ESTERQUATS, THEIR INTERMEDIATES, A PROCESS TO MAKE THE ESTERQUATS, AND THEIR USE

![Chemical structures](image)

**Abstract**

Esterquats, their intermediates, a process to make the esterquats, and their use as a fabric softener wherein said esterquats are of the formula: \(R_4\text{R}_5\text{N}^+\text{Z}^-\text{X}^-\) wherein \(Z\) is covalently bonded to the nitrogen atom and selected from the group of formulae (I)-(IV), and isomers thereof, wherein \(R_1\) and \(R_2\) are independently selected from linear or branched, saturated or unsaturated C\(_{5-22}\) hydrocarbyl, \(R_3\) is nothing or C\(_{1-20}\) hydrocarbyl, \(R_4\) is C\(_{1-6}\) alkyl or independent \(Z\), \(R_5\) is H, C\(_{1-6}\) alkyl, independent \(Z\), or the residue of the quaternizing agent, such as C\(_{1-30}\) alkyl or alkylken, preferably, C\(_{1-7}\) alkyl or alkylene, \(R_6\) is C\(_{1-6}\) alkyl or independent \(Z\), and \(X^-\) is an ion selected from CH\(_3\), Br\(^-\), F\(^-\), CH\(_3\)SO\(^-\), C\(_2\)H\(_5\)SO\(^-\), H\(_2\)PO\(_4\)^-, PO\(_4^2-\), H\(_3\)PO\(_4\)^-, HPO\(_4^{2-}\), HPO\(_3^{2-}\), HPO\(_2^{3-}\), nitrate\(^-\), formate\(^-\), acetate\(^-\), propionate\(^-\), tartrate\(^-\) and benzoate\(^-\), wherein the total charge of the anions equals the total charge of the cations and said process concerns the reaction of an unsaturated epoxide with an amine or a protonated amine, esterification of the formed intermediate, and optional quaternization.
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ESTERQUATS, THEIR INTERMEDIATES, A PROCESS TO MAKE THE
ESTERQUATS, AND THEIR USE

The invention relates to specific quaternary ammonium compounds having at
least one nitrogen-bonded moiety with at least one ester function (esterquats),
to intermediates for making such esterquats, to compositions comprising one or
more of these esterquats, to a process to make them, and to the use of the
esterquats as a fabric softener.

Quaternary ammonium compounds having a substituent on the nitrogen atom
with two ester groups are known. Also compositions comprising such
ammonium compounds are known. WO 97/47588, for instance, discloses how
2,3-dihydroxypropyl trimethyl ammonium chloride is reacted with lauric acid to
form a composition comprising the corresponding diester quaternary
compound, a propyl-diester-quat (PDQ).

The use of PDQ as a fabric softener is known to result in a very good softening
performance. However, the manufacture of these compounds is cumbersome
and the raw material N,N-dimethyl-1-amino propane-2,3-diol is costly. Also, the
epichlorohydrin typically used to make this raw material for PDQ is less desired
from an environmental viewpoint. Furthermore, it is noted that the production
of PDQ typically requires the use of a solvent, such as isopropanol, in the
quaternization reaction. However, such solvent can transesterify with the PDQ,
resulting in the formation of a contaminant (the fatty acid derivative of the
solvent) and a reduction of the softening performance of the PDQ-containing
product (because less diester compound is present). Therefore, there is a need
for alternative compounds with a better price/performance that can be produced
according to a process that puts less strain on the environment. Preferably, this
process does not require the use of solvents that can transesterify with fatty acid esters.

Our investigations have led us to a new and surprisingly simple process for making new types of quaternary compounds where at least one of the nitrogen substituents contains at least one ester group, and to new intermediates. These compounds, and compositions comprising these compounds, offer a good fabric softening performance and a better price/performance than the conventional fabric softeners. It is noted that the intermediates may be used as fabric softeners themselves. Also, the new process offers advantages from an environmental viewpoint because epichlorohydrin is not used. Furthermore, in one of the preferred embodiments the quaternization step of the amine is not performed towards the end of the synthesis of the surface-active compound, but already on the low-molecular weight amine. The process is improved by this, because it is much simpler to separate and purify the low-molecular weight quaternized amine than to perform such steps on the resulting surface-active compound, if so desired. More specifically, during the processing/washing of a surface-active compound, there is a tendency on the part of the compound to disperse. In another preferred embodiment, a trialkylamine is reacted with an epoxy alkene in the presence of an acid, in order to produce a quaternary intermediate. The process of this preferred embodiment obviates the use of epichlorohydrin as well as the use of undesired solvents. The quaternary intermediate can be esterified to give the preferred diesterquat fabric softeners. However, the monoesterquat that is formed may be useful as a fabric softener as well.

The new quaternary compounds according to the invention are of the formula:

\[ R_4[R_5R_6N^+Z]^- \cdot X^- \]
wherein Z is covalently bonded to the nitrogen atom, and of the following formulae (I-IV)

(I)

(II)

(III)

(IV)

or isomers thereof with the formulae:

(Ia)

(IIa)
wherein,
R₁ and R₂ are independently selected from linear or branched, saturated or unsaturated C₆-22 hydrocarbyl,
R₃ is nothing, or C₁-20 hydrocarbyl,
R₄ is C₁-6 alkyl, C₁-6 alkyne, or independent Z,
R₅ is H, C₁-6 alkyl, independent Z, or the residue of the quaternizing agent, such as C₁-30 alkyl or alkenyl, preferably, C₁-7 alkyl or alkenyl,
R₆ is C₁-6 alkyl or independent Z,
n is 1 or 2, and
X⁻ is an ion selected from Cl⁻, Br⁻, I⁻, F⁻, CH₃SO₄⁻, C₂H₅SO₄⁻, H₂PO₄⁻, HPO₄²⁻, PO₄³⁻, H₂PO₃⁻, H₂PO₄⁻, HPO₂⁻, nitrate⁻, formate⁻, acetate⁻, propionate⁻, tartrate⁻ and benzoate⁻, wherein the total charge of the anions equals the total charge of the cations. The compounds may be used after purification and/or isolation. Preferably, they are part of a composition comprising more than one of the compounds of formulae I-IV and isomers thereof, since extensive isolation steps for the individual compounds can then be refrained from. Purification may include a bleaching and/or adsorption step to convert and/or remove chemicals that cause discolouration.

Preferred are compounds of formula (I), given in full below

```
\[
\begin{array}{c}
\text{CH₃} \\
\text{O} \\
\text{R₄R₅R₆N⁺} \\
\text{O} \\
\text{R₁} \\
\text{O} \\
\text{O} \\
\text{R₂} \\
\text{X⁻} \\
\end{array}
\]
```

or isomers thereof, wherein R₁-R₆ have the meaning as presented above.

More preferably, the compounds of the above formulae I-IV, or the isomers thereof, are of the formula wherein R₁ and R₂ are C₁₂-18. Even more preferably, R₁ and R₂ have a carbon distribution within the range such as can be found in
commercial fatty acids. Also preferred compounds of formulae I-IV or isomers thereof are those of the formula wherein \( R_4 \) and \( R_8 \) are methyl or ethyl.

Other preferred compounds of formulae I-IV or isomers thereof are those of the formula wherein \( R_8 \) is methyl or ethyl.

Preferably, \( n \) is 1.

Further preferred compounds of formulae I-IV or isomers thereof are those of the formula wherein \( X^- \) is chloride, methyl sulfate, or ethyl sulfate.

Another embodiment of the invention is the process to make the compounds according to formulae I-IV and/or isomers thereof. This process involves the reaction of an unsaturated epoxide with an amine, preferably a dialkylamine or trialkylamine, after which the unsaturated, hydroxy group-substituted intermediate is reacted with, on average, 1-2 moles of fatty acid groups per mole of hydroxy group of the intermediate to form an ester. Using this ratio of fatty acid groups will ensure that at least some of the unsaturated bonds are reacted as well to form the preferred diesters.

It is noted that F.F. Blicke and J.H. Biel in *J. Am.Chem.Soc.* 79, 5508-5512 (1957) disclose that 1,2-epoxy-3-butene can be reacted with aqueous dimethylamine hydrochloride to form 1-dimethylamino-2-hydroxy-3-butene. However, it is not disclosed that such a compound can be further reacted with 1-2 moles of fatty acid per mole of this product to form the compounds according to the invention with good fabric softening performance.

Another embodiment of the invention concerns amines that can be formed as intermediates by reacting a dialkylamine with an unsaturated epoxide, followed by esterification. These intermediates are of the formula \( R_4[R_5NZ]_n \), wherein \( R_4 \), \( R_5 \), \( n \), and \( Z \) have the meaning specified above. These intermediates may be used as fabric softeners themselves. However, preferably they are quaternized
in conventional ways with agents of the formula \( R_5X \), in order to give the preferred products of the formulae I-IV and isomers thereof.

Intermediate products formed by reacting a trialkylamine with an unsaturated epoxide in the presence of an acid are preferably of the formula

\[
\begin{align*}
R_4R_6N^+ & - \text{R}_3\text{CH} = \text{CH} \text{OH} \quad (\text{VI}) \\
\text{R}_5R_6N^+ & - \text{X} -
\end{align*}
\]

or the isomers thereof according to the formula

\[
\begin{align*}
\text{R}_5R_6N^+ & \text{CH} = \text{CH} \text{OH} \\
\text{R}_4R_6N^+ & - \text{X} - (\text{VIIa})
\end{align*}
\]

wherein \( \text{R}_3-\text{R}_6 \) and \( \text{X} \) have the meaning specified above.

These intermediates can be easily transformed into the desired compounds of formulae I-IV by direct or trans-esterification. The products of formula VI, or the isomers thereof, may themselves be used as a fabric softener. Therefore, a composition comprising compounds I-IV may also contain intermediates of formula VI, without the fabric softening properties being adversely affected.

A further embodiment of the present invention constitutes the use of the compounds according to formulae I-VI, or the isomers thereof, as a fabric softener. Preferably, the compounds are compounds of formula I or the isomers thereof. The products are well-suited for this use because of their expected biodegradability, due to the two ester functions in the molecule, and their price/performance. It is noted that it is known in the art to further improve the performance of fabric softening compositions by combining the fabric softening compounds with a performance booster selected from the group consisting of cationic and non-ionic surfactants. When used in this fashion, the fabric softening compounds are even more effectively deposited on the textile fabric.
The amine that is reacted with the unsaturated epoxide is of the structure \( R_3[R_5R_6N]_n \), or the protonated form thereof with structure \( R_4[R_5R_6N^+H]_n \), wherein \( R_4, R_5, R_6, n, \) and \( X^+ \) have the meaning defined above. Preferably, a dialkylamine or trialkylamine is reacted with the unsaturated epoxide. Preferably, \( n \) is 1. Of the amines with \( n \) is 2, ethylene diamine is preferred.

The unsaturated epoxides that can be used are preferably 1,2-epoxyalkenes of the formula

\[
\begin{array}{c}
\text{O} \\
\text{R}_3
\end{array}
\]

wherein \( R_3 \) has the meaning defined above.

Preferred 1,2-epoxyalkenes are 1,2-epoxy-3-butene, 1,2-epoxy-4-pentene, 1,2-epoxy-5-hexene, 1,2-epoxy-6-heptene, 1,2-epoxy-3-pentene, 1,2-epoxy-3-hexene, 1,2-epoxy-3-heptene, and 1,2-epoxy-4-hexene. More preferred 1,2-epoxyalkenes are 1,2-epoxy-3-butene, 1,2-epoxy-3-pentene, 1,2-epoxy-3-hexene, and 1,2-epoxy-3-heptene. Most preferred is 1,2-epoxy-3-butene.

At least \( n \) mole of 1,2-epoxyalkene is to react with one mole of \( R_4[R_5R_6N]_n \), or the protonated form thereof with structure \( R_4[R_5R_6N^+H]_n \), according to the invention. However, if a monoalkylamine or ammonia is used to make the indicated dialkylamine or trialkylamine in situ by first reacting it with 1,2-epoxyalkene, then up to four moles of 1,2-epoxyalkene can be reacted per mole of amine.

The ester functions in the compounds of the invention typically are derived from saturated or unsaturated, linear or branched \( C_{6-30} \), preferably \( C_{8-22} \), more preferably \( C_{12-18} \), most preferably naturally occurring, fatty acids, optionally substituted with, e.g., one or more hydroxy groups. Preferred fatty acids used in making the compounds of the invention include coconut, palm, palm kernel, soybean, oleic, tallow, rapeseed, canola, behenic, eruca fatty acids, and
mixtures thereof. Preferably, a fatty acid mixture comprising at least 50 percent by weight (% w/w) of C_{12-18} fatty acids is used.

The acids may be used as such in a conventional direct esterification process, but also derivatives can be used, such as the corresponding acid chlorides or (mixed) anhydrides. In transesterification reactions typically a fatty acid ester is used. In such transesterification reactions preferably the methyl, ethyl and/or glycerol esters of the acids are used. Most preferred are mono-, di- and/or triglycerides of the acids. In the esterification process preferably a conventional (trans)esterification catalyst is used, such as hypophosphorous acid.

Preferably 50-100 percent, on a molar basis, of the hydroxy functions of the 1-amino-2-hydroxy-alkene intermediate formed after reacting amine and 1,2-epoxyalkene is esterified. More preferably, 75-100 mole percent of the hydroxy groups is esterified. Also, preferably 50-100 percent, on a molar basis, of the unsaturated functions of the 1-amino-2-hydroxy-alkene intermediate is reacted with fatty acid to give an ester function. More preferably, 75-100 mole percent of the alkene functions is esterified.

If a dialkylamine is used in the reaction with the 1,2-epoxyalkene, then the resulting diester bearing amine is to be quaternized in order to achieve the preferred fabric softening compounds according to the invention. The quaternization step is conventional, using agents of the formula R_3X, wherein R_3 and X have the meaning defined above. Examples of conventional quaternizing agents include, but are not limited to, dimethyl sulphate, diethyl sulphate, methyl chloride, methyl bromide, methyl iodide, benzyl chloride, benzyl bromide, allyl chloride, and allyl bromide.

However, in a preferred embodiment of the invention, the raw material is not a dialkylamine but a trialkylamine. When such trialkylamines are reacted with the unsaturated epoxide in the presence of a conventional activator for the ring
opening of epoxides, typically an acid, such as hydrochloric acid, sulfuric acid, hydrochloric salts of amines, the hydrochloric salt of pyridine, and the like, then the corresponding quaternary ammonium group-bearing unsaturated alcohol is formed. These alcohols can be converted to the corresponding mono- and/or diesterquaternary ammonium compounds by appropriate esterification. The esterification can be a direct esterification or a transesterification, both processes being known in the art for conventional esterification processes. Preferably, C_6-22 fatty acid groups are introduced into said esterification process, since such groups are needed to obtain the desired fabric softening effect.

The resulting quaternary ammonium compounds, having a nitrogen substituent with at least one ester group, can be used as is or after purification and/or isolation. The compounds, preferably the diester-bearing compounds, are pre-eminently suited for use as a biodegradable fabric softener.

The invention is elucidated by the following, non-optimized, examples.

**Materials used:**
- Hypophosphorous acid ex Aldrich
- Stearic acid ex Merck
- Dimethylaminobutenol ex Eastman
- Butadiene monoxide (1,2-epoxy-3-butene) ex Aldrich

**Procedure**

A mixture of N-(2-hydroxybut-3-en-1-yl)-N,N,N-trimethylammonium chloride and N-(1-hydroxybut-3-en-2-yl)-N,N,N-trimethylammonium chloride was produced by charging a 10 ml flask, equipped with a stirrer, with a mixture of 2.55 g (26.7 mmoles) trimethylamine hydrochloride, 0.06 g (0.5 mmole) of 47.7 %w/w of trimethylamine in water, and 2 g water, and cooling the mixture to 10°C. Then
2.09 g (29.9 mmoles) of 1,2-epoxybutene were added dropwise during 1 hour while the temperature was maintained at 10°C. Thereafter, the mixture was heated to 40°C and further reacted for 2 hours. Subsequently, 1 ml of 1M HCl was added, after which the water was evaporated at atmospheric pressure. The desired mixture remained.

Example 1
In a one-necked flask of 100 ml, equipped with stirrer and water separator, 1.58 g (10.2 mmoles) of a mixture of N,N-dimethyl-1-aminobut-3-en-2-ol hydrochloride and N,N-dimethyl-2-aminobut-3-en-1-ol hydrochloride with a purity of 97.5% by weight (%w/w) was combined with 6.07 g (21.4 mmoles) stearic acid. To the mixture, 1 ml of 50% w/w of hypophosphorous acid in water was added as a catalyst. The mixture was stirred for 4 hours at 160°C at a reduced pressure of 1 mbar.

A mixture of 2,3-di(stearołożyxy)but-1-yl)dimethylamine hydrochloride and 1,3-di(stearołożyxy)but-2-yl)dimethylamine hydrochloride was formed with a yield of 24 mole%, based on the amino compound. The product can be quaternized in conventional ways.

Example 2
In a one-necked flask of 100 ml, equipped with stirrer and water separator, 1 ml of 50% by weight hypophosphorous acid in water was added to a mixture of 1.71 g (10.3 mmoles) N-(2-hydroxybut-3-en-1-yl)-N,N,N-trimethylammonium chloride/N-(1-hydroxybut-3-en-2-yl)-N,N,N-trimethylammonium chloride and 5.96 g (21.0 mmoles) stearic acid. With stirring, the mixture was reacted at 160°C for 4 hours under vacuum at a pressure of 1.33 mbar.

A mixture of 2,3-di(stearołożyxy)but-1-yl)trimethyl ammonium chloride (or 1-N,N,N-trimethylammoniumchloride-butane-2,3-di-stearate) and 1-3-di-
(stearoyloxy)but-2-yl)trimethylammonium chloride was formed with a yield of 49 mole\%, based on the ammonium compound. The product has good fabric softening properties.
Claims

1. Quaternary ammonium compounds of the formula \( R_4[R_3R_6N^Z]_n X \), wherein \( Z \) is covalently bonded to the nitrogen atom and selected from the group of the following formulae (I-IV)

\[
\begin{align*}
(I) & \quad \begin{array}{c}
\text{H}_2 \\
\text{C}_2 \\
\text{R}_3 \\
\text{O} \\
\text{R}_2 \\
\end{array} \\
\text{R}_1 \\
\text{O} \\
\text{O} \\
\text{CH}_3 \\
\text{O} \\
\end{align*}

(II) \quad \begin{array}{c}
\text{H}_2 \\
\text{C}_2 \\
\text{R}_3 \\
\text{O} \\
\text{R}_2 \\
\end{array} \\
\text{O} \\
\text{H} \\
\text{CH}_3 \\
\text{O} \\
\end{align*}

(III) \quad \begin{array}{c}
\text{H}_2 \\
\text{C}_2 \\
\text{R}_3 \\
\text{O} \\
\text{R}_2 \\
\end{array} \\
\text{R}_1 \\
\text{O} \\
\text{O} \\
\text{CH}_3 \\
\text{O} \\
\end{align*}

(IV) \quad \begin{array}{c}
\text{H}_2 \\
\text{C}_2 \\
\text{R}_3 \\
\text{O} \\
\text{R}_2 \\
\end{array} \\
\text{R}_1 \\
\text{O} \\
\text{O} \\
\text{CH}_2 \\
\end{align*}

and the isomers of any of these compounds, wherein \( R_1 \) and \( R_2 \) are independently selected from linear or branched, saturated or unsaturated \( C_{6-22} \) hydrocarbyl, \( R_3 \) is nothing or \( C_{1-20} \) hydrocarbyl, \( R_4 \) is \( C_{1-6} \) alkyl, \( C_{1-6} \) alkyene, or independent \( Z \).
$R_5$ is H, C$_{1-6}$ alkyl, independent $Z$, or the residue of the quaternizing agent, such as C$_{1-30}$ alkyl or alkenyl, preferably, C$_{1-7}$ alkyl or alkenyl, $R_6$ is C$_{1-6}$ alkyl or independent $Z$, n is 1 or 2, and

$X^-$ is an ion selected from Cl$^-$, Br$^-$, I$^-$, F$^-$, CH$_3$SO$_4^-$, C$_2$H$_5$SO$_4^-$, H$_2$PO$_4^-$, HPO$_4^{2-}$, PO$_4^{3-}$, H$_2$PO$_3^-$, HPO$_3^{2-}$, H$_2$PO$_2^-$, HPO$_2^{2-}$, nitrate$^-$, formate$^-$, acetate$^-$, propionate$^-$, tartrate$^-$ and benzoate$^-$, wherein the total charge of the anions equals the total charge of the cations.

2. Compounds according to claim 1 of the formula

\[
\begin{array}{c}
\text{R}_4\text{R}_5\text{R}_6\text{N}^+ \\
\text{R}_1\text{O} \\
\text{CH}_3\text{O}
\end{array}
\begin{array}{c}
\text{R}_3\text{O} \\
\text{R}_2\text{N}^+ \\
\text{R}_1\text{O}
\end{array}
\xrightarrow{n} X^-, \text{ or isomers thereof,}
\]

wherein $R_1$-$R_6$, n, and $X^-$ have the meaning given in claim 1.

3. Compounds according to claim 2 of the formula

\[
\begin{array}{c}
\text{R}_4\text{R}_5\text{R}_6\text{N}^+ \\
\text{R}_1\text{O} \\
\text{CH}_3\text{O}
\end{array}
\begin{array}{c}
\text{R}_3\text{O} \\
\text{R}_2\text{N}^+ \\
\text{R}_1\text{O}
\end{array}
\xrightarrow{} X^-, \text{ or isomers thereof,}
\]

wherein $R_1$, $R_2$, $R_4$-$R_6$ and $X^-$ have the meaning given in claim 1.

4. Compounds according to any one of claims 1-3 wherein $R_1$ and $R_2$ are independently selected from linear or branched, saturated or unsaturated C$_{12-18}$ alkyl groups.
5. Compounds according to any one of the preceding claims, characterized in that \( R_4 \) and \( R_6 \) are methyl.

6. Compounds according to any one of the preceding claims wherein \( X^- \) is chloride, methyl sulfate or ethyl sulfate.

7. Intermediates for making one or more of the compounds of the preceding claims according to the formula \( R_4[R_6NZ]_n \), wherein \( R_4, R_6, n \), and \( Z \) have the meaning given in claim 1.

8. Compositions comprising one or more of the compounds according to any one of the preceding claims.

9. A process to make the compounds of claim 1 comprising the steps of:

   - reacting an unsaturated epoxide of the formula \( \text{O} \) \( \text{R}_3 \) \( \text{=C} \) with an amine or protonated amine of the formula \( R_4[R_6N]_n \) or \( R_4[R_6N^+H]_n \) \( X^- \), wherein \( R_3, R_4, R_6, n \), and \( X^- \) have the meaning given in claim 1, and

   - esterification of the intermediate with, on average, 1-2 moles of fatty acid derivatives, comprising the moieties \( R_1 \text{-C(O)}^-, R_2 \text{-C(O)}^- \) or mixtures thereof, per mole of OH groups of the intermediate,

   - an optional conventional quaternization either before or after said esterification step.

10. A process according to claim 9, characterized in that a trialkylamine is reacted with the unsaturated epoxide.

11. A process according to claim 9 or 10, characterized in that a product according to any one of claims 2-6 is formed.
12. Use of a compound or composition according to any one of claims 1-8 as a fabric softener.

13. Use according to claim 12 wherein the compound is used in combination with a conventional performance booster selected from the group consisting of cationic and non-ionic surfactants.
## INTERNATIONAL SEARCH REPORT

### A. CLASSIFICATION OF SUBJECT MATTER

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According to International Patent Classification (IPC) or to both national classification and IPC

### B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

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Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

### C. DOCUMENTS CONSIDERED TO BE RELEVANT

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Date of the actual completion of the international search

17 May 2000

Date of mailing of the international search report

24/05/2000

Name and mailing address of the ISA

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Rufet, J
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