Synthesis of substituted and unsubstituted 5-(1,3-diaryl-1-oxopropyl)pyrimidine (1H, 3H, 5H)-2,4,6-triones

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Reactions of 1,3-diaryl-2-propene-1-ones 2a-i give the corresponding 5-(1,3-diaryl-1-oxopropyl)pyrimidine-(1H,3H,5H)-2,4,6-triones 3a-i with barbituric acid 1 under refluxing condition without using any catalyst.

Keywords: Diarylpyrimidinetriones, diarylpropeneones, barbituric acid

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A number of barbituric acid derivatives 5-(1,3-diaryl-1-oxopropyl)pyrimidine(1H,3H,5H)-2,4,6-triones 3a-i were synthesized and characterized. These compounds seem to be new in literature.

The formation of compounds 3a-i may be explained by applying a general addition reaction of Michael type (Scheme I). These were formed during the synthesis of 1:2 adducts (2 being arylideneacetophenone) which may undergo subsequent intramolecular Aldol condensation leading to the formation of spiro structures 5. In continuation of the work 5-7 on barbituric acid derivatives, herein is reported the synthesis of compounds 3a-i in the present paper.

Compound 1 reacted with 1,3-diphenyl-2-propene-1-one 2a to give a yellowish crystalline solid at refluxing temperature in 50% aqueous ethanol. Its mass spectrum gave a peak at m/z 336 (M+). The 1H NMR spectrum of compound 3a showed besides other usual signals, two singlets at δ 11.08 (NH, 1H, H-1) and 11.03 (NH, 1H, H-3), one multiplet at δ 4.17 (1H, H-7), one doublet at δ 3.81 (J=4.0 Hz, 1H, H-5), two doublet of doublet at δ 4.04 (J1=8.5 Hz, J2=18.0 Hz, 1H, H-8) and δ 3.62 (J1=6.5 Hz, J2=18.5 Hz, 1H, H-8) and 13C NMR showed signals at δ 197.0 (C-9), 169.94 (C-4), 169.57 (C-6), 150.23 (C-2), 51.65 (C-5), 41.54 (C-7), 40.62 (C-8), on the basis of which it was assigned the structure 5-(1,3-diphenyl-1-oxopropyl)pyrimidine(1H,3H,5H)-2,4,6-triones 3a. Similarly 1 on condensation with other arylideneacetophenones 2b-i afforded the adducts 3b-i (Scheme I). The structures of all these compounds were assigned on the basis of their spectral data and elemental analysis.

Experimental Section

Melting points are uncorrected. IR spectra were recorded as KBr pellets using Shimadzu IR-470 infrared spectrometer in the range of 4000-400 cm⁻¹. 1H NMR spectra were recorded in DMSO-d₆ on a JEOL 500 MHz NMR spectrometer using TMS as internal standard (chemical shifts in δ, ppm). 13C NMR spectra were recorded in DMSO-d₆ at 125 MHz and MS on a JEOL JMS-HX 110A spectrometer. All the compounds gave satisfactory C, H and N analyses.

1,3-Diphenyl-2-propene-1-one 2a, 1-(4-chlorophenyl)-3-phenyl-2-propen-1-one 2b, 1-(4-methoxyphenyl)-3-phenyl-2-propen-1-one 2c, 1-(4-nitrophenyl)-3-phenyl-2-propen-1-one 2d, 1-(4-methylphenyl)-3-phenyl-2-propen-1-one 2e, 1-(4-methylphenyl)-3-phenyl-2-propen-1-one 2f, 1-(4-chlorophenyl)-3-(4-methoxyphenyl)-2-propen-1-one 2g, 1-(4-methylphenyl)-3-(4-methoxyphenyl)-2-propen-1-one 2h and
1,3-(4-dimethoxyphenyl)-2-propene-1-one were prepared by following primarily literature methods and by changing the reaction conditions whenever necessary. The reactions described in the present paper were carried out following a general procedure.

**General Procedure.** A mixture of barbituric acid (0.005 mole) and arylideneacetophenone (0.005 mole) was dissolved in 50% aqueous ethanol in a round bottomed flask equipped with a magnetic stirrer and a reflux condenser. The reaction mixture was refluxed for 18-20 h and was cooled and filtered. A solid mass obtained was purified by recrystallisation from rectified spirit. The progress of reaction and the formation of the products were monitored by TLC (eluting solvents, ethyl acetate: methanol, 10:1).

**5-(1,3-Diphenyl-1-oxopropyl)pyrimidine (1H, 3H, 5H)-2,4,6-trione, 3a:** Yellowish white solid. Yield 39%; m.p. 185-87°C; Rf 0.18; UV-Vis: 272 (π→π* of C=O), 242, 209 (π→π* of C=C); IR (KBr): 3220, 3100 (OH, NH), 1760, 1700, 1680 (C=O), 1590, 1575, 1530 cm⁻¹ (C=C, C=N); 1H NMR (DMSO-d6): δ 11.08 (s, 1H, NH, H-1), 11.03 (s, 1H, NH, H-3); 7.98-7.16 (m, 10H, aromatic), 4.17 (m, 1H, H-7), 3.81 (d, J=4.0, 1H, H-5); 3.62 (dd, J_vic=6.5, J_gem=18.5, 1H, H-8); 13C NMR (DMSO-d6): δ 197.0 (C-9), 169.94 (C-4), 169.59 (C-6), 150.23 (C-2), [139.38, 136.46, 133.10, 128.56, 128.20, 127.67, 127.12 (aromatic)], 51.65 (C-5), 41.54 (C-7), 40.62 (C-8); MS: m/z 336 (M⁺), 231, 216, 208, 132, 128, 105 (100%), 103, 102. Anal. Found: C, 67.95; H, 4.79; N, 8.29. Calcd. for C₁₉H₁₆N₂O₄: C, 67.88; H, 4.76; N, 8.32%.

**5-{1-(4-Chlorophenyl)-3-phenyl-1-oxopropyl}-pyrimidine (1H, 3H, 5H)-2,4,6-trione, 3b:** White solid. Yield 27%; m.p. 200-02°C; Rf 0.26; UV-Vis: 252 (π→π* of C=O), 210 (π→π* of C=C); IR (KBr): 3275, 3200 (OH, NH), 1730, 1690, 1670 (C=O), 1635, 1585, 1570, 1518 cm⁻¹ (C=C, C=N); 1H NMR (DMSO-d6): δ 11.07 (s, 1H, NH, H-1), 11.02 (s, 1H, NH, H-3); 8.0 (d, J=9.0, 2H, H-2′, 6′), 7.59 (d, J=9.0, 2H, H-2′, 6′), 7.25 (m, 3H, H-3′, 4′, 5′), 7.16 (d, J=7.0, 2H, H-3′, 5′), 4.15 (m, 1H, H-7), 4.03 (dd, J_vic=8.5, J_gem=18.5, 1H, H-8), 3.80 (d, J=3.5, 1H, H-5), 3.60 (dd, J_vic=6.5, J_gem=18.5, 1H, H-8); 13C NMR (DMSO-d6): δ 197.50 (C-9), 169.89 (C-6), 169.49 (C-4), 150.21 (C-2), [139.26, 138.02, 135.10, 129.80, 129.55, 128.81, 127.81, 127.15 (aromatic)], 51.57 (C-5), 41.47 (C-7), 40.64 (C-8); MS: m/z 370 (M⁺), 231, 216, 242, 166, 154, 139 (100%), 128, 111, 103, 102. Anal. Found: C, 61.33; H, 4.01; N, 7.44. Calcd. for C₁₉H₁₅N₂O₄Cl: C, 61.54; H, 4.07; N, 7.55%.

**5-{1-(4-Methylphenyl)-3-phenyl-1-oxopropyl}-pyrimidine (1H, 3H, 5H)-2,4,6-trione, 3c:** White solid. Yield 10%; m.p. 180-82°C; Rf 0.45; UV-Vis: δ values for protons H-1 and H-3 are exchangeable

† δ values for carbons C-4 and C-6 are exchangeable.
5-{1-(4-Nitrophenyl)-3-phenyl-1-oxopropyl}pyrimidine (1H, 3H, 5H)-2,4,6-trione, 3f: Light brownish solid. Yield 7%; m.p. 201-02°C; Rf 0.27; UV-Vis: 270 (π→π* of C=O), 207 (π→π* of C=C); IR (KBr): 3450, 3230 (OH, NH), 1740, 1710, 1670 (C=O), 1590, 1550 cm−1 (C=C, C=N); 13C NMR (DMSO-d6): δ 11.14 (s, 1H, NH, H-1), 11.07 (s, 1H, NH, H-3)jj, 8.09-7.25 (m, 14H, aromatic), 4.36 (d, 1H, H-7), 3.83 (d, J=3.5, 1H, H-5), 3.61 (dd, J=5.5, J=17.5, 1H, H-8); 13C NMR (DMSO-d6): δ 197.50 (C-9), 169.50 (C-4)jj, 169.37 (C-6)jj, 150.0 (C-2), [128.58, 128.23, 127.88, 127.65, 126.68 (aromatic)], 51.28 (C-5), 41.67 (C-7), 40.15 (C-8); MS: m/z 412 (M+), 413 (100%), 414, 230, 216, 284, 208, 196, 181, 153, 128, 103, 91, 77, 65, 43. Anal. Found: C, 72.55; H, 7.92; N, 6.65. Calcd. for C23H19N2O4: C, 72.50; H, 4.89; N, 6.79%.

5-{1-(4-Chlorophenyl)-3-phenyl-1-oxopropyl}pyrimidine (1H, 3H, 5H)-2,4,6-trione, 3g: Pink white solid. Yield 38%; m.p. 248-50°C; Rf 0.25; UV-Vis: 253 (π→π* of C=O), 210 (π→π* of C=C); IR (KBr): 3450, 3220 (OH, NH), 1740, 1700, 1690 (C=O), 1600, 1580, 1505 cm−1 (C=C, C=N)jj; 1H NMR (DMSO-d6): δ 11.06 (s, 1H, NH, H-1)jj, 11.02 (s, 1H, NH, H-3)jj, 8.0 (d, J=8.5, 2H, H-2´, 6´), 7.60 (d, J=8.5, 2H, H-2´, 6´), 7.06 (d, J=9.0, 2H, H-3´, 5´), 6.82 (d, J=8.5, 2H, H-3´, 5´), 4.11 (m, 1H, H-7), 4.00 (dd, J=8.5, J=18.25, 1H, H-8), 3.75 (d, J=3.5, 1H, H-5), 3.56 (dd, J=6.5, J=18.0, 1H, H-8), 3.70 (s, 3H, Ar-OCH3)jj; 13C NMR (DMSO-d6): δ 197.35 (C-9), 170.21 (C-4)jj, 169.80 (C-6)jj, 150.44 (C-2), [158.32, 138.15, 135.31, 131.10, 129.77, 128.83, 113.68 (aromatic)], 51.75 (C-5), 41.75 (C-7), 40.92 (C-8), 54.94 (Ar-OCH3); MS: m/z 400 (M+), 401, 402, 261, 246, 272, 166, 154, 139 (100%), 134, 132, 128, 111, 107, 103, 91, 65, 43. Anal. Found: C, 79.79; H, 4.22; N, 7.01. Calcd. for C23H20N2O3Cl: C, 79.93; H, 4.27; N, 6.99%.

5-{1-(4-Methylphenyl)-3-(4-methoxyphenyl)-1-oxopropyl}pyrimidine (1H, 3H, 5H)-2,4,6-trione, 3h: Yellow solid. Yield 6%; m.p. 261-63°C; Rf 0.28; UV-Vis: 257 (π→π* of C=O), 209 (π→π* of C=C);
IR (KBr): 3450, 3200 (OH, NH), 1750, 1700 (C=O), 1640, 1600, 1590, 1510 cm⁻¹ (C=C, C=N); ¹H NMR (DMSO-d₆): δ 11.05 (s, 1H, NH, H-1), 11.01 (s, 1H, NH, H-3), 7.88 (d, J = 8.0, 2H, H-2', 6''), 7.07 (d, J = 8.5, 2H, H-3', 5''), 6.83 (d, J = 9.0, 2H, H-2', 6'), 7.34 (d, J = 8.0, 2H, H-3', 5'), 4.12 (m, 1H, H-7), 3.96 (dd, Jvic = 8.0, Jgem = 18.0, 1H, H-8), 3.75 (d, J = 4.0, 1H, H-5), 3.54 (dd, Jvic = 6.75, Jgem = 18.0, 1H, H-5), 2.38 (s, 3H, Ar'-CH₃); ¹³C NMR (DMSO-d₆): δ 197.75 (C-9), 170.27 (C-4)‡, 169.83 (C-6)‡, 150.47 (C-2), [158.29, 143.60, 134.21, 131.21, 127.93, 113.67 (aromatic)], 51.87 (C-5), 41.06 (C-7), 40.72 (C-8), 38.19 (Ar-OCH₃), 21.09 (Ar'-CH₃); MS: m/z 380 (M +), 381, 382, 261, 253, 252, 247, 246, 134, 132, 128, 119 (100%), 107, 103, 91, 77. Anal. Found: C, 66.26; H, 5.34; N, 7.34. Calcd. for C₂₁H₂₀N₂O₅: C, 66.31; H, 5.30; N, 7.36%.

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References