SUPPLEMENTARY INFORMATION

Efficient Metal-Free Green Syntheses of 4*H*-Chromenes and 3-Aminoalkylated Indoles Using Reusable Graphite Oxide Carbocatalyst Under Aqueous and Solvent-Free Reaction Conditions

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1. Weight % of C and O in GO

Full results :

Processing option : All elements analyzed Number of iterations : 1

Element	Line	k factor	Absorbtion corrn.	Weight%	Weight% sigma	Atomic%	Standard
C	K_SERIES	1.706	1.0000	17.45	3.51	21.97	Standardless
O	K_SERIES	1.353	1.0000	82.55	3.51	78.03	Standardless
Totals				100.00			

2. Preparation of graphite oxide (GO)

Graphite Oxide (GO) was prepared in accordance with the modified Hummer's method from graphite powder. 20 mL of sulphuric acid (H_2SO_4, 98 %) was kept in a reaction flask. and the flask was kept in an ice bath. To this, 1 g of graphite (Sigma-Aldrich) and sodium nitrate (500 mg) was added. Then, 4 g of potassium permanganate ($KMnO_4$) was added slowly in portions over a period of an hour under stirring to avoid any explosion. After the addition is complete, the reaction mixture was allowed to stir for another hour. The mixture was then heated slowly to 45 ^oC and stirred maintaining the temperature for one more hour. After that, 20 mL of deionized (DI) water was added and heated (45 ^oC) for another 30 mins. Finally, 180 mL of DI water was mixed followed by dropwise addition of hydrogen peroxide (H_2O_2, 30 %) until the colour of the solution changed from dark brown to yellowish-brown. The prepared graphite oxide was then recovered by centrifugation and washed with DI water (3 X 10 mL), EtOH (3 X 10 mL), and diethyl ether (3 X 10 mL). After washing, the graphite oxide was allowed to dry under vacuum to obtain a yellowish-brown powder.

3. Calculation for TON and TOF

For a catalytic reaction system, the effectiveness of the catalyst can be expressed in terms of turnover number (TON) and turnover frequency (TOF). The TON is a unitless measure and can be calculated as:-

TON = % conversion × No. of moles of reactants/ No. of active sites of catalyst in moles

And TOF can be calculated as:-

$$TOF = TON/time$$

From TPD Analysis, 1g of the catalyst contains 39 mmol of active sites

Therefore, 1 mg of the catalyst contains 3.9×10^{-2} mmol of active sites

For 4*H*-Chromenes,

Since, 10 mg of the catalyst is used for the reaction

Hence, 10 mg of the catalyst contains 0.39 mmol of active sites

TON = 92 × 1/0.39

= 236

TOF = 236/3

=79 h^{-1}

For 3-amino alkylated indoles,

Since, 8 mg of the catalyst is used for the reaction

Hence, 8 mg of the catalyst contains 0.31 mmol of active sites

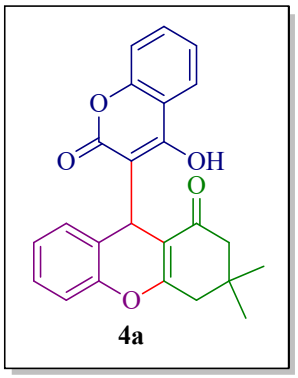
TON = 92 × 1/0.31

= 298

TOF = 236/6

=49 h^{-1}

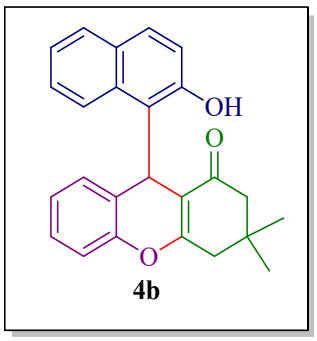
4. Spectral data of the compounds (4a-4q, 8a-8n and 8a* & 8l*)



4a

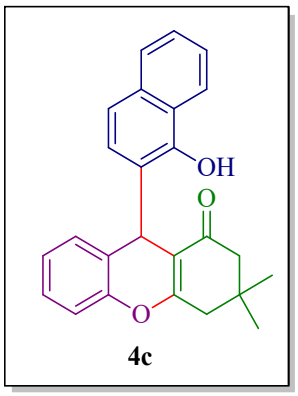
Colourless solid. M.p.: 232-234 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 11.03 (s, 1H), 7.98 (dd, *J*= 1.6 Hz, 1H), 7.45-7.41 (m, 1H), 7.26-7.22 (m, 1H), 7.20-7.14 (m, 2H), 7.06 (d, *J* = 8.0 Hz, 1H), 7.00 (d, *J* = 4.4 Hz, 2H), 5.03 (s, 1H), 2.65-2.49 (dd, *J* = 17.8, 45.8 Hz, 2H), 2.39-2.29

(dd, J =16.8, 25.2 Hz, 2H), 1.11 (s, 3H), 1.01 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 200.3, 169.1, 160.1, 159.9, 151.9, 150.1, 130.5, 127.5, 127.1, 123.9, 123.1, 122.6, 121.2, 115.9, 115.1, 114.9, 108.9, 108.1, 48.8, 40.5, 31.3, 28.2, 27.4, 26.1. [1]



4b

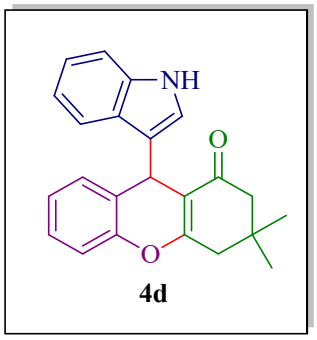
White solid. M.p.: 223-224 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 9.26 (s, 1H), 7.89-7.76 (m, 2H), 7.68 (d, J = 8.0 Hz, 1H), 7.42-7.36 (m, 2H), 7.34 (d, J = 8.8 Hz, 1H), 7.01-6.99 (m, 2H), 6.62-6.60 (m, 2H), 5.76 (s, 1H), 2.60 (s, 2H), 2.44 (dd, J = 16.6, 25.8 Hz, 2H), 1.14 (s, 3H), 0.99 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 200.7, 166.8, 152.8, 147.8, 132.7, 131.5, 131.0, 129.1, 128.7, 128.2, 127.9, 127.5, 125.2, 123.4, 121.5, 118.8, 117.4, 116.5, 113.9, 50.2, 41.5, 32.4, 29.0, 27.9, 27.2. [1]



4c

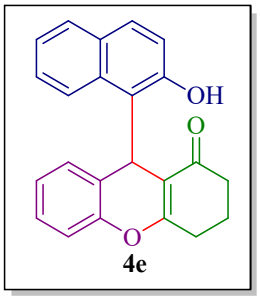
White solid. M.p.: 200-202 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 9.21 (s, 1H), 8.29 (d, J = 8.4 Hz, 1H), 7.82 (d, J = 8 Hz, 1H), 7.62 (t, J = 7.2 Hz, 1H), 7.56-7.53 (m, 2H), 7.09-6.98 (m, 3H), 6.69-6.65 (m, 1H), 6.55 (dd, J = 1.4, 7.8 Hz, 1H), 5.36 (s, 1H), 2.75 (dd, J = 17.6, 28 Hz, 2H), 2.44 (dd, J = 16.6, 24.2 Hz, 2H), 1.16 (s, 3H), 1.02 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 200.8, 166.9, 153.5, 145.0, 133.3, 132.9, 130.0, 127.9, 127.8, 127.2, 126.6, 125.2, 123.3, 121.2,

120.8, 119.4, 118.7, 114.0, 50.2, 41.7, 32.3, 31.0, 29.0, 27.3. HRMS (ESI) m/z calcd. for $C_{25}H_{22}O_3$ $[M+H]^+$ 371.1647, Found 371.1633.



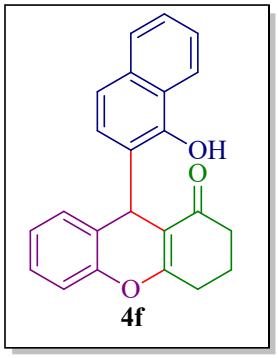
4d

Red solid. M.p.: 178-180 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 8.21 (s, 1H), 7.60 (d, *J*= 8 Hz, 1H), 7.47 (t, *J* = 4.2 Hz, 2H), 7.38-7.36 (m, 2H), 7.34-7.32 (m, 1H), 7.29-7.27 (m, 1H), 7.20-7.15 (m, 2H), 5.51 (s, 1H), 2.82 (dd, *J* = 17.6, 28 Hz, 2H), 2.47 (dd, *J* = 16.4, 28 Hz, 2H), 1.30 (s, 3H), 1.16 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 196.9, 163.8, 149.0, 136.0, 129.6, 126.9, 125.1, 124.7, 124.4, 121.8, 121.0, 119.9, 118.8, 118.5, 115.7, 112.1, 110.7, 50.4, 41.0, 31.6, 28.9, 28.5, 27.1. [1]



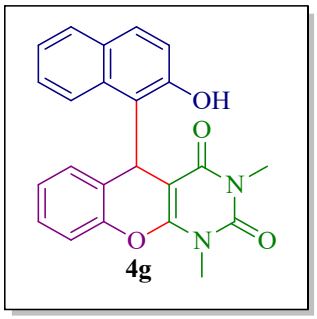
4e

White solid. M.p.: 258-259 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 9.44 (s, 1H), 7.80 (t, *J* = 7 Hz, 2H), 7.66 (d, *J* = 7.6 Hz, 1H), 7.41-7.32 (m, 3H), 7.02 (d, *J*= 3.6 Hz, 2H), 6.64-6.57 (m, 2H), 5.76 (s, 1H), 2.81- 2.66 (m, 2H), 2.60-2.44 (m, 2H), 2.14-1.96 (m, 2H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 201.0, 168.6, 153.0, 147.8, 132.6, 131.5, 131.0, 129.1, 128.9, 128.2, 128.0, 127.5, 125.2, 123.4, 121.4, 118.7, 117.5, 116.5, 115.1, 36.3, 28.1, 27.9, 19.9. [1]



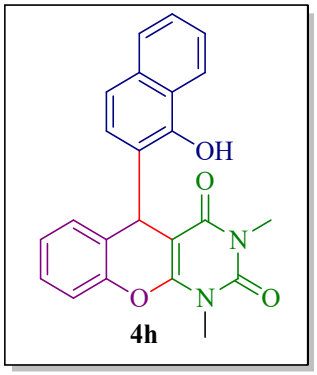
4f

White solid. M.p.: 207-209 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 9.36 (s, 1H), 8.31 (d, *J* = 8.4 Hz, 1H), 7.83 (d, *J* = 8 Hz, 1H), 7.63 (t, *J* = 7.4 Hz, 1H), 7.57-7.53 (m, 2H), 7.08-6.98 (m, 3H), 6.69 (t, *J* = 7.4 Hz, 1H), 6.53 (d, *J* = 8 Hz, 1H), 5.35 (s, 1H), 2.92-2.74 (m, 2H), 2.61-2.45 (m, 2H), 2.17-1.98 (m, 2H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 201.1, 168.8, 153.6, 145.0, 133.3, 132.8, 130.1, 128.0, 127.8, 127.2, 126.66, 126.65, 125.3, 123.3, 121.0, 120.8, 119.4, 118.6, 115.3, 36.4, 31.1, 28.0, 19.9. HRMS (ESI) m/z calcd. for $C_{23}H_{18}O_3$ $[M+H]^+$ 343.1334, Found 343.1329.



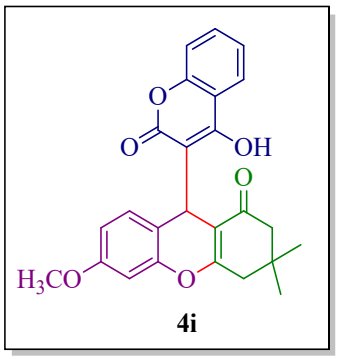
4g

Yellow solid. M.p.: 208-210 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 9.44 (s, 1H), 7.71-7.68 (m, 2H), 7.42 (d, *J* = 8 Hz, 1H), 7.34 (d, *J* = 8.8 Hz, 1H), 7.20 (d, *J* = 3.2 Hz, 2H), 7.18-7.12 (m, 2H), 6.97-6.93 (m, 1H), 6.88 (d, *J* = 7.6 Hz, 1H), 6.05 (s, 1H), 3.68 (s, 3H), 3.30 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 164.6, 153.7, 153.2, 150.1, 149.1, 131.8, 130.7, 130.0, 129.8, 129.1, 128.3, 126.3, 125.7, 124.6, 122.7, 122.6, 122.2, 121.0, 115.8, 89.3, 30.6, 29.4, 28.5. [1]



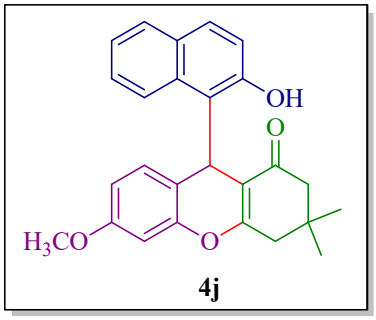
4h

Orange solid. M.p.: 148-150 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 10.09 (s, 1H), 8.45 (d, J = 8.4 Hz, 1H), 7.66 (d, J = 8 Hz, 1H), 7.50- 7.46 (m, 1H), 7.44-7.40 (m, 1H), 7.36-7.27 (m, 2H), 7.20 (d, J = 8.8 Hz, 1H), 7.18-7.14 (m, 1H), 7.09-7.07 (m, 1H), 6.61 (d, J = 8.8 Hz, 1H), 5.69 (s, 1H), 3.59 (s, 3H), 3.34 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 165.0, 153.4, 149.9, 149.8, 149.5, 133.4, 130.7, 128.5, 127.0, 126.8, 126.3, 126.1, 125.6, 125.4, 124.1, 123.4, 120.8, 116.2, 90.6, 31.9, 29.3, 28.6. HRMS (ESI) m/z calcd. for $C_{23}H_{18}N_2O_4$ $[M+Na]^+$ 409.1164, Found 409.1157.



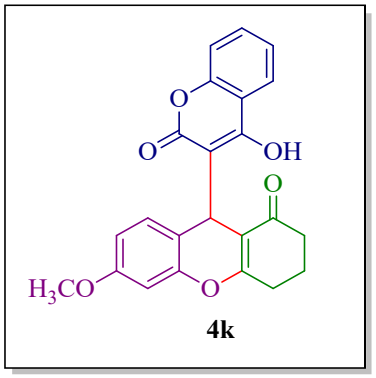
4i

Yellow solid. M.p.: 246-248 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 11.02 (s, 1H), 8.01 (d, J = 7.6 Hz, 1H), 7.49 (t, J = 7.6 Hz, 1H), 7.30 (t, J = 7.2 Hz, 1H), 7.20 (d, J = 8.4 Hz, 1H), 6.94 (d, J = 8.4 Hz, 1H), 6.66 (s, 1H), 6.61 (d, J = 8.4 Hz, 1H), 5.02 (s, 1H), 3.78 (s, 3H), 2.67 (dd, J = 17.8, 42.2 Hz, 2H), 2.43 (dd, J = 16.8, 26 Hz, 2H), 1.15 (s, 3H), 1.05 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 201.3, 169.9, 161.1, 160.7, 159.4, 153.0, 151.8, 131.5, 128.9, 124.1, 123.6, 117.0, 116.1, 114.1, 111.4, 110.2, 109.1, 101.3, 55.4, 49.8, 41.6, 32.3, 29.2, 27.9, 27.1. [1]



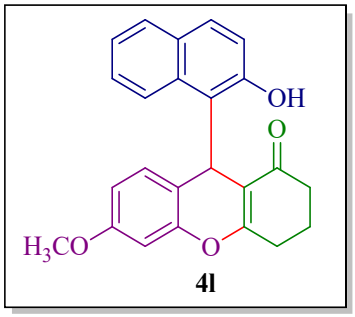
4j

White solid. M.p.: 255-257 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 9.40 (s, 1H), 7.79 (d J = 8.4 Hz, 2H), 7.67 (d, J = 8 Hz, 1H), 7.43-7.36 (m, 2H), 7.33 (d, J = 8.8 Hz, 1H), 6.57 (s, 1H), 6.48 (d, J = 8.8 Hz, 1H), 6.20 (d, J = 8.4 Hz, 1H), 5.67 (s, 1H), 3.68 (s, 3H), 2.60 (s, 2H), 2.44 (dd, J = 16.6, 24.2 Hz, 2H), 1.15 (s, 3H), 1.00 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 200.8, 166.6, 159.2, 153.8, 147.8, 131.5, 131.0, 129.3, 129.0, 128.2, 127.4, 125.2, 125.1, 123.5, 117.5, 116.5, 114.0, 108.1, 103.4, 55.1, 50.2, 41.5, 32.4, 29.0, 27.5, 27.2. [2]



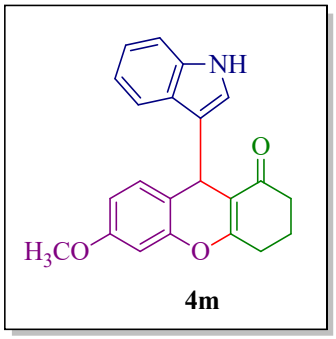
4k

White solid. M.p.: 186-188°C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 11.15 (s, 1H), 8.00, (dd, J = 1.4, 7.8 Hz, 1H), 7.49-7.45 (m, 1H), 7.29-7.27 (m, 1H), 7.20 (d, J = 8.4 Hz, 1H), 6.93 (d, J = 8.4 Hz, 1H), 6.66 (d, J = 2.4 Hz, 1H), 6.61 (dd, J = 2.6, 8.6 Hz, 1H), 5.02 (s, 1H), 3.78 (s, 3H), 2.85-2.78 (m, 1H), 2.69-2.57 (m, 1H), 2.55-2.43 (m, 2H), 2.13-1.97 (m, 2H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 201.8, 171.6, 161.2, 160.8, 159.4, 152.9, 151.7, 131.5, 128.9, 124.1, 123.6, 117.0, 116.1, 114.3, 111.6, 111.3, 109.2, 101.2, 55.4, 35.9, 28.04, 28.00, 19.9. [1]



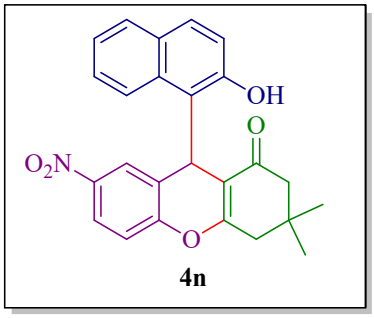
4l

White solid. M.p.: 232-234 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 9.55 (s, 1H), 7.79-7.77 (m, 2H), 7.66 (d, J = 8 Hz, 1H), 7.42-7.35 (m, 2H), 7.34 (d, J = 8.8 Hz, 1H), 6.57 (s, 1H), 6.45 (d, J = 8.8 Hz, 1H), 6.20 (d, J = 8.8 Hz, 1H), 5.67 (s, 1H), 3.68 (s, 3H), 2.81-2.66 (m, 2H), 2.61-2.44 (m, 2H), 2.17-1.96 (m, 2H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 201.2, 168.5, 159.3, 154.0, 147.8, 131.5 131.0, 129.5, 129.0, 128.2, 127.4, 125.2, 125.1, 123.4, 117.5, 116.5, 115.3, 108.0, 103.3, 55.1, 36.3, 27.8, 27.6, 19.9. [2]



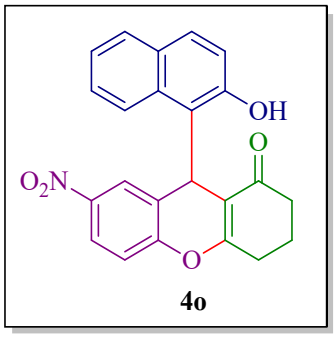
4m

White solid. M.p.: 206-208 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 7.97 (s, 1H), 7.41-7.36 (m, 2H), 7.25-7.24 (m, 1H), 7.21-7.17 (m, 1H), 7.14 (d, J = 2.4 Hz, 1H), 7.00-6.95 (m, 3H), 5.27 (s, 1H), 3.76 (s, 3H), 2.77-2.61 (m, 2H), 2.40-2.27 (m, 2H), 2.04-1.89 (m, 2H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 197.4, 165.8, 158.8, 150.1, 136.5, 130.6, 125.7, 122.4, 121.5, 120.7, 119.3, 119.1, 117.4, 114.3, 111.5, 111.1, 101.1, 55.4, 37.0, 28.9, 27.8, 20.4. HRMS (ESI) m/z calcd. for $C_{22}H_{19}NO_3$ $[M+Na]^+$ 368.1263, Found 368.1255. [2]



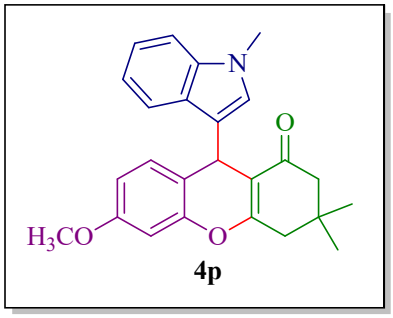
4n

White solid. M.p.: 246-248 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 10.46 (s, 1H), 7.93 (d, J = 9.2 Hz, 1H), 7.86 (d, J = 9.2 Hz, 1H), 7.82-7.81 (m, 1H), 7.49-7.45 (m, 2H), 7.41-7.38 (m, 3H), 7.10 (d, J = 9.2 Hz, 1H), 5.75 (s, 1H), 2.72 (dd, J = 17.8, 26.2 Hz, 2H), 2.48 (dd, J = 16.8, 25.6 Hz, 2H), 1.17 (s, 3H), 1.01 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 201.3, 168.3, 159.5, 147.9, 142.0, 133.1, 131.7, 130.5, 130.0, 128.7, 127.7, 125.5, 125.2, 124.3, 122.7, 119.3, 116.8, 115.7, 112.7, 50.0, 41.6, 32.4, 29.0, 28.2, 27.2. [2]



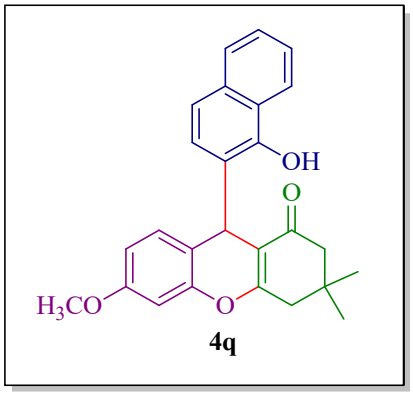
4o

Light Yellow solid. M.p.: 376-378 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 10.57 (s, 1H), 7.94 (dd, J = 2.4, 9.2 Hz, 1H), 7.86 (d, J = 8.8 Hz, 1H), 7.83-7.81 (m, 1H), 7.50-7.48 (m, 1H), 7.44-7,39 (m, 4H), 7.09 (d, J = 8.8 Hz, 1H), 5.75 (s, 1H), 2.90-2.71 (m, 2H), 2.65- 2.48 (m, 2H), 2.19-2.02 (m, 2H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 201.6, 169.9, 159.5, 147.9, 142.0, 133.1, 131.7, 130.4, 130.0, 128.7, 127.7, 125.5, 125.4, 124.3, 122.7, 119.2, 116.8, 115.8, 113.9, 36.2, 28.3, 28.0, 19.8. [2]



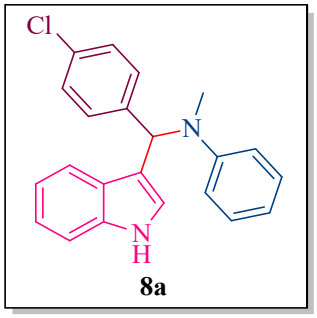
4p

Orange solid, M.p.: 160-162 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 7.40-7.30 (m, 2H), 7.25-7.17 (m, 2H), 7.11-7.00 (m, 3H), 6.96 (t, J = 7.4 Hz, 1H), 5.21 (s, 1H), 3.76 (s, 3H), 3.69 (s, 3H), 2.60 (dd, J = 17.2, 29.6 Hz, 2H), 2.25 (dd, J = 16.4, 26.0 Hz, 2H), 1.09 (s, 3H), 0.95 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 200.9, 166.7, 159.3, 154.5, 133.2, 130.6, 127.8, 127.3, 126.6, 125.3, 125.2, 123.3, 120.8, 119.5, 114.2, 107.9, 103.2, 55.1, 50.3, 41.7, 32.4, 30.9, 30.5, 29.0, 27.4. [3]



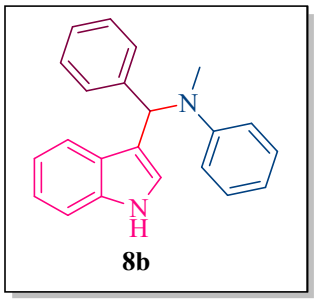
4q

Orange solid. M.p.: 200-202 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 9.33 (s, 1H), 8.29 (d, J = 8.4 Hz, 1H), 7.83 (d, J = 8 Hz, 1H), 7.63-7.59 (m, 1H), 7.56 (d, J = 8.4 Hz, 2H), 7.05 (d, J = 8.4 Hz, 1H), 6.56 (d, J = 2.4 Hz, 1H), 6.41 (d, J = 8.8 Hz, 1H), 6.26 (dd, J = 2.4, 8.8 Hz, 1H), 5.26 (s, 1H), 3.71 (s, 3H), 2.75 (dd, J = 17.6, 28.4 Hz, 2H), 2.44 (dd, J = 16.8, 22.8 Hz, 2H), 1.16 (s, 3H), 1.03 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 200.8, 166.6, 159.3, 153.9, 147.8, 131.5, 131.1, 129.3, 128.2, 127.4, 125.2, 123.5, 117.5, 116.5, 114.1, 108.2, 103.4, 55.1, 50.2, 41.6, 32.4, 29.0, 27.5, 27.2. HRMS (ESI) m/z calcd. for $C_{26}H_{24}O_4$ $[M+Na]^+$ 423.1572, Found 423.1563.



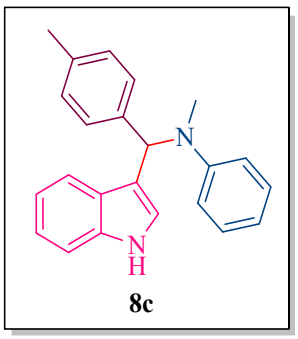
8a

White solid, M.p.: 128-130 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 7.91 (s, 1H,), 7.33 (d, *J* = 8 Hz, 1H), 7.25-7.20 (m, 4H), 7.17-7.13 (m, 3H), 7.01-6.96 (m, 3H), 6.54-6.52 (m, 3H), 5.52 (s,1H), 2.80 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 147.7, 143.3, 136.7, 132.3, 131.6, 130.3, 129.6, 128.3, 126.9, 124.0, 122.1, 120.1, 119.9, 119.4, 112.4, 111.1, 47.3, 30.9. ESI MS: m/z 347.48, 349.47 $[M+H]^+$. [4]



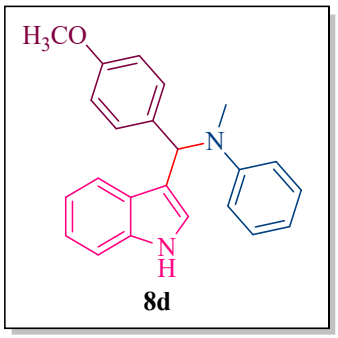
8b

White solid, M.p.: 125-127 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 7.82 (s, 1H), 7.25 (d, *J* = 8 Hz, 1H), 7.20-7.14 (m, 5H), 7.12-7.03 (m, 3H), 6.97 (d, *J* = 8.4 Hz, 2H), 6.91-6.84 (m, 1H), 6.47-6.45 (m, 3H), 5.48 (s, 1H), 2.72 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 147.6, 144.7, 136.7, 133.0, 130.1, 129.7, 129.4, 128.9, 128.2, 128.1, 127.1, 126.0, 124.0, 121.9, 120.5, 120.0, 119.2, 112.5, 112.4, 111.0, 47.9, 30.9. [4]



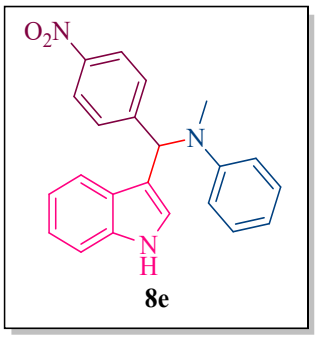
8c

White solid, M.p.: 168-170 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 7.87 (s, 1H), 7.31 (d, *J* = 8Hz, 1H), 7.25-7.22 (m, 1H), 7.15-7.11 (m, 8H), 6.98 (t, *J* = 7.4 Hz, 1H), 6.53 (m, 3H), 5.52 (s, 1H), 2.78 (s, 3H), 2.30 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 147.5, 141.8, 136.7, 135.4, 133.2, 129.7, 128.9, 128.8, 127.1, 124.0, 121.9, 120.6, 120.1, 119.2, 112.4, 111.0, 47.5, 30.9, 21.1.[5]



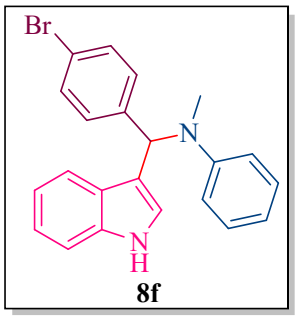
8d

White solid, M.p.: 158-160 °C , ^{1}H NMR ($CDCl_3$, 400 MHz): δ =7.93 (s, 1H), 7.35 (d, *J* = 8 Hz, 1H), 7.27 (s, 1H), 7.18-7.14 (m, 4H), 7.05 (d, *J* = 8.4 Hz, 2H), 7.00 (t, *J*= 7.4 Hz, 1H), 6.83 (d, *J* = 8.4 Hz, 2H), 6.58 (s, 1H), 6.56 (d, *J* = 8.4 Hz, 2H), 5.53 (s, 1H), 3.79 (s, 3H), 2.82 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 157.7, 147.4, 136.9, 136.7, 133.4, 129.8, 129.6, 127.0, 123.8, 121.9, 120.9, 120.1, 119.2, 113.5, 112.4, 110.9, 55.2, 47.0, 31.0. HRMS (ESI) m/z calcd. for $C_{23}H_{22}N_2O$ $[M+H]^+$ 343.1810, Found 343.1811.



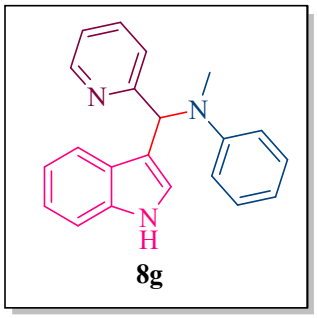
8e

Yellow solid, M.p.: 93.95 °C, ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 8.15 (d, *J* = 8.4 Hz, 2H), 8.08 (s, 1H), 7.41 (d, *J* = 8.8 Hz, 3H), 7.28-7.19 (m, 3H), 7.08-7.02 (m, 3H), 6.75 (m 2H), 6.60 (s, 1H), 5.68 (s, 1H), 2.87 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 152.5, 148.1, 146.3, 136.7, 131.0, 129.7, 129.6, 126.6, 124.0, 123.5, 122.3, 119.67, 119.63, 119.1, 112.4, 111.2, 47.9, 30.7. [4]



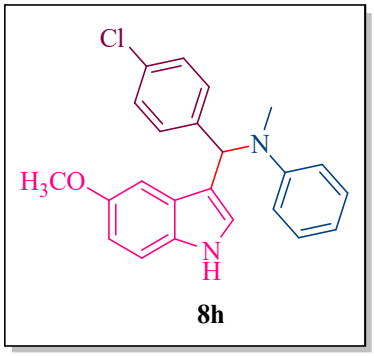
8f

Red solid, M.p.: 136-138 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 7.93 (s, 1H,), 7.37-7.32 (m, 3H), 7.24-7.14 (m, 3H), 7.10 (d, *J* = 8 Hz, 2H), 7.01-6.99 (m, 3H), 6.55 (m, 3H), 5.51 (s, 1H), 2.80 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 147.7, 143.8, 136.7, 132.2, 131.2, 130.7, 129.6, 126.8, 124.0, 122.1, 120.0, 119.9, 119.8, 119.4, 112.4, 111.1, 47.4, 30.9. HRMS (ESI) m/z calcd. for $C_{22}H_{19}BrN_2$ $[M+H]^+$ 391.0810, Found 391.0828.



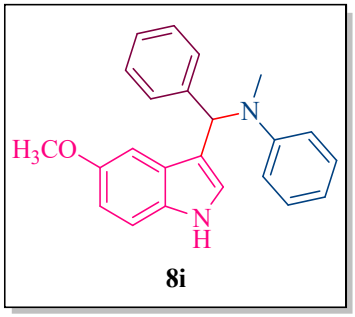
8g

Yellow solid, M.p.: 125-127 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ =8.58 (d, *J*= 4.4 Hz, 1H), 8.08 (s, 1H), 7.57-7.53 (m, 1H), 7.32 (d, *J* = 8.0 Hz, 1H), 7.22-7.13 (m, 3H), 7.11 (d, *J*= 8.4 Hz, 3H), 7.01-6.94 (m, 2H), 6.67 (s, 1H), 6.56 (d, *J* = 8.4 Hz, 2H), 5.72 (s, 1H), 2.80 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ =164.0, 149.3, 147.8, 136.7, 136.4, 129.6, 127.0, 123.8, 122.0, 121.2, 119.8, 119.3, 112.5, 111.0, 50.6, 30.9. HRMS (ESI) m/z calcd. for $C_{21}H_{19}N_3$ $[M+H]^+$ 314.1657, Found 314.1638.



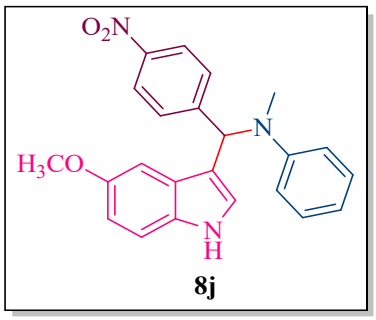
8h

White solid, M.p.: 110-112 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 7.85 (s, 1H), 7.24-7.21 (m, 4H), 7.16 (d, *J* = 8 Hz, 2H), 7.01 (d, *J* = 8.4 Hz, 2H), 6.83 (dd, *J* = 2.2, 8.6 Hz, 1H), 6.63 (d, *J* = 1.6 Hz, 1H), 6.55 (d, *J* = 8 Hz, 3H), 5.47 (s, 1H), 3.68 (s, 3H), 2.80 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 153.7, 147.7, 143.2, 132.3, 131.9, 131.6, 130.3, 129.6, 128.3, 127.3, 124.8, 119.7, 112.4, 112.0, 111.7, 101.9, 55.8, 47.3, 30.9. [5]



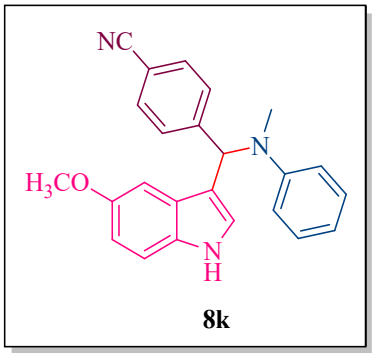
8i

White solid, M.p.: 108-110 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 7.81 (s, 1H), 7.27-7.16 (m, 7H), 7.05 (d, *J* = 8.4 Hz, 2H), 6.81 (dd, *J* = 2.2, 8.6 Hz, 1H), 6.65 (d, *J* = 2.4 Hz 1H), 6.54 (d, *J* = 8.4 Hz, 3H), 5.50 (s, 1H), 3.66 (s, 3H), 2.79 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 153.6, 147.6, 144.6, 132.8, 131.8, 129.7, 128.9, 128.1, 127.5, 125.9, 124.7, 120.2, 112.4, 111.9, 111.6, 102.0, 55.7, 47.9, 30.9. [6]



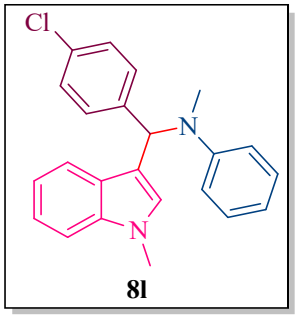
8j

Yellow solid, M.p.: 102-105 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 8.11 (d, *J* = 8.8 Hz, 2H), 7.99 (s, 1H), 7.38 (d, *J* = 8 Hz, 3H), 7.24 (d, *J* = 8.8 Hz, 1H), 7.00 (d, *J* = 8.4 Hz, 2H), 6.84 (dd, *J* = 2.4, 8.8 Hz, 1H), 6.60 (d, *J* = 2.4 Hz, 1H), 6.56-6.54 (m, 3H), 5.58 (s, 1H), 3.67 (s, 3H), 2.80 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 153.8, 152.6, 148.1, 146.2, 131.9, 131.0, 129.7, 129.6, 127.0, 124.9, 123.5, 118.5, 112.5, 112.1, 112.0, 101.6, 55.8, 47.8, 30.8. HRMS (ESI) m/z calcd. for $C_{23}H_{21}N_3O_3$ $[M+H]^+$ 388.1661, Found 388.1643.



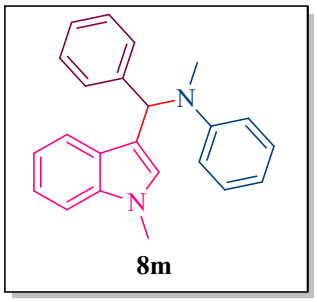
8k

White solid, M.p.: 145-148 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 7.93 (s, 1H), 7.55 (d, *J* = 8 Hz, 2H), 7.34 (d, *J* = 8 Hz, 3H), 7.25 (d, *J* = 8.4 Hz, 1H), 6.99 (d, *J* = 8.4 Hz, 2H), 6.84 (dd, *J* = 2, 8.4 Hz, 1H), 6.59 (d, *J* = 1.6 Hz, 1H), 6.56 (d, *J* = 8 Hz, 3H), 5.54 (s, 1H), 3.67 (s, 3H), 2.81 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 153.9, 150.1, 146.4, 132.8, 132.1, 131.8, 129.8, 129.7, 127.1, 124.8, 119.1, 118.6, 113.7, 112.2, 111.9, 109.9, 101.7, 55.8, 48.1, 31.6. HRMS (ESI) m/z calcd. for $C_{24}H_{21}N_3O$ $[M+H]^+$ 368.1763, Found 368.1756.



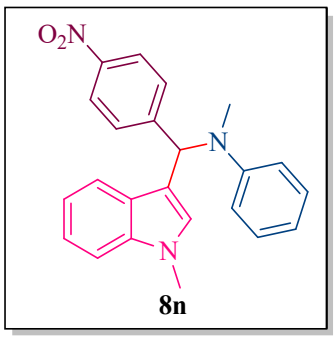
8l

White solid, M.p.: 86-88 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 7.21-7.06 (m, 8H), 6.92-6.88 (m, 3H), 6.50 (d, *J* = 8.4 Hz, 2H), 6.32 (s, 1H), 5.44 (s, 1H), 3.61 (s, 3H), 2.74 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 147.4, 143.4, 137.4, 132.9, 131.6, 130.3, 129.6, 128.7, 128.3, 127.2, 121.6, 120.0, 118.8, 118.4, 112.8, 109.1, 47.3, 32.7, 31.2. HRMS (ESI) m/z calcd. for $C_{23}H_{21}N_2Cl$ $[M+H]^+$ 361.1472, Found 361.1450.



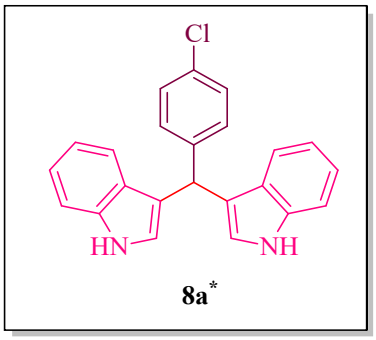
8m

Light Pink solid, M.p.: 110-112 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 7.27-7.21 (m, 7H), 7.19 (t, *J* = 7.6 Hz, 2H), 7.04 (d, *J* = 8.4 Hz, 2H), 6.97 (t, *J* = 7.4 Hz, 1H), 6.55 (d, *J* = 8.4 Hz, 2H), 6.41 (s, 1H), 5.55 (s, 1H), 3.66 (s, 3H), 2.79 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 147.5, 144.9, 137.5, 133.2, 129.8, 129.0, 128.8, 128.2, 127.5, 126.0, 121.6, 120.2, 119.0, 118.8, 112.5, 109.1, 48.0, 32.7, 31.1. [5]



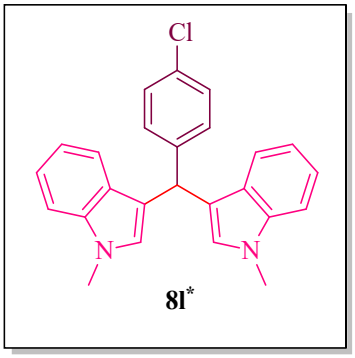
8n

Yellow solid, M.p.: 146-148 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 8.12 (d, *J* = 8.8 Hz, 2H), 7.39 (d, *J* = 8.8 Hz, 2H), 7.31 (d, *J* = 8.4 Hz, 1H), 7.22 (t, *J* = 7.8 Hz, 1H), 7.17 (d, *J* = 8 Hz, 1H), 7.01-6.96 (m, 4H), 6.57 (d, *J* = 8.4 Hz, 2H), 6.42 (s, 1H), 5.63 (s, 1H), 3.71 (s, 3H), 2,82 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 152.7, 148.0, 146.3, 137.4, 131.2, 129.7, 129.6, 128.7, 127.0, 123.5, 121.8, 119.7, 119.0, 117.4, 112.5, 109.3, 47.8, 32.7, 30.9. HRMS (ESI) m/z calcd. for $C_{23}H_{21}N_3O_2$ $[M+H]^+$ 372.1712, Found 372.1732.



8a*

Red solid, M.p.: 80-82 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 7.92 (s, 2H), 7.40-7.34 (m, 6H), 7.25-7.27 (m, 1H), 7.22-7.14 (m, 3H), 7.02 (t, *J* = 7.4 Hz, 2H), 6.66 (d, *J* = 1.6 Hz, 2H), 5.88 (s, 1H), [7]



8l*

White solid, M.p.: 208-210 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 7.41 (d, *J* = 7.6 Hz, 2H), 7.36-7.24 (m, 8H), 7.07 (t, *J* = 7.4 Hz, 2H), 6.56 (s, 2H), 5.90 (s, 1H), 3.73 (s, 6H). [8]

5. References

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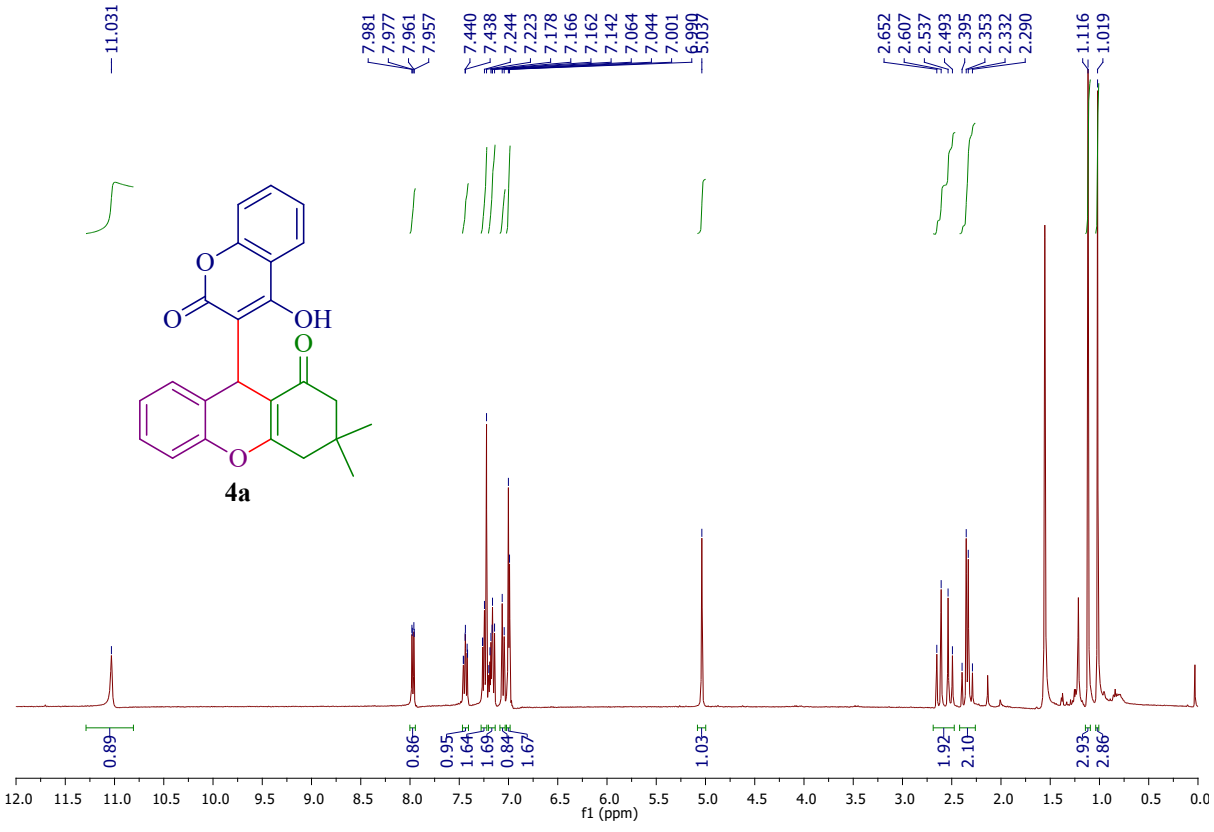
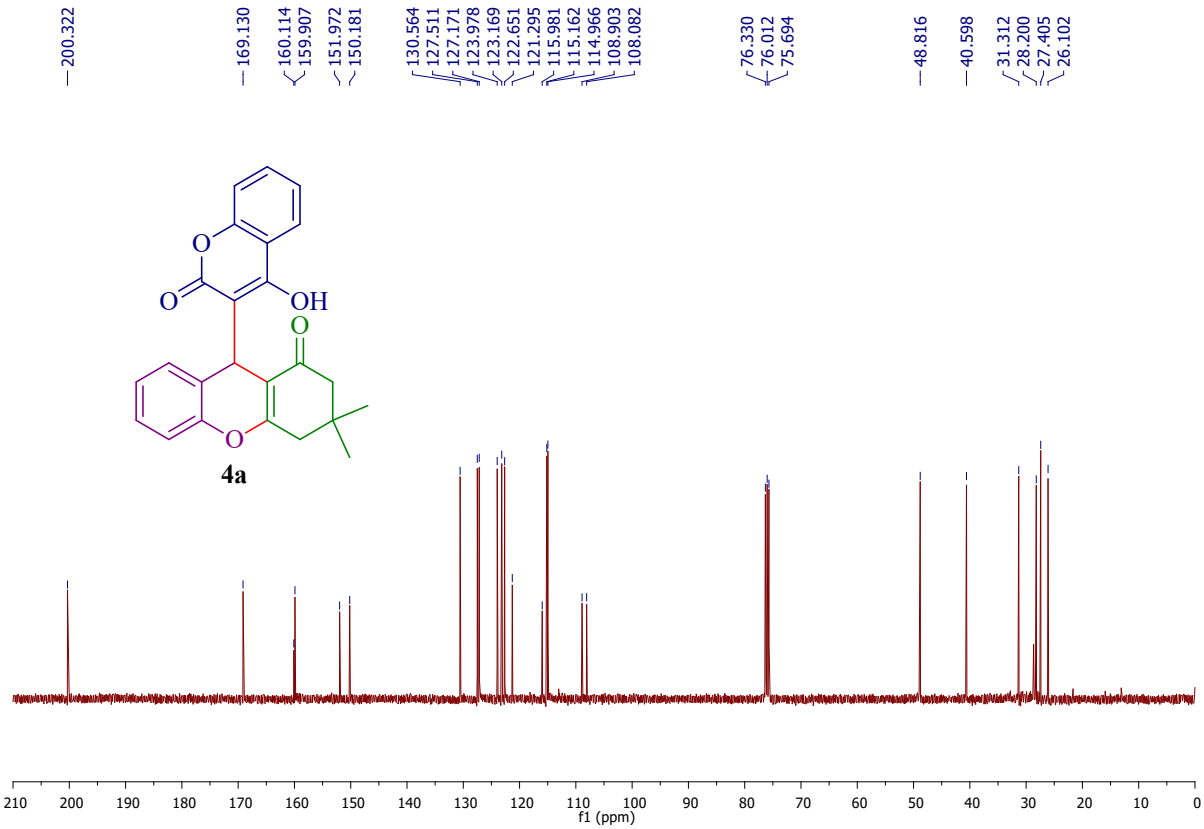
6. ^{1}H and ^{13}C NMR spectra of compounds (4a-4q, 8a-8n and 8a* & 8l*)

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159.907
151.972
150.181
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123.978
123.169
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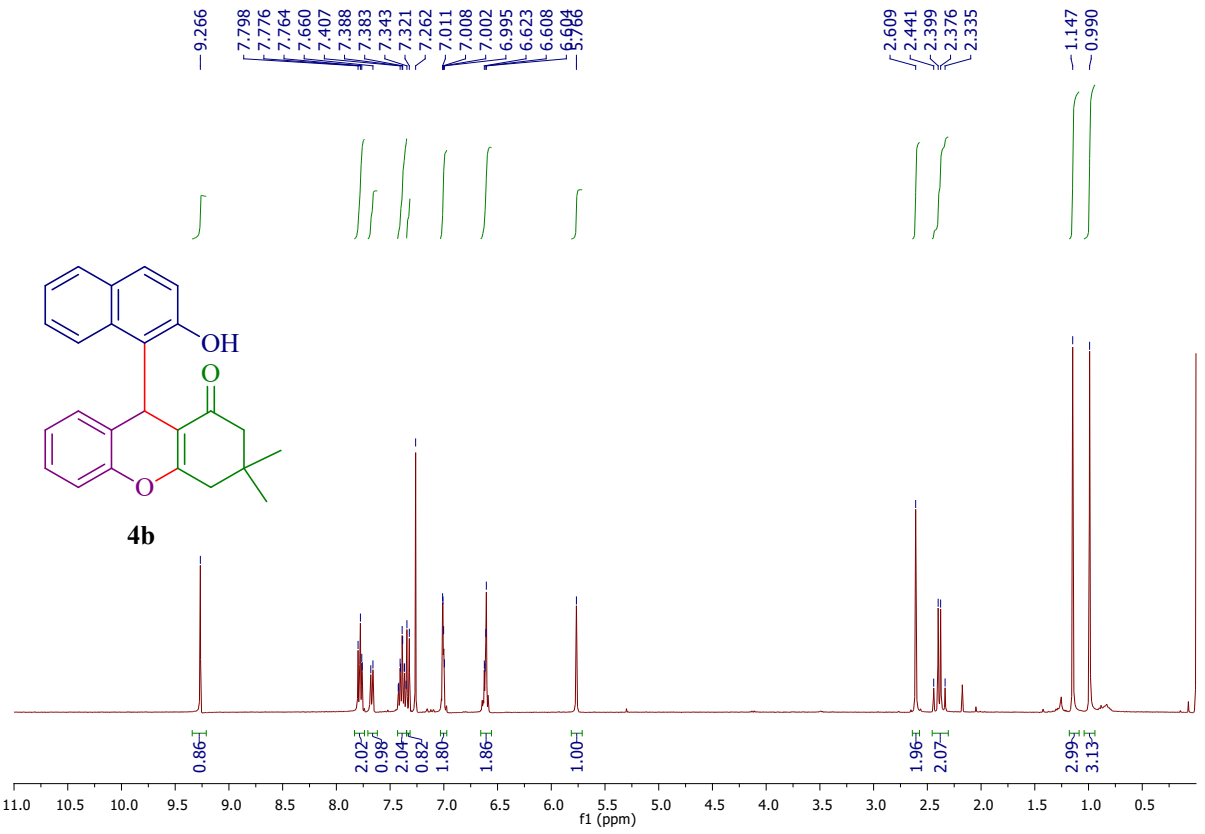
115.981
115.162
114.966
108.903
108.082
76.330
76.012
75.694
48.816
40.598
31.312
28.200
27.405
26.102
O
O
OH
O
O
f1 (ppm)

4a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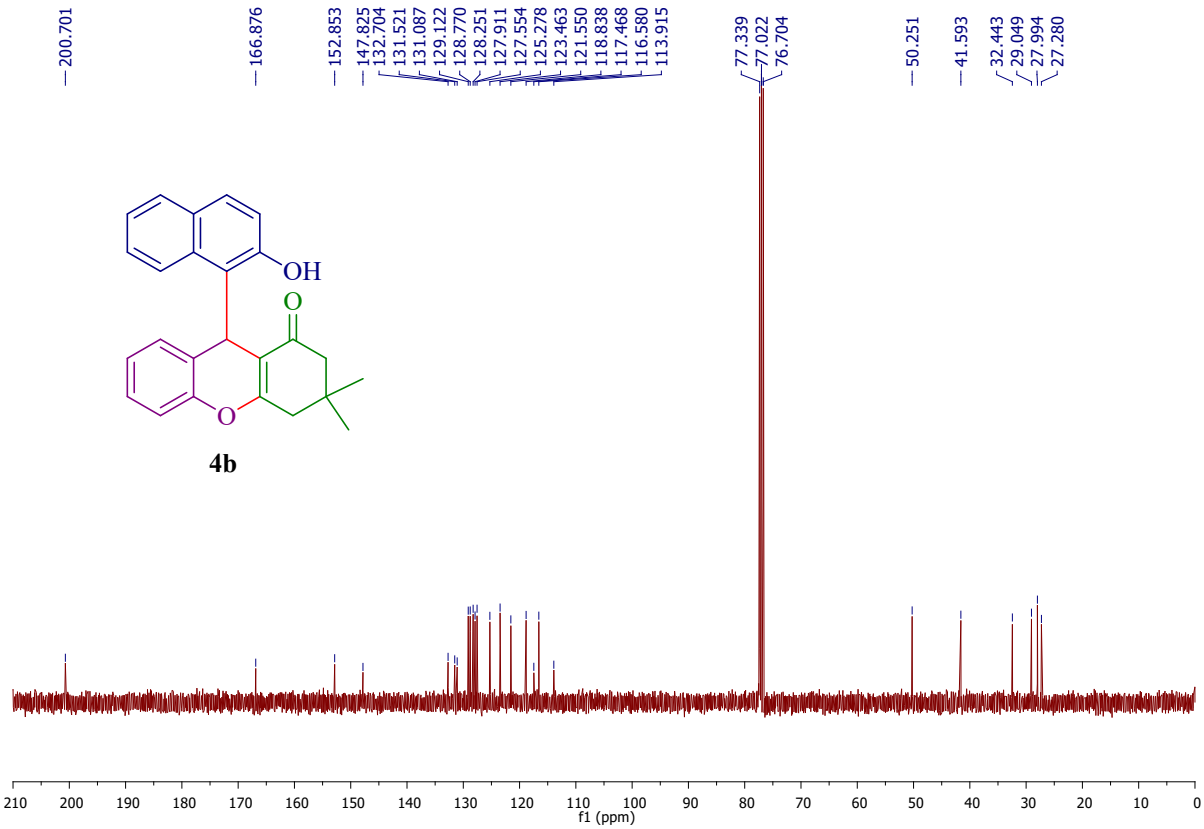
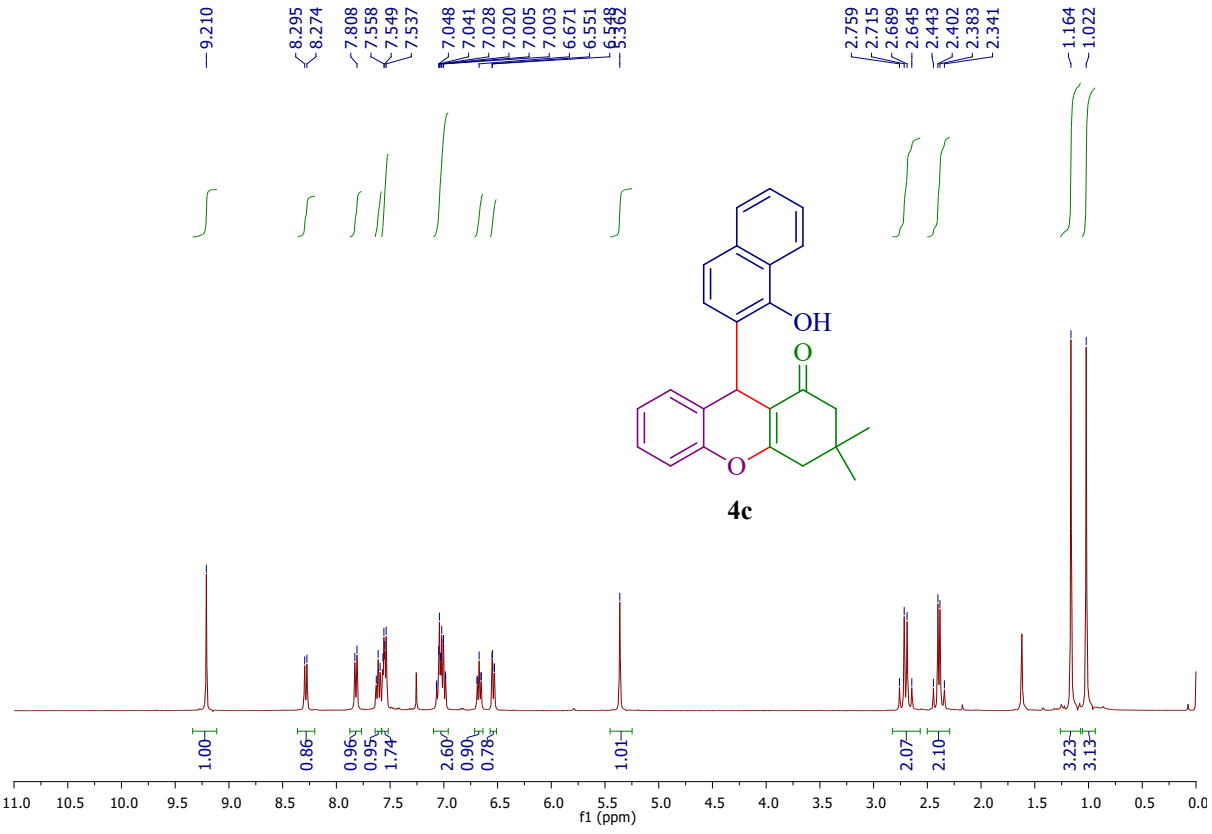
7.008
7.002
6.995
6.623
6.608
2.609
2.441
2.399
2.376
2.335
1.147
0.990
OH
O
O
0.86
2.02
0.98
2.04
0.82
1.80
1.86
1.00
1.96
2.07
2.99
3.13
f1 (ppm)

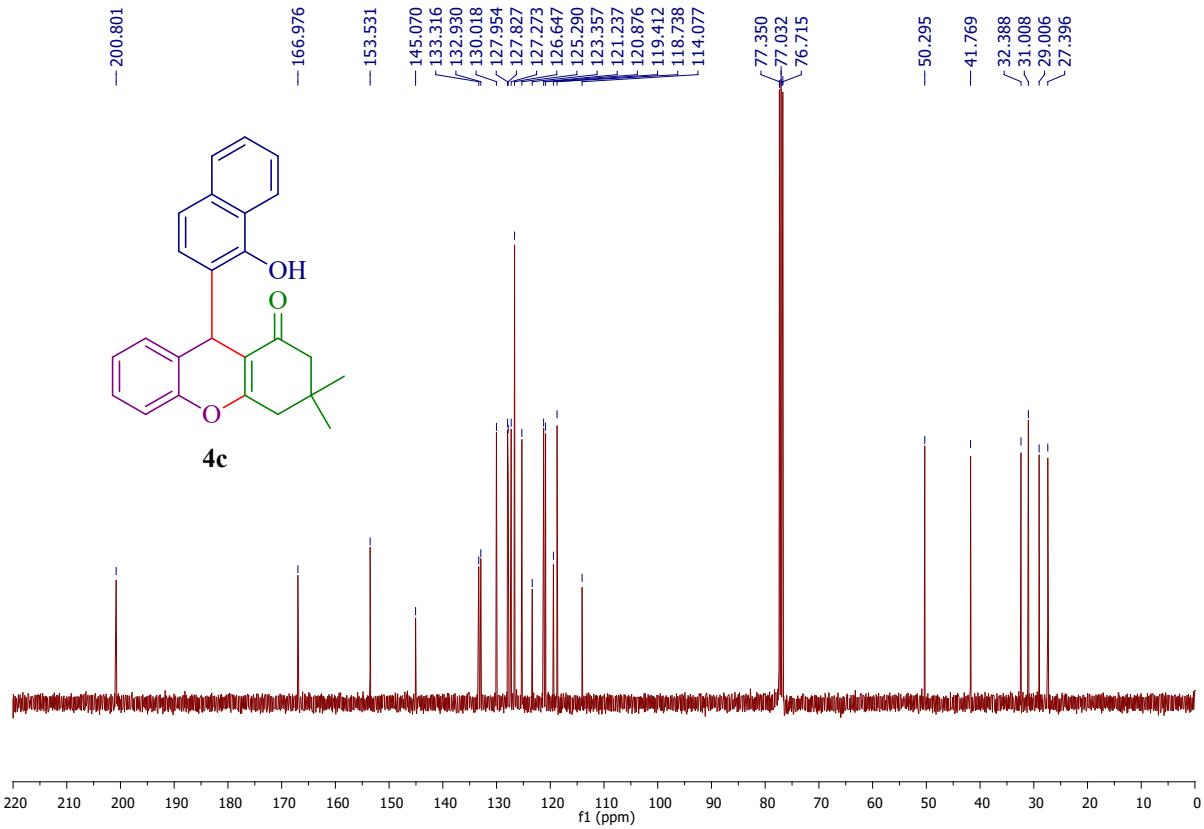
4b


200.701
166.876
152.853
147.825
132.704
131.521
131.087
129.122
128.770
128.251
127.911
127.554
125.278
123.463
121.550
118.838
117.468
116.580
113.915
77.339
77.022
76.704
50.251
41.593
32.443
29.049
27.994
27.280
OH
O
O
f1 (ppm)

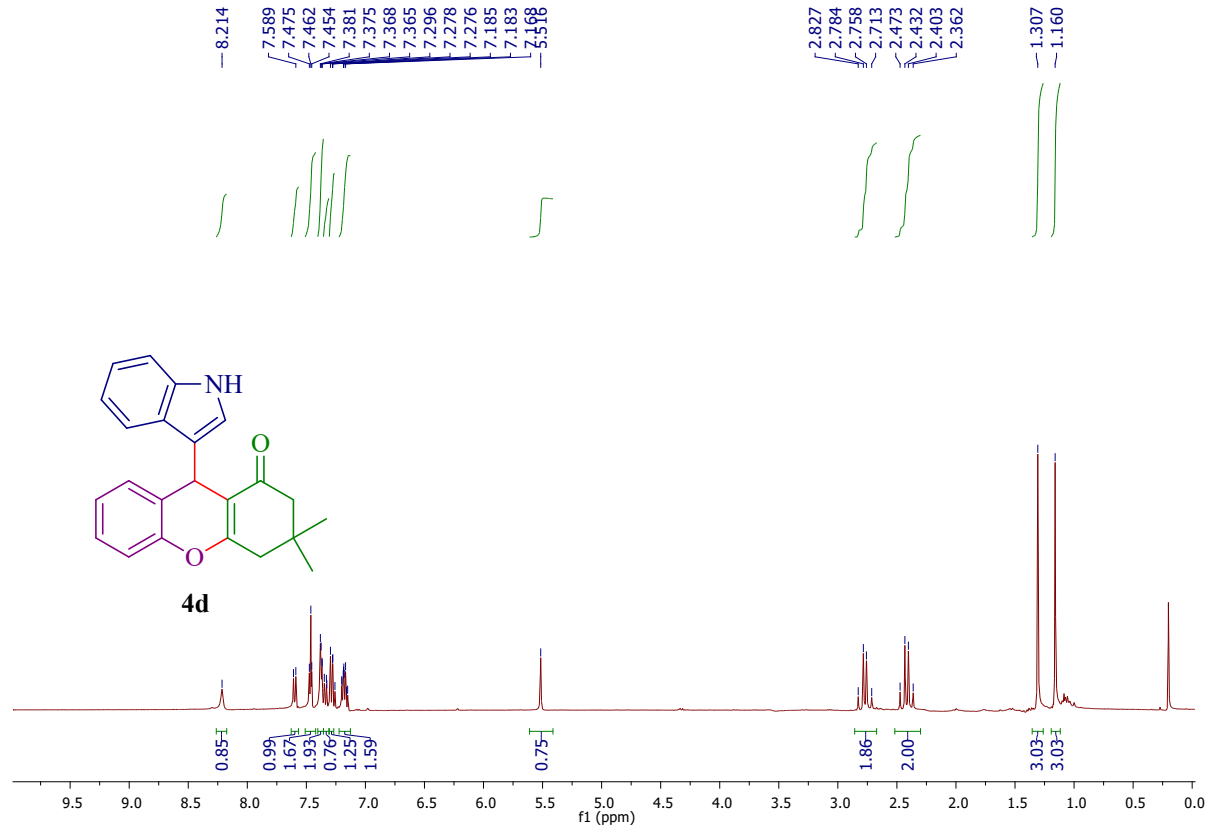
4b


9.210
8.295
8.274
7.808
7.558
7.549
7.537
7.048
7.041
7.028
7.020
7.005
7.003
6.671
6.551
2.759
2.715
2.689
2.645
2.443
2.402
2.383
2.341
1.164
1.022
OH
O
O
1.00
0.86
0.96
0.95
1.74
2.60
0.90
0.78
1.01
2.07
2.10
3.23
3.13
f1 (ppm)

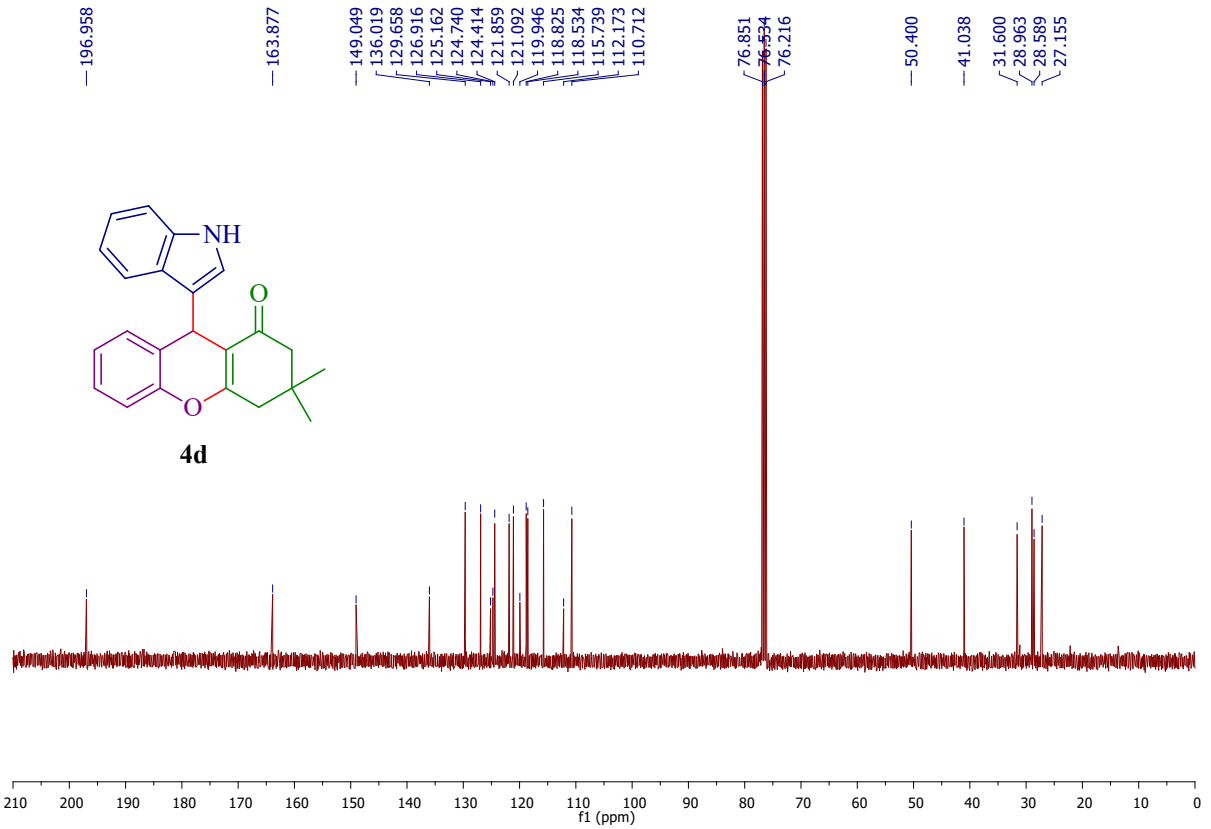
4c


200.801
166.976
153.531
145.070
133.316
132.930
130.018
127.954
127.827
127.273
126.647
125.290
123.357
121.237
120.876
119.412
118.738
114.077
77.350
77.032
76.715
50.295
41.769
32.388
31.008
29.006
27.396
OH
O
O
f1 (ppm)

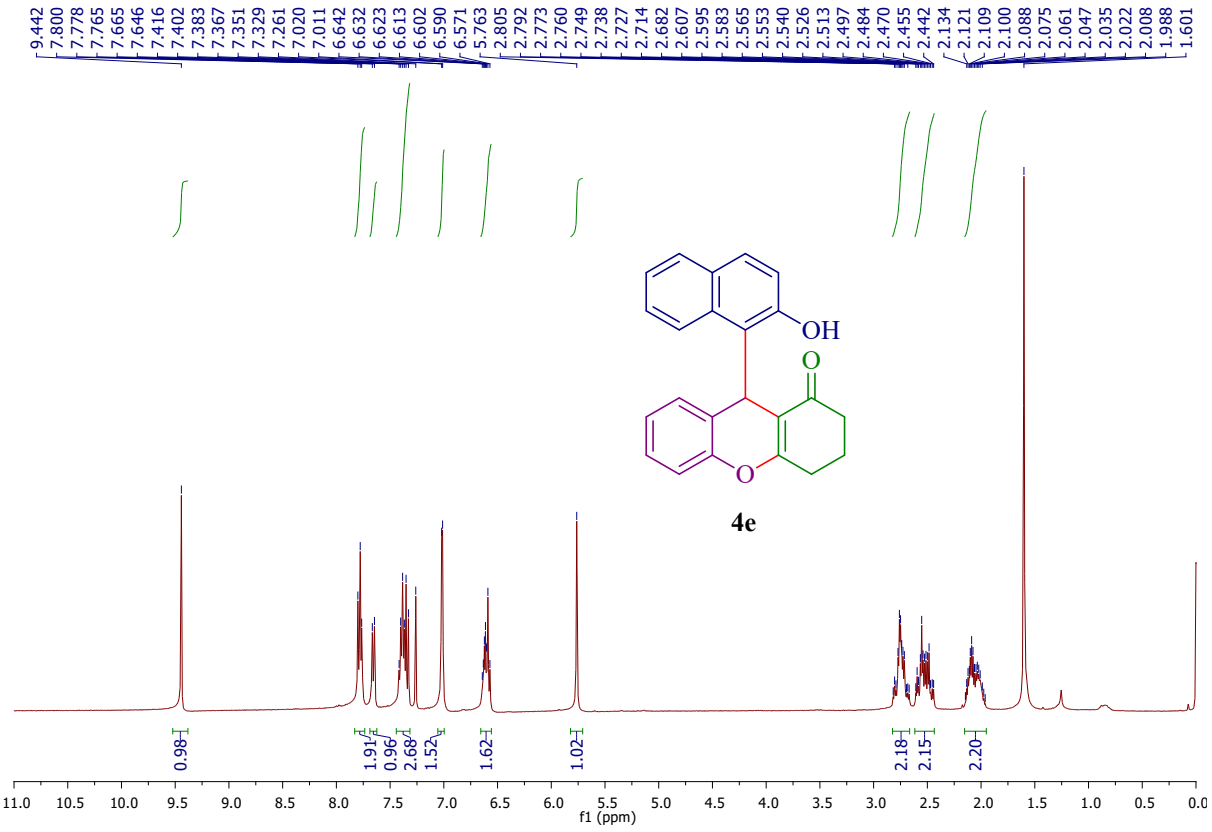
4c


8.214
7.589
7.475
7.462
7.454
7.381
7.375
7.368
7.365
7.296
7.278
7.276
7.185
7.183
7.168
5.510
2.827
2.784
2.758
2.713
2.473
2.432
2.403
2.362
1.307
1.160
NH
O
O
0.85
0.99
1.67
1.93
0.76
1.25
1.59
0.75
1.86
2.00
3.03
3.03
f1 (ppm)

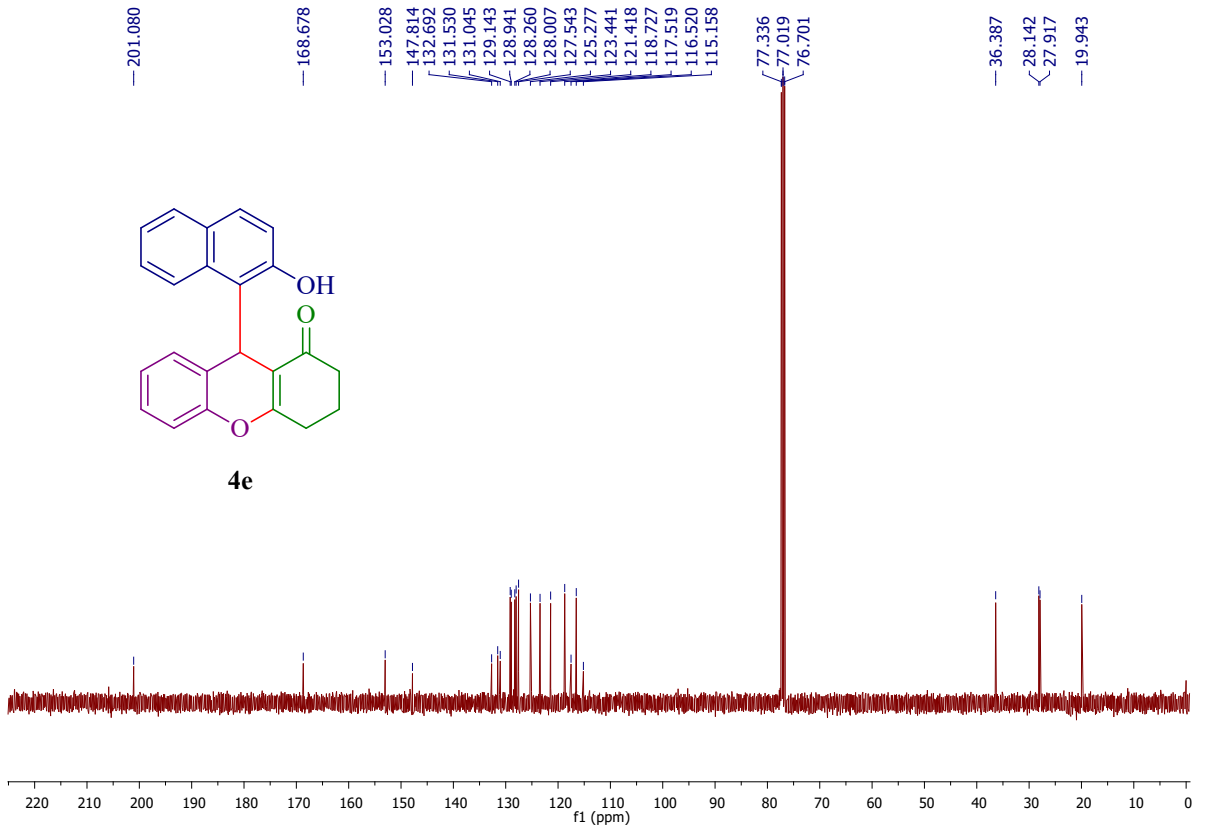
4d


196.958
163.877
149.049
136.019
129.658
126.916
125.162
124.740
124.414
121.859
121.092
119.946
118.825
118.534
115.739
112.173
110.712
76.851
76.534
76.216
50.400
41.038
31.600
28.963
28.589
27.155
NH
O
O
f1 (ppm)

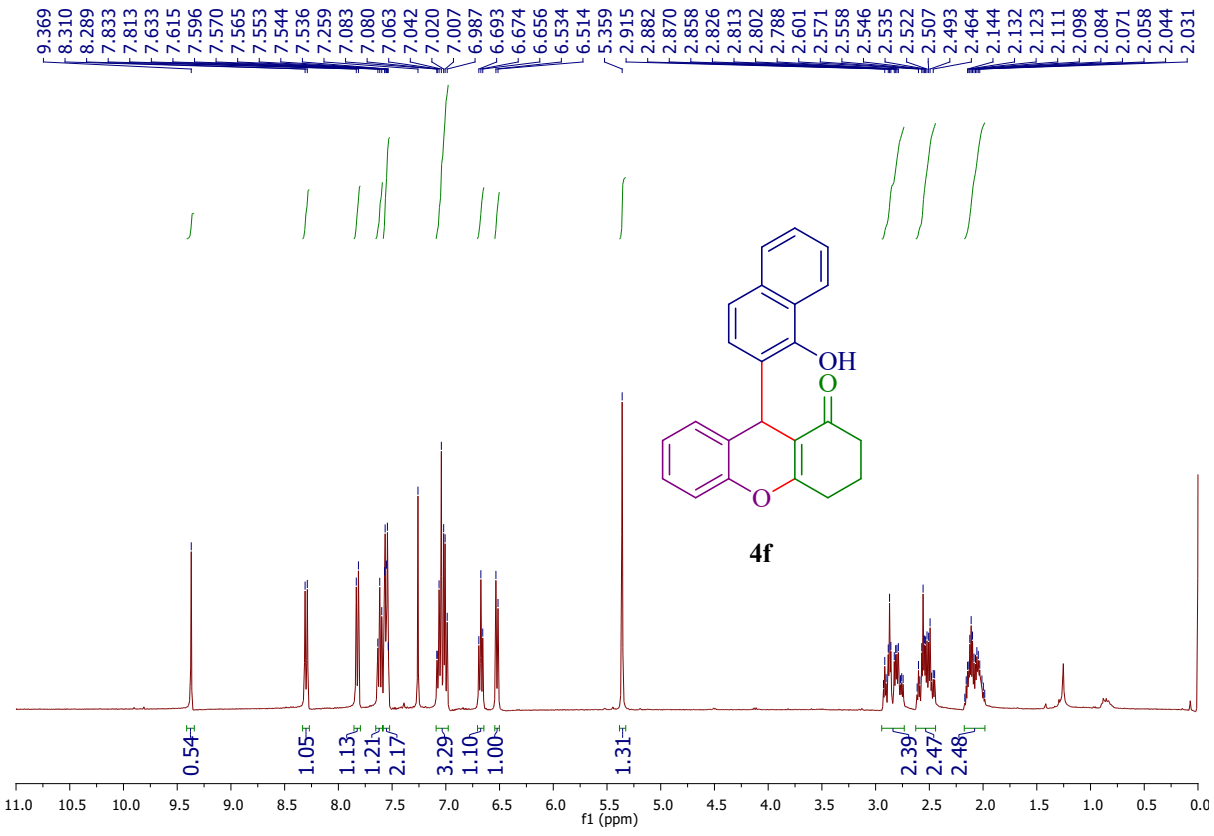
4d


9.442
7.800
7.778
7.765
7.665
7.646
7.416
7.402
7.383
7.367
7.351
7.329
7.261
7.020
7.011
6.642
6.632
6.623
6.613
6.602
6.590
6.571
5.763
2.805
2.792
2.773
2.760
2.749
2.738
2.727
2.714
2.682
2.607
2.595
2.583
2.565
2.553
2.540
2.526
2.513
2.497
2.484
2.470
2.455
2.442
2.134
2.121
2.109
2.100
2.088
2.075
2.061
2.047
2.035
2.022
2.008
1.988
1.601
OH
O
O
0.98
1.91
0.96
2.68
1.52
1.62
1.02
2.18
2.15
2.20
f1 (ppm)

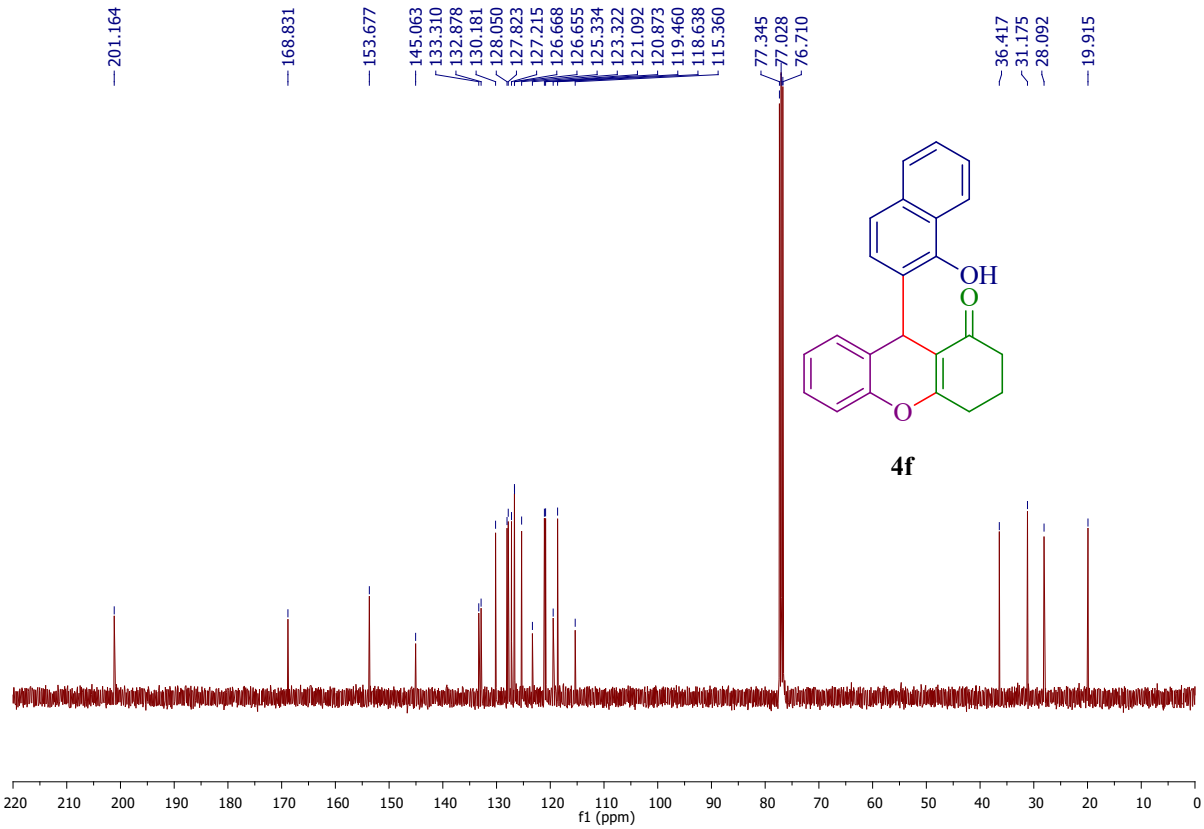
4e


201.080
168.678
153.028
147.814
132.692
131.530
131.045
129.143
128.941
128.260
128.007
127.543
125.277
123.441
121.418
118.727
117.519
116.520
115.158
77.336
77.019
76.701
36.387
28.142
27.917
19.943
OH
O
O
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)

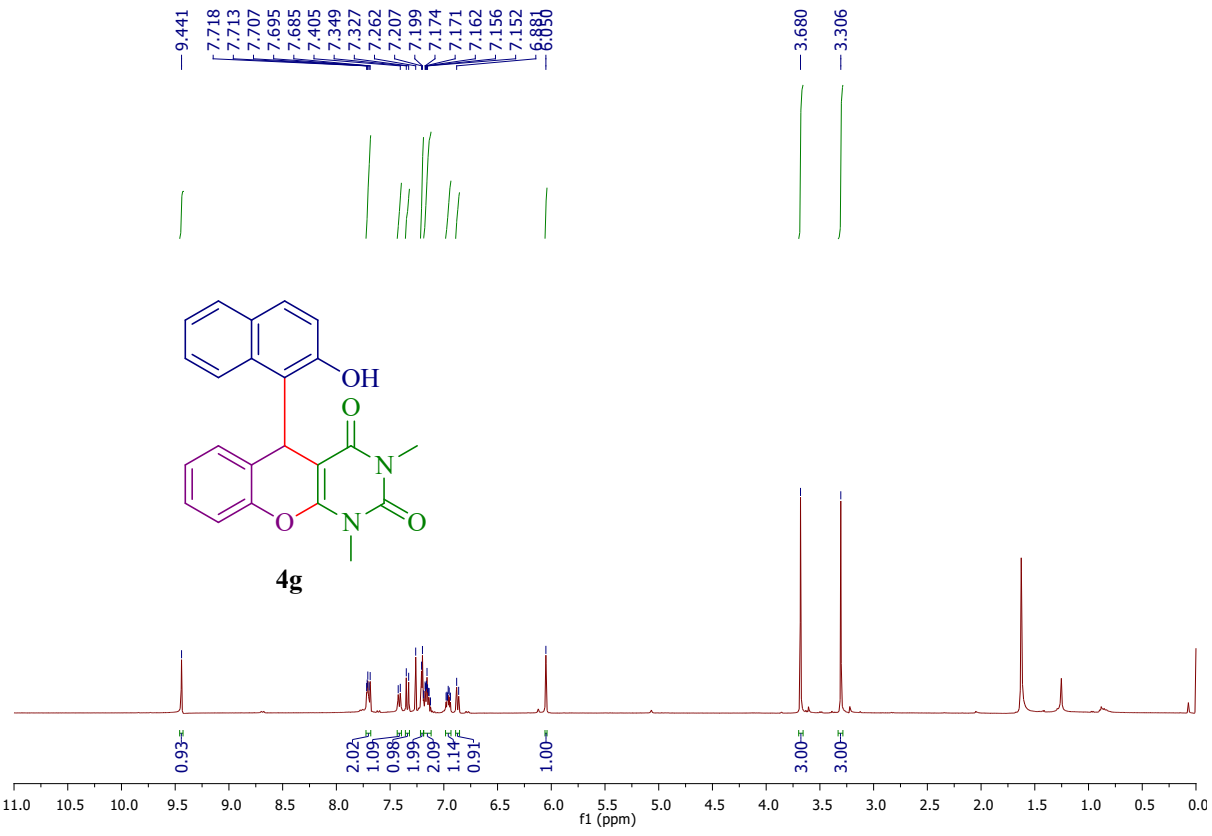
4e


9.369
8.310
8.289
7.833
7.813
7.633
7.615
7.596
7.570
7.565
7.553
7.544
7.536
7.259
7.083
7.080
7.063
7.042
7.020
7.007
6.987
6.693
6.674
6.656
6.534
6.514
5.359
2.915
2.882
2.870
2.858
2.826
2.813
2.802
2.788
2.601
2.571
2.558
2.546
2.535
2.522
2.507
2.493
2.464
2.144
2.132
2.123
2.111
2.098
2.084
2.071
2.058
2.044
2.031
OH
O
O
0.54
1.05
1.13
1.21
2.17
3.29
1.10
1.00
1.31
2.39
2.47
2.48
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)

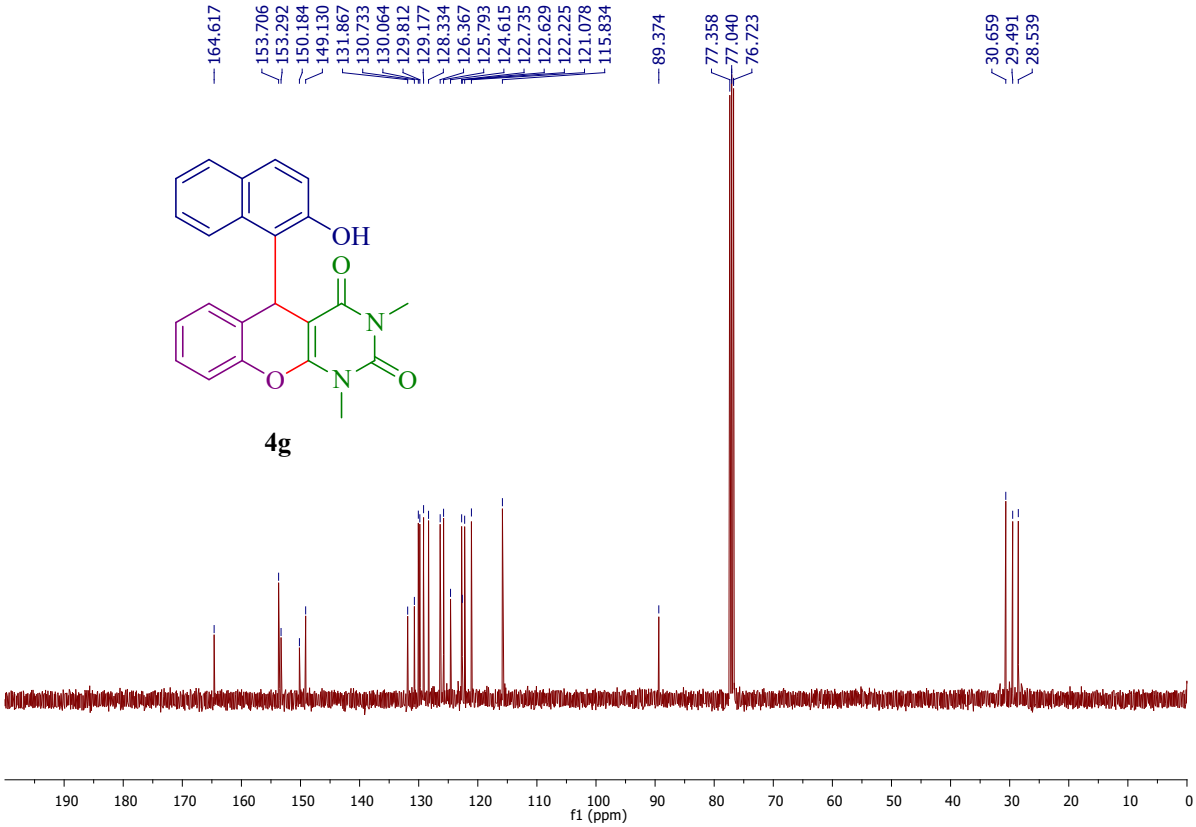
4f


201.164
168.831
153.677
145.063
133.310
132.878
130.181
128.050
127.823
127.215
126.668
126.655
125.334
123.322
121.092
120.873
119.460
118.638
115.360
77.345
77.028
76.710
36.417
31.175
28.092
19.915
OH
O
O
f1 (ppm)

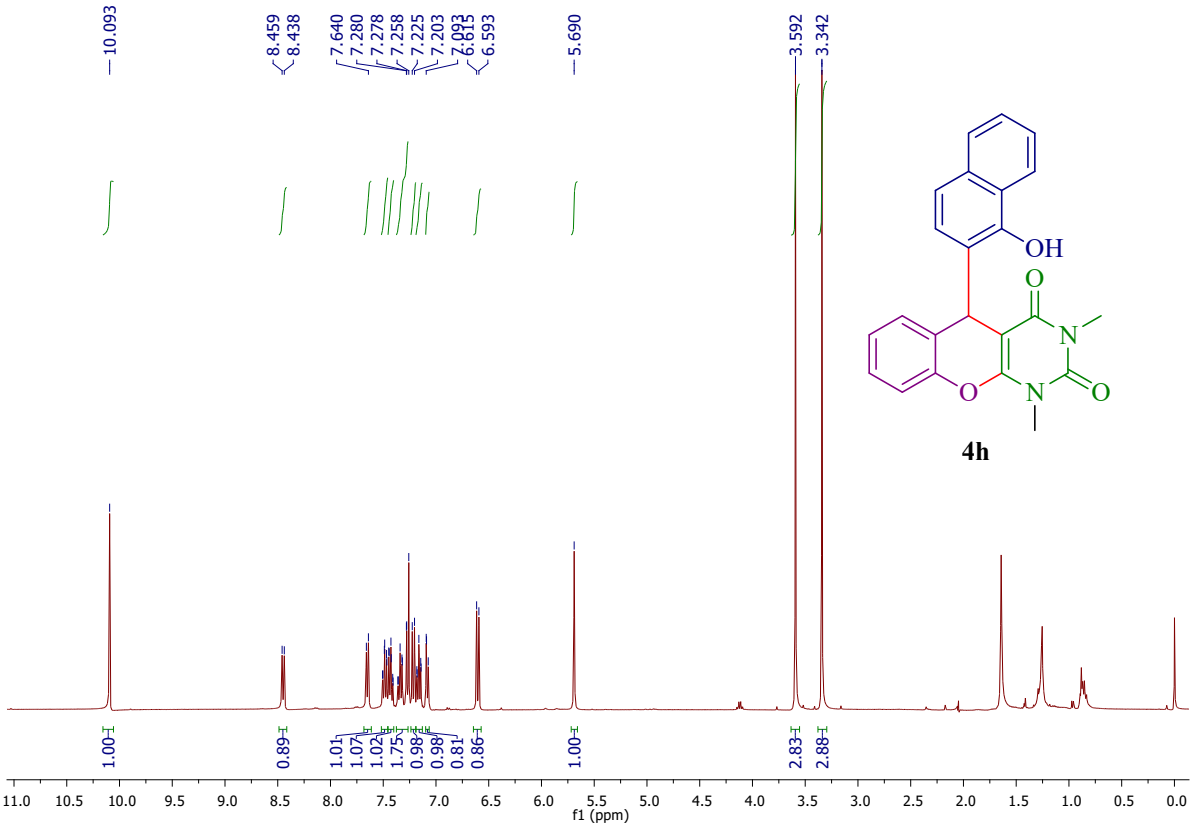
4f


9.441
7.718
7.713
7.707
7.695
7.685
7.405
7.349
7.327
7.262
7.207
7.199
7.174
7.171
7.162
7.156
7.152
6.881
6.850
3.680
3.306
OH
O
N
O
N
O
0.93
2.02
1.09
0.98
1.99
2.09
1.14
0.91
1.00
3.00
3.00
f1 (ppm)

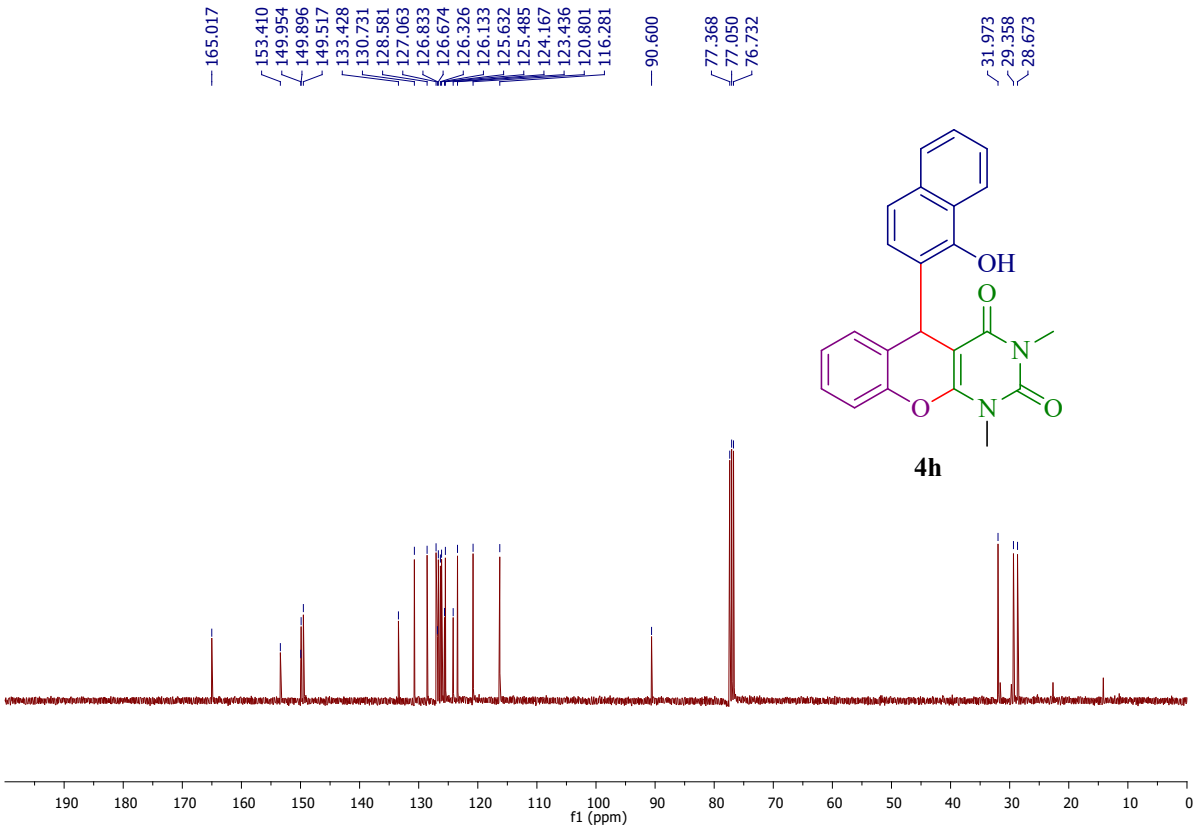
4g

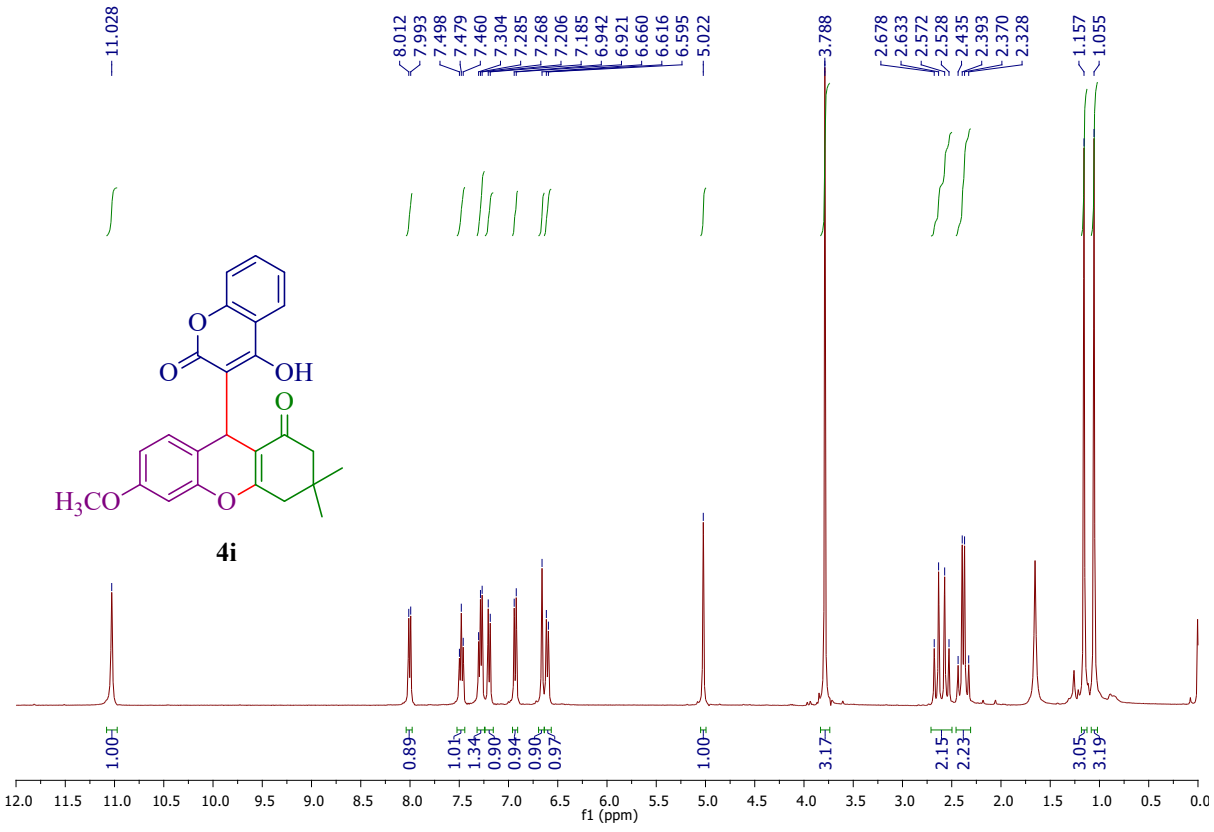

164.617
153.706
153.292
150.184
149.130
131.867
130.733
130.064
129.812
129.177
128.334
126.367
125.793
124.615
122.735
122.629
122.225
121.078
115.834
89.374
77.358
77.040
76.723
30.659
29.491
28.539
OH
O
N
O
N
O
f1 (ppm)

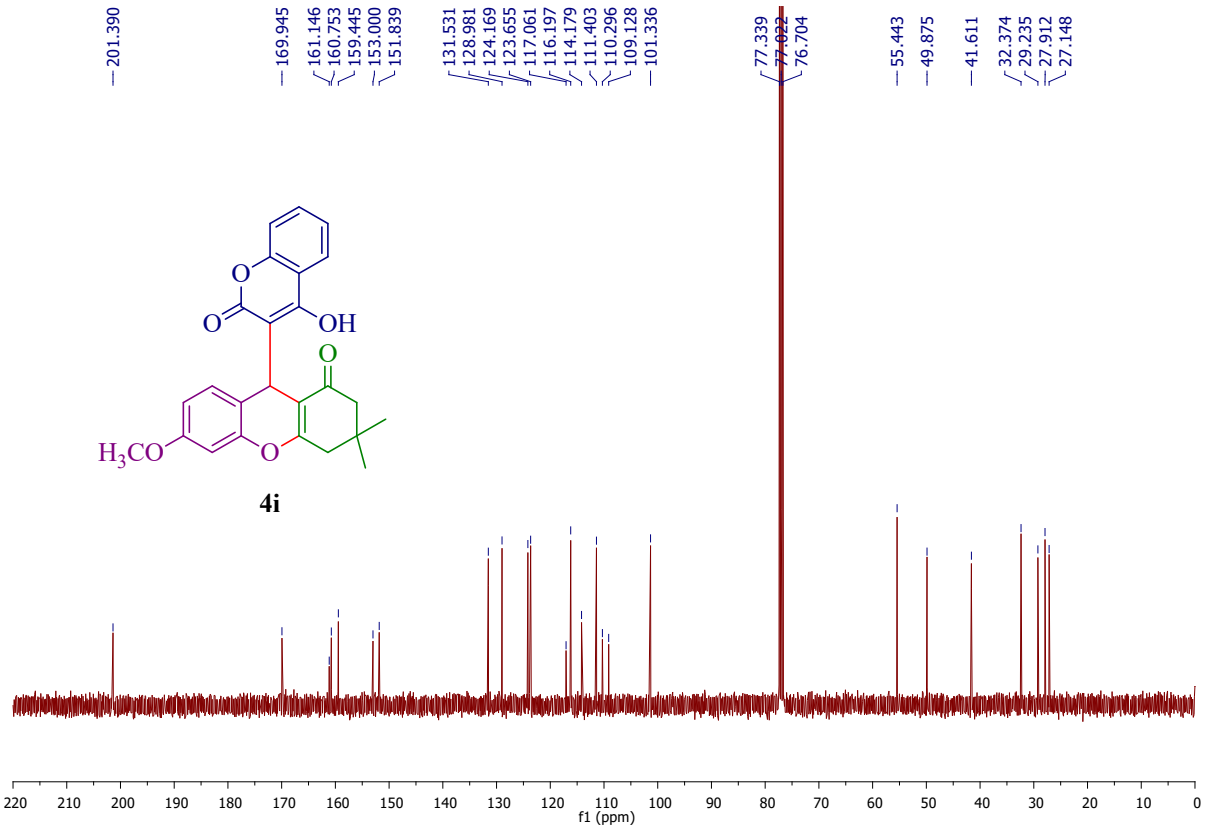
4g


10.093
8.459
8.438
7.640
7.280
7.278
7.258
7.225
7.203
7.093
6.615
6.593
5.690
3.592
3.342
OH
O
N
O
N
O
1.00
0.89
1.01
1.07
1.02
1.75
0.98
0.98
0.81
0.86
1.00
2.83
2.88
f1 (ppm)

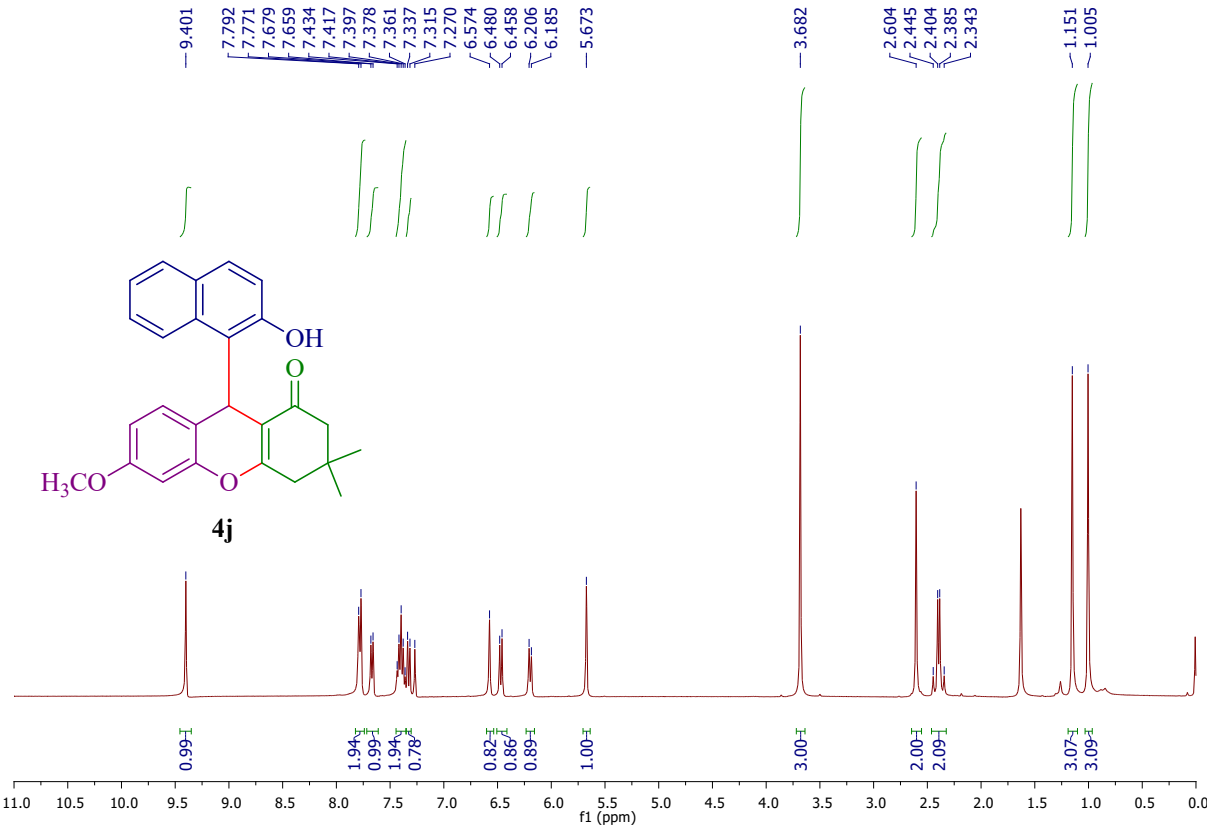
4h


165.017
153.410
149.954
149.896
149.517
133.428
130.731
128.581
127.063
126.833
126.674
126.326
126.133
125.632
125.485
124.167
123.436
120.801
116.281
90.600
77.368
77.050
76.732
31.973
29.358
28.673
OH
O
N
O
N
O
4h
f1 (ppm)

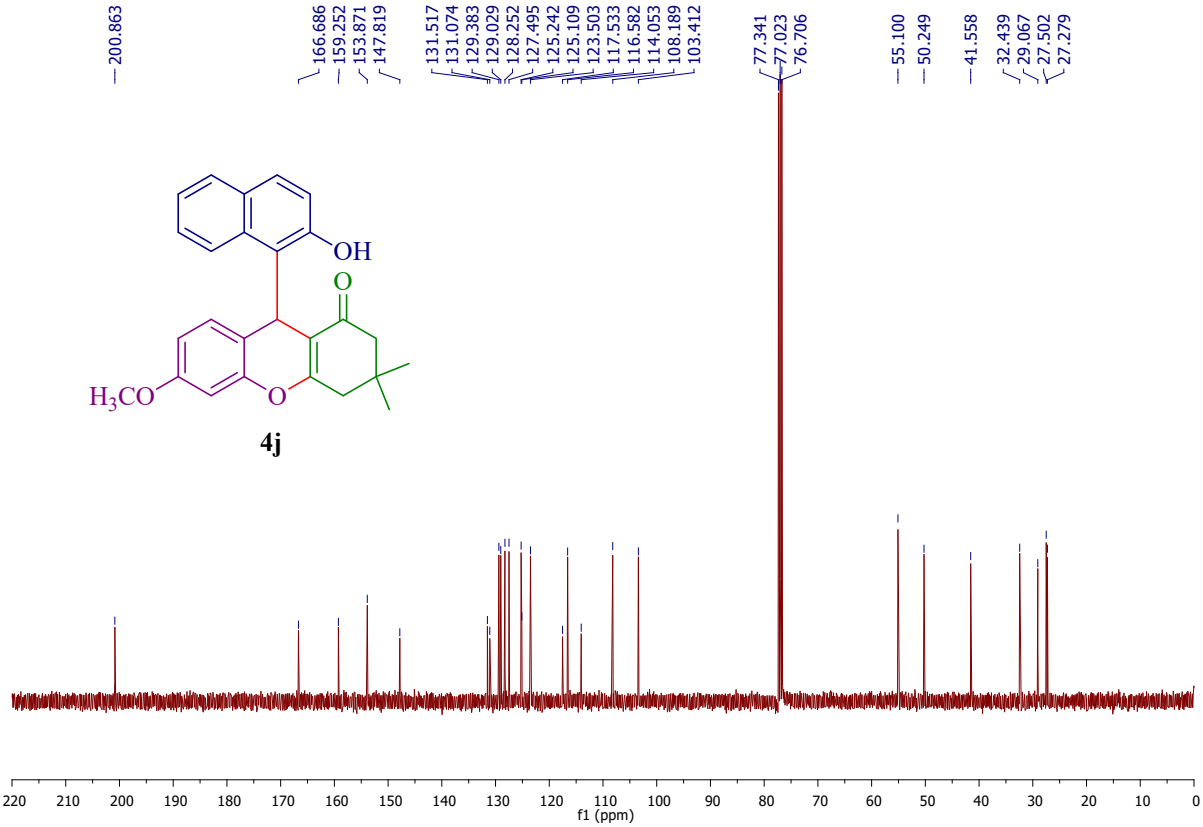

11.028
8.012
7.993
7.498
7.479
7.460
7.304
7.285
7.268
7.206
7.185
6.942
6.921
6.660
6.616
6.595
5.022
3.788
2.678
2.633
2.572
2.528
2.435
2.393
2.370
2.328
1.157
1.055
O
O
OH
O
H3CO
O
4i
1.00
0.89
1.01
1.34
0.90
0.94
0.90
0.97
1.00
3.17
2.15
2.23
3.05
3.19
f1 (ppm)


201.390
169.945
161.146
160.753
159.445
153.000
151.839
131.531
128.981
124.169
123.655
117.061
116.197
114.179
111.403
110.296
109.128
101.336
77.339
77.022
76.704
55.443
49.875
41.611
32.374
29.235
27.912
27.148
O
O
OH
O
H3CO
O
f1 (ppm)

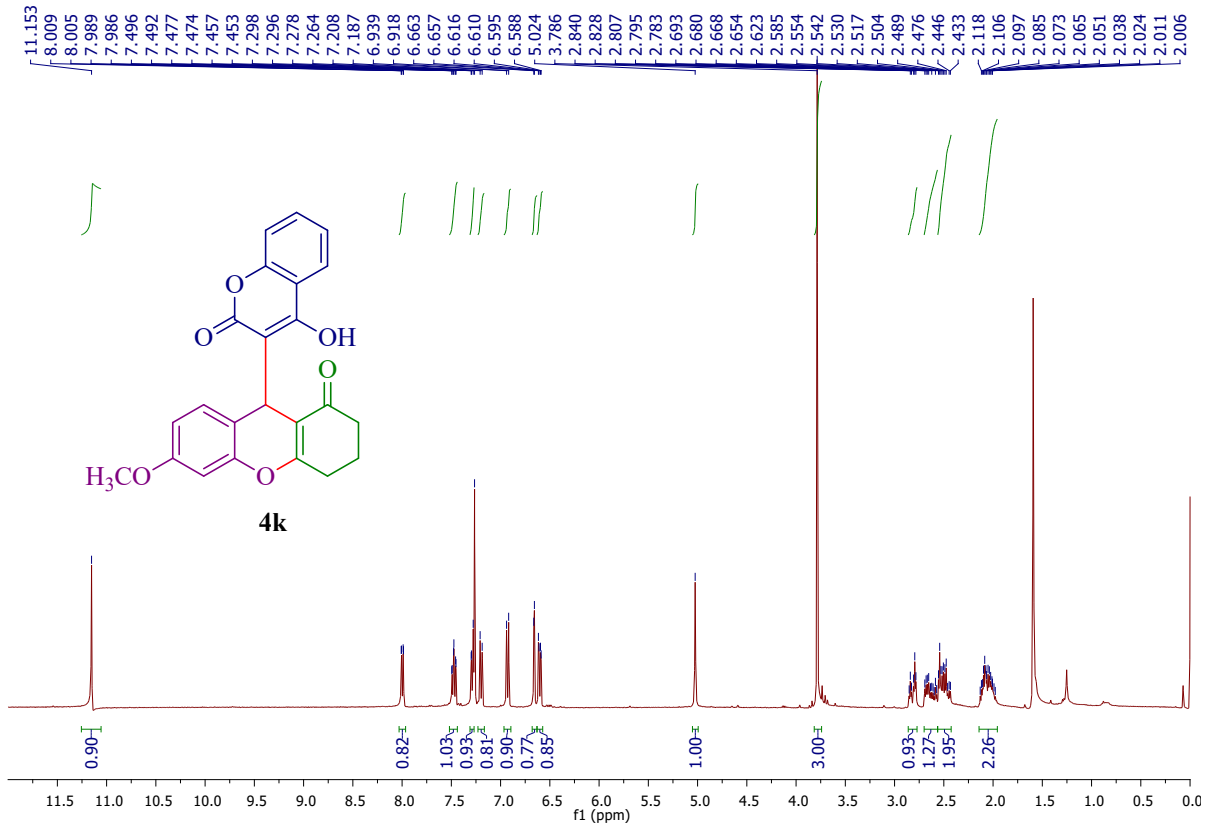
4i


9.401
7.792
7.771
7.679
7.659
7.434
7.417
7.397
7.378
7.361
7.337
7.315
7.270
6.574
6.480
6.458
6.206
6.185
5.673
3.682
2.604
2.445
2.404
2.385
2.343
1.151
1.005
OH
O
H3CO
O
0.99
1.94
0.99
1.94
0.78
0.82
0.86
0.89
1.00
3.00
2.00
2.09
3.07
3.09
f1 (ppm)

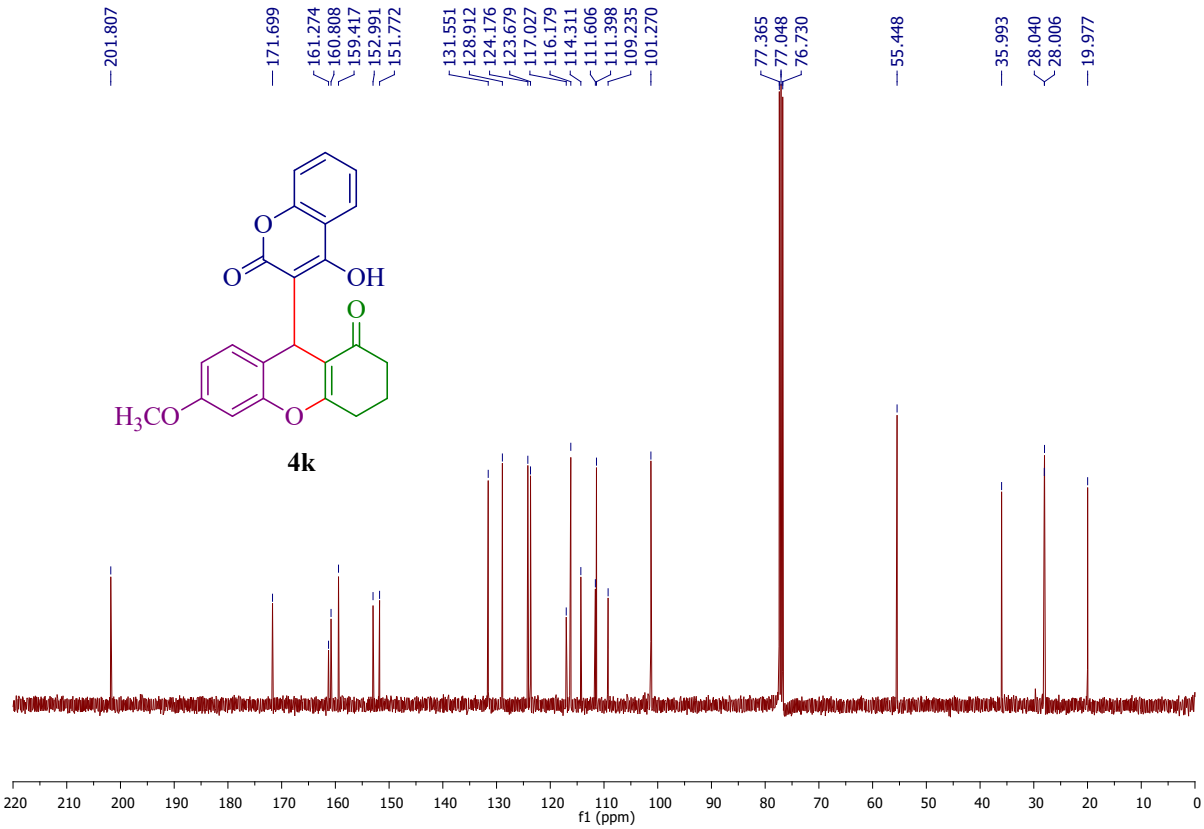
4j

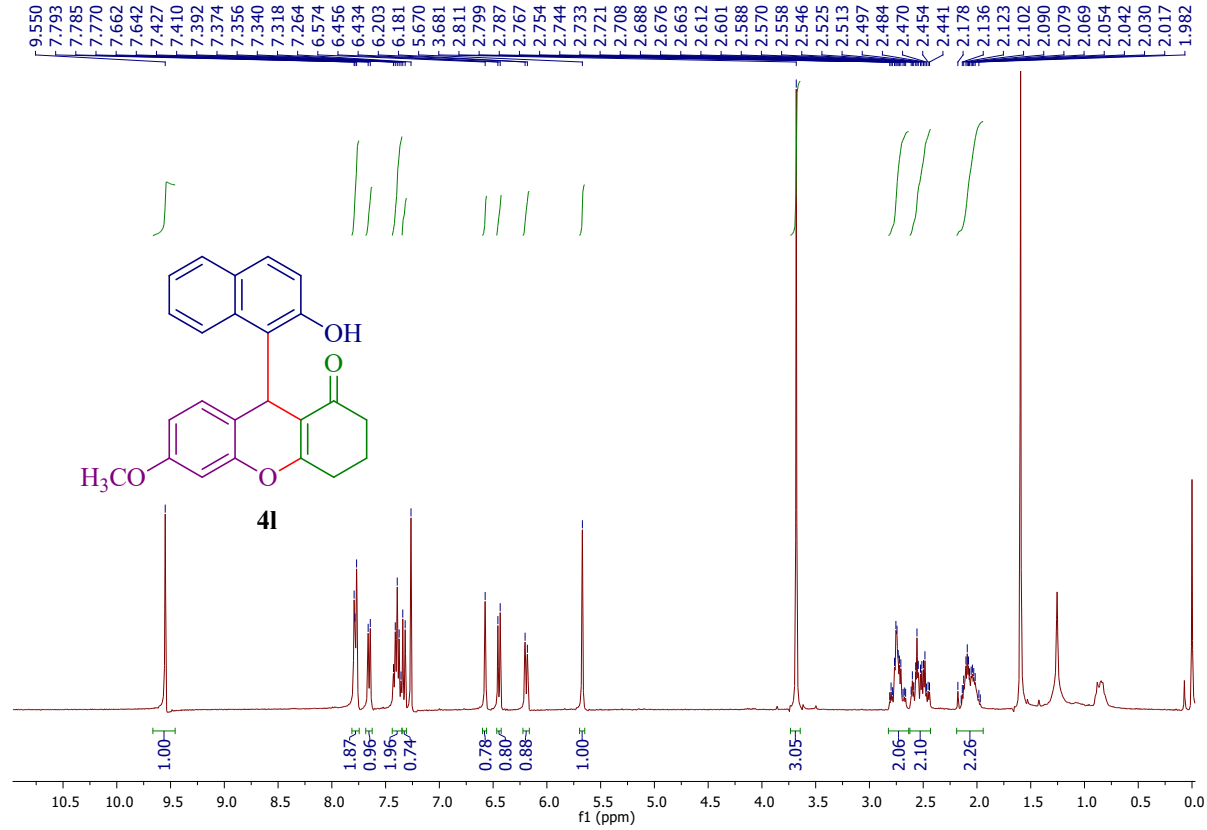

200.863
166.686
159.252
153.871
147.819
131.517
131.074
129.383
129.029
128.252
127.495
125.242
125.109
123.503
117.533
116.582
114.053
108.189
103.412
77.341
77.023
76.706
55.100
50.249
41.558
32.439
29.067
27.502
27.279
OH
O
H3CO
O
f1 (ppm)

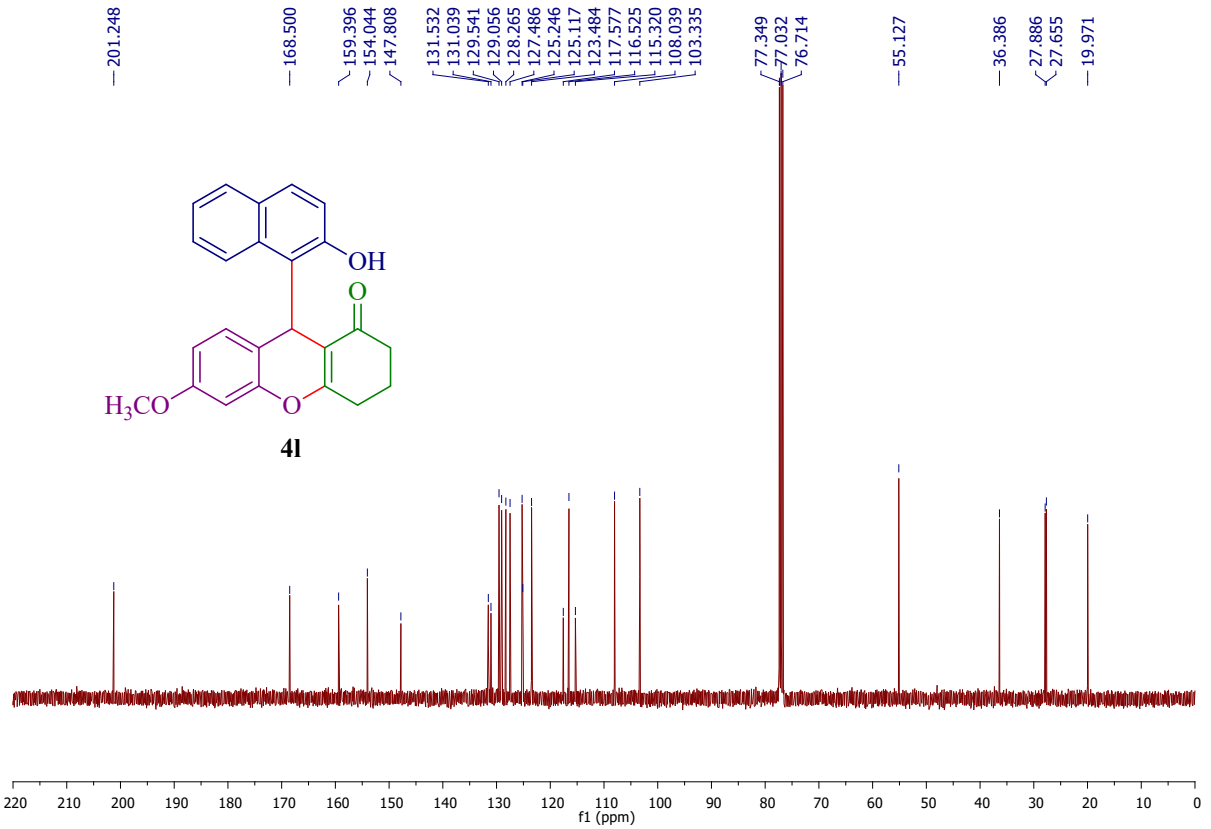
4j


11.153
8.009
8.005
7.989
7.986
7.496
7.492
7.477
7.474
7.457
7.453
7.298
7.296
7.278
7.264
7.208
7.187
6.939
6.918
6.663
6.657
6.616
6.610
6.595
6.588
5.024
3.786
2.840
2.828
2.807
2.795
2.783
2.693
2.680
2.668
2.654
2.623
2.585
2.554
2.542
2.530
2.517
2.504
2.489
2.476
2.446
2.433
2.118
2.106
2.097
2.085
2.073
2.065
2.051
2.038
2.024
2.011
2.006
O
O
OH
O
H3CO
O
0.90
0.82
1.03
0.93
0.81
0.90
0.77
0.85
1.00
3.00
0.93
1.27
1.95
2.26
f1 (ppm)

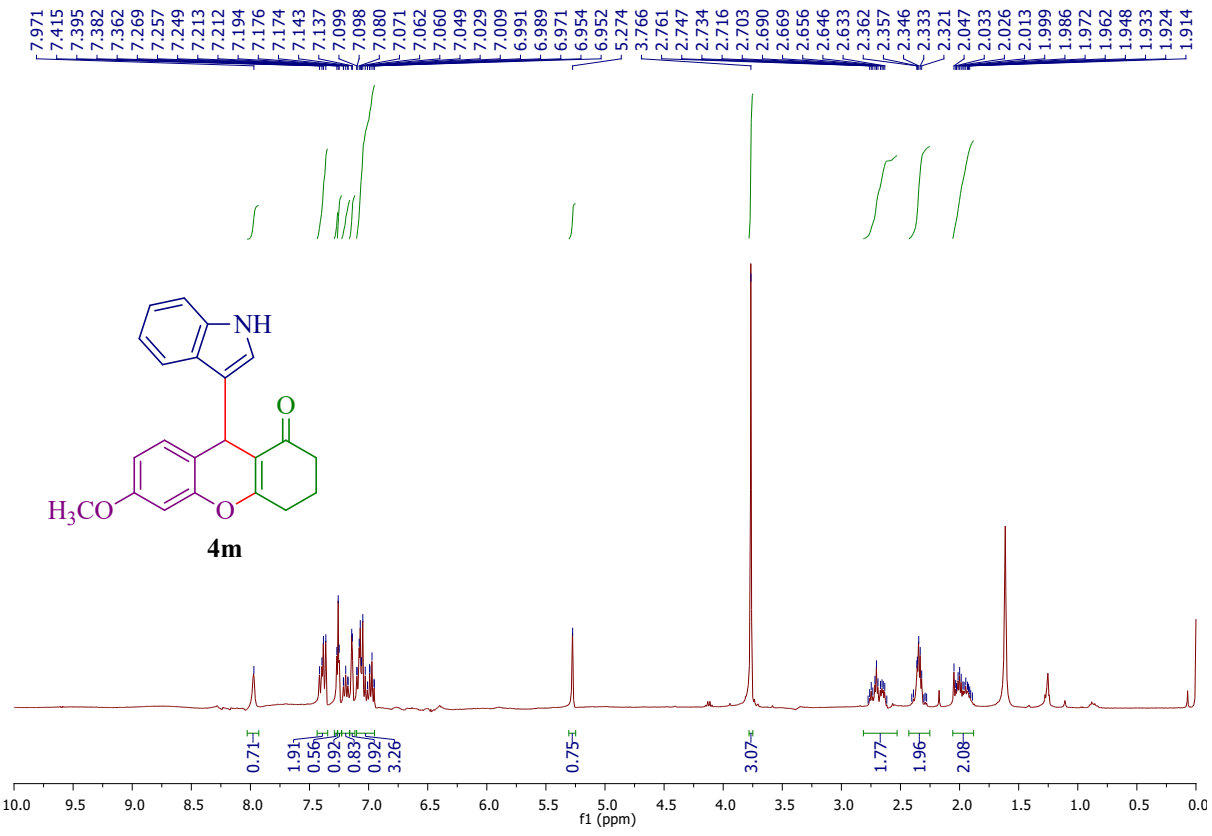
4k


201.807
171.699
161.274
160.808
159.417
152.991
151.772
131.551
128.912
124.176
123.679
117.027
116.179
114.311
111.606
111.398
109.235
101.270
77.365
77.048
76.730
55.448
35.993
28.040
28.006
19.977
O
O
OH
O
H3CO
O
4k
f1 (ppm)

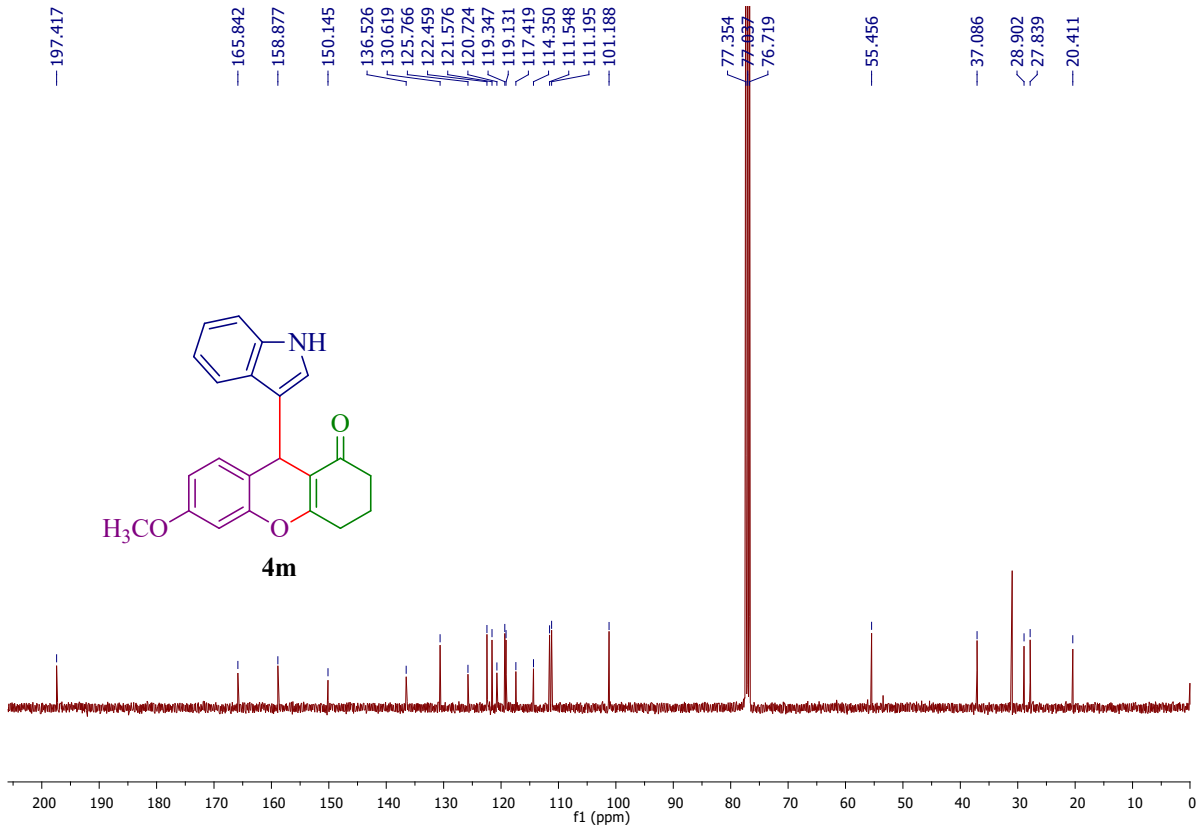

9.550
7.793
7.785
7.770
7.662
7.642
7.427
7.410
7.392
7.374
7.356
7.340
7.318
7.264
6.574
6.456
6.434
6.203
6.181
5.670
3.681
2.811
2.799
2.787
2.767
2.754
2.744
2.733
2.721
2.708
2.688
2.676
2.663
2.612
2.601
2.588
2.570
2.558
2.546
2.525
2.513
2.497
2.484
2.470
2.454
2.441
2.178
2.136
2.123
2.102
2.090
2.079
2.069
2.054
2.042
2.030
2.017
1.982
OH
O
H3CO
O
4l
1.00
1.87
0.96
1.96
0.74
0.78
0.80
0.88
1.00
3.05
2.06
2.10
2.26
f1 (ppm)


201.248
168.500
159.396
154.044
147.808
131.532
131.039
129.541
129.056
128.265
127.486
125.246
125.117
123.484
117.577
116.525
115.320
108.039
103.335
77.349
77.032
76.714
55.127
36.386
27.886
27.655
19.971
OH
O
H3CO
O
f1 (ppm)

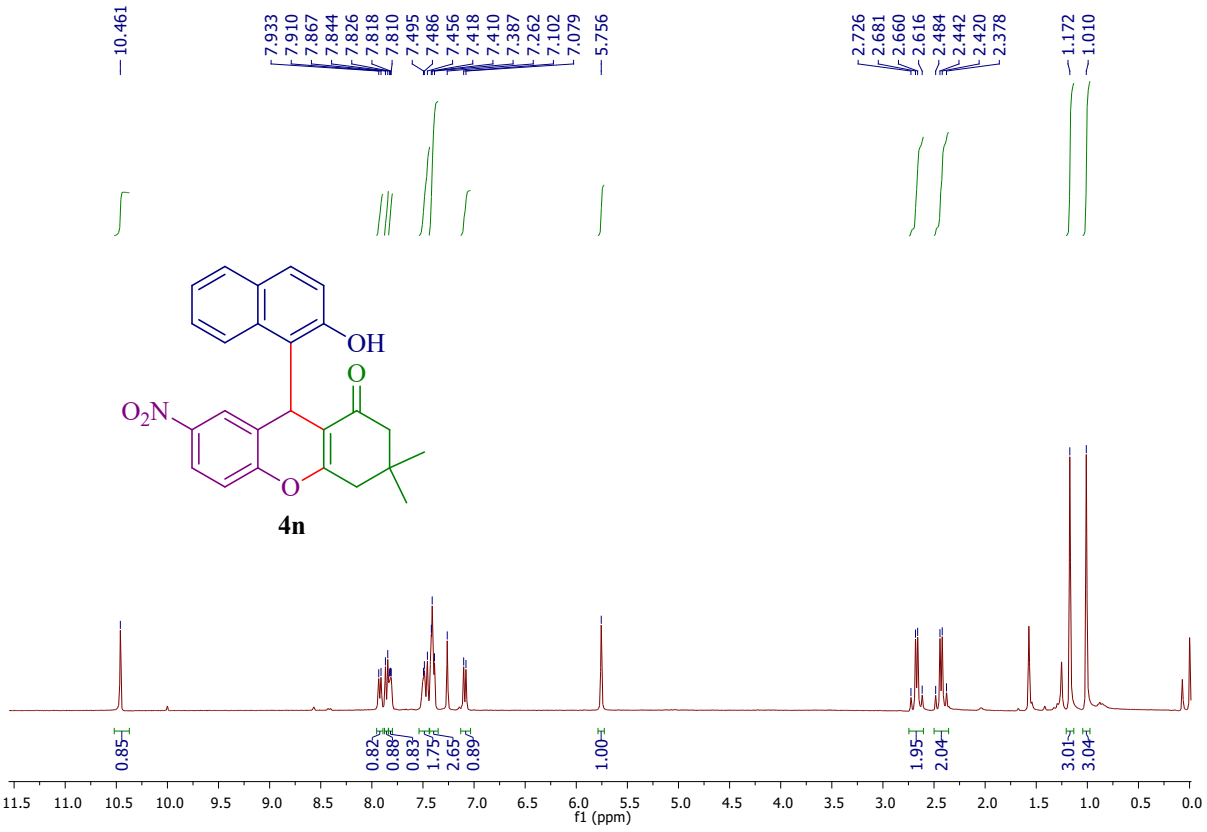
4l


7.971
7.415
7.395
7.382
7.362
7.269
7.257
7.249
7.213
7.212
7.194
7.176
7.174
7.143
7.137
7.099
7.098
7.080
7.071
7.062
7.060
7.049
7.029
7.009
6.991
6.989
6.971
6.954
6.952
5.274
3.766
2.761
2.747
2.734
2.716
2.703
2.690
2.669
2.656
2.646
2.633
2.362
2.357
2.346
2.333
2.321
2.047
2.033
2.026
2.013
1.999
1.986
1.972
1.962
1.948
1.933
1.924
1.914
NH
O
H3CO
O
0.71
1.91
0.56
0.92
0.83
0.92
3.26
0.75
3.07
1.77
1.96
2.08
f1 (ppm)

4m


197.417
165.842
158.877
150.145
136.526
130.619
125.766
122.459
121.576
120.724
119.347
119.131
117.419
114.350
111.548
111.195
101.188
77.354
77.037
76.719
55.456
37.086
28.902
27.839
20.411
NH
O
H_3CO
O
f1 (ppm)

4m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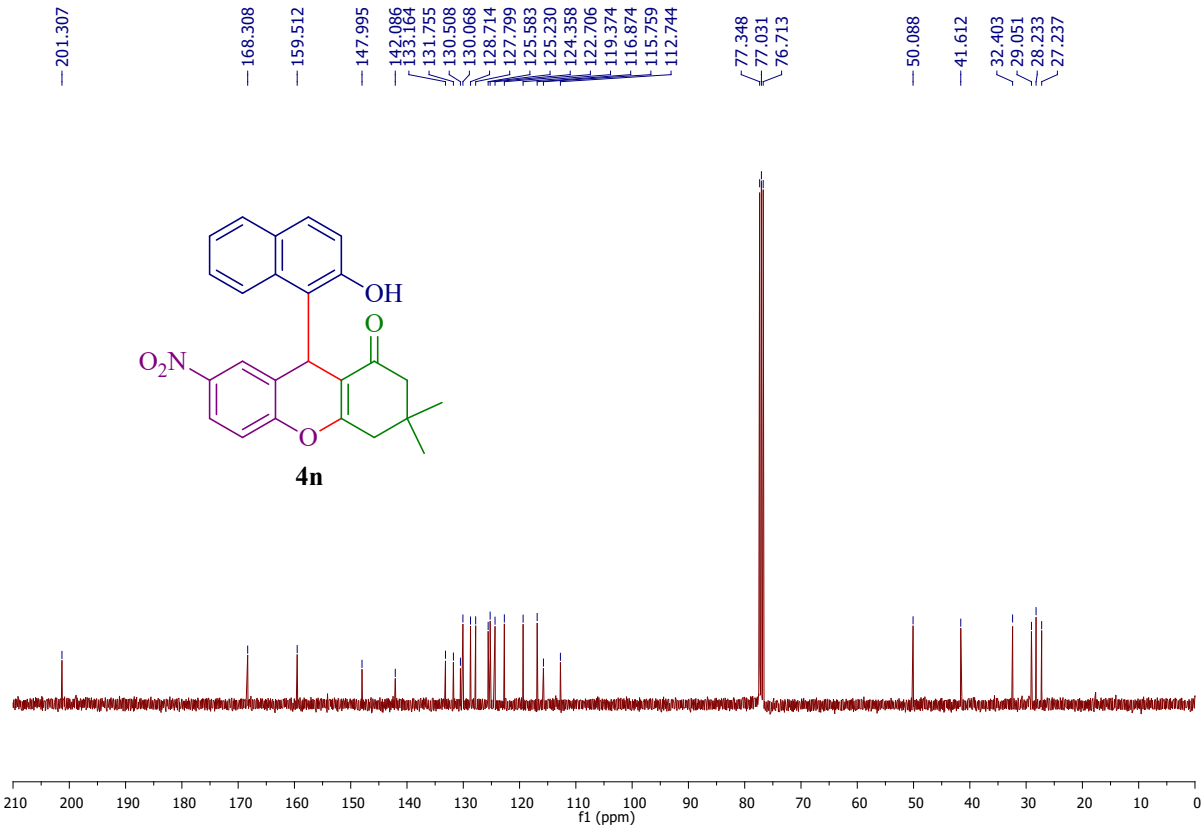
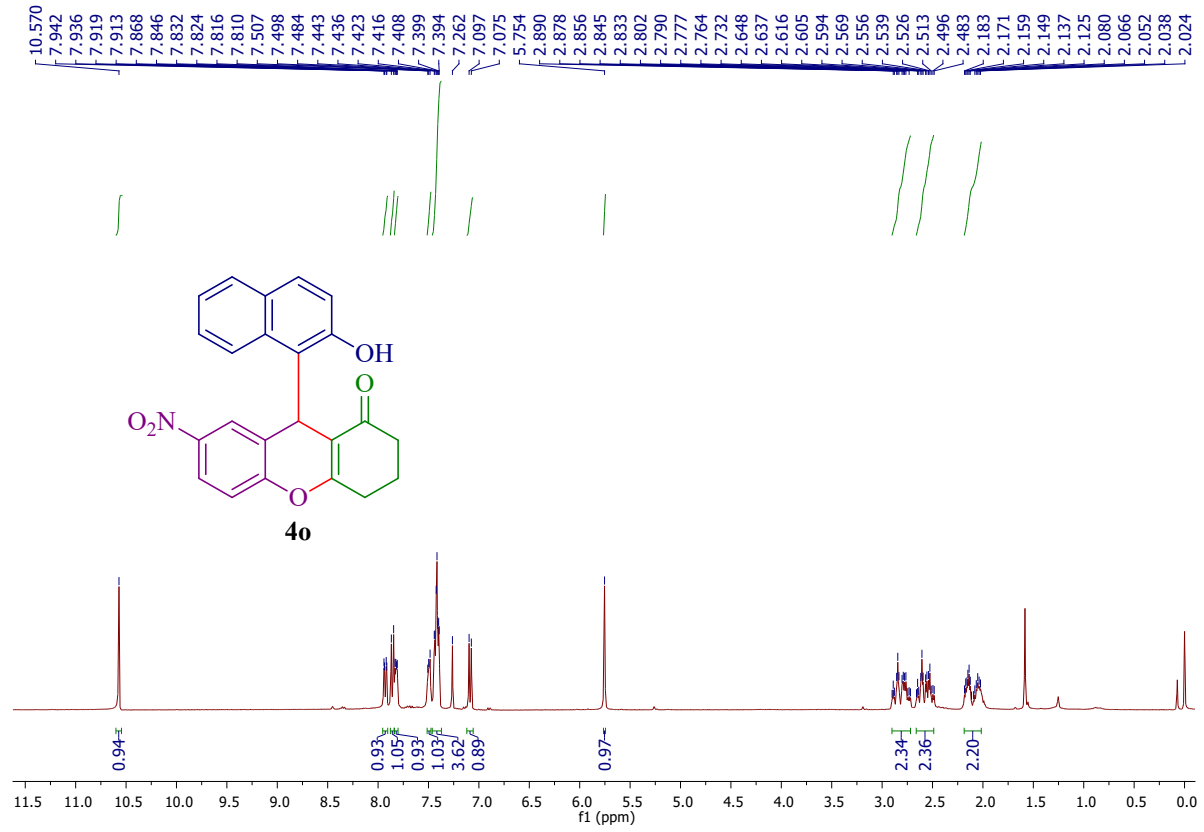
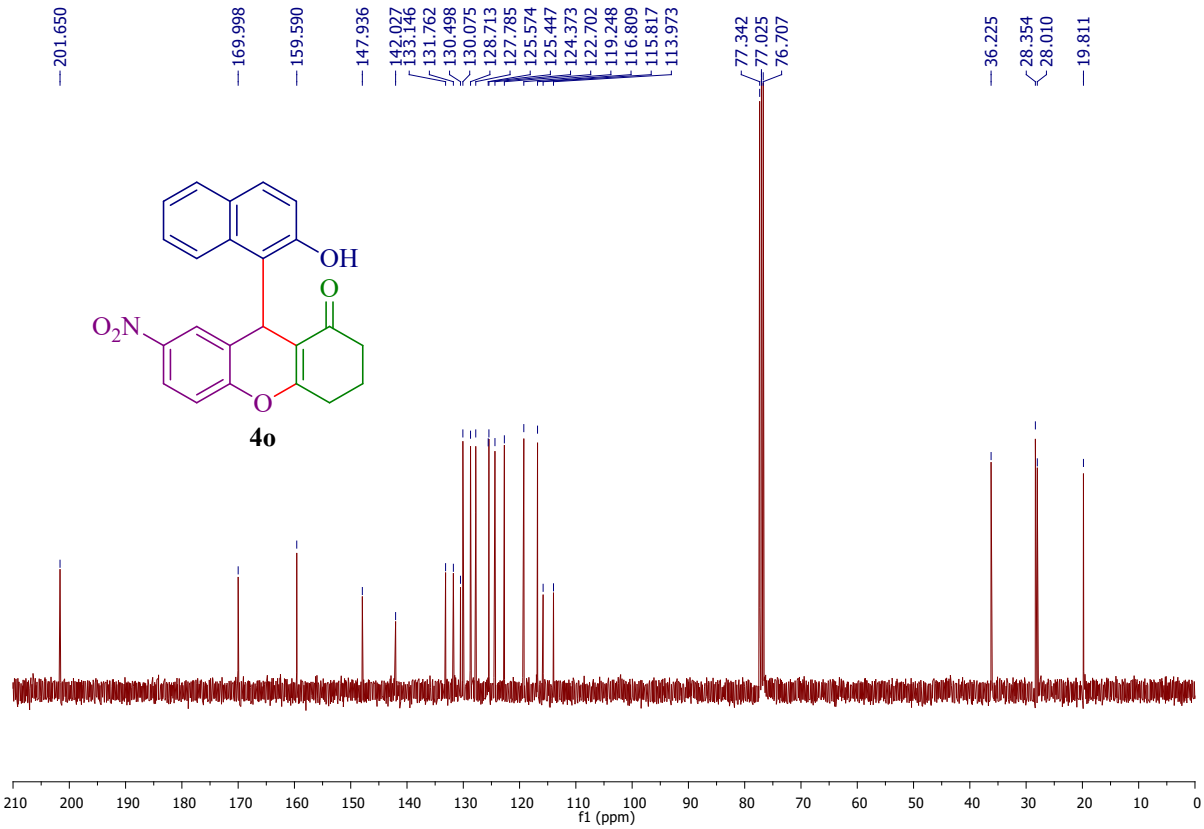
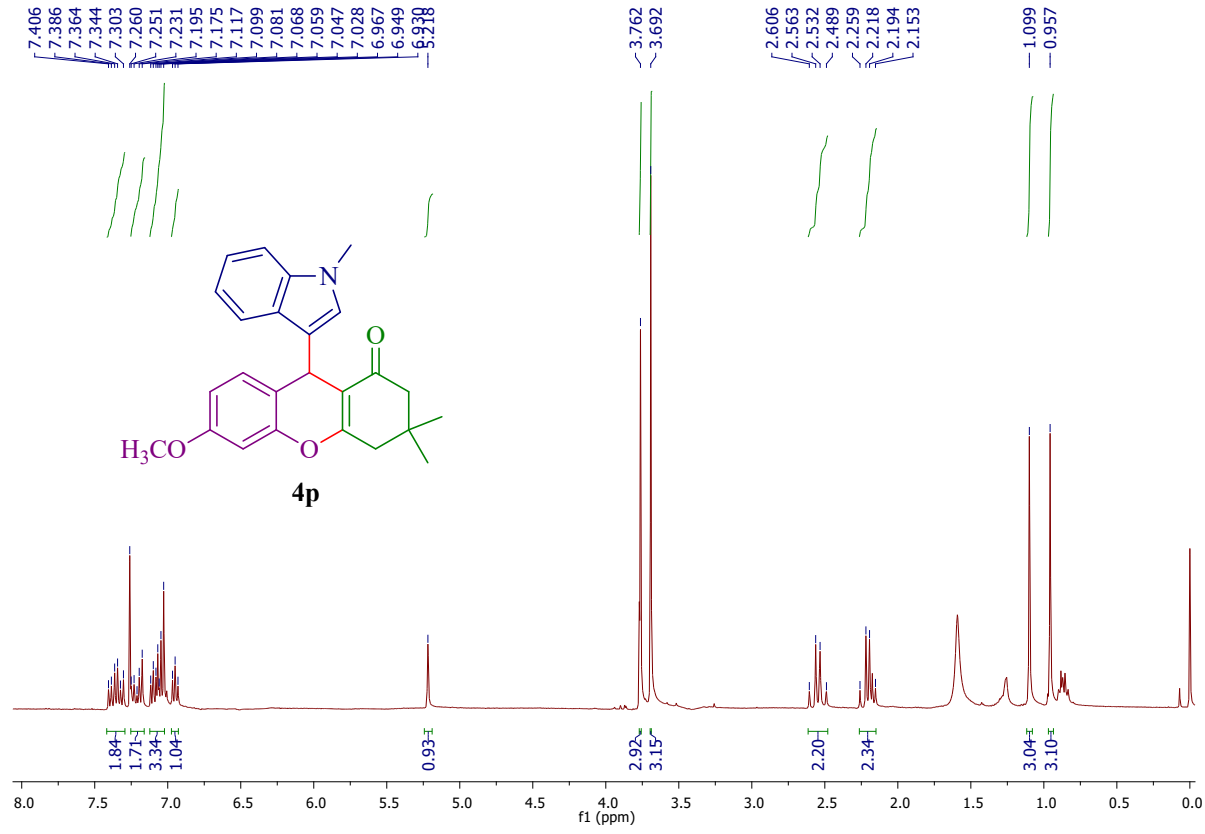
7.933
7.910
7.867
7.844
7.826
7.818
7.810
7.495
7.486
7.456
7.418
7.410
7.387
7.262
7.102
7.079
5.756
2.726
2.681
2.660
2.616
2.484
2.442
2.420
2.378
1.172
1.010
OH
O
O_2N
O
0.85
0.82
0.88
0.83
1.75
2.65
0.89
1.00
1.95
2.04
3.01
3.04
f1 (ppm)

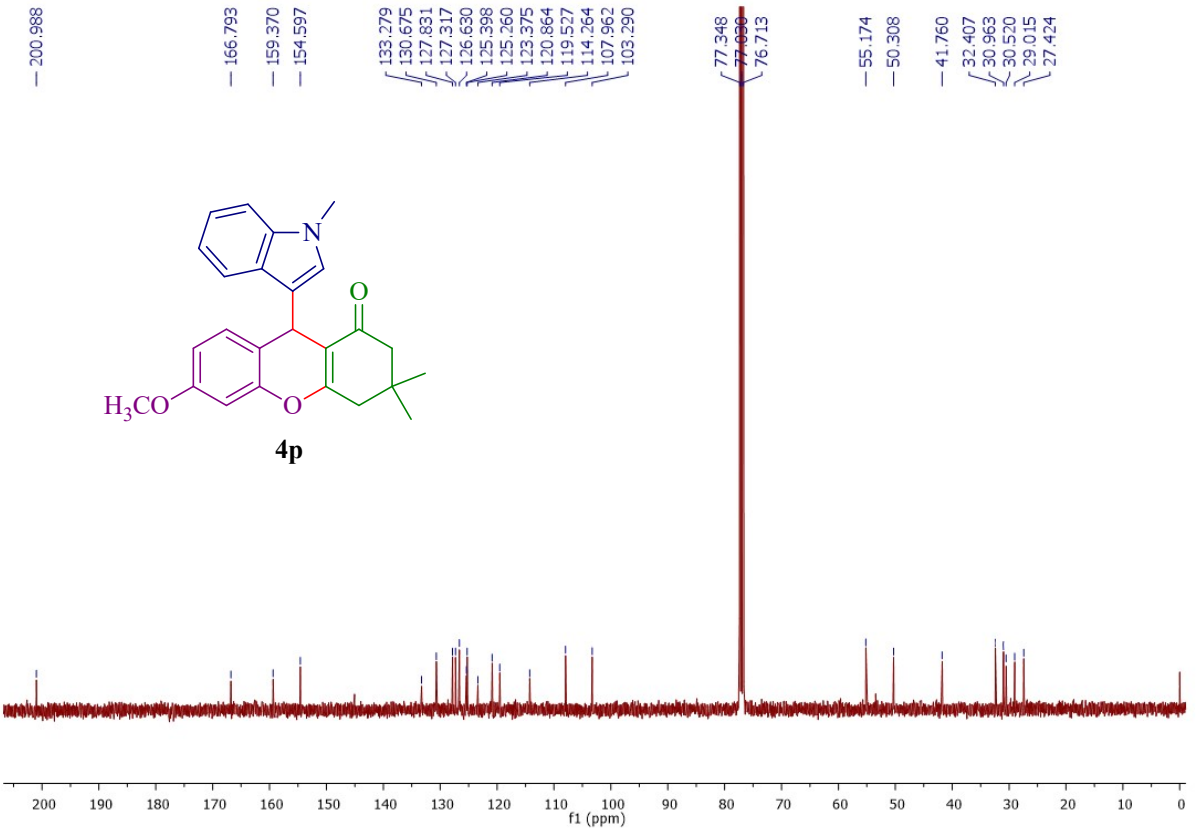
4n


201.307
168.308
159.512
147.995
142.086
133.164
131.755
130.508
130.068
128.714
127.799
125.583
125.230
124.358
122.706
119.374
116.874
115.759
112.744
77.348
77.031
76.713
50.088
41.612
32.403
29.051
28.233
27.237
OH
O
O2N
O
4n
f1 (ppm)

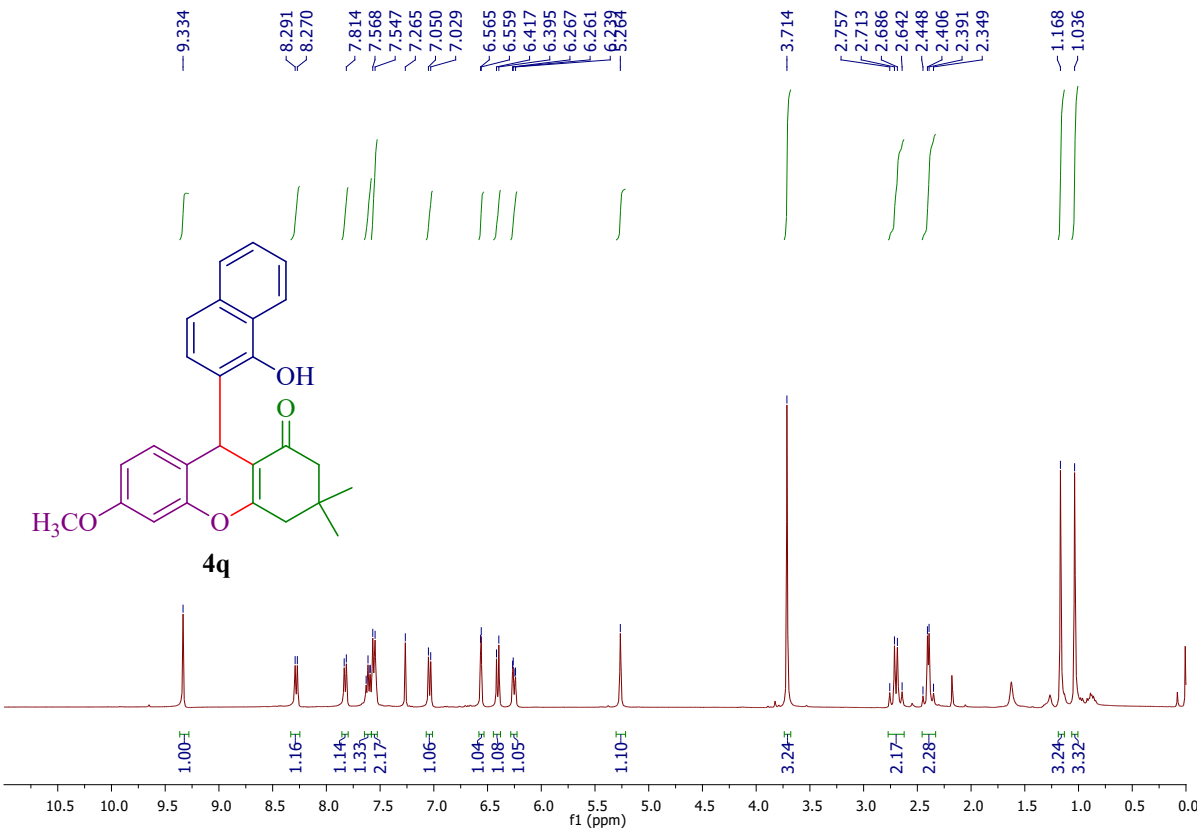

10.570
7.942
7.936
7.919
7.913
7.868
7.846
7.832
7.824
7.816
7.810
7.507
7.498
7.484
7.443
7.436
7.423
7.416
7.408
7.399
7.394
7.262
7.097
7.075
5.754
2.890
2.878
2.856
2.845
2.833
2.802
2.790
2.777
2.764
2.732
2.648
2.637
2.616
2.605
2.594
2.569
2.556
2.539
2.526
2.513
2.496
2.483
2.183
2.171
2.159
2.149
2.137
2.125
2.080
2.066
2.052
2.038
2.024
OH
O
O2N
O
4o
0.94
0.93
1.05
0.93
1.03
3.62
0.89
0.97
2.34
2.36
2.20
f1 (ppm)


201.650
169.998
159.590
147.936
142.027
133.146
131.762
130.498
130.075
128.713
127.785
125.574
125.447
124.373
122.702
119.248
116.809
115.817
113.973
77.342
77.025
76.707
36.225
28.354
28.010
19.811
OH
O
O_2N
O
4o
f1 (ppm)

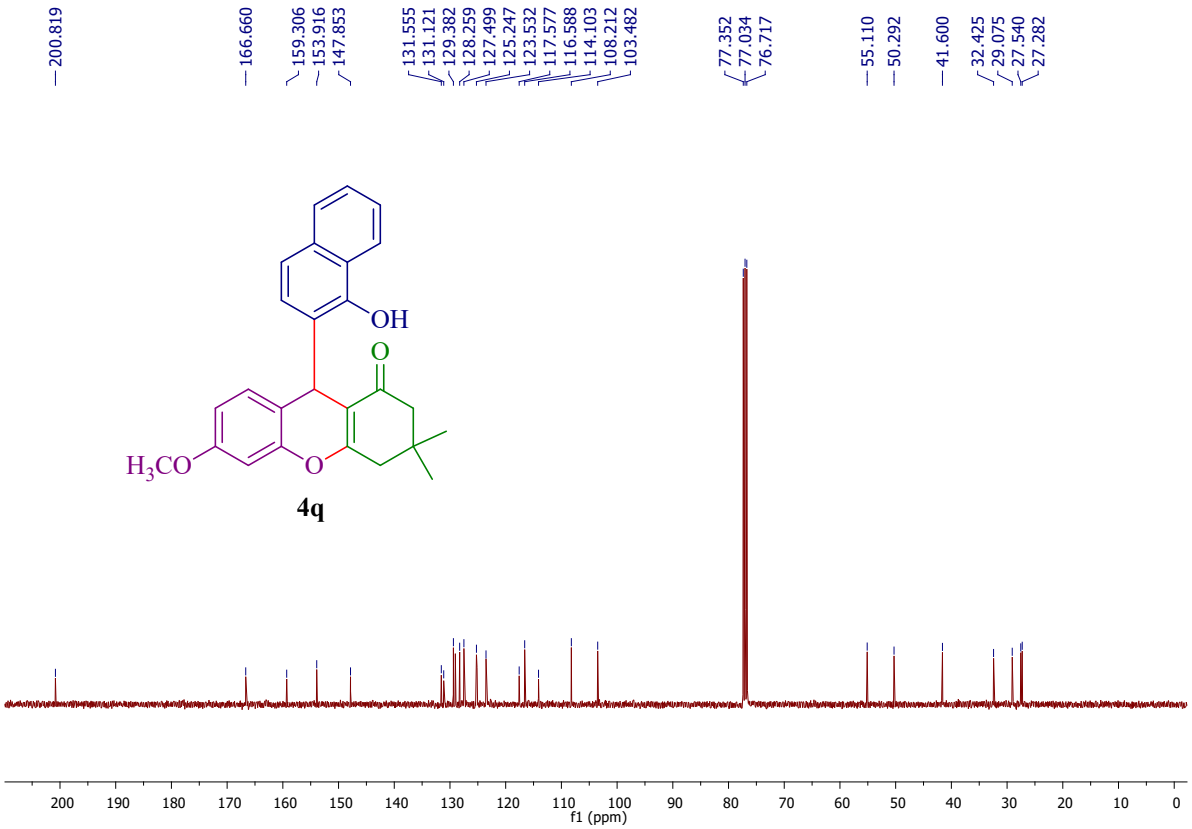

7.406
7.386
7.364
7.344
7.303
7.260
7.251
7.231
7.195
7.175
7.117
7.099
7.081
7.068
7.059
7.047
7.028
6.967
6.949
6.930
5.218
3.762
3.692
2.606
2.563
2.532
2.489
2.259
2.218
2.194
2.153
1.099
0.957
N
O
H_3CO
O
4p
1.84
1.71
3.34
1.04
0.93
2.92
3.15
2.20
2.34
3.04
3.10
f1 (ppm)

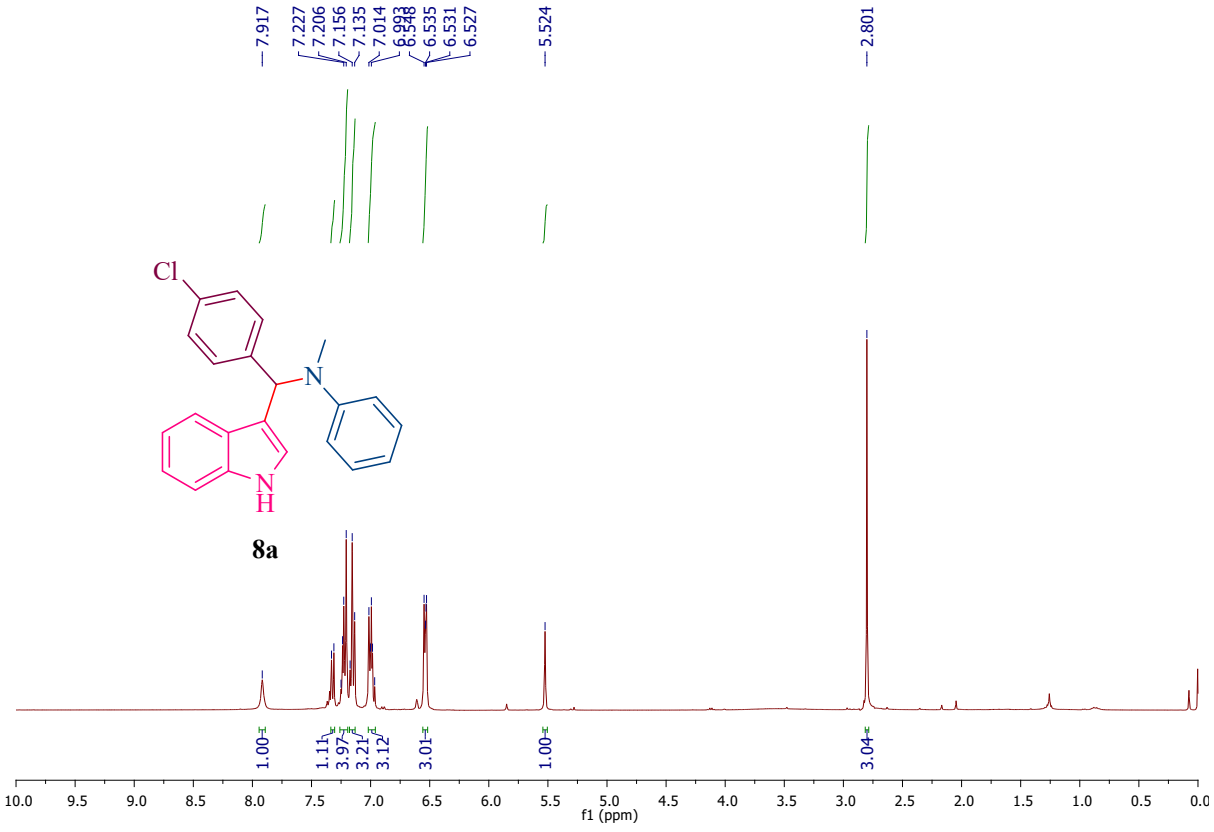

200.988
166.793
159.370
154.597
133.279
130.675
127.831
127.317
126.630
125.398
125.260
123.375
120.864
119.527
114.264
107.962
103.290
77.348
77.030
76.713
55.174
50.308
41.760
32.407
30.963
30.520
29.015
27.424
N
O
H3CO
O
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)

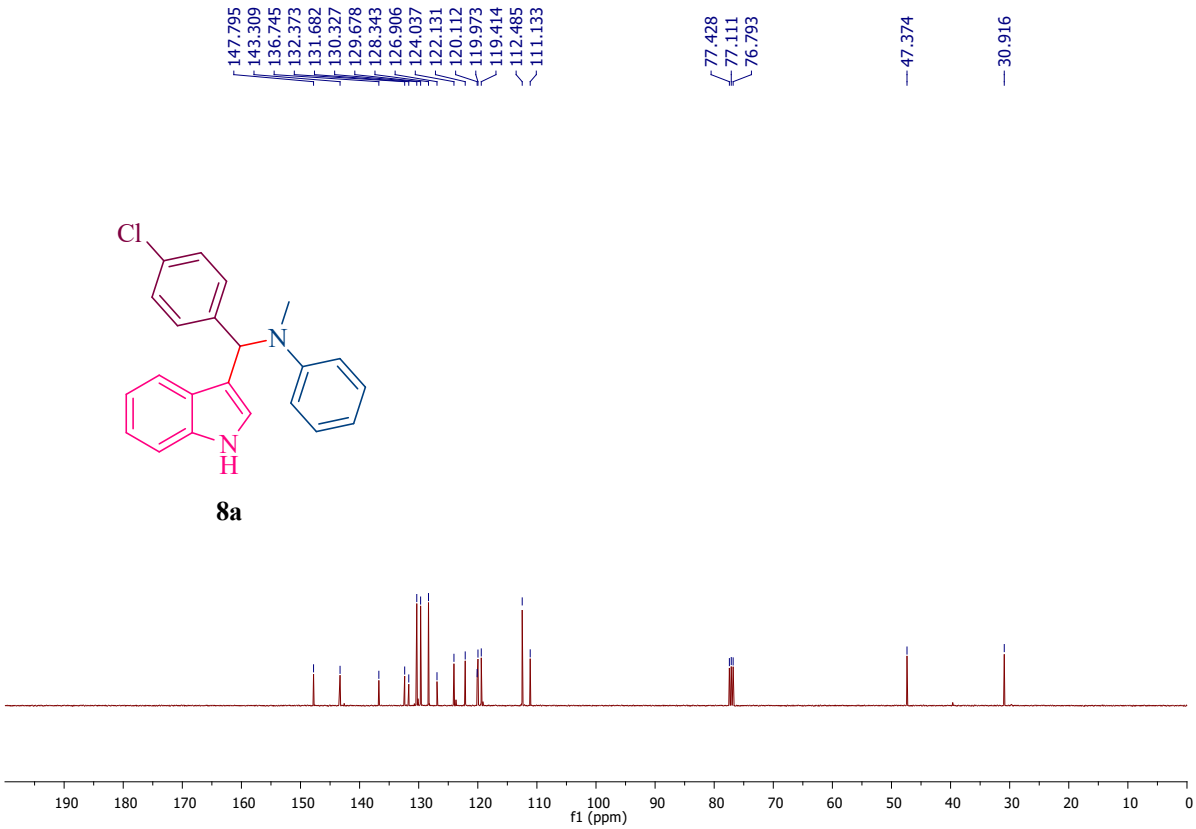
4p


9.334
8.291
8.270
7.814
7.568
7.547
7.265
7.050
7.029
6.565
6.559
6.417
6.395
6.267
6.261
3.714
2.757
2.713
2.686
2.642
2.448
2.406
2.391
2.349
1.168
1.036
OH
O
H3CO
O
1.00
1.16
1.14
1.33
2.17
1.06
1.04
1.08
1.05
1.10
3.24
2.17
2.28
3.24
3.32
10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)

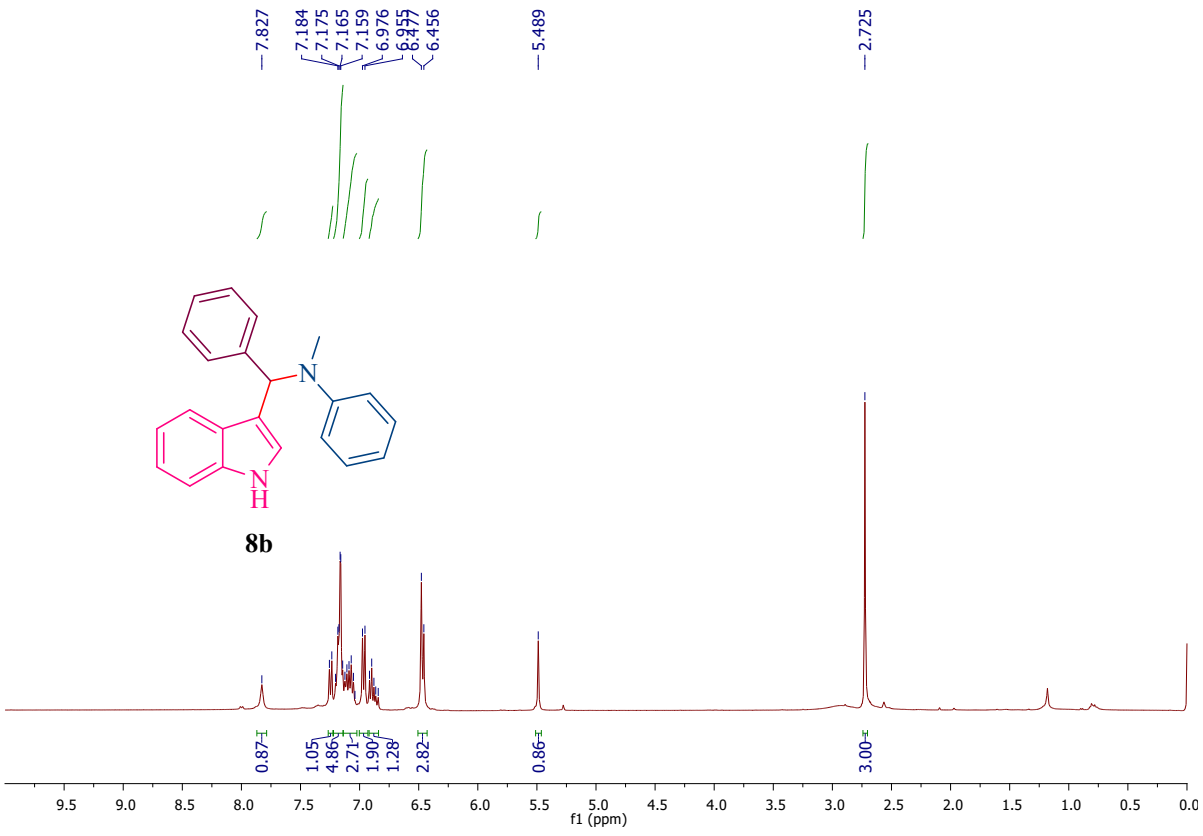
4q


200.819
166.660
159.306
153.916
147.853
131.555
131.121
129.382
128.259
127.499
125.247
123.532
117.577
116.588
114.103
108.212
103.482
77.352
77.034
76.717
55.110
50.292
41.600
32.425
29.075
27.540
27.282
OH
O
H3CO
O
4q
f1 (ppm)

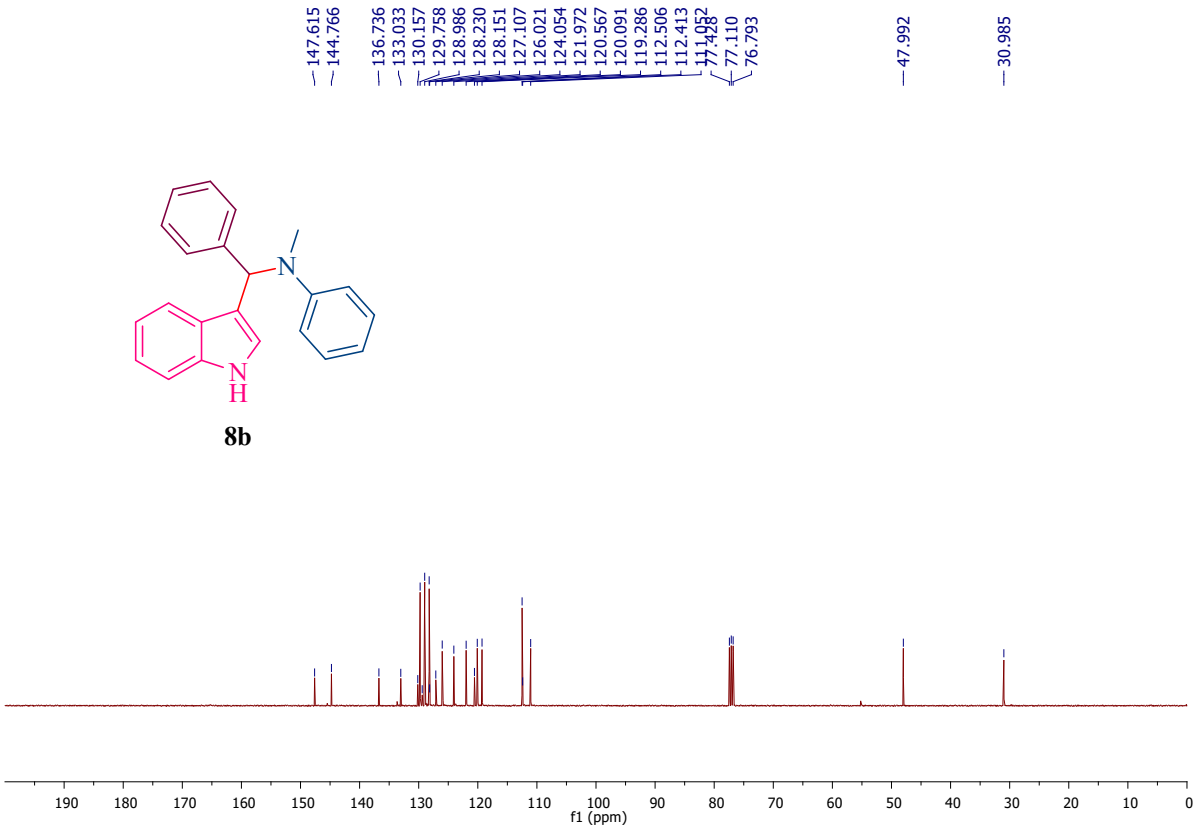

7.917
7.227
7.206
7.156
7.135
7.014
6.535
6.531
6.527
5.524
2.801
Cl
N
N
H
8a
1.00
1.11
3.97
3.21
3.12
3.01
1.00
3.04
f1 (ppm)


147.795
143.309
136.745
132.373
131.682
130.327
129.678
128.343
126.906
124.037
122.131
120.112
119.973
119.414
112.485
111.133
77.428
77.111
76.793
47.374
30.916
Cl
N
N
H
190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)

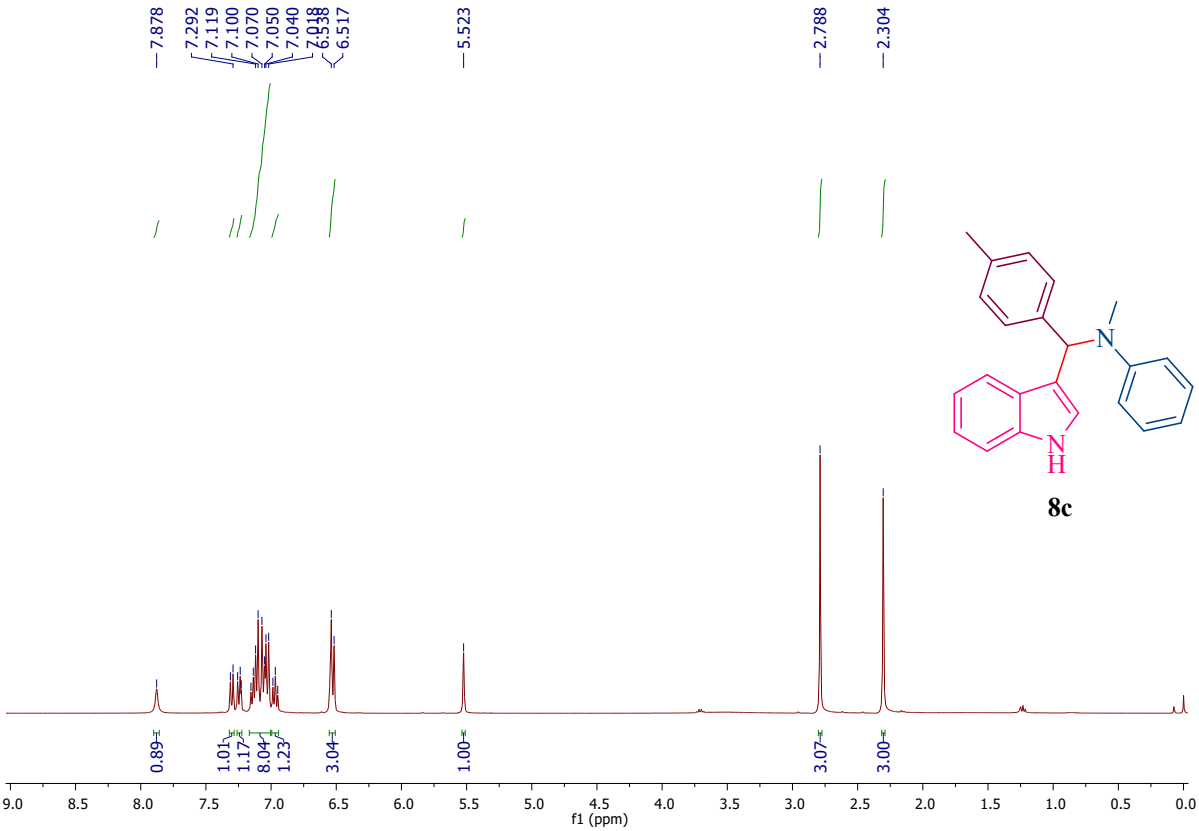
8a


7.827
7.184
7.175
7.165
7.159
6.976
6.955
6.477
6.456
5.489
2.725
N
N
H
0.87
1.05
4.86
2.71
1.90
1.28
2.82
0.86
3.00
9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)

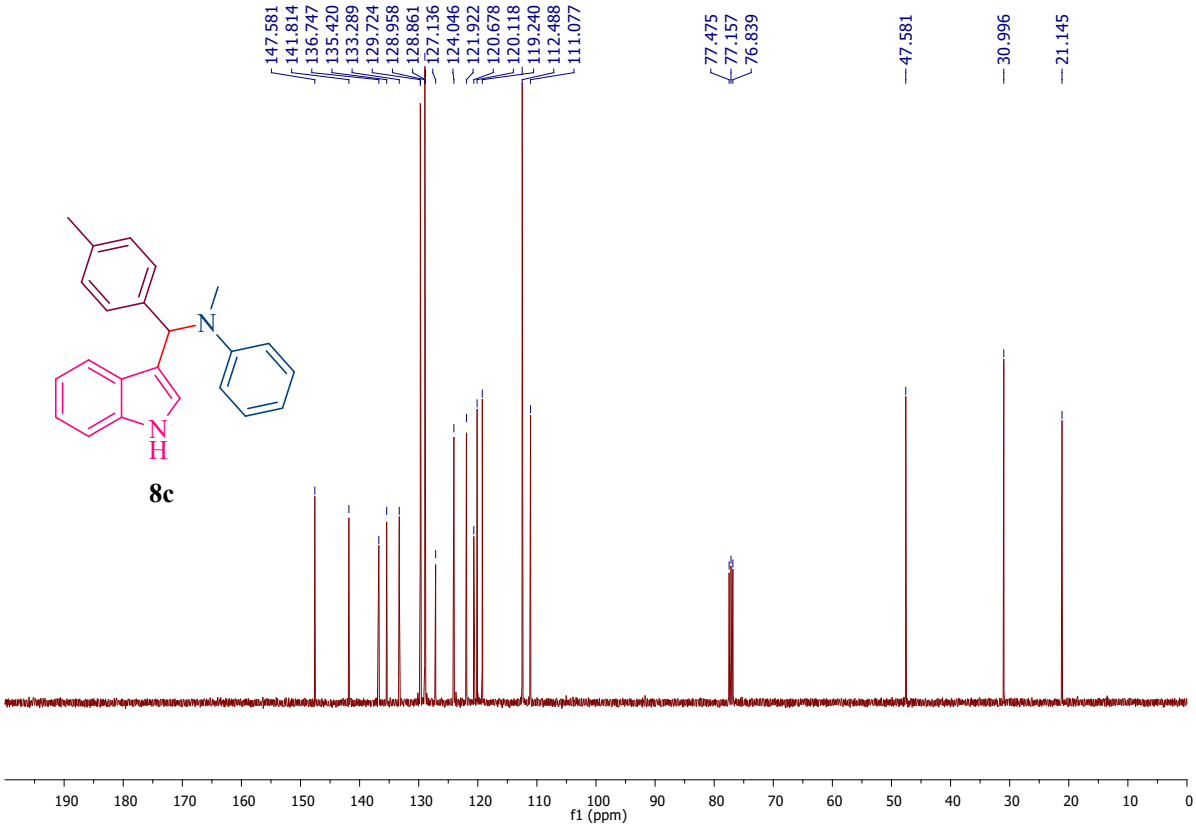
8b


147.615
144.766
136.736
133.033
130.157
129.758
128.986
128.230
128.151
127.107
126.021
124.054
121.972
120.567
120.091
119.286
112.506
112.413
111.052
77.428
77.110
76.793
47.992
30.985
N
N
H
f1 (ppm)

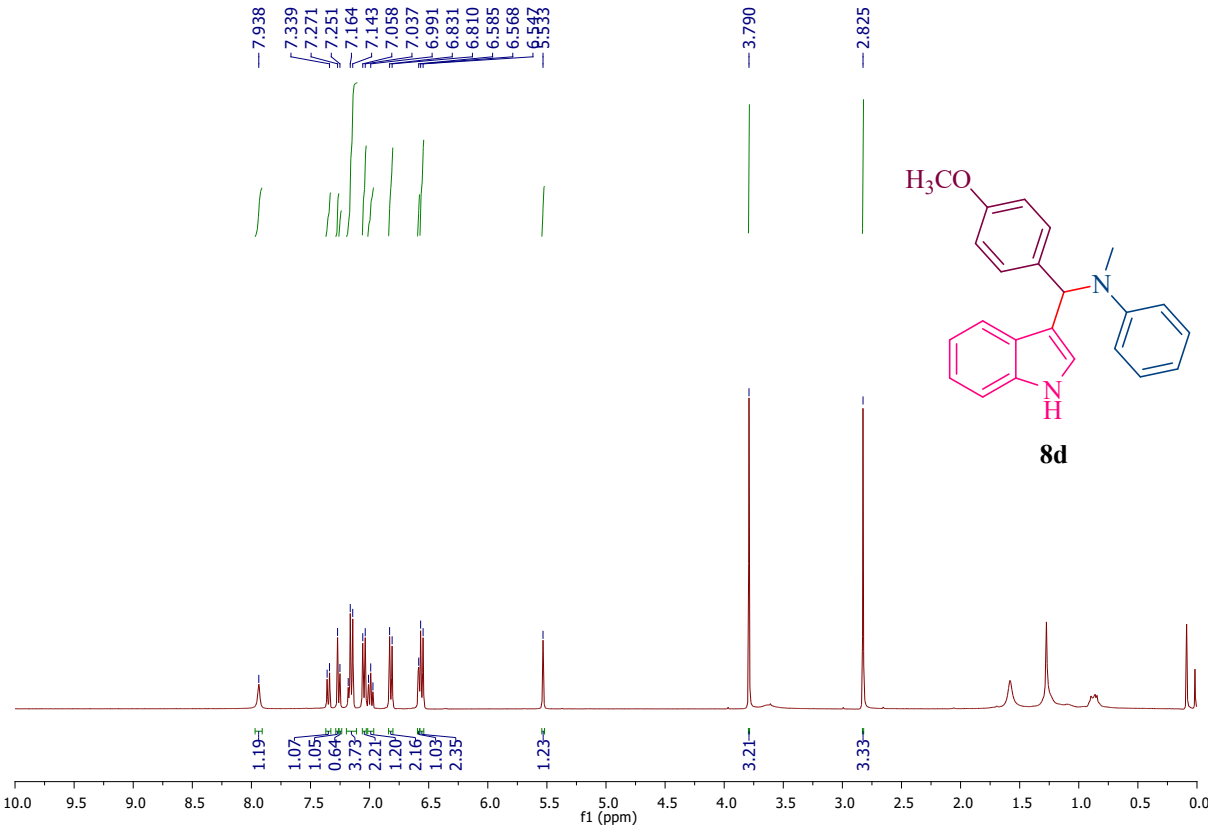
8b


7.878
7.292
7.119
7.100
7.070
7.050
7.040
6.538
6.517
5.523
2.788
2.304
N
N
H
0.89
1.01
1.17
8.04
1.23
3.04
1.00
3.07
3.00
f1 (ppm)

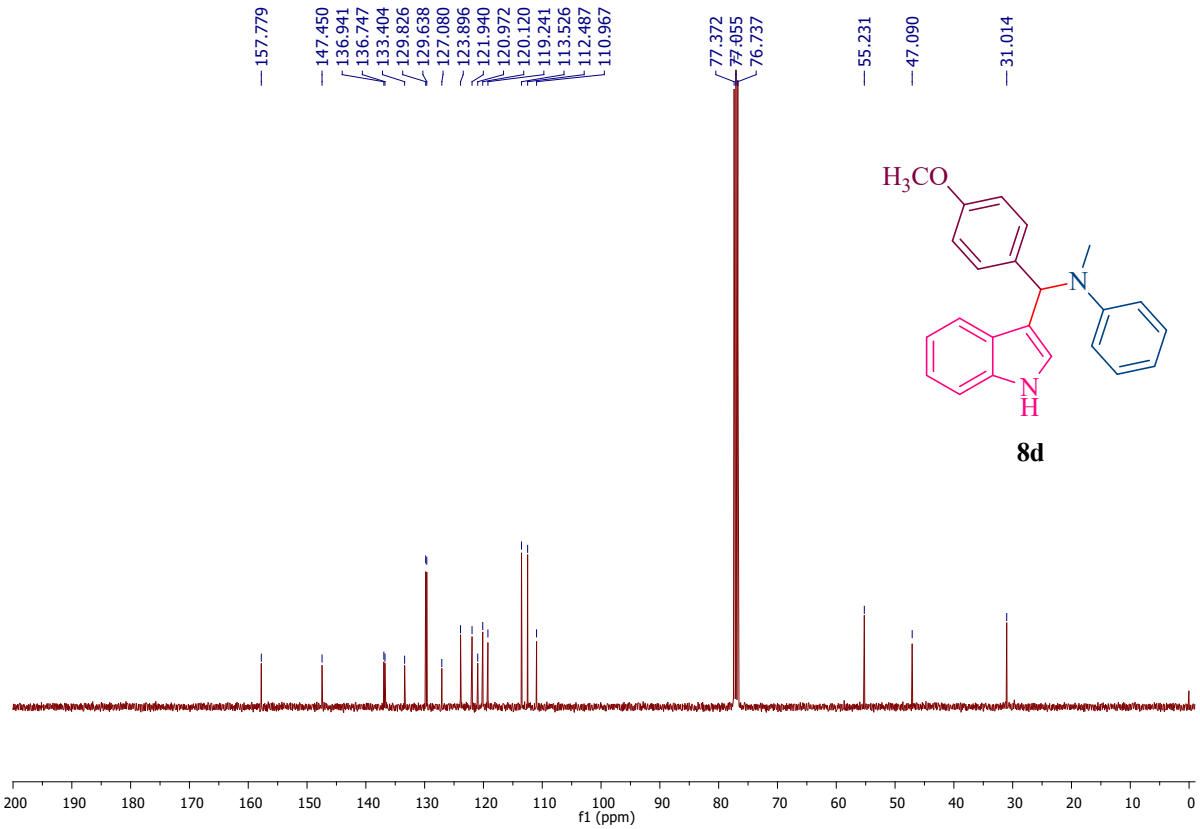
8c


147.581
141.814
136.747
135.420
133.289
129.724
128.958
128.861
127.136
124.046
121.922
120.678
120.118
119.240
112.488
111.077
77.475
77.157
76.839
47.581
30.996
21.145
N
N
H
f1 (ppm)

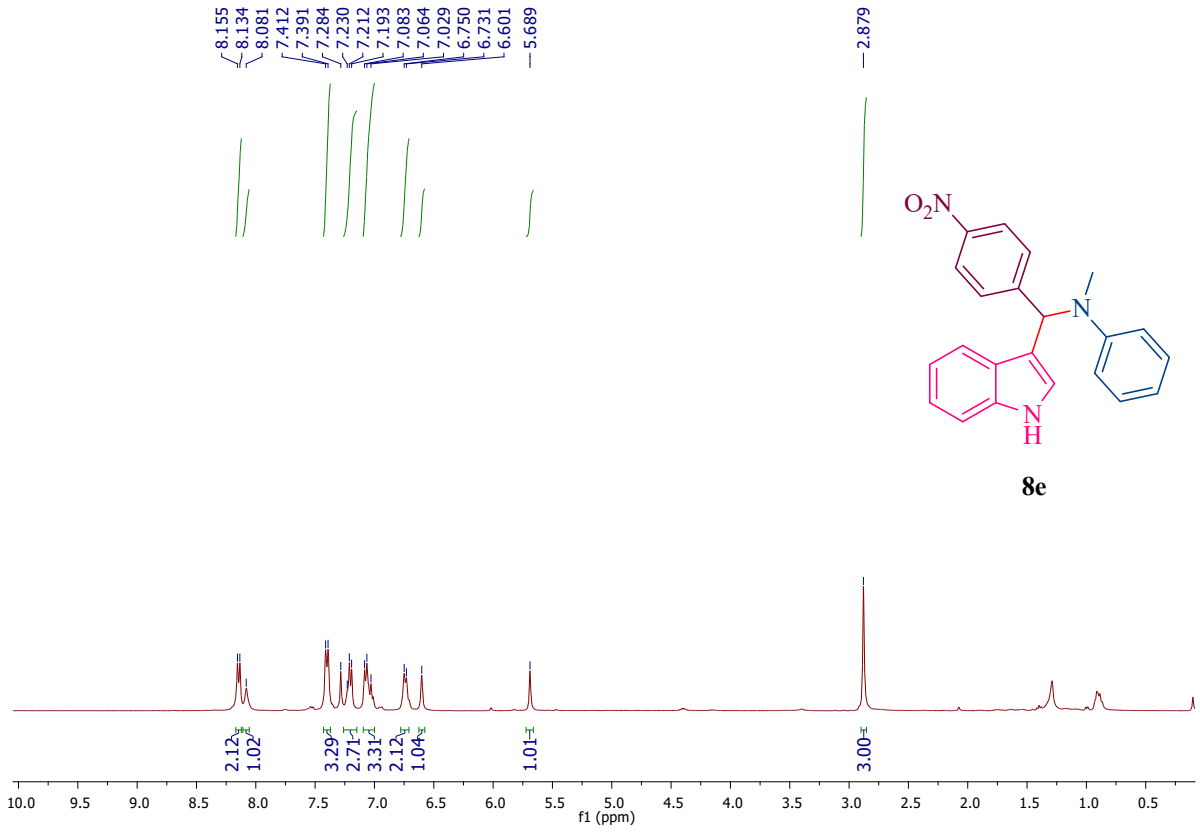
8c


7.938
7.339
7.271
7.251
7.164
7.143
7.058
7.037
6.991
6.831
6.810
6.585
6.568
6.547
5.533
3.790
2.825
H3CO
N
N
H
1.19
1.07
1.05
0.64
3.73
2.21
1.20
2.16
1.03
2.35
1.23
3.21
3.33
f1 (ppm)

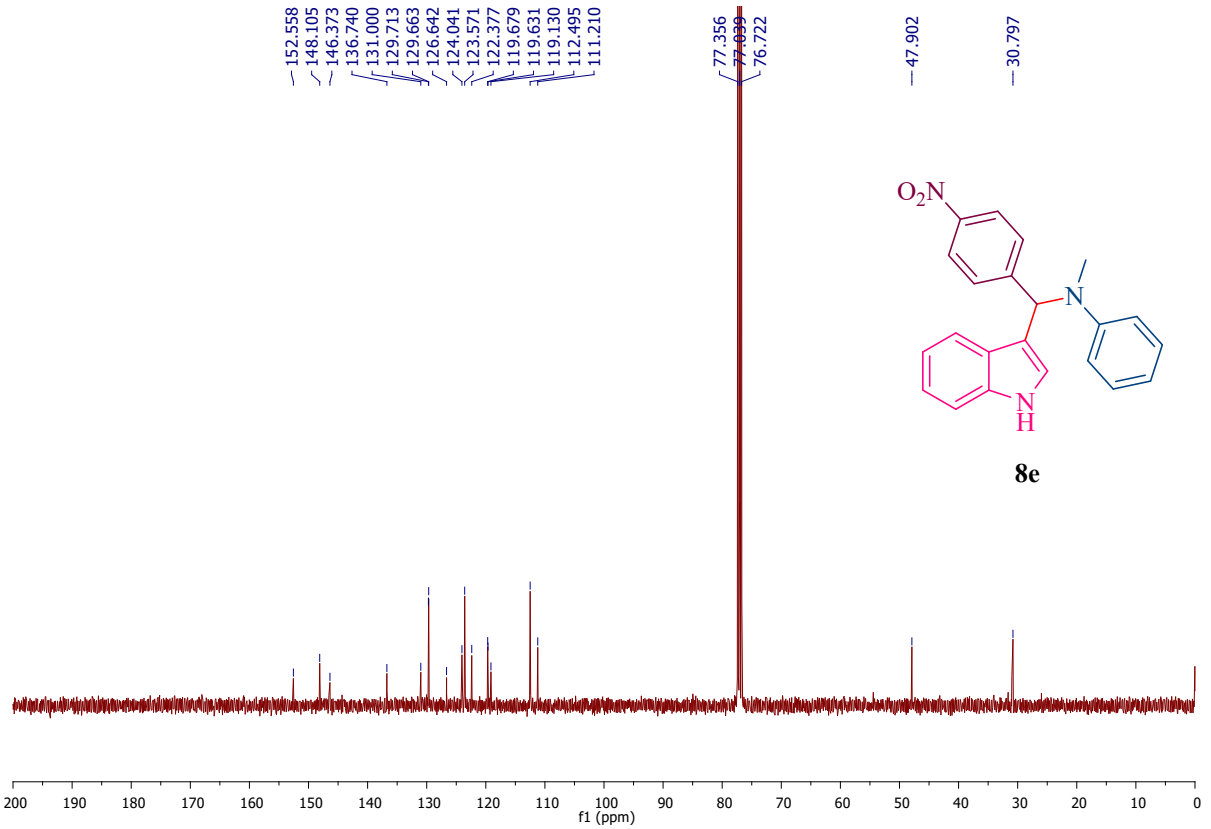
8d


157.779
147.450
136.941
136.747
133.404
129.826
129.638
127.080
123.896
121.940
120.972
120.120
119.241
113.526
112.487
110.967
77.372
77.055
76.737
55.231
47.090
31.014
H3CO
N
N
H
f1 (ppm)

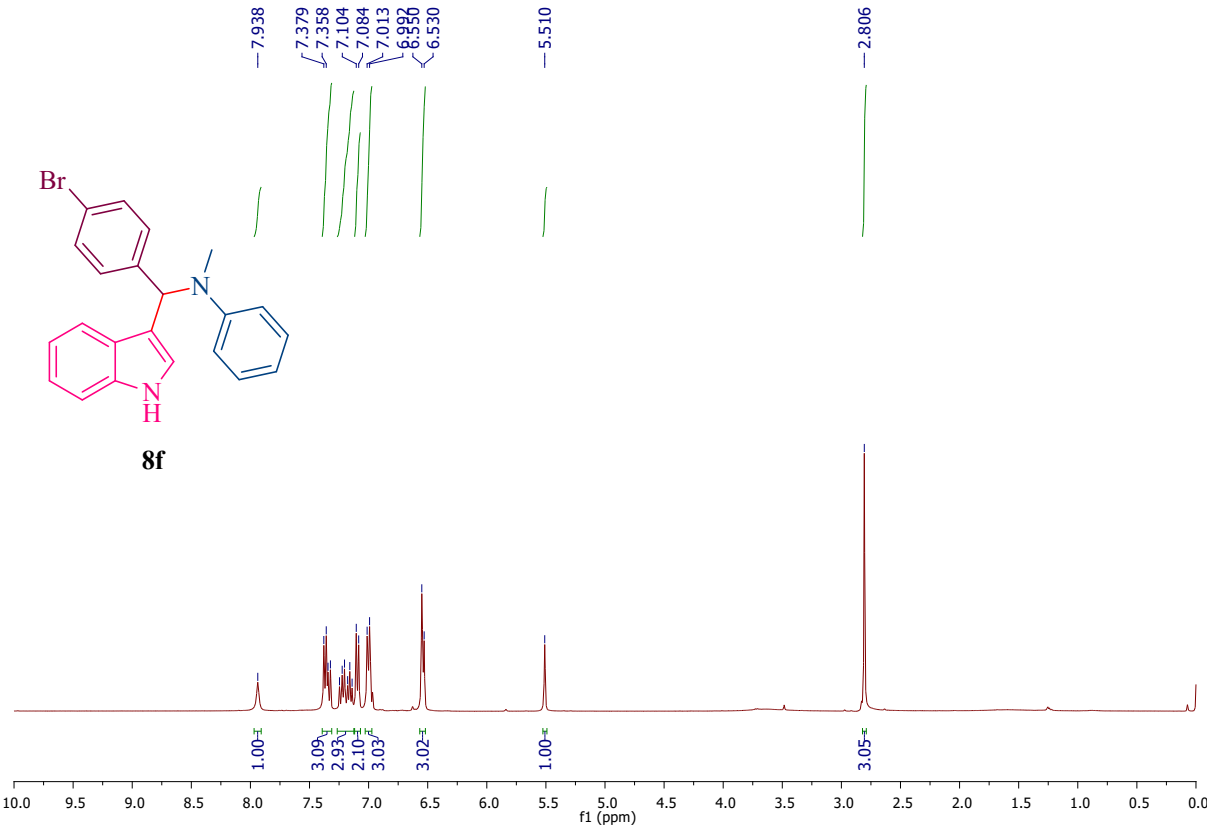
8d


8.155
8.134
8.081
7.412
7.391
7.284
7.230
7.212
7.193
7.083
7.064
7.029
6.750
6.731
6.601
5.689
2.879
O2N
N
N
H
2.12
1.02
3.29
2.71
3.31
2.12
1.04
1.01
3.00
f1 (ppm)

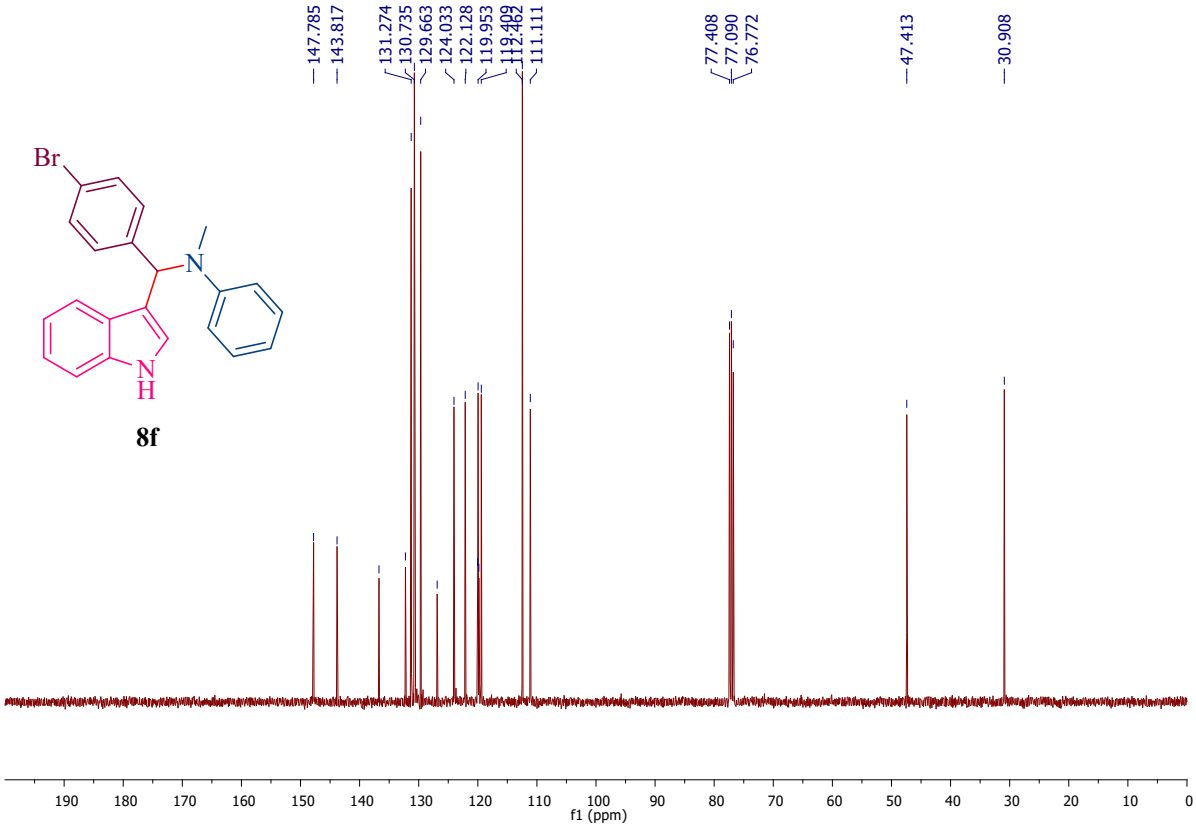
8e


152.558
148.105
146.373
136.740
131.000
129.713
129.663
126.642
124.041
123.571
122.377
119.679
119.631
119.130
112.495
111.210
77.356
77.039
76.722
47.902
30.797
O2N
N
N
H
f1 (ppm)

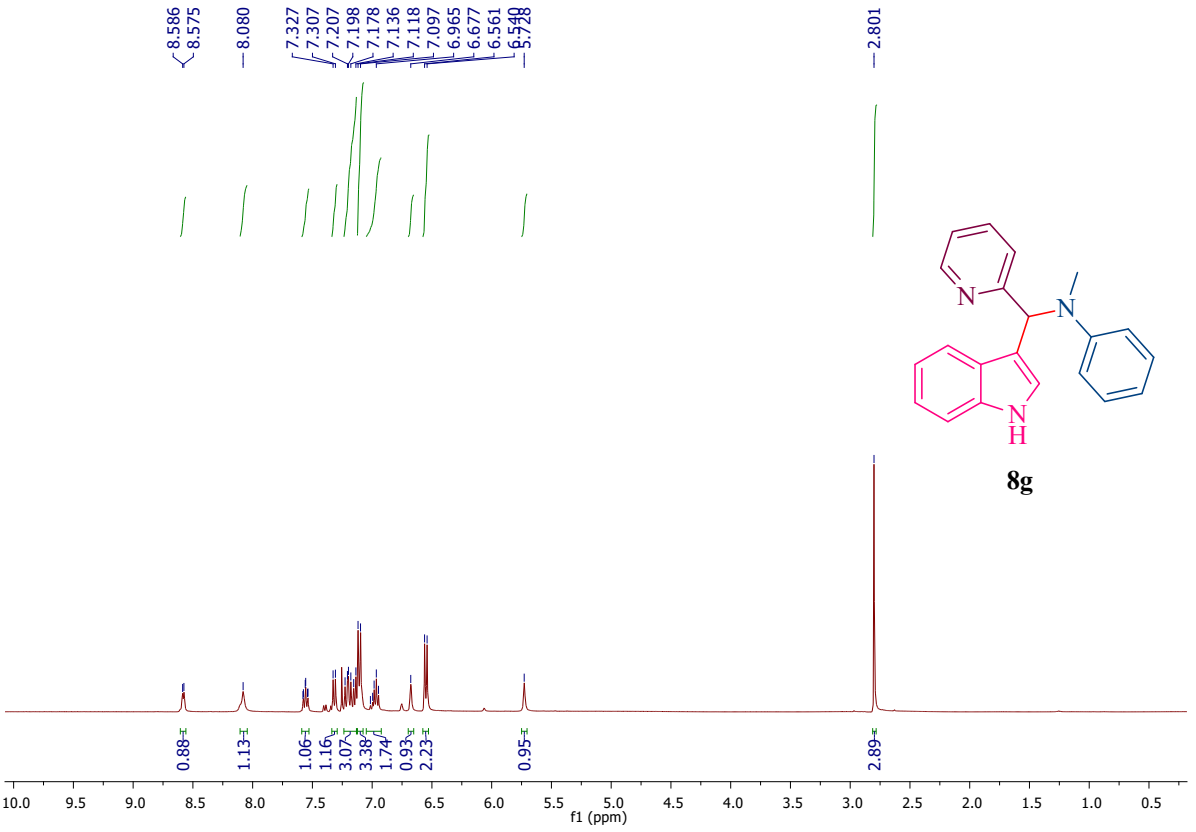
8e


7.938
7.379
7.358
7.104
7.084
7.013
6.992
6.550
6.530
5.510
2.806
Br
N
N
H
1.00
3.09
2.93
2.10
3.03
3.02
1.00
3.05
f1 (ppm)

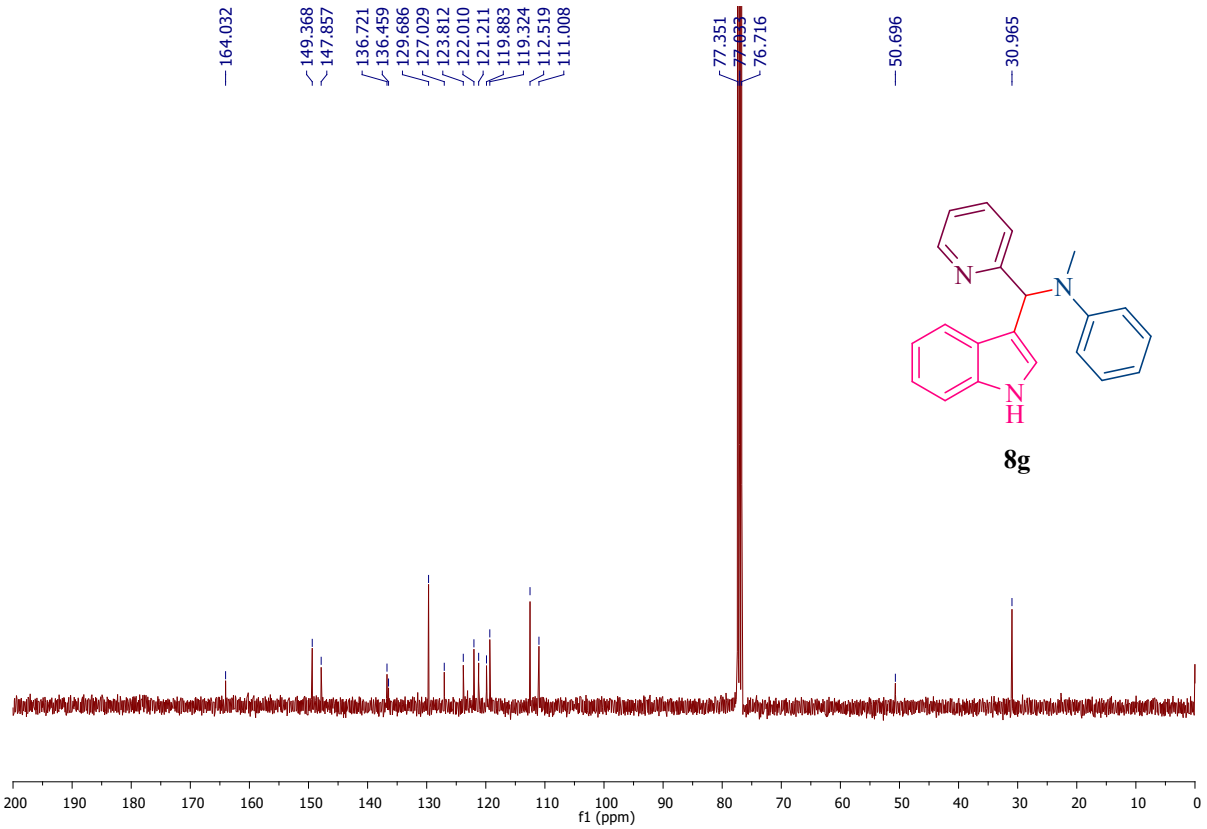
8f


147.785
143.817
131.274
130.735
129.663
124.033
122.128
119.953
111.111
77.408
77.090
76.772
47.413
30.908
Br
N
N
H
f1 (ppm)

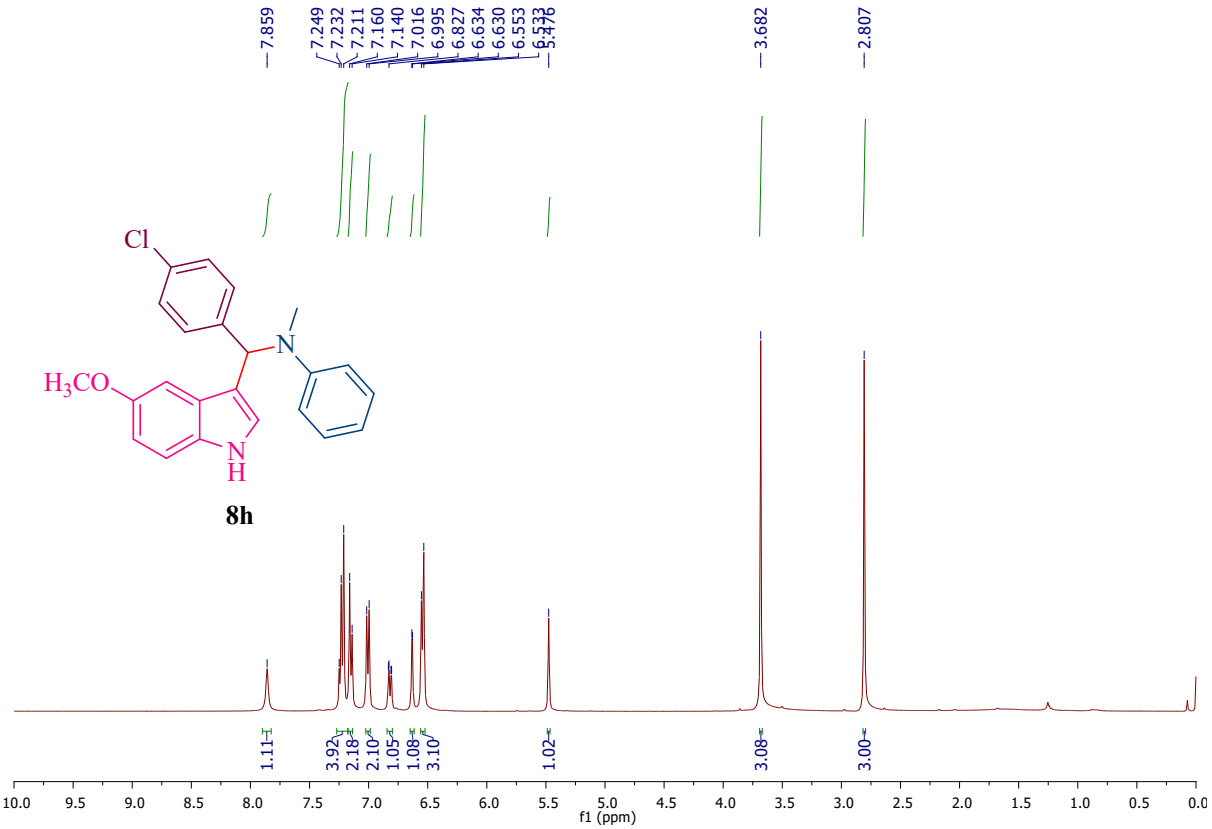
8f


8.586
8.575
8.080
7.327
7.307
7.207
7.198
7.178
7.136
7.118
7.097
6.965
6.677
6.561
6.540
2.801
N
N
N
H
0.88
1.13
1.06
1.16
3.07
3.38
1.74
0.93
2.23
0.95
2.89
f1 (ppm)

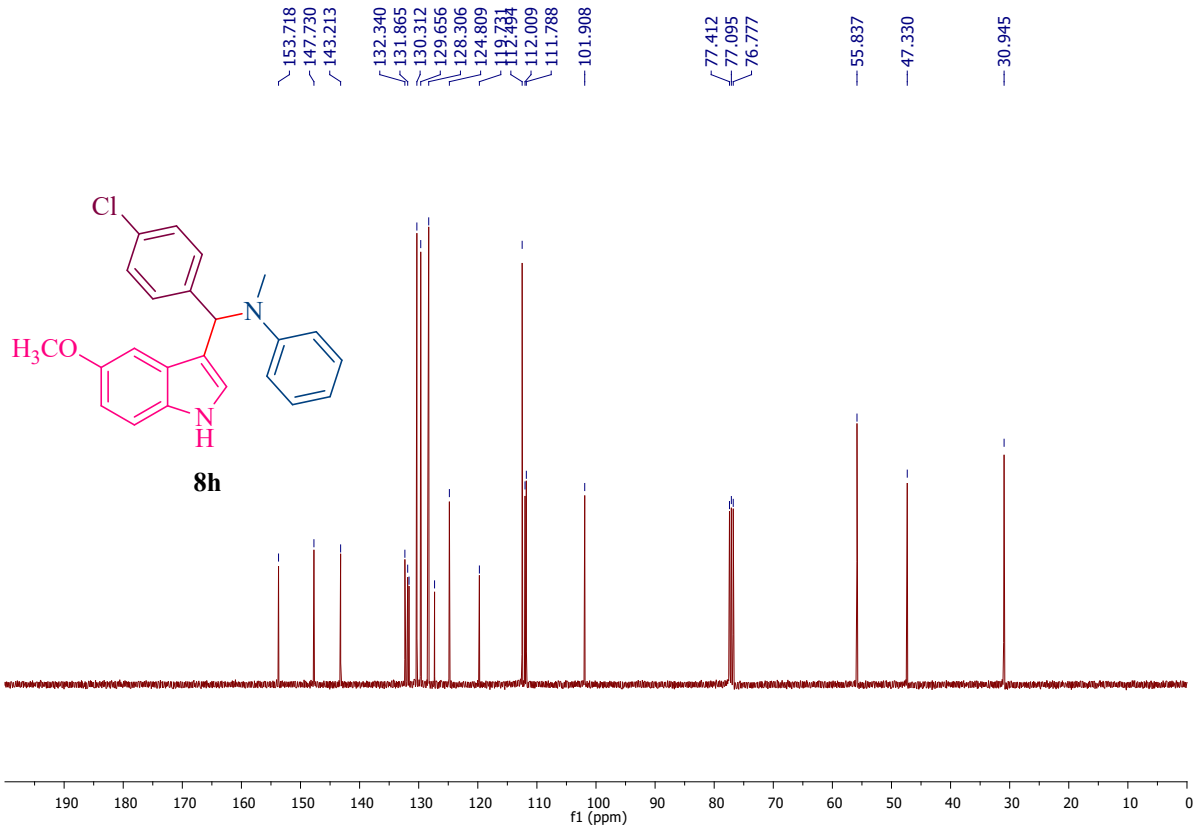
8g


164.032
149.368
147.857
136.721
136.459
129.686
127.029
123.812
122.010
121.211
119.883
119.324
112.519
111.008
77.351
77.033
76.716
50.696
30.965
f1 (ppm)

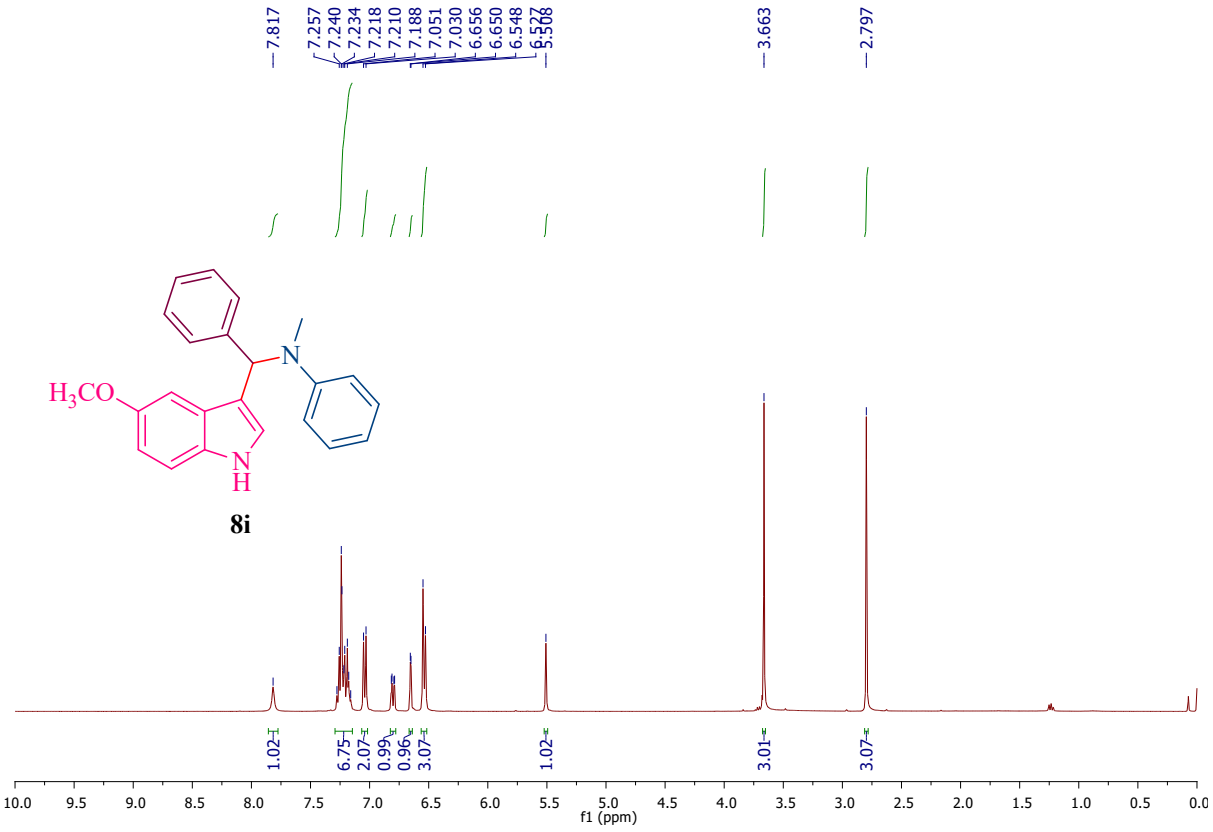
8g


7.859
7.249
7.232
7.211
7.160
7.140
7.016
6.995
6.827
6.634
6.630
6.553
3.682
2.807
1.11
3.92
2.18
2.10
1.05
1.08
3.10
1.02
3.08
3.00
f1 (ppm)

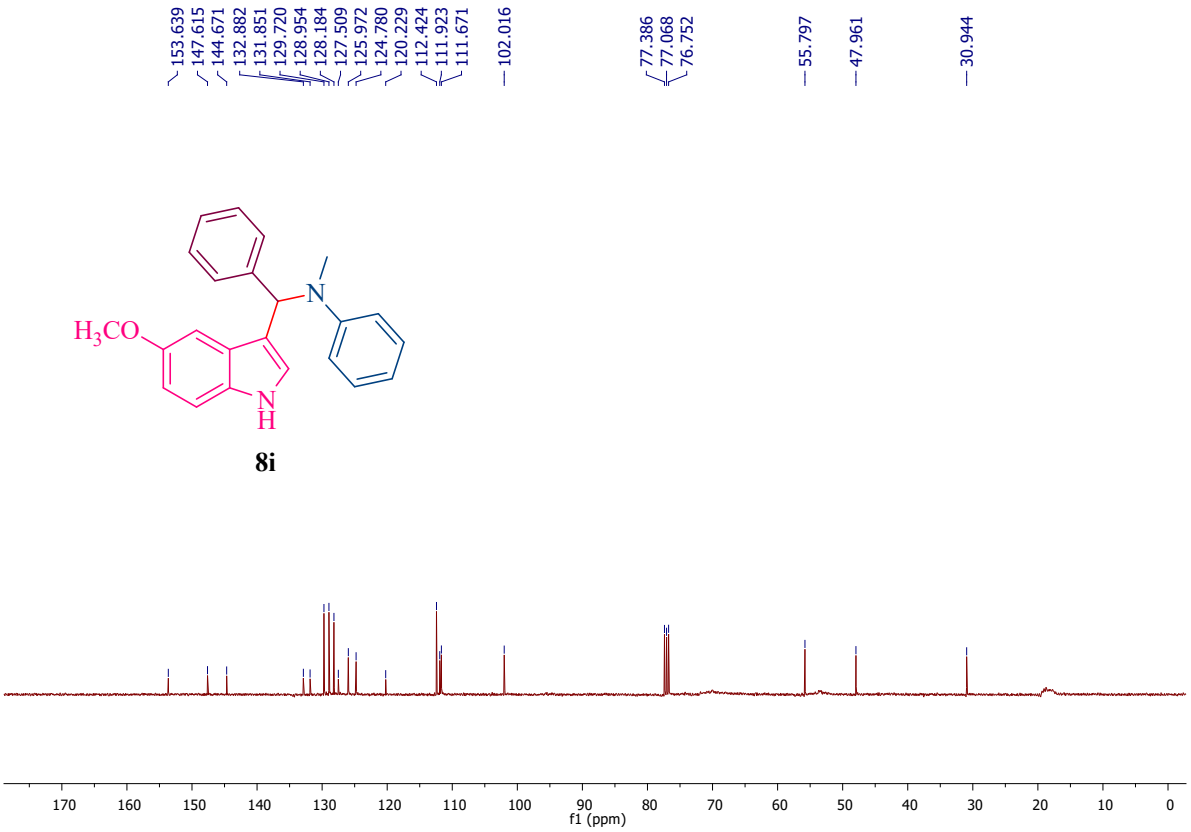
8h


153.718
147.730
143.213
132.340
131.865
130.312
129.656
128.306
124.809
119.731
112.494
112.009
111.788
101.908
77.412
77.095
76.777
55.837
47.330
30.945
Cl
N
H3CO
N
H
f1 (ppm)

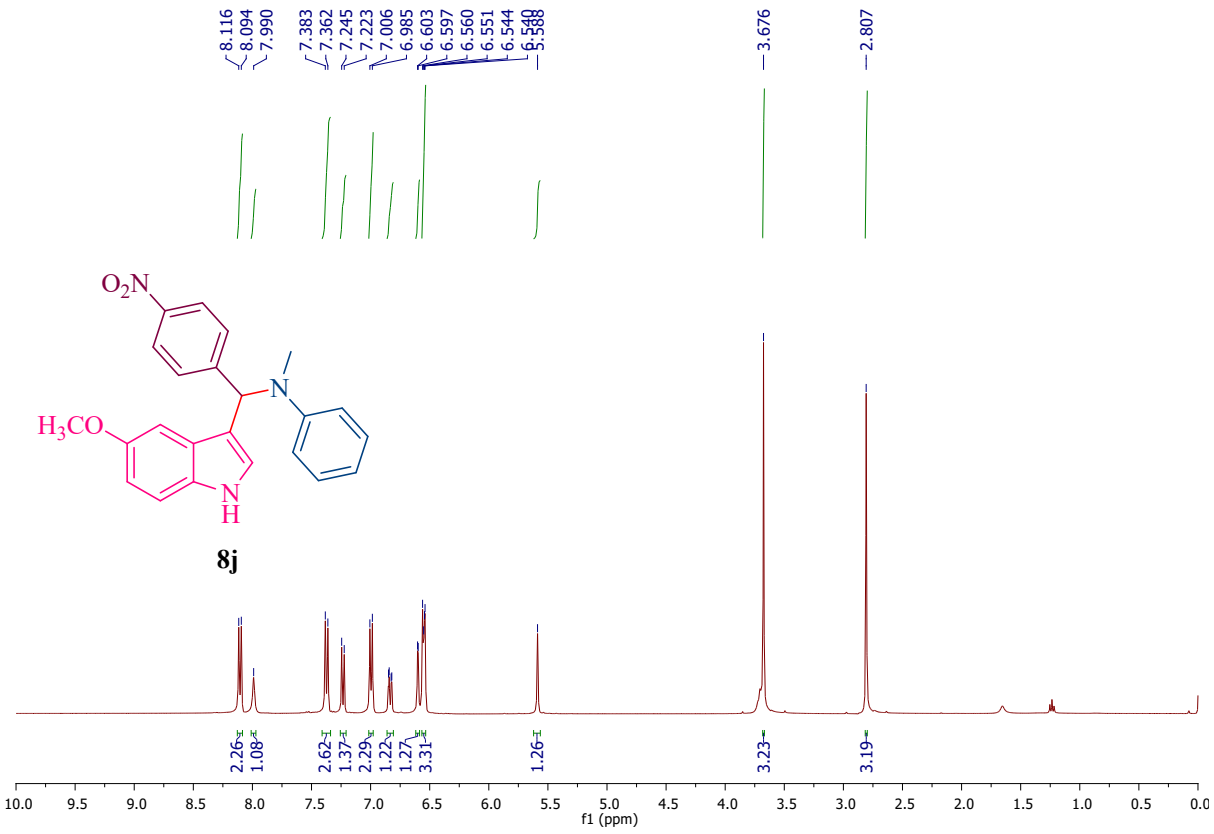
8h


7.817
7.257
7.240
7.234
7.218
7.210
7.188
7.051
7.030
6.656
6.650
6.548
6.527
6.508
3.663
2.797
N
H3CO
N
H
1.02
6.75
2.07
0.99
0.96
3.07
1.02
3.01
3.07
f1 (ppm)

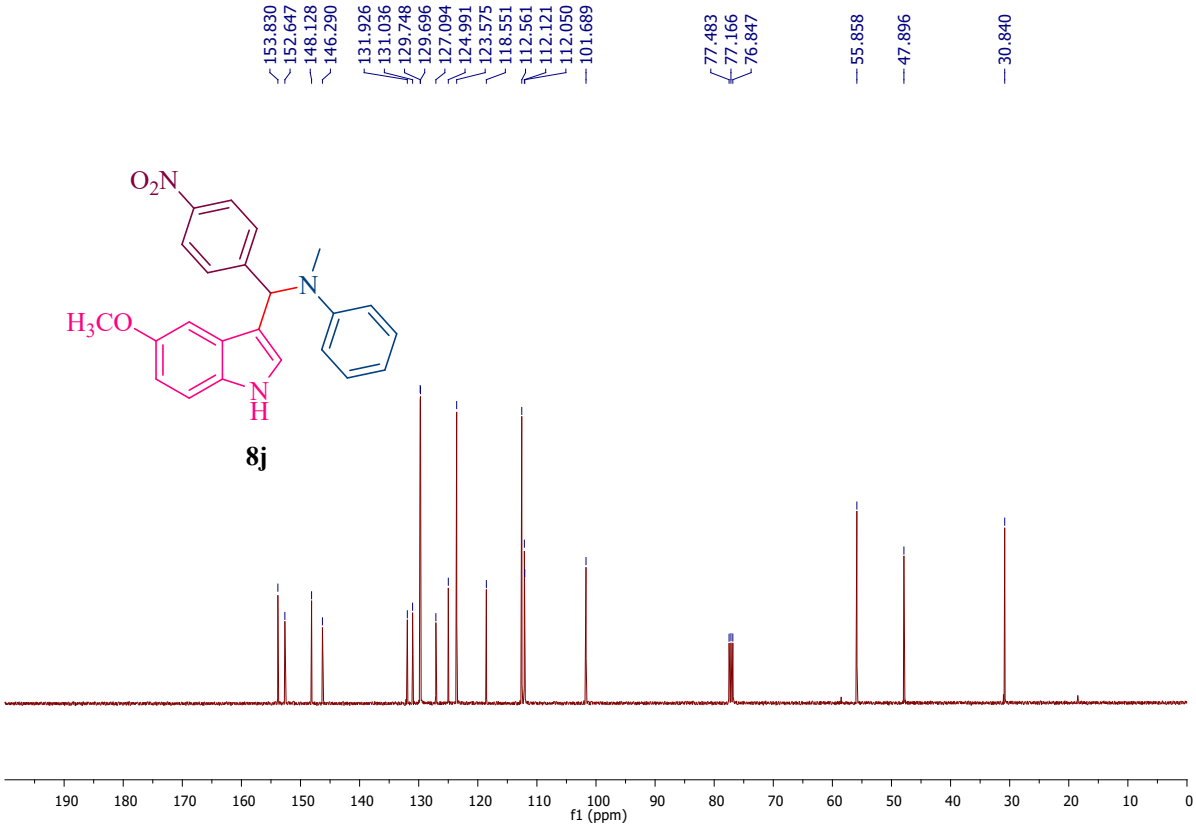
8i

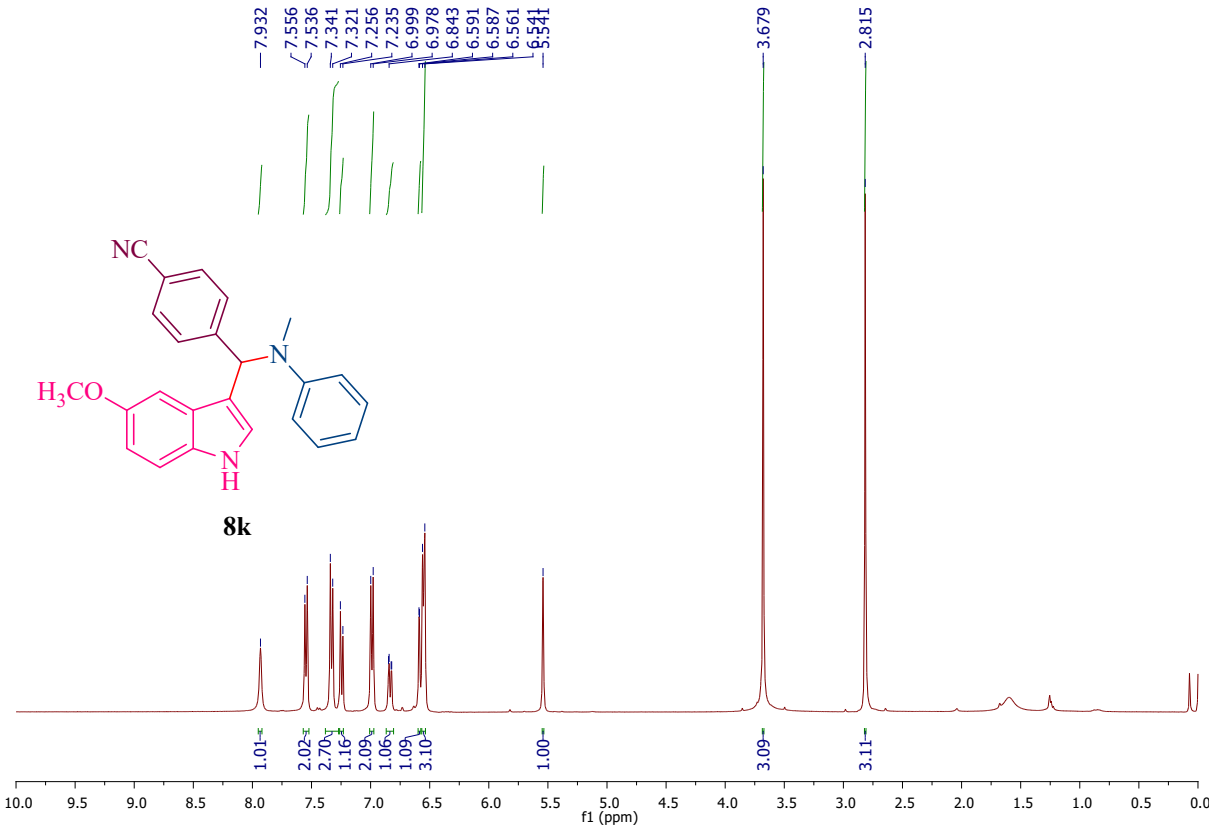

153.639
147.615
144.671
132.882
131.851
129.720
128.954
128.184
127.509
125.972
124.780
120.229
112.424
111.923
111.671
102.016
77.386
77.068
76.752
55.797
47.961
30.944
H3CO
N
N
H
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

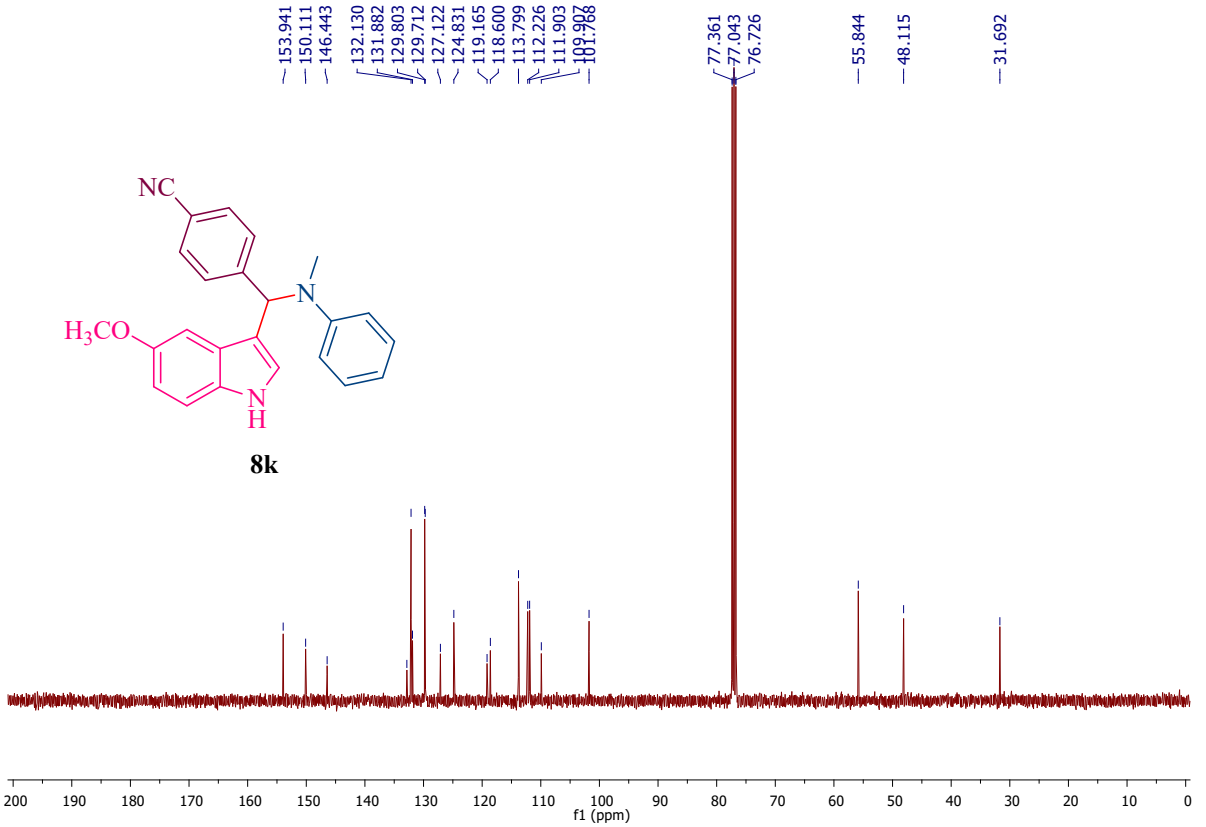
8i


8.116
8.094
7.990
7.383
7.362
7.245
7.223
7.006
6.985
6.603
6.597
6.560
6.551
6.544
3.676
2.807
O2N
H3CO
N
N
H
2.26
1.08
2.62
1.37
2.29
1.22
1.27
3.31
1.26
3.23
3.19
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

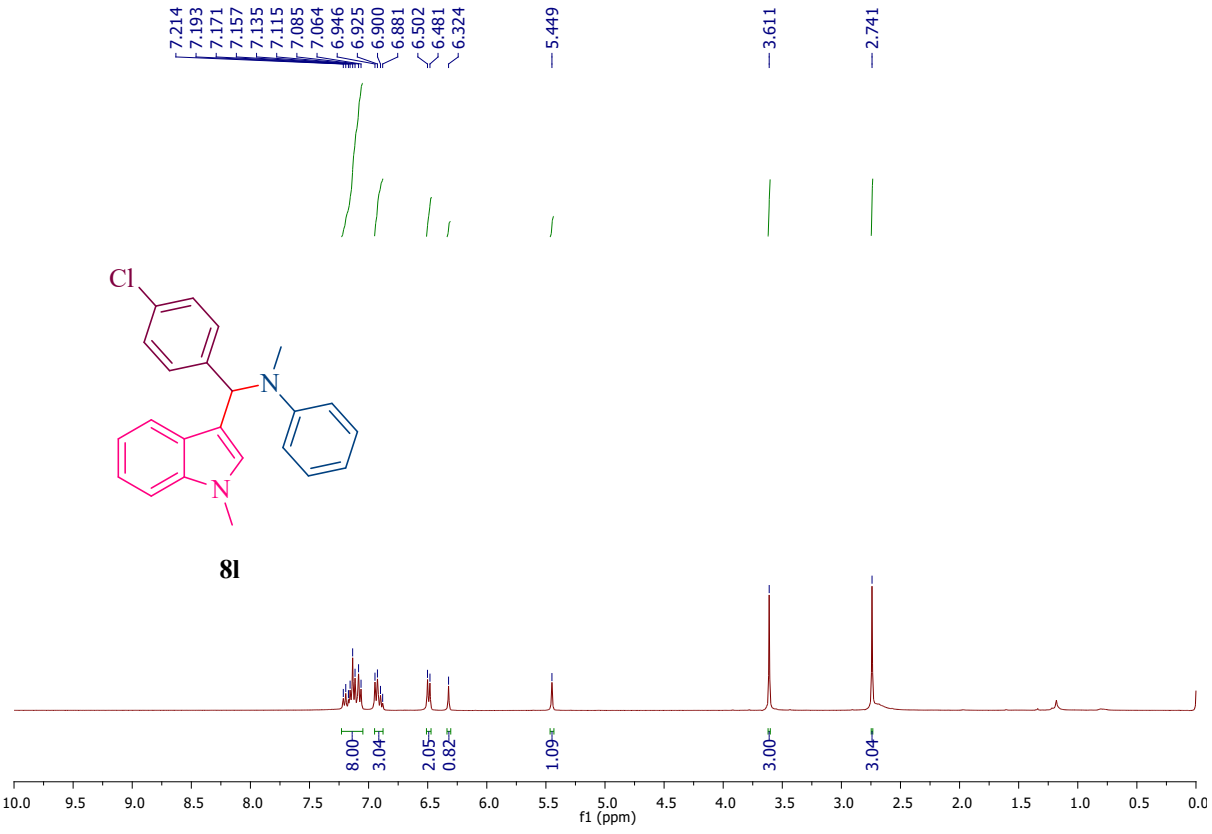
8j


153.830
152.647
148.128
146.290
131.926
131.036
129.748
129.696
127.094
124.991
123.575
118.551
112.561
112.121
112.050
101.689
77.483
77.166
76.847
55.858
47.896
30.840
O2N
N
H3CO
N
H
8j
f1 (ppm)

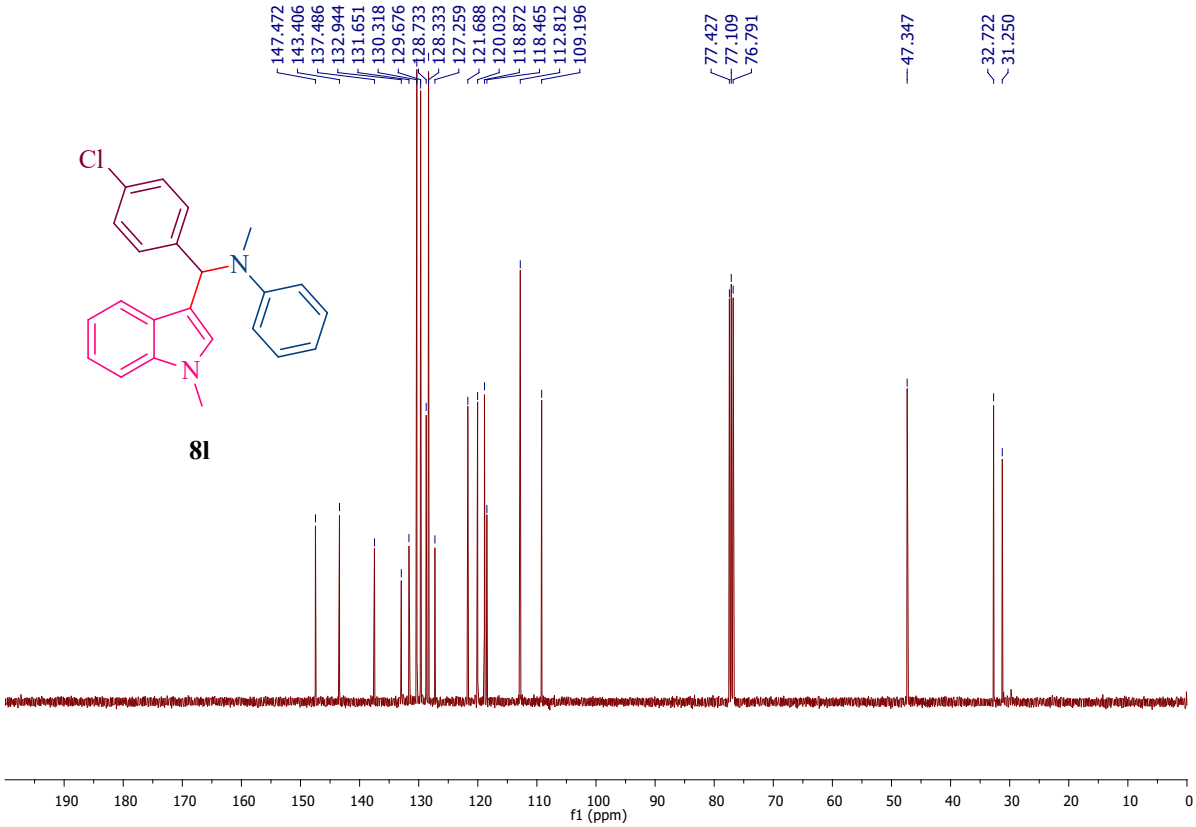

7.932
7.556
7.536
7.341
7.321
7.256
7.235
6.999
6.978
6.843
6.591
6.587
6.561
6.541
5.541
3.679
2.815
NC
N
H3CO
N
H
8k
1.01
2.02
2.70
1.16
2.09
1.06
1.09
3.10
1.00
3.09
3.11
f1 (ppm)

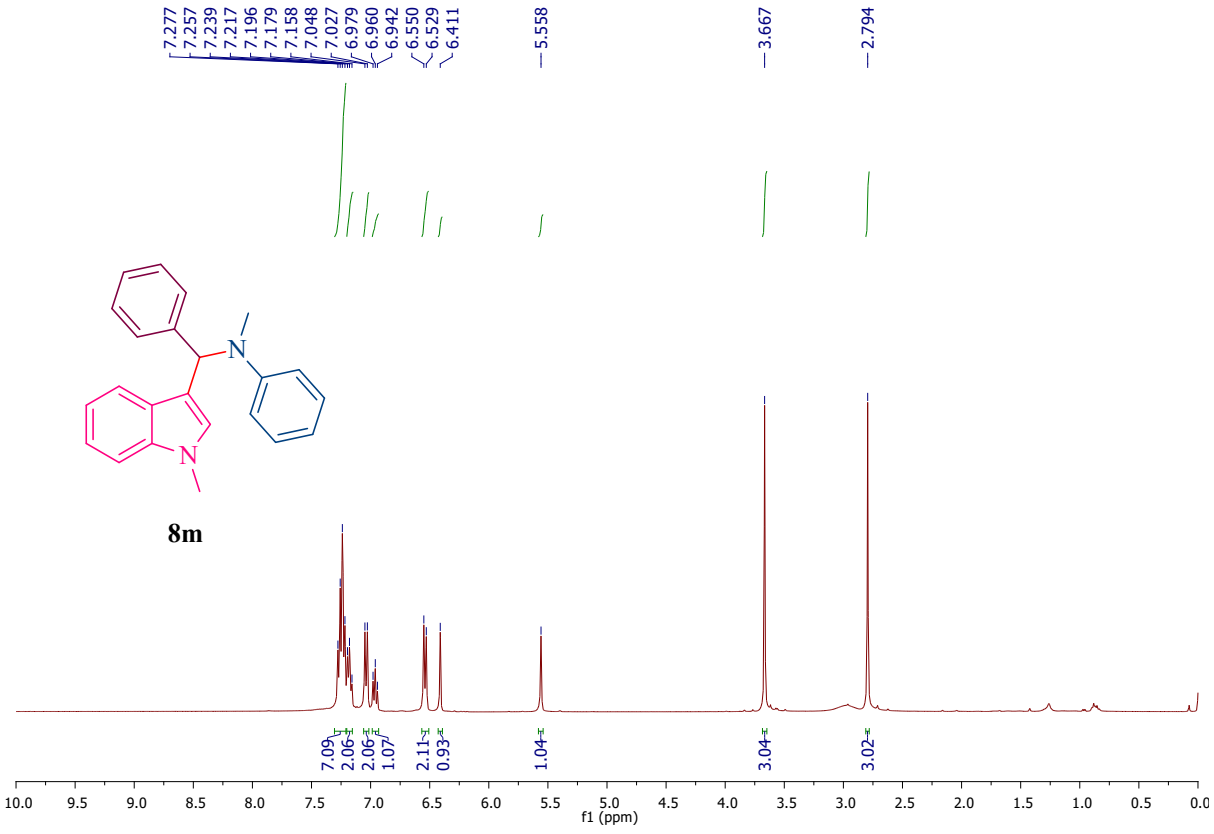

153.941
150.111
146.443
132.130
131.882
129.803
129.712
127.122
124.831
119.165
118.600
113.799
112.226
111.903
101.988
77.361
77.043
76.726
55.844
48.115
31.692
NC
H3CO
N
N
H
f1 (ppm)

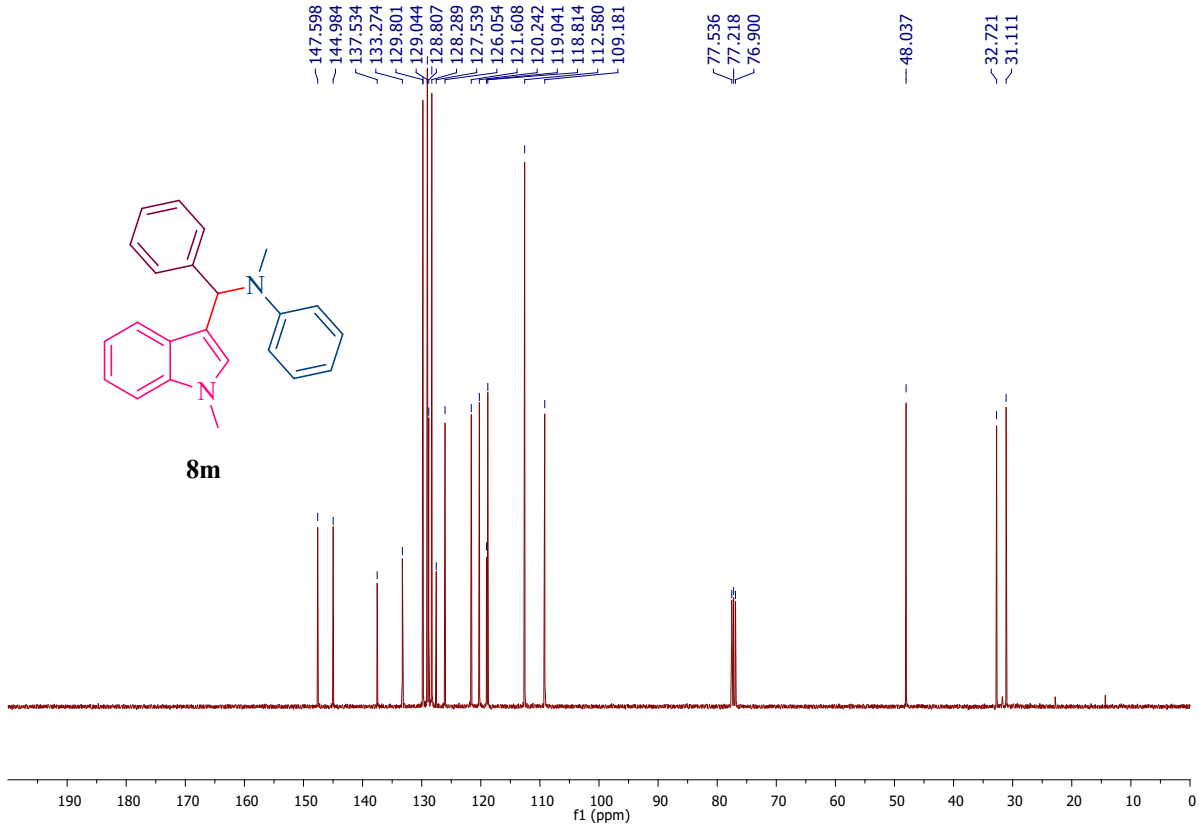
8k


7.214
7.193
7.171
7.157
7.135
7.115
7.085
7.064
6.946
6.925
6.900
6.881
6.502
6.481
6.324
5.449
3.611
2.741
Cl
N
N
8.00
3.04
2.05
0.82
1.09
3.00
3.04
f1 (ppm)

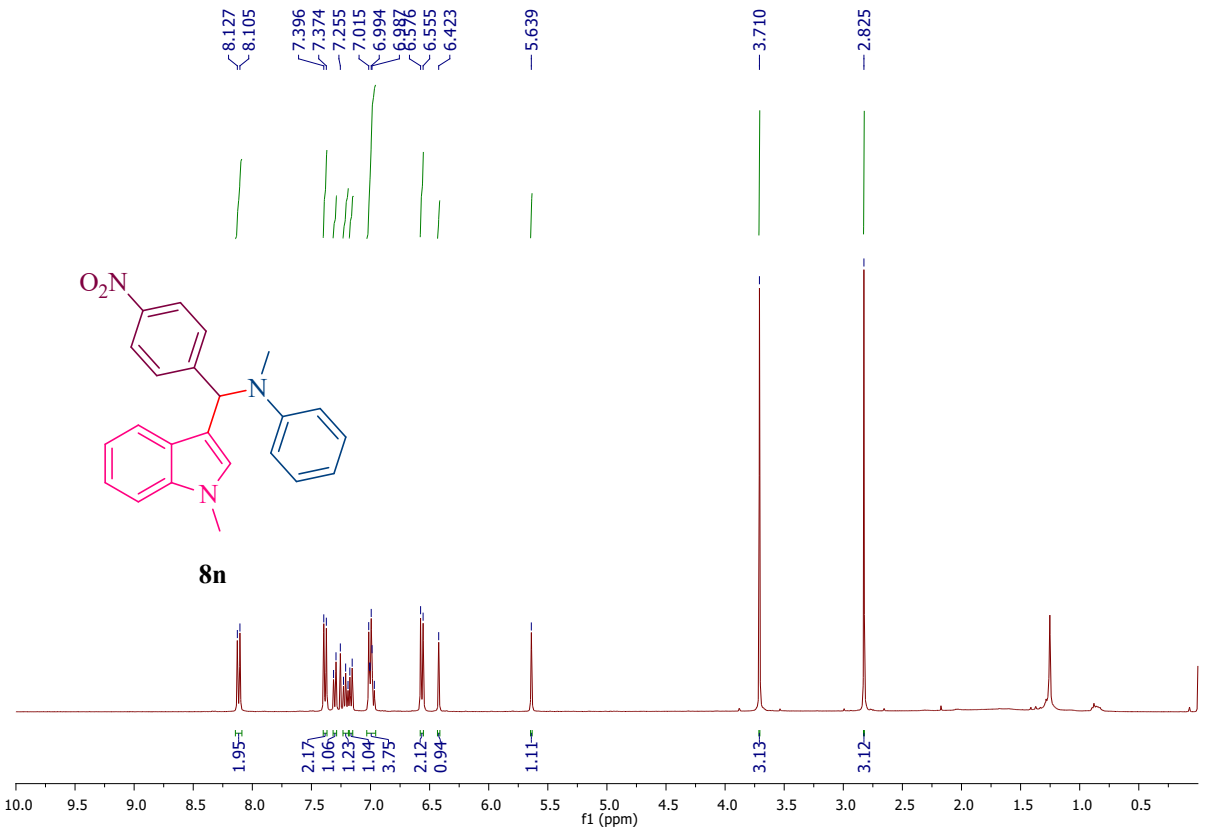
8l


147.472
143.406
137.486
132.944
131.651
130.318
129.676
128.733
128.333
127.259
121.688
120.032
118.872
118.465
112.812
109.196
77.427
77.109
76.791
47.347
32.722
31.250
Cl
N
N
8l
190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)

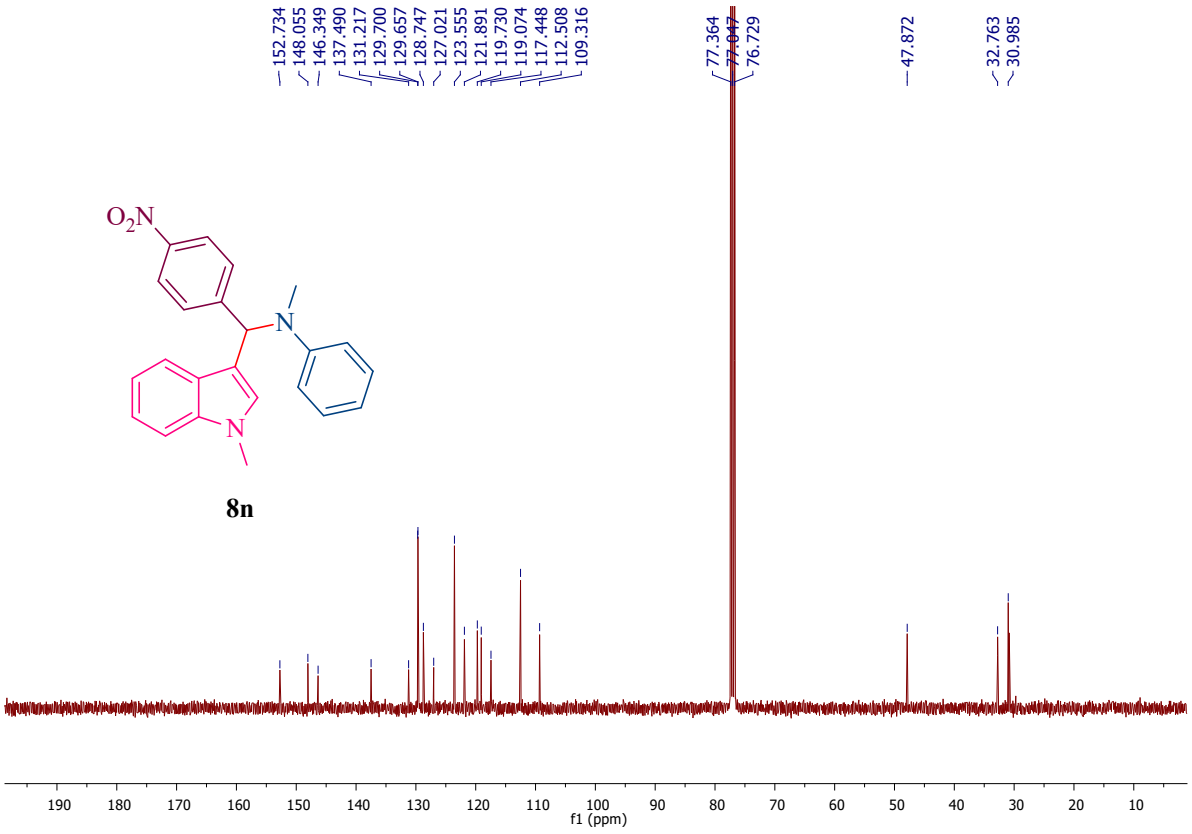

7.277
7.257
7.239
7.217
7.196
7.179
7.158
7.048
7.027
6.979
6.960
6.942
6.550
6.529
6.411
5.558
3.667
2.794
N
N
8m
7.09
2.06
2.06
1.07
2.11
0.93
1.04
3.04
3.02
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)


147.598
144.984
137.534
133.274
129.801
129.044
128.807
128.289
127.539
126.054
121.608
120.242
119.041
118.814
112.580
109.181
77.536
77.218
76.900
48.037
32.721
31.111
190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)

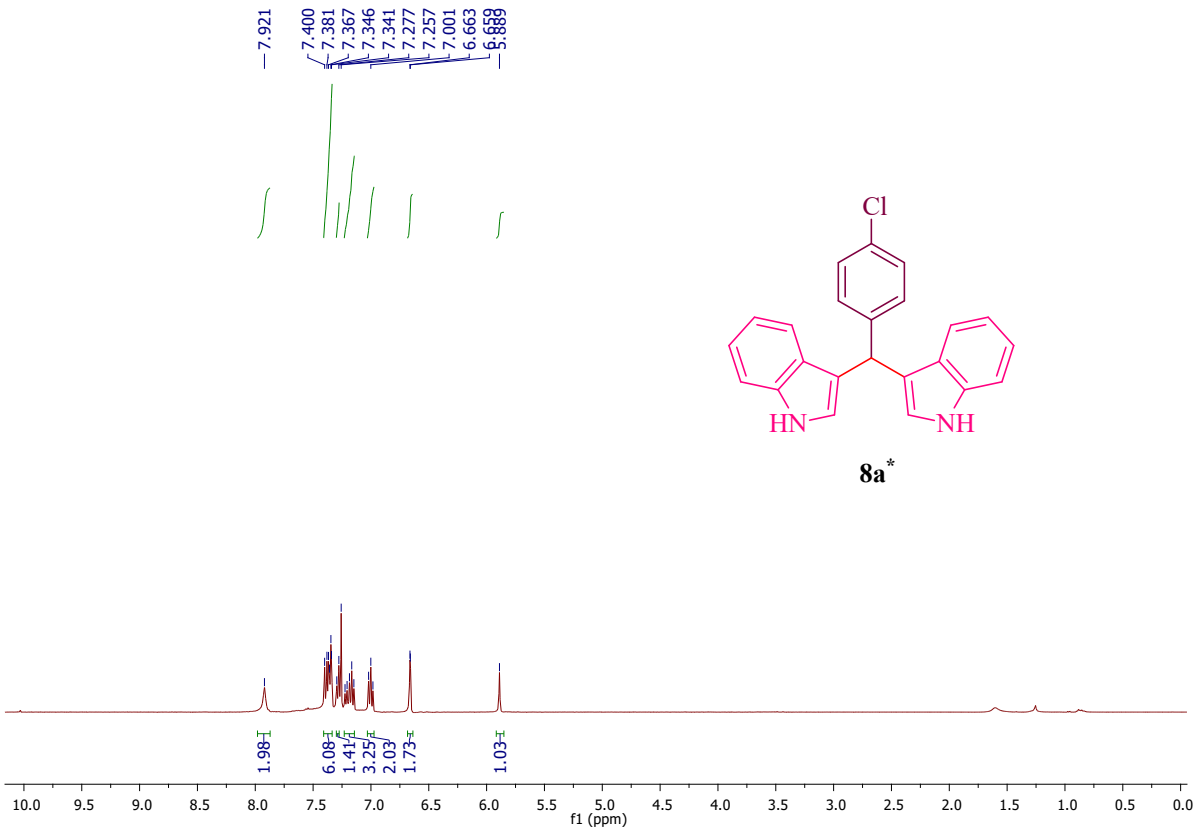
8m


8.127
8.105
7.396
7.374
7.255
7.015
6.994
6.555
6.423
5.639
3.710
2.825
1.95
2.17
1.06
1.23
1.04
3.75
2.12
0.94
1.11
3.13
3.12
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5
f1 (ppm)

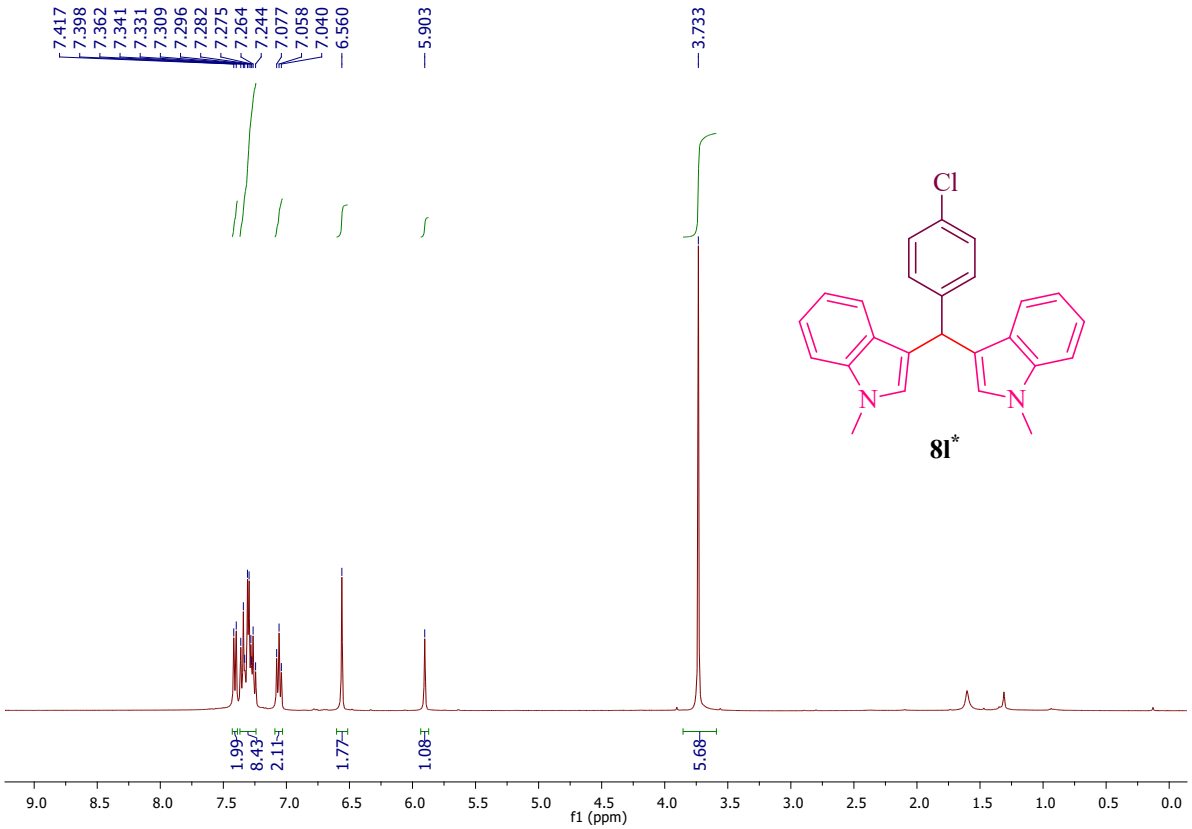
8n


152.734
148.055
146.349
137.490
131.217
129.700
129.657
128.747
127.021
123.555
121.891
119.730
119.074
117.448
112.508
109.316
77.364
77.047
76.729
47.872
32.763
30.985
O2N
N
N
190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)

8n


7.921
7.400
7.381
7.367
7.346
7.341
7.277
7.257
7.001
6.663
6.659
6.655
5.889
Cl
HN
NH
1.98
6.08
1.41
3.25
2.03
1.73
1.03
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)

8a*


7.417
7.398
7.362
7.341
7.331
7.309
7.296
7.282
7.275
7.264
7.244
7.077
7.058
7.040
6.560
5.903
3.733
Cl
N
N
1.99
8.43
2.11
1.77
1.08
5.68
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

8l*