

## Synthesis of some 7-methyl-3-(2-oxo-2*H*-chromen-3-yl)-5*H*[1,3]thiazolo[3,2-*a*]-pyrimidin-5-ones

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Reaction of 3-(2-amino-4-thiazolyl)coumarins with ethyl acetoacetate in a mixture of PPA and  $\text{POCl}_3$  give 7-methyl-3-(2-oxo-2*H*-chromen-3-yl)-5*H*-thiazolo[3,2-*a*]pyrimidin-5-ones in a single step. Alternatively condensation of 3-(2-amino-4-thiazolyl)coumarins **1** with acetoacetic ester (EAA) **2** results in the formation of Schiff bases **3a-g**. These on further reaction with polyphosphoric acid (PPA) and phosphorus oxychloride ( $\text{POCl}_3$ ) give corresponding cyclised compounds. 7-methyl-3-(2-oxo-2*H*-chromen-3-yl)-5*H*[1,3]thiazolo[3,2-*a*]pyrimidin-5-ones **4a-g**. The structures of newly prepared compounds have been confirmed from analytical and spectral data.

**Keywords:** Benzopyran-2-one, pyrimidin-5-ones, thiazoles, thiazolo pyrimidones

Coumarins constitute an important class of naturally occurring oxygen ring compounds<sup>1</sup>. The chemistry of Coumarin derivatives continues to draw attention of synthetic organic chemists due to their varied biological activities<sup>2-5</sup>. Further thiazoles<sup>6</sup> and coumarin derivatives with heterocyclic system at 3<sup>rd</sup> position exhibit promising biological activities<sup>7</sup>. A literature survey revealed that thiazoles are generally prepared by Hantzsch thiazole synthesis from  $\alpha$ -haloketones and thioureas and thioamides<sup>8</sup>. Later King *et al*<sup>9,10</sup> and other workers<sup>11</sup> synthesized amino thiazoles by replacing  $\alpha$ -haloketones with ketone and halogen. Despite this modification the method still remains cumbersome and time-consuming (24-25 hr reflux)<sup>12</sup>.

In continuation of earlier work on the synthesis of heterocyclic systems derived from coumarin<sup>13-16</sup>, synthesis of heterocyclic thiazolo pyrimidine-5-ones derived from coumarins is reported.

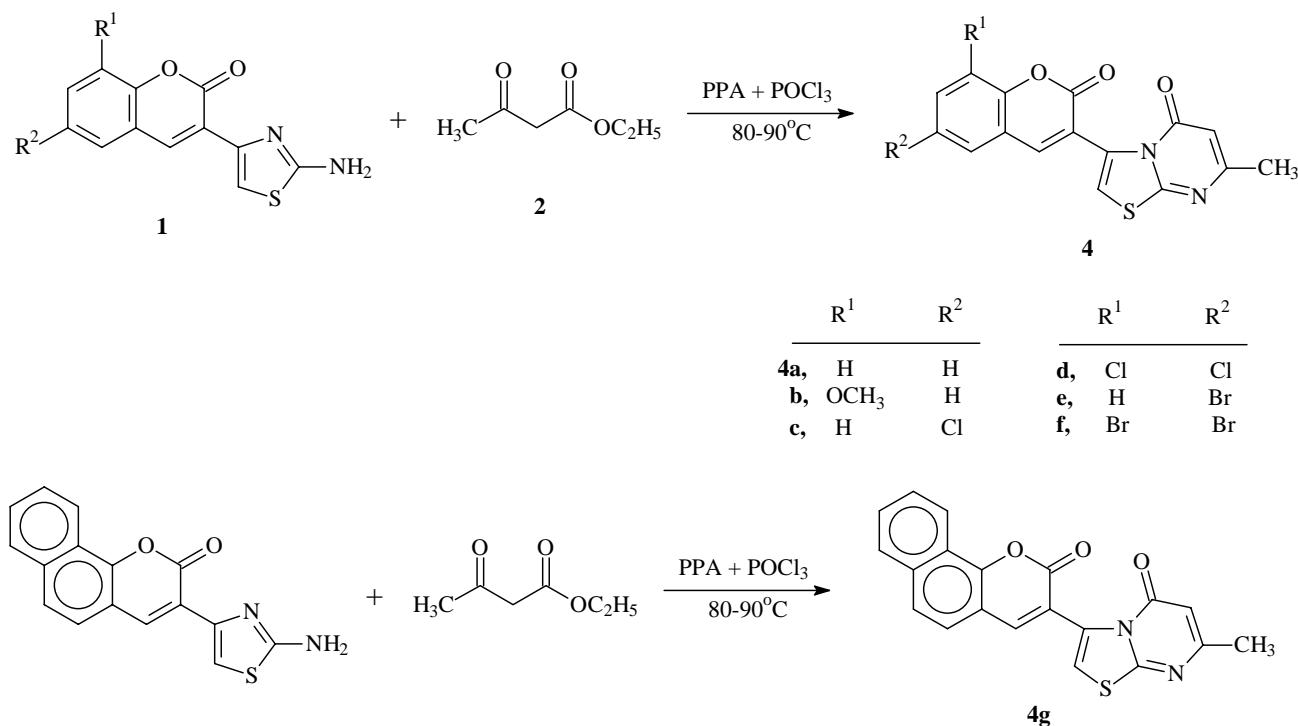
Cyclocondensation reaction of 3-(2-amino-4-thiazolyl)coumarin with ethyl acetoacetate in a mixture of  $\text{POCl}_3$  + PPA gave 7-methyl-3-(2-oxo-2*H*-chromen-3-yl)-5*H*[1,3]thiazolo[3,2-*a*]pyrimidin-5-ones **4** (**Scheme I**). This is a one step process.

Ring closure of the ester **3a-g** does not proceed in  $\text{POCl}_3$  or PPA alone and similar is true in the one pot synthesis of title compounds **4a-g**. Maximum yields of **4a-g** can be achieved, however, by adding some what more than a catalytic amount of PPA to the mixture containing reactants and  $\text{POCl}_3$ .  $\text{POCl}_3$  acts both as a

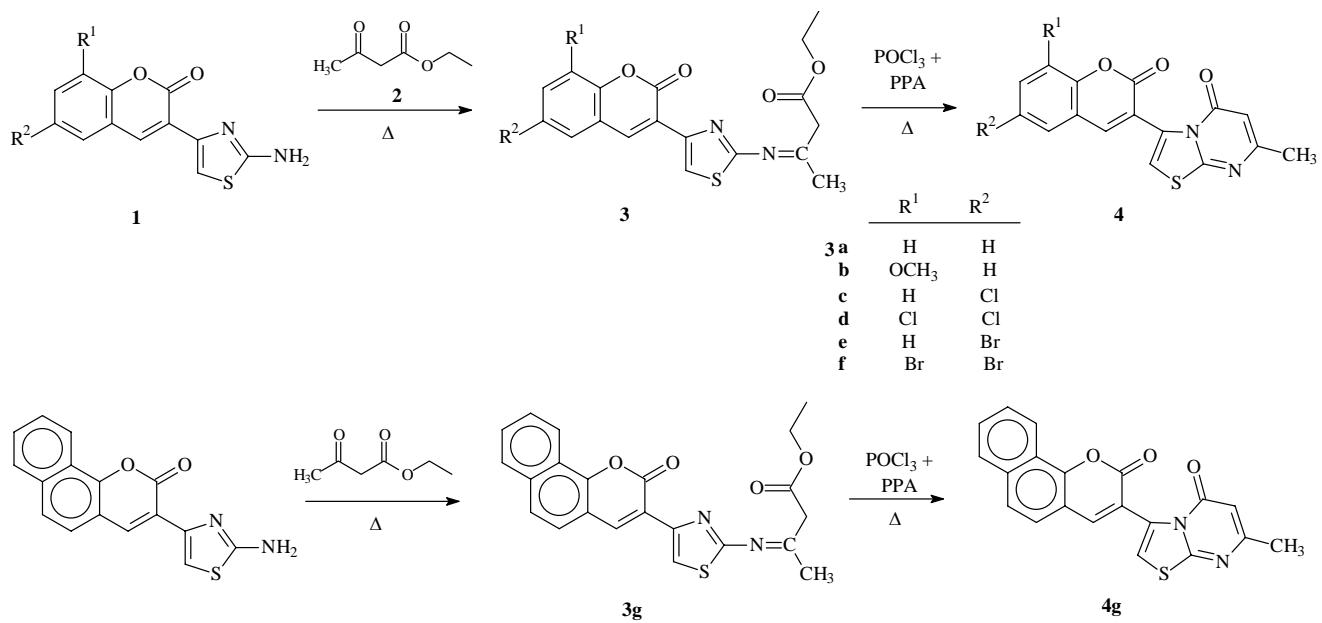
solvent and as an alcohol or water scavenger, rendering cyclization irreversible, but its presence is also advantageous during workup. The mixed reagent has already been used for the preparation of several nitrogen bridgehead systems<sup>17-19</sup> but its scope has not been studied in detail. The present article describes the importance of mixed reagent in the preparation of the title bridgehead system. The yields are maximum in one step process (70-85%).

Reaction of 3-(2-bromoacetyl) coumarins with thiourea resulted in the formation of 3-(2-amino-4-thiazolyl) coumarins **1**, condensation of these compounds with ethyl acetoacetate (EAA) **2** gave the corresponding schiff bases **3**. These on cyclization with a mixture of PPA+ $\text{POCl}_3$  gave the 7-methyl-3-(2-oxo-2*H*-chromen-3-yl)-5*H*[1,3]thiazolo[3,2-*a*]pyrimidin-5-ones (**Scheme II**). This is a two step process (yield 60-70%, **Table I**).

The characterization data for some representative compounds **3a-f** and **4a-f** has been given. The IR spectra of compounds **3a** showed prominent peaks 1615 (-C=N), 1651 (-COO) and 1716  $\text{cm}^{-1}$  (lactone, -C=O) consistent with the assigned structures. The <sup>1</sup>H NMR spectrum of **3a** showed a characteristic triplet for-CH<sub>3</sub> at  $\delta$  1.31 and singlet for -N=C-CH<sub>3</sub> at 2.48, quartet for -CH<sub>2</sub>- of ethyl at 4.25, at 4.89 for -CH<sub>2</sub>- of side chain. The coumarin C<sub>4</sub> proton appeared as singlet at  $\delta$  8.54. The remaining protons were observed in the usual region.



Scheme I (Method 1)



Scheme II (Method 2)

The IR spectra of compounds **4a** showed prominent peaks at 1684 (-C=O, pyrimidine) and 1720  $\text{cm}^{-1}$  (lactone, -C=O). The  $^1\text{H}$  NMR spectrum of **4a** showed characteristic singlet for  $-\text{CH}_3$  at  $\delta$  6.07 and coumarin C<sub>4</sub> proton appeared as singlet at 8.13. The remaining protons were observed in the usual region.

### Experimental Section

All melting points were determined in open capillaries with a cintex melting point apparatus. The purity of the compounds was checked by TLC plates. IR spectra were recorded on a Perkin-Elmer model 337 IR spectrophotometer.  $^1\text{H}$  NMR (300 MHz)

spectra were recorded on a Varian DPX 300 instrument in  $\text{CDCl}_3$  with tetramethylsilane as an internal standard. Chemical shifts are expressed in  $\delta$  ppm. Mass spectra (EI-MS) were determined on a Jeol-D-300 spectrometer at 70 eV. The 3-(2-bromoacetyl)-chromen-2-ones **1** were prepared by reported procedures<sup>12</sup>.

### Preparation of 7-methyl-3-(2-oxo-2H-chromen-3-yl)-5H-[1,3]thiazolo [3,2-a]-pyrimidin-5-one **4a**

A mixture of 3-(2-amino-4-thiazolyl)-2H-1-benzopyran-2-one (0.01 mole) and ethylacetacetate (0.01 mole) was suspended in  $\text{POCl}_3$  (0.03 mole) at RT. Then to the reaction-mixture freshly prepared polyphosphoric acid (0.003 mole) was added. The temperature of the mixture was raised to 90°C and maintained for 1 hr till the HCl evolution subsides. Reaction-mixture was cooled to 25°C and 25 mL of  $\text{H}_2\text{O}$  was added. Filtered the solid, washed with 5% aqueous  $\text{NaHCO}_3$  solution and recrystallized from methanol.

### Preparation of 3-[4-(2-oxo-2H-chromen-3-yl)-thiazol-2-ylimino]-butyric acid ethyl ester **3a**.

A mixture of 3-(2-amino-4-thiazolyl) coumarin (0.244 g, 1mmole) and ethylacetacetate (5 mL) was

**Table I**—Preparation of 7-methyl-3-(2-oxo-2H-chromen-3-yl)-thiazolo[3,2-a]-pyrimidin-5-ones and its derivatives **4** and preparation of 3-[4-(2-oxo-2H-chromen-3-yl)thiazol-2-ylimino]butyric acid ethyl ester **3a**

Compd	$\text{R}^1$ $\text{R}^2$	Yield (%)	
		Method-1	Method-2
<b>4a</b>	H	85	70
	H		
<b>4b</b>	$\text{OCH}_3$	82	68
	H		
<b>4c</b>	H	80	65
	Cl		
<b>4d</b>	Cl	78	66
	Cl		
<b>4e</b>	H	76	64
	Br		
<b>4f</b>	Br	75	67
	Br		
<b>4g</b>	—	84	60
<b>3a</b>			90
<b>3b</b>			88
<b>3c</b>			80
<b>3d</b>			84
<b>3e</b>			81
<b>3f</b>			86
<b>3g</b>			78

taken. The reaction-mixture was refluxed in an oil bath for about 4 hr, at 140°C, then the mixture was cooled at RT, the solid separated was filtered, dried and recrystallized from methanol. All the other compounds **3b-g** were prepared similarly.

**3-[4-(2-Oxo-2H-chromen-3-yl)-thiazol-2-ylimino]-butyric acid ethyl ester **3a**.** Yellow solid; m.p. 185°C; IR (KBr): 1557 (-C=C-), 1615 (-C=N), 1651 (-COO< ester), 1716 (lactone, -C=O)  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR ( $\text{CDCl}_3$ ):  $\delta$  1.31 (t, 3H,  $\text{CH}_3$  of  $-\text{CH}_2\text{-CH}_3$ ), 2.48 (s, 3H,  $\text{CH}_3$  of  $\text{N}=\text{C-CH}_3$ ), 4.25 (q, 2H,  $\text{CH}_2$  of ethyl) 4.89 (s, 2H,  $-\text{CH}_2-$ ), 7.30-7.58 (m, 4H, Ar-H), 7.99 (s, 1H,  $\text{C}_5\text{-H}$  of thiazole), 8.54 (s, 1H,  $\text{C}_4\text{-H}$  of coumarin). Anal. Calcd. for  $\text{C}_{18}\text{H}_{16}\text{N}_2\text{O}_4\text{S}$ : C, 60.66; H, 4.53; N, 7.86; S, 9.00. Found: C, 60.64; H, 4.50; N, 7.83; S, 8.98%.

**3-[4-(8-Methoxy-2-oxo-2H-chromen-3-yl)-thiazol-2-ylimino]-butyric acid ethylester **3b**.** Yellow solid; m.p. 172°C; IR (KBr): 1540 (C=C), 1614 (-C=N), 1645 (-COO, ester), 1713 (lactone, -C=O)  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR ( $\text{CDCl}_3$ ) :  $\delta$  1.30 (t, 3H,  $\text{CH}_3$  of  $-\text{CH}_2-$ ), 2.45 (s, 3H,  $\text{CH}_3$  of  $\text{N}=\text{C-CH}_3$ ), 4.25 (q, 2H,  $\text{CH}_2$  of ethyl), 4.89 (s, 2H,  $-\text{CH}_2-$ ), 7.35-7.55 (m, 3H, Ar-H), 8.0 (s, 1H,  $\text{C}_5\text{-H}$  of thiazole), 8.45 (s, 1H,  $\text{C}_4\text{-H}$  of coumarin). Anal. Calcd. for  $\text{C}_{19}\text{H}_{18}\text{N}_2\text{O}_5\text{S}$ : C, 59.06; H, 4.70; N, 7.25; S, 8.30. Found: C, 59.04; H, 4.67; N, 7.23; S, 8.27%.

**3-[4-(6-Chloro-2-oxo-2H-chromen-3-yl)-thiazol-2-ylimino]-butyric acid ethyl ester **3c**.** Yellow solid; m.p. 216°C; IR (KBr): 1540 (C=C), 1605 (-C=N), 1659 (-COO, ester), 1731 (lactone, -C=O)  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR ( $\text{CDCl}_3$ ) :  $\delta$  1.31 (t, 3H,  $\text{CH}_3$  of  $-\text{CH}_2\text{-CH}_3$ ), 2.50 (s, 3H,  $\text{CH}_3$  of  $\text{N}=\text{C-CH}_3$ ), 4.25 (q, 2H,  $\text{CH}_2-$  of ethyl), 4.98 (s, 2H,  $-\text{CH}_2-$ ), 7.30-7.56 (m, 3H, Ar-H), 8.10 (s, 1H,  $\text{C}_5\text{-H}$  of thiazole), 8.45 (s, 1H,  $\text{C}_4\text{-H}$  of coumarin). Anal. Calcd for  $\text{C}_{18}\text{H}_{15}\text{N}_2\text{O}_4\text{SCl}$ : C, 55.32; H, 3.87; N, 7.17; S, 8.20. Found: C, 55.30; H, 3.84; N, 7.14; S, 8.16%.

**3-[4-(6,8-Dichloro-2-oxo-2H-chromen-3-yl)-thiazol-2-ylimino]-butyric acid ethyl ester **3d**.** Yellow solid; m.p. 202°C; IR (KBr): 1538 (C=C), 1608 (-C=N), 1659 (-COO, ester), 1731 (lactone, -C=O)  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR ( $\text{CDCl}_3$ ):  $\delta$  1.30 (t, 3H,  $\text{CH}_3$  of  $\text{CH}_2\text{-CH}_3$ ), 2.54 (s, 3H,  $\text{CH}_3$  of  $\text{N}=\text{C-CH}_3$ ), 4.20 (q, 2H,  $\text{CH}_2$  of ethyl), 4.99 (s, 2H,  $-\text{CH}_2-$ ), 7.20 (d, 1H, Ar-H,  $J = 3\text{Hz}$ ), 7.75 (d, 1H, Ar-H,  $J = 3\text{Hz}$ ), 8.00 (s, 1H,  $\text{C}_5\text{-H}$  of thiazole), 9.38 (s, 1H,  $\text{C}_4\text{-H}$  of coumarin). Anal. Calcd. for  $\text{C}_{18}\text{H}_{14}\text{N}_2\text{O}_4\text{SCl}_2$ : C, 50.84; H, 3.32; N, 6.59; S, 7.54. Found: C, 50.81; H, 3.30; N, 6.55; S, 7.50%.

**3-[4-(6-Bromo-2-oxo-2H-chromen-3-yl)-thiazol-2-ylimino]-butyric acid ethyl ester **3e**.** Yellow solid;

m.p. 198°C; IR (KBr): 1540 (C=C), 1604 (-C=N), 1659 (-COO, ester), 1727 (lactone, -C=O)  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR ( $\text{CDCl}_3$ ):  $\delta$  1.31 (t, 3H,  $\text{CH}_3$  of  $\text{CH}_2\text{-CH}_3$ ), 2.54 (s, 3H,  $\text{CH}_3$  of N=C-), 4.25 (q, 2H, - $\text{CH}_2$ - of ethyl), 4.98 (s, 2H, - $\text{CH}_2$ -), 7.40 (d, 1H,  $\text{C}_8\text{-H}$  of coumarin,  $J$  = 7 Hz), 7.65 (d, 1H,  $\text{C}_7$  of coumarin,  $J$  = 8 Hz), 7.98 (d, 1H,  $\text{C}_5$  of coumarin,  $J$  = 2 Hz), 8.10 (s, 1H,  $\text{C}_5\text{-H}$  of thiazole), 8.55 (s, 1H,  $\text{C}_4$  of coumarin). Anal. Calcd. for  $\text{C}_{18}\text{H}_{15}\text{N}_2\text{O}_4\text{SBr}$ : C, 49.67; H, 3.47; N, 6.44; S, 7.37. Found: C, 49.64; H, 3.44; N, 6.40; S, 7.34%.

**3-[4-(6,8-Dibromo-2-oxo-2H-chromen-3-yl)-thiazol-2-ylimino]-butyric acid ethylester 3f.** Yellow solid; m.p. 218°C; IR (KBr): 1538 (C=C), 1625 (-C=N), 1659 (-COO, ester), 1739 (lactone, -C=O);  $^1\text{H}$  NMR ( $\text{CDCl}_3$ ):  $\delta$  1.35 (t, 3H,  $\text{CH}_3$  of ethyl), 2.50 (s, 3H,  $\text{CH}_3$  of N=C- $\text{CH}_3$ ), 4.20 (q, 2H, - $\text{CH}_2$ - of ethyl), 4.99 (s, 2H, - $\text{CH}_2$ -), 7.35 (d, 1H, Ar-H,  $J$  = 3Hz), 7.78 (d, 1H, Ar-H,  $J$  = 2Hz), 8.10 (s, 1H,  $\text{C}_5\text{-H}$  of thiazole), 8.55 (s, 1H,  $\text{C}_4\text{-H}$  of coumarin). Anal. Calcd. for  $\text{C}_{18}\text{H}_{14}\text{N}_2\text{O}_4\text{SBr}_2$ : C, 42.05; H, 2.74; N, 5.45; S, 6.24. Found: C, 42.03; H, 2.70; N, 5.42; S, 6.21%.

**3-[4(2-Oxo-2H-benzo[h] chromen-3-yl)-thiazol-2-ylimino]-butyric acid ethyl ester 3g.** Yellow solid; m.p. 197°C; IR (KBr): 1540 (C=C), 1610 (-C=N), 1635 (-COO, ester), 1722 (lactone, -C=O)  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR ( $\text{CDCl}_3$ ):  $\delta$  1.32 (t, 3H, - $\text{CH}_3$ - of  $\text{CH}_2\text{-CH}_3$ ), 2.56 (s, 3H,  $\text{CH}_3$ - of N=C- $\text{CH}_3$ ), 4.24 (q, 2H, - $\text{CH}_2$ - of ethyl), 4.98 (s, 2H, - $\text{CH}_2$ -), 7.52 – 7.99 (m, 6H, Ar-H), 8.03 (s, 1H,  $\text{C}_5\text{-H}$  of thiazole), 9.31 (s, 1H,  $\text{C}_4\text{-H}$  of coumarin). Anal. Calcd. for  $\text{C}_{22}\text{H}_{18}\text{N}_2\text{O}_4\text{S}$ : C, 65.01; H, 4.46; N, 6.89; S, 7.89. Found: C, 65.00; H, 4.44; N, 6.80; S, 7.85%.

**Preparation of 7-methyl-3-(2-oxo-2H-chromen-3-yl)-thiazolo[3,2-a] pyrimidin-5-one 4a.** A mixture of 3-[4-(2-oxo-2H-chromen-3-yl)-thiazol-2-ylimino]-butyric acid ethyl ester (0.01 mole) and polyphosphoric acid (0.025 mole) was heated to 80°C, then added phosphorus oxychloride (0.05 mole). The reaction-mixture was refluxed for about 2 hr, at 80-85°C. The mixture was cooled at RT and diluted with 20 mL of water. The solid separated was filtered, washed with 5% sodium bicarbonate solution and recrystallised from methanol. All the other compounds **4b-g** were prepared similarly.

**7-Methyl-3-(2-oxo-2H-chromen-3-yl)-thiazolo[3,2-a]pyrimidin-5-one 4a.** Brown solid; m.p. 230°C; IR (KBr): 1600 (-C=N), 1684 (-C=O, pyrimidone), 1720 (lactone, -C=O)  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR ( $\text{DMSO-}d_6$ ) :  $\delta$  2.28 (s, 3H, - $\text{CH}_3$ ), 6.07 (s, 1H, pyrimidone), 7.31-7.79 (m, 5H, Ar-H), 8.13 (s, 1H,  $\text{C}_4\text{-H}$  of coumarin).

Anal. Calcd. for  $\text{C}_{16}\text{H}_{10}\text{N}_2\text{O}_3\text{S}$ : C, 61.93; H, 3.25; N, 9.03; S, 10.33. Found: C, 61.90; H, 3.22; N, 9.00; S, 10.30%.

**3-(6-Chloro-2-oxo-2H-chromen-3-yl)-7-methyl-thiazolo[3,2-a]pyrimidin-5-one 4c.** Brown solid; m.p. >300°C; IR (KBr): 1603 (-C=N), 1676 (-C=O, pyrimidone), 1724 (lactone, -C=O)  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR ( $\text{DMSO-}d_6$ ):  $\delta$  2.28 (s, 3H, - $\text{CH}_3$ ), 6.08 (s, 1H, pyrimidone), 7.53 (d, 1H,  $J$ =8Hz, Ar-H), 7.62 (s, 1H,  $\text{C}_5$  of thiazole), 7.69-7.73 (dd, 1H, Ar-H), 7.92 (d, 1H,  $J$ =2.4Hz, Ar-H), 8.08 (s, 1H,  $\text{C}_4\text{-H}$  of coumarin). Anal. Calcd. for  $\text{C}_{16}\text{H}_{9}\text{N}_2\text{O}_3\text{SBr}$ : C, 55.74; H, 2.63; N, 8.13; S, 9.30. Found: C, 55.70; H, 2.60; N, 8.10; S, 9.26%.

**7-Methyl-3-(6-bromo-2-oxo-2H-chromen-3-yl)-thiazolo[3,2-a]pyrimidin-5-one 4e.** Brown solid; m.p. >264°C; IR (KBr): 1637 (-C=N), 1654 (-C=O, pyrimidone) and 1719 (lactone, -C=O)  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR ( $\text{DMSO-}d_6$ ):  $\delta$  2.50 (s, 3H, - $\text{CH}_3$ ), 7.20 (s, 1H, pyrimidone), 8.10 (s, 1H, of the thiazole), 7.4-7.8 (m, 3H, Ar-H) 8.10 (s, 1H, of thiazolo pyrimidone) and 8.50 (s, 1H,  $\text{C}_4$  of coumarin). Anal. Calcd. for  $\text{C}_{16}\text{H}_{9}\text{N}_2\text{O}_3\text{SBr}$ : C, 49.37; H, 2.33; N, 7.20; S, 8.24. Found: C, 49.34; H, 2.30; N, 7.17; S, 8.20%.

**7-Methyl-3-(6,8-dibromo-2-oxo-2H-chromen-3-yl)-thiazolo[3,2-a]-pyrimidin-5-one 4f.** Brown solid; m.p. >300°C; IR (KBr): 1610 (-C=N), 1660 (-C=O, pyrimidone) and 1720 (lactone, -C=O)  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR ( $\text{DMSO-}d_6$ ):  $\delta$  2.50 (s, 3H, - $\text{CH}_3$ ), 6.05 (s, 1H, pyrimidone), 8.05 (d, 1H, Ar-H), 8.15 (d, 1H, Ar-H), 8.3 (s, 1H,  $\text{C}_5$  of thiazole) and 8.5 (s, 1H,  $\text{C}_4$  of coumarin). Anal. Calcd. for  $\text{C}_{16}\text{H}_8\text{N}_2\text{O}_3\text{SBr}_2$ : C, 41.05; H, 1.72; N, 5.98; S, 6.85. Found: C, 41.03; H, 1.70; N, 5.94; S, 6.82%.

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