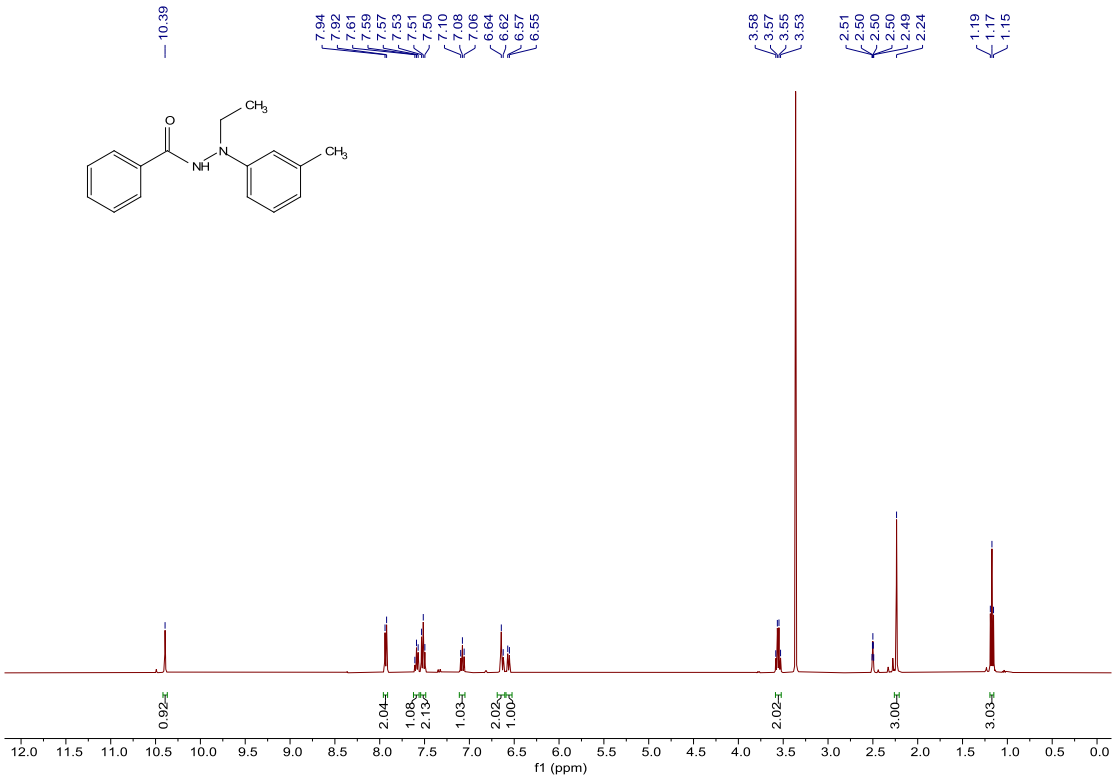
^{13}C{^{1}H} NMR of 75 in DMSO-d_6 (100 MHz)



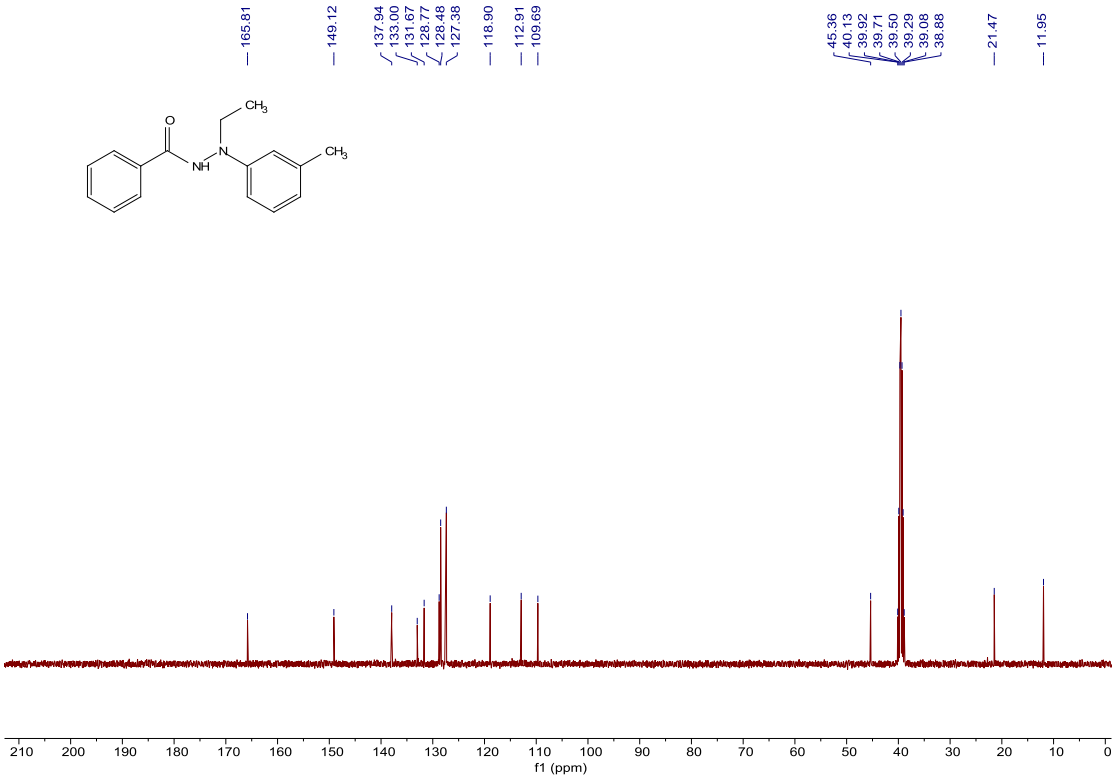
^{1}H NMR of 76 in DMSO-d_6 (400 MHz)



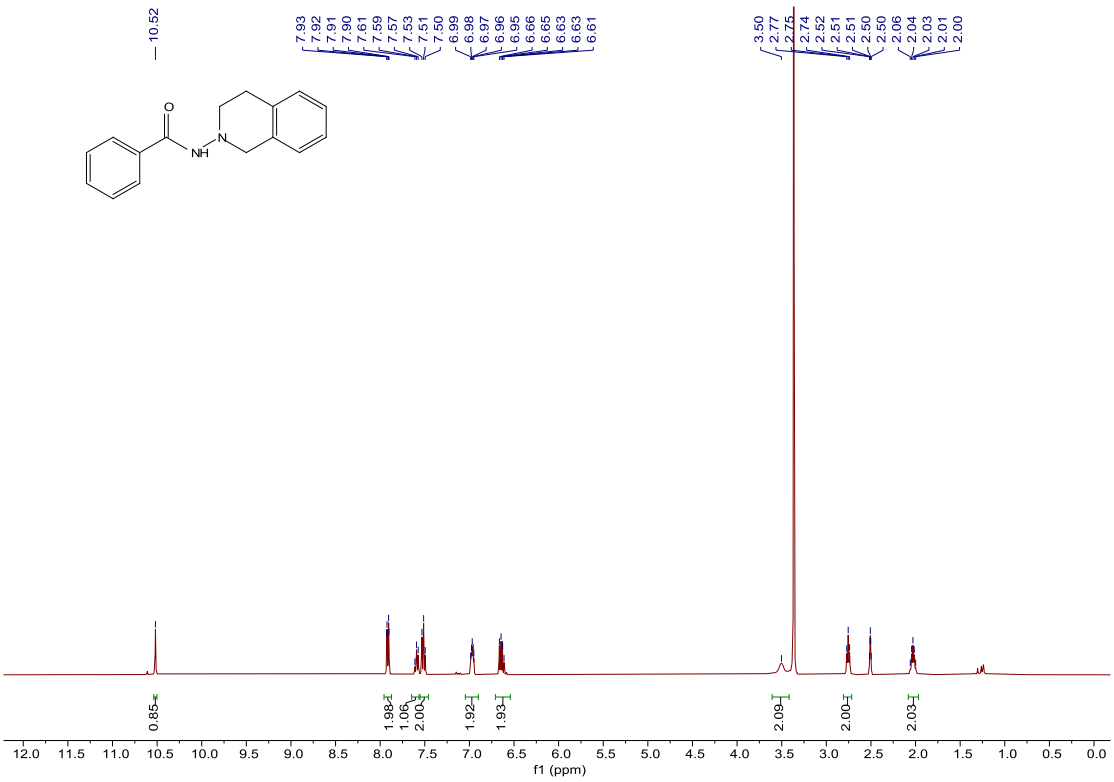
^{13}C{^{1}H} NMR of 76 in DMSO-d_6 (100 MHz)



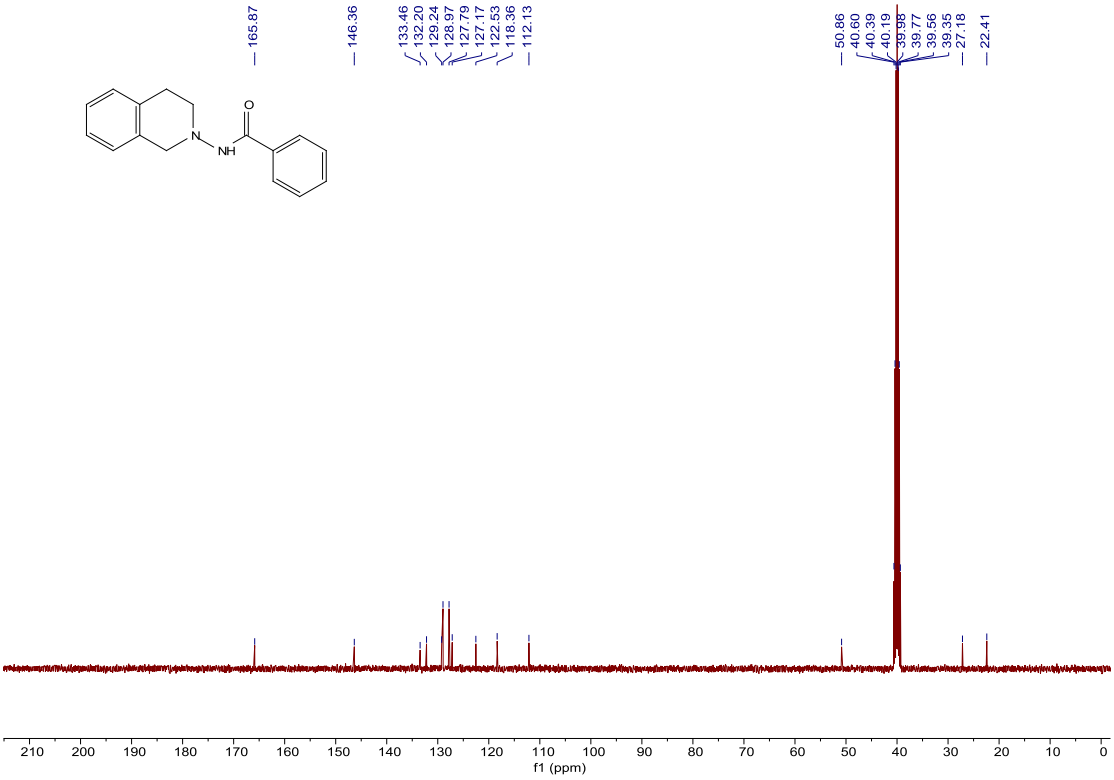
^{1}H NMR of 77 in DMSO-d_6 (400 MHz)



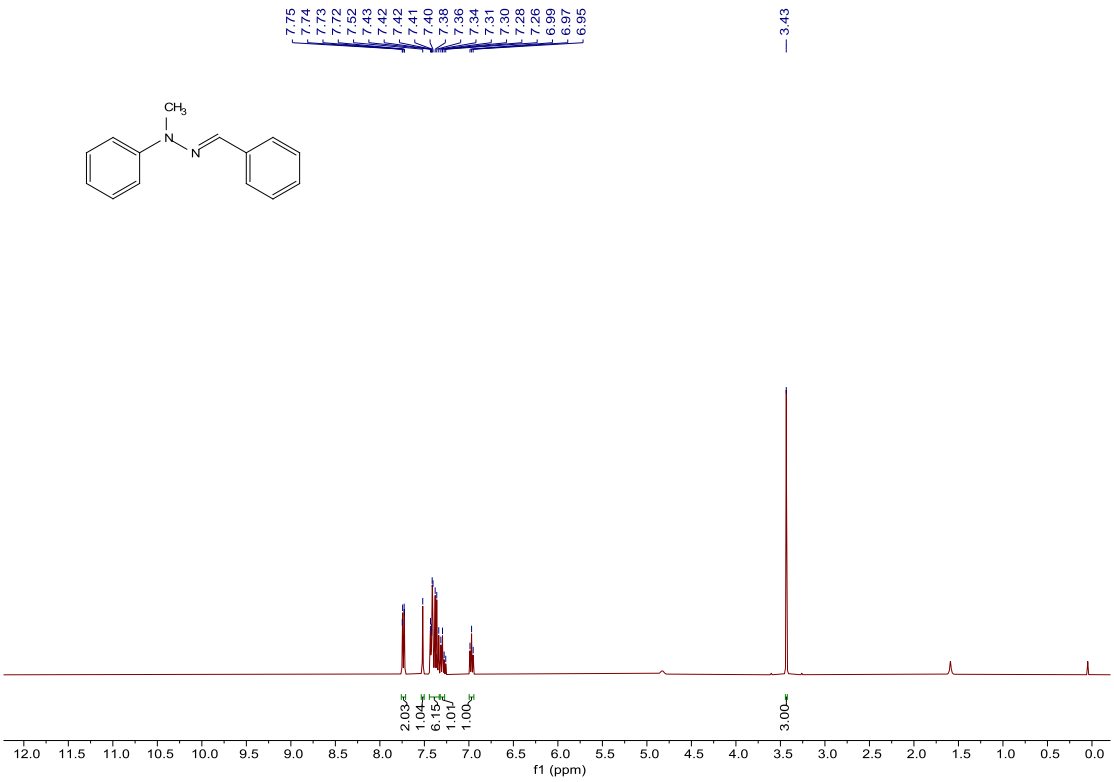
^{13}C{^{1}H} NMR of 77 in DMSO-d_6 (100 MHz)



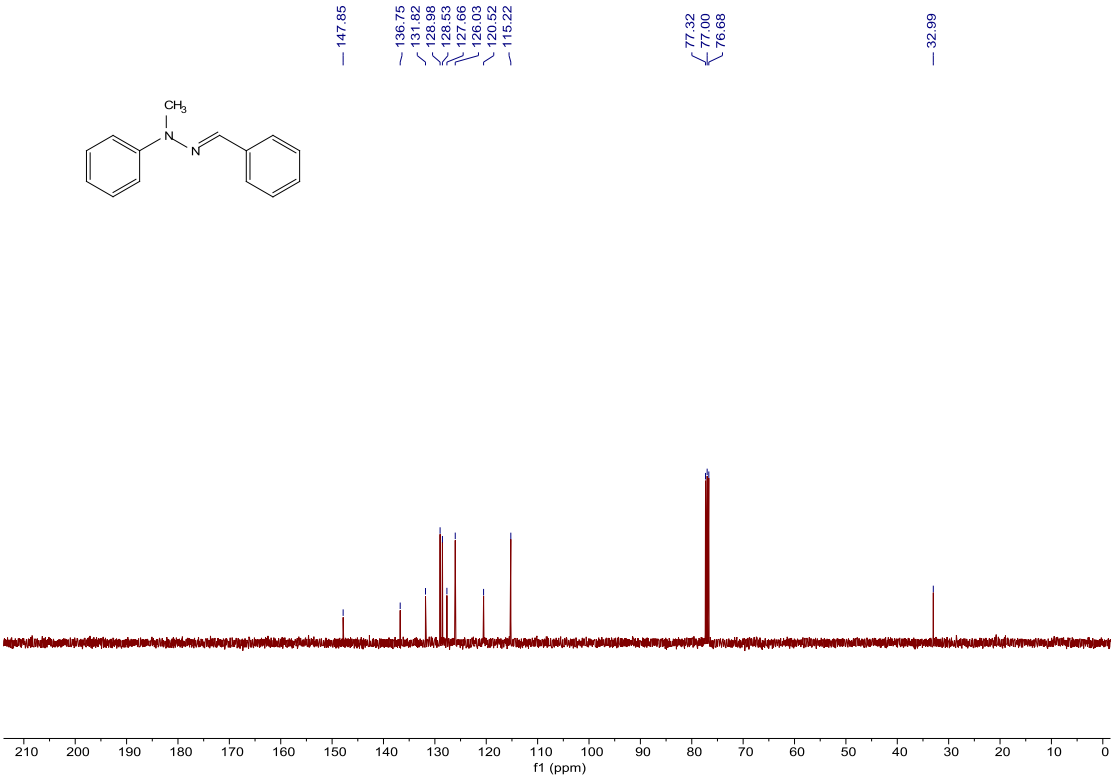
^{1}H NMR of 78 in DMSO-d_6 (400 MHz)



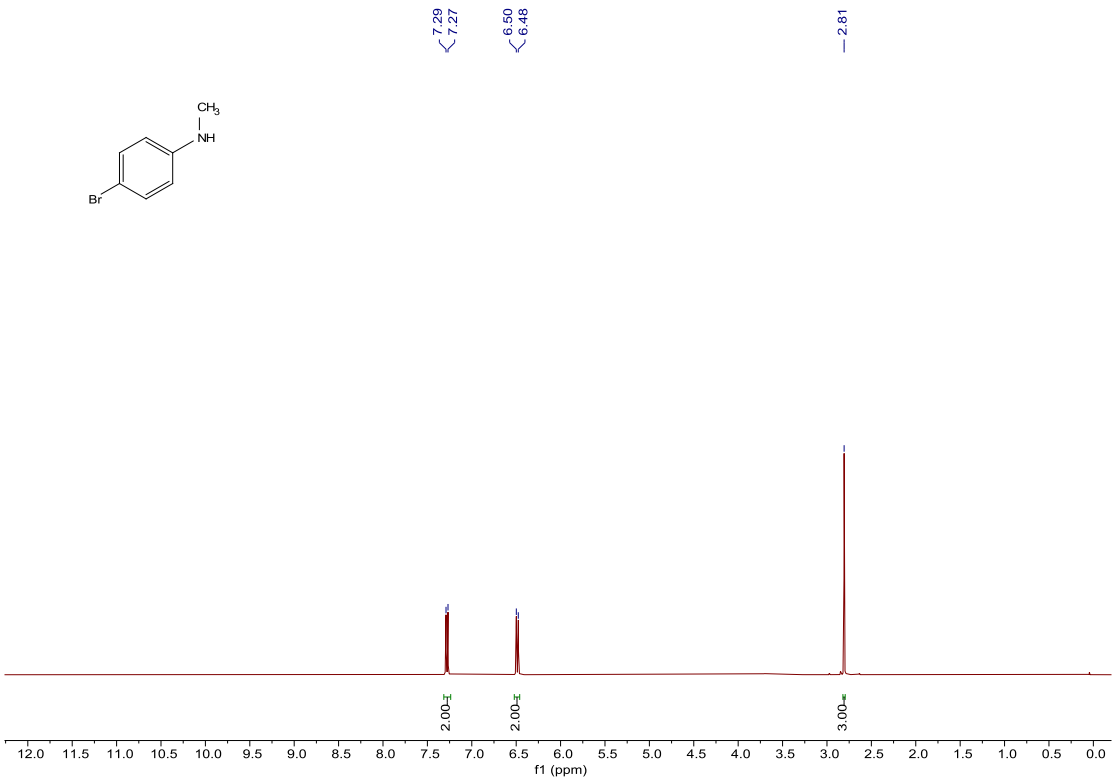
^{13}C{^{1}H} NMR of 78 in DMSO-d_6 (100 MHz)



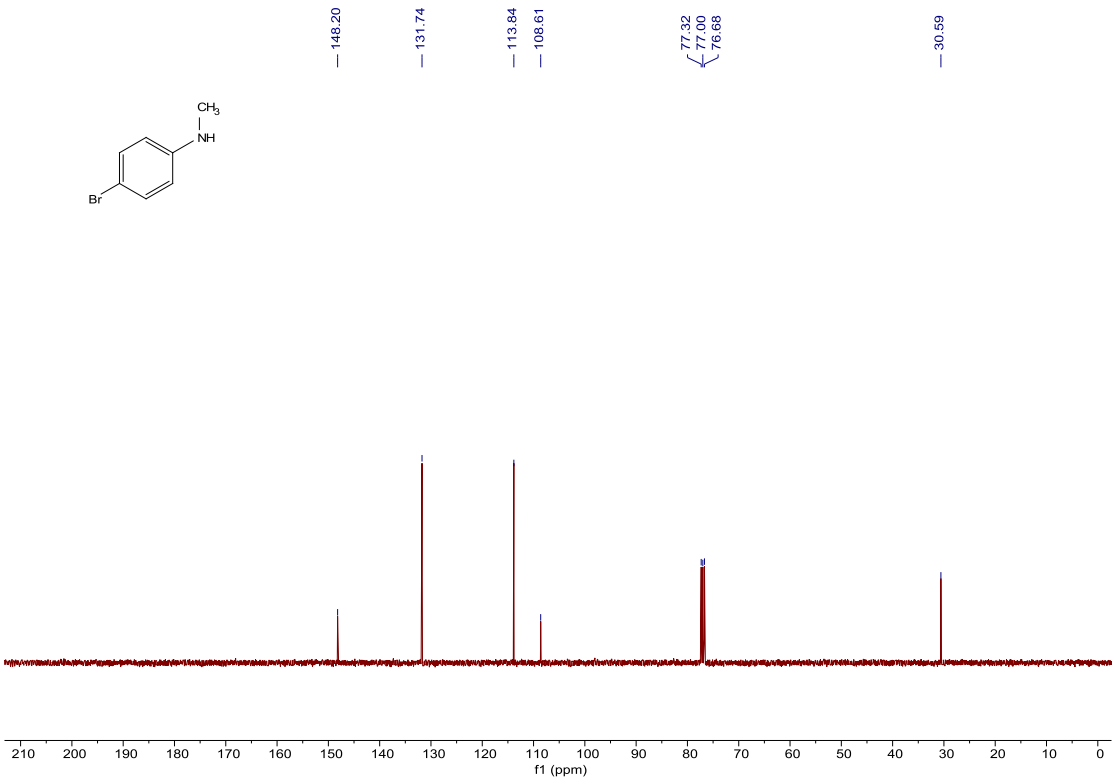
^{1}H NMR of 80 in $CDCl_3$ (400 MHz)



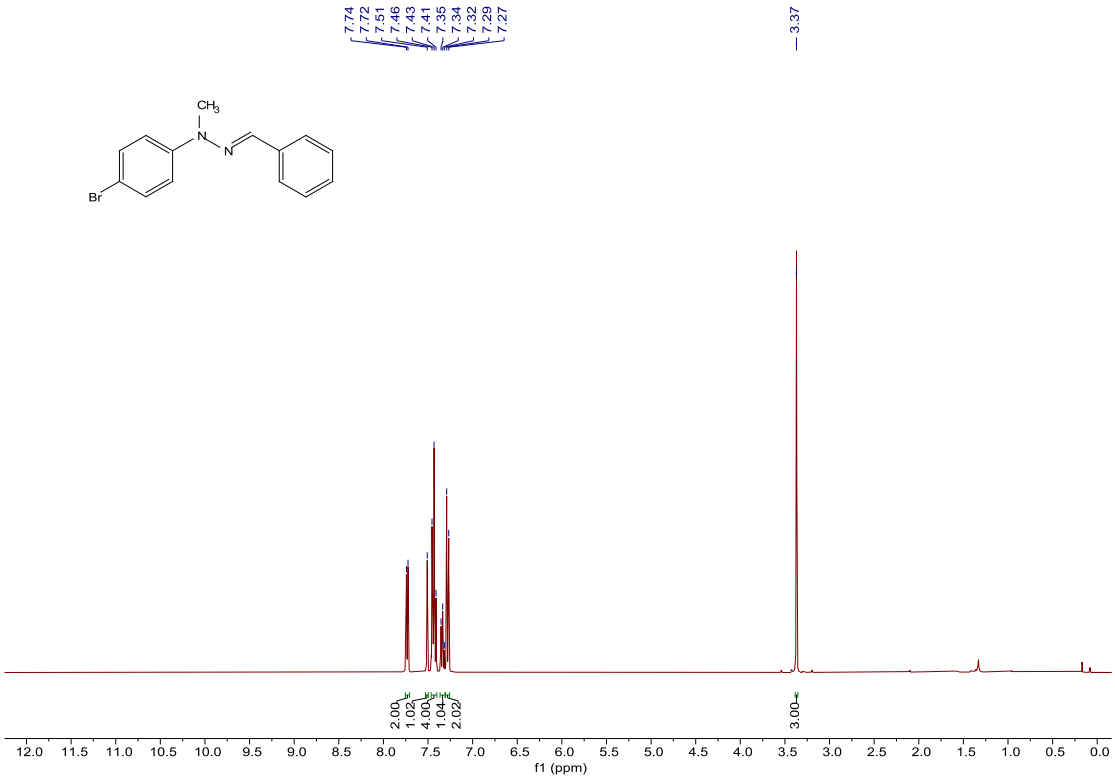
^{13}C{^{1}H} NMR of 80 in $CDCl_3$ (100 MHz)



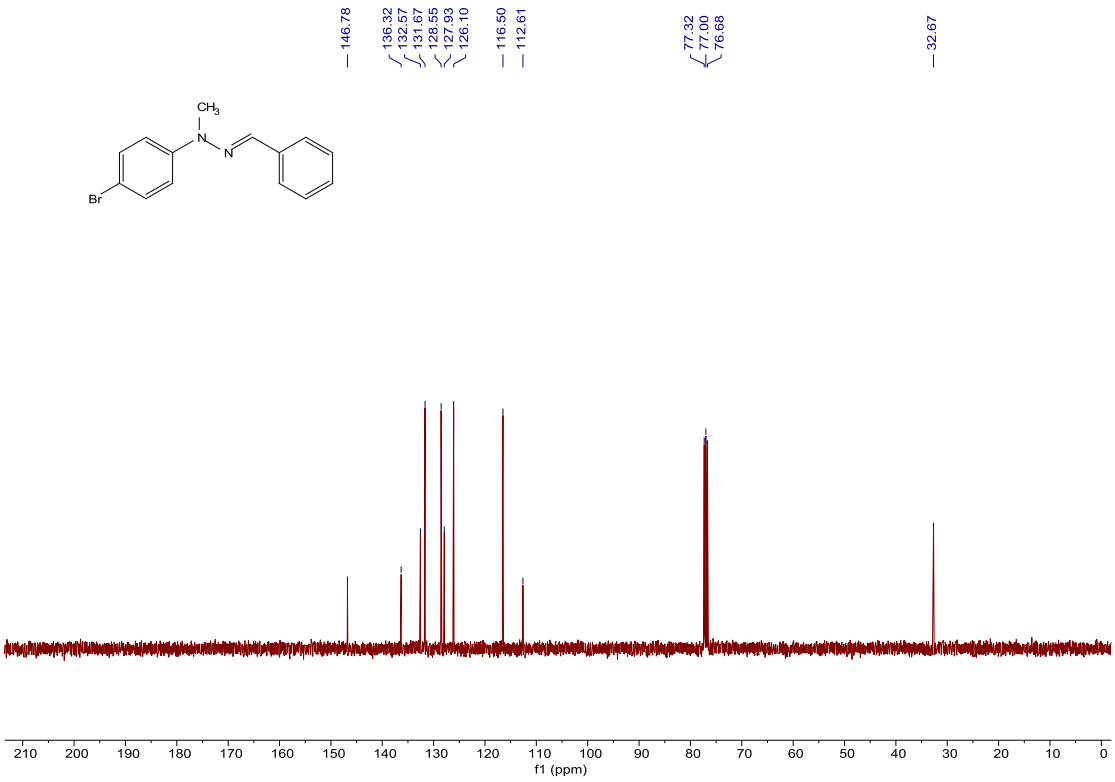
^{1}H NMR of 81 in $CDCl_3$ (400 MHz)



^{13}C{^{1}H} NMR of 81 in $CDCl_3$ (100 MHz)



^{1}H NMR of 83 in $CDCl_3$ (400 MHz)



^{13}C{^{1}H} NMR of 83 in $CDCl_3$ (100 MHz)