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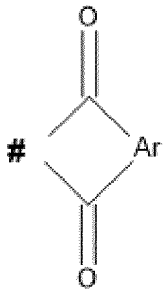
(54) USE OF POLYALKYLENEIMINE DERIVATIVES IN CLEANING COMPOSITIONS

(57) The use of a polymer having
a) a polyalkyleneimine backbone;
b) at least one aromatic moiety P1



(P1)

wherein
#
indicates the point of attachment to a nitrogen atom of the polyalkyleneimine backbone,
Ar
independently for each aromatic moiety P1 is 1,2-phenylene, 1,2-naphthylene, 2,3-naphthylene, or 1,8-naphthylene, wherein phenylene or naphthylene are unsubsituted or carry 1, 2, 3, 4, or 5 radicals which are selected from the group consisting of halogen, OH, C_1-C_4-alkyl, C_1-C_4-alkoxy, COOH, $CONH_2$, NO_2, NH_2, NH-CHO, NH- C_1-C_4-alkyl, and NH-(C=O)- C_1-C_4-alkyl;

c) at least one moiety P2,

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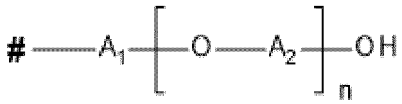


(P2)

wherein
#
indicates the point of attachment to a nitrogen atom of the polyalkyleneimine-backbone,
n
is an integer from 0 to 100,
A_1
is independently selected from linear or branched alkylenes having 2 to 18 carbon atoms,
A_2
for each individual repetition unit n A_2 is independently selected from linear or branched alkylenes having 2 to 18 carbon atoms,

in a cleaning composition.

Description

[0001] The present invention relates to the use of polyalkyleneimine derivatives in cleaning compositions and to cleaning compositions comprising polyalkyleneimine derivatives.

[0002] Removal of stains remains a challenge for all cleaning compositions. The removal of tough stains from both hard surfaces such as glass, stone, ceramic, porcelain, metal and wood and from fabrics and fabric surfaces such as textiles, fibers, filaments, and nonwovens needs to be continuously improved.

[0003] Of interest is for example the removal of bleachable stains by laundry detergents. Especially challenging is the removal of bleachable stains by use of liquid laundry detergents. A bleaching system commonly applied in powder laundry detergents is for example hydrogen peroxide, which is difficult to be formulated in liquid detergent systems because hydrogen peroxide is not stable in liquid system for prolonged periods of time as is required to ensure a shelf life of at least several months for the liquid detergent. Therefore, the performance of liquid detergents for removal of bleachable stains is until now limited and improved performance regarding removal of bleachable stains by laundry detergents is highly desirable.

[0004] Alkoxylated polyalkyleneimines are known as additives for stain removal in detergent formulations and are applied in liquid laundry detergents, but they only show limited performance with regard to removal of bleachable stains.

[0005] Alkoxylated polyalkyleneimine derivatives are so far only known as dispersants for pigments. WO2017/140538 discloses polyalkyleneimine based polyesters having polyester groups used as dispersant for pigments.

[0006] It was an object of the present invention to identify compounds in uses which comply with the above identifies objectives and needs. It was another objective of the present invention to identify compositions which comply with the above identifies objectives and needs.

[0007] This goal was achieved by the present invention as described herein below and as reflected in the claims.

[0008] Throughout this specification and the claims which follow, unless the context requires otherwise, the word "comprise", and variations such as "comprises" and "comprising", will be understood to imply the inclusion of a stated integer or step or group of integers or steps but not the exclusion of any other integer or step or group of integer or step. When used herein the term "comprising" can be substituted with the term "containing" or "including" or sometimes when used herein with the term "having".

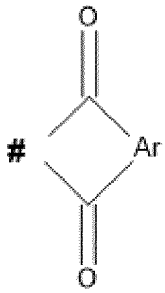
[0009] When used herein "consisting of" excludes any element, step, or ingredient not specified in the claim element. When used herein, "consisting essentially of" does not exclude materials or steps that do not materially affect the basic and novel characteristics of the claim.

[0010] In each instance herein any of the terms "comprising", "consisting essentially of" and "consisting of" may be replaced with either of the other two terms.

[0011] Generally, as used herein, the term "obtainable by" means that corresponding products do not necessarily have to be produced (i.e. obtained) by the corresponding method or process described in the respective specific context, but also products are comprised which exhibit all features of a product produced (obtained) by said corresponding method or process, wherein said products were actually not produced (obtained) by such method or process. However, the term "obtainable by" also comprises the more limiting term "obtained by", i.e. products which were actually produced (obtained) by a method or process described in the respective specific context.

[0012] The present invention relates to the use of a polymer having

a) a polyalkyleneimine backbone;
b) at least one aromatic moiety P1



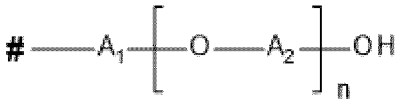
(P1)

wherein

# indicates the point of attachment to a nitrogen atom of the polyalkyleneimine backbone,

Ar independently for each aromatic moiety P1 is 1,2-phenylene, 1,2-naphthylene, 2,3-naphthylene, or 1,8-naphthylene, wherein phenylene or naphthylene are unsubsituted or carry 1, 2, 3, 4, or 5 radicals which are selected from the group consisting of halogen, OH, C_1-C_4-alkyl, C_1-C_4-alkoxy, COOH, $CONH_2$, NO_2, NH_2, NH-CHO, NH- C_1-C_4-alkyl, and NH-(C=O)- C_1-C_4-alkyl;

c) at least one moiety P2,



(P2)

wherein

# indicates the point of attachment to a nitrogen atom of the polyalkyleneimine backbone,

n is an integer from 0 to 100,

A_1 is independently selected from linear or branched alkylenes having 2 to 18 carbon atoms,

A_2 for each individual repetition unit n A_2 is independently selected from linear or branched alkylenes having 2 to 18 carbon atoms,

in a cleaning composition.

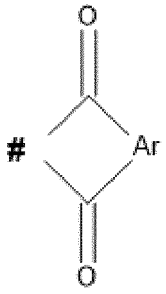
[0013] Additionally the present invention relates to a cleaning composition comprising

I. a surfactant and

II. a polymer having

a) a polyalkyleneimine backbone;

b) at least one aromatic moiety P1



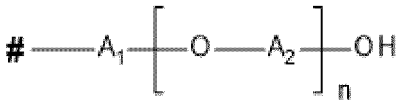
(P1)

wherein

# indicates the point of attachment to a nitrogen atom of the polyalkylene imine backbone,

Ar independently for each aromatic moiety P1 is 1,2-phenylene, 1,2-naphthylene, 2,3-naphthylene, or 1,8-naphthylene, wherein phenylene or naphthylene are unsubsituted or carry 1, 2, 3, 4, or 5 radicals which are selected from the group consisting of halogen, OH, C_1-C_4-alkyl, C_1-C_4-alkoxy, COOH, $CONH_2$, NO_2, NH_2, NH-CHO, NH- C_1-C_4-alkyl, and NH-(C=O)- C_1-C_4-alkyl;

c) at least one moiety P2,



(P2)

wherein

- # indicates the point of attachment to a nitrogen atom of the polyalkyleneimine backbone,
- n is an integer from 0 to 20,
- A_1 is independently selected from linear or branched alkylenes having 2 to 18 carbon atoms,
- A_2 for each individual repetition unit n A_2 is independently selected from linear or branched alkylenes having 2 to 18 carbon atoms.

[0014] In the following the various embodiments of the present invention are described in more detail.

[0015] The polymers of the present invention for use in cleaning compositions and for cleaning compositions have a polyalkylenimine backbone, to which the at least one aromatic moiety P1 and the at least one moiety P2 are attached.

[0016] The polyalkyleneimine backbone may be linear or branched. The polyalkyleneimine backbone is in particular a linear or branched poly(C_2-C_4-alkyleneimine), more particular a linear or branched polypropyleneimine, a linear or branched poly(ethyleneimine-co-propyleneimine) or a linear or branched polyethyleneimine or a mixture thereof. In one embodiment the polyalkyleneimine backbone is a linear or branched polyethyleneimine.

[0017] Before the modification with P1 and P2, the polyalkylenimine backbone has a weight average molecular weight (M_w) of from 100 to 10 000 g/mol. In one embodiment of the present invention the weight average molecular weight is in the range of from 250 to 5000 g/mol. In another embodiment the weight average molecular weight of the polyalkylenimine is in the range of from 600 to 2500 g/mol.

[0018] Both moieties P1 and P2 are attached to nitrogen atoms of the polyalkyleneimine backbone. It is apparent to a skilled person that polymers of the invention may have nitrogen atoms, which do not carry moieties P1 and P2. These nitrogen atoms are basic and susceptible to protonation or quaternization. Polymers of the invention, which have nitrogen atoms, which do not carry moieties P1 and P2, will usually have an amine number > 0 mg KOH/g, e.g. in the range from 2 to 150 mg KOH/g, in particular from 5 to 50 mg KOH/g. In one embodiment of the present invention the polyalkylene imine backbone in addition to the moieties P1 and P2 has protonated or quaternized amino groups. In this embodiment, the amount of protonated or quaternized amino groups is generally from 0.01 to 0.5 mol/kg.

[0019] In another embodiment the polyalkylene imine backbone in addition to the moieties P1 and P2 does not have quaternized amino groups. In this embodiment, the amount of quaternized amino groups is less than 0.01 mol/kg.

[0020] The polyalkyleneimines of the present invention have a molar ratio of from 0.01 to 0.5 aromatic moieties P1 per nitrogen atom in the polyalkyleneimine backbone. In another embodiment the molar ratio of aromatic moieties P1 to nitrogen atoms in the polyalkyleneimine backbone is in the range of from 0.02 to 0.4. In a further embodiment the molar ratio of aromatic moieties P1 to nitrogen atoms in the polyalkyleneimine backbone is in the range of from 0.1 to 0.3.

[0021] The aromatic moiety P1 is selected from the group consisting of 1,2-phenylene, 1,2-naphthylene, 2,3-naphthylene, and 1,8-naphthylene. The aromatic moiety P1 is bound to a nitrogen atom of the polyalkyleneimine backbone via two carbonyl groups, thereby forming a carboximide group. The point of attachment to the nitrogen atom of the polyalkyleneimine backbone is indicated by the symbol # in P1. The aryl or arylene moieties of P1 are unsubstituted or substituted, e.g. by 1, 2, 3, 4 or 5 radicals, which are preferably selected from halogen, OH, C1-C4-alkyl, C1-C4-alkoxy, COOH, CONH2, NH2, NO2, NH-CHO, NH-C1-C4-alkyl, and NH-(C=O)-C1-C4-alkyl. Within the polymers of the invention, the aromatic P1 moieties may be identical or different. Frequently, they are identical. In one embodiment, the Ar in the aromatic moiety P1 is 1,8-naphthylene. In a further embodiment, Ar in the aromatic moiety P1 is 1,8-naphthylene and the molar ratio of 1,8-naphthylene per nitrogen atom in the polyalkyleneimine backbone is in the range of from 0.01 to 0.3.

[0022] Each moiety P2 is attached to nitrogen atoms of the polyalkyleneimine backbone. The point of attachment to the nitrogen atom of the polyalkyleneimine backbone is indicated by the symbol # in P2. Each moiety P2 individually and independently from each other moiety P2 comprises a unit A1 which is independently selected from linear or branched alkylenes having 2 to 18 carbon atoms. Each moiety P2 individually and independently from each other moiety P2 comprises may comprise one or more units A2 which are independently selected from linear or branched alkylenes having 2 to 18 carbon atoms. In one embodiment of the present invention, A1 and A2 are independently for each repetition unit selected from the group consisting of ethylene, 1,2-propylene, 1,2-butylene, 2,3-butylene, i-butylene, pentenyl, hexyl, decenyl, dodecyl, tetradecenyl and hexadecenyl. In another embodiment A1 and A2 are independently for each repetition unit selected from the group consisting of ethylene, 1,2-propylene, and 1,2-butylene. In a further embodiment A1 and A2 are all ethylene.

In one embodiment the moiety P2 comprises at least two blocks of different alkylene groups, for example a first block formed by A1 and A2 may first be selected from ethylene, followed by a second block formed by A2 which may be selected from 1,2-propylene. In another embodiment the distribution of alkylene groups for individual units A1 and A2 is random.

[0023] In one embodiment, n is in the range of from 0 to 100. In another embodiment, n is in the range of from 1 to 50. In a further embodiment, n is in the range of from 10 to 35.

[0024] It is recognized that the polyalkyleneimines of the present disclosure may be asymmetrically alkoxylated, meaning that n for one individual moiety P2 is different from the n of another moiety P2. The number n is therefore the average number of substituents of all individual moieties P2.

[0025] In one embodiment, the molar ratio of from 0.1 to 0.99 moieties P2 per nitrogen atom in the polyalkyleneimine backbone. In another embodiment the molar ratio of moieties P2 to nitrogen atoms in the polyalkyleneimine backbone is in the range of from 0.5 to 0.98. In a further embodiment the molar ratio of moieties P2 to nitrogen atoms in the polyalkyleneimine backbone is in the range of from 0.7 to 0.9.

[0026] In one embodiment, Ar in the aromatic moiety P1 is 1,8-naphthylene, the molar ratio of 1,8-naphthylene per nitrogen atom in the polyalkyleneimine backbone is in the range of from 0.02 to 0.3, A1 and A2 in P2 are ethylene, n is in the range of from 10 to 35, and the molar ratio of P2 per nitrogen atom in the polyalkyleneimine backbone is in the range of from 0.7 to 0.98. The modified polyalkyleneimines can for example be obtained by the following process:

i. A polyalkyleneimine is reacted with at least one of the following dicarboxylic acids selected from the group consisting of substituted or unsubstituted 1,2-benzenedicarboxylic acid, substituted or unsubstituted 1,2-naphthalenedicarboxylic acid, substituted or unsubstituted 2,3-naphthalenedicarboxylic acid, and substituted or unsubstituted 1,8-naphthalenedicarboxylic acid.
ii. The product of step (ii) is reacted with at least one alkylene oxide using catalysts and reaction conditions well known in the art.

[0027] The polymer of the present invention is used in a cleaning composition. As used herein the phrase "cleaning composition" includes compositions and formulations de-signed for cleaning soiled material. Such cleaning compositions include but are not limited to laundry cleaning compositions and detergents, fabric softening compositions, fabric enhancing compositions, fabric freshening compositions, laundry prewash, laundry pretreat, laundry additives, spray products, dry cleaning agent or composition, laundry rinse additive, wash additive, post-rinse fabric treatment, ironing aid, dish washing compositions, hard surface cleaning compositions, unit dose formulation, delayed delivery formulation, detergent contained on or in a porous substrate or nonwoven sheet, and other suitable forms that may be apparent to one skilled in the art in view of the teachings herein. Such compositions may be used as a pre-laundering treatment, a post-laundering treatment, or may be added during the rinse or wash cycle of the laundering operation. The cleaning compositions may have a form selected from liquid, powder, single-phase or multi-phase unit dose, pouch, tablet, gel, paste, bar, or flake.

[0028] In one embodiment of the present invention the cleaning composition is a hard surface cleaning composition. In another embodiment the cleaning composition is a laundry detergent. In a further embodiment the cleaning composition is a liquid laundry detergent.

[0029] In one embodiment of the present invention the cleaning composition comprises in the range of from 0.01 to 5.0 wt-% based on the total weight of the cleaning composition of at least one polyalkyleneimine derivate according to the present invention. In a further embodiment the cleaning composition comprises in the range of from 0.1 to 1.0 wt-% based on the total weight of the cleaning composition of at least one polyalkyleneimine derivate according to the present invention. In another embodiment the amount of the polyalkyleneimine derivate according to the present invention in the cleaning composition is more than 0.01 wt.-% and less than 0.5 wt.-% based on the weight of the total cleaning composition.

[0030] The cleaning compositions comprise a surfactant system in an amount sufficient to provide desired cleaning properties. In some embodiments, the cleaning composition comprises, by weight of the composition, from about 1% to about 70% of a surfactant system. In other embodiments, the liquid cleaning composition comprises, by weight of the composition, from about 2% to about 60% of the surfactant system. In further embodiments, the cleaning composition comprises, by weight of the composition, from about 5% to about 30% of the surfactant system. The surfactant system may comprise a detersive surfactant selected from anionic surfactants, nonionic surfactants, cationic surfactants, zwitterionic surfactants, amphoteric surfactants, ampholytic surfactants, and mixtures thereof. Those of ordinary skill in the art will understand that a detersive surfactant encompasses any surfactant or mixture of surfactants that provide cleaning, stain removing, or laundering benefit to soiled material.

[0031] The cleaning compositions may also contain adjunct cleaning additives. Suitable adjunct cleaning additives include builders, structurants or thickeners, clay soil removal/anti-redeposition agents, polymeric soil release agents, polymeric dispersing agents, polymeric grease cleaning agents, enzymes, enzyme stabilizing systems, bleaching com-

pounds, bleaching agents, bleach activators, bleach catalysts, brighteners, dyes, hueing agents, dye transfer inhibiting agents, chelating agents, suds suppressors, softeners, and perfumes.

Methods

[0032] ^{1}H NMR measured in MeOD with Bruker Avance 400 MHz spectrometer.
[0033] pH is measured in 10 % aqueous solution.
[0034] Hydroxyl values are measured according to DIN 53240-1.
[0035] Weight average molecular weight was measured by GPC
[0036] Amine number was determined according to DIN 53176:2002-11.
[0037] The acid number was determined according to DIN 53402:1990-09.

Examples

[0038] The following liquid model composition was prepared:

Table 1. Single unit dose (SUD) detergent model formulation

Linear alkyl benzene sulfonic acid, salt of monoethnolamine	25
Alcohol ethoxylated (C13-C15 alcohol with 7 moles of EO)	25
1,2 propylene glycol	15
Glycerine	10
Citrate	1
Soap	8
Water	to 100

[0039] Compounds were added to a laundry liquor comprising a liquid model detergent (refer to Table 1) without polymer (additive dosage of 10% on weight of liquid model detergent (owod)) together with commercially obtained stained fabrics from Center of Test Materials CFT C-S-15 (Blueberry Juice) or from Swissatest Testmaterialien AG Empa 114 (Red Wine), respectively. Washing conditions were 1 g/L detergent, liquor 250 mL, 60 min, 40°C, 6-fold determination. After wash the fabrics were rinsed for 5 min at 30°C with 250 mL water, centrifuged and ironed. The fabrics were instrumentally assessed with a Datacolor reflection spectrometer Model Type SF600 before and after wash. From the reflection data readings ΔY were derived and further expressed in $\Delta\Delta Y$ values vs reference without polymer (baseline correction for pure bleach effect). The higher values of $\Delta\Delta Y$ are observed the more bleaching effect is found, respectively.
[0040] For reference, test results with the current market benchmark #C3 are given. Furthermore, reference values with the plain polyethyleneimine are shown for comparison as well as data with the respective alkoxylated backbone without 1,8-naphthalic anhydride functionalization.

Table 2. Results of primary detergency in SUD model formulation on commercially available stains CS15 and Empa 114 (6-fold determination each; abbreviations are explained in the paragraph below).

#	PEI backbone (g/mol)	molar ratio NA/NH	molar ratio EO/NH	$\Delta\Delta Y$ (CS15)	$\Delta\Delta Y$ (Empa 114)
C1	PEI800	0	0	-10,3	-9,6
C2	PEI2000	0	0	-13,6	-9,7
C3	PEI800	0	20	3,2	2,8
C4	PEI2000	0	15	2,5	2,6
C5	PEI2000	0	30	2,3	2,8
C6	PEI2000	0.21	0	-1,4	-0,1
C7	PEI800	0.21	0	-5,8	-3,4
C8	PEI800	0.10	0	-7,6	-5,3
1	PEI800	0.10	23	1,2	3,8
2	PEI800	0.10	35	3,0	4,1

(continued)

#	PEI backbone (g/mol)	molar ratio NA/NH	molar ratio EO/NH	ΔΔY (CS15)	ΔΔY (Empa 114)
3	PEI800	0.21	24	6,2	6,7
4	PEI2000	0.02	12	4,2	4,6
5	PEI2000	0.02	23	6,6	6,9

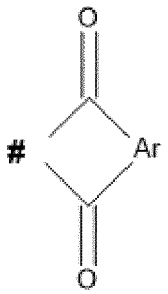
[0041] Molar ratio was calculated as follows (example for compound #3 in Table 2):

- 1 mole PEI 800 consisting of 14 repeating units, contains statistically 4.6 primary, 4.6 secondary, and 4.6 tertiary amine functionalities
- This 1 mole PEI 800 is reacted with 3 mole naphthylanhydride, leaving on average 1.6 primary and 4.6 secondary amine functionalities for later alkoxylation. Ratio NA/NH is thus calculated 3/14=0.21 (NA = 1,8 naphthalic anhydride; NH = amine function)
- Reacting with further 188 mole EO leads to EO/NH ratio of 24.
- All other compounds in the table have been prepared in a similar manner.

Claims

1. The use of a polymer having

a) a polyalkyleneimine backbone;
b) at least one aromatic moiety P1

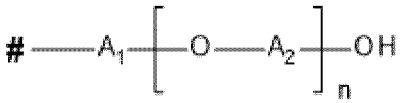


(P1)

wherein

# indicates the point of attachment to a nitrogen atom of the polyalkyleneimine backbone,
Ar independently for each aromatic moiety P1 is 1,2-phenylene, 1,2-naphthylene, 2,3-naphthylene, or 1,8-naphthylene, wherein phenylene or naphthylene are unsubsituted or carry 1, 2, 3, 4, or 5 radicals which are selected from the group consisting of halogen, OH, C_1-C_4-alkyl, C_1-C_4-alkoxy, COOH, $CONH_2$, NO_2, NH_2, NH-CHO, NH- C_1-C_4-alkyl, and NH-(C=O)- C_1-C_4-alkyl;

c) at least one moiety P2,



(P2)

wherein

# indicates the point of attachment to a nitrogen atom of the polyalkyleneiminebackbone,
n is an integer from 0 to 100,
A_1 is independently selected from linear or branched alkylenes having 2 to 18 carbon atoms,
A_2 for each individual repetition unit n A_2 is independently selected from linear or branched alkylenes having 2 to 18 carbon atoms,

in a cleaning composition.

2. The use of a polymer according to any of the previous claims wherein the ratio of moles of aromatic moieties P_1 to moles of nitrogen in the polyalkyleneimine backbone is in the range of from 0.01 to 0.5 and the ratio of moles of aliphatic moieties P_2 to moles of nitrogen in the polyalkyleneimine backbone is in the range of from 0.5 to 0.99.

3. The use of a polymer according to any of the previous claims wherein the polyalkyleneimine backbone is a linear or branched polyethyleneimine backbone.

4. The use of a polymer according to claim 4 wherein the polyethyleneimine backbone has a weight average molecular weight in the range of from 600 to 2500 Dalton.

5. The use of a polymer according to any of the previous claims wherein Ar is 1,8-naphthylene.

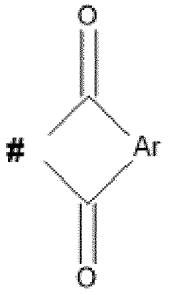
6. The use of a polymer according to any of the previous claims wherein n is of from 10 to 35.

7. The use of a polymer according to any of the previous claims wherein A_1 and/or A_2 are selected from the group consisting of ethylene, 1,2-propylene, and 1,2-butylene.

8. A cleaning composition comprising

I. a surfactant and
II. a polymer having

a) a polyalkyleneimine backbone;
b) at least one aromatic moiety P1



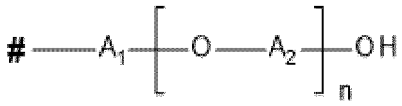
(P1)

wherein

# indicates the point of attachment to a nitrogen atom of the polyalkylene iminebackbone,

Ar independently for each aromatic moiety P1 is 1,2-phenylene, 1,2-naphthylene, 2,3-naphthylene, or 1,8-naphthylene, wherein phenylene or naphthylene are unsubsituted or carry 1, 2, 3, 4, or 5 radicals which are selected from the group consisting of halogen, OH, C_1-C_4-alkyl, C_1-C_4-alkoxy, COOH, $CONH_2$, NO_2, NH_2, NH-CHO, NH- C_1-C_4-alkyl, and NH-(C=O)- C_1-C_4-alkyl;

c) at least one moiety P2,



(P2)

wherein

# indicates the point of attachment to a nitrogen atom of the polyalkyleneiminebackbone,
n is an integer from 0 to 20,
A_1 is independently selected from linear or branched alkylenes having 2 to 18 carbon atoms,
A_2 for each individual repetition unit n A_2 is independently selected from linear or branched alkylenes having 2 to 18 carbon atoms.

9. The cleaning composition according to claim 9 wherein the ratio of moles of aromatic moieties P_1 to moles of nitrogen in the polyalkyleneimine backbone is in the range of from 0.01 to 0.5 and the ratio of moles of aliphatic moieties P_2 to moles of nitrogen in the polyalkyleneimine backbone is in the range of from 0.5 to 0.99.

10. The cleaning composition according to any one of claims 8 or 9, wherein the polyalkyleneimine backbone is a linear or branched polyethyleneimine backbone.

11. The cleaning composition according to claim 10 wherein the polyethyleneimine backbone has a weight average molecular weight in the range of from 600 to 2500 Dalton.

12. The cleaning composition according to any of claims 8 to 11 wherein Ar is 1,8-naphthylene.

13. The cleaning composition according to any of claims 8 to 12 wherein n is of from 10 to 35.

14. The cleaning composition according to any of claims 8 to 13 wherein A_1 and/or A_2 are selected from the group consisting of ethylene, 1,2-propylene, and 1,2-butylene.

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EUROPEAN SEARCH REPORT

Application Number
EP 20 15 7881

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The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
Munich	22 June 2020	Péntek, Eric

CATEGORY OF CITED DOCUMENTS

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