SYNTHESIS OF NEW THIAZOLO [3, 2-*a*: 4, 5-*d*] DIPYRIMIDINE DERIVATIVES

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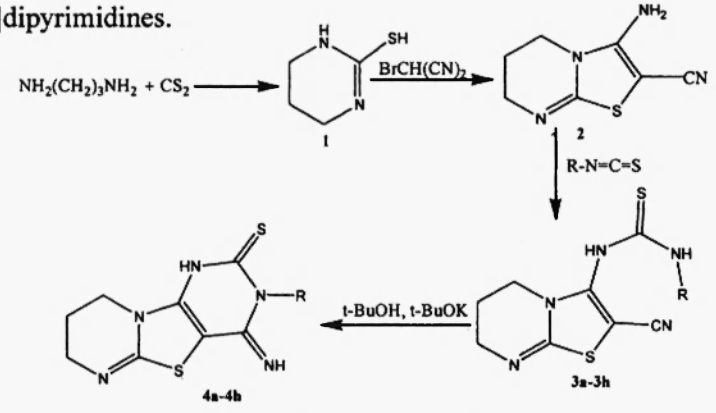
Abstract : Substituted thiazolo [3, 2-*a*] pyrimidines were successfully converted to their corresponding dipyrimidothiazoles by sequential treatment with various isothiocyanates and potassium *t*-butoxide in *t*-butyl alcohol.

Introduction

Interest in thiazolo[3, 2-*a*] pyrimidines and their fused derivatives arises from several reports on their diverse biological activities (1,2). These compounds have been described as being antimicrobial(3), anti-inflammatory (4) and enzyme inhibitor (1) agents and have found to be useful as insecticides (5). The most general approach for the synthesis of thiazolo[3, 2-*a*] pyrimidines involves annulation of a pyrimidine ring onto the preformed thiazole ring. This route involves reactions of 2- amino- thiazole with suitable bifunctional reagents (6). Annulation of a thiazole ring onto the preformed pyrimidine ring can be reached through heterocyclization of pyrimidine-2-thiole with bifunctional reagents (7-11). This procedure has the drawback of giving both regioisomers thiazolo[3, 2-*a*] and thiazolo [2, 3-*b*] pyrimidines (12). Recently we reported regioselective synthesis of 3-benzyl thiazolo [3, 2-*a*] pyrimidines through palladium catalyzed heteroannulation of acetylenic compounds (13). In continuation of our studies directed towards the synthesis of fused heterocycles (14), we had occasion to examine the heterocyclization reaction of 1, 4, 5, 6-tetrahydro-pyrimidinethiol with bromomalonitrile and isothiocyanates which is a less explored approach to the synthesis of thiazolo[3,2-a] pyrimidines and thiazolo[3, 2-*a*:4, 5-*d*]dipyrimidines.



a:R=Ph, **b**:R=3-CH_3-C_6H_4, **c**:R=4-CH_3-C_6H_4 , **d**:R=4-F-C_6H_4 , **e**:R=3-Cl-C_6H_4 , **f**:R=2-Br-C_6H_4 , **g**:R=3-Br-C_6H_4 , **h**:R=4-Br-C_6H_4

Results and Discussion

A. Preparation of thiazolo [3, 2-*a*] pyrimidines. The 1, 4, 5, 6-tetrahydro 2-pyrimidinethiol **1** which has been reported previously (15), was prepared in our laboratory from a simple procedure in >90% yield. Compound **1** reacts with bromomalonitrile in ethanol in the presence of sodium hydroxide at room temperature to give the thiazolo[3,2-a] pyrimidine **2** in 45% yield (Scheme). The structure **2** is derived from analytical and spectral data (IR, ^{1}HNMR and MS) as summarized in the experimental section. The IR spectrum of compound **2** exhibited characteristic absorption bands for NH_2 and CN groups at 3400-3300 and 2220 cm^{-1} respectively. The ^{1}HNMR spectrum recorded in deuterated dimethyl sulfoxide revealed three signals in the regions δ 1.8(quin), 3.7(t) and 3.9(t) ppm attributed to the three methylene moieties and one signal in the region δ 7.8(s) ppm which was removed on deuteration indicative of an NH_2 group in the molecule. The molecular ion of thiazolopyrimidine **2** was observed at m/z 180 corresponding to the molecular formula $C_7H_8N_4S$ (experimental section).

B. Preparation of thiazolodipyrimidines. Phenylthiourea derivatives **3**(a-h) of the foregoing compound **2**, readily available from thiazolopyrimidine **2** by treatment with appropriate isothiocyanates in dry acetonitrile in the presence of triethylamine at reflux temperature to give the corresponding pyrimidothiazolyl –N- phenyl thiourea derivatives **3**(a-h) as potential intermediates for the next step synthesis. When compounds **3**(a-h) were heated in *t*-butanol/ potassium *t*-butoxide at reflux temperature underwent ring closure to give thiazolodipyrimidines **4**(a-h) in moderate yields. The structures assigned to these compounds were substantiated by their spectral data (experimental section). For example the ^{1}HNMR spectrum of compound **4a** was devoid of the signal at δ 7.0-7.2 for two NH group of the precursor **3** but showed a signal for NH at 8.1 ppm and a downfield signal due to imine proton at 12.1-12.3 ppm indicating the construction of the pyrimidine ring around thiazolo nucleus.

Experimental

The melting points were recorded on an Electrothermal type 9100 melting point apparatus. The IR spectra were obtained on a 4300 Shimadzu Spectrometer for KBr pellets. The ^{1}HNMR (100 MHz) spectra were recorded on a Bruker AC 100 spectrometer using TMS as internal standard. The mass spectra were scanned on a Varian Mat CH-7 instrument at 70 eV. Elemental analysis was obtained on a Thermo Finnigan Flash EA microanalyzer.

1, 4, 5, 6–Tetrahydro- 2- pyrimidinethiol 1.

This compound was prepared from a mixture of 1,3-propanediamine according to a published procedure.[11] m.p. 211-213 °C; yield (90%); MS: m/z (%) 116(M^+, 5.5), 114 (100); IR: 3300 (NH) cm^{-1}; ^{1}HNMR δ 1.7 (quin, 2H,5- CH_2), 2.1 (s, 1H, SH), 3.1 (m, 4H,4,6- $2CH_2$), 7.8 (s, 1H, NH). *Anal.* Calcd for $C_4H_8N_2S$: C, 41.35; H, 6.94; N, 24.11; S 27.59. Found; C, 41.15; H, 6.81; N, 24.38; S, 27.34.

3- Amino- 6, 7- dihydro- 5*H*- thiazolo[3, 2-*a*] pyrimidine- 2- carbonitrile 2.

To a solution of **1** (0.1 mol), in ethanol (80 ml), bromomalononitrile (0.1mol) was added gradually. The reaction mixture was stirred at room temperature for 1 h. After that potassium hydroxide (0.1 mol) was added, and again the mixture was stirred for 1 h. The precipitated product was worked up by filtration and recrystalyzation from the ethanol. M.P. 226-227 °C; yield (45%); MS: m/z (%) 180(M^+, 2.1), 176(100); IR: 3400, 3300 (NH_2), 2220 (CN) cm^{-1}; ^{1}HNMR : δ 1.8 (m, 2H,6- CH_2), 3.7 (t, j = 6.2, 2H,7- CH_2), 3.9 (t, j = 6.2, 2H,5-CH_2), 7.8 (s, 2H, NH_2). *Anal.* Calcd for $C_7H_8N_4S$: C, 46.65; H, 4.47; N, 31.09; S 17.79. Found; C, 46.53; H, 4.66; N, 31.05; S, 17.82.

General procedure for preparation of N-(2-cyano- 6, 7- dihydro- 5H- thiazolo[3, 2-*a*] pyrimidine -3-yl)-N- phenyl thiourea derevitives 3(a-h)

To a solution of **2** (5.5 mmol, 1g), in dry MeCN (30 mL), Et_3N (7 mmol), each of the appropriate isothiocyanates (5.8 mmol) was added and refluxed for 12h. The mixture was cooled to r.t, and the precipitated product was collected by filtration and washed with EtOH. The yields were changed between 45-55%.

N-(2-cyano-6,7-dihydro-5*H*- thiazolo[3, 2-*a*] pyrimidine -3yl)-N-phenylthiourea 3a.

M.P. 341-343 °C; yield (55%); MS: m/z (%) 315 (M^+, 5.26), 313 (66.66), 312 (100); IR : 3420, 3210, 2225 cm^{-1}; ^{1}HNMR : δ 1.8 (m, 2H,6-CH_2), 3.5 (t, 2H, j = 6.2, 7-CH_2), 3.8 (t, 2H, j = 6.1, 5-CH_2), 6.9 (S, 2H, 2NH), 7.1-7.5 (m, 5H Ar). *Anal.* Calcd for $C_{14}H_{13}N_5S_2$: C, 53.31; H, 4.15; N, 22.20; S 20.33. Found; C, 53.45; H, 4.03; N, 21.98; S, 19.98.

N-(2-cyano-6, 7-dihydro-5*H*- thiazolo[3, 2-*a*] pyrimidine -3yl)-N-(3-methylphenyl) thiourea 3b.

M.P. 322-323 °C; yield (57%); MS: m/z (%) 329 (M^+, 5.13), 327 (28.05), 326(100); IR : 3400, 3240, 2300 cm^{-1}; ^{1}HNMR δ: 1.8 (m, 2H,6-CH_2), 2.3 (s, 3H, CH_3), 3.7 (t, 2H, j = 6.2, 7-CH_2), 4.1 (t, 2H, j= 6.2, 5-CH_2), 7 (S, 2H, 2NH), 7.2-7.5 (m, 4H Ar). *Anal.* Calcd for $C_{15}H_{15}N_5S_2$: C, 54.69; H, 4.59; N, 21.26; S 19.46. Found; C, 54.75; H, 4.65; N, 21.00; S, 19.38.

N-(2-cyano-6, 7-dihydro-5*H*- thiazolo[3, 2-*a*] pyrimidine -3yl)-N-(4-methylphenyl) thiourea 3c.

M.P. 329-330 °C; yield (49%); MS: m/z (%) 329 (M^+, 13.04), 326 (100); IR: 3400, 3245, 2270 cm^{-1}; ^{1}HNMR δ: 1.8 (m, 2H,6-CH_2), 2.4 (s, 3H, CH_3), 3.5 (t, 2H, j= 6.2, 7-CH_2), 3.8 (t, 2H, j= 6.2, 5-CH_2), 6.9 (S, 2H, 2NH), 7.1 (d, 2H, j= 8.3), 7.4 (d, 2H, j= 8.3). *Anal.* Calcd for $C_{15}H_{15}N_5S_2$: C, 54.69; H, 4.59; N, 21.26; S 19.46. Found; C, 54.75; H, 4.25; N, 21.28; S, 19.13.

N-(2-cyano-6, 7-dihydro-5*H*- thiazolo[3, 2-*a*] pyrimidine -3yl)-N-(4-fluorophenyl) thiourea 3d.

M.P.326-327 °C; yield (50%); MS: m/z (%) 333 (M^+, 10.42), 330 (100); IR : 3400, 3240, 2280 cm^{-1} ; ^{1}HNMR δ: 2.8 (m, 2H,6-CH_2), 3.4 (t, 2H, j= 6.2, 7-CH_2), 3.8 (t, 2H, j= 6.2, 5-CH_2), 7.1 (S, 2H, 2NH),

7.2-7.5 (t, 4H Ar). *Anal.* Calcd for $C_{14}H_{12}FN_5S_2$: C, 50.44; H, 3.63; N, 21.01; S 19.23. Found; C, 50.41; H, 3.48; N, 21.40; S, 19.17.

N-(3-chlorophenyl)-N-(2-cyano-6, 7-dihydro-5*H*- thiazolo[3, 2-*a*] pyrimidine -3yl) thiourea 3e.

M.P. 330-332 °C; yield (45%); MS: m/**z** (%) 351 (47), 349 (M^+, 95), 348 (100); IR : 3400, 3240, 2340 cm^{-1}; ^{1}HNMR δ: 1.8 (m, 2H,6-CH_2), 3.6 (t, 2H, j=7.0, 7-CH_2), 3.8 (t, 2H, j= 7.0, 5-CH_2), 7.1 (S, 2H, 2NH), 7.2-7.6 (m, 4H Ar). *Anal.* Calcd for $C_{14}H_{12}ClN_5S_2$: C, 48.06; H, 3.46; N, 20.02; S 18.33. Found; C, 48.30; H, 3.19; N, 20.12; S, 17.99.

N-(2-bromophenyl)-N-(2-cyano-6, 7-dihydro-5*H*- thiazolo[3, 2-*a*] pyrimidine -3yl) thiourea 3f.

M.P. 266-268 °C; yield (46%); MS: m/z (%) 395(5.55), 393 (M^+, 5.85), 310 (100); IR : 3400, 3235, 2280cm^{-1}; ^{1}HNMR δ: 1.8 (m, 2H,6-CH_2), 3.6 (t, 2H, j= 6.7, 7-CH_2), 3.8 (t, 2H, j= 6.7, 5-CH_2), 7.1 (S, 2H, 2NH), 7.2-7.8 (m, 4H, Ar). *Anal.* Calcd for $C_{14}H_{12}BrN_5S_2$: C, 42.65; H, 3.07; N, 17.76; S 16.26. Found; C, 42.86; H, 3.25; N, 17.40; S, 15.98.

N-(3-bromophenyl)-N-(2-cyano-6, 7-dihydro-5*H*- thiazolo[3, 2-*a*] pyrimidine -3yl) thiourea 3g.

M.P. 320-323 °C; yield (57%); MS: m/z (%) 395 (6.99), 393 (M^+, 7.00), 391 (100); IR : 3420, 3240, 2300 cm^{-1}; ^{1}HNMR δ: 1.8 (m, 2H,6-CH_2), 3.4 (t, 2H, j= 6.8, 7-CH_2), 3.8 (t, 2H, j= 6.9, 5-CH_2), 7.1 (S, 2H, 2NH), 7.2-7.7 (m, 4H Ar). *Anal.* Calcd for $C_{14}H_{12}BrN_5S_2$: C, 42.65; H, 3.07; N, 17.76; S 16.26. Found; C, 42.68; H, 3.18; N, 17.62; S, 16.09.

N-(4-bromophenyl)-N-(2-cyano-6, 7-dihydro-5*H*- thiazolo[3, 2-*a*] pyrimidine -3yl) thiourea 3h.

M.P. 279-281 °C; yield (49%); MS: m/z (%) 395 (99.89), 393 (M^+, 100); IR : 3400, 3240, 2280 cm^{-1}; ^{1}HNMR δ: 1.8 (m, 2H,6-CH_2), 3.6 (t, 2H, j= 6.2, 7-CH_2), 3.8 (t, 2H, j= 6.3, 5-CH_2), 7.1 (S, 2H, 2NH), 7.2 (d, 2H, j= 8.3), 7.7 (d, 2H, j= 8.3). *Anal.* Calcd for $C_{14}H_{12}BrN_5S_2$: C, 42.65; H, 3.07; N, 17.76; S 16.26. Found; C, 42.30; H, 3.25; N, 18.00; S, 15.96.

General procedure for the preparation of thiazolodipyrimidine deravative 4(a-h).

First, potassium (2 mmol) was dissolved in *t*-butyl alcohol (20 ml) then 1.5 mmol of each (**3a-h**) was added to it. After stirring for 30 minute at room temperature, the mixture was refluxed for 12h. Then, it was cooled and, neutralized with aqueous HCl. The resulting precipitate was filtered and washed with ethanol and water, respectively. The solvent of recrystallization was ethanol for each of compounds.

4- Imino- 3- phenyl-2-thioxo-1, 2, 3, 4, 8, 9-hexahydro- 7*H*- thiazolo[3,2-a:4,5-*d*] dipyrimidine 4a.

M.P.297-300 °C; yield (58%); MS: m/z (%) 315(M^+, 1.77), 313(30.09), 312(100); IR: 3400 (br, NH), 3020 cm^{-1}; ^{1}HNMR δ: δ 2.1 (m, 2H,8-CH_2), 3.6 (t, 2H, j= 7.1, 7-CH_2), 4 (t, 2H, j= 7.1, 9-CH_2), 7.1- 7.8 (m, 5H, Ar), 8.1(br, s, 1H, NH D_2O exchangeable), 12.3 (br, s, 1H, NH D_2O exchangeable). *Anal.* Calcd for $C_{14}H_{13}N_5S_2$: C, 53.31; H, 4.15; N, 22.20; S 20.33. Found; C, 53.11; H, 4.17; N, 22.43; S, 20.08.

3-(3-Methylphenyl)- 4-imino-1, 2, 3, 4, 8, 9-hexahydro- 7*H*- thiazolo [3,2-*a*:4,5-*d*] dipyrimidine 4b. M.P. 325-327 °C; yield (53%); MS: m/z (%) 329(M^+, 4.9), 327(27.56), 326(100); IR: 3430 (br, NH), 3000 cm^{-1}; ^{1}HNMR δ: δ 2.1 (m, 2H,8-CH_2), 2.4 (s, 3H, CH_3), 3.7 (t, 2H, j= 7.1, 7-CH_2), 4.1 (t, 2H, j= 7.1, 9-CH_2), 7.- 7.5 (m, 4H, Ar), 7.9 (br, s, 1H, NH D_2O exchangeable), 12.1 (br, s, 1H, NH D_2O exchangeable). *Anal.* Calcd for $C_{15}H_{15}N_5S_2$: C, 54.69; H, 4.59; N, 21.26; S 19.46. Found; C, 54.75; H, 4.64; N, 21.24; S, 18.37.

3-(4-Methylphenyl)- 4-imino-1, 2, 3, 4, 8, 9-hexahydro- 7*H*- thiazolo[3,2-*a*:4,5-*d*] dipyrimidine 4c. M.P. 268-270 °C; yield (46%); MS: m/z (%) 329(M^+, 12.71), 327(100) ; IR: 3400(b, NH),3000 cm^{-1}; ^{1}HNMR δ: δ 2.1 (m, 2H,8-CH_2), 2.4 (s, 3H, CH_3), 3.6 (t, 2H, j= 7.1, 7-CH_2), 4.1 (t, 2H, j= 7.1, 9-CH_2), 7.1 (d, 2H, j= 9.5), 7.4 (d, 2H, j= 9.5), 8 (br, s, 1H, NH D_2O exchangeable), 12.1 (br, s, 1H, NH D_2O exchangeable).). *Anal.* Calcd for $C_{15}H_{15}N_5S_2$: C, 54.69; H, 4.59; N, 21.26; S 19.46. Found; C, 54.68; H, 4.51; N, 21.38; S,19.27.

3-(4- Florophenyl)- 4- imino-1, 2, 3, 4, 8, 9-hexahydro- 7*H*- thiazolo[3,2-*a*:4,5-*d*] dipyrimidine 4d. M.P. 286-287 °C; yield (63%); MS: m/z (%) 333 (M^+, 13.11), 330(100) ; IR: 3420(b, NH), 3010 cm^{-1}; ^{1}HNMR δ: δ 2.1 (m, 2H,8-CH_2), 3.6 (t, 2H, j= 7.1, 7-CH_2), 4.1 (t, 2H, j= 7.1, 9-CH_2), 7.2- 7.6 (m, 4H, Ar), 8.2 (br, s, 1H, NH D_2O exchangeable), 12.3 (br, s, 1H, NH D_2O exchangeable). *Anal.* Calcd for $C_{14}H_{12}FN_5S_2$: C, 50.44; H, 3.63; N, 21.01; S 19.23. Found; C, 50.46; H, 3.58; N, 21.30; S, 18.98.

3-(3-Chlorophenyl) -4-imino-1, 2, 3, 4, 8, 9-hexahydro-7*H*-thiazolo [3, 2-*a*: 4, 5-*d*]dipyrimidine 4e. M.P. 261-263 °C; yield (59%); MS: m/z (%) 351(12), 349(M^+, 26.9), 348(100); IR: 3420(b, NH),3025 cm^{-1}; ^{1}HNMR δ: δ 2.1 (m, 2H,8- CH_2), 3.6 (t, 2H, j= 7.1, 7-CH_2), 4.1 (t, 2H, j= 7.1, 9-CH_2), 7.2- 7.7 (m, 4H, Ar), 8.2 (br, s, 1H, NH D_2O exchangeable), 12.3 (br, s, 1H, NH D_2O exchangeable). *Anal.* Calcd for $C_{14}H_{12}ClN_5S_2$: C, 48.08; H, 3.46; N, 20.02; S 18.33 Found; C, 48.03; H, 3.48; N, 20.21; S, 18.11.

3-(2- Bromophenyl)- 4- imino-1, 2, 3, 4, 8, 9-hexahydro- 7*H*- thiazolo[3,2-*a*:4,5-*d*] dipyrimidine 4f. M.P. 266-268 °C; yield (48%); MS: m/z (%) 395(6.15), 393(M^+, 6.86), 391(100); IR: 3420(b, NH), 3020 cm^{-1}; ^{1}HNMR δ: δ 2.1 (m, 2H, 8-CH_2), 3.7 (t, 2H, j= 6.7, 7-CH_2), 4.1 (t, 2H, j= 6.7, 9-CH_2), 7.3- 7.9 (m, 4H, Ar), 8.3 (br, s, 1H, NH D_2O exchangeable), 12.1 (br, s, 1H, NH D_2O exchangeable). *Anal.* Calcd for $C_{14}H_{12}BrN_5S_2$: C, 42.65; H, 3.07; N, 17.76; S 16.26. Found; C, 42.53; H, 3.18; N, 17.83; S, 15.99.

3-(3- Bromophenyl) - 4- imino-1, 2, 3, 4, 8, 9-hexahydro- 7*H*- thiazolo[3,2-*a*:4,5-*d*] dipyrimidine 4g. M.P. 265-267 °C; yield (60%); MS: m/z (%) 395(26.00), 393(M, 25.89), 391(100); IR: 3420(b, NH), 3020 cm^{-1}; ^{1}HNMR δ: δ 2.1 (m, 2H,8-CH_2), 3.7 (t, 2H, j= 7.0, 7-CH_2), 4.1 (t, 2H, j= 7.0, 9CH_2), 7.2- 7.8 (m, 4H, Ar), 8.2 (br, s, 1H, NH D_2O exchangeable), 12.2 (br, s, 1H, NH D_2O exchangeable).

Anal. Calcd for $C_{14}H_{12}BrN_5S_2$: C, 42.65; H, 3.07; N, 17.76; S 16.26. Found; C, 42.74; H, 3.10; N, 17.53; S, 16.01.

3-(4- Bromophenyl) - 4- imino-1, 2, 3, 4, 8, 9-hexahydro- 7*H*- thiazolo[3,2-*a*:4,5-*d*] dipyrimidine 4h. M.P. 265-266 °C; yield (46%); MS: m/z (%) 395(93.18), 393(M^{+} 94.32); IR: 3420(b, NH), 3020 cm^{-1}; ^{1}HNMR δ: δ 2.1 (m, 2H,8-CH_2), 3.6 (t, 2H, j= 7.1, 7-CH_2), 4.1 (t, 2H, j= 7.1, 9-CH_2), 7.2 (d, 2H, j= 9.5), 7.8 (d, 2H, j= 9.5), 8.2 (br, s, 1H, NH D_2O exchangeable), 12.2 (br, s, 1H, NH D_2O exchangeable). *Anal.* Calcd for $C_{14}H_{12}BrN_5S_2$: C, 42.65; H, 3.07; N, 17.76; S 16.26. Found; C, 42.38; H, 3.21; N, 17.77; S, 16.23.

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