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RESEARCHES ON QUINAZOLINES (TWENTY-FIRST PAPER). ON CERTAIN QUINAZOLINE OXYGEN ETHERS OF THE TYPE —N : C(OR)— AND THE ISOMERIC —NR.CO— COMPOUNDS.

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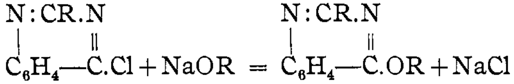
In a former article, by Bogert and Seil,[1] a résumé is given of the literature bearing upon the formation of —CO.NR— and —C(OR) : N— compounds when alpha-hydroxy derivatives of pyridines, quinolines, pyrimidines and quiazolines are treated with alkali and alkyl halide, together with the results of certain experiments along these lines with the nitroquinazolines. The preparation of the pure —NR.CO— compounds was described, and from the products obtained by treating nitro-4-quinazolones (nitro-4-hydroxyquinazolines) with alkali and alkyl halide there were isolated, in addition to the NR compounds, certain substances believed to be the corresponding oxygen ethers. At that time the separate synthesis of the pure oxygen ethers could not be undertaken because of the press of other work.

Besides the nitro ethers mentioned by Bogert and Seil,[2] the only other quinazoline oxygen ethers we have found in the literature (all prepared from the corresponding chlorquinazoline) are the 2,4-dimethoxyquinazoline of Abt,[3] the 2-methoxy-3-phenyl-4-quinazolone of McCoy,[4] the Bz-trichlor-2-methyl-4-ethoxyquinazoline of Dehoff,[5] the 5,6,7-trimethoxybenzoyleneurea of Pollak and Goldstein,[6] and the 2-ethoxy-Δ_2-4-quinazolone ("aethoxylcyanaminobenzoyl") of Griess.[7]

In resuming the study of quinazoline oxygen ethers, it has seemed best to begin with the simplest quinazolines instead of the nitro derivatives, and the pure oxygen ethers have been prepared by the action of alcoholates upon the chlorquinazolines:



$$\begin{array}{l}N{:}CR.N \\ |\qquad\ \ \| \\ C_6H_4—C.Cl\end{array} + NaOR = \begin{array}{l}N{:}CR.N \\ |\qquad\ \ \| \\ C_6H_4—C.OR\end{array} + NaCl$$

The isomeric —NR.CO— compounds were prepared by direct alkylation of the 4-quinazolones (4-hydroxyquinazolines) with alkali and alkyl halide, and by various special methods described in previous papers.

With the pure —C(OR) : N— and —CO.NR— isomers in hand, and

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[1] THIS JOURNAL, 29, 517 (1907).

[2] *Loc. cit.*

[3] *J. prakt. Chem.* [2], 39, 149 (1889).

[4] *Am. Chem. J.*, 21, 160 (1889).

[5] *J. prakt. Chem.* [2], 42, 354 (1890).

[6] *Ann.*, 351, 161 (1906).

[7] *Ber.*, 2, 415 (1869).
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their properties established, it was possible to study more closely the alkylation of 4-quinazolones (4-hydroxyquinazolines) by alkali and alkyl halide, and to determine the approximate amounts of oxygen and nitrogen ethers formed. We relied chiefly upon steam distillation to separate the OR and NR isomers, for the former are readily volatile with steam, while the latter (except in a few cases) are not carried over to any appreciable extent by it. With methyl, ethyl, *n*-propyl or *n*-butyl iodide, the product in every case was almost exclusively the NR compound, with only traces of oxygen ether.

The NR compounds are colorless, odorless solids, quite soluble in water, generally very difficultly volatile with steam, of higher melting point than the OR isomers, and not hydrolyzed by strong hydrochloric acid.

The OR compounds are oily liquids or low-melting solids, generally of pleasant odor, readily volatile with steam, are less soluble in water but more soluble in hydrochloric acid than the NR isomers, and are readily hydrolyzed by the mineral acids to the corresponding hydroxyquinazolines (4-quinazolones). Some of the simpler ones can be distilled undecomposed at ordinary pressure.

Of the quinazolines described in the following pages, those we believe to be new are the 3-*n*-propyl-4-hydroxyquinazoline, 3-*n*-butyl-4-hydroxyquinazoline, 1,3-diethyl-2,4-dihydroxyquinazoline, 1-(or 3-)*n*-propyl-2,4-dihydroxyquinazoline, Bz-trichlor-2-ethyl-4-chlorquinazoline, 4-methoxyquinazoline, 4-ethoxyquinazoline, 4-*n*-propyloxyquinazoline, 4-*n*-butyloxyquinazoline, Bz-trichlor-2-methyl-4-methoxyquinazoline, 2,4-diethoxyquinazoline, 2,4-di-*n*-propyloxyquinazoline, 2-methoxyquinazoline and 2-ethoxyquinazoline.

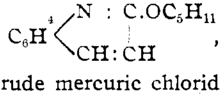
Experimental.

I. Quinoline Group.

Before taking up the quinazoline ethers, we carried out a few preliminary experiments in the quinoline group.

α-Chlorquinoline and sodium ethylate yielded α-ethoxyquinoline, whereas the action of ethyl bromide or iodide upon the sodium salt of carbostyril (α-hydroxyquinoline $\rightleftarrows$ α-quinolone) gave about equal amounts of oxygen and nitrogen ethers. This agrees with the experience of Friedländer and Ostermaier.[1] The oxygen ether gives a *mercuric chloride compound* melting at 136°–8°.

α-Chlorquinoline and sodium isoamylate yielded about 80 per cent. of the oxygen ether and about 20 per cent. of the NR compound. Isoamyl iodide and sodium carbostyril also gave both oxygen and nitrogen derivatives, while with silver carbostyril very little of the NR compound was formed.

2-Isoamyloxyquinoline,  $C_6H_4\left\langle\begin{array}{l}N : C.OC_5H_{11}\\ CH:CH\end{array}\right.$, is readily hydrolyzed by hydrochloric acid to carbostyril. Its crude mercuric chloride compound melts at 120–30°, and is less soluble in water than that of the isomeric NR compound.

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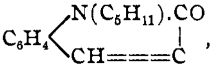
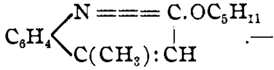
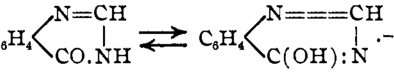
[1] *Ber.*, **14**, 1917 (1881); **15**, 335 (1882).

1-Isoamyl-2-quinolone,  $C_6H_4\left\langle\begin{array}{l}N(C_5H_{11}).CO\\ CH{=\!=}C\end{array}\right.$, forms a *mercuric chloride compound* which crystallizes in long needles, m. p. 89–90° (uncorr.), quite soluble in dilute hydrochloric acid and unhydrolyzed thereby.

2-Isoamyloxy Lepidine,  $C_6H_4\left\langle\begin{array}{l}N{=\!=\!=}C.OC_5H_{11}\\ C(CH_3):CH\end{array}\right.$.—By the interaction of the sodium salt of α-hydroxylepidine and isoamyliodide, a colorless compound was obtained, m. p. 120–40°, boiling above 360°, and rapidly hydrolyzed to α-hydroxylepidine again when boiled with hydrochloric acid. It is probable that it was the impure oxygen ether.

II. Quinazoline Group.

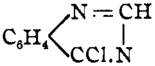
1. Derivatives of 4-Quinazolone (4-Hydroxyquinazoline).

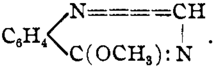
4-Quinazolone (4-Hydroxyquinazoline),  $C_6H_4\left\langle\begin{array}{l}N{=}CH\\ CO.NH\end{array}\right. \rightleftarrows C_6H_4\left\langle\begin{array}{l}N{=\!=\!=}CH\\ C(OH):N\end{array}\right.$.—This is easily obtained by heating anthranilic acid with formamide.[1] It crystallizes from water in fine, colorless needles, m. p. 215.5 – 216.5° (corr.).[2]

In certain of the reactions where 4-chlorquinazoline was hydrolyzed there was obtained a compound, crystallizing from water in yellowish warts and nodules, melting rather raggedly at 208–12° (uncorr.), and much more soluble in water than the ordinary 4-quinazolone to which it could be largely converted by heating in a sealed tube at 250–65°. On analysis, it gave the following figures:

Found: C, 65.80; H, 4.22: Calculated for $C_8H_6ON_2$: C, 65.75; H, 4.11.

Whether this is the labile 4-hydroxyquinazoline or only impure 4-quinazolone is a question not yet decided. The solubility in water supports the former rather than the latter assumption.

4-Chlorquinazoline,  $C_6H_4\left\langle\begin{array}{l}N{=}CH\\ CCl.N\end{array}\right.$, was prepared by the method of Gabriel[3] from 4-quinazolone, phosphorus penta- and oxychlorides. It was found that by using twice as much oxychloride as recommended by Gabriel, and continuing the heating for an hour after all was dissolved, better results were secured. The phosphorus chlorides used must be pure, or the chlorquinazoline will not be colorless. The pure chlorquinazoline melts at 96°.

4-Methoxyquinazoline,  $C_6H_4\left\langle\begin{array}{l}N{=\!=\!=}CH\\ C(OCH_3):N\end{array}\right.$.—4-Chlorquinazoline was treated with sodium methylate in absolute methyl alcohol solution. After the completion of the reaction in the cold, the mixture was warmed for a short time on the water bath, filtered from salt, the filtrate evaporated to dryness and the residue distilled with steam. A yellow oil separated in the distillate. This was collected with ether, the ethereal solution dried with calcium chloride and the ether distilled off. There remained a thin oil which soon solidified in rosettes of colorless crystals, m. p. 35.4°. From 30 g. of 4-quinazolone, 14 g. of this oxygen ether were obtained, and from the residue not volatilized by steam 12 g. of the quinazolone (from hydrolysis of the chloride) were recovered. On analysis, the ether gave the following figures:

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

[2] Hand, This Journal, **24**, 1048 (1902).

[3] *Ber.*, **29**, 1314 (1896).

Found: C, 67.32; H, 5.03; N, 17.65. Calculated for $C_9H_8ON_2$: C, 67.50; H, 5.0; N; 17.50.

The pure ether possesses a very pleasant odor. It is readily hydrolyzed by hydrochloric acid to the 4-quinazolone again. A small amount of the ether was boiled with acetic anhydride, the solution cooled, a little water added and the solution gently heated. On cooling, 4-quinazolone crystallized out.

4-Ethoxyquinazoline, $C_8H_5N_2(OC_2H_5)$, prepared from the chlorquinazoline and sodium ethylate, is a colorless oil, of pleasant ethereal odor, which solidifies in a freezing mixture to crystals which melt at 42–4°.

Found: N, 16.24. Calculated for $C_{10}H_{10}ON_2$: N, 16.06.

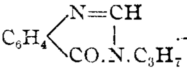
From 10 g. 4-quinazolone, 2.5 g. of the ethoxy compound were obtained and 7 g. 4-quinazolone recovered.

4-n-Propyloxyquinazoline, $C_8H_5N_2(OC_3H_7)$, from 4-chlorquinazoline and a solution of sodium propylate in absolute ethyl alcohol. Colorless oil, b. p. 257–60°, giving 4-quinazolone when hydrolyzed by hydrochloric acid. Eight grams of the 4-quinazolone gave 5.2 g. of the crude ether.

4-n-Butyloxyquinazoline, $C_8H_5N_2(OC_4H_9)$.—Oil, b. p. 263–5°, hydrolyzed to 4-quinazolone by hydrochloric acid. Yield of crude ether, about the same as for the above propyl ether.

4-Chlorquinazoline was also boiled in dry benzene solution with finely powdered potassium salts of 4-hydroxyquinazoline and of 2-methyl-4-hydroxyquinazoline, but in neither case was there any evidence of the formation of an oxygen ether.

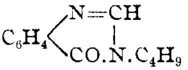
Of the NR compounds isomeric with the above oxygen ethers, the 3-methyl-4-quinazolone, m. p. 71°, has been described by Knape.[1]



3-n-Propyl-4-quinazolone, $C_6H_4\left<\begin{array}{l}N{=}CH\\ \quad\ |\\ CO.N.C_3H_7\end{array}\right.$.—4-Quinazolone was dissolved in alcohol containing the calculated amount of sodium, an equivalent quantity of *n*-propyl iodide was added and the whole heated on the water bath until neutral to litmus. The sodium iodide was then filtered out, the filtrate evaporated to dryness, and the residue distilled with steam. From the portion not volatile with steam crystals were obtained on concentrating, m. p. 82°.

Found: N. 14.98. Calculated for $C_{11}H_{12}ON_2$: N, 14.89.

A small amount of the same material was carried over by the steam, but no oxygen ether was found. From 10 g. 4-quinazolone, 6.8 g. of this nitrogen derivative resulted. It crystallizes from water in colorless needles, m. p. 82–3°. When heated in a sealed tube at 110–20° for an hour and a half with strong hydrochloric acid, it was unchanged.



3-n-Butyl-4-quinazolone, $C_6H_4\left<\begin{array}{l}N{=}CH\\ \quad\ |\\ CO.N.C_4H_9\end{array}\right.$, was prepared in a similar manner, using *n*-butyl instead of *n*-propyl iodide. From 10 g. 4-quinazolone, about an equal weight of this butyl derivative was obtained. A small amount was found in the steam distillate, together with what appeared to be traces of an oxygen ether.

Found: N, 13.80. Calculated for $C_{12}H_{14}ON_2$: N, 13.86.

It crystallizes from alcohol in colorless, silky needles, m. p. 73°, unchanged when heated with strong hydrochloric acid for an hour and a half at 110–20°.

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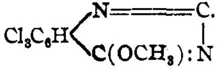
[1] *J. prakt Chem.* [2], **43**, 216 (1891).

2. *Derivatives of 2-Methyl- and 2-Ethyl-4-quinazolone (2-Methyl- and 2-Ethyl-4-hydroxyquinazoline).*

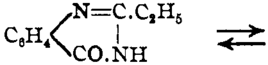
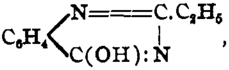
Bz-Trichlor-2-methyl-4-methoxyquinazoline,  $\mathrm{Cl_3C_6H}\left\langle\begin{array}{l}\mathrm{N{=\!=\!=}C.CH_3}\\ \qquad\quad |\\ \mathrm{C(OCH_3):N}\end{array}\right.$ —Dehoff[1] has shown that the action of phosphorus penta- and oxychlorides upon 2-methyl-4-quinazolone gives a Bz-trichlor-2-methyl-4-chlorquinazoline, m. p. 125°. Our experiments led to a similar result. With pure phosphorus chlorides and twice as much oxychloride as prescribed by Dehoff, better yields and practically colorless products were obtained. We have tried in vain to get a halogen in position 4 without at the same time introducing other halogens into the molecule. Evidently the quinazoline nucleus differs in this respect from the quinoline nucleus, for Ornstein[2] succeeded in changing 3-methyl carbostyril to 3-methyl-4-chlorquinoline without affecting the benzene nucleus.

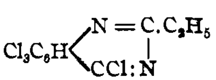
The Bz-trichlor-2-methyl-4-methoxyquinazoline was prepared from the above tetrachlor compound and sodium methylate in absolute methyl alcohol solution at 110°. It was separated from the products of the reaction by distillation with steam, and forms minute, colorless, silky needles, m. p. 87–8°. Yield, 2.4 g. pure ether from 3.2 g. tetrachlor derivative.

Found: Cl, 38.24. Calculated for $C_{10}H_7ON_2Cl_3$: Cl, 38.37.

Heated gently with concentrated hydrochloric acid for 15 minutes, the ether is hydrolyzed to the Bz-trichlor-2-methyl-4-quinazolone (m. p. 206–7°) of Dehoff.[3]

Bz-Trichlor-2-methyl-4-ethoxyquinazoline, $Cl_3C_9H_4N_2(OC_2H_5)$, was obtained by Dehoff[4] by the action of alcoholic sodium hydroxide solution upon the tetrachlor derivative. It melts at 75–6°. We have confirmed his work. The ether is readily hydrolyzed by strong hydrochloric acid.

2-Ethyl-4-quinazolone (2-Ethyl-4-hydroxyquinazoline),  $\mathrm{C_6H_4}\left\langle\begin{array}{l}\mathrm{N{=}C.C_2H_5}\\ \qquad |\\ \mathrm{CO.NH}\end{array}\right. \rightleftarrows$  $\mathrm{C_6H_4}\left\langle\begin{array}{l}\mathrm{N{=\!=\!=}C.C_2H_5}\\ \qquad\quad |\\ \mathrm{C(OH):N}\end{array}\right.$, has been described before by Bischler and Lang.[5] It melts at 234° (corr.).[6] We have obtained the same substance by boiling anthranilic acid with excess of propionic anhydride, distilling off the excess of the latter, boiling the residue with acetic anhydride, distilling off the excess of this anhydride, and then boiling the propionylanthranil thus obtained with strong ammonium hydroxide solution, the crude quinazoline being purified by solution in sodium hydroxide and reprecipitation with carbon dioxide.

Bz-Trichlor-2-ethyl-4-chlorquinazoline,  $\mathrm{Cl_3C_6H}\left\langle\begin{array}{l}\mathrm{N{=}C.C_2H_5}\\ \qquad |\\ \mathrm{CCl:N}\end{array}\right.$ —2-Ethyl-4-quinazolone, phosphorus penta- and oxychlorides, were heated together in a sealed tube for three and a half hours at 165°. The excess of phosphorus halides was removed by distillation, ice water was added to the residue, then dilute alkali to distinct alkalinity, and the mixture was warmed to dissolve unchlorinated quinazolone. When cold, the undissolved material was filtered out, dissolved in alcohol, the solution bone-blacked, concentrated, and water added. The tetrachlorquinazoline separated in crystals and was purified by recrystallization from dilute alcohol.

Found: Cl, 47.94. Calculated for $C_{10}H_6N_2Cl_4$: Cl, 47.97.

The pure substance forms minute, practically colorless needles, m. p. 80°.

Having failed in this case also to get a chlorine in position 4 without simultaneous

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[1] *J. prakt. Chem.* [2], **42**, 352 (1890).

[2] *Ber.*, **40**, 1095 (1907).

[3] *Loc. cit.*

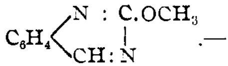
[4] *Ibid.*

[5] *Ber.*, **28**, 284 (1895).

[6] Gotthelf, This Journal, **23**, 619 (1901).

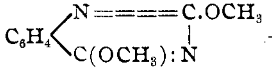
chlorination of the benzene nucleus, experiments were undertaken with 2,3-dimethyl-4-quinazolone, in the hope that under the influence of phosphorus halides it might yield the desired 2-methyl-4-chlorquinazoline with elimination of methyl chloride, as accomplished by Fischer[1] with N-alkyl α-pyridones and related compounds, and by Abt[2] in the case of N-methyl benzoyleneureas. The 2,3-dimethyl-4-quinazolone was prepared by alkylation of 2-methyl-4-quinazolone with caustic alkali and methyl iodide, and also by the action of methylamine upon acetanthranil. It melts at 110–11° (corr.)[3] and is slightly volatile with steam. When it was heated with phosphorus penta- and oxychlorides, the 3-methyl group was split off, chlorine was introduced in position 4, but again three more chlorine atoms entered the benzene nucleus, and the same Bz-trichlor-2-methyl-4-chlorquinazoline was obtained as from 2-methyl-4-quinazolone.

3. Derivatives of 2-Quinazolone (2-Hydroxyquinazoline).

2-Methoxyquinazoline,  $C_6H_4\langle\begin{array}{l}N : C.OCH_3\\ \quad\;\;|\\ CH : N\end{array}$.—2-Quinazolone (2-hydroxyquinazoline) was prepared by fusing *o*-aminobenzaldehyde with urea.[4] It was converted to 2-chlorquinazoline by heating it with phosphorus penta- and oxychlorides.[5] When this chlorquinazoline was heated with sodium methylate in absolute methyl alcohol solution, and the product distilled with steam, an oil passed over, which soon solidified in beautiful needles, of pleasant odor, and melting at 55–6°. Heated for an hour and a half with concentrated hydrochloric acid in a sealed tube at 110–20°, it was hydrolyzed to 2-quinazolone again. From 1 g. of the pure chlorquinazoline, 0.8 g. of the methoxy compound was obtained.

2-Ethoxyquinazoline,  $C_6H_4\langle\begin{array}{l}N = C.OC_2H_5\\ \quad\;\;|\\ CH : N\end{array}$, from 2-chlorquinazoline and sodium ethylate, was isolated as a yellow oil, solidifying in the ice-box to cubical crystals. It is volatile with steam, and hydrolyzed with hydrochloric acid to the 2-quinazolone again.

4. Derivatives of Benzoylene Urea (2,4-Dihydroxyquinazoline).

2,4-Dimethoxyquinazoline,  $C_6H_4\langle\begin{array}{l}N = C.OCH_3\\ \qquad\quad\;|\\ C(OCH_3) : N\end{array}$—The preparation of this ether from 2,4-dichlorquinazoline has been described by Abt.[6] We have found that it is easily volatile with steam and most conveniently purified in this way. Abt gives the melting-point as 66°. Our product melted at 67°.

2,4-Diethoxyquinazoline, $C_8H_4N_2(OC_2H_5)_2$, from the dichlorquinazoline and sodium ethylate, separated from the steam distillate in oily drops solidifying on cooling. Six g. benzoylene urea yielded 2 g. of the pure ether. On recrystallization, it formed colorless, silky needles of pleasant odor, melting at 50–1°.

Found: N, 12.81. Calculated for $C_{12}H_{14}O_2N_2$: N, 12.84.

Gently boiled for five minutes with strong hydrochloric acid, the ether is hydrolyzed to benzoylene urea.

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[1] *Ber.*, **32**, 1297 (1899).

[2] *J. prakt. Chem.* [2], **39**, 149 (1889).

[3] Bogert and Gotthelf, THIS JOURNAL, **22**, 532 (1900). Bogert and Seil, *Ibid.*, **29**, 531 (1907).

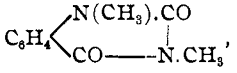
[4] Gabriel and Posner, *Ber.*, **28**, 1037 (1895). Gabriel and Stelzner, *Ibid.*, **29**, 1313 (1896).

[5] Gabriel and Stelzner, *Loc. cit.*

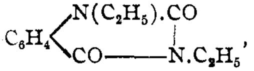
[6] *Loc. cit.*

2,4-Di-n-propyloxyquinazoline, $C_8H_4N_2(OC_3H_7)_2$, crystallizes from water in long, colorless needles, m. p. 40–1°. It has a pleasant odor, and is rapidly hydrolyzed to benzoylene urea by heating with strong hydrochloric acid.

Found: N, 11.5. Calculated for $C_{14}H_{18}O_2N_2$: N, 11.38.



1,3-Dimethylbenzoylene urea, $C_6H_4\left<\begin{matrix}N(CH_3).CO\\ \mid\\ CO\text{———}N.CH_3\end{matrix}\right.$, from benzoylene urea, alcoholic sodium hydroxide and methyl iodide was first obtained by Abt.[1] It is not hydrolyzed by concentrated hydrochloric acid. Abt gives the melting-point as 151°. Our product crystallized in colorless needles and melted at 163–5°.



1,3-Diethylbenzoylene urea, $C_6H_4\left<\begin{matrix}N(C_2H_5).CO\\ \mid\\ CO\text{———}N.C_2H_5\end{matrix}\right.$, from benzoylene urea, alcoholic sodium hydroxide and ethyl iodide, was purified by boiling it for a few minutes with strong hydrochloric acid (to hydrolyze any oxygen ether), neutralizing, collecting the separated crystals, washing them with dilute sodium hydroxide solution (to remove any benzoylene urea), and recrystallizing from alcohol. Minute colorless needles resulted, *m.* p. *105°–106°*.

Found: N, 12.98. Calculated for $C_{12}H_{14}O_2N_2$: N, 12.84.

1- (or *3-*)*n-Propylbenzoylene Urea*, $C_8H_5O_2N_2.C_3H_7$.—When benzoyleneurea was treated with alcoholic sodium hydroxide solution, *n*-propyl iodide added, and the mixture boiled under a return condenser, the monopropyl derivative resulted in every experiment, no matter how large the excess of propyl iodide present. On removal of the excess of alcohol and iodide, this propyl derivative tends to crystallize from the solution in needles. Distillation with steam, disclosed no oxygen ether. The crude propyl derivative was purified by dissolving it in sodium hydroxide solution and reprecipitating with carbon dioxide.

Found: N, 13.79. Calculated for $C_{11}H_{12}O_2N_2$: N, 13.73.

The pure substance forms minute, colorless, fluffy crystals, m. p. 171°. From 2 g. benzoylene urea 1.8 g. of the monopropyl derivative were obtained.

One gram of this monopropyl derivative was then dissolved in absolute propyl alcohol containing the calculated amount of sodium, *n*-propyl iodide was added, and the mixture boiled four hours under a return condenser, but no dipropyl derivative was found. 0.83 g. of the monopropyl compound was recovered unaltered (m. p. 171°).

This failure to get the dipropyl derivative is rather surprising, as the dimethyl and diethyl derivatives were prepared quite easily, and Abt[1] has shown that the former of these may be prepared from benzoylene urea, its 1- or 3-monomethyl derivatives, with alkali and methyl iodide.

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NEW BOOKS.

Quantitative Analyse durch Elektrolyse. Von ALEXANDER CLASSEN. Fünfte Auflage in durchaus neuer Bearbeitung. Unter Mitwirkung von H. CLOEREN. Berlin: Julius Springer. 1908. pp. xi + 336. Leinwand, Preis, 10 Mk.

This well-known work has undergone a thorough revision and enlargement. The theoretical introduction of 106 pages has been markedly

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[1] *Loc. cit.*