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2,921,885

ANTIBACTERIAL COMPOSITION

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This invention relates to new and improved compositions for the control of bacteria. More particularly it relates to compositions, e.g., mouthwashes, which comprise an antibacterial quaternary ammonium compound and an amide compound synergist.

Quaternary ammonium compounds have been used for years as antibacterial agents. They have been sold as solutions, in crystal form and mixed with fillers. Quaternary ammonium compounds have found use in antiseptics, deodorants, cleaning compositions and cosmetics and certain of them have been suggested for addition to mouth rinses.

Condensation products of fatty acylating materials and short chain amino acids have been recommended for incorporation in cosmetic preparations. At the present time substantially pure sodium N-lauroyl sarcosine is being employed in toothpaste because of its anti-caries activity. The sodium salts of N-substituted fatty acyl amides of short-chain amino acids have also been suggested as additives to oral preparations such as mouth washes.

Although both the quaternary ammonium compounds and the above-described amide compounds have been suggested singly as mouth rinse ingredients, so far as is known they have never heretofore been combined in a germicidal composition or mouthwash. The very good reason for this is that it is a recognized fact that most anionic active agents, if added to quaternary ammonium compounds, react with the quaternary compound and decrease the antibacterial action thereof. In some cases where the antibacterial action of quaternaries is undesirable it is intentionally destroyed completely by treatment with soap or other anionic material.

In accordance with the present invention an antibacterial composition comprises an antibacterial quaternary ammonium compound and a substantially saturated aliphatic acyl amide of a saturated monoaminocarboxylic acid compound having 2 to 6 carbon atoms, the aliphatic acyl group thereof having 10 to 18 carbon atoms, the amide being present in amount sufficient to improve the antibacterial action of the composition when it is dissolved in water.

It has been found that aqueous solutions of the above composition are more effective antibacterial agents than solutions of the quaternary compound alone. Thus, the antibacterial potency of a quaternary compound is increased noticeably by addition of the described amide compound, which is a highly surprising result. This increased antibacterial action is obtained at no additional cost when quaternary ammonium compound is added to compositions already containing such an amide compound, e.g., mouth washes, dentifrices and so forth.

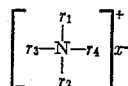
The addition to the composition of amide compound and quaternary of a water soluble nonionic surface active compound containing as a hydrophilic group a polyoxyethylene chain of 20-700 carbon atoms, increases the antibacterial power further.

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The antibacterial quaternary ammonium compound which is one of the essential elements of the invented composition is generally of the formula



where r_1 is a hydrophobic radical selected from the group consisting of aliphatic, unsaturated aliphatic, cycloaliphatic, aryl, aliphaticaryl and arylaliphatic radicals, of 10 to 26 carbon atoms, r_2 , r_3 and r_4 are each hydrocarbon radicals of 1 to 10 carbon atoms, the sum of the carbon atoms in the three radicals being from 3 to 12, and x is a salt-forming radical. The above formula may also be written as



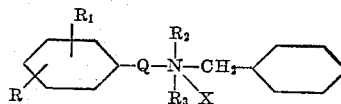
to better indicate the salt character of the quaternary compound.

The hydrophobic radical of the quaternary ammonium compound may be branched or normal chain alkyl phenoxy alkoxy alkyl, branched or long chain alkyl cresoxy alkoxy alkyl, long chain alkoxy aryl, branched or long chain alkyl phenoxy alkyl, long chain alkylaryl, halogen substituted long chain alkylaryl, arylalkyl, long chain alkyl, long chain alkenyl and cycloalkyl.

The remaining hydrocarbon radicals on the quaternary nitrogen are usually low molecular weight alkyl, preferably methyl or ethyl, or aryl, preferably phenyl, or arylalkyl, preferably benzyl, but may also include heterocycle-forming type substituents, as in pyridine or piperidine.

The salt forming radical x is not generally considered to be critical because the cationic portion of the molecule is the active antibacterial component. It may be any anionic radical capable of forming a water soluble salt. Chloride and bromide are preferred but halides generally, sulfates, phosphates, and other salt-forming ions are also satisfactory.

Among the quaternaries that may be incorporated in the invented antibacterial composition are those containing an ether linkage connecting the hydrophobic portion and the nitrogen atom. Of these, preferred are compounds of the formula



where R is an alkyl group of 6 to 12 carbon atoms, R_1 is selected from the group consisting of hydrogen and alkyl groups of 1 to 3 carbon atoms, R_2 and R_3 are alkyl groups of 1 to 3 carbon atoms, Q is an alkoxy intermediate linkage between the hydrophobe group and the quaternary nitrogen, having an oxygen atom adjacent the benzene ring of the hydrophobe and containing 2 to 6 carbon atoms, and X is a halide selected from the group consisting of bromine and chlorine. A particularly preferred quaternary of this formula is diisobutyl cresoxy ethoxy ethyl dimethyl benzyl ammonium chloride. Also preferred is diisobutyl phenoxy ethoxy ethyl dimethyl benzyl ammonium chloride. These two compounds are made and sold by Rohm and Haas Company, Philadelphia, Pa., as crystalline monohydrates under the trademark Hyamine, as Hyamine 10-X and Hyamine 1622 respectively.

Other examples of quaternaries that may be used in the present compositions are alkyl ammonium halides such as cetyl trimethyl ammonium bromide and alkylaryl

ammonium halides such as the long chain alkyl dimethyl benzyl ammonium chlorides. Also suitable are quaternaries having ester or amide intermediate linkages, e.g., N (lauroyl colamino formylmethyl) pyridinium chloride. Other contemplated quaternary ammonium salts are N-alkylpyridinium and N-alkylmorpholinium compounds, e.g., N-cetyl pyridinium bromide and quaternaries containing a substituted aromatic nucleus, e.g., chlorinated dodecylbenzyl trimethyl ammonium chloride.

The other essential component of the present antibacterial composition is a substantially saturated aliphatic acyl amide of a saturated aliphatic monoamino-carboxylic acid compound having 2 to 6 carbon atoms, the aliphatic acyl group thereof having 10 to 18 carbon atoms, which acts to improve the antibacterial action of the quaternary ammonium compound when the antibacterial composition is dissolved in an aqueous medium.

Among the suitable saturated aliphatic monoamino carboxylic acid compounds from which the higher amide may be derived are glycine, sarcosine, alanine, valine, 3-aminopropanoic acid and the like. Generally those amino acids will be used which have 2 to 5 carbon atoms, preferably the sarcosine and glycine homologues. Of the amino acids so encompassed sarcosine is most preferred.

The substantially saturated aliphatic acyl amides of this invention are those having about 10 to 18, preferably 12 to 16 carbon atoms in a fatty acyl radical, e.g., the lauroyl, myristoyl, palmitoyl derivatives.

The amide compound which comprises an essential element of the present antibacterial composition may be made in the known manner by condensing a higher fatty acylating substance, e.g., lauroyl chloride, with a salt of the amino acid in the presence of an alkaline acceptor for liberated hydrogen halide.

Generally, and preferably, the amide compounds are employed in the form of water-soluble salts. Such salts are the alkali metal, e.g., sodium, potassium, etc., ammonium, amine and alkylolamine salts. The terms "amide compound" and the term "amide of a . . . mono-aminocarboxylic acid compound . . ." includes amides of aminocarboxylic acids, and their water-soluble salts. Among the "amide compounds" are sodium N-lauroyl sarcosine, sodium N-palmitoyl sarcosine, lauroyl sarcosine, sodium N-myristoyl glycine, potassium N-lauroyl sarcosine and the like.

Care should be taken in the manufacture of the amide compounds so that a product may be obtained containing little fatty acid material. "Fatty acid material" includes higher fatty acids (of 10 or more carbon atoms) and soaps thereof. In mouth washes it is desirable that the amount of fatty acid material present be kept below 15% based on the weight of amide compound (active ingredient). Preferably the fatty acid material content is less than 10% on the same basis and it is best to keep it below 5%. Past experience has shown that if more than 15% of fatty acid material is present (based on active ingredient) the anti-carial properties of an oral preparation containing the amide compound will be decreased markedly, and it is well known that soap decreases the antibacterial effect of quaternary ammonium compounds.

On a weight basis the proportion of quaternary ammonium compound to amide compound should be between 1:25 and 1:2, preferably about 1:7, to have the amide compound act to improve the antibacterial activity of the quaternary.

The antibacterial composition may be made by mixing dry quaternary ammonium compounds or their dry hydrates with dry amide compounds, so obtaining solid compositions, usually particulate. Such dry or substantially dry compositions may later be diluted with solvents before use. Alternatively solutions (including partial dispersion) may be made initially. The particular concentrations of such solutions will depend to a great degree

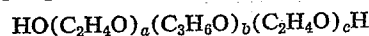
on (1) the use intended for the solution, (2) the particular quaternary ammonium compound, its antibacterial activity and limiting concentration for safe use for the purpose intended, and (3) the amide compound, its synergistic effect and other desired functions. In the case of mouth rinse compositions, for example, in general about 0.01 to 0.1% antibacterial quaternary ammonium compound and .05 to 0.5% of amide compound are employed in an aqueous or aqueous alcoholic medium, in proportion between 1:25 and 1:2.

In making solutions of the present composition, either during the manufacturing operation or at the time of use, various solvents or carrier liquids may be employed. Water (preferably deionized or distilled), lower molecular weight saturated aliphatic alcohols, water soluble lower polyhydric alcohols and mixtures thereof may be used. Included in the group from which suitable solvents and mixtures thereof may be selected are ethanol, glycerine, sorbitol, propylene glycol and the like. Other suitable solvents may be employed, even if toxic, when their poisonous nature is not objectionable, e.g., methanol, isopropanol in disinfectants for inanimate objects, antiseptics for external use only. Suitable known coloring, flavoring, perfuming and thickening agents and other usual adjuvants may be added to the dry solid and liquid compositions and to their solutions.

The concentration of active composition ingredients, the type and amount of solvent present and the presence of appropriate adjuvant materials are governed to a large extent by the end use of the composition. In a mouth rinse, for example, it is preferable that water constitute most of the solvent, from 50 to 95%, but ethanol may be used too in amounts between 5 and 25%, because of its antibacterial action, solvent properties (often needed to dissolve mouth rinse ingredients) and taste. Up to about 25 percent of other solvent may be included in mouth rinses.

It has been found that the addition of certain nonionic surface active agents to the invented antibacterial compositions increases their antibacterial action. Thus a water-soluble nonionic surface active compound containing, as a hydrophilic portion of its molecule, a polyoxyethylene of 10 to 300 oxyethylene groups and as a lipophilic portion a saturated aliphatic radical selected from the group consisting of alcohol radicals of 8 to 18 carbon atoms and polyoxypropylenes of molecular weight between 1000 and 2000 is a suitable additive. The amount of nonionic used should be from 1 to 5 times the combined weight of amide compound and quaternary.

In this group are the block copolymers of the formula



in which the ethylene oxide constitutes from 20 to 90% of the molecule by weight. These compounds are sold under the trade name "Pluronic" by Wyandotte Chemicals Corp., Wyandotte, Michigan. Pluronic F-68, a copolymer of the formula given in which the propylene oxide group is of a molecular weight of 1500 and 1800, and in which polymerized ethylene oxide is 80-90% by weight, is a preferred nonionic component because of its bland taste and solid form, in addition to low toxicity and excellent surface activity. In mouth rinses from 0.1 to 1.0% by weight should be employed for best results.

Of the class of nonionic compounds containing alcohol radicals as hydrophobes a preferred member is a polyoxyethylene tridecyl alcohol containing about 12 moles ethylene oxide. This product is marketed as Renex 30 by the Atlas Powder Company, Wilmington, Delaware.

Some of the nonionic products within the above descriptions are bitter to the taste. It is preferred to exclude such compounds from oral preparations or else to cover their taste with flavoring ingredients.

Solutions of the invented compositions have a general applicability, for antibacterial purposes, in various antibacterial products and operations. As examples the dis-

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closed compositions may be used as surface disinfectants for human skin, medical instruments, wall or floor surfaces, clothing and the like, as well as in mouth rinses.

In making the present antibacterial compositions suitable mixtures of quaternary ammonium compounds, amide compounds and specified nonionic compounds may be employed rather than compositions made from individual components.

The following examples are illustrative of the present invention but the invention is not limited to them. All amounts or percentages throughout this specification and in the claims are by weight unless otherwise indicated.

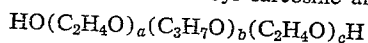
Example I

A solution of .03% diisobutyl cresoxy ethoxy ethyl dimethyl benzyl ammonium chloride and 0.2% sodium N-lauroyl sarcosine was made in distilled water. Standard disc-halo tests (described below) of this solution against *Micrococcus pyogenes*, var. *aureus* resulted in a halo 19.9 millimeters in diameter. The annular width of the halo was 3.6 mm. (the disc being 12.7 mm. in diameter). Similar tests of the named quaternary compound alone at concentrations of 0.03 and 0.06% gave halos of 16.2 and 16.6 mm. diameter and 1.75 and 1.95 mm. annular widths respectively. Solutions of 0.2 and 0.4% sodium N-lauroyl sarcosine exhibited no halos. There was evidence of slight inhibition of bacterial growth by the sarcosine derivative but there was no bacteriostasis. The above results show that the invented composition prevented bacterial growth over an area 135% greater than that in which .03% of quaternary compound alone prevented growth. Doubling the amount of quaternary to 0.06% resulted in only a 15% increase in area of bacteriostasis over that of the 0.03% solution.

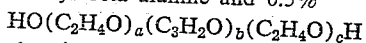
The disc halo tests described here are well known. Agar plates inoculated with *Micrococcus pyogenes*, var. *aureus* were prepared according to the method of section 141a.1, pages A-1 through A-4, of volume 1 of Compilation of Regulations for Tests and Methods of Assay and Certification of Antibiotic and Antibiotic-containing Drugs (Food and Drug Administration of the U.S. Dept. of Health, Education, and Welfare). Circular paper discs, 12.7 millimeters in diameter, are dipped in the solution whose antibacterial power is to be tested, drained and placed on the agar plates. The plates are then incubated at 37° C. for 24 hours after which they are inspected and the diameter of the clearly discernible "halo," or area of no bacterial growth, is recorded. In this specification every reported result is the average of four identical halo tests. A 0.1% solution of a control germicide, mercuric chloride (HgCl_2), gives a 19.8 mm. halo under these test conditions.

Example II

A water solution was made containing 0.03% diisobutyl cresoxy ethoxy ethyl dimethyl benzyl ammonium chloride, 0.2% sodium N-lauroyl sarcosine and 0.5%



in which the polypropylene oxide has a molecular weight of 1500 to 1800 and the polymerized ethylene oxide comprises 80-90% by weight. Disc-halo test results showed this solution to prevent bacterial growth in an area 153% greater than that obtained with the quaternary alone. When 0.25% polyoxyethylene tridecyl alcohol having about twelve ethylene oxide groups replaces the 0.50% $\text{HO}(\text{C}_2\text{H}_4\text{O})_a(\text{C}_3\text{H}_7\text{O})_b(\text{C}_2\text{H}_4\text{O})_c\text{H}$ in the above formula the area of no bacterial growth is even larger, 300% more than that from use of the quaternary alone. A solution of the above quaternary compound, 0.2% sodium N-lauroyl beta alanine and 0.5%



previously described in this example, made in distilled

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water and tested by the disc-halo method, prevented bacterial growth in an area 52% greater than that obtained with only the quaternary compound.

Example III

A mouth rinse (called mouth rinse "A") was made of the following formula:

	Percent
Diisobutyl cresoxy ethoxy ethyl dimethyl benzyl ammonium chloride	0.03
Sodium N-lauroyl sarcosine	0.2
$\text{HO}(\text{C}_2\text{H}_4\text{O})_a(\text{C}_3\text{H}_7\text{O})_b(\text{C}_2\text{H}_4\text{O})_c\text{H}$ wherein the polypropylene oxide has a molecular weight of 1500 to 1800 and polymerized ethylene oxide constitutes 80-90% by weight	0.5
Ethanol	15.0
Adjuvants (color, flavor, saccharine)	0.2
Water (distilled)	84.07

Standard disc-halo tests against *Micrococcus pyogenes*, var. *aureus* resulted in a halo diameter of 23.5 mm. The same tests gave a halo of 21.2 mm. against *Lactobacillus K*.

The above formula was modified to mouthrinse "B" by including 5.0% ethanol, 10.0% glycerine, 0.1% adjuvants and 84.17% water as well as the original amounts of the other three components. A halo 22.5 mm. in diameter was obtained against *Micrococcus pyogenes*, var. *aureus* and 22.5 mm. in the case of *Lactobacillus K*. These results were superior to those obtained from identical testing of several of the leading mouth rinses on the U.S. market. None of the tested rinses was more effective than the invented compositions.

Example IV

Mouthrinse A (formula given in Example III) was tested in vivo by a panel of 12 persons for its effectiveness against oral bacteria. Each person employed the subject mouth rinse on 2 different days and on 2 other days used a control saline rinse, giving a total of 24 results for each test.

Each subject brushed his teeth daily, according to his usual practice, using a paste dentifrice. Shortly after the noon meal of a test day he again brushed his teeth with that dentifrice to lower the microbial population of the mouth and make more reproducible the condition of his oral flora. Lowering the oral bacterial population also makes testing of a mouth rinse more stringent. Thirty minutes after this brushing a representative sample of the oral bacteria was taken by swishing 20 ml. of a 0.85% sterile saline solution in the oral cavity for thirty seconds. The bacterial count was determined by the usual plating methods, using thioglycollate agar as the growth medium. Thirty minutes later the subject rinsed his mouth with 20 ml. of either the test solution or the control saline solution. Ninety minutes after this rinsing a sample of the oral bacteria was taken and plated as before. The subjects took nothing by mouth in the time between use of the mouth rinse and collection of the specimen ninety minutes later.

After 72 hours' incubation at 37° C. the agar plates were examined and a bacterial count was made. Other experiments have proved that the bacterial counts obtained from consecutive saline rinses ½ hour apart are about the same. Considering then that the bacterial count immediately prior to use of the mouth rinse or control rinse is equal to that of thirty minutes earlier the arithmetic average (all figures being in millions) of bacteria found 90 minutes after using the invented mouth rinse was 26.6 compared to 69.9 before, a reduction of 61.9%. On the contrary, comparable figures for the control used were 141.9 and 68.3, showing an increase of 107.6%. If logarithmic means are taken instead of arithmetic averages the test mouth rinse causes

a reduction of bacteria from 46.5 to 25.5, a decrease of 45%. Corresponding results for the control are 112.8 and 43.8, indicating an increase of 158%. Other experimental work has shown that the decrease in bacterial count is not attributable to detergent or wetting effect of the present compositions. In those experiments mouth-washes containing detergents were found ineffective to reduce oral flora.

In the specification the word antibacterial has been used to describe the subject compositions. This word includes within its scope bactericidal, bacteriostatic and bacterial growth-inhibiting.

The above invention has been described in conjunction with various illustrative examples of antibacterial compositions and mouth rinses. It will be obvious to those skilled in the art that other variations and modifications of the invention can be made and that various equivalents can be substituted therein without departing from the principles disclosed herein or going outside the scope of the specification or purview of the claims.

What is claimed is:

1. An antibacterial composition comprising an antibacterial quaternary ammonium compound and a substantially saturated aliphatic acyl amide of a saturated aliphatic monoaminocarboxylic acid compound having 2 to 6 carbon atoms, the aliphatic acyl group thereof having 10 to 18 carbon atoms, the amide being present in amount sufficient to improve the antibacterial action of the quaternary ammonium compound when the composition is dissolved in an aqueous medium.

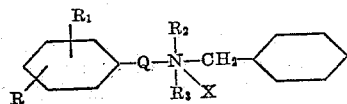
2. An antibacterial composition comprising an antibacterial quaternary ammonium compound and a substantially saturated aliphatic acyl amide of a saturated aliphatic monoaminocarboxylic acid compound having 2 to 6 carbon atoms, the aliphatic acyl group thereof having 10 to 18 carbon atoms and the proportion of quaternary compound to said amide being between 1:25 and 1:2.

3. An antibacterial composition comprising an antibacterial quaternary ammonium compound; a substantially saturated aliphatic acyl amide of a saturated aliphatic monoaminocarboxylic acid compound having 2 to 6 carbon atoms, the aliphatic acyl group thereof having 10 to 18 carbon atoms, the proportion of quaternary compound to said amide being between 1:25 and 1:2; and a water-soluble nonionic surface active compound containing as a hydrophilic portion a polyoxyethylene of 10 to 300 oxyethylene groups and, as a lipophilic portion, a saturated aliphatic radical selected from the group consisting of alcohol radicals having 8 to 18 carbon atoms and polyoxypropylenes of molecular weight between 1000 and 2000, the weight of nonionic surface active agent being between 1 and 5 times the combined weight of quaternary ammonium compound and amide.

4. An antibacterial composition comprising an antibacterial quaternary ammonium compound and a substantially saturated aliphatic acyl amide of sarcosine, the aliphatic acyl group having 10 to 18 carbon atoms, the proportion of quaternary compound to sarcosine amide being between 1:25 and 1:2.

5. An antibacterial composition comprising an antibacterial quaternary ammonium compound and a water-soluble salt of a substantially saturated aliphatic acyl amide of sarcosine, the aliphatic acyl group having 12 to 16 carbon atoms, the proportion of quaternary compound to sarcosine amide being between 1:25 and 1:2.

6. An antibacterial composition comprising an antibacterial quaternary ammonium compound of the formula



where R is an alkyl group of 6 to 12 carbon atoms, R₁

is selected from the group consisting of hydrogen and alkyl groups of 1 to 3 carbon atoms, R₂ and R₃ are alkyl groups of 1 to 3 carbon atoms, Q is an alkoxy intermediate linkage between the hydrophobe and quaternary nitrogen, having an oxygen atom adjacent the benzene ring of the hydrophobe and containing 2 to 6 carbon atoms, and X is a halide selected from the group consisting of bromine and chlorine, and a water soluble salt of an aliphatic acyl amide of sarcosine, the aliphatic acyl group having 12 to 16 carbon atoms, in a proportion, quaternary compound to salt of sarcosine amide, between 1:25 and 1:2.

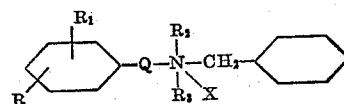
7. An antibacterial composition comprising diisobutyl cresoxy ethoxy ethyl dimethyl ammonium chloride and sodium N-lauroyl sarcosine in a proportion between 1:25 and 1:2.

8. A liquid antibacterial composition comprising an aqueous solution containing by weight about 1 part diisobutyl cresoxy ethoxy ethyl dimethyl ammonium chloride and about 7 parts sodium N-lauroyl sarcosine.

9. An antibacterial liquid mouth rinse comprising an aqueous medium containing by weight 0.01 to 0.1% of an antibacterial quaternary ammonium compound and 0.05 to 0.5% of a substantially saturated aliphatic acyl amide of a saturated aliphatic monoaminocarboxylic acid compound having 2 to 6 carbon atoms, the aliphatic acyl group thereof having 10 to 18 carbon atoms, the proportion of quaternary compound to said amide being between 1:25 and 1:2.

10. An antibacterial liquid mouth rinse comprising an aqueous medium containing by weight 0.01 to 0.1% of an antibacterial quaternary ammonium compound and 0.05 to 0.5% of a substantially saturated aliphatic acyl amide of sarcosine, the aliphatic acyl group thereof having 10 to 18 carbon atoms, the proportion of quaternary compound to sarcosine amide being between 1:25 and 1:2.

11. An antibacterial liquid mouth rinse comprising an aqueous medium containing by weight 0.01 to 0.1% of an antibacterial quaternary ammonium compound of the formula



where R is an alkyl group of 6 to 12 carbon atoms, R₁ is selected from the group consisting of hydrogen and alkyl groups of 1 to 3 carbon atoms, Q is an alkoxy intermediate linkage between the hydrophobe and quaternary nitrogen, having an oxygen atom adjacent the benzene ring of the hydrophobe and containing 2 to 6 carbon atoms, and X is a halide selected from the group consisting of bromine and chlorine, and 0.05 to 0.5% of a substantially saturated aliphatic acyl amide of sarcosine, the aliphatic acyl group thereof having 10 to 18 carbon atoms, the proportion of quaternary compound to sarcosine amide being between 1:25 and 1:2.

12. An antibacterial liquid mouth rinse comprising by weight 0.01 to 0.1% of diisobutyl cresoxy ethoxy ethyl dimethyl ammonium halide and 0.05 to 0.5% sodium N-lauroyl sarcosine, the proportion of quaternary compound to sarcosine amide being between 1:25 and 1:2, 5 to 25% ethyl alcohol and 50 to 95% water.

13. An antibacterial liquid mouth rinse comprising by weight about 0.03% diisobutyl cresoxy ethoxy ethyl dimethyl ammonium chloride, about 0.2% sodium N-lauroyl sarcosine having less than 10% higher fatty acid material based on the weight of said sarcosine compound, about 10% ethyl alcohol and about 90% water.

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