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3,283,005

p-XYLYLENE BIS QUATERNARY AMMONIUM COMPOUNDS

Phillip G. Abend, Cleveland Heights, Ohio, and Eric Jungermann, La Grange, Ill., assignors to Armour and Company, Chicago, Ill., a corporation of Delaware
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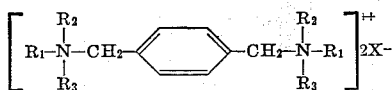
This invention relates to novel p-xylylene bis quaternary ammonium compounds. The compounds of this invention have particular utility as cationic fabric softeners in industrial and household applications. This invention is therefore also concerned with a method of softening fabrics wherein the compounds of this invention are applied to the fabrics.

Certain symmetrical diquaternary ammonium compounds have heretofore been disclosed, such as those described in United States Patent 2,933,529, issued April 19, 1960. As far as is known, however, no one heretofore has described or prepared any compounds which can be classed as p-xylylene bis quaternary ammonium compounds. This invention is therefore based in part on the discovery of such compounds, which have been found to be highly effective cationic fabric softeners.

In the use of cationic fabric softeners, such as dimethyl di-tallow ammonium chloride, an undesirable side effect is frequently obtained. While the fabric may be rendered anti-static, lint-free, and soft after application of the cationic agent, the fabric may tend to resist the absorption of water. Where water absorbency is an important property, such as with diapers, the tendency of the cationic fabric softener to produce a water-proofing effect, and to interfere with rewetting of the fabric, can be a serious problem. This problem tends to become more acute with repeated applications of the fabric softener. The quantity of the softener on the fabric will tend to build-up with each application even though there are intervening washings of the fabric, and the "rewet" property of the fabric can be drastically curtailed.

The discovery that fabric softeners coming within the novel class of compounds of this invention have better rewet properties than previously known cationic fabric softeners is therefore of considerable importance. This invention is therefore particularly concerned with novel p-xylylene bis quaternary ammonium compounds which can be employed as fabric softeners while obtaining superior rewet properties. It will be understood of course that the novel chemical compounds of this invention can be used for other purposes.

The bis quaternary ammonium compounds of this invention can be represented by the following structural formula:



In the above formula, R_1 is an aliphatic hydrocarbon group of from 14 to 22 carbon atoms, R_2 and R_3 are members of the class consisting of methyl, ethyl, hydroxyethyl and hydroxypropyl, and X is an ammonium salt anion. Preferably, R_1 is a saturated alkyl group of from 16 to 18 carbon atoms, such as an octadecyl group. R_1 can be derived from a mixed fatty acid source, the mixed alkyl and alkylene groups of the fatty acids of tallow are particularly desirable. The tallow hydrocarbon groups can be partially or completely hydrogenated to reduce the content of oleyl groups, thereby providing saturated hydrocarbon chains of from 16 to 18 carbon atoms, and predominately saturated C_{18} chains.

Notwithstanding the presence of the large phenyl or xylylene group in the center of the molecule, the com-

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pounds of this invention manifest good rewet properties when employed as cationic fabric softeners. However, the rewet properties tend to be even further improved where the R_2 and R_3 groups are hydroxyethyl or hydroxypropyl groups. A class of compounds wherein R_2 and R_3 are hydroxyethyl groups is particularly preferred. However, superior rewet properties are also obtained when R_1 is a methyl group.

As indicated, the quaternary ammonium compounds of this invention are in their salt form, that is, they are associated with an ammonium salt anion. For use as cationic fabric softeners, it is desirable to have the compounds readily dissolvable or dispersible in water, and the salt form tends to accomplish this result. While any of the known ammonium salt anions can be used, it is preferred to employ anions which are known to promote the water solubility or water dispersibility of the compounds. For example, X can be a halogen anion such as chloride, bromide, or iodine anions. Other anions can be substituted for various purposes, such as sulfate, methylsulfate, phosphate, acetate, citrate, tartrate, and the like. The compounds can be conveniently prepared, as will subsequently be described from reagents which result in X being chloride or other halide anion. However, after formation, the chloride or halide anion can be exchanged for other anions, such as those already listed. Procedures for accomplishing such an exchange are well-known in the art, such as metathesis with the halide form or by the use of anion-exchange resins.

Among the specific compounds coming within the scope of the present invention are the following: p-xylylene bis (dimethyloctadecylammonium chloride), p-xylylene bis (bishydroxyethyloctadecylammonium chloride), p-xylylene bis (bishydroxyethylhexadecylammonium chloride), p-xylylene bis (bishydroxyethyl tallow ammonium chloride), etc.

The compounds of this invention can be readily prepared by reacting a xylene halide, such as p-xylylene chloride or α,α' -dichloro-p-xylene, with a tertiary amine. This reaction can be readily carried out with the reactants dissolved in an inert organic solvent such as isopropanol, ethyl or butyl acetate, dimethyl formamide, etc. Two moles of the tertiary amine are reacted with each mole of the xylene halide to produce the bis quaternary ammonium compound. The reaction is preferably carried out at temperatures ranging from 50 to 150° C., or preferably around 100 to 120° C. The reaction can be conveniently carried out at the boiling point of the solvent employing refluxing. The compounds of this invention or method of preparation is further illustrated by the following specific examples:

EXAMPLE 1

A mixture consisting of p-xylylene chloride (0.25 mole, 43.8 g.), octadecyldimethyl amine (0.55 mole, 169 g.), 380 ml. of n-propyl alcohol and 20 ml. of water was refluxed for four hours. The solvent was removed by evaporation under reduced pressure. The residue was crystallized from a 4:1 mixture of ethyl acetate and isopropyl alcohol. The crystals were washed with petroleum ether and air-dried. Yield of p-xylylene bis (dimethyloctadecylammonium chloride) was 196 g., 99.5% active by titration, 99% of theory.

EXAMPLE 2

A solution of p-xylylene chloride (0.1 mole, 17.5 g.), and octadecylbishydroxyethyl amine (0.23 mole, 80.0 g.) in 200 ml. of N,N-dimethyl formamide was maintained at 110–120° for three and one-half hours, cooled to room temperature and then poured into 600 ml. of acetone. The white precipitate was filtered, washed with petroleum

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ether and air-dried. Yield of p-xylylene bis (bishydroxyethyl-octadecylammonium chloride) was 42 g., 94.5% active by titration, 46.5% of theory.

EXAMPLE 3

A solution of $\alpha'\alpha'$ -dichloro-p-xylene (0.1 mole, 17.5 g.), octadecylbishydroxyethyl amine (0.2 mole, 71.2 g.) and sodium iodide (3.0 g.) in 300 ml. of n-butylacetate was refluxed for two hours. The reaction mixture was allowed to cool down to 40° and 500 ml. of acetone was added. The white solid was filtered, washed with acetone and air-dried. Yield of p-xylylene bis (bishydroxyethylhexadecylammonium chloride) was 85 g., 94% active by titration, 90% of theory.

EXAMPLE 4

A solution of hexadecylbishydroxyethyl amine (0.4 mole, 131 g.), $\alpha'\alpha'$ -dichloro-p-xylene (0.2 mole, 35.0 g.) and sodium iodide (3.0 g.) in 400 ml. of n-butyl acetate was refluxed 1.5 hours, then allowed to cool down to 40° C. Five hundred mls. of acetone was added with stirring and the white solid filtered, then dried in a vacuum oven. Yield of p-xylylene bis (bishydroxyethylhexadecylammonium chloride) was 95 g., 98% active by titration, 55% of theory.

EXAMPLE 5

A solution of tallow bishydroxyethyl amine (0.4 mole, 140 g., I.V.=31.6) $\alpha'\alpha'$ -dichloro-p-xylene (0.2 mole, 35.0 g.), and sodium iodide (3.0 g.) in 400 ml. of n-butyl acetate was brought from 50° to 130° (reflux temperature) in 15 minutes and maintained there for 1 hour. The mixture was cooled down to 40° and poured into 1 liter of acetone. The white solid was filtered and dried by exposure to air over the weekend. Yield of p-xylylene bis (bishydroxyethyl tallow ammonium chloride) was 105 g., 100% active by titration, 60% of theory.

In employing the compounds of this invention as cationic fabric softeners, it is preferred to incorporate them in rinse water following the washing of the fabric. The softener compound can be used at a level ranging from .03 to .3% based on the weight of the fabric being treated (O.W.F.). For most purposes, however, the compounds will be applied at the rate of from .065 to 0.1% (O.W.F.). The cationic softeners are particularly effective with cellulosic fabrics such as cotton fabrics. For example, the softeners can be used to treat cotton diapers, cotton sheets and pillowcases, etc. Since the compounds are highly substantive to cellulosic fabrics, the treatment of such fabrics will usually remove substantially all of the softener compound from the water solution.

The compounds described in the foregoing examples were tested for softening activity on cotton fabrics at levels ranging from .075 to .1% (O.W.F.). They were found to be highly effective softeners. For example, p-xylylene bis (bishydroxyethyloctadecylammonium chloride) was found to be equal in softening activity at a level of .075% to Arquad 2HT at a level of 0.1% (O.W.F.). Arquad 2HT is a commercial cationic softener manufactured by Armour and Company which has become accepted as one of the most effective softeners previously known. The trade name "Arquad 2HT" designates that the compound is di(hydrogenated tallow) dimethyl ammonium chloride.

The compounds of Examples 1 and 2 were compared in re-wet characteristics with Arquad 2HT at levels of 0.1% (O.W.F.). Family-type wash bundles were used composed principally of cotton fabrics. The softener compounds were applied in the last rinse following wash-

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ing with a standard commercial alkyl aryl sulfonate detergent. The test was carried on through four successive applications, and the re-wet time in seconds was measured in terms of average sinking time after each application. The results are set forth below in Table A:

Table A

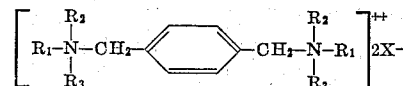
No. of Applications	Average Sinking Time (Seconds)		
	Ex. I	Ex. II	Arquad 2HT
0	8	6	6
1	13	13	20
2	11	8	92
3	8	13	>252
4	9	15	>200

It can be seen from the foregoing data that the compounds of Examples I and II provide excellent re-wet properties even after four applications, and that they are manifestly superior in this respect to Arquad 2HT.

While in the foregoing specification this invention has been described in considerable detail with reference to specific embodiments thereof, it will be apparent to those skilled in the art that the invention is susceptible to additional embodiments and that certain of the details described herein can be varied considerably without departing from the principles of the invention.

We claim:

1. As a composition of matter the compound represented by the formula



wherein R_1 is an aliphatic hydrocarbon group having from 14 to 22 carbon atoms, R_2 and R_3 are members of the class consisting of hydroxyethyl and hydroxypropyl, and X is an ammonium salt anion.

2. The compound of claim 1 wherein R_1 is an octadecyl group.

3. The compound of claim 1 wherein R_2 is hydroxyethyl.

4. The compound of claim 1 wherein R_1 is derived from tallow.

5. The compound p-xylylene bis (bishydroxy ethyloctadecylammonium chloride).

6. The compound p-xylylene bis (bishydroxyethylhexadecylammonium chloride).

7. The compound p-xylylene bis (bishydroxyethyl tallow ammonium chloride).

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CHARLES B. PARKER, *Primary Examiner*.

JULIUS GREENWALD, *Examiner*.

MAYER WEINBLATT, FLOYD D. HIGEL,
Assistant Examiners.