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Supporting Information

Synthesis and characterization of Bi(III) immobilized on triazine dendrimer-stabilized magnetic nanoparticles: a reusable catalyst for synthesis of aminonaphthoquinones and *bis*-aminonaphthoquinones

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1. General

The chemicals used in this work were purchased from Fluka and Merck chemical companies. Melting points were determined with a Stuart Scientific SMP2 apparatus. FT-IR spectra were recorded on a Nicolet-Impact 400D spectrophotometer. ^{1}H and ^{13}C NMR (400 and 100 MHz) spectra were recorded on a Bruker Avance 400 MHz spectrometer using DMSO-*d6* as solvent. Elemental analysis was performed on a LECO, CHNS-932 analyzer. Thermogravimetric analysis (TGA) was carried out on a Mettler TG50 instrument under air flow at a uniform heating rate of 20 ˚C min^{-1} in the range 30-600 ˚C. The TGA instrument was re-calibrated at frequent intervals with standards; the accuracy was always better than ±2.0%. The scanning electron microscope measurement was carried out on a Hitachi S-4700 field emission-scanning electron microscope (FE-SEM). The transmission electron microscopy (TEM) was carried out on a Philips CM10 instrument operating at 100 kV. The Bi content of the catalyst was determined by a Jarrell-Ash 1100 ICP analyzer. X-Ray diffraction (XRD) images were obtained with a Bruker XRD D8 Advance instrument with Co Kα radiation at 40 kV. The magnetic measurements were performed with a vibrating sample magnetometer (VSM) at Meghnatis Daghigh Kavir Co. The X-ray photoelectron spectroscopy (XPS) measurements were performed using a Gammadata-scienta ESCA200 hemispherical analyzer equipped with an Al (K**α** = 1486.6 eV) X-ray source.

2. Preparation of Bi(III) immobilized on triazinedendrimer-stabilized magnetic nanoparticles (Fe_3O_4@TDSN-Bi(III))

Preparation of $(DEA)_3T$

In a 50 mL round-bottomed flask, a mixture of cyanuric chloride (0.92 g, 5 mmol) and diethanolamine (1.73 g, 16.5 mmol) in 20 mL THF was stirred for 1 h at room temperature. DIPEA (8.61 mL, 49.6 mmol) was added and the resulting mixture was stirred for 1 h at room temperature and then 16 h at 80-85 ˚C. The progress of the reaction was monitored by TLC (eluent: CH_3OH/CH_2Cl_2; 1:10). After completion of the reaction, the solvent was evaporated to give a white solid with excellent yield (99%) and purity.

Preparation of G1

First, the MNPs was prepared according to the reported procedure.[1] In this manner, $FeCl_3 \cdot 6H_2O$ (11.0 g) and $FeCl_2 \cdot 4H_2O$ (4.0 g) were dissolved in 250 mL deionized water under N_2 with vigorous stirring at 85 ˚C, during which the pH of the solution adjusted to 9 with conc. $NH_3 \cdot H_2O$. The mixture was further stirred for 4 h and the resulting MNPs precipitates were washed with deionized water and ethanol until the pH reached 7. The black precipitate (MNPs) was collected with a permanent magnet. For coating of MNPs surface with triazine dendrimer, the MNPs (150 mg) in 7 mL EtOH/DMF (1:1) were sonicated for 5 min at room temperature, then DIPEA was added dropwise for a period of 10 min until the pH reached 9. Finally, $(DEA)_3T$ (50 mg) was added and the resulting mixture was dispersed under intense sonication for 8 h. The black precipitate (G1) was collected using a permanent magnet, washed several times with ethanol and acetone, and dried in a vacuum oven at 50˚C.

Preparation of CC2

The resulting G1 was dispersed in 10 mL THF by sonication. Then, cyanuric chloride (1.28 mmol, 0.23 g) and DIPEA (1.92 mmol, 0.33 mL) were added and the mixture was sonicated overnight under N_2 at room temperature. Finally, the black precipitate (CC2) was separated by a permanent magnet, washed with THF (2×10 mL) and acetone (10 mL),and dried in a vacuum oven at 50 ˚C.

Preparation of Fe_3O_4@TDSN (G2)

The obtained CC2 was dispersed in 10 mL CH_3CN by sonication at 70 ˚C. While the reaction mixture was vigorously stirring, diethanolamine (1.2 mmol, 0.12 mL) and DIPEA (3.6 mmol, 0.66 mL) were added and the mixture was stirred overnight at 70 ˚C. The resulting black precipitate (G2) was separated by a permanent magnet, washed with ethanol (2×10 mL) and acetone (10 mL) and dried in a vacuum oven at 50 ˚C.

3. Preparation of Fe_3O_4@TDSN-Bi(III)

A mixture of G2 (0.10 mmol g^{-1} Fe_3O_4) and $Bi(OTf)_3$ (2.0 mmol, 1.31 g) (dendrimer/metal; 1:20) was dispersed in THF (10 mL) under intense sonication and then the reaction mixture was shaken overnight at room temperature. The resulting solid material was separated by a permanent magnet, washed with *n*-hexane three times and dried under vacuum to afford Fe_3O_4@TDSN-Bi(III) catalyst as a black solid.

4. General procedure for synthesis of aminonaphthoquinone derivatives catalyzed by Fe_3O_4@TDSN-Bi(III)

A mixture of lawsone (1 mmol) aldehyde (1 mmol), amine (1 mmol) and Fe_3O_4@TDSN-Bi(III) (3 mol%, 50 mg) in EtOH (3 mL) was stirred at room temperature for the appropriate time according to Table 4. After completion of the reaction as indicated by TLC (eluent: petroleum ether/EtOAc, 1:1), the catalyst was easily separated by a permanent magnet and washed with EtOH (5 mL).The organic residue was filtered, washed with cold ethanol for several times and dried under vacuum to give the pure product.

5. General procedure for synthesis of symmetric *bis*-aminonaphthoquinones catalyzed by Fe_3O_4@TDSN-Bi(III)

A mixture of lawsone (1 mmol), dialdehyde (0.5 mmol), amine (1 mmol) and Fe_3O_4@TDSN-Bi(III) (3 mol%, 50 mg) in EtOH (3 mL) was stirred at room temperature for the appropriate time according to Table 5. The progress of the reaction was monitored by TLC (eluent: petroleum ether/EtOAc, 1:1). After completion of the reaction, the work-up was performed as described for the synthesis of aminonaphthoquinones and the pure product was obtained in 85-98% yields. (Table 5, entries 1-5). For synthesis of symmetric *bis*- aminonaphthoquinones from diamine, the related diamine (0.5 mmol) reacted with lawsone (1 mmol) and aldehyde (1 mmol) in the presence of the catalyst (3 mol%, 50 mg) under the same conditions and the pure product was obtained in 90-98% yields (Table 5, entries 6-8).

6. Synthesis of unsymmetric *bis*-aminonaphthoquinones catalyzed by Fe_3O_4@TDSN-Bi(III)

A mixture of terephthaldialdehyde (0.5 mmol), lawsone (1 mmol), amine **3** (0.5 mmol), amine **9** (0.5 mmol) and Fe_3O_4@TDSN-Bi(III) (3 mol%, 50 mg) in EtOH (3 mL) was stirred at room temperature for the time indicated in Scheme 4. The progress of the reaction was monitored by TLC (eluent: petroleum ether/EtOAc, 1:1). After completion of the reaction, the catalyst was easily separated by a permanent magnet and washed with EtOH (5 mL). The organic residue was filtered, washed with cold ethanol for several times and dried under vacuum to give the pure product in 93-95% yields.

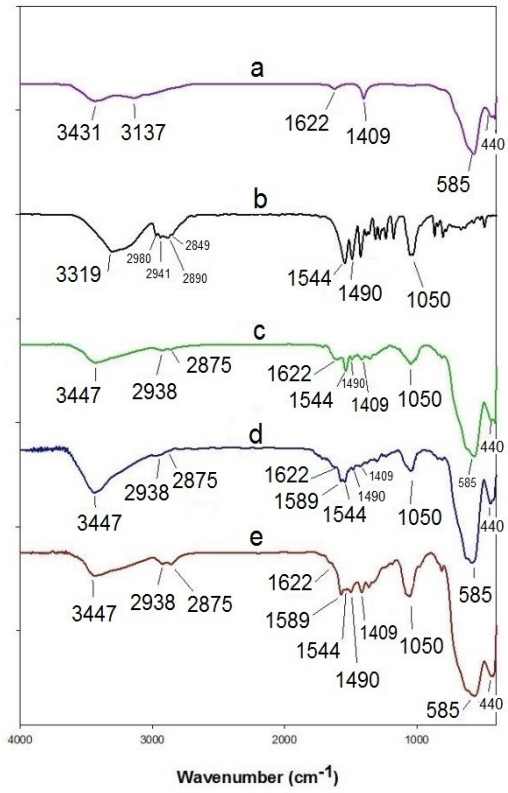


Figure 1. FT-IR spectra of: (a) Fe_3O_4, (b) $(DEA)_3T$, (c) G1, (d) CC2 and (e) G2 (Fe_3O_4@TDSN).

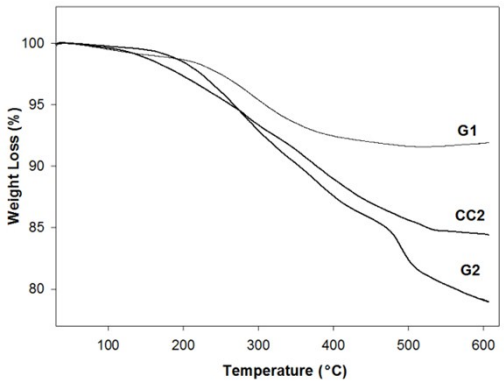


Figure 2. TGA spectra of G1, CC2 and G2.

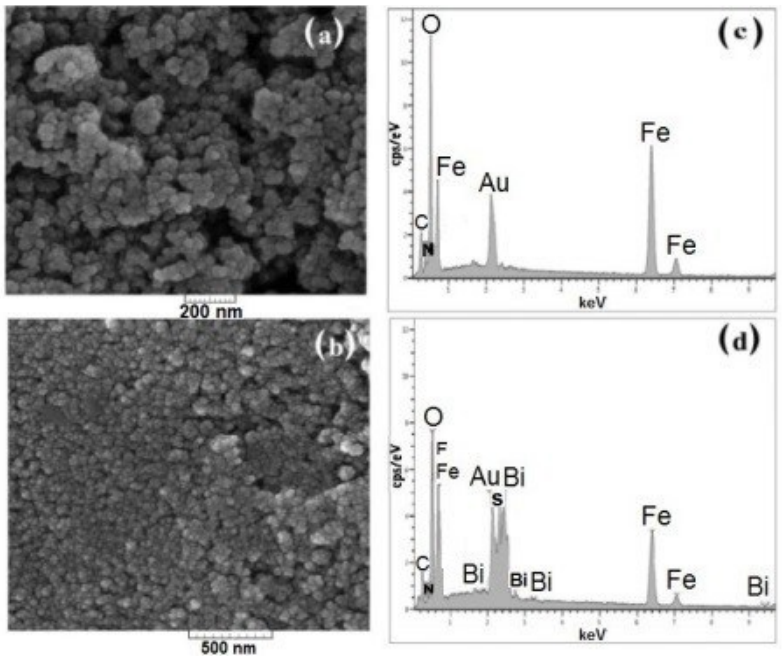


Figure 3. FE-SEM image of: (a) Fe_3O_4@TDSN and (b) Fe_3O_4@TDSN-Bi(III). SEM-EDX spectrum of: (c) Fe_3O_4@TDSN and (d) Fe_3O_4@TDSN-Bi(III).

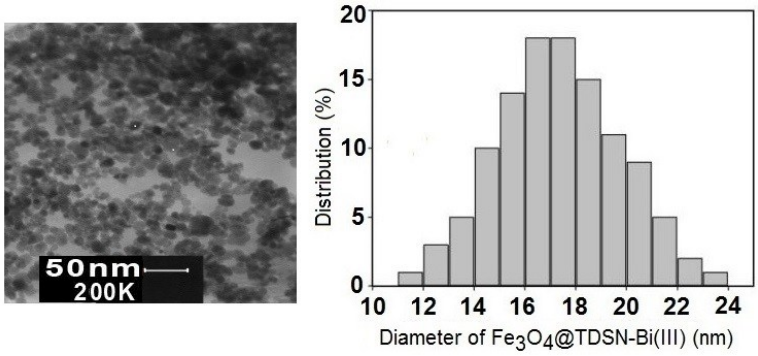


Figure 4. TEM image and particle size distribution for Fe_3O_4@TDSN-Bi(III) catalyst.

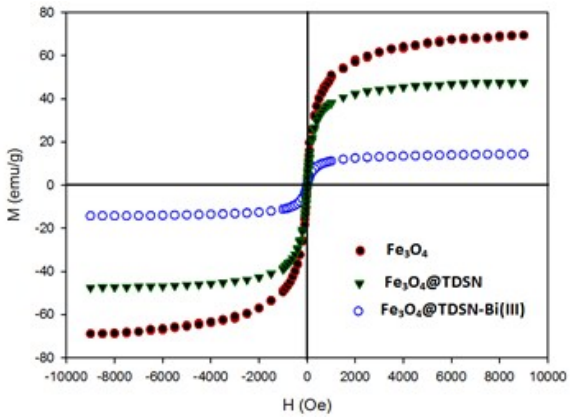


Figure 5. Magnetization curves for Fe_3O_4, Fe_3O_4@TDSN and Fe_3O_4@TDSN-Bi(III).

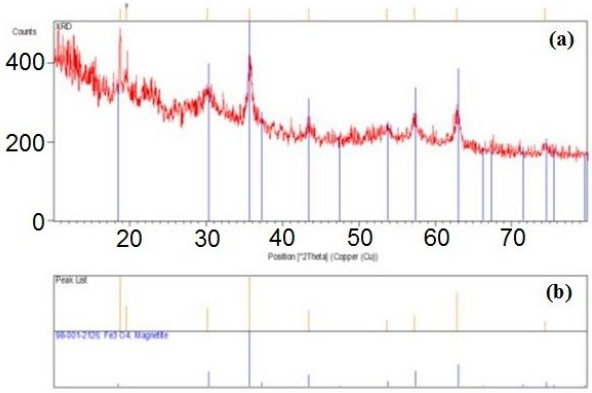


Figure 6. Powder X-ray diffraction patterns of: (a) Fe_3O_4@TDSN-Bi(III) and (b) Fe_3O_4@TDSN-Bi(III) and standard Fe_3O_4.

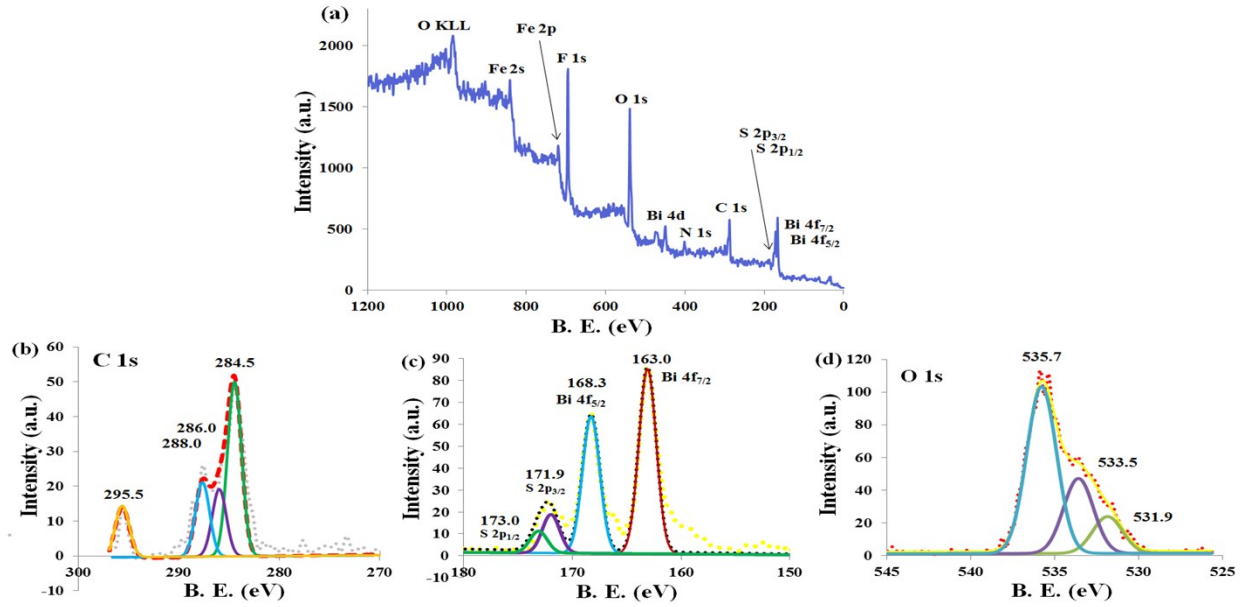
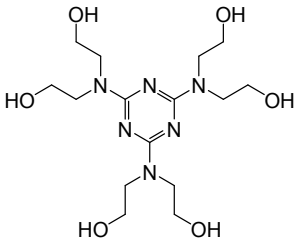


Figure 7. (a) XPS survey spectrum of Fe_3O_4@TDSN-Bi(III) and the high-resolution XPS spectra of: (b) C 1s, (c) Bi 4f and S 2p, and (d) O 1s.

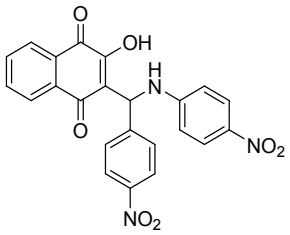
14. Spectroscopic data of the products:

2,2',2'',2''',2'''',2'''''-((1,3,5-Triazine-2,4,6-triyl)tris(azanetriyl))hexaethanol (Scheme 2, $(DEA)_3T$):



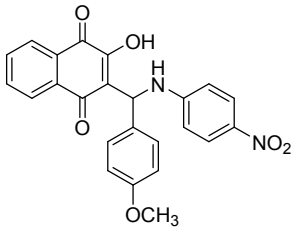
White solid, mp. 152-154 °C. IR (KBr): ν_{max} = 3319, 2980, 2941, 2890, 1544, 1490, 1424, 1272, 1182, 1050, 869, 804 cm^{-1}. ^1H NMR (400 MHz, DMSO-d_6): δ = 4.69 (s, 6H), 3.34 (s, 24H). ^{13}C NMR (100 MHz, DMSO-d6): δ = 164.3, 59.4, 50.5.

2-Hydroxy-3-((4-nitrophenyl)((4-nitrophenyl)amino)methyl)naphthalene-1,4-dione (Table 4, 4a):



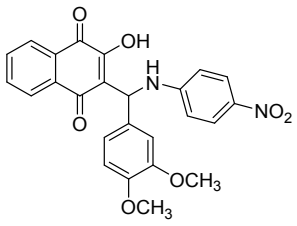
Yellow solid, mp. 135-1306 °C. (Lit.[2] 134-136 °C) IR (KBr): ν_{max} = 3366, 3076, 1677, 1642, 1599, 1508, 1329, 1262, 1182, 1116, 1051, 903, 898, 838, 796, 758 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 8.19 (d, J = 8.8 Hz, 2H), 7.93-8.04 (m, 4H), 7.86 (td, ^1J = 7.4 Hz, ^2J = 1.4 Hz, 1H), 7.82 (td, ^1J = 7.4 Hz, ^2J = 1.5 Hz, 1H), 7.69 (d, J = 8.6 Hz, 2H), 6.82 (d, J = 8.7 Hz, 2H), 6.24 (s, 1H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 182.1, 179.8, 155.7, 152.2, 146.9, 145.3, 135.6, 133.7, 132.4, 130.5, 128.9, 126.7, 124.9, 124.8, 124.8, 122.2, 119.2, 111.2, 49.4.

2-Hydroxy-3-((4-methoxyphenyl)((4-nitrophenyl)amino)methyl)naphthalene-1,4-dione (Table 4, 4b):



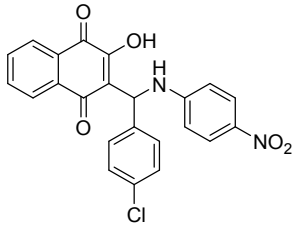
Yellow solid, mp. 220-222 °C (dec.). (Lit.[2] 222-224 °C (dec.)). IR (KBr): ν_{max} = 3447, 3188, 3015, 1673, 1625, 1591, 1505, 1343, 1269, 1243, 1108, 1045, 990, 898, 865, 845, 765 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 8.10 (dd, ^1J = 7.5 Hz, ^2J = 1.0 Hz, 2H), 7.99 (dd, ^1J = 7.5 Hz, ^2J = 1.2 Hz, 2H), 7.90 (td, ^1J = 7.4 Hz, ^2J = 1.4 Hz, 2H), 7.84 (td, ^1J = 7.4 Hz, ^2J = 1.4 Hz, 2H), 7.10-7.15 (m, 2H), 6.58-6.61 (m, 2H), 6.18 (s, 1H), 3.87 (s, 3H).

2-((3,4-Dimethoxyphenyl)((4-nitrophenyl)amino)methyl)-3-hydroxynaphthalene-1,4-dione (Table 4, 4c):



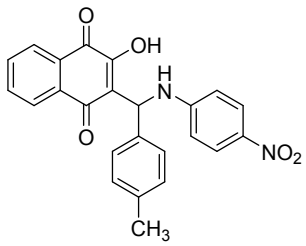
Red solid, mp. 165-167 °C (dec.). IR (KBr): ν_{max} = 3393, 3359, 3071, 1675, 1641, 1597, 1373, 1271, 1237, 1154, 1112, 1023, 914, 865, 838, 716 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 9.84 (s, 1H), 7.94 (d, J = 9.1 Hz, 2H), 7.76-7.88 (m, 4H), 7.08 (d, J = 1.8 Hz, 1H), 6.96 (dd, ^1J = 8.4 Hz, ^2J = 1.8 Hz, 1H), 6.88 (d, J = 8.4 Hz, 1H), 6.60 (d, J = 9.1 Hz, 2H), 6.05 (s,1H), 3.87 (s, 3H), 3.83 (s, 3H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 191.4, 183.6, 181.1, 155.6, 155.5, 149.1, 134.7, 134.5, 133.2, 133.1, 132.1, 129.8, 128.0, 126.4, 126.1, 118.9, 112.7, 112.2, 111.2, 109.3, 55.8, 55.5, 50.9. Anal. Calcd for $C_{25}H_{20}N_2O_7$: C, 65.21; H, 4.38; N, 6.08. Found: C, 65.07; H, 4.34; N, 6.17.

2-((4-Chlorophenyl)((4-nitrophenyl)amino)methyl)-3-hydroxynaphthalene-1,4-dione (Table 4, 4d):



Orange solid, mp. 132-133 °C. IR (KBr): ν_{max} = 3477, 3362, 3071, 1674, 1640, 1596, 1329, 1261, 1186, 1112, 1044, 906, 898, 784, 739 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 10.00 (s, 1H), 7.92-8.02 (m, 4H), 7.80-7.87 (m, 2H), 7.46 (d, J = 8.5 Hz, 2H), 7.38 (d, J = 8.6 Hz, 2H), 6.78 (d, J = 8.6 Hz, 2H), 6.12 (s, 1H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 183.4, 181.1, 155.5, 153.4, 139.9, 136.4, 134.8, 134.6, 132.1, 131.1, 130.0, 128.1, 127.5, 126.4, 126.1, 126.0, 112.2, 111.5, 50.3. Anal. Calcd for $C_{23}H_{15}ClN_2O_5$: C, 63.53; H, 3.48; N, 6.44. Found: C, 63.44; H, 3.46; N, 6.47.

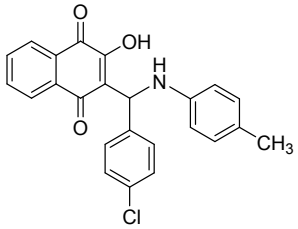
2-Hydroxy-3-(((4-nitrophenyl)amino)(p-tolyl)methyl)naphthalene-1,4-dione (Table 4, 4e):



Orange solid, mp. 145- 147 °C (dec.). IR (KBr): ν_{max}= 3641, 3354, 3071, 1660, 1643, 1598, 1324, 1260, 1183, 1112, 1047, 998, 898, 834, 773, 724 cm^{-1}. ^{1}H NMR (400 MHZ, DMSO-d_6): δ = 9.94 (s, 1H), 7.91-7.99 (m, 4H), 7.76-7.81 (m, 2H), 7.13 (d, J = 7.9 Hz, 2H), 7.00 (d, J = 7.9

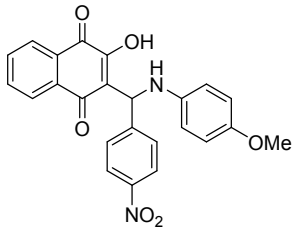
Hz, 2H), 6.79 (d, J = 8.2 Hz, 2H), 5.96 (s, 1H), 2.38 (s, 3H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 183.6, 181.1, 155.5, 153.5, 145.2, 136.2, 136.1, 135.6, 134.8, 131.6, 129.9, 128.1, 126.6, 126.3, 126.1, 126.0, 112.3, 111.3, 50.7, 21.3. Anal. Calcd for $C_{24}H_{18}N_2O_5$: C, 69.56; H, 4.38; N, 6.76. Found: C, 69.48; H, 4.40; N, 6.82.

2-((4-Chlorophenyl)(p-tolylamino)methyl)-3-hydroxynaphthalene-1,4-dione (Table 4, 4f):



Orange solid, mp. 151-153 ^{o}C (dec.). IR (KBr): ν_{max} = 3436, 3319, 3071, 1670, 1590, 1543, 1255, 1165, 1097, 1017, 985, 853, 819, 727 cm^{-1}. ^{1}H NMR (400 MHZ, DMSO-d_6): δ = 10.00 (s, 1H), 7.90-7.94 (m, 2H), 7.78 (td, ^1J = 7.5 Hz, ^2J = 1.3 Hz, 1H), 7.68-7.72 (m, 1H), 7.20-7.22 (m, 4H), 7.03 (d, J = 8.1 Hz, 2H), 6.81 (d, J = 8.1 Hz, 2H), 6.45 (s,1H), 2.32 (s, 3H). ^{13}C NMR (100 MHZ, DMSO-d_6): δ = 183.6, 182.7, 158.4, 140.5, 140.3, 134.9, 134.7, 132.3, 131.2, 130.5, 129.8, 129.7, 128.9, 127.5, 125.8, 125.3, 119.4, 113.4, 51.3, 20.3. Anal. Calcd for $C_{24}H_{18}ClNO_3$: C, 71.38; H, 4.49; N, 3.47. Found: C, 71.53; H, 4.47; N, 3.55.

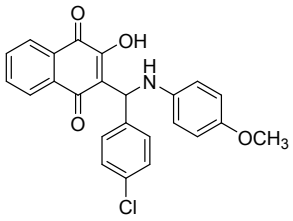
2-Hydroxy-3-(((4-methoxyphenyl)amino)(4-nitrophenyl)methyl)naphthalene-1,4-dione (Table 4, 4g):



Yellow solid, mp. 139-140 ^{o}C. IR (KBr): ν_{max} = 3370, 3071, 1670, 1620, 1594, 1510, 1260, 1236, 1174, 1105, 1026, 981, 860, 822, 708 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 8.05 (d, J = 8.8 Hz, 2H), 7.97 (dd, ^1J = 7.6 Hz, ^2J = 0.9 Hz, 1H), 7.92 (dd, ^1J = 7.4 Hz, ^2J = 0.9 Hz, 1H), 7.79 (td, ^1J = 7.5 Hz, ^2J = 1.2 Hz, 1H), 7.71 (td, ^1J = 7.5 Hz, ^2J = 1.2 Hz, 1H), 7.41 (d, J = 8.9 Hz, 2H), 7.14 (d, J = 8.9 Hz, 2H), 6.97 (d, J = 8.9 Hz, 2H), 6.63 (s, 1H), 3.80 (s, 3H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 183.0, 181.5, 156.0, 151.9, 148.5, 145.2, 140.6, 134.7, 133.9, 130.7,

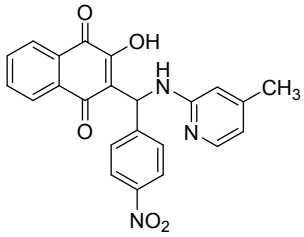
130.2, 128.3, 125.8, 125.7, 125.3, 114.8, 114.8, 114.6, 55.4, 52.5. Anal. Calcd for $C_{24}H_{18}N_2O_6$: C, 66.97; H, 4.22; N, 6.51. Found: C, 66.84; H, 4.16; N, 6.59.

2-((4-Chlorophenyl)((4-methoxyphenyl)amino)methyl)-3-hydroxynaphthalene-1,4-dione (Table 4, 4h):



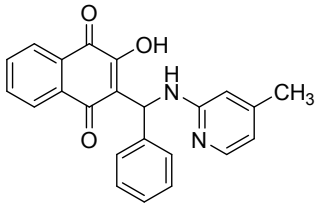
Yellow solid, mp. 127-128 °C (dec.). IR (KBr): ν_{max} = 3378, 3064, 1671, 1613, 1588, 1512, 1258, 1177, 1086, 1031, 975, 876, 830, 724 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 7.96 (dd, ^1J = 7.7 Hz, ^2J = 1.0 Hz, 1H), 7.90-7.92 (m, 1H), 7.77 (td, ^1J = 7.5 Hz, ^2J = 1.3 Hz, 1H), 7.70 (td, ^1J = 7.5 Hz, ^2J = 1.3 Hz, 1H), 7.15-7.22 (m, 4H), 7.11 (d, J = 8.9 Hz, 2H), 6.95 (d, J = 8.9 Hz, 2H), 6.51 (s, 1H), 3.78 (s, 3H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 183.0, 182.5, 157.3, 156.9, 143.7, 140.4, 135.5, 135.1, 132.1, 130.7, 129.9, 128.9, 127.6, 125.8, 125.2, 115.5, 114.8, 114.4, 55.4, 52.9. Anal. Calcd for $C_{24}H_{18}ClNO_4$: C, 68.66; H, 4.32; N, 3.34. Found: C, 68.52; H, 4.36; N, 3.23.

2-Hydroxy-3-(((4-methylpyridin-2-yl)amino)(4-nitrophenyl)methyl)naphthalene-1,4-dione (Table 4, 4i):



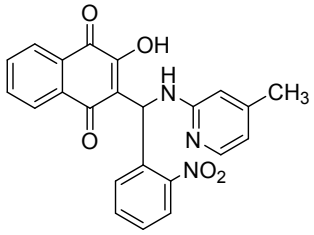
Orange solid, mp. 148-150 °C (dec.). IR (KBr): ν_{max} = 3444, 3262, 3075, 1667, 1631, 1590, 1537, 1281, 1110, 1056, 994, 898, 808, 792, 732 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 8.12 (d, J = 8.8 Hz, 2H), 8.08 (d, J = 8.8 Hz, 1H), 7.57- 7.73 (m, 6H), 6.91 (s, 1H), 6.63-6.67 (m, 2H), 5.99 (s, 1H), 2.29 (s, 3H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 192.3, 184.9, 179.6, 155.1, 153.2, 150.6, 145.8, 134.0, 133.9, 131.2, 130.9, 128.1, 127.1, 126.6, 125.7, 122.9, 114.2, 111.3, 50.4, 21.4. Anal. Calcd for $C_{23}H_{17}N_3O_5$: C, 66.50; H, 4.12; N, 10.12. Found: C, 66.91; H, 4.08; N, 10.01.

2-Hydroxy-3-(((4-methylpyridine-2-y1)amino)(phenl)methyl)naphthalene-1,4-dione (Table 4, 4j):



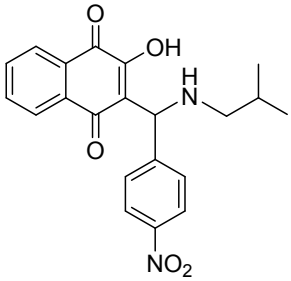
Orange solid, mp. 211-213 ^{o}C (dec.). IR (KBr): ν_{max} = 3442, 3265, 3065, 1675, 1590, 1510, 1277, 1234, 1188, 1119, 1059, 998, 898, 853, 792 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 10.02 (s, 1H), 7.97 (d, J = 7.2 Hz, 1H), 7.87-7.93 (m, 2H), 7.72-7.78 (m, 3H), 7.67 (td, ^{1}J = 7.5 Hz, ^{2}J = 1.0 Hz, 1H), 7.11-7.18 (m, 3H), 6.69 (s, 1H), 6.50- 6.53 (m, 2H), 2.22 (s, 3H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.2, 183.6, 182.4, 156.4, 151.9, 141.3, 140.9, 134.6, 133.8, 131.9, 130.9, 129.1, 127.7, 126.7, 125.7, 125.1, 122.3, 113.7, 109.9, 50.7, 20.9. Anal. Calcd for $C_{23}H_{18}N_2O_3$: C, 74.58; H, 4.90; N, 7.56. Found: C, 74.68; H, 4.85; N, 7.67.

2-Hydroxy-3-(((4-methylpyridine-2-y1)amino)(2-nitrophenyl)methyl)naphthalene-1,4-dione (Table 4, 4k):



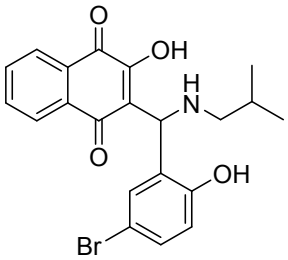
Orange solid, mp. 194-196 ^{o}C (dec.). IR (KBr): ν_{max} = 3431, 3289, 3066, 1679, 1647, 1590, 1529, 1276, 1211, 1119, 1043, 960, 838, 762, 754 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 10.24 (s, 1H), 7.92-7.96 (m, 3H), 7.87 (d, J = 7.4 Hz, 1H), 7.75 -7.77 (m, 2H), 7.68 (t, J = 7.4 Hz, 1H), 7.47-7.51 (m, 1H), 7.41 (d, J = 7.8 Hz, 1H), 6.84 (s, 1H), 6.50-6.53 (m, 2H), 2.22 (s, 3H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 189.9, 182.9, 181.8, 155.8, 152.7, 149.4, 139.9, 134.2, 134.1, 132.0, 131.6, 130.9, 127.5, 126.5, 125.7, 125.1, 124.3, 120.6, 113.8, 110.2, 47.4, 21.0. Anal. Calcd for $C_{23}H_{17}N_3O_5$: C, 66.50; H, 4.12; N, 10.12. Found: C, 66.61; H, 4.08; N, 10.24.

2-Hydroxy-3-((isobutylamino)(4-nitrophenyl)methyl)naphthalene-1,4-dione (Table 4, 4l):



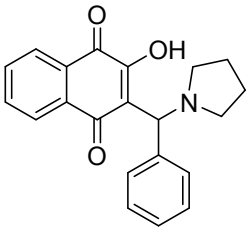
Yellow solid, mp. 171-173 °C (dec.). IR (KBr): ν_{max} = 3439, 3334, 3068, 2962, 2874, 1674, 1591, 1536, 1348, 1270, 1242, 1116, 1063, 1017, 991, 861, 739 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 8.28 (d, J = 8.8 Hz, 2H), 7.97 (dd, ^1J = 7.6 Hz, ^2J = 1.0 Hz, 1H), 7.92 (d, J = 8.8 Hz, 2H), 7.88 (dd, ^1J = 7.6 Hz, ^2J = 1.0 Hz, 1H), 7.77 (td, ^1J = 7.5 Hz, ^2J = 1.2 Hz, 1H), 7.64 (td, ^1J = 7.5 Hz, ^2J = 1.2 Hz, 1H), 5.68 (s, 1H), 2.70 (d, J = 7.0 Hz, 2H), 1.95-2.07 (m, 1H), 0.98 (d, J = 7.0 Hz, 6H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 183.9, 178.4, 170.6, 146.8, 145.8, 134.4, 133.8, 131.5, 131.0, 128.7, 128.0, 125.5, 125.1, 110.1, 58.5, 52.9, 25.5, 19.9. Anal. Calcd for $C_{21}H_{20}N_2O_5$: C, 66.31; H, 5.30; N, 7.36. Found: C, 66.40; H, 5.25; N, 7.43.

2-((5-Bromo-2-hydroxyphenyl)(isobutylamino)methyl)-3-hydroxynaphthalene-1,4-dione (Table 4, 4m):



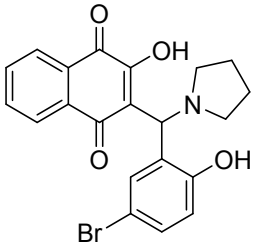
Orange solid, mp. 179-181 °C (dec.). IR (KBr): ν_{max} = 3373, 3173, 2962, 2938, 2874, 1677, 1590, 1523, 1275, 1234, 1173, 1112, 1067, 1013, 983, 898, 853, 701 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 7.99-8.01 (m, 1H), 7.93 (d, J = 7.4 Hz, 1H), 7.79-7.82 (m, 1H), 7.70-7.73 (m, 1H), 7.48 (s, 1H), 7.38 (d, J = 8.6 Hz, 1H), 6.90 (d, J = 8.6 Hz, 1H), 5.72 (s, 1H), 2.69 (d, J = 7.0 Hz, 2H), 1.92-2.02 (m, 1H), 0.98 (d, J = 7.0 Hz, 6H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 183.7, 182.1, 154.7, 154.3, 134.9, 134.7, 134.3, 131.2, 130.8, 125.6, 125.5, 122.2, 120.0, 119.1, 117.9, 116.6, 54.1, 53.3, 28.9, 20.1. Anal. Calcd for $C_{21}H_{20}BrNO_4$: C, 58.62; H, 4.68; N, 3.26. Found: C, 58.75; H, 4.63; N, 3.35.

2-Hydroxy-3-(phenyl(pyrrolidin-1-y1)methyl)naphthalene-1,4-dione (Table 4, 4n):



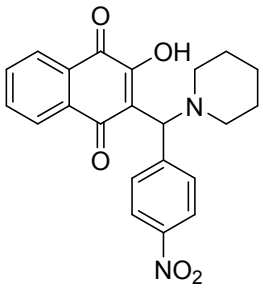
Red solid, mp. 178-179 °C (dec.). (Lit.[2] 177-179 °C (dec.)). IR (KBr): ν_{max} = 3330, 3064, 2976, 2872, 1681, 1587, 1535, 1272, 1220, 1181, 1143, 959, 840, 792, 767 cm^{-1}. ^1H NMR (400 MHz, DMSO-d_6): δ = 10.21 (br s, 1H), 7.92 (d, J = 7.1 Hz, 1H), 7.80 (d, J = 7.1 HZ, 1H), 7.67-7.71 (m, 3H), 7.56 (td, ^1J = 7.4 Hz, ^2J = 1.1 Hz, 1H), 7.32-7.35 (m, 2H), 7.26-7.29 (m, 1H), 5.50 (s,1H), 3.16 (br s, 4H), 1.94-1.95 (m, 4H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 184.4, 177.7, 170.2, 138.6, 134.7, 133.7, 131.4, 130.8, 128.3, 127.9, 127.3, 125.3, 125.1, 113.0, 67.1, 53.4, 23.2.

2-((5-Bromo-2-hydroxyphenyl)(pyrrolidin-1-y1)methyl)-3-hydroxynaphthalene-1,4-dione (Table 4, 4o):



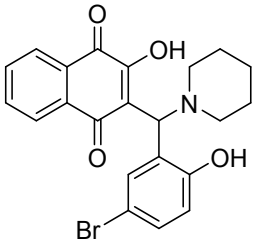
Red solid, mp. 200-201 °C. IR (KBr): ν_{max} = 3343, 3071, 2972, 2872, 1685, 1591, 1512, 1273, 1242, 1158, 1089, 1013, 945, 868, 815, 731 cm^{-1}. ^1H NMR (400 MHZ, DMSO-d_6): δ = 10.19 (br s, 2H), 7.92 (d, J = 7.5 Hz, 1H), 7.88 (s, 1H), 7.83 (d, J = 7.5 Hz, 1H), 7.71 (td, ^1J = 7.5 Hz, ^2J = 1.0 Hz, 1H), 7.59 (td, ^1J = 7.5 Hz, ^2J = 1.0 Hz, 1H), 7.26 (dd, ^1J = 8.6 Hz, ^2J = 2.4 Hz, 1H), 6.77 (d, J = 8.6 Hz,1H) 5.77 (s, 1H), 3.14 (br s, 4H), 1.92 (br s, 4H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 184.3, 178.7, 170.8, 154.4, 134.5, 133.8, 131.6, 131.4, 131.4, 130.9, 127.4, 125.3, 125.2, 118.3, 113.4, 110.1, 60.4, 53.1, 23.1. Anal. Calcd for $C_{21}H_{18}BrNO_4$: C, 58.89; H, 4.24; N, 3.27. Found: C, 59.03; H, 4.21; N, 3.16.

2-Hydroxy-3-((4-nitrophenyl)(piperidin-1-y1)methyl)naphthalene-1,4-dione (Table 4, 4p):



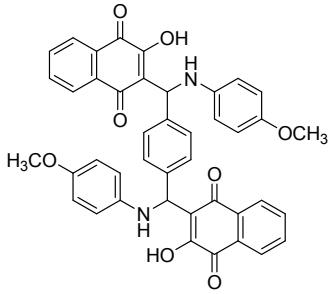
Orange solid, mp. 180-181 °C (dec.). IR (KBr): ν_{max} = 3350, 3058, 2996, 2873, 1684, 1586, 1539, 1341, 1267, 1143, 1112, 1013, 975, 936, 845, 800, 754 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 8.22 (d, J = 8.6 Hz, 2H), 7.89-7.92 (m, 3H), 7.81 (d, J = 7.0 Hz, 1H), 7.70 (td, ^1J = 7.5 Hz, ^2J = 1.0 Hz, 1H), 7.58 (td, ^1J = 7.5 Hz, ^2J = 1.0 Hz, 1H), 5.72 (s, 1H), 2.97-3.02 (m, 4H), 1.81 (br s, 2H), 1.55–1.67 (m, 4H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 183.9, 178.1, 170.4, 146.9, 134.5, 133.7, 131.5, 130.4, 129.4, 126.5, 125.4, 125.1, 110.8, 66.1, 51.2, 22.6, 22.2. Anal. Calcd for $C_{22}H_{20}N_2O_5$: C, 67.34; H, 5.14; N, 7.14. Found: C, 67.53; H, 5.07; N, 7.25.

2-((5-Bromo-2-hydroxyphenyl)(piperidin-1-yl)methyl)-3-hydroxynaphthalene-1,4-dione (Table 4, 4q):



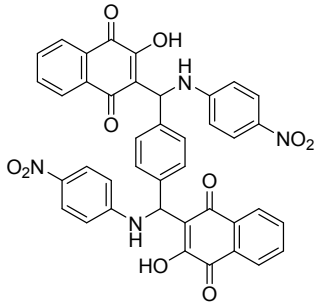
Orange solid, mp. 179-180 °C (dec.). IR (KBr): ν_{max} = 3307, 3066, 2946, 2858, 1690, 1588, 1515, 1273, 1211, 1150, 922, 891, 815, 731 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 7.90 (dd, ^1J = 7.6 Hz, ^2J = 0.7 Hz, 1H), 7.82 (dd, ^1J = 7.6 Hz, ^2J = 0.8 Hz, 1H), 7.70 (td, ^1J = 7.4 Hz, ^2J = 1.3 Hz, 1H), 7.56-7.64 (m, 2H), 7.24 (d, J = 8.6 Hz, 1H), 6.74 (d, J = 8.6Hz, 1H), 5.74 (s, 1H), 2.85 (br s, 4H), 1.69 (br s, 2H), 1.50-1.53 (m, 4H). ^{13}C NMR (100 MHZ, DMSO-d_6): δ = 183.6, 178.8, 170.7, 155.2, 134.9, 134.5, 133.7, 131.4, 130.9, 125.3, 125.2, 122.2, 119.9, 117.9, 116.5, 112.0, 61.0, 50.9, 22.1, 21.9. Anal. Calcd for $C_{22}H_{20}BrNO_4$: C, 59.74; H, 4.56; N, 3.17. Found: C, 59.91; H, 4.51; N, 3.27.

3,3'-(1,4-Phenylenebis(((4-methoxyphenyl)amino)methylene))bis(2-hydroxynaphthalene-1,4-dione) (Table 5, 6a):



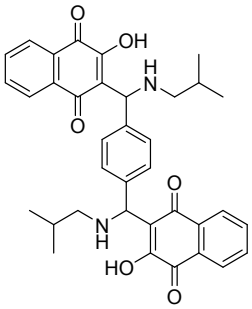
Dark yellow solid, mp. 160-162 °C (dec.). IR (KBr): ν_{max} = 3365, 3064, 1671, 1640, 1617, 1594, 1509, 1244, 1112, 1028, 983, 868, 731 cm^{-1}. ^1H NMR (400 MHz, DMSO-d_6): δ = 8.72 (s, 2H), 7.92-7.98 (m, 4H), 7.70-7.81 (m, 4H), 7.15 (d, J = 8.4 Hz, 4H), 6.97-7.02 (m, 8H), 6.55 (s, 2H), 3.76 (s, 6H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 182.9, 182.7, 157.8, 157.7, 144.1, 142.3, 134.0, 133.7, 132.3, 130.6, 127.6, 125.8, 125.3, 114.8, 114.4, 114.3, 55.4, 49.9. Anal. Calcd for $C_{42}H_{32}N_2O_8$: C, 72.82; H, 4.66; N, 4.04. Found: C, 72.73; H, 4.63; N, 4.14.

3,3'–(1,4-Phenylenebis(((4-nitrophenyl)amino)methylene))bis(2-hydroxynaphthalene-1,4-dione) (Table 5, 6b):



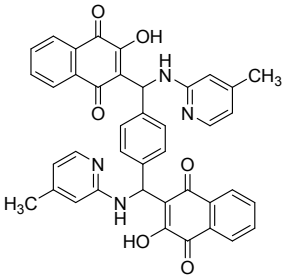
Orange solid, mp. 194-196 °C (dec.). IR (KBr): ν_{max} = 3387, 3330, 3073, 1698, 1645, 1596, 1524, 1504, 1326, 1261, 1181, 1112, 1051, 838, 731 cm^{-1}. ^1H NMR (400 MHz, DMSO-d_6): δ = 7.93-8.01 (m, 6H), 7.79-7.86 (m, 6H), 7.43 (s, 4H), 6.76 (d, J = 8.6 Hz, 4H), 6.11 (s, 2H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 183.4, 181.0, 156.1, 153.5, 139.8, 136.5, 135.6, 135.0, 131.6, 130.1, 127.2, 126.4, 126.1, 126.0, 112.3, 111.5, 50.8. Anal. Calcd for $C_{40}H_{26}N_4O_{10}$: C, 66.48; H, 3.63; N, 7.75. Found: C, 66.64; H, 3.61; N, 7.87.

3,3'-(1,4-Phenylenebis((isobutylamino)methylene))bis(2-hydroxynaphthalene-1,4-dione) (Table 5, 6c):



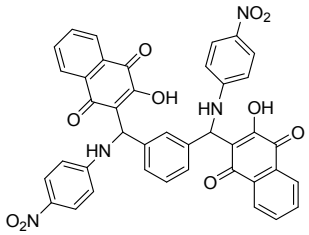
Red solid, mp. 170-171 °C (dec.). IR (KBr): ν_{max} = 3440, 3067, 2962, 2871, 1678, 1590, 1528, 1275, 1242, 1158, 1074, 1021, 983, 830, 739 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 7.98 (d, J = 8.0 Hz, 2H), 7.90 (d, J = 7.5 Hz, 2H), 7.72-7.75 (m, 2H), 7.67-7.70 (m, 2H), 7.01 (s, 4H), 5.41 (s, 2H), 2.62 (d, J = 7.0 Hz, 4H), 1.84- 1.94 (m, 2H), 0.89 (d, J = 6.7 Hz, 12H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 183.6, 182.4, 160.2, 137.8, 134.5, 133.9, 130.9, 130.8, 127.0, 125.1, 125.0, 108.9, 68.5, 45.7, 29.1, 20.4. Anal. Calcd for $C_{36}H_{36}N_2O_6$: C, 72.95; H, 6.12; N, 4.73. Found: C, 73.14; H, 6.06; N, 4.79.

3,3'(1,4-Phenylenebis(((4-methylpyridin-2-yl)amino)methylene))bis(2-hydroxynaphthalene-1,4- dione) (Table 5, 6d):



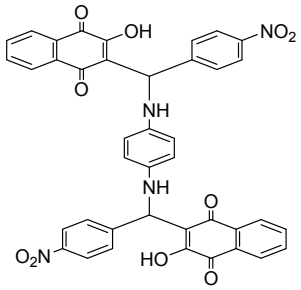
Red solid, mp. 188- 189 °C (dec.). IR (KBr): ν_{max} = 3403, 3208, 3066, 1674, 1640, 1592, 1564, 1275, 1226, 1181, 1127, 960, 807, 731 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 7.95 (d, J = 7.6 Hz, 2H), 7.86 (d, J = 7.4 Hz, 2H), 7.65-7.79 (m, 6H), 6.92 (s, 4H), 6.68 (s, 2H), 6.64 (d, J = 6.3 Hz, 2H), 6.59 (s, 2H), 2.28 (s, 6H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 183.4, 182.4, 163.8, 155.3, 153.7, 149.5, 138.6, 133.9, 133.6, 131.6, 130.7, 125.7, 125.2, 125.1, 121.8, 113.9, 110.8, 67.4, 21.1. Anal. Calcd for $C_{40}H_{30}N_4O_6$: C, 72.50; H, 4.56; N, 8.45. Found: C, 72.75; H, 4.50; N, 8.56.

3,3'-(1,3-Phenylenebis(((4-nitrophenyl)amino)methylene))bis(2-hydroxynaphthalene-1,4-dione) (Table 5, 6e):



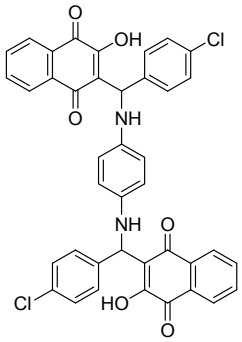
Orange solid, mp. 157-158 °C (dec.). IR (KBr): ν_{max} = 3378, 3190, 3074, 1670, 1646, 1596, 1504, 1313, 1276, 1188, 1112, 1051, 1013, 838, 731 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 7.97-7.99 (m, 4H), 7.75-7.93 (m, 8H), 7.30-3.35 (m, 4H), 6.65-6.74 (m, 4H), 6.08-6.09 (m, 2H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 183.4, 180.9, 155.6, 153.4, 142.3, 141.1, 136.3, 131.4, 130.1, 128.5, 127.6, 127.3, 126.3, 126.1, 126.0, 124.8, 124.7, 112.3, 111.3, 50.9, 50.6. Anal. Calcd for $C_{40}H_{26}N_4O_{10}$: C, 66.48; H, 3.63; N, 7.75. Found: C, 66.73; H, 3.56; N, 7.87.

3,3'-((1,4-Phenylenebis(azanediyl))bis((4-nitrophenyl)methylene))bis(2-hydroxynaphthalene-1,4- dione) (Table 5, 8a):



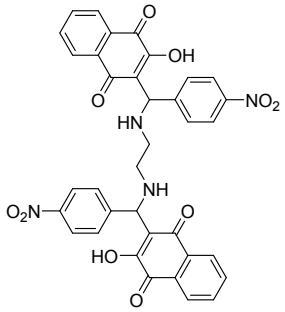
Red solid, mp. 180-181 °C (dec.). IR (KBr): ν_{max} = 3595, 3360, 3062, 1676, 1647, 1598, 1565, 1516, 1341, 1274, 1219, 1165, 1105, 1005, 853, 830, 730 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 8.31 (d, J = 8.8, 4H), 7.94-8.04 (m, 8H), 7.85 (td, ^1J = 7.4 Hz, ^2J = 1.2 Hz, 2H), 7.80 (td, ^1J = 7.4 Hz, ^2J = 1.2 Hz, 2H), 6.43 (br s, 4H), 6.16 (s, 2H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 183.4, 181.2, 156.8, 149.6, 146.2, 134.8, 134.3, 133.4, 131.7, 130.1, 129.6, 127.7, 125.8, 125.8, 115.7, 113.0, 50.9. Anal. Calcd for $C_{40}H_{26}N_4O_{10}$: C, 66.48; H, 3.63; N, 7.75. Found: C, 66.57; H, 3.60; N, 7.50.

3,3'-((1,4-Phenylenebis(azanediyl))bis((4-chlorophenyl)methylene))bis(2-hydroxynaphthalene-1,4- dione) (Table 5, 8b):



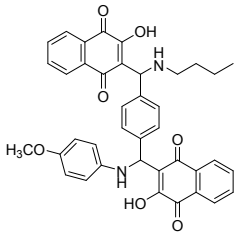
Dark yellow solid, mp. 145-147 °C (dec.). IR (KBr): ν_{max} =3442, 3363, 3067, 1674, 1634, 1595, 1513, 1279, 1211, 1150, 1089, 998, 960, 830, 724 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 7.95 (d, J = 7.0 Hz, 2H), 7.91 (dd, ^1J = 7.7 Hz, ^2J = 1.2 Hz, 2H), 7.78 (td, ^1J = 7.5 Hz, ^2J = 1.2 Hz, 2H), 7.70 (td, ^1J = 7.5 Hz, ^2J = 1.2 Hz, 2H), 7.16-7.22 (m, 8H), 6.46 (br s, 4H), 6.04 (s, 2H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 192.1, 182.6, 158.9, 140.4, 134.9, 134.0, 132.9, 132.3, 131.1, 130.2, 128.8, 127.5, 125.8, 125.2, 115.2, 113.6, 51.2. Anal. Calcd for $C_{40}H_{26}Cl_2N_2O_6$: C, 68.48; H, 3.74; N, 3.99. Found: C, 68.62; H, 3.71; N, 4.06.

3,3'-((Ethane-1,2-diylbis(azanediyl))bis((4-nitrophenyl)methylene))bis(2-hydroxynaphthalene-1,4- dione) (Table 5, 8c):



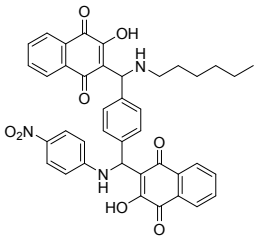
Yellow solid, mp. 190-192 °C (dec.). IR (KBr): ν_{max} = 3435, 3072, 2860, 1676, 1591, 1523, 1348, 1279, 1203, 1143, 1013, 960, 853, 739 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 8.65 (br s, 2H), 8.21 (d, J = 8.9 Hz, 4H), 7.86-7.92 (m, 4H), 7.81 (d, J = 8.7 Hz, 4H), 7.67-7.73 (m, 4H), 5.73 (s, 2H), 3.02 (s, 4H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 183.3, 180.7, 160.7, 146.8, 145.2, 135.0, 134.4, 131.3, 130.9, 128.1, 127.4, 125.8, 125.4, 111.0, 56.9, 41.4. Anal. Calcd for $C_{36}H_{26}N_4O_{10}$: C, 64.09; H, 3.88; N, 8.31. Found: C, 64.26; H, 3.84; N, 8.38.

2-((4-((Butylamino)(3-hydroxy-1,4-dioxo-1,4-dihydronaphthalen-2-yl)methyl)phenyl)((4-methoxyphenyl)amino)methyl)-3- hydroxynaphthalene-1,4-dione (Scheme 4, 10a):



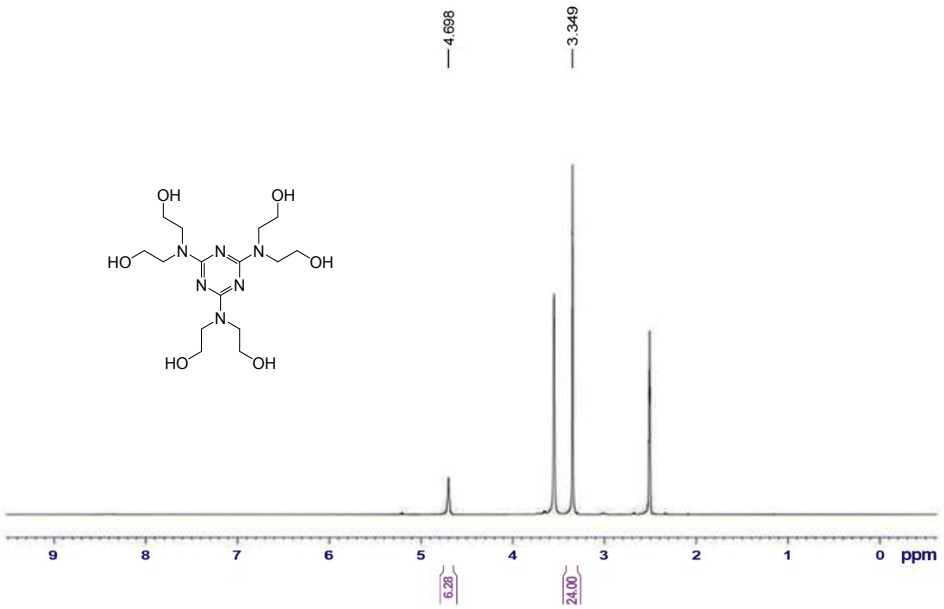
Orange solid, mp. 175-177 °C (dec). IR (KBr): ν_{max} = 3437, 3245, 3064, 2958, 2931, 2870, 2839, 1677, 1616, 1592, 1570, 1512, 1276, 1244, 1196, 1112, 1028, 983, 838, 746 cm^{-1}. ^1H NMR (400 MHz, DMSO-d_6): δ = 9.97 (br s, 2H), 7.88 (d, J = 7.5 Hz, 2H), 7.79 (d, J = 7.5Hz, 2H), 7.72-7.76 (m, 2H), 7.68 (t, J = 7.5 Hz, 2H), 7.54-7.58 (m, 4H), 7.08 (d, J = 8.2 Hz, 2H), 7.01 (d, J = 8.2 Hz, 2H), 5.47 (s, 2H), 3.79 (s, 3H), 2.84 (t, J = 7.4 Hz, 2H), 1.52-1.58 (m, 2H), 1.24-1.29 (m, 2H), 0.88 (t, J = 7.4 Hz, 3H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 183.6, 182.4, 158.4, 157.6, 143.8, 138.2, 137.5, 133.8, 133.7, 131.8, 130.8, 127.4, 126.8, 126.1, 125.7, 114.9, 114.4, 114.4, 60.3, 55.3, 55.2, 45.3, 27.6, 19.2, 13.7. Anal. Calcd for $C_{39}H_{34}N_2O_7$: C, 72.88; H, 5.33; N, 4.36. Found: C, 73.14; H, 5.29; N, 4.43.

2-((4-((Hexylamino)(3-hydroxy-1,4-dioxo-1,4-dihydronaphthalene-2-yl)methyl)phenyl)((4-nitrophenyl)amino)methyl)-3-hydroxynaphthalene-1,4-dione (Scheme 4, 10b):

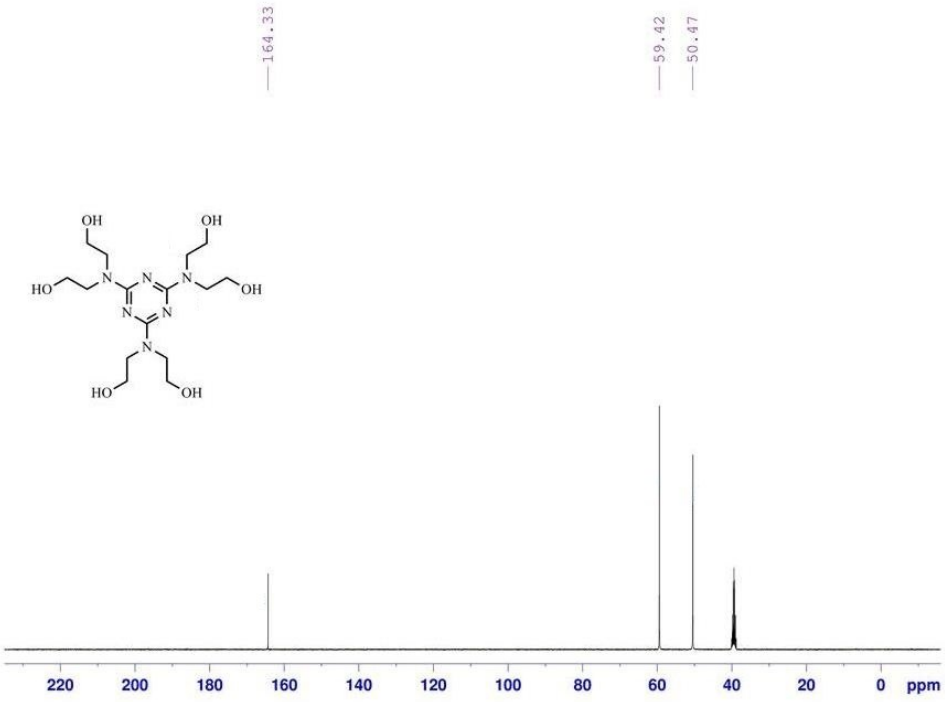


Orange solid, mp. 157-160 °C (dec.). IR (KBr): ν_{max} = 3444, 3069, 2955, 2928, 2859, 1675, 1637, 1595, 1567, 1278, 1211, 1158, 1067, 1013, 960, 922, 830, 739 cm^{-1}. ^1H NMR (400 MHz, DMSO-d_6): δ = 7.94 (d, J = 7.6 Hz, 2H), 7.87-7.89 (m, 2H), 7.84 (d, J = 7.5 Hz, 2H), 7.73 (td, ^1J = 7.4 Hz, ^2J = 1.1 Hz, 2H), 7.64 (t, J = 7.6 Hz, 2H), 6.87 (br s, 6H), 5.47 (s, 2H), 2.83 (t, J = 7.5 Hz, 2H), 1.54-1.59 (m, 2H), 1.21-1.28 (m, 6H), 0.86 (t, J = 7.0 Hz, 3H). ^{13}C NMR (100 MHz, DMSO-d_6): δ = 184.3, 183.5, 178.5, 170.5, 141.5, 135.0, 134.6, 133.7, 133.2, 131.8, 130.9, 127.3, 127.0, 126.8, 126.1, 125.7, 125.0, 110.9, 60.5, 58.6, 45.5, 31.0, 30.6, 26.9, 21.8, 13.8. Anal. Calcd for $C_{40}H_{35}N_3O_8$: C, 70.06; H, 5.14; N, 6.13. Found: C, 70.18; H, 5.09; N, 6.22.

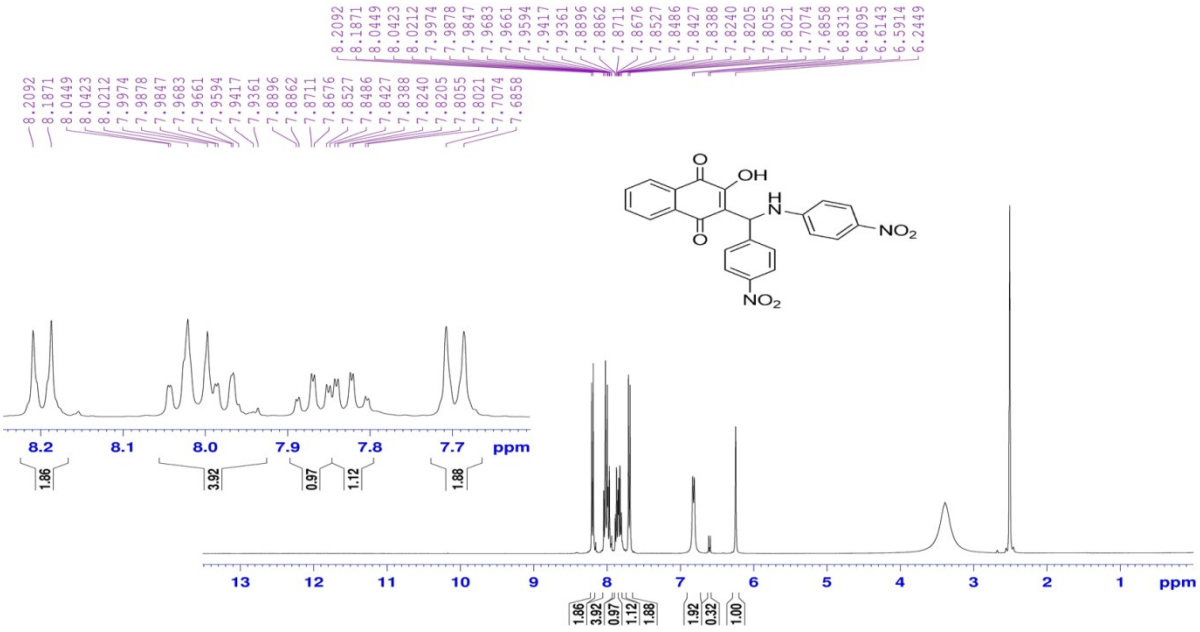
15. ^{1}H NMR and ^{13}C NMR Spectra of the Products:



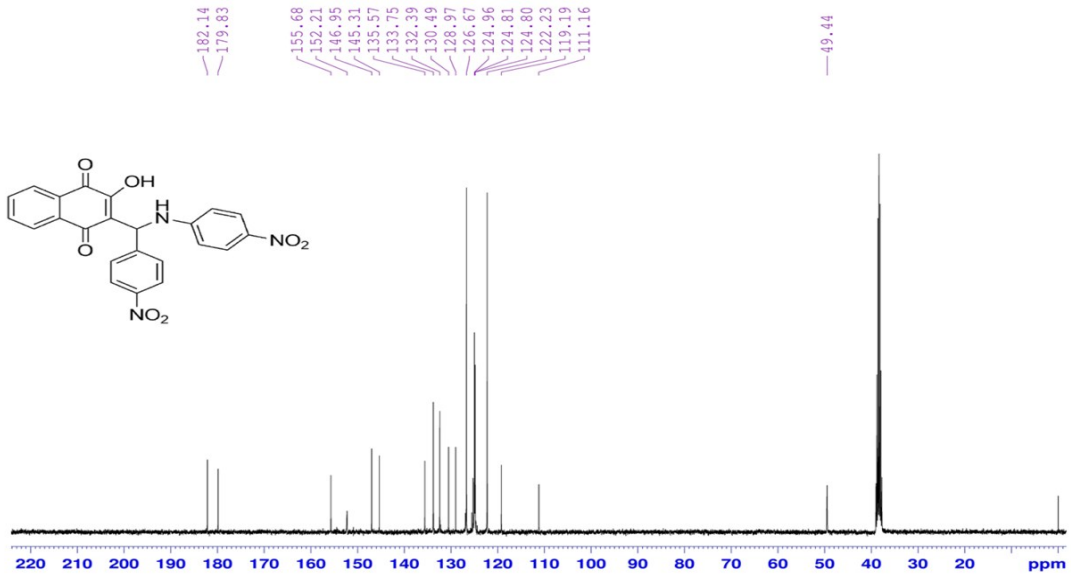
2,2',2'',2''',2'''',2'''''-((1,3,5-Triazine-2,4,6-triyl)tris(azanetriyl))hexaethanol ((DEA)$_3$T, Scheme 2); ^{1}H NMR (400 MHz, DMSO-d_6)



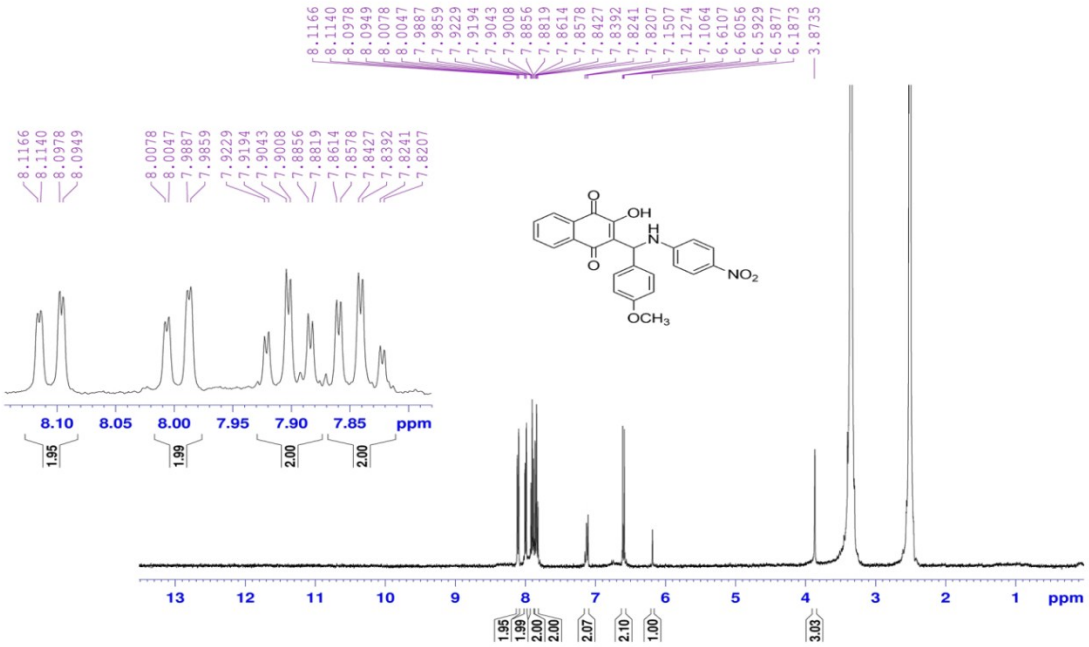
2,2',2'',2''',2'''',2'''''-((1,3,5-Triazine-2,4,6-triyl)tris(azanetriyl))hexaethanol ((DEA)$_3$T, Scheme 2); ^{13}C NMR (100 MHz, DMSO-d_6)



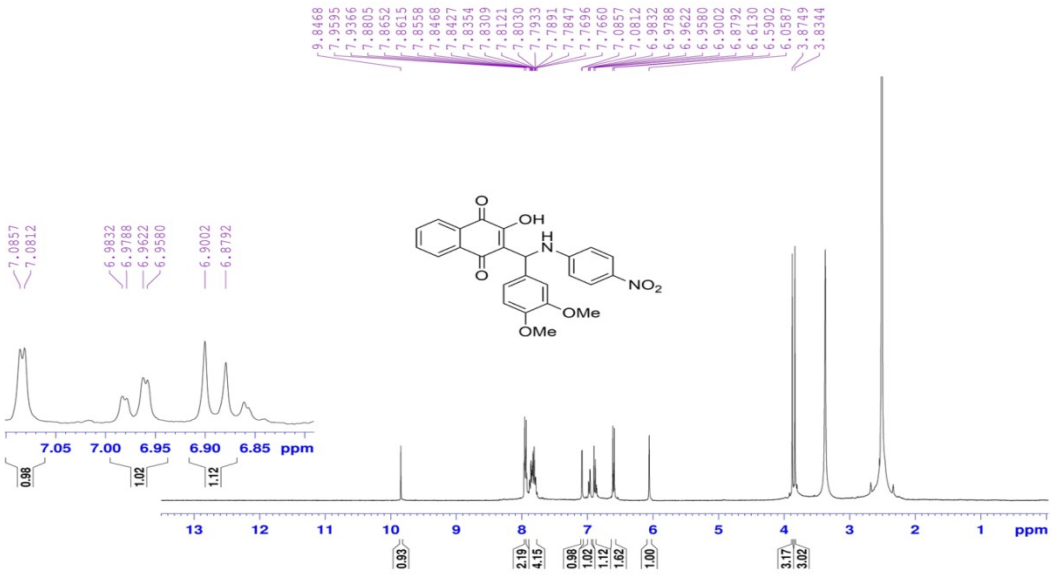
2- Hydroxy-3-((4- nitrophenyl)((4- nitrophenyl)amino)methyl)naphthalene-1,4-dione (Table 4, 4a); ^{1}H NMR (400 MHz, DMSO-d_6)



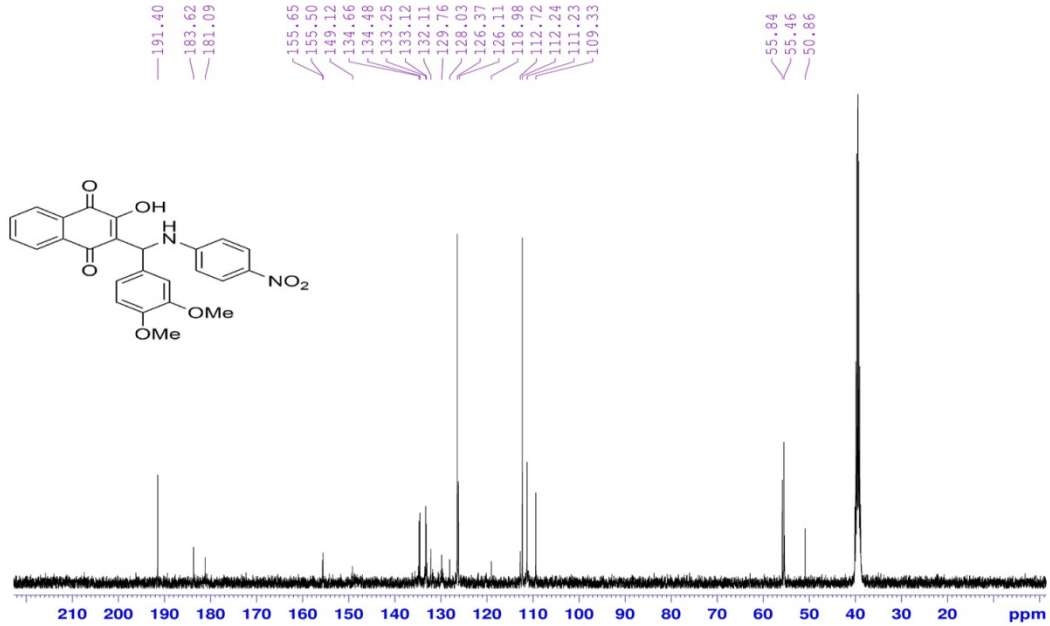
2- Hydroxy-3-((4- nitrophenyl)((4- nitrophenyl)amino)methyl)naphthalene-1,4-dione (Table 4, 4a); ^{13}C NMR (100 MHz, DMSO-d_6)



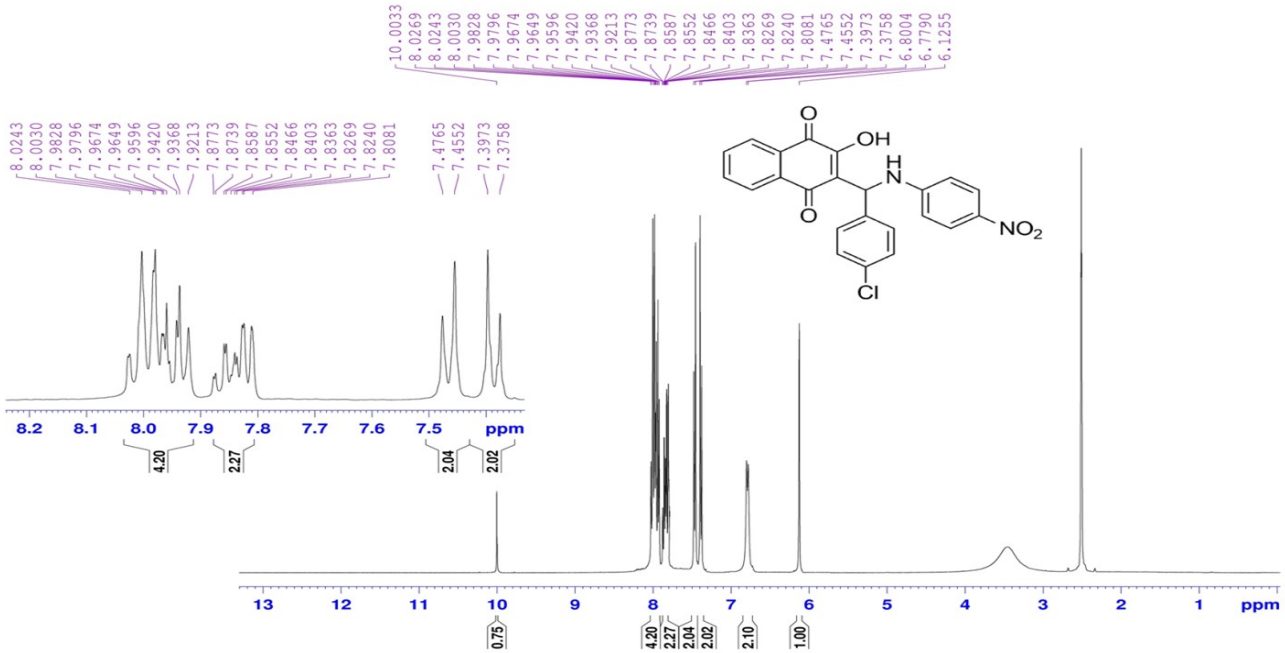
2-Hydroxy-3-((4-methoxyphenyl)((4-nitrophenyl)amino)methyl)naphthalene-1,4-dione (Table 4, 4b); ^{1}H NMR (400 MHz, DMSO-d_6)



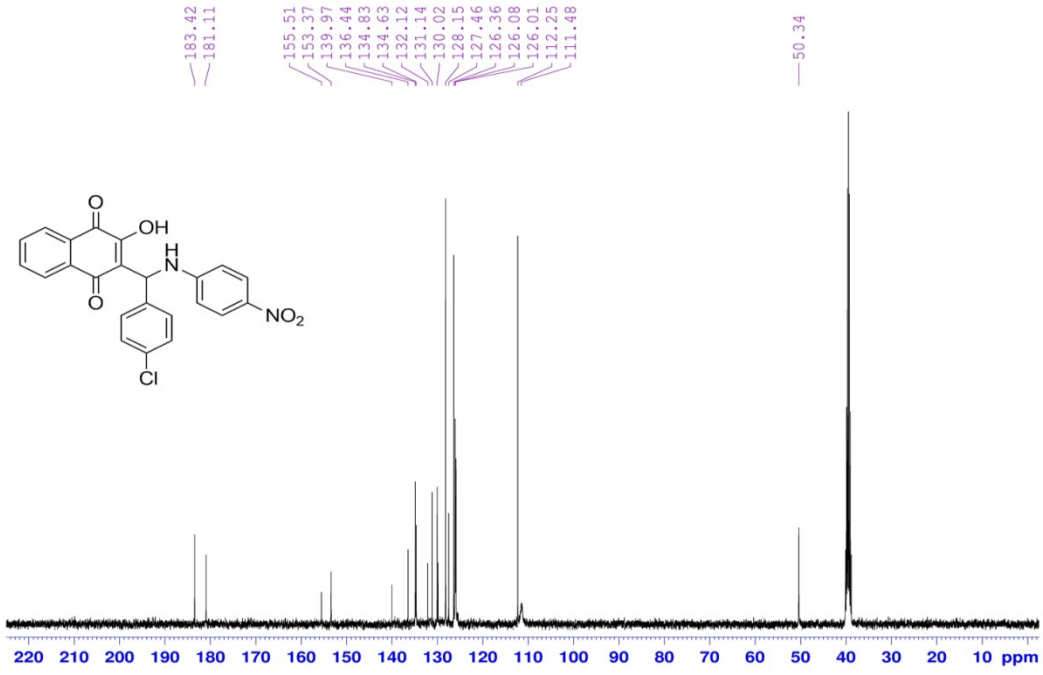
2-((3,4-Dimethoxyphenyl)((4-nitrophenyl)amino)methyl)-3-hydroxynaphthalene-1,4-dione (Table 4, 4c); ^{1}H NMR (400 MHz, DMSO-d_6)



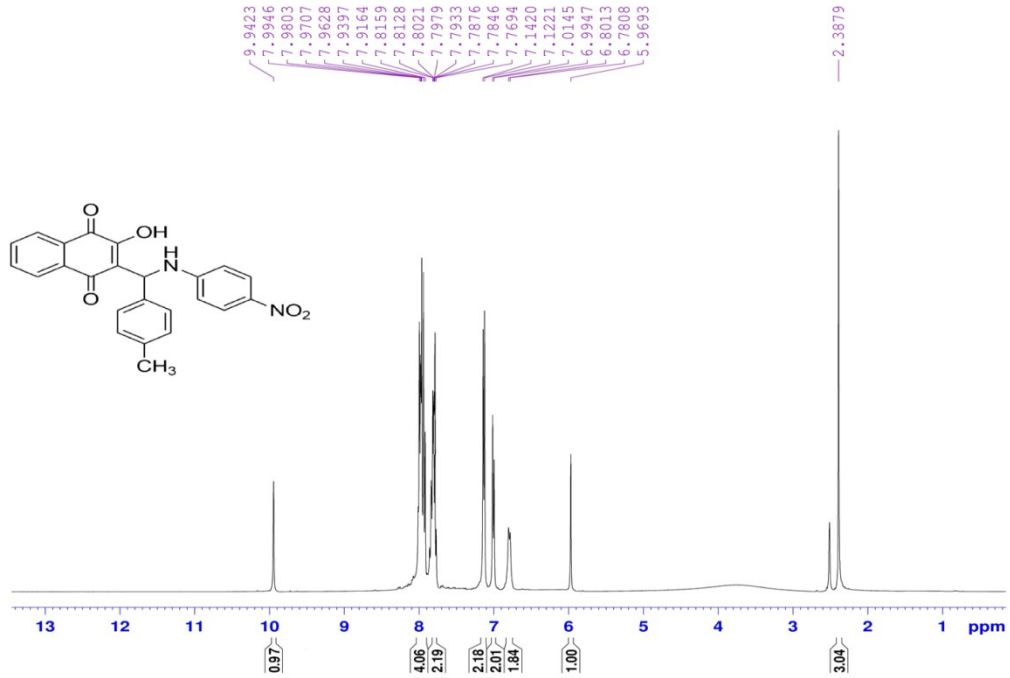
2-((3,4-Dimethoxyphenyl)((4-nitrophenyl)amino)methyl)-3-hydroxynaphthalene-1,4-dione (Table 4, 4c); ^{13}C NMR (100 MHz, DMSO-d_6)



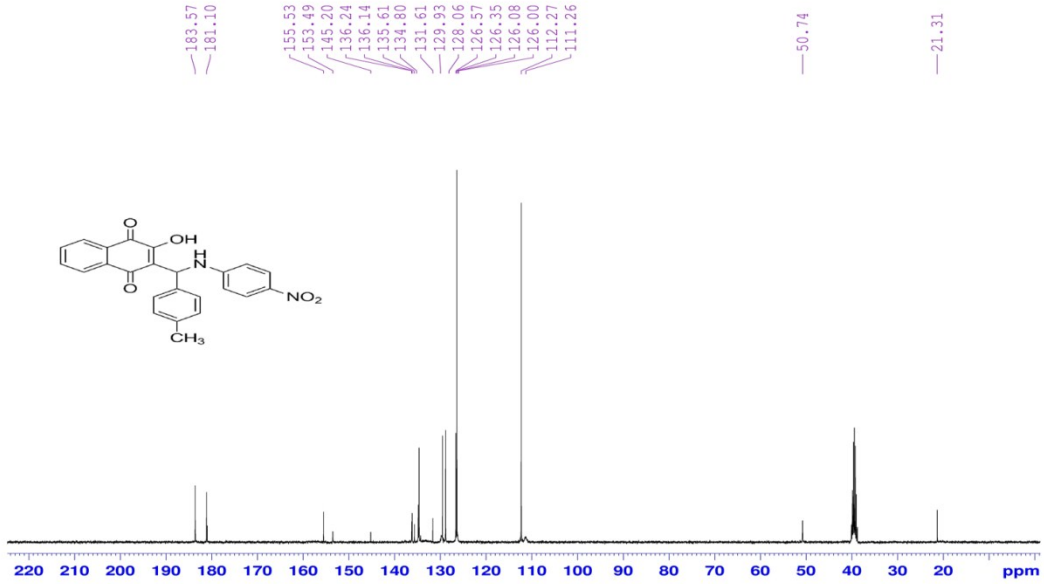
2-((4-Chlorophenyl)((4-nitrophenyl)amino)methyl)-3-hydroxynaphthalene-1,4-dione (Table 4, 4d) ; ^{1}H NMR (400 MHz, DMSO-*d*$_6$)



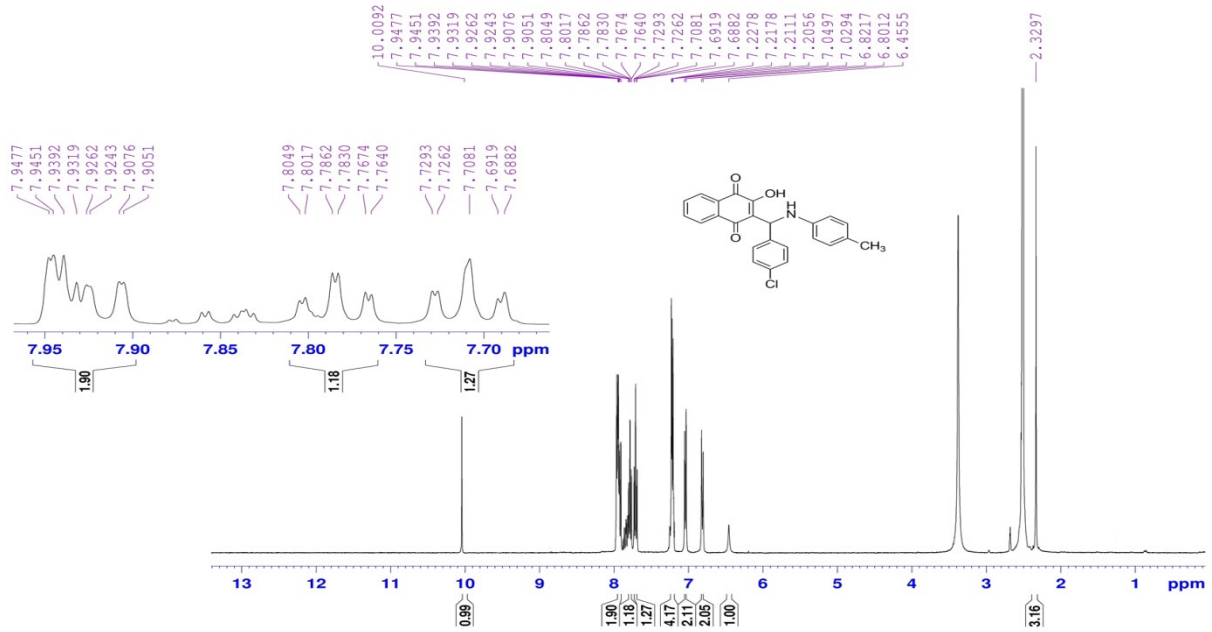
2-((4-Chlorophenyl)((4-nitrophenyl)amino)methyl)-3-hydroxynaphthalene-1,4-dione (Table 4, 4d) ; ^{13}C NMR (100 MHz, DMSO-*d*$_6$)



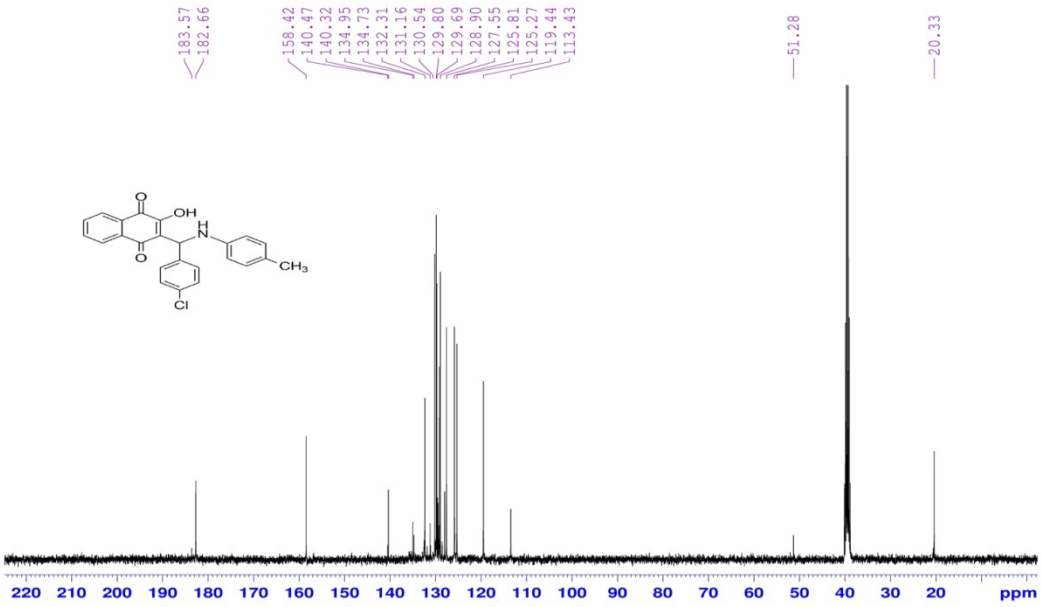
2-Hydroxy-3-(((4-nitrophenyl)amino)(p-tolyl)methyl)naphthalene-1,4-dione (Table 4, 4e) ; ^{1}H NMR (400 MHz, DMSO-d_6)



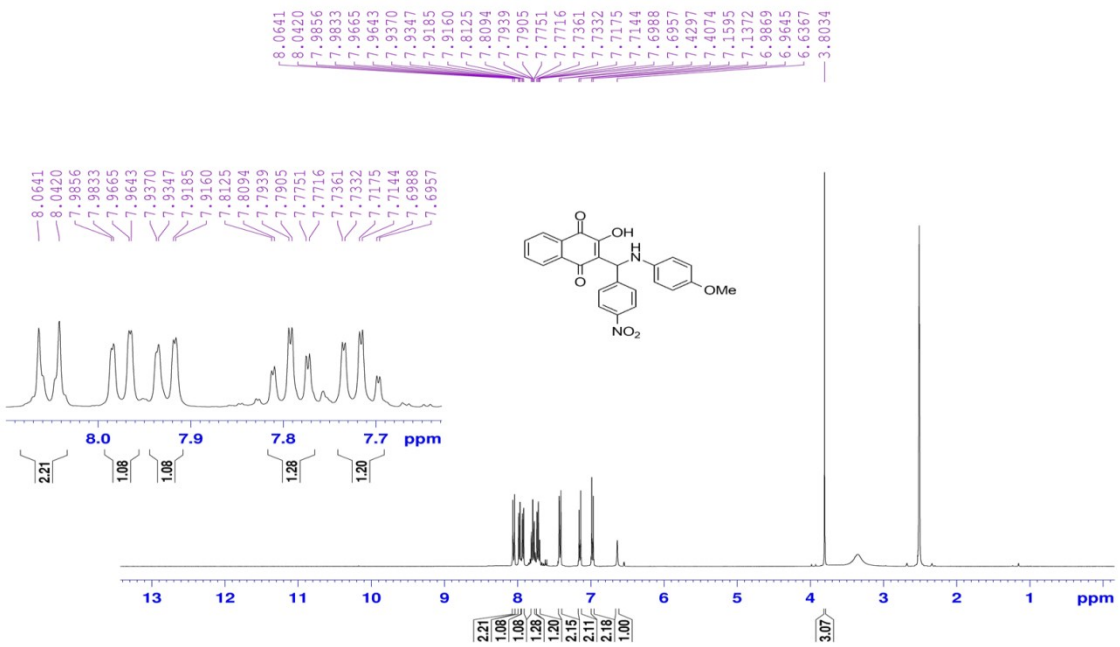
2-Hydroxy-3-(((4- nitrophenyl)amino)(p-tolyl)methyl)naphthalene-1,4-dione (Table 4, 4e) ; ^{13}C NMR (100 MHz, DMSO-d_6)



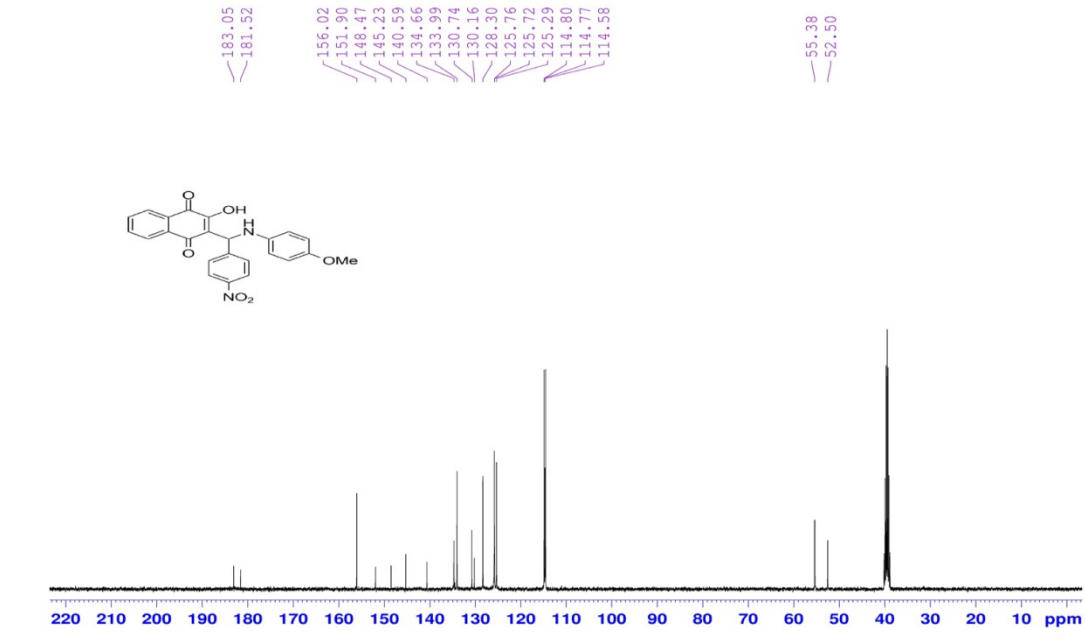
2- ((4-Chlorophenyl)(p-tolylamino)methyl)-3-hydroxynaphthalene-1,4-dione (Table 4, 4f) ; ^{1}H NMR (400 MHz, DMSO-d_6)



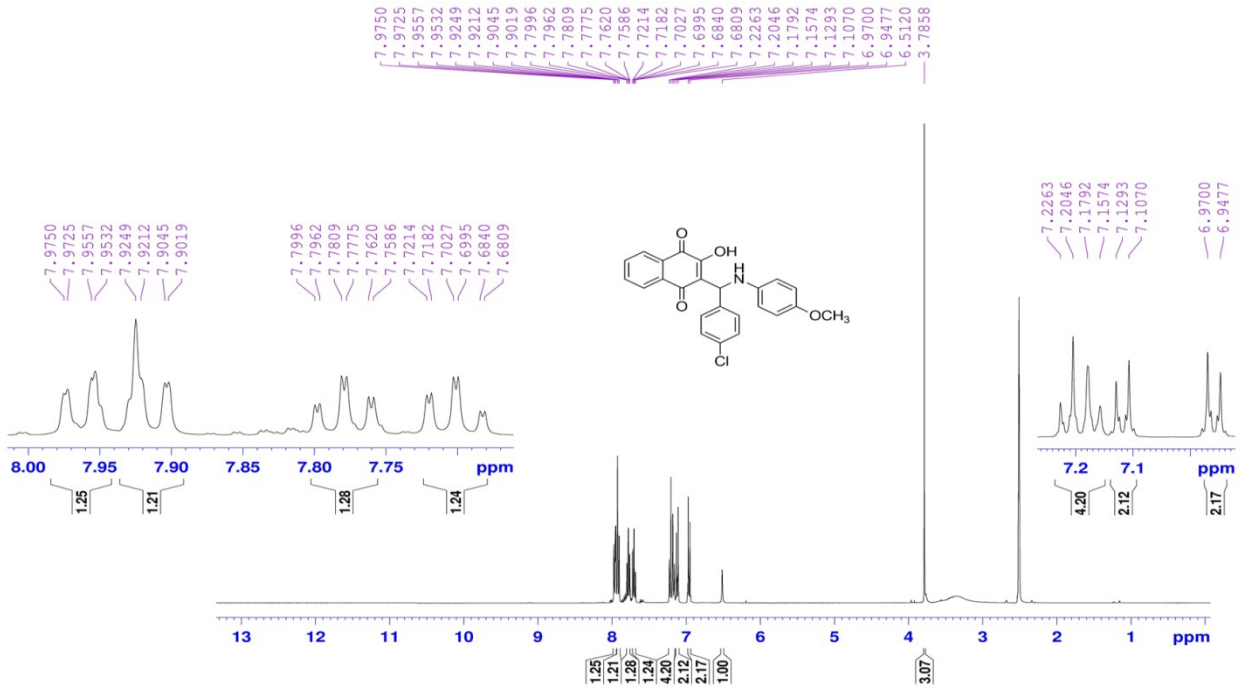
2- ((4-Chlorophenyl)(p-tolylamino)methyl)-3-hydroxynaphthalene-1,4-dione (Table 4, 4f) ; ^{13}C NMR (100 MHz, DMSO-d_6)



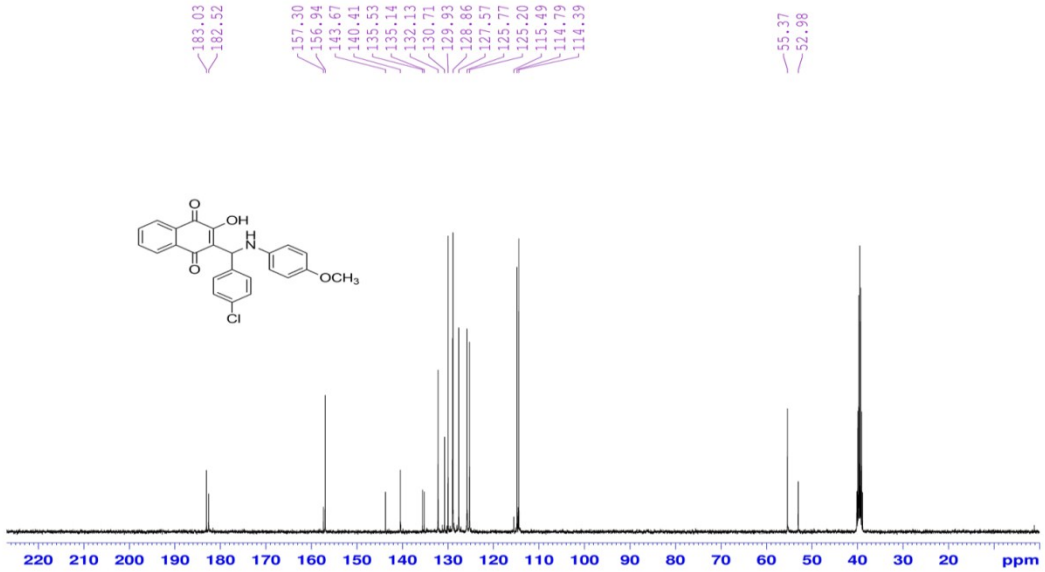
2-Hydroxy-3-(((4- methoxy phenyl) amino)(4-nitrophenyl)methyl)naphthalene-1,4- dione (Table 4, 4g); ^{13}C NMR (100 MHz, DMSO-d_6)



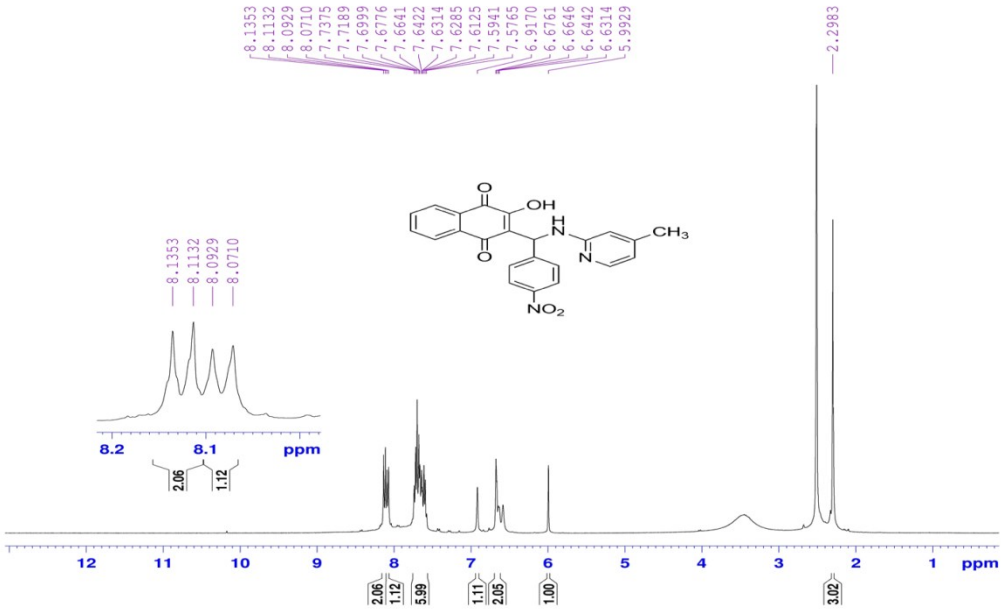
2-Hydroxy-3-(((4- methoxy phenyl) amino)(4-nitrophenyl)methyl)naphthalene-1,4- dione (Table 4, 4g); ^{1}H NMR (400 MHz, DMSO-d_6)



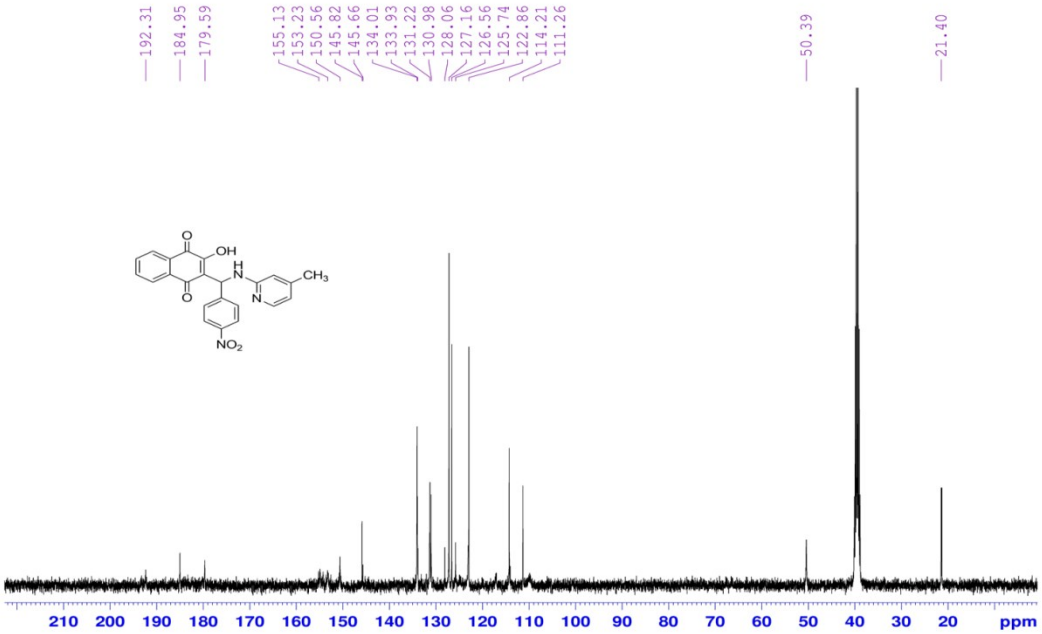
2-((4-Chlorophenyl)((4-methoxyphenyl)amino)methyl)-3- hydroxynaphthalene-1, 4-dione (Table 4, 4h); ^{1}H NMR (400 MHz, DMSO-d_6)



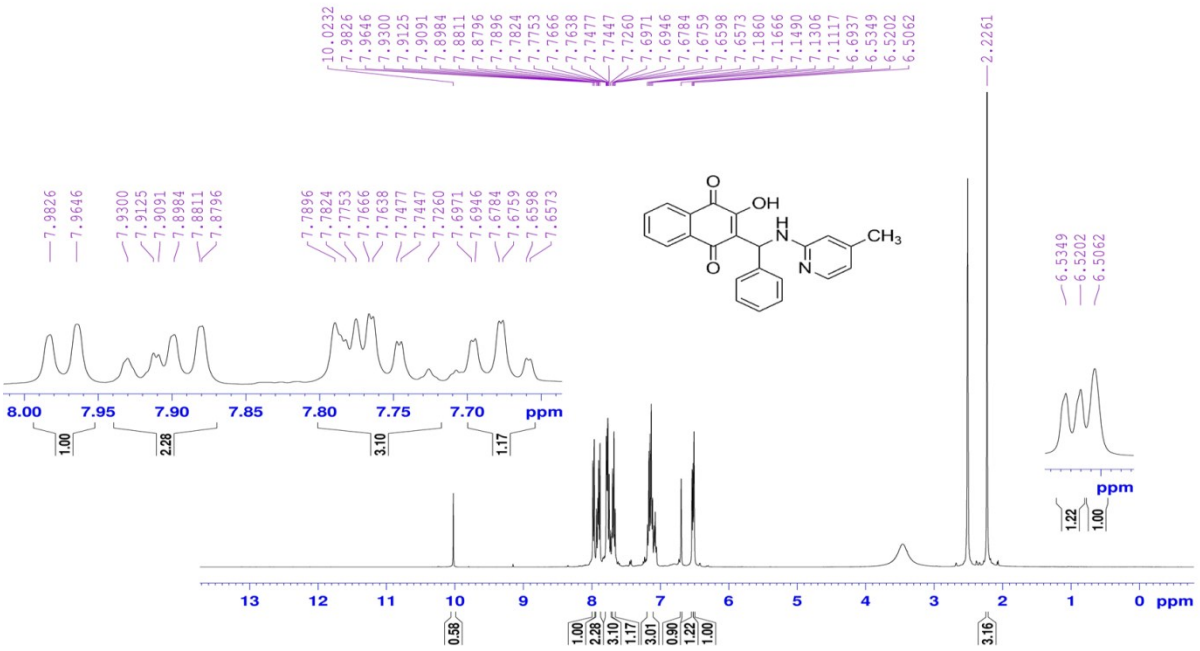
2-((4-Chlorophenyl)((4-methoxyphenyl)amino)methyl)-3- hydroxynaphthalene-1, 4-dione (Table 4, 4h); ^{13}C NMR (100 MHz, DMSO-d_6)



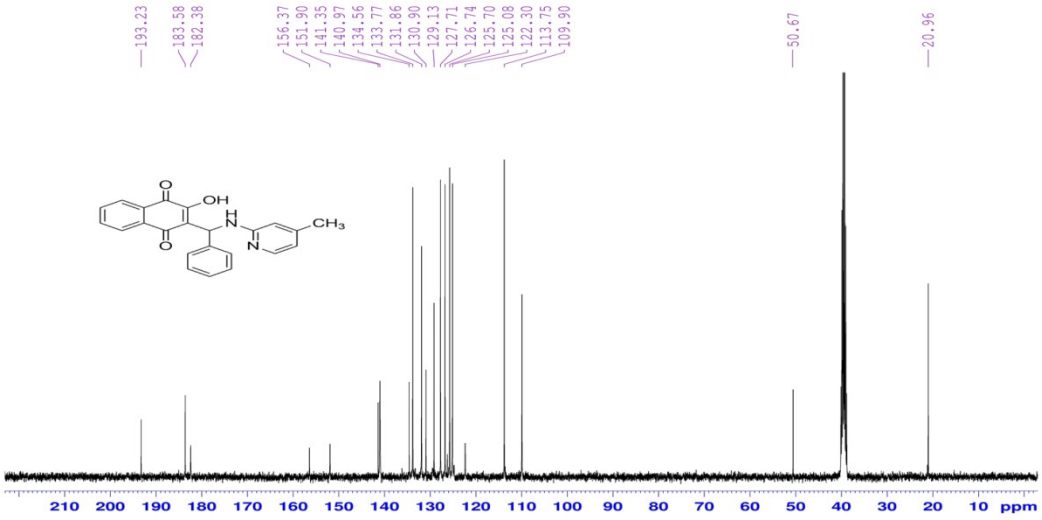
2-Hydroxy-3-(((4- methylpyridin-2-yl) amino)(4- nitrophenyl)methyl)naphthalene-1,4- dione (Table 4, 4i) ; ^{1}H NMR (400 MHz, DMSO-d_6)



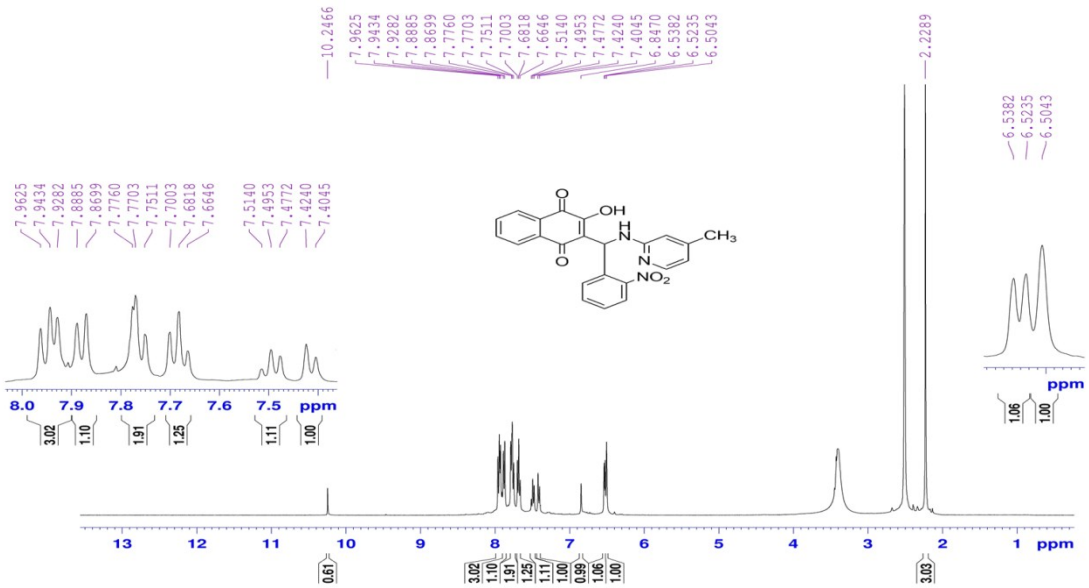
2-Hydroxy-3-(((4- methylpyridin-2-yl) amino)(4- nitrophenyl)methyl)naphthalene-1,4- dione (Table 4, 4i) ; ^{13}C NMR (100 MHz, DMSO-d_6)



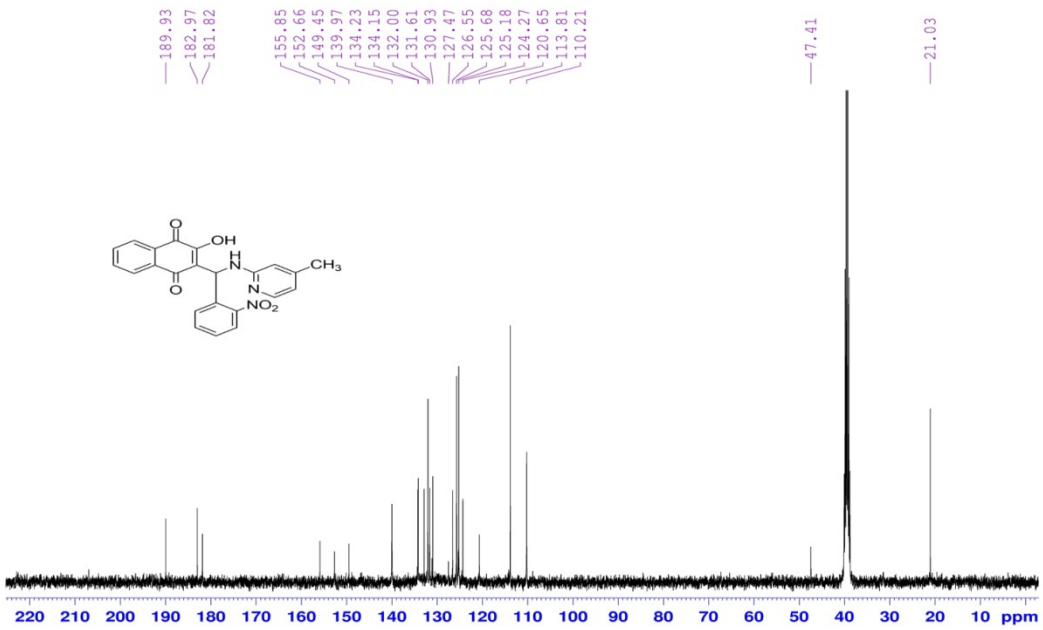
2-Hydroxy-3-(((4- methyl pyridine-2-y1)amino)(phenl) methyl)naphthalene-1,4-dione (Table 4, 4j) ; ^{1}H NMR (400 MHz, DMSO-d_6)



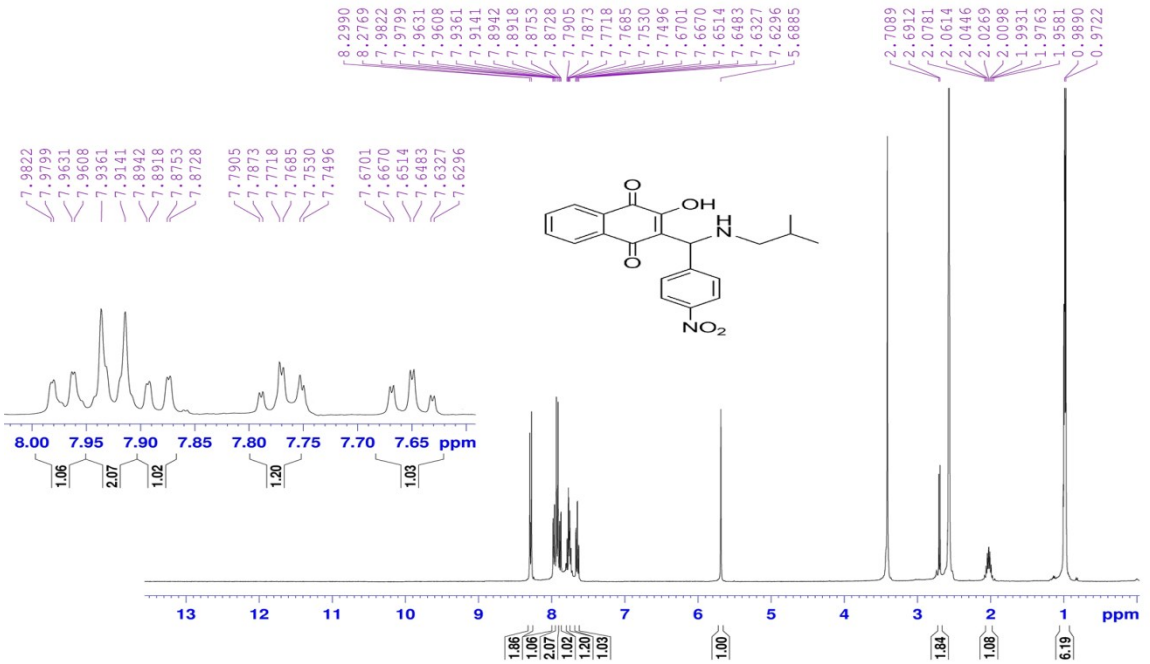
2-Hydroxy-3-(((4- methyl pyridine-2-y1)amino)(phenl) methyl)naphthalene-1,4-dione (Table 4, 4j) ; ^{13}C NMR (100 MHz, DMSO-d_6)



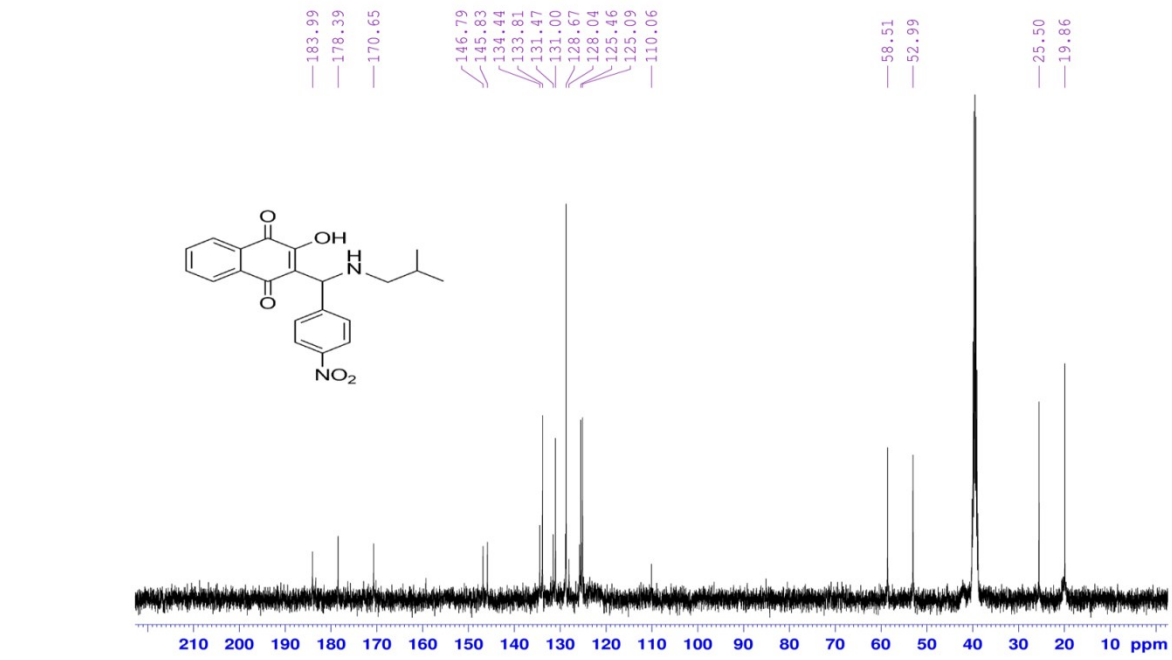
2-Hydroxy-3-(((4- methyl pyridine-2-y1)amino)(2-nitrophenyl)methyl)naphthalene- 1,4-dione (Table 4, 4k) ; ^{1}H NMR (400 MHz, DMSO-d_6)



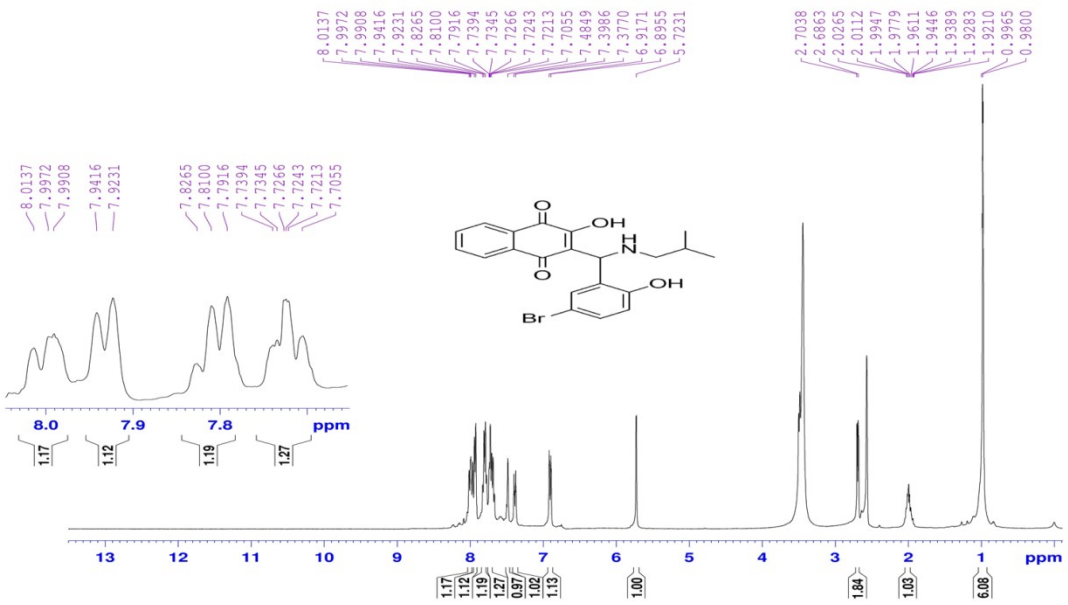
2-Hydroxy-3-(((4- methyl pyridine-2-y1)amino)(2-nitrophenyl)methyl)naphthalene- 1,4-dione (Table 4, 4k) ; ^{13}C NMR (100 MHz, DMSO-d_6)



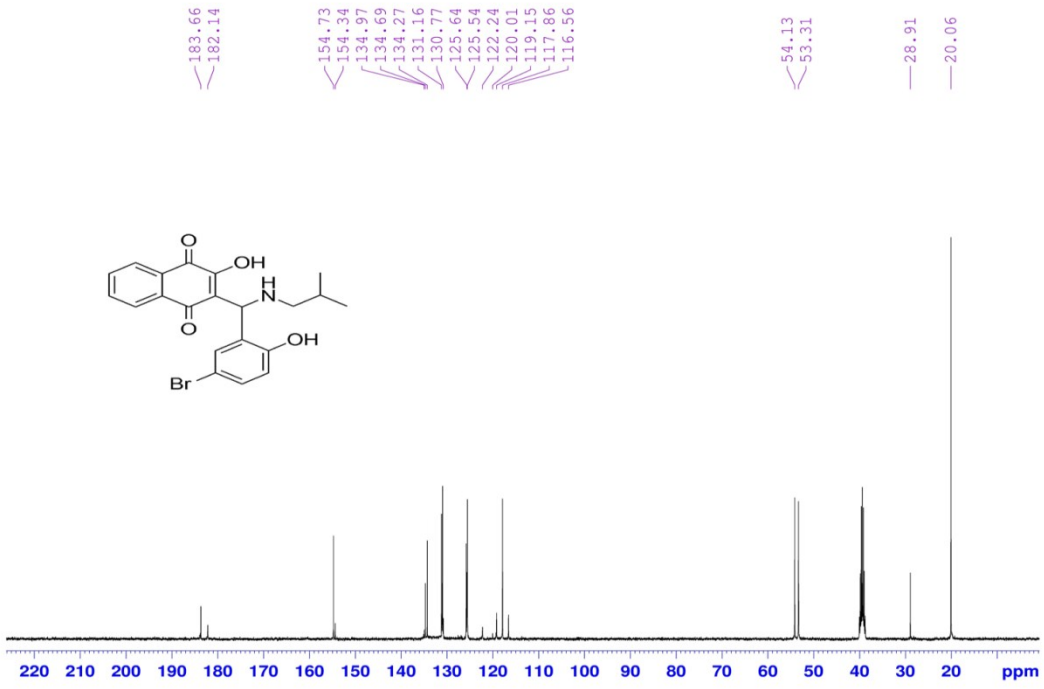
2- Hydroxy-3-((isobutylamino)(4-nitrophenyl)methyl)naphthalene-1,4-dione (Table 4, 4l) ; ^{1}H NMR (400 MHz, DMSO-d_6)



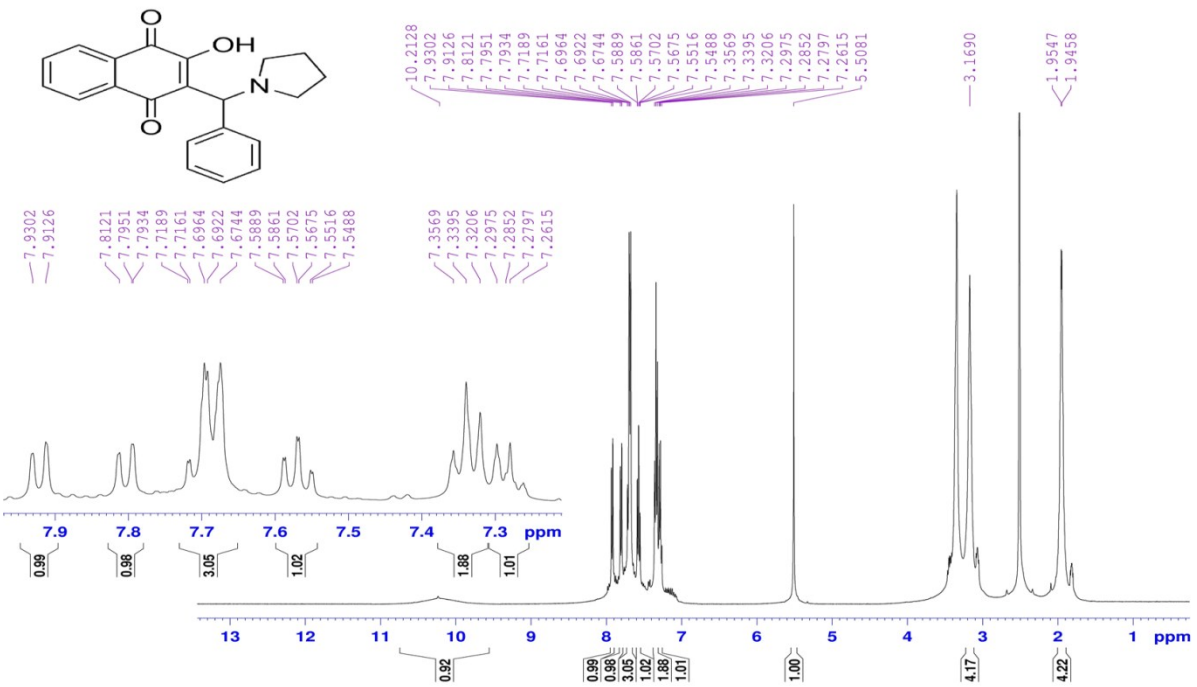
2- Hydroxy-3-((isobutylamino)(4-nitrophenyl)methyl)naphthalene-1,4-dione (Table 4, 4l) ; ^{13}C NMR (100 MHz, DMSO-d_6)



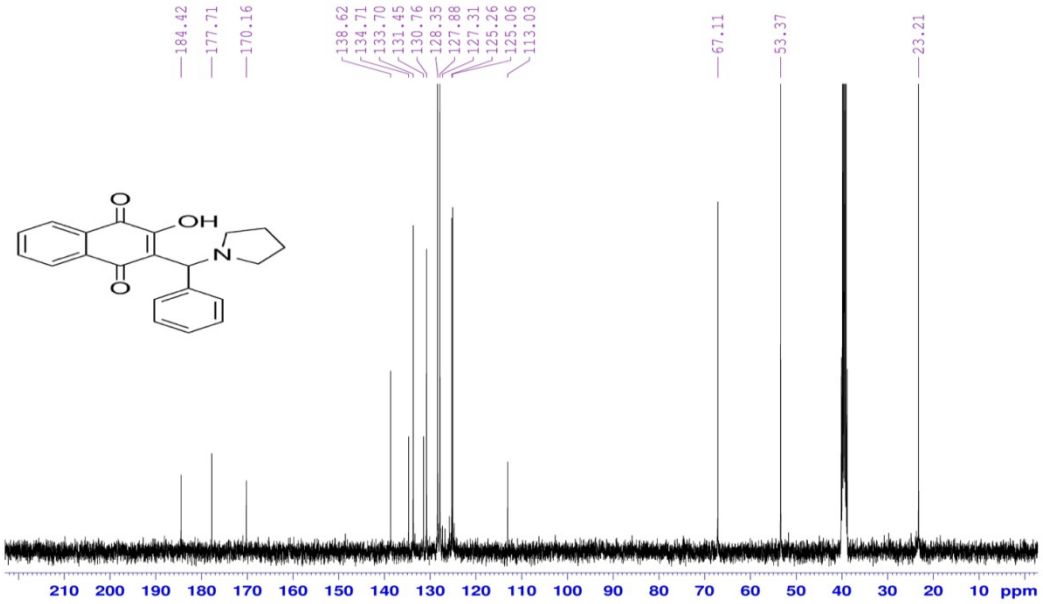
2-((5- Bromo-2- hydroxyphenyl)(isobutylamino)methyl)-3-hydroxynaphthalene-1,4-dione (Table 4, 4m); ^{1}H NMR (400 MHz, DMSO-d_6)



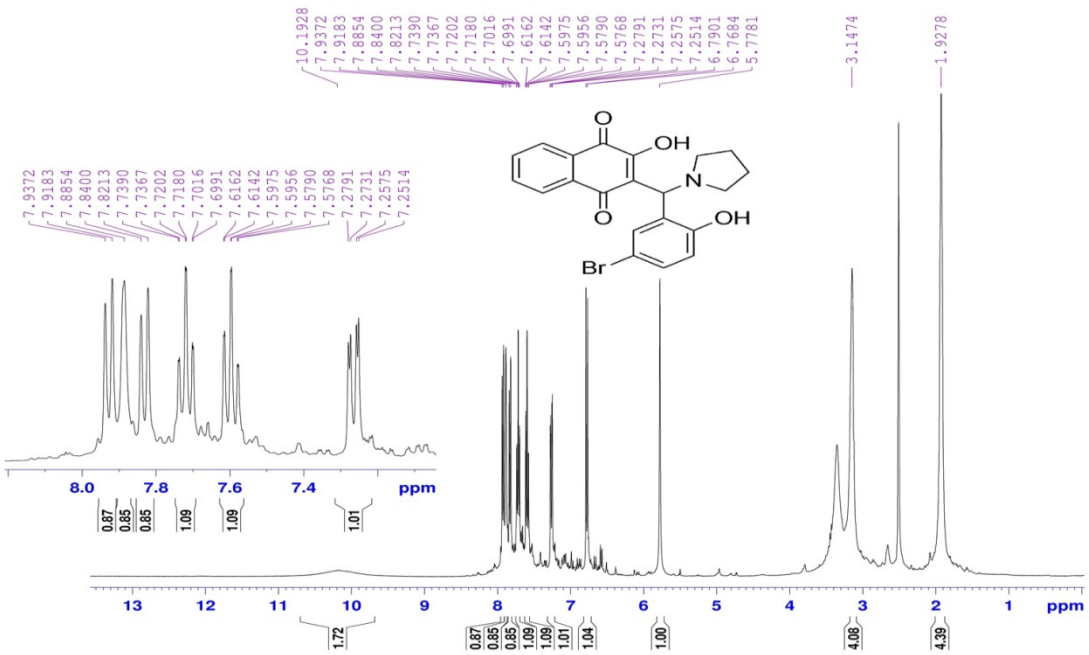
2-((5- Bromo-2- hydroxyphenyl)(isobutylamino)methyl)-3-hydroxynaphthalene-1,4-dione (Table 4, 4m); ^{13}C NMR (100 MHz, DMSO-d_6)



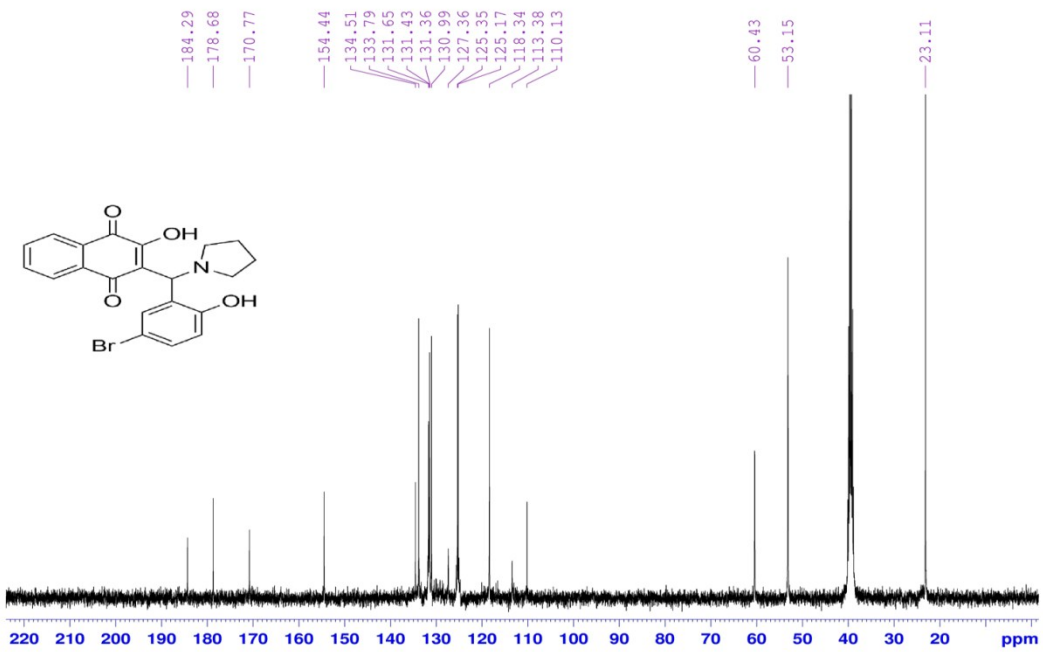
2- Hydroxy-3-(phenyl(pyrrolidin-1-yl)methyl)naphthalene-1,4-dione (Table 4, 4n) ; ^{1}H NMR (400 MHz, DMSO-d_6)



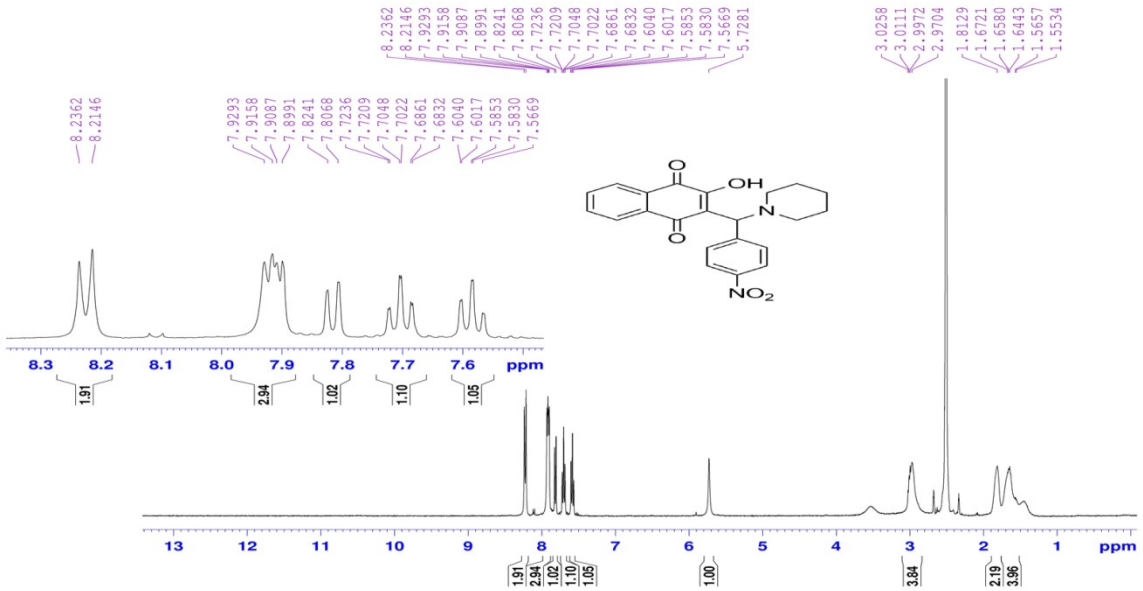
2- Hydroxy-3-(phenyl(pyrrolidin-1-yl)methyl)naphthalene-1,4-dione (Table 4, 4n) ; ^{13}C NMR (100 MHz, DMSO-d_6)



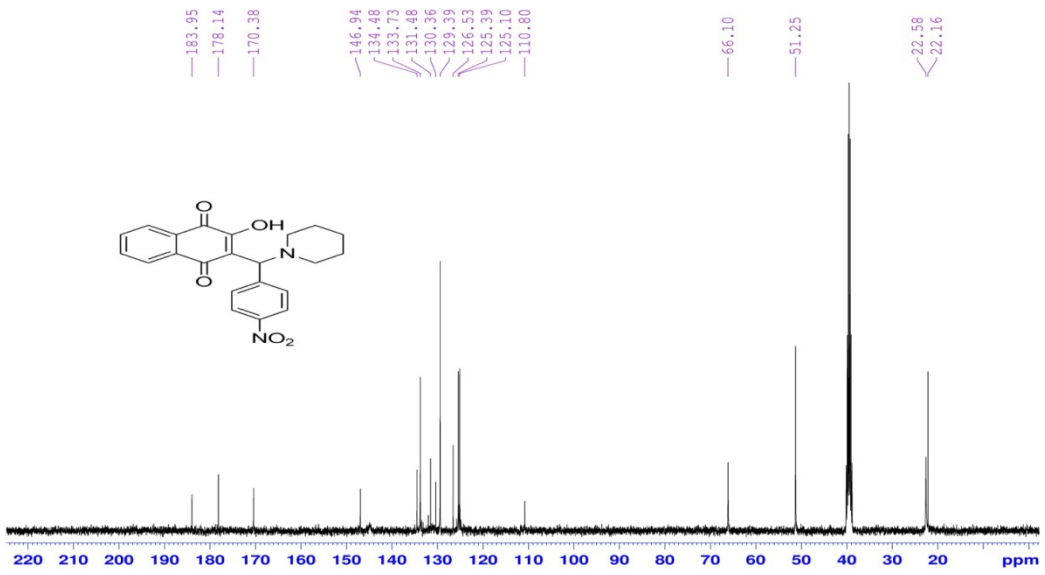
2- ((5-Bromo-2-hydroxyphenyl)(pyrrolidin-1-y1)methyl)-3-hydroxynaphthalene-1,4-dione (Table 4, 4o) ; ^{1}H NMR (400 MHz, DMSO-d_6)



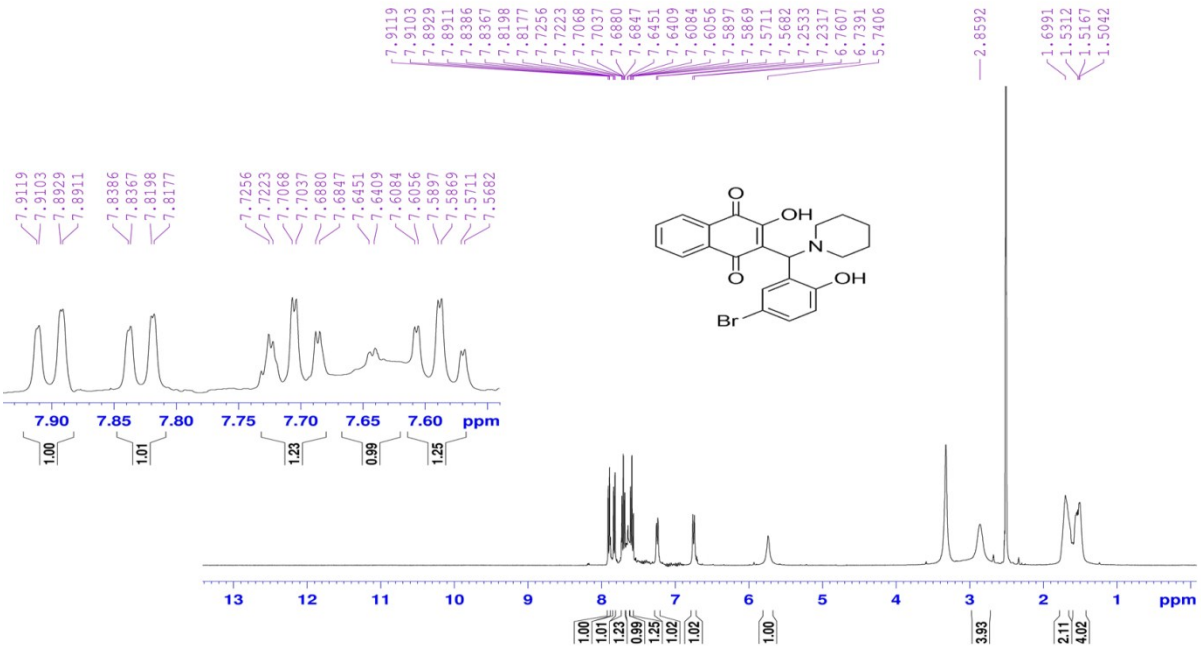
2- ((5- Bromo-2-hydroxyphenyl)(pyrrolidin-1-y1)methyl)-3-hydroxynaphthalene-1,4-dione (Table 4, 4o) ; ^{13}C NMR (100 MHz, DMSO-d_6)



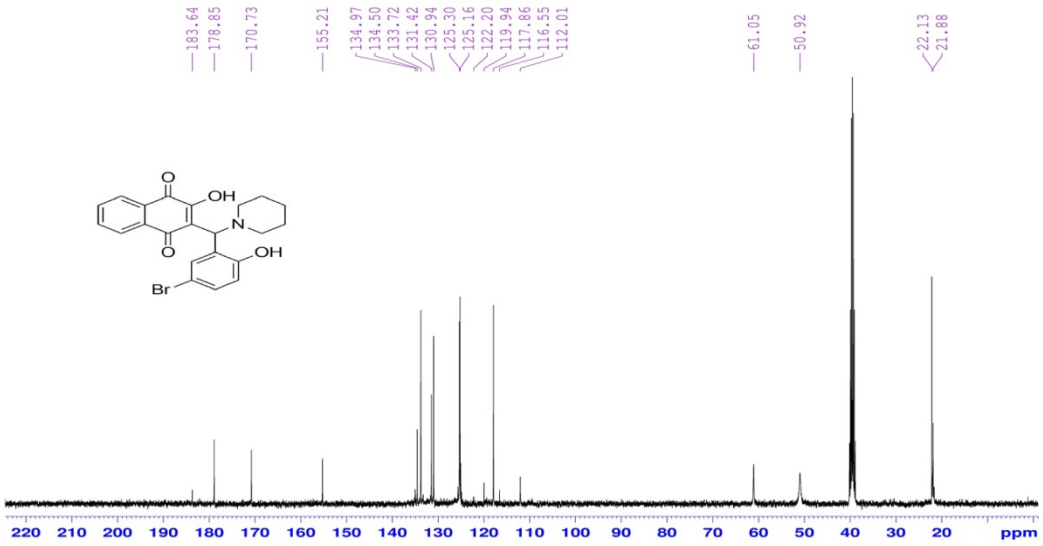
2- Hydroxy-3-((4-nitrophenyl)(piperidin-1-y1)methyl)naphthalene-1,4-dione (Table 4, 4p); ^{1}H NMR (400 MHz, DMSO-d_6)



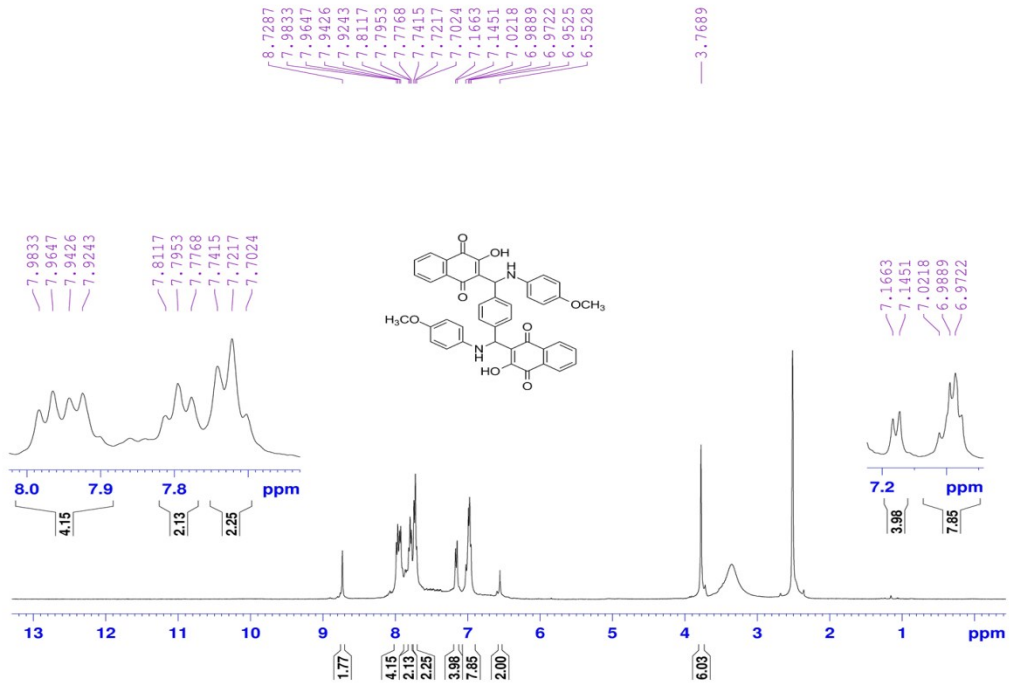
2- Hydroxy-3-((4-nitrophenyl)(piperidin-1-y1)methyl)naphthalene-1,4-dione (Table 4, 4p); ^{13}C NMR (100 MHz, DMSO-d_6)



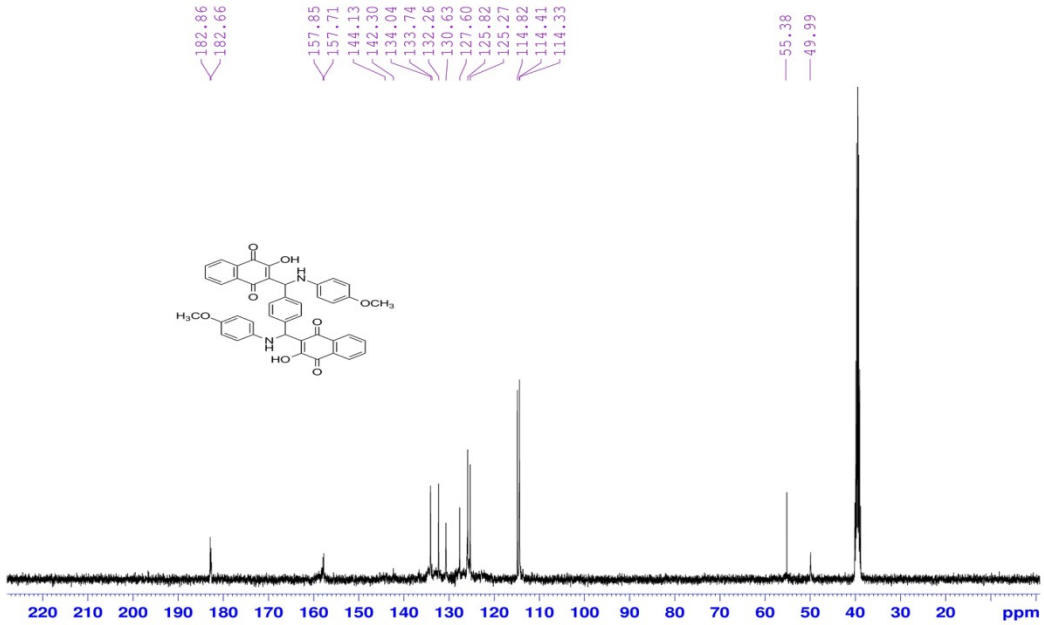
2-((5- Bromo-2- hydroxy phenyl) (piperidin-1- yl) methyl) -3- hydroxynaphthalene -1,4- dione (Table 4, 4q); ^{1}H NMR (400 MHz, DMSO-d_6)



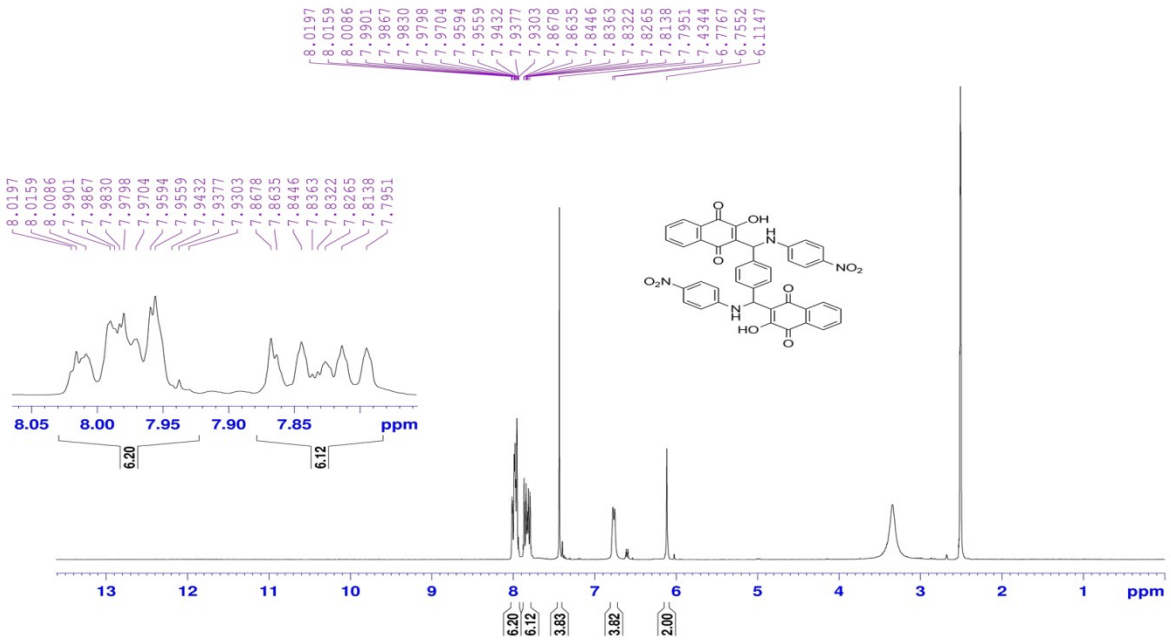
2-((5- Bromo-2- hydroxy phenyl) (piperidin-1- yl) methyl) -3- hydroxynaphthalene -1,4- dione (Table 4, 4q); ^{13}C NMR (100 MHz, DMSO-d_6)



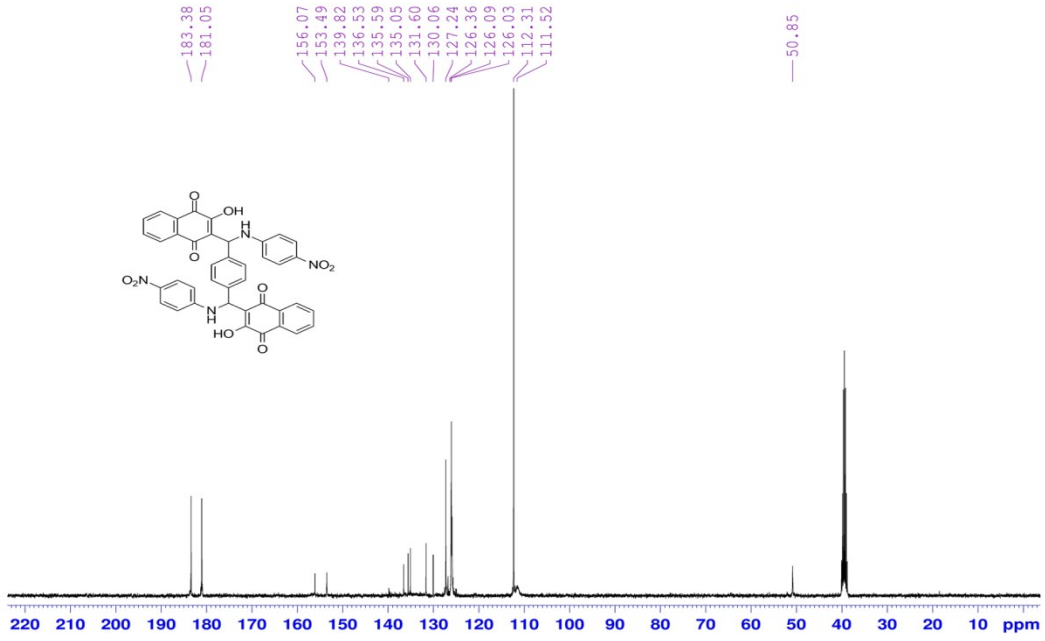
3,3'-(1,4-Phenylenebis(((4- methoxy phenyl)amino)methylene))bis(2-hydroxy naphthalene-1,4-dione) (Table 5, 6a); ^{1}H NMR (400 MHz, DMSO-d_6)



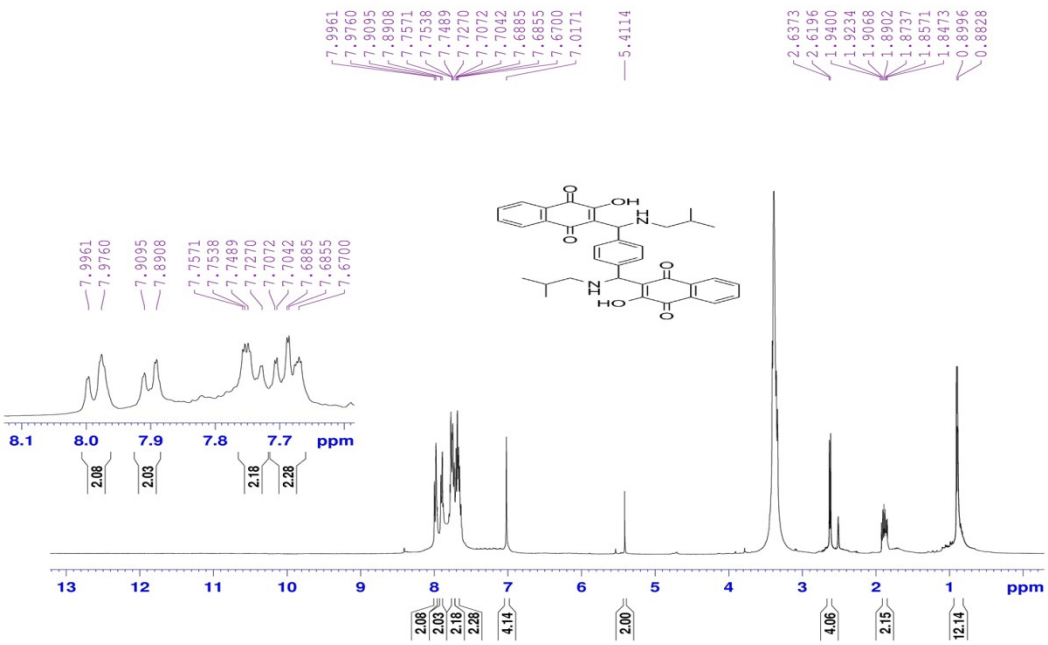
3,3'-(1,4-Phenylenebis(((4- methoxy phenyl)amino)methylene))bis(2-hydroxy naphthalene-1,4-dione) (Table 5, 6a); ^{13}C NMR (100 MHz, DMSO-d_6)



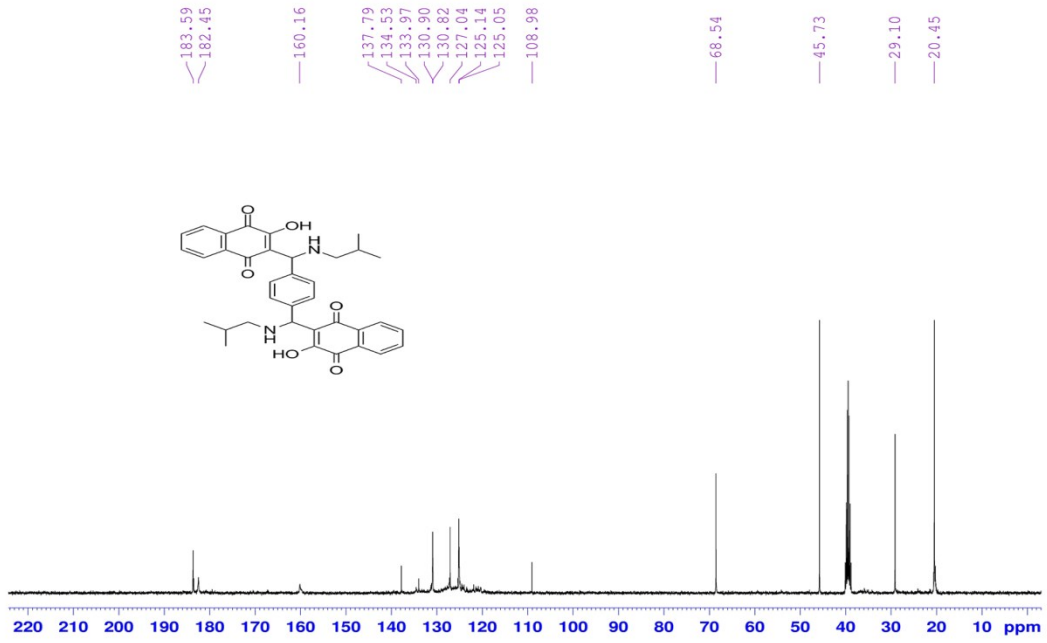
3,3'–(1,4-Phenylenebis (((4-nitrophenyl)amino)methylene))bis(2- hydroxynaphthalene-1,4-dione) (Table 5, 6b); ^{1}H NMR (400 MHz, DMSO-d_6)



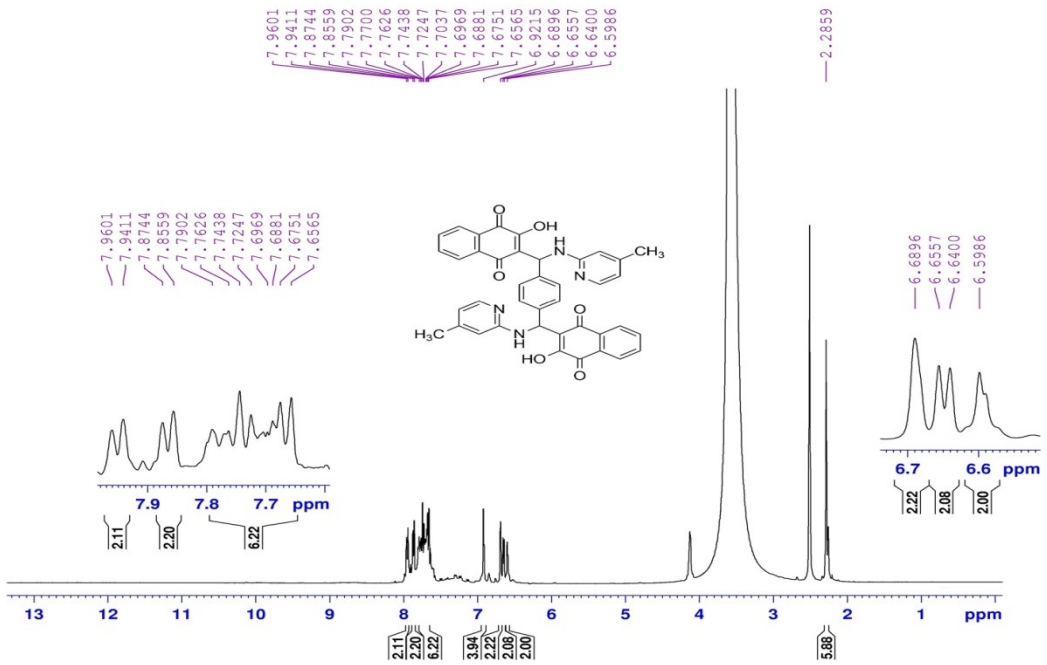
3,3'–(1,4-Phenylenebis(((4-nitrophenyl)amino)methylene))bis(2- hydroxynaphthalene-1,4-dione) (Table 5, 6b); ^{13}C NMR (100 MHz, DMSO-d_6)



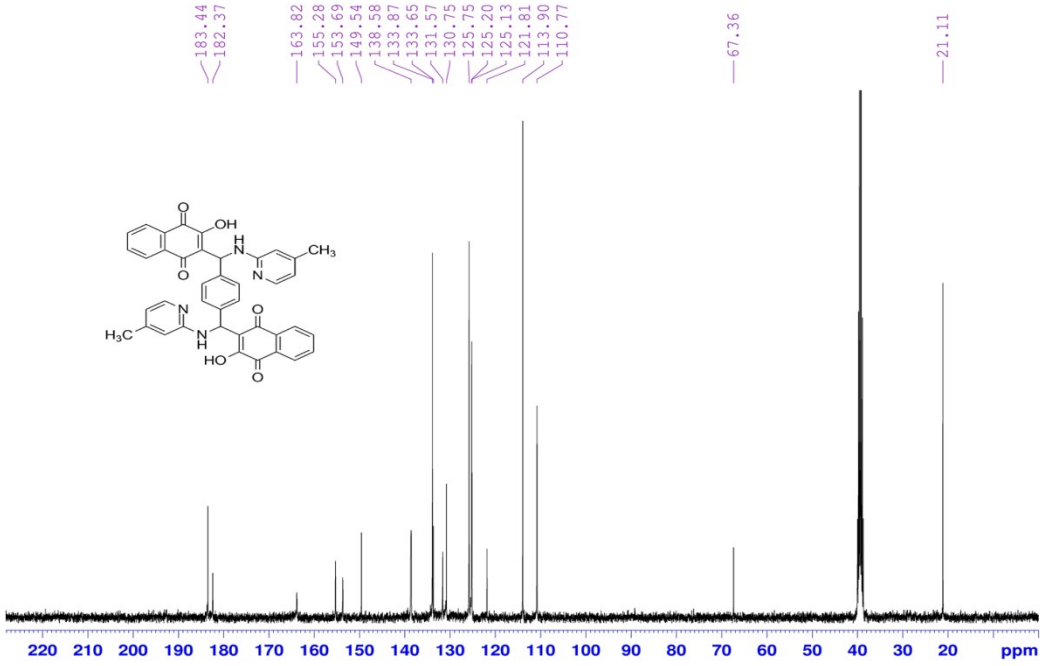
3,3'-(1,4-Phenylenebis((isobutylamino)methylene))bis(2- hydroxynaphthalene- 1,4- dione) (Table 5, 6c); ^{1}H NMR (400 MHz, DMSO-d_6)



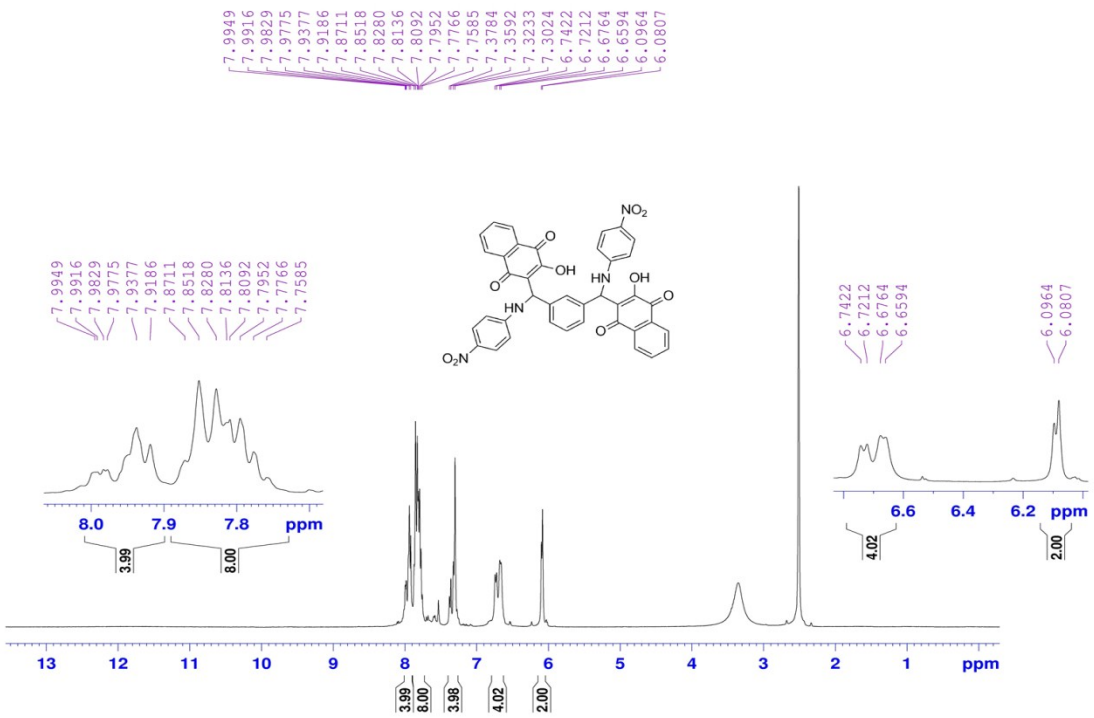
3,3'-(1,4-Phenylenebis((isobutylamino)methylene))bis(2- hydroxynaphthalene- 1,4- dione) (Table 5, 6c); ^{13}C NMR (100 MHz, DMSO-d_6)



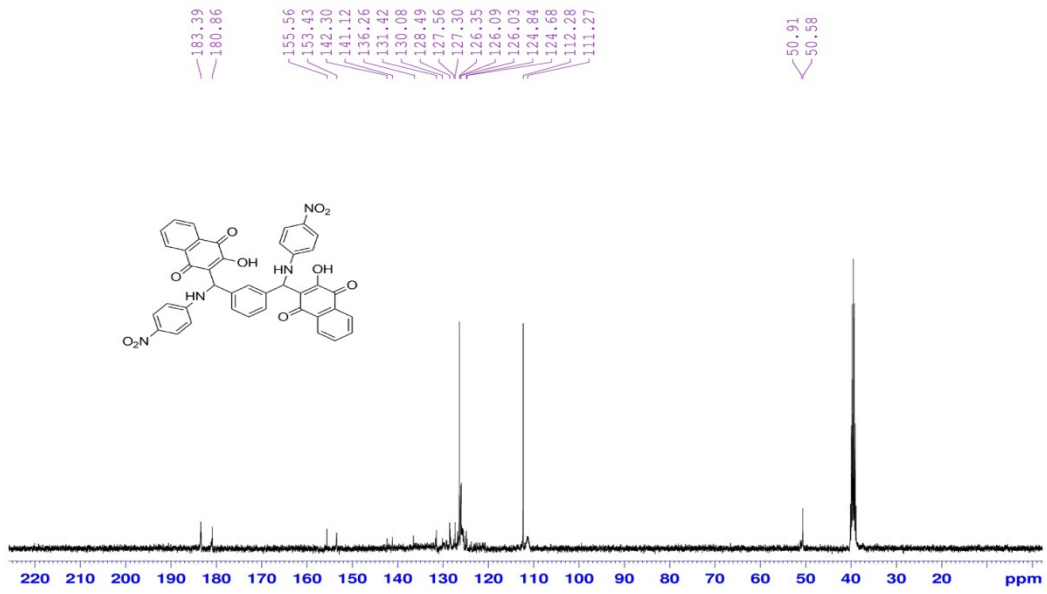
3,3'(1,4-Phenylenebis (((4-methylpyridin-2-yl) amino)methylene))bis(2-hydroxynaphthalene-1,4-dione) (Table 5, 6d); ^{1}H NMR (400 MHz, DMSO-d_6)



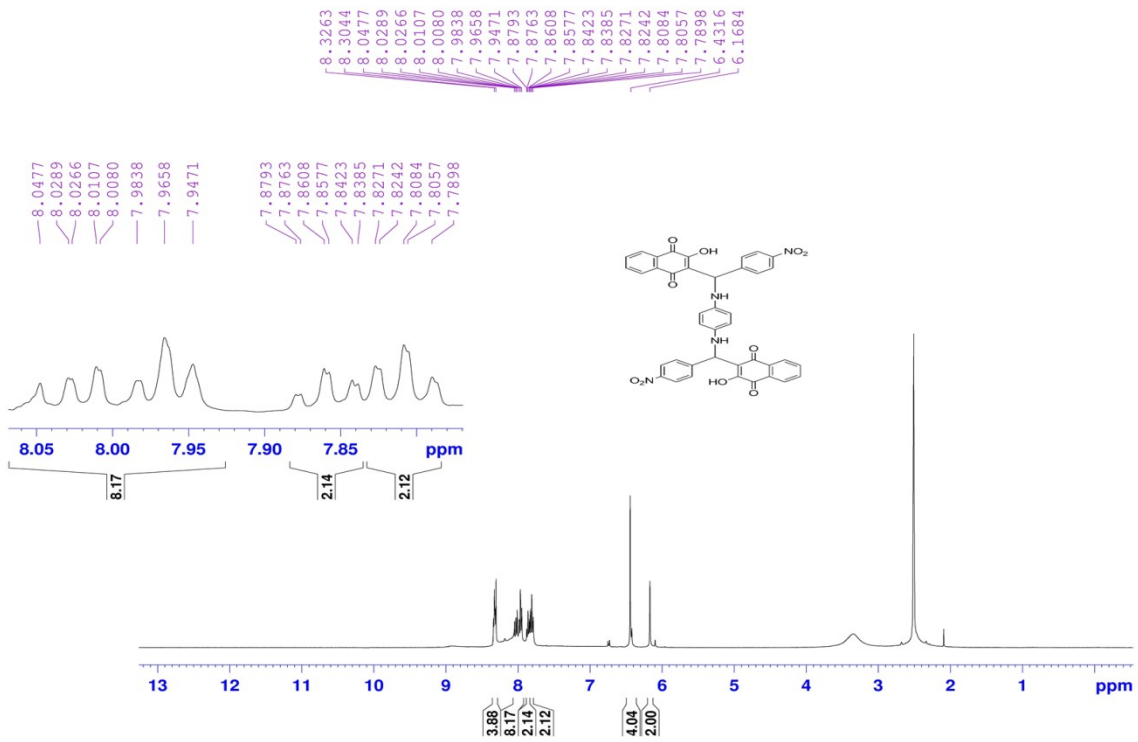
3,3'(1,4-Phenylenebis(((4-methylpyridin-2-yl) amino)methylene))bis(2-hydroxynaphthalene-1,4-dione) (Table 5, 6d); ^{13}C NMR (100 MHz, DMSO-d_6)



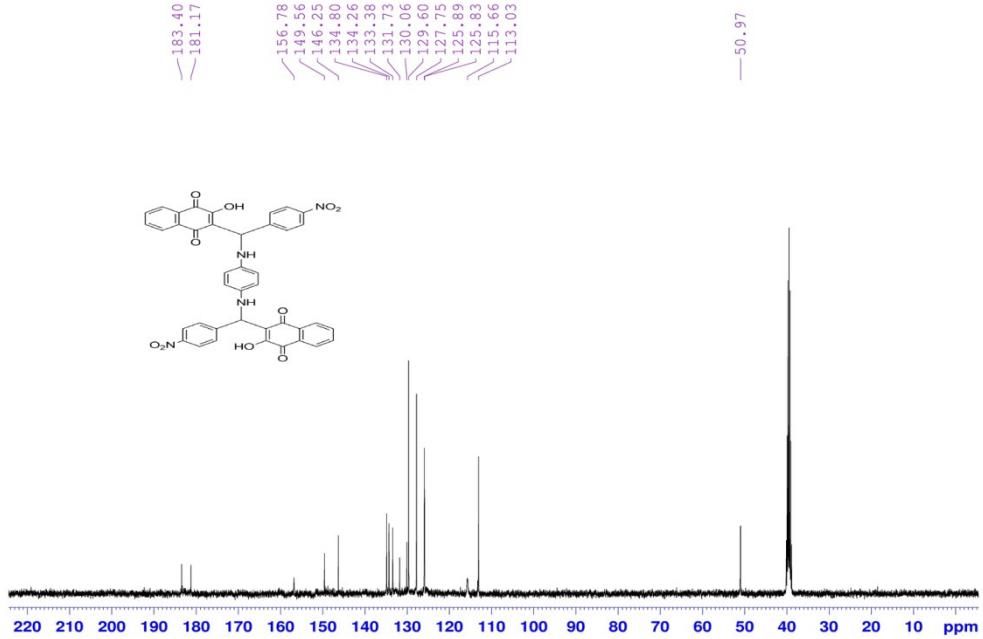
3,3'-(1,3-Phenylenebis(((4-nitrophenyl)amino)methylene))bis(2-hydroxy naphthalene-1,4-dione) (Table 5, 6e); ^{1}H NMR (400 MHz, DMSO-d_6)



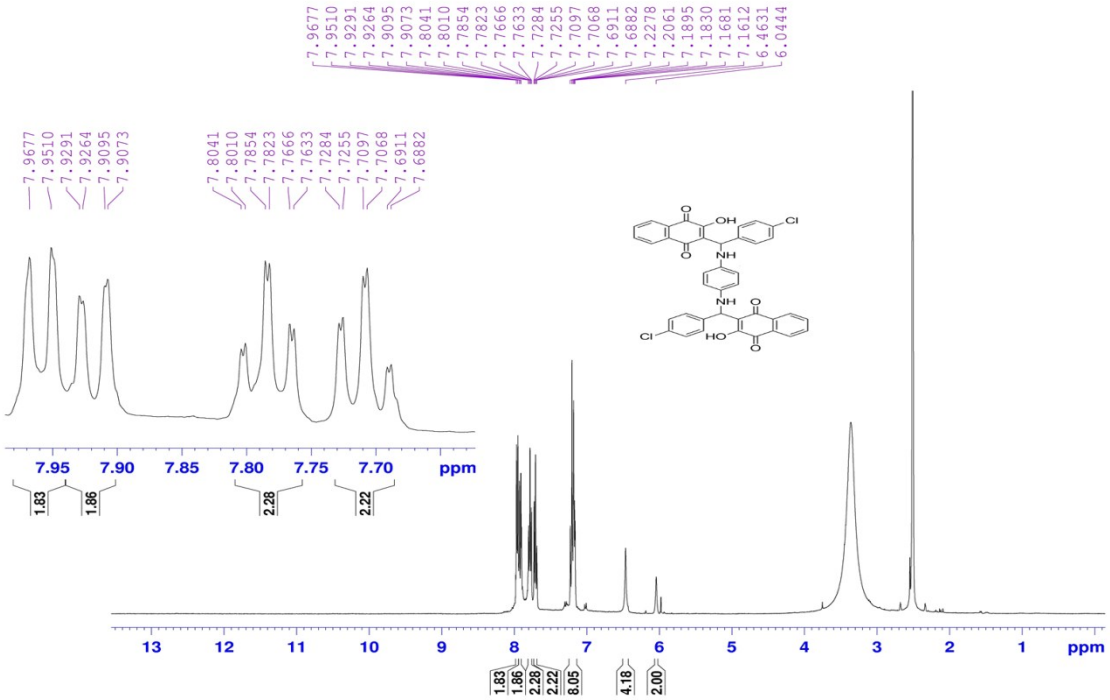
3,3'-(1,3-Phenylenebis(((4-nitrophenyl)amino)methylene))bis(2-hydroxynaphthalene-1,4-dione) (Table 5, 6e); ^{13}C NMR (100 MHz, DMSO-d_6)



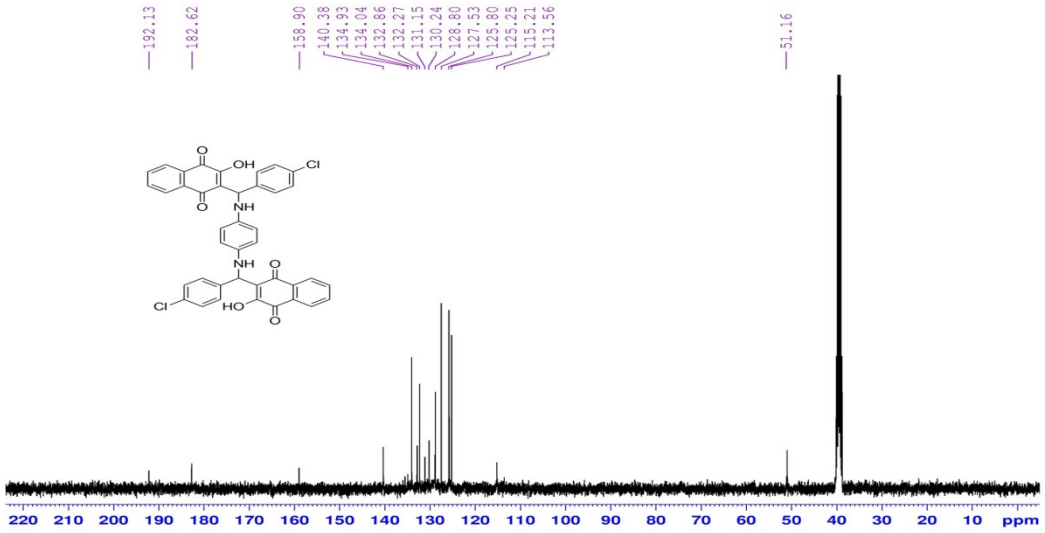
3,3'-((1,4-Phenylenebis(azanediyl))bis((4-nitrophenyl)methylene))bis (2-hydroxynaphthalene- 1,4-dione) (Table 5, 8a); ^{1}H NMR (400 MHz, DMSO-d_6)



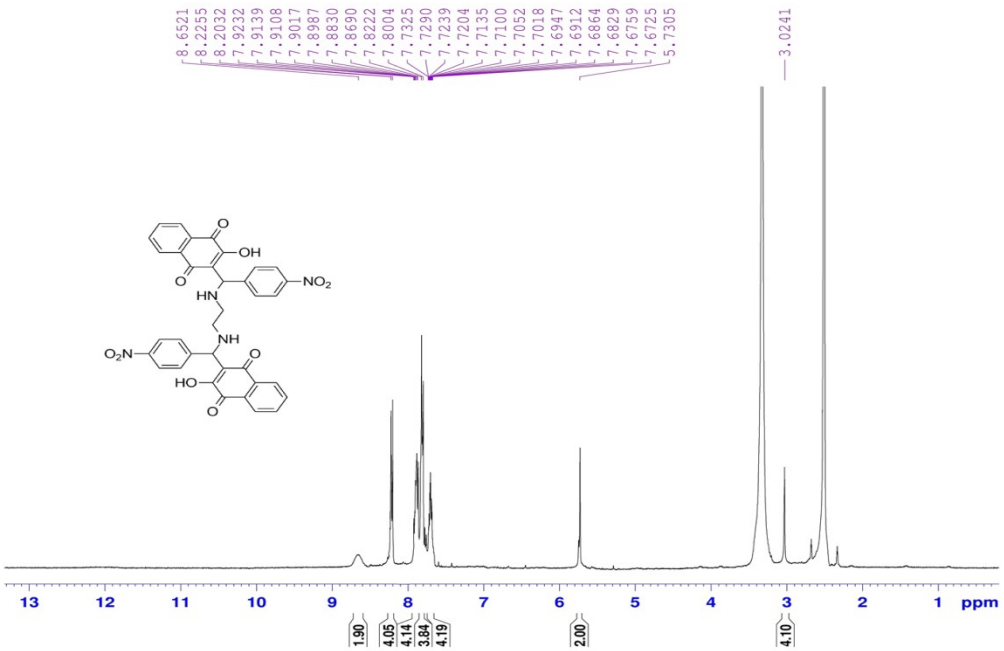
3,3'-((1,4-Phenylenebis(azanediyl))bis((4-nitrophenyl)methylene))bis (2-hydroxynaphthalene- 1,4-dione) (Table 5, 8a); ^{13}C NMR (100 MHz, DMSO-d_6)



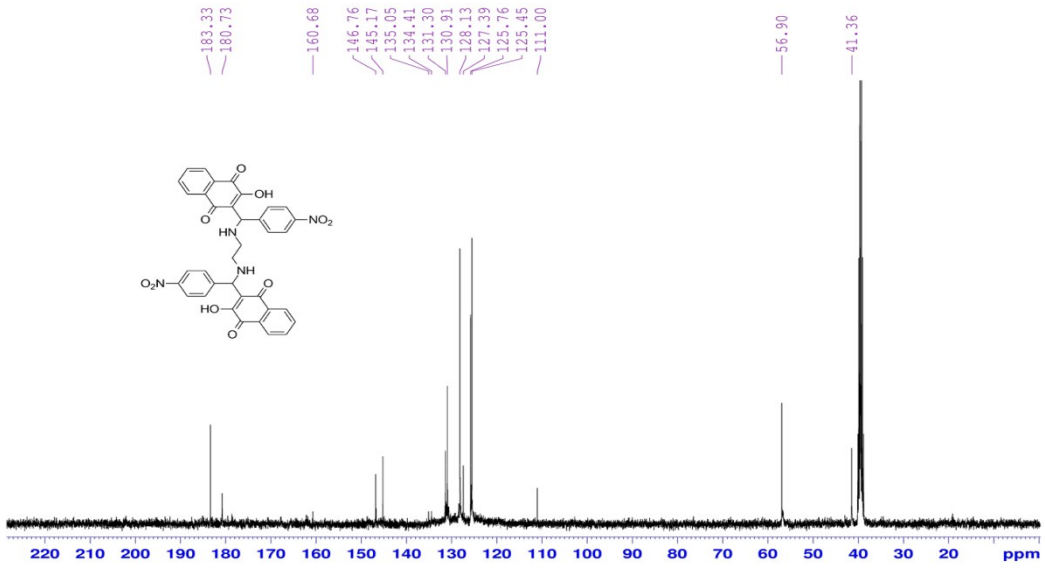
3,3'-((1,4-Phenylenebis(azanediyl))bis((4-chlorophenyl)methylene))bis(2-hydroxy naphthalene-1,4-dione) (Table 5, 8b); ^{1}H NMR (400 MHz, DMSO-d_6)



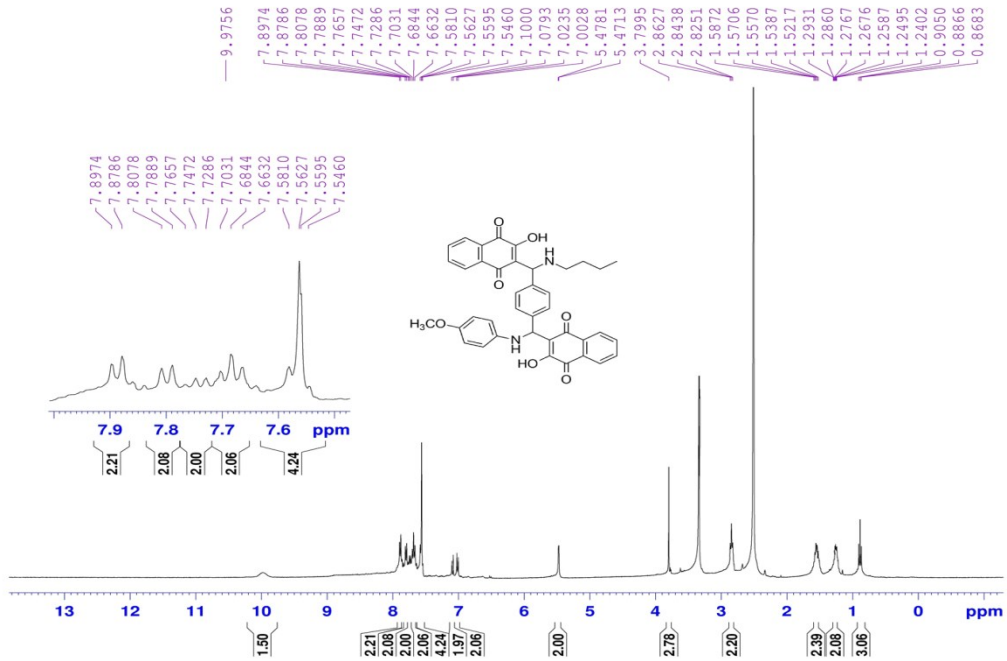
3,3'-((1,4-Phenylenebis(azanediyl))bis((4-chlorophenyl)methylene))bis(2-hydroxy naphthalene-1,4-dione) (Table 5, 8b); ^{13}C NMR (100 MHz, DMSO-d_6)



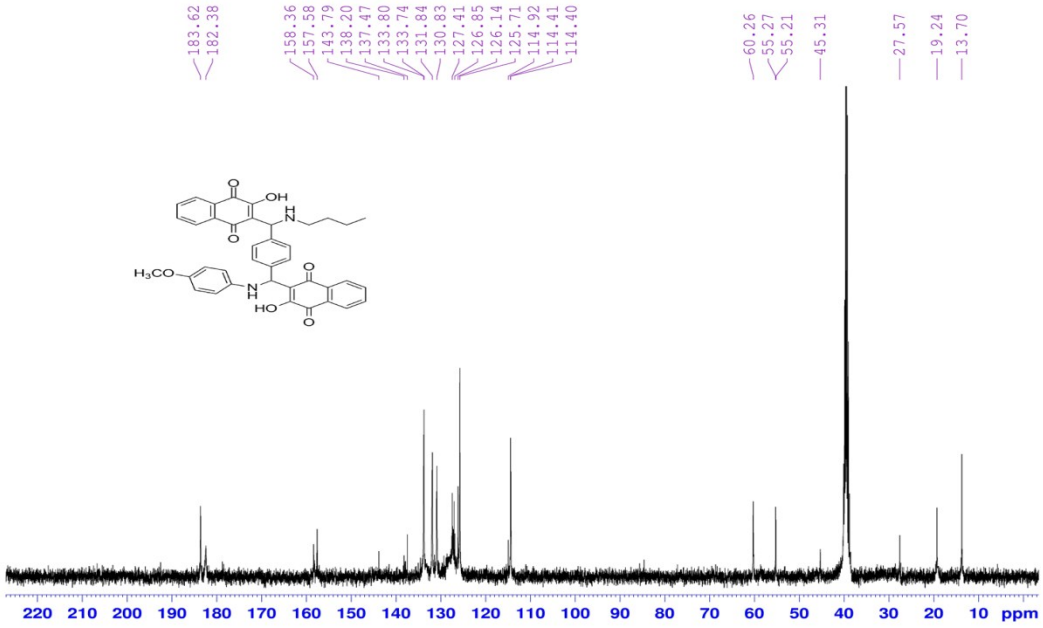
3,3'-((Ethane-1,2-diylbis(azanediyl))bis((4-nitrophenyl)methylene))bis(2-hydroxynaphthalene-1,4-dione) (Table 5, 8c); ^{1}H NMR (400 MHz, DMSO-d_6)



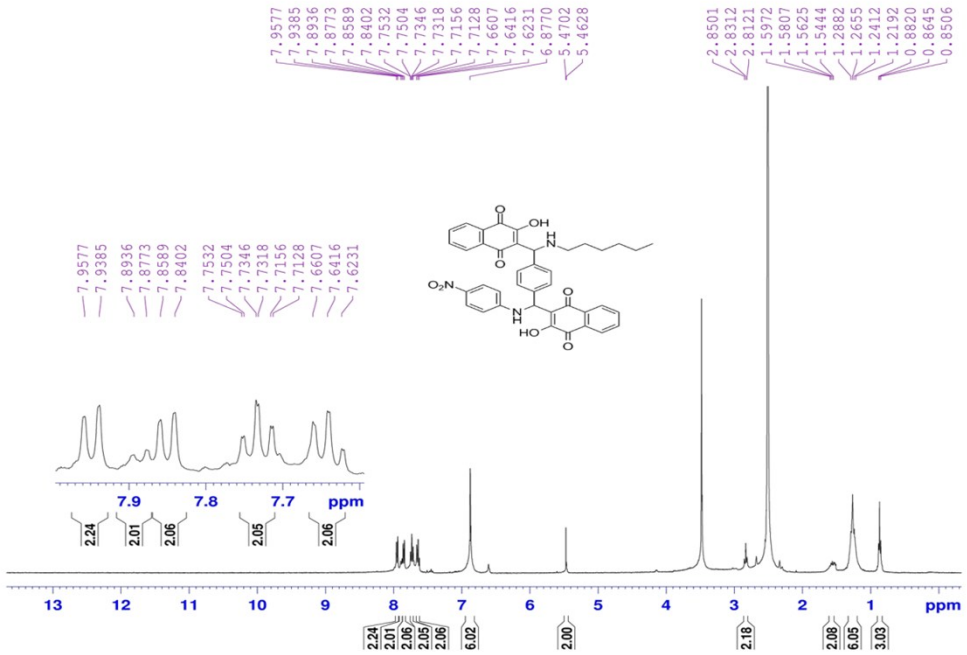
3,3'-((Ethane-1,2-diylbis(azanediyl))bis((4-nitrophenyl)methylene))bis(2-hydroxynaphthalene-1,4-dione) (Table 5, 8c); ^{1}H NMR (400 MHz, DMSO-d_6)



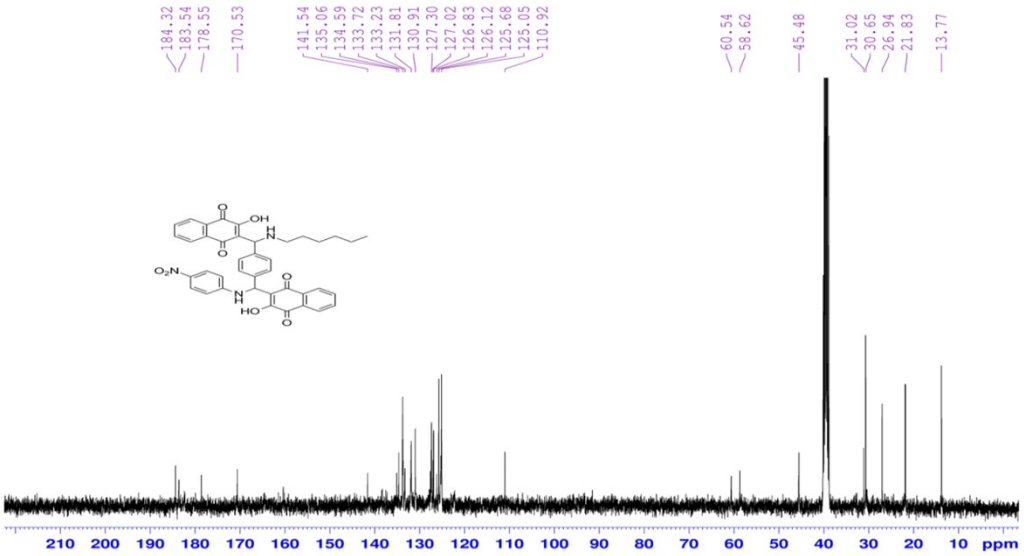
2-((4-((butylamino)(3-hydroxy-1,4-dioxo-1,4-dihydronaphthalen-2-yl)methyl)phenyl)((4-methoxyphenyl)amino)methyl)-3- hydroxynaphthalene-1,4-dione (Scheme 4, 10a); ^{1}H NMR (400 MHz, DMSO-d_6)



2-((4-((butylamino)(3-hydroxy-1,4-dioxo-1,4-dihydronaphthalen-2-yl)methyl)phenyl)((4-methoxyphenyl)amino)methyl)-3- hydroxynaphthalene-1,4-dione (Scheme 4, 10a); ^{13}C NMR (100 MHz, DMSO-d_6)



2-((4-((hexylamino)(3-hydroxy-1,4- dioxo-1, 4- dihydronaphthalene-2-yl)methyl)phenyl)((4-nitrophenyl)amino)methyl)-3-hydroxynaphthalene-1,4- dione (Scheme 4, 10b); ^{1}H NMR (400 MHz, DMSO-d_6)



2-((4-((hexylamino)(3-hydroxy-1,4- dioxo -1,4-dihydronaphthalene-2-yl)methyl)phenyl)((4-nitrophenyl)amino)methyl)-3-hydroxynaphthalene-1,4- dione (Scheme 4, 10b); ^{13}C NMR (100 MHz, DMSO-d_6)

1. A. Bee, R. Massart and S. Neveu, *J. Magn. Magn. Mater.*, 1995, **149**, 6-9.
2. R. G. Fiorot, J. F. Allochio Filho, T. M. C. Pereira, V. Lacerda Jr, R. B. dos Santos, W. Romão and S. J. Greco, *Tetrahedron Lett*. 2014, **55**, 4373-4377.