METHYL N-ARYLDITHIOCARBAMATES: USEFUL REAGENTS FOR THE ANNELATION OF PYRIMIDINES AND 1,3-OXAZINES TO FIVE-MEMBERED HETEROCYCLIC RINGS

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Abstract- The reaction of methyl N-aryldithiocarbamates with π-excessive heteroaromatic o-amino acid derivatives leads to the annelation of 4-oxo-2-thioxopyrimidines, 2,4-dioxopyrimidines or 2-arylamino-1,3-oxazines, depending on the reaction conditions.

The synthesis of fused bi- and polycyclic compounds by annelation of a pyrimidine ring to an existing ring constitutes a very large field, which has been recently reviewed[1]. The structure of the required starting compounds is mainly determined by the nature of the substituents on the pyrimidine ring and, as a general rule, systems with an amino group adjacent to another functional group are the most widely used[1]. Thus, when a 4-oxopyrimidine is to be synthesized, a ring with an amino group adjacent to a carboxylic acid derivative is generally employed, while 2-oxo and 2-thioxo groups are normally introduced by means of isocyanates and ureas or their sulfur analogues in the case of the latter.

In the present paper, and as a continuation of our studies in the field of fused heterocyclic compounds[2], we report a relatively simple general method for the annelation of pyrimidine and 1,3-oxazine rings to five-membered heteroaromatic systems, by the reaction of the appropriate o-amino acid derivatives with methyl N-aryldithiocarbamates[3] 3.

This method has certain advantages compared to previously reported methods:

- Starting compounds 3 are stable solids, obtained in one step from the corresponding anilines.
- The same reagent (i.e. 3) is used to prepare both the 2,4-dioxo and the 4-oxo-2-thioxo derivatives.
- This procedure can be modified to prepare fused 2-arylamino-1,3-oxazine derivatives which are uncommon. Some 6-amino-4-oxo-1H,4H-pyrazolo [3,4-d] [1,3] oxazines have in fact been prepared by us[4], although the corresponding 2-arylamino-4H,

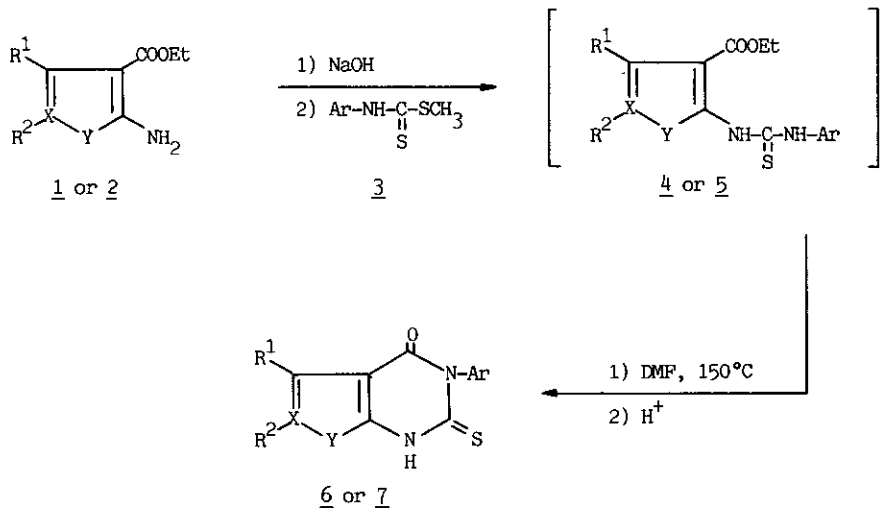
4-oxo-5,6,7,8-tetrahydro[1]benzothieno[2,3-d][1,3]oxazines 15 are, to the best of our knowledge, still unknown.

As can be seen, a large number of structures of potentially active compounds, derived either from benzo[b]thiophene[5] or from pyrazole -including analogues of the xanthine oxidase inhibitor oxyallopurinol[6]- can be easily prepared in one step.

For the sake of simplicity, the present paper has been arranged according to the most outstanding features of the newly-formed ring and the nomenclature of the final products is based on this criterion.

4-Oxo-2-thioxopyrimidines

Compounds 6 and 7 are easily obtained as shown in Scheme I:



For 1, 4 and 6: X=N, Y=N-C_6H_5, R^1=H

2, 5 and 7: X=C, Y=S, R^1-R^2=$(CH_2)_4$

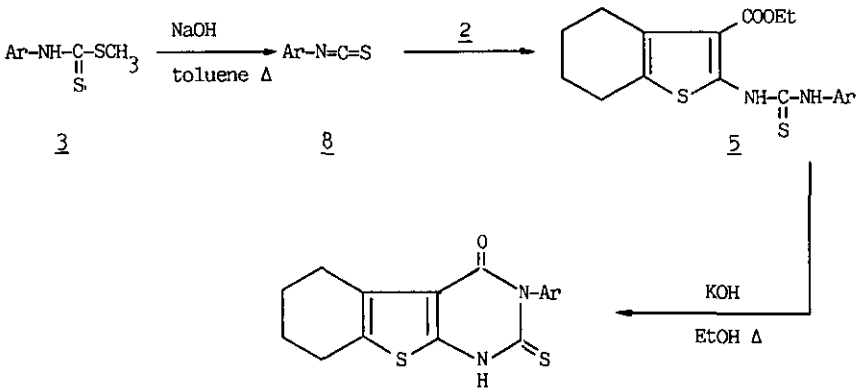
Ar: see tables

- Scheme I -

Compounds 6, 5-aryl-4-oxo-1-phenyl-6-thioxo-1H-4,5,6,7-tetrahydropyrazolo [3,4-d] pyrimidines, were prepared from 5-amino-4-ethoxycarbonyl-1-phenylpyrazole[7] 1 and the corresponding methyl N-aryldithiocarbamate 3. A basic medium is necessary in order to achieve greater reactivity of the o-aminoester and unless sodium hydroxide

is used, extensive decomposition occurs and yields are very low.

Compounds 7, 3-aryl-4-oxo-2-thioxo-1,2,3,4,5,6,7,8-octahydro[1]benzothieno[2,3-d] pyrimidines, are similarly obtained by refluxing the mixture for 7 to 10 h. With longer reaction times several by-products are formed, perhaps due to the rearrangement that 2-aminothiophenes undergo in a basic medium[8], especially when electron-withdrawing groups are attached to the thiophene ring, as is the case with 2. The structure of compounds 7 was confirmed by an independent synthesis in which 3 were also used as starting products. These were converted[9] to the corresponding arylisothiocyanates, which then reacted with 2 to give the thioureas 5. These were isolated and cyclized using ethanolic potassium hydroxide[10,11]. (Scheme II).



3 8 5 7

- Scheme II -

The main spectral features of compounds 6 are summarized below.

Ir (Nujol) and ^{1}H-nmr spectra of compounds 6

Compound	Ar	ν(cm^{-1}) NH	ν(cm^{-1}) C=O	δ(ppm); solvent CF_3-COOH H-3	Aromatics	Me
6a	C_6H_5	3120	1725	8.45	7.55-7.10(m,10H)	—
6b	4-Me-C_6H_4	3120	1705	8.50	7.6(s,5H);7.5-7.0(m,4H)	2.35(s,3H)
6c	4-MeO-C_6H_4	3120	1725	8.50	7.6(s,5H);7.2(s,4H)	4.0(s,3H)
6d	4-Cl-C_6H_4	3200-3120	1725	8.50	7.7-7.1(m,9H)	—
6e	2,4-di Me-C_6H_3	3350	1725	8.50	7.6(s,5H); 7.3-7.0(m,3H)	2.4(s,3H); 2.1(s,3H)
6f	3,5-di Me-C_6H_3	3120	1725	8.50	7.6(s,5H);7.3-6.8(m,3H)	2.35(s,6H)

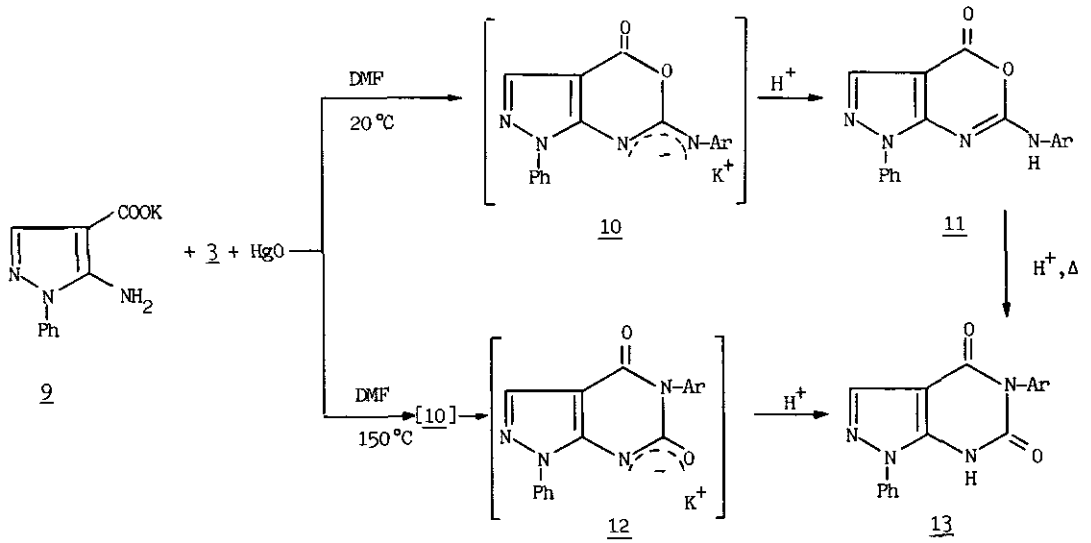
Compounds 7 show medium absorptions in the 3140-3100 cm^{-1} range (νN-H) and strong absorptions around 1230 cm^{-1}, attributable to ν(C=S). In their nmr spectra, the tetramethylene bridge shows its characteristic multiplets at δ 3.1-2.7 ppm (H-5 and H-8,4H) and δ 2.1-1.8 ppm(H-6 and H-7,4H). Additional data with regard to these compounds are shown in the table.

Ir (Nujol) and ^{1}H-nmr spectra of compounds 7

			δppm; solvent CF_3COOH	
Compound	Ar	ν(C=O)cm^{-1}	Ar	Me
7a	C_6H_5	1710	7.8-7.3(m,5H)	--
7b	4-Me-C_6H_4	1715	7.6-7.2(m,4H)	2.5(s,3H)
7c	4-MeO-C_6H_4	1715	7.4-7.0(m,4H)	4.0(s,3H)
7d	4-Cl-C_6H_4	1720	7.7-7.2(m,4H)	--

2,4- Dioxopyrimidines and 2-arylamino-1,3-oxazines

The synthesis of the pyrazole derivatives 11, 6-arylamino-4-oxo-1-phenyl-1H,4H-pyrazolo[3,4-d][1,3]oxazines, and 13, 5-aryl-4,6-dioxo-1-phenyl-4,5,6,7-tetrahydro-1H-pyrazolo[3,4-d]pyrimidines is easily carried out by the reaction of the potassium salt 9 with 3 in the presence of red mercury (II) oxide in dimethylformamide. When the mixture is maintained at room temperature the kinetic isomers 11 are formed after acidification (Scheme III).



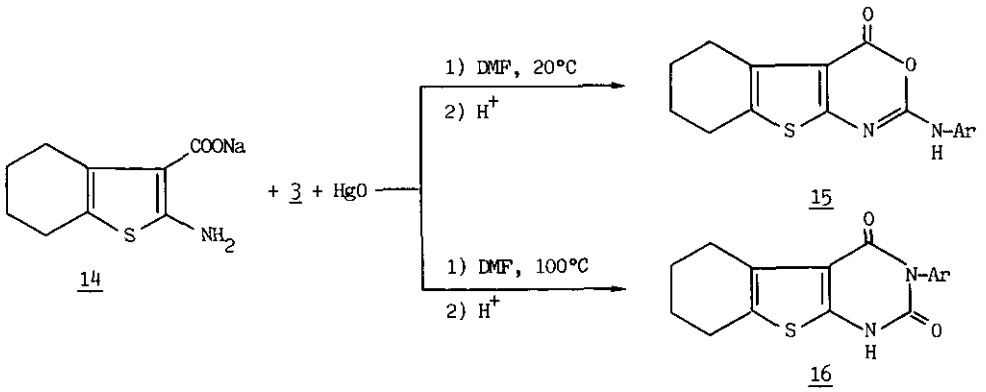
- Scheme III -

The reaction seems to proceed via the thiourea 4, which is desulfurated to the corresponding unstable carbodiimide, which is cyclized to the oxazine anion 10. Acidic treatment affords 11 in moderate to good yields. When the reaction is conducted at reflux temperature, anion 10 in all probability rearranges to the thermodynamically more stable anion 12 which, after acidic work-up, gives 13. Nevertheless, yields are only moderate and an alternative procedure for the synthesis of 13 was devised, namely, direct isomerization of compounds 11 by simply heating in acidic medium. Of all the acids used sulfuric acid turned out to give the best results. The ^{1}H-nmr spectra of compounds 11 and 13 (in CF_3-COOH) are quite similar to those of 6 and are not gathered here. The most outstanding difference between 11 and 13 lies in their ν(C=O) absorptions, with compounds 11 showing a strong band at a higher wavenumber (lactone type) than 13 (lactam and urea types).

Ir (Nujol) carbonyl absorptions of isomers 11 and 13

Ar	Compound	ν(C=O)cm^{-1}	Compound	ν(C_4=O)cm^{-1}	ν(C_6=O)
C_6H_5	11a	1760	13a	1735	1670
4-Me-C_6H_4	11b	1770	13b	1735	1670
4-MeO-C_6H_4	11c	1770	13c	1740	1670
4-Cl-C_6H_4	11d	1780	13d	1740	1660
2,4-diMe-C_6H_3	11e	1780	13e	1740	1660
3,5-diMe-C_6H_3	11f	1760	13f	1740	1675

Depending on reaction conditions the reaction of 3 with sodium salt 14 in the presence of red mercury (II) oxide gives 2-arylamino-4H-4-oxo-5,6,7,8-tetrahydro-[1]benzothieno[2,3-d][1,3]oxazines 15 or 3-aryl-2,4-dioxo-1,2,3,4,5,6,7,8-octahydro-[1]benzothieno[2,3-d]pyrimidines 16. (Scheme IV).



- Scheme IV -

In this case sodium salt was used since it gave better results than the corresponding potassium salt.

The ^{1}H-nmr spectra of compounds 15 and 16 (in CF_3-COOH) are very similar to those of 7, the most significant difference being in their ir spectra, as shown below.

Ir (Nujol) carbonyl absorptions of isomers 15 and 16

Ar	Compound	ν(C=O)cm^{-1}	Compound	ν(C$_4$=O)cm^{-1}	ν(C$_2$=O)
C_6H_5	15a	1745	16a	1730	1665
4-Me-C_6H_4	15b	1745	16b	1725	1665
4-MeO-C_6H_4	15c	1745	16c*	--	--
4-Cl-C_6H_4	15d	1745	16d	1730	1665

• Compound 16c could not be isolated.

To sum up, the main advantage of our procedure is that not only pyrimidinediones 13 and 16 can be prepared in one step -a typical reaction with isocyanates[12,13]- but also that their previously unreported isomers 11 and 15 can be synthesized under smooth conditions. This is of great interest since fused 2-amino-1,3-oxazines are quite elusive compounds[14], mainly due to the fact that reaction conditions tend to favour their thermodynamic isomers.

EXPERIMENTAL

Melting points were determined using a Büchi 510 apparatus and are uncorrected. Ir spectra were recorded on a Perkin-Elmer 283 instrument. ^{1}H-Nmr spectra were obtained on a Bruker WP-80-CW spectrometer with TMS as internal reference.

5-Aryl-4-oxo-1-phenyl-6-thioxo-1H-4,5,6,7-tetrahydropyrazolo[3,4-d]pyrimidines 6

(General Procedure)

To a solution of 5-amino-4-ethoxycarbonyl-1-phenylpyrazole[7] 1 (1.155 g, 5 mmol) in DMF (10 ml), aqueous 20 M sodium hydroxide (1 ml) was added. The mixture was stirred for 30 min and then the corresponding methyl N-aryldithiocarbamate[3] 3 (5 mmol) in DMF (10 ml) was added dropwise. Once all 3 had been added, the mixture was refluxed for 6-8 h and then cooled. The white precipitate (sodium formiate) was filtered off and the filtrate poured into water (200 ml) and acidified with concentrated hydrochloric acid (until pH=5) in an ice-water bath. The solid thus obtained was filtered off, dried, washed twice with ether and recrystallized.

6a: Yield 54%. Mp 193-195 °C(EtOH/H_2O). Anal. Calcd. for $C_{17}H_{12}N_4OS$ (320.37): C, 63.73%; H, 3.78; N, 17.49. Found: C, 63.92; H, 3.64; N, 17.69.

6b: Yield 65%. Mp 198-200 °C (EtOH/H_2O). Anal. Calcd. for $C_{18}H_{14}N_4OS$ (334.40): C, 64.65%; H, 4.22; N, 16.76. Found: C, 64.45; H, 4.08; N,16.95.

6c: Yield 50%. Mp 235-237 °C (EtOH/H_2O). Anal. Calcd. for $C_{18}H_{14}N_4O_2S$ (350.40): C, 61,70%; H, 4.03; N, 15,99. Found: C, 61.88; H, 3.89; N, 16.16.

6d: Yield 58%. Mp 230-232 °C(EtOH). Anal. Calcd. for $C_{17}H_{11}ClN_4OS$ (354.82): C, 57.54%; H, 3.13; N, 15.79. Found: C, 57.38; H, 3.30; N, 15.68.

6e: Yield 60%. Mp 123-125 °C (EtOH/H_2O). Anal. Calcd. for $C_{19}H_{16}N_4OS$ (348.42): C, 65.49%; H, 4.63; N, 16.08. Found: C, 65.55; H, 4.70; N, 16.00.

6f: Yield 55%. Mp 131-133 °C (EtOH /H_2O). Anal. Calcd. for $C_{19}H_{16}N_4OS$ (348.42): C, 65.49%; H, 4.63; N, 16.08. Found: C, 65.46; H, 4.69; N, 16.10.

N-(3-Ethoxycarbonyl-4,5,6,7-tetrahydrobenzo[b]thien-2-yl)-N'-arylthioureas 5

(General Procedure)

A mixture of 2-amino-3-ethoxycarbonyl-4,5,6,7-tetrahydrobenzo[b]thiophene 2 (1.125 g, 5 mmol) and the corresponding arylisothiocyanate[9] 8 (5 mmol) in ethanol (5 ml) was refluxed for 8-10 h and then cooled. The resulting precipitate was filtered off, washed with ethanol and dried.

5a: Yield 85%. Mp 182-183 °C. Anal. Calcd. for $C_{18}H_{20}N_2O_2S_2$ (360.49): C, 59.97%; H, 5.59; N, 7.77. Found: C, 60.06; H, 5.66; N, 7,69.

5b: Yield 75%. Mp 160-161 °C. Anal. Calcd. for $C_{19}H_{22}N_2O_2S_2$(374.51): C, 60.93%; H, 5.92; N, 7.48. Found: C, 60.99; H, 5.98; N, 7.39.

5c: Yield 80%. Mp 174-176 °C. Anal. Calcd. for $C_{19}H_{22}N_2O_3S_2$ (390.51): C, 58.43%; H, 5.68; N, 7.18. Found: C, 58.38; H, 5.72; N, 7.15.

5d: Yield 78%. Mp 205-207 °C. Anal. Calcd. for $C_{18}H_{19}ClN_2O_2S_2$(394.93): C, 54.74%; H, 4.85; N, 7.09. Found: C, 54.70; H, 4.81; N, 7.13.

3-Aryl-4-oxo-2-thioxo-1,2,3,4,5,6,7,8-octahydro[1]benzothieno[2,3-d]pyrimidines 7

(General Procedure)

METHOD A: Potassium hydroxide (0.28 g, 5 mmol) in ethanol (25 ml) was added to a solution of the corresponding thiourea 5 (5 mmol) in ethanol (25 ml). The mixture was refluxed for 2 h, cooled, poured into water (500 ml) -in an ice-water bath- and neutralized to litmus with concentrated hydrochloric acid. The white solid was filtered off, washed first with water then with ether and dried.

METHOD B: Aqueous 10M sodium hydroxide (0.5 ml) was added to a solution of 2-amino-3-ethoxycarbonyl-4,5,6,7-tetrahydrobenzo[b]thiophene 2 (1.125 g, 5 mmol) in DMF (10 ml). The mixture was stirred for 30 min and then the corresponding methyl N-aryldithiocarbamate 3 (5 mmol) in DMF (10 ml) was added dropwise. The mixture

was refluxed for 7-10 h, cooled, filtered and the filtrate was poured into cold water (200 ml) and acidified with concentrated hydrochloric acid (until pH=4-5). The precipitate was filtered off, washed with water, dried and recrystallized.

7a: Yield 97%(A), 65%(B). Mp 284-286 °C(CH_3CN). Anal. Calcd. for $C_{16}H_{14}N_2OS_2$ (314.42): C, 61.12%; H, 4.49; N, 8.91. Found: C, 61.07; H, 4.54; N, 8.99.

7b: Yield 93%(A), 75%(B). Mp 276-278 °C(CH_3CN). Anal. Calcd. for $C_{17}H_{16}N_2OS_2$ (328.45): C, 62.16%; H, 4.91; N, 8.53. Found: C, 62.19; H, 4.87; N, 8.59.

7c: Yield 98%(A), 74%(B). Mp 278-280 °C(CH_3CN). Anal. Calcd. for $C_{17}H_{16}N_2O_2S_2$ (344.45): C, 59.27%; H, 4.68; N, 8.13. Found: C, 59.33; H, 4.61; N, 8.09.

7d: Yield 93%(A), 80%(B). Mp 286-288 °C(CH_3CN). Anal. Calcd. for $C_{16}H_{13}ClN_2OS_2$ (348.87): C, 55.08%; H, 3.76; N, 8.03. Found: C, 55.12; H, 3.74; N, 8.07.

6-Arylamino-4-oxo-1-phenyl-1H,4H-pyrazolo[3,4-d][1,3]oxazines 11 (General Procedure)

To a suspension of potassium 5-amino-1-phenylpyrazole-4-carboxylate 9 (0.723 g, 3 mmol) and red mercury (II) oxide (0.756 g, 3.5 mmol) in DMF (10 ml), a solution of the corresponding methyl N-aryldithiocarbamate 3 (3 mmol) in DMF (10 ml) was added dropwise. The mixture was stirred for 15 h at room temperature and then filtered. The filtrate was poured into water (200 ml) -ice-water bath- and acidified with concentrated hydrochloric acid (until pH=5). The crude solid was filtered off, dried, washed with ether and recrystallized from acetonitrile.

11a: Yield 65%. Mp 220-222 °C. Anal. Calcd. for $C_{17}H_{12}N_4O_2$ (304.30): C, 67.09%; H, 3.98; N, 18.41. Found: C, 67.13; H, 4.01; N, 18.36.

11b: Yield 65%. Mp 222-224 °C. Anal. Calcd. for $C_{18}H_{14}N_4O_2$ (318.32): C, 67.91%; H, 4.43; N, 17.60. Found: C, 67.95; H, 4.40; N, 17.56.

11c: Yield 64%. Mp 209-211 °C. Anal. Calcd. for $C_{18}H_{14}N_4O_3$ (334.32): C, 64.66%; H, 4.22; N, 16.76. Found: C, 64.70; H, 4.28; N, 16.71.

11d: Yield 55%. Mp 227-229 °C. Anal. Calcd. for $C_{17}H_{11}ClN_4O_2$ (338.76): C, 60.27%; H, 3.27; N, 16,54.Found: C, 60.31; H, 3.33; N, 16.48.

11e: Yield 34%. Mp 165-167 °C. Anal. Calcd. for $C_{19}H_{16}N_4O_2$ (332.35): C, 68.66%; H, 4.85; N, 16.86. Found: C, 68.60; H, 4.88; N, 16.93.

11f: Yield 65%. Mp 209-211 °C. Anal. Calcd. for $C_{19}H_{16}N_4O_2$ (332.35): C, 68.66%; H, 4.85; N, 16.86. Found: C, 68.71; H, 4.83; N, 16.90.

5-Aryl-4,6-dioxo-1-phenyl-4,5,6,7-tetrahydro-1H-pyrazolo[3,4-d]pyrimidines 13

(General Procedure)

METHOD A: This method is identical to the one described for the synthesis of compounds 11, except that after the addition of 3 the reaction mixture was heated

under reflux for 4 h.

METHOD B: A solution of the corresponding compound 11 (2 mmol) in DMF (20 ml) containing four drops of concentrated sulfuric acid was heated at 100 °C for 2-3 h, then cooled and poured into water (100 ml). The mixture was stirred for 30 min and the precipitate was filtered off, dried, washed with ether and recrystallized.

13a: Yield 25%(A), 60%(B). Mp 225-227 °C(n-butanol). Anal. Calcd. for $C_{17}H_{12}N_4O_2$ (304.30): C, 67.09%; H, 3.98; N, 18.41. Found: C, 67.02; H, 3.92; N, 18.39.

13b: Yield 24%(A), 52%(B). Mp 275-277 °C (n-butanol). Anal. Calcd. for $C_{18}H_{14}N_4O_2$ (318.32): C, 67.91%; H, 4.43; N, 17.60. Found: C, 67.87; H, 4.48; N, 17.55.

13c: Yield 54%(A), 67%(B). Mp 208-210 °C. Anal. Calcd. for $C_{18}H_{14}N_4O_3$(334.32): C, 64.66%; H, 4.22; N, 16.76. Found: C, 64.60; H, 4.29; N, 16.80.

13d: Yield 49%(A), 75%(B). Mp 256-258 °C(n-butanol). Anal. Calcd. for $C_{17}H_{11}ClN_4O_2$ (338.74): C, 60.27%; H, 3.27; N, 16.54. Found: C, 60.23; H, 3.24; N, 16.59.

13e: Yield 30%(A), 51%(B). Mp 230-232 °C. Anal. Calcd. for $C_{19}H_{16}N_4O_2$ (332.35): C, 68.66%; H, 4.85; N, 16.86. Found: C, 68.59; H, 4.90; N, 16.83.

13f: Yield 31%(A), 60%(B). Mp 263-265 °C(n-butanol). Anal. Calcd. for $C_{19}H_{16}N_4O_2$ (332.35): C, 68.66%; H, 4.85; N, 16.86. Found: C, 68.62; H, 4.79; N, 16.88.

2-Arylamino-4H,4-oxo-5,6,7,8-tetrahydro[1]benzothieno[2,3-d][1,3]oxazines 15 (General Procedure)

To a suspension of sodium 2-amino-4,5,6,7-tetrahydrobenzo[b]thiophene-3-carboxylate 14 (1.095 g, 5 mmol) and red mercury (II) oxide (1.296 g, 6 mmol) in DMF (20 ml), a solution of the corresponding methyl N-aryldithiocarbamate 3 (5 mmol) in DMF (20 ml) was added dropwise. The mixture was then stirred for 20-24 h at room temperature and filtered. The filtrate was poured into cold water (400 ml) and acidified until pH=5 with concentrated hydrochloric acid. The resulting precipitate was filtered off, dried and recrystallized from n-butanol.

15a: Yield 72%. Mp 218-220 °C. Anal. Calcd. for $C_{16}H_{14}N_2O_2S$ (298.37): C, 64.41%; H, 4.73; N, 9.39. Found: C, 64.45; H, 4.79; N, 9.31.

15b: Yield 70%. Mp 235-237 °C. Anal. Calcd. for $C_{17}H_{16}N_2O_2S$ (312.39): C, 65.36%; H, 5.16; N, 8.97. Found: C, 65.40; H, 5.11; N, 8.93.

15c: Yield 75%. Mp 226-228 °C. Anal. Calcd. for $C_{17}H_{16}N_2O_3S$ (328.39): C, 62.18%; H, 4.91; N, 8.53. Found: C, 62.10; H, 4.98; N, 8.48.

15d: Yield 81%. Mp 243-245 °C. Anal. Calcd. for $C_{16}H_{13}ClN_2O_2S$ (332.81): C, 57.74%; H, 3.94; N, 8.42. Found: C, 57.69; H, 3.86; N, 8.50.

3-Aryl-2,4-dioxo-1,2,3,4,5,6,7,8-octahydro[1]benzothieno[2,3-d]pyrimidines 16

(General Procedure)

The procedure is identical with the one described for the synthesis of compounds 15, except that after the addition of 3 the reaction mixture was heated at 100°C for 8 h.

16a: Yield 70%. Mp 278-280 °C. Anal. Calcd. for $C_{16}H_{14}N_2O_2S$ (298.39): C, 64.41%; H, 4.73; N, 9.39. Found: C, 64.48; H, 4.79; N, 9.35.

16b: Yield 74%. Mp 284-286 °C. Anal. Calcd. for $C_{17}H_{16}N_2O_2S$ (312.38): C, 65.36%; H, 5.16; N, 8.97. Found: C, 65.29; H, 5.19; N, 8.94.

16d: Yield 70%. Mp 272-274 °C. Anal. Calcd. for $C_{16}H_{13}ClN_2O_2S$ (332.81): C, 57.74%; H, 3.94; N, 8.42. Found: C, 57.79; H, 3.99; N, 8.46.

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