

E = weight of quinone which would be left if oxidation were to diketo acid.

A. Gram.	B. cc.	C. Gram.	D. Gram.	E. Gram.
0.0226	1	0.0209	0.0210	0.0206
0.0226	6	0.0126	0.0131	0.0108
0.0226	8	0.0110	0.0100	0.0068
0.0226	10	0.0087	0.0069	0.0023

The results given in this table are not very good because the method was inaccurate but they confirm the conclusion arrived at from the previous experiments that there is no intermediate stage in the oxidation of β -naphthoquinone to phthalonic acid in alkaline solution. A series of experiments, in which the rate of oxidation of the quinone in alkaline solution was determined, also gave no indication of the existence of an intermediate product.

Further experiments to prepare the diketo acid by the saponification of phthalyl cyanide are now being carried out.

UNIVERSITY OF TORONTO.

[CONTRIBUTION FROM THE HAVEMEYER LABORATORIES OF COLUMBIA UNIVERSITY,
No. 174.]

RESEARCHES ON QUINAZOLINES (TWENTY-FOURTH PAPER). ON OXALYL ANTHRANILIC COMPOUNDS AND QUIN- AZOLINES DERIVED THEREFROM.¹

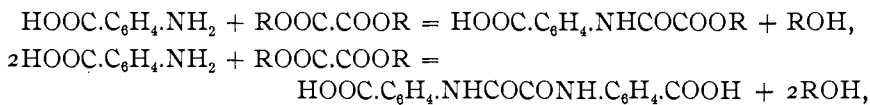
BY MARSTON TAYLOR BOGERT AND ROSS AIKEN GORTNER.

Received November 27, 1909.

In 1883, Kretschy² obtained by oxidation of kynurine, or of kynurenic acid, an acid which he called "kynuric acid." The succeeding year³ he showed that this "kynuric acid" was oxalyl anthranilic acid.

The following pages describe various derivatives of oxalyl anthranilic acid and condensation products obtained therefrom.

Mauthner and Suida⁴ have recorded the fact that when anthranilic acid is heated with ethyl oxalate, both oxalyl dianthranilic acid and ethoxalyl anthranilic acid are produced:



and we have used this method for the preparation of these substances, as well as for the methoxalyl anthranilic acid. Oxalyl dianthranilic

¹ Read at the meeting of the New York Section, March 5, 1909.

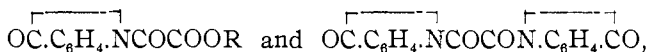
² *Monatsh. Chem.*, **4**, 157 (1883).

³ *Ibid.*, **5**, 30 (1884).

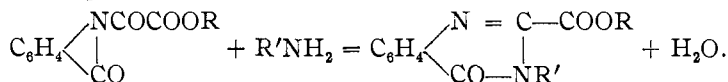
⁴ *Monatsh. Chem.*, **9**, 743 (1888).

acid was also prepared by the action of oxalyl chloride upon anthranilic acid.

By the action of acetic anhydride, these acyl anthranilic acids were changed to acyl anthranils,



which were then treated with various primary amines to get the corresponding quinazolines:¹



The reactions with the methoxalyl and ethoxalyl anthranils resulted in the formation of the expected quinazolines, the amines used being ammonia, methylamine, aniline, β -naphthylamine, hydrazine and phenylhydrazine.

Some of the results are, however, rather puzzling. Thus, when ethoxalyl anthranil was heated with alcoholic ammonia, the product was the

ammonium salt of the quinazoline acid, $\overline{\text{HN.CO.C}_6\text{H}_4.\text{N} = \text{C.COONH}_4}$; but when the anthranil was fused with urea, or boiled with an alcoholic

solution of urea, the quinazoline ester was obtained, $\overline{\text{HN.CO.C}_6\text{H}_4.\text{N} = \text{C.COOR}}$.

Ethoxalyl anthranil, heated with an alcoholic solution of methylamine,

gave the quinazoline amide, $\overline{\text{CH}_3\text{N.CO.C}_6\text{H}_4.\text{N} = \text{C.CONHCH}_3}$, and hydrazine similarly gave a hydrazide. Aniline, β -naphthylamine, and phenylhydrazine, on the other hand, did not displace the ester group, but gave quinazoline esters, even when the reaction was pushed to the point where a second molecule of the amine condensed with the keto

group of the quinazoline to an anil, $\overline{\text{C}_6\text{H}_5\text{N.C}(\text{:NC}_6\text{H}_5).\text{C}_6\text{H}_4.\text{N} = \text{C.COOR}}$.

Experimental.

Methoxalyl Anthranilic Acid, (1)HOOC.C₆H₄.NHCOCOOCH₃ (2).—An equimolecular mixture of anthranilic acid and methyl oxalate was heated at 140–55°. The mixture melted, methyl alcohol distilled off, and after 30 minutes' heating a hard cake remained. This was pulverized fine, washed with cold water and the residue repeatedly extracted with dilute alcohol. From the alcoholic extracts, combined and concentrated, crystals were obtained which on recrystallization from water melted at 176.5° (corr.). For analysis, they were further purified by recrystalli-

¹ Anschütz, Schmidt and Greiffenberg, *Ber.*, 35, 3480 (1902). Bogert and Chambers, *THIS JOURNAL*, 27, 649 (1905). Bogert and Seil, *Ibid.*, 27, 1305 (1905). Bogert and Steiner, *Ibid.*, 27, 1327 (1905); *et al.*

zation from a mixture of alcohol and benzene, washed with ligroin, and dried at 110°.

Calculated for $C_{10}H_9O_3N$: N, 6.27.
 Found: N, 6.17.

The acid dissolves readily in cold sodium carbonate solution with evolution of carbon dioxide. It is also soluble in hot water, alcohol, or benzene; difficultly soluble in cold water, practically insoluble in chloroform or ligroin.

In this preparation, some oxalyl dianthranilic acid is also formed.

Ethoxalyl Anthranilic Acid, $(1)HOOC.C_6H_4.NHCOCOOC_2H_5(2)$.—Mauthner and Suida¹ have already reported on the formation of this compound from anthranilic acid and ethyl oxalate. It was, however, first obtained by von Baeyer² by oxidizing ethyl indoxylate with sulphuric acid and potassium dichromate. von Baeyer gives its melting-point as 180–1°.

Twenty grams anthranilic acid were mixed with slightly more than an equimolecular amount of ethyl oxalate and the mixture heated for an hour at 140–50°, followed by ten minutes further heating at 160°. When cold, the melt was pulverized and extracted with boiling water. From the hot aqueous extract, colorless needles of ethoxalyl anthranilic acid separated on cooling, melting sharply at 184° (corr.). Yield, 27 grams. From the residue insoluble in water, 1.4 grams oxalyl dianthranilic acid were isolated.

Oxalyl Dianthranilic Acid (Oxanilide Di-o-carbonic Acid), $(1)HOOC.C_6H_4.NHCOCONH.C_6H_4.COOH(2)$.—That this acid is formed by heating together anthranilic acid and ethyl oxalate has already been noted by Mauthner and Suida.¹

We prepared it by heating 1 molecule of methyl or ethyl oxalate with 2 molecules of anthranilic acid for half an hour to an hour at 140–155°, removing methoxalyl- or ethoxalyl anthranilic acid by extracting the crude product with boiling water and dilute alcohol. We also prepared it by dissolving anthranilic acid in dry benzene and adding oxalyl chloride. The reaction was violent, with evolution of copious fumes of hydrochloric acid, and the precipitate was purified by washing (digestion) with benzene and with alcohol.

The purified products obtained by the above methods were identical. The substance is a white powder, decomposing at about 330° (uncorr.), and is insoluble in the ordinary neutral organic solvents. When freshly prepared from methyl oxalate, it seems to be appreciably soluble in alcohol, but on standing becomes practically insoluble in it.

Methoxalyl Anthranil, $OC.C_6H_4.N.COCOCH_3$.—Methoxalyl anthranilic

¹ *Loc. cit.*

² *Ber.*, 15, 777 (1882).

acid was boiled for several minutes with excess of acetic anhydride and the solution then concentrated to crystals. These crystals were pressed dry on a porous plate and recrystallized from dry benzene. They then melted sharply at 177.5° (corr.). As the methoxalyl anthranilic acid has a melting point very close to this (176.5° corr.), it was thought at first that we had only recovered the original substance unaltered, but on mixing with some of the methoxalyl anthranilic acid, the melting point was lowered to 150° (uncorr.). The pure dry substance gave the following figures on analysis:

Calculated for $C_{10}H_7O_4N$: N, 6.83.

Found: N, 7.02.

This anthranil crystallizes from dry benzene or acetic anhydride in pale brownish needles. It is very sensitive to moisture, being hydrolyzed to methoxalyl anthranilic acid by ordinary commercial absolute alcohol (99.8 per cent.) or by ordinary glacial acetic acid.

Ethoxalyl Anthranil, $(1)OC.C_6H_4.N.COOCOC_2H_5$, was prepared in similar manner from the ethoxalyl anthranilic acid and acetic anhydride. Recrystallized from acetic anhydride, it forms large, colorless plates, melting at $129-130^{\circ}$ (corr.). Yield, 81 per cent. of the theoretical.

Calculated for $C_{11}H_9O_4N$: N, 6.40.

Found: N, 6.67.

In contrast with the above methyl derivative, this ethoxalyl anthranil is not very easily hydrolyzed. It may even be recrystallized from relatively dilute alcohol without suffering any extensive hydrolysis. Boiling water rapidly hydrolyzes it to the ethoxalyl anthranilic acid again.

Oxalyl Dianthranil, $OC.C_6H_4.N.COCON.C_6H_4.CO$.—Five grams oxalyl anthranilic acid were boiled for three hours with excess of acetic anhydride, the solid changing from white to yellow. The insoluble material was filtered out and washed with acetic anhydride. Yield, 3.4 grams. For analysis, this crude product was boiled again for three hours with acetic anhydride, washed with the anhydride, and dried at 110° .

Calculated for $C_{16}H_8O_4N_2$: C, 65.75; H, 2.74; N, 9.59.

Found: C, 65.20, 64.97, 65.81; H, 3.20, 2.87, 3.05; N, 9.98.

The purified oxalyl dianthranil is a yellow powder, melting with decomposition at about 345° (uncorr.), and is insoluble in the ordinary neutral solvents. Boiling water slowly hydrolyzes it to anthranilic and oxalic acids. It is readily hydrolyzed by hot concentrated hydrochloric acid.

4-Quinazolone-2-carboxylic Acid (*4-Hydroxyquinazoline-2-carboxylic Acid*), $HN.CO.C_6H_4.N = C.CO_2H \rightleftharpoons N = C(OH).C_6H_4.N = C.CO_2H$.

Ammonium Salt, $C_8H_5ON_2 \cdot COONH_4$.—Two grams ethoxalyl anthranil were added to a slight excess of ammonia dissolved in 95 per cent. alcohol. The anthranil dissolved with evolution of heat and a white precipitate suddenly separated. The mixture was heated to 100° , the precipitate filtered out and washed repeatedly with 95 per cent. alcohol. Yield of this ammonium salt, nearly theoretical.

Calculated for $C_8H_5O_3N_3$: N, 20.28.

Found: N, 20.26.

The ammonium salt crystallizes from water in long silky colorless needles, melting at 229° (corr.) with evolution of ammonia and carbon dioxide.

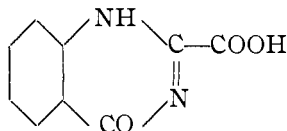
Free Acid.—Cold concentrated hydrochloric acid was added to the ammonium salt and then water. The free acid separated in a gelatinous condition and was crystallized from dilute alcohol. White silky needles resulted, softening at 227° and melting with evolution of carbon dioxide at 230° (corr.). The melt on cooling congealed in colorless needles, and re-melted at 214° (corr.). The melting point of 4-quinazolone (4-hydroxyquinazoline) as found by different investigators is variously stated from 209 to 216.5° .

Calculated for $C_8H_6O_3N_2$: N, 14.73.

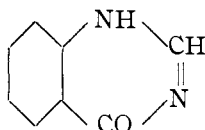
Found: N, 14.97.

The acid is easily soluble in hot alcohol, only slightly soluble in benzene, and very difficultly soluble in water. Boiled with mineral acids, it gradually loses carbon dioxide.

In 1885, Griess¹ described a compound which he called "carboxylcyanamidobenzoyl," and to which he assigned the formula



When this acid was heated, Griess found that it lost carbon dioxide giving another substance, which he named "carbimidamidobenzoyl" and formulated thus



Subsequent investigations of Weddige,² Knape,³ Bischler and Burkart,⁴

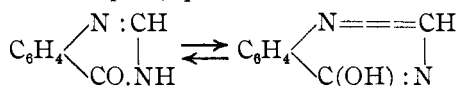
¹ *Ber.*, 18, 2418 (1885).

² *J. prakt. Chem.*, [2] 31, 124 (1885).

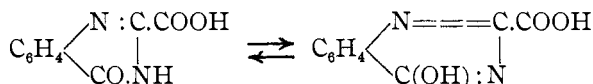
³ *Ibid.*, 43, 227 (1891).

⁴ *Ber.*, 26, 1349 (1893).

Niementovski,¹ and Bogert and Hand² showed that this "carbimidamidobenzoyl" was the simple 4-quinazolone and should be formulated



From this it follows that the formula for "carboxylcyanamidobenzoyl" should be written



Reisert and Grube³ have recently obtained this same acid by the rearrangement in dilute acid solution of *o*-cyanoxanilic acid, $\text{NC} \cdot \text{C}_6\text{H}_4 \cdot \text{NHCOCOOH}$.

Griess describes his acid as crystallizing in leaflets, but gives no melting point. Reisert and Grube, like ourselves, obtained the acid in needles. They give a melting point of 201–202°. We do not understand how they found so low a melting point, unless they did not recrystallize their product sufficiently. It does not appear from the article that they recrystallized their compound to constant melting point.

Ethyl Ester, $\text{C}_8\text{H}_8\text{ON}_2 \cdot \text{COOC}_2\text{H}_5$.—1.5 grams ethoxalyl anthranil and 0.42 gram urea were melted together at 125°. The temperature was then raised slowly to 140–150° and kept there for an hour. The cold melt was extracted with alcohol, and from the alcoholic extracts on cooling a crystalline material separated which was filtered out, washed with water and dried. The same substance was obtained when ethoxalyl anthranil (1 gram) and urea (0.33 gram) were boiled together in alcohol solution. For analysis, the compound was recrystallized from alcohol and dried at 110°.

Calculated for $\text{C}_{11}\text{H}_{10}\text{O}_3\text{N}_2$: N, 12.83.

Found: N, 12.90.

It crystallizes from alcohol in minute colorless needles, melting at 185.5° (corr.), and is insoluble in water or in mineral acids. It dissolves easily in solutions of the caustic alkalis, but not in strong sodium carbonate solution. Heated with aqueous ammonia, the ammonium salt of the acid (described above) results, apparently without any amide. That alcohol is split off by the action of alkalis was shown by the iodoform test.

3-Methyl-4-quinazolone-2-carbomethylamide, $\text{CH}_3\text{N} \cdot \text{CO} \cdot \text{C}_6\text{H}_4 \cdot \text{N} = \text{C} \cdot \text{CONHCH}_3$.—One molecule of ethoxalyl anthranil was added to an aqueous solution of slightly more than 2 molecules methylamine, and the solution

¹ *J. prakt. Chem.*, [2] 51, 564 (1895).

² *THIS JOURNAL*, 24, 1048 (1902).

³ *Ber.*, 42, 3713 and 3715 (1909).

concentrated to small bulk. On standing several days, beautiful pale-rose prisms separated, melting sharply at 160° (corr.). They were dried at 110° and analyzed:

Calculated for $C_{11}H_{11}O_2N_3$: N, 19.35.
Found: N, 19.51.

The compound is very soluble in water; much less soluble in alcohol.

Methyl 3-Phenyl-4-quinazolone-2-carboxylate, $C_6H_5N.CO.C_6H_4.N = C.COOC_6H_5$.—0.5 gram methoxalyl anthranil was mixed with a slight excess of aniline and the mixture heated carefully with the naked flame. As soon as the boiling commenced, the flame was removed and the action allowed to continue without further heating. Water was evolved and, on cooling, the mixture solidified. It was extracted with dilute alcohol, and from this extract small colorless plates separated on cooling, m. p. 203.5° (corr.) with previous softening in the neighborhood of about 120° (uncorr.). The yield was very poor. For analysis, the compound was recrystallized from dilute alcohol and dried at 110° .

Calculated for $C_{16}H_{12}O_3N_2$: N, 10.0.
Found: N, 9.78.

The ester is easily soluble in alcohol, but very slightly soluble in water. Heated with excess of aniline, it gives a difficultly soluble compound, probably similar to the anil described beyond.

Ethyl 3-Phenyl-4-quinazolone-2-carboxylate, $C_{14}H_9ON_2.COOC_2H_5$.—0.5 gram ethoxalyl anthranil was heated for a few minutes with excess of aniline, and the mass then cooled and extracted with 95 per cent. alcohol. Colorless plates separated from the alcoholic extract on cooling.

Calculated for $C_{17}H_{14}O_3N_2$: N, 9.50.
Found: N, 9.40.

This ester melts at 160° (corr.). It is but slightly soluble in water, easily soluble in alcohol. In cold potassium hydroxide solution, it is apparently insoluble, but dissolves on warming. It is not hydrolyzed readily by hot concentrated hydrochloric acid.

Ethyl 3-Phenyl-4-phenyliminoquinazoline-2-carboxylate,

$C_6H_5N.C(: NC_6H_5).C_6H_4.N = C.COOC_2H_5$.—This anil is obtained simultaneously with the above ester from the interaction of ethoxalyl anthranil and excess of aniline. If the heating is continued many minutes, the yield of anil is good. It can be separated easily from the keto ester by its insolubility in alcohol. For analysis, it was washed repeatedly with hot alcohol and dried at 110° .

Calculated for $C_{22}H_{19}O_2N_3$: N, 11.36.
Found: N, 11.40.

It is a colorless crystalline powder, insoluble in water and in all the

ordinary neutral organic solvents. It melts with effervescence at 291° (corr.).

Ethyl 3-β-Naphthyl-4-β-naphthyliminoquinazoline-2-carboxylate,

$C_{10}H_7N.C(: NC_{10}H_7).C_6H_4.N = C.COOC_2H_5$.—0.5 gram ethoxalyl anthranil and 0.5 gram β-naphthylamine were carefully melted together, and the temperature then gradually raised to the boiling point. The melt was allowed to cool, and was then thoroughly extracted with 95 per cent. alcohol. The insoluble residue was dried at 110° and analyzed.

Calculated for $C_{31}H_{23}O_2N_3$: N, 8.95.

Found: N, 8.83.

It is a grayish crystalline powder, insoluble in water, ethyl or amyl alcohol, acetone or benzene, and melts to an amber liquid at $253-4^{\circ}$ (corr.).

Ethyl 3-Anilino-4-quinazolone-2-carboxylate, $C_6H_5NHN.CO.C_6H_4.N = C.COOC_2H_5$.—An equimolecular mixture of ethoxalyl anthranilic acid and phenylhydrazine was heated to boiling. Water was evolved, but there was no evidence of any alcohol being given off. On cooling, a clear amber glass resulted. This was crystallized from alcohol, and the crystals obtained washed with dilute alcohol. Recrystallized from alcohol, long lemon-yellow needles were obtained, melting at 142° (corr.).

Calculated for $C_{17}H_{13}O_3N_3$: N, 13.58.

Found: N, 13.65.

The compound is insoluble in water, but dissolves readily in alcohol, benzene or acetone, giving bright yellow solutions. It is insoluble in cold solutions of caustic alkalies, but dissolves on heating. It does not dissolve in dilute mineral acids.

Sym. Di-(3-amino-4-quinazolone-2-carboxylic Acid) Hydrazide,

$H_2NN.CO.C_6H_4.N = C.CONHNHCO.C = N.C_6H_4.CO.NNH_2$.—One gram of hydrazine hydrate (in 50 per cent. aqueous solution) was diluted with 95 per cent. alcohol, 2 grams ethoxalyl anthranil gradually stirred in, and the mixture heated to boiling. The undissolved substance turned light yellow. It was filtered out and the yellow filtrate on standing deposited an additional amount of this same yellow amorphous material. It is slightly soluble in the ordinary neutral organic solvents, and melts at $157-158^{\circ}$ (corr.) with loss of ammonia, thereby becoming insoluble and infusible. Yield, 2.1 grams. For analysis, it was recrystallized from a large volume of absolute alcohol, and dried at 110° .

Calculated for $C_{18}H_{14}O_4N_8$: N, 27.60.

Found: N, 27.91.

Treated with acetic anhydride, a thick sirup resulted which defied all attempts to crystallize or purify it.

The same hydrazide was produced when methoxalyl anthranil was used instead of the ethoxalyl compound. Varying the proportions of anthranil and hydrazine did not appear to affect the result.

2-Carbazino-3-amino-4-quinazolone, $\text{H}_2\text{NN} \cdot \text{CO} \cdot \text{C}_6\text{H}_4 \cdot \text{N} = \text{C} \cdot \text{CONHNH}_2$, was obtained from its hydrochloride (see below) by dissolving the latter in water, adding potassium hydroxide solution to alkalinity and then acidifying with acetic acid. The precipitated free base was washed with water and crystallized from dilute alcohol acidified with acetic acid. Beautiful transparent plates were obtained, carrying water of crystallization. This water of crystallization was driven out at 100–110°, and the crystals then became opaque and melted at 202.5° (corr.).

Calculated for $\text{C}_9\text{H}_8\text{O}_2\text{N}_5$: N, 31.96.

Found: N, 31.70.

The substance is appreciably soluble in water and easily soluble in alcohol. When once dissolved, it does not separate again readily, and the loss on recrystallization is therefore considerable. A small amount tends to precipitate in an amorphous state.

Hydrochloride, $\text{C}_9\text{H}_8\text{O}_2\text{N}_5 \cdot 3\text{HCl}$.—One gram ethoxalyl anthranil was added to an aqueous solution of half a gram hydrazine hydrate diluted with 95 per cent. alcohol, and the mixture heated to boiling. The above *sym.* diquinazolone hydrazide (m. 157–158°) separated. Without removing the latter, concentrated hydrochloric acid was poured in and the mixture again boiled. The colorless hydrochloride which separated was washed with 95 per cent. alcohol. Yield, 0.8 gram. For analysis, it was recrystallized from a mixture of dilute hydrochloric acid and alcohol, washed with alcohol, and dried at 110°.

Calculated for $\text{C}_9\text{H}_8\text{O}_2\text{N}_5 \cdot 3\text{HCl}$: N, 21.43.

Found: N, 21.97.

It is freely soluble in water, but insoluble in most of the ordinary organic solvents, and melts at 190–191° (corr.). From strong hydrochloric acid, it crystallizes in colorless needles. As this monohydrazide is presumably formed by the hydrolysis of the dihydrazide which first separated, there should be produced simultaneously the 3-amino-4-quinazolone-2-carboxylic acid, but we have been unable to isolate any such compound from the mother liquors.

Acetyl Derivative, $\text{CH}_3\text{CONHN} \cdot \text{CO} \cdot \text{C}_6\text{H}_4 \cdot \text{N} = \text{C} \cdot \text{CONHNHCOCH}_3$.—0.8 gram 2-carbazino-3-amino-4-quinazolone hydrochloride and 0.6 gram fused sodium acetate were added to an excess of acetic anhydride and the solution boiled down to small bulk. More acetic anhydride was then added, the precipitated salt (NaCl) filtered out, and the filtrate boiled down twice more with acetic anhydride, finally carrying the evaporation

to dryness. The residual clear amber glass was dissolved in 95 per cent. alcohol, the solution concentrated and benzene added. The acetyl derivative separated in a fine yellowish powder, and was washed thoroughly with benzene and dried. Yield, 0.35 gram.

Calculated for $C_{13}H_{13}O_4N_5$: N, 23.10.

Found: N, 23.16.

The pure substance melts at 125° (corr.). It is easily soluble in water, alcohol or chloroform, but does not crystallize well from these solvents, evaporation of the solvent leaving finally a clear glass. On long boiling with benzene, small transparent plates were obtained, carrying benzene of crystallization.

Attempts to eliminate water from this compound and condense the side chains to a cycle proved unsuccessful.

NEW YORK, N. Y.

PREPARATION OF CHOLINE AND SOME OF ITS SALTS.

By R. R. RENSHAW.

Received November 22, 1909.

In connection with some work on the bactericidal properties of lecithins the author had occasion to prepare some of the salts of choline. In looking through the literature it was found that the choline used in recent investigations was prepared and isolated by methods which it was thought might be simplified.

Wurtz in 1867¹ prepared choline by heating trimethylamine with an excess of ethylene chlorohydrin, and later (1868) he and also Griess and Harrow² (1885) obtained it by the action of a concentrated solution of trimethylamine on ethylene oxide. The products were purified by means of the gold salt.

Later investigators have used different procedures or new methods. In 1889 Bode³ made choline by the prolonged action of dilute nitric acid on bromomethyltrimethylamine bromide which he had made from ethylene bromide by Hofmann's method. No yield was given. Gulewitsch in 1898⁴ in an extended article on choline described its preparation in quantity by a modification of Wurtz's method. Redistilled anhydrous ethylene chlorohydrin in large excess was heated with a 15-20 per cent. solution of trimethylamine in absolute alcohol for 24 hours in a water bath. The choline from the resulting slightly acid solution was separated partly as the mercuric, and partly as the platinic chloride double salts. Yield, 61 per cent. Krüger and Bergell in 1903⁵ got choline by the action of

¹ *Compt. rend.*, 65, 1015; *Ann. Spl.*, 6, 116.

² *Ber.*, 18, 707 (1885).

³ *Ann.*, 267, 268.

⁴ *Z. physiol. Chem.*, 24, 509.

⁵ *Ber.*, 36, 2901.