The Synthesis and Reactions of Some 1-(Nitroaryl)diaziridines

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1-(2,4-Dinitrophenyl)- and 1-(2,4,6-trinitrophenyl)diaziridines are prepared by treating appropriate diaziridines with 2,4-dinitrofluorobenzene and 2,4,6-trinitroanisole, respectively. The 1-(2,4-dinitrophenyl)-3,3-dialkyl- and 1-(2,4,6-trinitrophenyl)-3,3-dialkyldiaziridines are isomerized in refluxing toluene into 2,4-dinitrophenyl- and 2,4,6-trinitrophenylhydrazones. However, 1-(2,4-dinitrophenyl)-2,3-dialkyldiaziridines and 1-(2,4-dinitrophenyl)-2,3,3-trialkyldiaziridines are converted in refluxing toluene into 2-alkyl-6-nitrobenzotriazole 1-oxides and ketones. Acid hydrolysis of the 1-(nitroaryl)-2-alkyldiaziridines forms 1-nitroaryl-2-alkylhydrazines.

Previous work in this laboratory has shown that 1-(nitroaryl)aziridines are easily synthesized by treating an aziridine bearing an NH group with either 2,4,6-trinitroanisole or 2,4-dinitrofluorobenzene.[1] We now report that 1-(2,4-dinitrophenyl)- and 1-(2,4,6-trinitrophenyl)diaziridines can be similarly prepared. Some reactions of these compounds are also described. The only other 1-aryldiaziridine that has been made, but not isolated, is 1-phenyl-3,3-pentamethylenediaziridine.[2]

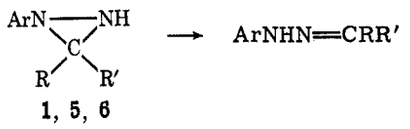
Results

Reaction of diaziridines having one or two NH groups with 2,4-dinitrofluorobenzene in ether containing triethylamine at room temperature or with 2,4,6-trinitroanisole in methanol led to 1-(2,4-dinitrophenyl)- and 1-(2,4,6-trinitrophenyl)diaziridines. These diaziridines are characterized in Table I. Picryl chloride was used in place of 2,4,6-trinitroanisole in a few instances, but the crude products that were obtained were less pure than when the anisole was employed.

The nmr spectra of the 1-(2,4-dinitrophenyl)- or 1-(2,4,6-trinitrophenyl)diaziridines were consistent with the proposed structures. This conclusion is based on the similar splitting patterns observed for the 3,3-pentamethylene protons in diaziridines **1**, **2**, **6**, and **7** and their progenitors, 3,3-pentamethylene- and 1-methyl-3,3-pentamethylenediaziridines. In both series of compounds the 3,3-pentamethylene group appears as a broad multiplet extending from δ 1.1 to 2.3. By contrast, the protons for this group in cyclohexanone 2,4-dinitrophenylhydrazone and cyclohexanone 2,4,6-trinitrophenylhydrazone [into which some of the 1-(nitroaryl)diaziridines can be isomerized; see below] form two multiplets, the four protons α to the carbon of the >C=N- group being separate and downfield at δ 2.0–2.7 while the remaining six protons are at δ 1.5–2.0.

Interestingly, each of the two aromatic hydrogens of 1-(2,4,6-nitrophenyl)-3,3-pentamethylenediaziridine (**6**) and 1-(2,4,6-trinitrophenyl)-2-methyl-3,3-pentamethylenediaziridine (**7**) appears as a sharp doublet. Models indicate that free rotation of the *o*-nitroaryl group is inhibited by the 3,3-pentamethylene group, thus rendering the aryl protons nonequivalent.

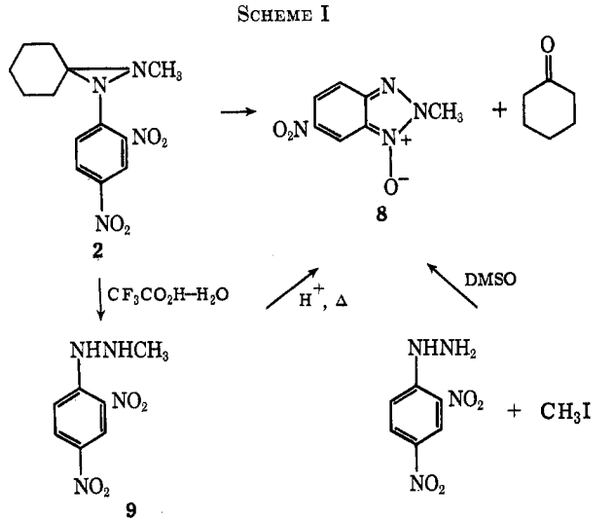
The 1-(2,4-dinitrophenyl)- and 1-(2,4,6-trinitrophenyl)diaziridines bearing an NH group (**1**, **5**, **6**) upon heating in toluene for about 3 hr rearrange into the corresponding known 2,4-dinitrophenyl- or 2,4,6-trinitrophenylhydrazones.



1, 5, 6

Heating of the 1-(2,4-dinitrophenyl)-2,3,3-trialkyldiaziridines **2**, **3**, and **4** did not give hydrazones but afforded instead 2-alkyl-6-nitrobenzotriazole 1-oxides and ketones. Thus 1-(2,4-dinitrophenyl)-2-methyl-3,3-pentamethylenediaziridine (**2**) was converted after 7 hr in refluxing toluene into 2-methyl-6-nitrobenzotriazole 1-oxide (**8**) and cyclohexanone (Scheme I).

SCHEME I



The cyclohexanone was identified by gas chromatography. The structure of **8** was confirmed by contrasting its nmr, ir, and mass spectra with those of the known isomeric 3-methyl-6-nitro-1,2,3-benzotriazole 1-oxide,[3] by elemental analyses, and by alternate syntheses. One synthesis involved treatment of 2,4-dinitrophenylhydrazine with methyl iodide in dimethyl sulfoxide at room temperature, while the other synthesis involved the acid-catalyzed dehydration of 1-methyl-2-(2,4-dinitrophenyl)hydrazine (**9**) (Scheme I). The acid-catalyzed dehydration of 1-aryl-2-(*o*-nitroaryl)-hydrazines into 2-arylbenzotriazole 1-oxides is a known reaction.[4,5] Compound **9**, although easily transformed into **8** by acid, remained unchanged after 7 hr in refluxing toluene. This clearly established that **9** was not a reaction intermediate in the conversion of **2** to **8**.

(1) H. W. Heine, G. J. Blosick, and G. B. Lowrie, *Tetrahedron Lett.*, 4801 (1968).

(2) E. Schmitz and R. Ohme, *Chem. Ber.*, **94**, 2166 (1961).

(3) O. L. Brady and C. V. Reynolds, *J. Chem. Soc.*, 1273 (1931).

(4) A. Mangini, *Gazz. Chim. Ital.*, **65**, 1191 (1935).

(5) (a) H. Goldstein and A. Jaquet, *Helv. Chim. Acta*, **24**, 30 (1941); (b) H. Goldstein and R. Stamm, *ibid.*, **35**, 1470 (1952).

TABLE I

1-(NITROARYL)DIAZIRIDINES FROM THE REACTION OF 2,4-DINITROFLUOROBENZENE AND 2,4,6-TRINITROANISOLE WITH DIAZIRIDINES[a]

ArNNCR²R³
|
R¹

Compd	Ar	R¹	R²		R³	Crude yield, %	Mp, °C
1	$2,4(O_2N)_2C_6H_3$	H		$-(CH_2)_5-$		81	123–125
2	$2,4(O_2N)_2C_6H_3$	CH_3		$-(CH_2)_5-$		81	92–93
3	$2,4(O_2N)_2C_6H_3$	$CH(CH_3)_2$	CH_3		CH_3	93	100–103
4	$2,4(O_2N)_2C_6H_3$	C_6H_{11}	H		C_2H_5	88	115–116
5	$2,4(O_2N)_2C_6H_3$	H	CH_3		C_2H_5	75	73–75
6	$2,4,6(O_2N)_3C_6H_2$	H		$-(CH_2)_5-$		71	132–134
7	$2,4,6(O_2N)_3C_6H_2$	CH_3		$-(CH_2)_5-$		94	108–109

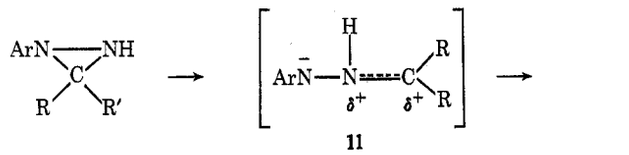
[a] Satisfactory analytical data (±0.4% for C, H, and N) were reported for all new compounds listed in the table: Ed.

Compound **9** was obtained by the hydrolysis of **2** in commercial trifluoroacetic acid (Scheme I). The structure of **9** was proved by mass spectrometry, elemental analyses, and comparison of its infrared and nmr spectra with those of the known isomeric 1-methyl-1-(2,4-dinitrophenyl)hydrazine prepared according to the method of Blanksma and Wackers.[6] The acid-catalyzed hydrolysis of compounds **3** and **4** to the corresponding 1-(2,4-dinitrophenyl)-2-isopropylhydrazine and 1-(2,4-dinitrophenyl)-2-cyclohexylhydrazine was also achieved in high yield. The acid hydrolysis of 1,2-dialkyldiaziridines to 1,2-dialkylhydrazines is a known reaction,[7] as is the hydrolysis of 1-alkyldiaziridines to 1-alkylhydrazines.[8]

The conversions of diaziridines **3** and **4** to the corresponding 2-alkylbenzotriazole 1-oxides were also achieved in good yields, but attempts to convert **7** to 2-methyl-4,6-dinitrobenzotriazole 1-oxide (**10**) were thwarted by extensive decomposition. Compound **10** was extracted from the reaction mixture with difficulty and in low yield.

Discussion

The rearrangement of the 1-(2,4-dinitrophenyl)- and 1-(2,4,6-trinitrophenyl)diaziridines bearing an NH group into hydrazones probably proceeds through the cleavage of the carbon–nitrogen bond of the diaziridine ring to give the dipolar intermediate **11**. Intermediate **11** subsequently forms the hydrazone. Other diaziridine derivatives have been shown to rearrange into hydrazones. For example, a number of 1,2-dibenzoyldiaziridines isomerized under mild conditions to ketone dibenzoylhydrazones.[9]

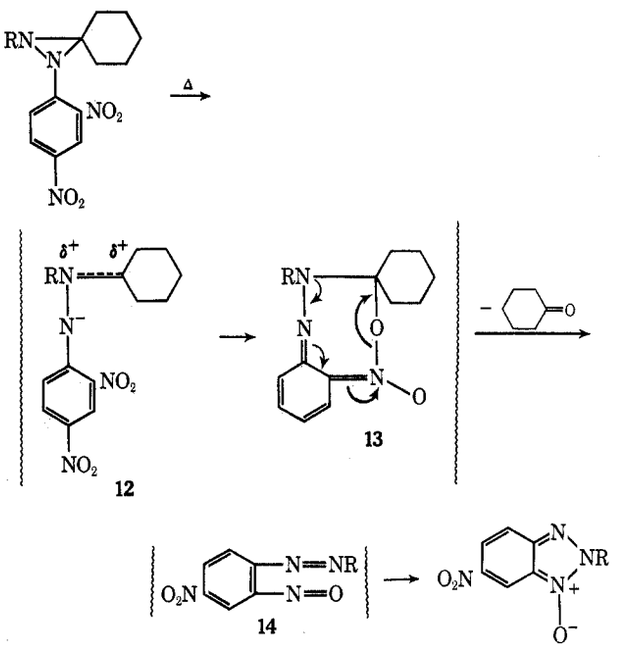


11

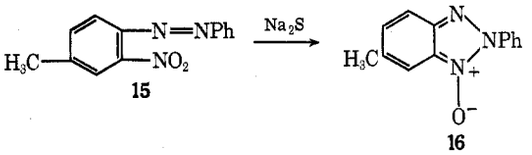
ArNHN=CRR′

One possible mechanism for the conversion of 1-(2,4-dinitrophenyl)-2,3,3-trialkyldiaziridines into benzotriazole 1-oxides and ketones involves the formation of a dipolar intermediate **12**, which undergoes ring closure to the intermediate **13**. Elimination of cyclohexanone from **13** would give the ortho nitroso azo intermediate **14**, which cyclizes into the benzotriazole 1-oxide (Scheme II).

SCHEME II

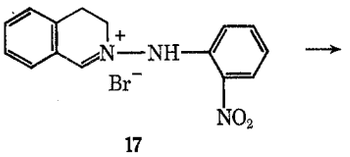


That an intermediate such as **14** could isomerize to a benzotriazole 1-oxide is supported by the observations that reducing agents such as sodium hydrosulfite, sodium sulfide, or hydrazine react with ortho nitro azo derivative **15** to form **16**.[10]



The ortho nitroso azo intermediate **18** similar to **14** has been proposed to account for the formation of the benzotriazole 1-oxide **19** when 2-(*o*-nitrophenylamino)-3,4-dihydroisoquinolinium bromide (**17**) is treated with pyridine.[11] Prior to this paper compound **19** was the only 2-alkylbenzotriazole 1-oxide reported.

(6) J. J. Blanksma and M. L. Wackers, *Recl. Trav. Chim. Pays-Bas*, **55**, 655 (1936).
(7) E. Schmitz, *Angew. Chem.*, **73**, 23 (1961).
(8) E. Schmitz and D. Habisch, *Chem. Ber.*, **95**, 680 (1962).
(9) E. Schmitz, D. Habisch, and Ch. Grundemann, *ibid.*, **100**, 142 (1967).
(10) E. Bamberg and R. Hubner, *Ber.*, **36**, 3822 (1903).
(11) R. Grashey, *Angew. Chem., Int. Ed. Engl.*, **1**, 158 (1962).



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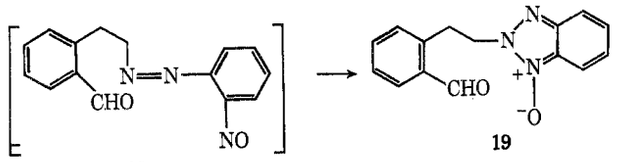
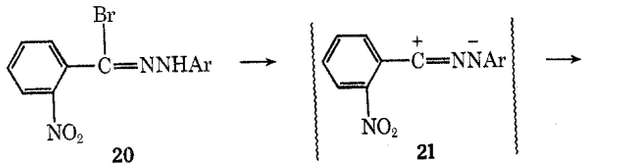
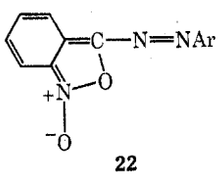


18 19

The solvolysis of *N*-(4-nitrophenyl)-*o*-nitrobenzhydrazonyl bromide (**20**) affords the azomethine imine **21**, an interesting counterpart to intermediate **12**. Interaction of the nitro group of **21** with the neighboring positive carbon yields 3-(4-nitrophenylazo)-anthranil 1-oxide (**22**).[12]



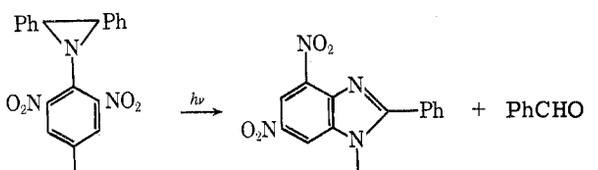
20 21



22

Ar = *p*-$O_2NC_6H_4$

The formation of 2-alkylbenzotriazole 1-oxides from 1-(2,4-dinitrophenyl)-2,3,3-trialkyldiaziridines resembles the photochemical conversion of 1-(2,4,6-trinitrophenyl)-2,3-diphenylaziridine (**23**) into 1-hydroxy-2-phenyl-4,6-dinitrobenzimidazole (**24**) and benzaldehyde.[1]



23 24

Experimental Section

Compounds 1–5.—To a mixture of 4 mmol of triethylamine and 4 mmol of the appropriate diaziridine (3,3-pentamethylene-, 1-methyl-3,3-pentamethylene-, 1-isopropyl-3,3-dimethyl-, 1-cyclohexyl-3-ethyl-, 3-methyl-3-ethyldiaziridines) in 200 ml of dry ether was added all at once a solution of 4 mmol of 2,4-dinitrofluorobenzene in 40 ml of ether. The reaction mixture was allowed to stand for 18 hr and then the solvent was evaporated. One milliliter of CH_3OH was added to the residue and the mixture was slurried until a fine yellow powder was obtained which was filtered. The 1-(2,4-dinitrophenyl)diaziridines were recrystallized from a small quantity of methanol, special care being taken that the period of recrystallization was of short duration and that the recrystallizing solution was cooled immediately after the dissolution of the diaziridine.

Compounds 6 and 7.—A solution of 4 mmol of either 3,3-pentamethylene- or 1-methyl-3,3-pentamethylenediaziridine in 10 ml of CH_3OH was added to 4 mmol of 2,4,6-trinitroanisole in 10 ml of CH_3OH. The reaction mixture was allowed to stand at room temperature overnight and then the solvent was evaporated. The same care was taken in recrystallizing **6** and **7** as with compounds **1–5**.

Rearrangements of 1, 5, and 6.—A solution of **1**, **5**, or **6** in toluene was refluxed for 4 hr. Evaporation of the toluene gave the known cyclohexanone 2,4-dinitrophenylhydrazone, butanone 2,4-dinitrophenylhydrazone, and cyclohexanone 2,4,6-trinitrophenylhydrazone, respectively, in quantitative yields.

Conversion of 2 to 8.—A solution of 223 mg of **2** in 10 ml of toluene was refluxed for 7.5 hr. Evaporation of the solvent gave 144 mg (97%) of **8**, mp 200–201°. After two recrystallizations from ethanol **8** had mp 200–202°; molecular ion *m/e* 194; uv max (absolute EtOH) 271 nm (ε 18,000), 204 (14,000); ir (Nujol) 6.21, 6.70, 7.44, 7.66, 7.95, 8.10, 8.59, 9.40, 9.73, 10.89, 11.16, 12.00, 12.70, 13.20, 13.59, 14.54 μ; nmr ($CDCl_3$) δ 4.30 (s, 3, CH_3), 7.80 (d, 1, *J* = 9.5 Hz, 4-H), 8.20 (pair of d's, 1, $J_{4,5}$ = 9.5, $J_{5,7}$ = 2.0 Hz, 5-H), 8.70 (d, 1, *J* = 2.0 Hz, 7-H).

Anal. Calcd for $C_7H_6N_4O_3$: C, 43.29; H, 3.11; N, 28.85. Found: C, 43.38; H, 3.28; N, 28.72.

Conversion of 3 to 2-Isopropyl-6-nitrobenzotriazole 1-Oxide.—A solution of 200 mg of **3** in 10 ml of toluene was refluxed for 3 hr and the toluene was then evaporated to give a quantitative yield of **22**. After recrystallization from ethanol **22** melted at 151–153°; uv max (absolute EtOH) 271 nm (ε 20,000), 204 (16,000); nmr ($CDCl_3$) δ 1.70 (d, 6, 2 CH_3's), 5.63 [m, 1, $(CH_3)_2$**CH**], 7.92 (d, 1, *J* = 9.5 Hz, 4-H), 8.30 (pair of d's, 1, $J_{4,5}$ = 9.5, $J_{5,7}$ = 2.0 Hz, 5-H), 8.85 (d, 1, *J* = 2.0 Hz, 7-H).

Anal. Calcd for $C_9H_{10}N_4O_3$: C, 48.65; H, 4.53; N, 25.21. Found: C, 48.67; H, 4.74; N, 24.85.

Conversion of 4 to 2-Cyclohexyl-6-nitrobenzotriazole 1-Oxide.—A solution of 202 mg of **4** in 10 ml of toluene was refluxed for 14 hr. Evaporation of the solvent gave 174 mg (98%) of **23**. Recrystallization from ethanol gave **23**, mp 120–122°.

Anal. Calcd for $C_{12}H_{14}N_4O_3$: C, 54.93; H, 5.37; N, 21.36. Found: C, 55.28; H, 5.35; N, 21.48.

Reaction of Methyl Iodide with 2,4-Dinitrophenylhydrazine. Preparation of 8.—To 1.982 g (0.01 mol) of 2,4-dinitrophenylhydrazine in 11 ml of DMSO was added 1.420 g (0.01 mol) of CH_3I. The reaction mixture was allowed to stand at room temperature for 42 hr. The mixture was cooled in an ice bath and about 5 ml of H_2O was added gradually. The precipitate of **8** (270 mg) was collected and water was added to the filtrate until an oil settled out. This oil solidified after 12 hr and an additional 905 mg of **8** was filtered. The total yield of **8** was 1.175 g (60%).

Conversion of 2 to 9.—A solution of 273 mg of **2** in 1 ml of commercial CF_3CO_2H was allowed to stand overnight at room temperature. The solvent was evaporated and 0.5 ml of CH_3OH was added to the residue. Trituration of the mixture gave 137 mg (69%) of **9**. These recrystallizations from CH_3OH gave **9**, mp 125–127°.

Anal. Calcd for $C_7H_8N_4O_4$: C, 39.63; H, 3.80; N, 26.41. Found: C, 39.72; H, 4.06; N, 26.37.

Conversion of 3 to 1-(2,4-Dinitrophenyl)-2-isopropylhydrazine.—A solution of 378 mg of **3** in 4 ml of CF_3CO_2H was stirred for 2 hr and then the solvent was evaporated in a hood. The residue was recrystallized from ethanol to give 278 mg (86%) of **24**, mp 106–108°.

Anal. Calcd for $C_9H_{12}N_4O_4$: C, 45.00; H, 5.03; N, 23.32. Found: C, 45.30; H, 4.97; N, 22.90.

Conversion of 4 to 1-(2,4-Dinitrophenyl)-2-cyclohexylhydrazine.—A mixture of 1.099 g of **4** and 5 ml of CF_3CO_2H was stirred for 5 min and then the solvent was evaporated. A small quantity of MeOH was added to the dark brown oil. Trituration of the mixture gave 732 mg of **25**. Recrystallization from acetonitrile gave **25**, mp 160–162.5°.

Anal. Calcd for $C_{12}H_{16}N_4O_4$: C, 51.42; H, 5.75; N, 19.99. Found: C, 51.54; H, 6.03; N, 19.89.

Preparation of 8 from 9.—A mixture of 50 mg of **9**, 10 ml of CH_3OH, and 3 drops of concentrated hydrochloric acid was stirred for 7 hr. The solvent was evaporated to give 39 mg of crude **8**. Recrystallization gave dark-colored crystals of **8**, mp 195–198°. The infrared spectrum was identical with that of **8** prepared by heating **2** in toluene.

Preparation of 22 from 25.—A mixture of 67 mg of **24**, 10 ml of EtOH, and 1 drop of concentrated hydrochloric acid was refluxed for 3.5 hr. The solvent was evaporated to give 49 mg (79%) of crude **22**.

(12) A. F. Hegarty, M. Cashman, J. B. Aylward, and F. L. Scott, *J. Chem. Soc. B*, 1879 (1971).

Synthesis of 23 from 25.—A mixture of 130 mg of **25**, 10 ml of MeOH, and 1 drop of concentrated hydrochloric acid was refluxed for 2 hr. Evaporation of the solvent gave 98 mg (81%) of crude **23**, mp 105–110°.

Registry No.—**1**, 35042-51-4; **2**, 35040-15-4; **3**, 35040-16-5; **4**, 35040-17-6; **5**, 35040-18-7; **6**, 35040-19-8; **7**, 35040-20-1; **8**, 35040-21-2; **9**, 35040-22-3; **23**, 35040-27-8; 2-isopropyl-6-nitrobenzotriazole 1-oxide, 35040-23-4; 2-cyclohexyl-6-nitrobenzotriazole 1-oxide, 35040-24-5; 1-(2,4-dinitrophenyl)-2-isopropylhydrazine 35040-25-6; 1-(2,4-dinitrophenyl)-2-cyclohexylhydrazine, 35040-26-7.

Acknowledgment.—We thank the Camille and Henry Dreyfus Foundation, Inc., and the Petroleum Research Fund for financial aid and Professor C. C. Sweeley for several mass spectra.

cis-8,9-Dihydroisoxazolo[5,4-*d*]pyrimidine-4,6(5*H*,7*H*)-diones

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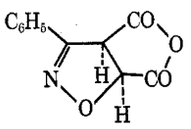
The Lossen rearrangement of *cis*-3-phenyl-2-isoxazoline-*N*-benzenesulfonyloxy-4,5-dicarboximide with aqueous ammonia or methylamine produced a mixture of *cis*-3-phenyl-5-ureido-2-isoxazoline-4-carboxamides and the corresponding 4-carboxylic acids (80–90% yield). Cyclization of these ureido acids with 3.3 *N* hydrochloric acid at 100° furnished the title compounds. Structures of all products were established *via* their pmr and mass spectra; the stereochemistry of the 4,5-disubstituted 2-isoxazolines was shown to be *cis* with $J_{4,5}$ consistently between 9 and 12 Hz. Mechanisms for this selective Lossen degradation are discussed.

As part of our continuing interest in condensed uracils[2] as potential antimetabolites, we explored syntheses of isoxazolo[4,5- and 5,4-*d*]pyrimidinediones.[3] The initial plan to utilize 4,5-isoxazoledicarboxylic esters[4] and to convert these by the standard method[2] to the corresponding bishydroxamates, was thwarted when the latter could not be isolated. Thus, this approach to build the uracil system onto the isoxazole ring *via* the modified Lossen rearrangement of the 4,5-bishydroxamates[2] was abandoned. An alternate route to the isoxazolo pyrimidine system is reported below.

1,3-Dipolar addition of benzonitrile oxide to maleic anhydride produced *cis*-3-phenyl-2-isoxazoline-4,5-dicarboxylic anhydride (**1**).[6,7] It was planned to degrade **1**, *via* the Lossen reaction, to one, or both, of the corresponding β-amino acids[8,9] and then build up, in this instance, the dihydrouracil system. Hydroxylamine smoothly transformed **1** to the corresponding *N*-hydroxyimide, **2a**, which was characterized by an acetate, **2b**, and sulfonate, **2c**. An instantaneous reaction took place between **2c** and ammonia to give the ureido acid and amide, **3a** and **3b**, respectively. The isomers **4a** and **4b** could not be detected. This type of Lossen degradation parallels that of *N*-sulfonyloxyphthalimides with amines reported by Kühle and Wegler.[9] However, these authors found their intermediate *o*-ureidobenzoic acid derivatives spontaneously cyclized to 2,4-quin-



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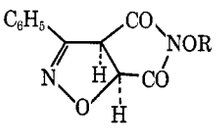
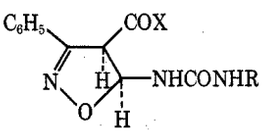
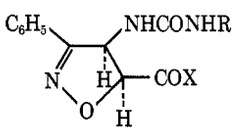


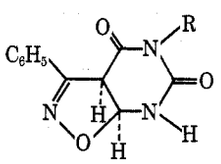
2a, R = H
b, R = $COCH_3$
c, R = $SO_2C_6H_5$



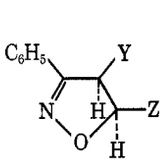
3a, X = OH; R = H
b, X = NH_2; R = H
c, X = OH; R = CH_3
d, X = $NHCH_3$; R = CH_3



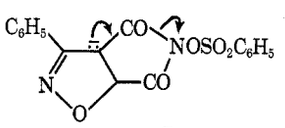
4a, X = OH; R = H
b, X = NH_2; R = H



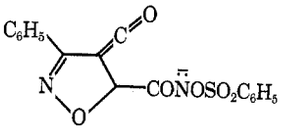
5a, R = H
b, R = CH_3



6a, Y = CO_2H; Z = $CONH_2$
b, Y = H; Z = $CONH_2$
c, Y = CO_2H; Z = CONHOH



7



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(1) Abstracted from the Ph.D. Dissertation of W. J. T., University of Illinois (Medical Center), 1972.

(2) L. Bauer and C. S. Mahajanshetti, *J. Heterocycl. Chem.*, **5**, 331 (1968), and references cited therein.

(3) Recent papers in this field are by G. Desimoni and P. Grünanger, *Gazz. Chim. Ital.*, **98**, 25 (1968); *Tetrahedron*, **23**, 687 (1967); P. Rajagopalan and C. N. Talaty, *ibid.*, **23**, 3541 (1967).

(4) These esters are readily available from the addition of acetylenedicarboxylic esters to nitrile oxides; see ref 5.

(5) C. Grundmann and P. Grünanger, "The Nitrile Oxides," Springer-Verlag, New York, N. Y., 1971.

(6) (a) This anhydride was synthesized in somewhat larger scale from the original paper of A. Quilico, G. S. D'Alcontres, and P. Grünanger, *Gazz. Chim. Ital.*, **80**, 479 (1950); N. S. Isaacs, "Experiments in Physical Organic Chemistry," Macmillan, London, 1969, p 261.

(7) A. Quilico, "Five- and Six-Membered Compounds with Nitrogen and Oxygen," Interscience, New York, N. Y., 1962 p 95.

(8) This approach was demonstrated originally by us [L. Bauer and S. Miarka, *J. Amer. Chem. Soc.*, **79**, 1983 (1957)], later by Kühle and Wegler (ref 9). A recent example of this type of degradation is described by V. L. Plakidin, N. M. Zadorozhnyi and Z. I. Krasota, *J. Org. Chem. USSR*, **6**, 1493 (1970).

(9) E. Kühle and R. Wegler [*Justus Liebigs Ann. Chem.*, **616**, 183 (1958)] found that *N*-(*p*-chlorobenzenesulfonyloxy)phthalimide rearranged with gaseous ammonia at 25° in benzene–dioxane and gave *o*-ureidobenzamide (72%), but cyclized in 10% aqueous ammonia solution. It is plausible that the driving force for this cyclization was the formation of the aromatic 2,4-quinazolinedione.