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⑤④ **Hydrophilic oxygen permeable polymers.**

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⑥④ Designated Contracting States :
AT DE ES FR GB

⑤⑥ References cited :
**US-A- 3 004 950
US-A- 4 576 973**

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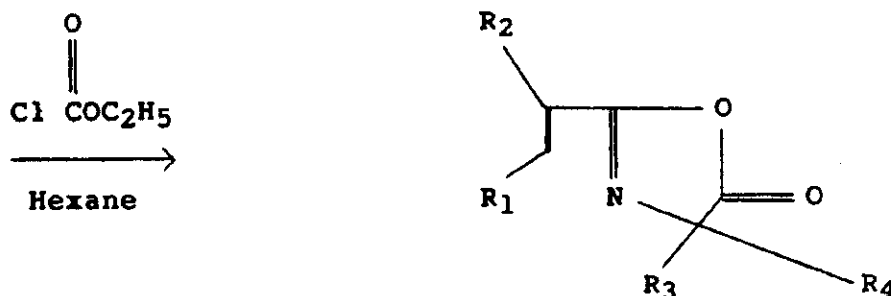
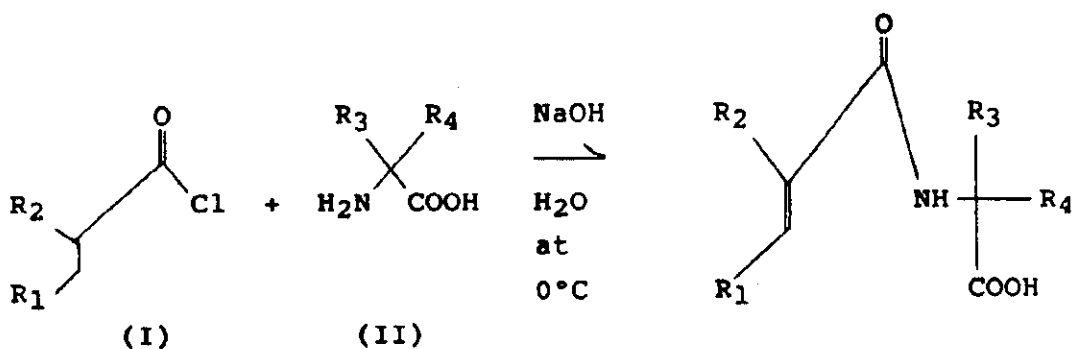
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and R³ and R⁴ independently denote alkyl radicals with one to six carbon atoms or R³ and R⁴ taken together represent cyclohexyl.

Specific examples of the useful internal wetting agents include 2-isopropenyl-4,4-dimethyl-2-oxazolin-5-one (IPDMO); 2-vinyl-4,4-dimethyl-2-oxazolin-5-one (VDMO); spiro-4'-(2' isopropenyl-2'-oxazolin-5-one) cyclohexane (IPCO); spiro-4'-(2'-Vinyl-2'-oxazolin-5'-one) cyclohexane (VCO), and 2-(1-propenyl)-4,4-dimethyl-2-oxazolin-5-one (PDMO).

These compounds have two important features which make them particularly preferred comonomers with acrylate or methacrylate functional polysiloxane monomers: (1) They are relatively nonpolar and are thus highly compatible with polysiloxane monomers thus forming optically clear monomer mixtures and polymers, and (2) They are converted to highly polar amino acids upon mild hydrolysis. Thus they can be copolymerized into a hydrophobic polymeric matrix and then hydrolyzed to render the matrix substantially hydrophilic.

The internal wetting agents can be prepared by the general reaction sequence:



40 The first step is a Shotten-Bauman acylation of an amino acid. The polymerizable functionality is introduced by using either acryloyl or methacryloyl chloride. The second step involves a ring closure with a chloroformate to yield the desired oxazolinone. The product is isolated and purified by the usual procedures of organic chemistry.

45 The internal wetting agents can be copolymerized with acrylate- or methacrylate- functional endcapped siloxane monomers to form polymeric materials which have high oxygen permeabilities. These polymeric materials may then be hydrolyzed in order to convert the oxazolinone components into amino acids. In general, the hydrolysis step will follow the general reaction of:

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wherein

E is either hydrogen or methyl

D is branched or normal alkyl having 3 to 6 carbon atoms, preferably 3 to 4 carbon atoms

Z is either hydrogen or methyl and

n is an integer from 3 to 8 and preferably from 4 to 6.

Illustrative of the foregoing cycloalkyl toughening agents are the following: Menthyl methacrylate, menthyl acrylate, tertiarybutylcyclohexyl methacrylate, isopropylcyclopentyl acrylate, tertiarypentylcycloheptyl methacrylate, tertiarybutylcyclohexyl acrylate, isohexylcyclopentyl acrylate and methylisopentyl cyclooctyl acrylate.

Useful polycyclic toughening agents are described and defined in US-A- 4,355,147. These agents are a polycyclic acrylate or methacrylate selected from the group consisting of isobornyl acrylate, isobornyl methacrylate, dicyclopentadienyl acrylate, dicyclopentadienyl methacrylate, adamantyl acrylate, adamantyl methacrylate isopinocampyl acrylate and isopinocampyl methacrylate.

Illustrative of useful fluorinated methacrylate toughening agents are: octafluoropentylmethacrylate, trifluoromethylmethacrylate, and pentafluoroethyl methacrylate.

The toughening agent suitably may be present in an amount from 90 to 10 parts by weight per 10 to 90 parts by weight of the above-described monomers. More preferably the modifier is present in the amount of 70 to 10 parts, more preferably yet the modifier is 45 to 15 parts.

The polymeric materials of the present invention are formed by: (1) mixing the monomers together optionally with a toughening agent as described above; (2) adding a polymerization initiator; (3) subjecting the monomer/initiator mixture to a source of ultraviolet or actinic radiation and curing said mixture to a solid or elastomeric state.

Typical polymerization initiators include free radical generating polymerization initiators of the type illustrated by acetyl peroxide, lauroyl peroxide, decanoyl peroxide, copryl peroxide, benzoyl peroxide, tertiary butyl peroxyvalate, diisopropyl peroxy carbonate, tertiary butyl peroctoate, and α,α -azobis-isobutyronitrile. Ultraviolet free radical initiators illustrated by diethoxyacetophenone can also be used. The curing process will of course depend upon the initiator used and the physical characteristics of the comonomer mixture such as viscosity. In any event, the level of initiator employed generally will vary within the range of 0.01 to 2 weight percent of the mixture of monomers.

Typical formulations will comprise 1 to 98 weight percent siloxane monomer, 1 to 50 weight percent of the defined internal agents, and 1 to 80 weight percent toughening agent. Preferably, the composition will comprise 60 to 95 weight percent siloxane monomer, 4 to 20 weight percent toughening agent and 1 to 20 weight percent of the novel wetting agent.

EXAMPLES

A. Synthesis of 2-isopropenyl-4,4-dimethyl-2-oxazolin-5-one (IPDMO)

1. Methacryloylation: In a 500 ml 3-neck round bottom flask equipped with a mechanical stirrer, thermometer and an addition funnel, 51.5 grams (0.5 mole) of α -aminoisobutyric acid (Aldrich) and 40 grams (1 mole) of NaOH were dissolved in ~ 150 ml of H₂O. The reaction flask was cooled to 0~-5°C with MeOH-ice bath and 0.5 mole of methacryloyl chloride (distilled, Aldrich) was added dropwise while the temperature of the reaction mixture kept below 0°C. After stirring for an additional hour, the reaction mixture was acidified with concentrated HCl to a pH of about 3 to precipitate out the intermediate, which was then filtered, washed with cold H₂O and air dried. Further drying was accomplished in vacuum oven at 80°C overnight to obtain 44.9 grams (0.26 mole; 53%) N-methacryloyl- α -aminoisobutyric acid (MAIBA), which was suitable for IPDMO synthesis. m.p. 157-159°C.

2. Cyclization; Synthesis of IPDMO: In a 500 ml dry 3-neck round bottom flask, 0.26 mole of MAIBA was dispersed in 300 ml of dry hexane and allowed to react with 0.26 mole of alkylchloroformate with mechanical stirring by dropwise addition of triethylamine (0.52 mole) while the temperature of the reaction mixture was maintained at 45 ~ 50°C. During the addition, the copious evolution of carbon dioxide and the formation of white precipitate of TEA-HCl were observed. The reaction mixture was stirred for an additional two hours. After cooling to room temperature, white precipitate was filtered off and hexane evaporated off yielding an oil. Pure IPDMO was obtained after two times of recrystallization in hexane at dry ice-acetone temperature. Yield was 29 grams (73%).

B. Synthesis of 2-vinyl-4,4-dimethyl-2-oxazolin-5-one (VDMO)

1. Acrylation was achieved by adopting a procedure similar to step 1 of the synthesis of IPDMO except acryloyl chloride was used in place of methacryloyl chloride. The intermediate N-acryloyl- α -aminobutyric acid (AAIBA) was produced.

2. AAIBA was reacted with triethylamine and ethyl chloroformate in a fashion similar to step 2 of the synthesis of IPDMO to produce VDMO.

C. Synthesis of Spiro-4'-(2'-isopropenyl-2-oxazolin-5'-one) Cyclohexane (IPCO)

The synthetic method used to synthesize IPDMO was followed except 1-amino-1-cyclohexane carboxylic acid was used in place of α -aminoisobutyric acid.

D. Synthesis of Spiro-4'-(2'-vinyl-2'-oxazolin-5'-one) Cyclohexane (VCO)

The synthetic method employed in the synthesis of VDMO was used except 1-amino-1-cyclohexane carboxylic acid was used in place of α -aminoisobutyric acid.

E. Synthesis of 2-(1-propenyl)-4,4-dimethyl-2-oxazolin-5-one (PDMO)

The method employed in the synthesis of VDMO was employed except crotonyl chloride was used in place of methacryloyl chloride and α -aminoisobutyric acid was used.

F. Synthesis and Characterization of Polymeric Materials**1. Polydimethylsiloxane Based Materials**

Methacrylate end capped polydimethylsiloxane (PDMS) with an average degree of polymerization of about 180 was compounded with isobornylmethacrylate (IBOMA) and VDMO in the weight proportion of 85/5/5 (PDMS/IBOMA/VDMO). The mixture was stirred, a free radical initiator was added and polymerized by exposure to ultraviolet radiation. The resultant polymeric material was characterized for modulus, tensile strength, elongation, oleic acid uptake, and oxygen permeability (D_K). The material was purified by extraction with toluene and also the internal wetting agent (VDMO) was hydrolyzed in the polymeric material by boiling in buffered saline solution for 20 minutes. Both the toluene extracted sample and toluene extracted/hydrolyzed sample were characterized. The results of these tests are reported in Table 1.

TABLE 1**Polydimethylsiloxane Based Material**

<u>Material</u>	<u>Youngs Modulus MPa (g/mm²)</u>	<u>Tensile Strength MPa (g/mm²)</u>	<u>Elongation %</u>	<u>Oleic %</u>	<u>D_K</u>
95/5/5 (PDMS /IBOMA/VDMO)	1.049 (107)	0.922 (94)	127	4.7	200
toluene extracted	5.108 (521)	1.392 (142)	35	4.4	200
extracted/ hydrolyzed	8.902 (908)	1.951 (199)	37	6.0	200

Comparison with Prior Art Polymeric Materials

The polymeric material from Example F was immersed in a buffered solution for a period of time under

controlled conditions. As a control polymeric material combining the wetting agents described by US-A-4,652,622 was subjected to the same conditions and the wettability of each sample was monitored. The polymeric material of the present invention maintains its wettability to a greater degree than the state of the art wetting agent (the control).

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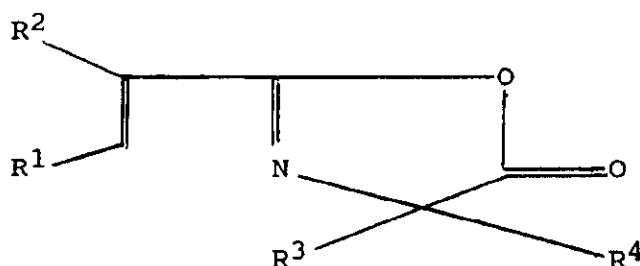
Claims

Claims for the following Contracting States : AT, DE, FR, GB

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1. A composition formed by polymerizing a mixture comprising:
 - (a) an acrylate- or methacrylate-functional endcapped siloxane monomer; and
 - (b) an internal wetting agent represented by the general formula

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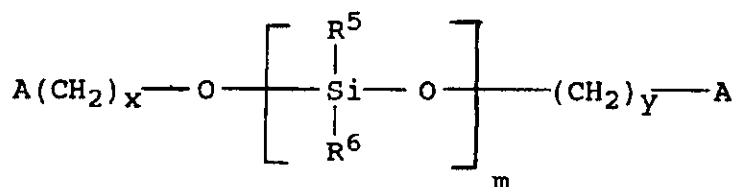
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where R¹ and R² independently denote a hydrogen atom or a lower alkyl radical with one to six carbon atoms, and R³ and R⁴ independently denote alkyl radicals with one to six carbon atoms or R³ and R⁴ taken together represent a cyclohexyl radical.

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2. A composition according to Claim 1, wherein the siloxane monomer is represented by the formula:

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wherein A denotes an acrylate or methacrylate radical, R⁵ and R⁶ independently denote alkyl radicals with one to six carbon atoms or phenyl radicals, x and y are on the average one to six, and m is at least one.

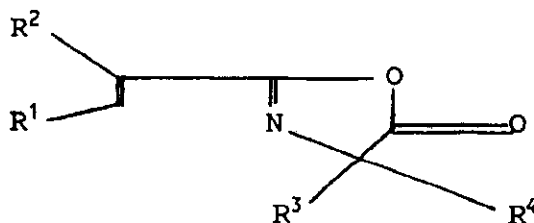
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3. A composition according to Claim 1 or Claim 2, comprising 1-98 weight percent of the siloxane monomer (a), 1-50 weight percent of said internal wetting agent (b) and from 1 to 80 weight percent of a toughening agent.
4. A composition according to Claim 3, which comprises: 60-95 weight percent of the siloxane monomer (a); 1-20 weight percent of said internal wetting agent (b); and 4-20 weight percent of said toughening agent.
5. A composition according to any preceding claim, wherein said internal wetting agent is 2-vinyl-4,4-dimethyl-2-oxazolin-5-one.
6. A composition according to any one of Claims 1-4, wherein said internal wetting agent is 2-isopropenyl-4,4-dimethyl-2-oxazolin-5-one.
7. A contact lens made from a composition according to any preceding claim.

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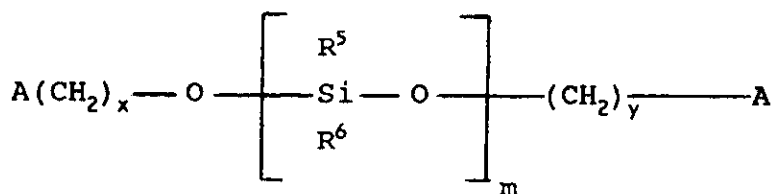
Claims for the following Contracting State : ES

1. A process for preparing a composition, formed by polymerizing a mixture comprising:
 (a) an acrylate- or methacrylate-functional endcapped siloxane monomer; and
 (b) an internal wetting agent represented by the general formula



where R¹ and R² independently denote a hydrogen atom or a lower alkyl radical with one to six carbon atoms, and R³ and R⁴ independently denote alkyl radicals with one to six carbon atoms or R³ and R⁴ taken together represent a cyclohexyl radical.

2. A process according to Claim 1, wherein the siloxane monomer is represented by the formula:



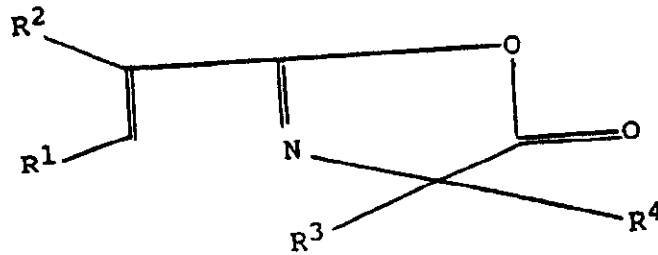
wherein A denotes an acrylate or methacrylate radical, R⁵ and R⁶ independently denote alkyl radicals with one to six carbon atoms or phenyl radicals, x and y are on the average one to six, and m is at least one.

3. A process according to Claim 1 or Claim 2, wherein said mixture comprises 1-98 weight percent of the siloxane monomer (a), 1-50 weight percent of said internal wetting agent (b) and from 1 to 80 weight percent of a toughening agent.
4. A process according to Claim 3, wherein said mixture comprises: 60-95 weight percent of the siloxane monomer (a); 1-20 weight percent of said internal wetting agent (b); and 4-20 weight percent of said toughening agent.
5. A process according to any preceding claim, wherein said internal wetting agent is 2-vinyl-4,4-dimethyl-2-oxazolin-5-one.
6. A process according to any one of Claims 1-4, wherein said internal wetting agent is 2-isopropenyl-4,4-dimethyl-2-oxazolin-5-one.
7. A contact lens made from a composition prepared by a process according to any preceding claim.

Patentansprüche

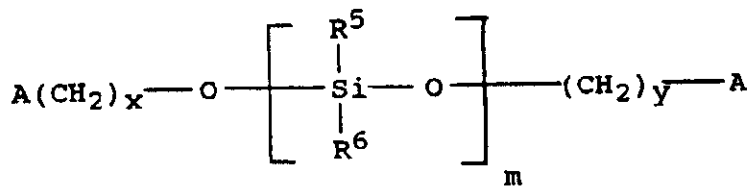
Patentansprüche für folgende Vertragsstaaten : AT, DE, FR, GB

1. Zusammensetzung, gebildet durch das Polymerisieren einer Mischung, die umfaßt:
 (a) ein mit funktioneller Acrylat- oder Methacrylatgruppe endverkapptes Siloxanmonomer; und
 (b) ein inneres Benetzungsmittel der allgemeinen Formel:



worin R¹ und R² unabhängig voneinander für ein Wasserstoffatom oder einen Niederalkylrest mit 1 bis 6 C-Atomen stehen und R³ und R⁴ unabhängig voneinander für Alkylreste mit 1 bis 6 C-Atomen stehen oder R³ und R⁴ zusammengenommen einen Zylohexylrest darstellen.

2. Zusammensetzung nach Anspruch 1, worin das Siloxanmonomer von der Formel

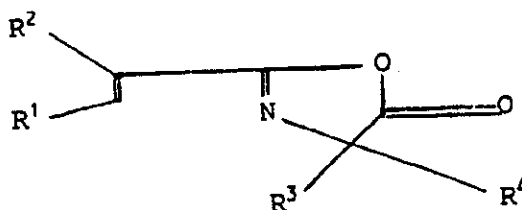


dargestellt wird, worin A einen Acrylat- oder Methacrylatrest bezeichnet, R⁵ und R⁶ unabhängig voneinander Alkylreste mit 1 bis 6 C-Atomen oder Phenylreste bezeichnen, x und y durchschnittlich 1 bis 6 sind und m zumindest 1 ist.

3. Zusammensetzung nach Anspruch 1 oder 2, die 1 bis 98 Gew.-% des Siloxanmonomers (a), 1 bis 50 Gew.-% des genannten inneren Benetzungsmittels (b) und von 1 bis 80 Gew.-% eines festigkeitserhöhenden Mittels umfaßt.
4. Zusammensetzung nach Anspruch 3, die umfaßt: 60 bis 95 Gew.-% des Siloxanmonomers (a); 1 bis 20 Gew.-% des genannten inneren Benetzungsmittels (b); und 4 bis 20 Gew.-% des genannten festigkeitserhöhenden Mittels.
5. Zusammensetzung nach einem der vorhergehenden Ansprüche, worin das genannte innere Benetzungsmittel 2-Vinyl-4,4-dimethyl-2-oxazolin-5-on ist.
6. Zusammensetzung nach einem der Ansprüche 1 bis 4, worin das genannte innere Benetzungsmittel 2-Isopropenyl-4,4-dimethyl-2-oxazolin-5-on ist.
7. Kontaktlinse, die aus einer Zusammensetzung nach einem der vorhergehenden Ansprüche hergestellt ist.

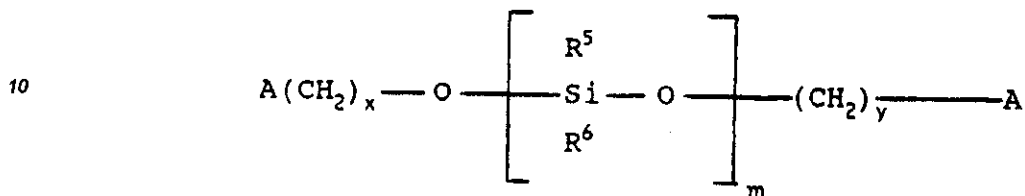
Patentansprüche für folgenden Vertragsstaat : ES

1. Verfahren zur Herstellung einer Zusammensetzung, umfassend das Polymerisieren einer Mischung, die umfaßt:
- (a) ein mit funktioneller Acrylat- oder Methacrylatgruppe endverkapptes Siloxanmonomer; und
- (b) ein inneres Benetzungsmittel allgemeinen Formel:



worin R¹ und R² unabhängig voneinander für ein Wasserstoffatom oder einen Niederalkylrest mit 1 bis 6 C-Atomen stehen und R³ und R⁴ unabhängig voneinander für Alkylreste mit 1 bis 6 C-Atomen stehen oder R³ und R⁴ zusammengenommen einen Zyklohexylrest darstellen.

- 5 2. Verfahren nach Anspruch 1, worin das Siloxanmonomer von der Formel



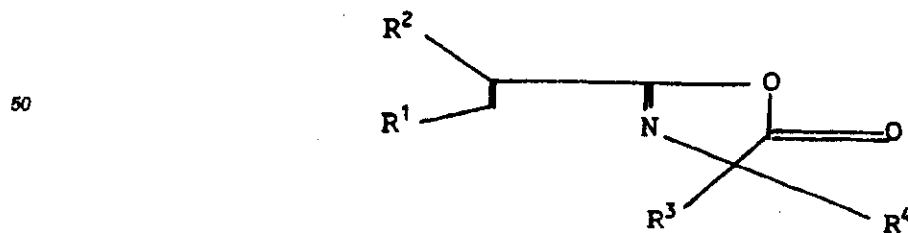
dargestellt wird, worin A einen Acrylat- oder Methacrylatrest bezeichnet, R⁵ und R⁶ unabhängig voneinander Alkylreste mit 1 bis 6 C-Atomen oder Phenylreste bezeichnen, x und y durchschnittlich 1 bis 6 sind und m zumindest 1 ist.

- 20 3. Verfahren nach Anspruch 1 oder 2, worin die genannte Mischung 1 bis 98 Gew.-% des Siloxanmonomers (a), 1 bis 50 Gew.-% des genannten inneren Benetzungsmittels (b) und von 1 bis 80 Gew.-% eines festigkeitserhöhenden Mittels umfaßt.
- 25 4. Verfahren nach Anspruch 3, worin die genannte Mischung umfaßt: 60 bis 95 Gew.-% des Siloxanmonomers (a); 1 bis 20 Gew.-% des genannten inneren Benetzungsmittels (b); und 4 bis 20 Gew.-% des genannten festigkeitserhöhenden Mittels.
- 30 5. Verfahren nach einem der vorhergehenden Ansprüche, worin das genannte innere Benetzungsmittel 2-Vinyl-4,4-dimethyl-2-oxazolin-5-on ist.
- 35 6. Verfahren nach einem der Ansprüche 1 bis 4, worin das genannte innere Benetzungsmittel 2-Isopropenyl-4,4-dimethyl-2-oxazolin-5-on ist.
7. Kontaktlinse, die aus einer Zusammensetzung hergestellt ist, die nach einem Verfahren nach einem der vorhergehenden Ansprüche hergestellt ist.

Revendications

40 **Revendications pour les Etats contractants suivants : AT, DE, FR, GB**

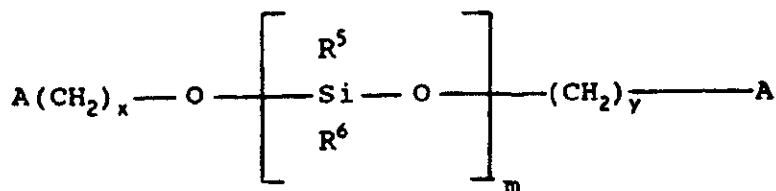
1. Composition formée par polymérisation d'un mélange comprenant:
- a) un monomère de siloxane comprenant des groupes terminaux à fonction acrylate ou méthacrylate;
- et
- 45 b) un agent mouillant interne représenté par la formule générale



dans laquelle R¹ et R² représentent indépendamment un atome d'hydrogène ou un radical alkyle inférieur avec de 1 à 6 atomes de carbone, et R³ et R⁴ représentent indépendamment des radicaux alkyle

avec de 1 à 6 atomes de carbone ou R³ et R⁴ pris ensemble représentent un radical cyclohexyle.

2. Une composition selon la revendication 1, dans laquelle le monomère de siloxane est représenté par la formule:

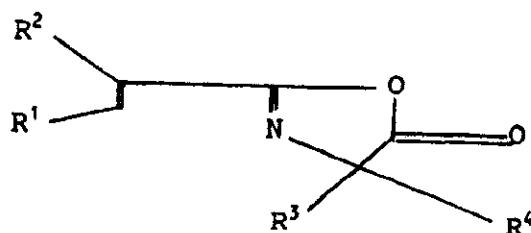


dans laquelle A représente un acrylate ou un méthacrylate, R⁵ et R⁶ représentent indépendamment des radicaux alkyle avec de 1 à 6 atomes de carbone ou des radicaux phényles, x et y sont sur la moyenne de 1 à 6, et m vaut au moins 1.

3. Composition selon la revendication 1 ou 2, comprenant de 1 à 98 % en poids de monomère siloxane (a), 1 à 50 % en poids de l'agent mouillant interne, et de 1 à 80 % en poids d'un durcisseur.
4. Composition selon la revendication 3, qui comporte de 60 à 95 % en poids du monomère de siloxane (a), de 1 à 20 % en poids de l'agent mouillant interne (b) et de 4 à 20 % en poids du durcisseur.
5. Composition selon l'une quelconque des revendications précédentes, dans laquelle l'agent mouillant interne est le 2-vinyl-4,4-diméthyl-2-oxazoline-5-one.
6. Composition selon l'une quelconque des revendications 1 à 4, dans laquelle l'agent mouillant interne est le 2-isopropényl-4,4-diméthyl-2-oxazoline-5-one.
7. Lentille de contact fabriquée à partir d'une composition selon l'une des revendications précédentes.

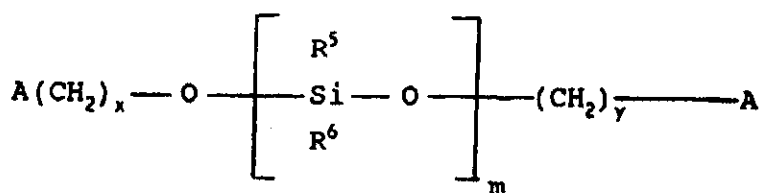
Revendications pour l'Etat contractant suivant : ES

1. Procédé de préparation d'une composition comportant la polymérisation d'un mélange qui comprend:
- un monomère de siloxane comprenant des groupes terminaux à fonction acrylate ou méthacrylate;
 - et
 - un agent mouillant interne représenté par la formule générale



dans laquelle R¹ et R² représentent indépendamment un atome d'hydrogène ou un radical alkyle inférieur avec de 1 à 6 atomes de carbone, et R³ et R⁴ représentent indépendamment des radicaux alkyle avec de 1 à 6 atomes de carbone ou R³ et R⁴ pris ensemble représentent un radical cyclohexyle.

2. Procédé selon la revendication 1, dans lequel le monomère de siloxane est représenté par la formule:



dans laquelle A représente un acrylate ou un méthacrylate, R⁵ et R⁶ représentent indépendamment des radicaux alkyle avec de 1 à 6 atomes de carbone ou des radicaux phényles, x et y sont sur la moyenne de 1 à 6, et m vaut au moins 1.

3. Procédé selon la revendication 1 ou 2, dans lequel ledit mélange comprend de 1 à 98 % en poids de monomère siloxane (a), 1 à 50 % en poids de l'agent mouillant interne, et de 1 à 80 % en poids d'un durcisseur.
4. Procédé selon la revendication 3, dans lequel ledit mélange comporte de 60 à 95 % en poids du monomère de siloxane (a), de 1 à 20 % en poids de l'agent mouillant interne (b) et de 4 à 20 % en poids du durcisseur.
5. Procédé selon l'une quelconque des revendications précédentes, dans lequel l'agent mouillant interne est le 2-vinyl-4,4-diméthyl-2-oxazoline-5-one.
6. Procédé selon l'une quelconque des revendications 1 à 4, dans lequel l'agent mouillant interne est le 2-isopropényl-4,4-diméthyl-2-oxazoline-5-one.
7. Lentille de contact fabriquée à partir d'une composition préparée au moyen du procédé selon l'une des revendications précédentes.

REGISTER ENTRY FOR EP0328340

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Priority claimed:

09.02.1988 in United States of America - doc: 153901

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Title HYDROPHILIC OXYGEN PERMEABLE POLYMERS.

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Classified to

C08F G02B

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HYDROPHILIC OXYGEN PERMEABLE POLYMERS.

13.11.1989 EPO: Search report published on 13.12.1989

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NY 14602, United States of America

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