Studies in Pyrimidine-Annelated Heterocycles[1] by Tandem Cyclization: Regioselective Synthesis of [6,6]Pyranopyran by Intramolecular [1,6] Michael Addition[†]

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Introduction

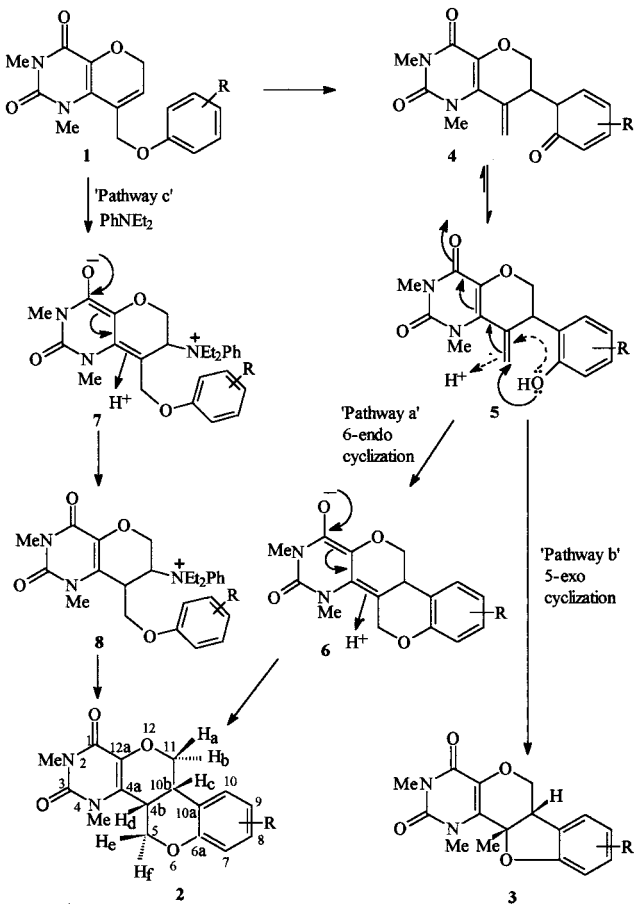
The Claisen rearrangement has been an excellent method for the formation of C−C bonds useful for the construction of heterocyclic rings in fused heterocycles.[2] In continuation of our work on the sigmatropic rearrangement of propargylic ethers of 1,3-dimethyl-5-hydroxypyrimidine-2,4-dione,[3] we have recently reported the regioselective synthesis of a number of 8-aryloxymethyl-1,3-dimethyl-6*H*-pyrano[3,2-*d*]pyrimidine-2,4-diones[4] (**1a**−**e**). These compounds possess an allyl aryl ether moiety which can undergo further Claisen rearrangement to give polyheterocycles **2** and/or **3**. Herein we report the results of this investigation.

Results and Discussion

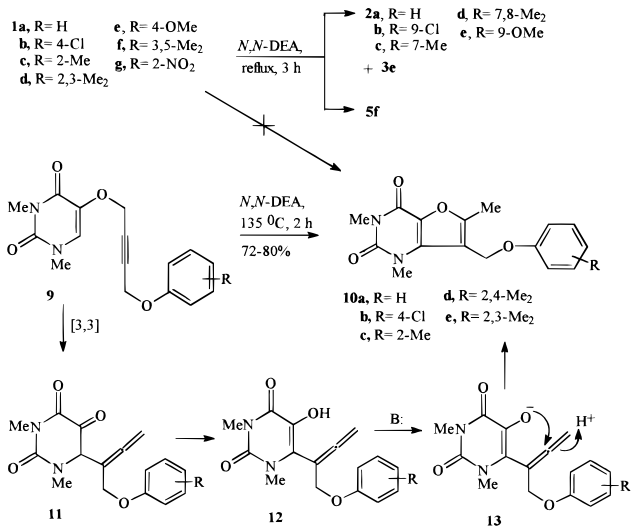
The substrate **1** when subjected to heating above 200 °C is expected to generate phenolic intermediate **5** by a [3,3] sigmatropic rearrangement and enolization. A close examination of the structure of the phenolic intermediate **5** reveals that the phenolic OH group is suitably juxtaposed for a possible intramolecular [1,6] Michael addition (6-endo cyclization) to the dienone system to give [6,6] pyranopyran **2** (pathway a) or it may undergo a normal 5-exo cyclization to afford furopyran system **3** (pathway b). Intermolecular [1,6] Michael addition of *N*,*N*-DEA to the dienone system of **1** followed by a Friedel−Crafts cyclization which is irreversible may also provide [6,6]-pyranopyran **2** (Scheme 1).

N,*N*-Diethylaniline is a versatile solvent for conducting Claisen rearrangement. Therefore, to test the aforementioned possibilities substrate, **1a** was refluxed in *N*,*N*-diethylaniline for 3 h to give a white crystalline solid, mp 204 °C, in 82% yield. The structure for **2a** was assigned on the basis of its elemental analysis and ^{13}C, DEPT, $^{1}H-^{1}H$ COSY, and HETCOR spectra (Scheme 2). Chemical shifts are not separated even under 500 MHz in solvents such as $CDCl_3$, MeOD, and acetone-d_6. The 500 MHz ^{1}H NMR spectrum in C_6H_6 exhibited relatively

Scheme 1



Scheme 2



better separation of chemical shifts and was partly utilized for the assignment of protons. However, during a decoupling study the two desired protons H_c and H_d were not well-separated in C_6H_6 solution. In this case the use of $CDCl_3$ solution was more appropriate. The aliphatic protons H_a, H_b, H_c, H_d, H_e, and H_f were identified by a combination of DEPT, HMBC, and HMQC experiments in $CDCl_3$ solution: δ 3.35 (dt, J = 11.2, 4.6 Hz, H_d), 3.45 (dt, J = 11.7, 4.7 Hz, H_c), 3.94 (t, J = 11.2

[†] This paper is dedicated to Professor B. S. Thyagarajan of the University of Texas at San Antonio on the occasion of his 69th birth anniversary.

(1) For our previous paper, see: Majumdar, K. C.; Das, U.; Jana, N. K. *J. Org. Chem.* **1998**, *63*, 3550.

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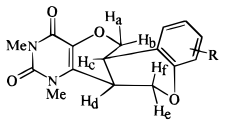

Figure 1.

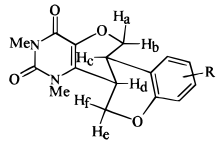

Figure 2.

Hz, H_f), 4.04 (t, J = 11.7 Hz, H_b), 4.50 (ddd, J = 1.1, 4.2, 11.2 Hz, H_a), 4.57 (ddd, J = 1.5, 3.8, 11.3 Hz, H_e). The *cis* stereochemistry was established from decoupling and NOE experiments. Thus decoupling of proton H_c simplifies proton H_d to a dd (J = 4.3, 11.1 Hz). Similarly, irradiation of H_d simplies proton H_c to a dd (J = 4.2, 11.2 Hz). More importantly, irradiation of the two axial protons H_b and H_f together leaves the two ring juncture protons as triplets (J = 5 Hz). Additional support for the *cis* stereochemistry was also obtained from molecular mechanics (MM2) calculations. The *cis* isomer is found to be more stable than the *trans* isomer for **2a** by 3.76 kcal/mol (Figure 1 and Figure 2). Other substrates **1b**−**g** were also subjected to similar treatment. Substrates **1b**, **1c**, and **1d** produced similar products **2b**, **2c**, and **2d** in 72%, 70%, and 70% yields, respectively. However, substrate **1e** gave a mixture of two products **2e** (40%) and **3e** (40%) which were easily separated by column chromatography over silica gel (Scheme 1). That the furopyran derivative **3e** bears a *cis* stereochemistry at the ring juncture is evident from its ^{1}H NMR coupling constant of J = 4.8 Hz. Benzofuro[3,2-*c*]benzopyran derivatives obtained by a similar reaction are known to possess *cis* stereochemistry at the ring juncture.[5] Substrate **1f** did not provide a product arising from cyclization, and **5f** was isolated in 20% yield. The structure of **5f** was assigned on the basis of its elemental analysis and ^{1}H NMR signals at δ 2.20 (s, 6H, 2ArC**H**$_3$), 3.40 (s, 3H, NC**H**$_3$), 3.50 (s, 3H, NC**H**$_3$), 4.20−4.48 (m, 3H, OC**H**$_2$−C**H**−), 4.46 (s, 1H, O**H**), 5.36 (bs, 1H), and 5.52 (bs, 1H) due to *exo*-methylene. Substrate **1g** decomposed to give tarry material from which no tractable product could be obtained. Nitroaryloxy-containing substrates are known to decompose at elevated temperature.[5] Product **3** was obtained only in the case of substrate **1e**, and the phenol intermediate **5f** was obtained only in the case of substrate **1f**. The presence of a sufficiently electron rich group (in this case OMe) on the phenolic moiety seems to bring about the 5-exocyclization (Scheme 1).

Substrates **1a**−**f** were prepared from 5-(4-aryloxybut-2-ynyloxy)-1,3-dimethylpyrimidine-2,4-diones **9** by the thermal [3,3] sigmatropic rearrangement in refluxing chlorobenzene. Therefore, we were interested in carrying out the thermal rearrangement of substrates **9** directly using *N*,*N*-diethylaniline at 130 °C. However, this did not give the products **1** or **2**. Instead, 7-aryloxymethyl-1,3-dimethyl-6-methylfuro[3,2-*d*]pyrimidine-2,4-diones **10a**−**e** were obtained in 72−80% yields (Scheme 2). The formation of **10** from butynyl ether **9** can be explained[6] by an initial [3,3] sigmatropic rearrangement to give **11** and enolization to **12** which undergoes cyclization in the presence of *N*,*N*-DEA to give product **10** (Scheme 2).

The present result is quite different from earlier reports in this area. Similar thermal rearrangements of a number of analogous systems have been reported. 4-Aryloxymethylchrom-3-ene is known[7] to give benzofuro-[3,2-*c*][1]-6a,11a-dihydro-11-methylbenzopyran. 7-Chromenylmethyloxy-4-methylcoumarin[8] and 7-chromenylmethyloxyflavones[9] afforded angularly fused furopyrans. 4-Aryloxymethylpyrano[3,2-*c*]coumarin furnished the phenolic product[10] whereas 1-aryloxymethylpyrano[2,3-*c*]-quinolin-5(6*H*)-ones produced only 1-aryloxymethyl-2-methylfuro[2,3-*c*]quinolin-4(5*H*)-one via an unusual ring contraction.[11] Therefore, this synthesis of fused [6,6]-pyranopyran is noteworthy. Moreover, recently there has been a flurry of activity on the synthesis of [6,6]pyranopyrans.[12] The uracil moiety has been incorporated in the presently synthesized [6,6]pyranopyrans. 5-Substituted uracils have been developed as drugs[13−18] and enzyme inhibitors,[19] but functionalization of uracil at C-5 and C-6 usually requires rather sophisticated and tedious reactions conditions.[20,21] These [6,6]pyranopyran derivatives have the potential to be useful as drugs. The

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methodology described here seems to be simple and facile for the synthesis of these compounds.

Experimental Section

Melting points are uncorrected. UV absorption spectra were recorded in ethanol. IR spectra were run for KBr disks (solid). ^{1}H NMR spectra were performed at the Indian Institute of Chemical Biology, Calcutta, and Department of Chemistry, Nottingham University, England. Elemental analyses and mass spectra were carried out at Regional Sophisticated Instrumentation Centre (Central Drug Research Institute), Lucknow. Silica gel 60−120 mesh was used for chromatographic separation. Petroleum ether refers to the fraction boiling between 60 and 80 °C.

General Procedure for the Preparation of 1,3-Dimethyl-5-[4-aryloxybut-2-ynyloxy]uracils (9a−g). These compounds were prepared according to a published procedure.[4] Compounds **9a**−**e** have already been reported.[4]

1,3-Dimethyl-5-[4-(2,3-dimethylphenoxy)but-2-ynyloxy]-uracil (9f): yield 92%; mp 144 °C; λ_{max} (log ϵ) 224 (3.3), 277 (3.2) nm; IR (KBr) ν_{max} 1680, 1620 cm^{-1}; ^{1}H NMR ($CDCl_3$, 100 MHz) δ 2.12 (s, 3H), 2.24 (s, 3H), 3.16 (s, 3H), 3.32 (s, 3H), 4.72 (s, 4H), 6.72−7.16 (m, 4H); MS *m*/*z* 328 (M^+). Anal. Calcd for $C_{18}H_{20}N_2O_4$: C, 65.85; H, 6.10; N, 8.54. Found: C, 65.97; H, 6.31; N, 8.32.

General Procedure for the Preparation of 8-Aryloxymethyl-1,3-dimethyl-6*H*-pyrano[3,2-*d*]pyrimidine-2,4-diones (1a−g). These compounds were prepared according to a published procedure.[4] Compounds **1a**−**e** have already been reported.[4]

1,3-Dimethyl-8-[3,5-dimethylphenoxymethyl]-6*H*-pyrano-[3,2-*d*]pyrimidine-2,4-dione (1f): yield 90%; mp 114 °C; UV λ_{max} (log ϵ) 224 (3.3), 340 (2.8) nm; IR (KBr) ν_{max} 1680, 1620 cm^{-1}; ^{1}H NMR ($CDCl_3$, 100 MHz) δ 2.08 (s, 3H), 2.24 (s, 3H), 3.40 (s, 3H), 3.42 (s, 3H), 4.60 (d, J = 6 Hz, 2H), 4.80 (s, 2H), 6.24 (t, J = 5 Hz, 1H), 6.56 (m, 3H); MS *m*/*z* 328 (M^+). Anal. Calcd for $C_{18}H_{20}N_2O_4$: C, 65.85; H, 6.10; N, 8.54. Found: C, 65.64; H, 6.39; N, 8.78.

General Procedure for the Rearrangement of 8-Aryloxymethyl-6*H*-pyrano[3,2-*d*]pyrimidine-2,4-diones (1a−g) in *N*,*N*-Diethylaniline. A mixture of compound **1** (1 mmol) in *N*,*N*-diethylaniline (5 mL) was refluxed for 3 h. The reaction mixture was cooled, poured into ice-cold 6 N HCl (30 mL), and extracted with $CHCl_3$ (3 × 20 mL). The $CHCl_3$ layer was washed with water and dried over Na_2SO_4. Removal of $CHCl_3$ gave a viscous liquid which was chromatographed over silica gel. Elution of the column with benzene−ethyl acetate (3:1) furnished products **2a**−**e**, **3e**, and **5f**.

(4b,10b-*cis*)-2,4-Dimethyl-4b,5,10b,11-tetrahydro-4*H*-6,12-dioxa-2,4-diazachrysene-1,3-dione (2a): yield 82%; mp 204 °C; UV λ_{max} (log ϵ) 223 (3.2), 283 (2.9) nm; IR (KBr) ν_{max} 1650, 1620 cm^{-1}; ^{1}H NMR (C_6D_6, 500 MHz) δ 2.34 (dt, J = 11.3, 4.7 Hz, 1H, H_d), 2.72−2.78 (m, 1H, H_c), 2.76 (s, 3H), 3.29 (t, J = 11.33 Hz, 1H, H_e), 3.44 (s, 3H), 3.45 (t, J = 11.33 Hz, 1H, H_a), 3.81 (ddd, J = 11.3, 4.1, 1.4 Hz, 1H, H_f), 4.12 (br dd, J = 11.3, 4.11 Hz, 1H, H_b), 6.75 (dd, J = 6.49, 1.1 Hz, 1H, C_7-**H**), 6.85 (dt, J = 1.1, 7.4 Hz, 1H, C_8-**H**), 7.05 (dd, J = 6.49, 1.1 Hz, 1H, C_{10}-**H**), 7.15 (dt, J = 1.1, 7.4 Hz, 1H, C_9-**H**);^{1}H NMR ($CDCl_3$, 500 MHz) δ 3.35 (dt, J = 11.2, 4.6 Hz, H_d), 3.41 (s, 3H), 3.45 (dt, J = 11.7, 4.7 Hz, H_c), 3.54 (s, 3H), 3.94 (t, J = 11.2 Hz, H_f), 4.04 (t, J = 11.7 Hz, H_b), 4.50 (ddd, J = 1.1, 4.2, 11.2 Hz, H_a), 4.57 (ddd, J = 1.5, 3.8, 11.3 Hz, H_e), 6.92 (dd, J = 8, 1.2 Hz, 1H), 7.01 (dt, J = 1.2, 7.48 Hz, 1H), 7.19 (dd, J = 8, 1.2 Hz, 1H), 7.24 (dt, J = 1.2, 7.48 Hz, 1H); ^{13}C ($CDCl_3$, 60 MHz) 28.6 (CH_3), 30.9 (CH_3), 31.1 (C_{4b}), 32.4 (C_{10b}), 65.1 (C_{11}), 67.1 (C_5), 117.3 (C_7), 118.1 (C_{10a}), 121.7 (C_9), 127.0 (C_{4a}), 129.1 (C_8), 130.0 (C_{10}), 131.9 (C_{12a}), 150.0 (C_1), 154.4 (C_{6a}), 158.5 (C_3); MS *m*/*z* 300 (M^+). Anal. Calcd for $C_{16}H_{16}N_2O_4$: C, 64.00; H, 5.33; N, 9.33. Found: C, 64.15; H, 5.53; N, 9.03.

(4b,10b-*cis*)-9-Chloro-2,4-dimethyl-4b,5,10b,11-tetrahydro-4*H*-6,12-dioxa-2,4-diazachrysene-1,3-dione (2b): yield 72%; mp 242 °C; UV λ_{max} (log ϵ) 225 (3.2), 292 (2.9) nm; IR (KBr) ν_{max} 1650, 1620 cm^{-1}; ^{1}H NMR ($CDCl_3$, 100 MHz) δ 3.16−3.32 (m, 2H), 3.36 (s, 3H), 3.48 (s, 3H), 3.92 (dt, J = 4, 11 Hz, 2H), 4.36−4.56 (m, 2H), 6.76−6.88 (m, 1H), 7.12−7.24 (m, 2H); MS *m*/*z* 334, 336 (M^+). Anal. Calcd for $C_{16}H_{15}ClN_2O_4$: C, 57.49; H, 4.49; N, 8.38. Found: C, 57.37; H, 4.59; N, 8.58.

(4b,10b-*cis*)-2,4,7-Trimethyl-4b,5,10b,11-tetrahydro-4*H*-6,12-dioxa-2,4-diazachrysene-1,3-dione (2c): yield 70%; mp 176 °C; UV λ_{max} (log ϵ) 223 (3.2), 283 (2.9) nm; IR (KBr) ν_{max} 1660, 1620 cm^{-1}; ^{1}H NMR ($CDCl_3$, 100 MHz) δ 2.24 (s, 3H), 3.24−3.40 (m, 2H), 3.48 (s, 3H), 3.60 (s, 3H), 3.97 (dt, J = 4, 11 Hz, 2H), 4.48−4.68 (m, 2H), 6.78−7.24 (m, 3H); MS *m*/*z* 314 (M^+). Anal. Calcd for $C_{17}H_{18}N_2O_4$: C, 64.97; H, 5.73; N, 8.92. Found: C, 64.77; H, 5.63; N, 8.71.

(4b,10b-*cis*)-2,4,7,8-Tetramethyl-4b,5,10b,11-tetrahydro-4*H*-6,12-dioxa-2,4-diazachrysene-1,3-dione (2d): yield 70%; mp 220 °C; UV λ_{max} (log ϵ) 223 (3.3), 284 (3.0) nm; IR (KBr) ν_{max} 1660, 1630 cm^{-1}; ^{1}H NMR ($CDCl_3$, 100 MHz) δ 2.12 (s, 3H), 2.24 (s, 3H), 3.16−3.36 (m, 2H), 3.40 (s, 3H), 3.52 (s, 3H), 3.86 (t, J = 11 Hz, 2H), 4.36−4.64 (m, 2H), 6.80 (d, J = 8 Hz, 1H), 6.94 (d, J = 8 Hz, 1H); MS *m*/*z* 328 (M^+). Anal. Calcd for $C_{18}H_{20}N_2O_4$: C, 65.85; H, 6.10; N, 8.54. Found: C, 65.97; H, 6.30; N, 8.33.

(4b,10b-*cis*)-2,4-Dimethyl-9-methoxy-4b,5,10b,11-tetrahydro-4*H*-6,12-dioxa-2,4-diazachrysene-1,3-dione (2e): yield 40%; mp 174 °C; UV λ_{max} (log ϵ) 224 (3.3), 295 (3.1) nm; IR (KBr) ν_{max} 1650, 1630 cm^{-1}; ^{1}H NMR (DMSO-d_6, 300 MHz) δ 3.18 (s, 3H), 3.21−3.36 (m, 2H), 3.41 (s, 3H), 3.70 (s, 3H), 3.88 (dt, J = 4.1, 11.4 Hz, 2H), 4.42 (dt, J = 4.2, 11.4 Hz, 2H), 6.77 (d, J = 1.5 Hz, 2H), 6.94 (s, 1H); MS *m*/*z* 330 (M^+). Anal. Calcd for $C_{17}H_{18}N_2O_5$: C, 61.82; H, 5.45; N, 8.48. Found: C, 61.92; H, 5.25; N, 8.78.

(4b,9b-*cis*)-2,4-Dimethyl-4b-methyl-4b,9b,10-trihydro-4*H*-8-methoxy-2,4-diazabenzofuro[3,2-*c*]benzopyran-1,3-dione (3e): yield 40%; mp 160 °C; UV λ_{max} (log ϵ) 225 (3.1), 294 (2.9) nm; IR (KBr) ν_{max} 1650, 1620 cm^{-1}; ^{1}H NMR ($CDCl_3$, 300 MHz) δ 1.71 (s, 3H), 3.39 (s, 3H), 3.41−3.44 (m, 1H), 3.57 (t, J = 11 Hz, 1H), 3.74 (s, 3H), 3.78 (s, 3H), 4.36 (dd, J = 11, 4.8 Hz, 1H), 6.77 (d, J = 2 Hz, 2H), 6.82 (d, J = 5.4 Hz, 1H); MS *m*/*z* 330 (M^+). Anal. Calcd for $C_{17}H_{18}N_2O_5$: C, 61.82; H, 5.45; N, 8.48. Found: C, 61.97; H, 5.75; N, 8.18.

1,3-Dimethyl-7-[3,5-dimethyl-2-hydroxyphenyl]-8-methylene-6,7-dihydropyrano[3,2-*d*]pyrimidine-2,4-dione (5f): yield 20%; mp 214 °C; UV λ_{max} (log ϵ) 223 (3.2), 321 (2.9) nm; IR (KBr) ν_{max} 3300 (br), 1650, 1630 cm^{-1}; ^{1}H NMR ($CDCl_3$, 100 MHz) δ 2.20 (s, 6H), 3.40 (s, 3H), 3.50 (s, 3H), 4.20−4.48 (m, 3H), 4.76 (s, 1H, O**H**), 5.36 (bs, 1H), 5.52 (bs, 1H), 6.72−6.96 (m, 2H); MS *m*/*z* 328 (M^+). Anal. Calcd for $C_{18}H_{20}N_2O_4$: C, 65.85; H, 6.10; N, 8.54. Found: C, 65.75; H, 6.33; N, 8.74.

General Procedure for the Rearrangement of 1,3-Dimethyl-5-[4-aryloxybut-2-ynyloxy]uracils (9a−e). Compound **9** (1 mmol) was heated in *N*,*N*-diethylaniline (5 mL) at 135 °C for 2 h. The reaction mixture was cooled, poured into ice-cold 6 N HCl (30 mL), and extracted with $CHCl_3$ (3 × 20 mL). The $CHCl_3$ layer was washed with water and dried over Na_2SO_4. Removal of $CHCl_3$ gave a viscous liquid which was chromatographed over silica gel. Elution of the column with benzene−ethyl acetate (3:1) furnished products **10a**−**e**.

1,3,6-Trimethyl-7-phenoxymethylfuro[3,2-*d*]pyrimidine-2,4-dione (10a): yield 80%; mp 186 °C; UV λ_{max} (log ϵ) 223 (3.1), 277 (3.0) nm; IR (KBr) ν_{max} 1650, 1630 cm^{-1}; ^{1}H NMR ($CDCl_3$, 200 MHz) δ 2.46 (s, 3H), 3.43 (s, 3H), 3.61 (s, 3H), 4.95 (s, 2H), 6.94−7.08 (m, 3H), 7.30−7.38 (m, 2H); MS *m*/*z* 300 (M^+). Anal. Calcd for $C_{16}H_{16}N_2O_4$: C, 64.00; H, 5.33; N, 9.33. Found: C, 64.23; H, 5.53; N, 9.12.

1,3,6-Trimethyl-7-[4-chlorophenoxymethyl]furo[3,2-*d*]-pyrimidine-2,4-dione (10b): yield 75%; mp 208 °C; UV λ_{max} (log ϵ) 224 (3.4), 280 (3.1) nm; IR (KBr) ν_{max} 1700, 1660 cm^{-1}; ^{1}H NMR ($CDCl_3$, 100 MHz) δ 2.52 (s, 3H), 3.52 (s, 3H), 3.72 (s, 3H), 5.00 (s, 2H), 6.96 (dd, J = 8, 2 Hz, 2H), 7.40 (dd, J = 8, 2 Hz, 2H); MS *m*/*z* 334, 336 (M^+). Anal. Calcd for $C_{16}H_{15}ClN_2O_4$: C, 57.49; H, 4.49; N, 8.38. Found: C, 57.28; H, 4.69; N, 8.59.

1,3,6-Trimethyl-7-(2-methylphenoxymethyl)furo[3,2-*d*]-pyrimidine-2,4-dione (10c): yield 72%; mp 178 °C; UV λ_{max} (log ϵ) 223 (3.2), 283 (2.9) nm; IR (KBr) ν_{max} 1660, 1640 cm^{-1}; ^{1}H NMR ($CDCl_3$, 100 MHz) δ 2.18 (s, 3H), 2.45 (s, 3H), 3.43 (s, 3H), 3.63 (s, 3H), 4.95 (s, 2H), 6.94 (t, J = 7 Hz, 2H), 7.18 (d, J = 7 Hz, 2H); MS *m*/*z* 314 (M^+). Anal. Calcd for $C_{17}H_{18}N_2O_4$: C, 64.97; H, 5.73; N, 8.92. Found: C, 64.77; H, 5.94; N, 8.62.

1,3,6-Trimethyl-7-(2,4-dimethylphenoxymethyl)furo[3,2-*d*]pyrimidine-2,4-dione (10d): yield 80%; mp 160 °C; UV λ_{max}

(log ϵ) 223 (3.3), 279 (3.2) nm; IR (KBr) ν_{max} 1700, 1660 cm^{-1}; ^1H NMR ($CDCl_3$, 100 MHz) δ 2.20 (s, 3H), 2.34 (s, 3H), 2.54 (s, 3H), 3.52 (s, 3H), 3.72 (s, 3H), 5.00 (s, 2H), 6.84−7.24 (m, 3H); MS *m*/*z* 328 (M^+). Anal. Calcd for $C_{18}H_{20}N_2O_4$: C, 65.85; H, 6.10; N, 8.54. Found: C, 65.77; H, 6.35; N, 8.78.

1,3,6-Trimethyl-7-(2,3-dimethylphenoxymethyl)furo[3,2-*d*]pyrimidine-2,4-dione (10e): yield 72%; mp 192 °C; UV λ_{max} (log ϵ) 224 (3.4), 277 (3.2) nm; IR (KBr) ν_{max} 1700, 1670 cm^{-1}; ^1H NMR ($CDCl_3$, 100 MHz) δ 2.18 (s, 3H), 2.32 (s, 3H), 2.50 (s, 3H), 3.54 (s, 3H), 3.72 (s, 3H), 5.00 (s, 2H), 6.80−7.24 (m, 3H); MS *m*/*z* 328 (M^+). Anal. Calcd for $C_{18}H_{20}N_2O_4$: C, 65.85; H, 6.10; N, 8.54. Found: C, 65.71; H, 6.36; N, 8.77.

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