Supplementary Information

An aerobic copper-catalyzed multi-component reaction strategy for *N'*,*N'*-diaryl acylhydrazines synthesis: reactions and mechanism

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1.General Methods

Melting points were tested using a digital melting point apparatus and are uncorrected. Infrared (IR) spectra data were measured on an infrared spectrometer using KBr pellets. ^{1}H and $^{13}C\{^{1}H\}$ NMR spectra were performed on a Bruker Advance 400 nuclear magnetic resonance (400 MHz NMR) spectrometer using $CDCl_3$ or DMSO-d_6 as the solution and tetramethylsilane (TMS) as the internal standard. Gas chromatography-mass spectrometry (GC−MS) data were collected using electron ionization. The data of high resolution mass spectrometry (HRMS) were recorded on a high-resolution mass spectrometer (LCMS-IT-TOF). The crystal data were recorded on a diffractometer (Rigaku Oxford diffraction supernova dual source, Gu at zero) equipped with an AtlasS_2 charge-coupled device using Cu Kα radiation (1.54178 Å) in a scan mode. Thin-layer chromatography (TLC) and column chromatography were performed on commercially available 100−400 mesh silica gel. The starting materials, including aldehydes and aryl hydrazines were purchased from Innochem (Beijing) Technology Co., Ltd. of China. Unless otherwise noted, all purchased chemicals were used without further purification.

2. Representative Procedure for the Synthesis of *N′*,*N′*-Diaryl Acylhydrazines.

In a flame-dried test tube with a stir bar, benzaldehyde **1** (31.8 mg, 0.30 mmol), phenylhydrazine **2** (21.6 mg, 0.20 mmol), $Cu(OTf)_2$ (28.9 mg, 0.08 mmol), and K_2HPO_4 (52.2 mg, 0.30 mmol) were added into CH_3CN (2.0 mL). The reaction was performed at 0 ℃ in under an air atmosphere for 12 h (monitored by TLC). After the completion of the reaction, the solvent was evaporated and then filtered through an inch of silica gel. The filtrate was concentrated and purified by chromatography on a silica gel using petroleum ether/ethyl acetate (v/v = 10/1) as an eluent to provide the desired product **3** (21.7 mg, yield of 75%).

For the gram-scale synthesis of **3**, to a solution of benzaldehyde **1** (1.590 g, 15.0 mmol), phenylhydrazine **2** (1.080 g, 10.0 mmol), $Cu(OTf)_2$ (1,445g, 4 mmol), and K_2HPO_4 (2.610 g, 15.0 mmol) in 100 mL of CH_3CN was added under air atmosphere. The solution was stirred at 0 ℃ for 12 h. The mixture was concentrated and purified

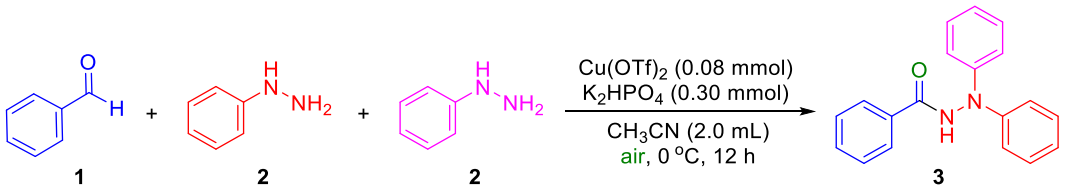
by flash column chromatography on silica gel using the mixture of petroleum ether/ ethyl acetate (PE/EA = 10/1) as an eluent to provide the desired compound **3** (0.710 g, yield of 49%).

3. Representative Procedure for the Synthesis of 64-66.

In a 25 mL test tube with a stir bar, **3** (28.8 mg, 0.1 mmol), bromides (0.2 mmol), Cs_2CO_3 (65.2 mg, 0.2 mmol) were added into CH_3CN (2.0 mL) at room temperature. The reaction was performed at room temperature for 12 hours. After completion of the reaction (monitored by TLC), water (10 mL) was added to the reaction mixture, and the resulting mixture was extracted with ethyl acetate (5 mL×3). The combined organic layers were then dried over $MgSO_4$, filtered, and concentrated in vacuum. The residue was purified by flash chromatography on silica gel using the mixture of petroleum ether/ ethyl acetate (PE/EA = 10/1) to provide the desired product.

4. Optimization of the Reaction Conditions

Table S1. The Effect of the Amount of Benzaldehyde for 3[a]

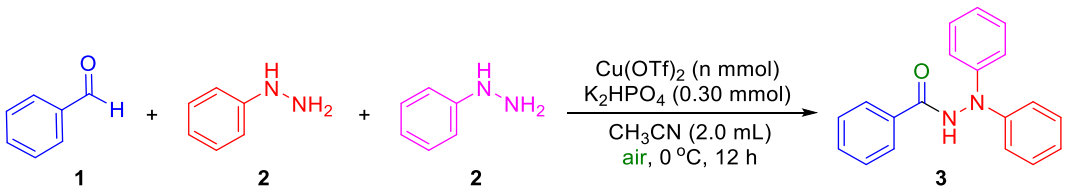


1 2 2 3

entry	**1** (x mmol)	yield of **3** (%)[b]
1	0.30	75
2	0.20	70
3	0.10	45
4	0.05	53

[a]Reaction conditions: **1** (x mmol), **2** (0.20 mmol), $Cu(OTf)_2$ (0.08 mmol), and K_2HPO_4 (0.30 mmol) in CH_3CN (2.0 mL) at 0 ℃ for 12 h under an air atmosphere. [b] Isolated yields.

Table S2. The Effect of the Amount of $Cu(OTf)_2$ for 3[a]

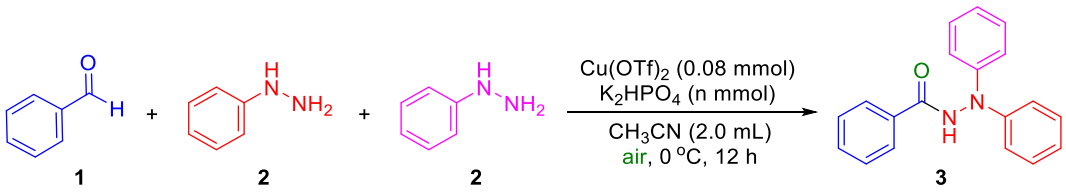


1 2 2 3

entry	$Cu(OTf)_2$ (x mmol)	yield of **3** (%)[b]
1	0.10	75
2	0.08	75
3	0.06	65
4	0.04	50
5	0.02	45

[a]Reaction conditions: **1** (0.30 mmol), **2** (0.20 mmol), $Cu(OTf)_2$ (x mmol), and K_2HPO_4 (0.30 mmol) in CH_3CN (2.0 mL) at 0 ℃ for 12 h under an air atmosphere. [b] Isolated yields.

Table S3. The Effect of the the Amount of K_2HPO_4 for 3[a]

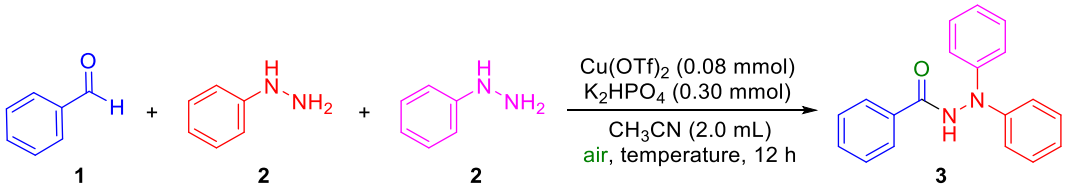


1 2 2 3

entry	K_2HPO_4 (x mmol)	yield of **3** (%)[b]
1	0.40	75
2	0.30	75
3	0.20	70
4	0.10	61

[a]Reaction conditions: **1** (0.30 mmol), **2** (0.20 mmol), $Cu(OTf)_2$ (0.08 mmol), and K_2HPO_4 (x mmol) in CH_3CN (2.0 mL) at 0 ℃ for 12 h under an air atmosphere. [b]Isolated yields.

Table S4. The Effect of the Reaction Temperature for 3[a]

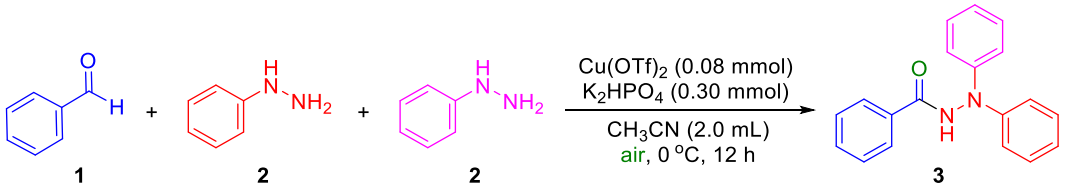


1 2 2 3

entry	T (°C)	yield of **3** (%)[b]
1	20	67
2	10	70
3	0	75
4	- 10	75

[a]Reaction conditions: **1** (0.30 mmol), **2** (0.20 mmol), $Cu(OTf)_2$ (0.08 mmol), and K_2HPO_4 (0.30 mmol) in CH_3CN (2.0 mL) for 12 h under an air atmosphere. [b]Isolated yields.

Table S5. The Effect of the Reaction Time for 3[a]



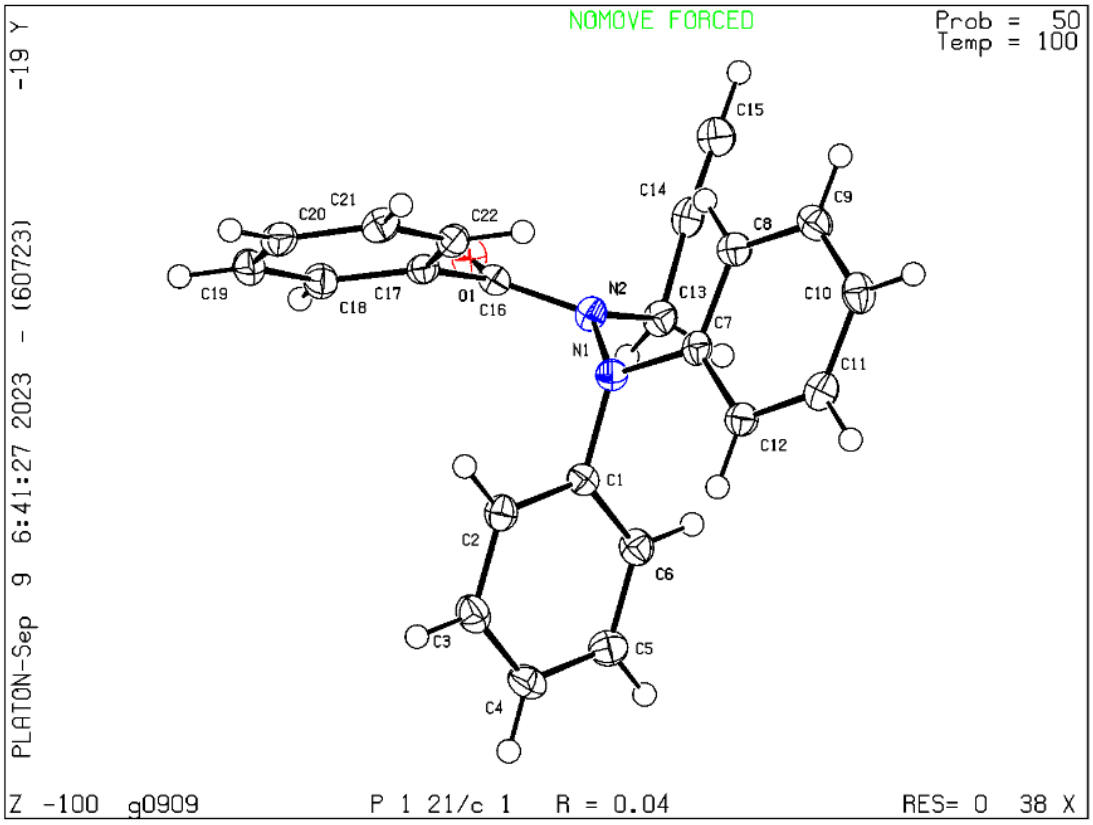
1 2 2 3

entry	Reaction time (h)	yield of **3** (%)[b]
1	6	65
2	12	75
3	18	75
4	24	75

[a]Reaction conditions: **1** (0.30 mmol), **2** (0.20 mmol), $Cu(OTf)_2$ (0.08 mmol), and K_2HPO_4 (0.30 mmol) in CH_3CN (2.0 mL) at 0 ℃ under an air atmosphere. [b]Isolated yields.

5. X-Ray Crystallography Data of 66

The crystal growth procedure: Compound 66 (25 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 provide crystal 66. The ellipsoid contour % probability is 50%.



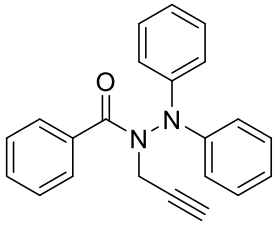


Figure S1. The Crystal Structure of 66

The CCDC number of **66** is 2294001, the detail information please see 66.cif document.

checkCIF/PLATON report

Structure factors have been supplied for datablock(s) g0909

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: g0909

```
Bond precision:   C-C = 0.0016 A          Wavelength=1.54184

Cell:             a=9.6382(1)     b=8.9234(1)         c=19.9546(2)
                  alpha=90        beta=99.063(1)      gamma=90
Temperature:      100 K

                 Calculated                Reported
Volume           1694.78(3)                1694.78(3)
Space group      P 21/c                    P 1 21/c 1
Hall group       -P 2ybc                   -P 2ybc
Moiety formula   C22 H18 N2 O              C22 H18 N2 O
Sum formula      C22 H18 N2 O              C22 H18 N2 O
Mr               326.38                    326.38
Dx,g cm-3        1.279                     1.279
Z                4                         4
Mu (mm-1)        0.623                     0.623
F000             688.0                     688.0
F000'            689.92
h,k,lmax         12,11,24                  12,10,24
Nref             3447                      3353
Tmin,Tmax        0.914,0.940               0.888,1.000
Tmin'            0.911

Correction method= # Reported T Limits: Tmin=0.888 Tmax=1.000
AbsCorr = MULTI-SCAN

Data completeness= 0.973           Theta(max)= 74.259

R(reflections)= 0.0353( 3051)                wR2(reflections)=
                                             0.0933( 3353)
S = 1.031                  Npar= 226
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The following ALERTS were generated. Each ALERT has the format
test-name_ALERT_alert-type_alert-level.
Click on the hyperlinks for more details of the test.

Alert level G

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PLAT230_ALERT_2_G Hirshfeld Test Diff for    C13      --C14      .        7.1 s.u.
PLAT912_ALERT_4_G Missing # of FCF Reflections Above STh/L=  0.600         84 Note
PLAT941_ALERT_3_G Average HKL Measurement Multiplicity ..........        2.6 Low
PLAT978_ALERT_2_G Number C-C Bonds with Positive Residual Density.         7 Info
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0 **ALERT level A** = Most likely a serious problem - resolve or explain
0 **ALERT level B** = A potentially serious problem, consider carefully
0 **ALERT level C** = Check. Ensure it is not caused by an omission or oversight
4 **ALERT level G** = General information/check it is not something unexpected

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

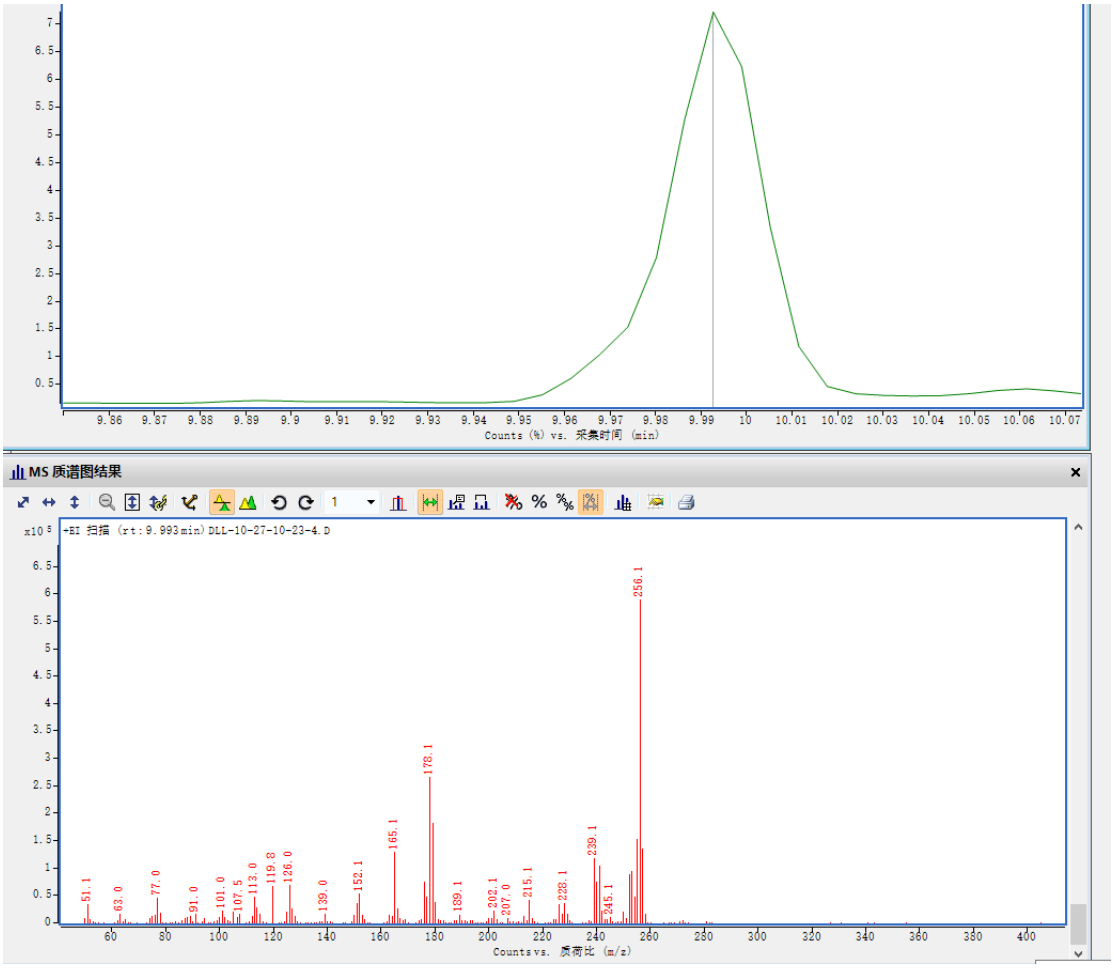
Publication of your CIF in IUCr journals

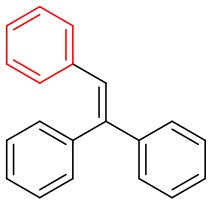
A basic structural check has been run on your CIF. These basic checks will be run on all CIFs submitted for publication in IUCr journals (*Acta Crystallographica*, *Journal of Applied Crystallography*, *Journal of Synchrotron Radiation*); however, if you intend to submit to *Acta Crystallographica Section 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.

6. GC-MS/HRMS of 67





67

Figure S2. GC-MS of 67

(RT = 9.95-10.02 min, MS (EI, 70 eV) *m/z*: 256, 239, 178, 165, 77.)

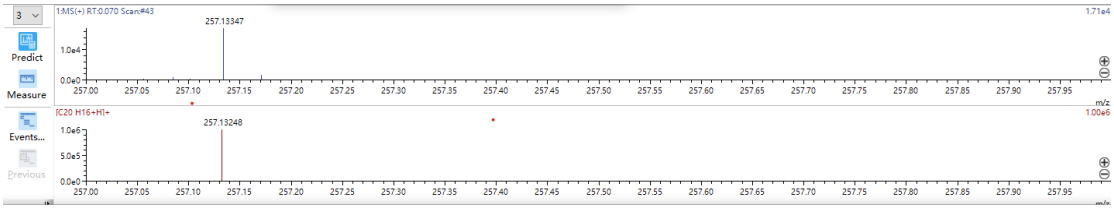
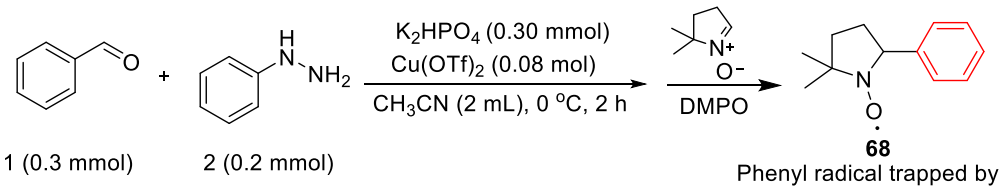


Figure S3. HRMS of 67

HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{20}H_{17}$ 257.13248, found 257.13347.

7. Electron Paramagnetic Resonance (EPR) Experiments



In a flame-dried test tube with a stir bar, benzaldehyde **1** (31.8 mg, 0.30 mmol), phenylhydrazine **2** (21.6 mg, 0.20 mmol), $Cu(OTf)_2$ (28.9 mg, 0.08 mmol), and K_2HPO_4 (52.2 mg, 0.30 mmol) were added into CH_3CN (1.0 mL). The reaction was performed at 0 ℃ in under an air atmosphere for 2 h. Then, DMPO (50 μL) was added into the reaction mixture and stirred for another 1 min. Next, the solution sample was analyzed by EPR. We propose that this free radical signal belongs to the same carbon free radical (g = 2.00598, A_N = 14.70 G, A_H = 21.70 G). Meanwhile, DMPO-benzene adducts **68** was confirmed by HRMS.

When the experiment was carried out in the absence of copper catalyst, the signal of adducts **68** became weaken (g = 2.00595, A_N = 14.80 G, A_H = 21.80 G), suggesting that copper catalyst could promote the generation of phenyl radical intermediates.

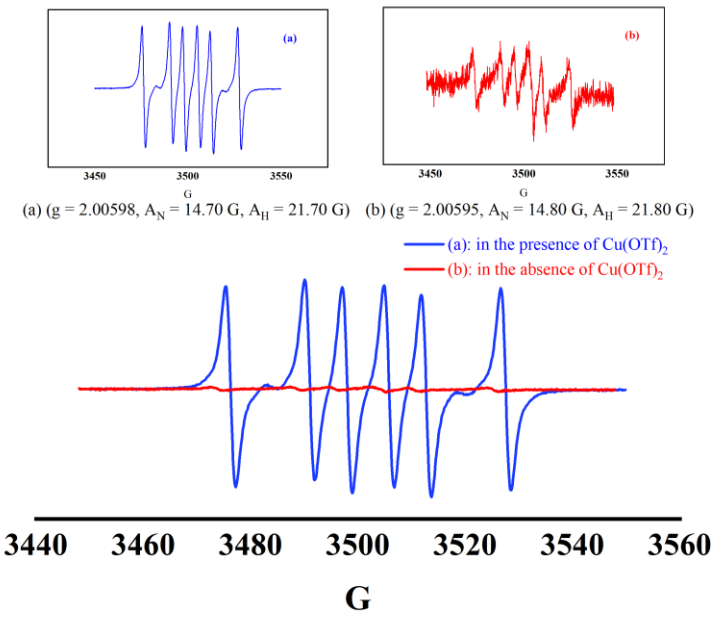
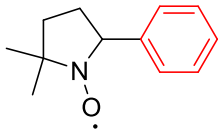


Figure S4. EPR Experiments with Spin-Trapping Reagents for the Detection of Phenyl Radicals

8. HRMS of 68

Formula Predictor Result	C12 H16 N O
Mass	190.123
Error Margin	20 ppm
DBE Range	Not Used
Electron Ions	Both configurations
HC Ratio	Not Used
Nitrogen Rule	Not Used

#	Score	Pred. (M)	Pred. m/z	Meas. m/z	Diff. (mDa)	Formulae (M)	Ion	Diff. (ppm)	Iso Score	DBE
2	92.03	190.12319	190.12264	190.12300	0.36	C12 H16 N O	[M]+	1.894	92.25	5.5



68 (HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $C_{12}H_{17}NO$ 190.1226, found 190.1230)

Figure S5. HRMS of 68

9. GC-MS of 70, 71, 72, 73

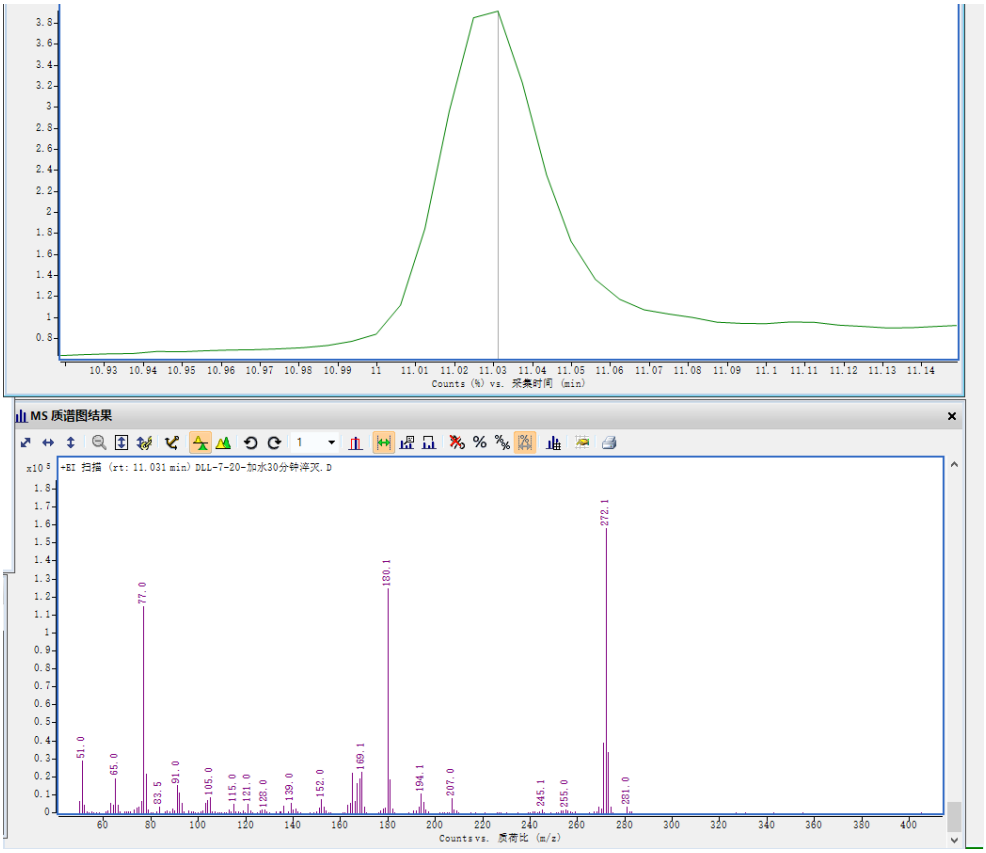
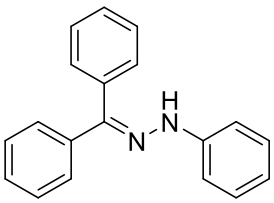


Figure S6. GC-MS of 70



70: (RT = 11.00-11.08 min, MS (EI, 70 eV) *m/z*: 272, 194, 180, 169, 77.)

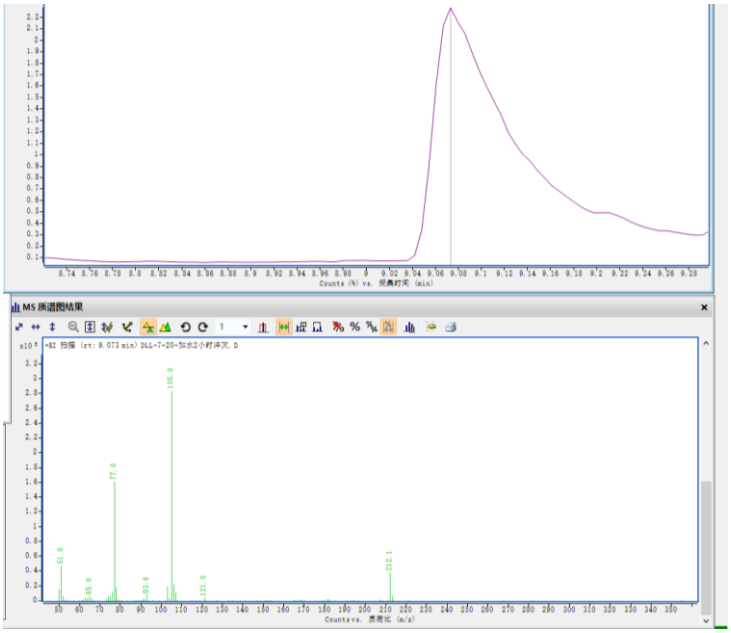
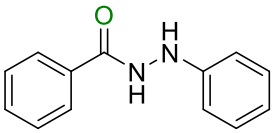


Figure S7. GC-MS of 71



71: (RT = 9.04-9.20 min, MS (EI, 70 eV) *m/z*: 212, 121, 105, 77, 51.)

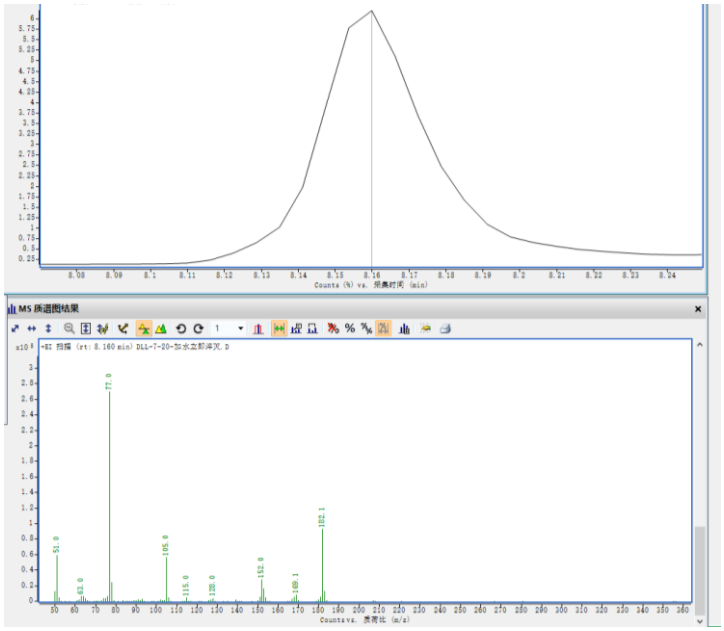
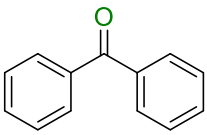


Figure S8. GC-MS of 72



72: (RT = 8.13-8.19 min, MS (EI, 70 eV) *m/z*: 182, 169, 152, 105, 77.)

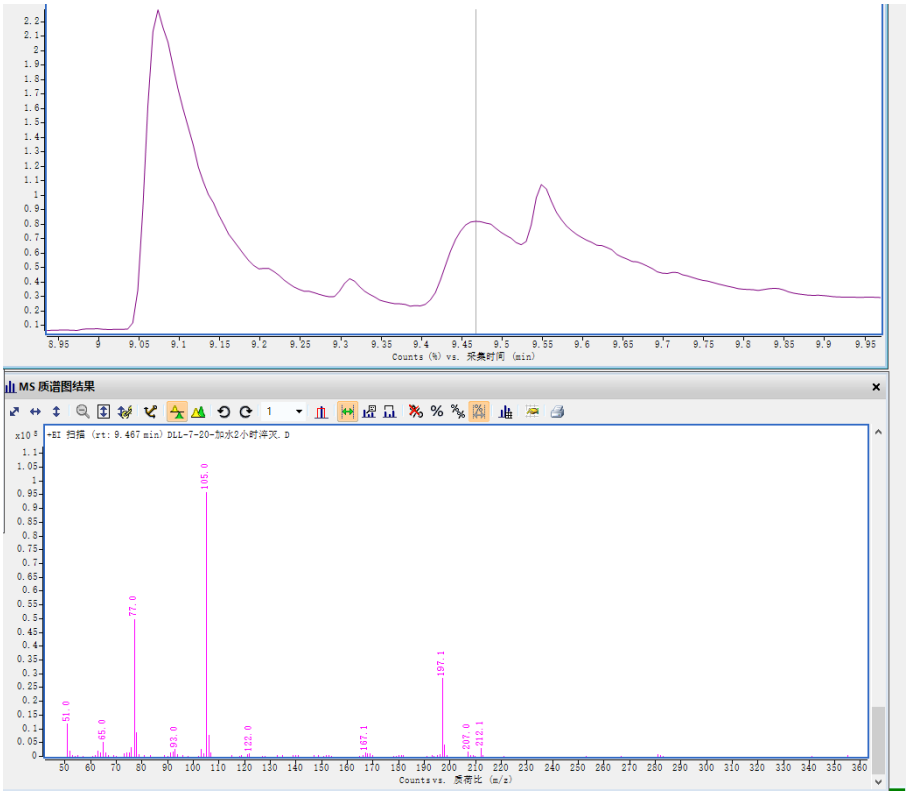
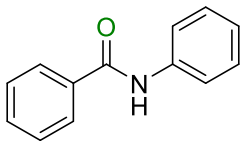
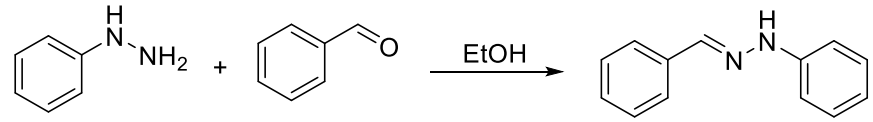


Figure S9. GC-MS of 73



73: (RT = 9.40-9.52 min, MS (EI, 70 eV) *m/z*: 197, 105, 77, 65, 51.)

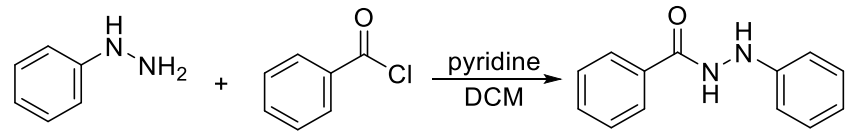
10. Preparation of (*E*)-1-Benzylidene-2-phenylhydrazine (69)



phenylhydrazine benzaldehyde **69**

In a flame-dried test tube with a stir bar, phenylhydrazine (108 mg, 1.0 mmol) and benzaldehyde (106 mg, 1.0 mmol) were added into anhydrous ethanol (10 mL) at 0 ℃. After the completion of the reaction, the precipitate was filtrated, washed with water (3 × 5 mL) and dried to provide the desired product **69** (195.0 mg, yield of 99%).

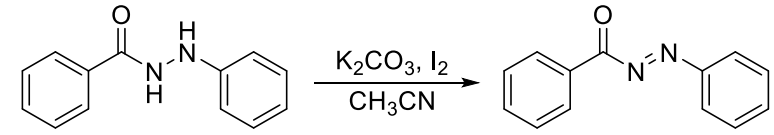
11. Preparation of *N'*-Phenylbenzohydrazide (71)



phenylhydrazine benzoyl chloride **71**

In a flame-dried test tube with a stir bar, phenylhydrazine (108 mg, 1.0 mmol), benzoyl chloride (140 mg, 1.0 mmol), and pyridine (158 mg, 2.0 mmol) were added into dichloromethane (10 mL) at 0 ℃. After the completion of the reaction, the precipitate was filtrated, washed with petroleum ether (2 × 5 mL) and dried to provide the desired product 71 (170.0 mg, yield of 80%).

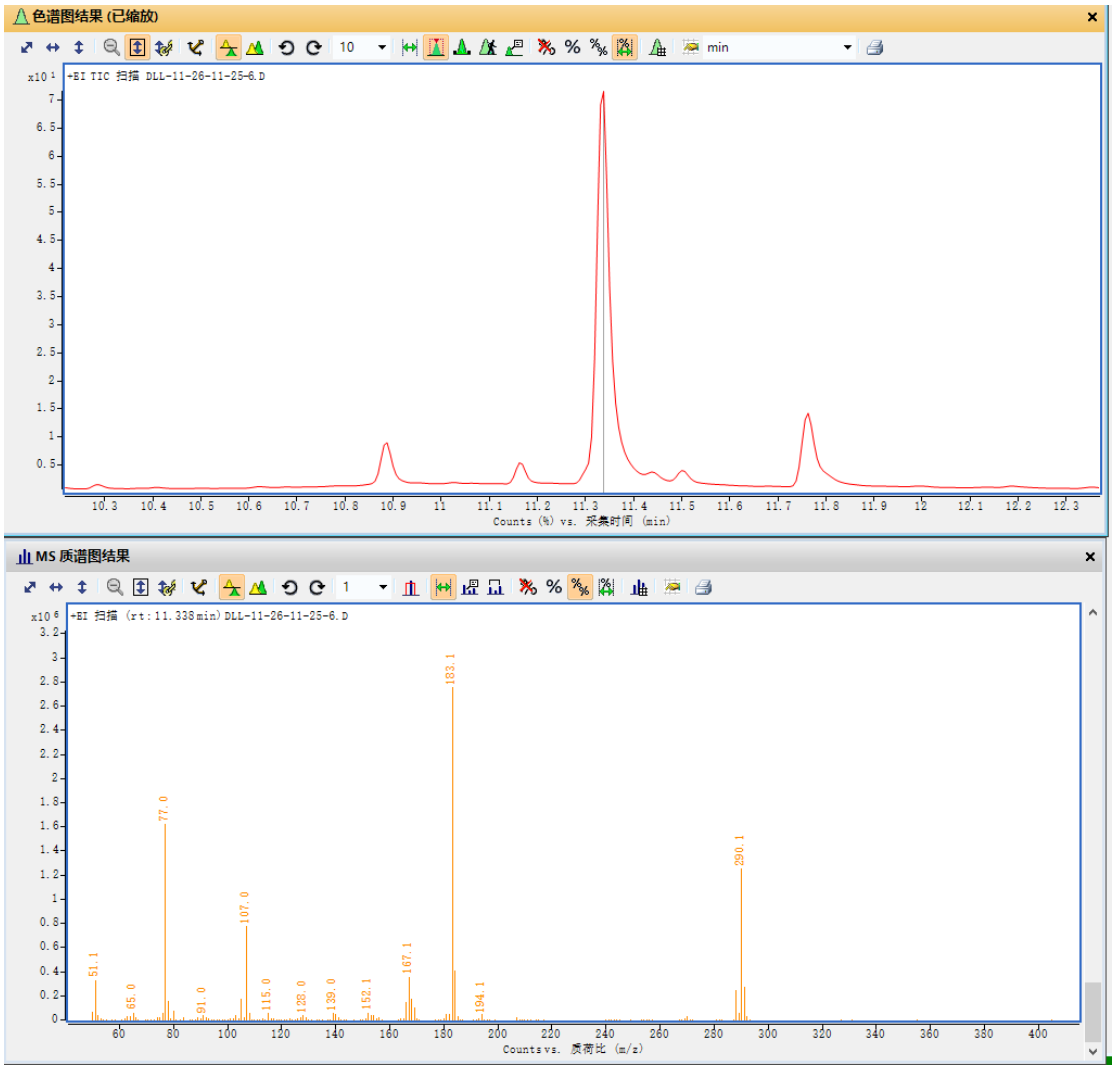
12. Preparation of (*E*)-phenyl(phenyldiazenyl)methanone (74)



N'-phenylbenzohydrazide

In a flame-dried test tube with a stir bar, *N'*-phenylbenzohydrazide (106 mg, 0.5 mmol), K_2CO_3 (207 mg, 1.5 mmol), I_2 (152 mg, 0.6 mmol) were added into CH_3CN (5 mL). The reaction was performed at room temperature for 40 min, then quenched with 5% $Na_2S_2O_3$ (5 mL), diluted with brine (10 mL), and extracted with ethyl acetate (3 × 15 mL). The combined organic layer was dried over anhydrous Na_2SO_4, concentrated, and purified through silica gel column chromatography using petroleum ether/ethyl acetate (v/v = 10/1) as an eluent to afford the desired product **74** (100.8 mg, yield of 96%).

13. GC-MS/HRMS of ^{18}O-3



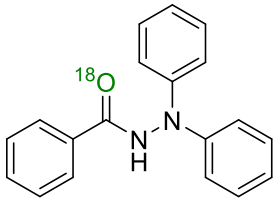
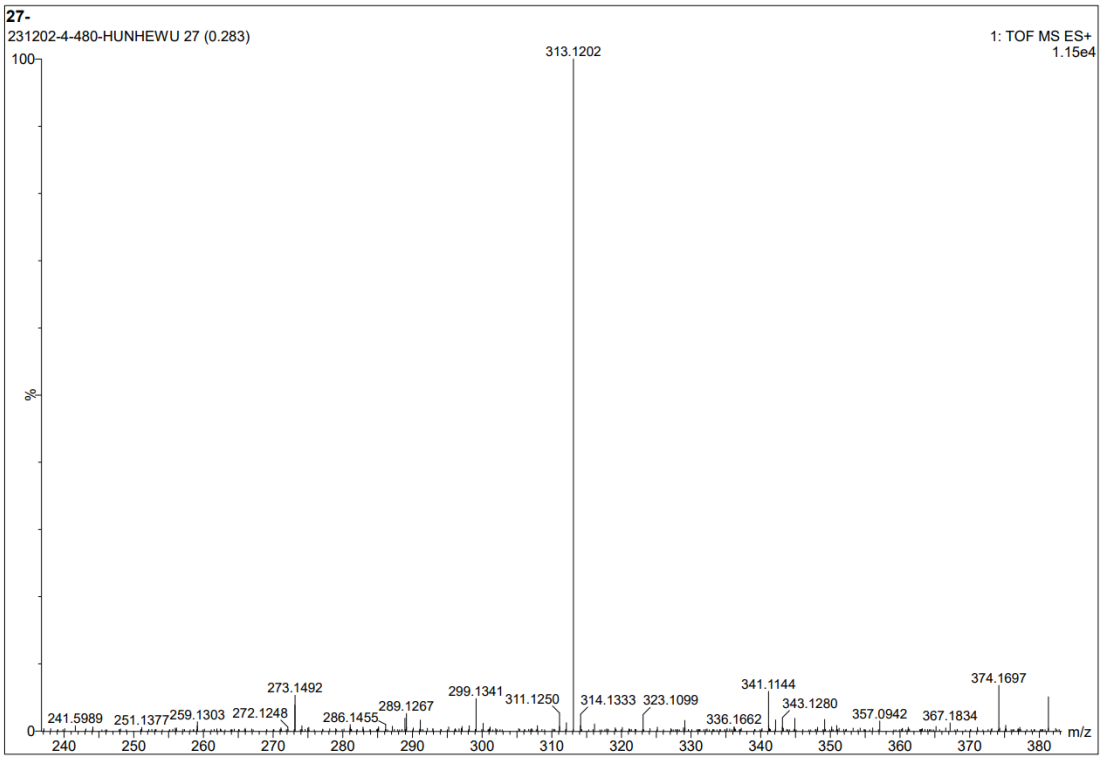


Figure S10. GC-MS of ^{18}O-3

(RT = 11.30-11.40 min, MS (EI, 70 eV) *m/z*: 290, 183, 167, 107, 77.)



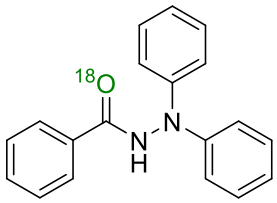
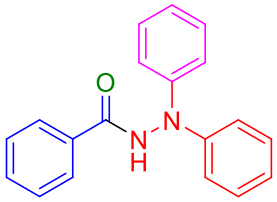


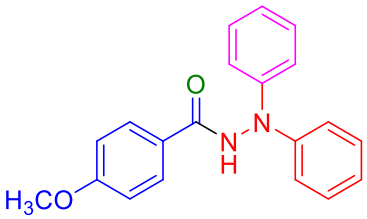
Figure S11. HRMS of ^{18}O-3

(HRMS: *m/z* [M + Na]$^+$ calcd for $C_{19}H_{16}N_2Na^{18}O^+$ 313.1197, found 313.1202.)

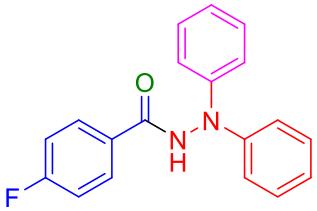
14. Characterization Data



*N',N'-Diphenylbenzohydrazide (**3**).*[1] Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 75% yield (21.7 mg, 0.08 mmol); mp 185-186 ℃; IR (KBr, cm^{-1}) 3267, 3059, 1656, 1589, 1523, 1495, 1311, 1271, 1028, 887, 747, 691; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 11.24 (s, 1H), 7.96 (d, J = 8.0 Hz, 2H), 7.61 (t, J = 8.0 Hz, 1H), 7.53 (t, J = 8.0 Hz, 2H), 7.31 (t, J = 8.0 Hz, 4H), 7.18 (d, J = 8.0 Hz, 4H), 6.99 (t, J = 8.0 Hz, 2H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 166.3, 146.2, 132.9, 132.5, 129.5, 129.1, 127.9, 122.6, 119.2; MS (EI, 70 eV) *m/z* 288, 183, 167, 105, 77.

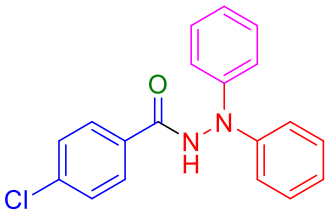


*4-Methoxy-N',N'-diphenylbenzohydrazide (**4**).* Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 69% yield (22.0 mg, 0.07 mmol); mp 146-147 ℃; IR (KBr, cm^{-1}) 3293, 3058, 2932, 2835, 1654, 1605, 1588, 1495, 1335, 1315, 1247, 1178, 1028, 888, 747; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 11.11 (s, 1H), 7.95 (s, 2H), 7.30 (t, J = 8.0 Hz, 4H), 7.17 (s, 4H), 7.07 (d, J = 8.0 Hz, 2H), 6.98 (t, J = 8.0 Hz, 2H), 3.83 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.8, 162.7, 146.3, 130.2, 129.5, 125.0, 122.5, 119.1, 114.3, 55.9; MS (EI, 70 eV) *m/z* 318, 183, 167, 135, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{20}H_{19}N_2O_2$ 319.1441, found 319.1460.

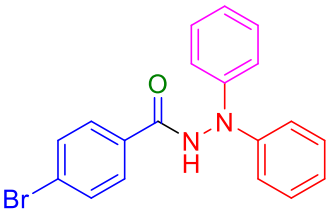


*4-Fluoro-N',N'-diphenylbenzohydrazide (**5**).* Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 75% yield (23.0 mg, 0.08 mmol); mp 123-124 ℃; IR (KBr, cm^{-1}) 3261, 3060, 1655, 1590, 1498, 1321, 1237, 1055, 1028, 853, 750; ^{1}H

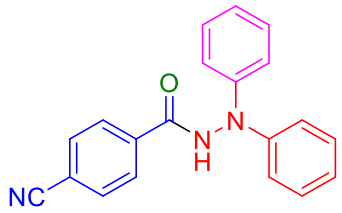
NMR (400 MHz, DMSO- d_6, ppm) δ 11.27 (s, 1H), 8.07-8.03 (m, 2H), 7.38 (t, J = 12.0 Hz, 2H), 7.31 (t, J = 8.0 Hz, 4H), 7.18 (d, J = 8.0 Hz, 4H), 7.00 (t, J = 8.0 Hz, 2H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.2, 164.8 (d, J = 248.0 Hz), 146.2, 130.7 (d, J = 10.0 Hz), 129.6, 129.4 (d, J = 3.0 Hz), 122.7, 119.2, 116.1 (d, J = 21.0 Hz); MS (EI, 70 eV) *m/z* 306, 183, 167, 123, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{19}H_{16}N_2OF$ 307.1241, found 307.1257.



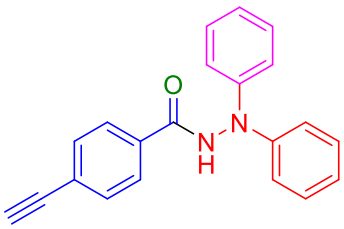
4-Chloro-N',N'-diphenylbenzohydrazide ***(6)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 66% yield (21.3 mg, 0.07 mmol); mp 134-135 ℃; IR (KBr, cm^{-1}) 3268, 3049, 1657, 1591, 1495, 1316, 1271, 1026, 847, 747; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 11.30 (s, 1H), 7.97 (d, J = 8.0 Hz, 2H), 7.62 (d, J = 8.0 Hz, 2H), 7.31 (t, J = 8.0 Hz, 4H), 7.16 (d, J = 8.0 Hz, 4H), 7.00 (t, J = 8.0 Hz, 2H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.3, 146.1, 137.4, 131.7, 129.9, 129.6, 129.2, 122.7, 119.2; MS (EI, 70 eV) *m/z* 322, 183, 169, 139,77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{19}H_{16}N_2OCl$ 323.0946, found 323.0963.



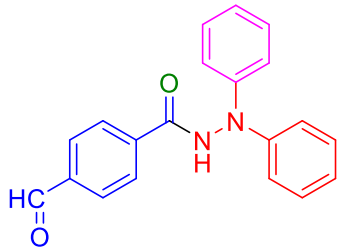
4-Bromo-N',N'-diphenylbenzohydrazide ***(7)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 66% yield (24.2 mg, 0.07 mmol); mp 210-211 ℃; IR (KBr, cm^{-1}) 3265, 3038, 1658, 1589, 1494, 1480, 1335, 1271, 1074, 886, 745; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 11.31 (s, 1H), 7.89 (d, J = 8.0 Hz, 2H), 7.75 (d, J = 8.0 Hz, 2H), 7.31 (t, J = 8.0 Hz, 4H), 7.16 (d, J = 8.0 Hz, 4H), 7.00 (t, J = 8.0 Hz, 2H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.5, 146.1, 132.1, 132.1, 130.0, 129.6, 126.3, 122.7, 119.2; MS (EI, 70 eV) *m/z* 368, 183, 169, 139, 77; HRMS (ESI) *m/z* $[M + Na]^+$ calcd for $C_{19}H_{15}N_2OBrNa$ 389.0260, found 389.0281.



4-Cyano-N',N'-diphenylbenzohydrazide ***(8)***. Eluent, petroleum ether/ethyl acetate (v/v = 3/1); light yellow solid in 72% yield (22.5 mg, 0.07 mmol); mp 251-252 ℃; IR (KBr, cm^{-1}) 3423, 3262, 3059, 2231, 1661, 1590, 1496, 1335, 1275, 1027, 858, 748; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 11.48 (s, 1H), 8.11 (d, J = 8.0 Hz, 2H), 8.02 (d, J = 8.0 Hz, 2H), 7.32 (t, J = 8.0 Hz, 4H), 7.18 (d, J = 8.0 Hz, 4H), 7.02 (t, J = 12.0 Hz, 2H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.1, 146.0, 137.0, 133.2, 129.6, 128.8, 122.9, 119.3, 118.7, 114.8; MS (EI, 70 eV) *m/z* 313, 183, 169, 139, 77; HRMS (ESI) *m/z* $[M + Na]^+$ calcd for $C_{20}H_{15}N_3ONa$ 336.1107, found 336.1132.

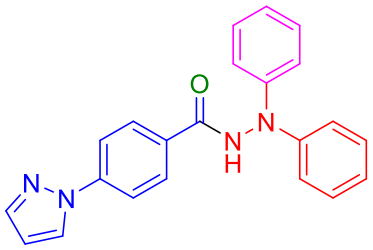


4-Ethynyl-N',N'-diphenylbenzohydrazide ***(9)***. Eluent, petroleum ether/ethyl acetate (v/v = 3/1); yellow solid in 30% yield (9.4 mg, 0.03 mmol); mp 214-215 ℃; IR (KBr, cm^{-1}) 3279, 3060, 3004, 2100, 1655, 1590, 1495, 1307, 1076, 893, 748, 692; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 11.30 (s, 1H), 7.96 (d, J = 8.0 Hz, 2H), 7.64 (d, J = 8.0 Hz, 2H), 7.31 (t, J = 8.0 Hz, 4H), 7.17 (d, J = 8.0 Hz, 4H), 7.00 (t, J = 8.0 Hz, 2H), 4.40 (s, 1H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.6, 146.1, 133.4, 132.4, 129.6, 128.3, 125.7, 122.7, 119.2, 83.7, 83.2; MS (EI, 70 eV) *m/z* 312, 183, 167, 129, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{21}H_{17}N_2O$ 313.1335, found 313.1343.

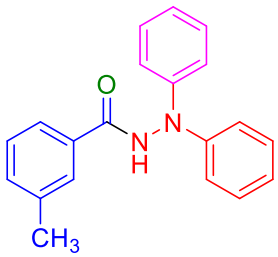


4-Formyl-N',N'-diphenylbenzohydrazide ***(10)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 51% yield (16.1 mg, 0.05 mmol); mp 126-127 ℃;

IR (KBr, cm^{-1}) 3420, 3041, 1704, 2850, 1658, 1495, 1323, 1206, 1026, 829, 751, 694; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 11.42 (s, 1H), 10.12 (s, 1H), 8.13 (d, J = 8.0 Hz, 2H), 8.06 (d, J = 8.0 Hz, 2H), 7.32 (t, J = 8.0 Hz, 4H), 7.18 (d, J = 4.0 Hz, 4H), 7.01 (t, J = 8.0 Hz, 2H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 193.4, 165.4, 146.1, 138.8, 137.9, 130.1, 129.6, 128.7, 122.8, 119.3; MS (EI, 70 eV) *m/z* 316, 183, 169, 133, 77; HRMS (ESI) *m/z* $[M + Na]^+$ calcd for $C_{20}H_{16}N_2O_2Na$ 339.1104, found 339.1125.

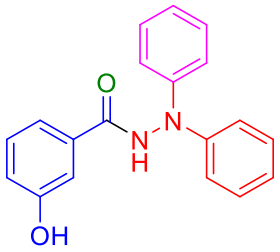


N',N'-Diphenyl-4-(1H-pyrazol-1-yl)benzohydrazide ***(11)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 65% yield (23.0 mg, 0.07 mmol); mp 140-141 ℃; IR (KBr, cm^{-1}) 3397, 3274, 3059, 1655, 1611, 1590, 1495, 1395, 1291, 1205, 1026, 937, 849, 746; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 11.28 (s, 1H), 8.64 (s, 1H), 8.09 (d, J = 8.0 Hz, 2H), 8.02 (d, J = 8.0 Hz, 2H), 7.82 (s, 1H), 7.31 (t, J = 8.0 Hz, 4H), 7.18 (d, J = 8.0 Hz, 4H), 7.00 (t, J = 8.0 Hz, 2H), 6.61 (s, 1H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.5, 146.2, 142.6, 142.3, 130.1, 129.6, 128.7, 122.6, 119.2, 118.4, 111.0, 109.0; MS (EI, 70 eV) *m/z* 354, 183, 171, 143, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{22}H_{19}N_4O$ 355.1553, found 315.1571.

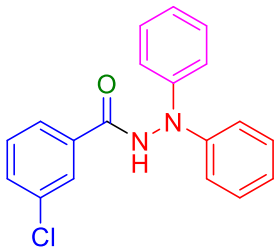


3-Methyl-N',N'-diphenylbenzohydrazide ***(12)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 86% yield (26.0 mg, 0.09 mmol); mp 147-148 ℃; IR (KBr, cm^{-1}) 3443, 3254, 3037, 2923, 2855, 1660, 1589, 1493, 1315, 1277, 1026, 934, 830, 748, 693; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 11.18 (s, 1H), 7.79-7.75 (m, 2H), 7.42 (d, J = 4.0 Hz, 2H), 7.31 (t, J = 8.0 Hz, 4H), 7.17 (d, J = 8.0 Hz, 4H), 6.99 (t, J = 8.0 Hz, 2H), 2.39 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 166.4, 146.2, 138.4, 133.02, 132.96, 129.5, 129.0, 128.5, 125.1, 122.6, 119.2, 21.4;

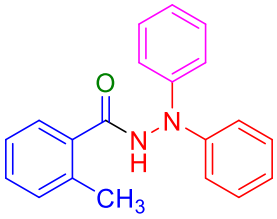
MS (EI, 70 eV) *m/z* 302, 183, 169, 119, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{20}H_{19}N_2O$ 303.1492, found 303.1513.



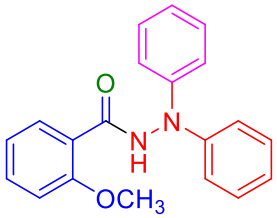
3-Hydroxy-N',N'-diphenylbenzohydrazide ***(13)***. Eluent, petroleum ether/ethyl acetate (v/v = 3/1); light yellow oily liquid in 50% yield (15.2 mg, 0.05 mmol); IR (KBr, cm^{-1}) 3259, 3037, 1655, 1588, 1494, 1308, 1219, 1079, 998, 847, 748, 691; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 11.13 (s, 1H), 9.84 (s, 1H), 7.39 (d, J = 8.0 Hz, 2H), 7.35-7.29 (m, 6H), 7.16 (d, J = 8.0 Hz, 4H), 6.99 (t, J = 8.0 Hz, 2H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 166.5, 158.0, 146.2, 134.4, 130.2, 129.5, 122.6, 119.5, 119.2, 118.4, 114.9; MS (EI, 70 eV) *m/z* 304, 183, 169, 121, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{19}H_{17}N_2O_2$ 305.1285, found 305.1307.



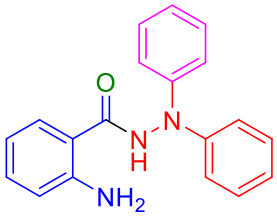
3-Chloro-N',N'-diphenylbenzohydrazide ***(14)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 81% yield (26.1 mg, 0.08 mmol); mp 181-182 ℃; IR (KBr, cm^{-1}) 3247, 3026, 1660, 1590, 1494, 1312, 1246, 1079, 926, 893, 745, 691; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 11.36 (s, 1H), 8.02 (s, 1H), 7.93 (d, J = 8.0 Hz, 1H), 7.68 (d, J = 8.0 Hz, 1H), 7.57 (t, J = 8.0 Hz, 1H), 7.32 (t, J = 8.0 Hz, 4H), 7.19 (d, J = 8.0 Hz, 4H), 7.01 (t, J = 8.0 Hz, 2H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.0, 146.1, 134.9, 134.0, 132.3, 131.1, 129.6, 127.8, 126.7, 122.8, 119.3; MS (EI, 70 eV) *m/z* 322, 183, 169, 139, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{19}H_{16}N_2OCl$ 323.0946, found 323.0964.



2-Methyl-N',N'-diphenylbenzohydrazide ***(15)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 88% yield (26.6 mg, 0.09 mmol); mp 184-185 ℃; IR (KBr, cm^{-1}) 3217, 3037, 2925, 2849, 1662, 1589, 1493, 1316, 1247, 1157, 899, 747, 693; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.96 (s, 1H), 7.52 (d, J = 8.0 Hz, 1H), 7.42 (t, J = 8.0 Hz, 1H), 7.37-7.31 (m, 6H), 7.23 (d, J = 8.0 Hz, 4H), 7.03 (t, J = 8.0 Hz, 2H), 2.38 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 168.9, 146.3, 136.4, 135.0, 131.2, 130.5, 129.6, 127.7, 126.2, 122.7, 119.2, 19.8; MS (EI, 70 eV) *m/z* 302, 183, 169, 119, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{20}H_{19}N_2O$ 303.1492, found 303.1513.

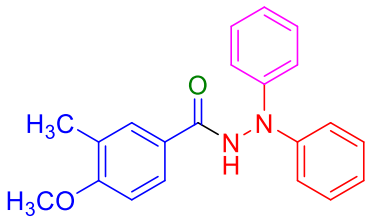


2-Methoxy-N',N'-diphenylbenzohydrazide ***(16)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 65% yield (20.7 mg, 0.07 mmol); mp 121-122 ℃; IR (KBr, cm^{-1}) 3365, 3061, 2921, 2840, 1659, 1600, 1589, 1495, 1296, 1243, 1024, 898, 750, 693; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.72 (s, 1H), 7.61 (d, J = 8.0 Hz, 1H), 7.51 (t, J = 8.0 Hz, 1H), 7.32 (t, J = 8.0 Hz, 4H), 7.23 (d, J = 8.0 Hz, 4H), 7.18 (d, J = 8.0 Hz, 1H), 7.06 (t, J = 8.0 Hz, 1H), 7.00 (t, J = 8.0 Hz, 2H), 3.93 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 166.1, 157.4, 146.2, 132.8, 130.3, 129.4, 123.3, 122.4, 121.0, 119.1, 112.4, 56.3; MS (EI, 70 eV) *m/z* 318, 183, 167, 135, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{20}H_{19}N_2O_2$ 319.1441, found 319.1464.

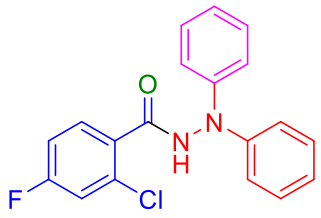


2-Amino-N',N'-diphenylbenzohydrazide ***(17)***. Eluent, petroleum ether/ethyl acetate (v/v = 3/1); white solid in 36% yield (10.9 mg, 0.04 mmol); mp 136-137 ℃;

IR (KBr, cm^{-1}) 3423, 3241, 3061, 1651, 1618, 1589, 1494, 1305, 1157, 1026, 823, 749, 700; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.92 (s, 1H), 7.70 (d, J = 8.0 Hz, 1H), 7.30 (t, J = 8.0 Hz, 4H), 7.22 (t, J = 8.0 Hz, 1H), 7.17 (d, J = 4.0 Hz, 4H), 6.98 (t, J = 4.0 Hz, 2H), 6.76 (d, J = 8.0 Hz, 1H), 6.58 (t, J = 8.0 Hz, 1H), 6.42 (s, 2H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 168.7, 150.7, 146.3, 133.1, 129.5, 128.5, 122.4, 119.1, 117.1, 115.3, 112.5; MS (EI, 70 eV) *m/z* 303, 184, 169, 120, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{19}H_{18}N_3O$ 304.1444, found 304.1422.

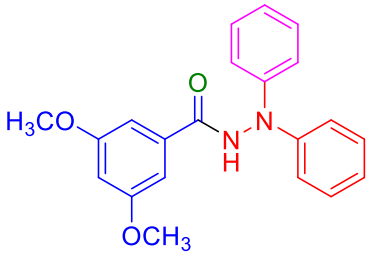


4-Methoxy-3-methyl-N',N'-diphenylbenzohydrazide ***(18)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 67% yield (22.3 mg, 0.07 mmol); mp 169-170 ℃; IR (KBr, cm^{-1}) 3422, 3060, 2922, 2850, 1655, 1607, 1590, 1496, 1259, 1129, 1026, 824, 750, 693; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 11.05 (s, 1H), 7.86 (d, J = 8.0 Hz, 1H), 7.81 (s, 1H), 7.30 (t, J = 8.0 Hz, 4H), 7.18 (d, J = 8.0 Hz, 4H), 7.05 (d, J = 8.0 Hz, 1H), 6.97 (t, J = 8.0 Hz, 2H), 3.86 (s, 3H), 2.21 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 166.0, 160.8, 146.4, 130.2, 129.5, 127.6, 126.2, 124.6, 122.4, 119.1, 110.4, 56.0, 16.5; MS (EI, 70 eV) *m/z* 332, 183, 169, 149, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{21}H_{21}N_2O_2$ 333.1598, found 333.1619.

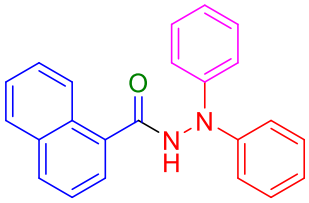


2-Chloro-4-fluoro-N',N'-diphenylbenzohydrazide ***(19)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 57% yield (19.4 mg, 0.06 mmol); mp 139-140 ℃; IR (KBr, cm^{-1}) 3422, 3204, 3037, 1672, 1599, 1492, 1307, 1258, 1151, 1027, 922, 875, 750, 693; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 11.13 (s, 1H), 7.68-7.64 (m, 1H), 7.62-7.59 (m, 1H), 7.40-7.33 (m, 5H), 7.23 (d, J = 8.0 Hz, 4H), 7.04 (t, J = 8.0 Hz, 2H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.5, 163.0 (d, J = 249.0 Hz), 146.0, 132.3 (d, J = 11.0 Hz), 131.8 (d, J = 4.0 Hz), 131.3 (d, J = 9.0 Hz), 129.6, 122.9, 119.4, 117.8 (d, J = 15.0 Hz), 115.1 (d, J = 21.0 Hz); MS (EI, 70 eV) *m/z* 340, 183, 167, 129, 77; HRMS (ESI) *m/z* $[M + Na]^+$ calcd for

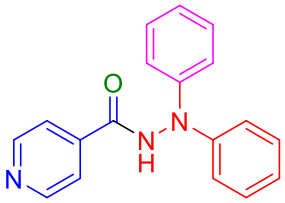
$C_{19}H_{14}N_2OFClNa$ 363.0671, found 363.0689.



3,5-Dimethoxy-N',N'-diphenylbenzohydrazide (***20***). Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 70% yield (24.4 mg, 0.07 mmol); mp 150-151 ℃; IR (KBr, cm^{-1}) 3248, 3039, 2938, 2838, 1658, 1593, 1494, 1457, 1350, 1329, 1206, 1157, 1064, 927, 845, 747, 692; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 11.22 (s, 1H), 7.32 (t, J = 8.0 Hz, 4H), 7.18 (d, J = 8.0 Hz, 6H), 7.00 (t, J = 8.0 Hz, 2H), 6.74 (s, 1H), 3.82 (s, 6H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.8, 161.0, 146.2, 134.8, 129.6, 122.6, 119.2, 105.8, 104.5, 56.0; MS (EI, 70 eV) *m/z* 348, 183, 165, 137, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{21}H_{21}N_2O_3$ 349.1547, found 349.1569.

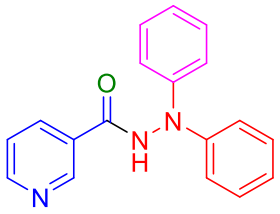


N',N'-Diphenyl-1-naphthohydrazide (***21***). Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 72% yield (24.3 mg, 0.07 mmol); mp 205-206 ℃; IR (KBr, cm^{-1}) 3421, 3213, 3060, 1660, 1590, 1493, 1296, 1201, 1025, 1005, 899, 750, 693; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 11.24 (s, 1H), 8.18 (s, 1H), 8.11 (d, J = 8.0 Hz, 1H), 8.03 (t, J = 8.0 Hz, 1H), 7.83 (s, 1H), 7.65-7.59 (m, 3H), 7.38 (t, J = 8.0 Hz, 4H), 7.31 (s, 4H), 7.05 (t, J = 8.0 Hz, 2H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 168.4, 146.3, 133.7, 132.4, 131.2, 130.5, 129.7, 128.9, 127.6, 127.0, 126.2, 125.5, 125.3, 122.8, 119.3; MS (EI, 70 eV) *m/z* 338, 183, 155, 127, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{23}H_{19}N_2O$ 339.1492, found 339.1510.

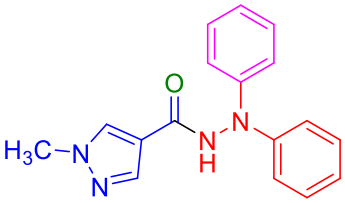


N',N'-Diphenylisonicotinohydrazide (***22***). Eluent, petroleum ether/ethyl acetate (v/v = 3/1); white solid in 70% yield (20.2 mg, 0.07 mmol); mp 112-113 ℃; IR (KBr, cm^{-1}) 3252, 3041, 1663, 1588, 1523, 1494, 1314, 1276, 1063, 901, 749, 692; ^1H

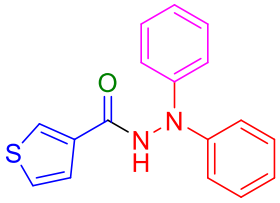
NMR (400 MHz, DMSO-d_6, ppm) δ 11.53 (s, 1H), 8.80 (d, J = 8.0 Hz, 2H), 7.86 (d, J = 8.0 Hz, 2H), 7.32 (t, J = 8.0 Hz, 4H), 7.17 (d, J = 8.0 Hz, 4H), 7.01 (t, J = 8.0 Hz, 2H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 164.9, 156.0, 145.9, 139.9, 129.6, 122.9, 121.8, 119.3; MS (EI, 70 eV) *m/z* 289, 183, 167, 139, 77; HRMS (ESI) *m/z* [M + H]$^+$ calcd for $C_{18}H_{16}N_3O$ 290.1288, found 290.1301.



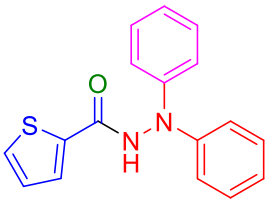
N',N'-Diphenylnicotinohydrazide ***(23)***. Eluent, petroleum ether/ethyl acetate (v/v = 3/1); white solid in 69% yield (19.9 mg, 0.07 mmol); mp 138-139 ℃; IR (KBr, cm^{-1}) 3229, 3038, 1661, 1590, 1494, 1313, 1152, 1025, 898, 747, 697; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 11.43 (s, 1H), 9.12 (s, 1H), 8.79 (d, J = 4.0 Hz, 1H), 8.29 (d, J = 8.0 Hz, 1H), 7.58 (dd, J = 8.0 Hz, J = 4.0 Hz, 1H), 7.32 (t, J = 8.0 Hz, 4H), 7.18 (d, J = 8.0 Hz, 4H), 7.01 (t, J = 8.0 Hz, 2H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.0, 153.6, 148.9, 146.0, 135.8, 129.6, 128.6, 124.2, 122.8, 119.2; MS (EI, 70 eV) *m/z* 289, 183, 167, 139, 77; HRMS (ESI) *m/z* [M + H]$^+$ calcd for $C_{18}H_{16}N_3O$ 290.1288, found 290.1306.



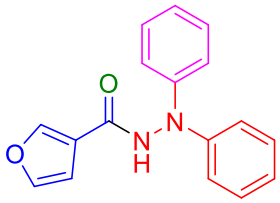
1-Methyl-N',N'-diphenyl-1H-pyrazole-4-carbohydrazide ***(24)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 70% yield (20.4 mg, 0.07 mmol); mp 186-187 ℃; IR (KBr, cm^{-1}) 3249, 3108, 2919, 2850, 1647, 1588, 1559, 1494, 1301, 1224, 1024, 870, 745, 696; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 10.84 (s, 1H), 8.25 (s, 1H), 7.97 (s, 1H), 7.29 (t, J = 8.0 Hz, 4H), 7.13 (d, J = 8.0 Hz, 4H), 6.98 (t, J = 8.0 Hz, 2H), 3.88 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 161.9, 146.3, 139.0, 133.0, 129.5, 122.5, 119.0, 116.2, 39.3; MS (EI, 70 eV) *m/z* 292, 183, 167, 109, 77; HRMS (ESI) *m/z* [M + H]$^+$ calcd for $C_{17}H_{17}N_4O$ 293.1397, found 293.1411.



N',N'-Diphenylthiophene-3-carbohydrazide ***(25)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 68% yield (20.0 mg, 0.07 mmol); mp 127-128 ℃; IR (KBr, cm^{-1}) 3420, 3229, 3026, 1656, 1589, 1541, 1495, 1317, 1026, 999, 851, 747, 692; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 11.10 (s, 1H), 8.31 (s, 1H), 7.69-7.67 (m, 1H), 7.60 (d, J = 4.0 Hz, 1H), 7.30 (t, J = 8.0 Hz, 4H), 7.15 (d, J = 8.0 Hz, 4H), 6.99 (t, J = 8.0 Hz, 2H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 162.0, 146.2, 135.6, 130.4, 129.5, 127.8, 127.2, 122.6, 119.1; MS (EI, 70 eV) *m/z* 294, 183, 169, 111, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{17}H_{15}N_2OS$ 295.0900, found 295.0913.

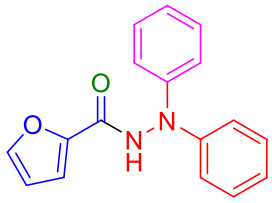


N',N'-Diphenylthiophene-2-carbohydrazide ***(26)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 78% yield (22.9 mg, 0.08 mmol); mp 112-113 ℃; IR (KBr, cm^{-1}) 3422, 3240, 3061, 1645, 1590, 1535, 1494, 1312, 1250, 1026, 850, 747, 693; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 11.25 (s, 1H), 7.96 (d, J = 4.0 Hz, 1H), 7.89 (d, J = 4.0 Hz, 1H), 7.31 (t, J = 8.0 Hz, 4H), 7.23 (t, J = 8.0 Hz, 1H), 7.16 (d, J = 8.0 Hz, 4H), 7.00 (t, J = 8.0 Hz, 2H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 161.3, 146.2, 137.5, 132.5, 129.6, 128.8, 122.7, 120.2, 119.1; MS (EI, 70 eV) *m/z* 294, 183, 169, 111, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{17}H_{15}N_2OS$ 295.0900, found 295.0914.

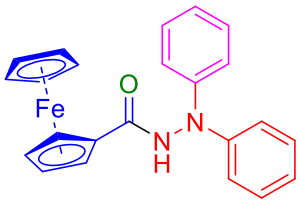


N',N'-Diphenylfuran-3-carbohydrazide ***(27)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 57% yield (15.9 mg, 0.06 mmol); mp 216-217 ℃; IR (KBr, cm^{-1}) 3213, 3063, 1651, 1589, 1523, 1494, 1326, 1276, 1163, 1020, 876,

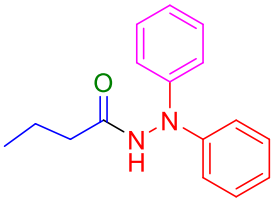
748, 692; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.97 (s, 1H), 8.36 (s, 1H), 7.81 (d, J = 4.0 Hz, 1H), 7.31 (t, J = 8.0 Hz, 4H), 7.17 (s, 4H), 7.01-6.96 (m, 3H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 161.8, 146.3, 146.2, 144.9, 129.6, 122.7, 120.9, 119.1, 109.3; MS (EI, 70 eV) *m/z* 278, 183, 167, 95, 77; HRMS (ESI) *m/z* [M + H]$^{+}$ calcd for $C_{17}H_{15}N_2O_2$ 279.1128, found 279.1142.



N',N'-Diphenylfuran-2-carbohydrazide ***(28)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 75% yield (20.9 mg, 0.08 mmol); mp 105-106 ℃; IR (KBr, cm^{-1}) 3421, 3028, 1669, 1590, 1494, 1311, 1177, 1025, 1006, 861, 752, 695; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 11.16 (s, 1H), 7.93 (s, 1H), 7.30 (t, J = 8.0 Hz, 5H), 7.13 (d, J = 8.0 Hz, 4H), 7.00 (t, J = 8.0 Hz, 2H), 6.70-6.69 (m, 1H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 157.8, 146.5, 146.2, 139.0, 129.5, 122.7, 119.2, 115.4, 112.4; MS (EI, 70 eV) *m/z* 278, 183, 169, 95, 77; HRMS (ESI) *m/z* [M + H]$^{+}$ calcd for $C_{17}H_{15}N_2O_2$ 279.1128, found 279.1142.

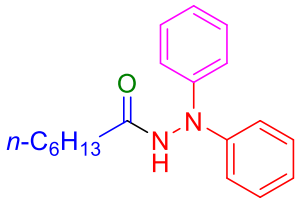


N',N'-Diphenylferrocenylhydrazide ***(29)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); red brown solid in 50% yield (19.8 mg, 0.05 mmol); mp >300 ℃; IR (KBr, cm^{-1}) 3419, 3010, 1661, 1647, 1590, 1507, 1495, 1374, 1297, 1026, 830, 745, 692; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.57 (s, 1H), 7.32 (t, J = 8.0 Hz, 4H), 7.19 (d, J = 8.0 Hz, 4H), 7.00 (t, J = 8.0 Hz, 2H), 4.93 (s, 2H), 4.46 (s, 2H), 4.20 (s, 5H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 169.4, 146.4, 129.9, 122.5, 119.2, 74.5, 71.0, 69.7, 68.8; MS (EI, 70 eV) *m/z* 396, 213, 185, 129, 77; HRMS (ESI) *m/z* [M + H]$^{+}$ calcd for $C_{23}H_{21}N_2OFe$ 397.0998, found 397.1021.

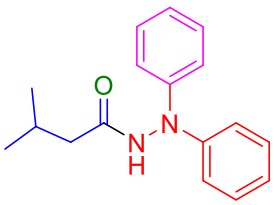


N',N'-Diphenylbutyrohydrazide ***(30)***. Eluent, petroleum ether/ethyl acetate (v/v =

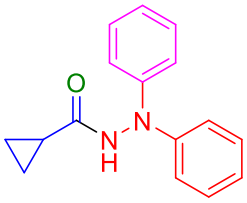
10/1); white solid in 79% yield (20.1 mg, 0.08 mmol); mp 113-114 ℃; IR (KBr, cm^{-1}) 3418, 3257, 3022, 1666, 1591, 1519, 1497, 1332, 1197, 1026, 826, 746, 690; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 10.48 (s, 1H), 7.28 (t, J = 8.0 Hz, 4H), 7.07 (d, J = 8.0 Hz, 4H), 6.97 (t, J = 8.0 Hz, 2H), 2.19 (t, J = 8.0 Hz, 2H), 1.64-1.55 (m, 2H), 0.90 (t, J = 8.0 Hz, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 172.0, 146.3, 129.5, 122.5, 119.1, 35.6, 18.9, 14.1; MS (EI, 70 eV) *m/z* 254, 184, 168, 115, 77; HRMS (ESI) *m/z* $[M + H]^+$ Calcd for $c_{16}H_{19}N_2O$ 255.1492, found 255.1495.



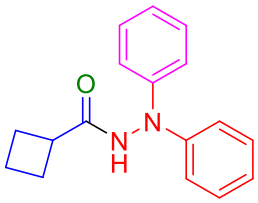
*N',N'-Diphenylheptanehydrazide (**31**).* Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 79% yield (23.4 mg, 0.08 mmol); mp 120-121 ℃; IR (KBr, cm^{-1}) 3263, 3011, 2928, 2852, 1670, 1590, 1520, 1495, 1329, 1185, 1028, 886, 745, 690; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 10.52 (s, 1H), 7.27 (t, J = 8.0 Hz, 4H), 7.08 (d, J = 8.0 Hz, 4H), 6.97 (t, J = 8.0 Hz, 2H), 2.21 (t, J = 8.0 Hz, 2H), 1.57 (t, J = 8.0 Hz, 2H), 1.28 (s, 6H), 0.87 (t, J = 8.0 Hz, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 172.5, 146.3, 129.4, 123.2, 119.1, 33.6, 31.4, 28.8, 25.4, 22.5, 14.4; MS (EI, 70 eV) *m/z* 296, 184, 168, 139, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{19}H_{25}N_2O$ 297.1961, found 297.1973.



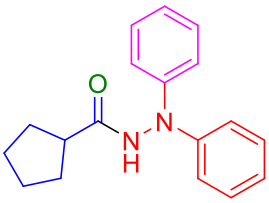
*3-Methyl-N',N'-diphenylbutanehydrazide (**32**).* Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 77% yield (20.6 mg, 0.08 mmol); mp 107-108 ℃; IR (KBr, cm^{-1}) 3422, 3267, 3024, 2958, 2871, 1666, 1591, 1496, 1330, 1199, 1026, 946, 886, 746, 691; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 10.50 (s, 1H), 7.29 (t, J = 8.0 Hz, 4H), 7.09 (d, J = 8.0 Hz, 4H), 6.97 (t, J = 8.0 Hz, 2H), 2.11-2.04 (m, 3H), 0.93 (d, J = 4.0 Hz, 6H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 171.5, 146.3, 129.4, 122.5, 119.1, 42.8, 26.0, 22.8; MS (EI, 70 eV) *m/z* 268, 184, 168, 115, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{17}H_{21}N_2O$ 269.1648, found 269.1656.



N',N'-Diphenylcyclopropanecarbohydrazide ***(33)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 62% yield (15.6 mg, 0.06 mmol); mp 183-184 ℃; IR (KBr, cm^{-1}) 3269, 3024, 2922, 2853, 1664, 1591, 1521, 1495, 1400, 1327, 1275, 1194, 957, 824, 745, 692; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.74 (s, 1H), 7.30 (t, J = 8.0 Hz, 4H), 7.09 (d, J = 8.0 Hz, 4H), 6.98 (t, J = 8.0 Hz, 2H), 1.73-1.67 (m, 1H), 0.79 (d, J = 8.0 Hz, 4H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 172.9, 146.4, 129.5, 122.5, 119.1, 12.4, 7.1; MS (EI, 70 eV) *m/z* 252, 184, 168, 115, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{16}H_{17}N_2O$ 253.1335, found 253.1346.

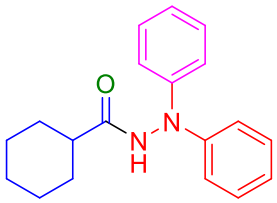


N',N'-Diphenylcyclobutanecarbohydrazide ***(34)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 73% yield (19.4 mg, 0.07 mmol); mp 158-159 ℃; IR (KBr, cm^{-1}) 3242, 3036, 2939, 2855, 1663, 1590, 1534, 1494, 1383, 1274, 1191, 956, 895, 746, 693; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.38 (s, 1H), 7.28 (t, J = 8.0 Hz, 4H), 7.07 (d, J = 8.0 Hz, 4H), 6.97 (t, J = 8.0 Hz, 2H), 3.21-3.13 (m, 1H), 2.20 (t, J = 8.0 Hz, 2H), 2.12 (d, J = 8.0 Hz, 2H), 2.01-1.92 (m, 1H), 1.86-1.78 (m, 1H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 173.7, 146.3, 129.4, 122.4, 119.0, 37.2, 24.8, 18.5; MS (EI, 70 eV) *m/z* 266, 184, 167, 115, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{17}H_{19}N_2O$ 267.1492, found 267.1503.

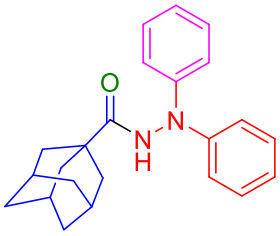


N',N'-Diphenylcyclopentanecarbohydrazide ***(35)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 75% yield (21.0 mg, 0.08 mmol); mp 126-127 ℃; IR (KBr, cm^{-1}) 3238, 3037, 2955, 2867, 1664, 1590, 1533, 1495, 1297, 1201, 1028, 996, 895, 746, 692; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.53 (s, 1H), 7.29 (t, J = 8.0 Hz, 4H), 7.09 (d, J = 8.0 Hz, 4H), 6.97 (t, J = 8.0 Hz, 2H), 2.76-2.68 (m, 1H),

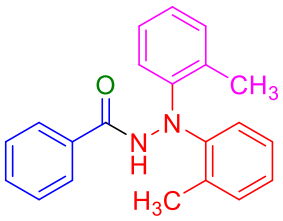
1.85 (d, J = 8.0 Hz, 2H), 1.72-1.63 (m, 4H), 1.54 (t, J = 8.0 Hz, 2H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 175.2, 146.3, 129.5, 122.4, 119.0, 42.6, 30.2, 26.2; MS (EI, 70 eV) *m/z* 280, 184, 167, 115, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{18}H_{21}N_2O$ 281.1648, found 281.1661.



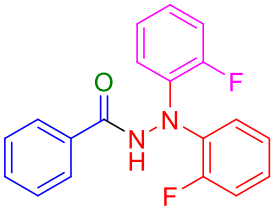
N',N'-Diphenylcyclohexanecarbohydrazide (***36***). Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 65% yield (19.1 mg, 0.07 mmol); mp 206-207 ℃; IR (KBr, cm^{-1}) 3250, 3025, 2929, 2853, 1665, 1590, 1495, 1332, 1193, 1028, 897, 744, 693; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.45 (s, 1H), 7.28 (t, J = 8.0 Hz, 4H), 7.08 (d, J = 8.0 Hz, 4H), 6.96 (t, J = 8.0 Hz, 2H), 2.31-2.24 (m, 1H), 1.81-1.73 (m, 4H), 1.66-1.63 (m, 1H), 1.45-1.36 (m, 2H), 1.32-1.16 (m, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 174.9, 146.3, 129.4, 122.4, 118.9, 42.5, 29.4, 25.8, 25.6; MS (EI, 70 eV) *m/z* 294, 184, 168, 115, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{19}H_{23}N_2O$ 295.1805, found 295.1820.



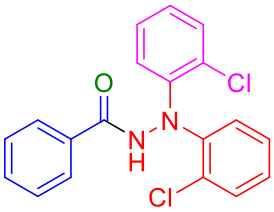
(1R,3R,5S)-N',N'-Diphenyladamantane-1-carbohydrazide (***37***). Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 48% yield (16.6 mg, 0.05 mmol); mp 259-260 ℃; IR (KBr, cm^{-1}) 3283, 3037, 2906, 2850, 1656, 1590, 1494, 1338, 1276, 1180, 930, 833, 743, 691; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.31 (s, 1H), 7.27 (t, J = 8.0 Hz, 4H), 7.08 (d, J = 8.0 Hz, 4H), 6.96 (t, J = 8.0 Hz, 2H), 2.01 (s, 3H), 1.91 (s, 6H), 1.70 (s, 6H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 176.2, 146.3, 129.3, 122.1, 118.7, 38.8, 36.5, 28.0; MS (EI, 70 eV) *m/z* 346, 183, 167, 135, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{23}H_{27}N_2O$ 347.2118, found 347.2136.



*N',N'-Di-o-tolylbenzohydrazide (**38**)*. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 55% yield (17.4 mg, 0.06 mmol); mp 205-206 ℃; IR (KBr, cm^{-1}) 3237, 3055, 2921, 2851, 1652, 1601, 1579, 1525, 1489, 1463, 1309, 1266, 1122, 1025, 945, 892, 754, 693; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 10.97 (s, 1H), 7.88 (d, J = 8.0 Hz, 2H), 7.56 (t, J = 8.0 Hz, 1H), 7.48 (t, J = 8.0 Hz, 2H), 7.19 (d, J = 4.0 Hz, 2H), 7.14 (t, J = 8.0 Hz, 2H), 7.02 (t, J = 8.0 Hz, 4H), 2.07 (s, 6H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.0, 146.6, 133.4, 132.1, 132.0, 131.6, 128.9, 127.8, 126.8, 124.3, 122.1, 18.7; MS (EI, 70 eV) *m/z* 316, 194, 180, 105, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{21}H_{21}N_2O$ 317.1648, found 317.1672.

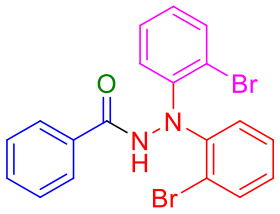


*N',N'-Bis(2-fluorophenyl)benzohydrazide (**39**)*. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 60% yield (19.4 mg, 0.06 mmol); mp 164-165 ℃; IR (KBr, cm^{-1}) 3420, 3266, 3063, 1661, 1502, 1318, 1261, 1026, 1001, 825, 749; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 11.42 (s, 1H), 7.94 (d, J = 4.0 Hz, 2H), 7.61 (t, J = 8.0 Hz, 1H), 7.53 (t, J = 8.0 Hz, 2H), 7.23-7.13 (m, 6H), 7.11-7.08 (m, 2H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 166.2, 154.3 (d, J = 245.0 Hz), 134.6 (d, J = 8.0 Hz), 132.8, 132.5, 129.1, 128.0, 125.1 (d, J = 4.0 Hz), 124.7 (d, J = 8.0 Hz), 122.3, 116.6 (d, J = 20.0 Hz); MS (EI, 70 eV) *m/z* 324, 219, 205, 105, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{19}H_{15}N_2OF_2$ 325.1147, found 325.1171.

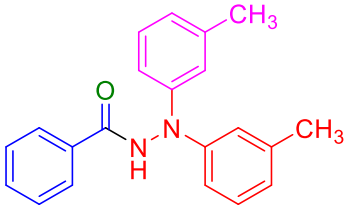


*N',N'-Bis(2-chlorophenyl)benzohydrazide (**40**)*. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 60% yield (21.4 mg, 0.06 mmol); mp 138-139 ℃; IR (KBr, cm^{-1}) 3420, 3225, 3048, 1659, 1538, 1474, 1316, 1269, 1026, 944, 889, 753, 688; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 11.26 (s, 1H), 7.89 (d, J = 8.0 Hz, 2H), 7.58 (t, J = 8.0 Hz, 1H), 7.49 (t, J = 8.0 Hz, 2H), 7.43 (d, J = 8.0 Hz, 2H), 7.30 (t, J = 8.0 Hz, 2H), 7.19 (d, J = 8.0 Hz, 2H), 7.13 (t, J = 8.0 Hz, 2H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.6, 143.9, 133.0, 132.3, 131.0, 129.0, 128.09, 128.05,

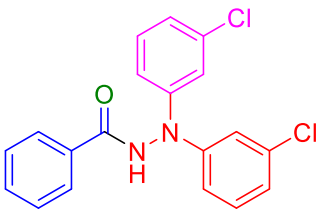
127.1, 125.9, 124.5; MS (EI, 70 eV) *m/z* 356, 321, 251, 183, 77; HRMS (ESI) *m/z* [M + H]$^+$ calcd for $C_{19}H_{15}N_2OCl_2$ 357.0556, found 357.0582.



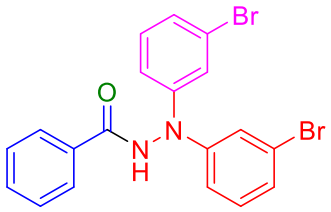
N',N'-Bis(2-bromophenyl)benzohydrazide ***(41)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 52% yield (23.1 mg, 0.05 mmol); mp 201-202 ℃; IR (KBr, cm^{-1}) 3421, 3055, 1659, 1471, 1316, 1027, 1000, 827, 752, 692; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 11.22 (s, 1H), 7.91 (d, J = 8.0 Hz, 2H), 7.62 (d, J = 12.0 Hz, 2H), 7.57 (d, J = 8.0 Hz, 1H), 7.49 (t, J = 8.0 Hz, 2H), 7.35 (t, J = 8.0 Hz, 2H), 7.18 (d, J = 12.0 Hz, 2H), 7.07 (t, J = 8.0 Hz, 2H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 165.4, 145.3, 134,4, 133.0, 132.3, 128.9, 128.6, 128.1, 126.3, 125.1, 117.9; MS (EI, 70 eV) *m/z* 444, 365, 183, 105, 77; HRMS (ESI) *m/z* [M + H]$^+$ calcd for $C_{19}H_{15}N_2OBr_2$ 444.9546, found 444.9563.



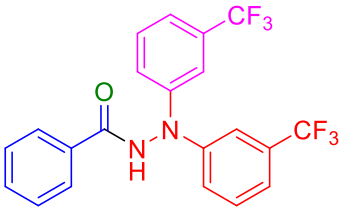
N',N'-Di-m-tolylbenzohydrazide ***(42)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 62% yield (19.6 mg, 0.06 mmol); mp 109-110 ℃; IR (KBr, cm^{-1}) 3422, 3060, 2921, 2852, 1660, 1602, 1582, 1489, 1312, 1261, 1026, 1002, 904, 772, 692; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 11.15 (s, 1H), 7.95 (d, J = 8.0 Hz, 2H), 7.61 (t, J = 8.0 Hz, 1H), 7.53 (t, J = 8.0 Hz, 2H), 7.18 (t, J = 8.0 Hz, 2H), 6.95 (d, J = 8.0 Hz, 4H), 6.81 (d, J = 8.0 Hz, 2H), 2.25 (s, 6H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 166.2, 146.4, 138.7, 133.0, 132.4, 129.3, 129.1, 127.9, 123.3, 119.7, 116.5, 21.7; MS (EI, 70 eV) *m/z* 316, 211, 197, 105, 77; HRMS (ESI) *m/z* [M + H]$^+$ calcd for $C_{21}H_{21}N_2O$ 317.1648, found 317.1672.



N',N'-Bis(3-chlorophenyl)benzohydrazide ***(43)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 50% yield (17.8 mg, 0.05 mmol); mp 104-105 ℃; IR (KBr, cm^{-1}) 3424, 3065, 1662, 1585, 1477, 1310, 1026, 994, 898, 770, 704; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 11.38 (s, 1H), 7.96 (d, J = 4.0 Hz, 2H), 7.64 (t, J = 8.0 Hz, 1H), 7.55 (t, J = 8.0 Hz, 2H), 7.36 (t, J = 8.0 Hz, 2H), 7.15 (t, J = 4.0 Hz, 4H), 7.10 (d, J = 8.0 Hz, 2H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 166.3, 147.1, 134.1, 132.8, 132.4, 131.4, 129.2, 128.0, 123.0, 118.9, 118.2; MS (EI, 70 eV) *m/z* 356, 251, 201, 105, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{19}H_{15}N_2OCl_2$ 357.0556, found 357.0582.

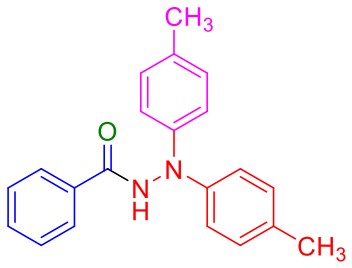


N',N'-Bis(3-bromophenyl)benzohydrazide ***(44)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 58% yield (25.8 mg, 0.06 mmol); mp 120-121 ℃; IR (KBr, cm^{-1}) 3424, 3059, 1666, 1581, 1475, 1308, 1026, 1007, 897, 770, 693; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 11.38 (s, 1H), 7.95 (d, J = 8.0 Hz, 2H), 7.64 (t, J = 8.0 Hz, 1H), 7.55 (t, J = 8.0 Hz, 2H), 7.30-7.28 (m, 4H), 7.24-7.18 (m, 4H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 166.4, 147.2, 132.9, 132.3, 131.7, 129.2, 128.0, 126.0, 122.5, 121.8, 118.6; MS (EI, 70 eV) *m/z* 444, 341, 166, 105, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{19}H_{15}N_2OBr_2$ 444.9546, found 444.9577.

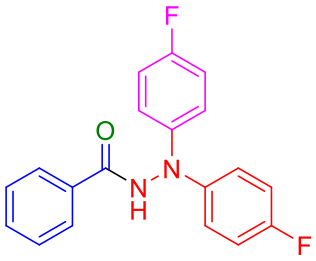


N',N'-Bis(3-(trifluoromethyl)phenyl)benzohydrazide ***(45)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 55% yield (23.3 mg, 0.06 mmol); mp 154-155 ℃; IR (KBr, cm^{-1}) 3250, 3037, 1661, 1591, 1496, 1458, 1336, 1168, 1123, 1071, 904, 876, 788, 696; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 11.54 (s, 1H), 7.99 (d, J = 4.0 Hz, 2H), 7.63 (t, J = 8.0 Hz, 1H), 7.59-7.50 (m, 6H), 7.46 (s, 2H), 7.39 (d, J = 8.0 Hz, 2H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 166.6, 146.3, 132.6 (d, J = 56.0 Hz), 131.1, 130.7 (d, J = 31.0 Hz), 129.2, 128.0, 125.7, 123.2, 123.0, 119.8 (d, J = 4.0 Hz), 115.4 (d, J = 4.0 Hz); MS (EI, 70 eV) *m/z* 424, 305, 235, 105, 77;

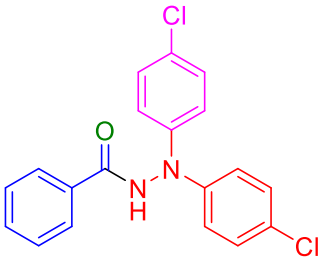
HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{21}H_{15}N_2OF_6$ 425.1083, found 425.1102.



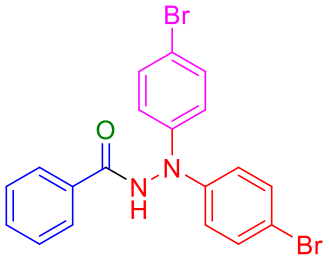
N',N'-Di-p-tolylbenzohydrazide (***46***). Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 62% yield (19.6 mg, 0.06 mmol); mp 119-120 ℃; IR (KBr, cm^{-1}) 3422, 3028, 2922, 2857, 1661, 1509, 1324, 1299, 1026, 1006, 812, 699; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 11.13 (s, 1H), 7.94 (d, J = 8.0 Hz, 2H), 7.60 (t, J = 8.0 Hz, 1H), 7.52 (t, J = 8.0 Hz, 2H), 7.10 (d, J = 8.0 Hz, 4H), 7.04 (d, J = 8.0 Hz, 4H), 2.25 (s, 6H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 166.2, 144.2, 133.1, 132.4, 131.3, 129.9, 129.0, 127.9, 119.2, 20.7; MS (EI, 70 eV) *m/z* 316, 211, 197, 105, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{21}H_{21}N_2O$ 317.1648, found 317.1672.



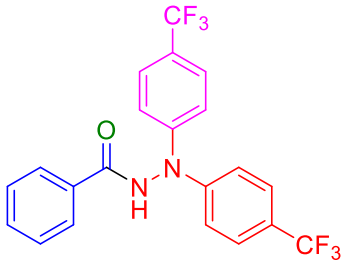
N',N'-Bis(4-fluorophenyl)benzohydrazide (***47***). Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 54% yield (17.5 mg, 0.05 mmol); mp 110-111 ℃; IR (KBr, cm^{-1}) 3259, 3053, 1661, 1505, 1313, 1290, 1226, 1153, 1027, 904, 824, 695; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 11.24 (s, 1H), 7.95 (d, J = 8.0 Hz, 2H), 7.61 (t, J = 8.0 Hz, 1H), 7.53 (t, J = 8.0 Hz, 2H), 7.14 (d, J = 4.0 Hz, 8H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 166.4, 158.2 (d, J = 238.0 Hz), 143.1 (d, J = 2.0 Hz), 132.8 (d, J = 26.0 Hz), 129.1, 128.5 (d, J = 79.0 Hz), 127.9, 121.1 (d, J = 8.0 Hz), 116.2 (d, J = 23.0 Hz); MS (EI, 70 eV) *m/z* 324, 219, 105, 95, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{19}H_{15}N_2OF_2$ 325.1147, found 325.1157.



N',N'-Bis(4-chlorophenyl)benzohydrazide ***(48)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 52% yield (18.5 mg, 0.05 mmol); mp 134-135 ℃; IR (KBr, cm^{-1}) 3221, 3059, 1658, 1491, 1322, 1089, 1026, 819, 691; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 11.32 (s, 1H), 7.94 (d, J = 8.0 Hz, 2H), 7.63 (t, J = 8.0 Hz, 1H), 7.54 (t, J = 8.0 Hz, 2H), 7.37 (d, J = 4.0 Hz, 4H), 7.18 (d, J = 4.0 Hz, 4H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 166.3, 144.8, 132.7, 132.6, 129.5, 129.1, 128.0, 126.6, 120.9; MS (EI, 70 eV) *m/z* 356, 251, 237, 105, 77; HRMS (ESI) *m/z* [M + H]$^+$ calcd for $C_{19}H_{15}N_2OCl_2$ 357.0556, found 357.0580.

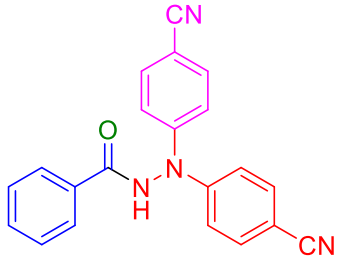


N',N'-Bis(4-bromophenyl)benzohydrazide ***(49)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 46% yield (20.4 mg, 0.05 mmol); mp 138-139 ℃; IR (KBr, cm^{-1}) 3422, 3228, 3061, 1654, 1580, 1487, 1317, 1026, 1008, 816, 760, 692; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 11.33 (s, 1H), 7.94 (d, J = 8.0 Hz, 2H), 7.63 (t, J = 8.0 Hz, 1H), 7.54 (t, J = 8.0 Hz, 2H), 7.49 (d, J = 8.0 Hz, 4H), 7.13 (d, J = 12.0 Hz, 4H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 166.3, 145.2, 132.7, 132.5, 132.4, 129.1, 128.0, 121.3, 114.4; MS (EI, 70 eV) *m/z* 444, 341, 327, 105, 77; HRMS (ESI) *m/z* [M + H]$^+$ calcd for $C_{19}H_{15}N_2OBr_2$ 444.9546, found 444.9576.

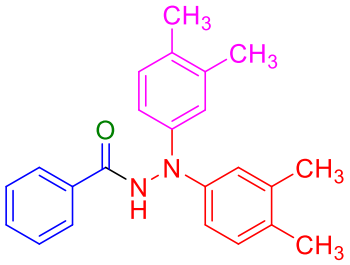


N',N'-Bis(4-(trifluoromethyl)phenyl)benzohydrazide ***(50)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 45% yield (19.1 mg, 0.05 mmol); mp 127-128 ℃; IR (KBr, cm^{-1}) 3226, 3059, 1660, 1608, 1519, 1319, 1160, 1107, 1067, 1026, 1006, 907, 832, 691; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 11.58 (s, 1H), 7.97 (d, J = 8.0 Hz, 2H), 7.69 (d, J = 8.0 Hz, 4H), 7.64 (d, J = 8.0 Hz, 1H), 7.56 (t, J = 8.0 Hz, 2H), 7.40 (d, J = 8.0 Hz, 4H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ

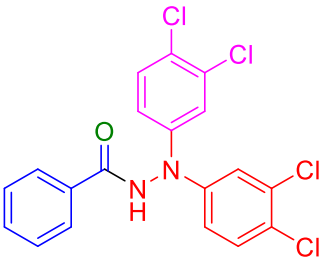
166.3, 148.4, 132.6 (d, J = 70.0 Hz), 129.2, 128.0, 127.1 (q, J = 4.0 Hz), 126.2, 123.5, 123.3 (d, J = 32.0 Hz), 119.3; MS (EI, 70 eV) *m/z* 424, 319, 235, 105, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{21}H_{15}N_2OF_6$ 425.1083, found 425.1094.



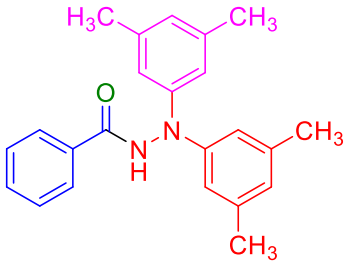
N',N'-Bis(4-cyanophenyl)benzohydrazide ***(51)***. Eluent, petroleum ether/ethyl acetate (v/v = 3/1); white gel in 32% yield (10.8 mg, 0.03 mmol); IR (KBr, cm^{-1}) 3199, 3070, 2223, 1679, 1596, 1502, 1331, 1300, 1252, 1176, 1025, 1007, 830, 699; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 11.68 (s, 1H), 7.98 (d, J = 8.0 Hz, 2H), 7.79 (d, J = 8.0 Hz, 4H), 7.65 (t, J = 8.0 Hz, 1H), 7.56 (t, J = 8.0 Hz, 2H), 7.39 (d, J = 8.0 Hz, 4H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 166.3, 148.4, 134.2, 133.0, 132.0, 129.2, 128.1, 119.6, 119.5, 105.2; MS (EI, 70 eV) *m/z* 338, 219, 191, 105, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{21}H_{15}N_4O$ 339.1240, found 339.1265.



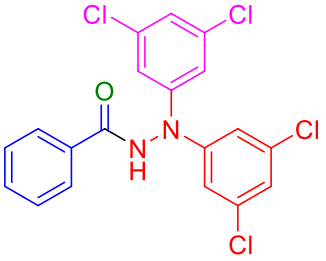
N',N'-Bis(3,4-dimethylphenyl)benzohydrazide ***(52)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 60% yield (20.7 mg, 0.06 mmol); mp 113-114 ℃; IR (KBr, cm^{-1}) 3397, 3268, 3053, 2920, 2850, 1657, 1608, 1503, 1488, 1311, 1286, 1026, 895, 806, 693; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 11.03 (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), 7.03 (d, J = 8.0 Hz, 2H), 6.91 (s, 2H), 6.85 (d, J = 8.0 Hz, 2H), 2.15 (s, 12H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 166.1, 144.6, 137.0, 133.2, 132.3, 130.3, 130.1, 129.0, 127.9, 120.5, 116.8, 20.2, 19.1; MS (EI, 70 eV) *m/z* 344, 239, 225, 105, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{23}H_{25}N_2O$ 345.1961, found 345.1985.



*N',N'-Bis(3,4-dichlorophenyl)benzohydrazide (**53**)*. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 40% yield (17.0 mg, 0.04 mmol); mp 184-185 ℃; IR (KBr, cm^{-1}) 3422, 3251, 3068, 1659, 1582, 1473, 1311, 1131, 1026, 1007, 902, 814, 693; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 11.45 (s, 1H), 7.95 (d, J = 4.0 Hz, 2H), 7.64 (t, J = 8.0 Hz, 1H), 7.55 (t, J = 8.0 Hz, 4H), 7.36 (d, J = 4.0 Hz, 2H), 7.18 (dd, J = 8.0 Hz, J = 4.0 Hz, 2H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 166.3, 145.4, 132.9, 132.1, 131.6, 129.2, 128.1, 125.1, 120.9, 119.9; MS (EI, 70 eV) *m/z* 424, 307, 235, 105, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{19}H_{13}N_2OCl_4$ 424.9777, found 424.9805.

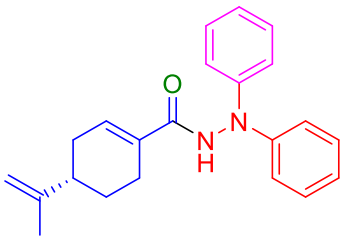


*N',N'-Bis(3,5-dimethylphenyl)benzohydrazide (**54**)*. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 54% yield (18.6 mg, 0.05 mmol); mp 145-146 ℃; IR (KBr, cm^{-1}) 3423, 3284, 3057, 2921, 2853, 1660, 1590, 1486, 1339, 1288, 1196, 1027, 1006, 832, 731; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 11.06 (s, 1H), 7.94 (d, J = 8.0 Hz, 2H), 7.61 (t, J = 8.0 Hz, 1H), 7.53 (t, J = 8.0 Hz, 2H), 6.75 (s, 4H), 6.63 (s, 2H), 2.21 (s, 12H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 166.1, 146.6, 138.4, 133.1, 132.4, 129.1, 127.9, 124.2, 117.2, 21.6; MS (EI, 70 eV) *m/z* 344, 239, 225, 105, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{23}H_{25}N_2O$ 345.1961, found 345.1983.

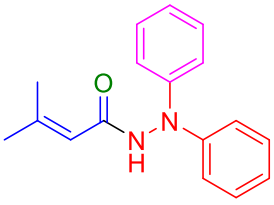


*N',N'-Bis(3,5-dichlorophenyl)benzohydrazide (**55**)*. Eluent, petroleum ether/ethyl

acetate (v/v = 10/1); white solid in 43% yield (18.2 mg, 0.04 mmol); mp 224-225 ℃; IR (KBr, cm^{-1}) 3242, 3083, 1654, 1575, 1446, 1312, 1114, 1006, 837, 803, 690; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 11.47 (s, 1H), 7.95 (d, J = 8.0 Hz, 2H), 7.65 (t, J = 8.0 Hz, 1H), 7.55 (t, J = 12.0 Hz, 2H), 7.28 (d, J = 4.0 Hz, 2H), 7.18 (d, J = 4.0 Hz, 4H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 166.4, 147.5, 135.2, 133.1, 131.9, 129.2, 128.1, 123.2, 118.3; MS (EI, 70 eV) *m/z* 424, 307, 269, 105, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{19}H_{13}N_2OCl_4$ 424.9777, found 424.9808.

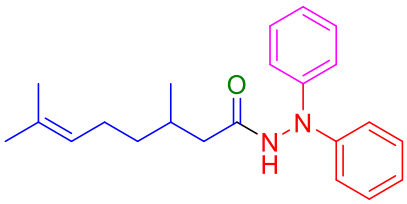


(S)-N',N'-Diphenyl-4-(prop-1-en-2-yl)cyclohex-1-ene-1-carbohydrazide ***(58)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 56% yield (18.6 mg, 0.06 mmol); mp 189-190 ℃; IR (KBr, cm^{-1}) 3267, 3060, 2962, 2921, 1659, 1633, 1589, 1496, 1308, 1239, 1072, 893, 746, 697; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 10.55 (s, 1H), 7.28 (t, J = 8.0 Hz, 4H), 7.11 (d, J = 4.0 Hz, 4H), 6.97 (t, J = 8.0 Hz, 2H), 6.73 (s, 1H), 4.75 (s, 2H), 2.44-2.41 (m, 1H), 2.28 (s, 1H), 2.16-2.06 (m, 2H), 1.87-1.83 (m, 1H), 1.74 (s, 3H), 1.48-1.38 (m, 1H), 1.24 (d, J = 8.0 Hz, 1H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 167.3, 149.1, 146.3, 133.7, 132.0, 129.4, 122.4, 119.1, 109.7, 40.0, 30.7, 27.1, 24.8, 21.0; MS (EI, 70 eV) *m/z* 332, 183, 167, 149, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{22}H_{25}N_2O$ 333.1961, found 333.1979.

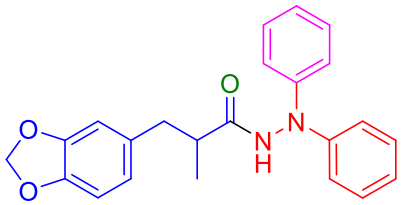


3-Methyl-N',N'-diphenylbut-2-enehydrazide ***(59)***. Eluent, petroleum ether/ethyl acetate (v/v = 5/1); yellow solid in 40% yield (10.6 mg, 0.04 mmol); mp 170-171 ℃; IR (KBr, cm^{-1}) 3249, 3061, 3007, 2921, 2851, 1661, 1641, 1590, 1495, 1332, 1277, 1165, 1078, 841, 745, 691; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 10.41 (s, 1H), 7.28 (t, J = 8.0 Hz, 4H), 7.07 (d, J = 8.0 Hz, 4H), 6.97 (t, J = 8.0 Hz, 2H), 5.78 (s, 1H), 2.10 (s, 3H), 1.86 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 165.9, 153.0,

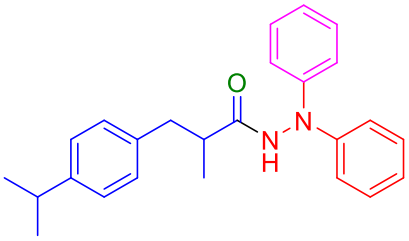
146.3, 129.5, 122.4, 119.1, 116.3, 27.5, 20.0; MS (EI, 70 eV) *m/z* 266, 184, 168, 115, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{17}H_{19}N_2O$ 267.1492, found 267.1502.



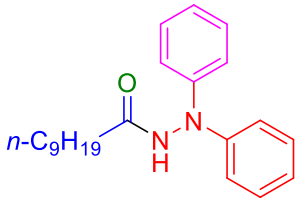
3,7-Dimethyl-N',N'-diphenyloct-6-enehydrazide ***(60)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 64% yield (21.5 mg, 0.06 mmol); mp 68-69 ℃; IR (KBr, cm^{-1}) 3254, 3025, 2963, 2925, 2855, 1666, 1591, 1495, 1377, 1331, 1276, 1028, 825, 747, 692; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 10.50 (s, 1H), 7.27 (t, J = 8.0 Hz, 4H), 7.08 (d, J = 8.0 Hz, 4H), 6.97 (t, J = 8.0 Hz, 2H), 5.08 (t, J = 8.0 Hz, 1H), 2.24-2.19 (m, 1H), 2.07-1.92 (m, 4H), 1.66 (s, 3H), 1.58 (s, 3H), 1.38-1.30 (m, 1H), 1.22-1.14 (m, 1H), 0.91 (d, J = 4.0 Hz, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 171.6, 146.3, 131.1, 129.4, 124.8, 122.5, 119.1, 41.1, 36.8, 30.2, 26.0, 25.3, 19.9, 18.0; MS (EI, 70 eV) *m/z* 336, 184, 168, 115, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{22}H_{29}N_2O$ 337.2274, found 337.2287.



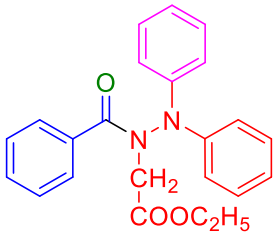
3-(Benzo[d][1,3]dioxol-5-yl)-2-methyl-N',N'-diphenylpropanehydrazide ***(61)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 70% yield (26.2 mg, 0.07 mmol); mp 140-141 ℃; IR (KBr, cm^{-1}) 3422, 3259, 3036, 2934, 2851, 1668, 1591, 1489, 1443, 1248, 1032, 1007, 923, 822, 747, 701; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 10.43 (s, 1H), 7.21-7.15 (m, 5H), 6.99-6.92 (m, 4H), 6.85 (d, J = 8.0 Hz, 2H), 6.81 (s, 1H), 6.68 (dd, J = 8.0 Hz, J = 4.0 Hz, 1H), 6.02 (s, 1H), 5.99 (s, 1H), 2.79-2.70 (m, 2H), 2.62 (dd, J = 8.0 Hz, J = 4.0 Hz, 1H), 1.12 (d, J = 4.0 Hz, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 174.5, 147.6, 146.1, 146.0, 134.0, 129.3, 122.5, 109.9, 108.5, 101.2, 39.5, 39.3, 18.2; MS (EI, 70 eV) *m/z* 374, 184, 168, 135, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{23}H_{23}N_2O_3$ 375.1703, found 375.1730.



3-(4-Isopropylphenyl)-2-methyl-N',N'-diphenylpropanehydrazide ***(62)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 73% yield (27.2 mg, 0.07 mmol); mp 116-117 ℃; IR (KBr, cm^{-1}) 3241, 3027, 2963, 2927, 2870, 1668, 1590, 1534, 1496, 1458, 1215, 1178, 1098, 816, 747, 691; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.43 (s, 1H), 7.19-7.12 (m, 9H), 6.99-6.93 (m, 5H), 2.94-2.89 (m, 1H), 2.86-2.81 (m, 1H), 2.75-2.70 (m, 1H), 2.64-2.59 (m, 1H), 1.23 (dd, J = 8.0 Hz, J = 4.0 Hz, 6H), 1.12 (d, J = 8.0 Hz, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 174.6, 146.5, 146.0, 137.5, 129.4, 129.3, 126.5, 122.2, 118.8, 39.3, 33.5, 24.5, 24.4, 18.4; MS (EI, 70 eV) *m/z* 372, 184, 168, 133, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{25}H_{29}N_2O$ 373.2274, found 373.2301.

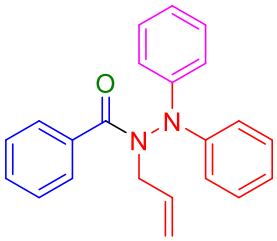


N',N'-Diphenyldecanehydrazide ***(63)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 73% yield (24.7 mg, 0.07 mmol); mp 104-105 ℃; IR (KBr, cm^{-1}) 3267, 3024, 2917, 2850, 1671, 1591, 1522, 1496, 1331, 1276, 1183, 1027, 963, 745, 689; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.47 (s, 1H), 7.28 (t, J = 8.0 Hz, 4H), 7.07 (d, J = 8.0 Hz, 4H), 6.97 (t, J = 8.0 Hz, 2H), 2.20 (t, J = 8.0 Hz, 2H), 1.56 (t, J = 8.0 Hz, 2H), 1.26 (s, 12H), 0.87 (t, J = 8.0 Hz, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 172.2, 146.3, 129.4, 122.5, 119.1, 33.6, 31.8, 29.4, 29.1, 29.1, 29.0, 25.4, 22.6, 14.4; MS (EI, 70 eV) *m/z* 338, 184, 168, 115, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{22}H_{31}N_2O$ 339.2431, found 339.2456.

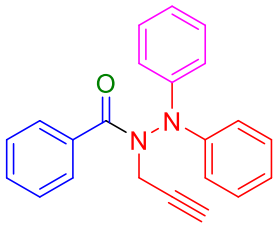


Ethyl N-Benzoyl-N-(diphenylamino)glycinate ***(64)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white gel in 85% yield (31.8 mg, 0.09 mmol); IR (KBr, cm^{-1})

3445, 3063, 2980, 2933, 1750, 1659, 1590, 1492, 1417, 1381, 1299, 1199, 1030, 991, 753, 696; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 7.44 (d, J = 8.0 Hz, 3H), 7.32 (q, J = 8.0 Hz, 6H), 7.07 (t, J = 8.0 Hz, 2H), 6.98 (d, J = 8.0 Hz, 4H), 4.41 (s, 2H), 4.07 (q, J = 8.0 Hz, 2H), 1.15 (t, J = 8.0 Hz, 3H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 172.4, 168.0, 144.8, 134.6, 131.4, 130.0, 128.5, 127.4, 124.2, 120.2, 61.1, 50.8, 14.4; MS (EI, 70 eV) *m/z* 374, 269, 168, 105, 77; HRMS (ESI) *m/z* [M + H]$^{+}$ calcd for $C_{23}H_{23}N_2O_3$ 375.1703, found 375.1730.

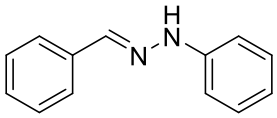


N-Allyl-N',N'-diphenylbenzohydrazide ***(65)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white gel in 82% yield (26.9 mg, 0.08 mmol); IR (KBr, cm^{-1}) 3422, 3063, 2922, 2852, 1657, 1590, 1492, 1385, 1315, 1293, 1028, 924, 750, 695; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 7.62-7.55 (m, 2H), 7.37 (d, J = 8.0 Hz, 3H), 7.29 (t, J = 16.0 Hz, 4H), 7.17 (d, J = 8.0 Hz, 1H), 7.03 (t, J = 8.0 Hz, 2H), 6.96 (d, J = 8.0 Hz, 3H), 5.96-5.86 (m, 1H), 5.08 (t, J = 8.0 Hz, 2H), 4.36 (d, J = 4.0 Hz, 2H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 172.4, 144.8, 135.7, 133.3, 130.7, 129.8, 128.3, 127.1, 123.6, 119.7, 119.0, 51.7; MS (EI, 70 eV) m/z 328, 287, 223, 168, 77; HRMS (ESI) *m/z* [M + H]$^{+}$ calcd for $C_{22}H_{21}N_2O$ 329.1648, found 329.1672.

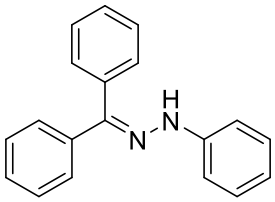


N',N'-Diphenyl-N-(prop-2-yn-1-yl)benzohydrazide ***(66)***. Eluent, petroleum ether/ethyl acetate (v/v = 10/1); white solid in 92% yield (30.0 mg, 0.09 mmol); mp 146-147 ℃; IR (KBr, cm^{-1}) 3423, 3061. 1654, 1590, 1493, 1388, 1282, 1029, 830, 751, 695; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 7.38 (t, J = 8.0 Hz, 3H), 7.29 (q, J = 8.0 Hz, 6H), 7.06 (d, J = 4.0 Hz, 2H), 7.01 (t, J = 8.0 Hz, 4H), 4.49 (s, 2H), 3.23 (s, 1H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 171.9, 144.3, 135.0, 131.1, 129.9,

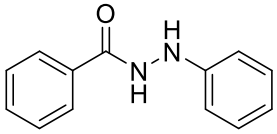
128.4, 127.2, 123.8, 119.6, 79.2, 76.2, 38.0; MS (EI, 70 eV) m/z 326, 287, 221, 168, 77; HRMS (ESI) *m/z* $[M]^+$ calcd for $C_{22}H_{19}N_2O$ 326.1414, found 326.1437.



(E)-1-Benzylidene-2-phenylhydrazine (***69***). Light yellow solid in 99% yield (195.0 mg, 1.00 mmol); mp 159-160 ℃; IR (KBr, cm^{-1}) 3313, 3056, 3027, 1602, 1593, 1522, 1494, 1433, 1262, 1136, 1066, 930, 882, 750, 691; ^1H NMR (400 MHz, DMSO-d_6, ppm) δ 10.34 (s, 1H), 7.90 (s, 1H), 7.66 (d, J = 8.0 Hz, 2H), 7.40 (t, J = 8.0 Hz, 2H), 7.29 (t, J = 8.0 Hz, 1H), 7.24 (t, J = 8.0 Hz, 2H), 7.11 (d, J = 8.0 Hz, 2H), 6.77 (t, J = 8.0 Hz, 1H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6, ppm) δ 145.8, 136.9, 136.3, 129.6, 129.1, 128.4, 126.1, 119.3, 112.5; MS (EI, 70 eV) *m/z* 196, 167, 119, 92, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{13}H_{13}N_2$ 197.1073, found 197.1091.

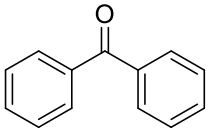


1-(Diphenylmethylene)-2-phenylhydrazine (***70***). White solid; mp 137-138 ℃; IR (KBr, cm^{-1}) 3323, 3051, 3026, 1600, 1557, 1500, 1489, 1332, 1247, 1171, 1070, 998, 884, 777, 699; ^1H NMR (400 MHz, $CDCl_3$, ppm) δ 7.59-7.55 (m, 4H), 7.52-7.49 (m, 2H), 7.33-7.22 (m, 7H), 7.07 (d, J = 8.0 Hz, 2H), 6.83 (t, J = 8.0 Hz, 1H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 144.7, 144.2, 138.4, 132.8, 129.8, 129.30, 129.28, 129.21, 129.24, 128.1, 126.5, 120.1, 113.0. MS (EI, 70 eV) *m/z*: 272, 194, 180, 169, 77; HRMS (ESI) *m/z* $[M + H]^+$ calcd for $C_{19}H_{17}N_2$ 273.1386, found 273.1393.

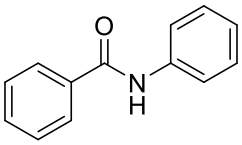


N'-Phenylbenzohydrazide (***71***). White solid in 80% yield (170.0 mg, 0.80 mmol); mp 170-171 ℃; IR (KBr, cm^{-1}) 3329, 3266, 3057, 1644, 1603, 1578, 1523, 1496,

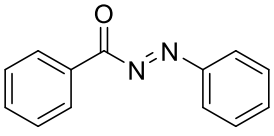
1088, 902, 750, 691; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.39 (s, 1H), 7.95 (d, J = 8.0 Hz, 2H), 7.91 (s, 1H), 7.59 (t, J = 8.0 Hz, 1H), 7.52 (t, J = 8.0 Hz, 2H), 7.18 (t, J = 8.0 Hz, 2H), 6.82 (d, J = 8.0 Hz, 2H), 6.74 (t, J = 8.0 Hz, 1H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 166.9, 150.0, 133.5, 132.1, 129.2, 129.0, 127.8, 119.2, 112.8; MS (EI, 70 eV) *m/z* 212, 121, 105, 77, 51; HRMS (ESI) *m/z* [M + H]$^{+}$ calcd for $C_{13}H_{13}N_2O$ 213.1022, found 213.1039.



*Benzophenone (**72**).*[2] White solid; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.81-7.80 (m, 4H), 7.59 (t, J = 8.0 Hz, 2H), 7.48 (t, J = 8.0 Hz, 4H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$, ppm) δ 196.8, 137.6, 132.4, 130.1, 128.3; MS (EI, 70 eV) *m/z*: 272, 194, 180, 169, 77; MS (EI, 70 eV) *m/z*: 182, 169, 152, 105, 77.



*N-Phenylbenzamide (**73**).*[3] White solid; ^{1}H NMR (400 MHz, DMSO-d_6, ppm) δ 10.29 (s, 1H), 7.98 (d, J = 8.0 Hz, 2H), 7.82 (d, J = 8.0 Hz, 2H), 7.62-7.53 (m, 3H), 7.37 (t, J = 8.0 Hz, 2H), 7.12 (t, J = 8.0 Hz, 1H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6, ppm) δ 166.1, 139.7, 135.5, 132.0, 129.1, 128.9, 128.1, 124.1, 120.8; MS (EI, 70 eV) *m/z*: 197, 105, 77, 65, 51.



*(E)-Phenyl(phenyldiazenyl)methanone (**74**).*[4] Red oily liquid in 96% yield (100.8 mg, 0.48 mmol); IR (KBr, cm^{-1}) 3060, 1690, 1602, 1494, 1451, 1316, 1290, 1026, 750, 709, 692; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.09 (d, J = 8.0 Hz, 2H), 8.03 (d, J = 8.0 Hz, 2H), 7.69 (t, J = 8.0 Hz, 1H), 7.63-7.53 (m, 5H); ^{13}C{^{1}H} NMR (100 MHz,

$CDCl_3$, ppm) δ 182.1, 152.1, 134.5, 133.4, 130.9, 130.6, 129.4, 128.9, 123.6.

Reference

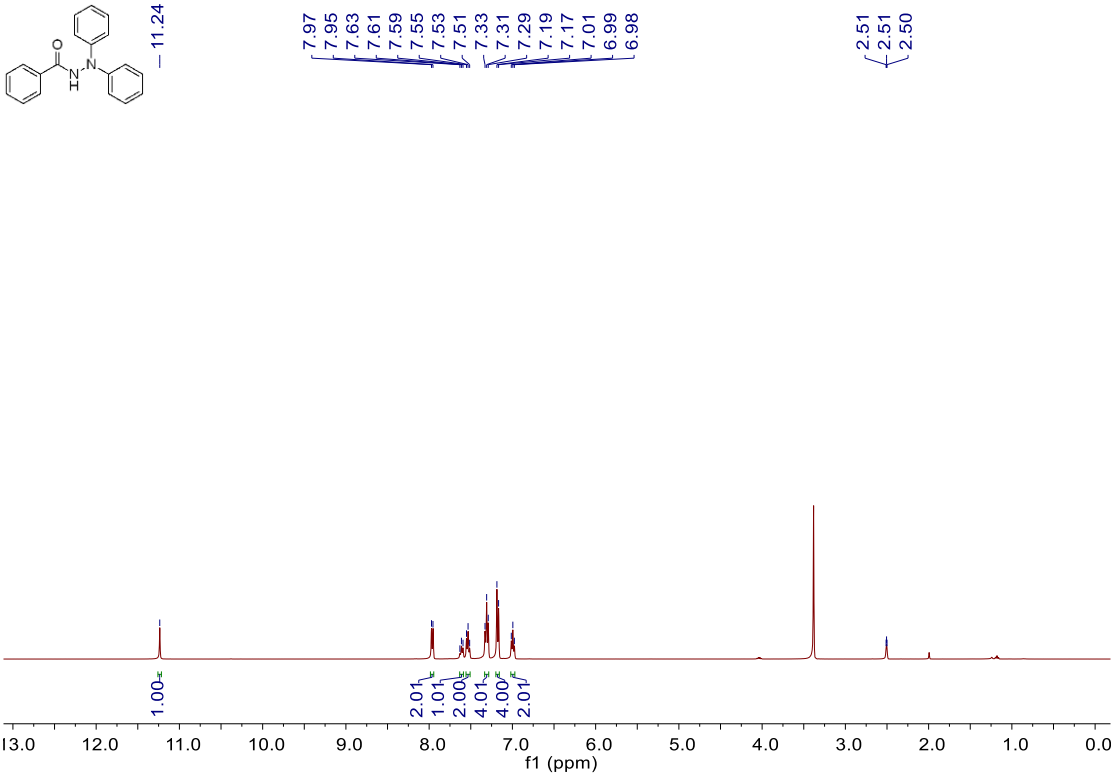
(1) Zhang, J.-Q.; Huang, G.-B.; Weng, J.; Lu, G.; Chan, A. S. C. Copper(II)-Catalyzed Coupling Reaction: An Efficient and Regioselective Approach to *N′*,*N′*-Diaryl Acylhydrazines. *Org. Biomol. Chem.* **2015**, *13*, 2055-2063.

(2) Ren, L.; Jiao, N. Pd/Cu-Cocatalyzed Aerobic Oxidative Carbonylative Homocoupling of Arylboronic Acids and CO: A Highly Selective Approach to Diaryl Ketones. *Chem.-Asian J.* **2014**, *9*, 2411-2414.

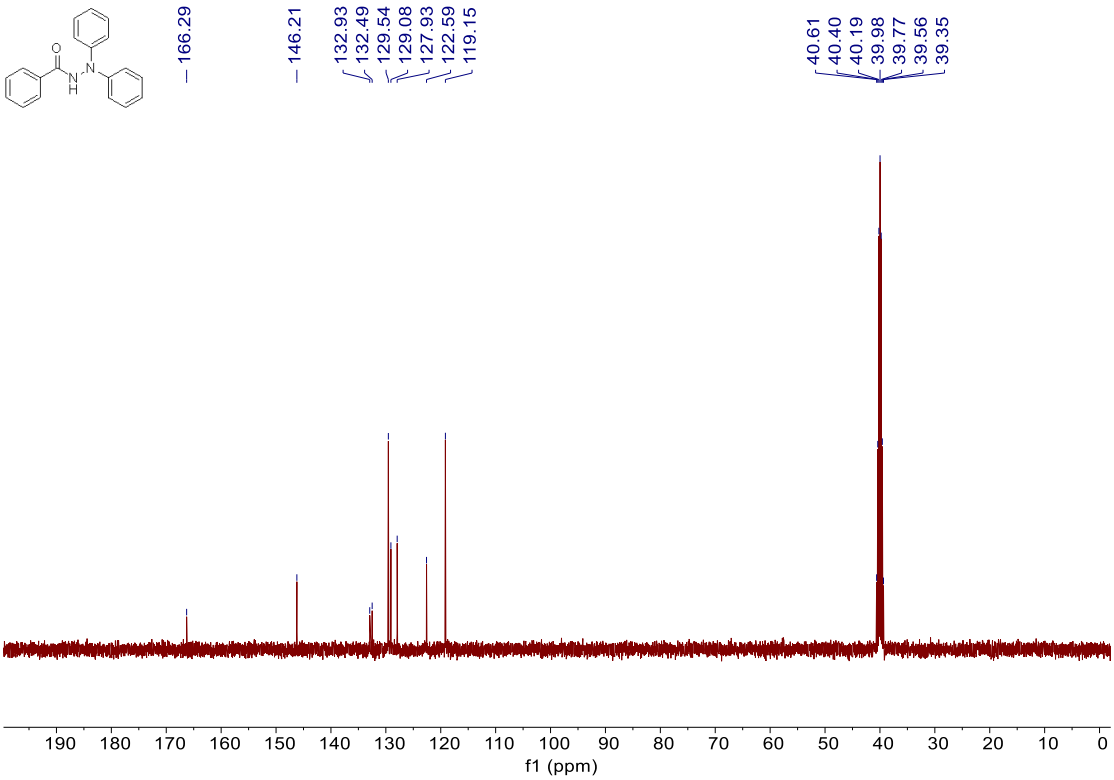
(3) Zhang, R.; Yao, W.; Qian, L,; Sang, W.; Yuan, Y.; Du, M., Cheng, H.; Chen, C.; Qin, X. A practical and sustainable protocol for direct amidation of unactivated esters under transition-metal-free and solvent-free conditions. *Green Chem.* **2021**, *23*, 3972-3982.

(4) Zhao, Q.; Ren, L.; Hou, J.; Yu, W.; Chang, J. Annulation Reactions of In-Situ-Generated *N*-(Het)aroyldiazenes with Isothiocyanates Leading to 2-Imino-1,3,4-oxadiazolines. *Org. Lett.* **2019**, *21*, 210-213.

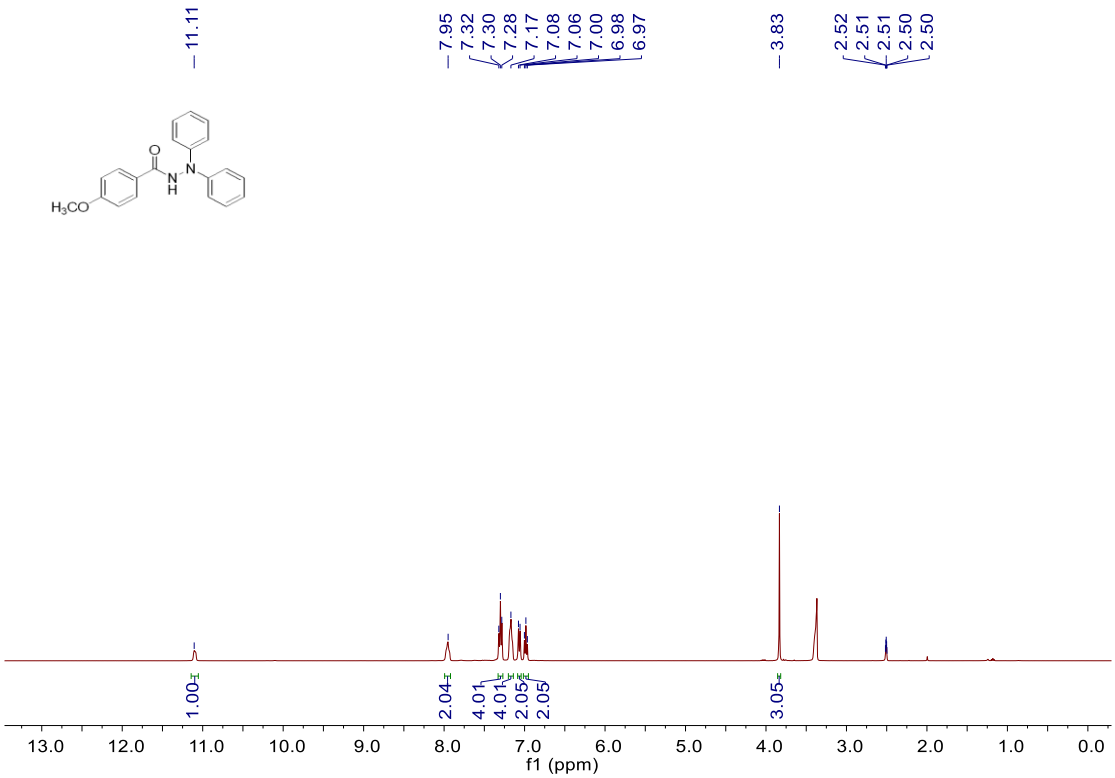
15. NMR Spectra



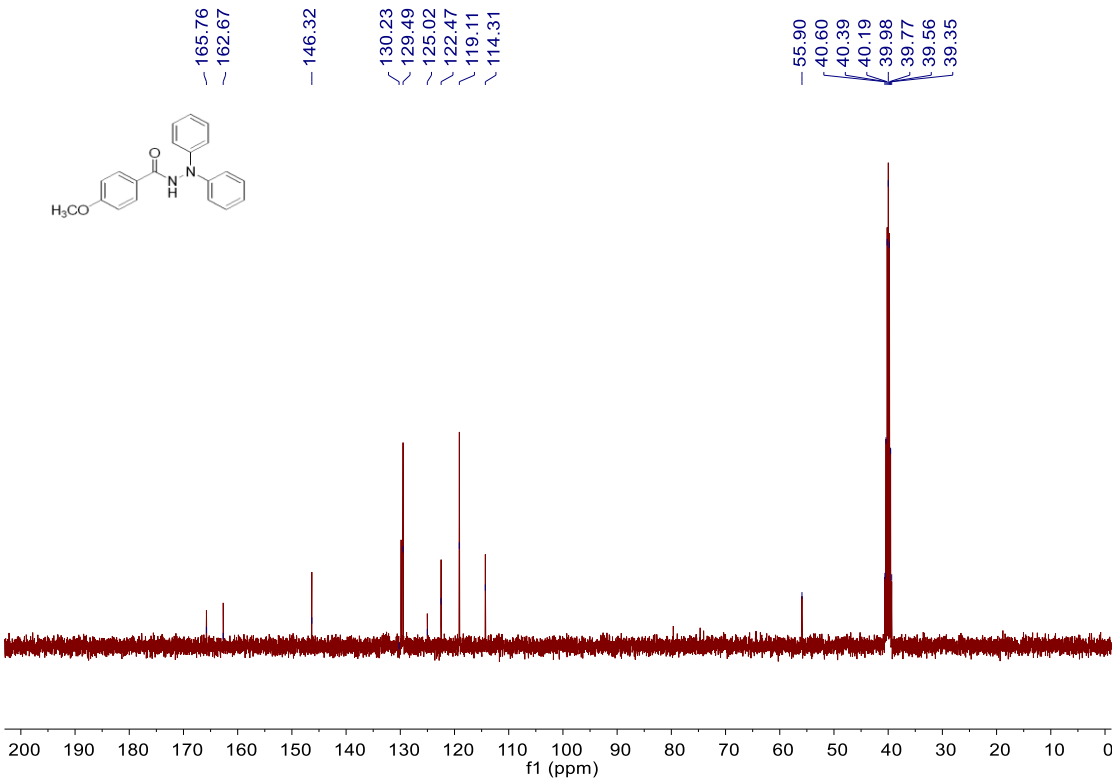
^{1}H NMR of 3 in DMSO-d_6 (400 MHz)



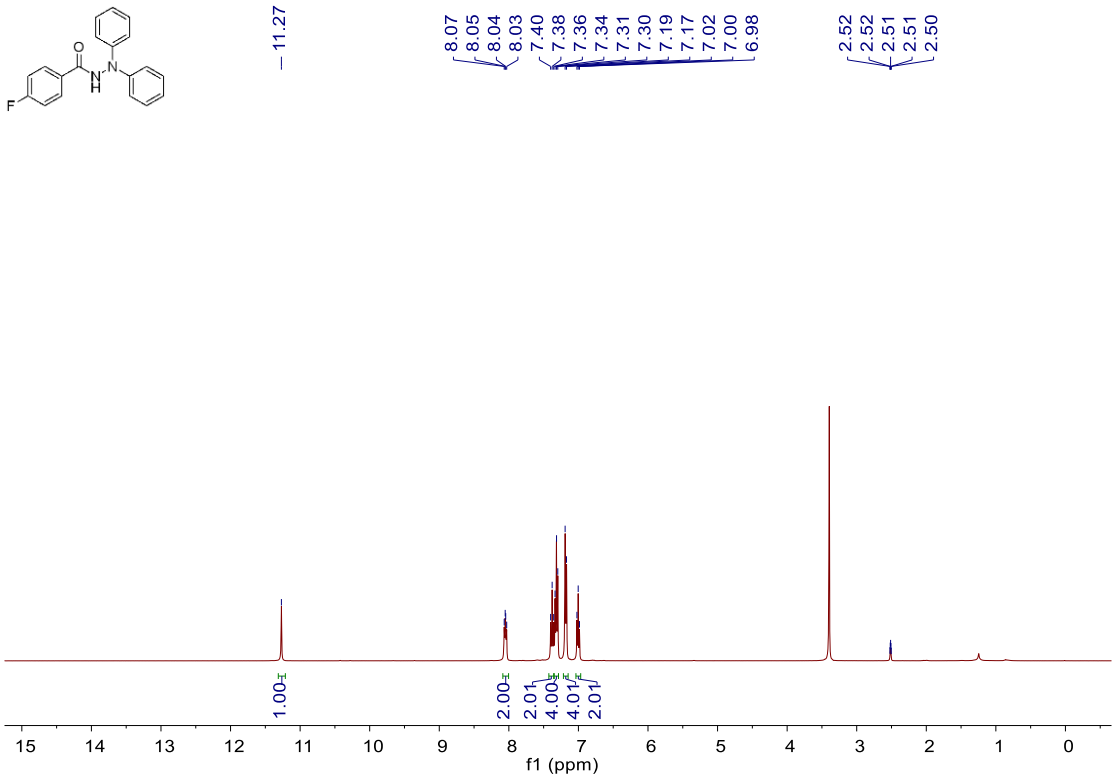
^{13}C{^{1}H} NMR of 3 in DMSO-d_6 (100 MHz)



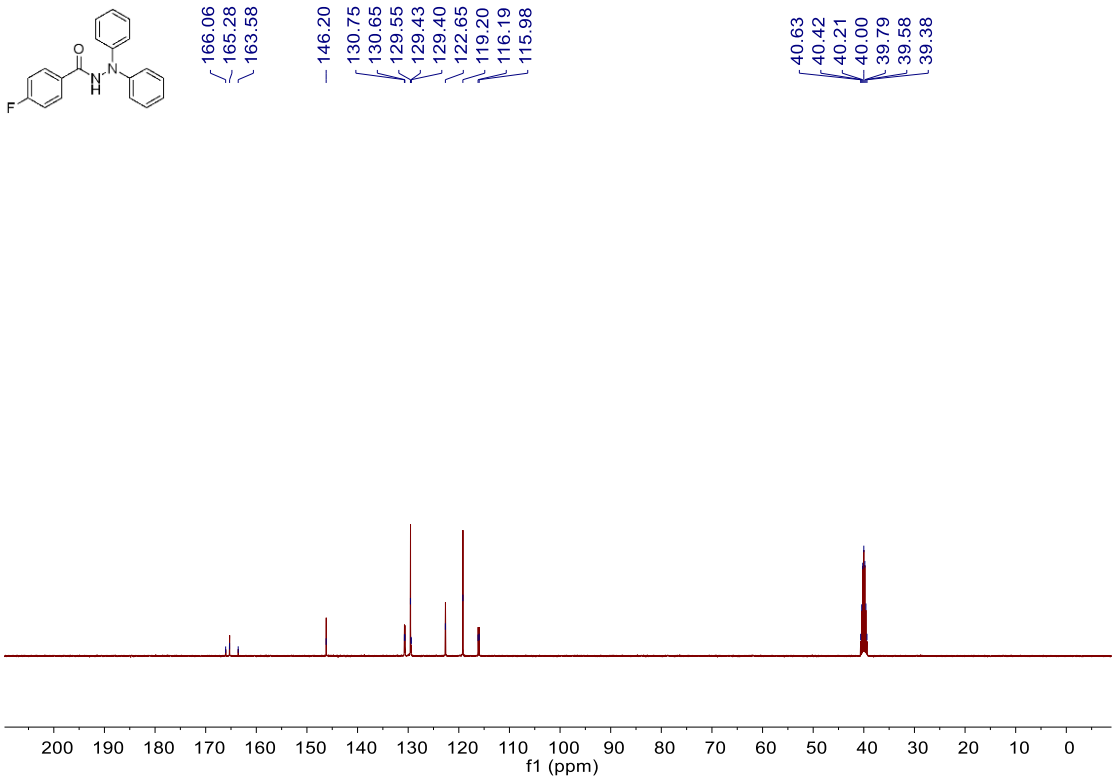
^{1}H NMR of 4 in DMSO-d_6 (400 MHz)



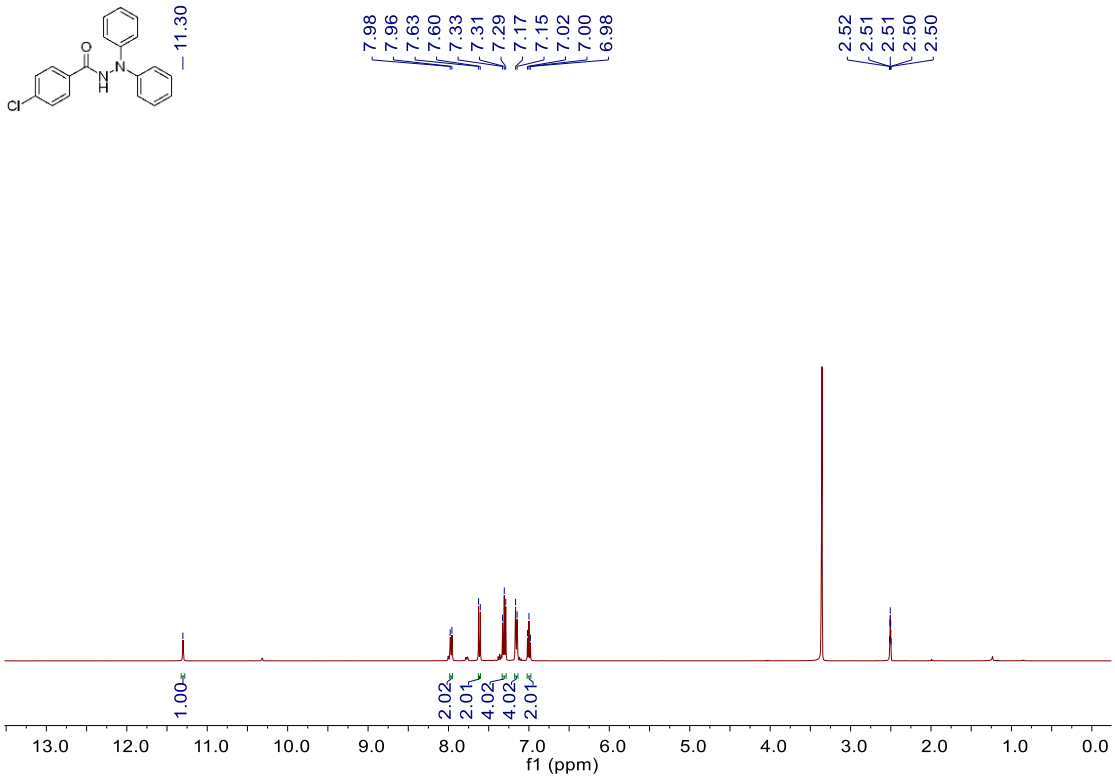
^{13}C{^{1}H} NMR of 4 in DMSO-d_6 (100 MHz)



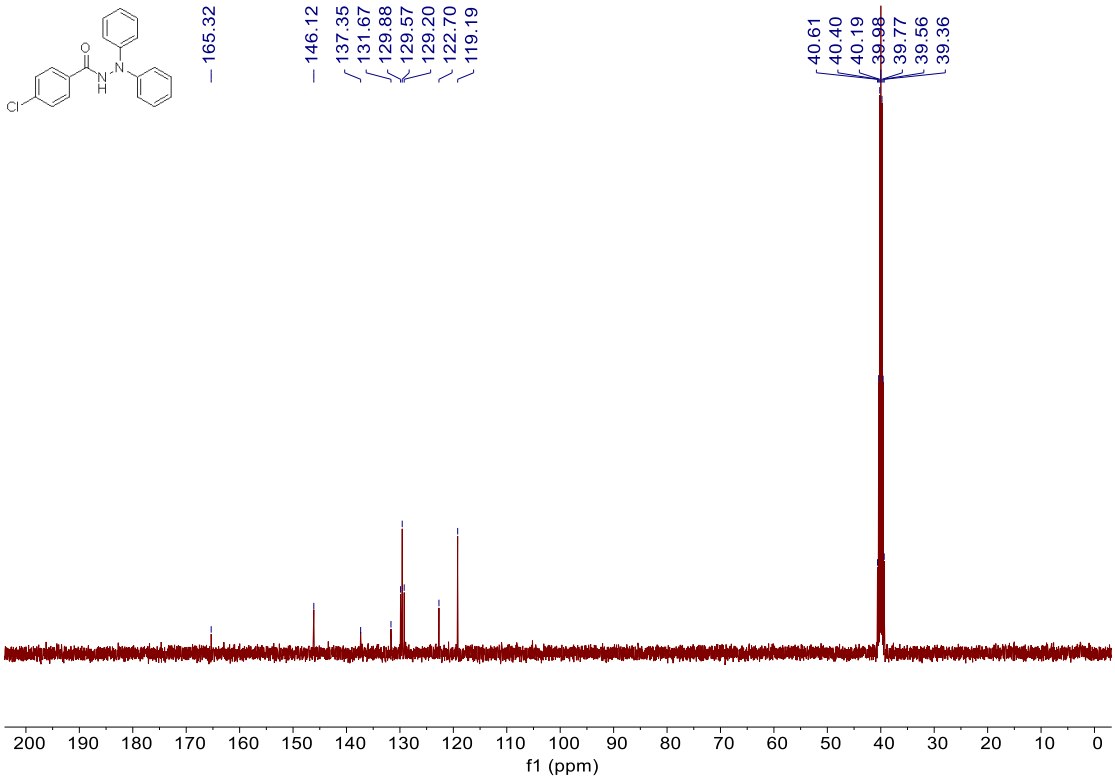
^{1}H NMR of 5 in DMSO-d_6 (400 MHz)



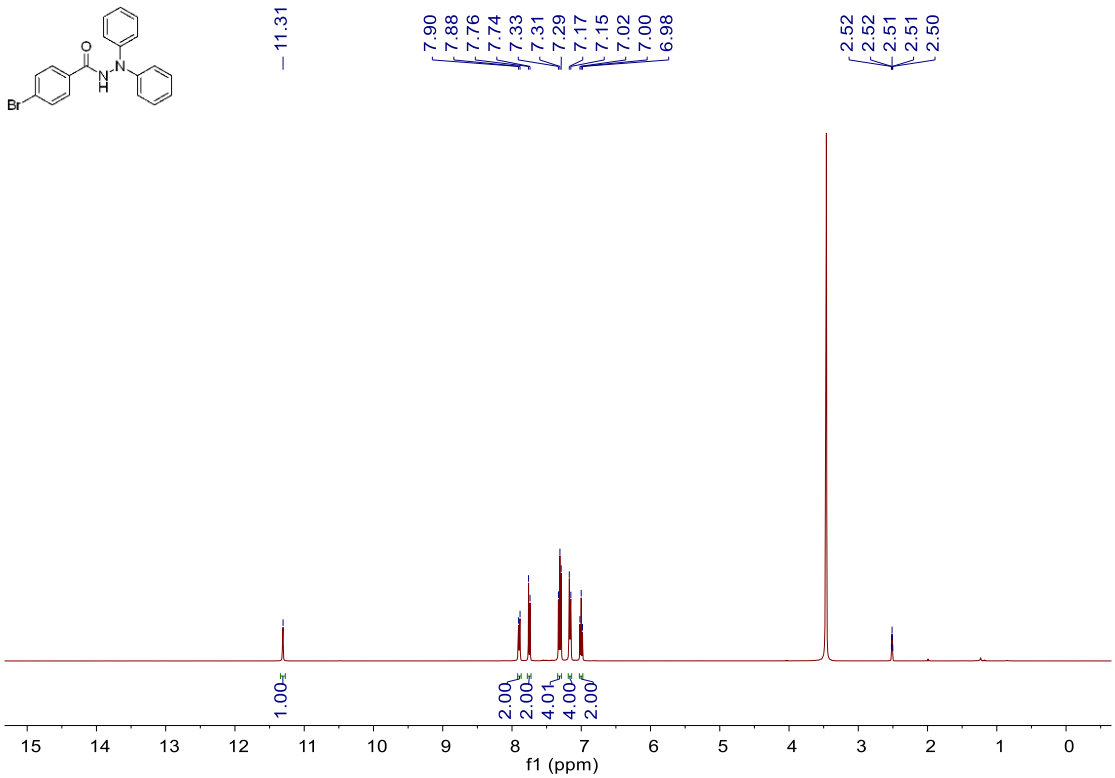
^{13}C{^{1}H} NMR of 5 in DMSO-d_6 (100 MHz)



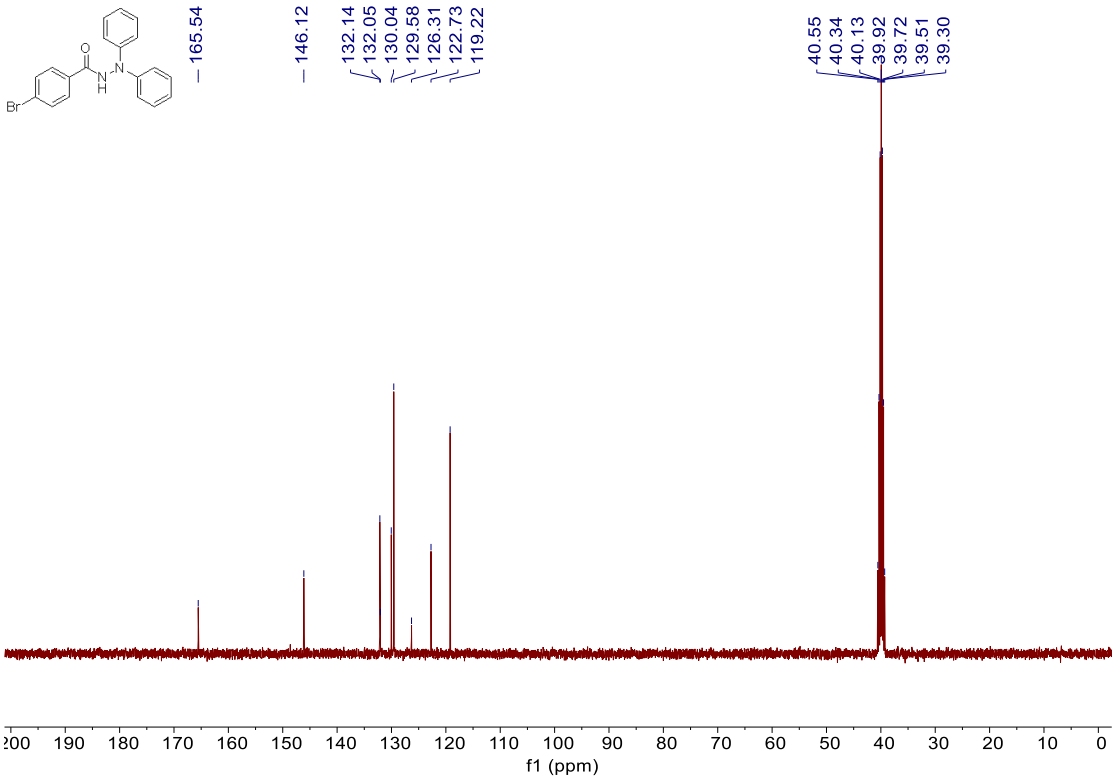
^{1}H NMR of 6 in DMSO-d_6 (400 MHz)



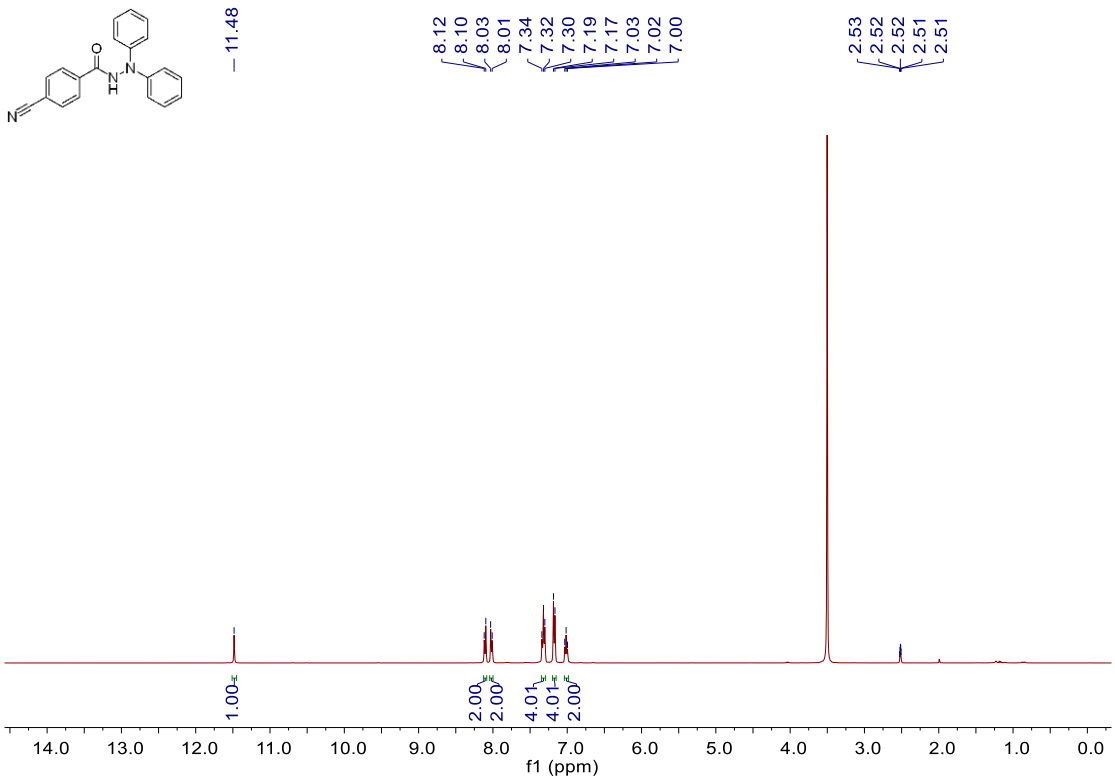
^{13}C{^{1}H} NMR of 6 in DMSO-d_6 (100 MHz)



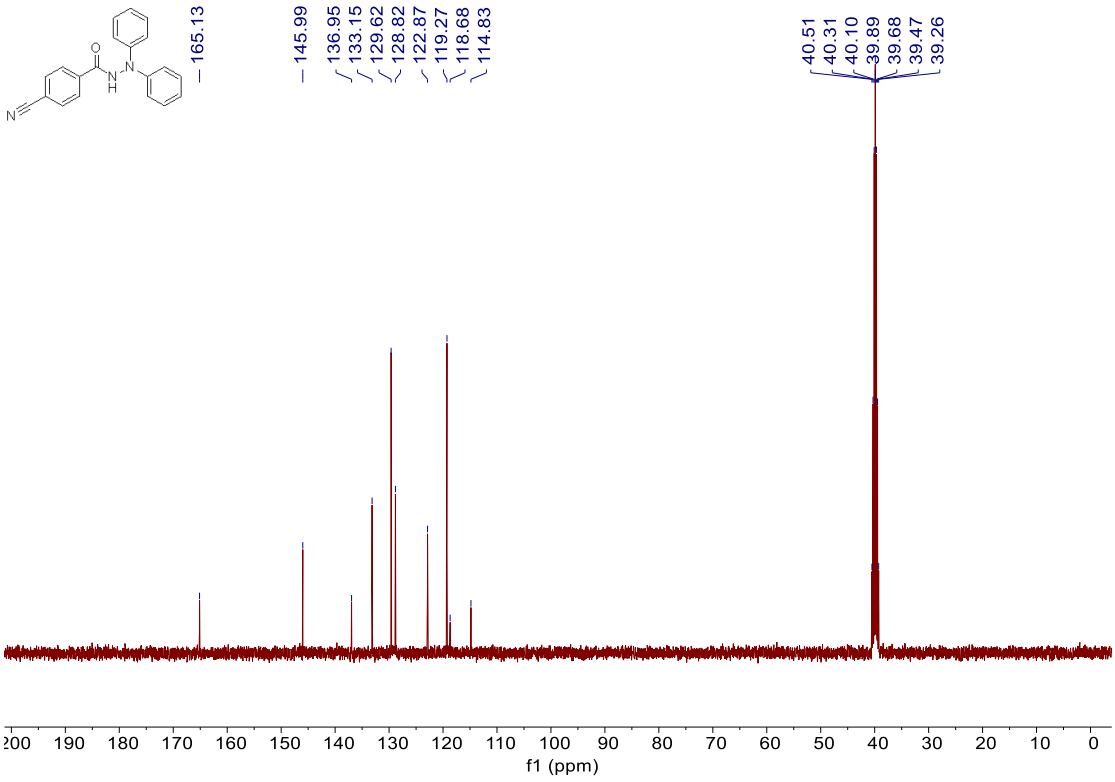
^{1}H NMR of 7 in DMSO-d_6 (400 MHz)



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



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



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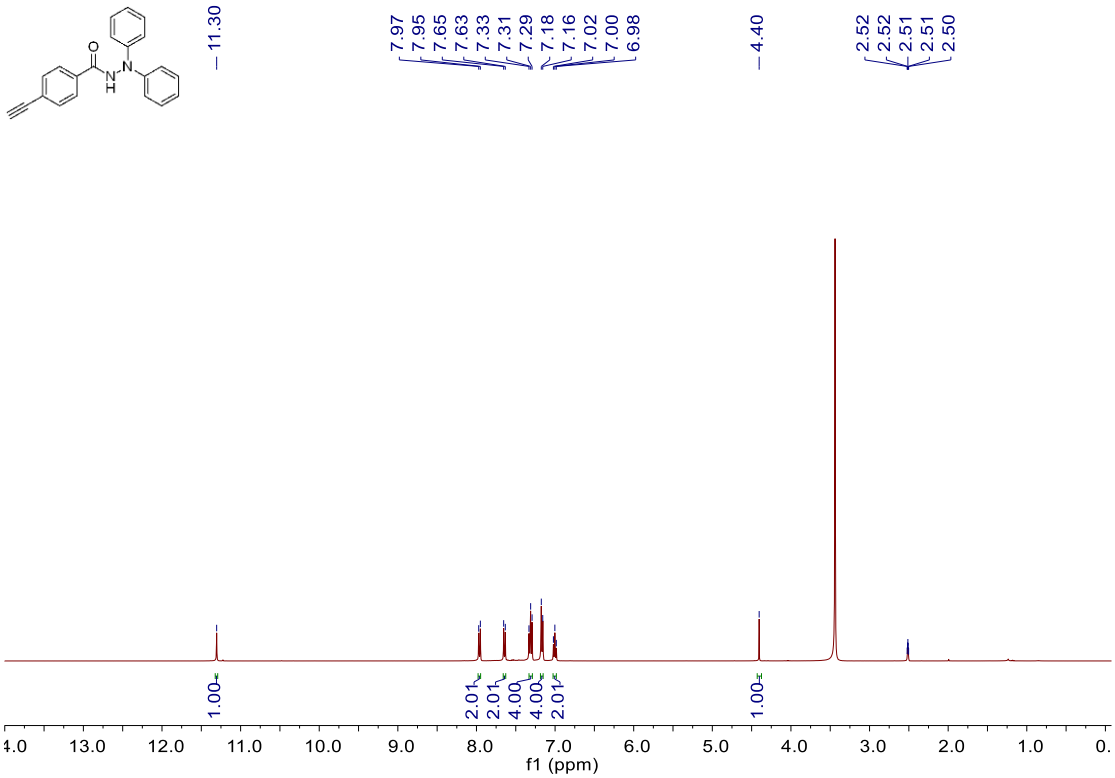
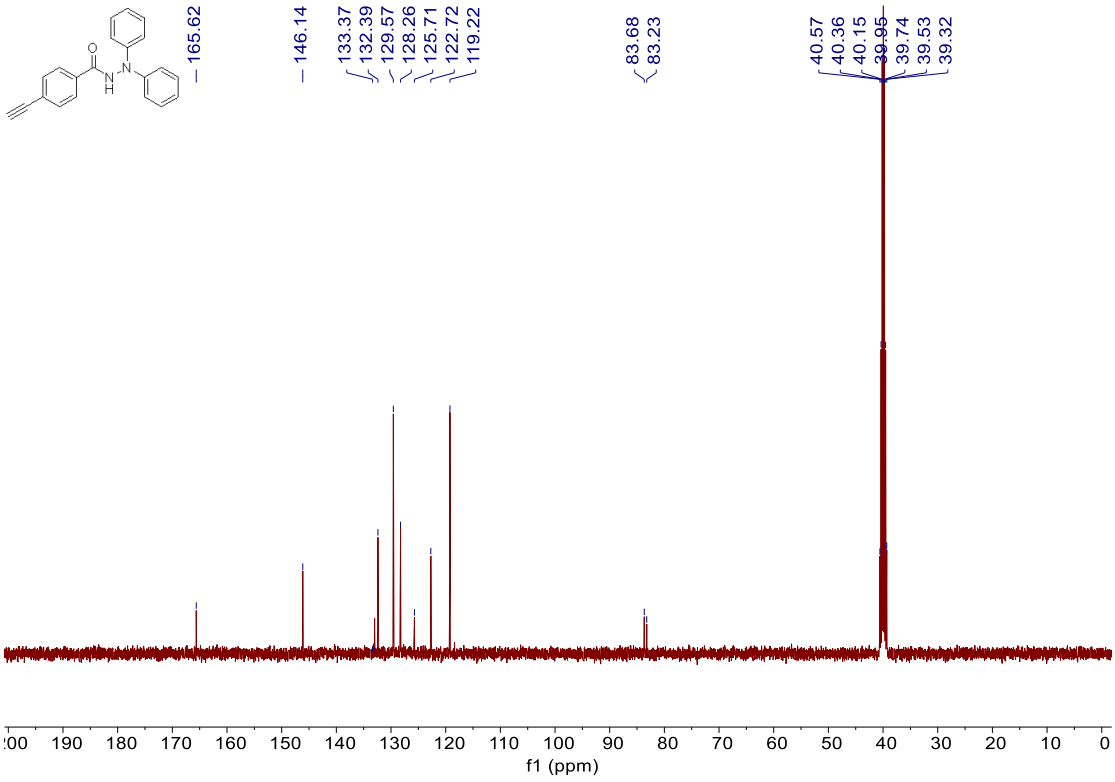
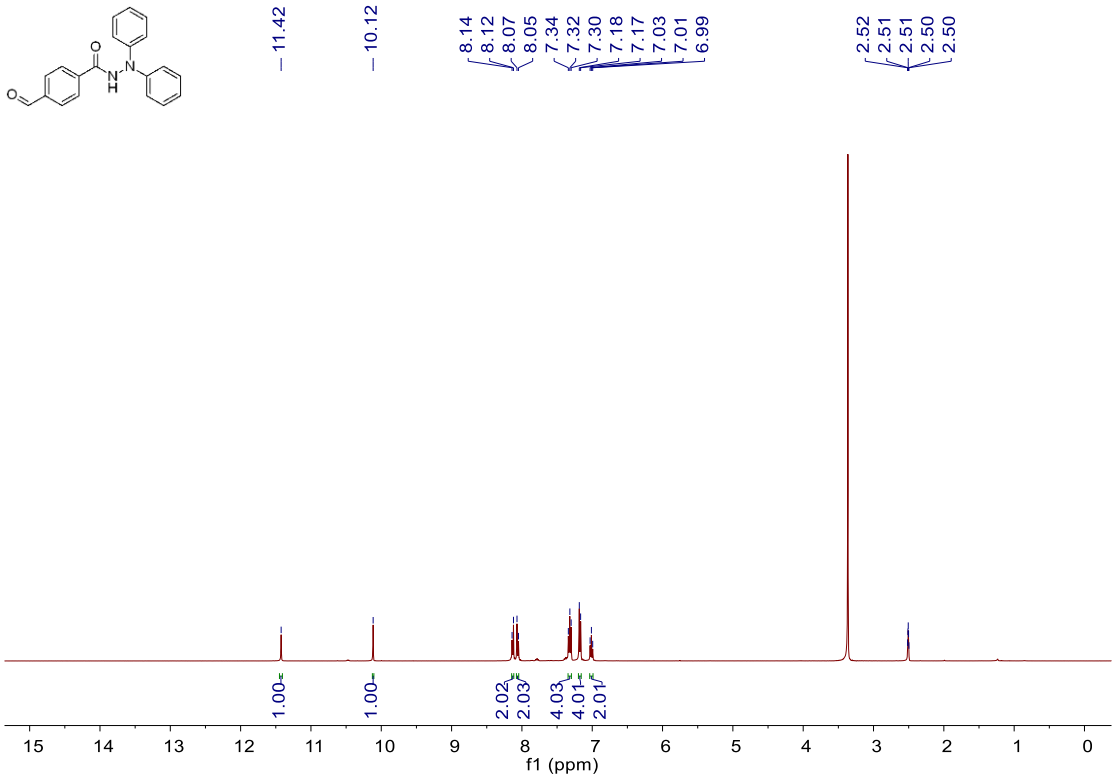


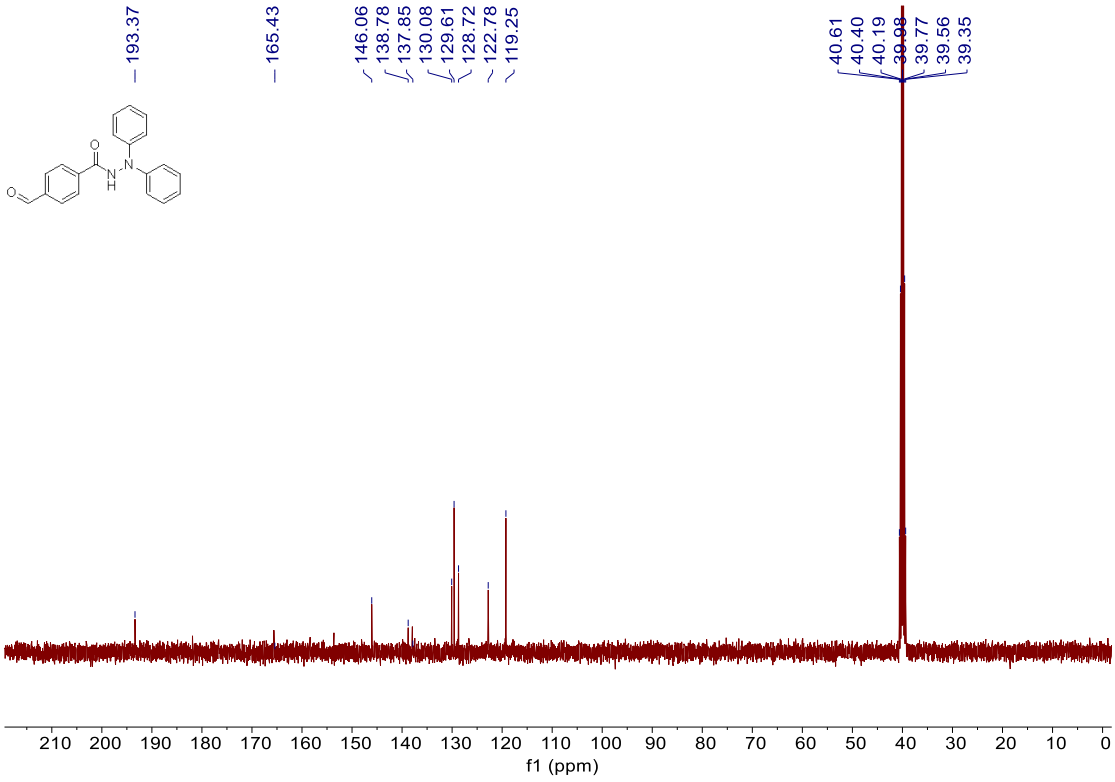
^{1}H NMR of 9 in DMSO-d_6 (400 MHz)



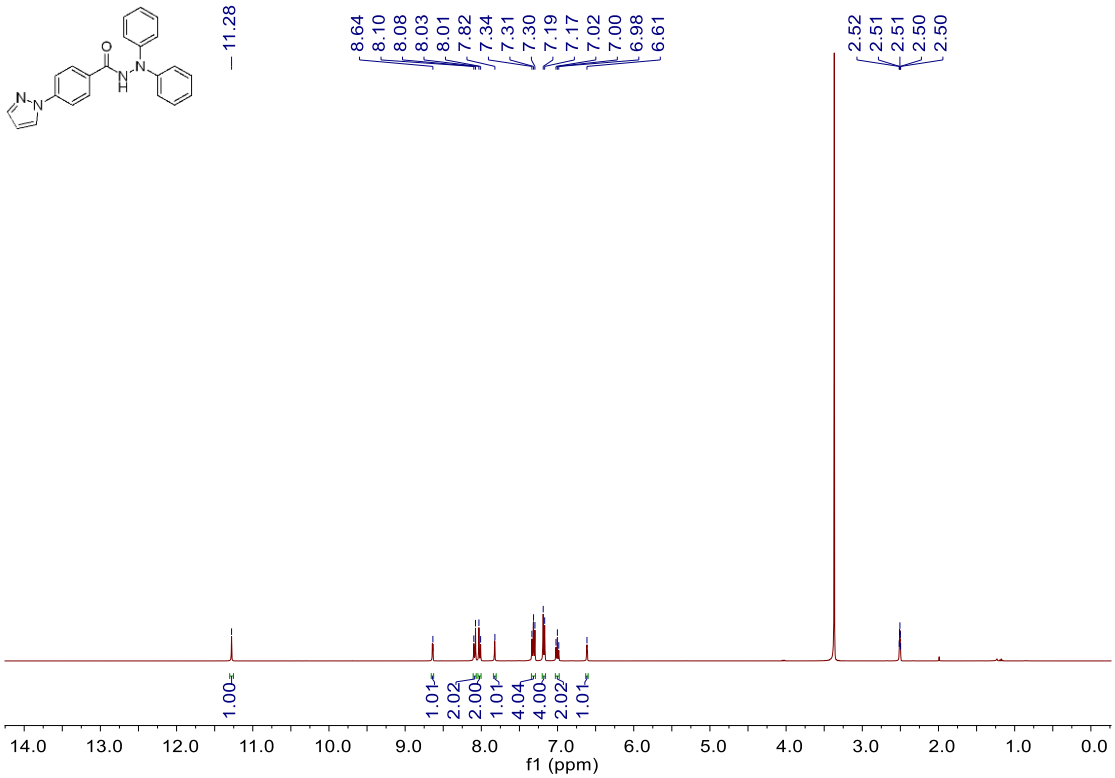
^{13}C{^{1}H} NMR of 9 in DMSO-d_6 (100 MHz)



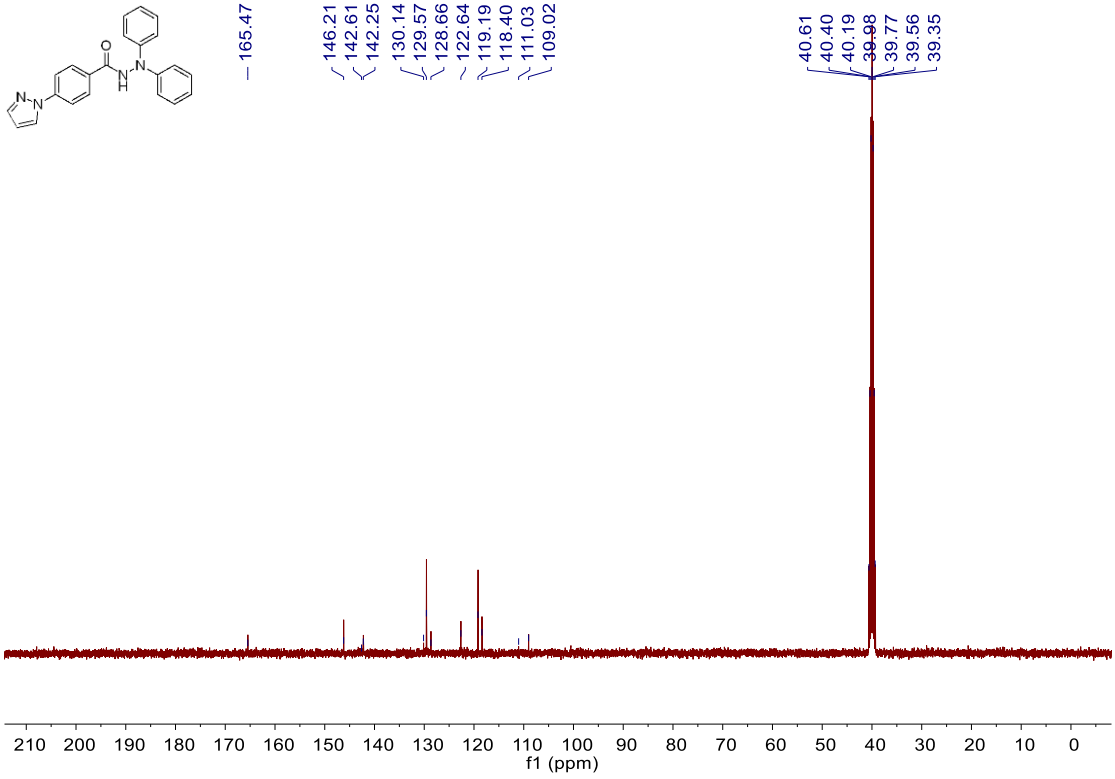
^{1}H NMR of 10 in DMSO-d_6 (400 MHz)



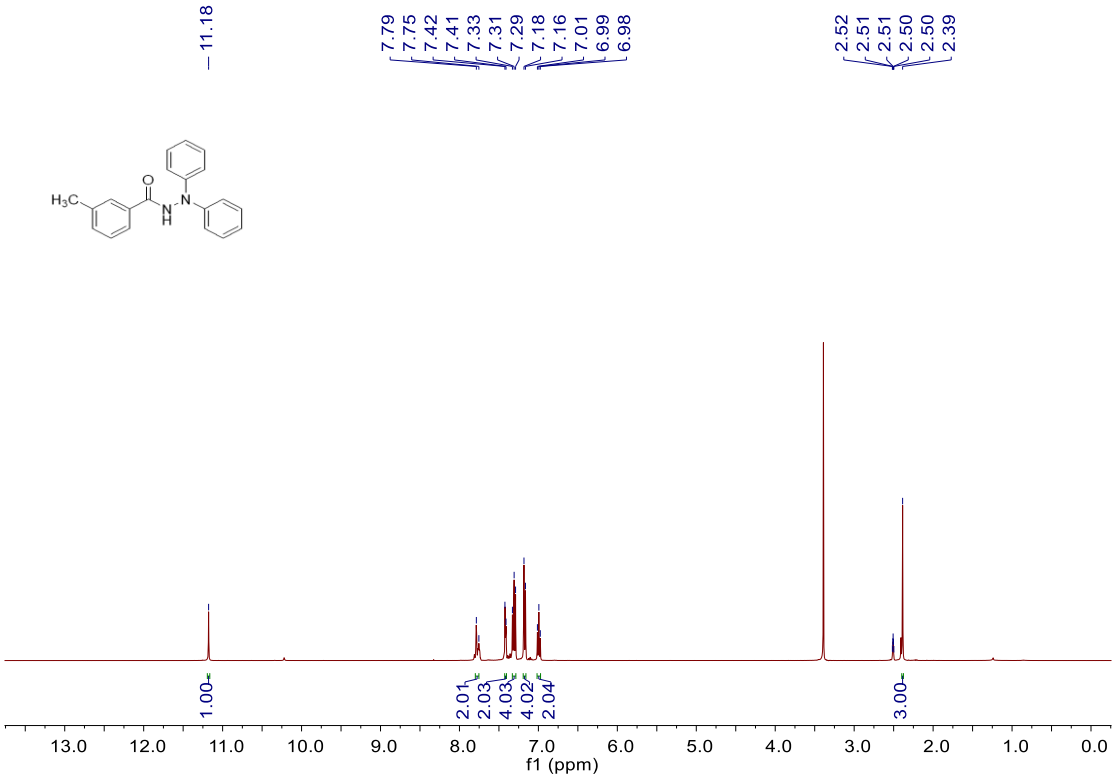
^{13}C{^{1}H} NMR of 10 in DMSO-d_6 (100 MHz)



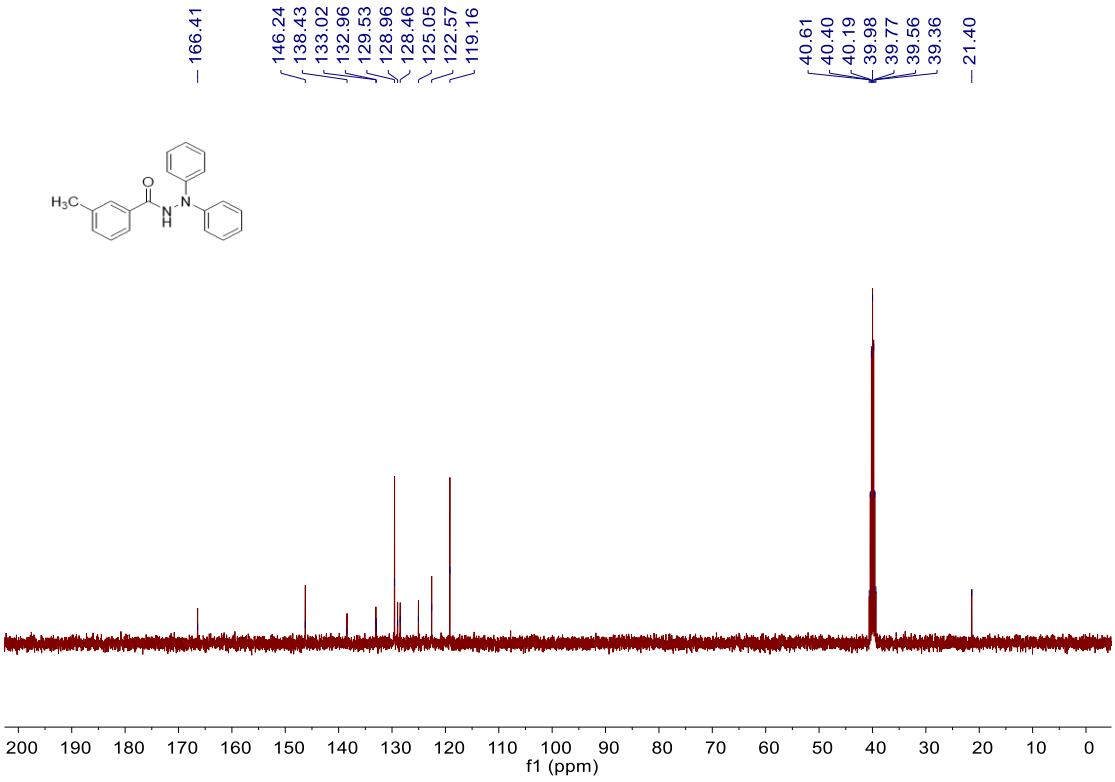
^{1}H NMR of 11 in DMSO-d_6 (400 MHz)



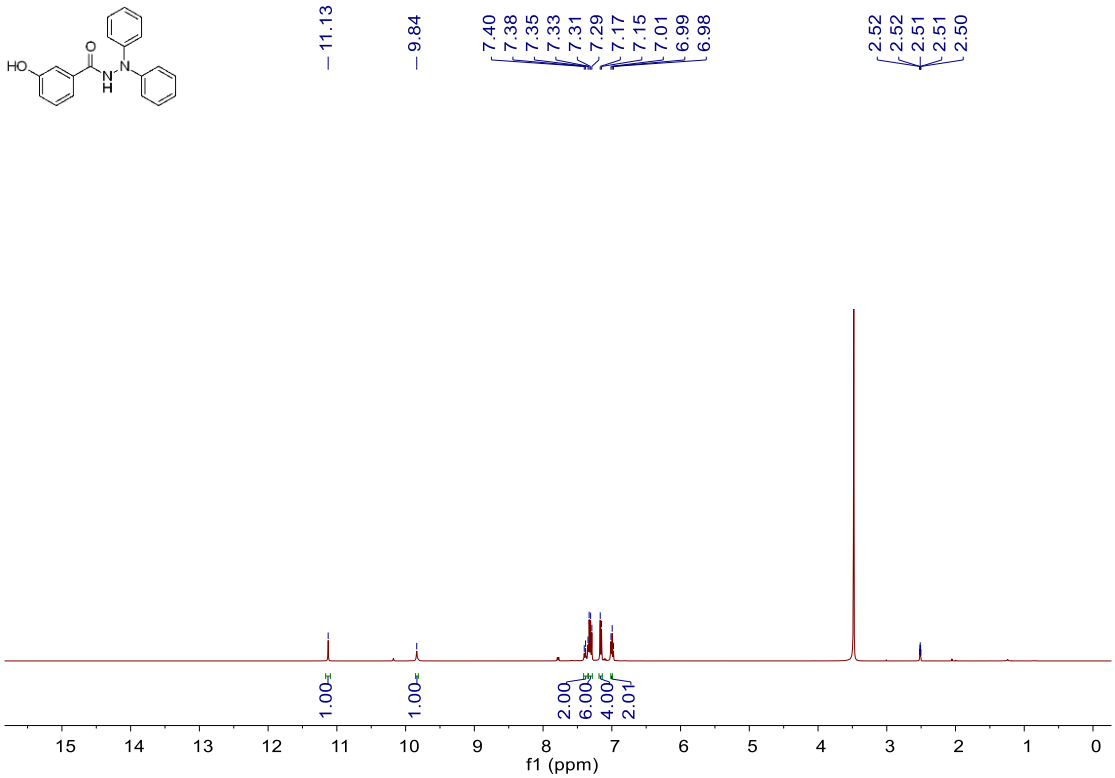
^{13}C{^{1}H} NMR of 11 in DMSO-d_6 (100 MHz)



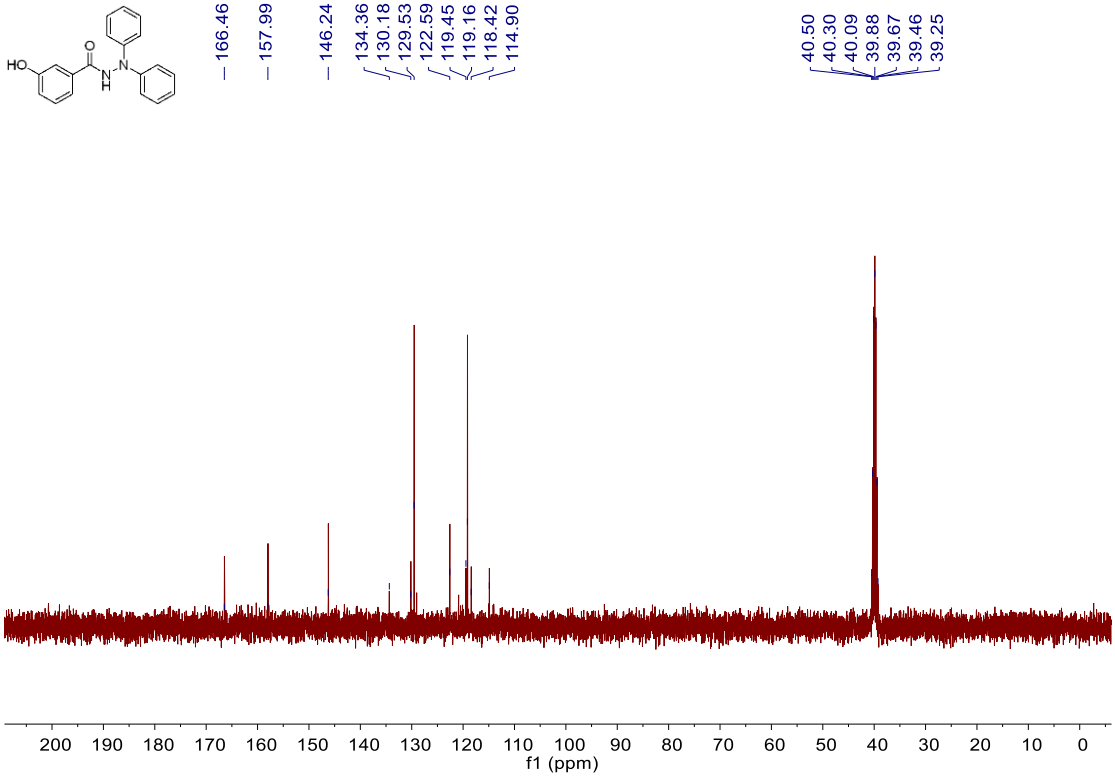
^{1}H NMR of 12 in DMSO-d_6 (400 MHz)



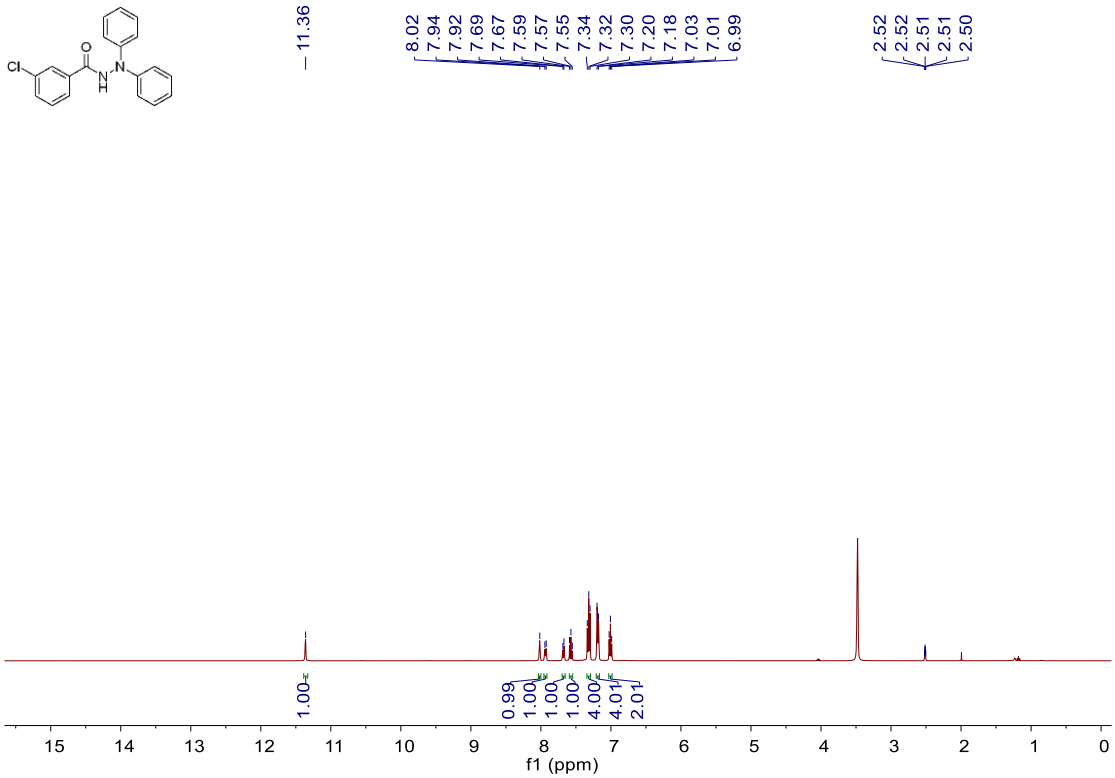
^{13}C{^{1}H} NMR of 12 in DMSO-d_6 (100 MHz)



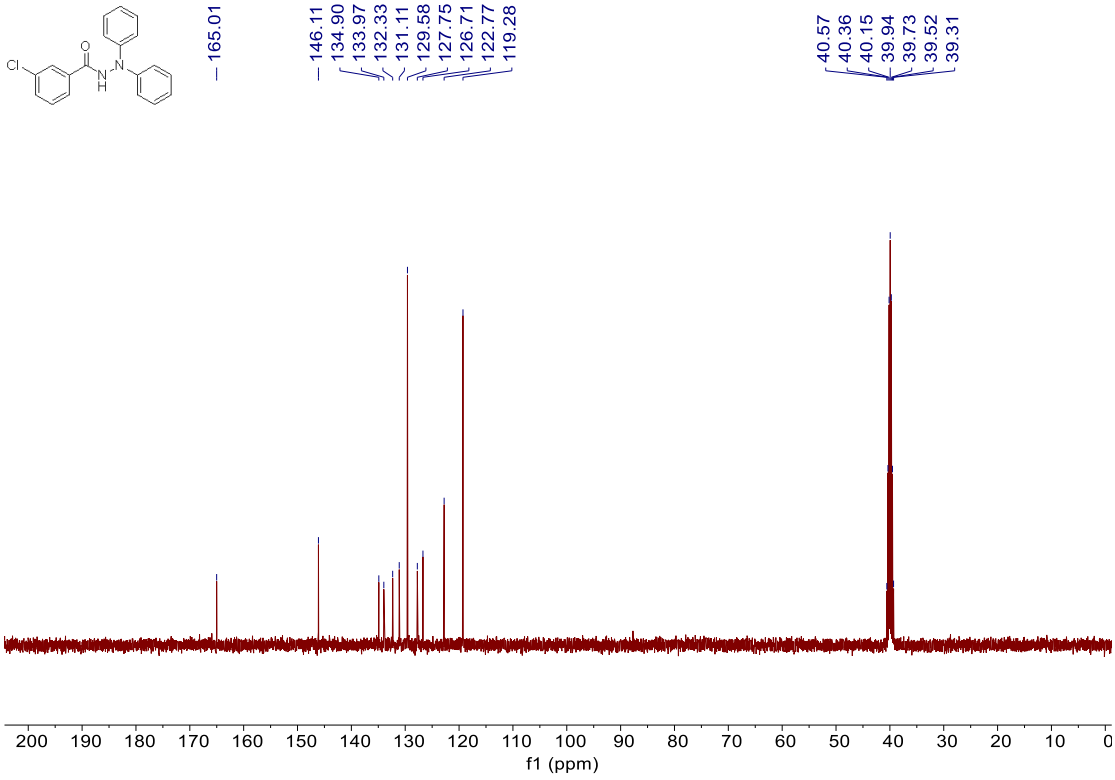
^{1}H NMR of 13 in DMSO-d_6 (400 MHz)



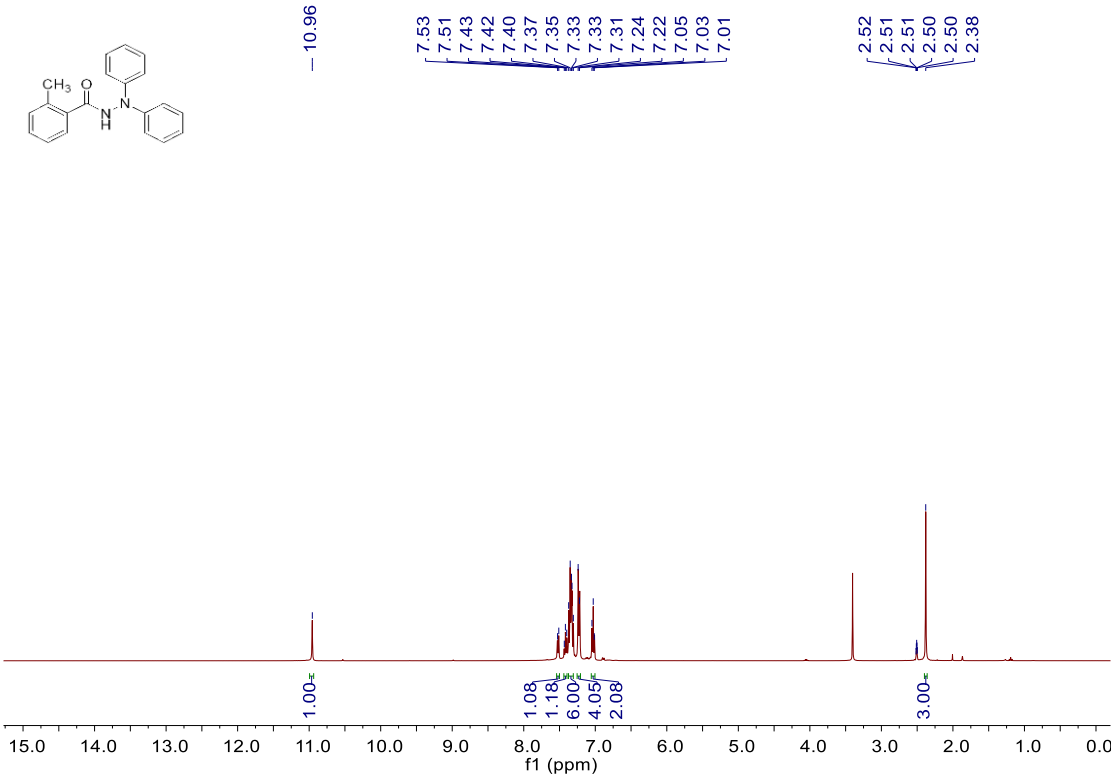
^{13}C{^{1}H} NMR of 13 in DMSO-d_6 (100 MHz)



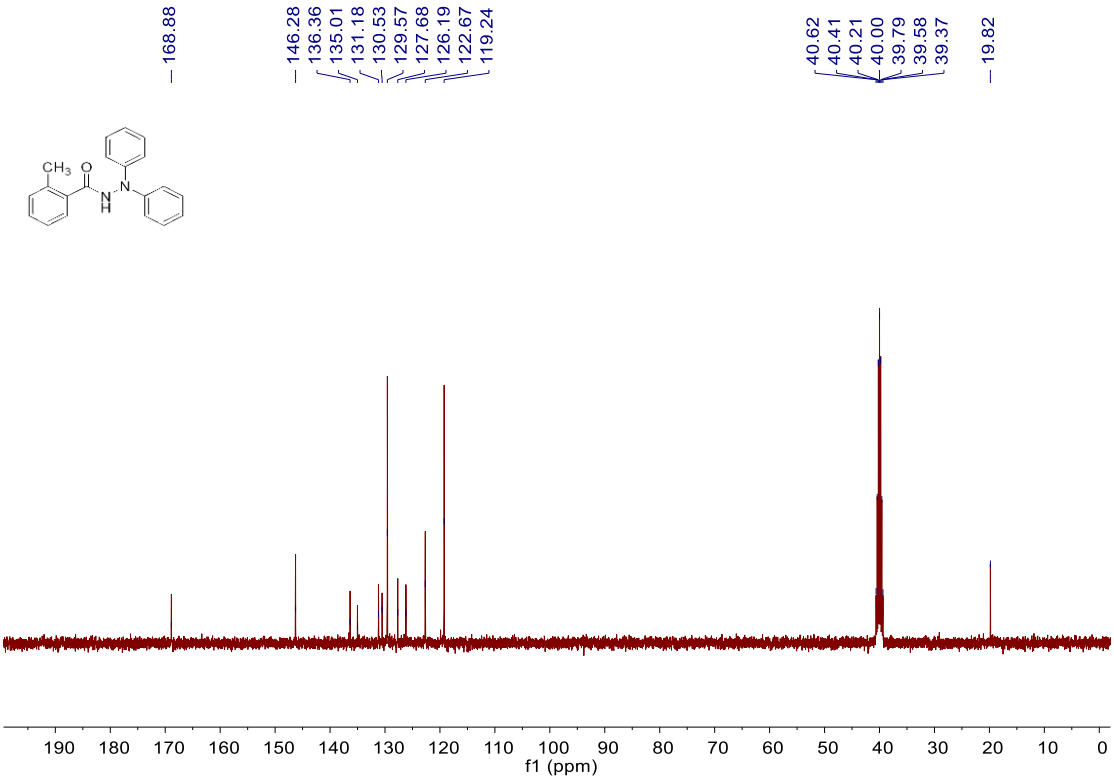
^{1}H NMR of 14 in DMSO-d_6 (400 MHz)



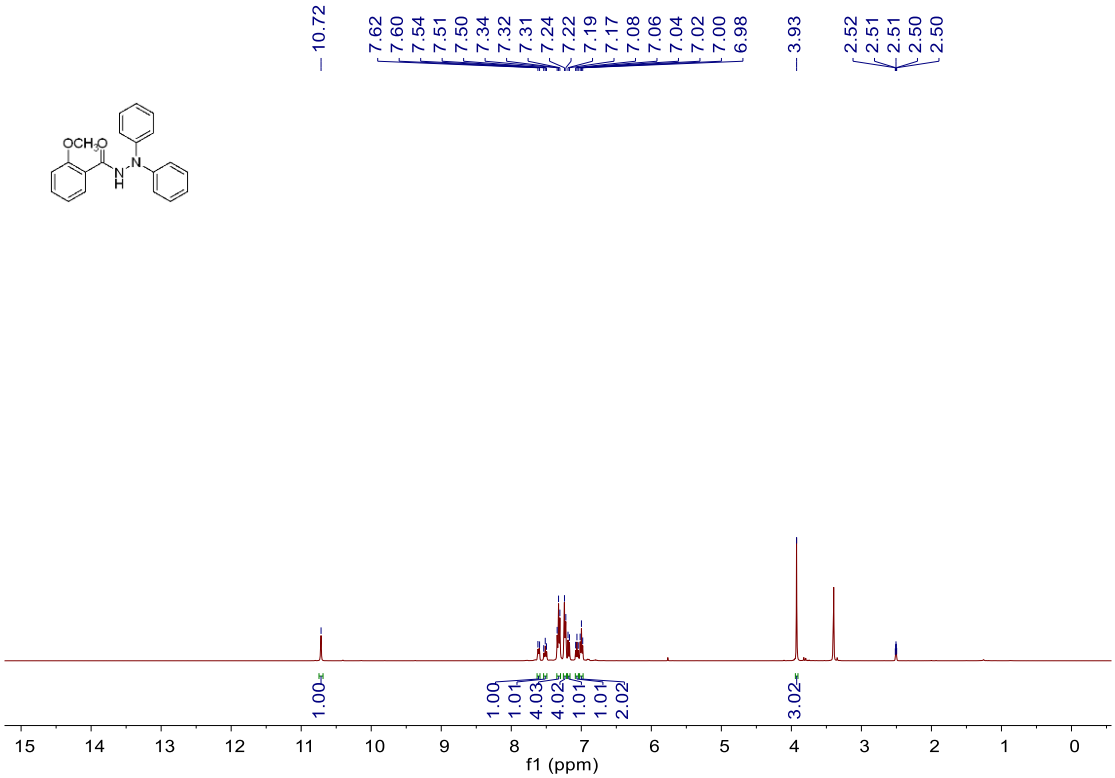
^{13}C{^{1}H} NMR of 14 in DMSO-d_6 (100 MHz)



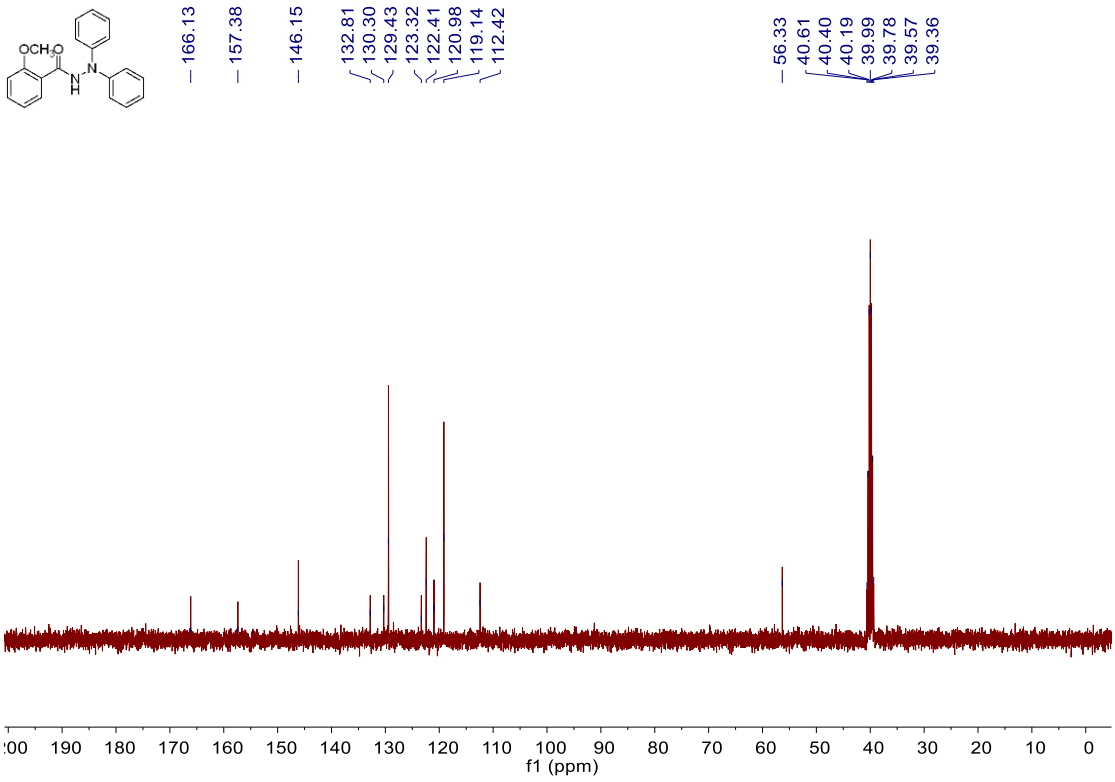
^{1}H NMR of 15 in DMSO-d_6 (400 MHz)



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



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



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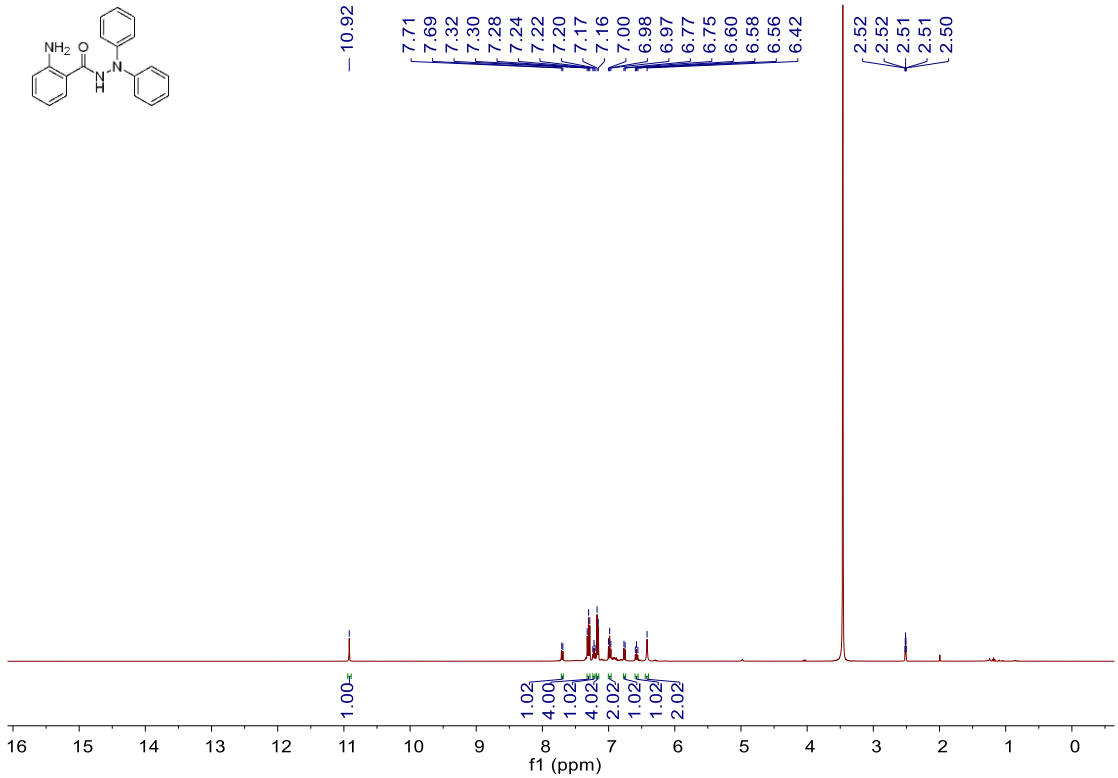
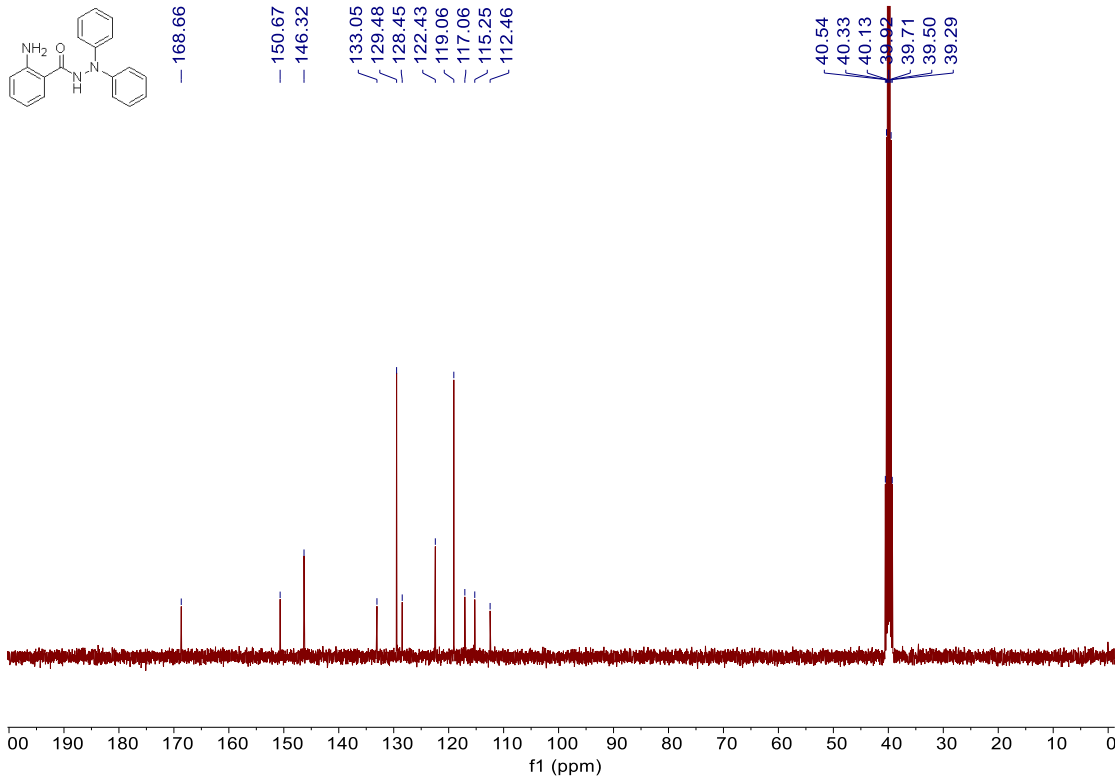
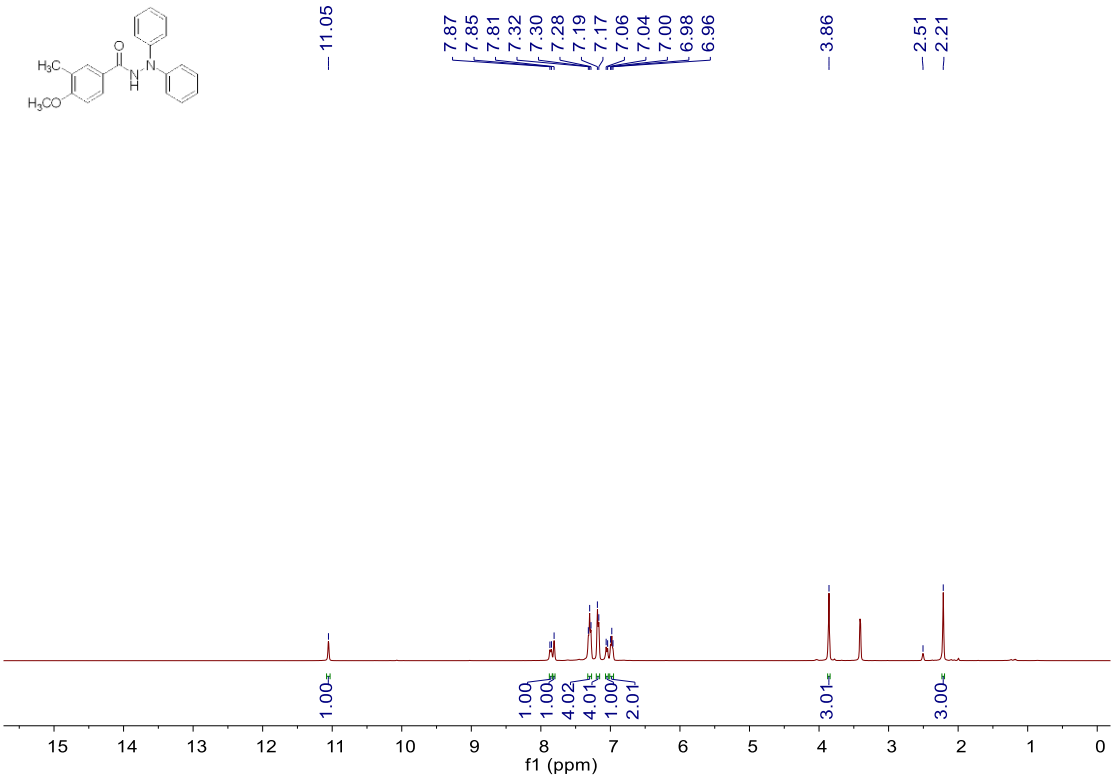


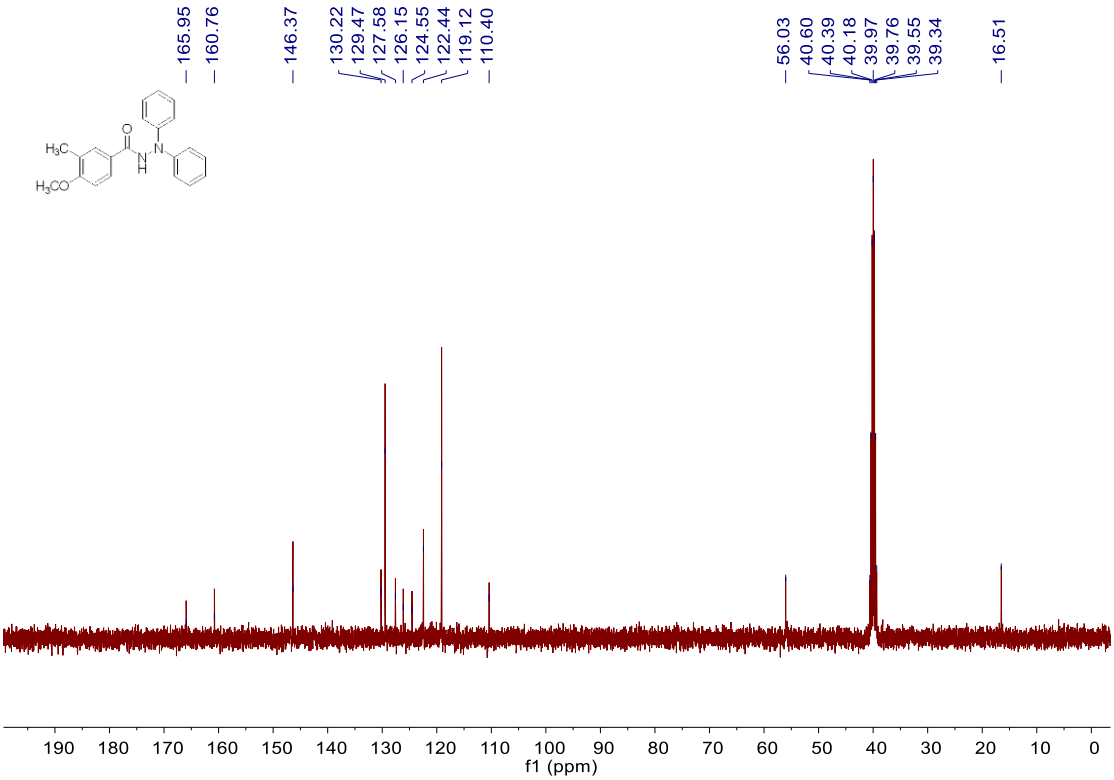
^{1}H NMR of 17 in DMSO-d_6 (400 MHz)



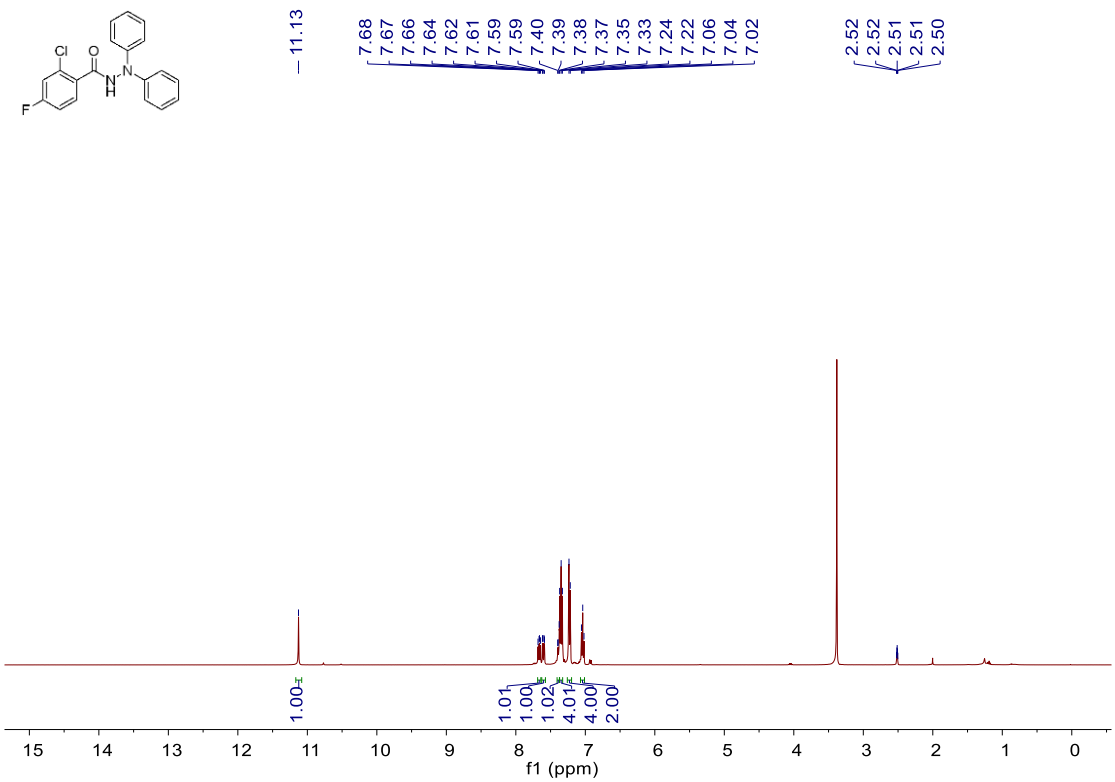
^{13}C{^{1}H} NMR of 17 in DMSO-d_6 (100 MHz)



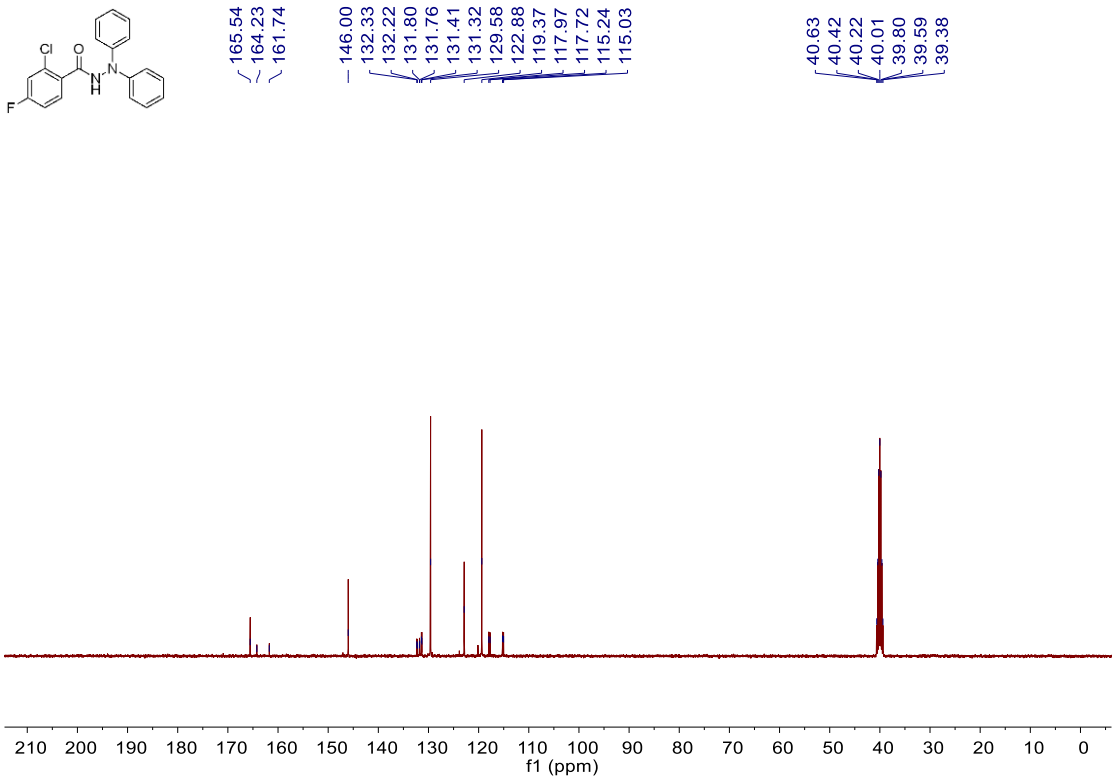
^{1}H NMR of 18 in DMSO-d_6 (400 MHz)



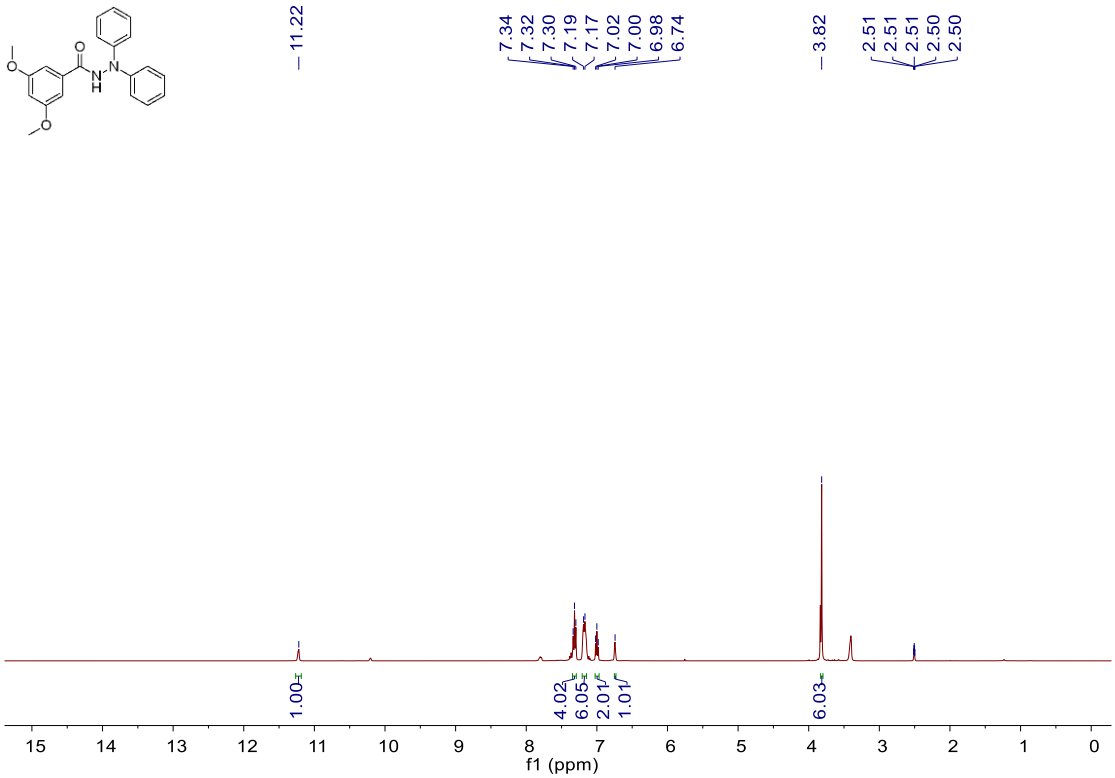
^{13}C{^{1}H} NMR of 18 in DMSO-d_6 (100 MHz)



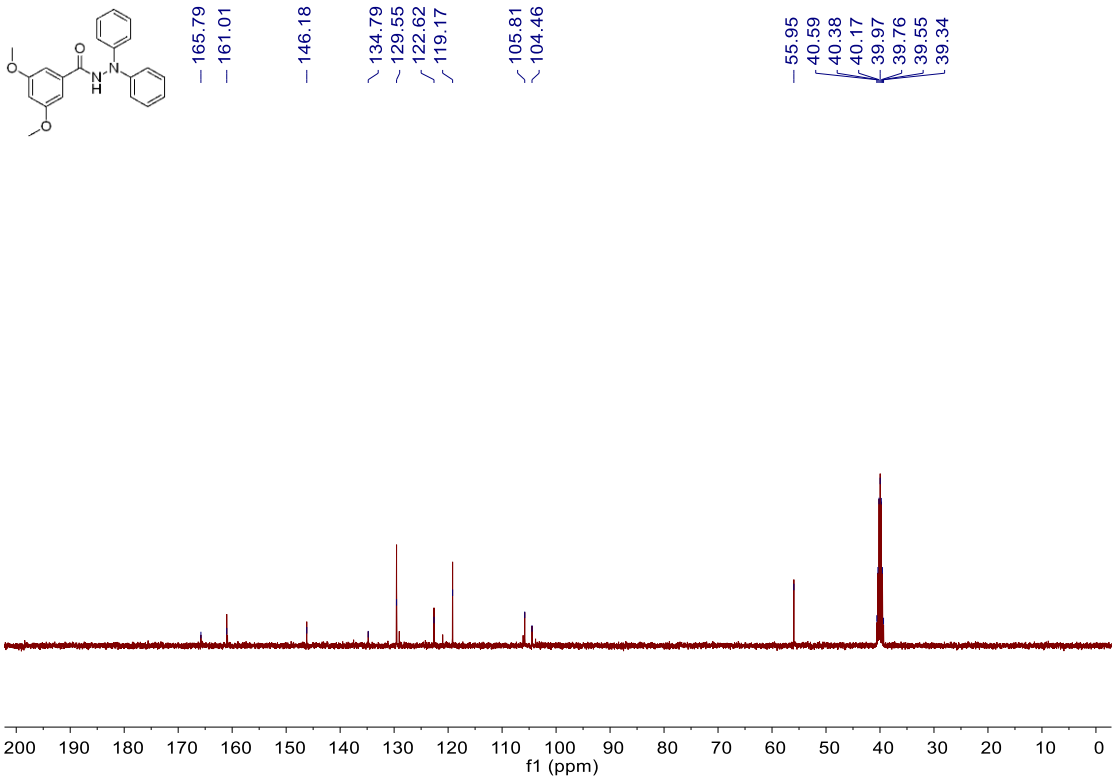
^{1}H NMR of 19 in DMSO-d_6 (400 MHz)



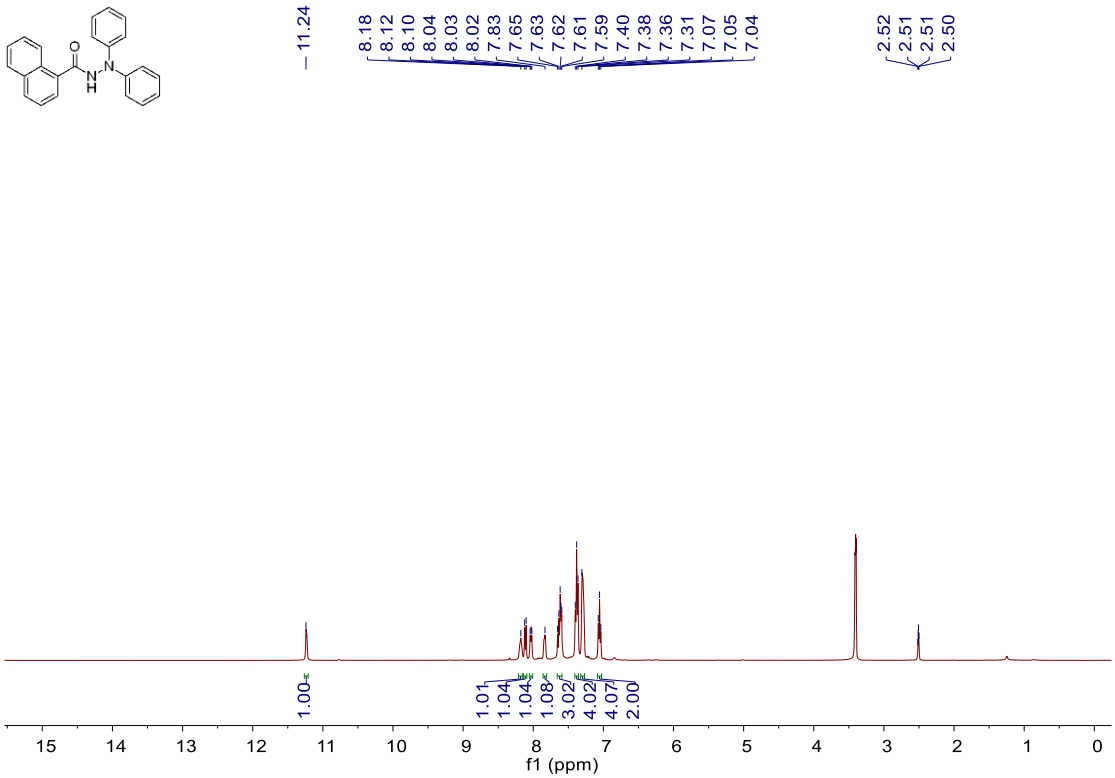
^{13}C{^{1}H} NMR of 19 in DMSO-d_6 (100 MHz)



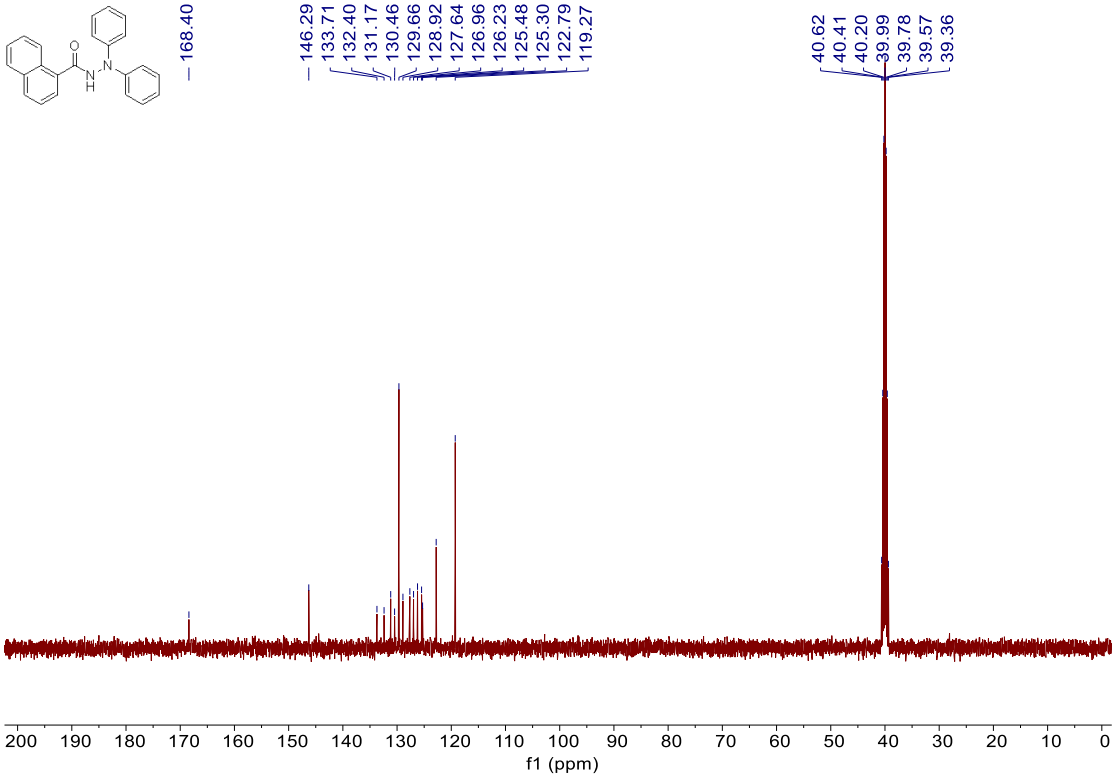
^{1}H NMR of 20 in DMSO-d_6 (400 MHz)



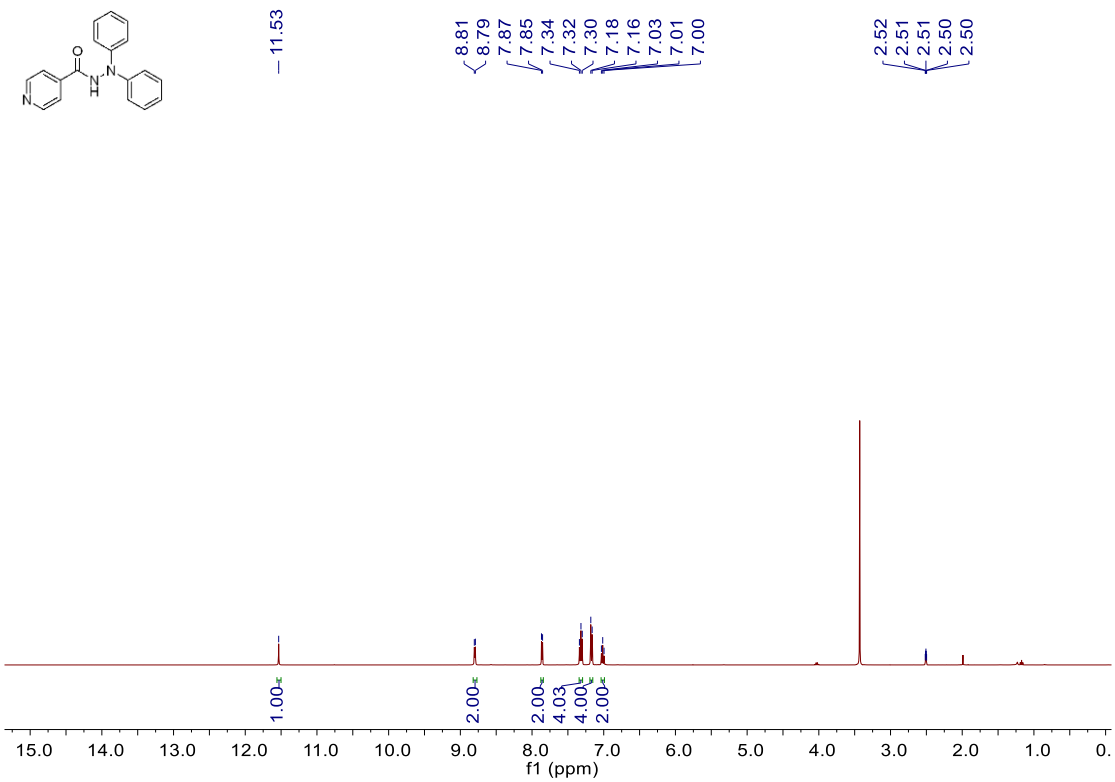
^{13}C{^{1}H} NMR of 20 in DMSO-d_6 (100 MHz)



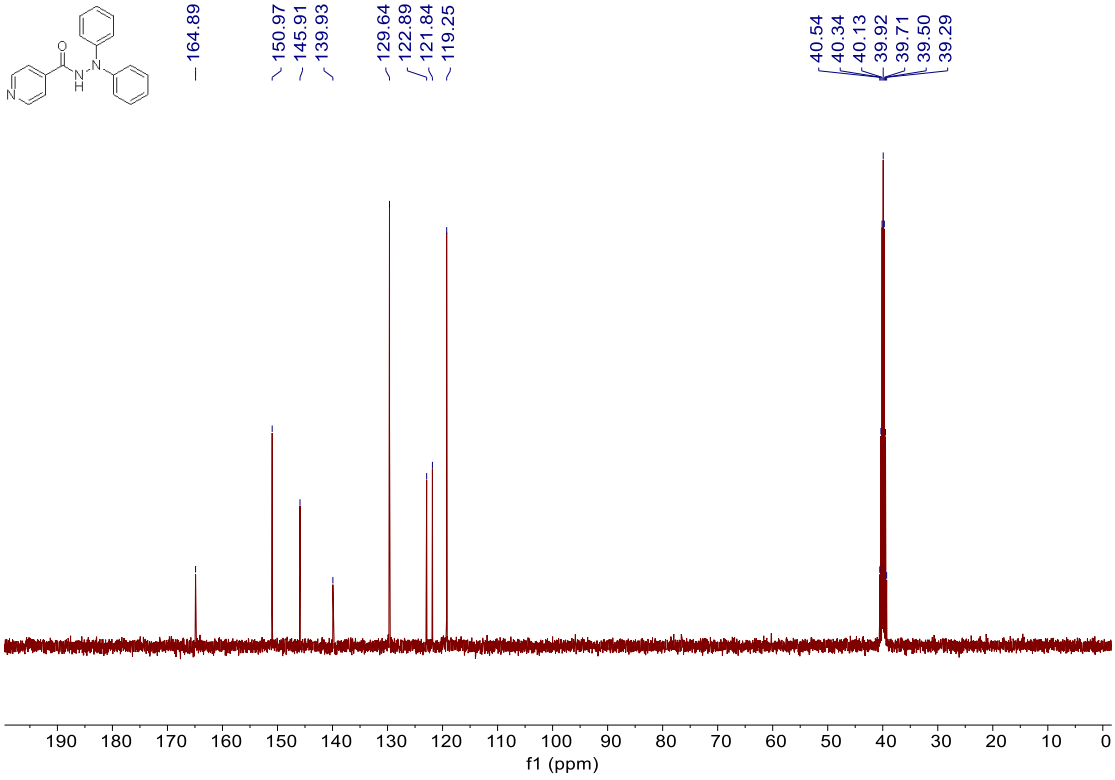
^{1}H NMR of 21 in DMSO-d_6 (400 MHz)



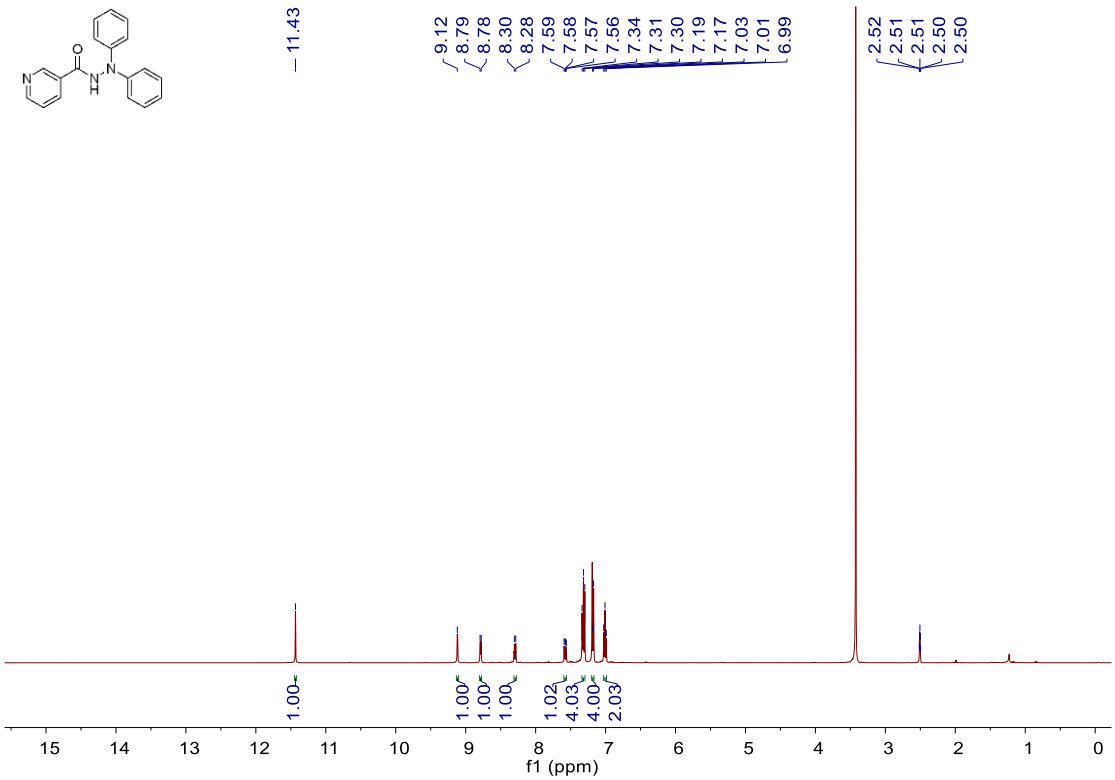
^{13}C{^{1}H} NMR of 21 in DMSO-d_6 (100 MHz)



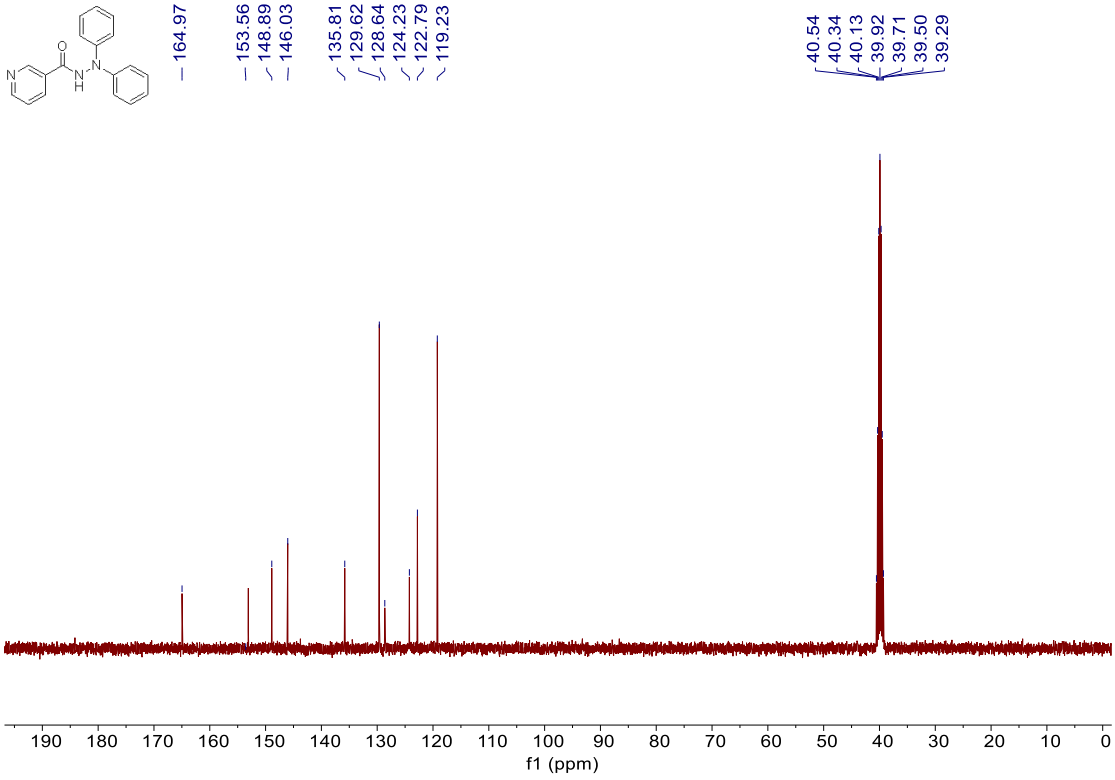
^{1}H NMR of 22 in DMSO-d_6 (400 MHz)



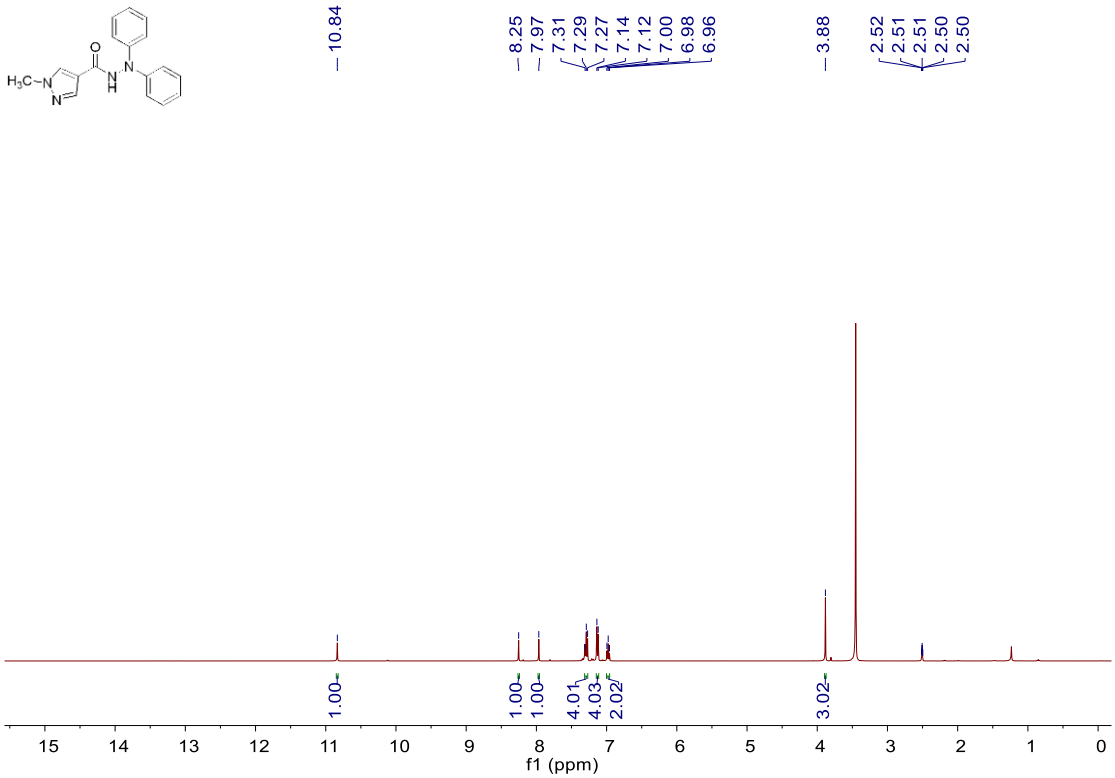
^{13}C{^{1}H} NMR of 22 in DMSO-d_6 (100 MHz)



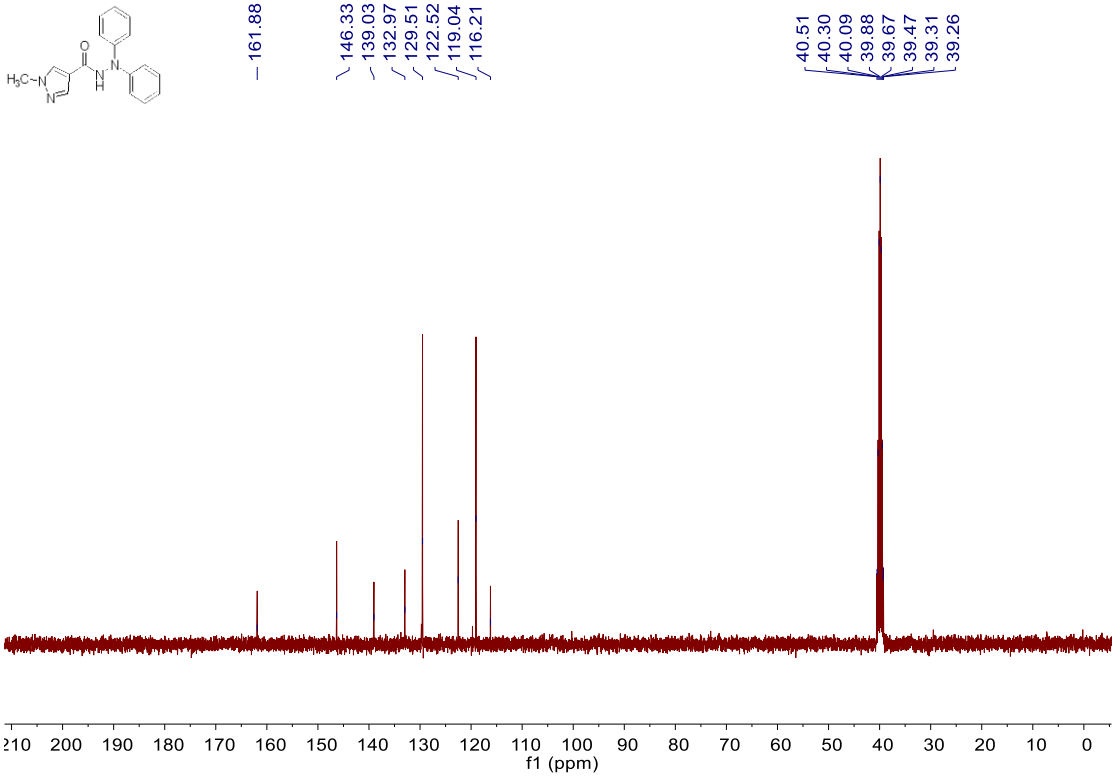
^{1}H NMR of 23 in DMSO-d_6 (400 MHz)



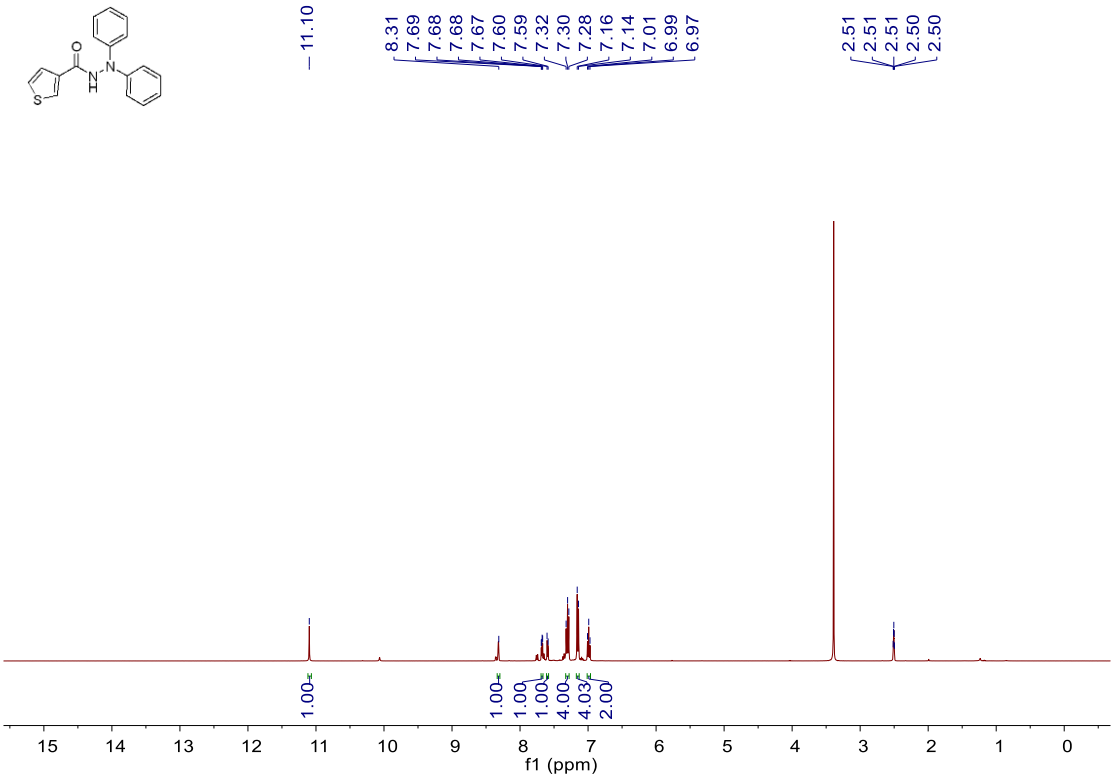
^{13}C{^{1}H} NMR of 23 in DMSO-d_6 (100 MHz)



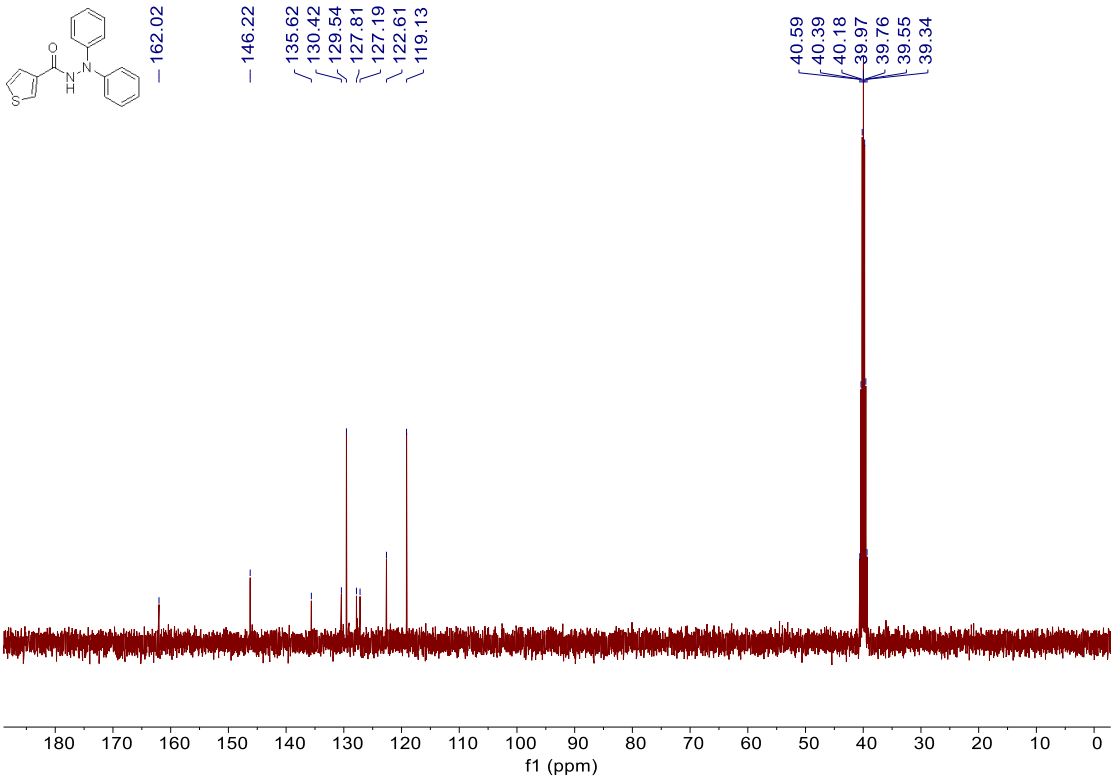
^{1}H NMR of 24 in DMSO-d_6 (400 MHz)



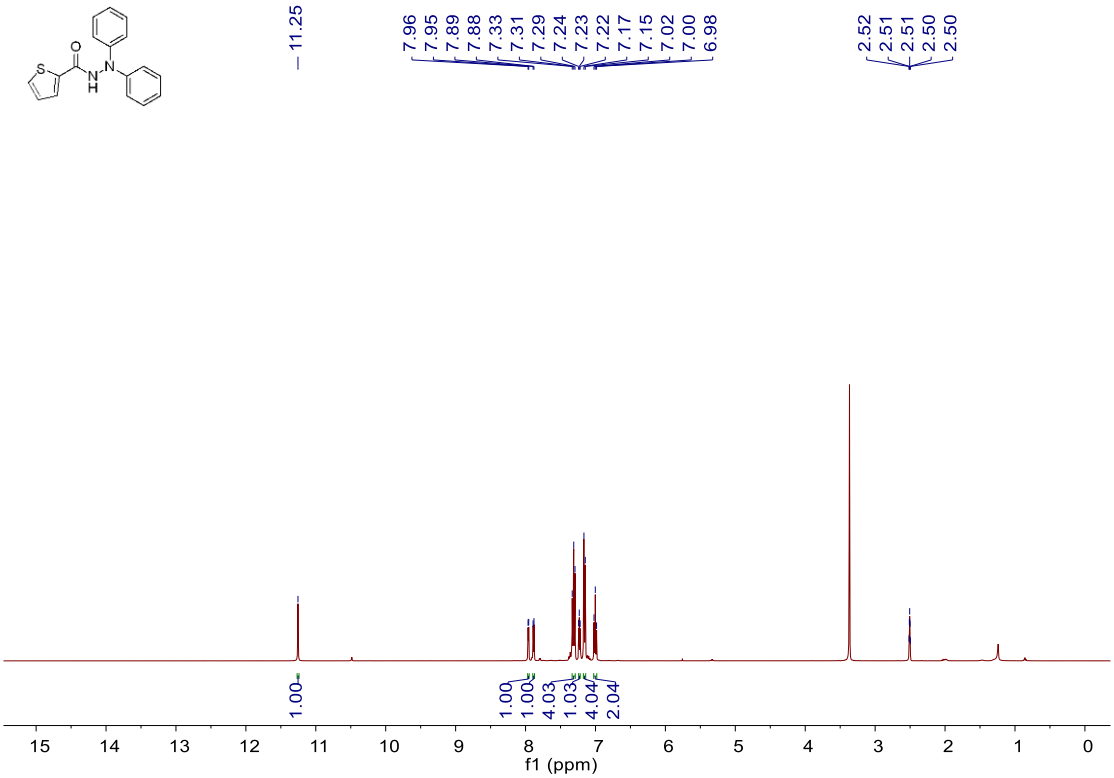
^{13}C{^{1}H} NMR of 24 in DMSO-d_6 (100 MHz)



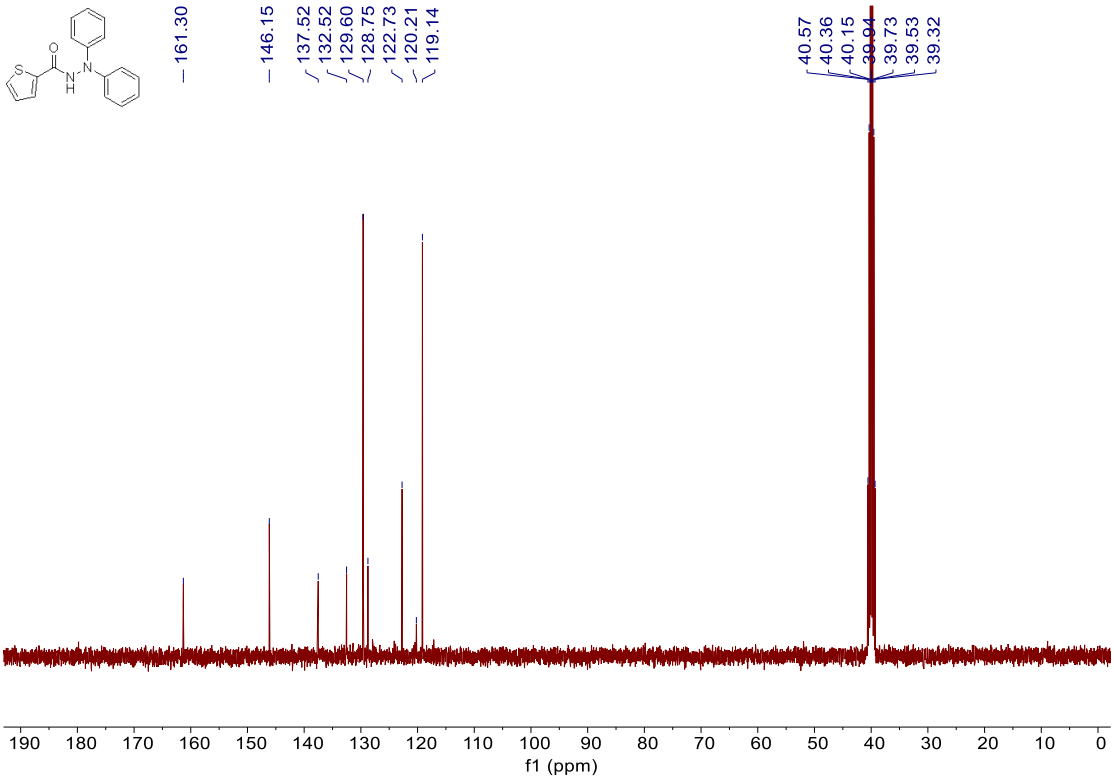
^{1}H NMR of 25 in DMSO-d_6 (400 MHz)



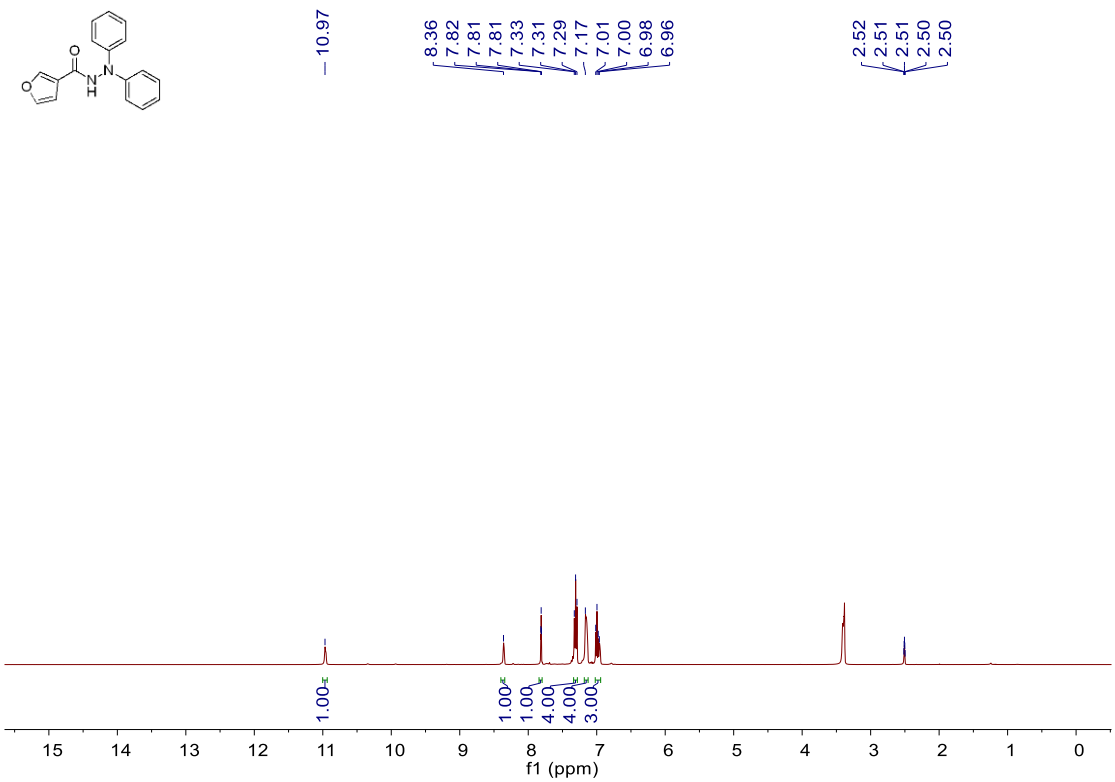
^{13}C{^{1}H} NMR of 25 in DMSO-d_6 (100 MHz)



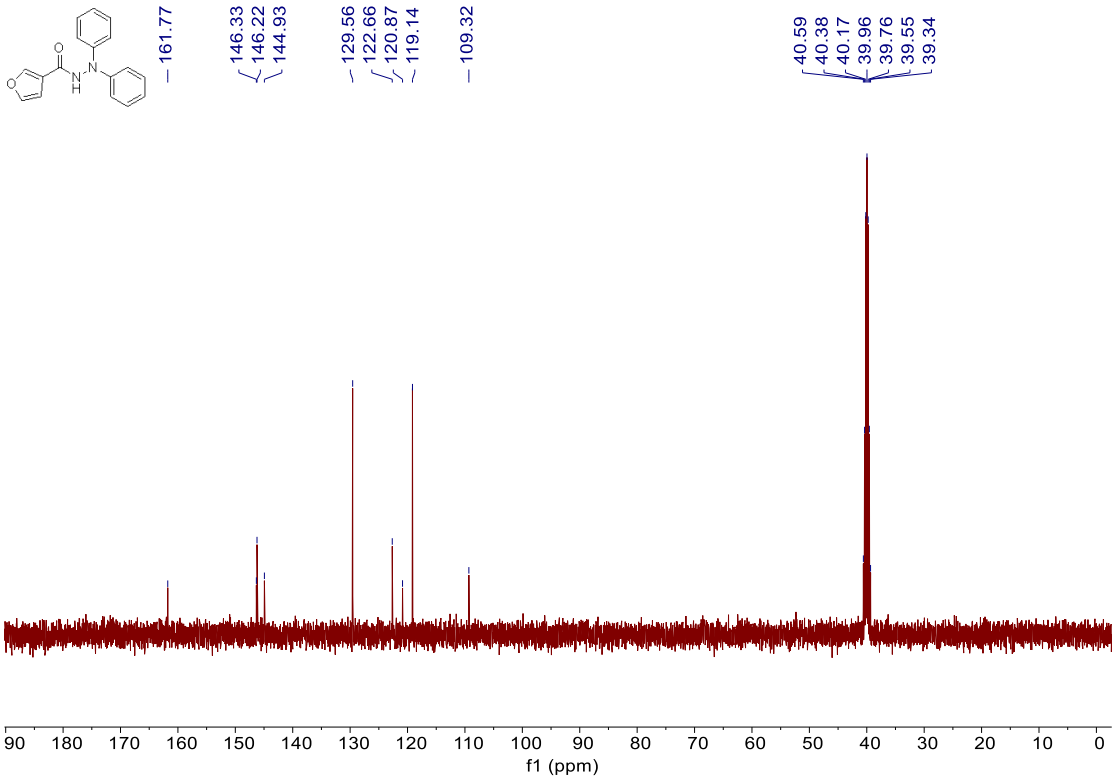
^{1}H NMR of 26 in DMSO-d_6 (400 MHz)



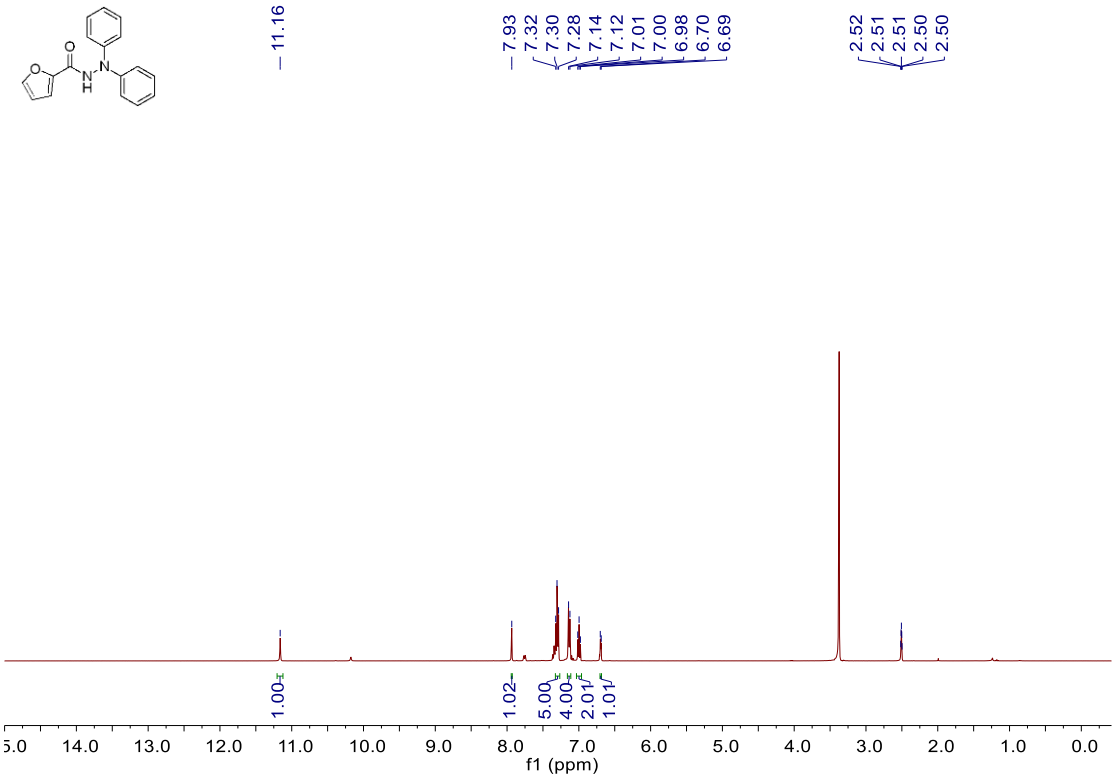
^{13}C{^{1}H} NMR of 26 in DMSO-d_6 (100 MHz)



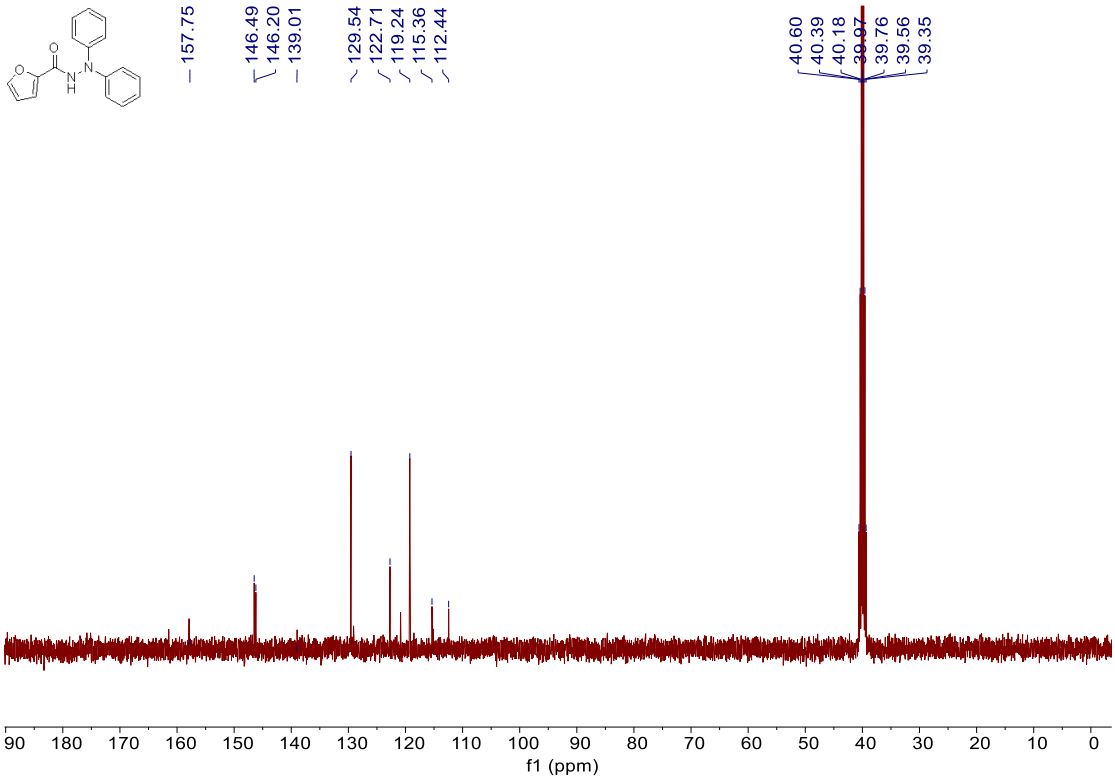
^{1}H NMR of 27 in DMSO-d_6 (400 MHz)



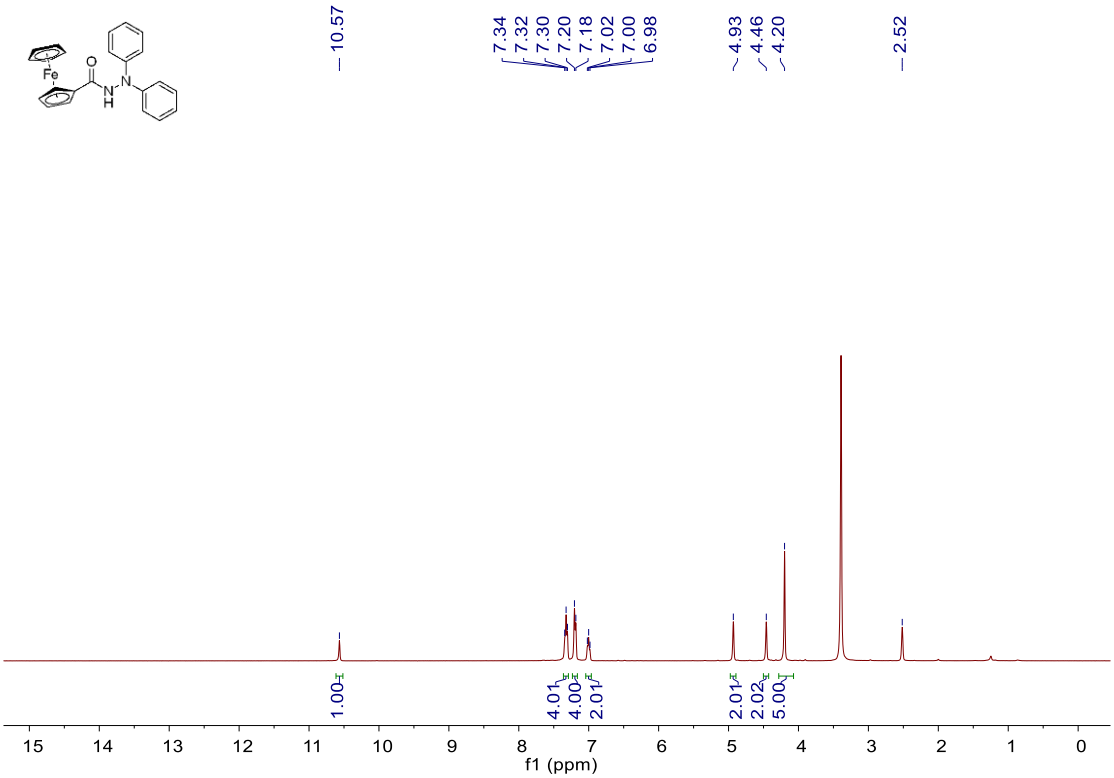
^{13}C{^{1}H} NMR of 27 in DMSO-d_6 (100 MHz)



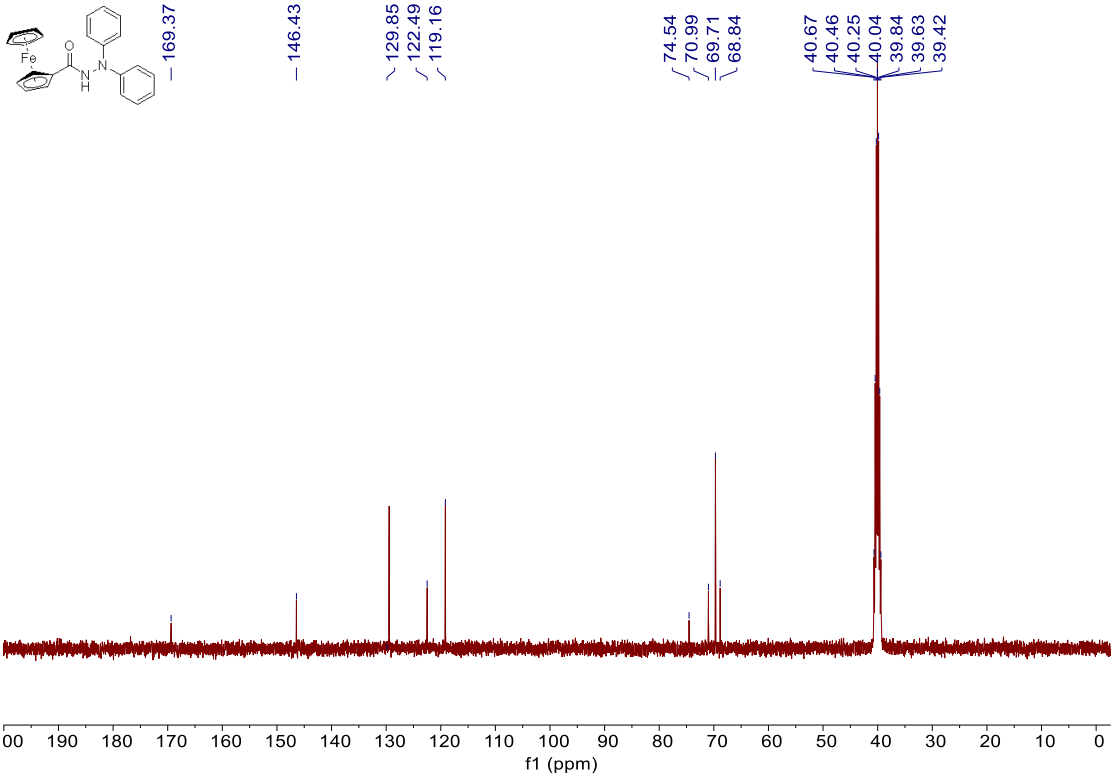
^{1}H NMR of 28 in DMSO-d_6 (400 MHz)



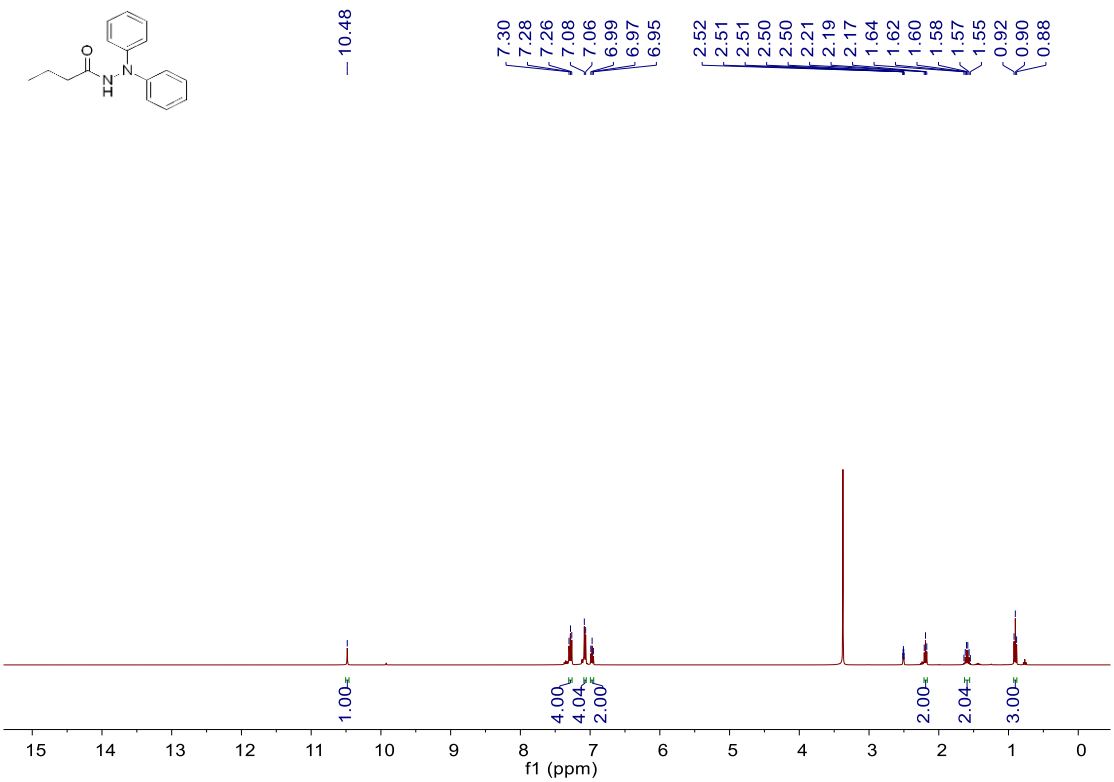
^{13}C{^{1}H} NMR of 28 in DMSO-d_6 (100 MHz)



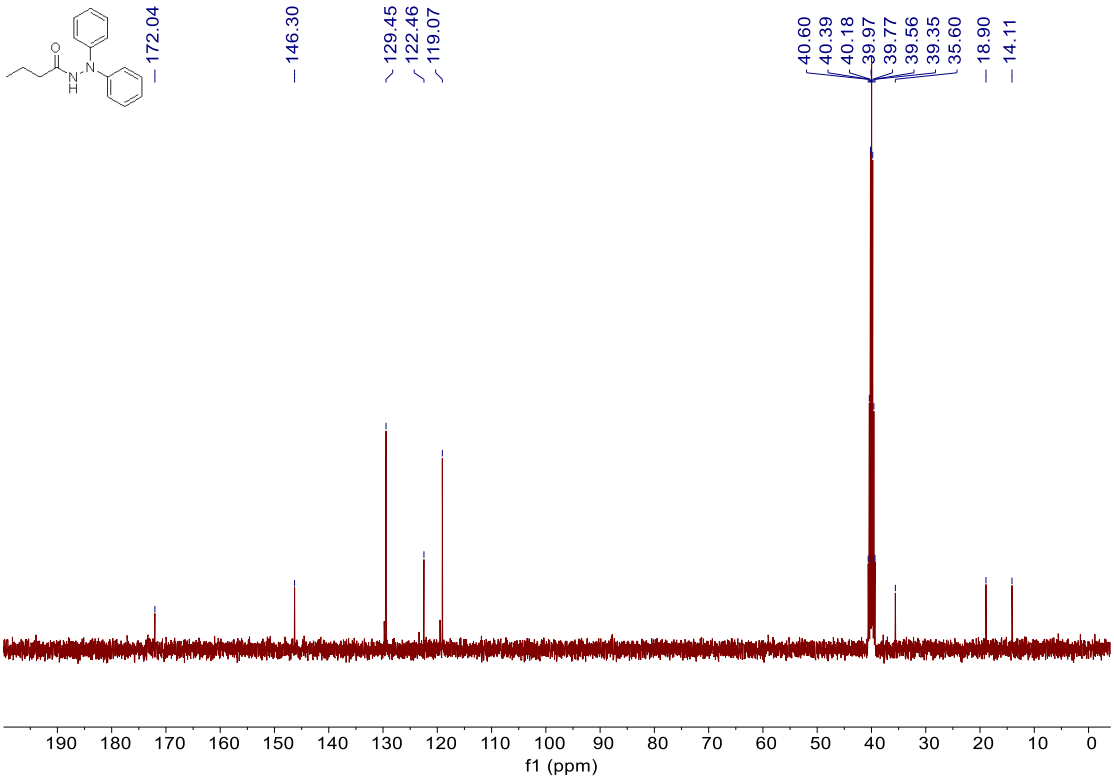
^{1}H NMR of 29 in DMSO-d_6 (400 MHz)



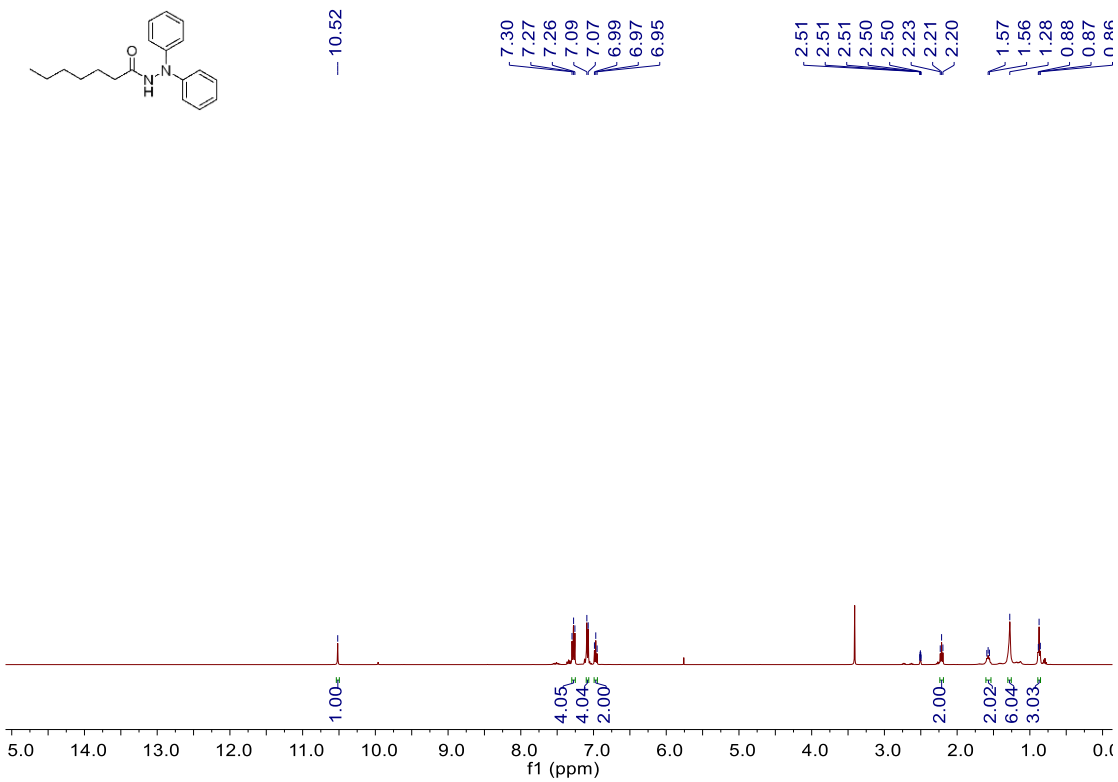
^{13}C{^{1}H} NMR of 29 in DMSO-d_6 (100 MHz)



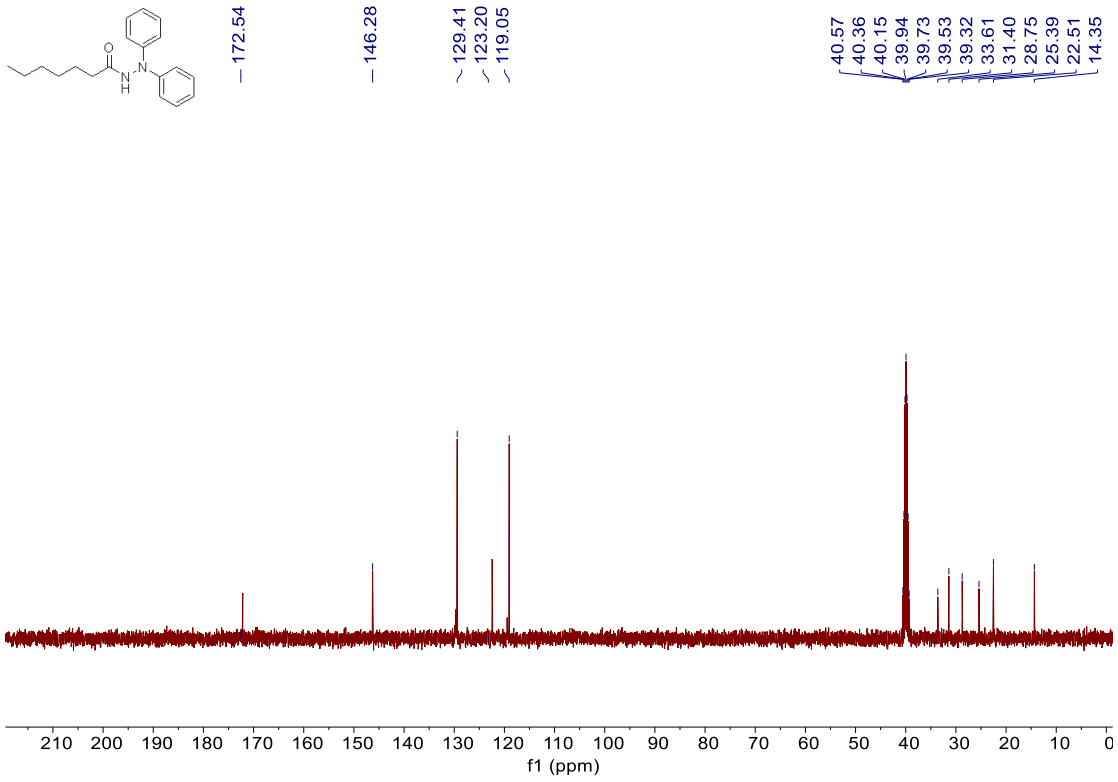
^{1}H NMR of 30 in DMSO-d_6 (400 MHz)



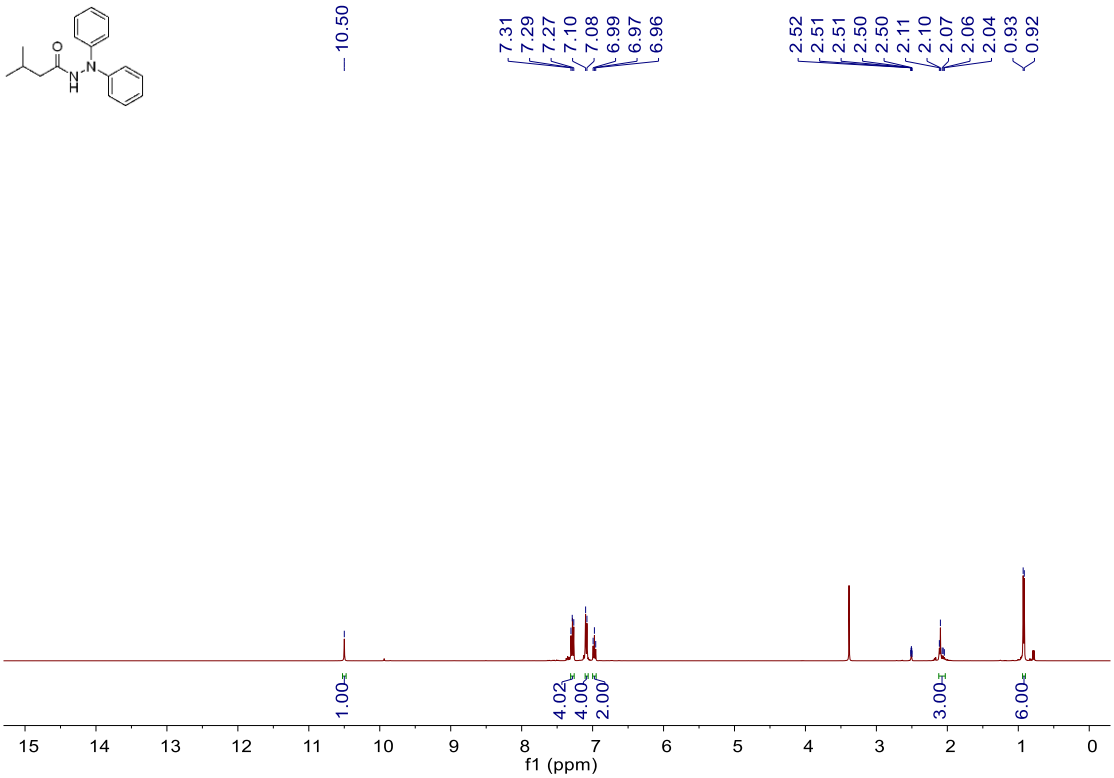
^{13}C{^{1}H} NMR of 30 in DMSO-d_6 (100 MHz)



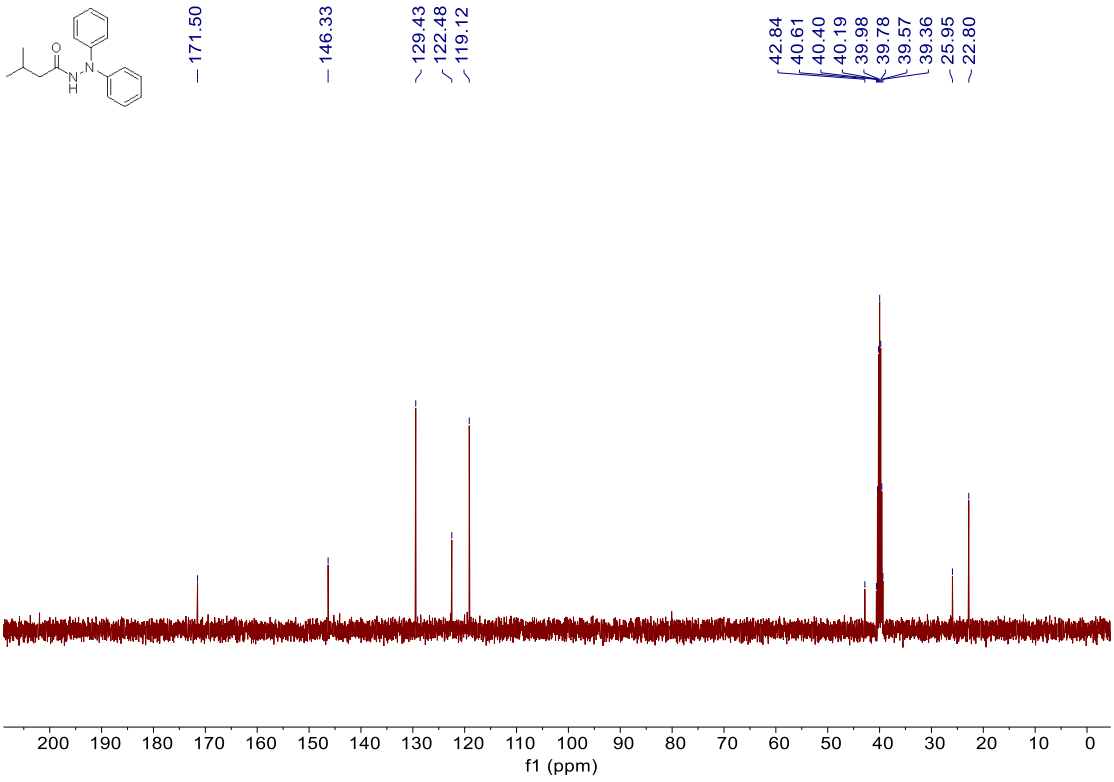
^{1}H NMR of 31 in DMSO-d_6 (400 MHz)



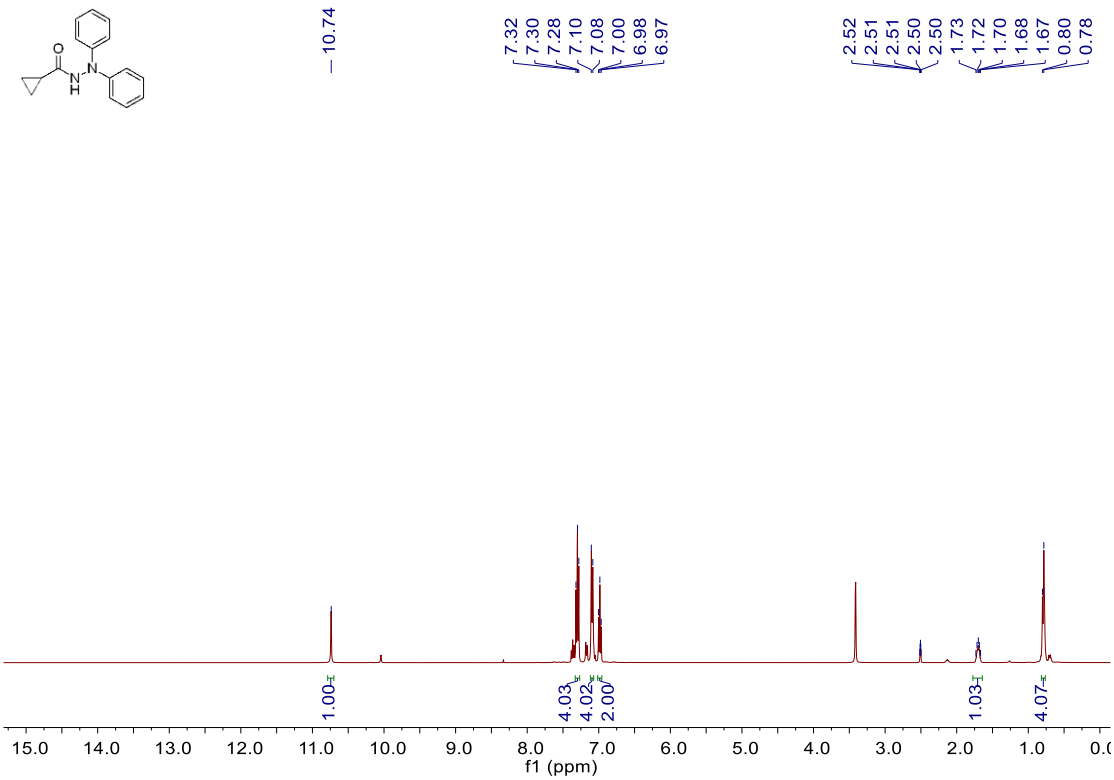
^{13}C{^{1}H} NMR of 31 in DMSO-d_6 (100 MHz)



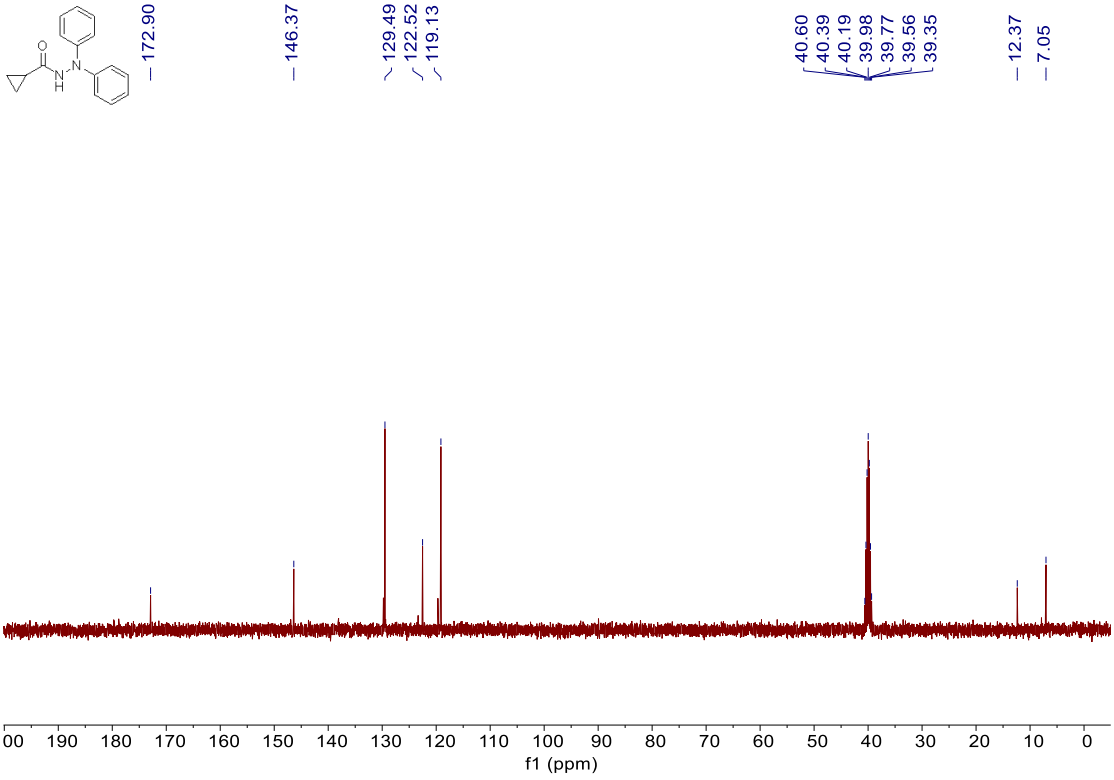
^{1}H NMR of 32 in DMSO-d_6 (400 MHz)



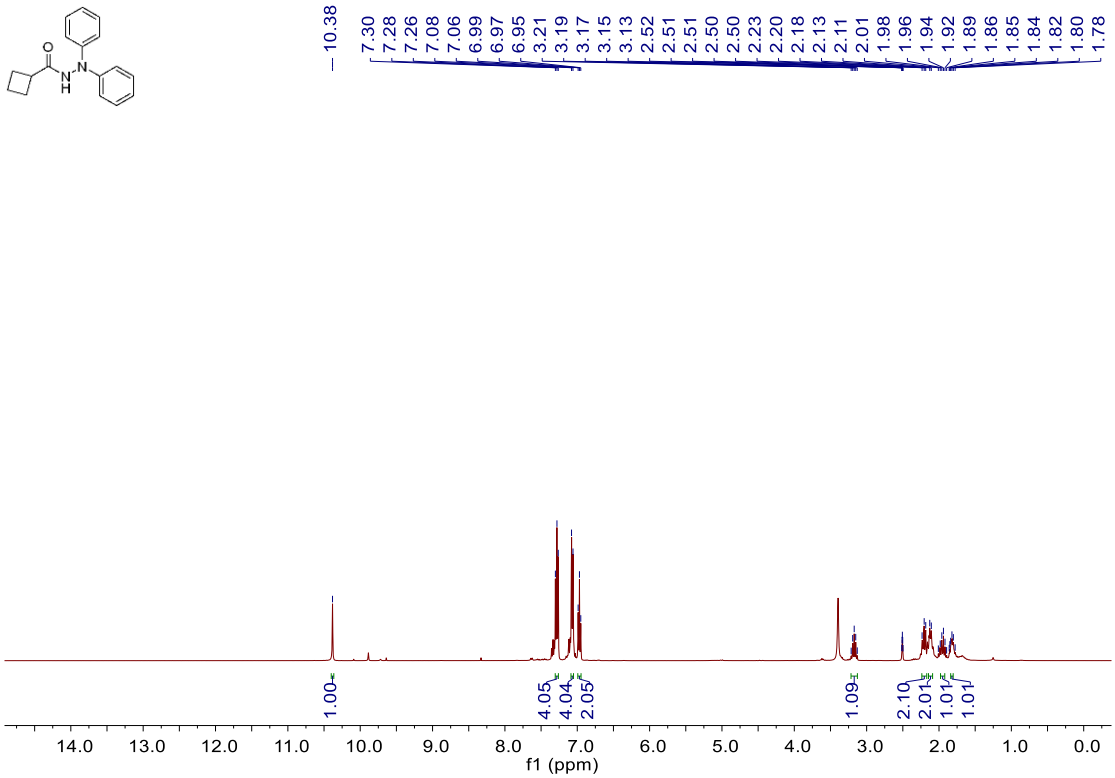
^{13}C{^{1}H} NMR of 32 in DMSO-d_6 (100 MHz)



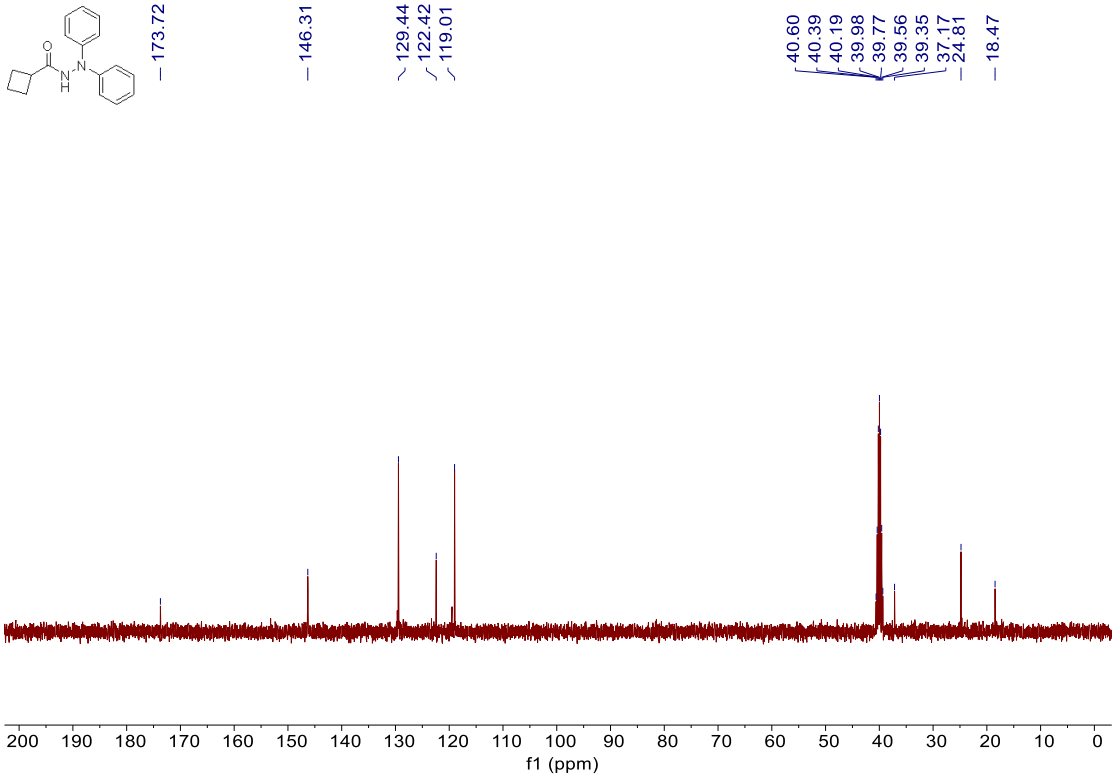
^{1}H NMR of 33 in DMSO-d_6 (400 MHz)



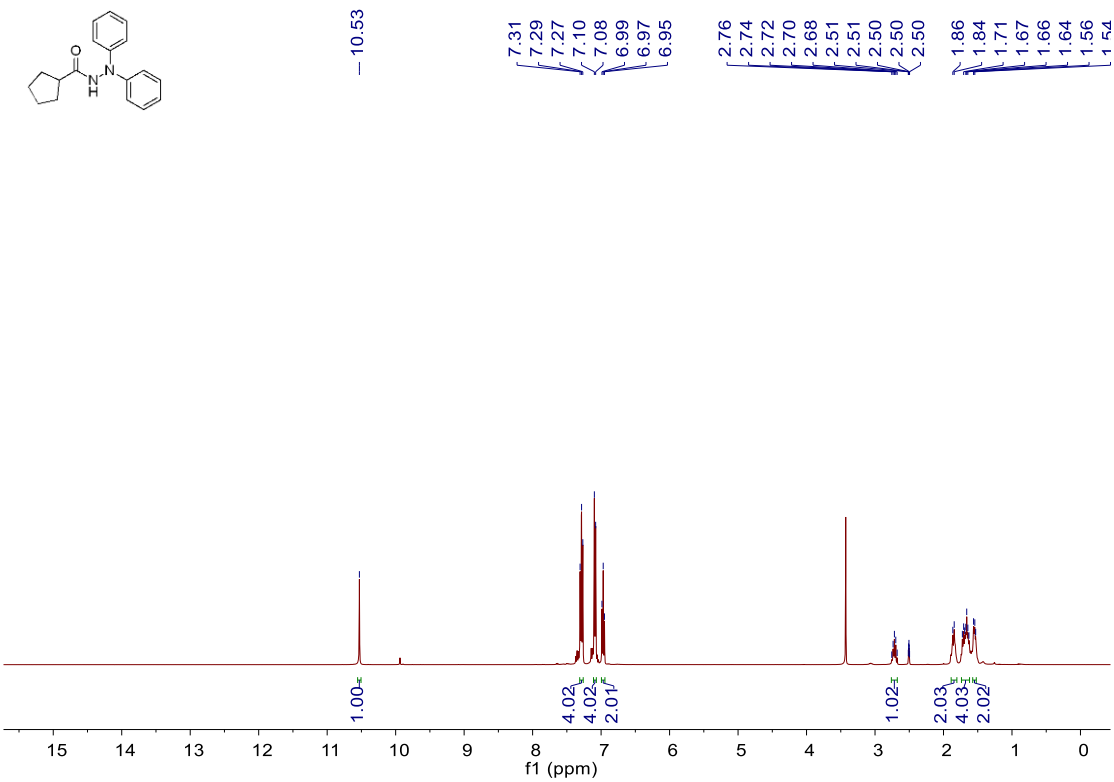
^{13}C{^{1}H} NMR of 33 in DMSO-d_6 (100 MHz)



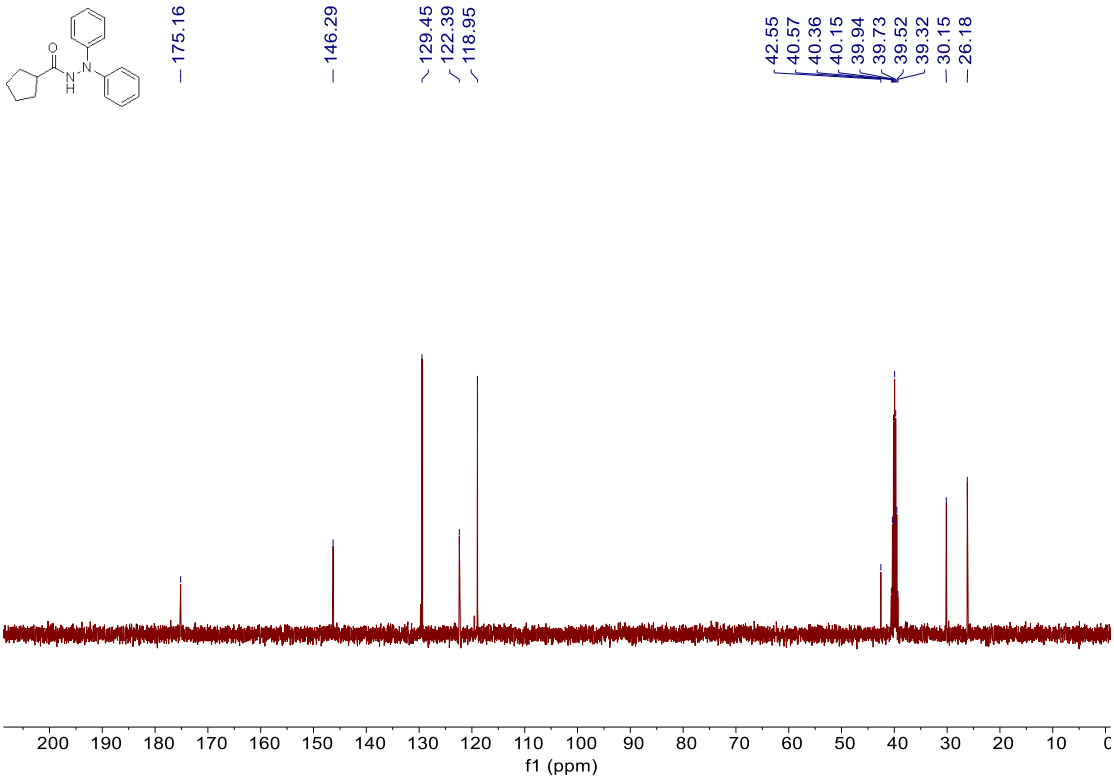
^{1}H NMR of 34 in DMSO-d_6 (400 MHz)



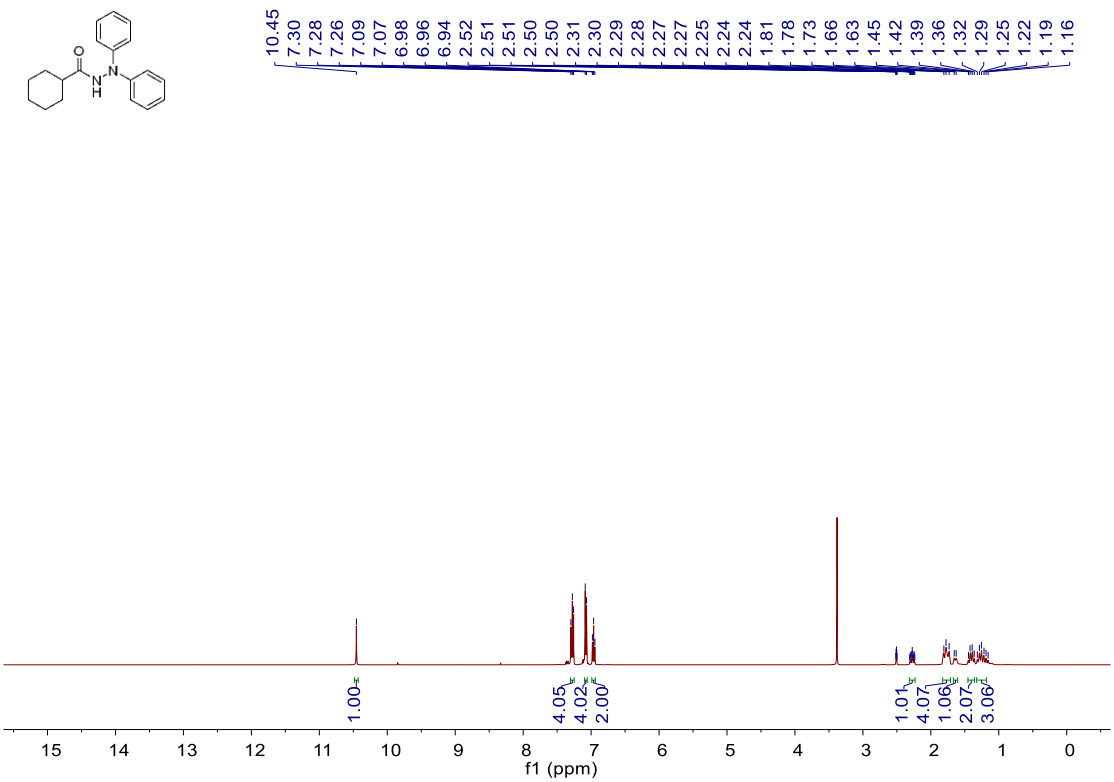
^{13}C{^{1}H} NMR of 34 in DMSO-d_6 (100 MHz)



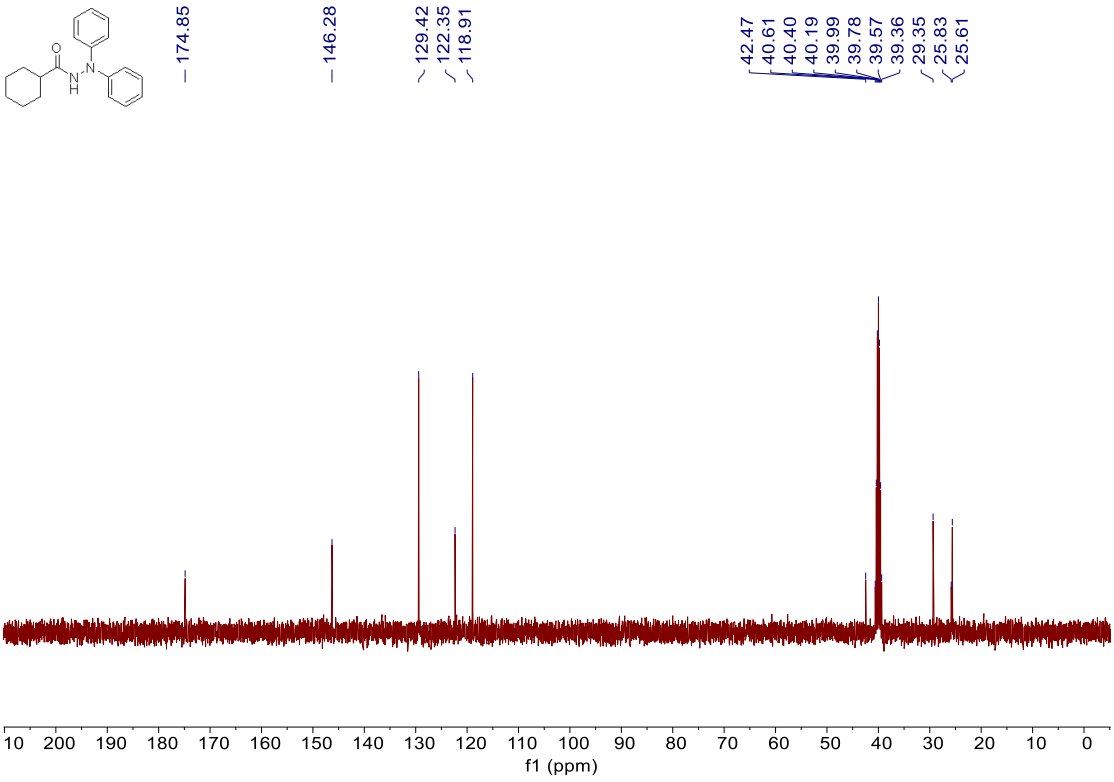
^{1}H NMR of 35 in DMSO-d_6 (400 MHz)



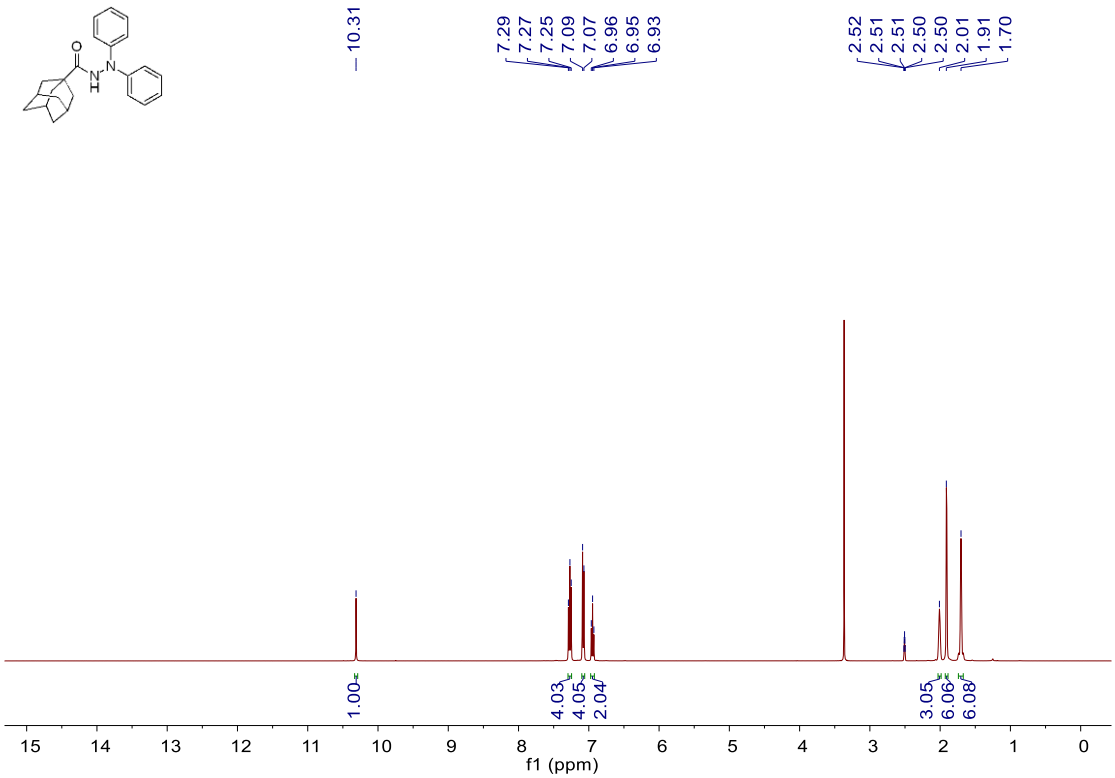
^{13}C{^{1}H} NMR of 35 in DMSO-d_6 (100 MHz)



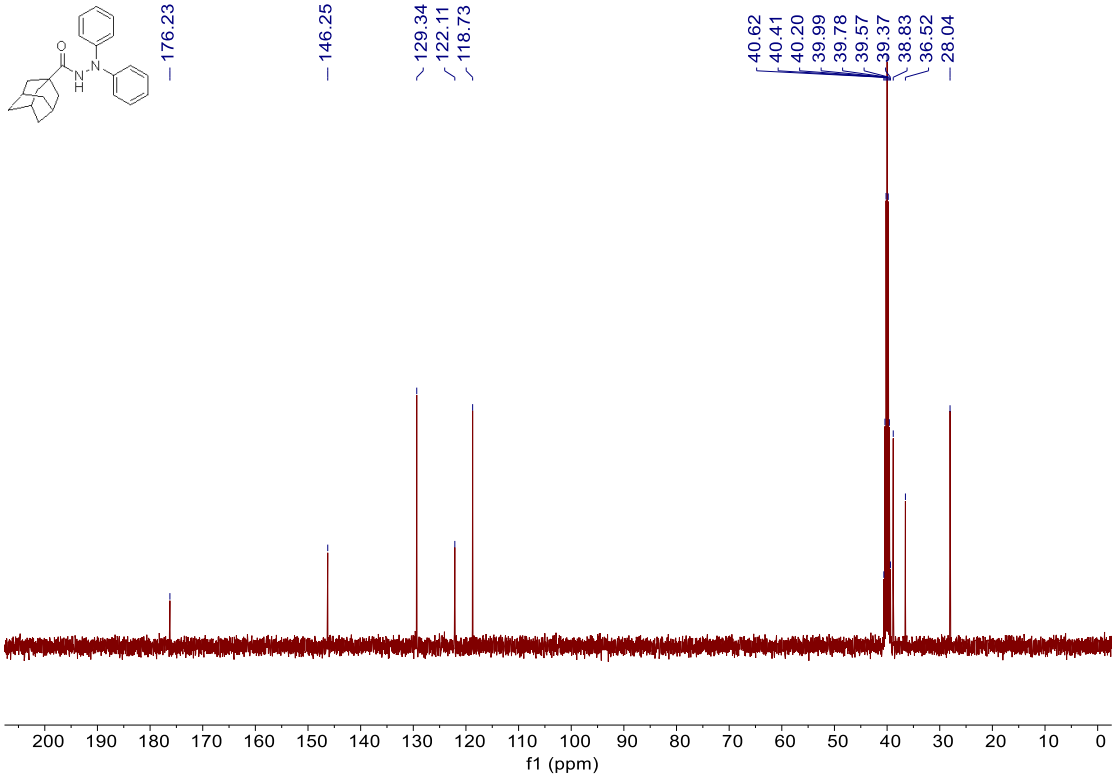
^{1}H NMR of 36 in DMSO-d_6 (400 MHz)



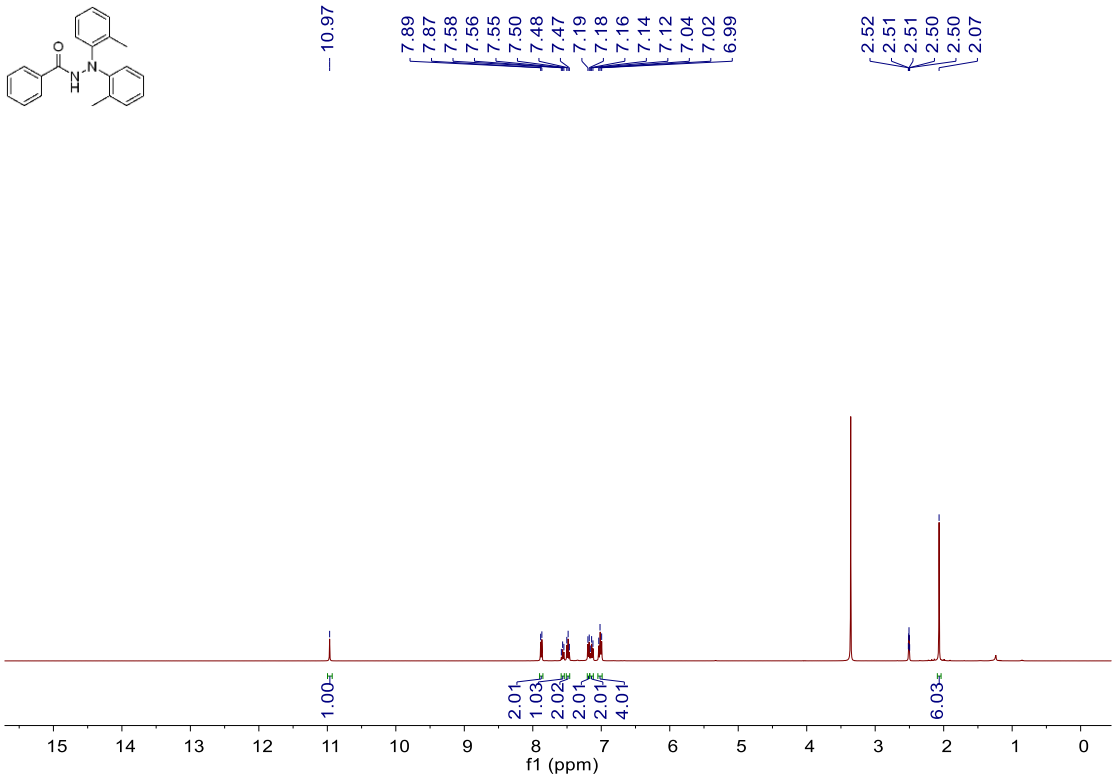
^{13}C{^{1}H} NMR of 36 in DMSO-d_6 (100 MHz)



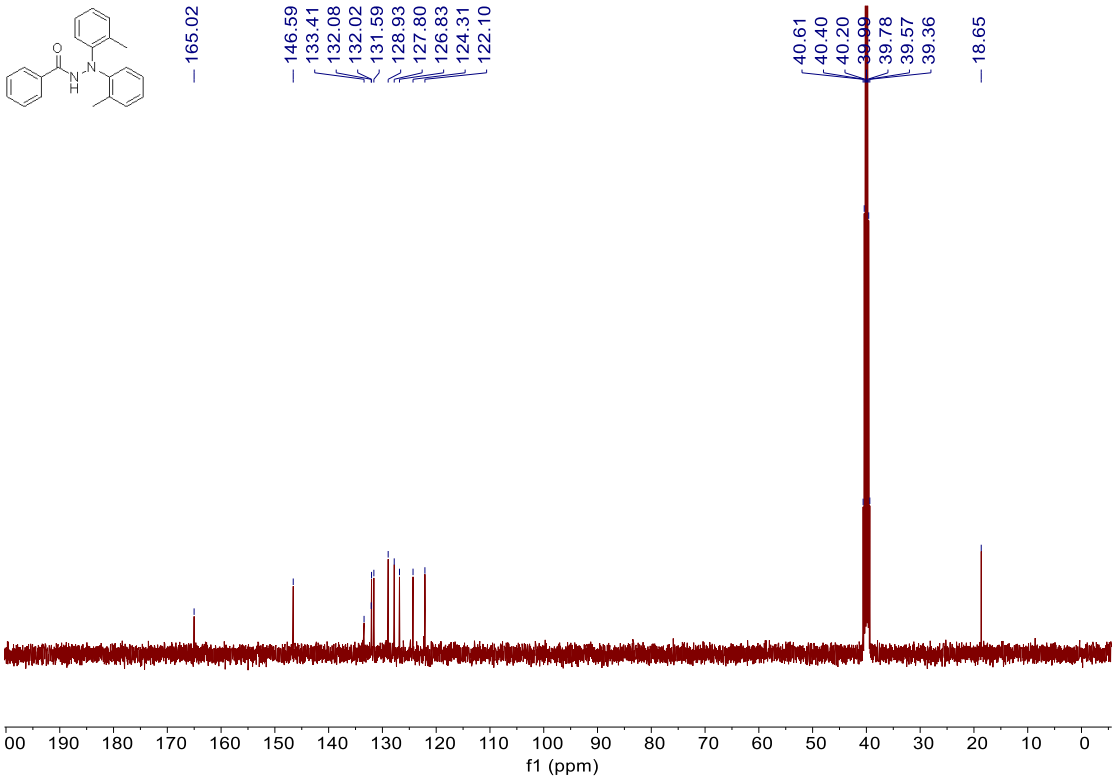
^{1}H NMR of 37 in DMSO-d_6 (400 MHz)



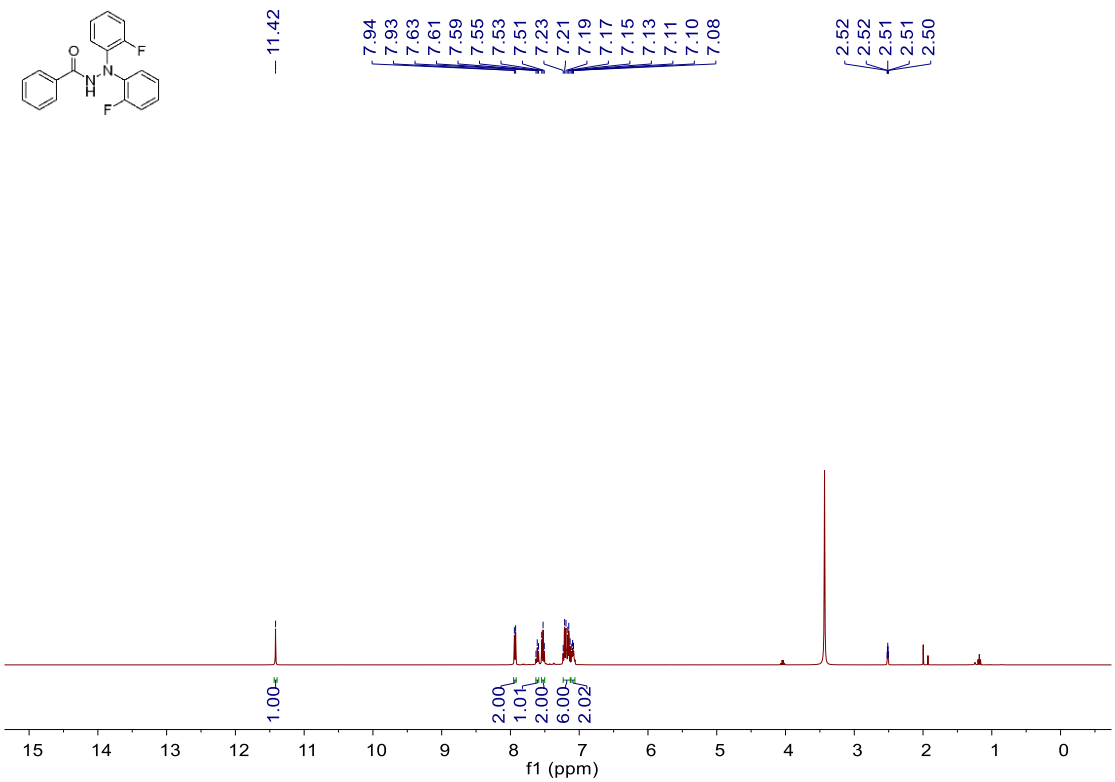
^{13}C{^{1}H} NMR of 37 in DMSO-d_6 (100 MHz)



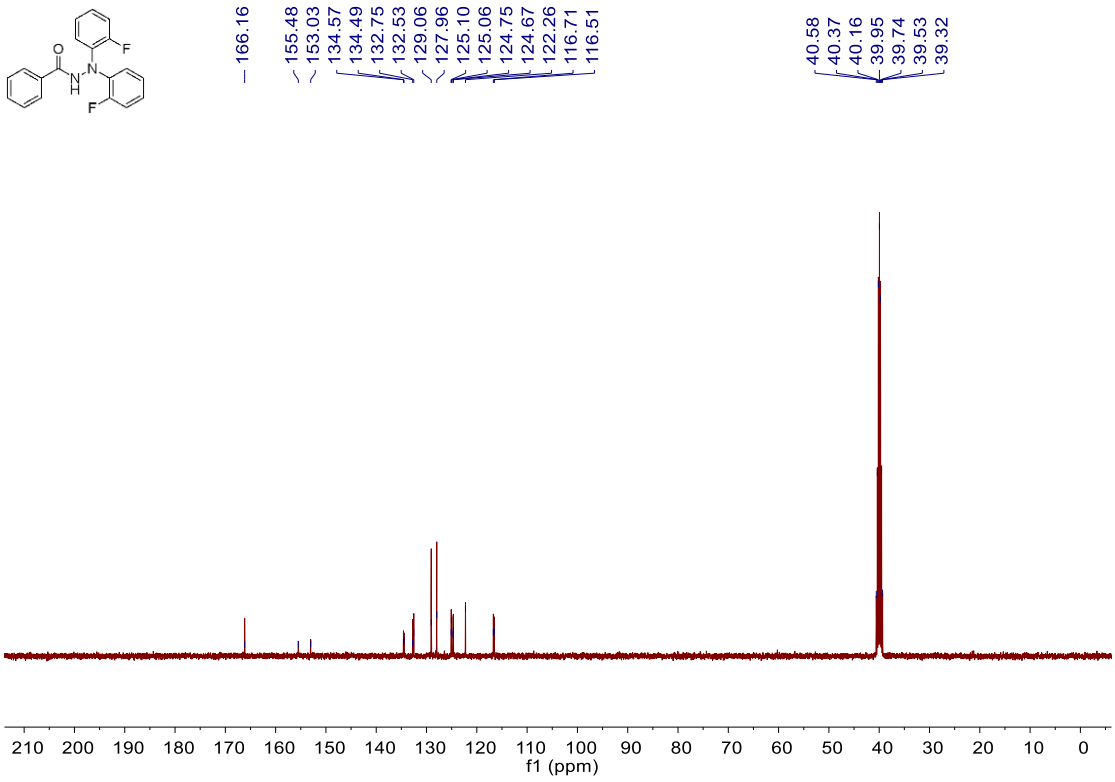
^{1}H NMR of 38 in DMSO-d_6 (400 MHz)



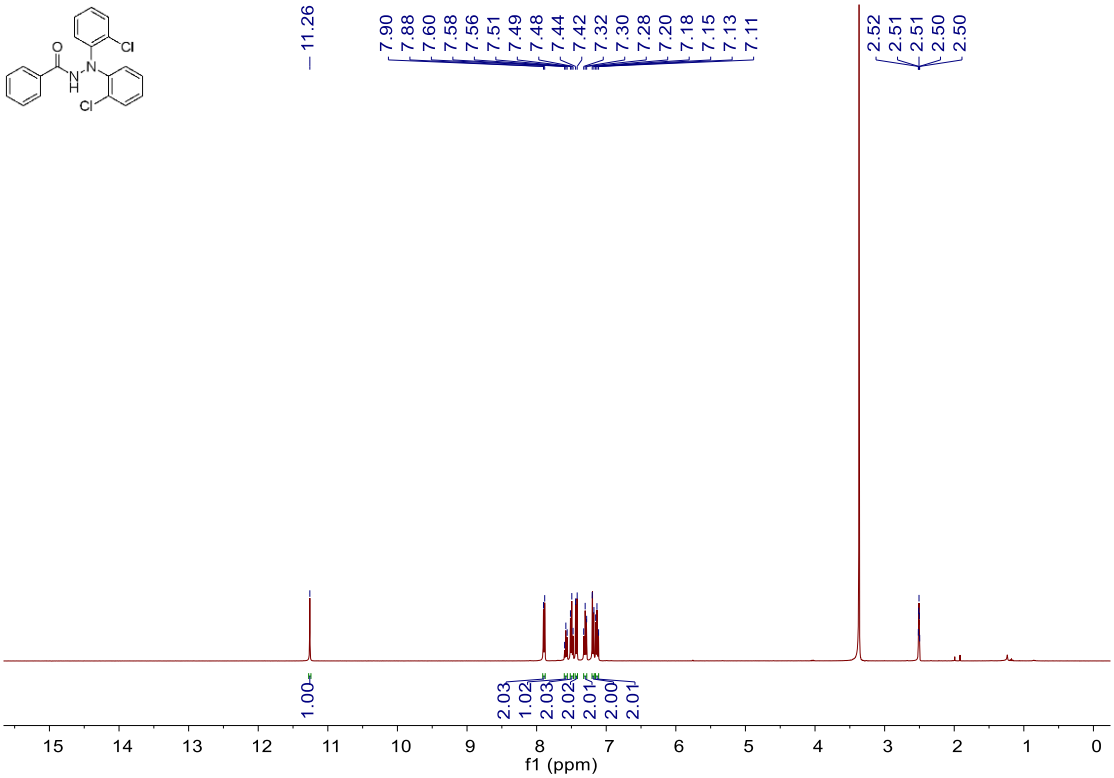
^{13}C{^{1}H} NMR of 38 in DMSO-d_6 (100 MHz)



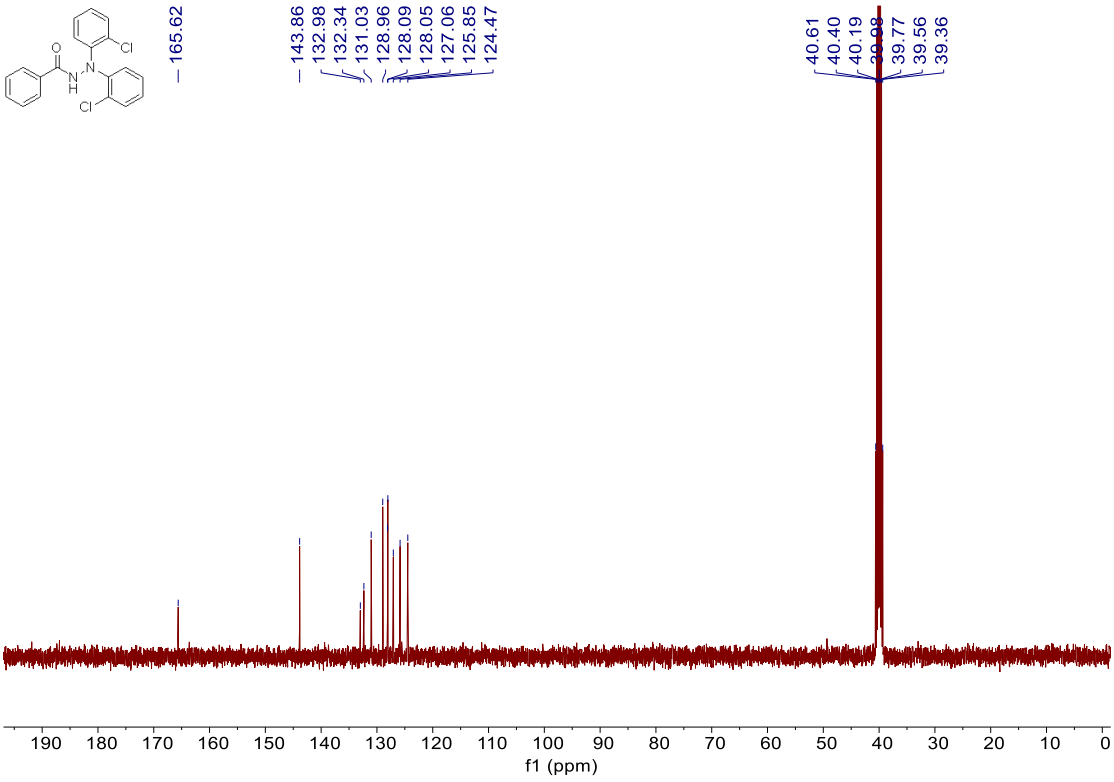
^{1}H NMR of 39 in DMSO-d_6 (400 MHz)



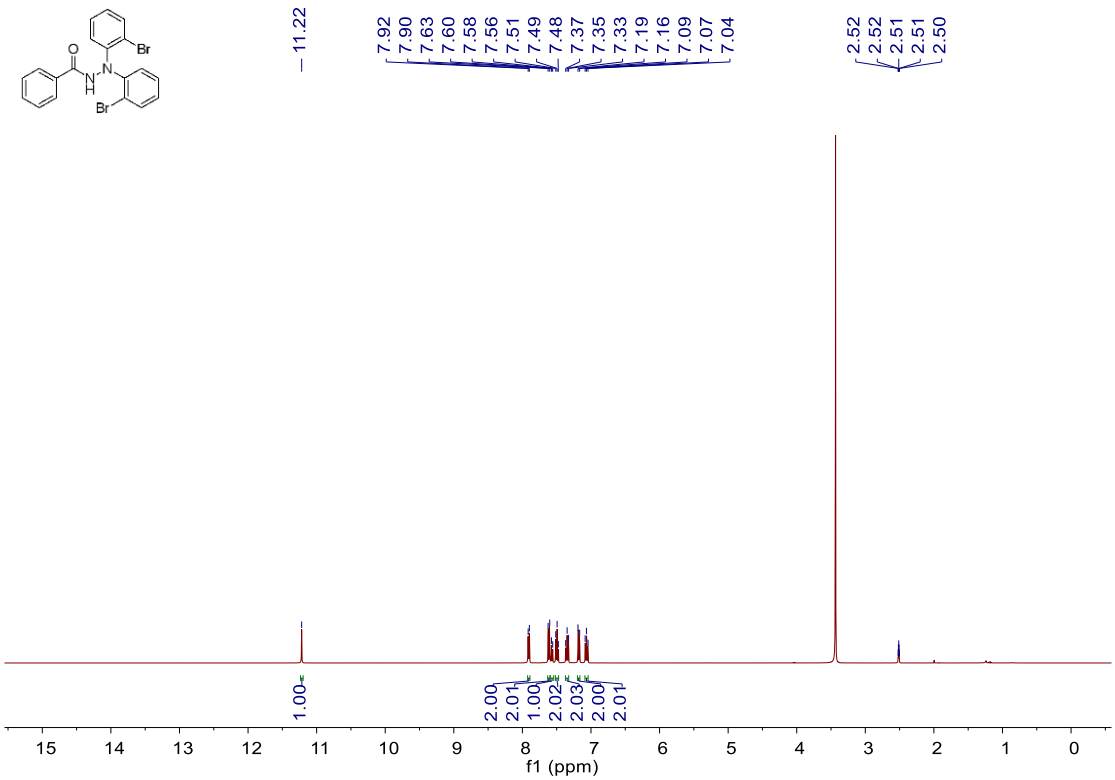
^{13}C{^{1}H} NMR of 39 in DMSO-d_6 (100 MHz)



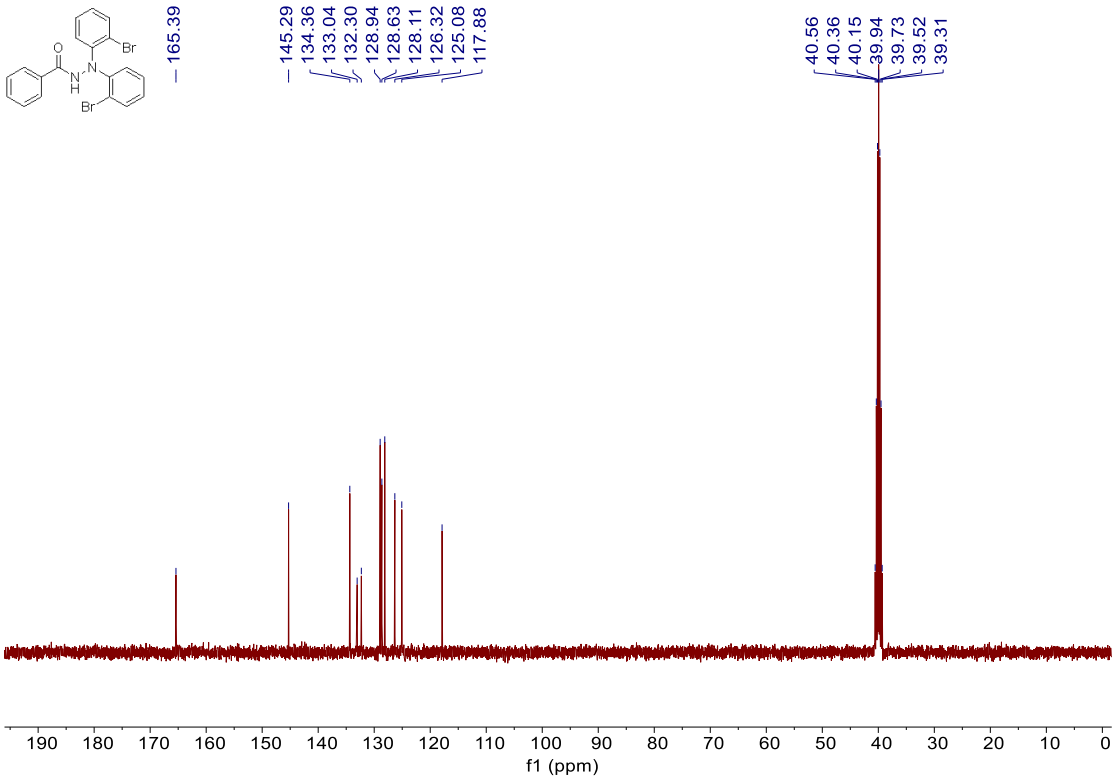
^{1}H NMR of 40 in DMSO-d_6 (400 MHz)



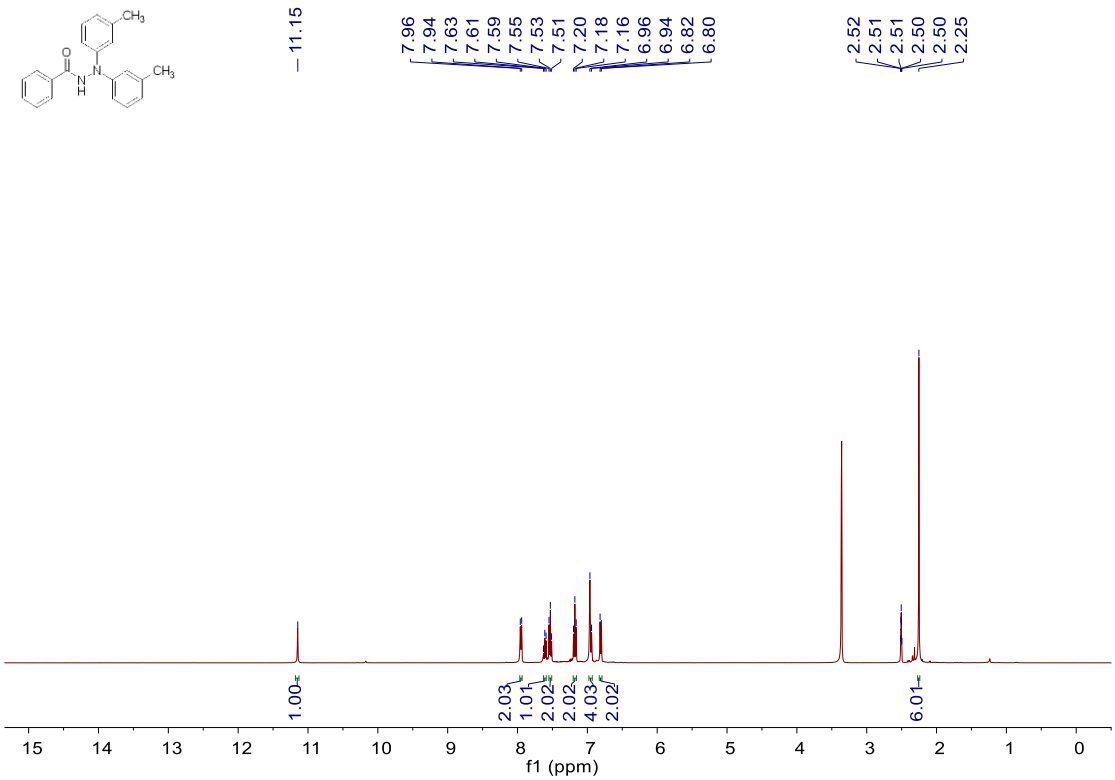
^{13}C{^{1}H} NMR of 40 in DMSO-d_6 (100 MHz)



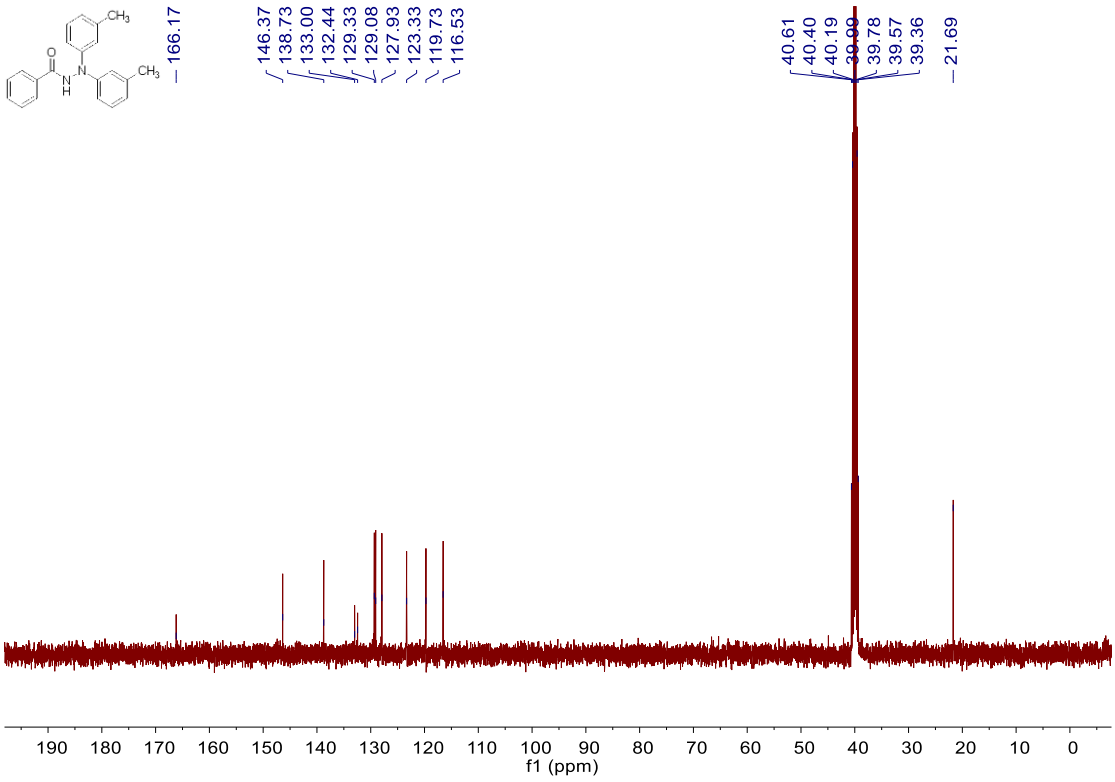
^{1}H NMR of 41 in DMSO-d_6 (400 MHz)



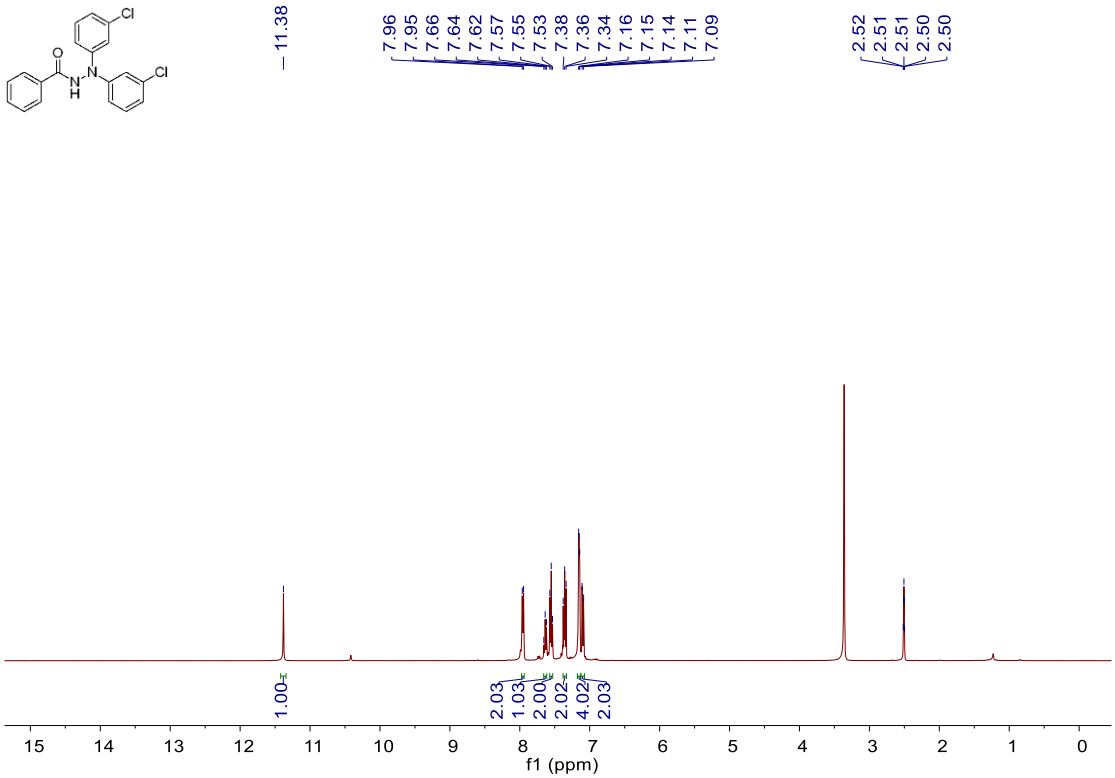
^{13}C{^{1}H} NMR of 41 in DMSO-d_6 (100 MHz)



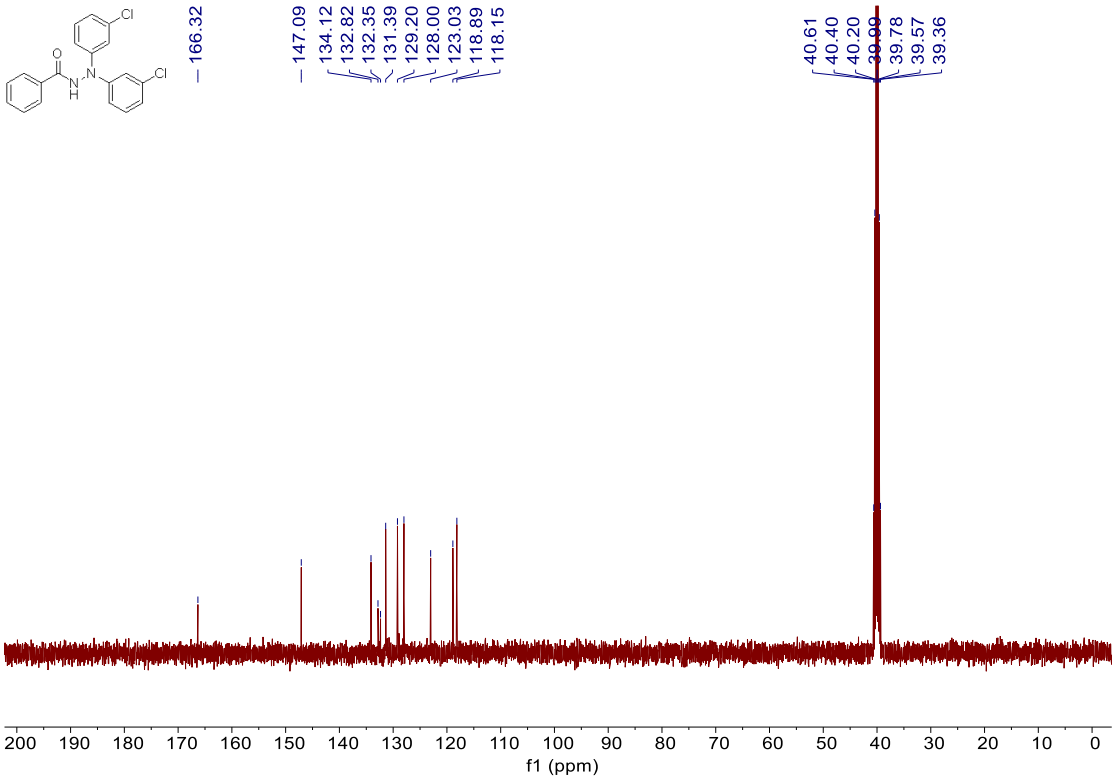
^{1}H NMR of 42 in DMSO-d_6 (400 MHz)



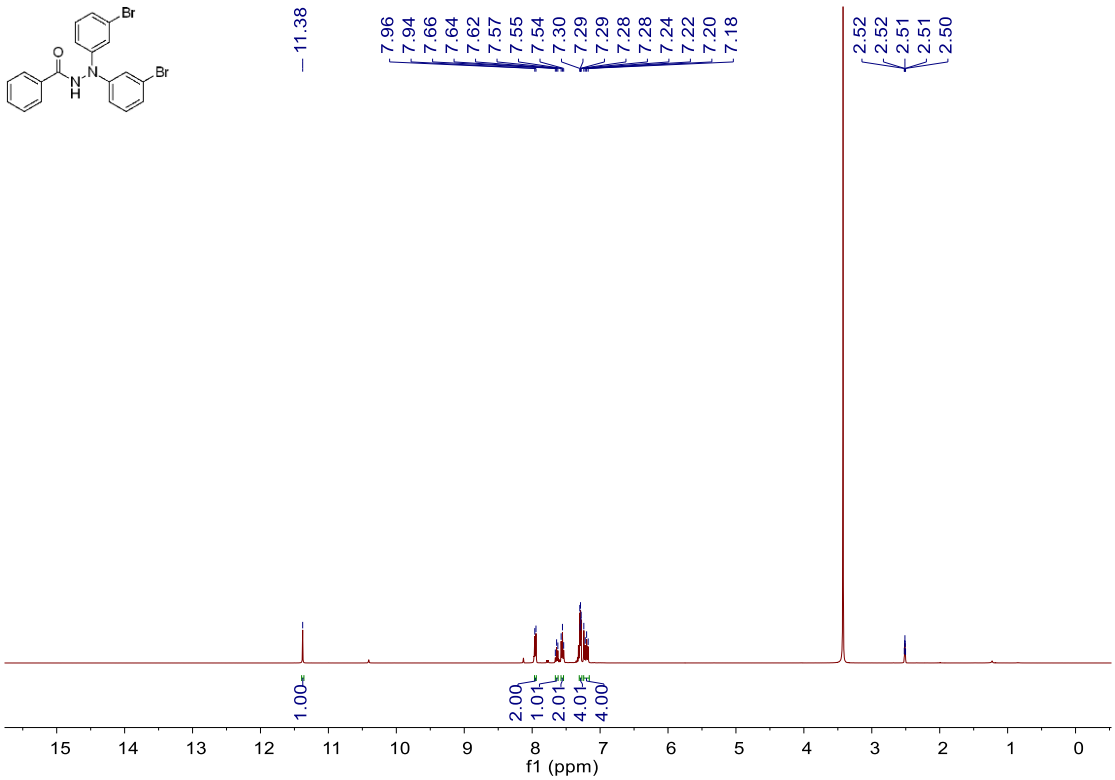
^{13}C{^{1}H} NMR of 42 in DMSO-d_6 (100 MHz)



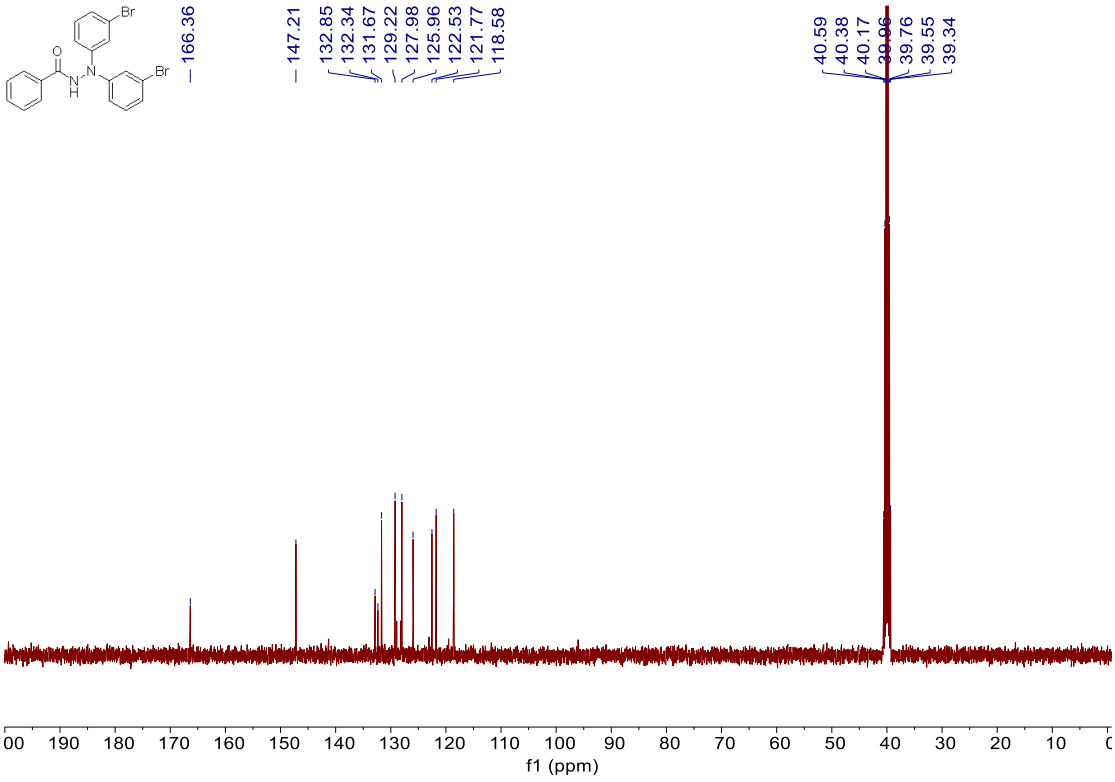
^{1}H NMR of 43 in DMSO-d_6 (400 MHz)



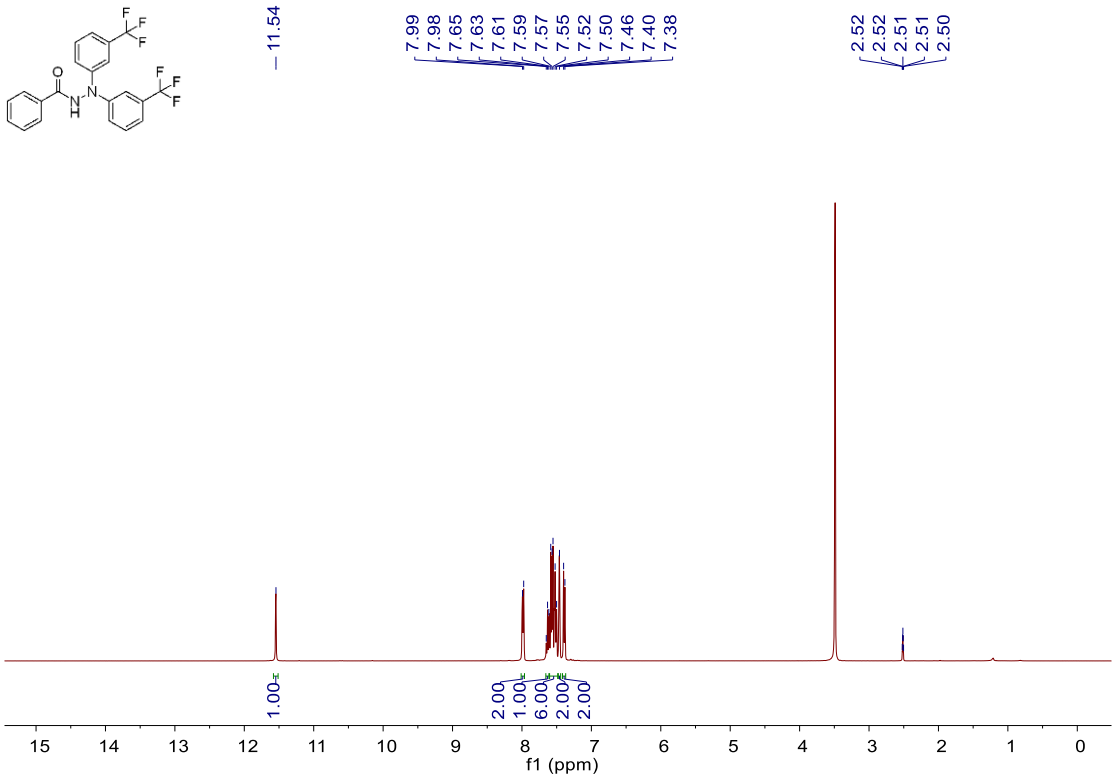
^{13}C{^{1}H} NMR of 43 in DMSO-d_6 (100 MHz)



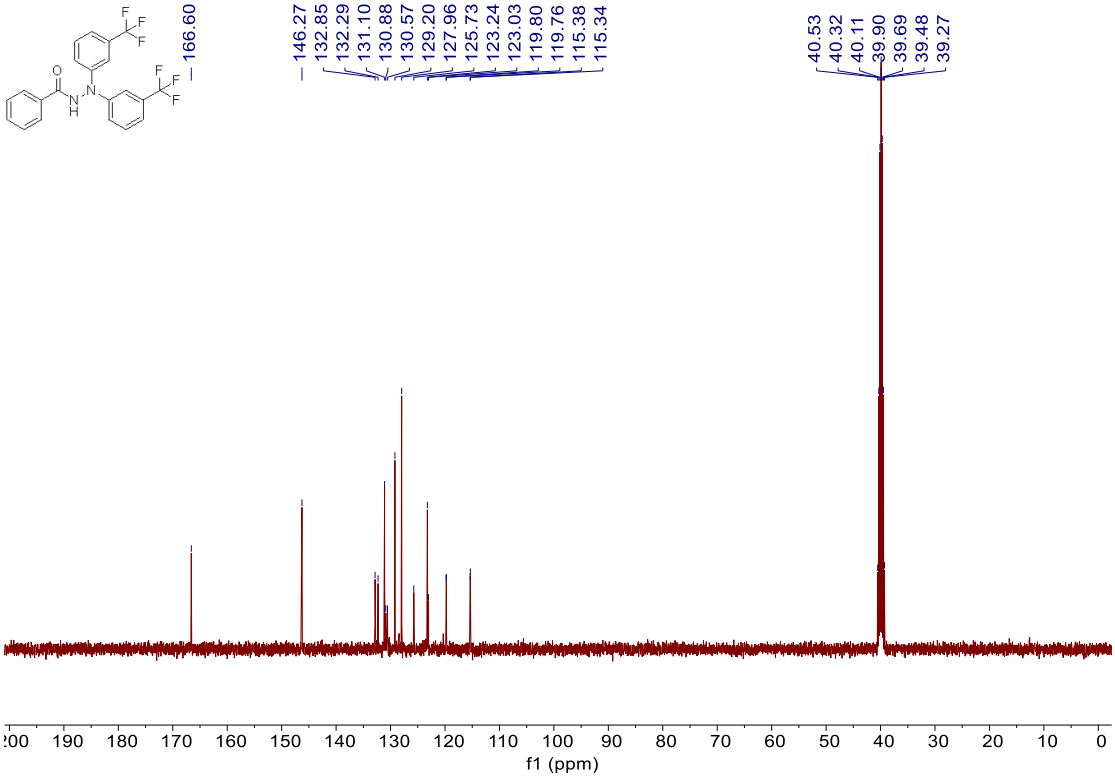
^{1}H NMR of 44 in DMSO-d_6 (400 MHz)



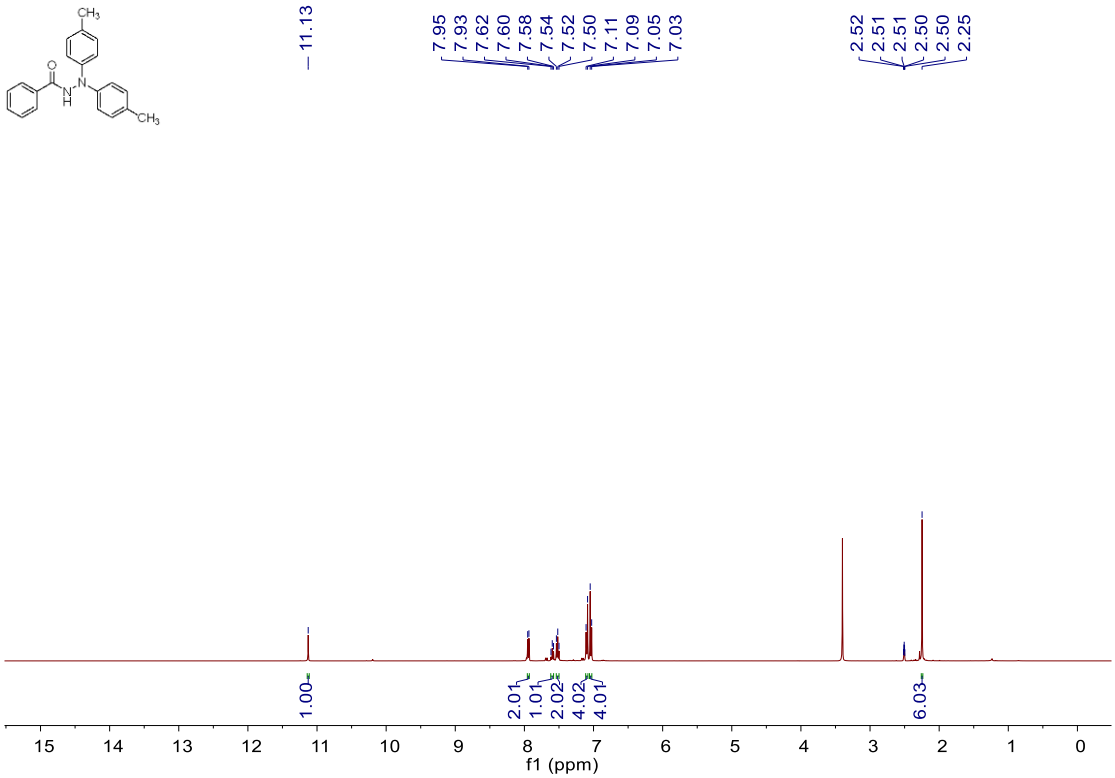
^{13}C{^{1}H} NMR of 44 in DMSO-d_6 (100 MHz)



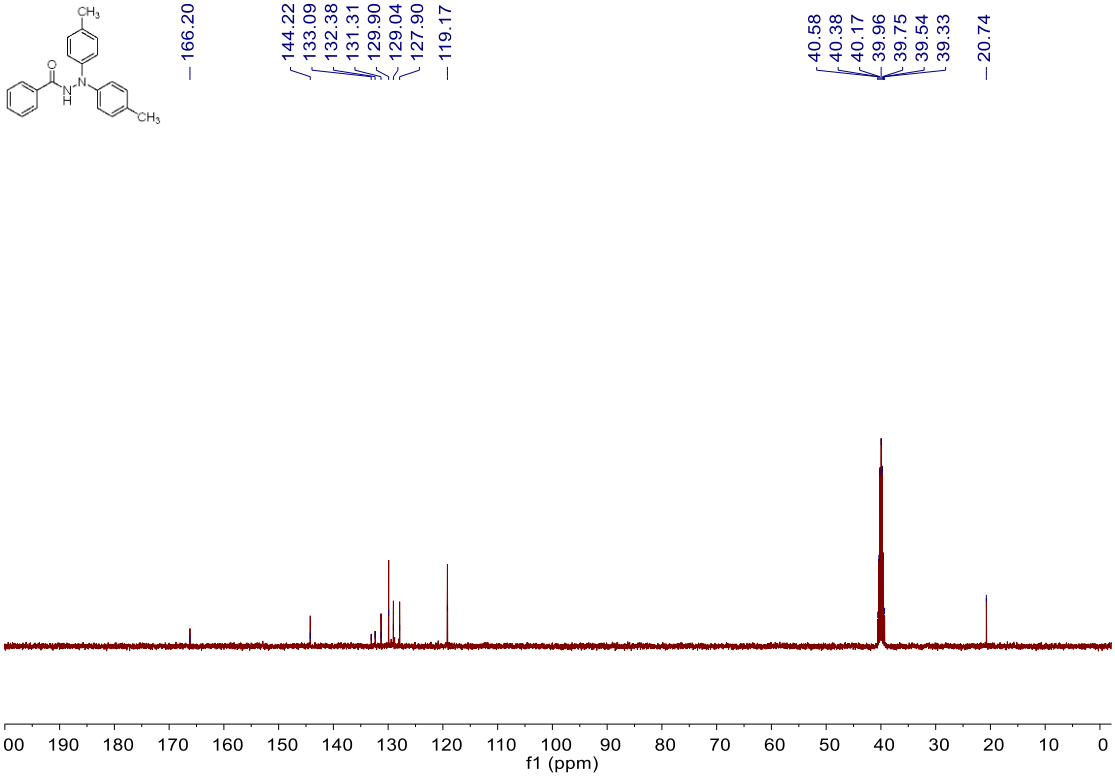
^{1}H NMR of 45 in DMSO-d_6 (400 MHz)



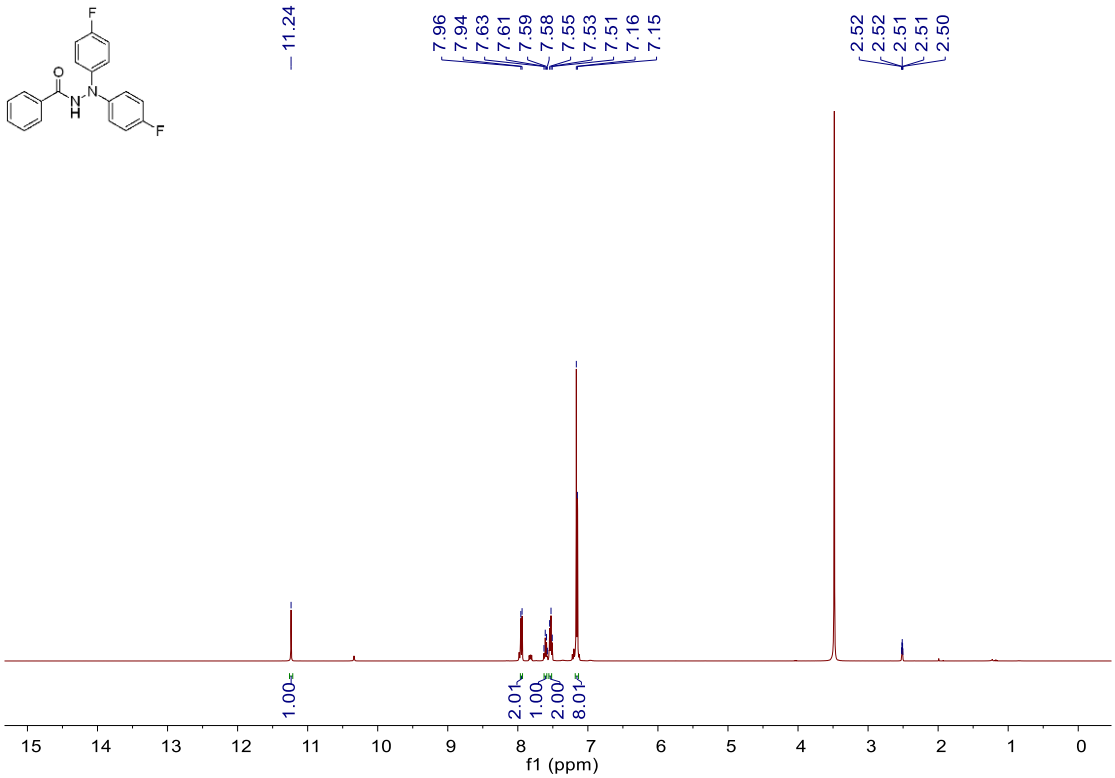
^{13}C{^{1}H} NMR of 45 in DMSO-d_6 (100 MHz)



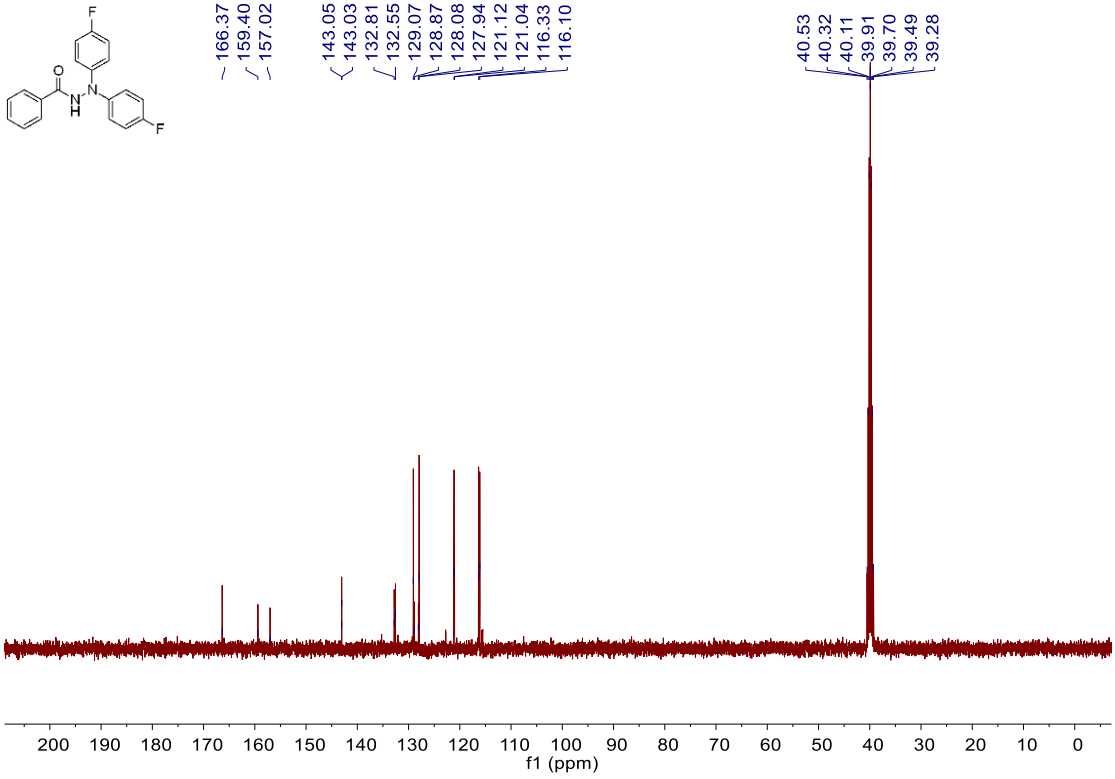
^{1}H NMR of 46 in DMSO-d_6 (400 MHz)



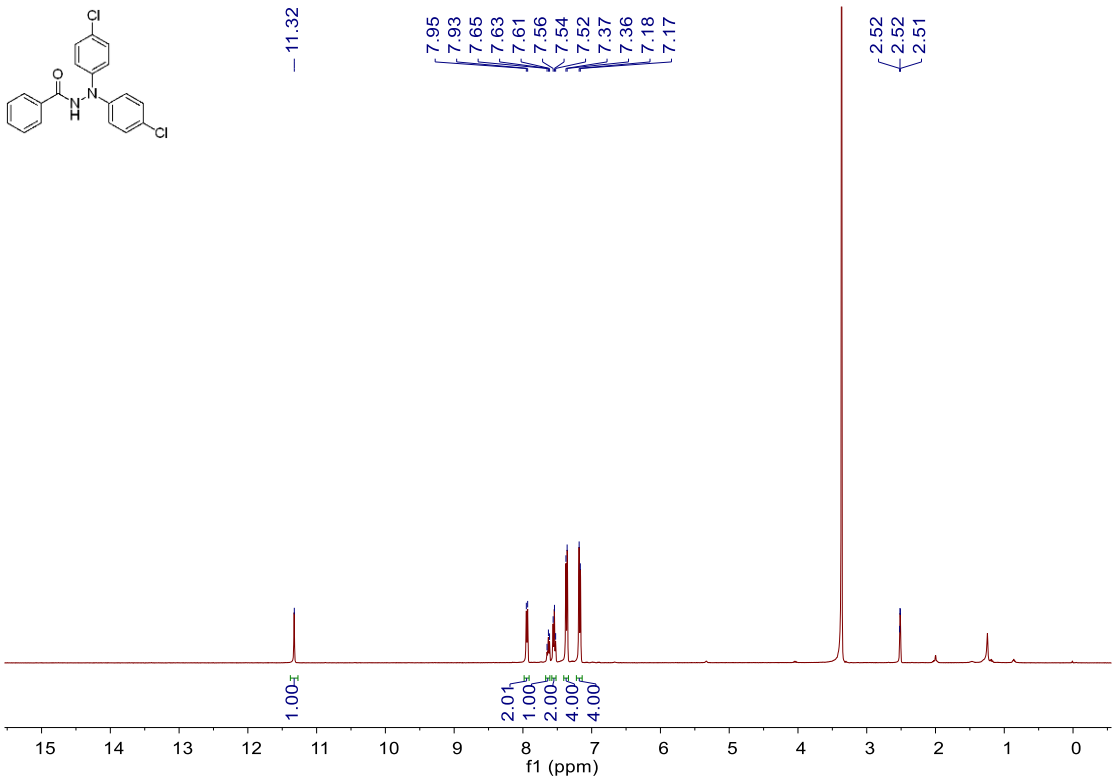
^{13}C{^{1}H} NMR of 46 in DMSO-d_6 (100 MHz)



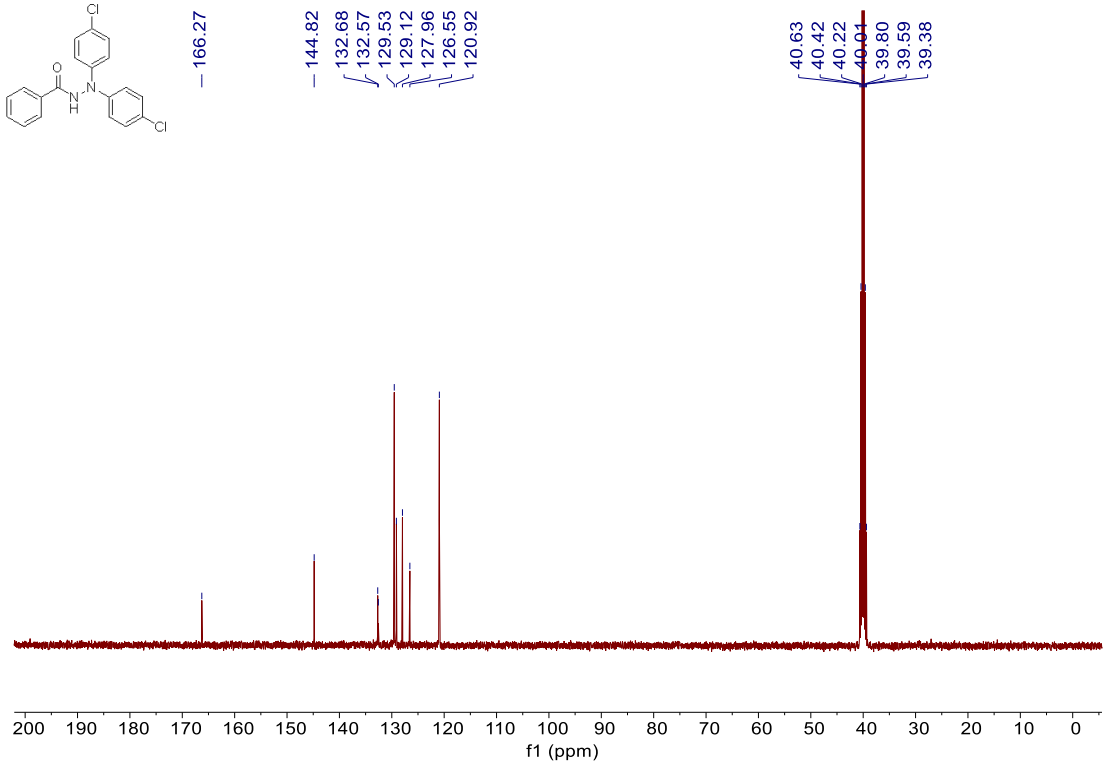
^{1}H NMR of 47 in DMSO-d_6 (400 MHz)



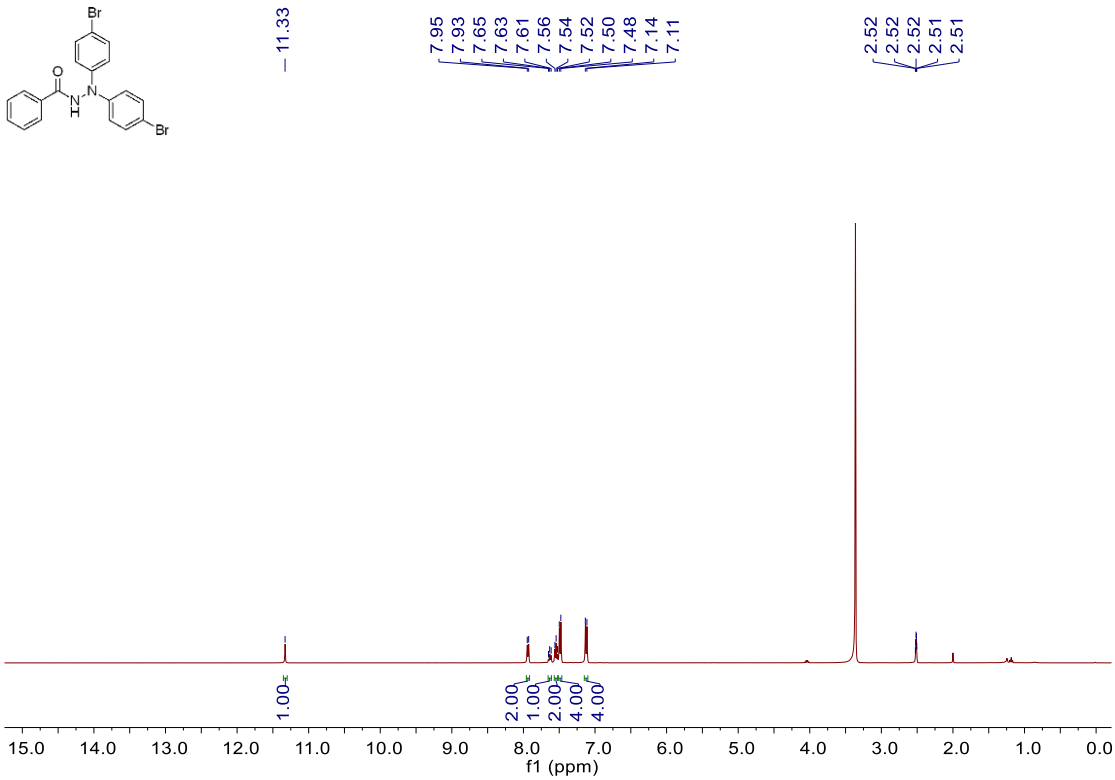
^{13}C{^{1}H} NMR of 47 in DMSO-d_6 (100 MHz)



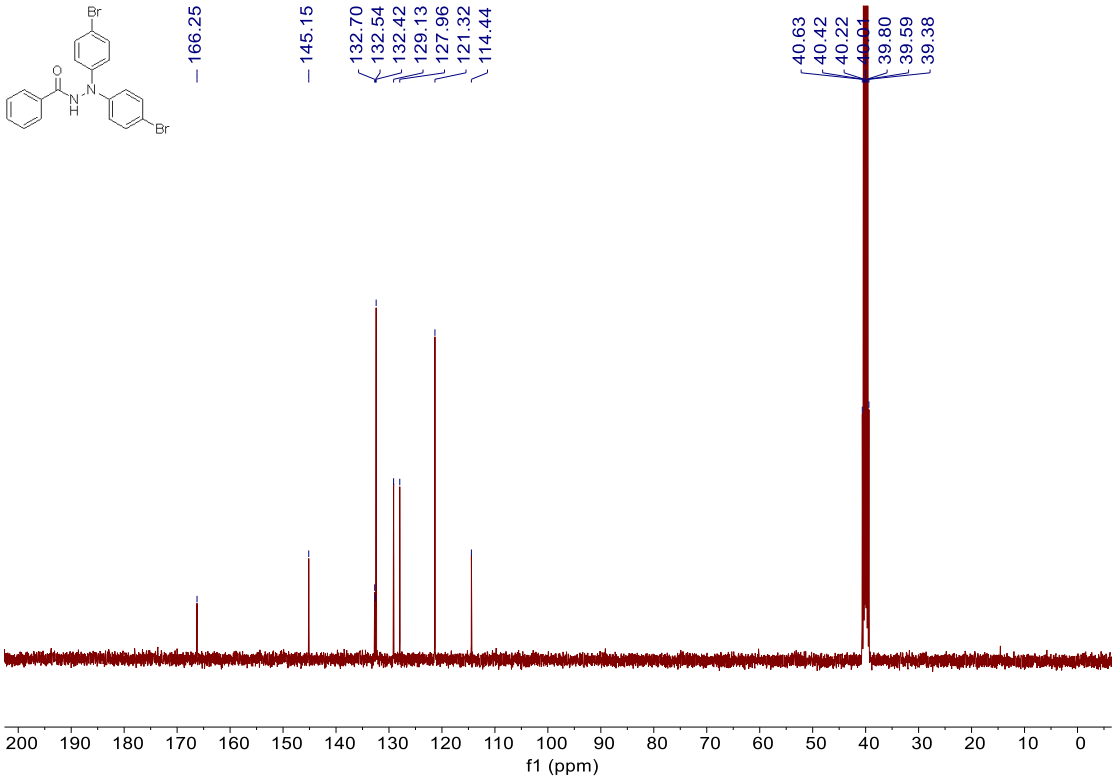
^{1}H NMR of 48 in DMSO-d_6 (400 MHz)



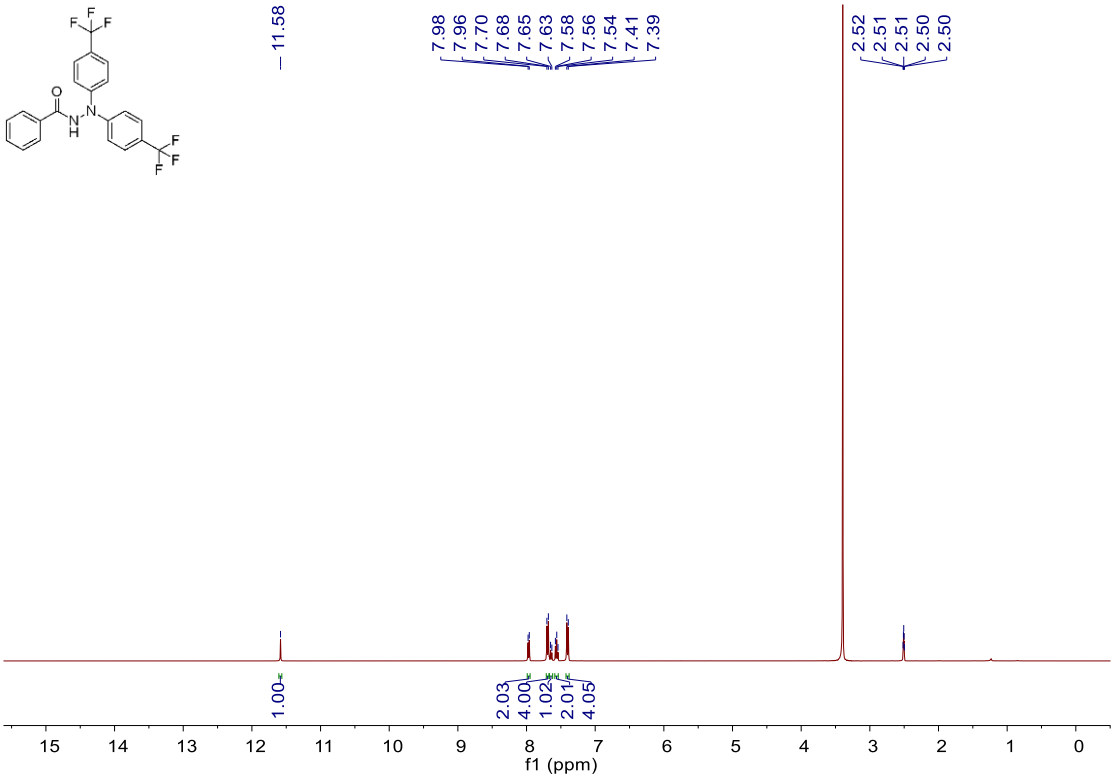
^{13}C{^{1}H} NMR of 48 in DMSO-d_6 (100 MHz)



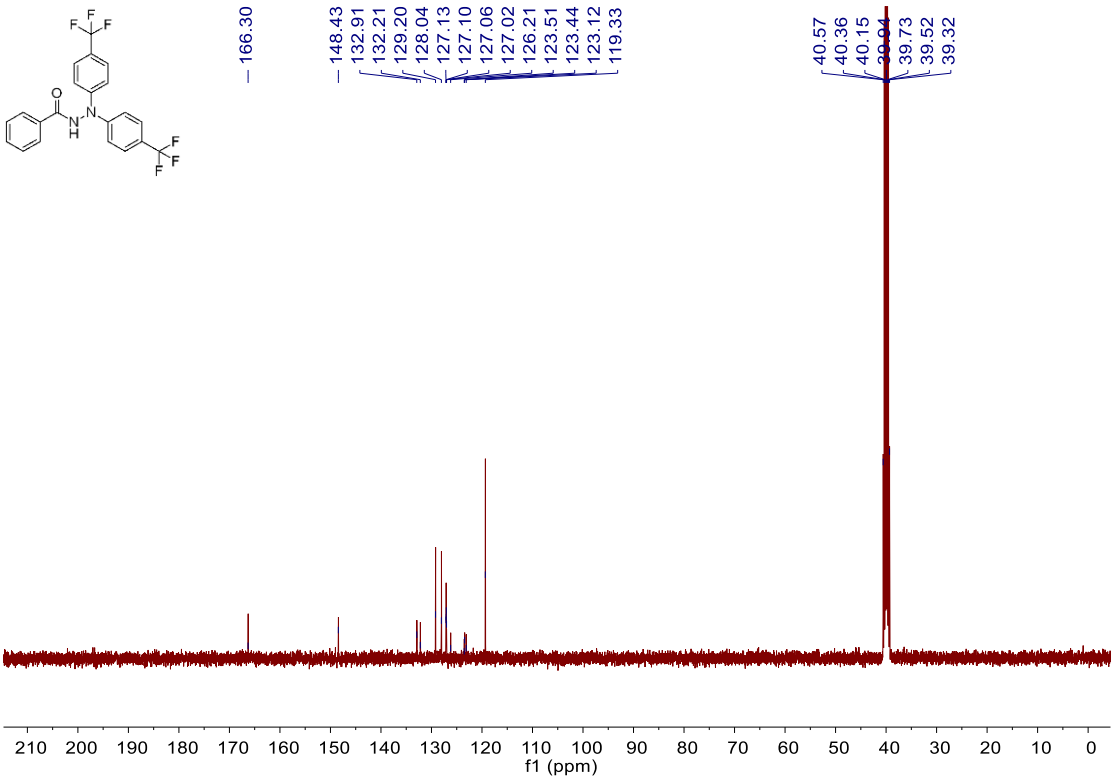
^{1}H NMR of 49 in DMSO-d_6 (400 MHz)



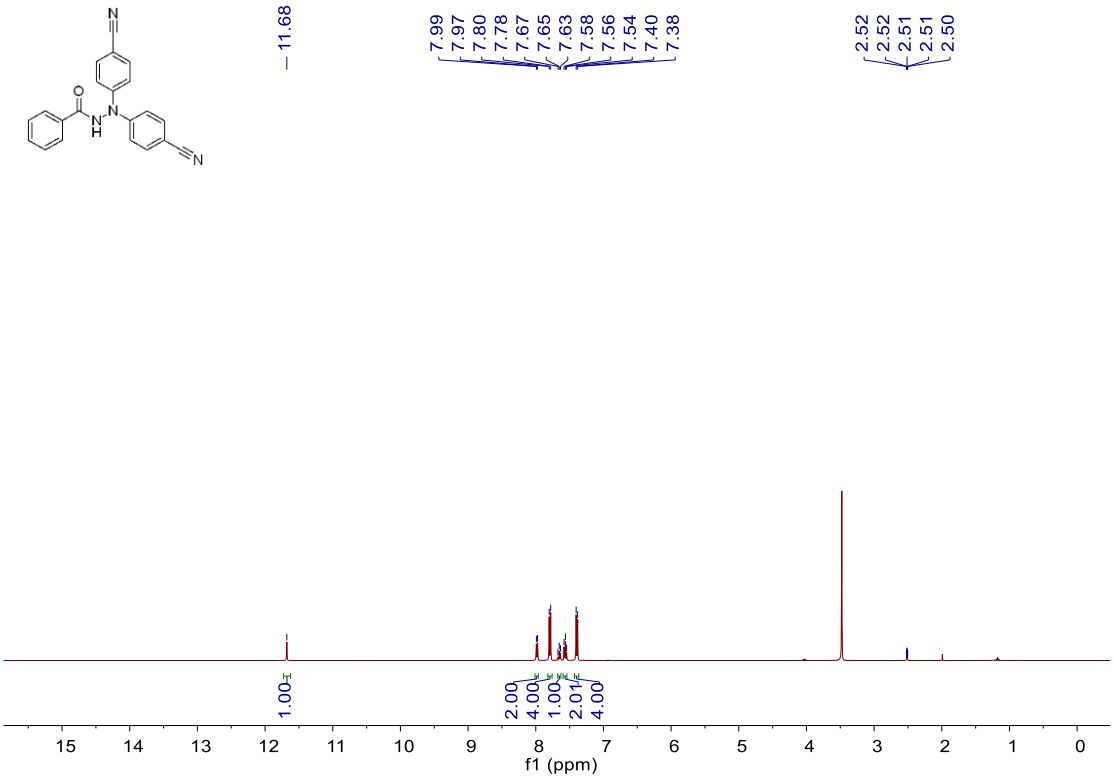
^{13}C{^{1}H} NMR of 49 in DMSO-d_6 (100 MHz)



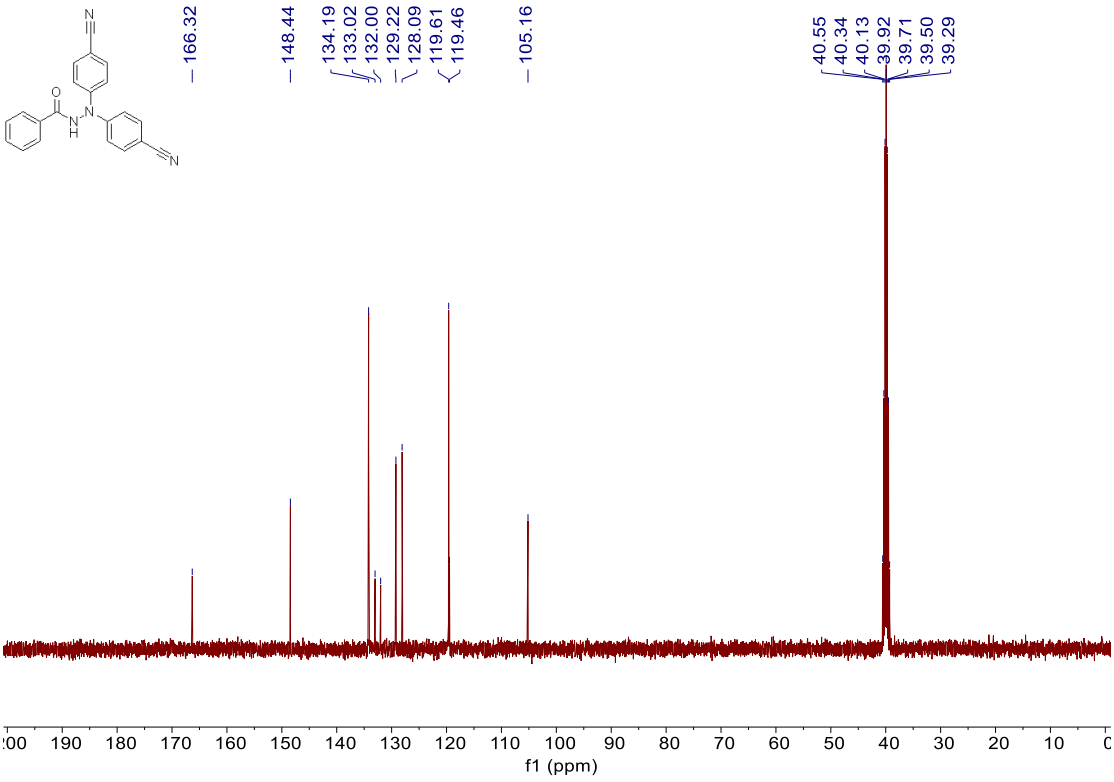
^{1}H NMR of 50 in DMSO-d_6 (400 MHz)



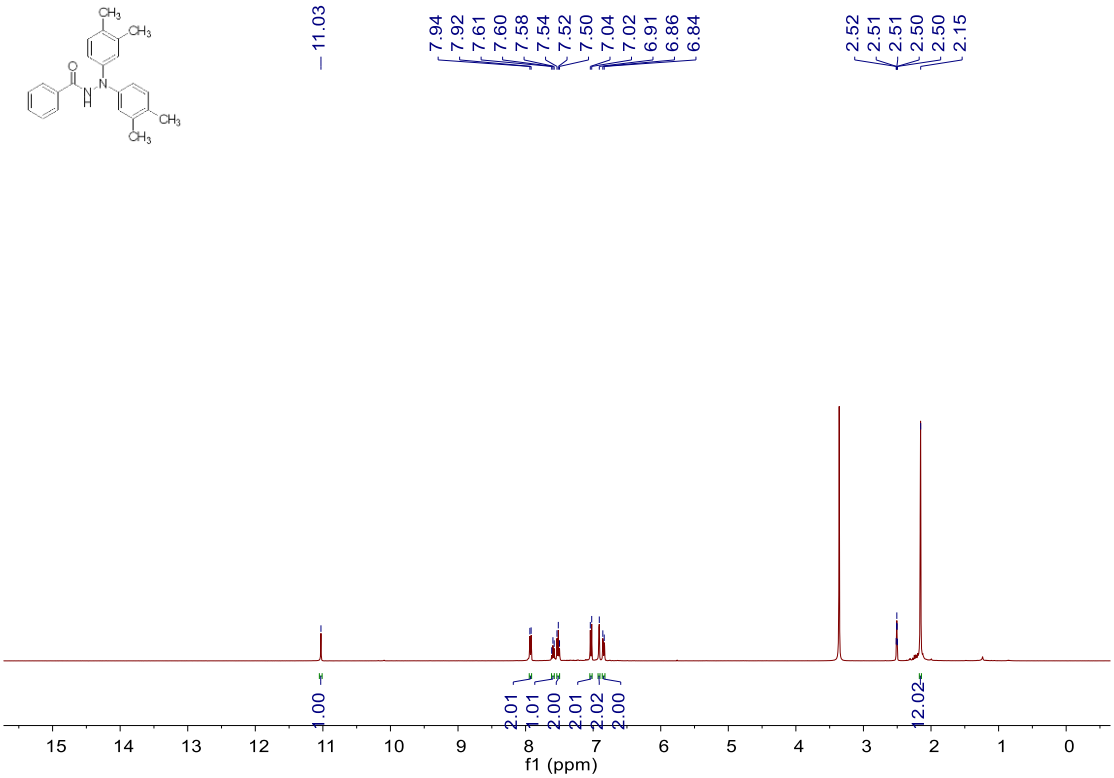
^{13}C{^{1}H} NMR of 50 in DMSO-d_6 (100 MHz)



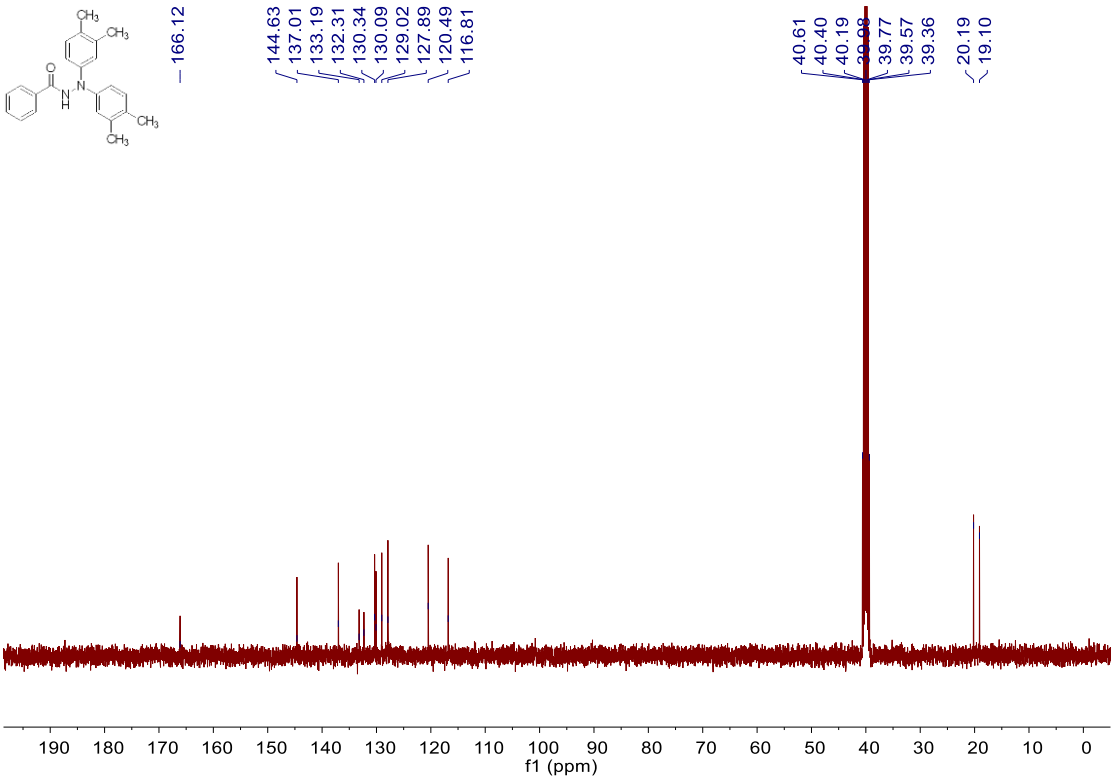
^{1}H NMR of 51 in DMSO-d_6 (400 MHz)



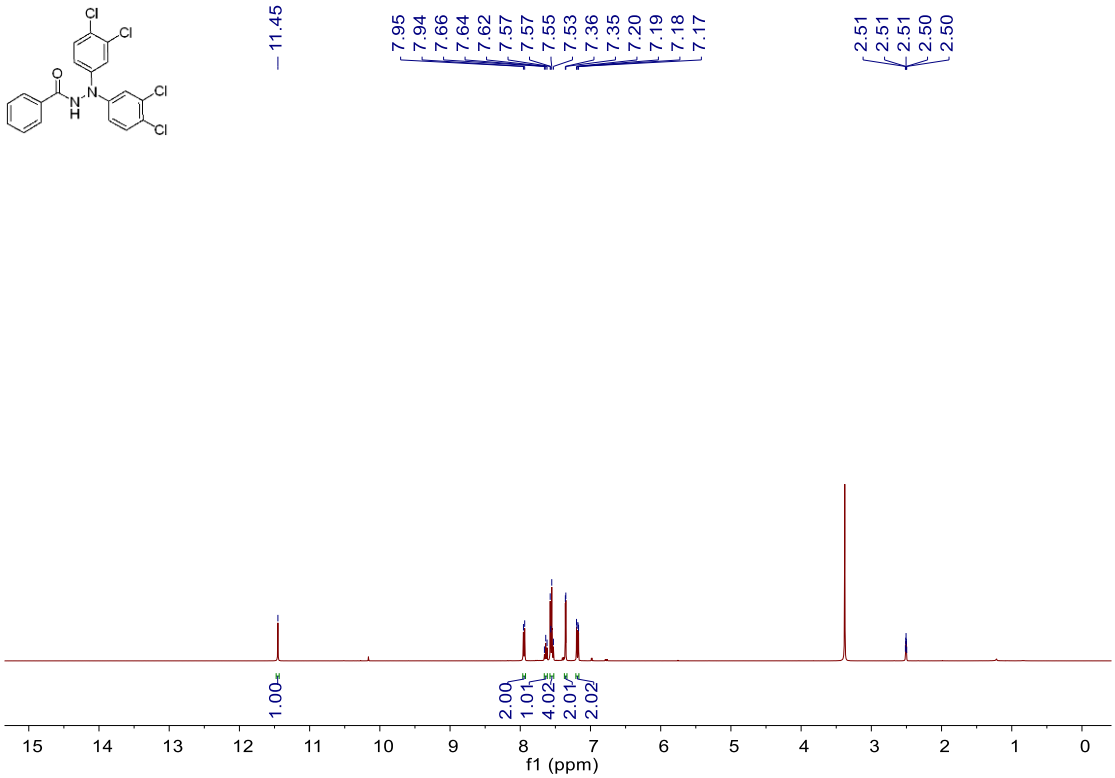
^{13}C{^{1}H} NMR of 51 in DMSO-d_6 (100 MHz)



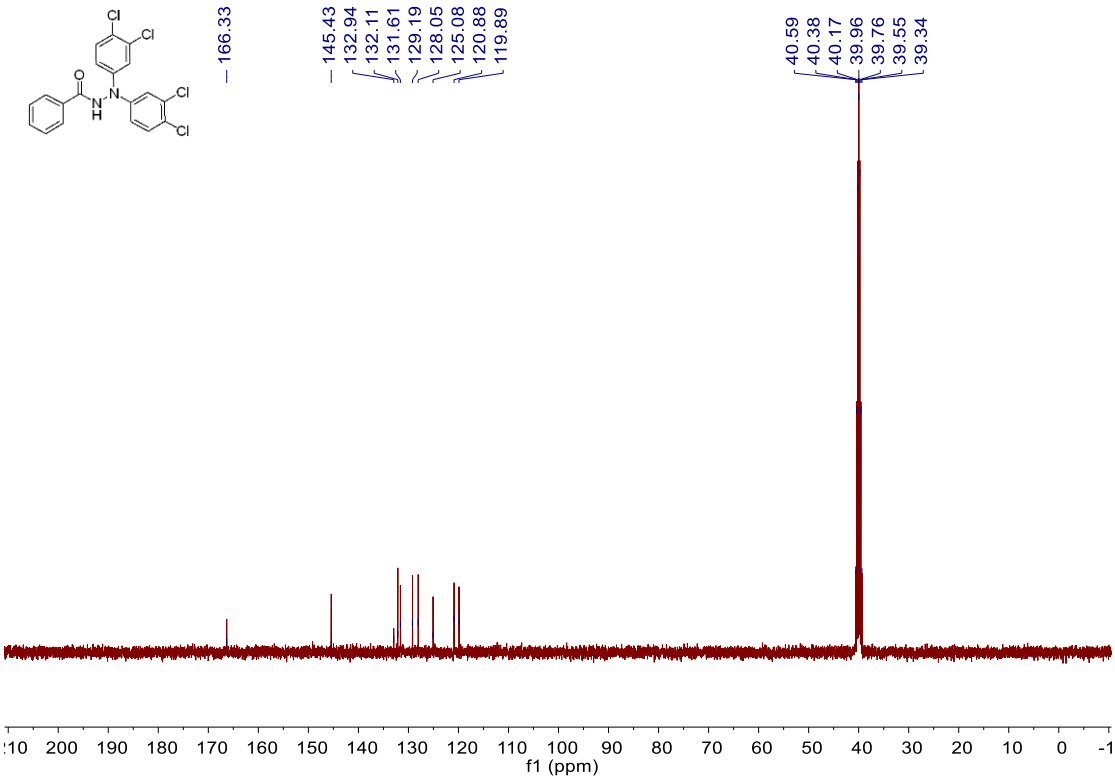
^{1}H NMR of 52 in DMSO-d_6 (400 MHz)



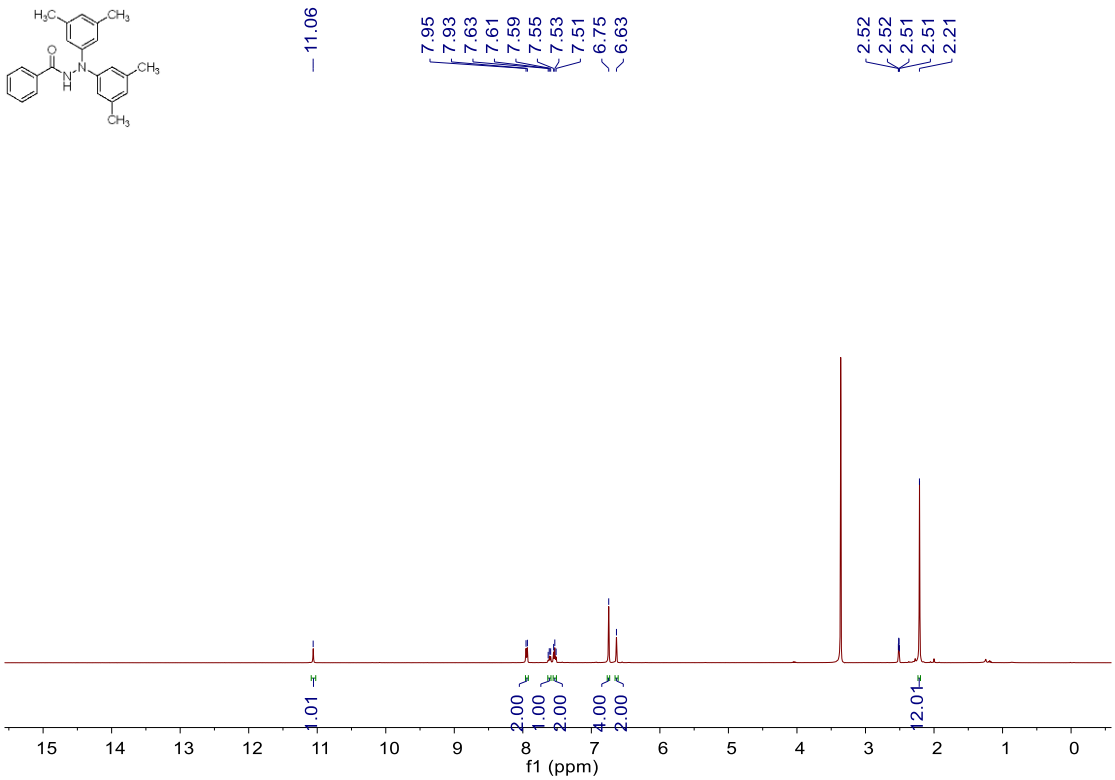
^{13}C{^{1}H} NMR of 52 in DMSO-d_6 (100 MHz)



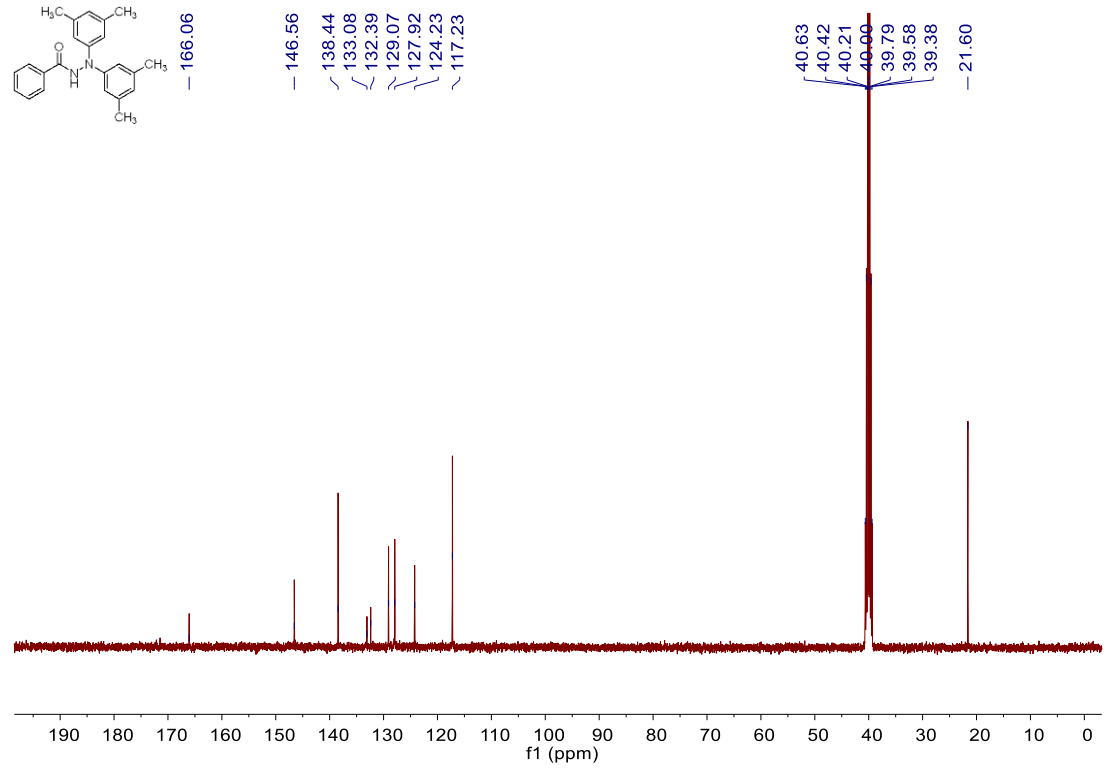
^{1}H NMR of 53 in DMSO-d_6 (400 MHz)



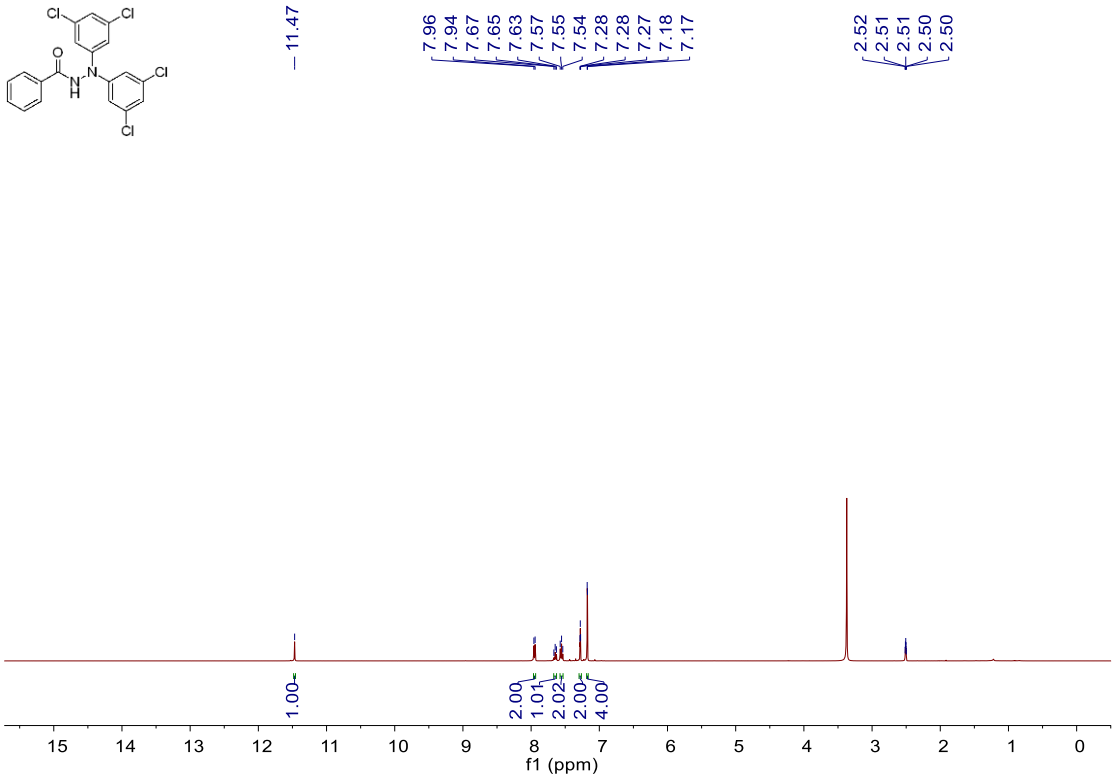
^{13}C{^{1}H} NMR of 53 in DMSO-d_6 (100 MHz)



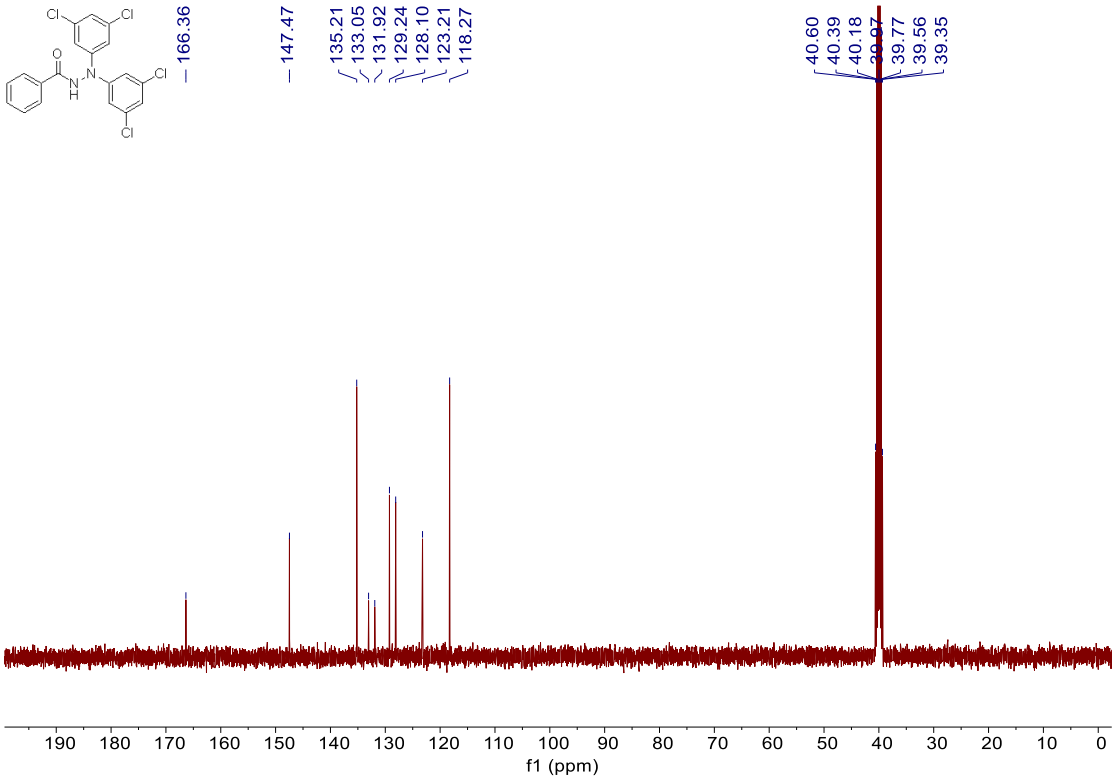
^{1}H NMR of 54 in DMSO-d_6 (400 MHz)



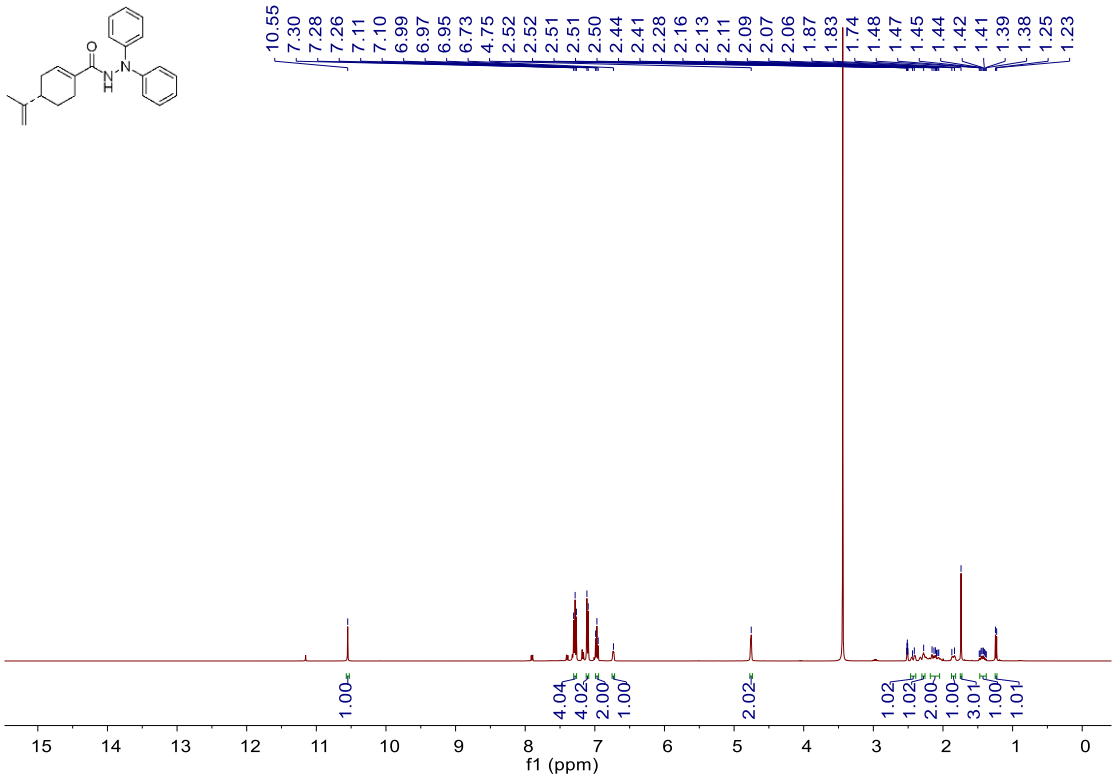
^{13}C{^{1}H} NMR of 54 in DMSO-d_6 (100 MHz)



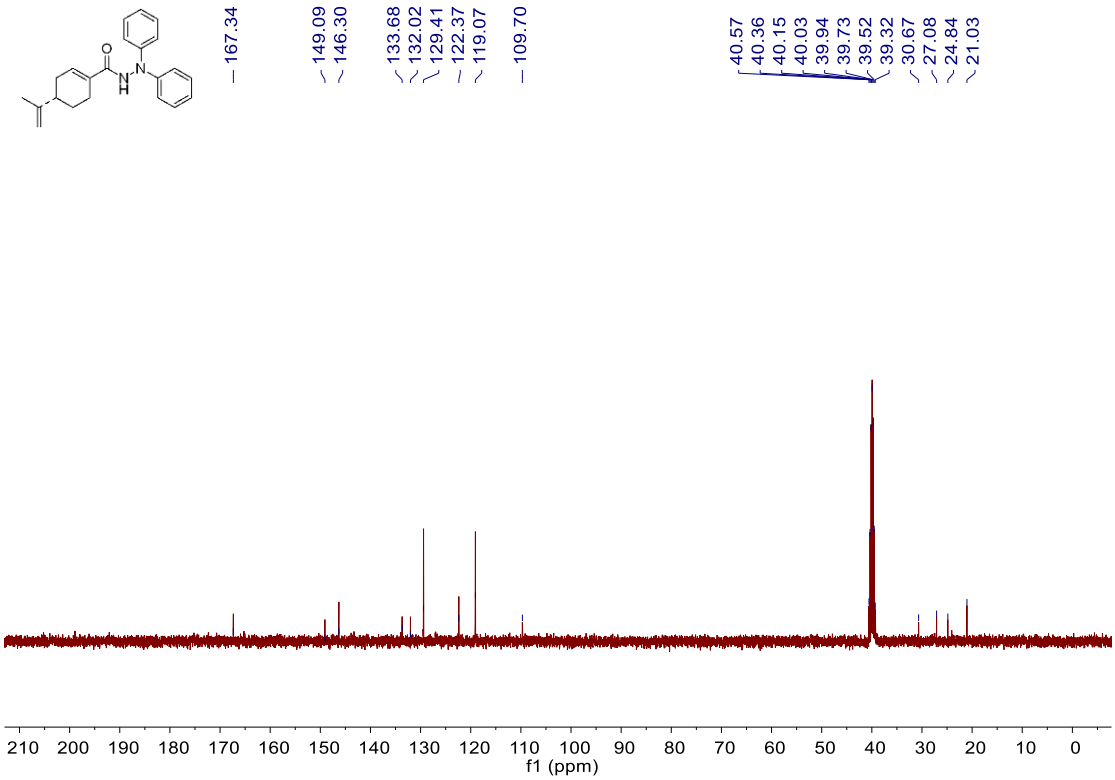
^{1}H NMR of 55 in DMSO-d_6 (400 MHz)



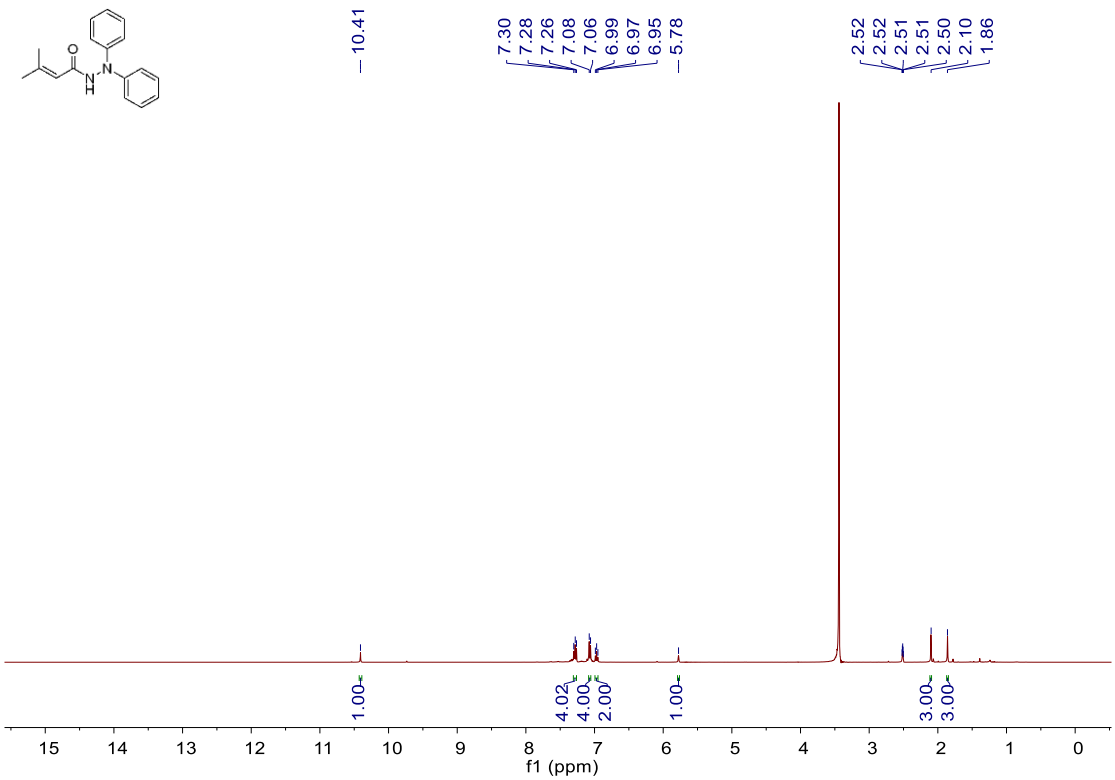
^{13}C{^{1}H} NMR of 55 in DMSO-d_6 (100 MHz)



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



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



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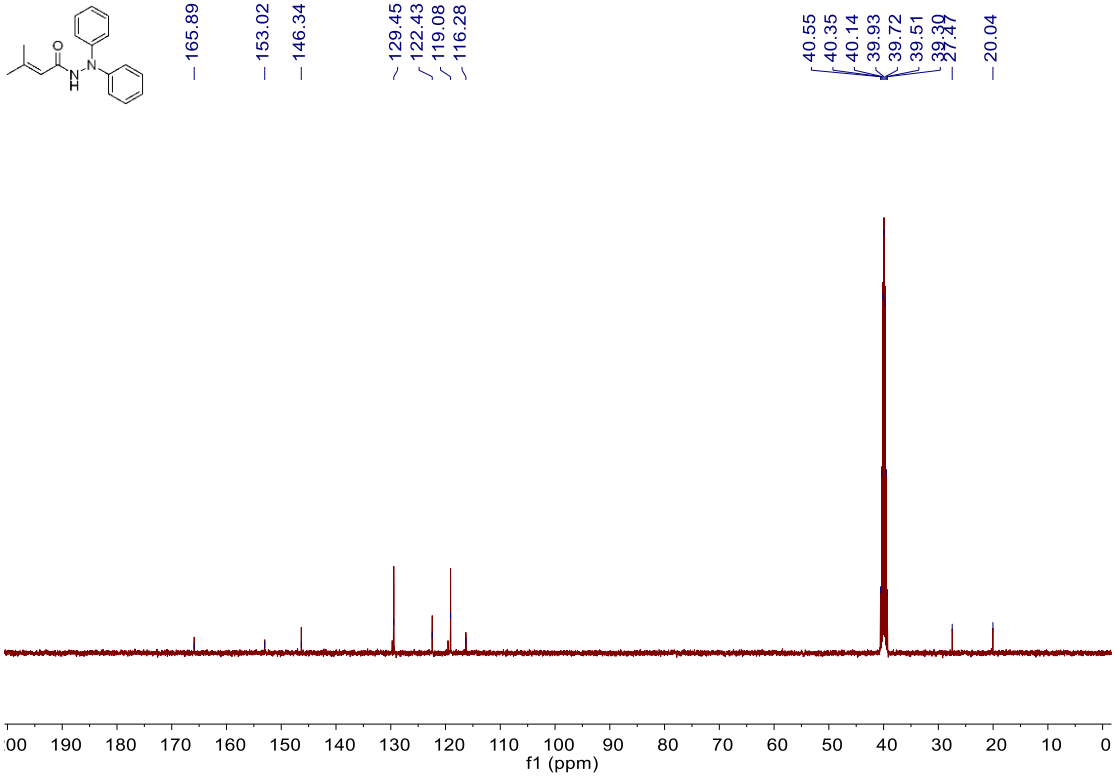
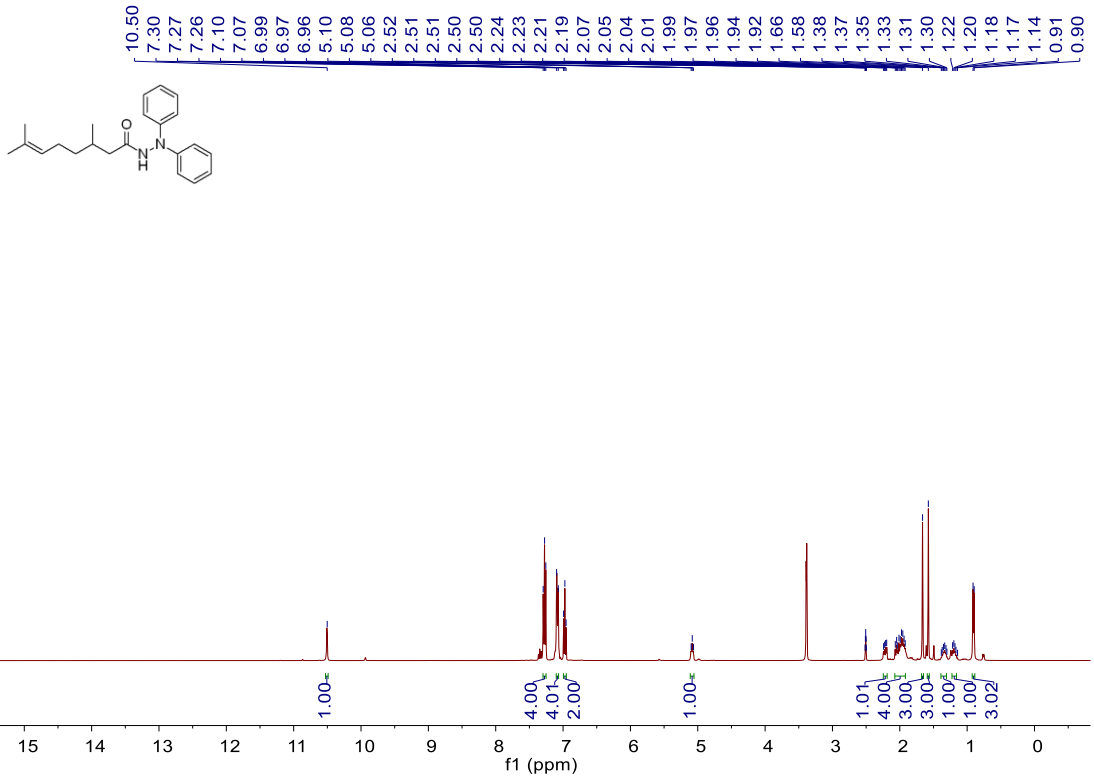
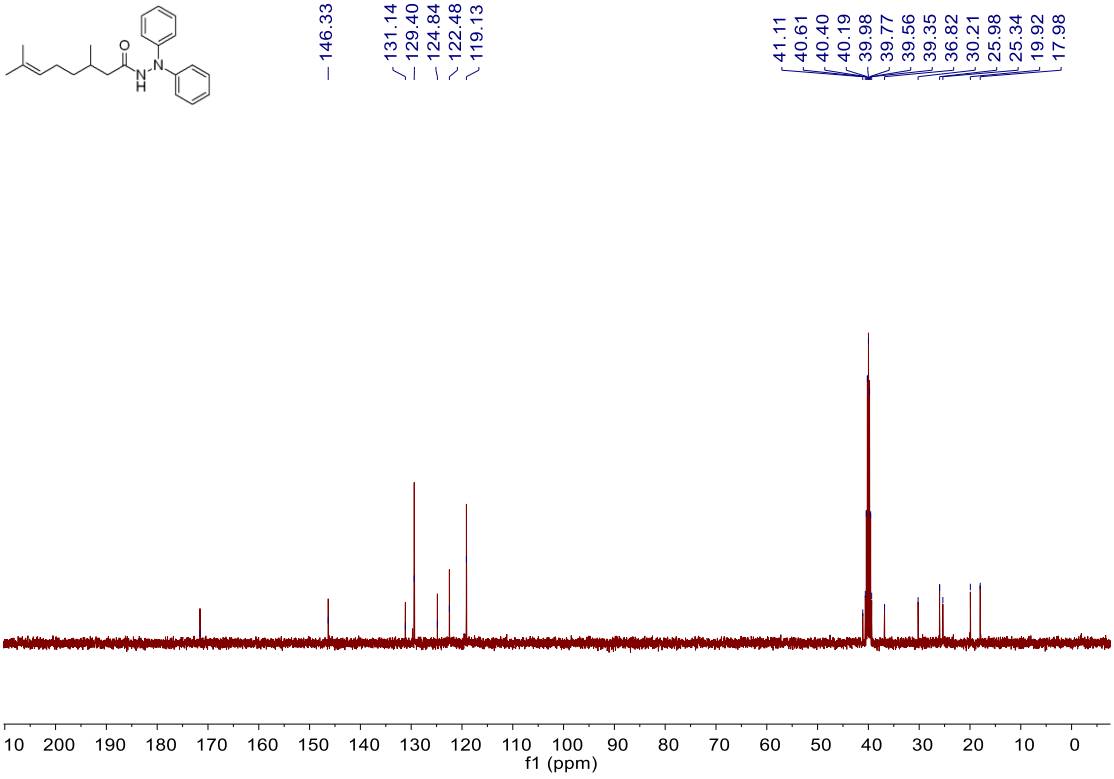


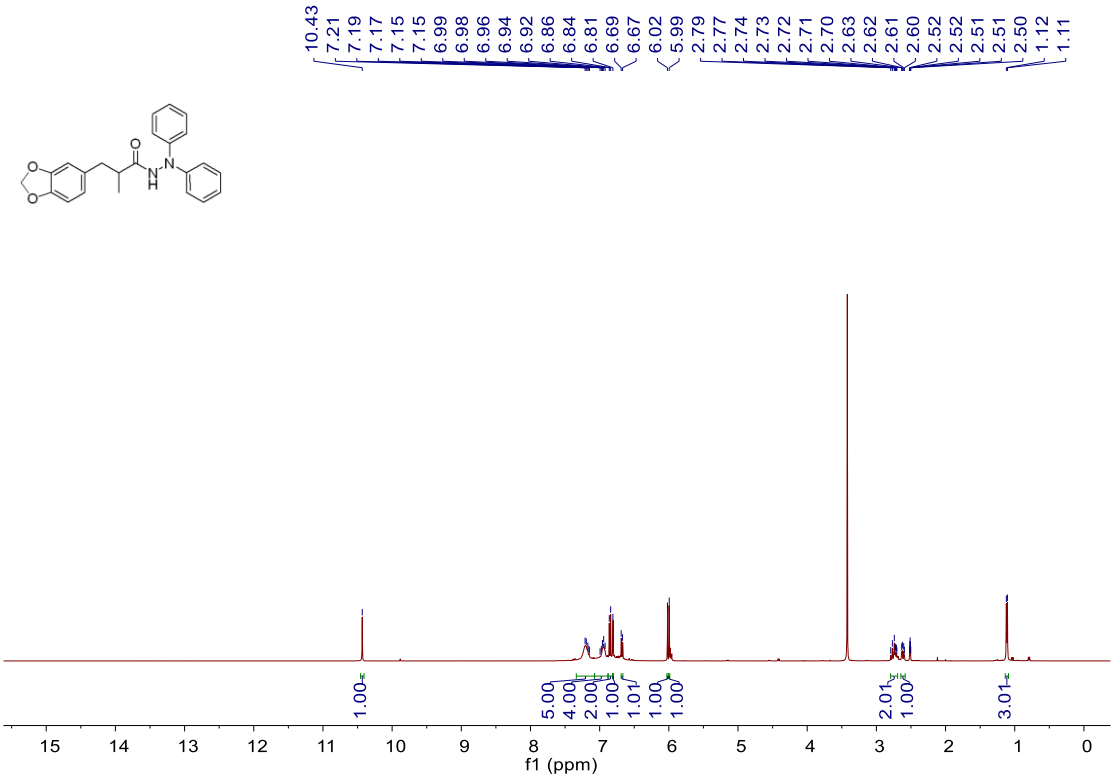
^{13}C{^{1}H} NMR of 59 in DMSO-d_6 (100 MHz)



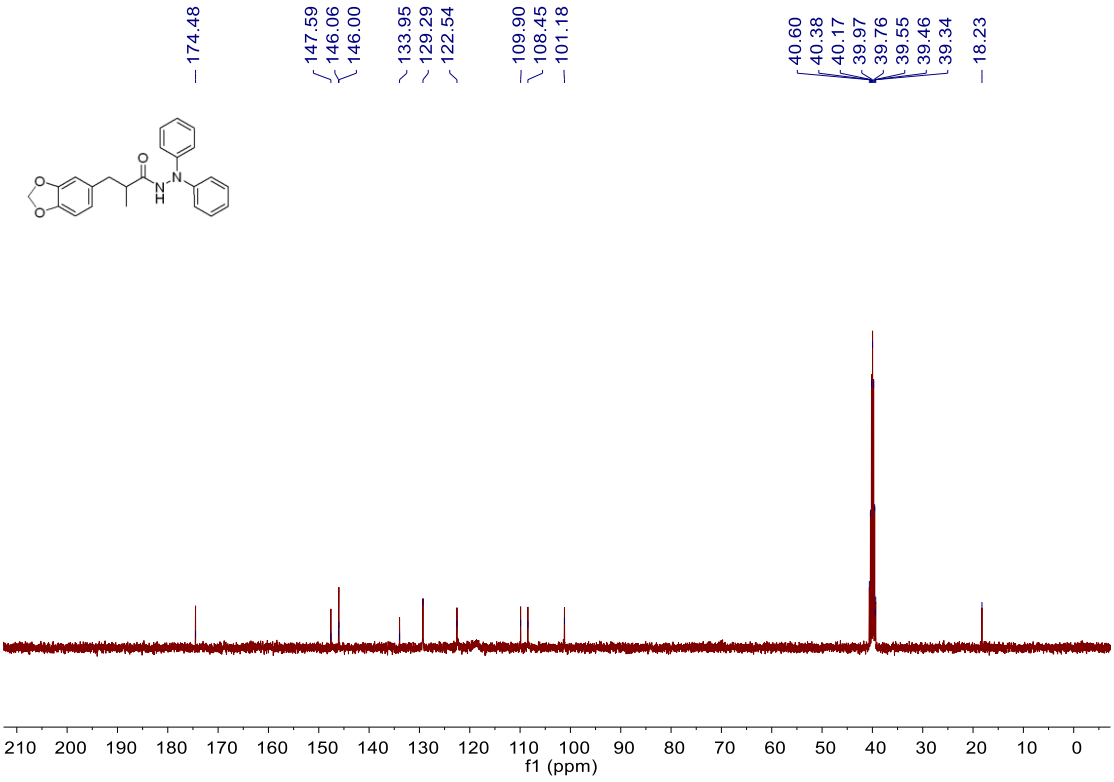
^{1}H NMR of 60 in DMSO-d_6 (400 MHz)



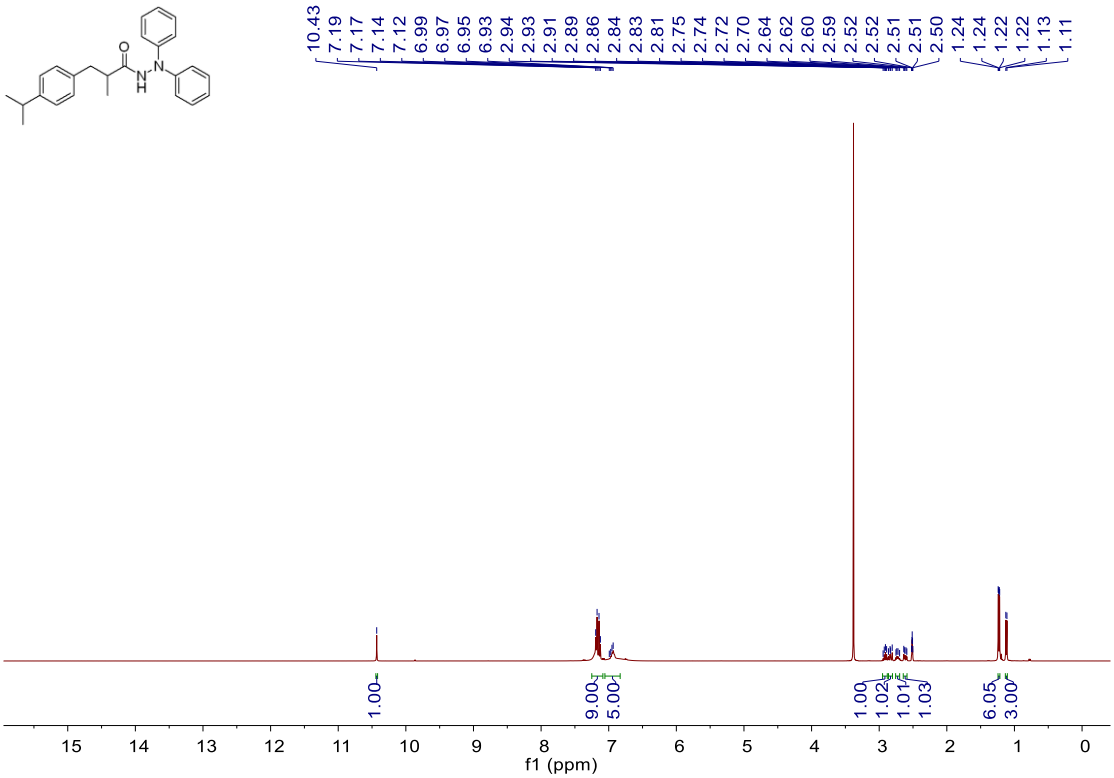
^{13}C{^{1}H} NMR of 60 in DMSO-d_6 (100 MHz)



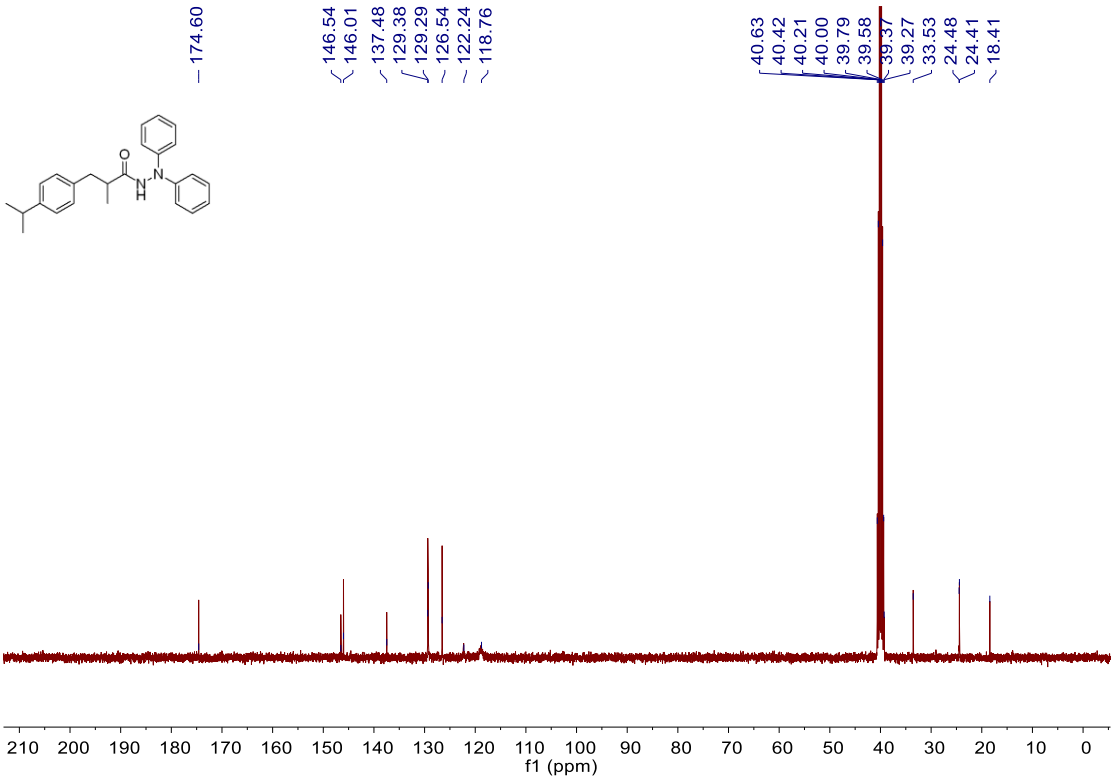
^{1}H NMR of 61 in DMSO-d_6 (400 MHz)



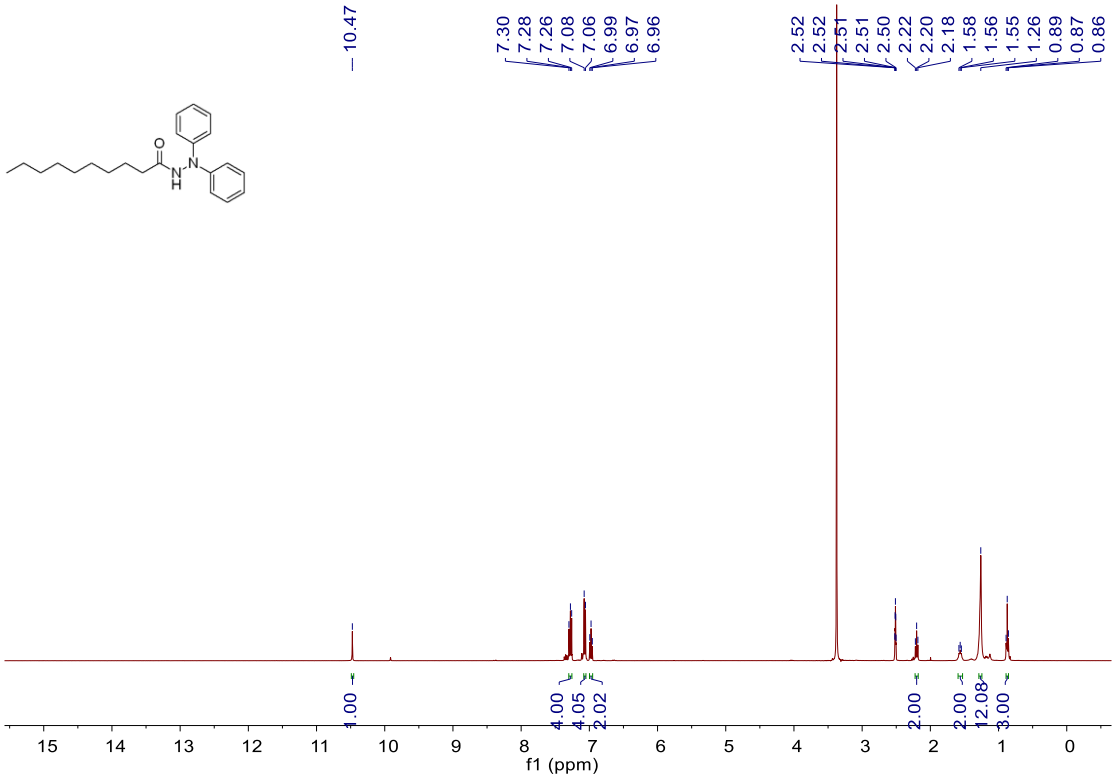
^{13}C{^{1}H} NMR of 61 in DMSO-d_6 (100 MHz)



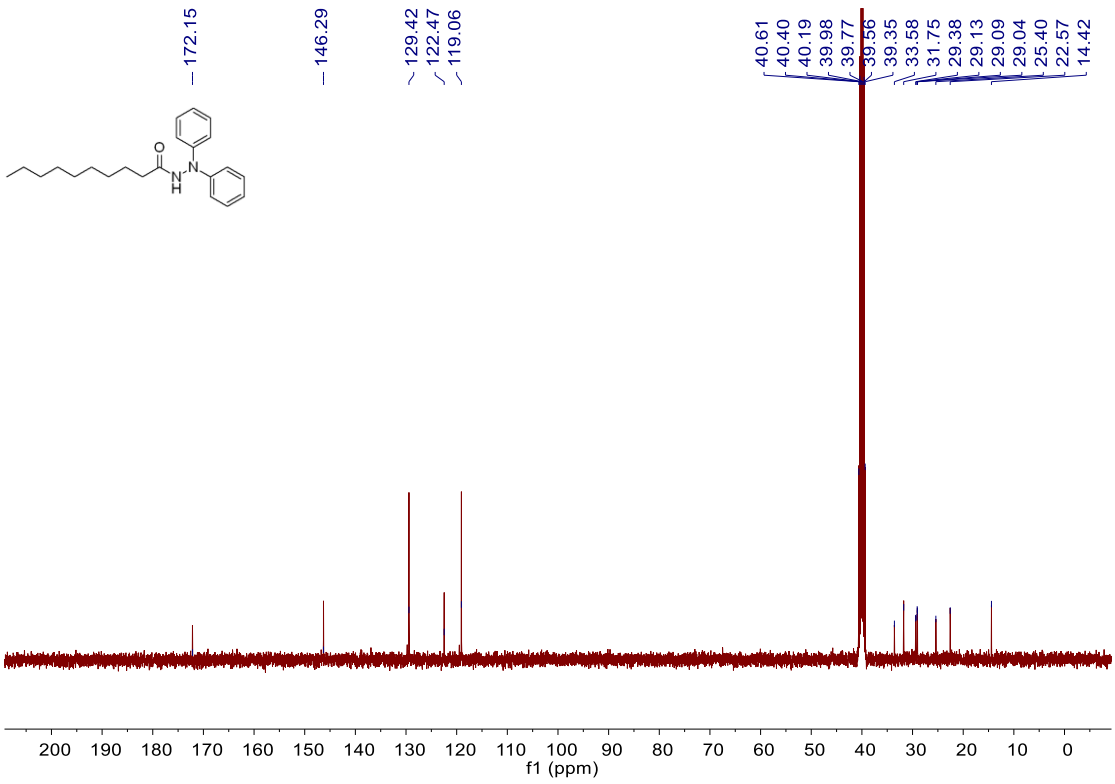
^{1}H NMR of 62 in DMSO-d_6 (400 MHz)



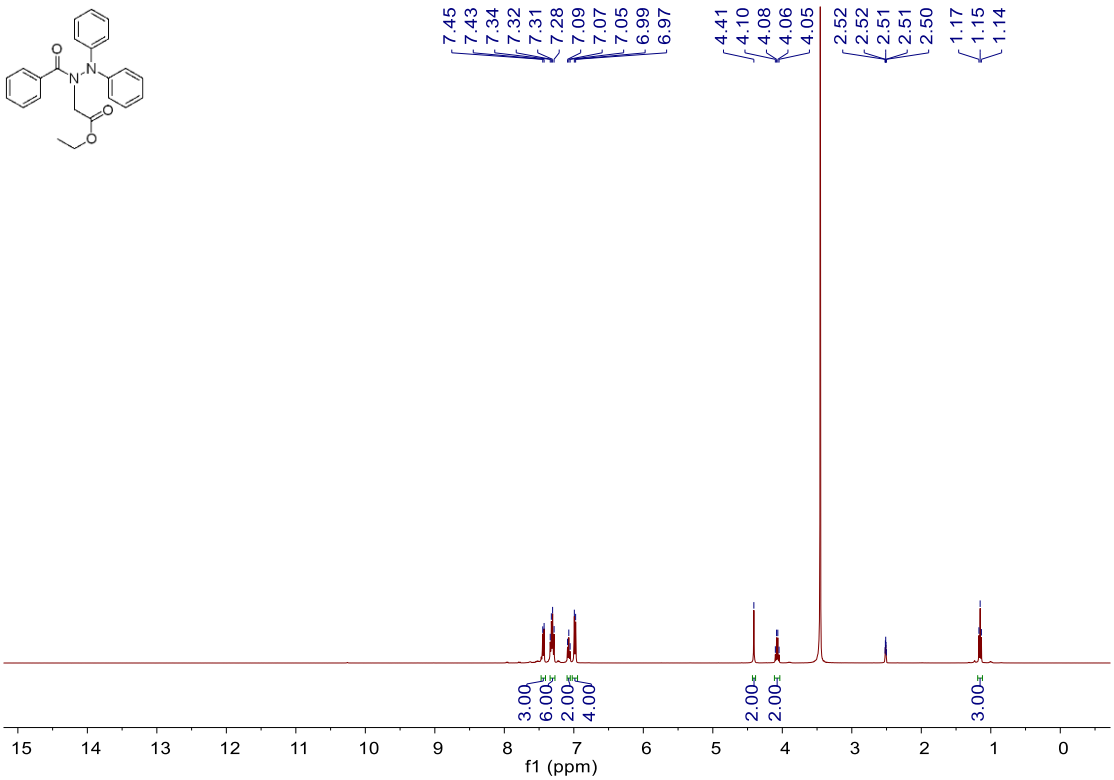
^{13}C{^{1}H} NMR of 62 in DMSO-d_6 (100 MHz)



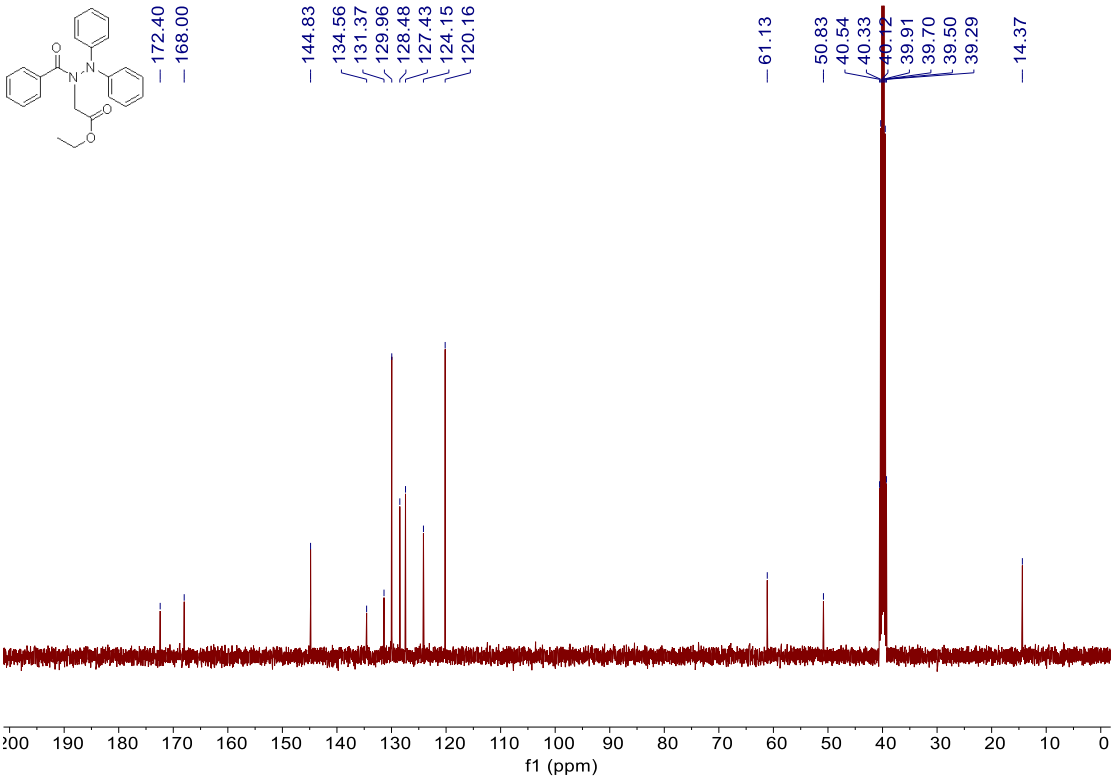
^{1}H NMR of 63 in DMSO-d_6 (400 MHz)



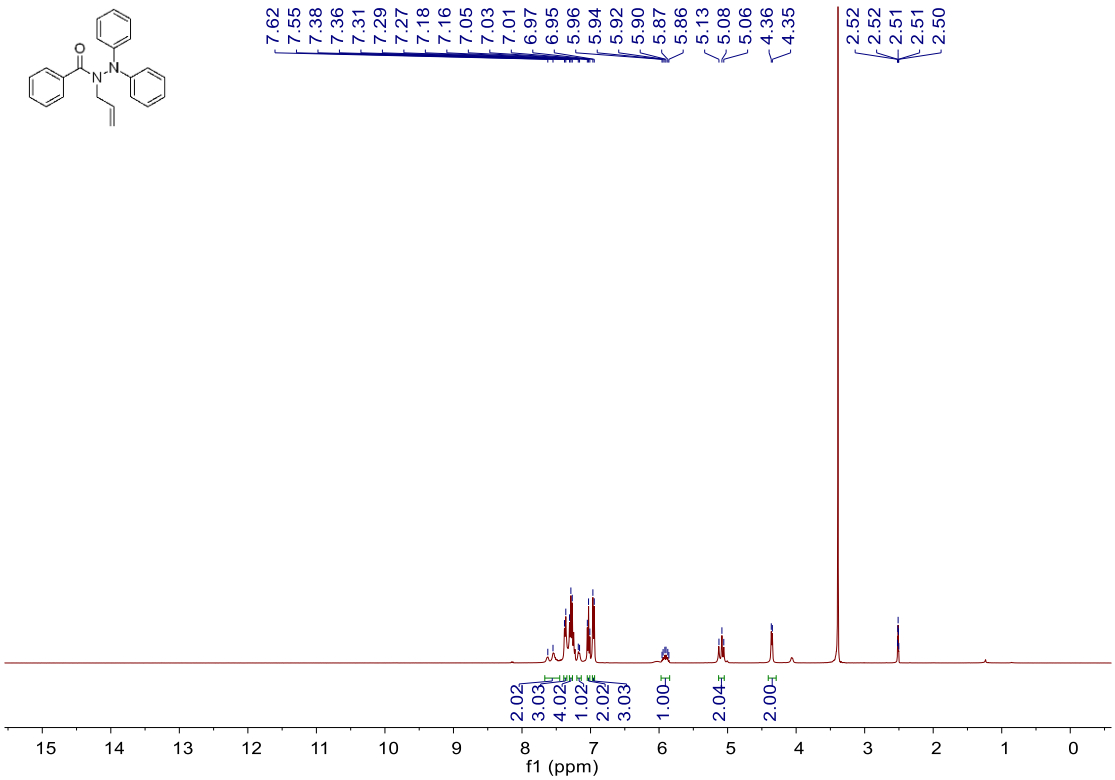
^{13}C{^{1}H} NMR of 63 in DMSO-d_6 (100 MHz)



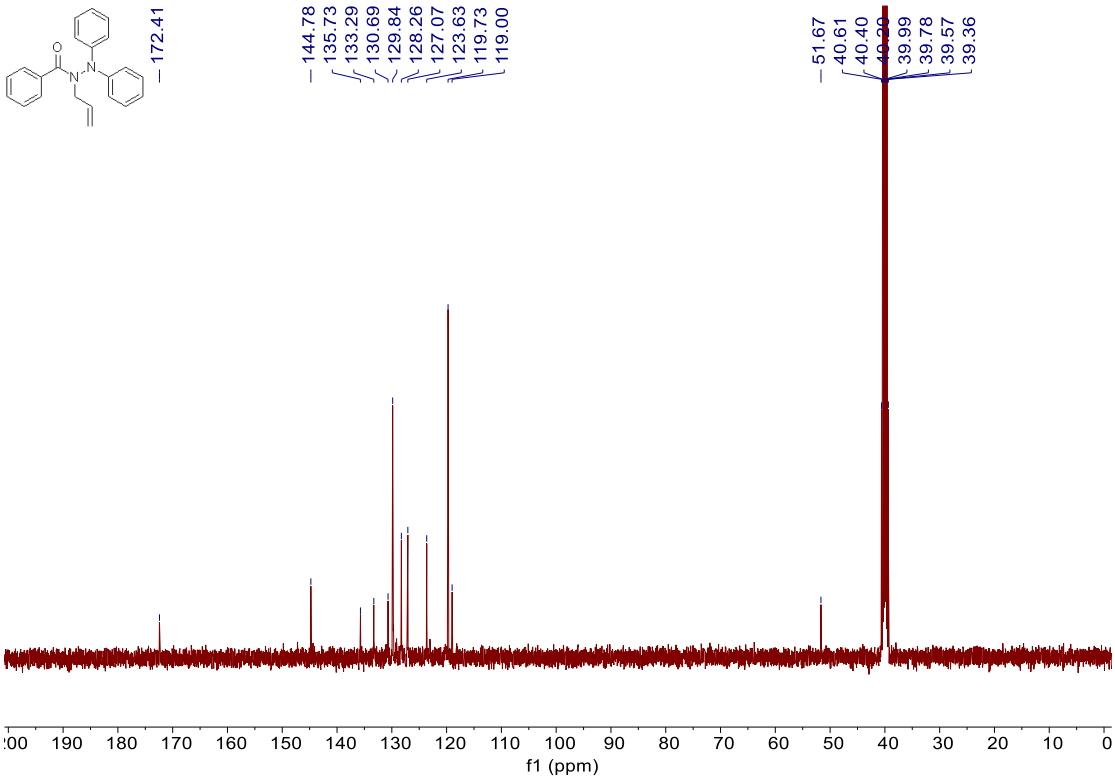
^{1}H NMR of 64 in DMSO-d_6 (400 MHz)



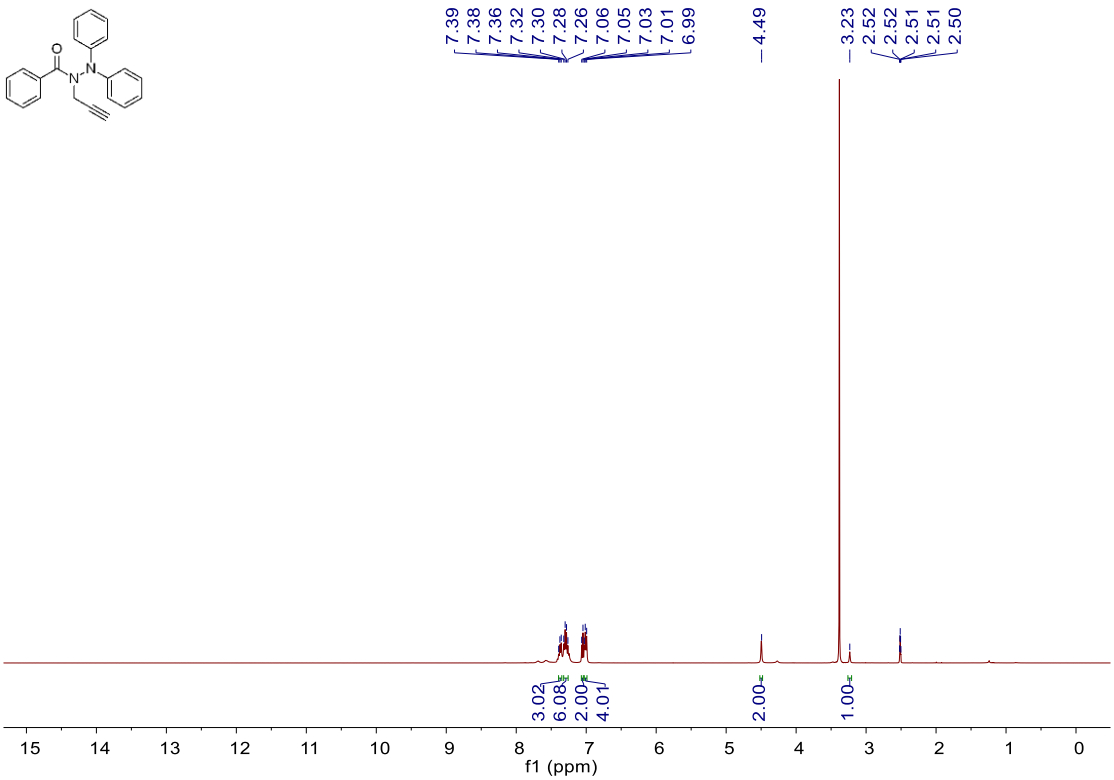
^{13}C{^{1}H} NMR of 64 in DMSO-d_6 (100 MHz)



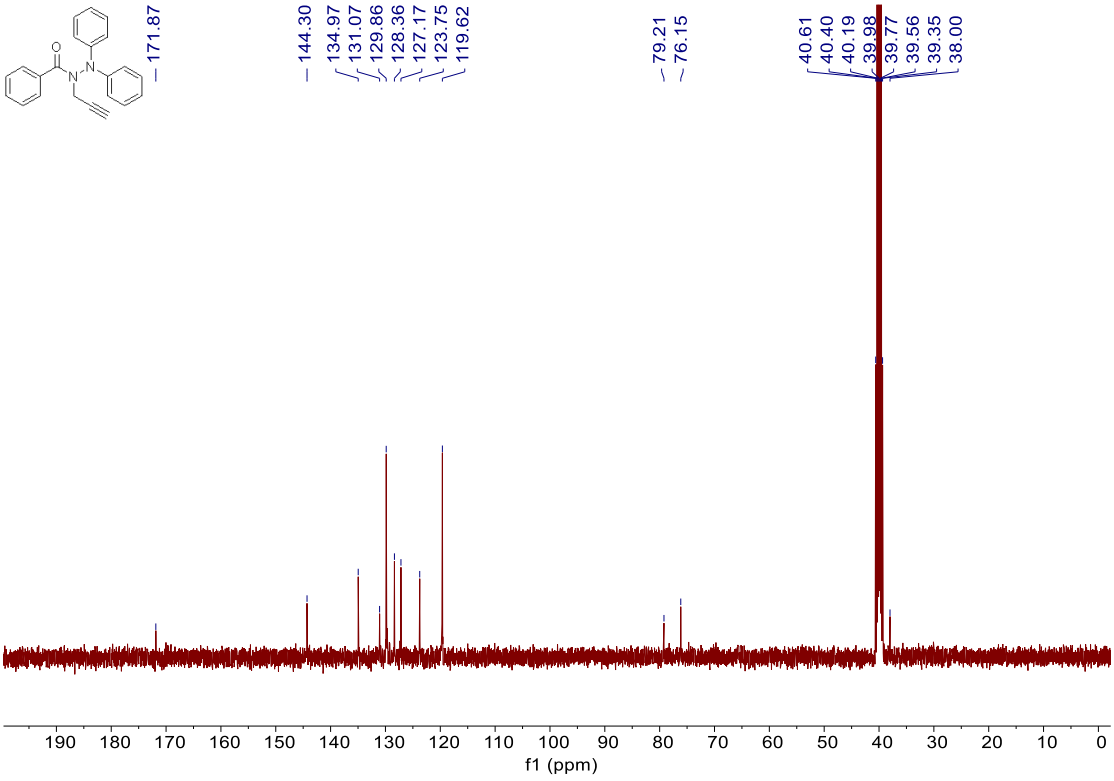
^{1}H NMR of 65 in DMSO-d_6 (400 MHz)



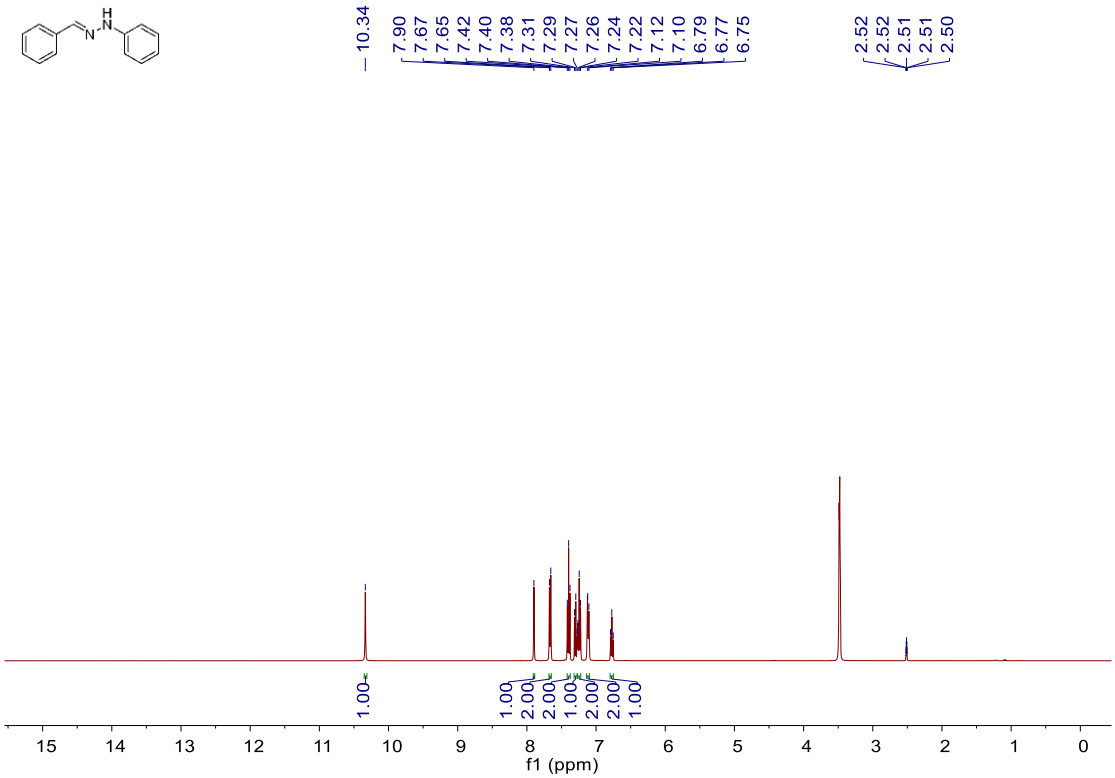
^{13}C{^{1}H} NMR of 65 in DMSO-d_6 (100 MHz)



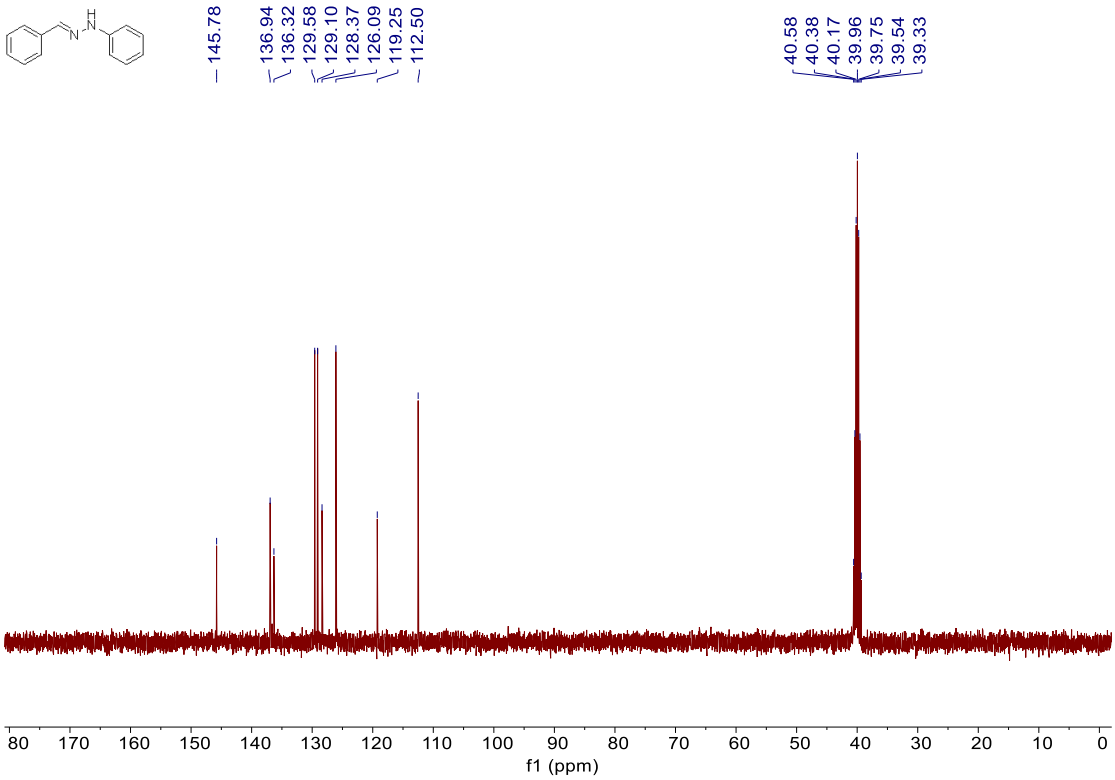
^{1}H NMR of 66 in DMSO-d_6 (400 MHz)



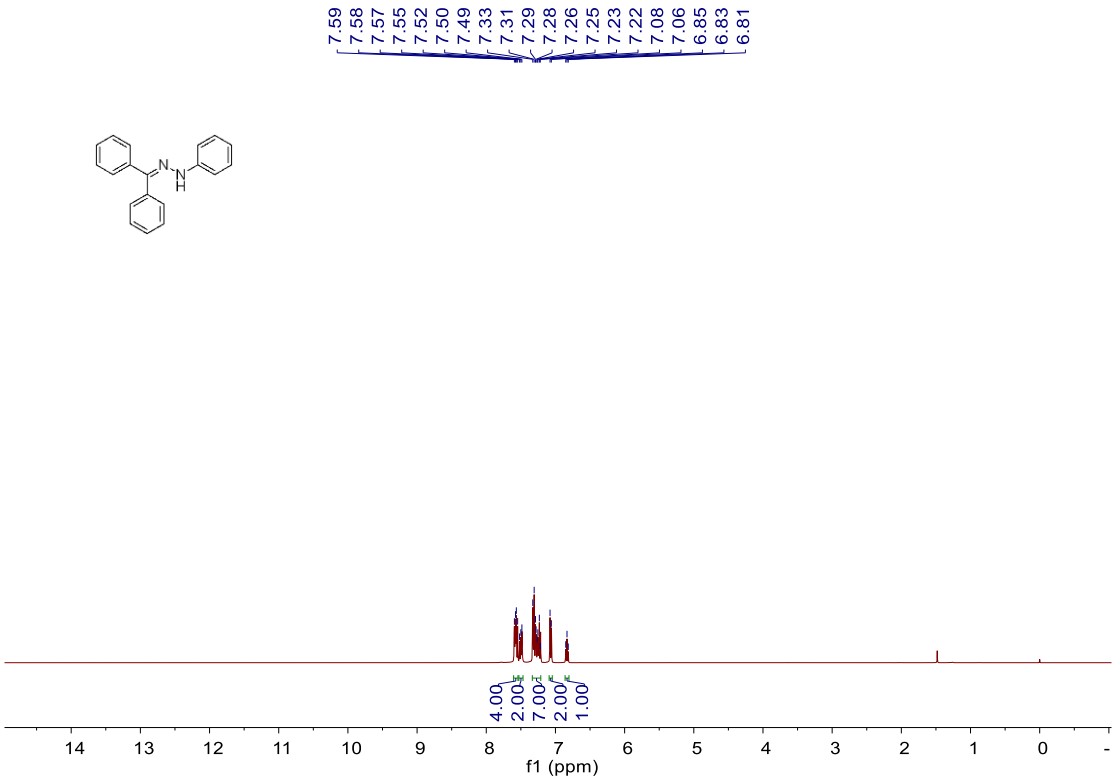
^{13}C{^{1}H} NMR of 66 in DMSO-d_6 (100 MHz)



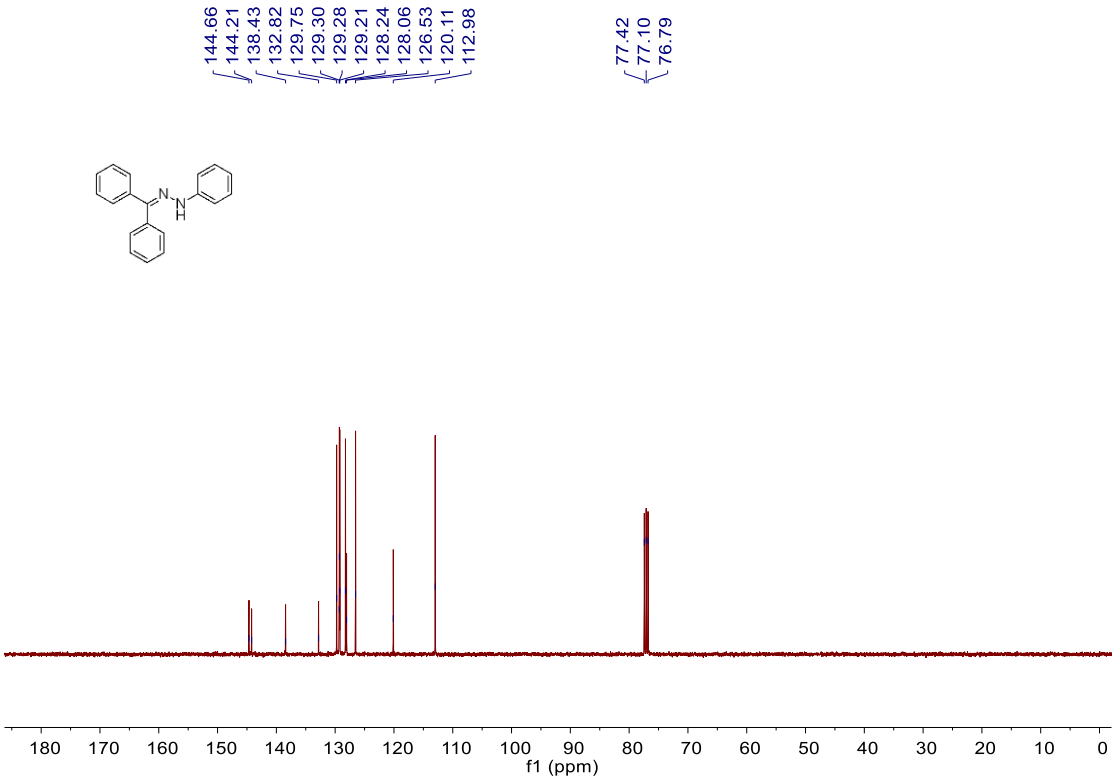
^{1}H NMR of 69 in DMSO-d_6 (400 MHz)



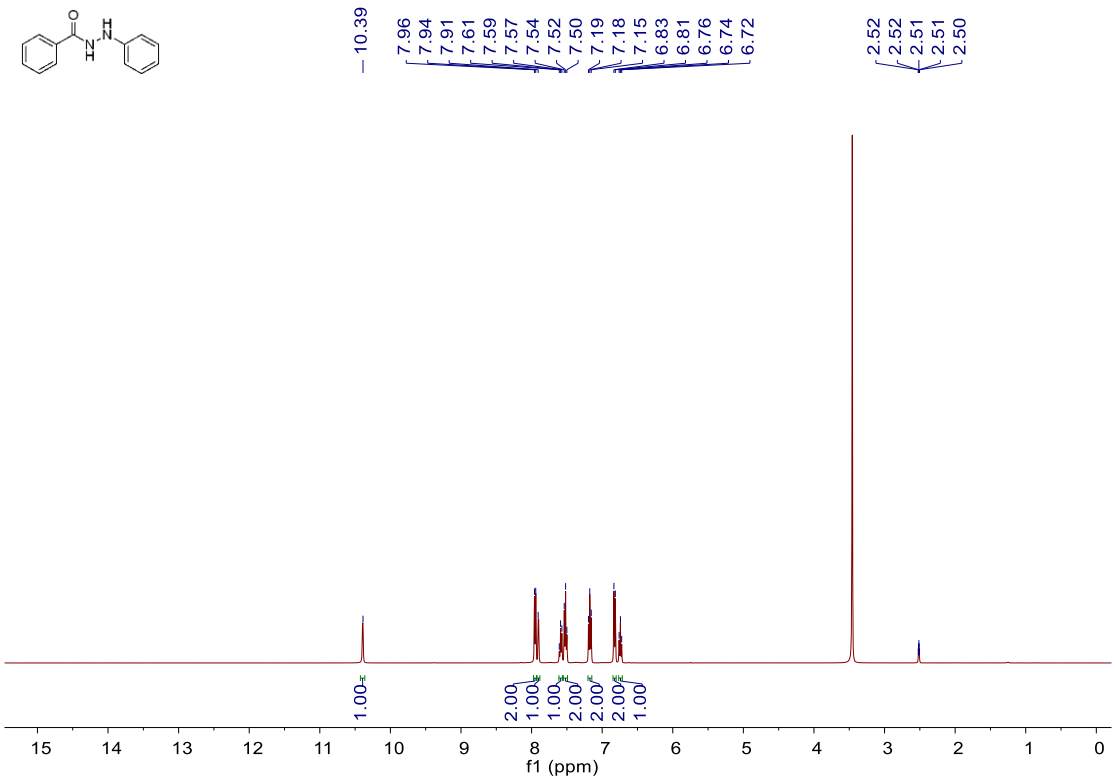
^{13}C{^{1}H} NMR of 69 in DMSO-d_6 (100 MHz)



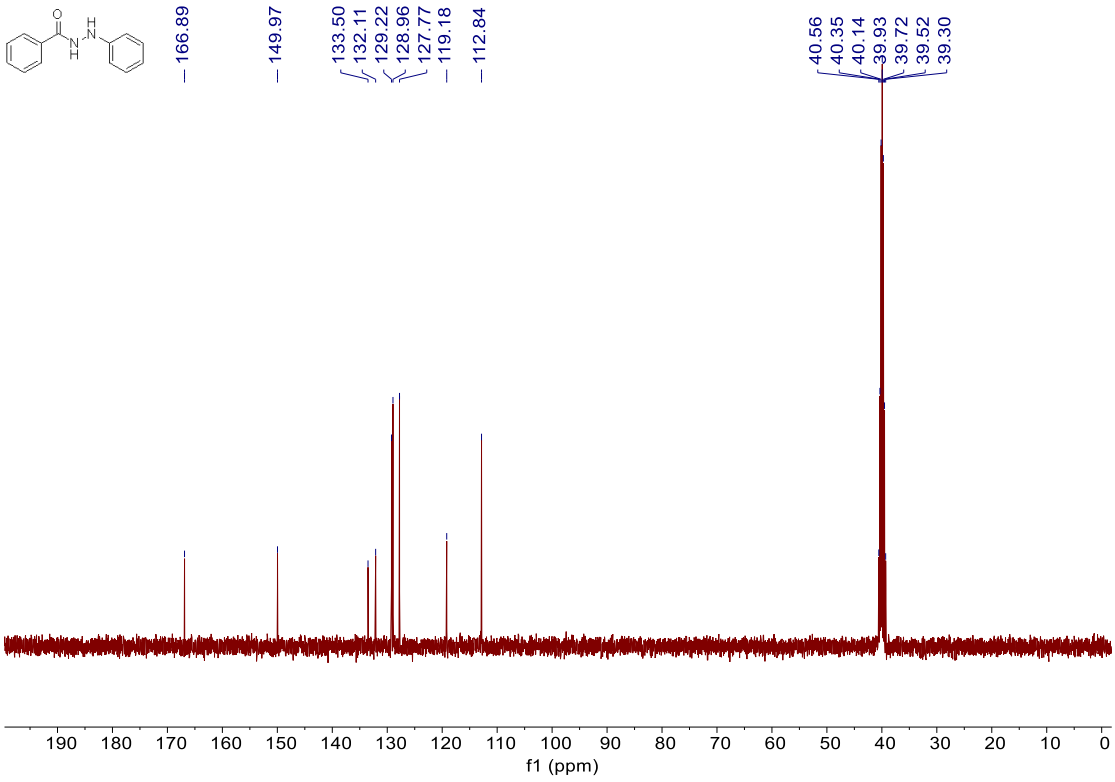
^{1}H NMR of 70 in $CDCl_3$ (400 MHz)



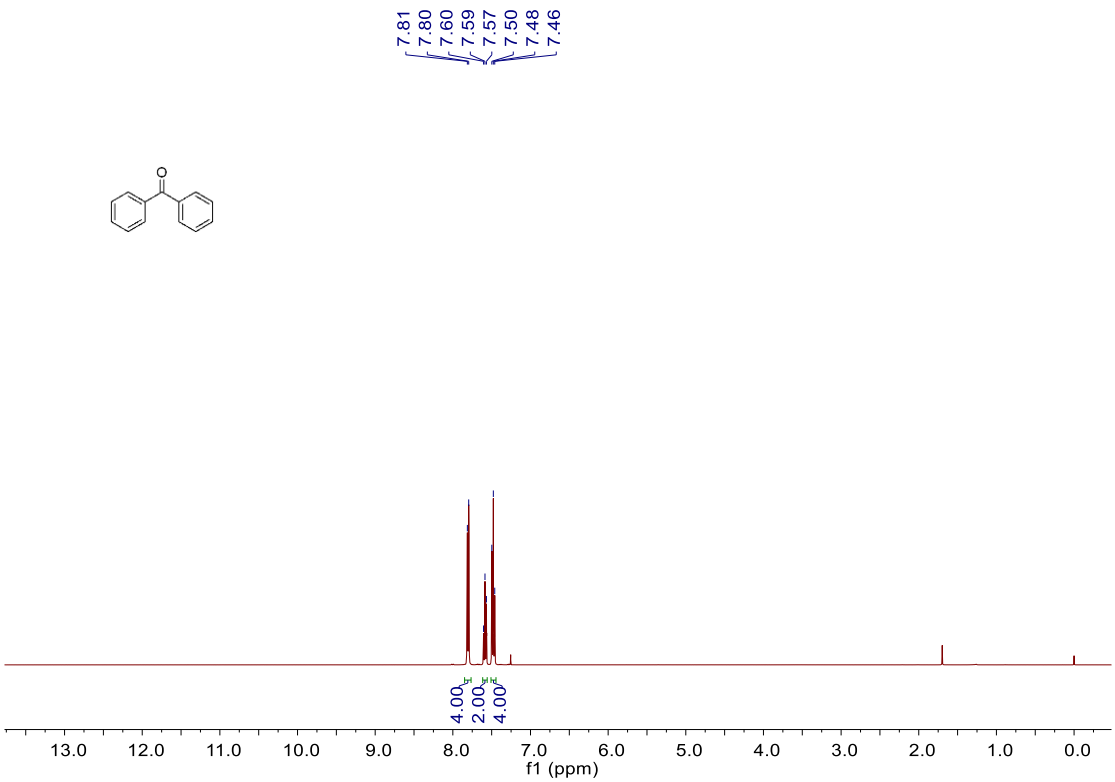
^{13}C{^{1}H} NMR of 70 in $CDCl_3$ (100 MHz)



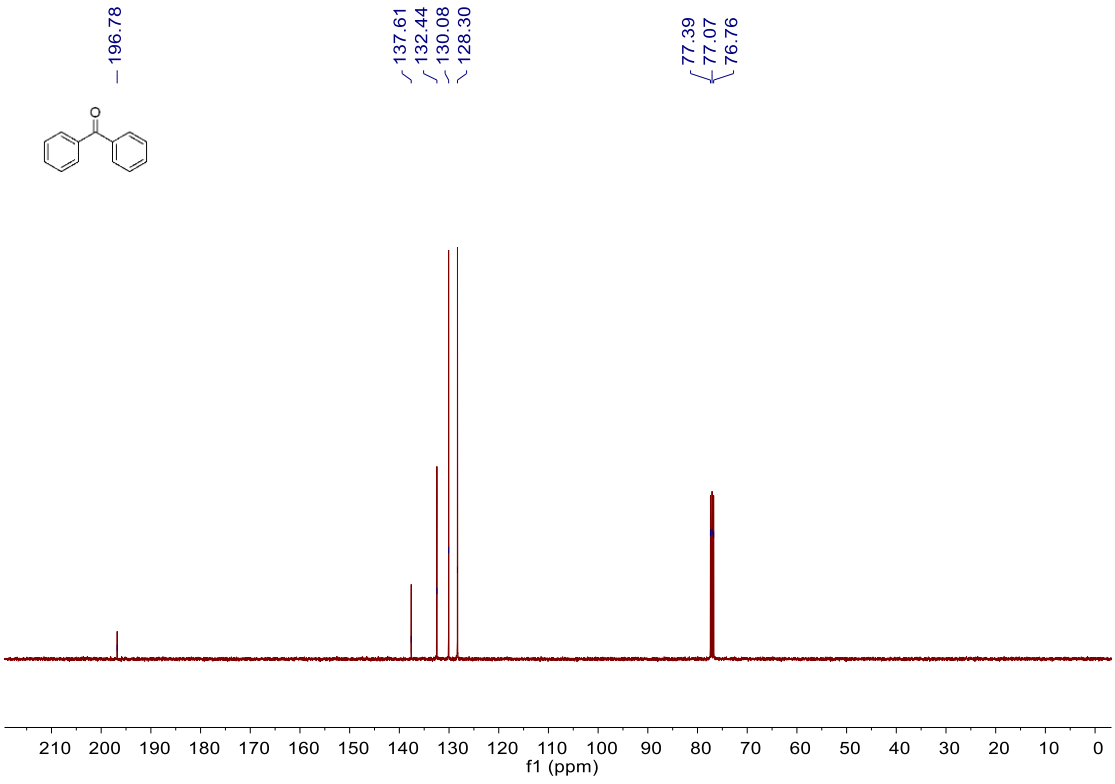
^{1}H NMR of 71 in DMSO-d_6 (400 MHz)



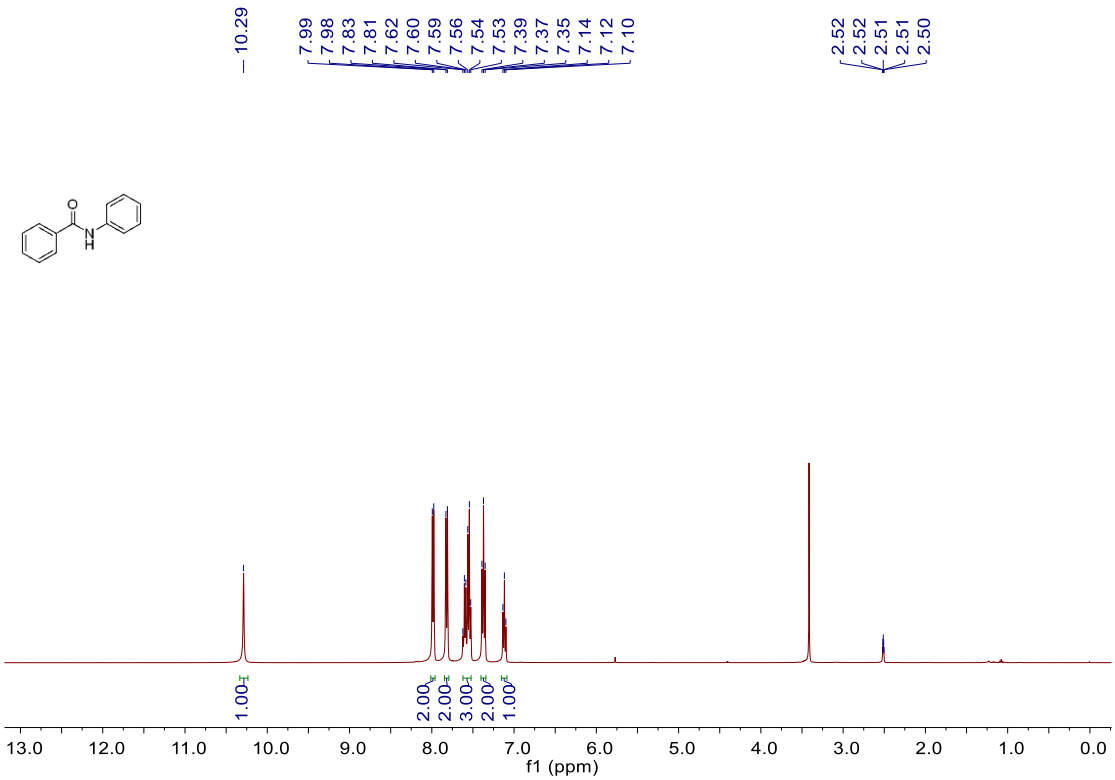
^{13}C{^{1}H} NMR of 71 in DMSO-d_6 (100 MHz)



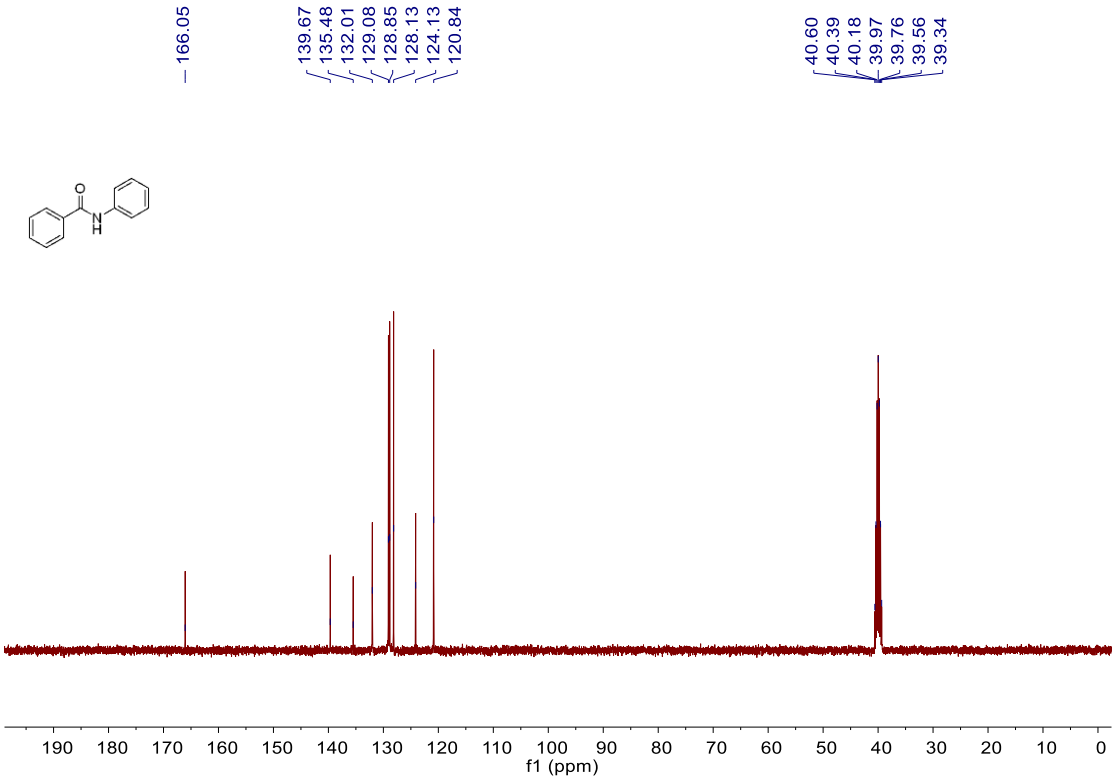
^{1}H NMR of 72 in $CDCl_3$ (400 MHz)



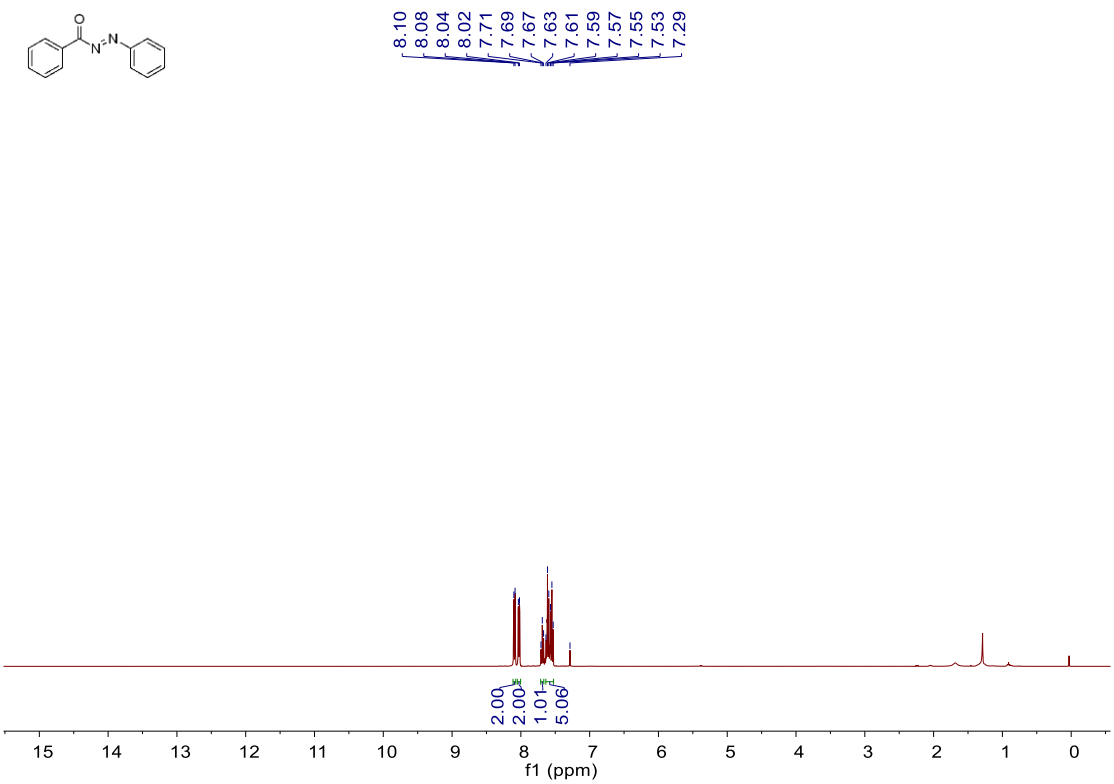
^{13}C{^{1}H} NMR of 72 in $CDCl_3$ (100 MHz)



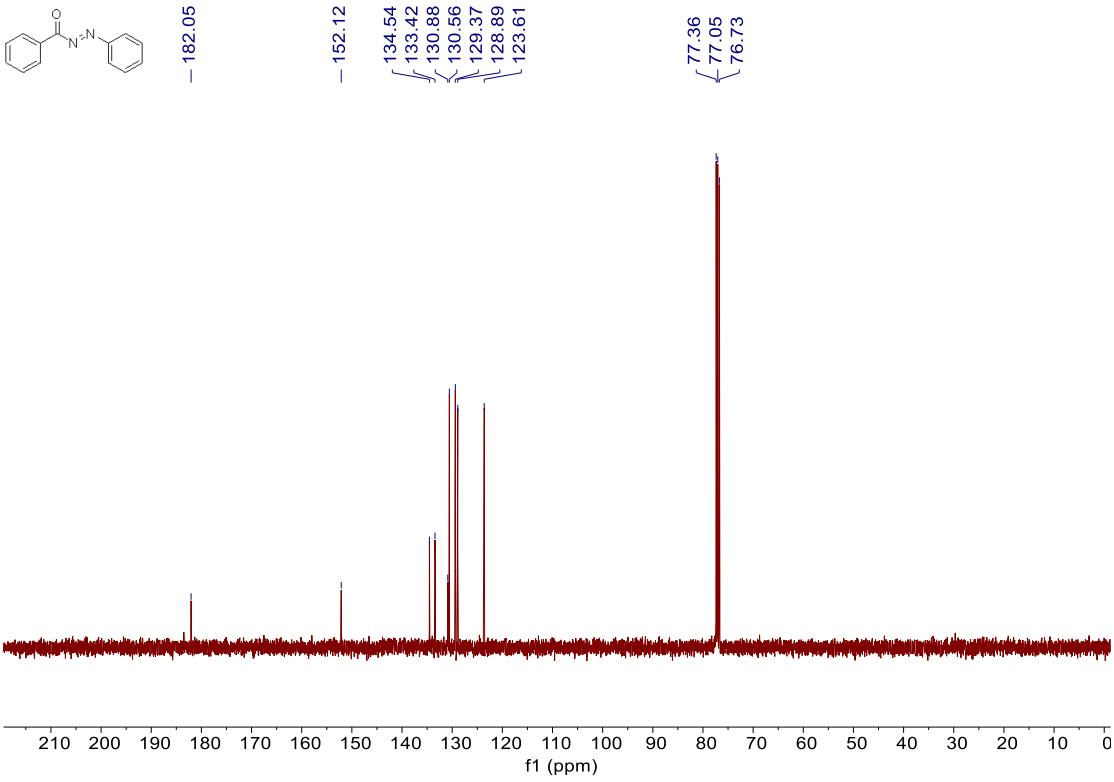
^{1}H NMR of 73 in DMSO-d_6 (400 MHz)



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



^{1}H NMR of 74 in $CDCl_3$ (400 MHz)



^{13}C{^{1}H} NMR of 74 in $CDCl_3$ (100 MHz)