Supplementary Information (SI) for Organic & Biomolecular Chemistry.

Ru(II)-catalyzed oxidative C-H spiroannulation of 4-arylquinazolin-2-ones with alkynes.

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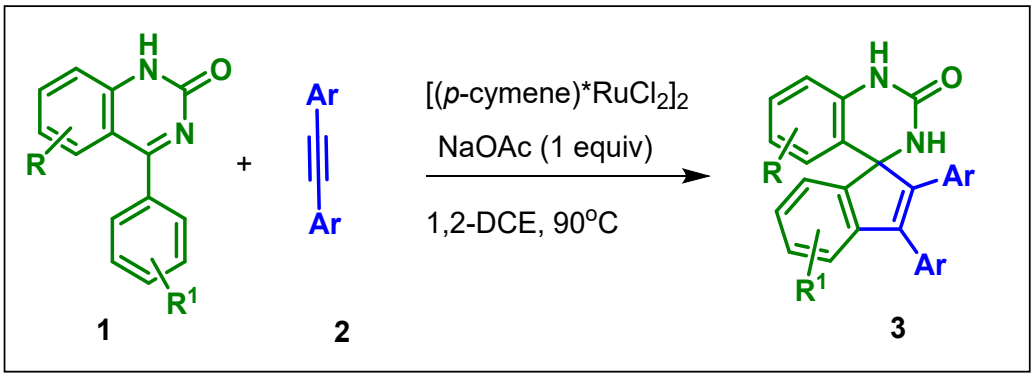

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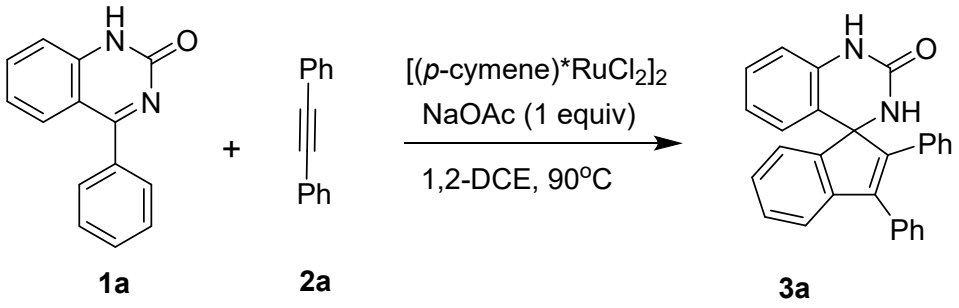
1. **Experimental**

All solvents were dried by a standard literature procedure. Crude products were purified by column chromatography on silica gel (60–120 or 100-200 mesh). Thin layer chromatography (**TLC**) plates were visualized by exposure to ultraviolet light at 254 nm, and by exposure to iodine vapours and/or by exposure to methanolic acidic solution of *p*-anisaldehyde followed by heating (<1 min) on a hot plate (~250°C). Organic solvents were concentrated on rotary evaporator at 35–40 °C. Melting points (**m.p**) were measured on Buchi B-540. **^{1}H** and **^{13}C NMR** (proton-decoupled) spectra were recorded in $CDCl_3$ on 300, 400 or 500 MHz NMR spectrometer. Chemical shifts (δ) were reported in parts per million (ppm) with respect to TMS as an internal standard. Coupling constants (*J*) are quoted in hertz (Hz). Mass spectra and **HRMS** were recorded on mass spectrometer by Electrospray ionization (ESI) or Atmospheric pressure chemical ionization (APCI) technique.

Typical Procedure:



1a **2a** **3a**

4-phenylquinazolin-2(1*H*)-one

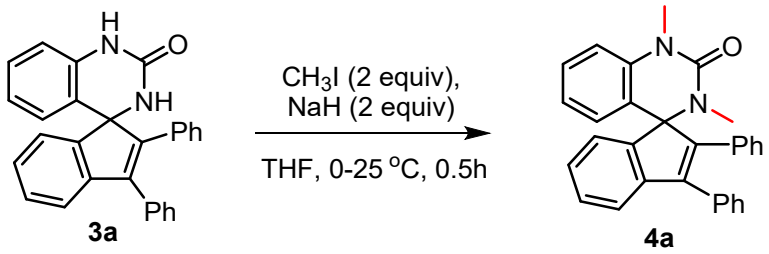
To an oven dried sealed tube equipped with a stir bar were charged with 4-phenylquinazolin-2(1*H*)-one (**1a**, 100 mg, 1.0 equiv), diphenylacetylene (**2a**, 80 mg, 1.0 equiv) in 3 mL of 1,2-DCE, followed by the addition of $[(p\text{-cymene})RuCl_2]_2$ (8.3 mg, 3 mol%) and NaOAc (60 mg, 1.0 equiv) as an additive at room temperature. The resulting mixture was stirred at 90 °C for 4-6h and then diluted with DCM. Then the mixture was diluted water and the aqueous layer was extracted with DCM (2×10 mL). The organic layer was evaporated and the residue was purified by column chromatography (R_f= 0.5) (SiO_2, EtOAc:*n*-Hexane, 4/6) to give the **3a** as a white solid in 94% (169 mg) yield.

Synthetic transformations:

***N*-Alkylation:**

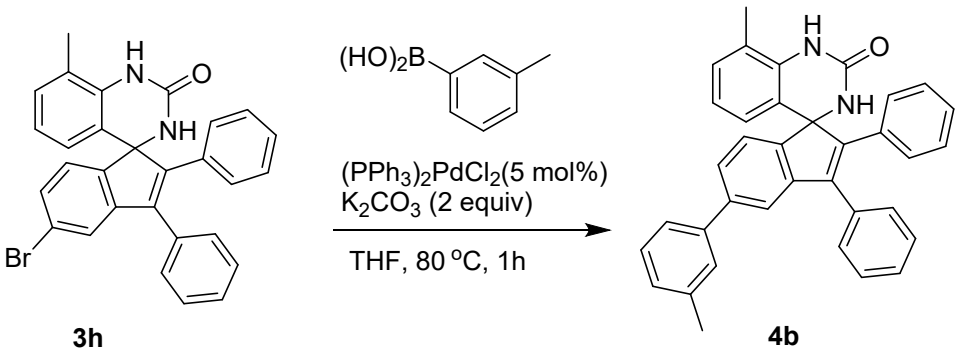
To an oven dried sealed tube was equipped with a stir bar and charged with **3a** (1 equiv), NaH (2 equiv) in THF at 0-5 °C and then CH_3I (2 equiv) was added *via* syringe while stirring. Then

the reaction mixture was stirred at room temp for 0.5 h and diluted with ethyl acetate. The mixture was filtered through celite bed and washed with ethyl acetate and then concentrated under reduced pressure. The residue was purified by column chromatography on silica gel (EtOAc/hexane) to afford the product in 95%yield.



Suzuki Coupling

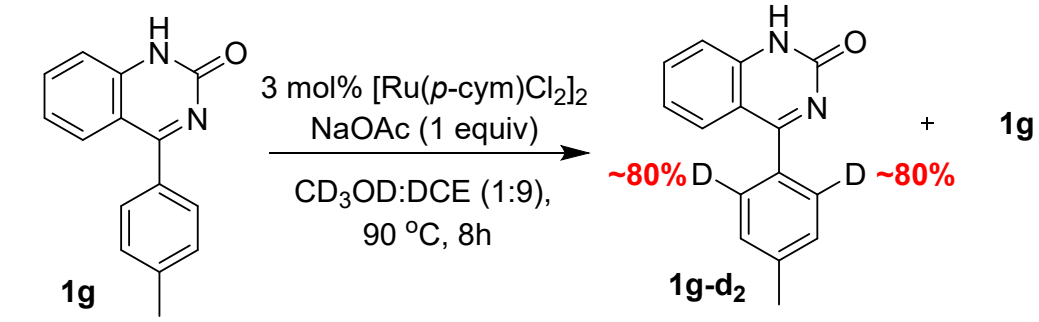
A sealed tube containing $PdCl_2(PPh_3)_2$ (10 mol%) and **3h** (1 equiv) and *m*-tolylboronic acid (1.1 equiv) K_2CO_3 (2 equiv) was purged with nitrogen three times and then THF (3ml) was added *via* syringe. The reaction mixture was stirred at 80 °C for 1h and then diluted with ethyl acetate and filtered through celite bed and washed with ethyl acetate. The solvent was concentrated under reduced pressure. The residue was purified by column chromatography on silica gel (EtOAc/hexane) to afford the product in 93% yield.



3. Mechanistic Studies:

(a) H/D exchange experiment:

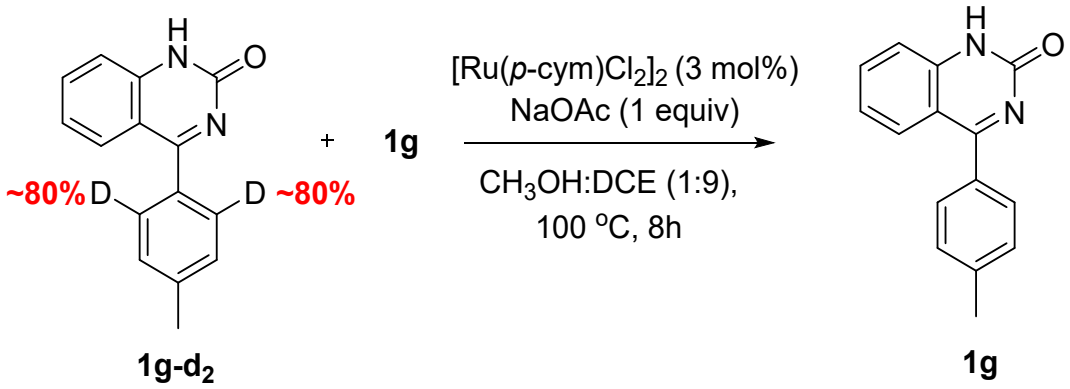
To an oven dried sealed tube equipped with a stir bar were charged with 4-(*p*-tolyl)quinazolin-2(1H)-one (**1g**, 1.0 equiv) and CD_3OD:DCE (1:9, 3 mL), followed by the addition of [Ru(*p*-cym)Cl_2]$_2$ catalyst (3 mol%) and NaOAc (1 equiv) as an additive at room temperature. The resulting mixture was stirred at 90 °C for 8h and then concentrated under reduced pressure. The residue was purified by column chromatography on silica gel using EtOAc/hexane as an eluent to afford the mixture of products **1g** and **1g-d_2.** The deuterium content of the product was 80%, which was determined by ^{1}H NMR.



4-(*p*-tolyl)quinazolin-2(1*H*)-one

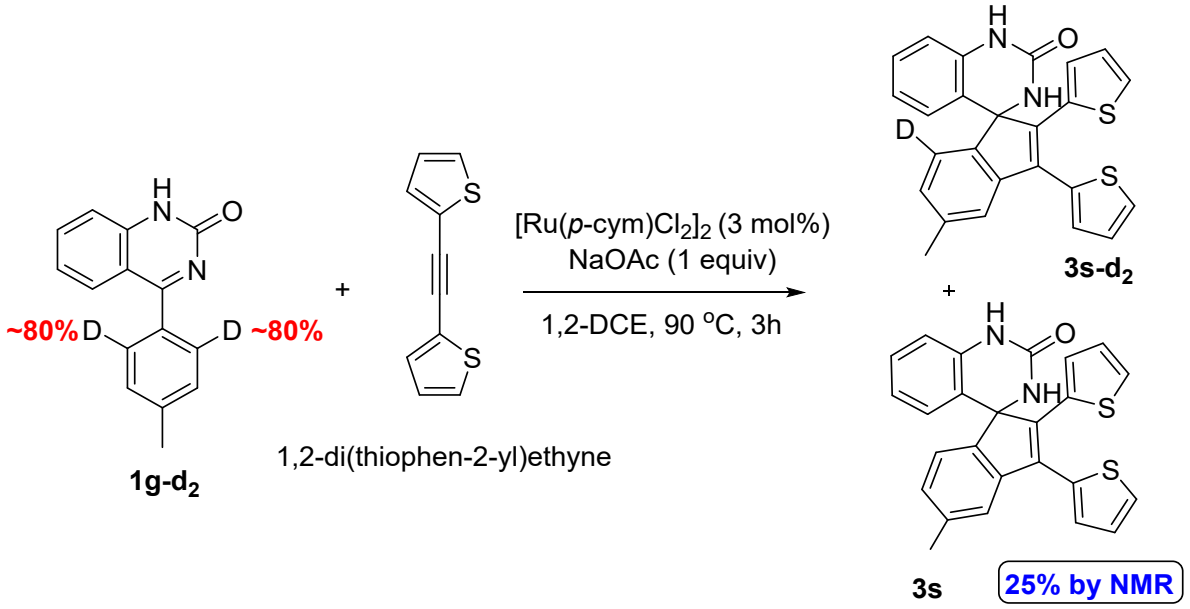
(b) D/H exchange experiment:

To an oven dried sealed tube equipped with a stir bar and charged with deuterated 4-(*p*-tolyl)quinazolin-2(1*H*)-one (**1g-d$_2$**, 1.0 equiv) and CH_3OH:DCE (1:9, 3 mL), followed by addition of [Ru(*p*-cym)Cl_2]$_2$ catalyst (3 mol%) and NaOAc as an additive (1 equiv) at room temperature. The resulting mixture was stirred at 90 °C for 8h and then concentrated under reduced pressure. The residue was purified by column chromatography on silica gel using EtOAc/hexane as an eluent to afford the product **1g**.

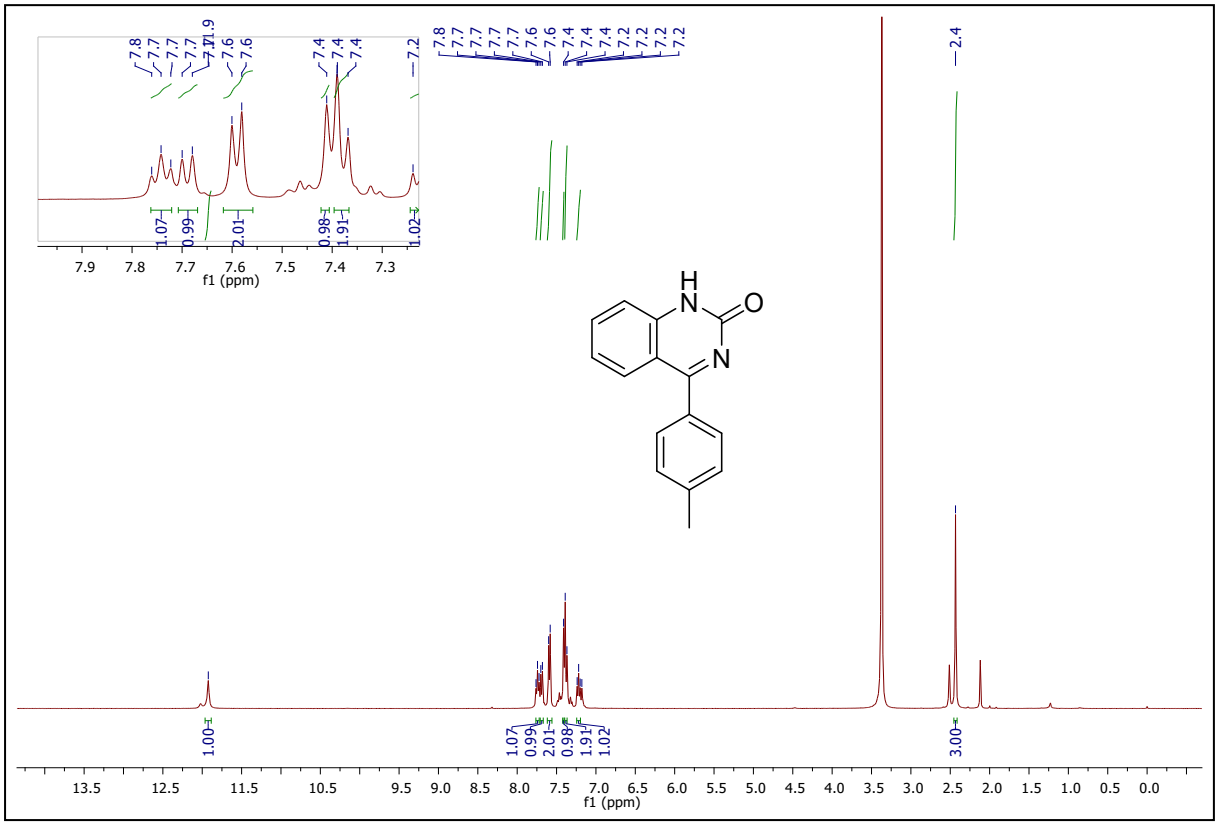


(c) H/D scrambling experiment with alkyne partner

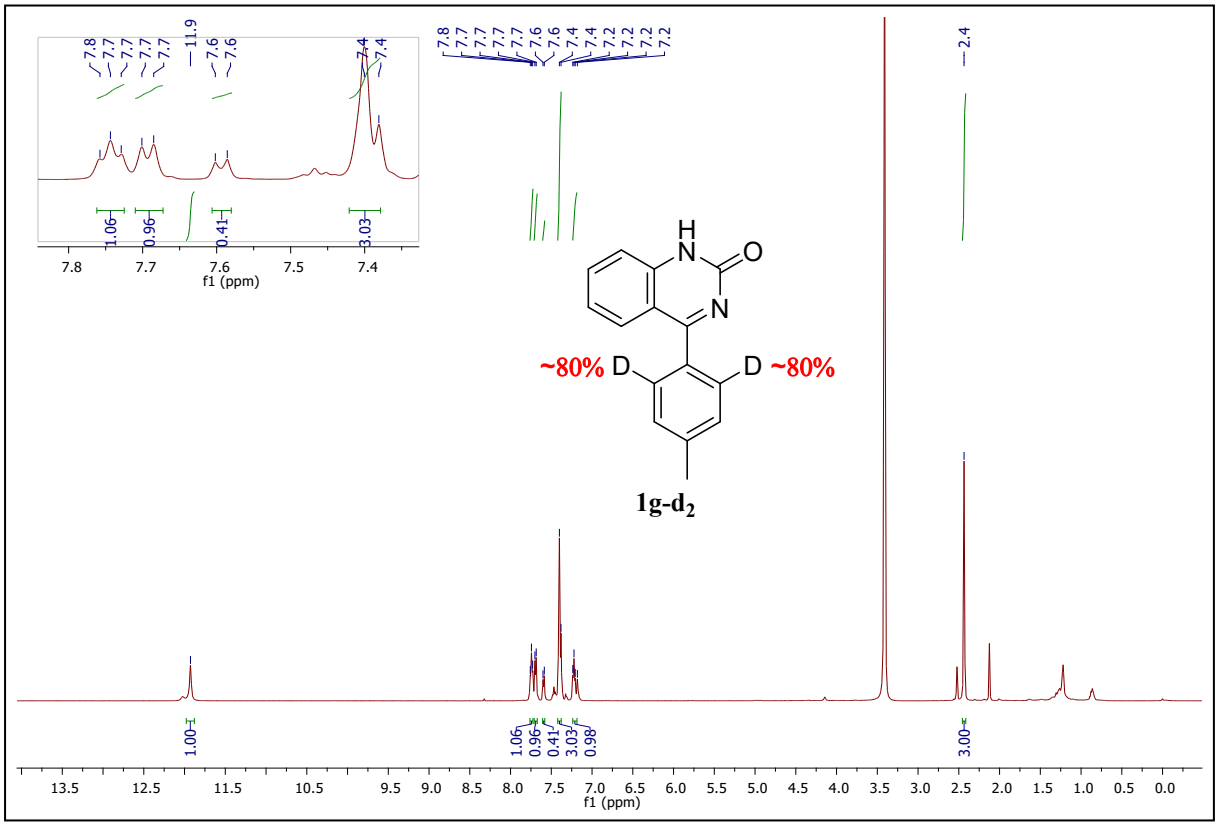
To an oven dried sealed tube equipped with a stir bar were charged with **1g-d$_2$** (1.0 equiv) and 1,2-di(thiophen-2-yl)ethyne (**2e**, 1 equiv) under the optimized conditions. The resulting mixture was stirred at 90 °C for 3h and then concentrated under reduced pressure. The residue was purified by column chromatography on silica gel using EtOAc/hexane to give the desired product, which was a mixture of **3s** and **3s-d$_2$**. The ratio of **3s** and **3s-d$_2$** was determined by ^{1}H NMR.



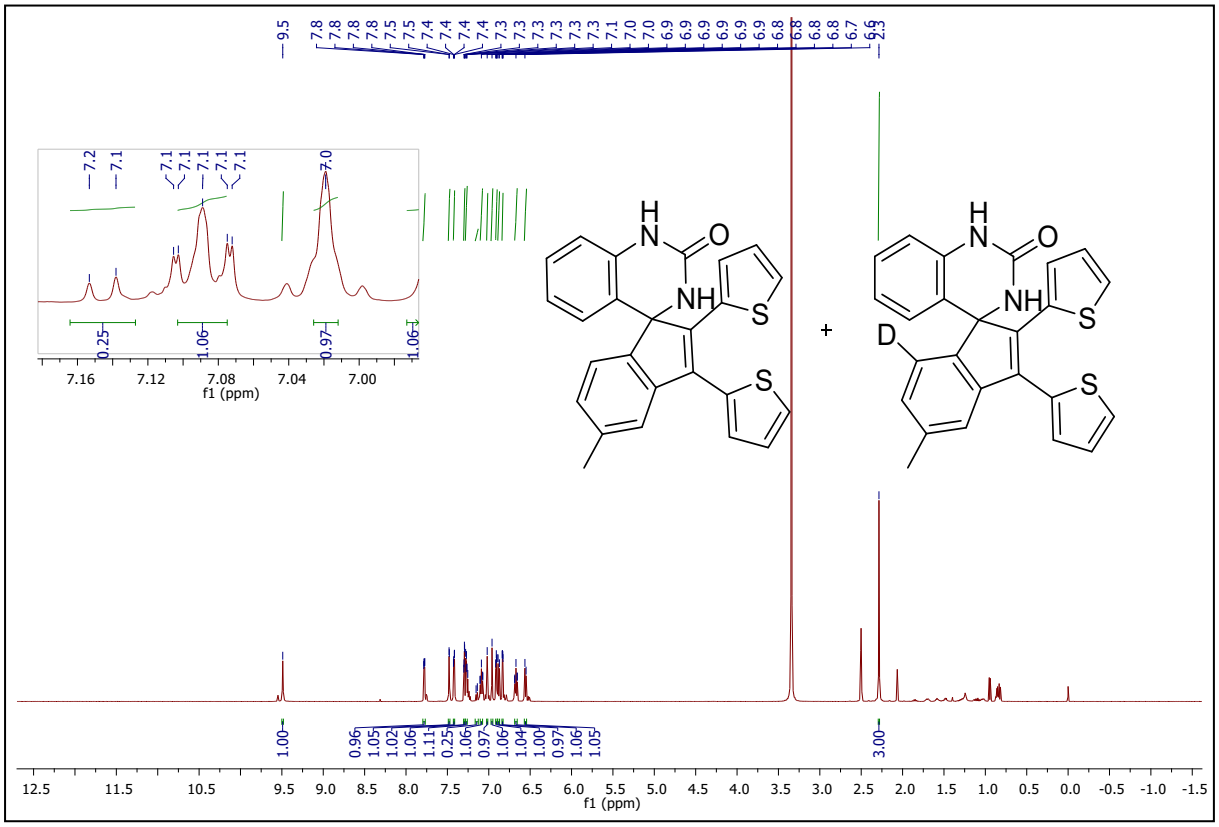
^{1}H NMR (400 MHz, DMSO-d_6) spectrum of 1g:



^{1}H NMR (500 MHz, DMSO-d_6) spectrum of 1g and 1g-d_2 mixture:

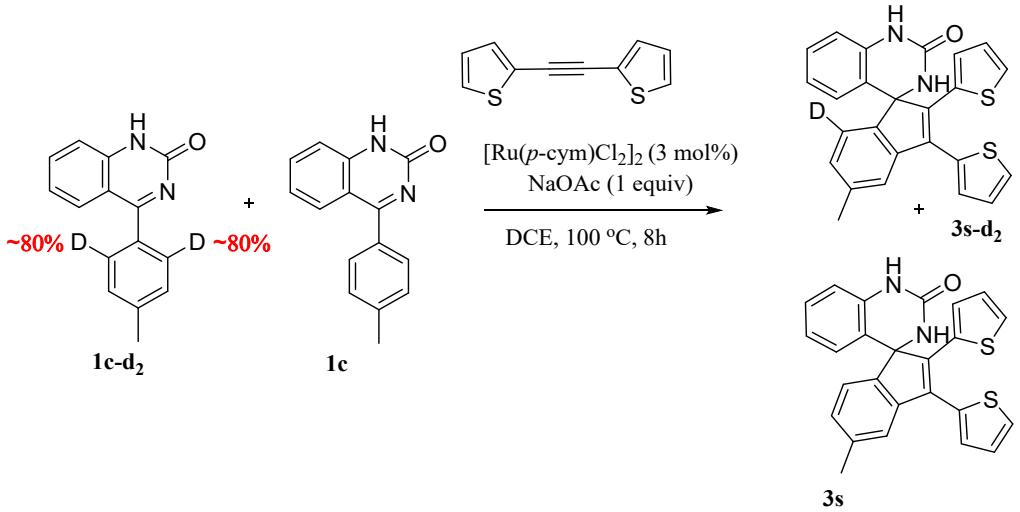


^{1}H NMR (400MHz, DMSO-d_6) spectrum of 3s and 3s-d_2 mixture:

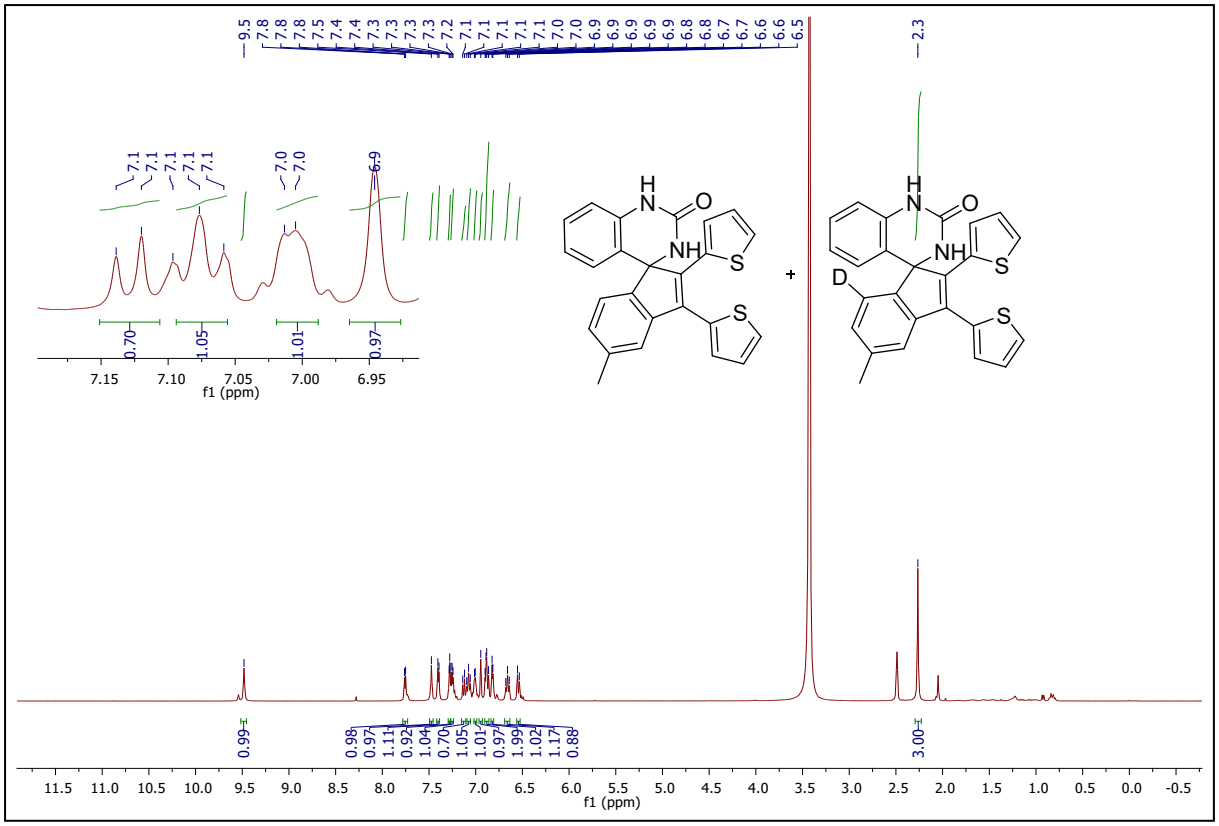


(d) Competitive KIE experiment between 1g and 1g-d_2:

To an oven dried sealed tube equipped with a stir bar were charged with 4-(*p*-tolyl)quinazolin-2(1*H*)-one (**1c**, 1.0 equiv), **1cd$_2$** (1.0 equiv) and 1,2-di(thiophen-2-yl)ethyne (**2e**, 1.0 equiv) under optimized conditions. The resulting mixture was stirred at 90 °C for 4h and then concentrated under reduced pressure. The residue was purified by column chromatography on silica gel using EtOAc/hexane to give the desired product, which is a mixture of **3s** and **3s-d$_2$**. The ratio of product formation of **3s** and **3s-d$_2$** was determined by ^{1}H NMR. The calculated K_H/K_D=2.33.



^{1}H NMR (500MHz, $CDCl_3$) spectrum of 3s and 3s-d_2 mixture:



Characterization data of products

2,3-Diphenyl-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3a:

White solid (0.169g, 94%) mp 272-274 ^{o}C, ^{1}H NMR (300 MHz, DMSO) δ 9.28 (s, 1H), 7.49 (s, 1H), 7.41 (s, 1H), 7.39 (s, 1H), 7.36 (s, 2H), 7.34 (s, 1H), 7.32 (d, *J* = 1.4 Hz, 1H), 7.30 (s, 1H), 7.25 (d, *J* = 6.9 Hz, 1H), 7.18 (d, *J* = 7.2 Hz, 1H), 7.13 (d, *J* = 2.2 Hz, 2H), 7.11 (s, 1H), 7.08 (d, *J* = 4.8 Hz, 1H), 7.03 (d, *J* = 1.9 Hz, 1H), 7.01 (d, *J* = 4.1 Hz, 1H), 6.81 (d, *J* = 7.9 Hz, 1H), 6.73 (s, 1H), 6.72 (s, 1H).^{13}C NMR (101 MHz, $CDCl_3$) δ 154.2, 151.6, 148.3, 141.9, 140.1, 137.1, 134.0, 133.8, 131.6, 129.5, 129.3, 128.9, 128.8, 128.6, 128.1, 128.0, 127.6, 127.5, 125.3, 123.9, 122.7, 121.2, 119.6, 115.0, 72.6. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{28}H_{21}N_2O$: 401.1654 found: 401.1647.

5-Methoxy-2,3-diphenyl-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3b:

Pale yellow solid (0.165g, 97%) 256-258 ^{o}C^{1}H NMR (500 MHz, DMSO) δ 9.23 (s, 1H), 7.44 (s, 1H), 7.42 – 7.40 (m, 1H), 7.38 (s, 1H), 7.36 – 7.32 (m, 3H), 7.22 (d, *J* = 8.2 Hz, 1H), 7.13 – 7.09 (m, 3H), 7.09 – 7.06 (m, 1H), 7.00 (d, *J* = 1.5 Hz, 1H), 6.99 (d, *J* = 2.2 Hz, 1H), 6.81 – 6.77 (m, 2H), 6.75 – 6.70 (m, 2H), 6.67 (d, *J* = 2.2 Hz, 1H), 3.73 (s, 3H). ^{13}C NMR (101 MHz, DMSO) δ 160.3, 153.4, 150.0, 145.8, 143.6, 139.4, 138.7, 135.0, 134.4, 129.9, 129.5, 129.1, 128.7, 128.3, 128.1, 127.7, 125.2, 124.4, 121.8, 119.8, 114.9, 112.2, 107.0, 71.8, 55.8. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{29}H_{22}N_2O_2$: 431.1760 found: 431.1754.

5-Methyl-2,3-diphenyl-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3c:

White solid (0.165g, 94%) mp 284-286 ^{o}C, ^{1}H NMR (500 MHz, DMSO) δ 9.24 (s, 1H), 7.44 (s, 1H), 7.39 (d, *J* = 7.1 Hz, 2H), 7.34 (t, *J* = 6.6 Hz, 3H), 7.18 (d, *J* = 7.5 Hz, 1H), 7.11 – 7.07 (m, 4H), 7.04 (d, *J* = 7.8 Hz, 1H), 7.01 (d, *J* = 1.4 Hz, 1H), 6.99 (d, *J* = 2.2 Hz, 1H), 6.97 (s, 1H), 6.79 (d, *J* = 7.9 Hz, 1H), 6.72 (d, *J* = 6.3 Hz, 2H), 2.30 (s, 3H).^{13}C NMR (126 MHz, DMSO) δ 153.5, 150.8, 148.8, 142.2, 139.9, 138.6, 138.2, 135.0, 134.6, 129.9, 129.5, 129.1, 128.8, 128.3, 128.1, 127.6, 125.2, 123.3, 121.8, 121.5, 119.7, 114.9, 72.1, 21.6. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{29}H_{23}N_2O$: 415.1810 found: 415.1791.

5-Fluoro-2,3-diphenyl-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3d:

White solid (0.153g, 88%) mp 266-268 ^{o}C, ^{1}H NMR (400 MHz, DMSO-D_6) δ 9.29 (s, 1H), 7.52 (s, 1H), 7.41 (d, *J* = 1.5 Hz, 1H), 7.39 (s, 1H), 7.36 (s, 1H), 7.34 – 7.30 (m, 2H), 7.15 – 7.12 (m, 3H), 7.11 – 7.09 (m, 1H), 7.09 – 7.04 (m, 1H), 7.03 – 6.98 (m, 2H), 6.91 (dd, *J* = 9.1, 2.4 Hz, 1H), 6.80 (d, *J* = 7.9 Hz, 1H), 6.77 – 6.71 (m, 2H). ^{13}C NMR (101 MHz, DMSO) δ 162.2 (d, J_{C-F} = 245.2 Hz), 161.9 (d, J_{C-F} = 245.0 Hz), 153.2, 152.6, 147.3, 141.6, 139.6, 138.0, 131.8 (d, J_{C-F} = 8.3 Hz), 131.7 (d, J_{C-F} = 8.3 Hz), 121.2 (d, J_{C-F} = 14.7 Hz), 116.2 (d, J_{C-F} = 21.4 Hz), 115.5 (d, J_{C-F} = 21.3 Hz)., 130.8, 130.3, 129.3, 129.1, 128.0, 125.2, 124.4, 123.7, 121.2 (d, *J* = 14.7 Hz), 116.8, 116.2 (d, J_{C-F} = 21.4 Hz), 115.5 (d, J_{C-F} = 21.3 Hz), 72.0. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{28}H_{20}N_2FO$: 419.1560 found: 419.1547.

5-Chloro-2,3-diphenyl-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3e:

White solid (0.154g, 91%) mp 252-254^{o}C,^{1}H NMR (300 MHz, DMSO) δ 9.45 (s, 1H), 7.64 (s, 1H), 7.44 – 7.40 (m, 1H), 7.39 (s, 1H), 7.37 (s, 2H), 7.35 (d, *J* = 3.5 Hz, 3H), 7.29 (d, *J* = 6.7 Hz, 1H), 7.21 (d, *J* = 7.5 Hz, 1H), 7.18 – 7.12 (m, 4H), 7.06 – 7.00 (m, 2H), 6.83 (d, *J* = 8.6 Hz, 1H), 6.61 (d, *J* = 2.1 Hz, 1H). ^{13}C NMR (101 MHz, DMSO) δ 153.1, 152.9, 148.2, 141.8, 140.2, 138.0, 134.6, 134.2, 129.8, 129.5, 129.2, 129.0, 128.4, 128.3, 127.9, 127.8, 125.1, 124.4, 123.6, 121.6, 121.2, 116.8, 72.1. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{28}H_{20}N_2OCl$: 435.1031found: 431.1020.

5-Fluoro-2,3-diphenyl-1'H-spiro[indene-1,4'-quinazolin]-2'(3'H)-one 3f:

Pale yellow solid (0.155g. 89%) 268-270 ^{o}C, ^{1}H NMR (300 MHz, DMSO) δ 9.27 (s, 1H), 7.50 (s, 1H), 7.42 – 7.38 (m, 1H), 7.36 (s, 1H), 7.35 – 7.32 (m, 2H), 7.31 (d, *J* = 1.2 Hz, 1H), 7.29 (d, *J* = 5.1 Hz, 1H), 7.14 – 7.11 (m, 2H), 7.11 – 7.07 (m, 2H), 7.06 – 7.02 (m, 1H), 6.88 (dd, *J* = 9.1, 2.4 Hz, 1H), 6.78 (d, *J* = 7.8 Hz, 1H), 6.73 – 6.68 (m, 2H). ^{13}C NMR (126 MHz, DMSO) δ 163.1 (d, J_{C-F} = 242.9 Hz), 153.3, 150.0, 149.0, 143.8 (d, J_{C-F} = 8.7 Hz), 138.8, 138.3, 133.3 (d, J_{C-F} = 6.1 Hz), 133.0, 132.4, 131.5, 131.3, 129.5, 129.2, 128.6, 125.2 (d, J_{C-F} = 8.8 Hz), 125.1, 122.0, 118.4, 115.1, 114.1 (d, J_{C-F} = 23.1 Hz), 108.3 (d, J_{C-F} = 24.1 Hz),71.9. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{28}H_{20}N_2FO$: 419.1560 found: 419.1547

8'-Bromo-2,3-diphenyl-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3g:

Grey solid, (0.134g,84%) mp 248-250 °C, ^{1}H NMR (500 MHz, DMSO) δ 8.12 (s, 1H), 7.94 (d, *J* = 1.5 Hz, 1H), 7.49 – 7.46 (m, 2H), 7.46 (d, *J* = 6.2 Hz, 1H), 7.42 (s, 2H), 7.41 (s, 2H), 7.40 – 7.38 (m, 1H), 7.32 (td, *J* = 7.4, 0.9 Hz, 1H), 7.25 (d, *J* = 7.5 Hz, 1H), 7.20 (d, *J* = 2.0 Hz, 2H), 7.19 (d, *J* = 1.8 Hz, 1H), 7.09 (d, *J* = 2.0 Hz, 1H), 7.08 – 7.07 (m, 1H), 6.85 (dd, *J* = 7.7, 0.8 Hz, 1H), 6.79 (t, *J* = 7.8 Hz, 1H). ^{13}C NMR (126 MHz, DMSO) δ 152.9, 152.9, 147.8, 141.8, 140.6, 136.2, 134.5, 134.3, 132.6, 129.9, 129.5, 129.1, 128.4, 128.3, 127.9, 127.8, 124.9, 123.5, 122.2, 121.2, 108.2, 72.4. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{28}H_{20}N_2OBr$: 479.0759 found: 479.0755.

5-Bromo-8'-methyl-2,3-diphenyl-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3h:

White solid (0.140g, 89%) mp 285-287 °C, ^{1}H NMR (500 MHz, DMSO) δ 9.40 (s, 1H), 7.57 (d, *J* = 1.3 Hz, 1H), 7.38 (d, *J* = 7.3 Hz, 2H), 7.35 – 7.31 (m, 4H), 7.26 – 7.24 (m, 1H), 7.21 (d, *J* = 7.5 Hz, 1H), 7.14 – 7.11 (m, 3H), 7.07 (d, *J* = 7.8 Hz, 1H), 6.99 (d, *J* = 2.5 Hz, 1H), 6.98 (d, *J* = 3.8 Hz, 2H), 6.74 (d, *J* = 8.6 Hz, 1H), 6.70 (d, *J* = 2.1 Hz, 1H), 2.30 (s, 3H). ^{13}C NMR (101 MHz, DMSO) δ 153.1, 150.2, 148.5, 142.1, 140.2, 138.6, 138.3, 134.6, 134.3, 131.7, 129.7, 129.5, 129.2, 128.4, 128.3, 127.9, 127.2, 123.4, 122.3, 121.8, 117.1, 112.7, 71.7, 21.6. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{29}H_{22}N_2OBr$: 493.0916 found: 493.0897.

2,3-Bis(4-methoxyphenyl)-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3i:

Pale yellow solid (0.199g, 96%) mp 199-201 °C, ^{1}H NMR (400 MHz, DMSO) δ 10.09 (s, 1H), 7.77 (s, 1H), 7.23 (s, 3H), 7.15 (s, 2H), 6.93 (d, *J* = 33.3 Hz, 4H), 6.82 (s, 2H), 6.64 (s, 3H), 3.76 (s, 3H), 3.62 (s, 3H). ^{13}C NMR (101 MHz, DMSO) δ 159.1, 158.7, 153.6, 153.5, 147.3, 142.2, 138.8, 138.7, 133.1, 131.1, 130.8, 130.3, 128.7, 127.2, 126.7, 125.1, 123.3, 121.8, 120.7, 119.8, 114.9, 114.6, 113.7, 72.1, 55.5, 55.3. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{30}H_{24}N_2O_3$: 461.1865 found: 461.1845.

2,3-Di-m-tolyl-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3j:

Pale yellow solid (0.164g, 85%) mp214-216 °C, ^{1}H NMR (400 MHz, DMSO) δ 9.28 (s, 1H), 7.46 (s, 1H), 7.31 (d, *J* = 6.3 Hz, 2H), 7.28 (d, *J* = 7.3 Hz, 1H), 7.25 (s, *J* = 10.1 Hz, 1H), 7.21 (s, 1H), 7.10 (s, 1H), 7.07 (d, *J* = 6.8 Hz, 1H), 6.98 (d, *J* = 7.7 Hz, 1H), 6.93 (d, *J* = 7.8 Hz, 1H), 6.85 (s, 1H), 6.80 (d, *J* = 7.6 Hz, 2H), 6.71 (d, *J* = 7.9 Hz, 2H).^{13}C NMR (101 MHz, DMSO) δ 153.5, 148.5, 142.1, 139.7, 138.8, 138.1, 136.7, 134.8, 134.5, 130.3, 129.8, 129.0, 128.9, 128.8, 128.3, 127.9, 127.4, 127.2, 126.7, 125.1, 123.5, 121.8, 120.9, 119.5, 114.9, 72.4, 21.6, 21.5. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{30}H_{25}N_2O$: 429.1967 found: 429.1950.

2,3-Bis(3-(trifluoromethyl)phenyl)-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3k:

White solid (0.207g, 86%) mp278-280 °C, ^{1}H NMR (400 MHz, DMSO) δ 9.49 (s, 1H), 7.75 (d, *J* = 7.5 Hz, 1H), 7.72 (s, 1H), 7.69 (d, *J* = 7.7 Hz, 1H), 7.65 (s, 1H), 7.62 (s, 1H), 7.49 (d, *J* = 7.1 Hz, 1H), 7.40 (d, *J* = 7.3 Hz, 1H), 7.37 (s, 1H), 7.35 (s, 3H), 7.30 (d, *J* = 7.2 Hz, 1H), 7.21 (d, *J* = 7.1 Hz, 1H), 7.12 (t, *J* = 7.3 Hz, 1H), 6.89 (d, *J* = 8.0 Hz, 1H), 6.85 (d, *J* = 7.4 Hz,

1H), 6.75 (t, *J* = 7.2 Hz, 1H).^{13}C NMR (101 MHz, DMSO) δ 153.4, 153.2, 147.8, 141.0, 140.2, 138.8, 135.5, 135.0, 133.5, 130.5, 130.2, 129.8, 129.5, 129.2, 129.2, 128.9, 128.3, 126.3 (q, J_{C-F} = 32.3 Hz), 126.3 (q, J_{C-F} = 32.4 Hz), 125.4, 124.6, 124.4 (q, J_{C-F} = 272.4 Hz), 124.4 (q, J_{C-F} = 272.4 Hz).122.1, 121.1, 118.5, 115.1, 72.4. ^{19}F NMR (376 MHz, DMSO) δ -61.5, -61.7. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{30}H_{19}N_2OF_6$: 537.1402 found: 537.1418.

2,3-Bis(4-fluorophenyl)-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3l:

Pale yellow solid (0.165g, 84%) 295-297 °C, ^{1}H NMR (400 MHz, DMSO) δ 9.29 (s, 1H), 7.52 (d, *J* = 1.6 Hz, 1H), 7.43 – 7.39 (m, 2H), 7.37 – 7.31 (m, 4H), 7.14 (d, *J* = 1.6 Hz, 1H), 7.12 (d, *J* = 3.4 Hz, 1H), 7.11 – 7.09 (m, 1H), 7.09 – 7.04 (m, 1H), 7.03 – 6.98 (m, 2H), 6.91 (dd, *J* = 9.1, 2.4 Hz, 1H), 6.80 (d, *J* = 7.9 Hz, 1H), 6.77 – 6.71 (m, 2H). ^{13}C NMR (126 MHz, DMSO) δ 162.1 (d, *J* = 245.0 Hz), 161.8 (d, *J* = 245.0 Hz), 153.5, 153.3, 147.7, 141.6, 139.2, 138.8, 131.9 (d, *J* = 8.0 Hz), 131.6 (d, *J* = 8.1 Hz), 131.1 (d, *J* = 2.7 Hz), 130.5 (d, *J* = 2.4 Hz), 129.0, 128.9, 127.7, 125.1, 123.6, 121.9, 120.9, 119.1, 116.2 (d, *J* = 21.4 Hz), 115.3 (d, *J* = 21.2 Hz), 115.0, 72.3. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{28}H_{19}N_2F_2O$: 437.1465 found: 437.1453.

2,3-Bis(4-chlorophenyl)-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3m:

White solid (0.182g, 86%) mp 214-216 °C, ^{1}H NMR (400 MHz, DMSO) δ 9.34 (s, 1H), 7.51 (s, 1H), 7.50 (s, 1H), 7.48 (s, 1H), 7.36 (d, *J* = 8.4 Hz, 2H), 7.33 (d, *J* = 6.1 Hz, 2H), 7.27 (d, *J* = 7.7 Hz, 1H), 7.24 (d, *J* = 8.3 Hz, 2H), 7.19 (d, *J* = 7.4 Hz, 1H), 7.12 – 7.08 (m, 1H), 7.01 (d, *J* = 8.3 Hz, 2H), 6.82 (d, *J* = 7.9 Hz, 1H), 6.71 (t, *J* = 6.9 Hz, 2H). ^{13}C NMR (101 MHz, DMSO) δ 153.5, 153.3, 147.7, 141.3, 139.5, 138.8, 133.6, 133.1, 133.0, 132.7, 131.6, 131.4, 129.4, 129.04, 129.0, 128.5, 127.9, 125.1, 123.6, 122.0, 121.0, 118.9, 115.1, 72.4. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{28}H_{19}N_2OCl_2$: 469.0874 found: 469.0852.

2,3-Bis(4-bromophenyl)-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3n:

White solid (0.207g, 83%) mp 232-234 °C, ^{1}H NMR (400 MHz, DMSO) δ 9.31 (s, 1H), 7.61 (s, 1H), 7.59 (s, 1H), 7.49 (s, 1H), 7.36 (s, 1H), 7.34 (s, 1H), 7.31 (d, *J* = 3.8 Hz, 1H), 7.30 – 7.29 (m, 2H), 7.27 (s, 1H), 7.25 (d, *J* = 6.9 Hz, 1H), 7.19 – 7.15 (m, 1H), 7.10 – 7.05 (m, 1H), 6.94 (s, 1H), 6.92 (s, 1H), 6.80 (d, *J* = 7.9 Hz, 1H), 6.73 – 6.66 (m, 2H). ^{13}C NMR (126 MHz, DMSO) δ 153.5, 153.3, 147.8, 141.2, 139.5, 138.8, 134.0, 133.4, 132.3, 131.8, 131.7, 131.4, 129.1, 129.0, 127.9, 125.1, 123.7, 122.0, 121.8, 121.5, 121.0, 118.9, 115.1, 72.4. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{28}H_{19}N_2Br_2O$: 556.9864 found: 556.9854.

5-Chloro-2,3-bis(4-fluorophenyl)-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3o:

Pale yellow solid (0.156g, 85%) mp 273-275 °C, ^{1}H NMR (400 MHz, DMSO) δ 9.47 (s, 1H), 7.62 (s, 1H), 7.41 (d, *J* = 5.8 Hz, 1H), 7.39 (d, *J* = 5.5 Hz, 1H), 7.37 (s, 1H), 7.35 (s, 1H), 7.30 (d, *J* = 6.9 Hz, 1H), 7.25 (t, *J* = 8.8 Hz, 2H), 7.21 (d, *J* = 7.3 Hz, 1H), 7.17 (dd, *J* = 8.6, 2.2 Hz, 1H), 7.05 (s, 2H), 7.04 (s, 2H), 6.83 (d, *J* = 8.6 Hz, 1H), 6.62 (d, *J* = 2.0 Hz, 1H).

^{13}C NMR (101 MHz, DMSO) δ 162.2 (d, J_{C-F} = 245.2 Hz), 161.9 (d, J_{C-F} = 245.0 Hz), 153.2, 152.6, 147.3, 141.6, 139.6, 138.0, δ 131.8 (d, J_{C-F} = 8.3 Hz), 131.7 (d, J_{C-F} = 8.3 Hz), 130.8 (d, J_{C-F} = 2.4 Hz), 130.3 (d, J_{C-F} = 2.4 Hz), 129.3, 129.1, 128.0, 125.2, 124.4, 123.7, 121.3, 121.2, 116.8, 116.2 (d, J_{C-F} = 21.4 Hz), 115.5 (d, J_{C-F} = 21.3 Hz), 72.0. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{28}H_{18}N_2ClOF_2$: 471.1496 found: 471.1509.

2,3-Bis(4-chlorophenyl)-8'-fluoro-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3p:

White solid (0.176, 87%) mp 273-275 °C, ^{1}H NMR (400 MHz, DMSO) δ 9.32 (s, 1H), 7.51 (s, 1H), 7.48 (s, 1H), 7.46 (s, 1H), 7.35 (s, 1H), 7.33 (s, 1H), 7.32 – 7.30 (m, 1H), 7.24 (s, 1H), 7.22 (s, 1H), 7.11 – 7.03 (m, 2H), 6.98 (s, 1H), 6.96 (s, 1H), 6.94 (dd, *J* = 9.0, 2.3 Hz, 1H), 6.79 (d, *J* = 7.8 Hz, 1H), 6.75 – 6.68 (m, 2H). ^{13}C NMR (126 MHz, DMSO) δ 163.1 (d, J_{C-F} = 242.9 Hz), 153.3, 150.0, 149.0, 143.8 (d, J_{C-F} = 8.7 Hz), 138.8, 138.3, 133.3 (d, J_{C-F} = 6.1 Hz), 133.0, 132.4, 131.5, 131.3, 129.5, 129.2, 128.6, 125.2 (d, J_{C-F} = 8.8 Hz), 125.1, 122.0, 118.4, 115.1, 114.1 (d, J_{C-F} = 23.1 Hz), 108.3 (d, J_{C-F} = 24.1 Hz), 71.8. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{28}H_{18}N_2Cl_2O$: 487.0780 found: 487.0763.

Diethyl 2'-oxo-2',3'-dihydro-1'*H*-spiro[indene-1,4'-quinazoline]-2,3-dicarboxylate 3q:

White solid (0.148g, 84%) mp 230-232 °C, ^{1}H NMR (500 MHz, DMSO) δ 9.43 (s, 1H), 7.37 – 7.36 (m, 1H), 7.36 – 7.34 (m, 2H), 7.26 – 7.24 (m, 1H), 7.03 (t, *J* = 7.7 Hz, 1H), 6.77 (d, *J* = 8.0 Hz, 1H), 6.58 (t, *J* = 7.5 Hz, 1H), 6.15 (d, *J* = 7.7 Hz, 1H), 4.31 (q, *J* = 7.1 Hz, 2H), 4.01 – 3.94 (m, 2H), 1.25 (t, *J* = 7.1 Hz, 3H), 1.02 (t, *J* = 7.1 Hz, 3H). ^{13}C NMR (101 MHz, DMSO) δ 164.5, 162.5, 153.2, 153.1, 143.6, 140.3, 139.3, 136.8, 130.7, 129.7, 129.1, 124.8, 124.5, 123.0, 121.5, 117.3, 115.0, 70.7, 62.0, 61.1, 14.6, 14.1. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{22}H_{21}N_2O_5$: 393.1450 found: 393.1439.

5-Chloro-2,3-dipropyl-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3r:

White solid (0.119g, 83%) mp193-195 °C, ^{1}H NMR (500 MHz, DMSO) δ 9.50 (s, 1H), 7.28 (d, *J* = 9.9 Hz, 1H), 7.27 (s, 1H), 7.20 (s, 1H), 7.16 (d, *J* = 7.3 Hz, 1H), 7.14 – 7.09 (m, 2H), 6.85 (d, *J* = 8.6 Hz, 1H), 6.16 (d, *J* = 2.1 Hz, 1H), 2.47 – 2.42 (m, 2H), 2.25 (m, 1H), 2.04 (m, 1H), 1.59 (m, 2H), 1.35 – 1.28 (m, 1H), 1.26 – 1.15 (m, 1H), 0.96 (t, *J* = 7.3 Hz, 3H), 0.80 (t, *J* = 7.3 Hz, 3H). ^{13}C NMR (126 MHz, DMSO) δ 153.6, 152.3, 149.2, 143.2, 138.1, 137.5, 128.9, 128.6, 126.4, 124.8, 124.3, 123.1, 122.6, 119.6, 116.5, 71.3, 28.1, 27.2, 22.5, 21.9, 15.1, 14.5. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{22}H_{23}N_2ClO$: 367.1534 found: 367.1525.

2,3-Di(thiophen-2-yl)-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3s:

Pale yellow solid (0.169g, 91%) mp 274-276 °C, ^{1}H NMR (400 MHz, DMSO) δ 9.52 (s, 1H), 7.78 (d, *J* = 4.9 Hz, 1H), 7.53 (s, 1H), 7.43 (d, *J* = 4.9 Hz, 1H), 7.31 (t, *J* = 6.8 Hz, 2H), 7.27 (t, *J* = 4.2 Hz, 2H), 7.23 (d, *J* = 7.3 Hz, 1H), 7.18 (d, *J* = 7.4 Hz, 1H), 7.10 (t, *J* = 7.6 Hz, 1H), 6.93 – 6.91 (m, 1H), 6.90 (d, *J* = 7.8 Hz, 1H), 6.85 (d, *J* = 3.2 Hz, 1H), 6.68 (t, *J* = 7.4 Hz, 1H), 6.58 (d, *J* = 7.5 Hz, 1H). ^{13}C NMR (101 MHz, DMSO) δ 153.1, 153.0, 144.0, 141.6, 138.5,

135.6, 134.4, 132.4, 129.1, 128.9, 128.5, 128.3, 128.1, 127.8, 127.1, 124.9, 123.3, 122.0, 120.9, 119.5, 115.1, 71.8. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{24}H_{17}N_2OS_2$: 413.0782 found: 413.0775.

5-Methoxy-2,3-di(thiophen-2-yl)-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3t:

Pale yellow solid (0.165g, 94%) mp 152-154 ^oC, ^1H NMR (400 MHz, DMSO) δ 9.52 (s, 1H), 7.79 (dd, *J* = 5.0, 0.9 Hz, 1H), 7.66 – 7.61 (m, 1H), 7.58 – 7.53 (m, 1H), 7.50 (s, 1H), 7.44 (d, *J* = 5.8 Hz, 1H), 7.32 (d, *J* = 2.6 Hz, 1H), 7.27 (dd, *J* = 4.9, 3.6 Hz, 1H), 7.18 (d, *J* = 8.2 Hz, 1H), 7.09 (t, *J* = 7.6 Hz, 1H), 6.93 – 6.90 (m, 1H), 6.85 (d, *J* = 3.4 Hz, 1H), 6.78 (dd, *J* = 8.2, 2.2 Hz, 1H), 6.69 – 6.65 (m, 1H), 6.58 (d, *J* = 7.6 Hz, 1H), 3.72 (s, 3H). ^{13}C NMR (101 MHz, DMSO) δ 160.5, 153.1, 145.4, 143.2, 138.4, 135.6, 134.3, 131.9, 129.3, 129.2, 129.0, 128.6, 128.4, 128.2, 127.1, 125.0, 124.2, 121.9, 119.9, 115.0, 112.7, 106.7, 71.3, 55.8. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{25}H_{19}N_2O_2S_2$: 443.0888 found: 443.0869.

5-Methyl-2,3-di(thiophen-2-yl)-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3u:

Pale yellow solid (0.166g, 92%) mp 132-134 ^oC, ^1H NMR (400 MHz, DMSO) δ 9.47 (s, 1H), 7.78 – 7.75 (m, 1H), 7.47 (s, 1H), 7.40 (d, *J* = 4.0 Hz, 1H), 7.28 (d, *J* = 2.3 Hz, 1H), 7.27 – 7.24 (m, 1H), 7.13 (d, *J* = 7.5 Hz, 1H), 7.08 (t, *J* = 7.6 Hz, 1H), 7.01 (d, *J* = 7.1 Hz, 1H), 6.95 (s, 1H), 6.87 (d, *J* = 8.4 Hz, 1H), 6.82 (d, *J* = 3.1 Hz, 1H), 6.66 (t, *J* = 7.2 Hz, 1H), 6.54 (d, *J* = 7.5 Hz, 1H), 2.27 (s, 3H). ^{13}C NMR (126 MHz, DMSO) δ 153.1, 150.4, 144.4, 141.8, 138.5, 138.4, 135.7, 134.5, 132.4, 129.0, 128.9, 128.5, 128.5, 128.3, 128.2, 128.0, 127.8, 127.1, 124.9, 123.0, 122.0, 121.5, 119.8, 115.1, 71.5, 21.5. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{25}H_{19}N_2OS_2$: 427.0939 found: 427.0927.

3-(4-Chlorophenyl)-5-methyl-2-phenyl-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3v:

White solid (0.105g, 55%) mp 263-265 ^oC, ^1H NMR (300 MHz, DMSO) δ 8.54 (s, 1H), 7.58 (s, 1H), 7.44 (d, *J* = 6.5 Hz, 1H), 7.42 – 7.38 (m, 2H), 7.36 (d, *J* = 1.7 Hz, 1H), 7.34 (d, *J* = 1.5 Hz, 1H), 7.32 (d, *J* = 4.1 Hz, 1H), 7.31 – 7.29 (m, 1H), 7.25 (d, *J* = 6.5 Hz, 1H), 7.22 (s, 1H), 7.18 (d, *J* = 9.2 Hz, 2H), 7.02 (s, 1H), 7.00 (s, 1H), 6.96 (d, *J* = 6.6 Hz, 1H), 6.66 (t, *J* = 7.5 Hz, 1H), 6.59 (d, *J* = 7.1 Hz, 1H), 2.20 (s, 3H). ^{13}C NMR (126 MHz, DMSO) δ 153.7, 153.6, 149.0, 141.4, 139.0, 136.9, 134.7, 133.4, 132.9, 131.4, 130.4, 129.9, 129.3, 128.8, 128.3, 127.8, 127.7, 123.5, 123.3, 122.9, 121.6, 120.8, 119.3, 72.4, 17.9. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{29}H_{22}N_2OCl$: 449.1421 found: 449.1409

2-(4-Chlorophenyl)-5-methyl-3-phenyl-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3v':

White solid (0.070g, 37%) mp 264-266 ^1H NMR (300 MHz, DMSO) δ 8.52 (s, 1H), 7.58 (s, 1H), 7.48 (s, 1H), 7.45 (s, 1H), 7.37 (s, 1H), 7.34 (s, 1H), 7.31 (s, 1H), 7.29 (s, 1H), 7.24 (d, *J* = 6.9 Hz, 1H), 7.18 (s, 1H), 7.15 – 7.12 (m, 3H), 7.04 (d, *J* = 1.8 Hz, 1H), 7.03 – 7.00 (m, 1H), 6.95 (d, *J* = 6.1 Hz, 1H), 6.66 (d, *J* = 7.7 Hz, 1H), 6.62 (s, 1H), 2.19 (s, 3H). ^{13}C NMR (75

MHz, DMSO) δ 153.8, 153.6, 147.0, 141.6, 140.8, 136.9, 134.2, 133.9, 132.5, 131.6, 130.5, 129.5, 129.2, 128.8, 128.5, 128.3, 127.7, 123.5, 123.4, 122.7, 121.7, 121.1, 119.3, 72.3, 17.9. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{29}H_{22}N_2OCl$: 449.1421 found: 449.1409.

3-Phenyl-2-(phenylethynyl)-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3w:

Grey solid, (0.160g, 84%) mp ^{1}H NMR (500 MHz, DMSO) δ 9.59 (s, 1H), 7.78 (d, *J* = 1.2 Hz, 1H), 7.77 (s, 1H), 7.58 (t, *J* = 7.6 Hz, 2H), 7.50 (d, *J* = 7.5 Hz, 1H), 7.48 (d, *J* = 1.9 Hz, 1H), 7.47 (d, *J* = 7.9 Hz, 1H), 7.43 – 7.39 (m, 1H), 7.36 (dd, *J* = 3.9, 2.5 Hz, 5H), 7.26 (d, *J* = 2.7 Hz, 1H), 7.26 – 7.25 (m, 1H), 7.14 – 7.10 (m, 1H), 6.92 (d, *J* = 8.1 Hz, 1H), 6.72 – 6.68 (m, 1H), 6.40 (d, *J* = 7.3 Hz, 1H). ^{13}C NMR (126 MHz, DMSO) δ 154.1, 151.4, 143.9, 141.0, 139.3, 133.4, 133.2, 131.5, 129.5, 129.4, 129.3, 129.2, 129.1, 128.9, 128.6, 124.8, 124.6, 122.7, 121.8, 121.8, 119.8, 114.8, 97.9, 86.0, 71.9. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{30}H_{21}N_2O$: 425.1654 found: 425.1652.

5-Methoxy-3-phenyl-2-(phenylethynyl)-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3x:

Grey solid,(0.180g, 88%) mp ^{1}H NMR (400 MHz, DMSO) δ 9.59 (s, 1H), 7.78 (d, *J* = 7.1 Hz, 2H), 7.62 – 7.57 (m, 2H), 7.51 (d, *J* = 7.0 Hz, 1H), 7.44 (s, 1H), 7.37 (s, 3H), 7.27 (s, 3H), 7.16 – 7.11 (m, 1H), 6.95 (d, *J* = 8.3 Hz, 2H), 6.92 (s, 1H), 6.72 (t, *J* = 7.0 Hz, 1H), 6.43 (d, *J* = 7.4 Hz, 1H), 3.80 (s, 3H). ^{13}C NMR (126 MHz, DMSO) δ 160.6, 154.2, 143.4, 143.3, 142.7, 139.2, 134.7, 133.1, 131.5, 129.5, 129.4, 129.3, 129.2, 129.0, 128.8, 125.4, 124.9, 122.7, 121.7, 120.2, 114.7, 113.5, 107.6, 98.0, 86.0, 71.3, 55.9. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{31}H_{23}N_2O_2$: 455.1750 found: 455.1764.

1'-Methyl-2,3-diphenyl-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'*H*)-one 3y:

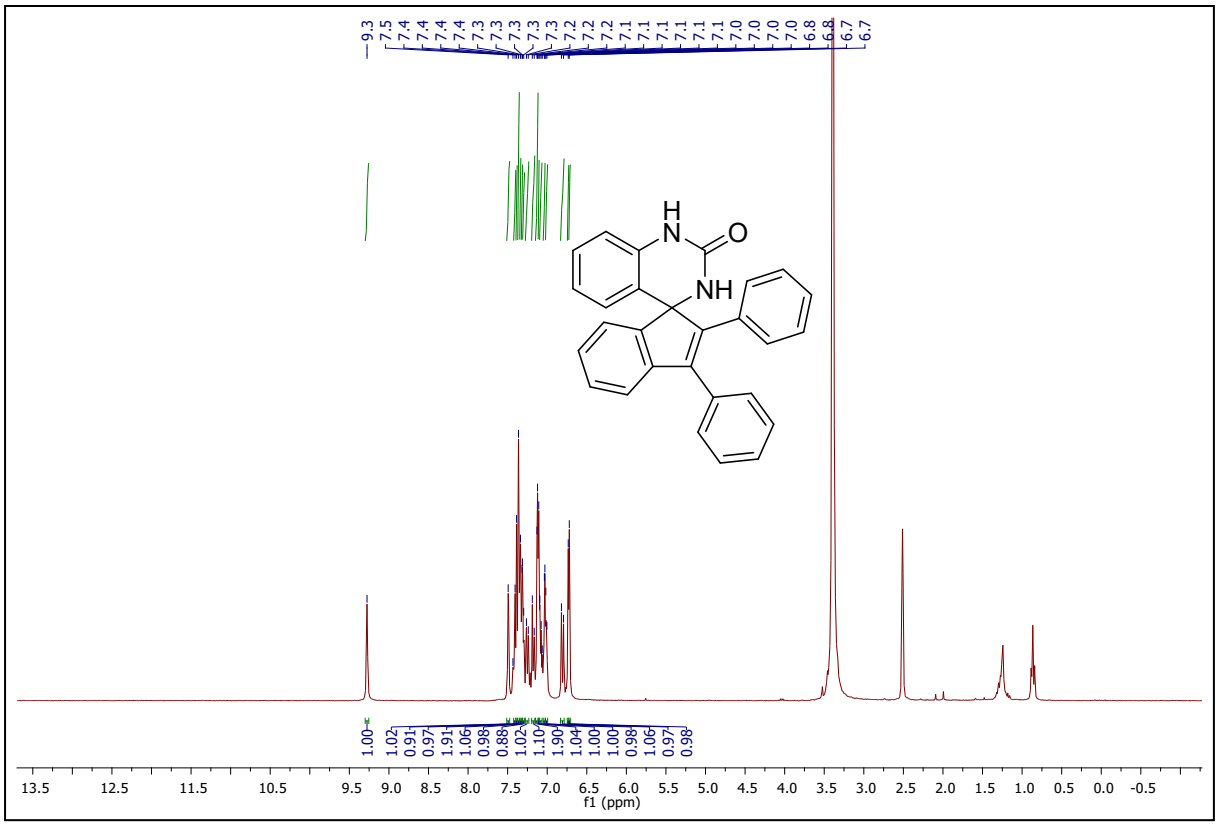
White solid, (0.164g, 94%) mp 282-284 ^{o}C, ^{1}H NMR (500 MHz, DMSO) δ 7.74 (s, 1H), 7.41 – 7.39 (m, 1H), 7.38 (s, 1H), 7.35 (d, *J* = 7.0 Hz, 3H), 7.33 – 7.31 (m, 1H), 7.30 (d, *J* = 4.6 Hz, 1H), 7.26 – 7.21 (m, 2H), 7.18 (d, *J* = 7.5 Hz, 1H), 7.12 – 7.07 (m, 3H), 6.99 (d, *J* = 1.7 Hz, 1H), 6.98 (d, *J* = 2.4 Hz, 1H), 6.96 (d, *J* = 8.5 Hz, 1H), 6.86 – 6.82 (m, 1H), 6.79 (dd, *J* = 7.7, 1.5 Hz, 1H), 3.13 (s, 3H). ^{13}C NMR (101 MHz, DMSO) δ 153.9, 152.6, 148.2, 142.1, 140.1, 140.0, 134.8, 134.4, 129.9, 129.5, 129.2, 129.1, 129.0, 128.3, 128.1, 127.7, 127.6, 124.9, 123.5, 122.2, 122.0, 121.0, 114.3, 111.3, 71.4, 29.3. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{29}H_{23}N_2O$: 415.1810 found: 415.1791.

1'-Isopropyl-7'-methyl-2,3-diphenyl-1'*H*-spiro[indene-1,4'-quinazolin]-2'(3'H)-one 3z:

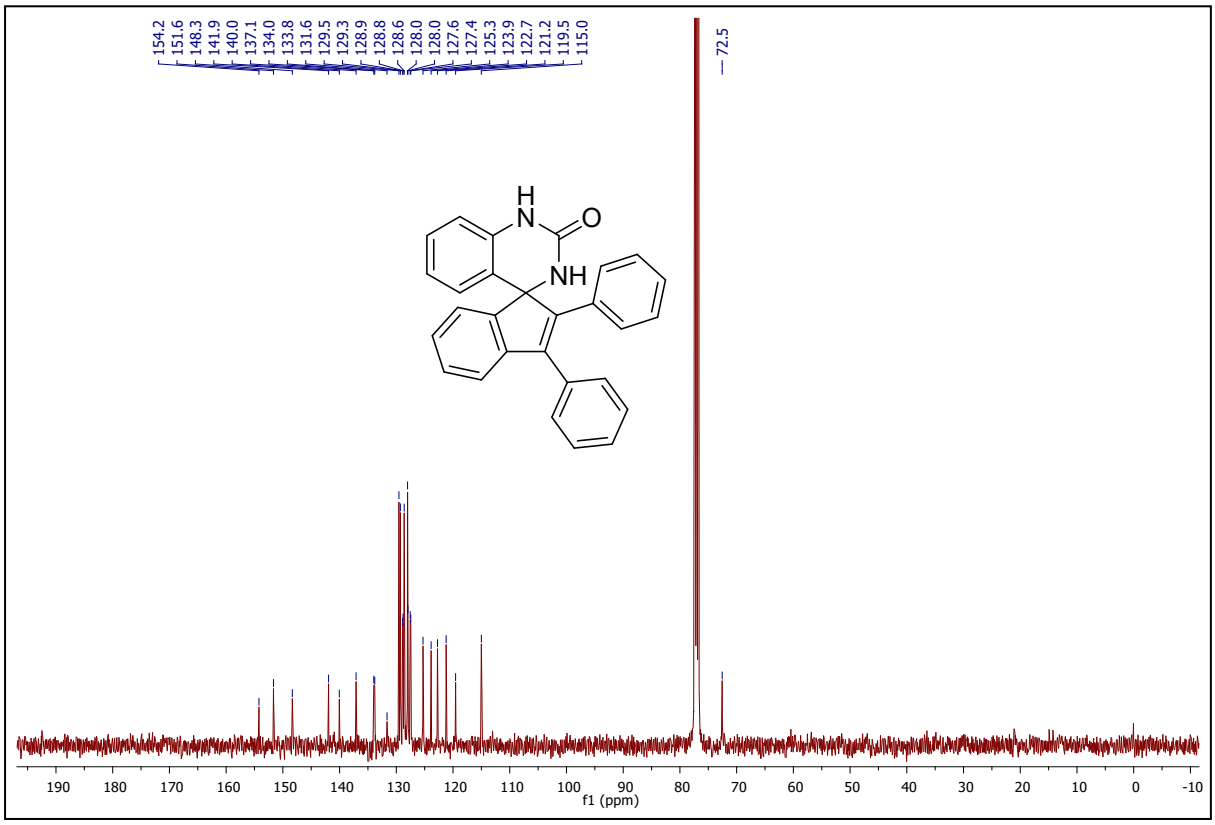
Grey solid, (0.136g, 83%) mp ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.43 (d, *J* = 6.4 Hz, 1H), 7.37 (s, 2H), 7.35 (s, 2H), 7.27 (s, 2H), 7.23 – 7.18 (m, 2H), 7.08 (s, 1H), 7.05 (d, *J* = 6.5 Hz, 2H), 6.99 (d, *J* = 7.0 Hz, 2H), 6.91 (s, 1H), 6.61 (s, 2H), 5.24 (s, 1H), 4.62 (s, 1H), 2.26 (s, 3H), 1.26 (s, 3H), 1.14 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 141.8, 139.0, 138.3, 134.6, 129.7, 129.4, 128.6,128.5,128.2, 127.8, 127.2, 124.1, 124.0, 120.9, 69.0, 32.0, 22.0, 21.2. HRMS (ESI) *m/z*: $[M+H]^+$ calcd for $C_{32}H_{29}N_2O$:457.2280 found:457.2290.

4. NMR spectra of products:

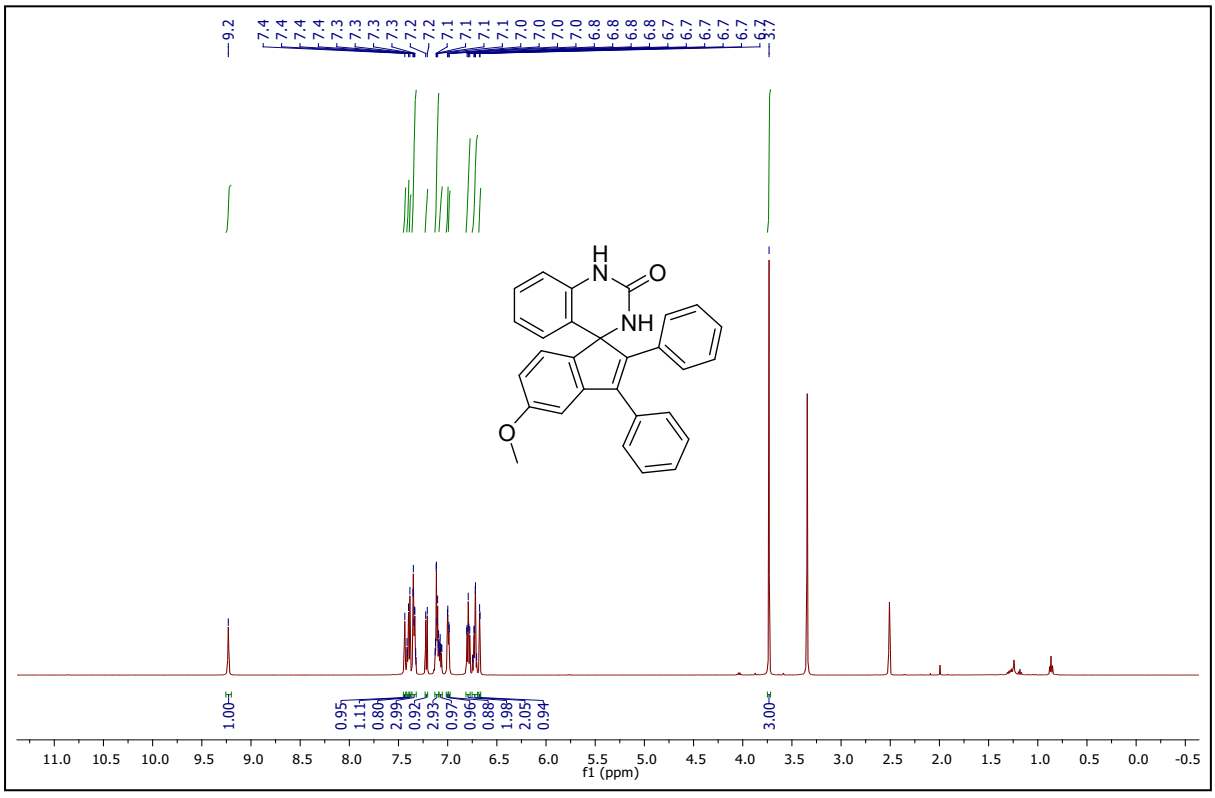
^{1}H NMR (400 MHz, DMSO) spectrum of 3a:



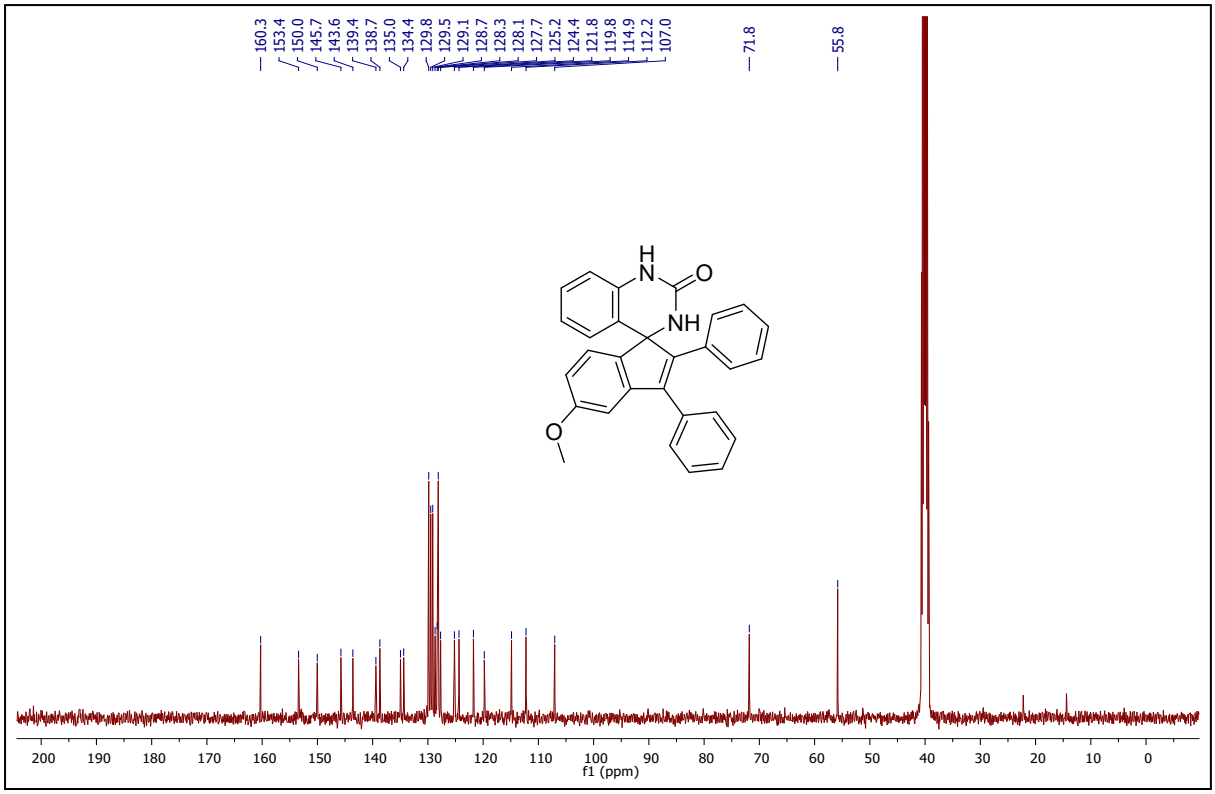
^{13}C NMR (101 MHz, DMSO) spectrum of 3a:



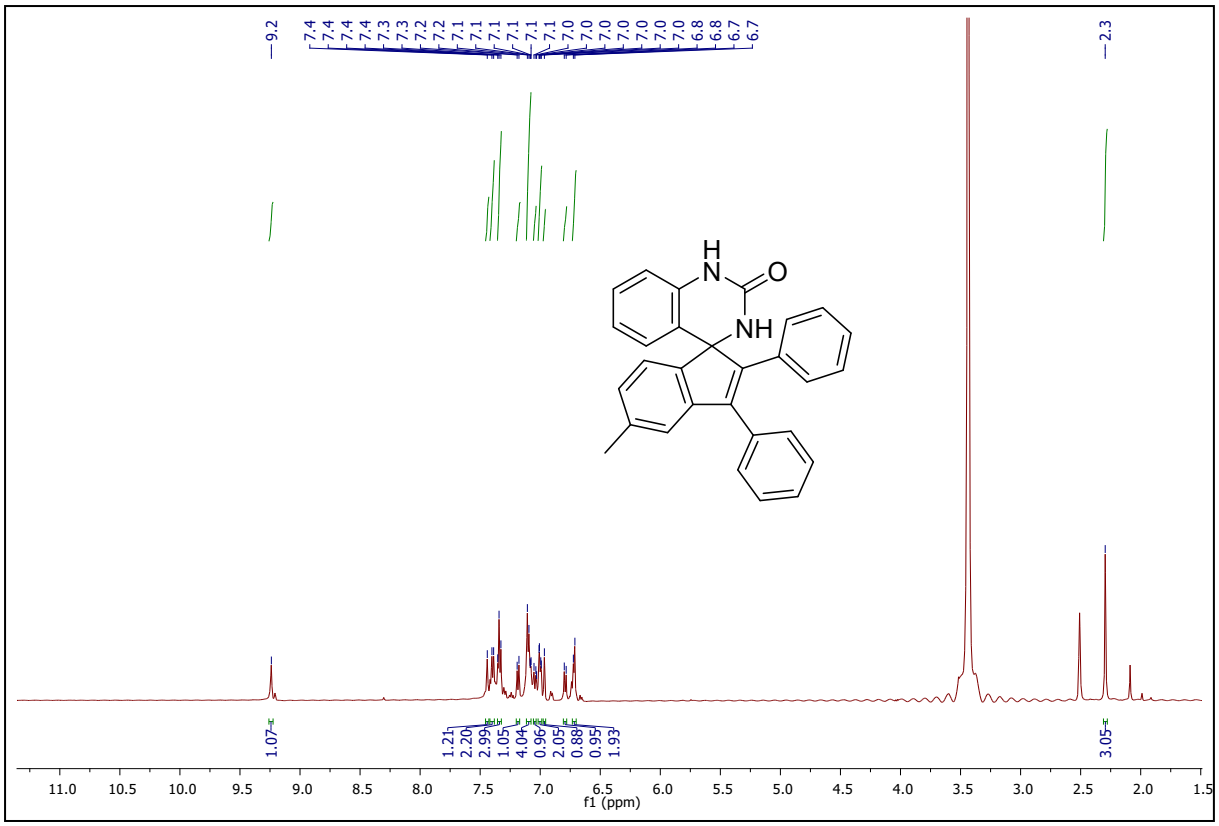
^{1}H NMR (400 MHz, DMSO) spectrum of 3b:



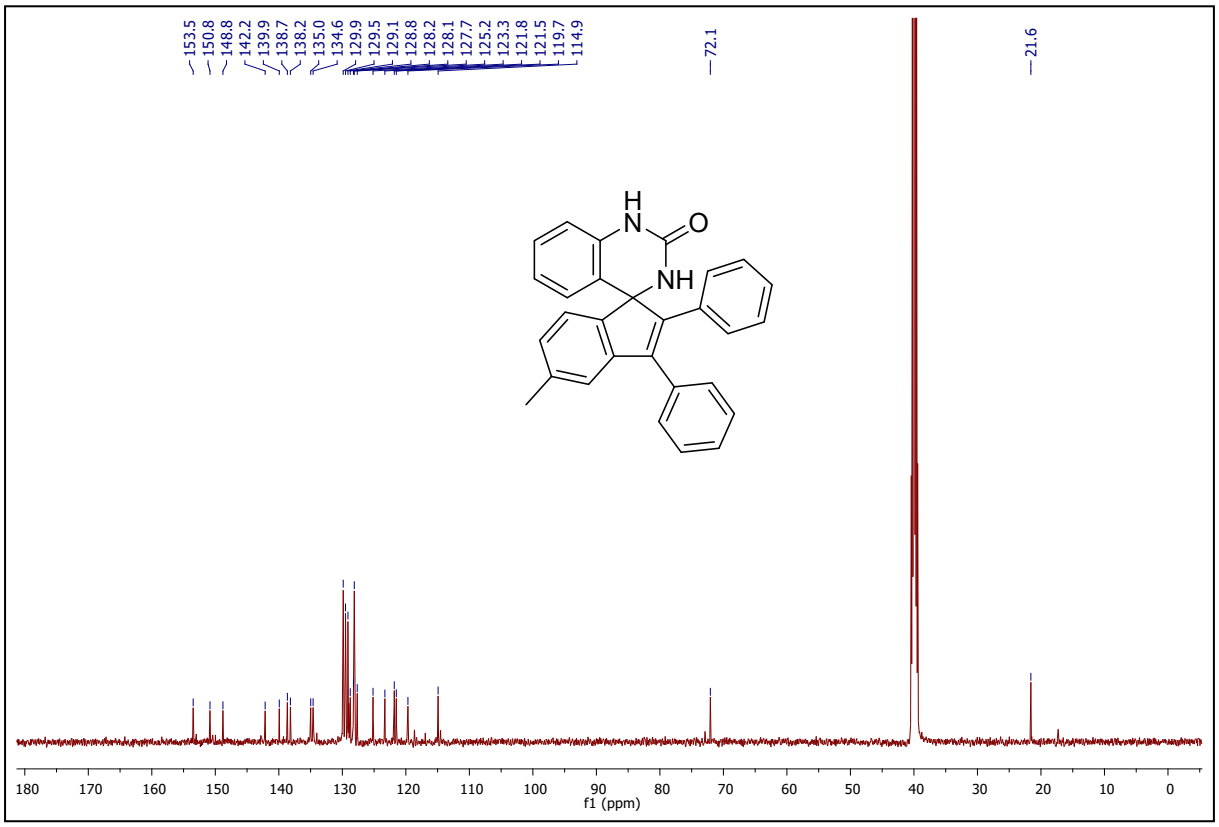
^{13}C NMR (101 MHz, DMSO) spectrum of 3b:



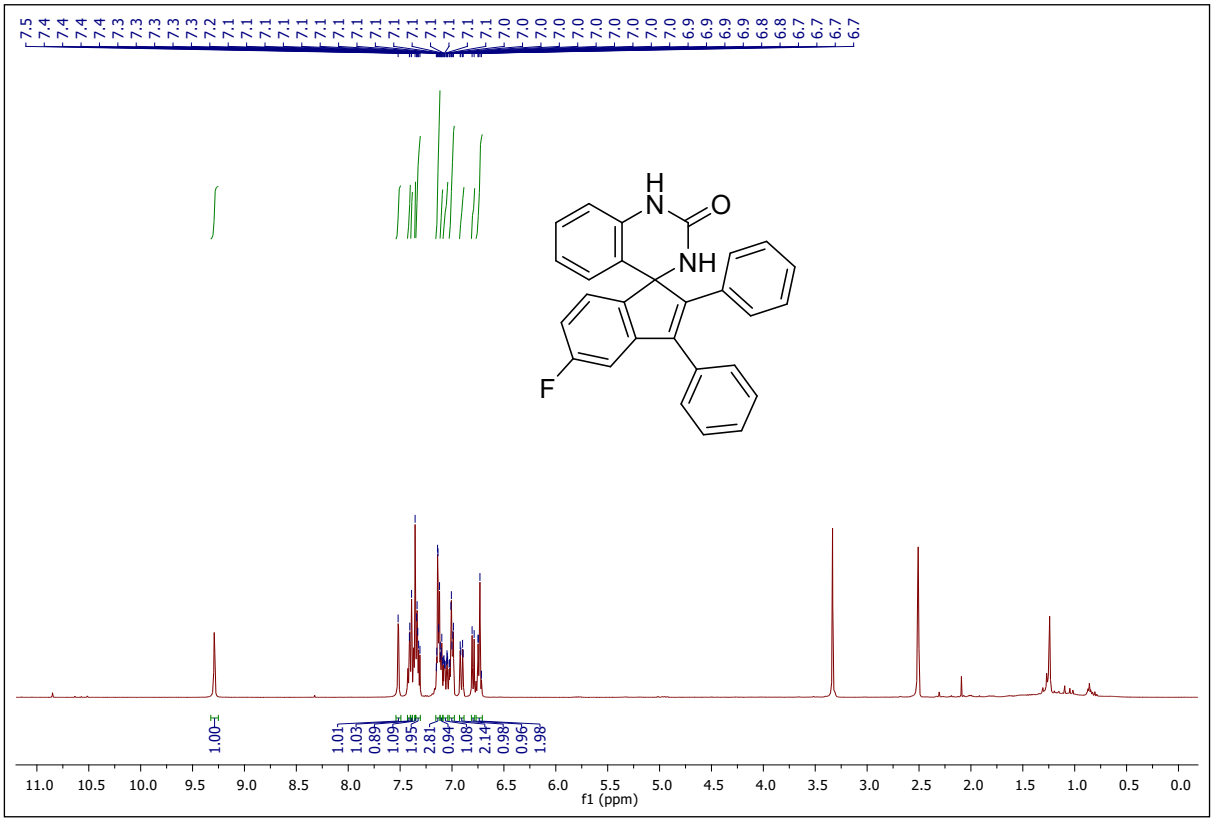
[1]H NMR (500 MHz, DMSO) spectrum of 3c:



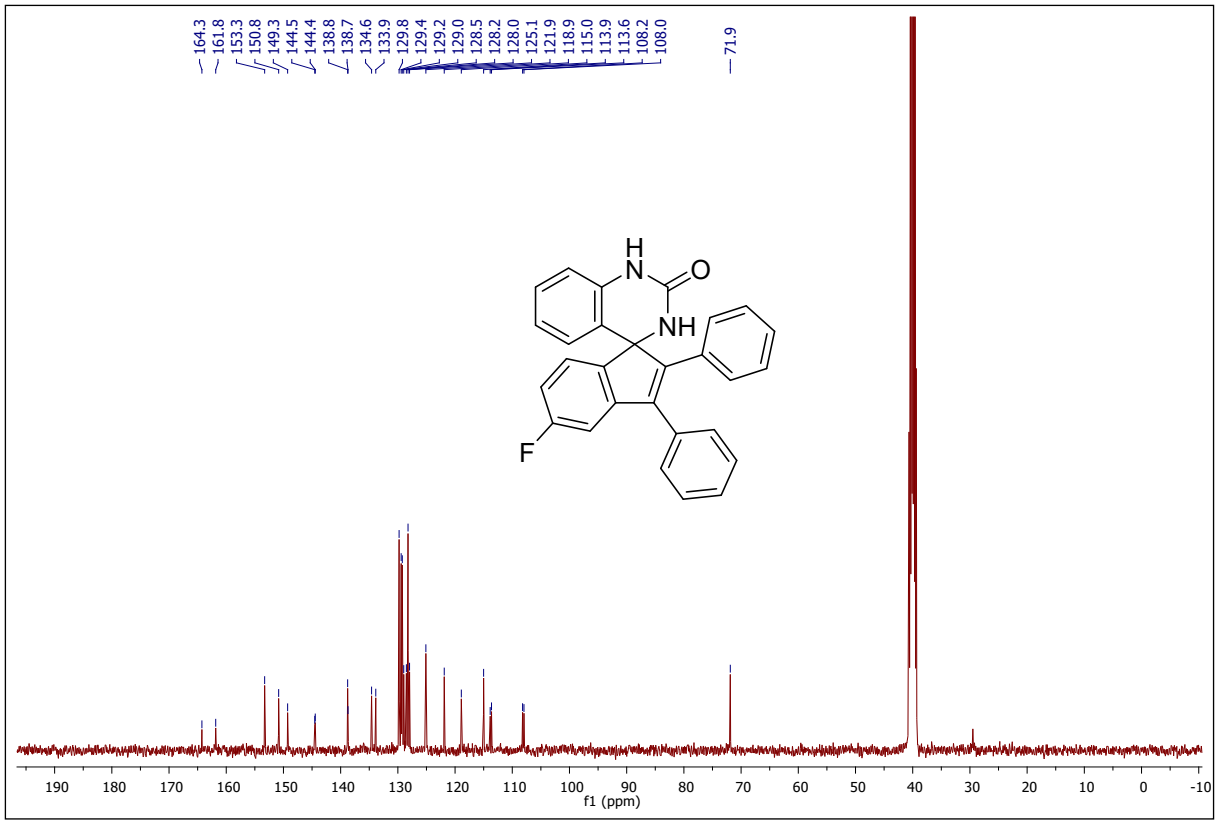
^{13}C NMR (101 MHz, DMSO) spectrum of 3c:



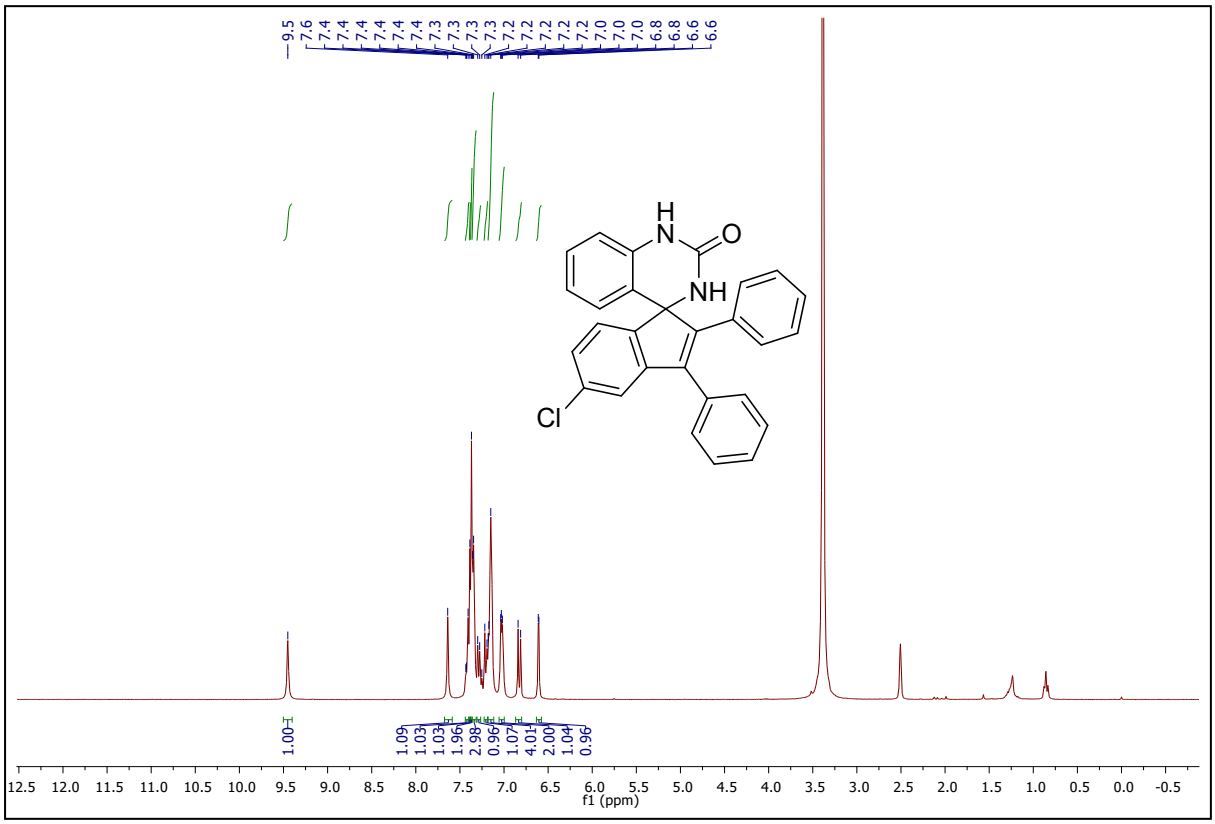
^{1}H NMR (400 MHz, DMSO) spectrum of 3d:



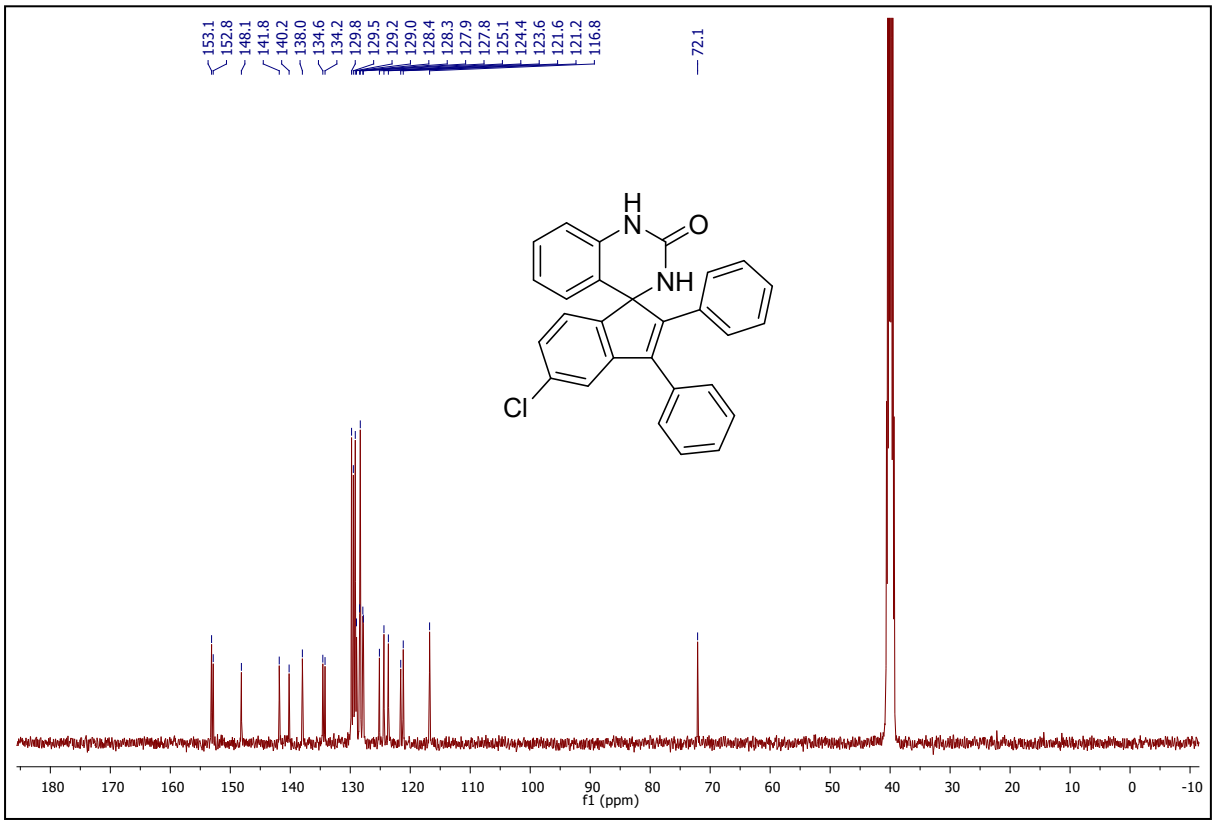
^{13}C NMR (101 MHz, DMSO) of compound 3d:



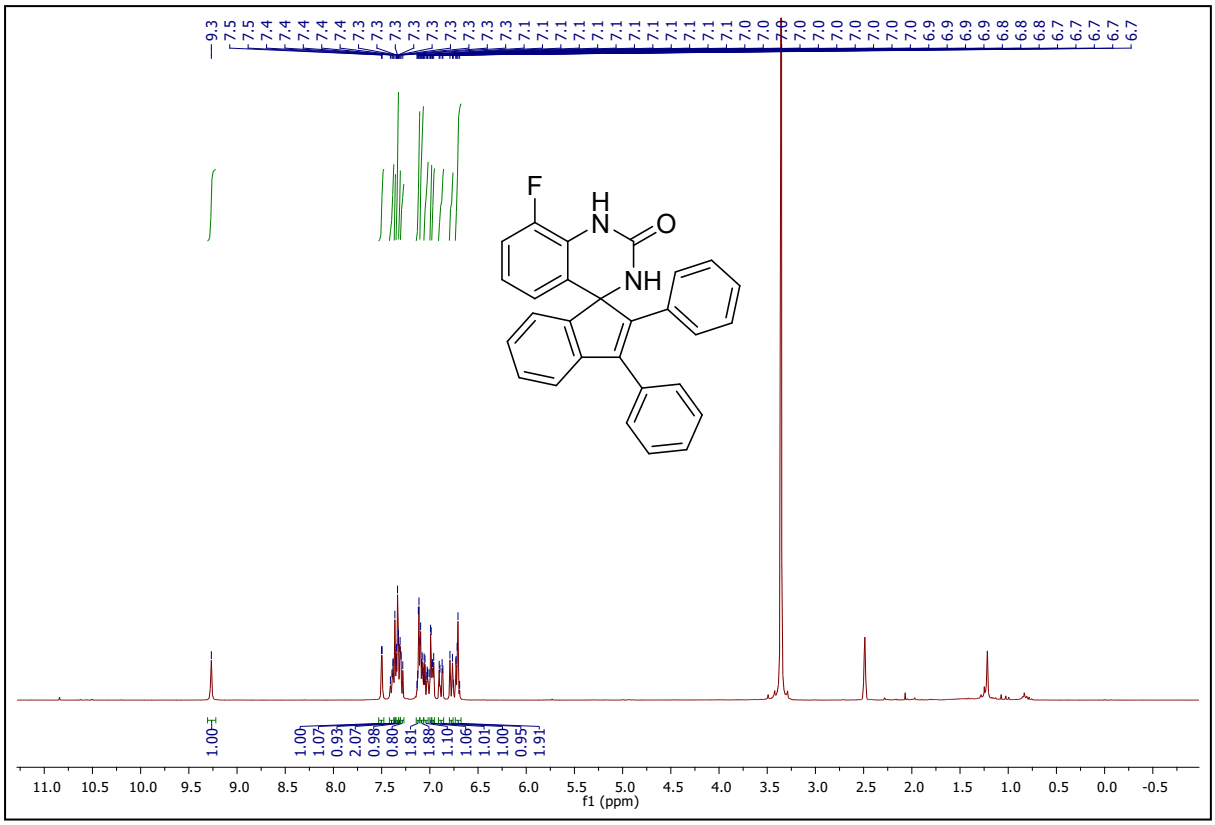
^{1}H NMR (300 MHz, DMSO) spectrum of 3e:



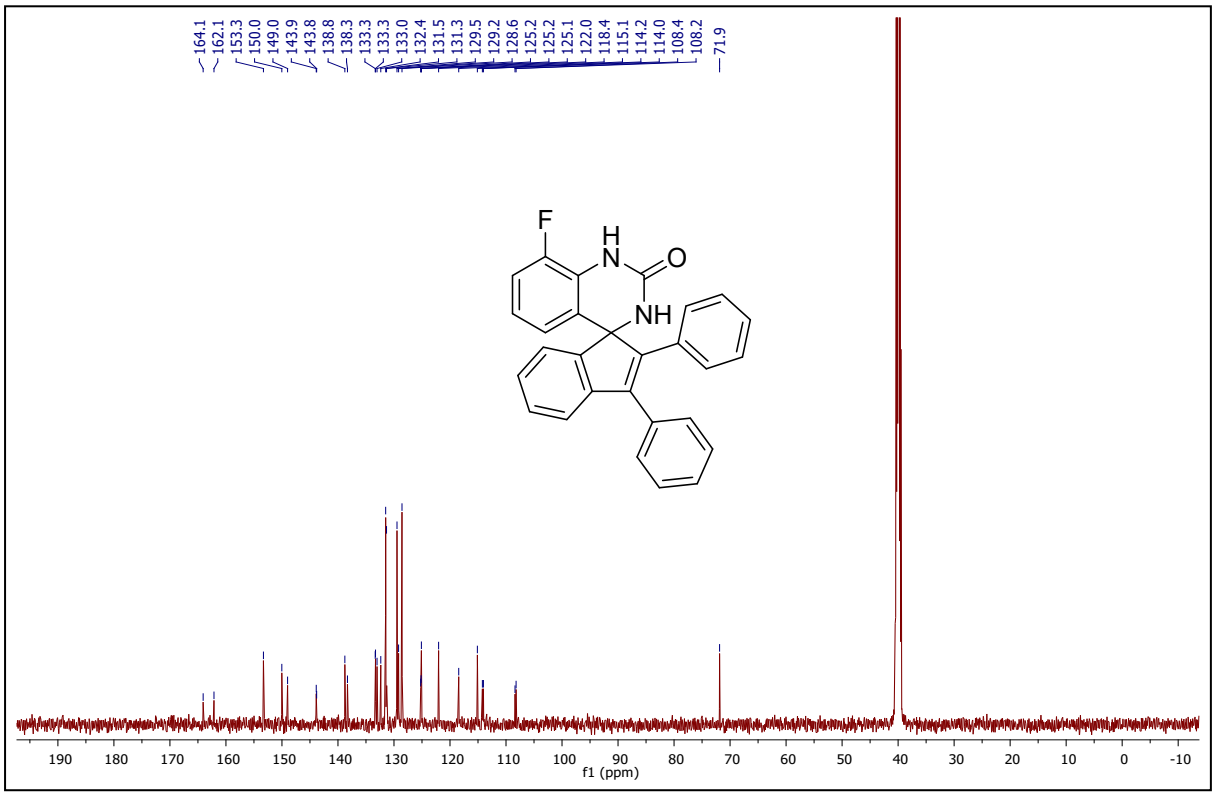
^{13}C NMR (101 MHz, DMSO) spectrum of 3e:



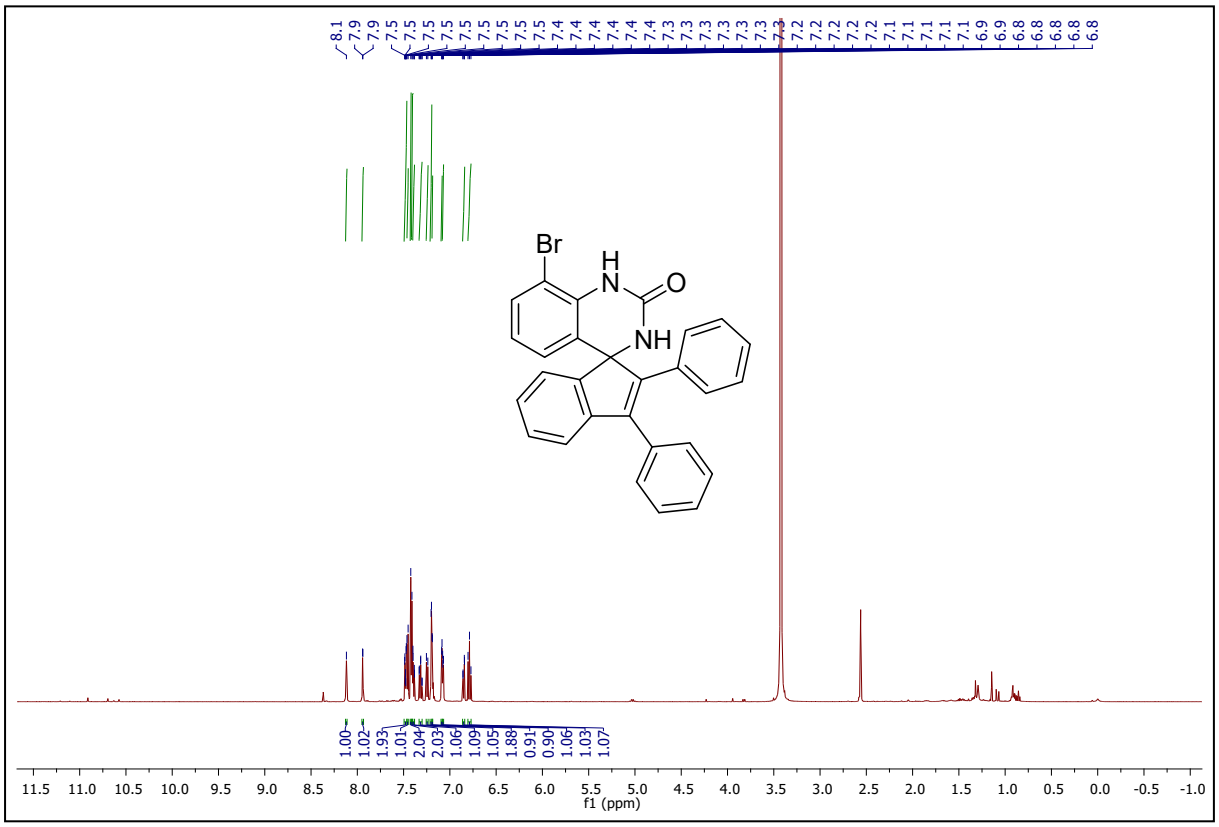
^{1}H NMR (300 MHz, DMSO) spectrum of 3f:



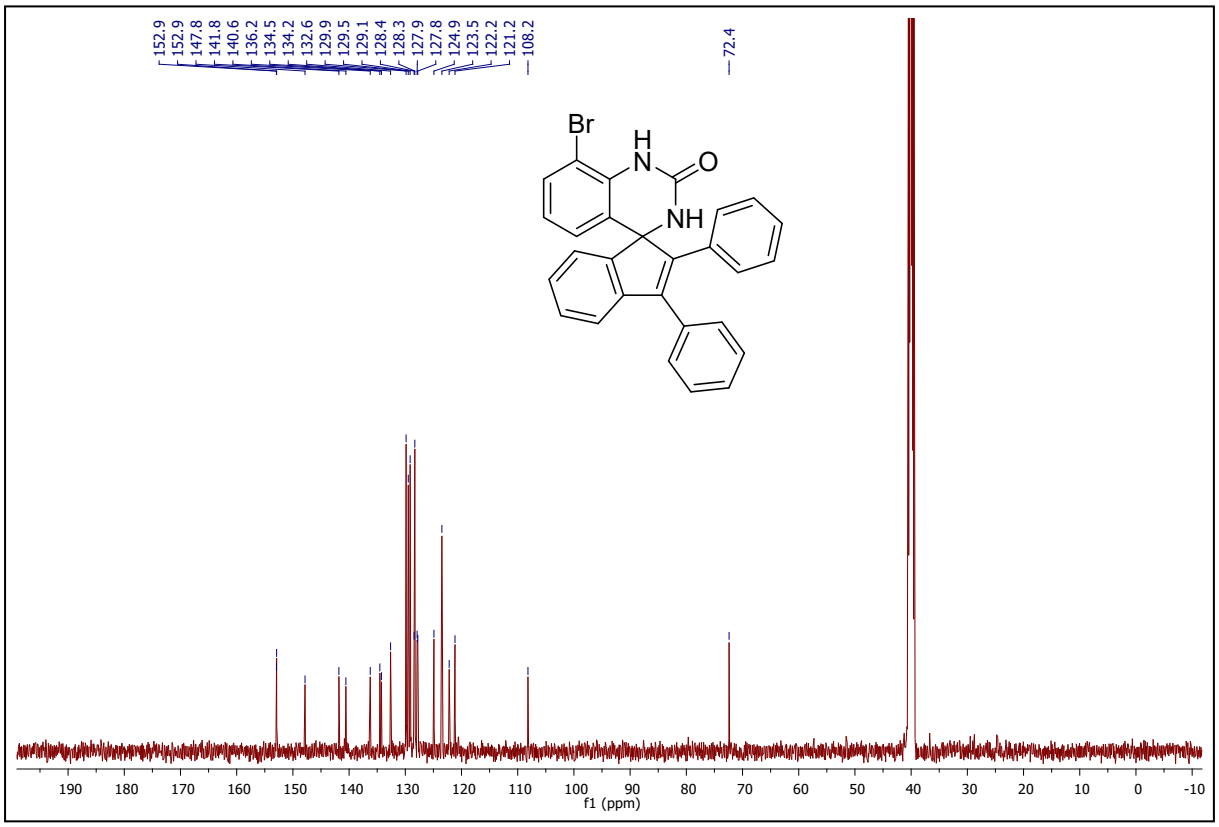
^{13}C NMR (126 MHz, DMSO) spectrum of 3f:



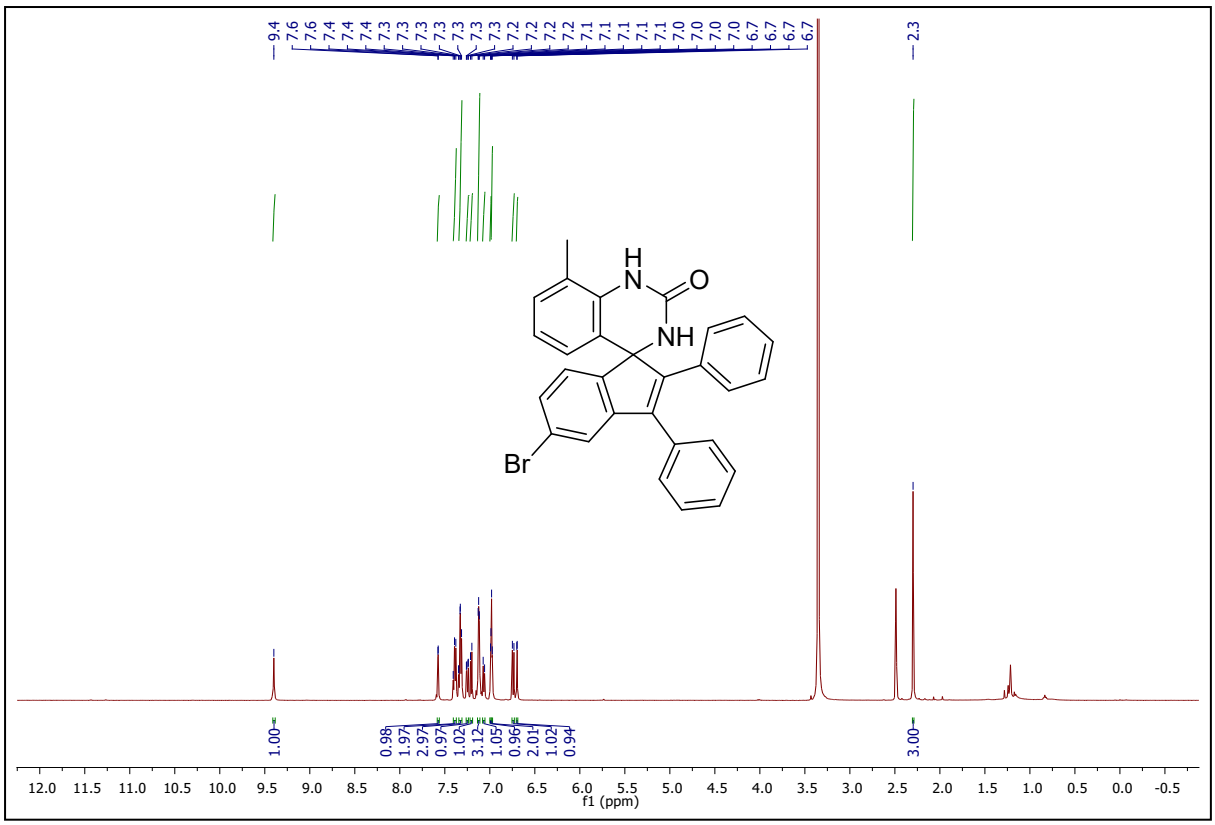
^{1}H NMR (500 MHz, DMSO) spectrum of 3g:



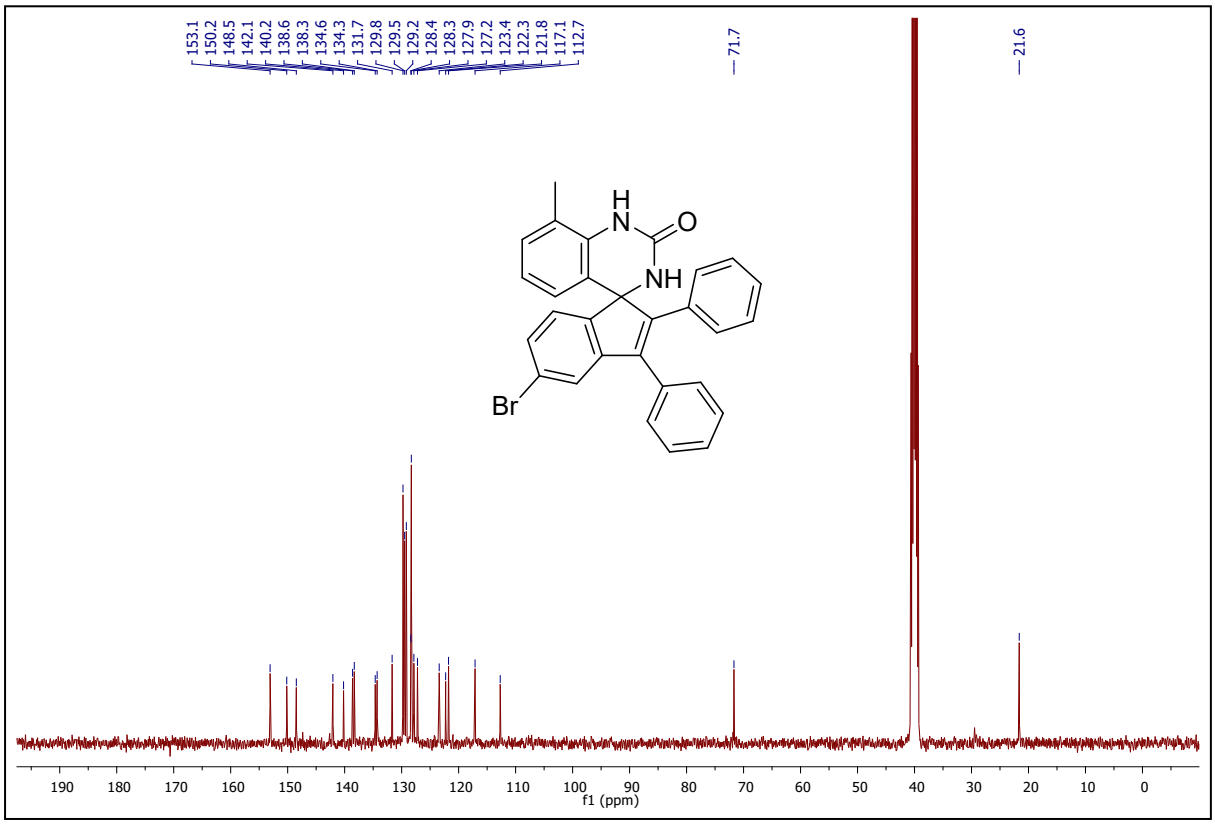
^{13}C NMR (126 MHz, DMSO) of compound 3g:



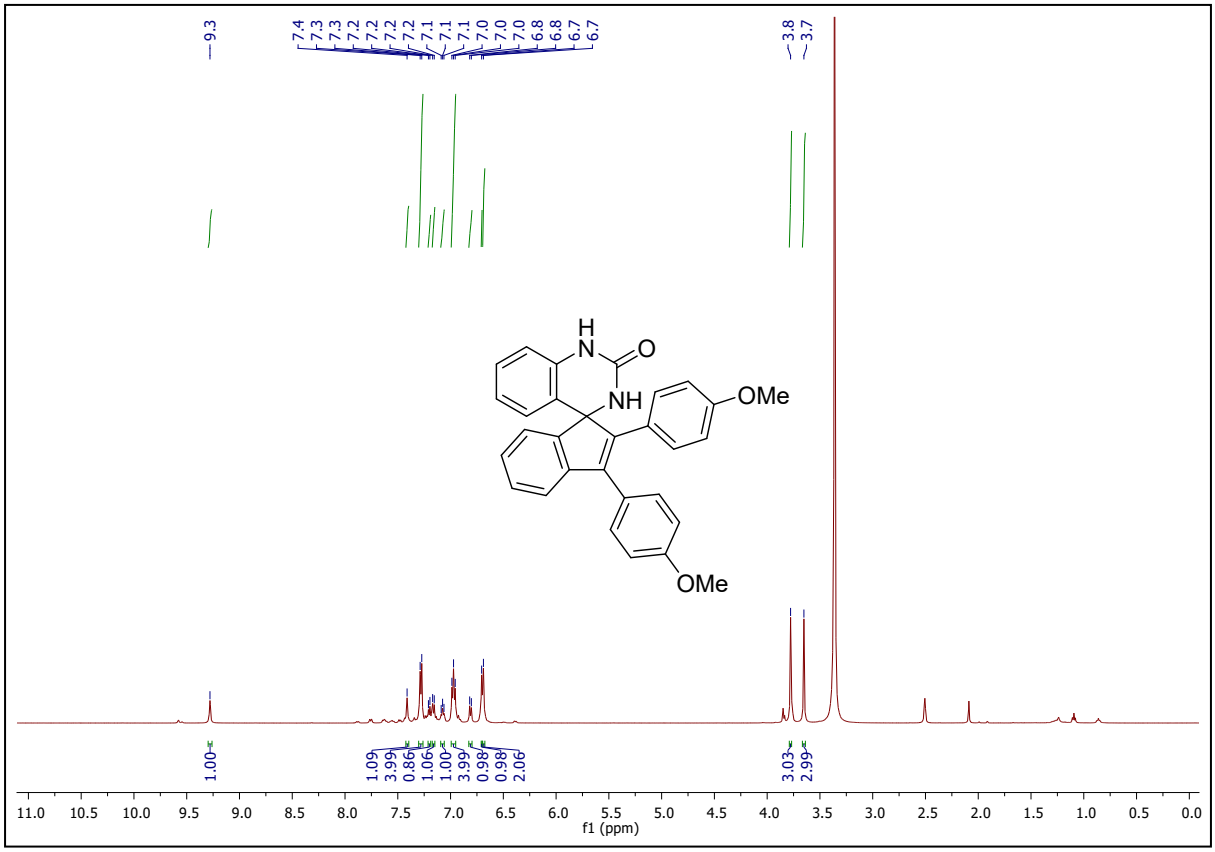
^{1}H NMR (500 MHz, DMSO) spectrum of 3h:



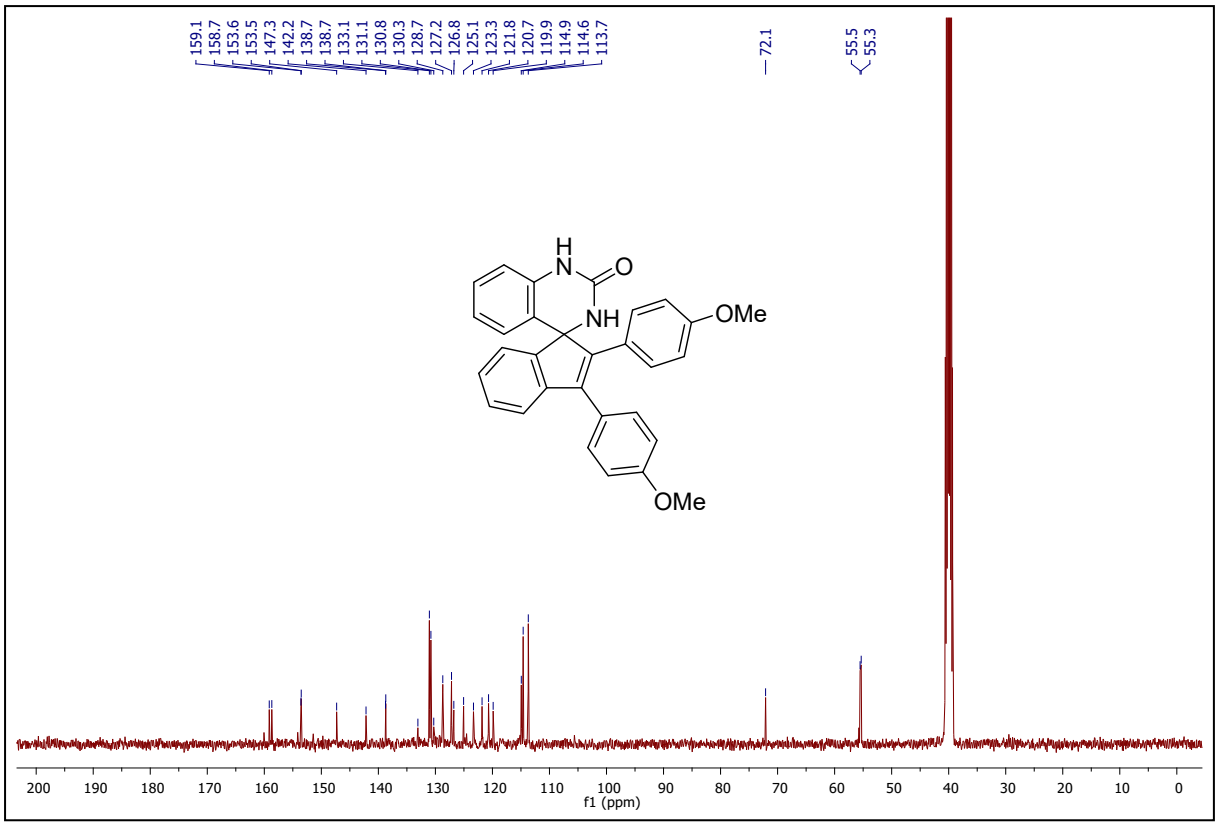
^{13}C NMR (101 MHz, DMSO) spectrum of 3h:



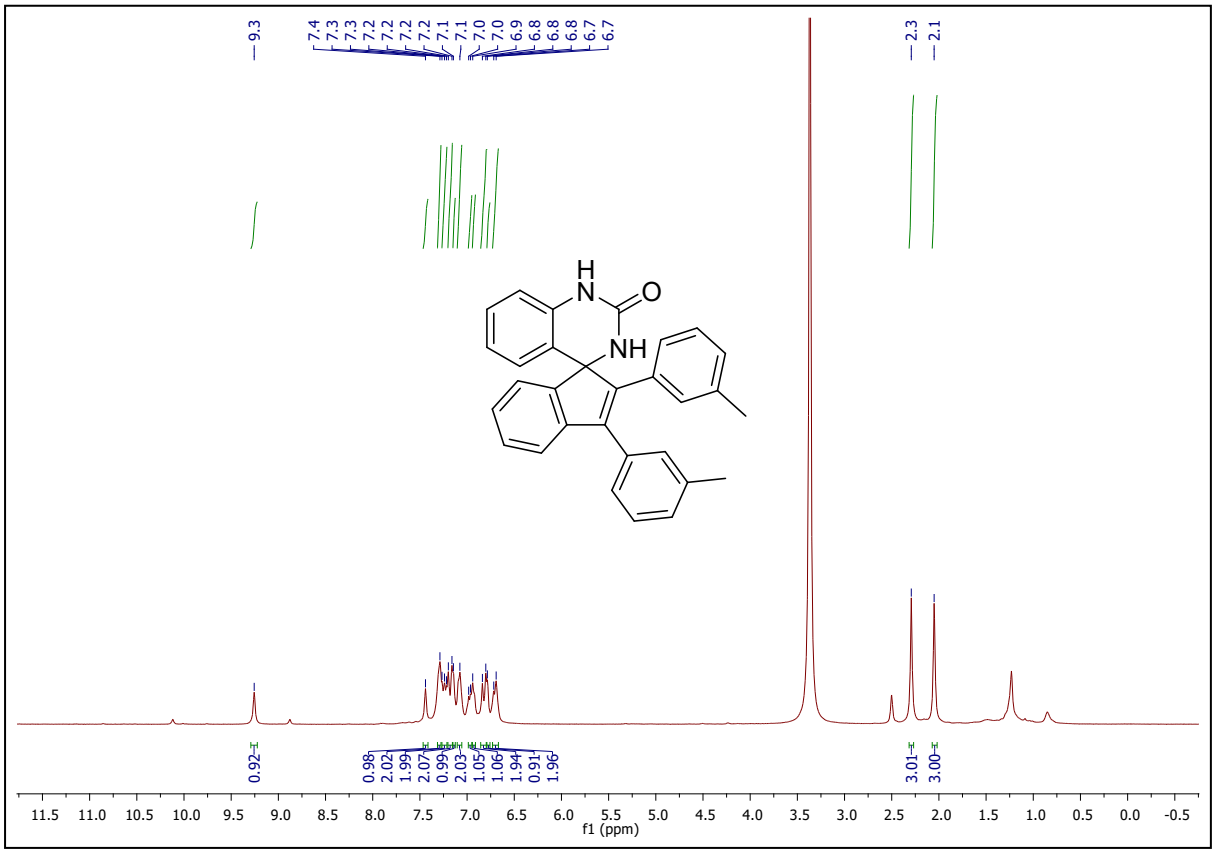
^{1}H NMR (400 MHz, DMSO) spectrum of 3i:



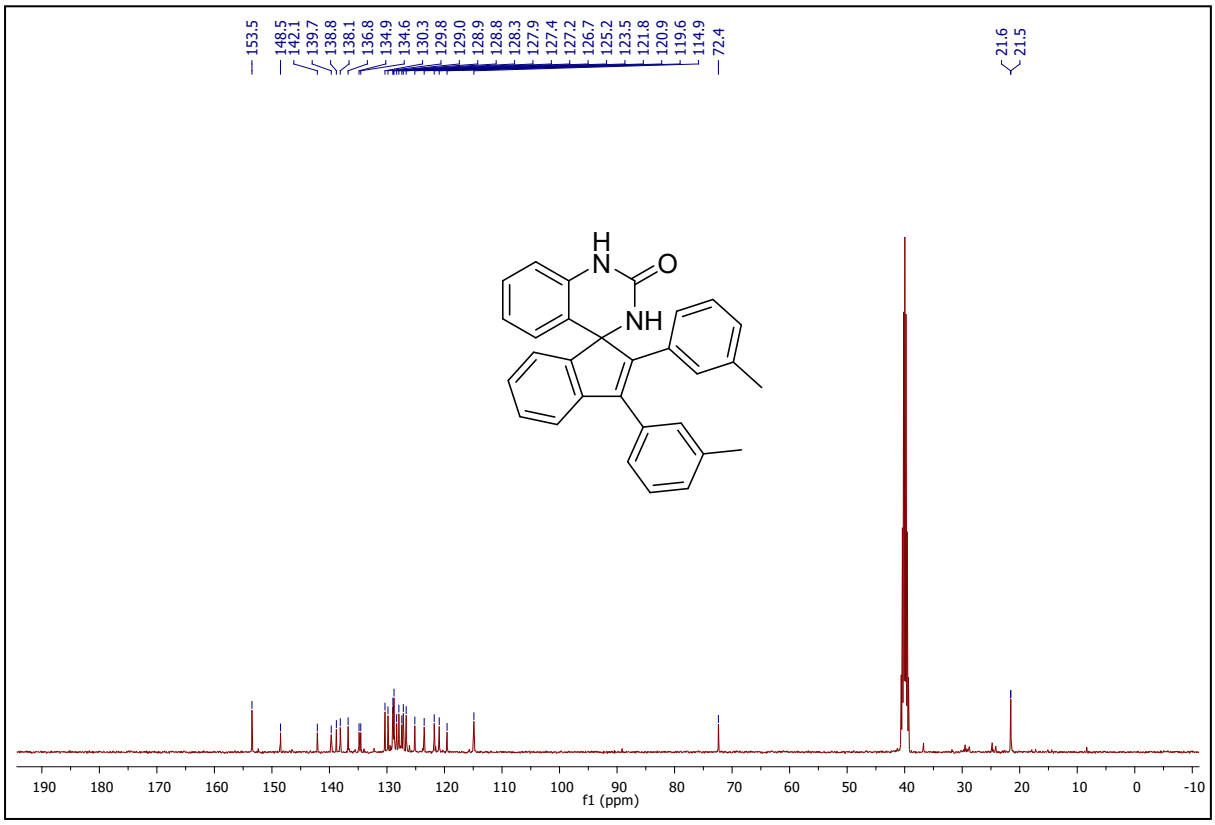
^{13}C NMR (101 MHz, DMSO) of compound 3i:



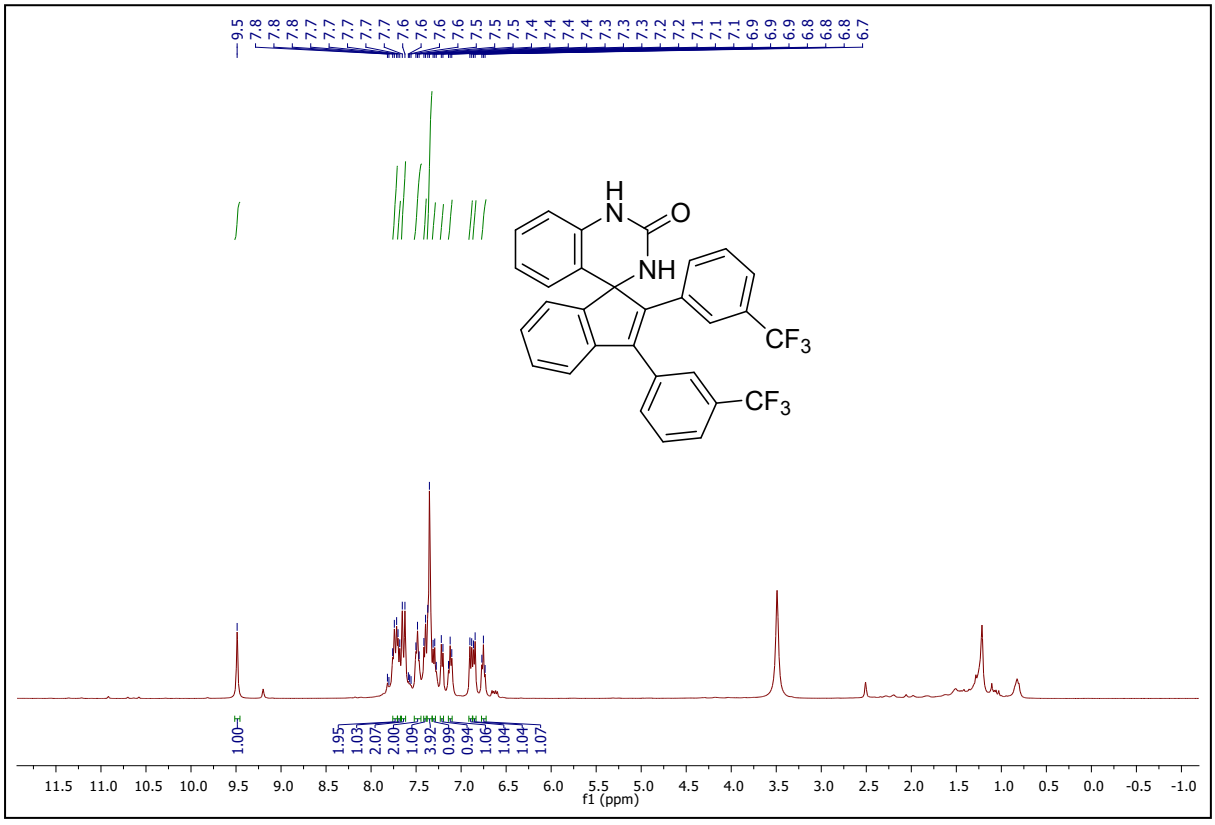
^{1}H NMR (400 MHz, DMSO) spectrum of 3j:



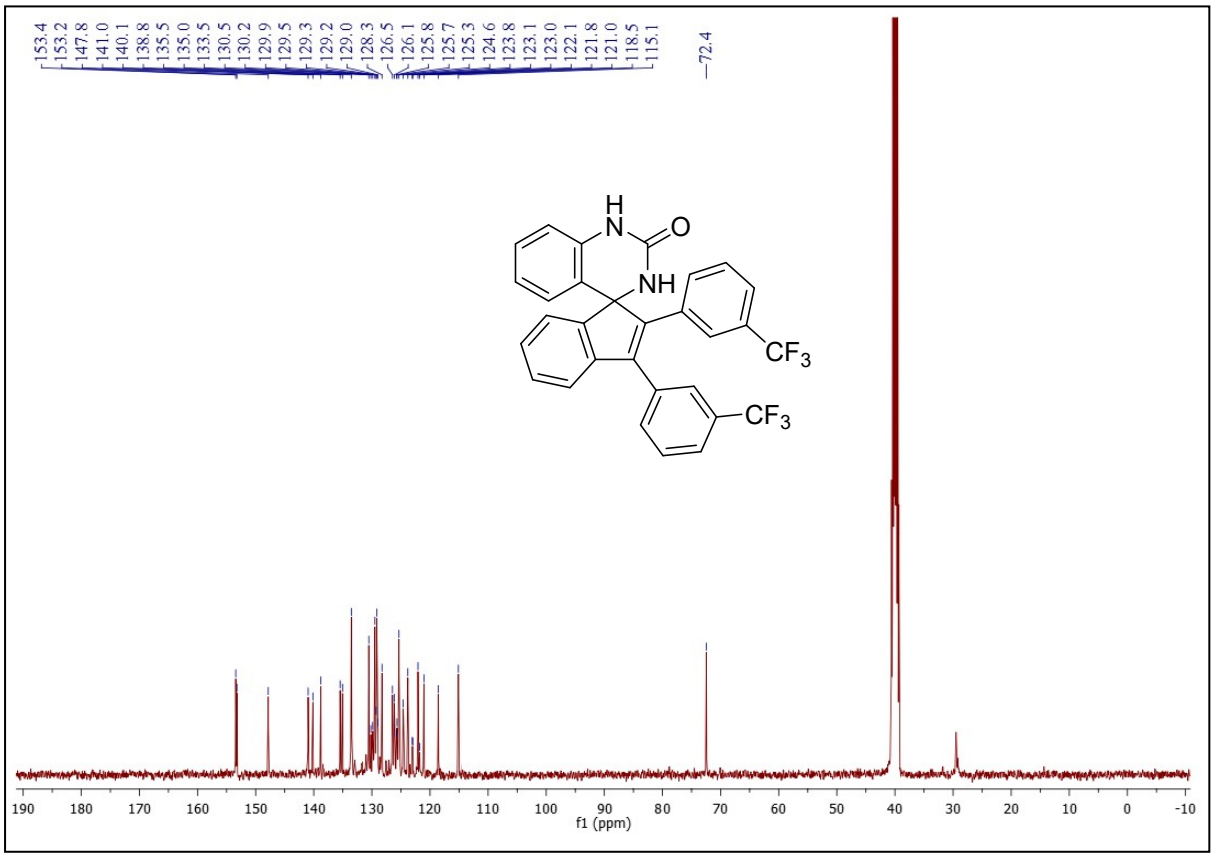
^{13}C NMR (101 MHz, DMSO) spectrum of 3j:



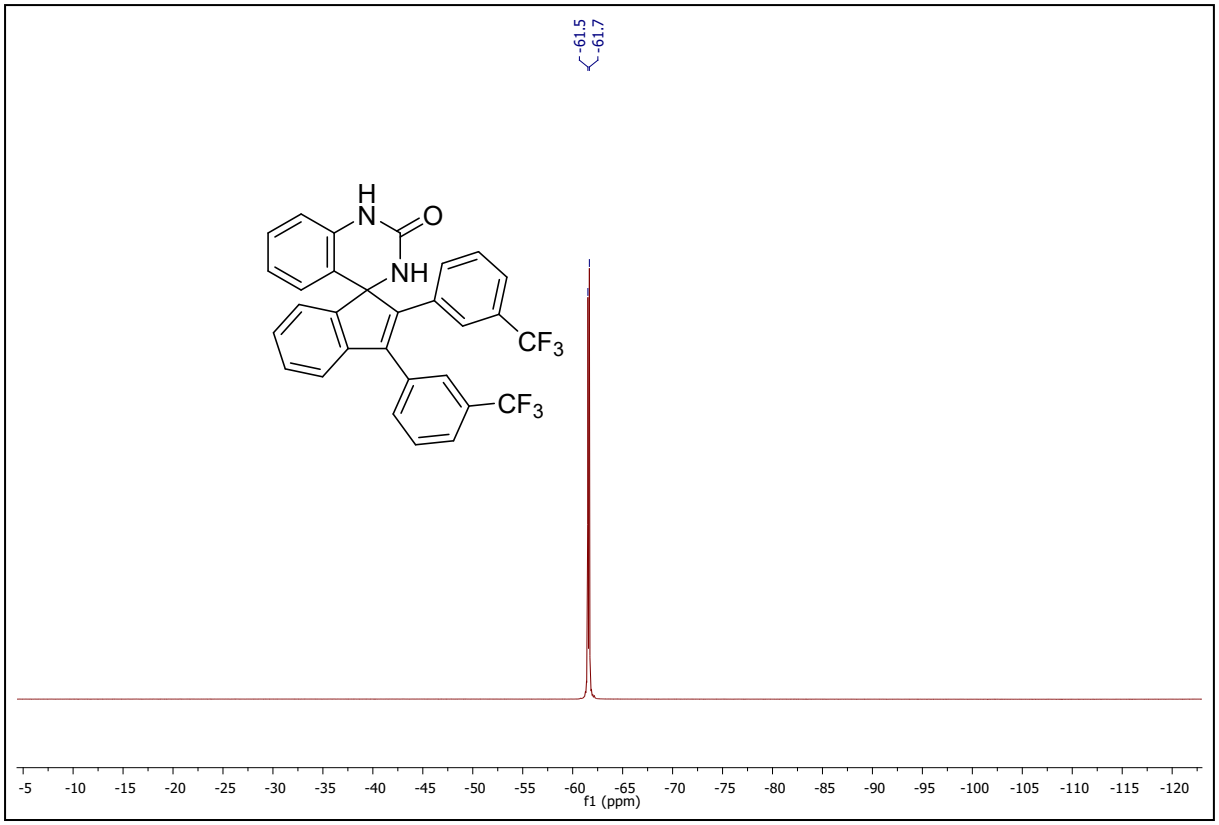
^{1}H NMR (400 MHz, DMSO) spectrum of 3k:



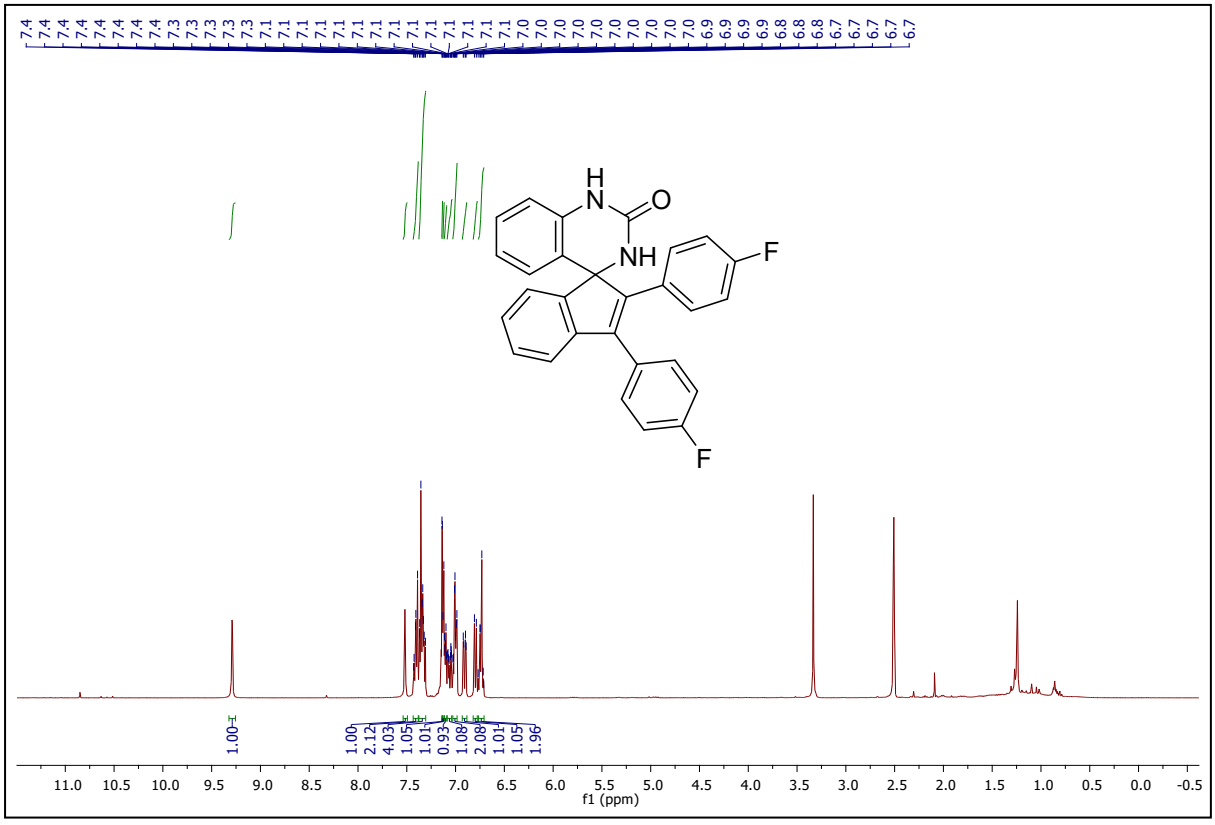
^{13}C NMR (126 MHz, DMSO) spectrum of 3k:



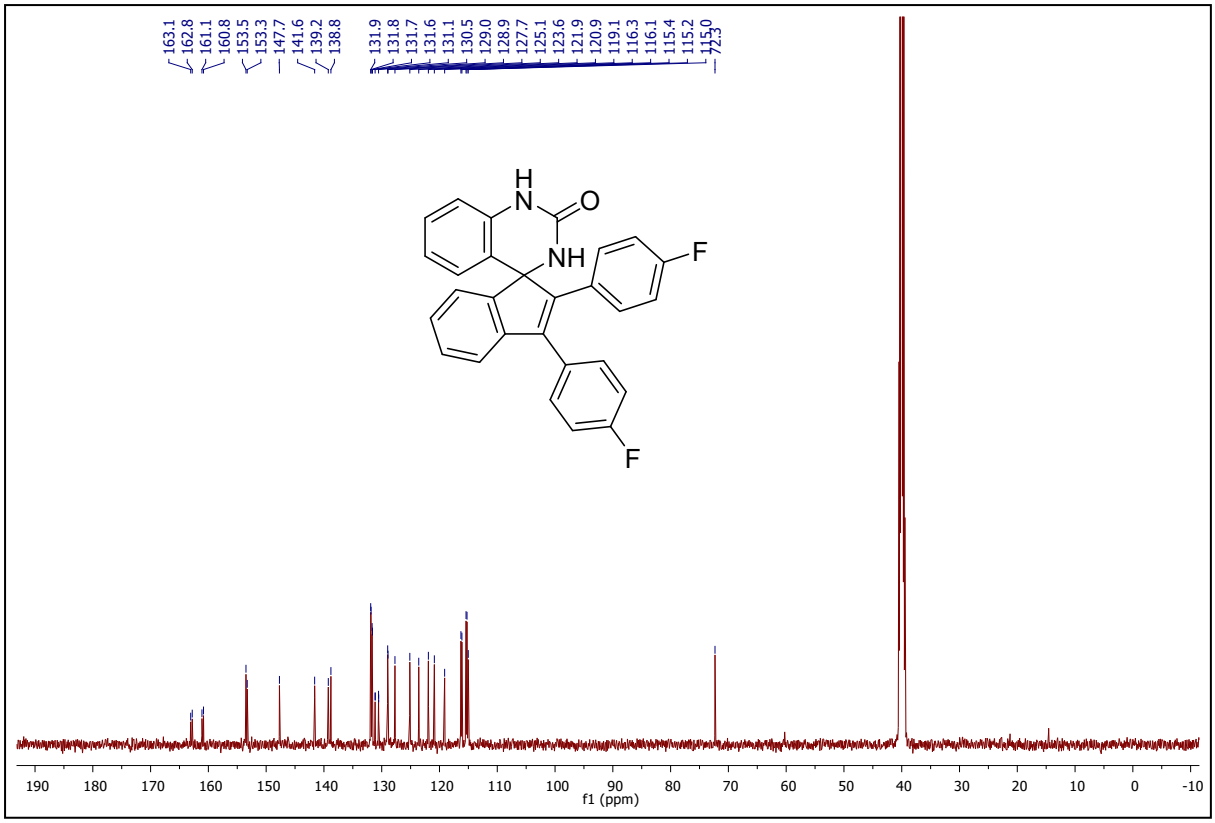
^{19}F NMR (376 MHz, DMSO) spectrum of 3k:



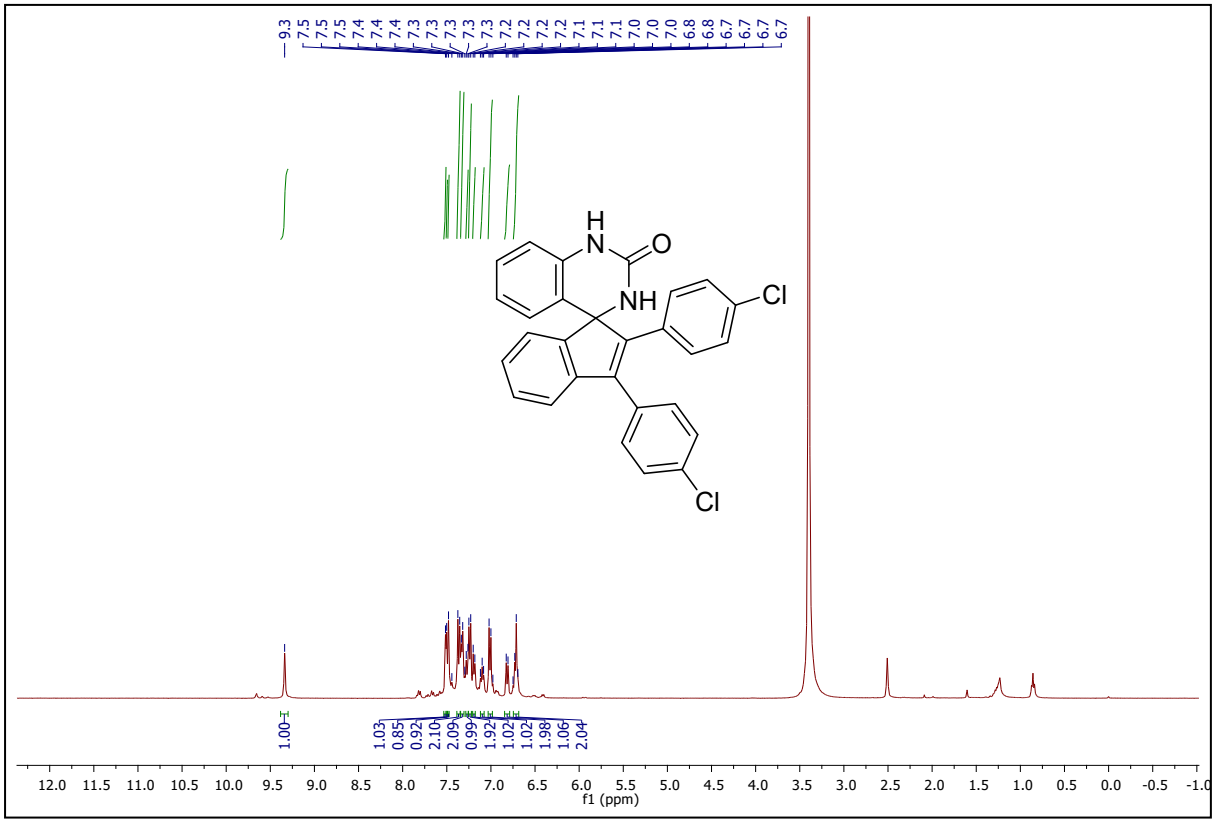
^{1}H NMR (300 MHz, DMSO) spectrum of 3l:



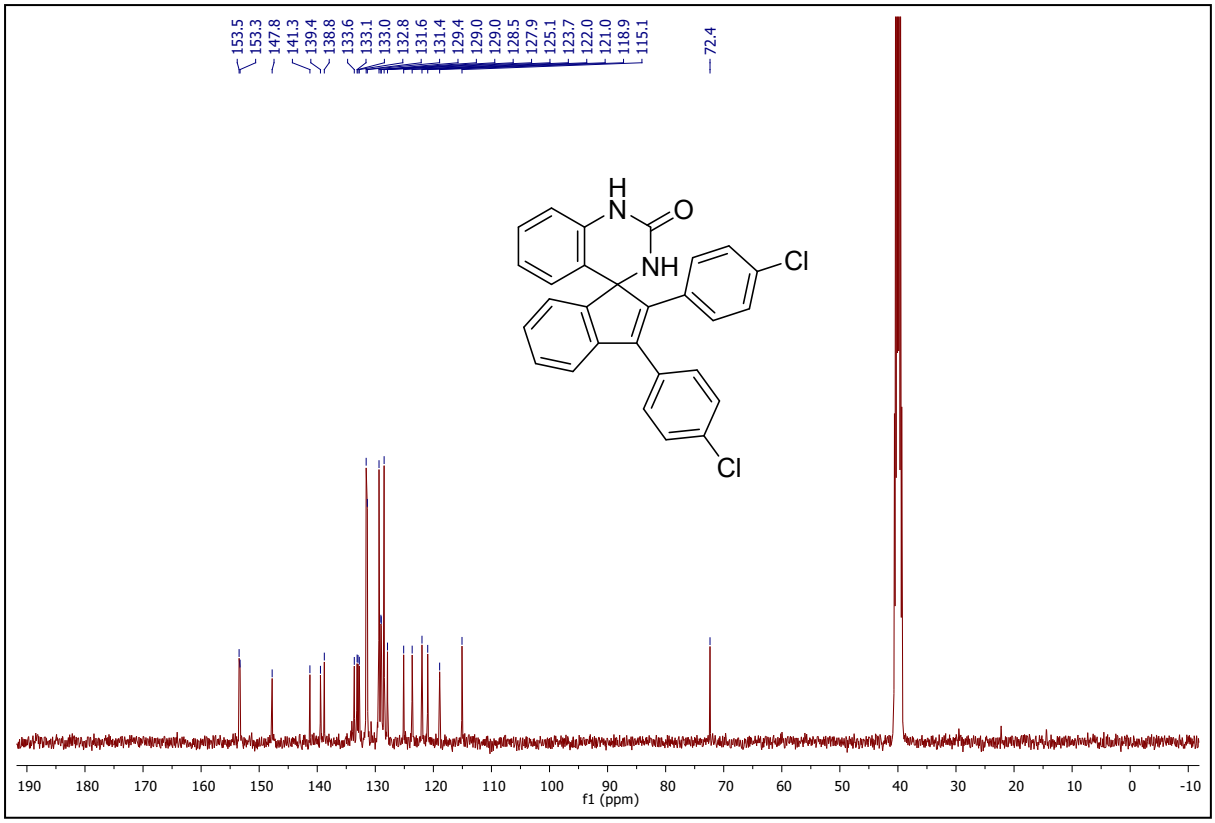
^{13}C NMR (101 MHz, DMSO) spectrum of 3l:



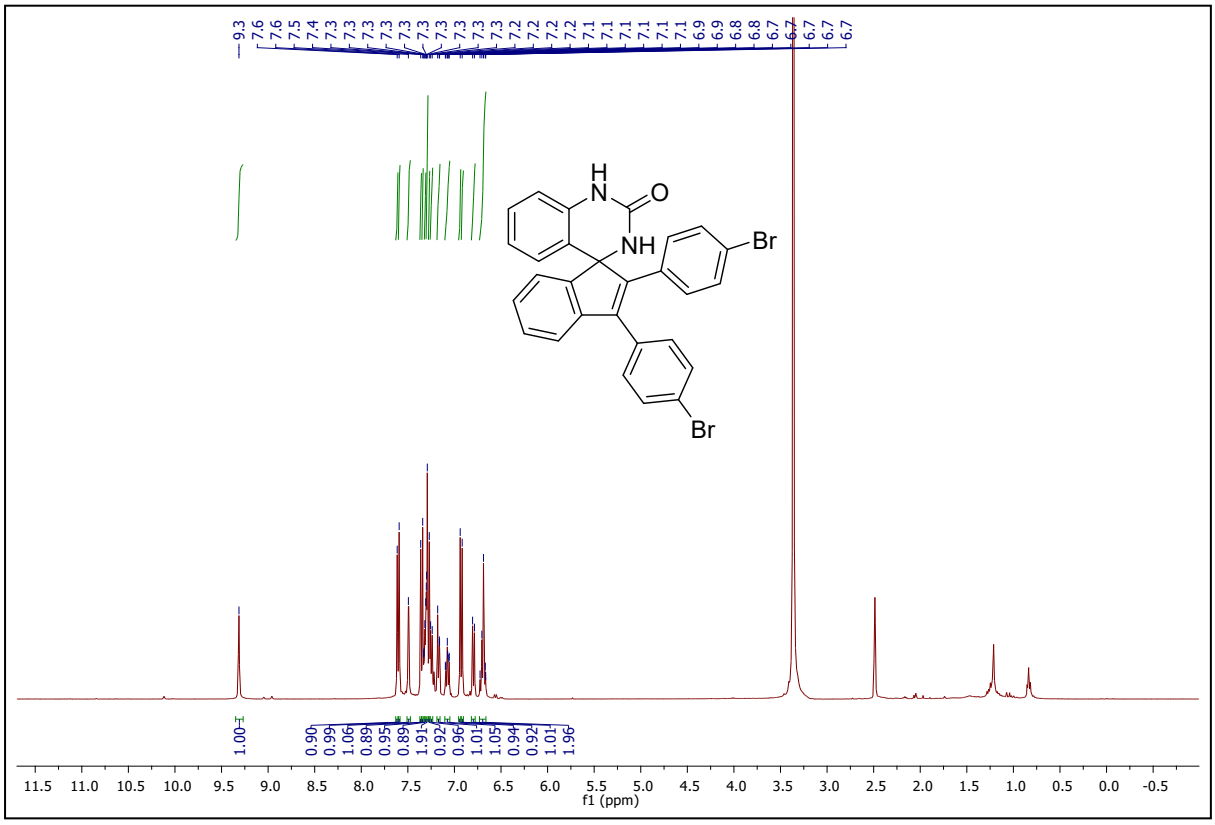
[1]H NMR (400 MHz, DMSO) spectrum of 3m:



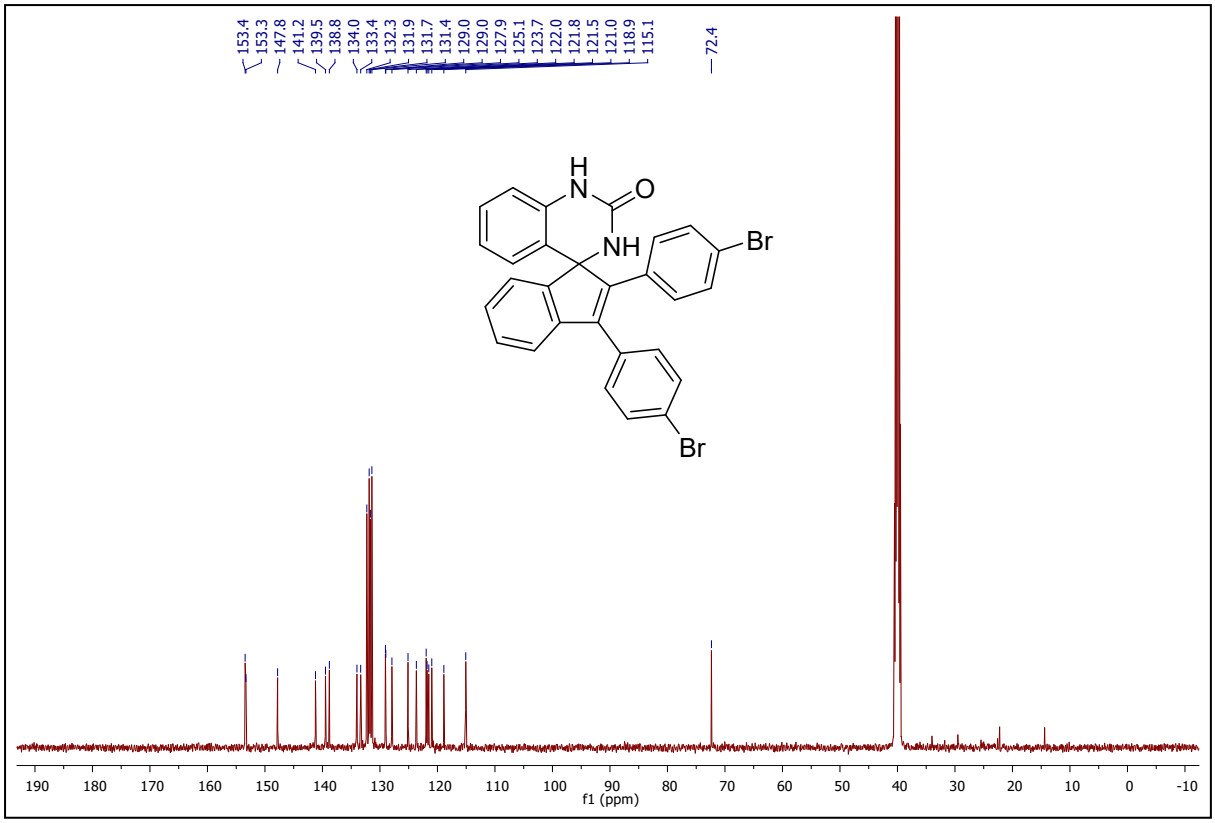
[13]C NMR (101 MHz, DMSO) spectrum of 3m:



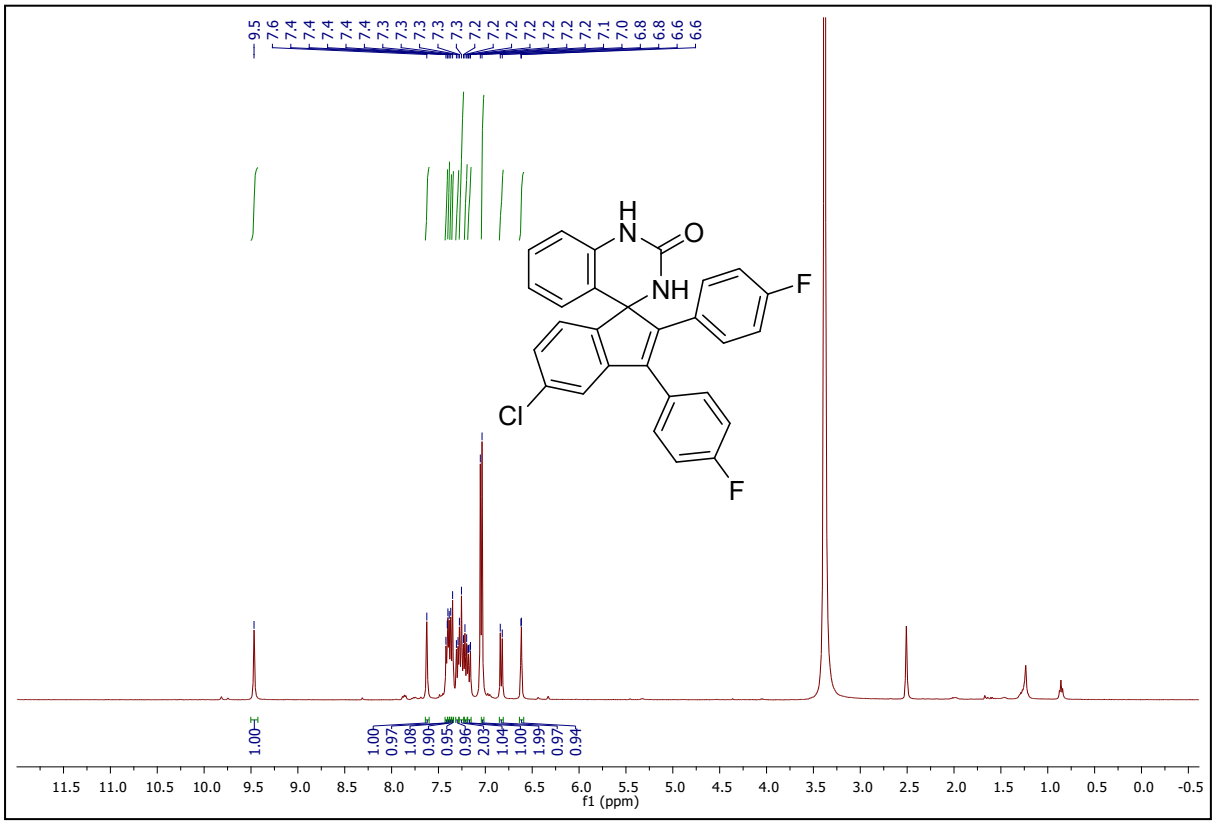
[1]H NMR (400 MHz, DMSO) spectrum of 3n:



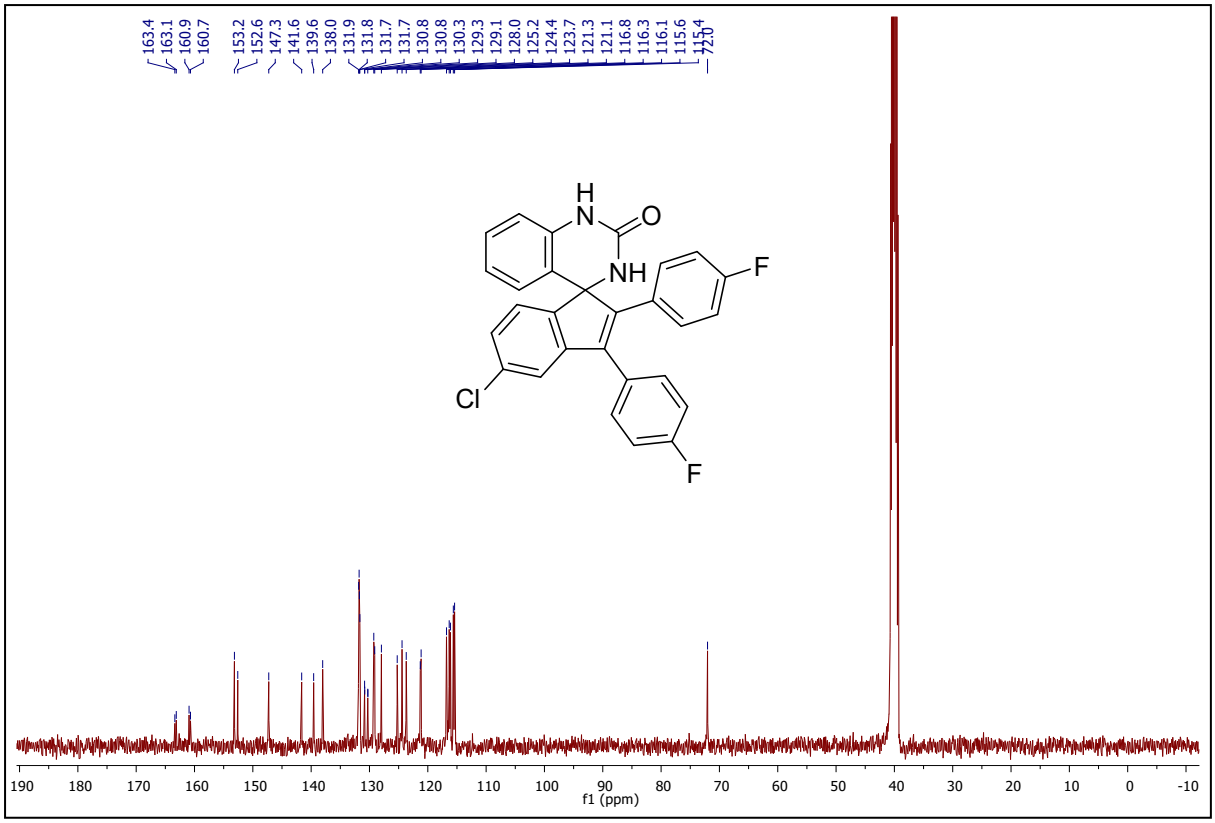
[13]C NMR (101 MHz, DMSO) spectrum of 3n:



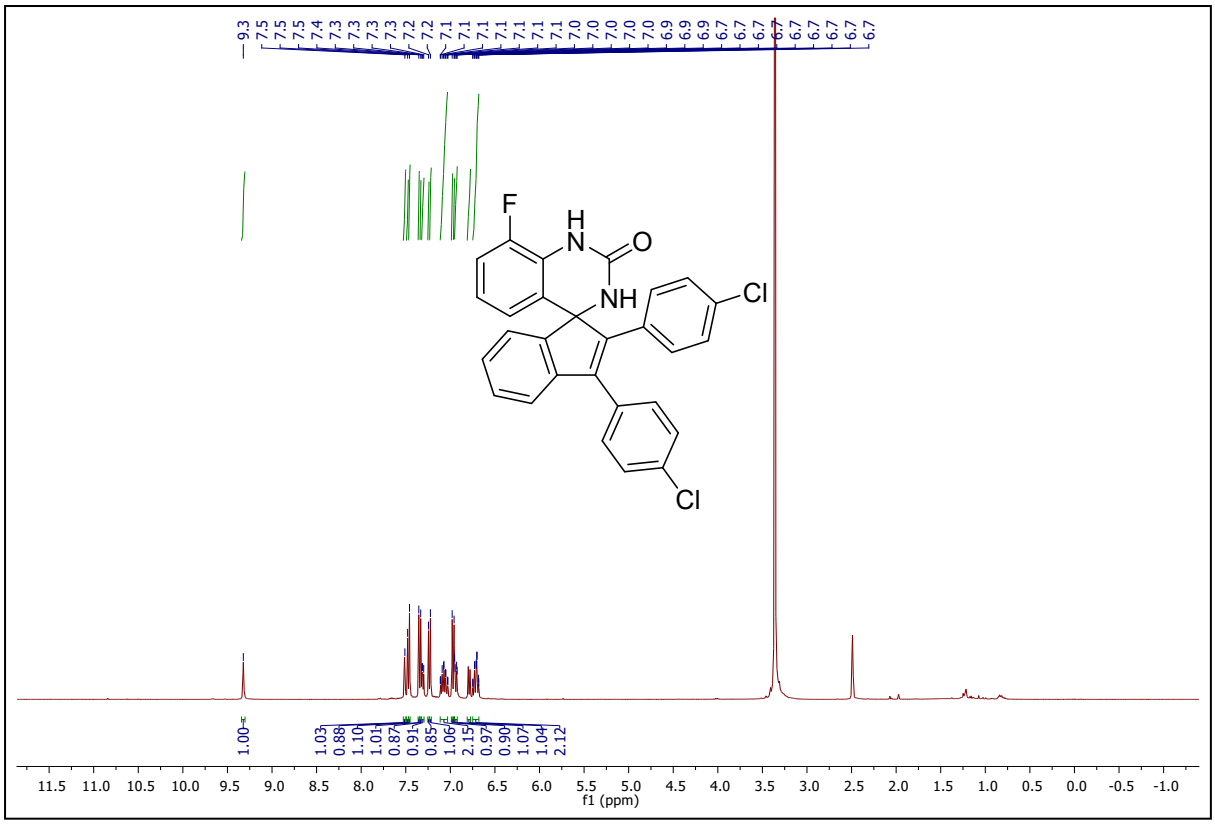
[1]H NMR (400 MHz, DMSO) spectrum of 3o:



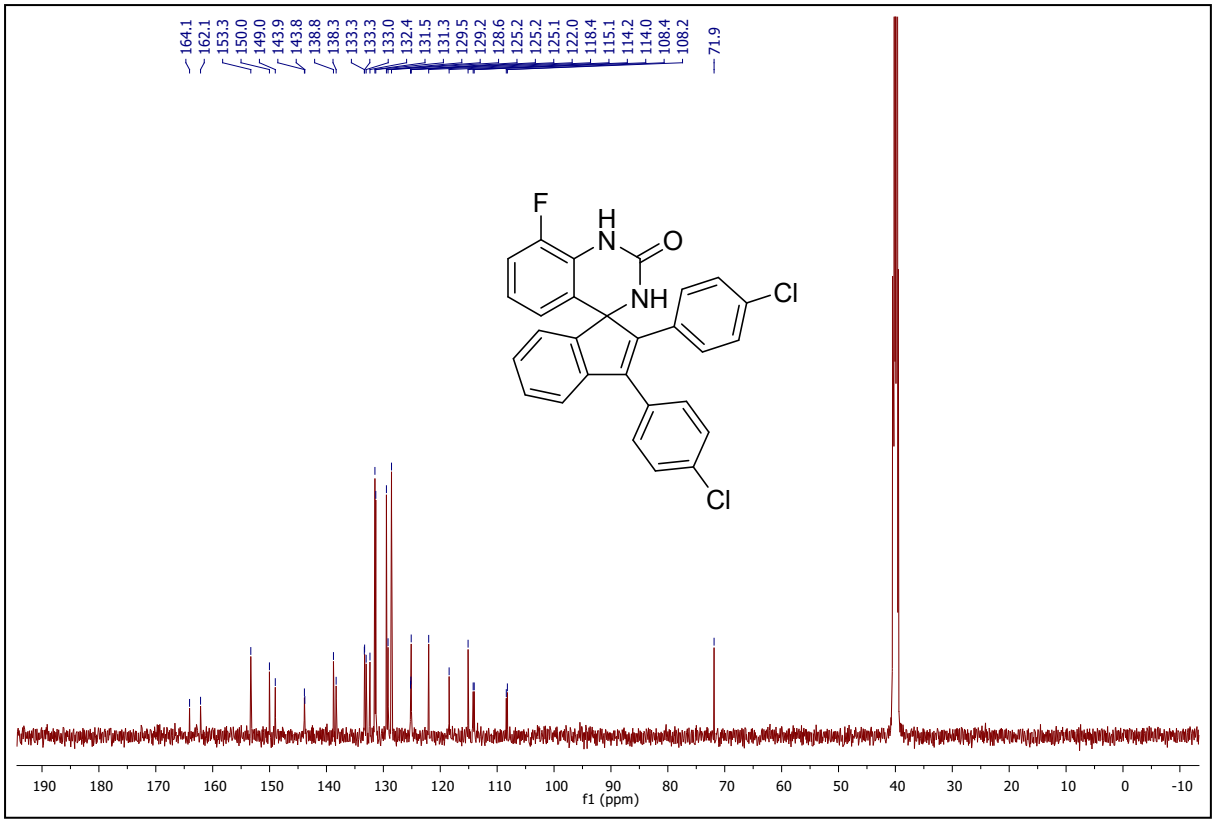
[13]C NMR (101 MHz, DMSO) spectrum of 3o:



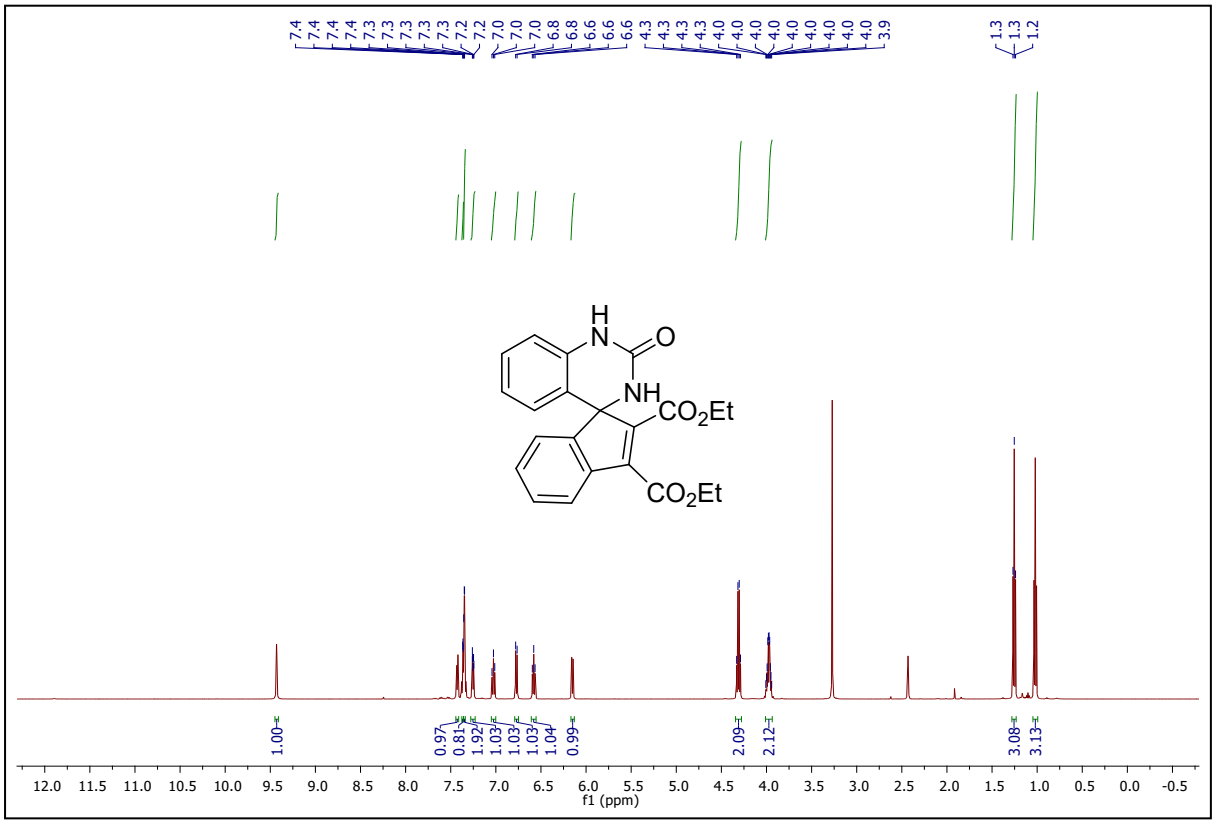
[1]H NMR (400 MHz, DMSO) spectrum of 3p:



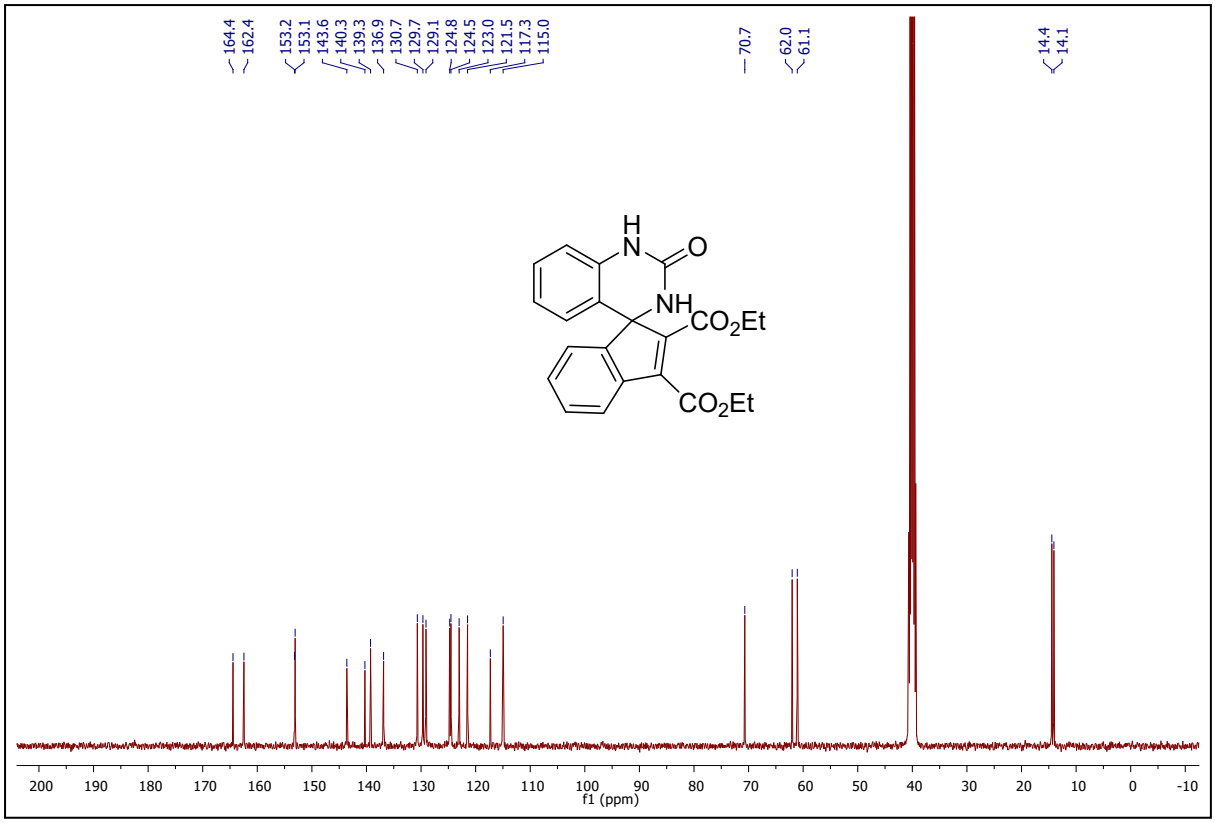
[13]C NMR (126 MHz, DMSO) spectrum of 3p:



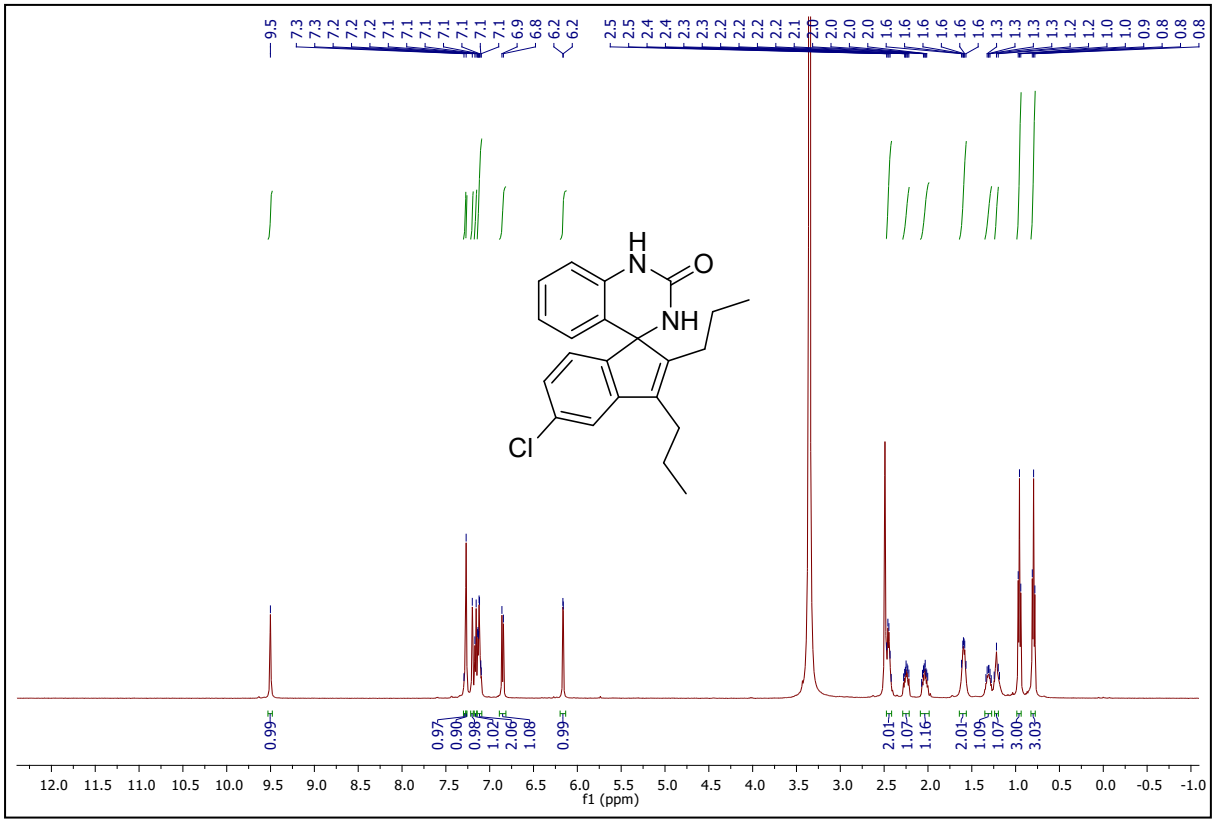
^{1}H NMR (500 MHz, DMSO) spectrum of 3q:



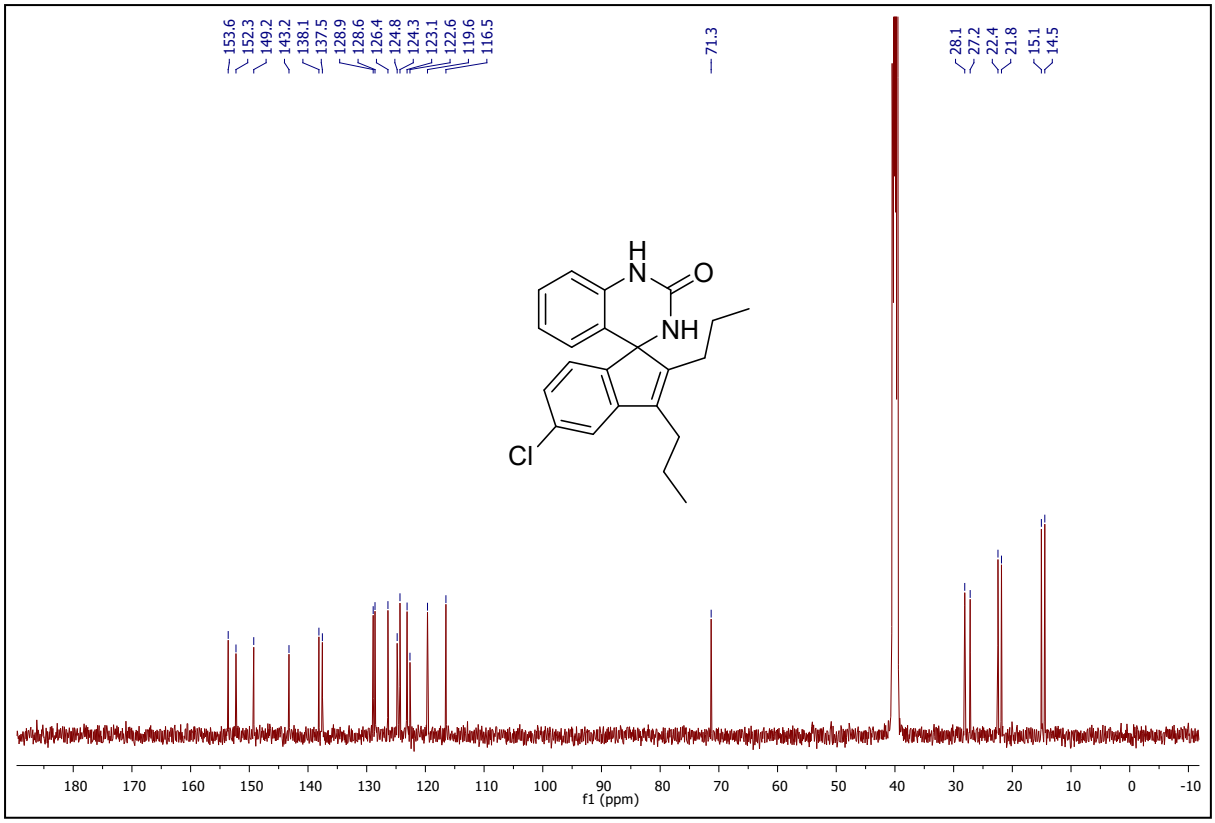
^{13}C NMR (101 MHz, DMSO) spectrum of 3q:



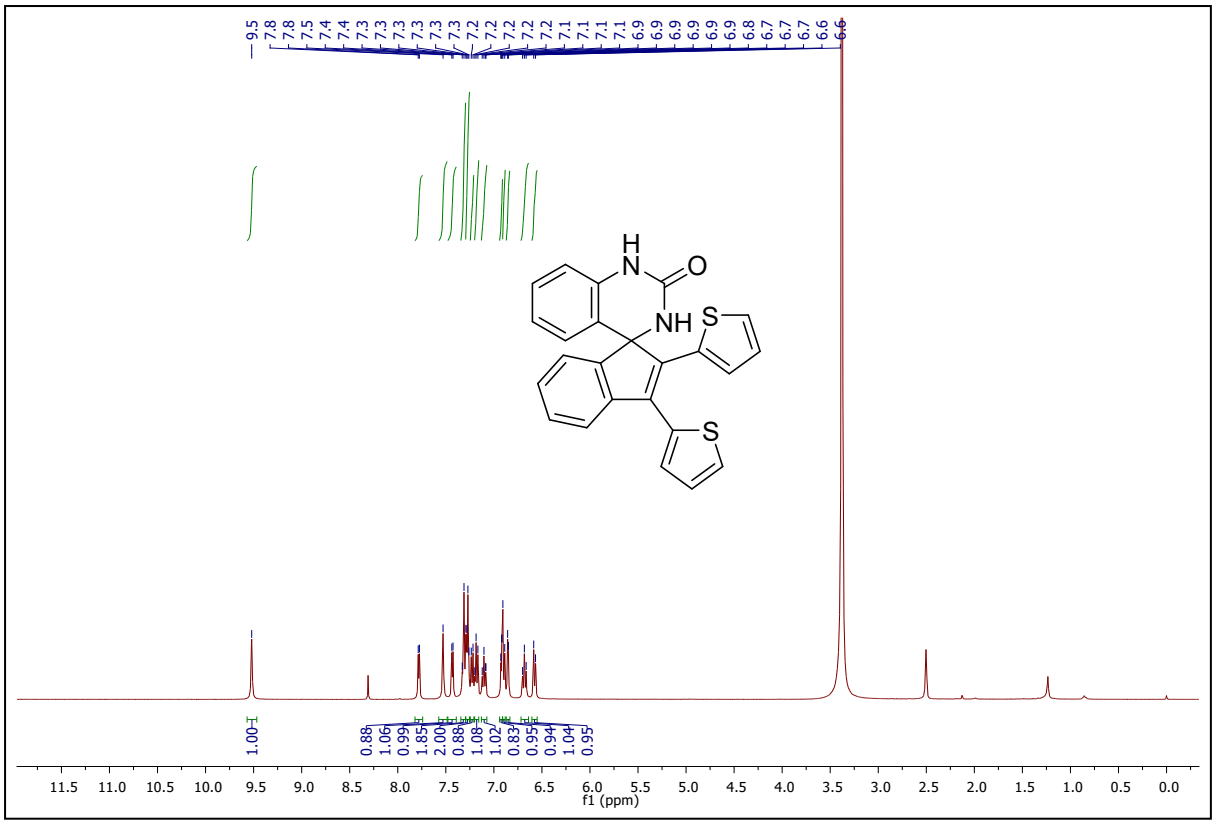
[1]H NMR (500 MHz, DMSO) spectrum of 3r:



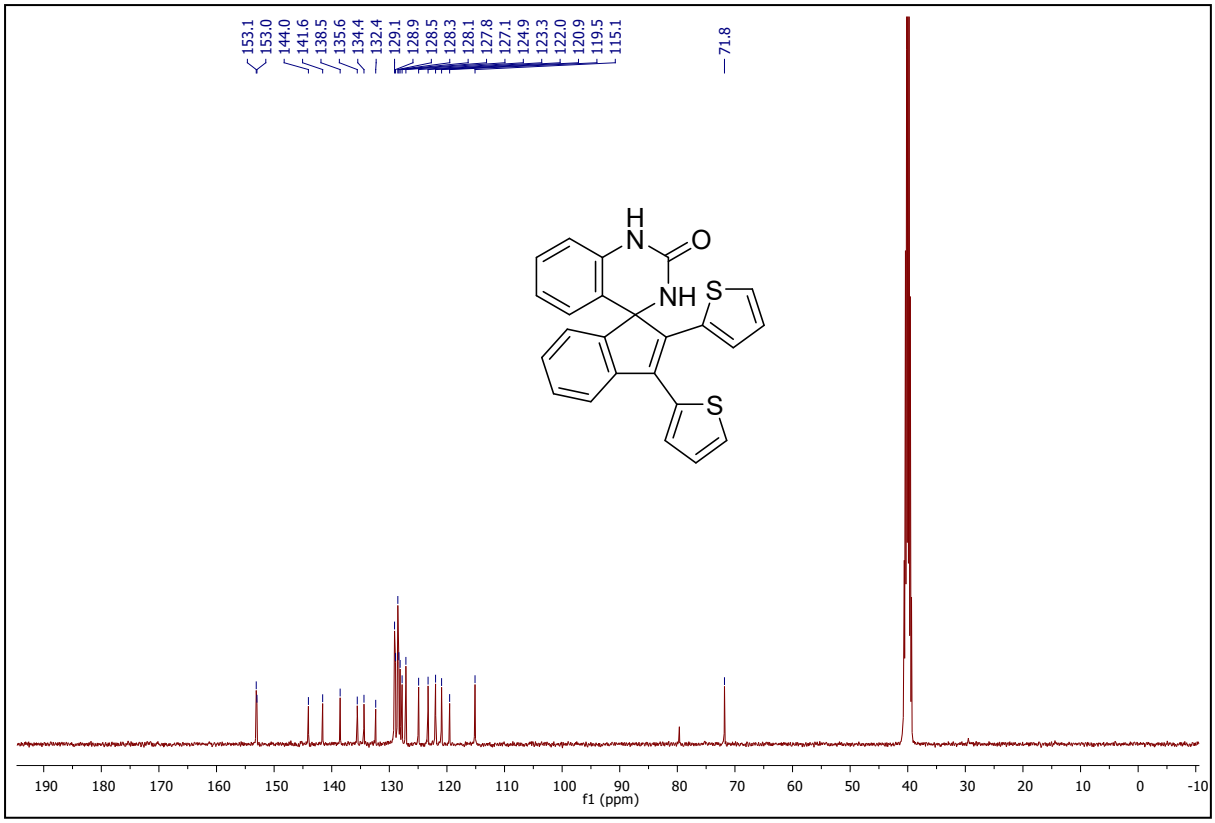
[13]C NMR (101 MHz, DMSO) spectrum of 3r:



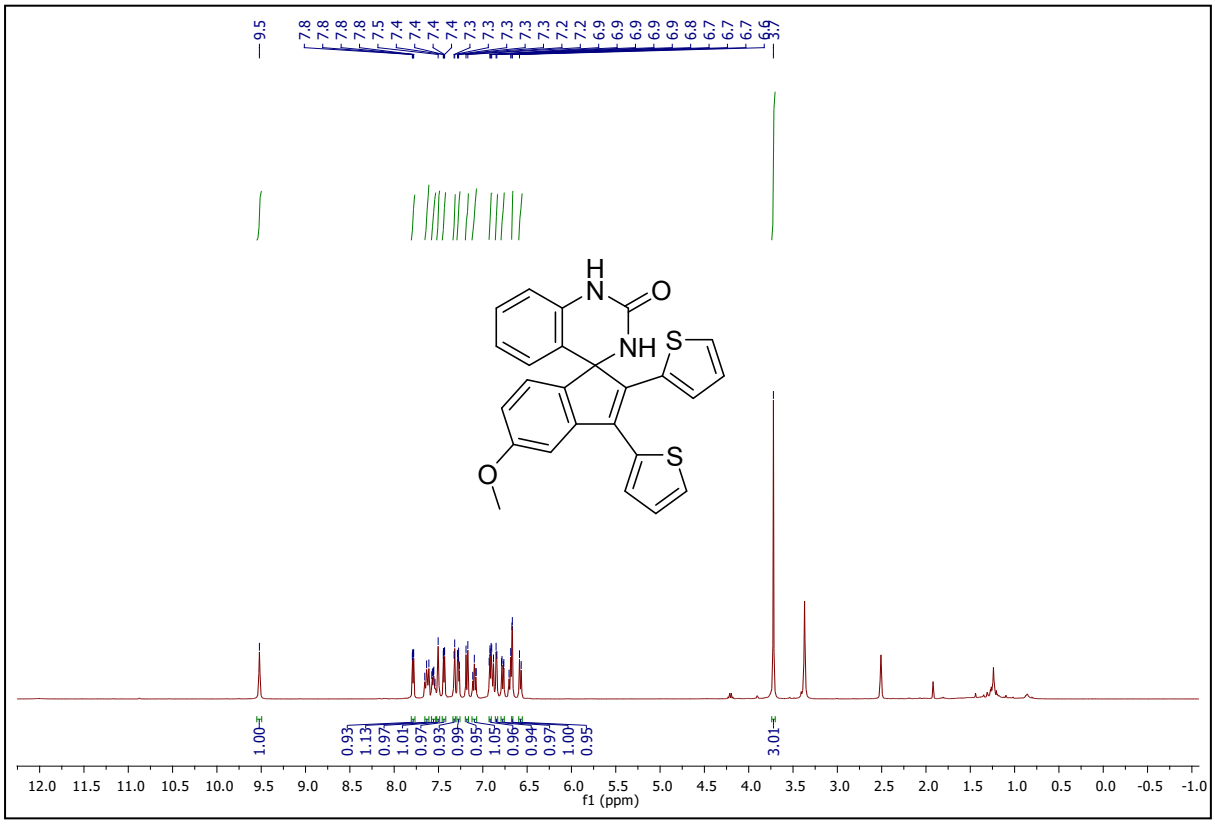
^{1}H NMR (400 MHz, DMSO $_{3}$) spectrum of 3s:



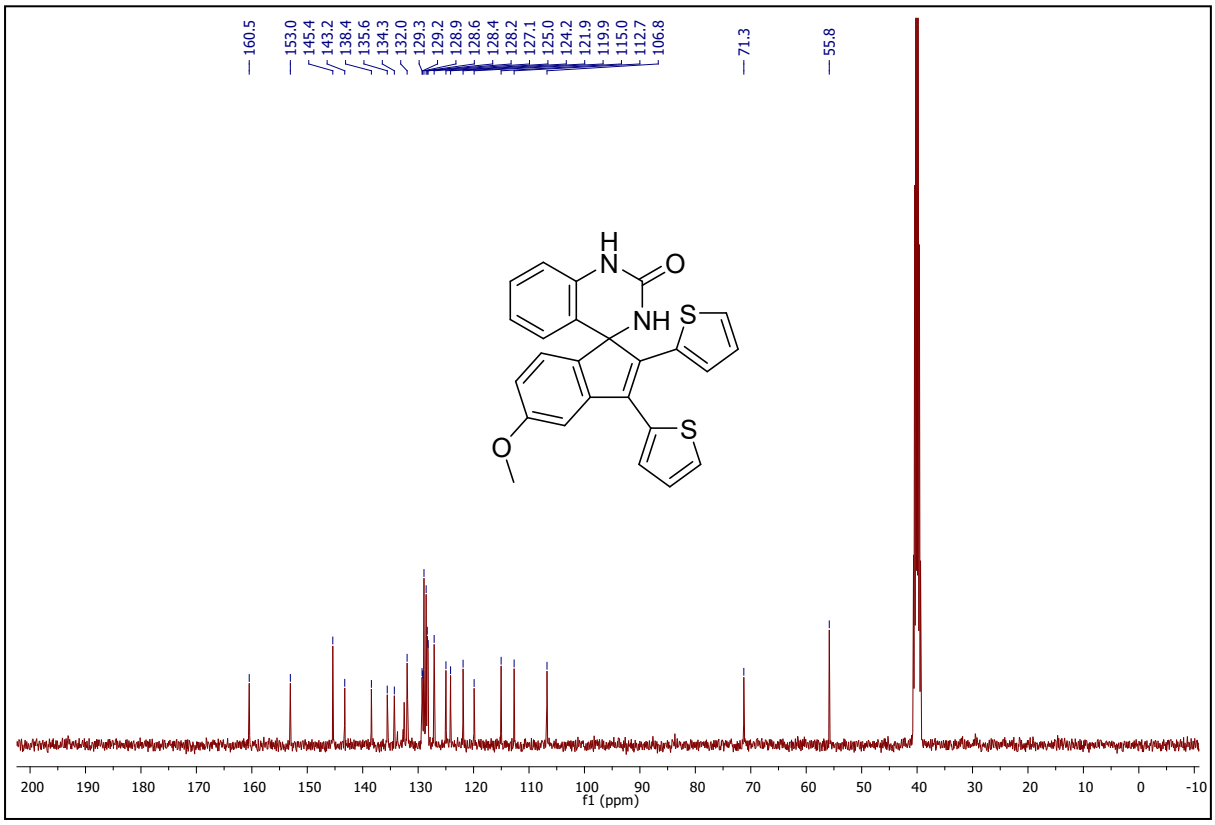
^{13}C NMR (101 MHz, DMSO) spectrum of 3s:



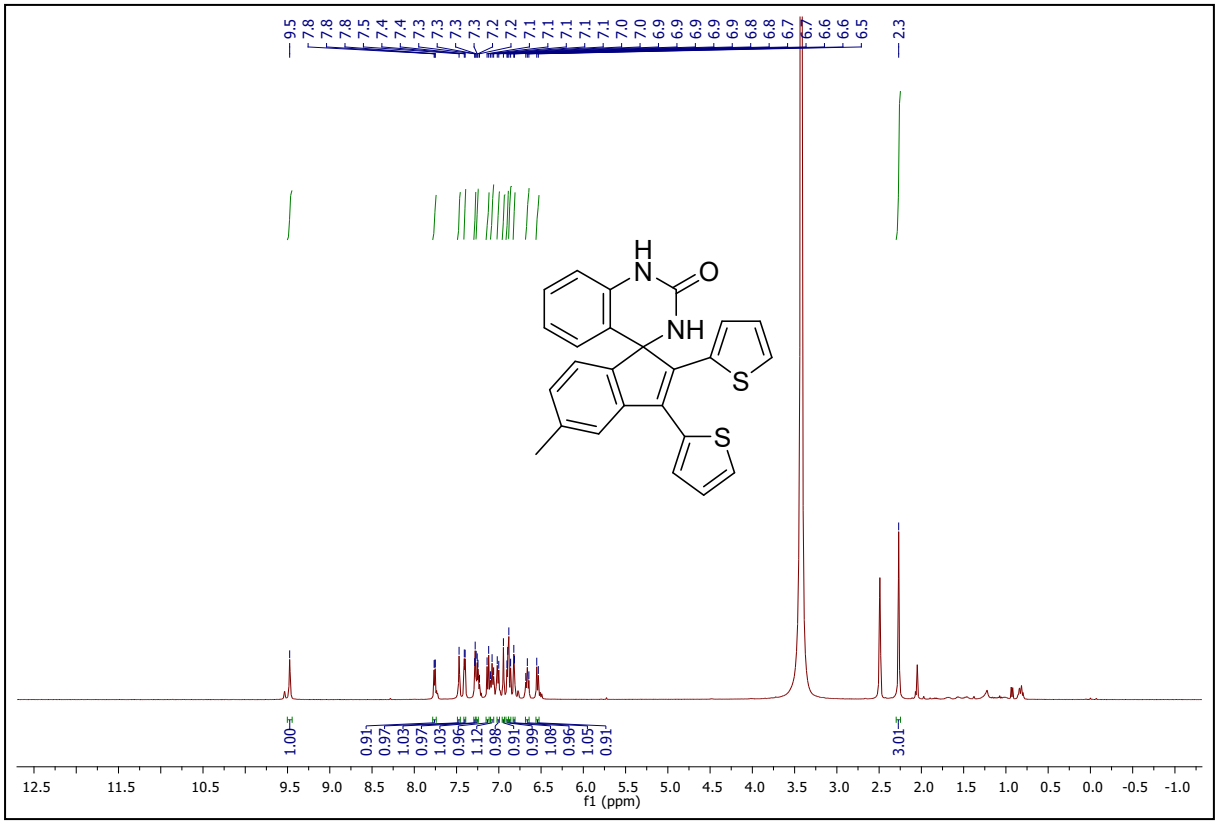
[1]H NMR (400 MHz, DMSO) spectrum of 3t:



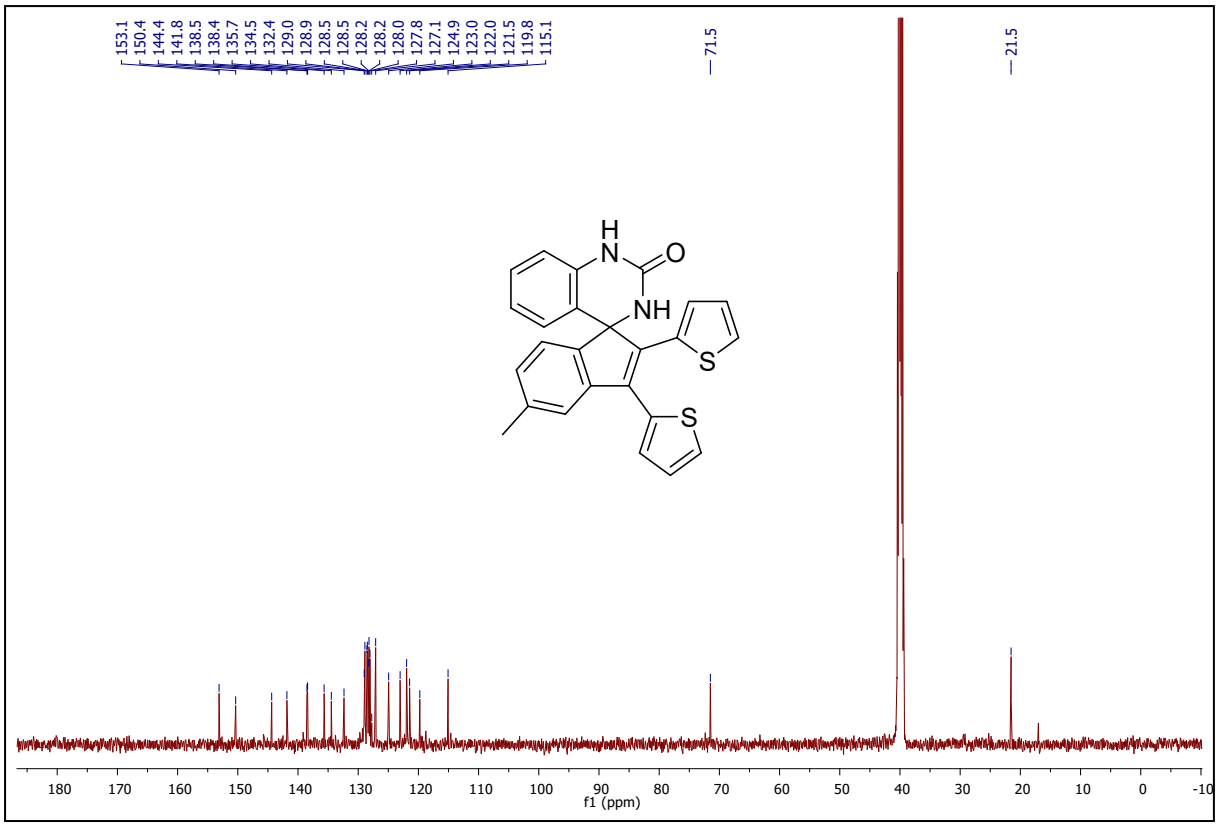
^{13}C NMR (101 MHz, DMSO) spectrum of 3t:



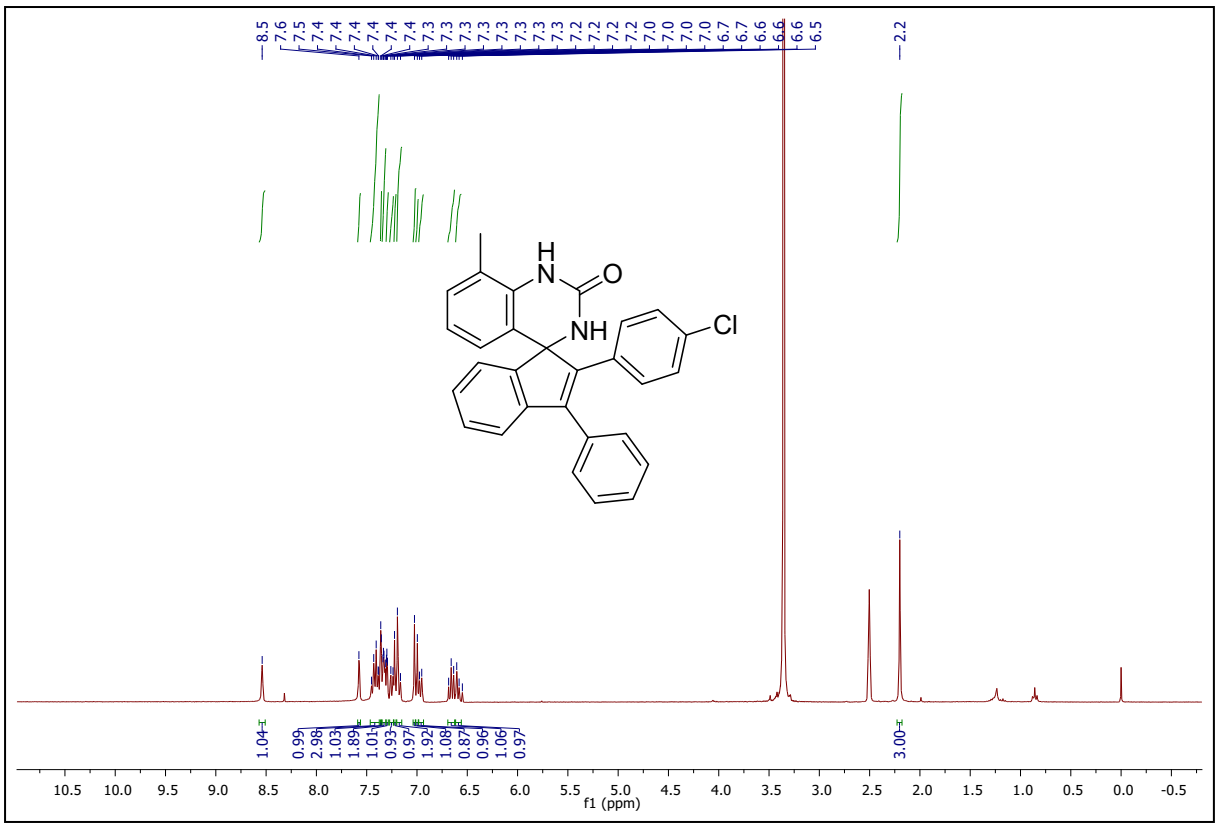
^{1}H NMR (400 MHz, DMSO) spectrum of 3u:



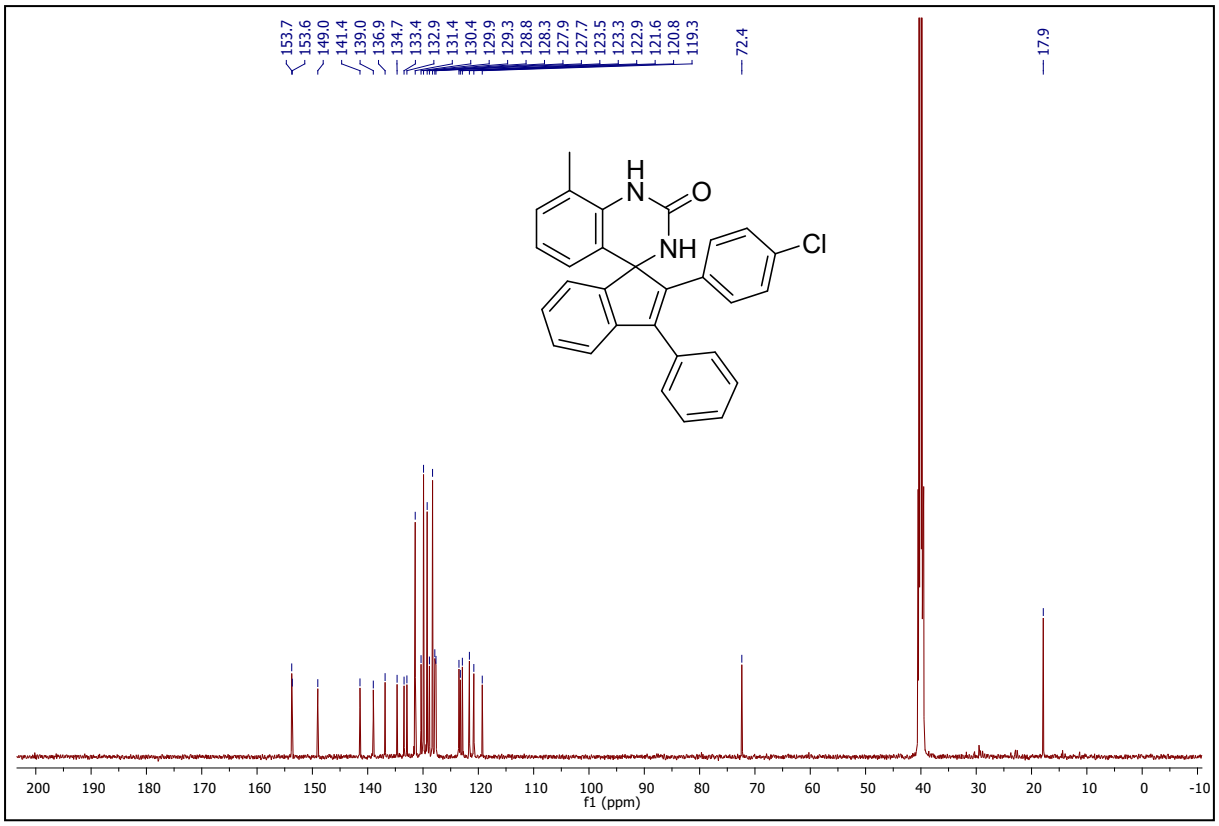
^{13}C NMR (101 MHz, DMSO) of compound 3u:



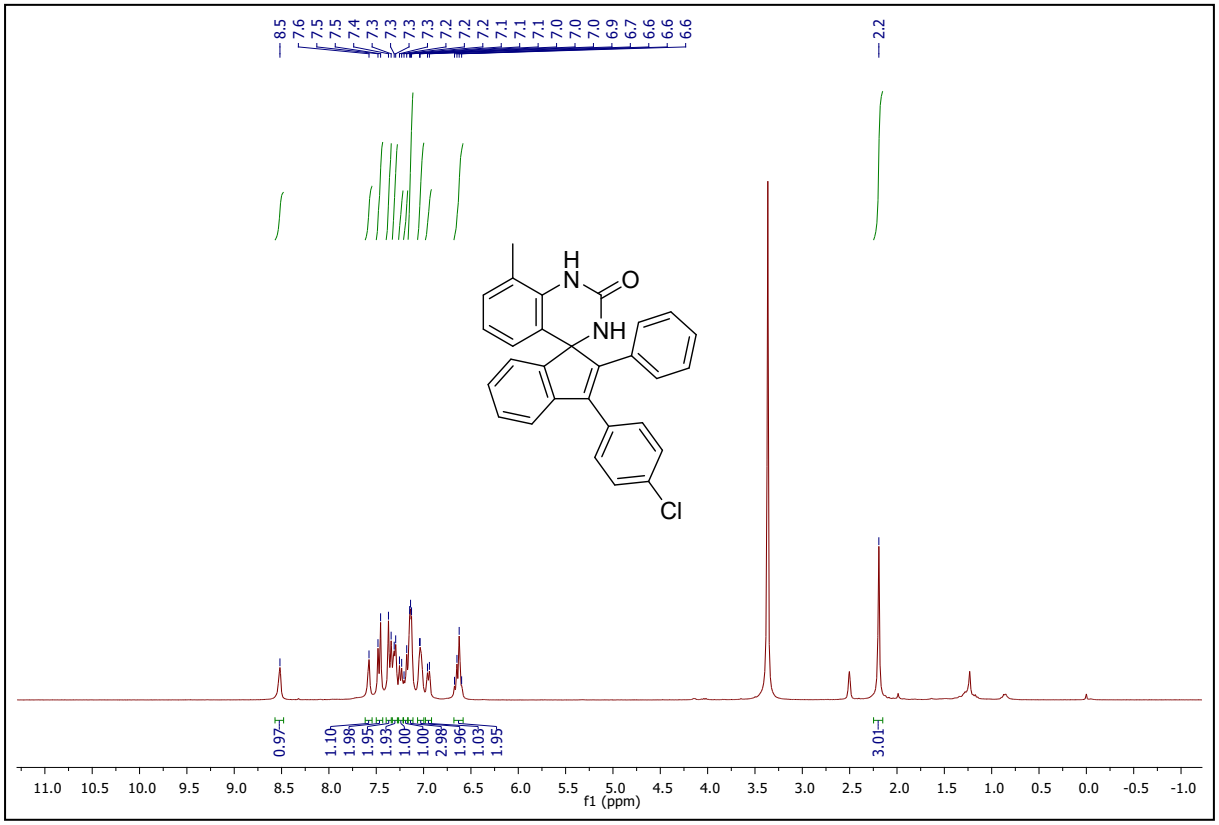
^{1}H NMR (400 MHz, DMSO) spectrum of 3v:



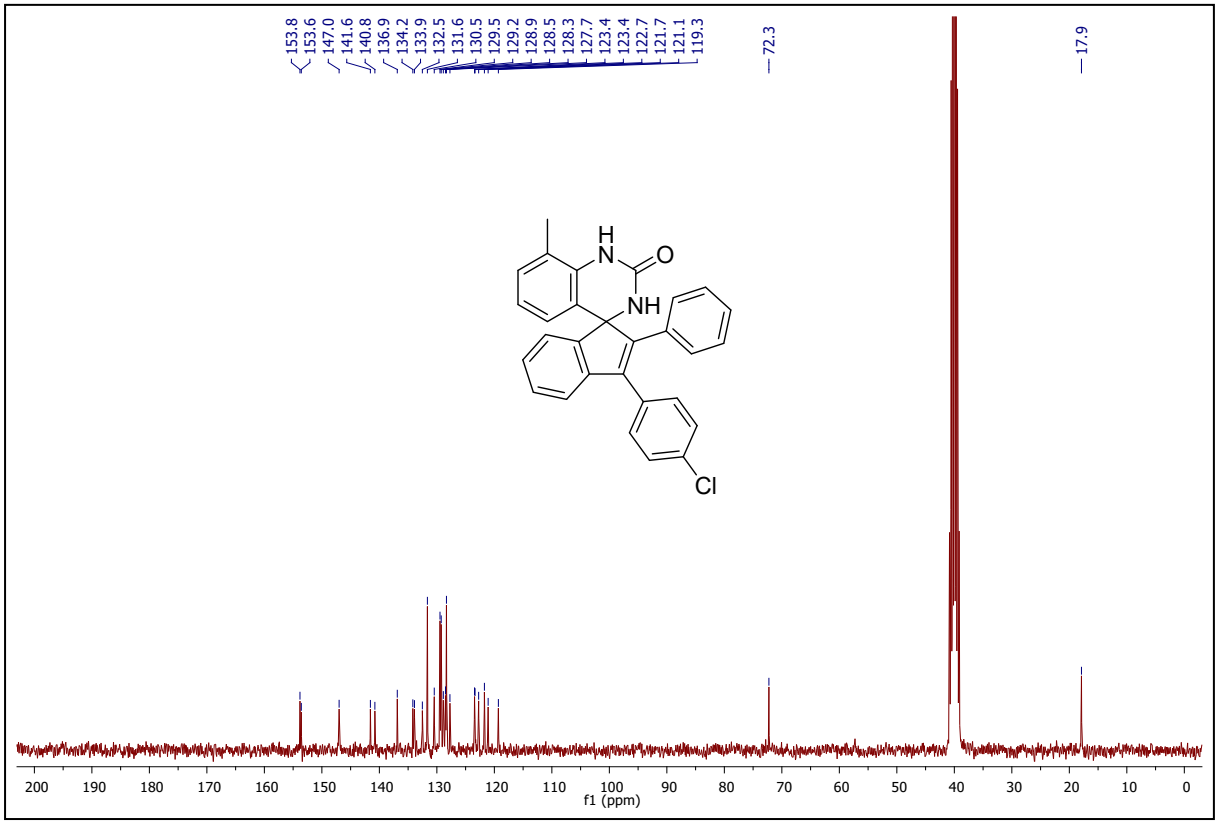
^{13}C NMR (101 MHz, DMSO) of compound 3v:



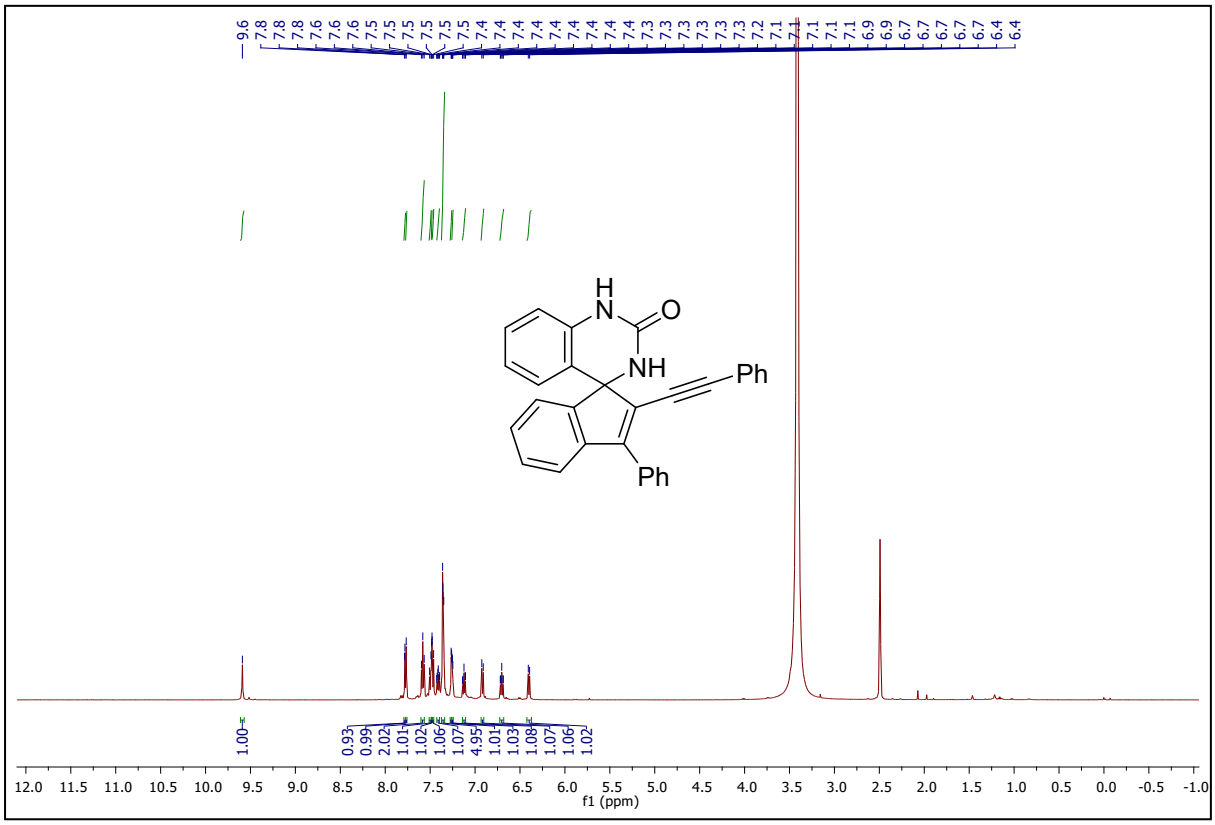
^{1}H NMR (400 MHz, DMSO) spectrum of 3v′:



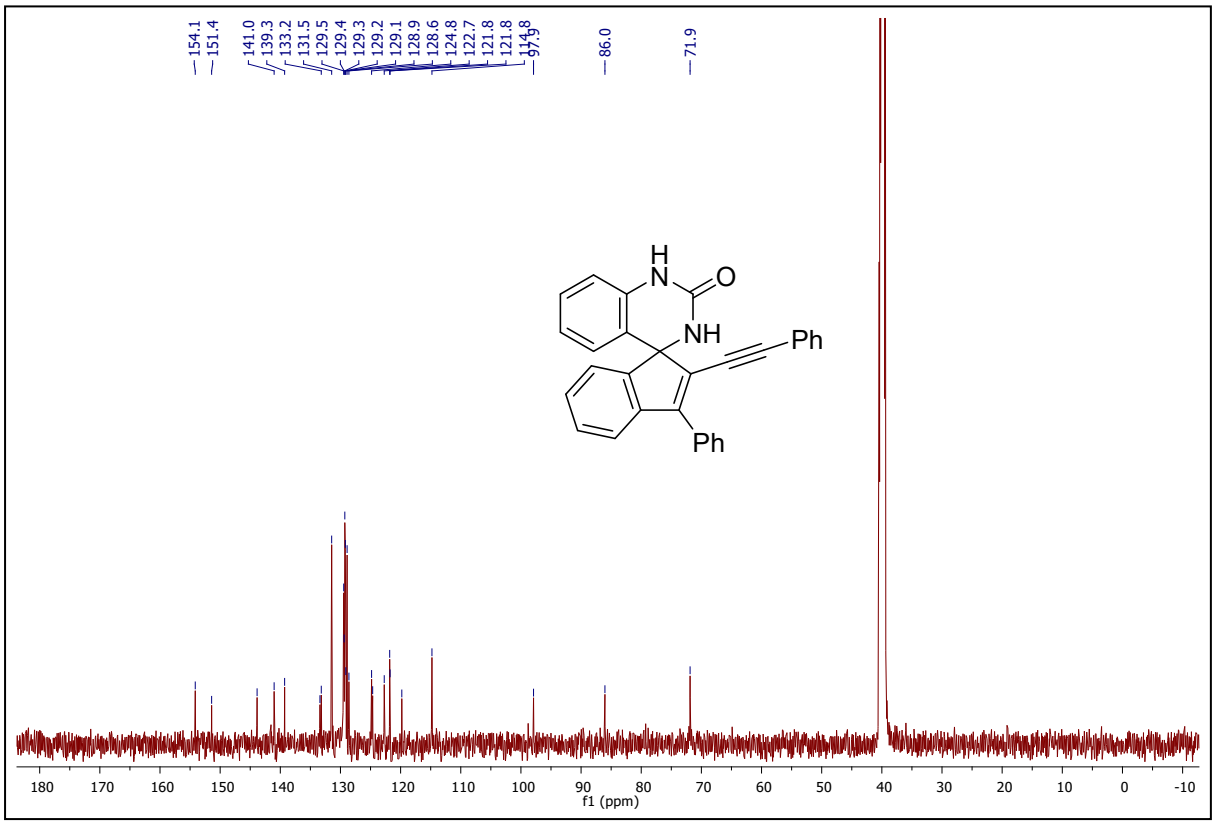
^{13}C NMR (101 MHz, DMSO) of compound 3v′:



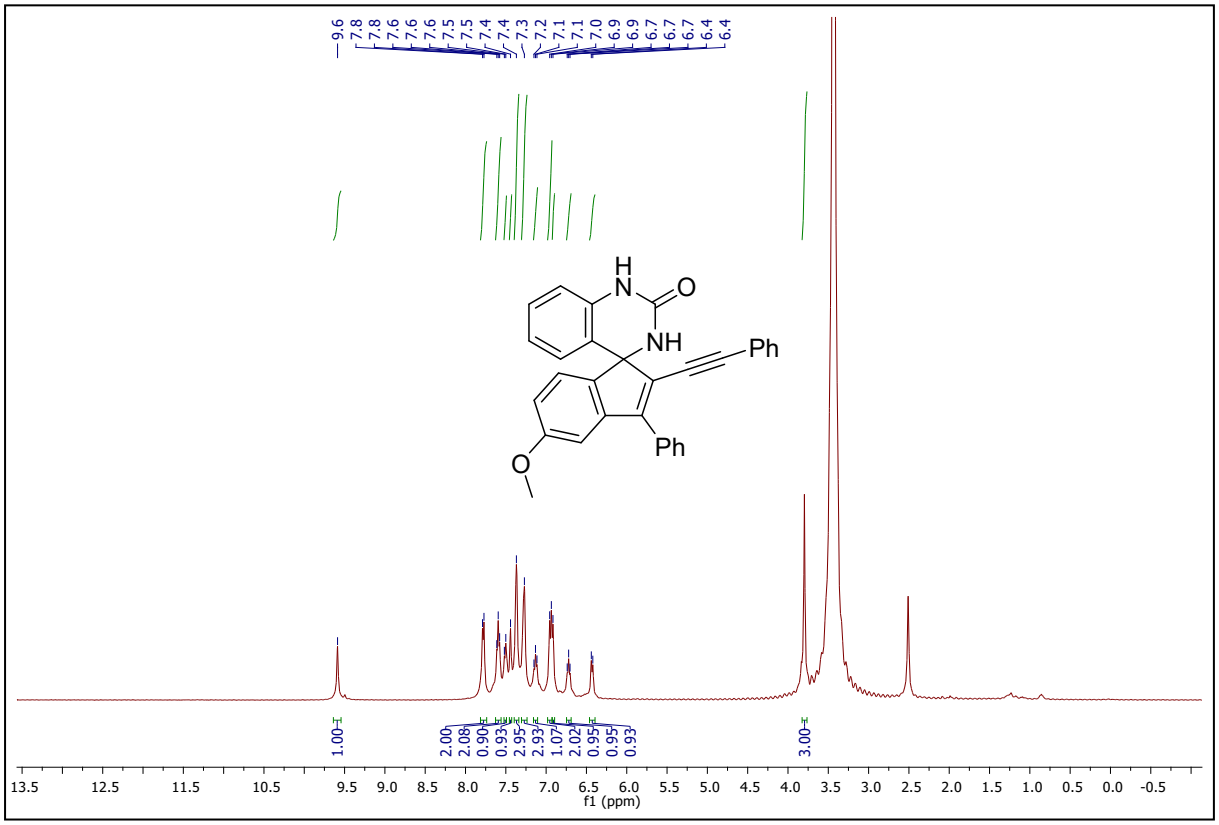
^{1}H NMR (500 MHz, DMSO) spectrum of 3w:



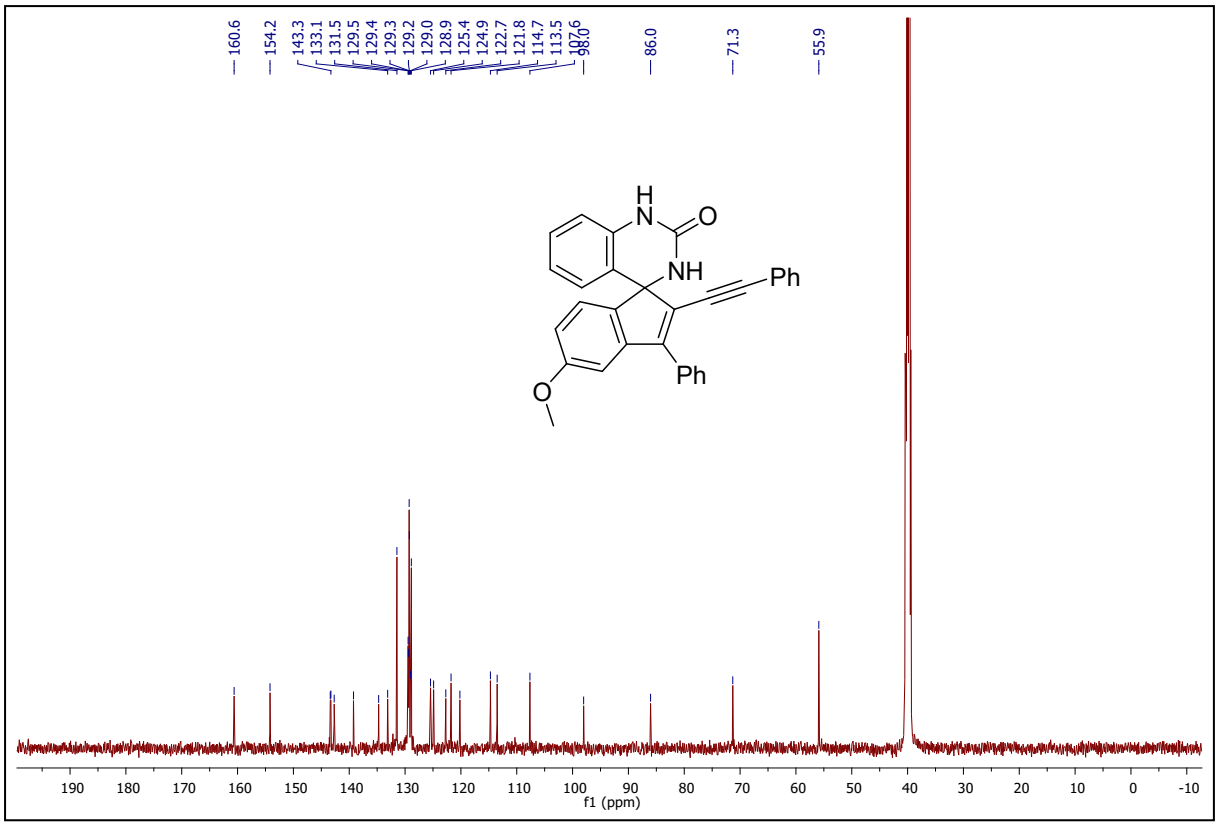
^{13}C NMR (101 MHz, DMSO) of compound 3w:



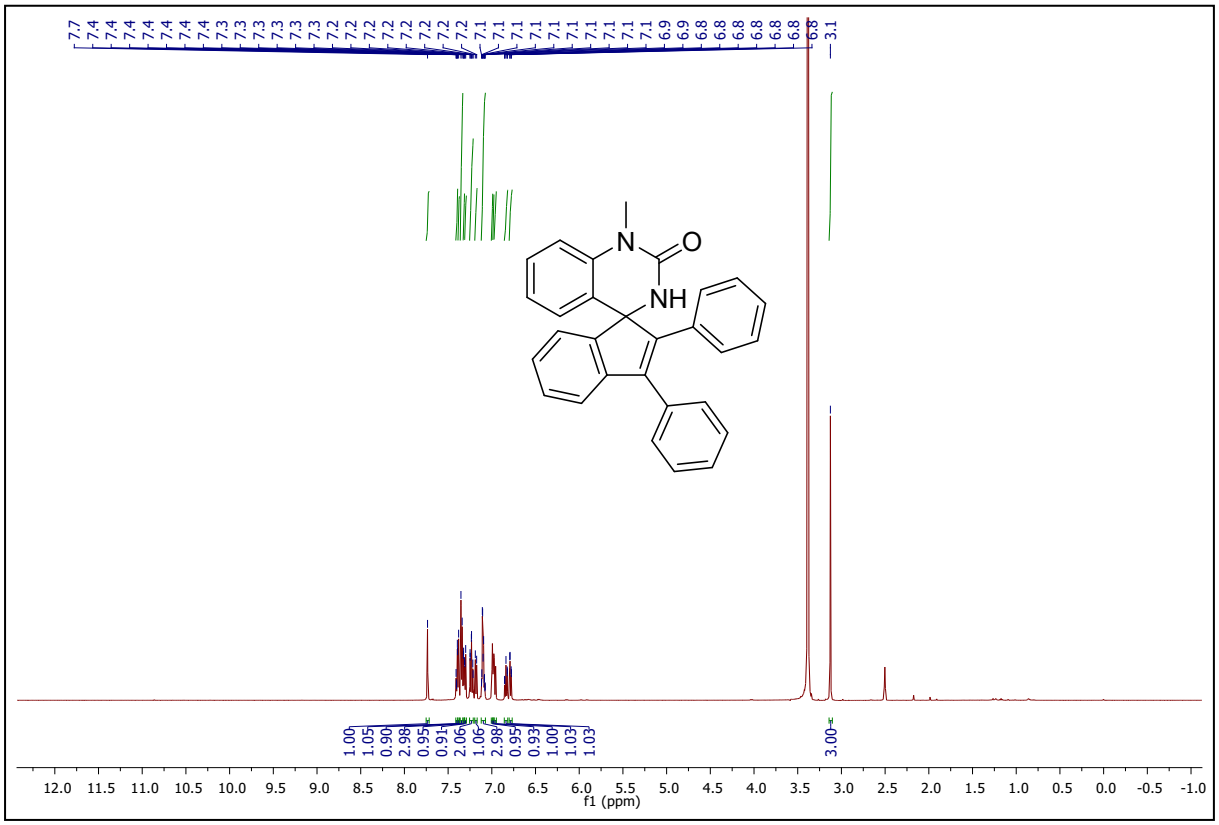
^{1}H NMR (500 MHz, DMSO) spectrum of 3x:



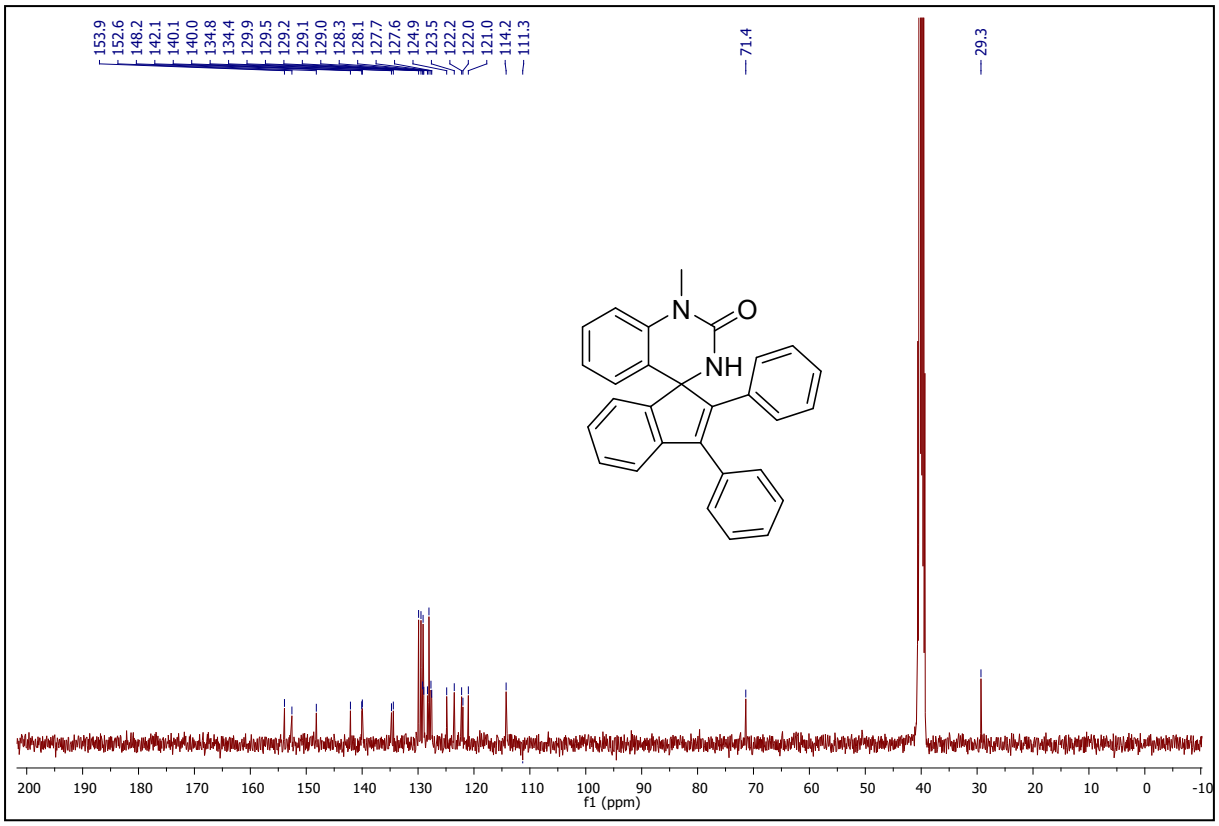
^{13}C NMR (101 MHz, DMSO) of compound 3x:



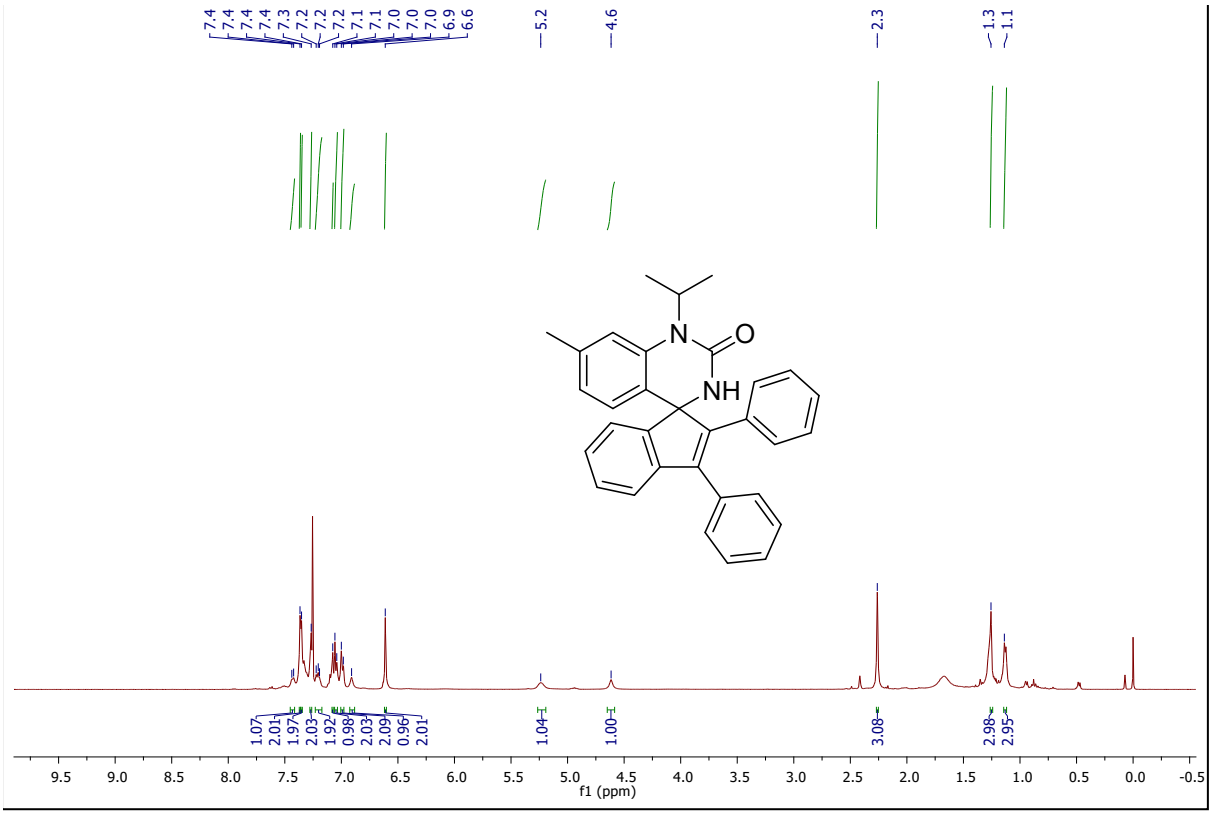
^{1}H NMR (300 MHz, DMSO) spectrum of 3y:



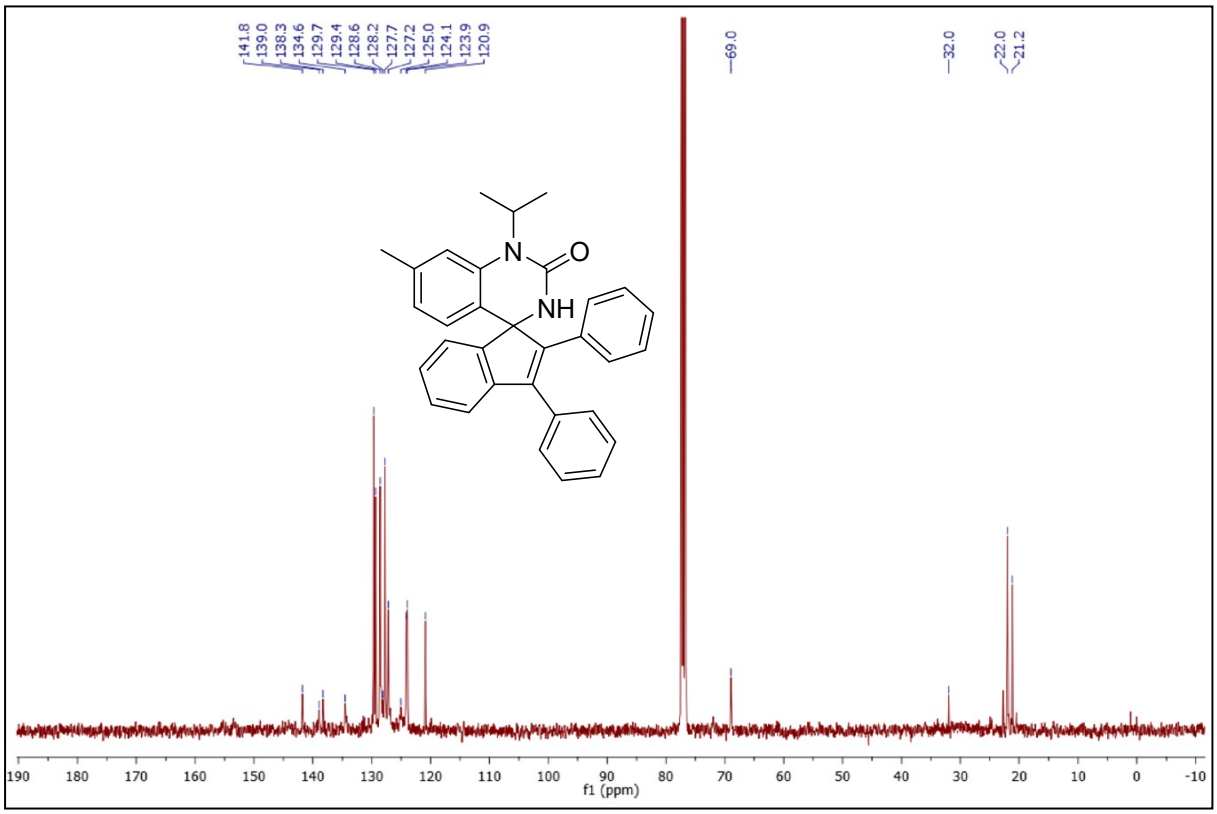
^{13}C NMR (101 MHz, DMSO) of compound 3y:



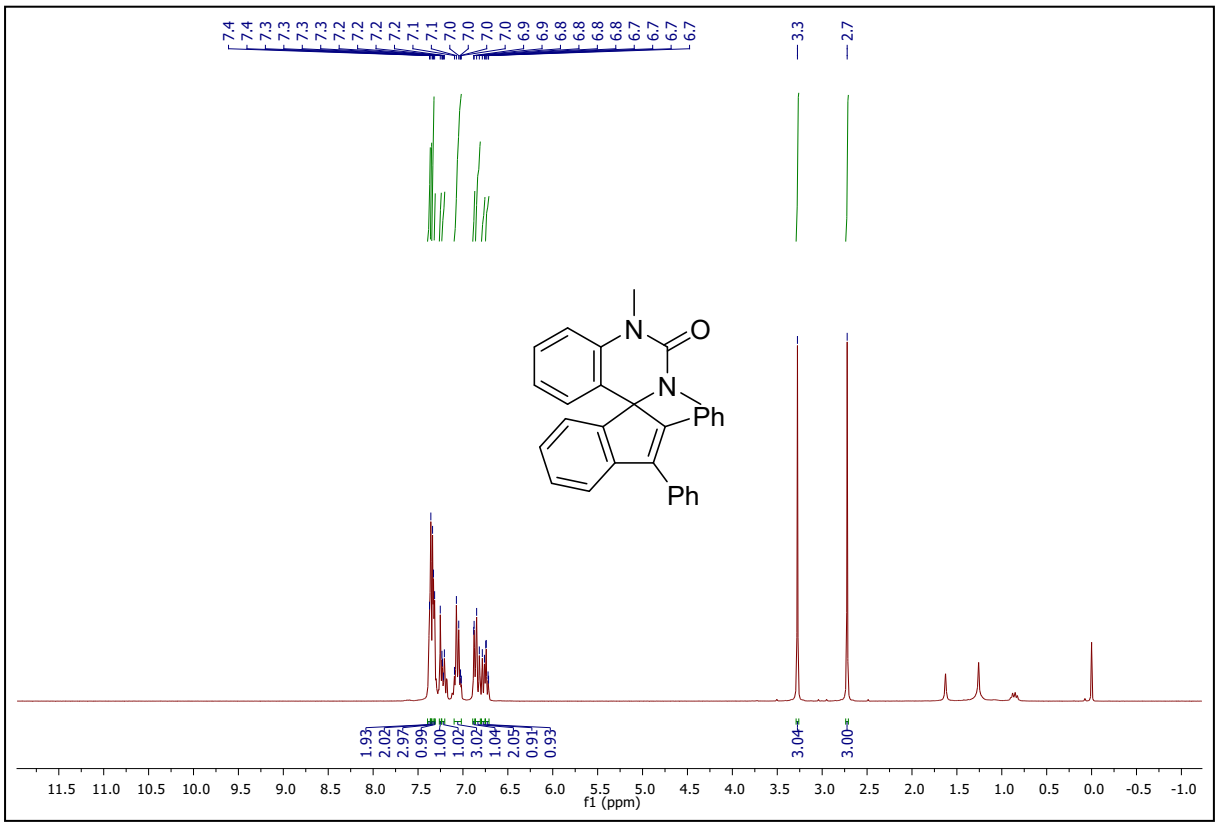
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of 3z:



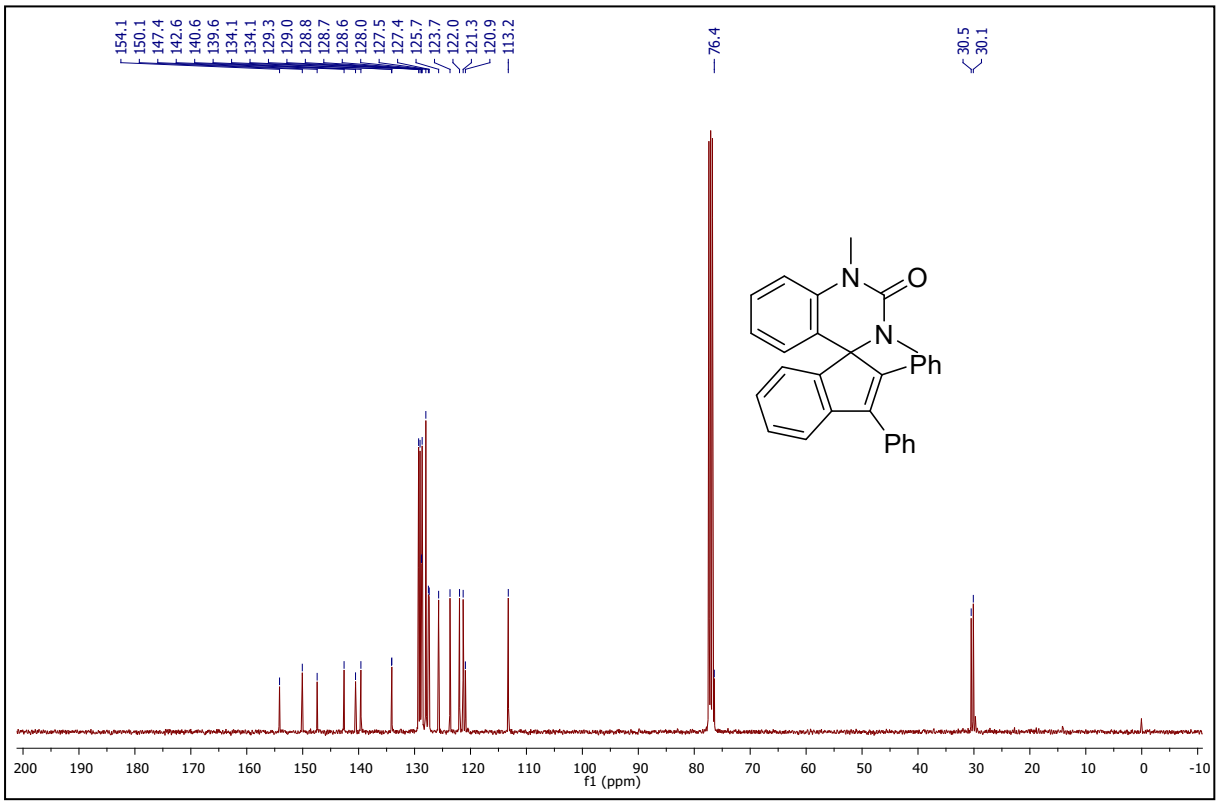
^{13}C NMR (101MHz, $CDCl_3$) spectrum of 3z:



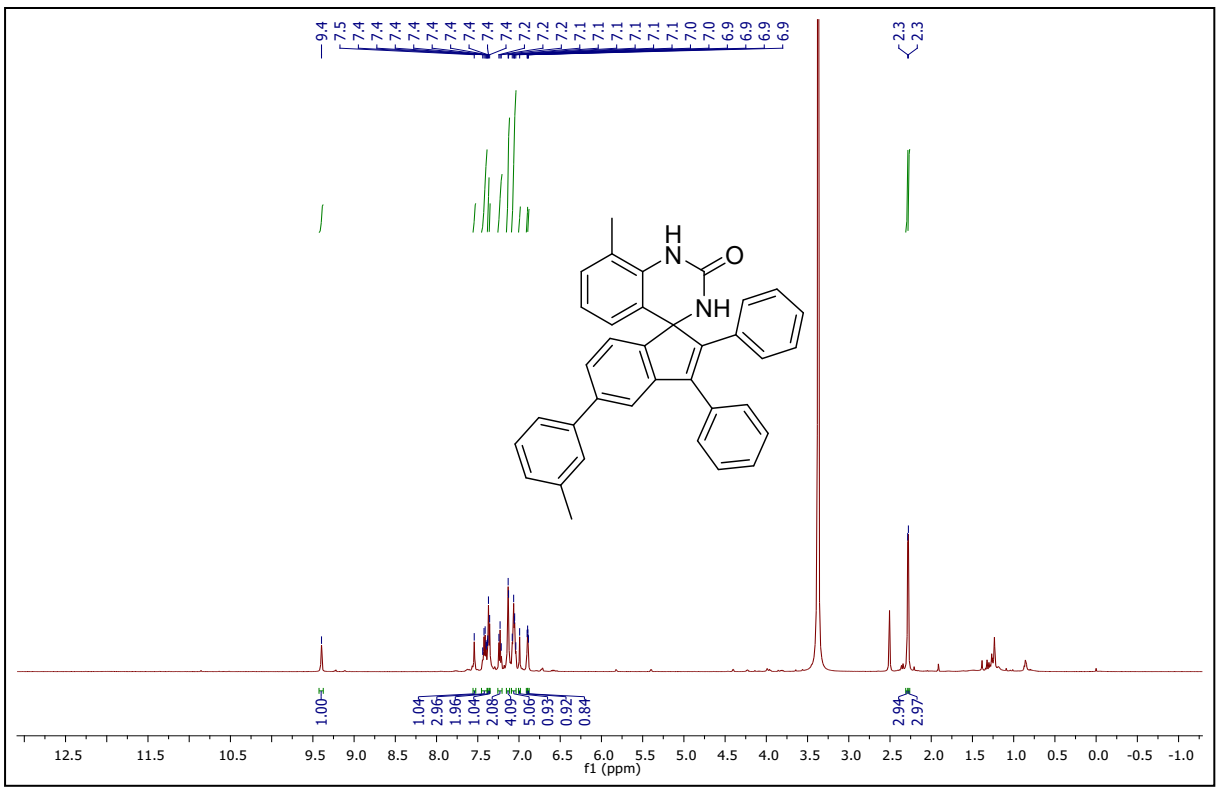
^{1}H NMR (300 MHz, $CDCl_3$) spectrum of 4a:



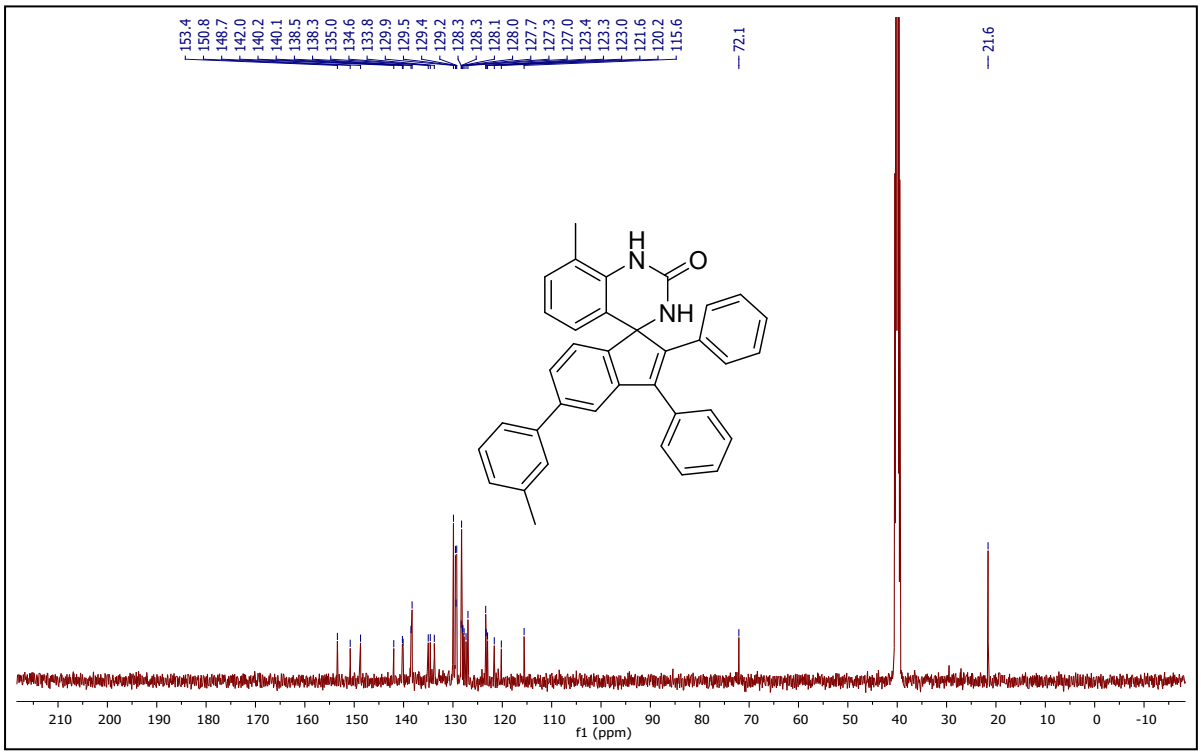
^{13}C NMR (101 MHz, $CDCl_3$) of compound 4a:



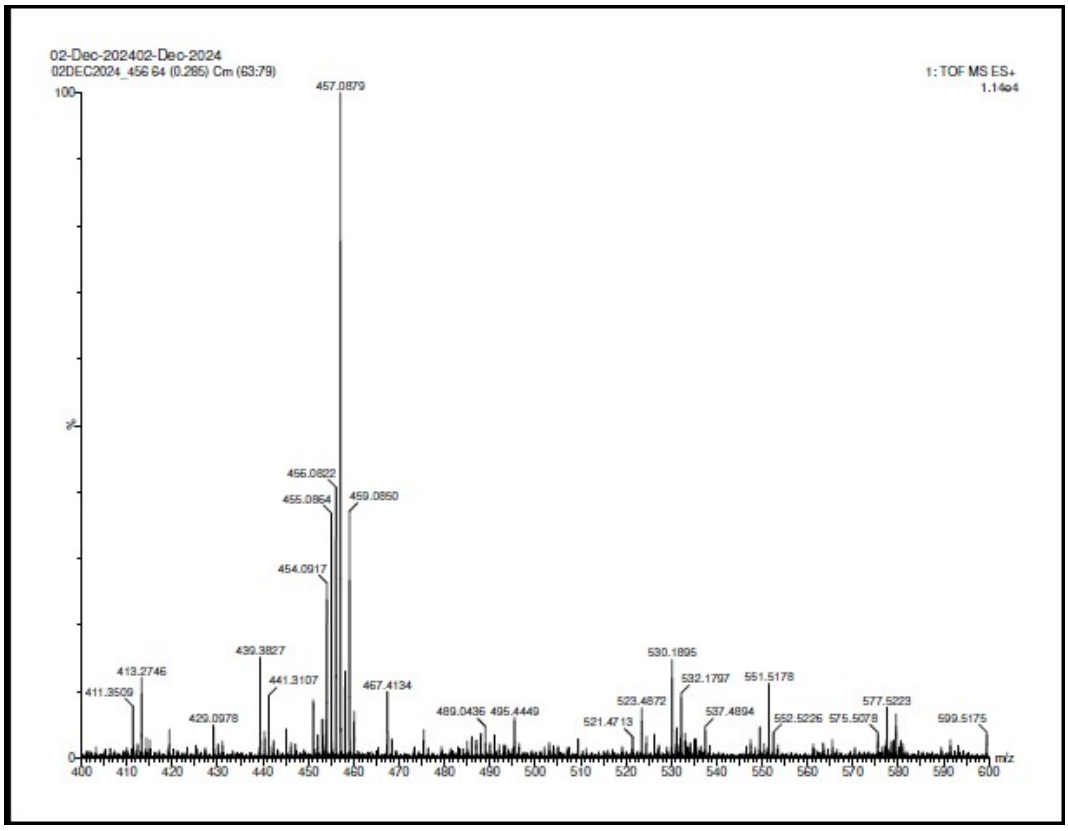
^{1}H NMR (400 MHz, $CDCl_3$) spectrum of 4b:



^{13}C NMR (101 MHz, DMSO) of compound 4b:



HRMS spectra of metallocycle



Mass	Calc. Mass	mDa	PPM	DBE	formula
457.0879	457.0854	2.5	5.5	14.5	$C_{24}H_{23}N_2ORu$

X-ray Crystallography.

X-ray data for the compound was collected at room temperature on a Bruker D8 QUEST instrument with an IμS Mo microsource (λ = 0.7107 Å) and a PHOTON-III detector. The raw data frames were reduced and corrected for absorption effects using the Bruker Apex 3 software suite programs [1]. The structure was solved using intrinsic phasing method [2] and further refined with the SHELXL [2] program and expanded using Fourier techniques. Anisotropic displacement parameters were included for all non-hydrogen atoms. The N-H atoms were located in the difference Fourier map and its positions and isotropic displacement parameters were refined. All C bound H atoms were positioned geometrically and treated as riding on their parent C atoms [C-H = 0.93-0.97 Å, and Uiso(H) = 1.5Ueq(C) for methyl H or 1.2Ueq(C) for other H atoms].

Crystal structure determination of 3v

Crystal Data for $C_{29}H_{21}N_2OCl$ (M =448.93 g/mol): triclinic, space group P-1 (no. 2), a = 9.8411(19) Å, b = 10.4582(19) Å, c = 10.930(2) Å, α = 93.183(6)°, β = 93.558(7)°, γ = 90.622(7)°, V = 1120.9(4) Å 3 , Z = 2, T = 294.15 K, μ(MoKα) = 0.196 mm -1 , Dcalc = 1.330 g/cm 3 , 22507 reflections measured (4.148° ≤ 2Θ ≤ 56.7°), 5553 unique (R int = 0.0485, R sigma = 0.0530) which were used in all calculations. The final R 1 was 0.0491 (I > 2σ(I)) and wR 2 was 0.1480 (all data). CCDC 2420260 deposition number contains the supplementary crystallographic data for this paper which can be obtained free of charge at https://www.ccdc.cam.ac.uk/structures/

1. Bruker (2016). APEX3, SAINT and SADABS. Bruker AXS, Inc., Madison, Wisconsin, USA.
2. Sheldrick G. M. (2015).ActaCrystallogr C71: 3-8.