Electronic Supplementary Material (ESI) for New Journal of Chemistry.

Preparation, characterization and catalytic application of nano-Fe_3O_4-DOPA-SnO_2 having high TON and TOF for non-toxic and sustainable synthesis of dihydroquinazolinone derivatives

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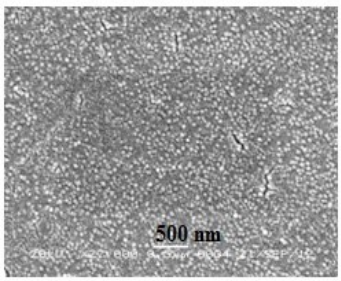
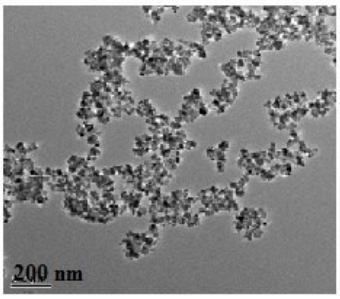
General Remarks:

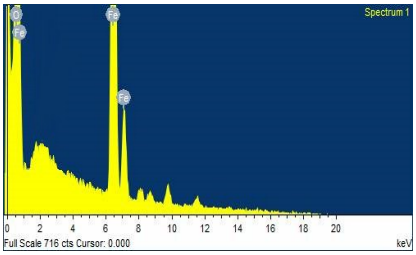
Melting points were determined in open capillaries and are uncorrected. IR spectra were recorded on Spectrum BX FT-IR, Perkin Elmer (υ_{max} in cm^{-1}) on KBr disks. ^{1}H NMR and ^{13}C NMR (400 MHz and 100 MHz respectively) spectra were recorded on Bruker Avance II-400 spectrometer in $CDCl_3$ and $CDCl_3$+ DMSO-d_6 (chemical shifts in δ with TMS as internal standard). Mass spectra were recorded using Waters ZQ-4000. Transmission Electron Microscope (TEM) images were recorded on JEOL JSM 100CX. Scanning electron microscope (SEM) and Energy Dispersive X-ray (EDX) images were obtained by using JSM-6360 (JEOL). ICP-AES analysis was performed by using ARCOS, Simultaneous ICP Spectrometer, Powder XRD was recorded on Bruker D8 XRD instrument SWAX. Thermogravimetric analysis (TGA) was recorded on a Perkin Elmer Precisely STA 6000 simultaneous thermal analyzer. CHN were recorded on CHN-OS analyzer (Perkin Elmer 2400, Series II). Silica gel G (E-merck, India) was used for TLC.



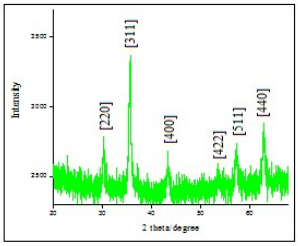
(a)



(b)



(c)



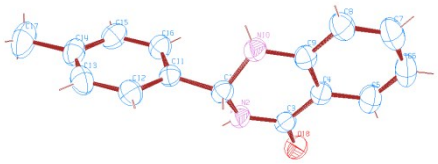
(d)

Fig SI 1: (a) SEM, (b) TEM, (c) EDX and (d) powder XRD of Fe_3O_4 NPS

X-ray crystallography

X-ray diffraction data were collected at 293 K with Mo Kα radiation (λ = 0.71073 Å) using Agilent Xcalibur (Eos, Gemini) diffractometer equipped with a graphite monochromator. Software used for data collection are CrysAlis PRO (Agilent, 2011), data reduction CrysAlis PRO and cell refinement CrysAlis PRO. Structures were solved by direct methods and refined by full-matrix least-squares calculation using SHELXS-97 and SHELXL-97.

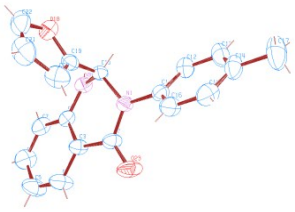
Table S.I.1.X-ray crystallography data for compound **6f** (**CCDC 1063941**).



Empirical formula	$C_{15}H_{14}N_2O$
Formula weight	238.29
Crystal system	Monoclinic
Space group	$P2_1/C$
a(Å)	13.027(2)
b(Å)	8.8840(12)
c(Å)	11.4246(17)
α(°)	90
β(°)	109.209(18)
γ(°)	90
Volume (Å)	1248.5(4)
T(K)	289.84(17)

Absorption coefficient (μ/mm^{-1})	0.081
Total reflection collected	5990
Independent reflection	2884[R(int) = 0.0211]
2θ range (°)	6.16 to 57.54°
Final R Indexes [1>=2σ(I)]	R_1 = 0.0524, wR_2 = N/A
Final R indexes [all data]	R_1 = 0.0758, wR_2 = 0.1531
Goodness-of-fit on F^2	1.062

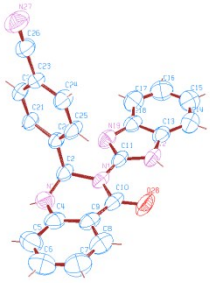
Table S.I.2.X-ray crystallography data for compound **6k** (**CCDC 1524323**).



Empirical formula	$C_{19}H_{16}N_2O_2$
Formula weight	304.35
Crystal system	Monoclinic
Space group	*P2$_1$/n*
a(Å)	14.3966(17)
b(Å)	6.8726(5)
c(Å)	17.724(2)
α(°)	90
β(°)	113.844(15)
γ(°)	90
Volume (Å)	1603.9(4)

T(K)	293.25(10)
Absorption coefficient (μ/mm^{-1})	0.083
Total reflection collected	15027
Independent reflection	3799[R(int) = 0.0427]
2θ range (°)	6.18 to 57.28°
Final R Indexes [1>=2σ(I)]	R_1 = 0.0719, wR_2 = N/A
Final R indexes [all data]	R_1 = 0.1018, wR_2 = 0.1570
Goodness-of-fit on F^2	1.074

Table S.I.3.X-ray crystallography data for compound **6m** (**CCDC 1535780**)

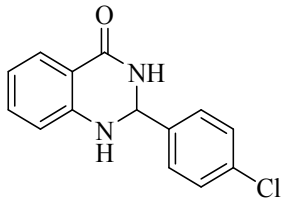


Empirical formula	$C_{22}H_{15}N_5O$
Formula weight	365.40
Crystal system	Monoclinic
Space group	C2/c
a(Å)	23.175(4)
b(Å)	9.9645(11)
c(Å)	16.245(2)
α(°)	90
β(°)	107.625(18)
γ(°)	90

Volume (Å)	3575.2(10)
T(K)	294.1(3)
Absorption coefficient (μ/mm^{-1})	0.088
Total reflection collected	7321
Independent reflection	3646 [R_{int} = 0.0275, R_{sigma} = 0.0502]
2θ range (°)	6.48 to 52.74°
Final R Indexes [1>=2σ(I)]	R_1 = 0.0552, wR_2 = 0.1072
Final R indexes [all data]	R_1 = 0.0958, wR_2 = 0.1284
Goodness-of-fit on F^2	1.074

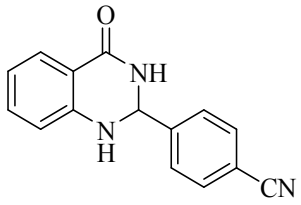
SPECTRAL DATA

1. Compound 6a



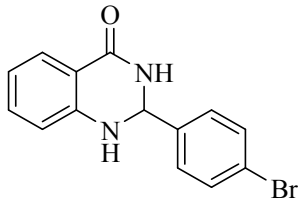
Light yellow solid. M.P.: 205-206 ^{0}C. IR (KBr): 3310, 3067, 1668, 1656, 752 cm^{-1}. ^{1}H NMR (DMSO-d_6, 400 MHz): δ = 7.71 (d, *J* = 8.0 Hz, 1H), 7.40 (d, *J* = 8.0 Hz, 2H), 7.27 (t, *J* = 8.0 Hz, 2H), 7.16 (t, *J* = 6.0 Hz, 1H), 6.71-6.65 (m, 2H), 6.60 (d, *J* = 8.0 Hz, 1H), 5.71 (s, 1H), 5.56 (brs,1H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 159.5, 142.1, 132.7, 130.0, 129.3, 128.6, 123.7, 123.5, 123.0, 122.1, 120.9, 113.8, 109.4, 62.7. ESI- MS: *m/z* 259, 261 $[M + H]^+$. Anal. Cacld for $C_{14}H_{11}ClN_2O$: C, 65.00; H, 4.29; N, 10.83. Found: C, 65.18; H, 4.35; N, 10.62.

2. Compound 6b



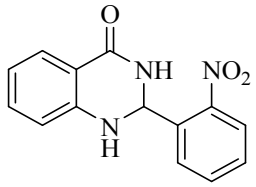
Light brown solid. M.P.: 250-252 ^{0}C. IR (KBr): 3354, 3337, 2228, 1667 cm^{-1}. ^{1}H NMR (DMSO-d_6, 400 MHz): δ = 8.18 (brs, 1H), 7.81 (d, *J* = 4.0 Hz, 1H), 7.71 (brs, 4H), 7.28 (t, *J* = 8.0 Hz, 1H), 6.91 (brs, 1H), 6.79 (d, *J* = 8.0 Hz, 1H), 6.75 (t, *J* = 8.0 Hz, 1H), 5.89 (s, 1H). ^{13}C NMR ($CDCl_3$ + DMSO-d_6, 100 MHz): δ = 163.9, 146.8, 146.4, 133.3, 131.8, 127.4, 118.1, 117.5, 114.5, 114.3, 111.4, 65.81. ESI- MS: *m/z* 250 $[M + H]^+$. Anal. Cacld for $C_{15}H_{11}N_3O$: C, 72.28; H, 4.45; N, 16.86. Found: C, 72.36; H, 4.56; N, 16.62.

3. Compound 6c



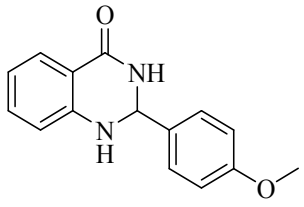
Light yellow solid. M.P.: 206-207 ^{0}C. IR (KBr): 3310, 1655 cm $^{-1}$. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 7.95 (d, *J* = 8.0 Hz, 1H), 7.60 (d, *J* = 8.0 Hz, 2H), 7.50 (d, *J* = 8.0 Hz, 2H), 7.37 (t, *J* = 6.0 Hz, 1H), 6.94 (t, *J* = 6.0 Hz, 1H), 6.69 (d, *J* = 8.0 Hz, 1H), 5.89 (s, 1H), 5.86 (brs, 1H), 4.38 (brs, 1H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 164.6, 146.9, 137.6, 134.2, 132.3, 129.1, 128.7, 119.9, 114.6, 68.5. ESI- MS: *m/z* 303, 305 $[M+H]^+$. Anal. Cacld for $C_{14}H_{11}BrN_2O$: C, 55.47; H, 3.66; N, 9.24. Found: C, 55.68; H, 3.95; N, 9.15.

4. Compound 6d



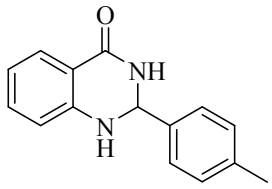
Light yellow solid. M.P.: 193-194 ^{0}C. IR (KBr): 3411, 1658, 1507, 1348 cm^{-1}. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 7.98 (d, *J* = 8.0 Hz, 1H), 7.86 (t, *J* = 8.0 Hz, 2H), 7.62 (t, *J* = 8.0 Hz, 1H), 7.48 (t, *J* = 8.0 Hz, 1H), 7.27 (t, *J* = 8.0 Hz, 1H), 6.81 (t, *J* = 8.0 Hz, 1H), 6.58 (d, *J* = 4.0 Hz, 1H), 6.51 (brs, 1H), 6.36 (s, 1H), 4.98 (brs, 1H). ^{13}C NMR ($CDCl_3$ + DMSO-d_6, 100 MHz): δ = 163.4, 146.6, 145.4, 135.6, 133.16, 133.13, 128.6, 127.8, 127.0, 124.0, 117.5, 113.6, 113.3, 62.0. ESI- MS: *m/z* 270 $[M + H]^+$. Anal. Cacld for $C_{14}H_{11}N_3O_3$: C, 62.45; H, 4.12; N, 15.61. Found: C, 62.41; H, 4.05; N, 15.62.

5. Compound 6e



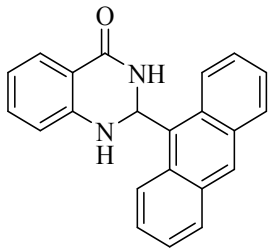
Light yellow solid. M.P.: 192-193 ^{0}C. IR (KBr): 3456, 3301, 1653, 1247 cm^{-1}. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 7.96 (d, *J* = 8.0 Hz, 1H), 7.54 (d, *J* = 8.0 Hz, 2H), 7.36 (t, *J* = 8.0 Hz, 1H), 6.97 (d, *J* = 8.0 Hz, 2H), 6.93 (t, *J* = 8.0 Hz, 1H), 6.68 (d, *J* = 8.0 Hz, 2H), 5.86 (s, 1H), 5.75 (brs,1H), 4.36 (brs, 1H), 3.85 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 164.8, 160.9, 147.3, 134.0, 130.5, 128.8, 128.7, 119.6, 114.5, 114.3, 68.6, 55.4. ESI- MS: *m/z* 255 [M + H]$^+$. Anal. Cacld for $C_{15}H_{14}N_2O_2$: C, 70.85; H, 5.55; N, 11.02. Found: C, 70.68; H, 5.59; N, 10.88.

6. Compound 6f



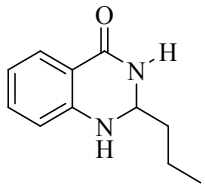
Light yellow solid. M.P.: 233-235 ^{0}C. IR (KBr): 3314, 2926, 1673, 1659 cm^{-1}. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 7.64 (d, *J* = 8.0 Hz, 1H), 7.30 (d, *J* = 4.0 Hz, 2H), 7.11-7.04 (m, 3H), 6.97 (brs, 1H), 6.60 (d, *J* = 8.0 Hz, 2H), 5.64 (s, 1H), 2.93 (brs, 1H), 2.20 (s, 3H). ^{13}C NMR ($CDCl_3$ + DMSO-d_6, 100 MHz): δ = 169.5, 152.9, 143.6, 142.1, 138.3, 134.0, 132.6, 132.0, 122.7, 119.9, 119.5, 72.7, 25.9. ESI- MS: *m/z* 239 [M + H]$^+$. Anal. Cacld for $C_{15}H_{14}N_2O$: C, 75.61; H, 5.92; N, 11.76. Found: C, 75.78; H, 5.95; N, 11.67.

7. Compound 6g



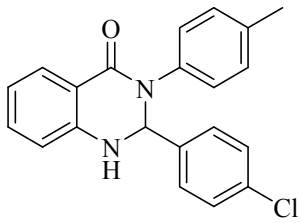
Yellow solid. M.P.: 237-239 ^{0}C. IR (KBr): 3366, 3203, 1678 cm^{-1}. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 8.50 (s, 1H), 8.04-7.98 (m, 3H), 7.45 (s, 5H), 7.35 (t, *J* = 6.0Hz, 1H), 7.27 (d, *J* = 8.0 Hz, 1H), 7.10-7.01 (m, 1H), 6.95 (t, *J* = 8.0 Hz, 1H), 6.65-6.60 (m, 1H), 6.13 (s, 1H), 4.56 (s, 1H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 165.9, 148.3, 134.2, 132.6, 131.7, 131.0, 130.5, 129.4, 129.1, 125.3, 125.2, 119.7, 117.2, 116.0, 114.7, 63.9. ESI- MS: *m/z* 325 $[M + H]^+$. Anal. Cacld for $C_{22}H_{16}N_2O$: C, 81.46; H, 4.97; N, 8.64. Found: C, 81.73; H, 5.05; N, 8.86.

8. Compound 6h



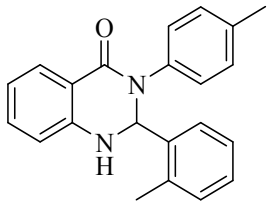
Red solid. M.P.: 173-175 ^{0}C. IR (KBr): 3214,3021, 1670 cm^{-1}. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 7.89 (d, *J* = 4.0 Hz, 1H), 7.31 (t, *J* = 4.0 Hz, 1H), 6.87 (t, *J* = 6.0 Hz, 1H), 6.67 (d, *J* = 4.0 Hz, 1H), 6.24 (brs, 1H), 4.90 (t, *J* = 4.0 Hz, 1H), 4.22 (brs, 1H), 1.78 (q, 2H), 1.52-1.46 (m, 2H), 1.01 (t, *J* = 4.0 Hz, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 165.7, 147.6, 134.1, 128.8, 119.6, 116.1, 114.9, 65.3, 37.8, 17.6, 14.0. ESI- MS: *m/z* 191 $[M + H]^+$. Anal. Cacld for $C_{11}H_{14}N_2O$: C, 69.45; H, 7.42; N, 14.73. Found: C, 69.39; H, 7.67; N, 14.59.

9. Compound 6i



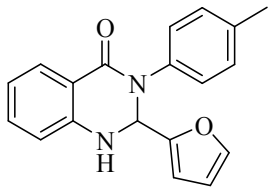
Light yellow solid. M.P.: 243-245 ^{0}C. IR (KBr): 3306, 3064, 1667 cm^{-1}. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 8.03 (d, *J* = 12.0 Hz, 1H), 7.34-7.23 (m, 5H), 7.11 (d, *J* = 8.0 Hz, 2H), 7.06 (d, *J* = 8.0 Hz, 2H), 6.92 (t, *J* = 6.0 Hz, 1H), 6.65 (d, *J* = 8.0 Hz, 1H), 6.06 (s, 1H), 4.73 (brs, 1H), 2.29 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 163.1, 145.0, 138.4, 137.6, 136.8, 134.7, 133.9, 129.7, 129.0, 128.9, 128.2, 126.7, 119.8, 116.8, 114.9, 74.0, 21.0. ESI- MS: *m/z* 349, 351 $[M + H]^+$. Anal. Cacld for $C_{21}H_{17}ClN_2O$: C, 72.31; H, 4.91; N, 8.03. Found: C, 72.58; H, 4.95; N, 7.92.

10. Compound 6j



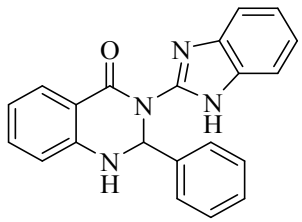
Light yellow solid. M.P.: 201-204 ^{0}C. IR (KBr): 3297, 2926, 1632 cm $^{-1}$. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 7.98 (d, *J* = 8.0 Hz, 1H), 7.51 (d, *J* = 8.0 Hz, 1H), 7.24-7.23 (m, 1H), 7.12-7.04 (m, *2*H), 7.01-6.96 (m, 4H), 6.83 (t, *J* = 6.0 Hz, 2H), 6.54 (d, *J* =12.0 Hz, 1H) 6.31 (s, 1H), 4.50 (brs, 1H), 2.24 (s, 3H), 2.19 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 158.5, 140.2, 132.3, 131.9, 131.4, 129.8, 128.5, 125.8, 124.2, 123.7, 123.5, 122.3, 121.6, 121.1, 114.1, 111.3, 109.5, 66.3, 15.8, 14.0. ESI- MS: *m/z* 329 $[M + H]^+$. Anal. Cacld for $C_{22}H_{20}N_2O$: C, 80.74; H, 6.14; N, 8.53. Found: C, 80.59; H, 5.98; N, 8.38.

11. Compound 6k



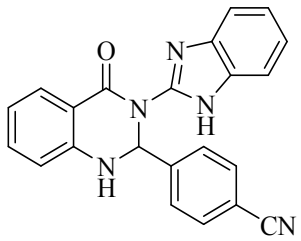
Light yellow solid. M.P.: 161-163 ^{0}C. IR (KBr): 3356, 3113, 1673 cm^{-1}. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 7.95 (d, *J* = 8.0 Hz, 1H), 7.27 (t, *J* = 8.0 Hz, 2H), 7.20 (brs, 3H), 7.12 (d, *J* = 8.0 Hz, 2H), 6.86 (t, *J* = 8.0 Hz, 1H), 6.64 (d, *J* = 8.0 Hz, 1H), 6.25 (brs, 1H), 6.18 (brs, 1H), 5.96 (s, 1H), 2.28 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 162.6, 152.2, 145.1, 142.7, 138.0, 136.7, 133.6, 129.6, 128.9, 126.1, 119.9, 117.3, 115.0, 110.4, 109.0, 68.6, 21.0. ESI-MS: *m/z* 305 $[M + H]^+$. Anal. Cacld for $C_{19}H_{16}N_2O_2$: C, 74.98; H, 5.30; N, 9.20. Found: C, 74.77; H, 5.55; N, 9.33.

12. Compound 6l



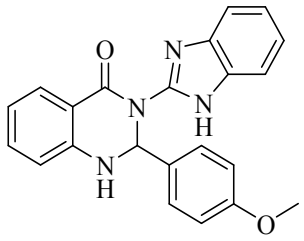
Off white. solid. M.P.: 102-105 ^{0}C. IR (KBr): 3336, 3109, 1694 cm^{-1}. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 11.69 (brs, 1H), 8.00 (d, *J* = 8.0 Hz, 1H), 7.63 (d, *J* = 4.0 Hz, 2H), 7.46-7.37 (m, 4H), 7.27-7.22 (m, 5H), 6.95 (t, *J* = 8.0 Hz, 1H), 6.84 (d, *J* = 12.0 Hz, 1H), 5.35 (d, *J* = 4.0 Hz, 1H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 163.3, 150.4, 145.5, 135.2, 134.1, 131.2, 129.1, 129.0, 128.6, 128.4, 126.0, 122.3, 120.0, 116.7, 116.3, 116.1, 110.7, 67.7. ESI- MS: *m/z* 341 $[M + H]^+$. Anal. Cacld for $C_{21}H_{16}N_4O$: C, 74.10; H, 4.74; N, 16.46. Found: C, 74.03; H, 4.79; N, 16.28.

13. Compound 6m



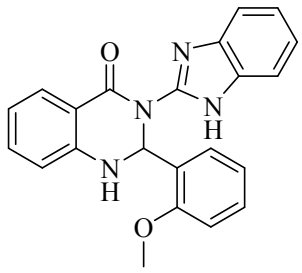
Light yellow solid. M.P.: 143-145 ^{0}C. IR (KBr): 3106, 2228, 1697 cm^{-1}. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 11.6 (brs, 1H), 8.03 (d, *J* = 8.0 Hz, 1H), 7.69 (d, *J* = 4.0 Hz, 1H), 7.65-7.64(m, 1H), 7.55 (s, 4H), 7.50 (t, *J* = 8.0 Hz, 3H), 7.31 (s, 1H), 7.04 (t, *J* = 8.0 Hz, 1H), 6.94 (d, *J* = 8.0 Hz, 1H), 5.33 (d, *J* = 4.0 Hz, 1H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 162.8, 147.8, 144.7, 144.5, 139.9, 135.5, 132.9, 132.4, 132.1, 129.9, 129.3, 127.0, 122.6, 120.9, 118.3, 118.0, 116.9, 116.7, 112.4, 110.9, 67.0. ESI- MS: *m/z* 366 $[M + H]^+$. Anal. Cacld for $C_{22}H_{15}N_5O$: C, 72.32; H, 4.14; N, 19.17. Found: C, 72.29; H, 4.11; N, 18.98.

14. Compound 6n



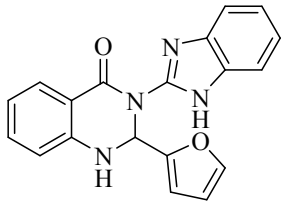
Brown solid. M.P.: 291-293 ^{0}C. IR (KBr): 3326, 3103, 1695 cm^{-1}. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 11.67 (brs, 1H), 7.99 (d, *J* = 4.0 Hz, 1H), 7.63 (d, *J* = 4.0 Hz, 1H), 7.57 (d, *J* = 4.0 Hz, 1H), 7.44-7.38 (m, 2 H), 7.30 (d, *J* = 4.0 Hz, 2H), 7.24-7.21 (m, 2H), 6.93 (t, *J* = 6.0 Hz, 1H), 6.82 (d, *J* = 8.0 Hz, 1H), 6.74 (d, *J* = 8.0 Hz, 2H), 5.22 (s, 1H), 3.70 (s, 3H). ^{13}C NMR ($CDCl_3$ +DMSO-d_6, 100 MHz): δ = 163.3, 159.2, 146.6, 134.9, 131.8, 131.4, 128.7, 127.3, 121.9, 118.6, 115.7, 114.3, 114.2, 113.6, 113.1, 67.2, 55.0. ESI- MS: *m/z* 371 $[M + H]^+$. Anal. Cacld for $C_{22}H_{18}N_4O_2$: C, 71.34; H, 4.90; N, 15.13. Found: C, 71.14; H, 5.07; N, 15.04.

15. Compound 6o



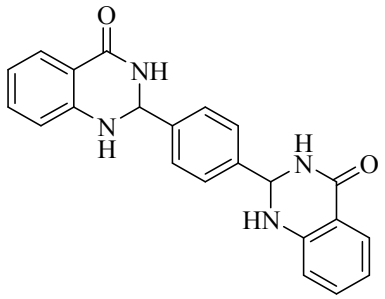
Brown solid. M.P.: 275-277 ^{0}C. IR (KBr): 3325, 3110, 1691 cm^{-1}. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 11.79 (brs, 1H), 8.00 (d, *J* = 4.0 Hz, 1H), 7.67 (d, *J* = 1.6 Hz, 1H), 7.57-7.55 (m, 1H), 7.44-7.42 (m, 1H), 7.21-7.19 (m, 4H), 6.99 (d, *J* = 8.0 Hz, 1H), 6.93 (d, *J* = 8.0 Hz, 1H), 6.84 (t, *J* = 6.0 Hz, 1H), 6.75 (t, *J* = 6.0 Hz, 1H), 6.61 (d, *J* = 8.0 Hz, 1H), 5.70 (s, 1H), 4.01 (s, 3H). ESI- MS: *m/z* 371 $[M + H]^+$. Anal. Cacld for $C_{22}H_{18}N_4O_2$: C, 71.34; H, 4.90; N, 15.13. Found: C, 71.12; H, 5.07; N, 15.25.

16. Compound 6p



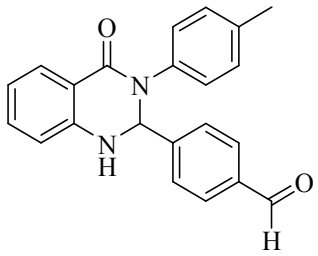
Light Brown solid. M.P.: 148-150 ^{0}C. IR (KBr): 3416, 3093, 1636, 1662 cm^{-1}. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 11.59 (brs, 1H), 8.02 (d, *J* = 4.0 Hz, 1H), 7.65 (d, *J* = 8.0 Hz, 1H), 7.57 (d, *J* = 3.2 Hz, 1H), 7.47-7.39 (m, 3H), 7.25-7.23 (m, 2H), 6.97 (t, *J* = 6.0 Hz, 1H), 6.81 (d, *J* = 4.0 Hz, 1H), 6.17 (s, 2H), 5.19 (s, 1H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 168.8, 142.5, 141.9, 134.5, 134.4, 133.0, 132.4, 131.6, 131.4, 130.6, 130.5, 130.1, 122.3, 122.1, 120.2, 111.8, 111.6, 110.3, 60.4. ESI- MS: *m/z* 331 $[M + H]^+$. Anal. Cacld for $C_{19}H_{14}N_4O_2$: C, 69.08; H, 4.27; N, 16.96. Found: C, 68.89; H, 4.08; N, 17.09.

17. Compound 6q



Light brown solid. M.P.: 244-246 ^{0}C. IR (KBr): 3416, 3212, 1663, 1669 cm^{-1}. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 10.04 (s, 1H), 7.94 (d, *J* = 8.0 Hz, 2H), 7.87 (d, *J* = 8.0 Hz, 2H), 7.78 (d, *J* = 8.0 Hz, 2H), 7.62 (s, 1H), 7.43 (s, 2H), 7.32 (t, *J* = 6.0 Hz, 2H), 6.91 (brs, 1H), 6.85 (t, *J* = 8.0 Hz, 2H), 6.77 (d, *J* = 8.0 Hz, 1H), 5.96 (s, 1H), 5.89 (s, 1H). ^{13}C NMR ($CDCl_3$ + DMSO-d_6, 100 MHz): δ = 164.0, 147.4, 141.6, 133.0, 129.3, 127.2, 126.6, 117.1, 114.7, 114.2, 66.3. ESI- MS: *m/z* 371 $[M + H]^+$. Anal. Cacld for $C_{22}H_{18}N_4O_2$: C, 71.34; H, 4.90; N, 15.13. Found: C, 71.18; H, 5.09; N, 15.07.

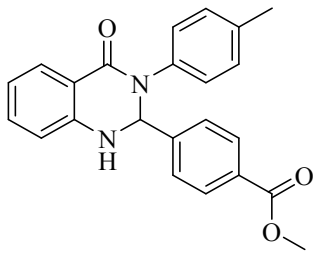
18. Compound 6r



Light brown solid. M.P.: 198-201 ^{0}C. IR (KBr): 3412, 3211, 1689 cm^{-1}. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 9.96 (s, 1H), 8.04 (d, *J* = 8.0 Hz, 1H), 7.81 (d, *J* = 8.0 Hz, 2H), 7.55 (d, *J* = 4.0 Hz, 2H), 7.35 (t, *J* =8.0 Hz, 1H), 7.23 (brs, 1H), 7.12-7.06 (m, 4H), 6.94 (t, *J* = 8.0 Hz, 1H), 6.67 (d, *J* = 8.0 Hz, 1H), 6.16 (s, 1H), 2.30 (s, 3H). ^{13}C NMR ($CDCl_3$ + DMSO-d_6, 100 MHz): δ = 196.5, 167.6, 152.0, 150.8, 142.9, 141.0, 140.9, 138.6, 134.6, 134.3, 134.1, 133.2,

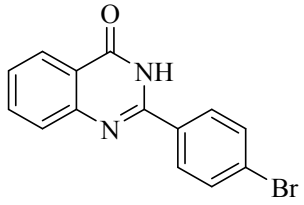
132.1, 130.9, 123.1, 120.6, 119.7, 78.2, 25.7. ESI- MS: *m/z* 343 $[M + H]^+$. Anal. Cacld for $C_{22}H_{18}N_2O_2$: C, 77.17; H, 5.30; N, 8.18. Found: C, 77.28; H, 5.09; N, 8.20.

19. Compound 6s



Brown solid. M.P.: 210-213 ^{0}C. IR (KBr): 3329, 3000, 1720, 1639 cm^{-1}. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 8.03 (d, *J* = 8.0 Hz, 1H), 7.95 (d, *J* = 8.0 Hz, 2H), 7.45 (d, *J* = 8.0 Hz, 2H), 7.33 (t, *J* = 8.0 Hz, 1H), 7.27 (brs, 1H), 7.11-7.05 (m, 4 H), 6.93 (t, *J* = 8.0 Hz, 1H), 6.65 (d, *J* = 8.0 Hz, 1H), 6.13 (s, 1H), 3.89 (s, 3H), 2.29 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 166.5, 163.0, 144.9, 144.7, 137.6, 136.9, 133.9, 130.6, 130.0, 129.7, 129.0, 126.8, 126.6, 119.8, 116.8, 114.9, 74.2, 52.2, 21.0. ESI- MS: *m/z* 373 $[M + H]^+$. Anal. Cacld for $C_{23}H_{20}N_2O_3$: C, 74.18; H, 5.41; N, 7.52. Found: C, 74.24; H, 5.39; N, 7.67.

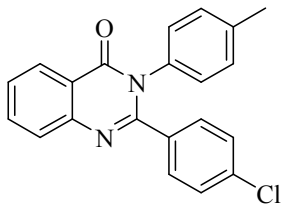
20. Compound 7a



Off white solid. M.P.: 319-321 ^{0}C. IR (KBr): 3309, 1639, 1671cm^{-1}. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 7.87 (d, *J* = 4.0 Hz, 1H), 7.53 (d, *J* = 8.0 Hz, 2H), 7.43 (d, *J* = 12.0 Hz, 2H), 7.30 (t, *J* = 8.0 Hz, 1H), 6.87 (t, *J* = 8.0 Hz, 1H), 6.62 (d, *J* = 8.0 Hz, 1H), 4.31 (brs, 1H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 160.3, 148.0, 147.8, 147.7, 147.3, 136.4, 133.5, 127.0, 126.2,

125.7, 125.2, 120.9. ESI- MS: *m/z* 301, 303 $[M + H]^+$. Anal. Cacld for $C_{14}H_9BrN_2O$: C, 55.84; H, 3.01; N, 9.30. Found: C, 56.01; H, 3.09; N, 9.07.s

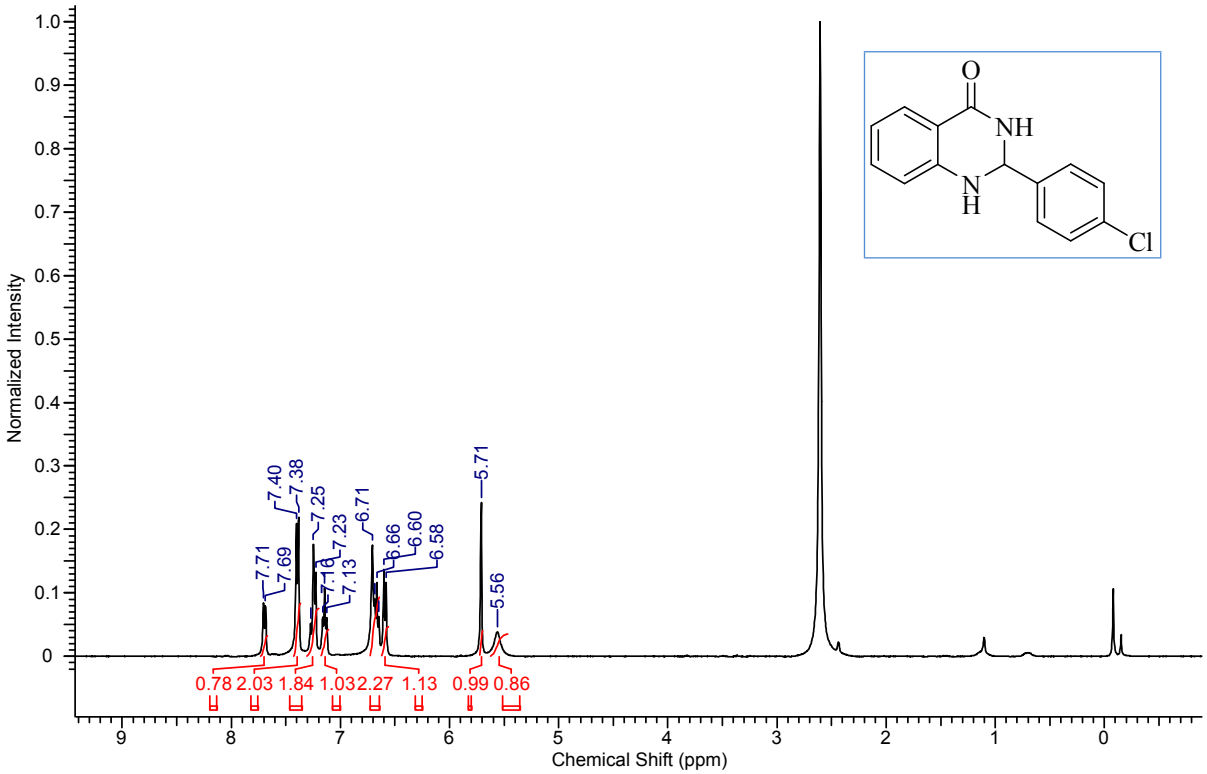
21. Compound 7b

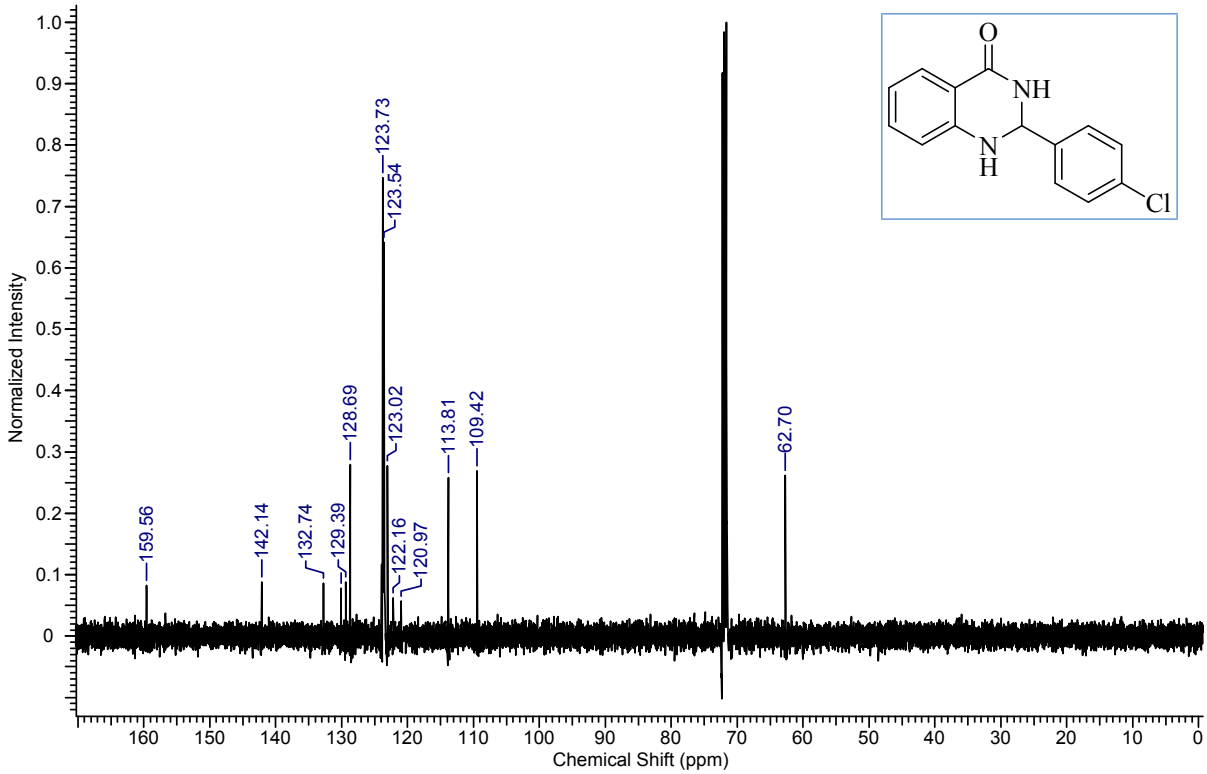


White solid. M.P.: 183-185 ^{0}C. IR (KBr): 2918, 1646, 1665 cm^{-1}. ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 8.28 (d, *J* = 8.0 Hz, 1H), 8.20 (d, *J* = 8.0 Hz, 2H), 7.78 (d, *J* = 4.0 Hz, 2H), 7.51-7.48 (m, 7H), 2.60 (3H). ^{13}C NMR (DMSO-d_6, 100 MHz): δ = 164.5, 147.9, 138.6, 137.1, 133.2, 128.9, 127.5, 126.9, 117.7, 114.8, 114.4, 20.9. ESI- MS: *m/z* 347, 349 $[M + H]^+$. Anal. Cacld for $C_{21}H_{15}ClN_2O$: C, 72.73; H, 4.36; N, 8.08. Found: C, 72.59; H, 4.57; N, 8.01.

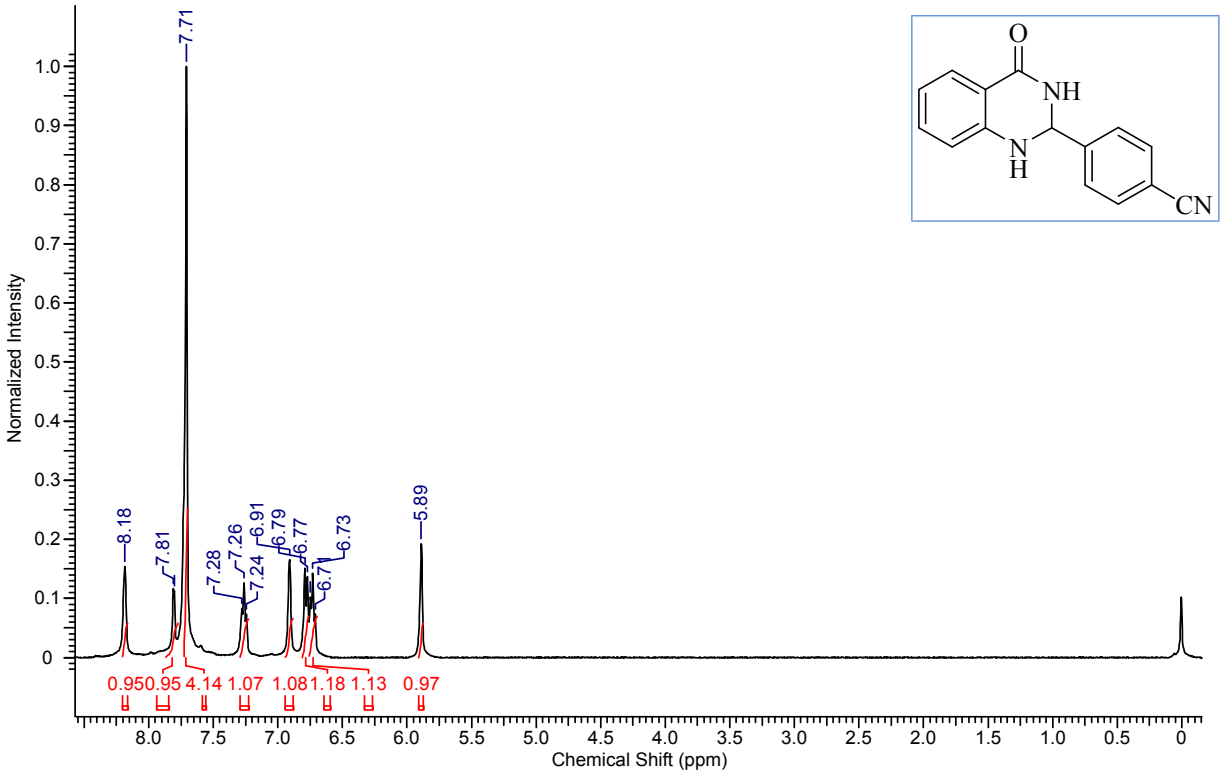
^{1}H and ^{13}C NMR spectra

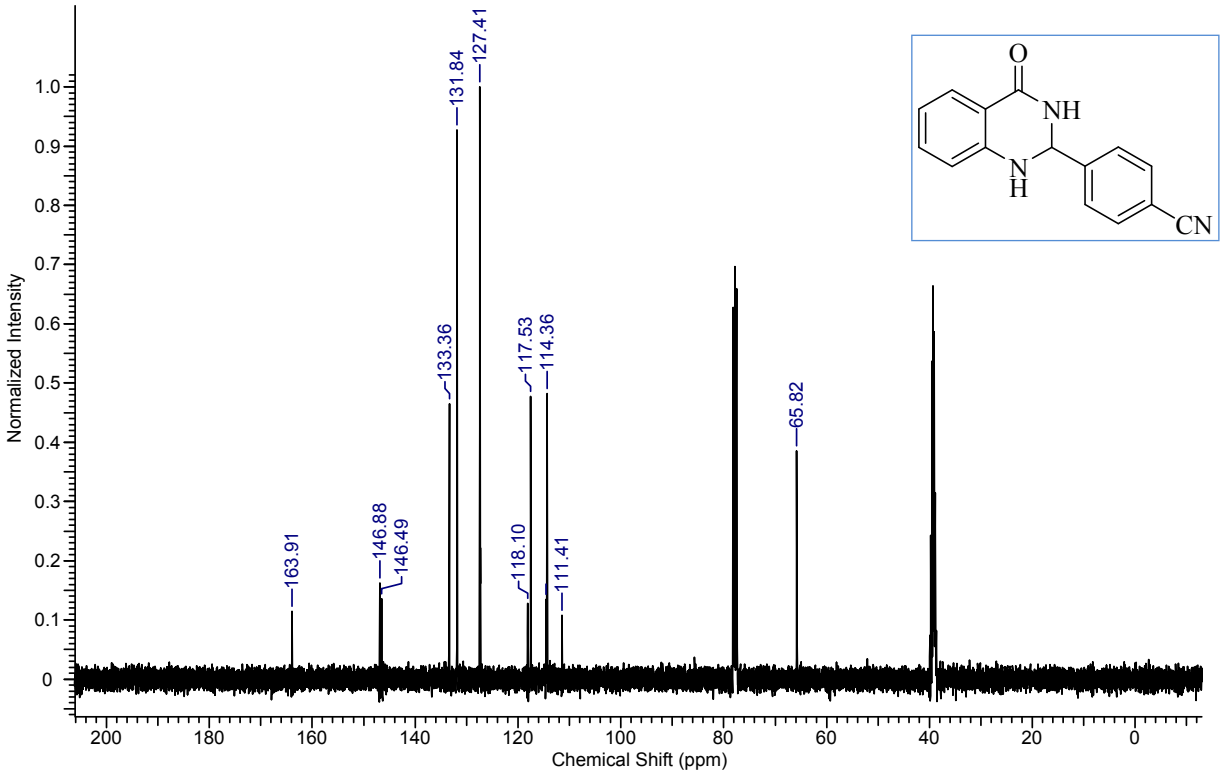
1. **6a**



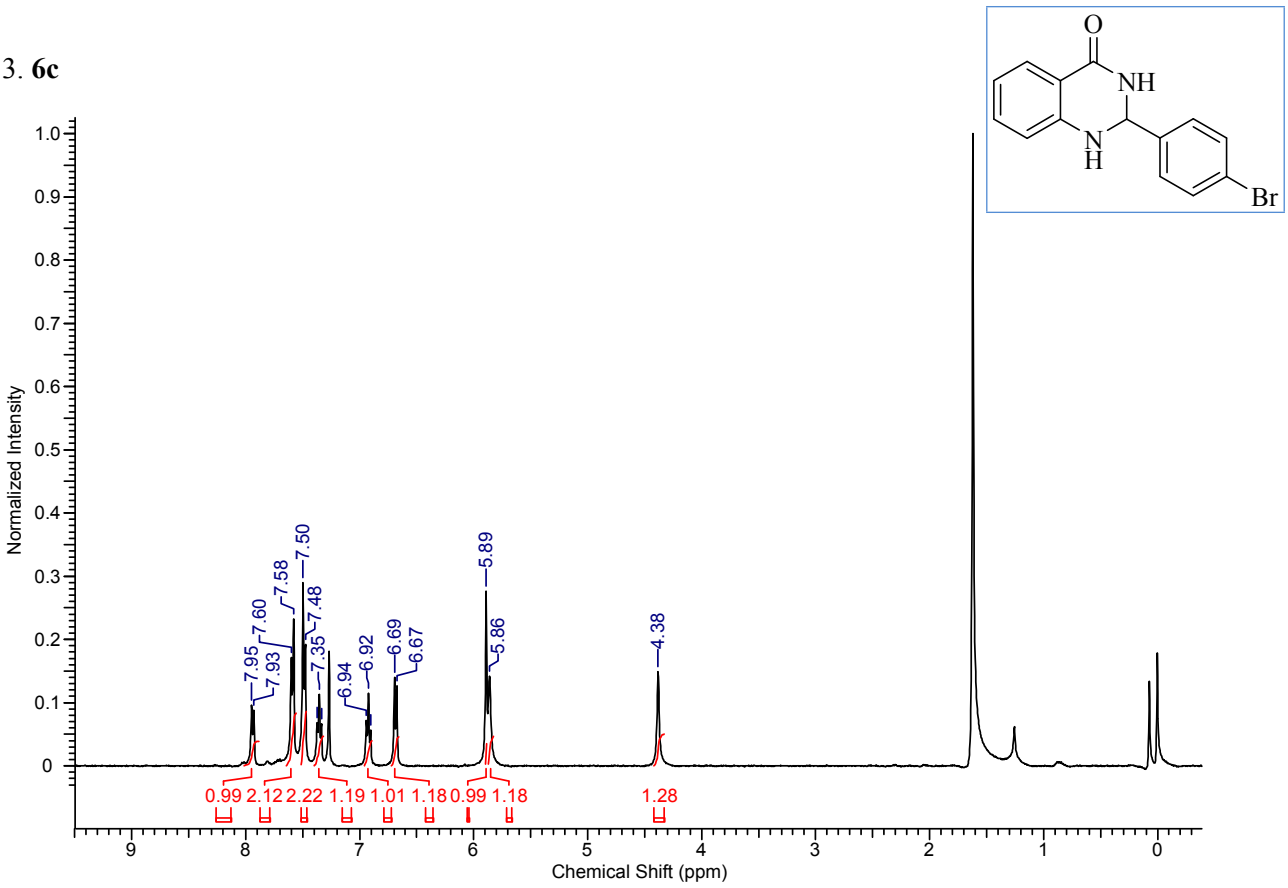


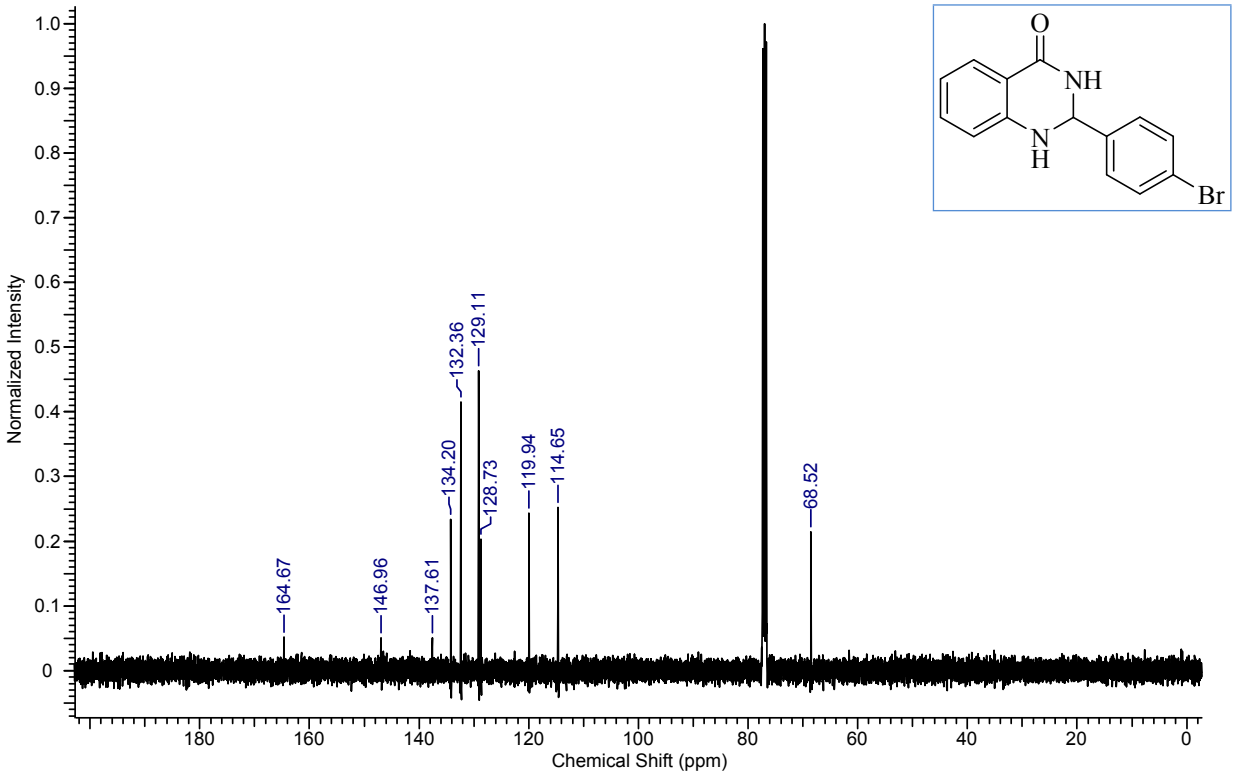
2. **6b**


Normalized Intensity
Chemical Shift (ppm)
7.71
8.18
7.81
7.28
7.26
7.24
6.91
6.79
6.77
6.73
6.71
5.89
0.95 0.95 4.14 1.07 1.08 1.18 1.13 0.97
O
NH
N
H
CN

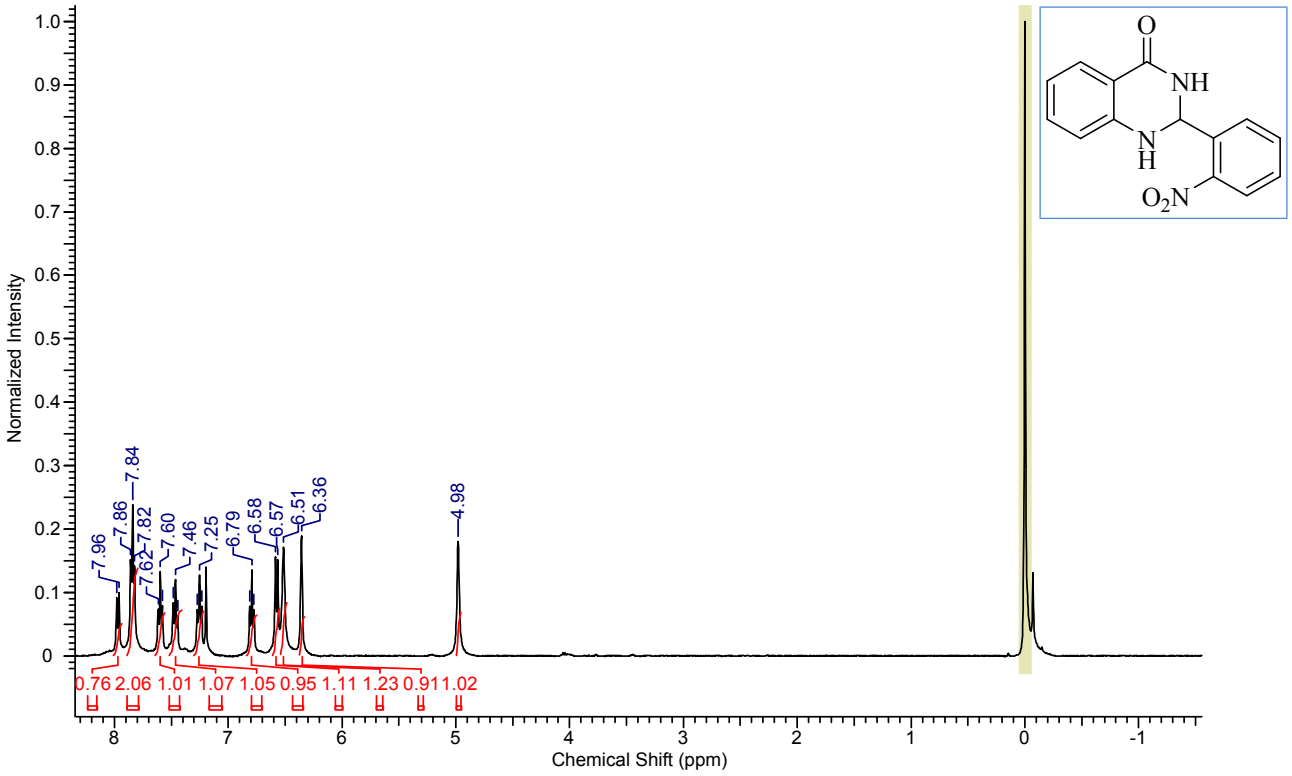

Normalized Intensity
Chemical Shift (ppm)
131.84
127.41
133.36
117.53
114.36
65.82
146.88
146.49
163.91
118.10
111.41
O
NH
N
H
CN

3. **6c**


O
NH
N
H
Br
Normalized Intensity
Chemical Shift (ppm)
7.95
7.93
7.60
7.58
7.50
7.48
7.35
6.94
6.92
6.69
6.67
5.89
5.86
4.38
0.99 2.12 2.22 1.19 1.01 1.18 0.99 1.18
1.28


O
NH
N
H
Br
Normalized Intensity
Chemical Shift (ppm)
164.67
146.96
137.61
134.20
132.36
129.11
128.73
119.94
114.65
68.52

4. **6d**


Normalized Intensity
Chemical Shift (ppm)
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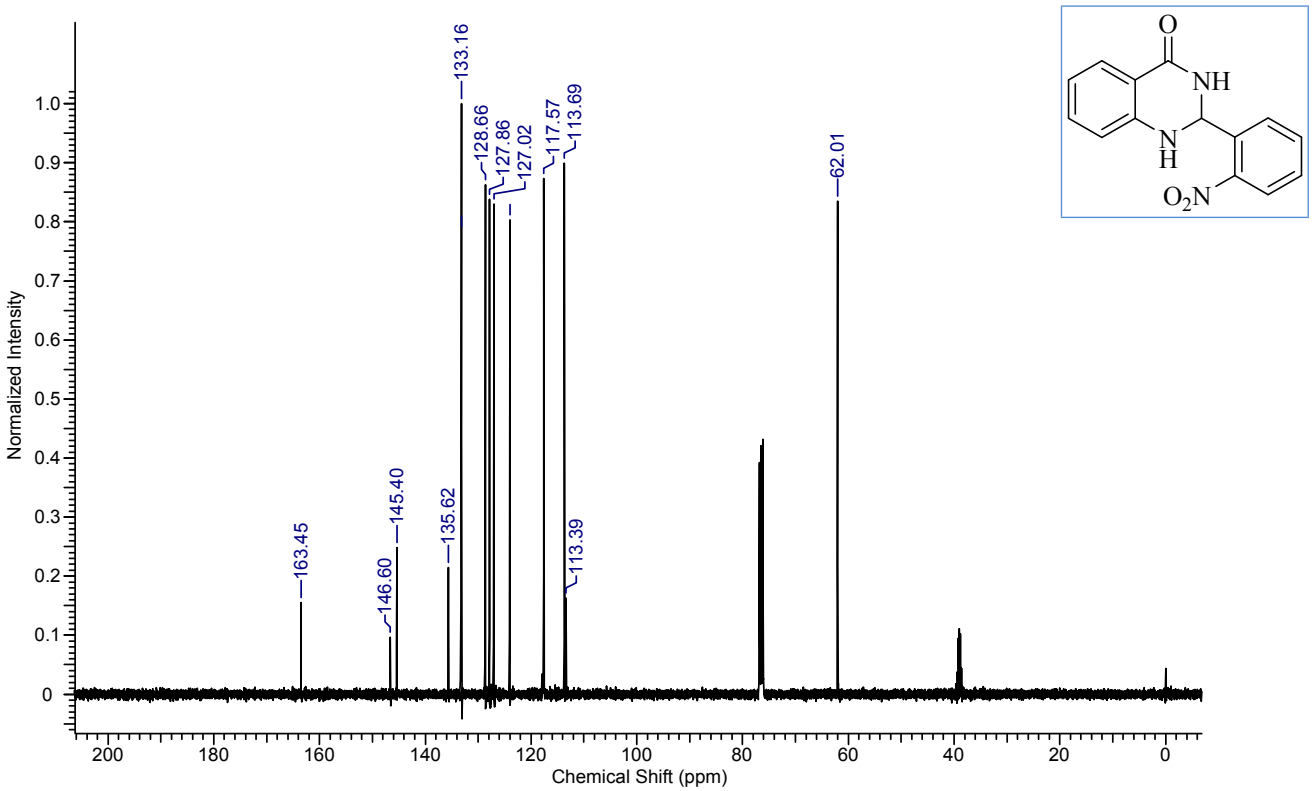
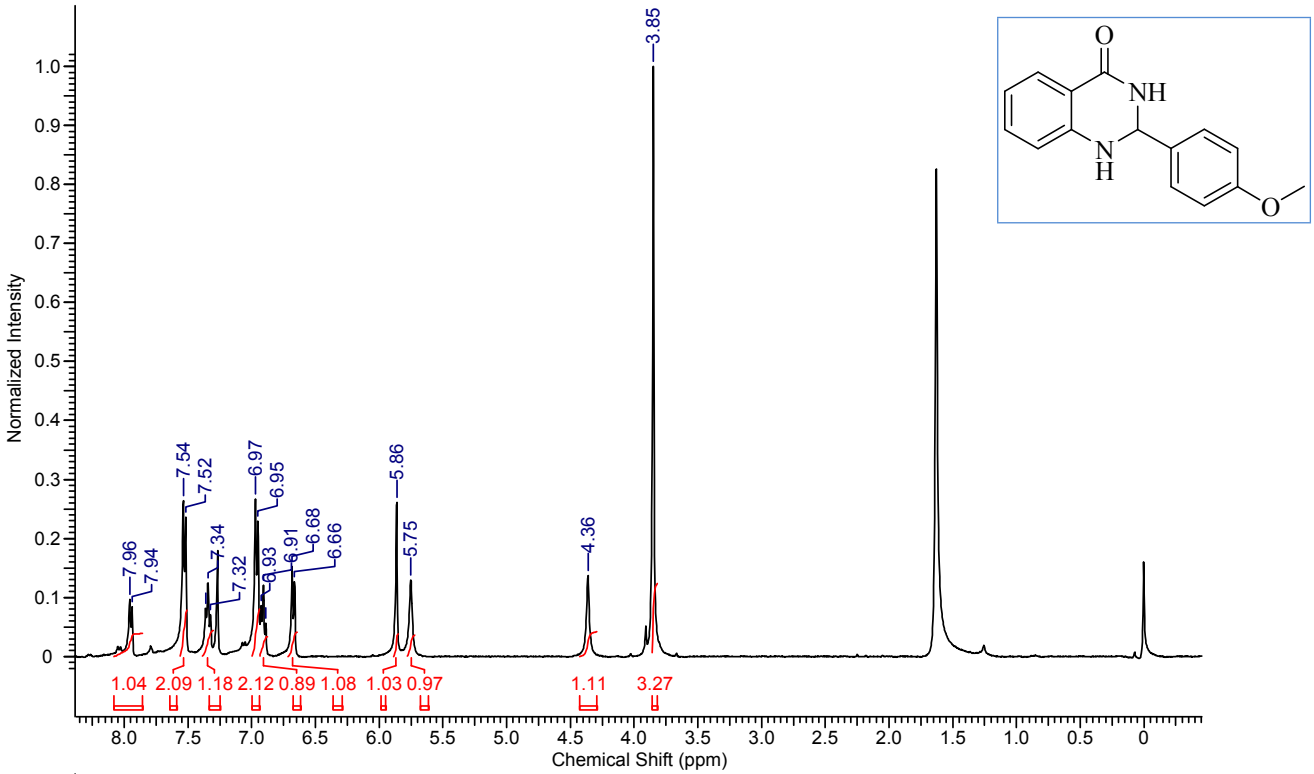
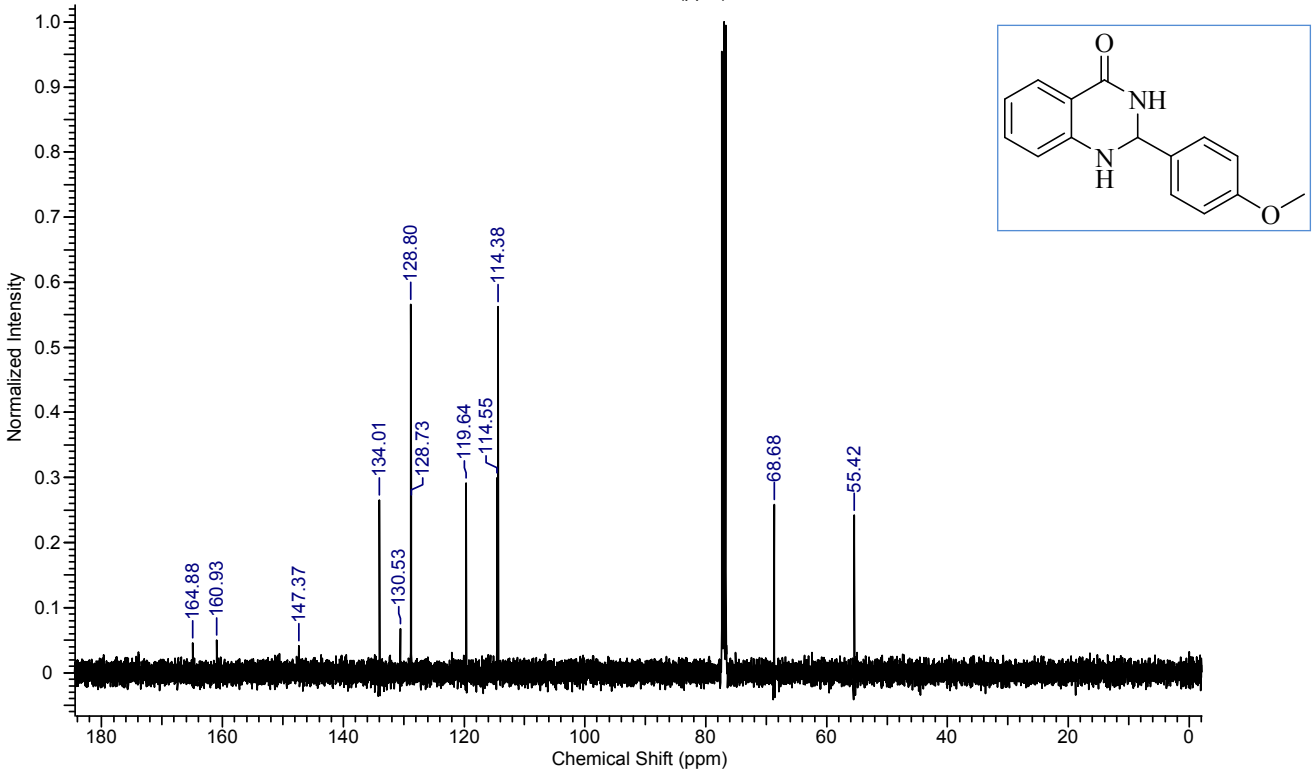
7.86
7.84
7.82
7.62
7.60
7.46
7.25
6.79
6.58
6.57
6.51
6.36
4.98
0.76 2.06 1.01 1.07 1.05 0.95 1.11 1.23 0.91 1.02
O
NH
N
H
O2N

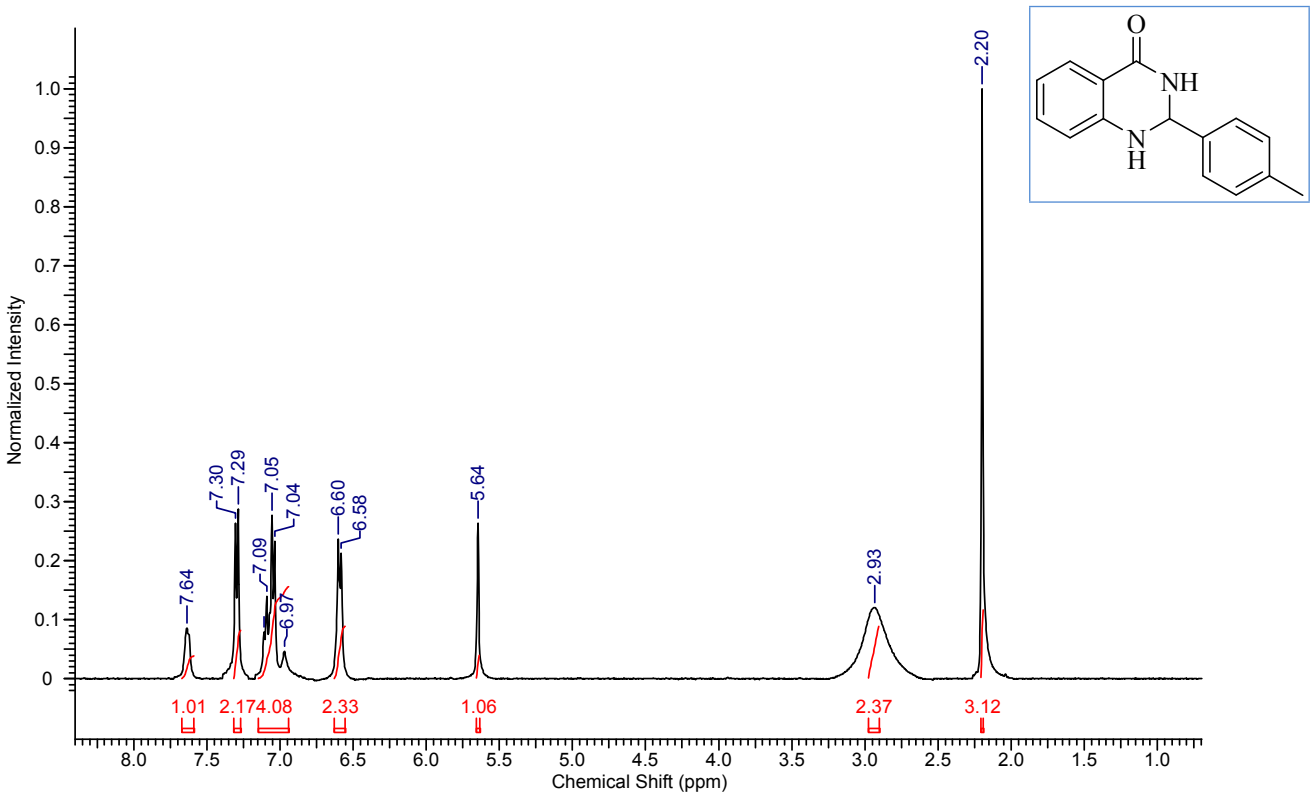

Normalized Intensity
Chemical Shift (ppm)
163.45
146.60
145.40
135.62
133.16
128.66
127.86
127.02
117.57
113.69
113.39
62.01
O
NH
N
H
O2N

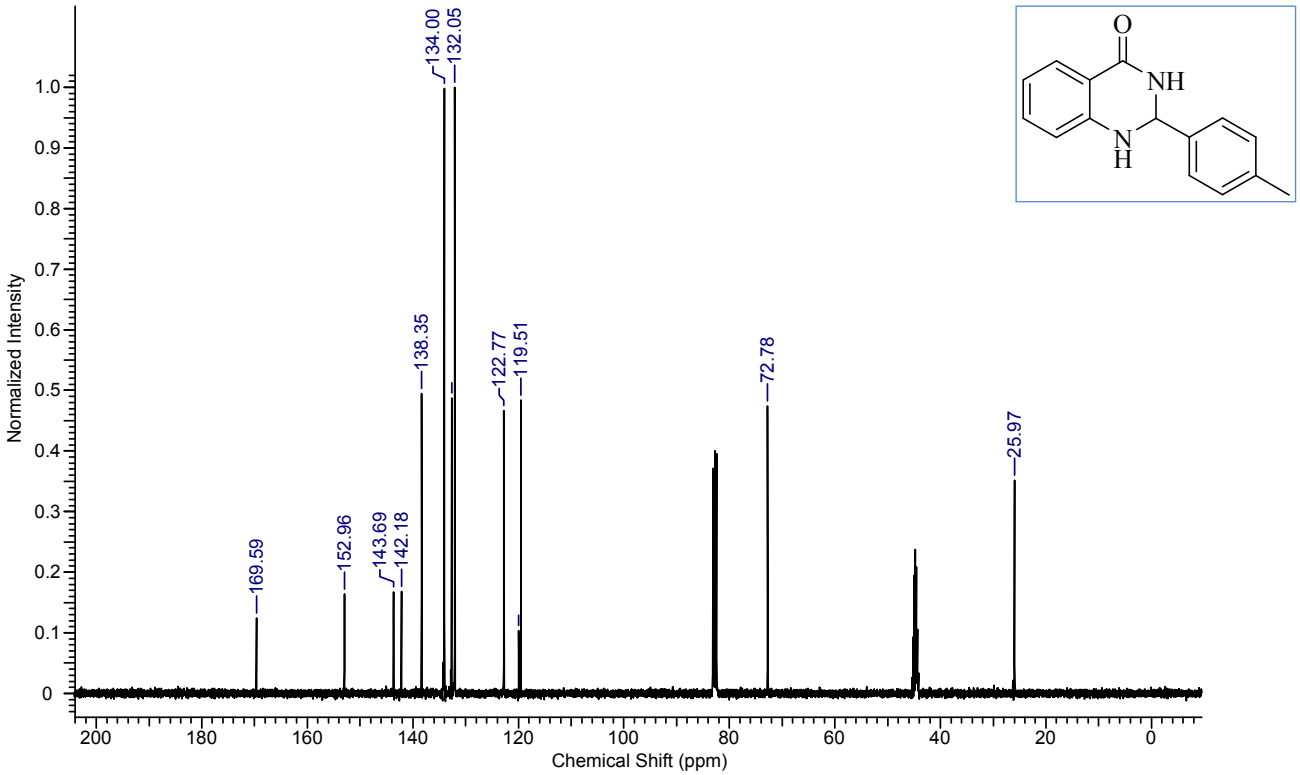
5. **6e**


Normalized Intensity
Chemical Shift (ppm)
3.85
7.96
7.94
7.54
7.52
7.34
7.32
6.97
6.95
6.93
6.91
6.68
6.66
5.86
5.75
4.36
1.04 2.09 1.18 2.12 0.89 1.08 1.03 0.97
1.11
3.27
O
NH
N
H
O

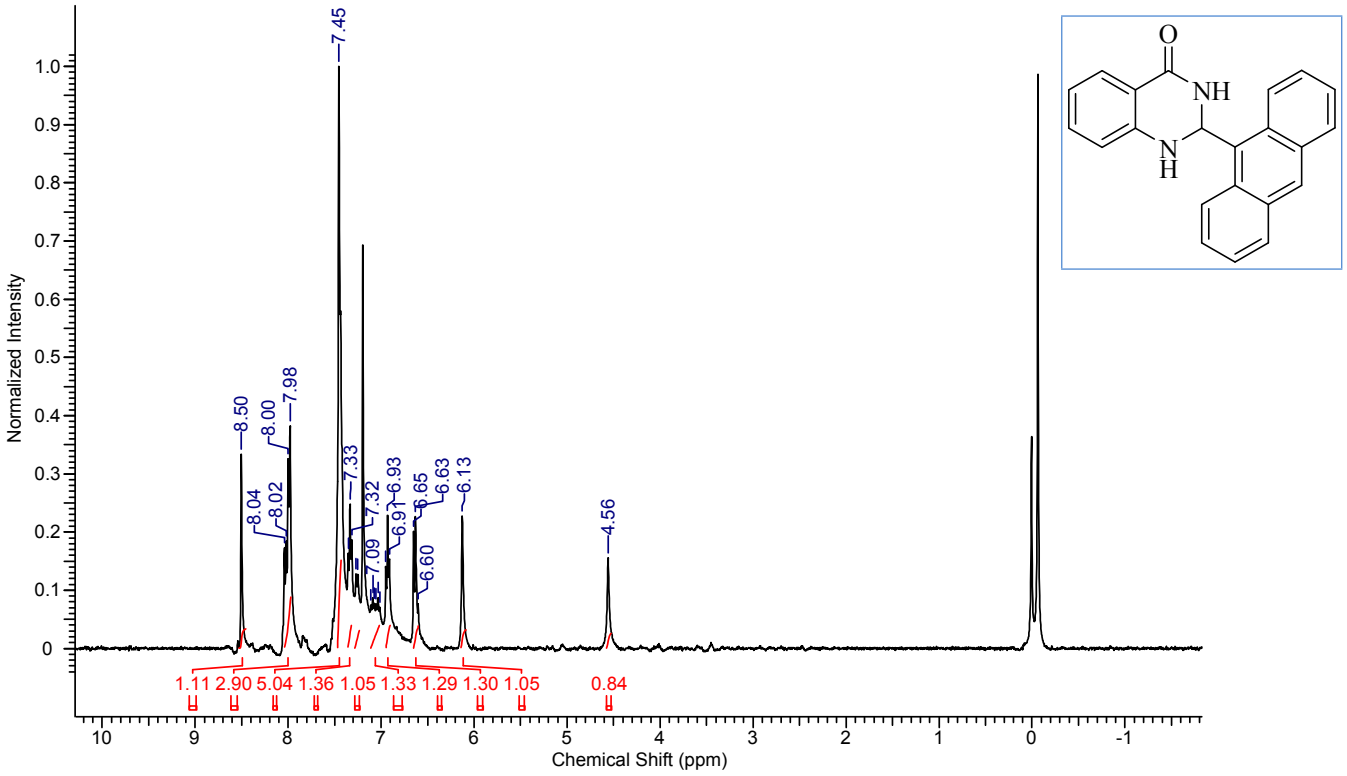

Normalized Intensity
Chemical Shift (ppm)
164.88
160.93
147.37
134.01
130.53
128.80
128.73
119.64
114.55
114.38
68.68
55.42
O
NH
N
H
O

6. **6f**


O
NH
N
H
2.20
7.64
7.30
7.29
7.09
7.05
7.04
6.97
6.60
6.58
5.64
2.93
1.01
2.17
4.08
2.33
1.06
2.37
3.12
Normalized Intensity
Chemical Shift (ppm)


O
NH
N
H
134.00
132.05
138.35
122.77
119.51
72.78
25.97
169.59
152.96
143.69
142.18
Normalized Intensity
Chemical Shift (ppm)

7. **6g**


Normalized Intensity
Chemical Shift (ppm)
7.45
8.50
8.00
7.98
8.04
8.02
7.33
7.32
7.09
6.93
6.91
6.65
6.63
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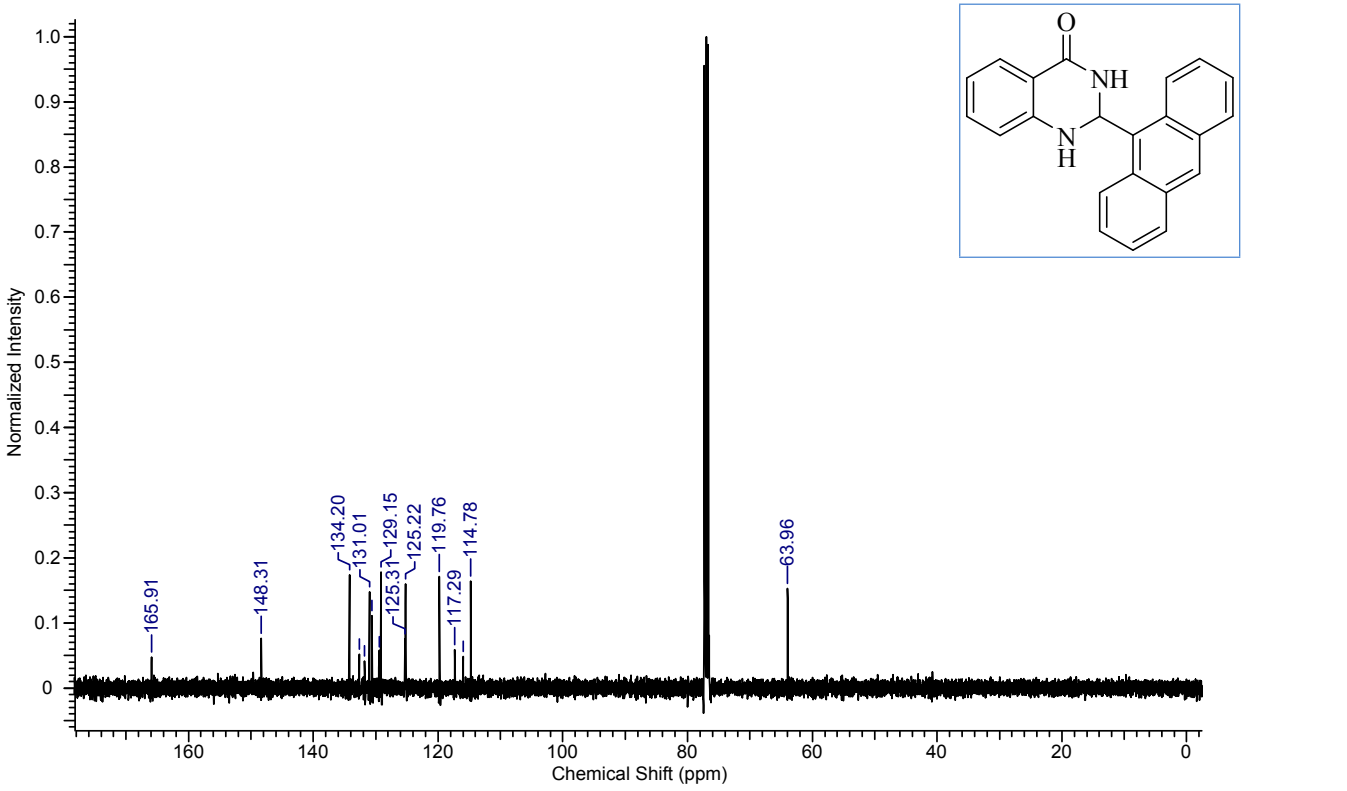
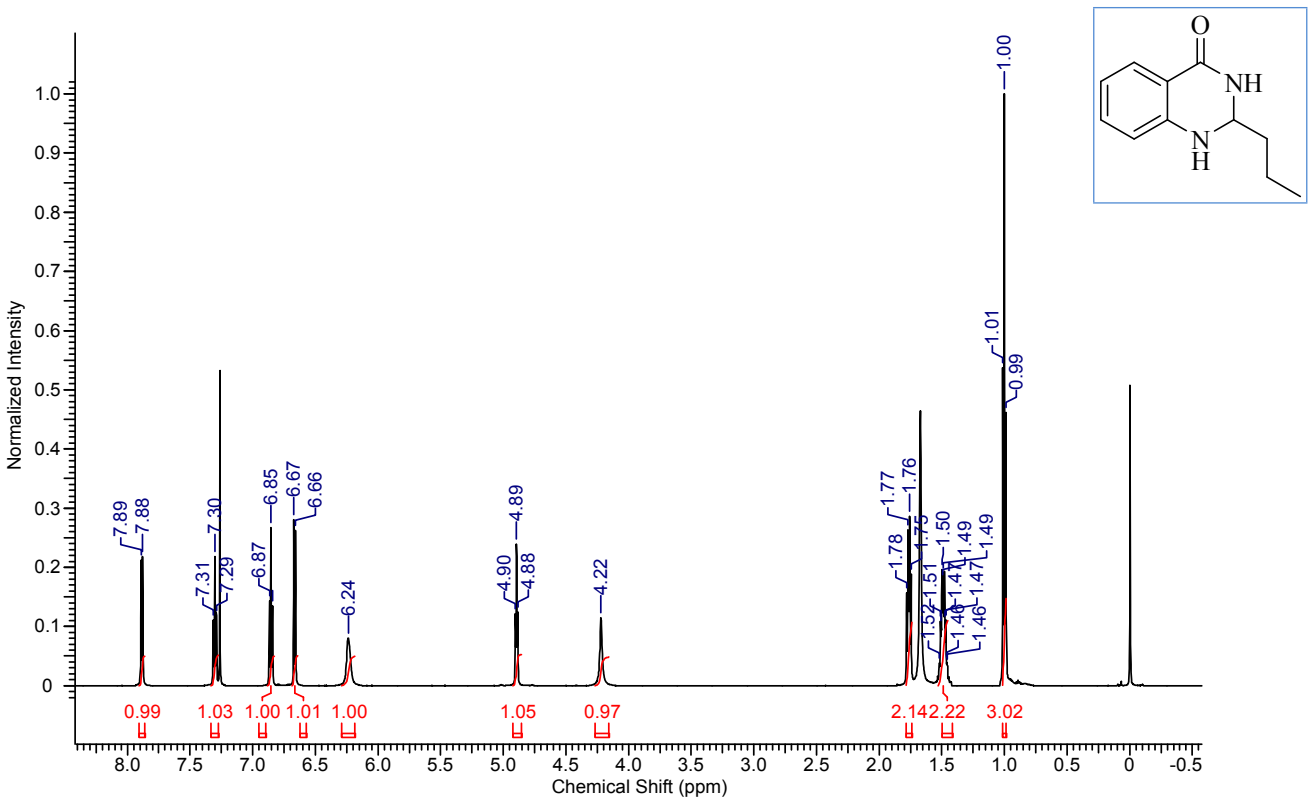
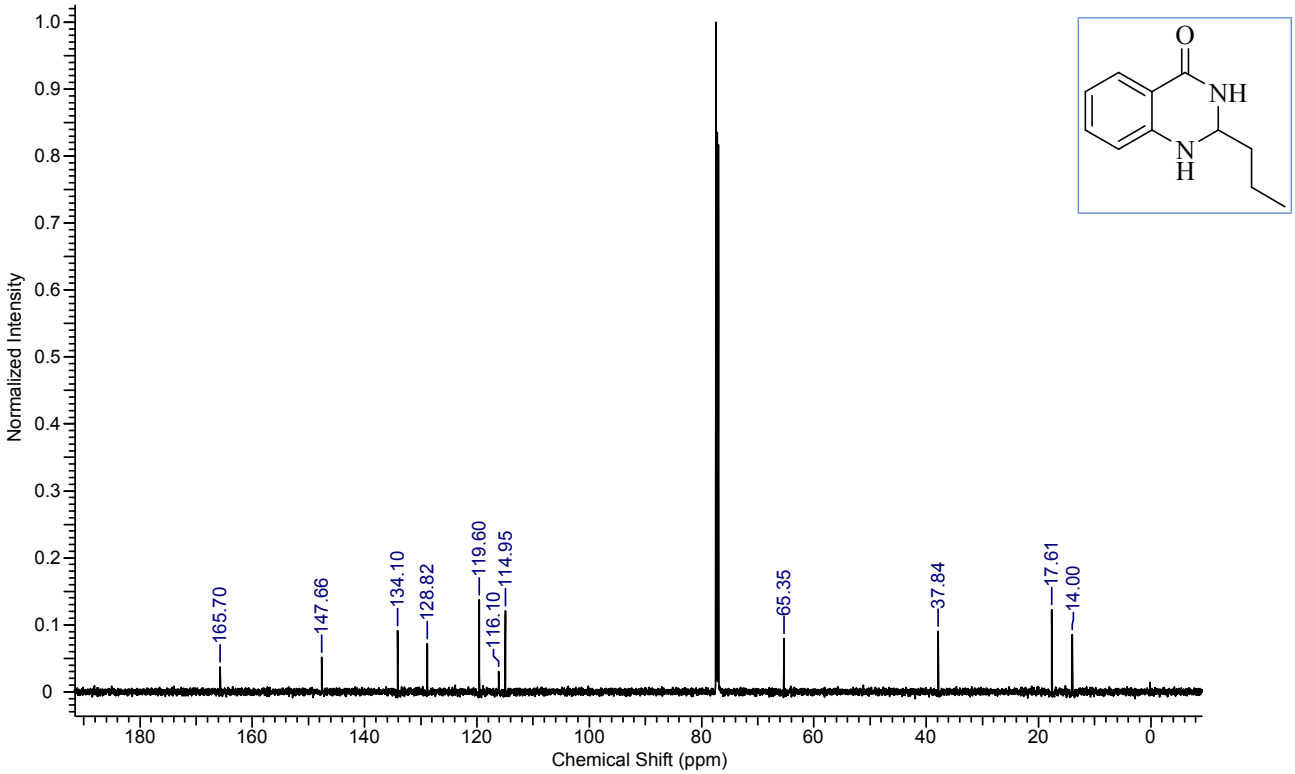
0.84
O
NH
N
H

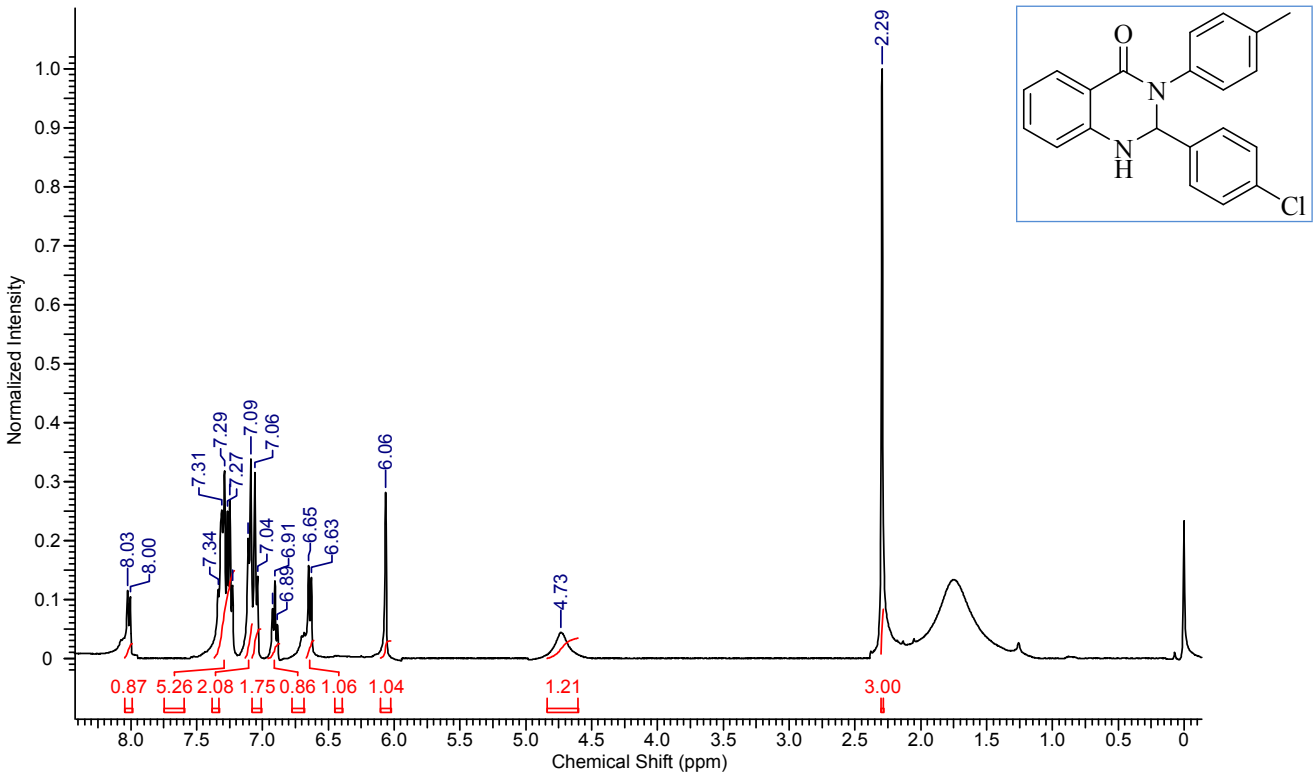

Normalized Intensity
Chemical Shift (ppm)
165.91
148.31
134.20
131.01
129.15
125.31
125.22
119.76
117.29
114.78
63.96
O
NH
N
H

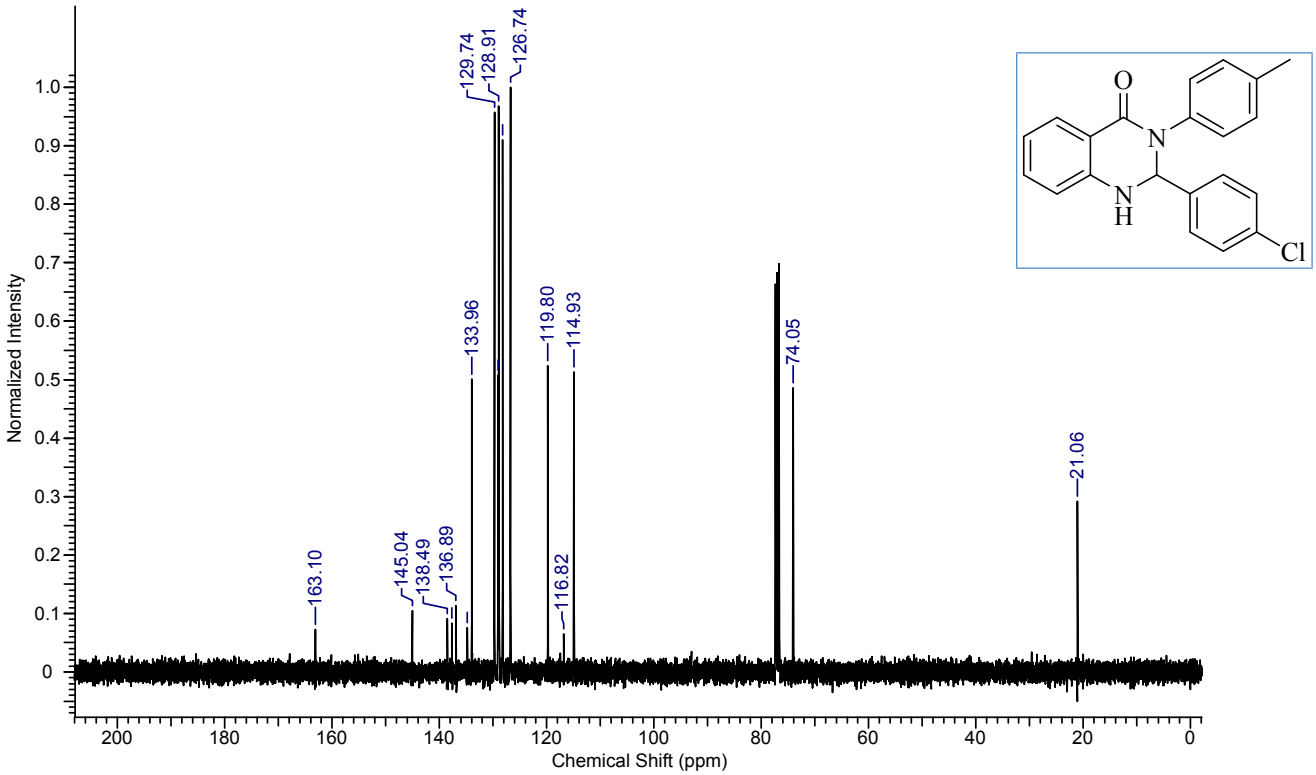
8. **6h**


O
NH
N
H
Normalized Intensity
Chemical Shift (ppm)
7.89
7.88
7.31
7.30
7.29
6.87
6.85
6.67
6.66
6.24
4.90
4.89
4.88
4.22
1.78
1.77
1.76
1.75
1.51
1.50
1.49
1.48
1.47
1.46
1.01
1.00
0.99
0.99
1.03
1.00
1.01
1.00
1.05
0.97
2.14
2.22
3.02

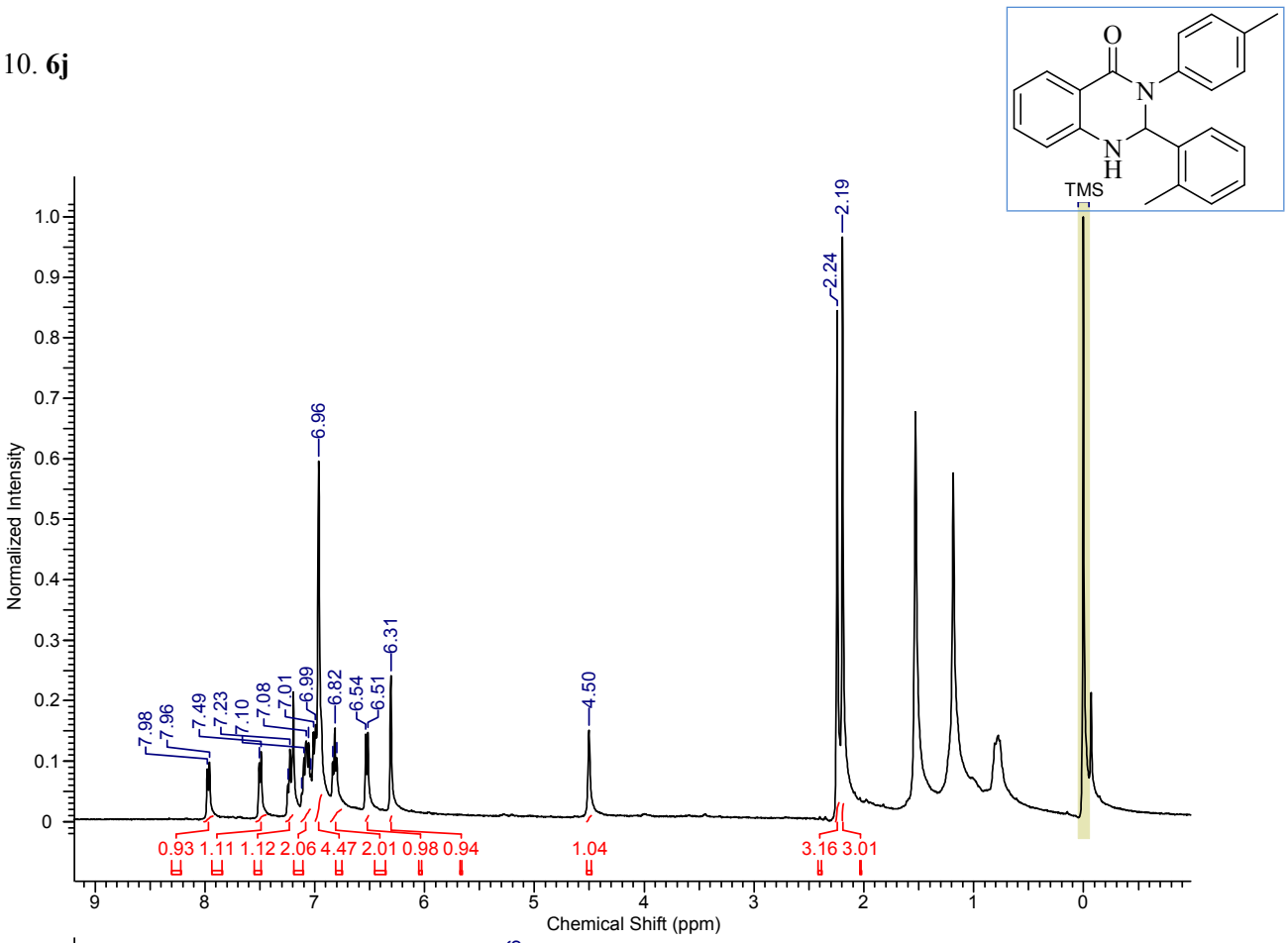

O
NH
N
H
Normalized Intensity
Chemical Shift (ppm)
165.70
147.66
134.10
128.82
119.60
116.10
114.95
65.35
37.84
17.61
14.00

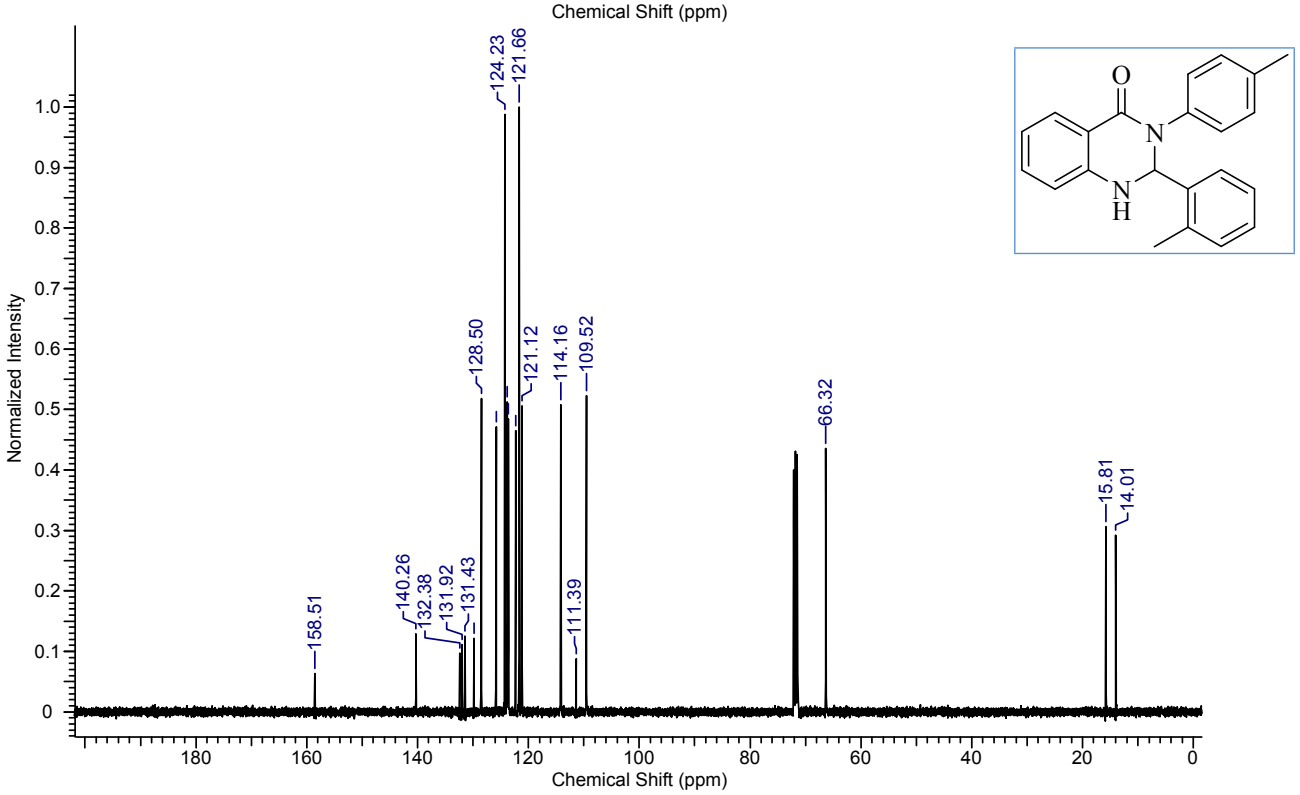
9. **6i**


Normalized Intensity
Chemical Shift (ppm)
8.03
8.00
7.34
7.31
7.29
7.27
7.09
7.06
7.04
6.91
6.89
6.65
6.63
6.06
4.73
2.29
0.87
5.26
2.08
1.75
0.86
1.06
1.04
1.21
3.00
O
N
N
H
Cl

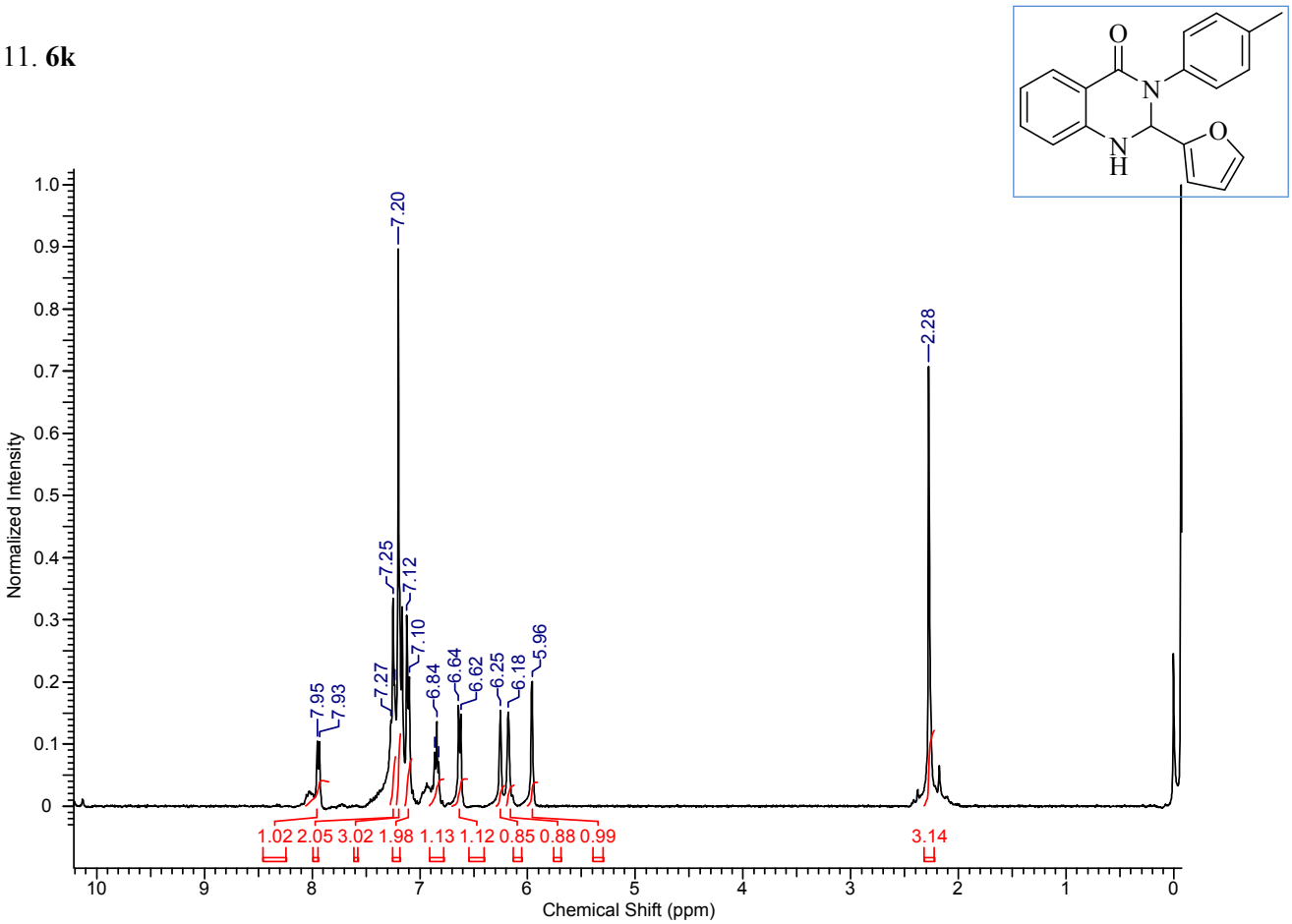

Normalized Intensity
Chemical Shift (ppm)
163.10
145.04
138.49
136.89
133.96
129.74
128.91
126.74
119.80
116.82
114.93
74.05
21.06
O
N
N
H
Cl

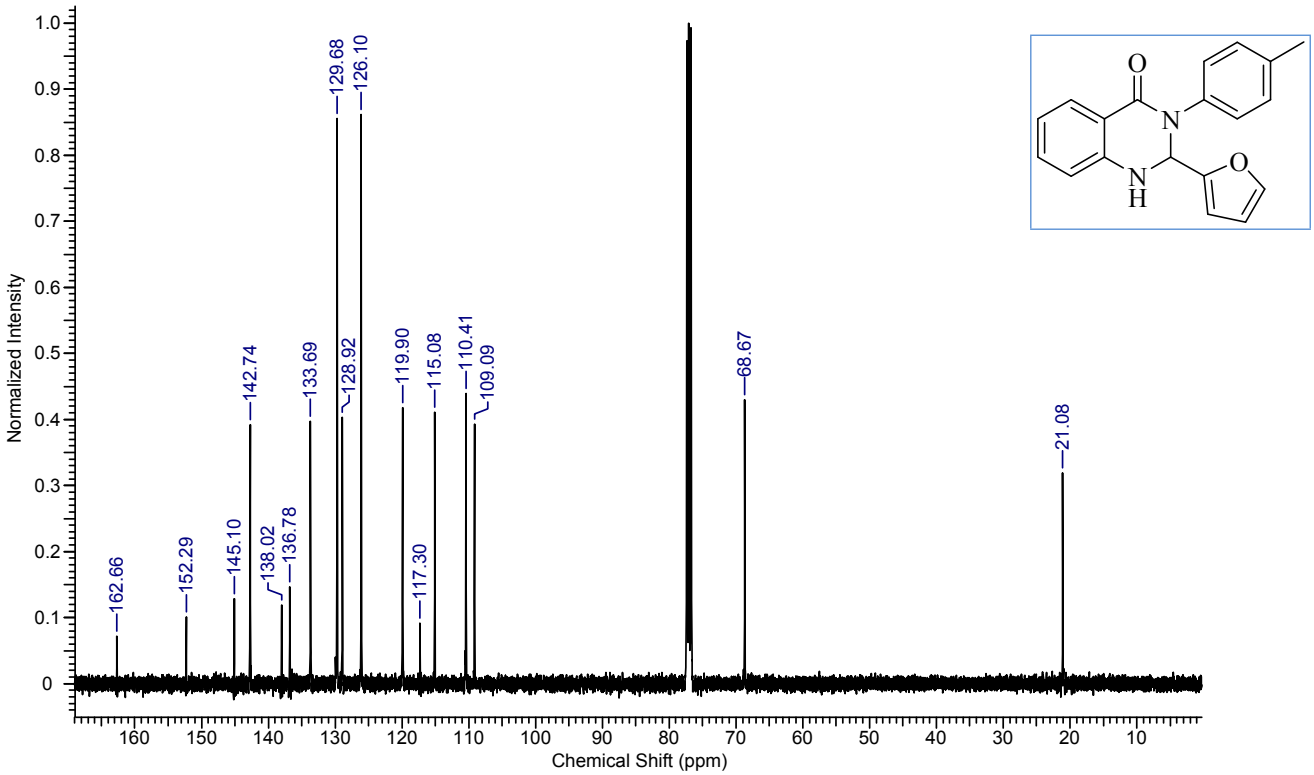
10. **6j**


O
N
N
H
TMS
Normalized Intensity
Chemical Shift (ppm)
7.98
7.96
7.49
7.23
7.10
7.08
7.01
6.99
6.96
6.82
6.54
6.51
6.31
4.50
2.24
2.19
0.93 1.11 1.12 2.06 4.47 2.01 0.98 0.94
1.04
3.16 3.01

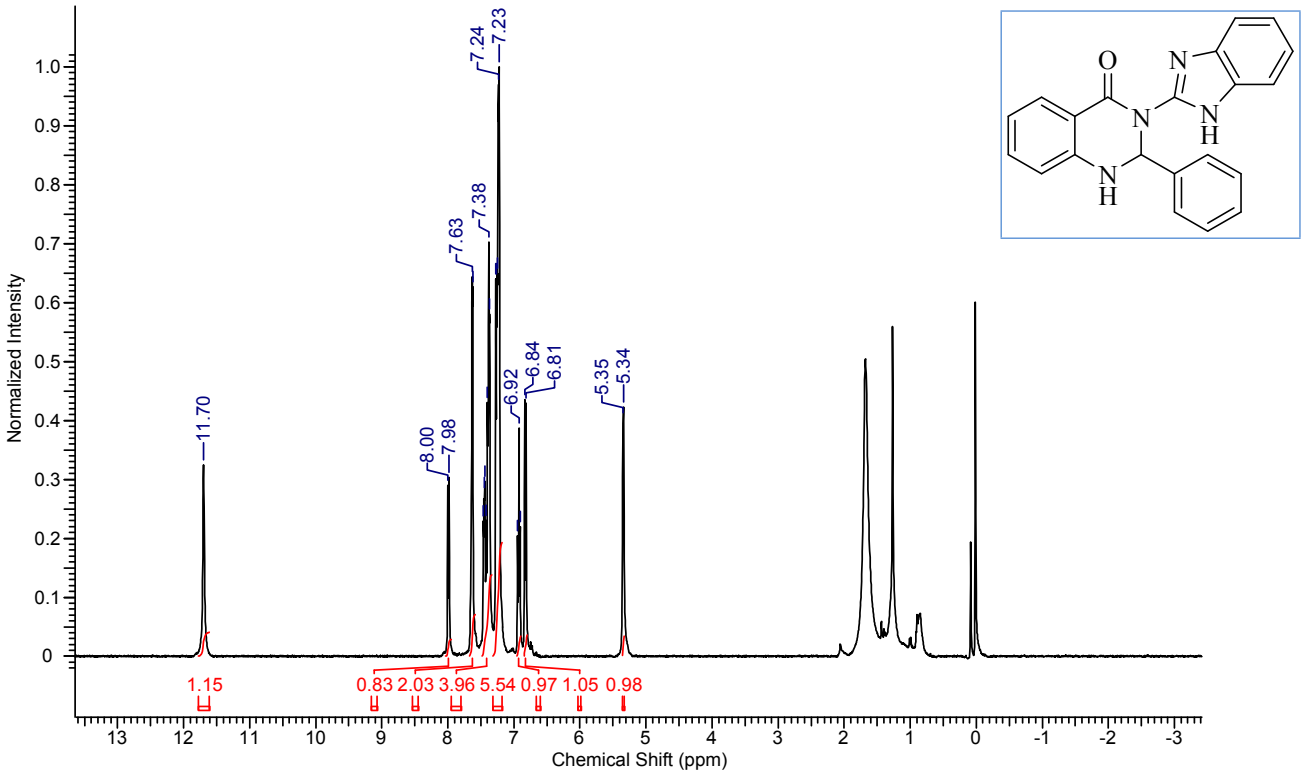

O
N
N
H
Normalized Intensity
Chemical Shift (ppm)
158.51
140.26
132.38
131.92
131.43
128.50
124.23
121.66
121.12
114.16
111.39
109.52
66.32
15.81
14.01

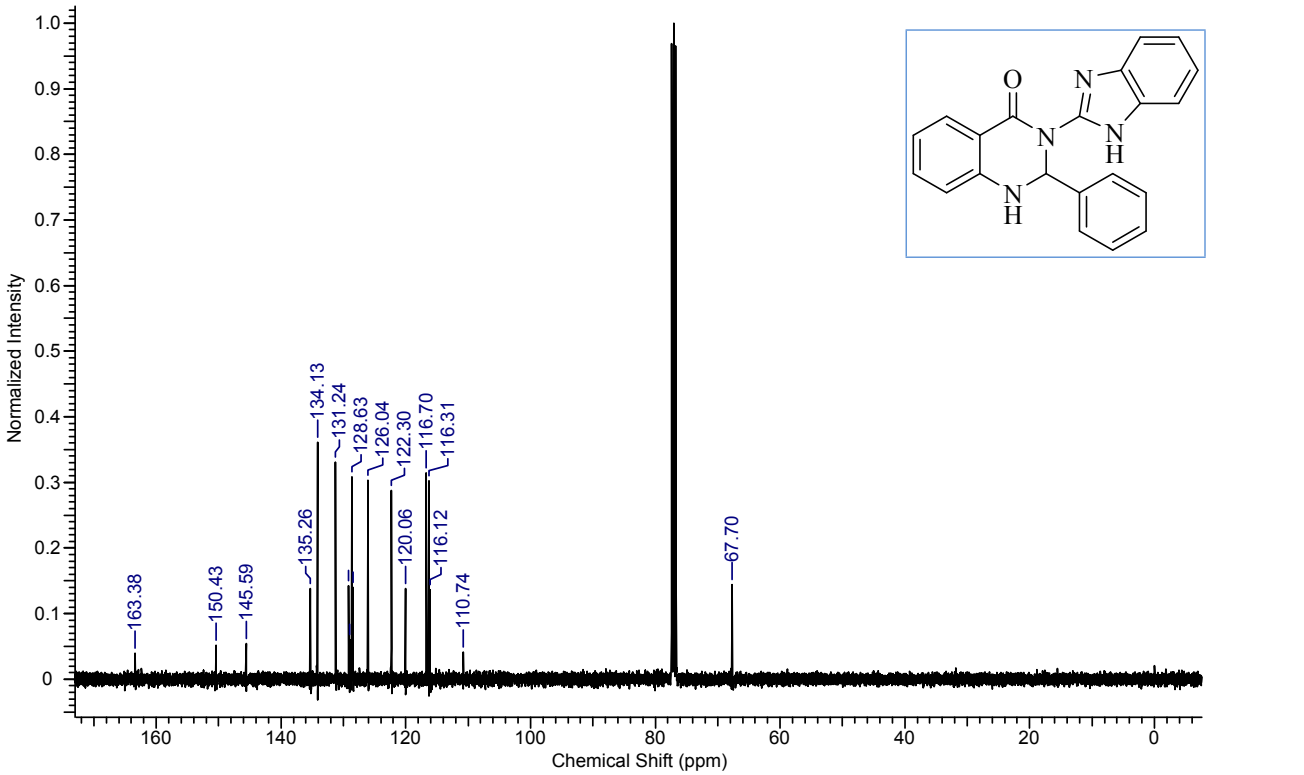
11. **6k**


Normalized Intensity
Chemical Shift (ppm)
7.95
7.93
7.27
7.25
7.20
7.12
7.10
6.84
6.64
6.62
6.25
6.18
5.96
2.28
1.02 2.05 3.02 1.98 1.13 1.12 0.85 0.88 0.99
3.14

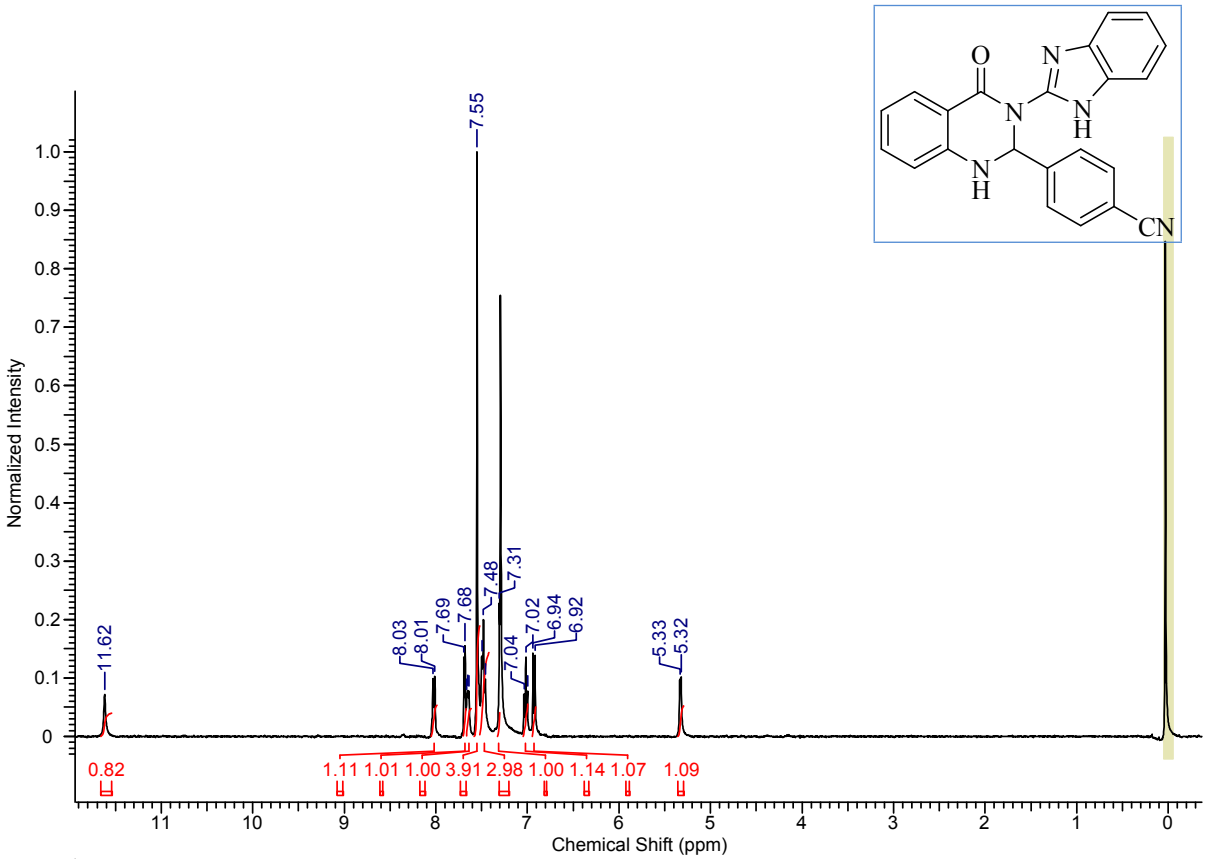

Normalized Intensity
Chemical Shift (ppm)
162.66
152.29
145.10
142.74
138.02
136.78
133.69
129.68
128.92
126.10
119.90
117.30
115.08
110.41
109.09
68.67
21.08

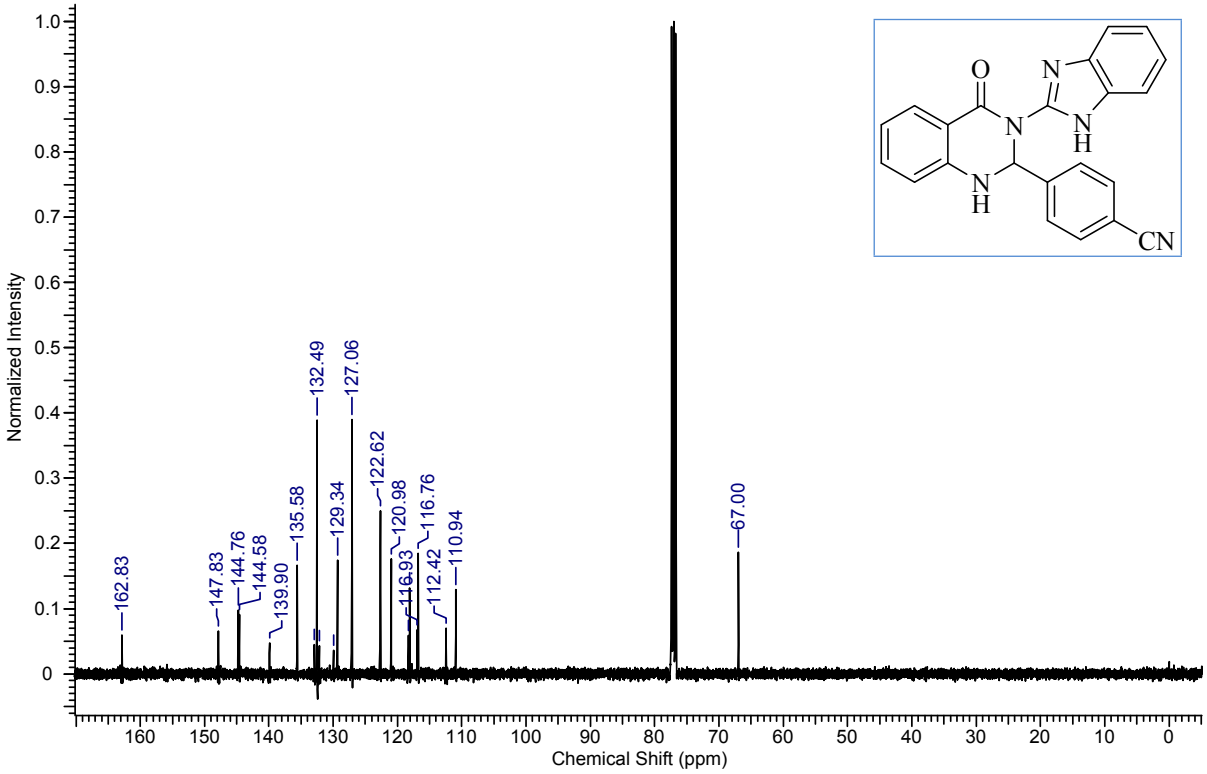
12. **6l**


Normalized Intensity
Chemical Shift (ppm)
11.70
8.00
7.98
7.63
7.38
7.24
7.23
6.92
6.84
6.81
5.35
5.34
1.15
0.83 2.03 3.96 5.54 0.97 1.05 0.98
O
N
N
H
N
H

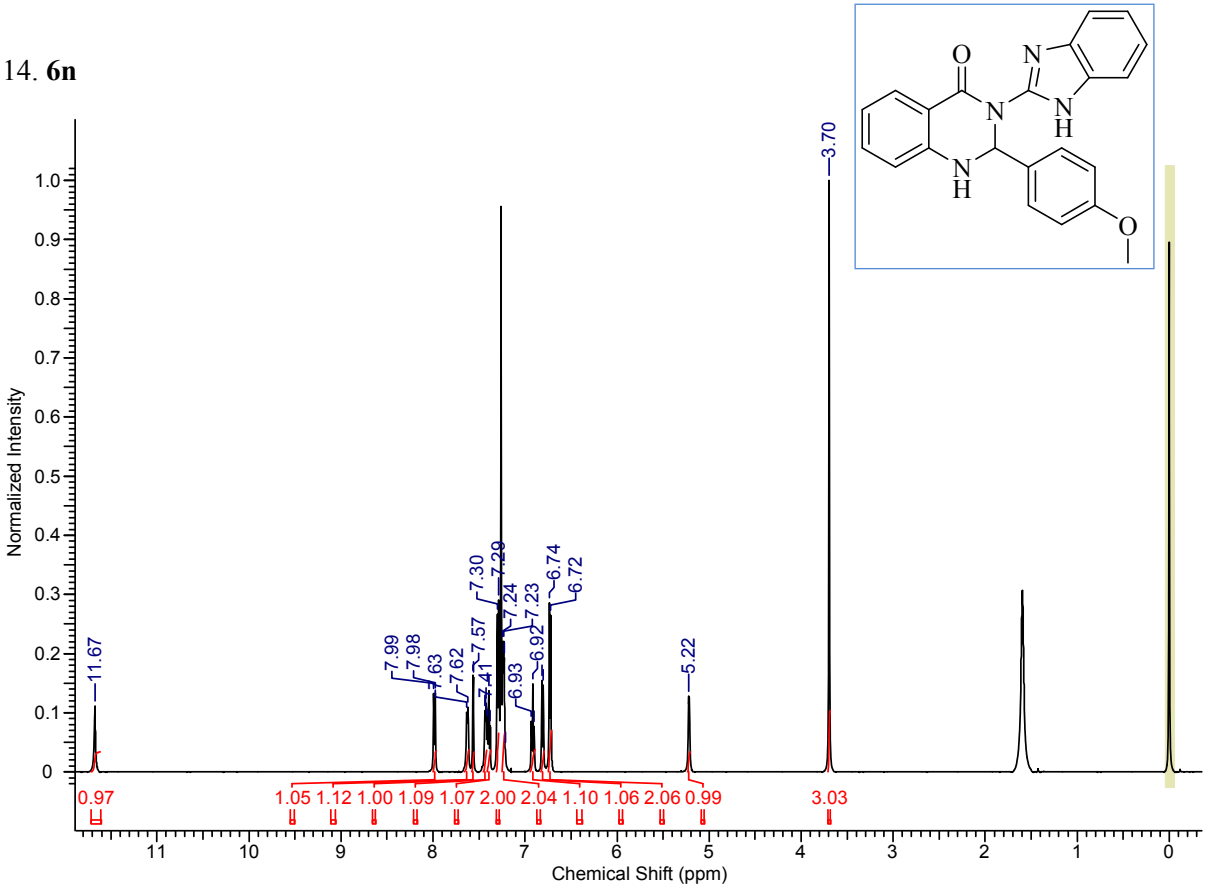

Normalized Intensity
Chemical Shift (ppm)
163.38
150.43
145.59
135.26
134.13
131.24
128.63
126.04
122.30
120.06
116.70
116.31
116.12
110.74
67.70
O
N
N
H
N
H

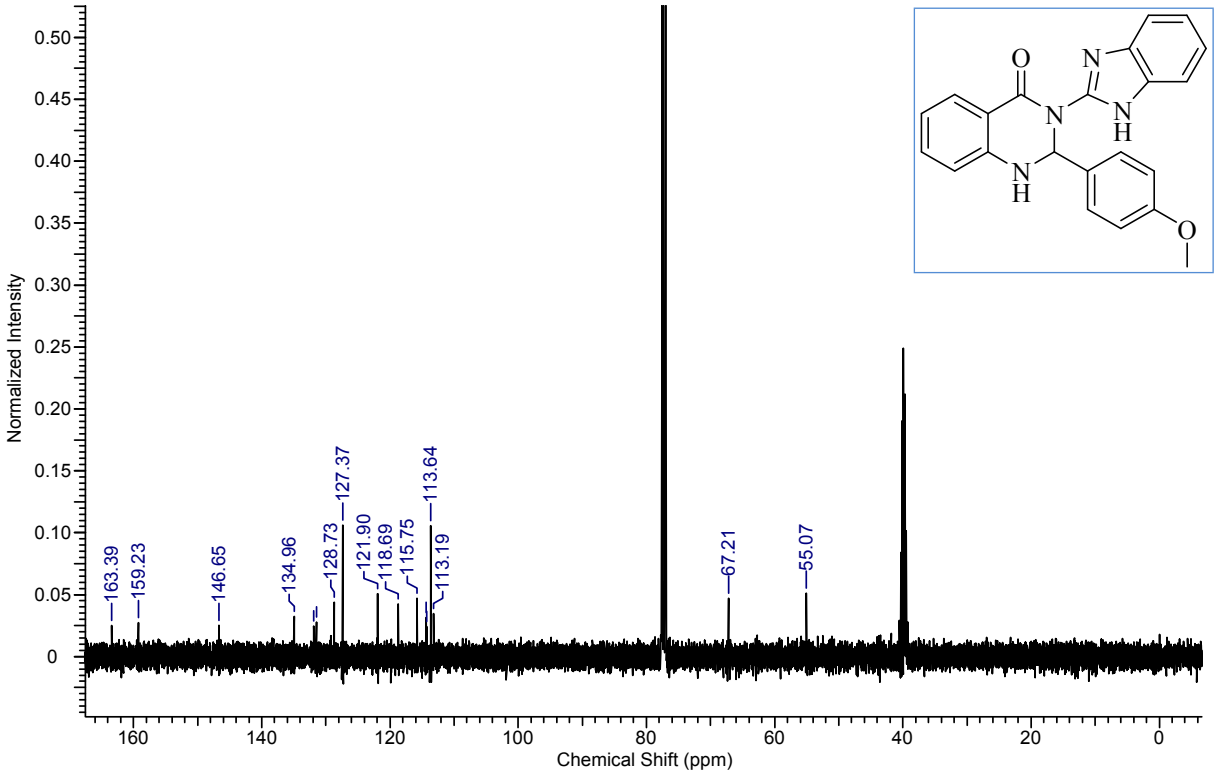
13. **6m**


O
N
N
H
N
H
CN
Normalized Intensity
11.62
8.03
8.01
7.69
7.68
7.55
7.48
7.31
7.04
7.02
6.94
6.92
5.33
5.32
0.82
1.11 1.01 1.00 3.91 2.98 1.00 1.14 1.07
1.09
Chemical Shift (ppm)

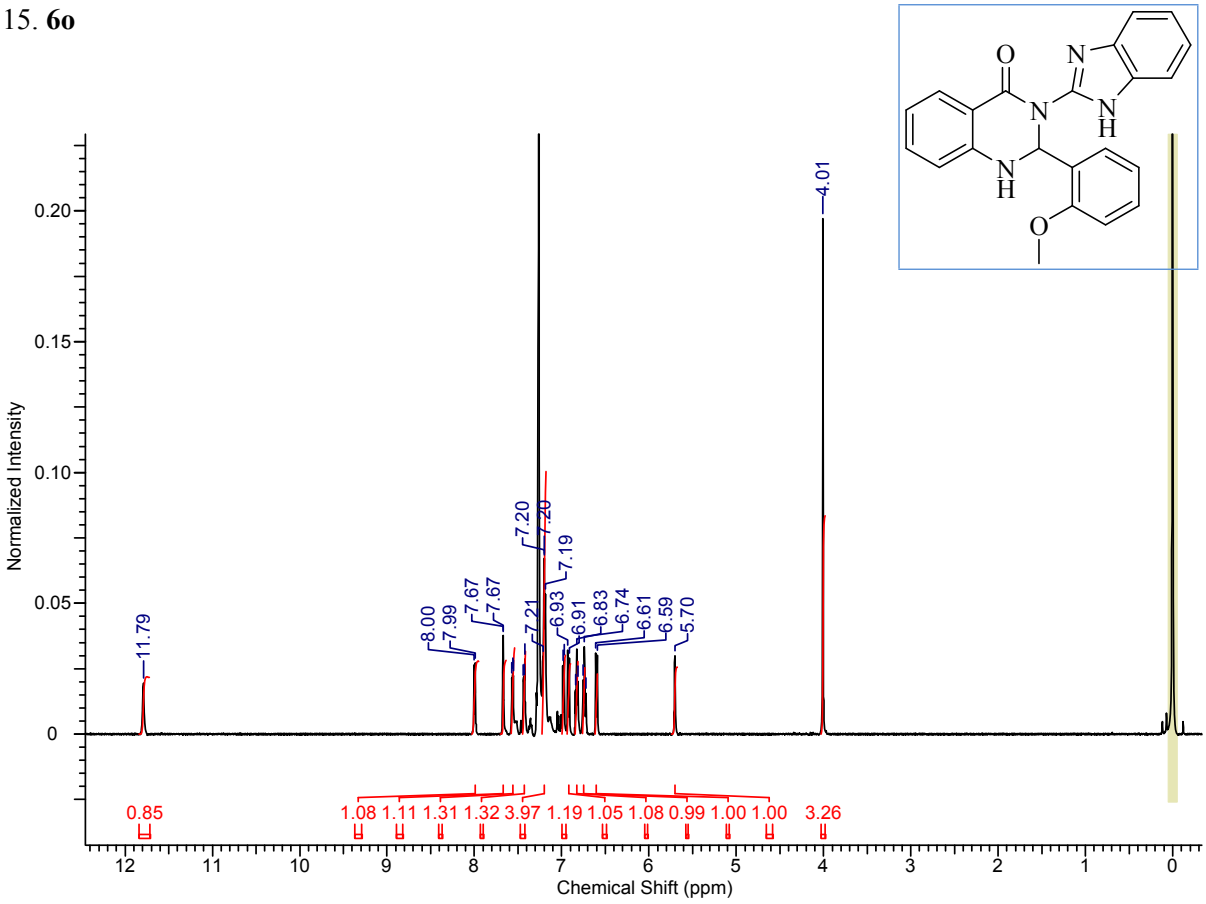

O
N
N
H
N
H
CN
Normalized Intensity
162.83
147.83
144.76
144.58
139.90
135.58
132.49
129.34
127.06
122.62
120.98
116.93
116.76
112.42
110.94
67.00
Chemical Shift (ppm)

14. **6n**

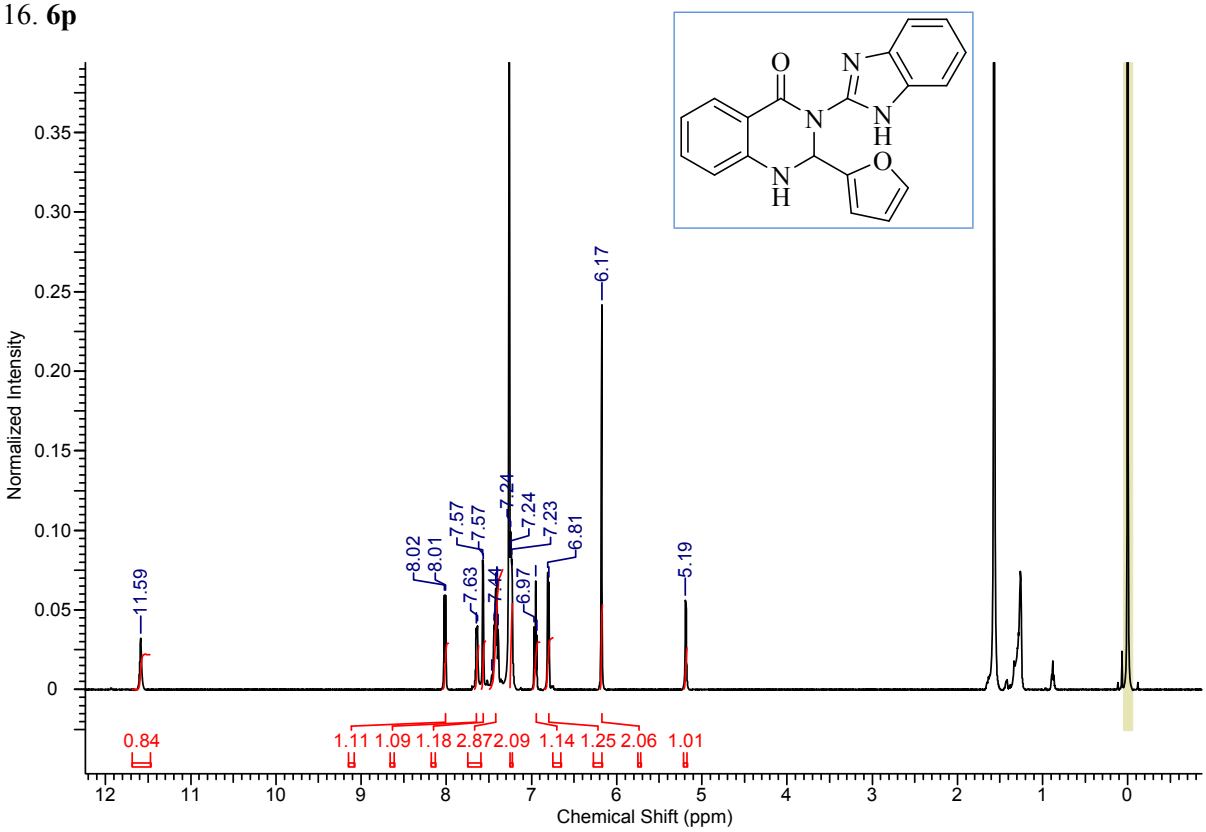

Normalized Intensity
Chemical Shift (ppm)
11.67
7.99
7.98
7.63
7.62
7.57
7.41
7.30
7.29
7.24
7.23
6.93
6.92
6.74
6.72
5.22
3.70
0.97
1.05 1.12 1.00 1.09 1.07 2.00 2.04 1.10 1.06 2.06 0.99
3.03

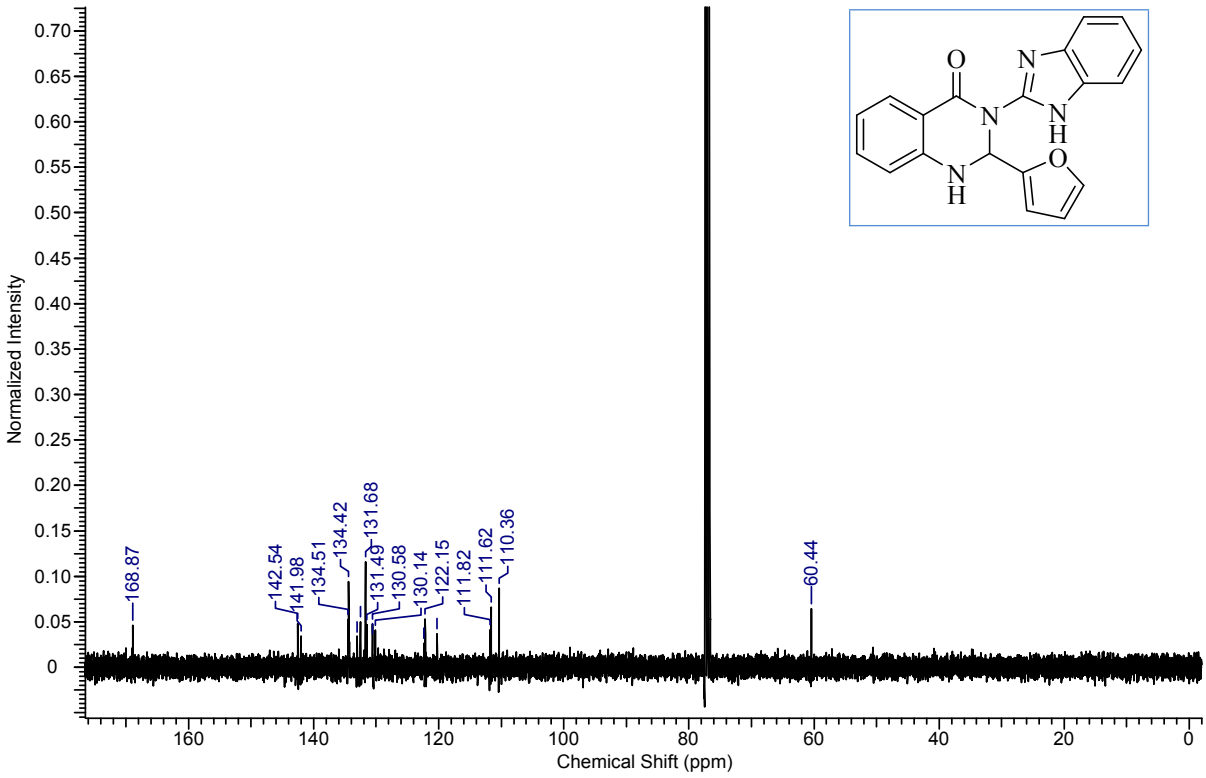

Normalized Intensity
Chemical Shift (ppm)
163.39
159.23
146.65
134.96
128.73
127.37
121.90
118.69
115.75
113.64
113.19
67.21
55.07

15. **6o**

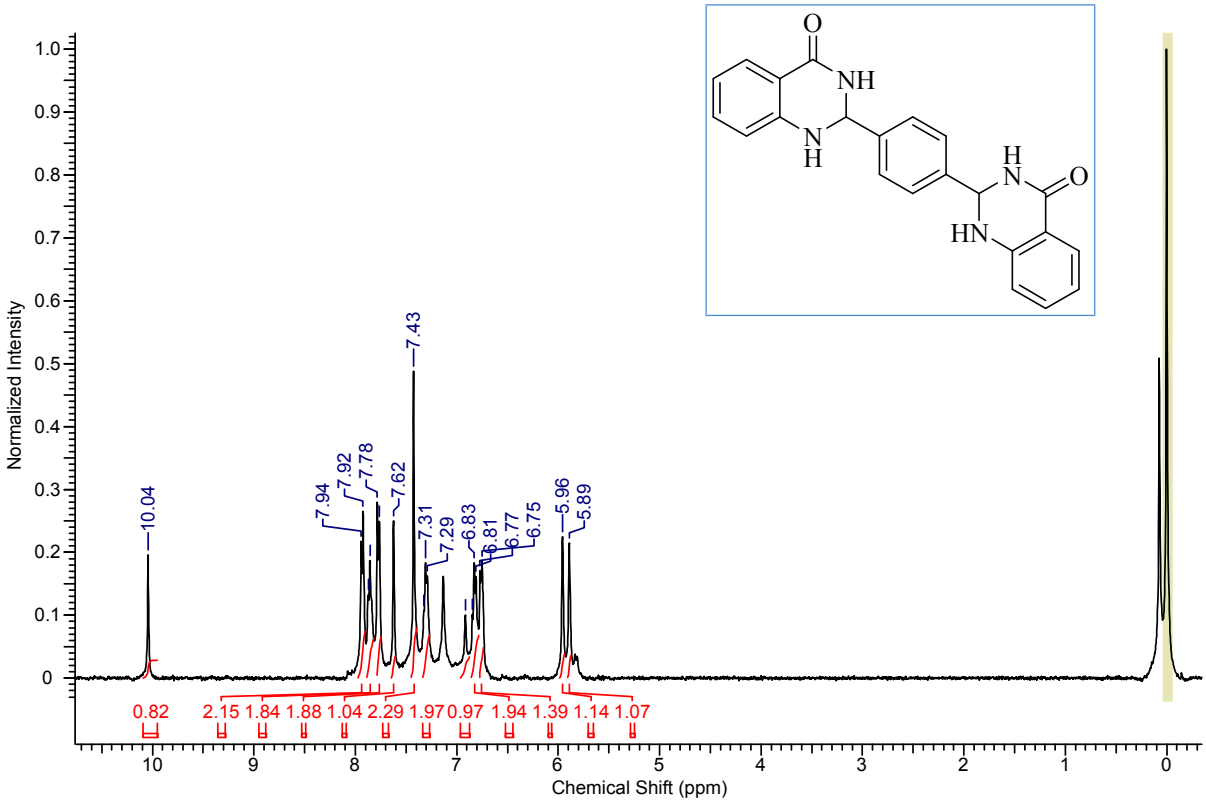

Normalized Intensity
Chemical Shift (ppm)
11.79
8.00
7.99
7.67
7.67
7.21
7.20
7.20
7.19
6.93
6.91
6.83
6.74
6.61
6.59
5.70
4.01
0.85
1.08 1.11 1.31 1.32 3.97 1.19 1.05 1.08 0.99 1.00 1.00 3.26

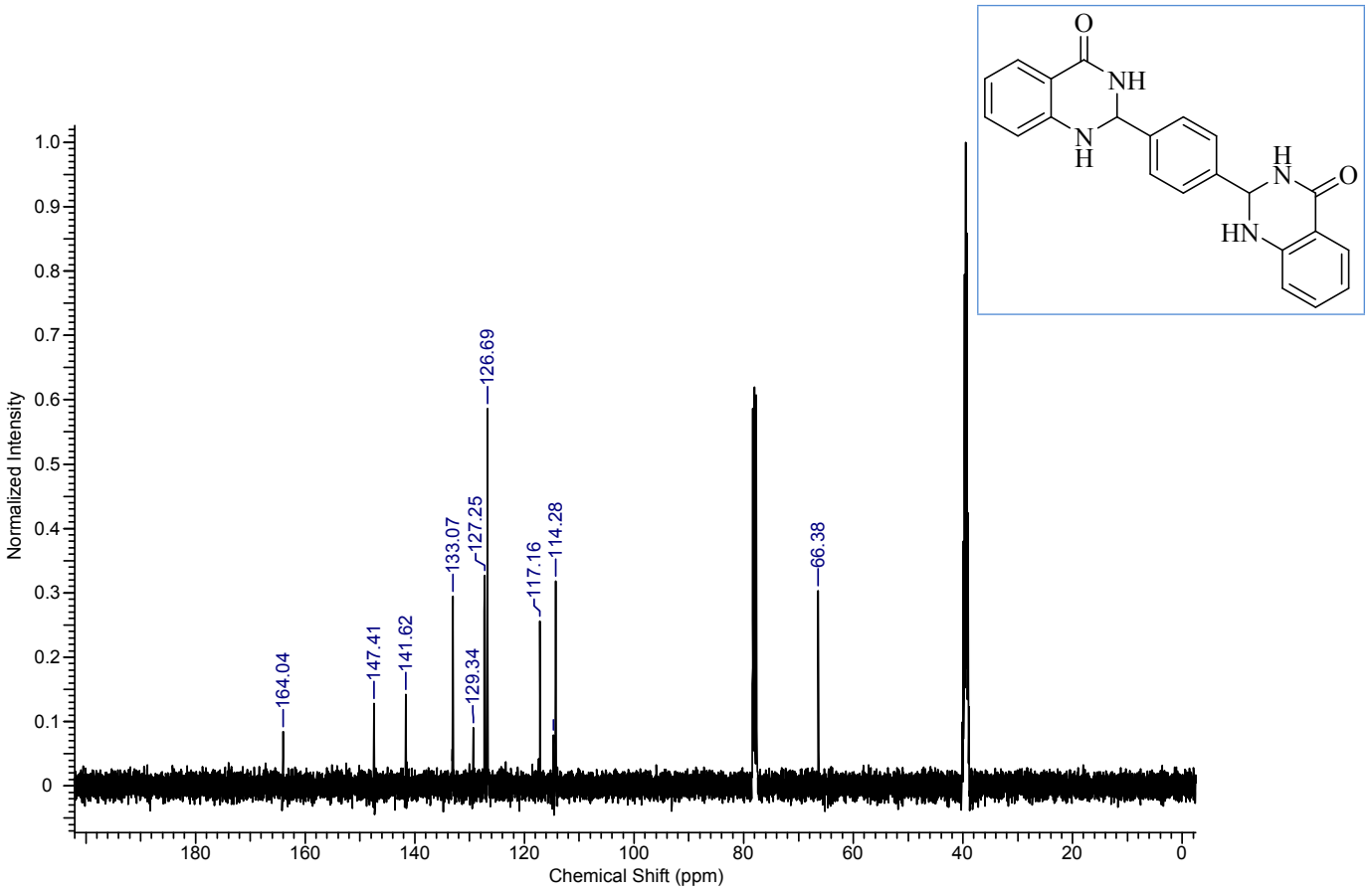
16. **6p**


Normalized Intensity
Chemical Shift (ppm)
11.59
8.02
8.01
7.57
7.57
7.24
7.24
7.23
7.63
7.44
6.97
6.81
6.17
5.19
0.84
1.11 1.09 1.18 2.87 2.09 1.14 1.25 2.06 1.01

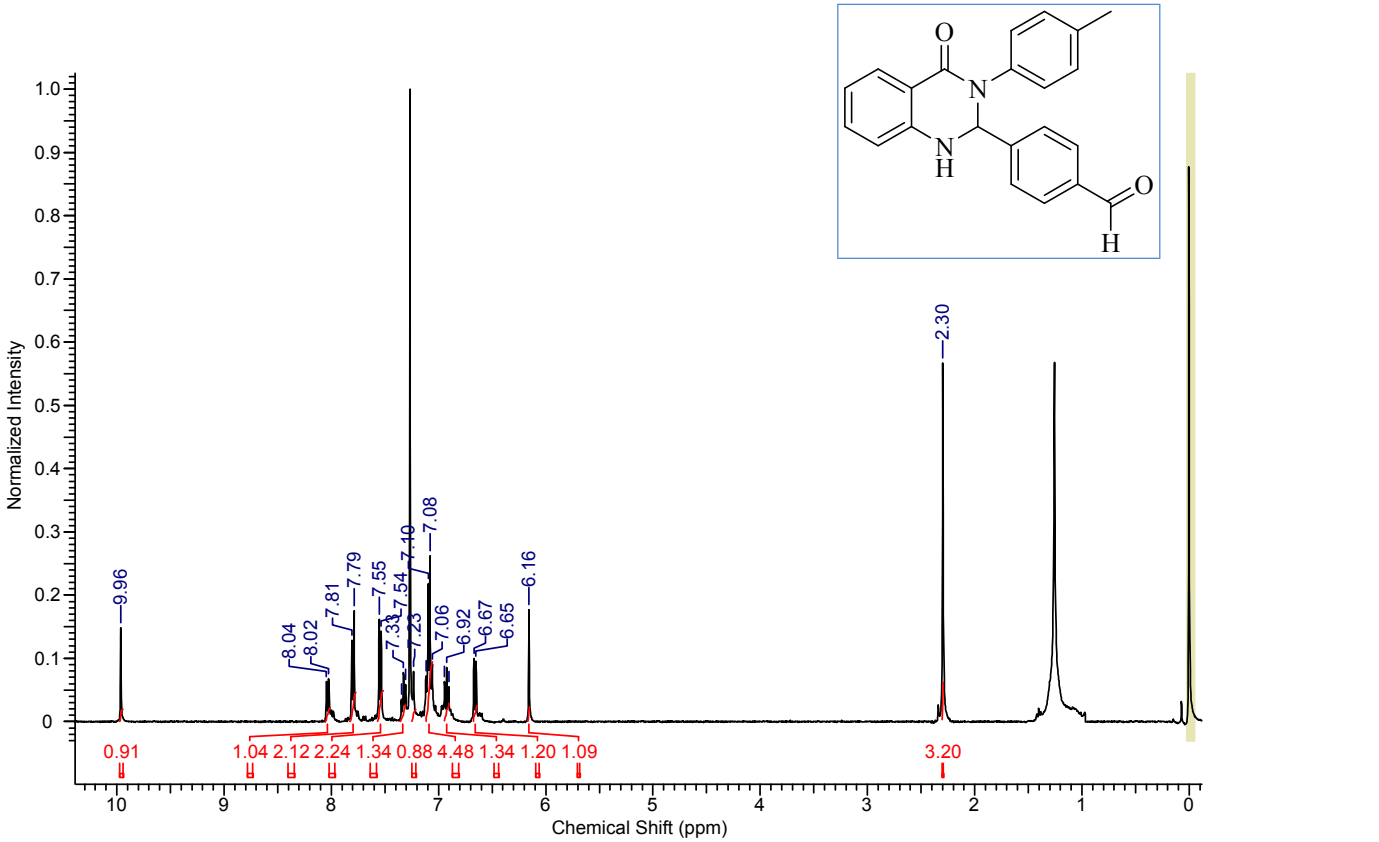

Normalized Intensity
Chemical Shift (ppm)
168.87
142.54
141.98
134.51
134.42
131.68
131.49
130.58
130.14
122.15
111.82
111.62
110.36
60.44
O
N
N
H
N
H
O

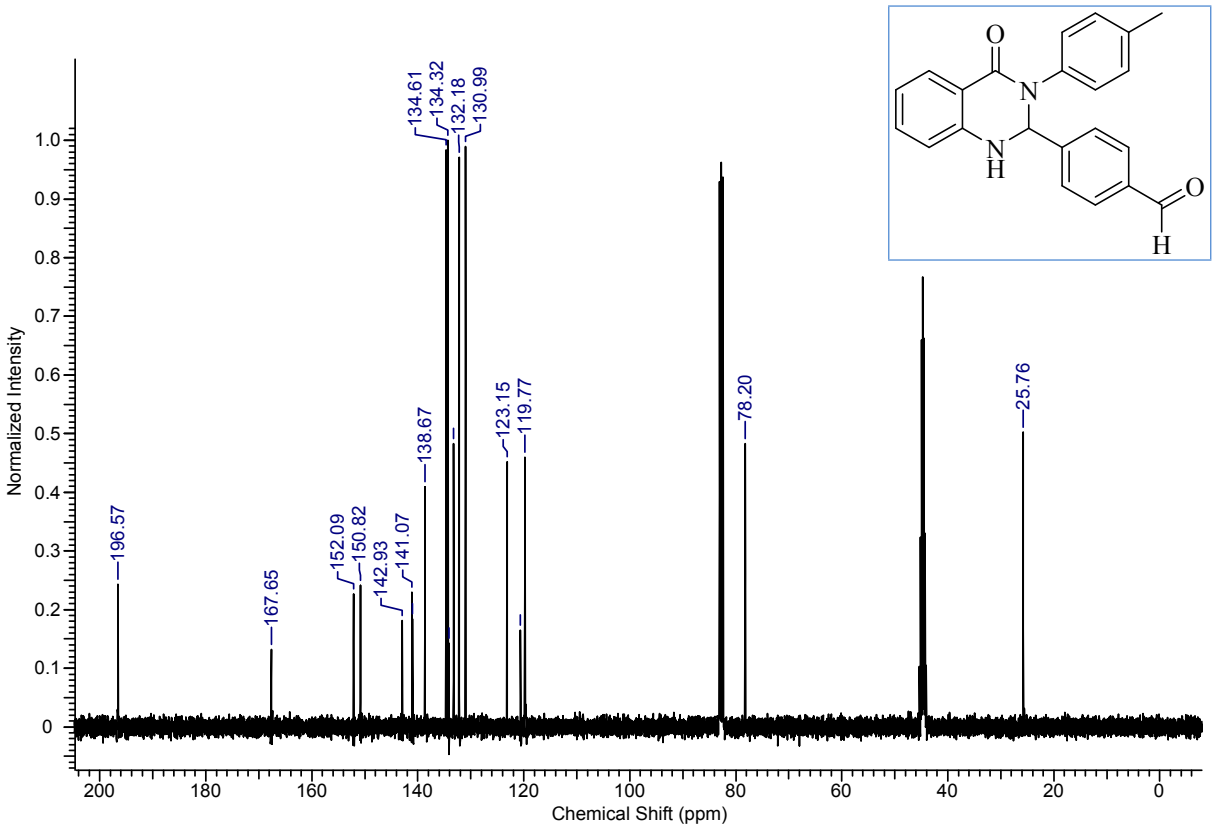
17. **6q**


Normalized Intensity
Chemical Shift (ppm)
10.04
7.94
7.92
7.78
7.62
7.43
7.31
7.29
6.83
6.81
6.77
6.75
5.96
5.89
0.82
2.15 1.84 1.88 1.04 2.29 1.97 0.97 1.94 1.39 1.14 1.07
O
NH
N
H
H
N
O
HN

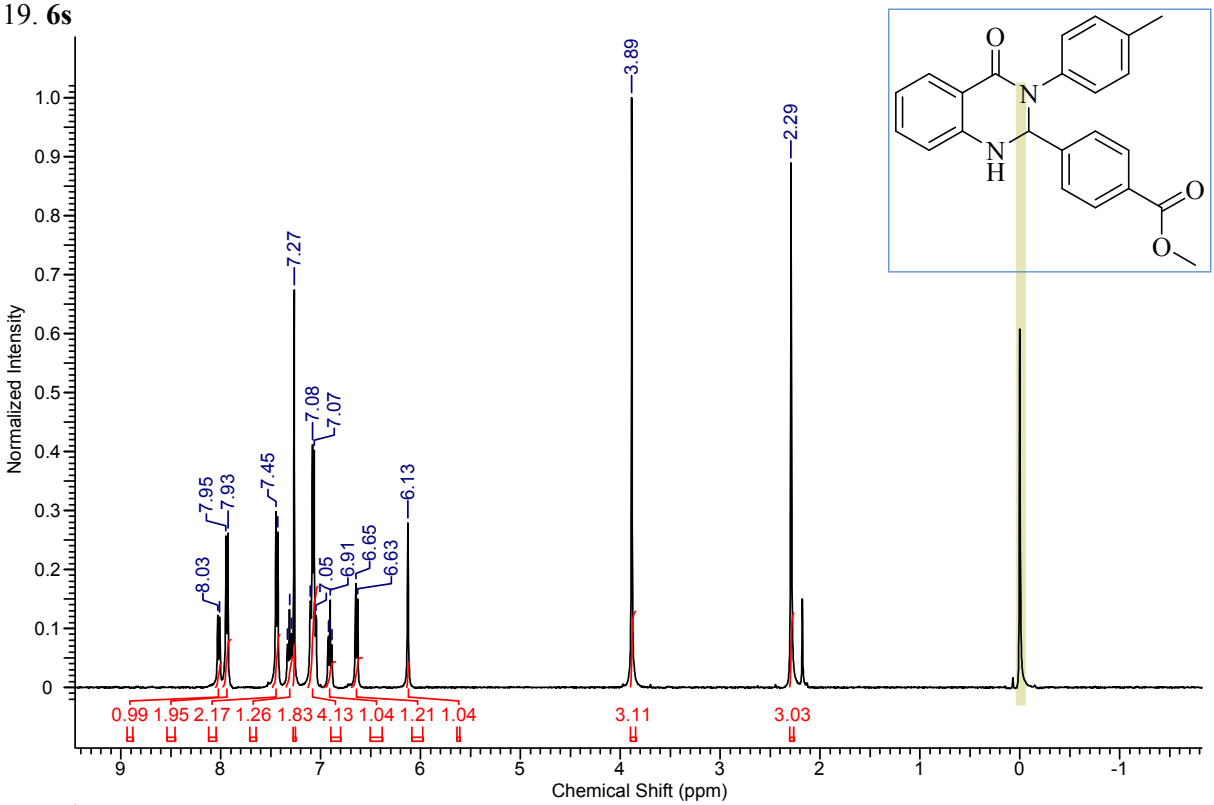

O
NH
N
H
H
N
O
HN
Normalized Intensity
Chemical Shift (ppm)
164.04
147.41
141.62
133.07
129.34
127.25
126.69
117.16
114.28
66.38

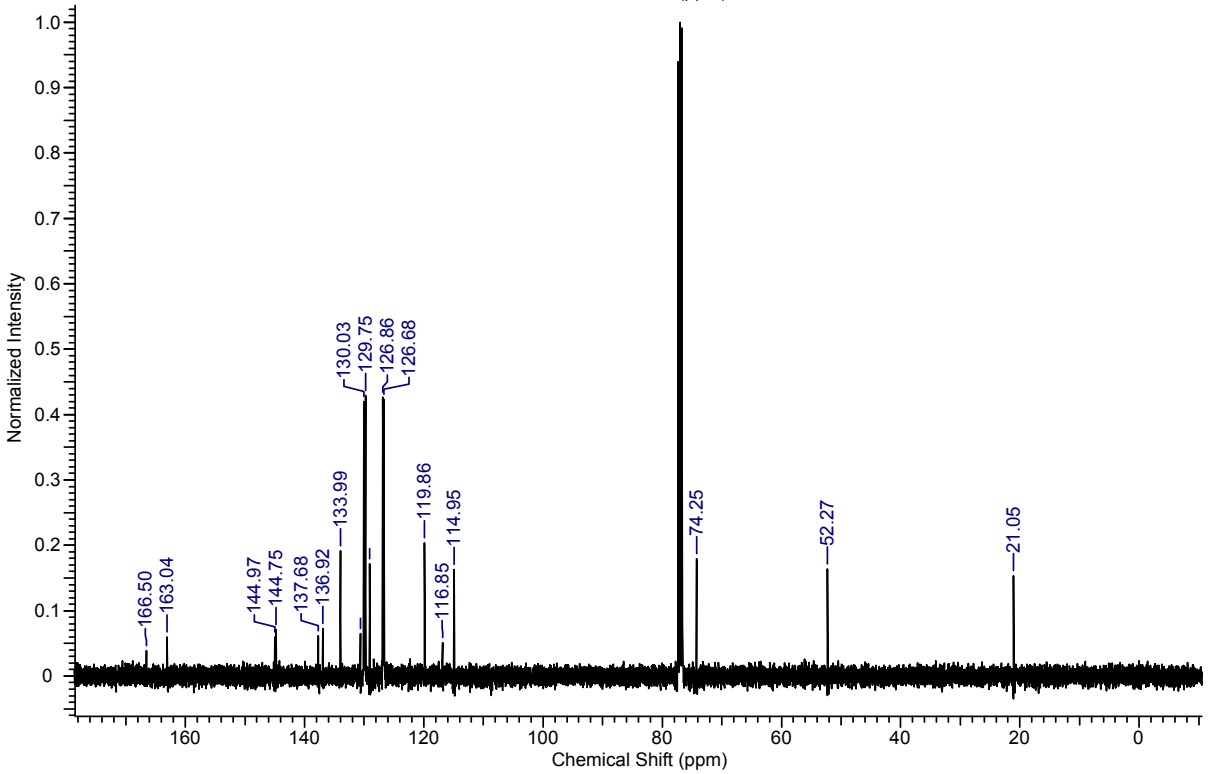
18. **6r**


O
N
N
H
O
H
Normalized Intensity
Chemical Shift (ppm)
9.96
8.04
8.02
7.81
7.79
7.55
7.54
7.40
7.33
7.25
7.08
7.06
6.92
6.67
6.65
6.16
2.30
0.91
1.04 2.12 2.24 1.34 0.88 4.48 1.34 1.20 1.09
3.20

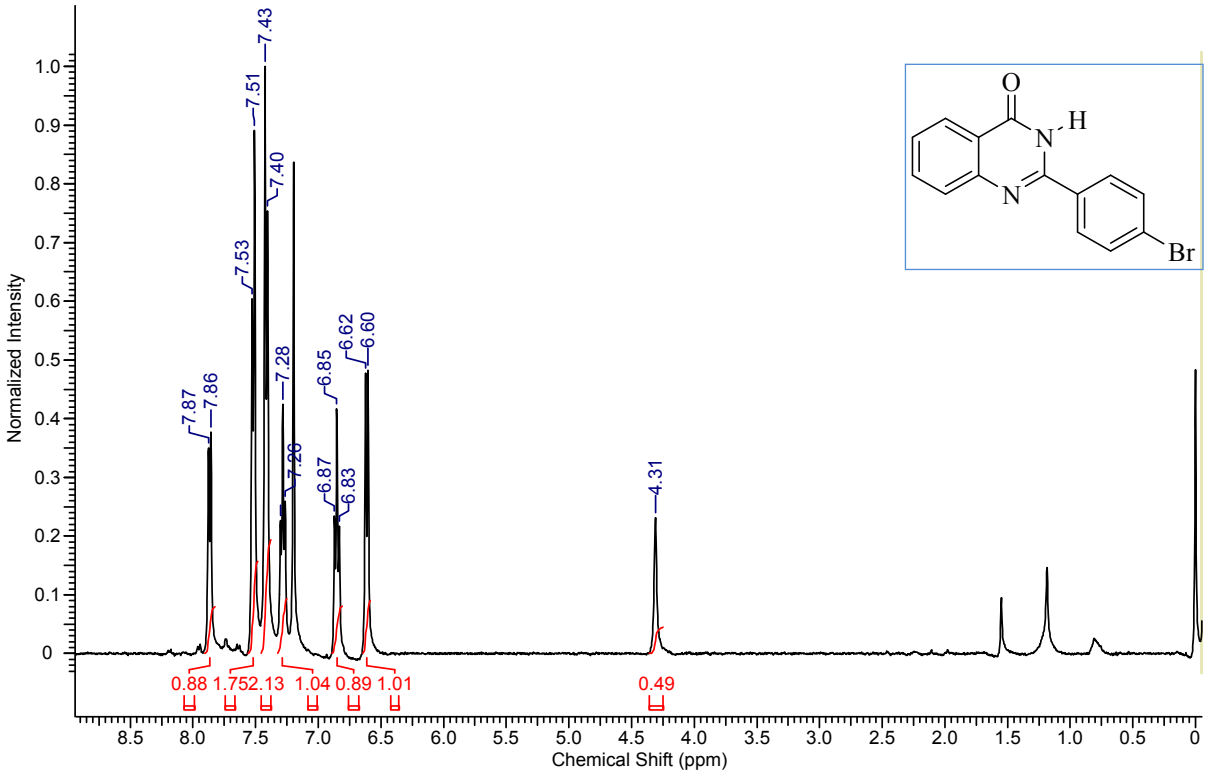

O
N
N
H
O
H
Normalized Intensity
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
196.57
167.65
152.09
150.82
142.93
141.07
138.67
134.61
134.32
132.18
130.99
123.15
119.77
78.20
25.76
200
180
160
140
120
100
80
60
40
20
0
Chemical Shift (ppm)

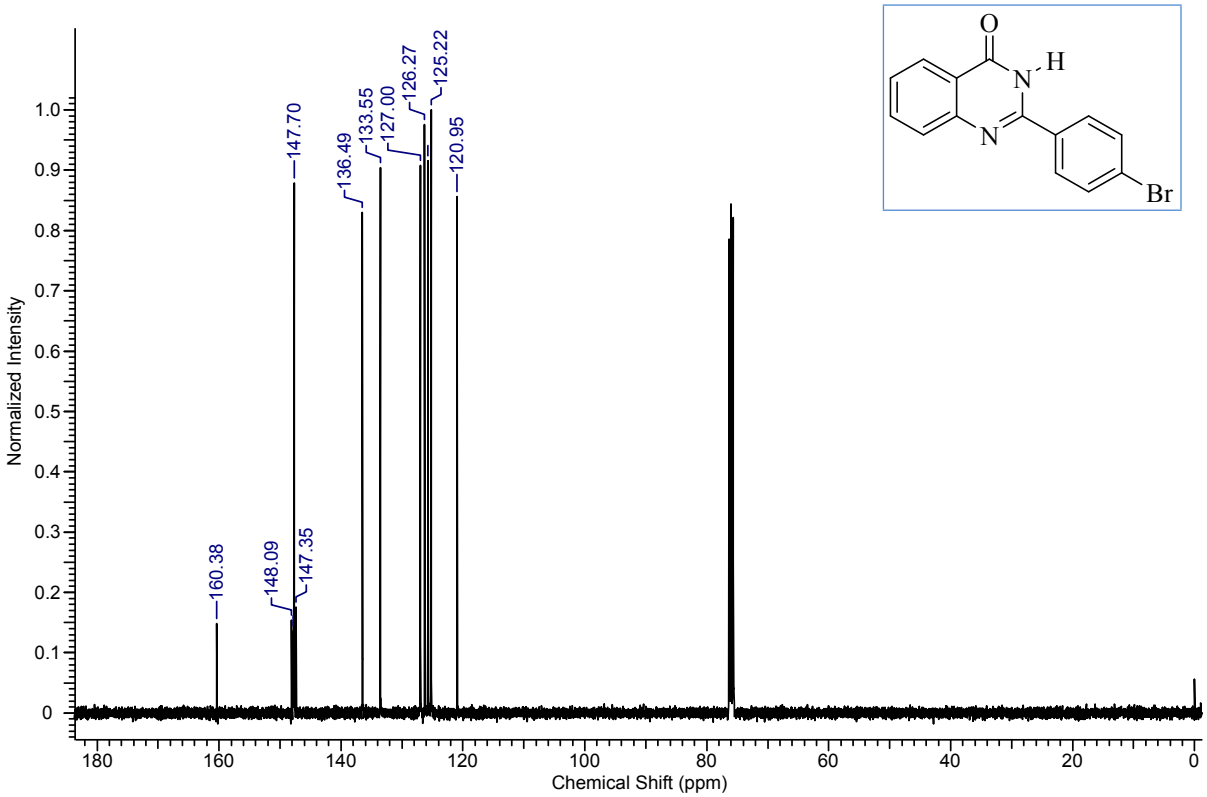
19. **6s**


Normalized Intensity
Chemical Shift (ppm)
3.89
2.29
7.27
7.95
7.93
7.45
7.08
7.07
6.13
8.03
7.05
6.91
6.65
6.63
0.99 1.95 2.17 1.26 1.83 4.13 1.04 1.21 1.04
3.11
3.03
O
N
N
H
O
O

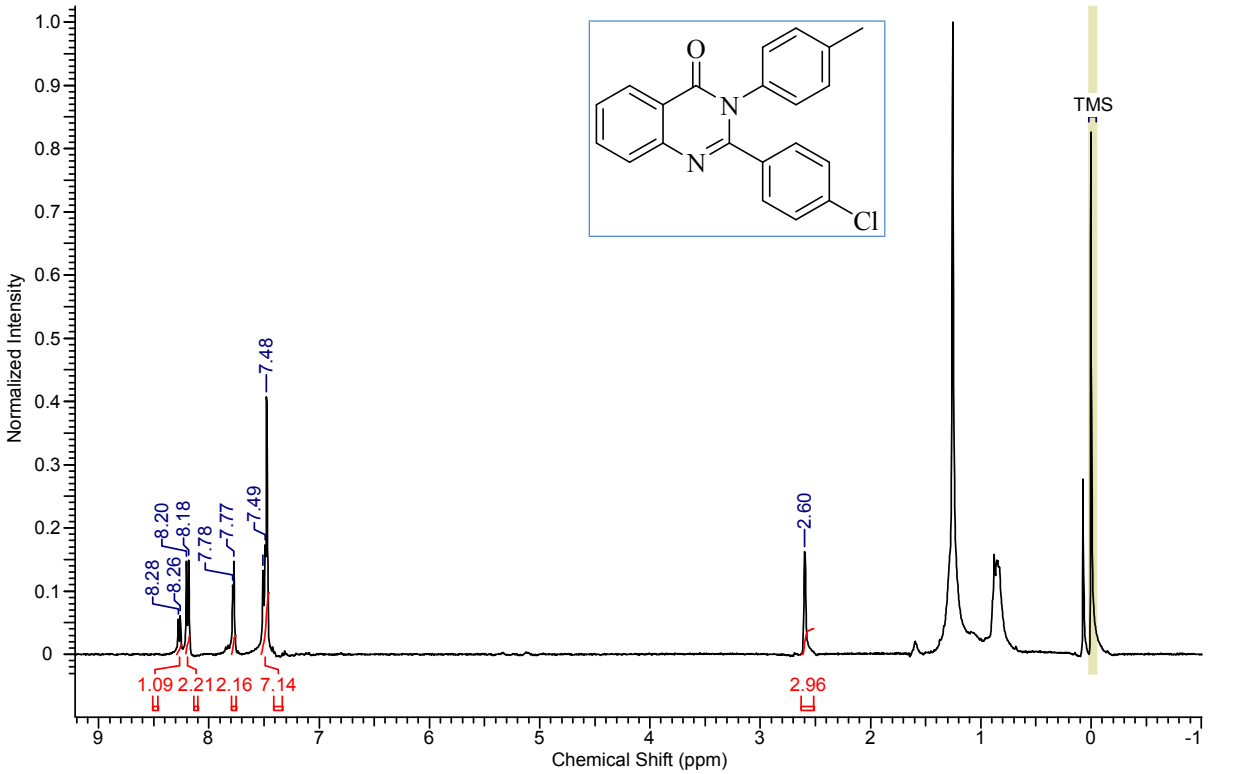

Normalized Intensity
Chemical Shift (ppm)
130.03
129.75
126.86
126.68
133.99
119.86
114.95
74.25
52.27
21.05
166.50
163.04
144.97
144.75
137.68
136.92
116.85

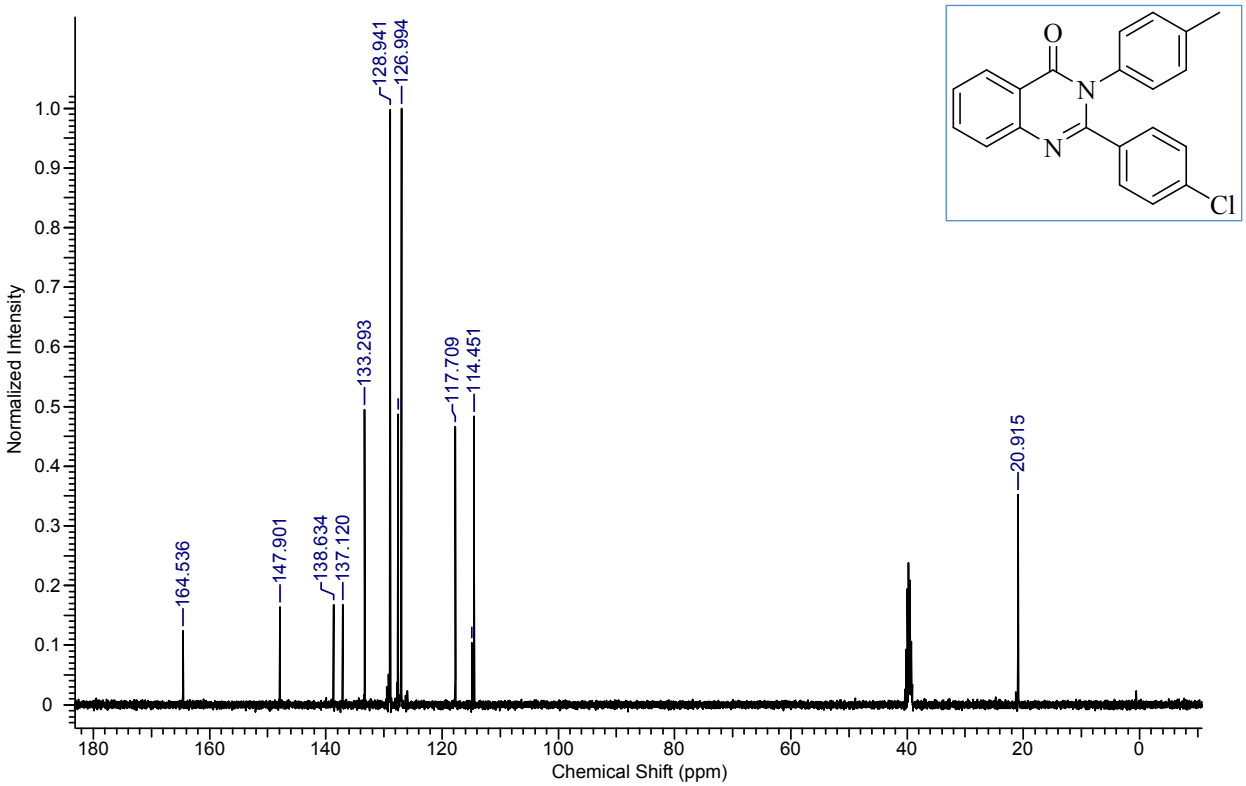
20. **7a**


Normalized Intensity
Chemical Shift (ppm)
7.87
7.86
7.53
7.51
7.43
7.40
7.28
7.26
6.87
6.85
6.83
6.62
6.60
4.31
0.88
1.75
2.13
1.04
0.89
1.01
0.49
O
N
H
N
Br


Normalized Intensity
Chemical Shift (ppm)
160.38
148.09
147.70
147.35
136.49
133.55
127.00
126.27
125.22
120.95
O
N
H
N
Br

21. **7b**


Normalized Intensity
Chemical Shift (ppm)
TMS
8.28
8.26
8.20
8.18
7.78
7.77
7.49
7.48
2.60
1.09
2.21
2.16
7.14
2.96
O
N
N
Cl


Normalized Intensity
Chemical Shift (ppm)
164.536
147.901
138.634
137.120
133.293
128.941
126.994
117.709
114.451
20.915
O
N
N
Cl