Anal. Calcd for $C_{13}H_{13}N_2I$: C, 48.20; H, 4.01; N, 8.65. Found: C, 48.12; H, 4.16; N, 8.52.

N,N′-Dimethyl-4,4′-pyridocyanine iodide (V) and **N,N′,N″-trimethyl-4,4′,4″-tripyridocyanine (VI)** were synthesized following the procedure of Sprague and Brooker.[16] After two recrystallizations from ethanol dye VI was obtained as dark red crystals which showed green dichroism, yield 41%. The mother liquors were concentrated and filtered from a second crop of VI. The filtrate was chromatographed with acetone–methanol (9:1) on alumina and dye V was eluted in a yellow zone. After concentration of the solution, V was precipitated with ether and recrystallized from a little ethanol, yield *ca.* 5%.

Anal. Calcd for $C_{13}H_{15}N_2I$ (V): C, 47.85; H, 4.60; N, 8.59. Found: C, 47.76; H, 4.99; N, 8.52. Calcd for $C_{19}H_{21}N_3I$ (VI): C, 41.83; H, 3.85; N, 7.71. Found: C, 41.75; H, 4.05; N, 7.62.

N,N′-Diethyl-2,2′-azapyridocyanine Iodide (VII, R = C_2H_5).—Di(2-pyridyl)amine (**12**) (2.0 g, 0.012 mol) and 5.5 g (0.036 mol) of iodoethane were heated to 100° for 4 hr in a sealed glass tube. The product was washed with ether and twice recrystallized from ethanol, yield 2.9 g (70%).

Anal. Calcd for $C_{14}H_{18}N_3I$: C, 47.30; H, 5.07; N, 11.83. Found: C, 47.20; H, 5.10; N, 11.70.

N,N′-Methylene-2,2′-azapyridocyanine Iodide (VIII).—A mixture of 3.0 g (0.018 mol) of di(2-pyridyl)amine (**12**) and 7.0 g (0.026 mol) of diiodomethane was heated to 100° for 4 hr in a sealed glass tube. The reaction product was washed with ether, then extracted with methanol, and chromatographed on alumina, first with acetone–methanol (9:1). The blue fluorescing zone was collected, concentrated, precipitated with ether, and once more chromatographed with acetonitrile, yield 0.6 g (10%).

Anal. Calcd for $C_{11}H_{10}N_3I \cdot H_2O$: C, 40.12; H, 3.65; N, 12.80. Found: C, 40.45; H, 3.50; N, 12.89.

Monoalkylation Product of 2,4′-Dipyridylamine (IX, R = CH_3).—A mixture of 1.0 g (0.006 mol) of 2,4′-dipyridylamine (**13**) and 2.5 g (0.018 mol) of iodomethane was heated to 100° for 3 hr in a sealed glass tube. The reaction product was washed with ether and recrystallized from ethanol, yield 1.5 g (80%). A similar reaction using 2.8 g (0.018 mol) of iodoethane in place of iodomethane yielded 1.5 g (80%) of IX (R = C_2H_5).

Anal. Calcd for $C_{11}H_{12}N_3I$ (R = CH_3): C, 42.17; H, 3.83; N, 13.42. Found: C, 42.15; H, 3.95; N, 13.30.

N,N′-Dimethyl-2,4′-azapyridocyanine Iodide (X).—2,4′-Dipyridylamine (**13**) (1.0 g, 0.006 mol) and 2.2 g (0.018 mol) of dimethyl sulfate were heated until reaction began. After it had subsided, the reaction mixture was dissolved in a little ammonia containing potassium iodide and then chromatographed on alumina with acetone–methanol (1:1). The blue fluorescing zone was collected and concentrated, and the product was precipitated with ether. It was again chromatographed as above, and once more with acetonitrile, yield 0.6 g (30%).

Anal. Calcd for $C_{12}H_{14}N_3I$: C, 44.03; H, 4.28; N, 12.84. Found: C, 44.03; H, 4.44; N, 12.81.

N,N′-Dimethyl-4,4′-azapyridocyanine Iodide (XI).—A mixture containing 1.0 g (0.006 mol) of di(4-pyridyl)amine (**15**) and 2.5 g (0.018 mol) of iodomethane was heated to 100° for 2 hr in a sealed glass tube. The reaction product was washed with ether, dissolved in a little concentrated ammonia, and chromatographed on alumina with acetonitrile. The blue fluorescing zone was collected and concentrated, and the product precipitated with ether. It was then dried *in vacuo* for 12 hr at 100° where it lost acetonitrile that was enclosed in the crystals, yield 1.5 g (80%).

Anal. Calcd for $C_{12}H_{14}N_3I$: C, 44.03; H, 4.28; N, 12.84. Found: C, 43.99; H, 4.32; N, 12.81.

Registry No.—**3**, 1132-37-2; **12**, 1202-34-2; **13**, 33932-96-6; **15**, 1915-42-0; II, 38222-62-7; III bromide, 16521-09-8; III iodide, 38222-64-9; V, 16521-11-2; VI, 16610-36-9; VII (R = Et), 22013-57-6; VIII, 22013-58-7; IXa (R = Me), 38222-69-4; IXa (R = Et), 38222-70-7; IXb (R = Me), 38222-71-8; IXb (R = Et), 38222-72-9; X, 38222-73-0; XI, 22013-60-1.

Acknowledgment.—Thanks are due to Professor G. Scheibe for helpful discussions and for his encouragement of this research, and to the Deutsche Forschungsgemeinschaft for financial support.

Synthesis and Structure of a Trimer of 4,5-Dihydropyridazine

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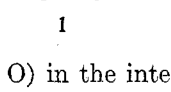
The synthesis and X-ray structure of the trimer of 4,5-dihydropyridazine (11), 4,4a,9,9a,14,14a-hexahydro-3*H*,8*H*,13*H*-tripyridazino[1,6-*a*:1′,6′-*c*:1″,6″-*e*]-*s*-triazine (13), is described.

Dipolar structures or ylides (**1**) have been assigned to a number of "1,3-diradical" (**2**) systems. These intermediates have been examined theoretically,[1] observed spectroscopically,[2,3] trapped with dipolarophiles,[4,5] and studied kinetically.[4-6]

We were interested in preparing the oxadiazole ring system, (**3**), in an attempt to evaluate ylide character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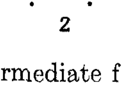
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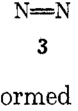
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(**1**, X = O) in the intermediate formed from thermolysis of **3**.[7]

Results and Discussion

The most likely entry into the oxadiazole ring system appeared to be through the Diels–Alder addition of furan (**4**) and an azodicarboxylate (**5**). This reaction with diethyl azodicarboxylate has been reported, although the adduct **6a** was only poorly characterized because of its instability.[8-12] Critical factors in its formation seem to be temperature, stoichiometry, and length of time allowed for formation of adduct. Apparently, once formed, the adduct undergoes additional

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[1] E. F. Hayes and A. K. Q. Siu, *J. Amer. Chem. Soc.*, **93**, 2090 (1971).

[2] T. Do-Mihn, A. M. Trozzolo, and G. W. Griffin, *ibid.*, **92**, 1402 (1970).

[3] D. R. Arnold and L. A. Karnischky, *ibid.*, **92**, 1404 (1970).

[4] R. Huisgen and H. Mäder, *ibid.*, **93**, 1777 (1971).

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[6] R. M. Kellogg, S. Wassenaar, and J. Buter, *Tetrahedron Lett.*, 4689 (1970).

[7] Several examples of the thiadiazole (sulfur analog of **3**) ring system are known; *cf.* ref 6; W. J. Middleton, *J. Org. Chem.*, **34**, 3201 (1969); D. H. R. Barton, E. H. Smith, and B. J. Willis, *Chem. Commun.*, 1226 (1970); R. M. Kellogg and S. Wassenaar, *Tetrahedron Lett.*, 1987 (1970); A. P. Krapcho, D. R. Rao, M. P. Silvon, and B. Abegaz, *J. Org. Chem.*, **36**, 3885 (1971).

[8] K. Alder and H. Niklas, *Justus Liebigs Ann. Chem.*, **585**, 81 (1954).

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[10] J. G. Kuderna, J. W. Sims, J. F. Wikstrom, and S. B. Soloway, *J. Amer. Chem. Soc.*, **81**, 382 (1959).

[11] K. N. Zelenin and I. P. Bezhan, *Zh. Org. Khim.*, **2**, 1524 (1966).

[12] K. N. Zelenin and I. P. Bezhan, *Dokl. Akad. Nauk SSSR*, **191**, 295 (1970).

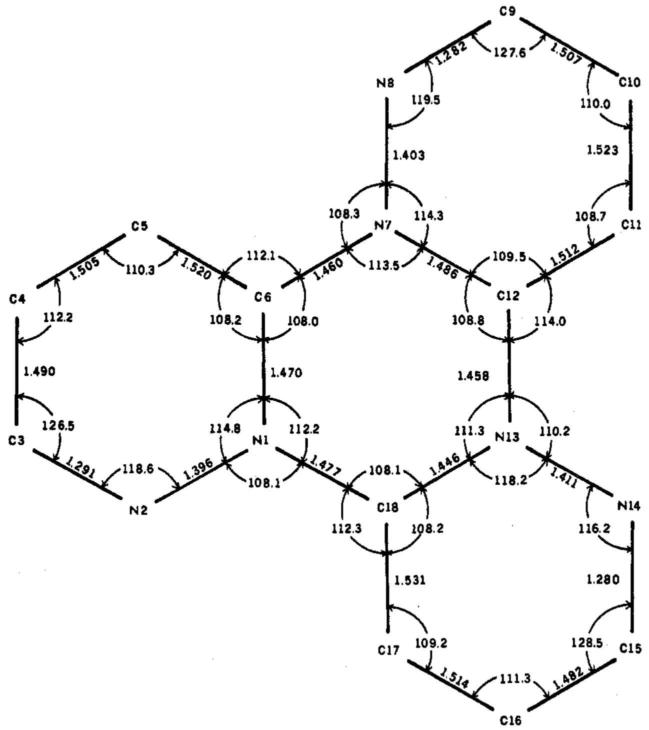
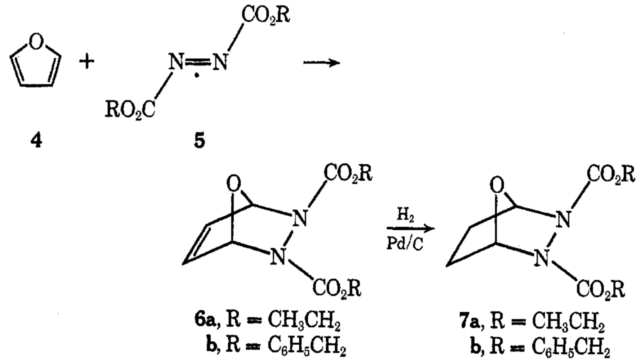
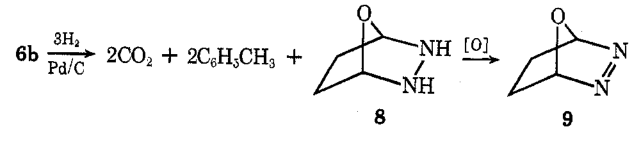


Figure 1.—Structural parameters for 4,4a,9,9a,14,14a-hexahydro-3*H*,8*H*,13*H*-tripyridazino[1,6-*a*:1′,6′-*c*:1″,6″-*e*]-*s*-triazine (**13**). The numbers shown are for crystallographic convenience and do not correspond to those used in the nomenclature.

reactions, giving polymeric products. Short time periods and near equal stoichiometry allowed us to isolate, after reduction, compound **7a** which could be readily characterized. However, hydrolysis of **7a** with



methanolic KOH proceeded rapidly at room temperature to yield considerable highly insoluble polymeric material. Repeated extraction of the product mass followed by chromatography yielded a small amount of material with mp 139.5–140.5° which was assigned the formula $C_{12}H_{18}N_6$ (**13**, *vide infra*) on the basis of its mass spectrum and elemental analysis. It was decided that nonaqueous conditions were necessary for isolation of the hydrazo product **8** which could then be oxidized to the desired oxadiazole derivative **9**. The benzyloxy group, commonly used in peptide syntheses as a protecting group, seemed ideal for this purpose.[13] It was expected that reduction of **6b** would lead directly to **8**. However, it was found that the hydrogenation could be controlled so as to isolate **7b**, while more rigorous conditions gave the same compound isolated from the aqueous hydrolysis of **7a** ($C_{12}H_{18}N_6$). Analysis of spectral data including a 220-MHz nmr was not sufficient to assign a structure. An X-ray structure on a single crystal of the sample identified the compound as 4,4a,-9,9a,14,14a-hexahydro-3*H*,8*H*,13*H*-tripyridazino[1,6-*a*:1′,6′-*c*:1″,6″-*e*]-*s*-triazine (**13**) with the dimensions shown in Figure 1. Figure 2 shows the perspective view.



$$6b \xrightarrow[\text{Pd/C}]{3H_2} 2CO_2 + 2C_6H_5CH_3 + \mathbf{8} \xrightarrow{[O]} \mathbf{9}$$

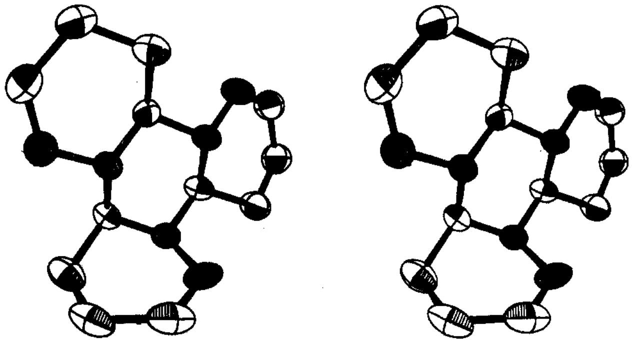
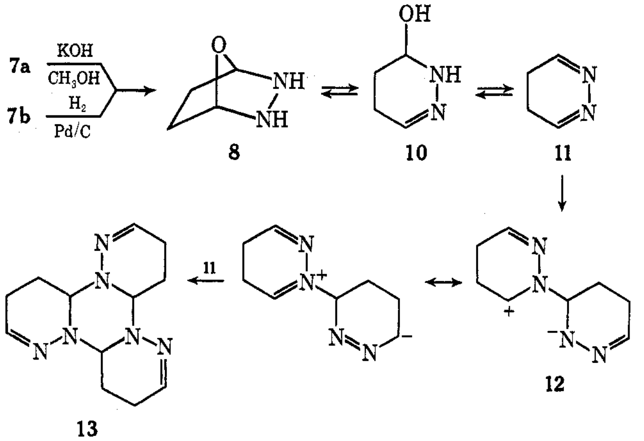
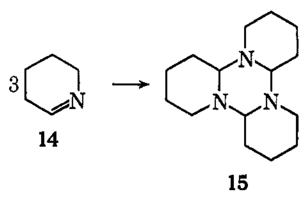


Figure 2.—Perspective view of **13**. The nitrogen atoms are completely shaded.

Since both **7a** and **7b** give rise to the same final product, it seems reasonable to assume a common intermediate. This intermediate (**8**) could tautomerize to **10** which, through loss of water, would give 4,5-dihydropyridazine (**11**). The trimerization of **11** can be imagined to involve formation of a "dimer," probably a resonance stabilized 1,4 dipole (**12**), which can add to another molecule of 4,5-dihydropyridazine. There are numerous examples of 1,4-dipolar cycloadditions involving products which resulted from combination of three fragments.[14] A similar trimerization, though





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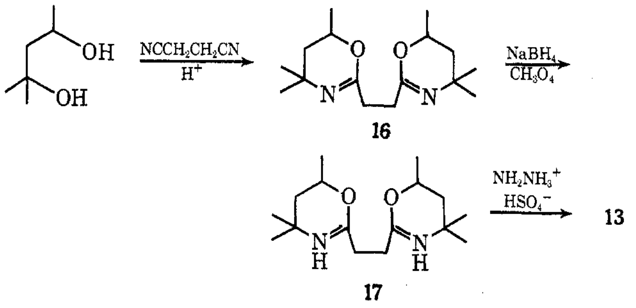
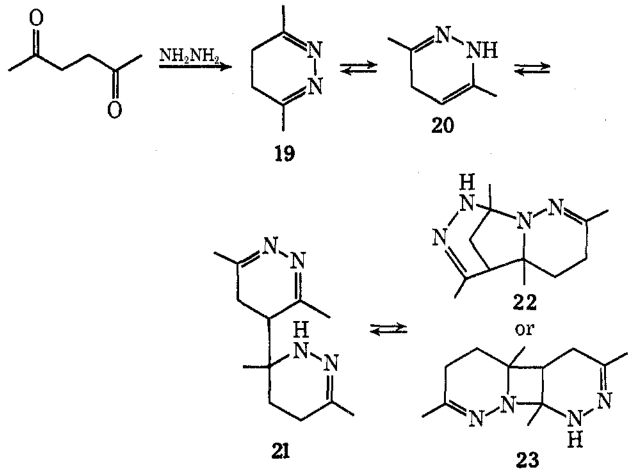
[14] R. Huisgen, *Z. Chem.*, **8**, 290 (1968).

possibly not by the same mechanism, has been reported by Schöpf and coworkers[15] (**14** → **15**).

A natural approach in establishing the intermediacy of **11** is the independent synthesis of **11** from succinaldehyde and hydrazine. Because of the lability of succinaldehyde it was decided that good use could be made of the Meyers aldehyde reagent[16-18] which has been shown to be the procedure of choice for preparing a variety of carbonyl derivatives. The bis(oxazine) **16**[17] in reduced from **17**[18] is actually a protected form of succinaldehyde.[16] Treatment of **17** with hydrazine hydrogen sulfate afforded **13** presumably through **11**.



Part of the instability of dihydropyridazines can apparently be alleviated when bulky groups are incorporated into the ring. 3,6-Diphenyl-4,5-dihydropyridazine has been isolated and is reasonably stable.[19] In contrast, acetonylactetone (**18**) and hydrazine give a tricyclic dimer.[20,21] The authors[21] postulate **22** and **23** as possible structures for the dimer, preferring **22** on the basis of ^{13}C nmr data. It is thought to arise by a tautomerization of 3,6-dimethyl-4,5-dihydropyridazine (**19**)



followed by a series of addition reactions. Models of the trimer of 2,6-dimethyl-4,5-dihydropyridazine, with a structure corresponding to that of **13**, illustrate that the compound would be extremely strained as all three methyl groups would have to assume axial positions on the central rings' "chair-like" configuration (see Figure 2). This basically must account for the difference in reactivity between **13** and **19**.

Experimental Section

Synthesis of Diethyl 7-Oxa-2,3-diazabicyclo[2.2.1]hept-5-ene-2,3-dicarboxylate (6a).—Diethyl azodicarboxylate (10 g) was stirred with furan (20 ml) for 3 hr. The reaction flask was protected from light. After 3 hr furan was removed under reduced pressure to give a colorless viscous liquid. This adduct was found to be unstable toward ethanol, light, acid, base, silica gel, alumina, and high temperature. The nmr of the product consisted of a triplet at 1.35 ppm (6 H, $-CH_2CH_3$), a quartet at 4.30 (4 H, $-CH_2CH_3$), a singlet at 6.50 (2 H), and a singlet at 6.75 ppm (2 H). The carbonyl absorption occurred at 1725 cm^{-1}.

Reduction of Diethyl 7-Oxa-2,3-diazabicyclo[2.2.1]hept-5-ene-2,3-dicarboxylate.—Diethyl 7-oxa-2,3-diazabicyclo[2.2.1]hept-5-ene-2,3-dicarboxylate (**6a**) obtained as above was hydrogenated over 10% Pd/C catalyst using 15 psi of hydrogen and ether as solvent. After 1 hr of shaking with hydrogen, the ethereal solution was filtered and concentrated to give a solid product which was recrystallized from methanol to give 7.5 g of diethyl 7-oxadiazabicyclo[2.2.1]heptane-2,3-dicarboxylate (**7a**), mp 103–104°. The nmr spectrum consisted of a triplet at 1.37 (6 H, $-CH_2CH_2$), a singlet at 2.15 (4 H, $-CH_2CH_2-$), a quartet at 4.20 (4 H, $-CH_2CH_3$), and a singlet at 6.00 ppm [2 H, -C(N)H-O]. *Anal.* Calcd for $C_{10}H_{16}N_2O_5$: C, 49.17; H, 6.60; N, 11.46. Found: C, 49.20; H, 6.68; N, 11.46.

Synthesis of Dibenzyl 7-Oxa-2,3-diazabicyclo[2.2.1]hept-5-ene-2,3-dicarboxylate (6b).—Dibenzyl azodicarboxylate (5 g) was stirred with furan (20 ml) for 1 hr in a reaction flask protected from light. The excess furan was removed from the adduct under reduced pressure. The nmr consisted of a singlet at 5.25 (4 H, $-CH_2C_6H_5$), a multiplet at 6.65 (4 H, -CH=CH-, OCHN) and a singlet at 7.38 ppm (10 H, $-C_6H_5$). The carbonyl absorption occurred at 1725 cm^{-1}.

Reduction of Dibenzyl 7-Oxa-2,3-diazabicyclo[2.2.1]hept-5-ene-2,3-dicarboxylate.—Dibenzyl 7-oxa-2,3-diazabicyclo[2.2.1]-hept-5-ene-2,3-dicarboxylate (**6b**) obtained as above was hydrogenated over 10% Pd/C catalyst using 12 psi of hydrogen and ether as a solvent. After 1 hr the ethereal solution was filtered and concentrated to give 6.7 g of dibenzyl 7-oxa-2,3-diazabicyclo-[2.2.1]heptane-2,3-dicarboxylate (**7b**) as a viscous liquid.

The nmr consisted of a singlet at 1.93 (4 H, $-CH_2CH_3-$), a singlet at 5.14 (4 H, $-CH_2C_6H_5$), a singlet at 6.00 (2 H, -NCHO), and a singlet at 7.25 ppm (10 H, $-C_6H_5$). The carbonyl absorption occurred at 1725 cm^{-1}. *Anal.* Calcd for $C_{20}H_{20}O_5N_2$: C, 65.24; H, 5.47; N, 7.60. Found: C, 64.88; H, 5.57; N, 7.64.

Decomposition of Diethyl 7-Oxa-2,3-diazabicyclo[2.2.1]heptane-2,3-dicarboxylate (7a).—A solution of diethyl 7-oxa-2,3-diazabicyclo[2.2.1]heptane-2,3-dicarboxylate (**7a**, 6.0 g) in methanol (20 ml) was added to an aqueous methanolic solution of potassium hydroxide (6 g) under a nitrogen atmosphere. The reaction contents were then stirred for 4 hr and poured in 200 ml of water. After adjusting the pH to 7 the mixture was extracted with three 100-ml portions of ether. The ether extracts were dried over anhydrous magnesium sulfate and concentrated to give a yellow solid which was chromatographed on a silica gel column using ether as an eluting solvent. The product so obtained was recrystallized from ether-petroleum ether to give 250 mg of compound **13**, mp 139.5–140.5°.

Hydrogenolysis of Dibenzyl 7-Oxa-2,3-diazabicyclo[2.2.1]heptane-2,3-dicarboxylate (7b).—A solution of dibenzyl 7-oxa-2,3-diazabicyclo[2.2.1]heptane-2,3-dicarboxylate (**7b**, 5.0 g) in ether (200 ml) was hydrogenated using 10% Pd/C (1.1 g) and 30 psi of hydrogen pressure for 24 hr. The catalyst was removed by filtration and the solution was concentrated under reduced pressure. The presence of toluene in the mixture of products was confirmed by nmr. Ether, 10 ml, was added to the residue and the insoluble part of the residue was separated by decantation. Removal of the ether gave 500 mg of viscous product which was chromatographed on silica gel column using benzene and then ether as eluting solvents. Compound **13** was recrystallized from ether petroleum ether to give 200 mg of product, mp 139.5–140.5°.

The nmr consisted of a complex multiplet at 1.70–2.70 (12 H, $-CH_2$), a complex multiplet from 3.50 to 3.75 (3 H, -N-CH-N-),

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(16) A. I. Meyers and N. Nazarenko, *J. Amer. Chem. Soc.*, **94**, 3243 (1972), and references listed therein.

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(20) C. G. Overberger, N. P. Byrd, and R. B. Mesrobian, *ibid.*, **78**, 1961 (1956).

(21) P. De Mayo, J. B. Stothers, and M. C. Usselman, *Can. J. Chem.*, **50**, 613 (1972).

and a singlet at 6.82 ppm (3 H, -N=CH-). The ir indicated a prominent bond at 1620 cm^{-1} (-C=N-) and the mass spectrum showed a parent ion at 246. *Anal.* Calcd for $C_{12}H_{18}N_6$: C, 58.49; H, 7.33; N, 34.12. Found: C, 58.34; H, 7.29; N, 33.90.

Synthesis of 4,4a,9,9a,14,14a-Hexahydro-3*H*,8*H*,13*H*-tripyridazino[1,6-*a*:1',6'-*c*:1'',6''-*e*]-*s*-triazine (13) from 1,2-Bis(tetrahydro-1,3-oxazin-2-yl)ethane (17).—1,2-Bis(tetrahydro-1,3-oxazin-2-yl)ethane[18] (**17**) was prepared by sodium borohydride reduction of 1,2-bis(dihydro-1,3-oxazin-2-yl)ethane[17] (**16**). A mixture of crude **17** (4.0 g, 3.5 mmol), hydrazine hydrogen sulfate (2.0 g, 15.9 mmol), and 400 ml of methanol was stirred under nitrogen at room temperature for 24 hr. The methanol was removed and after addition of water, the solution was extracted with several portions of ether. The dried ether extract upon concentrating yielded 300 mg of crude product which was washed with 2 ml of petroleum ether to remove the color. Recrystallization from ether gave 150 mg of pure product, mp 139–140°. The mixture melting point and the mass spectra of this sample and the one prepared from hydrogenation of **7b** indicated, that the compounds were identical.

X-Ray Analysis of 13.—A crystal of dimensions 0.17 × 0.22 × 0.11 mm, mounted on a G.E. XRD-5 diffractometer, displayed *mmm* symmetry and axial extinctions for (h00) when h was odd; (0k0) when k was odd; (00l) when l was odd, uniquely identifying the space group as $P2_12_12_1$. Lattice constants were determined from a least-squares refinement of ten carefully measured 2θ values (1° take-off angle and a 0.05° slit) at $2\theta > 74°$ where the Cu Kα_2 doublet is resolvable. The resultant lattice constants and their estimated standard deviations are $a = 14.641 \pm 0.003$ Å, $b = 16.118 \pm 0.002$ Å, and $c = 5.228 \pm 0.001$ Å.

Data Collection and Refinement

Three dimensional intensity data were collected on a G.E. XRD-490 fully automated diffractometer by the stationary crystal–stationary counter method using balanced nickel and cobalt filters and Cu Kα radiation. A total of 1278 reflections were measured to a 2θ maximum of 130° (d = 0.85 Å), and of these 1017 (80%) were considered statistically acceptable by the criterion

$$[I_{\mathrm{Ni}} - 2\sigma(I_{\mathrm{Ni}})] - [I_{\mathrm{Co}} + 2\sigma(I_{\mathrm{Co}})] > 75 \text{ counts}$$

for 10-sec counting times. The data were corrected for $\alpha_1 - \alpha_2$ splitting[22] as a function of 2θ and for absorption as a function of φ (linear $\mu = 7.0$ cm^{-1} and a ratio of observed transmission factors of 1.38:1.00). Lorentz–polarization corrections were made and the intensities were reduced to structure amplitudes in the usual manner.

Normalized structure magnitudes were computed using a $K(s)$ curve[23] and two sets of tangent refinements were generated using $|E|$ greater than 1.3 in each. E maps were generated for each, one of which contained a random set of peaks and the other contained 18 peaks at chemically reasonable distances and angles. So as not to prejudice our approach, initially all atoms were assigned a carbon scattering factor curve.[24] Five cycles of block-diagonal least squares, using $1/\sigma^2$ weights, led to a value of $R = 0.12$. The assignment of the six nitrogen atoms was easily made at this stage on the basis of temperature factors, electron density peak heights, and such chemical evidence as bond distances and bond angles. With all of the atoms now properly assigned, additional cycles of isotropic least squares, conversion to anisotropic temperature factors, and further anisotropic refinement lowered the value of the reliability index to $R = 0.11$.

The 18 hydrogen atoms were readily located for a difference electron density map phased by the nonhydrogens. The absence of any peaks of height greater than 0.2 e/Å^3 about the six peaks earlier presumed to be nitrogens corroborates that assignment. Least-squares refinement was continued varying the coordinates of all atoms with the nonhydrogens having varying anisotropic temperature factors and the hydrogens having fixed isotropic temperature factors of 5.0 Å^2. The refinement converged at a value of $R = 0.046$ with all shifts less than one-tenth the estimated standard deviation of the respective parameter.[25]

Registry No.—**4,** 110-00-9; **5a,** 1972-28-7; **5b,** 2449-05-0; **6a,** 13925-26-3; **6b,** 37819-02-6; **7a,** 37819-03-7; **7b,** 37819-04-8; **13,** 37819-05-9; **16,** 37819-06-0; **17,** 37819-07-1.

Acknowledgment.—We appreciate substantive discussions with Professor A. I. Meyers regarding the oxazine ring system.

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[24] Scattering factors for nitrogen and carbon were taken from D. Cromer and J. Waber, *ibid.*, **18**, 104 (1965), whereas scattering factors for hydrogen were taken from "International Tables for X-ray Crystallography," Vol. III, Kynoch Press, Birmingham, England, 1968.

[25] Listings of structure factors, coordinates, and anisotropic temperature factors will appear following these pages in the microfilm edition of this volume of the journal. Single copies may be obtained from the Business Operations Office, 1155 Sixteenth St., N.W., Washington, D. C. 20036, by referring to code number JOC-73-1102. Remit check or money order for $3.00 for photocopy or $2.00 for microfiche.