Synthesis of 5,5'-(arylmethylene)bis(pyrimidinone) derivatives in aqueous media

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The condensation and addition reactions of aromatic aldehydes and 4,6-dihydroxypyrimidine or 2,4-dihydroxy-6-aminopyrimidine in water in the presence of triethylbenzylammonium chloride (TEBAC) afford a one-pot synthesis of 5,5'-(arylmethylene)bis[6-hydroxypyrimidine-4(3H)-one]s and 5,5'-(arylmethylene)bis[6-aminopyrimidine-2,4(1H,3H)-dione]s. These compounds were characterised by elemental analysis and IR and ^{1}H NMR spectra and further confirmed by a single crystal X-ray diffraction analysis.

Keywords: 5,5'-(arylmethylene)bis(pyrimidinone), aromatic aldehyde, 4,6-dihydroxypyrimidine, 2,4-dihydroxy-6-aminopyrimidine, aqueous media

The importance of uracil and its annelated derivatives is well recognised by synthetic[1-4] as well as biological[5-10] chemists. With the development of clinically useful anticancer and antiviral drugs,[11-14] there has recently been remarkable interest in the synthetic manipulations of uracils.[15-17] Jiang and Roberts[18] reported the synthesis of 5,5'-{(perfluorophenyl) methylene}bis(6-amino-1,3-dimethylpyrimidine-2,4-dione) in glacial acetic acid under nitrogen. But the yield was only 44%. Azizian *et al.*[19] reported the synthesis of bis(6-amino-pyrimidonyl)methanes using thermal (in ethanol at 80 °C) or microwave-assisted solvent-free methods. Moskvin *et al.*[20] reported the synthesis of 5,5'-methylenebis(4,6-dihydroxy-2-methylthiopyrimidines) by condensation of 4,6-dihydroxy-2-methylthiopyrimidine with formaldehyde and aromatic or heterocyclic aldehydes in ethanol. However, they were reacted in organic solvents or had low yields.[18]

The need to reduce the amount of toxic waste and byproducts arising from chemical processes requires increasing emphasis on the use of less toxic and environmentally compatible materials in the design of new synthetic methods.[21-23] In one of the most promising approaches, water is used as the reaction medium.[24-26] Breslow and Rideout,[27,28] showed that hydrophobic effects could strongly enhance the rate of several organic reactions and rediscovered the use of water as a solvent in organic chemistry in the 1980s. In recent years, there has been increasing recognition that water is an attractive medium for many organic reactions.[29-34] The aqueous medium is less expensive, less dangerous and more environment-friendly than an organic solvent. Generally, the low solubility[35] of most reagents in water is not an obstacle to reactivity, which on the contrary, is reduced with the use of cosolvents. Based on our previous studies on the use of water as the solvent for carrying out carbon-carbon bond-forming reactions under heterogeneous catalysis,[36-39] we report here a novel synthesis of 5,5'-(arylmethylene)bis(pyrimidinone) derivatives using water as the reaction medium.

When aromatic aldehydes **1** and 4,6-dihydroxypyrimidine **2** were stirred at 90 °C for 8–14 h in water in the presence of triethylbenzylammonium chloride (TEBAC), the desired products 5,5'-(arylmethylene)bis[6-hydroxypyrimidine-4(3H)-one]s **3** were obtained in moderate to good yields (Scheme 1). The results are summarised in Table 1.

Similarly, the reaction of aromatic aldehyde **1** and 2,4-dihydroxy-6-aminopyrimidine **4** under the same reaction conditions afforded 5,5'-(arylmethylene)bis[6-aminopyrimidine-2,4(1H,3H)-dione]s **5** (Scheme 2) and the results are summarised in Table 2.

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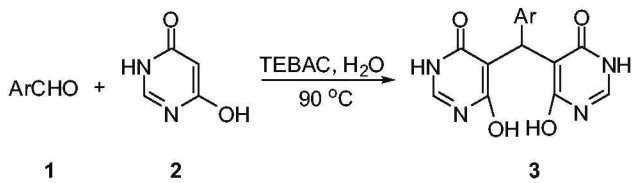
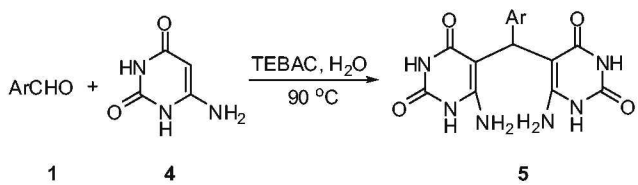


1 **2** **3**

Scheme 1

Table 1 The syntheses of **3** in aqueous media

Entry	Ar	t/h	Isolated yield/%
3a	3-$NO_2C_6H_4$	10	79
3b	4-BrC_6H_4	9	62
3c	2-$NO_2C_6H_4$	12	73
3d	4-HOC_6H_4	12	86
3e	3,4-$Cl_2C_6H_3$	14	75
3f	4-$NO_2C_6H_4$	8	84



1 **4** **5**

Scheme 2

Table 2 The syntheses of **5** in aqueous media

Entry	Ar	t/h	Isolated yield/%
5a	3-$NO_2C_6H_4$	8	96
5b	4-$NO_2C_6H_4$	6	94
5c	2-ClC_6H_4	12	81
5d	4-ClC_6H_4	10	86
5e	3-ClC_6H_4	8	91

The structures of the compounds **3** and **5** were established on the basis of spectroscopic data, particularly ^{1}H NMR analysis. The structure of **3c** was further confirmed by X-ray diffraction analysis. Crystallographic data are presented in Table 3. An ORTEP plot and the atom numbering are given in Fig. 1. The structure exhibits intermolecular hydrogen bonds: N2–H…O2 ($-x + 2$, $-y$, $-z$), N3–H…N1 ($x - 1/2$, $-y + 1/2$, $z - 1/2$) and O1–H…O3 ($x + 1/2$, $-y + 1/2$, $z + 1/2$) and an intramolecular hydrogen bond: O4–H…O1, which helps in stabilising the crystal structure.

Table 3 Crystal data and structure refinement of compound **3c**

Empirical formula	$C_{39}H_{43}N_{13}O_{15}$
Formula weight	933.86
Temperature	298(2) K
Wavelength	0.71073 Å
Crystal system	Monoclinic
Space group	$P2_1/n$
Unit cell dimensions	a = 11.6719(15) Å α = 90°
	b = 15.0583(18) Å β = 93.631(2)°
	c = 12.6443(16) Å γ = 90°
Volume	2217.9(5) Å^3
Z	2
Density (calculated)	1.398 mg/m^3
Absorption coefficient	0.110 mm^{-1}
F(000)	976
Crystal size	0.31 × 0.25 × 0.13 mm
Theta range for data collection	2.45 to 25.03°
Index ranges	$-13 \le h \le 13$, $-17 \le k \le 17$, $-8 \le l \le 15$
Reflections collected	11498
Independent reflections	3912 [R(int) = 0.0343]
Absorption correction	Semi-empirical from equivalents
Refinement method	Full-matrix least-squares on F^2
Data/restraints/parameters	3912/0/344
Goodness-of-fit on F^2	1.058
Final R indices [I>2 sigma(I)]	R^1 = 0.0571, wR^2 = 0.1538
R indices (all data)	R^1 = 0.0994, wR^2 = 0.1920
Largest diff. peak and hole	0.439 and –0.482 eÅ^{-3}

A reasonable mechanism for the formation of the products **3** is outlined in Scheme 3. The reaction occurs by an initial formation of a Knoevenagel condensation product, from the aromatic aldehyde **1** and 4,6-dihydroxypyrimidine **2**. Michael addition than occurs between the Knoevenagel condensation product and another 4,6-dihydroxypyrimidine **2** followed by isomerisation to give product **3**.

In summary, the conversion of aromatic aldehydes and 4,6-dihydroxypyrimidine or 2,4-dihydroxy-6-aminopyrimidine into5,5'-(arylmethylene)bis[6-hydroxypyrimidine-4(3H)-one]s and 5,5'-(arylmethylene)bis[6-aminopyrimidine-2,4(1H,3H)-dione]s can be efficiently performed in water as a solvent using a catalytic amount of TEBAC. Compared to the previous methods, this method has the advantages of good yields, low cost, simple operation and an environmentally benign procedure.

Experimental

Melting points are uncorrected. IR spectra were recorded on a Tensor 27 spectrometer in KBr with absorptions in cm^{-1}. ^{1}H NMR spectra were determined on a Bruker-400 MHz spectrometer using DMSO-d_6 solutions. Chemical shifts are expressed in ppm downfield from internal tetramethylsilane. Microanalyses were carried out on a Perkin-Elmer 2400 II elemental analyser. X-ray diffraction was recorded on a Smart-1000 diffractometer.

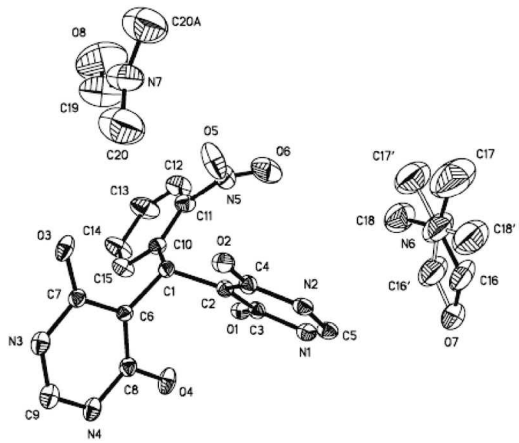


Fig. 1 The X-ray crystal structure of compound **3c**.

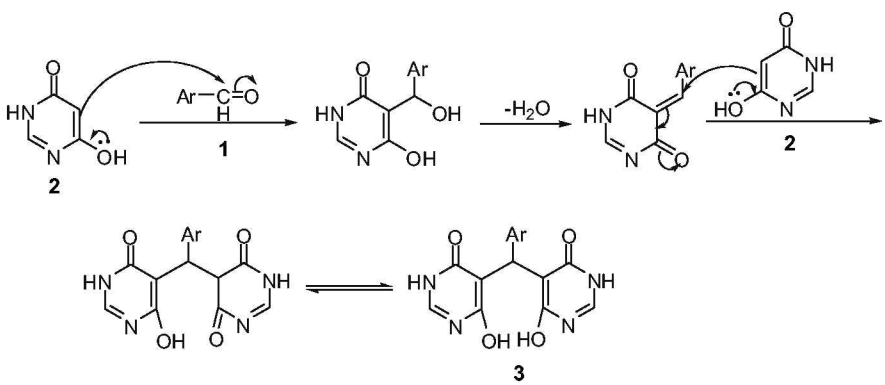
General procedure for the synthesis of 5,5'-(arylmethylene)bis(pyrimidinone) (**3** and **5**): A mixture of aromatic aldehyde (**1**) (2 mmol), 4,6-dihydroxypyrimidine (**2**) or 2,4-dihydroxy-6-aminopyrimidine (**4**) (4 mmol) and TEBAC (0.15 g) in H_2O (10 ml) was stirred for 6–14 h at 90 °C, then cooled to room temperature. The crystalline powder formed was collected by filtration, washed with water and recrystallised from DMF and dried at 120 °C *in vacuo* to give pure **3** or **5**.

5,5'-(3-nitrophenylmethylene)bis[6-hydroxypyrimidine-4(3H)-one] (**3a**): M.p. 266–268 °C. IR: ν/cm^{-1} 3500–2400, 1684, 1610, 1560, 1441, 1382, 1350, 1288, 1220, 1140, 1095, 1059, 927, 898, 867, 823, 789, 756, 721, 694. ^{1}H NMR (DMSO-d_6): δ 6.25 (1H, s, CH), 7.48–7.56 (2H, m, ArH), 7.78 (1H, s, ArH), 8.03 (1H, d, J = 7.6 Hz, ArH), 8.21 (2H, s, 2 × C^2-H), 12.65 (4H, br., s, 2 × OH, 2 × NH). Found: C, 50.56; H, 3.02; N, 19.45. Calcd for $C_{15}H_{11}N_5O_6$: C, 50.43; H, 3.10; N, 19.60%.

5,5'-(4-bromophenylmethylene)bis[6-hydroxypyrimidine-4(3H)-one] (**3b**): M.p. 258–260 °C. IR: ν/cm^{-1} 3200–2500, 1653, 1558, 1487, 1448, 1397, 1294, 1243, 1134, 1096, 1073, 1009, 916, 845, 793, 698. ^{1}H NMR (DMSO-d_6): δ 6.13 (1H, s, CH), 6.94 (2H, d, J = 8.4 Hz, ArH), 7.39 (2H, d, J = 8.4 Hz, ArH), 8.14 (2H, s, 2 × C^2-H), 12.59 (4H, br., s, 2 × OH, 2 × NH). Found: C, 46.28; H, 2.91; N, 14.17. Calcd for $C_{15}H_{11}BrN_4O_4$: C, 46.06; H, 2.83; N, 14.32%.

5,5'-(2-nitrophenylmethylene)bis[6-hydroxypyrimidine-4(3H)-one] (**3c**): M.p. > 300 °C. IR: ν/cm^{-1} 3200–2500, 1652, 1620, 1566, 1522, 1470, 1431, 1386, 1361, 1308, 1250, 1234, 1098, 1061, 903, 855, 835, 804, 780, 724. ^{1}H NMR (DMSO-d_6): δ 6.36 (1H, s, CH), 7.22 (1H, d, J = 7.6 Hz, ArH), 7.39 (1H, d, J = 7.6 Hz, ArH), 7.52 (1H, d, J = 7.6 Hz, ArH), 7.65 (1H, d, J = 7.6 Hz, ArH), 8.10 (2H, s, 2 × C^2-H), 12.23 (4H, br., s, 2 × OH, 2 × NH). Found: C, 50.32; H, 3.04; N, 19.34. Calcd for $C_{15}H_{11}N_5O_6$: C, 50.43; H, 3.10; N, 19.60%.

5,5'-(4-hydroxyphenylmethylene)bis[6-hydroxypyrimidine-4(3H)-one] (**3d**): M.p. > 300 °C. IR: ν/cm^{-1} 3500-2500, 1674, 1620, 1557, 1512, 1445, 1391, 1291, 1243, 1173, 1095, 1060, 912, 837, 793, 775. ^{1}H NMR (DMSO-d_6): δ 6.07 (1H, s, CH), 6.60 (2H, d, J = 8.0 Hz, ArH), 6.76 (2H, d, J = 8.0 Hz, ArH), 8.10 (2H, s, 2 × C^2-H), 9.07 (1H, s, OH), 12.52 (4H, br., s, 2 × OH, 2 × NH). Found: C, 55.02; H, 3.61; N, 17.29. Calcd for $C_{15}H_{12}N_4O_5$: C, 54.88; H, 3.68; N, 17.07%.



Scheme 3

5,5'-(3,4-dichlorophenylmethylene)bis[6-hydroxypyrimidine-4(3H)-one] (**3e**): M.p. 275–276 °C. IR: ν/cm^{-1} 3500–2500, 1668, 1560, 1470, 1429, 1388, 1294, 1232, 1134, 1096, 1059, 1029, 921, 808, 791, 747, 725. ^{1}H NMR (DMSO-d_6): δ 6.14 (1H, s, CH), 6.99 (1H, d, J = 8.4 Hz, ArH), 7.14 (1H, s, ArH), 7.47 (1H, d, J = 8.4 Hz, ArH), 8.17 (2H, s, 2 × C^2-H), 12.63 (4H, br., s, 2 × OH, 2 × NH). Found: C, 47.18; H 2.75; N, 14.83. Calcd for $C_{15}H_{10}Cl_2N_4O_4$: C, 47.27; H, 2.64; N, 14.70%.

5,5'-(4-nitrophenylmethylene)bis[6-hydroxypyrimidine-4(3H)-one] (**3f**): M.p. 257–259 °C. IR: ν/cm^{-1} 3200-2500, 1672, 1632, 1558, 1529, 1437, 1388, 1345, 1291, 1138, 1095, 1058, 911, 854, 830, 794, 764, 724. ^{1}H NMR (DMSO-d_6): δ 6.25 (1H, s, CH), 7.27 (2H, d, J = 8.8 Hz, ArH), 8.10 (2H, d, J = 8.8 Hz, ArH), 8.20 (2H, s, 2 × C^2–H), 12.66 (4H, br., s, 2 × OH, 2 × NH). Found: C 50.35; H 2.95; N 19.43. Calcd for $C_{15}H_{11}N_5O_6$: C 50.43; H 3.10; N 19.60%.

5,5'-(3-nitrophenylmethylene)bis[6-aminopyrimidine-2,4(1H,3H)-dione] (**5a**): M.p. > 300 °C. IR: ν/cm^{-1} 3421, 3311, 3242, 3189, 3074, 2988, 1699, 1623, 1592, 1539, 1458, 1393, 1338, 1290, 1234, 1192, 1093, 1014, 930, 852, 828, 794, 762, 733, 708. ^{1}H NMR (DMSO-d_6): δ 6.01 (4H, br., s, 2 × NH$_2$), 6.25 (1H, s, CH), 7.59 (1H, t, J = 8.0 Hz, ArH), 7.79 (1H, d, J = 7.6 Hz, ArH), 7.08 (1H, d, J = 8.0 Hz, ArH), 8.23 (1H, s, ArH), 10.07 (2H, br., s, 2 × NH), 10.44 (2H, br., s, 2 × NH). Found: C, 46.75; H, 3.52; N, 25.17. Calcd for $C_{15}H_{13}N_7O_6$: C, 46.52; H, 3.38; N, 25.32%.

5,5'-(4-nitrophenylmethylene)bis[6-aminopyrimidine-2,4(1H,3H)-dione] (**5b**): M.p. > 300 °C. IR: ν/cm^{-1} 3444, 3312, 3167, 1708, 1641, 1624, 1600, 1532, 1518, 1457, 1403, 1351, 1233, 1175, 1108, 1050, 871, 829, 763, 737, 706. ^{1}H NMR (DMSO-d_6): δ 5.98 (4H, br., s, 2 × NH$_2$), 6.20 (1H, s, CH), 7.64 (2H, d, J = 8.0 Hz, ArH), 8.17 (2H, d, J = 8.0 Hz, ArH), 10.07 (2H, br., s, 2 × NH), 10.44 (2H, br., s, 2 × NH). Found: C, 46.43; H, 3.46; N, 25.16. Calcd for $C_{15}H_{13}N_7O_6$: C, 46.52; H, 3.38; N, 25.32%.

5,5'-(2-chlorophenylmethylene)bis[6-aminopyrimidine-2,4 (1H,3H)-dione] (**5c**): M.p. > 300 °C. IR: ν/cm^{-1} 3365, 3170, 1714, 1654, 1634, 1595, 1455, 1391, 1168, 1092, 1024, 886, 775. ^{1}H NMR (DMSO-d_6): δ 5.31 (1H, s, CH), 6.70 (4H, br., s, 2 × NH$_2$), 7.03–7.07 (2H, m, ArH), 7.16 (1H, d, J = 8.0 Hz, ArH), 7.25 (1H, t, J = 8.0 Hz, ArH), 10.33 (2H, br., s, 2 × NH), 10.55 (2H, br., s, 2 × NH). Found: C, 47.97; H, 3.53; N, 22.48. Calcd for $C_{15}H_{13}ClN_6O_4$: C, 47.82; H, 3.48; N 22.31%.

5,5'-(4-chlorophenylmethylene)bis[6-aminopyrimidine-2,4 (1H,3H)-dione] (**5d**): M.p. > 300 °C. IR: ν/cm^{-1} 3336, 3164, 1715, 1635, 1551, 1529, 1522, 1489, 1457, 1393, 1296, 1240, 1179, 1093, 1022, 911, 841, 774. ^{1}H NMR (DMSO-d_6): δ 5.28 (1H, s, CH), 6.17 (4H, br., s, 2 × NH$_2$), 7.09 (2H, d, J = 8.4 Hz, ArH), 7.25 (2H, d, J = 8.4 Hz, ArH), 10.07 (2H, br., s, 2 × NH), 10.52 (2H, br., s, 2 × NH). Found: C 48.02; H 3.35; N 22.19. Calcd for $C_{15}H_{13}ClN_6O_4$: C, 47.82; H, 3.48; N, 22.31%.

5,5'-(3-chlorophenylmethylene)bis[6-aminopyrimidine-2,4 (1H,3H)-dione] (**5e**): M.p. > 300 °C. IR: ν/cm^{-1} 3279, 3156, 1709, 1658, 1623, 1523, 1456, 1394, 1217, 1180, 1154, 1096, 807, 784. ^{1}H NMR (DMSO-d_6): δ 5.31 (1H, s, CH), 6.70 (4H, br., s, 2 × NH$_2$), 7.06 (2H, s, ArH), 7.17 (1H, d, J = 8.0 Hz, ArH), 7.25 (1H, t, J = 8.0 Hz, ArH), 10.33 (2H, br., s, 2 × NH), 10.53 (2H, br., s, 2 × NH). Found: C, 47.73; H, 3.34; N, 22.37. Calcd for $C_{15}H_{13}ClN_6O_4$: C, 47.82; H, 3.48; N, 22.31%.

X-ray crystal analysis of **3c**

X-ray diffraction data were collected on a BRUKER SMART 1000 CCD detector with graphite-monochromatised Mo-Kα radiation (λ = 0.71073 Å) for compound **3c**. The structures have been solved by direct methods using the program SHELXL 97[40] and Fourier difference techniques. Refinement has been by full-matrix least-squares method on F^2 using SHELXL 97.[41] CCDC 705256 contains the supplementary crystallographic data for this paper. The data can be obtained free of charge from the Cambridge Crystallographic Data Centre via www.ccdc.cam.ac.uk/data_request.cif.

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