

29. The Amino-derivatives of Pentaerythritol. Part III. The Formation and Thermal Decomposition of Some Quaternary Salts of Tetrakisdimethylamino-methylmethane.

By GEOFFREY M. GIBSON and FREDERICK G. MANN.

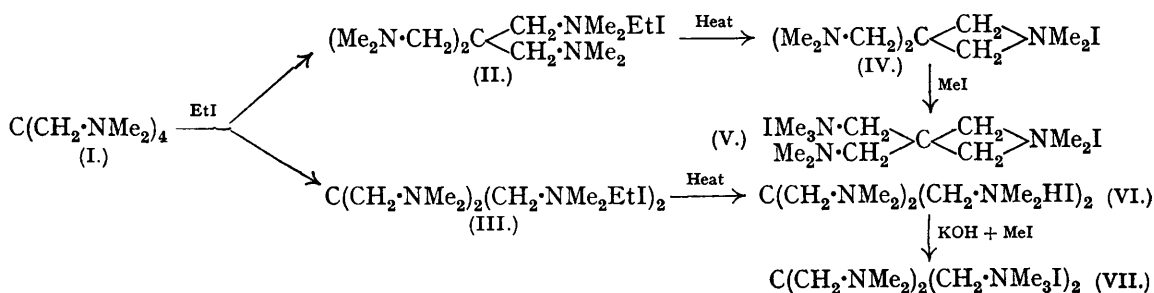
The reaction of this base with methyl iodide has been described in Part I (J., 1938, 1588). It is now shown that the base reacts with ethyl iodide and benzyl iodide to give mixtures of the mono- and bis-quaternary salts, and with allyl iodide to give the monoallyliodide and the allyliodide-hydriodide. Combination with three or four molecules of these alkyl iodides could not be obtained. The thermal decomposition of these compounds has been studied.

The reaction of the base with benzyl iodide in certain circumstances, however, causes rupture of the amine molecule with the formation of dibenzyltrimethylammonium iodide and the dibenzyliodide of the "pyro"-base described in Part II (preceding paper).

The mechanism of the above reactions is discussed.

TETRAKISDIMETHYLAMINOMETHYLMETHANE, $C(CH_2 \cdot NMe_2)_4$, reacts readily with methyl iodide, but even when the latter is in considerable excess, only the dimethiodide is formed; this is unchanged by boiling methyl iodide but on fusion gives the tetramethiodide (Part I; Litherland and Mann, J., 1938, 1588). In view of this abnormal result, the reactions of this octamethyl base with ethyl, allyl, and benzyl iodides have been investigated.

When the octamethyl base (I) is treated with ethyl iodide in the cold, the acetone-soluble monoethiodide (II) is formed; if, however, the original mixture is boiled for a short period and then set aside, the acetone-insoluble, very deliquescent *diethiodide* (III) is formed. No higher ethiodide could be detected. The solubility and instability of the monoethiodide (II) prevented its isolation; its presence was inferred, however, from the fact that when boiled in acetone solution it readily gave $\beta\beta$ -bisdimethylaminomethyl-N-methyltrimethyleneimine monomethiodide (IV), a compound which is similarly formed also by the decomposition of the monoallyliodide and monobenzyliodide of the base (I) (see below). The

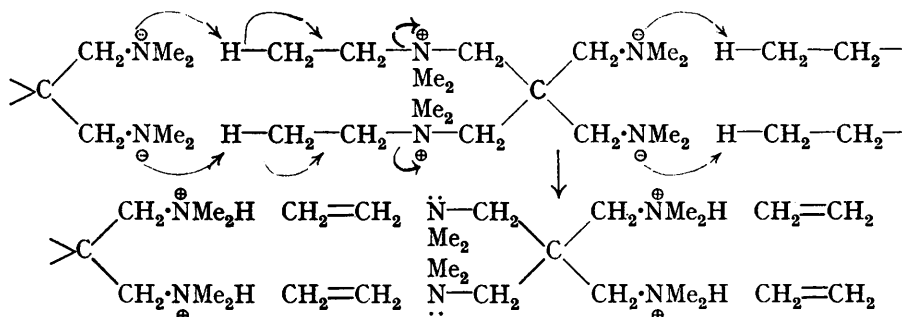


cyclic monomethiodide (IV), when treated with an excess of methyl iodide, combined with only one more molecule, giving the *dimethiodide* (V); apparently the same factors that inhibit the formation of the tetramethiodide of the octamethyl base (I) here inhibit that of the expected trimethiodide.

When the diethiodide (III) was heated, the *dihydriodide* (VI) of the octamethyl base was formed, accompanied by a vigorous evolution of gas, presumably ethylene; the identity of (VI) was confirmed by the fact that when it was treated in turn with potassium hydroxide and methyl iodide, the normal dimethiodide (VII) of the octamethyl base was obtained.

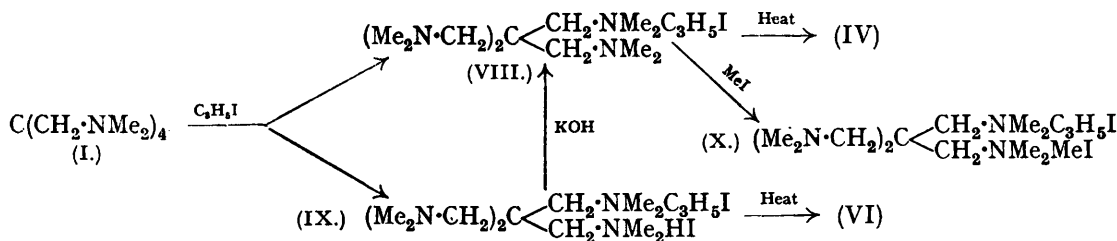
The reason for this marked difference in the thermal decomposition of the diethiodide (III) and the dimethiodide is not far to seek. One of the chief factors in the decomposition of quaternary ammonium hydroxides (and many salts) containing an ethyl group is the elimination of a β -proton from

this ethyl group by the hydroxyl ion, with the formation of ethylene, water, and a tertiary amine (Hanhart and Ingold, J., 1927, 997). A parallel reaction is possible with the diethiodide (III), but now, however, the lone pair of electrons on the two tertiary nitrogen atoms of one molecule will fulfil the function of the hydroxyl ions, and will extract the β -protons from the two ethyl groups of a second molecule. The process will therefore be :



This mechanism explains the formation of the dihydriodide and the evolution of ethylene. The dimethiodide, on the other hand, could clearly give no such reaction, nor by a parallel reaction could the methyl groups split off as methylene radicals and so furnish ethylene, as there is no known example of methyl groups behaving thus during quaternary ammonium decomposition (Hanhart and Ingold, *loc. cit.*); hence the entirely different behaviour of the dimethiodide on heating.

When the octamethyl base (I) was treated with an excess of allyl iodide in the cold, the only products isolated were the acetone-soluble *monoallyliodide* (VIII) and the acetone-insoluble *monoallyliodide monohydriodide* (IX); the constitution of the latter was confirmed by the fact that potassium hydroxide converted it into (VIII). The compound (VIII) on heating passed smoothly over into the cyclic trimethyleneimine compound (IV); when, however, it was treated even with an excess of methyl iodide, only the *monoallyliodide monomethiodide* (X) was formed. The allyliodide hydriodide (IX) on fusion gave the dihydriodide (VI) of the octamethyl base, a reaction similar to that of the diethiodide.



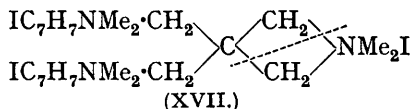
The occurrence of the allyliodide hydriodide (IX) was unexpected, and was not due to the *thermal* decomposition of an intermediate product, since the compound (IX) could be isolated by the use of cold solvents alone. It is possible, however, that a very unstable diallyliodide was formed, and even at room temperature decomposed by a reaction similar to that shown above for the diethiodide but now affecting only one quaternary allylammonium ion. The allyliodide hydriodide (IX) so formed would have greater stability and therefore require heating before the decomposition was repeated with the formation of the dihydriodide (VI). This we regard as the most probable explanation of the formation of the above compounds. Furthermore, the fact that a stable diallyliodide of the octamethyl base does not apparently exist, whereas the diethiodide (III), dibenzyl iodide (XIV), and also mixed salts such as the allyliodide-methiodide (X) and the allyliodide-benzyl iodide (XVI) (see below) have all been isolated is not unexpected, for von Braun (*Annalen*, 1911, **382**, 5) has shown that in quaternary ammonium decompositions the ease of elimination of groups is allyl > benzyl > ethyl > methyl; clearly, therefore, the diallyliodide should be the least stable of the above bisquaternary salts.

The action of benzyl iodide on the octamethyl base followed that of ethyl and allyl iodides, with certain remarkable differences. When the base and benzyl iodide were mixed with cooling in the absence of a solvent, a pale brown, brittle glass was obtained (cooling was essential, otherwise the considerable heat evolution caused almost complete decomposition of the mixture). The glass was separated by crystallisation from acetone and alcohol into dibenzyl dimethylammonium iodide (XI) and the dibenzyl iodide of the "pyro"-base (XII) described in Part II, *i.e.*, $\alpha\gamma$ -bisdimethylamino- β -methylpropene. It

The reason for the formation of dibenzyltrimethylammonium iodide (XI) and the dibenzylidene (XII) of the "pyro"-base when the octamethyl base (I) was treated with excess benzyl iodide in the absence of a solvent is obscure, but may be due to the monobenzylidene (XIII) of the octamethyl base initially formed undergoing ready decomposition into the trimethyleneimine compound (IV) and benzyl-

dimethylamine. The latter would then unite with benzyl iodide to give (XI), whilst the compound (IV), under the influence of the excess of benzyl iodide, would first give a mono- or di-benzyl derivative such as (XVII) and the latter would then decompose by fission of the ring as described in Part II (p. 167), forming the unstable precursor of the "pyro"-base dibenzylidide (XII).

It may be objected that this radical cleavage of the molecule (XVII) is unlikely to occur without vigorous heating. It is well known, however, that benzyl groups in quaternary ammonium derivatives have an exceptional lability and that such derivatives in consequence often decompose very much more readily than most other quaternary derivatives (cf. Graymore, J., 1941, 39).



One striking result emerging from this investigation is that the octamethyl base (I), when treated with an excess of alkyl iodide under normal conditions, apparently never unites with more than 2 mols. of it. Even this formation of a bisquaternary salt occurs readily only with methyl iodide; in the above experiments with ethyl and benzyl iodide the mono- was always formed in much greater yield than the bisquaternary salt, and the addition of the second molecule of alkyl iodide must presumably be much slower than that of the first. Furthermore, only the dimethiodide on heating gave the quadri-quaternary salt, the other dialkylidides giving the dihydriodide. It was suggested in Part I that this extreme difficulty in forming quadri-quaternary salts was probably partly steric, but was due mainly to increasing energy being required for successive alkyl iodide additions, *i.e.*, these additions changed steadily from exothermic to endothermic reactions. The steric factor is probably a comparatively minor one, since other cases are known where this factor can hardly be present at all; *e.g.*, it has been shown that trimethyl- and triethyltrimethylenetriamine when treated with excess methyl or ethyl iodide, in spite of a vigorous reaction, give only the mono-quaternary salt (Graymore, J., 1938, 1311; Blundell and Graymore, J., 1939, 1787). Phenazine, when treated with methyl sulphate even at 120°, also gives only a mono-quaternary salt (Kehrmann and Havas, *Ber.*, 1913, 46, 343). Similarly, both isomeric forms of 5:10-di-*p*-tolyl-5:10-dihydroarsanthren unite with only 1 mol. of methyl iodide, the second arsenic atom remaining persistently 3-covalent (Chatt and Mann, J., 1940, 1184). The electronic explanation of the behaviour of the octamethyl base is probably that the positive pole on one nitrogen atom formed by the union of the first alkyl iodide molecule exerts a marked inductive effect which in turn causes a certain restraint on the lone-electron pairs of the remaining three nitrogen atoms. This restraint hinders but does not prevent the addition of a second alkyl iodide molecule. After this second addition, however, the increased inductive restraint on the lone pairs of the remaining two nitrogen atoms prevents further quaternary-salt formation, except in the case of the methiodides, where a high temperature suffices for this purpose. A similar mechanism can clearly be put forward to account for the behaviour of the above trimethylenetriamine, phenazine, and arsanthren compounds, and, of course, for the fact that the trimethyleneimine methiodide (IV) will unite with only one more molecule of methyl iodide.

EXPERIMENTAL.

Reaction with Ethyl Iodide.—(a) A solution of the octamethyl base (I) in ethyl iodide (8 mols.) was boiled under reflux for 15 mins. and then set aside in the dark for 7 days. Some preparations had then deposited colourless crystals of the *diethiodide* (III), which were collected, triturated thoroughly with acetone, and recrystallised from alcohol. Other similar preparations, however, sometimes deposited a dark yellow syrup; in this case, the excess of ethyl iodide was allowed to evaporate at room temperature and the residual syrup was stirred vigorously with acetone, crystallisation of the diethiodide usually occurring at once; sometimes, however, the crude syrup dissolved completely in the acetone, which then slowly deposited the colourless crystals. The diethiodide was thus obtained as very deliquescent crystals, m. p. 128° (Found: N, 9.7; I, 45.3. $\text{C}_{17}\text{H}_{42}\text{N}_4\text{I}_2$ requires N, 10.1; I, 45.65%). The acetone washings contained appreciable quantities of the monoethiodide. Unless the base (I) and the ethyl iodide employed in the above preparation are pure, the yellow syrup obtained will not give a crystalline product on treatment with acetone.

(b) When, however, the preliminary heating was omitted from otherwise identical experiments, a homogeneous solution was obtained. A test portion, when evaporated and treated with acetone, gave no precipitate of the diethiodide. The main solution, which presumably contained the monoethiodide, was therefore boiled under reflux for 15 mins., evaporated to dryness on the water-bath, and then dissolved in hot acetone. The solution, on standing overnight, deposited a small crop of the slightly impure dihydriodide (VI), m. p. 204–207°, formed apparently by decomposition of traces of the diethiodide. The mother-liquor, after filtration, was then evaporated to dryness, the residue heated on the water-bath for several hours (during which a faint amine odour was evolved), and then recrystallised from acetone, colourless crystals of $\beta\beta$ -*bis*-dimethylaminomethyl-*N*-methyltrimethyleneimine monomethiodide (IV), m. p. 206.5–207.5°, being deposited; a mixture of these crystals with those of the dihydriodide (VI) had m. p. 181–185°. Further successive

crops of (IV) were obtained by repeating the evaporation of the acetone mother-liquor to dryness, followed by heating and recrystallisation as before. The united crops, when recrystallised from alcohol, gave the pure monomethiodide (IV), m. p. 208—208.5° (decomp.) (Found: C, 41.0; H, 8.3; N, 12.5; I, 38.4. $C_{11}H_{26}N_3I$ requires C, 40.4; H, 8.0; N, 12.8; I, 38.8%). The identity of (IV) as a methiodide and not a hydriodide was confirmed by the fact that ether extracted no amine from a mixture of (IV) with concentrated aqueous potash.

When a mixture of the monomethiodide (IV) and of methyl iodide was boiled under reflux for 6 hours, and the excess of methyl iodide then evaporated, the residue, after extraction with cold acetone, consisted of much unchanged monomethiodide and the corresponding dimethiodide (V). This residue, when recrystallised from alcohol, furnished the monomethiodide. Dilution of the alcoholic mother-liquors with acetone caused crystallisation of the *dimethiodide*, which, when washed with acetone and recrystallised from alcohol, separated as the *dihydrate*, m. p. 123—124° (decomp.; efferv.) (Found: N, 8.1; I, 50.8. $C_{12}H_{28}N_3I_2 \cdot 2H_2O$ requires N, 8.3; I, 50.3%).

Thermal Decomposition of the Diethiodide (III).—The carefully dried diethiodide was plunged into an oil-bath at 128°, and the temperature was then increased to 135—140° and kept thereat for 20 mins., by which time effervescence had ceased and a reddish-brown syrup remained. Addition of acetone to a solution of this syrup in a minimum of alcohol caused separation of a pale brown solid, which was recrystallised from alcohol, washed with acetone until colourless, and again recrystallised. The *monohydrated dihydriodide* (VI) of the octamethyl base was thus obtained, m. p. 208—209° (Found: C, 29.9; H, 6.8; N, 11.0; I, 49.3. $C_{13}H_{34}N_4I_2 \cdot H_2O$ requires C, 30.1; H, 7.0; N, 10.8; I, 49.0%). The identity of this compound was confirmed by saturating its aqueous solution with potassium hydroxide and then extracting it with ether. Evaporation of the ether gave an oil which rapidly combined with methyl iodide to give the dimethiodide of the octamethyl base, m. p. 145—146°, unchanged by admixture with an authentic sample.

Reaction with Allyl Iodide.—Allyl iodide (4 mols.) was slowly added to the octamethyl base (I) with stirring and ice-water cooling, and the mixture set aside in the dark overnight. The viscous product was well stirred with a small quantity of cold acetone; the latter was then decanted and on keeping (particularly if diluted with ethyl acetate) deposited the pure *monoallyliodide* (VIII) of the octamethyl base as colourless crystals which on heating melted with effervescence at 145—146°, then resolidified and melted again with effervescence at 207°, these values being unchanged after recrystallisation from alcoholic ethyl acetate (Found: C, 46.5; H, 9.1; N, 13.6; I, 30.7. $C_{16}H_{37}N_4I$ requires C, 46.6; H, 9.0; N, 13.6; I, 30.8%).

The sticky insoluble residue from the acetone extraction, when vigorously triturated with fresh acetone, ultimately solidified, giving thus crystals of the *monoallyliodide monohydriodide* (IX), m. p. 153—154°; these, when triturated afresh with alcohol and acetone and then recrystallised from alcohol, gave the pure salt (IX), m. p. 157—158° (Found: C, 35.4; H, 7.0; N, 10.5; I, 46.2. $C_{16}H_{37}N_4I_2$ requires C, 35.6; H, 7.0; N, 10.4; I, 47.0%). It is clear from the above results that the compound (IX) must be formed during the original reaction and at room temperature, and did not arise when the products of this reaction were in contact with hot solvents.

A 0.86% methyl-alcoholic potassium hydroxide solution (6.0 c.c., *i.e.*, slightly less than 1 mol. of KOH) was added to a suspension of the hydriodide (IX; 0.5 g.) in methyl alcohol. The clear solution was evaporated to dryness at room temperature, extracted with cold acetone to remove a trace of unchanged material, and the acetone solution filtered and evaporated. The residue, when recrystallised from alcoholic ethyl acetate, furnished the monoallyliodide (VIII), melting with effervescence at 145—146°, and resolidifying and finally remelting at 207—208°, these values being unchanged by admixture with an authentic sample.

Thermal Decompositions.—(a) *Of the monoallyliodide (VIII).* The substance was heated at 160—165° for a few minutes, during which an amine odour was evolved. The residue, which solidified on cooling, was washed with acetone until colourless, and thus furnished the pure trimethyleneimine compound (IV), m. p. 210—210.5°, unchanged by admixture with previous specimens (Found: I, 39.1%).

(b) *Of the allyliodide hydriodide (IX).* The hydriodide was heated at 170—175° for 15 mins. An alcoholic solution of the brown, sticky residue was diluted with acetone, and the colourless needles which separated were recrystallised from alcohol, furnishing the dihydriodide (VI), m. p. and mixed m. p. 210.5—211.5° (Found: I, 48.5%).

A mixture of the monoallyliodide (VIII) and methyl iodide was boiled under reflux for 4 hours, and the excess of methyl iodide then evaporated: the residue, after thorough washing with acetone, furnished the pure and very deliquescent *monoallyliodide monomethiodide* (X), m. p. 114—115° (decomp., efferv.), unchanged by recrystallisation from alcohol (Found: N, 9.8; I, 46.0. $C_{17}H_{40}N_4I_2$ requires N, 10.1; I, 45.8%). When a thoroughly dry sample of this compound was plunged into an oil-bath at 115—120° and kept at this temperature for 30 mins., a sharp odour, unlike that of an aliphatic amine, was first evolved, followed by that of an amine itself. The viscous, pale brown residue, when mixed with alcohol, gave colourless crystals in small yield; these were purified by adding water to their suspension in boiling alcohol until a clear solution was obtained. Cooling gave crystals, which when thoroughly dried did not melt below 355° and were apparently the tetramethiodide of the octamethyl base (I); the small amount available made identification uncertain (Found: C, 24.6; H, 6.1; N, 7.5; I, 61.4. Calc. for $C_{17}H_{40}N_4I_4$: C, 25.1; H, 5.45; N, 6.9; I, 62.5%). When an aqueous solution of this compound was treated with potassium hydroxide (1 mol.) and then evaporated

to dryness and thoroughly extracted with alcohol to remove any potassium iodide, the N:I ratio in the residue was still 1:1; hence the above compound could not have been the trimethyliodide hydriodide, $C(CH_2 \cdot NMe_2)_4 \cdot 3CH_3I \cdot HI$.

Reaction with Benzyl Iodide.—(1) The base (I) (5 g.) was chilled in ice-water, and freshly prepared benzyl iodide (10.5 c.c., 4 mols.) slowly added. The mixture was vigorously shaken to prevent crystallisation of the benzyl iodide, and the corked flask containing the mixture returned to the ice-water and there set aside for six days, during which the mixture rapidly became gelatinous, and ultimately set to a hard, pale reddish-brown glass, which when gently warmed melted to a viscous liquid. (a) A portion of this glass was dissolved in boiling water, and the solution, after spontaneous cooling and filtration from a slight gummy deposit, gave on stirring colourless crystals of dibenzylidimethylammonium iodide (XI), m. p. after recrystallisation from water, 191—192° (Found: C, 54.6; H, 6.0; N, 4.0; I, 36.2. Calc. for $C_{16}H_{20}NI$: C, 54.5; H, 5.7; N, 4.0; I, 36.1%). Emde (*Arch. Pharm.*, 1909, **247**, 355) gives m. p. 191°. (b) An alcoholic solution of a second portion slowly deposited a cream-coloured, crystalline powder, m. p. 162—164°, which on further alcoholic crystallisation gave colourless crystals of the dibenzyl iodide (XII) of the "pyro"-base, m. p. 168—169°, unchanged by admixture with an authentic sample (Found: N, 4.7; I, 44.1. Calc. for $C_{22}H_{32}N_2I_2$: N, 4.85; I, 43.9%). The identity of this compound was further confirmed by conversion into the dibenzylpicrate, m. p. 199—201°, also unchanged by admixture with a known sample (Found: N, 14.45. Calc. for $C_{34}H_{36}O_{14}N_8$: N, 14.4%). An aqueous solution of (XII) was kept at 100° for 2.5 hours, and the unchanged compound (XII), m. p. 166°, then crystallised; a similar result was obtained in hot alcoholic solution. It is clear, therefore, that in the above preparation the compound (XI) could not have arisen by decomposition of (XII). (c) The main bulk of the original glass was now dissolved in much boiling acetone, which on cooling and standing for 2 days slowly deposited small, pale yellow crystals. These were collected and washed with acetone until colourless, and were now only slightly soluble in boiling acetone; either the solubility of this compound had originally been increased by the presence of other substances (a common occurrence in this investigation) or, less probably, the compound had been formed in the hot acetone solution. The colourless crystals, twice recrystallised from alcohol, gave the pure dibenzyl iodide (XII), m. p. and mixed m. p. 168—169°. The alcoholic mother-liquors, on spontaneous evaporation, deposited these colourless crystals and also large, very pale yellow crystals of the iodide (XI), m. p. 191—192°, mixed and unmixed.

(2) When, however, the above quantities of the base (I) and benzyl iodide, each now dissolved in ether (20 c.c.), were mixed with ice-cooling, and the mixture set aside for 24 hours in the dark, a crystalline solid (A) separated, and a hard glassy film (B) remained attached to the walls of the flask. The crystals (A) were collected, washed with ether, and dissolved in a minimum of hot acetone-ethyl acetate (1:1 by vol.); after 24 hours a small crop of the colourless crystalline *monohydrate* of the *dibenzyl iodide* (XIV), m. p. 128—129° (efferv.), unchanged by recrystallisation from alcohol, was collected (Found: C, 46.6; H, 7.3; N, 7.9; I, 36.5. $C_{27}H_{46}N_4I_2 \cdot H_2O$ requires C, 46.4; H, 6.9; N, 8.0; I, 36.4%). The acetone-ethyl acetate mother-liquor, chilled in ice, then deposited a larger crop of the pure *monobenzyliodide* (XIII), m. p. 146—147° (with resolidification and remelting at 190—196°), unchanged by recrystallisation from ethyl acetate containing a trace of alcohol (Found: C, 51.8; H, 8.4; N, 12.3. $C_{20}H_{38}N_4I$ requires C, 51.9; H, 8.4; N, 12.1%). It is noteworthy that the compound (XIV) is only slightly, and the compound (XIII) readily, soluble in cold acetone.

The glass (B), when boiled with acetone containing a small quantity of alcohol, was converted into colourless crystals, which, washed with acetone and dried, had m. p. 139—145°. These were boiled with sufficient alcohol to dissolve about half the material. The insoluble portion, recrystallised from much alcohol, gave the *benzyliodide hydriodide* (XV) of the octamethyl base, m. p. 170° (Found: C, 39.7; H, 7.0; N, 9.5; I, 42.8. $C_{20}H_{42}N_4I_2$ requires C, 40.6; H, 6.8; N, 9.5; I, 43.0%). The alcoholic mother-liquor slowly deposited crystals which were again fractionally extracted with hot alcohol; the latter extract, filtered and cooled, deposited the hydrated dibenzyl iodide (XIV), m. p. 128—128.5° after further recrystallisation (Found: N, 7.7; I, 36.1%). Further quantities of (XIV) were also slowly deposited from the alcoholic acetone with which the glass (B) had originally been treated.

Thermal Decompositions.—(a) *Of the monobenzyliodide (XIII).* This compound, heated at 150—155° for 5 mins., gave a mixture of a liquid and a crystalline solid which, when washed in turn with ether and acetone, furnished the trimethyleneimine compound (IV); this, after crystallisation from alcohol, had m. p. and mixed m. p. 209—210° (Found: I, 39.3%). The ethereal extract reacted readily with methyl iodide to give benzyltrimethylammonium iodide, m. p. 177—178° after recrystallisation from alcoholic ethyl acetate (Found: I, 45.7. Calc. for $C_{10}H_{16}NI$: I, 45.8%). Emde (*loc. cit.*) gives m. p. 179°.

(b) *Of the dibenzyl iodide (XIV).* An alcoholic solution of (XIV) was boiled under reflux for 6 hours, the solvent removed, and the syrupy residue dissolved in hot acetone. Cooling gave colourless crystals of the dihydriodide (VI), which, after being washed with acetone and recrystallised from alcohol, had m. p. and mixed m. p. 212—213°.

A concentrated acetone solution of the monobenzyliodide (XIII), to which excess of allyl iodide had been added, was set aside in the dark for 3 days. The crystalline *monobenzyliodide-allyliodide* (XVI) was then collected and washed with acetone; m. p. 145—146° (efferv.) unchanged by recrystallisation from alcohol (Found: C, 43.2; H, 7.3; N, 8.9. $C_{23}H_{44}N_4I_2$ requires C, 43.8; H, 7.0; N, 8.9%).

All the compounds isolated in this investigation were dried overnight in a vacuum over phosphoric anhydride,

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and the hydrates recorded must clearly have considerable stability. It is noteworthy that many alkyl halides of the octamethyl base (I) possess m. p.'s which, although sharp, vary considerably with the rate of heating. All mixed m. p.'s were therefore determined side by side with the original material.

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THE UNIVERSITY CHEMICAL LABORATORY, CAMBRIDGE.

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