

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

(19) World Intellectual Property

Organization

International Bureau

(43) International Publication Date

02 September 2021 (02.09.2021)



(10) International Publication Number

WO 2021/171908 A1

(51) International Patent Classification:

A61K 8/55 (2006.01) A61K 8/81 (2006.01)

A61K 8/73 (2006.01) A61Q 5/00 (2006.01)

(21) International Application Number:

PCT/JP2021/003438

(22) International Filing Date:

26 January 2021 (26.01.2021)

(25) Filing Language:

English

(26) Publication Language:

English

(30) Priority Data:

2020-030603 26 February 2020 (26.02.2020) JP

2002953 26 March 2020 (26.03.2020) FR

(71) Applicant (for all designated States except AL): L'OREAL

[FR/FR]; 14 RUE ROYALE, 75008 PARIS (FR).

(72) Inventors; and

(71) Applicants (for AL only): MARLIAC Marie-Ade-

line [FR/JP]; c/o NIHON L'OREAL K.K., KSP R&D,

3-2-1, Sakado, Takatsu-ku, Kawasaki-shi, Kanagawa,

2130012 (JP). YAMAMOTO Mariko [JP/JP]; c/o NIHON

L'OREAL K.K., KSP R&D, 3-2-1, Sakado, Takatsu-ku,

Kawasaki-shi, Kanagawa, 2130012 (JP). DU Yao [CN/JP];

c/o NIHON L'OREAL K.K., KSP R&D, 3-2-1, Sakado,

Takatsu-ku, Kawasaki-shi, Kanagawa, 2130012 (JP). ISO-

JIMA Tatsushi [JP/JP]; c/o NIHON L'OREAL K.K., KSP

R&D, 3-2-1, Sakado, Takatsu-ku, Kawasaki-shi, Kana-

gawa, 2130012 (JP).

(74) Agent: MURAYAMA Yasuhiko; 1-9-2, Marunouchi,

Chiyoda-ku, Tokyo, 1006620 (JP).

(81) Designated States (unless otherwise indicated, for every

kind of national protection available): AE, AG, AL, AM,

AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ,

CA, CH, CL, CN, CO, CR, CU, CZ, DE, DJ, DK, DM, DO,

DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN,

HR, HU, ID, IL, IN, IR, IS, IT, JO, KE, KG, KH, KN, KP,

KR, KW, KZ, LA, LC, LK, LR, LS, LU, LY, MA, MD, ME,

MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ,

OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SA,

SC, SD, SE, SG, SK, SL, ST, SV, SY, TH, TJ, TM, TN, TR,

TT, TZ, UA, UG, US, UZ, VC, VN, WS, ZA, ZM, ZW.

(84) Designated States (unless otherwise indicated, for every

kind of regional protection available): ARIPO (BW, GH,

(54) Title: COMPOSITION COMPRISING POLYION COMPLEX AND FATTY ALCOHOL

(57) Abstract: The present invention relates to a composition, preferably a cosmetic composition, and more preferably a cosmetic composition for keratin fibers such as hair, comprising: (a) at least one polyion complex comprising at least one ionic polymer selected from the group consisting of cationic polymers, anionic polymers, amphoteric polymers, and mixtures thereof, and at least one crosslinker selected from the group consisting of non-polymeric acids having two or more pKa values or salt(s) thereof, non-polymeric bases having two or more pKb values or salt(s) thereof, and mixtures thereof; (b) at least one fatty alcohol; and (c) water. The composition according to the present invention can provide keratin fibers with improved textures in terms of all of smoothness, softness, evenness of coating and tip moisturization.



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## DESCRIPTION

## TITLE OF INVENTION

5 COMPOSITION COMPRISING POLYION COMPLEX AND FATTY ALCOHOL

## TECHNICAL FIELD

10 The present invention relates to a composition, preferably a cosmetic composition, and more preferably a cosmetic composition for keratin fibers such as hair, which includes a polyion complex.

## BACKGROUND ART

15 A polyion complex, which is formed with an anionic polymer and a cationic polymer, has already been known.

For example, WO 2017/104221 discloses a composition which is useful for cosmetic treatments and comprises at least one polyion complex particle comprising at least one cationic polymer, at least one anionic polymer and at least one non-polymeric acid having two or more pKa values.

Also, WO 2018/230673 discloses a composition which includes such a polyion complex particle and an oil, which may further include an oil-gelling agent. The oil-gelling agent is used to enhance the stability of the composition.

## DISCLOSURE OF INVENTION

It has been discovered that when a composition including a polyion complex is used for cosmetically treating keratin fibers such as hair, textures such as smoothness, softness, evenness of coating, and tips moisturization of the treated keratin fibers need to be improved.

Thus, an objective of the present invention is to provide a composition including a polyion complex, which can provide keratin fibers such as hair with improved textures in terms of all of smoothness, softness, evenness of coating and tip moisturization.

The above objective of the present invention can be achieved by a composition, preferably a cosmetic composition, and more preferably a cosmetic composition for keratin fibers such as hair, comprising:

- 40 (a) at least one polyion complex comprising  
at least one ionic polymer selected from the group consisting of cationic polymers, anionic polymers, amphoteric polymers, and mixtures thereof,  
and  
at least one crosslinker selected from the group consisting of non-polymeric acids having two or more pKa values or salt(s) thereof, non-polymeric bases having two or more pKb values or salt(s) thereof, and mixtures thereof;
- 45 (b) at least one fatty alcohol; and  
(c) water.

50 The (a) polyion complex(es) may comprise

- (i) at least one cationic polymer and at least one non-polymeric acid having two or more pKa values or salt(s) thereof;
- (ii) at least one cationic polymer, at least one anionic polymer, and at least one non-polymeric acid having two or more pKa values or salt(s) thereof and/or at least one non-polymeric base having two or more pKb values or salt(s) thereof,
- (iii) at least one cationic polymer, at least one amphoteric polymer, and at least one non-polymeric acid having two or more pKa values or salt(s) thereof and/or at least one non-polymeric base having two or more pKb values or salt(s) thereof,
- (iv) at least one cationic polymer, at least one anionic polymer, at least one amphoteric polymer, and at least one non-polymeric acid having two or more pKa values or salt(s) thereof and/or at least one non-polymeric base having two or more pKb values or salt(s) thereof,
- (v) at least one anionic polymer and at least one non-polymeric base having two or more pKb values or salt(s) thereof,
- (vi) at least one anionic polymer, at least one amphoteric polymer, and at least one non-polymeric acid having two or more pKa values or salt(s) thereof and/or at least one non-polymeric base having two or more pKb values or salt(s) thereof, or
- (vii) at least one amphoteric polymer and at least one non-polymeric acid having two or more pKa values or salt(s) thereof and/or at least one non-polymeric base having two or more pKb values or salt(s) thereof.

The cationic polymer may be selected from the group consisting of polyquaternium-4, polyquaternium-6, polyquaternium-7, polyquaternium-10, polyquaternium-24, polyquaternium-67, and a mixture thereof.

The amount of the ionic polymer(s) in the composition according to the present invention may be from 0.01% to 10% by weight, preferably from 0.05% to 5% by weight, and more preferably from 0.1% to 1% by weight, relative to the total weight of the composition.

The non-polymeric acid having two or more pKa values or salt(s) thereof may be selected from the group consisting of terephthalylidene dicamphor sulfonic acid and salts thereof (Mexoryl SX), Yellow 6 (Sunset Yellow FCF), ascorbic acid, phytic acid, citric acid, tartaric acid, and salts thereof, and a mixture thereof.

The amount of the crosslinker(s) in the composition according to the present invention may be from 0.001% to 10% by weight, preferably from 0.005% to 5% by weight, and more preferably from 0.01% to 1% by weight, relative to the total weight of the composition.

The (b) fatty alcohol may have a structure R-OH wherein R is chosen from saturated and unsaturated, linear and branched radicals containing from 8 to 40 carbon atoms.

The amount of the (b) fatty alcohol in the composition according to the present invention may be from 0.1% to 15% by weight, preferably from 0.5% to 10% by weight, and more preferably from 1% to 5% by weight, relative to the total weight of the composition.

The amount of the (c) water in the composition according to the present invention may be from 50% to 99% by weight, preferably from 60% to 97% by weight, and more preferably from 70% to 95% by weight, relative to the total weight of the composition.

The pH of the composition according to the present invention may be from 3 to 9, preferably from 3.5 to 8.5, and more preferably from 4 to 8.

5 The composition according to the present invention may further comprise (d) at least one fatty material other than the (b) fatty alcohol, preferably at least one oil and/or at least one wax.

10 The composition according to the present invention may further comprise (e) at least one texture agent, preferably selected from hydrophilic nonionic polymeric thickeners, and more preferably selected from hydrophilic nonionic polymeric associative thickeners and hydrophilic nonionic polysaccharide thickeners.

15 The present invention also relates to a cosmetic process for keratin fibers such as hair, comprising applying to the keratin fibers the composition according to the present invention; and drying the composition to form a cosmetic film on the keratin fibers.

20 The present invention also relates to a process for preparing a film, preferably a cosmetic film, comprising: applying onto keratin fibers such as hair, the composition according to the present invention; and drying the composition.

The present invention also relates to a film, preferably a cosmetic film, comprising:  
25 (a) at least one polyion complex comprising at least one ionic polymer selected from the group consisting of cationic polymers, anionic polymers, amphoteric polymers, and mixtures thereof, and at least one crosslinker selected from the group consisting of non-polymeric acids having two or more pKa values or salt(s) thereof, non-polymeric bases having two or  
30 more pKb values or salt(s) thereof, and mixtures thereof; and  
(b) at least one fatty alcohol.

### BEST MODE FOR CARRYING OUT THE INVENTION

35 After diligent research, the inventors have discovered that it is possible to provide a composition which includes a polyion complex and can provide keratin fibers such as hair with improved textures in terms of all of smoothness, softness, evenness of coating and tip moisturization.

40 The term “evenness of coating” refers to a feeling to touch such that keratin fibers such as hair are evenly coated with a film formed by the composition according to the present invention.

The term “tip moisturization” refers to a feeling to touch such that the tips of the keratin fibers such as hair are moisturized.

45 Thus, the composition according to the present invention comprises:  
(a) at least one polyion complex comprising at least one ionic polymer selected from the group consisting of cationic polymers, anionic polymers, amphoteric polymers, and mixtures thereof,  
50 and

at least one crosslinker selected from the group consisting of non-polymeric acids having two or more pKa values or salt(s) thereof, non-polymeric bases having two or more pKb values or salt(s) thereof, and mixtures thereof;

(b) at least one fatty alcohol; and

5 (c) water.

It may be preferable that the (a) polyion complex is in the form of a particle.

10 The composition according to the present invention can provide keratin fibers such as hair with improved textures with regard to all of smoothness, softness, evenness of coating and tip moisturization. Even for dried keratin fibers, the composition can provide them with tip moisturization.

15 It is preferable that the composition according to the present invention is intended for cosmetically treating keratin fibers such as hair. The composition according to the present invention can preferably be used as a cosmetic composition of the leave-on type or rinse-off type for keratin fibers such as hair.

20 The composition according to the present invention is, in particular, useful for caring keratin fibers such as hair.

Hereinafter, the composition and the like according to the present invention will be explained in a more detailed manner.

25 [Polyion Complex]

The composition according to the present invention includes (a) at least one polyion complex. Two or more different types of (a) polyion complexes may be used in combination. Thus, a single type of (a) polyion complex or a combination of different types of (a) polyion complexes may be used.

30 It may be preferable that the (a) polyion complex is in the form of a particle. The size of the polyion complex particle may be from 5 nm to 100  $\mu\text{m}$ , preferably from 100 nm to 50  $\mu\text{m}$ , more preferably from 200 nm to 40  $\mu\text{m}$ , and even more preferably from 500 nm to 30  $\mu\text{m}$ . A particle size less than 1  $\mu\text{m}$  can be measured by a dynamic light scattering method, and a particle size more than 1  $\mu\text{m}$  can be measured by an optical microscope. This particle size may be based on a number-average diameter.

40 The amount of the (a) polyion complex(es) in the composition according to the present invention may be 0.01% by weight or more, preferably 0.05% by weight or more, and more preferably 0.1% by weight or more, relative to the total weight of the composition.

45 The amount of the (a) polyion complex(es) in the composition according to the present invention may be 25% by weight or less, preferably 20% by weight or less, and more preferably 15% by weight or less, relative to the total weight of the composition.

50 The amount of the (a) polyion complex(es) in the composition according to the present invention may be from 0.01% to 25% by weight, preferably from 0.05% to 20% by weight, and more preferably from 0.1% to 15% by weight, relative to the total weight of the composition.

It may be preferable that the positive charge of the (a) polyion complex is larger than the amount of negative charge of the (a) polyion complex so that the total or net charge of the (a) polyion complex be positive. In other words, it may be preferable that the (a) polyion complex, as a whole, be positively charged.

The (a) polyion complex can be formed by at least one ionic polymer and at least one crosslinker. The ionic polymer has positive and/or negative charges. Also, the crosslinker may have positive or negative charge(s). In any event, it may be preferable that the amount of positive charge of the (a) polyion complex is larger than the amount of negative charge of the (a) polyion complex. Thus, it may be preferable that the total or net charge of the (a) polyion complex formed by the ionic polymer and the crosslinker, after offsetting or counterbalancing the positive and negative charges provided by the ionic polymer and the crosslinker, be positive, i.e., above zero, preferably 0.1 or more, more preferably 0.3 or more, and even more preferably 0.5 or more, and preferably 10 or less, more preferably 5 or less, and even more preferably 3 or less.

The ratio of the cation/anion on the (a) polyion complex(es) in the composition according to the present invention may be from 1.01 to 5 which can be determined by the ratio of cation equivalent/g divided by anion equivalent/g, preferably from 1.05 to 4, and more preferably from 1.1 to 3, even more preferably from 1.5 to 2.

The (a) polyion complex in the form of a particle may be present at the interface between a fatty phase comprising the (b) fatty alcohol (and, if present, the (d) fatty material such as oil) and the (c) water. Thus, the (a) polyion complex may form, for instance, an emulsion. For example, if the (c) water constitutes a continuous phase and the fatty phases constitute dispersed phases, the (a) polyion complex can form an O/W emulsion which may be similar to a so-called Pickering emulsion.

Alternatively, the (a) polyion complex may form a capsule having a hollow. The fatty phase comprising the (b) fatty alcohol (and, if present, the (d) fatty material such as oil) may be present in the hollow. In other words, the fatty phase can be incorporated into the capsule. The wall of the capsule may be composed of a continuous layer or film formed from the (a) polyion complex. While not wishing to be bound by theory, it is believed that the (a) polyion complex can re-organize at the interface of the fatty phase and the (c) water to spontaneously form a capsule having a hollow to include the fatty phase. For example, a continuous phase constituted with the (c) water and dispersed phases constituted with the fatty phases in the capsule can form an O/W emulsion which may also be similar to a so-called Pickering emulsion.

The above would mean that the (a) polyion complex itself is amphiphilic and insoluble in fatty material such as oil, or water.

{Ionic Polymer and Crosslinker}

The (a) polyion complex includes at least one ionic polymer selected from the group consisting of cationic polymers, anionic polymers, amphoteric polymers, and mixtures thereof, and

at least one crosslinker selected from the group consisting of non-polymeric acids having two or more pKa values or salt(s) thereof, non-polymeric bases having two or more pKb values or salt(s) thereof, and mixtures thereof.

5 In a preferable embodiment, the (a) polyion complex comprises:

- (i) at least one cationic polymer and at least one non-polymeric acid having two or more pKa values or salt(s) thereof;
- (ii) at least one cationic polymer, at least one anionic polymer, and at least one non-polymeric acid having two or more pKa values or salt(s) thereof and/or at least one  
10 non-polymeric base having two or more pKb values or salt(s) thereof,
- (iii) at least one cationic polymer, at least one amphoteric polymer, and at least one non-polymeric acid having two or more pKa values or salt(s) thereof and/or at least one non-polymeric base having two or more pKb values or salt(s) thereof,
- (iv) at least one cationic polymer, at least one anionic polymer, at least one amphoteric  
15 polymer, and at least one non-polymeric acid having two or more pKa values or salt(s) thereof and/or at least one non-polymeric base having two or more pKb values or salt(s) thereof,
- (v) at least one anionic polymer and at least one non-polymeric base having two or more pKb values or salt(s) thereof,
- (vi) at least one anionic polymer, at least one amphoteric polymer, and at least one non-polymeric acid having two or more pKa values or salt(s) thereof and/or at least one non-polymeric base having two or more pKb values or salt(s) thereof, or  
20 (vii) at least one amphoteric polymer and at least one non-polymeric acid having two or more pKa values or salt(s) thereof and/or at least one non-polymeric base having two or more pKb values or salt(s) thereof.  
25

There is no limit to the type of the cationic, anionic and amphoteric polymers. Two or more different types of cationic polymers may be used in combination. Thus, a single type of cationic polymer or a combination of different types of cationic polymers may be used. Two  
30 or more different types of anionic polymers may be used in combination. Thus, a single type of anionic polymer or a combination of different types of anionic polymers may be used. Two or more different types of amphoteric polymers may be used in combination. Thus, a single type of amphoteric polymer or a combination of different types of amphoteric polymers may be used.  
35

In the above component (ii), the ratio of the amount, for example the chemical equivalent, of the cationic polymer(s)/the anionic polymer(s) may be 0.05-18, preferably 0.1-10, and more preferably 0.5-5.0. In particular, it may be preferable that the number of the cationic groups of the cationic polymer(s)/the number of anionic groups of the anionic polymer(s) be 0.05-18,  
40 more preferably 0.1-10; and even more preferably 0.5-5.0.

In the above component (iii), the ratio of the amount, for example chemical equivalent, of the cationic polymer(s)/the amphoteric polymer(s) may be 0.05-18, preferably 0.1-10, and more preferably 0.5-5.0. In particular, it may be preferable that the number of the cationic groups of the cationic polymer(s)/the number of cationic and anionic groups of the amphoteric polymer(s) be 0.05-18, more preferably 0.1-10, and even more preferably 0.5-5.0.  
45

In the above component (vi), the ratio of the amount, for example chemical equivalent, of the anionic polymer(s)/the amphoteric polymer(s) may be 0.05-18, preferably 0.1-10, and more preferably 0.5-5.0. In particular, it may be preferable that the number of the anionic groups of  
50

the anionic polymer(s)/the number of cationic and anionic groups of the amphoteric polymer(s) be 0.05-18, more preferably 0.1-10, and even more preferably 0.5-5.0.

5 The amount of the ionic polymer(s) in the composition according to the present invention may be 0.01% by weight or more, preferably 0.05% by weight or more, and more preferably 0.1% by weight or more, relative to the total weight of the composition.

10 The amount of the ionic polymer(s) in the composition according to the present invention may be 10% by weight or less, preferably 5% by weight or less, and more preferably 1% by weight or less, relative to the total weight of the composition.

15 The amount of the ionic polymer(s) in the composition according to the present invention may be from 0.01% to 10% by weight, preferably from 0.05% to 5% by weight, and more preferably from 0.1% to 1% by weight, relative to the total weight of the composition.

The amount of the crosslinker(s) in the composition according to the present invention may be 0.001% by weight or more, preferably 0.005% by weight or more, and more preferably 0.01% by weight or more, relative to the total weight of the composition.

20 The amount of the crosslinker(s) in the composition according to the present invention may be 10% by weight or less, preferably 5% by weight or less, and more preferably 1% by weight or less, relative to the total weight of the composition.

25 The amount of the crosslinker(s) in the composition according to the present invention may be from 0.001% to 10% by weight, preferably from 0.005% to 5% by weight, and more preferably from 0.01% to 1% by weight, relative to the total weight of the composition.

(Cationic Polymer)

30 A cationic polymer has a positive charge density. The charge density of the cationic polymer may be from 0.01 meq/g to 20 meq/g, preferably from 0.05 to 15 meq/g, and more preferably from 0.1 to 10 meq/g.

35 The molecular weight of the cationic polymer may be 1000 or more, preferably 50000 or more, more preferably 100000 or more, and even more preferably 1000000 or more.

Unless otherwise defined in the descriptions, "molecular weight" means a number-average molecular weight.

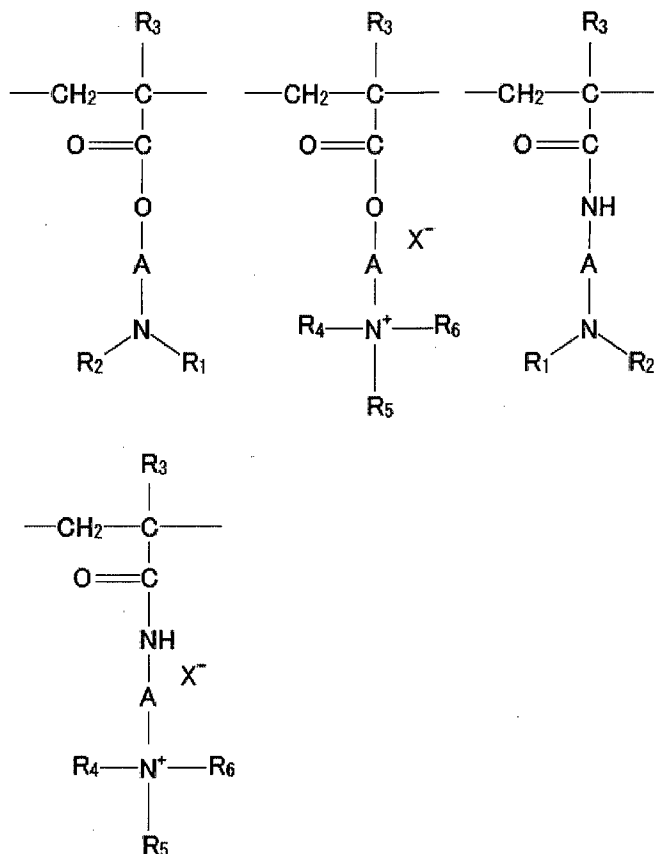
40 The cationic polymer may have at least one positively chargeable and/or positively charged moiety selected from the group consisting of a primary, secondary or tertiary amino group, a quaternary ammonium group, a guanidine group, a biguanide group, an imidazole group, an imino group, and a pyridyl group. The term (primary) "amino group" here means a group of  $-\text{NH}_2$ .

45 The cationic polymer may be a homopolymer or a copolymer. The term "copolymer" is understood to mean both copolymers obtained from two kinds of monomers and those obtained from more than two kinds of monomers, such as terpolymers obtained from three kinds of monomers.

50

The cationic polymer may be selected from natural and synthetic cationic polymers. Non-limiting examples of the cationic polymers are as follows.

- 5 (1) Homopolymers and copolymers derived from acrylic or methacrylic esters and amides and comprising at least one unit chosen from units of the following formulas:



wherein:

- 10  $R_1$  and  $R_2$ , which may be identical or different, are chosen from hydrogen and alkyl groups comprising from 1 to 6 carbon atoms, for instance, methyl and ethyl groups;
- 15  $R_3$ , which may be identical or different, is chosen from hydrogen and  $\text{CH}_3$ ;
- the symbols A, which may be identical or different, are chosen from linear or branched alkyl groups comprising from 1 to 6 carbon atoms, for example, from 2 to 3 carbon atoms and hydroxyalkyl groups comprising from 1 to 4 carbon atoms;
- 15  $R_4$ ,  $R_5$ , and  $R_6$ , which may be identical or different, are chosen from alkyl groups comprising from 1 to 18 carbon atoms and benzyl groups, and in at least one embodiment, alkyl groups comprising from 1 to 6 carbon atoms; and
- X is an anion derived from an inorganic or organic acid, such as methosulphate anions and halides, for instance chloride and bromide.

- 20 The copolymers of family (1) may also comprise at least one unit derived from comonomers which may be chosen from acrylamides, methacrylamides, diacetone acrylamides, acrylamides and methacrylamides substituted on the nitrogen atom with ( $\text{C}_1\text{-C}_4$ ) lower alkyl groups, groups derived from acrylic or methacrylic acids and esters thereof, vinyl lactams such as vinylpyrrolidone and vinylcaprolactam, and vinyl esters.

- 25 Examples of copolymers of family (1) include, but are not limited to:

copolymers of acrylamide and of dimethylaminoethyl methacrylate quaternized with dimethyl sulphate or with a dimethyl halide,  
copolymers of acrylamide and of methacryloyloxyethyltrimethylammonium chloride described, for example, in European Patent Application No. 0 080 976,  
5 copolymers of acrylamide and of methacryloyloxyethyltrimethylammonium methosulphate, quaternized or nonquaternized vinylpyrrolidone/dialkylaminoalkyl acrylate or methacrylate copolymers, described, for example, in French Patent Nos. 2 077 143 and 2 393 573,  
dimethylaminoethyl methacrylate/vinylcaprolactam/vinylpyrrolidone terpolymers,  
10 vinylpyrrolidone/methacrylamidopropyltrimethylamine copolymers, quaternized vinylpyrrolidone/dimethylaminopropylmethacrylamide copolymers, and  
crosslinked methacryloyloxy(C<sub>1</sub>-C<sub>4</sub>)alkyltri(C<sub>1</sub>-C<sub>4</sub>)alkylammonium salt polymers such as the polymers obtained by homopolymerization of dimethylaminoethyl methacrylate quaternized  
with methyl chloride, or by copolymerization of acrylamide with dimethylaminoethyl  
15 methacrylate quaternized with methyl chloride, the homopolymerization or copolymerization being followed by crosslinking with a compound containing an olefinic unsaturation, for  
example, methylenebisacrylamide.

(2) Cationic cellulose derivatives such as cellulose ether derivatives comprising quaternary ammonium groups described, for example, in French Patent No. 1 492 597, such as the  
20 polymers sold under the names "JR" (JR 400, JR 125, JR 30M) or "LR" (LR 400, LR 30M) by the company Union Carbide Corporation. These polymers are also defined in the CTFA dictionary as quaternary ammoniums of hydroxyethylcellulose that have reacted with an epoxide substituted with a trimethylammonium group.

(3) Cationic cellulose derivatives such as cellulose copolymers and cellulose derivatives grafted with a water-soluble monomer of quaternary ammonium, and described, for example,  
25 in U.S. Pat. No. 4,131,576, such as hydroxyalkylcelluloses, for instance, hydroxymethyl-, hydroxyethyl-, and hydroxypropylcelluloses grafted, for example, with a salt chosen from methacryloylethyltrimethylammonium, methacrylamidopropyltrimethylammonium, and  
30 dimethyldiallylammonium salts. Commercial products corresponding to these polymers include, for example, the products sold under the name "Celquat® L 200" and "Celquat® H 100" by the company National Starch.

(4) Non-cellulose-based cationic polysaccharides described in U.S. Pat. Nos. 3,589,578 and  
35 4,031,307, such as guar gums comprising cationic trialkylammonium groups, cationic hyaluronic acid, and dextran hydroxypropyl trimonium chloride. Guar gums modified with a salt, for example the chloride, of 2,3-epoxypropyltrimethylammonium (guar hydroxypropyltrimonium chloride) may also be used.

40 Such products are sold, for instance, under the trade names JAGUAR® C13 S, JAGUAR® C15, JAGUAR® C17, and JAGUAR® C162 by the company MEYHALL.

(5) Polymers comprising piperazinyl units and divalent alkylene or hydroxyalkylene groups comprising straight or branched chains, optionally interrupted with at least one entity chosen  
45 from oxygen, sulphur, nitrogen, aromatic rings, and heterocyclic rings, and also the oxidation and/or quaternization products of these polymers. Such polymers are described, for example, in French Patent Nos. 2 162 025 and 2 280 361.

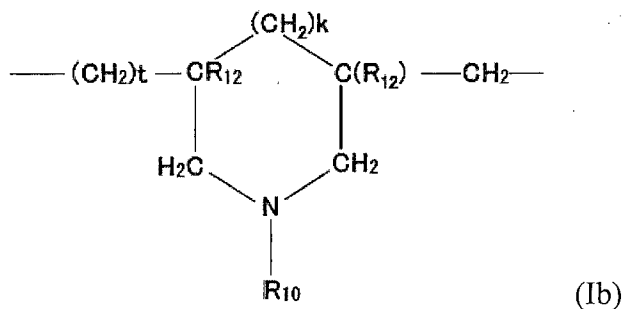
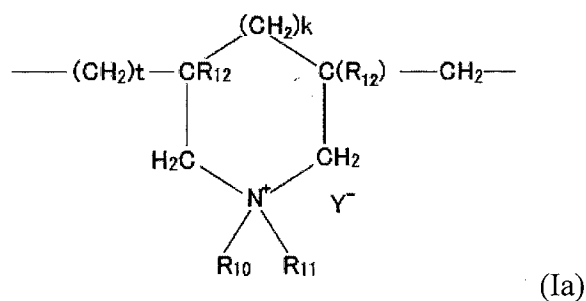
(6) Water-soluble polyamino amides prepared, for example, by polycondensation of an acidic  
50 compound with a polyamine; these polyamino amides possibly being crosslinked with an

entity chosen from epihalohydrins; diepoxides; dianhydrides; unsaturated dianhydrides; bisunsaturated derivatives; bishalohydrins; bisazetidiniums; bishaloacydiamines; bisalkyl halides; oligomers resulting from the reaction of a difunctional compound which is reactive with an entity chosen from bishalohydrins, bisazetidiniums, bishaloacydiamines, bisalkyl halides, epihalohydrins, diepoxides, and bisunsaturated derivatives; the crosslinking agent being used in an amount ranging from 0.025 to 0.35 mol per amine group of the polyamino amide; these polyamino amides optionally being alkylated or, if they comprise at least one tertiary amine function, they may be quaternized. Such polymers are described, for example, in French Patent Nos. 2 252 840 and 2 368 508.

(7) Polyamino amide derivatives resulting from the condensation of polyalkylene polyamines with polycarboxylic acids, followed by alkylation with difunctional agents, for example, adipic acid/dialkylaminohydroxyalkyldialkylenetriamine polymers in which the alkyl group comprises from 1 to 4 carbon atoms, such as methyl, ethyl, and propyl groups, and the alkylene group comprises from 1 to 4 carbon atoms, such as an ethylene group. Such polymers are described, for instance, in French Patent No. 1 583 363. In at least one embodiment, these derivatives may be chosen from adipic acid/dimethylaminohydroxypropyldiethylenetriamine polymers.

(8) Polymers obtained by reaction of a polyalkylene polyamine comprising two primary amine groups and at least one secondary amine group, with a dicarboxylic acid chosen from diglycolic acid and saturated aliphatic dicarboxylic acids comprising from 3 to 8 carbon atoms. The molar ratio of the polyalkylene polyamine to the dicarboxylic acid may range from 0.8:1 to 1.4:1; the polyamino amide resulting therefrom being reacted with epichlorohydrin in a molar ratio of epichlorohydrin relative to the secondary amine group of the polyamino amide ranging from 0.5:1 to 1.8:1. Such polymers are described, for example, in U.S. Pat. Nos. 3,227,615 and 2,961,347.

(9) Cyclopolymers of alkylallylamine and cyclopolymers of dialkylallyl-ammonium, such as homopolymers and copolymers comprising, as the main constituent of the chain, at least one unit chosen from units of formulas (Ia) and (Ib):



wherein:  
 k and t, which may be identical or different, are equal to 0 or 1, the sum k+t being equal to 1;

R<sub>12</sub> is chosen from hydrogen and methyl groups;

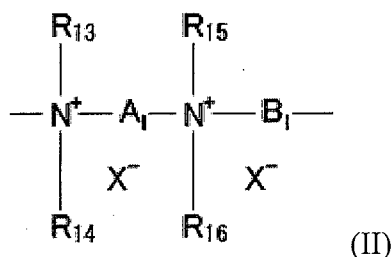
R<sub>10</sub> and R<sub>11</sub>, which may be identical or different, are chosen from alkyl groups comprising from 1 to 6 carbon atoms, hydroxyalkyl groups in which the alkyl group comprises, for example, from 1 to 5 carbon atoms, and lower (C<sub>1</sub>-C<sub>4</sub>)amidoalkyl groups, or R<sub>10</sub> and R<sub>11</sub> may form, together with the nitrogen atom to which they are attached, heterocyclic groups such as piperidinyl and morpholinyl; and

γ is an anion such as bromide, chloride, acetate, borate, citrate, tartrate, bisulphate, bisulphite, sulphate, and phosphate. These polymers are described, for example, in French Patent No. 2 080 759 and in its Certificate of Addition 2 190 406.

In one embodiment, R<sub>10</sub> and R<sub>11</sub>, which may be identical or different, are chosen from alkyl groups comprising from 1 to 4 carbon atoms.

Examples of such polymers include, but are not limited to, (co)polydiallyldialkyl ammonium chloride such as the dimethyldiallylammonium chloride homopolymer sold under the name "MERQUAT® 100" by the company CALGON (and its homologues of low weight-average molecular mass) and the copolymers of diallyldimethylammonium chloride and of acrylamide sold under the name "MERQUAT® 550".

Quaternary diammonium polymers comprising at least one repeating unit of formula (II):



wherein:

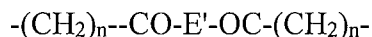
R<sub>13</sub>, R<sub>14</sub>, R<sub>15</sub>, and R<sub>16</sub>, which may be identical or different, are chosen from aliphatic, alicyclic, and arylaliphatic groups comprising from 1 to 20 carbon atoms and lower hydroxyalkyl aliphatic groups, or alternatively R<sub>13</sub>, R<sub>14</sub>, R<sub>15</sub>, and R<sub>16</sub> may form, together or separately, with the nitrogen atoms to which they are attached, heterocycles optionally comprising a second heteroatom other than nitrogen, or alternatively R<sub>13</sub>, R<sub>14</sub>, R<sub>15</sub>, and R<sub>16</sub>, which may be identical or different, are chosen from linear or branched C<sub>1</sub>-C<sub>6</sub> alkyl groups substituted with at least one group chosen from nitrile groups, ester groups, acyl groups, amide groups, -CO-O-R<sub>17</sub>-E groups, and -CO-NH-R<sub>17</sub>-E groups, wherein R<sub>17</sub> is an alkylene group and E is a quaternary ammonium group;

A<sub>1</sub> and B<sub>1</sub>, which may be identical or different, are chosen from polymethylene groups comprising from 2 to 20 carbon atoms, which may be linear or branched, saturated or unsaturated, and which may comprise, linked or intercalated in the main chain, at least one entity chosen from aromatic rings, oxygen, sulphur, sulphoxide groups, sulphone groups, disulphide groups, amino groups, alkylamino groups, hydroxyl groups, quaternary ammonium groups, ureido groups, amide groups, and ester groups, and

X<sup>-</sup> is an anion derived from an inorganic or organic acid;

A<sub>1</sub>, R<sub>13</sub>, and R<sub>15</sub> may form, together with the two nitrogen atoms to which they are attached, a piperazine ring;

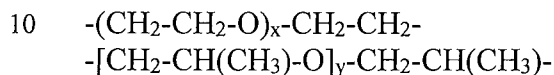
if A<sub>1</sub> is chosen from linear or branched, saturated or unsaturated alkylene or hydroxyalkylene groups, B<sub>1</sub> may be chosen from:



5

wherein E' is chosen from:

a) glycol residues of formula -O-Z-O-, wherein Z is chosen from linear or branched hydrocarbon-based groups and groups of the following formulas:



10

wherein x and y, which may be identical or different, are chosen from integers ranging from 1 to 4, which represent a defined and unique degree of polymerization, and numbers ranging from 1 to 4, which represent an average degree of polymerization;

15

b) bis-secondary diamine residue such as piperazine derivatives;

c) bis-primary diamine residues of formula -NH-Y-NH-, wherein Y is chosen from linear or branched hydrocarbon-based groups and the divalent group -CH<sub>2</sub>-CH<sub>2</sub>-S-S-CH<sub>2</sub>-CH<sub>2</sub>-; and

d) ureylene groups of formula -NH-CO-NH-.

20

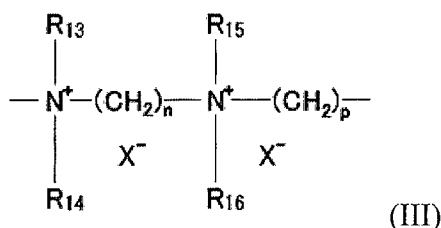
In at least one embodiment, X<sup>-</sup> is an anion such as chloride or bromide.

Polymers of this type are described, for example, in French Patent Nos. 2 320 330; 2 270 846; 2 316 271; 2 336 434; and 2 413 907 and U.S. Pat. Nos. 2,273,780; 2,375,853; 2,388,614; 2,454,547; 3,206,462; 2,261,002; 2,271,378; 3,874,870; 4,001,432; 3,929,990; 3,966,904; 4,005,193; 4,025,617; 4,025,627; 4,025,653; 4,026,945; and 4,027,020.

25

Non-limiting examples of such polymers include those comprising at least one repeating unit of formula (III):

30



wherein

R<sub>13</sub>, R<sub>14</sub>, R<sub>15</sub>, and R<sub>16</sub>, which may be identical or different, are chosen from alkyl and hydroxyalkyl groups comprising from 1 to 4 carbon atoms, n and p, which may be identical or different, are integers ranging from 2 to 20, and X<sup>-</sup> is an anion derived from an inorganic or organic acid.

35

(11) Polyquaternary ammonium polymers comprising units of formula (IV):



As the (co)polyamines, it is preferable to use (co)polylysines. Polylysine is well known. Polylysine can be a natural homopolymer of L-lysine that can be produced by bacterial fermentation. For example, polylysine can be  $\epsilon$ -Poly-L-lysine, typically used as a natural preservative in food products. Polylysine is a polyelectrolyte which is soluble in polar solvents such as water, propylene glycol and glycerol. Polylysine is commercially available in various forms, such as poly D-lysine and poly L-lysine. Polylysine can be in salt and/or solution form.

#### 10 (14) Cationic Polyaminoacids

As the cationic polymer, it may be possible use cationic polyaminoacids, which may be cationic homopolymers or copolymers, with a plurality of amino groups and carboxyl groups. The amino group may be a primary, secondary, tertiary or quaternary amino group. The amino group may be present in a polymer backbone or a pendent group, if present, of the cationic polyaminoacids. The carboxyl group may be present in a pendent group, if present, of the cationic polyaminoacids.

As examples of the cationic polyaminoacids, mention may be made of cationized collagen, cationized gelatin, steardimoiium hydroxypropyl hydrolyzed wheat protein, cocodimonium hydroxypropyl hydrolyzed wheat protein, hydroxypropyltrimonium hydrolyzed conchiolin protein, steardimonium hydroxypropyl hydrolyzed soy protein, hydroxypropyltrimonium hydrolyzed soy protein, cocodimonium hydroxypropyl hydrolyzed soy protein, and the like.

It may be preferable that the cationic polymer be selected from the group consisting of cyclopolymers of alkyldiallylamine and cyclopolymers of dialkyldiallylammonium such as (co)polydiallyldialkyl ammonium chloride, (co)polyamines such as (co)polylysines, cationic (co)polyaminoacids such as cationized collagen, and salts thereof.

It may also be preferable that the cationic polymer be selected from cationic polysaccharides.

The charge density of the cationic polysaccharide may be from 0.01 meq/g to 20 meq/g, preferably from 0.05 to 15 meq/g, and more preferably from 0.1 to 10 meq/g.

The molecular weight of the cationic polysaccharide may be 1000 or more, preferably 50,000 or more, more preferably 100,000 or more, and even more preferably 1,000,000 or more.

Unless otherwise defined in the description, "molecular weight" means a number average molecular weight.

The cationic polysaccharide may have at least one positively chargeable and/or positively charged moiety selected from the group consisting of a primary, secondary or tertiary amino group, a quaternary ammonium group, a guanidine group, a biguanide group, an imidazole group, an imino group, and a pyridyl group. The term (primary) "amino group" here means the group  $-\text{NH}_2$ . It is preferable that the (a) cationic polysaccharide have at least one quaternary ammonium group.

The cationic polysaccharide may be a homopolymer or a copolymer. The term "copolymer" is understood to mean both copolymers obtained from two kinds of monomers and those

obtained from more than two kinds of monomers, such as terpolymers obtained from three kinds of monomers.

The cationic polysaccharide may be selected from natural and synthetic cationic polysaccharides.

It is preferable that the cationic polysaccharide be selected from cationic cellulose polymers. Non-limiting examples of the cationic cellulose polymers are as follows.

(15) Cationic cellulose polymers having at least one quaternary ammonium group comprising at least one fatty chain, such as alkyl, arylalkyl or alkylaryl groups comprising at least 8 carbon atoms. It may be preferable that the cationic cellulose polymers be quaternized hydroxyethyl celluloses modified with at least one quaternary ammonium group comprising at least one fatty chain, such as alkyl, arylalkyl or alkylaryl groups comprising at least 8 carbon atoms, or mixtures thereof. The alkyl radicals borne by the quaternary ammonium group may preferably contain from 8 to 30 carbon atoms, especially from 10 to 30 carbon atoms. The aryl radicals preferably denote phenyl, benzyl, naphthyl or anthryl groups. More preferably, the cationic cellulose polymer may comprise at least one quaternary ammonium group including at least one C<sub>8</sub>-C<sub>30</sub> hydrocarbon group. Examples of quaternized alkylhydroxyethylcelluloses containing C<sub>8</sub>-C<sub>30</sub> fatty chains that may be mentioned include the products Quatrisoft LM 200, Quatrisoft LM-X 529-18-A, Quatrisoft LM-X 529-18B (C12 alkyl) and Quatrisoft LM-X 529-8 (C18 alkyl) or Softcat Polymer SL100, Softcat SX-1300X, Softcat SX-1300H, Softcat SL-5, Softcat SL-30, Softcat SL-60, Softcat SK-MH, Softcat SX-400X, Softcat SX-400H, SoftCat SK-L, Softcat SK-M, and Softcat SK-H, sold by the company Dow Chemical, and the products Crodacel QM, Crodacel, QL (C12 alkyl) and Crodacel QS (C18 alkyl) sold by the company Croda.

It may be even more preferable that the cationic polymer be selected from the group consisting of polyquaternium-4, polyquaternium-6, polyquaternium-7, polyquaternium-10, polyquaternium-24, polyquaternium-67, and a mixture thereof.

The amount of the cationic polymer(s) in the composition according to the present invention may be 0.01% by weight or more, preferably 0.05% by weight or more, and more preferably 0.1% by weight or more, relative to the total weight of the composition.

The amount of the cationic polymer(s) in the composition according to the present invention may be 10% by weight or less, preferably 5% by weight or less, and more preferably 1% by weight or less, relative to the total weight of the composition.

The amount of the cationic polymer(s) in the composition according to the present invention may be from 0.01% to 10% by weight, preferably from 0.05% to 5% by weight, and more preferably from 0.1% to 1% by weight, relative to the total weight of the composition.

(Anionic Polymer)

An anionic polymer has a negative charge density. The charge density of the anionic polymer may be from 0.1 meq/g to 20 meq/g, preferably from 1 to 15 meq/g, and more preferably from 4 to 10 meq/g if the anionic polymer is a synthetic anionic polymer, and the average substitution degree of the anionic polymer may be from 0.1 to 3.0, preferably from 0.2 to 2.7, and more preferably from 0.3 to 2.5 if the anionic polymer is a natural anionic polymer.

The molecular weight of the anionic polymer may be 1,000 or more, preferably 10,000 or more, more preferably 50,000 or more, and even more preferably 100,000 or more.

- 5 The anionic polymer may have at least one negatively chargeable and/or negatively charged moiety selected from the group consisting of a sulfuric group, a sulfate group, a sulfonic group, a sulfonate group, a phosphoric group, a phosphate group, a phosphonic group, a phosphonate group, a carboxylic group, and a carboxylate group.
- 10 The anionic polymer may be a homopolymer or a copolymer. The term "copolymer" is understood to mean both copolymers obtained from two kinds of monomers and those obtained from more than two kinds of monomers, such as terpolymers obtained from three kinds of monomers.
- 15 The anionic polymer may be selected from natural and synthetic anionic polymers.

The anionic polymer may comprise at least one hydrophobic chain.

- 20 The anionic polymer, which may comprise at least one hydrophobic chain, may be obtained by copolymerization of a monomer (a) chosen from carboxylic acids comprising  $\alpha,\beta$ -ethylenic unsaturation (monomer a') and 2-acrylamido-2-methylpropanesulphonic acid (monomer a'') with a non-surface-active monomer (b) comprising an ethylenic unsaturation other than (a) and/or a monomer (c) comprising an ethylenic unsaturation resulting from the reaction of an acrylic monomer comprising an  $\alpha,\beta$ -monoethylenic unsaturation or of an isocyanate monomer
- 25 comprising a monoethylenic unsaturation with a monohydric nonionic amphiphilic component or with a primary or secondary fatty amine.

Thus, the anionic polymer with at least one hydrophobic chain may be obtained by two synthetic routes:

- 30 - either by copolymerization of the monomers (a') and (c), or (a'), (b) and (c), or (a'') and (c), or (a''), (b) and (c),
- or by modification (and in particular esterification or amidation) of a copolymer formed from the monomers (a') or from the monomers (a') and (b), or (a'') and (b), by a monohydric nonionic amphiphilic compound or a primary or secondary fatty amine.

- 35 Mention may in particular be made, as 2-acrylamido-2-methylpropanesulphonic acid copolymers, of those disclosed in the article "Micelle formation of random copolymers of sodium 2-(acrylamido)-2-methylpropanesulfonate and nonionic surfactant macromonomer in water as studied by fluorescence and dynamic light scattering – *Macromolecules*, 2000, Vol. 33, No. 10 – 3694-3704" and in applications EP-A-0 750 899 and EP-A-1 069 172.
- 40

The carboxylic acid comprising an  $\alpha,\beta$ -monoethylenic unsaturation constituting the monomer (a') can be chosen from numerous acids and in particular from acrylic acid, methacrylic acid, crotonic acid, itaconic acid and maleic acid. It is preferably acrylic or methacrylic acid.

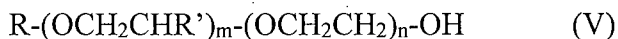
- 45 The copolymer can comprise a monomer (b) comprising a monoethylenic unsaturation which does not have a surfactant property. The preferred monomers are those which give water-insoluble polymers when they are homopolymerized. They can be chosen, for example, from C<sub>1</sub>-C<sub>4</sub> alkyl acrylates and methacrylates, such as methyl acrylate, ethyl acrylate, butyl acrylate
- 50 or the corresponding methacrylates. The more particularly preferred monomers are methyl

acrylate and ethyl acrylate. The other monomers which can be used are, for example, styrene, vinyltoluene, vinyl acetate, acrylonitrile and vinylidene chloride. Unreactive monomers are preferred, these monomers being those in which the single ethylenic group is the only group which is reactive under the polymerization conditions. However, monomers which comprise groups which react under the effect of heat, such as hydroxyethyl acrylate, can optionally be used.

The monomer (c) is obtained by reaction of an acrylic monomer comprising  $\alpha,\beta$ -monoethylenic unsaturation, such as (a), or of an isocyanate monomer comprising monoethylenic unsaturation with a monohydric nonionic amphiphilic compound or a primary or secondary fatty amine.

The monohydric nonionic amphiphilic compounds or the primary or secondary fatty amines used to produce the nonionic monomer (c) are well known. The monohydric nonionic amphiphilic compounds are generally alkoxyated hydrophobic compounds comprising an alkylene oxide forming the hydrophilic part of the molecule. The hydrophobic compounds are generally composed of an aliphatic alcohol or an alkylphenol, in which compounds a carbonaceous chain comprising at least six carbon atoms constitutes the hydrophobic part of the amphiphilic compound.

The preferred monohydric nonionic amphiphilic compounds are compounds having the following formula (V):



in which R is chosen from alkyl or alkylene groups comprising from 6 to 30 carbon atoms and alkylaryl groups having alkyl radicals comprising from 8 to 30 carbon atoms, R' is chosen from alkyl groups comprising from 1 to 4 carbon atoms, n is a mean number ranging from approximately 1 to 150 and m is a mean number ranging from approximately 0 to 50, provided that n is at least as great as m.

Preferably, in the compounds of formula (V), the R group is chosen from alkyl groups comprising from 12 to 26 carbon atoms and alkylphenyl groups in which the alkyl group is C<sub>8</sub>-C<sub>13</sub>; the R' group is the methyl group; m = 0 and n = 1 to 25.

The preferred primary and secondary fatty amines are composed of one or two alkyl chains comprising from 6 to 30 carbon atoms.

The monomer used to form the nonionic urethane monomer (c) can be chosen from highly varied compounds. Use may be made of any compound comprising a copolymerizable unsaturation, such as an acrylic, methacrylic or allylic unsaturation. The monomer (c) can be obtained in particular from an isocyanate comprising a monoethylenic unsaturation, such as, in particular,  $\alpha,\alpha$ -dimethyl-m-isopropenylbenzyl isocyanate.

The monomer (c) can be chosen in particular from acrylates, methacrylates or itaconates of oxyethylenated (1 to 50 EO) C<sub>6</sub>-C<sub>30</sub> fatty alcohol, such as steareth-20 methacrylate, oxyethylenated (25 EO) behenyl methacrylate, oxyethylenated (20 EO) monocetyl itaconate, oxyethylenated (20 EO) monostearyl itaconate or the acrylate modified by polyoxyethylenated (25 EO) C<sub>12</sub>-C<sub>24</sub> alcohols and from dimethyl-m-isopropenylbenzyl



chosen from vinyl acetate, vinyl alcohol, vinylpyrrolidone, olefins comprising from 2 to 20 carbon atoms, such as octadecene, ethylene, isobutylene, diisobutylene or isooctylene, and styrene, the maleic acid comonomers optionally being partially or completely hydrolysed.

5 Use will preferably be made of hydrophilic polymers, that is to say polymers having a solubility of water of greater than or equal to 2 g/l.

In an advantageous aspect of the present invention, the maleic acid copolymer may have a molar fraction of maleic acid units of between 0.1 and 1 and preferably between 0.4 and 0.9.

10 The weight-average molar mass of the maleic acid copolymer may be between 1,000 and 500,000 and preferably between 1,000 and 50,000.

It is preferable that the maleic acid copolymer be a styrene/maleic acid copolymer, and more preferably sodium styrene/maleic acid copolymer.

15 Use will preferably be made of a copolymer of styrene and of maleic acid in a 50/50 ratio.

20 Use may be made, for example, of the styrene/maleic acid (50/50) copolymer, in the form of an ammonium salt at 30% in water, sold under the reference SMA1000H® by Cray Valley or the styrene/maleic acid (50/50) copolymer, in the form of a sodium salt at 40% in water, sold under the reference SMA1000HNa® by Cray Valley.

25 The use of the styrene/maleic acid copolymer such as sodium styrene/maleic acid copolymer can improve the wettability of a film prepared by the composition according to the present invention.

30 It may be preferable that the anionic polymer be selected from the group consisting of polysaccharides such as alginic acid, hyaluronic acid, and cellulose polymers, anionic (co)polyaminoacids such as (co)polyglutamic acids, (co)poly(meth)acrylic acids, (co)polyamic acids, (co)polystyrene sulfonate, (co)poly(vinyl sulfates), dextran sulfate, chondroitin sulfate, (co)polymaleic acids, polyfumaric acids, maleic acid (co)polymers, and salts thereof.

35 The amount of the anionic polymer(s) in the composition according to the present invention may be 0.01% by weight or more, preferably 0.05% by weight or more, and more preferably 0.1% by weight or more, relative to the total weight of the composition.

40 The amount of the anionic polymer(s) in the composition according to the present invention may be 10% by weight or less, preferably 5% by weight or less, and more preferably 1% by weight or less, relative to the total weight of the composition.

45 The amount of the anionic polymer(s) in the composition according to the present invention may be from 0.01% to 10% by weight, preferably from 0.05% to 5% by weight, and more preferably from 0.1% to 1% by weight, relative to the total weight of the composition.

(Amphoteric Polymer)

An amphoteric polymer has both a positive charge density and a negative charge density.

The positive charge density of the amphoteric polymer may be from 0.01 meq/g to 20 meq/g, preferably from 0.05 to 15 meq/g, and more preferably from 0.1 to 10 meq/g.

5 The negative charge density of the amphoteric polymer may be from 0.01 meq/g to 20 meq/g, preferably from 0.05 to 15 meq/g, and more preferably from 0.1 to 10 meq/g.

The molecular weight of the amphoteric polymer may be 1000 or more, preferably 2000 or more, more preferably 3000 or more, and even more preferably 10,000 or more.

10 Unless otherwise defined in the descriptions, "molecular weight" means a number average molecular weight.

The amphoteric polymer may have  
at least one positively chargeable and/or positively charged moiety selected from the group  
15 consisting of a primary, secondary or tertiary amino group, a quaternary ammonium group, a guanidine group, a biguanide group, an imidazole group, an imino group, and a pyridyl group, and  
at least one negatively chargeable and/or negatively charged moiety selected from the group  
20 consisting of a sulfuric group, a sulfate group, a sulfonic group, a sulfonate group, a phosphoric group, a phosphate group, a phosphonic group, a phosphonate group, a carboxylic group, and a carboxylate group.

The amphoteric polymer may be a homopolymer or a copolymer. The term "copolymer" is  
25 understood to mean both copolymers obtained from two kinds of monomers and those obtained from more than two kinds of monomers, such as terpolymers obtained from three kinds of monomers.

The amphoteric polymers which can be used in accordance with the present invention may be  
30 chosen from the polymers containing K and M units distributed randomly in the polymer chain where K denotes a unit which is derived from a monomer containing at least one basic nitrogen atom and M denotes a unit which is derived from an acidic monomer containing one or more carboxylic or sulphonic groups or alternatively K and M may denote groups which are derived from zwitterionic monomers of carboxybetaines or of sulphobetaines. K and M  
35 may also denote a cationic polymer chain containing primary, secondary, tertiary or quaternary amine groups, in which at least one of the amine groups carries a carboxylic or sulphonic group linked through a hydrocarbon radical or alternatively K and M form part of a chain of a polymer with an  $\alpha,\beta$ -dicarboxylic ethylene unit in which one of the carboxylic groups has been caused to react with a polyamine containing one or more primary or  
40 secondary amine groups.

The amphoteric polymers corresponding to the definition given above which are more particularly preferred are chosen from the following polymers:

(1) The polymers resulting from the copolymerization of a monomer derived from a vinyl  
45 compound carrying a carboxylic group such as more particularly acrylic acid, methacrylic acid, maleic acid, alpha-chloroacrylic acid, and from a basic monomer derived from a substituted vinyl compound containing at least one basic atom such as more particularly dialkylaminoalkyl methacrylate and acrylate, dialkylaminoalkylmethacrylamide and  
50 acrylamide. Such compounds are described in U.S. Patent No. 3,836,537. There may also be mentioned the sodium acrylate/acrylamidopropyltrimethylammonium chloride copolymer

sold under the name POLYQUART KE 3033 by the company HENKEL. The vinyl compound may also be a dialkyldiallylammonium salt such as dimethyldiallylammonium chloride. The copolymers of acrylic acid and of the latter monomer are provided under the names MERQUAT 280, MERQUAT 295, MERQUAT 2003 PR, MERQUAT 3330 PR, and  
 5 MERQUAT PLUS 3330 by the company Lubrizol.

(2) The polymers containing units which are derived from:

- a) at least one monomer chosen from acrylamides or methacrylamides substituted on the nitrogen by an alkyl radical,
- 10 b) at least one acidic comonomer containing one or more reactive carboxylic groups, and
- c) at least one basic comonomer such as esters with primary, secondary, tertiary and quaternary amine substituents of acrylic and methacrylic acids and the product of quaternization of dimethylaminoethyl methacrylate with dimethyl or diethyl sulphate.

15 The N-substituted acrylamides or methacrylamides most particularly preferred according to the invention are groups whose alkyl radicals contain from 2 to 12 carbon atoms and more particularly N-ethylacrylamide, N-tert-butylacrylamide, N-tert-octylacrylamide, N-octylacrylamide, N-decylacrylamide, N-dodecylacrylamide as well as the corresponding methacrylamides.

20 The acidic comonomers are chosen more particularly from acrylic, methacrylic, crotonic, itaconic, maleic and fumaric acids as well as the alkyl monoesters having 1 to 4 carbon atoms of maleic or fumaric anhydrides or acids.

25 The basic comonomers preferred are methacrylates of aminoethyl, butylaminoethyl, N,N'-dimethylaminoethyl, N-tert-butylaminoethyl.

Particularly used are the copolymers whose CTFA name (4th ed. 1991) is Octylacrylamide/acrylates/butylaminoethylmethacrylate copolymer such as the products sold  
 30 under the name AMPHOMER or LOVOCRYL 47 by the company NATIONAL STARCH.

(3) The partially or completely alkylated and crosslinked polyaminoamides derived from polyaminoamides of the following general formula:

35  $[-\text{CO}-\text{R}_4-\text{CO}-\text{Z}-]$

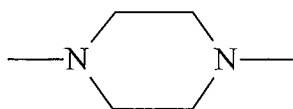
in which  $\text{R}_4$  represents a divalent radical derived from a saturated dicarboxylic acid, a mono- or dicarboxylic aliphatic acid with ethylenic double bond, an ester of a lower alkanol having 1 to 6 carbon atoms of these acids or a radical which is derived from the addition of any one of  
 40 the said acids with a bis-primary or bis-secondary amine, and Z denotes a radical of a bis-primary, mono- or bis-secondary polyalkylene-polyamine and preferably represents:

a) in the proportions of 60 to 100 mol%, the radical

45  $-\text{NH}-[(\text{CH}_2)_x-\text{NH}-]_p-$

where  $x=2$  and  $p=2$  or 3, or alternatively  $x=3$  and  $p=2$ , this radical being derived from the diethylenetriamine, triethylenetetraamine or dipropylenetriamine;

b) in the proportions of 0 to 40 mol%, the radical (IV) above, in which  $x=2$  and  $p=1$  and which is derived from ethylenediamine, or the radical which is derived from piperazine:

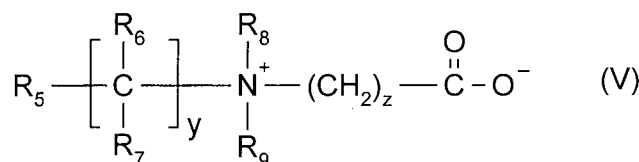


c) in the proportions of 0 to 20 mol%, the radical -NH-(CH<sub>2</sub>)<sub>6</sub>-NH- which is derived from hexamethylenediamine, these polyamino amines being crosslinked by adding a bifunctional crosslinking agent chosen from the epihalohydrins, diepoxides, dianhydrides, bis-unsaturated derivatives, by means of 0.025 to 0.35 mol of crosslinking agent per amine group of the polyamino amide and alkylated by the action of acrylic acid, chloroacetic acid or of an alkanesultone or of their salts.

The saturated carboxylic acids are preferably chosen from the acids having 6 to 10 carbon atoms such as adipic, 2,2,4-trimethyladipic and 2,4,4-trimethyladipic acid, terephthalic acid, the acids with ethylene double bond such as for example acrylic, methacrylic and itaconic acids.

The alkanesultones used in the alkylation are preferably propane or butanesultone, and the salts of the alkylating agents are preferably the sodium or potassium salts.

(4) The polymers containing zwitterionic units of formula:



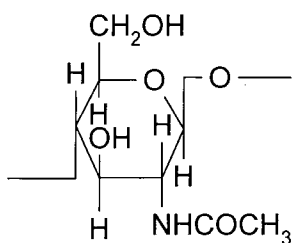
in which

R<sub>5</sub> denotes a polymerizable unsaturated group such as an acrylate, methacrylate, acrylamide or methacrylamide group, y and z represent an integer from 1 to 3, R<sub>6</sub> and R<sub>7</sub> represent a hydrogen atom, methyl, ethyl or propyl, R<sub>8</sub> and R<sub>9</sub> represent a hydrogen atom or an alkyl radical such that the sum of the carbon atoms in R<sub>8</sub> and R<sub>9</sub> does not exceed 10.

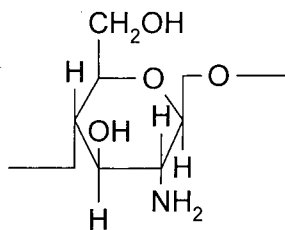
The polymers comprising such units may also comprise units derived from nonzwitterionic monomers such as dimethyl or diethylaminoethyl acrylate or methacrylate or alkyl acrylates or methacrylates, acrylamides or methacrylamides or vinyl acetate.

By way of example, there may be mentioned the copolymer of butyl methacrylate/dimethylcarboxymethylammonioethyl methacrylate such as the product sold under the name DIAFORMER Z301 by the company SANDOZ.

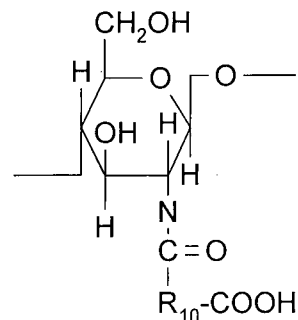
(5) The polymers derived from chitosan containing monomeric units corresponding to the following formulae (VI), (VII), and (VIII):



(VI)

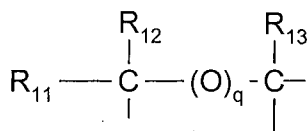


(VII)



(VIII)

the (VI) unit being present in proportions of from 0 to 30%, the (VII) unit in proportions of from 5 to 50% and the (VIII) unit in proportions of from 30 to 90%, it being understood that in this (VIII) unit, R<sub>10</sub> represents a radical of formula:



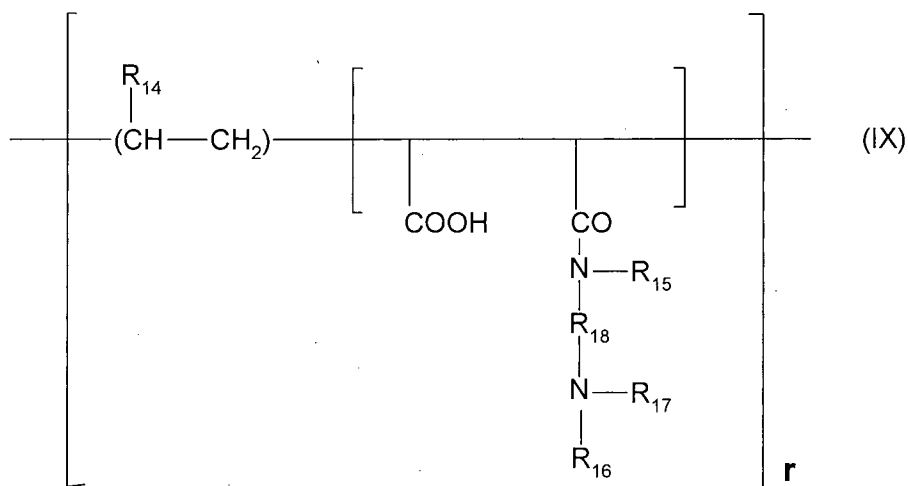
in which

if q=0, R<sub>11</sub>, R<sub>12</sub> and R<sub>13</sub>, which are identical or different, each represent a hydrogen atom, a methyl, hydroxyl, acetoxy or amino residue, a monoalkylamine residue or a dialkylamine residue optionally interrupted by one or more nitrogen atoms and/or optionally substituted with one or more amine, hydroxyl, carboxyl, alkylthio or sulphonic groups, or an alkylthio residue whose alkyl group carries an amino residue, at least one of the R<sub>11</sub>, R<sub>12</sub> and R<sub>13</sub> radicals being in this case a hydrogen atom;

or if q=1, R<sub>11</sub>, R<sub>12</sub> and R<sub>13</sub> each represent a hydrogen atom, as well as the salts formed by these compounds with bases or acids.

(6) The polymers derived from the N-carboxyalkylation of chitosan such as N-carboxymethyl chitosan or N-carboxybutyl chitosan sold under the name "EVALSAN" by the company JAN DEKKER.

(7) The polymers corresponding to the general formula (IX) such as those described for example in French Patent 1,400,366:

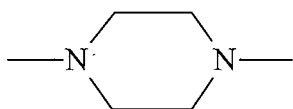


in which  $R_{14}$  represents a hydrogen atom, a  $CH_3O$ ,  $CH_3CH_2O$  or phenyl radical,  $R_{15}$  denotes hydrogen or a lower alkyl radical such as methyl or ethyl,  $R_{16}$  denotes hydrogen or a lower alkyl radical such as methyl or ethyl,  $R_{17}$  denotes a lower alkyl radical such as methyl or ethyl or a radical corresponding to the formula:  $-R_{18}-N(R_{16})_2$ ,  $R_{18}$  representing a group  $-CH_2-CH_2-$ ,  $-CH_2-CH_2-CH_2-$  or  $-CH_2-CH(CH_3)-$ ,  $R_{16}$  having the meanings mentioned above, as well as the higher homologues of these radicals and containing up to 6 carbon atoms.

(8) Amphoteric polymers of the -D-X-D-X- type chosen from:  
 a) the polymers obtained by the action of chloroacetic acid or sodium chloroacetate on the compounds containing at least one unit of formula:

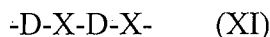


where D denotes a radical

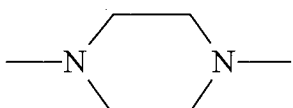


and X denotes the symbol E or E', E or E', which are identical or different, denote a bivalent radical which is an alkylene radical with a linear or branched chain containing up to 7 carbon atoms in the principal chain which is unsubstituted or substituted with hydroxyl groups and which may contain, in addition, oxygen, nitrogen or sulphur atoms, 1 to 3 aromatic and/or heterocyclic rings; the oxygen, nitrogen and sulphur atoms being present in the form of ether, thioether, sulphoxide, sulphone, sulphonium, alkylamine or alkenylamine groups, or hydroxyl, benzylamine, amine oxide, quaternary ammonium, amide, imide, alcohol, ester and/or urethane groups.

b) The polymers of formula:



where D denotes a radical



and X denotes the symbol E or E' and, at least once, E'; E having the meaning indicated above and E' is a bivalent radical which is an alkylene radical with a linear or branched chain having up to 7 carbon atoms in the principal chain, which is unsubstituted or substituted with one or more hydroxyl radicals and containing one or more nitrogen atoms, the nitrogen atom being substituted with an alkyl chain optionally interrupted by an oxygen atom and necessarily containing one or more carboxyl functional groups or one or more hydroxyl functional groups and betainized by reaction with chloroacetic acid or sodium chloroacetate.

(9) The copolymers  $(C_1-C_5)$ alkyl vinyl ether/maleic anhydride partially modified by semiamidation with an N,N-dialkylaminoalkylamine such as N,N-dimethylaminopropylamine or by semiesterification with an N,N-dialkanolamine. These copolymers may also contain other vinyl comonomers such as vinylcaprolactam.

The amphoteric polymers particularly preferred according to the invention are those of the family (1), particularly those containing a salt of dialkyldiallyl ammonium as a cationic monomer.

5 The amphoteric polymers may be chosen from polyquaternium-22, polyquaternium-39, polyquaternium-53, polyquaternium-64, polyquaternium-51, polyquaternium-61 and mixtures thereof. Polyquaternium-39 and polyquaternium-53, for example the product Merquat 3330 PR and Merquat 2003 PR, sold by Lubrizol, are more preferable.

10 The amount of the amphoteric polymer(s) in the composition according to the present invention may be 0.01% by weight or more, preferably 0.05% by weight or more, and more preferably 0.1% by weight or more, relative to the total weight of the composition.

15 The amount of the amphoteric polymer(s) in the composition according to the present invention may be 10% by weight or less, preferably 5% by weight or less, and more preferably 1% by weight or less, relative to the total weight of the composition.

20 The amount of the amphoteric polymer(s) in the composition according to the present invention may be from 0.01% to 10% by weight, preferably from 0.05% to 5% by weight, and more preferably from 0.1% to 1% by weight, relative to the total weight of the composition.

#### (Preferable Embodiments)

It is preferable that the composition according to the present invention comprises:

- 25 (a) at least one polyion complex comprising  
at least one cationic polymer, preferably at least two cationic polymers, and more preferably polyquaternium-67 and polyquaternium-6, and  
at least one non-polymeric acid having two or more pKa values or salt(s) thereof, as a crosslinker,  
30 (b) at least one fatty alcohol; and  
(c) water.

35 It is preferable that the cationic polymer be selected from cationic polysaccharides. It is more preferable that the cationic polymer be selected from cationic cellulose polymers. It is even more preferable that the cationic polymer be selected from cationic cellulose polymers having at least one quaternary ammonium group comprising at least one fatty chain, as explained in the above (15) for the section of (Cationic Polymer).

40 It may be preferable that the polyion complex comprise no anionic polymer.

#### (Non-Polymeric Acid Having Two or More Acid Dissociation Constants)

45 As a crosslinker for the ionic polymer, the composition according to the present invention may include at least one non-polymeric acid having two or more pKa values or salt(s) thereof, i.e., at least one non-polymeric acid having two or more acid dissociation constants or salt(s) thereof. The pKa value (acid dissociation constant) is well known to those skilled in the art, and should be determined at a constant temperature such as 25°C.

The non-polymeric acid having two or more pKa values or salt(s) thereof can be included in the (a) particle. The non-polymeric acid having two or more pKa values can function as a crosslinker for the cationic polymer, anionic polymer and amphoteric polymers.

5 The term "non-polymeric" here means that the acid is not obtained by polymerizing two or more monomers. Therefore, the non-polymeric acid does not correspond to an acid obtained by polymerizing two or more monomers such as polycarboxylic acid.

10 It is preferable that the molecular weight of the non-polymeric acid having two or more pKa values or salt(s) thereof be less than 1000, preferably 800 or less, and more preferably 700 or less.

15 There is no limit to the type of the non-polymeric acid having two or more pKa values or salt(s) thereof. Two or more different types of non-polymeric acids having two or more pKa values or salts thereof may be used in combination. Thus, a single type of a non-polymeric acid having two or more pKa values or a salt thereof or a combination of different types of non-polymeric acids having two or more pKa values or salts thereof may be used.

20 The term "salt" in the present specification means a salt formed by addition of suitable base(s) to the non-polymeric acid having two or more pKa values, which may be obtained from a reaction with the non-polymeric acid having two or more pKa values with the base(s) according to methods known to those skilled in the art. As the salt, mention may be made of metal salts, for example salts with alkaline metal such as Na and K, and salts with alkaline earth metal such as Mg and Ca, and ammonium salts.

25 The non-polymeric acid having two or more pKa values or salt(s) thereof may be an organic acid or salt(s) thereof, and preferably a hydrophilic or water-soluble organic acid or salt(s) thereof.

30 The non-polymeric acid having two or more pKa values may have at least two acid groups selected from the group consisting of a carboxylic group, a sulfuric group, a sulfonic group, a phosphoric group, a phosphonic group, a phenolic hydroxyl group, and a mixture thereof.

35 The non-polymeric acid having two or more pKa values may be a non-polymeric polyvalent acid.

The non-polymeric acid having two or more pKa values may be selected from the group consisting of dicarboxylic acids, disulfonic acids, and diphosphoric acids, and a mixture thereof.

40 The non-polymeric acid having two or more pKa values or salt(s) thereof may be selected from the group consisting of oxalic acid, malonic acid, succinic acid, glutaric acid, adipic acid, pimelic acid, suberic acid, azelaic acid, sebacic acid, fumaric acid, maleic acid, malic acid, citric acid, aconitic acid, oxaloacetic acid, tartaric acid, and salts thereof; aspartic acid, glutamic acid, and salts thereof; terephthalylidene dicamphor sulfonic acid or salts thereof (Mexoryl SX), Benzophenone-9; phytic acid, and salts thereof; Red 2 (Amaranth), Red 102 (New Coccine), Yellow 5 (Tartrazine), Yellow 6 (Sunset Yellow FCF), Green 3 (Fast Green FCF), Blue 1 (Brilliant Blue FCF), Blue 2 (Indigo Carmine), Red 201 (Lithol Rubine B), Red 202 (Lithol Rubine BCA), Red 204 (Lake Red CBA), Red 206 (Lithol Red CA), Red 207 (Lithol Red BA), Red 208 (Lithol Red SR), Red 219 (Brilliant Lake Red R), Red 220 (Deep

Maroon), Red 227 (Fast Acid Magenta), Yellow 203 (Quinoline Yellow WS), Green 201 (Alizanine Cyanine Green F), Green 204 (Pyranine Conc), Green 205 (Light Green SF Yellowish), Blue 203 (Patent Blue CA), Blue 205 (Alfazurine FG), Red 401 (Violamine R), Red 405 (Permanent Re F5R), Red 502 (Ponceau 3R), Red 503 (Ponceau R), Red 504 (Ponceau SX), Green 401 (Naphtol Green B), Green 402 (Guinea Green B), and Black 401 (Naphtol Blue Black); folic acid, ascorbic acid, erythorbic acid, and salts thereof; cystine and salts thereof; EDTA and salts thereof; glycyrrhizin and salts thereof; and a mixture thereof.

It may be preferable that the non-polymeric acid having two or more pKa values or salt(s) thereof be selected from the group consisting of terephthalylidene dicamphor sulfonic acid and salts thereof (Mexoryl SX), Yellow 6 (Sunset Yellow FCF), ascorbic acid, phytic acid, citric acid, tartaric acid, and salts thereof, and a mixture thereof.

The amount of the non-polymeric acid having two or more pKa values or salt(s) thereof in the composition according to the present invention may be 0.001% by weight or more, preferably 0.005% by weight or more, and more preferably 0.01% by weight or more, relative to the total weight of the composition.

The amount of the non-polymeric acid having two or more pKa values or salt(s) thereof in the composition according to the present invention may be 10% by weight or less, preferably 5% by weight or less, and more preferably 1% by weight or less, relative to the total weight of the composition.

The amount of the non-polymeric acid having two or more pKa values or salt(s) thereof in the composition according to the present invention may be from 0.001% to 10% by weight, preferably from 0.005% to 5% by weight, and more preferably from 0.01% to 1% by weight, relative to the total weight of the composition.

(Non-Polymeric Base Having Two or More Base Dissociation Constants)

As a crosslinker for the ionic polymer, the composition according to the present invention may include at least one non-polymeric base having two or more pKb values or salt(s) thereof, i.e., at least one non-polymeric base having two or more base dissociation constants or salt(s) thereof. The pKb value (base dissociation constant) is well known to those skilled in the art, and should be determined at a constant temperature such as 25°C.

The non-polymeric base having two or more pKb values or salt(s) thereof can be included in the (a) particle. The non-polymeric base having two or more pKb values can function as a crosslinker for the cationic polymer, anionic polymer and amphoteric polymers.

The term "non-polymeric" here means that the base is not obtained by polymerizing two or more monomers. Therefore, the non-polymeric base does not correspond to a base obtained by polymerizing two or more monomers such as polyallylamine.

It is preferable that the molecular weight of the non-polymeric base having two or more pKb values or salt(s) thereof be 1000 or less, preferably 800 or less, and more preferably 700 or less.

There is no limit to the type of the non-polymeric base having two or more pKb values or salt(s) thereof. Two or more different types of non-polymeric bases having two or more pKb

values or salts thereof may be used in combination. Thus, a single type of a non-polymeric base having two or more pK<sub>b</sub> values or a salt thereof or a combination of different types of non-polymeric bases having two or more pK<sub>b</sub> values or salts thereof may be used.

5 The term "salt" in the present specification means a salt formed by addition of suitable acid(s) to the non-polymeric base having two or more pK<sub>b</sub> values, which may be obtained from a reaction with the non-polymeric base having two or more pK<sub>b</sub> values with the acid(s) according to methods known to those skilled in the art. As the salt, mention may be made of ammonium salts, for example salts with inorganic acid such as HCl and HNO<sub>3</sub>, and salts with  
10 organic acid such as carboxylic acids and sulfonic acids.

The non-polymeric base having two or more pK<sub>b</sub> values or salt(s) thereof may be an organic base or salt(s) thereof, and preferably a hydrophilic or water-soluble organic base or salt(s) thereof.

15 The non-polymeric base having two or more pK<sub>b</sub> values may have at least two basic groups selected from the group consisting of an amino group, a guanidine group, a biguanide group, an imidazole group, an imino group, a pyridyl group and a mixture thereof.

20 The non-polymeric base having two or more pK<sub>b</sub> values may be selected from the group consisting of non-polymeric diamines such as ethylenediamine, propylenediamine, pentanediamine, hexanediamine, urea and derivatives thereof and guanidine and derivatives thereof, non-polymeric polyamines such as spermine and spermidine, basic amino acids, and a mixture thereof.

25 The non-polymeric base having two or more pK<sub>b</sub> values or salt(s) thereof may be selected from the group consisting of arginine, lysine, histidine, cysteine, cystine, tyrosine, tryptophan, ornithine, and a mixture thereof.

30 It may be preferable that the non-polymeric base having two or more pK<sub>b</sub> values or salt(s) thereof be selected from the group consisting of arginine, lysine, histidine, and a mixture thereof.

35 The amount of the non-polymeric base having two or more pK<sub>b</sub> values or salt(s) thereof in the composition according to the present invention may be 0.001% by weight or more, preferably 0.005% by weight or more, and more preferably 0.01% by weight or more, relative to the total weight of the composition.

40 The amount of the non-polymeric base having two or more pK<sub>b</sub> values or salt(s) thereof in the composition according to the present invention may be 10% by weight or less, preferably 5% by weight or less, and more preferably 1% by weight or less, relative to the total weight of the composition.

45 The amount of the non-polymeric base having two or more pK<sub>b</sub> values or salt(s) thereof in the composition according to the present invention may be from 0.001% to 10% by weight, preferably from 0.005% to 5% by weight, and more preferably from 0.01% to 1% by weight, relative to the total weight of the composition.

[Fatty Alcohol]

50

The composition according to the present invention comprises (b) at least one fatty alcohol. Two or more different types of (b) fatty alcohols may be used in combination. Thus, a single type of (b) fatty alcohol or a combination of different types of (b) fatty alcohols may be used.

5 The term “fatty” here means the inclusion of a relatively large number of carbon atoms. Thus, alcohols which have 6 or more, preferably 8 or more, and more preferably 10 or more carbon atoms are encompassed within the scope of fatty alcohols. The fatty alcohols may be saturated or unsaturated. The fatty alcohol may be linear or branched. Two or more fatty alcohols may be used in combination.

10 The (b) fatty alcohol may have the structure R-OH wherein R is chosen from saturated and unsaturated, linear and branched radicals containing from 8 to 40 carbon atoms, for example from 8 to 30 carbon atoms. In at least one embodiment, R is chosen from C<sub>12</sub>-C<sub>24</sub> alkyl and C<sub>12</sub>-C<sub>24</sub> alkenyl groups. R may be or may not be substituted with at least one hydroxyl group.

15 Non-limiting examples of the (b) fatty alcohols that may be mentioned include lauryl alcohol, cetyl alcohol, stearyl alcohol, oleyl alcohol, behenyl alcohol, linoleyl alcohol, undecylenyl alcohol, palmitoleyl alcohol, arachidonyl alcohol, erucyl alcohol, cetearyl alcohol, and mixtures thereof.

20 Examples of suitable fatty alcohols include, but are not limited to, cetyl alcohol, cetearyl alcohol, stearyl alcohol, behenyl alcohol, oleyl alcohol, and mixtures thereof.

25 The fatty alcohol may represent a mixture of fatty alcohols, which means that several species of fatty alcohol may coexist, in the form of a mixture, in a commercial product.

According to at least one embodiment, the fatty alcohol used in the composition according to the present invention is chosen from a mixture of cetyl alcohol and cetearyl alcohol (cetearyl alcohol).

30 The amount of the (b) fatty alcohol in the composition according to the present invention may be 0.1% by weight or more, preferably 0.5% by weight or more, and more preferably 1% by weight or more, relative to the total weight of the composition.

35 The amount of the (b) fatty alcohol in the composition according to the present invention may be 15% by weight or less, preferably 10% by weight or less, and more preferably 5% by weight or less, relative to the total weight of the composition.

40 The amount of the (b) fatty alcohol in the composition according to the present invention may be from 0.1% to 15% by weight, preferably from 0.5% to 10% by weight, and more preferably from 1% to 5% by weight, relative to the total weight of the composition.

[Water]

45 The composition according to the present invention comprises (c) water.

The (c) water can form an aqueous phase of the composition according to the present invention.

The amount of the (c) water may be 50% by weight or more, preferably 60% by weight or more, and more preferably 70% by weight or more, relative to the total weight of the composition.

- 5 The amount of the (c) water may be 99% by weight or less, preferably 97% by weight or less, and more preferably 95% by weight or less, relative to the total weight of the composition.

10 The amount of the (c) water may be from 50% to 99% by weight, preferably from 60% to 97% by weight, and more preferably from 70% to 95% by weight, relative to the total weight of the composition.

[Fatty Material]

15 The composition according to the present invention may comprise (d) at least one fatty material other than the (b) fatty alcohol. Two or more different types of (d) fatty materials may be used in combination. Thus, a single type of (d) fatty material or a combination of different types of (d) fatty materials may be used.

20 The term "fatty material" here means an organic compound that is insoluble in water at ordinary temperature (25°C) and at atmospheric pressure (760 mmHg) (solubility of less than 5% by weight, preferably less than 1% by weight, and more preferably less than 0.1% by weight). The fatty material may contain, in its structure, a sequence of at least two siloxane groups or at least one hydrocarbon-based chain containing at least 6 carbon atoms. In addition, the fatty substances may be soluble in organic solvents under the same temperature and pressure conditions, for instance chloroform, ethanol, benzene or  
25 decamethylcyclopentasiloxane.

30 The (d) fatty material may be in the form of a liquid or a solid. Here, "liquid" and "solid" mean that the fatty material is in the form of a liquid or a paste (non-solid) or solid, respectively, at ambient temperature (25°C) under atmospheric pressure (760 mmHg or 10<sup>5</sup> Pa).

35 The (d) fatty material may be in the form of a paste or a solid at ambient temperature and under atmospheric pressure. As examples of the (d) fatty material in the form of a solid, mention may be made of, for example, waxes.

40 The term "wax" here means that the fatty material is substantially in the form of a solid at room temperature (25°C) under atmospheric pressure (760 mmHg), and has a melting point generally of 35°C or more. As the waxy fatty material, waxes generally used in cosmetics can be used alone or in combinations thereof.

45 For example, the wax may be selected from carnauba wax, beeswax, microcrystalline waxes, ozokerites, hydrogenated jojoba oil, polyethylene waxes such as the wax sold under the name "Performalene 400 Polyethylene" by the company New Phase Technologies, silicone waxes, for instance poly(C<sub>24</sub>-C<sub>28</sub>)alkylmethyl dimethylsiloxane, such as the product sold under the name "Abil Wax 9810" by the company Goldschmidt, shea butter, palm butter, the C<sub>20</sub>-C<sub>40</sub> alkyl stearate sold under the name "Kester Wax K82H" by the company Kester Keunen, stearyl benzoate, shellac wax, and mixtures thereof. For example, a wax selected from  
50 carnauba wax, candelilla wax, ozokerites, hydrogenated jojoba oil and polyethylene waxes can be used. In at least one embodiment, the wax is preferably selected from candelilla wax,

beeswax, and ozokerite, and mixtures thereof.

On the other hand, the (d) fatty material may be in the form of a liquid or a paste, at ambient temperature and under atmospheric pressure. As examples of fatty material in the form of a liquid or a paste, mention may be made of oil.

It is preferable that the (d) fatty material be selected from oils.

Here, "oil" means a fatty compound or substance which is in the form of a liquid or a paste (non-solid) at room temperature (25°C) under atmospheric pressure (760 mmHg). As the oils, those generally used in cosmetics can be used alone or in combination thereof. These oils may be volatile or non-volatile.

The oil may be selected from the group consisting of oils of plant or animal origin, synthetic oils, silicone oils, and hydrocarbon oils.

As examples of plant oils, mention may be made of linseed oil, camellia oil, macadamia nut oil, corn oil, mink oil, olive oil, avocado oil, sasanqua oil, castor oil, apricot oil, coconuts oil, safflower oil, jojoba oil, sunflower oil, almond oil, rapeseed oil, sesame oil, soybean oil, peanut oil, and mixtures thereof.

As examples of animal oils, mention may be made of squalene and squalane.

As examples of synthetic oils, mention may be made of alkane oils such as isododecane and isohexadecane, ester oils, ether oils, and artificial triglycerides.

The ester oils are preferably liquid esters of saturated or unsaturated, linear or branched C<sub>1</sub>-C<sub>26</sub> aliphatic monoacids or polyacids and of saturated or unsaturated, linear or branched C<sub>1</sub>-C<sub>26</sub> aliphatic monoalcohols or polyalcohols, the total number of carbon atoms of the esters being greater than or equal to 10.

Preferably, for the esters of monoalcohols, at least one from among the alcohol and the acid from which the esters of the present invention are derived is branched.

Among the monoesters of monoacids and of monoalcohols, mention may be made of ethyl palmitate, ethyl hexyl palmitate, isopropyl palmitate, dicaprylyl carbonate, alkyl myristates such as isopropyl myristate or ethyl myristate, isocetyl stearate, 2-ethylhexyl isononanoate, isononyl isononanoate, isodecyl neopentanoate and isostearyl neopentanoate.

Esters of C<sub>4</sub>-C<sub>22</sub> dicarboxylic or tricarboxylic acids and of C<sub>1</sub>-C<sub>22</sub> alcohols, and esters of monocarboxylic, dicarboxylic or tricarboxylic acids and of non-sugar C<sub>4</sub>-C<sub>26</sub> dihydroxy, trihydroxy, tetrahydroxy or pentahydroxy alcohols may also be used.

Mention may especially be made of: diethyl sebacate; isopropyl lauroyl sarcosinate; diisopropyl sebacate; bis(2-ethylhexyl) sebacate; diisopropyl adipate; di-n-propyl adipate; dioctyl adipate; bis(2-ethylhexyl) adipate; diisostearyl adipate; bis(2-ethylhexyl) maleate; triisopropyl citrate; triisocetyl citrate; triisostearyl citrate; glyceryl trilactate; glyceryl trioctanoate; trioctyldodecyl citrate; trioleyl citrate; neopentyl glycol diheptanoate; diethylene glycol diisononanoate.

As ester oils, one can use sugar esters and diesters of C<sub>6</sub>-C<sub>30</sub> and preferably C<sub>12</sub>-C<sub>22</sub> fatty acids. It is recalled that the term "sugar" means oxygen-bearing hydrocarbon-based compounds containing several alcohol functions, with or without aldehyde or ketone functions, and which comprise at least 4 carbon atoms. These sugars may be monosaccharides, oligosaccharides or polysaccharides.

Examples of suitable sugars that may be mentioned include sucrose (or saccharose), glucose, galactose, ribose, fucose, maltose, fructose, mannose, arabinose, xylose and lactose, and derivatives thereof, especially alkyl derivatives, such as methyl derivatives, for instance methylglucose.

The sugar esters of fatty acids may be chosen especially from the group comprising the esters or mixtures of esters of sugars described previously and of linear or branched, saturated or unsaturated C<sub>6</sub>-C<sub>30</sub> and preferably C<sub>12</sub>-C<sub>22</sub> fatty acids. If they are unsaturated, these compounds may have one to three conjugated or non-conjugated carbon-carbon double bonds.

The esters according to this variant may also be selected from monoesters, diesters, triesters, tetraesters and polyesters, and mixtures thereof.

These esters may be, for example, oleates, laurates, palmitates, myristates, behenates, cocoates, stearates, linoleates, linolenates, caprates and arachidonates, or mixtures thereof such as, especially, oleopalmitate, oleostearate and palmitostearate mixed esters, as well as pentaerythrityl tetraethyl hexanoate and dipentaerythrityl hexacaprylate/hexacaprate.

More particularly, use is made of monoesters and diesters and especially sucrose, glucose or methylglucose monooleates or dioleates, stearates, behenates, oleopalmitates, linoleates, linolenates and oleostearates.

An example that may be mentioned is the product sold under the name Glucate® DO by the company Amerchol, which is a methylglucose dioleate.

As examples of preferable ester oils, mention may be made of diisopropyl adipate, dioctyl adipate, 2-ethylhexyl hexanoate, ethyl laurate, cetyl octanoate, octyldodecyl octanoate, isodecyl neopentanoate, myristyl propionate, 2-ethylhexyl 2-ethylhexanoate, 2-ethylhexyl octanoate, 2-ethylhexyl caprylate/caprate, methyl palmitate, ethyl palmitate, isopropyl palmitate, dicaprylyl carbonate, isopropyl lauroyl sarcosinate, isononyl isononanoate, ethylhexyl palmitate, isohexyl laurate, hexyl laurate, isocetyl stearate, isopropyl isostearate, isopropyl myristate, isodecyl oleate, glyceryl tri(2-ethylhexanoate), pentaerythrityl tetra(2-ethylhexanoate), 2-ethylhexyl succinate, diethyl sebacate, polyglyceryl-6 polyricinoleate, polyglyceryl-2 triisostearate, glyceryl stearate, and mixtures thereof.

As examples of artificial triglycerides, mention may be made of capryl caprylyl glycerides, glyceryl trimyristate, glyceryl tripalmitate, glyceryl trilinolenate, glyceryl trilaurate, glyceryl tricaprate, glyceryl tricaprylate, glyceryl tri(caprate/caprylate) and glyceryl tri(caprate/caprylate/linolenate).

As examples of silicone oils, mention may be made of linear organopolysiloxanes such as dimethylpolysiloxane, methylphenylpolysiloxane, methylhydrogenpolysiloxane, and the like; cyclic organopolysiloxanes such as cyclohexasiloxane, octamethylcyclotetrasiloxane,

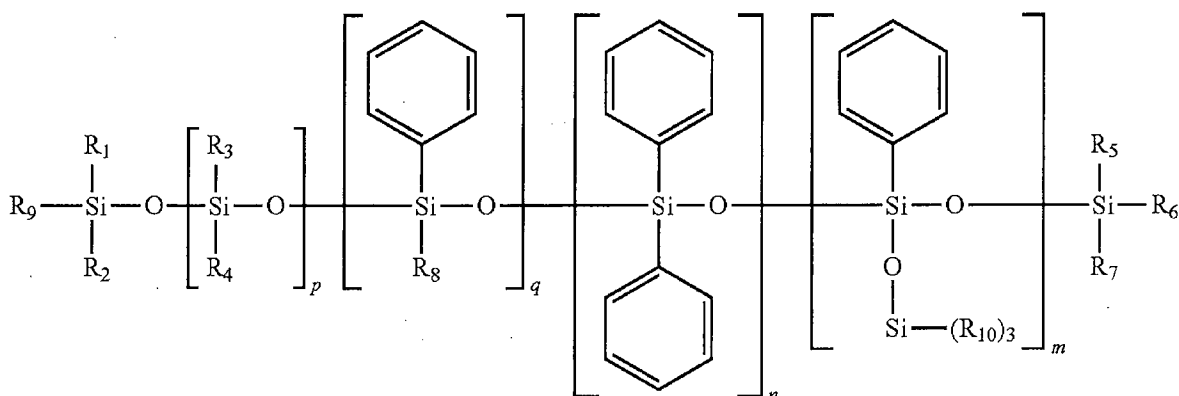


- the Silbione® oils of the 47 and 70 047 series or the Mirasil® oils sold by Rhodia, for instance the oil 70 047 V 500 000;
- the oils of the Mirasil® series sold by the company Rhodia;
- the oils of the 200 series from the company Dow Corning, such as DC200 with a viscosity of 60 000 mm<sup>2</sup>/s; and
- the Viscasil® oils from General Electric and certain oils of the SF series (SF 96, SF 18) from General Electric.

Mention may also be made of polydimethylsiloxanes containing dimethylsilanol end groups known under the name dimethiconol (CTFA), such as the oils of the 48 series from the company Rhodia.

Among the silicones containing aryl groups, mention may be made of polydiarylsiloxanes, especially polydiphenylsiloxanes and polyalkylarylsiloxanes such as phenyl silicone oil.

The phenyl silicone oil may be chosen from the phenyl silicones of the following formula:



in which

R<sub>1</sub> to R<sub>10</sub>, independently of each other, are saturated or unsaturated, linear, cyclic or branched C<sub>1</sub>-C<sub>30</sub> hydrocarbon-based radicals, preferably C<sub>1</sub>-C<sub>12</sub> hydrocarbon-based radicals, and more preferably C<sub>1</sub>-C<sub>6</sub> hydrocarbon-based radicals, in particular methyl, ethyl, propyl or butyl radicals, and

m, n, p and q are, independently of each other, integers 0 to 900 inclusive, preferably 0 to 500 inclusive, and more preferably 0 to 100 inclusive, with the proviso that the sum n+m+q is other than 0.

Examples that may be mentioned include the products sold under the following names:

- the Silbione® oils of the 70 641 series from Rhodia;
- the oils of the Rhodorsil® 70 633 and 763 series from Rhodia;
- the oil Dow Corning 556 Cosmetic Grade Fluid from Dow Corning;
- the silicones of the PK series from Bayer, such as the product PK20;
- certain oils of the SF series from General Electric, such as SF 1023, SF 1154, SF 1250 and SF 1265.

As the phenyl silicone oil, phenyl trimethicone (R<sub>1</sub> to R<sub>10</sub> are methyl; p, q, and n = 0; m=1 in the above formula) is preferable.

The organomodified liquid silicones may especially contain polyethyleneoxy and/or polypropyleneoxy groups. Mention may thus be made of the silicone KF-6017 proposed by Shin-Etsu, and the oils Silwet® L722 and L77 from the company Union Carbide.

5 Hydrocarbon oils may be chosen from:

- linear or branched, optionally cyclic, C<sub>6</sub>-C<sub>16</sub> lower alkanes. Examples that may be mentioned include hexane, undecane, dodecane, tridecane, and isoparaffins, for instance isohexadecane, isododecane and isodecane; and
  - linear or branched hydrocarbons containing more than 16 carbon atoms, such as
- 10 liquid paraffins, liquid petroleum jelly, polydecenes and hydrogenated polyisobutenes such as Parleam®, and squalane.

As preferable examples of hydrocarbon oils, mention may be made of, for example, linear or branched hydrocarbons such as isohexadecane, isododecane, squalane, mineral oil (e.g., liquid

15 paraffin), paraffin, vaseline or petrolatum, naphthalenes, and the like; hydrogenated polyisobutene, isoeicosan, and decene/butene copolymer; and mixtures thereof.

It may be preferable that the (d) fatty material be not selected from silicone oils. In other words, it may be preferable that the composition according to the present invention comprises

20 no silicone oil.

The amount of the (d) fatty material(s) in the composition according to the present invention may be 0.1% by weight or more, preferably 0.5% by weight or more, and more preferably 1%

25 by weight or more, relative to the total weight of the composition.

The amount of the (d) fatty material(s) in the composition according to the present invention may be 15% by weight or less, preferably 10% by weight or less, and more preferably 5% by weight or less, relative to the total weight of the composition.

30 The amount of the (d) fatty material(s) in the composition according to the present invention may be from 0.1% to 15% by weight, preferably from 0.5% to 10% by weight, and more preferably from 1% to 5% by weight, relative to the total weight of the composition.

(Fatty Phase)

35 The (b) fatty alcohol and, if present, the (d) fatty material can form a fatty phase.

The amount of the (d) fatty material(s) in the composition according to the present invention may be 0.1% by weight or more, preferably 0.5% by weight or more, and more preferably 1%

40 by weight or more, relative to the total weight of the composition.

The amount of the (d) fatty material(s) in the composition according to the present invention may be 20% by weight or less, preferably 15% by weight or less, and more preferably 10% by weight or less, relative to the total weight of the composition.

45

The amount of the (d) fatty material(s) in the composition according to the present invention may be from 0.1% to 20% by weight, preferably from 0.5% to 15% by weight, and more preferably from 1% to 10% by weight, relative to the total weight of the composition.

50 [Texture Agent]

The composition according to the present invention may comprise (e) at least one texture agent. Two or more different types of (e) texture agents may be used in combination. Thus, a single type of (e) texture agent or a combination of different types of (e) texture agents may be used.

The term "texture agent" here means any agent which can thicken the system or the composition.

The (e) texture agent may be selected from hydrophilic thickeners.

It is preferable that the (e) texture agent be selected from hydrophilic nonionic polymeric thickeners. The hydrophilic nonionic polymeric thickener here means a hydrophilic nonionic thickener based on at least one polymer.

It is more preferable that the (e) texture agent be selected from hydrophilic nonionic polymeric associative thickeners and hydrophilic nonionic polysaccharide thickeners.

{Hydrophilic Nonionic Polymeric Associative Thickener}

The hydrophilic nonionic polymeric associative thickeners here means a nonionic thickener comprising at least one polymer with both at least one hydrophilic unit and at least one hydrophobic unit, such as a least one C<sub>8</sub>-C<sub>30</sub> fatty chain, and is hydrophilic as a whole such that it can thicken an aqueous phase.

The hydrophilic nonionic polymeric associative thickeners may, for example, be chosen from:

- (i) celluloses modified with groups comprising at least one fatty chain; examples that may be mentioned include:
  - hydroxyethylcelluloses modified with groups comprising at least one fatty chain chosen from alkyl, arylalkyl and alkylaryl groups, and in which the alkyl groups are, for example, C<sub>8</sub>-C<sub>22</sub>, such as the product Natrosol Plus Grade 330 CS(C<sub>1</sub>-C<sub>6</sub> alkyls) sold by the company Aqualon, and the product Bermocoll EHM 100 sold by the company Berol Nobel, and
  - celluloses modified with polyalkylene glycol alkylphenyl ether groups, such as the product Amercell Polymer HM-1500 (polyethylene glycol (15) nonylphenyl ether) sold by the company Amerchol;
- (ii) hydroxypropyl guar modified with groups comprising at least one fatty chain, such as the product Esaflor HM 22 (C<sub>22</sub> alkyl chain) sold by the company Lamberti, and the products Miracare XC95-3 (C<sub>14</sub> alkyl chain) and RE205-1 (C<sub>20</sub> alkyl chain) sold by the company Rhodia Chimie;
- (iii) polyether-polyurethanes comprising at least one fatty chain, such as C<sub>10</sub>-C<sub>30</sub> alkyl or alkenyl groups, for instance the products Elfacos T 210 and Elfacos T 212 sold by the company Akzo or the products Aculyn 44 and Aculyn 46 sold by the company Rohm & Haas;
- (iv) copolymers of vinylpyrrolidone and of hydrophobic fatty-chain monomers; examples that may be mentioned include:
  - the products Antaron V216 and Ganex V216 (vinylpyrrolidone/hexadecene copolymer) sold by the company I.S.P., and
  - the products Antaron V220 and Ganex V220 (vinylpyrrolidone/eicosene copolymer) sold by the company I.S.P.;

- (v) copolymers of C<sub>1</sub>-C<sub>6</sub> alkyl acrylates or methacrylates and of amphiphilic monomers comprising at least one fatty chain, such as the oxyethylenated methyl methacrylate/stearyl acrylate copolymer sold by the company Goldschmidt under the name Antil 208;
- 5 (vi) copolymers of hydrophilic acrylates or methacrylates and of hydrophobic monomers comprising at least one fatty chain, such as polyethylene glycol methacrylate/lauryl methacrylate copolymer.

10 It may be preferable that the hydrophilic nonionic polymeric associative thickener be selected from polyether-polyurethanes. The polyether-polyurethanes may have both at least one hydrophilic moiety and at least one hydrophobic moiety. More particularly, they may contain, in their polymer chain, both hydrophilic sequences most often of a polyoxyethylenated nature and hydrophobic sequences which may be aliphatic linkages alone and/or cycloaliphatic and/or aromatic linkages.

15 Preferably, these polyether-polyurethanes comprise at least two lipophilic hydrocarbon chains, having from 6 to 30 carbon atoms, preferably from 6 to 20, separated by a hydrophilic sequence, it being possible for the hydrocarbon chains to be pendent chains or chains at the end of a hydrophilic sequence. In particular, it is possible for one or more pendent chains to be envisaged. In addition, the polyether-polyurethanes may comprise a hydrocarbon chain at  
20 one end or at both ends of a hydrophilic sequence.

The polyether-polyurethanes may comprise polyblocks, in particular in triblock form. The hydrophobic sequences may be at each end of the polymer chain (for example: triblock  
25 copolymer with hydrophilic central sequence) or distributed both at the ends and in the polymer chain (for example: polyblock copolymers). The same polymers may also be in the form of graft units or may be star-shaped.

30 The hydrophilic nonionic polymeric associative thickeners can form a network in water in which the hydrophobic part connects to form quasi-micelles.

Therefore, the hydrophilic nonionic polymeric associative thickeners can increase viscosity or consistency of the composition according to the present invention. Thus, after application of the composition according to the present invention, it can recover the original elasticity of the  
35 composition quickly.

The nonionic polyether-polyurethanes containing a fatty chain may be triblock copolymers whose hydrophilic sequence is a polyoxyethylenated chain comprising from 50 to 1000 oxyethylenated groups.  
40

The nonionic polyether-polyurethanes comprise a urethane bond between the hydrophilic sequences, hence the origin of the name.

45 By extension, those whose hydrophilic sequences are linked by other chemical bonds to the hydrophobic sequences are also included among the nonionic polyether-polyurethanes containing a hydrophobic chain.

By way of examples of nonionic polyether-polyurethanes containing a hydrophobic chain which can be used in the present invention, it is also possible to use Rheolate® 205 containing  
50 a urea functional group sold by the company RHEOX or else the Rheolates® 208, 204 or 212,

as well as Acrysol RM 184®.

There may also be mentioned the product ELFACOS T210® containing a C<sub>12</sub>-C<sub>14</sub> alkyl chain and the product ELFACOS T212® containing a C<sub>18</sub> alkyl chain from AKZO.

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The product DW 1206B® from ROHM & HAAS containing a C<sub>20</sub> alkyl chain and with a urethane bond, sold at 20% dry matter content in water, may also be used.

It is also possible to use solutions or dispersions of these polymers in particular in water or in an aqueous-alcoholic medium. By way of examples of such polymers, there may be mentioned Rheolate® 255, Rheolate® 278 and Rheolate® 244 sold by the company RHEOX. It is also possible to use the product DW 1206F and DW 1206J provided by the company ROHM & HAAS.

10 The above-described polyether-polyurethanes which can be used can also be chosen from those described in the article by G. Fønnum, J. Bakke and Fk. Hansen-Colloid Polym. Sci 271, 380-389 (1993).

As the above-described polyether-polyurethanes, mention may be made of polyether-polyurethanes comprising in their chain at least one polyoxyethylenated hydrophilic block and at least one of hydrophobic blocks containing at least one sequence chosen from aliphatic sequences, cycloaliphatic sequences, and aromatic sequences.

It may be preferable that the polyether-polyurethanes comprise at least two hydrocarbon-based lipophilic chains having from 8 to 30 carbon atoms, separated by a hydrophilic block, and wherein the hydrocarbon-based chains are chosen from pendent chains and chains at the end of the hydrophilic block.

According to a specific form of the present invention, use will be made of a polyurethane/polyether that may be obtained by polycondensation of at least three compounds comprising (i) at least one polyethylene glycol comprising from 150 to 180 mol of ethylene oxide, (ii) a polyoxyethylenated stearyl alcohol comprising 100 mol of ethylene oxide, and (iii) a diisocyanate.

Such polyurethane/polyethers are sold especially by the company Elementis under the name Rheolate FX 1100® and Rheoluxe 811®, which is a polycondensate of polyethylene glycol containing 136 mol of ethylene oxide, of stearyl alcohol polyoxyethylenated with 100 mol of ethylene oxide and of hexamethylene diisocyanate (HDI) with a weight-average molecular weight of 40000 (INCI name: PEG-136/Steareth-100/HDI Copolymer).

According to another specific form of the present invention, use will be made of a polyurethane/polyether that may be obtained by polycondensation of at least three compounds comprising (i) at least one polyethylene glycol comprising from 150 to 180 mol of ethylene oxide, (ii) stearyl alcohol or decyl alcohol, and (iii) at least one diisocyanate.

Such polyurethane/polyethers are sold in particular by the company Rohm & Haas under the names Aculyn 46® and Aculyn 44®.

Aculyn 46® having the INCI name: PEG-150/Stearyl Alcohol/SMDI Copolymer, is a polycondensate of polyethylene glycol comprising 150 or 180 mol of ethylene oxide, of

50

stearyl alcohol and of methylenebis(4-cyclohexyl isocyanate) (SMDI) at 15% by weight in a matrix of maltodextrin (4%) and water (81%) (INCI name: PEG-150/Stearyl Alcohol/SMDI Copolymer).

5 Aculyn 44® (PEG-150/Decyl Alcohol/SMDI Copolymer) is a polycondensate of polyethylene glycol comprising 150 or 180 mol of ethylene oxide, of decyl alcohol and of methylenebis(4-cyclohexyl isocyanate) (SMDI) at 35% by weight in a mixture of propylene glycol (39%) and water (26%) (INCI name: PEG-150/Decyl Alcohol/SMDI Copolymer).

10 As the polyether-polyurethanes, it may be preferable to use a compound represented by the following formula (1):



15 wherein  $R^1$  represents a hydrocarbon group,  $R^2$  and  $R^4$  independently represent alkylene groups having 2 to 4 carbon atoms, which alkylene groups may be identical or different from each other, or a phenylethylene group,  $R^3$  represents a hydrocarbon group, which may optionally have a urethane bond,  $R^5$  represents a branched chain or secondary hydrocarbon group,  $m$  represents a number of at least 2,  $h$  represents a number of at least 1,  $k$  represents a number within the range of 1 to 500, and  $n$  represents a number within the range of 1 to 200.

The hydrophobically modified polyurethane that is represented by the general formula (1) shown above is obtained by, for example, reacting at least one polyether polyol that is represented by the formula  $R^1-[(O-R^2)_k-OH]_m$ , at least one polyisocyanate that is represented by the formula  $R^3-(NCO)_{h+1}$ , and at least one polymonoalcohol that is represented by the formula  $HO-(R^4-O)_n-R^5$ .

25 In such cases,  $R^1$  to  $R^5$  in the general formula (1) are determined by the compounds  $R^1-[(O-R^2)_k-OH]_m$ ,  $R^3-(NCO)_{h+1}$  and  $HO-(R^4-O)_n-R^5$ . The loading ratios among the three compounds are not limited particularly and should preferably be such that the ratio of the isocyanate group derived from the polyisocyanate to the hydroxyl group derived from the polyether polyol and the polyether monoalcohol is selected within the range of NCO/OH of between 0.8:1 and 1.4:1.

35 The polyether polyol compound that is represented by the formula  $R^1-[(O-R^2)_k-OH]_m$  and that may be used preferably for obtaining the polyether-polyurethane represented by the general formula (1) may be obtained from addition polymerization of an  $m$ -hydric polyol with an alkylene oxide, such as ethylene oxide, propylene oxide, butylene oxide, or epichlorohydrin, or with styrene oxide, and the like.

40 The polyols should preferably be di- to octa-hydric polyols. Examples of the di- to octa-hydric polyols include dihydric alcohols, such as ethylene glycol, propylene glycol, butylene glycol, hexamethylene glycol, and neopentyl glycol; trihydric alcohols, such as glycerol, trioxy isobutane, 1,2,3-butanetriol, 1,2,3-pentanetriol, 2-methyl-1,2,3-propanetriol, 2-methyl-2,3,4-butanetriol, 2-ethyl-1,2,3-butanetriol, 2,3,4-pentanetriol, 2,3,4-hexanetriol, 4-propyl-3,4,5-heptanetriol, 2,4-dimethyl-2,3,4-pentanetriol, pentamethylglycerol, pentaglycerol, 1,2,4-butanetriol, 1,2,4-pentanetriol, trimethylolpropane, and trimethylolpropane; tetrahydric alcohols, such as pentaerythritol, 1,2,3,4-pentanetetrol, 2,3,4,5-hexanetetrol, 1,2,4,5-pentanetetrol, and 1,3,4,5-hexanetetrol; pentahydric alcohols, such as adonitol, arabitol, and xylitol; hexahydric alcohols, such as dipentaerythritol, sorbitol, mannitol, and iditol; and

octahydric alcohols, such as sucrose.

Also,  $R^2$  is determined by the alkylene oxide, styrene oxide, or the like, which is subjected to the addition. Particularly, for availability and excellent effects, an alkylene oxide having 2 to 4 carbon atoms, or styrene oxide is preferable.

The alkylene oxide, styrene oxide, or the like, to be subjected to the addition may be subjected to single polymerization, or random polymerization or block polymerization of at least two members. The procedure for the addition may be a conventional procedure. Also, the polymerization degree  $k$  may be selected within the range of 0 to 1,000, preferably within the range of 1 to 500, and more preferably within the range of 10 to 200. Further, the ratio of the ethylene group occupying  $R^2$  should preferably be within the range of 50 to 100 mass % with respect to the total quantity of  $R^2$ . In such cases, the hydrophilic nonionic polymeric associative thickener appropriate for the purposes of the present invention is obtained.

Furthermore, the molecular weight of the polyether polyol compound that is represented by the formula  $R^1-[(O-R^2)_k-OH]_m$  should preferably be selected within the range of 500 to 100,000, and should more preferably be selected within the range of 1,000 to 50,000.

The polyisocyanate that is represented by the formula  $R^3-(NCO)_{n+1}$  and that may be used preferably for obtaining the hydrophobically modified polyether urethane represented by the general formula (1) employed in accordance with the present invention is not limited particularly in so far as the polyisocyanate has at least two isocyanate groups in the molecule. Examples of the polyisocyanates include aliphatic diisocyanates, aromatic diisocyanates, alicyclic diisocyanates, biphenyl diisocyanate, phenylmethane diisocyanate, phenylmethane triisocyanate, and phenylmethane tetraisocyanate.

Also, it is possible to employ dimers and trimers (isocyanurate bonds) of the above-enumerated polyisocyanates. Further, it is possible to employ biuret obtained by a reaction with an amine.

Furthermore, it is possible to employ a polyisocyanate having a urethane bond obtained by a reaction of the aforesaid polyisocyanate compound and a polyol. As the polyol, di- to octahydric polyols are preferable, and the above-enumerated polyols are preferable. In cases where a tri- or higher-hydric polyisocyanate is used as the polyisocyanate that is represented by the formula  $R^3-(NCO)_{n+1}$ , it is preferable to employ the aforesaid polyisocyanate having the urethane bond.

The polyether monoalcohol that is represented by the formula  $HO-(R^4-O)_n-R^5$  and that may be used preferably for obtaining the hydrophobically modified polyether urethane represented by the general formula (1) employed in accordance with the present invention is not limited particularly in so far as the polyether monoalcohol is a polyether of a straight chain, branched chain, or secondary monohydric alcohol. The polyether monoalcohol may be obtained by addition polymerization of the straight chain, branched chain, or secondary monohydric alcohol with an alkylene oxide, such as ethylene oxide, propylene oxide, butylene oxide, or epichlorohydrin, or with styrene oxide, and the like.

The compound represented by the general formula (1) may be produced by, for example, heating at a temperature of 80 to 90°C for 1 to 3 hours and thereby causing a reaction to occur in the same manner as that in the ordinary reaction of a polyether and an isocyanate.

As the compound represented by the general formula (1), polyethyleneglycol-240/decyltetradeceth-20/hexamethylene diisocyanate copolymer is preferable. The polyethyleneglycol-240/decyltetradeceth-20/hexamethylene diisocyanate copolymer is referred to also as PEG-240/HDI copolymer bis-decyltetradeceth-20 ether.

According to the present invention, it is preferable that the hydrophilic nonionic polymeric associative thickener be selected from Steareth-100/PEG-136/HDI Copolymer sold by the company Rheox under the name of Rheolate FX 1100, PEG-240/HDI Copolymer Bis-decyltetradeceth-20 ether sold by the company Asahi Denka under the name of Adekanol GT-700, and mixtures thereof.

#### {Hydrophilic Nonionic Polysaccharide Thickeners}

The hydrophilic nonionic polysaccharide thickener here means a hydrophilic nonionic thickener based on at least one polysaccharide, preferably as a backbone of the thickener.

The hydrophilic nonionic polysaccharide thickener may be chosen from those described, for example, in "Encyclopedia of Chemical Technology", Kirk-Othmer, Third Edition, 1982, volume 3, pp. 896-900, and volume 15, pp. 439-458, in "Polymers in Nature" by E. A. MacGregor and C. T. Greenwood, published by John Wiley & Sons, Chapter 6, pp. 240-328, 1980, and in "Industrial Gums--Polysaccharides and their Derivatives", edited by Roy L. Whistler, Second Edition, published by Academic Press Inc., the content of these three publications being entirely incorporated by reference.

Specifically, the hydrophilic nonionic polysaccharide thickener may be chosen, for example, from glucans, modified and unmodified starches (such as those derived, for example, from cereals, for instance wheat, corn or rice, from vegetables, for instance yellow pea, and tubers, for instance potato or cassava), amylose, amylopectin, glycogen, dextrans, celluloses and derivatives thereof (methylcelluloses, hydroxyalkylcelluloses, ethyl hydroxyethylcelluloses, and carboxymethylcelluloses), mannans, xylans, lignins, arabans, galactans, galacturonans, chitin, chitosans, glucuronoxylans, arabinoxylans, xyloglucans, glucomannans, pectic acids and pectins, alginic acid and alginates, arabinogalactans, carrageenans, agars, glycosaminoglucans, gum arabics, gum tragacanth, ghatti gums, karaya gums, carob gums, galactomannans, such as guar gums, and nonionic derivatives thereof (e.g., hydroxypropyl guar) and xanthan gums, and mixtures thereof.

As the hydrophilic nonionic polysaccharide thickener, for example, starches, guar gums, inulin, and celluloses and derivatives thereof may preferably be used.

Among the starches that may be used, mention may be made, for example, of macromolecules in the form of polymers comprising elemental moieties that are anhydroglucose units. The number of these moieties and their assembly make it possible to distinguish between amylose (linear polymer) and amylopectin (branched polymer). The relative proportions of amylose and of amylopectin, and also their degree of polymerization, can vary as a function of the botanical origin of the starches.

The botanical origin of the starch molecules used may be cereals or tubers. Thus, the starches can be, for example, chosen from corn starch, rice starch, cassava starch, tapioca starch, barley starch, potato starch, wheat starch, sorghum starch and pea starch.

Starches are generally in the form of a white powder which is insoluble in cold water and which has an elementary particle size ranging from 3 to 100 microns.

- 5 The starches may optionally be C<sub>1</sub>-C<sub>6</sub> hydroxyalkylated or C<sub>1</sub>-C<sub>6</sub> acylated (such as acetylated). The starches may also have undergone heat treatments.

10 Distarch phosphates or of compounds rich in distarch phosphate, for instance the products sold under the references Prejel VA-70-T AGGL (gelatinized hydroxypropylated cassaya distarch phosphate) or Prejel TK1 (gelatinized cassaya distarch phosphate) or Prejel 200 (gelatinized acetylated cassaya distarch phosphate) by the company Avebe, or Structure ZEA from National Starch (hydroxypropylated corn distarch phosphate), may also be used.

The guar gums may be modified or unmodified.

15 The unmodified guar gums are, for example, the products sold under the name Vidogum GH 175 by the company Unipectine and under the names Meyro-Guar 50 and Jaguar C by the company Meyhall.

- 20 The modified nonionic guar gums are, for example, modified with C<sub>1</sub>-C<sub>6</sub> hydroxyalkyl groups.

Among hydroxyalkyl groups, mention may be made, for example, of hydroxymethyl, hydroxyethyl, hydroxypropyl and hydroxybutyl groups.

25 These guar gums are well known in the state of the art and can be prepared, for example, by reacting corresponding alkene oxides, such as propylene oxides, with guar gum so as to obtain a guar gum modified with hydroxypropyl groups.

30 The degree of hydroxyalkylation, which corresponds to the number of alkylene oxide molecules consumed by the number of free hydroxyl functions present on the guar gum, may, for example, range from 0.4 to 1.2.

Such nonionic guar gums optionally modified with hydroxyalkyl groups are sold, for example, under the trade names Jaguar HP-8 COS, Jaguar HP-60, Jaguar HP-120, and Jaguar HP-120 by the company Solvay.

35 Among the celluloses that are used are, for example, hydroxyethylcellulose and hydroxypropylcelluloses. Mention may be made of the products sold under the names Klucel EF, Klucel H, Klucel MF and Klucel G by the company Ashland.

- 40 Alternatively, as the hydrophilic nonionic polysaccharide thickener, polysaccharides derived from microorganisms may also preferably be used.

The polysaccharide derived from microorganisms means polysaccharide produced by microorganisms such as germ or bacteria.

45 The polysaccharide derived from microorganisms is not polysaccharide derived from plants. Thus, it may be preferable that polysaccharide derived from microorganisms is not based on cellulose.

- 50 As examples of the polysaccharide derived from microorganisms, mention may be made of

cardollan, xanthan gum, Jellan gum, dextran, pullulan, sclerotium gum, and mixtures thereof.

It may be preferable that the polysaccharide derived from microorganisms be selected from the group consisting of sclerotium gum, xanthan gum and mixtures thereof.

5 It may be even more preferable that the (e) texture agent be selected from the group consisting of agar, guar gum, hydroxypropyl guar gum, sclerotium gum and PEG-240/HDI Copolymer Bis-Decyltetradeceth-20 Ether.

10 The amount of the (e) texture agent in the composition according to the present invention may be 0.01% by weight or more, preferably 0.05% by weight or more, and more preferably 0.1% by weight or more, relative to the total weight of the composition.

15 The amount of the (e) texture agent in the composition according to the present invention may be 15% by weight or less, preferably 10% by weight or less, and more preferably 5% by weight or less, relative to the total weight of the composition.

20 The amount of the (e) texture agent in the composition according to the present invention may be from 0.01% to 15% by weight, preferably from 0.05% to 10% by weight, and more preferably from 0.1% to 5% by weight, relative to the total weight of the composition.

[Cosmetic Active Ingredient]

25 The composition according to the present invention may comprise at least one (additional) cosmetic active ingredient. There is no limitation to the additional cosmetic active ingredient as long as it is different from the (a) polyion complex and the (b) fatty alcohol, as well as, if present, the (d) fatty material and the (e) texture agent. Two or more additional cosmetic active ingredients may be used in combination. Thus, a single type of additional cosmetic active ingredient or a combination of different types of additional cosmetic active ingredients  
30 may be used.

35 Among the additional cosmetic active ingredients to be used, mention may be made of hydrophobic or water-insoluble UV filters, anti-oxidants, cleansing agents, free radical scavengers, moisturizers, whitening agents, liporegulators, anti-acne agents, antidandruff agents, anti-aging agents, softeners, anti-wrinkle agents, keratolytic agents, fresheners, antibacterial agents, antifungal agents, antiperspirants, deodorants, skin conditioners, anesthetics, nourishing agents, and sebum absorbers or moisture absorbers.

40 The composition according to the present invention may comprise the additional cosmetic active ingredient(s) in an amount of from 0.01% to 30% by weight, preferably from 0.1% to 20% by weight, and more preferably from 1% to 10% by weight, relative to the total weight of the composition.

[pH]

45 The pH of the composition according to the present invention may be from 3 to 9, preferably from 3.5 to 8.5, and more preferably from 4 to 8.

50 At a pH of from 3 to 9, the (a) polyion complex can be very stable.

The pH of the composition according to the present invention may be adjusted by adding at least one alkaline agent and/or at least one acid, other than the crosslinker, to be incorporated into the (a) polyion complex. The pH of the composition according to the present invention may also be adjusted by adding at least one buffering agent.

5

(Alkaline Agent)

The composition according to the present invention may comprise at least one alkaline agent. Two or more alkaline agents may be used in combination. Thus, a single type of alkaline agent or a combination of different types of alkaline agents may be used.

10

The alkaline agent may be an inorganic alkaline agent. It is preferable that the inorganic alkaline agent be selected from the group consisting of ammonia; alkaline metal hydroxides; alkaline earth metal hydroxides; alkaline metal phosphates and monohydrogenophosphates such as sodium phosphate or sodium monohydrogen phosphate.

15

As examples of the inorganic alkaline metal hydroxides, mention may be made of sodium hydroxide and potassium hydroxide. As examples of the alkaline earth metal hydroxides, mention may be made of calcium hydroxide and magnesium hydroxide. As an inorganic alkaline agent, sodium hydroxide is preferable.

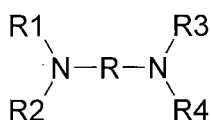
20

The alkaline agent may be an organic alkaline agent. It is preferable that the organic alkaline agent be selected from the group consisting of monoamines and derivatives thereof; diamines and derivatives thereof; polyamines and derivatives thereof; basic amino acids and derivatives thereof; oligomers of basic amino acids and derivatives thereof; polymers of basic amino acids and derivatives thereof; urea and derivatives thereof; and guanidine and derivatives thereof.

25

As examples of the organic alkaline agents, mention may be made of alkanolamines such as mono-, di- and tri-ethanolamine, and isopropanolamine; urea, guanidine and their derivatives; basic amino acids such as lysine, ornithine or arginine; and diamines such as those described in the structure below:

30



wherein R denotes an alkylene such as propylene optionally substituted by a hydroxyl or a C<sub>1</sub>-C<sub>4</sub> alkyl radical, and R<sub>1</sub>, R<sub>2</sub>, R<sub>3</sub> and R<sub>4</sub> independently denote a hydrogen atom, an alkyl radical or a C<sub>1</sub>-C<sub>4</sub> hydroxyalkyl radical which may be exemplified by 1,3-propanediamine and derivatives thereof. Arginine, urea and monoethanolamine are preferable.

35

The alkaline agent(s) may be used in a total amount of from 0.01% to 10% by weight, preferably from 0.05% to 5% by weight, more preferably from 0.1% to 1% by weight, relative to the total weight of the composition, depending on their solubility.

40

(Acid)

The composition according to the present invention may comprise at least one acid other than the crosslinker to be incorporated into the (a) particle. Two or more acids may be used in combination. Thus, a single type of acid or a combination of different types of acids may be used.

45

As the acid, mention may be made of any inorganic or organic acids, preferably inorganic acids, which are commonly used in cosmetic products. A monovalent acid and/or a polyvalent acid may be used. A monovalent acid such as citric acid, lactic acid, sulfuric acid, phosphoric acid and hydrochloric acid (HCl) may be used. Lactic acid may be preferable.

The acid(s) may be used in a total amount of from 0.01% to 10% by weight, preferably from 0.05% to 5% by weight, more preferably from 0.1% to 1% by weight, relative to the total weight of the composition, depending on their solubility.

(Buffering Agent)

The composition according to the present invention may comprise at least one buffering agent. Two or more buffering agents may be used in combination. Thus, a single type of buffering agent or a combination of different types of buffering agents may be used.

As the buffering agent, mention may be made of an acetate buffer (for example, acetic acid + sodium acetate), a phosphate buffer (for example, sodium dihydrogen phosphate + di-sodium hydrogen phosphate), a citrate buffer (for example, citric acid + sodium citrate), a borate buffer (for example, boric acid + sodium borate), a tartrate buffer (for example, tartaric acid + sodium tartrate dihydrate), Tris buffer (for example, tris(hydroxymethyl)aminomethane), Hepes buffer (4-(2-hydroxyethyl)-1-piperazineethanesulfonic acid).

[Optional Additives]

The composition according to the present invention may comprise, in addition to the aforementioned components, components typically employed in cosmetics, for example, surfactants or emulsifiers, preservatives such as phenoxyethanol and caprylyl glycol, organic non-volatile solvents, natural extracts derived from vegetables, and the like, within a range which does not impair the effects of the present invention.

It is preferable that the composition according to the present invention comprise no pigments or dyes.

The composition according to the present invention may comprise the above optional additive(s) in an amount of from 0.01% to 50% by weight, preferably from 0.05% to 30% by weight, and more preferably from 0.1% to 10% by weight, relative to the total weight of the composition.

However, the composition according to the present invention may include a very limited amount of surfactant(s) or emulsifier(s). The amount of the surfactant(s) or emulsifier(s) in the composition according to the present invention may be 1% by weight or less, preferably 0.1% by weight or less, and more preferably 0.01% by weight or less, relative to the total weight of the composition. It is in particular preferable that the composition according to the present invention include no surfactant or emulsifier.

[Composition]

Since the composition according to the present invention comprises (c) water, the composition according to the present invention can comprise at least one aqueous phase.

The aqueous phase may comprise at least one C<sub>2</sub>-C<sub>6</sub> monohydric alcohol. Two or more C<sub>2</sub>-C<sub>6</sub> monohydric alcohols may be used in combination.

5 The C<sub>2</sub>-C<sub>6</sub> monohydric alcohol suitable for the present invention may comprise from 2 to 5 carbon atoms, preferably from 2 to 4 carbon atoms, such as ethanol, isopropanol, propanol or butanol.

10 Ethanol and isopropanol, and preferably ethanol, are very particularly suitable for the present invention.

15 The amount of the C<sub>2</sub>-C<sub>6</sub> monohydric alcohol in the composition according to the present invention may be 20% by weight or less, preferably 15% by weight or less, and more preferably 10% by weight or less, relative to the total weight of the composition. On the other hand, the amount of the C<sub>2</sub>-C<sub>6</sub> monohydric alcohol in the composition according to the present invention is 5% by weight or more, preferably 6% by weight or more, and more preferably 7% by weight or more, relative to the total weight of the composition. For example, the amount of the C<sub>2</sub>-C<sub>6</sub> monohydric alcohol may be from 5% to 20% by weight, preferably from 6% to 15% by weight, and more preferably from 7% to 10% by weight, in relation to the total weight of the composition.

25 The aqueous phase may comprise polyhydric alcohols containing 2 to 8 carbon atoms such as propylene glycol, ethylene glycol, 1,3-butylene glycol, dipropylene glycol, diethylene glycol, pentyleneglycol, hexyleneglycol, glycerin, and mixtures thereof.

30 The amount of the polyhydric alcohol(s) such as glycols, if present, in the aqueous phase according to the present invention may range from 0.1% to 15% by weight, preferably from 0.5% to 12% by weight, and more preferably from 1% to 8% by weight, relative to the total weight of the composition.

35 As the composition according to the present invention comprises (b) at least one fatty alcohol and, if present, (d) at least one fatty material, the composition according to the present invention can comprise at least one fatty phase.

40 If the (d) fatty material comprises oil, the composition according to the present invention can easily be in the form of an emulsion, an O/W emulsion or a W/O emulsion. It is preferable that the composition according to the present invention be in the form of an O/W emulsion, because it can provide a fresh sensation due to the (c) water which forms the outer phase thereof.

45 It may be more preferable that the amount of the surfactant(s) or emulsifier(s) in the emulsion, in particular an O/W emulsion, be 1% by weight or less, preferably 0.1% by weight or less, and more preferably 0.01% by weight or less, relative to the total weight of the composition, because the surfactant(s) may negatively affect water-resistance. It is in particular preferable that the emulsion, in particular an O/W emulsion include no surfactant or emulsifier.

50 The composition according to the present invention may be intended to be used as a cosmetic composition. Thus, the cosmetic composition according to the present invention may be intended for application onto keratin fibers. Keratin fibers here means fibers containing keratin as a main constituent element, and examples thereof include hair, eyelash, eyebrow,

and the like. It is preferable that the cosmetic composition according to the present invention be used for a cosmetic process for keratin fibers, and more preferably hair.

5 The composition according to the present invention can preferably be used as a leave-on or rinse-off cosmetic composition for keratin fibers such as hair. The "leave-on" here means that the composition according to the present invention is not removed from keratin fibers after being applied onto the keratin fibers. The "rinse-off" here means that the composition according to the present invention is removed, by rinsing, from keratin fibers after being applied onto the keratin fibers. Water can be used for rinsing. Even after rinsing off, the  
10 composition according to the present invention can remain to form a film or a coating on keratin fibers such as hair.

15 The composition according to the present invention can be prepared by mixing the above essential and optional ingredients in accordance with any of the processes which are well known to those skilled in the art.

#### [Process and Use]

20 The present invention also relates to a cosmetic process for keratin fibers such as hair, comprising: applying to the keratin fibers the composition according to the present invention; and drying the composition to form a cosmetic film on the keratin fibers,  
and  
25 a process for preparing a film, preferably a cosmetic film, comprising: applying onto keratin fibers such as skin, the composition according to the present invention; and drying the composition.

30 Furthermore, the present invention also relates to a film, preferably a cosmetic film, comprising:  
(a) at least one polyion complex comprising at least one ionic polymer selected from the group consisting of cationic polymers, anionic polymers, amphoteric polymers, and mixtures thereof,  
and  
35 at least one crosslinker selected from the group consisting of non-polymeric acids having two or more pKa values or salt(s) thereof, non-polymeric bases having two or more pKb values or salt(s) thereof, and mixtures thereof; and  
(b) at least one fatty alcohol.

40 The present invention may also relate to a use of the composition according to the present invention for the preparation of a cosmetic film on keratin fibers such as hair.

The cosmetic process here means a non-therapeutic cosmetic method for caring and/or styling keratin fibers such as hair.

45 The above film, preferably the cosmetic film, is resistant to water with a pH of 7 or less, and is removable with water with a pH of more than 7, preferably 8 or more, and more preferably 9 or more.

In other words, the above film, preferably the cosmetic film, can be water-resistant under neutral or acidic conditions such as a pH of 7 or less, preferably in a range of 6 or more and 7 or less, and more preferably in a range of 5 or more and 7 or less, while the above film preferably the cosmetic film, can be removed under alkaline conditions such as a pH of more than 7, preferably 8 or more, and more preferably 9 or more. The upper limit of the pH is preferably 13, more preferably 12, and even more preferably 11.

Accordingly, the above film, preferably the cosmetic film, can be water-resistant, and therefore, it can remain on keratin fibers such as hair even if the surface of the keratin fibers is wet due to, for example sweat and rain. On the other hand, the above film, preferably the cosmetic film, can be easily removed from keratin fibers such as hair under alkaline conditions. Therefore, the above film, preferably the cosmetic film, is difficult to remove with water, while it can be easily removed with, for example, a soap which can provide alkaline conditions.

Furthermore, the above film may have cosmetic effects such as absorbing or adsorbing malodor, changing the appearance of keratin fibers such as hair, changing the feel to the touch of the keratin fibers, and/or protecting the keratin fibers from, for example, dirt or pollutant, due to the properties of the polyion complex in the film, even if the film does not include any cosmetic active ingredient.

If the above film includes at least one additional cosmetic active ingredient other than the (b) fatty alcohol and, if present, the (d) fatty material, the film can have cosmetic effects provided by the additional cosmetic active ingredient(s).

The present invention may also relate to a use of (b) at least one fatty alcohol in a cosmetic composition for keratin fibers such as hair, comprising:

- (a) at least one polyion complex comprising
  - at least one ionic polymer selected from the group consisting of cationic polymers, anionic polymers, amphoteric polymers, and mixtures thereof,
  - and
  - at least one crosslinker selected from the group consisting of non-polymeric acids having two or more pKa values or salt(s) thereof, non-polymeric bases having two or more pKb values or salt(s) thereof, and mixtures thereof;

and

- (c) water

in order for the composition to provide keratin fibers such as hair with improved textures in terms of all of smoothness, softness, evenness of coating and tip moisturization. The composition may further comprise (d) at least one fatty material other than the (b) fatty alcohol.

The explanations regarding the ingredients (a) to (d) in the composition according to the present invention can apply to those in the use according to the present invention.

## EXAMPLES

The present invention will be described in a more detailed manner by way of examples. However, they should not be construed as limiting the scope of the present invention.

### Example 1 and Comparative Example 1-2

## [Preparations]

5 Compositions according to Example 1 and Comparative Examples 1-2 were prepared as shown below.

## (Example 1)

10 0.75 g of hydroxypropyl guar was dissolved in about 92 g of water to obtain a mixture of hydroxypropyl guar and water. The mixture was heated with a water bath to 70°C. Then, 0.259 g of polyquaternium-67 was added to the mixture and dispersed completely. Heating the mixture of hydroxypropyl guar, polyquaternium-67 and water with the water bath was then stopped, and the water bath was removed. Then, 0.616 g of a 40 wt% polyquaternium-6 aqueous solution was added to the mixture. Further, 0.035 g of sodium hydroxide and 0.21 g  
15 of a 50 wt% phytic acid aqueous were added to the mixture. Then, the mixture was homogenized with a homogenizer to obtain a composition including a polyion complex. Furthermore, 0.30 g of caprylyl glycol was added into the above mentioned mixture. Then, the mixture was homogenized with a homogenizer.

20 Next, a premix, which had been composed of 2.00 g of cetearyl alcohol, 1.00 g of sunflower oil, and 2.00 g of isopropyl myristate, was heated to 80°C and was added to the mixture obtained by the above steps and homogenized with a homogenizer at high mixing speed. Also, 0.50 g of phenoxyethanol was then added to the mixture and homogenized with a homogenizer at high mixing speed.

25 Next, the pH of the mixture thus obtained was adjusted to about 7.5 by adding NaOH, followed by homogenizing with a homogenizer.

30 A uniform composition in the form of a milk was obtained.

## (Comparative Example 1)

35 0.75 g of hydroxypropyl guar was dissolved in about 92 g of water to obtain a mixture of hydroxypropyl guar and water. The mixture was heated with a water bath to 70°C. Then, 0.259 g of polyquaternium-67 was added to the mixture and dispersed completely. Heating the mixture of hydroxypropyl guar, polyquaternium-67 and water with the water bath was then stopped, and the water bath was removed. Then, 0.616 g of a 40 wt% polyquaternium-6 aqueous solution was added to the mixture. Furthermore, 0.30 g of caprylyl glycol was added into the above mentioned mixture. Then, the mixture was homogenized with a homogenizer.

40 Next, a premix, which had been composed of 2.00 g of cetearyl alcohol, 1.00 g of sunflower oil, and 2.00 g of isopropyl myristate, was heated to 80°C and was added to the mixture obtained by the above steps and homogenized with a homogenizer at high mixing speed. Also, 0.50 g of phenoxyethanol was then added to the mixture and homogenized with a  
45 homogenizer at high mixing speed.

Next, the pH of the mixture thus obtained was adjusted to about 7.5 by adding a 90wt% lactic acid aqueous solution, followed by homogenizing with a homogenizer.

50 A uniform composition in the form of a milk was obtained.

## (Comparative Example 2)

0.75 g of hydroxypropyl guar was dissolved in about 92 g of water to obtain a mixture of  
5 hydroxypropyl guar and water. The mixture was heated with a water bath to 70°C. Then,  
0.259 g of polyquaternium-67 was added to the mixture and dispersed completely. Heating  
the mixture of hydroxypropyl guar, polyquaternium-67 and water with the water bath was  
then stopped, and the water bath was removed. Then, 0.616 g of a 40 wt% polyquaternium-6  
10 aqueous solution was added to the mixture. Further, 0.035 g of sodium hydroxide and 0.21 g  
of a 50 wt% phytic acid aqueous were added to the mixture. Then, the mixture was  
homogenized with a homogenizer to obtain a composition including a polyion complex.  
Furthermore, 0.30 g of caprylyl glycol was added into the above mentioned mixture. Then,  
the mixture was homogenized with a homogenizer.

15 Next, a premix, which had been composed of 2.00 g of sunflower oil, 1.00 g of shea butter  
and 2.00 g of isopropyl myristate, was heated to 80°C and was added to the mixture obtained  
by the above steps and homogenized with a homogenizer at high mixing speed. Also, 0.50 g  
of phenoxyethanol was then added to the mixture and homogenized with a homogenizer at  
20 high mixing speed.

Next, the pH of the mixture thus obtained was adjusted to about 7.5 by adding NaOH,  
followed by homogenizing with a homogenizer.

A uniform composition in the form of a milk was obtained.

25 The formulation of the composition according to Example 1 and Comparative Examples 1-2  
is shown in Table 1. The numerical values for the amounts of the components shown in Table  
1 are all based on “% by weight” as raw materials.

30

Table 1

		Ex. 1	Comp. Ex. 1	Comp. Ex. 2
Polyquaternium-67		0.259	0.259	0.259
40wt% Polyquaternium-6 Aqueous Solution		0.616	0.616	0.616
Sodium Hydroxide		0.035	-	0.035
50wt% Phytic Acid Aqueous Solution		0.21	-	0.21
Cetearyl Alcohol		2.00	2.00	-
Sunflower Oil		1.00	1.00	2.00
Shea Butter		-	-	1.00
Isopropyl Myristate		2.00	2.00	2.00
Hydroxypropyl Guar		0.75	0.75	0.75
Phenoxyethanol		0.50	0.50	0.50
Caprylyl Glycol		0.30	0.30	0.30
Sodium Hydroxide		qs pH 7.5	-	qs pH 7.5
Lactic Acid		-	qs pH 7.5	-
Water		qsp 100	qsp 100	qsp 100
pH		7.5	7.5	7.5
Fatty Phase (wt%)		5	5	5
Leave-on	Softness of Dried Hair	Good	Poor	Poor
	Smoothness of Dried Hair	Very Good	Very Good	Good
	Evenness of Coating	Very Good	Very Good	Poor
	Tip moisturization	Very Good	Very Good	Poor
Rinse-off	Softness of Dried Hair	Good	Poor	Good
	Smoothness of Dried Hair	Very Good	Very Good	Good
	Evenness of Coating	Very Good	Good	Good
	Tip moisturization	Very Good	Good	Poor

5 [Evaluations]

{Leave-on}

10 Using a hair swatch, the textures of softness and smoothness of dried hair, evenness of coating and tip moisturization, after the application of each of the compositions according to Example 1 and Comparative Examples 1-2 onto the hair swatch followed by drying, were assessed by 6

panelists in accordance with the following criteria. The benchmark means a hair swatch to which the composition has not been applied.

Very Good: 4-6 panelists considered better than the benchmark

5 Good: 2 or 3 panelists considered better than the benchmark

Poor: 0 or 1 panelist considered better than the benchmark

The results are shown in Table 1.

10 The composition according to Example 1 was able to provide hair with better texture in terms of all of smoothness and softness, evenness of coating and tip moisturization than the composition according to Comparative Examples 1-2.

{Rinse-off}

15 Using a hair swatch, the textures of softness and smoothness of dried hair, evenness of coating and tip moisturization, after the application of each of the compositions according to Example 1 and Comparative Examples 1-2 onto the hair swatch followed by rinsing and drying, were assessed by 6 panelists in accordance with the following criteria. The benchmark means a  
20 hair swatch to which the composition has not been applied.

Very Good: 4-6 panelists considered better than the benchmark

Good: 2 or 3 panelists considered better than the benchmark

Poor: 0 or 1 panelist considered better than the benchmark

25

The results are shown in Table 1.

30 The composition according to Example 1 was able to provide hair with better texture in terms of all of smoothness and softness, evenness of coating and tip moisturization than the composition according to Comparative Examples 1-2.

## CLAIMS

1. A composition, preferably a cosmetic composition, and more preferably a cosmetic composition for keratin fibers such as hair, comprising:
- 5 (a) at least one polyion complex comprising  
at least one ionic polymer selected from the group consisting of cationic polymers, anionic polymers, amphoteric polymers, and mixtures thereof,  
and  
10 at least one crosslinker selected from the group consisting of non-polymeric acids having two or more pKa values or salt(s) thereof, non-polymeric bases having two or more pKb values or salt(s) thereof, and mixtures thereof;
- (b) at least one fatty alcohol; and  
(c) water.
- 15 2. The composition according to Claim 1, wherein the (a) polyion complex(es) comprise(s)
- (i) at least one cationic polymer and at least one non-polymeric acid having two or more pKa values or salt(s) thereof;
- 20 (ii) at least one cationic polymer, at least one anionic polymer, and at least one non-polymeric acid having two or more pKa values or salt(s) thereof and/or at least one non-polymeric base having two or more pKb values or salt(s) thereof,
- (iii) at least one cationic polymer, at least one amphoteric polymer, and at least one non-polymeric acid having two or more pKa values or salt(s) thereof and/or at least one non-polymeric base having two or more pKb values or salt(s) thereof,
- 25 (iv) at least one cationic polymer, at least one anionic polymer, at least one amphoteric polymer, and at least one non-polymeric acid having two or more pKa values or salt(s) thereof and/or at least one non-polymeric base having two or more pKb values or salt(s) thereof,
- 30 (v) at least one anionic polymer and at least one non-polymeric base having two or more pKb values or salt(s) thereof,
- (vi) at least one anionic polymer, at least one amphoteric polymer, and at least one non-polymeric acid having two or more pKa values or salt(s) thereof and/or at least one non-polymeric base having two or more pKb values or salt(s) thereof, or
- 35 (vii) at least one amphoteric polymer and at least one non-polymeric acid having two or more pKa values or salt(s) thereof and/or at least one non-polymeric base having two or more pKb values or salt(s) thereof.
- 40 3. The composition according to Claim 1 or 2, wherein the cationic polymer is selected from the group consisting of polyquaternium-4, polyquaternium-6, polyquaternium-7, polyquaternium-10, polyquaternium-24, polyquaternium-67, and a mixture thereof.
- 45 4. The composition according to any one of Claims 1 to 3, wherein the amount of the ionic polymer(s) in the composition is from 0.01% to 10% by weight, preferably from 0.05% to 5% by weight, and more preferably from 0.1% to 1% by weight, relative to the total weight of the composition.

5. The composition according to any one of Claims 1 to 4 wherein the non-polymeric acid having two or more pKa values or salt(s) thereof is selected from the group consisting of terephthalylidene dicamphor sulfonic acid and salts thereof (Mexoryl SX), Yellow 6 (Sunset Yellow FCF), ascorbic acid, phytic acid, citric acid, tartaric acid and salts thereof, and a mixture thereof.
6. The composition according to any one of Claims 1 to 5, wherein the amount of the crosslinker(s) in the composition is from 0.001% to 10% by weight, preferably from 0.005% to 5% by weight, and more preferably from 0.01% to 1% by weight, relative to the total weight of the composition.
7. The composition according to any one of Claims 1 to 6, wherein the (b) fatty alcohol has a structure R-OH wherein R is chosen from saturated and unsaturated, linear and branched radicals containing from 8 to 40 carbon atoms.
8. The composition according to any one of Claims 1 to 7, wherein the amount of the (b) fatty alcohol in the composition is from 0.1% to 15% by weight, preferably from 0.5% to 10% by weight, and more preferably from 1% to 5% by weight, relative to the total weight of the composition.
9. The composition according to any one of Claims 1 to 8, wherein the amount of the (c) water in the composition is from 50% to 99% by weight, preferably from 60% to 97% by weight, and more preferably from 70% to 95% by weight, relative to the total weight of the composition.
10. The composition according to any one of Claims 1 to 9, wherein the pH of the composition is from 3 to 9, preferably from 3.5 to 8.5, and more preferably from 4 to 8.
11. The composition according to any one of Claims 1 to 10, wherein the composition further comprises (d) at least one fatty material other than the (b) fatty alcohol, preferably at least one oil and/or at least one wax.
12. The composition according to any one of Claims 1 to 11, wherein the composition further comprises (e) at least one texture agent, preferably selected from hydrophilic nonionic polymeric thickeners, and more preferably selected from hydrophilic nonionic polymeric associative thickeners and hydrophilic nonionic polysaccharide thickeners.
13. A cosmetic process for keratin fibers such as hair, comprising applying to the keratin fibers the composition according to any one of Claims 1 to 12; and drying the composition to form a cosmetic film on the keratin fibers.
14. A process for preparing a film, preferably a cosmetic film, comprising: applying onto keratin fibers such as skin, the composition according to any one of Claims 1 to 12; and drying the composition.

15. A film, preferably a cosmetic film, comprising:
- (a) at least one polyion complex comprising  
at least one ionic polymer selected from the group consisting of cationic  
polymers, anionic polymers, amphoteric polymers, and mixtures thereof,  
and  
at least one crosslinker selected from the group consisting of non-polymeric  
acids having two or more pKa values or salt(s) thereof, non-polymeric bases  
having two or more pKb values or salt(s) thereof, and mixtures thereof; and
- (b) at least one fatty alcohol.

5

10

INTERNATIONAL SEARCH REPORT

International application No  
PCT/JP2021/003438

A. CLASSIFICATION OF SUBJECT MATTER  
 INV. A61K8/55 A61K8/73 A61K8/81 A61Q5/00  
 ADD.  
 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)  
 A61K A61Q  
 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)  
 EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2017/104221 A1 (OREAL [FR]; KASAI TAKEHIRO [JP] ET AL.) 22 June 2017 (2017-06-22) cited in the application the whole document	3-14
X	WO 2018/230673 A1 (OREAL [FR]; SHIROYA TOSHIFUMI [JP] ET AL.) 20 December 2018 (2018-12-20) cited in the application the whole document	3-14
X	WO 2018/230740 A1 (OREAL [FR]; SHIROYA TOSHIFUMI [JP] ET AL.) 20 December 2018 (2018-12-20) the whole document	3-14
	----- -/--	

Further documents are listed in the continuation of Box C.

See patent family annex.

\* Special categories of cited documents :

<p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier application or patent but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p>	<p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art</p> <p>"&amp;" document member of the same patent family</p>
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Date of the actual completion of the international search  12 May 2021	Date of mailing of the international search report  25/05/2021
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Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Authorized officer  Yon, Jean-Michel
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## INTERNATIONAL SEARCH REPORT

International application No  
PCT/JP2021/003438

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2019/116877 A1 (OREAL [FR]; SHIROYAMA TOSHIFUMI [JP] ET AL.) 20 June 2019 (2019-06-20) the whole document	3-14
X	----- WO 2017/104585 A1 (OREAL [FR]; KASAI TAKEHIKO [JP] ET AL.) 22 June 2017 (2017-06-22) the whole document -----	3-14

# INTERNATIONAL SEARCH REPORT

International application No.  
PCT/JP2021/003438

## Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1.  Claims Nos.:  
because they relate to subject matter not required to be searched by this Authority, namely:
  
2.  Claims Nos.: **1, 2, 15(completely); 3-14(partially)**  
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:  
**see FURTHER INFORMATION sheet PCT/ISA/210**
  
3.  Claims Nos.:  
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

## Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1.  As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
  
2.  As all searchable claims could be searched without effort justifying an additional fees, this Authority did not invite payment of additional fees.
  
3.  As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
  
4.  No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

### Remark on Protest

- The additional search fees were accompanied by the applicant's protest and, where applicable, the payment of a protest fee.
- The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.
- No protest accompanied the payment of additional search fees.

**FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210**

Continuation of Box II.2

Claims Nos.: 1, 2, 15(completely); 3-14(partially)

Present claim 1 relates to an extremely large number of possible compounds. Support and disclosure in the sense of Article 6 and 5 PCT is to be found however for only a very small proportion of the compounds claimed, see paragraphs [0323], [0324] et [0337] à [0339], as well as the examples).

The non-compliance with the substantive provisions is to such an extent, that the search was performed taking into consideration the non-compliance in determining the extent of the search of claim 1 (PCT Guidelines 9.19 and 9.23).

The search of claim 1 was restricted to those claimed compounds which appear to be supported and a generalisation of their structural formulae.

Consequently, compounds which have been searched are those of present claim 3 (definition of the cationic polymer) and present claim 5 (non-polymeric acid having two or more pKa values).

The applicant's attention is drawn to the fact that claims relating to inventions in respect of which no international search report has been established need not be the subject of an international preliminary examination (Rule 66.1(e) PCT). The applicant is advised that the EPO policy when acting as an International Preliminary Examining Authority is normally not to carry out a preliminary examination on matter which has not been searched. This is the case irrespective of whether or not the claims are amended following receipt of the search report or during any Chapter II procedure. If the application proceeds into the regional phase before the EPO, the applicant is reminded that a search may be carried out during examination before the EPO (see EPO Guidelines C-IV, 7.2), should the problems which led to the Article 17(2) PCT declaration be overcome.

## INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/JP2021/003438

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