

Supporting Information

Fabrication of porous ultrathin carbon nitride nanosheets catalysts for the enhanced photocatalytic activity of N- and O- heterocyclic compound synthesis

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Results and discussion

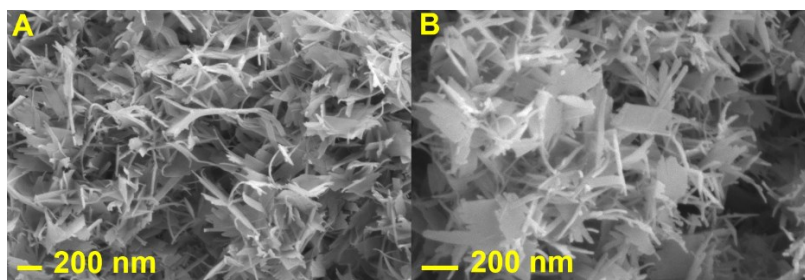


Figure S1. The CuO NB without calcination (A) and calcination at 350 °C for 120 min.

As shown in Figure S1, the CuO NB without calcination displays NB structure. After calcination at 350 °C for 120 min, the morphology has little change.

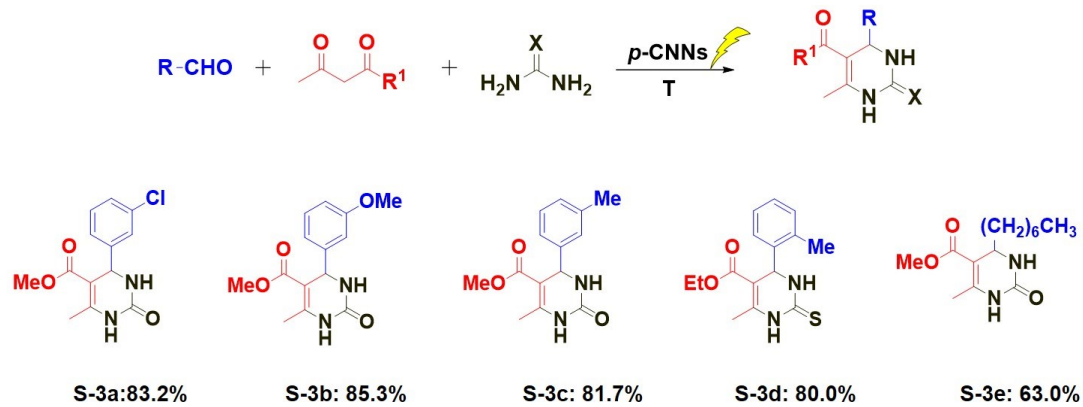


Figure S2. Photocatalytic synthesis of Biginelli reaction.

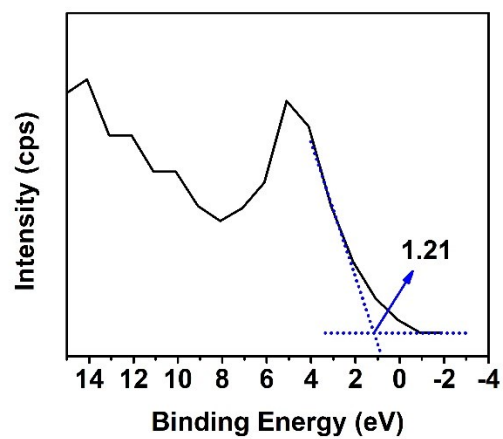


Figure S3. , XPS valence band spectrum of *p*-CNNs.

¹H NMR and ¹³C NMR of the products

3a: ¹H NMR (DMSO-*d*₆, 400 MHz, Me₄Si, 20 °C): δ ppm = 9.18 (s, 1H, NH), 7.72 (s, 1H, NH), 7.33-7.22 (m, 5H, Ar-H), 5.14 (s, 1H, CH), 3.97 (q, *J* = 8.0 Hz, 2H, OCH₂CH₃), 2.24 (s, 3H, CH₃), 1.08 (t, *J* = 8.0 Hz, 3H, OCH₂CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆, δ ppm): 165.31, 152.11, 148.32, 144.84, 128.36, 127.23, 126.22, 99.23, 59.16, 53.94, 17.75, 14.05.

3b: ¹H NMR (DMSO-*d*₆, 400 MHz, Me₄Si, 20 °C): δ ppm = 9.31, (s, 1H, OH), 9.08 (s, 1H, NH), 7.59 (s, 1H, NH), 7.01 (d, *J* = 8.0 Hz, 2H, Ar-H), 6.67 (d, *J* = 8.0 Hz, 2H, Ar-H), 5.03 (d, *J* = 4.0 Hz, 1H, CH), 3.96 (q, *J* = 8.0 Hz, 2H, OCH₂CH₃), 2.22 (s, 3H, CH₃), 1.08 (t, *J* = 8.0 Hz, 3H, OCH₂CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆, δ ppm): 165.44, 156.51, 152.16, 147.75, 135.43, 127.39, 114.96, 99.72, 59.09, 53.42, 17.72, 14.09.

3c: ¹H NMR (DMSO-*d*₆, 400 MHz, Me₄Si, 20 °C): δ ppm = 9.13 (s, 1H, NH), 7.65 (s, 1H, NH), 7.13 (d, *J* = 8.0 Hz, 2H, Ar-H), 6.87 (d, *J* = 8.0 Hz, 2H, Ar-H), 5.08 (d, *J* = 4.0 Hz, 1H, CH), 3.96 (q, *J* = 8.0 Hz, 2H, OCH₂CH₃), 3.71 (s, 3H, CH₃), 2.23 (s, 3H, CH₃), 1.09 (t, *J* = 8.0 Hz, 3H, OCH₂CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆, δ ppm): 165.38, 158.44, 152.15, 148.01, 137.05, 127.39, 113.71, 99.57, 59.15, 55.05, 53.33, 17.75, 14.10.

3d: ¹H NMR (DMSO-*d*₆, 400 MHz, Me₄Si, 20 °C): δ ppm = 9.13 (s, 1H, NH), 7.66 (s, 1H, NH), 7.10 (s, 4H, Ar-H), 5.09 (d, *J* = 4.0 Hz, 1H, CH), 3.96 (q, *J* = 8.0 Hz, 2H, OCH₂CH₃), 2.25 (s, 3H, CH₃), 2.22 (s, 3H, CH₃), 1.09 (t, *J* = 8.0 Hz, 3H, OCH₂CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆, δ ppm): 165.56, 152.38, 148.34, 142.14, 136.57, 129.09, 126.34, 99.62, 59.37, 53.82, 20.84, 17.95, 14.30.

3e: ^1H NMR (DMSO- d_6 , 400 MHz, Me $_4$ Si, 20 °C): δ ppm = 9.20 (s, 1H, OH), 9.05 (s, 1H, NH), 7.55 (s, 1H, NH), 6.90-6.66 (m, 3H, Ar-H), 5.09 (d, J = 4.0 Hz, 1H, CH), 3.95 (q, J = 8.0 Hz, 2H, OCH $_2$ CH $_3$), 2.21 (s, 3H, OCH $_3$), 2.06 (s, 3H, CH $_3$), 1.09 (t, J = 8.0 Hz, 3H, OCH $_2$ CH $_3$); ^{13}C NMR (100 MHz, DMSO- d_6 , δ ppm): 165.45, 154.56, 152.17, 147.61, 135.34, 128.64, 124.54, 123.38, 114.28, 99.79, 59.09, 53.95, 17.75, 16.16, 14.10.

3f: ^1H NMR (DMSO- d_6 , 400 MHz, Me $_4$ Si, 20 °C): δ ppm = 9.22 (s, 1H, NH), 7.75 (s, 1H, NH), 7.39 (d, J = 8.0 Hz, 2H, Ar-H), 7.24 (d, J = 8.0 Hz, 2H, Ar-H), 5.13 (d, J = 4.0 Hz, 1H, CH), 3.97 (q, J = 8.0 Hz, 2H, OCH $_2$ CH $_3$), 2.24 (s, 3H, CH $_3$), 1.08 (t, J = 8.0 Hz, 3H, OCH $_2$ CH $_3$); ^{13}C NMR (100 MHz, DMSO- d_6 , δ ppm): 165.21, 151.93, 148.72, 143.79, 131.78, 128.40, 128.18, 98.83, 59.26, 53.41, 17.79, 14.07.

3g: ^1H NMR (DMSO- d_6 , 400 MHz, Me $_4$ Si, 20 °C): δ ppm = 9.23 (s, 1H, NH), 7.76 (s, 1H, NH), 7.52 (d, J = 8.0 Hz, 2H, Ar-H), 7.19 (d, J = 8.0 Hz, 2H, Ar-H), 5.12 (d, J = 4.0 Hz, 1H, CH), 3.97 (q, J = 8.0 Hz, 2H, OCH $_2$ CH $_3$), 2.24 (s, 3H, CH $_3$), 1.08 (t, J = 8.0 Hz, 3H, OCH $_2$ CH $_3$); ^{13}C NMR (100 MHz, DMSO- d_6 , δ ppm): 165.21, 151.96, 148.74, 144.21, 131.33, 128.57, 120.33, 98.79, 59.29, 53.51, 17.82, 14.08.

3h: ^1H NMR (DMSO- d_6 , 400 MHz, Me $_4$ Si, 20 °C): δ ppm = 9.20 (s, 1H, NH), 7.73 (s, 1H, NH), 7.27-7.23 (m, 2H, Ar-H), 7.16-7.11 (m, 2H, Ar-H), 5.14 (d, J = 4.0 Hz, 1H, CH), 4.00-3.94 (m, 2H, OCH $_2$ CH $_3$), 2.24 (s, 3H, CH $_3$), 1.08 (t, J = 8.0 Hz, 3H, OCH $_2$ CH $_3$); ^{13}C NMR (100 MHz, DMSO- d_6 , δ ppm): 162.24, 162.51, 160.10, 151.97, 148.52, 141.14 (d, J = 16.0 Hz), 128.28 (d, J = 32.0 Hz), 115.22 (d, J = 88.0 Hz), 99.13, 59.21, 53.33, 17.77, 14.06.

3i: ^1H NMR (DMSO- d_6 , 400 MHz, Me $_4$ Si, 20 °C): δ ppm = 9.17 (s, 1H, NH), 7.71 (s, 1H, NH), 7.25-7.21 (m, 1H, Ar-H), 6.82-6.76 (m, 3H, Ar-H), 5.11 (d, J = 4.0 Hz, 1H, CH), 3.98 (q, J = 8.0 Hz, 2H, OCH $_2$ CH $_3$), 3.71 (s, 3H, OCH $_3$), 2.23 (s, 3H, CH $_3$), 1.10 (t, J = 8.0 Hz, 3H, OCH $_2$ CH $_3$); ^{13}C NMR (100 MHz, DMSO- d_6 , δ ppm): 165.80, 159.66, 152.17, 148.88, 146.77, 130.01, 118.69, 112.85, 112.58, 99.60, 59.68, 55.41, 54.19, 18.21, 14.56.

3j: ^1H NMR (DMSO- d_6 , 400 MHz, Me $_4$ Si, 20 °C): δ ppm = 9.14 (s, 1H, NH), 7.67 (s, 1H, NH), 7.21-7.17 (m, 1H, Ar-H), 7.05-7.00 (m, 3H, Ar-H), 5.10 (d, J = 4.0 Hz, 1H, CH), 4.00-3.95 (m, 2H, OCH $_2$ CH $_3$), 2.67 (s, 3H, OCH $_3$), 2.30 (s, 3H, CH $_3$), 1.09 (t, J = 8.0 Hz, 3H, OCH $_2$ CH $_3$); ^{13}C NMR (100 MHz, DMSO- d_6 , δ ppm): 165.34, 152.12, 148.21, 144.84, 137.32, 128.30, 127.88, 126.83, 123.33, 99.27, 59.15, 53.93, 21.13, 17.77, 14.16.

3k: ^1H NMR (DMSO- d_6 , 400 MHz, Me $_4$ Si, 20 °C): δ ppm = 8.97 (s, 1H, NH), 7.19 (s, 1H, NH), 4.12-4.10 (s, 2H, OCH $_2$ CH $_3$), 4.08-4.00 (m, 1H, CH), 2.14 (s, 3H, CH $_3$), 1.20 (s, 3H, CH $_3$), 1.16 (s, 3H, OCH $_2$ CH $_3$); ^{13}C NMR (100 MHz, DMSO- d_6 , δ ppm): 165.83, 152.99, 148.23, 100.95, 59.54, 47.76, 23.87, 18.15, 14.71.

3l: ^1H NMR (DMSO- d_6 , 400 MHz, Me $_4$ Si, 20 °C): δ ppm = 8.90 (s, 1H, NH), 7.27 (s, 1H, NH), 4.09 (d, J = 8.0 Hz, 2H, OCH $_2$ CH $_3$), 4.05-3.99 (m, 1H, CH), 2.15 (s, 3H, CH $_3$), 1.42 (q, J = 4.0 Hz, CH $_2$ CH $_3$), 1.19 (s, 3H, CH $_2$ CH $_3$), 1.17 (s, 3H, OCH $_2$ CH $_3$); ^{13}C NMR (100 MHz, DMSO- d_6 , δ ppm): 165.49, 152.78, 148.42, 98.71, 59.00, 51.31, 29.61, 17.69, 14.21, 8.48.

3m: ^1H NMR (DMSO- d_6 , 400 MHz, Me $_4$ Si, 20 °C): δ ppm = 9.26 (s, 1H, NH), 7.77 (s,

1H, NH), 7.39 (d, $J = 8.0$ Hz, 2H, Ar-H), 7.25 (d, $J = 8.0$ Hz, 2H, Ar-H), 5.14 (d, $J = 4.0$ Hz, 1H, CH), 3.52 (s, 3H, OCH₃), 2.24 (s, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆, δ ppm): 165.73, 152.00, 149.01, 143.61, 131.84, 128.47, 128.13, 98.62, 53.28, 80.84, 17.86.

3n: ¹H NMR (DMSO-*d*₆, 400 MHz, Me₄Si, 20 °C): δ ppm = 9.24 (s, 1H, NH), 7.76 (s, 1H, NH), 7.27-7.24 (s, 2H, Ar-H), 7.15-7.11 (m, 2H, Ar-H), 5.14 (d, $J = 4.0$ Hz, 1H, CH), 3.52 (s, 3H, OCH₃), 2.24 (s, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆, δ ppm): 165.77, 162.55, 160.14, 152.02, 148.82, 140.95 (d, $J = 16.0$ Hz), 128.22 (d, $J = 32.0$ Hz), 115.29 (d, $J = 88.0$ Hz), 98.92, 53.18, 50.79, 17.84.

3o: ¹H NMR (DMSO-*d*₆, 400 MHz, Me₄Si, 20 °C): δ ppm = 9.19 (s, 1H, NH), 7.69 (s, 1H, NH), 7.15 (d, $J = 8.0$ Hz, 2H, Ar-H), 6.87 (d, $J = 8.0$ Hz, 2H, Ar-H), 5.10 (d, $J = 4.0$ Hz, 1H, CH), 3.70 (s, 3H, OCH₃), 3.51 (s, 3H, CH₃), 2.4 (s, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆, δ ppm): 165.91, 158.51, 152.26, 148.34, 136.89, 127.38, 113.80, 99.37, 55.07, 53.25, 50.78, 17.83.

3p: ¹H NMR (DMSO-*d*₆, 400 MHz, Me₄Si, 20 °C): δ ppm = 9.34 (s, 1H, OH), 9.15 (s, 1H, NH), 7.65 (s, 1H, NH), 7.01 (s, 2H, Ar-H), 6.68 (s, 2H, NH), 5.03 (s, 1H, CH), 3.51 (s, 3H, OCH₃), 2.23 (s, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆, δ ppm): 166.37, 157.03, 152.68, 148.58, 135.66, 127.80, 115.51, 99.89, 53.67, 51.23, 18.23.

3q: ¹H NMR (DMSO-*d*₆, 400 MHz, Me₄Si, 20 °C): δ ppm = 10.29 (s, 1H, OH), 9.59 (s, 1H, NH), 9.43 (s, 1H, NH), 7.01 (s, 2H, Ar-H), 6.71 (s, 2H, Ar-H), 5.07 (s, 1H, CH), 3.53 (s, 3H, OCH₃), 2.28 (s, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆, δ ppm): 174.34, 166.19, 157.43, 145.35, 134.37, 128.18, 115.79, 101.15, 53.89, 51.62, 17.79.

3r: ^1H NMR (DMSO- d_6 , 400 MHz, Me $_4$ Si, 20 °C): δ ppm = 8.99 (s, 1H, NH), 7.20 (s, 1H, NH), 4.11 (t, J = 4.0 Hz, 1H, CH), 3.59 (s, 3H, OCH $_3$), 2.14 (s, 3H, CH $_3$), 1.08 (s, 3H, CH $_3$); ^{13}C NMR (100 MHz, DMSO- d_6 , δ ppm): 165.81, 152.46, 147.97, 100.26, 50.73, 46.25, 23.41, 17.70.

12-phenyl-9,9-dimethyl-8,9,10,12-tetrahydrobenzo[*a*]xanthen-11-one: ^1H NMR (DMSO- d_6 , 400 MHz, Me $_4$ Si, 20 °C): δ ppm = 8.05 (s, 1H), 7.92 (s, 2H), 7.61-7.04 (m, 8H), 5.57 (s, 1H), 2.72-2.57 (m, 2H), 2.36-2.10 (m, 2H), 1.06 (s, 3H), 0.88 (s, 3H); ^{13}C NMR (100 MHz, DMSO- d_6 , δ ppm): 195.88, 163.81, 147.11, 144.85, 131.05, 130.59, 129.07, 128.51, 128.10, 127.11, 126.18, 124.95, 123.24, 117.28, 117.13, 113.19, 50.08, 40.19, 34.06, 31.90, 28.80, 26.18.

5-phenyl-1(4-methoxyphenyl)-3[(4-methoxyphenyl)-amino]-1*H*-pyrrol-2(5*H*)-one: ^1H NMR (DMSO- d_6 , 400 MHz, Me $_4$ Si, 20 °C): δ ppm = 7.86 (s, 1H), 7.47 (s, 2H), 7.21 (s, 7H), 6.85 (s, 4H), 6.11 (s, 1H), 5.92 (s, 1H), 3.68 (s, 6H); ^{13}C NMR (100 MHz, DMSO- d_6 , δ ppm): 166.34, 156.17, 153.36, 138.28, 135.50, 132.63, 130.19, 128.66, 127.65, 126.82, 123.50, 118.28, 114.29, 113.86, 107.24, 62.81, 55.16, 55.08.

S-3a: ^1H NMR (DMSO- d_6 , 400 MHz, Me $_4$ Si, 20 °C): δ ppm = 9.29 (s, 1H, NH), 7.69 (s, 1H, NH), 7.40-7.38 (m, 1H, Ar-H), 7.30-7.26 (m, 3H, Ar-H), 5.60 (s, 1H, CH), 3.44 (s, 3H, OCH $_3$), 2.29 (s, 3H, CH $_3$); ^{13}C NMR (100 MHz, DMSO- d_6 , δ ppm): 165.48, 151.42, 149.47, 141.54, 131.67, 129.47, 129.13, 128.67, 127.77, 97.73, 51.39, 50.74, 17.75.

S-3b: ^1H NMR (DMSO- d_6 , 400 MHz, Me $_4$ Si, 20 °C): δ ppm = 9.26 (s, 1H, NH), 7.78 (s, 1H, NH), 7.25-7.23 (m, 1H, Ar-H), 6.91-6.73 (m, 3H, Ar-H), 5.12 *s, 1H, CH), 3.72

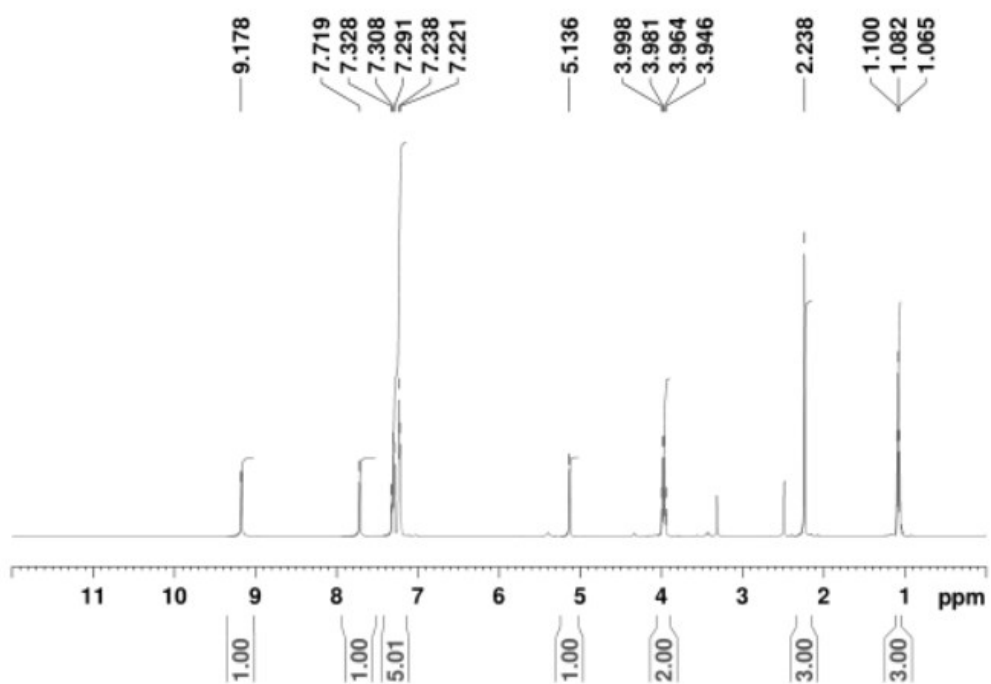
(s, 3H, OCH₃), 3.55 (s, 3H, OCH₃), 2.26 (s, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆, δ ppm): 165.91, 159.31, 157.76, 152.31, 148.80, 146.18, 129.67, 118.18, 112.43, 112.22, 98.89, 55.01, 53.69, 50.91, 17.85.

S-3c: ¹H NMR (DMSO-*d*₆, 400 MHz, Me₄Si, 20 °C): δ ppm = 9.21 (s, 1H, NH), 9.09 (s, 1H, NH), 7.57 (s, 1H, Ar-H), 6.89 (s, 1H, Ar-H), 6.83-6.80 (m, 1H, Ar-H), 6.68-6.66 (m, 1H, Ar-H), 5.01 (d, *J* = 4.0 Hz, 1H, CH), 3.51 (s, 3H, OCH₃), 2.22 (s, 3H, CH₃), 2.07 (s, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆, δ ppm): 165.94, 154.62, 152.19, 147.97, 135.09, 128.53, 124.48, 123.51, 114.33, 99.47, 53.34, 50.70, 17.80, 16.18.

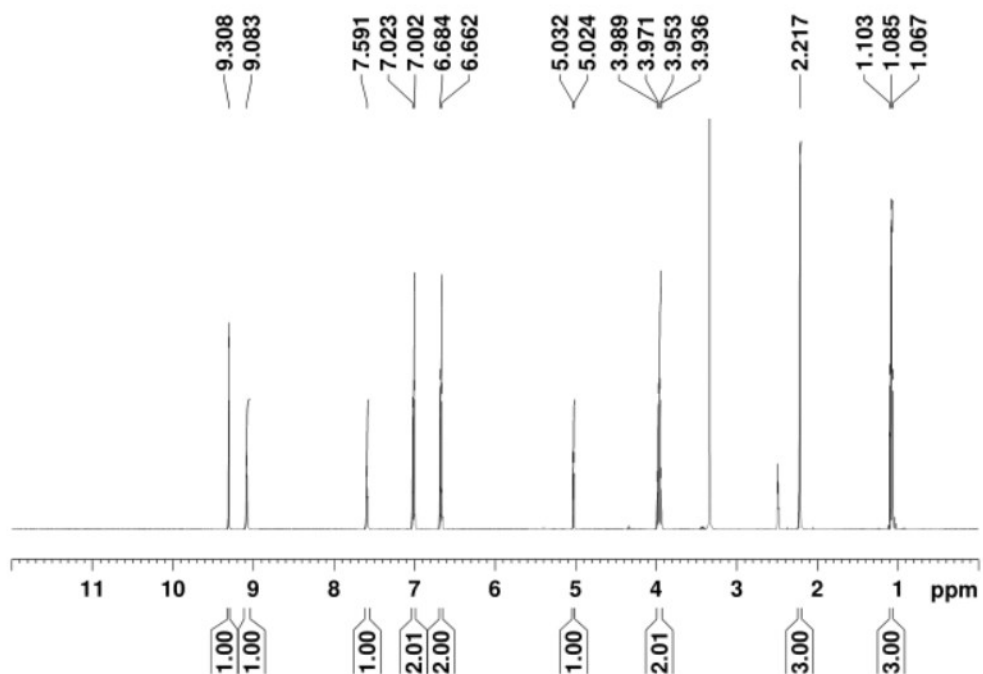
S-3d: ¹H NMR (DMSO-*d*₆, 400 MHz, Me₄Si, 20 °C): δ ppm = 9.18 (s, 1H, NH), 7.66 (s, 1H, NH), 7.16-7.12 (m, 4H, Ar-H), 5.40 (s, 1H, CH), 3.88 (s, 2H, OCH₂CH₃), 2.42 (s, 3H, CH₃), 2.29 (s, 3H, CH₃), 0.99 (s, 3H, OCH₂CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆, δ ppm): 165.71, 152.07, 148.95, 143.75, 135.13, 130.56, 127.62, 126.99, 99.62, 59.54, 50.88, 19.12, 18.15, 14.39.

S-3e: ¹H NMR (DMSO-*d*₆, 400 MHz, Me₄Si, 20 °C): δ ppm = 8.93 (s, 1H, NH), 7.30 (s, 1H, NH), 4.03-4.01 (s, 1H, CH), 3.59 (s, 3H, OCH₃), 2.14 (s, 3H, (CH₂)₆CH₃), 1.37-1.22 (m, 12H, (CH₂)₆CH₃), 0.86 (s, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆, δ ppm): 165.91, 152.70, 148.44, 99.16, 50.69, 50.06, 36.68, 31.22, 28.46, 23.59, 21.99, 17.71, 13.93.

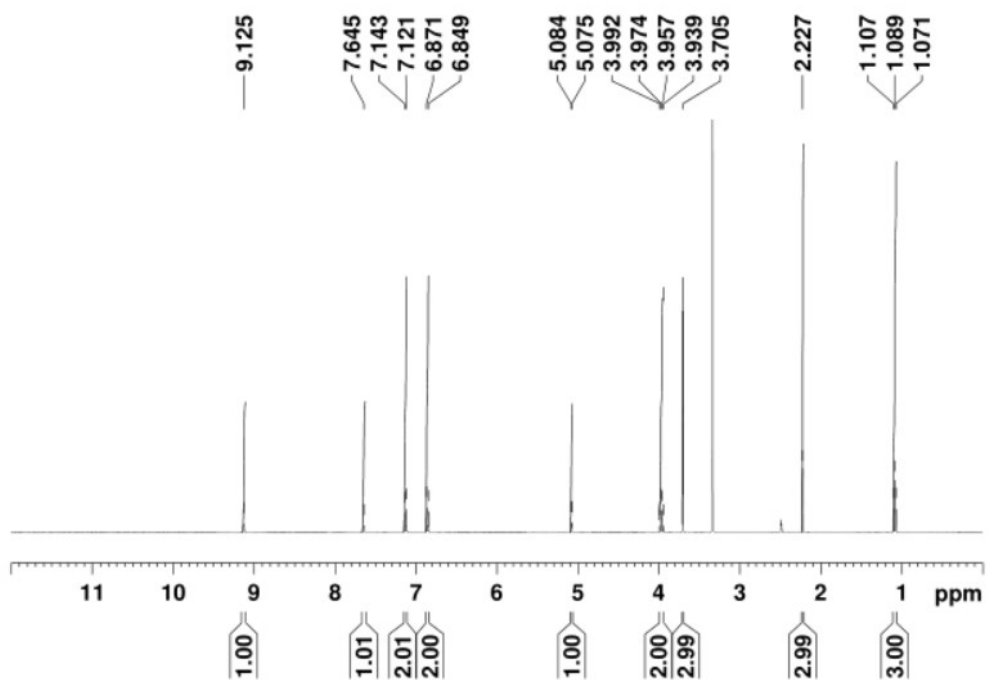
3a-¹H NMR:



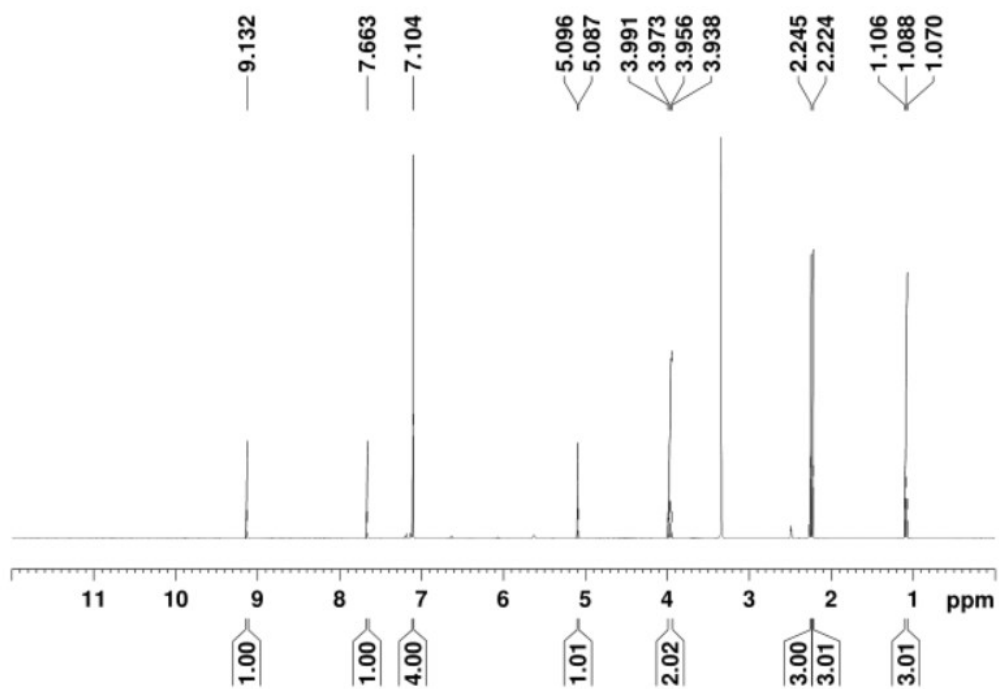
3b-¹H NMR:



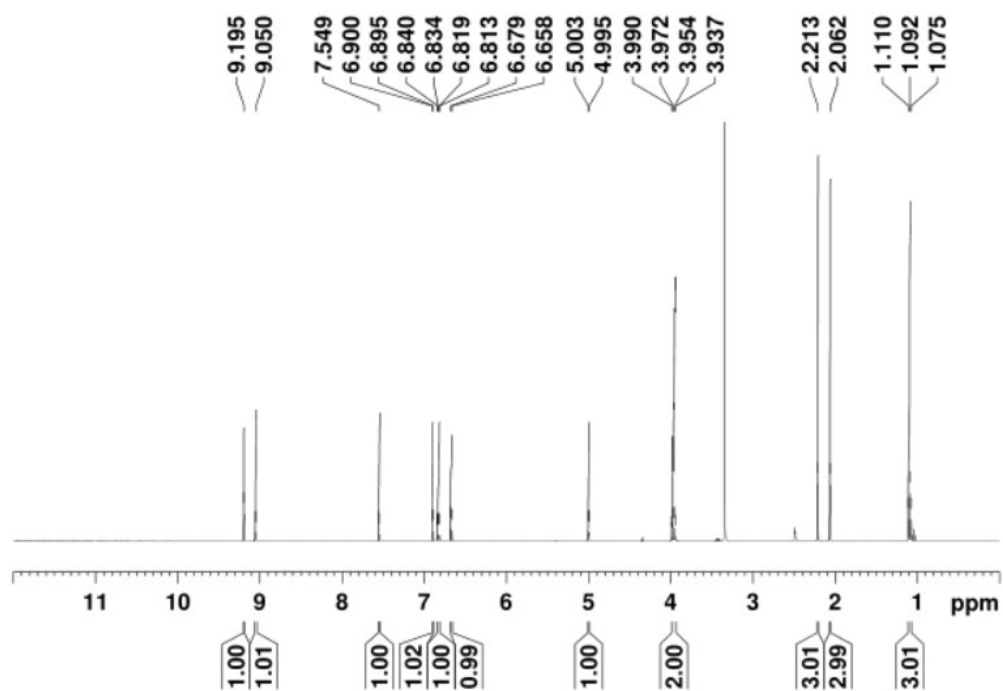
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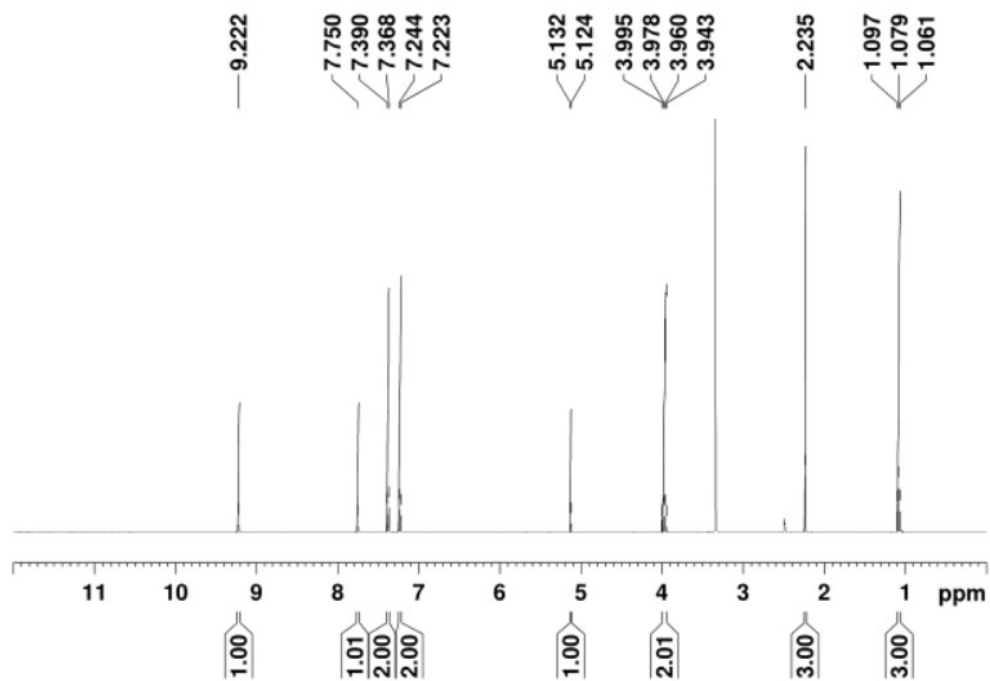
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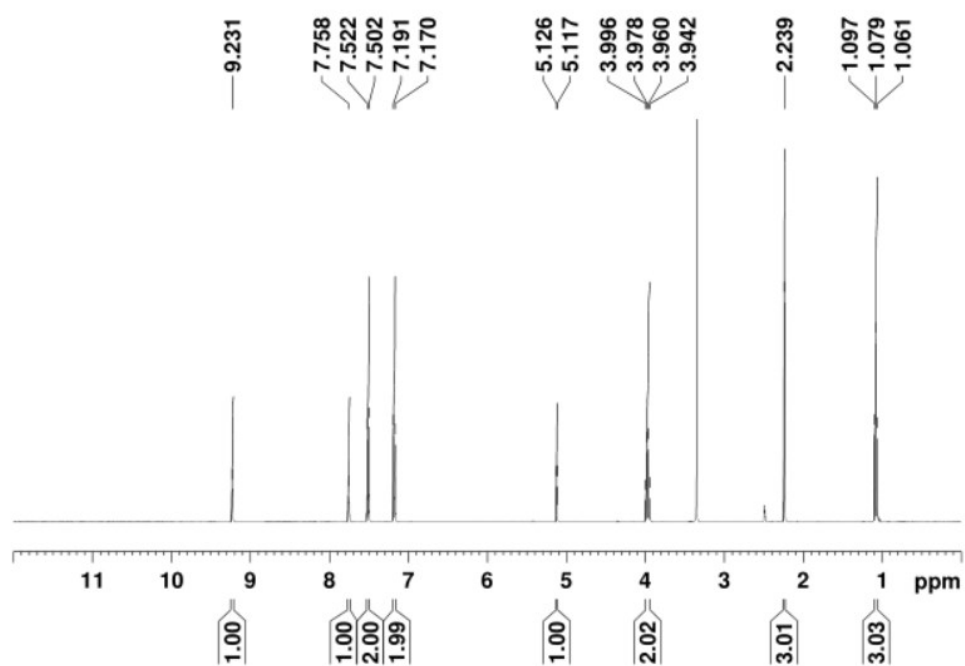
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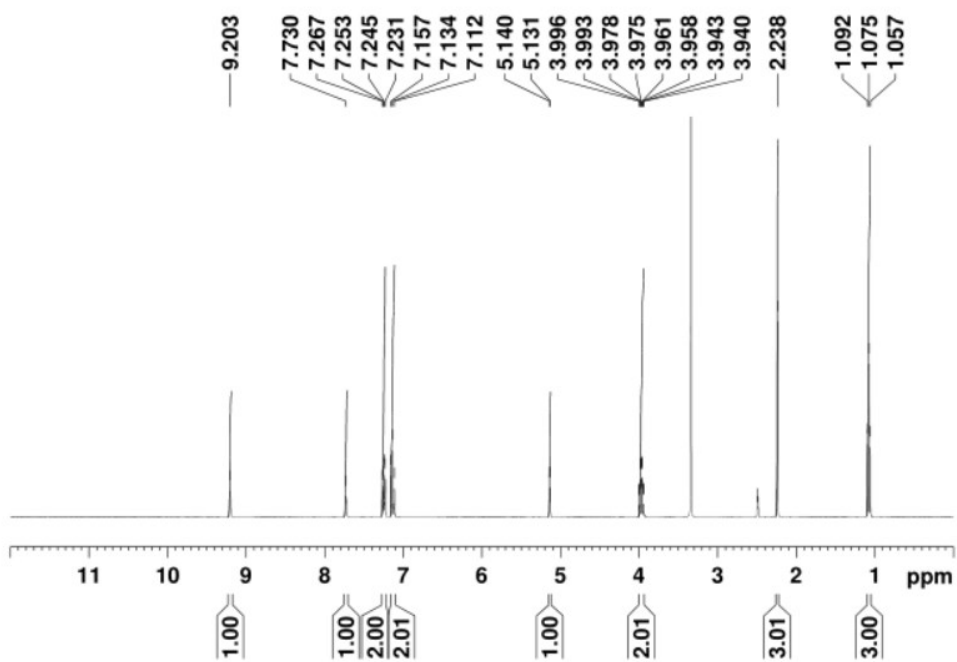
3f-¹H NMR:



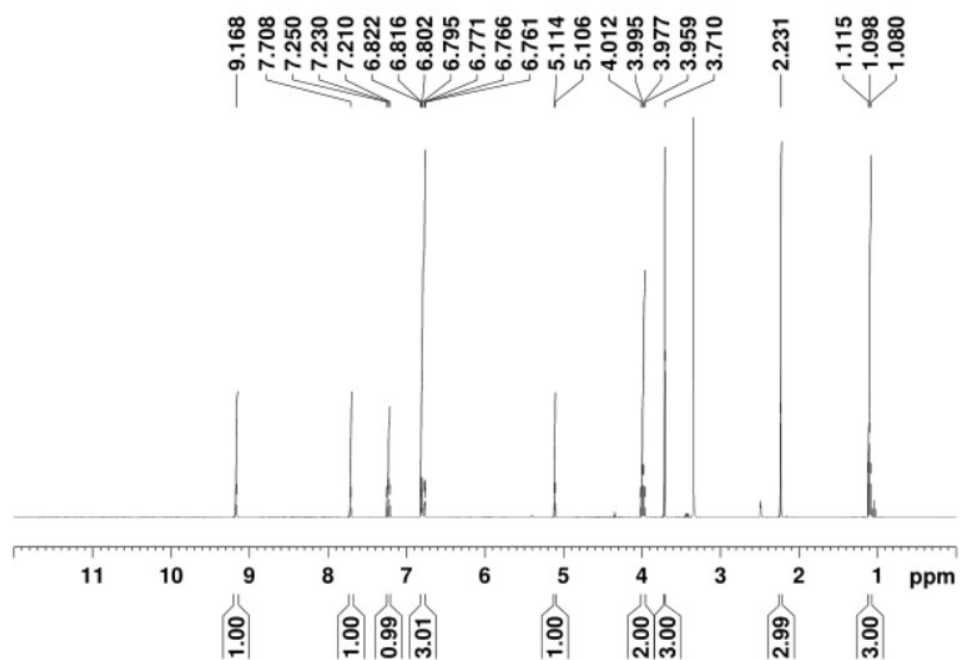
3g-¹H NMR:



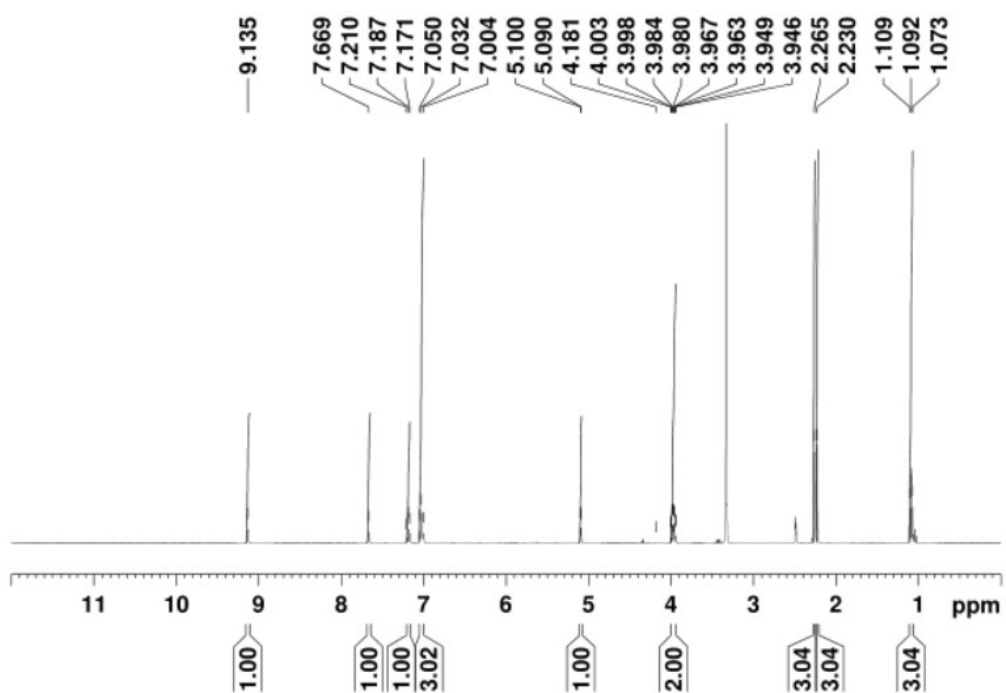
3h-¹H NMR:



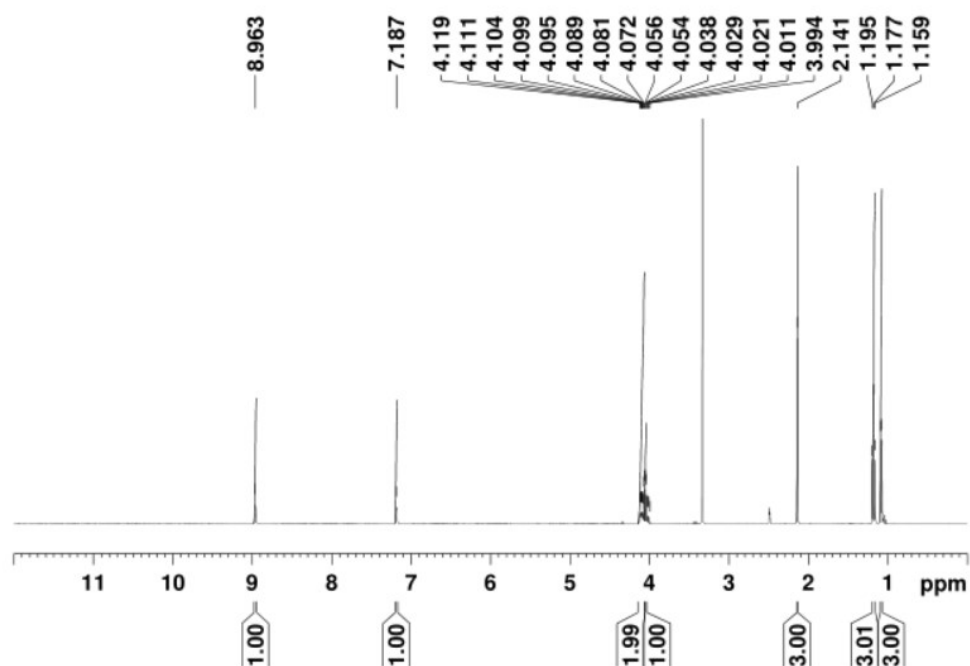
3i-¹H NMR:



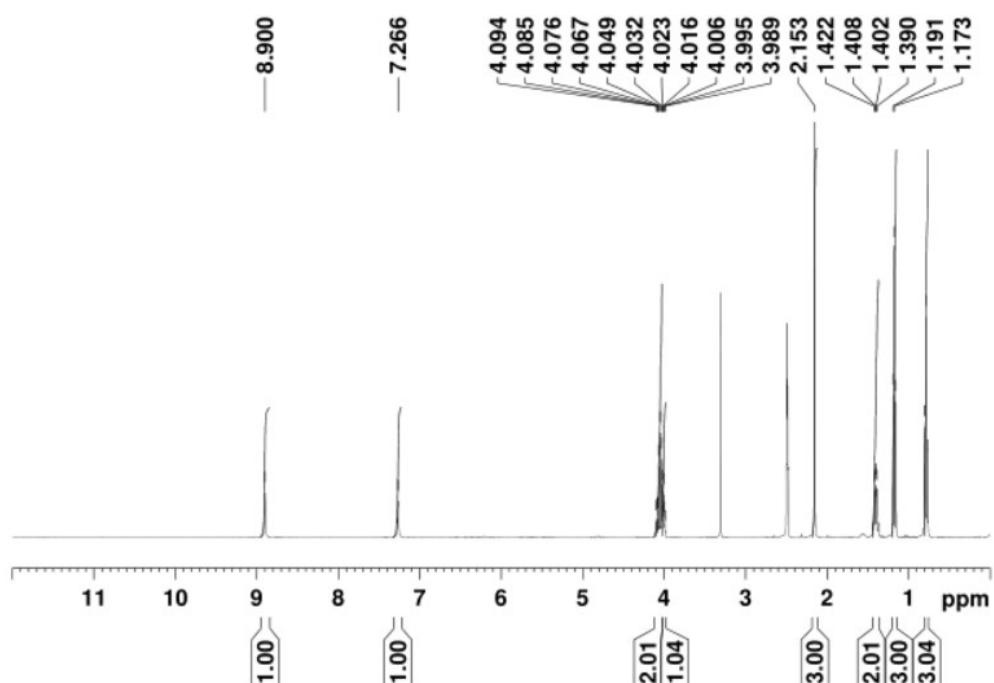
3j-¹H NMR:



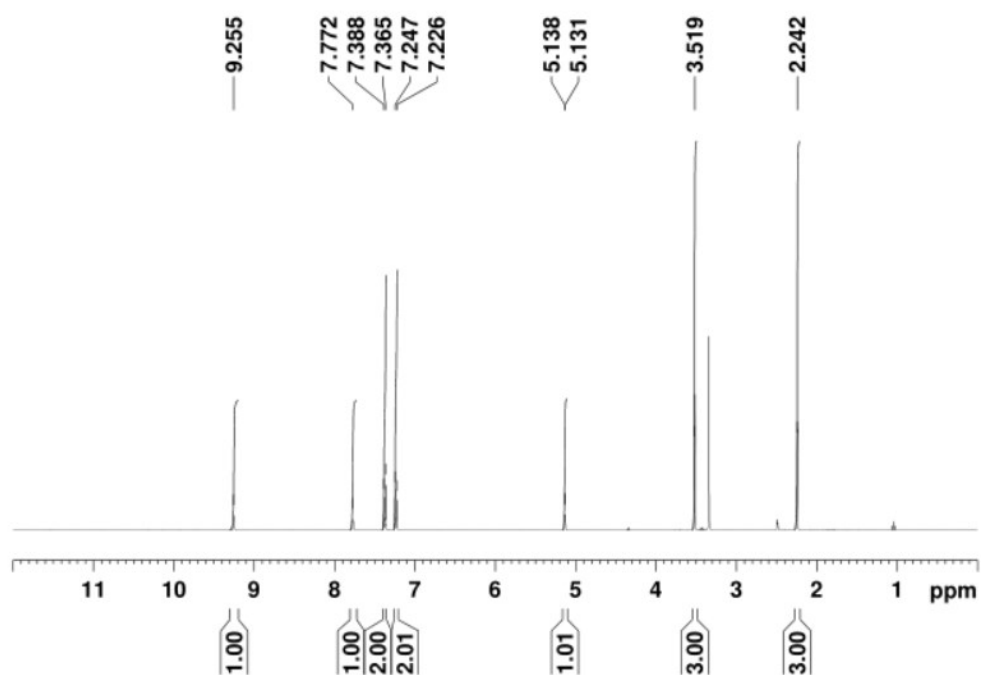
3k-¹H NMR:



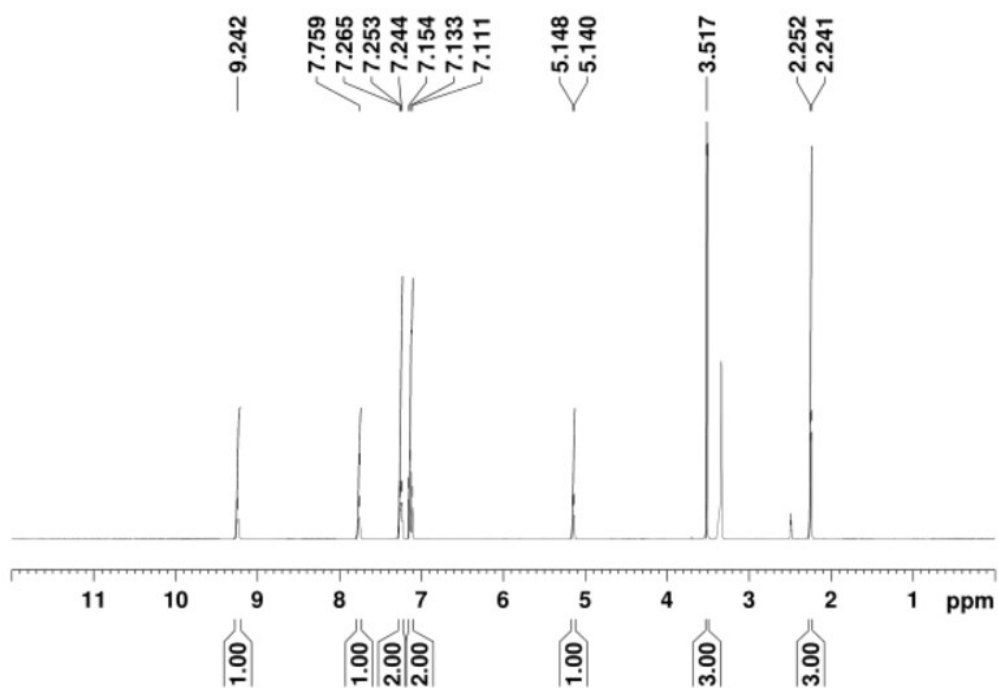
3l-¹H NMR:



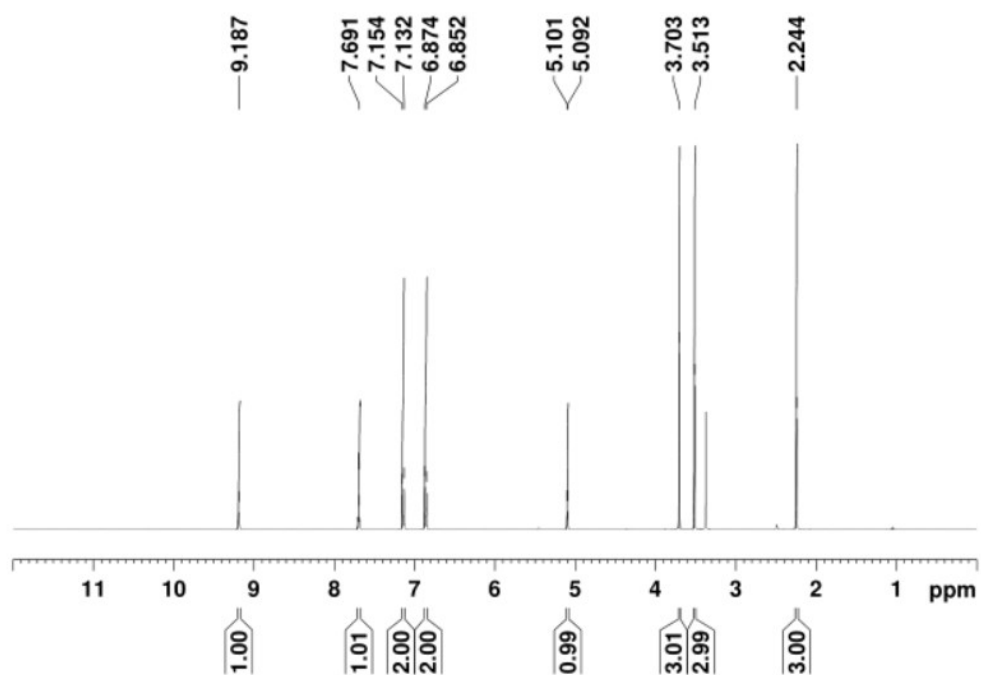
3m-¹H NMR:



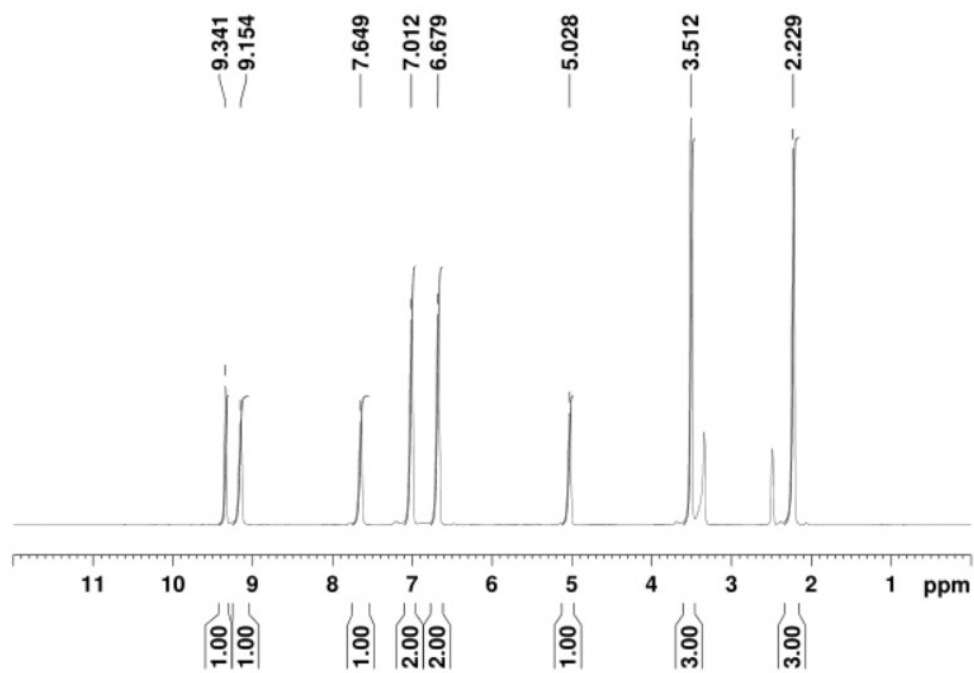
3n-¹H NMR:



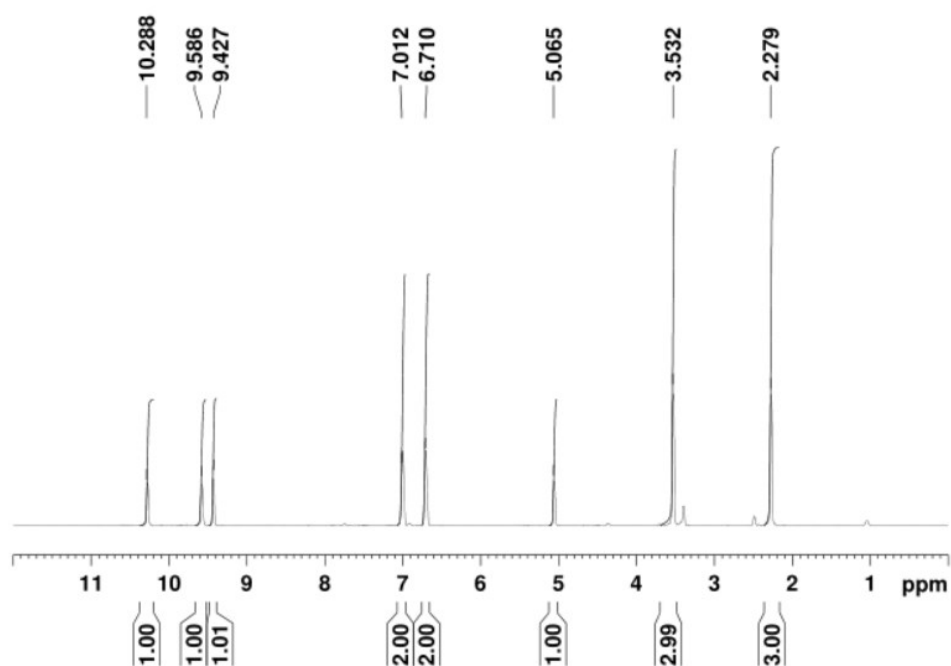
3o-¹H NMR:



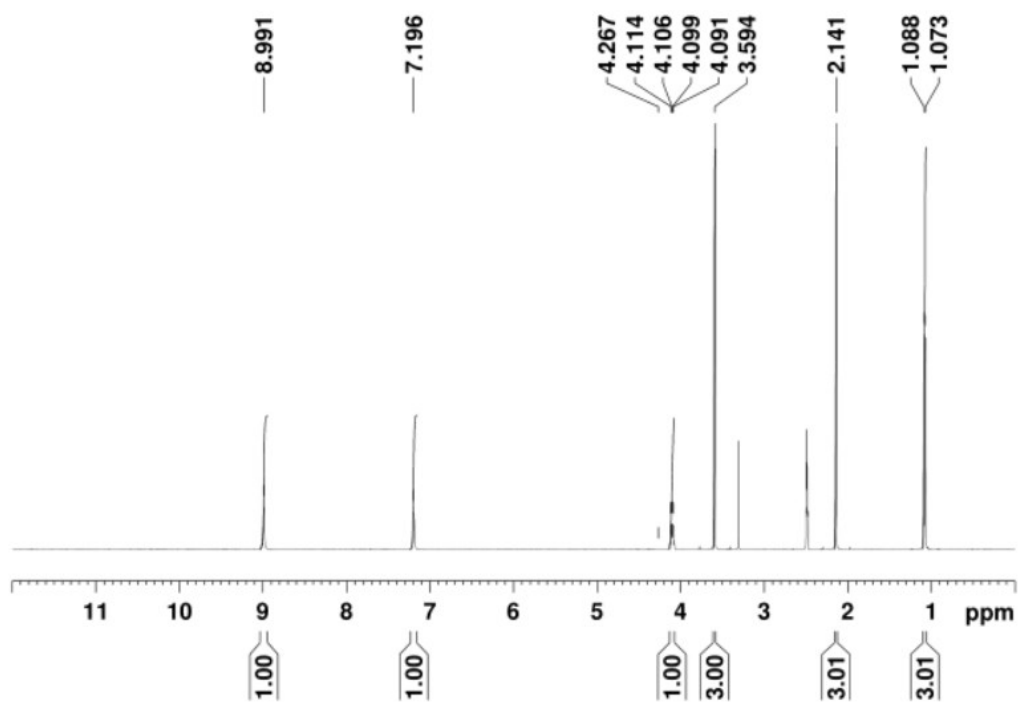
3p-¹H NMR:



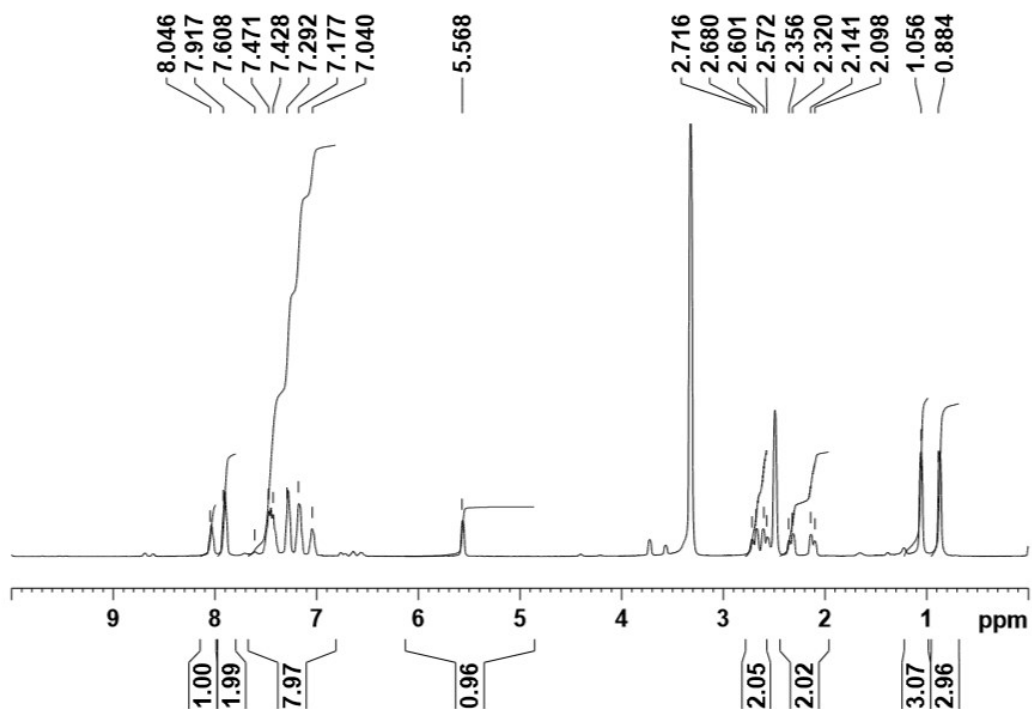
3q-¹H NMR:



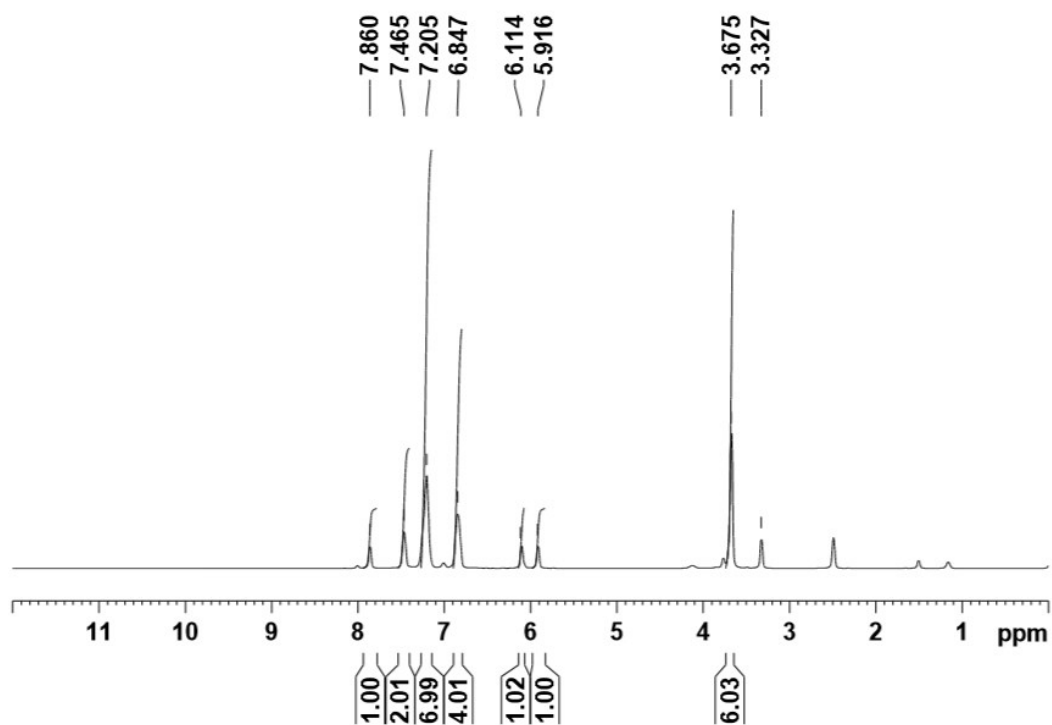
3r-¹H NMR:



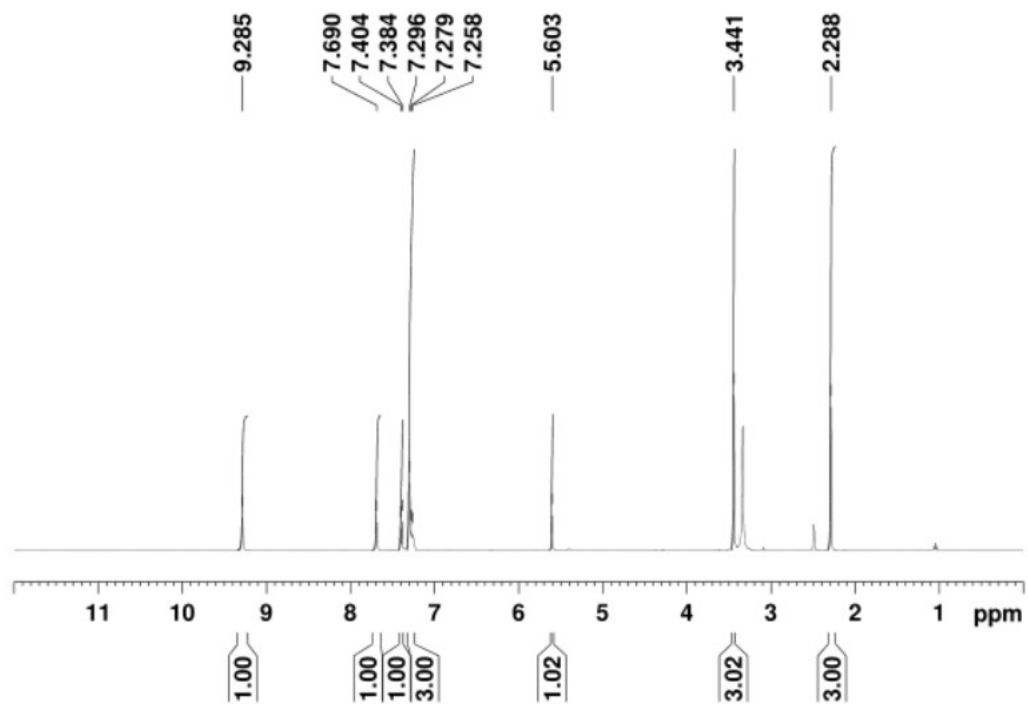
12-phenyl-9,9-dimethyl-8,9,10,12-tetrahydrobenzo[*a*]xanthen-11-one-¹H NMR:



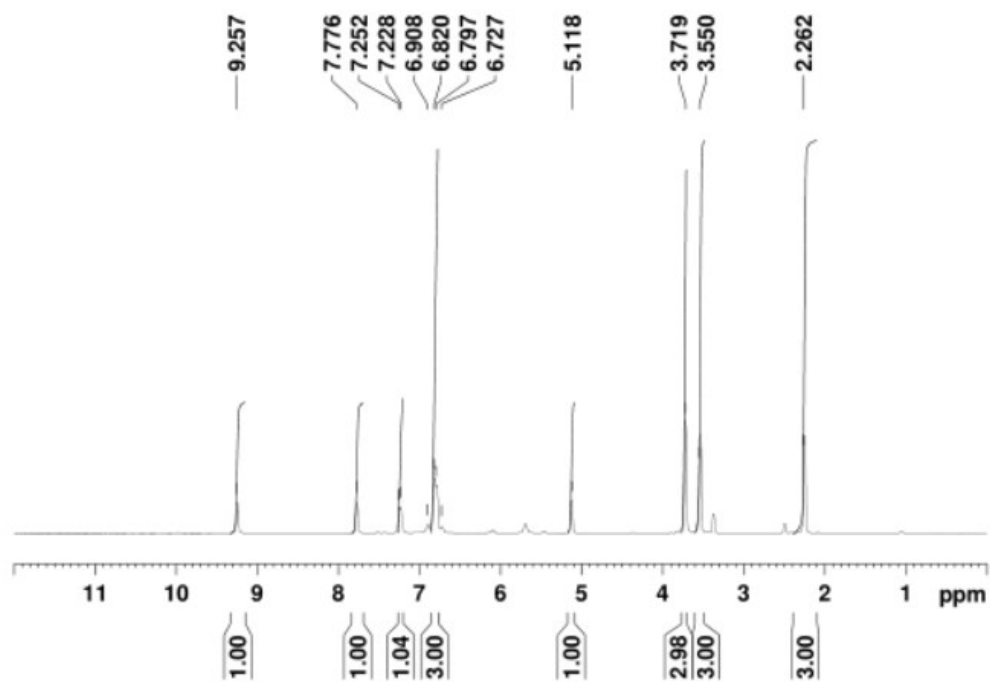
5-phenyl-1(4-methoxyphenyl)-3[(4-methoxyphenyl)-amino]-1*H*-pyrrol-2(5*H*)-one-¹H NMR:



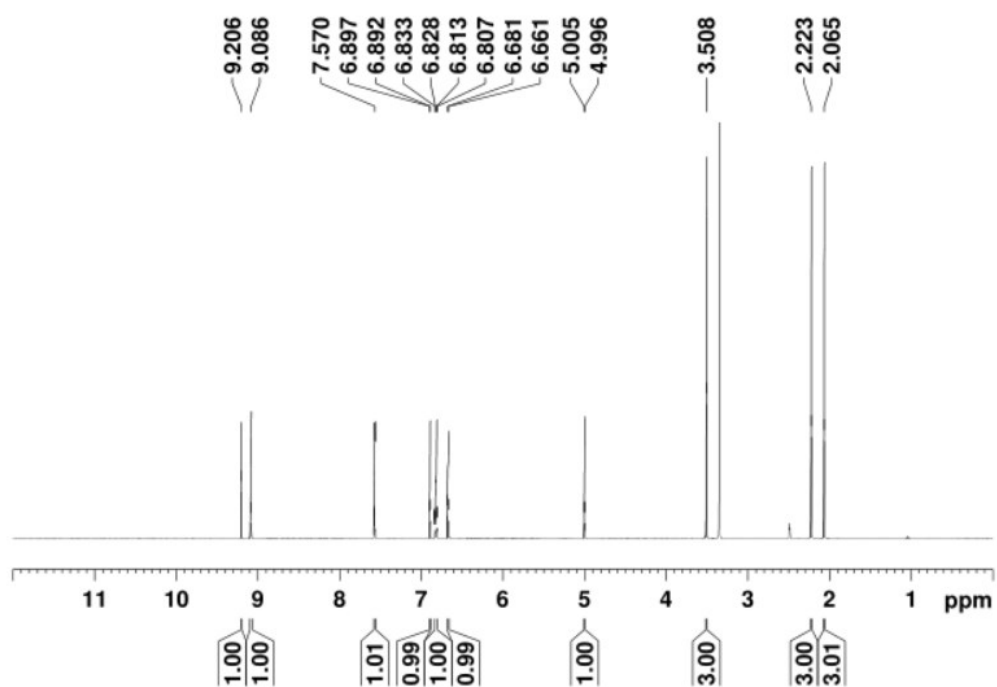
S-3a-¹H NMR:



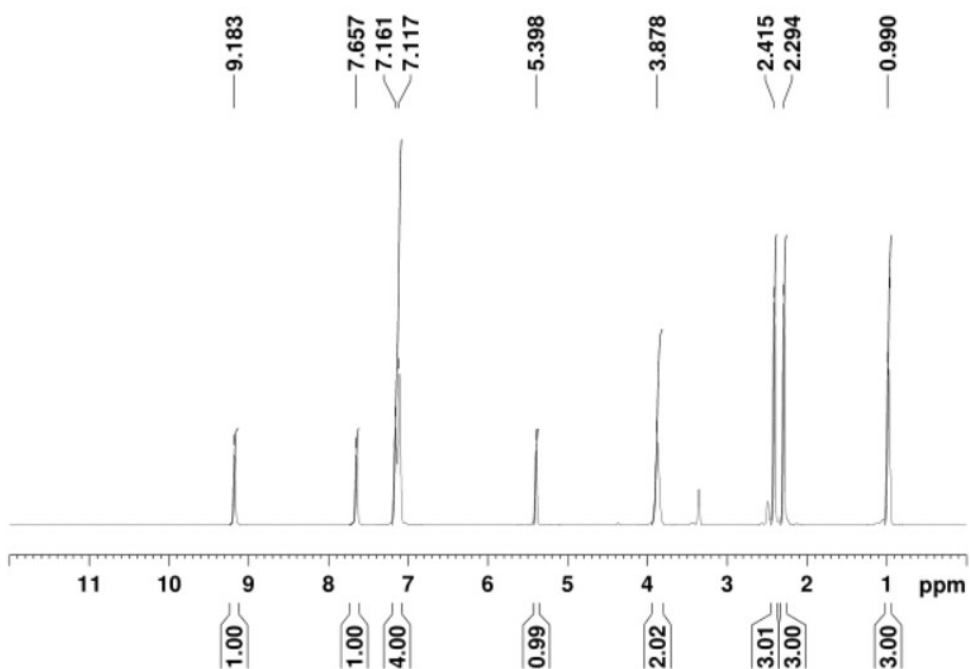
S-3b-¹H NMR:



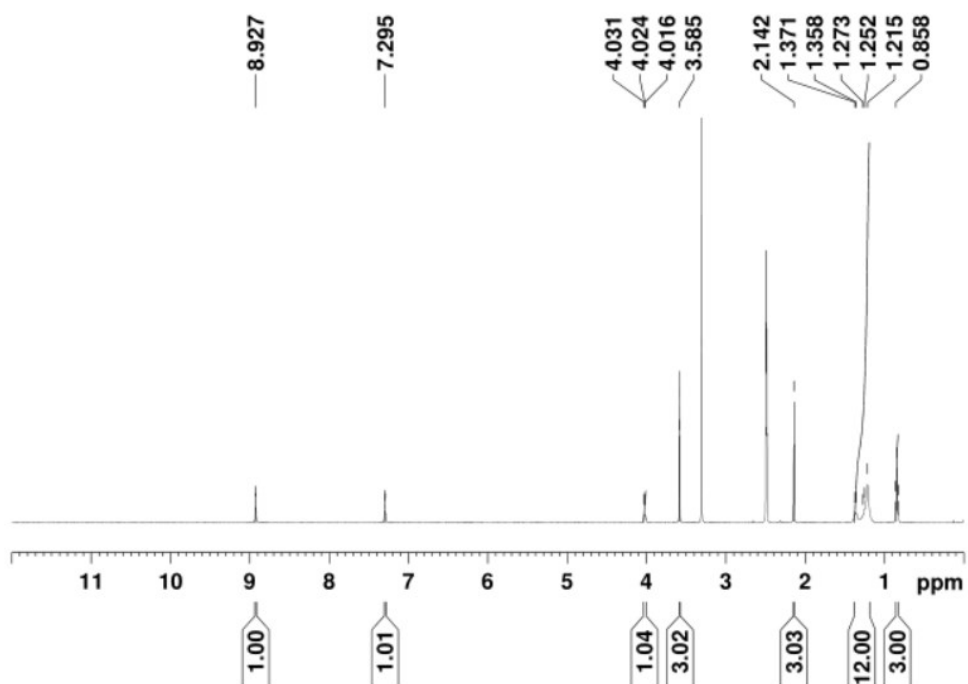
S-3c- ^1H NMR:



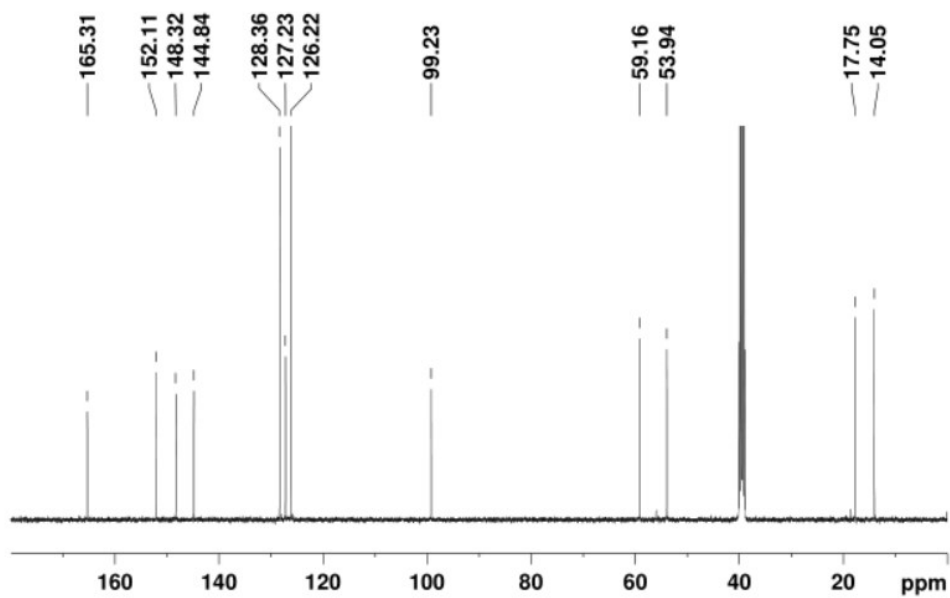
S-3d- ^1H NMR:



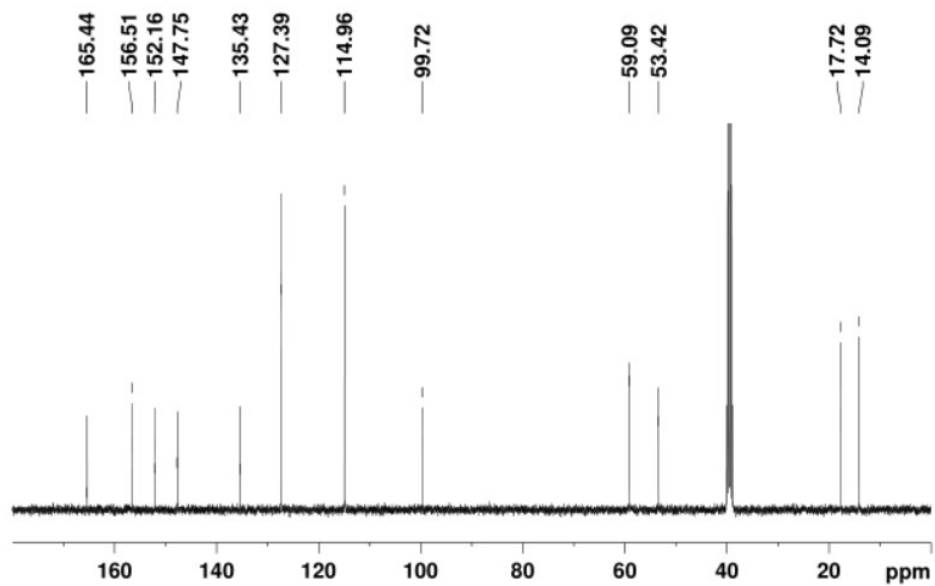
S-3e-¹H NMR:



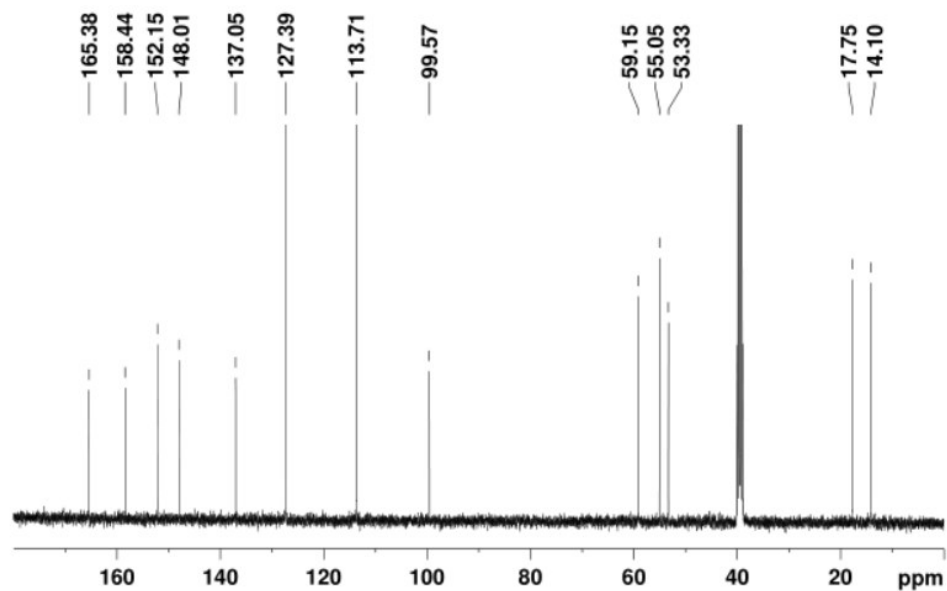
3a-¹³C NMR:



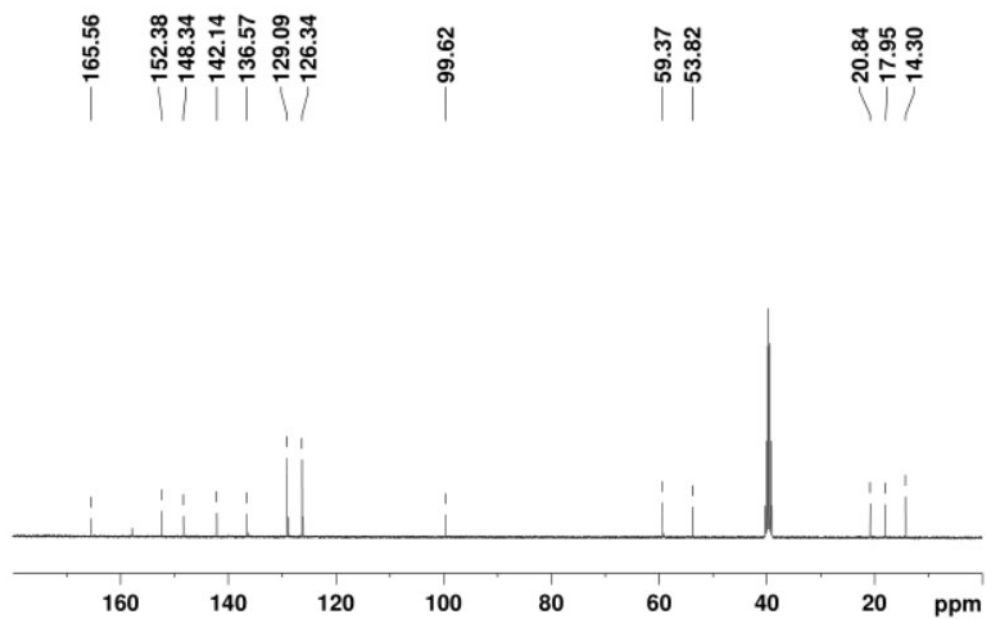
3b-¹³C NMR:



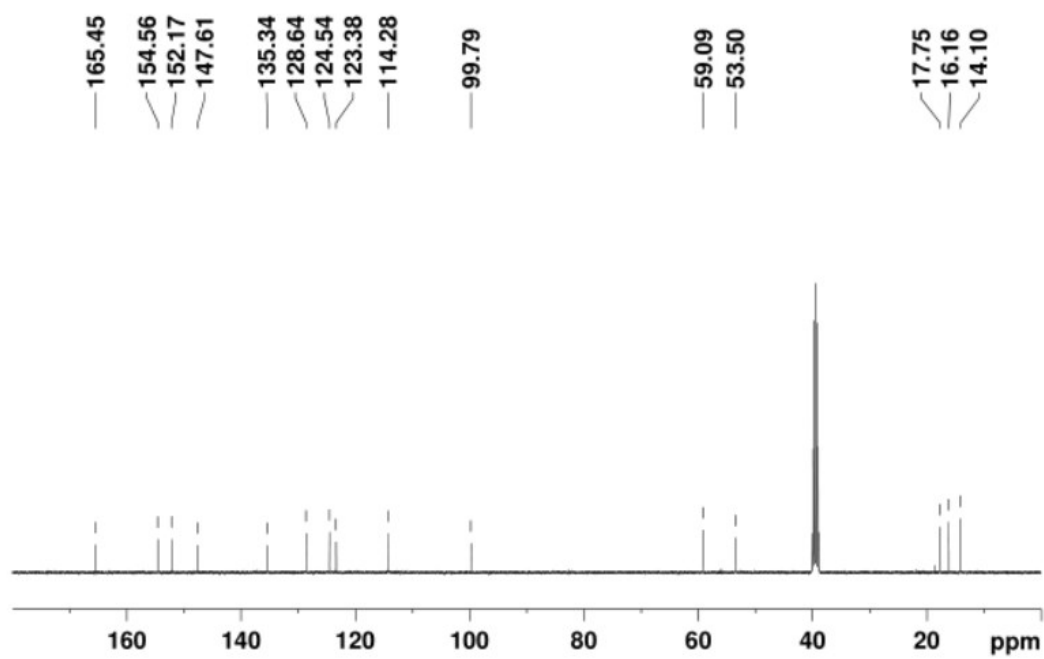
3c-¹³C NMR:



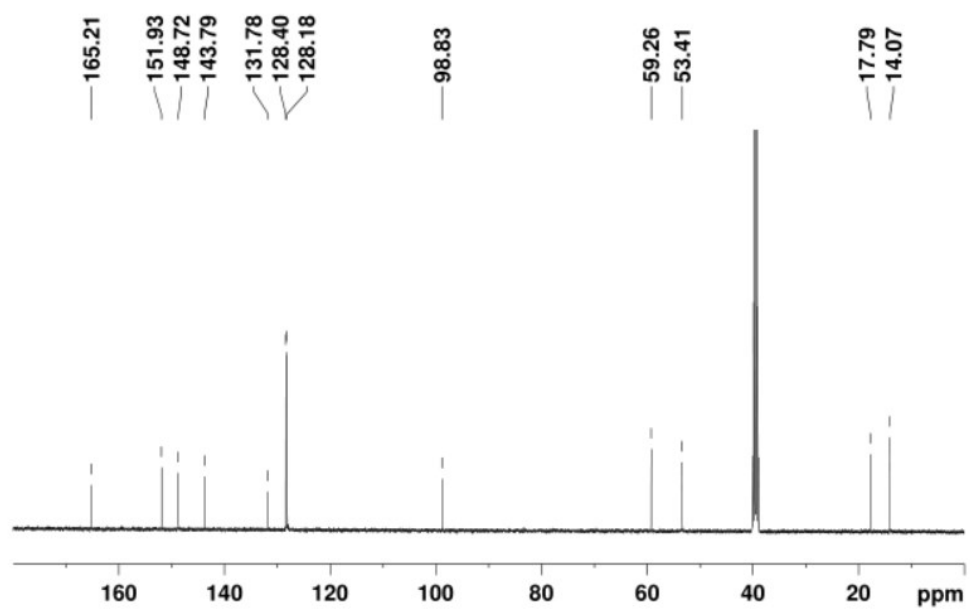
3d-¹³C NMR:



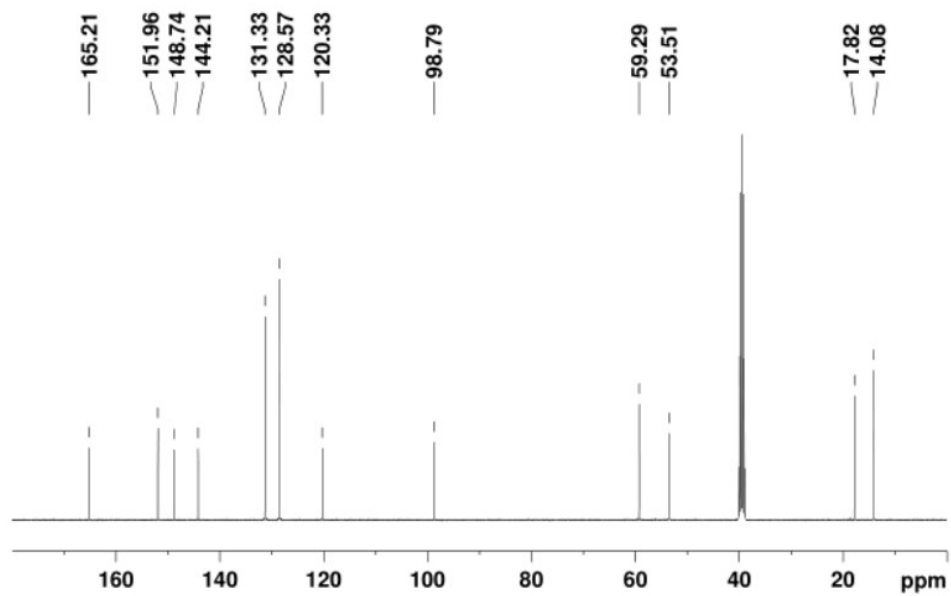
3e-¹³C NMR:



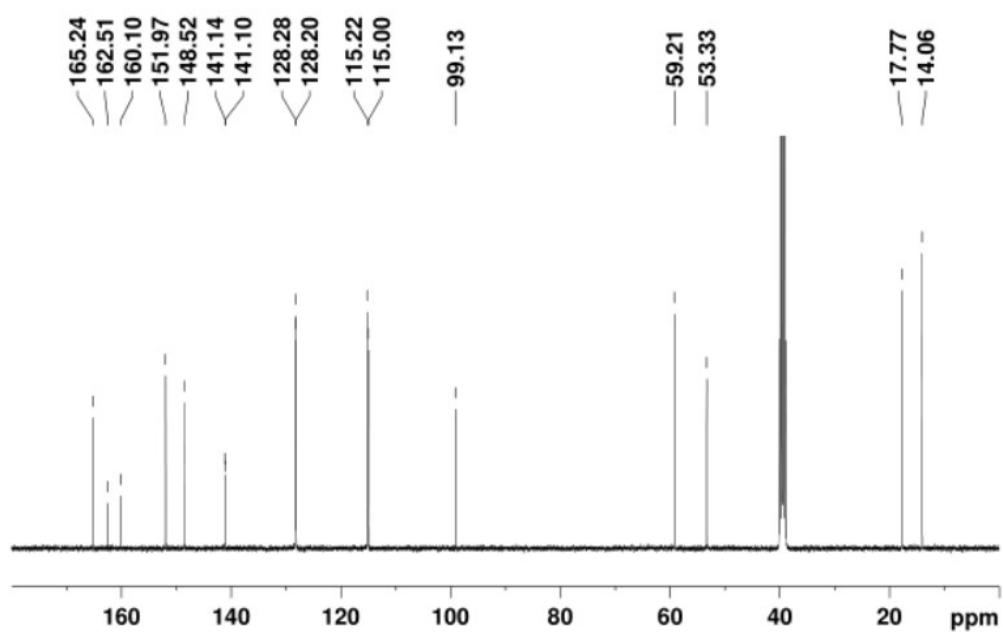
3f-¹³C NMR:



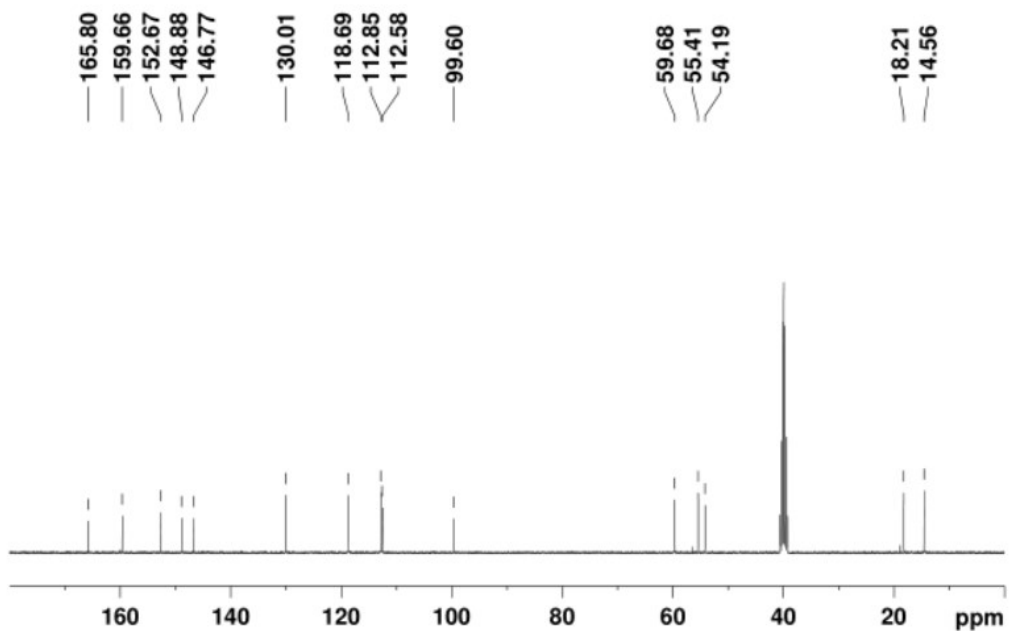
3g-¹³C NMR:



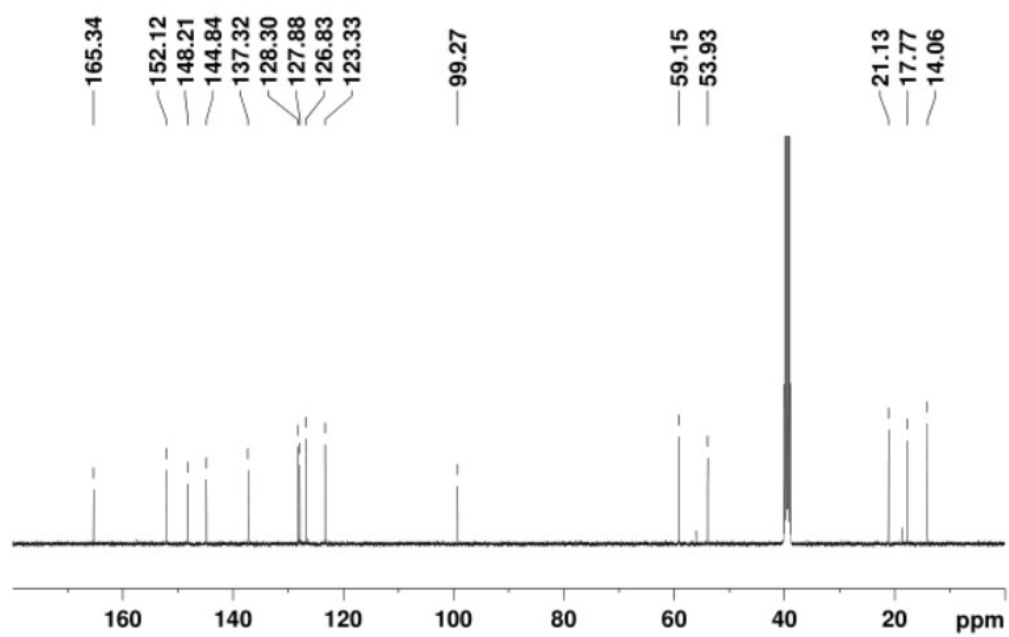
3h-¹³C NMR:



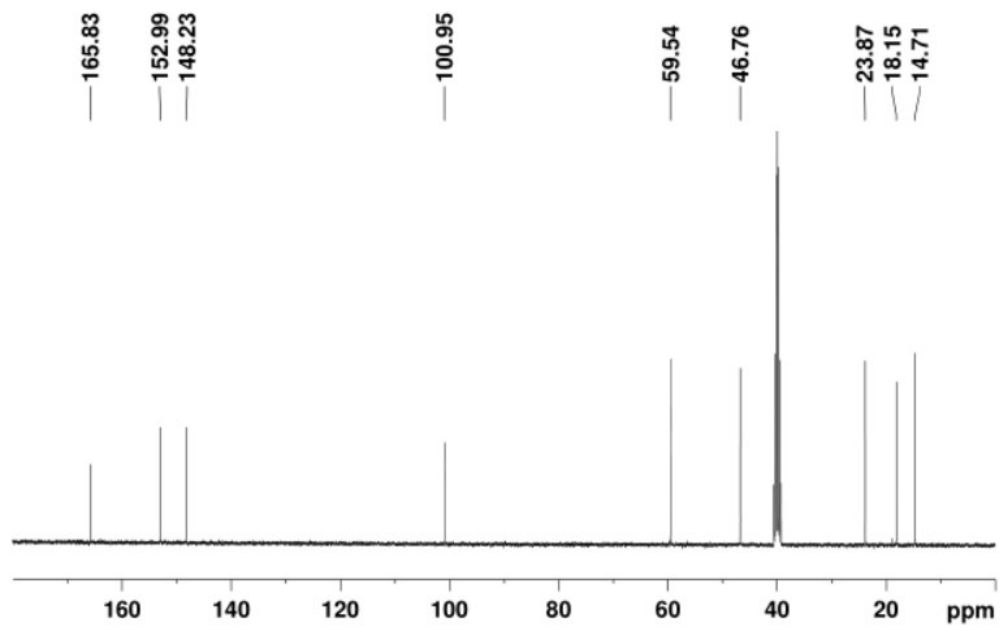
3i-¹³C NMR:



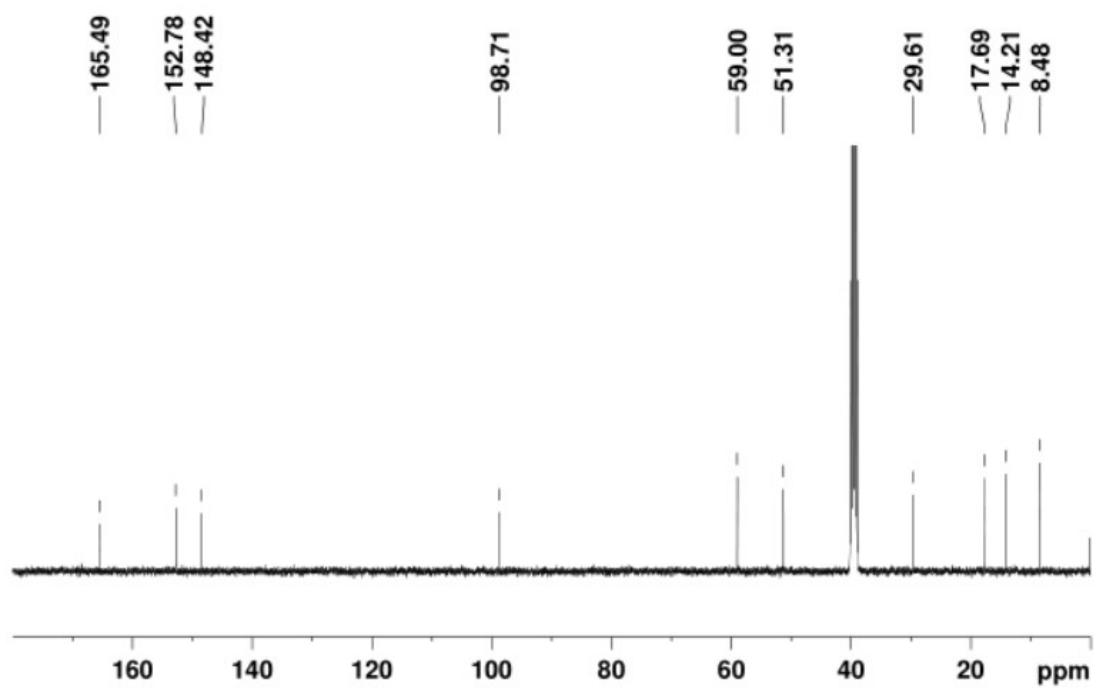
3j-¹³C NMR:



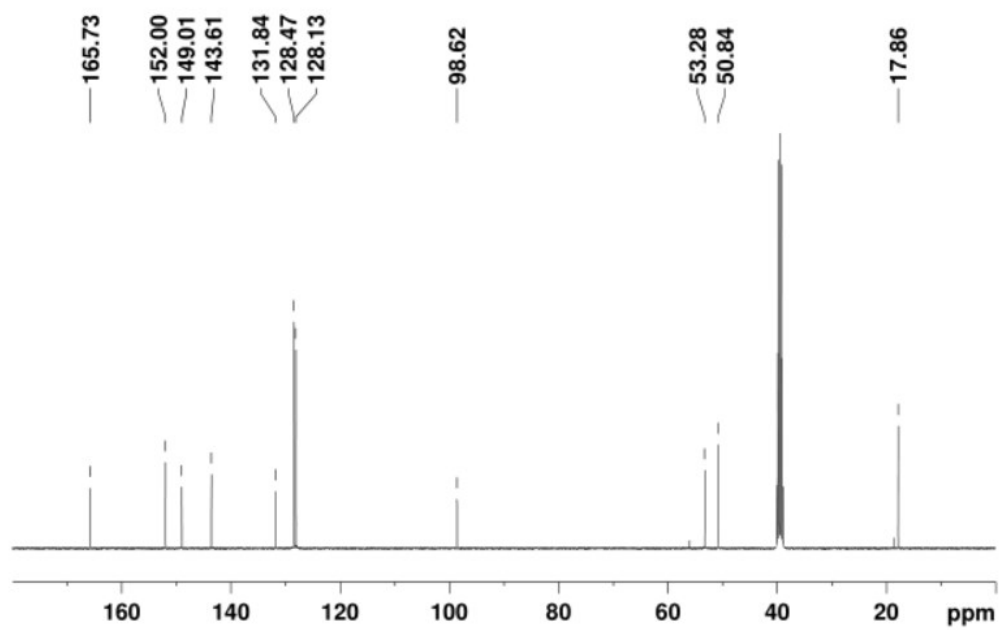
3k-¹³C NMR:



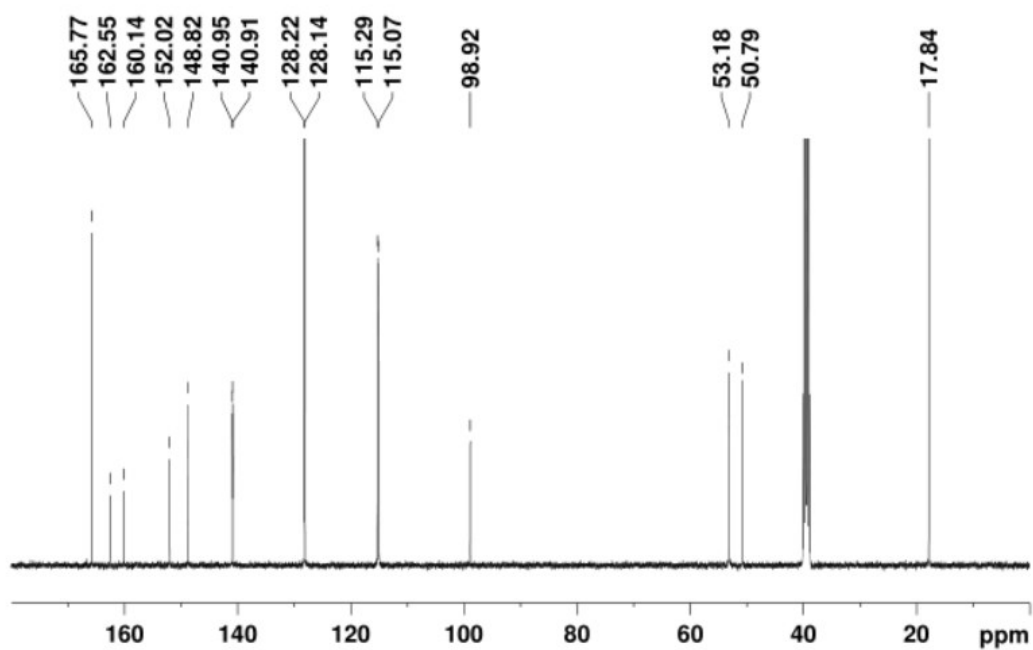
3l-¹³C NMR:



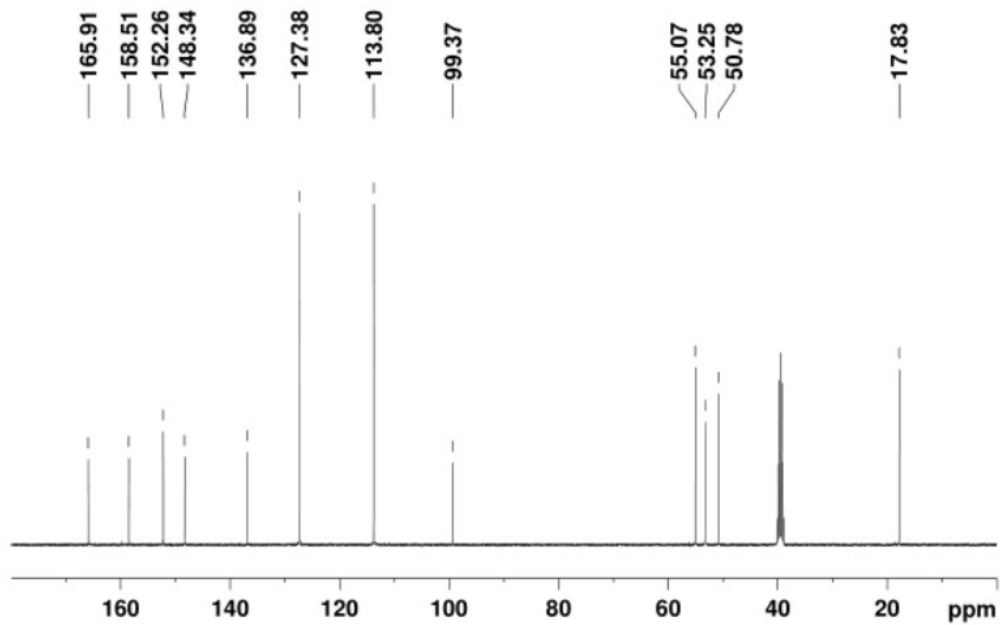
3m-¹³C NMR:



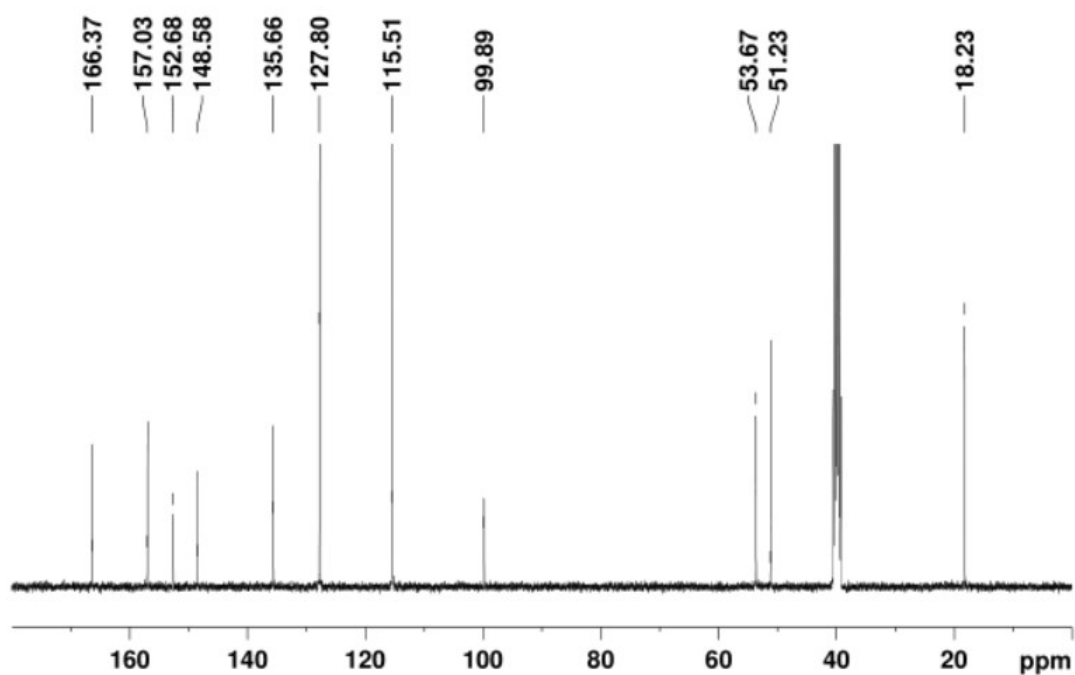
3n-¹³C NMR:



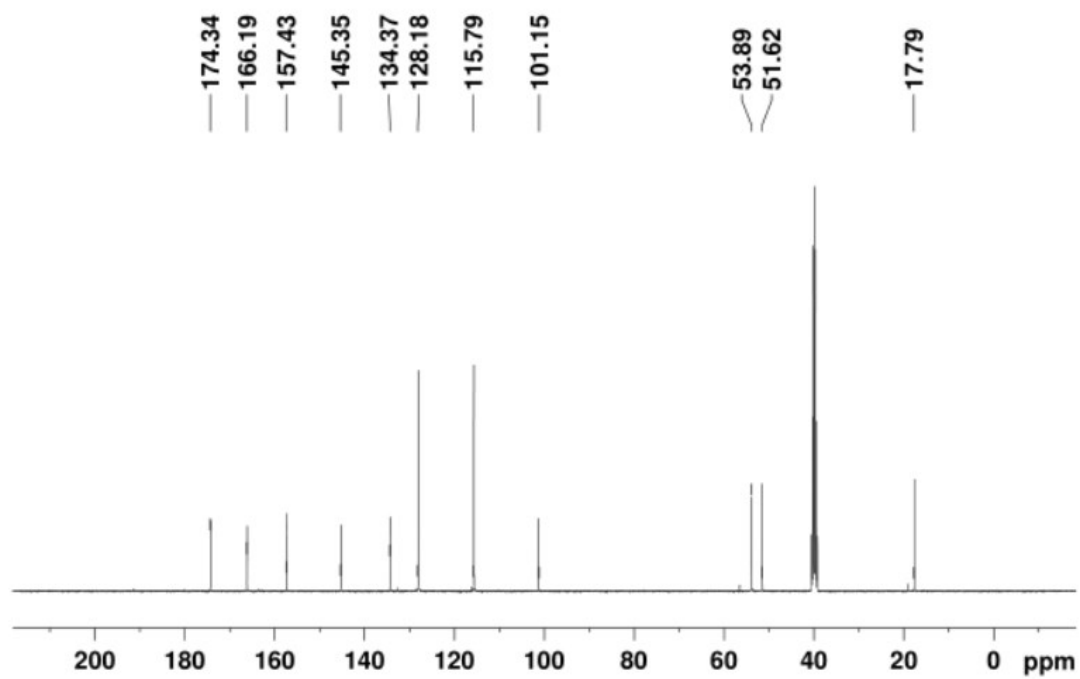
3o-¹³C NMR:



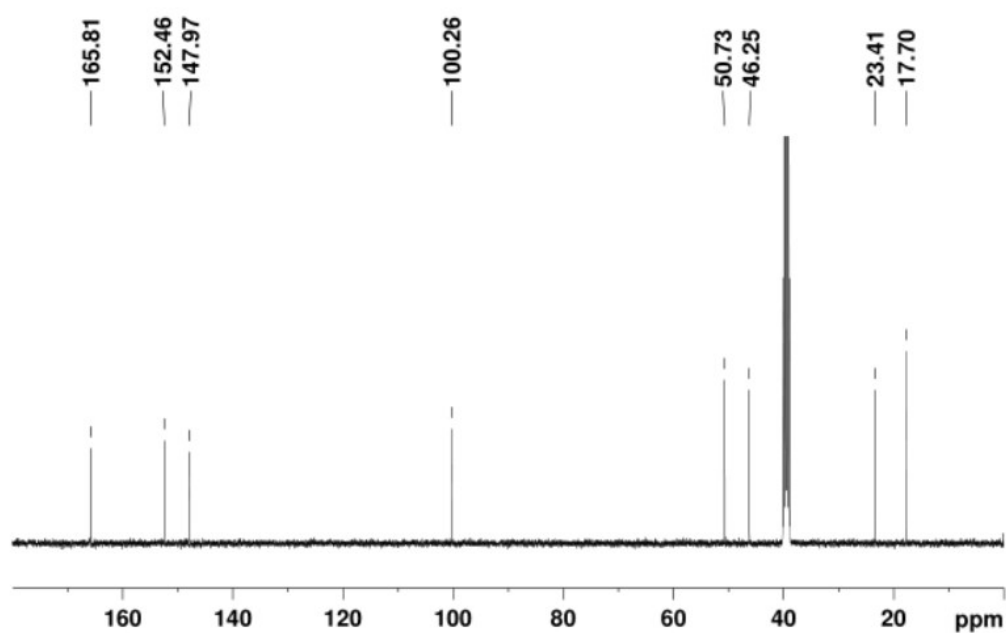
3p- ^{13}C NMR:



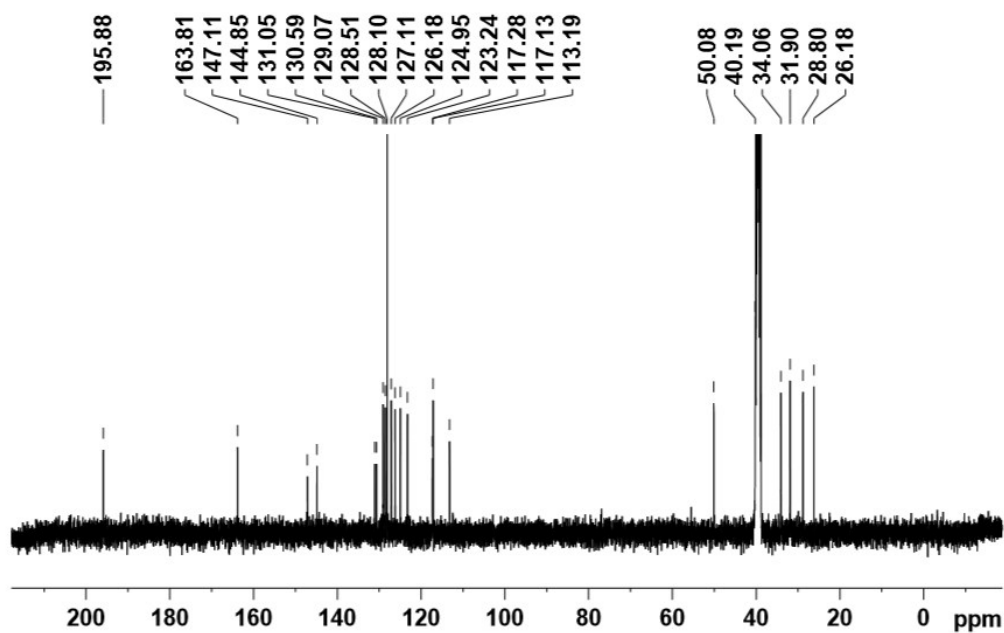
3q- ^{13}C NMR:



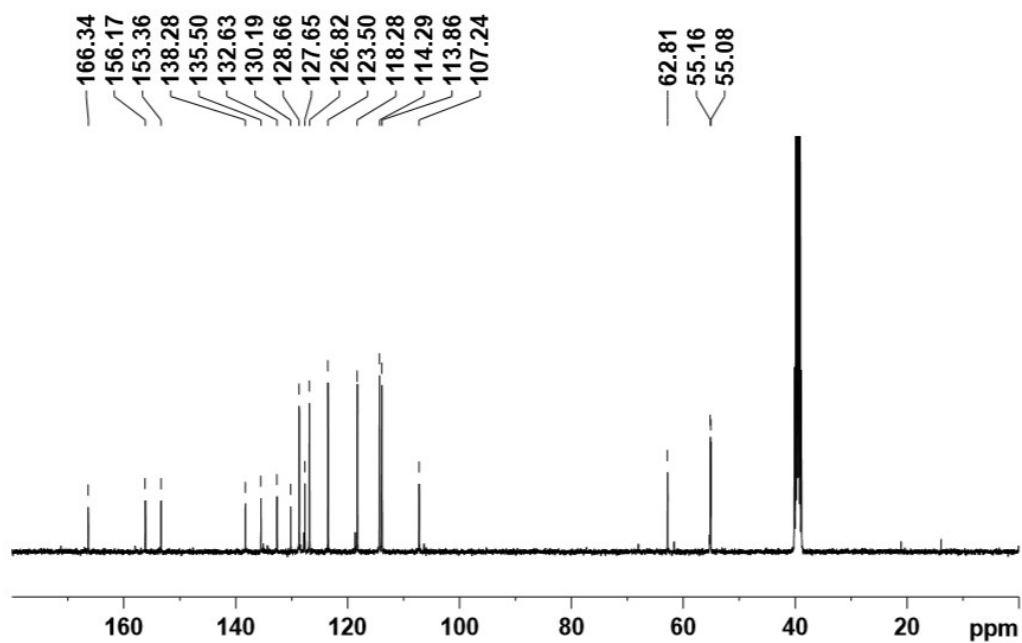
3r-¹³C NMR:



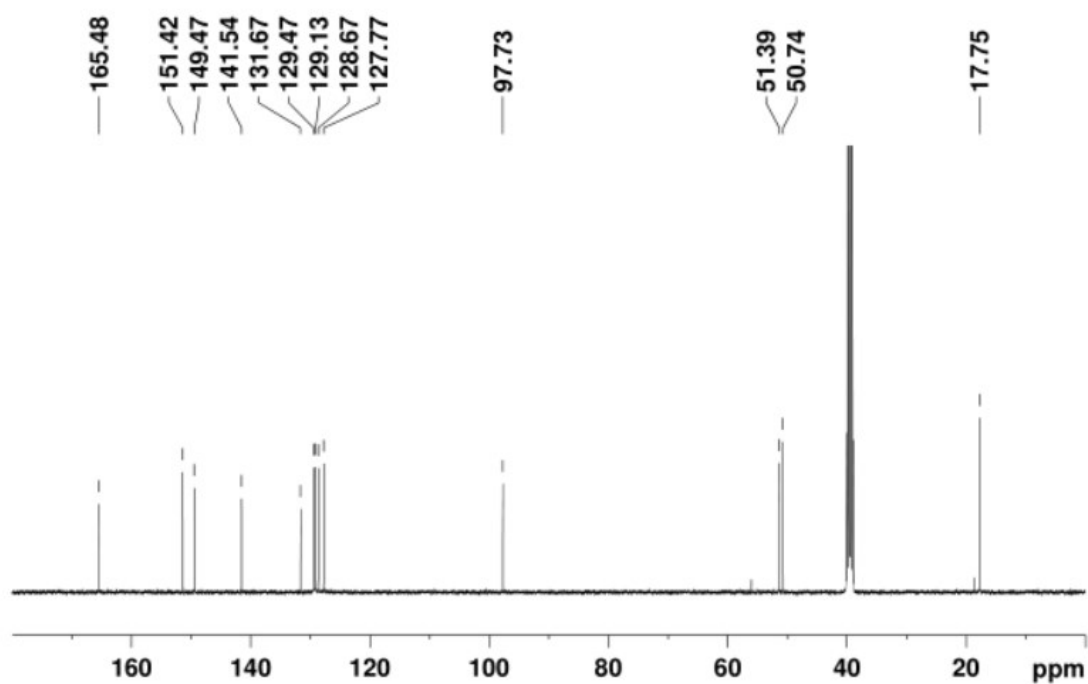
12-phenyl-9,9-dimethyl-8,9,10,12-tetrahydrobenzo[*a*]xanthen-11-one-¹³C NMR:



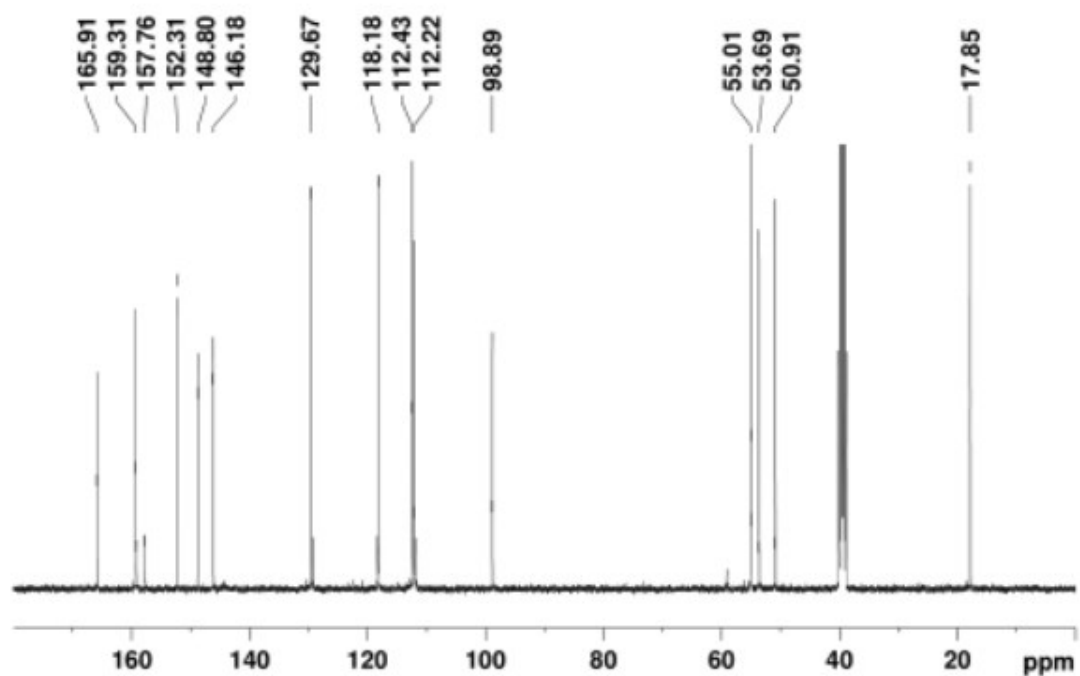
5-phenyl-1(4-methoxyphenyl)-3[(4-methoxyphenyl)-amino]-1*H*-pyrrol-2(5*H*)-
one-¹³C NMR:



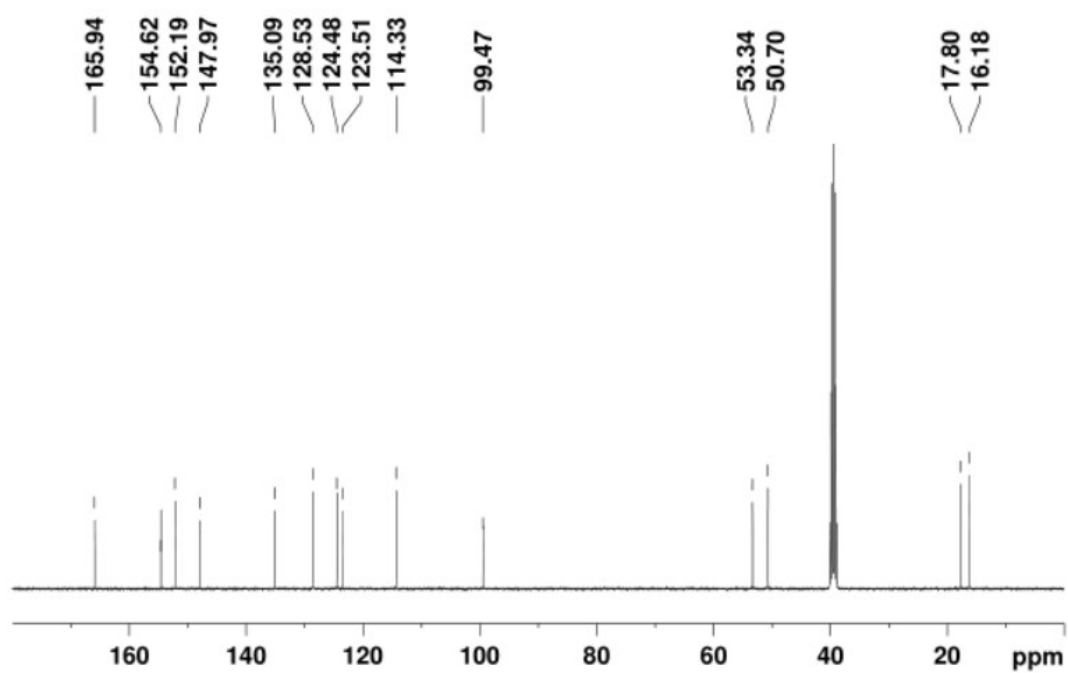
S-3a-¹³C NMR:



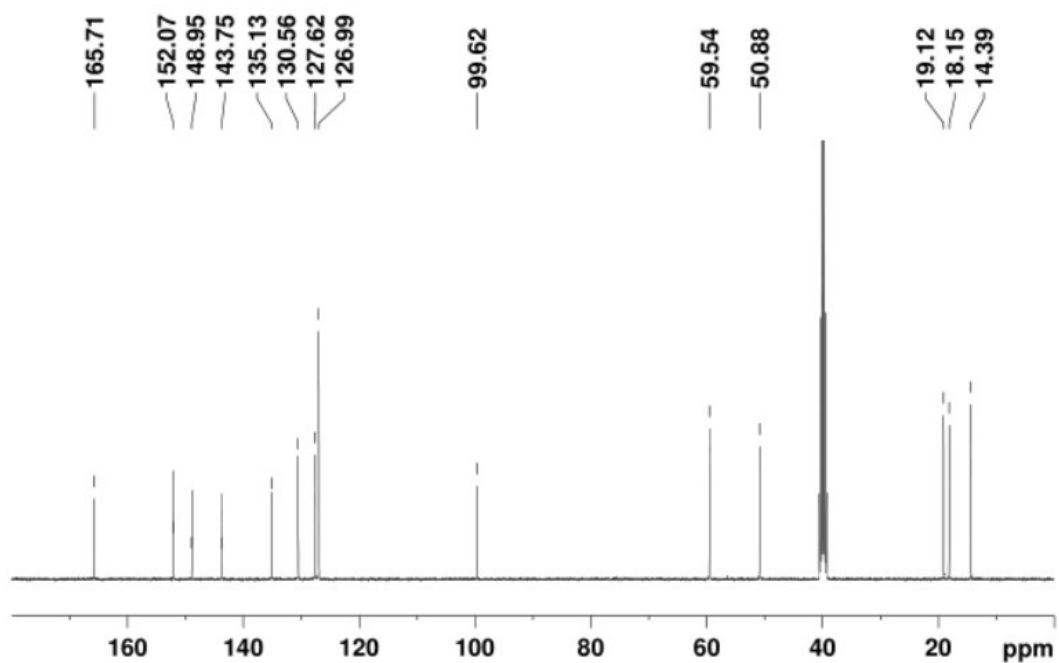
S-3b-¹³C NMR:



S-3c-¹³C NMR:



S-3d- ^{13}C NMR:



S-3e- ^{13}C NMR:

