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PREPARATION OF STABLE, SOLID, WATER-SOLUBLE, SURFACE-ACTIVE COMPOSITIONS CONTAINING UREA AND A QUATERNARY AMMONIUM COMPOUND

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This invention relates to improvements in granular, water-soluble, surface-active compositions containing effective proportions of water-soluble, surface-active quaternary ammonium compounds.

A large number of water-soluble, surface-active quaternary ammonium compounds are known. In general, these compounds are substituted ammonium salts in which all four of the hydrogens of the ammonium radical are substituted by organic radicals at least one of which contains a chain of eight to sixteen carbon atoms. While it is possible to produce such water-soluble, surface-active quaternary ammonium compounds which are solids at ordinary atmospheric temperatures, the cost of the special raw materials required makes the cost of such compounds prohibitive in terms of commercial application. Further, although such compounds may be described as "solids," they frequently exhibit an oiliness or greasiness which causes them to agglomerate rapidly when originally flaked or granulated.

For example, coconut oil is a low cost raw material for the production of a particularly valuable group of such quaternary ammonium compounds, the distribution in length of the carbon chains of the constituent fatty acids being such that the quaternary ammonium compounds formed by conversion of the mixed fatty acids to amines followed by methylation and reaction with benzyl chloride are unusually effective surface-active and bactericidal agents. However, even when substantially anhydrous, these compounds are normally either an oily liquid or a greasy solid.

Other known water-soluble, surface-active quaternary ammonium compounds include a lauryl dimethyl benzyl ammonium chloride, cetyl trimethyl ammonium bromide, heptadecyl trimethyl ammonium chloride, lauryl pyridinium chloride, octadecyl pyridinium bromide, decyl chlorobenzyl diethyl ammonium chloride, undecyl benzyl diethyl ammonium bromide, and the like.

These quaternary ammonium compounds are effective in dilutions of as little as $\frac{1}{1000}$ %, $\frac{1}{100}$ % or even $\frac{1}{10000}$ %, for example in sanitizing vessels and utensils used in connection with food and drink, and are commonly used in aqueous solutions of such concentrations. For this reason, and the further reason that they are conveniently prepared in aqueous solution, these compounds are commonly manufactured and distributed as aqueous solutions containing from about 10% to

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about 50% of the quaternary ammonium compound.

In general, the ultimate consumer would prefer, for a variety of practical reasons, to receive such quaternary ammonium compounds as a granular solid, but for the oily or greasy characteristic previously mentioned. Further, since these compounds depress the freezing point of their solutions very little, containers in which such aqueous solutions are shipped, stored or handled must be protected against freezing to avoid losses resulting from resultant container damage. Of course, any such solid product must be of the same order of solubility as the quaternary ammonium compound itself if its practical value is to be maintained.

This problem has been recognized for some time and various expedients for dealing with it have been suggested. Probably the most effective of such proposals has been that of fusing the quaternary ammonium compound with urea. In some proportions, reasonably dry solids can be thus prepared but a fusion temperature upwards of about 110°-115° C. is usually required to obtain a homogeneous product and such temperatures initiate a curious decomposition that continues for an indefinite period even after the fusion is solidified and cooled to ordinary atmospheric temperatures. Such decomposition not only involves substantial losses of the effective agent but also renders the product objectionable during handling and in use.

Proposals involving the admixture of some inorganic salt with the quaternary ammonium compound, to take up any water present, have been less than successful for the reason that the difficulties involved are not to be solved merely by removing or binding the water present. The product remains an oily or greasy product tending to agglomerate.

This invention relates particularly to improvements in granular, water-soluble, surface-active compositions containing effective proportions of a water-soluble, surface-active quaternary ammonium compound and urea.

A mixture of urea in major proportion and such quaternary ammonium compounds in minor but effective proportion can be fused to produce a homogeneous melt, but the temperature required to obtain a homogeneous fusion from such simple mixtures usually exceeds 110°-115° C. and such temperatures initiate the curious decomposition previously mentioned. The decomposition products include ammonia and more odoriferous reaction products and the decomposition reaction,

once initiated, continues with evolution of such decomposition products for a long period. This decomposition is not arrested by cooling and solidifying the fusion or melt.

We have discovered that mixtures of urea comprising not more than about 80% by weight on the total of urea, about 12%–22% by weight on the urea of such water-soluble, surface-active quaternary ammonium compounds and about 4%–6.5% by weight on the total of urea and quaternary ammonium compound of sodium acetate can be fused to produce a homogeneous melt at temperatures above normal atmospheric temperature but not exceeding about 110° C., usually at about 95°–100° C., and that at such temperatures the decomposition previously mentioned does not occur. Some negligible decomposition may occur but it does not continue after the melt is cooled and solidified. This surprising effect enables us to produce a stable, granular, water-soluble homogeneous surface-active composition comprising an effective proportion of the quaternary ammonium compound by producing a fusion in this manner and then cooling the fusion and either flaking or granulating the fusion as it cools and solidifies or grinding the solidified melt.

The granular product of our invention comprises urea, but not more than about 80% by weight on the total product of urea, about 12%–22% by weight on the urea of a water-soluble, surface-active quaternary ammonium compound and about 4%–6.5% by weight on the total of urea and quaternary ammonium compound of sodium acetate. This product may contain as much as 15% by weight on the total of urea, quaternary ammonium compound and sodium acetate of water. The quaternary ammonium compound may be supplied to the fusion operation, for example, as an aqueous solution containing 50% by weight of the quaternary ammonium compound. The direct product of such fusions will, within the stated limits, be stable solids. Or, the quaternary ammonium compound may be dehydrated, partially or to substantially anhydrous condition, before being supplied to the fusion operation. Again, the melt produced by fusing a concentrated aqueous solution of the quaternary ammonium compound with the urea and sodium acetate may be dehydrated after solidification. In any dehydration of the quaternary ammonium compounds or products containing them, temperatures involving decomposition are to be avoided. One particularly advantageous product of our invention comprises urea, but not more than about 80%, by weight on the total product of urea, about 12%–22% by weight on the urea of a water-soluble, surface-active quaternary ammonium compound, about 4%–6.5% by weight on the total of urea and quaternary ammonium compound of sodium acetate and about 8.5%–14% by weight on the total of urea, quaternary ammonium compound and sodium acetate of water.

The proportions stated are important. If the proportion of urea is increased, higher temperatures are required to produce a clear, homogeneous melt. If the proportion of quaternary ammonium compound, or that of water, is increased, the products do not solidify satisfactorily, in particular the products do not flake or granulate readily. If the proportion of sodium acetate is decreased, the fusion temperature required tends to increase and the products do not solidify satisfactorily, in particular the products do not flake or granulate readily. If the proportion of sodium

acetate is increased, the products become hygroscopic and tend to deliquesce and to agglomerate.

The following examples will illustrate the practice and the products of our invention. In these examples, all parts are parts by weight.

Example I

A water-soluble, surface-active quaternary ammonium compound was derived from coconut oil fatty acids, by conventional processing, by conversion of the fatty acids to the corresponding cocoa amines, methylation to form dimethyl cocoa amines and treatment with benzyl chloride to produce cocoa dimethyl benzyl ammonium chloride in the form of a 50% by weight aqueous solution.

20 parts of this solution of the quaternary ammonium compound, 75 parts of urea and 5 parts of anhydrous sodium acetate were fused, with stirring, to a homogeneous melt at a temperature of 103° C., fusion being carried out in an oil bath. This fusion was cooled, solidified and granulated and this granular product was air dried to a total water content of about 1% by weight. The product had no ammoniacal odor and it remained granular and free flowing on prolonged exposure to air at 80° F. with a relative humidity of 68%. Glasses rinsed in a solution of the product containing 400 parts per million of the quaternary ammonium compound were free of odor.

Example II

20 parts of the same solution of the quaternary ammonium compound, 75 parts of urea and 5 parts of anhydrous sodium acetate were fused to a clear melt at a temperature of 95–100° C. in a stainless steel oil jacketed pan. A stainless steel roll, cooled by internal circulation of water at 10° C., dipped into the fusion in the pan, was slowly revolved while the film formed on the roll was scraped off. The film on the roll had a temperature of about 21° C. as it was removed from the roll and was about 0.040" thick. About 33 lbs. of the flake product were recovered per square foot of roll surface per hour. The product was odorless and remained granular on prolonged exposure to air at 80° F. with a relative humidity of 68%.

Other materials may be compounded with the product of our invention. For example, other surface-active materials such as sodium lauryl sulfate, potassium mono-naphthalene sulfonate and alkyl aryl polyether alcohols, and alkylol amides, sodium carbonate, sodium bicarbonate, normal and acid orthophosphates, polyphosphates such as triphosphate and tetraphosphate, polymetaphosphates, pyrophosphates and borates including borax may be included in the composition.

The products of our invention may also include, for example, 2%–5% by weight on the total of urea and quaternary ammonium compound of glacial acetic acid, concentrated sulfuric acid or syrupy phosphoric acid.

The process of our invention is, in a broader aspect, applicable to the production of homogeneous, solid products comprising urea and such water-soluble, surface-active quaternary ammonium compounds in cast shapes, for example, rather than in granular form. If the product is not to be granular, the proportions of quaternary ammonium compound on the urea may be varied over wider limits than those previously stated for granular products. For the production of such non-granular solid products, the

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proportion of the quaternary ammonium compound may range from 3% to 35% by weight on the urea.

We claim:

1. A method of compounding stable, solid, water-soluble, surface-active compositions which comprises fusing together urea, in major proportion but not more than 80% by weight on the total composition of urea, and 12%–22% by weight on the urea of a water-soluble, surface-active quaternary ammonium salt in which all four hydrogens of the ammonium radical are substituted by organic radicals at least one of which contains a chain of 8 to 16 carbon atoms and 4%–6.5% by weight on the total of urea and quaternary ammonium compound of sodium acetate at a temperature above normal atmospheric temperature and not exceeding 110° C. and cooling and solidifying the fusion.

2. A method of compounding stable, solid, water-soluble, surface-active compositions which comprises fusing together urea, in major propor-

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tion but not more than 80% by weight on the total composition of urea, and 3%–35% by weight on the urea of a water-soluble, surface-active quaternary ammonium salt in which all four hydrogens of the ammonium radical are substituted by organic radicals at least one of which contains a chain of 8 to 16 carbon atoms and 4%–6.5% by weight on the total of urea and quaternary ammonium compound of sodium acetate at a temperature above normal atmospheric temperature and not exceeding 110° C. and cooling and solidifying the fusion.

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The following references are of record in the file of this patent:

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