The Reaction of Phenyl Isocyanate with *N*-Methyl-2-pyrrolidinone

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Received February 16, 1973

The reaction of 1-methyl-2-pyrrolidinone with phenyl isocyanate at 140 and 210° was studied. Besides 1-methyl-2-phenyliminopyrrolidine (**2**), a pyrroline carbanilide (**7**) and pyrroloquinolines (**10, 11**) and pyrrolopyrimidines (**12, 13**) are formed. The reaction mechanism is discussed and earlier reported findings on this reaction are corrected.

Aryl isocyanates react at elevated temperature with a variety of compounds containing carbonyl groups to give imines and carbon dioxide. The method is used for the preparation of azomethines, ketimines, amidines, and guanidines from the corresponding aromatic aldehydes, benzophenones, and N-persubstituted amides and ureas.[1-5] In some cases, other products are obtained owing to successive interaction of the formed imines with excess isocyanate. Good examples are the extensively studied reactions of *N,N*-dimethylformamide[6-8] and *N,N*-dimethylacetamide[9] with aryl isocyanates.

In connection with thermostability studies of *N*-methyl-2-pyrrolidinone (NMP, **1**)-isocyanate mixtures, we found that phenyl isocyanate reacts with **1** at temperatures above 100°. Generation of carbon dioxide was observed when an equimolar mixture of the two components was kept at 140° for 16 hr. During the reaction the isocyanate was consumed completely (as shown by the disappearance of the NCO band at 2270 cm^{-1} in the ir) while about 65% of **1** remained unchanged.

N-Methyl-2-phenyliminopyrrolidine (**2**), expected to be the main product, can be isolated after removal of unreacted **1** from the semisolid reaction mixture by distillation in only 18% yield (based on converted **1**). From the residue seven solid compounds were isolated by repeated fractional crystallization from different solvent combinations (see Table I and Scheme I). Carbanilide, triphenyl isocyanurate, the pyrroline-3-carboxanilide **7** and the hexahydropyrrolopyrimidine **12** are the major products, whereas the pyrroloquinolines **10** and **11** and the hexahydropyrrolopyrimidine **13** together account for only 1-2%.

Structure assignments of the new compounds are based on elemental analysis, nmr, ir, and in some cases mass spectroscopy (see Experimental Section); the known compounds (carbanilide, triphenyl isocyanurate, and **2**) were compared with authentic samples. The yields given vary somewhat with work-up methods but are correct in the order of magnitude.

The product ratio can be changed drastically by increasing the reaction temperature. For example, a 1:1 mixture of **1** and phenyl isocyanate kept for 4 hr at 210° yielded 41% of **2**, only 7% of the pyrrolopyrimidine **12**, and 16% of the pyrroloquinoline **11** (58% of **1** is recovered unchanged). The yield of the pyrroloquinoline **10** remained under 2% and **7**, **13**, and triphenyl isocyanurate could not be detected among the reaction products.

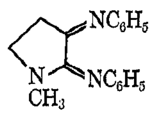
TABLE I

YIELDS[a] OF PRODUCTS OBTAINED IN REACTIONS OF *N*-METHYL-2-PYRROLIDINONE WITH PHENYL ISOCYANATE AT 140 AND 210°

Compd	Yield, % 140°	Yield, % 210°
1-Methyl-2-phenyliminopyrrolidine (**2**)	18	41
1-Methyl-2-phenylamino-2-pyrrolino-3-carboxanilide (**7**)	18	
1-Methyl-4-hydroxy-1,2-dihydro-3*H*-pyrrolo[2,3-*b*]quinoline (**10**)	0.4	1.8
1-Methyl-4-anilino-1,2-dihydro-3*H*-pyrrolo[2,3-*b*]quinoline (**11**)	0.8	16
7-Methyl-1,3-diphenyl-2,4-dioxohexahydro-7*H*-pyrrolo[2,3-*d*]pyrimidine (**12**)	31	7
7-Methyl-1,3-diphenyl-4-phenylimino-2-oxohexahydro-7*H*-pyrrolo[2,3-*d*]pyrimidine (**13**)	<0.2	

[a] Yields based on consumed **1**.

These results are in contrast to a recent report in which a mixture of NMP and phenyl isocyanate (molar ratio 1:1) was kept for 4 hr at 235°[10] and carbanilide and 1-methyl-2,3-diphenyliminopyrrolidone (**14**), mp 227-228°, were claimed to be the reaction products. For **14**, the structure of **9** is also discussed as a possibility but discarded, since the ir spectrum of the product did not show absorption bands characteristic of the >C=C=N- group. The given C, H, and N values of the elemental analysis do agree better with a molecular formula of $C_{18}H_{17}N_3$ (**9** or **11**) rather than $C_{17}H_{17}N_3$ (**14**). Despite a difference in melting points for **14** (227-228°) and **11** (238°), a comparison of the ir (which also differ slightly, possibly owing to deviations in the spectrophotometers used) and nmr spectra (**14** taken in CF_3COOH, **11** in CD_3COOD) suggests the identity of the compounds. The two triplets for the four protons of the neighboring CH_2 groups show that positions 4 and 5 on the pyrrolidine ring are not involved in the product formation.



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(1) E. Niwa, H. Aoki, H. Tanaka, K. Munakata, and M. Namiki, *Chem. Ber.*, **99**, 3932 (1966).

(2) H. Staudinger and R. Endle, *Ber. Deut. Chem. Ges.*, **50**, 1042 (1917).

(3) R. Richter, *Chem. Ber.*, **102**, 938 (1969).

(4) M. L. Weiner, *J. Org. Chem.*, **25**, 2245 (1960).

(5) P. A. Argabright and V. J. Sinkey, *Chem. Ind. (London)*, 857 (1966).

(6) H. Ulrich, B. Tucker, F. A. Stuber, and A. A. R. Sayigh, *J. Org. Chem.*, **33**, 3928 (1968).

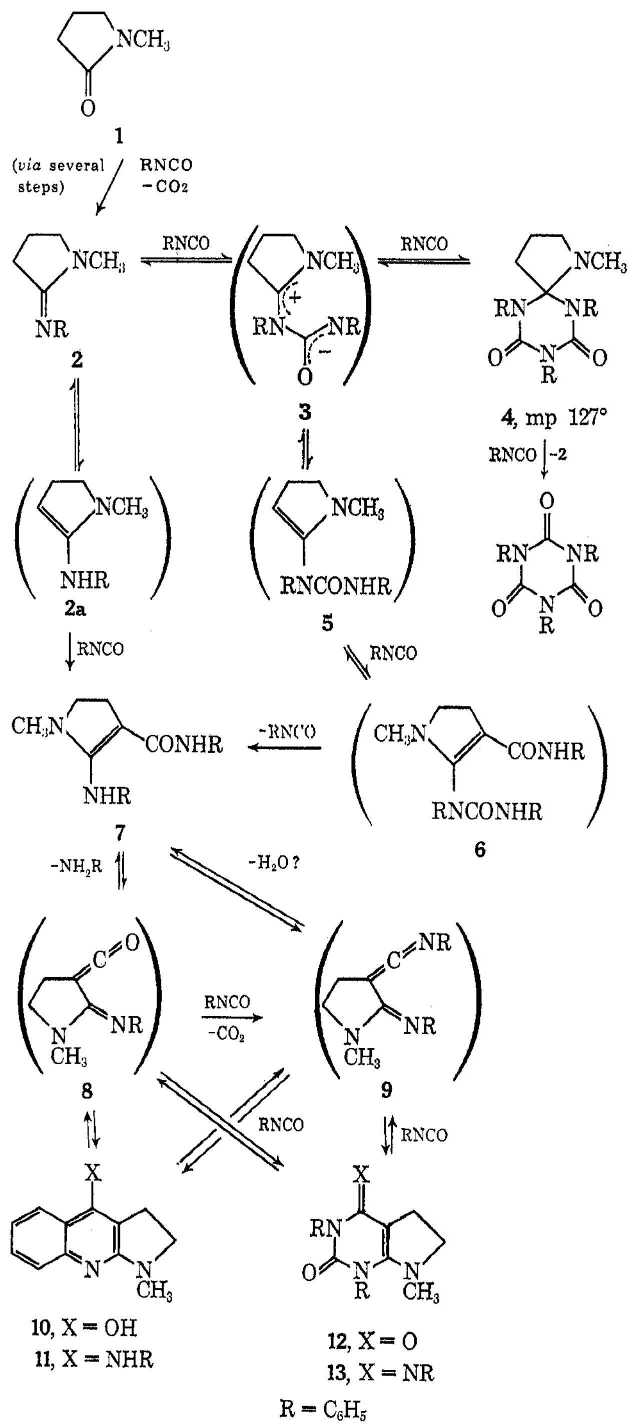
(7) E. Dyer, T. E. Majewski, and J. D. Travis, *J. Org. Chem.*, **33**, 3931 (1968).

(8) R. Richter and H. Ulrich, *J. Org. Chem.*, **36**, 2005 (1971).

(9) E. Dyer, T. E. Majewski, T. J. Nycz, and J. D. Travis, *J. Heterocycl. Chem.*, **9**, 955 (1972).

(10) S. Terney, J. Keating, J. Zielinski, J. Hakala, and H. Sheffer, *J. Polym. Sci., Part A-1*, **8**, 683 (1970).

SCHEME I

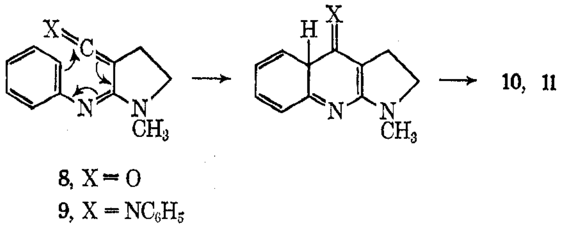


A possible pathway for the formation of all the reaction products is given in Scheme I. The conversion of the amide **1** into the amidine **2** proceeds *via* [2 + 2] cycloaddition with phenyl isocyanate followed by ring scission of the adduct to give **2** and carbon dioxide. Reactions of this type are described in detail elsewhere.[1,2] Subsequent interaction of the amidine **2** with phenyl isocyanate proceeds *via* substitution in the 3 position to give **7**. In Scheme I two possible pathways to the substituted vinylogous urea **7** are shown, which basically differ in the site of the initial isocyanate attack. In the sequence **2** → **3** → **5** → **6** → **7**, isocyanate attacks first on the exocyclic imino nitrogen, whereas in **2** → **2a** → **7** isocyanate attack is preceded by proton shift in **2** to the ketene aminal **2a**. Both reactions are known to occur with other imines and iso(thio)cyanates.[11-16]

Since **2** and phenyl isocyanate form the 2:1 cycloadduct **4** at room temperature in high yield[17] in a reaction generally known to proceed *via* a 1,4 dipole of type **3**,[18,19] it seems logical to assume an initial N attack of isocyanate also for the formation of **7**. Proton shift in **3** and C attack of phenyl isocyanate on the substituted ketene aminal **5** is followed by the loss of 1 mol of isocyanate initially bonded to the exocyclic nitrogen. The last step, **6** → **7**, is aided by a basicity decrease on the exocyclic nitrogen owing to the electron-withdrawing effect of the carboxamide group in position 3. In this reaction sequence phenyl isocyanate acts as catalyst in the proton shift in **2** *via* **3** to **5**.

No evidence for a thermal equilibrium **2** ⇄ **2a** could be obtained by independently recording the ir spectrum of a solution of **2** in *o*-dichlorobenzene in the NH stretching region (2900–3500 cm^{-1}) at various temperatures (32–165°). If any **2a** is formed at elevated temperatures, the amount is small and below the sensitivity limits of the spectrophotometer.

The trimerization of phenyl isocyanate to triphenyl isocyanurate might involve the compounds **2**, **3**, and **4**, as outlined in the reaction scheme, but a catalytic effect can also be expected from **1**, since it is known that N-substituted amides such as *N*,*N*-dimethylformamide trimerize aryl isocyanates.[20] Thermal dissociation of the vinylogous urea **7** could give either aniline and the phenylimino ketene **8**, or water and the phenylimino-*N*-phenyl ketenimine **9** (water or aniline would be constantly removed from the mixture by reacting with phenyl isocyanate to produce carbanilide, which has been isolated from the reaction mixture). Both ketene intermediates **8** and **9** can undergo cycloaddition reactions as shown in Scheme I. In the presence of excess phenyl isocyanate, intermolecular [4 + 2] addition produces the pyrimidine derivatives **12** and **13**, while intramolecular cyclization, as indicated in eq 1, gives the pyrroloquinolines **10** and **11**.



→ 10, 11 (1)

Reactions with a similar cyclization step were reported recently[21,22] and related intermediates are also

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(11) R. Richter, *Chem. Ber.*, **105**, 82 (1972).

(12) L. Capuano and H. J. Schrepfer, *Chem. Ber.*, **105**, 2539 (1972).

(13) S. J. Love and J. A. Moore, *J. Org. Chem.*, **33**, 2361 (1968).

(14) J. P. Chupp and E. R. Weiss, *J. Org. Chem.*, **33**, 2357 (1968).

(15) R. Huisgen, R. Grashey, J. M. Vernon, and R. Kunz, *Tetrahedron*, **21**, 3311 (1965).

(16) R. Richter and H. Ulrich, *Justus Liebigs Ann. Chem.*, **743**, 10 (1971).

(17) The absence of **4** among the products of reactions at 140 and 210° is not surprising, since the compound melts under dissociation at 130 132°.

(18) See R. Richter, *Chem. Ber.*, **101**, 3002 (1968).

(19) R. Huisgen, M. Morikawa, K. Herbig, and E. Brunn, *Chem. Ber.*, **100**, 1094 (1967).

(20) H. Ulrich, "Cycloaddition Reaction of Heterocumulenes," Academic Press, New York, N. Y., 1967, p 125.

(21) G. Kollenz, H. Igel, and E. Ziegler, *Monatsh. Chem.*, **103**, 450 (1972).

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discussed in the Conrad–Limpach quinoline synthesis.[23,24]

Since N,N′-disubstituted ureas are not known to lose water to give carbodiimides on heating, the direct formation of **9** from **7** is doubtful. It is more likely that **9** is formed from **8** and phenyl isocyanate in another [2 + 2] cycloaddition reaction.

Confirmation for a conversion of **8** to **9** was obtained in the thermolysis of **12**, which yielded not only the expected pyrroloquinoline **10** (32%) but also **11** (36%).

Experimental Section[25]

Reaction of 1-Methyl-2-pyrrolidinone with Phenyl Isocyanate. A. At 140°.—A mixture of 238 g (2.0 mol) of phenyl isocyanate and 198 g (2.0 mol) of **1** was kept at 140° for 16 hr. Carbon dioxide was generated during the first period of reaction, totaling 32.2 g, while the color of the mixture turned to deep yellow. After cooling to room temperature, 129.2 g of **1** (65%) was recovered by vacuum distillation, bp 40° (0.1 mm). A semisolid residue remained from which, on treatment with methanol and methanol–diethyl ether, solid fractions were obtained. The following separation procedure was typical, and reproducible with only minor deviations in amounts and composition of the fractions.

Treatment of the residue with 400 ml of methanol gave a colorless precipitate, 149 g (fraction 1), after filtering and washing with 100 ml of methanol. On concentrating the filtrate, a second portion of crystals, 51 g (fraction 2), did separate. The remaining filtrate was further concentrated and distilled at reduced pressure, giving 22.5 g of **2** (fraction 3), bp 85–90° (0.01 mm), which was identical in ir comparison with authentic material.[26]

Treatment of the distillation residue with methanol gave 7.2 g (fraction 4), which was collected by filtration and washed with methanol. The combined filtrate and methanol washings gave, on cooling to −10° for 17 hr, another crop of crystals, 5.9 g (fraction 5), after filtration and methanol washing. Addition of diethyl ether to the filtrate to beginning turbidity and scratching led to the separation of more crystals, which were collected after standing for several hours, 12.5 g (fraction 6) after washing with methanol–diethyl ether and finally diethyl ether. Another crop of crystalline material, 2.1 g (fraction 7), was obtained on concentrating the filtrate *in vacuo*. Filtrate and methanol–diethyl ether washings were again concentrated at reduced pressure, which led to the separation of 3.8 g (fraction 8) of colorless crystals.

No further solid material could be obtained from the remaining yellow-brownish resinous residue. Tlc with benzene–acetone (1:1) showed that most of the solid fractions were mixtures of two or more components.

Separation of Solid Fractions. Fraction 1.—The mixture was dissolved in 300 ml of hot *N,N*-dimethylformamide (DMF) and the resulting solution was diluted with 700 ml of diethyl ether, which caused separation of 39.0 g of **7-methyl-1,3-diphenyl-2,4-dioxo-1,2,3,4,5,6-hexahydro-7*H*-pyrrolo[2,3-*d*]pyrimidine** (**12**), mp 227–228°, colorless plates, ir (KBr) 1698, 1668 cm⁻¹ (C═O), R_f 0.71.

Anal. Calcd for $C_{19}H_{17}N_3O_2$: C, 71.45; H, 5.37; N, 13.16. Found: C, 71.41; H, 5.36; N, 13.43.

On concentrating the DMF–diethyl ether filtrate *in vacuo* a second crop of crystals was obtained. The material, a mixture by tlc, was dissolved in 1700 ml of hot acetone and the solution deposited on cooling 36.5 g of carbanilide, mp 245°, R_f 0.9, identical in ir comparison with authentic material. On evaporation of the filtrate to dryness colorless crystals were obtained, which were recrystallized from warm DMF. Thus, 20.7 g of **1-methyl-2-phenylamino-2-pyrrolino-3-carboxanilide** (**7**), mp 170–172° (from acetone), colorless needles, were isolated: ir (KBr) 3250 (NH), 1662, 1639 cm^{-1} (C═O, C═N, or C═C); R_f 0.24.

Anal. Calcd for $C_{18}H_{19}N_3O$: C, 73.79; H, 6.53; N, 14.33. Found: C, 73.75; H, 6.53; N, 14.33.

From the filtrates, more material of the same compounds was isolated by repeated recrystallization from the named solvents totaling 43.9 g of carbanilide, 42.9 g of **12**, and 26.0 g of **7**.

Fraction 2.—On treating the mixture with DMF–diethyl ether and acetone as described above for fraction 1, a total of 3.7 g of carbanilide, 22.0 g of **12**, and 10.6 g of **7** could be isolated in pure form. The combined filtrates yielded on evaporation to dryness a crystal mixture containing a further component. Separation by column chromatography on silica gel (Bio-Rad Laboratories, Richmond, Calif.) with DMF as eluent gave 0.5 g of **7-methyl-1,3-diphenyl-4-phenylimino-2-oxo-1,2,3,4,5,6-hexahydro-7*H*-pyrrolo[2,3-*d*]pyrimidine** (**13**), mp 258–260° (from acetone), colorless needles, ir (KBr) 1682, 1646 cm^{-1} (C═O, C═N), R_f 0.82.

Anal. Calcd for $C_{25}H_{22}N_4O$: C, 76.12; H, 5.62; N, 14.20. Found: C, 76.34; H, 5.64; N, 14.10.

Fraction 4.—Tlc did indicate the presence of only one component, which was identified as triphenyl isocyanurate, mp 275°, R_f 0.9, identical in ir comparison with authentic material.

Fraction 5.—The separation into components was omitted, since tlc did show only the presence of carbanilide, **12**, **7**, and traces of triphenyl isocyanurate.

Fraction 6.—Recrystallization from hot DMF gave a crystal mixture, which on boiling with acetone left 0.6 g of **1-methyl-4-hydroxy-1,2-dihydro-3*H*-pyrrolo[2,3-*b*]quinoline** (**10**), undissolved: mp >300° (from DMF); colorless crystals; ir (KBr) 1638, 1589 cm^{-1} (quinoline); uv λ_{max}^{EtOH} (log ε) 222 nm (4.47), 243 (4.34), 319 (4.17); R_f 0.43 (benzene/DMF, 7:3).

Anal. Calcd for $C_{12}H_{12}N_2O$: C, 71.98; H, 6.04; N, 13.99; mol wt, 200. Found: C, 71.90; H, 6.06; N, 13.92; mol wt, 200 (from mass spectral data).

From the acetone filtrate 5.9 g of **12** and from the DMF filtrate 5.0 g of triphenyl isocyanurate were obtained.

Fraction 7 did consist of pure **7**.

Fraction 8.—Repeated recrystallizations from DMF–diethyl ether gave 1.7 g of **1-methyl-4-anilino-1,2-dihydro-3*H*-pyrrolo[2,3-*b*]quinoline** (**11**): mp 238°; colorless crystals; ir (KBr) 1640, 1600, 1595, 1574 (quinoline system), 765, 755, 698 cm^{-1} (CH out of plane vibrations for mono- and 1,2-disubstituted benzene); nmr (CD_3COOD) δ 2.40 (t, 2, CH_2, J = 8 Hz), 3.15 (s, 3, CH_3), 3.68 (t, CH_2, J = 8 Hz), 6.98–8.45 (m, 9, aromatic H), NH exchanged by CD_3COOD; uv λ_{max}^{EtOH} (log ε) 206 nm (4.51), 247 (4.52), 330 (4.10); R_f 0.62.

Anal. Calcd for $C_{18}H_{17}N_3$: C, 78.51; H, 6.22; N, 15.26; mol wt, 275. Found: C, 78.79; H, 6.29; N, 15.38; mol wt, 275 (from mass spectral data).

The following total amounts of product were obtained: 22.5 g (18%) of **2**, 38.7 g (18%) of **7**, 0.6 g (0.4%) of **10**, 1.7 g (0.8%) of **11**, 70.8 g (31%) of **12**, 0.5 g (<0.2%) of **13**, 47.6 g of carbanilide, and 16.0 g of triphenyl isocyanurate.

B. At 210°.—A mixture of 99 g (1.0 mol) of **1** and 119 g (1.0 mol) of phenyl isocyanate was kept for 4 hr at 210°. During the reaction, carbon dioxide was generated and the mixture became highly viscous. For isolation and identification the procedure outlined above was followed. The unreacted **1**, 58.4 g (59%), was removed by initial vacuum distillation, bp 40° (0.1 mm), and solid fractions were obtained from the residue on treatment with methanol and methanol–diethyl ether; the amidine **2**, 29.4 g (41%), was also obtained by vacuum distillation from the residue at bp 80–90° (0.1 mm). By repeated recrystallization from solvents as outlined in A, the following amounts of products were obtained pure: 1.5 g (1.8%) of **10**, 18 g (16%) of **11**, and 9.2 g (7%) of **12**.

4′,6′-Dioxo-1-methyl-1′,3′,5′-triphenylspiro[pyrrolidine-2,2′-perhydro-*s*-triazine] (**4**).[18]—A mixture of 3.5 g (0.02 mol) of **2** and 4.8 g (0.04 mol) of phenyl isocyanate was kept for 17 hr at room temperature. Colorless crystals were formed during the reaction. Treatment with diethyl ether and filtration yielded 8.0 g (97%) of **4**, mp 130–132° (from chloroform–diethyl ether), ir (KBr) 1670, 1710 cm^{-1} (C═O).

Anal. Calcd for $C_{25}H_{24}N_4O_2$: C, 72.79; H, 5.87; N, 13.58. Found: C, 72.44; H, 5.92; N, 13.46.

Thermal Decomposition of 12.—A sample of 3.0 g (<0.01 mol) of **12** was heated in an open flask to ∼240° and kept at that

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[23] M. Conrad and L. Limpach, *Ber. Deut. Chem. Ges.*, **20**, 944 (1887).

[24] H. M. Blatter and H. Lukaszewski, *Tetrahedron Lett.*, 855 (1964).

[25] Infrared, uv, mass, and nmr spectra were recorded using P-E 625, Cary 14, CH-4, and Varian T-60 (TMS as internal standard) spectrophotometers, respectively. Tlc was performed on precoated silica gel plates (Quantum Ind., Fairfield, N. J.). All melting points were determined on a Fisher-Johns apparatus and are uncorrected.

[26] H. Bredereck and K. Bredereck, *Chem. Ber.*, **94**, 2278 (1961).

temperature for 5 min. Evolution of a colorless gas was observed. The reaction mixture was diluted with 10 ml of methanol, and the yellowish, crystalline precipitate was collected and washed with ether. Thus, 1.6 g of a mixture of starting material and 11 was obtained. Recrystallization from DMF–diethyl ether yielded 0.95 g (36%) of 11, mp 237°, identical in ir comparison with material obtained above. Dilution of the methanol filtrate with diethyl ether gave 0.6 g (32%) of **10**, mp >300°, identical in ir comparison with material obtained above.

Registry No.—**1,** 872-50-4; **2,** 7544-93-6; **4,** 40387-20-0; **7,** 40387-21-1; **10,** 40387-22-2; **11,** 40387-23-3; **12,** 40387-24-4; **13,** 40387-25-5; phenyl isocyanate, 103-71-9.

Quinazolines. I. The Oxidation of Indole-1,2-dicarboximides and Subsequent Conversion of Their Oxidation Products to Quinazolinones

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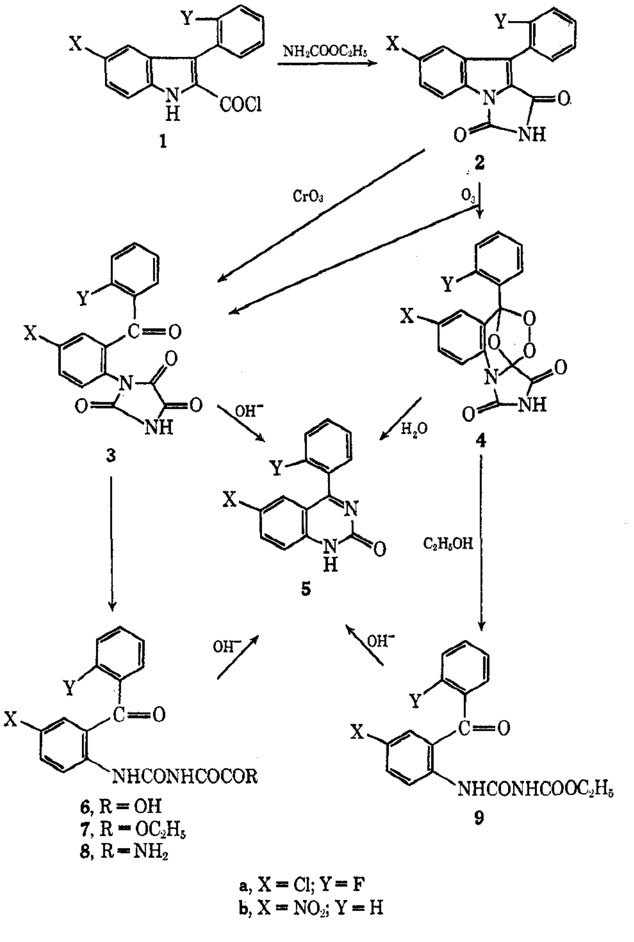
Received March 6, 1973

Chromic acid oxidation of indole-1,2-dicarboximides 2 led to imidazolidinetriones 3, which on hydrolysis with base gave the corresponding dihydroquinazolinones 5. Ozonolysis of 2, on the other hand, resulted in the formation and isolation of crystalline ozonides 4. On simply heating with water, the ozonides 4 were readily converted into 5 in nearly quantitative yields. The mechanism for this conversion is discussed.

In the past few years, we have reported the syntheses of 1,4-benzodiazepine ring systems by the oxidative ring cleavage of indoles bearing a substituent such as 2-aminomethyl,[1] 1-aminoethyl,[2] and 1-phthalimidoacetyl groups.[3] We have now extended our studies to another heterocyclic system, dihydroquinazolinone. Several reports have appeared in the literature on syntheses of the quinazoline ring system by rearrangement reactions of other heterocyclic structures, such as isatins,[4] quinolines,[5] and 1,4-benzodiazepines,[6] bearing an N-monosubstituted carbamoyl group at N-1. These methods, however, led only to the tetrahydroquinazolinones, rather than to the dihydro derivatives, because of the presence of a substituent on the carbamoyl nitrogen. We turned our attention to the synthesis and oxidation of indole-1,2-dicarboximides 2. By analogy with the previously described conversion[2] of pyrazino[1,2-*a*]indol-1(2*H*)-ones into 2,3-dihydro-1*H*-1,4-benzodiazepines, compounds 2 seemed likely to produce the desired dihydroquinazolinones 5 by oxidative cleavage of the indole ring, followed by hydrolysis of the imidazolidinetriones 3 thus obtained (Scheme I).

The synthesis of 2 was achieved by condensation of indole-2-carboxylic acid chlorides[1c,d] with urethane.[7] The ir spectrum of 2 showed the expected NH absorptions and two carbonyl bands at relatively high frequencies (1790 and 1728 cm^{-1}), consistent with the hydantoin structure and in good agreement with those observed in the spectrum of *N*-phenylindole-1,2-dicarboximide.[8]

SCHEME I



Chromic Acid Oxidation.—When compounds 2 were treated with chromic acid in acetic acid at 60–70°, the expected imidazolidinetriones 3 were obtained together with small amounts of 5. While **3a** was isolated only in amorphous form, **3b** formed a crystalline etherate, which on heating *in vacuo* was converted to free, crystalline **3b**. Their ir spectra showed carbonyl bands at 1750 cm^{-1} with a shoulder near 1790 cm^{-1}, owing to imidazolidinetrione structure,[9] as well as the benzophenone C=O absorption at 1670 cm^{-1}.

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[1] (a) H. Yamamoto, S. Inaba, T. Hirohashi, and K. Ishizumi, *Chem. Ber.*, **101**, 4245 (1968); (b) S. Inaba, T. Hirohashi, and H. Yamamoto, *Chem. Pharm. Bull.*, **17**, 1263 (1969); (c) S. Inaba, K. Ishizumi, and H. Yamamoto, *ibid.*, **19**, 263 (1971); (d) S. Inaba, K. Ishizumi, K. Mori, and H. Yamamoto, *ibid.*, **19**, 722 (1971).

[2] S. Inaba, K. Ishizumi, T. Okamoto, and H. Yamamoto, *ibid.*, **20**, 1628 (1972).

[3] K. Ishizumi, K. Mori, S. Inaba, and H. Yamamoto, *ibid.*, **21**, 1027 (1973).

[4] (a) L. Capuano and M. Welter, *Chem. Ber.*, **101**, 3671 (1968); (b) L. Capuano, M. Welter, and R. Zander, *ibid.*, **103**, 2394 (1970).

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[6] T. Masuda, S. Fujii, and K. Naito, Japanese Patent 15,500 (1971); *Chem. Abstr.*, **75**, 36104z (1971).

[7] The synthesis of indole-1,2-dicarboximide itself is achieved by condensing ethyl indole-2-carboxylate with the sodium derivative of urethane: J. D. Dutcher and A. Kjaer, *J. Amer. Chem. Soc.*, **73**, 4139 (1951).

[8] E. P. Papadopoulos and S. B. Bedrosian, *J. Org. Chem.*, **33**, 4551 (1968).

[9] H. Ulrich and A. A. R. Sayigh, *J. Org. Chem.*, **30**, 2781 (1965).