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# An Easy Direct Conversion of Pyridine- and Pyrimidine-Thiones into Multi-Fused Heterocyclic Compounds

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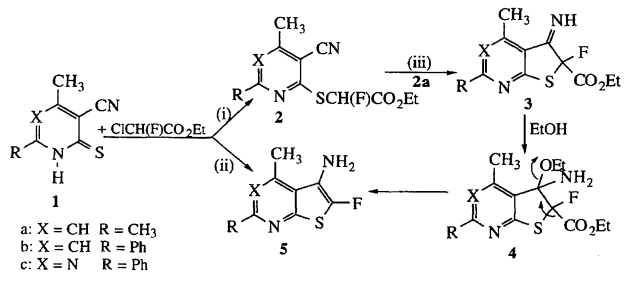
The reaction of azine-thiones with ethyl chlorofluoroacetate afforded 2-fluorothienoazines. The latter underwent self-condensation and gave multi-fused heterocyclic compounds. A wide range of unique heterocycles could be obtained on treatment of 2-fluorothienoazines with nitrogen nucleophilic reagents. The reactivity of the pyrimidine-thiones towards a variety of electrophilic reagents was studied. Chemical and spectroscopic evidence of the newly synthesized compounds are described.

Pyridine- and pyrimidine-thiones and their derivatives[1] constitute an important class of heterocyclic compounds, which are of considerable interest due to their versatility in chemical conversions such as the synthesis of inaccessible annulated heterocyclic systems,[2] and their uses as active ingredients in fungicidal and antimicrobial agents.[2] The introduction of a fluorine atom into heterocyclic rings often leads to compounds of biological and pharmaceutical importance, such as specific irreversible enzyme inhibitors or paracatalytic inactivators with high specificity.[3] Although these heterocycles have been extensively used in organic synthesis,[4,5] there is no report in the literature on their utility as reagents for building up fluoroheterocyclic compounds. This paper reports our findings on the reaction of pyridine- and pyrimidine-thiones **1a—c** with ethyl chlorofluoroacetate, in a novel synthesis of fluorothienoazines, which dimerized into multi-fused heterocyclic compounds in a one-pot reaction.

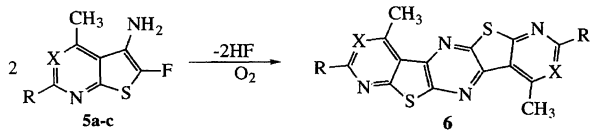
Treatment of **1a—c** with ethyl chlorofluoroacetate in tetrahydrofuran containing a slight excess of triethylamine (TEA) at room temperature afforded the corresponding sulfides **2** in excellent yields (Scheme 1). The structure of **2**[3e] was established by elemental analysis and spectroscopic methods



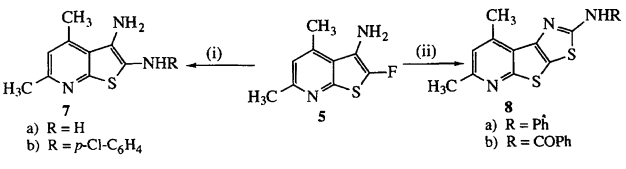

Scheme 1.

(Table 1). Thus, for example, the mass spectrum of **2a** had a molecular formula of $C_{12}H_{13}FN_2O_2S$ ($m/z$ = 268) and its $^1$H NMR spectrum showed the methyl and ethyl ester groups together with a doublet signal at $\delta$ = 7.24 for the methine proton coupled with a fluorine atom with the large coupling constant $J_{HF}$ = 50.4 Hz. In addition, the $^{19}$F NMR spectrum data supported the proposed structure as it showed a doublet fluorine signal at $\delta$ = −88.1 coupled with a hydrogen atom (Table 2).

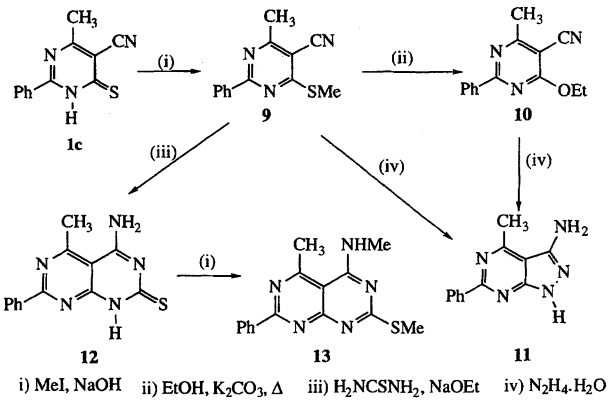
On the other hand, it was found that the change of solvent from THF to ethanol resulted in the formation of unexpected products **5** in high yields. The structure of **5** was established by various spectroscopic studies and by elemental analysis. For example, the mass spectrum of **5a** showed a molecular ion peak, $m/z$ = 196, which corresponds to the molecular formula of $C_9H_9FN_2S$, indicating the loss of a molecule of diethyl carbonate. $^{19}$F NMR showed a singlet signal at $\delta$ = −76.9. Fluorothienoazines **5** were assumed to be formed via the corresponding cyclized iminothienoazines **3**. Indeed treatment of **2a** with triethylamine in acetonitrile



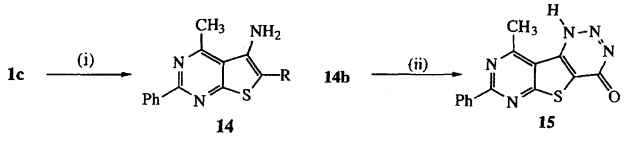

Scheme 2.




Scheme 3.




Scheme 4.




Scheme 5.

gave **3a**, which upon dissolution in ethanol and standing at room temperature gave **5a** directly. Similarly, we could prepare **5b** and **5c**, which were difficult to prepare in our previous anodical fluorination.[3e]

At 100 °C, 2-fluorothienoazines **5** undergo self-condensa-

Table 1. Yields, Mps, Color, Mass, and Elemental Analyses for Compounds **2c**, **3a**, **5c**, **6a—c**, **7a,b**, **8a,b**, **9**, **10**, **11**, **12**, **13**, **14a—d**, and **15** (compounds **2a,b** and **5a,b** cited in Ref. 3e)

| Compd | Yield | Mp | Color | MS (EI) | Elemental analysis | | |
|---|---|---|---|---|---|---|---|
| | % | °C | | *m/z* ($M^+$) | C | H | N |
| **2c** | 71 | 160—161 | Yellow | 331 | 58.00 (57.70) | 4.22 (4.40) | 12.68 (12.70) |
| **3a** | 65 | 121—122 | Pale Yellow | 268 | 53.72 (53.90) | 4.88 (4.60) | 10.44 (10.40) |
| **5c** | 60 | 171—172 | Light green | 259 | 60.23 (60.50) | 3.86 (3.80) | 16.21 (16.10) |
| **6a** | 70 | 270—270 | Dark pink | 350 | 61.71 (61.60) | 4.00 (3.80) | 16.00 (16.40) |
| **6b** | 80 | >290 | Dark blue | 474 | 70.88 (70.90) | 3.79 (3.80) | 11.81 (11.60) |
| **6c** | 50 | >290 | Dark green | 476 | 65.54 (65.60) | 3.36 (3.40) | 17.64 (17.60) |
| **7a** | 90 | 92—92 | Green | 193 | 55.95 (56.00) | 5.69 (5.70) | 21.76 (21.70) |
| **7b** | 88 | 122—123 | Green | 303 | 59.30 (59.10) | 4.61 (4.60) | 13.38 (13.80) |
| **8a** | 76 | 181—182 | Golden yellow | 311 | 61.73 (61.70) | 4.18 (4.20) | 13.50 (13.50) |
| **8b** | 75 | 210—211 | Golden yellow | 339 | 60.17 (60.20) | 3.83 (3.80) | 12.38 (12.40) |
| **9** | 92 | 133—135 | Colorless | 241 | 64.73 (64.63) | 4.56 (4.67) | 17.42 (17.33) |
| **10** | 92 | 90—92 | Colorless | 239 | 70.29 (70.39) | 5.43 (5.30) | 17.57 (17.45) |
| **11** | 80 | 160—162 | Pale yellow | 225 | 64.00 (63.85) | 4.88 (4.95) | 31.11 (31.24) |
| **12** | 80 | >300 | Pale yellow | 269 | 57.99 (57.81) | 4.08 (4.18) | 26.08 (26.15) |
| **13** | 85 | >300 | Colorless | 297 | 60.58 (60.73) | 5.05 (4.81) | 23.55 (23.70) |
| **14a** | 87 | 230—231 | Pale green | 266 | 63.15 (63.35) | 3.75 (3.61) | 21.05 (21.44) |
| **14b** | 89 | 212—214 | Pale green | 285 | 59.15 (58.93) | 4.22 (4.35) | 19.71 (19.60) |
| **14c** | 100 | 170—171 | Pale green | 284[a] | 63.60 (63.50) | 4.59 (4.69) | 14.84 (14.71) |
| **14d** | 85 | >300 | Pale green | 345 | 69.56 (69.45) | 4.34 (4.45) | 12.17 (12.01) |
| **15** | 90 | >300 | Colorless | 295 | 56.94 (56.82) | 3.05 (3.17) | 23.64 (23.51) |

a) MS(CI), *m/z* 284 ($M^+ + 1$).

Table 2. Spectral Data for Compounds **2c**, **3a**, **5c**, **6a—c**, **7a,b**, **8a,b**, **9**, **10**, **11**, **12**, **13**, **14a—d**, and **15** (IR, $^1$H NMR, and $^{19}$F NMR) (The spectral data of compounds **2a,b** and **5a,b** cited in Ref. 3e)

| Compd | IR ($cm^{-1}$) | $^1$H NMR ($\delta_H$) | $^{19}$F NMR ($\delta_F$) |
|---|---|---|---|
| **2c** | 2210 (CN); 1785 (C=O) | 1.32 (t, 3H, $CH_3$, $J$=7.5 Hz), 2.4 (s, 3H, $CH_3$), 4.21 (q, 2H, $CH_2$, $J$=7.5 Hz), 7.21 (d, 1H, CH, $J_{HF}$=50.1 Hz), 7.32—7.81 (m, 5H, aromatic H) | −89.51 (d, $J_{FH}$=50.3 Hz) |
| **3a** | 1795 (C=O) | 1.34 (t, 3H, $CH_3$, $J$=7.2 Hz), 2.45 (s, 3H, $CH_3$), 2.56 (s, 3H, $CH_3$), 4.35 (q, 2H, $CH_2$, $J$=7.2 Hz), 6.92 (s, 1H, CH), 7.31 (s, 1H) | −65.81 (S) |
| **5c** | 3400—3200 ($NH_2$) | 2.61 (s, 3H, $CH_3$), 4.32 (br, exch., 2H, $NH_2$), 6.92—7.49 (m, 5H, aromatic H) | −70.90 (S) |
| **6a** | 3000, 2450 ($CH_3$) | 2.51 (s, 6H, $2CH_3$), 2.67 (s, 6H, $2CH_3$), 6.80 (s, 1H, ring H), 6.91 (s, 1H, ring H) | |
| **6b** | 1660 (C=N) | (insoluble in DMSO) | |
| **6c** | 1665 (C=N) | (insoluble in DMSO) | |
| **7a** | 3400—3250 ($NH_2$) | 2.50 (s, 3H, $CH_3$), 2.59 (s, 3H), 3.91 (br, exch., 4H, $2NH_2$), 6.86 (s, 1H, ring H) | |
| **7b** | 3400—3250 ($NH_2$, NH) | 2.51 (s, 3H, $CH_3$), 2.60 (s, 3H, $CH_3$), 3.50 (br, exch., 2H, $NH_2$), 6.71—7.13 (m, 5H, aromatic H), 8.10 (s, 1H, NH) | |
| **8a** | 3400—3200 ($NH_2$, NH) | 2.51 (s, 3H, $CH_3$), 2.61 (s, 3H, $CH_3$), 6.71—7.24 (m, 6H, aromatic protons), 8.40 (s, exch., 1H, NH) | |
| **8b** | 3400—3200 ($NH_2$, NH) 1680 (C=O) | 2.51 (s, 3H, $CH_3$), 2.61 (s, 3H, $CH_3$), 6.85—7.24 (m, 6H, aromatic protons), 10.51 (s, exch., 1H, NH) | |
| **9** | 2216 (CN) | 2.68 (s, 3H, $CH_3$), 2.9 (s, 3H, $SCH_3$), 7.44—7.52 (m, 2 H, Ph), 8.41—8.43 (m, 3H, Ph) | |
| **10** | 2222 (CN) | 1.48 (t, 3 H, $CH_3$, $J$=6.48 Hz), 2.68 (s, 3H, $CH_3$), 4.64 (q, 2H, $CH_2$, $J$=7.07 Hz), 7.44—7.52 (m, 3H, Ph), 8.41—8.44 (m, 2H, Ph) | |
| **11** | 3418, 3384, 3181 ($NH_2$, NH) | 12.62 (s, 3 H, $CH_3$), 5.4 (br, exch., 2H, $NH_2$), 7.44—7.51 (m, 3H, Ph), 8.3—8.4 (m, 2H, Ph), 12.03 (br, exch., 1H, NH) | |
| **12** | 3425, 3015 ($NH_2$) | The sample is sparingly soluble in DMSO | |
| **13** | 3180 (NH) | 2.64 (s, 3H, $CH_3$), 3( s, 3 H, $SCH_3$), 3.21 (d, 3H, $NHCH_3$, $J$=4.69 Hz), 6.08 (br, exch., 1H, NH), 7.48 (m, 3H, Ph), 8.6 (m, 2H, Ph) | |
| **14a** | 3330, 3232 ($NH_2$) 2206 (CN) | 2.73 (s, 3H, $CH_3$), 7.04 (br, exch., 2H, $NH_2$), 7.49—7.78 (m, 3H, Ph), 8.53—8.56 (m, 2H, Ph) | |
| **14b** | 3420, 3315 ($NH_2$) 1695 (C=O) | 2.70 (s, 3H, $CH_3$), 6.91 (br, exch., 2H, $NH_2$), 7.20 (br, exch., 2H, $NH_2$), 7.45—7.50 (m, 3H, Ph), 8.60—8.62 (m, 2H, Ph) | |
| **14c** | 3417, 3291 ($NH_2$), 1651 (C=O) | 2.61 (s, 3H, $CH_3$), 2.78 (s, 3H, $COCH_3$), 7.44—7.47 (m, 3H, Ph), 8.61—8.63 (m, 2H, Ph) | |
| **14d** | 3460, 3290 ($NH_2$), 1687 (C=O) | 2.61 (s, 3H, $CH_3$), 7.11 (s, exch., 2H, $NH_2$), 7.45—8.61 (m, 10 aromatic H) | |
| **15** | 3089 (NH), 1667 (C=O) | 2.61 (s, 3H, $CH_3$), 7.44—7.49 (m, 3H, Ph), 8.60—8.62 (m, 2H, Ph), 13.56 (br, exch., 1H, NH) | |

tion to the dimeric cyclic compounds **6**. This interesting reaction also occurs slowly during the storage of the monomer **5** even at low temperatures. The very characteristic colors (**6a**: pink, **6b**: blue and **6c**: green color) of these products indicates that they are highly π-conjugated systems (Scheme 2). To our knowledge, this is the first synthesis of multi-fused heterocylic compounds in an essentially one-pot reaction.

The fluorine atom of **5** is also highly reactive to external amine nucleophiles. Thus, the fluorine atom of **5a** could be replaced by passing $NH_3$ gas in ether solution to give the diamino derivative **7a**. Similarly **5a** reacted with *p*-chloroaniline to yield **7b**. Compound **5a** also reacted with isothiocyanates to yield thiazolo[2′,3′ : 5,4]thieno[2,3-*b*]pyridine derivatives **8a,b** (Scheme 3).

The pyrimidine-thione **1c** could be converted into the *S*-methyl derivatives **9** by being dissolved in ethanol and sodium hydroxide, and treated with MeI at room temperature. Compound **9** showed high reactivity towards nucleophilic reagents. Thus, it reacted readily with ethanol in the presence of $K_2CO_3$ to give the ethoxypyrimidine derivative **10**

(Scheme 4). Upon refluxing compound **9** with $N_2H_4 \cdot H_2O$, a product of the condensation via elimination of methanethiol was formed. This product can be proposed as structure **11**. The pyrazolo[3,4-*d*]pyrimidine **11** was established on the basis of the IR spectrum, which revealed the absence of the cyano group stretching band and the presence of an amino group at 3418 and 3384 $cm^{-1}$. Similarly, the ethoxypyrimidine derivative **10** could be converted into **11** by refluxing in $N_2H_4 \cdot H_2O$. Compound **9** reacted readily with thiourea to give pyrimidino[4,5-*d*]pyrimidinethione **12**, which underwent *S*-methylation to afford compound **13** on treatment with MeI in the presence of NaOH.

This synthetic methodology of thienopyrimidine could be generalized using a variety of $\alpha$-haloacyl compounds to yield **14a—d** respectively. A unique multi-fused 1,2,3-triazine species (**15**) was obtained in excellent yield (90%) by treatment of **14b** with nitrous acid (Scheme 5).

## Experimental

All melting points are uncorrected. IR spectra were recorded (KBr) on a Pye Unicam SP 1000 spectrophotometer. $^1$H NMR and $^{19}$F NMR spectra were taken at 270 MHz using $CDCl_3$ as a solvent. The chemical shifts for $^1$H and $^{19}$F NMR are given in $\delta$ downfield from internal TMS and upfield from external $CF_3COOH$, respectively. High-resolution mass spectra were taken on a Hitachi M-80B GC-mass spectrometer and with GS/MS INCOS XL Finnigan MAT. Analytical data were obtained from the Microanalytical Data Center at Cairo University, Egypt and at the University of Salford, Salford M5, 4WT, UK. Pyridine- and pyrimidine-thione derivatives[4] **1a—c** were prepared according to procedures in the literature.[4,5]

**Preparation of 2-Pyridyl Sulfides (2a,b) and 4-Pyrimidinyl Sulfides (2c).** Ethyl chlorofluoroacetate (0.01 mol) was added to a solution of **1a—c** (0.01 mol) in THF (20 ml) containing triethylamine (0.012 mol). The reaction mixture was stirred at room temperature overnight, then passed through a short column (3 cm) of silica gel to yield almost pure fluorinated sulfides (**2a—c**).

**2-(Ethoxycarbonyl)-2-fluoro-3-imino-4,6-dimethyl-2,3-dihydrothieno[2,3-*b*]pyridine (3a).** Potassium carbonate (0.2 gm, 0.15 mmol) was added to a solution of **2a** (0.1 mmol) in dry acetonitrile (5 ml). The reaction mixture was stirred at room temperature for 1 h. After filtration of insoluble material, the filtrate was evaporated under vacuum at room temperature, to yield **3a** (170 mg).

**General Procedure for 5a—c.** A solution of **2** (0.1 mmol) in ethanol (5 ml) containing potassium carbonate (0.15 mmol) was stirred at room temperature for 3 h. After filtration of insoluble material, the filtrate was evaporated under vacuum to yield **5a—c** in pure form.

**Dimerization of 5a—c. General Procedure for 6a—c.** Compound **5** (0.01 mmol) was refluxed for 1 h in xylene. The solid product was collected and washed three times with hot ethanol.

**4,6-Dimethylthieno[2,3-*b*]pyridine-2,3-diamine (7a).** A solution of **5a** (0.1 mmol) in dry ether (10 ml) in a conical flask fitted with a bubbler stopper was placed in an ice bath. Dry $NH_3$ gas was bubbled into the solution (care was taken to remove moisture as much as possible). The passage of $NH_3$ gas was continued for 30 min. The solution then was filtered by gravity to remove any impurities, and kept in a refrigerator overnight before being evaporated under vacuum to yield pure **7a**.

**4,6-Dimethyl-2-*p*-chloroanilinothieno[2,3-*b*]pyridine-3-amine (7b).** *p*-Chloroaniline (0.1 mmol) and $K_2CO_3$ (0.1 mmol) was added to a solution of **5a** (0.1 mmol) in dry acetonitrile (10 ml). The reaction mixture was stirred at room temperature overnight. After filtration of insoluble material the filtrate was evaporated under vacuum to yield **7b**.

**2-Anilino-4,6-dimethylthiazolo[2′,3′ : 5,4]thieno[2,3-*b*]pyridine (8a).** Phenyl isothiocyanate (0.1 mmol) was added to a solution of **5a** (0.1 mmol) in dioxane (10 ml) containing $K_2CO_3$ (0.2 g, 0.15 mmol). The reaction mixture was stirred for 48 h, then refluxed for an additional 0.5 h. After filtration of insoluble material, the filtrate was evaporated under vacuum to yield **8a**, which was recrystallized from absolute ethanol.

**2-Benzamido-4,6-dimethylthiazolo[2′,3′ : 5,4]thieno[2,3-*b*]-pyridine (8b).** Benzoyl chloride (0.1 mmol) was added to suspension of ammonium thiocyanate (0.7 g, 0.1 mmol) in dry dioxane (20 ml). The reaction mixture was refluxed for 1 min, then treated with **5a** (0.1 mmol) at room temperature for 2 h. The reaction mixture was refluxed for 10 min. After filtration of insoluble material, the filtrate was evaporated under vacuum to yield **8b** which was recrystallized from absolute ethanol.

**4-Methyl-6-methylthio-2-phenylpyrimidine-5-carbonitrile (9).** Pyrimidine-2(1*H*)-thione (**1**) (0.01 mol) was dissolved in ethanol (25 ml) and sodium hydroxide (10%, 10 ml), then treated with methyl iodide (2 ml) dropwise, with stirring, at room temperature. The reaction mixture was left being stirred at room temperature for 2 h. The solid obtained was recrystallized from ethanol to give pure **9** as colorless crystals.

**4-Ethoxy-6-methyl-2-phenylpyrimidine-5-carbonitrile (10).** Pyrimidine-5-carbonitrile (**9**) (0.01 mol) was dissolved in ethanol (20 ml) contining anhydrous potassium carbonate (1.5 g), and left under reflux for 3 h. The solid obtained was recovered by filtration and washed several times with water.

**3-Amino-4-methyl-6-phenyl-1*H*-pyrazolo[3,4-*d*]pyrimidine (11).** A mixture of pyrimidine-5-carbonitrile (**9**) (0.01 mol) or (**10**) (0.01 ml) and hydrazine hydrate (2 ml) in ethanol (20 ml) was left under reflux for 3 h. The solid obtained after evaporation was collected and recrystallized from ethanol.

**4-Amino-5-methyl-7-phenylpyrimidino[4,5-*d*]pyrimidine-2(1*H*)-thione (12).** A mixture of pyrimidine-5-carbonitrile (**9**) (0.01 mol) and thiourea (0.01 mol) in ethanol (20 ml) and sodium ethoxide (sodium metal 0.46 g, 0.02 mol in 10 ml EtOH) was left under reflux for 4 h. The solid obtained after acidification with concd HCl was recrystallized from DMF/ethanol.

**5-Methyl-4-methylamino-2-methylthio-7-phenylpyrimidino-[4,5-*d*]pyrimidine (13).** Pyrimidino[4,5-*d*]pyrimidine (**12**) (0.01 mol) was dissolved in ethanol (20 ml) contining sodium hydroxide (10 ml, 10%) by warming, and methyl iodide (2 ml excess) was added with stirring for 2 h. The solid obtained was recrystallized from ethanol.

**Reaction of 1c with Alkyl Halides: General Procedure:** Equimolar amounts of pyrimidine-2(1*H*)-thione (**1c**) (0.01 mol) and $\alpha$-halo acyl compounds (0.01 mol) in absolute ethanol containing anhydrous potassium carbonate (2 g, 15 mol) were refluxed for 3 h. The reaction mixture was diluted by water and the solid formed was collected by filtration and recrystallized from ethanol.

**9-Methyl-7-phenylpyrimidino[4′,5′ : 5,4]thieno[3,2-*d*]-1,2,3-triazin-4(1*H*)-one (15).** 3-Amino-4-methylthieno[2,3-*d*]-pyrimidine-2-carboxamide (**14b**) (0.01 mol) in acetic acid (25 ml) was treated with sodium nitrite (0.14 g, 0.02 mol) portionwise with stirring at room temperature, and left being stirred for 1 h. The solid was collected and purified by recrystallization from acetic acid.

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