Anal. Calcd. for $C_{19}H_{31}ClN_2OS$: N, 7.51; Cl, 9.50. Found: N, 7.38; Cl, 9.64.

2-Butyloxyquinoline-4-carboxylic Acid Derivatives.—These compounds were readily prepared from 2-butyloxyquinoline-4-carbonyl chloride[8] and the amine or alcohol in dry benzene.

2-(2-Diethylaminoethylmercapto)-ethyl 2-butyloxyquinoline-4-carboxylate hydrochloride, VII, white leaflets from ethyl acetate, m. p. 125.8–127.0°.

Anal. Calcd. for $C_{22}H_{33}ClN_2O_3S$: N, 6.35; Cl, 8.04. Found: N, 6.20; Cl, 8.03.

2-(3-(Piperidyl-1)-propylmercapto)-ethyl 2-butyloxyquinoline-4-carboxylate hydrochloride, VII, waxy white needles from isopropyl alcohol, m. p. 118.4–120.4°.

Anal. Calcd. for $C_{24}H_{35}ClN_2O_3S$: N, 6.01; Cl, 7.61. Found: N, 6.01; Cl, 7.56.

N-(2-(2-Diethylaminoethylmercapto)-ethyl 2-butyloxyquinoline-4-carboxamide, VIII, long slender white needles from Skellysolve B, m. p. 63.5–64.5°.

Anal. Calcd. for $C_{22}H_{33}N_3O_2S$: N, 10.41; S, 7.94. Found: N, 10.31; S, 8.08.

The **citrate** formed tiny white prisms from absolute alcohol–ethyl acetate, m. p. 87.5–90.5° (dec.).

Anal. Calcd. for $C_{28}H_{41}N_3O_9S$: N, 7.05; S, 5.38. Found: N, 6.83; S, 5.22.

(8) Gardner and Hammel, THIS JOURNAL, **58**, 1360 (1936), report 37–51% yields of 2-alkoxycinchonyl chlorides, with accompanying by-products of the 2-alkoxycinchoninic acid hydrochloride. Modification of their procedure, by inclusion of a one hour reflux period of the acid–thionyl chloride–benzene mixture, increased the yields of acid chlorides to 95–97%, with no formation of 2-alkoxycinchoninic acid hydrochloride.

Summary

There has been described the preparation of a series of esters and amides derived from sulfur-containing amines.

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[CONTRIBUTION FROM THE WELLCOME RESEARCH LABORATORIES]

A Family of Long-Acting Depressors[1,2]

BY RICHARD BALTZLY, JOHANNES S. BUCK,[3] EDWIN J. DE BEER AND FREDERICK J. WEBB[4]

Some years ago Ide and Buck[5] prepared a number of N-methyltetrahydro*iso*quinolines by the cyclization of the appropriate N-methylphenethylamines with formalin and hydrochloric acid. A pharmacological study was performed by Fassett and Hjort,[6] who found all but one member of this group to be weak pressors or depressors with transient action, while 6-ethoxy-N-methyltetrahydro*iso*quinoline was an extremely potent depressor with a long period of action. While correlation of physiological action with chemical structure is on essentially an empirical basis at present, such correlation within classes of related compounds usually is fairly good. It was therefore felt that the physiological results cast grave doubt on the purity and identity of this sample of 6-ethoxy-N-methyltetrahydro*iso*quinoline.

An authentic specimen of 6-ethoxy-2-methyltetrahydro*iso*quinoline hydrochloride was prepared by an alternative route.[7] The new sample had none of the remarkable physiological activity of the older specimen. Further fractionation of material prepared according to the formalin procedure afforded a sample more nearly resembling the authentic (new) specimen. De-ethylation of the original material also yielded a sample of 6-hydroxy-N-methyl tetrahydro*iso*quinoline hydrochloride identical with an authentic specimen of this substance. It was thus evident that the Ide and Buck material was preponderantly 6-ethoxy-N-methyl tetrahydro*iso*quinoline hydrochloride contaminated by a small amount of a highly potent depressor substance whose physical properties and composition were not markedly different from those of the major component.

A number of formalin cyclizations of 3-ethoxyphenethylmethylamine followed by attempts to fractionate the reaction products gave rather irregular results. Usually such preparations, like the original one, produced strong, lasting depression of blood pressure when administered intravenously to anesthetized dogs in doses of 1 mg./kg. body weight. One specimen, a chloroform-insoluble hydrochloride obtained by successive precipitations from chloroform solution by ethyl acetate, was about four times as active.

Serious progress in elucidating the nature of the unknown depressor was not made until a change in the amine under study was effected. Cyclization of a phenethylamine to a tetrahydroisoquinoline by the formalin–hydrochloric acid procedure is effective only when there is an activating group para to the cyclization-point. Ortho and para methoxyphenethylmethylamines are not cyclized by this method. When these amines were treated by the usual procedure, however, the products were by far the most active depressors so far obtained. It was now possible to secure a fall in the blood-pressure of 100 mm. of mercury lasting for an hour or more with doses of the

(1) Presented before the Division of Medicinal Chemistry of the American Chemical Society, Washington Meeting, August, 1948.

(2) The work here reported is part of a project carried out in collaboration with a pharmacological group in these laboratories headed successively by A. M. Hjort and E. J. de Beer. The detailed pharmacological study will be published separately.

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(4) Present address: Firestone Rubber Co., Akron, Ohio.

(5) Ide and Buck, THIS JOURNAL, **59**, 726 (1937).

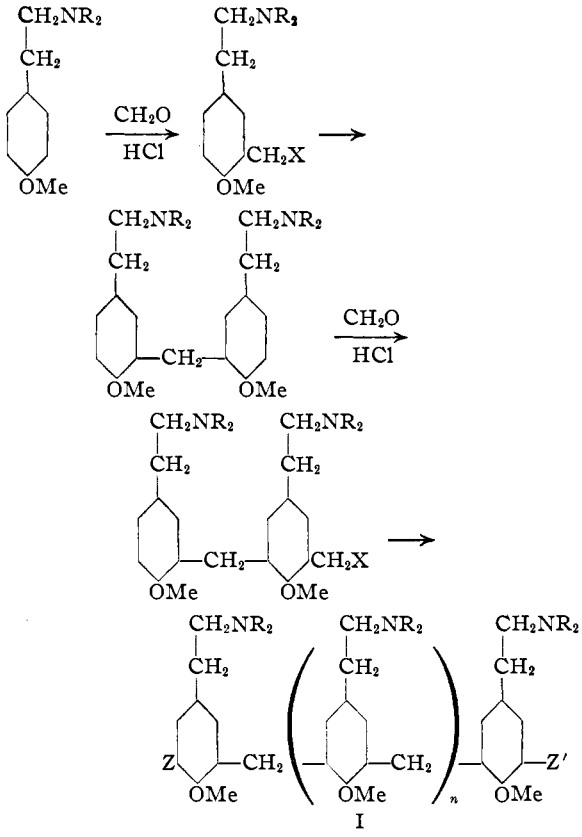
(6) Fassett and Hjort, *J. Pharmacol. Exptl. Therap.*, **63**, 253 (1938).

(7) Hjort, de Beer, Buck and Randall, *ibid.*, **76**, 64, 252 (1942).

order of 0.1 mg./kg. body weight.[8] This indicated that the depressor type was not a tetrahydro*iso*quinoline.

Two main possibilities remained: cross-linking between nitrogen atoms by formaldehyde, and formation of methylene links between aromatic nuclei. The first possibility was eliminated when it was found that hordenine methyl ether and its metho-chloride both yielded preparations of maximum potency. It was therefore probable that the depressor type was formed by the sort of reaction shown in Chart I.

CHART I



wherein X might be Cl or OH and Z and Z′ could be H or CH_2OH or CH_2Cl. The conditions of the reaction are essentially those of a mild chloromethylation and formation of a condensation polymer as indicated in Chart I is not at all surprising.

The optimum conditions for the condensation were found to be when the formalin was approximately equimolecular with the amine, when the hydrochloric acid was about 6*N*, and when heating was continued for about four hours on the steam-bath. The products so obtained were light colored glasses when dried thoroughly. They exhibited the usual amine-hydrochloride solubilities and could be ground to powders under ether. Precipitation from absolute ethanolic solution by ethyl acetate and ether under controlled conditions gave filterable powders that appeared granular to gross inspection and which were not clearly crystalline or non-crystalline under the microscope. Such powders, however, were incapable of seeding out more material from solutions in alcohol brought to the point of turbidity by addition of ether, and undoubtedly were not truly crystalline. Further fractionation revealed a variety of components.

When a "Standard Preparation" from *p*-methoxyphenethylmethylamine was dissolved in water, perchloric acid and ammonium perchlorate were added and the solution was refrigerated, a solid perchlorate separated. This material, doubtfully crystalline to start with, became definitely crystalline on recrystallization from water and could be transformed to a crystalline hydrochloride. This hydrochloride had a composition consistent with that of a "dimer" (I, $n = 0$, $Z = Z' = H$). The base corresponding gave a molecular weight in agreement with this formulation. The "dimer" base distils at about 120° at 0.5 μ pressure. On the basis of the amounts of perchlorate isolable about a third of the "standard preparation" is "dimer." This material is nearly inactive physiologically giving a weak depression of short duration.

After removal of the "dimer" as its perchlorate, partial basifications followed by extraction with ether and later ether–ethyl acetate mixtures produced a series of base fractions. All were oils and were increasingly viscous and diminishingly soluble in ether as the basification proceeded. The more ether-soluble fractions gave "molecular weights" about right for a "trimer," the less ether-soluble fractions were probably more highly polymerized; their Rast values were about right for the "tetramer." In high vacuum the "trimer fraction" was partially distillable, perhaps with some decomposition. The distillate (boiling around 200° at 0.3 μ pressure) was highly active; it left a residue on redistillation. It is not clear whether this residue consists of higher molecular material initially present or produced by further condensation during the distillation. Some of these residues proved to be reactive on testing, but some were too insoluble for investigation.

The above experiments indicated that the physiological activity was associated with trimeric and tetrameric condensation products. Attempts at crystallizing such fractions were not successful and further fractionation appeared required. The most fruitful results were obtained by the use of Craig's method of counter-current distribution[9] on the product from the condensation

(8) We have not been able to improve on this potency. Some fractions obtained subsequently were undoubtedly more homogeneous and probably more potent. Unfortunately, the physiological assay will not differentiate clearly between preparations whose activity is in the ratio of 3:2. As will be seen this is about the greatest ratio of potencies that could be expected between a pure component and a total reaction mixture of this type.

(9) Craig, *J. Biol. Chem.*, **155**, 519 (1944); Craig, Golumbic, Mighton and Titus, *ibid.*, **161**, 321 (1945); Williamson and Craig, *ibid.*, **168**, 687 (1947).

of *p*-methoxyphenethyldimethylamine with formaldehyde. This polymer is perhaps less active than that from the corresponding secondary amine but appeared more suitable for investigation. An eight-plate fractionation between 50% benzene–hexane and 75% methanol–water yielded two fractions. The more hydrophobic of these, which contained about two-thirds of the active material, was presumably free of hydroxymethyl groups since it gave no gas in a Zerewitinoff machine. The more hydrophilic fraction could be separated further by a repeated distribution using 55% methanol as the lower phase. The separation was only approximate but indicates the presence of low polymers having one hydroxymethyl group and of another type having two. All of these fractions were highly potent, and equally potent within the limits of accuracy of the physiological assay. So far, however, no active fraction has yielded crystalline salts. It is to be expected that molecules of this type would not crystallize readily and failure to crystallize them is not in itself proof of non-homogeneity but their physical properties taken together with the analytical data seem to indicate that separation had been made only between types, not between species.[10]

Condensations between formaldehyde and amines with shorter side chains resulted in qualitatively similar products. Those from *p*-methoxybenzylmethylamine and *p*-methoxybenzyldimethylamine were somewhat less potent and more toxic. The proportion of dimer was higher and the general state of polymerization appeared to be lower. This would be anticipated on the general principles of aromatic substitution since a cationic group only one stage removed from a benzene ring is well known to interfere appreciably with substitution.[11]

Condensation products of the same general type were also obtained from *o*-anisidine. The polymeric aromatic bases had no depressor action but the derived methiodides were fairly potent. This would seem to show that activity is dependent on the presence of groups that would be cationic under physiological conditions and suggests that a molecule of the approximate dimensions of a "trimer" and possessing two or three properly spaced cationic groups should exhibit strong depressor action. Experiments in this direction have been in progress and will be reported separately.

Experimental

Physiological Testing.—Since evaluation of the chemical work was at all times dependent on physiological assay a few remarks about the methods used and their interpretation are desirable even though the pharmacology of these depressors is to be reported elsewhere. In Table I are shown typical results selected from the very large number available. The preparations tested are identified by code numbers referring to the section "Chemical Manipulations."

TABLE I

POTENCIES OF TYPICAL DEPRESSOR PREPARATIONS

Specimen (Code number)	Potency	Specimen (Code number)	Potency
IV-132	++	GC-60	+
Q-190C	+++	GC-55	+
M-25	++	GC-125 I	++++
M-27	++	GC-125 II	++++
GD-6	+	GC-69A	++++
GC-142-1	+	GC-69B	++++
GC-142-2	++	GC-131-2	++++
GC-142-3	+++	GC-131-3	++++
GC-142-4	++++	GC-132-8	++++
GC-142-5	+++	M-124 res.	++++
M-96	+++	GC-114	++++
GC-81 II	++++	GC-114 II	++++
M-118	++++	GC-104 II	+
M-114	++++	GC-110	+++

Assay was carried out by injecting aqueous solutions intravenously into anesthetized dogs in the usual set-up for observation of blood pressure. In the earlier period of this work chief attention was given to discovering the minimum effective dose for the preparation used. This led to irregularities since relatively few animals could be tested and individual variations in sensitivity were considerable. It was later decided to administer standard doses (usually 0.4 mg./kg.) of the more potent preparations and to evaluate the response by the magnitude and duration of depression. Such methods might distinguish one preparation from another of twice the potency but smaller differences are not detected with certainty.

Potency is indicated in Table I by the notations ++++, +++, etc. The maximum potency is ascribed to preparations that in doses of 0.02–0.05 mg./kg. body weight lowered the blood pressure by about 30 mm. (of mercury) and whose effect lasted at least fifteen minutes *or*, by the later method of testing, gave depressions of 90 mm. or more lasting at least one hour in doses of 0.1–0.4 mg./kg. Preparations giving only short depressions (five to ten minutes) of small amplitude in doses of 1 mg./kg. are indicated with +. The notations +++ and ++ show samples of intermediate activity.

Chemical Preparations.—Sample IV-132 was the original specimen of "6-ethoxy-2-methyltetrahydroisoquinoline hydrochloride."[5]

Q-190-C was prepared from a repeat preparation of IV-132 by repeated solution in chloroform and precipitation with ether. Q-190-C represented a final, chloroform-insoluble fraction.

Standard Preparations.—For these preparations the base was suspended in water and the whole chilled in ice. An exact equivalent of formalin solution was added followed by sufficient concentrated hydrochloric acid to make the final concentration 6 *N*. (As an example, 12.4 g. of N-methylhomoanisylamine was suspended in 15 cc. of water and 5.85 cc. of formalin solution added slowly, followed by 30 cc. of cold concentrated hydrochloric acid.) The flask was then covered with a watch-glass and heated on the steam-bath for four hours. All preparations subsequently identified as "standard preparation" were subjected to this treatment. GC-142-1 received one hour of heating, GC-142-2 two hours and the other GC-142 samples the number of hours indicated by the last digits. It will be observed that GC-142-4 was the most potent. A minor variation was to extract the amine-formaldehyde reaction mixture with ether before adding acid, to evapo-

(10) The occurrence of methylene substitution in positions other than those ortho to the methoxyl groups, while doubtless of minor quantitative importance, is to be expected and would add to the complexity of the polymer mixture, while presumably having little influence on the physiological properties.

(11) *Cf.* Goss, Houbart and Ingold, *J. Chem. Soc.*, 250 (1927); Ingold and Wilson, *ibid.*, 810 (1927).

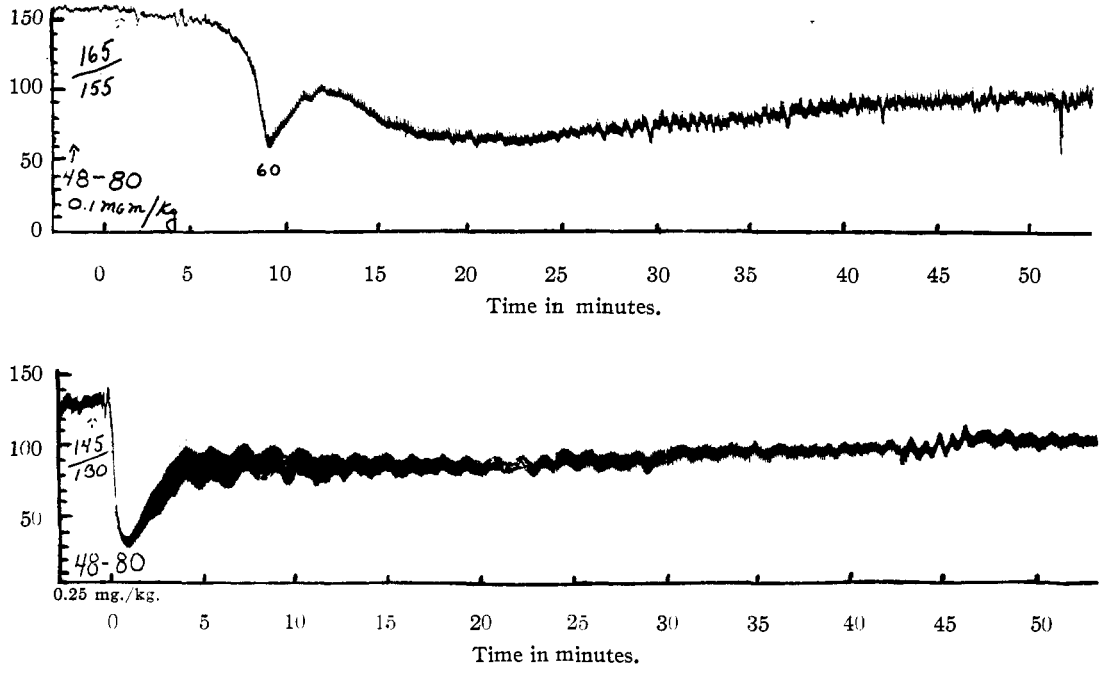


Fig. 1.—Blood pressure tracings for "Standard Preparations." The vertical scale indicates pressure in mm.

rate the ether and heat with acid as before. The products so obtained were not distinguishable from those previously described and the term "standard preparation" is intended to cover both. After completion of the heating period, the solution was evaporated *in vacuo* on the steam-bath and the residue was dissolved in a minimum of absolute ethanol (about 40 cc. for the quantity previously quoted). To the hot ethanolic solution was added an equal volume of ethyl acetate and the solution, which set to a jelly on cooling, was refrigerated overnight. About two volumes of anhydrous ether were then stirred in whereby the jelly was broken up to a colorless suspension of granular appearance. This was filtered rapidly, washed with anhydrous ether and dried *in vacuo*. Figure 1 shows typical blood pressure tracings with a "standard preparation" from *p*-methoxyphenethylmethylamine administered in two dose levels as indicated.

M-25 was a "standard preparation" from 3,4-dimethoxyphenethylmethylamine, M-27 from 2,3-dimethoxyphenethylmethylamine, GD-6 from hordenine. M-96 was a "standard preparation" from hordenine methyl ether and M-118 from hordenine methyl ether methochloride. The five samples GC-142-1 through GC-142-5 were obtained from *p*-methoxyphenethylmethylamine as previously indicated. M-114 was prepared from *o*-methoxyphenethyldimethylamine, GC-114 from *p*-methoxybenzylmethylamine, and GC-104 II from *o*-anisidine. GC-110 was the quaternary salt formed by the methylation of GC-104 II with methyl iodide in methanol (in the presence of sodium carbonate).

Isolation of "Dimers"

GC-60 from *p*-Methoxyphenethylmethylamine.—This was prepared by a variation of the "standard preparation" using perchloric acid instead of hydrochloric. To 6.4 g. of base was added 3.0 cc. of formalin and 20 cc. of 20% perchloric acid in the cold. The mixture was heated two hours on the steam-bath and placed in the refrigerator. The gummy solid that separated was crystallized twice from water giving 3.7 g. of a colorless micro crystalline powder. The perchlorate was converted to the base and transformed to the hydrochloride which melted at 261–262° (m. p. not raised by four recrystallizations from ethanol-ether mixtures).

Anal. Calcd. for $C_{21}H_{32}Cl_2N_2O_2$: C, 60.69; H, 7.77 Calcd. for $C_{32}H_{49}Cl_3N_3O_3$: C, 60.9; H, 7.84. Found: C, 60.91; H, 7.30.

The base was again liberated from its salt and was distilled at 0.4 μ pressure, bath temperature, 125–130°. By the Rast method the base showed a molecular weight of 329 (calcd. for $C_{21}H_{30}N_2O_2$, 342). On reconversion to the hydrochloride the m. p. was 264–265°.

GC-55 from 4-Methoxy-3-methylphenethyldimethylamine.—This amine was condensed with formalin and hydrochloric acid in the expectation that the "dimer" would be obtained to the virtual exclusion of other material. Actually, considerable amounts of more highly polymerized bases were formed (as shown by distillation of the base in high vacuum). The presumed "dimer" was isolated in poor yield from the reaction mixture by repeated crystallizations from ethanol–ether mixtures, m. p. 221–222°.

Anal. Calcd. for $C_{25}H_{40}Cl_2N_2O_2$: C, 63.66; H, 8.56. Calcd. for $C_{36}H_{57}Cl_3N_3O_3$: C, 63.88; H, 8.61. Found: C, 63.47; H, 8.51.

The analysis fits either a "dimer" or a "trimer," but in view of the failure to obtain any "trimer" crystalline that possibility appears remote. Some of the starting amine was recovered from the reaction mixture but since its hydrochloride melts at 181–182°, the 221–222° material is clearly a condensation product.

GC-125-I and GC-125-II.—These were products from an attempt to conduct a condensation so as to favor formation of a "trimer." Four grams of formalin solution, 9.65 g. of 4-methoxy-3-methylphenethyldimethylamine and 48 g. of concentrated hydrochloric acid were held at 30–40° for two hours during which time a current of hydrogen chloride was passed through the solution. The reaction mixture was then evaporated *in vacuo*. It was hoped that at this stage the main constituent would be a mono-chloromethyl derivative. To the residue from evaporation was then added 5.4 g. of *p*-methoxyphenethyldimethylamine hydrochloride and 10 cc. of concentrated hydrochloric acid and the solution was heated on the steam-bath for seven hours. The reaction mixture was again taken down *in vacuo*, redissolved in water and basified. GC-125-I was the fraction of base soluble in ether while GC-125-II was insoluble in ether but soluble in

ethyl acetate. The former fraction, as a base, had a "molecular weight" of 520 (Rast method). Both fractions were tested as hydrochlorides.

GC-114-II.—A portion (275 mg.) of base from the "standard preparation" with *p*-methoxyphenethylmethylamine (GC-114) was subjected to distillation up to 130° at 0.3 μ pressure. There distilled 115 mg. of oil giving a "molecular weight" of 244 (calcd. for "dimer," 314). The residue (GC-114-II), weighed 140 mg. and showed a "molecular weight" of 496 (calcd. for "trimer," 477).

Counter-current Distribution.—A "standard preparation" from hordenine methyl ether was largely freed of "dimer" by partial basification of its aqueous solution and solvent extraction. Fraction GC-81-II represented about 70% of the original preparation, about 25% being in the "dimer" fraction first removed. Two 1-g. portions of GC-81-II were separately subjected to distributions by the Craig method (using separatory funnels) between 50-50 benzene-hexane and 75% aqueous methanol (120 cc. of each solvent mixture in each funnel). On completion of an "eight-plate" distribution (with Tube 0 containing the initial upper layer and Tube 8 the final lower layer) the contents of each funnel was evaporated separately. The evaporation residues from the tubes in Run 2 were as follows: Tube 0, 50 mg.; Tube 1, 155 mg.; Tube 2, 205 mg.; Tube 3, 170 mg.; Tube 4, 110 mg.; Tube 5, 60 mg.; Tube 6, 65 mg.; Tube 7, 100 mg.; Tube 8, 110 mg. It was evident that about two-thirds of the material was contained in Tubes 0–4, inclusive, and that a separate hydrophilic component was concentrated in Tubes 7–8. Run 1 was an almost exact duplicate, but Tube 8 was lost by an accident.

The combined contents of Tubes 1–3 of the two runs amounting to 1.08 g. was redistributed and now gave a curve indicating homogeneity (Tubes 6, 7 and 8 contained 30, 15 and 20 mg., respectively). The contents of Tubes 2 and 3, transformed into hydrochlorides, were tested as GC-131-2 and GC-131-3. GC-131-2 gave no gas in the Zerewitinoff machine.

The combined contents of Tube 7 of Run 1, Tubes 7 and 8 of Run 2 and Tubes 7 and 8 of a third similar run, totaling 490 mg. was redistributed in the same fashion. The contents of Tubes 0–8 were as follows: Tube 0, 15 mg.; Tube 1, 5 mg.; Tube 2, 5 mg.; Tube 3, 10 mg.; Tube 4, 10 mg.; Tube 5, 35 mg.; Tube 7, 80 mg.; Tube 7, 155 mg.; Tube 8, 185 mg. The contents of Tube 8 was analyzed (as the base).

Anal. Calcd. for $C_{37}H_{55}N_3O_5$ (trimer with two hydroxy methyl groups): C, 71.40; H, 8.84; m. wt., 621. Found: C, 72.18; H, 8.72; m. wt., 630.

In a Zerewitinoff machine 14.77 mg. gave 1.20 cc. of gas at 25° and 771 mm. = 1.12 cc. at 0° and 760 mm. Calcd. for m. wt. 621 and two active hydrogen atoms: 1.07 cc. The remainder of this sample was dissolved in dilute hydrochloric acid and tested as GC-132-8.

One-tenth g. of fraction GC-131-2 was distilled as base, three fractions being taken. The first, boiling up to 175° at 0.2 μ weighed 40 mg. and gave a "molecular weight" of 296. Distillation was continued on the residue but nothing came over below 195°. A total of 25 mg. distilled, mainly at 208°. This showed a "molecular weight" of 572. (Calcd. for "trimer" with no hydroxy methyl groups, 561.) The residue weighed 25 mg. and showed a "molecular weight" of 557. It seems likely that the material suffered some decomposition.

Several larger distributions having been made, several terminal fractions amounting to 655 mg. were united and redistributed between 50-50 benzene–hexane and 55% aqueous methanol. The contents of Tubes 0–8 were as follows: Tube 0, 135 mg.; Tube 1, 95 mg.; Tube 2, 70 mg.; Tube 3, 75 mg.; Tube 4, 55 mg.; Tube 5, 40 mg.; Tube 6, 50 mg.; Tube 7, 45 mg.; Tube 8, 90 mg. The contents of Tube 0 gave a "molecular weight" of 746 (in camphor). The "tetramer" with one hydroxy methyl group would have mol. wt., 782. 27.8 mg. in the Zerewitinoff machine gave 0.61 cc. of gas (reduced to 0° and 760 mm.). Calcd. for mol. wt. of 782 with one active hydrogen, gas = 0.80 cc.

The contents of Tube 8 was not sufficiently soluble in camphor for a molecular weight determination. In borneol (observed mol. m. p. depression 33°; lit. value, 35.6°) the "molecular weight" was 567 (or 612 using the literature m. p. depression). A "trimer" with two hydroxy methyl groups would have mol. wt. = 621. In the Zerewitinoff machine 25.6 mg. gave 1.57 cc. of gas (reduced to 0° and 760 mm.). Calcd. for "trimer" with two active hydrogens, 1.85 cc.

Acknowledgment.—The authors wish to express their gratitude to Mr. Samuel Blackman, who performed the microanalyses, and to Mr. Lloyd Wnuck for assistance in preparing the blood pressure tracings.

Summary

1. The condensation of formaldehyde with alkoxyphenalkylamines in the presence of acid results in mixtures of low-polymeric bases that have been only partially separated into their constituents.

2. Those polymers having three or four residues of the parent amine joined by methylene groups between the rings have a pronounced and enduring effect in lowering the blood pressure.

3. Evidence is presented that some of these polymers have one or two hydroxy methyl groups attached (presumably to the terminal rings). Presence or absence of these groups is not critical to the effect on the blood pressure.

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