Supporting Information

One-pot, efficient synthesis of fused quinoline analogues *via* room temperature C(sp^3)-N cleavage/aromatization reaction

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Supporting information

Total number of pages: 51

Table of Context

Materials:

All reactions were performed in an oven-dried glassware in the presence of air atmosphere. Maleic anhydride and acetic anhydride were purchased from Qualigens; sodium acetate, were purchased from Merck. Substituted anilines were purchased from Merck, HiMedia and Aldrich. *N*,*N*-Dimethylaniline, *N*-substituted anilines and *t*-butyl hydroperoxide (TBHP) were purchased from Otto and Aldrich respectively. Silver nitrate, potassium persulfate, DMSO were purchased from Fisher Scientific, Merck, Loba Chemie. Substituted aldehydes, dimedone, ammonium acetate and zinc chloride were purchased from Aldrich, Merck, Avra, Aldrich , Merck , Avra Silica gel 100-200 mesh size (Code 95178) and 200-400 mesh size (Code 5699D00500) were purchased from SRL. Sodium chloride (Assay = 99.5%) and anhydrous sodium sulphate were purchased from SRL Chemical. The final products were purified by column chromatography. Melting points were analyzed using melting point apparatus and the melting points are uncorrected. ^{1}H-NMR was recorded in Jeol NMR 600 MHz and ^{13}C-NMR in 150 MHz spectrometer using TMS as an internal standard and $CDCl_3$ and/or DMSO-d_6 as solvent. High resolution mass spectrometry (HRMS) was recorded on a WATERS – XEVO G2-XS-QToF High-Resolution Mass Spectrometer. TBHP is an organic peroxide and it is toxic in nature. Hence, all the necessary precautions need to be taken while handling TBHP.

Methods:

Representative procedure for the synthesis of pyrrolo-fused quinolines (4a):

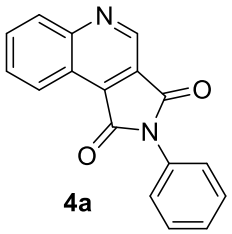
A round bottom flask was charged with **3a** (29.2 mg, 0.1 mmol) or **5a** (acridine derivative (34.9 mg, 0.1 mmol)) and DMSO (2 mL) then $AgNO_3$ (5.1 mg, 0.03 mmol), $K_2S_2O_8$ (40.6 mg, 0.15 mmol) were added. The reaction mixture was stirred at room temperature in air atmosphere for 8h. The progress of the reaction was monitored by TLC. After the completion of the reaction, the reaction mixture was quenched with brine solution (10 mL) and water (5 mL) is added into the reaction mixture. The reaction mixture was extracted with ethyl acetate

(2 × 10 mL). The combined organic layer was dried over anhydrous sodium sulphate, filtrated and the filtrate was evaporated under reduced pressure to afford crude residue. The crude residue was purified by column chromatography using silica gel (100-200 mesh) with hexane and ethyl acetate as eluent (9:1) to afford pure product.

To evaluate the synthetic utility of the given method, we have carried out **4a** synthesis in gram scale. The product **4a** was obtained in 78% yield.

The synthesis of *N*-arylmaleimides/*N*-alkylmaleimides, 3,3,6,6-tetramethyl-9-phenyldecahydroacridine-1,8(2H,5H)-dione and tetrahydroisoquinoline substrates were performed by closely following the previously reported procedure.[1-3]

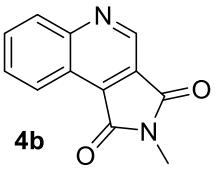
2-phenyl-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione (4a)



4a

Pale yellow solid, Yield: 24 mg (88%),mp: 135-137 ºC (136–137 ºC). [4] ^{1}H-NMR (600 MHz, $CDCl_3$) δ 9.47 (s, 1H), 8.92 -8.91 (m, 1H), 8.31 (d, *J* = 8.4 Hz, 1H), 7.99-7.96 (m, 1H), 7.86-7.83 (m, 1H), 7.57–7.54 (m, 2H) 7.50–7.45 (m, 3H). ^{13}C-NMR (150 MHz, $CDCl_3$) δ 167.3, 166.7, 152.2, 143.8, 135.2, 132.9, 131.2, 130.4, 130.3, 129.3, 128.5, 126.6, 125.1, 123.4, 121.6. HRMS (m/z): $[M + H]^+$ calc. for $C_{17}H_{11}N_2O_2$: 275.0815; found: 275.0831.

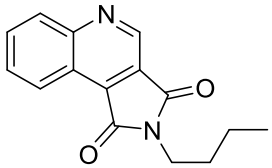
2-methyl-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione (4b)



4b

Yellow solid, Yield: 19.62 mg (92%),mp: 74-76 ºC (77–78 ºC). [4] ^{1}H-NMR (600 MHz, $CDCl_3$) δ 9.37 (s, 1H), 8.86-8.84 (m, 1H), 8.25 (d, *J* = 8.4 Hz, 1H), 7.95–7.92 (m, 1H), 7.82-7.79 (m, 1H), 3.26 (s, 3H). ^{13}C-NMR (150 MHz, $CDCl_3$) δ 168.5, 167.9, 152.1, 143.4, 135.8, 132.7, 130.3, 130.1, 125.1, 124.0, 121.5, 24.2. HRMS (m/z): $[M + H]^+$ calc. for $C_{12}H_9N_2O_2$: 213.0659; found: 213.0666.

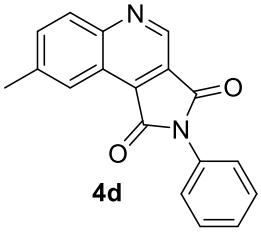
2-butyl-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione (4c)



4c

White solid, Yield: 23 mg (90%),mp: 62-64 ºC ^{1}H-NMR (600 MHz, $CDCl_3$) δ 9.36 (s, 1H), 8.86-8.84 (m, 1H), 8.25 (d, *J* = 8.4 Hz, 1H), 7.94-7.92 (m, 1H), 7.81-7.79 (m, 1H), 3.76 (t, *J* = 7.2 Hz, 2H), 1.74-1.69 (m, 2H), 1.43-1.39 (m, 2H), 0.97 (t, *J* = 7.8 Hz, 3H). ^{13}C-NMR (150 MHz, $CDCl_3$) δ 168.5, 167.9, 152.1, 143.5, 135.7, 132.7, 130.3, 130.1, 125.1, 123.9, 121.6, 38.1, 30.7, 20.2, 13.7. HRMS (m/z): $[M + H]^+$ calc. for $C_{15}H_{15}N_2O_2$: 255.1128; found: 255.1122

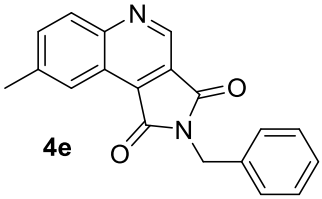
8-methyl-2-phenyl-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione(4d)



4d

Yellow solid, Yield: 23.57 mg (82%),mp: 160-162 ºC (160–161 ºC). [4] ^{1}H-NMR (600 MHz, $CDCl_3$) δ 9.39 (s, 1H), 8.67 (brs, 1H), 8.18 (d, *J* = 9 Hz, 1H), 7.80 (dd, J = 8.4 Hz, J = 1.8 Hz, 1H), 7.57–7.54 (m, 2H), 7.49–7.44 (m, 3H), 2.65 (s, 3H). ^{13}C-NMR (150 MHz, $CDCl_3$) δ 167.5, 166.8, 151.1, 142.8, 141.1, 135.3, 134.2, 131.3, 130.0, 129.3, 128.4, 126.6, 123.7, 123.4, 121.7, 22.0. HRMS (m/z): $[M + H]^+$ calc. for $C_{18}H_{13}N_2O_2$: 289.0972; found: 289.0978.

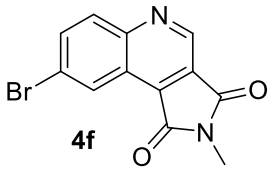
2-benzyl-8-methyl-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione(4e)



4e

Pale yellow solid, Yield: 22.98 mg (76%),mp: 140-142 ºC ^{1}H-NMR (600 MHz, $CDCl_3$) δ 9.28 (s 1H), 8.59 (s 1H), 8.11 (d, *J* = 8.4 Hz, 1H), 7.74 (dd, *J* = 9.0 Hz, 2.4 Hz 1H), 7.48–7.46 (m, 2H), 7.35–7.33 (m, 2H), 7.30–7.27 (m, 1H), 4.90 (s, 2H) 2.62 (s, 3H). ^{13}C-NMR (150 MHz, $CDCl_3$) δ 168.3, 167.6, 150.9, 142.4, 140.8, 136.0, 135.1, 134.6 129.8, 128.8, 128.7, 128.1, 123.7, 123.5, 121.6, 41.8, 22.0. HRMS (m/z): $[M + H]^+$ calc. for $C_{19}H_{15}N_2O_2$: 303.1128; found: 303.1137

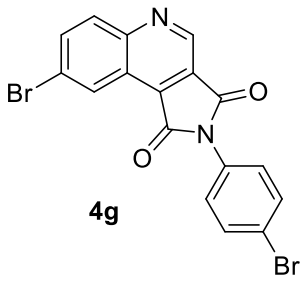
8-bromo-2-methyl-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione (4f)



4f

Pale yellow solid, Yield: 23.87 mg (82%),mp: 212-214 ºC (209–210 ºC). [4] ^{1}H-NMR (600 MHz, $CDCl_3$) δ 9.36 (s, 1H), 9.01 (d, *J* = 2.4 Hz, 1H), 8.11 (d, *J* = 9 Hz, 1H), 7.99 (dd, *J*= 9 Hz, *J*= 1.8 Hz, 1H), 3.26 (s, 3H). ^{13}C-NMR (150 MHz, $CDCl_3$) δ 167.9, 167.4, 150.5, 143.6, 136.2, 134.7, 131.7, 127.1, 125.0, 124.5, 122.3, 24.3. HRMS (m/z): $[M + H]^+$ calc. for $C_{12}H_8BrN_2O_2$: 290.9764; found: 290.9760.

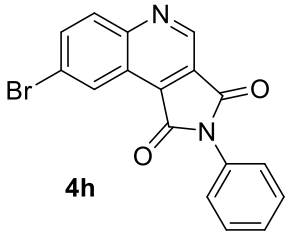
8-bromo-2-(4-bromophenyl)-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione (4g)



4g

Pale yellow solid, Yield: 33 mg (77%), mp: 216-218 ºC ^{1}H-NMR (600 MHz, $CDCl_3$) δ 9.46 (s, 1H), 9.06 (d, *J* = 1.8 Hz, 1H), 8.16 (d, *J* = 9.0 Hz, 1H), 8.04 (dd, *J* = 9.0 Hz, 2.4 Hz, 1H),7.70-7.67 (m, 2H), 7.40-7.38 (m, 2H). ^{13}C-NMR (150 MHz, $CDCl_3$) δ 166.4, 165.9, 150.8, 144.0, 136.6, 134.0, 132.5, 131.8, 130.1, 127.9, 127.2, 125.4, 123.8, 122.4, 122.3. HRMS (m/z): $[M + H]^+$ calc. for $C_{17}H_9Br_2N_2O_2$: 430.9025; found: 430.9027

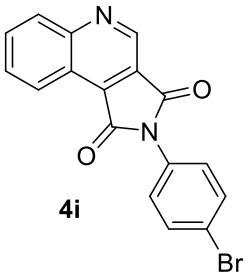
8-bromo-2-phenyl-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione(4h)



4h

Pale yellow solid, Yield: 28 mg (80%), mp: 210-112ºC (209–210ºC). [4] ^{1}H-NMR (600 MHz, $CDCl_3$) δ 9.46 (s, 1H), 9.07 (d, *J* = 1.8 Hz, 1H), 8.16 (d, *J* = 9.0 Hz, 1H), 8.02 (dd, *J* = 9.0 Hz, 2.4 Hz, 1H), 7.57 -7.54 (m, 2H), 7.48 -7.47 (m, 3H). ^{13}C-NMR (150 MHz, $CDCl_3$) δ 166.7, 166.2, 150.6, 144.0, 136.4, 134.1, 131.7, 131.0, 129.3, 128.5, 127.2, 126.5, 125.2, 124.0, 122.3. HRMS (m/z): $[M + H]^+$ calc. for $C_{17}H_{10}BrN_2O_2$: 352.9920; found: 352.9926.

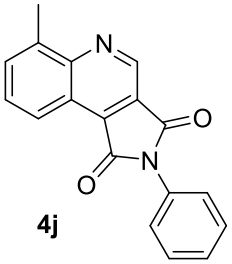
2-(4-bromophenyl)-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione(4i)



4i

Pale yellow solid, Yield: 28 mg (80%); mp: 142-144 °C (136–137 °C). [4] ^{1}H-NMR (600 MHz, $CDCl_3$) δ 9.46 (s, 1H), 8.90 -8.89 (m, 1H), 8.31 (d, *J* = 8.4 Hz, 1H), 8.00 -7.97 (m, 1H), 7.86 -7.83 (m, 1H), 7.69-7.67 (m, 2H), 7.41 -7.39 (m, 2H). ^{13}C-NMR (150 MHz, $CDCl_3$) δ 166.9, 166.3, 152.2, 143.7, 135.0, 133.0, 132.5, 130.4, 130.2, 127.9, 127.8, 125.0, 123.2, 122.2, 121.5. HRMS (m/z): $[M + H]^+$ calc. for $C_{17}H_{10}BrN_2O_2$: 352.9920; found: 352.9919.

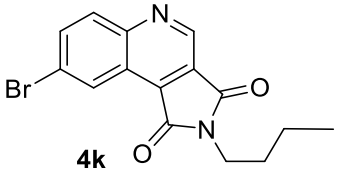
6-methyl-2-phenyl-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione(4j)



4j

White solid, Yield: 28.3 mg (79%), mp: 214-216°C. ^{1}H-NMR (600 MHz, $CDCl_3$) δ 9.43 (s, 1H), 8.74 (d, *J* = 8.4 Hz, 1H), 7.79 (d, *J* = 7.2 Hz, 1H), 7.71-7.68 (m, 1H), 7.56-7.53 (m, 2H), 7.49-7.44 (m, 3H) 2.87 (s, 3H).^{13}C-NMR (150 MHz, $CDCl_3$) δ 167.4, 166.8, 151.3, 142.3, 138.5, 135.1, 132.9, 131.3, 130.1, 129.2, 128.4, 126.6, 123.0, 122.8, 121.6, 18.6. HRMS (m/z): $[M + H]^+$ calc. for $C_{18}H_{13}N_2O_2$: 289.0972; found: 289.0967

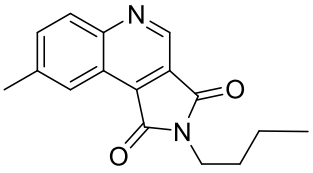
8-bromo-2-butyl-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione (4k)



4k

White solid, Yield: 27.61 mg (83%),mp: 100-102 °C ^{1}H-NMR (600 MHz, $CDCl_3$) δ 9.35 (s, 1H), 9.01 (d, *J* = 1.8 Hz, 1H), 8.11 (d, *J* = 9 Hz, 1H), 7.98 (dd, *J* = 9 Hz, *J* = 1.8 Hz 1H), 3.76 (t, *J* = 7.2 Hz, 2H), 1.73-1.68 (m, 2H), 1.43-1.37 (m, 2H), 0.97 (t, *J* = 7.2 Hz, 3H). ^{13}C-NMR (150 MHz, $CDCl_3$) δ 167.9, 167.4, 150.5, 143.7, 136.2, 134.6, 131.7, 127.1, 124.9, 124.4, 122.3, 38.2, 30.6, 20.1, 13.6. HRMS (m/z): $[M + H]^+$ calc. for $C_{15}H_{14}BrN_2O_2$: 333.0233; found: 333.0242.

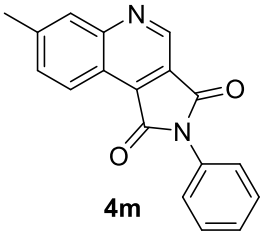
2-butyl-8-methyl-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione (4l)



4l

White solid, Yield: 22.39 mg (83%),mp: 78-80 °C ^{1}H-NMR (600 MHz, $CDCl_3$) δ 9.28 (s, 1H), 8.61 (s, 1H), 8.13 (d, *J* = 9 Hz, 1H), 7.75 (d, *J* = 8.4 Hz, 1H), 3.75 (t, *J* = 7.2 Hz, 2H), 2.63 (s, 3H), 1.73-1.69 (m, 2H), 1.44-1.37 (m, 2H), 0.97(s, *J* = 7.2 Hz, 3H). ^{13}C-NMR (150 MHz, $CDCl_3$) δ δ 168.7, 168.0, 150.8, 142.4, 140.7, 135.0, 134.7, 129.8, 123.8, 123.5, 121.6, 38.0, 30.7, 22.0, 20.1, 13.6. HRMS (m/z): $[M + H]^+$ calc. for $C_{16}H_{17}N_2O_2$: 269.1285; found: 269.1306

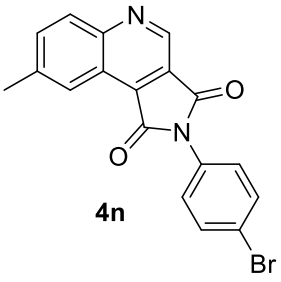
7-methyl-2-phenyl-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione (4m)



4m

Yellow solid, Yield: 21.97 mg (76%),mp: 145-147 °C (149-150 °C). [4] ^{1}H-NMR (600 MHz, $CDCl_3$) δ 9.42 (s, 1H), 8.79 (d, *J* = 9 Hz, 1H), 8.08 (s, 1H), 7.67 (dd, J = 8.4 Hz, J = 1.2 Hz, 1H), 7.57–7.54 (m, 2H), 7.49–7.44 (m, 3H) 2.67 (s, 3H). ^{13}C-NMR (150 MHz, $CDCl_3$) 167.4, 166.8, 152.5, 144.1, 143.8, 135.1, 132.7, 131.3, 129.3, 129.28, 128.4, 126.6, 124.6, 122.6, 119.6, 22.4. HRMS (m/z): $[M + H]^+$ calc. for $C_{18}H_{13}N_2O_2$:289.0972; found: 289.0978.

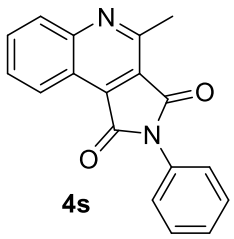
2-(4-bromophenyl)-8-methyl-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione (4n)



4n

Pale yellow solid, Yield: 29.29 mg (80%), mp: 196-198 °C (193–194 °C). [4] ^{1}H-NMR (600 MHz, $CDCl_3$) δ 9.39 (s, 1H), 8.66 (s, 1H), 8.19 (d, *J* = 9.0 Hz, 1H), 7.80 (dd, *J* = 9.0 Hz, 1.8 Hz, 1H), 7.69 -7.66 (m, 2H), 7.41 -7.39 (m, 2H), 2.66 (s, 3H). ^{13}C-NMR (150 MHz, $CDCl_3$) δ 166.8, 166.3, 152.2, 143.7, 135.0, 132.9, 132.5, 130.4, 130.2, 127.9, 127.8, 125.0, 123.2, 122.2, 121.4. HRMS (m/z): $[M + H]^+$ calc. for $C_{18}H_{12}BrN_2O_2$:367.0077; found: 367.0079.

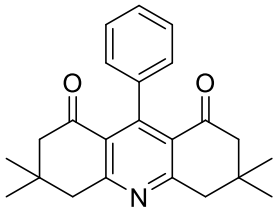
4-methyl-2-phenyl-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione (4s)



4s

Pale yellow solid, Yield: 22 mg (77%), mp: 194-196 °C ^{1}H-NMR (600 MHz, $CDCl_3$) δ 8.89 - 8.87 (m, 1H), 8.18 (d, *J* = 8.4 Hz, 1H), 7.94-7.91 (m, 1H), 7.75-7.74 (m, 1H), 7.56–7.54 (m, 2H) 7.48–7.45 (m, 3H), 3.11 (s, 3H). ^{13}C NMR (150 MHz, $CDCl_3$) δ 167.4, 166.8, 151.3, 142.3, 138.3, 135.1, 132.9, 131.3, 130.1, 129.2, 128.4, 126.6, 123.0, 122.8, 121.6, 18.6. HRMS (m/z): $[M + H]^+$ calc. for $C_{18}H_{13}N_2O_2$: 289.0972; found: 289.0970.

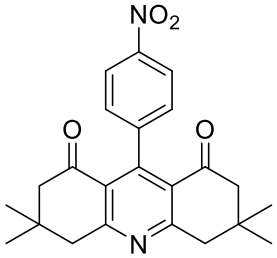
3,3,6,6-tetramethyl-9-phenyl-3,4,6,7-tetrahydroacridine-1,8(2H,5H)-dione (6a)



6a

White solid, Yield: 29.92 mg (86%),mp: 198-200 °C ^{1}H-NMR (600 MHz, $CDCl_3$) δ 7.39-7.38 (m, 3H), 7.02-7.00 (m, 2H), 3.11 (s, 4H), 2.46 (s, 4H), 1.12 (s, 12H). ^{13}C-NMR (150 MHz, $CDCl_3$) δ 197.1, 165.9, 152.0, 138.5, 127.7, 127.0, 126.4, 125.5, 54.0, 48.0, 32.4, 28.3. HRMS (m/z): $[M + H]^+$ calc. for $C_{23}H_{26}NO_2$: 348.1958; found: 348.1992.

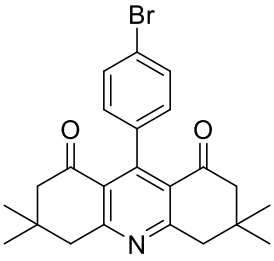
3,3,6,6-tetramethyl-9-(4-nitrophenyl)-3,4,6,7-tetrahydroacridine-1,8(2H,5H)-dione (6b)



6b

White solid, Yield: 31.66 mg (81%),mp: 238-239 °C ^{1}H-NMR (600 MHz, $CDCl_3$) δ 8.27-8.24 (m, 2H), 7.17-7.15 (m, 2H), 3.14 (s, 4H), 2.47 (s, 4H), 1.13 (s, 12H). ^{13}C-NMR (150 MHz, $CDCl_3$) δ 197.0, 166.6, 149.6, 146.8, 146.7, 127.4, 124.7, 123.2, 53.7, 47.9, 32.4, 28.2. HRMS (m/z): $[M + 2H]^+$ calc. for $C_{23}H_{25}N_2O_4$: 393.1809; found: 393.1842.

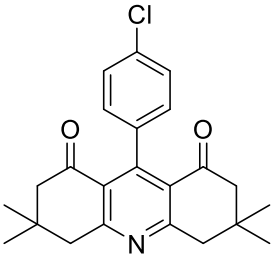
9-(4-bromophenyl)-3,3,6,6-tetramethyl-3,4,6,7-tetrahydroacridine-1,8(2H,5H)-dione (6c)



6c

White solid, Yield: 34.80 mg (82%),mp: 164-166 ^{o}C ^{1}H-NMR (600 MHz, $CDCl_3$) δ 7.51-7.49 (m, 2H), 6.90-6.87 (m, 2H), 3.11 (s, 4H), 2.47 (s, 4H), 1.12 (s, 12H). ^{13}C-NMR (150 MHz, $CDCl_3$) δ 197.2, 166.1, 150.8, 137.0, 133.0, 128.0, 127.9, 125.3, 53.9, 48.0, 32.4, 28.2. HRMS (m/z): $[M + H]^+$ calc. for $C_{23}H_{25}BrNO_2$: 426.1063; found: 426.1075

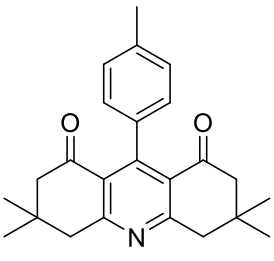
9-(4-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7-tetrahydroacridine-1,8(2H,5H)-dione (6d)



6d

White solid, Yield: 31.95 mg (84%),mp: 158-160 ^{o}C ^{1}H-NMR (600 MHz, $CDCl_3$) δ 7.36-7.34 (m, 2H), 6.95-6.93 (m, 2H), 3.11 (s, 4H), 2.47 (s, 4H), 1.12 (s, 12H). ^{13}C-NMR (150 MHz, $CDCl_3$) δ 197.2, 166.1, 150.8, 137.6, 130.9, 128.2, 125.3, 121.1, 53.9, 48.0, 32.4, 28.2. HRMS (m/z): $[M + H]^+$ calc. for $C_{23}H_{25}ClNO_2$: 382.1568; found: 382.1575.

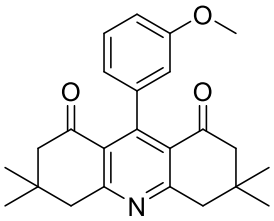
3,3,6,6-tetramethyl-9-(p-tolyl)-3,4,6,7-tetrahydroacridine-1,8(2H,5H)-dione (6e)



6e

White solid, Yield: 31.73 mg (88%),mp: 195-197 ^{o}C ^{1}H-NMR (600 MHz, $CDCl_3$ δ 7.20 (d, *J* = 7.8 Hz, 2H), 6.90 (d, *J* = 8.4 Hz, 2H), 3.10 (s, 4H), 2.47 (s, 4H), 2.41 (s, 3H), 1.11 (s, 12H). ^{13}C-NMR (150 MHz, $CDCl_3$) δ 197.3, 165.8, 152.3, 136.6, 135.4, 128.5, 126.4, 125.7, 54.0, 47.9, 32.4, 28.3, 21.5. HRMS (m/z): $[M + H]^+$ calc. for $C_{24}H_{28}NO_2$: 362.2115; found: 362.2130.

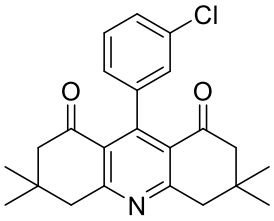
9-(3-methoxyphenyl)-3,3,6,6-tetramethyl-3,4,6,7-tetrahydroacridine-1,8(2H,5H)-dione (6f)



6f

White solid, Yield: 32 mg (86%), mp: 280-282°C. ^{1}H-NMR (600 MHz, $CDCl_3$) δ 7.31 (t, *J* = 7.8 Hz, 1H), 6.91-6.89 (m, 1H), 6.63-6.61 (m, 1H), 6.54-6.53 (m, 1H), 3.79 (s, 3H), 3.10 (s, 4H), 2.46 (s, 4H), 1.12 (s, 6H), 1.11 (s, 6H). ^{13}C-NMR (150 MHz, $CDCl_3$) δ 196.8, 165.8, 159.1, 151.6, 139.8, 128.6, 125.5, 119.2, 113.0, 111.6, 55.0, 53.9, 48.0, 32.3, 28.3, 28.2. HRMS (m/z): $[M + H]^+$ calc. for $C_{24}H_{28}NO_3$: 378.2064; found: 378.2073.

9-(3-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7-tetrahydroacridine-1,8(2H,5H)-dione (6g)

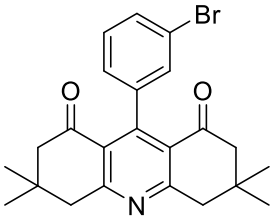


6g

White solid, Yield: 32 mg (85%), mp: 125-1272°C. ^{1}H-NMR (600 MHz, $CDCl_3$) δ 7.35-7.30 (m, 2H), 6.97 (brs, 1H), 6.91(d, *J* = 7.2 Hz, 1H), 3.11 (s, 4H), 2.47 (s, 2H), 2.46 (s, 2H), 1.12 (s, 6H), 1.11 (s, 6H). ^{13}C-NMR (150 MHz, $CDCl_3$) δ 196.8, 166.1, 150.2, 140.3, 133.6, 128.8, 127.0, 126.5, 125.2, 124.8, 53.8, 47.9, 32.3, 28.3, 28.1. HRMS (m/z): $[M + H]^+$ calc. for $C_{23}H_{25}ClNO_2$: 382.1568; found: 382.1612.

9-(3-bromophenyl)-3,3,6,6-tetramethyl-3,4,6,7-tetrahydroacridine-1,8(2H,5H)-dione

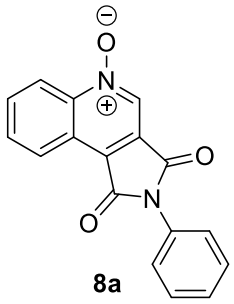
(6h)



6h

White solid, Yield: 37.9 mg (89%), mp: 120-122°C. ^{1}H-NMR (600 MHz, $CDCl_3$) δ 7.50-7.48 (m,1H), 7.27-7.24 (m, 1H), 7.124-7.119 (m, 1H), 6.97-6.95 (m, 1H), 3.11 (s, 4H), 2.48 (s, 2H), 2.47 (s, 2H), 1.13 (s, 6H), 1.12 (s, 6H). ^{13}C-NMR (150 MHz, $CDCl_3$) δ 196.8, 166.1, 150.2, 140.5, 129.9, 129.2, 129.1, 125.3, 125.2, 121.8, 53.8, 47.8, 32.3, 28.3, 28.1. HRMS (m/z): $[M + H]^+$ calc. for $C_{23}H_{25}BrNO_2$: 426.1063; found: 426.1107

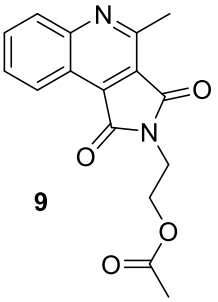
1,3-dioxo-2-phenyl-2,3-dihydro-1H-pyrrolo[3,4-c]quinoline 5-oxide (8a)



8a

White solid, Yield: 26.09 mg (90%),mp: 198-200 ^{o}C ^{1}H-NMR (600 MHz, $CDCl_3$) δ 8.91 (d, *J* = 7.8 Hz, 1H), 8.88 (s, 1H), 8.77 (d, *J* = 9 Hz, 1H), 7.96-7.89 (m, 2H), 7.56-7.54 (m, 2H), 7.47-7.44 (m, 3H). ^{13}C-NMR (150 MHz, $CDCl_3$) δ 165.8, 164.9, 145.1, 132.9, 131.9, 131.0, 129.7, 129.3, 128.6, 127.1, 126.5, 126.1, 124.4, 121.5, 120.6. HRMS (m/z): $[M + H]^+$ calc. for $C_{17}H_{11}N_2O_2$: 291.0764; found: 291.0766.

2-(4-methyl-1,3-dioxo-1,3-dihydro-2H-pyrrolo[3,4-c]quinolin-2-yl)ethyl acetate (9)



9

White solid, Yield: 14 mg (49.0 %), mp: 125-127 ^{o}C; ^{1}H-NMR (600 MHz, $CDCl_3$) δ 8.81 (d, *J* = 7.2 Hz, 1H)), 8.138 (d, *J* = 8.4 Hz, 1H), 7.90-7.88 (m, 1H), 7.73-7.71 (m, 1H), 4.36 (t, *J* = 5.4 Hz, 2H)), 4.015 (t, *J* = 5.4 Hz, 2H)), 3.06 (s, 3H), 2.04 (s, 3H). ^{13}C NMR (150 MHz, $CDCl_3$) δ 170.9, 168.3, 168.1, 155.0, 151.7, 136.0, 132.8, 129.3, 129.0, 124.9, 122.0, 120.7, 61.5, 37.2, 22.1, 20.8. HRMS (m/z): $[M + H]^+$ calc. for $C_{16}H_{15}N_2O_4$: 299.1026; found: 299.1034.

Representative procedure for the one-pot, two-step synthesis of pyrrolidine fused quinolines:

In a round bottom flask, *N*,*N*-dimethylaniline (18.2 mg, 0.15 mmol), *N*-phenylmaleimide (17.3 mg, 0.1 mmol), TBHP (5.0 M-6.0 M in Decane, 0.1 mmol, 18 µL) and copper(II) acetate (1.82 mg, 0.01 mmol) were taken and stirred at neat condition at room temperature for 2 hours. The reaction mixture was then diluted with DMSO (2 mL) and $AgNO_3$ (5.1 mg, 0.03 mmol), $K_2S_2O_8$ (40.6 mg, 0.15 mmol) were added, the progress of the reaction was monitored by TLC. After the completion

of the reaction, the reaction mixture was quenched with brine solution (10 mL) and water (5 mL) is added into the reaction mixture. The aqueous layer was extracted with ethyl acetate (2 × 10 mL). The combined organic extracts was washed with water (1 x 5 mL) and brine solution (2 × 5 mL) and dried over anhydrous Na_2SO_4. The solution was concentrated and eluted through a silica gel column (petroleum ether/ethyl acetate 9:1) to give corresponding compound **4a**.

2-phenyl-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione (4a)
Pale yellow solid, Yield: 22 mg (79%)

2-methyl-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione (4b)
Yellow solid, Yield: 19 mg (88%)

2-butyl-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione (4c)
White solid, Yield: 22 mg (87%)

8-methyl-2-phenyl-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione (4d)
Yellow solid, Yield: 22mg (76%)

2-benzyl-8-methyl-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione (4e)
Pale yellow solid, Yield: 27 mg (72%)

8-bromo-2-methyl-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione (4f)
Pale yellow solid, Yield: 27 mg (75%)

8-bromo-2-(4-bromophenyl)-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione (4g)
Pale yellow solid, Yield: 36 mg (84%)

8-bromo-2-phenyl-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione (4h)
Pale yellow solid, Yield: 26 mg (76%)

2-(4-bromophenyl)-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione (4i)
Pale yellow solid, Yield: 24 mg (68%)

6-methyl-2-phenyl-1H-pyrrolo[3,4-c]quinoline-1,3(2H)-dione (4j)
White solid, Yield: 17.3 mg (62%)

Synthesis of quinolone N-oxide (8a) [5]

Based on literature method, *m*-chloroperoxybenzoic acid (0.15 mmol) was added to a solution of the corresponding **4a** (0.1 mmol) in DCM (5 mL) at 0 °C. After 12 hours of stirring at room temperature, the starting material conversion monitored by TLC, a saturated aqueous

$NaHCO_3$ solution (20 mL) was added to the reaction mixture. DCM (10 mL x 3) was used to extract the reaction mixture. The organic extracts were then dried over anhydrous Na_2SO_4, filtered, and concentrated under low reduced pressure. Using column chromatography and EtOAc/n-hexane on silica gel, the crude product was purified.

References:

(1). (a) K. Eloh, M. Demurtas, M. G. Mura, A. Deplano, V. Onnis, N. Sasanelli, A. Maxia and P.Caboni, *J. Agric. Food Chem*., **2016**, 64, 4876–4881. (b) J. Y. Hwang, A. Y. Ji, S. H. Lee and E. J. Kang, *Org. Lett*., **2019**, 22, 16–21.

(2). M. Udayakumar, K. Jagatheeswaran, S. S. Ganesan, N. S. Venkataramanan, S. Madan Kumar, K. Byrappa and S. Thamotharan, *Journal of Molecular Structure*, **2017**, 1133, 510–518.

(3). Prasanna Kumari, B. Naveen, P. Suresh Kumar and S. Selva Ganesan, *Chem. Pap*., **2022**, 77, 151–158.

(4) K. Sharma, B. Das and P. Gogoi, *New J. Chem*., **2018**, 42, 18894–1890

(5) J. Jeong, P. Patel, H. Hwang, and S. Chang, *Org. Lett.* **2014**, 16, 4598–4601

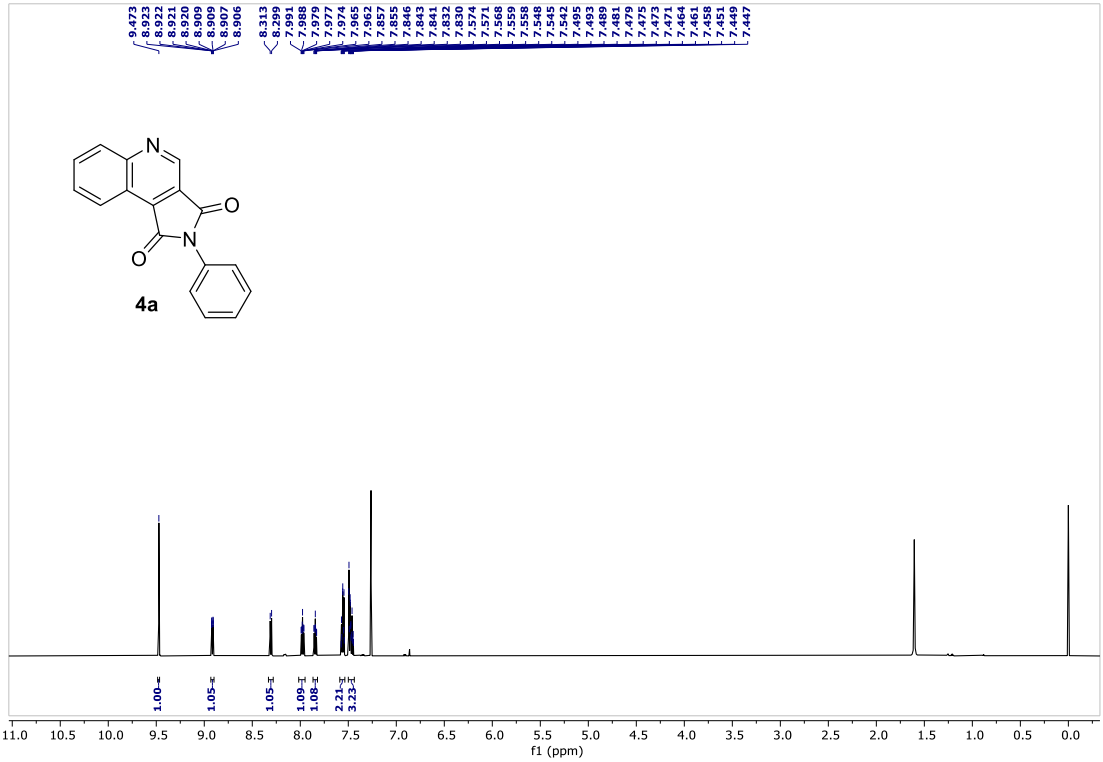


Figure S1: ^{1}H-NMR (600 MHz, $CDCl_3$) of compound **4a**

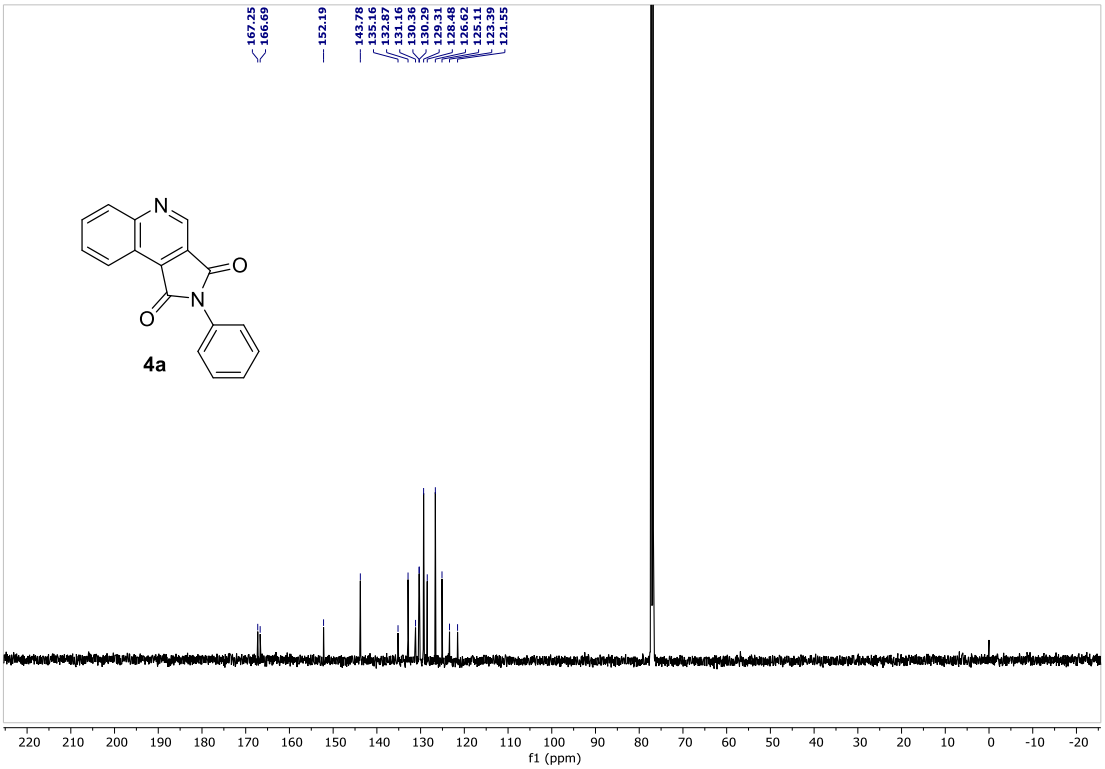
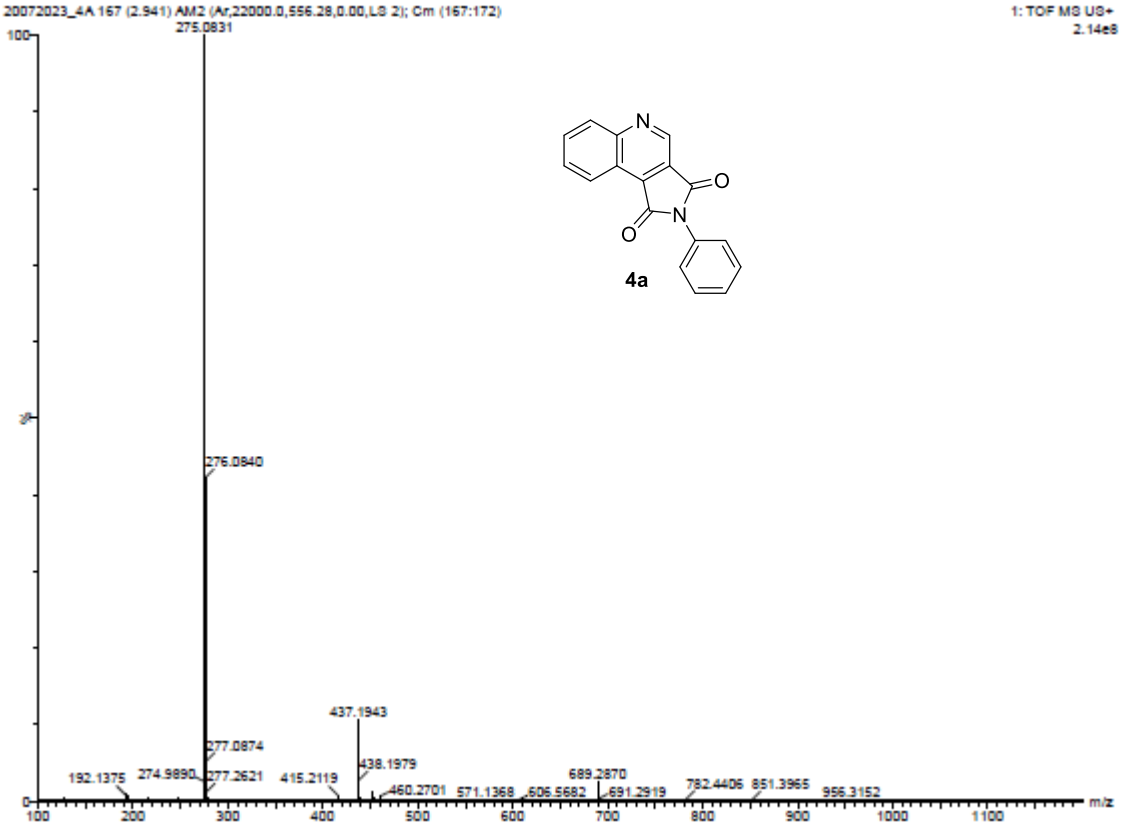


Figure S2: ^{13}C-NMR (150 MHz, $CDCl_3$) of compound **4a**



F

igure S3: HRMS spectra of compound **4a**

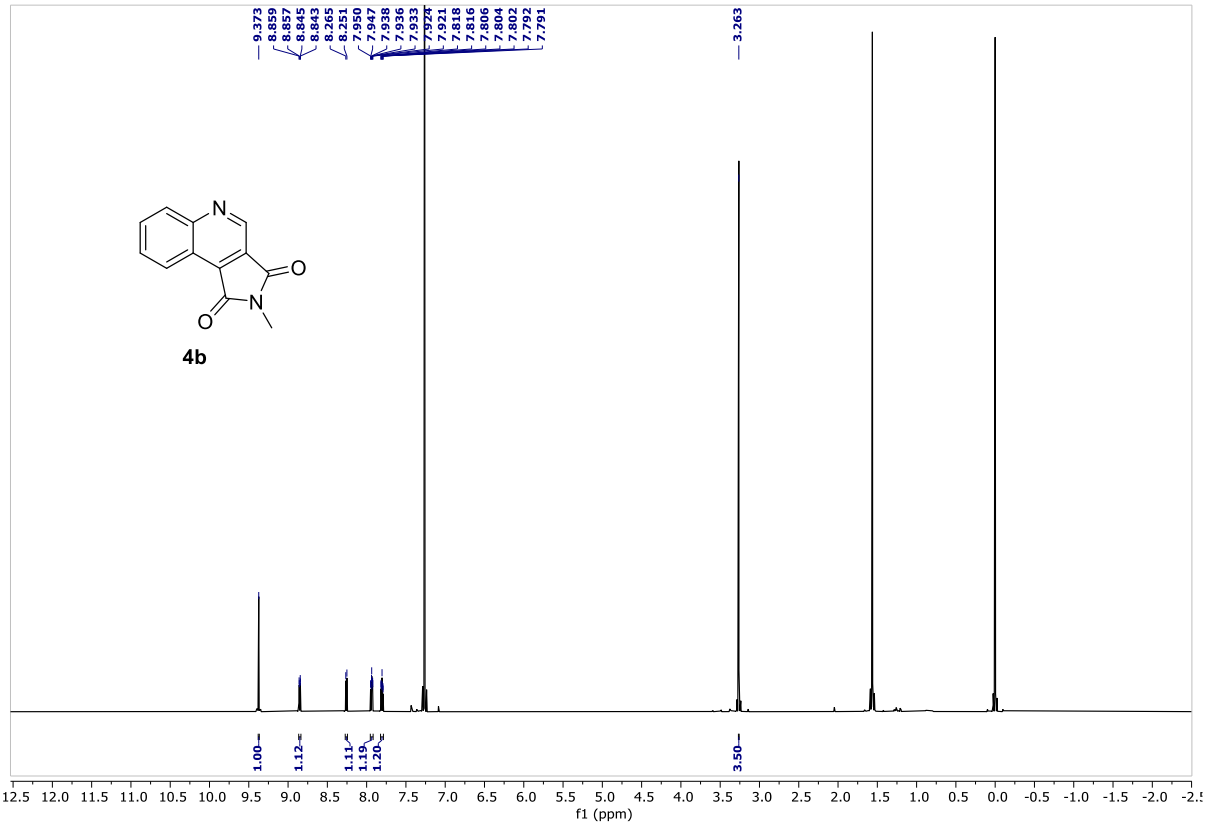


Figure S4: ^{1}H-NMR (600 MHz, $CDCl_3$) of compound **4b**

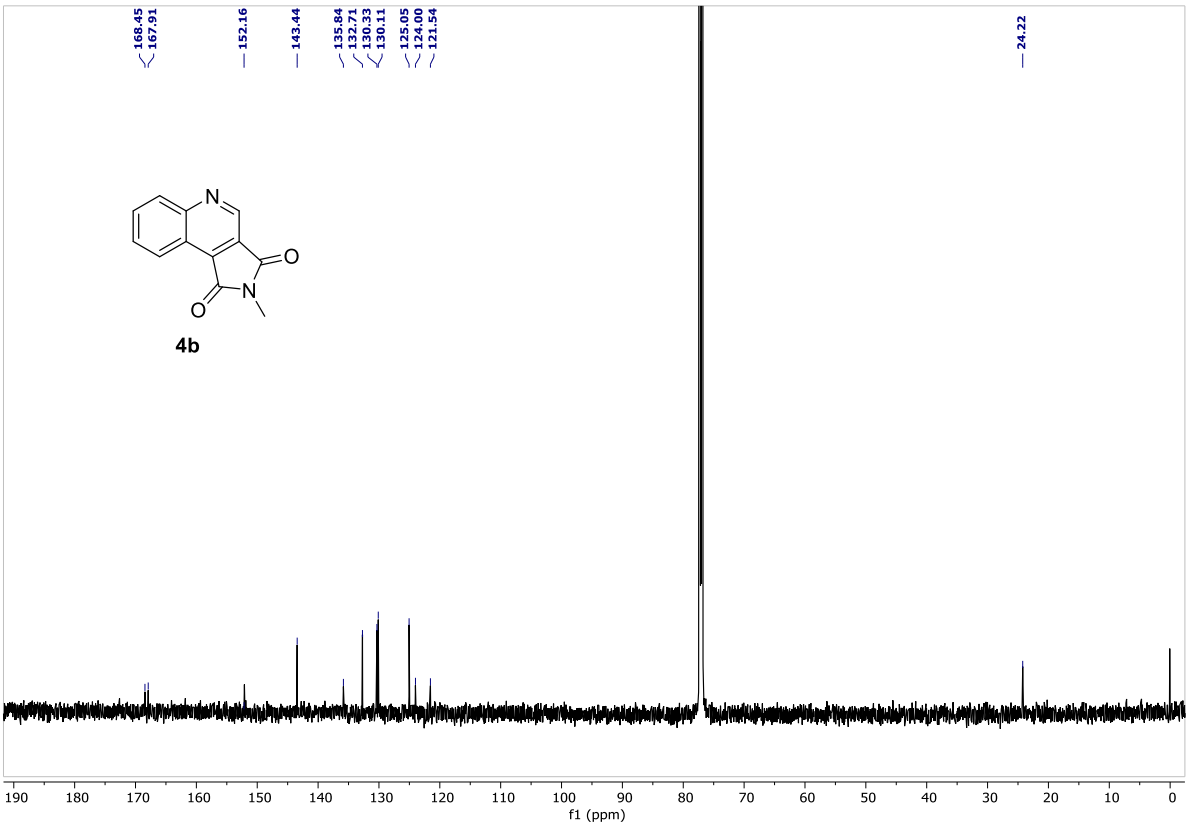


Figure S5: ^{13}**C-NMR** (150 MHz, $CDCl_3$) of compound **4b**

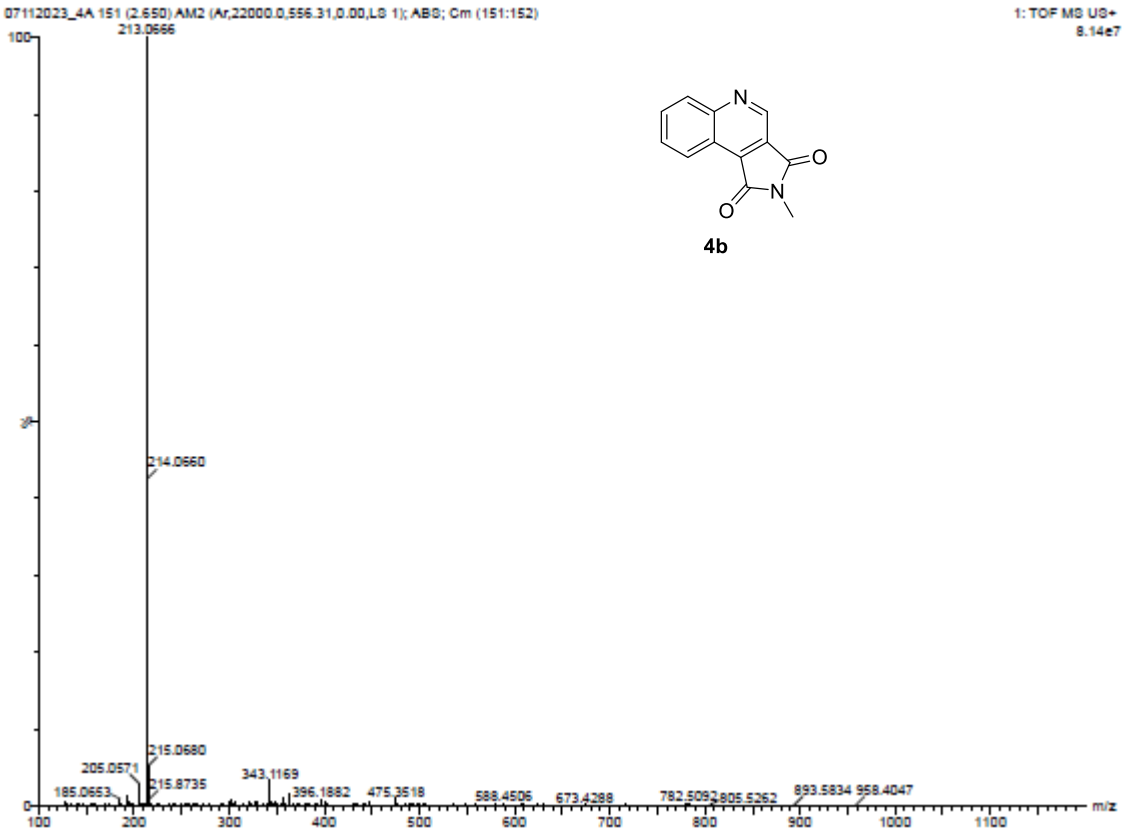


Figure S6: HRMS spectra of compound **4b**

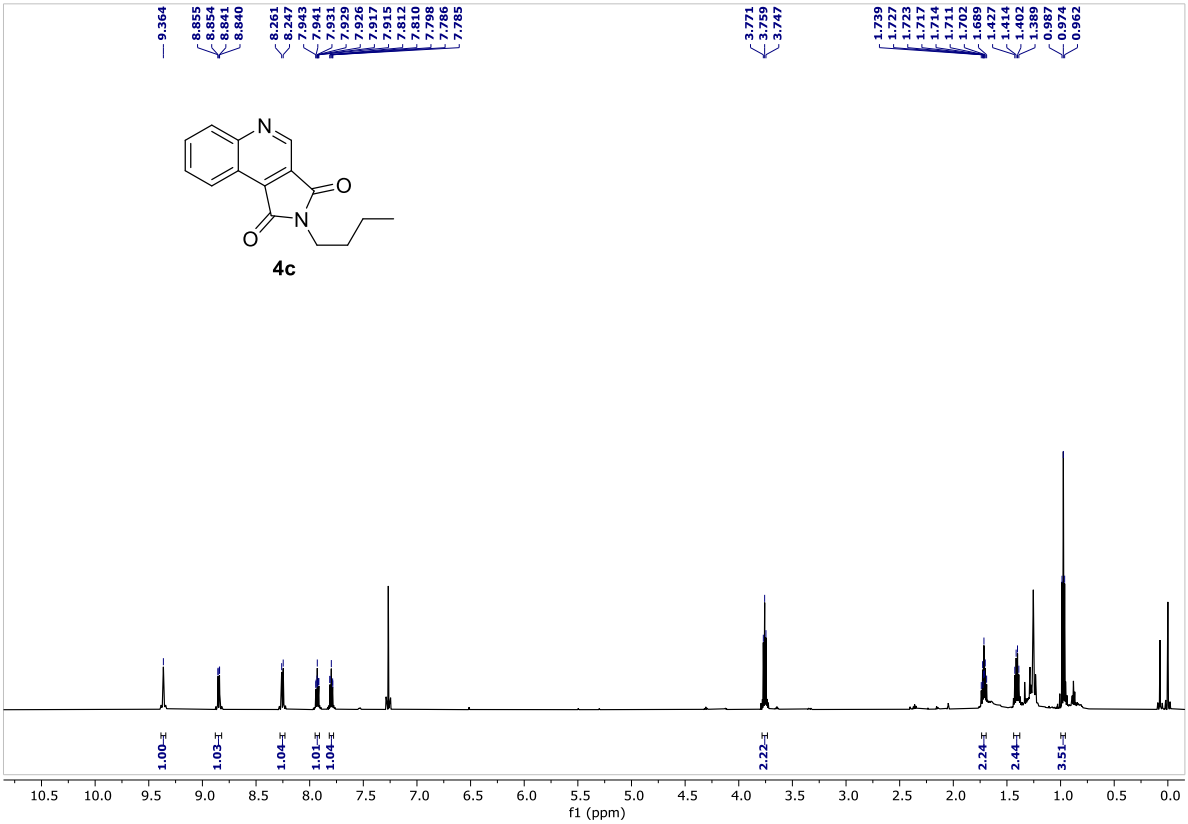


Figure S7: ^{1}H-NMR (600 MHz, $CDCl_3$) of compound **4c**

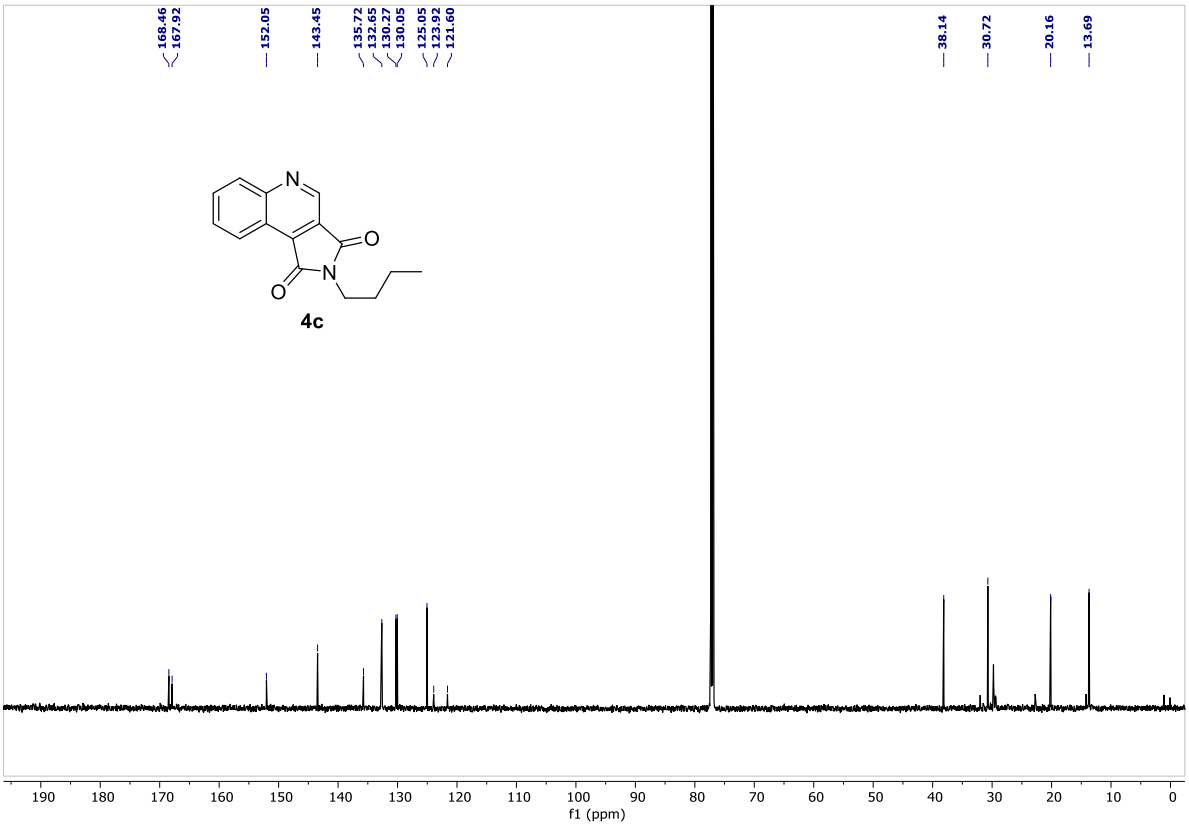


Figure S8: ^{13}C-NMR (150 MHz, $CDCl_3$) of compound **4c**

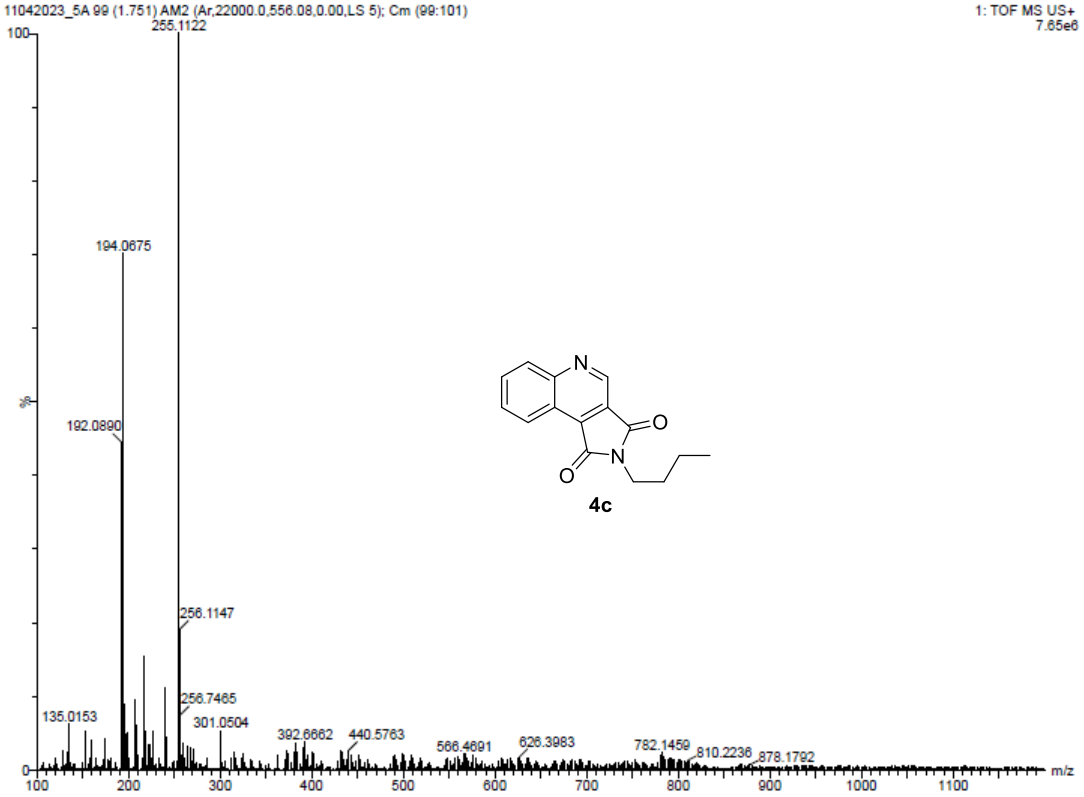


Figure S9: HRMS spectra of compound **4c**

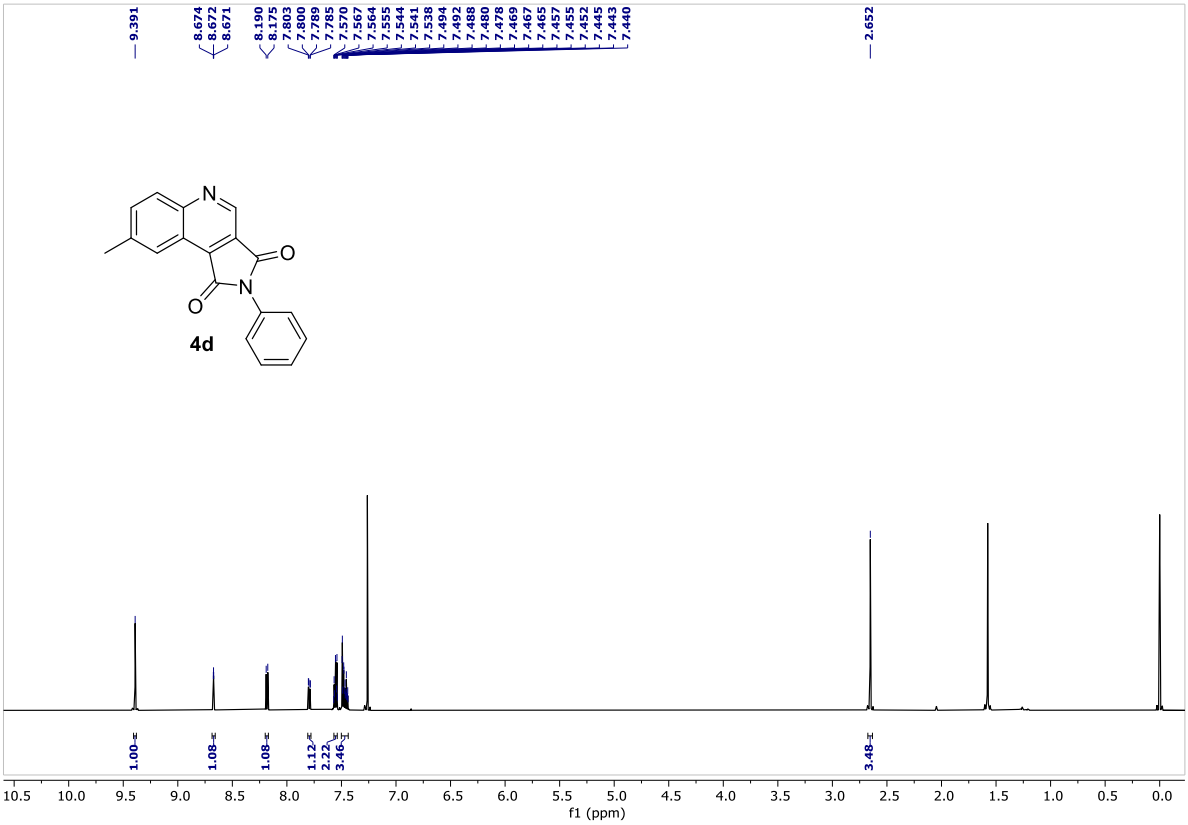


Figure S10: **^{1}H-NMR** (600 MHz, $CDCl_3$) of compound **4d**

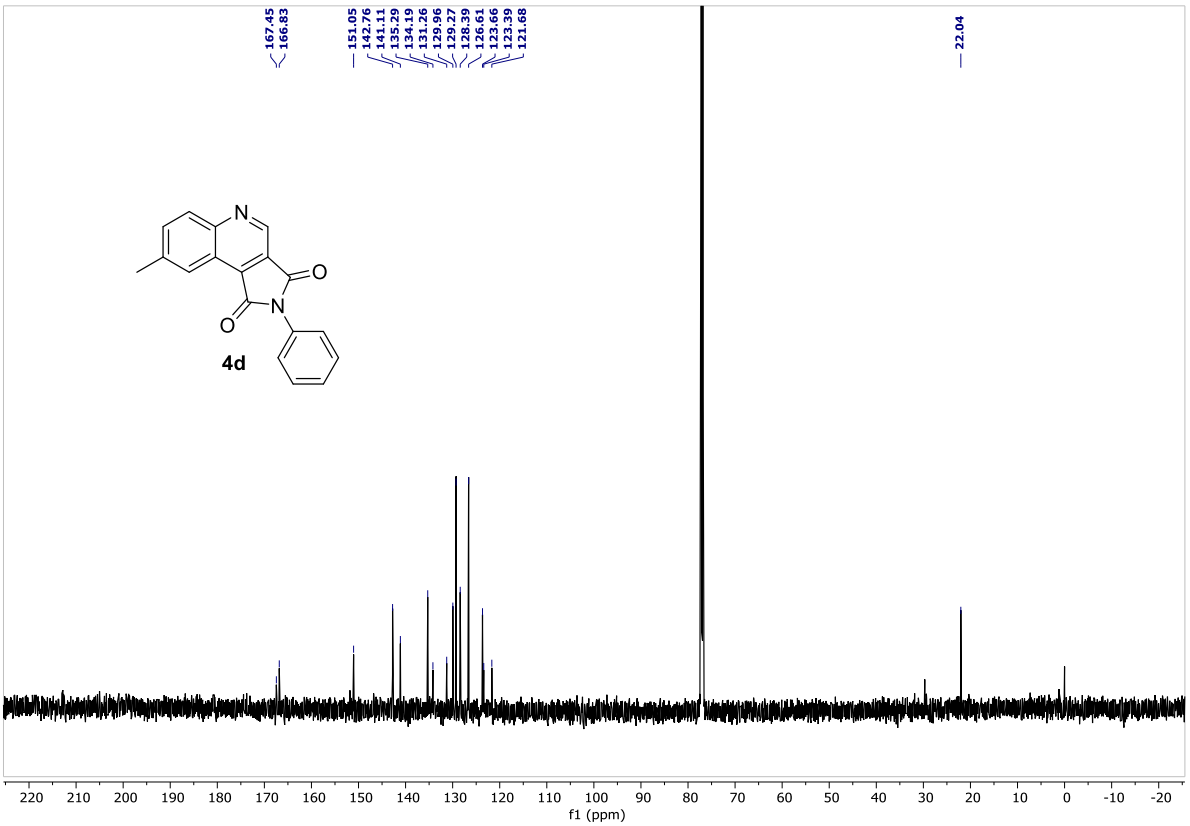


Figure S11: ^{13}**C-NMR** (150 MHz, $CDCl_3$) of compound **4d**

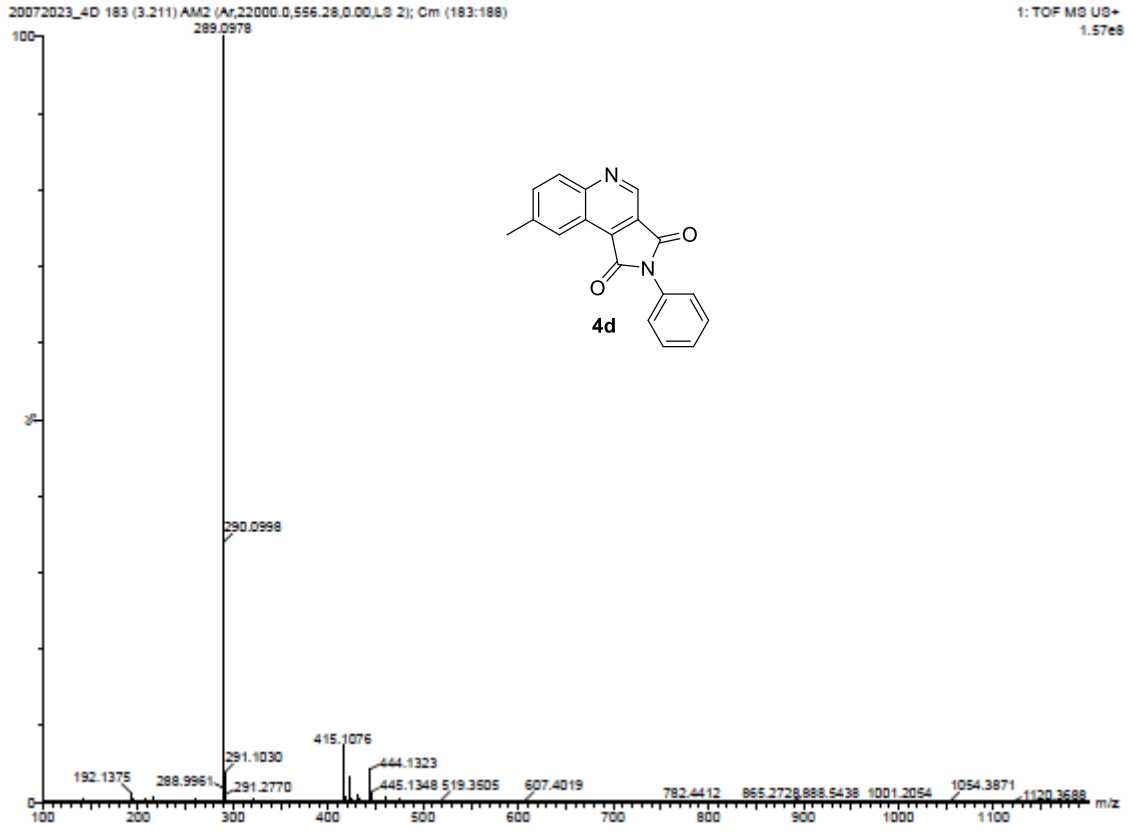


Figure S12: HRMS spectra of compound **4d**

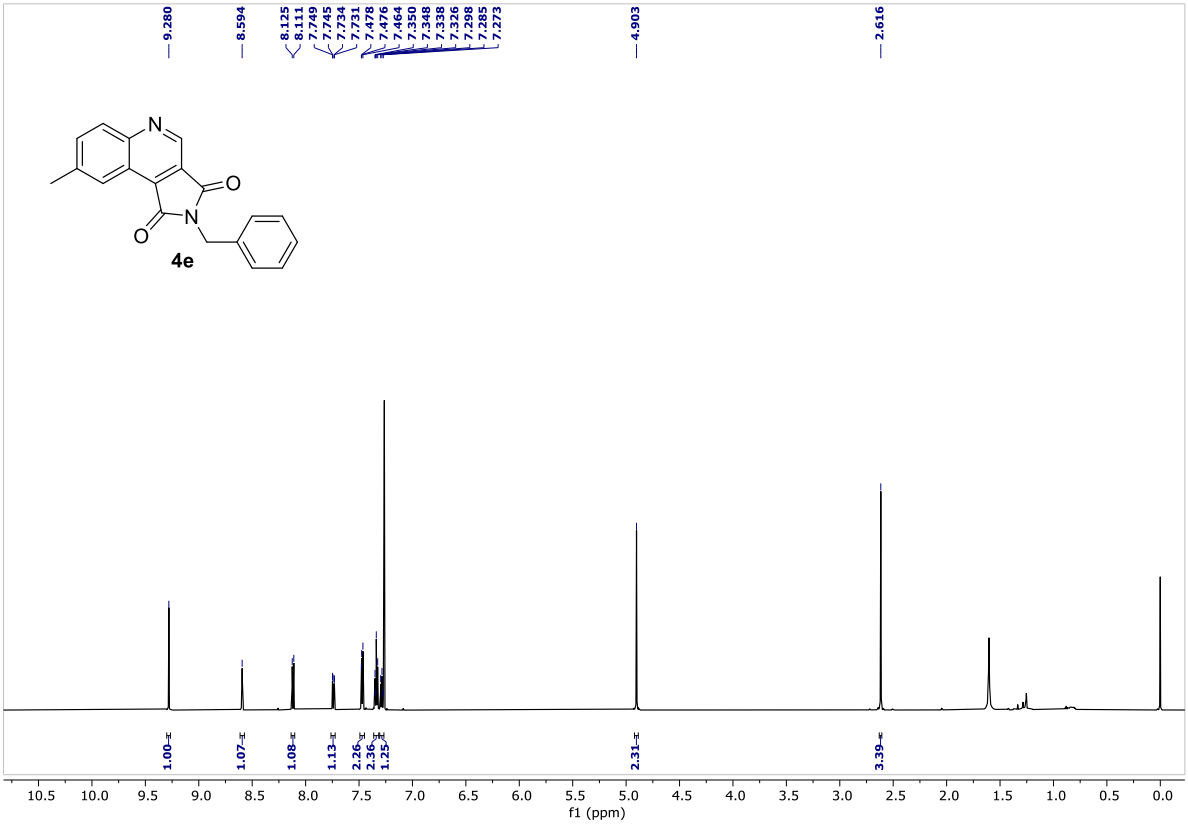


Figure S13: ^{1}H-NMR (600 MHz, $CDCl_3$) of compound **4e**

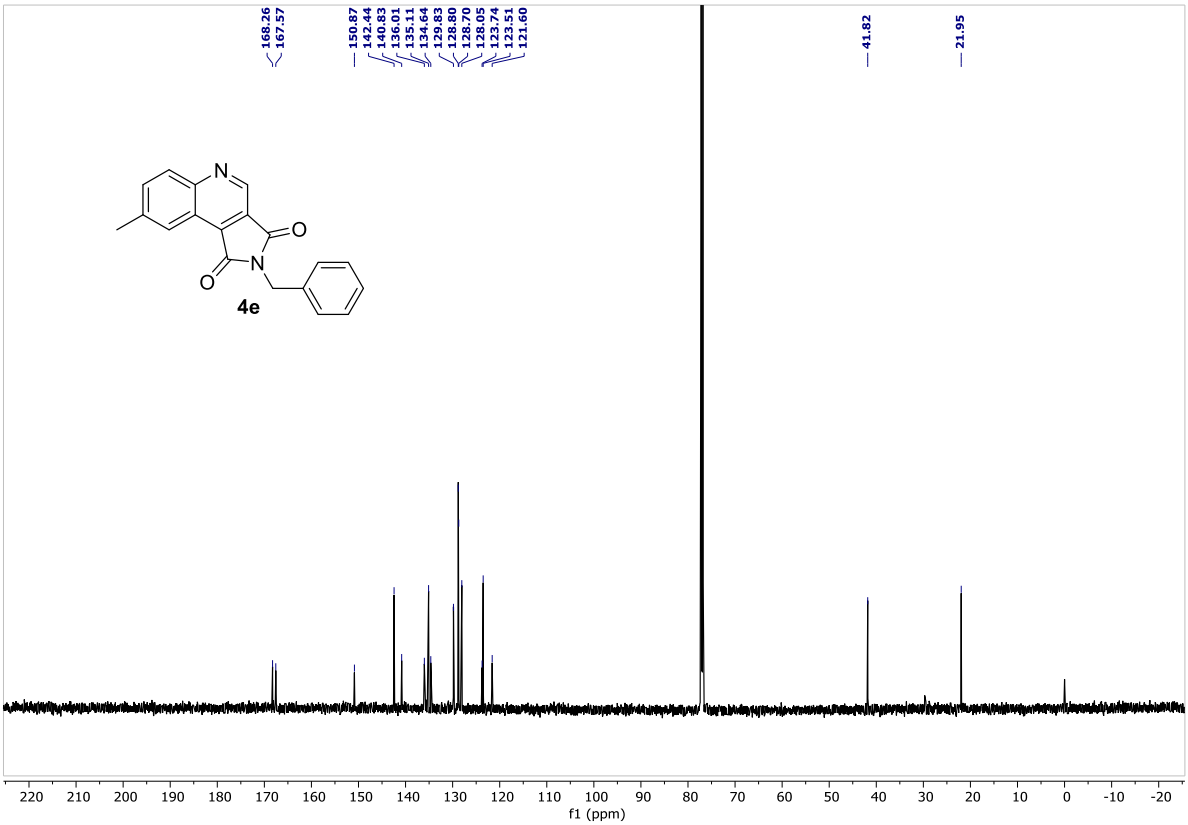


Figure S14: ^{13}C-NMR (150 MHz, $CDCl_3$) of compound **4e**

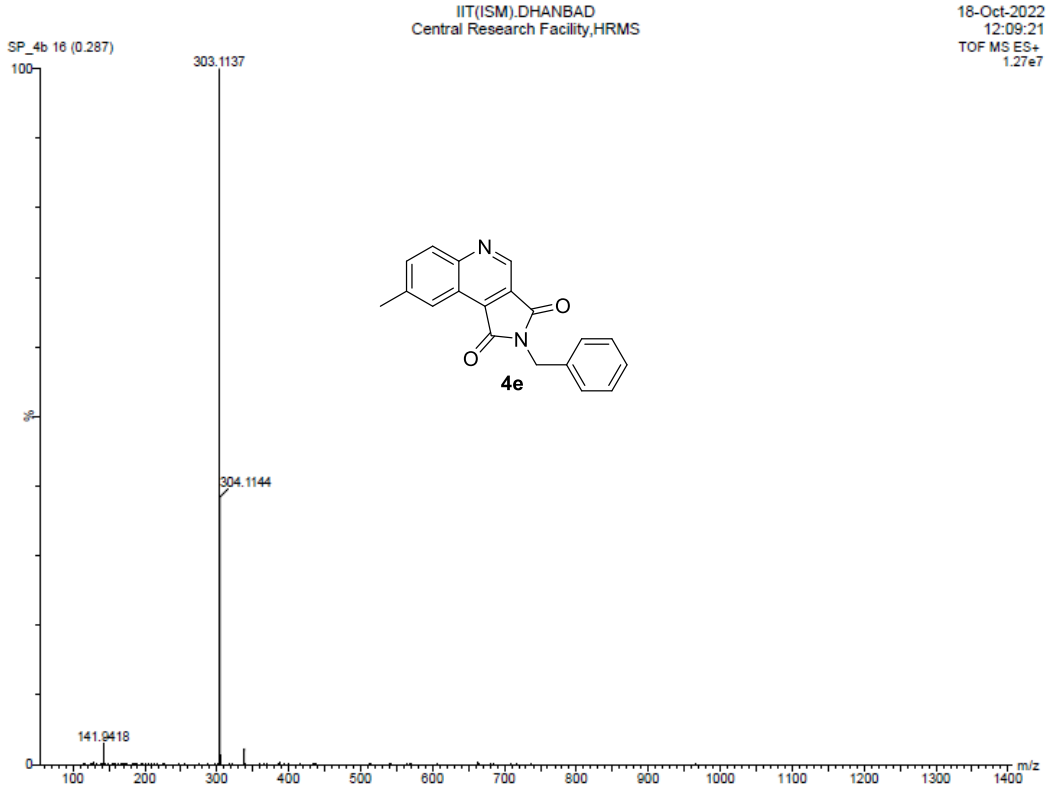


Figure S15: HRMS spectra of compound **4e**

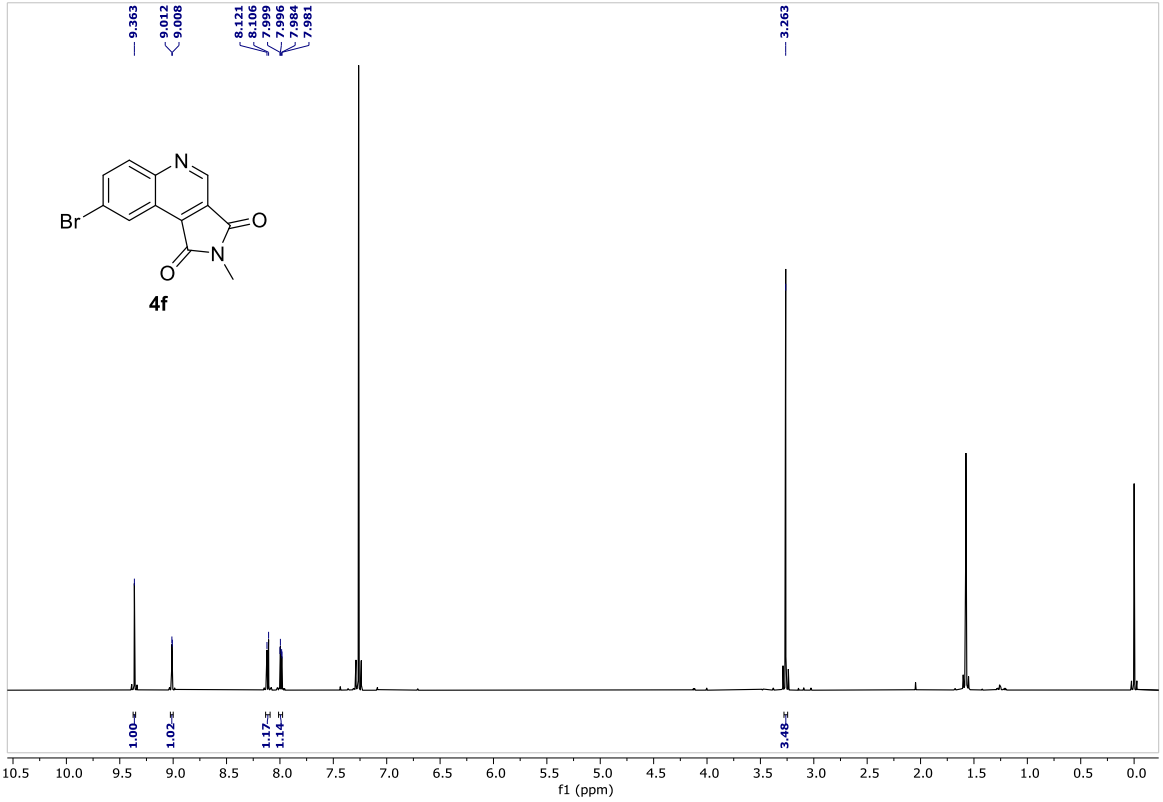


Figure S16: **^{1}H-NMR** (600 MHz, $CDCl_3$) of compound **4f**

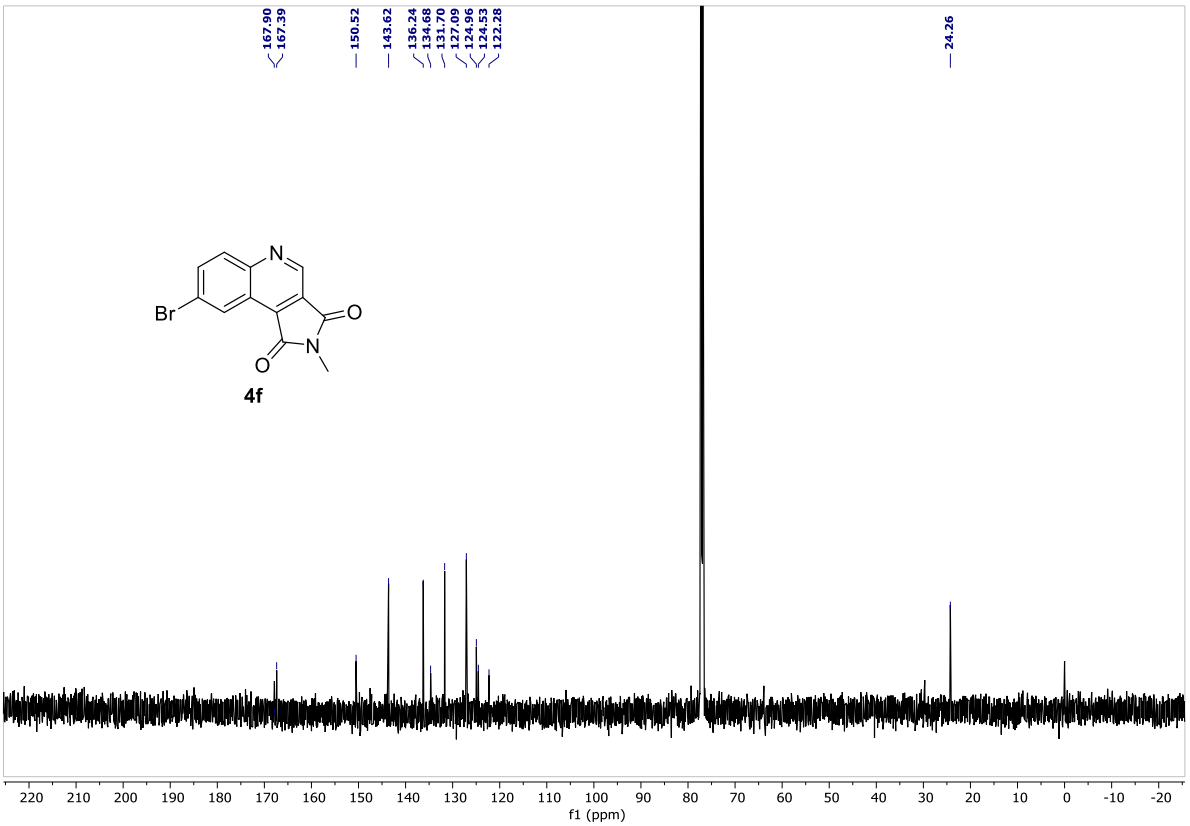


Figure S17: ^{13}C-NMR (150 MHz, $CDCl_3$) of compound **4f**

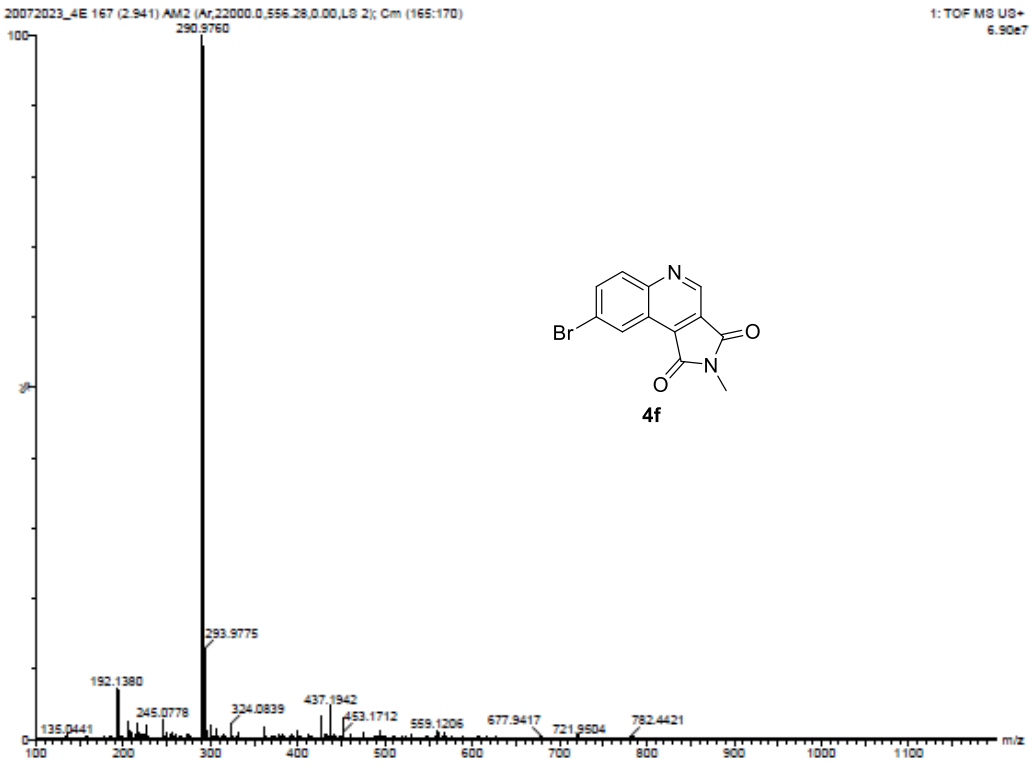


Figure S18: HRMS spectra of compound **4f**

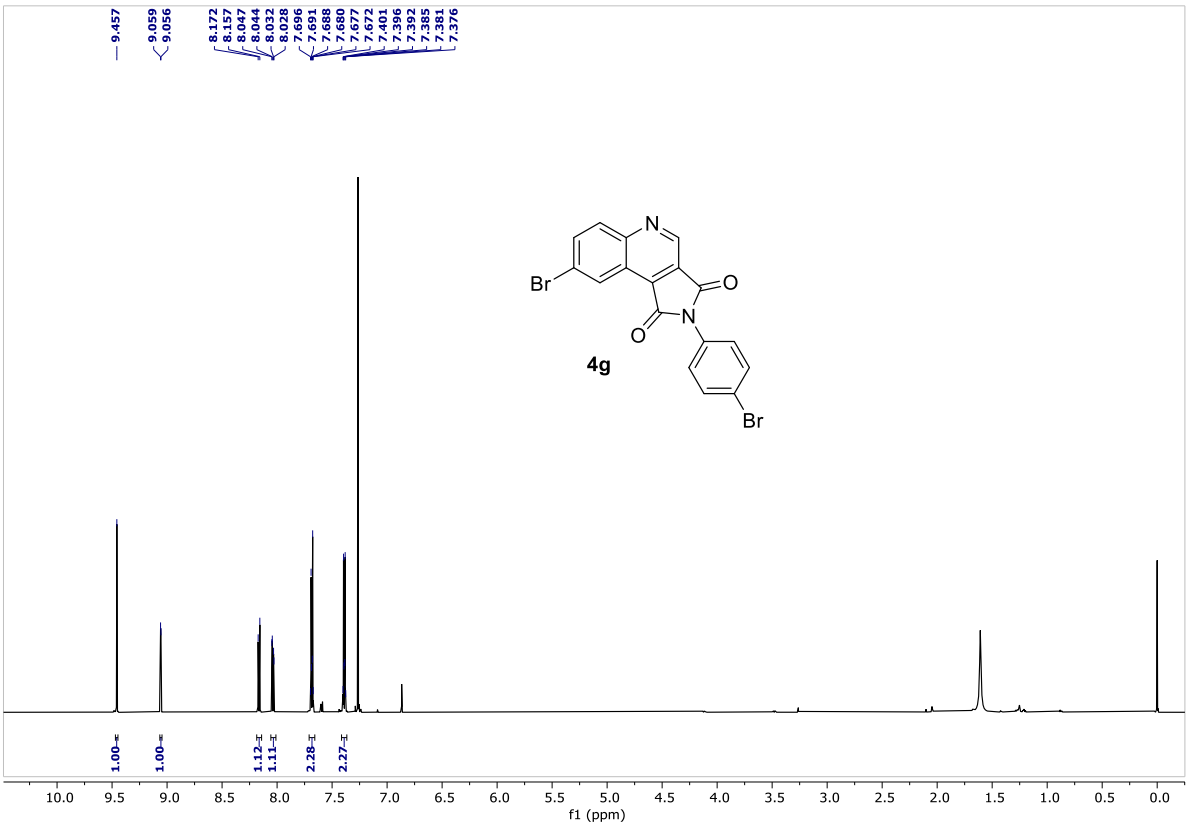


Figure S19: ^{1}H-NMR (600 MHz, $CDCl_3$) of compound **4g**

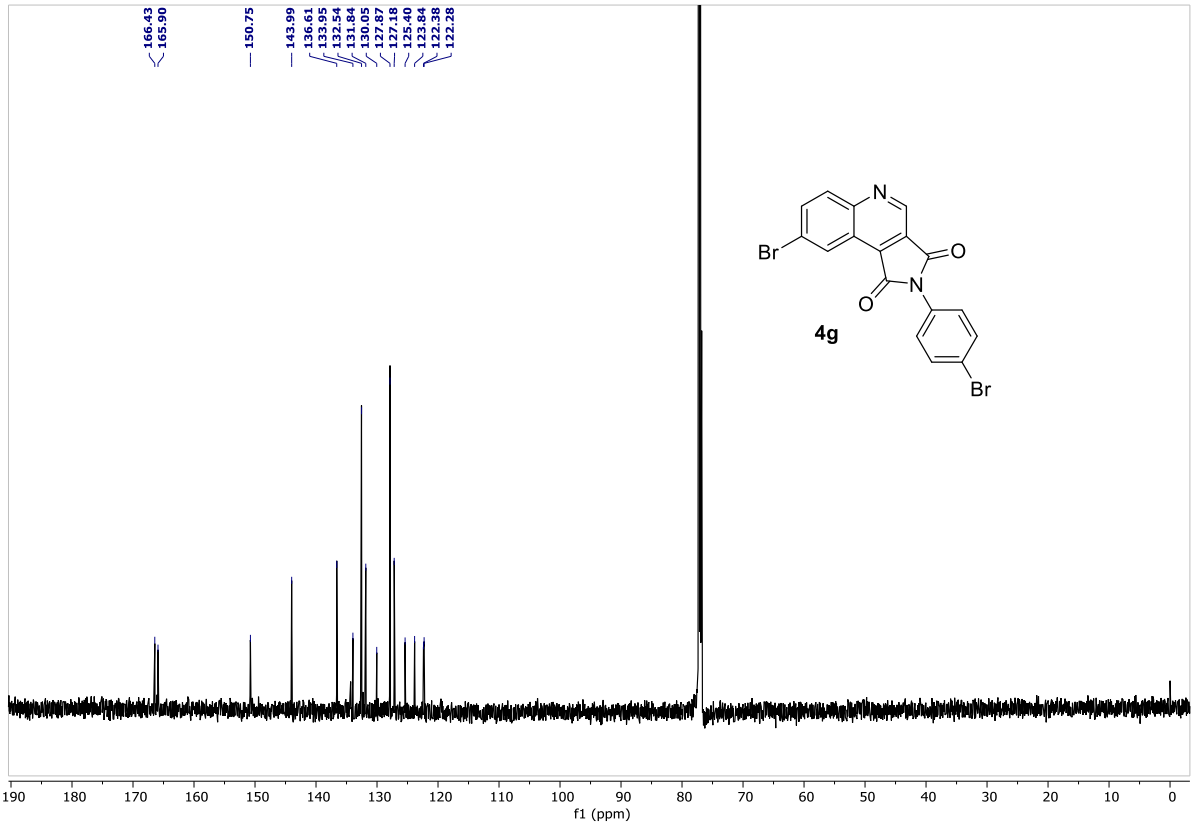


Figure S20: ^{13}C-NMR (150 MHz, $CDCl_3$) of compound **4g**

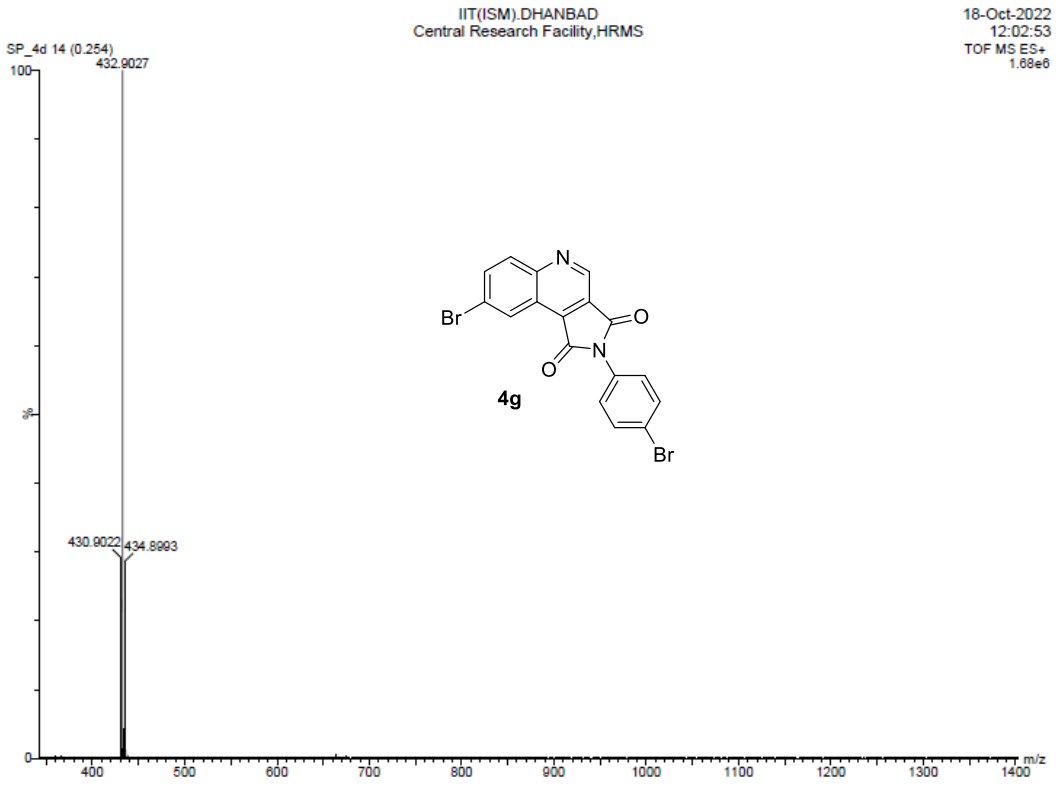


Figure S21: HRMS spectra of compound **4g**

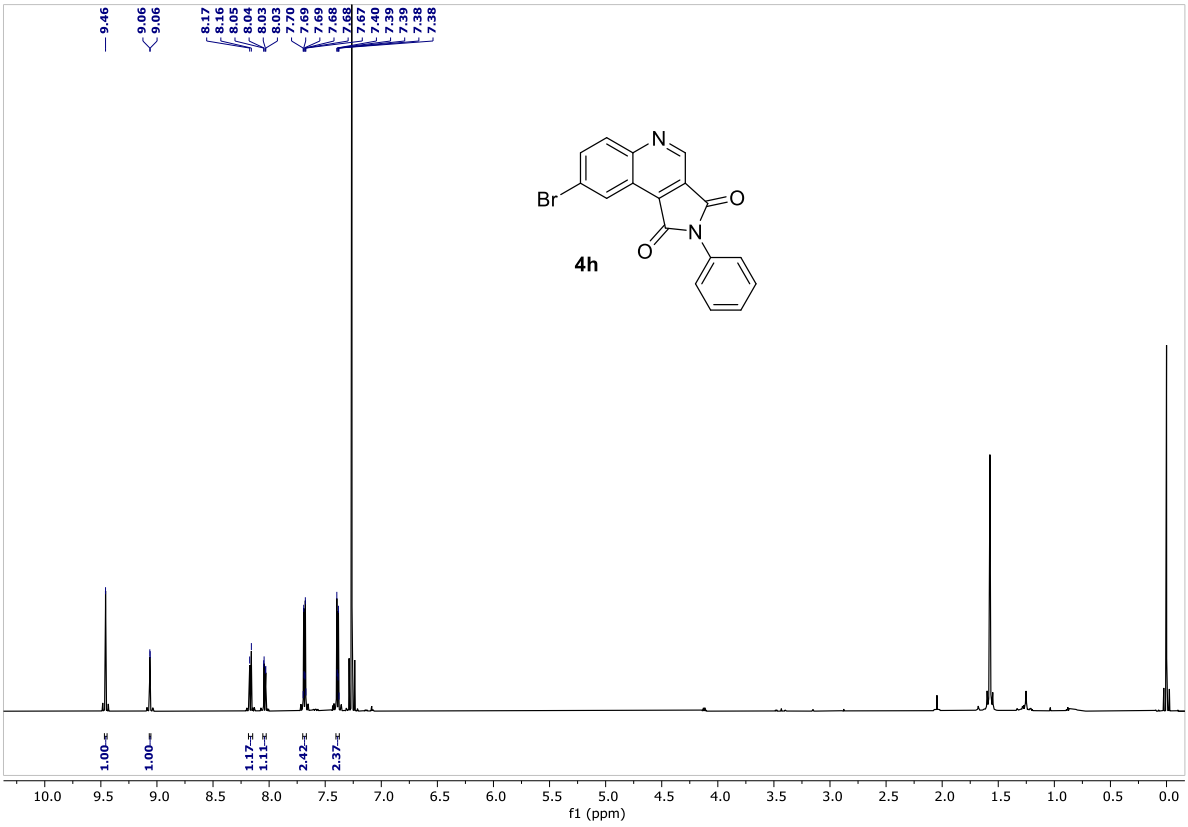


Figure S22: **^{1}H-NMR** (600 MHz, $CDCl_3$) of compound **4h**

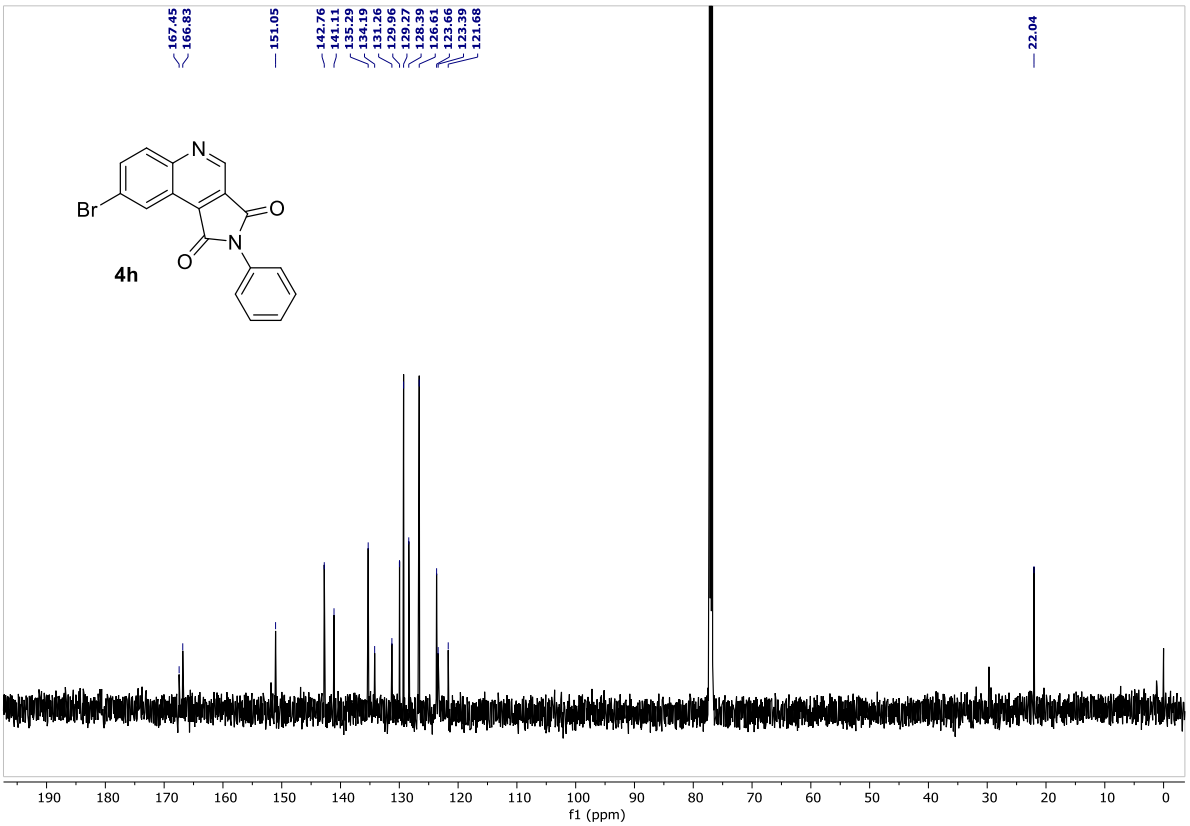


Figure S23: ^{13}**C-NMR** (150 MHz, $CDCl_3$) of compound **4h**

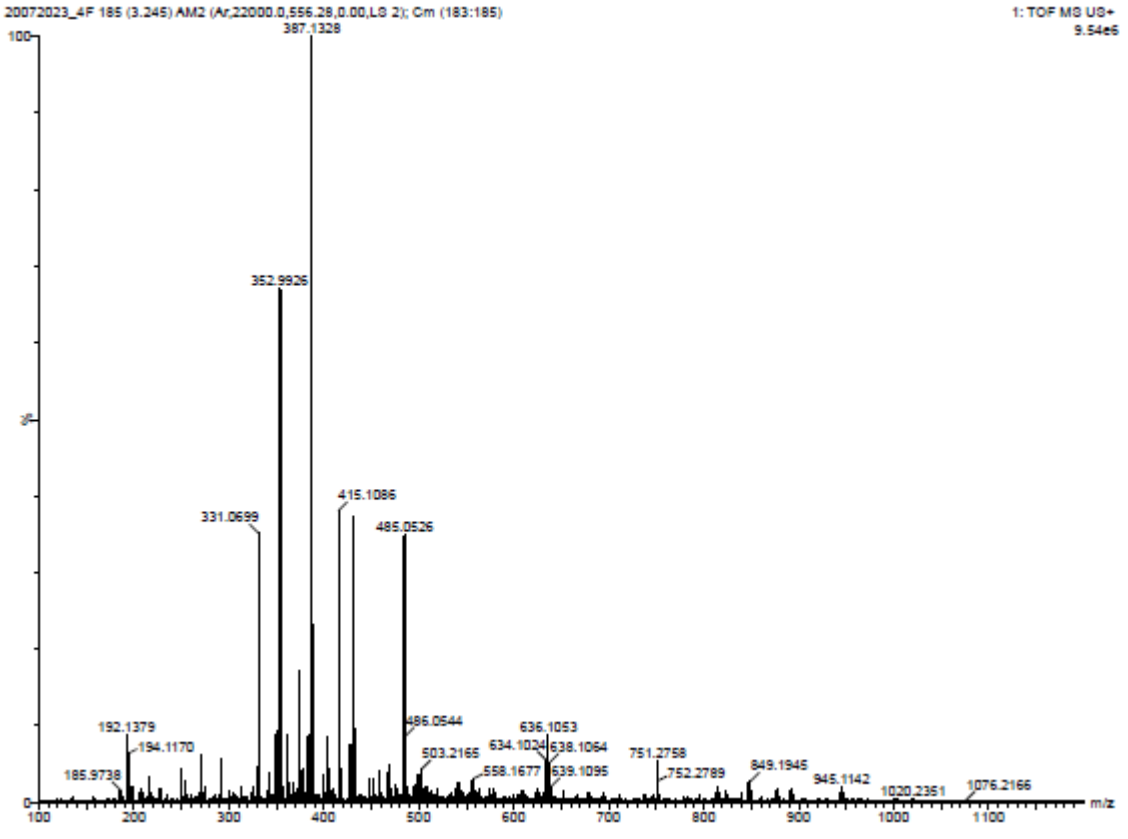


Figure S24: HRMS spectra of compound **4h**

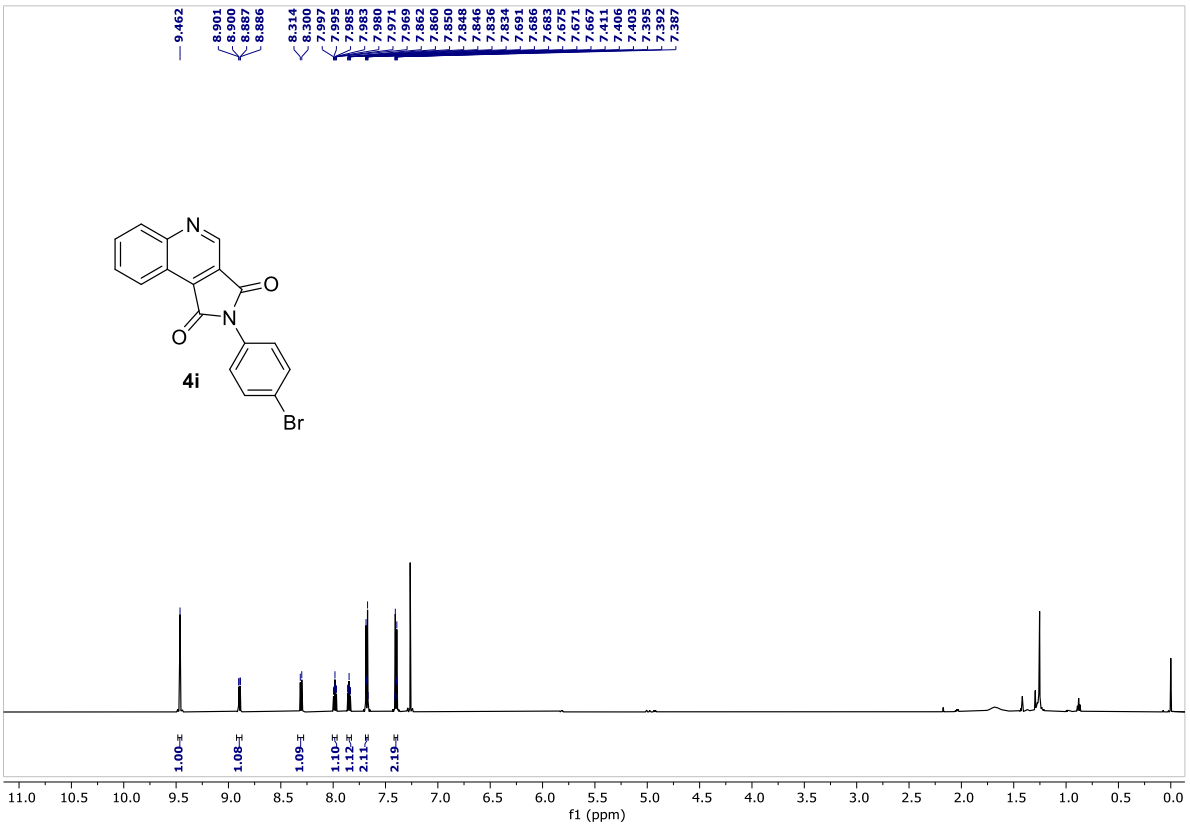


Figure S25: ^{1}H-NMR (600 MHz, $CDCl_3$) of compound **4i**

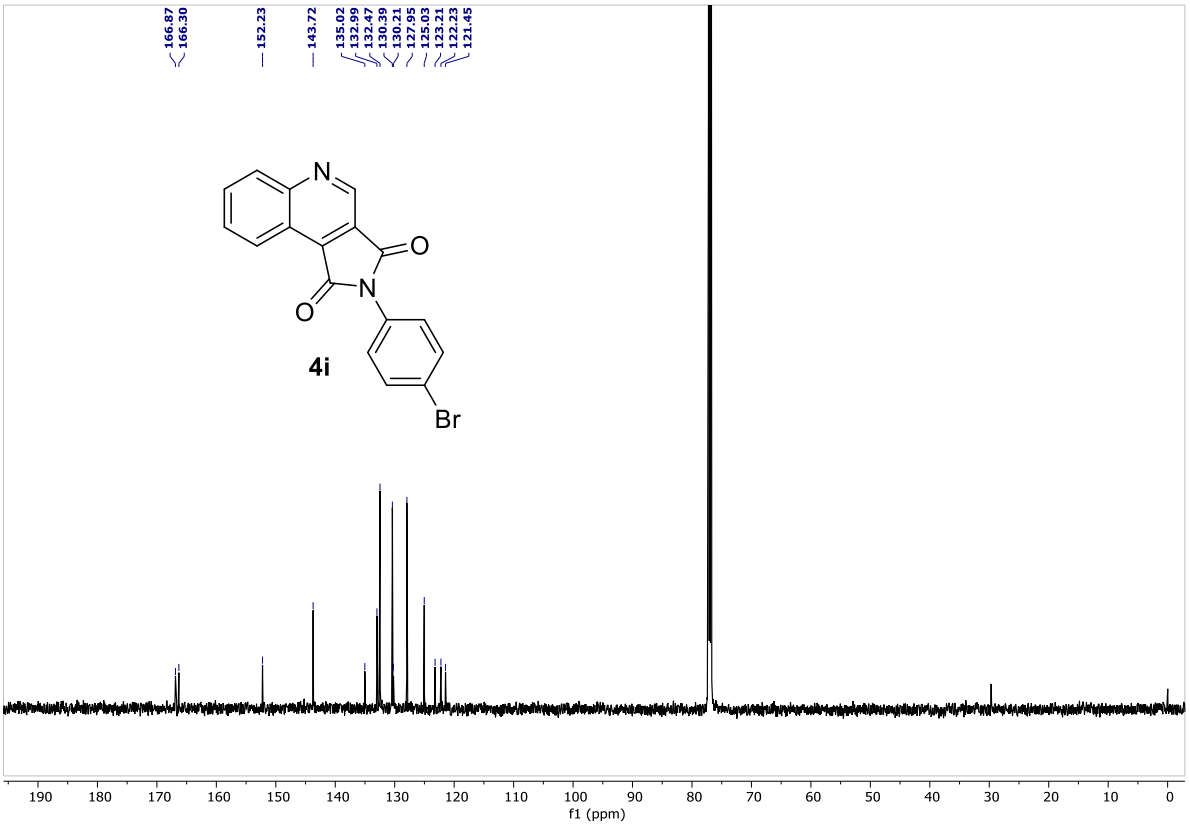


Figure S26: ^{13}C-NMR (150 MHz, $CDCl_3$) of compound **4i**

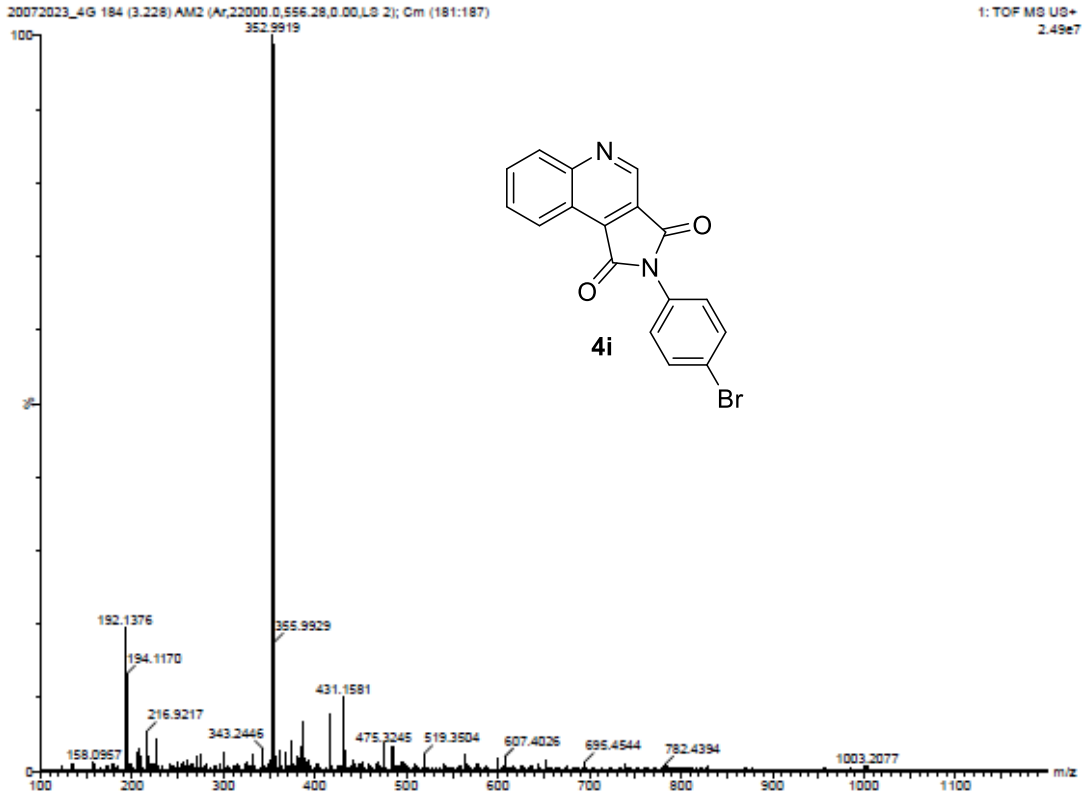


Figure S27: HRMS spectra of compound **4i**

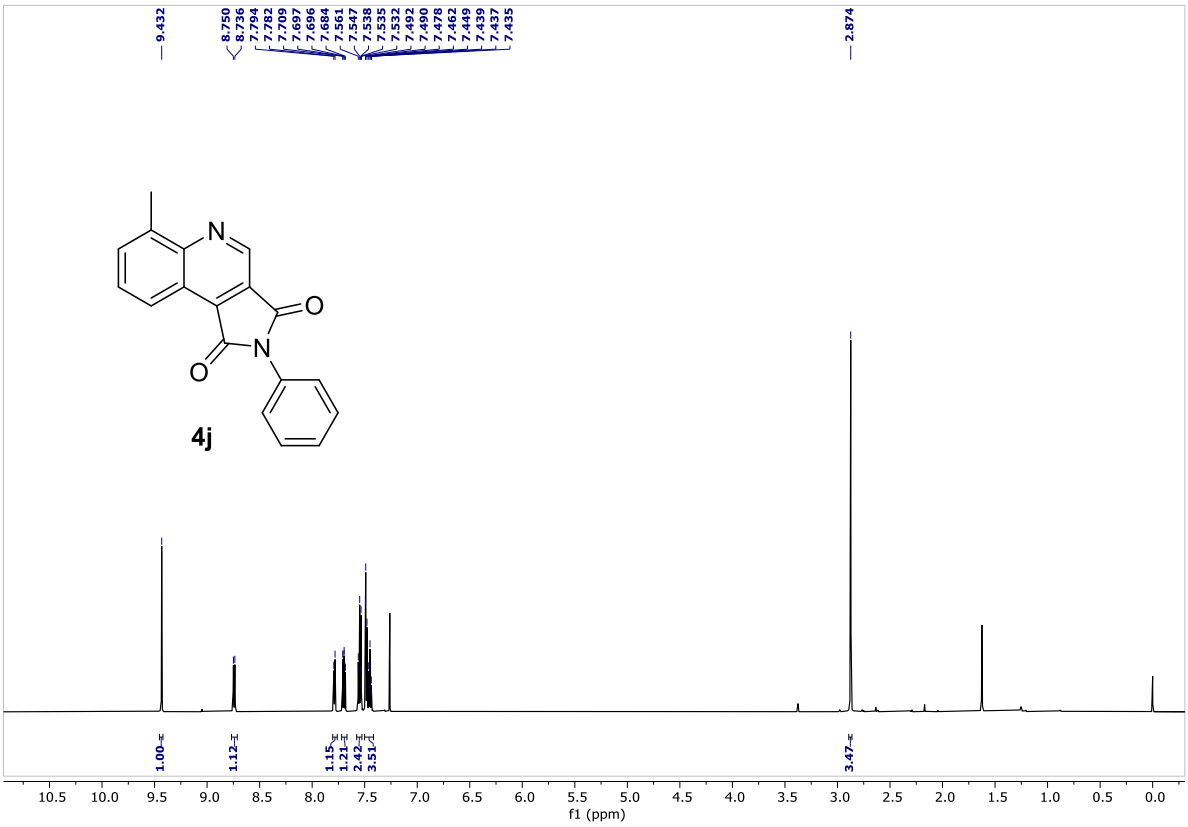


Figure S28: **^{1}H-NMR** (600 MHz, $CDCl_3$) of compound **4j**

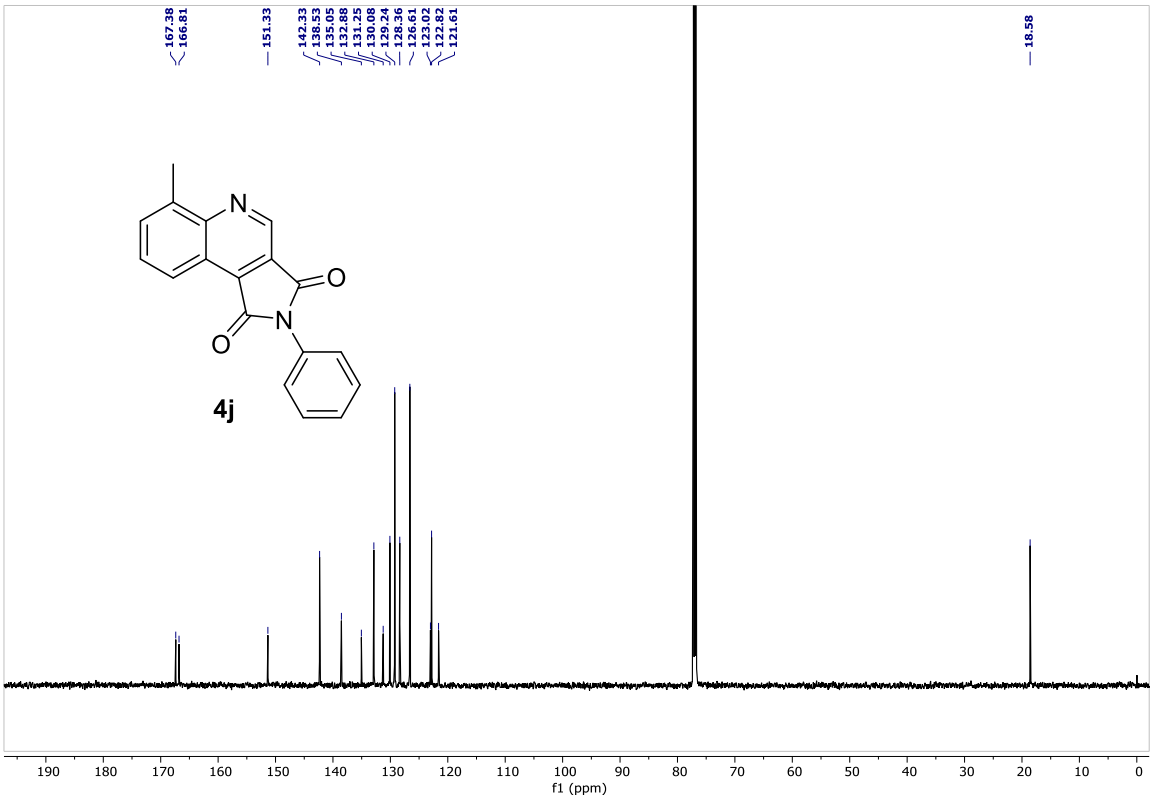


Figure S29: ^{13}C-NMR (150 MHz, $CDCl_3$) of compound **4j**

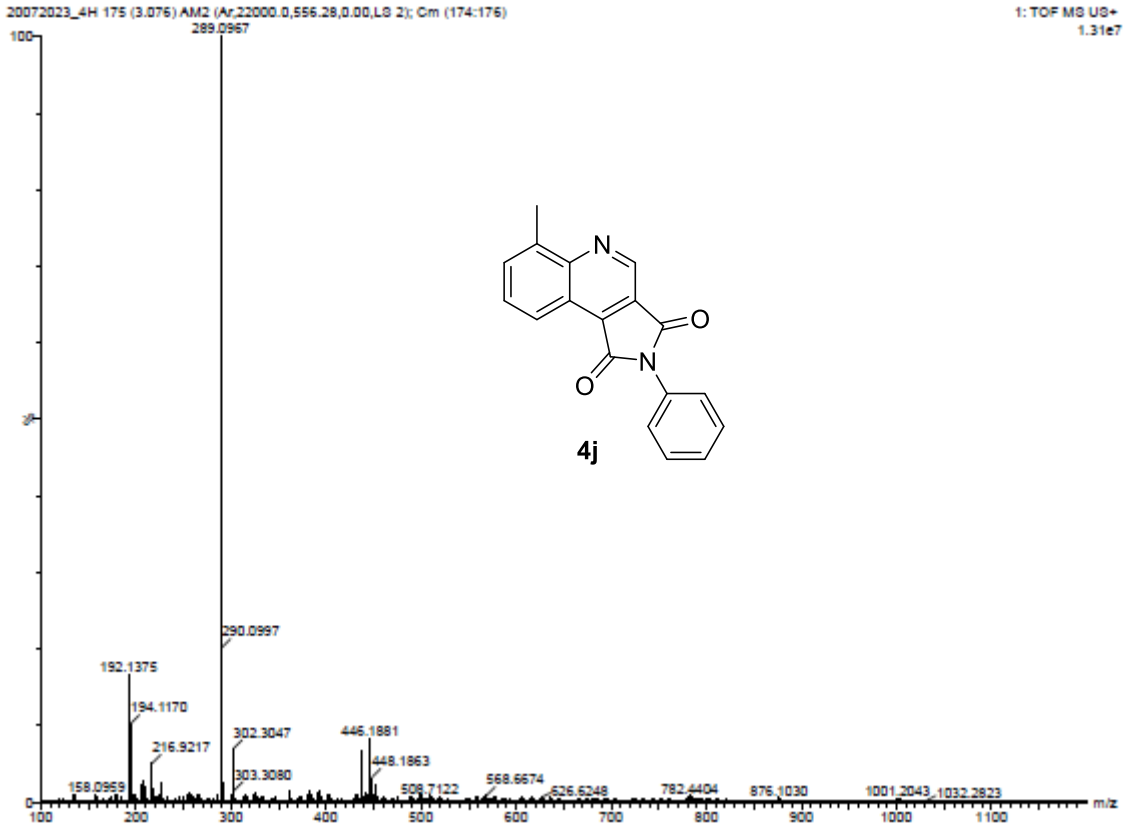


Figure S30: HRMS spectra of compound **4j**

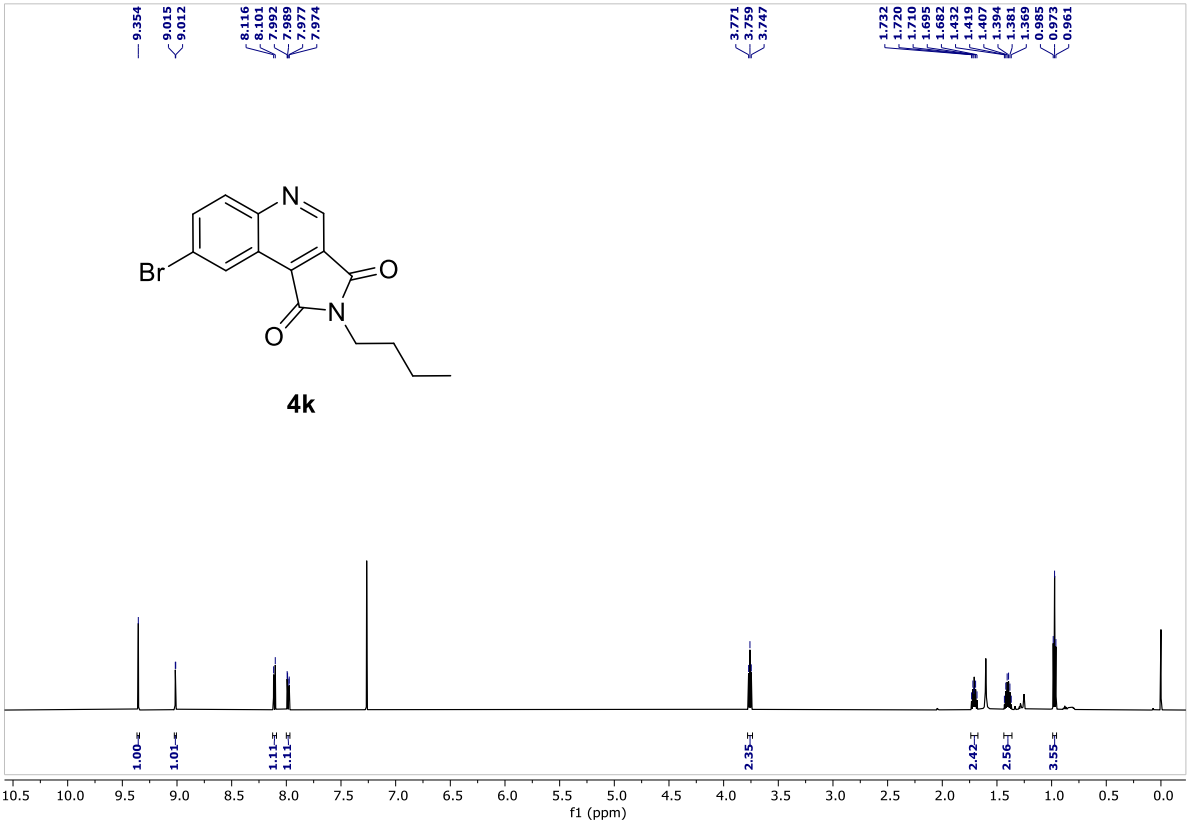


Figure S31: ^{1}H-NMR (600 MHz, $CDCl_3$) of compound **4k**

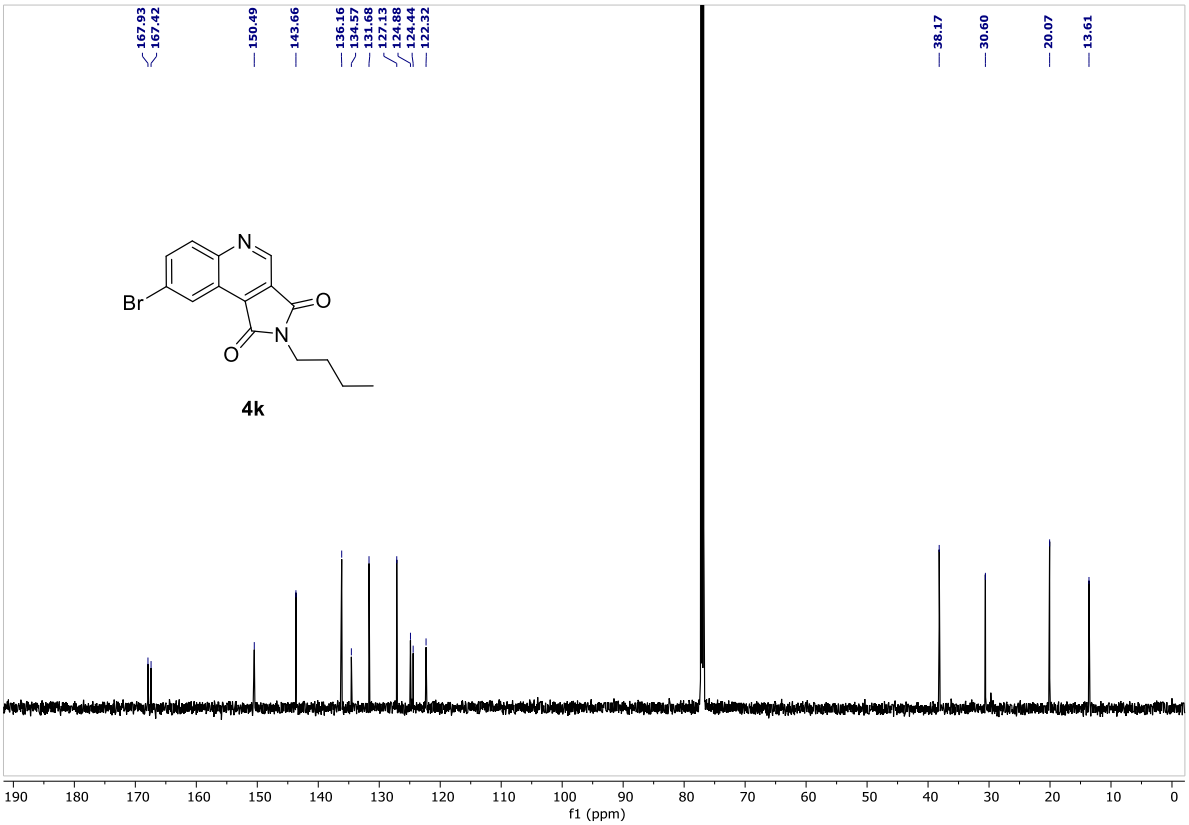


Figure S32: ^{13}C-NMR (150 MHz, $CDCl_3$) of compound **4k**

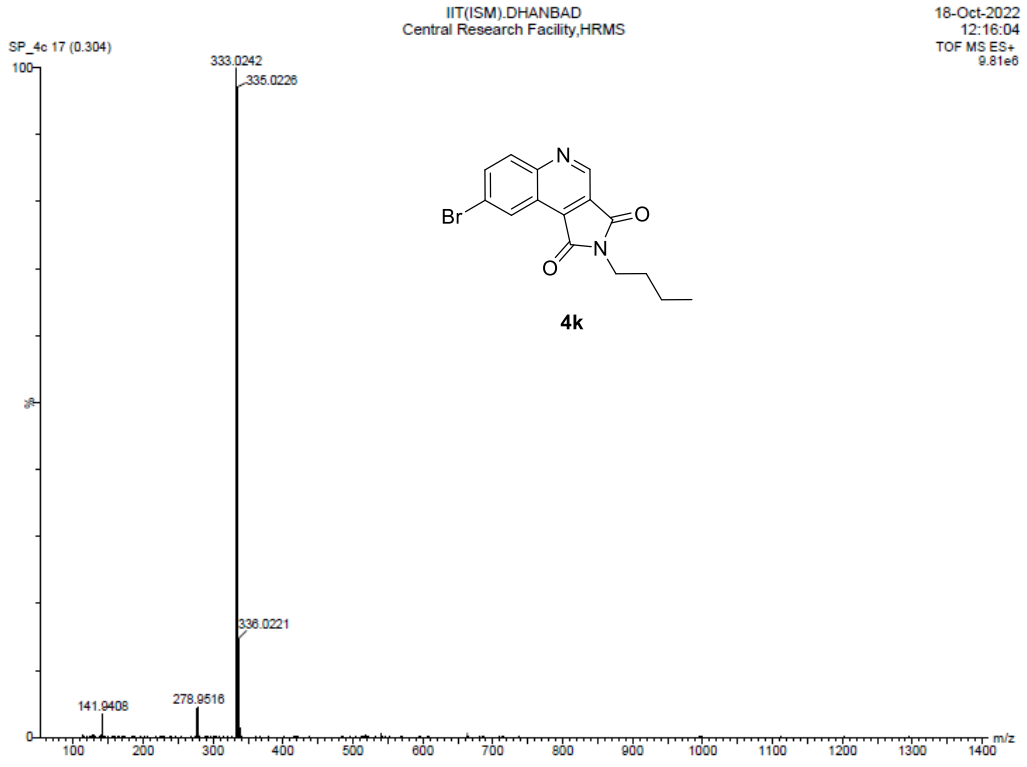


Figure S33: HRMS spectra of compound **4k**

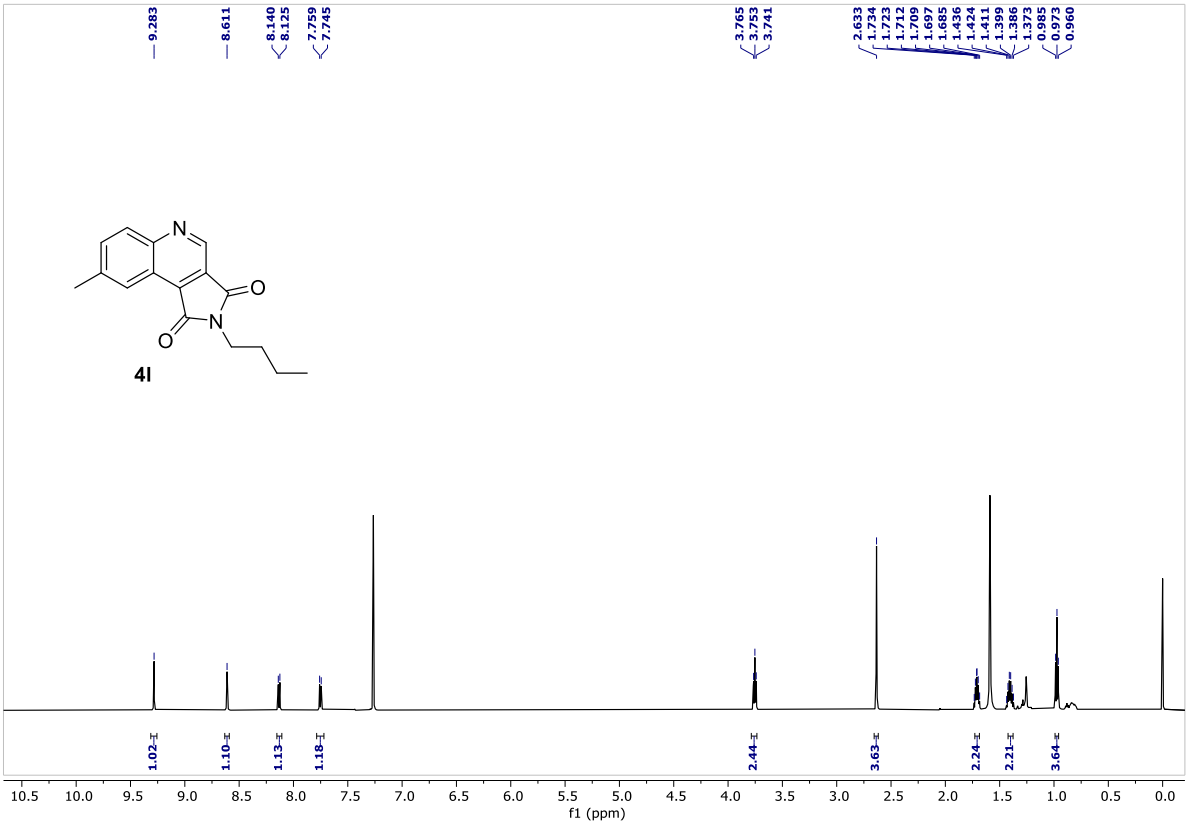


Figure S34: **^{1}H-NMR** (600 MHz, $CDCl_3$) of compound **4l**

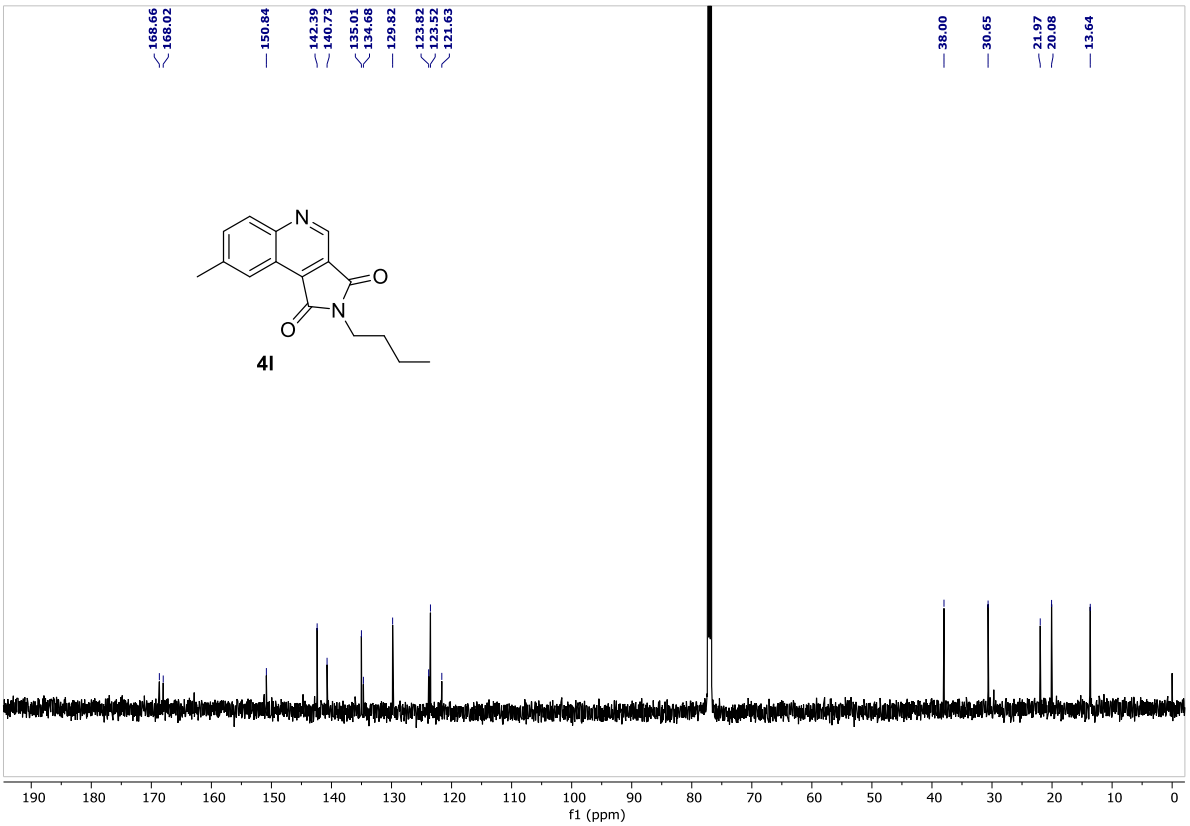


Figure S35: ^{13}**C-NMR** (150 MHz, $CDCl_3$) of compound **4l**

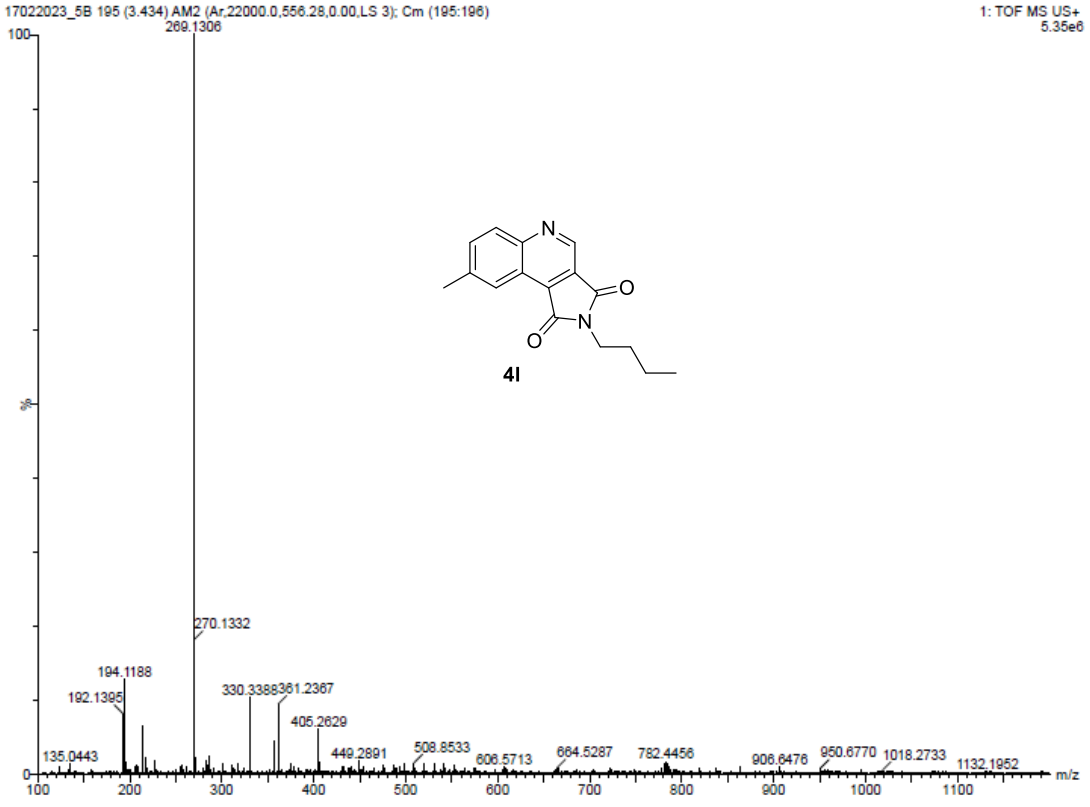


Figure S36: HRMS spectra of compound **4l**

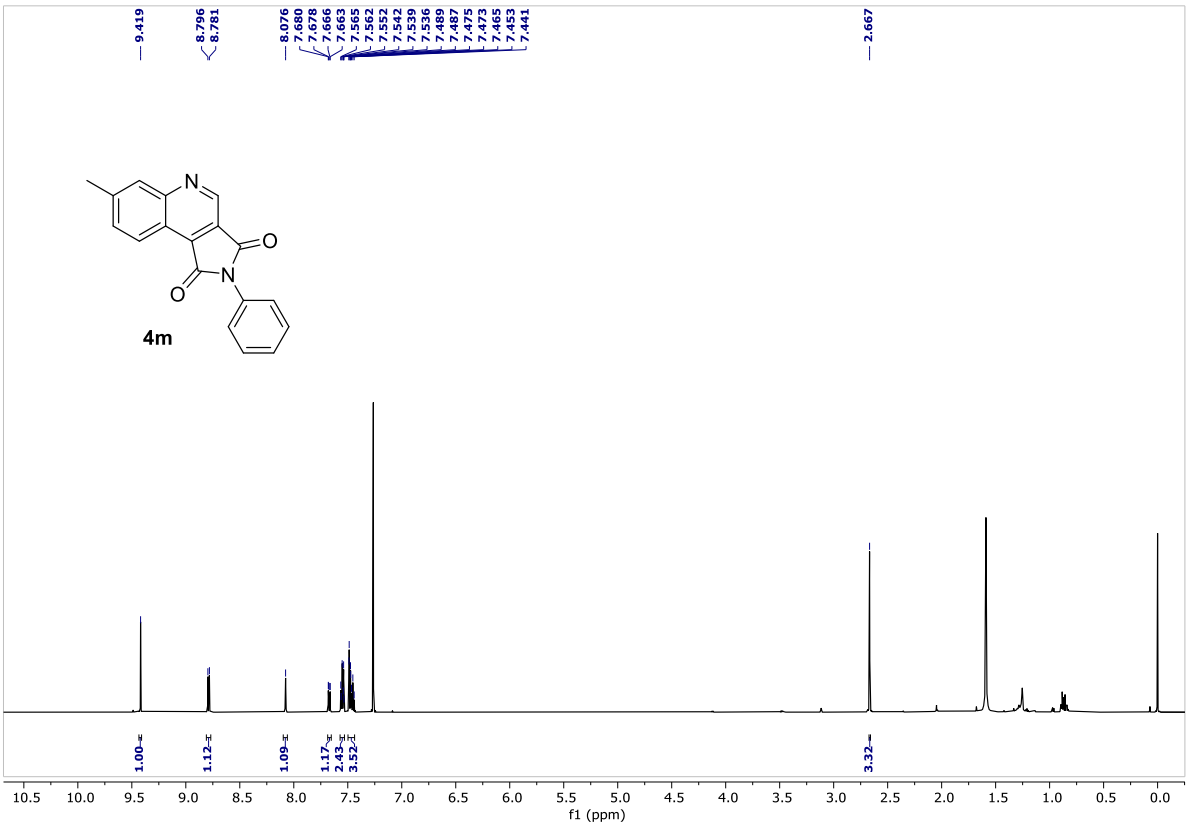


Figure S37: ^{1}H-NMR (600 MHz, $CDCl_3$) of compound **4m**

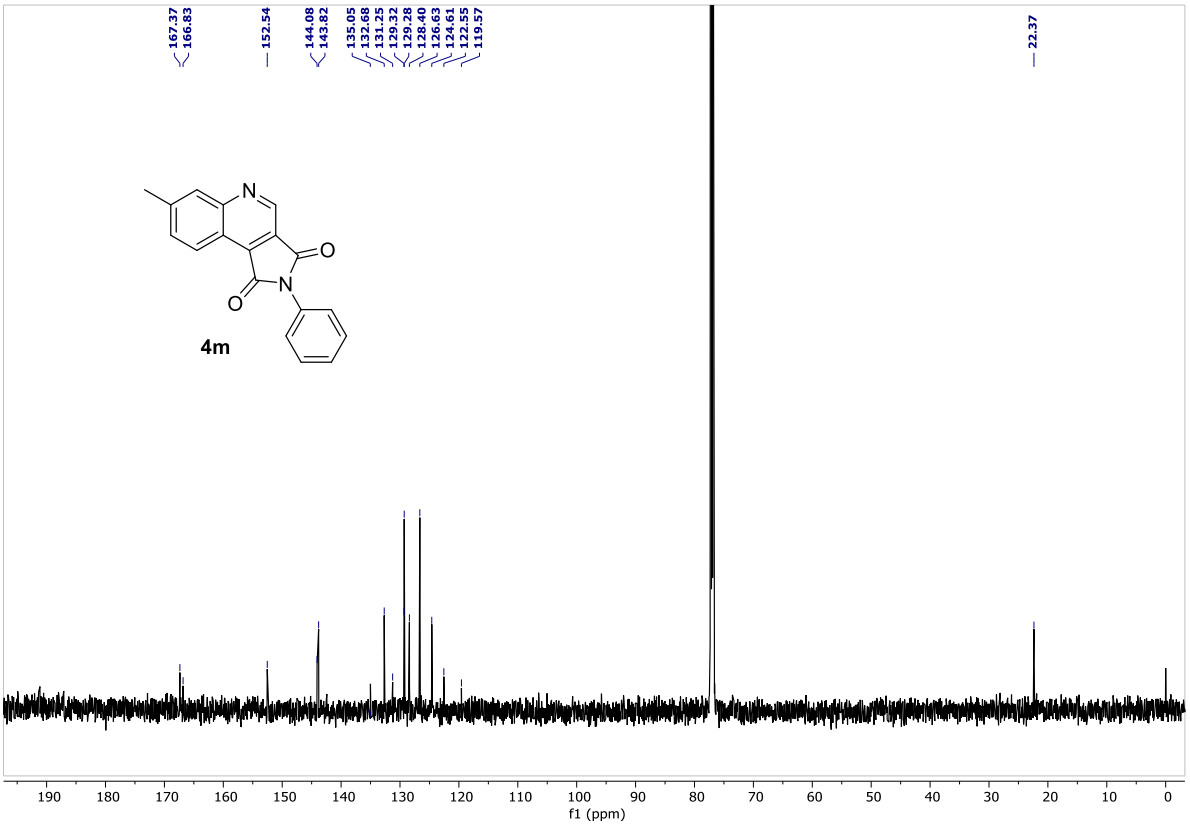


Figure S38: ^{13}C-NMR (150 MHz, $CDCl_3$) of compound **4m**

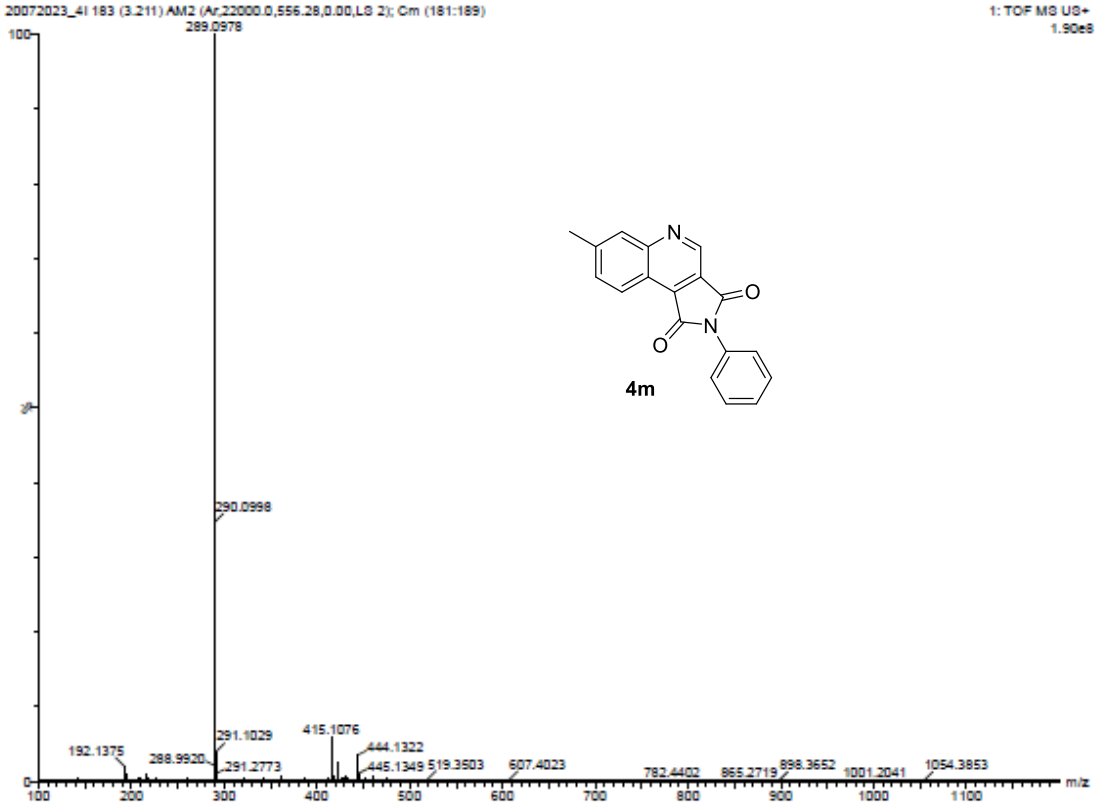


Figure S39: HRMS spectra of compound **4m**

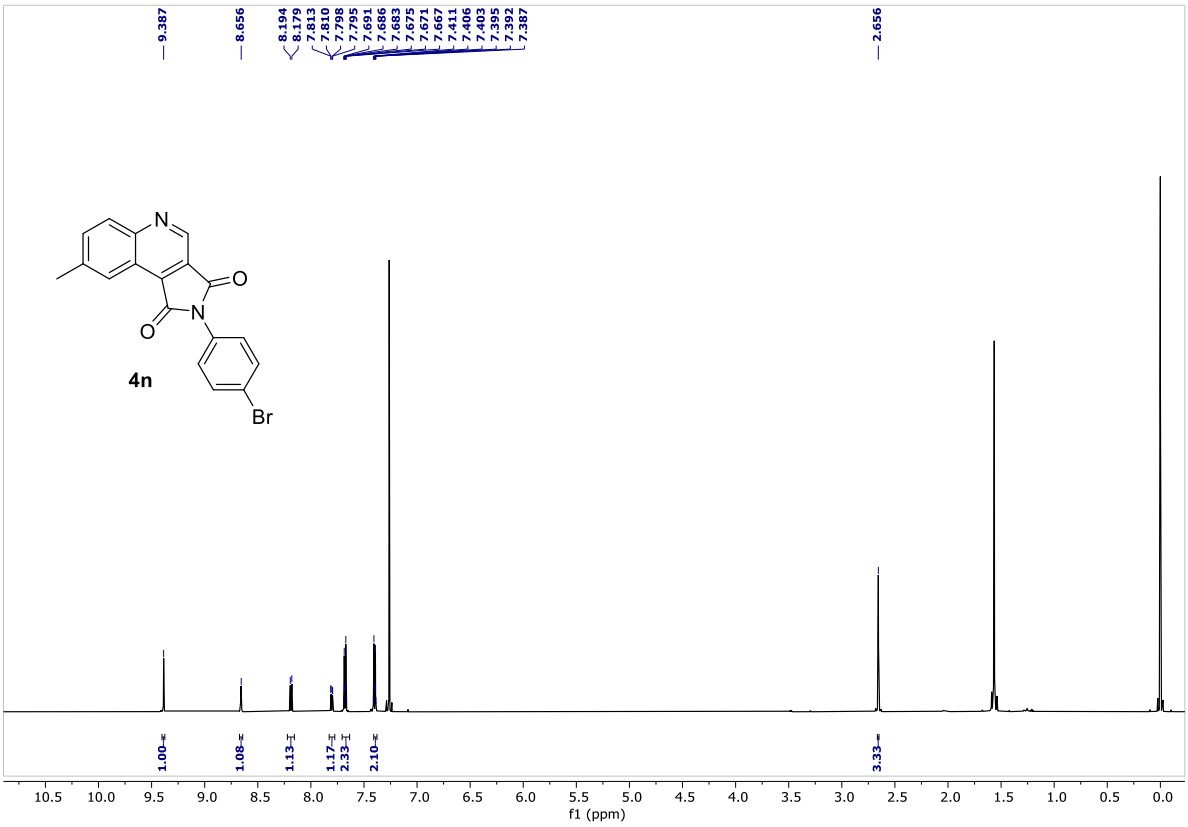


Figure S40: **^{1}H-NMR** (600 MHz, $CDCl_3$) of compound **4n**

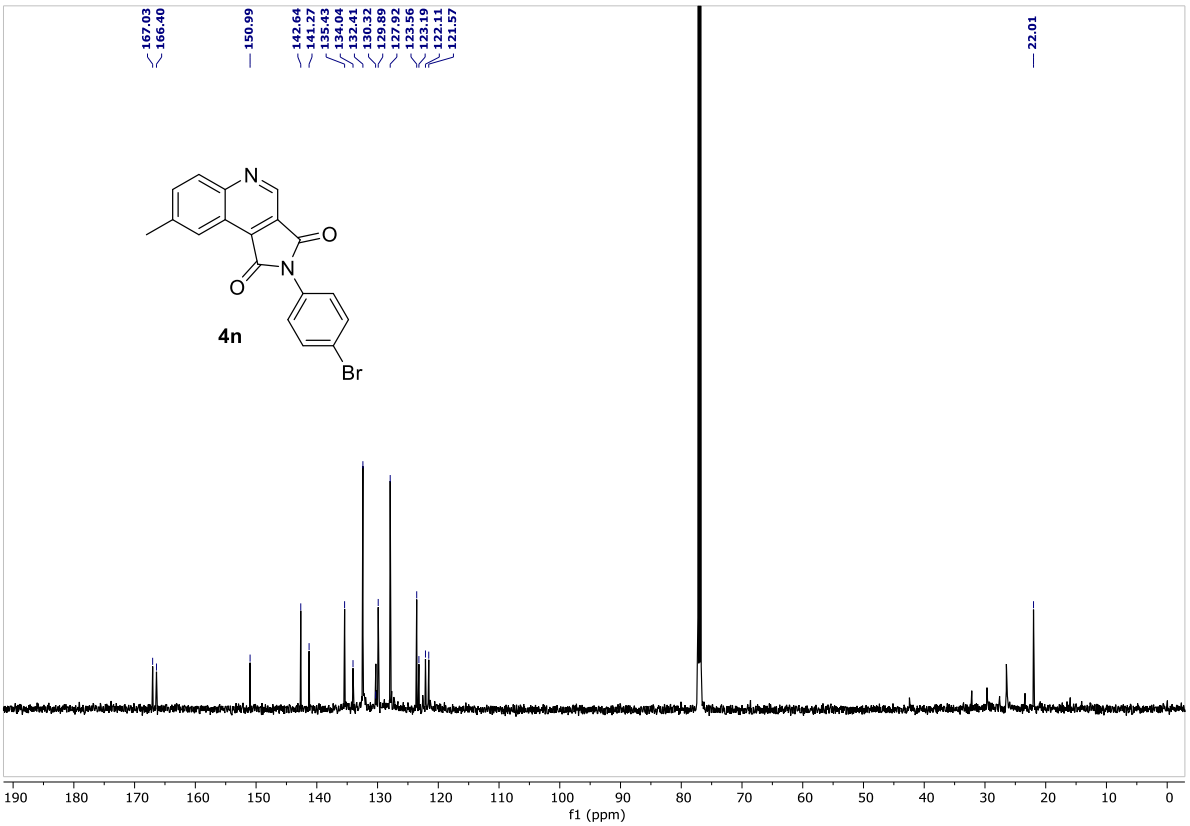


Figure S41: ^{13}**C-NMR** (150 MHz, $CDCl_3$) of compound **4n**

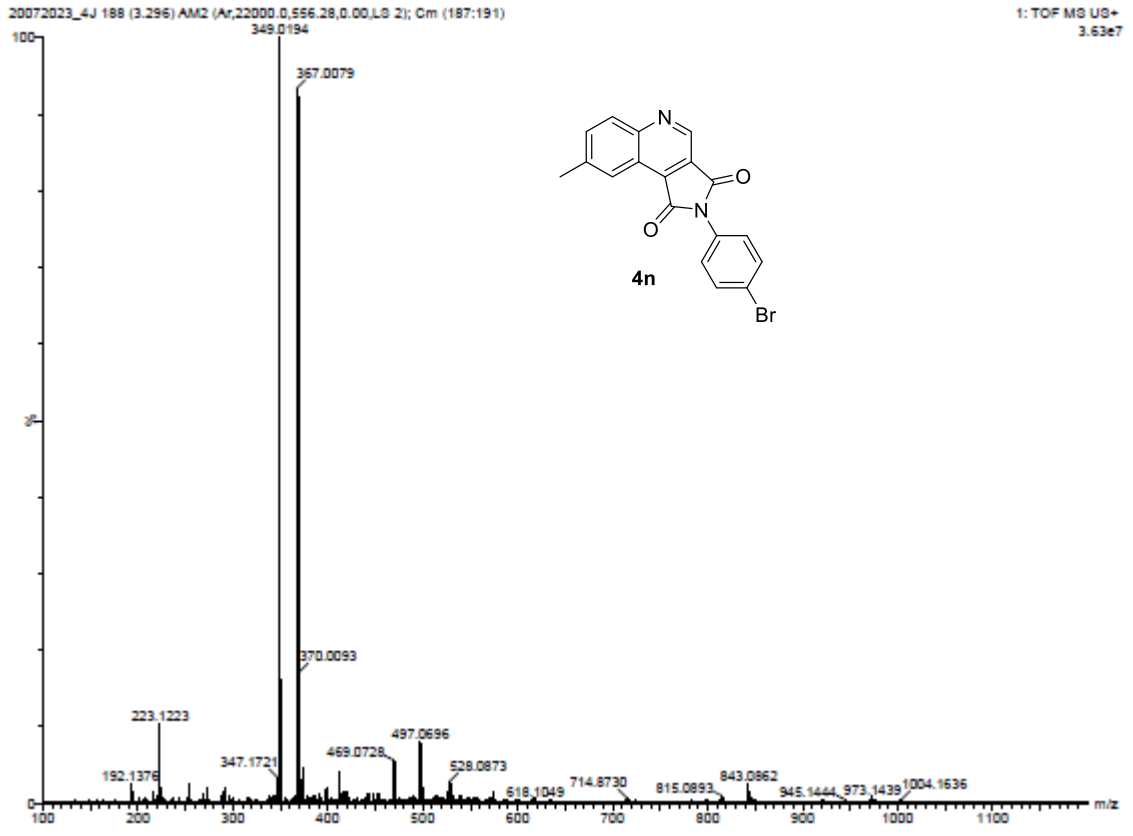


Figure S42: HRMS spectra of compound **4n**

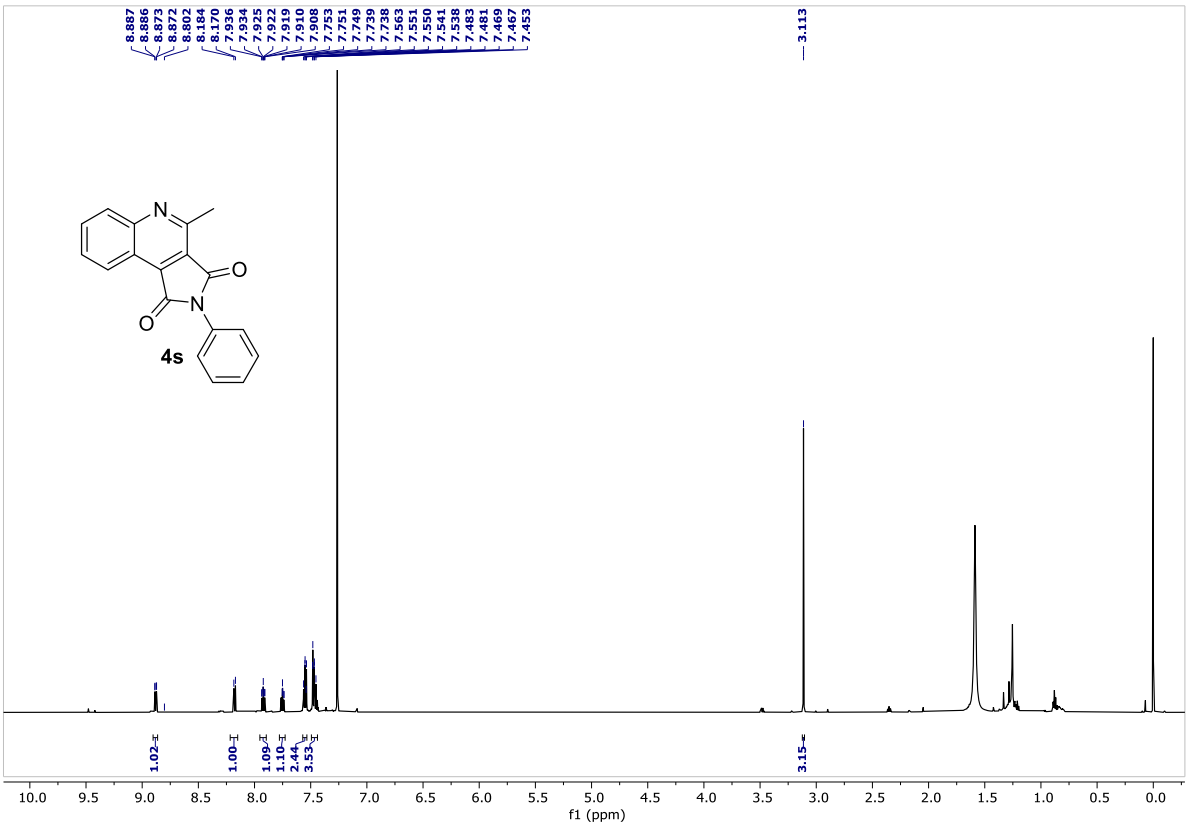


Figure S43: ^{1}H-NMR (600 MHz, $CDCl_3$) of compound **4u**

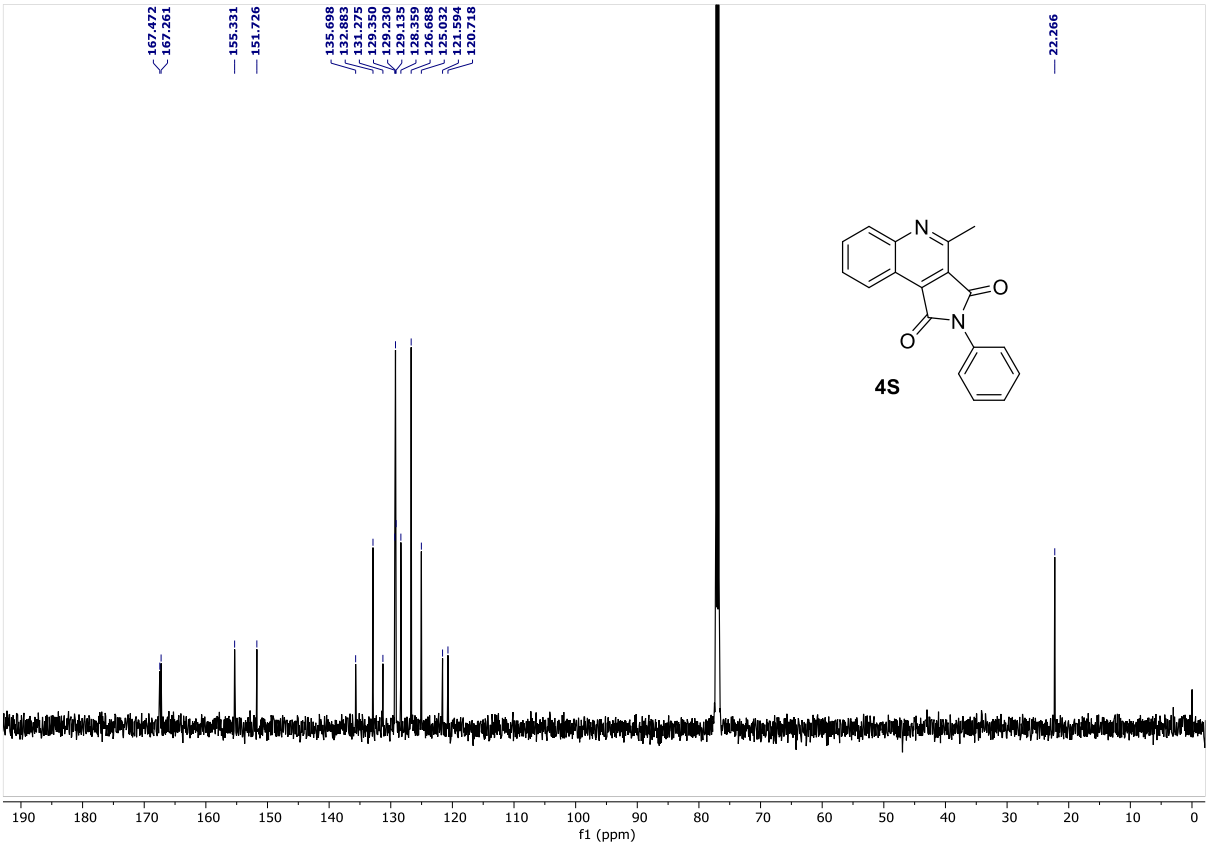


Figure S44: ^{13}C-NMR (150 MHz, $CDCl_3$) of compound **4u**

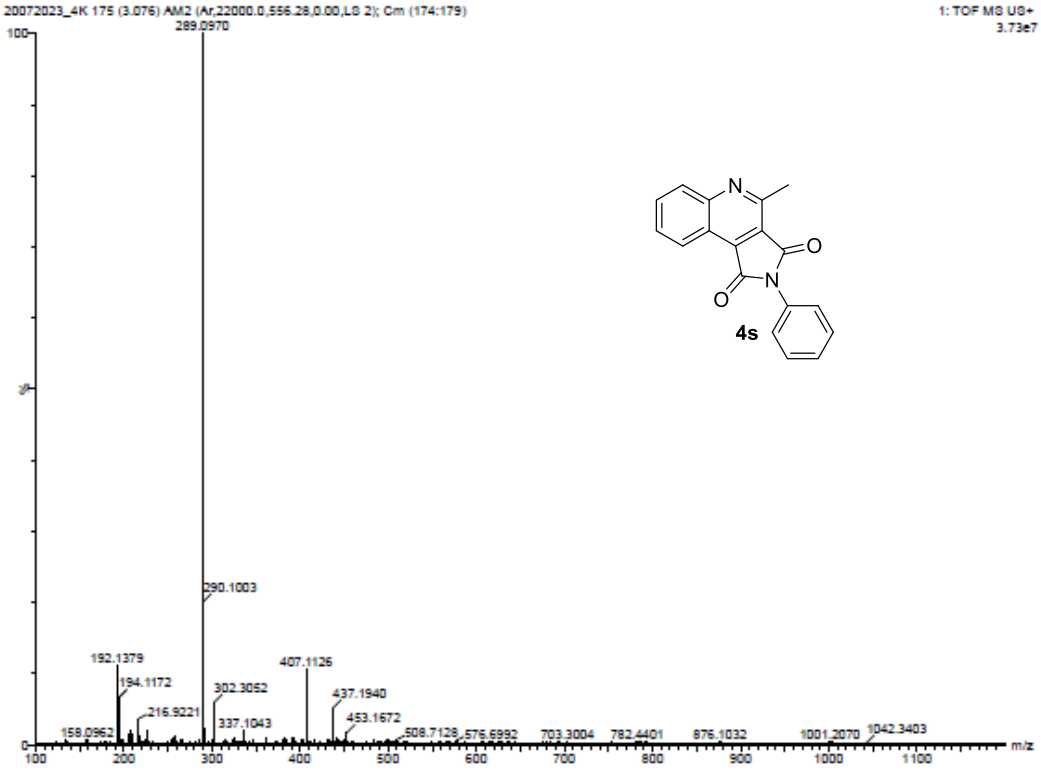


Figure S45: HRMS spectra of compound **4s**

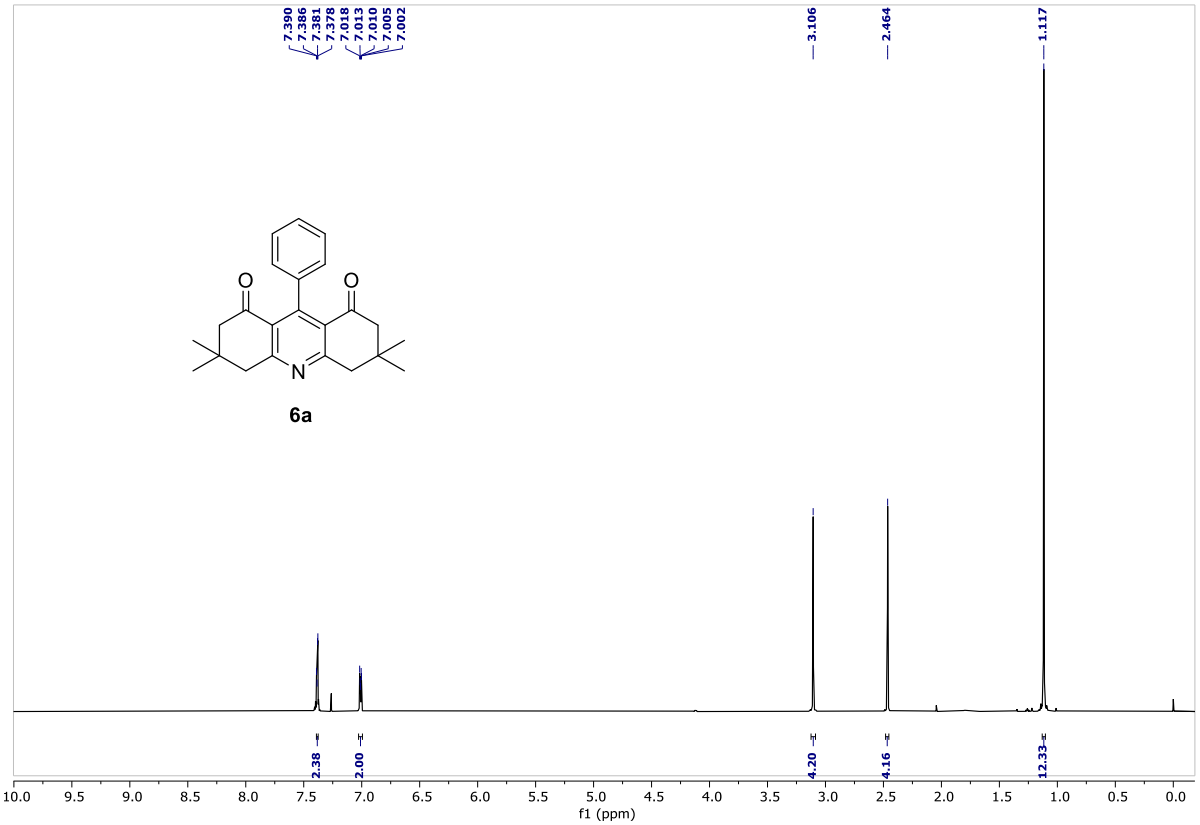


Figure S46: ^{1}H-NMR (600 MHz, $CDCl_3$) of compound **6a**

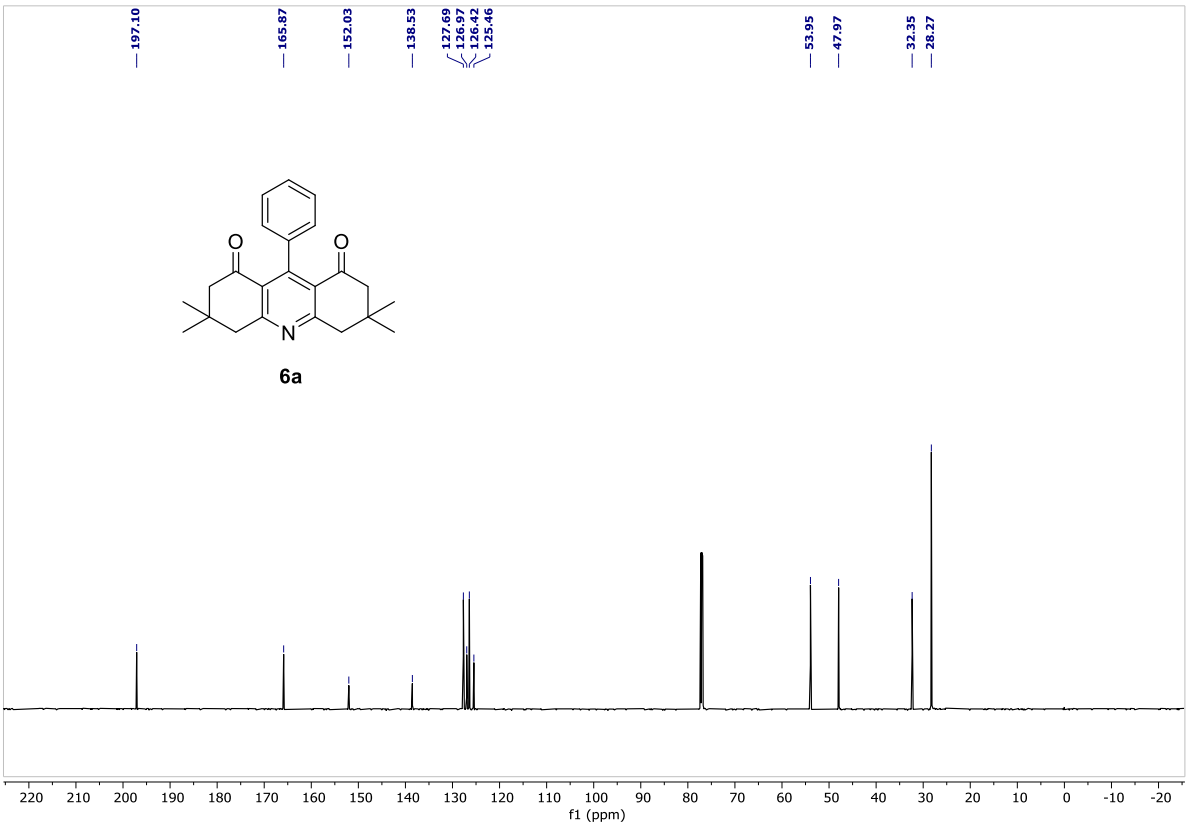


Figure S47: ^{13}**C-NMR** (150 MHz, $CDCl_3$) of compound **6a**

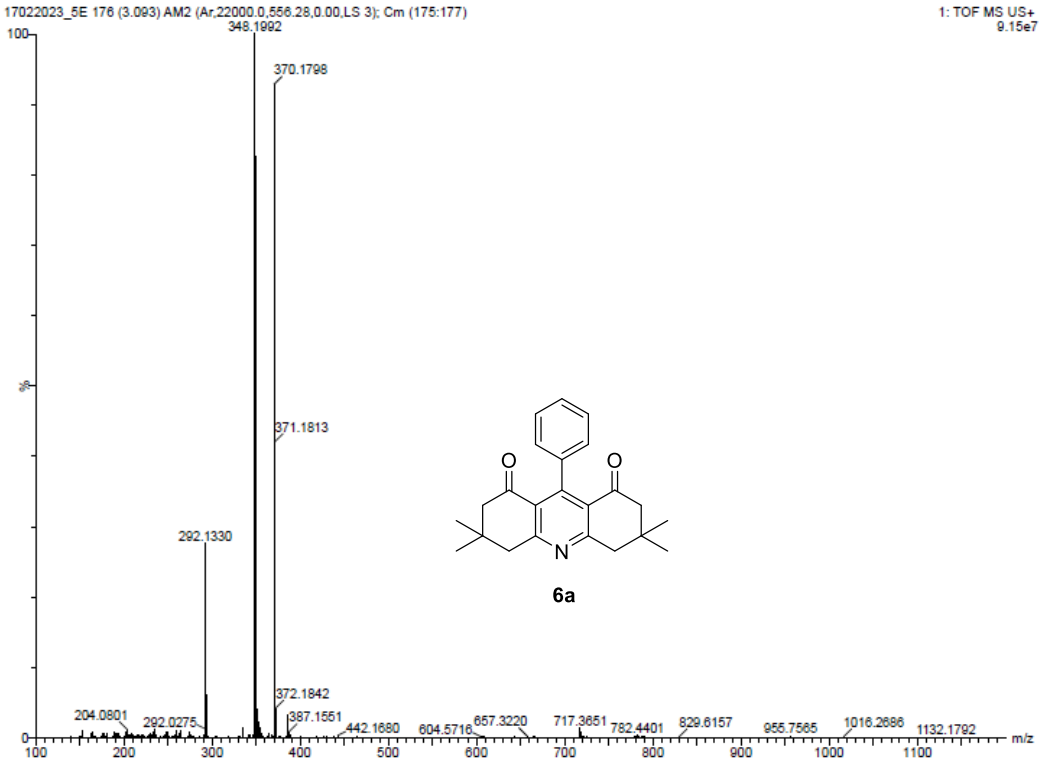


Figure S48: HRMS spectra of compound **6a**

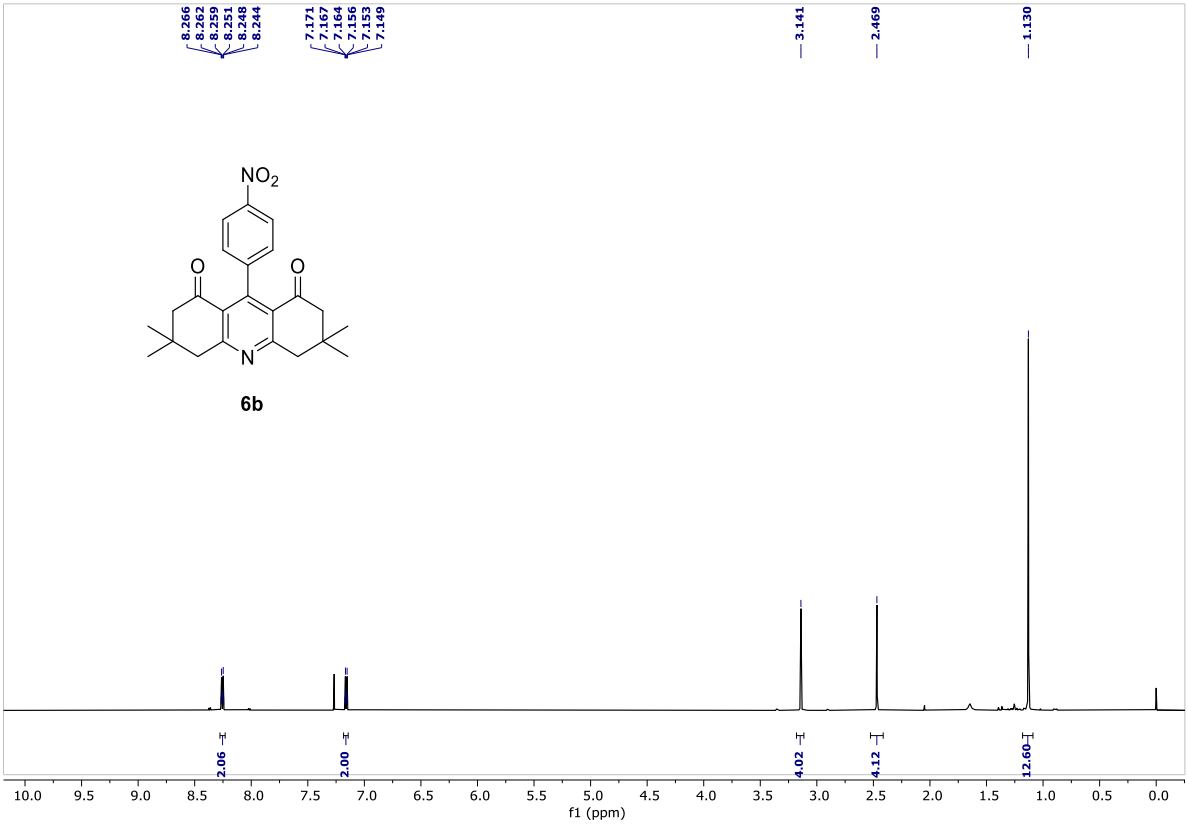


Figure S49: ^{1}H-NMR (600 MHz, $CDCl_3$) of compound **6b**

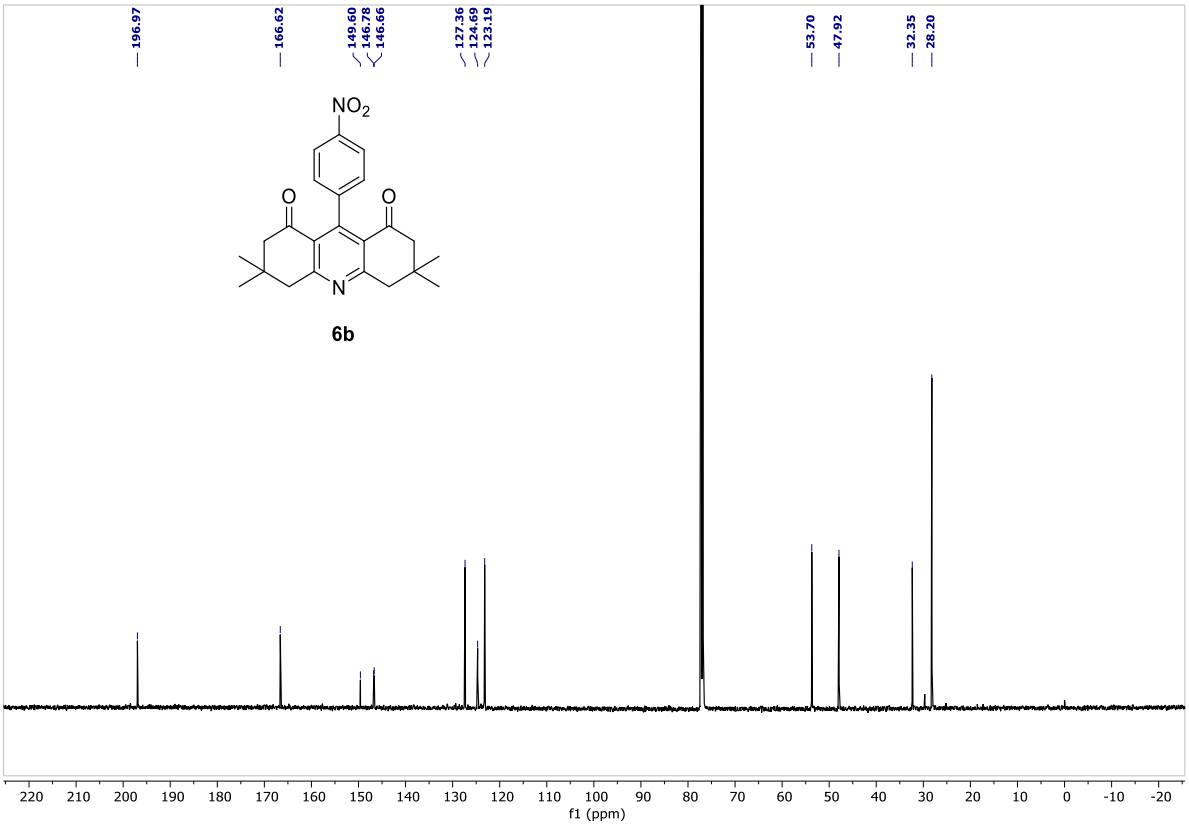


Figure S50: ^{13}C-NMR (150 MHz, $CDCl_3$) of compound **6b**

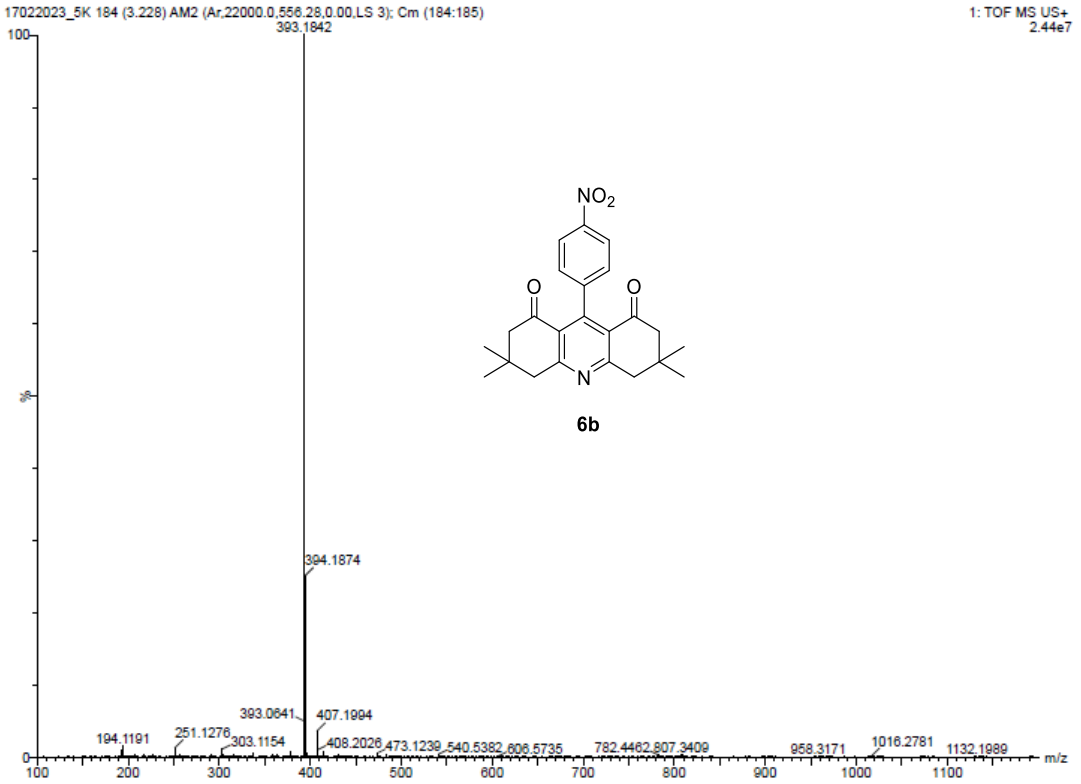


Figure S51: HRMS spectra of compound **6b**

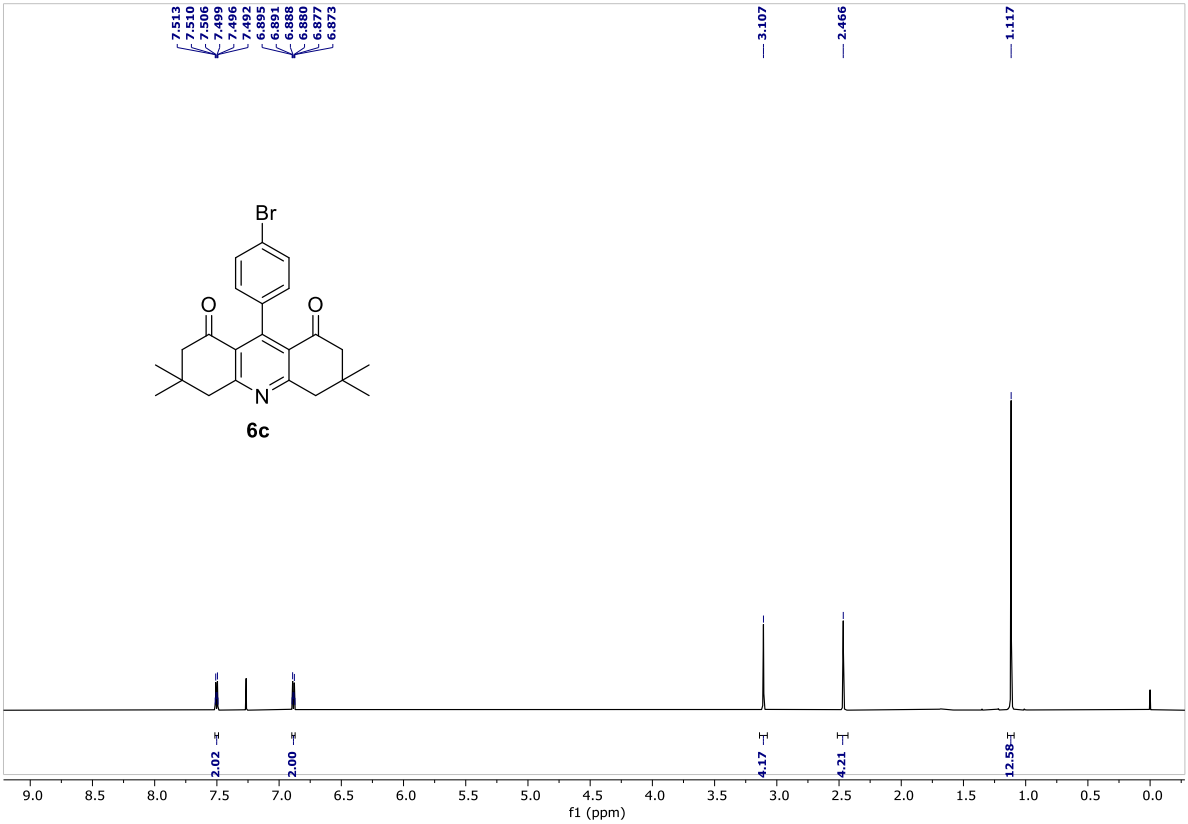


Figure S52: **^{1}H-NMR** (600 MHz, $CDCl_3$) of compound **6c**

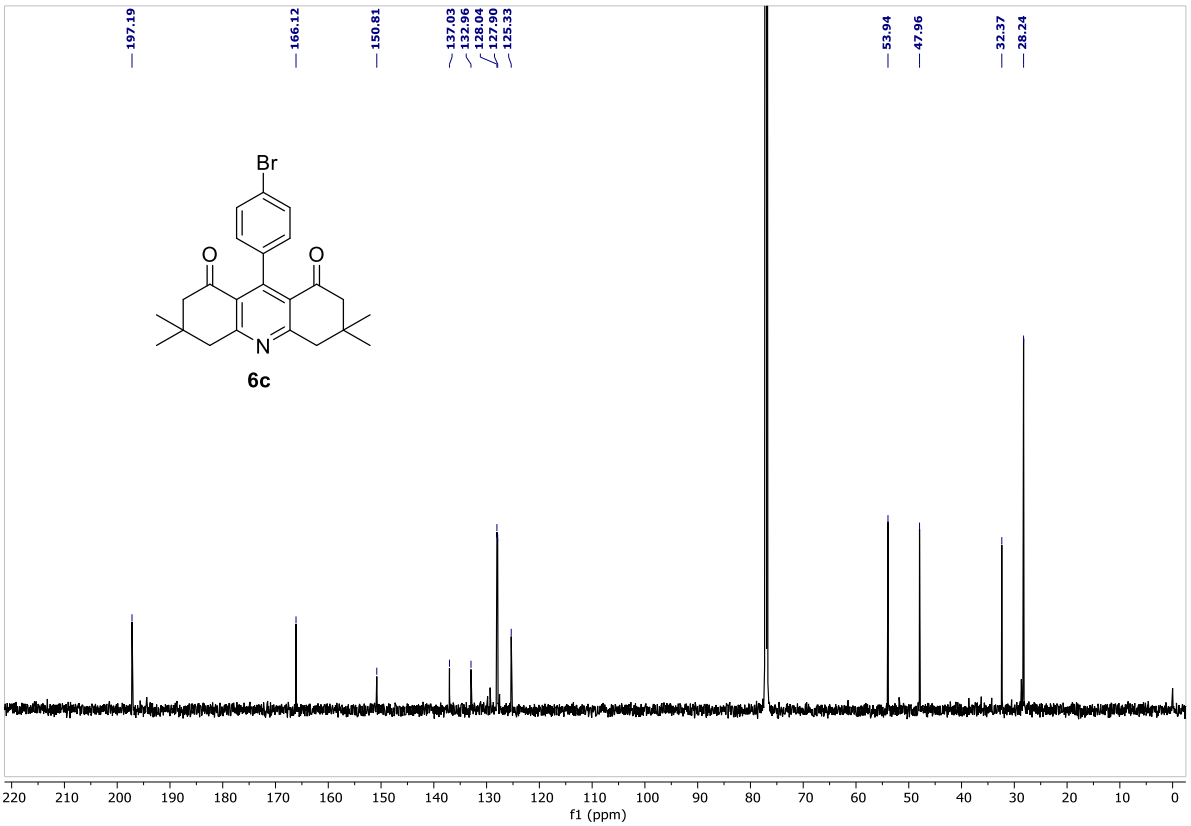


Figure S53: ^{13}**C-NMR** (150 MHz, $CDCl_3$) of compound **6c**

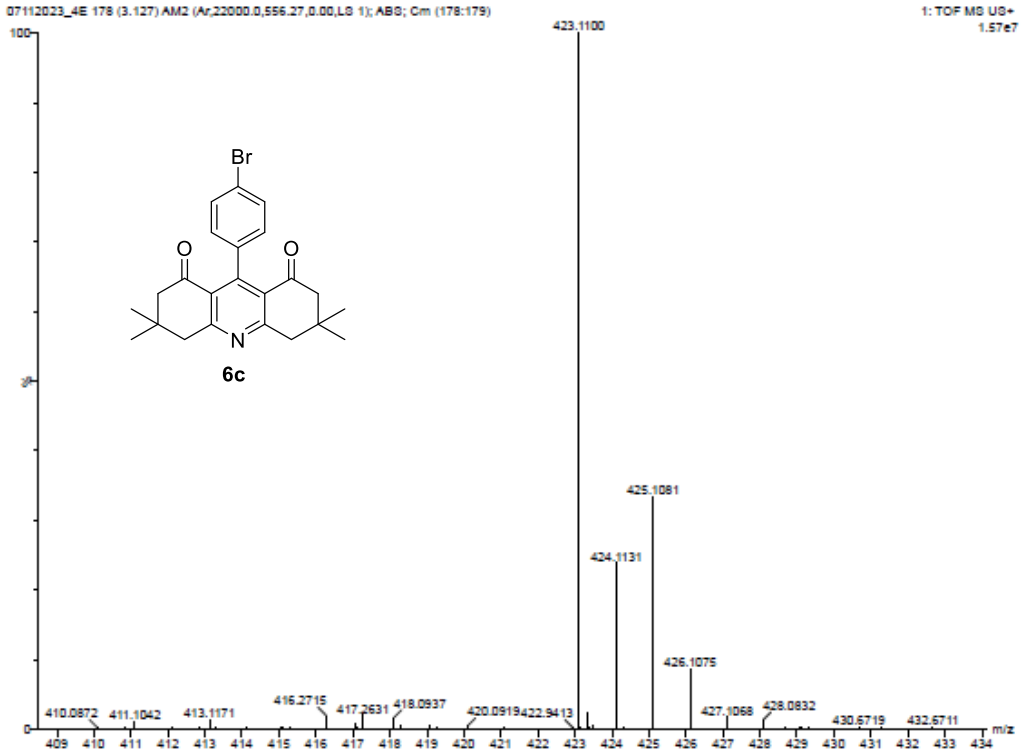


Figure S54: HRMS spectra of compound **6c**

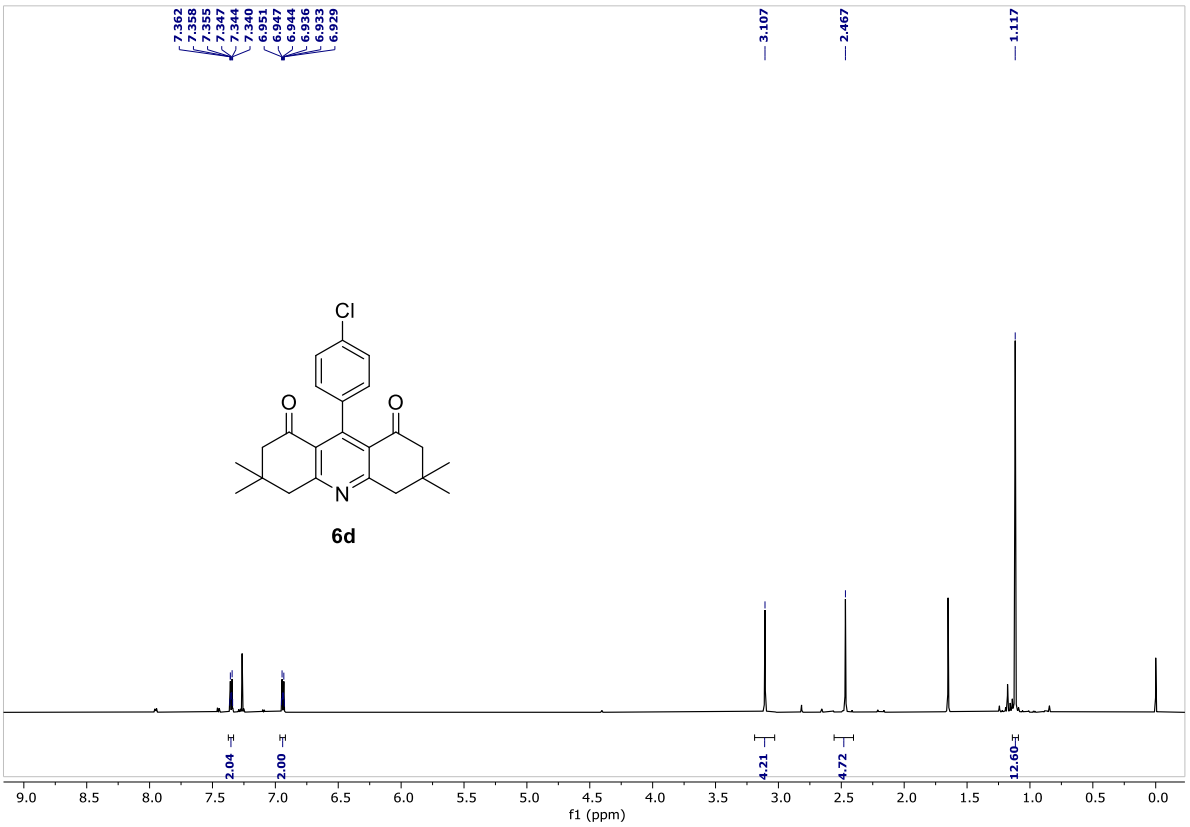


Figure S55: ^{1}H-NMR (600 MHz, $CDCl_3$) of compound **6d**

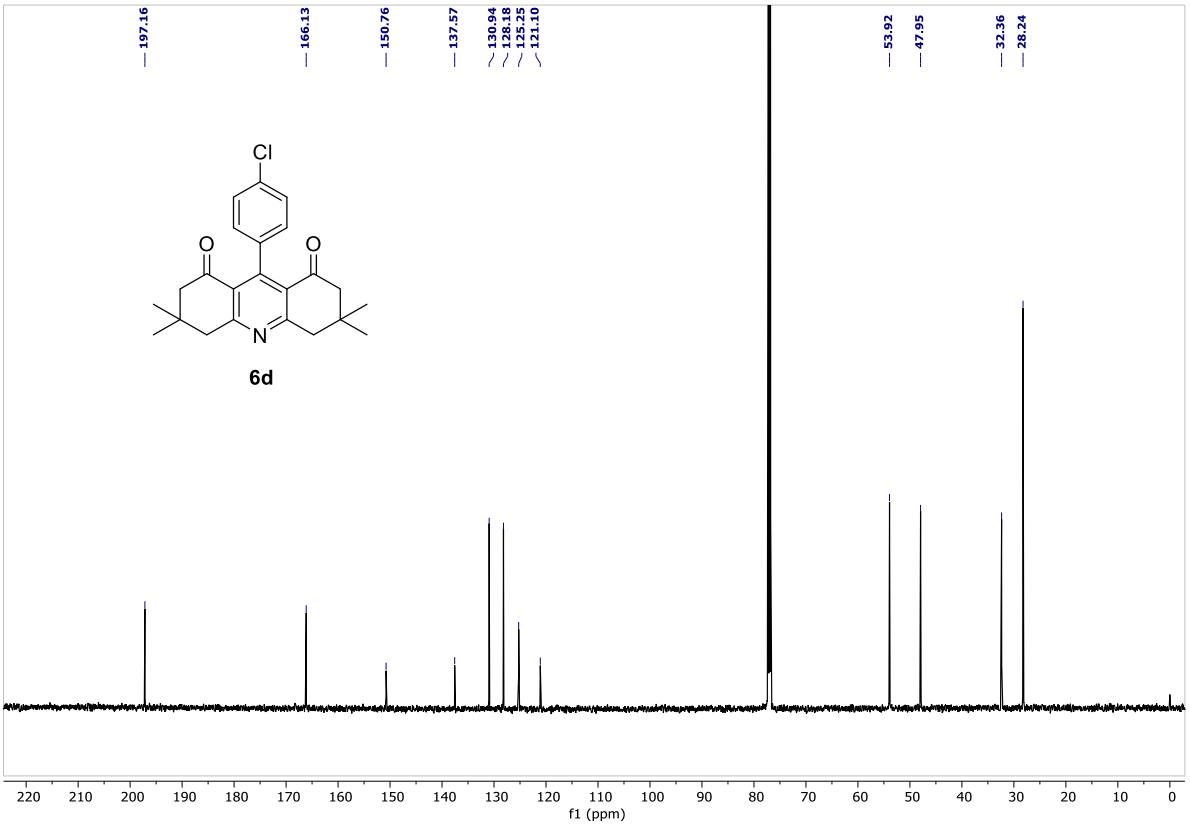


Figure S56: ^{13}C-NMR (150 MHz, $CDCl_3$) of compound **6d**

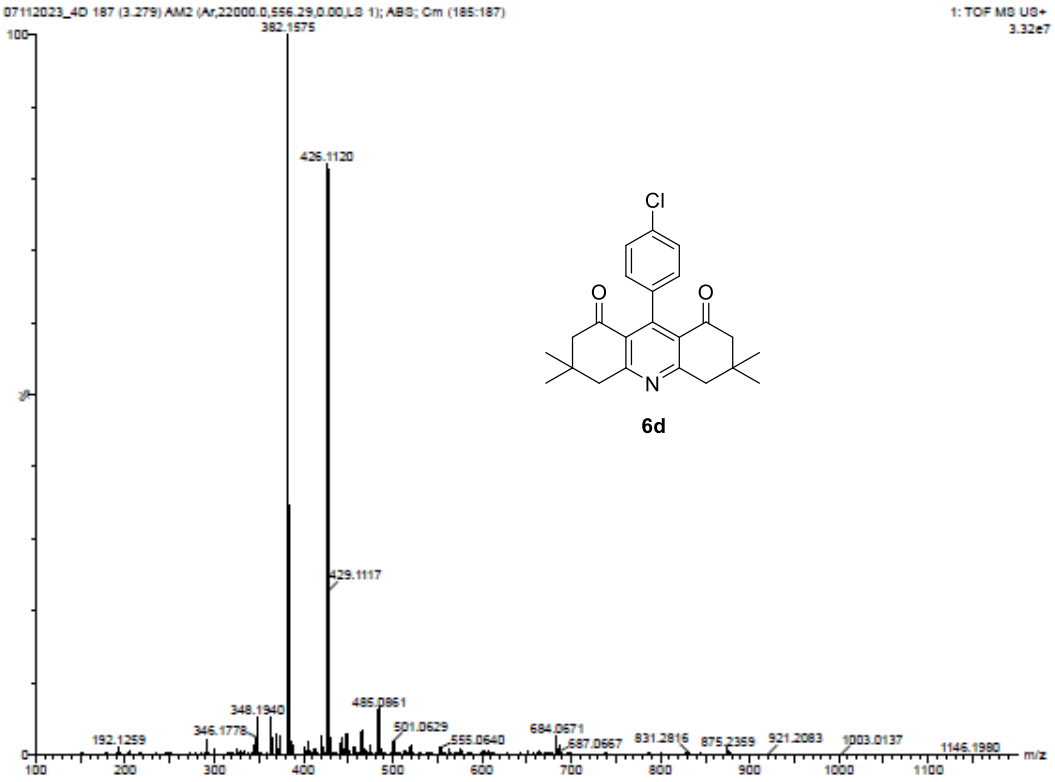


Figure S57: HRMS spectra of compound **6d**

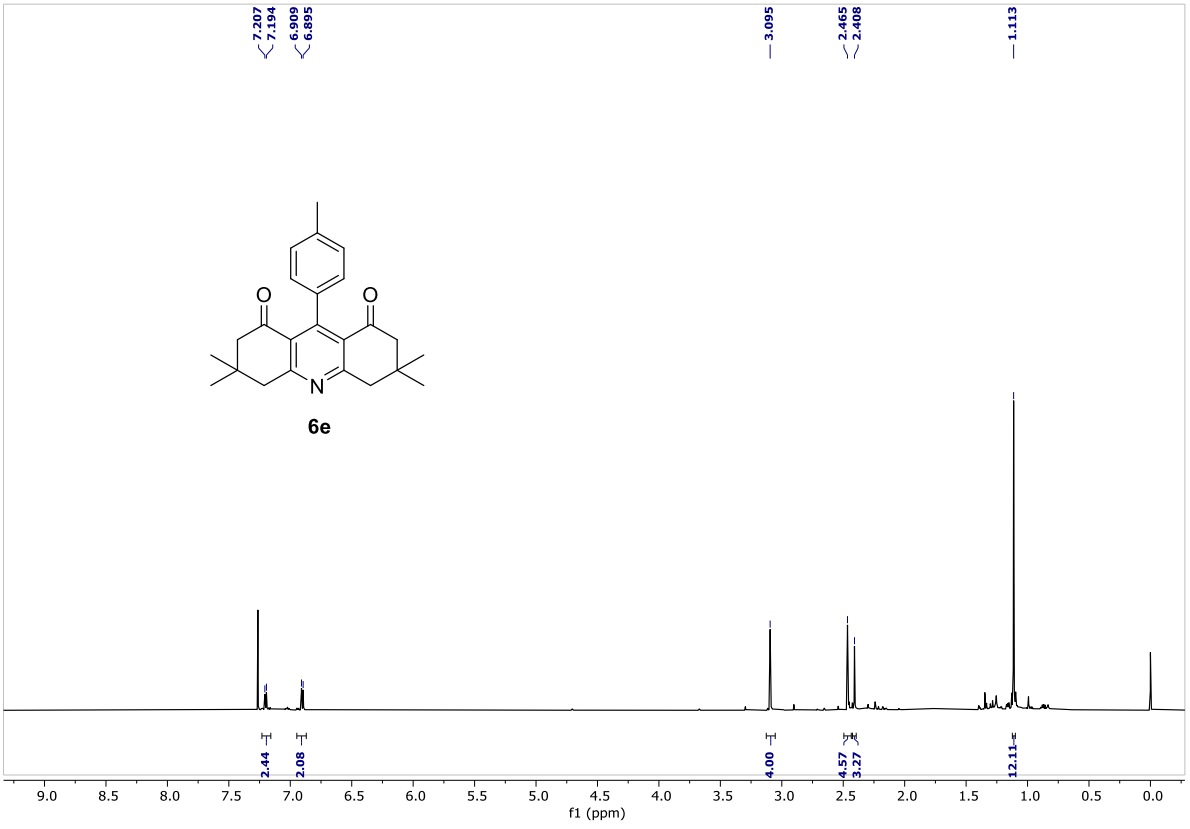


Figure S58: **^{1}H-NMR** (600 MHz, $CDCl_3$) of compound **6e**

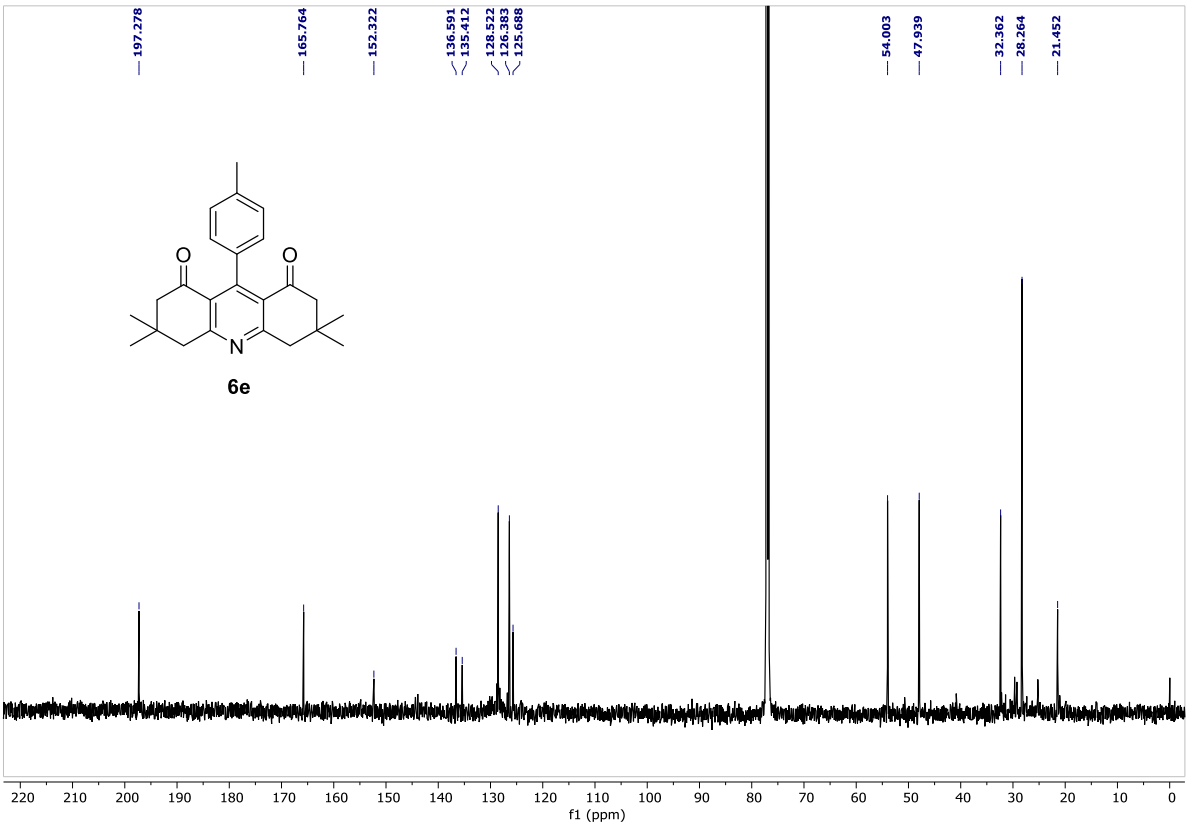


Figure S59: ^{13}**C-NMR** (150 MHz, $CDCl_3$) of compound **6e**

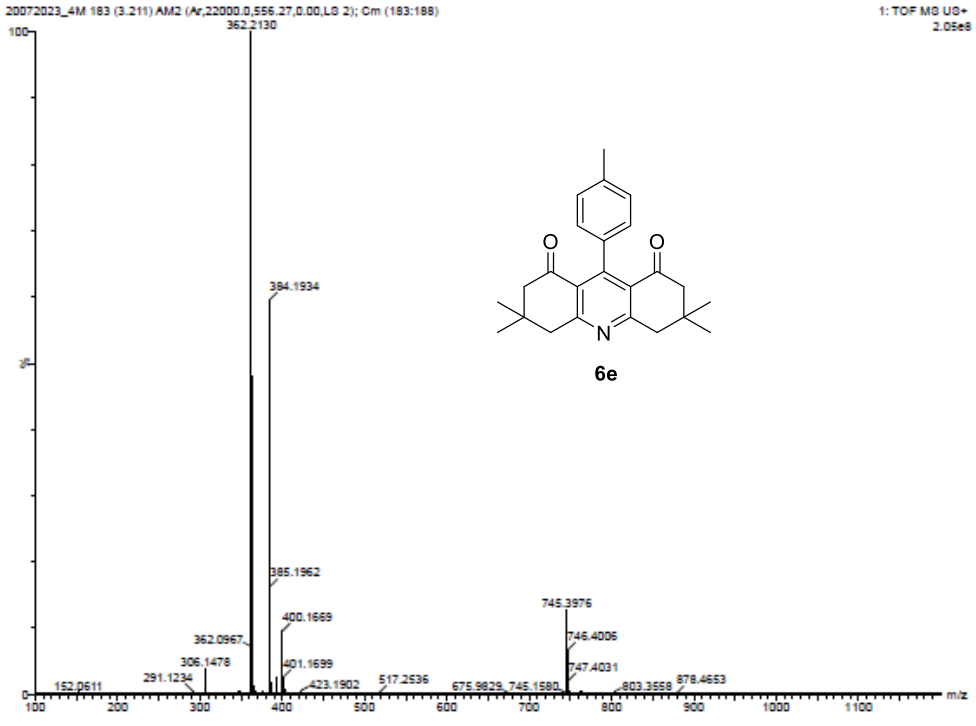


Figure S60: HRMS spectra of compound **6e**

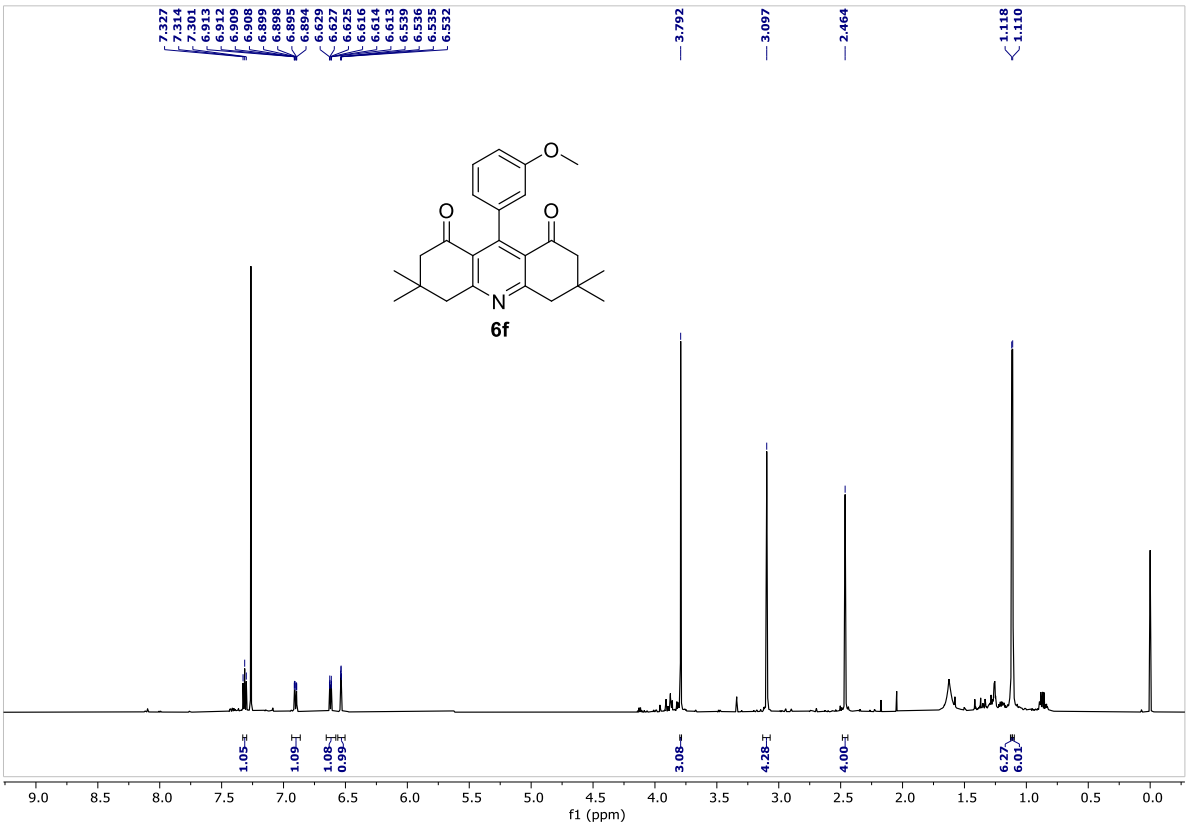


Figure S61: ^{1}H-NMR (600 MHz, $CDCl_3$) of compound **6f**

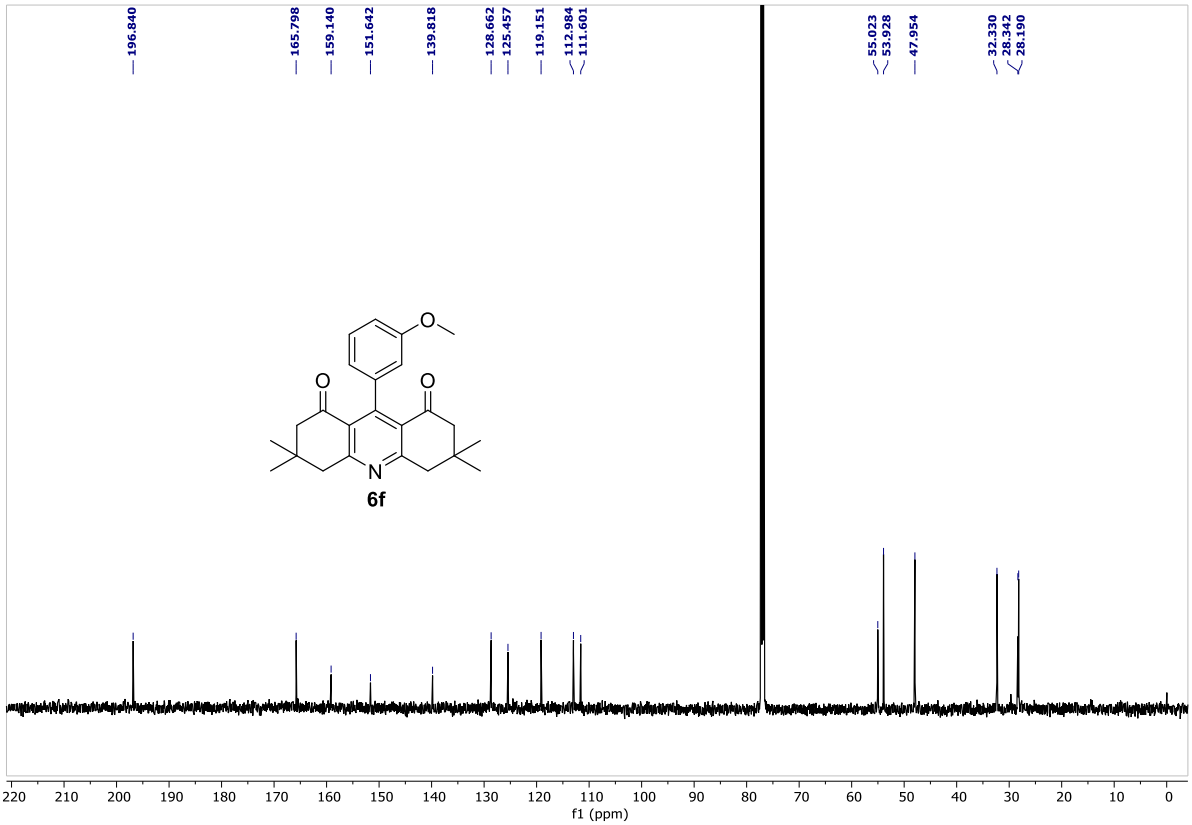


Figure S62: ^{13}C-NMR (150 MHz, $CDCl_3$) of compound **6f**

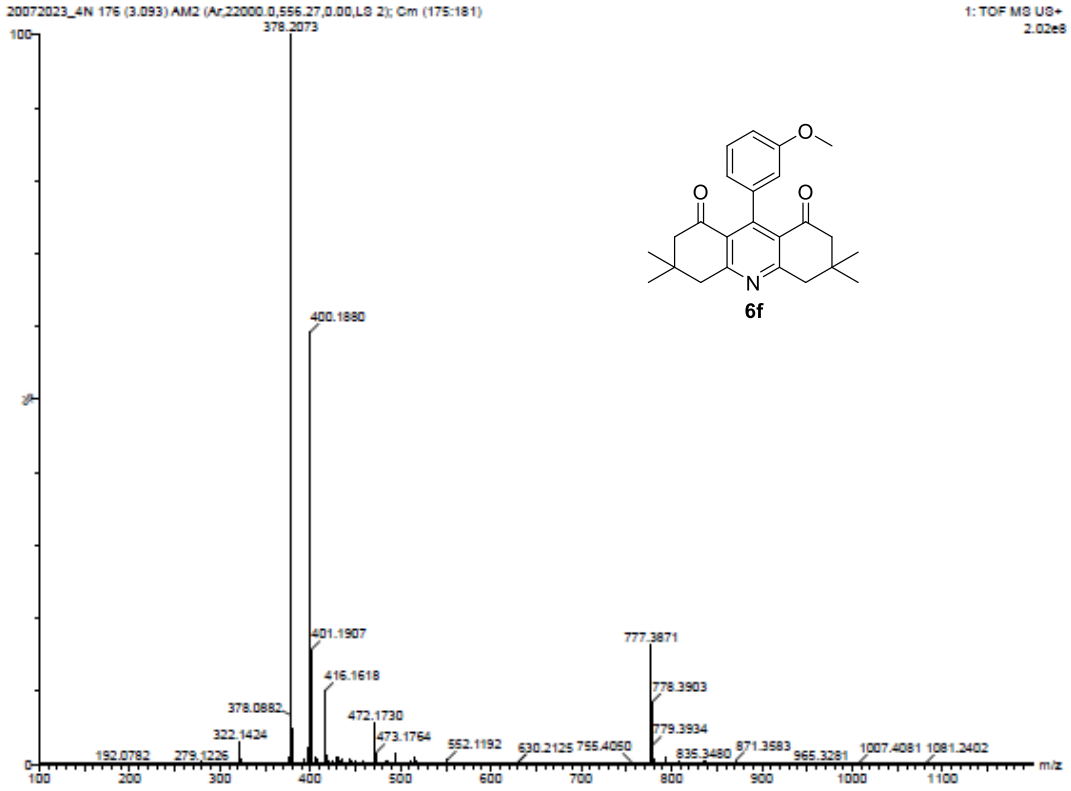


Figure S63: HRMS spectra of compound **6f**

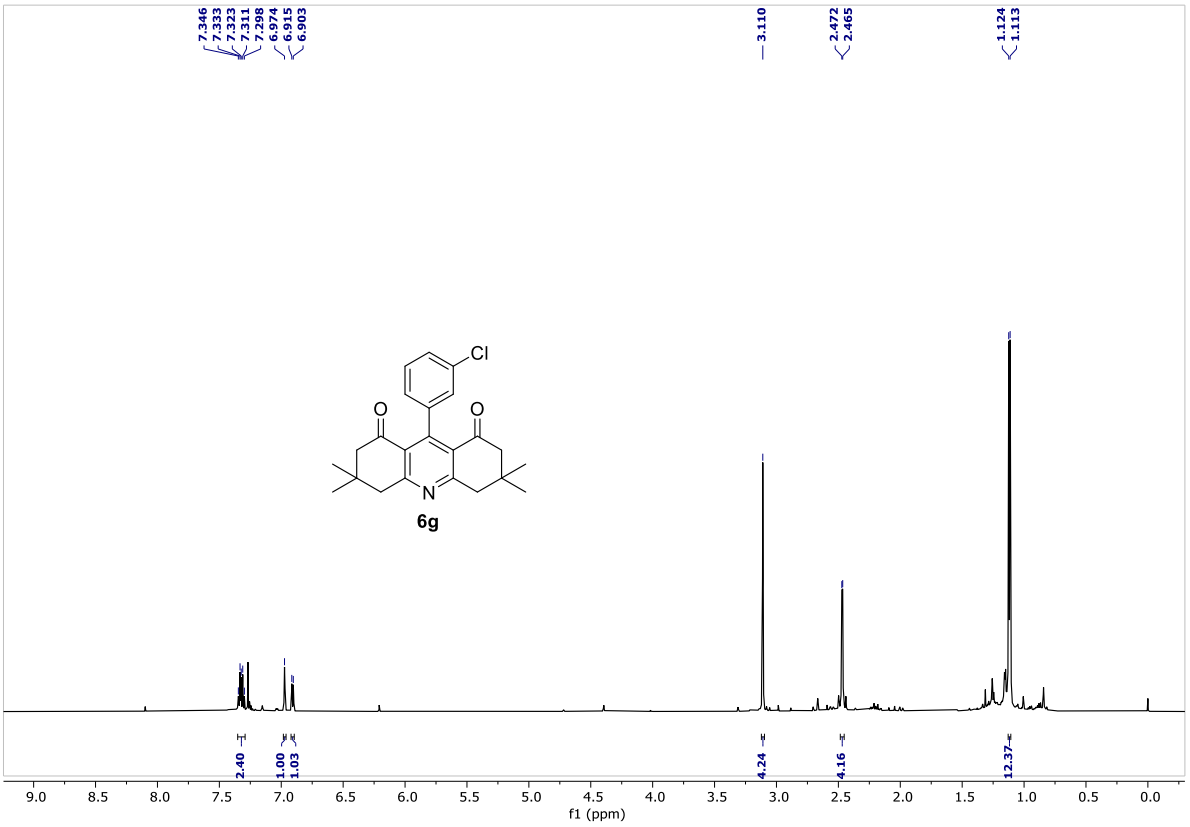


Figure S64: ^{1}H-NMR (600 MHz, $CDCl_3$) of compound **6g**

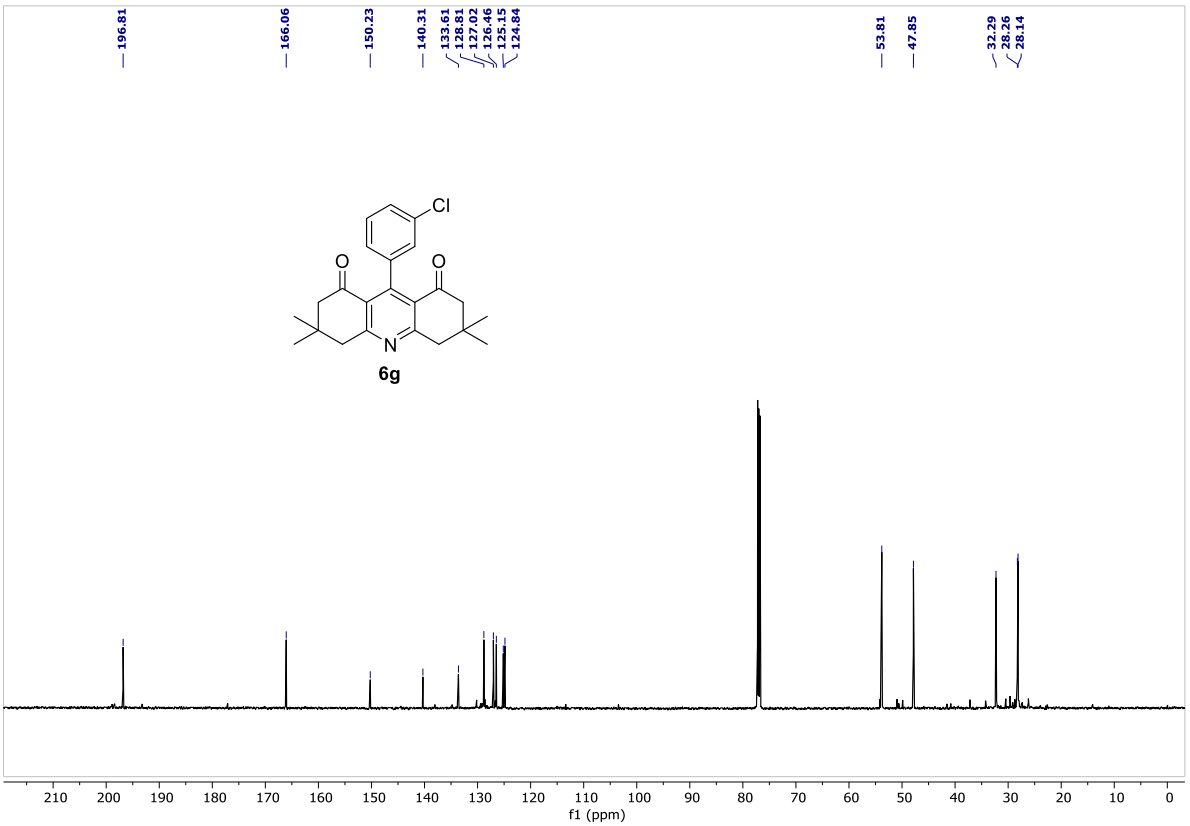


Figure S65: ^{13}**C-NMR** (150 MHz, $CDCl_3$) of compound **6g**

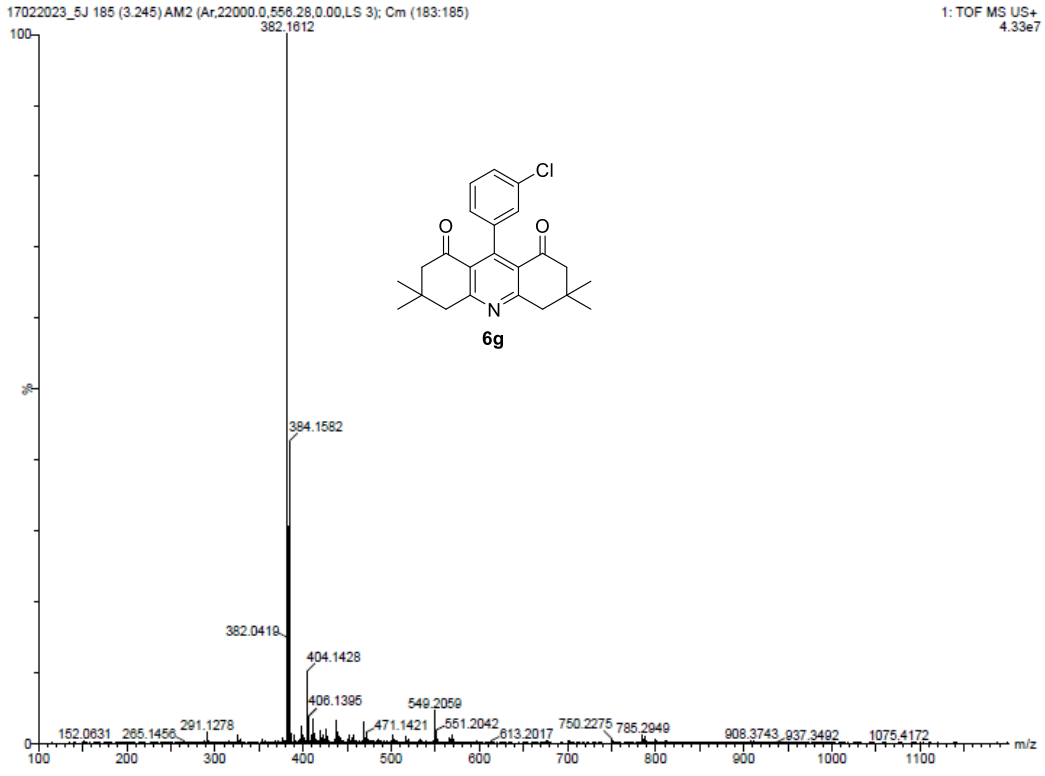


Figure S66: HRMS spectra of compound **6g**

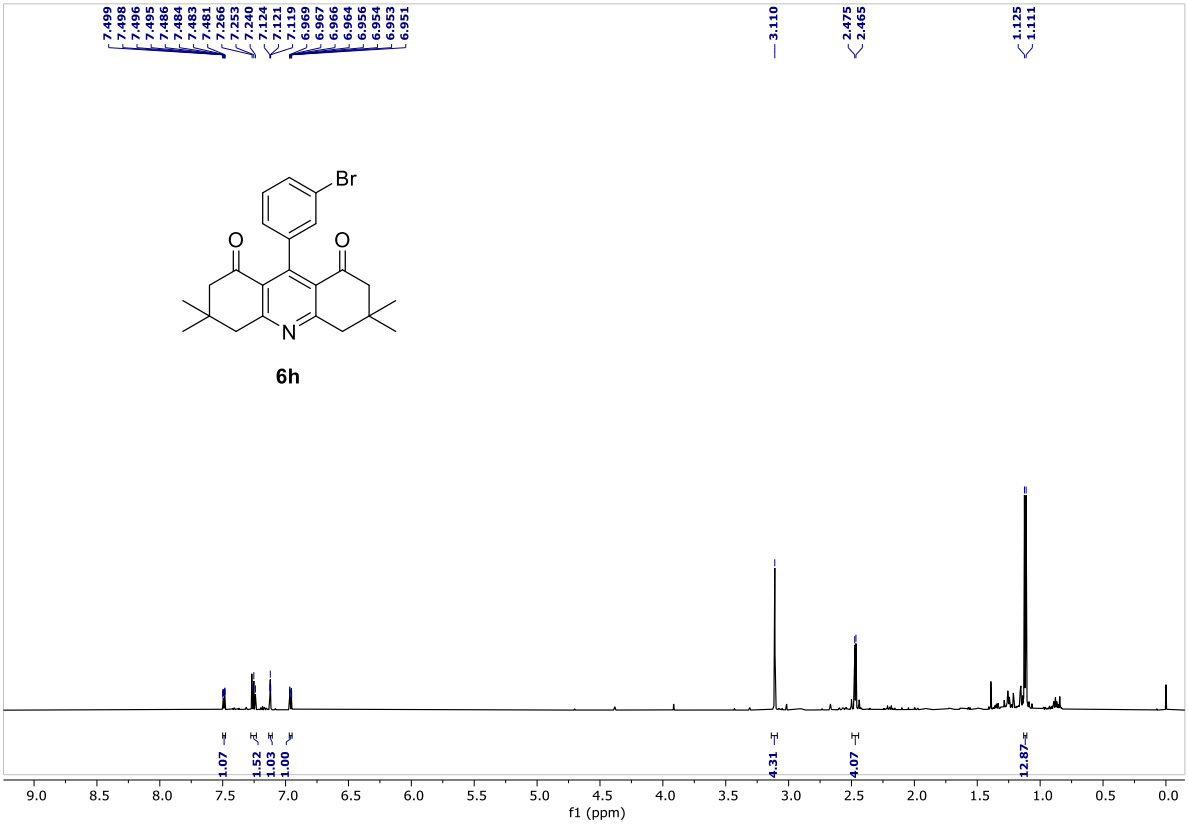


Figure S67: ^{1}H-NMR (600 MHz, $CDCl_3$) of compound **6h**

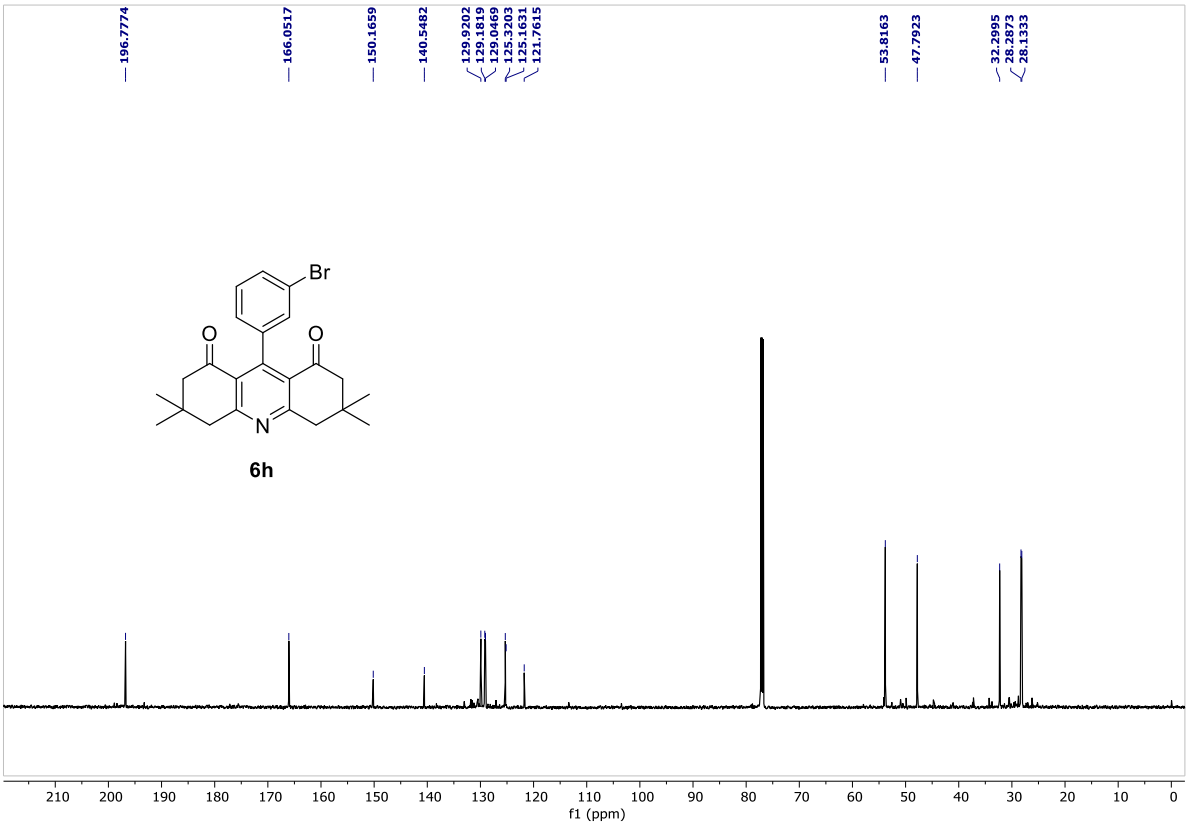


Figure S68: ^{13}C-NMR (150 MHz, $CDCl_3$) of compound **6h**

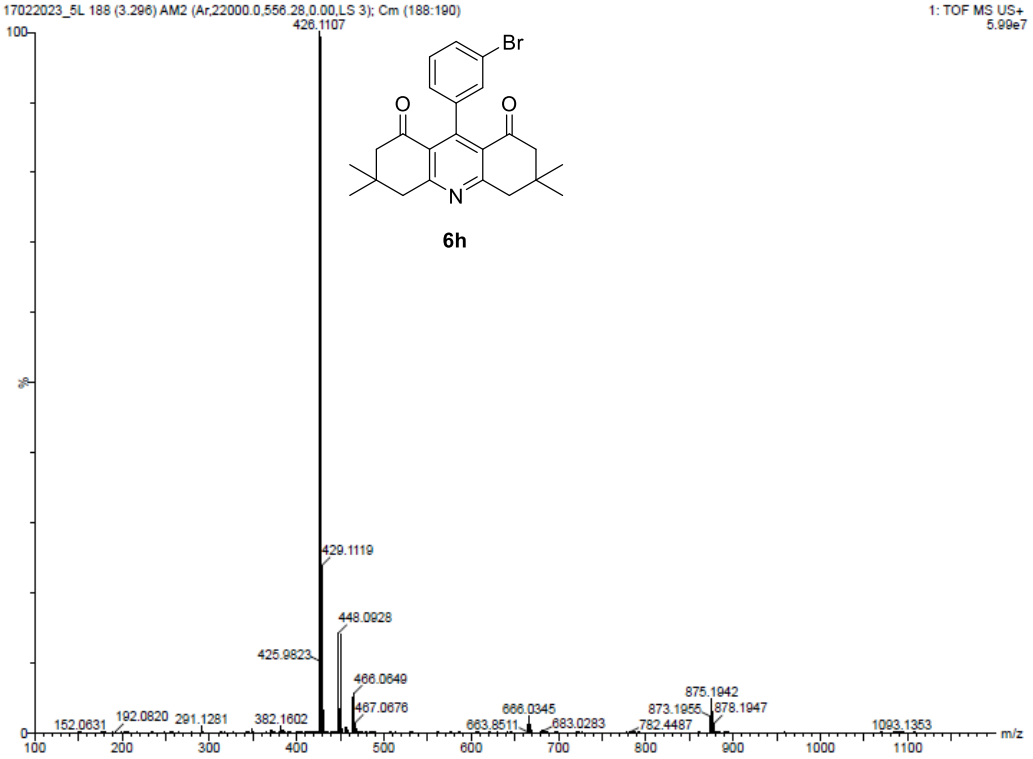


Figure S69: HRMS spectra of compound **6h**

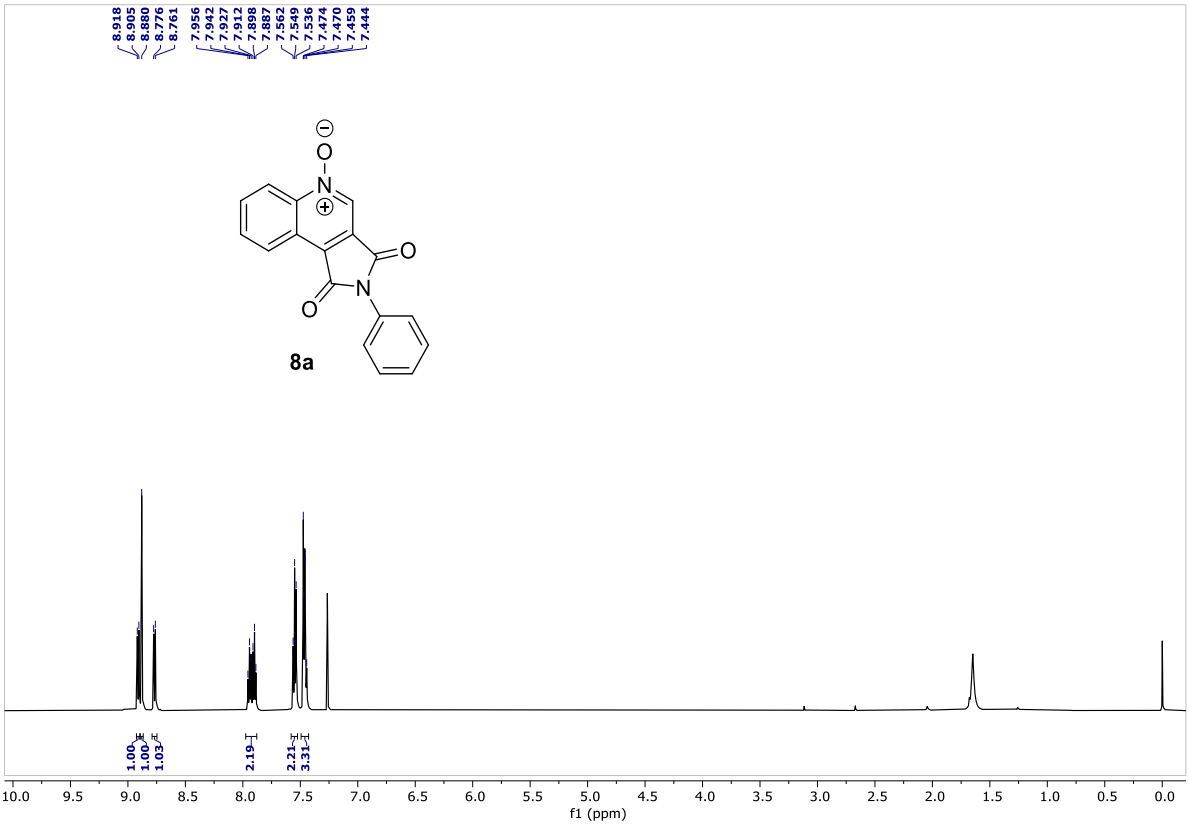


Figure S70: **^{1}H-NMR** (600 MHz, $CDCl_3$) of compound **8a**

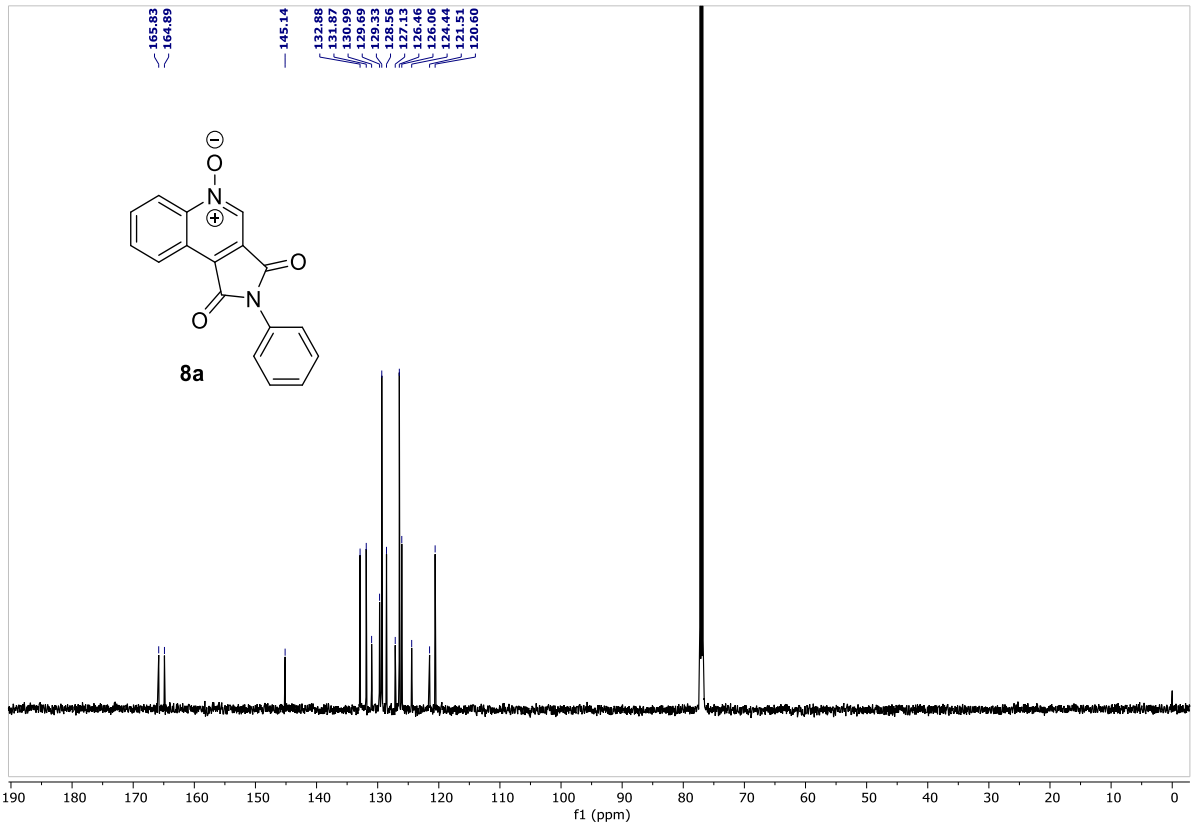


Figure S71: ^{13}C-NMR (150 MHz, $CDCl_3$) of compound **8a**

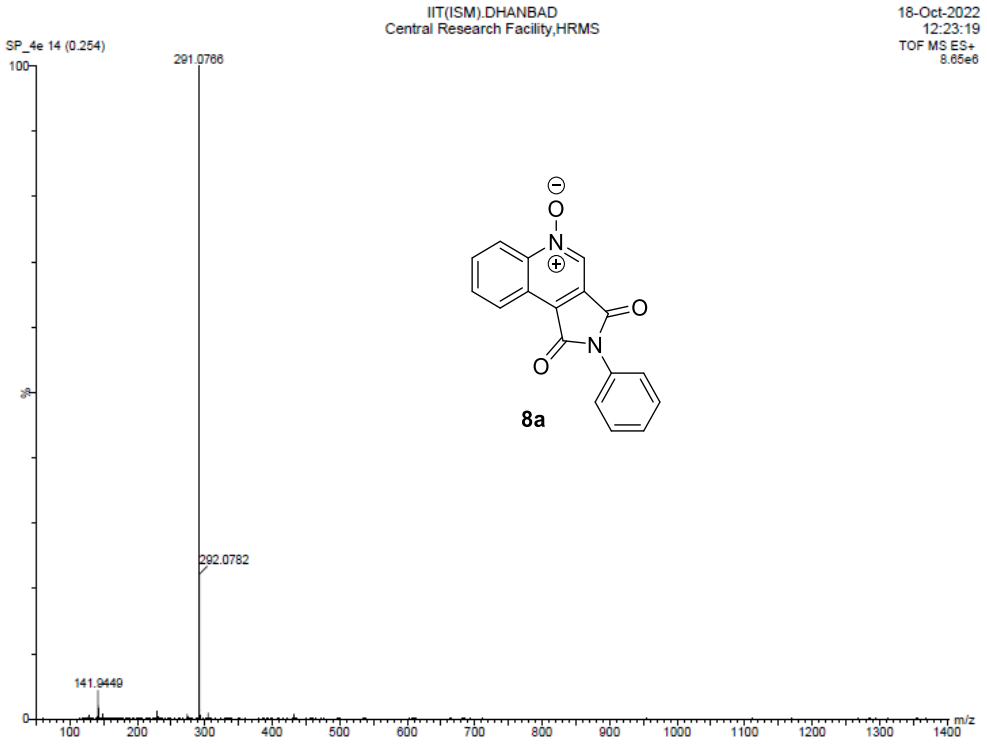


Figure S72: HRMS spectra of compound **8a**

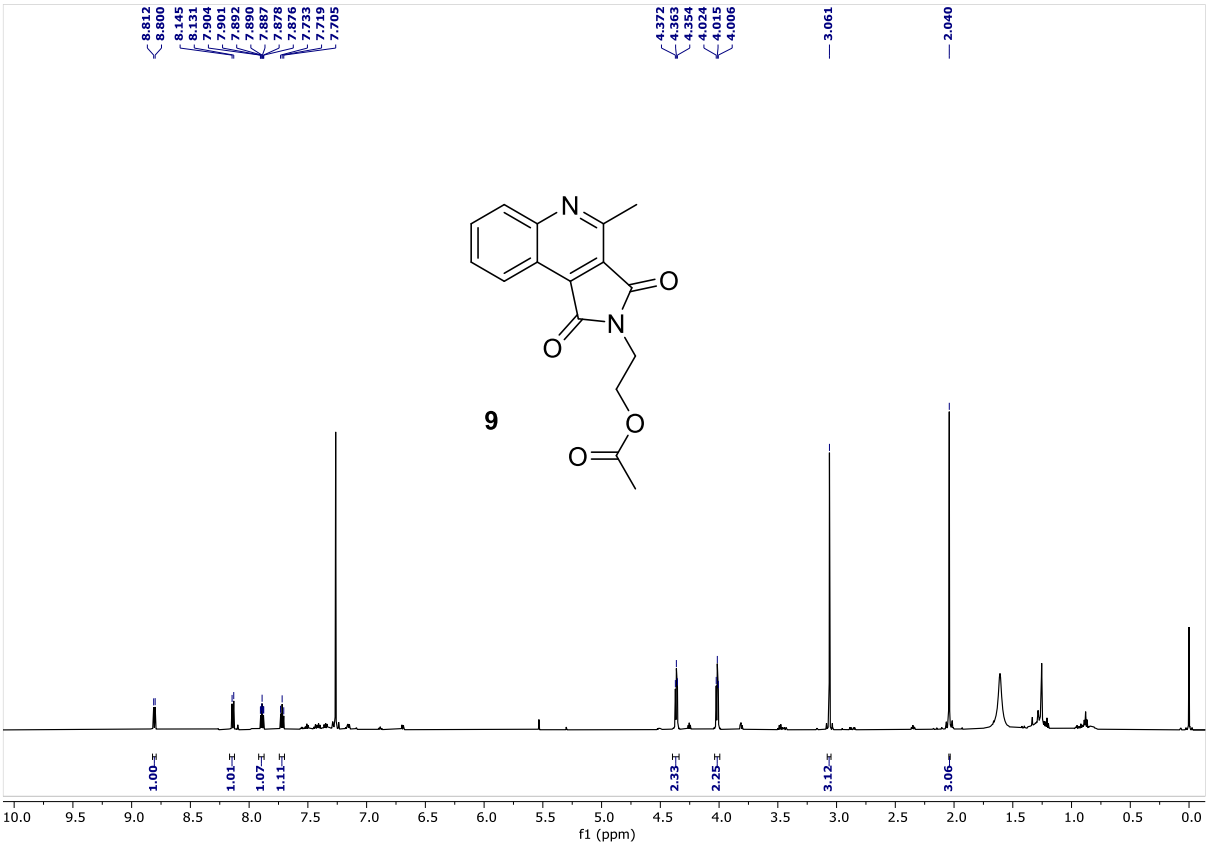


Figure S73: ^{1}H-NMR (600 MHz, $CDCl_3$) of compound **9**

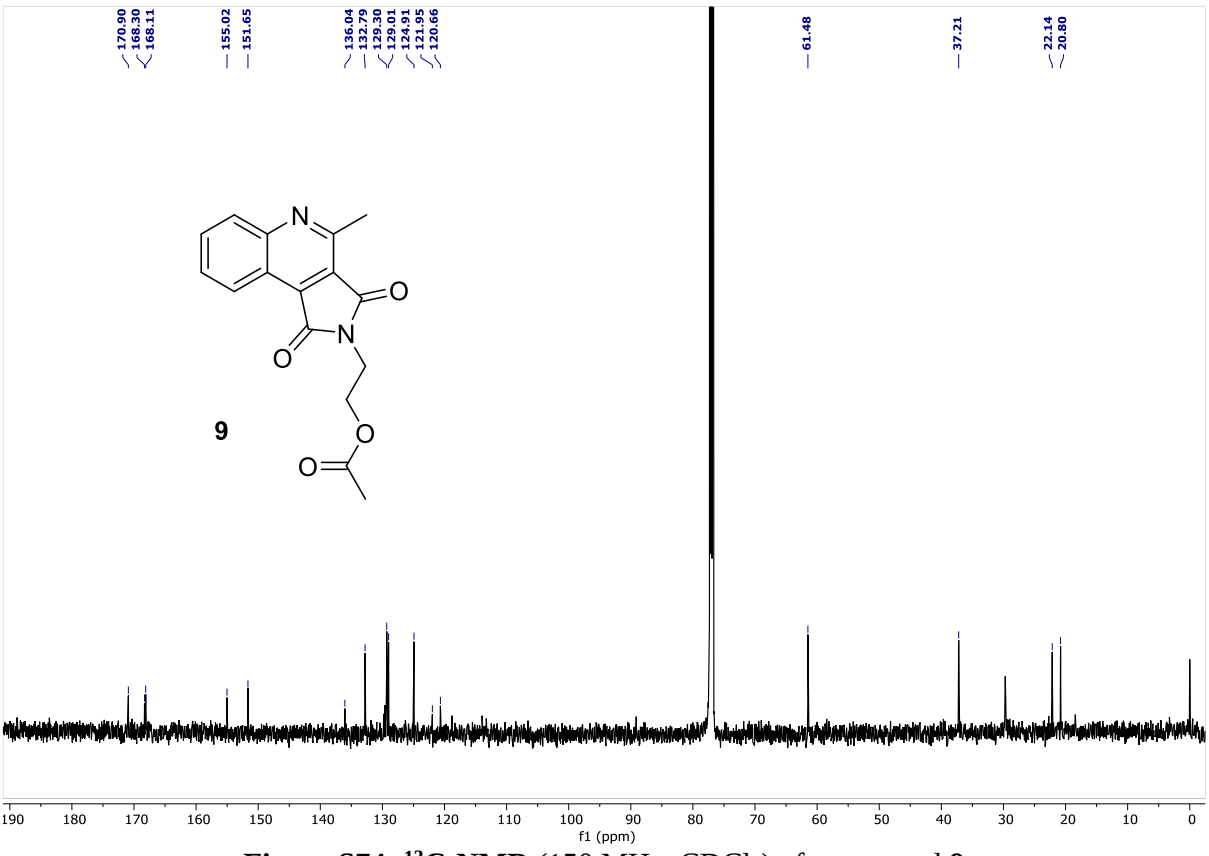


Figure S74: ^{13}C-NMR (150 MHz, $CDCl_3$) of compound **9**

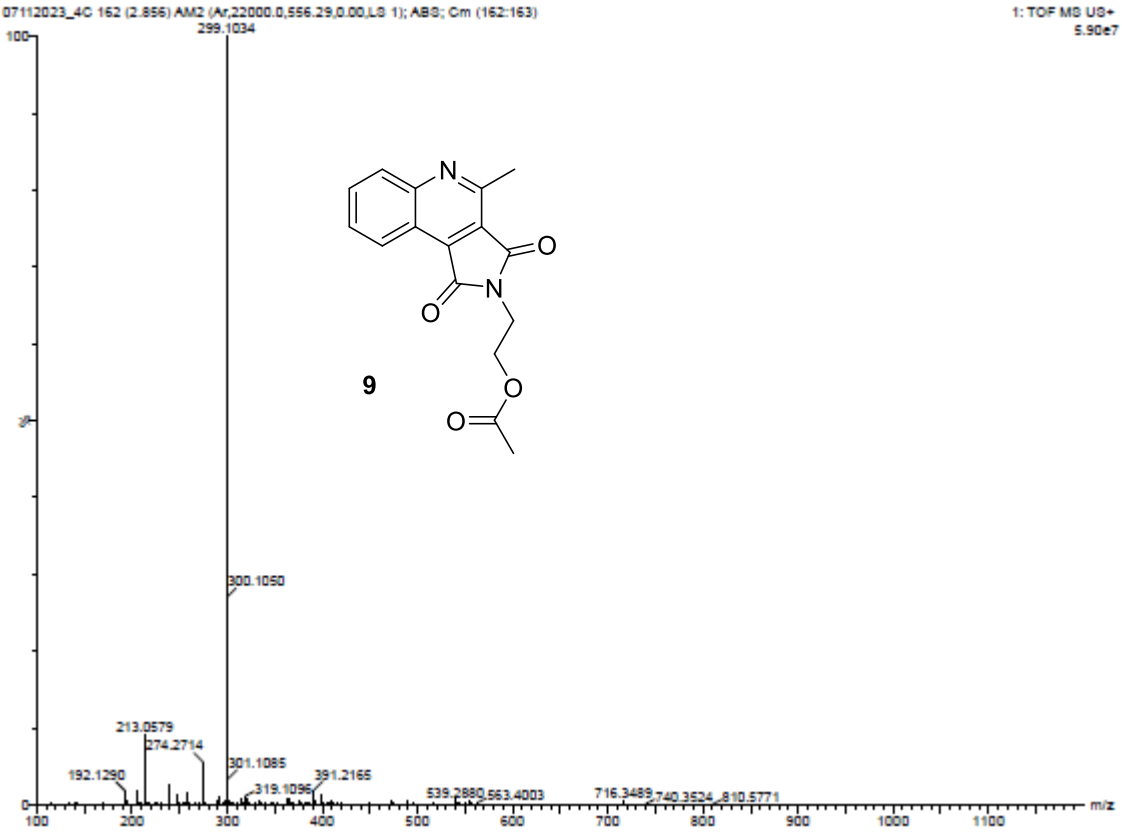


Figure S75: HRMS spectra of compound **9**