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(54) Title: ALKYLSILANE CONTACT LENS AND POLYMER (57) Abstract Alkylsilane polymers and optical contact lenses fabricated therefrom.		

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ALKYLSILANE CONTACT LENS AND POLYMER
Cross-Reference to Related Application

This is a continuation-in-part of my
copending application Serial No. 641,594, filed August
5 17, 1984, now to be abandoned.

Field of the Invention

This invention relates to optical contact
lenses and materials therefor and, in particular, to
alkylsilane polymers and alkylsilane polymer contact
10 lenses.

Background of the Invention

Many polymeric materials have been evaluated
for potential utility as contact lens material, but a
very limited number of materials have been found to form
15 contact lenses which are satisfactory. Advances in
contact lens materials and techniques have come in small
steps, which have been excruciatingly slow and
difficult. Polymers and methods which appeared
attractive have fallen by the wayside. The problems are
20 myriad and predictability is low. It is difficult and
frequently impossible to predict optical quality,
strength and flexibility, resistance to protein
build-up, machining and fabrication characteristics,
dimensional stability, oxygen permeability, and general
25 biological compatability. It is impossible to predict,
or even to speculate as to possible optical, oxygen
permeability, and biological characteristics of
structural and industrial silanes such as disclosed by
Campbell, U.S. Patent No. 2,958,681 for example.

30 Reference is made to the literature, in
texts, treatises and technical literature which describe
silicon compounds, commonly referred to as silanes,

particularly alkylsilanes. While the present invention departs from this chemistry in important and substantial ways, this body of chemistry is fundamental to the present invention.

5 Silane chemistry is quite well known and reported in the literature. An excellent treatment of the chemistry of silanes is given by Sommers, L.H.; Mitch, F.A; and Goldberg, G.M., "Synthesis and properties of Compounds with a Framework of Alternate Silicon and Carbon Atoms, 10 J.A.C.S., 71, 2746, (1949). Surveys of this body of chemistry are found in KIRK-OTHMER, **ENCYCLOPEDIA OF CHEMICAL TECHNOLOGY**, 3rd Ed. at Vol. 20, pp 887-911. The chemistry of organosilicon compounds is described in **ORGANOSILICON COMPOUNDS**, Bazant, Chvalovsky and 15 Rathovsky, Academic Press, Inc., New York, 1965.

 The literature on contact lenses and contact lenses and contact lenses containing silicone compounds is massive, including hundreds of patents. This massive body of literature is not considered analogous to the present 20 invention except as to the general techniques for forming optical contact lenses, e.g. cutting and polishing.

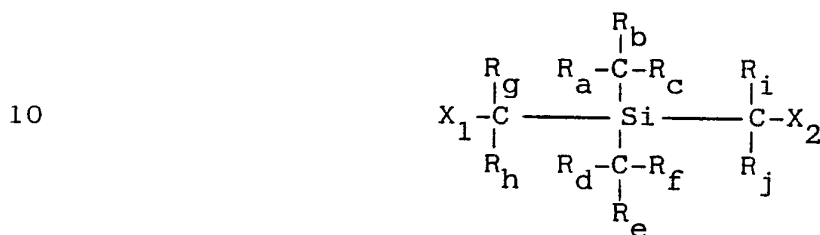
 Silanes have been utilized in preparative organic chemistry and for a number of specialty applications, including waterproofing compounds for mortar and fabrics 25 and the like, as accelerators in some polymer operations, and as intermediates in the preparation of organosiloxanes.

 While the chemistry, vis-a-vis reaction conditions, of alkyl silanes is known and reasonably well 30 understood, it has not, to the inventor's knowledge, been proposed to use such materials as the principal constituent polymer in contact lenses. In particular, the unique characteristics of such contact lenses has not been reported, insofar as is known to the inventor. Given the 35 uncertainty as to lens characteristics of given polymer

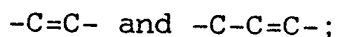
systems, there was no reason to expect that such materials would be useful as lens polymers.

Summary of the Invention

The present invention relates to a novel class of contact lenses comprising polymers resulting from the polymerization or copolymerization alkyl silanes, having the general structure:

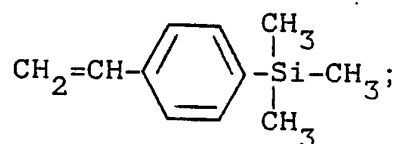


wherein R_a through R_j are hydrogen or alkyl, aryl, aralkyl, or silyl moieties, which may include vinyl, allyl, acrylyl, acrylic, methacrylic, ethacrylic, or pyrrolidinonyl substituents and may also contain up to about 35 weight percent siloxyl, and wherein either X_1 or X_2 , or both X_1 and X_2 are vinyl polymerizable group containing moieties. The term "vinyl polymerizable group" is used here in a particular sense to mean a polymerizable group containing the carbon-carbon double bond which is polymerized in the formation of polyvinyl polymers, i.e. the following structures:

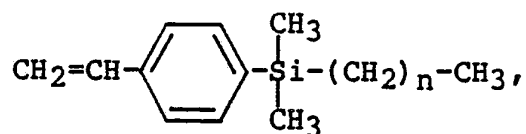


exemplary of which vinyl polymerizable groups are: vinyl, allyl, acrylyl, acrylyl, methacrylyl, or styryl.

Exemplary of the monomers suitable for forming the polymers and copolymers of this invention are the following:

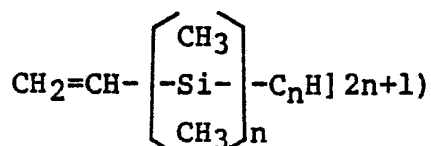


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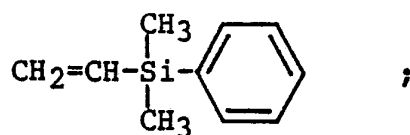


wherein n is a positive integer from 1 to 5,
preferably 1 to 3;

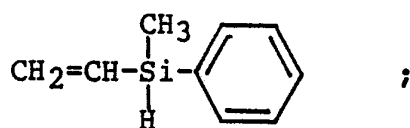
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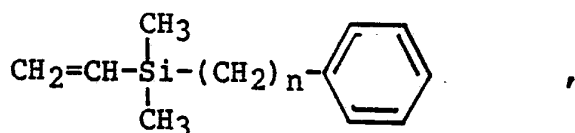
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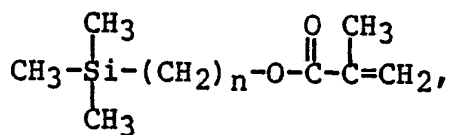


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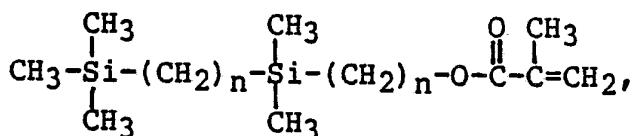
wherein n is a positive integer from 1 to 5,
preferably 1 to 3;

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wherein n is a positive integer from 1 to 5,
preferably 1 to 3;

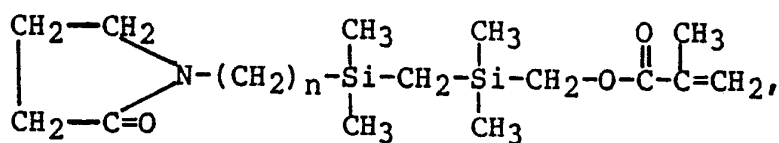
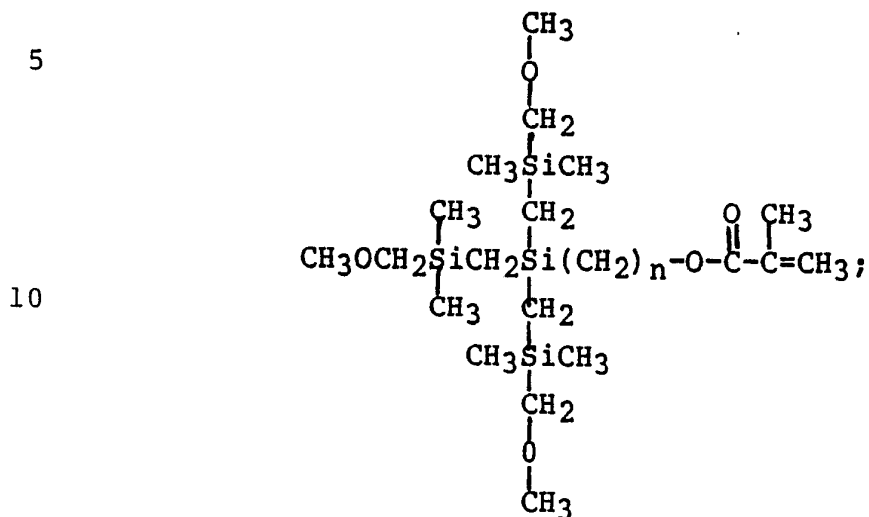
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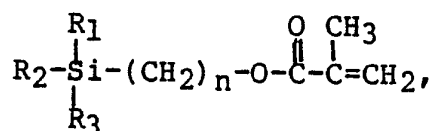
wherein n is a positive integer from 1 to 5,
preferably 1 to 3;



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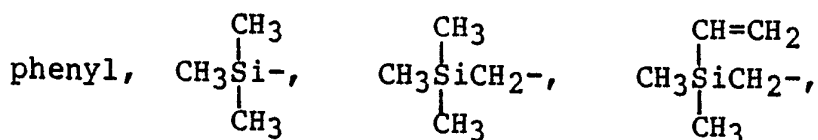
wherein n is a positive integer from 1 to 5,
preferably 1 to 3;

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wherein n is a positive integer from 1 to 5,
preferably 1 to 3 and R_1 , R_2 and R_3 are selected from
the group consisting of methyl,

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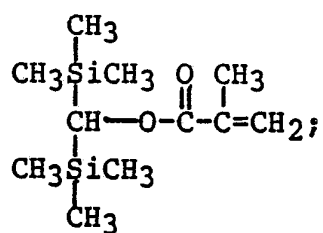


alkylmethoxy, phenylmethyl, and N-alkyl-pyrrol-
idinonyl

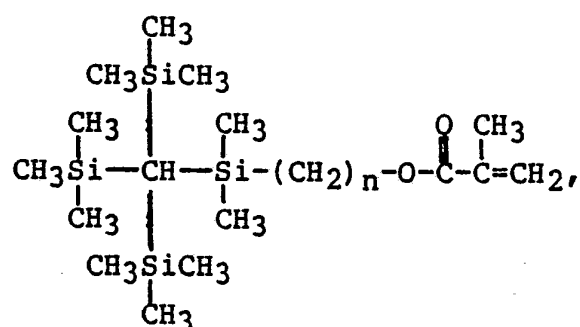
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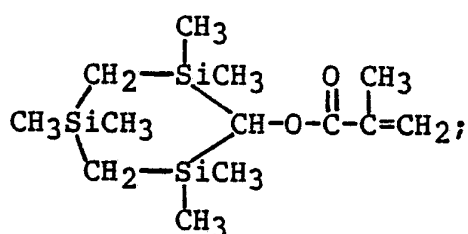
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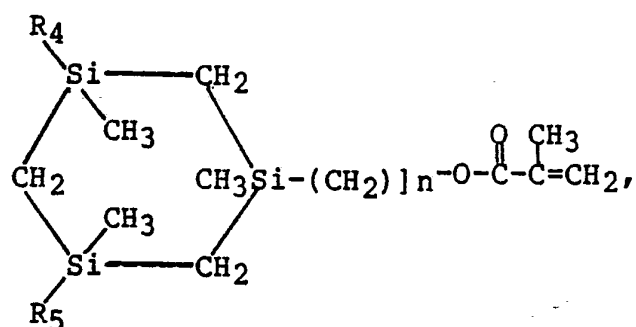
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wherein n is a positive integer from 1 to 5, preferably 1 to 3;

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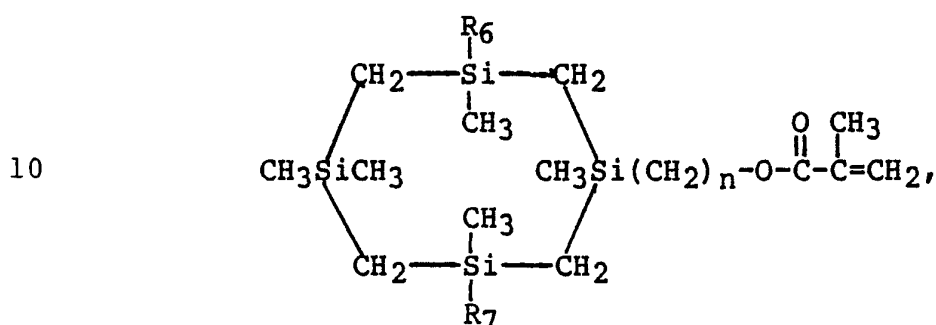
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wherein n is a positive integer from 1 to 5, preferably 1 to 3, and R_4 and R_5 are selected from the group consisting of methyl,

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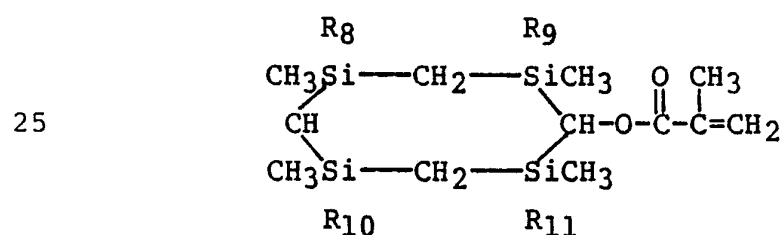
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phenyl, $\text{CH}_3\text{Si}(\text{CH}_3)_2-$, $\text{CH}_3\text{Si}(\text{CH}_3)\text{CH}_2-$, $\text{CH}_3\text{Si}(\text{CH}_3)\text{CH}_2-$,
 alkylmethoxy, phenylmethyl, and N-alkyl-pyrrol-
 idinonyl



15 wherein n is a positive integer from 1 to 5, preferably 1 to 3, and R_6 and R_7 are selected from the group consisting of methyl,

phenyl, $\text{CH}_3\text{Si}(\text{CH}_3)_2-$, $\text{CH}_3\text{Si}(\text{CH}_3)\text{CH}_2-$, $\text{CH}_3\text{Si}(\text{CH}_3)\text{CH}_2-$,
 alkylmethoxy, phenylmethyl, and N-alkyl-pyrrol-
 idinonyl

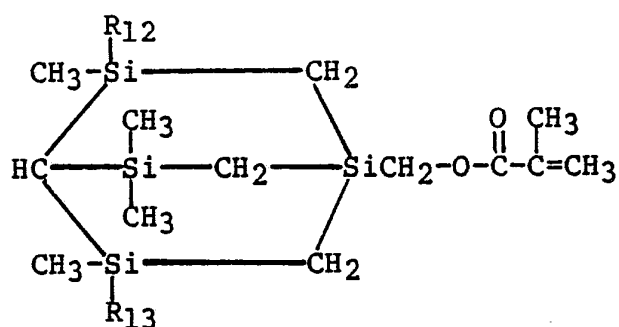


30 wherein n is a positive integer from 1 to 5, preferably 1 to 3, and $R_8 - R_{11}$ are selected from the group consisting of methyl,

phenyl, $\text{CH}_3\text{Si}(\text{CH}_3)_2-$, $\text{CH}_3\text{Si}(\text{CH}_3)\text{CH}_2-$, $\text{CH}_3\text{Si}(\text{CH}_3)\text{CH}_2-$,
 alkylmethoxy, phenylmethyl, and N-alkyl-pyrrol-
 idinonyl and

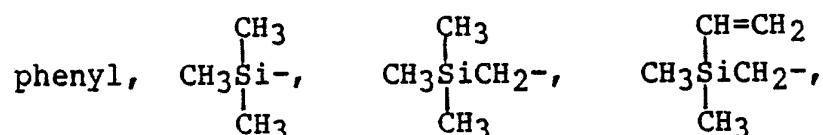
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wherein n is a positive integer from 1 to 5, preferably 1 to 3, and R_{12} and R_{13} are selected from the group consisting of methyl,

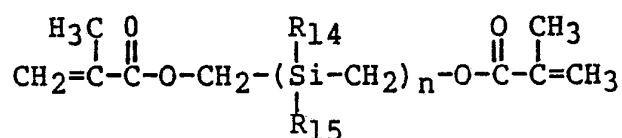


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alkylmethoxy, phenylmethyl, and N-alkylpyrrolidinonyl.

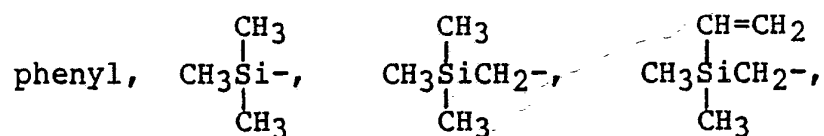
Silanes with two polymerizable groups and polymers thereof and lenses of such polymers are also contemplated within the scope of the invention. Exemplary of such monomers are:

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wherein n is a positive integer from 1 to 5, preferably 1 to 3, and R_{14} and R_{15} are selected from the group consisting of methyl,

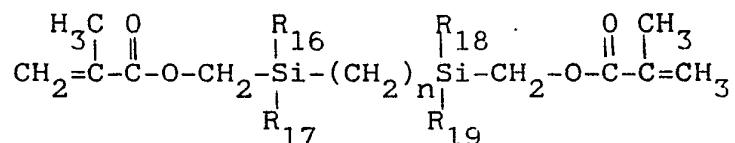


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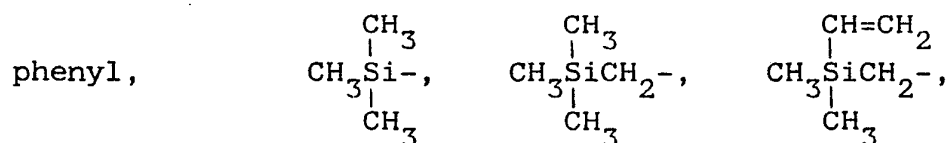
alkylmethoxy, phenylmethyl, and N-alkyl-pyrrolidinonyl

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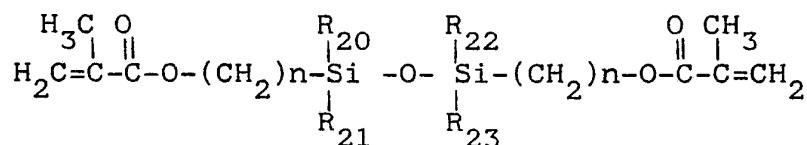


wherein n is a positive integer from 1 to 5, preferably 1 to 3, and R_{16} to R_{19} are selected from the group consisting of methyl,

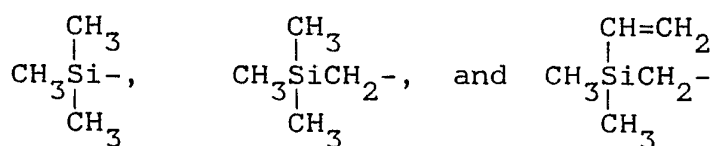


alkylmethoxy, phenylmethyl, and N-alkylpyrrolidinonyl.

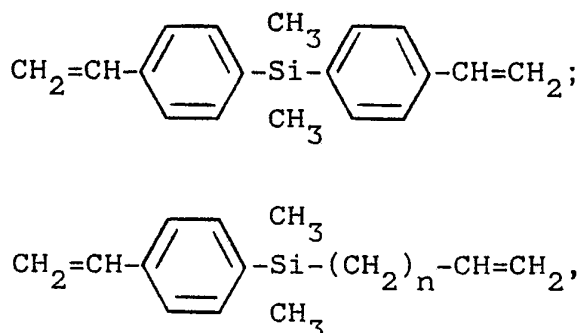
A monomer which is predominantly silane is:



wherein n is a positive integer from 1 to 5, preferably 1 to 3, and R_{20} to R_{23} are selected from the group consisting of



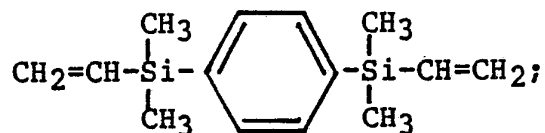
Other monomers which include two polymerizable groups include:



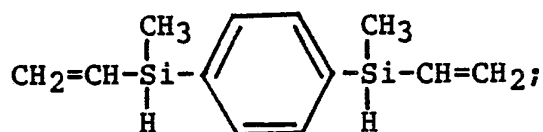
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wherein n is a positive integer from 1 to 5, preferably 1 to 3,;

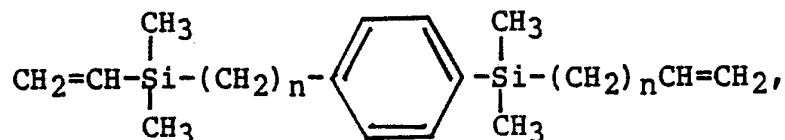
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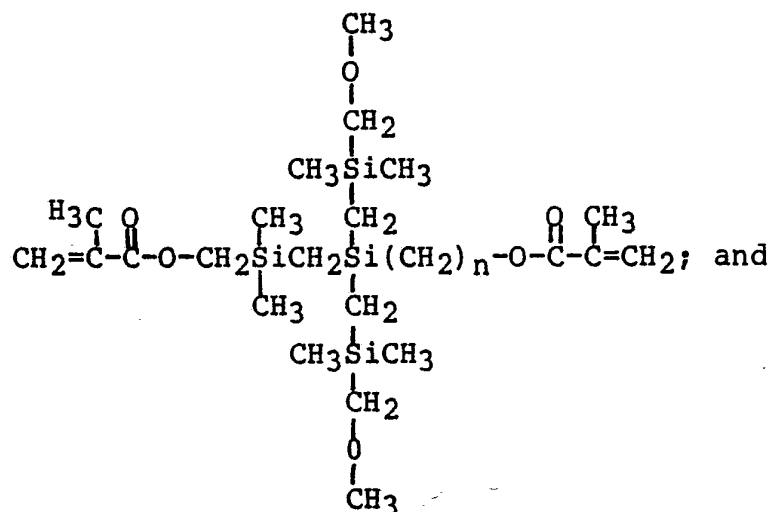


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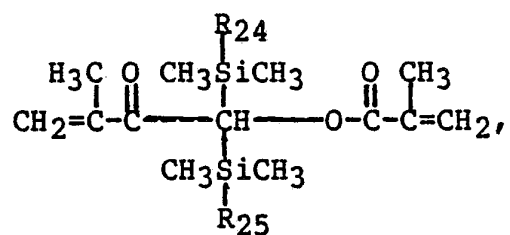
wherein n is a positive integer from 1 to 5, preferably 1 to 3,;

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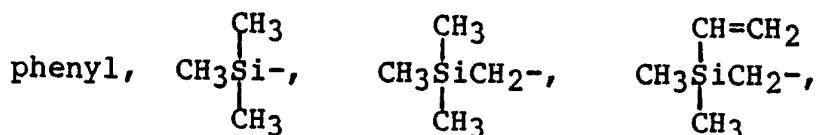
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wherein R₂₄ and R₂₅ are selected from the group consisting of methyl,



alkylmethoxy, phenylmethyl, and N-alkylpyrrolidinonyl.

In general, vinyl, allyl, acrylallyl, acrylic, methacrylic or ethacrylic derivatives of the compounds referred to which include one or more polymerizable groups such as vinyl, allyl, acrylic, methacrylic or ethacrylic may be considered equivalent to the specific, exemplary monomers, and polymers and copolymers of the same may be used as contact lens materials and lenses.

The alkylsilane polymer lenses of this invention have been discovered to have extremely beneficial, and most unexpected and unpredictable properties as contact lenses. For example, the most comparable lenses, of siloxyl based polymers, have an oxygen permeability, Dk value (see, e.g. Fatt, I. and St. Helen, R., Oxygen Tension Under an Oxygen-Permeable Contact Lens, American Journal of Optrometry, July 1971, pp.545-555, for a discussion of Dk values) in the 20's, the highest being about 30 to 32. The alkylsilane polymer lenses of this invention have a calculated Dk value of as high as 40 or more! The extremely high Dk value, as shown by wearer comfort, has been demonstrated for the contact lenses of this invention. In addition, these alkylsilane polymer lenses have an even greater resistance to protein contamination than the silicone polymer lenses. These alkylsilane polymer lenses are also harder and, very surprisingly, can be made wettable by inclusion of appropriate hydrophylic substituents much easier than comparable silicone polymer contact lenses! These very surprising advantages, coupled with good optical quality could not have been

predicted, or even guessed at in advance. These lens polymers can be formulated with a relatively high phenyl substituent content, giving lenses having a high index of refraction which can be made thinner and lighter than conventional contact lenses, and more easily fabricated into bifocal lenses than is possible with conventional and know lens polymers. Surface characteristics can be modified by inclusion of specific moieties in the polymer; for example, methoxy alkyl, ethoxy alkyl, or n-alkylpyrrolidinone may be included to improve wettability. Monomers having two polymerizable groups may be used, thus resulting in a fully crosslinked lens polymer. It is even possible to prepare highly hydrated lenses from the polymers of this invention!

Description of the Preferred Embodiment

No new silane chemistry, per se, is involved in the present invention; rather, it has been discovered that alkylsilane polymer contact lenses have most unexpected and unpredicted advantages over other lenses and, more particularly, over the most comparable lenses, those formed of silicone polymers.

The alkylsilanes used in forming the polymers from which the lenses of the present invention are manufactured are most conveniently prepared by the action of a polymerizable vinyl group containing moiety, e.g. methacrylic acid, on a chloroalkyl or bromoalkyl substituted silane, such as chloromethyl trimethylsilane or di-chloromethyl dimethylsilane, in the presence of a base such as pyridine or triethyl amine. The higher homologues of the series are conveniently prepared by the action of the Grignard Reagent of a silane, such as trimethylsilylmethyl magnesium chloride on a chlorosilyl-alkyl methacrylate, e.g. trichlorosilyl propyl methacrylate, to give tris(trimethylsilylmethyl) silyl-propyl methacrylate.

Example A**Trimethylsilyl Methyl Methacrylate**

Trimethylsilylmethyl methacrylate was prepared as follows: Methacrylic acid (29.6 g) was dissolved in dry ether (600 ml), sodium carbonate (18.6 g) was added slowly to form the sodium salt. Chloromethyl trimethyl silane (42.2 g) was added to the gelatinous solid formed from the preceeding salt forming reaction, followed by the addition of hydroquinone (1.00 g). The mixture was refluxed for 72 hours, washed with water, dried over magnesium sulfate, filtered and distilled giving a 21.7 g of product which boiled at 29.5°C. at 0.3 mm Hg, 37% of theoretical yield. The product was washed with basic carbonate solution until the wash was free of color and then washed with distilled water to remove any hydroquinone which may have been carried over during distillation, and dried over magnesium sulfate and stored under refrigeration.

Example B**Phenyldimethylsilyl Methyl Methacrylate**

Phenyldimethylsilyl Methyl Methacrylate was prepared by reacting phenyl dimethylul chloromethyl silane (161 g) with methacrylic acid (132 g) and triethylamine (132 g) in benzene (300 ml), with hydroquinone (1.0 g) added to inhibit polymerization during the reaction. The mixture was refluxed for 64 hours. The resulting product was washed, dried and distilled, and the boiling point of the product was found to be 86°C. at 0.1 mm Hg. The yield, 105.2 g, was 51.5% of theoretical. The product was further washed and stored as in example I.

These procedures are, of course, well known reactions. Similar reactions and techniques are suitable for the preparation of the monomers of interest.

Lens Manufacture

The following general technique was followed in the

preparation of lens blanks and lenses:

Monomers in the specified ratio and initiator were thoroughly mixed and dried over magnesium sulfate and filtered. The dried, filtered monomer mixture was placed
 5 in molds under nitrogen atmosphere and cured by slowly raising the temperature to about 100°C. for about 2 hours followed by a reduction to a post-cure temperature of about 80°C. for a post cure of about 15 hours. The resulting lens blank was examined and is then machined to form contact
 10 lenses according to conventional procedures for the manufacture of contact lenses.

The following examples of lenses formed by the technique described exemplify the invention.

Lens Material No. 1

15	Trimethylsilyl methyl methacrylate	45%*
	Methyl methacrylate	41%
	N-vinyl pyrrolidinone	3%
	Methacrylic acid	6%
	Ethylene glycol dimethacrylate	5%
20	Initiator**	(Trace)
	* Percentages in all examples by weight.	
	** 2,2'azobis-2,4-dimethyl-4-methoxy- valeronitrile 0.001-0.5% in all Lens examples	
	Initial Cure Temperature	102°C.
25	Initial Cure Time	2 hours.
	Post Cure Temperature	82-85°C.
	Post Cure Time	16 hours.
	Lens Qualities:	
	Clarity	Excellent
30	Hardness	Good
	Machinability	Good
	Wettability	Good
	Dimensional stability	Excellent

Lens Material No. 2

35	Trimethylsilyl methyl methacrylate	30%*
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	Methyl methacrylate	56%
	N-vinyl pyrrolidinone	3%
	Methacrylic acid	5%
	Ethylene glycol dimethacrylate	6%
5	Initiator**	(Trace)
	Initial Cure Temperature	76°C.
	Initial Cure Time	3/4 hours.
	Post Cure Temperature	52-58°C.
	Post Cure Time	18 hours.
10	Lens Qualities:	
	Clarity	Excellent
	Hardness	Good
	Machinability	Good
	Wettability	Good
15	Dimensional stability	Excellent

Lens Material No. 3

	Trimethylsilyl methyl methacrylate	60%*
	Methyl methacrylate	26%
	N-vinyl pyrrolidinone	3%
20	Methacrylic acid	5%
	Ethylene glycol dimethacrylate	6%
	Initiator**	(Trace)
	Initial Cure Temperature	69°C.
	Initial Cure Time	3/4 hours.
25	Post Cure Temperature	52-58°C.
	Post Cure Time	18 hours.
	Lens Qualities:	
	Clarity	Excellent
	Hardness	Good
30	Machinability	Good
	Wettability	Good
	Dimensional stability	Excellent

Lens Material No. 4

	Trimethylsilyl methyl methacrylate	45%*
35	Methyl methacrylate	41%

16

	N-vinyl pyrrolidinone	3%
	Methacrylic acid	6%
	Ethylene glycol dimethacrylate	5%
	Initiator**	(Trace)
5	Initial Cure Temperature	102°C.
	Initial Cure Time	2 hours.
	Post Cure Temperature	82-85°C.
	Post Cure Time	16 hours.
	Lens Qualities:	
10	Clarity	Excellent
	Hardness	Good
	Machinability	Good
	Wettability	Good
	Dimensional stability	Excellent
15	Lens Material No. 5	
	Trimethylsilyl methyl methacrylate	30%*
	Methyl methacrylate	56%
	N-vinyl pyrrolidinone	3%
	Methacrylic acid	5%
20	Ethylene glycol dimethacrylate	6%
	Initiator**	(Trace)
	Initial Cure Temperature	76°C.
	Initial Cure Time	3/4 hours.
	Post Cure Temperature	52-58°C.
25	Post Cure Time	18 hours.
	Lens Qualities:	
	Clarity	Excellent
	Hardness	Good
	Machinability	Good
30	Wettability	Good
	Dimensional stability	Excellent
	Lens Material No. 6	
	Trimethylsilyl methyl methacrylate	60%*
	Methyl methacrylate	26%
35	N-vinyl pyrrolidinone	3%

17

	Methacrylic acid	5%
	Ethylene glycol dimethacrylate	6%
	Initiator**	(Trace)
	Initial Cure Temperature	69°C.
5	Initial Cure Time	3/4 hours.
	Post Cure Temperature	52-58°C.
	Post Cure Time	18 hours.
	Lens Qualities:	
	Clarity	Excellent
10	Hardness	Good
	Machinability	Good
	Wettability	Good
	Dimensional stability	Excellent

Lens Material No. 7

15	Trimethylsilyl methyl methacrylate	86%*
	N-vinyl pyrrolidinone	3%
	Methacrylic acid	5%
	Ethylene glycol dimethacrylate	5%
	Initiator**	(Trace)
20	Initial Cure Temperature	57°C.
	Initial Cure Time	3/4 hours.
	Post Cure Temperature	57°C.
	Post Cure Time	21 hours.
	Lens Qualities:	
25	Clarity	Excellent
	Hardness	Good
	Machinability	Good
	Wettability	Good
	Dimensional stability	Exceptional

Lens Material No. 8

	Trimethylsilyl methyl methacrylate	40%*
	Methyl methacrylate	31%
	N-vinyl pyrrolidinone	3%
35	1,3,Bis(methacryloxy propyl)1,1';3,3'- tetrakis(trimethylsiloxy)disiloxane	20%

18

	Methacrylic acid	6%
	Initiator**	(Trace)
	Initial Cure Temperature	80°C.
	Initial Cure Time	3/4 hours.
5	Post Cure Temperature	55-58°C.
	Post Cure Time	21 hours.
	Lens Qualities:	
	Clarity	Excellent
	Hardness	Good
10	Machinability	Good
	Wettability	Good
	Dimensional stability	Excellent
	Lens Material No. 9	
	Trimethylsilyl methyl methacrylate	25%*
15	Methyl methacrylate	47%
	N-vinyl pyrrolidinone	3%
	Trimethoxysilyl propyl methacrylate	25%
	Initiator**	Trace
	Initial Cure Temperature	58°C.
20	Initial Cure Time	1 1/4 hours.
	Post Cure Temperature	58°C.
	Post Cure Time	20 hours.
	Lens Qualities:	
	Clarity	Excellent
25	Hardness	Good
	Machinability	Good
	Wettability	Good
	Dimensional stability	Excellent
	Lens Material No. 10	
30	Phenylmethylsilyl methylmethacrylate	50%*
	Methyl methacrylate	36%
	N-vinyl pyrrolidinone	3%
	Methacrylic acid	6%
	Ethylene glycol dimethacrylate	5%
35	Initiator**	Trace

19

	Initial Cure Temperature	58°C.
	Initial Cure Time	1 1/4 hours.
	Post Cure Temperature	58°C.
	Post Cure Time	20 hours.
5	Lens Qualities:	
	Clarity	Excellent
	Hardness	Good
	Machinability	Good
	Wettability	Good
10	Dimensional stability	Excellent
	This lens material has an exceptionally high refractive index, making it ideally suited to the manufacture of bifocal and thin lenses.	
	Phenyldimethylsilyl methylemethacrylate	60%*
15	Methyl methacrylate	26%
	N-vinyl pyrrolidinone	3%
	Methacrylic acid	6%
	Ethylene glycol dimethacrylate	5%
	Initiator**	Trace
20	Initial cure temperature	104°
	Initial cure time	2 hours
	Post Cure Temperature	83-85°C.
	Post Cure Time	16 hours.
	Lens Qualities:	
25	Clarity	Excellent
	Hardness	Good
	Machinability	Good
	Wettability	Good
	Dimensional stability	Excellent
30	This lens material has an exceptionally high refractive index, making it ideally suited to the manufacture of bifocal and thin lenses.	
	Lens Material No. 12	
	Phenylmethysilyl methylemethacrylate	86%*
35	N-vinyl pyrrolidinone	3%

20

	Methacrylic acid	6%
	Ethylene glycol dimethacrylate	5%
	Initiator**	Trace
	Initial Cure Temperature	104°C.
5	Initial Cure Time	2 hours.
	Post Cure Temperature	84-86°C.
	Post Cure Time	18 hours.
	Lens Qualities:	
	Clarity	Excellent
10	Hardness	Good
	Machinability	Good
	Wettability	Good
	Dimensional stability	Excellent
15	This lens material has an exceptionally high refractive index, making it ideally suited to the manufacture of bifocal and thin lenses.	

Lens Material No. 13

	Trimethoxysilyl propylmethacrylate	45%*
	Methyl methacrylate	41%
20	N-vinyl pyrrolidinone	3%
	Methacrylic acid	5%
	Ethylene glycol dimethacrylate	5%
	Initiator**	Trace
	Initial Cure Temperature	102°C.
25	Initial Cure Time	2 hours.
	Post Cure Temperature	82-85°C.
	Post Cure Time	16 hours.
	Lens Qualities:	
	Clarity	Excellent
30	Hardness	Good
	Machinability	Good
	Wettability	Good
	Dimensional stability	Excellent

Lens Material No. 14

35	Trimethylsilyl methyl methacrylate	45%*
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	Hydroxyethyl methacrylate	10%
	N-vinyl pyrrolidinone	50%
	Methacrylic acid	4%
	Ethylene glycol dimethacrylate	1%
5	Initiator**	Trace
	Initial Cure Temperature	57°C.
	Initial Cure Time	2 hours.
	Post Cure Temperature	57°C.
	Post Cure Time	20 hours.
10	Lens Qualities:	
	Clarity	Excellent
	Hardness	Good
	Machinability	Good
	Wettability	Good
15	Dimensional stability	Excellent
	On hydrating in 0.9% saline solution, this lens material reached an equilibrium hydration level of 42.7%, providing a clear, rigid hydrating lens material.	
	Lens Material No. 15	
20	Phenyldimethylsilyl methylmethacrylate	20%*
	Hydroxyethyl methacrylate	74%
	Methacrylic acid	5%
	Triethylene glycol dimethacrylate	5%
	Initiator**	Trace
25	Initial Cure Temperature	102°C.
	Initial Cure Time	2 hours.
	Post Cure Temperature	84-86°C.
	Post Cure Time	18 hours.
	Lens Qualities:	
30	Clarity	Excellent
	Hardness	Good
	Machinability	Good
	Wettability	Good
	Dimensional stability	Excellent
35	This lens material has an exceptionally high refractive	

index, making it ideally suited to the manufacture of bifocal and thin lenses, and, additionally, reached an equilibrium of 18.5% hydration in 0.9% saline, thus providing a clear, rigid hydrating lens material.

Lens Material No. 16

Phenyltetramethyldisilylmethylene-

	methylemethacrylate	50%*
	Methyl methacrylate	38.5%
	Methacrylic acid	6%
10	Ethylene glycol dimethacrylate	5%
	N-vinylpyrrolidinone	3%
	Initiator**	Trace
	Initial Cure Temperature	100°C.
	Initial Cure Time	2 hours.
15	Post Cure Temperature	82-83°C.
	Post Cure Time	17 hours.
	Lens Qualities:	
	Clarity	Excellent
	Hardness	Good
20	Machinability	Good
	Wettability	Good
	Dimensional stability	Excellent

This lens material has an exceptionally high refractive index, making it ideally suited to the manufacture of bifocal and thin lenses.

Discussion and Equivalents

30 These lens materials had excellent optical properties and some had outstanding refractive index characteristics. All were ideal for the manufacture of high quality exceptionally comfortable lenses. Some had moderate to high hydration capacity, in addition to being excellent, clear comfortable lens characteristics.

One of the important discoveries of this invention is that these lens materials are far more comfortable for the wearer than the most nearly comparable lenses formed of

siloxanyl polymers, the silicone polymers of the prior art and of my earlier filed copending patent applications. Wearer comfort is somewhat subjective but reflects real differences. Two objective observations are believed to explain the unexpectedly high comfort factor of the lenses of this invention. First, oxygen permeability is very high, thus contributing to healthier eye tissue and greater comfort. Second, these lens materials are exceptionally resistant to the buildup of proteins on the lens surfaces.

A third, highly unexpected, factor believed to contribute to wearer comfort is the wettability of the lens materials of this invention. Silanes have typically been used in waterproofing applications and one would predict a highly hydrophobic lens material. Quite surprisingly, however, the lenses of this invention are quite hydrophylic and, indeed, in some formulations, hydrate to a moderate to high level.

Another surprising characteristic of lens materials of the present invention is that it is possible to form excellent lens materials with excellent optical, refractive, mechanical and comfort properties without the presence of methyl methacrylate, or with only very minor amounts of methyl methacrylate

It will be readily understood by those skilled in the art that the foregoing lens material formulations are only exemplary of a vast number of lens materials and lenses which can be manufactured within the scope of this invention. Many analogous and homologous monomers of the silane family may be substituted for those shown in the examples. Initiators may be selected from among the many which are suitable for initiating the polymerization of vinyl group containing monomers.

In general, the alkyl silanes of this invention comprise greater than 5% and preferably greater than 20% of the polymeric lenses and lens materials and my comprise

up to about 95%, preferably up to about 90%, of such materials and lenses. In the preferred embodiment, the lens material is formed from the polymerization of alkyl silanes with a cross-linking monomer and a monomer, such as
 5 n-vinylpyrrolidinone or hydroxyethyl methacrylate, or both, which contributes to the wettability or hydration of the lens, or to both wettability and hydration of lenses. This hydrophylic constituent may comprise, preferably, at least 2 to 3% and may comprise up to about 75% or more of the
 10 polymeric lens material.

Exemplary percentages of selected formulations are shown in the following tables:

I

	Trimethylsilyl methyl methacrylate	5 to 95 weight %
15	Methyl methacrylate	1 to 50 weight %
	N-vinyl pyrrolidinone	1 to 50 weight %
	Methacrylic acid	1 to 10 weight %
	Ethylene glycol dimethacrylate	1 to 10 weight %

II

20	Trimethylsilyl methyl methacrylate	20 to 90 weight %
	Methyl methacrylate	20 to 60 weight %
	N-vinyl pyrrolidinone	1 to 10 weight %

III

	Trimethylsilyl methyl methacrylate	20 to 90 weight %
25	N-vinyl pyrrolidinone	3 to 60 weight %
	Methacrylic acid	1 to 10 weight %

IV

	Trimethylsilyl methyl methacrylate	20 to 95 weight %
	N-vinyl pyrrolidinone	5 to 80 weight %

30

V

	Trimethylsilyl methyl methacrylate	25 to 95 weight %
	Hydroxyethyl methacrylate	5 to 75 weight %

VI

	Trimethylsilyl methyl methacrylate	5 to 95 weight %
35	Methyl methacrylate	5 to 90 weight %

25

1,3,Bis(methacryloxy propyl)1,1';3,3'-

tetrakis(trimethylsiloxy)disiloxane 5 to 35 weight %

VII

Phenylmethysilyl methylmethacrylate 5 to 95 weight %
 5 Methyl methacrylate 5 to 50 weight %

VIII

Phenylmethysilyl methylmethacrylate 5 to 95 weight %
 Methacrylic acid 5 to 50 weight %

IX

10 Trimethoxysilyl propylmethacrylate 5 to 95 weight %
 Methyl methacrylate 5 to 60 weight %

X

Trimethoxysilyl propylmethacrylate 5 to 95 weight %
 Hydroxyethyl methacrylate 5 to 50 weight %

15

XI

Phenyldimethysilyl methylmethacrylate 10 to 95 weight %
 Hydroxyethyl methylmethacrylate 5 to 90 weight %

XII

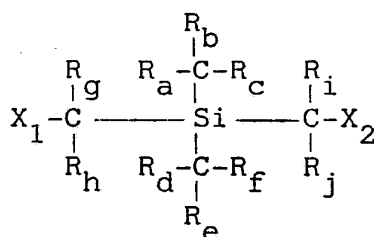
Phenyltetramethyldisilylmethylene-
 20 methylmethacrylate 5 to 95 weight %
 Methacrylic acid 5 to 60 weight %

Typically, in the preferred embodiments, a hydrophylic monomer would also comprise the polymerization mix.

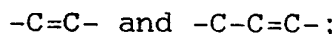
25 . It has also been found advantageous to include methacrylic acid and/or methyl methacrylate as a monomer in the formation of the polymerized lense material; however, one of the surprising discoveries of this invention is that high quality lenses can be formed without either of these
 30 constituents.

Polymeric materials resulting from the polymerization or copolymerization alkyl silanes, having the following general structures and lenses formed therefrom are within the contemplation and concept of this
 35 invention:

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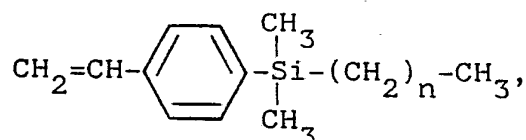
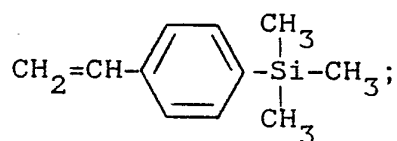
wherein R_a through R_j are hydrogen or alkyl, aryl, aralkyl, or silyl moieties, which may include vinyl, allyl, acrylyl, acrylic, methacrylic, ethacrylic, or pyrrolidinonyl substituents and may also contain up to about 35 weight percent siloxyl, and wherein either X_1 or X_2 , or both X_1 and X_2 are vinyl polymerizable group containing moieties. The term "vinyl polymerizable group" is used here in a particular sense to mean a polymerizable group containing the carbon-carbon double bond which is polymerized in the formation of polyvinyl polymers, i.e. the following structures:



exemplary of which vinyl polymerizable groups are:

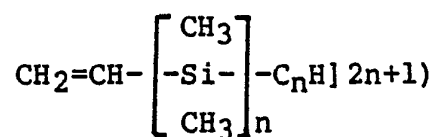
vinyl, allyl, acrylyl, acrylic, methacrylyl, or styryl.

Exemplary of the monomers suitable for forming the polymers and copolymers of this invention are the following:

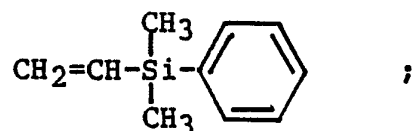


wherein n is a positive integer from 1 to 5, preferably 1 to 3;

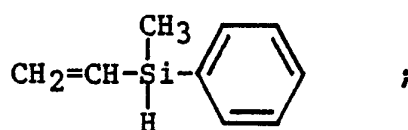
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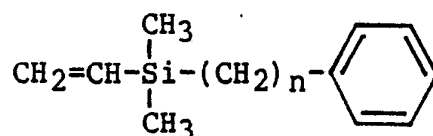
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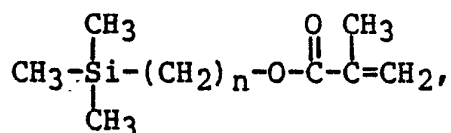


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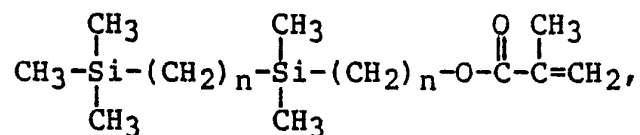
wherein n is a positive integer from 1 to 5,
preferably 1 to 3;

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wherein n is a positive integer from 1 to 5,
preferably 1 to 3;

25



wherein n is a positive integer from 1 to 5,
preferably 1 to 3;

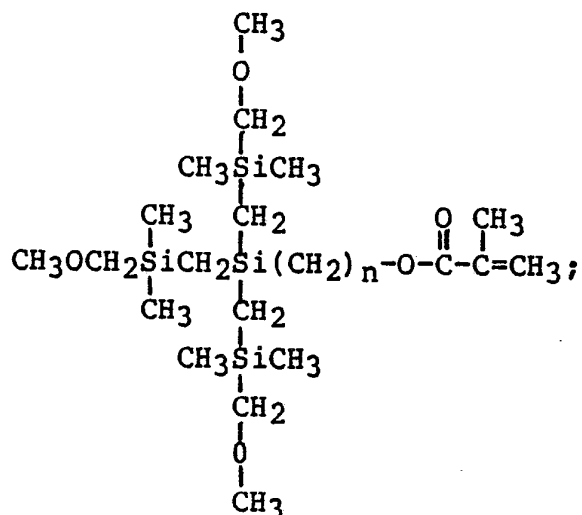
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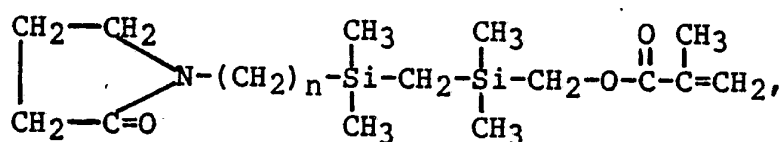
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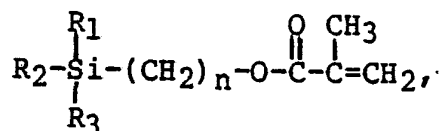


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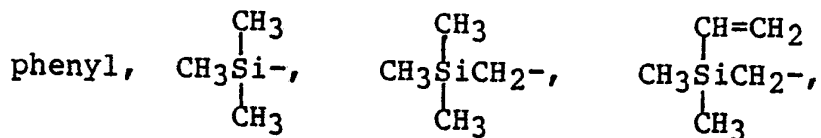
wherein n is a positive integer from 1 to 5, preferably 1 to 3;

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wherein n is a positive integer from 1 to 5, preferably 1 to 3 and R_1 , R_2 and R_3 are selected from the group consisting of methyl,

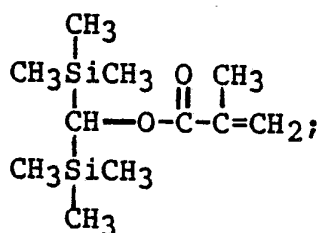
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alkylmethoxy, phenylmethyl, and N-alkyl-pyrrolidinonyl

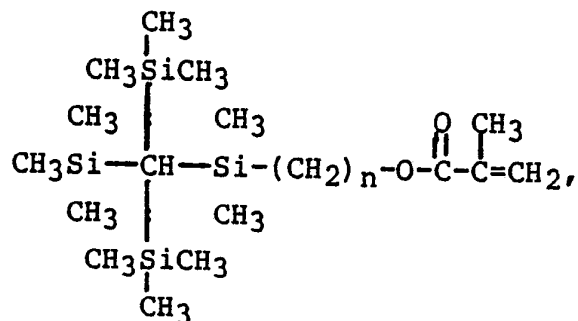
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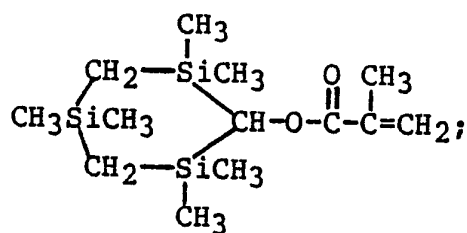
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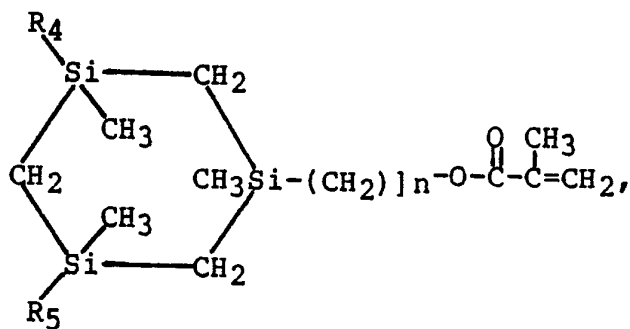
wherein n is a positive integer from 1 to 5, preferably 1 to 3;

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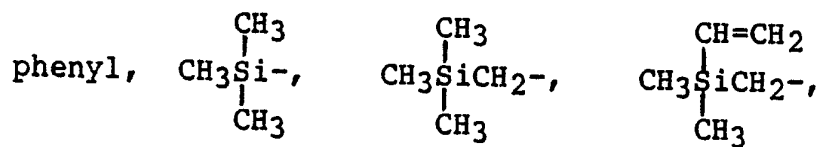
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wherein n is a positive integer from 1 to 5, preferably 1 to 3, and R_4 and R_5 are selected from the group consisting of methyl,

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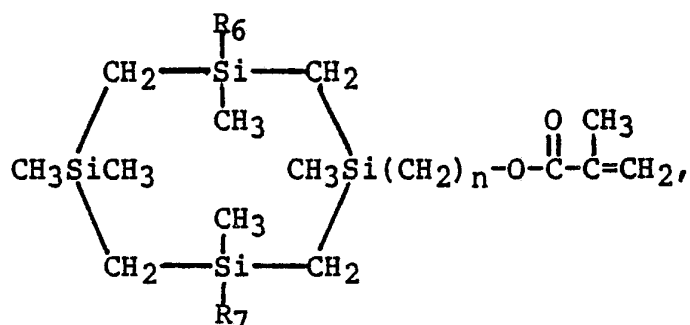


alkylmethoxy, phenylmethyl, and N-alkyl-pyrrolidinonyl

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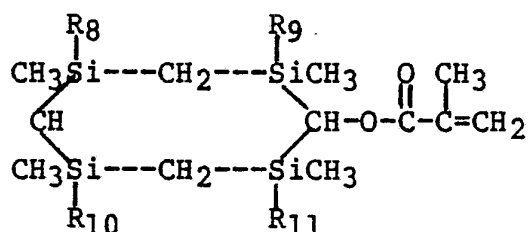
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wherein n is a positive integer from 1 to 5, preferably 1 to 3, and R_6 and R_7 are selected from the group consisting of methyl,

15

phenyl, $\text{CH}_3\text{Si}(\text{CH}_3)_2-$, $\text{CH}_3\text{Si}(\text{CH}_3)_2\text{CH}_2-$, $\text{CH}_3\text{Si}(\text{CH}_3)_2\text{CH}_2-$, $\text{CH}=\text{CH}_2$, $\text{CH}_3\text{Si}(\text{CH}_3)_2\text{CH}_2-$, alkylmethoxy, phenylmethyl, and N-alkyl-pyrrolidinonyl

20



wherein n is a positive integer from 1 to 5, preferably 1 to 3, and $\text{R}_8 - \text{R}_{11}$ are selected from the group consisting of methyl,

25

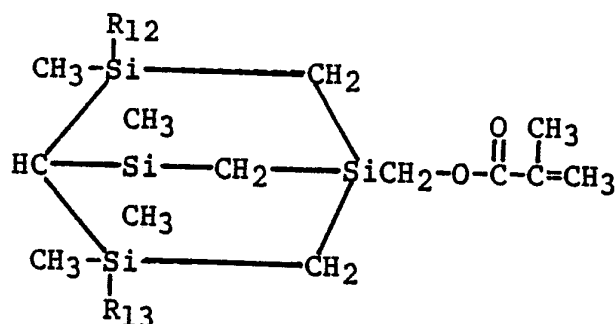
phenyl, $\text{CH}_3\text{Si}(\text{CH}_3)_2-$, $\text{CH}_3\text{Si}(\text{CH}_3)_2\text{CH}_2-$, $\text{CH}_3\text{Si}(\text{CH}_3)_2\text{CH}_2-$, $\text{CH}=\text{CH}_2$, $\text{CH}_3\text{Si}(\text{CH}_3)_2\text{CH}_2-$, alkylmethoxy, phenylmethyl, and N-alkyl-pyrrolidinonyl and

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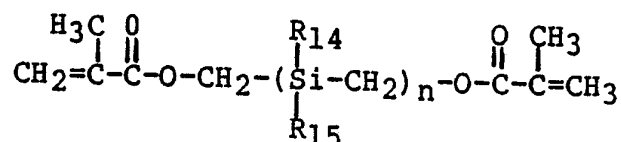
wherein n is a positive integer from 1 to 5, preferably 1 to 3, and R_{12} and R_{13} are selected from the group consisting of methyl,

15

phenyl, $\text{CH}_3\text{Si}(\text{CH}_3)_2-$, $\text{CH}_3\text{Si}(\text{CH}_3)\text{CH}_2-$, $\text{CH}_3\text{Si}(\text{CH}_3)\text{CH}_2-$, $\text{CH}=\text{CH}_2$, $\text{CH}_3\text{Si}(\text{CH}_3)\text{CH}_2-$, alkylmethoxy, phenylmethyl, and N-alkylpyrrolidinonyl.

20

Silanes with two polymerizable groups and polymers thereof and lenses of such polymers are also contemplated within the scope of the invention. Exemplary of such monomers are:



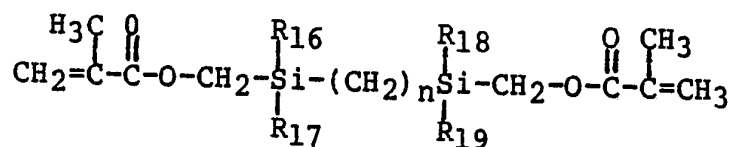
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wherein n is a positive integer from 1 to 5, preferably 1 to 3, and R_{14} and R_{15} are selected from the group consisting of methyl,

30

phenyl, $\text{CH}_3\text{Si}(\text{CH}_3)_2-$, $\text{CH}_3\text{Si}(\text{CH}_3)\text{CH}_2-$, $\text{CH}=\text{CH}_2$, $\text{CH}_3\text{Si}(\text{CH}_3)\text{CH}_2-$, alkylmethoxy, phenylmethyl, and N-alkyl-pyrrolidinonyl

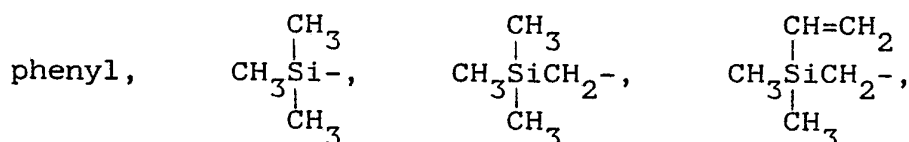
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wherein n is a positive integer from 1 to 5, preferably 1 to 3, and R_{16} to R_{19} are selected from the group consisting of methyl,

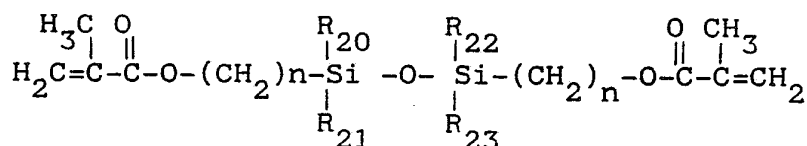
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alkylmethoxy, phenylmethyl, and N-alkylpyrrol-idinonyl.

10

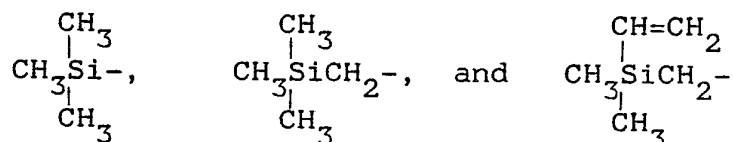
A monomer which is predominantly silane is:



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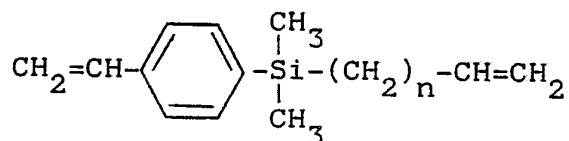
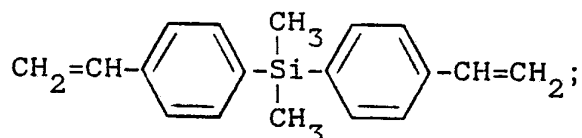
wherein n is a positive integer from 1 to 5, preferably 1 to 3, and R_{20} to R_{23} are selected from the group consisting of:

20



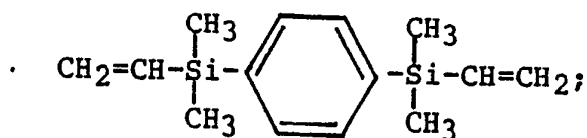
Other monomers which include two polymerizable groups include:

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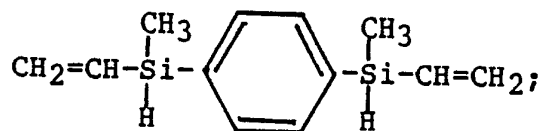


wherein n is a positive integer from 1 to 5, preferably 1 to 3;

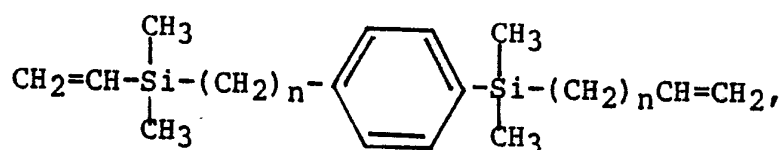
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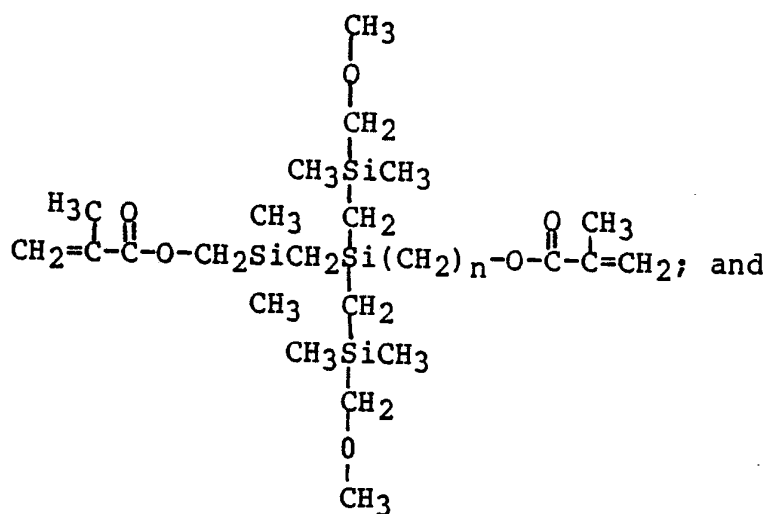
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wherein n is a positive integer from 1 to 5, preferably 1 to 3,;

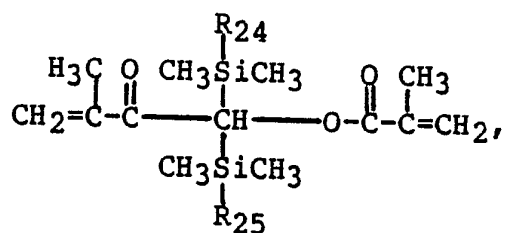
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wherein R_{24} and R_{25} are selected from the group consisting of methyl,

35

34

phenyl, $\text{CH}_3\overset{\text{CH}_3}{\underset{\text{CH}_3}{\text{Si}}}-$, $\text{CH}_3\overset{\text{CH}_3}{\underset{\text{CH}_3}{\text{Si}}}\text{CH}_2-$, $\text{CH}_3\overset{\text{CH}=\text{CH}_2}{\underset{\text{CH}_3}{\text{Si}}}\text{CH}_2-$,
 alkylmethoxy, phenylmethyl, and N-alkylpyrrol-
 idinonyl.

In general, vinyl, allyl, acrylallyl, acrylic, methacrylic or ethacrylic derivatives of the compounds referred to which include one or more polymerizable groups such as vinyl, allyl, acrylic, methacrylic or ethacrylic may be considered equivalent to the specific, exemplary monomers, and polymers and copolymers of the same may be used as contact lens materials and lenses.

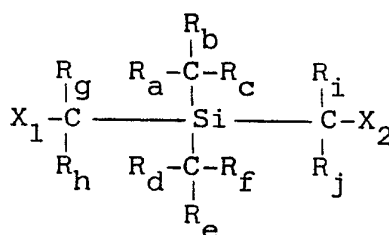
Industrial Application

This invention is useful in the optical industry and, particularly, in the manufacture of optical contact lenses for the correction of certain human visual defects.

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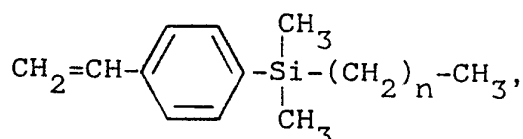
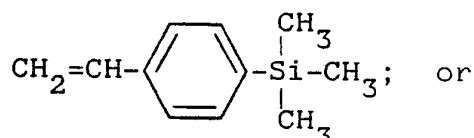
WHAT IS CLAIMED IS:

1. Contact lenses comprising polymers resulting from the polymerization or copolymerization alkyl silanes, having the general structure:



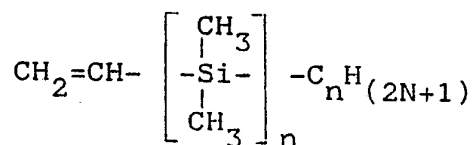
wherein R_a through R_j are hydrogen or alkyl, aryl, aralkyl, or silyl moieties, which may include vinyl, allyl, acrylyl, acrylic, methacrylic, ethacrylic, or pyrrolidinonyl substituents and may also contain up to about 35 weight percent siloxyl, and wherein either X_1 or X_2 , or both X_1 and X_2 are vinyl polymerizable group containing moieties.

2. Contact lenses comprising polymers and copolymers resulting from the polymerization or copolymerization of one or more of the following:



wherein n is a positive integer from 1 to 3.

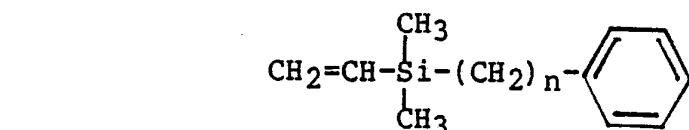
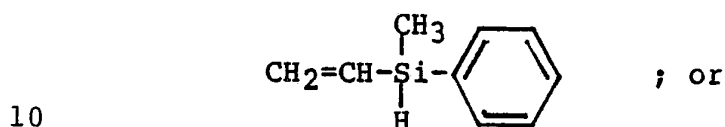
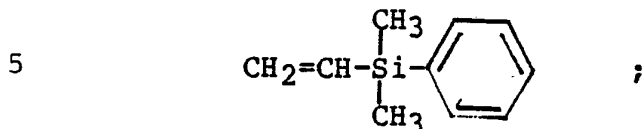
3. Contact lenses comprising polymers and copolymers resulting from the polymerization or copolymerization of:



wherein n is a positive integer from 1 to 3.

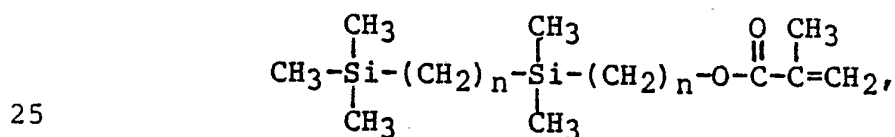
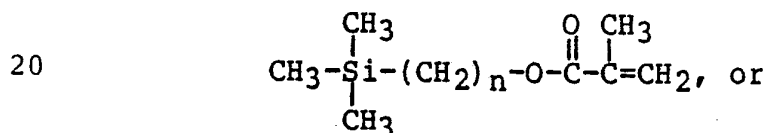
36

4. Contact lenses comprising polymers and copolymers resulting from the polymerization or copolymerization of one or more of:



wherein n is a positive integer from 1 to 3.

5. Contact lenses comprising polymers and copolymers resulting from the polymerization or copolymerization of one or more of:



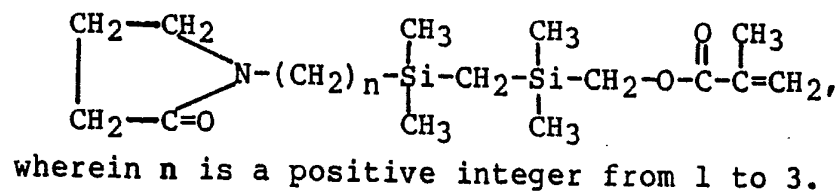
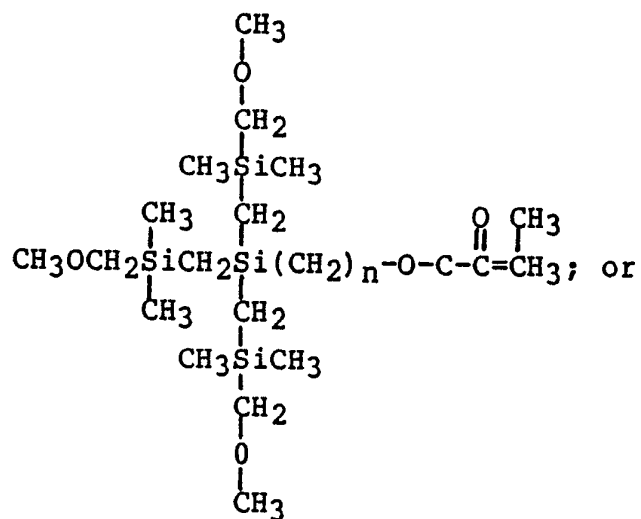
wherein n is a positive integer from 1 to 3.

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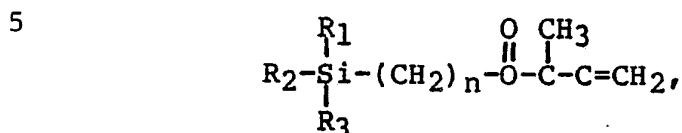
37

6. Contact lenses comprising polymers and copolymers resulting from the polymerization or copolymerization of one or more of the following:

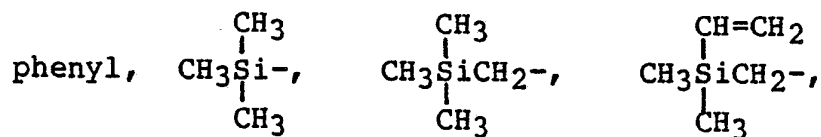


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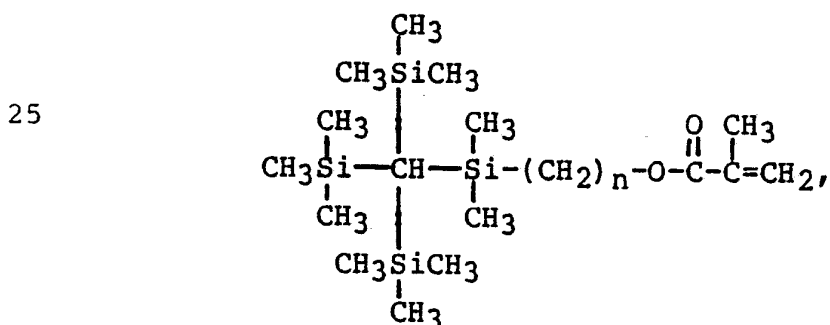
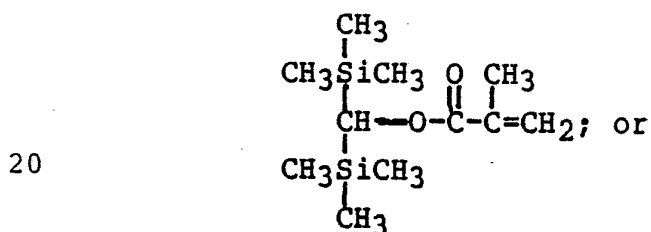
7. Contact lenses comprising polymers and copolymers resulting from the polymerization or copolymerization of one or more of the following:



wherein n is a positive integer from 1 to 3 and R_1 , R_2 and R_3 are selected from the group consisting of methyl,

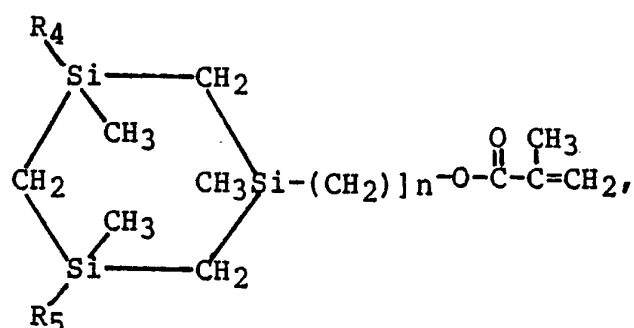
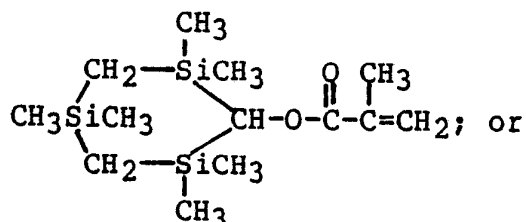


alkylmethoxy, phenylmethyl, and N-alkyl-pyrrolidinonyl;

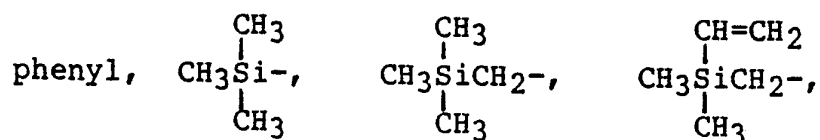


wherein n is a positive integer from 1 to 3.

8. Contact lenses comprising polymers and copolymers resulting from the polymerization or copolymerization of one or more of the following:



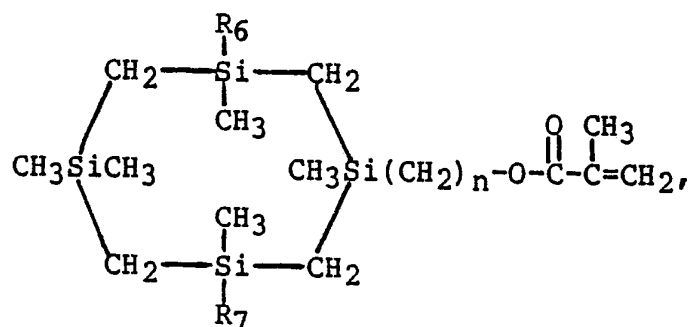
wherein n is a positive integer from 1 to 5, preferably 1 to 3, and R_4 and R_5 are selected from the group consisting of methyl,



alkylmethoxy, phenylmethyl, and N-alkyl-pyrrolidinonyl.

e.

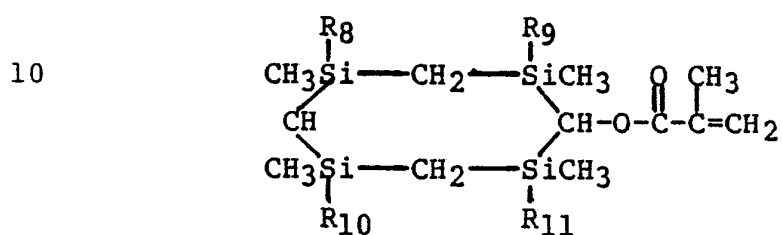
9. Contact lenses comprising polymers and copolymers resulting from the polymerization or copolymerization of one or more of the following:



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wherein n is a positive integer from 1 to 5, preferably 1 to 3, and R_6 and R_7 are selected from the group consisting of methyl,

5 phenyl, $\text{CH}_3\text{Si}(\text{CH}_3)_2-$, $\text{CH}_3\text{Si}(\text{CH}_3)_2\text{CH}_2-$, $\text{CH}_3\text{Si}(\text{CH}_3)_2\text{CH}_2-$,
 alkylmethoxy, phenylmethyl, and N-alkyl-pyrrolidinonyl;



15 wherein n is a positive integer from 1 to 5, preferably 1 to 3, and $R_8 - R_{11}$ are selected from the group consisting of methyl,

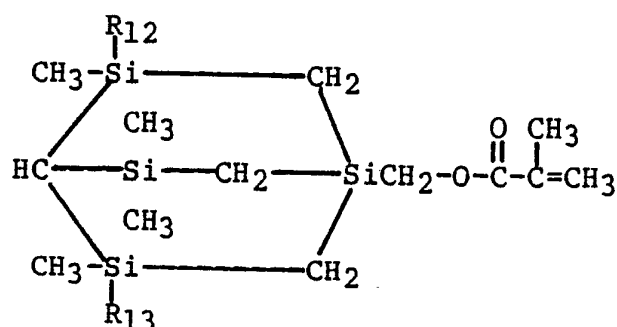
20 phenyl, $\text{CH}_3\text{Si}(\text{CH}_3)_2-$, $\text{CH}_3\text{Si}(\text{CH}_3)_2\text{CH}_2-$, $\text{CH}_3\text{Si}(\text{CH}_3)_2\text{CH}_2-$,
 alkylmethoxy, phenylmethyl, and N-alkyl-pyrrolidinonyl; or

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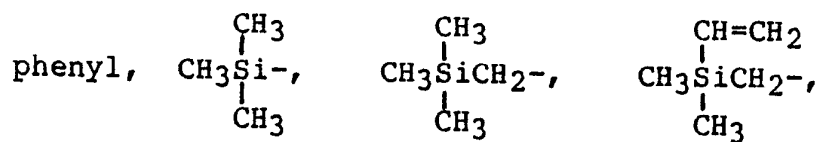
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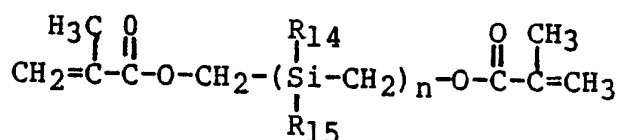


wherein n is a positive integer from 1 to 5, preferably 1 to 3, and R_{12} and R_{13} are selected from the group consisting of methyl,

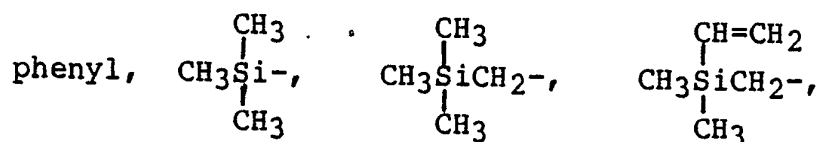


alkylmethoxy, phenylmethyl, and N-alkylpyrrolidinonyl.

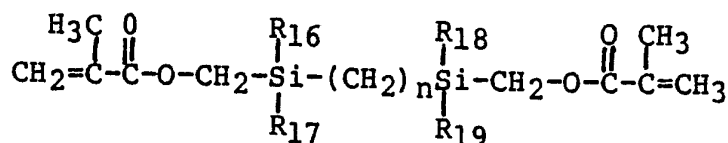
10. Contact lenses comprising polymers and copolymers resulting from the polymerization or copolymerization of one or more of the following:



wherein n is a positive integer from 1 to 5, preferably 1 to 3, and R_{14} and R_{15} are selected from the group consisting of methyl,



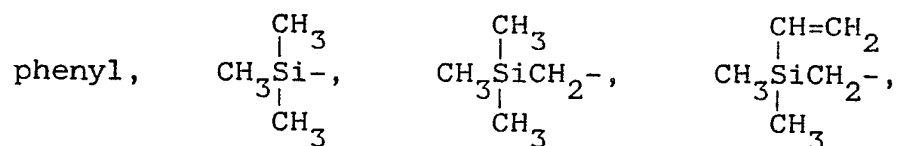
alkylmethoxy, phenylmethyl, and N-alkylpyrrolidinonyl; or



wherein n is a positive integer from 1 to 5, preferably 1 to 3, and R_{16} to R_{19} are selected from the group

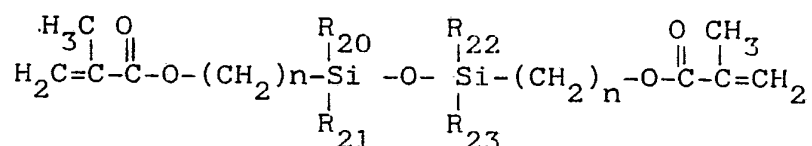
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consisting of methyl,

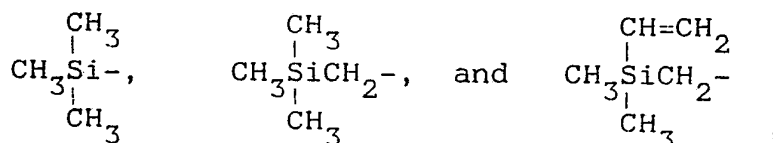


alkylmethoxy, phenylmethyl, and N-alkylpyrrolidinonyl.

11. Contact lenses comprising polymers and copolymers resulting from the polymerization or copolymerization of one or more of the following:



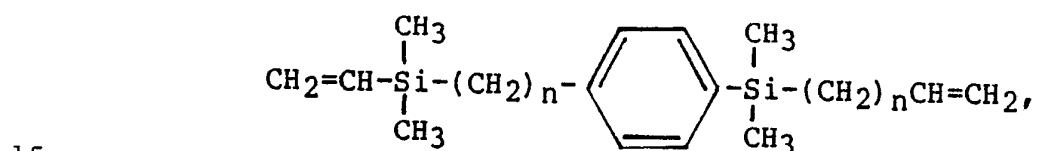
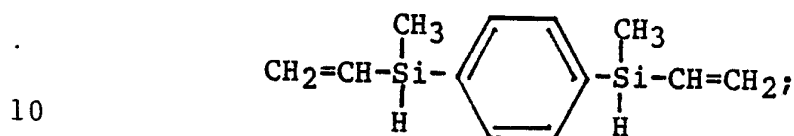
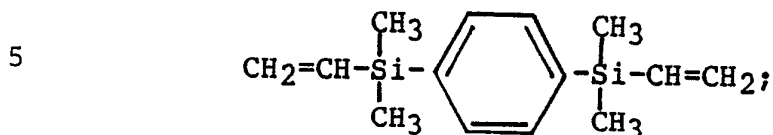
wherein n is a positive integer from 1 to 5, preferably 1 to 3, and R_{20} to R_{23} are selected from the group consisting of,



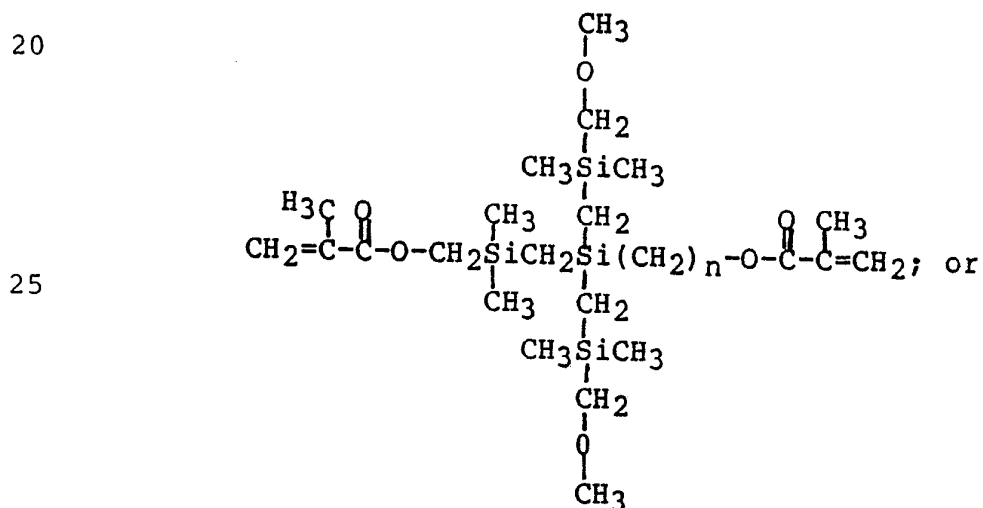
the polymer being characterized as predominantly a silane polymer.

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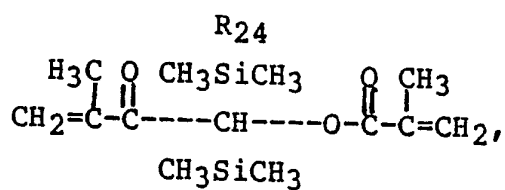
12. Contact lenses comprising polymers and copolymers resulting from the polymerization or copolymerization of one or more of the following:



wherein n is a positive integer from 1 to 5, preferably 1 to 3;

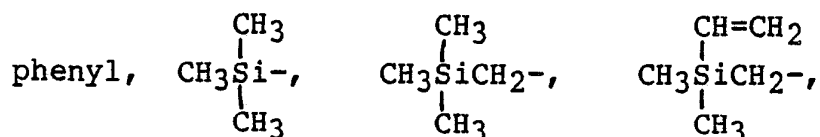


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wherein R₂₄ and R₂₅ are selected from the group consisting of methyl,



alkylmethoxy, phenylmethyl, and N-alkylpyrrolidinonyl.

13. A polymeric contact lens comprising the polymer resulting from the copolymerization of the following:

Trimethylsilyl methyl methacrylate	5 to 95 weight %
Methyl methacrylate	1 to 50 wieght %
N-vinyl pyrrolidinone	1 to 50 wieght %
Methacrylic acid	1 to 10 weight %
Ethylene glycol dimethacrylate	1 to 10 weight %.

14. A polymeric contact lens comprising the polymer resulting from the copolymerization of the following:

Trimethylsilyl methyl methacrylate	20 to 90 weight %
Methyl methacrylate	20 to 60 weight %
N-vinyl pyrrolidinone	1 to 10 weight %.

15. A polymeric contact lens comprising the polymer resulting from the copolymerization of the following:

Trimethylsilyl methyl methacrylate	20 to 90 weight %
N-vinyl pyrrolidinone	3 to 60 weight %
Methacrylic acid	1 to 10 weight %.

16. A polymeric contact lens comprising the polymer resulting from the copolymerization of the following:

Trimethylsilyl methyl methacrylate	20 to 95 weight %
N-vinyl pyrrolidinone	5 to 80 weight %.

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17. A polymeric contact lens comprising the polymer resulting from the copolymerization of the following:

5 Trimethylsilyl methyl methacrylate 25 to 95 weight %
Hydroxyethyl methacrylate 5 to 75 weight %.

18. A polymeric contact lens comprising the polymer resulting from the copolymerization of the following:

10 Trimethylsilyl methyl methacrylate 5 to 95 weight %
Methyl methacrylate 5 to 90 weight %.

1,3,Bis(methacryloxy propyl)1,1',3,3'-
tetrakis(trimethylsiloxy)disiloxane 5 to 35 weight %.

21. A polymeric contact lens comprising the polymer resulting from the copolymerization of the following:

15 Phenylmethethylsilyl methylmethacrylate 5 to 95 weight %
Methyl methacrylate 5 to 50 weight %.

22. A polymeric contact lens comprising the polymer resulting from the copolymerization of the following:

20 Phenylmethethylsilyl methylmethacrylate 5 to 95 weight %
Methacrylic acid 5 to 50 weight %.

23. A polymeric contact lens comprising the polymer resulting from the copolymerization of the following:

25 Trimethoxysilyl propylmethacrylate 5 to 95 weight %
Methyl methacrylate 5 to 60 weight %.

24. A polymeric contact lens comprising the polymer resulting from the copolymerization of the following:

30 Trimethoxysilyl propylmethacrylate 5 to 95 weight %
Hydroxyethyl methacrylate 5 to 50 weight %.

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25. A polymeric contact lens comprising the polymer resulting from the copolymerization of the following:

Phenyldimethylsilyl methacrylate 10 to 95 weight %
5 Hydroxyethyl methacrylate 5 to 90 weight %.

26. A polymeric contact lens comprising the polymer resulting from the copolymerization of the following:

Phenyltetramethyldisilylmethylene-
10 methacrylate 5 to 95 weight %
Methacrylic acid 5 to 60 weight %.

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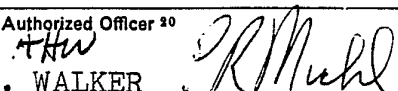
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INTERNATIONAL SEARCH REPORT

International Application No PCT/US85/01522

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) ³ According to International Patent Classification (IPC) or to both National Classification and IPC INT. CL. 4 C08L 83/00 U.S. CL. 523/107; 526/258, 264, 279, 281, 307.7		
II. FIELDS SEARCHED		
Minimum Documentation Searched ⁴		
Classification System	Classification Symbols	
U.S.	523/107; 526/258, 264, 279, 281, 307.7	
Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched ⁵		
III. DOCUMENTS CONSIDERED TO BE RELEVANT ¹⁴		
Category *	Citation of Document, ¹⁶ with indication, where appropriate, of the relevant passages ¹⁷	Relevant to Claim No. ¹⁸
A	US, A, 2,958,681 PUBLISHED 01 NOVEMBER 1960 CAMPBELL	1-26
A	US, A, RE 31406 PUBLISHED 04 OCTOBER 1983 GAYLORD	1-26
A	US, A, 4,463,149 PUBLISHED 31 JULY 1984 ELLIS	1-26
A, P	US, A, 4,507,452 PUBLISHED 26 MARCH 1985 FOLEY	1-26
<div style="display: flex; justify-content: space-between;"> <div style="width: 45%;"> <p>* Special categories of cited documents: ¹⁵</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document 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> </div> <div style="width: 45%;"> <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</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>"&" document member of the same patent family</p> </div> </div>		
IV. CERTIFICATION		
Date of the Actual Completion of the International Search ³		Date of Mailing of this International Search Report ³
25 SEPTEMBER 1985		07 OCT 1985
International Searching Authority ¹		Signature of Authorized Officer ²⁰
ISA/US		 A.H. WALKER