Supporting Information

Multicomponent Domino Synthesis of 6-Azabicyclo[3.2.1]octanes and Their Cascade Ring Expansion to 2-Azabicyclo[4.3.1]decanes

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I. General Information

All reagents were purchased from Alfa Aesar and Sigma-Aldrich were used without further purification unless otherwise specified. Melting points of all the synthesized compounds were determined on an electrothermal melting point apparatus using an open capillary tube and are uncorrected. For all the prepared compounds, ^{1}H NMR (400 MHz) and ^{13}C NMR (100 MHz) spectra were recorded on Bruker Avance 400 MHz NMR spectrometer with tetramethylsilane as the internal reference. ESI-mass spectra were taken on an Agilent 6530B Q-TOF mass spectrometer. The solvent used for photophysical studies were of spectral grade. Data collection was performed with a Xcalibur, Eos diffractometer. The crystal was kept at 298 K during data collection. Using Olex2, the structure was solved with the ShelXS structure solution program using Direct Methods and refined with the ShelXL [3] refinement package using Least Squares minimization. The single crystal of **5a** and **7a** suitable for X-ray analysis were obtained by slowly volatilizing a mixed solution of MeOH and DMF in a semi-sealed glass vial bottle (5 mL) at room temperature. (10 mg of compound was dissolved in 2mL of DMF before ca. 1 mL of MeOH was added).

II. Experimental section

Starting material **3** was synthesized following the literature [1] and the procedure is given below.

1. General procedure for synthesis of compound **3**

Into a 25 mL RB flask, *o*-phthalaldehyde **1** (1 mmol), 2-cyanoacetamide **2a** (1.1 mmol) ammonium acetate (1.2 mmol) and triethylamine (20 mol%) were added successively in 10 mL of ethanol and were stirred at room temperature. Precipitate started to form in 20 minutes was monitored by TLC and left to run for about 3h for completion. Simple filtration from methanol (20mL) afforded clean product **3** in 75% yield.

2. General procedure for synthesis of compound **5**

(**5a** as an example) To a 25ml RB flask, compound **3** (1 mmol) and anisaldehyde **4** (2.1 mmol), malononitrile **2a** (2.1 mmol) and piperidine (1 mmol) were added successively in ethanol (10 mL) at room temperature The reaction mixture was stirred vigorously for 6 h and the precipitated white solid was filtered and washed with methanol/diethylether to yield the product **5a**, without any further purification needed.

3. General procedure for synthesis of compound **7**

(**7a** as an example) To a 25ml RB flask, compound **3** (1 mmol) and 4-(trifluoromethyl)benzaldehyde **4j** (3.1 mmol), malononitrile **2b** (3.1 mmol) and piperidine (2 mmol) were added successively in ethanol (10 mL) at room temperature The reaction mixture was stirred vigorously for 8 h and the precipitated shiny yellow solid was filtered and washed with methanol/diethylether to yield the pure product **7a**, without any further purification needed.

III. UV-Vis and solid emission spectra of Compound 3

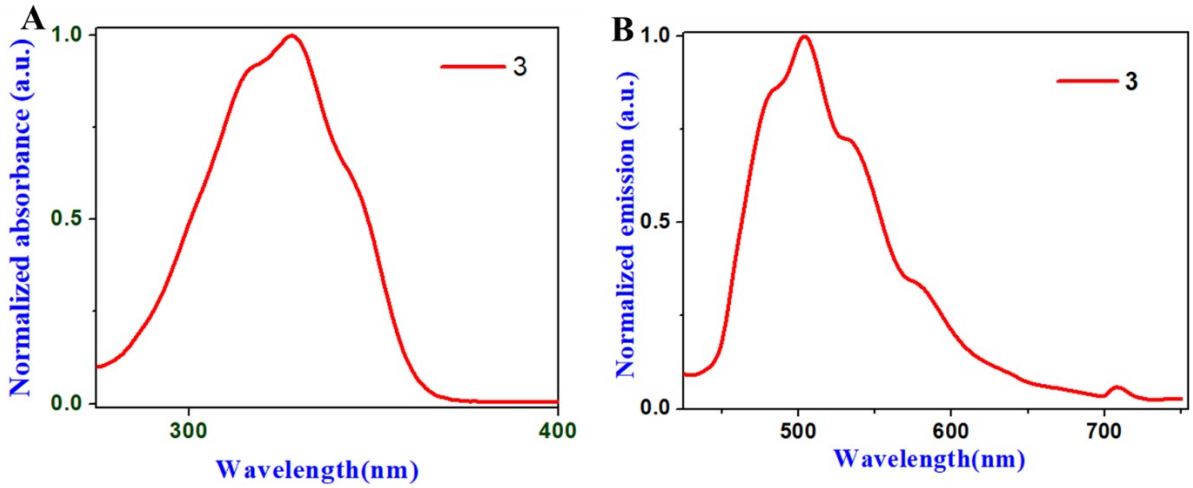


Figure S1. (**A**) UV−vis spectra of **3** measured in DMF at 2.5 μM and (**B**) Emission spectra of **3** in solid state.

IV. X-ray Crystal Data

1. X-ray Crystal Data of **3** (CCDC NO. 2212407).

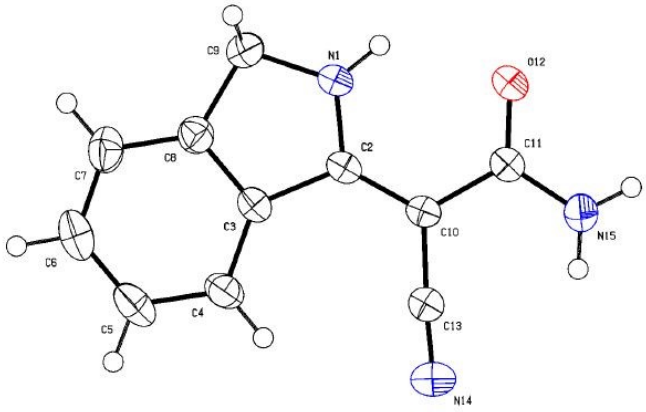


Figure S2. Molecular structure of compound **3** presented using thermal ellipsoids of the 50% probability.

Table S1. Crystal data and structure refinement for compound **3**

	Compound **3**
Empirical formula	$C_{11}H_9N_3O$
Formula weight	199.21
Temperature/K	297
Crystal system	triclinic
Space group	P-1
a/Å	7.3731(5)
b/Å	7.4734(6)
c/Å	9.4400(9)
α/°	113.196(8)
β/°	90.392(7)
γ/°	100.287(6)
Volume/Å^3	468.71(7)
Z	2
ρ_{calc}g/cm^3	1.412
μ/mm^{-1}	0.095
F(000)	208.0
Crystal size/mm^3	0.15 × 0.12 × 0.11
Radiation	MoKα (λ = 0.71073)
2Θ range for data collection/°	7.032 to 58.58
Index ranges	$-9 \le h \le 9$, $-9 \le k \le 10$, $-12 \le l \le 12$
Reflections collected	5180
Independent reflections	2186 [R_{int} = 0.0166, R_{sigma} = 0.0187]
Data/restraints/parameters	2186/0/136
Goodness-of-fit on F^2	1.057
Final R indexes [I>=2σ (I)]	R_1 = 0.0463, wR_2 = 0.1169

Final R indexes [all data]	$R_1 = 0.0537$, $wR_2 = 0.1244$
Largest diff. peak/hole / e Å^{-3}	0.22/-0.34

2. X-ray Crystal Data of **5a** (CCDC NO. 2211929).

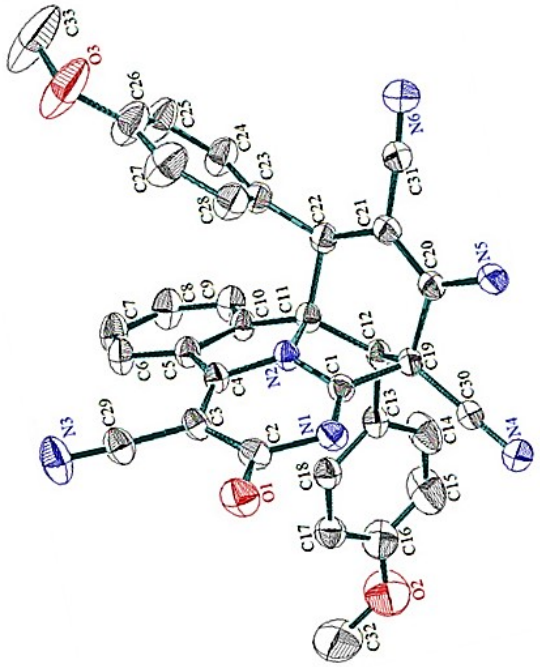


Figure S3. Molecular structure of compound **5a** presented using thermal ellipsoids of the 50% probability.

Table S2. Crystal data and structure refinement for compound **5a**

	Compound 5a
Empirical formula	$C_{24}H_{16}N_{4.36}O_{2.18}$
Formula weight	400.41
Temperature/K	295(2)
Crystal system	orthorhombic
Space group	Pccn
a/Å	27.0007(18)
b/Å	21.4303(10)
c/Å	9.5534(4)

α/°	90
β/°	90
γ/°	90
Volume/Å^3	5527.9(5)
Z	11
ρ_{calc}g/cm^3	1.323
μ/mm^{-1}	0.088
F(000)	2288.0
Crystal size/mm^3	0.52 × 0.28 × 0.2
Radiation	Mo Kα (λ = 0.71073)
2Θ range for data collection/°	8.184 to 58.416
Index ranges	-36 ≤ h ≤ 28, -28 ≤ k ≤ 28, -12 ≤ l ≤ 11
Reflections collected	29729
Independent reflections	6745 [R_{int} = 0.0421, R_{sigma} = 0.0323]
Data/restraints/parameters	6745/0/389
Goodness-of-fit on F^2	1.017
Final R indexes [I>=2σ (I)]	R_1 = 0.0500, wR_2 = 0.1076
Final R indexes [all data]	R_1 = 0.0824, wR_2 = 0.1232
Largest diff. peak/hole / e Å^{-3}	0.18/-0.18

3. X-ray Crystal Data of **7a** (CCDC NO. 2212386).

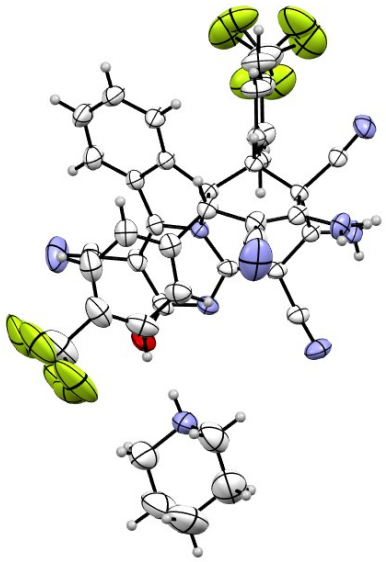


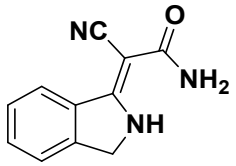
Figure S4. Molecular structure of compound **7a** presented using thermal ellipsoids of the 50% probability.

Table S3. Crystal data and structure refinement for compound **7a**

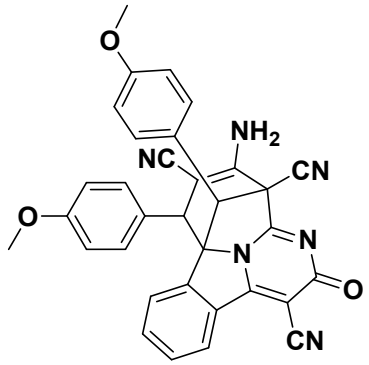
	Compound 7a
Empirical formula	C36 H18 F6 N8 O
Formula weight	923.92
Temperature/K	296.15
Crystal system	triclinic
Space group	P-1
a/Å	13.9052(8)
b/Å	13.9260(9)
c/Å	14.7055(13)
α/°	117.811(2)
β/°	105.207(3)
γ/°	99.479(2)
Volume/Å^3	2288.9(3)
Z	2
ρ_{calc}g/cm^3	1.341
μ/mm^{-1}	0.104
F(000)	960.0
Crystal size/mm^3	0.2 × 0.15 × 0.15
Radiation	MoKα (λ = 0.71073)
2Θ range for data collection/°	6.056 to 49.996
Index ranges	-16 ≤ h ≤ 16, -16 ≤ k ≤ 16, -17 ≤ l ≤ 17
Reflections collected	91057
Independent reflections	8049 [R_{int} = 0.0376, R_{sigma} = 0.0172]
Data/restraints/parameters	8049/258/677
Goodness-of-fit on F^2	1.031
Final R indexes [I>=2σ (I)]	R_1 = 0.0693, wR_2 = 0.1979
Final R indexes [all data]	R_1 = 0.0857, wR_2 = 0.2226

Largest diff. peak/hole / e Å^{-3}	0.98/-0.48

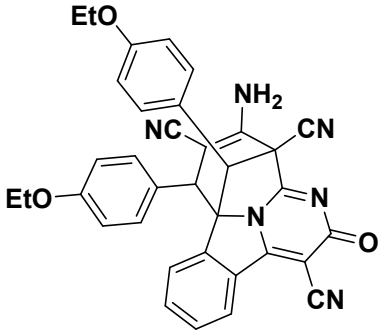
V. Analytical Data of Products



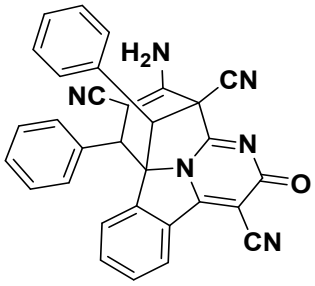
2-cyano-2-(isoindolin-1-ylidene)acetamide (3): Simple filtration from ethanol/methanol afforded the product **3** as Pale green solid, 150 mg, 75% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 10.61 (s, 1H), 8.50 – 8.31 (m, 1H), 7.72 (dt, J = 7.6, 1.0 Hz, 1H), 7.66 (td, J = 7.4, 1.1 Hz, 1H), 7.58 (td, J = 7.6, 1.3 Hz, 1H), 6.87 (s, 2H), 4.74 (s, 2H). **^{13}C NMR** (101 MHz, DMSO-d_6) δ 169.46, 163.19, 143.48, 132.96, 131.47, 127.92, 123.97, 123.51, 120.58, 64.16, 51.93. **HRMS-ESI** (m/z) calcd for $C_{11}H_{10}N_3O$ $[M + H]^+$: 200.0824; Found 200.0817.



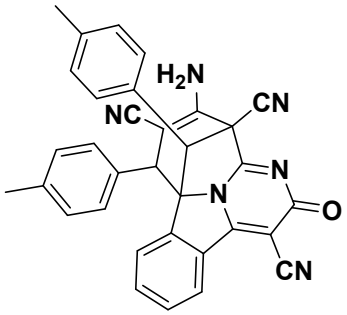
5-amino-7,12-bis(4-methoxyphenyl)-2-oxo-2H-3,3a1-diaza-4,7a-methanocyclohepta[jk]fluorene-1,4,6(7H)-tricarbonitrile (5a): Simple filtration from methanol/diethylether afforded the product **5a** as white solid, 346.2 mg, 63% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 7.85 (d, J = 7.8 Hz, 1H), 7.67 – 7.56 (m, 2H), 7.42 (td, J = 7.7, 1.1 Hz, 1H), 7.19 (s, 2H), 6.84 – 6.31 (m, 8H), 5.32 (s, 1H), 4.99 (s, 1H), 3.60 (s, 3H), 3.59 (s, 3H). **^{13}C NMR** (101 MHz, DMSO-d_6) δ) 167.71, 162.04, 159.68, 159.08, 157.12, 151.06, 143.34, 134.91, 131.19, 131.02, 127.04, 126.45, 124.90, 124.80, 118.64, 114.98, 113.93, 113.88, 112.04, 86.19, 80.62, 75.90, 63.11, 57.54, 55.41, 55.38, 49.45. **HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{33}H_{23}N_6O_3$ 551.1832; Found 551.1833.



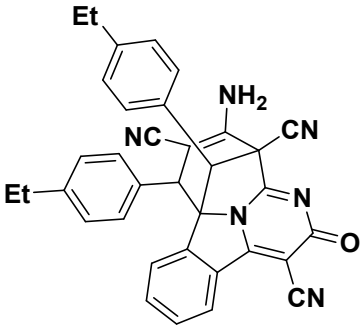
5-amino-7,12-bis(4-ethoxyphenyl)-2-oxo-2H-3,3a1-diaza-4,7a-methanocyclohepta[jk]fluorene-1,4,6(7H)-tricarbonitrile (5b): Simple filtration from methanol/diethylether afforded the product **5b** as white solid, 335mg, 58% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 7.85 (d, J = 7.8 Hz, 1H), 7.67 – 7.56 (m, 2H), 7.47 – 7.38 (m, 1H), 7.19 (s, 2H), 6.85 – 6.28 (m, 8H), 5.31 (s, 1H), 4.98 (s, 1H), 3.84 (p, J = 6.8 Hz, 4H), 1.22 (t, J = 4 Hz 3H), 1.19 (t, J = 4 Hz 3H). **^{13}C NMR** (101 MHz, DMSO-d_6) δ 167.72, 162.03, 159.00, 158.37, 157.13, 151.06, 143.37, 134.90, 131.18, 131.00, 126.89, 126.46, 124.77, 124.74, 118.64, 115.34, 114.38, 113.92, 112.04, 86.13, 80.65, 75.88, 63.38, 63.31, 63.09, 57.49, 49.44, 14.95. **HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{35}H_{27}N_6O_3$ 579.2144; Found 579.2150.



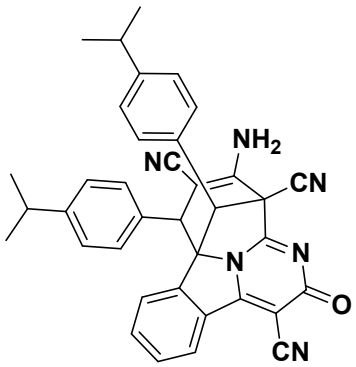
5-amino-2-oxo-7,12-diphenyl-2H-3,3a1-diaza-4,7a-methanocyclohepta[jk]fluorene-1,4,6(7H)-tricarbonitrile (5c): Simple filtration from methanol/diethylether afforded the product **5c** as white solid, 383 mg, 78% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 7.87 (d, J = 7.8 Hz, 1H), 7.58 (td, J = 7.7, 0.9 Hz, 1H), 7.50 (d, J = 7.8 Hz, 1H), 7.41 – 7.31 (m, 1H), 7.27 (s, 2H), 7.23 – 7.08 (m, 4H), 7.08 – 6.59 (m, 6H), 5.40 (s, 1H), 5.07 (s, 1H). **^{13}C NMR** (101 MHz, DMSO-d_6) δ 167.18, 161.38, 156.47, 150.74, 142.45, 134.68, 134.28, 132.48, 130.68, 130.34, 129.14, 128.86, 127.94, 126.08, 124.13, 118.11, 113.31, 111.47, 85.60, 79.80, 74.84, 62.96, 56.62, 49.61. **HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{31}H_{19}N_6O$ 491.1620; Found 491.1617.



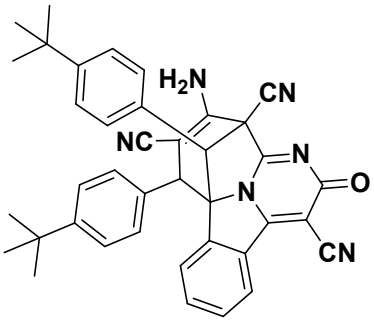
5-amino-2-oxo-7,12-di-p-tolyl-2H-3,3a1-diaza-4,7a-methanocyclohepta[jk]fluorene-1,4,6(7H)-tricarbonitrile (5d): Simple filtration from methanol/diethylether afforded the product **5d** as white solid, 393.6 mg, 76% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 7.86 (d, J = 7.8 Hz, 1H), 7.66 – 7.49 (m, 2H), 7.38 (t, J = 7.7 Hz, 1H), 7.22 (s, 2H), 7.04 – 6.46 (m, 8H), 5.33 (s, 1H), 5.01 (s, 1H), 2.09 (s, 6H). ^{13}**C NMR** (101 MHz, DMSO-d_6) δ 167.27, 161.62, 156.72, 150.66, 142.74, 138.31, 137.09, 134.44, 131.80, 130.80, 130.45, 129.78, 129.59, 128.73, 126.08, 124.28, 118.19, 113.45, 111.58, 85.58, 79.98, 75.28, 62.96, 56.82, 49.34, 20.55, 20.52. **HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{33}H_{23}N_6O$ 519.1933; Found 519.1940.



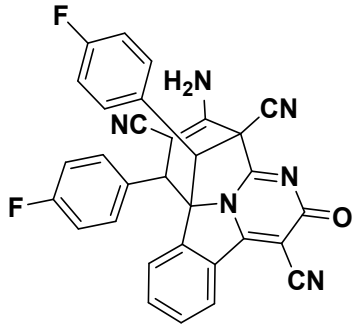
5-amino-7,12-bis(4-ethylphenyl)-2-oxo-2H-3,3a1-diaza-4,7a-methanocyclohepta[jk]fluorene-1,4,6(7H)-tricarbonitrile (5e): Simple filtration from methanol/diethylether afforded the product **5e** as white solid, 404 mg, 74% yield. mp > 290 °C. ^{1}H NMR (400 MHz, DMSO-d_6) δ 7.85 (d, J = 7.8 Hz, 1H), 7.58 (t, J = 7.7 Hz, 1H), 7.49 (d, J = 7.8 Hz, 1H), 7.34 (t, J = 7.7 Hz, 1H), 7.23 (s, 2H), 7.08 – 6.34 (m, 8H), 5.33 (s, 1H), 5.01 (s, 1H), 2.39 (d, J = 7.5 Hz, 2H), 2.36 (d, J = 5.2 Hz, 2H), 0.98 (t, J = 7.6 Hz, 6H). ^{13}**C NMR** (101 MHz, DMSO-d_6) δ 167.23, 161.40, 156.54, 150.68, 144.25, 143.29, 142.69, 134.34, 131.88, 130.63, 130.42, 129.77, 128.45, 127.36, 126.04, 124.13, 118.18, 113.38, 111.52, 85.53, 79.90, 75.06, 62.78, 56.82, 49.35, 27.46, 27.39, 15.20, 14.74. **HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{35}H_{27}N_6O$ 547.2246; Found 547.2255.



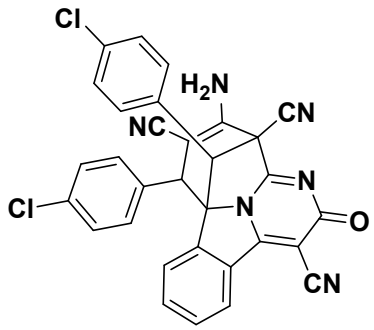
5-amino-7,12-bis(4-isopropylphenyl)-2-oxo-2H-3,3a1-diaza-4,7a-methanocyclohepta[jk]fluorene-1,4,6(7H)-tricarbonitrile (5f): Simple filtration from methanol/diethylether afforded the product **5f** as white solid, 407.4 mg, 71% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 7.83 (d, J = 7.8 Hz, 1H), 7.57 (t, J = 7.6 Hz, 1H), 7.44 (d, J = 7.7 Hz, 1H), 7.31 (t, J = 7.7 Hz, 1H), 7.24 (s, 2H), 7.08 – 6.47 (m, 8H), 5.33 (s, 1H), 4.99 (s, 1H), 2.66 (m, 2H), 0.99 (d, J = 6.8 Hz, 12H). **^{13}C NMR** (101 MHz, DMSO-d_6) δ 167.23, 161.39, 156.54, 150.64, 144.29, 143.32, 142.68, 134.35, 131.85, 130.63, 130.42, 129.74, 128.45, 127.30, 126.03, 124.13, 118.15, 113.36, 111.51, 85.53, 79.90, 75.15, 62.77, 56.83, 49.36, 27.46, 27.39, 15.19, 14.73. **HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{37}H_{31}N_6O$ 575.2559; Found 575.2563.



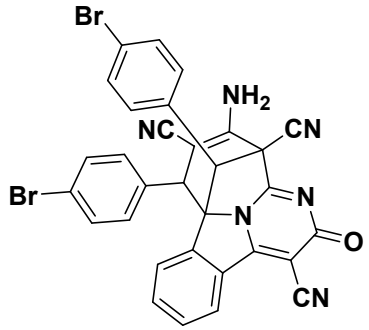
5-amino-7,12-bis(4-(tert-butyl)phenyl)-2-oxo-2H-3,3a1-diaza-4,7a-methanocyclohepta[jk]fluorene-1,4,6(7H)-tricarbonitrile (5g): Simple filtration from methanol/diethylether afforded the product **5g** as white solid, 440 mg, 73% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 7.79 (d, J = 7.7 Hz, 1H), 7.56 (t, J = 7.7 Hz, 1H), 7.42 (d, J = 7.8 Hz, 1H), 7.32 (t, J = 7.6 Hz, 1H), 7.18 (d, J = 6.3 Hz, 4H), 7.08 – 6.46 (m, 6H), 5.30 (s, 1H), 4.95 (s, 1H), 1.06 (s, 9H), 1.05 (s, 9H). **^{13}C NMR** (101 MHz, DMSO-d_6) δ 167.87, 161.53, 156.81, 151.80, 151.36, 150.93, 143.33, 134.89, 131.99, 131.20, 131.03, 130.13, 126.61, 126.46, 125.05, 124.57, 118.76, 113.93, 112.03, 86.26, 80.33, 75.49, 62.95, 57.68, 50.07, 34.76, 34.58, 31.40, 31.30. **HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{39}H_{35}N_6O$ 603.2872; Found 603.2883.



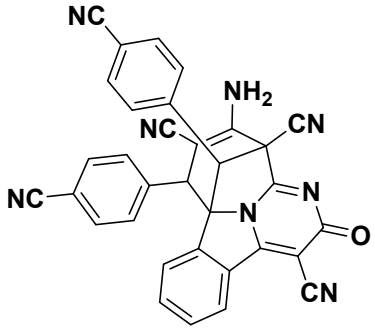
5-amino-7,12-bis(4-fluorophenyl)-2-oxo-2H-3,3a1-diaza-4,7a-methanocyclohepta[jk]fluorene-1,4,6(7H)-tricarbonitrile (5h): Simple filtration from methanol/diethylether afforded the product **5h** as white solid, 368 mg, 70% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 7.87 (d, J = 7.8 Hz, 1H), 7.70 – 7.54 (m, 2H), 7.43 (t, J = 7.7 Hz, 1H), 7.30 (s, 2H), 7.16 – 6.36 (m, 8H), 5.45 (s, 1H), 5.10 (s, 1H). ^{13}C NMR (101 MHz, DMSO-d_6) δ 167.57, 163.34 (d, $J_{C\text{-}F}$ = 40.4 Hz), 161.83, 160.90 (d, $J_{C\text{-}F}$ = 39.1 Hz), 157.54, 156.80, 151.29, 142.64, 134.92, 131.48 (d, $J_{C\text{-}F}$ = 3.2 Hz), 131.37, 130.88, 129.37 (d, $J_{C\text{-}F}$ = 3.1 Hz), 126.49, 124.83 (d, $J_{C\text{-}F}$ = 3.4 Hz), 118.54, 116.69 (d, $J_{C\text{-}F}$ = 21.8 Hz), 113.84, 111.91, 86.59, 80.23, 75.11, 62.72, 57.25, 49.32. **^{19}F NMR** (376 MHz, DMSO-d6) δ -112.15 (ddd, $J_{F\text{-}H}$ = 13.8, 8.9, 5.9 Hz, 1F), -113.78 (tt, $J_{F\text{-}H}$ = 11.2, 6.4 Hz, 1F). **HRMS (ESI)** m/z: [M + H]$^+$ calcd for $C_{31}H_{17}F_2N_6O$ 527.1432; Found 527.1429.



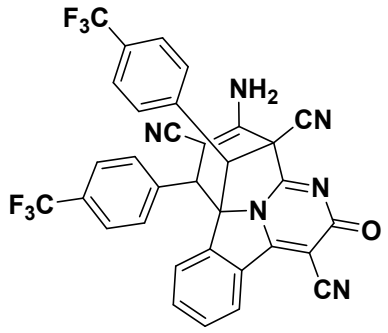
5-amino-7,12-bis(4-chlorophenyl)-2-oxo-2H-3,3a1-diaza-4,7a-methanocyclohepta[jk]fluorene-1,4,6(7H)-tricarbonitrile (5i): Simple filtration from methanol/diethylether afforded the product **5i** as white solid, 362 mg, 65% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 7.87 (d, J = 7.8 Hz, 1H), 7.67 – 7.54 (m, 2H), 7.42 (td, J = 7.7, 1.0 Hz, 1H), 7.37 – 7.23 (m, 4H), 7.08 (br s, 6H), 5.45 (s, 1H), 5.11 (s, 1H).**^{13}C NMR** (101 MHz, DMSO-d_6) δ 167.04, 161.31, 156.23, 150.88, 141.89, 134.49, 133.85, 133.58, 132.52, 132.16, 131.46, 131.01, 130.32, 129.27, 126.01, 124.37, 118.03, 113.36, 111.37, 86.23, 79.47, 74.25, 62.35, 56.60, 48.92. **HRMS (ESI)** m/z: [M + H]$^+$ calcd for $C_{31}H_{17}Cl_2N_6O$ 559.0841; Found 559.0842.



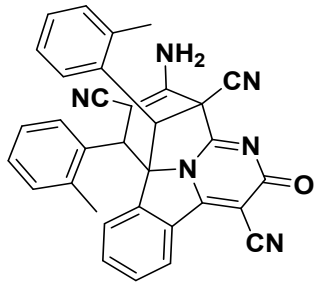
5-amino-7,12-bis(4-bromophenyl)-2-oxo-2H-3,3a1-diaza-4,7a-methanocyclohepta[jk]fluorene-1,4,6(7H)-tricarbonitrile (5j): Simple filtration from methanol/diethylether afforded the product **5j** as white solid, 439 mg, 68% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 7.88 (d, J = 7.8 Hz, 1H), 7.67 – 7.54 (m, 2H), 7.48 – 7.05 (m, 8H), 6.75 (br s, 3H), 5.43 (s, 1H), 5.09 (s, 1H). **^{13}C NMR** (101 MHz, DMSO-d_6) δ 167.19, 167.19, 161.43, 161.43, 156.36, 156.35, 150.99, 141.98, 134.66, 134.37, 132.34, 131.95, 131.15, 130.44, 126.13, 124.53, 122.46, 121.34, 118.17, 113.50, 111.50, 86.38, 79.45, 74.34, 62.55, 56.66, 49.12. **HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{31}H_{17}N_6O$ 489.1463; Found 489.1469.



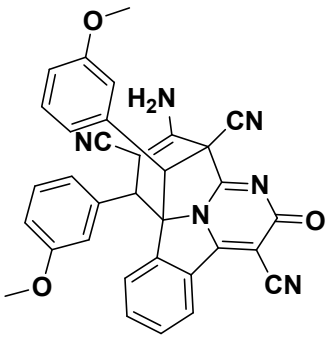
5-amino-7,12-bis(4-cyanophenyl)-2-oxo-2H-3,3a1-diaza-4,7a-methanocyclohepta[jk]fluorene-1,4,6(7H)-tricarbonitrile (5k): Simple filtration from methanol/diethylether afforded the product **5k** as white solid, 389 mg, 72% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 7.91 (d, J = 7.8 Hz, 1H), 7.71 (t, J = 7.4 Hz, 3H), 7.65 – 7.54 (m, 3H), 7.51 – 7.39 (m, 4H), 7.11 (s, 3H), 5.59 (s, 1H), 5.25 (s, 1H). **^{13}C NMR** (101 MHz, DMSO-d_6) δ 165.00, 159.19, 154.04, 149.20, 139.24, 138.52, 135.71, 132.61, 131.17, 129.26, 128.25, 124.12, 122.52, 116.26, 116.01, 115.89, 111.37, 109.78, 109.30, 108.84, 84.62, 77.20, 71.50, 60.70, 54.44, 47.55. **HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{33}H_{17}N_8O$ 541.1525; Found 541.1512.



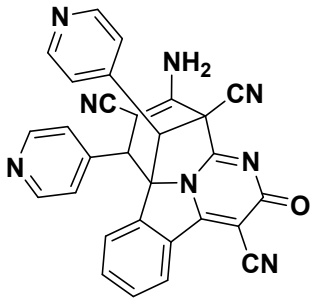
5-amino-2-oxo-7,12-bis(4-(trifluoromethyl)phenyl)-2H-3,3a1-diaza-4,7a-methanocyclohepta[jk]fluorene-1,4,6(7H)-tricarbonitrile (5l): Simple filtration from methanol/diethylether afforded the product **5l** as white solid, 407 mg, 65% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 7.94 (d, J = 7.8 Hz, 1H), 7.68 – 7.57 (m, 3H), 7.55 (d, J = 7.8 Hz, 1H), 7.48 – 7.35 (m, 4H), 7.13 (s, 4H), 5.61 (s, 1H), 5.28 (s, 1H). ^{13}C NMR (101 MHz, DMSO-d_6) δ 166.99, 161.14, 155.97, 151.10, 141.47, 139.56, 136.98, 134.53, 131.06, 130.28, 128.74 (q, $J_{C\text{-}F}$ = 31.9 Hz), 126.12 (q, $J_{C\text{-}F}$ = 6.4 Hz), 125.05 (d, $J_{C\text{-}F}$ = 22.5 Hz), 124.28, 122.35 (d, $J_{C\text{-}F}$ = 22.7 Hz), 118.01, 113.34, 111.30, 86.52, 79.17, 73.76, 62.49, 56.62, 49.35.**^{19}F NMR** (376 MHz, DMSO-d6) δ -61.13, -61.47. **HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{33}H_{17}F_6N_6O$ 627.1367; Found 627.1370.



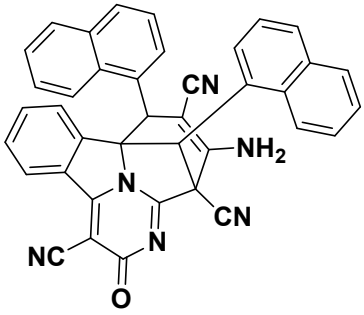
5-amino-2-oxo-7,12-di-o-tolyl-2H-3,3a1-diaza-4,7a-methanocyclohepta[jk]fluorene-1,4,6(7H)-tricarbonitrile (5m): Simple filtration from methanol/diethylether afforded the product **5m** as white solid, 336 mg, 65% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) 7.74 (d, J = 7.7 Hz, 1H), 7.55 (t, J = 7.4 Hz, 1H), 7.50 (dd, J = 7.7, 1.1 Hz, 1H), 7.37 (td, J = 7.6, 0.9 Hz, 1H), 7.18 – 7.07 (m, 3H), 7.02 – 6.97 (m, 1H), 6.97 – 6.88 (m, 2H), 6.85 (td, J = 7.6, 1.5 Hz, 1H), 6.78 (dd, J = 7.6, 1.7 Hz, 1H), 6.60 (dd, J = 7.6, 1.7 Hz, 1H), 5.75 (dd, J = 8.0, 1.2 Hz, 1H), 5.42 (d, J = 19.3 Hz, 1H), 2.61 (s, 3H), 2.09 (s, 3H). **^{13}C NMR** (101 MHz, DMSO-d_6) δ 167.80, 161.33, 156.70, 150.51, 142.55, 138.42, 136.76, 134.77, 133.95, 131.96, 131.62, 131.30, 131.10, 131.03, 129.40, 128.48, 127.86, 127.46, 126.50, 125.50, 124.85, 124.13, 118.63, 113.91, 111.91, 86.59, 80.35, 77.31, 59.56, 57.74, 45.40, 21.01, 20.37. **HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{33}H_{23}N_6O$ 519.1933; Found 519.1942.



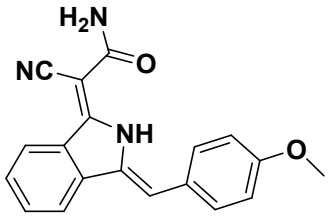
5-amino-7,12-bis(3-methoxyphenyl)-2-oxo-2H-3,3a1-diaza-4,7a-methanocyclohepta[jk]fluorene-1,4,6(7H)-tricarbonitrile (5n): Simple filtration from methanol/diethylether afforded the product **5n** as white solid, 357.2 mg, 65% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 7.92 (d, J = 7.8 Hz, 1H), 7.65 (t, J = 7.7 Hz, 1H), 7.58 (d, J = 7.8 Hz, 1H), 7.43 (t, J = 7.7 Hz, 1H), 7.27 (s, 2H), 7.17 – 6.91 (m, 3H), 6.73 (dd, J = 8.3, 2.1 Hz, 1H), 6.64 (d, J = 6.4 Hz, 1H), 6.38 – 6.16 (m, 3H), 5.37 (s, 1H), 5.04 (s, 1H), 3.61 (s, 3H), 3.50 (s, 3H). **^{13}C NMR** (101 MHz, DMSO-d_6) δ 167.56, 161.99, 159.66, 157.25, 151.13, 143.05, 136.70, 134.89, 134.33, 131.32, 130.99, 130.94, 129.74, 126.72, 124.75, 123.44, 118.46, 114.78, 114.08, 113.60, 111.86, 85.78, 80.24, 75.62, 63.39, 57.23, 55.43, 55.38, 50.21. **HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{33}H_{23}N_6O_3$ 551.1832; Found 551.1823.



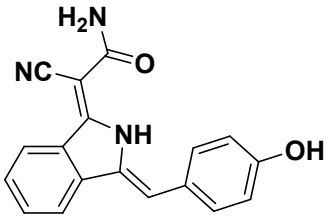
5-amino-2-oxo-7,12-di(pyridin-4-yl)-2H-3,3a1-diaza-4,7a-methanocyclohepta[jk]fluorene-1,4,6(7H)-tricarbonitrile (5o): Simple filtration from methanol/diethylether afforded the product **5o** as white solid, 305 mg, 62% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 8.40 (d, J = 5.3 Hz, 2H), 8.22 (br s, 2H), 7.94 (d, J = 7.7 Hz, 1H), 7.63 (t, J = 7.7 Hz, 0H), 7.58 (d, J = 7.7 Hz, 1H), 7.48 – 7.40 (m, 3H), 6.86 (s, 4H), 5.50 (s, 1H), 5.14 (s, 1H). **^{13}C NMR** (101 MHz, DMSO-d_6) δ 167.09, 161.18, 156.10, 151.30, 150.70, 149.60, 143.80, 141.22, 140.91, 134.71, 131.38, 130.41, 126.23, 124.58, 118.03, 118.00, 113.43, 111.33, 86.74, 78.77, 73.29, 62.12, 56.28, 49.11. **HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{29}H_{17}N_8O$ 493.1525; Found 493.1518.



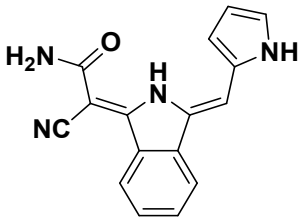
5-amino-7,12-di(naphthalen-1-yl)-2-oxo-2H-3,3a1-diaza-4,7a-methanocyclohepta[jk]fluorene-1,4,6(7H)-tricarbonitrile (5p): Simple filtration from methanol/diethylether afforded the product **5p** as white solid, 437 mg, 74% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 9.12 (d, J = 8.8 Hz, 1H), 8.59 (d, J = 8.7 Hz, 1H), 7.91 (t, J = 7.1 Hz, 2H), 7.77 (d, J = 8.2 Hz, 1H), 7.70 (q, J = 7.3 Hz, 3H), 7.48 (dd, J = 8.1, 4.6 Hz, 2H), 7.44 – 7.24 (m, 6H), 7.08 (d, 1H), 6.96 (t, J = 7.4 Hz, 1H), 6.88 (t, 1H), 6.59 (s, 1H), 6.49 (d, J = 7.4 Hz, 1H), 6.42 (s, 1H). **^{13}C NMR** (101 MHz, DMSO-d_6) δ 13C NMR (101 MHz, DMSO) δ 167.85, 161.58, 156.78, 150.83, 141.55, 133.74, 133.50, 133.44, 131.90, 131.85, 131.72, 130.92, 130.66, 130.03, 129.26, 129.21, 129.07, 128.69, 127.57, 127.05, 126.49, 126.18, 126.05, 125.70, 125.49, 124.94, 124.37, 124.02, 123.21, 118.83, 113.82, 112.23, 86.04, 81.04, 77.54, 58.71, 57.40, 43.55. **HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{39}H_{23}N_6O$ 591.1933; Found 591.1942.



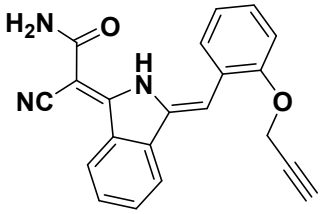
2-cyano-2-((Z)-3-((Z)-4-methoxybenzylidene)isoindolin-1-ylidene)acetamide (6a): The filtered crude solid (mixture of **5a** and **6a**) was washed with methanol/diethyl ether; compound **6a** was recovered from the filtrate after solvent removal under reduced pressure as a yellowish-green solid, 54 mg, 17% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 12.80 (s, 1H), 8.33 (d, J = 7.9 Hz, 1H), 8.10 (d, J = 7.8 Hz, 1H), 7.72 (t, J = 7.5 Hz, 1H), 7.62 (t, J = 7.6 Hz, 1H), 7.52 (d, J = 8.4 Hz, 2H), 7.39 (s, 2H), 7.07 (d, J = 8.3 Hz, 2H), 7.00 (s, 1H), 3.80 (s, 3H). **HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{19}H_{16}N_3O_2$ 318.1242; Found 318.1269. Compound 6a was unstable in standard NMR solvent, tending to form aggregates. As a result, reliable ^{13}C NMR spectra could not be obtained.



2-cyano-2-((Z)-3-((Z)-4-hydroxybenzylidene)isoindolin-1-ylidene)acetamide (6b): Simple filtration from methanol afforded the product **6b** as yellowish green solid, 221 mg, 73% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 12.82 (s, 1H), 9.99 (s, 1H), 8.35 (d, J = 8.0 Hz, 1H), 8.12 (d, J = 7.8 Hz, 1H), 7.73 (t, J = 7.5 Hz, 1H), 7.62 (t, J = 7.7 Hz, 1H), 7.56 – 7.29 (m, 4H), 7.00 (s, 1H), 6.91 (d, J = 8.2 Hz, 2H).^{13}**C NMR** (101 MHz, DMSO-d_6) δ 168.99, 157.89, 157.07, 136.57, 132.28, 131.91, 130.14, 129.72, 128.89, 125.66, 123.56, 120.88, 119.14, 116.43, 109.30, 68.01. **HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{18}H_{14}N_3O_2$ 304.1086; Found 304.1081.

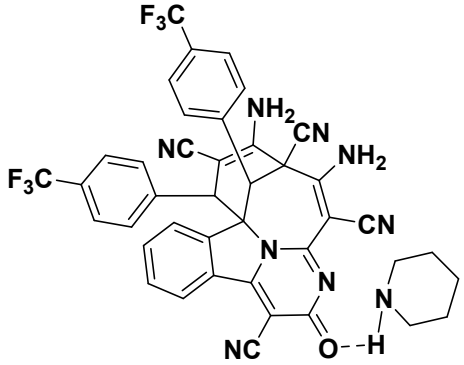


2-((1Z,3Z)-3-((1H-pyrrol-2-yl)methylene)isoindolin-1-ylidene)-2-cyanoacetamide (6c): Simple filtration from methanol afforded the product **6d** as yellowish green solid, 201 mg, 73% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 12.67 (s, 1H), 11.42 (s, 1H), 8.35 (d, J = 8.0 Hz, 1H), 7.97 (d, J = 7.8 Hz, 1H), 7.70 (t, J = 7.6 Hz, 1H), 7.58 (t, J = 7.6 Hz, 1H), 7.34 (s, 2H), 7.05 (d, J = 2.7 Hz, 1H), 6.86 (s, 1H), 6.60 (t, J = 3.1 Hz, 1H), 6.35 (q, J = 2.7 Hz, 1H). ^{13}**C NMR** (101 MHz, DMSO-d_6) δ 169.13, 155.92, 136.05, 131.80, 129.77, 129.61, 128.30, 127.21, 123.71, 121.50, 120.27, 119.37, 111.33, 109.10, 100.55, 67.51. **HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{16}H_{13}N_4O$ 277.1089; Found 277.1086.

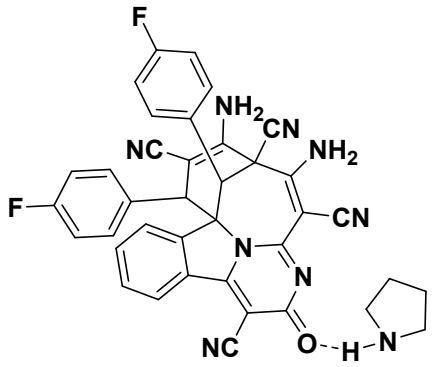


2-cyano-2-((Z)-3-((Z)-2-(prop-2-yn-1-yloxy)benzylidene)isoindolin-1-ylidene)acetamide (6d): Simple filtration from methanol afforded the product **6c** as yellowish green solid, 208 mg, 61% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 12.50 (s, 1H), 8.38 (d, J = 7.9 Hz, 1H), 8.17 (d, J = 7.8 Hz, 1H), 7.75 (t, J = 7.5 Hz, 1H), 7.66 (t, J = 7.7 Hz, 1H), 7.57 (dd, J = 7.8, 1.6 Hz, 1H), 7.46 – 7.34 (m, 4H), 7.12 (t, J = 7.4 Hz, 1H), 6.99 (s, 1H), 5.04 (d, J = 2.5

Hz, 2H), 3.59 (t, J = 2.3 Hz, 1H). **^{13}C NMR** (101 MHz, DMSO-d_6) δ 168.45, 156.49, 154.37, 136.32, 134.67, 132.03, 130.52, 130.47, 129.80, 129.45, 123.76, 123.69, 121.98, 121.24, 119.15, 113.50, 104.31, 79.04, 78.79, 69.00, 55.96. **HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{21}H_{16}N_3O$ 326.1293; Found 326.1296.

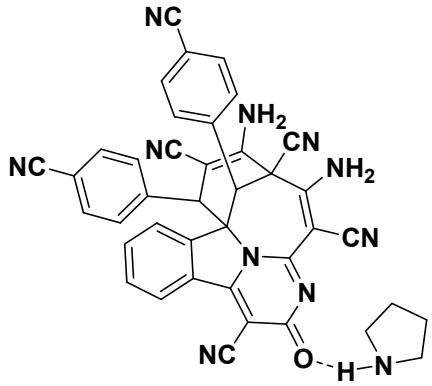


5,7-diamino-2-oxo-9,14-bis(4-(trifluoromethyl)phenyl)-2H-3,3a1-diaza-6,9a-methanocyclonona[jk]fluorene-1,4,6,8(9H)-tetracarbonitrile with Piperidine (7a): Simple filtration from methanol/diethylether afforded the product **7a** as pale yellow solid, 540 mg, 78% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 8.68 – 8.02 (m, 4H), 7.75 – 7.65 (m, 2H), 7.56 (d, J = 8.4 Hz, 2H), 7.51 (d, J = 6.9 Hz, 1H), 7.45 (t, J = 7.4 Hz, 1H), 7.34 – 7.25 (m, 3H), 7.18 (d, J = 8.0 Hz, 1H), 6.50 (s, 2H), 5.93 (d, J = 8.1 Hz, 1H), 5.65 (s, 1H), 4.64 (s, 1H), 3.03 (t, 4H), 1.65 (p, J = 6.2, 5.8 Hz, 4H), 1.61 – 1.40 (m, 2H).^{13}**C NMR** (101 MHz, DMSO-d_6) δ 163.87, 156.46, 155.27, 153.26, 152.14, 143.46, 139.32, 137.99, 133.42, 130.62 (q, $J_{C\text{-}F}$ = 3.5 Hz), 130.15, 129.00 (d, J = 9.8 Hz), 128.84, 128.65 (d, J = 2.4 Hz), 125.80 – 125.23 (m), 124.82 (d), 124.46 – 123.79 (q, $J_{C\text{-}F}$ =7.1 Hz), 123.60 (d, $J_{C\text{-}F}$ = 3.8 Hz), 122.56, 122.45, 122.26 (d, $J_{C\text{-}F}$ = 4.2 Hz), 122.06, 117.87, 117.28 (d, J = 2.6 Hz), 115.06, 84.34, 73.92 (d, J = 4.4 Hz), 57.15, 53.51, 45.84, 43.78, 22.26, 21.64. **^{19}F NMR** (376 MHz, DMSO-d6) δ -61.20, -61.29. **HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{36}H_{19}F_6N_8O$ 693.1585; Found 693.1585.

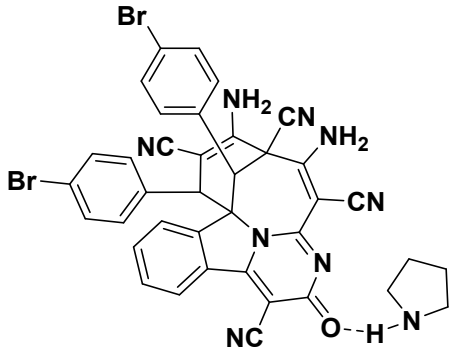


5,7-diamino-9,14-bis(4-fluorophenyl)-2-oxo-2H-3,3a1-diaza-6,9a-methanocyclonona[jk]fluorene-1,4,6,8(9H)-tetracarbonitrile with pyrrolidine (7b): Simple filtration from methanol/diethylether afforded the product **7b** as pale yellow solid, 432

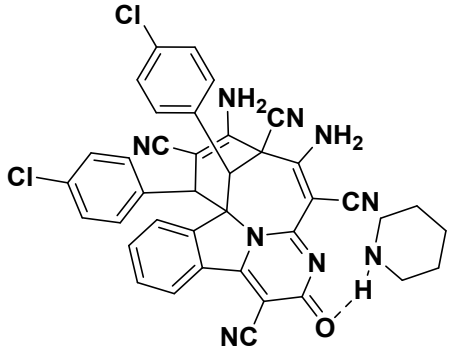
mg, 73% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 8.51 (s, 2H), 8.31 – 8.02 (m, 1H), 7.69 (dd, J = 8.3, 6.8 Hz, 2H), 7.45 (t, J = 7.6 Hz, 1H), 7.27 – 6.82 (m, 6H), 6.66 (td, J = 8.7, 2.8 Hz, 1H), 6.35 (s, 2H), 5.83 – 5.63 (m, 1H), 5.47 (s, 1H), 4.51 (s, 1H), 3.25 – 3.03 (m, 4H), 1.98 – 1.67 (m, 4H). ^{13}**C NMR** (101 MHz, DMSO-d_6) δ 164.17, 163.70, 162.98 (d, $J_{C\text{-}F}$ = 4.3 Hz), 160.54 (d, $J_{C\text{-}F}$ = 4.5 Hz), 156.85, 155.43, 152.99, 144.12, 133.32, 131.73 (q, $J_{C\text{-}F}$ = 12.0, 7.5 Hz), 130.91 (d, $J_{C\text{-}F}$ = 3.0 Hz), 130.12 (d, $J_{C\text{-}F}$ = 8.9 Hz), 129.97 – 129.64 (m), 128.80, 123.65, 122.09 (d, J = 9.5 Hz), 117.91, 117.45, 115.48 (d, $J_{C\text{-}F}$ = 5.0 Hz), 115.28 (d, $J_{C\text{-}F}$ = 4.1 Hz), 115.04, 114.03, 113.82, 83.89, 75.41, 74.63, 74.10, 57.30, 52.89, 45.41, 44.98, 23.66. **^{19}F NMR** (376 MHz, DMSO-d6) δ -113.22, -113.82. **HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{34}H_{19}F_2N_8O$ 593.1649; Found 593.1655.



5,7-diamino-9,14-bis(4-cyanophenyl)-2-oxo-2H-3,3a1-diaza-6,9a-methanocyclonona[jk]fluorene-1,4,6,8(9H)-tetracarbonitrile with pyrrolidine (7c): Simple filtration from methanol/diethylether afforded the product **7c** as pale yellow solid, 442 mg, 73% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 8.59 (s, 2H), 8.21 (d, J = 7.9 Hz, 1H), 7.74 (d, J = 8.5 Hz, 1H), 7.72 – 7.67 (m, 2H), 7.64 (d, J = 8.8 Hz, 2H), 7.47 (t, J = 7.6 Hz, 1H), 7.33 (dd, J = 8.0, 1.6 Hz, 1H), 7.30 – 7.21 (m, 3H), 6.55 (s, 2H), 5.93 (dd, J = 8.1, 1.6 Hz, 1H), 5.65 (s, 1H), 4.64 (s, 1H), 3.14 (t, J = 7.2 Hz, 4H), 2.09 – 1.73 (m, 4H).^{13}**C NMR** (101 MHz, DMSO-d_6) δ 163.96, 163.39, 156.56, 155.23, 153.29, 143.25, 140.28, 138.66, 133.59, 133.03, 132.41, 131.88, 131.05, 130.87, 130.30, 129.23, 128.70, 128.65, 123.66, 122.31, 121.91, 118.26, 118.06, 117.76, 117.12, 115.19, 111.45, 110.95, 84.23, 74.08, 73.99, 73.82, 70.74, 56.99, 53.61, 46.02, 44.98, 23.67.**HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{36}H_{19}N_{10}O$ 607.1743; Found 607.1749.

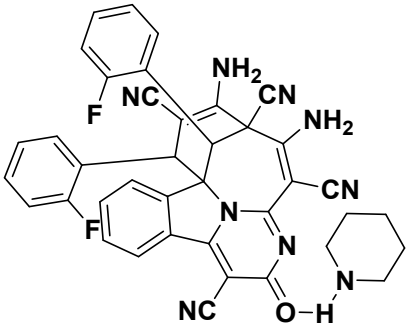


5-amino-7,12-bis(4-fluorophenyl)-2-oxo-2H-3,3a1-diaza-4,7a-methanocyclohepta[jk]fluorene-1,4,6(7H)-tricarbonitrile with pyrrolidine (7d): Simple filtration from methanol/diethylether afforded the product **7d** as pale yellow solid, 463 mg, 65% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 8.57 (s, 2H), 8.23 (d, J = 7.8 Hz, 1H), 7.97 – 7.67 (m, 3H), 7.51 (t, J = 7.6 Hz, 1H), 7.45 – 7.36 (m, 3H), 7.32 – 7.04 (m, 3H), 7.02 (dd, J = 8.4, 2.4 Hz, 1H), 6.47 (s, 2H), 5.73 (dd, J = 8.3, 2.4 Hz, 1H), 5.53 (s, 1H), 4.56 (s, 1H), 3.22 – 3.16 (m, 4H), 1.96 – 1.85 (m, 4H). ^{13}**C NMR** (101 MHz, DMSO-d_6) δ 164.57, 164.10, 157.21, 155.84, 153.52, 144.30, 134.53, 133.82, 133.26, 132.80, 132.27, 131.96, 131.49, 130.62, 130.59, 130.53, 130.47, 129.24, 124.03, 122.71, 122.47, 121.97, 118.31, 117.81, 115.81, 84.58, 75.42, 74.64, 74.51, 57.59, 53.63, 49.08, 45.44, 24.12. **HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{34}H_{19}Br_2N_8O$ 713.0049; Found 713.0044.



5,7-diamino-9,14-bis(4-chlorophenyl)-2-oxo-2H-3,3a1-diaza-6,9a-methanocyclonona[jk]fluorene-1,4,6,8(9H)-tetracarbonitrile with piperidine (7e): Simple filtration from methanol/diethylether afforded the product **6e** as pale yellow solid, 381 mg, 61% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 8.45 (s, 1H), 8.12 (d, J = 7.8 Hz, 1H), 7.96 – 7.56 (m, 2H), 7.44 (t, J = 7.7 Hz, 1H), 7.19 (d, J = 8.4 Hz, 3H), 7.07 (d, J = 8.4 Hz, 2H), 7.01 (dd, J = 8.4, 2.3 Hz, 1H), 6.86 (dd, J = 8.3, 2.3 Hz, 1H), 6.29 (s, 2H), 5.75 (dd, J = 8.4, 2.3 Hz, 1H), 5.40 (s, 1H), 4.51 (s, 1H), 3.01 (t, 4H), 1.64 (p, J = 5.7 Hz, 4H), 1.55 (p, J = 5.5, 2.6 Hz, 2H). ^{13}**C NMR** (101 MHz, DMSO-d_6) δ 164.70, 164.17, 157.30, 155.90, 153.48, 144.18, 134.04, 133.90, 133.77, 133.44, 132.75, 132.02, 131.92, 130.46, 130.27, 129.14,

129.01, 128.56, 127.59, 124.00, 122.67, 122.59, 118.31, 117.79, 115.80, 84.45, 75.47, 74.79, 74.43, 57.65, 53.44, 46.07, 44.30, 22.67, 22.03. **HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{34}H_{19}Cl_{12}N_8O$ 974.7944; Found 625.1065.



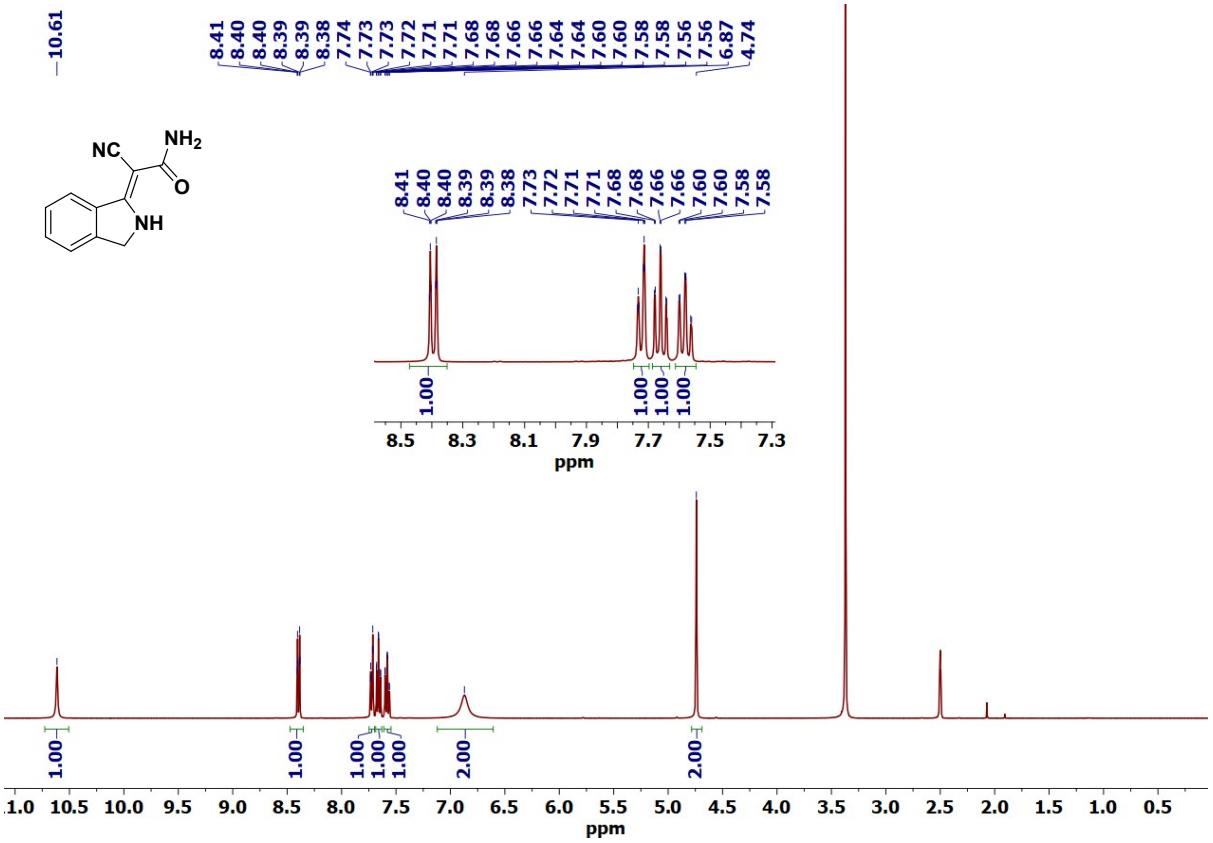
5,7-diamino-9,14-bis(2-fluorophenyl)-2-oxo-2H-3,3a1-diaza-6,9a-methanocyclonona[jk]fluorene-1,4,6,8(9H)-tetracarbonitrile with piperidine (7f): Simple filtration from methanol/diethylether afforded the product **7f** as pale yellow solid, 414 mg, 70% yield. mp > 290 °C. ^{1}**H NMR** (400 MHz, DMSO-d_6) δ 8.17 (d, J = 8.0 Hz, 3H), 7.70 – 7.61 (m, 2H), 7.42 (t, J = 7.5 Hz, 1H), 7.25 – 7.20 (m, 1H), 7.17 (dd, J = 7.1, 2.5 Hz, 1H), 7.13 – 7.05 (m, 2H), 7.00 (t, J = 7.5 Hz, 1H), 6.93 (t, J = 7.8 Hz, 1H), 6.85 (t, J = 7.4 Hz, 1H), 6.72 (dd, J = 10.4, 8.3 Hz, 1H), 6.32 (s, 2H), 5.78 (d, J = 2.1 Hz, 1H), 5.20 (s, 1H), 3.12 (t, J = 7.1 Hz, 4H), 2.06 – 1.73 (m, 4H). **^{19}F NMR** (376 MHz, DMSO-d6) δ -112.98, -113.66. **HRMS (ESI)** m/z: $[M + H]^+$ calcd for $C_{34}H_{19}F_2N_8O$ 593.1650; Found 593.1654.

VI. References

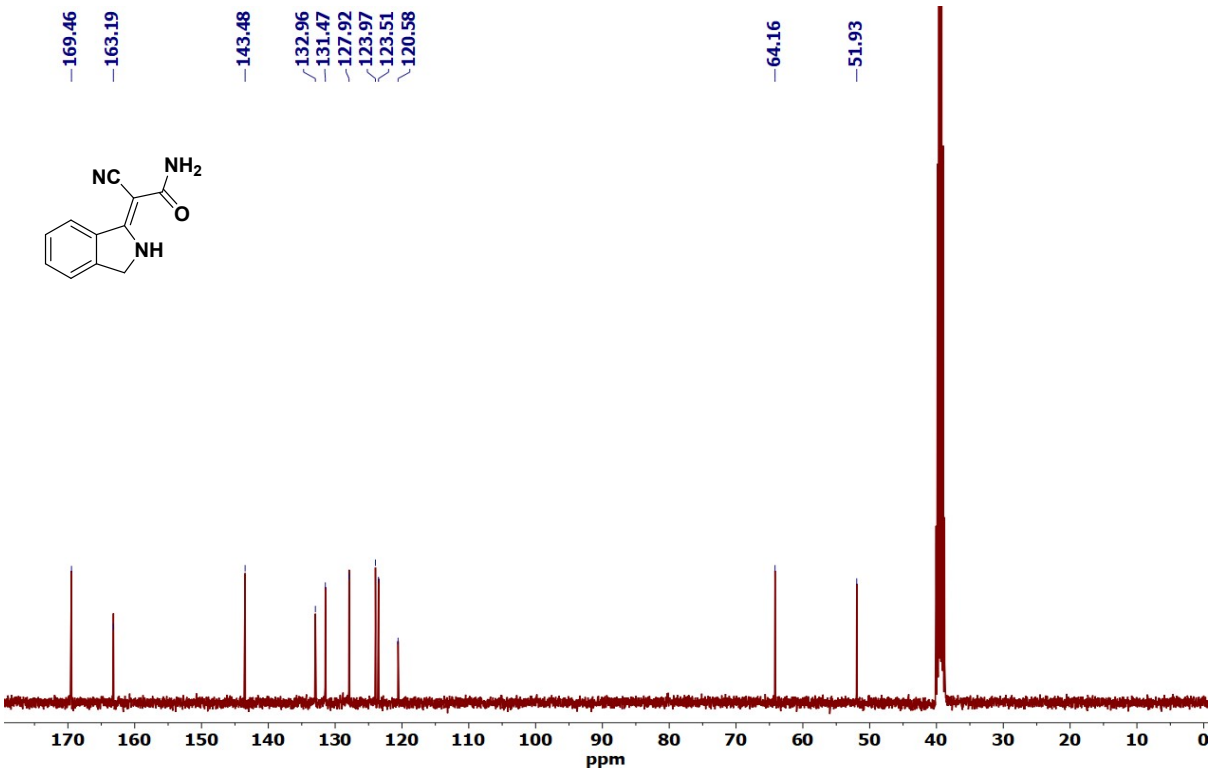
1. Natarajan, N.; Jayabal, E.; Venkatesan, R.; Vasuki, G. Multicomponent Synthesis of Pyrrolo[2,1-a]Isoindolylidene-Malononitrile (PIYM) Fluorophores and Their Photophysical Properties. *Org. Biomol. Chem.* **2022**, *20* (46), 9192–9206.

VII. ^{1}H and ^{13}C NMR and ^{19}F NMR Spectra

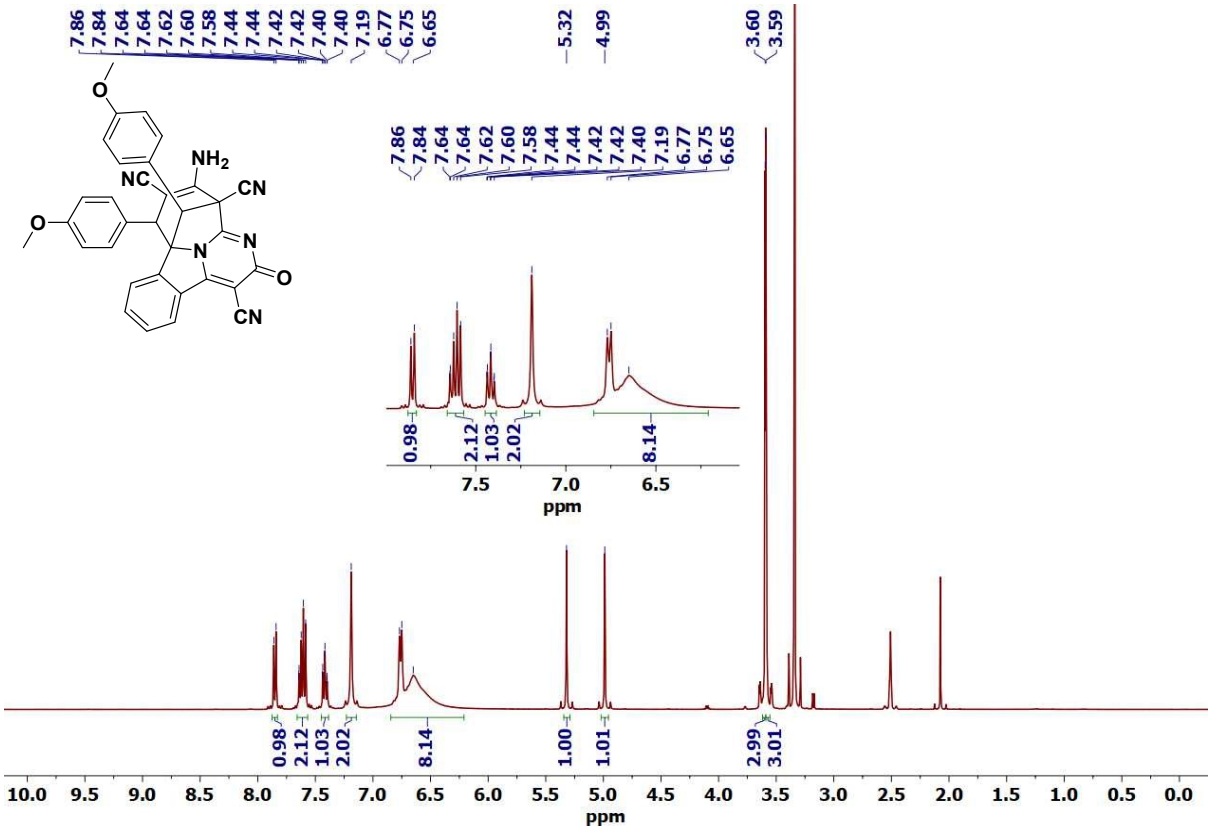
^{1}H NMR spectra of **3** (400 MHz, DMSO-d_6)



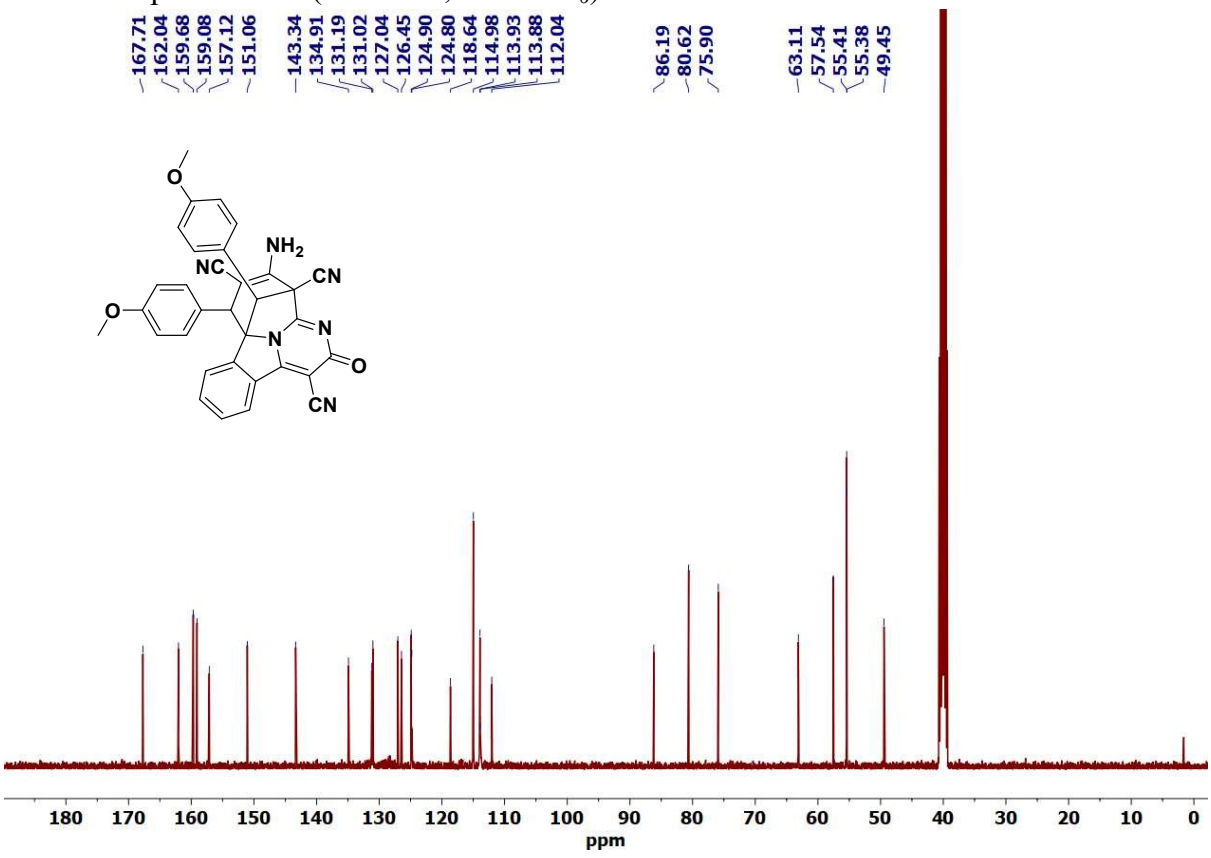
^{13}C NMR spectra of **3** (101 MHz, DMSO-d_6)



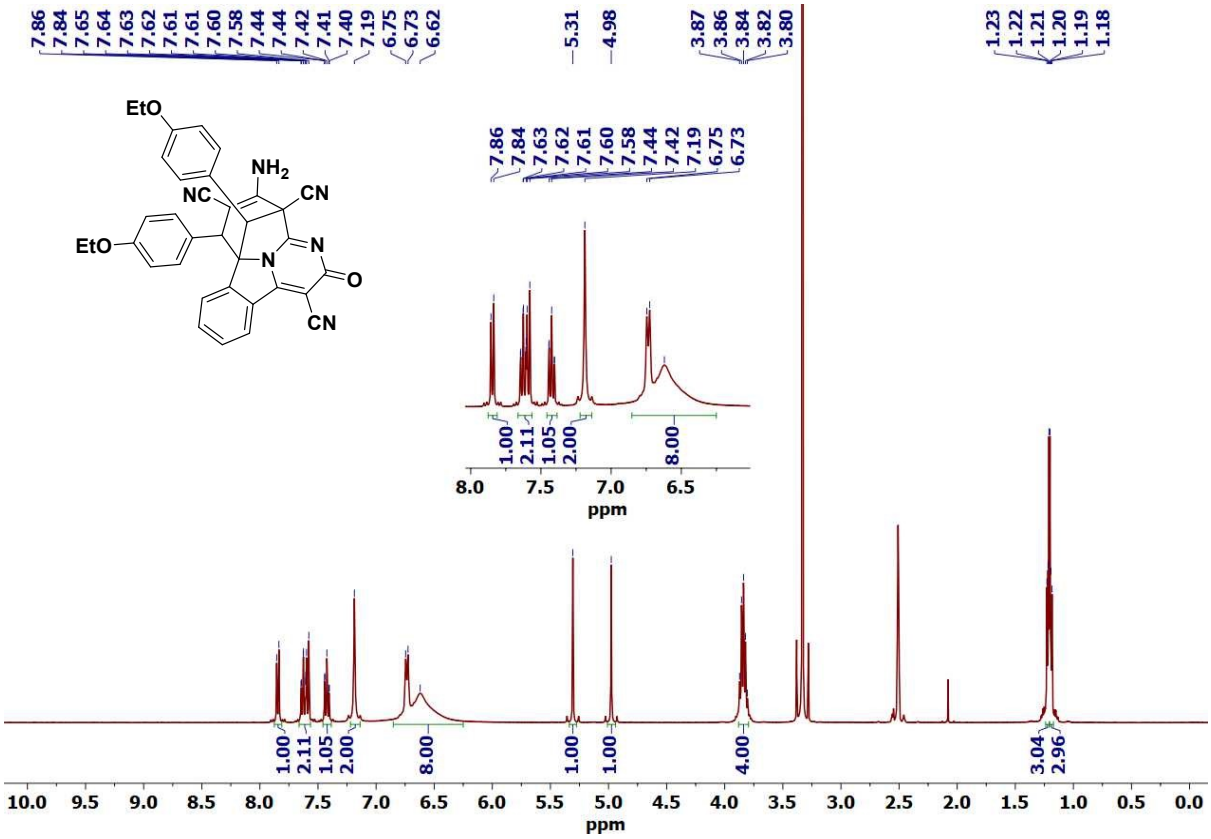
^{1}H NMR spectra of **5a** (400 MHz, DMSO-d_6)



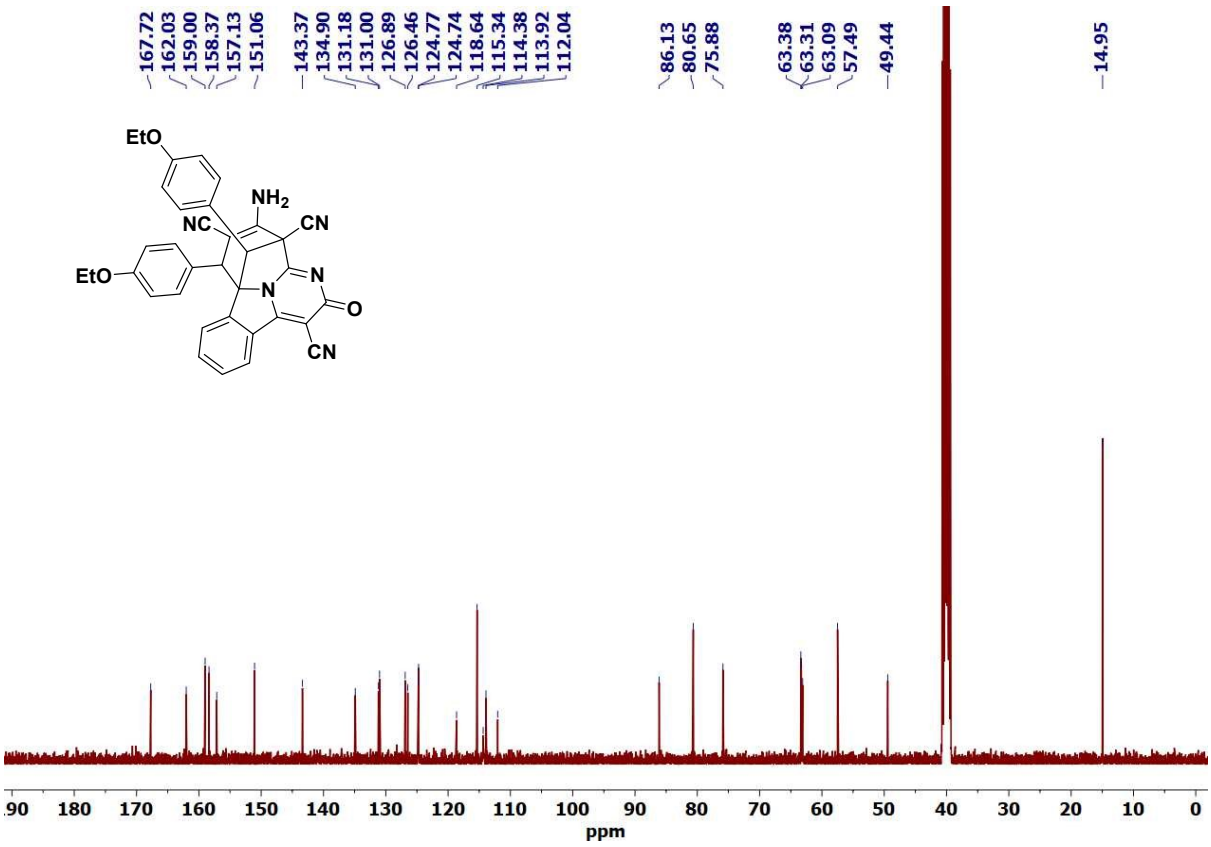
^{13}C NMR spectra of **5a** (101 MHz, DMSO-d_6)



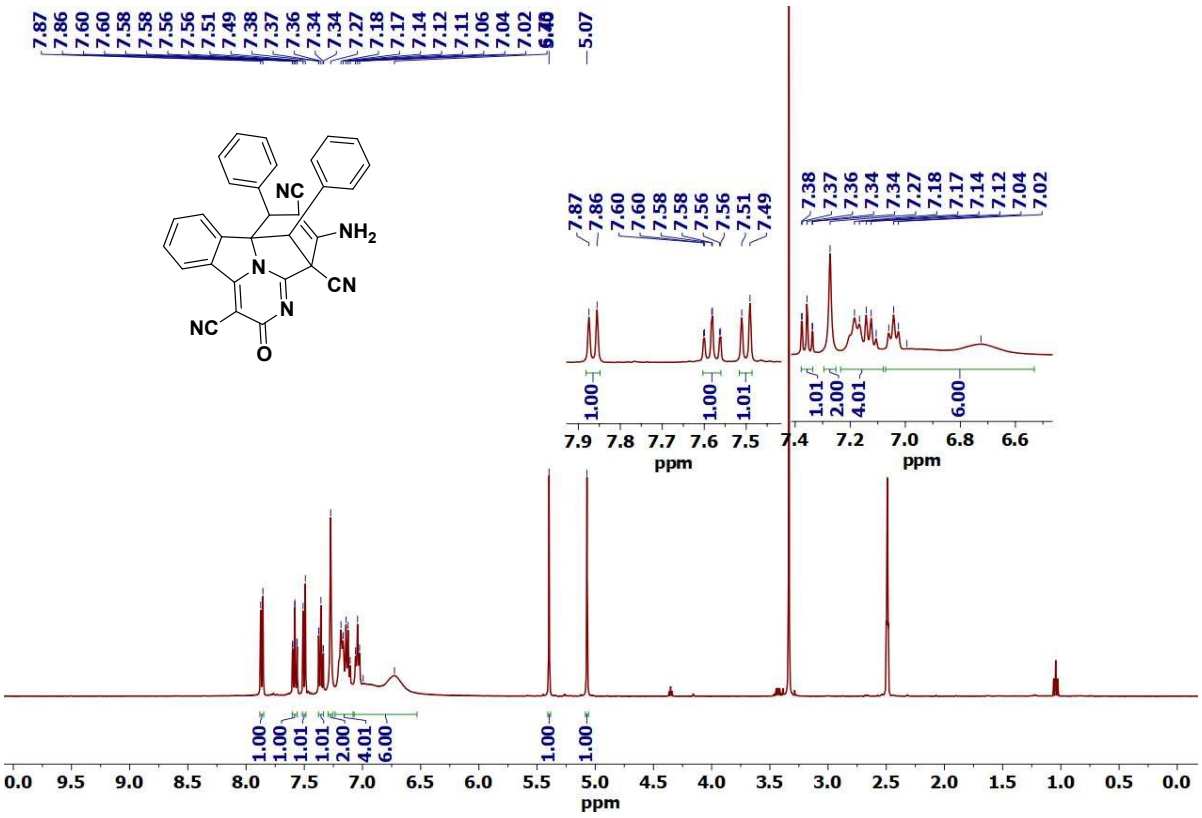
^{1}H NMR spectra of **5b** (400 MHz, DMSO-d_6)



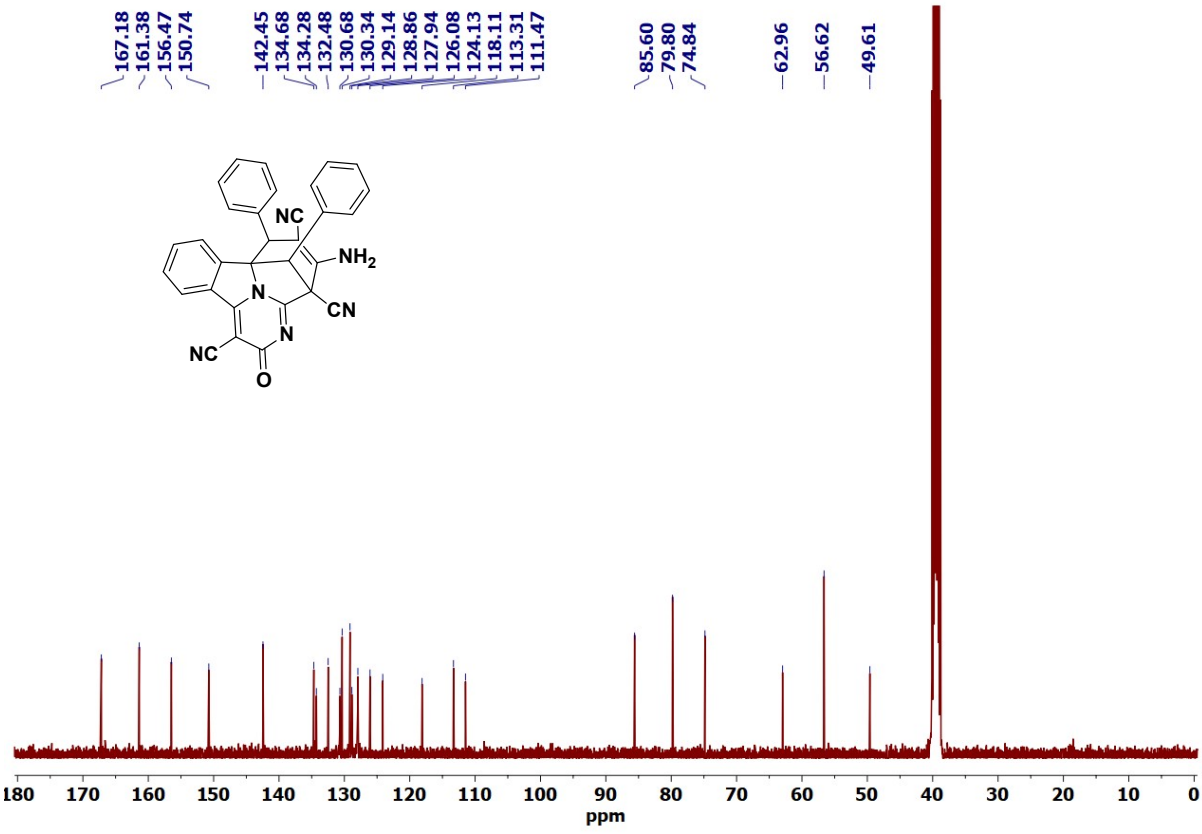
^{13}C NMR spectra of **5b** (101 MHz, DMSO-d_6)



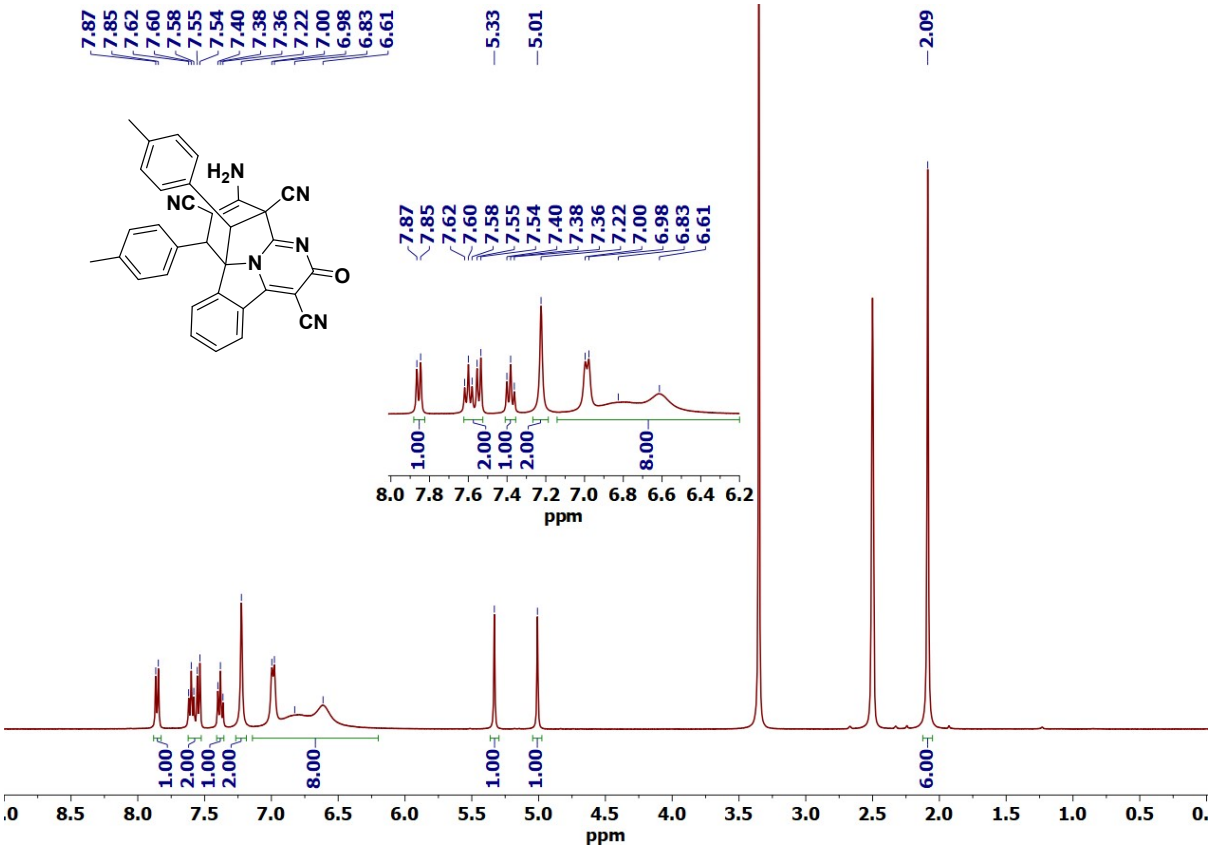
^{1}H NMR spectra of **5c** (400 MHz, DMSO-d_6)



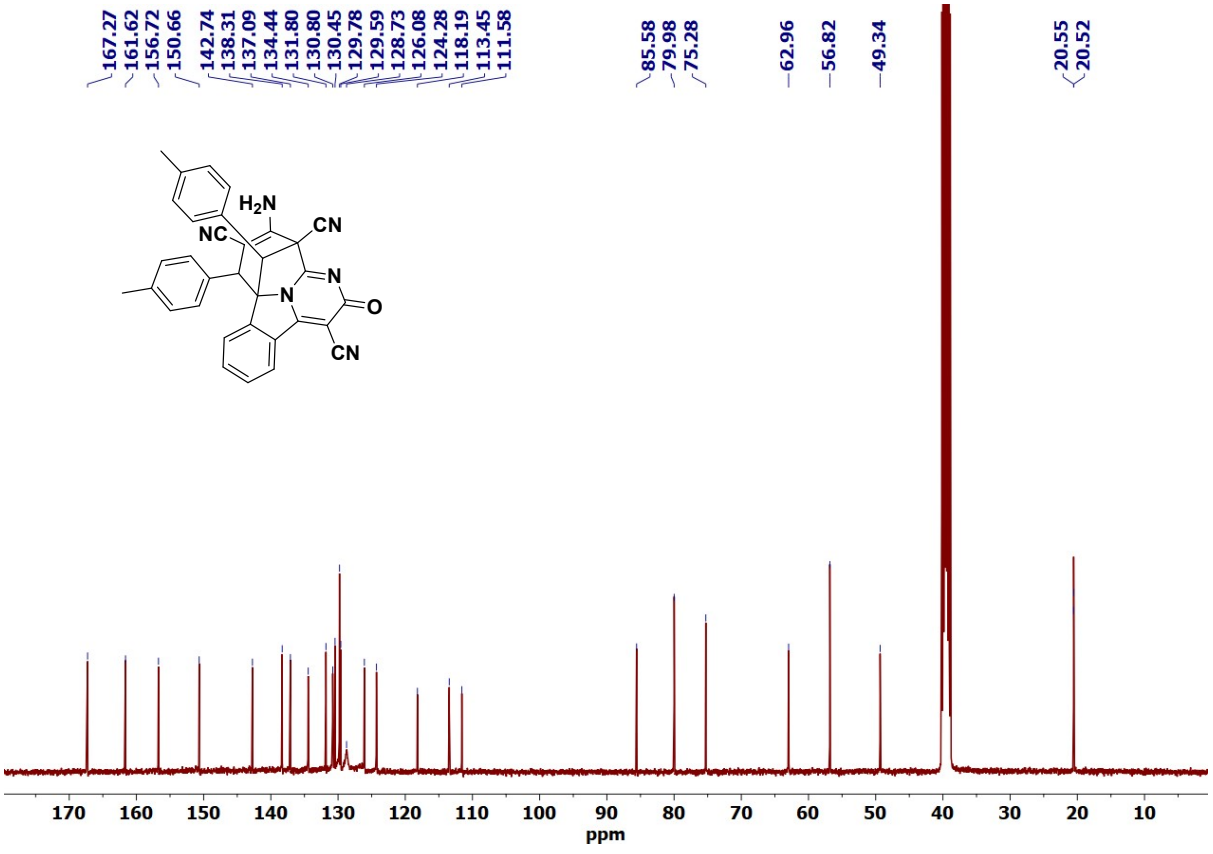
^{13}C NMR spectra of **5c** (101 MHz, DMSO-d_6)



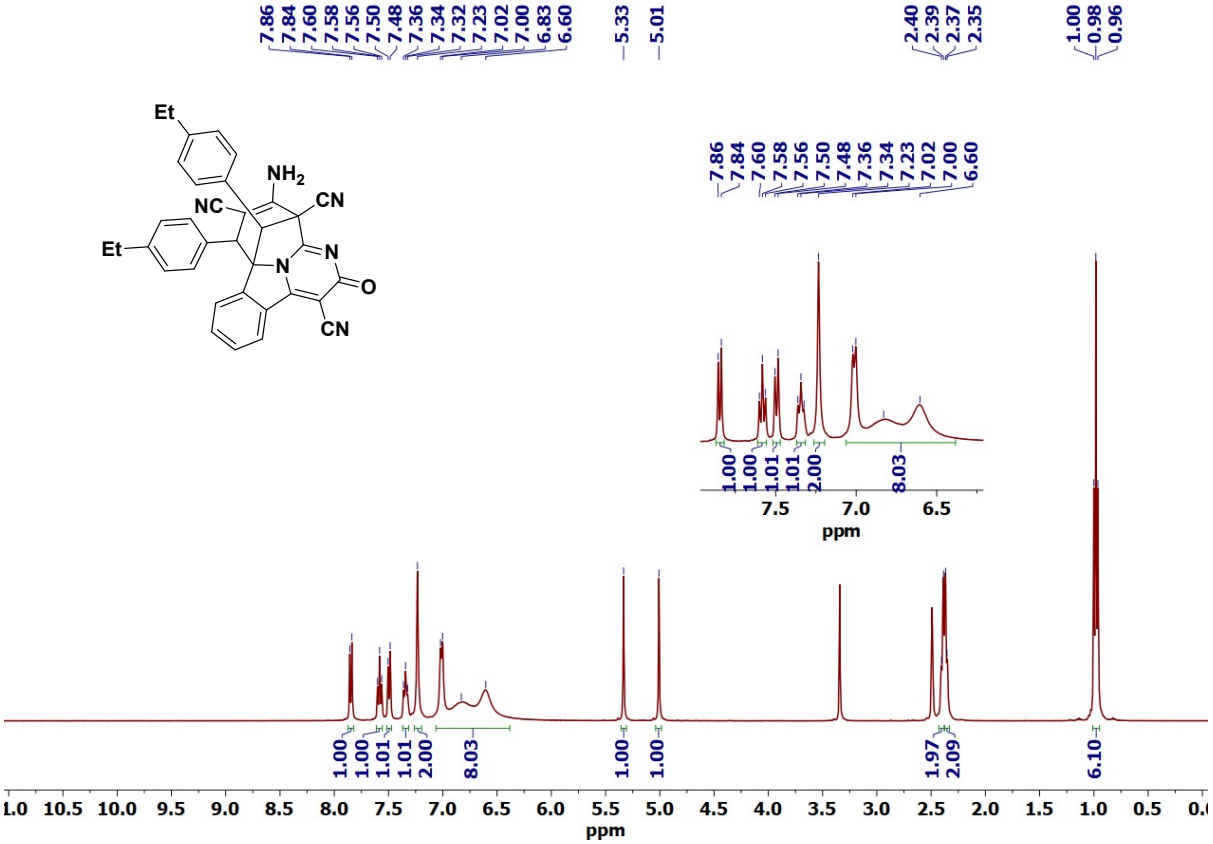
^{1}H NMR spectra of **5d** (400 MHz, DMSO-d_6)



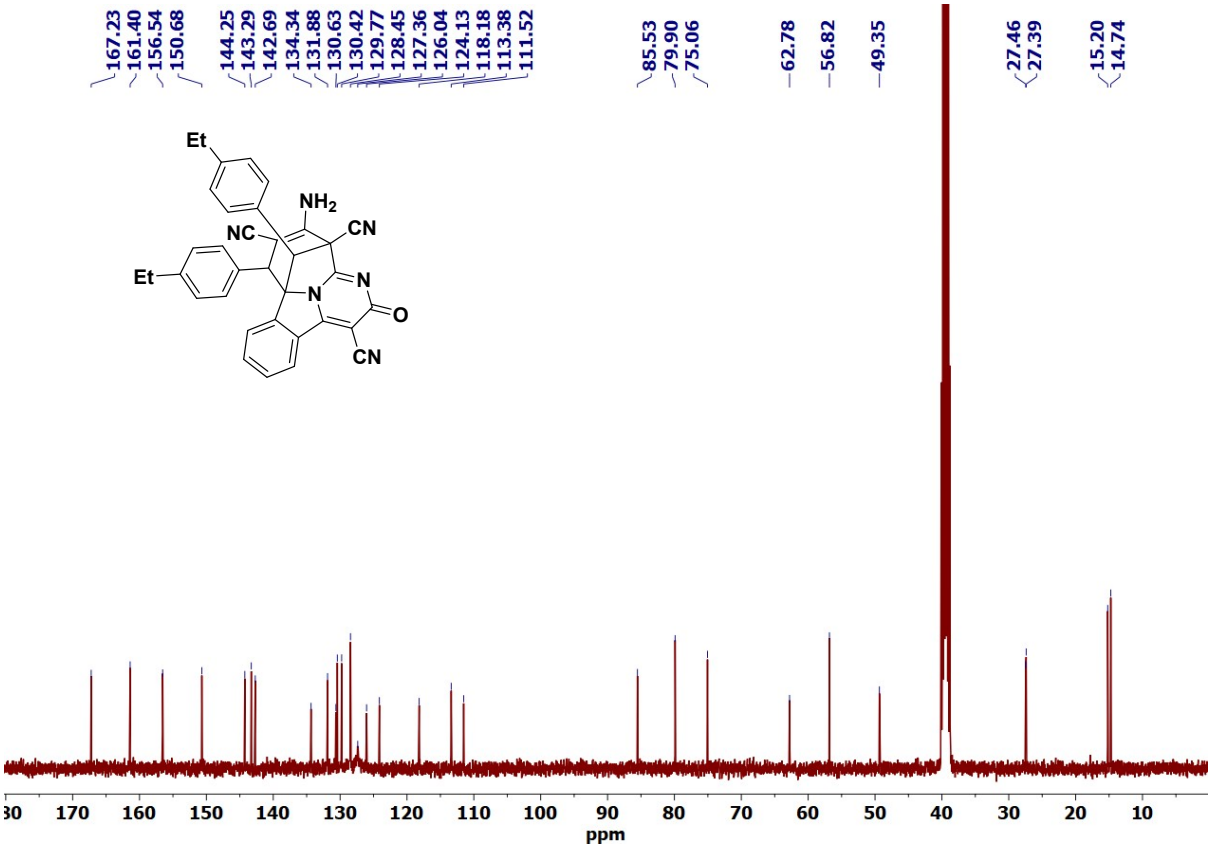
^{13}C NMR spectra of **5d** (101 MHz, DMSO-d_6)



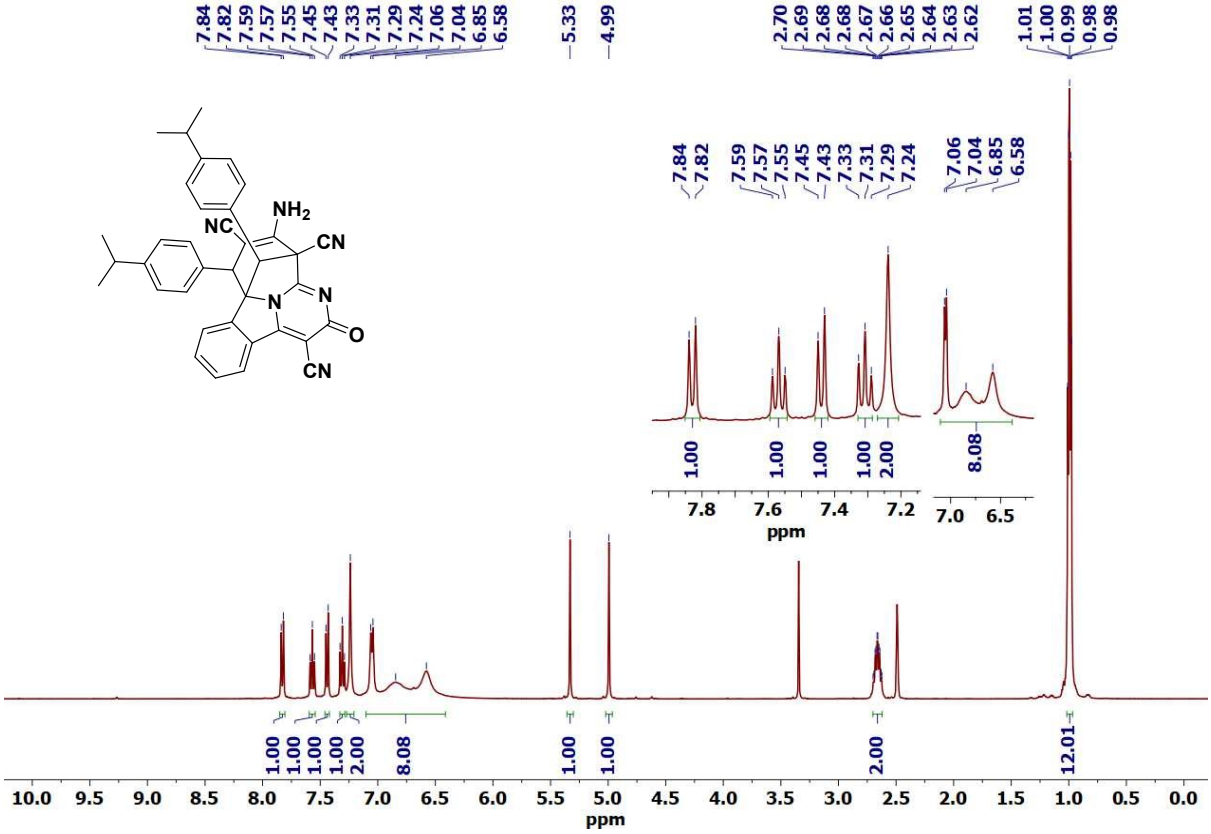
^{1}H NMR spectra of **5e** (400 MHz, DMSO-d_6)



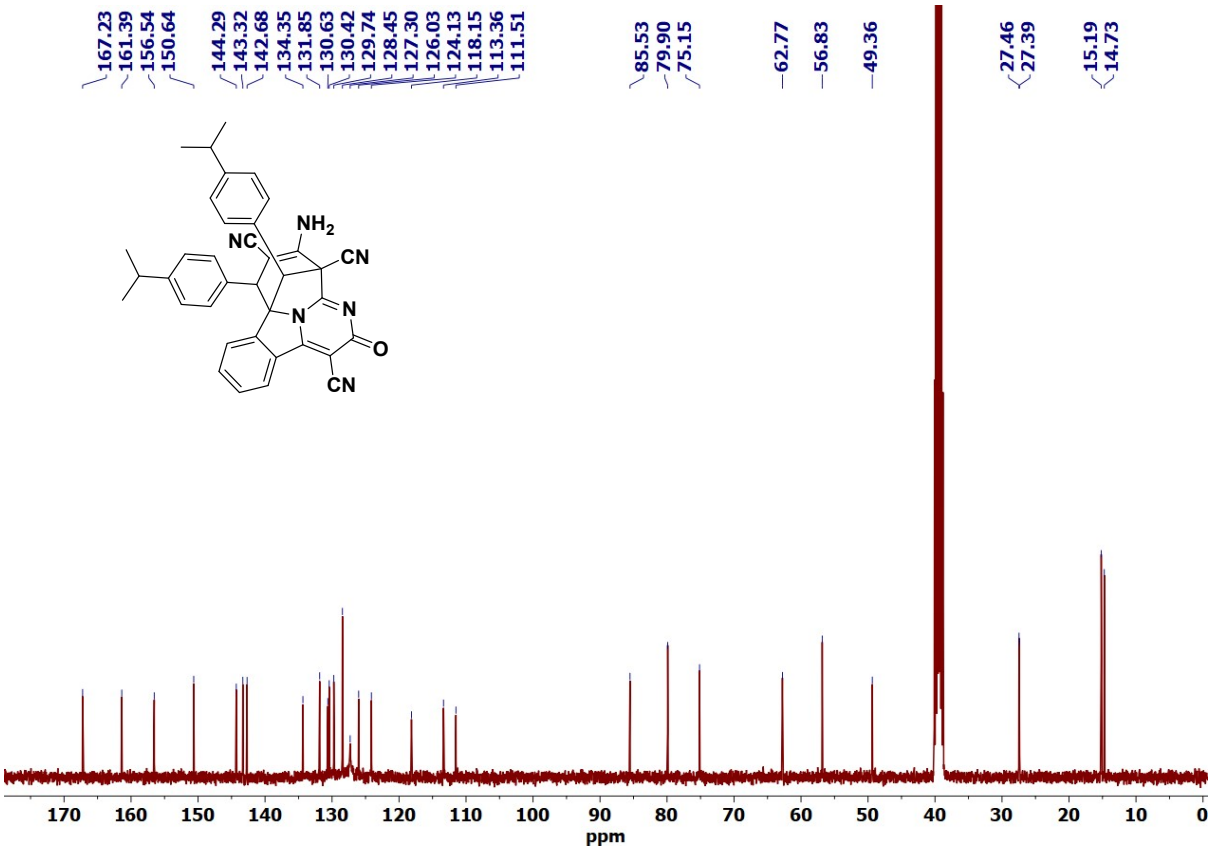
^{13}C NMR spectra of **5e** (101 MHz, DMSO-d_6)



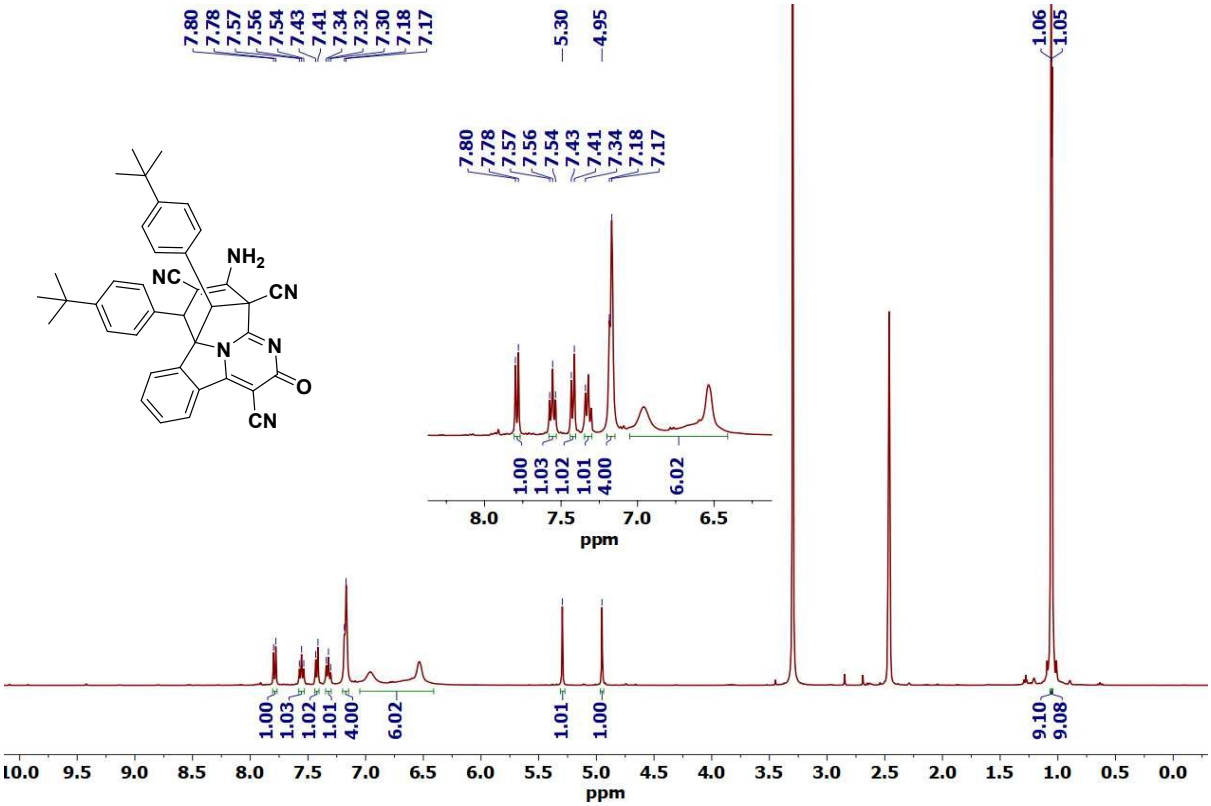
^{1}H NMR spectra of **5f** (400 MHz, DMSO-d_6)



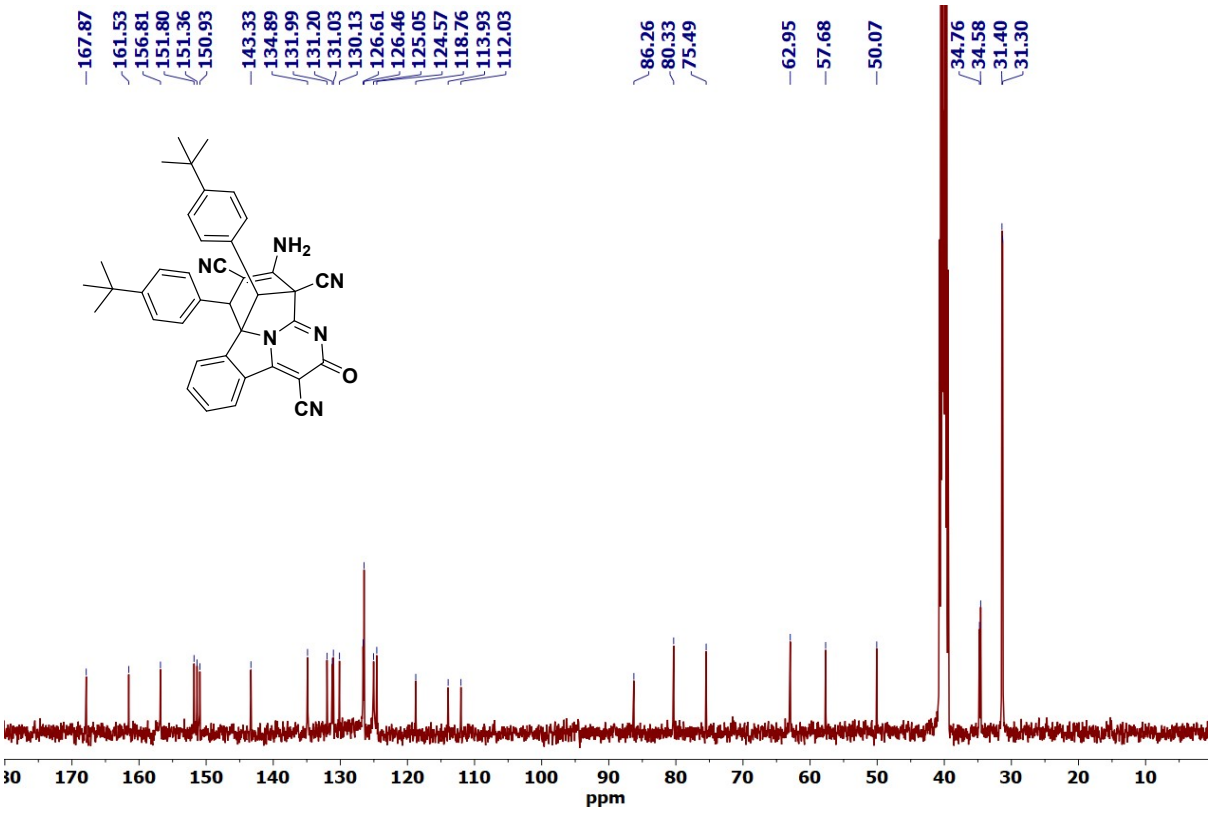
^{13}C NMR spectra of **5f** (101 MHz, DMSO-d_6)



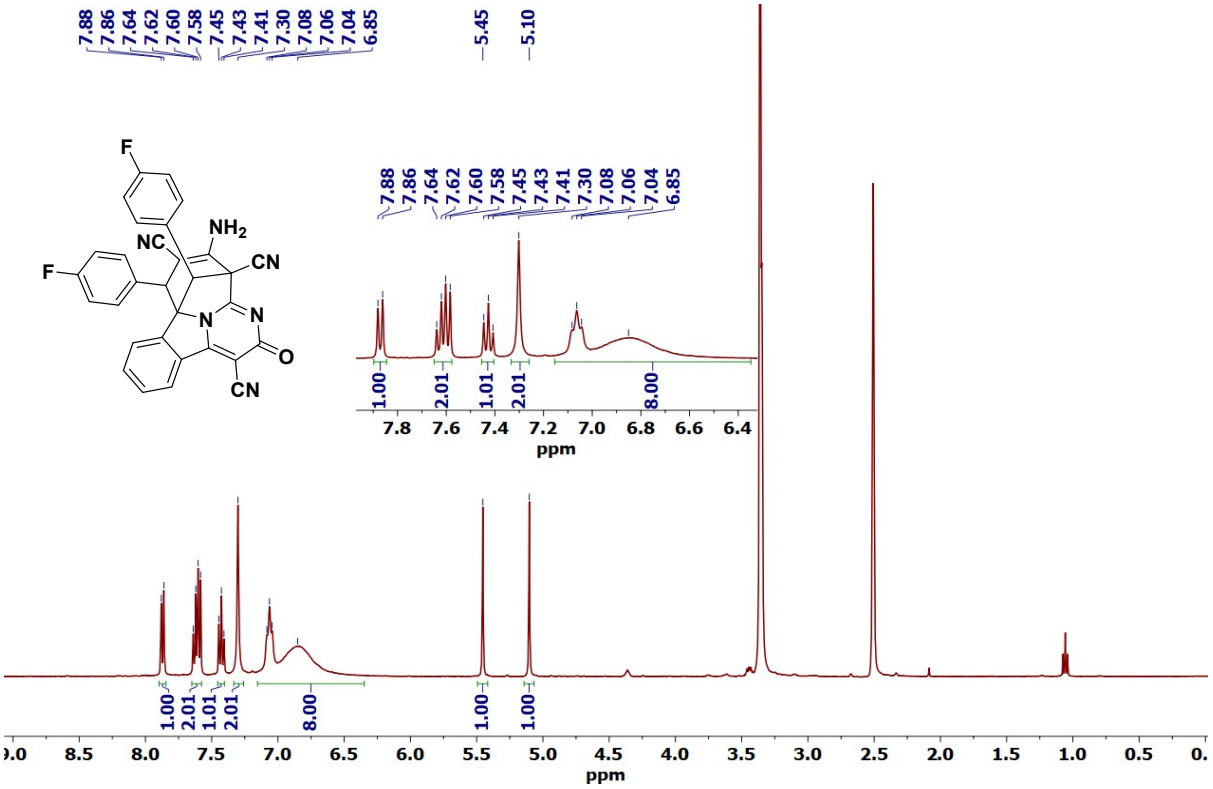
^{1}H NMR spectra of **5g** (400 MHz, DMSO-d_6)



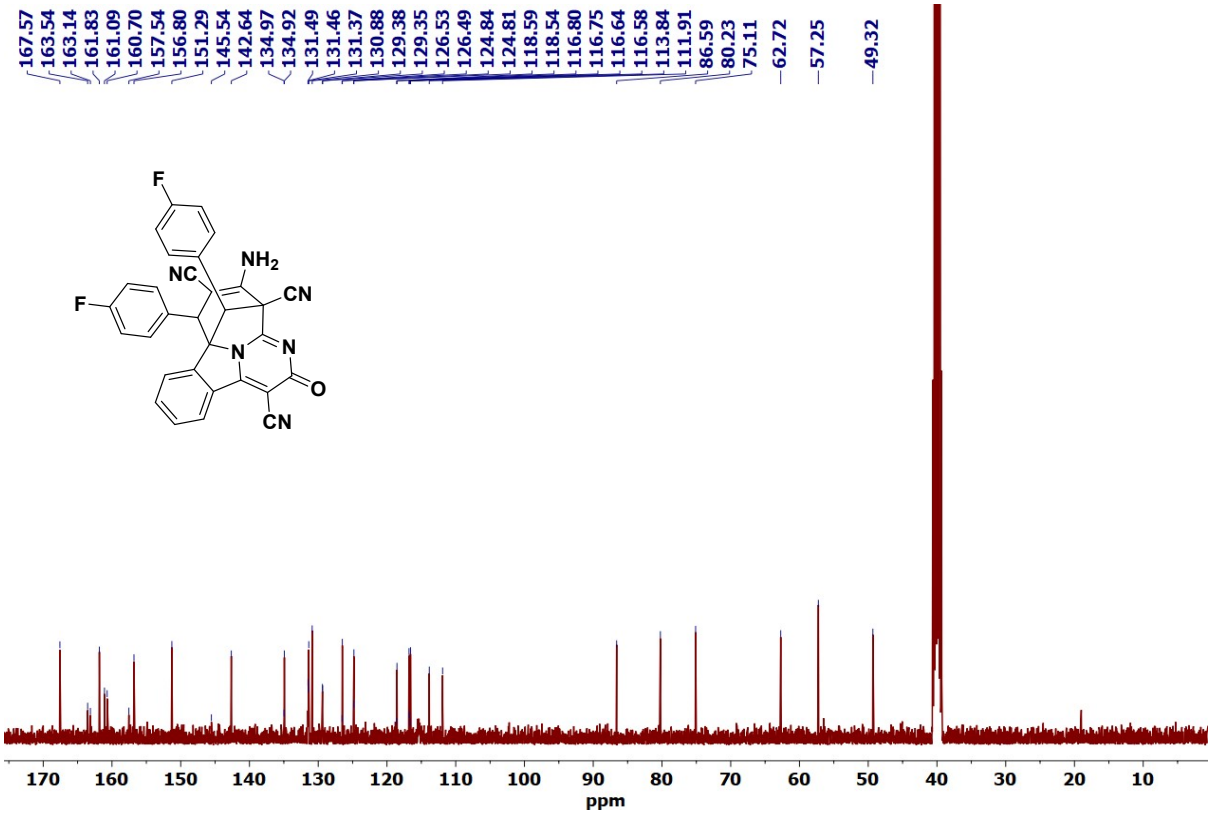
^{13}C NMR spectra of **5g** (101 MHz, DMSO-d_6)



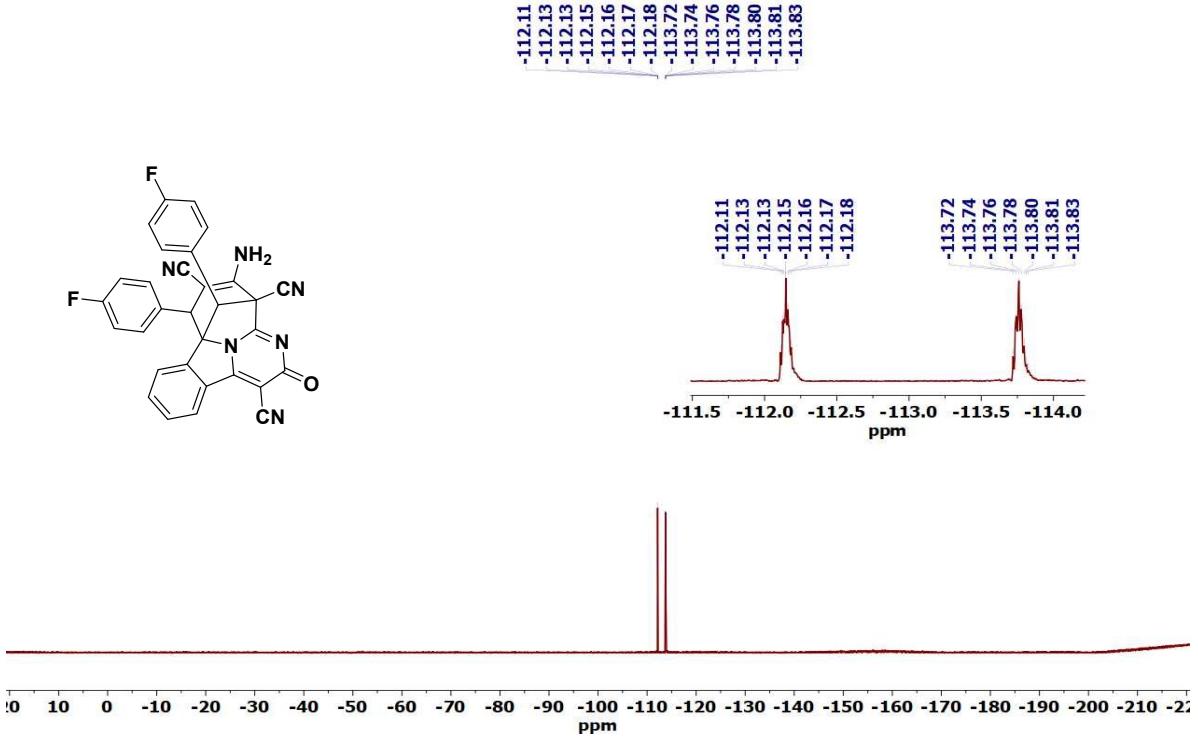
^{1}H NMR spectra of **5h** (400 MHz, DMSO-d_6)



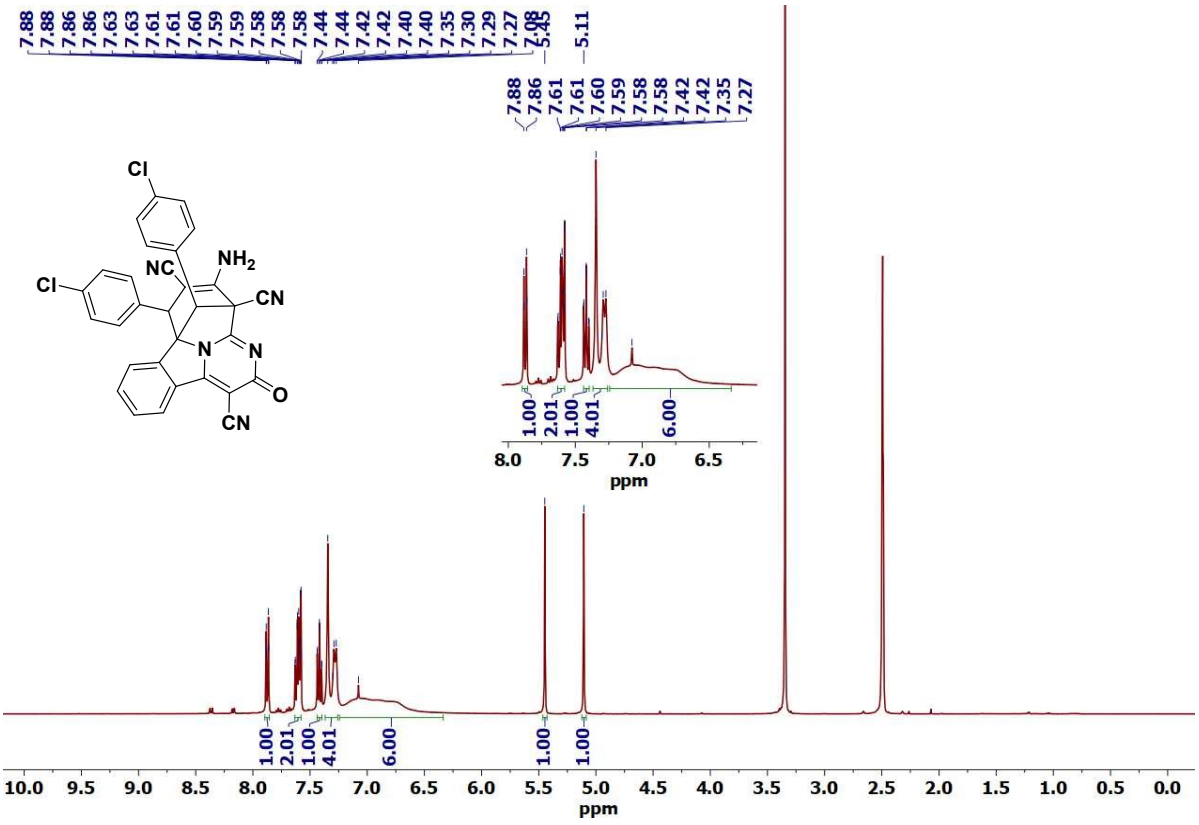
^{13}C NMR spectra of **5h** (101 MHz, DMSO-d_6)



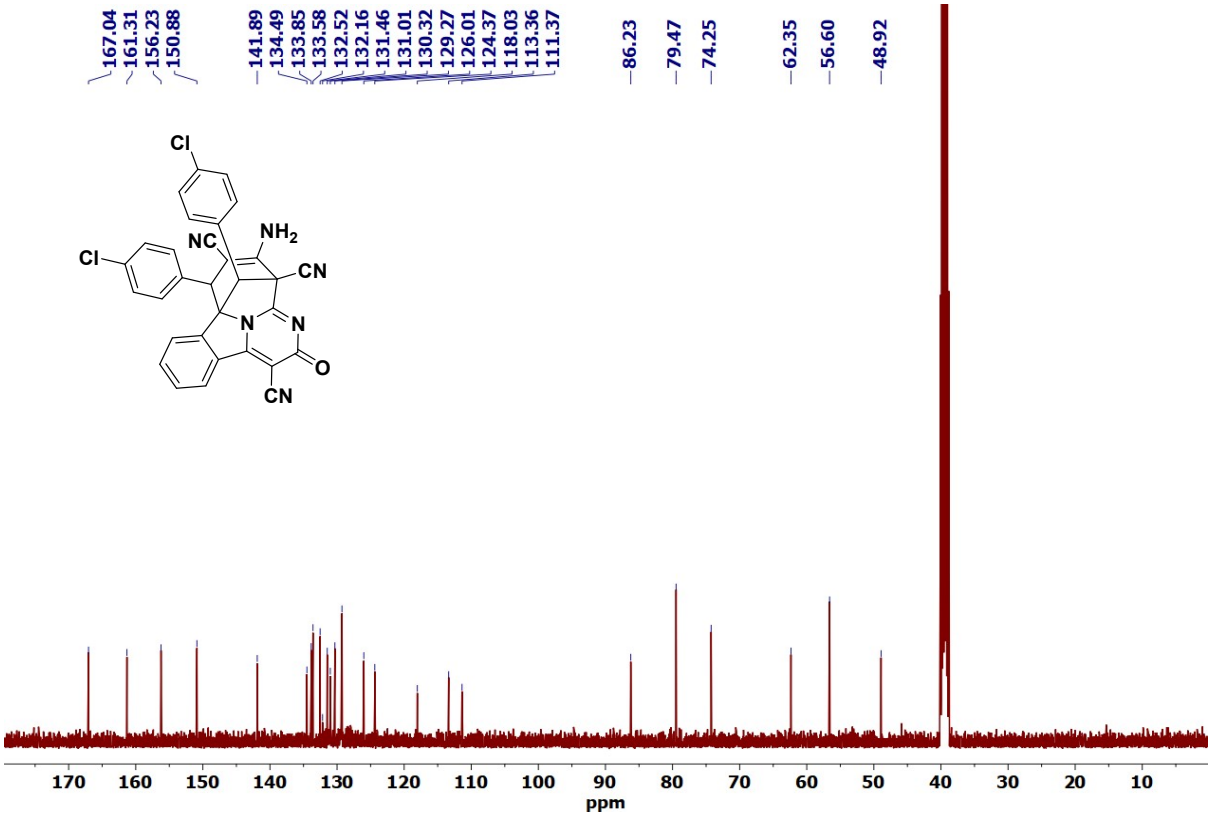
^{19}F NMR spectra of **5h** (376 MHz, DMSO-d_6)



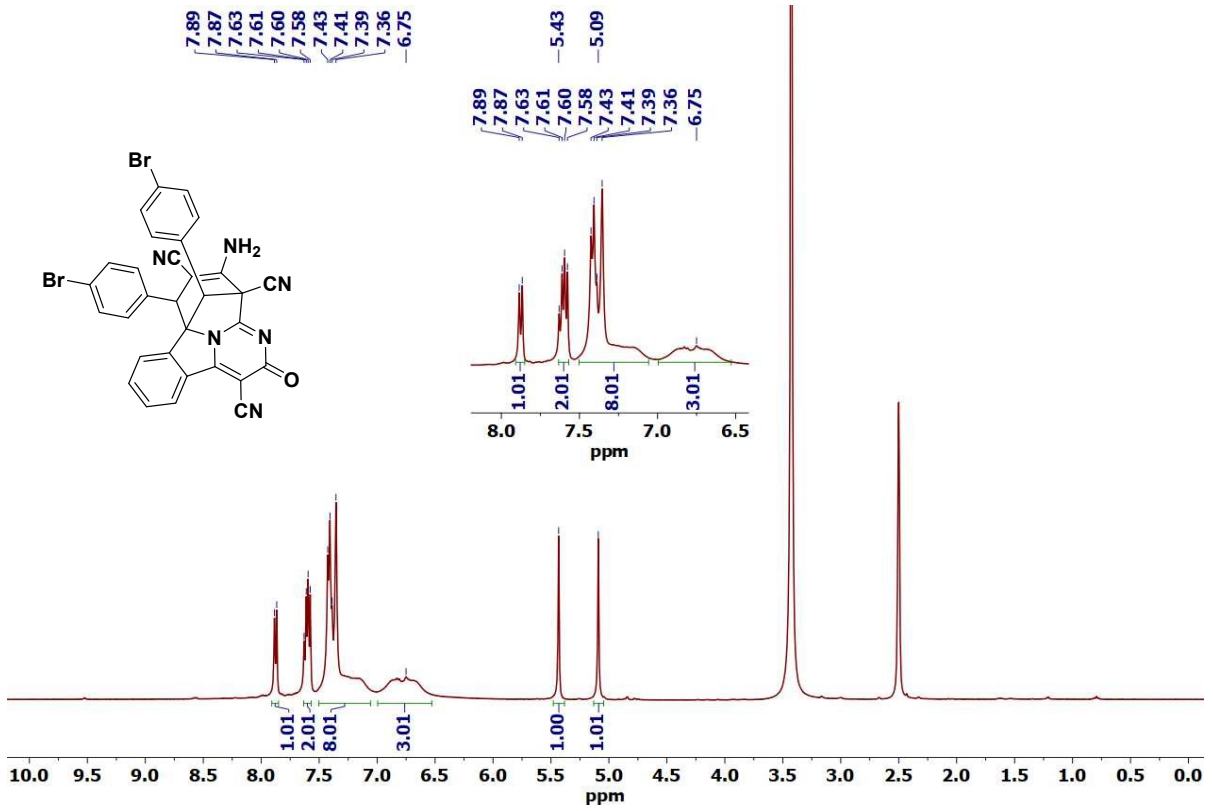
^{1}H NMR spectra of **5i** (400 MHz, DMSO-d_6)



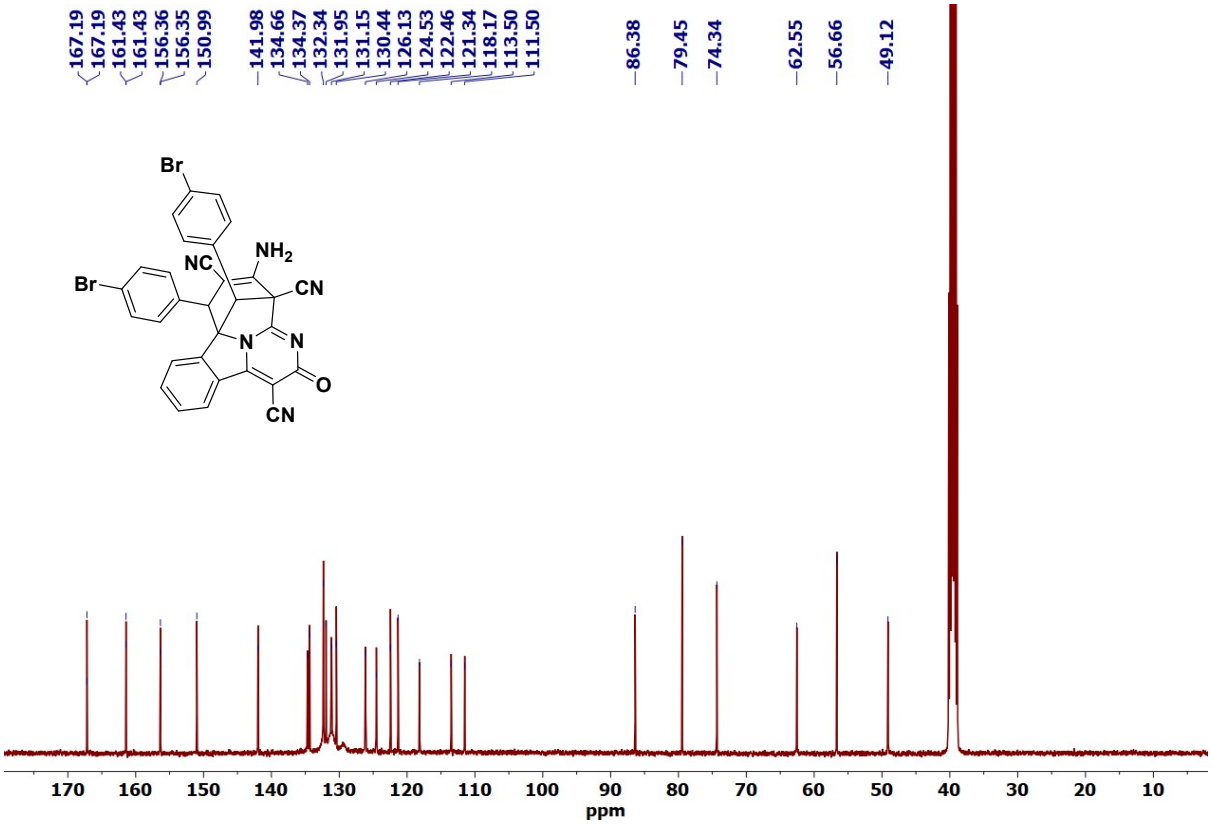
^{13}C NMR spectra of **5i** (101 MHz, DMSO-d_6)



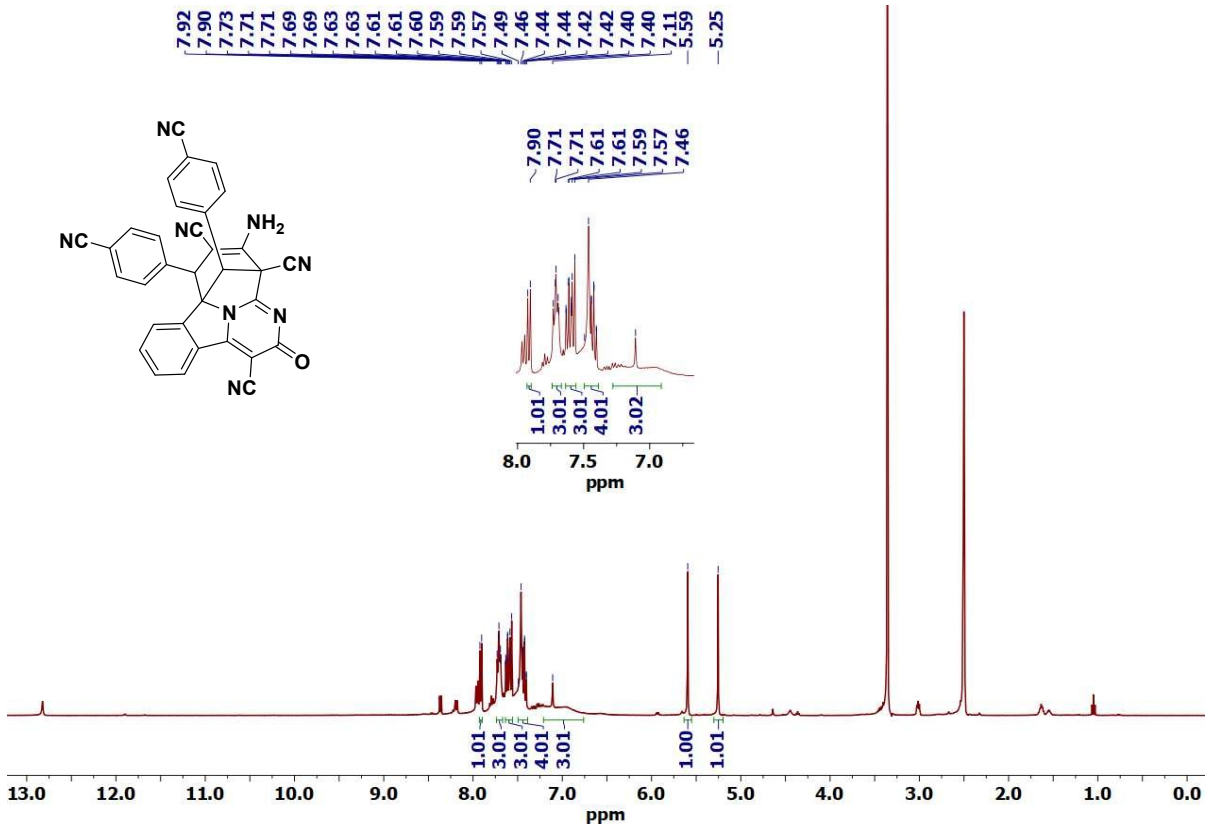
^{1}H NMR spectra of **5j** (400 MHz, DMSO-d_6)



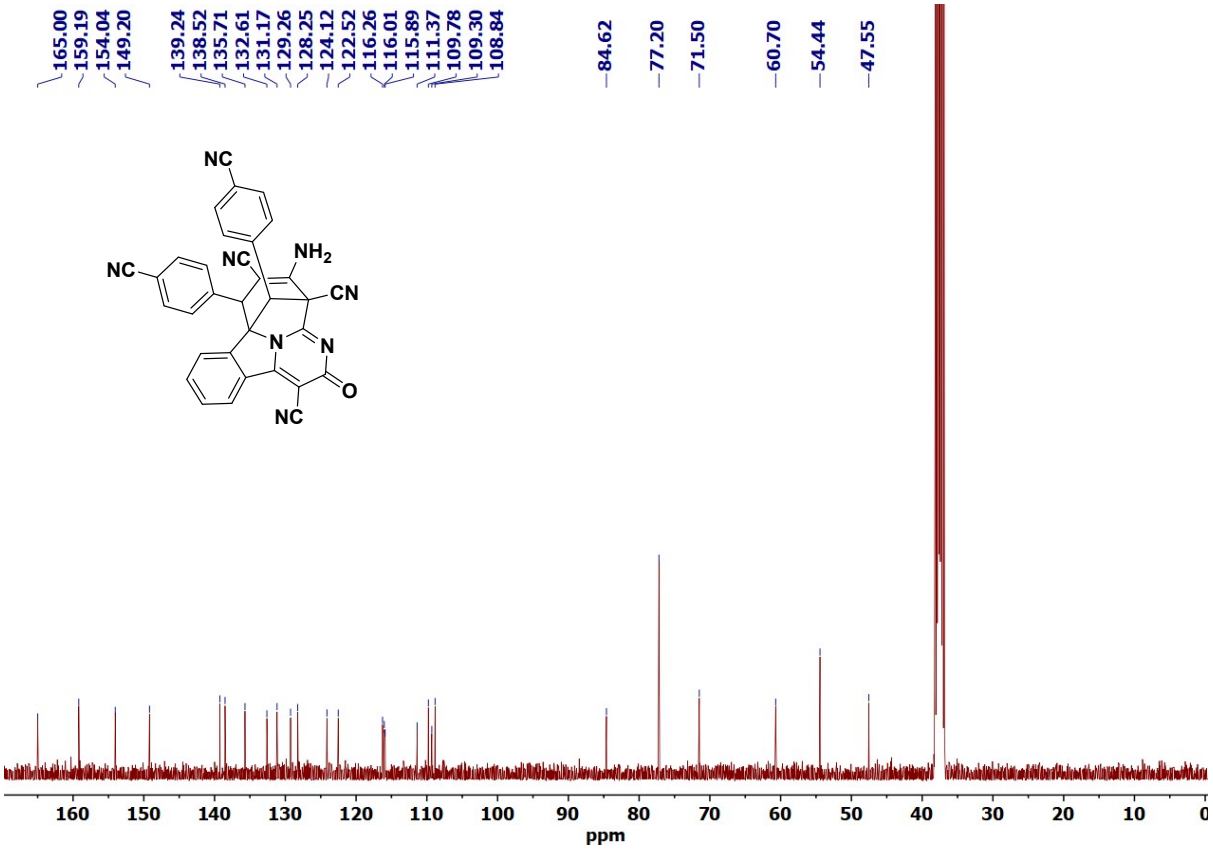
^{13}C NMR spectra of **5j** (101 MHz, DMSO-d_6)



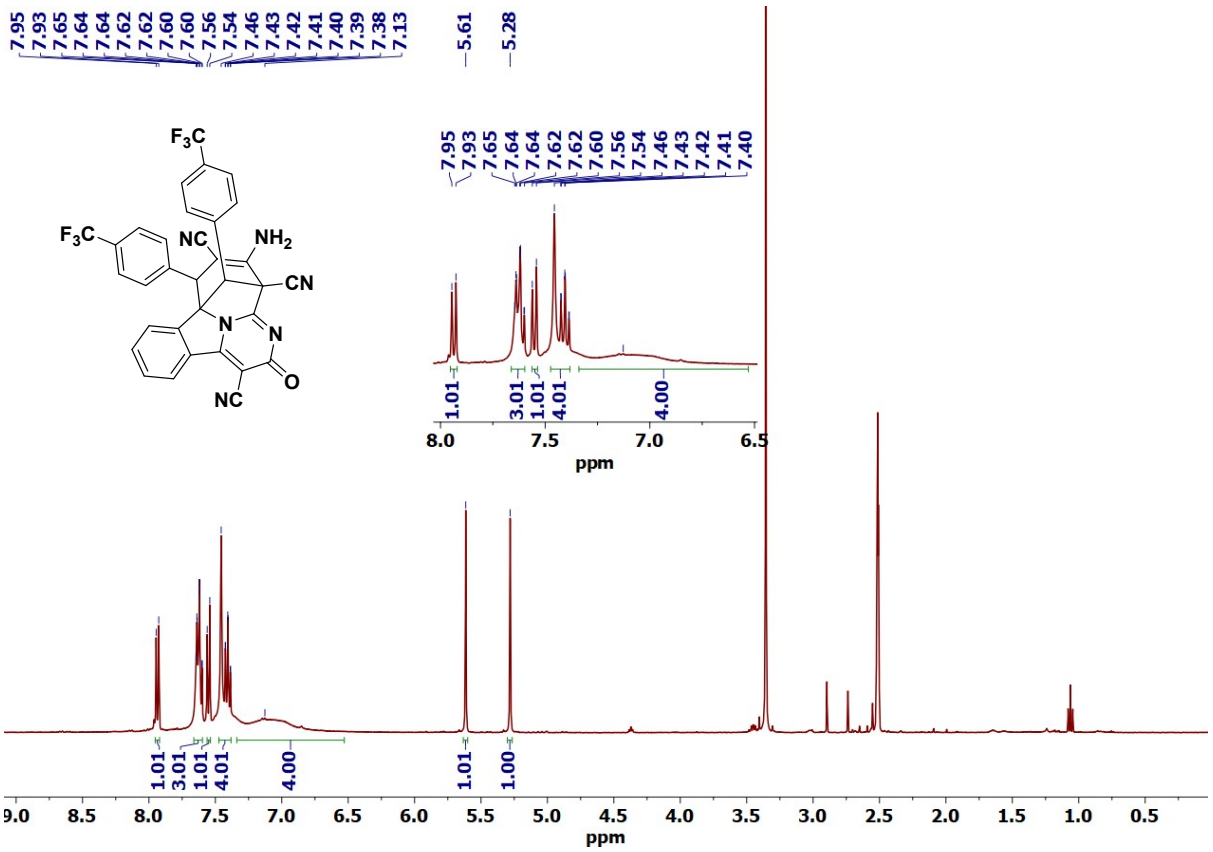
^{1}H NMR spectra of **5k** (400 MHz, DMSO-d_6)



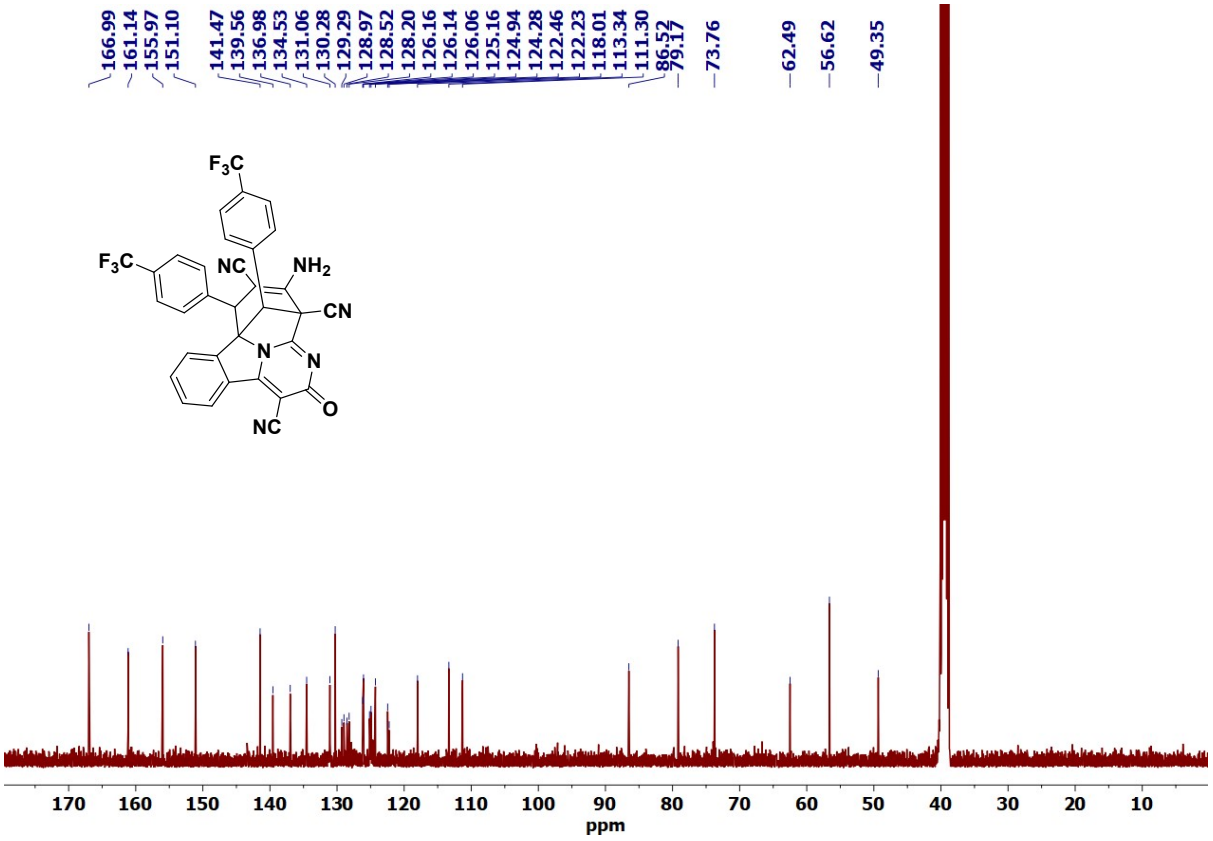
^{13}C NMR spectra of **5k** (101 MHz, DMSO-d_6)



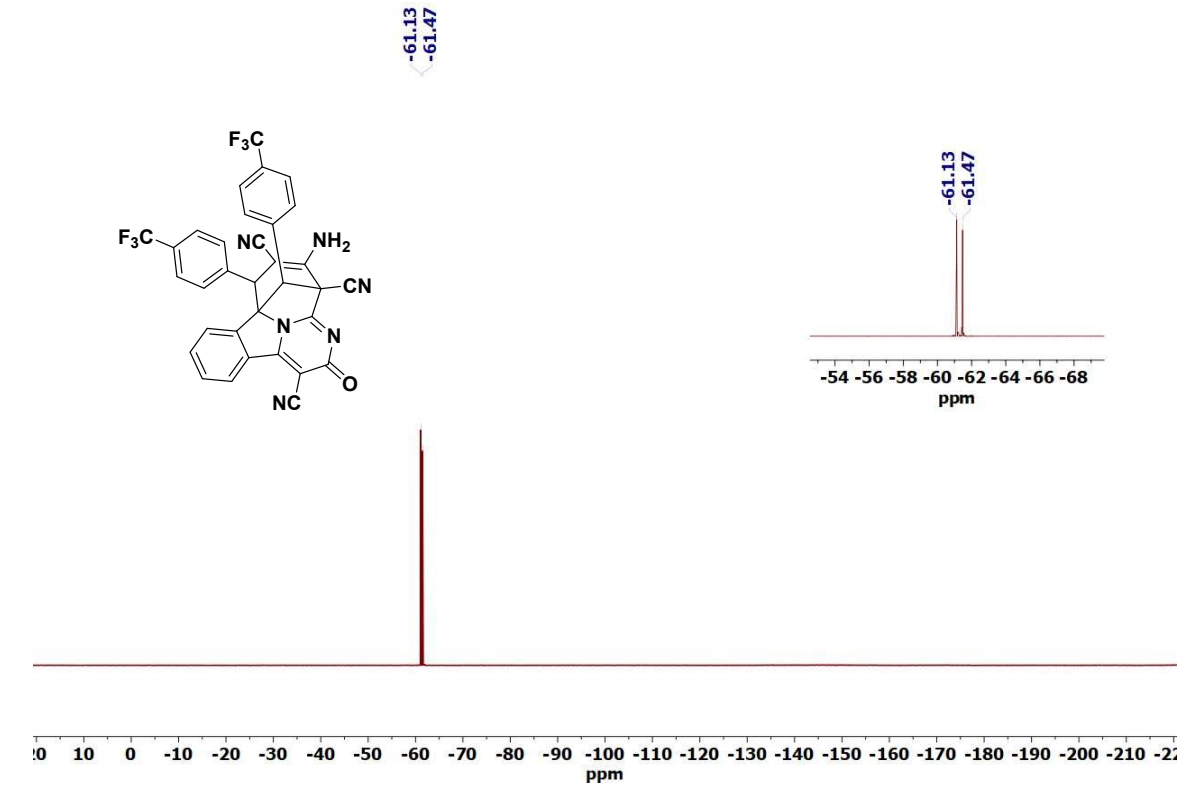
^{1}H NMR spectra of **5l** (400 MHz, DMSO-d_6)



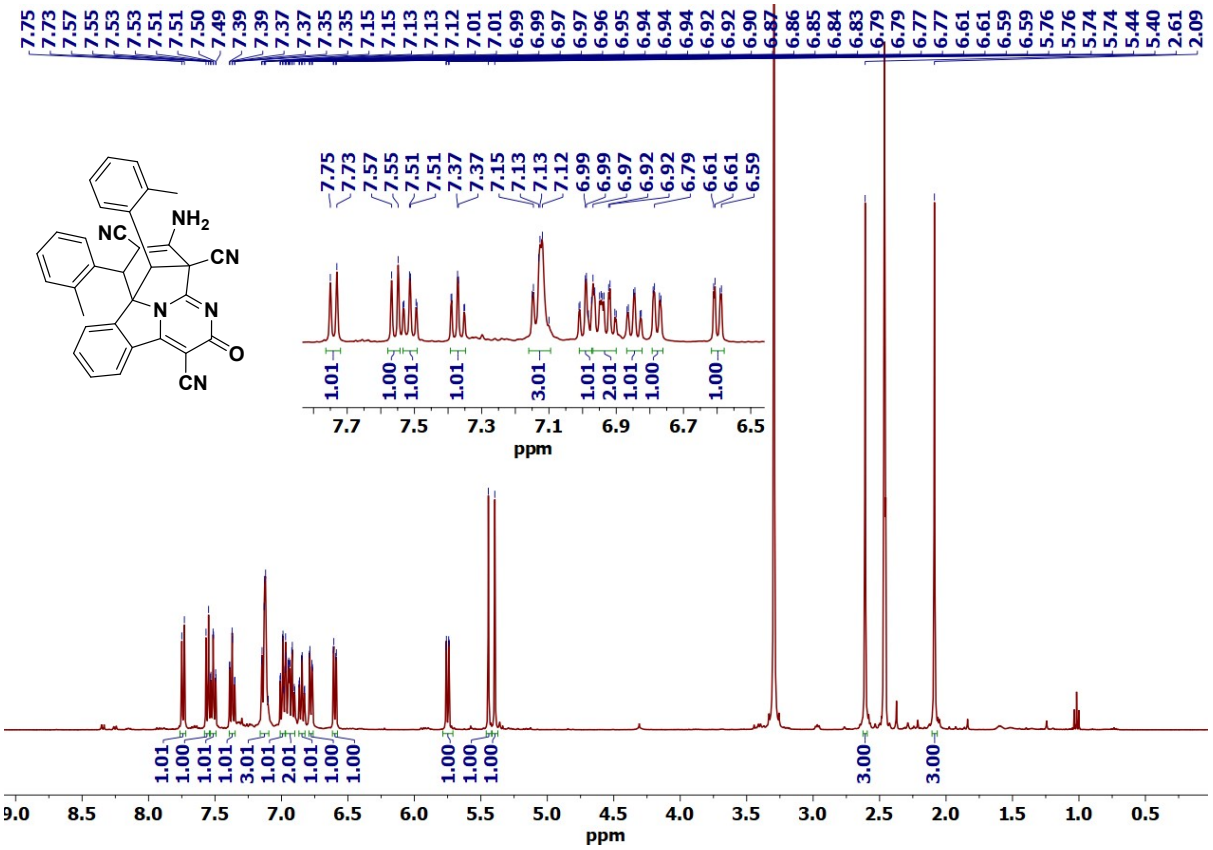
^{13}C NMR spectra of **5l** (101 MHz, DMSO-d_6)



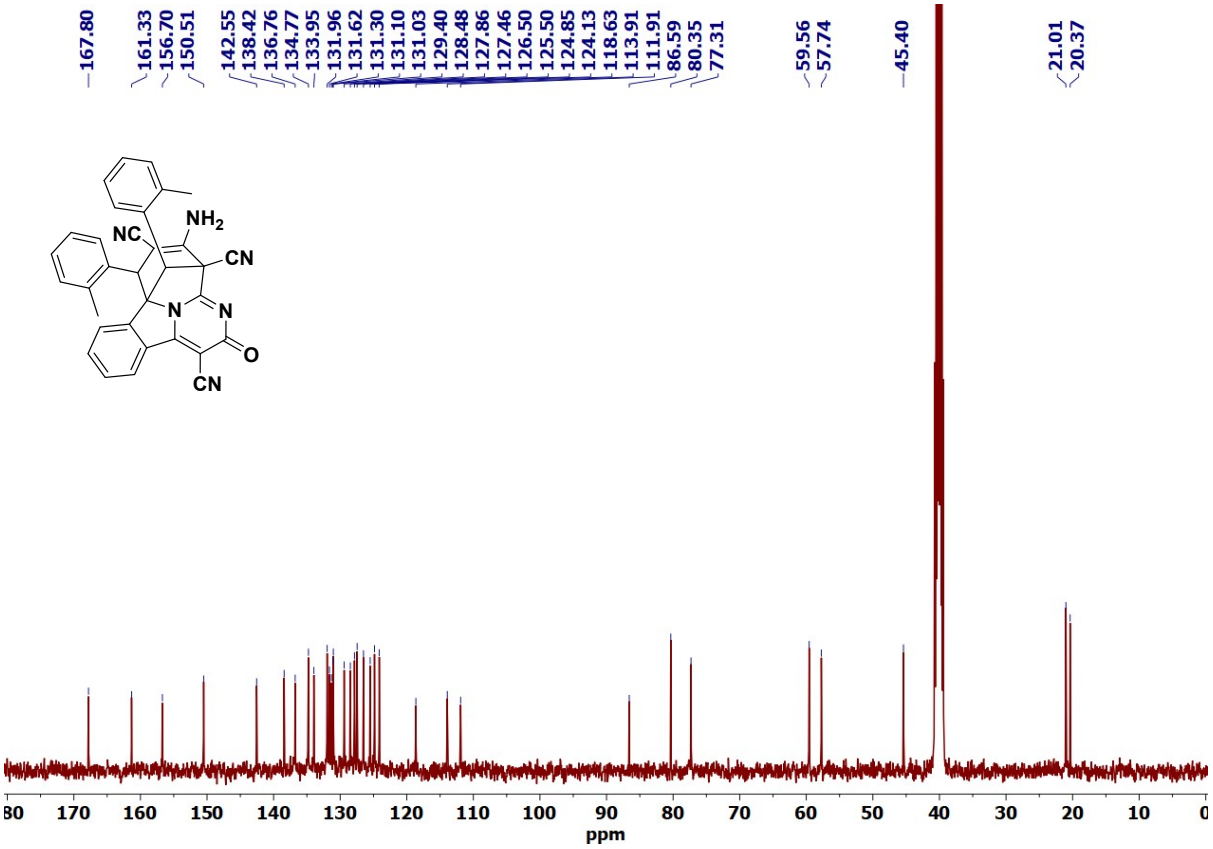
^{19}F NMR spectra of **5l** (376 MHz, DMSO-d_6)



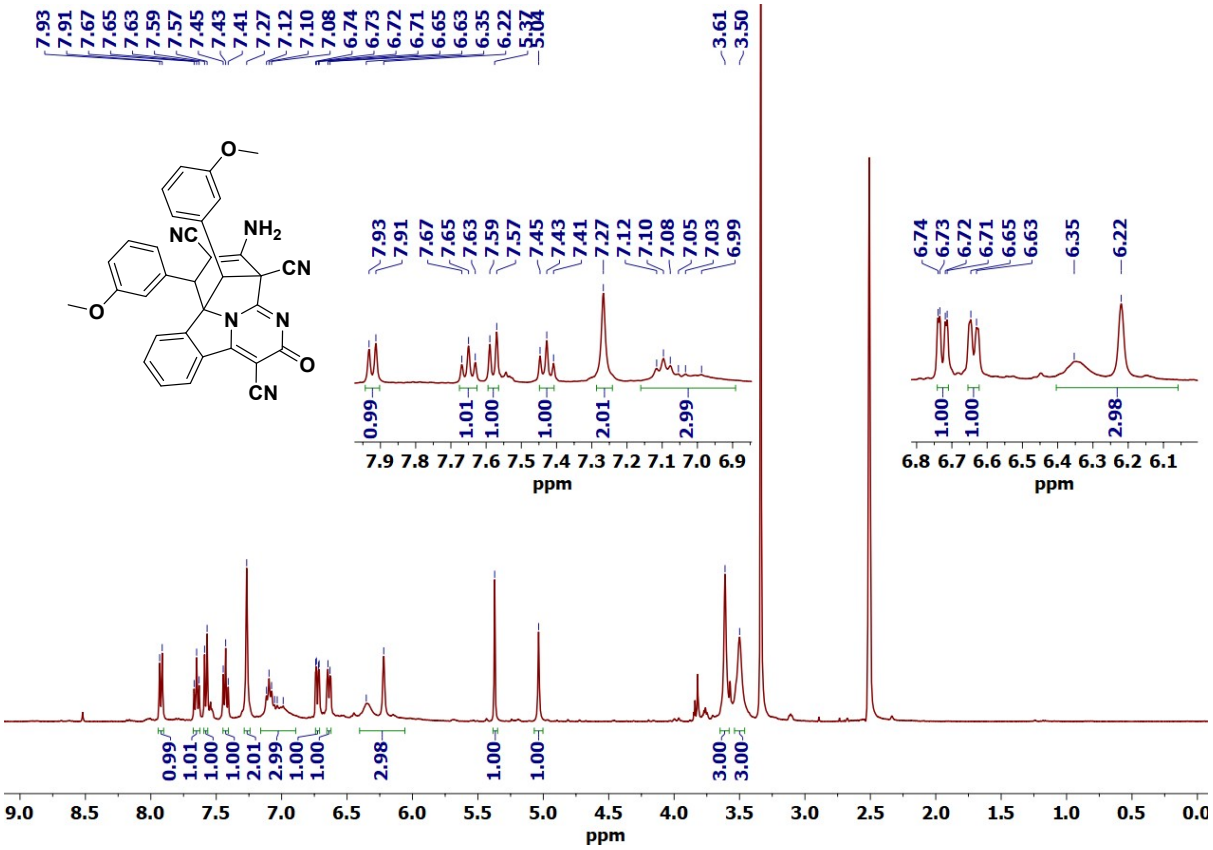
^{1}H NMR spectra of **5m** (400 MHz, DMSO-d_6)



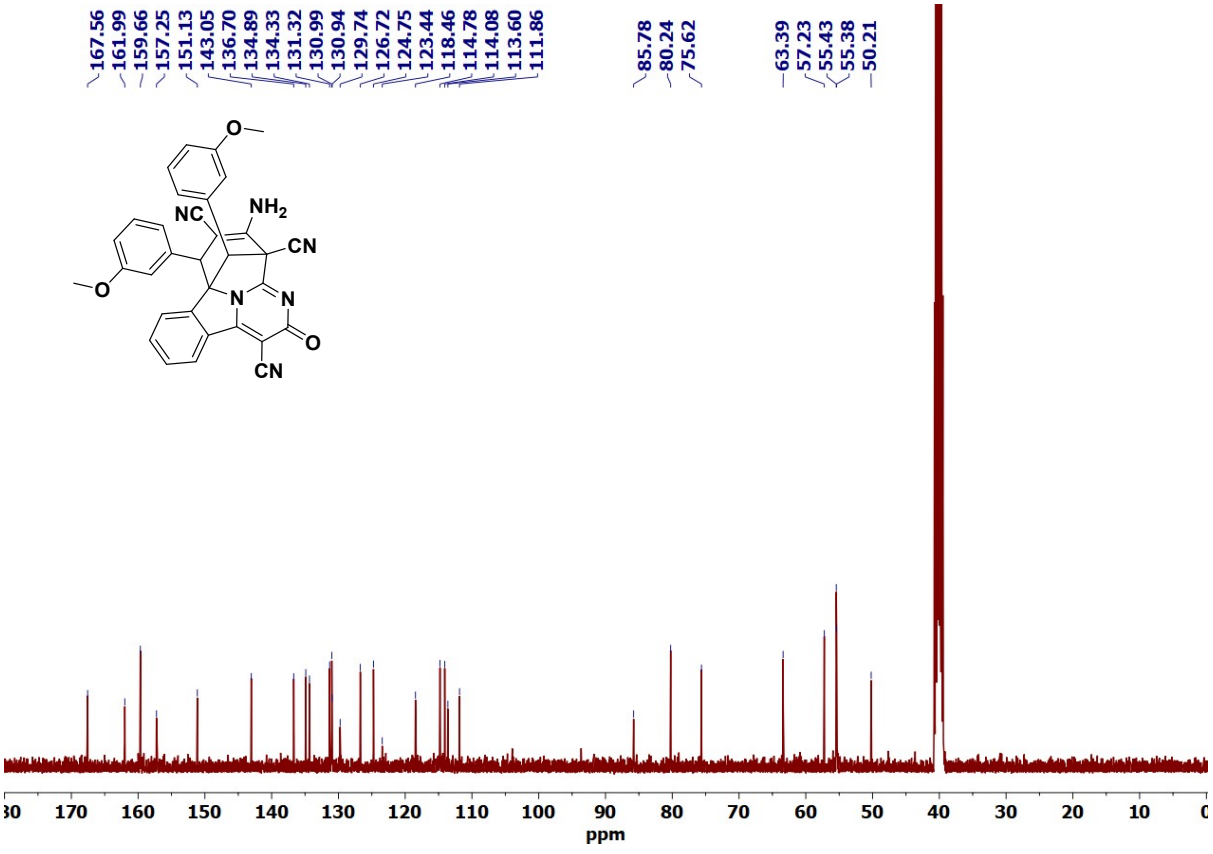
^{13}C NMR spectra of **5m** (101 MHz, DMSO-d_6)



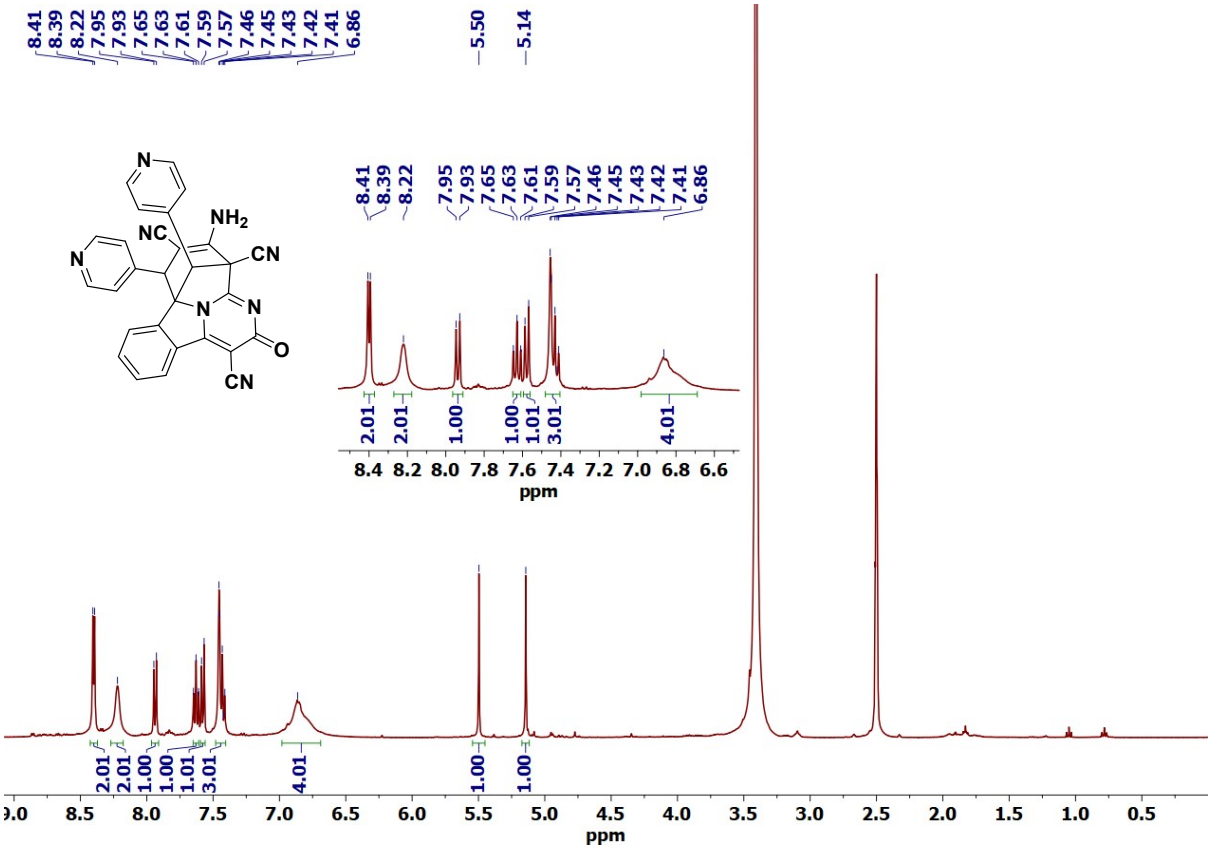
^{1}H NMR spectra of **5n** (400 MHz, DMSO-d_6)



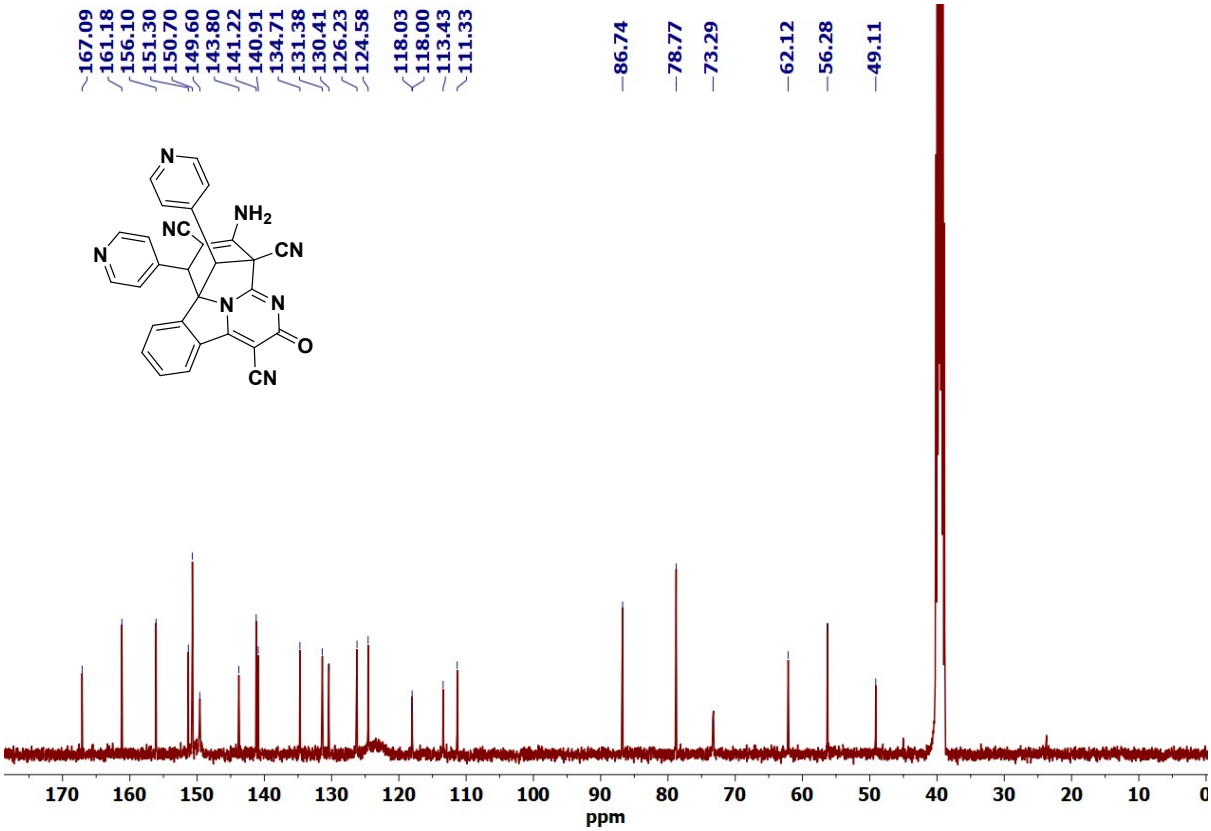
^{13}C NMR spectra of **5n** (101 MHz, DMSO-d_6)



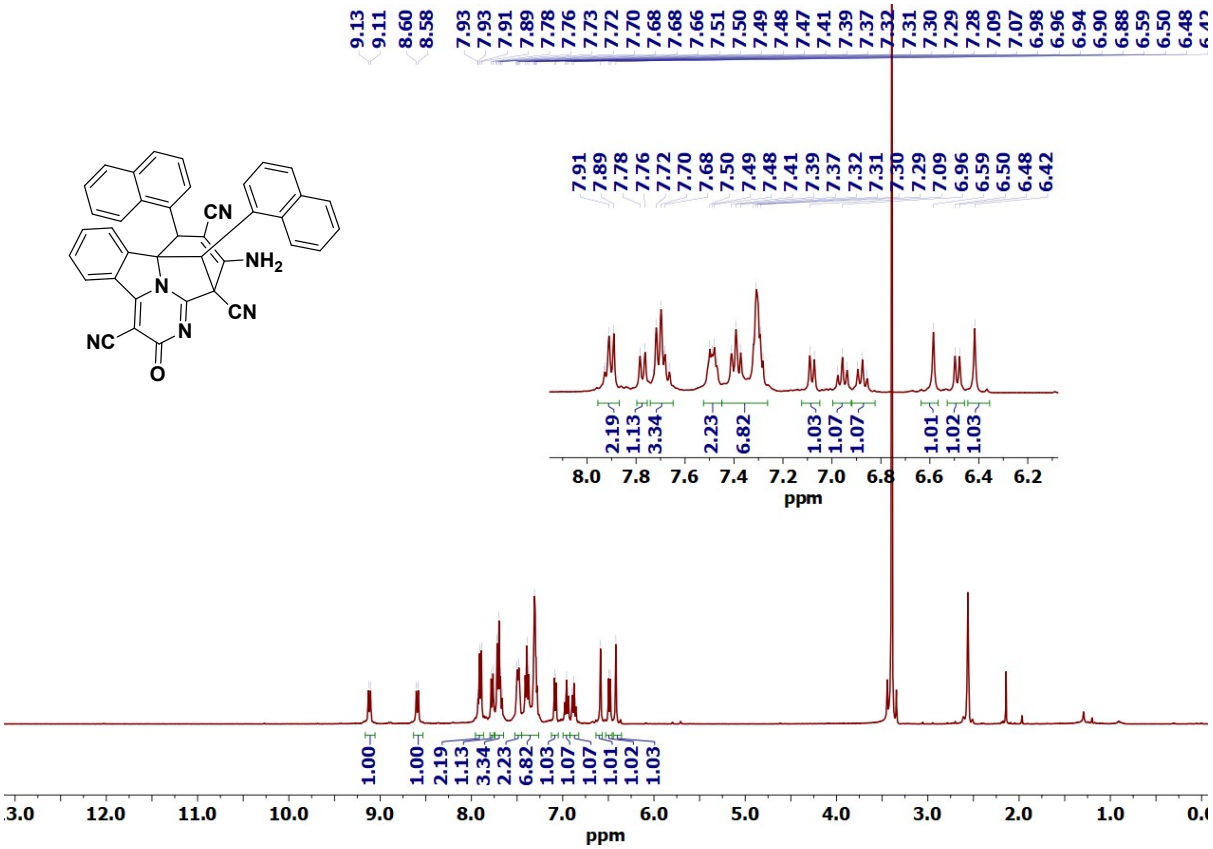
^{1}H NMR spectra of **5o** (400 MHz, DMSO-d_6)



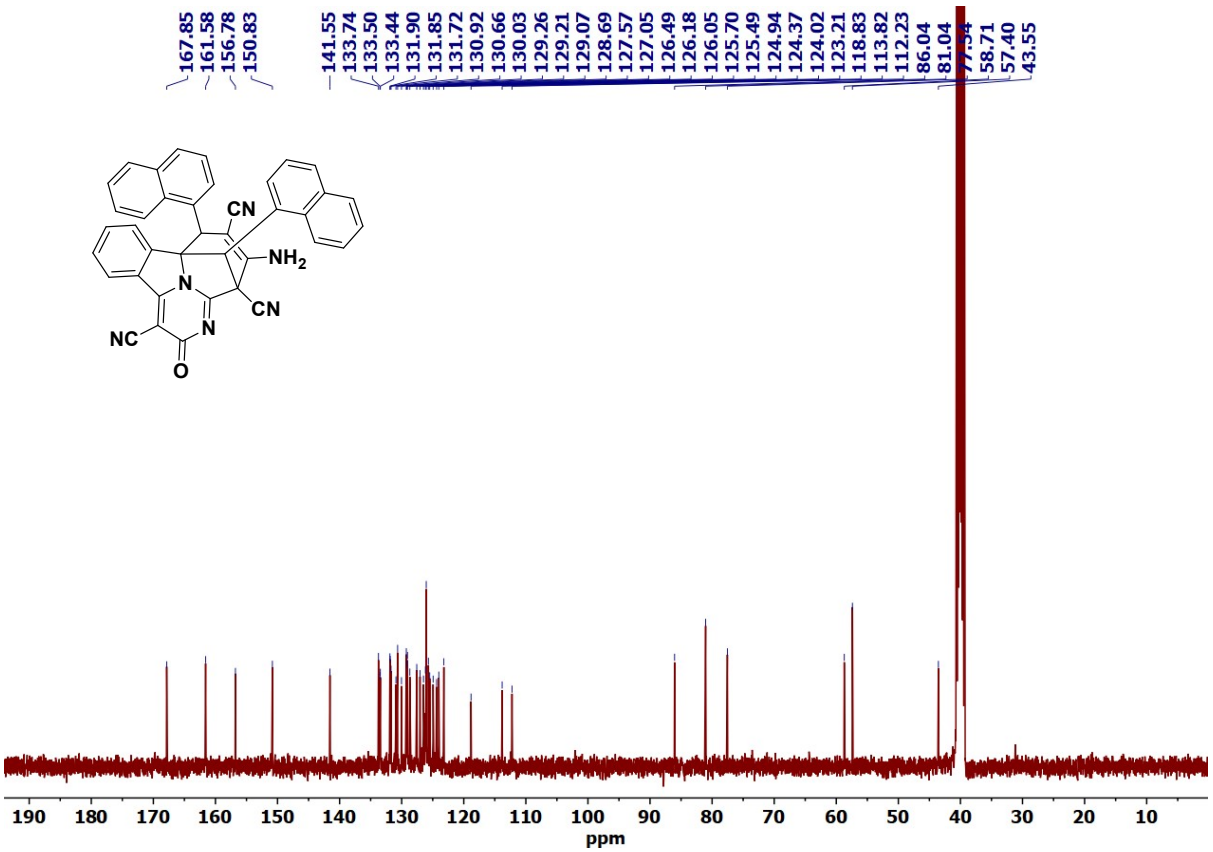
^{13}C NMR spectra of **5o** (101 MHz, DMSO-d_6)



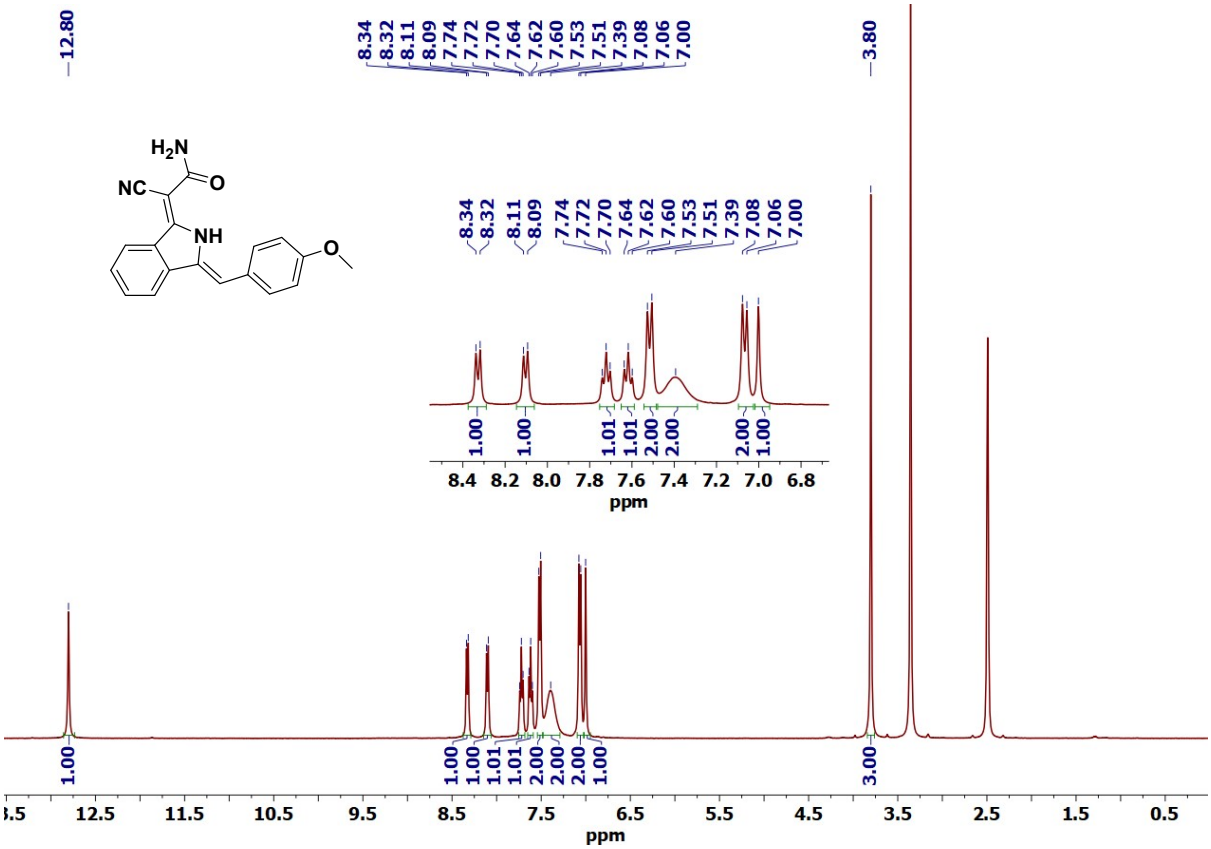
^{1}H NMR spectra of **5p** (400 MHz, DMSO-d_6)



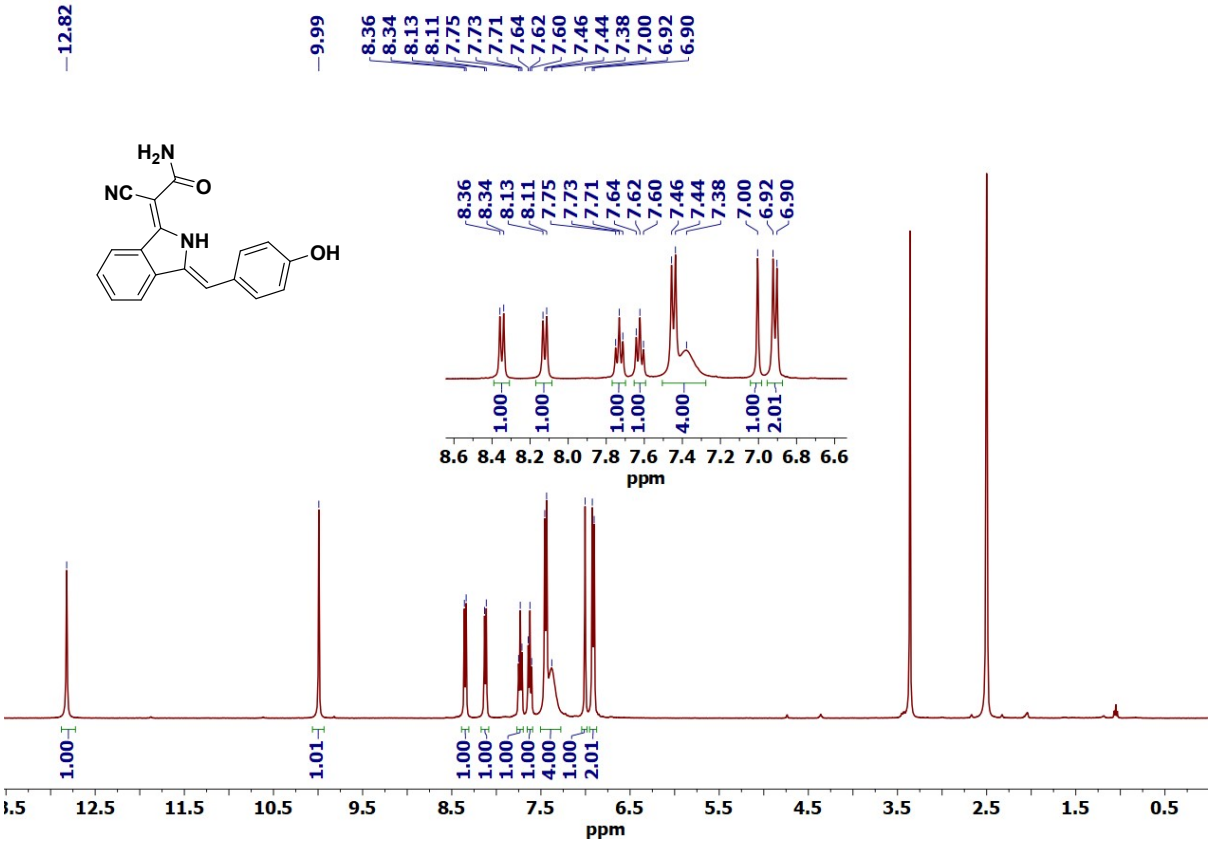
^{13}C NMR spectra of **5p** (101 MHz, DMSO-d_6)



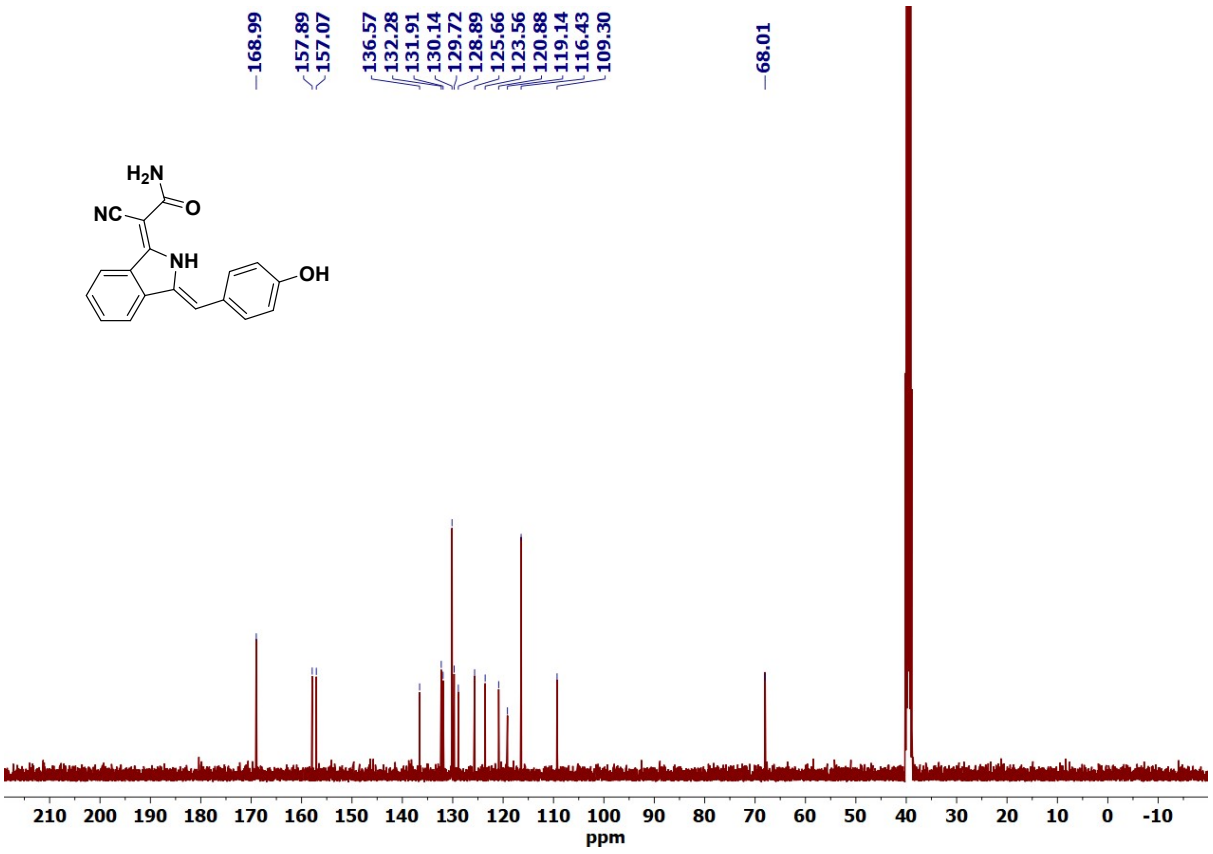
^{1}H NMR spectra of 6**a** (400 MHz, DMSO-d_6)



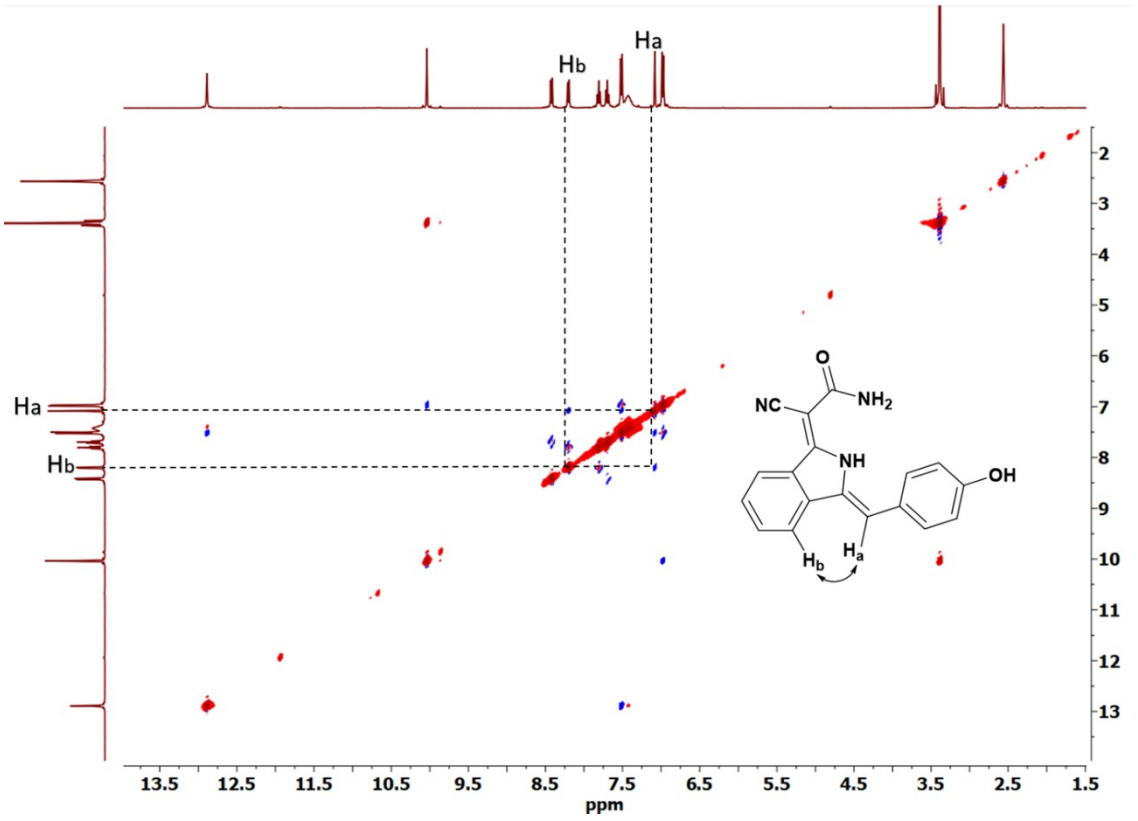
^{1}H NMR spectra of **6b** (400 MHz, DMSO-d_6)



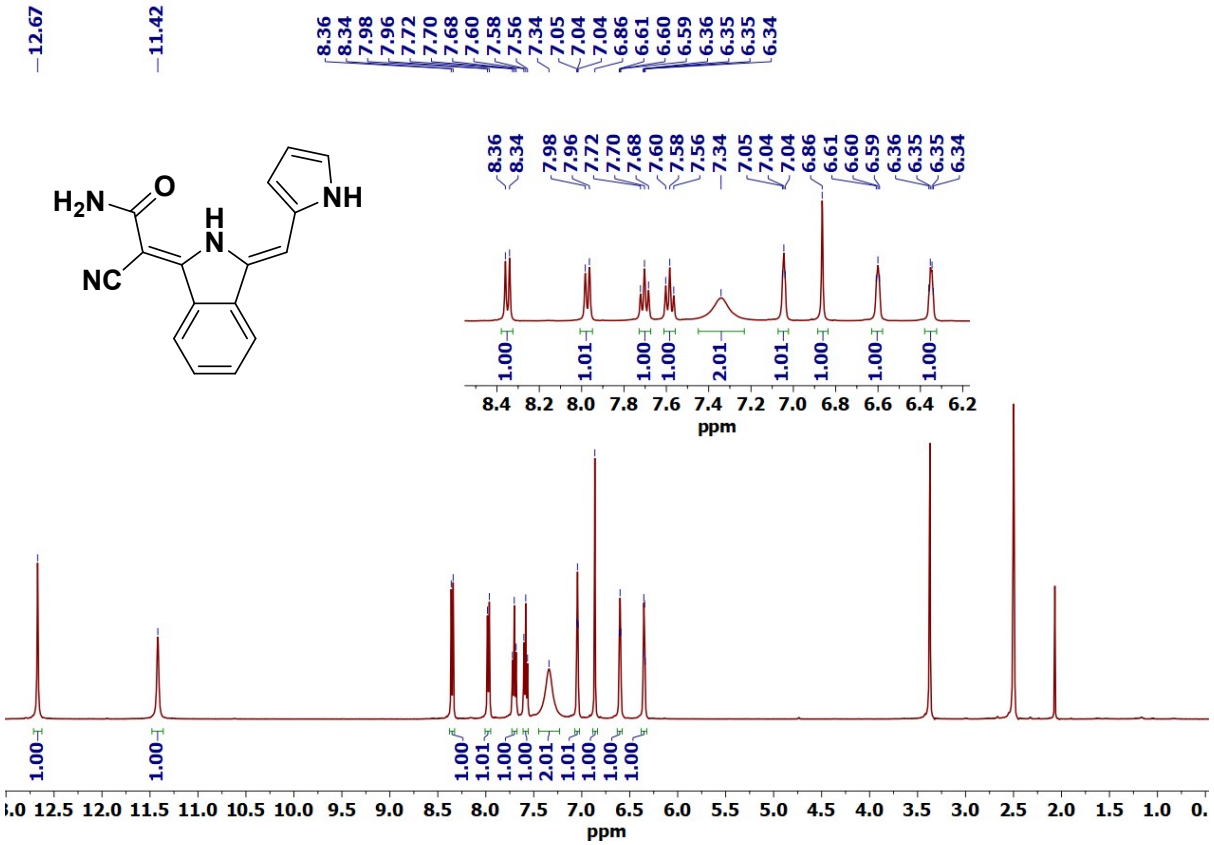
^{13}C NMR spectra of **6b** (101 MHz, DMSO-d_6)



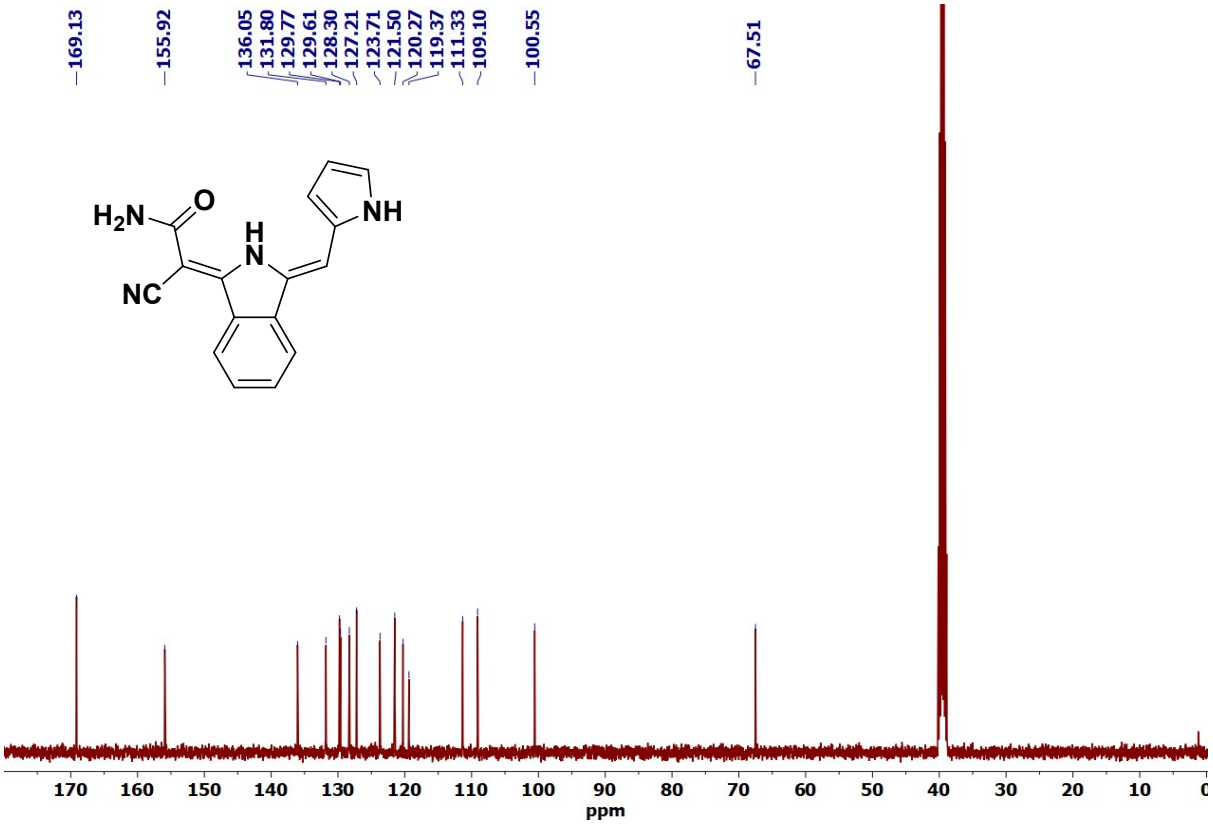
NOE spectra of **6b** (400 MHz, DMSO-d_6)



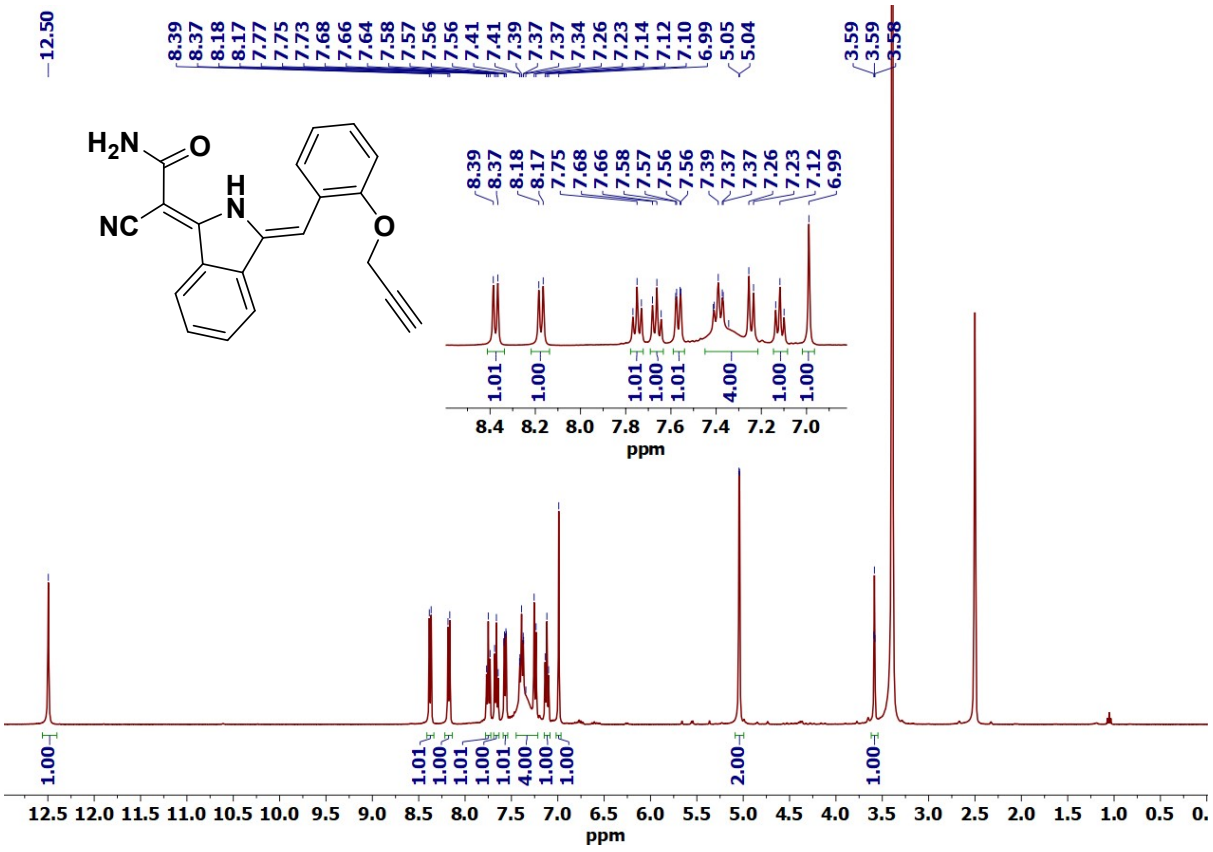
^{1}H NMR spectra of **6c** (400 MHz, DMSO-d_6)



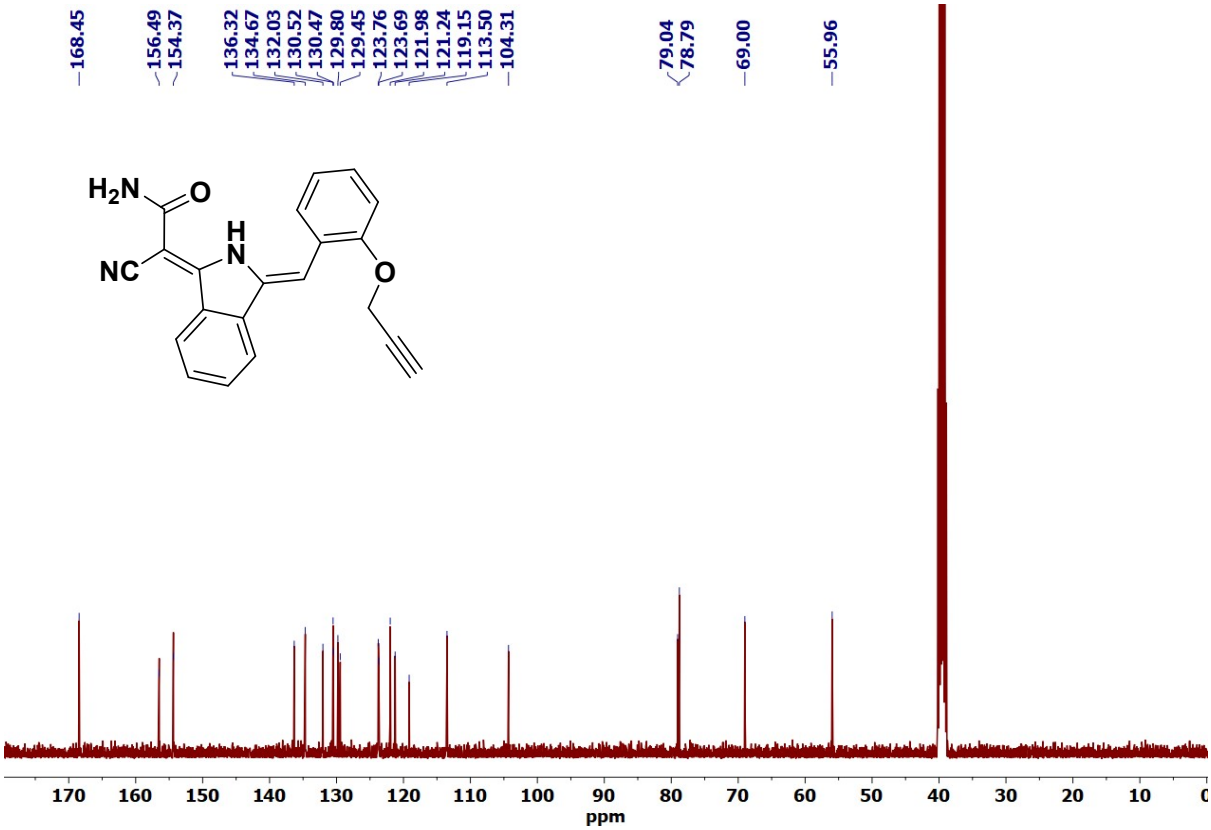
^{13}C NMR spectra of **6c**(101 MHz, DMSO-d_6)



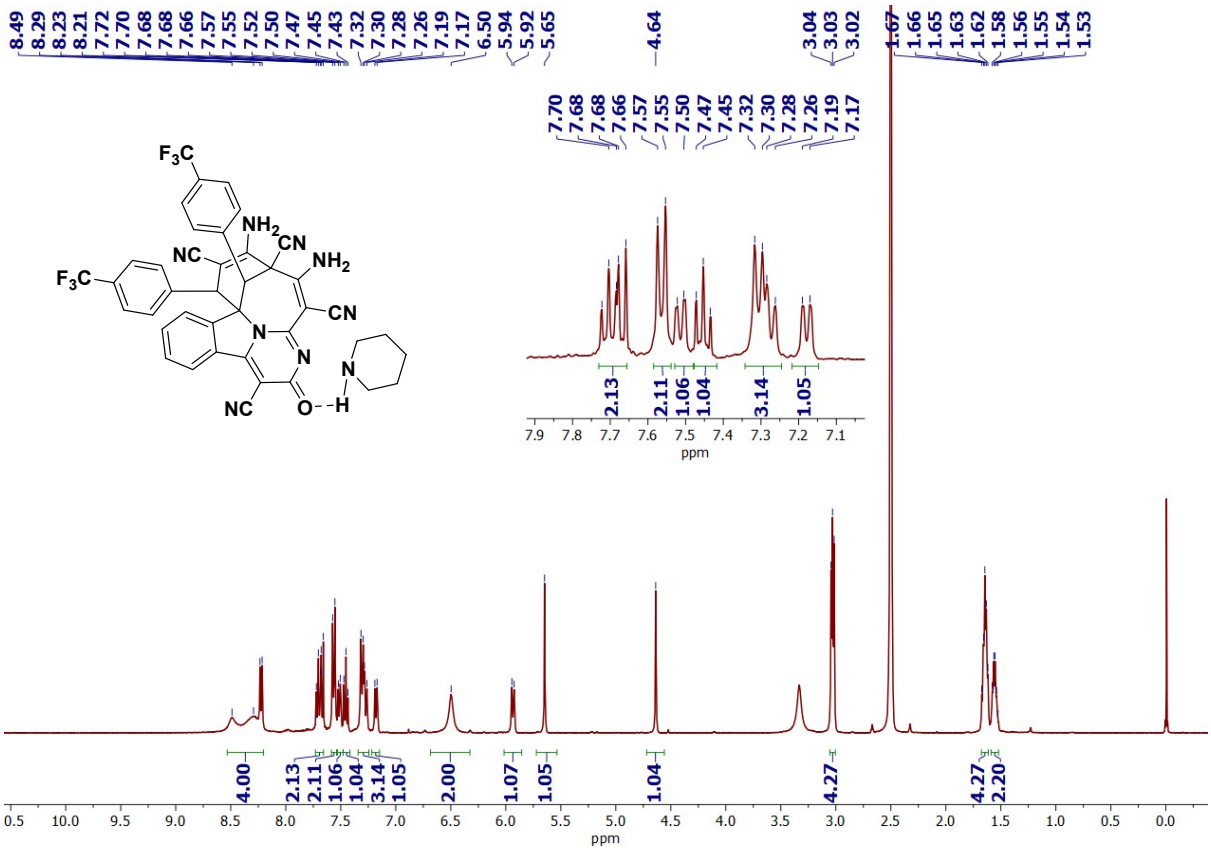
^{1}H NMR spectra of **6d** (400 MHz, DMSO-d_6)



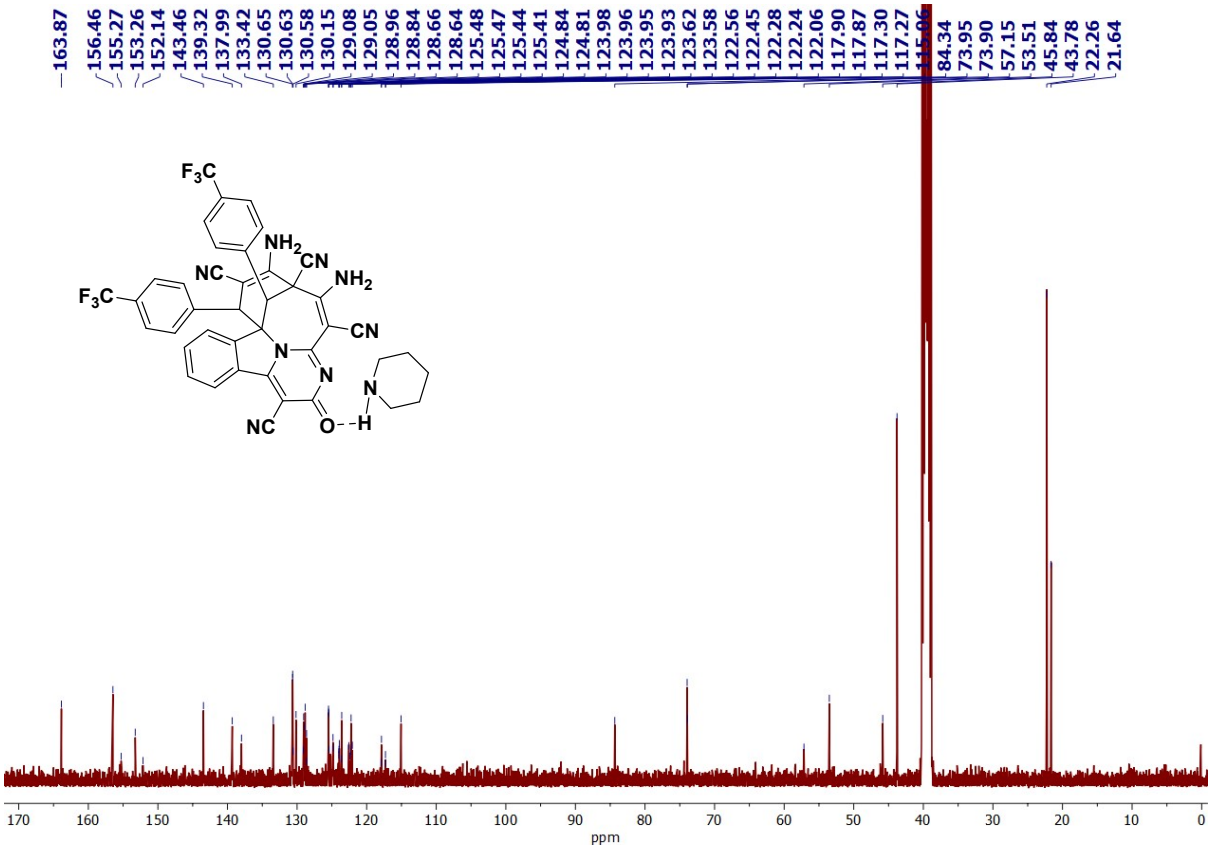
^{13}C NMR spectra of **6d** (101 MHz, DMSO-d_6)



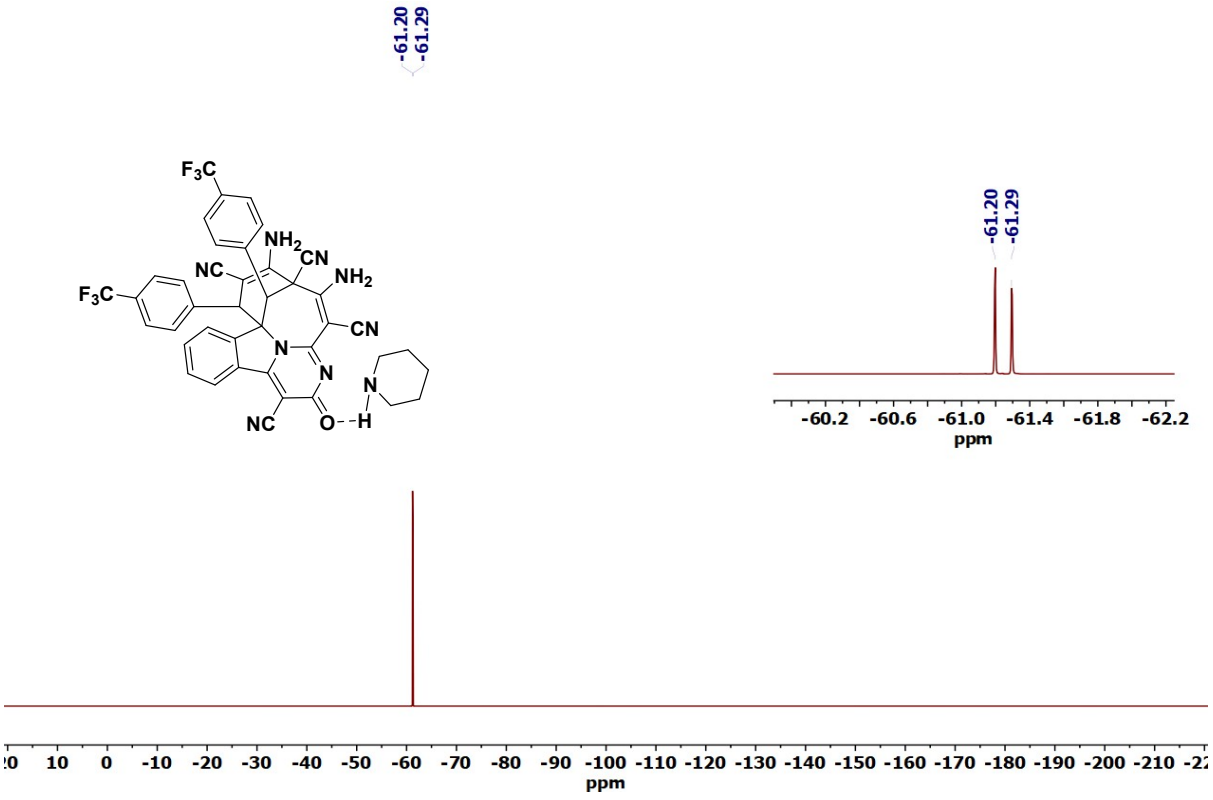
^{1}H NMR spectra of **7a** (400 MHz, DMSO-d_6)



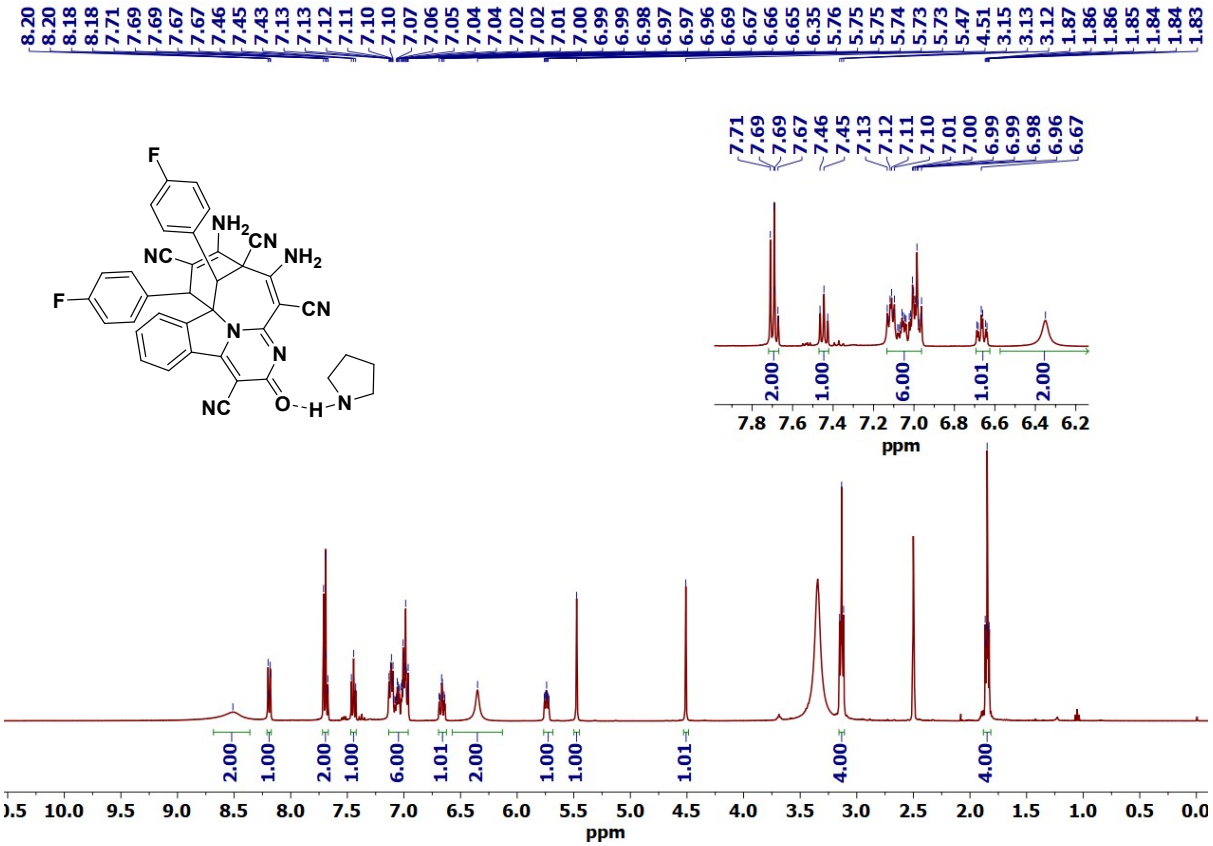
^{13}C NMR spectra of **7a** (101 MHz, DMSO-d_6)



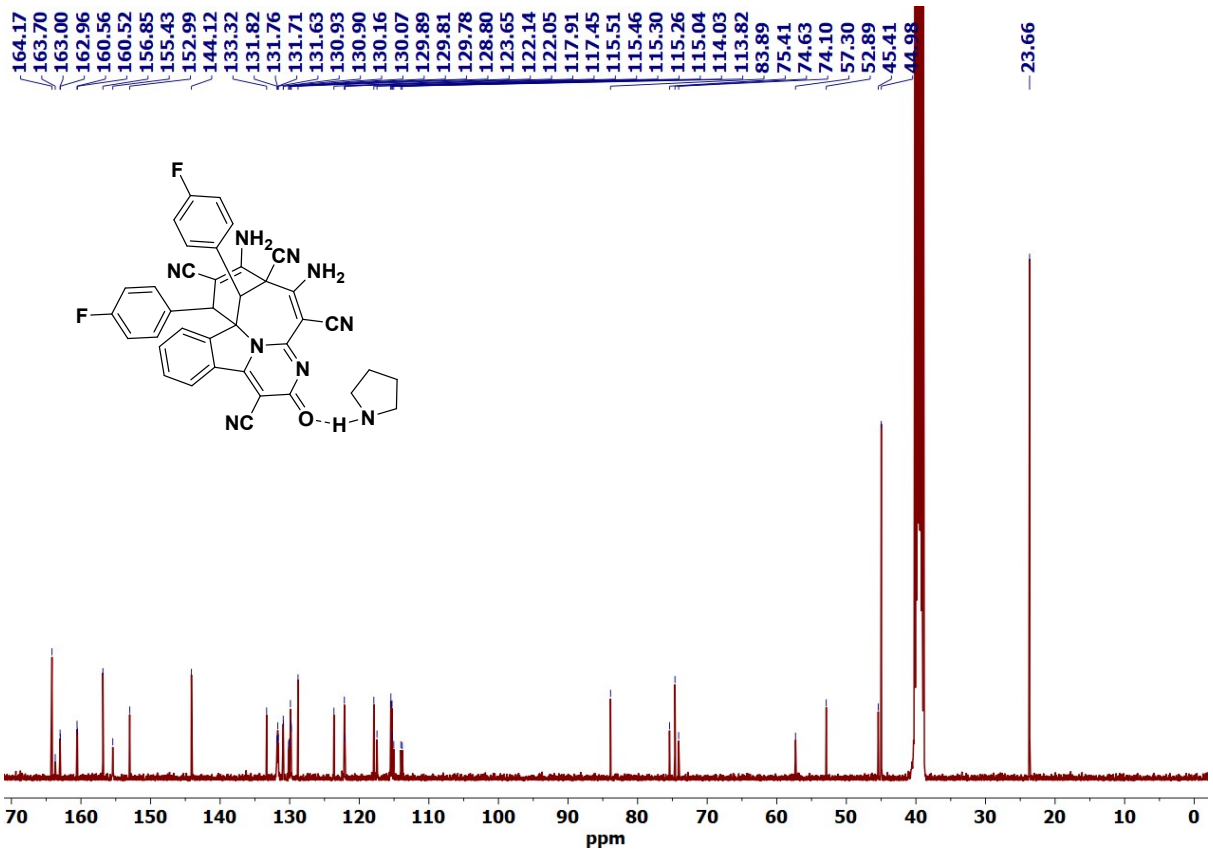
^{19}F NMR spectra of **7a** (376 MHz, DMSO-d_6)



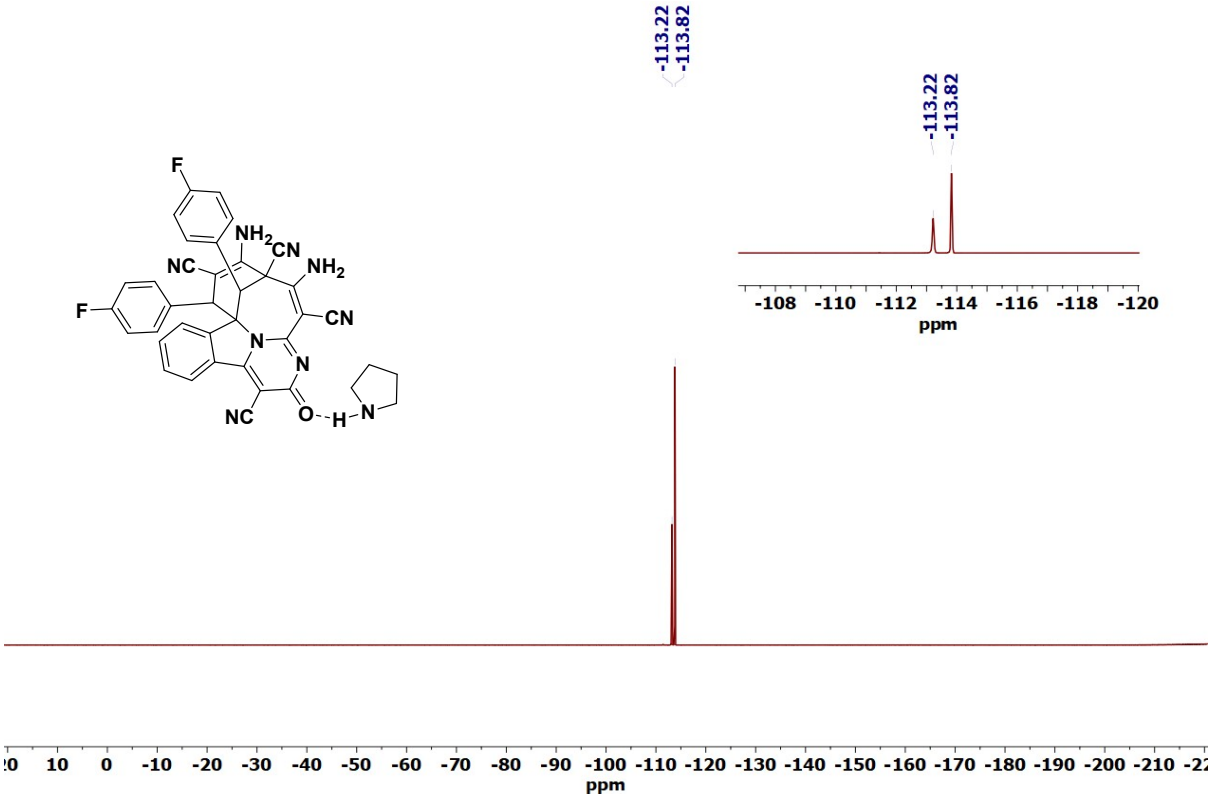
^{1}H NMR spectra of **7b** (400 MHz, DMSO-d_6)



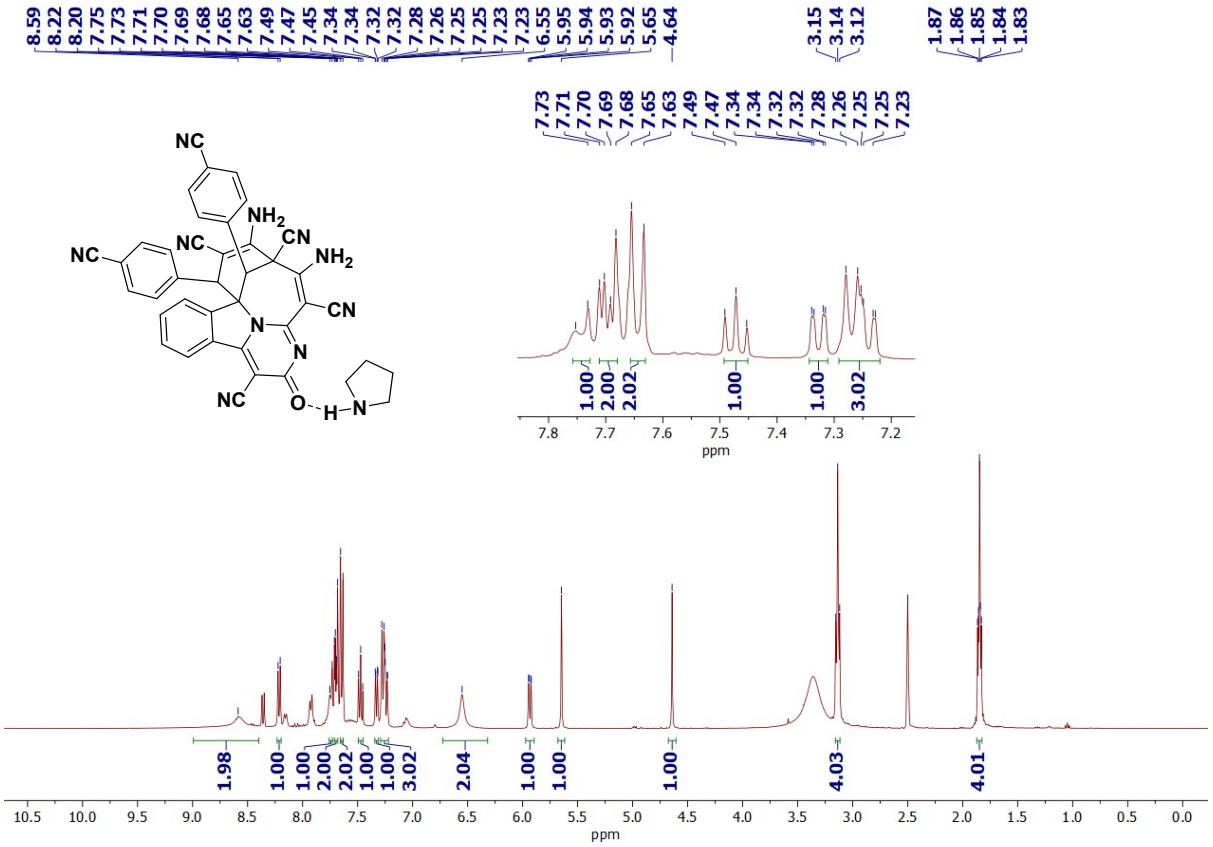
^{13}C NMR spectra of **7b** (101 MHz, DMSO-d_6)



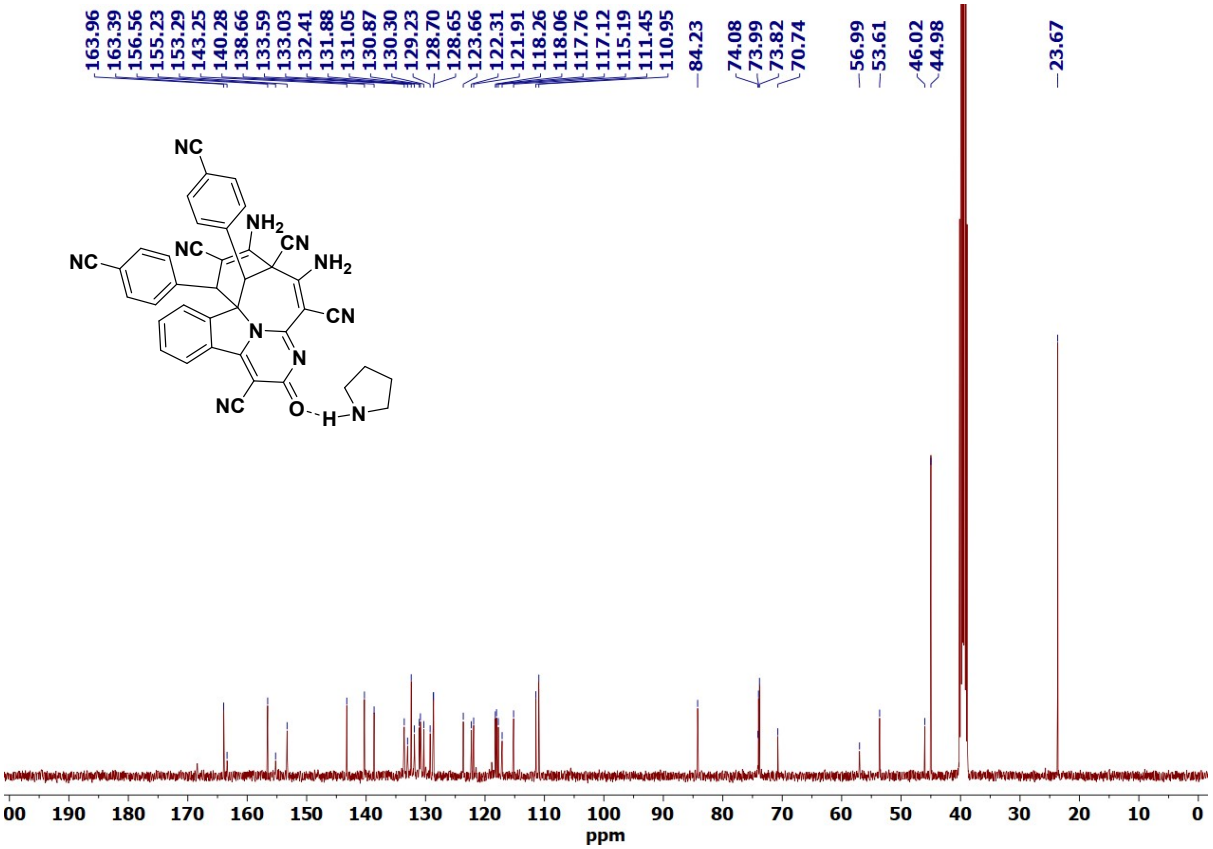
^{19}F NMR spectra of **7b** (376 MHz, DMSO-d_6)



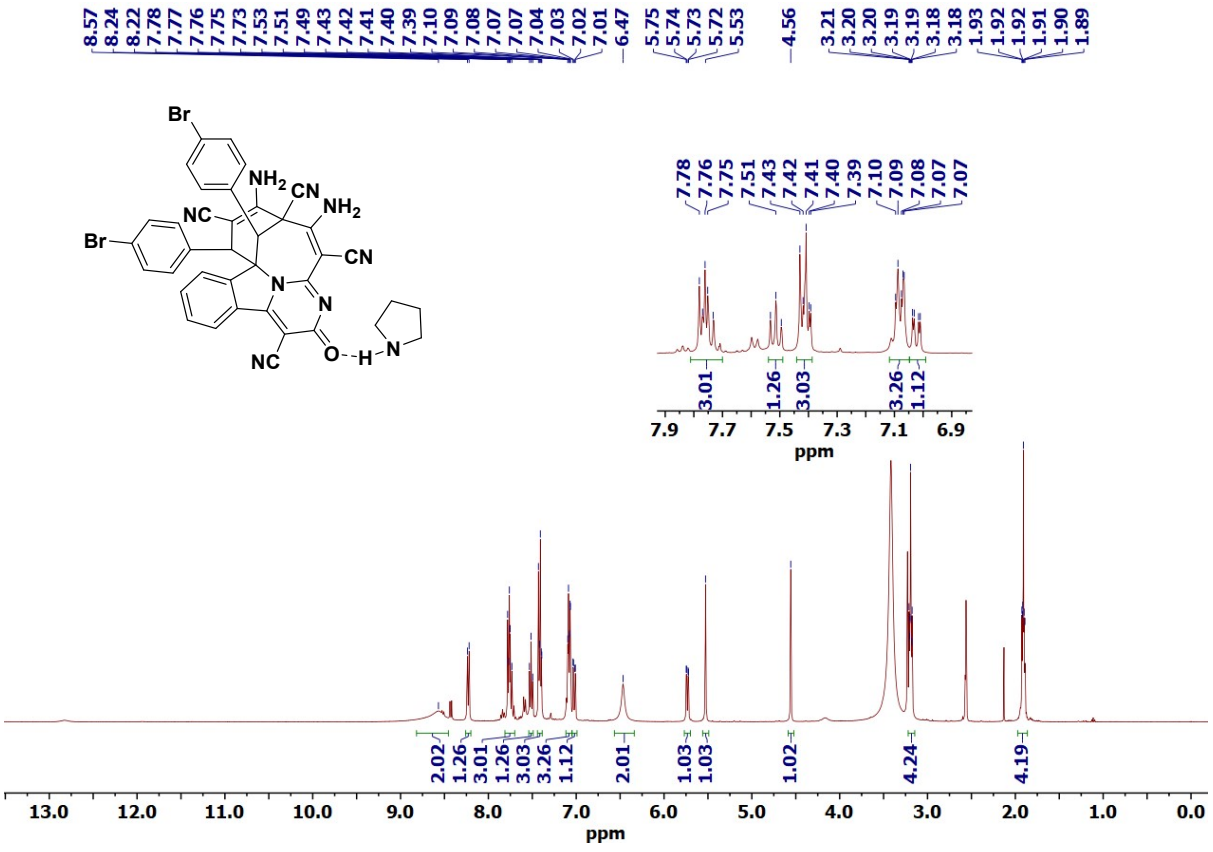
^{1}H NMR spectra of **7c** (400 MHz, DMSO-d_6)



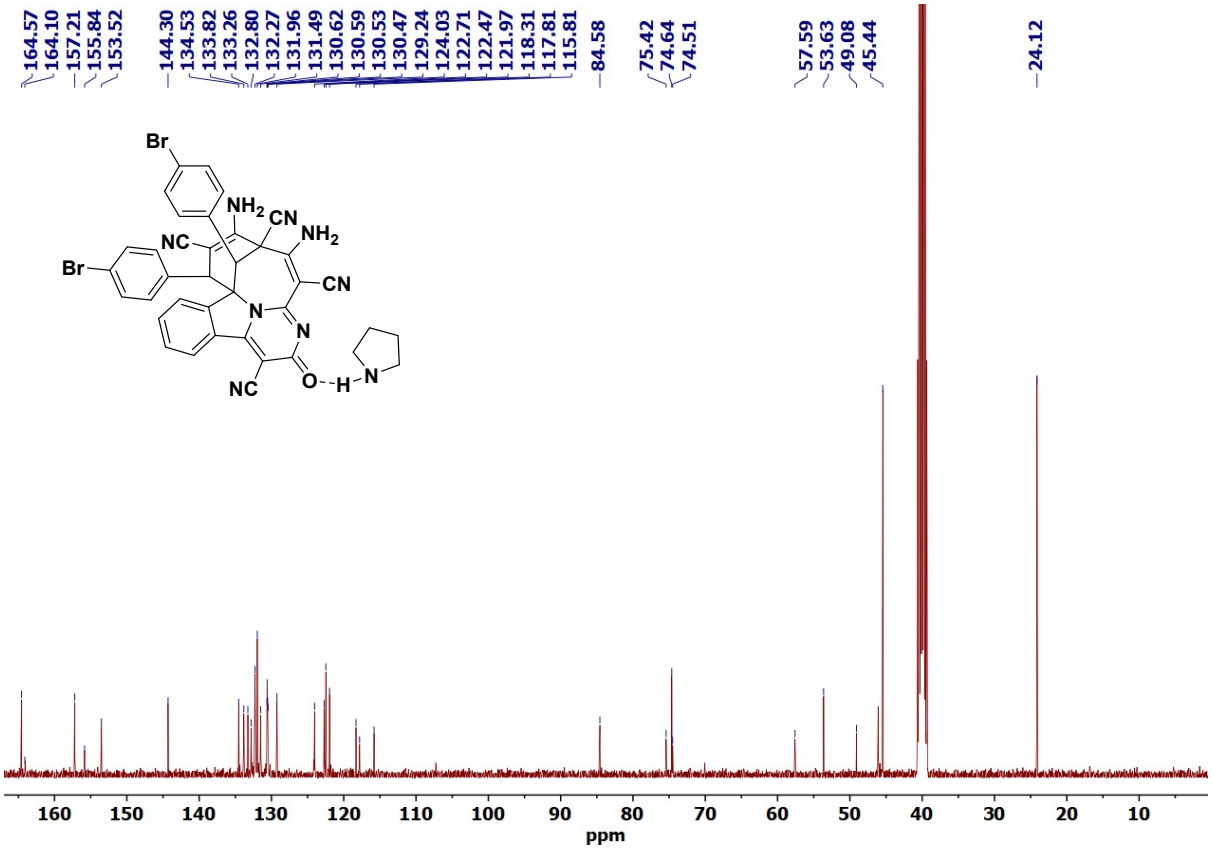
^{13}C NMR spectra of **7c** (101 MHz, DMSO-d_6)



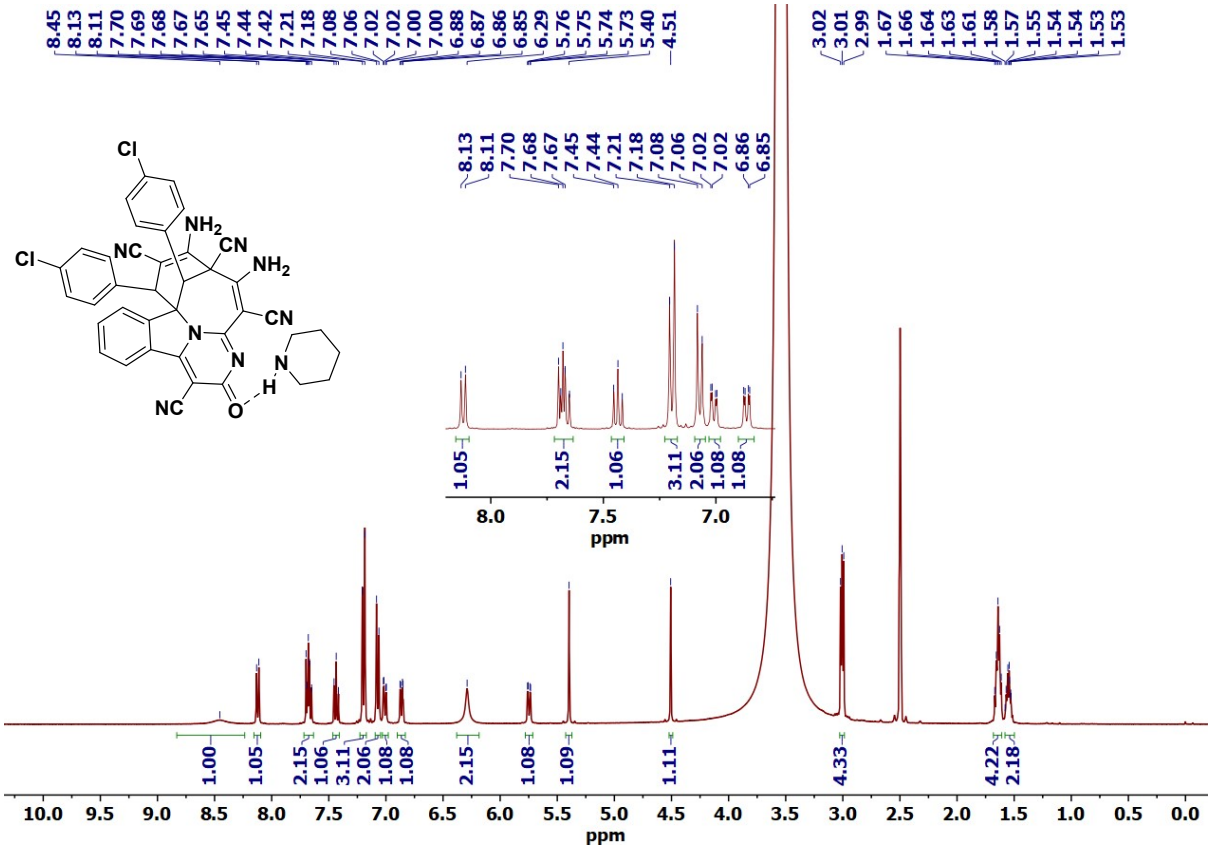
^{1}H NMR spectra of **7d** (400 MHz, DMSO-d_6)



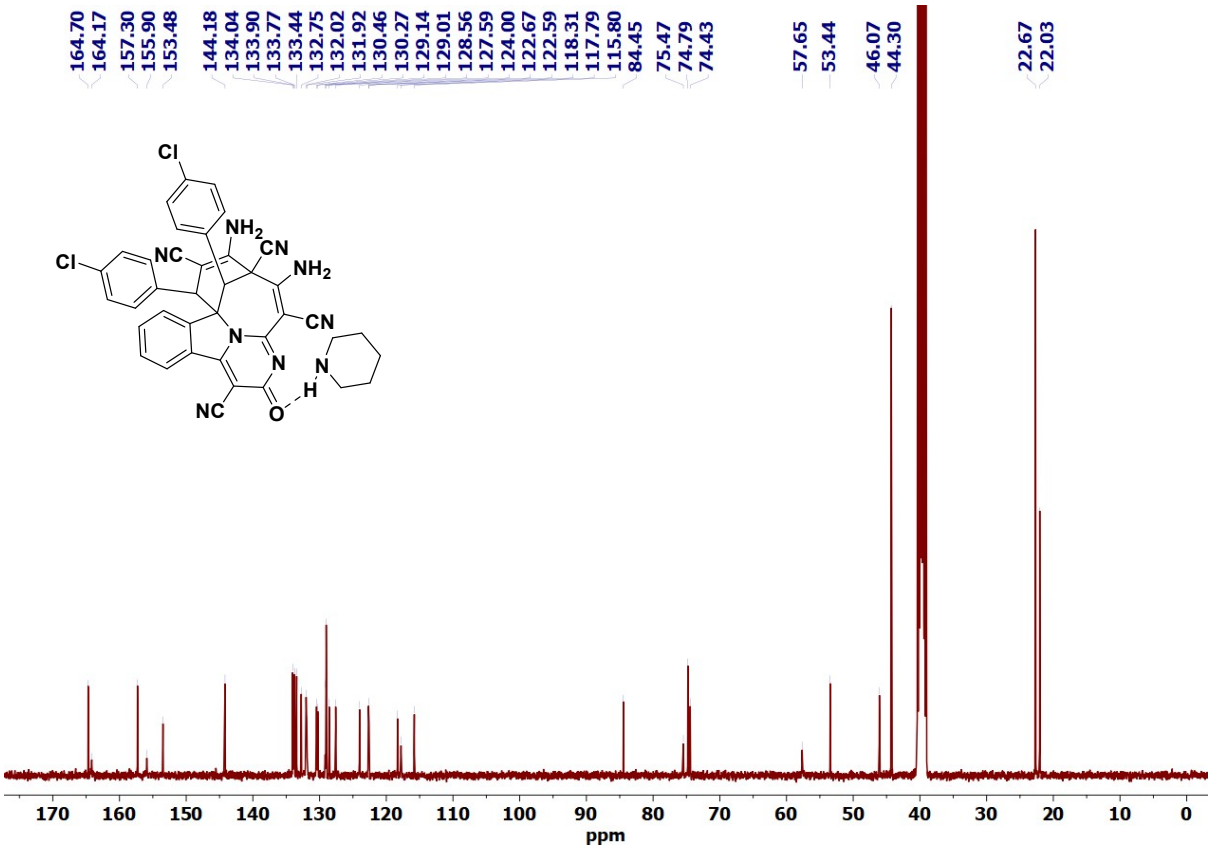
^{13}C NMR spectra of **7d** (101 MHz, DMSO-d_6)



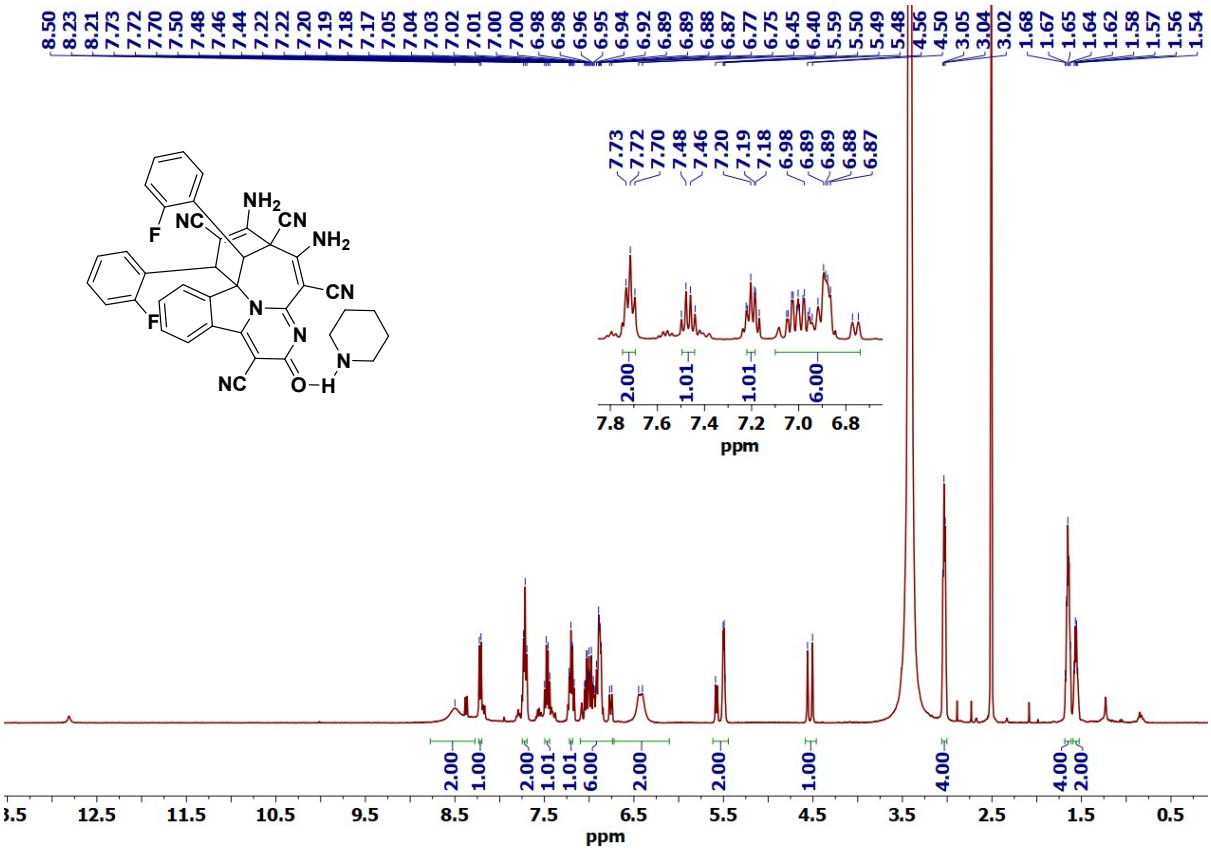
^{1}H NMR spectra of **7e** (400 MHz, DMSO-d_6)



^{13}C NMR spectra of **7e** (101 MHz, DMSO-d_6)



^{1}H NMR spectra of **7f** (400 MHz, DMSO-d_6)



^{19}F NMR spectra of **7f** (376 MHz MHz, DMSO-d_6)