United States Patent Office 2,750,389 Patented June 12, 1956

2,750,389

QUATERNARY AMMONIUM COMPOUNDS AND PROCESS OF PREPARING THEM

Raymond A. Pingree, Cranston, and Chester U. Stevens, Saylesville, R. I., assignors to Crown Chemical Corporation, Providence, R. I., a corporation of Rhode Island

No Drawing. Application June 24, 1954, Serial No. 439,127

20 Claims. (Cl. 260—295)

This invention relates to new quaternary ammonium compounds. This invention also relates to the process for the manufacture of these aforesaid compounds.

It is an object of the present invention to prepare new quaternary ammonium compounds which are water-dispersible, the exact chemical formula of which is not known, by a series of steps in the synthesis thereof and the intermediate compounds which are known. The quaternary ammonium compounds of this invention are characterized by the capacity of imparting permanent water-repellency to textile materials.

In the U. S. patent granted to Alfred Wm. Baldwin and Eric Everard Walker, identified as No. 2,146,392, patented February 7, 1939, there is described a quaternary ammonium salt depiced as having the general formula

$$R{-}CO{-}NH{-}CH_2{-}\underset{\displaystyle X}{\underset{|}{N}}(tert)$$

where R stands for an aliphatic hydrocarbon radical of at least 10 carbon atoms and X stands for an anion of a hydrogen halide acid. Some of the specific compounds of the patent are stearamidomethylpyridinium chloride, the corresponding sulfate; the lauryl and the oleyl derivatives of the methyl pyridinium chloride. Those whose hydrocarbon radical is of a higher fatty acid, these inventors aver are useful as agents in textile treating processes. There is employed in the preparation of these compounds an intermediate which is formed from an acid amide of the formula $R{-}CO{-}NH_2$, when formaldehyde is a reactant, otherwise a hydroxy-methyl-amide.

There are other quaternary ammonium compounds capable of imparting water repellency to textile fibers which compounds are derived from the reaction of pyridine and an intermediate prepared from the amide of a higher fatty acid, formaldehyde and an acid. None of these is the same as the products which will now be described.

According to the present invention, we initially react a halomethyl acyl amide of the general formula

$$R.CONH.CH_2X$$

where R is a paraffin hydrocarbon radical and X is a halogen, with an alkyl nitrile; the acyl group of said amide has a chain length of not less than 10 carbon atoms and the alkyl nitrile has a chain length of not less than 10 carbon atoms. This resulting compound is treated with formaldehyde and a hydrogen halide, specifically hydrogen chloride, to form a halomethyl compound of this first intermediate, specifically the chloromethyl derivative; the exact composition of this second intermediate has not been definitely established. This last described compound is then reacted with a tertiary amine from the group consisting of pyridine and the C-alkyl substituted pyridines. These include all isomers of picoline, lutidine and collidine. There is formed a quaternary ammonium compound which is water dispersible.

We have not been able to establish proof of the structure of the compounds formed in our process. We believe, however, that the reaction between the alkyl nitrile and the chloromethyl-acylamide does proceed to form an imino chloride, thus:

$$R'CN + RCONH.CH_2Cl \longrightarrow R'\underset{\displaystyle Cl}{\underset{|}{C}}{=}NCH_2NHCOR$$

and that the chloromethylation of the imino chloride proceeds:

$$R'\underset{\displaystyle Cl}{\underset{|}{C}}{=}NCH_2NHCOR + HCHO + HCl \longrightarrow$$

$$R'\underset{\displaystyle Cl}{\underset{|}{C}}{=}NCH_2\overset{\displaystyle CH_2Cl}{\overset{|}{N}}CO.R + H_2O$$

The final quaternary compound would be:

$$R'\underset{\displaystyle Cl}{\underset{|}{C}}{=}NCH_2\overset{\displaystyle CH_2N(tert)\,Cl}{\overset{|}{N}}CO.R$$

Alternatively according to the invention the new quaternary ammonium compounds may be manufactured by reacting together a hydroxymethyl amide and an alkyl nitrile, the acyl radical of which amide and the straight chain hydrocarbon of the organic nitrile each having the same significance as presently described, with hydrogen chloride to form the first product or intermediate. It is believed that the hydrogen chloride initially reacts with the hydroxymethyl amide to form the acyl amido methly chloride which reacts with the alkyl nitrile. This first intermediate in situ in turn is then reacted with formaldehyde and hydrogen chloride to form the chloromethyl derivative of this first intermediate, which second compound is then reacted with the tertiary amine of the type previously described to form the quaternary ammonium compound of the present invention.

Another alternative is to react the paraffin organic acid amide with the alkyl nitrile, and in the presence of formaldehyde or a polymeric form thereof and an inorganic halide which releases a hydrogen halide, such as one of the phosphorus poly-halides or thionyl chloride, then reacting this product with pyridine or a C-alkyl substituted pyridine. It is believed that the order of reactions is the same as in the general presentation supra.

Suitable halomethyl acyl amides, specifically the chloromethyl acyl amide, for use in the present process are those whose acyl group is of the fatty acid type and has a chain length of not less than 10 carbon atoms. As the chain length of the acid amide falls below C_{16} water-repellency of treated fabrics becomes progressively poorer. The preferred length is C_{16} and above, up to and including C_{22}; no adverse effect has been observed with such. Inasmuch as the acid amide derivatives, whose acyl group has a length greater than C_{22}, are not commercially available, there is a practical range imposed in the selection. And, in addition, the quaternary ammonium compounds from the members of such the C_{16}–C_{22} range possess less water solubility. Specific chloromethyl amides of the higher fatty acids having ten or more carbon atoms are such amides of the following acids: capric, lauric, myristic, palmitic, stearic, arachidic, behenic, cerotic, melissic and montanic.

One chemical designation of the above discussed reactant is an acyl amido methyl chloride and other designation a chloromethyl acyl amide.

The alkyl nitriles which are used in the process should have a chain length of not less than 10 carbon atoms. As the chain length thereof falls below C_{16}, performance, i. e., water-repellency of treated fabrics, becomes progressively poorer. As is the case with the acid amides no adverse effect has been observed with this reactant whose

chain length is above C_{16}, the preference being C_{16} to C_{22} inclusive. The element of commercial availability of the highest members of this series and the aspect of decreasing water solubility of the quaternary ammonium compounds from the members of such are aspects which deserve mention. Specific compounds are: caprinitrile (decane nitrile), myristonitrile (tetradecane nitrile), palmitonitrile (hexadecane nitrile), stearonitrile (octadecane nitrile), arachidonitrile (eicosane nitrile), cerotonitrile (hexacosane nitrile), melissonitrile (triacontane nitrile), the designations in parenthesis being one system of nomenclature.

The formaldehyde employed in the reaction in the second step referred to above in the general description of the present invention may be in the monomeric or polymeric form. As a matter of practical use paraformaldehyde is the form employed.

In reference to the C-alkyl substituted pyridines which may be employed instead of pyridine itself in the final step of the synthesis of the quaternary ammonium compounds, the selection of the various isomers of picoline which is methyl pyridine, of lutidine which is dimethyl pyridine and of collidine is partly one of commercially availability and cost factor. The isomers of this last mentioned compound are 4-ethyl-2-methyl; 3-ethyl-4-methyl and 2, 4, 6 trimethyl pyridine. Another polysubstituted hydrocarbon pyridine is 2-methyl-5-ethyl pyridine.

In place of hydrogen chloride other compounds of chlorine which release hydrogen chloride, such as phosphorus trichloride (PCl_3), phosphorus pentachloride (PCl_5) and thionyl chloride ($SOCl_2$), may be used.

Only the chloromethyl acid amides are commercially available, whereas the bromo and iodo derivatives are not. The chloromethyl acid amides are preferred anyway.

Unsaturated hydrocarbon acid amides and nitriles may be tolerated in mixtures with the fully saturated compounds. Commercial amides and nitriles containing up to 5% of oleic acid derivatives have been successfully used in the process. Appreciably higher percentages of the unsaturated hydrocarbon compounds cause a serious loss of water repellency in the performance of the resulting quaternary ammonium compounds.

It is preferred to carry out the preparation of the intermediates and the quaternary ammonium compound in the presence of an inert liquid solvent for the reactants which prevents the mass from solidifying during the process, thus facilitating handling and control. Usually in the last step of the synthesis considerably less organic liquid solvent is present however. Some of the liquid organic solvents of this type are: benzene, dioxane, ethylene dichloride, isoheptane and ethyl acetate.

Because of the difficulty of isolating some of the chloromethyl acyl amides which are reacted with the alkyl nitrile as the first step, we have found it convenient to prepare the chloromethyl acid amide in situ by the conventional methods.

Suitable proportions in preparing the quaternary ammonium compounds of the present invention and the intermediates in the synthesis are 1 mol equivalent of a chloromethyl acyl amide of the formula given in the fourth paragraph of the presentation, approximately 0.5–1.5 mol equivalents of a straight chain alkyl nitrile containing not less than 10 C atoms, approximately 0.5–1.5 mol equivalents of formaldehyde and 0.5–1.5 mol equivalents of a chlorinating agent which may be hydrogen chloride or an inorganic compound capable of releasing hydrogen chloride under the conditions of operations, and approximately 0.5–1.5 mol equivalents of a tertiary amine from the group consisting of pyridine and C-alkyl substituted pyridines.

Preferred temperature range for the reaction between the chloromethyl acyl amide and the alkyl nitriles, and also for the chloromethylation of this intermediate with formaldehyde and hydrogen chloride is 60° to 75° C. inclusive. Formation of the final product, the pyridinium compound, takes place readily at temperatures of 50° to 75° C. inclusive. No advantage has been observed by conducting these reactions at any specific temperatures within the limits stated. These temperatures should not be considered as critical. Below the lowest temperatures mentioned supra the speed of reaction tends to slacken. Generally speaking, temperatures above the highest in these ranges offer no advantage in yield, and increase of temperature entails a costlier condition of operation. The reaction temperature necessary to give optimum yields varies with the reactants and proportions.

It will be understood that the acyl amido methyl chloride reactant need not be that of one fatty acid, since mixtures of two or more of the general formula

$$R.CONH.CH_2X$$

where R denotes an aliphatic hydrocarbon and X a halogen, may be employed. And mixtures of the other reactants, to wit, the alkyl nitrile and the tertiary amine, etc., may be employed. To confer latitude in properties this may be desirable.

The new quaternary ammonium compounds of the present invention have the characteristic that they impart a high degree of water-repellency to textile materials, including fabrics, yarns and thread. This water-repellency is, moreover, not seriously reduced by repeated launderings and/or dry cleanings. We have found that the water-repellent characteristics of cellulosic fabrics treated with compounds of the present invention are more resistant to laundering than those which are the invention of U. S. Patent #2,146,392, to which patent reference has previously been made, wherein the acyl-containing radical is the same. These compounds can be successfully applied to cellulosic fibers (cotton, linen, regenerated cellulose rayon, etc.), cellulose ester fibers (acetate rayon), polyamide fibers ("nylon"), protein fibers (wool, "Vicara," silk), acylic fibers ("Acrilan," "Orlon," "Dynel") and polyester fibers ("Dacron"). The treatment of textile materials with the compounds of the present invention is more fully described in our copending patent application Serial No. 439,128 filed of even date herewith. There is left a water-repellent residue or finish on the treated fiber. The compounds of the present invention thus have uses of practical technical merit.

No contention is made that the products secured in the subsequent examples are substantially pure. The quaternary ammonium compounds are customarily left in the reaction mass, and the latter is used as a whole in the process of treatment of textiles to confer thereto water-repellent properties.

Without limiting our invention to any particular procedure, the following examples will serve to illustrate our preferred mode of operation:

EXAMPLE I

Three hundred thirteen (313) grams of methylol stearamide is mixed with six hundred (600) grams of benzene to form a slurry. While agitating vigorously and maintaining the temperature at 40° to 60° C., forty (40) grams of hydrogen chloride gas is slowly bubbled beneath the surface. Stirring is continued for 10 minutes after the hydrogen chloride has been added.

To the chloromethyl stearamide (stearyl amido methyl) thus formed is added two hundred sixty-five (265) grams of octadecane nitrile (stearonitrile). The temperature is raised to 65° to 70° C. and stirring is continued for one (1) hour. At the end of this time thirty-four (34) grams of 95% paraformaldehyde is added, and then thirty-six (36) grams of hydrogen chloride gas is bubbled in slowly beneath the surface of the mixture while the temperature is maintained at 65° to 75° C. Stirring is

continued for thirty (30) minutes after the hydrogen chloride has been added, and the temperature is then lowered to about 50° C. The mixture is then placed under partial vacuum until about five hundred (500) grams of benzene has been stripped off.

To the remaining material is then slowly added ninety-five (95) grams of pyridine while maintaining the temperature at 65° to 70° C. Stirring is continued for thirty (30) minutes after the pyridine has been added. The reaction is then complete; and the product when cooled to room temperature is a firm homogeneous paste having a slight odor of pyridine. It is readily dispersible in water at 60° C.

In this example, the preparation of the stearyl amido methyl chloride in situ forms no part of the present invention.

This product was tested for its capacity to impart water-repellency to cotton fabric as more fully described under Water-Repellency Test A just below Example IV.

EXAMPLE II

Two hundred eighty-five (285) grams of methylol palmitamide is mixed with five hundred (500) grams of ethyl acetate. To this slurry is added slowly, while agitating vigorously, forty-six (46) grams of phosphorus trichloride. Stirring is continued for ten (10) minutes after the phosphorus trichloride has been added, while maintaining the temperature of 50° to 60° C. throughout the entire reaction period.

To the chloromethyl palmitamide (palmityl amido methyl chloride) thus formed is added two hundred sixty-five (265) grams of octadecane nitrile and the reaction is allowed to proceed for 30 minutes at 70° to 75° C. while stirring is continued. At the end of this time the temperature is lowered to 60° to 65° C., thirty-four (34) grams of 95% paraformaldehyde is added and then forty-six (46) grams of phosphorus trichloride is run in slowly. After the phosphorus trichloride has been added, stirring is continued for 15 minutes while maintaining the temperature at 65° to 70° C. The mixture is then cooled to 50° to 55° C. and is subjected to partial vacuum until about four hundred fifty (450) grams of ethyl acetate has been stripped off.

To the remaining material is then added slowly ninety-eight (98) grams of beta picoline while maintaining the temperature at 60° to 75° C. Stirring is continued for 30 minutes after the picoline has been added. The resulting product is a tan colored paste at room temperature and is readily dispersible in water at 50° C.

EXAMPLE III

Three hundred thirteen (313) grams of methylol stearamide and one hundred eighty-one (181) grams of dodecane nitrile are mixed with two hundred (200) grams of benzene and two hundred (200) grams of isoheptane. While stirring vigorously and maintaining the temperature at 50° to 60° C. forty (40) grams of hydrogen chloride gas is bubbled in slowly beneath the surface of the mixture. The temperature thereof is then raised to 70° to 75° C. and reaction is completed within about 30 minutes.

After that step thirty-four (34) grams of 95% paraformaldehyde is added, and thirty-six (36) grams of hydrogen chloride gas is again bubbled in beneath the surface. Stirring is continued for 15 minutes. The mixture is then cooled to 50° to 60° C. and subjected to partial vacuum until about two hundred (200) grams of mixed solvent has been removed.

To the remaining material is then added slowly one hundred thirty (130) grams of 2-methyl-5-ethyl pyridine while maintaining the temperature at 60° to 75° C. Stirring is continued for 30 minutes and the mixture is then cooled to room temperature. The resulting product is a mixture of a quaternary ammonium compound and solvent. It is a smooth tan colored paste which is readily dispersible in water at 45° C.

EXAMPLE IV

Two hundred eighty-three (283) grams of stearamide and two hundred sixty-five (265) grams of octadecane nitrile are mixed with two hundred (200) grams of dioxane and two hundred (200) grams of benzene. The mixture is heated to about 70° C. and stirred until the amide and nitrile are dissolved. The solution is then cooled to about 55° C. and thirty-two (32) grams of 95% paraformaldehyde is added. While stirring vigorously forty-six (46) grams of phosphorus trichloride is slowly added and the temperature allowed to rise to 65° to 75° C. Stirring is continued for 15 minutes after the phosphorus trichloride has been added. The mixture is cooled to about 55° C., then thirty-two (32) grams of 95% paraformaldehyde and forty-six (46) grams of phosphorus trichloride are added and the reaction is continued at 65° to 75° C. for 30 minutes. The mixture is then cooled to 50° to 60° C. and approximately two hundred (200) grams of mixed solvent is distilled off under reduced pressure.

To the remaining material is then slowly added ninety-five (95) grams of pyridine while maintaining the temperature at 65° to 75° C. Stirring is continued for 15 minutes after the pyridine has been added and the mixture is then cooled to room temperature. The final product is a firm homogeneous paste which is readily dispersible in warm water.

This last product (Example IV) was tested for its water-repellency to viscose rayon, i. e., regenerated cellulose rayon, as more fully described in test B below.

The water-repellency tests

A. Forty (40) grams of the product prepared in Example I is dispersed by stirring into two hundred (200) cubic centimeters of water at 65° C. When completely dispersed one hundred fifty-five (155) cubic centimeters of cold water and five (5) grams of sodium acetate are added. A piece of cotton poplin is immersed in this treating bath until thoroughly wetted and then the excess liquid is removed by passing between a pair of squeeze rolls. The cloth is then placed in an oven held at 110° C. until just barely dry. It is then placed in a second oven held at a temperature of 155° to 160° C. for a period of 90 seconds. After this heat treatment the cloth is immersed for a few seconds in an aqueous solution containing 0.1% sodium carbonate and 0.1% sodium lauryl sulfate, at about 40° C., to neutralize residual acidity. It is then rinsed thoroughly in warm water and dried at 110° to 120° C. After being permitted to regain its normal moisture content the treated cloth is found to possess a high degree of water repellency.

The product of each of Examples II, III and IV was similarly tested and the same condition of the cotton poplin was observed after this test had been performed.

B. Thirty (30) grams of the product prepared in Example IV is dispersed by stirring into one hundred fifty (150) cubic centimeters of water at 55° C. When completely dispersed, two hundred seventeen (217) cubic centimeters of cold water and three (3) grams of sodium acetate are added. A piece of spun viscose rayon challis is immersed in this treating bath until thoroughly wetted and then the excess liquid is removed by squeezing. The cloth is then placed in a ventilated oven held at 150° to 160° C. for 4 minutes. It is then immersed for a few seconds in an aqueous solution containing 0.1% sodium carbonate and 0.5% isopropanol, after which it is rinsed thoroughly in warm water and dried at about 110° C. After regaining its normal moisture content the treated cloth is found to possess a high degree of water repellency.

The product of Examples II, III and IV was similarly tested and the same condition of the spun viscose rayon was observed after this second test had been performed.

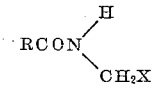
It will be understood that many variations and modifications may be made in the details of procedure above set

forth without departing from the spirit of this invention.

In the claims the term "formaldehyde" shall be understood as covering gaseous formaldehyde, paraformaldehyde, or any other compounds which yield formaldehyde in chemical reactions. Where the term "hydrogen chloride" appears in the claims, it will be understood as covering it per se or any other compounds which yield hydrogen chloride in chemical reactions under conditions used in the present process. We do not claim as part of our invention the preparation of either the methylol acyl amide or the halomethyl acyl amides.

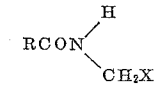
We claim as our invention:

1. The process of producing quaternary ammonium compounds, which are water-dispersible per se, which comprises reacting a chloromethyl acid amide of the general formula



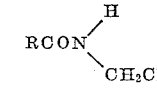
wherein R designates a saturated aliphatic hydrocarbon radical of 9 to 21 carbon atoms inclusive and X stands for chlorine, with a straight chain alkyl nitrile containing 10 to 22 carbon atoms inclusive, reacting this intermediate product with formaldehyde and hydrogen chloride to form a second intermediate product, the temperature of reaction in forming these aforesaid intermediate products being substantially 60° to 75° C. inclusive, and thereafter reacting this latter product at temperatures of substantially 50° to 75° C. inclusive with a tertiary amine from the group consisting of pyridine and the C-alkyl substituted pyridines, the alkyl group of which does not contain more than 3 carbon atoms.

2. The process of producing quaternary ammonium compounds which are water-dispersible per se, which comprises reacting without thermal decomposition 1 mol equivalent of a chloromethyl acid amide of the general formula



wherein R designates a saturated aliphatic hydrocarbon radical of 9 to 21 carbon atoms inclusive and X stands for chlorine, with approximately 0.5–1.5 mols of a straight chain alkyl nitrile containing 10 to 22 carbon atoms, inclusive, then reacting this intermediate product with approximately 0.5–1.5 mol equivalents of formaldehyde and approximately 0.5–1.5 mol equivalents of hydrogen chloride to form a second intermediate product, the temperature of reaction in forming these aforesaid intermediate products being substantially 60° to 75° C. inclusive, and thereafter reacting this latter product at temperatures of substantially 50° to 75° C. inclusive with 0.5–1.5 mol equivalents of a tertiary amine from the group consisting of pyridine and the C-alkyl substituted pyridines, the alkyl group of which does not contain more than 3 carbon atoms.

3. The process of producing quaternary ammonium compounds which are water-dispersible per se, which comprises reacting a chloromethyl acid amide of the general formula



wherein R designates a saturated aliphatic hydrocarbon radical of 15 to 21 carbon atoms inclusive with a straight chain alkyl nitrile containing 16 to 22 carbon atoms inclusive, then reacting this intermediate product with formaldehyde and hydrogen chloride to form a second intermediate, the temperature of reaction in forming these aforesaid intermediate products being substantially 60° to 75° C. inclusive, and thereafter reacting this latter product at temperature of substantially 50° to 75° C. inclusive with a tertiary amine from the group consisting of pyridine and C-alkyl substituted pyridines, the alkyl group of which does not contain more than 3 carbon atoms.

4. The process set out in claim 1, wherein the first two steps at least carried out in the presence of an organic solvent for the reactants.

5. The process of producing a quaternary ammonium compound, which comprises reacting stearyl amidomethyl chloride with stearonitrile, then reacting this intermediate product with paraformaldehyde and hydrogen chloride gas, the reactions being carried out in the presence of a volatile organic solvent for the reactants while heating to substantially 60° to 75° C. inclusive and with good agitation, removing the major portion of the solvent and thereafter thoroughly mixing pyridine with the second intermediate product and reacting therewith at temperatures of substantially 50° to 75° inclusive until the reaction is substantially complete and a mass which is pasty on cooling is secured.

6. The process of producing a quaternary ammonium compound, which comprises reacting palmityl amidomethyl chloride with stearonitrile, then reacting this intermediate product with paraformaldehyde and hydrogen chloride gas, the reactions being carried out in the presence of a volatile organic solvent for the reactants while heating to substantially 60° to 75° C. inclusive and with good agitation, removing the major portion of the solvent and thereafter thoroughly mixing picoline with the second intermediate product and reacting therewith at temperatures of substantially 50° to 75° C. inclusive until the reaction is substantially complete and a mass which is pasty on cooling is secured.

7. The process of producing a quaternary ammonium compound, which comprises reacting methylol stearamide with dodecane nitrile and hydrogen chloride, then adding paraformaldehyde and hydrogen chloride gas to this intermediate product, the aforesaid reactions being carried out in the presence of a volatile organic solvent for the reactants while heating to substantially 60° to 75° C. inclusive, and with good agitation, removing part of the solvent and thereafter mixing 2-methyl-5-ethyl pyridine with this second intermediate product and reacting therewith at temperatures of substantially 50° to 75° C. inclusive until the reaction is substantially complete and a mass which is pasty on cooling is secured.

8. The process of producing a quaternary ammonium compound, which comprises reacting brought about by heating to 60° to 75° C. inclusive stearamide, octadecane nitrile and paraformaldehyde in the presence of a phosphorus halide under good agitation while the organic reactants are dissolved in a volatile organic solvent therefor, then removing much of the solvent, and adding and reacting pyridine with this product at temperatures of substantially 50° to 75° C. inclusive while continuing to stir and until a firm homogeneous paste product upon cooling is secured.

9. The produce secured by the process set out in claim 1 supra.

10. The product secured by the process set out in claim 2 supra.

11. The product secured by the process set out in claim 3 supra.

12. The product secured by the process set out in claim 4 supra.

13. The product secured by the process set out in claim 5 supra.

14. The product secured by the process set out in claim 6 supra.

15. The product secured by the process set out in claim 7 supra.

16. The product secured by the process set out in claim 8 supra.

17. The process of producing quaternary ammonium compounds which comprises reacting a primary unsub-

stituted saturated aliphatic organic acid amide, whose acyl radical has 9 to 21 carbon atoms inclusive, with a straight chain alkyl nitrile containing 10 to 22 carbon atoms inclusive, and formaldehyde in the presence of a volatile organic solvent and hydrogen chloride while heating to substantially 60° to 75° C. inclusive, then removing much of the organic solvent, thereafter reacting this product at temperatures of substantially 50° to 75° C. inclusive, with a member of the group consisting of pyridine and C-alkyl substituted pyridines, the alkyl group of which does not contain more than 3 carbon atoms.

18. The product secured by the process set out in claim 17 supra.

19. The process of producing a quaternary ammonium compound, which comprises reacting a methylol aliphatic acyl amide, the acyl group of which is saturated and has 10 to 22 carbon atoms inclusive, with a straight chain alkyl nitrile containing 10 to 22 carbon atoms inclusive,

and hydrogen chloride gas, then reacting formaldehyde and hydrogen chloride gas with this intermediate product, the aforesaid steps being carried out at temperatures of substantially 60° to 75° C. inclusive in the presence of a volatile organic solvent for the organic reactants and with good agitation, removing part of the organic solvent and thereafter adding a member of the group consisting of pyridine and C-alkyl substituted pyridines and reacting this with this second intermediate product at temperatures of substantially 50° to 75° C. inclusive to produce the quarternary ammonium compound.

20. The product secured by the process set out in claim 19 supra.

References Cited in the file of this patent

UNITED STATES PATENTS

2,510,007	Zerner	May 30, 1950
2,654,758	Papa	Oct. 6, 1953