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3,123,641

CATION-ACTIVE SURFACE ACTIVE TRILOWER-ALKYLPOLYPROPYLENEOXY QUATERNARY AMMONIUM COMPOUNDS

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9 Claims. (Cl. 260-567.6)

My invention relates to the production of novel cation-active surface active chemical compounds which are useful for bactericidal, germicidal, antiseptic, algaecidal, fungicidal, textile softening, corrosion inhibition, antistatic, emulsifying, foam modifying, ore beneficiation, and various other purposes for which certain heretofore known cationic surface active agents have been employed or suggested for use.

The specific cation-active surface active agents of my present invention are of the quaternary ammonium type and they are characterized by particularly advantageous properties, simplicity of production and markedly low cost. Certain of them, for instance, those derived from low molecular weight alkyl secondary amines and cyclic amines, can be represented by the formula

(I)



wherein R is an alkyl radical containing from 1 to 3 and more particularly from 1 to 2 carbon atoms, R₁ is an alkyl radical containing from 1 to 3 carbon atoms, or



may be a radical of a cyclic amine containing not more than 6 carbon atoms, R₂ is a polyoxyalkylene radical containing at least 18 carbon atoms and derived from an α-epoxide containing at least 3 carbon atoms, especially propyleneoxide. In those cases where R₂ is derived from propyleneoxide, there will be at least 6 of such propyleneoxide groups in R₂ and, more particularly, it is desired that R₂ contain from 8 to 30 oxypropylene groups. Where R₂ is derived from α-epoxides containing more than 3 carbon atoms, a lesser number of such oxyalkylene groups can be present, subject to the limitation of R₂ containing at least 18 atoms. R₃ is a lower alkyl or alkenyl radical containing from 1 to 4 carbon atoms, and A is an anion. It is important, for the achievements of the results to which my invention is directed, that the R radical, where it is alkyl, contain not more than 3 carbon atoms and especially that it contain from 1 to 2 carbon atoms and, hence, that it be methyl or ethyl. Likewise, except where

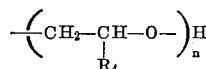


is derived from a cyclic amine as hereafter pointed out, it is important that the R₁ radical does not contain over 3 carbon atoms, and, in any event, the maximum number of carbon atoms in R and R₁ does not exceed a total of more than 6, and more particularly does not exceed a total of 4 or 5. While R₂ may contain a minimum of 6 of the aforesaid oxyalkylene, particularly oxypropylene, groups, and may contain 40, 50 or 60 or even 100 or more of said groups, it is especially advantageous that the minimum number of said groups be 8 and the preferred range is 8 to 25 or 30 of said groups. The term "α-epoxide," as used herein, is intended to cover those compounds, of the type here involved, where there are two vicinal carbon

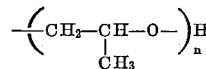
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atoms connected to one oxygen atom independently of the position of such group in the molecule.

The terms "polyoxyalkylene," "polyoxypropylene" and "polyoxyethylene," to the extent that they are used herein or in the claims, are employed in their usual or conventional manner. Thus, in the case of the polyoxyalkylene radical derived from α-epoxides containing from 3 to 6 carbon atoms, to wit, R₂ in the above formula, the same may be represented by



where R₄ is an alkyl radical containing from 1 to 4 carbon atoms, and, as indicated above, since R₂ contains a minimum of 18 carbon atoms, n is at least 6 in the case of the polyoxyalkylene radical being polyoxypropylene and n is at least 3 in the case of the polyoxyalkylene radical being polyoxyethylene. As indicated above, n is usually up to 60 or more. Where the polyoxyalkylene radical is polyoxypropylene, it may be represented by the formula



n advantageously being from 6 to 30.

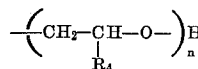
The foregoing illustrate the polyoxyalkylene and polyoxypropylene radicals where the vicinal carbons are terminal. In the broader, but less preferred, embodiments of my invention, however, the vicinal carbons need not be terminal. A typical illustration of the latter is a polyoxyalkylene radical derived from 2,3-butylene oxide.

At least most of those of the particularly preferred quaternary ammonium compounds of my invention which are derived from lower monoalkyl amines can be represented by the formula

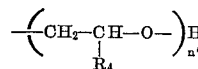
(II)



where R is an alkyl radical containing from 1 to 3, and especially from 1 to 2, carbon atoms, R₁ is



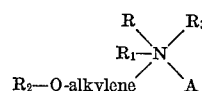
and R₂ is



where R₄ is an alkyl radical containing from 1 to 4 carbon atoms, n and n' are numbers the sum of which does not exceed 60, the total number of carbon atoms in R₁ and R₂ being at least 18; R₃ is a hydrocarbon radical selected from the group consisting of alkyl and alkenyl radicals containing not more than 4 carbon atoms; and A is an anion.

Various of those particularly preferred quaternary ammonium compounds of my invention which are derived from lower dialkyl monoalkanolamines can be represented by the formula

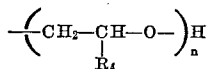
(III)



where R is an alkyl radical containing from 1 to 3, and especially from 1 to 2, carbon atoms; R₁ is an alkyl

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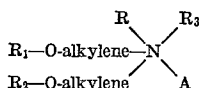
radical containing from 1 to 3 carbon atoms; alkylene contains not more than 3 carbon atoms, R_2-O is



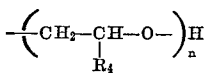
where R_4 is an alkyl radical containing from 1 to 4 carbon atoms, n is a number up to and including 60, the total number of carbon atoms in R_2 being at least 18; R_3 is a hydrocarbon radical selected from the group consisting of alkyl and alkenyl radicals containing not more than 4 carbon atoms; and A is an anion.

Again, various of those of the particularly preferred quaternary ammonium compounds of my invention which are derived from lower monoalkyl dialkanolamines can be represented by the formula

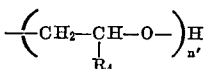
(IV)



where R is an alkyl radical containing from 1 to 3, and especially from 1 to 2, carbon atoms, alkylene contains not more than 3 carbon atoms, R_1-O is



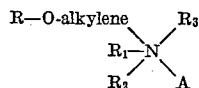
and R_2-O is



where R_4 is an alkyl radical containing from 1 to 4 carbon atoms, n and n' are numbers the sum of which does not exceed 60, the total number of carbon atoms in R_1 and R_2 being at least 18; R_3 is a hydrocarbon radical selected from the group consisting of alkyl and alkenyl radicals containing not more than 4 carbon atoms; and A is an anion.

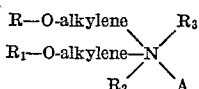
Still others of the quaternary ammonium compounds of my invention can be represented by the formulae

(V)

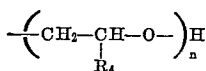


and

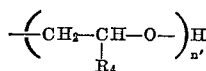
(VI)



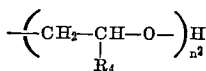
where alkylene contains not more than 3 carbon atoms, $R-O$ is



R_2-O is



and R_2-O is



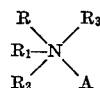
in which R_4 is an alkyl radical containing from 1 to 4 carbon atoms, n , n' and n^2 are numbers the sum of which does not exceed 60, the total number of carbon atoms in R , R_1 and R_2 being at least 18; R_3 is a hydrocarbon radical selected from the group consisting of alkyl and alkenyl radicals containing not more than 4 carbon atoms; and A is an anion.

Most of the quaternary ammonium compounds of the present invention fall into the category of or correspond to reaction products of quaternizing esters having a hydrocarbon alkyl or alkenyl radical containing not more than

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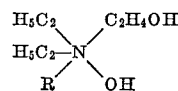
4 carbon atoms, and an anion, with adducts of (a) aliphatic amines or cyclic amines containing a reactive hydrogen and a total of not more than 6 carbon atoms and in which no single aliphatic radical contains more than 3 carbon atoms, with (b) and α -epoxide containing at least 3 and advantageously from 3 to 8 carbon atoms, the molal ratio of (a) to (b) being 1 of (a) to at least 6 of (b) when said α -epoxide contains 3 carbon atoms and ranging down to at least 2.5 of (b) when said α -epoxide contains 8 carbon atoms.

Still others of the quaternary ammonium compounds of my invention can be represented by the formula

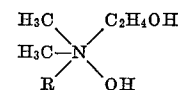


where R is an alkyl radical containing from 1 to 3 carbon atoms, R_1 is an alkyl radical containing from 1 to 3 carbon atoms or is the same as R_3 , R_2 is a polyoxyalkylene radical containing at least 18 carbon atoms and derived from an α -epoxide containing from 3 to 8 carbon atoms, R_3 is a hydroxy-alkyl radical containing from 2 to 4 carbon atoms, and A is hydroxyl or other anion. Such compounds are readily produced by reacting the adducts, such as those described hereafter in Examples a to v, with ethyleneoxide in an aqueous medium in accordance with known quaternizing techniques. Typical of such compounds are those corresponding to the formulae

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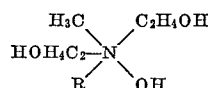


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and

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where R is a polyoxypropylene or polyoxybutylene radical containing say from 6 to 30 oxypropylene or oxybutylene groups.

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While, as indicated, many of the novel and useful cation-active surface active agents of my invention can be represented by the above formulae, they may, in general, best be described in the form of reaction products of identified ingredients, as hereafter pointed out, since the active constituent or constituents thereof need not be separated out and recovered as such, the reaction products being usable as such for many purposes without the necessity for resorting to concentration, purification or recovery steps. Thus, for example, although this will be pointed out and described in more detail hereafter, quaternary ammonium compounds which are particularly useful and which may simply and inexpensively be produced are reaction products of quaternizing ester in the form of alkyl and alkenyl halides, sulfates and sulfonates containing from 1 to 4 carbon atoms exemplified by methyl chloride, allyl chloride, methyl chloracetate, and 1 and 2 carbon atom alkyl sulfates, with adducts of (a) alkyl amines having a reactive hydrogen and containing from 1 to 4 carbon atoms, such as dimethylamine and diethylamine, with (b) propyleneoxide or butyleneoxides, said adducts containing 1 mol of (a) to from 6, and preferably from 8, to 30 mols of (b). Purification steps can, if desired, be employed as, for instance, separating unreacted tertiary amines by distillation, etc.

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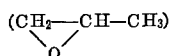
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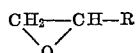
It is important to note that it is critical to my present invention that the oxyalkylene groups be derived from an α -epoxide containing at least 3 carbon atoms. Ethyleneoxide, for instance, is not operative in the prac-

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tice of my invention since it produces compounds whose properties are radically different from those obtained in accordance with my invention and which would be useless for the purposes for which the cation-active surface active agents of my invention are adapted and intended. The most important embodiment of the α -epoxides whose use is contemplated by my invention in the production of the cation-active surface active agents is propylene-oxide



The α -epoxides containing 4, 5, 6 or more carbon atoms can be used but, for general reasons of lack of economy or commercial availability, are not preferred. Typical of such latter α -epoxides are butylene oxides such as 1,2-butylene oxide, pentylene epoxides, hexylene epoxides, heptylene epoxides, octylene epoxides, dodecylene epoxides, and C_{16} – C_{18} olefin oxides or epoxides, and, as well, styrene epoxide. It will be seen that most of those of the α -epoxides in which the vicinal carbons are terminal can conveniently be represented by the formula



where R is an alkyl radical. In the case of propylene oxide, R is CH_3 , and in the representation of the group of propylene oxide, butylene oxides, pentylene oxide and hexylene oxide R contains from 1 to 4 carbon atoms. In the higher molecular weight alkylene oxides, R may contain as high as 16 carbon atoms.

While, as I have stated above, the use of ethyleneoxide as the sole alkylene oxide is inoperative for my purposes, small proportions thereof, not to exceed 25% and better still not to exceed 20% by weight of the propylene-oxide or other α -epoxide employed, can be utilized as hereafter described. Mixtures of two or more of the α -epoxides containing at least 3 carbons can, of course, be used as such, as well as in conjunction with ethylene-oxide, subject to the limitation that the ethyleneoxide must not exceed the aforesaid 20% to 25% of the weight of said α -epoxides.

The cation-active surface active agents of my invention are advantageously prepared by initially condensing a lower molecular weight amine, for instance, one containing at least one alkyl radical having from 1 to 3 carbon atoms and no radical containing more than 3 carbon atoms linked to the nitrogen atom of the amine, or by initially condensing a cyclic amine such as piperidine containing not more than 6 carbon atoms, with propyleneoxide or other α -epoxide containing at least 3 carbon atoms, advantageously in the presence of a conventional oxyalkylation catalyst such as sodium hydroxide or potassium hydroxide, whereby to produce an intermediate or adduct containing at least 6 oxypropylene groups. The amines used in the reaction to produce said adducts must contain a reactive hydrogen, which may be hydrogen directly attached to the nitrogen of the amine or which may be in the form of hydroxyl hydrogen, secondary alkyl amines being especially desirable, particularly dimethylamine, diethylamine and methylethylamine. The resulting adduct is then reacted with a quaternizing ester in the form of lower alkyl or alkenyl halides or lower alkyl esters of sulfuric acid or lower alkyl esters of sulfonic acids, or lower alkyl and alkenyl haloacetates, to convert said adduct into the cation-active surface active agents of my present invention. Illustrative examples of such lower molecular weight alkyl halides, haloacetates, sulfates, etc. are methyl chloride, ethyl chloride, methyl bromide, ethyl bromide, methyl iodide, allyl chloride, allyl bromide, methyl chloracetate, ethyl chloracetate, vinyl chloracetate, dimethyl sulfate, and diethyl sulfate. In order to obtain cation-active surface active agents having the required properties in accordance with my invention, the quaternizing alkyl and alkenyl esters, for instance, the halides, sulfates and sulfonates, should con-

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tain not more than 4 carbon atoms and, better still, should contain only 1 to 2 carbon atoms. The speed of quaternization in any given instance will be governed by the reactants and the reaction conditions including the temperature and solvent medium utilized.

Illustrative examples of the intermediate condensation products or adducts which are subsequently converted into the cation-active surface active agents or quaternary ammonium compounds encompassed by my present invention are the following:

(a) Condensation products of 1 mol of dimethylamine with 6 to 10 mols of propyleneoxide.

(b) Condensation products of 1 mol of dimethylamine with 20 mols of propyleneoxide.

(c) Condensation products of 1 mol of dimethylamine with a mixture of 25 mols of propyleneoxide and 5 mols of ethyleneoxide.

(d) Condensation products of 1 mol of dimethylamine with 1 to 2 mols of ethyleneoxide, reacted with 20 mols of propyleneoxide.

(e) Condensation products of 1 mol of methylamine with 30 to 40 mols of propyleneoxide.

(f) Condensation products of 1 mol of methylethylamine with 10 to 60 mols of propyleneoxide.

(g) Condensation product of 1 mol of diethylamine with 30 mols of propyleneoxide.

(h) Condensation products of 1 mol of dimethylamine with 10 to 20 mols of propyleneoxide and then with 1 mol of ethyleneoxide.

(i) Condensation products of 1 mol of methylpropylamine with 10 to 25 mols of propyleneoxide.

(j) Condensation products of 1 mol of dimethylamine with 10 to 15 mols of butyleneoxide.

(k) Condensation products of 1 mol of diethylamine with 10 to 18 mols of butyleneoxide.

(l) Condensation products of 1 mol of piperidine with 10 to 25 mols of propyleneoxide.

(m) Condensation products of 1 mol of methylpiperidine with 10 to 25 mols of propyleneoxide.

(n) Condensation products of 1 mol of pyrrolidine with 10 to 25 mols of propyleneoxide.

(o) condensation products of $\frac{1}{2}$ mol of methylamine and $\frac{1}{2}$ mol of diethylamine with 6 to 3 mols of propyleneoxide.

(p) Condensation products of $\frac{1}{3}$ mol of dimethylamine and $\frac{2}{3}$ mol of diethylamine with 10 to 25 mols of propyleneoxide.

(q) Condensation products of 1 mol of morpholine with from 6 to 30 mols of propyleneoxide.

(r) Condensation products of 1 mol of diethylethanolamine with 8 to 20 mols of propyleneoxide.

(s) Condensation products of 1 mol of diallylethanolamine with 8 to 25 mols of propyleneoxide.

(t) Condensation products of 1 mol of furfurylamine with 8 to 25 mols of propyleneoxide.

(u) Condensation products of 1 mol of 2-amino-1,3-propanediol with 8 to 25 mols of propyleneoxide.

(v) Condensation products of 1 mol of diethylamine with 6 to 25 mols of propyleneoxide.

The amines which are utilized in the production of the intermediate adducts or condensation products with the propylene oxide or other α -epoxides are particularly advantageously of aliphatic or acyclic character, although they also include cyclic secondary amines. In all cases, they contain at least one reactive hydrogen directly attached to nitrogen or in a hydroxy group. Illustrative examples of such amines are alkyl amines, e.g., methylamine, dimethylamine, ethylamine, diethylamine, methylethylamine, propylamine, dipropylamine; alkyl hydroxy amines, e.g., monomethylethylthanolamine, monoethylthanolamine, monomethyldiethanolamine, dimethylethanolamine, diethylethanolamine and diallylethanolamine; polyamines such as ethylenediamine, diethylenetriamine and triethylenetetraamine and higher polyamines in which there is at least one short chain alkyl group, as for in-

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stance, monomethyl ethylenediamine, dimethyl ethylenediamine and trimethyl ethylenediamine; N,N-dimethyl-N',N'-diisopropanol propylenediamine - 1,3; furfurylamine; cyclic secondary amines containing not more than 6 carbon atoms, typical examples of which are morpholine, piperidine, methylpiperidine, and pyrrolidine. Such amines may contain nonreactive substituents such as nitro groups, ether and thioether groups, CN groups, and the like, but, in general, best results will be obtained with the unsubstituted amines. Compatible mixtures of any two or more of said amines, in various proportions, can, of course, be utilized in the production of the intermediate condensation products.

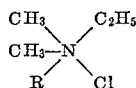
The radical A can be any negative or salt-forming radical as, for instance, halogen such as chlorine, bromine and iodine; hydroxy; sulfate, alkyl-sulfuric acid such as methyl sulfuric acid or ethyl sulfuric acid (methosulfate or ethosulfate); nitrate; phosphate; acetate; formate; carbonate; lower alkyl (1 to 4 carbon atoms) sulfonic acids; and the like. Of particular importance are those of the cation-active surface active agents of my invention in which the anion is chlorine, bromine, methyl chloroacetate, methosulfate, and ethosulfate. Various of said anionic radicals can be introduced directly into the molecules; and, in the case of others, they can be made, for instance, by substitution for halogen in the cation-active surface active agents of my present invention by known techniques as, for instance, by metathesis procedures.

The following examples are illustrative of the production of typical cation-active surface active agents in accordance with my invention. It will be understood that other reactants can be utilized, proportions modified, temperature and other conditions, and concentration or purification procedures employed, all within the guiding principles taught herein, without departing from the essential teachings disclosed herein.

Example 1

(a) 46 grams (1 mol) of dimethylamine were placed in an autoclave from which the air was displaced by nitrogen. 58 grams (1 mol) of propyleneoxide were pumped into the dimethylamine in said autoclave and reacted while maintaining the temperature below 25 degrees C. The resulting compound was heated to about 150 degrees C. and 25 mols of propyleneoxide were added to the autoclave over a period of 6 hours while maintaining a nitrogen pressure in the autoclave, said reaction being advantageously carried out in the presence of 2 to 3 grams of potassium hydroxide as a catalyst. The rate of addition of the propyleneoxide was controlled so that the pressure in the autoclave did not exceed 2 atmospheres and the temperature controlled so as not to exceed 200 degrees C. and preferably at about 150 degrees C. Upon completion of the reaction, the reaction mixture was cooled to below 100 degrees C. and flushed out with gaseous nitrogen.

(b) To 77 grams of the intermediate condensation product or adduct of part (a) hereof there were added 8 grams of methanol and 4 grams of ethyl chloride and the resulting mixture was refluxed, with stirring, for 5 hours. During this period, the temperature of reflux rose somewhat and the viscosity of the reaction mixture increased appreciably. A yield in excess of 90% of theoretical of the following product was obtained:



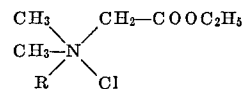
wherein R is a polyoxypropylene radical containing about 25 oxypropylene groups.

Example 2

(a) To 150 grams of the intermediate condensation product or adduct of part (a) of Example 1 there were

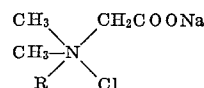
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added 13.5 grams of ethyl chloroacetate and 15 grams of methanol. The resulting mixture was heated at reflux temperature for 7 hours during which period the reflux temperature rose to about 100 degrees C. and the viscosity increased appreciably. A yield of about 94% of the following compound was obtained:



wherein R is a polyoxypropylene radical containing about 25 oxypropylene groups.

(b) To the product of part (a) of this Example 2 there were added 50 ml. of water and 1 ml. of concentrated hydrochloric acid, and heating at reflux was continued to hydrolyze the ester to the free acid. The latter was neutralized with sodium hydroxide to form the sodium salt, the final compound corresponding to the formula

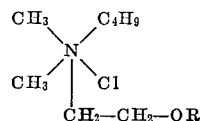


where R is the same as set forth above.

Example 3

(a) 93 grams (1 mol) of dimethylethanolamine were reacted with 1480 grams (20 mols) of propyleneoxide in the manner described in the latter part (a) of Example 1.

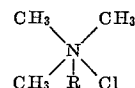
(b) 50 grams of the adduct of part (a) of this Example 3 were reacted with 5 grams of butyl chloride, in methanol, in the manner described in part (b) of Example 1. The final reaction product, in a yield of in excess of 90%, comprised the following product:



where R is a polyoxypropylene radical containing approximately 20 oxypropylene groups.

Example 4

50 grams of the adduct of part (a) of Example 1 were reacted with 3 grams of methyl chloride in the manner described in part (b) of Example 1 to produce a product having the following formula:



where R is a polyoxypropylene radical containing about 25 oxypropylene groups.

Example 5

The process was carried out as described in Example 1 except that, in part (a) thereof, in place of the final 25 mols of propyleneoxide, there was used a mixture of 22 mols of propyleneoxide and 2 mols of ethyleneoxide.

This application is a continuation-in-part of my application Serial No. 524,297, filed July 25, 1955, and now abandoned.

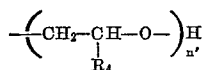
What I claim as new and desire to protect by Letters Patent of the United States is:

1. A quaternary ammonium compound having the formula

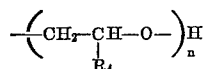


wherein R is an alkyl radical containing from 1 to 3 carbon atoms, R₁ is a member selected from the group

consisting of an alkyl radical containing from 1 to 3 carbon atoms and



and components wherein R and R₁ are joined together to form with the N atom a member of the group consisting of piperidine, methyl piperidine and pyrrolidine; R₂ is



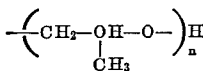
where R₄ is an alkyl radical containing from 1 to 4 carbon atoms, *n* and *n'* are numbers the sum of which does not exceed 60, the total number of carbon atoms in R₂ being at least 18; R₃ is a hydrocarbon radical selected from the group consisting of alkyl and alkenyl radicals containing from 1 to 4 carbon atoms; and A is an anion.

2. A quaternary ammonium compound in accordance with claim 1, wherein R₄ is CH₃.

3. A quaternary ammonium compound having the formula



wherein R is an alkyl radical containing from 1 to 3 carbon atoms; R₁ is an alkyl radical containing from 1 to 3 carbon atoms; R₂ is

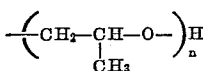


wherein *n* is a number from 6 to 60; R₃ is an alkyl radical having from 1 to 4 carbon atoms; and A is an anion.

4. A quaternary ammonium compound having the formula



wherein R is methyl; R₁ is an alkyl radical containing from 1 to 3 carbon atoms; R₂ is

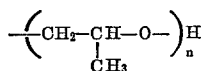


wherein *n* is a number from 6 to 30; R₃ is an alkyl radical having from 1 to 4 carbon atoms; and A is an anion.

5. A quaternary ammonium compound having the formula



wherein R and R₁ are each ethyl; R₂ is



wherein *n* is a number from 6 to 30; R₃ is an alkyl radical containing from 1 to 4 carbon atoms; and A is chlorine.

6. A quaternary ammonium compound in accordance with claim 1 wherein up to 20% by weight of R₂ is replaced by a polyoxyethylene radical.

7. A quaternary ammonium compound having the formula



wherein R is an alkyl radical containing from 1 to 3 carbon atoms, R₁ is an alkyl radical containing from 1 to 3 carbon atoms, and compounds wherein R and R₁ are joined together to form with the N atom a member of the group consisting of piperidine, methyl piperidine and pyrrolidine; R₂ is a polyoxypropylene-polyoxyethylene radical in which the number of oxypropylene groups is from 6 to 60 and in which the ethylene oxide constitutes not more than 20% by weight of said radical; R₃ is a hydrocarbon radical selected from the group consisting of alkyl and alkenyl radicals containing from 1 to 4 carbon atoms; and A is an anion.

8. A quaternary ammonium compound having the formula



where R is an alkyl radical containing from 1 to 3 carbon atoms, R₁ is an alkyl radical containing from 1 to 3 carbon atoms, and compounds wherein R and R₁ are joined together to form with the N atom a cyclic amine radical containing not more than 6 carbon atoms, with the proviso that the total number of carbon atoms in R and R₁ does not exceed 6; R₂ is a polyoxyalkylene radical containing at least 18 carbon atoms and derived from an α-epoxide containing from 3 to 6 carbon atoms, said α-epoxide consisting of carbon, hydrogen and oxygen, and the total number of oxyalkylene groups in said polyoxyalkylene radical not exceeding 60, R₃ is a hydrocarbon radical selected from the group consisting of alkyl and alkenyl radicals containing from 1 to 4 carbon atoms; and A is an anion.

9. A quaternary ammonium compound in accordance with claim 8, wherein up to 25% by weight of R₂ is replaced by a polyoxyethylene radical.

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