2-Chloro-4,5-dihydroimidazole. Part VII.[1] Reactions with Carbon Disulfide and Potassium *O*-Alkyl Dithiocarbonates. Synthesis and Transformations of 2,3,7,8-Tetrahydro-5*H*-diimidazo[2,1-*b*:1′,2′-*e*][1,3,5]thiadiazine-5-thione

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2-Chloro-4,5-dihydroimidazole **1** reacted with carbon disulfide to give 2,3,7,8-tetrahydro-5*H*-diimidazo[2,1-*b*:1′,2′-*e*][1,3,5]thiadiazine-5-thione **5**. Acid hydrolysis of **5** gave imidazolidine-2-thione **6**, which reacted with amines, dialkylhydrazines, and alkoxides to give thioureas **9**, thiosemicarbazides **10** and thiocarbamates **11**, respectively. Reaction of the hemisulfate of **1** with excess of potassium *O*-alkyl dithiocarbonates afforded diesters of thiodicarbonic acid **13**.

Recently, Yasumoto *et al.* have reported[2] on the cyclocondensation of dialkylcyanamides with carbon disulfide at high pressure leading to the formation of 1,3,5-thiadiazine derivatives. We now wish to describe that 2-chloro-4,5-dihydroimidazole **1** containing a chloroamidine moiety reacted with CS_2 under mild conditions to give diimidazo[1,3,5]-thiadiazine **5**. The reaction outlines the value of compound **1** in the constructions of novel nitrogen-bridgehead heterocyclic systems.[3]

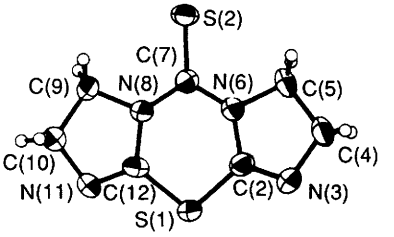


Fig. 1 Molecular structure of compound **5**

Results and Discussion

Treatment of the compound **1** with carbon disulfide in the presence of triethylamine at 20 °C for 48 h afforded the title 1,3,5-thiadiazine **5** in 54% yield. The mechanistic pathway for this reaction was deduced as shown in Scheme 1.

The initial step is the addition of the N–H group of **1** to the C=S double bond of the disulfide, followed by an internal nucleophilic substitution of the chlorine atom to give imidazothiazetidine **3**. Then, the intermediate **3** reacts with a second molecule of **1** yielding the chloro derivative **4**. The final product is formed by 1,3,5-thiadiazine ring closure in a second intramolecular displacement of the chlorine with a sulfur atom. It is noteworthy that the intermediate imidazothiazetidine **3** cannot be isolated from the reaction mixture, apparently due to its high susceptibility to the nucleophilic ring opening.

The ^1H NMR spectrum of **5** revealed two multiplets at δ 4.0 and 4.25, and a pattern typical of the AA′BB′ system. The ^{13}C NMR spectrum exhibits four resonances: two signals indicative of CH_2 groups at δ 51.1 and 53.0, as well as signals at δ 145.6 and 167.9 due to the C=N and C=S groups, respectively. Molecular structure obtained from X-ray analysis of compound **5** is shown in Fig. 1.

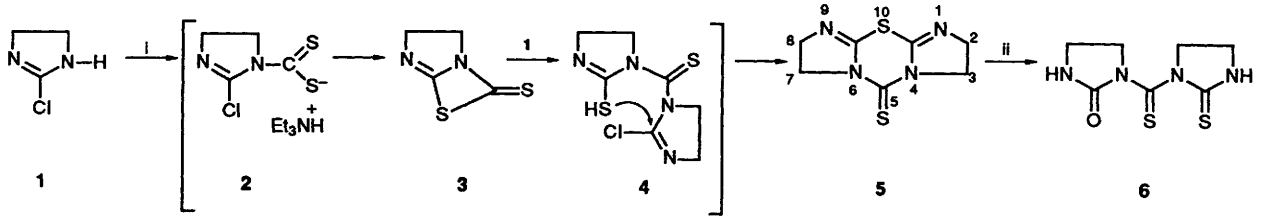
Examination of the reactivity of thiadiazine **5** revealed its relative inertness to basic hydrolysis with 5% NaOH. Acid hydrolysis, however, with 10% hydrochloric acid at 20 °C gave a high yield of imidazolidine-2-thione **6** as a result of C–S bond cleavage.

The structural assignment of compound **6** is based on the following evidence: the mass spectrum exhibits a molecular ion peak at *m/z* 230 (30.2%); in the IR spectrum, the N–H and C=O absorptions are observed at ν_{max} 3215 and 1730 cm^{-1}, respectively; and the ^{13}C NMR spectrum shows three peaks due to trigonal carbon atoms at δ 179.5 (C=S), 179.0 (C=S) and 152.5 (C=O).

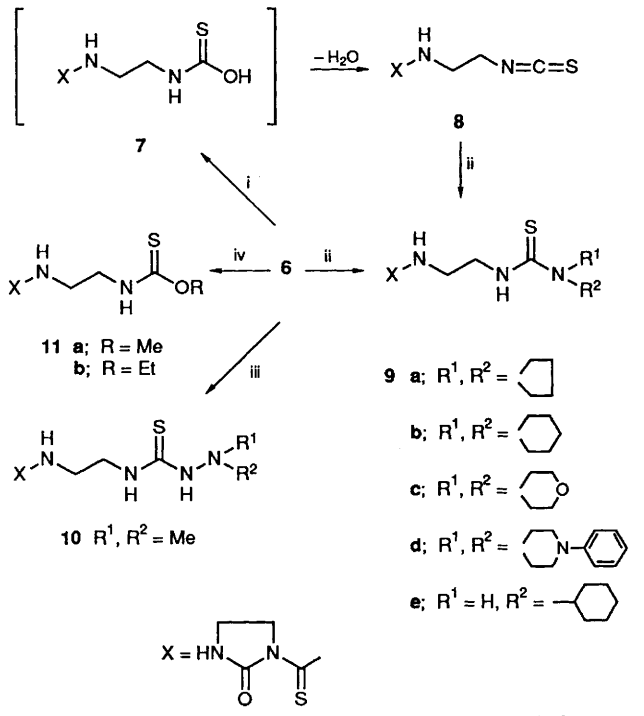
Compound **6** was applied to the synthesis of variously functionalized ethylenediamines **9**, **10** and **11** (Scheme 2). The method is based on our finding that **6**, which can be considered a dithiotriuret derivative, reacted selectively with nucleophilic reagents to give the products of the nucleophilic ring opening of the imidazolidine-2-thione moiety. Thus, refluxing of **6** in water for 10 min led to the formation of isothiocyanate **8** *via* elimination of water from the initially formed thiocarbamic acid **7**.

In the IR spectrum of **8**, characteristic bands for the N=C=S group are present (ν_{max} 2120 and 2190 cm^{-1}) together with a band at ν_{max} 1725 cm^{-1} assignable to the C=O group.

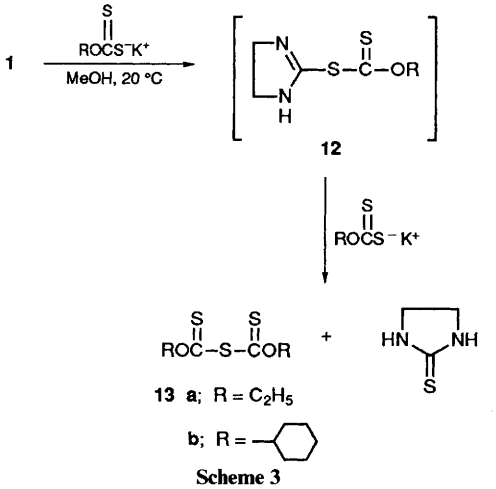
The reaction of **6** with primary and secondary amines, as well as *N*,*N*-dialkylhydrazines carried out in boiling ethanol afforded thioureas **9** and thiosemicarbazides **10**. In a related reaction compound **6** on treatment with sodium alkoxide at 20 °C was transformed into the thiocarbonate **11**.



Scheme 1 *Reagents and conditions:* i, CS_2/Et_3N, CH_2Cl_2, 20 °C; ii, 10% HCl, 20 °C



Scheme 2 *Reagents and conditions:* i, H_2O, reflux; ii, R^1R^2NH; iii, $R^1R^2NNH_2$; iv, RO^-Na^+



Scheme 3

The spectral properties of compounds **9**, **10** and **11** are in full accord with the assigned structures. Moreover, the reactions of isothiocyanate **8** with suitable amines gave the products **9** with identical spectroscopic and physicochemical characteristics to those obtained from **6**.

We further investigated the reaction of **1** with potassium *O*-alkyl dithioarbonates (xanthates) and found that it provides a versatile method for preparation of the diesters of thiodicarbonic acid of type **13** (Scheme 3).

The reaction involves formation of the unstable 2-imidazoline derivative **12**, which *in situ* undergoes reaction with a second molecule of the xanthate to give products **13**. The best results were obtained when the hemisulfate of **1** was treated with a three-fold excess of the xanthate in methanol at room temperature for 0.5 h.

Compounds of type **13** are the well known thioacylating agents.[4,5]

Experimental

Melting points were determined with a Büchi capillary apparatus and are uncorrected. Instrumentation: ^1H NMR, Varian VXR 300 instrument at 300 MHz using tetramethylsilane as the internal standard; ^{13}C NMR, Varian XL 200 instrument using the resonace of solvent for callibration; MS, LKB 900 S instrument (electron impact at 70 eV); IR, Specord M-80 instrument.

2,3,7,8-*Tetrahydro*-5H-*diimidazo*[2,1-b:1′,2′-e][1,3,5]*thiadiazine*-5-*thione* **5**.—To a solution of **1**[6] (5 g, 0.05 mol) in CH_2Cl_2 (60 cm^3), carbon disulfide (6 cm^3, 0.1 mol) and triethylamine (6.9 cm^3, 0.05 mol) were added. The mixture was kept at room temperature for 48 h, after which the triethylamine hydrochloride that had precipitated was filtered off using suction. The filtrate was evaporated to dryness and the residue obtained was washed with water and Et_2O. The resulting crude compound **5** was purified by recrystallization from benzene (2.8 g, 54%), m.p. 191–192 °C (Found: C, 39.9; H, 3.7; N, 26.2. $C_7H_8N_4S_2$ requires C, 39.6; H, 3.8; N, 26.4%); $\delta_H[(CD_3)_2SO]$ 4.0 (m, 4 H) and 4.25 (m, 4 H); $\delta_C[(CD_3)_2SO]$ 51.1, 53.0, 145.6 and 167.9; *m/z* 212 (M^+, 45%).

1-[(2-*Thioxoimidazolidin*-1-*yl*)*thiocarbonyl*]*imidazolidin*-2-*one* **6**.—Compound **5** (5 g, 23 mmol) was dissolved in 10% hydrochloric acid (50 cm^3) and the reaction mixture was stirred at room temperature for 0.5 h. The product **6** that precipitated was collected by filtration, washed with water and dried in a dessicator over P_2O_5, yield 5 g (93%), m.p. 161–164 °C (Found: C, 36.8; H, 4.4; N, 24.1. $C_7H_{10}N_4OS_2$ requires C, 36.5; H, 4.4; N, 24.3%); $\delta_H[(CD_3)_2SO]$ 3.4 (m, 4 H) and 4.0 (m, 4 H); $\delta_C[(CD_3)_2SO]$ 36.0, 41.1, 48.6, 52.4, 152.5, 179.0 and 179.5; ν_{max}/cm^{-1} 3215 (NH), 1730 (C=O), 1535, 1350, 1245 and 1215; *m/z* 230 (M^+, 30), 171 (48), 170 (20), 145 (100), 129 (58) and 112 (35).

2-[(2-*Oxoimidazolidin*-1-*yl*)*thiocarbonylamino*]*ethyl Isothiocyanate* **8**.—Compound **6** (1 g, 4.3 mmol) was suspended in H_2O (20 cm^3) and the mixture was refluxed for 10 min. The insoluble material was filtered off, and the filtrate cooled to room temperature. The solid that precipitated was collected by suction to give compound **8** (0.7 g, 70%), m.p. 109–111 °C (Found: C, 36.4; H, 4.7. $C_7H_{10}N_4OS_2$ requires C, 36.5; H, 4.4%); ν_{max}/cm^{-1} 3175 (N–H), 2190, 2120 (N=C=S), 1725 (C=O), 1535, 1390, 1345 and 1265; $\delta_H(CDCl_3)$ 3.45 (t, 2 H), 3.9 (m, 4 H) and 4.25 (t, 2 H).

Reactions of **6** *with Aliphatic Amines and* N,N-*Dialkylhydrazines. General Procedure*.—Compound **6** (1 g, 4.3 mmol) and suitable amine or hydrazine (6 mmol) were heated in boiling ethanol (10 cm^3) for 0.5 h. Then the reaction mixture was cooled to 5 °C and the resulting product **9** or **10** was separated by suction filtration and purified by recrystallization.

According to the above procedure the following were prepared.

1-{2-[(2-*Oxoimidazolidin*-1-*yl*)*thiocarbonylamino*]*ethylthiocarbamoyl*}*pyrrolidine* **9a**.—This was prepared from **6** and pyrrolidine in 92% yield, m.p. 198–200 °C (methanol) (Found: C, 43.5; H, 6.1. $C_{11}H_{19}N_5OS_2$ requires C, 43.8; H, 6.3%); ν_{max}/cm^{-1} 3185, 1725 (C=O), 1550, 1470, 1400 and 1265; $\delta_H[(CD_3)_2SO]$ 2.0 (m, 4 H), 3.6 (m, 6 H), 3.8 (m, 4 H), 4.2 (t, 2 H), 7.5 (s, 1 H, N–H), 8.1 (s, 1 H, N–H) and 10.5 (s, 1 H, N–H); *m/z* 301 (M^+, 15%), 230(16), 171(48), 170(11), 145(100), 129(13), 71(25) and 70(39).

1-{2-[(2-*Oxoimidazolidin*-1-*yl*)*thiocarbonylamino*]*ethylthiocarbamoyl*}*piperidine* **9b**.—This was prepared from **6** and piperidine in 71% yield, m.p. 182–184 °C (methanol) (Found: C, 49.5; H, 6.7. $C_{12}H_{21}N_5OS_2$ requires C, 45.7; H, 6.7%);

Table 1 Crystal data for compound **5**

Formula	$C_7H_8N_4S_2$
M	212.29
Crystal system	monoclinic
Space group	$P2_1/a$
Unit cell parameters	
a	8.230(1) Å
b	14.634(2) Å
c	8.254(1) Å
β	116.80(1) Å
v	4
Z	1.59 g cm^{-3}
μ(Cu-Kα)	50.1 cm^{-1}

ν_{max}/cm^{-1}, 3185, 1725 (C=O), 1550, 1455, 1390, 1310 and 1265; δ_H[$(CD_3)_2SO$] 1.7 (m, 6 H), 3.5 (t, 2 H), 3.9 (m, 8 H), 4.2 (t, 2 H), 7.9 (s, 1 H, NH), 8.1 (s, 1 H, N–H) and 10.6 (s, 1 H, N–H).

4-{2-[(2-*Oxoimidazolidin*-1-*yl*)*thiocarbonylamino*]*ethylthiocarbamoyl*}*morpholine* **9c**.—This was prepared from **6** and morpholine in 52% yield, m.p. 185–187 °C (methanol) (Found: C, 41.4; H, 6.2; $C_{11}H_{19}N_5O_2S_2$ requires C, 41.6; H, 6.0%). ν_{max}/cm^{-1} 3230, 1725 (C=O), 1630, 1550, 1395 and 1265; δ_H[$(CO_3)_2SO$] 3.4 (m, 4 H), 3.8 (m, 10 H), 4.2 (t, 2 H), 8.0 (s, 1 H, N–H), 8.1 (s, 1 H, N–H) and 10.5 (s, 1 H, N–H).

1-{2-[(2-*Oxoimidazolidin*-1-*yl*)*thiocarbonylamino*]*ethylthiocarbamoyl*}-4-*phenylpiperazine* **9d**.—This was prepared from **6** and *N*-phenylpiperazine in 51% yield, m.p. 195–197 °C (ethanol) (Found: C, 52.2; H, 6.5; $C_{17}H_{24}N_6OS_2$ requires C, 52.0; H, 6.2%). ν_{max}/cm^{-1} 3185, 1725 (C=O), 1600, 1545, 1390, 1315 and 1265; δ_H[$(CD_3)_2SO$] 3.3 (m, 6 H), 3.5–4.2 (m, 10 H), 7.0 (m, 3 H), 7.35 (m, 2 H), 8.0 (s, 2 H, N–H) and 10.5 (s, 1 H, N–H).

1-*Cyclohexyl*-3-{2-[(2-*oxoimidazolidin*-1-*yl*)*thiocarbonylamino*]*ethyl*}*thiourea* **9e**.—This was prepared from **6** and cyclohexylamine in 48% yield, m.p. 197–199 °C (DMF–H_2O) (Found: C, 47.1; H, 7.1; $C_{13}H_{23}N_5OS_2$ requires C, 47.4; H, 7.0%). ν_{max}/cm^{-1} 3215, 1725 (C=O), 1535, 1470, 1390, 1265, 1230 and 1170; δ_H[$(CD_3)_2SO$] 1.3–2.3 (m, 10 H), 3.6 (m, 3 H), 3.8 (m, 4 H), 4.25 (m, 2 H), 7.6 (s, 2 H, N–H), 8.1 (s, 1 H, N–H) and 10.7 (s, 1 H, N–H).

1,1-*Dimethyl*-4-{2-[(2-*oxoimidazolidin*-1-*yl*)*thiocarbonylamino*]*ethyl*}*thiosemicarbazide* **10**.—This was prepared from **6** and *N*,*N*-dimethylhydrazine in 71% yield, m.p. 168–170 °C (ethanol). (Found: C, 37.0; H, 6.0. $C_9H_{18}N_6OS_2$ requires C, 37.2; H, 6.2%). ν_{max}/cm^{-1} 3215, 1715 (C=O), 1550, 1390, 1265 and 1210; δ_H[$(CD_3)_2SO$] 2.6 (s, 6 H), 3.5 (m, 2 H), 3.8 (m, 4 H), 4.25 (m, 2 H), 8.1 (s, 1 H, N–H), 8.25 (s, 1 H, N–H), 8.8 (s, 1 H, N–H) and 10.5 (s, 1 H, N–H); *m*/*z* 290 (M^+, 56%), 172 (22), 171 (69), 170 (16), 146 (23), 145 (56), 103 (20), 102 (43) and 60 (100).

O-*Methyl* N-{2-[(2-*Oxoimidazolidin*-1-*yl*)*thiocarbonylamino*]*ethyl*}*thiocarbamate* **11a**.—Compound **6** (1 g, 4.3 mmol) was added to a solution of sodium methoxide (0.54 g, 10 mmol) in methanol (10 cm^3) and the reaction mixture was stirred at room temperature for 10 min. Water (10 cm^3) was then added and the solution was neutralized with 10% hydrochloric acid. After being cooled to 5 °C, the solid that precipitated was collected by suction and recrystallized from methanol (0.9 g, 79%), m.p. 163–165 °C (ethanol) (Found: 36.3; H, 5.1. $C_8H_{14}N_4O_2S_2$ requires C, 36.6; H, 5.4%); ν_{max}/cm^{-1} 3260, 1700, 1535, 1375, 1330, 1265 and 1185; δ_H[$(CD_3)_2SO$] 3.2–3.9 (m, 6 H), 3.6 (s, 3 H, OCH_3) and 4.4 (m, 2 H); *m*/*z* 262 (M^+, 100%), 161 (34), 103 (20), 102 (66), 88 (24) and 85 (18).

Table 2 Fractional atomic co-ordinates with esd's in parentheses

Atom	*x*/*a*	*y*/*b*	*z*/*c*
S(1)	−0.2654(2)	0.4910(1)	0.0118(2)
S(2)	0.0651(2)	0.2078(1)	0.2588(2)
C(2)	−0.0492(9)	0.4748(4)	0.1950(9)
N(3)	0.0392(7)	0.5412(4)	0.2885(8)
C(4)	0.2097(10)	0.5032(5)	0.4318(10)
C(5)	0.1982(9)	0.3995(4)	0.4153(8)
N(6)	0.0240(7)	0.3876(3)	0.2450(7)
C(7)	−0.0401(8)	0.3056(4)	0.1670(8)
N(8)	−0.1937(6)	0.3072(3)	0.0051(6)
C(9)	−0.2878(8)	0.2242(4)	−0.0979(8)
C(10)	−0.4425(8)	0.2668(5)	−0.2661(8)
N(11)	−0.4499(6)	0.3639(3)	−0.2267(6)
C(12)	−0.3080(8)	0.3812(4)	−0.0797(8)

O-*Ethyl* N-{2-[(2-*oxoimidazolidin*-1-*yl*)*thiocarbonylamino*]*ethyl*}*thiocarbamate* **11b**.—This compound was prepared from **6** and sodium ethoxide according to the procedure described for **11a**, yield 66%, m.p. 149–151 °C (methanol–water) (Found: C, 38.8; H, 5.5. $C_9H_{16}N_4O_2S_2$ requires C, 39.1; H, 5.8%); ν_{max}/cm^{-1} 3310, 1700, 1525, 1430, 1375, 1330, 1265 and 1185; δ_H[$(CD_3)_2SO$] 1.25 (t, 3 H), 3.2–3.8 (m, 6 H), 4.1 (q, 2 H) and 4.4 (m, 2 H).

Thiodicarbonate O,O-*Diethyl Ester* **13a**.—To a solution of potassium *O*-ethyl dithiocarbonate (12.3 g, 75 mmol) in methanol (50 cm^3) was added hemisulfate of compound **1** (5 g, 25 mmol) and the reaction mixture was stirred at room temperature for 0.5 h. Water (50 cm^3) was added and the crude product that precipitated was separated by suction filtration. Recrystallization from methanol gave 4.4 g (91%) of **13a**, m.p. 52 °C (lit.,[4] 52–53 °C); δ_H($CDCl_3$) 1.5 (t, 6 H) and 4.65 (q, 4 H).

Thiodicarbonate O,O-*Dicyclohexyl Ester* **13b**.—This was prepared from hemisulfate of compound **1** and potassium *O*-cyclohexyl dithiocarbonate according to the procedure described for **13a**, yield 85%, m.p. 78–79 °C (methanol) (Found: C, 52.7; H, 6.6. $C_{14}H_{22}O_2S_3$ requires C, 52.8; H, 6.9%); δ_H($CDCl_3$) 1.2–1.5 (m, 6 H), 1.55–1.7 (m, 5 H), 1.8 (m, 4 H), 2.1 (m, 4 H) and 5.5 (m, 2 H); δ_C($CDCl_3$) 23.8, 25.1, 30.9, 84.3 and 204.5; *m*/*z* 318 (M^+, 2%), 236 (11), 177 (20), 95 (17), 84 (53), 82 (100) and 80 (50).

Crystal Structure Analysis of Compound **5**.—Colourless crystals obtained from methanol solution were twinned plates. A single crystal of dimensions 0.5 × 0.5 × 0.05 mm was cut from a twinned specimen. The lattice parameters given in Table 1 were determined by a least-squares fitting of the setting angles of 15 reflections (2θ in the range 17 to 27°). Intensities of reflections were measured on a Syntex $P2_1$ diffractometer with graphite monochromatized Cu-Kα radiation (λ = 1.541 78 Å) to $2\theta_{max}$ = 115°. Lorentz and polarization corrections were applied to the net intensities, absorption was ignored. Integrated intensities were obtained by peak profile analysis according to Lehmann and Larsen.[7] No significant intensity variation was observed for two standard reflections. Out of 1361 measured reflections 1092 had $I > 2\sigma(I)$ and they were considered observed. The structure was solved straightforwardly by direct methods with program MULTAN80.[8] The positions and anisotropic thermal parameters of non-hydrogen atoms were refined by full-matrix least-squares method. The positions of the hydrogen atoms were calculated from

Table 3 Bond distances and angles with esd's in parentheses

	Distance/Å		Bond angles/°
S(1)–C(2)	1.756(6)	C(2)–S(1)–C(12)	100.2(3)
S(2)–C(7)	1.668(6)	S(1)–C(2)–N(3)	120.6(6)
C(2)–N(6)	1.392(8)	C(2)–N(3)–C(4)	106.3(6)
C(4)–C(5)	1.523(9)	C(4)–C(5)–N(6)	101.2(5)
N(6)–C(7)	1.352(7)	C(5)–N(6)–C(7)	123.6(5)
N(8)–C(9)	1.485(7)	S(2)–C(7)–N(6)	122.3(5)
C(9)–C(10)	1.531(8)	S(2)–C(7)–N(8)	121.5(5)
N(11)–C(12)	1.274(6)	C(7)–N(8)–C(9)	124.1(5)
S(1)–C(12)	1.743(6)	N(8)–C(9)–C(10)	101.1(5)
C(2)–N(3)	1.250(8)	C(10)–N(11)–C(12)	107.0(5)
N(3)–C(4)	1.477(8)	S(1)–C(12)–N(11)	121.0(5)
C(5)–N(6)	1.498(7)	S(1)–C(2)–N(6)	121.0(5)
C(7)–N(8)	1.364(6)	N(3)–C(2)–N(6)	118.4(6)
N(8)–C(12)	1.398(7)	N(3)–C(4)–C(5)	107.8(6)
C(10)–N(11)	1.465(9)	C(2)–N(6)–C(5)	106.0(5)
		C(2)–N(6)–C(7)	130.5(6)
		N(6)–C(7)–N(8)	116.2(5)
		C(7)–N(8)–C(12)	128.4(5)
		C(9)–N(8)–C(12)	106.7(5)
		C(9)–C(10)–N(11)	107.1(5)
		N(8)–C(12)–N(11)	116.4(5)
		C(1)–C(12)–N(8)	122.6(5)

geometrical conditions [d(C–H) 1.08 Å]. The H atoms contributed to F_c but their parameters were not refined. Empirical isotropic extinction parameter x was used to correct F_c according to $F' = F_c(1 - xF_c^2/\sin\theta)$; x converged at $2(1) \times 10^{-6}$. Final values of R and R_w are 0.091 and 0.111, respectively. The weights were $w = 1/[\sigma^2(F_o) + 0.000\,05F_o^2]$. The highest peak in the final difference map of 1.21 e Å^{-3} was located near S(2), the lowest peak was −0.67 e Å^{-3}. The scattering factors used in calculations were those included in SHELX76.[9] Calculations were performed using programs: SHELX76 for the refinement of the structure, ORTEP[10] for drawings and programs written by Jaskólski[11] for molecular geometry analysis.

Final positional parameters of **5** are given in Table 2, molecular dimensions in Table 3. Hydrogen atom co-ordinates and thermal parameters are deposited at the CCDC.*

* See Instructions for Authors, *J. Chem. Soc., Perkin Trans. 1*, 1991, Issue 1.

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Paper 1/04133I
Received 7*th August* 1991
Accepted 11*th September* 1991