



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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<b>(21) International Application Number:</b> PCT/US98/11393 <b>(22) International Filing Date:</b> 10 June 1998 (10.06.98)  <b>(30) Priority Data:</b> 60/049,338            10 June 1997 (10.06.97)            US 08/911,795            15 August 1997 (15.08.97)            US  <b>(71) Applicant:</b> HENKEL CORPORATION [US/US]; Suite 200, 2500 Renaissance Boulevard, Gulph Mills, PA 19406 (US).  <b>(72) Inventors:</b> WIGGINS, Michael, S.; 2405 Hillock Court, Lansdale, PA 19446 (US). DEVORE, David, I.; 112 Alberts Way, Langhorne, PA 19047 (US). BROADBENT, Ronald, W.; 737 Jarrett Road, Horsham, PA 19044 (US).  <b>(74) Agent:</b> MILLSON, Henry, E., Jr.; Henkel Corporation, Suite 200, 2500 Renaissance Boulevard, Gulph Mills, PA 19406 (US).		<b>(81) Designated States:</b> AU, CA, JP, MX, European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).  <b>Published</b> <i>With international search report.</i>
<b>(54) Title:</b> CROSS-LINKED REACTION PRODUCTS OF ALKOXYLATED ALCOHOLS AND ALKYLENE GLYCOLS		
<b>(57) Abstract</b>  The reaction product of A) a linking compound of formula (I): $R^1(X)_3$ , wherein each X group is a halogen or one X group is halogen and two X groups with 2 carbon atoms from the $R^1$ group and an oxygen atom form an epoxy group, and $R^1$ is an alkanetriyl group containing from 3 to 10 carbon atoms; B) a compound of formula (II): $R^2(OA)_n-OH$ , wherein $R^2$ is an aliphatic group containing from 4 to 36 carbon atoms, n is a number of from 1 to 200, and each OA group is independently an ethyleneoxy, 1,2-propyleneoxy, or 1,2-butyleneoxy group; and C) a compound of formula (III): $H-(OA)_m-OH$ , wherein OA is as defined above, and m is a number of from 1 to 500; processes for its preparation; and method for its use as a thickener in aqueous compositions.		

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## CROSS-LINKED REACTION PRODUCTS OF ALKOXYLATED ALCOHOLS AND ALKYLENE GLYCOLS

### 1. Field of the Invention

This invention relates to polymeric compounds useful as thickeners for aqueous compositions, especially hydrophilic emulsion polymer latexes.

### 5 2. Background of the Invention

Thickeners for aqueous systems are often required to obtain the rheological properties needed for various applications. Such aqueous-based systems include latex paints, printing inks, protective coatings for paper and metal, and the like. Known thickeners include natural polymers such as  
10 casein and alginates, and synthetic materials such as certain cellulose derivatives, acrylic polymers, and polyurethane polymers.

### SUMMARY OF THE INVENTION

The present invention relates to compounds useful as thickening  
15 agents for aqueous compositions, especially hydrophilic latex paints, to aqueous dispersions or solutions containing the compounds, to methods for their preparation, and to aqueous compositions containing these compounds.

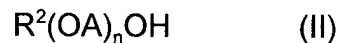
The compounds of the invention are the reaction products of

A) a linking compound of formula I

20  $R^1(X)_3$  (I)

wherein each X group is a halogen atom or one X group is a halogen atom and two X groups with two carbon atoms in the R<sup>1</sup> group and an oxygen atom form an epoxy group, and R<sup>1</sup> is an alkanetriyl group containing from 3 to 10 carbon atoms;

5 B) a compound of formula II



wherein R<sup>2</sup> is an aliphatic group containing from 4 to 36 carbon atoms, n is a number of from 1 to 200, and each OA group is independently an ethyleneoxy, 1,2-propyleneoxy, or 1,2-butyleneoxy group; and

10

C) a compound of formula III



wherein OA is as defined above, and m is a number of from 1 to 500;

15

wherein the ratio of moles of component A) to the sum of the OH equivalents in components B) plus C) is from 0.7:1 to 0.99:1.

### DETAILED DESCRIPTION OF THE INVENTION

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Other than in the operating examples, or where otherwise indicated, all numbers expressing quantities of ingredients or reaction conditions used herein are to be understood as modified in all instances by the term "about".

As stated above, this invention relates to compounds and aqueous solutions or dispersions thereof useful as thickening agents for aqueous

compositions, especially hydrophilic latex paints, wherein the compounds of the invention are the reaction products of

- A) a linking compound of formula I



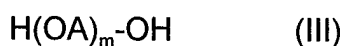
5 wherein each X group is a halogen atom or one X group is a halogen atom and two X groups with two carbon atoms in the R<sup>1</sup> group and an oxygen atom form an epoxy group, and R<sup>1</sup> is an alkanetriyl group containing from 3 to 10 carbon atoms;

- B) a compound of formula II



wherein R<sup>2</sup> is an aliphatic group containing from 4 to 36 carbon atoms, n is a number of from 1 to 200, and each OA group is independently an ethyleneoxy, 1,2-propyleneoxy, or 1,2-butyleneoxy group; and

- 15 C) a compound of formula III



wherein OA is as defined above, and m is a number of from 1 to 500;

wherein the ratio of moles of component A) to the sum of the  
20 OH equivalents in components B) plus C) is from 0.7:1 to 0.99:1, and preferably from 0.7:1 to 0.9:1, and more preferably from 0.8:1 to 0.9:1.

The linking compound of formula I is preferably epichlorohydrin although other epihalohydrins can also be used. Also, trihaloalkanes can be

used, such as 1,2,3-trichloropropane, 1,2,4-trichlorobutane, 1,3,6-trichlorohexane, and the like. Instead of chlorine in the epihalohydrins and the trihaloalkanes, the corresponding bromine and iodine compounds can also be used, including compounds containing two or even all three of the  
5 above halogens.

The compound of formula II is an aliphatic alkoxyated alcohol. The R<sup>2</sup> group in formula II is preferably a straight or branched chain alkyl group containing from 12 to 20 carbon atoms, more preferably from 16 to 20 carbon atoms, and most preferably is a C<sub>20</sub> Guerbet alkyl group. However, the R<sup>2</sup>  
10 group can also be a linear or branched alkenyl or alkynyl group, a saturated carbocyclic moiety, an unsaturated carbocyclic moiety having one or more multiple bonds, a saturated heterocyclic moiety, or an unsaturated heterocyclic moiety having one or more multiple bonds. The number of alkoxy groups, i.e. the value for n in formula II, is preferably from 80 to 120,  
15 and more preferably from 80 to 100. The OA groups are preferably all ethyleneoxy groups.

The compound of formula III is a polyalkylene glycol in which the OA groups are as defined above for the compounds of formula II. The OA groups are preferably all ethyleneoxy groups. The value for m in formula III is  
20 preferably a number from 50 to 500.

With respect to components A), B) and C), one or more of these components can be a mixture of compounds falling within the respective formulas therefor.

It is critical for the compounds of the invention that the ratio of moles of

component A) to the sum of the OH equivalents in components B) plus C) be in the range of 0.7:1 to 0.99:1 since a ratio of 1:1 or greater will result in the formation of an aqueous gel rather than an aqueous solution which is required for use as a thickening agent for aqueous compositions such as latex paints. Conversely, a ratio of less than 0.7:1 will result in less branching and accordingly less effective thickening activity per unit weight of active compound. The compounds of the invention are designed to be branched compounds, whereas prior to the present invention it was believed that branching would be detrimental to the performance of thickeners for latex paints.

The molar ratios of components used to prepare the present compounds can be selected from the following ranges:

Component A):component B):component C) = (0.5 to 1):1:(0.1 to 1), provided that the ratio of moles of A) to the sum of the OH equivalents in B) plus C) must be in the range of from 0.7:1 to 0.99:1. Also, the degree of branching can be further increased as desired by increasing the ratio of component B) to component C) in the reaction product.

The compounds of the invention are preferably prepared in aqueous solution. These aqueous solutions typically contain from 15 to 40% by weight of solids, preferably from 20 to 40% by weight of solids. In addition, these aqueous solutions can also contain from 1 to 30% by weight of a water-miscible cosolvent, which acts as a viscosity modifier. Suitable cosolvents include liquid polyols, liquid ethoxylated and/or propoxylated C<sub>1-8</sub> alcohols, or liquid ethoxylated and/or propoxylated C<sub>1-8</sub> carboxylic acids. A liquid polyol is

any compound having two or more -OH groups which is a liquid at room temperature, examples of which include but are not limited to ethylene glycol, 1,2-propylene glycol, 1,3-propylene glycol, and 1,2-butylene glycol. A liquid ethoxylated and/or propoxylated C<sub>1-8</sub> alcohol is any aliphatic alcohol

5 ethoxylated and/or propoxylated to any degree of ethoxylation or propoxylation and which is a liquid. Compounds in which the -OH group of the liquid ethoxylated and/or propoxylated C<sub>1-8</sub> alcohol is etherified with a C<sub>1-4</sub> alkyl group are also included in this group. A liquid ethoxylated and/or

10 propoxylated C<sub>1-8</sub> carboxylic acid is any aliphatic carboxylic acid ethoxylated and/or propoxylated to any degree of ethoxylation or propoxylation and which is a liquid. Preferred viscosity modifiers include butoxy triglycol (triethylene glycol monobutyl ether), butyl carbitol (diethylene glycol monobutyl ether), or 1,2-propylene glycol. Also preferred are combinations of butoxy triglycol, butyl carbitol, and 1,2-propylene glycol which total from 1% to 30% by weight

15 of the total composition.

The process of the invention used to prepare the aqueous solutions containing the compounds of the invention is preferably carried out using the following steps:

l) components B) and C), i.e. the aliphatic alkoxyated alcohol and

20 the polyalkylene glycol components, are reacted together in the presence of an organic solvent that will azeotrope water, e.g. benzene, toluene, or xylene and in the presence of from 1 to 1.5 equivalents, based on OH groups, of an alkali metal hydroxide, preferably aqueous concentrated sodium hydroxide, or an alkali or alkaline earth metal lower alkoxide, e.g. sodium methoxide, under

reflux conditions, preferably at atmospheric pressure, to remove water of reaction and any water introduced with the alkali metal hydroxide through azeotropic distillation. The reflux temperature is of course dependent on the organic solvent used in this step. The reaction is continued until substantially  
5 all of the water in the reaction mixture is removed, e.g. to a residual water content of 0.2% by weight or less. The reaction mixture is then preferably cooled.

II) component A), i.e. the linking compound of formula I, is then added to the reaction mixture from step I) and reacted at a temperature of  
10 from 60°C to the reflux temperature of the organic solvent, preferably at a temperature of from 100 to 110°C.

III) the organic solvent is then removed from the reaction mixture resulting from step II), preferably by vacuum distillation.

IV) water is then added to the reaction product from step III) to form  
15 an aqueous solution having a solids content of from 15 to 40% by weight, preferably from 20 to 40% by weight.

As discussed above, a cosolvent can then optionally be added to the aqueous solution.

The compounds of the invention and the aqueous solutions containing  
20 them are useful as thickeners for aqueous compositions, particularly for use in hydrophilic emulsion polymer latexes. They can be added to aqueous compositions so that from 0.1 to 20% by weight of the compounds of the invention are present, based on the solids content of the aqueous composition.

There are a number of advantages for using the compounds of the invention. For example, smaller quantities of the present compounds are needed compared to conventional thickeners to achieve the desired degree of thickening of emulsion polymer latexes. In addition, the resulting thickened latexes have better flow, better leveling, better scrub, and better spatter resistance than cellulosic and polyacrylic thickeners. Moreover, the thickened latexes have better ageing stability than urethane associative thickeners.

The invention will be illustrated but not limited by the following examples.

10

## EXAMPLES

### Example 1

57 millimoles of tridecyl alcohol ethoxylate (100 mole average) and 19 millimoles of PEG-8000 were added to a one liter reactor. 300 grams of toluene were then added and the mixture was heated until it melted. 95 millimoles of 50% aqueous sodium hydroxide were added to the melt, and the mixture heated to reflux to drive off water. When the water level in the mixture was below 0.2%, 86 millimoles of epichlorohydrin were added. The reaction was then carried out at 90-100°C until epoxide titration showed the absence of oxirane groups. The toluene was distilled off under vacuum and the residue was diluted with 15% by weight of butyl carbitol as a viscosity reducing cosolvent, and sufficient water to produce a solution containing 40%

polymer solids.

### Comparative Example 1

The procedure of Example 1 was repeated except that 57 millimoles of  
 5 epichlorohydrin, 400 grams of water, and 200 grams of butyl carbitol were  
 used.

The products of the above examples were then tested in a 0.35 micron  
 vinyl-acrylic hydrophilic latex water-based paint. The results are shown in  
 TABLE I below.

10

TABLE I

Example	Ratio <sup>1</sup> ECH/OH	Gms <sup>2</sup> /Pint Paint	ICI <sup>3</sup>	KU <sup>4</sup>
1	0.9	1.0	0.9	75
1	0.9	2.0	2.0	92
Comp. 1	0.6	2.0	0.2	<53
Comp. 1	0.6	10.0	1.0	53

15

- 20
- 1 - ratio of moles of epichlorohydrin to the OH equivalents of tridecyl alcohol ethoxylate plus PEG-8000.
  - 2 - grams of water/cosolvent/product mixture
  - 3 - ICI is a viscosity measurement for paint to measure its brush drag.  
 The measurement (poise units) is carried out on a cone and plate

viscometer at a shear rate of 8,000-10,000  $\text{sec}^{-1}$ .

- 4 - KU is a low shear viscosity measurement at 100  $\text{sec}^{-1}$  for paint to measure its stirrability in Krebs Units (KU).

5

### Example 2

59.7 millimoles of cetyl/stearyl alcohol ethoxylate (100 mole ethoxy average) and 15 millimoles of PEG-8000 were added to a one liter reactor. 400 grams of toluene were then added and the mixture was heated until it melted. 90 millimoles of 50% aqueous sodium hydroxide were added and the mixture heated to reflux to drive off water. When the water level in the mixture was below 0.2%, 65.3 millimoles of epichlorohydrin were added. The reaction was then carried out at 90-100°C until epoxide titration showed the absence of oxirane groups. The toluene was distilled off under vacuum and the residue was diluted with 15% by weight of butyl carbitol as a viscosity reducing cosolvent, and sufficient water to produce a solution containing 30% polymer solids.

### Examples 3-7

20 The process of Example 2 was repeated except that the quantities of components set forth in Table II were used in these examples.

TABLE II

Example	mmol cetyl/stearyl alcohol ethoxylate	mmol PEG-8000	Toluene, grams	mmol NaOH	mmol epichloro- hydrin
3	73.1	18.2	400	110	79.2
4	70	10.5	350	91	73
5	78.8	8	350	105	86
6	68.5	10.3	350	98	71.3
7	63.7	12.8	350	90	64.3

Example 8

10 The water/cosolvent/product mixtures obtained in Examples 2 through 7 were added to contractors' paint in the quantities shown in TABLE III below, and the product viscosities (in cps) and the initial and equilibrium ICI and KU values were measured and are also given in TABLE III.

TABLE III

Example	Ethoxylated alcohol	% PEG- 8000 in product	ECH/OH ratio	Product viscosity <sup>2</sup> (cps)	lbs mixture in 100 gal paint	Initial ICI/KU	Equilibrium ICI/KU
2	cetyl/stearyl	25	0.72	2200	6.44	0.8/91	0.8/93
3	cetyl/stearyl	25	0.75	1920	7.40	1.3/98	1.1/96
4	cetyl/stearyl	15	0.8	1880	6.35	0.7/92	0.6/91
5	C <sub>16</sub> Guerbet	10	0.9	1500	7.29 <sup>1</sup>	1.0/89	1.0/89
6	C <sub>20</sub> Guerbet	10	0.85	740	6.34 <sup>1</sup>	1.0/71	1.0/73
7	C <sub>20</sub> Guerbet	20	0.7	4200	4.99 <sup>1</sup>	0.5/91	0.5/90

1 - extra butyl carbitol added to disperse thickener in paint

2 - Brookfield viscosity (spindle #3, at 30 r.p.m.)

What is claimed is:

1. The branched reaction product of

A) a linking compound of formula I



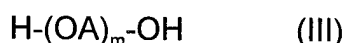
wherein each X group is a halogen atom or one X group is a halogen atom and two X groups together with two carbon atoms from the R<sup>1</sup> group and an oxygen atom form an epoxy group and R<sup>1</sup> is an alkanetriyl group containing from 3 to 10 carbon atoms;

B) a compound of formula II



wherein R<sup>2</sup> is an aliphatic group containing from 4 to 36 carbon atoms, n is a number of from 1 to 200, and each OA group is independently an ethyleneoxy, 1,2 -propyleneoxy, or 1,2-butyleneoxy group; and

C) a compound of formula III



wherein OA is as defined above, and m is a number of from 1 to 500; wherein the ratio of moles of component A) to the sum of the OH equivalents in components B) plus C) is from 0.7:1 to 0.99:1.

2. The product of claim 1 wherein said ratio is from 0.7:1 to 0.9:1.

3. The product of claim 1 wherein component A) is epichlorohydrin.

4. The product of claim 1 wherein component A) is a trichloroalkane.

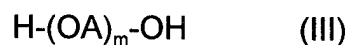
5. The product of claim 1 wherein in component B) all OA groups are ethyleneoxy groups.
6. The product of claim 1 wherein in component C) all OA groups are ethyleneoxy groups.
7. The product of claim 1 wherein in component B) the R<sup>2</sup> group is a straight or branched alkyl group containing from 16 to 20 carbon atoms.
8. The product of claim 5 wherein in component B) the R<sup>2</sup> group is a C<sub>20</sub> Guerbet alkyl group.
9. The product of claim 1 wherein in component B), n is a number of from 80 to 120.
10. The product of claim 9 wherein n is a number of from 80 to 100.
11. The product of claim 1 wherein in component B), the R<sup>2</sup> group contains from 12 to 20 carbon atoms.
12. The product of claim 1 wherein in component C), m is a number of from 50 to 500.
13. The product of claim 1 wherein the product is in the form of an aqueous liquid.
14. The product of claim 1 wherein the product is in the form of an aqueous liquid; in component B) the R<sup>2</sup> group is a straight or branched alkyl group containing from 16 to 20 carbon atoms and n is a number of from 80 to 120; and in component C) m is a number of from 50 to 500.
15. The product of claim 14 wherein the ratio of moles of A) to the sum of the OH equivalents in B) + C) is in the range of 0.7:1 to 0.9:1.
16. The product of claim 14 wherein component A) is epichlorohydrin.

17. The product of claim 14 wherein in components B) and C) all OA groups are ethyleneoxy groups.
18. The product of claim 17 wherein n is a number of from 80 to 100.
19. The product of claim 1 in the form of an aqueous liquid having a solids content of from about 15 to about 40% by weight.
20. The aqueous liquid of claim 19 wherein the aqueous liquid also contains a glycol ether cosolvent.
21. The aqueous liquid of claim 19 which also contains a surfactant.
22. In an aqueous composition, the improvement wherein a thickening-effective quantity of the reaction product of claim 1 is present therein.
23. In a latex paint, the improvement wherein a thickening-effective quantity of the reaction product of claim 1 is present therein.
24. A process comprising the steps of

- i) reacting a compound of formula II

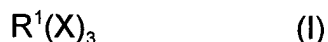


wherein  $R^2$  is an aliphatic group containing from 4 to 36 carbon atoms, n is a number of from 1 to 200, and each OA group is independently an ethyleneoxy, 1,2 -propyleneoxy, or 1,2-butyleneoxy group; and a compound of formula III



wherein OA is as defined above, and m is a number of from 1 to 500, in an organic solvent that azeotropes water, in the presence of an alkali metal alkoxide or hydroxide, and under azeotropic distillation conditions to remove water, to obtain a reaction mixture containing reaction product;

- ii) adding to the reaction mixture from step i) a linking compound of formula I



wherein each X group is a halogen atom or one X group is a halogen atom and two X groups together with two carbon atoms from the R<sup>1</sup> group and an oxygen atom form an epoxy group and R<sup>1</sup> is an alkanetriyl group containing from 3 to 10 carbon atoms, and reacting the compound of formula I with the reaction mixture from step i);

- iii) removing the organic solvent from the resulting reaction product; and
- iv) adding water to said reaction product to form an aqueous solution or dispersion having a solids content of from about 15 to about 40% by weight.

25. The process of claim 24 wherein the organic solvent is benzene, toluene, xylene, or mixtures thereof.

26. The process of claim 24 wherein the reaction mixture from step A) contains no more than 0.2% by weight of water.

27. The process of claim 24 wherein step II) is carried out at a temperature of from about 60°C to the reflux temperature of the organic solvent.

28. The process of claim 27 wherein said temperature is in the range of from about 100 to about 110°C.

29. The process of claim 24 wherein in step IV the solids content is from about 20 to about 40% by weight.

30. The process of claim 24 wherein a cosolvent is also added to reduce

the viscosity of the aqueous solution.

31. The process of claim 30 wherein the cosolvent is a glycol ether.
32. The process of claim 24 wherein a surfactant is added to the aqueous solution from step IV.
33. The process of claim 24 wherein in step III the organic solvent is removed by vacuum distillation.
34. The process of claim 24 wherein in step I from 1 equivalent to 1.5 equivalents of an alkali metal hydroxide are present per equivalent of hydroxy groups.
35. The product of the process of claim 24.

INTERNATIONAL SEARCH REPORT

International application No.  
PCT/US98/11393

**A. CLASSIFICATION OF SUBJECT MATTER**

IPC(6) :C08K 5/00, 5/06  
US CL :524/376, 377, 378

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)

U.S. : 524/376, 377, 378

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 practicable, search terms used)

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	Database WPIDS, London: Derwent Publications Ltd., AN 92-209431, EP 491232 A1, (BADI) BASF AG, "High mol. wt. polyether ol(s) - obtd. by reaction of cpds. contg. active hydrogen atoms, epoxide(s), etc.", abstract, 24 June 1992, Vol. 26, see the entire abstract.	1-35

Further documents are listed in the continuation of Box C.  See patent family annex.

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