Synthesis and Properties of 1,4-Dimethylpyridazino[4,5-*d*]pyridazines (1)

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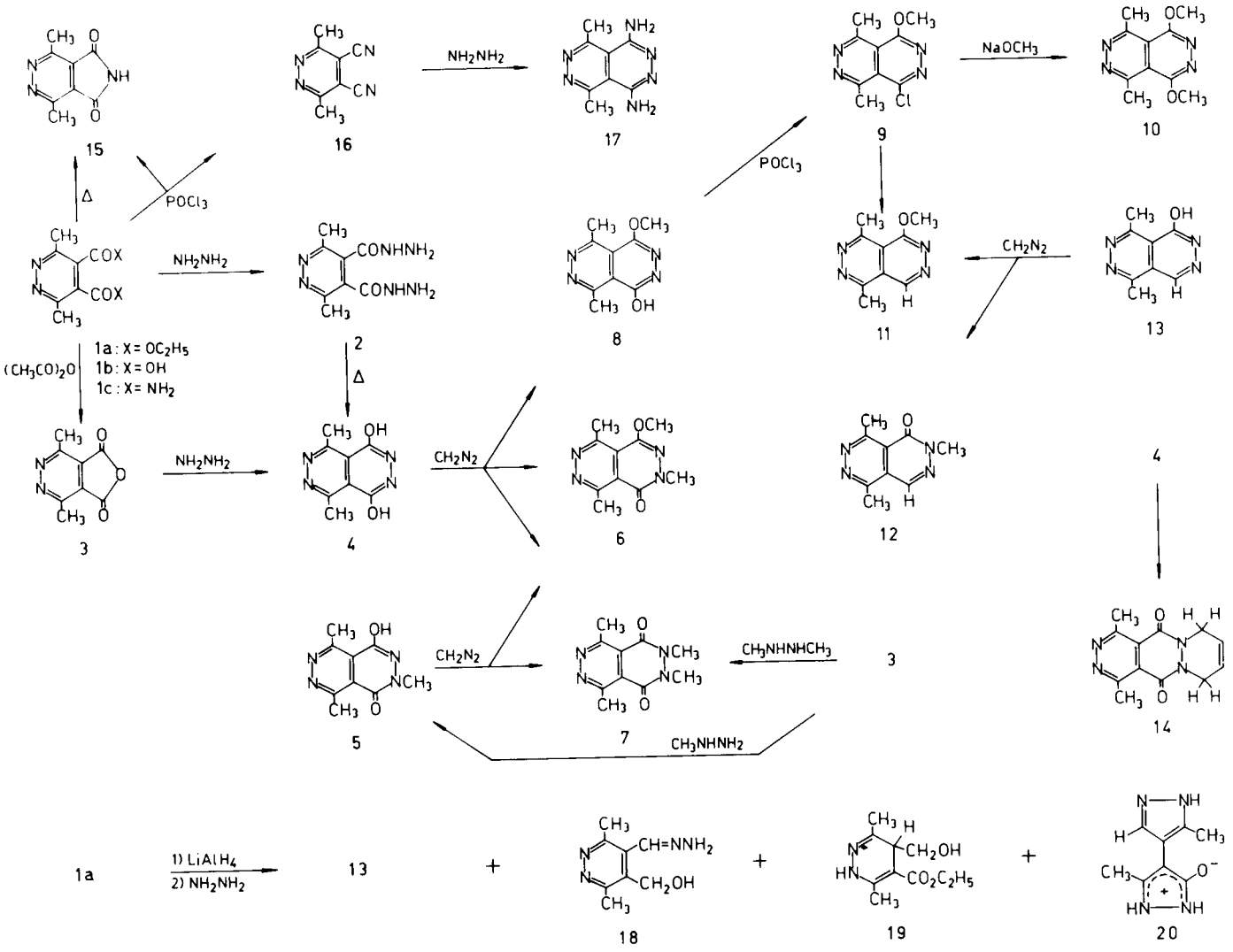
The synthesis and properties of several 1,4-dimethylpyridazino[4,5-*d*]pyridazines are described. Treatment of diethyl 3,6-dimethylpyridazine-4,5-dicarboxylate (**1a**) with lithium aluminium hydride and hydrazine did not afford the expected 1,4-dimethylpyridazino[4,5-*d*]pyridazine (**21b**) but a mixture of compounds **13**, **18**, **19**, and **20**, whose structures were deduced from spectroscopic data.

As a part of our research (2a-e) on the pyridazino[4,5-*d*]-pyridazine system, this paper reports the synthesis of several 1,4-dimethyl derivatives.

1,4-Dihydroxy-5,8-dimethylpyridazino[4,5-*d*]pyridazine (**4**) was prepared by a modification of Jones' method (3) which gave in our hands unsatisfactory results; con-



densation of the ester (**1a**) with hydrazine hydrate at room temperature afforded the dihydrazide (**2**) which cyclized to **4** by heating at 180-190°. Compound **4** was also obtained in quantitative yield by refluxing 3,6-dimethylpyridazine-4,5-dicarboxylic acid anhydride (**3**) with anhydrous hydrazine in glacial acetic acid.

The anhydride (**3**) was synthesised from the acid (**1b**) with boiling acetic anhydride.

When compound **4** was allowed to react with an excess of diazomethane for 24 hours, a mixture of 1-methoxy-3,5,8-trimethyl-3*H*-pyridazino[4,5-*d*]pyridazin-4-one (**6**) and 2,3,5,8-tetramethyl-2*H*,3*H*-pyridazino[4,5-*d*]pyridazine-1,4-dione (**7**) with a small amount of 1-methoxy-4-hydroxy-5,8-dimethylpyridazino[4,5-*d*]pyridazine (**8**) was obtained. The yield of **8** could be improved if the reaction was carried out with two moles of the alkylating agent and stopped after 2 hours. The structures of these compounds followed from chemical evidence. The methoxy derivative (**8**) was different from the isomer **5**, prepared by condensation of **3** with methylhydrazine; methylation of **5** with diazomethane yielded a mixture of **6** and **7**, the latter being also obtained by treatment of **3** with *N*,*N*′-dimethylhydrazine.

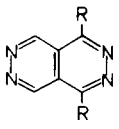
Chlorination of **8** with phosphorus oxychloride afforded 1-methoxy-4-chloro-5,8-dimethylpyridazino[4,5-*d*]pyridazine (**9**) which was converted into **10** by reaction with sodium methoxide. Catalytic dehalogenation of **9** gave 1-methoxy-5,8-dimethylpyridazino[4,5-*d*]pyridazine (**11**).

Compound **4** reacted with lead tetraacetate in the presence of butadiene to yield the adduct **14** whose structure followed from spectroscopic evidence. The ir spectrum shows a strong band at 1635 cm^{-1} for the carbonyl groups and the nmr spectrum exhibits two multiplets at δ 6.08 (2H) and 4.6 (4H) ppm and a singlet at δ 3.17 (6H) ppm attributable to the $-CH_2-CH=CH-CH_2-$ system and to the ring methyl groups respectively.

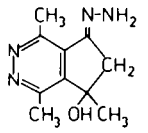
Despite the unsuccessful attempts reported by Bilton and Linstead (4) to dehydrate the dicarboxamide (**1c**) into 3,6-dimethyl-4,5-dicyanopyridazine (**16**), the latter was obtained in 50% yield by refluxing **1c** with phosphorus oxychloride. From this reaction we also isolated a small amount of the known imide (**15**) (4,5) which could be prepared in high yield by cyclization of **1c** in boiling hexamethylphosphoric triamide.

The dinitrile (**16**) reacted with hydrazine at room temperature to give 1,4-diamino-5,8-dimethylpyridazino[4,5-*d*]pyridazine (**17**).

Attempts to prepare 1,4-dimethylpyridazino[4,5-*d*]-pyridazine (**21b**) by the method employed for the parent compound (**21a**) (2a) were unsuccessful. When the ester (**1a**) was treated with lithium aluminium hydride in anhydrous tetrahydrofuran at -70°, and the reduction products allowed to react with hydrazine, compounds **19** and **20** were isolated as the main products with lesser amounts of the hydrazone (**18**) and 1-hydroxy-5,8-dimethylpyridazino-[4,5-*d*]pyridazine (**13**). The structure of **13** was deduced from spectroscopic data (2e) and confirmed by its conversion into a mixture of *N*-methyl- (**12**) and *O*-methyl- (**11**) derivatives by treatment with diazomethane.



21a : R=H
21b: R=CH_3



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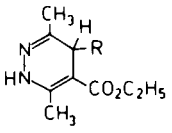

23a: R=$CO_2C_2H_5$
23b: R=CHO

The uv spectrum of **18** with maxima at 209, 235, and 310 nm closely resembles that of the hydrazone (**22**) (2c); the nmr spectrum (see experimental) strongly supports the assigned structure.

Compound **19** for which nine tautomeric structures could be taken into consideration, was formulated as ethyl 1,4-dihydro-3,6-dimethyl-4-hydroxymethylpyridazine-5-carboxylate on spectral evidence. The ir spectrum in carbon tetrachloride shows two bands at 3620 (OH) and 3435 (NH) cm^{-1} and a strong band at 1690 cm^{-1} attributable to a conjugated ester carbonyl group; the uv spectrum showing two maxima at 234 and 326 nm, is nearly identical to that of the known ester (**23a**) (6). The nmr spectrum in deuteriochloroform exhibits two exchangeable peaks at δ 7.56 (NH) and 2.40 (OH) ppm and a structured singlet at δ 3.53 (3H) ppm due to the proton and the methylene group at 4-position; this signal changes into a well resolved AB_2 pattern when the spectrum is recorded in hexadeuteriodimethylsulfoxide in the presence of deuterium oxide.

On the grounds of the known conversion of the dihydro-ester (**23a**) into 3,3′-dihydroxy-5,5′-dimethyl-4,4′-bispyrazole by treatment with hydrazine (6,7), the formation of compound **20** could be rationalized by the action of the same reagent on the intermediate formyl derivative **23b**.

Although the structure of **20** was definitively established by an X-ray analysis (8), its zwitterionic character could be deduced from the ir spectrum showing a broad band between 3300 and 2000 cm^{-1} (maximum at 2700 cm^{-1}) and a strong band at 1595 cm^{-1} for the NH^+ and $C\text{-}O^-$ groups respectively.

EXPERIMENTAL

All melting points are uncorrected. Unless otherwise stated, the infrared spectra were measured for potassium bromide discs with a Perkin-Elmer 457 Spectrometer and the ultraviolet spectra were taken in methanol with a Cary 14 Spectrophotometer. ^1H nmr spectra were recorded with a Varian A-56/60 instrument; chemical shifts are reported in ppm downfield from internal tetramethylsilane. Silica gel plates (Merck F_{254}) were used for analytical and preparative tlc. Light petroleum refers to the fraction boiling at 75-120°. For the spectroscopic data of compounds **4-13** see reference (2e).

3,6-Dimethylpyridazine-4,5-dicarboxylic Acid Dihydrazide (**2**).

A solution of diethyl 3,6-dimethylpyridazine-4,5-dicarboxylate (**1a**) (1 g.) in methanol (10 ml.) was treated with 98% hydrazine hydrate (0.6 ml.) and left overnight at room temperature. Removal of the solvent gave a yellow solid which was treated with water (5 ml.) and filtered to yield the dihydrazide (**2**) as its monohydrate (0.76 g., 79%). An analytical sample obtained by crystallization from water melted between 160 and 180°, solidified again at higher temperature and then decomposed above 300°; ir: 3500, 3320, 3240, 1675, 1620, 1570, 1545, and 1400 cm^{-1}; nmr (DMSO-d_6 + deuterium oxide): δ 2.57 (s, 6, 2 x CH_3).

Anal. Calcd. for $C_8H_{12}N_6O_2.H_2O$: C, 39.67; H, 5.83; N, 34.69. Found: C, 39.53; H, 5.88; N, 34.74.

3,6-Dimethylpyridazine-4,5-dicarboxylic Acid Anhydride (**3**).

3,6-Dimethylpyridazine-4,5-dicarboxylic acid monohydrate (**1b**) (5 g.) was dissolved in boiling acetic acid anhydride (500 ml.) and the resulting solution was treated with charcoal and filtered hot. On cooling, compound **3** separated as yellow plates (3.45 g., 82%), m.p. 233-235° dec., (after a further crystallization from acetic anhydride); ir (nujol); 1860, 1845, 1797, and 1400 cm^{-1}; uv (chloroform): 304 (log ε 3.58) and 312 sh (log ε 3.5) nm.

Anal. Calcd. for $C_8H_6N_2O_3$: C, 53.94; H, 3.39; N, 15.72. Found: C, 53.99; H, 3.38; N, 15.86.

1,4-Dihydroxy-5,8-dimethylpyridazino[4,5-*d*]pyridazine (**4**).

A) A mixture of **3** (3.45 g.) and anhydrous hydrazine (3.2 ml.) in glacial acetic acid (180 ml.) was refluxed for 1 hour and allowed to stand overnight to give compound **4** (3.15 g.) which was separated by filtration, washed with ethyl ether and dried. Evaporation of the acetic mother liquors left a solid residue which was treated with water and filtered to yield a second crop of the dihydroxy derivative **4** (0.52 g., quantitative yield); it darkened above 300° and decomposed between 320 and 330° (after crystallization from water) [lit (3) 320° dec.].

Anal. Calcd. for $C_8H_8N_4O_2$: C, 50.00; H, 4.20; N, 29.15. Found: C, 49.88; H, 4.20; N, 29.11.

B) The dihydrazide **2** (0.15 g.) was heated at 180-190° for 3-4 hours to give a yellow solid (0.11 g.) identical (m.p. and ir spectrum) to material prepared as above.

2,5,8-Trimethyl-2*H*,3*H*-pyridazino[4,5-*d*]pyridazine-1,4-dione (**5**).

A mixture of the anhydride **3** (1.33 g.) and methyl hydrazine (1.68 ml.) in glacial acetic acid (50 ml.) was refluxed with stirring for 1 hour. Removal of the solvent left a solid residue which was treated with water (20 ml.) and filtered to give the dione (**5**) (0.85 g.); acidification of the mother liquors with concentrated hydrochloric acid (*p*H 1-2) precipitated a further 0.05 g. of **5** (overall yield 58%), m.p. 280° dec. (from water).

Anal. Calcd. for $C_9H_{10}N_4O_2$: C, 52.42; H, 4.89; N, 27.17. Found: C, 52.65; H, 4.77; N, 27.38.

2,3,5,8-Tetramethyl-2*H*,3*H*-pyridazino[4,5-*d*]pyridazine-1,4-dione (**7**).

Compound **3** (0.5 g.) and *N*,*N*'-dimethylhydrazine (1 ml.) were refluxed in glacial acetic acid (50 ml.) for 2 hours. The semi-solid residue left after removal of the solvent was kept for a long time in a vacuum dessiccator (sulfuric acid and potassium hydroxide), treated with water and extracted with chloroform (4 x 30 ml.). Evaporation of the combined extracts afforded a yellow solid which was sublimed at 120° and 0.02 mm Hg to give the tetramethyl derivative (**7**) (0.32 g., 51.5%), m.p. 193-194° (from ethyl acetate).

Anal. Calcd. for $C_{10}H_{12}N_4O_2$: C, 54.54; H, 5.49; N, 25.44. Found: C, 54.49; H, 5.47; N, 25.24.

Methylation of Compound **4** with Diazomethane.

A) A suspension of **4** (0.3 g., 1.563 mmoles) in ethyl ether (30 ml.) and methanol (5 ml.) was treated with ethereal diazomethane (0.33 g., 7.85 mmoles) and allowed to stand for 24 hours. The reaction mixture was filtered to give 1-methoxy-4-hydroxy-5,8-dimethylpyridazino[4,5-*d*]pyridazine (**8**) (0.02 g.), m.p. 262-263° dec. (from methanol).

Anal. Calcd. for $C_9H_{10}N_4O_2$: C, 52.42; H, 4.89; N, 27.17. Found: C, 52.29; H, 4.93; N, 29.96.

Evaporation of the filtrate gave a yellow-brown solid which was resolved into two components by preparative layer chromatography with chloroform as developer.

The faster running band gave 1-methoxy-3,5,8-trimethyl-3*H*-pyridazino[4,5-*d*]pyridazin-4-one (**6**) (0.2 g.), m.p. 160-161° (after sublimation at 60-70° and 0.05 mm Hg and crystallization from cyclohexane).

Anal. Calcd. for $C_{10}H_{12}N_4O_2$: C, 54.54; H, 5.49; N, 25.44. Found: C, 54.32; H, 5.54; N, 25.21.

The second band yielded a yellow solid which was sublimed at 100° and 0.03 mm Hg to give compound **7** (0.025 g.) identical (m.p. and ir spectrum) with material prepared as above.

B) A suspension of **4** (0.5 g., 2.6 mmoles) in ethyl ether (50 ml.) and methanol (10 ml.) was treated with ethereal diazomethane (0.22 g., 5.2 mmoles) and allowed to stand for 2 hours. The solid obtained by filtration of the reaction mixture was washed with benzene and treated with hot chloroform; the unreacted starting material (0.2 g.) was filtered off and the chloroform solution was evaporated to dryness to give the methoxy derivative (**8**) (0.07 g.).

Methylation of Compound **5** with Diazomethane.

Methylation of **5** (0.2 g., 0.97 mmoles) with ethereal diazomethane (0.1 g., 2.38 mmoles) carried out essentially as described for **4** (method A) gave compounds **6** (0.13 g.) and **7** (0.035 g.).

1-Methoxy-4-chloro-5,8-dimethylpyridazino[4,5-*d*]pyridazine (**9**).

Compound **8** (0.15 g.) was added to freshly distilled phosphorus oxychloride (20 ml.) heated at 100° and the mixture was refluxed for 2 hours. Removal of the solvent under reduced pressure left a sticky brown residue which was treated with crushed ice, made weakly alkaline with cold saturated sodium carbonate solution, and extracted with ethyl ether. Evaporation of the dried (sodium sulfate) extracts afforded a yellow-brown solid which was sublimed at 75° and 0.03 mm Hg to give the chloro derivative (**9**) (0.13 g., 80%), m.p. 134-135° (after purification by layer cromatography with chloroform as developer, and crystallization from light petroleum).

Anal. Calcd. for $C_9H_9N_4OCl$: C, 48.10; H, 4.01; N, 24.94; Cl, 15.80. Found: C, 48.40; H, 4.10; N, 24.84; Cl, 16.10.

1,4-Dimethoxy-5,8-dimethylpyridazino[4,5-*d*]pyridazine (**10**).

The chloro derivative (**9**) (0.05 g.) was added to a solution of sodium (0.05 g.) in anhydrous methanol (10 ml.) and refluxed for 1 hour. The residue left after removal of the solvent was treated with water and extracted with chloroform; evaporation of the extracts gave compound **10** (0.045 g., 92%), m.p. 166-167° (after sublimation at 80° and 0.03 mm Hg and crystallization from light petroleum).

Anal. Calcd. for $C_{10}H_{12}N_4O_2$: C, 54.54; H, 5.49; N, 25.44. Found: C, 54.58; H, 5.29; N, 25.52.

1-Methoxy-5,8-dimethylpyridazino[4,5-*d*]pyridazine (**11**).

A solution of **9** (0.075 g.) in ethyl ether (25 ml.) was added to a suspention of 5% palladium charcoal (0.075 g.) in aqueous sodium hydroxide (0.02 *N*, 17 ml.) previously saturated with hydrogen,

and the mixture was hydrogenated at room temperature and atmospheric pressure until 1 mole of the gas had been consumed. The two layers were separated and filtered and the aqueous solution was extracted several times with ethyl ether; the extracts were combined with the ethereal solution, dried (sodium sulfate) and evaporated to dryness. The residual product was sublimed at 80° and 0.05 mm Hg to give **11** (0.03 g., 47%), m.p. 153-154° (after purification by layer chromatography with chloroform-methanol (96:4 v/v) as developer, and crystallization from cyclohexane).

Anal. Calcd. for $C_9H_{10}N_4O$: C, 56.83; H, 5.30; N, 29.46. Found: C, 56.97; H, 5.40; N, 29.8.

1,4-Dimethyl-5,8-dihydro-9,10-dioxo-2,3,5a,9a-tetraazaanthracene (**14**).

A suspension of **4** (0.5 g.) in methylene chloride (20 ml.) containing butadiene (1.37 g.) was treated with lead tetraacetate (85-90%, 1.3 g.) and stirred vigorously at room temperature until no more of the latter was consumed (*ca.* 36 hours). The reaction mixture was filtered and the filtrate was evaporated to give a yellow-brown sticky product which was triturated with ethyl ether; sublimation at 125-130° and 0.04 mm Hg afforded the adduct (**14**) (0.4 g., 63%) as yellow needles, m.p. 196-198° (after several crystallizations from ethyl acetate); uv: 284 (log ε 3.62) and 370 (log ε 3.49) nm.

Anal. Calcd. for $C_{12}H_{12}N_4O_2$: C, 59.01; H, 4.95; N, 22.94. Found: C, 58.90; H, 4.98; N, 23.12.

3,6-Dimethylpyridazine-4,5-dicarboximide (**15**).

A suspension of the carboxamide (**1c**) (0.2 g.) in hexamethylphosphoric triamide (2 ml.) was gently refluxed until evolution of ammonia ceased. The resulting solution was cooled and diluted with water (5 ml.) to give compound **15** (0.15 g., 83%), as yellow plates; an analytical sample obtained by crystallization from methanol, gradually darkened above 260° and largely decomposed at about 303° [lit (4) 240° dec., (5) 290° dec.].

Anal. Calcd. for $C_8H_7N_3O_2$: C, 54.24; H, 3.98; N, 23.72. Found: C, 54.21; H, 3.83; N, 23.58.

3,6-Dimethyl-4,5-dicyanopyridazine (**16**).

A mixture of **1c** (1.5 g.) and freshly distilled phosphorus oxychloride (60 ml.) was stirred at room temperature for 5-6 hours and then refluxed for 24 hours. Removal of the solvent under reduced pressure left a brown residue which was treated with crushed ice, made slightly alkaline with cold saturated sodium carbonate solution and extracted with ethyl ether. Evaporation of the dried (sodium sulfate) extracts, gave a yellow-brown solid which was sublimed at 75° and 0.05 mm Hg to yield compound **16** (0.61 g., 50%), m.p. 178-179° (from carbon tetrachloride); ir: 2240, 1435, 1405, 1105, and 1020 cm^{-1}; uv: 234 (log ε 3.78), 297 (log ε, 3.56), and 305 (log ε 3.54); nmr (deuteriochloroform): δ 3.01 (s, 6, 2 x CH_3).

Anal. Calcd. for $C_8H_6N_4$: C, 60.75; H, 3.82; N, 35.42. Found: C, 60.84; H, 3.84; N, 35.46.

Further sublimation of the reaction product at 130-140° and 0.05 mm Hg afforded the dicarboximide (**15**) (0.07 g.) identical (m.p. and ir spectrum) with material prepared as above.

1,4-Diamino-5,8-dimethylpyridazino[4,5-*d*]pyridazine (**17**).

95% Hydrazine (1.2 ml.) was added to a solution of **16** (0.2 g.) in methanol (10 ml.); the mixture was set aside overnight at room temperature and then kept in the refrigerator for 2 days. The yellow crystalline solid which separated was filtered off, washed with little methanol and dried *in vacuo* (potassium hydroxide) to give the diamino derivative (**17**) (0.17 g.) which gradually darkened above 260° and melted at 288-290° dec. Evaporation of the filtrate afforded a residue which was dried (sulfuric acid and potassium hydroxide), treated with a small amount of methanol and filtered to give a second crop of **17** (0.03 g., overall yield 74%); ir (nujol): 3320, 3110, 2730, 1660, 1615, 1545, 1455, 1410, 1385, 1350, 1060, and 1030 cm^{-1}; uv: 220 (log ε 4.16), 279 (log ε 3.65), and 305 sh (log ε 3.60) nm; nmr (DMSO-d_6 + deuterium oxide): δ 2.69 (s, 3, CH_3), 2.82 (s, 3, CH_3), and 3.2 (s, 3, CH_3OH).

Anal. Calcd. for $C_8H_{10}N_6.CH_3OH$: C, 48.64; H, 6.35; N, 37.81. Found: C, 48.63; H, 6.47; N, 37.43.

Reaction of Compound **4** with Lithium Aluminum Hydride and Hydrazine.

A solution of the ester (**1a**) (10 g.) in anhydrous tetrahydrofuran (50 ml.) was added dropwise under nitrogen to a stirred suspension of lithium aluminum hydride (3.96 g.) in the same solvent (100 ml.) at −70° and stirring was continued at the same temperature for 1.5 hours. The reaction mixture was then hydrolyzed very slowly with aqueous acetic acid (50%; 10 ml.) in tetrahydrofuran (20 ml.) and the suspension was rapidly filtered through a sintered glass funnel into a flask containing 95% hydrazine (4 ml.) in methanol (40 ml.) at −70°. The inorganic material was washed with tetrahydrofuran (3 x 15 ml.) and the washings were collected in the same flask. The solution was then stirred at −70° under nitrogen for 1 hour, allowed to rise slowly to room temperature and set aside overnight. The reaction mixture was filtered and the filtrate evaporated under reduced pressure to give a semi-solid yellow residue which was kept in a vacuum dessiccator (sulfuric acid and potassium hydroxide) for several days and then treated with water (5-10 ml.). The crude product separated by filtration and the solid recovered by concentration of the aqueous mother liquors were combined, dried, and extracted (Soxhlet) with ethyl ether to give ethyl 1,4-dihydro-3,6-dimethyl-4-hydroxymethylpyridazine-5-carboxylate (**19**) (1.9 g.), m.p. 123-125° (after two crystallizations from ethyl acetate).

Anal. Calcd. for $C_{10}H_{16}N_2O_3$: C, 56.59; H, 7.60; N, 13.20. Found: C, 56.54; H, 7.62; N, 13.14.

Further extraction of the residue with chloroform afforded a solid (0.8 g.) which was treated with hot benzene (400 ml.) and filtered to yield the hydrazone of 3,6-dimethyl-4-hydroxymethyl-5-formylpyridazine (**18**) (0.4 g.); m.p. 190° (after treatment with dilute aqueous ammonium hydroxide (5 ml.) and crystallization from methanol); ir: 3320, 3160, 2910, 2840, 2720, 1570, 1520, 1395, 1030, and 1015 cm^{-1}; nmr (DMSO-d_6): δ 7.98 (s, 1, $-CH=N-$), 7.57 (bd s, 2, NH_2), 4.95 (bd t, J 5-6 Hz, 1, OH), 4.57 (bd d, J 5-6 Hz, 2, CH_2), and 2.63 (s, 6, 2 x CH_3).

Anal. Calcd. for $C_8H_{12}N_4O$: C, 53.32; H, 6.71; N, 31.09. Found: C, 53.48; H, 6.74; N, 31.29.

Evaporation of the benzene solution gave 1-hydroxy-5,8-dimethylpyridazino[4,5-*d*]pyridazine (**13**) (0.4 g.) which was purified by treatment with aqueous hydrochloric acid (*p*H 3) and crystallization from methanol, m.p. 271-272°.

Anal. Calcd. for $C_8H_8N_4O$: C, 54.54; H, 4.58; N, 31.80. Found: C, 54.25; H, 4.51; N, 31.71.

The solid recovered from the Soxhlet (1.1 g.) largely consisted of 3-hydroxy-5,5′-dimethyl-4,4′-bis pyrazole (**20**) which crystallized from water as white needles, m.p. > 330°; uv (water): 246 (log ε 3.92) nm; nmr (DMSO-d_6 + deuterium oxide): δ 7.40 (s, 1, CH), 2.17 (s, 3, CH_3), and 2.08 (s, 3, CH_3).

Anal. Calcd. for $C_8H_{10}N_4O$: C, 53.92; H, 5.66; N, 31.44. Found: C, 53.74; H, 5.73; N, 31.46.

Methylation of Compound **13** with Diazomethane.

A suspension of **13** (0.17 g.) in ethyl ether (30 ml.) and methanol (5 ml.) was treated with ethereal diazomethane (0.13 g.) and allowed to stand overnight. Removal of the solvent left a sticky residue which was treated with light petroleum to give a solid (0.16 g.) containing (nmr spectrum) 2,5,8-trimethyl-2*H*-pyridazino[4,5-*d*]pyridazin-1-one (**12**) as main product, with a small amount (*ca.* 10%) of 1-methoxy-5,8-dimethylpyridazino[4,5-*d*]pyridazine (**11**). An analytical sample of **12** was obtained by preparative layer chromatography with ethyl acetate as developer and crystallization from light petroleum, m.p. 158-159°.

Anal. Calcd. for $C_9H_{10}N_4O$: C, 56.83; H, 5.30; N, 29.46. Found: C, 56.70; H, 5.23; N, 29.57.

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