nium compounds. A nanofiltration method specifically adapted for this catalyst system has not hitherto been disclosed.

Extensive research has revealed that membranes comprising a separation-active layer of crosslinked silicone acrylates and/or polydimethylsiloxane and/or polyimide are useful for removing such a catalyst system from the reaction mixture of an epoxidation of cyclic unsaturated C12 compounds. The separation-active layer may comprise each of these three membrane materials individually or in combination.

The disclosure thus provides a method of removing a homogeneous catalyst system from a reaction mixture wherein a membrane comprising at least one separation-active layer separates the reaction mixture in such a way that the homogeneous catalyst system is concentrated in the membrane retentate at least to an extent wherein the reaction mixture comprises at least one partially epoxidized cyclic unsaturated compound comprising twelve carbon atoms and the membrane separation-active layer comprises crosslinked silicone acrylates and/or polydimethylsiloxane (PDMS) and/or polyimide.

In an aspect, there is provided a method to prepare an epoxidized cyclic unsaturated compound comprising twelve carbon atoms, comprising:
epoxidizing a cyclic unsaturated compound comprising 12 carbon atoms with hydrogen peroxide in a two phase organic aqueous system in the presence of a homogeneous catalyst and phase transfer reagent;
separating and removing the aqueous phase to obtain an organic phase containing the epoxidized cyclic unsaturated compound comprising twelve carbon atoms, the homogeneous catalyst and the phase transfer reagent;
contacting the organic phase with a membrane under a transmembrane pressure of 5 to 80 bar to obtain a permeate comprising the epoxidized cyclic unsaturated compound comprising twelve carbon atoms and a retentate comprising the homogeneous catalyst and phase transfer reagent;
wherein the membrane is a composite membrane comprising a support material and a membrane separation-active layer comprising a crosslinked silicone acrylate and

the homogeneous catalyst comprises an oxide, a mixed oxide, an oxoacid, a salt of an oxoacid, a carbonyl derivative, a sulphide, a chloride, an oxychloride or a stearate of an element selected from the group consisting of tungsten, molybdenum and vanadium.

The reaction mixture comprises two immiscible or poorly miscible liquid phases. One of the phases comprises substantially water (aqueous phase). This phase may additionally comprise hydrogen peroxide, catalyst, phase transfer reagent and traces of the cyclic unsaturated C12 compound, traces of the epoxidized cyclic unsaturated C12 compound and traces of the descendent product thereof such as diols for example. The other phase (organic phase) typically comprises substantially epoxidized cyclic unsaturated C12 compounds as epoxidation reaction product. Said phase may additionally comprise the reaction mixture comprising the unmodified cyclic unsaturated C12 reactant compounds, the catalyst and the phase transfer reagent.

The two liquid phases are separated before the reaction mixture is supplied to the membrane. Said separation may be carried out using a phase separation vessel for example. As such, the reaction mixture is applied to the membrane as a monophasic mixture rather than a biphasic mixture. It is preferable to subject the non-aqueous (organic) phase to membrane separation. The catalyst may also be removed from the

Claims

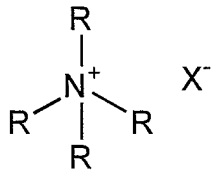
1. A method to prepare an epoxidized cyclic unsaturated compound comprising twelve carbon atoms, comprising:
epoxidizing a cyclic unsaturated compound comprising 12 carbon atoms with hydrogen peroxide in a two phase organic aqueous system in the presence of a homogeneous catalyst and phase transfer reagent;
separating and removing the aqueous phase to obtain an organic phase containing the epoxidized cyclic unsaturated compound comprising twelve carbon atoms, the homogeneous catalyst and the phase transfer reagent;
contacting the organic phase with a membrane under a transmembrane pressure of 5 to 80 bar to obtain a permeate comprising the epoxidized cyclic unsaturated compound comprising twelve carbon atoms and a retentate comprising the homogeneous catalyst and phase transfer reagent;
wherein the membrane is a composite membrane comprising a support material and a membrane separation-active layer comprising a crosslinked silicone acrylate and
the homogeneous catalyst comprises an oxide, a mixed oxide, an oxoacid, a salt of an oxoacid, a carbonyl derivative, a sulphide, a chloride, an oxychloride or a stearate of an element selected from the group consisting of tungsten, molybdenum and vanadium.

2. The method according to claim 1, wherein the aqueous phase comprises the hydrogen peroxide.

3. The method according to claim 1, wherein the cyclic unsaturated compound comprising twelve carbon atoms is cyclododecene.

4. The method according to claim 3, wherein the phase transfer reagent is a tertiary or quaternary ammonium compound.

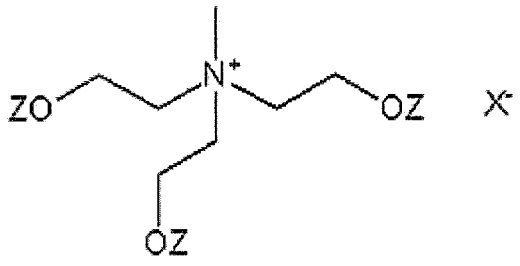
5. The method according to claim 4, wherein the phase transfer reagent is a quaternary ammonium compound and is an esterquat of formula (I)



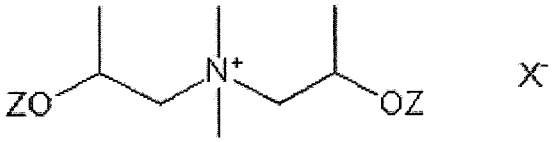
(I)

where the substituents R are independently identical or different alkyl or alcohol groups comprising from 1 to 4 carbon atoms and at least one of the substituents R is an alcohol group which comprises from 1 to 4 carbon atoms and which has been esterified with a saturated or unsaturated fatty acid comprising from 1 to 30 carbon atoms and X^- is a counteranion.

6. The method according to claim 4, wherein the phase transfer reagent is a quaternary ammonium compound and is a quaternary ammonium compound ester according to formulae (II) and (III)



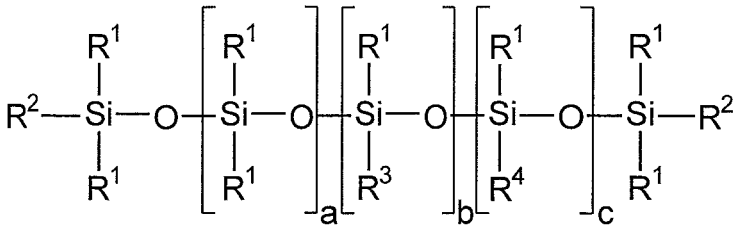
(II)



(III)

where Z denotes one or more saturated or unsaturated fatty acids comprising from 1 to 30 carbon atoms and X^- is a counteranion.

7. The method according to claim 1, wherein the silicone acrylate is of formula IV:



(IV)

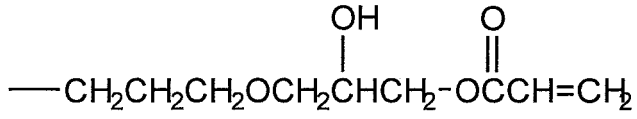
wherein

a is from 1 to 500, b is from 1 to 25 and c is from 0 to 20,

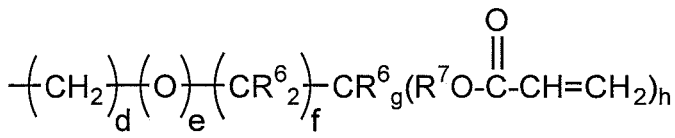
the substituents R^1 are independently identical or different alkyl or aryl groups which comprise from 1 to 30 carbon atoms and which may bear at least one ether, ester, epoxy and/or hydroxy group,

the substituents R^2 are independently identical or different substituents selected from the group consisting of R^1, R^3 and R^4,

the substituents R^3 are independently identical or different acrylate groups of formulae V or VI:



(V)



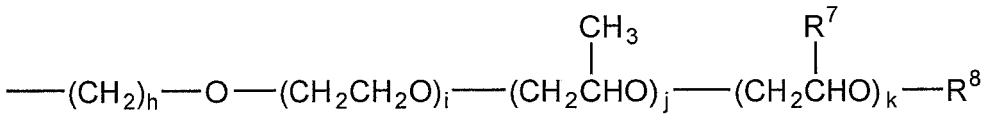
(VI),

wherein d is from 0 to 12, e is from 0 to 1, f is from 0 to 12, g is from 0 to 2, h is from 1 to 3 and g + h is 3,

the substituents R^6 are independently identical or different alkyl or aryl groups comprising from 1 to 30 carbon atoms or hydrogen,

the groups R^7 are identical or different divalent hydrocarbon radicals,

the substituents R^4 are independently identical or different polyether groups of formula (VII):



(VII),

wherein i is from 0 to 12, j is from 0 to 50, k is from 0 to 50 and l is from 0 to 50,

the substituents R^8 are identical or different alkyl or aryl groups comprising from 2 to 30 carbon atoms and

R^9 is an alkyl, aryl or acyl group comprising from 2 to 30 carbon atoms or hydrogen.

8. The method according to claim 1, wherein the separation-active layer is arranged on a carrier material selected from the group consisting of polyacrylonitrile, polyimide, polyether ether ketone, polyvinylidene fluoride, polyamide, polyamide-imide, polyethersulphone, polybenzimidazole, sulphonated polyether ketone, polyethylene, polypropylene and mixtures thereof.

9. The method according to claim 1, wherein the applied transmembrane pressure is from 10 to 60 bar.

10. The method according to claim 1, wherein the separation is carried out at a temperature of from 25°C to 160°C.

11. The method according to claim 7, wherein the groups R^7 are identical or different divalent hydrocarbon radicals of the formula $-CR^6_2-$.

12. The method according to claim 7, wherein the groups R^7 are CH_2.

13. The method according to claim 1, wherein the epoxidizing and the separating are carried out simultaneously.