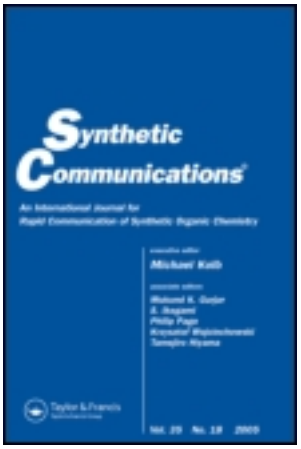

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Simple and Efficient Procedure for the Synthesis of Novel 1,3-Diphenyl-2-azaphenalene Derivatives via One-Pot Multicomponent Reactions

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SIMPLE AND EFFICIENT PROCEDURE FOR THE SYNTHESIS OF NOVEL 1,3-DIPHENYL-2-AZAPHENALENE DERIVATIVES VIA ONE-POT MULTICOMPONENT REACTIONS

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Faculty of Sciences, Department of Chemistry, Arak University, Arak, Iran

A new, one-pot multicomponent reaction of two molecules of aromatic aldehydes with 2,7-naphthalenediol and ammonium hydrogen phosphate is described as an efficient and direct procedure for the preparation of novel 1,3-diphenyl-2-azaphenalene derivatives in a mixture of EtOH-H_2O (3:1) under reflux conditions.

Keywords: Condensation; 1,3-diphenyl-2-azaphenalene; multicomponent; naphthalenediol; one-pot reaction

INTRODUCTION

Multicomponet reactions (MCRs) are of increasing importance in organic and medicinal chemistry.[1–3] MCRs offer significant advantages over conventional linear-type syntheses for their high degree of atom economy, convergence, ease of execution, and broad applications. MCRs are particularly useful to generate diverse chemical libraries of druglike molecules for biological screening.[4,5] There has been tremendous development in three- or four-component reactions, especially the Biginelli,[6–8] Passerini,[9,10] Ugi,[11–14] and Mannich[15,16] reactions, which have further led to a renaissance of MCRs. Nevertheless, great efforts have been and still are being made to find and develop new MCRs.

Straightforward synthesis of 1-(α-aminobenzyl)-2-naphthol (the Betti base) from 2-naphthol, benzaldehyde, and ammonia was reported for the first time by Betti at the beginning of the 20th century.[17] The Betti procedure can be interpreted as an extension of Mannich condensation, with formaldehyde replaced by an aromatic aldehyde, the secondary amine replaced by ammonia, and the C-H acid replaced by an electron-rich aromatic compound such as 2-naphthol.[18] As a consequence of the potential utility of Mannich-type phenolic bases, the amioalkylation of naphthol derivatives is a subject of current chemical interest.[19–21]

Considering the attention given to the condensation of hydroxyaromatic compounds with aldehydes and amines or ammonia, it is rather surprising that

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products from such reactions involving the use of 2,7-naphthalenediol have not been described in the literature. This general type of reaction has provided a convenient route to a variety of complex substances that are accessible only with difficulty by other methods. It has been found that the course of the condensation and the type of products obtained depend upon the several factors, including the nature of the hydroxyaromatic compounds,[22] amine,[23–25] and aldehyde compounds.[26–28]

RESULTS AND DISCUSSION

During the course of our recent studies directed toward the development of practical, safe, and environmentally friendly procedures for some important transformations,[29–33] we found a simple and efficient procedure for the synthesis of novel 1,3-diphenyl-2-azaphenalene derivatives from the condensation of 2,7-naphthalenediol, aromatic aldehydes, and ammonium hydrogen phosphate in a mixture of EtOH-H_2O (3:1) under reflux conditions (Fig. 1).

For the preliminary study, 2,7-naphthalenediol (1 mmol), benzaldehyde (2 mmol), and 25% aqueous ammonia (0.5 mL) in ethanol (3 mL) were stirred under reflux conditions (Betti-type procedure). It was found that 4,9-dihydroxy-1,3-diphenyl-2, 3-dihydro-2-azaphenalene was obtained with yield of 43% after 30 h. To improve the yield of the reaction, we carried out this condensation reaction with different ammonium salts instead of aqueous ammonia, and the best results were obtained when 2 equivalents of ammonium hydrogen phosphate were used (Table 1, entry 3).

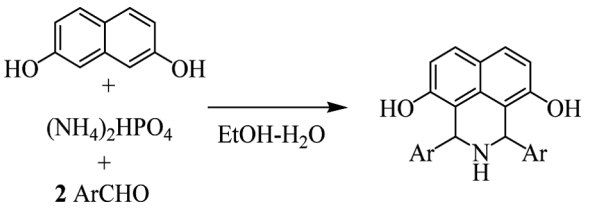


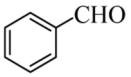
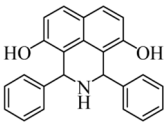
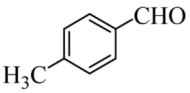
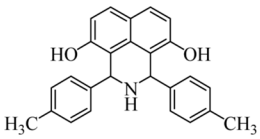
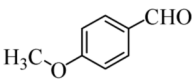
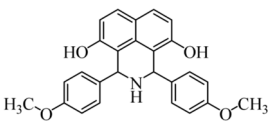
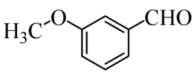
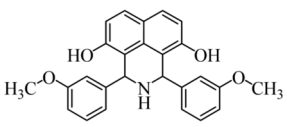
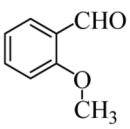
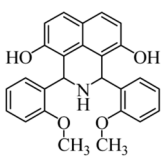
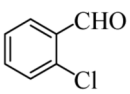
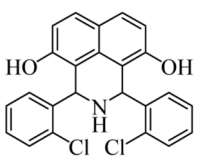
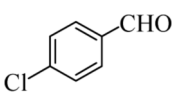
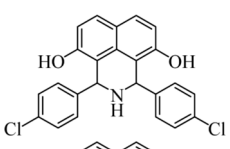
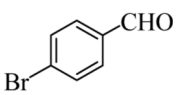
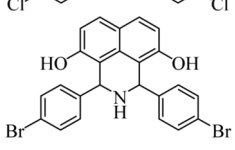
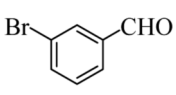
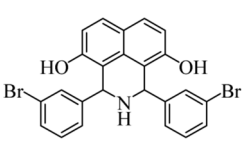
Figure 1. Synthesis of 1,3-diphenyl-2-azaphenalene derivatives.

Table 1. Optimization of reaction condition for the condensation of 2,7-naphthalenediol, benzaldehyde, and ammonia

Entry	Ammonium salts	Time (h)	Yield (%)[a]
1	NH_3	30	43
2	NH_4Cl	30	0
3	$(NH_4)_2HPO_4$	18	70
4	$(NH_4)_2SO_4$	30	0
5	$(NH_4)_2CO_3$	30	58
6	$(NH_4)HSO_4$	30	0
7	NH_4NO_3	30	0
8	$(NH_4)_2HPO_4$	12	85[b]

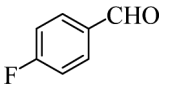
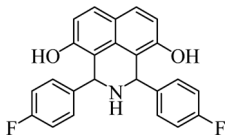
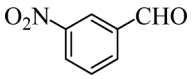
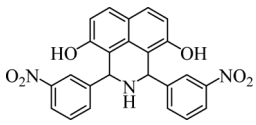
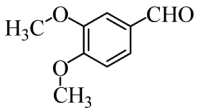
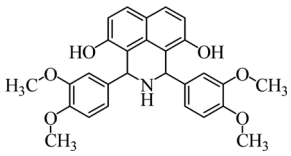
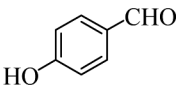
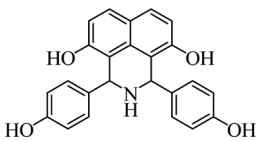
[a]Isolated yields.
[b]This reaction was carried out in EtOH-H_2O (3:1) as a solvent.

Table 2. Synthesis of 4,9-dihydroxy-1,3-diaryl-2,3-dihydro-2-azaphenalenes

Entry	Aldehyde (Ar)	Product[a]	Time (h)	Yield (%)[b]	Mp (°C)
1			12	85	202–203
2			10	83	230–232
3			12	81	223–224
4			9	89	219–220
5			10	78	215–216
6			12	75	203–205
7			8	90	207–209
8			8	92	211–212
9			10	84	208–209

(*Continued*)

Table 2. Continued

Entry	Aldehyde (Ar)	Product[a]	Time (h)	Yield (%)[b]	Mp (°C)
10			12	85	199–200
11			18	82	196–197
12			15	83	215–217
13			10	80	204–206

[a]The product were characterized by IR, ^{1}H NMR, ^{13}C NMR, and elemental analysis.
[b]Isolated yields.

During the course of our investigations of these reaction conditions, we found that addition of water to the reaction mixture increased the yield of product up to 85% (Table 1, entry 8), probably due to the increase of the solubility of $(NH_4)_2HPO_4$. Thus, EtOH-H_2O (3:1) was chosen as a solvent for all further reactions.

To extend the preparative utility and generality of this MCR, the reaction of 2,7-naphthalenediol with various aromatic aldehydes, bearing electron-withdrawing groups (such as nitro, halide), electron-donating groups (such as methyl, methoxy), and ammonium hydrogen phosphate was carried out in ethanol–water (3:1) under reflux conditions and afforded the corresponding 1,3-diphenyl-2-azaphenalene derivatives in good to excellent yields. The results are summarized in Table 2.

On the other hand, aliphatic aldehydes such as propionaldehyde or butyraldehyde and heteroaromatic aldehydes such as 2-pyridinecarbaldehyde or furfural were also examined under the same conditions, but the corresponding products were isolated in only trace amounts.

The plausible mechanism for this MCR can be explained by the Betti reaction.[17] 2,7-Naphthalenediol was reacted with aromatic aldehydes and ammonia in a molar ratio of 1:2:1 to yield naphthoxazine or isomeric Schiff base similar to 2-naphthol in the Betti reaction. In view of the tautomeric capability of these condensation products,[27,34,35] the Schiff base intermediate furnished the

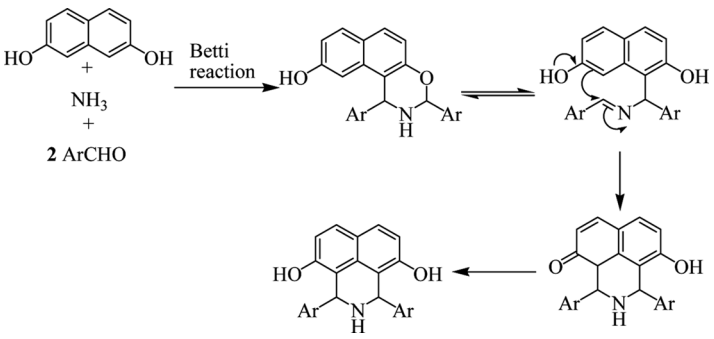


Figure 2. The most probable mechanism for synthesis of 1,3-diphenyl-2-azaphenalene derivatives.

1,3-diphenyl-2-azaphenalene derivatives upon an intermolecular cyclization (Mannich-type reaction) and keto-enol tautomerization. The most probable mechanism for this MCR is illustrated in Fig. 2.

CONCLUSION

In conclusion, we have described a novel, efficient, and one-pot procedure for the preparation of 4,9-dihydroxy-1,3-diaryl-2,3-dihydro-2-azaphenalenes from four-component condensation reactions of aromatic aldehydes, 2,7-naphthalenediol, and ammonium hydrogen phosphate in ethanol–water (3:1) under reflux conditions. In addition to the efficiency and simplicity provided by this procedure, ease of workup, excellent yields of products, and environmental friendliness make the method advantageous. This work is currently in progress, and the results will be reported in due course.

EXPERIMENTAL

All products were characterized by a comparison of their spectral [infrared (IR), ^{1}H NMR, ^{13}C NMR, and elemental analysis] data. All yields refer to isolated products. IR spectra were prepared on a Galaxy series Fourier transform (FT)–IR 5000 spectrophotometer using KBr discs. NMR spectra were recorded on a Brucker spectrophotometer (300 MHz) in dimethylsulfoxide (DMSO-d_6) using tetramethylsilane (TMS) as an internal standard. Elemental analyses were performed on a Vario EL III elemental analyzer.

General Procedure for Synthesis of 4,9-Dihydroxy-1,3-diaryl-2,3-dihydro-2-azaphenalenes

A mixture of 2,7-naphthalenediol (1 mmol), aldehyde (2 mmol), and ammonium hydrogen phosphate (2 mmol) in ethanol–water (3:1, 4 mL) was stirred under reflux conditions for an appropriate time as indicated in Table 2. The progress of the reaction was monitored by thin-layer chromatography (TLC). After completion of the reaction, saturated aqueous NaCl (20 mL) was added to it, the suspension was stirred

for 60 min, and the precipitate was filtered, washed with water, and air dried. The crude products were washed with a mixture of ethyl acetate/n-hexane, (20 mL, 1:4) and dried in vacuum at 100 °C for 4 h to afford the pure products.

Characterization Data

4,9-Dihydroxy-1,3-diphenyl-2,3-dihydro-2-azaphenalene (Table 2, Entry 1). IR (KBr): $\nu_{max} = 3610$, 3377–2850, 1633, 1581, 1516, 1454, 1421, 1323, 1288, 1157, 1041, 977, 827, 754, 702 cm^{-1}. ^1H NMR (300 MHz, DMSO-d_6): $\delta = 9.86$ (s, 2H, disappeared on D_2O exchange), 7.78 (d, $J = 8.1$, 2H), 7.35–7.31 (m, 6H), 7.21 (d, $J = 7.0$, 4H), 6.86 (d, $J = 8.0$, 2H), 5.44 (s, 2H), 3.12 (br, 1H, disappeared on D_2O exchange). ^{13}C NMR (75 MHz, DMSO-d_6): $\delta = 152.5$, 137.1, 130.5, 129.9, 129.4, 128.9, 127.8, 121.9, 115.7, 109.5, 54.2. Anal. calcd. for $C_{24}H_{19}NO_2$: C, 81.56; H, 5.42; N, 3.96. Found: C, 81.42; H, 5.35; N, 3.89.

4,9-Dihydroxy-1,3-di(4-methylphenyl)-2,3-dihydro-2-azaphenalene (Table 2, entry 2). IR (KBr): $\nu_{max} = 3470$, 3322–2860, 1626, 1512, 1488, 1365, 1275, 1089, 1014, 1024, 830 cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6): $\delta = 9.20$ (br, 2H, disappeared on D_2O exchange), 7.63 (d, $J = 8.6$ Hz, 2H), 7.22 (d, $J = 7.3$ Hz, 4H), 6.98 (d, $J = 7.3$ Hz, 4H), 6.85 (d, $J = 8.7$ Hz, 2H), 5.13 (s, 2H), 3.06 (br, 1H, disappeared on D_2O exchange), 2.22 (s, 6H); ^{13}C NMR (75 MHz, DMSO-d_6): $\delta = 151.8$, 138.0, 131.3, 130.6, 130.1, 128.2, 127.9, 122.9, 115.8, 115.5, 54.1, 20.9. Anal. calcd. for $C_{26}H_{23}NO_2$: C, 81.86; H, 6.08; N, 3.67. Found: C, 81.71; H, 6.13; N, 3.52.

4,9-Dihydroxy-1,3-di(4-methoxylphenyl)-2,3-dihydro-2-azaphenalene (Table 2, entry 3). IR (KBr): $\nu_{max} = 3473$, 3332–2833, 1626, 1599, 1516, 1489, 1425, 1369, 1261, 1147, 1039, 835, 781, 700 cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6): $\delta = 9.17$ (br, 2H, disappeared on D_2O exchange), 7.55 (d, $J = 7.3$ Hz, 2H), 7.10 (d, $J = 6.8$, 4H), 6.71–6.63 (m, 6H), 5.12 (s, 2H), 3.68 (s, 6H) 3.10 (br, 1H, disappeared on D_2O exchange); ^{13}C NMR (75 MHz, DMSO-d_6): $\delta = 156.6$, 150.3, 134.3, 131.7, 129.2, 128.5, 122.8, 115.8, 114.9, 111.2, 55.6, 53.2. Anal. calcd. for $C_{26}H_{23}NO_4$: C, 75.53; H, 5.61; N, 3.39. Found: C, 75.39; H, 5.74; N, 3.28.

4,9-Dihydroxy-1,3-di-(3-methoxyphenyl)-2,3-dihydro-2-azaphenalene (Table 2, entry 4). IR (KBr): $\nu_{max} = 3493$, 3300–2833, 1624, 1599, 1516, 1489, 1452, 1369, 1147, 1039, 835, 700 cm^{-1}. ^1H NMR (300 MHz, DMSO-d_6): $\delta = 9.16$ (s, 2H, disappeared on D_2O exchange), 7.57 (d, $J = 8.5$, 2H), 7.12 (t, $J = 7.6$, 2H), 6.85 (d, $J = 8.7$, 2H), 6.74 (d, $J = 7.5$, 2H), 6.66–6.63 (m, 4H), 5.14 (s, 2H), 3.65 (s, 6H), 3.02 (br, 1H, disappeared on D_2O exchange). ^{13}C NMR (75 MHz, DMSO-d_6): $\delta = 159.4$, 150.5, 146.7, 132.1, 129.1, 127.7, 122.7, 120.8, 116.3, 115.2, 114.3, 111.6, 55.3, 53.9. Anal. calcd. for $C_{26}H_{23}NO_4$: C, 75.53; H, 5.61; N, 3.39. Found: C, 75.39; H, 5.54; N, 3.30.

4,9-Dihydroxy-1,3-di-(2-methoyphenyl)-2,3-dihydro-2-azaphenalene (Table 2, entry 5). IR (KBr): $\nu_{max} = 3371$, 3234–2837, 1626, 1512, 1417, 1323, 1265, 1163, 1045, 796, 698 cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6): $\delta = 9.17$ (br, 2H, disappeared on D_2O exchange), 7.61 (d, $J = 8.8$ Hz, 2H), 6.99 (d, $J = 8.1$ Hz, 2H),

6.86–6.68 (m, 4H), 6.60–6.53 (m, 4H), 5.24 (s, 2H), 3.76 (s, 6H), 3.17 (br, 1H, disappeared on D_2O exchange); ^{13}C NMR (75 MHz, DMSO-d_6): $\delta = 157.4$, 150.5, 146.7, 132.1, 129.1, 127.7, 122.7, 120.8, 116.3, 115.2, 114.3, 111.6, 55.2, 53.9. Anal. calcd. for $C_{26}H_{23}NO_4$: C, 75.53; H, 5.61; N, 3.39. Found: C, 75.35; H, 5.72; N, 3.28.

4,9-Dihydroxy-1,3-di-(2-chlorophenyl)-2,3-dihydro-2-azaphenalene (Table 2, entry 6). IR (KBr): $\nu_{max} = 3468$–2830, 1624, 1514, 1428, 1396, 1275, 1132, 1026, 883, 700, 664 cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6): $\delta = 9.28$ (br, 2H, disappeared on D_2O exchange), 7.57 (d, $J = 8.6$ Hz, 2H), 7.10–7.01 (m, 8H), 6.86 (d, $J = 8.7$ Hz, 2H), 5.12 (s, 2H), 3.14 (br, 1H, disappeared on D_2O exchange); ^{13}C NMR (75 MHz, DMSO-d_6): $\delta = 150.8$, 146.3, 134.1, 132.8, 129.6, 129.4, 128.5, 127.7, 127.3, 122.7, 115.6, 114.5, 53.3. Anal. calcd. for $C_{24}H_{17}Cl_2NO_2$: C, 68.26; H, 4.06; N, 3.32. Found: C, 68.12; H, 4.21; N, 3.38.

4,9-Dihydroxy-1,3-di-(4-chlorophenyl)-2,3-dihydro-2-azaphenalene (Table 2, entry 7). IR (KBr): $\nu_{max} = 3535$, 3387–2862, 1626, 1596, 1512, 1489, 1367, 1275, 1089, 1014, 829 cm^{-1}. ^1H NMR (300 MHz, DMSO-d_6): $\delta = 9.26$ (s, 2H, disappeared on D_2O exchange), 7.58 (d, $J = 8.7$, 2H), 7.27 (d, $J = 8.2$, 4H), 7.09 (d, $J = 8.2$, 4H), 6.86 (d, $J = 8.7$, 2H), 5.13 (s, 2H), 3.07 (br, 1H, disappeared on D_2O exchange). ^{13}C NMR (75 MHz, DMSO-d_6): $\delta = 150.6$, 144.1, 132.0, 131.2, 130.2, 128.1, 127.9, 122.8, 115.9, 115.3, 53.3. Anal. calcd. for $C_{24}H_{17}Cl_2NO_2$: C, 68.26; H, 4.06; N, 3.32. Found: C, 68.10; H, 4.10; N, 3.25.

4,9-Dihydroxy-1,3-di-(4-bromophenyl)-2,3-dihydro-2-azaphenalene (Table 2, entry 8). IR (KBr): $\nu_{max} = 3487$, 3279–2862, 1626, 1599, 1514, 1489, 1367, 1300, 1089, 1014, 829, 781, 756, 687 cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6): $\delta = 9.29$ (br, 2H, disappeared on D_2O exchange), 7.63 (d, $J = 8.3$ Hz, 2H), 7.35 (d, $J = 7.9$ Hz, 4H), 7.16 (d, $J = 7.8$ Hz, 4H), 6.92 (d, $J = 8.4$ Hz, 2H), 5.16 (s, 2H), 3.11 (br, 1H, disappeared on D_2O exchange); ^{13}C NMR (75 MHz, DMSO-d_6): $\delta = 150.9$, 146.0, 131.6, 131.4, 130.1, 128.6, 122.5, 121.6, 115.3, 114.8, 53.4. Anal. calcd. for $C_{24}H_{17}Br_2NO_2$: C, 56.39; H, 3.35; N, 2.74. Found: C, 56.30; H, 3.29; N, 2.80.

4,9-Dihydroxy-1,3-di-(3-bromophenyl)-2,3-dihydro-2-azaphenalene (Table 2, entry 9). IR (KBr): $\nu_{max} = 3484$, 3370–2896, 1624, 1594, 1512, 1487, 1311, 1275, 1090, 1132, 881 cm^{-1}. ^1H NMR (300 MHz, DMSO-d_6): $\delta = 9.30$ (s, 2H, disappeared on D_2O exchange), 7.57 (d, $J = 8.7$, 2H), 7.35 (d, $J = 7.8$, 2H), 7.21–7.15 (m, 4H), 7.08 (d, $J = 7.6$, 2H), 6.85 (d, $J = 8.7$, 2H), 5.12 (s, 2H), 3.09 (br, 1H, disappeared on D_2O exchange). ^{13}C NMR (75 MHz, DMSO-d_6): $\delta = 150.7$, 147.7, 131.8, 131.0, 130.4, 129.6, 128.2, 127.5, 122.7, 121.7, 115.5, 115.4, 53.6. Anal. calcd. for $C_{24}H_{17}Br_2NO_2$: C, 56.39; H, 3.35; N, 2.74. Found: C, 56.26; H, 3.37; N, 2.81.

4,9-Dihydroxy-1,3-di-(4-flurophenyl)-2,3-dihydro-2-azaphenalene (Table 2, entry 10). IR (KBr): $\nu_{max} = 3491$, 3338–2820, 1625, 1516, 1368, 1276, 1119, 1024, 825, 790, 656 cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6): $\delta = 9.31$ (br, 2H, disappeared on D_2O exchange), 7.60 (d, $J = 8.4$ Hz, 2H), 7.11 (d, $J = 7.6$ Hz, 4H), 7.06 (d, $J = 7.5$ Hz, 4H), 6.87 (d, $J = 8.3$ Hz, 2H), 5.21 (s, 2H), 3.11 (br, 1H, disappeared on D_2O exchange); ^{13}C NMR (75 MHz, DMSO-d_6): $\delta = 163.0$, 150.9,

140.0, 131.7, 130.3, 130.2, 128.2, 122.6, 115.4, 115.1, 53.2. Anal. calcd. for $C_{24}H_{17}F_2NO_2$: C, 74.03; H, 4.40; N, 3.60. Found: C, 73.87; H, 4.53; N, 3.47.

4,9-Dihydroxy-1,3-di-(3-nitrophenyl)-2,3-dihydro-2-azaphenalene (Table 2, entry 11). IR (KBr): $\nu_{max} = 3471$, 3276–2864, 1626, 1526, 1512, 1424, 1346, 1321, 1275, 1156, 1024, 832, 699 cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6): $\delta = 9.46$ (br, 2H, disappeared on D_2O exchange), 8.06 (d, $J = 7.5$ Hz, 2H), 7.93 (s, 2H), 7.64–7.50 (m, 6H), 6.90 (d, $J = 8.8$ Hz, 2H), 5.30 (s, 2H), 3.17 (br, 1H, disappeared on D_2O exchange); ^{13}C NMR (75 MHz, DMSO-d_6): $\delta = 151.8$, 148.0, 146.3 133.1, 131.6, 129.7, 128.4, 120.8, 123.0, 121.8, 115.2, 114.8, 53.4. Anal. calcd. for $C_{24}H_{17}N_3O_6$: C, 65.01; H, 3.86; N, 9.48. Found: C, 64.82; H, 3.93; N, 9.34.

4,9-Dihydroxy-1,3-di(3,4-dimethoyphenyl)-2,3-dihydro-2-azaphenalene (Table 2, entry 12). IR (KBr): $\nu_{max} = 3493$, 3332–2833, 1626, 1599, 1516, 1489, 1425, 1369, 1261, 1147, 1039, 835, 781, 700 cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6): $\delta = 9.11$ (br, 2H, disappeared on D_2O exchange), 7.66 (d, $J = 8.7$ Hz, 2H), 6.88 (d, $J = 8.5$ Hz, 2H), 6.75–6.64 (m, 4H), 6.56 (d, $J = 7.6$ Hz, 2H), 5.12 (s, 2H), 3.74 (s, 6H), 3.67 (s, 6H), 3.13 (br, 1H, disappeared on D_2O exchange); ^{13}C NMR (75 MHz, DMSO-d_6): $\delta = 157.8$, 153.6, 150.5, 146.7, 132.1, 129.1, 127.7, 122.7, 120.8, 115.2, 114.3, 111.6, 55.2, 55.0, 53.9. Anal. calcd. for $C_{28}H_{27}NO_6$: C, 71.02; H, 5.75; N, 2.96. Found: C, 69.84; H, 5.83; N, 2.83.

4,9-Dihydroxy-1,3-di-(4-hydroxyphenyl)-2,3-dihydro-2-azaphenalene (Table 2, entry 13). IR (KBr): $\nu_{max} = 3519$, 3211–2823, 2696, 1632, 1604, 1543, 1516, 1429, 1309, 1248, 1174, 1130, 831, 773, 657 cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6): $\delta = 9.01$ (br, 4H, disappeared on D_2O exchange), 7.53 (d, $J = 7.4$ Hz, 2H), 6.95–6.84 (m, 6H), 6.60–6.50 (m, 4H), 5.08 (s, 2H), 3.03 (br, 1H, disappeared on D_2O exchange); ^{13}C NMR (75 MHz, DMSO-d_6): $\delta = 156.4$, 150.7, 134.2, 132.0, 129.4, 127.7, 122.6, 116.0, 115.3, 115.0, 53.3. Anal. calcd. for $C_{24}H_{19}NO_4$: C, 74.49; H, 4.97; N, 3.63. Found: C, 74.36; H, 4.86; N, 3.52.

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