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### Facile Syntheses of Some Thieno[2,3-d] Pyrimidine Derivatives

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## Facile Syntheses of Some Thieno[2,3-d] Pyrimidine Derivatives

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*In one-pot synthesis 2-arylidene-5,6,7,8-tetrahydrothiazolo[3,2-a] cyclopenteno-thieno[2,3-d] pyrimidine-3,5-diones (3) were prepared via the reaction of a ternary mixture of 2-thioxo-1,3,4,5,6,7-hexahydr cyclopentinothieno [2,3-d]-4-one (2), chloroacetic acid and a proper aldehyde. Compound 2 reacted with 3-chloropent-2, 4-dione in ethanolic potassium hydroxide yielding the S-acetyl acetone derivative 5f. The latter compound reacted with hydrazine hydrate and phenyl hydrazine yielded the 2-pyrazolthio derivative 10a, b. Compound 5f also underwent cyclization on heating with acetic acid—pyridine solution to give 11. The 2-methylthio derivative 5a, when treated with hydrogen peroxide gave the corresponding oxidized product 9.*

**Keywords** 1,3-Diketones; pyrazoleyl; thiazolo; thienopyrimidones

## INTRODUCTION

Our interest in thieno [2,3-d] pyrimidine, and their oxo and thioxo derivatives synthesis, emerges from the numerous reports on their biological and physiological activities.<sup>1–15</sup>

We report a facile and convenient route for the synthesis of polynuclear heterocyclic substituted thienopyrimidone, as thiazolothienopyrimidine and pyrazolothienopyrimidine derivatives. Using 1,2,3,5,6,7-hexahydrocyclopentenothieno-[2,3-d]-pyrimidin-4-(4H)one (2), as starting material.

Thus, heating ethyl 2-amino-4H,5H,6H-cycloheptnothiophene-3-carboxylate (1), prepared according to Karl Gewald method,<sup>16</sup> with potassium thiocyanate in dry dioxane in presence of concentrated hydrochloric acid, followed by cyclization with acetic acid gave compound 2 in good yield.

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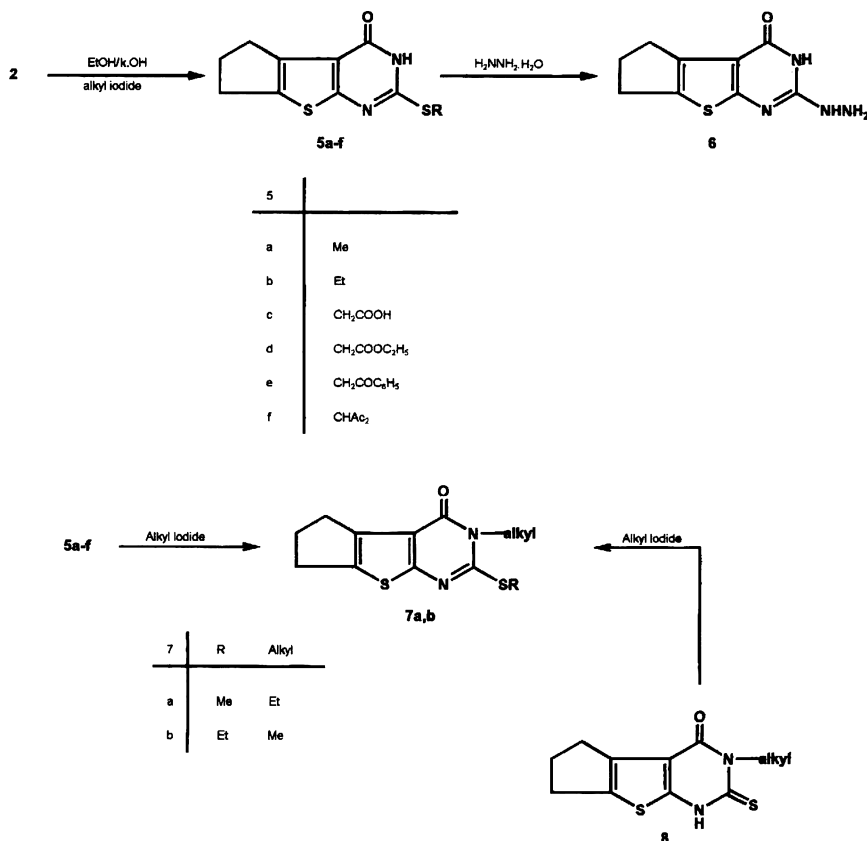


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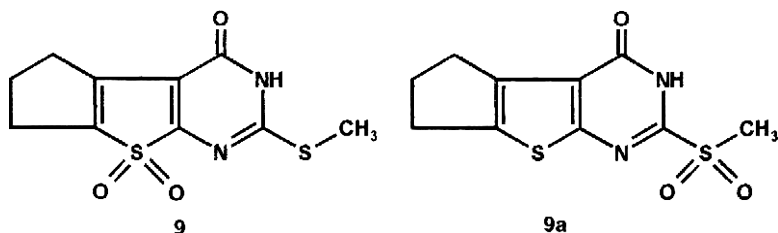
(Scheme 2). Hydrazine hydrate compounds **5a,b** gave the 2-hydrazino derivative **6**, which is conclusive for the structure **5a,b**.



SCHEME 2

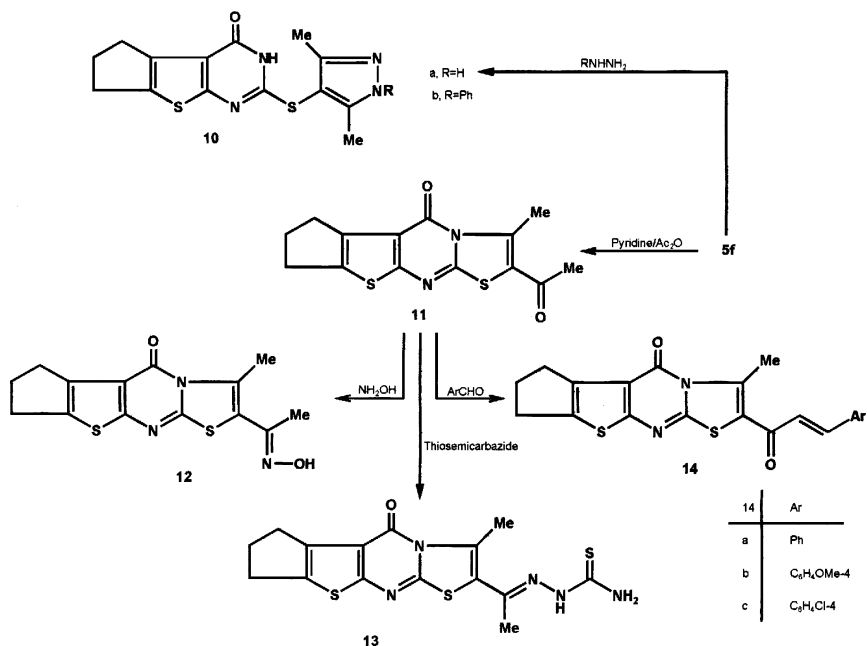
The 2-alkylthio derivatives **5a,b** underwent further alkylation on treatment with alkyl iodide in aqueous ethanolic sodium hydroxide to give the N-3 alkylated products **7a,b** (Scheme 2). Structural assignments of **7** are based on an independent preparation of **7a** by methylation of compound **8** with methyl iodide.<sup>17</sup>

Oxidation of **5a** with hydrogen peroxide in acetic acid yielded structure **9**, rather than its isomeric sulphone derivative **9a**. Structure **9** was preferred upon **9a** on the basis of  $^1\text{H}$ -NMR studies. Thus,  $^1\text{H}$ -NMR spectra ( $\text{DMSO}-d_6$ ) of both **5a** and **9** gave the same value for the methyl group at  $\delta$  2.40 ppm, which proves that the oxidation takes place on the sulfur atom of the thiophene ring only.



Compound **5f**, reacted as 1,3-diketone with hydrazine hydrate and phenyl hydrazine, to afford the cyclized products **10a,b** respectively (Scheme 3). The IR spectra of these compounds displayed absorption bands at  $1665\text{ cm}^{-1}$  (CO). The  $^1\text{H-NMR}$  spectrum ( $\text{CF}_3\text{COOD}$ :  $\text{CDCl}_3/2:1$ ) of **10a**, showed signals at  $\delta$  2.56 ( $\text{CH}_2$ ).

Heating **5f** in a mixture of acetic anhydride-pyridine afforded the cyclized product **11**. The IR spectrum of **11** showed no absorption bands in the NH region. The  $^1\text{H-NMR}$  spectrum ( $\text{CDCl}_3$ :  $\text{DMSO-}d_6/1:2$ ) showed signals at  $\delta$  2.30 ( $\text{CH}_2$ ),  $\delta$  2.45 ( $\text{CH}_3$ ),  $\delta$  2.70 ( $\text{CH}_2$ ), and  $\delta$  3.00 ( $\text{CH}_3$ ). In the  $^{13}\text{C-NMR}$  spectrum, resonances for 9sp<sup>2</sup> and 5sp<sup>3</sup> hybridized carbon atoms were observed. The mass spectrum showed  $m/e$  304 (100%



SCHEME 3

M<sup>+</sup>) Compound **11** reacted with each of hydroxylamine hydrochloride and thiosemicarbazide to give derivative **12** and **13** respectively. IR, <sup>1</sup>H-NMR, and mass spectra of **12** gave data, which support the assigned structures (Experimental section). On the other hand, condensation of **11** with aromatic aldehyde gave 2-cinnamoyl derivatives **14a-c**.

The IR spectra of **14b** displayed two carbonyl absorption bands at 1700 and 1695 cm<sup>-1</sup>. The <sup>1</sup>H-NMR spectrum (DMSO-d<sub>6</sub>) of **14b**, showed signals for three methylenic protons at  $\delta$  2.40,  $\delta$  2.95,  $\delta$  3.15 (CH<sub>3</sub>), and the AA'BB' spectrum for aromatic protons at (6.95 and 7.59 (J = 10 Hz). The vinyl protons appeared at (7.76 and 7.78 (J = 16 Hz) indicating the trans configuration of the double bond.

## EXPERIMENTAL

Solid compounds were recrystallized to constant melting points and dried in vacuum in drying pistol containing sodium hydroxide. All melting points are uncorrected and were taken in open capillaries on a Gallen Kamp Apparatus. Microanalyses were carried out at the Microanalytical Unite, National Research Centre and Faculty of Science, Cairo University. IR spectra were carried out on FT/IR 300 E Jasco using KBr discs. <sup>1</sup>H-NMR spectra were measured in DMSO or CDCl<sub>3</sub>, using Joel Ex. 270 NMR spectrometer. Signals were measured with reference to TMS as an internal standard. The Mass spectra were recorded on Finigan SSQ 7000 spectrometer. All reactions were followed up by TLC using CHCl<sub>3</sub>/MeOH (9:1, v/v) and/or ethyl acetate/Benzene (7:3) and detected under UV Lamp.

### Ethyl-2-amino-4H, 5H, 6H-cyclopentenothiophene-3-carboxylate (I)

A mixture of cyclopentanone (0.84 g, 0.01 mol), diethylamine (0.73 g, 0.01 mol), ethylcyanoacetate (1.13 g, 0.01 mol), and ethanol (1 mL.) was stirred at room temperature, then sulfur (0.32 g, 0.01 mol) was added drop wise, over a period of one h. The reaction mixture was then stirred at 40°C for 5 h (the reaction mixture was followed by TLC). The reaction mixture was left to cool at room temperature overnight to form orange crystals. The formed crystals were filtered off and recrystallized from benzene (20 ml) to yield the title product as orange needles (0.161 g, 76%); m.p. 111–112°C; [C<sub>10</sub>H<sub>13</sub>NSO<sub>2</sub>] (211.3); found C, 56.63%; H, 5.96%; N, 6.63%. requires C, 56.85%; H, 6.20%; N, 6.63%; IR (potassium bromide) cm<sup>-1</sup>: 3411 (br NH), 2904 (CH) and 1666 (CO); <sup>1</sup>H-NMR (CF<sub>3</sub>COOD: CDCl<sub>3</sub>, 1:1) (ppm: 1.30 (t, 3H, CH<sub>3</sub>), 2.60 (m, 2H, CH<sub>2</sub>), 2.75

(m, 4H, 2CH<sub>2</sub>), 4.25 (q, 2H, ester CH<sub>2</sub>), and 5.80 (br s, 2H, NH<sub>2</sub>, D<sub>2</sub>O exchangeable); <sup>13</sup>C-NMR (CF<sub>3</sub>COOD:CDCl<sub>3</sub>/1:1) (ppm: 14.5 (CH<sub>3</sub>); 27.3, 28.9, 30.8 and 59.4 (4CH<sub>2</sub>); 102.8, 103.2, 121.4 and, 142.8 (the thiophene ring carbon atoms) and 166.4 (CO). MS (EI+Q1MS LMR UP LR): 211.0 (M<sup>+</sup>) 100%.

### **1, 2, 3, 5, 6, 7-Hexahydrocyclopentenothieno [2,3-d] pyrimidine-4(4H)-one (2)**

Compound **1** (2.11 g, 0.01 mol), potassium thiocyanate (1.94 g, 0.02 mol), and concentrated hydrochloric acid solution (30 mL) was refluxed in dioxane (30 mL) for 5 h (the reaction mixture was followed by TLC). The reaction mixture was cooled and poured into water (100 mL). The deposited precipitate was filtered off and recrystallized from dioxane (30 mL). The product was dissolved in 10% sodium hydroxide solution, and then neutralized with concentrated hydrochloric acid solution to afford colorless precipitate. The formed precipitate was collected by filtration, and it was washed with water and recrystallized from dioxane (30 mL) to yield the title product as colorless powder (1.64 g, 73%); mp. 340–42°C; [C<sub>9</sub>H<sub>8</sub>N<sub>2</sub>S<sub>2</sub>O] (224.3); found: C, 48.2% 6; H, 3.55%; N, 12.49%. Requires: C, 48.19%; H, 3.59%; N, 12.49%; IR (potassium bromide) cm<sup>-1</sup>: 3400, 3293 (broad NH), 2904 (CH) and 1670 (CO); <sup>1</sup>H-NMR (DMSO-d<sub>6</sub>) δ ppm: 2.34 (m, 2H, CH<sub>2</sub>); 2.81 (m, 4H, 2CH<sub>2</sub>); 12.13 (s, 1H, SH) and 13.18 (br s, 1H, NH, D<sub>2</sub>O exchangeable); <sup>13</sup>C-NMR (DMSO-d<sub>6</sub>) ppm: 27.65, 28.15, and 28.38 (CH<sub>2</sub>); 113.4, 133.4, 139.8, and 154.5 (thiophene ring carbon atoms), 156.5 (CS) and 172.9 (CO). MS (EI+Q1MS LMR UP LR): 224.0 (M<sup>+</sup>) 100%.

### **2-Arylmethylene-2,3,4,6,7,8-hexahydrothiazolo[3,2-a] cyclopentenothieno[2,3-d]-Pyrimidin-5H-3, 5-dione 3a–d—General Procedure**

#### **Method A**

A mixture of compound **2** (2.24 g, 0.01 mol), chloroacetic acid (0.95 g, 0.01 mol), appropriate aromatic aldehyde (0.01 mol), and anhydrous sodium acetate (0.02 mol) were stirred under reflux in glacial acetic acid (30 mL) and acetic anhydride (15 mL) for 3 h. The reaction mixture was cooled and poured into water (100 mL). The deposited precipitate was filtered off and recrystallized from the appropriate solvent to yield the title product.



### Method B

A mixture of compound **4** (2.64 g, 0.01 mol), the appropriate aromatic aldehyde (0.01 mol) and anhydrous sodium acetate (0.02 mol) were stirred under reflux in glacial acetic acid (30 ml) and acetic anhydride (15 mL) for 2 h. The reaction mixture was allowed to cool to room temperature and poured into water (100 mL). The deposited precipitate was filtered off and recrystallized from the appropriate solvent to yield the title product.

#### **2-(Phenylmethylene)-2,3,4,6,7,8-hexahydrothiazolo[3,2-a]cyclopentenothieno[2,3-d] primidin-5H-3,5-dione (3a)**

The compound was recrystallized from dimethylformamide (20 mL) to yield the title product as light pale yellow crystals (2.85 g, 81%); m.p. 296–298°C; [C<sub>18</sub>H<sub>12</sub>N<sub>2</sub>S<sub>2</sub>O<sub>2</sub>] (352.3); Found C, 61.35%; H, 3.40%; N, 7.87%. Required: C, 61.36%; H, 3.40%; N, 7.95%; IR (potassium bromide) cm<sup>-1</sup>: 2900 (CH) and 1760, 1697 (2CO); <sup>1</sup>H-NMR (CF<sub>3</sub> COOD: CDCl<sub>3</sub>/1:1) δ ppm: 2.55 (m, 2H, CH<sub>2</sub>), 3.04 (m, 4H, 2CH<sub>2</sub>), 7.59 (s, 1H, CH), and 7.60–7.67 (m, 5H, phenyl protons); <sup>13</sup>C-NMR (CF<sub>3</sub>COOD: CDCl<sub>3</sub>/1:1) δ ppm: 28.7, 29.6 and 30.2 (CH<sub>2</sub>); 67.4 (CH); 109.2, 113.6, 113.8, 118.1, 118.4, 122.3, 122.7, 130.7, 132.3, 133.2, 133.7, 142.4, and 143.5 (thiophene carbon atoms, pyrimidine carbon atom, thiazol carbon atom, and phenyl carbon atoms), 173.6 and 175.2 (2CO); MS (EI+MS/70 ev/T = 225°C) 352 (M<sup>+</sup>) 26.41%, 323 (M-COH) 12.86%, and 133.9 (C<sub>6</sub>H<sub>5</sub>CH -CS<sup>+</sup>) 100%.

#### **2-(4-Methoxyphenylmethylene)-2,3,4,6,7,8-hexahydrothiazolo [3,2-a] cyclopentenothieno [2,3-d]pyrimidine- 5H-3,5-dione (3b)**

The compound was recrystallized from dimethylformamide (20 mL) to yield the title product as yellow crystals (3.17 g, 83%); m.p. 261–262°C; [C<sub>19</sub>H<sub>14</sub>N<sub>2</sub>S<sub>2</sub>O<sub>3</sub>] (382.5); Found: C, 59.11%; H, 4.16%; N, 6.58%. Required: C, 59.66%; H, 3.68%; N, 7.33%; IR (potassium bromide) cm<sup>-1</sup>: 3432 (broad OH); 2926 (CH aliphatic) and 1707, 1695 (2CO); <sup>1</sup>H-NMR (CF<sub>3</sub> COOD: CDCl<sub>3</sub>/1:1) δ ppm: 2.57 (m, 2H, CH<sub>2</sub>), 3.05 (m, 4H, 2CH<sub>2</sub>), 3.96 (s, 3H, OCH<sub>3</sub>), 7.26 (d, 2H, J = 13 Hz, aromatic protons), 7.61 (d, 2H, J = 13 Hz, aromatic protons), and 8.20 (s, 1H, CH, ethylenic proton); <sup>13</sup>C-NMR (CF<sub>3</sub>COOD: CDCl<sub>3</sub>/1:1) δ ppm: 28.7, 29.5, and 30.2 (CH<sub>2</sub>); 56.5 (CH<sub>3</sub>); 67.6 (CH); 108.4, 113.0, 116.3, 117.5, 118.3, 122.0, 126.3, 134.8, 142.5, 142.7, 155.6, and 157.2 (thiophene ring carbon atoms and aromatic carbon atoms, pyrimidine ring carbon atoms, thiazol ring carbon atoms), and 170.8, 175.0 (2CO); MS (EI+MS/70 ev/T: 230°C) 382

(M<sup>+</sup>)97.5%, 533 (M-HCO) 12.5%, 191 (M-C<sub>10</sub>H<sub>8</sub>SO<sub>2</sub>) 10.42%, and 164 (C<sub>9</sub>H<sub>8</sub>SO) 100%.

**2-(4-Chlorophenylmethylene)-2,3,4,6,7,8-hexahydrothiazolo [3,2-a]cyclopentenothieno [2,3-d] pyrimidin-5H-3,5-dione (3c)**

The product was recrystallized from dimethylformamide (20 mL) to yield the title compound as yellow crystals (2.93 g, 76%); m.p. > 300°C; [C<sub>18</sub>H<sub>11</sub>N<sub>2</sub>S<sub>2</sub>O<sub>2</sub>Cl] (286.9); Found: C, 56.11%; H, 2.88%; N, 7.01%. Required: C, 55.88%; H, 2.87%; N, 7.24%; IR (potassium bromide) cm<sup>-1</sup>: 3020 (CH aryl), 293 (CH aliphatic), 1760, 1693 (2CO); <sup>1</sup>H-NMR(DMSO-d<sub>6</sub>) δ ppm: 2.47 (m, 2H, CH<sub>2</sub>), 2.84 (m, 4H, 2CH<sub>2</sub>), 7.61 (d, 2H, J=10 Hz aromatic protons), 7.65 (d, 2H, J=10 Hz, aromatic protons), and 7.98 (s, 1H, ethylenic proton); MS (EI+MS/70 ev/T=250°C) 386 (M<sup>+</sup>) 18.5%, 247 (M-HCO) 8.52%, 222 (C<sub>9</sub>H<sub>5</sub>OCl) 12.20%, and 190 (C<sub>9</sub>H<sub>5</sub>SOCl) 16.00%.

**2-(4-Fluorophenylmethylene)-2,3,4,6,7,8-hexahydrothiazolo [3,2-a]cyclopentenothieno [2,3-d] pyrimidine-5H-3,5-dione (3d)**

The product was recrystallized from dimethylformamide (20 mL) to yield the title compound as yellow crystals (2.88 g, 78%), m.p. > 300°C; [C<sub>18</sub>H<sub>11</sub>N<sub>2</sub>S<sub>2</sub>O<sub>2</sub>F] (370.8); Found: C, 58.27%; H, 2.9%7; N, 7.66%. Required: C, 58.36%; H, 2.99%; N, 7.56%; IR (potassium bromide) cm<sup>-1</sup>: 3020 (CH aryl), 2946 (CH aliphatic) and 1760, 1693 (2CO); <sup>1</sup>H-NMR (CF<sub>3</sub>COOD; CDCl<sub>3</sub>/1:1) δ ppm:2.62 (m, 2H, CH<sub>2</sub>), 3.07 (m, 4H, 2CH<sub>2</sub>), 7.27 (d, 2H, J = 10 Hz aromatic protons), 7.57 (s, 1H, ethylenic proton), and 8.25 (d, 2H, J = 10 Hz, aromatic protons). MS (EI+Q1MS LMR UP LR): 370.0 (M<sup>+</sup>) 100%.

**2,3,4,6,7,8-Hexahydro-5H-thiazolo[3,2-a]cyclopeitlnothieno [2,3-d] pyrimidin-3,5-diones (4)**

A mixture of comopound **2** (2.24 g, 0.01 mol), chloroacetic acid (0.95 g, 0.01 mol) and anhydrous sodium acetate (1.64 g, 0.01 mol) was heated gently, with stirring on a water bath (60°C) for 2 h. The reaction mixture was allowe to cool to room temperature and poured into water (100 mL). The deposited precipitate was filtered off and recrystallized from dimethylformamide (20 mL) to afford the title product as a colorless powder (1.61 g, 61%), m.p. > 300°C; [C<sub>11</sub>H<sub>8</sub>N<sub>2</sub>S<sub>2</sub>O<sub>2</sub>] (264.3); Found: C, 49.62; H, 3.16; N, 10.22. Required: C, 49.98; H, 3.05; N, 10.60%; IR (potassium bromide) cm<sup>-1</sup>: 2928 (CH aliphatic), and 1668, 1665 (2CO); <sup>1</sup>H-NMR (DMSO-d<sub>6</sub>) δ ppm: 2.35 (m, 2H, CH<sub>2</sub>), 2.80 (m, 4H, 2CH<sub>2</sub>) and

3.45 (s, 2H, CH<sub>2</sub>); MS (EI+QIMSLMR UP LR): 264.0 (M<sup>+</sup>) 8.2%, 224.0 (Base peak) 100% and 191.0 (C<sub>2</sub>SO<sub>2</sub>).

### **2-(S-alkyl)-3, 5, 6, 7-tetrahydrocyclopentenothieno [2, 3-d] pyrimidin-4(4H)-one (5a-f) – General Method**

To a warmed ethanolic potassium hydroxide solution [prepared by dissolving potassium hydroxide (0.56 g, 0.01 mol) in ethanol (50 mL)] was added compound **2** (2.24 g, 0.01 mol). The heating was continued for 30 min, the mixture was allowed to cool to room temperature, and the proper halo compound (0.012 mol) was added. The mixture was stirred under reflux for 5 h, then cooled to room temperature and poured onto water (100 mL). The solid product so precipitated was filtered off, washed with water, dried, and recrystallized from the appropriate solvent to produce **5a-f**.

### **2-(Methylthio)-3, 5, 6, 7-tetrahydrocyclopentenothieno [2, 3-d] pyrimidin-4(4H)-one **5a****

From compound **2** (2.24 g, 0.01 mol) and methyl iodide (1.72 g, 0.012 mol). The compound was re-crystallized from dioxane (30 mL) to yield the title product as colorless crystals (1.57 g, 66%), m.p. 273–75°C, [C<sub>10</sub>H<sub>10</sub>N<sub>2</sub>S<sub>2</sub>O] (238.3); Found: C, 50.10%; H, 3.90%; N, 11.55%. Required: C, 50.39%; H, 4.22%; N, 11.75%; IR (potassium bromide) cm<sup>-1</sup>: 3400 (broad NH), 2920 (CH alkyl) and 1680 (CO); <sup>1</sup>H-NMR (DMSO-d<sub>6</sub>) δ ppm (m, 2H, CH<sub>2</sub>), 2.45 (s, 3H, SCH<sub>3</sub>), 2.85 (m, 4H, 2CH<sub>2</sub>), and 8.65 (br s, 1H, NH, D<sub>2</sub>O exchangeable); <sup>13</sup>C-NMR (DMSO-d<sub>6</sub>) δ ppm: 22.9, 27.3, 28.6, and 28.8 (3CH<sub>2</sub> + CH<sub>3</sub>); 116.5, 135.3, 139.3, 156.2, and 157.7 (thienopyrimidone carbon atoms) and 168.3 (CO). MS (EI-MS/70 eV/T = 230°C): 238.0 (M<sup>+</sup>) 18.72%.

### **2-(Ethylthio)-3, 5, 6, 7-tetrahydrocyclopentenothieno [2, 3-d] pyrimidin-4 (4H)-one **5b****

From compound **2** (2.24 g, 0.01 mol) and ethyl iodide (1.86 g, 0.012 mol). The compound was recrystallized from ethanol to yield the title product as colorless crystals (1.70 g, 63%), m.p. 188–189°C; [C<sub>11</sub>H<sub>12</sub>N<sub>2</sub>S<sub>2</sub>O] (252.2); Found: C, 51.73%; H, 4.51%; N, 10.77%. Required: C, 52.35%; H, 4.79%; N, 11.10%; IR (potassium bromide) cm<sup>-1</sup>: 3400 (broad NH), 2925 (CH alkyl) and 1660 (CO); <sup>1</sup>H-NMR (DMSO-d<sub>6</sub>) δ ppm: 1.40 (t, 3H, CH<sub>3</sub>), 2.45 (m, 2H, CH<sub>2</sub>), 2.90 (t, 2H, CH<sub>2</sub>), 3.05 (t, 2H, CH<sub>2</sub>), and 9.70 (br s, 1H, NH, D<sub>2</sub>O exchangeable); <sup>13</sup>C-NMR (DMSO-d<sub>6</sub>) δ ppm: 13.9 (CH<sub>3</sub>); 24.7 (SCH<sub>3</sub>); 27.5, 28.6, and 28.9 (CH<sub>2</sub>); 116.7, 135.6, 139.4, 154.5, and 158.7 (thienopyrimidone carbon

atoms), and 168.6 (CO); MS (EI-MS/70  $\text{ev/T} = 230^\circ\text{C}$ ): 252.2 ( $\text{M}^+$ ) 18.72%.

**2-(Aceticacidthio)-3, 5, 6, 7-tetrahydrocyclopentenothieno [2,3-d] pyrimidin-4 (4H) one 5c**

From compound **2** (2.24 g, 0.01 mol) and chloroacetic acid (1.14 g, 0.012 mol). The compound was recrystallized from dioxane (25 mL) to yield the title product as colorless crystals (1.70 g, 66%), m.p. 242–243°C;  $[\text{C}_{11}\text{H}_{10}\text{N}_2\text{S}_2\text{O}_3]$  (282.3); Found: C, 45.97%; H, 3.32%; N, 9.83%. Required: C, 46.79%; H, 3.57%; N, 9.92%; IR (potassium bromide)  $\text{cm}^{-1}$ : 3240 (broad NH), 2925 (CH alkyl) and 1720, 1688 (2CO);  $^1\text{H-NMR}$  ( $\text{CDCl}_3$ :DMSO- $\text{d}_6$ /1:1)  $\delta$  ppm: 2.45 (m, 2H,  $\text{CH}_2$ ), 2.85 (m, 4H,  $2\text{CH}_2$ ), 3.95 (s, 2H,  $\text{CH}_2$ ), and 5.85 (br s, 1H, NH,  $\text{D}_2\text{O}$  exchangeable);  $^{13}\text{C-NMR}$  ( $\text{CDCl}_3$ : DMSO- $\text{d}_6$ /1:1)  $\delta$  ppm: 27.6, 28.6, 29.0, and 32.6 ( $\text{CH}_2$ ); 116.9, 135.7, 139.5, 154.2, and 158.1 (thienopyrimidone carbon atom), and 168.1, 169.5 (2CO); MS (EI-MS/70  $\text{ev/T} = 240^\circ\text{C}$ ): 282 ( $\text{M}^+$ ) 87% and 264 ( $\text{M}-18$ ) 100%.

**2-(Ethylacetatethio)-3, 5, 6, 7-tetrahydrocyclopentenothieno [2, 3-d] primidin-4 (4H)-one (5d)**

From compound **2** (2.24 g, 0.01 mol) and ethyl bromoacetate (2.00g, 0.012 mol). The compound was recrystallized from ethanol (25 mL) to yield the titled product as colorless crystals (2.20 g, 71%), m.p. 202–203°C;  $[\text{C}_{13}\text{H}_{14}\text{N}_2\text{S}_2\text{O}_3]$  (310.38); Found C, 49.75%; H, 4.22%; N, 9.30%; S, 21.20%. Required: C, 50.30%; H, 4.55%; N, 9.03%; S, 20.66%; IR (potassium bromide)  $\text{cm}^{-1}$ : 3220 (broad NH), 2930 (CH alkyl), and 1720, 1680 ( $\text{C}=\text{N}$ ) and 1480 ( $\text{C}=\text{C}$ );  $^1\text{H-NMR}$  ( $\text{CDCl}_3$ )  $\delta$  ppm: 1.40 (t, 3H,  $\text{CH}_3$ ), 2.45 (m, 2H,  $\text{CH}_2$ ), 2.90 (t, 2H,  $\text{CH}_2$ ), 3.05 (t, 2H,  $\text{CH}_2$ ), 3.25 (q, 2H,  $\text{CH}_2$ ), 3.55 (s, 2H,  $\text{CH}_2$ ), and 9.18 (br s, 1H, NH,  $\text{D}_2\text{O}$  exchangeable); MS (EI + Q1 MS LMR UP LR): 310.0 ( $\text{M}^+$ ) 27.12%.

**2-(Phenacylthio)-3, 5, 6, 7-tetrahydrocyclopentenothieno [2, 3-d] pnmid-in-4(4H)-one (5e)**

From compound **2** (2.42 g, 0.01 mol) and phenacyl bromide (2.38 g, 0.012 mol). The compound was recrystallized from dioxane (30 mL) to yield the title product as pale yellow crystals (2.63 g, 77%), m.p. 257–258°C;  $[\text{C}_{17}\text{H}_{14}\text{N}_2\text{S}_2\text{O}_2]$  (342.4); Found: C, 59.63%; H, 4.22%; N, 8.25%; S, 17.77%. Required: C, 59.62%; H, 4.12%; N, 8.18%; S, 18.72%. IR (potassium bromide)  $\text{cm}^{-1}$ : 3448 (broad OH and NH), 3050 (CH aromatic), 2911 (CH alkyl) and 1680, 1670 (2CO);  $^1\text{H-NMR}$  (DMSO- $\text{d}_6$ )  $\delta$  ppm: 2.30 (m, 2H,  $\text{CH}_2$ ), 2.80 (m, 4H,  $2\text{CH}_2$ ), 4.85 (s, 2H,  $\text{CH}_2$ ),

7.50–7.75 (m, 5H, phenyl protons), and 11.20 (br s, 1H, NH, D<sub>2</sub>O exchangeable). MS (EI+Q1MS LMR UP LR): 222.0 (M<sup>+</sup>) 100%.

**2-(Acetylacetonethio)-3, 5, 6, 7-tetrahydrocyclopentenothieno [2,3-d]-pyrimidin-4(4H)-one. (5f)**

From Compound **2** (2.24 g, 0.01 mol) and chloroacetylacetone (1.61 g, 0.012 mol). The compound was recrystallized from dioxane (30 mL) to yield the title product as yellow crystals (2.58 g, 80%), m.p. 260–262°C; [C<sub>14</sub>H<sub>14</sub>N<sub>2</sub>S<sub>2</sub>O<sub>3</sub>] (322.4); Found: C, 52.24%; H, 4.35%; N, 8.66%. Required: C, 52.15%; H, 4.37%; N, 8.69%; IR (potassium bromide) cm<sup>-1</sup>: 3420 (broad NH, and OH), 2860 (CH alkyl) and 1690, 1590 (2CO); <sup>1</sup>H-NMR (DMSO-d<sub>6</sub>) δ ppm: 2.25 (s, 6H, 2CH<sub>3</sub>), 2.38 (m, 2H, CH<sub>2</sub>), 2.85 (m, 4H, 2CH<sub>2</sub>), 7.81 (s, 1H, CH<sub>2</sub>) and 12.82 (br s, 1H, NH, D<sub>2</sub>O exchangeable); MS (EI+Q1 MS LMRUPLR): 322.1 (M<sup>+</sup>) 87.89%.

**2-Hydrazino-3,5,6,7-tetrahydrocyclopentenothieno [2,3-d] pyrimidin-4(4H)-one (6)**

A mixture of compound **5a** (2.38g, 0.01 mol) and hydrazine hydrate (99–100%) (7 mL, 0.03 mol) in dioxane (20 mL) and ethanol (10 mL) was heated under reflux for 4 h. The solid that separated upon cooling the reaction mixture was filtered off and recrystallized from dioxane-dimethylformamide (4:1, 25 mL) to yield the title compound as a colorless crystals (1.44 g, 65%), m.p. > 300°C; [C<sub>9</sub>H<sub>10</sub>N<sub>4</sub>SO](222.0); Found: C, 48.51%; H, 4.46%; N, 24.91%. Required: C, 48.63%; H, 4.54%; N, 25.21%; IR (potassium bromide) cm<sup>-1</sup>: 3322 (NH<sub>2</sub>), 3245 (NH), 2913 (CH alkyl), and 1666 (CO); <sup>1</sup>H-NMR (DMSO-d<sub>6</sub>) δ ppm: 2.30 (m, 2H, CH<sub>2</sub>), 2.80 (m, 4H, 2CH<sub>2</sub>), and 8.20 (br s, 1H, NH, D<sub>2</sub>O exchangeable); MS (EI+Q1MS LMR UP LR): 222.0 (M<sup>+</sup>) 100%.

**2-Alkylthio-3-alkyl-3,5,6,7-tetrahydrocyclopentano[2,3-d] pyrimidin-4-one (7a,b) – General Procedure**

To a warmed ethanolic sodium ethoxide solution [prepared by dissolving (0.23 g, 0.01 mol) of sodium metal in absolute ethanol (30 mL)] was added each of compound **5a** or **5b** (0.01 mol), the heating was continued for 30 min, the mixture was allowed to cool to room temperature, and the proper alkyl iodide (0.012 mol) was added. The mixture was stirred under reflux for 3 h, cooled to room temperature, and poured into cold water (100 mL). The solid soprecipitated was filtered off, washed

with water, dried and recrystallized from the appropriate solvent to produce **7a,b**.

**2-Ethylthio-3-methyl-4,5,6,7-tetrahydrocyclopentenothieno [2,3-d]pyrimidin-4-one (7a)**

From compound **5b** (2.52 g, 0.01 mol) and methyl iodide (1.72 g, 0.012 mol). The compound was recrystallized from ethanol (20 mL) to yield the title product as colorless crystals (2.16 g, 81%), m.p. 197–198°C; [C<sub>12</sub>H<sub>14</sub>N<sub>2</sub>S<sub>2</sub>O](266.3); Found: C, 54.2%; H, 4.90%; N, 9.89%. Required: C, 54.11%; H, 5.30%; N, 10.52%; IR (potassium bromide) cm<sup>-1</sup>: 2850 (CH alkyl) and 1666 (CO); <sup>1</sup>H-NMR (CDCl<sub>3</sub>) δ ppm: 1.40 (t, 3H, CH<sub>3</sub>), 2.45 (m, 2H, CH<sub>3</sub>), 2.65 (s, 3H, CH<sub>3</sub>), 2.75 (t, 2H, CH<sub>2</sub>), 3.00 (t, 2H, CH<sub>3</sub>), and 4.15 (q, 2H, CH<sub>2</sub>). MS (EI + Q1 MS LMR UP LR): 266 (M<sup>+</sup>) 12%.

**2-Methylthio-3-ethyl-4,5,6,7-tetrahydrocyclopentenothieno [2,3-d]pyrimidin-4-one (7b)**

From compound **5a** (2.38 g, 0.01 mol) and ethyl iodide (1.86 g, 0.012 mol). The compound was recrystallized from ethanol (25 mL) to yield the title product as colorless crystals (2.05 g, 77%), m.p. 207–208°C; [C<sub>12</sub>H<sub>14</sub>N<sub>2</sub>S<sub>2</sub>O] (266.3); Found: C, 53.64%; H, 4.93%; N, 9.70%. Required: C, 54.2%; H, 5.30%; N, 10.52%; IR (potassium bromide) cm<sup>-1</sup>: 2911 (CH alkyl) and 1677 (CO); <sup>1</sup>H-NMR (CDCl<sub>3</sub>) δ ppm: 1.40 (t, 3H, CH<sub>3</sub>), 2.45 (m, 2H, CH<sub>3</sub>), 2.60 (s, 3H, CH<sub>3</sub>), 2.90 (t, 2H, CH<sub>2</sub>), 3.05 (t, 2H, CH<sub>2</sub>), and 4.00 (q, 2H, CH<sub>2</sub>); MS (EI + Q1 MS LMR UP LR): 266 (M<sup>+</sup>) 17%.

**2-(Methylthio)-3,5,6,7-tetrahydrocyclopentenothieno[2,3-d]-pyrimidin-S,S-dioxid-4(4H)-one (9)**

A mixture of compound **5a** (2.24 g, 0.01 mol) and hydrogen peroxide (5 mL) in acetic acid (30 mL) was heated gently with stirring for 6 h; the reaction mixture was allowed to cool to room temperature. The deposited precipitate was filtered off, and recrystallized from dioxane (25 mL) to yield the title product as a colorless crystals (1.56 g, 58%); m.p. 284–286°C; [C<sub>13</sub>H<sub>10</sub>N<sub>2</sub>S<sub>2</sub>O<sub>3</sub>] (270.23); Found: C, 44.12%; H, 3.66%; N, 9.75%; S, 23.11%. Required: C, 44.43%; H, 3.73%; N, 10.3% 6; S, 23.72%; IR (potassium bromide) cm<sup>-1</sup>: 3151 (NH), 2907 (CH alkyl) and 1666 (CO); <sup>1</sup>H-NMR (DMSO-d<sub>6</sub>) δ ppm: 2.35 (m, 2H, CH<sub>2</sub>), 2.45 (s, 3H, SCH<sub>3</sub>), 2.80 (t, 2H, CH<sub>2</sub>), 2.90 (t, 2H, CH<sub>2</sub>), and 11.80 (br s, 1H, NH, D<sub>2</sub>O exchangeable); MS (EI + Q1 MS LMR UP LR): 270.0 (M<sup>+</sup>) 12%.

**2-(3,5-Dimethyl-1H-or(1-phenyl)pyrazol-4-ylthio)-3,5,6,7-tetrahydrocyclopenteno[2,3-d] pyrimidin-4-one (10a,b) – General Procedure**

A mixture of compound **5f** (3.22 g, 0.01 mol) and hydrazine hydrate (99–100%) or phenyl hydrazine hydrochloride (0.01 mol) in dioxane (20 mL) and ethanol (10 mL) was stirred under reflux for 8 h. The reaction mixture was allowed to cool to room temperature and then poured into cold water (100 mL). The deposited precipitate was filtered off, dried, and recrystallized from the appropriate solvent to produce **10a,b**.

**2-(3,5-Dimethyl-1H)pyrazol-4-ylthio)-3,4,5,6,7-tetrahydrocyclopentanothieno[2,3-d]pyrimidin-4-one(10a)**

From compound **5f** (3.22 g, 0.01 mol) and hydrazine hydrate (10 mL). The compound was recrystallized from dioxane (25 mL) to yield the title product as light colorless crystals (2.45 g, 77%), m.p. > 300°C; [C<sub>14</sub>H<sub>14</sub>N<sub>4</sub>S<sub>2</sub>O] (318.4); Found: C, 51.89%; H, 4.13%; N, 16.82%. Required: C, 52.81%; H, 4.43%; N, 17.60%; IR (potassium bromide) cm<sup>-1</sup>: 3420 (NH), 2880 (CH alkyl), and 1660 (CO); <sup>1</sup>H-NMR (CDCl<sub>3</sub>: CF<sub>3</sub>COOD/1:2) δ ppm: 2.56 (m, 2H, CH<sub>2</sub>), 2.61 (s, 6H, 2CH<sub>3</sub>), 3.00 (m, 4H, 2CH<sub>2</sub>), and 8.85 (br s, 1H, NH, D<sub>2</sub>O exchangeable); <sup>13</sup>C-NMR (CDCl<sub>3</sub>: CF<sub>3</sub>COOD/1:2) δ ppm: 10.8 and 10.9 (CH<sub>3</sub>); 29.0, 29.6, and 30.5 (CH<sub>2</sub>), 113.5, 118.0, 118.7, 122.6, 141.5, 144.5, 154.1, and 154.4 (thienopyrimidone carbon atoms and pyrazol ring carbon atoms), and 164.6 (CO); MS (EI-MS/70 eV/T = 265°C): 318 (M<sup>+</sup>) 36.29%.

**2-(3,5-Dimethyl-1-phenylpyrazol-4-ylthio)-3,4,5,6,7-tetrahydrocyclopenteno[2,3-d]pyrimidin-4-one(10b)**

From compound **5f** (3.22 g, 0.01 mol) and phenyl hydrazine hydrochloride (1.45 g, 0.01 mol). The compound was recrystallized from dioxane (30 mL) to yield the title product as light pale yellow crystals (2.60 g, 66%); m.p. 276–277°C; [C<sub>20</sub>H<sub>18</sub>N<sub>4</sub>S<sub>2</sub>O] (394.5); Found: C, 60.81%; H, 4.43%; N, 13.87%. Required: C, 60.88%; H, 4.50%; N, 14.20%; IR (potassium bromide) cm<sup>-1</sup>: 3448 (broad NH), 3050 (CH aromatic), 2826 (CH alkyl) and 1670 (CO); <sup>1</sup>H-NMR (DMSO-d<sub>6</sub>) δ ppm: 2.20 (s, 3H, CH<sub>3</sub>), 2.35 (s, 3H, CH<sub>3</sub>), 2.47 (m, 2H, CH<sub>2</sub>), 2.85 (m, 4H, 2CH<sub>2</sub>), 7.43–7.53 (m, 5H, phenyl protons), and 12.30 (br s, 1H, NH, D<sub>2</sub>O exchangeable); <sup>13</sup>C-NMR (DMSO-d<sub>6</sub>) δ ppm: 11.4 and 11.8 (CH<sub>3</sub>); 27.4, 28.6, and 28.9 (CH<sub>2</sub>); 101.2, 117.1, 124.4, 127.8, 129.2, 135.9, 139.4, 139.5, 144.7, 152.1, 152.8, 154.6, and 157.1 (Thienopyrimidone carbon atoms, pyrazol ring carbon atoms, and phenyl carbon atoms) and 168.8 (CO); MS (EI-MS/70 eV/T = 210°C): 394 (M<sup>+</sup>) 52.51%.

## 2-Acetyl-3-methylthiazolo [3,2-a]-5,6,7,8-tetrahydrocyclopent- enothieno [2,3-d] pyrimidin-5-one (11)

A solution of compound **5f** (3.22 g, 0.01 mol) in a mixture of acetic anhydride/pyridine (20 mL: 10 mL) was stirred under reflux for 3 h. The reaction mixture was allowed to cool to room temperature, then the deposited precipitate was filtered off, dried, and recrystallized from dioxane (30 mL) to yield the title product as a light yellow crystals (2.47 g, 81%), m.p. 206–208°C; [C<sub>14</sub>H<sub>12</sub>N<sub>2</sub>S<sub>2</sub>O<sub>2</sub>] (304.3); Found: C, 55.25%; H, 3.82%; N, 9.29%. Required: C, 55.2%; H, 3.97%; N, 9.21%; IR (potassium bromide) cm<sup>-1</sup>: 2962 (CH alkyl) and 1698, 1660 (2CO); <sup>1</sup>H-NMR (DMSO-d<sub>6</sub>) δ ppm: 2.30 (m, 2H, CH<sub>2</sub>), 2.45 (s, 3H, CH<sub>3</sub>), 2.70 (m, 4H, 2CH<sub>2</sub>), and 3.00 (s, 3H, CH<sub>3</sub>); <sup>13</sup>C-NMR (DMSO-d<sub>6</sub>) δ ppm: 16.5 and 27.6 (CH<sub>3</sub>), 29.1, 29.3, and 30.6 (CH<sub>2</sub>), 114.9, 121.2, 136.2, 139.9, 142.4, 156.6, and 157.3 (thiazolothienoprimumidone carbon atoms) and 167.4, 190.5 (CO); MS (EI-MS/70 eV/T = 160°C): 304 (M<sup>+</sup>) 100%.

## 2-(Acetoxime)-3-methylthiazolo [3,2-a]-5,6,7,8-tetrahydro cyclopentenothieno [2,3-d] pyrimidin-5-one (12)

A mixture of compound **11** (3.04 g, 0.01 mol) and hydroxylamine hydrochloride (0.70 g, 0.01 mol) in dioxane (30 mL) and catalytic amount of piperidine was added. The reaction mixture was stirred under reflux for 8 h, and then it was allowed to cool to room temperature and poured into water (100 mL). The deposited precipitate was filtered off, dried, and recrystallized from dimethylformamide (25 mL) to yield the title product as yellow crystals (1.94 g, 61%), m.p. 237–238°C; [C<sub>14</sub>H<sub>13</sub>N<sub>3</sub>S<sub>2</sub>O<sub>2</sub>] (319.38); found: C, 52.70; H, 3.77; N, 12.60. Required: C, 52.65; H, 4.10; N, 13.16%; IR (potassium bromide) cm<sup>-1</sup>: 3450 (broad OH and NH), 2880 (CH alkyl) and 1680 (CO); <sup>1</sup>H-NMR (DMSO-d<sub>6</sub>) δ ppm: 2.35 (m, 2H, CH<sub>2</sub>), 2.40 (s, 3H, CH<sub>3</sub>), 2.8 (s, 3H, CH<sub>3</sub>), and 2.85 (m, 4H, 2CH<sub>2</sub>); MS (EI-Q1 MS LMR UP LR): 319 (M<sup>+</sup>) 100%; 360 (m/z) 100%; 318 (m/z) 70% and 302 (m/z) 80%.

## 2-(Acetothiosemicarbazone)-3-methylthiazolo [3, 2-a]-5, 6, 7, 8-tetrahydrocyclopenteno-thieno [2,3-d] pyrimidin-4-one (13)

A mixture of compound **11** (3.04 g, 0.01 mol) and thiosemicarbazide (0.91 g, 0.01 mol) in dioxane (30 mL) and catalytic amount of piperidine was added. The reaction mixture was stirred under reflux for 8 h, and then it was allowed to cool to room temperature and poured into water (100 mL). The deposited precipitate was filtered off, dried, and recrystallized from dimethylformamide (25 mL) to yield the title product as



yellow crystals (2.72 g, 72%), m.p. 244–246°C; [C<sub>15</sub>H<sub>15</sub>N<sub>5</sub>S<sub>3</sub>O] (377.5); Found: C, 46.80; H, 3.45; N, 18.61. Required: C, 47.72; H, 4.01; N, 18.55%; IR (potassium bromide) cm<sup>-1</sup>: 3450 (NH<sub>2</sub>), 3150 (NH), 2925 (CH alkyl), and 1689 (CO); <sup>1</sup>H-NMR (DMSO-*d*<sub>6</sub>) δ ppm: 2.30 (s, 3H, CH<sub>3</sub>), 2.40 (m, 2H, CH<sub>2</sub>), 2.85 (s, 3H, CH<sub>3</sub>), 2.95 (m, 4H, 2CH<sub>2</sub>), 7.45 (br s, 2H, NH<sub>2</sub>, D<sub>2</sub>O exchangeable), and 10.65 (br s, 1H, NH, D<sub>2</sub>O exchangeable); MS (EI+Q1 MS LMR UP LR) 377.0 (M<sup>+</sup>) 11.75%.

**2-Cinnamoyl (derivatives)-3-methylthiazolo [3,2-*a*]-5,6,7,8-tetrahydrocyclopenteno-thieno [2, 3-*d*] pyrimidin-5-one (14a-c) – General Procedure**

A mixture of compound **11** (3.22 g, 0.01 mol), the proper aromatic aldehyde (0.01 mol), and a catalytic amount of piperidine was heated at 170–180°C in a test tube for 3 h. The product was solidified by cooling and addition of methanol (50 mL). The precipitate so formed was collected by filtration and recrystallized from the proper solvent to produce **14a-c**.

**2-Cinnamoyl-3-methyl-thiazolo [3,2-*a*]-5,6,7,8-tetrahydrocyclopentenothieno [2,3-*d*] pyrimidin-5-one (14a)**

From compound **11** (3.22 g, 0.01 mol) and benzaldehyde (1.06 g, 0.01 mol). The compound was recrystallized from dioxane (30 mL) to yield the title compound as yellow crystals (2.47 g, 63%), mp. 235–236°C; [C<sub>21</sub>H<sub>16</sub>N<sub>2</sub>S<sub>2</sub>O<sub>2</sub>] (392.48); Found: C, 64.7; H, 3.71; N, 6.77. Required: C, 64.26; H, 4.11; N, 7.14%; IR (potassium bromide) cm<sup>-1</sup>: 3020 (CH aromatic), 2920 (CH alkyl) and 1693, 1680 (2CO); <sup>1</sup>H-NMR (CDCl<sub>3</sub>) δ ppm: 2.45 (m, 2H, CH<sub>2</sub>), 2.95 (t, 2H, CH<sub>2</sub>), 3.10 (t, 2H, CH<sub>2</sub>), 3.15 (s, 3H, CH<sub>3</sub>), 7.05 (d, 1H, *J* = 10 Hz, ethylenic proton), 7.45–7.80 (m, 5H, phenyl protons) and 7.85 (d, 1H, *J* = 10 Hz, ethylenic proton). MS/70 ev/T = 160°C): 304 (M<sup>+</sup>) 100%.

**2-(*p*-Methoxycinnamoyl)-3-methylthiazolo [3,2-*a*]-5,6,7,8-tetrahydrocyclopenteno-thieno [2,3-*d*] pyrimidin-5-one (14b)**

From compound **11** (3.22 g, 0.01 mol) and 4-methoxybenzaldehyde (1.36 g, 0.01 mol). The compound was recrystallized from dioxane (30 mL) to yield the title product as yellow crystals (2.58 g, 61%), m.p. 246–248°C; [C<sub>22</sub>H<sub>18</sub>N<sub>2</sub>S<sub>2</sub>O<sub>2</sub>] (422.5); Found: C, 62.31; H, 3.91; N, 6.50%. Required: C, 62.54; H, 4.29; N, 6.63%; IR (potassium bromide) cm<sup>-1</sup>: 3025 (CH aromatic), 2929 (CH alkyl), 1700, 1695 (2CO), 1519 (C=N), and 14,900 (C=C); <sup>1</sup>H-NMR (CDCl<sub>3</sub>) δ ppm: 2.45 (m, 2H, CH<sub>2</sub>), 2.90 (t, 2H, CH<sub>2</sub>), 3.05 (t, 2H, CH<sub>3</sub>), 3.2 (s, 3H, CH<sub>3</sub>), 3.90 (s, 3H, OCH<sub>3</sub>), 6.90 (d, 1H, *J* = 10 Hz, ethylene proton), 7.00–7.60 (m, 4H, phenyl protons),

and 7–80 (d, 1H,  $J = 10\text{H}_Z$ , ethylenic proton). MS/70 eV/T = 160°C): 422 ( $M^+$ ) 100%.

**2-(*P*-Chlorocmnamoyl)-3-methylthiazolo  
[3,2-*a*]-5,6,7,8-tetrahydrocyclopenteno-thieno [2,3-*d*]  
pyrimidin-5-one (14c)**

From compound **11** (3.22 g, 0.01 mol) and 4-chlorobenzaldehyde (1.41 g, 0.01 mol). The compound was re crystallized from dioxane (30 mL) to yield the title product as yellow crystals (2.82 g, 66%), m.p. 272–275°C,  $[\text{C}_{21}\text{H}_{15}\text{N}_2\text{S}_2\text{O}_2\text{Cl}]$  (426.92); Found: C, 59.10; H, 3.7; N, 6.42. Required: C, 59.07; H, 3.54; N, 6.56%; IR (potassium bromide)  $\text{cm}^{-1}$ : 3015 (CH aromatic), 2929 (CH alkyl) and 1700, 1689 (2CO);  $^1\text{H-NMR}$  ( $\text{CDCl}_3$ )  $\delta$  ppm: 2.45 (m, 2H,  $\text{CH}_2$ ), 2.85 (t, 2H,  $\text{CH}_2$ ), 3.00 (t, 2H,  $\text{CH}_2$ ), 3.25 (s, 3H,  $\text{CH}_3$ ), 6.85 (d, 1H,  $J = 10\text{H}_Z$ , ethylenic proton), 7.00–7.65 (m, 4H, phenyl protons), and 7.85 (d, 1H,  $J = 10\text{H}_Z$ , ethylenic proton). MS/70 eV/T = 160°C): 426.0 ( $M^+$ ) 100%.

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