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Supporting Informations

Molecular diversity from the three-component reaction of 2-hydroxy-1,4-naphthaquinone, aldehydes and 6-aminouracils: a reaction condition dependent MCR.

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Table of content **Page No.**

Experimental

Starting materials and solvents are commercially available and used without further purification. The purity of the synthesized compounds were ascertained by thin layer chromatography on silica gel GF 254 in ethyl acetate using iodine vapours as detecting agent. Melting points were determined by the melting point apparatus using capillary tube method and are uncorrected. IR spectra were recorded on a Shimadzu FTIR spectrophotometer. ^{1}H NMR and ^{13}C NMR spectra were recorded in $CDCl_3$ and DMSO-d_6 at ambient temperature on a Bruker Avance II 400 or 500 MHz spectrophotometer (100/125 MHz for ^{13}C). NMR chemical shifts are reported on the δ scale (ppm) downfield from tetramethyl silane (δ = 0.0 ppm). Data are reported as follows: chemical shift, multiplicity (s = singlet, d = doublet, t = triplet, q = quartet, m = multiplet, brs = broad singlet). HRMS analysis were carried out using BRUKER Impact HD mass spectrometer. Microwave irradiation was carried out with Initiator 2.5 Microwave Synthesizers from Biotage, Uppsala, Sweden.

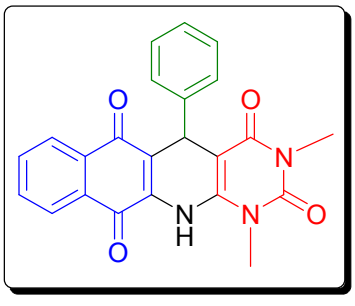
General procedures and Product Characterisation

General Procedure for the Synthesis of Fused Polycyclic *N*-heterocycles (4).

A mixture of 2-hydroxy-1,4-naphthaquinone **1** (1.0 mmol), 6-Aminouracil derivatives **2a-b** (1.0 mmol), and aldehydes **3a-o** (1.0 mmol) in 2 ml acetic acid/water (1:1) was introduced in a 2-5 mL reaction vial, the mixture was irradiated for 15 min, keeping temperature at 130 ^{o}C (Absorption Level: High; Fixed Hold Time and 200 W). The reaction mixture was then cooled to room temperature and the solid was filtered off, and was washed with water-ethanol or water-DMSO mixture to get the pure desired three component product **4**.

Product Charaterisation

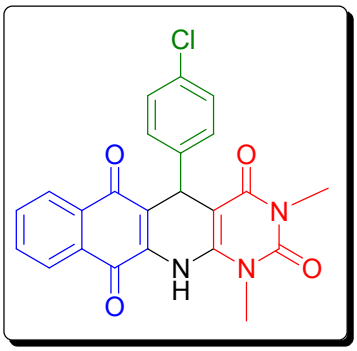
1,3-Dimethyl-5-phenylbenzo[*G*]pyrimido[4,5-*b*]quinoline-2,4,6,11(1*h*,3*h*,5*h*,12*h*)-tetrone (4aa)



Yield: 79%; Maroon solid; mp = 297-299 ^{o}C; IR (υ_{max}, KBr, cm^{-1}): 3365, 3073, 2941, 1702, 1681, 1657, 1635, 1608, 1589, 1492, 1435, 1296, 1149, 1037, 975, 867, 763; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 8.84 (s, 1H), 8.06 (d, *J* = 8.0 Hz, 1H), 7.94 (d, *J* = 8.0 Hz, 1H), 7.82 (t, *J* = 8.0 Hz, 2H), 7.34 (d, *J* = 8.0 Hz, 2H), 7.21 (t, *J* = 8.0 Hz, 2H), 7.12 (t, *J* = 8.0 Hz, 1H), 5.26 (s,

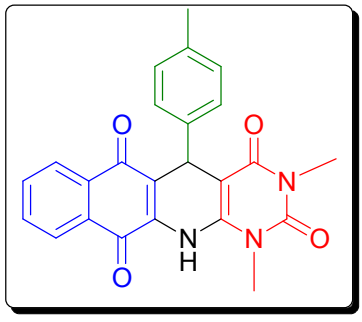
1H), 3.61 (s, 3H), 3.15 (s, 3H) ppm; ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 181.8, 178.7, 160.5, 150.5, 144.6, 143.5, 137.8, 135.1, 133.7, 131.6, 130.1, 128.2, 127.9, 126.7, 126.0, 125.9, 119.9, 89.6, 34.9, 29.7, 27.8 ppm; HRMS (ESI-TOF) calcd for $C_{23}H_{17}N_3O_4$ $[M + H]^+$ 400.1297, found 400.1278.

1,3-dimethyl-5-*p*-chlorophenyl-benzo[*G*]pyrimido[4,5-*b*]quinoline-2,4,6,11(1*h*,3*h*,5*h*, 12*h*)-tetrone (4ab)



Yield: 87%; Maroon solid; mp = 260-262 ^{o}C; IR (KBr, cm^{-1}): 3383, 2978, 1681, 1655, 1647, 1607, 1597, 1485, 1257, 1153, 1037, 926, 894, 729; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 8.85 (s, 1H), 8.06 (d, J = 8.0 Hz, 1H), 7.94 (d, J = 8.0 Hz, 1H), 7.84 (t, J = 8.0 Hz, 1H), 7.81 (t, J = 8.0 Hz, 1H), 7.35 (d, J = 8.0 Hz, 2H), 7.25 (d, J = 8.0 Hz, 2H), 5.24 (s, 1H), 3.59 (s, 3H), 3.14 (s, 3H) ppm; ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 185.8, 180.9, 163.6, 158.5, 154.4, 150.1, 137.6, 134.2, 133.4, 131.7, 130.6, 130.2, 128.7, 127.8, 126.0, 125.6, 123.2, 85.4, 34.4, 30.4, 28.2 ppm; HRMS (ESI-TOF) calcd for $C_{23}H_{16}ClN_3O_4$ $[M + H]^+$ 434.0907, found 434.0898.

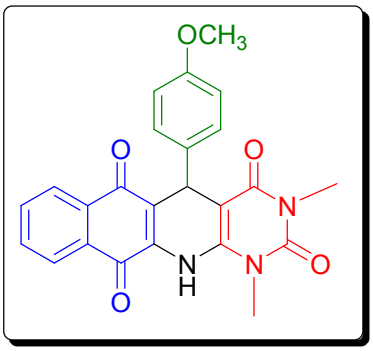
1,3-dimethyl-5-*p*-tolyl-benzo[*G*]pyrimido[4,5-*b*]quinoline-2,4,6,11(1*h*,3*h*,5*h*,12*h*)-tetrone (4ac)



Yield: 90%; Maroon solid; mp = 241-243 ^{o}C; IR (KBr, cm^{-1}): 3323, 3019, 1711, 1666, 1607, 1568, 1497, 1432, 1267, 1169, 1032, 967, 874, 767; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 8.82 (s, 1H), 8.07 (d, J = 8.0 Hz, 1H), 7.95 (d, J = 8.0 Hz, 1H), 7.84 (t, J = 8.0 Hz, 1H), 7.80 (d, J = 8.0 Hz,

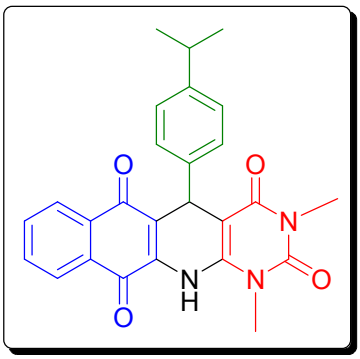
1H), 7.21 (d, *J* = 8.0 Hz, 2H), 7.01 (d, *J* = 8.0 Hz, 2H), 5.22 (s, 1H), 3.60 (s, 3H), 3.14 (s, 3H), 2.20 (s, 3H) ppm; ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 185.8, 181.0, 163.5, 158.5, 154.4, 150.1, 137.7, 134.3, 133.4, 131.7, 130.6, 130.2, 128.8, 127.9, 126.0, 125.7, 123.2, 85.4, 34.4, 30.4, 28.2, 20.4 ppm; HRMS (ESI-TOF) calcd for $C_{24}H_{19}N_3O_4$ $[M + H]^+$ 414.1453, found 414.1446.

1,3-dimethyl-5-*p*-methoxyphenyl-benzo[*G*]pyrimido[4,5-*b*]quinoline-2,4,6,11(1*h*,3*h*,5*h*,12 *h*)-tetrone (4ad)



Yield: 85%; Maroon solid; mp = 342-343 ^{o}C; IR (KBr, cm^{-1}): 3348, 2986, 1701, 1651, 1628, 1609, 1523, 1458, 1149, 1087, 821, 775; ^{1}H NMR (400 MHz, $CDCl_3$) δ_H: 8.06 (d, *J* = 8.0 Hz, 1H), 8.00 (d, *J* = 8.0 Hz, 1H), 7.71 (t, *J* = 8.0 Hz, 1H), 7.66 (t, *J* = 8.0 Hz, 1H), 7.51 (s, 1H), 7.31 (d, *J* = 8.0 Hz, 2H), 6.76 (d, *J* = 8.0 Hz, 2H), 5.35 (s, 1H), 3.70 (s, 3H), 3.64 (s, 3H), 3.28 (s, 3H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ_C: 181.8, 179.4, 161.0, 158.6, 150.8, 142.2, 136.5, 135.9, 135.1, 133.2, 132.3, 129.8, 129.2, 126.7, 126.2, 121.1, 113.8, 90.7, 55.0, 40.9, 35.0, 28.9 ppm; HRMS (ESI-TOF) calcd for $C_{24}H_{19}N_3O_5$ $[M + H]^+$ 430.1402, found 430.1411.

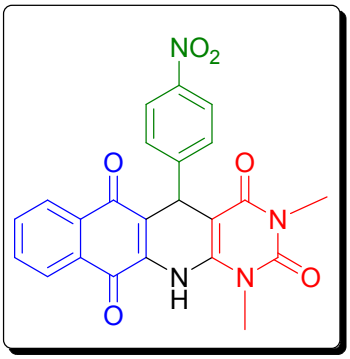
1,3-dimethyl-5-*p*-isopropylphenyl-benzo[*G*]pyrimido[4,5-*b*]quinoline-2,4,6,11(1*h*,3*h*,5*h*, *12 h*)-tetrone (4ae)



Yield: 82%; Caramine red solid; mp = 322-323 ^{o}C; IR (KBr, cm^{-1}): 3287, 3005, 2958, 1712, 1651, 1609, 1530, 1485, 1439, 1296, 1153, 1035, 968, 837, 752; ^{1}H NMR (400 MHz, DMSO-

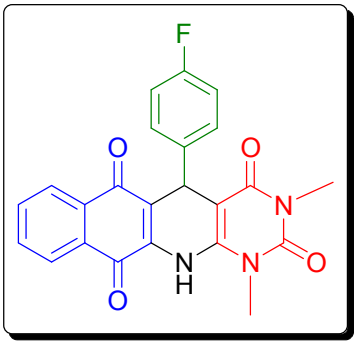
d$_6$) δ_H: 8.89 (s, 1H), 8.07 (d, *J* = 8.0 Hz, 1H), 7.95 (d, *J* = 8.0 Hz, 1H), 7.87 (t, *J* = 8.0 Hz, 1H), 7.85 (t, *J* = 8.0 Hz, 1H), 7.23 (d, *J* = 8.0 Hz, 2H), 7.09 (d, *J* = 8.0 Hz, 2H), 5.22 (s, 1H), 3.59 (s, 3H), 3.13 (s, 3H), 2.63-2.82 (m, 1H), 1.12 (d, *J* = 8.0 Hz, 6H) ppm; ^{13}C NMR (100 MHz, DMSO-*d*$_6$) δ_C: 181.8, 179.6, 161.1, 150.9, 147.7, 142.2, 141.4, 136.1, 135.2, 133.2, 132.4, 129.9, 128.0, 126.9, 126.7, 126.3, 121.4, 90.8, 35.4, 33.6, 28.9, 28.3, 23.9 ppm; HRMS (ESI-TOF) calcd for $C_{26}H_{23}N_3O_4$ $[M + H]^+$ 442.1766, found 442.1758.

1,3-dimethyl-5-*p*-nitrophenyl-benzo[*G*]pyrimido[4,5-*b*]quinoline-2,4,6,11(1*h*,3*h*,5*h*,12*h*)-tetrone (4af)



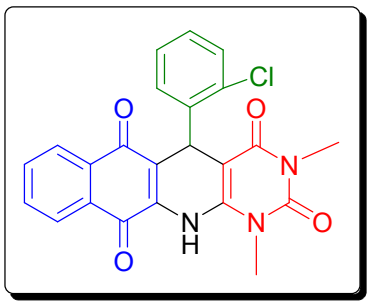
Yield: 64%; Brown solid; mp = 311-312 ^oC; IR (KBr, cm^{-1}): 3391, 3067, 2958, 1712, 1681, 1667, 1631, 1607, 1558, 1467, 1350, 1296, 1161, 1046, 883, 752; ^{1}H NMR (400 MHz, DMSO-*d*$_6$) δ_H: 8.89 (s, 1H), 8.09-8.07 (m, 3H), 7.94 (d, *J* = 8.0 Hz, 1H), 7.83-7.80 (m, 2H), 7.64 (d, *J* = 8.0 Hz, 2H), 5.39 (s, 1H), 3.62 (s, 3H), 3.14 (s, 3H) ppm; ^{13}C NMR (100 MHz, DMSO-*d*$_6$) δ_C: 181.5, 178.5, 160.5, 151.6, 150.4, 146.2, 143.8, 138.2, 134.9, 133.6, 131.5, 130.1, 129.4, 126.0, 125.9, 123.2, 118.6, 88.8, 35.6, 29.8, 27.7 ppm; HRMS (ESI-TOF) calcd for $C_{23}H_{16}N_4O_6$ $[M + H]^+$ 445.1148, found 445.1139.

1,3-dimethyl-5-*p*-fluorophenyl-benzo[*G*]pyrimido[4,5-b]quinoline-2,4,6,11(1*h*,3*h*,5*h*, 12*h*)-tetrone (4ag)



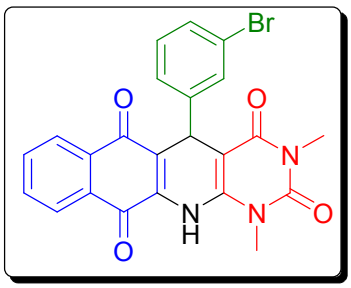
Yield: 71%; Maroon solid; mp = 321-322 ^{o}C; IR (KBr, cm^{-1}): 3344, 3070, 2962, 1693, 1654, 1616, 1496, 1296, 1149, 1045, 972, 898, 756; ^{1}H NMR (400 MHz, $CDCl_3$) δ_H: 8.11 (d, *J* = 8.0 Hz, 1H), 8.05 (d, *J* = 8.0 Hz, 1H), 7.76 (t, *J* = 8.0 Hz, 1H), 7.73 (t, *J* = 8.0 Hz, 1H), 7.53 (s, 1H), 7.39 (t, *J* = 8.0 Hz, 2H), 6.94 (t, *J* = 8.0 Hz, 2H), 5.47 (s, 1H), 3.66 (s, 3H), 3.30 (s, 3H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ_C: 181.0, 170.3, 163.6, 161.7, 159.3, 158.5, 154.4, 150.1, 134.3, 133.4, 131.7, 130.6, 128.7, 126.0, 125.6, 123.5, 114.7, 85.6, 34.2, 30.4, 28.1 ppm; HRMS (ESI-TOF) calcd for $C_{23}H_{16}FN_3O_4$ $[M + H]^+$ 418.1203, found 418.1212.

1,3-dimethyl-5-*o*-chlorophenyl-benzo[*G*]pyrimido[4,5-*b*]quinoline-2,4,6,11(1*h*,3*h*,5*h*, 12*h*)-tetrone (4ah)



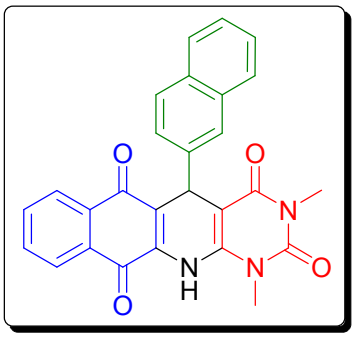
Yield: 90%; Red solid; mp = 321-323 ^{o}C; IR (KBr, cm^{-1}): 3379, 3074, 2951, 1701, 1658, 1635, 1609, 1589, 1493, 1435, 1261, 1180, 1037, 975, 867, 763; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 8.79 (s, 1H), 8.07 (d, *J* = 8.0 Hz, 1H), 7.91 (d, *J* = 8.0 Hz, 1H), 7.80 (t, *J* = 8.0 Hz, 2H), 7.45 (d, *J* = 8.0 Hz, 1H), 7.25 (d, *J* = 8.0 Hz, 1H), 7.18 (t, *J* = 8.0 Hz, 1H), 7.14 (t, *J* = 8.0 Hz, 1H), 5.62 (s, 1H), 3.63 (s, 3H), 3.12 (s, 3H) ppm; ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 185.8, 181.0, 163.6, 161.7, 159.3, 158.5, 154.4, 150.1, 134.4, 134.3, 134.2, 133.4, 131.7, 130.6, 128.6, 126.0, 125.6, 123.5, 114.5, 85.6, 34.2, 30.4, 28.1 ppm; HRMS (ESI-TOF) calcd for $C_{23}H_{16}ClN_3O_4$ $[M + H]^+$ 434.0907, found 434.0912.

1,3-dimethyl-5-*m*-bromophenyl-benzo[*G*]pyrimido[4,5-*b*]quinoline-2,4,6,11(1*h*,3*h*,5*h*, 12*h*) -tetrone (4ai)



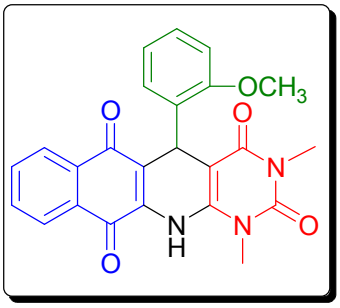
Yield: 78%; Caramine red solid; mp = 273-275 ^{o}C; IR (KBr, cm^{-1}): 3363, 2954, 1685, 1654, 1639, 1601, 1589, 1473, 1273, 1072, 972, 748; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 8.86 (s, 1H), 8.08 (d, J = 8.0 Hz, 1H), 7.96 (d, J = 8.0 Hz, 1H), 7.82 (t, J = 8.0 Hz, 2H), 7.51-7.50 (m, 1H), 7.34 (d, J = 8.0 Hz, 1H), 7.30 (d, J = 8.0 Hz, 1H), 7.17 (t, J = 8.0 Hz, 1H), 5.24 (s, 1H), 3.61 (s, 3H), 3.16 (s, 3H) ppm; ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 181.6, 178.7, 160.5, 150.4, 147.0, 143.6, 137.9, 134.9, 133.5, 131.6, 130.7, 130.2, 130.1, 129.5, 127.1, 126.0, 125.9, 121.5, 119.1, 89.2, 35.2, 29.7, 27.7 ppm; HRMS (ESI-TOF) calcd for $C_{23}H_{16}BrN_3O_4$ $[M + H]^+$ 478.0402, found 478.0423.

1,3-dimethyl-5-(2-naphthyl)benzo[*G*]pyrimido[4,5-*b*]quinoline-2,4,6,11(1*h*,3*h*,5*h*,12*h*)-tetr one (4aj)



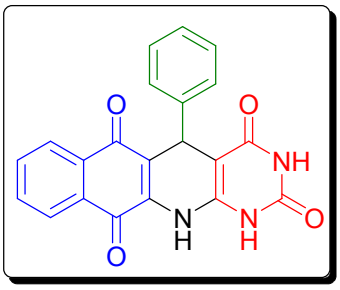
Yield: 85%; Caramine red solid; mp = 279-281^{o}C; IR (KBr, cm^{-1}): 3383, 3055, 1693, 1654, 1616, 1496, 1296, 1045, 972, 860, 748; ^{1}H NMR (400 MHz, $CDCl_3$) δ_H: 8.89 (s, 1H), 8.08 (d, J = 8.0 Hz, 1H), 7.93 (d, J = 8.0 Hz, 1H), 7.83-7.78 (m, 4H), 7.75 (t, J = 8.0 Hz, 2H), 7.56 (d, J = 8.0 Hz, 1H), 7.41 (t, J = 8.0 Hz, 2H), 5.44 (s, 1H), 3.65 (s, 3H), 3.14 (s, 3H) ppm; ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 186.1, 181.0, 170.1, 163.8, 158.8, 154.5, 150.2, 135.9, 134.1, 133.3, 133.0, 131.8, 131.5, 130.6, 127.5, 127.4, 127.1, 126.1, 125.7, 125.6, 125.2, 124.3, 123.2, 85.9, 34.9, 30.4, 28.1 ppm; HRMS (ESI-TOF) calcd for $C_{27}H_{19}N_3O_4$ $[M + H]^+$ 450.1453, found 450.1471.

1,3-dimethyl-5-*o*-methoxyphenyl-benzo[*G*]pyrimido[4,5-*b*]quinoline-2,4,6,11(1*h*,3*h*,5*h*,12 *h*)-tetrone (4ak)



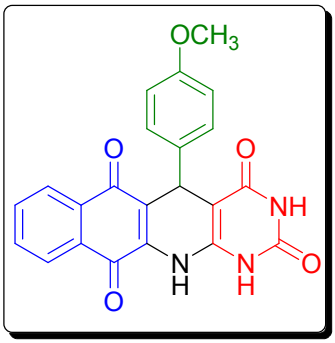
Yield: 92%; Maroon solid; mp = 313-315 ^{o}C; IR (KBr, cm^{-1}): 3352, 3162, 3041, 2958, 1693, 1655, 1618, 1500, 1438, 1296, 1049, 972, 844, 798; ^{1}H NMR (500 MHz, DMSO-d$_6$) δ_H: 8.98 (s, 1H), 8.06 (d, *J* = 5.0 Hz, 1H), 7.89 (d, *J* = 10.0 Hz, 1H), 7.83 (t, *J* = 10.0 Hz, 2H), 7.36 (d, *J* = 10.0 Hz, 1H), 7.12 (t, *J* = 10.0 Hz, 1H), 6.84 (t, *J* = 10.0 Hz, 2H), 5.32 (s, 1H), 3.70 (s, 3H), 3.60 (s, 3H), 3.09 (s, 3H) ppm; ^{13}C NMR (125 MHz, DMSO-*d*$_6$) δ_C: 182.3, 179.5, 165.2, 160.9, 158.8, 155.9, 151.1, 144.5, 138.9, 135.6, 134.0, 132.1, 131.9, 128.6, 126.4, 126.3, 120.2, 119.2, 112.4, 88.9, 55.9, 34.3, 30.2, 28.1 ppm; HRMS (ESI-TOF) calcd for $C_{24}H_{19}N_3O_5$ $[M + H]^+$ 430.1402, found 430.1421 .

5-phenylbenzo[*G*]pyrimido[4,5-*b*]quinoline-2,4,6,11(1*h*,3*h*,5*h*,12*h*)-tetrone (4ba)



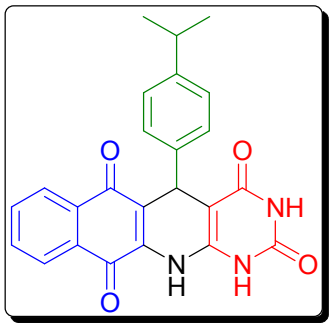
Yield: 89%; Red solid; mp = 299-300 ^{o}C; IR (KBr, cm^{-1}): 3401, 3347, 3257, 3004, 1728, 1661, 1604, 1587, 1426, 1286, 1157, 1016, 972, 861, 748; ^{1}H NMR (400 MHz, DMSO-*d*$_6$) δ_H: 10.92 (s, 1H), 10.15 (s, 1H), 9.43 (s, 1H), 8.05 (d, *J* = 8.0 Hz, 1H), 7.92 (d, *J* = 8.0 Hz, 1H), 7.85 (t, *J* = 8.0 Hz, 1H), 7.81 (t, *J* = 8.0 Hz, 1H), 7.33 (d, *J* = 8.0 Hz, 2H), 7.22 (t, *J* = 8.0 Hz, 2H), 7.13 (t, *J* = 8.0 Hz, 1H), 5.11 (s, 1H) ppm; ^{13}C NMR (100 MHz, DMSO-*d*$_6$) δ_C: 181.8, 179.0, 162.6, 149.4, 145.0, 143.7, 137.9, 135.0 133.5, 131.8, 130.1, 128.1, 128.0, 126.5, 125.9, 125.8, 118.5, 88.5, 34.3 ppm; HRMS (ESI-TOF) calcd for $C_{21}H_{13}N_3O_4$ $[M + H]^+$ 372.0984, found 372.1004.

5-*p*-methoxyphenylbenzo[*G*]pyrimido[4,5-*b*]quinoline-2,4,6,11(1*h*,3*h*,5*h*,12*h*)-tetrone (4bd)



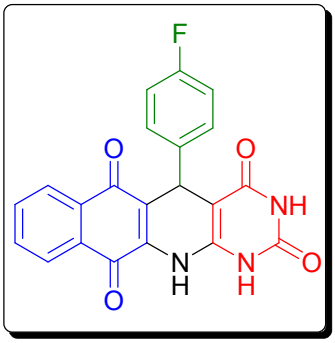
Yield: 91%; Caramine red solid; mp = 309-310 ^{o}C; IR (KBr, cm^{-1}): 3417, 3328, 3159, 2978, 1716, 1651, 1608, 1543, 1435, 1296, 1165, 1014, 937, 860, 759; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 10.92 (brs, 1H), 9.78 (brs, 2H), 8.04 (d, J = 8.0 Hz, 1H), 7.92 (d, J = 8.0 Hz, 1H), 7.83 (t, J = 8.0 Hz, 1H), 7.81 (t, J = 8.0 Hz, 1H), 7.23 (d, J = 8.0 Hz, 2H), 6.77 (d, J = 8.0 Hz, 2H), 5.07 (s, 1H), 3.67 (s, 3H) ppm; ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 181.6, 178.6, 160.5, 150.4, 149.6, 143.8, 138.2, 134.9, 133.6, 132.0, 131.5, 130.1, 129.1, 126.0, 125.9, 118.7, 118.6, 109.5, 88.8, 54.9, 35.7 ppm; HRMS (ESI-TOF) calcd for $C_{22}H_{15}N_3O_5$ $[M + H]^+$ 402.1089, found 402.1078.

5-*p*-isopropylphenylbenzo[*G*]pyrimido[4,5-*b*]quinoline-2,4,6,11(1*h*,3*h*,5*h*,12*h*)-tetrone (4be)



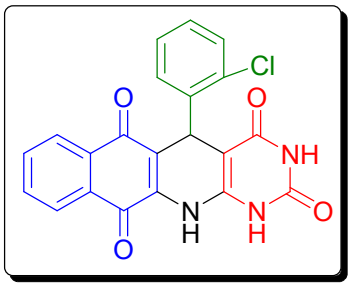
Yield: 87%; Orange solid; mp = 317-320 ^{o}C; IR (KBr, cm^{-1}): 3159, 3055, 2958, 1716, 1651, 1612, 1589, 1508, 1435, 1296, 1149, 1014, 933, 837, 752; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 10.88 (s, 1H), 10.02 (s, 1H), 9.58 (s, 1H), 8.05 (d, J = 8.0 Hz, 1H), 7.94 (d, J = 8.0 Hz, 1H), 7.80 (t, J = 8.0 Hz, 2H), 7.24 (d, J = 8.0 Hz, 2H), 7.07 (d, J = 8.0 Hz, 2H), 5.12 (s, 1H), 2.81-2.78 (m, 1H), 1.12 (d, J = 8.0 Hz, 6H) ppm; ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 185.9, 181.1, 163.6, 158.6, 154.4, 150.1, 145.5, 135.5, 134.3, 133.4, 131.7, 130.5, 126.6, 126.0, 125.9, 125.7, 123.7, 85.7, 34.4, 32.9, 23.9 ppm; HRMS (ESI-TOF) calcd for $C_{24}H_{19}N_3O_4$ $[M + H]^+$ 414.1453, found 414. 1468.

5-*p*-fluorophenylbenzo[*G*]pyrimido[4,5-*b*]quinoline-2,4,6,11(1*h*,3*h*,5*h*,12*h*)-tetrone (4bg)



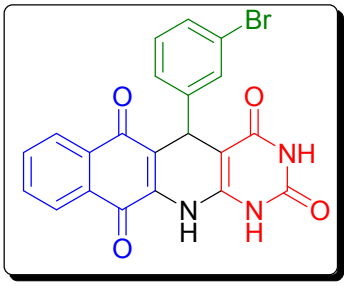
Yield: 89%; Orange solid; mp = 309-310 ^{o}C; IR (KBr, cm^{-1}): 3342, 3271, 3174, 3008, 1739, 1647, 1597, 1438, 1296, 1153, 1014, 929, 867, 752; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 10.86 (s, 1H), 10.21 (s, 1H), 9.69 (s, 1H), 8.06 (d, *J* = 8.0 Hz, 1H), 7.95 (d, *J* = 8.0 Hz, 1H), 7.81 (t, *J* = 8.0 Hz, 1H), 7.79 (t, *J* = 8.0 Hz, 1H), 7.37 (t, *J* = 8.0 Hz, 2H), 6.96 (t, *J* = 8.0 Hz, 2H), 5.18 (s, 1H) ppm; ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 181.7, 178.9, 162.6, 149.4, 143.6, 141.2, 137.9, 134.9, 133.4, 131.8, 130.1, 129.9, 125.9, 125.8, 118.1, 114.8, 114.6, 88.4, 33.8 ppm; HRMS (ESI-TOF) calcd for $C_{21}H_{12}FN_3O_4$ $[M + H]^+$ 390.0890, found 390.0913.

5-*o*-chlorophenylbenzo[*G*]pyrimido[4,5-*b*]quinoline-2,4,6,11(1*h*,3*h*,5*h*,12*h*)-tetrone (4bh)



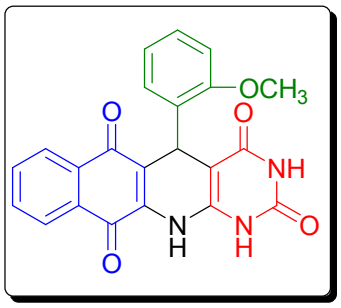
Yield: 87%; Caramine red solid; mp = 327-328 ^{o}C; IR (KBr, cm^{-1}): 3525, 3298, 3248, 3070, 2985, 1720, 1647, 1593, 1473, 1296, 1153, 1076, 937, 759; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 10.76 (s, 1H), 10.04 (s, 1H), 9.51 (s, 1H), 8.06 (d, *J* = 8.0 Hz, 1H), 7.91 (d, *J* = 8.0 Hz, 1H), 7.78 (t, *J* = 8.0 Hz, 1H), 7.75 (t, *J* = 8.0 Hz, 1H), 7.44 (d, *J* = 8.0 Hz, 1H), 7.25 (d, *J* = 8.0 Hz, 1H), 7.18 (t, *J* = 8.0 Hz, 1H), 7.10 (t, *J* = 8.0 Hz, 1H), 5.54 (s, 1H) ppm; ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 185.9, 181.1, 163.6, 158.6, 154.4, 150.2, 138.4, 134.3, 133.5, 131.7, 130.6, 128.1, 126.7, 126.1, 125.7, 125.6, 123.5, 117.5, 114.9, 85.6, 34.7 ppm; HRMS (ESI-TOF) calcd for $C_{21}H_{12}ClN_3O_4$ $[M + H]^+$ 406.0594, found 406.0615.

5-*m*-bromophenylbenzo[*G*]pyrimido[4,5-*b*]quinoline-2,4,6,11(1*h*,3*h*,5*h*,12*h*)-tetrone (4bi)



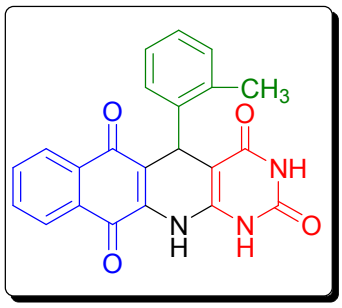
Yield: 92%; Caramine red solid; mp = 285-286 ^{o}C; IR (KBr, cm^{-1}): 3347, 3257, 3163, 2981, 1724, 1647, 1585, 1427, 1207, 1161, 1015, 941, 848, 752; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 10.80 (s, 1H), 10.04 (s, 1H), 9.53 (s, 1H), 8.08 (d, J = 8.0 Hz, 1H), 7.99 (d, J = 8.0 Hz, 1H), 7.79-7.72 (m, 2H), 7.52 (s, 1H), 7.34 (d, J = 8.0 Hz, 1H), 7.26 (d, J = 8.0 Hz, 1H), 7.14 (t, J = 8.0 Hz, 1H), 5.24 (s, 1H) ppm; ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 181.4, 178.8, 162.6, 160.0, 150.4, 149.5, 146.9, 141.7, 139.4, 138.9, 137.4, 134.6, 130.9, 130.8, 129.6, 129.3, 126.7, 125.8, 119.3, 89.4, 34.2 ppm; HRMS (ESI-TOF) calcd for $C_{21}H_{12}BrN_3O_4$ $[M + H]^+$ 450.0089, found 450.0106.

5-*o*-methoxyphenylbenzo[*G*]pyrimido[4,5-*b*]quinoline-2,4,6,11(1*h*,3*h*,5*h*,12*h*)-tetrone (4bk)



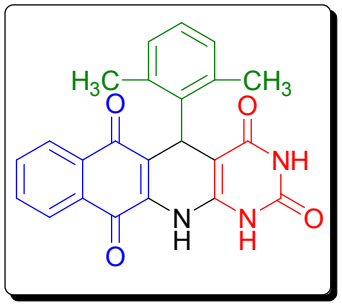
Yield: 96 %; Maroon solid; mp = 323-325 ^{o}C; IR (KBr, cm^{-1}): 3438, 3367, 3165, 3043, 2937, 1678, 1662, 1645, 1612, 1501, 1426, 1293, 1055, 965, 847, 762; ^{1}H NMR (500 MHz, $CDCl_3$) δ_H: 10.75 (s, 1H), 10.09 (s, 1H), 9.37 (s, 1H), 8.03 (d, J = 8.0 Hz, 1H), 7.87-7.77 (m, 3H), 7.32 (d, J = 10.0 Hz, 1H), 7.12 (t, J = 10.0 Hz, 1H), 6.89-6.82 (m, 2H), 5.22 (s, 1H), 3.68 (s, 3H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ_C: 182.1, 179.8, 162.9, 158.5, 149.9, 144.4, 138.7, 165.6, 133.8, 132.6, 131.9, 128.4, 126.4, 126.3, 120.4, 118.2, 112.5, 87.9, 56.2, 32.9 ppm; HRMS (ESI-TOF) calcd for $C_{22}H_{15}N_3O_5$ $[M + H]^+$ 402.1089, found 402.1068.

5-*o*-methylphenylbenzo[*G*]pyrimido[4,5-*b*]quinoline-2,4,6,11(1*h*,3*h*,5*h*,12*h*)-tetrone (4bl)



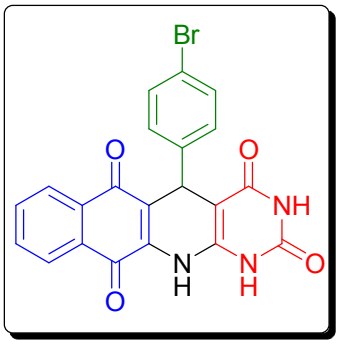
Yield: 93%; Caramine red solid; mp = 320-321 ^{o}C; IR (KBr, cm^{-1}): 3478, 3365, 3241, 3057, 2968, 1698, 1661, 1648, 1611, 1443, 1297, 1061, 971, 837, 791; ^{1}H NMR (500 MHz, DMSO-d_6) δ_H: 10.84 (s, 1H), 10.17 (s, 1H), 9.31 (s, 1H), 8.05 (d, J = 10.0 Hz, 1H), 7.89-7.88 (m, 1H), 7.83–7.80 (m, 2H), 7.15 (d, J = 10.0 Hz, 1H), 7.04 (d, J = 10.0 Hz, 1H), 7.02-6.97 (m, 2H), 5.19 (s, 1H), 2.79 (s, 3H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ_C: 179.6, 163.1, 149.8, 145.9, 143.7, 136.2, 135.5, 133.9, 129.8, 129.2, 126.9, 126.8, 126.5, 126.3, 125.1, 124. 0, 120.4, 90.5, 31.1, 20.0 ppm;HRMS (ESI-TOF) calcd for $C_{22}H_{15}N_3O_4$ $[M + H]^+$ 386.1140, found 386.1158.

5-*2,6*-dimethylphenylbenzo[*G*]pyrimido[4,5-*b*]quinoline-2,4,6,11(1*h*,3*h*,5*h*,12*h*)-tetrone (4bm)



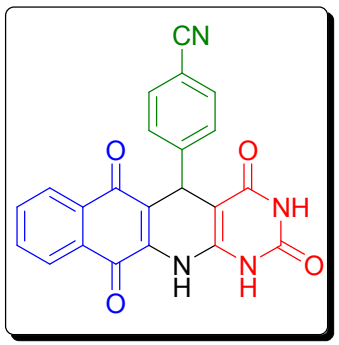
Yield: 87%; Maroon solid; mp = 314-315 ^{o}C; IR (KBr, cm^{-1}): 3457, 3264, 3132, 3065, 2934, 1698, 1672, 1657, 1614, 1432, 1290, 1055, 945, 823, 762; ^{1}H NMR (500 MHz, DMSO-d_6) δ_H: 10.76 (s, 1H), 10.08 (s, 1H), 9.37 (s, 1H), 8.02-8.00 (m, 1H), 7.82-7.78 (m, 3H), 6.95-6.83 (m, 3H), 5.55 (s, 1H), 2.86 (s, 3H), 2.14 (s, 3H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ_C: 182.5, 179.3, 178.6, 162.9, 149.7, 143.9, 140.3, 139.4, 138.9, 137.3, 135.5, 133.8, 132.3, 130.4, 130.2, 128.5, 126.4, 118.1, 87.5, 32.1, 21.9, 19.0 ppm; HRMS (ESI-TOF) calcd for $C_{23}H_{17}N_3O_4$ $[M + H]^+$ 400.1297, found 400.1316 .

5-*p*-bromophenylbenzo[*G*]pyrimido[4,5-*b*]quinoline-2,4,6,11(1*h*,3*h*,5*h*,12*h*)-tetrone (4bn)



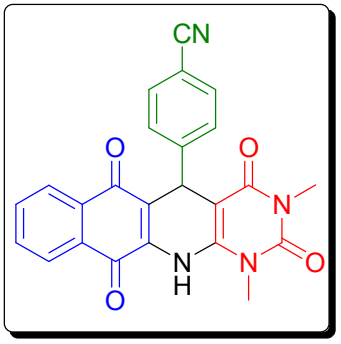
Yield: 87%; Orange solid; mp = 293-294 ^{o}C; IR (KBr, cm^{-1}): 3347, 3257, 3163, 2981, 1724, 1647, 1585, 1427, 1207, 1161, 1015, 941, 848, 752; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 10.89 (s, 1H), 10.14 (s, 1H), 9.53 (s, 1H), 8.06 (d, J = 8.0 Hz, 1H), 7.95 (d, J = 8.0 Hz, 1H), 7.77 (t, J = 8.0 Hz, 2H), 7.52 (d, J = 8.0 Hz, 1H), 7.33 (d, J = 8.0 Hz, 1H), 7.28 (d, J = 8.0 Hz, 1H), 7.16 (d, J = 8.0 Hz, 1H), 5.14 (s, 1H) ppm; ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 181.5, 178.9, 162.5, 149.3, 147.3, 143.6, 137.9, 134.8, 133.1, 131.7, 130.8, 129.9, 129.3, 126.9, 125.9, 121.4, 117.9, 88.1, 34.5 ppm; HRMS (ESI-TOF) calcd for $C_{21}H_{12}BrN_3O_4$ $[M + H]^+$ 450.0089, found 450.0104.

5-*p*-cyanophenylbenzo[*G*]pyrimido[4,5-*b*]quinoline-2,4,6,11(1*h*,3*h*,5*h*,12*h*)-tetrone (4bo)



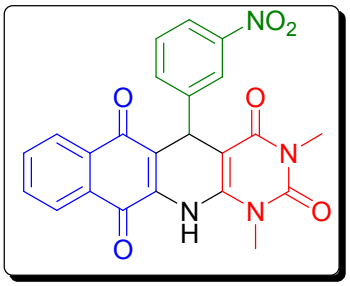
Yield: 78%; Orange solid; mp = 293-294 ^{o}C; IR (KBr, cm^{-1}): 3379, 3257, 3166, 3028, 2223, 1724, 1667, 1601, 1446, 1292, 1157, 1076, 952, 860, 741; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 10.88 (s, 1H), 10.17 (s, 1H), 9.83 (s, 1H), 8.05 (d, J = 8.0 Hz, 1H), 7.92 (d, J = 8.0 Hz, 1H), 7.85 (t, J = 8.0 Hz, 1H), 7.81 (t, J = 8.0 Hz, 1H), 7.23 (d, J = 8.0 Hz, 2H), 7.08 (t, J = 8.0 Hz, 2H), 5.08 (s, 1H) ppm; ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 185.8, 181.0, 163.5, 158.5, 154.4, 150.1, 137.7, 134.3, 133.4, 131.7, 130.6, 130.2, 128.8, 127.9, 126.0, 125.7, 123.2, 118.5, 85.4, 34.4 ppm; HRMS (ESI-TOF) calcd for $C_{22}H_{12}N_4O_4$ $[M + H]^+$ 397.0936, found 397.0955.

1,3-dimethyl-5-*p*-cyanophenyl-benzo[*G*]pyrimido[4,5-*b*]quinoline-2,4,6,11(1*h*,3*h*,5*h*, 12*h*)-tetrone (4ao)



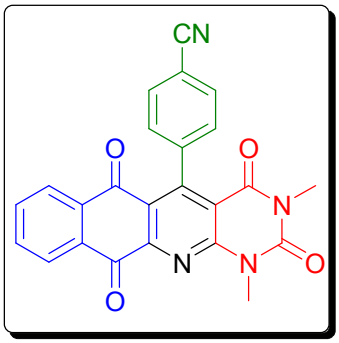
Yield: 38%; Reddish brown solid; mp = 317-318 ^{o}C; IR (KBr, cm^{-1}): 3298, 3152, 3082, 2251, 1716, 1667, 1605, 1446, 1157, 1083, 840, 763; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 8.89 (s, 1H), 8.07 (d, J = 8.0 Hz, 1H), 7.93 (d, J = 8.0 Hz, 1H), 7.86 (t, J = 8.0 Hz, 2H), 7.71 (d, J = 8.0 Hz, 2H), 7.55 (d, J = 8.0 Hz, 2H), 5.29 (s, 1H), 3.58 (s, 3H), 3.12 (s, 3H) ppm; ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 181.7, 179.6, 163.6, 158.9, 150.5, 149.7, 135.1, 133.8, 132.1, 131.5, 130.2, 129.2, 126.6, 126.1, 123.5, 118.8, 118.5, 109.4, 88.8, 35.7, 29.8, 27.8 ppm; HRMS (ESI-TOF) calcd for $C_{24}H_{16}N_4O_4$ $[M + H]^+$ 425.1249, found 425.1268.

1,3-dimethyl-5-*m*-nitrophenyl-benzo[*G*]pyrimido[4,5-*b*]quinoline-2,4,6,11(1*h*,3*h*,5*h*,12*h*)-tetrone (4ap)



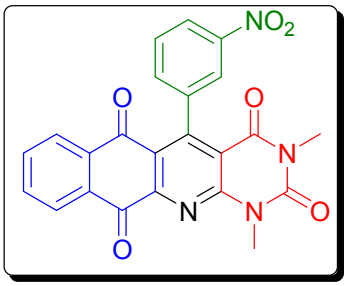
Yield: 35%; Reddish brown solid; mp = 310-312 ^{o}C; IR (KBr, cm^{-1}): 3363, 3089, 2955, 1685, 1654, 1639, 1616, 1528, 1473, 1296, 1161, 1045, 972, 887, 783; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 8.83 (s, 1H), 8.06 (d, J = 8.0 Hz, 1H), 7.93 (d, J = 8.0 Hz, 1H), 7.82-7.75 (m, 2H), 7.60 (d, J = 8.0 Hz, 2H), 7.53 (d, J = 8.0 Hz, 2H), 5.33 (s, 1H), 3.59 (s, 3H), 3.13 (s, 3H) ppm; ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 181.4, 178.5, 165.8, 160.5, 157.3, 150.4, 149.5, 143.7, 134.8, 133.4, 131.8, 131.5, 130.1, 129.1, 126.0, 125.9, 118.9, 118.5, 109.7, 88.9, 35.7, 29.7, 27.6 ppm; HRMS (ESI-TOF) calcd for $C_{23}H_{16}N_4O_6$ $[M + H]^+$ 445.1148, found 445.1166.

Compound 6ao



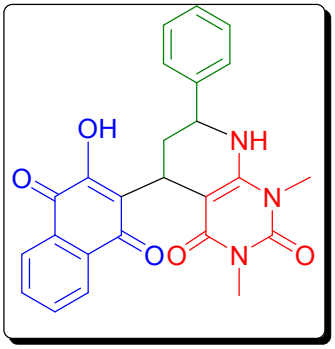
Yield: 52%; Yellow solid; mp = 317-320 ^{o}C; IR (KBr, cm^{-1}): 3078, 2954, 2222, 1712, 1663, 1612, 1523, 1446, 1384, 1227, 1161, 1083, 852, 763; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 8.77 (d, J = 8.0 Hz, 1H), 8.07 (d, J = 8.0 Hz, 1H), 7.94 (t, J = 8.0 Hz, 1H), 7.83 (d, J = 8.0 Hz, 2H), 7.75 (t, J = 8.0 Hz, 3H), 3.82 (s, 3H) 3.16 (s, 3H) ppm; ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 186.9, 181.0, 164.3, 158.1, 154.7, 150.3, 149.9, 143.6, 133.6, 132.9, 131.4, 130.5, 125.9, 125.5, 124.0, 118.9, 118.5, 109.7, 85.7, 30.2, 27.9 ppm; HRMS (ESI-TOF) calcd for $C_{24}H_{14}N_4O_4$ [M + H]$^{+}$ 423.1093, found 423.1112.

Compound 6ap



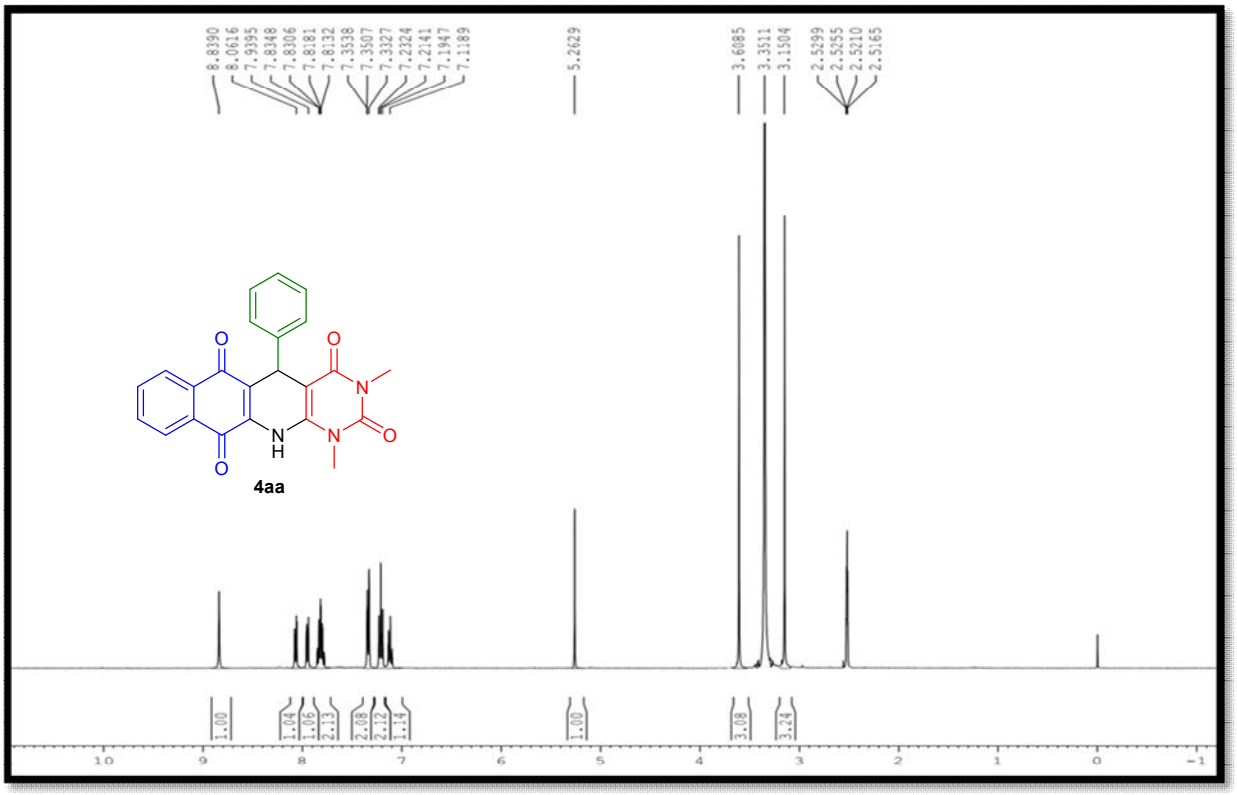
Yield: 58%; Yellow solid; mp = 305-307 ^{o}C; IR (KBr, cm^{-1}): 3078, 2954, 1712, 1681, 1658, 1647, 1612, 1527, 1469, 1238, 1161, 1045, 887, 752; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 8.76 (d, J = 8.0 Hz, 1H), 8.27 (d, J = 8.0 Hz, 1H), 8.06 (t, J = 8.0 Hz, 2H), 7.93 (t, J = 8.0 Hz, 1H), 7.75 (t, J = 8.0 Hz, 1H), 7.71 (t, J = 8.0 Hz, 1H), 7.59 (d, J = 8.0 Hz, 1H), 3.82 (s, 1H) 3.34 (s, 3H) ppm; ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 178.2, 177.3, 159.2, 156.0, 153.2, 152.7, 150.5, 147.4, 140.1, 135.3, 133.5, 132.4, 128.9, 128.1, 127.2, 121.8, 108.6, 89.4, 30.1, 28.1 ppm; HRMS (ESI-TOF) calcd for $C_{23}H_{14}N_4O_6$ [M + H]$^{+}$ 443.0991, found 443.1012.

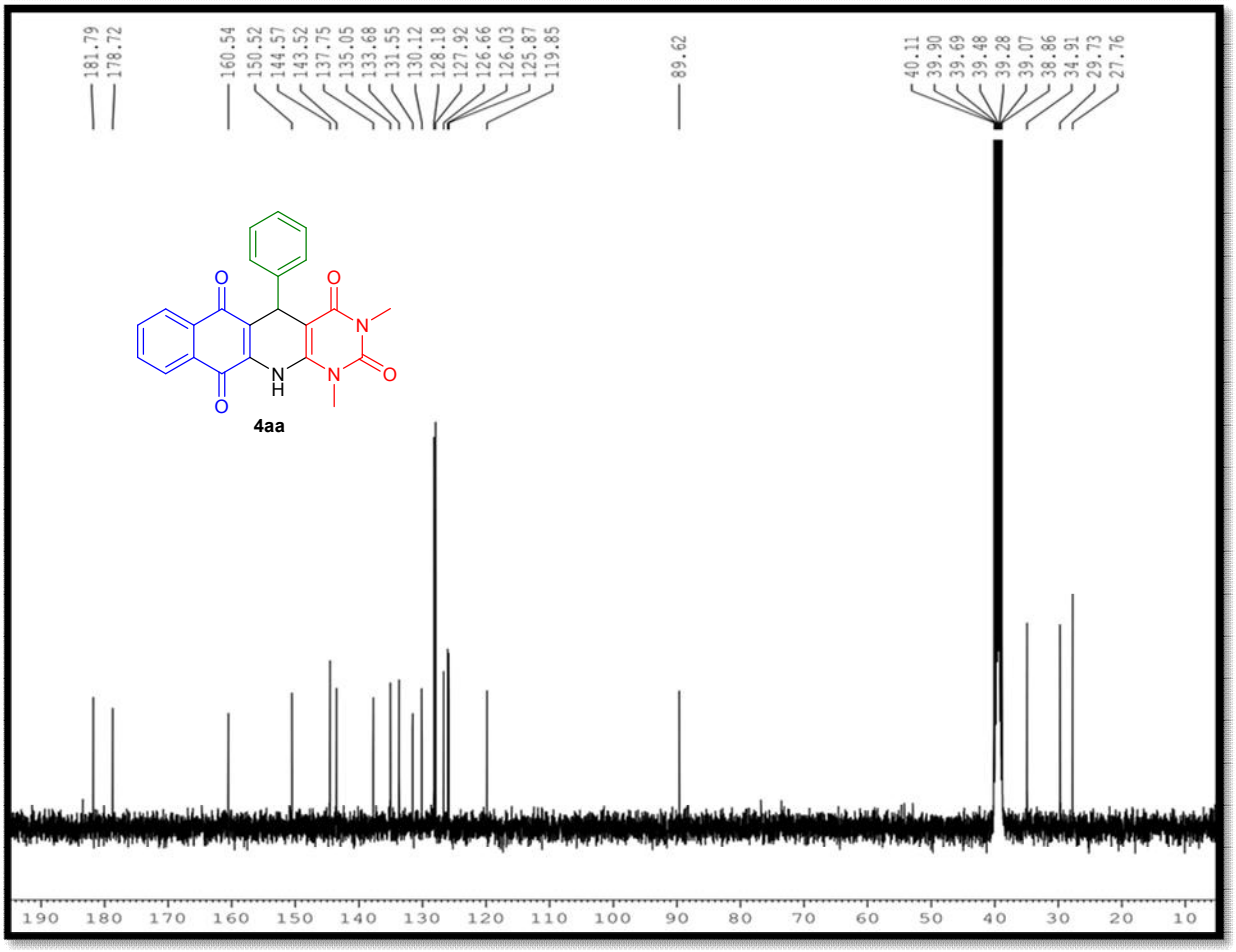
6-amino-5-((2*E*)-1-(1,4-dihydro-2-hydroxy-1,4-dioxonaphthalen-3-yl)-3-phenylallyl)-1,3-dimethylpyrimidine-2,4(1*H*,3*H*)-dione (7as)



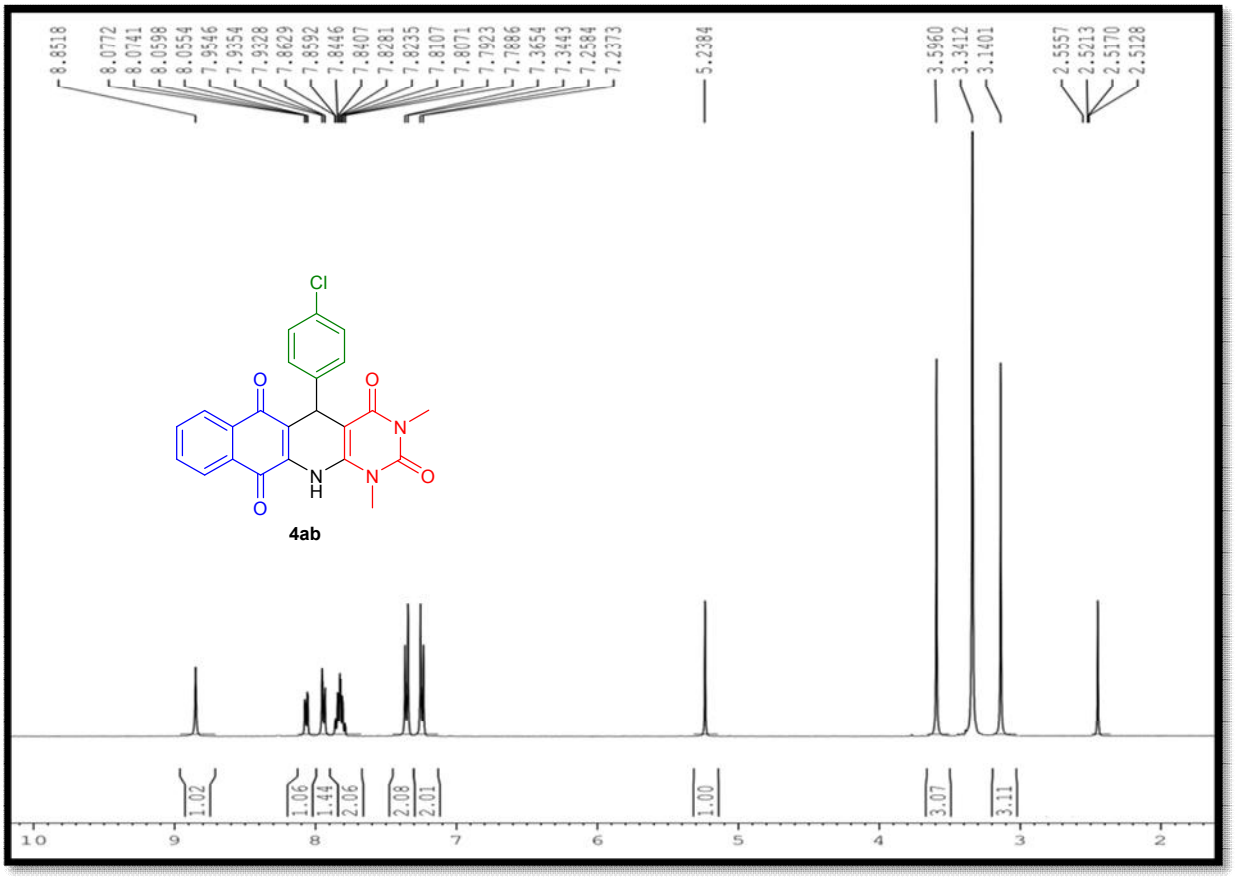
Yield: 85%; Caramine red solid; mp = 300-301 ^{o}C; IR (KBr, cm^{-1}): 3363, 3159, 3055, 2943, 1681, 1657, 1620, 1577, 1477, 1215, 1029, 972, 875, 748; ^{1}H NMR (400 MHz, $CDCl_3$) δ_H: 13.38 (s, 1H), 8.01 (d, *J* = 8.0 Hz, 1H), 7.95 (d, *J* = 8.0 Hz, 1H), 7.05 (t, *J* = 8.0 Hz, 1H), 7.79 (t, *J* = 8.0 Hz, 1H), 7.30 (t, *J* = 8.0 Hz, 2H), 7.22 (s, 2H), 7.19 (d, *J* = 8.0 Hz, 1H), 6.87 (s, 1H), 4.65-4.61 (m, 1H), 4.18-4.17 (m, 1H), 3.28 (s, 3H), 3.12 (s, 3H), 2.45-2.49 (m, 1H), 1.81-1.84 (m, 1H) ppm; ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 183.2, 181.1, 160.4, 156.3, 151.1, 149.8, 145.6, 134.9, 133.2, 132.1, 129.7, 128.1, 127.5, 125.9, 125.7, 120.7, 82.5, 43.1, 35.3, 31.3, 29.2, 27.3 ppm; HRMS (ESI-TOF) calcd for $C_{25}H_{21}N_3O_5$ $[M + H]^+$ 444.1559, found 444.1570.

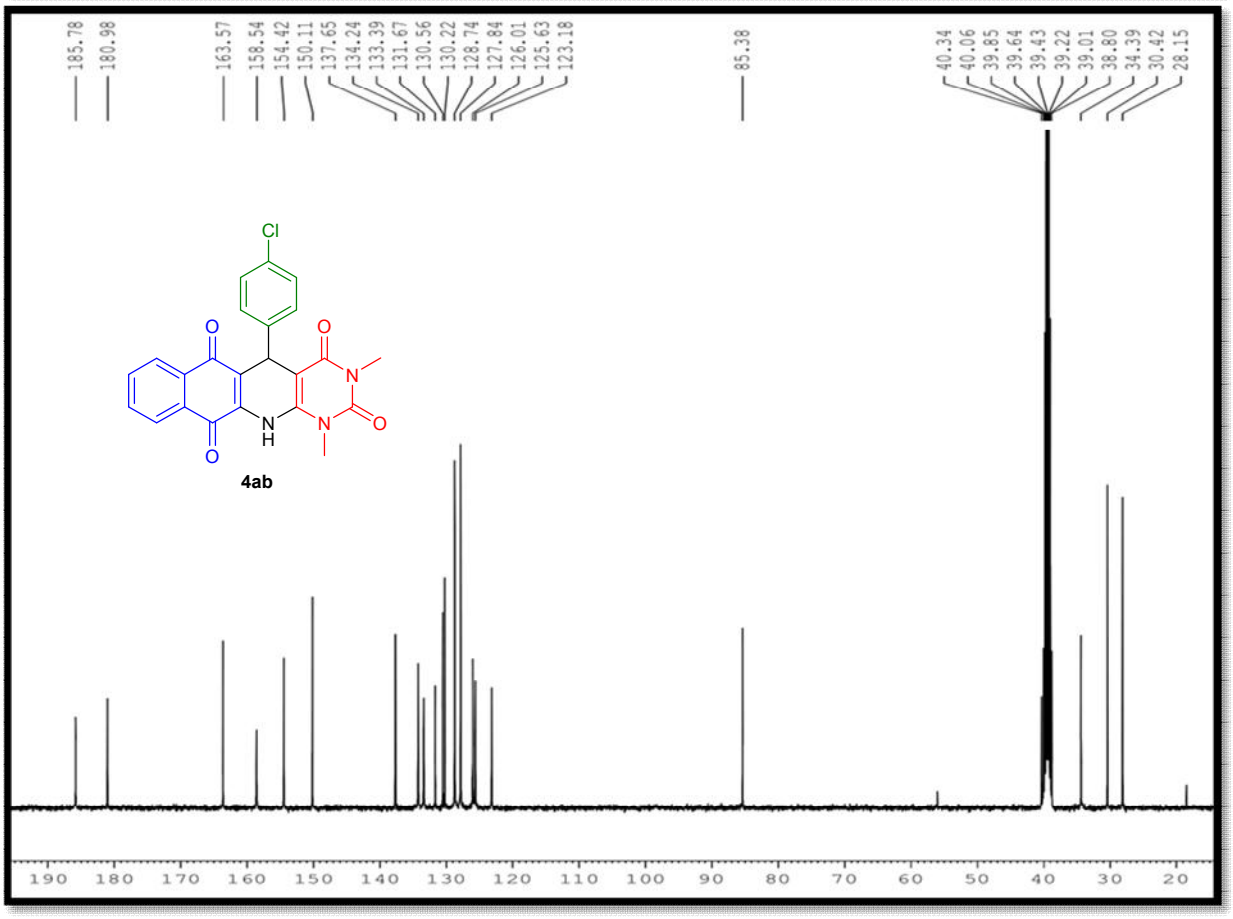
^{1}H & ^{13}C NMR of compound **4aa**



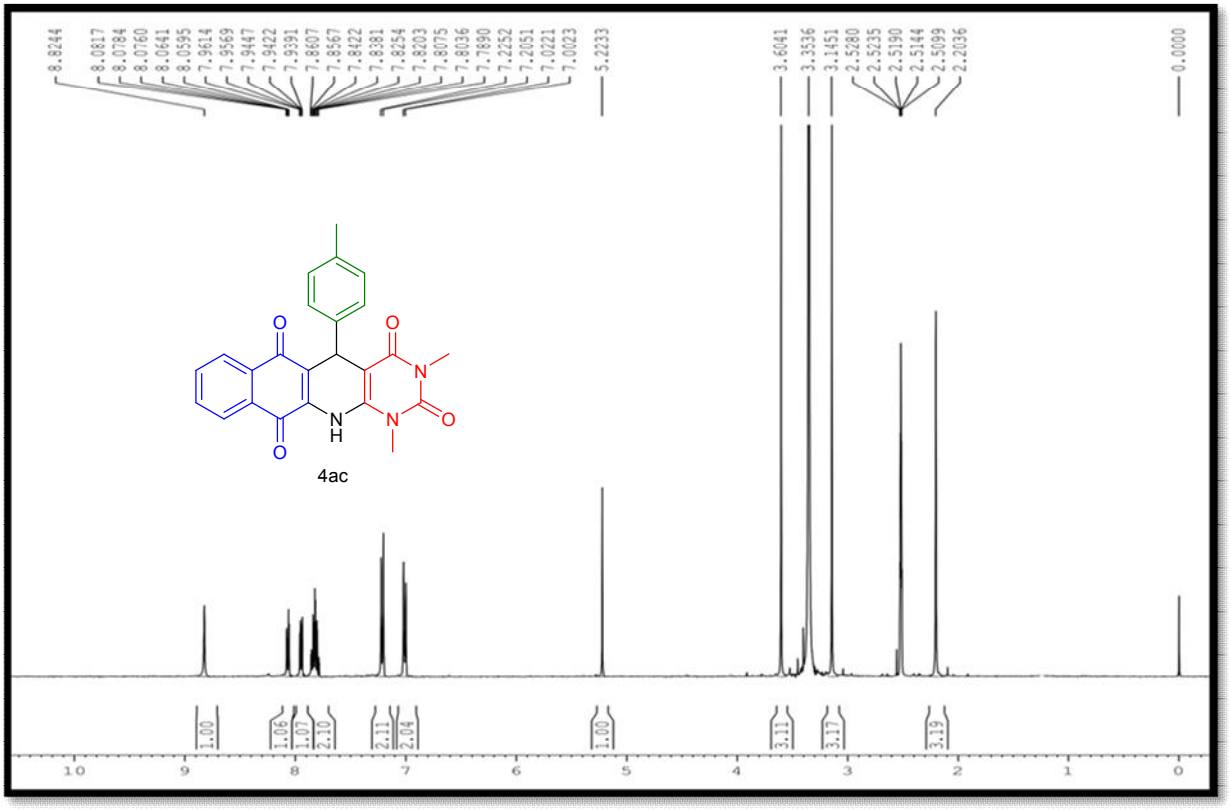


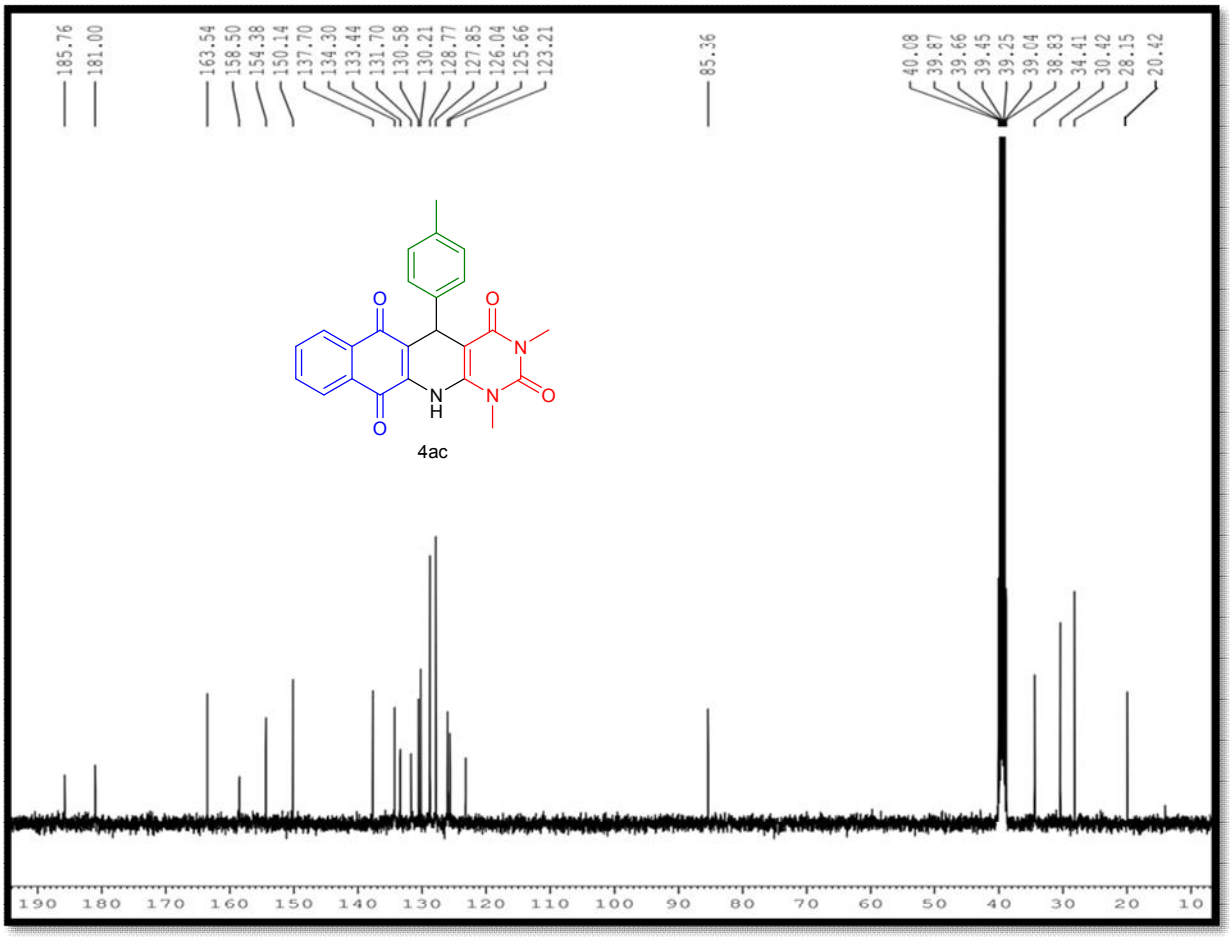
^{1}H & ^{13}C NMR of compound **4ab**



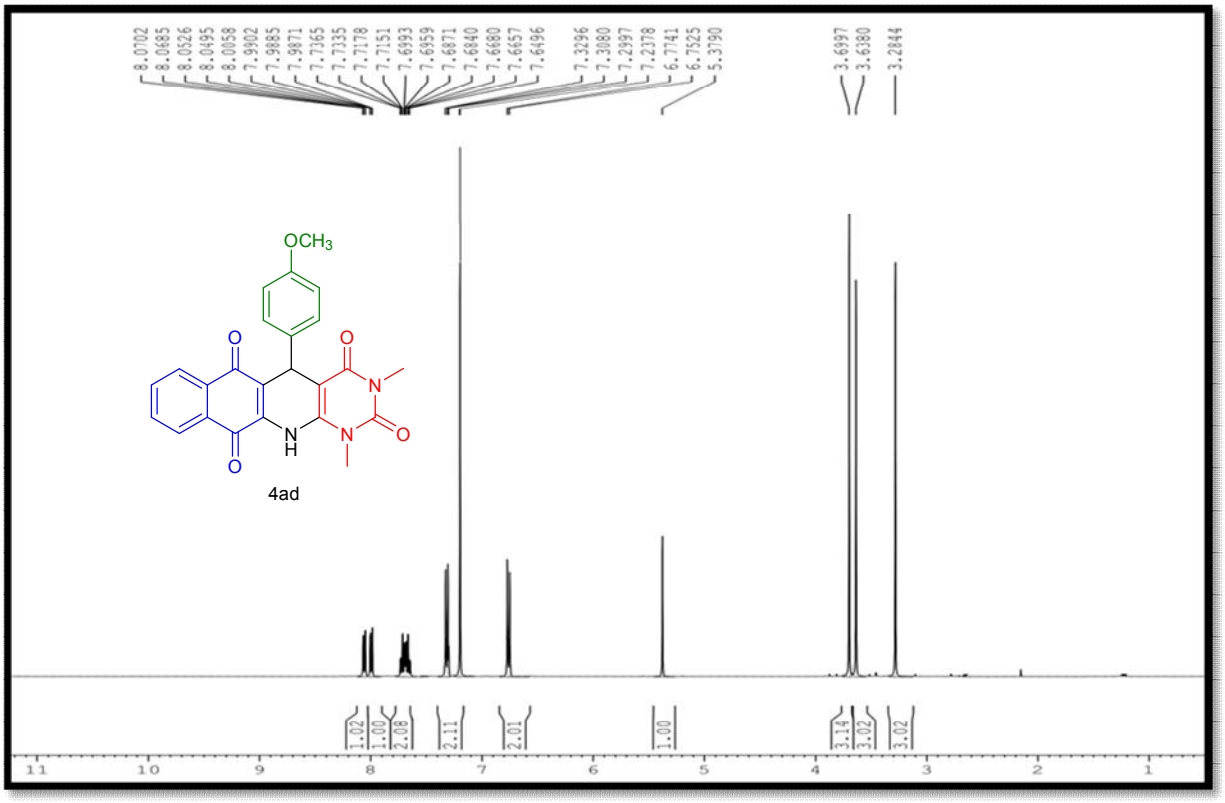


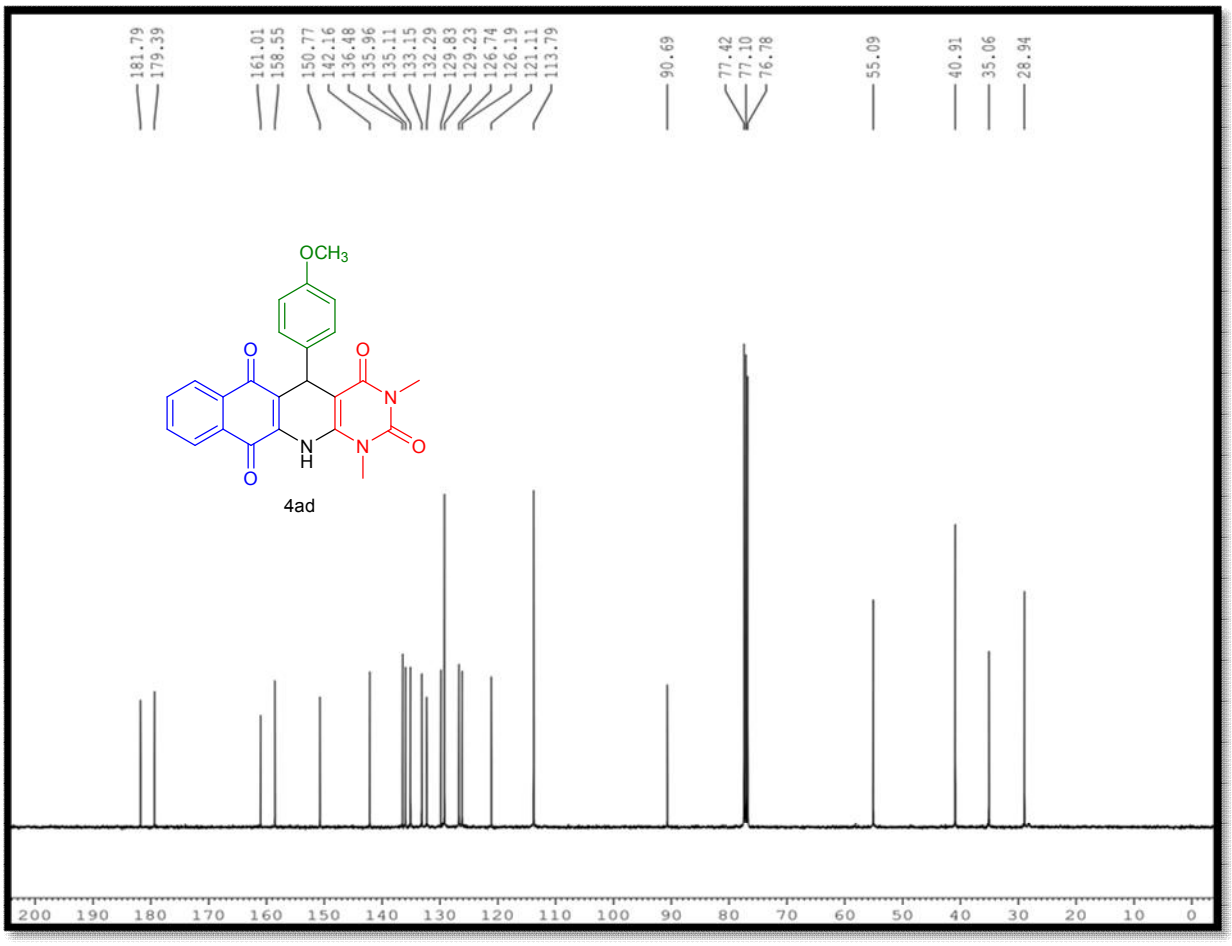
^{1}H &^{13}C NMR of compound **4ac**



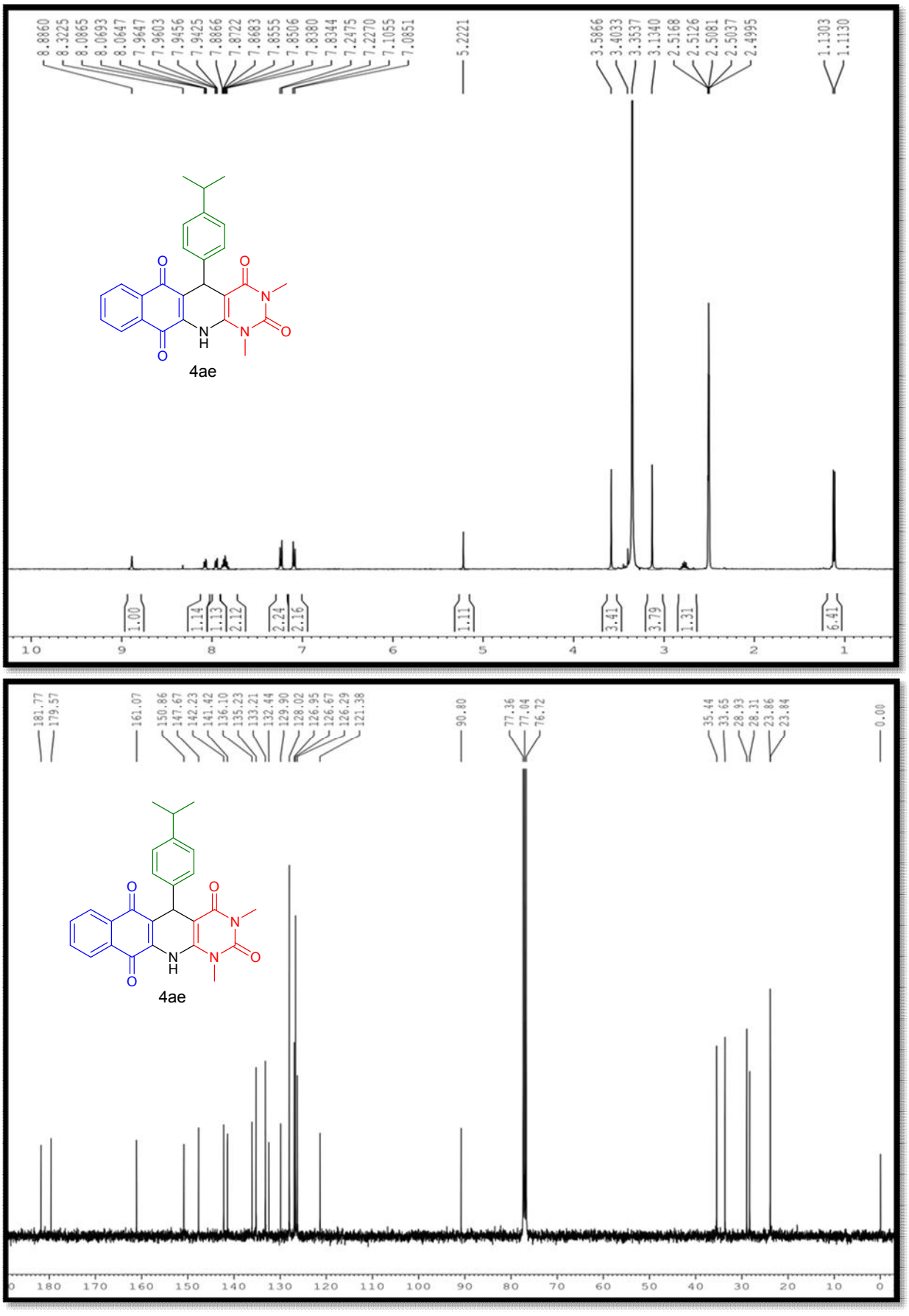


^{1}H & ^{13}C NMR Spectra of compound **4ad**

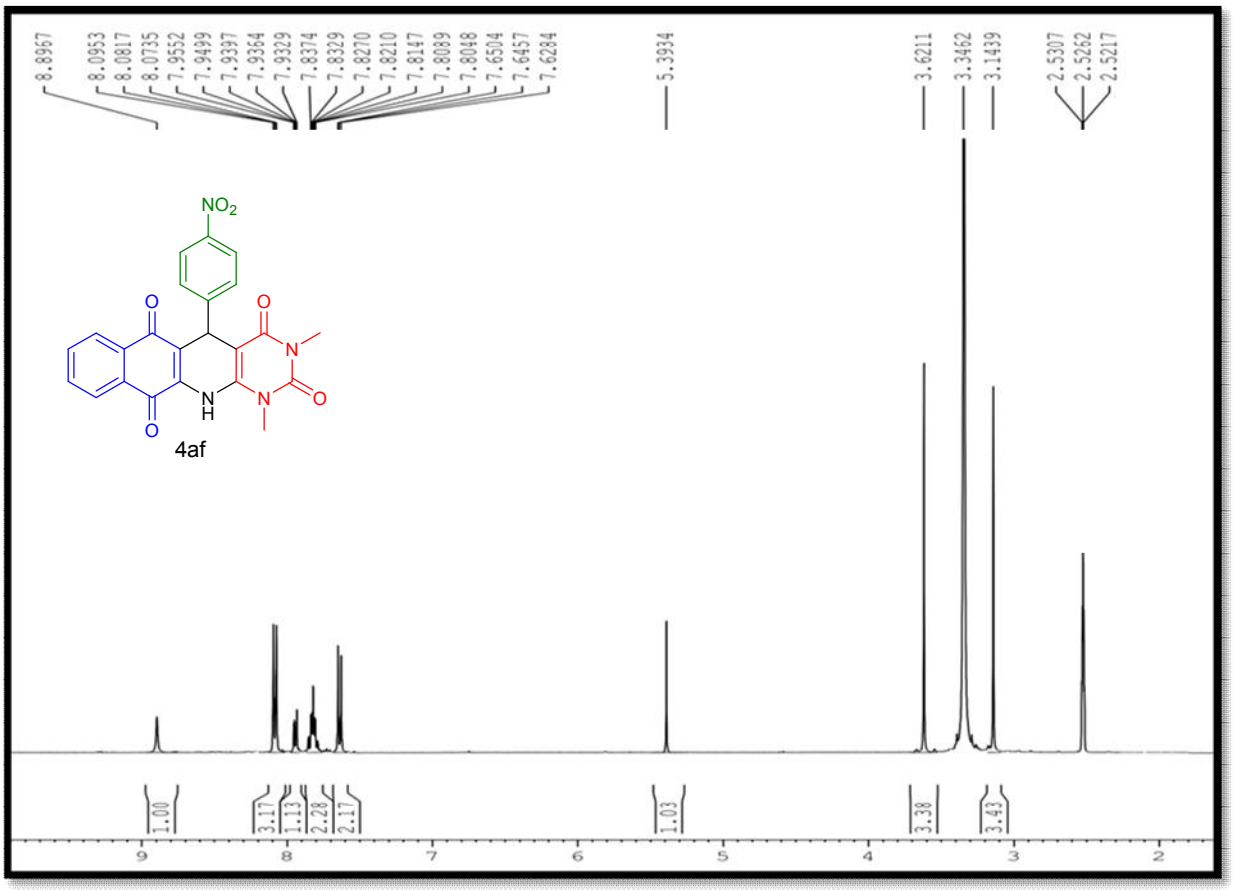


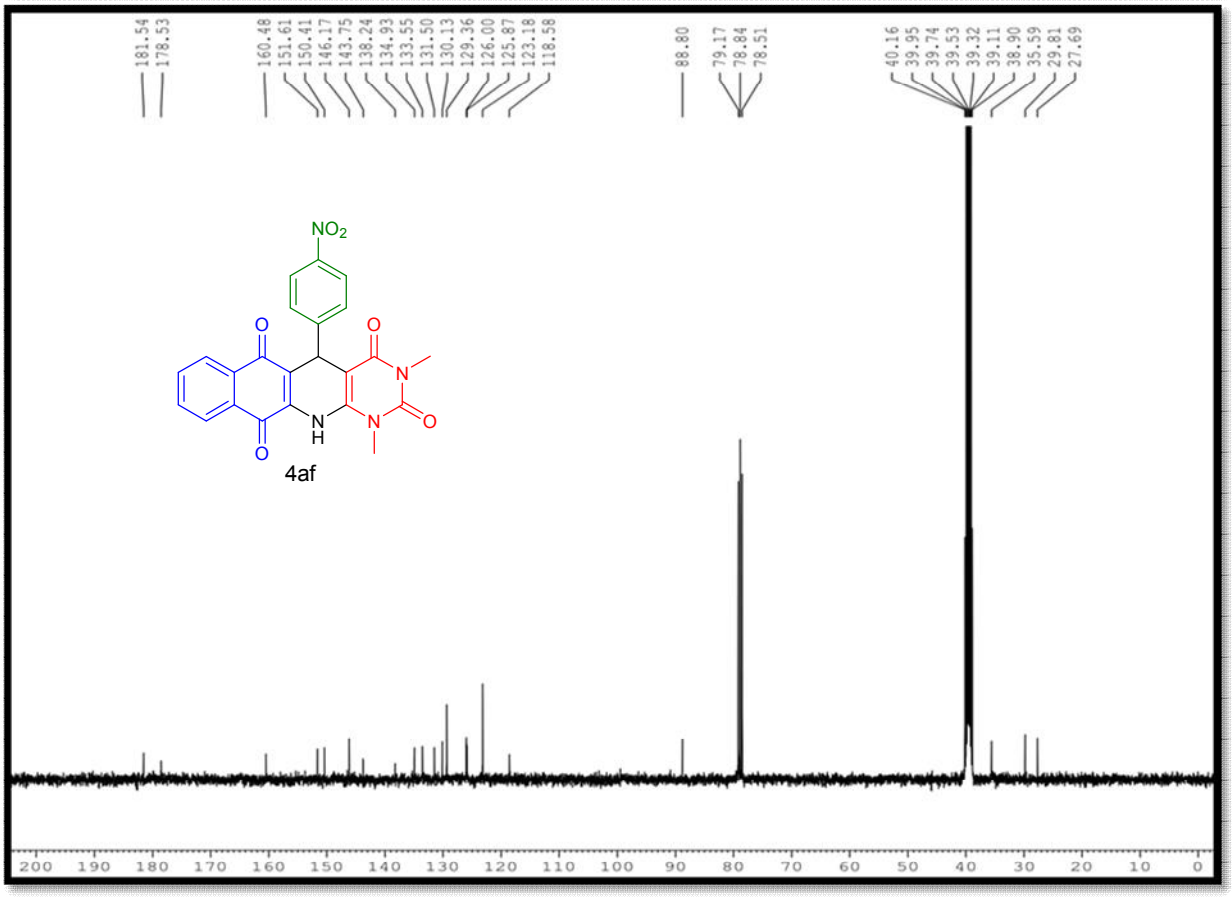


^{1}H & ^{13}C NMR Spectra of compound **4ae**

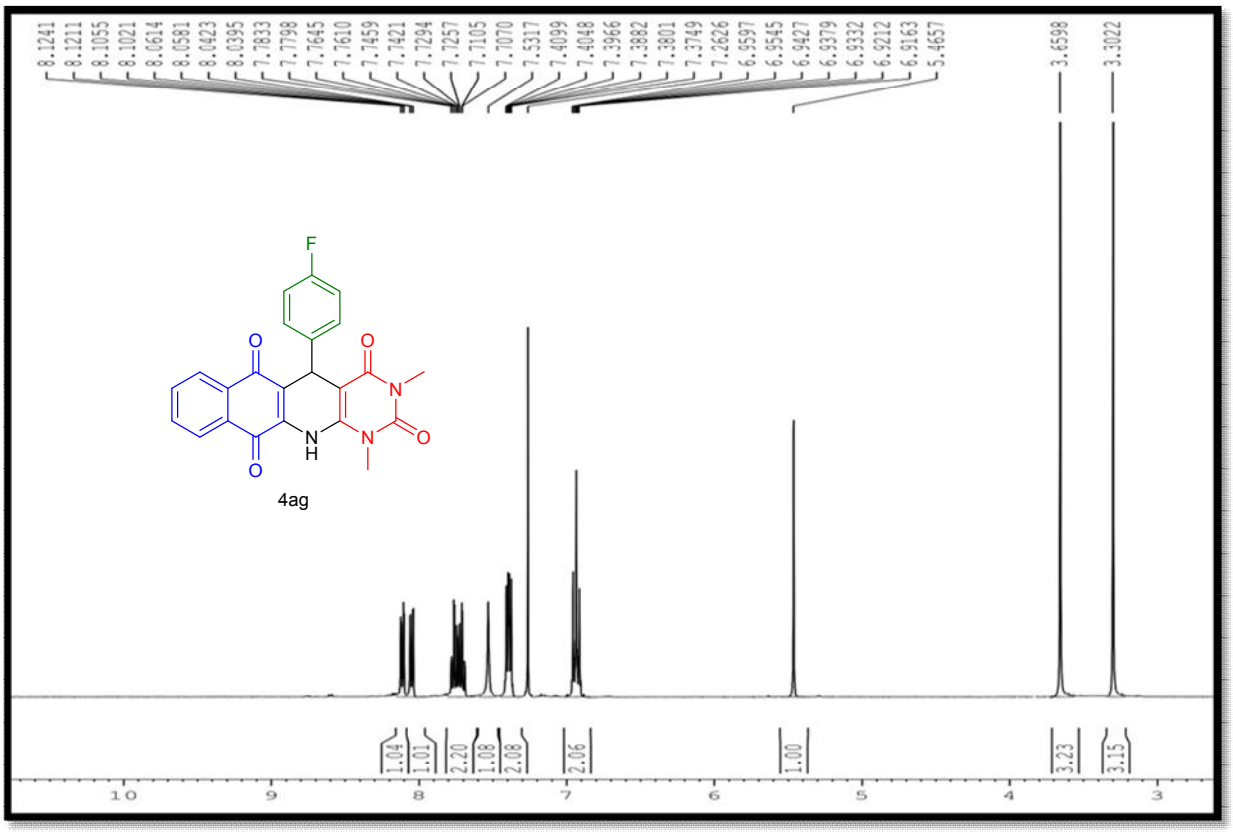


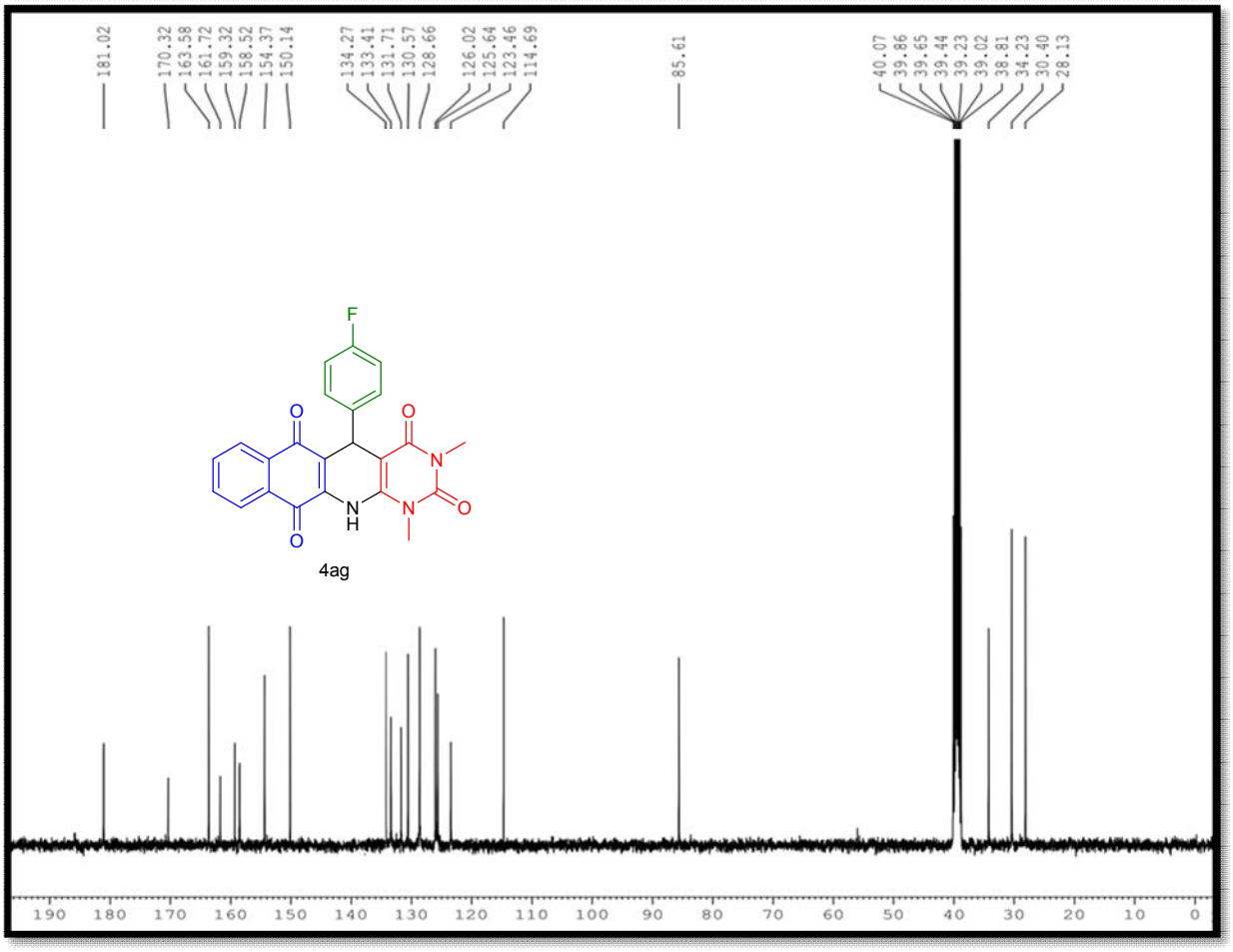
^{1}H & ^{13}C NMR Spectra of compound **4af**



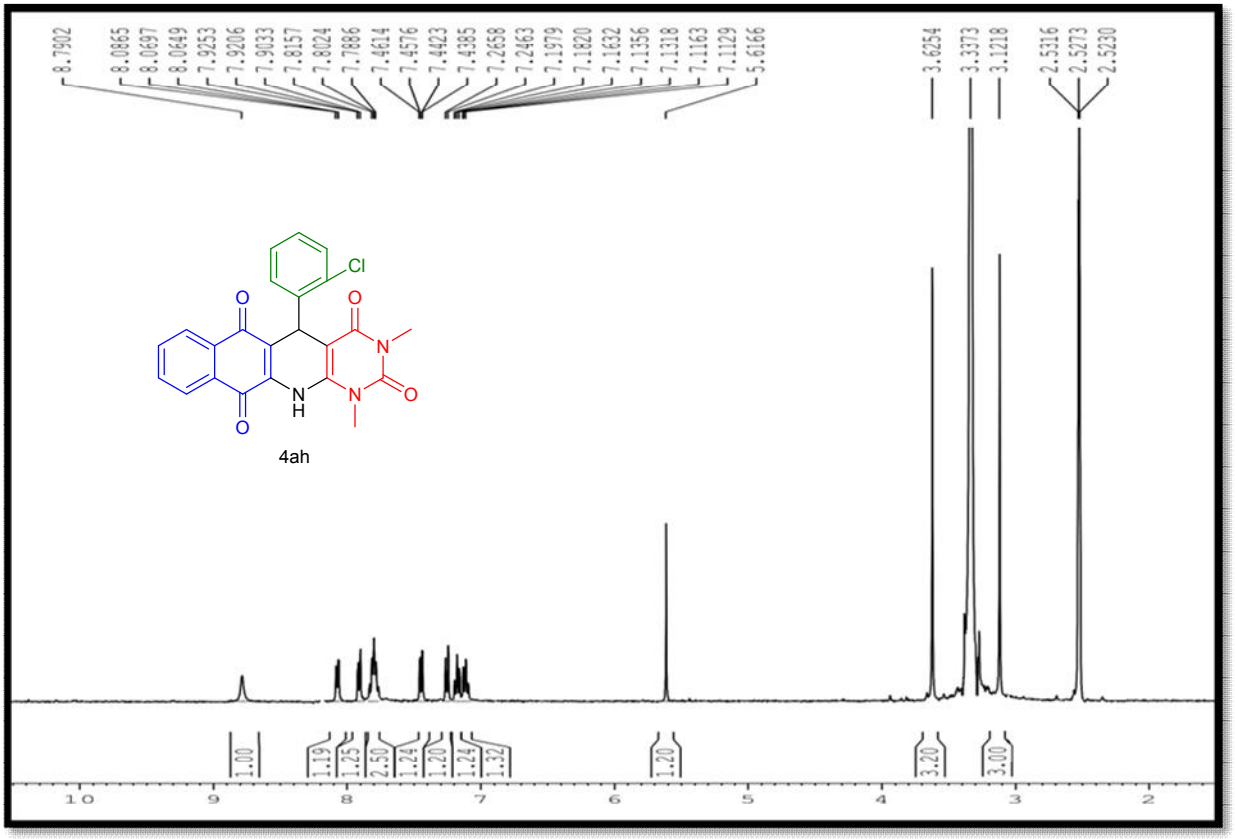


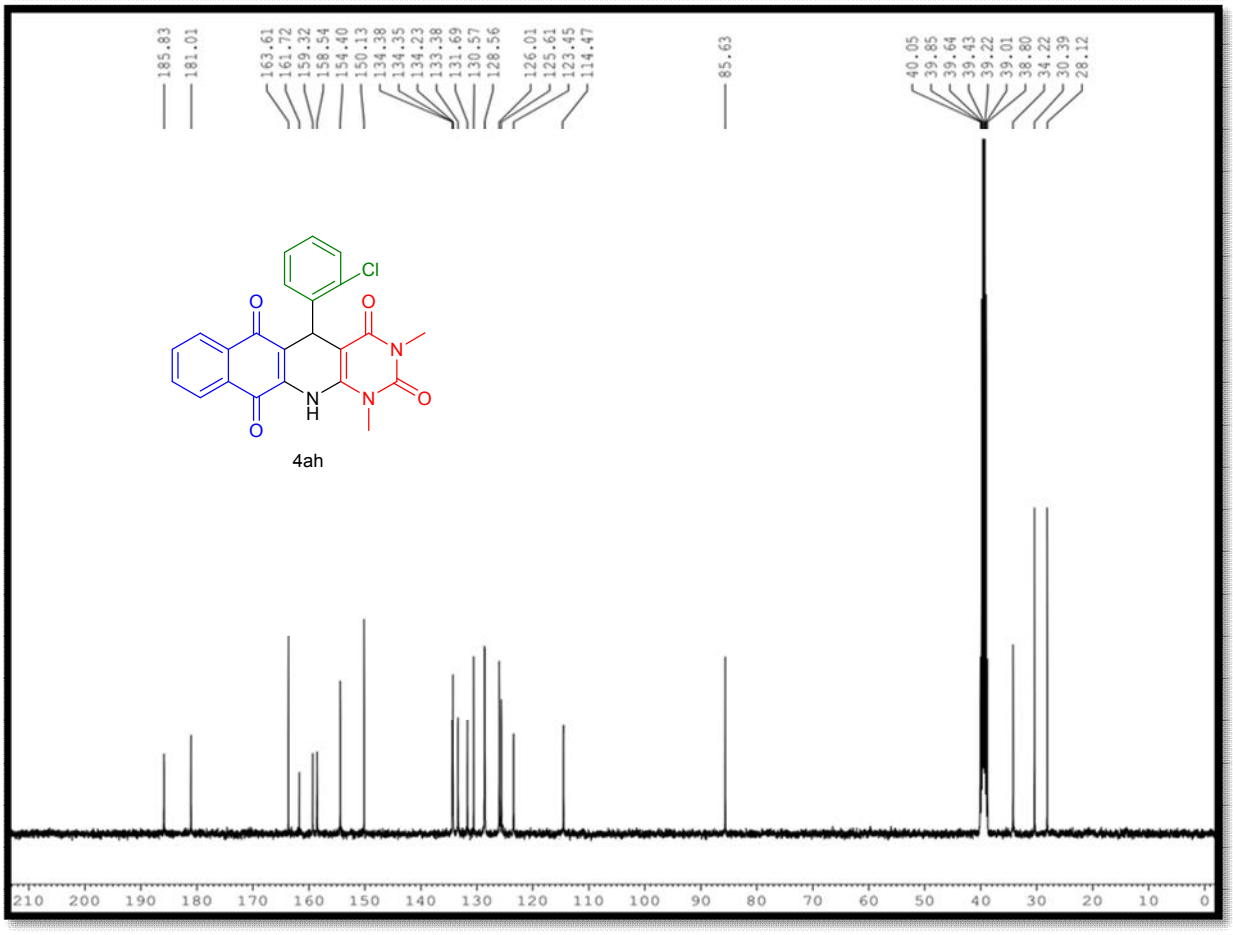
^{1}H & ^{13}C NMR Spectra of compound **4ag**



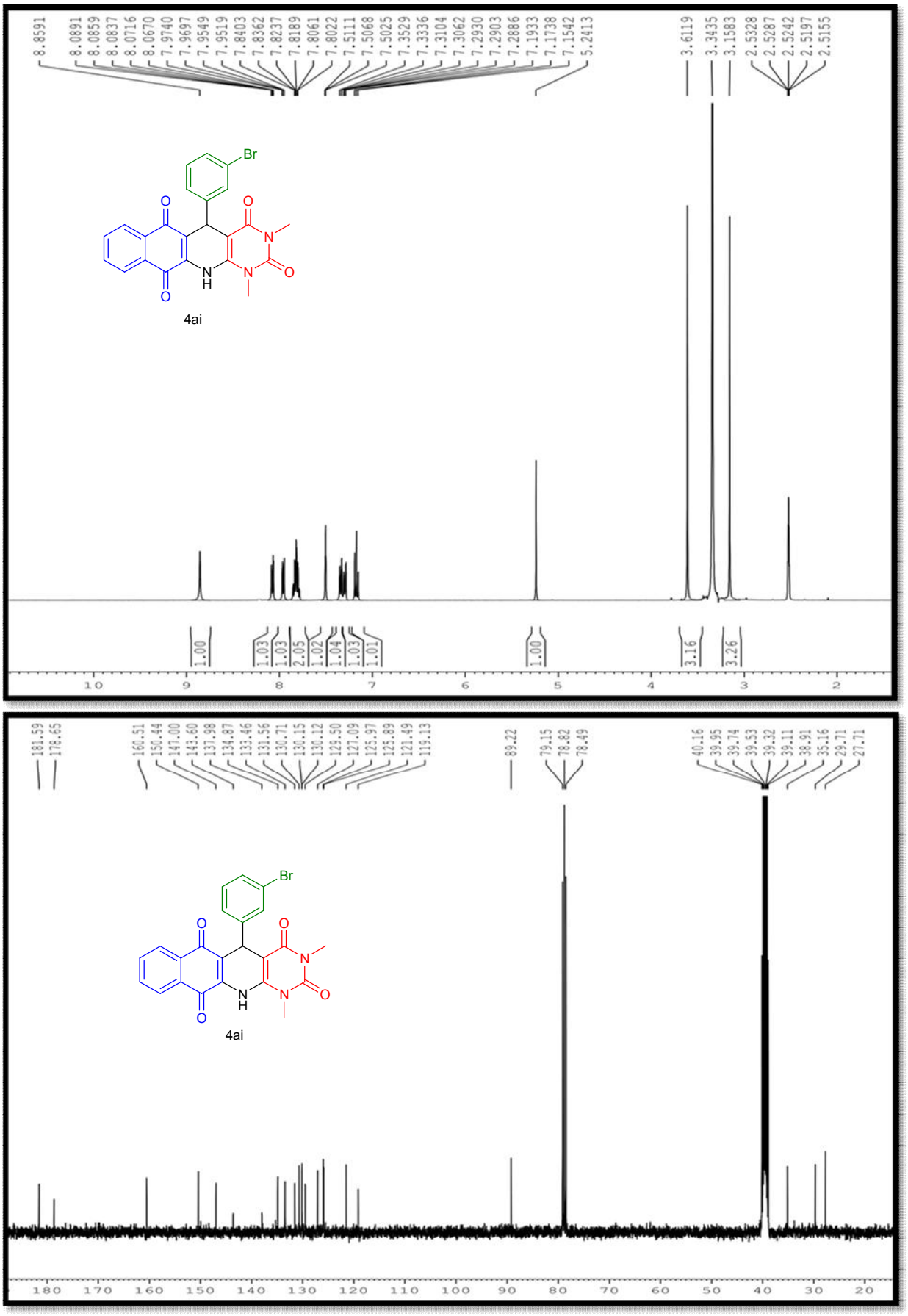


^{1}H & ^{13}C NMR Spectra of compound **4ah**

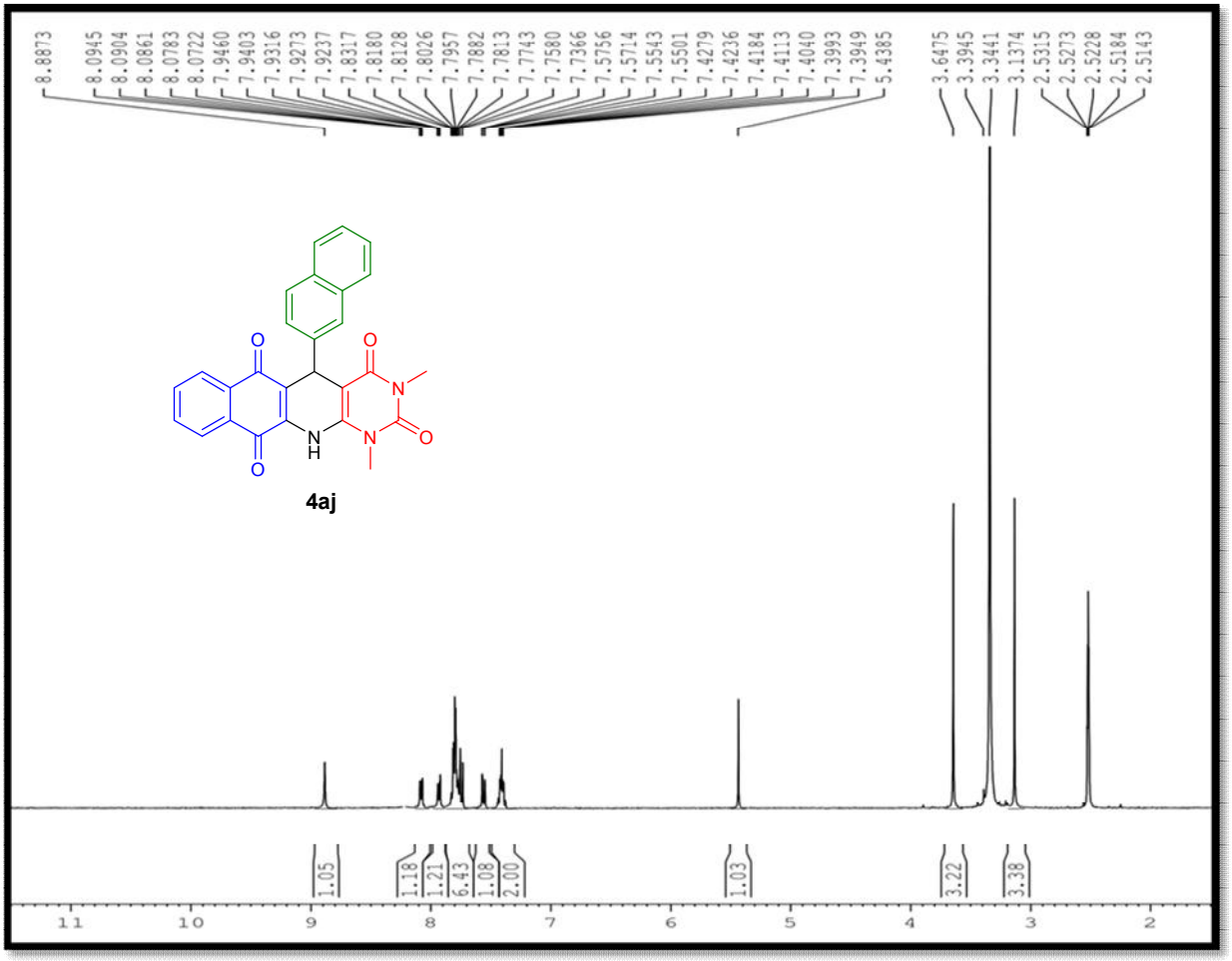


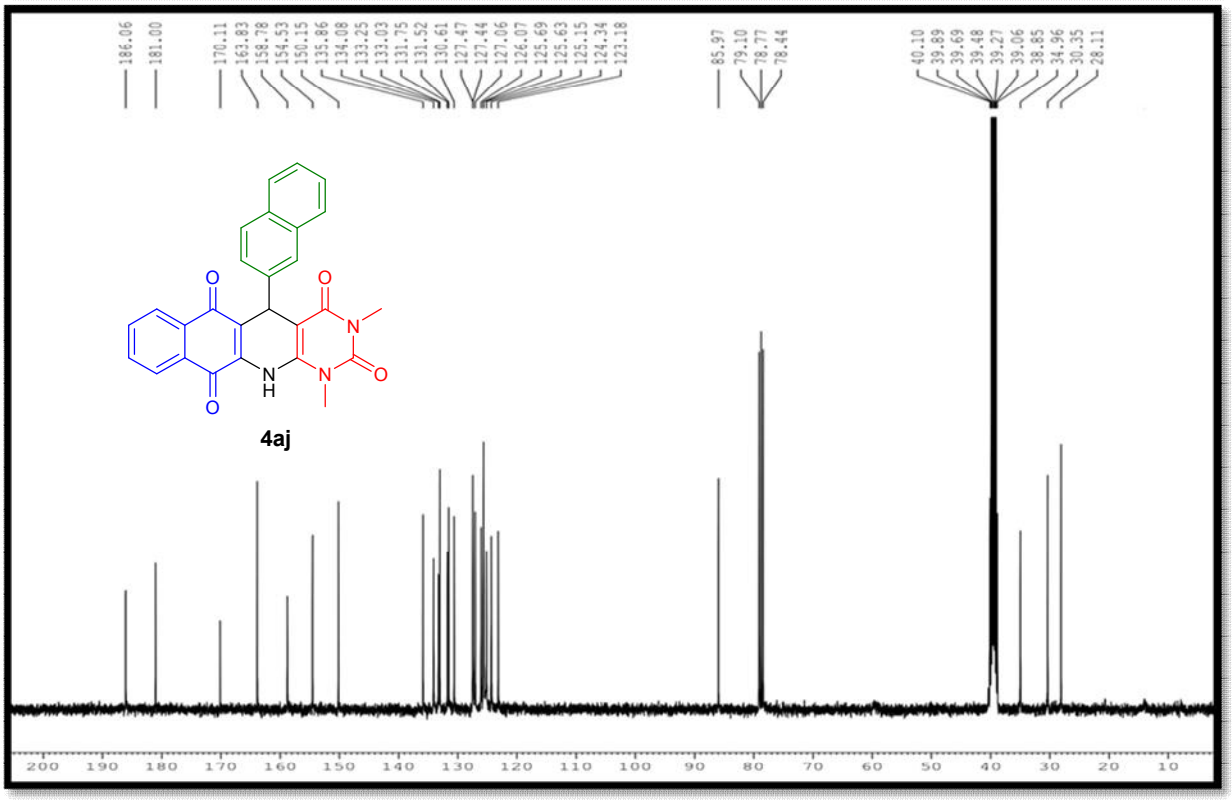


^{1}H & ^{13}C NMR Spectra of compound **4ai**

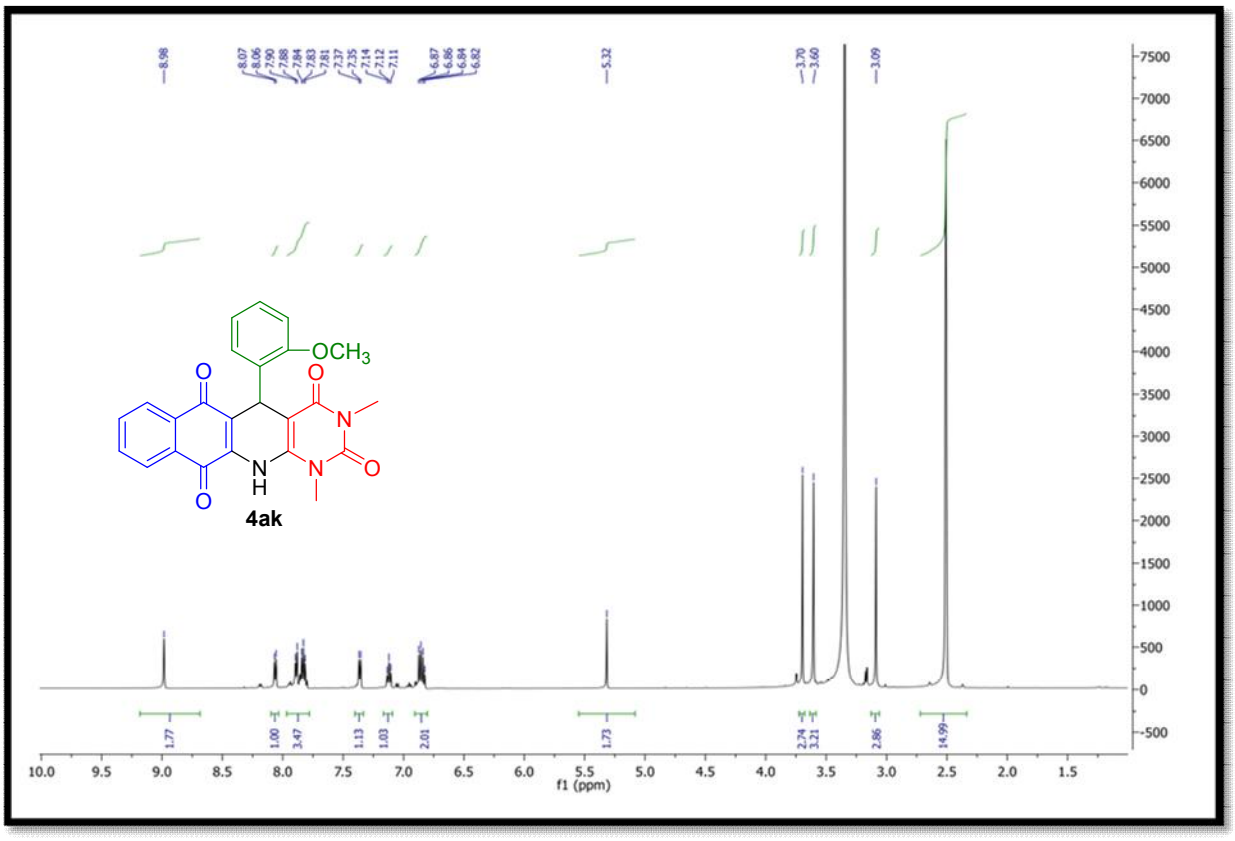


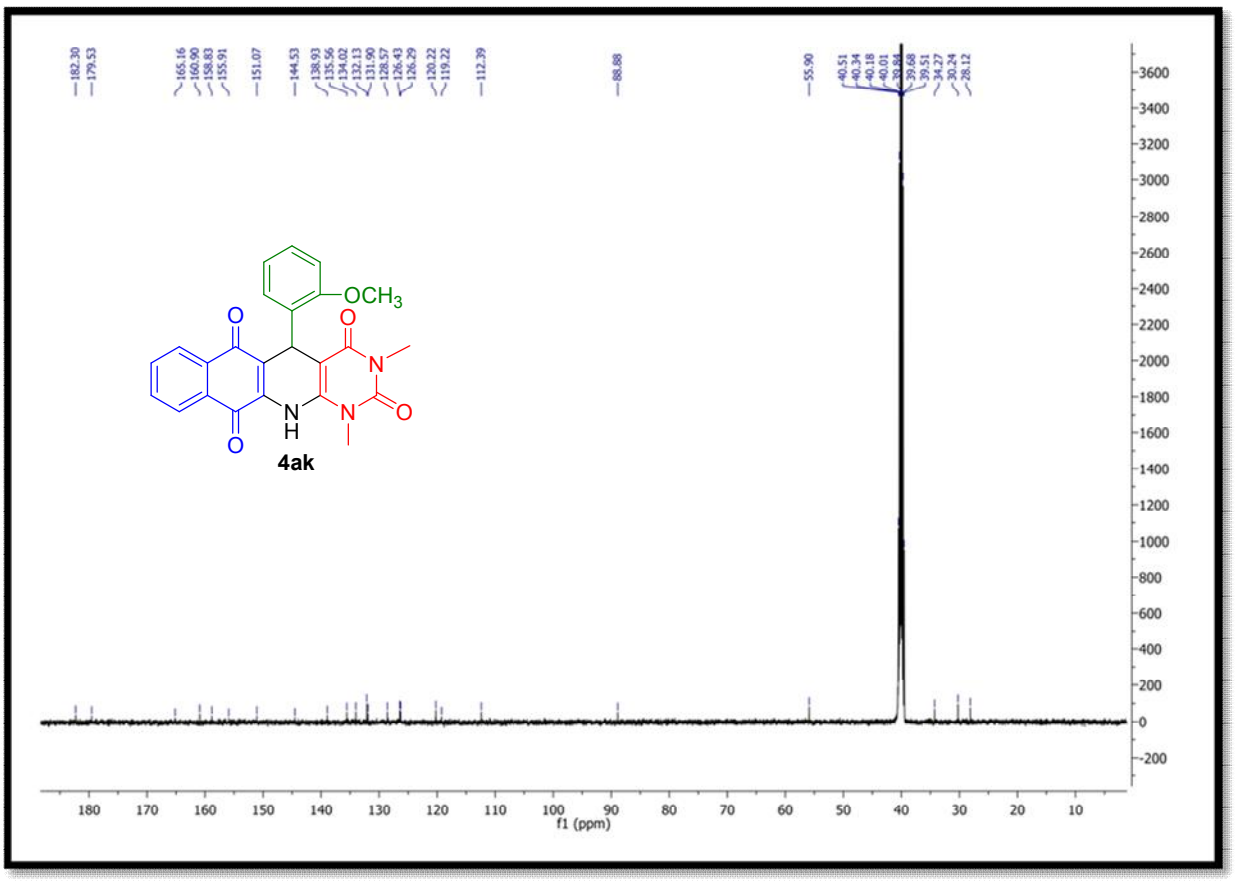
^{1}H & ^{13}C NMR Spectra of compound **4aj**



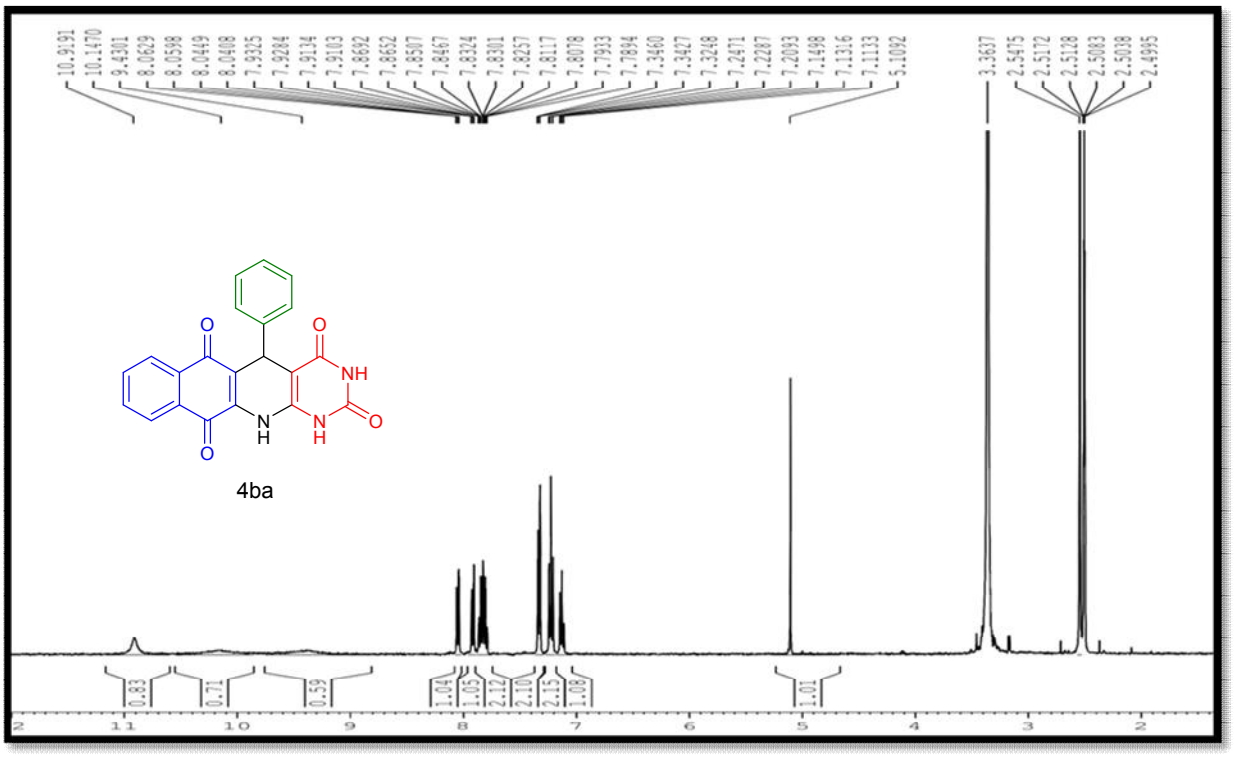


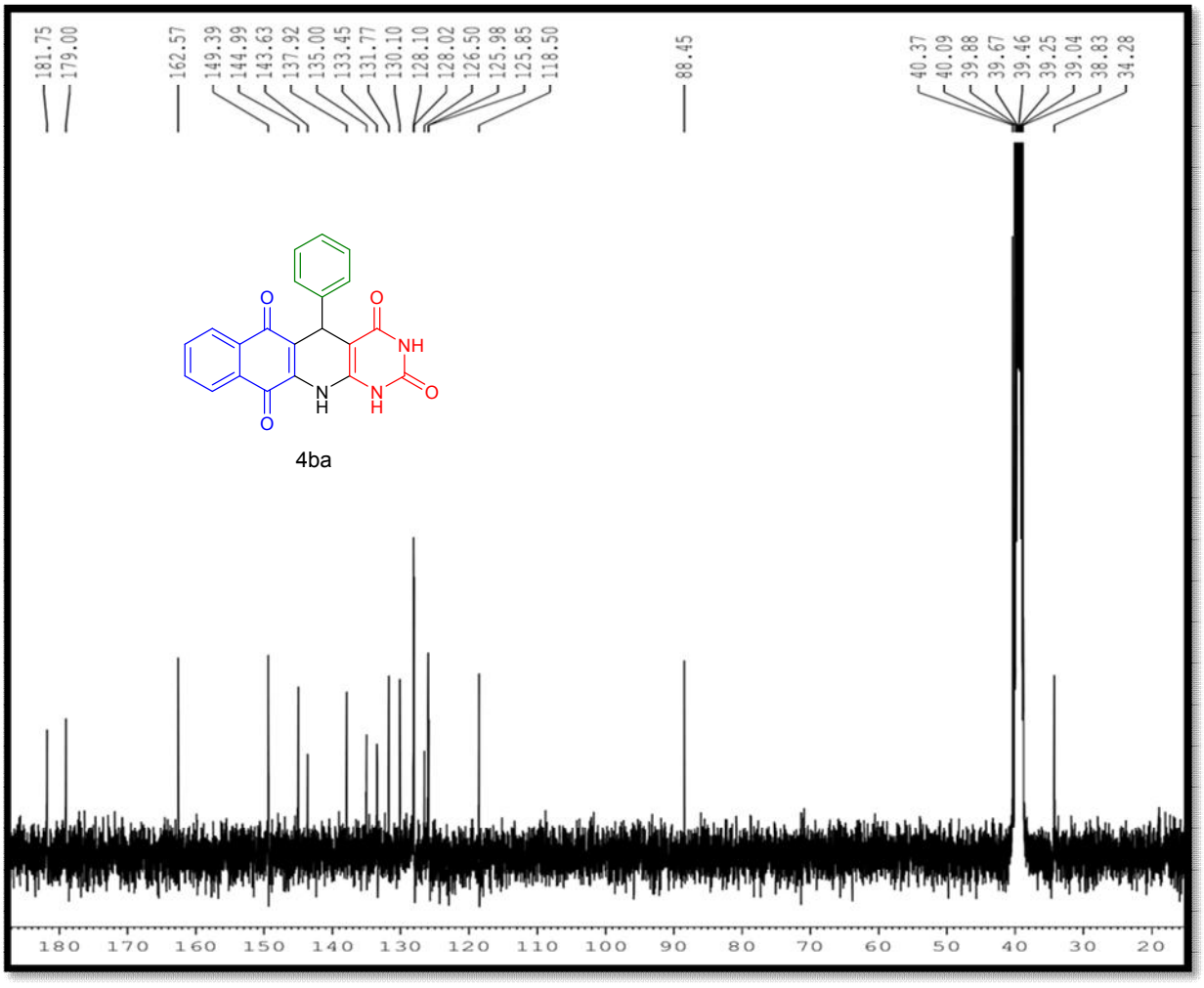
^{1}H NMR Spectra of compound **4ak**



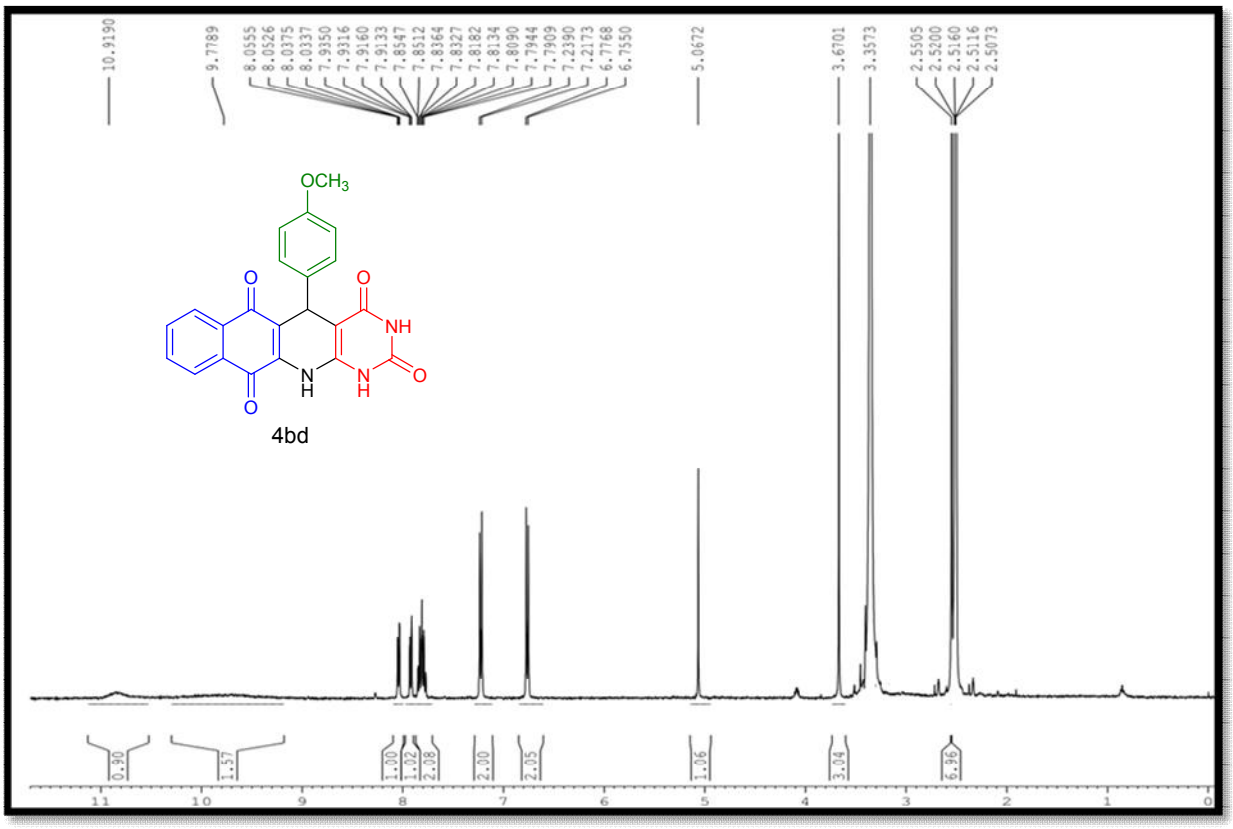


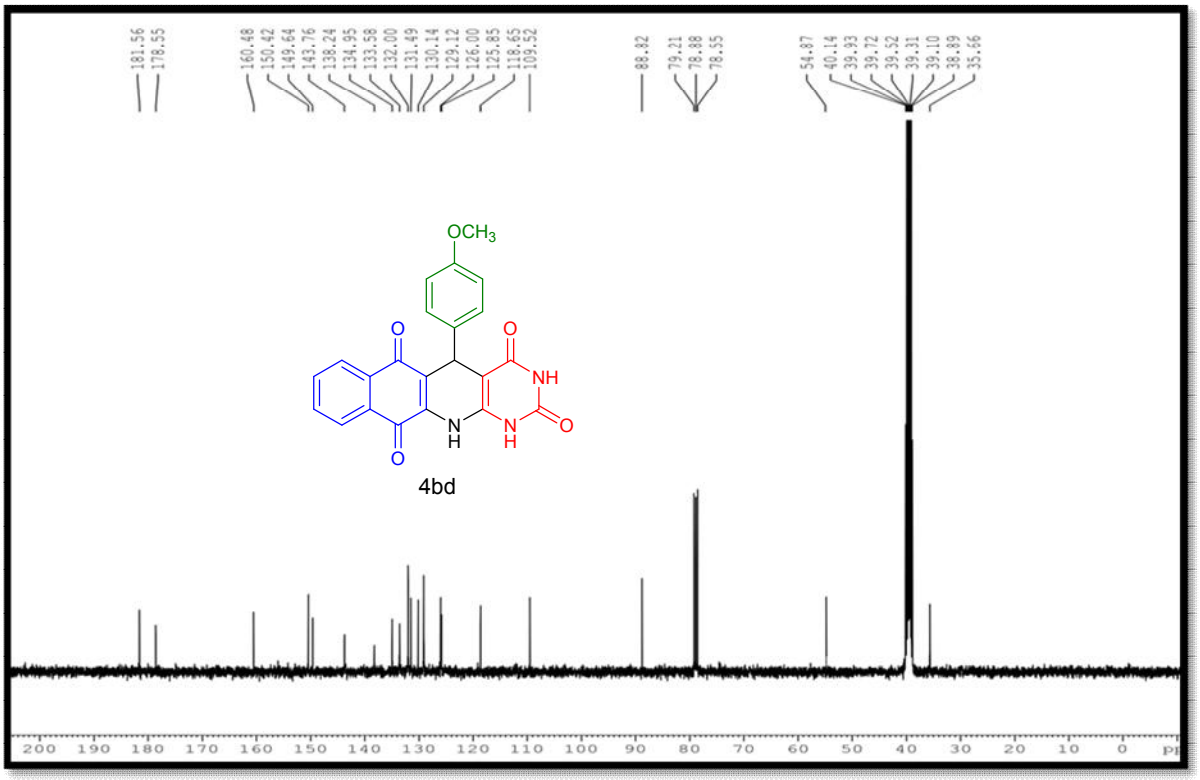
^{1}H & ^{13}C NMR Spectra of compound **4ba**



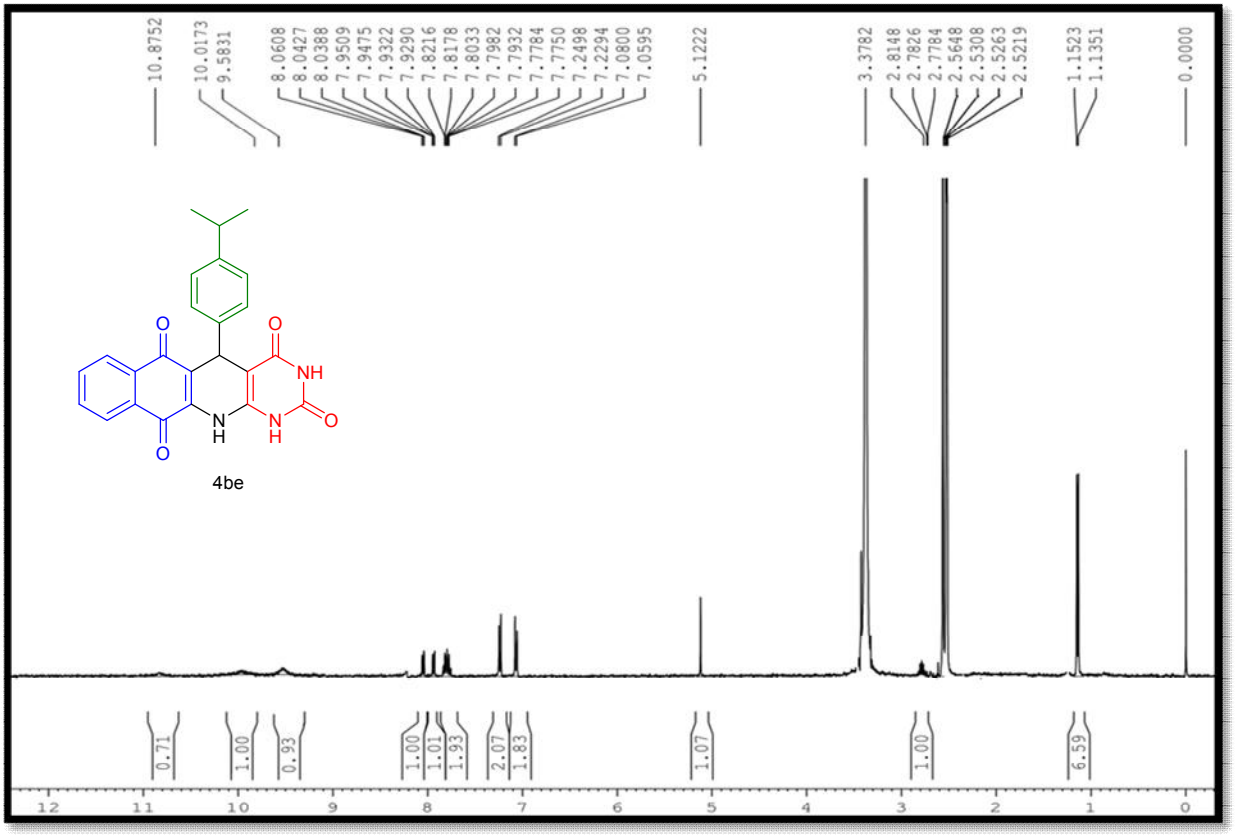


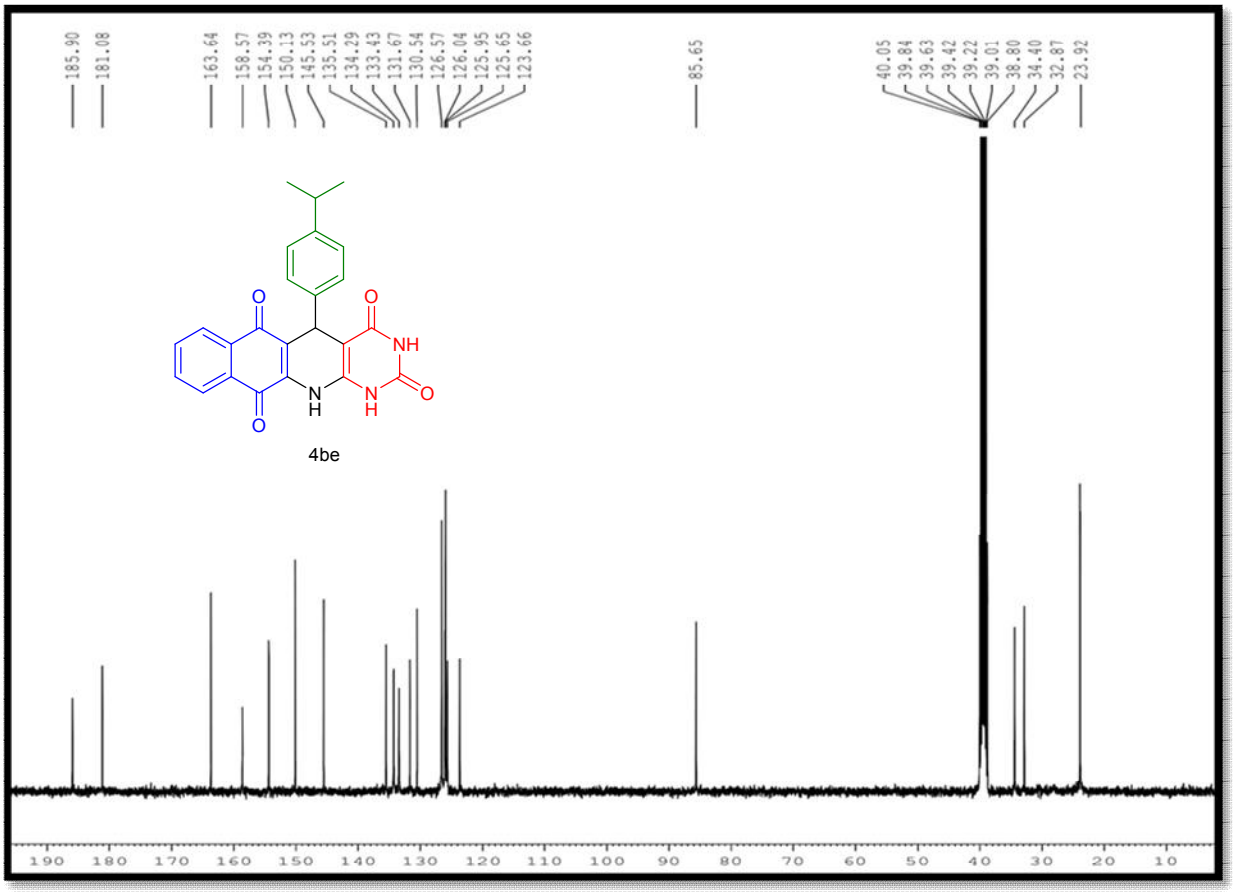
^{1}H & ^{13}C NMR Spectra of compound **4bd**



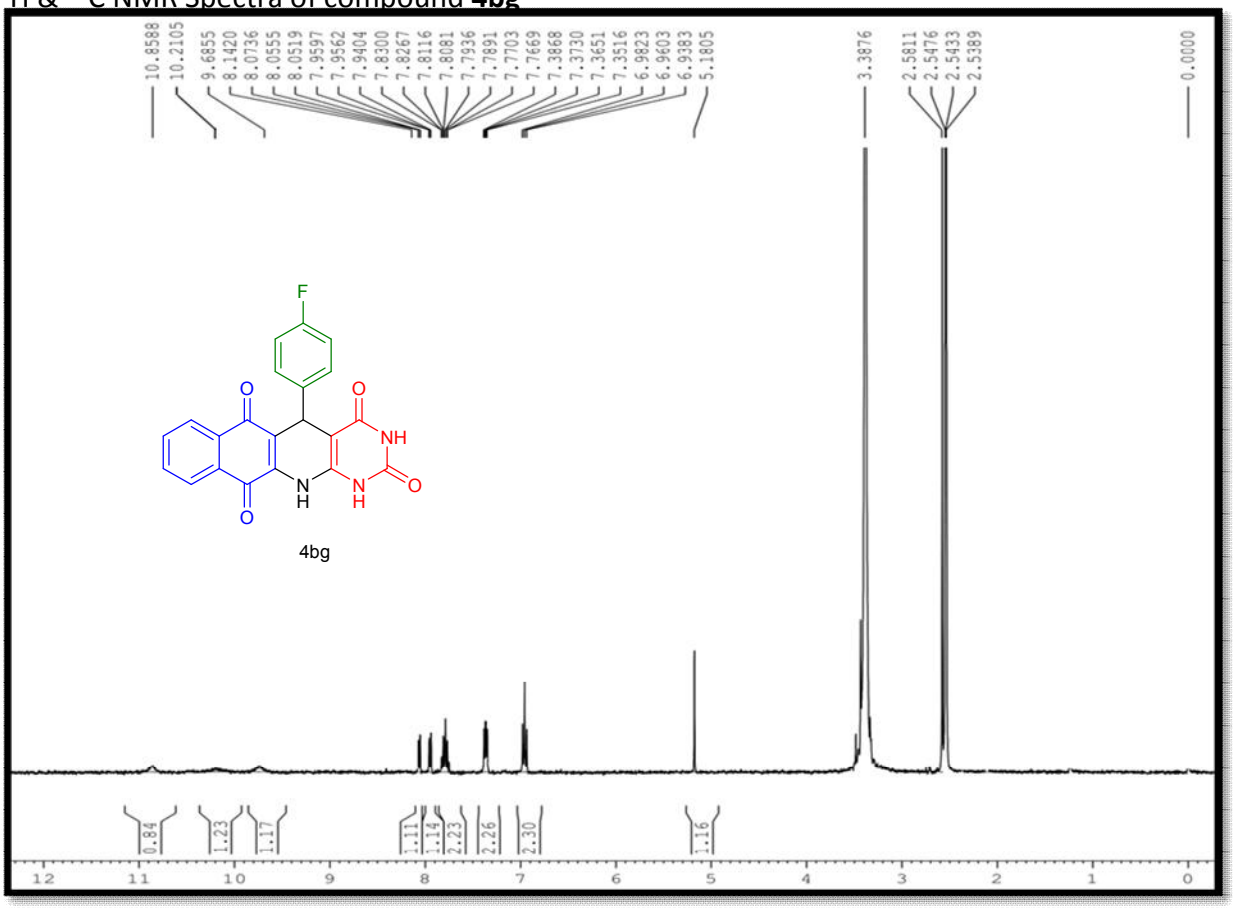


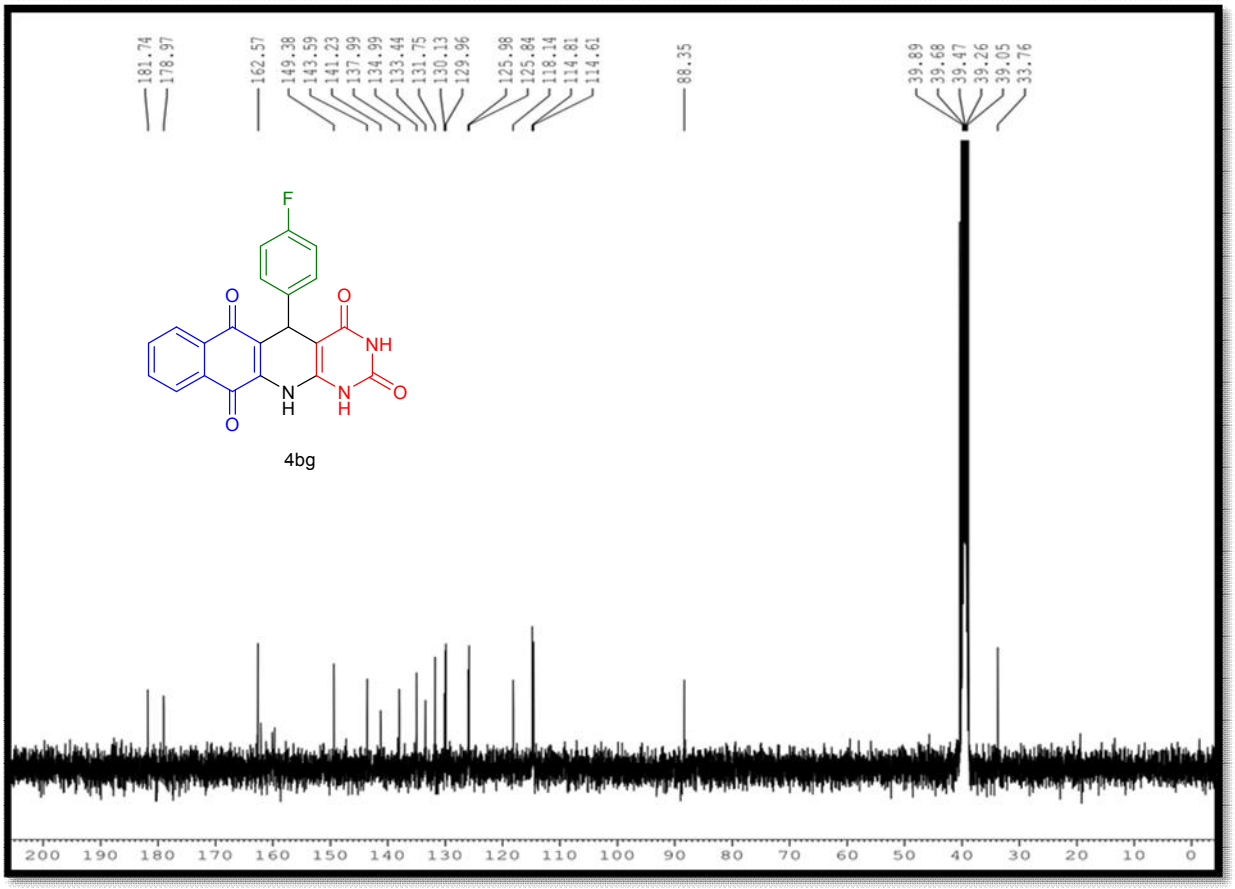
^{1}H & ^{13}C NMR Spectra of compound **4be**



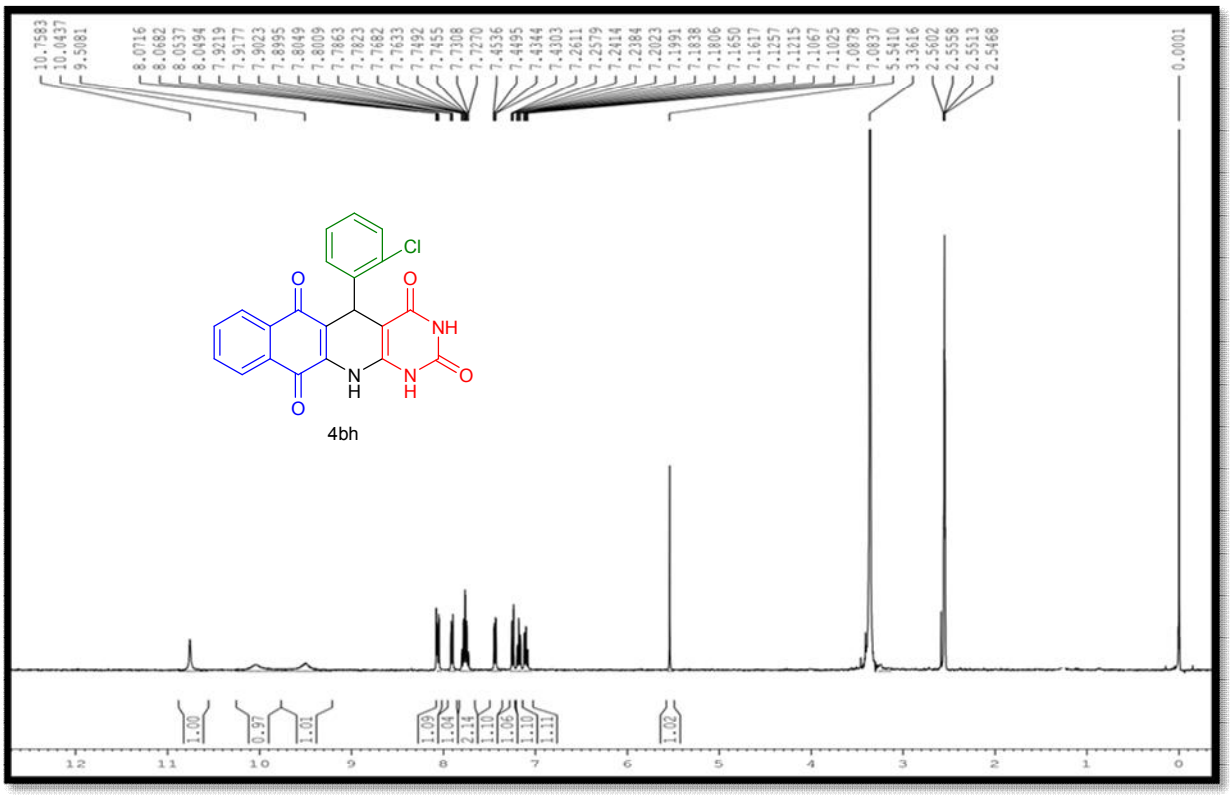


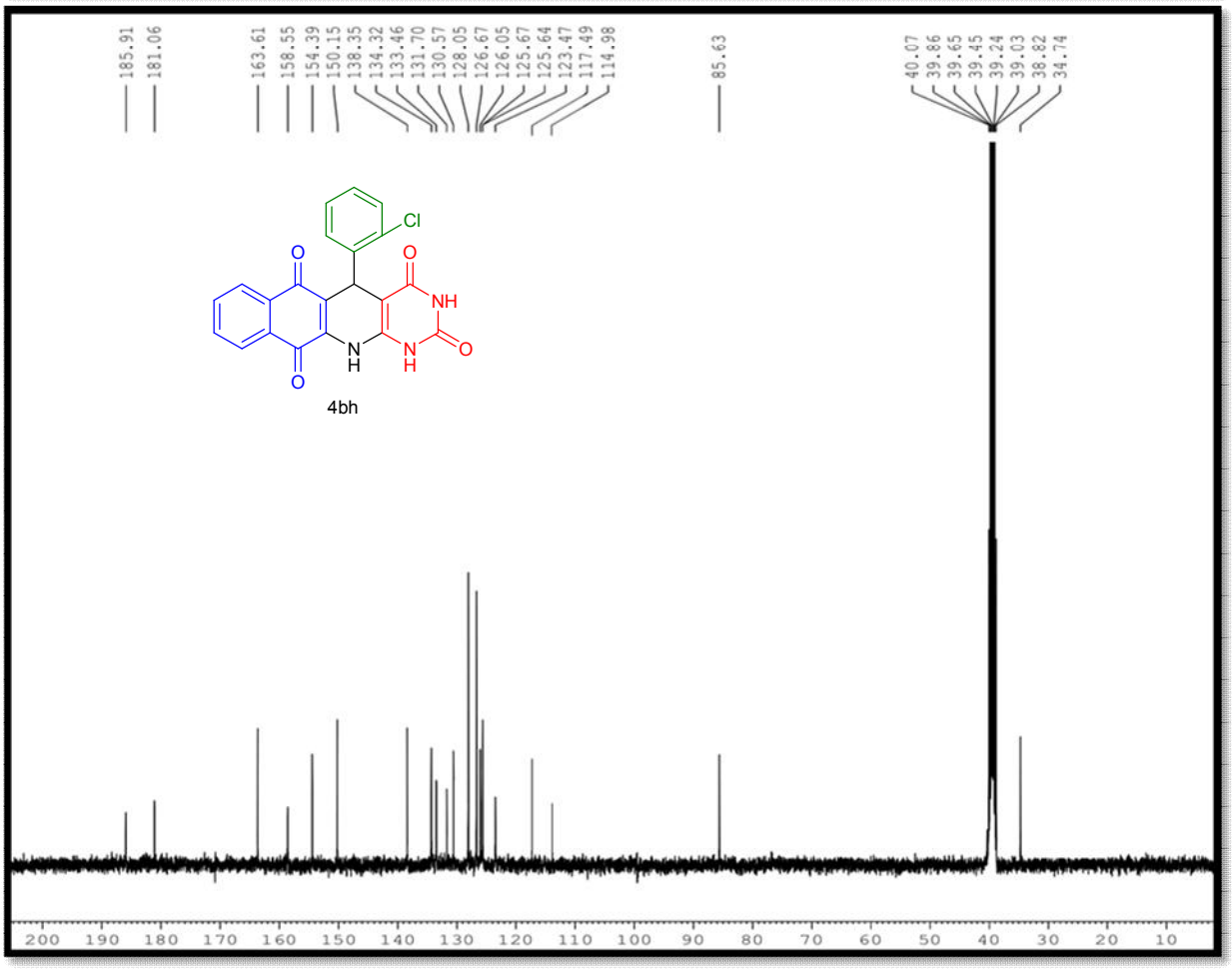
^{1}H & ^{13}C NMR Spectra of compound **4bg**



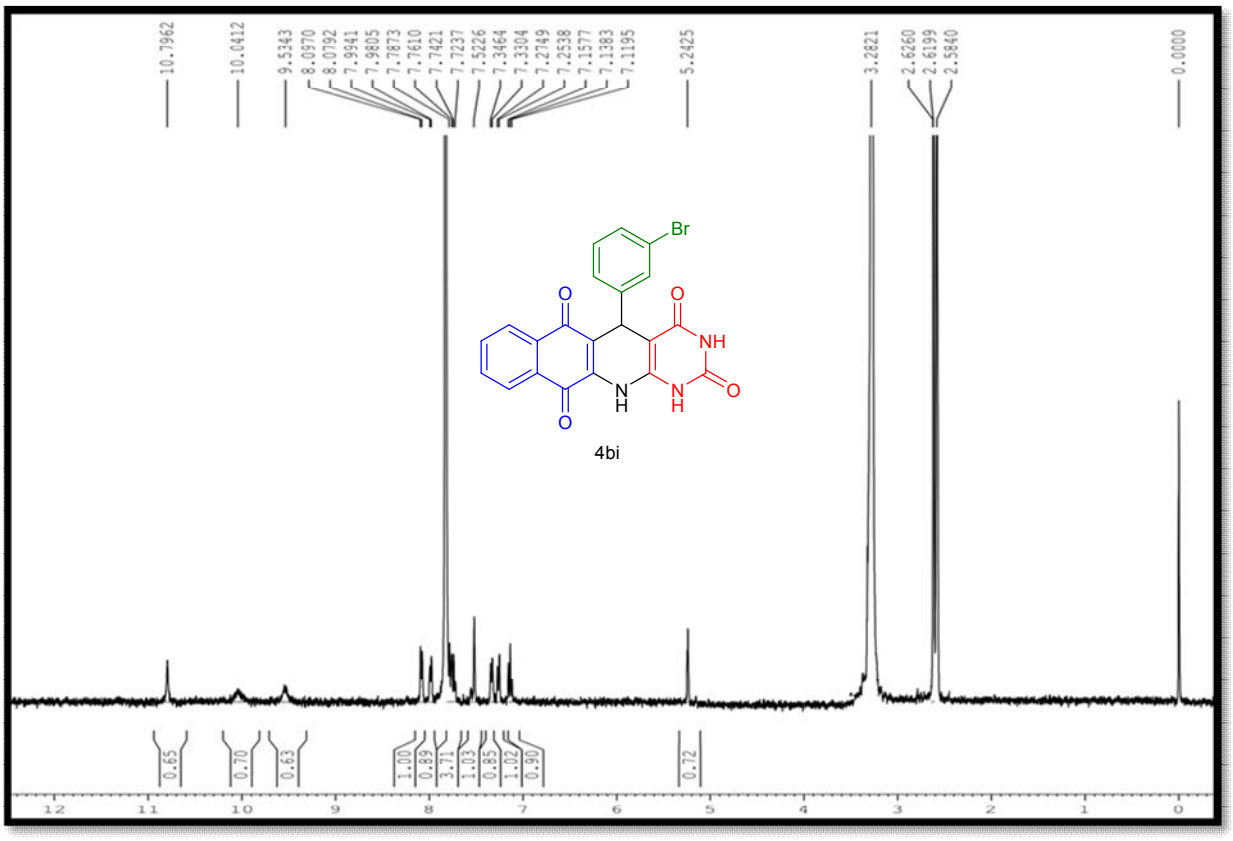


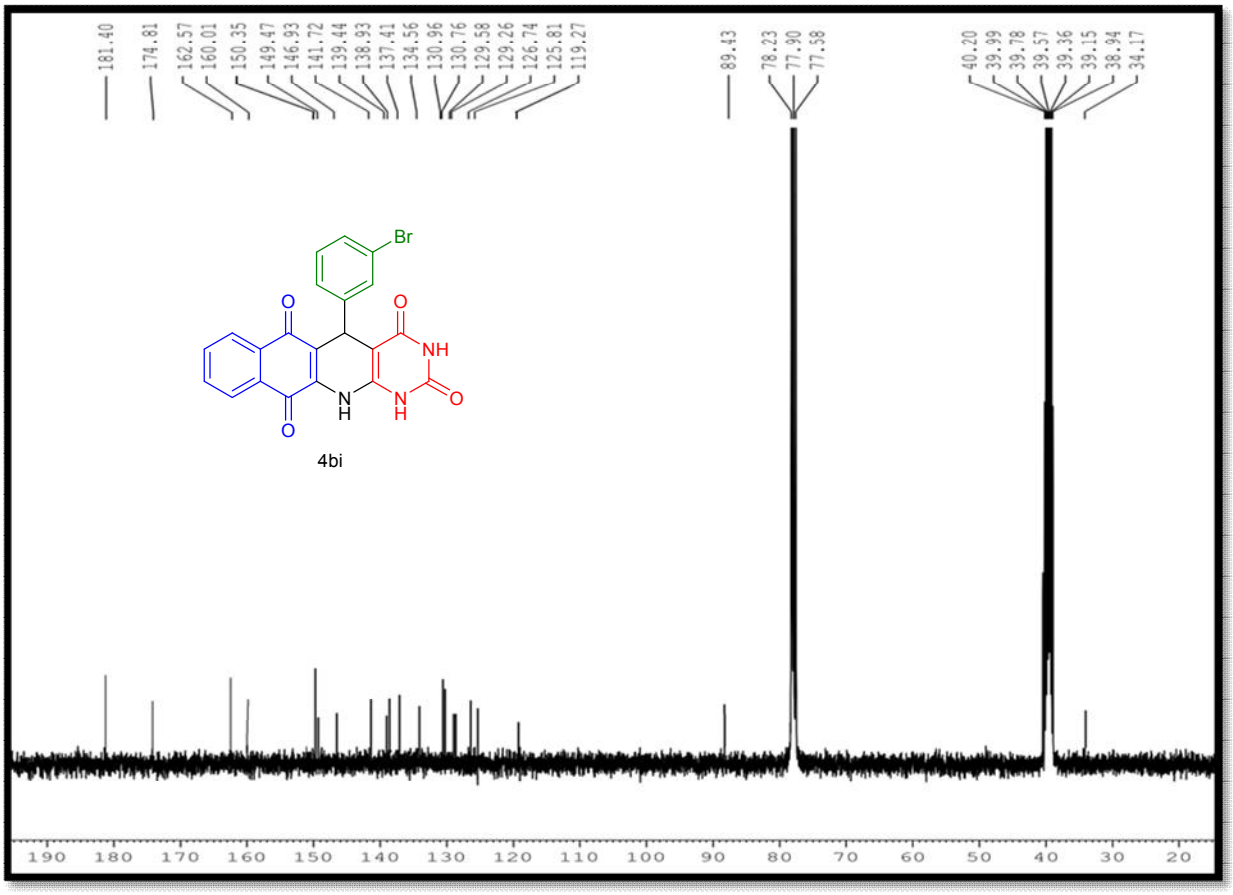
^{1}H & ^{13}C NMR Spectra of compound **4bh**



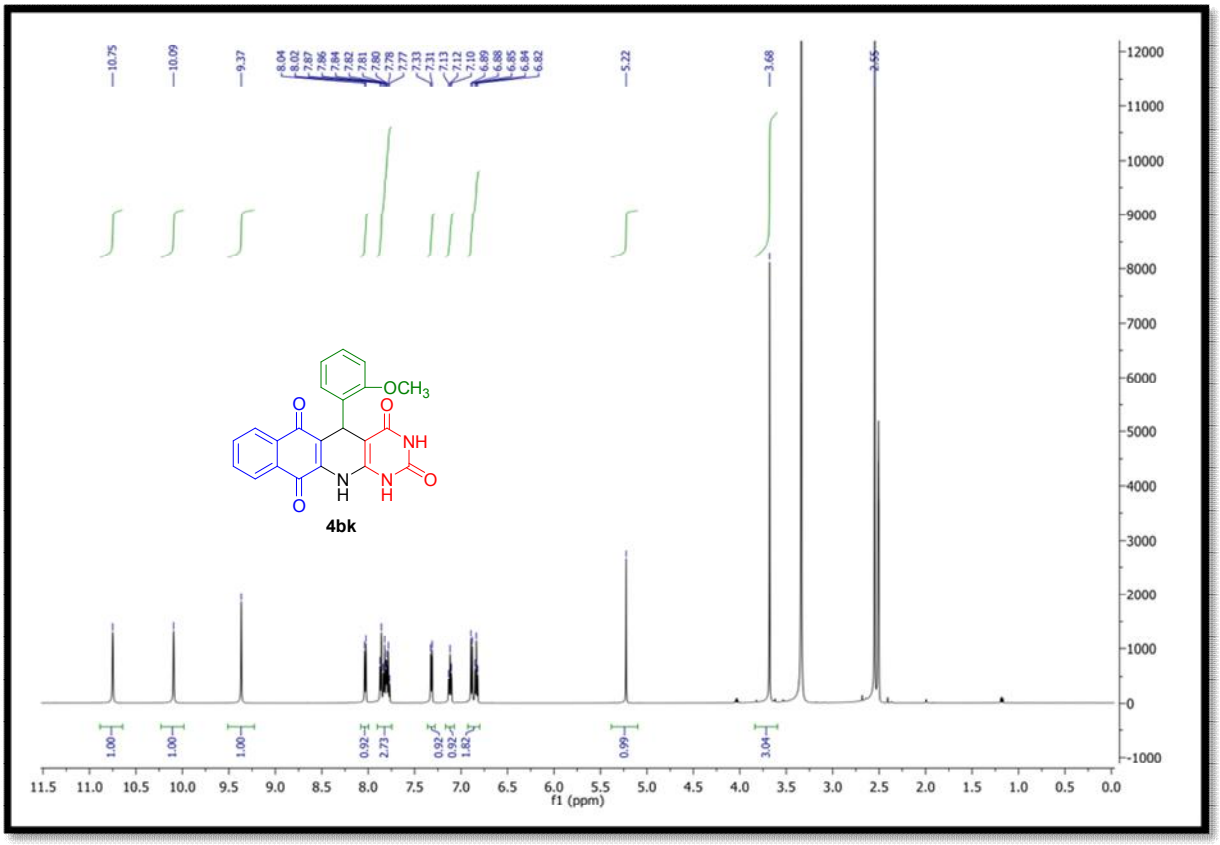


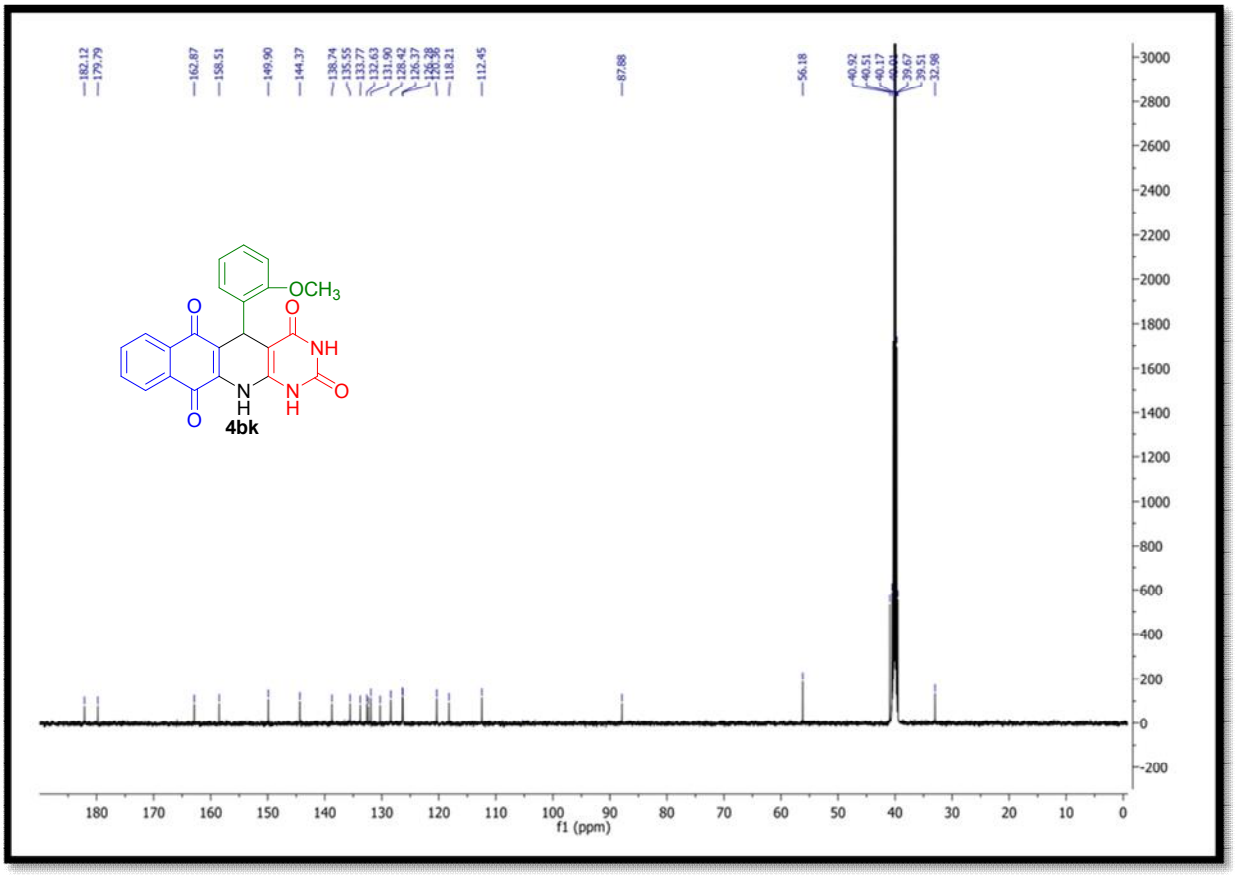
^{1}H & ^{13}C NMR Spectra of compound **4bi**



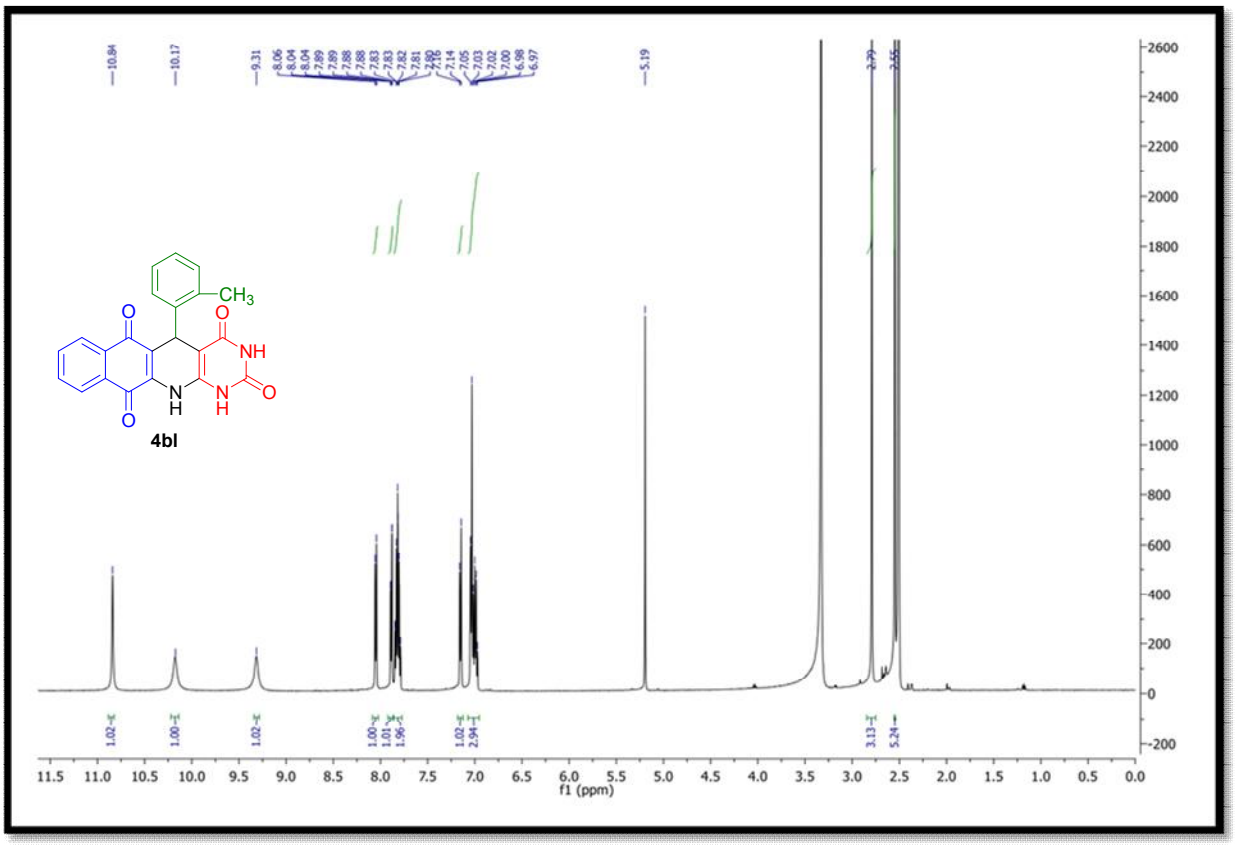


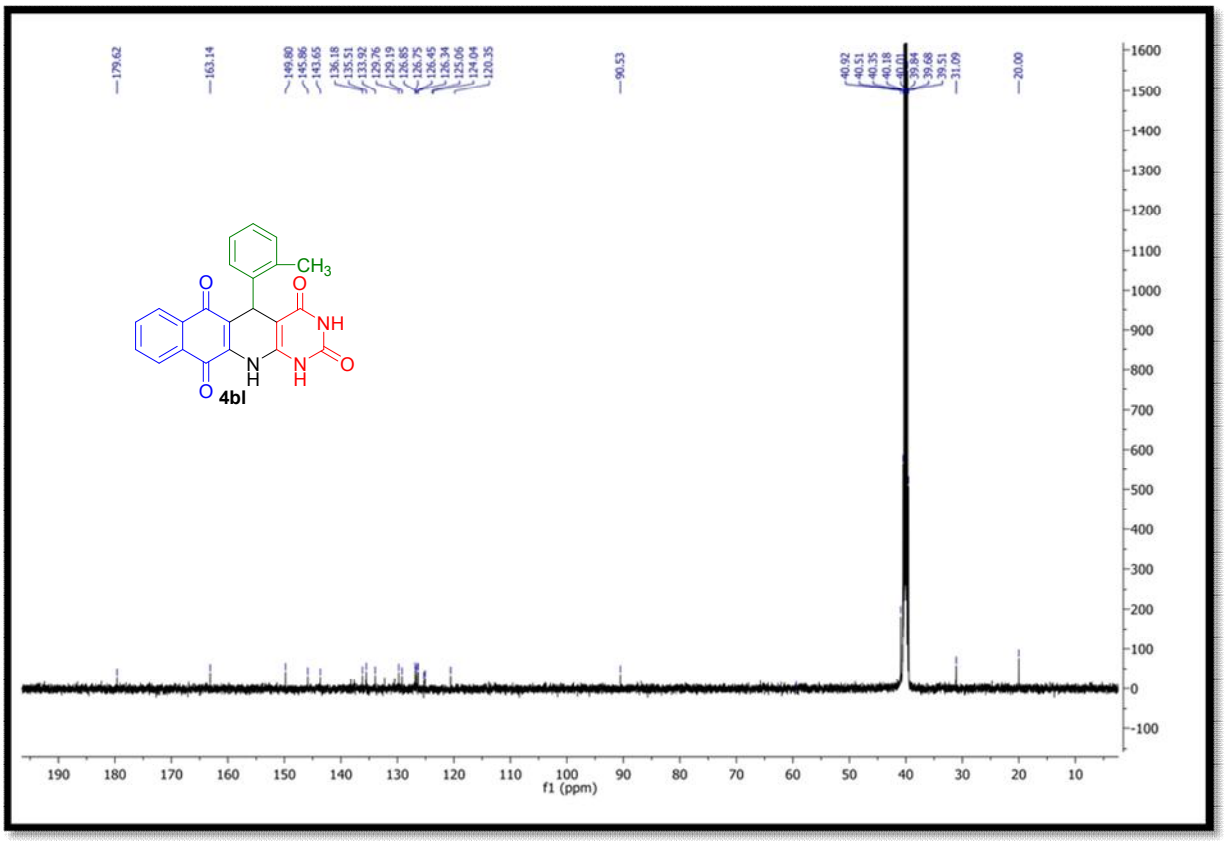
^{1}H &^{13}C NMR of compound **4bk**



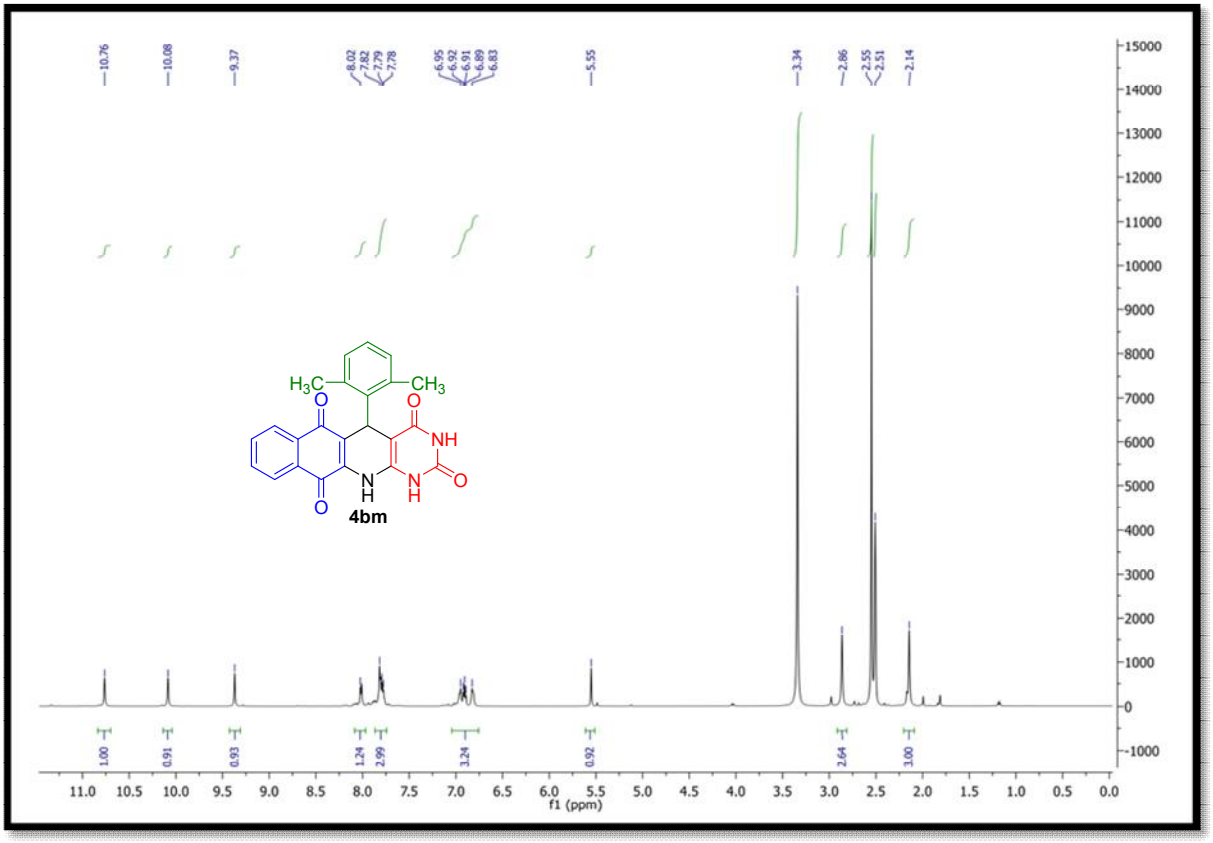


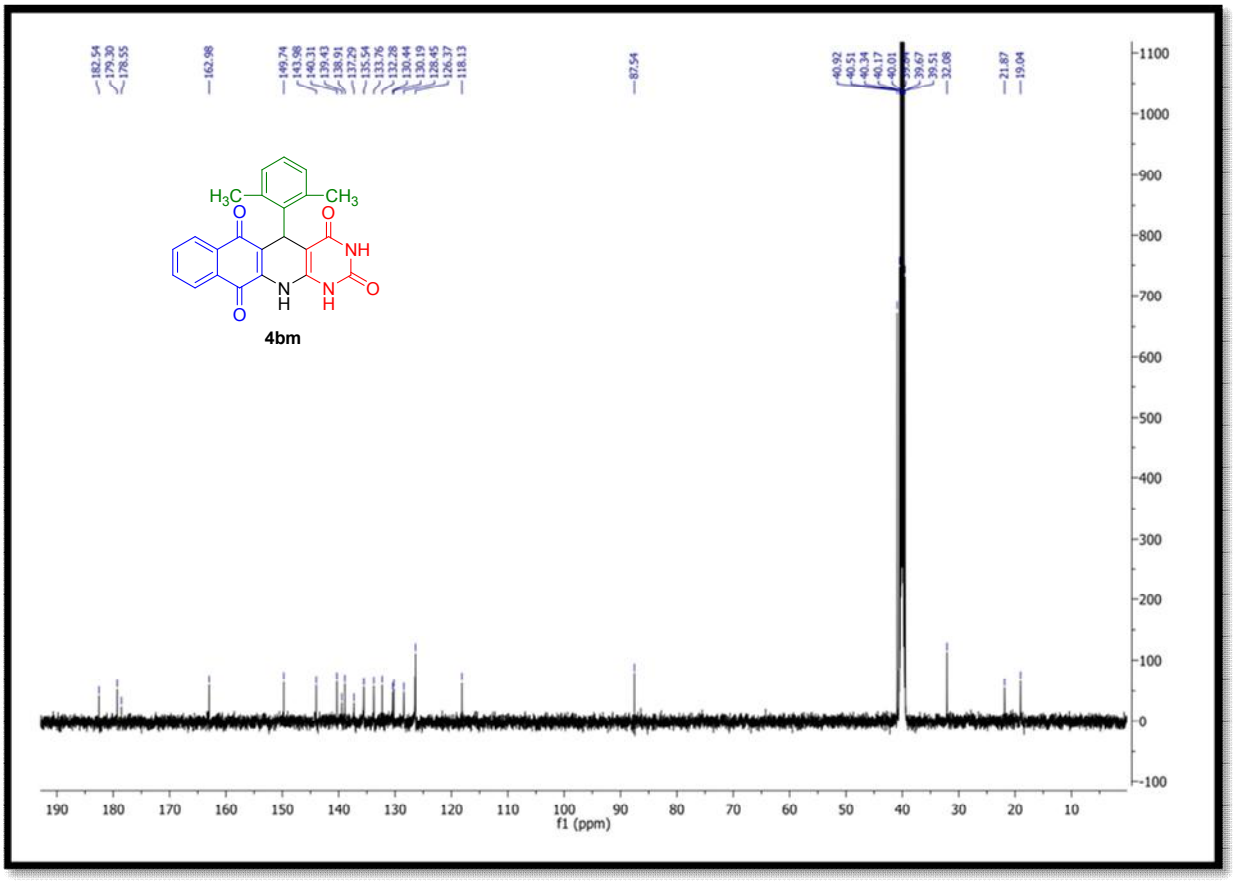
^{1}H &^{13}C NMR of compound **4bl**



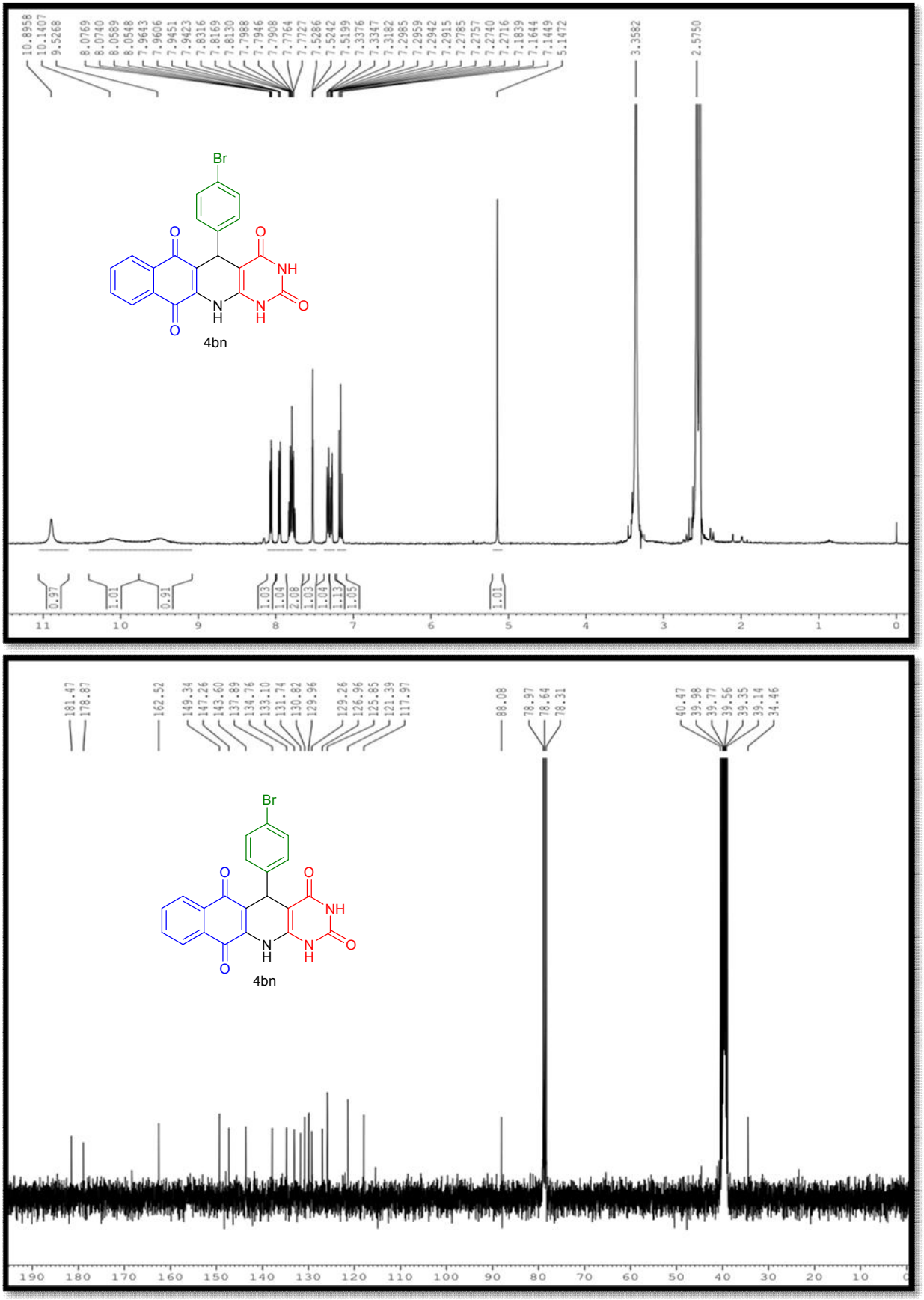


^{1}H &^{13}C NMR of compound **4bm**

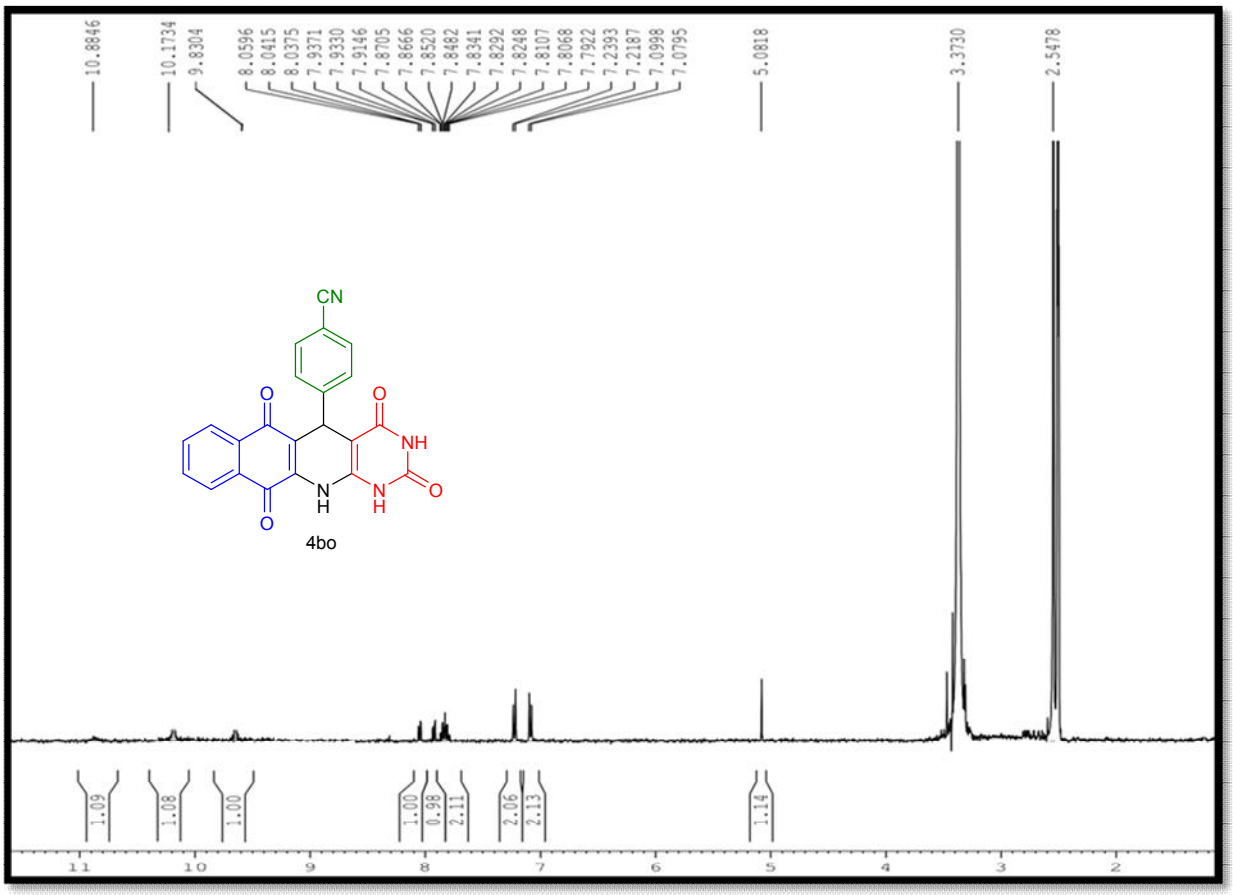


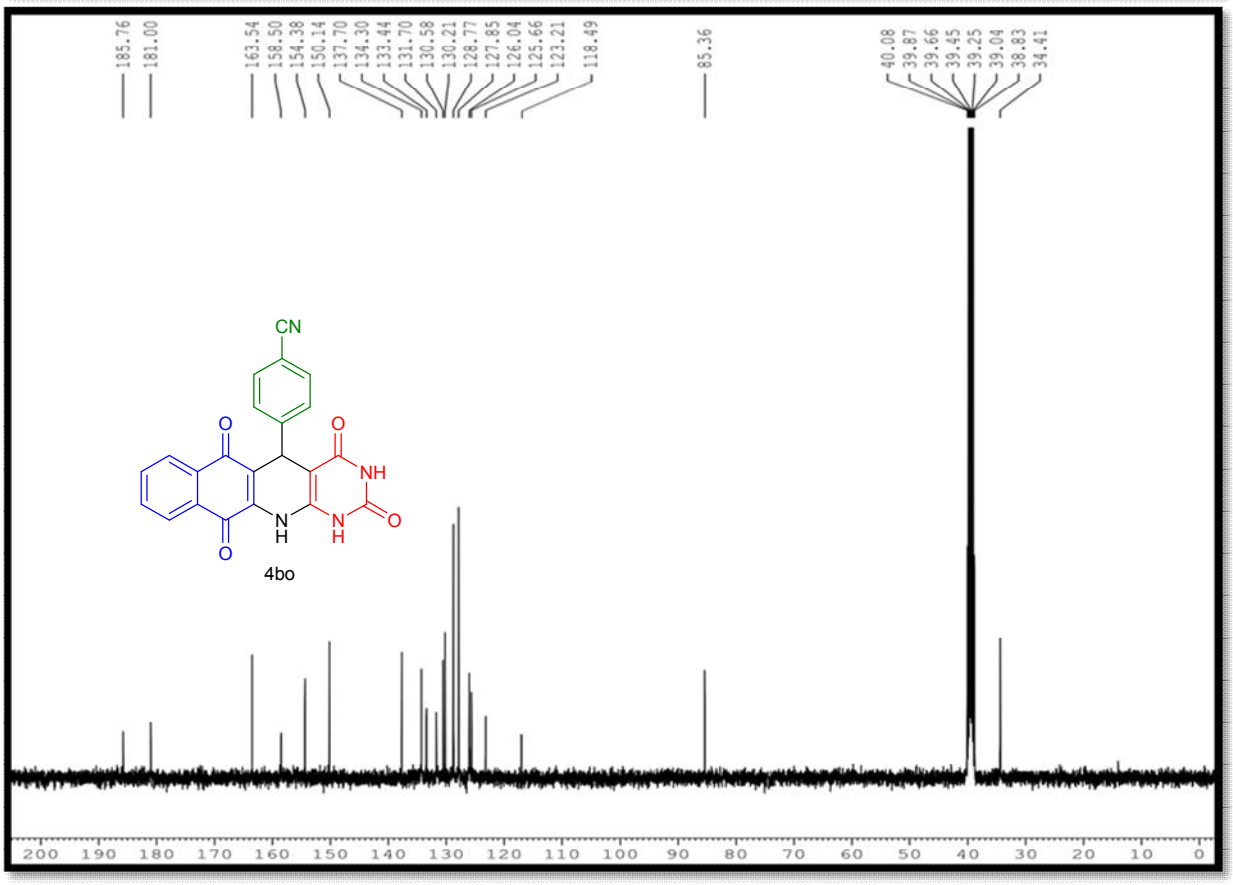


^{1}H & ^{13}C NMR Spectra of compound **4bn**

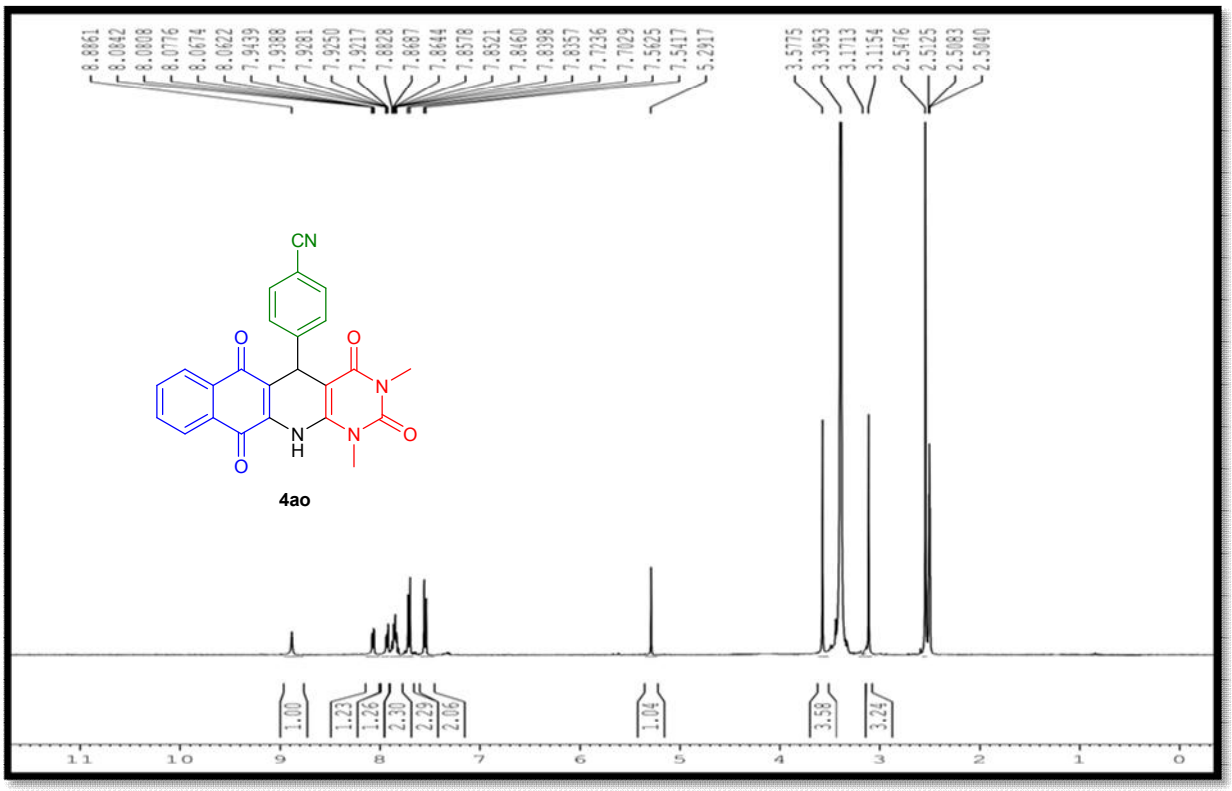


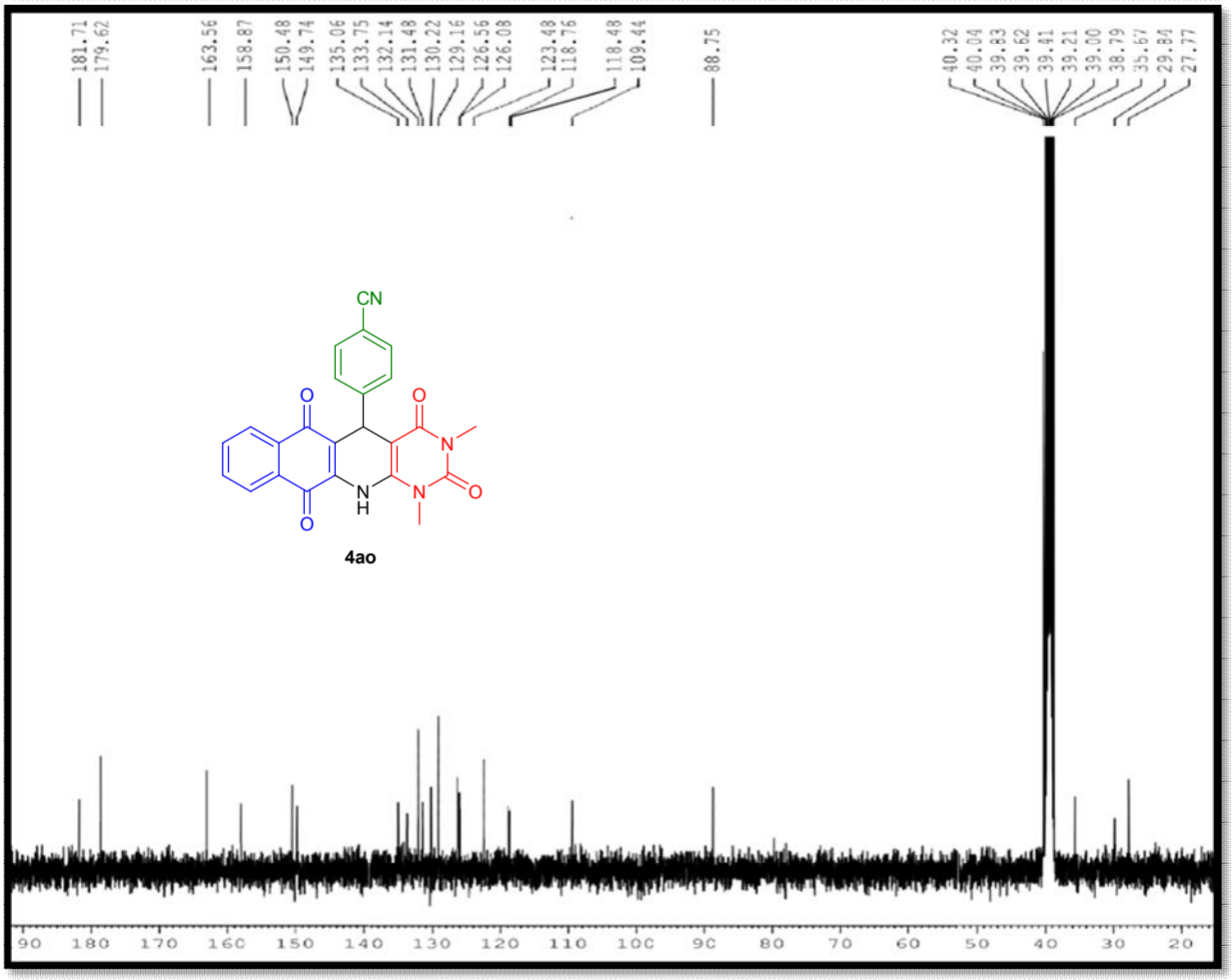
^{1}H & ^{13}C NMR Spectra of compound **4bo**



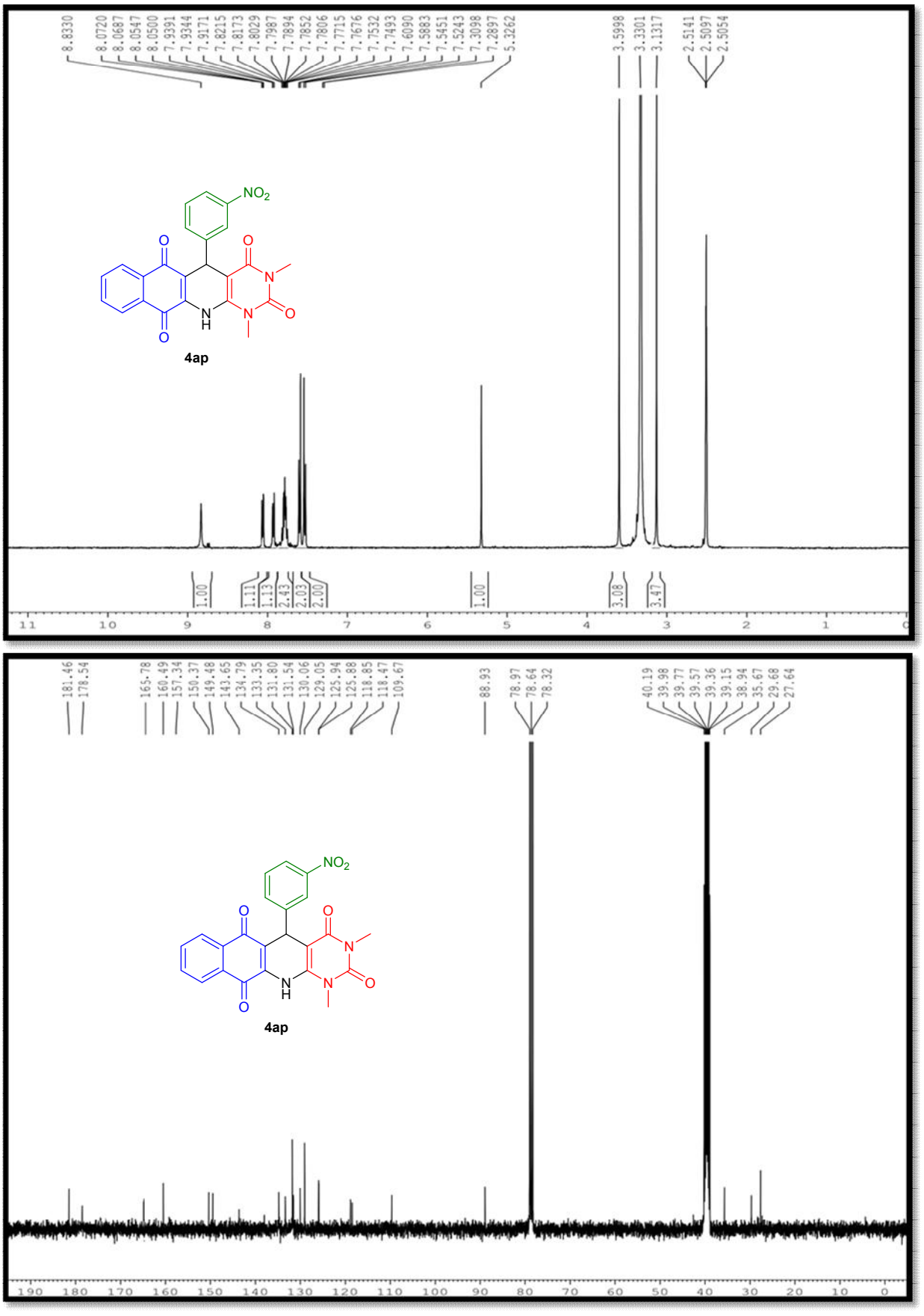


^{1}H & ^{13}C NMR Spectra of compound **4ao**

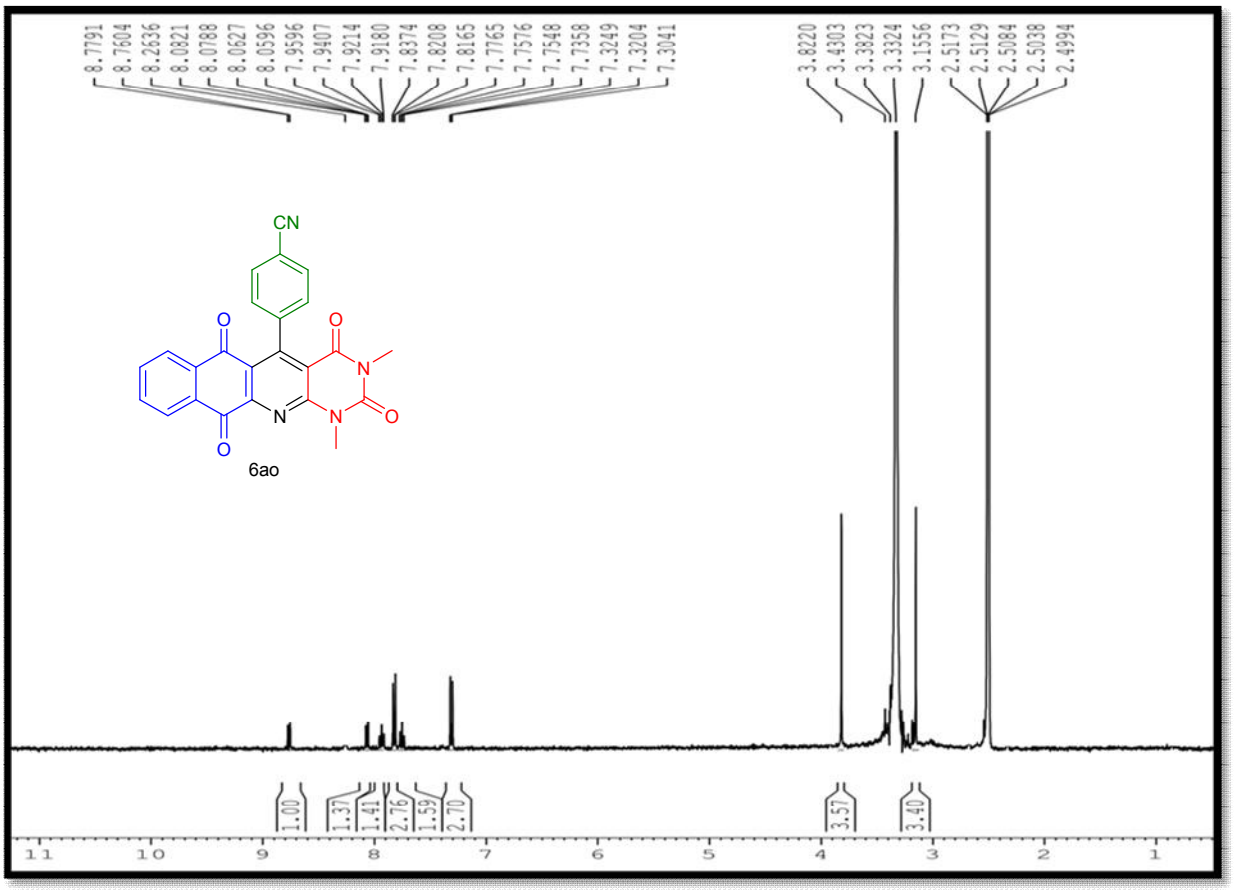


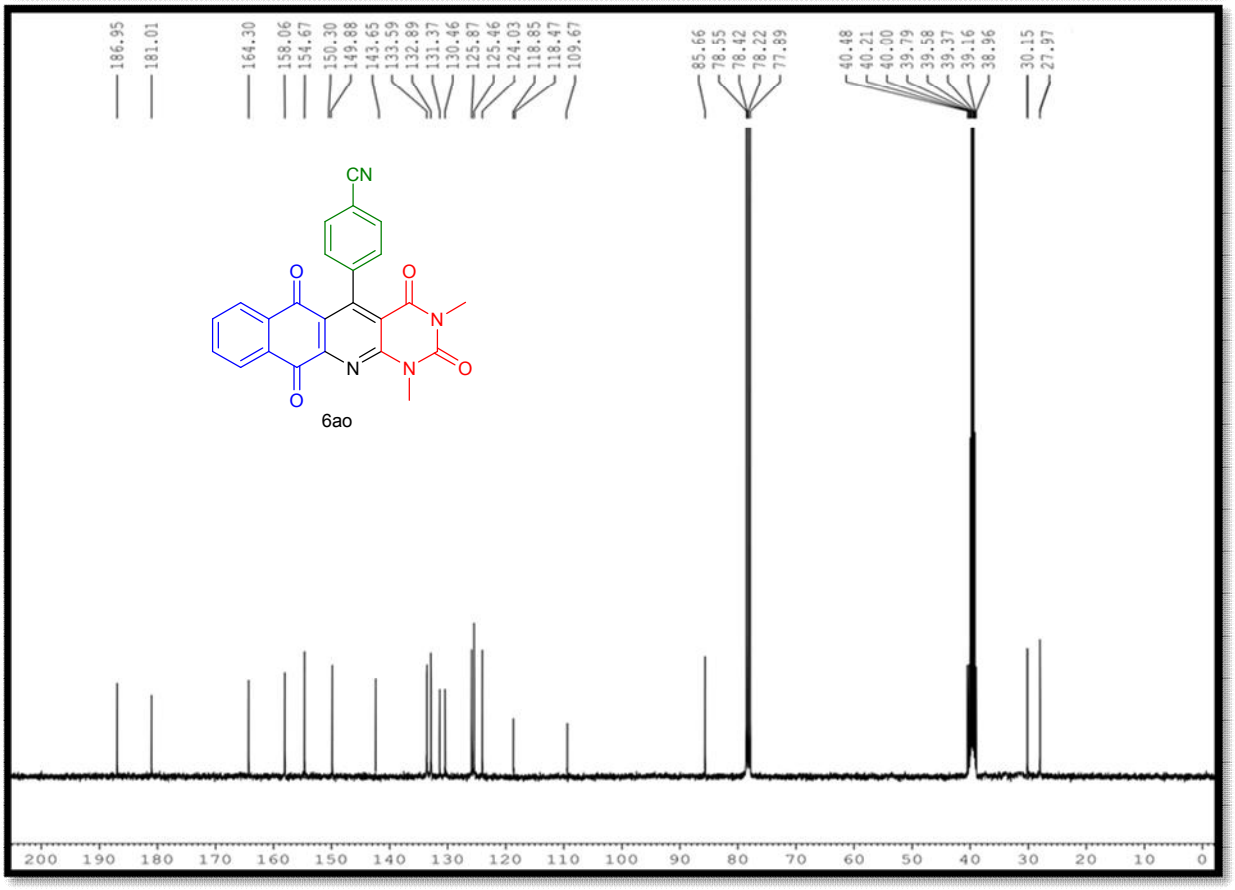


^{1}H & ^{13}C NMR Spectra of compound **4ap**

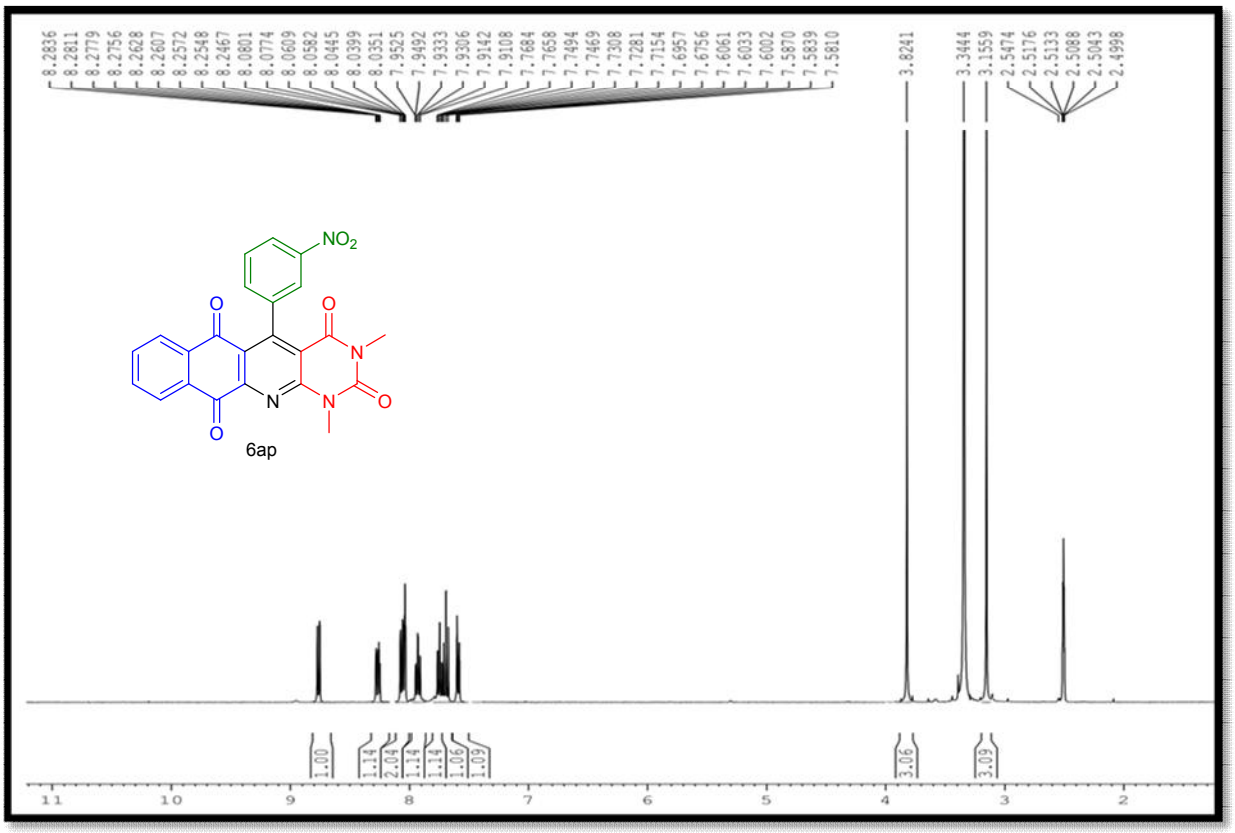


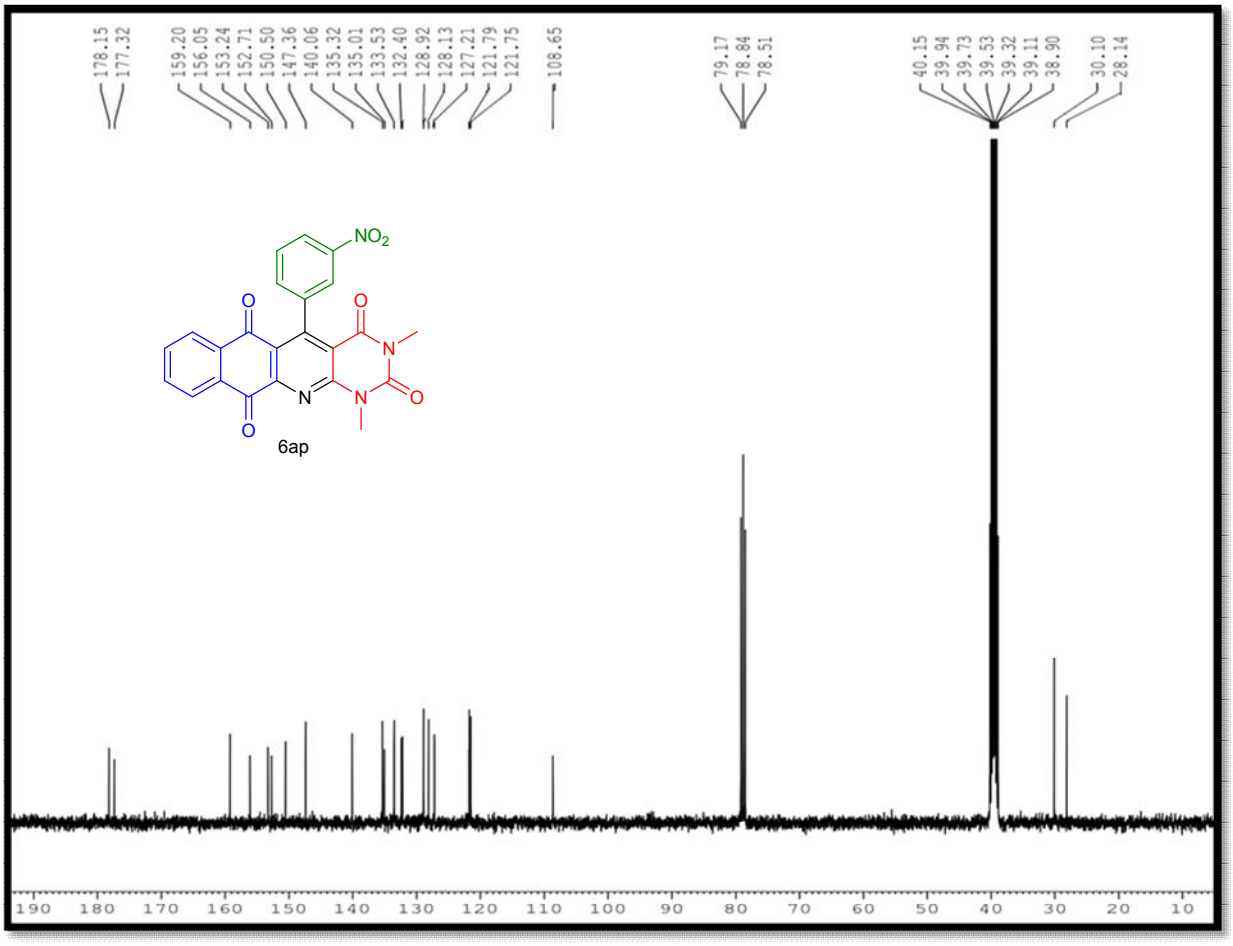
^{1}H & ^{13}C NMR Spectra of compound **6ao**





^{1}H & ^{13}C NMR Spectra of compound **6ap**





^{1}H & ^{13}C NMR Spectra of compound **7as**