***SHORT PAPER***

*J. Chem. Research (S),*
2001, 439–441

# The reaction of dimethyl N-cyanodithioiminocarbonate with amino- and oxo-azoles: a new general synthesis of methylsulfanylazoloazines†

**Galal H. Elgemeie* and Soha A. Sood**

*Chemistry Department, Faculty of Science, Helwan University, Ain-Helwan, Cairo, Egypt*

A novel and efficient method for the synthesis of a new variety of methylsulfanylazoloazines by the reaction of dimethyl N-cyanodithioiminocarbonate with diazoles containing amino and active methylene functions. The synthetic potential of the method is demonstrated.

**Keywords:** dimethyl N-cyanodithioiminocarbonate, methylsulfanylazoloazines

The synthesis and reactions of dimethyl N-cyanodithioiminocarbonate, in the preparation of fused heterocycles, has attracted much attention.[1,2] We are developing new procedures for the synthesis of thioguanine and mercaptopurine analogues and other antimetabolites.[3–5] We have recently reported different successful approaches for the synthesis of mercaptopurine analogues using ketene dithioacetals.[6–8] In an extension to this work, we now report a novel synthesis of methylsulfanylazoloazines by the reaction of dimethyl N-cyanodithioiminocarbonate **2** with diazoles containing amino and active methylene functions. Thus, it has been found that dimethyl N-cyanodithioiminocarbonate **2** reacts with 3,5-diaminopyrazoles **3** in refluxing ethanol containing catalytic amounts of piperidine gave the corresponding 4-methylthiopyrazolo[1,5-a]-1,3,5-triazines **6a–f** in good yield. The structure of compounds **6a–f** were established by their elemental analysis and spectroscope data (IR, $^1$H NMR, $^{13}$C NMR and MS). The analytical data for **6a** revealed a molecular formula $C_{12}H_{12}N_8S$, ($M^+ = 300$). $^1$H NMR spectroscopy was used to confirm this structure for the product. Thus, the $^1$H NMR spectrum revealed a signal at δ 2.61 ppm assigned to a $SCH_3$ group, a multiplet at δ 7.12–7.81 ppm assigned to the aromatic protons and broad singlet at δ 6.97 and 8.03 ppm assigned to an amino group. The formation of **6** from the reaction of **2** and **3** is assumed to proceed via the intermediate **5**, which cyclised to yield the end products **6**. Although, one may argue that the reaction of **2** with 3,5-diaminopyrazoles **3** may involve the exocyclic pyrazole nitrogen leading to the other possible 4-amino regioisomers, it is known that the endocyclic ring nitrogen is the most nucleophilic centre in the molecule and in a basic medium, it will initially add to the unsaturated double bond of **2**.[9] Compounds **6** bearing latent functional substituents were found useful for the synthesis of other derivatives. Thus, it has been found that compounds **6** reacted with hydrazine or aniline in refluxing ethanol containing catalytic amounts of piperidine to afford the corresponding hydrazino or anilino derivatives **7a–f**, **7g–l**, respectively. The structure of compounds **7** was established on the basis of elemental analysis and spectral data. Thus, the mass spectrum of **7d** was compatible with the molecular formula $C_{11}H_{11}ClN_{10}$ ($M^+ = 318$), and the $^1$H NMR spectrum contained three broad bands at δ 4.57, 6.81 and 7.23 ppm assigned to three amino group and a broad singlet at δ 9.31 ppm assigned to an NH group. The behaviour of dithioacetals **2** towards azolones was also investigated. Thus, it has been found that compound **2** reacted with 3-methyl-5-oxopyrazole **8** in refluxing ethanol containing catalytic amounts of piperidine to yield the corresponding 2-imino-2,7-dihydro-5-methyl-4-methylthio-pyrazolo[4,5-e]-1,3-oxazine derivatives **10** in good yields. The structure of compounds **10a,b** was established on the basis of their elemental analysis and spectral data (IR, $^1$H NMR and MS). Thus, structure **10b** followed from the IR which revealed the absence of a CN band, whilst its mass spectrum which showed a molecular formula $C_{13}H_{12}N_4OS$ ($M^+ = 272$). The $^1$H NMR was used to confirm this structure for the product. Thus, the $^1$H NMR spectrum revealed a band δ 2.64 ppm assigned to a $SCH_3$ group, a multiplet at δ 7.20–8.00 ppm assigned to the aromatic protons and a broad singlet at δ 7.10 ppm associated with an imino group. The formation of **10** from the reaction of **2** and **8** may proceed via the initial Michael addition of the active methylene in **8** to the double bond of **2** to yield the intermediate **9**. This Michael adduct then cyclises via the elimination of $CH_3SH$ followed by cyclisation to the cyano group to give the stable product **10**.

The reaction of **2** with 2-cyanomethylbenzimidazole **11** can be utilised for the synthesis of the interesting 2-amino-5-cyano-4-(methylthio)pyrimidino[1,6-a]benzimidazole **14.** Thus, it has been found that **11** reacted with **2** in dioxane containing a catalytic amount of potassium hydroxide for 24 h at room temperature to give the 4-(methylthio)pyrimidino[1,6-a]benzimidazole **14**. The structure of **14** was established for the reaction products on the basis of its elemental analysis and spectroscopic data (IR, $^1$H NMR, $^{13}$C NMR and MS). Thus, structure **14** is supported by its analytical data which revealed a molecular formula $C_{12}H_9N_5S$ ($M^+ = 255$). $^1$H NMR spectroscopy was used to confirm this structure for the product. Thus, $^1$H NMR spectroscopy revealed a broad band in the range δ = 8.43 ppm assigned to an amino group. The formation of **14** from the reaction of **11** and **2** may proceed via the initial Michael addition of the active methylene group of **11** to the double bond of **2**. The Michael adduct which was formed then cyclised smothly via elimination of $CH_3SH$ and addition to the cyano group to yield the final product **14**.

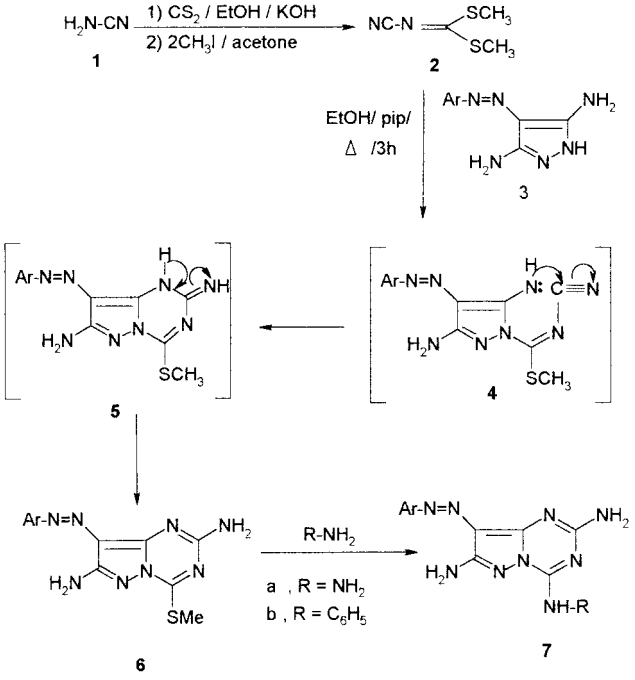
In summary, we have achieved a regiospecific synthesis of interesting mercaptopurine analogues by the reaction of dimethyl N-cyanodithioiminocarbonate with diazoles containing amino and active methylene functions. The compounds which have been synthesised are being subjected to biological testing.

## Experimental

All melting points were determined in open glass capillaries on a Gallenkamp melting point apparatus and are uncorrected. Their spectra were obtained (KBr disc) on a Perkin Elmer / 1650. FT-IR insturment. The $^1$H NMR and $^{13}$C NMR. Spectra were measured in a Varian

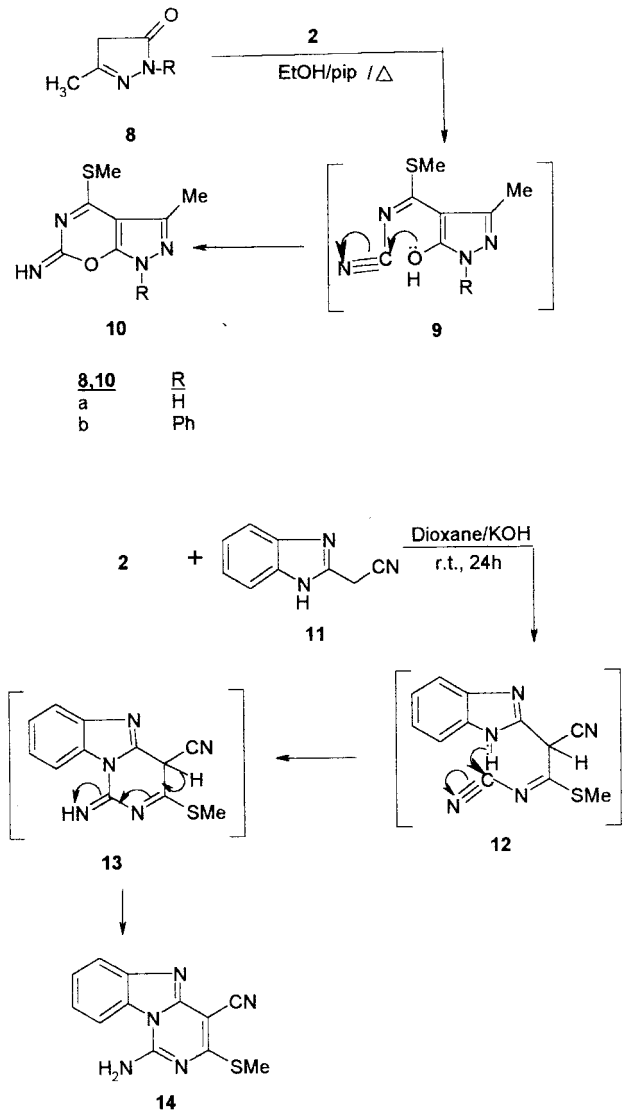
* To receive any correspondence.

† This is a Short Paper, there is therefore no corresponding material in *J Chem. Research (M).*



| 6 | Ar | 7 | R | Ar | 7 | R | Ar |
|---|---|---|---|---|---|---|---|
| a) | $C_6H_5$ | a) | $NH_2$ | $C_6H_5$ | g) | $C_6H_5$ | $C_6H_5$ |
| b) | 4- $H_3C$-$C_6H_4$ | b) | $NH_2$ | 4- $H_3C$-$C_6H_4$ | h) | $C_6H_5$ | 4-$H_3C$ – $C_6H_4$ |
| c) | 4-$H_3CO$-$C_6H_4$ | c) | $NH_2$ | 4-$H_3CO$-$C_6H_4$ | i) | $C_6H_5$ | 4-$H_3CO$-$C_6H_4$ |
| d) | 4- Cl – $C_6H_4$ | d) | $NH_2$ | 4- Cl – $C_6H_4$ | j) | $C_6H_5$ | 4-Cl-$C_6H_4$ |
| e) | 4-$O_2N$- $C_6H_4$ | e) | $NH_2$ | 4-$O_2N$- $C_6H_4$ | k) | $C_6H_5$ | 4-$O_2N$-$C_6H_4$ |
| f) | 2,5–Cl,Cl-$C_6H_3$ | f) | $NH_2$ | 2,5–Cl,Cl-$C_6H_3$ | l) | $C_6H_5$ | 2,5- Cl,Cl-$C_6H_3$ |

**Scheme 1**



**Scheme 2**

400 or wilmad 270 MHZ spectrometer for $(CD_3)_2$ SO solutions using $SiMe_4$ as internal standard. Mass spectra were recorded in a Varian MAT 112 spectrometer. Analytical data were obtained from the Micro Analytical Data Center at Cairo University.

*8-Arylazo-2,7-diamino-4-(methylthio)pyrazolo[1,5-a]-1,3,5-triazines* **6a–f**: *general procedure:* A suspension of dimethyl N-cyanodithioiminocarbonate **2** (0.01 mol) in ethanol (30 ml) was refluxed with 4-arylazo-3,5-diamino- -pyrazoles **3** (0.01 mol) and three drops of piperidine, for 3 hours. The reaction mixture was left to cool to room temperature. The crystals separating on cooling were filtered off, and recrystallised from the appropriate solvent.

**6a**: Yellow crystals, m.p. 300°C, (from EtOH), yield (85%). IR(KBr) $\nu_{max}$/cm$^{-1}$3425 and 3271 ($NH_2$) and 1643 (C = N). $^1$H NMR (DMSO) δ 2.61 (s, 3H, $SCH_3$), 6.97 (s, 2H, $NH_2$), 7.12–7.81 (m, 5H, $C_6H_5$), 8.03 (s, 2H, $NH_2$). $^{13}$C NMR (DMSO) δ 13.19 ($SCH_3$), 113.55 (C-9), 122.07–130.97 (phenyl carbons), 149.43 (C-8), 152.52 (C-7), 152.90 (C-4), 158.58 (C-2). $C_{12}H_{12}N_8S$ ($M^+$ = 300), Calcd: C, 48.0; H, 4.0, N, 37.3; S, 10.66. Found: C, 48.2; H, 4.3; N, 37.5; S, 10.6 %. **6b**: Brown crystals, m.p. 270ºC (from EtOH), yield (89%). IR (KBr)$\nu_{max}$/cm$^{-1}$ 3441 and 3376 ($NH_2$) and 1670 (C=N). $^1$H NMR (DMSO) δ 2.36 (s, 3H, $CH_3$), 2.60 (s, 3H, $SCH_3$), 6.94(s, 2H, $NH_2$), 7.21–7.89 (m, 5H , $C_6H_5$), 8.55 (s, 2H, $NH_2$). $^{13}$C NMR (DMSO) δ 11.85 ($CH_3$), 20.77 ($SCH_3$), 113.22 (C-9), 123.81–138.44 (phenyl carbons), 149.18 (C-8), 152.60 (C-7), 158.50 (C-4), 159.29 (C-2). $C_{13}H_{14}N_8S$, Calcd: C, 49.68; H, 4.45; N, 35.66; S,10.19. Found: C, 59.4; H, 4.2; N, 35.3; S,10.2%. **6c**: Brown cystals, m.p. > 300°C (from EtOH), yield (79%). IR (KBr) $\nu_{max}$ / cm$^{-1}$ 3432 ($NH_2$) and 1634 (C = N). $C_{13}H_{14}N_8OS$, Calcd: C, 47.27; H, 4.24; N, 33.93; S, 9.69. Found: C, 47.5; H, 4.1; N, 33.7; S, 9.8%. **6d**: Brown, m.p. > 300°C (from EtOH), yield (88%). IR (KBr) $\nu_{max}$/ cm$^{-1}$ 3428 ($NH_2$), 1635 (C = N). $^1$H NMR (DMSO) δ 2.61 (s, 3H, $SCH_3$), 7.13 (s, 2H, $NH_2$), 7.50–7.72 (m, 4H, $C_6H_4$), 7.75 (s, 2H, $NH_2$). $^{13}$C NMR (DMSO) δ 11.80 ($SCH_3$), 122.28 (C-9), 124.91–131.98 (phenyl carbons), 151.63 (C-8), 152.39 (C-7), 158.62 (C-4), 159.38 (C-2). $C_{12}H_{11}ClN_8S$ ($M^+$ = 334), Calcd: C, 43.04; H, 3.29; N, 33.48; S, 9.57 Found: C, 43.0; H, 3.21; N, 33.5; S, 9.4 %. **6e**: Yellow crystals, m.p. > 300°C (from EtOH), yield (83%). IR (KBr) $\nu_{max}$ / cm$^{-1}$ 3418 ($NH_2$) and 1644 (C = N). $^1$H NMR (DMSO) δ 2.58 (s, 3H, $SCH_3$), 7.20–7.86 (m, 4H, $C_6H_4$), 8.31 (s, br, 2H, $NH_2$), 8.40 (s, br, 2H, $NH_2$). $^{13}$C NMR (DMSO) δ 13.21 ($SCH_3$), 113.67 (C-9), 128.13–129.03 (phenyl carbons), 149.52 (C-8), 152.61 (C-7), 153.02 (C-4), 158.67 (C-2). $C_{12}H_{11}N_9O_2S$ ($M^+$ = 345), Calcd: C, 41.73; H, 3.18; N, 36. 52; S, 9.27. Found: C, 41.8; H, 3.2; N, 36.6; S, 9.3 %. **6f:** Red crystals, m.p. > 300°C (from EtOH), yield (86%). IR (KBr) $\nu_{max}$ / cm$^{-1}$ 3407 ($NH_2$) and 1635 (C = N). $^1$H NMR (DMSO) δ 2.61 (s, 3H, $SCH_3$), 7.14–7.82 (m, 3H, $C_6H_3$), 8.00 (s, br, 2H, $NH_2$), 8.45 (s, br, 2H, $NH_2$). $^{13}$C NMR (DMSO) δ13.55 ($SCH_3$), 115.78 (C-9), 129.20-132.57 (phenyl carbons), 149.15 (C-8), 152.12 (C-7), 159.03 (C-4), 159.88 (C-2). $C_{12}H_{10}Cl_2N_8S$ ($M^+$ = 368), Calcd: C, 39.02; H, 2.71; N, 30.35; S, 8.67. Found: C, 39.2; H, 2.6; N, 30.6, S, 8.5 %.

*8-Arylazo–2,7-diamino4-hydrazinopyrazolo[1,5-a]–1,3,5-triazines* **7a-f**: *general procedure*: A mixture of **6** (0.01 mol) and hydrazine (0.01 mol) in ethanol (30 ml) containing a catalytic amounts of piperidine, was refluxed for 3 hours. The solution was cooled and the precipitate was filtered off and crystallised from the appropriate solvent.

**7a**: Yellow crystals, m.p. 270°C (from EtOH), yield (74%). IR (KBr) $\nu_{max}$/cm$^{-1}$ 3479, 3377 and 3313 ($NH_2$, NH) and 1589 (C = N). $C_{11}H_{12}N_{10}$, Calcd: C, 46.47; H, 4.22; N, 49.29. Found: C, 46.5; H, 4.3; N, 49.6 %. **7b**: Brown crystals, m.p. > 300ºC ( from EtOH), yield (81%). IR (KBr) $\nu_{max}$ /cm$^{-1}$ 3400 and 3230 ($NH_2$, NH), 1586 (C = N). $^1$H NMR (DMSO) δ 2.38 (s, 3H, $CH_3$), 6.74 (s, br, 2H, $NH_2$), 6.93 (s, br, 2H, $NH_2$), 7.20–7.91 (m, 4H, $C_6H_4$), 8.96 (s, br, 2H, $NH_2$), 9.33 (s, br, 1H, NH). $C_{12}H_{14}N_{10}$, Calcd: C, 48.32; H, 4.70; N, 46.97. Found: C, 48.6; H, 4.8; N, 47.1%. **7c**: Brown crystals, m.p. 275°C (from EtOH), yield (84%). IR (KBr) $\nu_{max}$/cm$^{-1}$ 3357 and 3150 ($NH_2$, NH). $C_{12}H_{14}N_{10}O$, Calcd: C, 45.85; H, 4.45; N, 44.58. Found: C,

46.1; H, 4.4; N, 44.7 %. **7d**: Brown crystals, m.p. > 300ºC ( from EtOH), yield (86%). IR (KBr) $\nu_{max}/cm^{-1}$ 3484, 3298 and 3146 ($NH_2$, NH) and 1624 (C = N). $^1H$ NMR (DMSO) δ 4.57 (s, br, 2H, $NH_2$), 6.81 (s, br, 2H, $NH_2$), 7.23 (s, br, 2H, $NH_2$), 7.42–7.73 (m, 4H,$C_6H_4$), 9.31 (s, br, 1H, NH). $^{13}C$ NMR (DMSO) δ 116.43 (C-9), 128.90–131.37 (phenyl carbons), 149.29 (C-8), 150.92 (C-7), 151.13 (C-4), 160.97 (C-2). $C_{11}H_{11}ClN_{10}$ ($M^+$ = 318), Calcd: C, 41.44; H, 3.45; N, 43.95. Found: C, 41.9; H, 3.7; N, 44.1%. **7e**: Yellow crystals, m.p. > 300°C ( from EtOH), yield (81%). IR (KBr) $\nu_{max}/cm^{-1}$ 3320, 3114 ($NH_2$, NH), and 1597 (C = N). $^1H$ NMR (DMSO) δ 3.27 (s, br, 2H, $NH_2$), 6.91 (s, br, 2H, $NH_2$), 7.70–7.91 (m, 4H, $C_6H_4$), 8.27 (s, br, 2H, $NH_2$), 8.31 (s, br, 1H, NH). $^{13}C$ NMR (DMSO) δ 112.38 (C-9), 122.35-126.91 (phenyl carbons), 145.11 (C-8), 149.28 (C-7), 157.72 (C-4), 162.22 (C-2). $C_{11}H_{11}N_{11}O_2$, Calcd: C, 40.12 ; H, 3.34; N, 46.80. Found: C, 40.5; H, 3.6 ; N, 47.0%. **7f**: Red crystals, m.p. > 300°C (from EtOH), yield (78%). IR (KBr) $\nu_{max}/cm^{-1}$ 3402, 3318 and 3112 ($NH_2$, NH) and 1617 (C = N). $C_{11}H_{10}Cl_2N_{10}$, Calcd: C, 37.39; H, 2.83; N, 39.66. Found: C, 37.4; H, 3.0 ; N, 39.6%.

*4-Anilino-8-arylazo-2,7-diaminopyrazolo[1,5-a]–1,3,5-triazines* **7g–l**: *general procedure:* A mixture of **6** (0.01 mol) and aniline (0.01 mol) were heated at 150°C (bath temperature) for 30 min, the resulting solid product was triturated with water, filtered off and crystallised from the appropriate solvent.

**7g**: Yellow crystals, m.p. 230°C ( from EtOH), yield (70%). IR (KBr) $\nu_{max}/cm^{-1}$ 3411, 3310 and 3101 ($NH_2$, NH) and 1650 (C = N). $^1H$ NMR (DMSO) δ 6.46 (s, br, 2H, $NH_2$), 7.00–7.99 (m, 10H, $C_6H_5$ and $C_6H_5$), 8.01 (s, 2br, 2H, $NH_2$), 9.42 (s, br, 1H, NH). $^{13}C$ NMR (DMSO) δ 113.39 (C-9), 120.77–128.85 (2 phenyl carbons), 148.48 (C-8), 148.24 (C-7), 158. 13 (C-4), 160.01 (C-2). $C_{17}H_{15}N_9$, Calcd: C, 59.13; H, 4.35; N, 36.52. Found: C, 59.0; H, 4.1; N, 36.5 %. **7h**: Yellow crystals, m.p. 300°C ( from EtOH), yield (73%). IR (KBr) $\nu_{max}/cm^{-1}$ 3271 and 3110 ($NH_2$, NH ), 1612 (C = N). $^1H$ NMR (DMSO) δ 2.36 ($CH_3$), 6.57 (s, br, 2H, $NH_2$), 7.00–7.91 (m, 9H, $C_6H_5$ and $C_6H_4$), 8.48 (s, br, 2H, $NH_2$), 9.35 (s, br, 1H, NH). $C_{18}H_{17}N_9$, Calcd: C, 60.77; H, 4.74; N, 35.0. Found: C,60.1; H, 4.8; N, 34.0 %. **7i**: Brown crystals, m.p. 300°C ( from EtOH), yield (80%). IR (KBr) $\nu_{max}/cm^{-1}$ 3358 and 3173 ($NH_2$, NH) and 1644 (C = N). $C_{18}H_{17}N_9O$, Calcd: C, 57.60; H, 4.53; N, 33.60. Found: C, 57.9; H, 4.7; N, 33.9%. **7j**: Brown, m.p. 300°C (from EtOH), yield (80%). IR (KBr) $\nu_{max}/cm^{-1}$ 3434 ($NH_2$, NH), 1624 (C = N). $^1H$ NMR (DMSO) δ 4.11 (s, br, 2H, $NH_2$), 5.01 (s, br, 1H, NH), 6.51 (s, br, 2H, $NH_2$), 7.04–7.77 (m, 9H, $C_6H_5$ and $C_6H_4$). $C_{17}H_{14}ClN_9$ ($M^+$ = 379) Calcd: C, 53.75 ; H, 3.69; N, 33.21. Found: C, 54.0; H, 3.9; N, 33.5%. **7k**: Pale yellow, m.p. > 350°C ( from EtOH), yield (79%). IR (KBr) $\nu_{max}/cm^{-1}$ 3645 and 3405 ($NH_2$, NH) and 1645 (C = N). $C_{17}H_{14}N_{10}O_2$, Calcd: C, 52.30; H, 3.58; N, 35.89. Found: C, 52.6; H, 3.7; N, 36.0%.**7l**: Red crystals, m.p. 285–287°C ( from EtOH), yield (87%). IR (KBr) $\nu_{max}/cm^{-1}$ 3396, 3280 and 3126 ($NH_2$, NH), and 1610 (C = N).$C_{17}H_{14}Cl_2N_9$, Calcd: C, 49.15; H, 3.37; N, 30.36. Found: C,49.1; H, 3.6; N, 30.3%.

*2-Imino-2,7-dihydro–5-methyl-4-methylthio-7-substituted- pyrazolo[4,5,-e]-1,3-oxazine* **10a, b**: *general procedure:* A suspension of dimethyl N-cyanodithioiminocarbonate **2** (0.01 mol) in ethanol (30 ml) was refluxed with 3-methyl 1-substituted 5-oxopyrazole **8** (0.01 mol) and three drops of piperidine for 3 hours, the reaction mixture was left to cool to room temperature the crystals separation on cooling were filtered off and crystallised from the appropriate solvent.

**10a**: Pale yellow, m.p. 140°C ( from EtOH), yield (70%). IR (KBr) $\nu_{max}/cm^{-1}$ 3738 and 3651 (NH). $C_7H_8N_4OS$, Calcd: C, 42.86; H, 4.48; N, 28.57; S, 16.33. Found: C,42.7; H, 4.7; N, 28.5; S, 16.2.%. **10b**: Yellow, m.p. 210°C ( from EtOH), yield (85%). IR (KBr) $\nu_{max}/cm^{-1}$ 3371 and 3176 (NH) and 2925 ($CH_3$).

$^1H$ NMR (DMSO) δ 2.28 (s, 3H, $CH_3$), 2.64 (s, 3H, $SCH_3$), 7.10 (s, br, 1H, imine NH). 7.20-8.00 (m, 5H, $C_6H_5$), $^{13}C$ NMR (DMSO) δ 16.5 ($CH_3$), 17.8 ($SCH_3$), 120.91–130.42 (phenyl carbons), 140.02 (C-9), 143.54 (C-5), 147.74 (C-4), 152.44 (C-8), 165.59 (C-2). $C_{13}H_{12}N_4OS$ ($M^+$ = 272), Calcd: C, 57.35; H, 4.41; N, 20.59; S, 11.76. Found: C, 57.0; H, 4.3; N, 20.2; S, 11.8.%.

*2-Amino-5-cyano-4-methylthiopyrimidino[1,6-a]benzimidazole* **14**: *general procedure:* A suspension of dimethyl N-cyanodithioiminocarbonate **2** (0.01 mol) was added to a stirred solution of 2-cyanomethylbenzo [1,2-b] imidazole **11** (0.01 mol) in a dry dioxane (50 ml) containing potassium hydroxide (0.01 mol). After stirring overnight at room temperature the reaction mixture was diluted with cold water (50 ml). The resulting solid product was collected by filtration and recrystallised from the appropriate solvent.

**14**: Yellow, m.p. > 300°C (from EtOH), yield (87%). IR (KBr) $\nu_{max}/cm^{-1}$ 3853 and 3465 ($NH_2$), 2210 (CN) and 1655 (C = N). $^1H$ NMR (DMSO) δ 2.60 (s,3H,$SCH_3$), 7.19–7.71 (m, 4H, $C_6H_4$), 8.43 (s, br, 2H, $NH_2$). $^{13}C$ NMR (DMSO) δ 12.33 ($SCH_3$), 112.47 (C-5), 114.5 (CN), 120.18–128.50 ($C_6H_4$), 144.64 (C-4), 151.95 (C-13), 164.44 (C-2). $C_{12}H_9N_5S$ ($M^+$ = 255), Calcd: C, 56.47; H, 3.53; N, 27.45; S, 12.56. Found: C, 56.8; H, 3.7; N, 27.9; S, 12.3%.

*Received 22 March 2001; accepted 10 June 2001*
*Paper 01/809*

## References

1 M.T. Cocco and A. Maccioni, *Synthesis*, 1991, **7**, 529.
2 Y. Tominaga, K. Ogata, S. Kohra, M. Hojo and A. Hosomi, *Tetrahedron Lett.*, 1991, **32**, 5987.
3 G.E.H. Elgemeie, A.H. Elghandour, A.M. Elzanate and S.A. Ahmed, *J. Chem. Soc. Perkin Trans. 1*, 1997, 3285.
4 G.E.H. Elgemeie, A.M. Attia, D.S. Farag and S.M. Sherif, *J. Chem. Soc. Perkin Trans. 1*, 1994, 1285.
5 G.E.H. Elgemeie and B.A. Hussein, *Tetrahedron*., 1994, **50**, 199.
6 G.E.H. Elgemeie, H.A. Ali and A.M. Elzanate, *J. Chem. Research (S)*, 1996, 340.
7 G.E.H. Elgemeie, A.H. Elghandour, A.M. Elzanate and W.A. Masoud *J. Chem. Research (S)*, 1998, 164.
8 G.E.H. Elgemeie, A.H. Elghandour, A.M. Elzanate and S.A. Ahmed, *J. Chem. Research (S)*, 1998, 162.
9 G.E.H. Elgemeie *et al*. *Advances in Heterocyclic Chemistry*, Ed. A.R. Katritzky, Academic Press, Vol. 41, pp. 320–367 1987.