

Method for Making Photochromic Contact Lenses

This invention is related to a method for producing photochromic contact lenses, in particular, preferably photochromic silicone hydrogel contact lenses, capable of undergoing a reversible color change upon exposure to UV-irradiation or high-energy-violet light (HEVL) (with wavelengths from 380 nm to 440 nm). This invention also provides photochromic contact lenses or more preferably photochromic silicone hydrogel contact lenses made according to a method of the invention.

BACKGROUND

Conventionally, UV-absorbing compounds have been incorporated into contact lenses to make UV-absorbing contact lenses to protect eyes from UV-radiations. Recently, there have been proposed various photochromic contact lenses for protecting eyes from UV-radiations (see, e.g., U.S. Pat. Nos. 4681412, 6017121, 6174464, 6224945, 7261844, 7368072, 7556750, 7560056, 7584630, 7641337, 7999989, 8158037, 8697770, 8741188, 9052438, 9097916, 9377569, 9465234, 9904074, 10197707). Those proposed photochromic contact lenses contains photochromic compounds which are capable of undergoing a reversible color change, e.g., from "clear" to blue or other color, upon exposure to UV/HEVL irradiation.

Typically, photochromic contact lenses can be obtained by adding a photochromic compound in a polymerizable composition (i.e., lens formulation) for forming the contact lenses and then by curing the polymerizable composition in contact lens molds according to a cast-molding lens manufacturing process. A conventional cast-molding lens manufacturing process comprises: curing (polymerizing) of a polymerizable composition (lens formulation) within disposable plastic molds typically consisting of two single-use mold halves; opening molds; optionally but preferably delensing (i.e., removing cast-molded lenses from the molds); and being subjected to various post-molding processes including extraction, hydration, packaging, and sterilization, etc.

However, the Lightstream Technology™ (Alcon) has not been applied to make photochromic contact lenses from a lens-forming composition (i.e., a polymerizable composition comprising several polymerizable components). The Lightstream Technology™ involves (1) a lens-forming (i.e., polymerizable) composition (which is a fluid at room temperature), (2) reusable molds produced in high precision, and (3) curing under a spatial limitation of actinic radiation (e.g., UV) for a very short period of time (e.g., less than about 30 seconds). Lenses produced according to the Lightstream Technology™ can have high consistency and high fidelity to the original lens design, because of use of reusable, high precision molds. In addition, contact lenses with high quality can be produced at relatively lower cost due to the short curing time and a high production yield.

Therefore, there is still a need for a method for making photochromic contact lenses (preferably photochromic silicone hydrogel contact lenses) from a polymerizable composition according to the Lightstream Technology™.

SUMMARY

In one aspect, the invention provides a method for producing photochromic contact lenses, the method comprising the steps of: (1) providing a mold for making a contact lens, wherein the mold has a first mold half with a first molding surface defining the anterior surface of a contact lens and a second mold half with a second molding surface defining the posterior surface of the contact lens, wherein said first and second mold halves are configured to receive each other such that a cavity is formed between said first and second molding surfaces; (2) introducing a polymerizable composition into the cavity, wherein the polymerizable composition comprises (a) at least one hydrophilic (meth)acrylamido monomer, (b) at least one siloxane-containing (meth)acrylamido monomer, (c) at least one polysiloxane vinylic crosslinker, (d) at least one photochromic compound, and (e) from about 0.05% to about 1.5% by weight of at least one an acyl germanium photoinitiator capable of initiating a free-radical polymerization under irradiation with a light source including a light in the region of about 420 to about 500 nm, wherein the sum of the amounts of components (a) to (e) is at least 90% by weight relative to total amount of all polymerizable components in the polymerizable composition, wherein the polymerizable composition has a curing time of about 80 seconds or less as determined in photo-rheology study using a LED light source which provide a visible light with a spectral centroid of about 452nm and a total irradiance of about 54 mW/cm²; and (3) irradiating (preferably under a spatial limitation of actinic radiation) the polymerizable composition in the mold for about 80 seconds or less with a visible light in a region of from 420 to 500 nm, so as to form the photochromic silicone hydrogel contact lens, wherein the formed photochromic silicone hydrogel contact lens comprises an anterior surface defined by the first molding surface and an opposite posterior surface defined by the second molding surface and is capable of undergoing reversible change from a colorless or light-colored state to a colored state upon exposure to UV or HEVL irradiation.

In another aspect, the invention provide a polymerizable composition suitable for actinically cast-molding photochromic silicone hydrogel contact lenses (preferably under a spatial limitation of actinic radiation). The polymerizable composition is a fluid composition that comprises (a) at least one hydrophilic (meth)acrylamido monomer, (b) at least one siloxane-containing (meth)acrylamido monomer, (c) at least one polysiloxane vinylic crosslinker, (d) at least one photochromic compound, and (e) from about 0.05% to about 1.5% by weight of at least one an acyl germanium photoinitiator capable of initiating a free-

radical polymerization under irradiation with a light source including a light in the region of about 420 to about 500 nm, wherein the sum of the amounts of components (a) to (e) is at least 90% by weight relative to total amount of all polymerizable components in the polymerizable composition, wherein the polymerizable composition has a curing time of about 80 seconds or less as determined in photo-rheology study using a LED light source which provide a visible light with a spectral centroid of about 452 nm and a total irradiance of about 54 mW/cm².

The invention provides in a further aspect photochromic contact lenses obtained according to a method of the invention.

BRIEF DESCRIPTIONS OF THE DRAWINGS

Figure 1 shows the UV-Vis absorption spectra of a spiropyran photochromic dye, 2-(3',3'-Dimethyl-6-nitrospiro[chromene-2,2'-indolin]-1'-yl)ethyl methacrylate (20 ppm) in 1-propanol.

Figure 2 shows the UV-Vis absorption spectra of a naphthopyran photochromic dye, 3,3-Diphenyl-3H-naphtho[2,1-b]pyran (20 ppm) in 1-propanol.

Figure 3 shows the photo-rheology studies of various lens formulations (Control, SP1_{0.5%} and NP1_{0.5%}).

Figure 4 shows the UV-Vis transmission spectra of a photochromic contact lens (SP1_{0.5%}) at "on" and "off" states.

Figure 5 shows the fading kinetics studies at room temperature (absorption at 550nm) of a photochromic contact lens (SP1_{0.5%}).

Figure 6 shows the photo-rheology studies of various lens formulations (1 – Control; 2 – Ge-PC_{0.2%}, 1st run; and 3 – Ge-PC_{0.2%}, 2nd run).

Figure 7 shows the UV/visible absorption spectra of a photochromic silicone hydrogel contact lens made from lens formulation Ge-PC_{0.2%} at room temperature as function of time immediately after UV activation.

Figure 8 shows the fading kinetics of a photochromic silicone hydrogel contact lens made from lens formulation Ge-PC_{0.2%} at room temperature.

Figure 9 shows photo fatigue test of a photochromic silicone hydrogel contact lens made from lens formulation Ge-PC_{0.2%} for 10 cycles at room temperature.

DETAILED DESCRIPTION

Unless defined otherwise, all technical and scientific terms used herein have the same meaning as commonly understood by one of ordinary skill in the art to which this invention belongs. Generally, the nomenclature used herein and the laboratory procedures are well known and commonly employed in the art. Conventional methods are used for

these procedures, such as those provided in the art and various general references. Where a term is provided in the singular, the inventors also contemplate the plural of that term. The nomenclature used herein and the laboratory procedures described below are those well known and commonly employed in the art.

"About" as used herein means that a number referred to as "about" comprises the recited number plus or minus 1-10% of that recited number.

"Optional" or "optionally" means that the subsequently described event or circumstance can or cannot occur, and that the description includes instances where the event or circumstance occurs and instances where it does not.

An "ophthalmic device", as used herein, refers to a contact lens (hard or soft), an intraocular lens, a corneal onlay, other ophthalmic devices (e.g., stents, glaucoma shunt, or the like) used on or about the eye or ocular vicinity.

"Contact Lens" refers to a structure that can be placed on or within a wearer's eye. A contact lens can correct, improve, or alter a user's eyesight, but that need not be the case. A contact lens can be of any appropriate material known in the art or later developed, and can be a soft lens, a hard lens, or a hybrid lens. A "silicone hydrogel contact lens" or "SiHy contact lens" refers to a contact lens comprising a silicone hydrogel material.

A "hydrogel" or "hydrogel material" refers to a crosslinked polymeric material which has three-dimensional polymer networks (i.e., polymer matrix), is insoluble in water, but can hold at least 10% by weight of water in its polymer matrix when it is fully hydrated (or equilibrated).

A "silicone hydrogel" or "SiHy" refers to a silicone-containing hydrogel obtained by copolymerization of a polymerizable composition comprising at least one silicone-containing monomer or at least one silicone-containing macromer or at least one crosslinkable silicone-containing prepolymer.

As used in this application, the term "non-silicone hydrogel" refers to a hydrogel that is theoretically free of silicon.

"Hydrophilic," as used herein, describes a material or portion thereof that will more readily associate with water than with lipids.

A "vinyl monomer" refers to a compound that has one sole ethylenically unsaturated group, is soluble in a solvent, and can be polymerized actinically or thermally.

The term "soluble", in reference to a compound or material in a solvent, means that the compound or material can be dissolved in the solvent to give a solution with a concentration of at least about 0.5% by weight at room temperature (i.e., a temperature of from 21°C to 27°C).

The term "insoluble", in reference to a compound or material in a solvent, means that the compound or material can be dissolved in the solvent to give a solution with a

concentration of less than 0.01% by weight at room temperature (as defined above).

The term “ethylenically unsaturated group” is employed herein in a broad sense and is intended to encompass any groups containing at least one $>C=C<$ group. Exemplary

ethylenically unsaturated groups include without limitation (meth)acryloyl ($\text{—}\overset{\text{O}}{\underset{\text{||}}{\text{C}}}\overset{\text{CH}_3}{\text{—}}\text{C}=\text{CH}_2$ and/or $\text{—}\overset{\text{O}}{\underset{\text{||}}{\text{C}}}\text{—CH}=\text{CH}_2$), allyl, vinyl, styrenyl, or other $C=C$ containing groups.

As used herein, “actinically” in reference to curing, crosslinking or polymerizing of a polymerizable composition, a prepolymer or a material means that the curing (e.g., crosslinked and/or polymerized) is performed by actinic irradiation, e.g., UV/visible light irradiation, or the like. Thermal curing or actinic curing methods are well-known to a person skilled in the art.

An “acrylic monomer” refers to a vinylic monomer having one sole (meth)acryloyl group. Examples of acrylic monomers includes (meth)acryloxy [or(meth)acryloyloxy] monomers and (meth)acrylamido monomers.

An “(meth)acryloxy monomer” or “(meth)acryloyloxy monomer” refers to a vinylic monomer having one sole group of $\text{—O—}\overset{\text{O}}{\underset{\text{||}}{\text{C}}}\overset{\text{CH}_3}{\text{—}}\text{C}=\text{CH}_2$ or $\text{—O—}\overset{\text{O}}{\underset{\text{||}}{\text{C}}}\text{—CH}=\text{CH}_2$.

An “(meth)acrylamido monomer” refers to a vinylic monomer having one sole group of $\text{—NR}^0\text{—}\overset{\text{O}}{\underset{\text{||}}{\text{C}}}\overset{\text{CH}_3}{\text{—}}\text{C}=\text{CH}_2$ or $\text{—NR}^0\text{—}\overset{\text{O}}{\underset{\text{||}}{\text{C}}}\text{—CH}=\text{CH}_2$ in which R^0 is H or $C_1\text{—}C_4$ alkyl.

The term “(meth)acrylamide” refers to methacrylamide and/or acrylamide.

The term “(meth)acrylate” refers to methacrylate and/or acrylate.

An “N-vinyl amide monomer” refers to an amide compound having a vinyl group ($\text{—CH}=\text{CH}_2$) that is directly attached to the nitrogen atom of the amide group.

The term “ene group” refers to a monovalent radical of $\text{CH}_2=\text{CH—}$ or $\text{CH}_2=\text{CCH}_3\text{—}$ that is not covalently attached to an oxygen or nitrogen atom or a carbonyl group.

An “ene monomer” refers to a vinylic monomer having one sole ene group.

A “hydrophilic vinylic monomer”, a “hydrophilic acrylic monomer”, a “hydrophilic (meth)acryloxy monomer”, or a “hydrophilic (meth)acrylamido monomer”, as used herein, respectively refers to a vinylic monomer, an acrylic monomer, a (meth)acryloxy monomer, or a (meth)acrylamido monomer, which typically yields a homopolymer that is water-soluble or can absorb at least 10 percent by weight of water.

A “hydrophobic vinylic monomer”, a “hydrophobic acrylic monomer”, a “hydrophobic (meth)acryloxy monomer”, or a “hydrophobic (meth)acrylamido monomer”, as used herein, respectively refers to a vinylic monomer, an acrylic monomer, a (meth)acryloxy monomer, or a (meth)acrylamido monomer, which typically yields a homopolymer that is insoluble in water and can absorb less than 10% by weight of water.

As used in this application, the term “vinyl crosslinker” refers to an organic compound having at least two ethylenically unsaturated groups. A “vinyl crosslinking agent” refers to a vinyl crosslinker having a molecular weight of 700 Daltons or less.

A siloxane, which often also described as a silicone, refers to a molecule having at least one moiety of $-\text{Si}-\text{O}-\text{Si}-$ where each Si atom carries two organic groups as substituents.

A “polysiloxane segment” or “polydiorganosiloxane segment” interchangeably refers

to a polymer chain segment (i.e., a divalent radical) of $\left[\begin{array}{c} \text{R}_{\text{S1}} \\ | \\ \text{---Si---O---} \\ | \\ \text{R}_{\text{S2}} \end{array} \right]_{\text{SN}} \begin{array}{c} \text{R}_{\text{S1}} \\ | \\ \text{---Si---} \\ | \\ \text{R}_{\text{S2}} \end{array}$ in which SN is an integer of 3 or larger and each of R_{S1} and R_{S2} independent of one another are selected from the group consisting of: $\text{C}_1\text{--C}_{10}$ alkyl; phenyl; $\text{C}_1\text{--C}_4\text{--alkyl-substituted phenyl}$; $\text{C}_1\text{--C}_4\text{--alkoxy-substituted phenyl}$; phenyl- $\text{C}_1\text{--C}_6\text{--alkyl}$; $\text{C}_1\text{--C}_{10}$ fluoroalkyl; $\text{C}_1\text{--C}_{10}$ fluoroether; aryl; aryl $\text{C}_1\text{--C}_{18}$ alkyl; $-\text{alk}-(\text{OC}_2\text{H}_4)_{\gamma 1}-\text{OR}^0$ (in which alk is $\text{C}_1\text{--C}_6$ alkylene diradical, R^0 is H or $\text{C}_1\text{--C}_4$ alkyl and $\gamma 1$ is an integer from 1 to 10); a $\text{C}_2\text{--C}_{40}$ organic radical having at least one functional group selected from the group consisting of hydroxyl group ($-\text{OH}$), carboxyl group ($-\text{COOH}$), amino group ($-\text{NR}_{\text{N1}}\text{R}_{\text{N1}}'$), amino linkages of $-\text{NR}_{\text{N1}}-$, amide linkages of $-\text{CONR}_{\text{N1}}-$, amide of $-\text{CONR}_{\text{N1}}\text{R}_{\text{N1}}'$, urethane linkages of $-\text{OCONH}-$, and $\text{C}_1\text{--C}_4$ alkoxy group, or a linear hydrophilic polymer chain, in which R_{N1} and R_{N1}' independent of each other are hydrogen or a $\text{C}_1\text{--C}_{15}$ alkyl; and a photochromic organic radical having a photochromic group.

A “polydiorganosiloxane vinyl crosslinker” or “polysiloxane vinyl crosslinker” interchangeably refers to a compound comprising at least one polysiloxane segment and at least two ethylenically-unsaturated groups.

A “linear polydiorganosiloxane vinyl crosslinker” or “linear polysiloxane vinyl crosslinker” interchangeably refers to a compound comprising a main chain which includes at least one polysiloxane segment and is terminated with one ethylenically-unsaturated group at each of the two ends of the main chain.

A “chain-extended polydiorganosiloxane vinyl crosslinker” or “chain-extended polysiloxane vinyl crosslinker” interchangeably refers to a compound comprising at least two ethylenically-unsaturated groups and at least two polysiloxane segments each pair of which are linked by one divalent radical.

The term “photochromic compound” refers to a compound that has one colorless (or light-colored) form and one colored form and can undergo reversible change from the colorless form (or light-colored form) (or so-called “deactivated form” to the colored form (or so-called “activated form”) upon exposure to UV or HEVL irradiation.

The term “colorless or light-colored stated” or “inactivated state” in reference to a photochromic contact lens means the original state of the photochromic contact lens before

the photochromic contact lens is irradiated with UV and/or HEVL. In this state, the photochromic contact lens typically is colorless or shows a faint color as observed by a naked eye.

The term “colored stated” or “activated state” in reference to a photochromic contact lens means a state of the photochromic contact lens when the photochromic contact lens is being irradiated with UV and/or HEVL. In this state, the photochromic contact lens typically shows a dark color as observed by a naked eye.

The term “fluid” as used herein indicates that a material is capable of flowing like a liquid.

As used in this application, the term “clear” in reference to a polymerizable composition means that the polymerizable composition is a transparent solution or liquid mixture having a light transmissibility of 85% or greater (preferably 90% or greater) in the range between 400 to 700 nm.

A free radical initiator can be either a photoinitiator or a thermal initiator. A “photoinitiator” refers to a chemical that initiates free radical crosslinking/polymerizing reaction by the use of UV and/or visible light. A “thermal initiator” refers to a chemical that initiates radical crosslinking/polymerizing reaction by the use of heat energy.

The term “acyl germanium photoinitiator” refers to an organogermanium compound that is a germanium-based Norrish Type I photoinitiator and comprises at least one acrylcarbonyl group connected to germanium. Examples of such acyl germanium photoinitiators are described in U.S. Pat. Nos. 7,605,190 and 10,324,311.

The term “curing time” in reference to a polymerizable composition means a time required for the measured elastic modulus (G') to reach a plateau in the photo-rheology study in which the polymerizable composition is placed between a quartz plate that allows actinic radiation (e.g., a visible light with a spectral centroid of about 452nm and a total irradiance of about 54 mW/cm²) to pass through and a rheometer.

As used in this application, the term “polymer” means a material formed by polymerizing/crosslinking one or more monomers or macromers or prepolymers or combinations thereof.

A “macromer” or “prepolymer” refers to a compound or polymer that contains ethylenically unsaturated groups and has a number average molecular weight of greater than 700 Daltons.

As used in this application, the term “molecular weight” of a polymeric material (including monomeric or macromeric materials) refers to the number-average molecular weight unless otherwise specifically noted or unless testing conditions indicate otherwise. A skilled person knows how to determine the molecular weight of a polymer according to known methods, e.g., GPC (gel permeation chromatography) with one or more of a

refractive index detector, a low-angle laser light scattering detector, a multi-angle laser light scattering detector, a differential viscometry detector, a UV detector, and an infrared (IR) detector; MALDI-TOF MS (matrix-assisted desorption/ionization time-of-flight mass spectroscopy); ^1H NMR (Proton nuclear magnetic resonance) spectroscopy, etc.

The term "monovalent radical" refers to an organic radical that is obtained by removing a hydrogen atom from an organic compound and that forms one bond with one other group in an organic compound. Examples include without limitation, alkyl (by removal of a hydrogen atom from an alkane), alkoxy (or alkoxyl) (by removal of one hydrogen atom from the hydroxyl group of an alkyl alcohol), thiyl (by removal of one hydrogen atom from the thiol group of an alkylthiol), cycloalkyl (by removal of a hydrogen atom from a cycloalkane), cycloheteroalkyl (by removal of a hydrogen atom from a cycloheteroalkane), aryl (by removal of a hydrogen atom from an aromatic ring of the aromatic hydrocarbon), heteroaryl (by removal of a hydrogen atom from any ring atom), amino (by removal of one hydrogen atom from an amine), etc.

The term "divalent radical" refers to an organic radical that is obtained by removing two hydrogen atoms from an organic compound and that forms two bonds with other two groups in an organic compound. For example, an alkylene divalent radical (i.e., alkylenyl) is obtained by removal of two hydrogen atoms from an alkane, a cycloalkylene divalent radical (i.e., cycloalkylenyl) is obtained by removal of two hydrogen atoms from the cyclic ring.

In this application, the term "substituted" in reference to an alkyl or an alkylenyl means that the alkyl or the alkylenyl comprises at least one substituent which replaces one hydrogen atom of the alkyl or the alkylenyl and is selected from the group consisting of hydroxyl (-OH), carboxyl (-COOH), -NH₂, sulfhydryl (-SH), C₁-C₄ alkyl, C₁-C₄ alkoxy, C₁-C₄ alkylthio (alkyl sulfide), C₁-C₄ acylamino, C₁-C₄ alkylamino, di-C₁-C₄ alkylamino, and combinations thereof.

The term "terminal ethylenically-unsaturated group" refers to one ethylenically-unsaturated group at one of the two ends of the main chain (or backbone) of an organic compound as known to a person skilled in the art.

A "blending vinylic monomer" refers to a vinylic monomer capable of dissolving both hydrophilic and hydrophobic components of a polymerizable composition to form a solution.

"Post-curing surface treatment", in reference to a SiHy lens bulk material or a SiHy contact lens, means a surface treatment process that is performed after the SiHy lens bulk material or the SiHy contact lens is formed by curing (i.e., thermally or actinically polymerizing) a SiHy lens formulation.

The term "silicone hydrogel lens formulation" or "SiHy lens formulation" interchangeably refers to a polymerizable composition that comprises all necessary polymerizable components for producing a SiHy contact lens or a SiHy lens bulk material as

well known to a person skilled in the art.

A "UV-absorbing vinylic monomer" refers to a compound comprising an ethylenically-unsaturated group and a UV-absorbing moiety which can absorb or screen out UV radiation in the range from 200 nm to 400 nm as understood by a person skilled in the art.

A "HEVL-absorbing vinylic monomer" refers to a compound comprising an ethylenically-unsaturated group and a HEVL-absorbing moiety which can absorb or screen out HEVL (high-energy-violet-light) radiation in the range from 380 nm to 440 nm as understood by a person skilled in the art.

"UVA" refers to radiation occurring at wavelengths between 315 and 380 nanometers; "UVB" refers to radiation occurring between 280 and 315 nanometers; "Violet" refers to radiation occurring at wavelengths between 380 and 440 nanometers.

"UVA transmittance" (or "UVA %T"), "UVB transmittance" or "UVB %T", and "violet-transmittance" or "Violet %T" are calculated by the following formula.

$$\text{UVA \%T} = \text{Average \% Transmission between 315 nm and 380 nm} \times 100$$

$$\text{UVB \%T} = \text{Average \% Transmission between 280 nm and 315 nm} \times 100$$

$$\text{Violet \%T} = \text{Average \% Transmission between 380 nm and 440 nm} \times 100$$

A "spatial limitation of actinic radiation" refers to an act or process in which energy radiation in the form of rays is directed by, for example, a mask or screen or combinations thereof, to impinge, in a spatially restricted manner, onto an area having a well defined peripheral boundary. A spatial limitation of UV radiation is obtained by using a mask or screen having a radiation (e.g., UV and/or visible light) permeable region, a radiation (e.g., UV and/or visible light) impermeable region surrounding the radiation-permeable region, and a projection contour which is the boundary between the radiation-impermeable and radiation-permeable regions, as schematically illustrated in the drawings of U.S. Pat. Nos. 6,800,225 (Figs. 1-11), 6,627,124 (Figs. 1-9), 7,384,590 (Figs. 1-6), and 7,387,759 (Figs. 1-6). The mask or screen allows to spatially project a beam of radiation (e.g., UV radiation and/or visible radiation) having a cross-sectional profile defined by the projection contour of the mask or screen. The projected beam of radiation (e.g., UV radiation and/or visible radiation) limits radiation impinging on a lens formulation located in the path of the projected beam from the first molding surface to the second molding surface of a mold. The resultant contact lens comprises an anterior surface defined by the first molding surface, an opposite posterior surface defined by the second molding surface, and a lens edge defined by the sectional profile of the projected UV and/or visible beam (i.e., a spatial limitation of radiation). The radiation used for the crosslinking is radiation energy, especially UV radiation (and/or visible radiation), gamma radiation, electron radiation or thermal radiation, the radiation energy preferably being in the form of a substantially parallel beam in order on the one hand to achieve good restriction and on the other hand efficient use of the energy.

The intrinsic "oxygen permeability", Dk_i , of a material is the rate at which oxygen will pass through a material. As used in this application, the term "oxygen permeability (Dk)" in reference to a hydrogel (silicone or non-silicone) or a contact lens means a corrected oxygen permeability (Dk_c) which is measured at about 34-35°C and corrected for the surface resistance to oxygen flux caused by the boundary layer effect according to the procedures described in ISO 18369-4. Oxygen permeability is conventionally expressed in units of barrers, where "barrer" is defined as $[(\text{cm}^3 \text{ oxygen})(\text{cm}) / (\text{cm}^2)(\text{sec})(\text{mm Hg})] \times 10^{-9}$.

The "oxygen transmissibility", Dk/t , of a lens or material is the rate at which oxygen will pass through a specific lens or material with an average thickness of t [in units of mm] over the area being measured. Oxygen transmissibility is conventionally expressed in units of barrers/mm, where "barrers/mm" is defined as $[(\text{cm}^3 \text{ oxygen})/(\text{cm}^2)(\text{sec})(\text{mm Hg})] \times 10^{-9}$.

The term "modulus" or "elastic modulus" in reference to a contact lens or a material means the tensile modulus or Young's modulus which is a measure of the stiffness of a contact lens or a material in tension. A person skilled in the art knows well how to determine the elastic modulus of a SiHy material or a contact lens. For example, all commercial contact lenses have reported values of elastic modulus.

A "coating" in reference to a contact lens means that the contact lens has, on its surfaces, a thin layer of a material that is different from the bulk material of the contact lens and obtained by subjecting the contact lens to a surface treatment.

"Surface modification" or "surface treatment", as used herein, means that an article has been treated in a surface treatment process, in which (1) a coating is applied to the surface of the article, (2) chemical species are adsorbed onto the surface of the article, (3) the chemical nature (e.g., electrostatic charge) of chemical groups on the surface of the article are altered, or (4) the surface properties of the article are otherwise modified. Exemplary surface treatment processes include, but are not limited to, a surface treatment by energy (e.g., a plasma, a static electrical charge, irradiation, or other energy source), chemical treatments, the grafting of hydrophilic vinylic monomers or macromers onto the surface of an article, mold-transfer coating process disclosed in U.S. Pat. No. 6719929, the incorporation of wetting agents into a lens formulation for making contact lenses proposed in U.S. Pat. Nos. 6367929 and 6822016, reinforced mold-transfer coating disclosed in U.S. Pat. No. 7858000, and a hydrophilic coating composed of covalent attachment or physical deposition of one or more layers of one or more hydrophilic polymer onto the surface of a contact lens disclosed in U.S. Pat. Nos. 8147897, 8409599, 8557334, 8529057, and 9505184.

A "hydrophilic surface" in reference to a SiHy material or a contact lens means that the SiHy material or the contact lens has a surface hydrophilicity characterized by having an averaged water contact angle of about 90 degrees or less, preferably about 80 degrees or

less, more preferably about 70 degrees or less, more preferably about 60 degrees or less.

An "average contact angle" refers to a water contact angle (static water contact angle measured by Sessile Drop), which is obtained by averaging measurements of at least 3 individual contact lenses.

In general, the invention is directed to a method for making photochromic contact lenses, in particular, SiHy contact lenses, from a polymerizable composition based on the Lightstream Technology™. The invention is partly based on the discovery that by selecting the combination of photochromic compound, acylgermanium photoinitiator, and (meth)acrylamide-type monomers and/or macromers in preparing a polymerizable composition for making photochromic SiHy contact lenses as well as a visible radiation as curing radiation source, the curing time of the polymerizable composition in a mold can be shortened so as to make the polymerizable composition suitable for making photochromic SiHy contact lenses based on the Lightstream Technology™.

The relatively-short curing time involved in the invention can ensure that contact lenses with high quality lens edge are produced at a lower cost. It is believed that a relatively longer curing time may adversely affect the quality of lens edge of contact lenses made according to the Lightstream Technology™ and can increase the product cost by lowering the production output. It is also believed that a relatively longer curing time may cause permanent damage to a photochromic compound because a photochromic compound is susceptible to degradation by free radicals upon its relatively long exposure to free radicals (irradiation). Although one might be able to shorten the curing time of a mixture by increasing the curing light intensity and/or the concentration of a photoinitiator in the mixture, the high curing light intensity and high photoinitiator concentration have limits and may not be sufficient to reduce the curing time enough for the Lightstream Technology™. Moreover, the high curing light intensity and high photoinitiator concentration may cause degradation of the photochromic compound. These measures may cause resultant lenses with undesirable physical properties (such as, for example, fragile and high content of extractable due to unpolymerized monomers) and with insufficient photoactivities. In addition, ineffective or uneven photo-polymerization of the polymerizable composition due to the presence of a photochromic compound may be minimized or eliminated. With a polymerizable composition of the invention, a curing time of less than about 30 seconds, for example, can be achieved with a LED light source (e.g., a LED light source with a spectral centroid of 452nm and a total irradiance of 54 mW/cm²). Photochromic SiHy contact lenses made from such a polymerizable composition according to the Lightstream Technology can have high lens edge quality and relatively low lens production cost because of the relatively short curing time.

In one aspect, the invention provides a method for producing photochromic contact

lenses, the method comprising the steps of: (1) obtaining a mold, wherein the mold has a first mold half with a first molding surface defining the anterior surface of a contact lens and a second mold half with a second molding surface defining the posterior surface of the contact lens, wherein said first and second mold halves are configured to receive each other such that a cavity is formed between said first and second molding surfaces; (2) introducing a polymerizable composition into the cavity, wherein the polymerizable composition comprises (a) at least one hydrophilic (meth)acrylamido monomer, (b) at least one siloxane-containing (meth)acrylamido monomer, (c) at least one polysiloxane vinylic crosslinker (with two or more ethylenically-unsaturated groups), (d) at least one photochromic compound, preferably in an amount from about 0.005% to about 1.0% by weight (preferably from about 0.01% to about 0.8% by weight, more preferably from about 0.02% to about 0.6% by weight, even more preferably from about 0.05% to about 0.5% by weight), (relative to the total weight of the polymerizable composition), and (e) from about 0.05% to about 1.5% by weight of at least one an acyl germanium photoinitiator relative to the total weight of the polymerizable composition, wherein the acyl germanium photoinitiator is capable of initiating a free-radical polymerization under irradiation with a light source including a light in the region of about 420 to about 500 nm, wherein the sum of the amounts of components (a) to (e) is at least 90% by weight (preferably at least 93% by weight, more preferably at least 96% by weight, even more preferably at least 98% by weight) relative to total amount of all polymerizable components in the polymerizable composition, wherein the polymerizable composition has a curing time of about 80 seconds or less (preferably about 65 seconds or less, more preferably about 50 seconds or less, even more preferably about 35 seconds or less) as determined in photo-rheology study using a LED light source which provide a visible light with a spectral centroid of about 452nm and a total irradiance of about 54 mW/cm²; and (3) irradiating (preferably under a spatial limitation of actinic radiation) the polymerizable composition in the mold for about 80 seconds or less (preferably about 65 seconds or less, more preferably about 50 seconds or less, even more preferably about 35 seconds or less) with a visible light in a region of from 420 to 500 nm, so as to form the photochromic SiHy contact lens, wherein the formed photochromic SiHy contact lens comprises an anterior surface defined by the first molding surface and an opposite posterior surface defined by the second molding surface and is capable of undergoing reversible change from a colorless or light-colored state to a colored state upon exposure to UV or HEVL irradiation.

In another aspect, the invention provides a polymerizable composition for actinically cast-molding photochromic SiHy contact lenses (preferably under a spatial limitation of actinic radiation). The polymerizable composition is a fluid composition which comprises (a) at least one hydrophilic (meth)acrylamido monomer, (b) at least one siloxane-containing (meth)acrylamido monomer, (c) at least one polysiloxane vinylic crosslinker (with two or

more ethylenically-unsaturated groups), (d) at least one photochromic compound, preferably in an amount from about 0.005% to about 1.0% by weight (preferably from about 0.01% to about 0.8% by weight, more preferably from about 0.02% to about 0.6% by weight, even more preferably from about 0.05% to about 0.5% by weight), (relative to the total weight of the polymerizable composition), and (e) from about 0.05% to about 1.5% by weight of at least one an acyl germanium photoinitiator relative to the total weight of the polymerizable composition, wherein the acyl germanium photoinitiator is capable of initiating a free-radical polymerization under irradiation with a light source including a light in the region of about 420 to about 500 nm, wherein the sum of the amounts of components (a) to (e) is at least 60% by weight (preferably at least 65% by weight, more preferably at least 70% by weight, even more preferably at least 75% by weight) relative to total amount of all polymerizable components in the polymerizable composition, wherein the polymerizable composition has a curing time of about 80 seconds or less (preferably about 65 seconds or less, more preferably about 50 seconds or less, even more preferably about 35 seconds or less) as determined in photo-rheology study using a LED light source which provide a visible light with a spectral centroid of about 452nm and a total irradiance of about 54 mW/cm².

Lens molds for making contact lenses including SiHy contact lenses are well known to a person skilled in the art and, for example, are employed in cast molding or spin casting. For example, a mold (for cast molding) generally comprises at least two mold sections (or portions) or mold halves, i.e. first and second mold halves. The first mold half defines a first molding (or optical) surface and the second mold half defines a second molding (or optical) surface. The first and second mold halves are configured to receive each other such that a lens forming cavity is formed between the first molding surface and the second molding surface. The molding surface of a mold half is the cavity-forming surface of the mold and in direct contact with the polymerizable composition.

Methods of manufacturing mold sections for cast-molding a contact lens are generally well known to those of ordinary skill in the art. The process of the present invention is not limited to any particular method of forming a mold. In fact, any method of forming a mold can be used in the present invention. The first and second mold halves can be formed through various techniques, such as injection molding or lathing. Examples of suitable processes for forming the mold halves are disclosed in U.S. Pat. Nos. 4444711; 4460534; 5843346; and 5894002.

Virtually all materials known in the art for making molds can be used to make molds for making contact lenses. For example, polymeric materials, such as polyethylene, polypropylene, polystyrene, PMMA, Topas® COC grade 8007-S10 (clear amorphous copolymer of ethylene and norbornene, from Ticona GmbH of Frankfurt, Germany and Summit, New Jersey), or the like can be used. Other materials that allow UV light

transmission could be used, such as quartz glass and sapphire.

Preferably, a reusable mold suitable for spatial limitation of radiation is used in the invention, the projected beam of radiation (e.g., radiation from the light source including the light in the region of 420 nm to 550 nm) limits radiation (e.g., UV or visible light radiation) impinging on the polymerizable composition located in the path of the projected beam from the first molding surface to the second molding surface of the reusable mold. The resultant contact lens comprises an anterior surface defined by the first molding surface, an opposite posterior surface defined by the second molding surface, and a lens edge (with sharp edge and high quality) defined by the sectional profile of the projected radiation beam (i.e., a spatial limitation of radiation). Examples of reusable molds suitable for spatial limitation of radiation include without limitation those disclosed in U.S. Pat. Nos. 6627124, 6800225, 7384590, and 7387759.

For example, a preferred reusable mold comprises a first mold half having a first molding surface and a second mold half having a second molding surface. The two mold halves of the preferred reusable mold are not touching each other, but there is a thin gap of annular design arranged between the two mold halves. The gap is connected to the mold cavity formed between the first and second molding surfaces, so that excess mixture can flow into the gap. It is understood that gaps with any design can be used in the invention.

In a preferred embodiment, at least one of the first and second molding surfaces is permeable to a crosslinking radiation. More preferably, one of the first and second molding surfaces is permeable to a crosslinking radiation while the other molding surface is poorly permeable to the crosslinking radiation.

The reusable mold preferably comprises a mask which is fixed, constructed or arranged in, at or on the mold half having the radiation-permeable molding surface. The mask is impermeable or at least of poor permeability compared with the permeability of the radiation-permeable molding surface. The mask extends inwardly right up to the mold cavity and surrounds the mold cavity so as to screen all areas behind the mask with the exception of the mold cavity.

The mask may preferably be a thin chromium layer, which can be produced according to processes as known, for example, in photo and UV/visible light lithography. Other metals or metal oxides may also be suitable mask materials. The mask can also be coated with a protective layer, for example of silicon dioxide if the material used for the mold or mold half is quartz.

Alternatively, the mask can be a masking collar made of a material comprising a UV/visible light-absorber and substantially blocks curing radiation therethrough as described in U.S. Pat. No. 7387759. In this preferred embodiment, the mold half with the mask comprises a generally circular disc-shaped transmissive portion and a masking collar having

an inner diameter adapted to fit in close engagement with the transmissive portion, wherein said transmissive portion is made from an optically clear material and allows passage of curing energy therethrough, and wherein the masking collar is made from a material comprising a light-blocker and substantially blocks passage of curing energy therethrough, wherein the masking collar generally resembles a washer or a doughnut, with a center hole for receiving the transmissive portion, wherein the transmissive portion is pressed into the center opening of the masking collar and the masking collar is mounted within a bushing sleeve.

Reusable molds can be made of quartz, glass, sapphire, CaF_2 , a cyclic olefin copolymer (such as for example, Topas[®] COC grade 8007-S10 (clear amorphous copolymer of ethylene and norbornene) from Ticona GmbH of Frankfurt, Germany and Summit, New Jersey, Zeonex[®] and Zeonor[®] from Zeon Chemicals LP, Louisville, KY), polymethylmethacrylate (PMMA), polyoxymethylene from DuPont (Delrin), Uitem[®] (polyetherimide) from G.E. Plastics, PrimoSpire[®], etc.. Because of the reusability of the mold halves, a relatively high outlay can be expended at the time of their production in order to obtain molds of extremely high precision and reproducibility. Since the mold halves do not touch each other in the region of the lens to be produced, i.e. the cavity or actual molding surfaces, damage as a result of contact is ruled out. This ensures a high service life of the molds, which, in particular, also ensures high reproducibility of the contact lenses to be produced and high fidelity to the lens design.

In accordance with the invention, any hydrophilic (meth)acrylamido monomers can be in this invention. Examples of preferred hydrophilic (meth)acrylamido monomers are described later in this application. It is understood that any hydrophilic (meth)acrylamido monomers other than those specifically described later in this application can also be used in this invention.

Any siloxane-containing (meth)acrylamido monomers can be used in the invention. Preferably, siloxane-containing (meth)acrylamido monomers are those (meth)acrylamido monomers each of which comprises a bis(trialkylsilyloxy)alkylsilyl group, a tris(trialkylsilyloxy)-silyl group, or a polysiloxane chain having 2 to 30 siloxane units and terminated with an alkyl, hydroxyalkyl or methoxyalkyl group, as described later in this application. Such preferred siloxane-containing (meth)acrylamido monomers can be obtained from the commercial suppliers, or alternatively prepared according to known procedures, e.g., similar to those described in U.S. Pat. No. 9315669.

In accordance with the invention, any polysiloxane vinylic crosslinkers can be used in this invention. Examples of preferred polysiloxane vinylic crosslinkers include without limitation α,ω -(meth)acryloxy-terminated polydimethylsiloxanes of various molecular weight; α,ω -(meth)acrylamido-terminated polydimethylsiloxanes of various molecular weight; α,ω -

vinyl carbonate-terminated polydimethylsiloxanes of various molecular weight; α,ω -vinyl carbamate-terminated polydimethylsiloxane of various molecular weight; bis-3-methacryloxy-2-hydroxypropyloxypropyl polydimethylsiloxane of various molecular weight; N,N,N',N'-tetrakis(3-methacryloxy-2-hydroxypropyl)- α,ω -bis-3-aminopropyl-polydimethylsiloxane of various molecular weight; the reaction products of glycidyl methacrylate with amino-functional polydimethylsiloxanes; the reaction products of an azlactone-containing vinylic monomer (any one of those described above) with hydroxyl-functional polydimethylsiloxanes; polysiloxane-containing macromer selected from the group consisting of Macromer A, Macromer B, Macromer C, and Macromer D described in US 5,760,100; polysiloxane vinylic crosslinkers disclosed in U.S. Pat. Nos. 4136250, 4153641, 4182822, 4189546, 4259467, 4260725, 4261875, 4343927, 4254248, 4355147, 4276402, 4327203, 4341889, 4486577, 4543398, 4605712, 4661575, 4684538, 4703097, 4833218, 4837289, 4954586, 4954587, 5010141, 5034461, 5070170, 5079319, 5039761, 5346946, 5358995, 5387632, 5416132, 5449729, 5451617, 5486579, 5962548, 5981675, 6039913, 6762264, 7423074, 8163206, 8480227, 8529057, 8835525, 8993651, 9187601, 10081697, 10301451, and 10465047.

One class of preferred polysiloxane vinylic crosslinkers are di-(meth)acryloyloxy-terminated polysiloxane vinylic crosslinkers each having dimethylsiloxane units and hydrophilized siloxane units each having one methyl substituent and one monovalent C_4-C_{40} organic radical substituent having 2 to 6 hydroxyl groups, more preferably a polysiloxane vinylic crosslinker of formula (H), are described later in this application and can be prepared according to the procedures disclosed in U.S. Pat. No. 10081697.

Another class of preferred polysiloxane vinylic crosslinkers are vinylic crosslinkers each of which comprises one sole polydiorganosiloxane segment and two terminal (meth)acryloyl groups, which can be obtained from commercial suppliers; prepared by reacting glycidyl (meth)acrylate (meth)acryloyl chloride with a di-amino-terminated polydimethylsiloxane or a di-hydroxyl-terminated polydimethylsiloxane; prepared by reacting isocyanatoethyl (meth)acrylate with di-hydroxyl-terminated polydimethylsiloxanes prepared by reacting an amino-containing acrylic monomer with di-carboxyl-terminated polydimethylsiloxane in the presence of a coupling agent (a carbodiimide); prepared by reacting a carboxyl-containing acrylic monomer with di-amino-terminated polydimethylsiloxane in the presence of a coupling agent (a carbodiimide); or prepared by reacting a hydroxyl-containing acrylic monomer with a dihydroxy-terminated polydisiloxane in the presence of a diisocyanate or diepoxy coupling agent.

Other classes of preferred polysiloxane vinylic crosslinkers are chain-extended polysiloxane vinylic crosslinkers each of which has at least two polydiorganosiloxane segments linked by a linker between each pair of polydiorganosiloxane segments and two

terminal ethylenically unsaturated groups, which can be prepared according to the procedures described in U.S. Pat. Nos. 5034461, 5416132, 5449729, 5760100, 7423074, 8529057, 8835525, 8993651, 9187601, 10301451, and 10465047.

Any photochromic compounds can be used in the invention. Various photochromic compounds are disclosed in the patents and published patent applications and can be obtained from commercial sources or prepared by following the procedures described in the patents and literatures. Examples of preferred photochromic compounds include without limitation naphthopyrans, indeno-fused naphthopyrans (i.e., indeno-naphthopyrans), heterocyclic ring-fused naphthopyrans, benzopyrans, phenanthropyrans, quinopyrans, quinolinopyrans, fluoroanthenopyrans, anthracene-fused pyrans, tetracene-fused pyrans, spiro(benzindoline)naphthopyrans, spiro(indoline)naphthopyrans, spiro(indoline)benzopyrans, spiro(indoline)quinopyrans, spiro(indoline)pyrans, naphthoxazines, spirobenzopyrans, spirobenzothiopyrans, naphthacenediones, benzoxazines, spirooxazines, naphthoxazines, spiro(benzindoline)naphthoxazines, spiro(indoline)naphthoxazines, spiro(indoline)pyrido-benzoxazines, spiro(indoline)benzoxazines, spiro(benzindoline)benzoxazines, spiro(benzindoline)pyridobenzoxazines, spiro(indoline)fluoranthenoxazines, spiro(indoline)-quinoxazines, spiropiperidine-naphthopyrans, spiro(indoline)pyronobenzoxazinones, benzospiropyrans, naphthospiropyrans, spirobenzoxazine-pyrrolopyridines, spironaphthoxazine-pyrrolopyridines, spiro-oxazepin-benzoxazines, spiro-oxazepin-naphthoxazines, spiro(indoline)benzothiazoloxazines, spiro(indoline)benzopyrroloxazines, spiro(indoline)quinazolino-oxazines, spiro(indoline)-anthracenobenzoxazines, benzofurobenzopyrans, benzothienobenzopyrans, naphthofurobenzopyrans, benzopyrano-fusednaphthopyrans, spiro(isoindoline)-naphthoxazines, spiro(isoindoline)benzoxazines, etc. Such photochromic compounds are disclosed in U.S. Pat. Nos. 3100778, 3562172, 3567605, 3578602, 3671543, 4215010, 4342668, 4440672, 4634767, 4636561, 4637698, 4699473, 4719296, 4720547, 4772700, 4784474, 4785097, 4816584, 4818096, 4826977, 4831142, 4880667, 4929693, 4931219, 4931221, 4959471, 4980089, 4986934, 5055576, 5066818, 5110922, 5114621, 5139707, 5166345, 5171636, 5180524, 5186867, 5200116, 5238931, 5238981, 5244602, 5274132; 5340857, 5369158, 5384077, 5395567, 5399687, 5405958, 5411679, 5429774, 5451344, 5458814; 5458815, 5464567, 5466398, 5514817; 5520853, 5552090, 5552091, 5565147, 5573712; 5578252, 5585042, 5623005, 5637262, 5637709, 5645767; 5650098, 5651923, 5656206; 5658500, 5658501, 5674432, 5698141, 5723072, 5728758, 5730908, 5744070, 5759450, 5783116, 5801243, 5808063, 5811034, 5831090, 5833885, 5869658; 5879592, 5891368, 5955520; 5961892; 5981634, 5998520, 6004486, 6017121, 6018059; 6019914, 6022495, 6022497; 6030555, 6034193, 6065836, 6106744, 6106744, 6107395, 6113814, 6146554; 6149841, 6153126, 6221284, 6248264;

6296785, 6315928; 6342459; 6348604, 6353102, 6414057, 6478988, 6630597, 7556750, 7584630, 7999989, 8158037, 8697770, 8698117, 8741188, 9029532, 9028728, 9052438, 9097916, 9465234, 9487499, 9904074, 10197707, 10501446, 10532997, and 10502998 and are also described in the texts, Techniques in Chemistry, Volume III. "Photochromism", Chapter 3 (Glenn H. Brown, Editor, John Wiley and Sons, Inc., New York, 1971) and in "Chromic Phenomena: Technological Applications of Colour Chemistry" (P. Bamfield, RSC Books (2001)). Derivatives of these compounds that include various Substituents can be Synthesized from this teaching by people skilled in the art.

In a preferred embodiment, a polymerizable photochromic compound is used in the invention. Examples of preferred photochromic vinylic monomers include polymerizable naphthopyrans, polymerizable benzopyrans, polymerizable indenonaphthopyrans, polymerizable phenanthropyrans, polymerizable spiro(benzindoline)-naphthopyrans, polymerizable spiro(indoline)benzopyrans, polymerizable spiro(indoline)-naphthopyrans, polymerizable spiro(indoline)quinopyrans, polymerizable spiro(indoline)-pyrans, polymerizable naphthoxazines, polymerizable spirobenzopyrans; polymerizable spirobenzopyrans, polymerizable spirobenzothiopyrans, polymerizable naphthacenediones, polymerizable spirooxazines, polymerizable spiro(indoline)naphthoxazines, polymerizable spiro(indoline)-pyridobenzoxazines, polymerizable spiro(benzindoline)pyridobenzoxazines, polymerizable spiro(benzindoline)naphthoxazines, polymerizable spiro(indoline)-benzoxazines, polymerizable diarylethenes, and combinations thereof, as disclosed in U.S. Pat. Nos. 4929693, 5166345 6017121, 7556750, 7584630, 7999989, 8158037, 8697770, 8741188, 9052438, 9097916, 9465234, 9904074, 10197707, 6019914, 6113814, 6149841, 6296785, and 6348604.

Alternatively, a polymerizable photochromic compound can be obtained by reacting a reactive (meth)acrylamido or (meth)acryloxy monomer having a first reactive functional group ($-\text{COCl}$, $-\text{COBr}$, $-\text{COOH}$, $-\text{NHR}_{\text{N}2}$, $-\text{NCO}$, $-\text{OH}$, $-\text{CHO}$, $\text{---}\overset{\text{O}}{\text{V}}$, $\text{---}\overset{\text{N}}{\text{V}}\text{R}_0$, or $-\text{N}\text{---}\text{V}\text{---}\text{R}_0$ in which R_0 is hydrogen or methyl and $\text{R}_{\text{N}2}$ is hydrogen, a linear or branched $\text{C}_1\text{--}\text{C}_{15}$ alkyl, cyclohexyl, cyclopentyl, a substituted or unsubstituted phenyl, or a substituted- or unsubstituted-phenyl- $\text{C}_1\text{--}\text{C}_6$ alkyl) with a reactive photochromic compound having a second reactive functional group ($-\text{COOH}$, $-\text{NHR}_{\text{N}2}$, $-\text{N}\text{---}\text{NH}$, $\text{---}\text{NH}$, $\text{---}\text{NH}$, $-\text{NCO}$, $-\text{OH}$, $-\text{SH}$, $-\text{CHO}$, $\text{---}\overset{\text{O}}{\text{V}}$, $\text{---}\overset{\text{N}}{\text{V}}\text{R}_0$, or $-\text{N}\text{---}\text{V}\text{---}\text{R}_0$, in the absence or presence of a coupling agent (i.e., having two of the reactive functional groups listed above) under well known coupling reaction conditions, to form a photochromic compound.

Non-limiting examples of coupling reactions under various reaction conditions between a pair of matching co-reactive functional groups selected from the group preferably

consisting of primary group, secondary amino group, hydroxyl group, carboxyl group, acid anhydride group, aldehyde group, isocyanate group, epoxy group, aziridine group, azlactone group and thiol group, are given below for illustrative purposes. A primary/secondary amino group reacts with aldehyde or ketone group to form a Schiff base which may further be reduced into an amine bond; a primary/secondary amino group –NHR (in which R is hydrogen or C₁-C₆ alkyl) reacts with an acid chloride or bromide group or with an acid anhydride group to form an amide linkage (-CO-NR-); an amino group –NHR reacts with a N-hydroxysuccinimide ester group to form an amide linkage; an amino group –NHR reacts with a carboxylic acid group in the presence of a coupling agent – carbodiimide (e.g., 1-ethyl-3-(3-dimethylaminopropyl) carbodiimide (EDC), N,N'-dicyclohexylcarbodiimide (DCC), 1-cyclohexyl-3-(2-morpholinoethyl) carbodiimide, diisopropyl carbodiimide, or mixtures thereof) and N-hydroxysuccinimide to form an amide linkage; an amino group –NHR reacts (ring-opening) with an azlactone group to form an alkylene-diamido linkage (-CONH-CT₁T₂-(CH₂)_{p1}-CONH-); an amino group –NHR reacts with an isocyanate group to form a urea linkage (-NR-C(O)-NH- with R as defined above); an amino group –NHR reacts with an epoxy or aziridine group to form an amine bond (-C-NR-); a hydroxyl reacts with an isocyanate to form a urethane linkage; a hydroxyl reacts with an epoxy or aziridine to form an ether linkage (-O-); a hydroxyl reacts with an acid chloride or bromide group or with an acid anhydride group to form an ester linkage; a carboxyl group reacts with an epoxy group to form an ester bond; a thiol group (-SH) reacts with an isocyanate to form a thiocarbamate linkage (-N-C(O)-S-); a thiol group reacts with an epoxy or aziridine to form a thioether linkage (-S-); a thiol group reacts with an acid chloride or bromide group or with an acid anhydride group to form a thioester linkage; a thiol group reacts with an azlactone group in the presence of a catalyst to form a linkage (-CONH-CT₁T₂-(CH₂)_{p1}-CO-S-); a thiol group reacts with an ene group or vinylsulfonyl group based on thiol-ene "click" reaction under thiol-ene reaction conditions to form a thioether linkage (-S-); a thiol group reacts with a (meth)acryloyl group based on Michael Addition under appropriate reaction conditions to form a thioether linkage.

The reactions conditions for the above described coupling reactions are taught in textbooks and are well known to a person skilled in the art.

In accordance with the invention, coupling agents, each of which has two reactive functional groups, may be used in the coupling reactions. A coupling agent having two reactive functional groups can be: a diisocyanate compound; a di-acid halide compound; a di-carboxylic acid compound; a di-carboxylic acid anhydride compound; a diamine compound; a diol compound; a di-epoxy compound; a di-aziridine compound; di-azlactone compound; a di-ene compound; a divinylsulfone compound; a di-thiol compound; a thiolactone compound; an amino acid compound; a hydroxy-containing amine compound; an

amine compound having one hydroxyl or ketone group; a hydroxy-containing carboxylic acid compound; a mercaptane having a hydroxyl, carboxyl or amino group.

For example, a diisocyanate, di-carboxylic acid (preferably a di-carboxylic acid anhydride), di-aziridine, di-epoxy, or di-azlactone compound can be used in the coupling of the first and second reactive functional groups when they are different or same and are selected from the group consisting of hydroxyl, primary amino, secondary amino, and thiol groups; a di-amine, di-hydroxyl, di-thiol, hydroxy-containing amine, or hydroxy-containing thiol compound can be used in the coupling of the first and second reactive functional groups when they are different or same and are selected from the group consisting of isocyanate, epoxy, aziridine, and carboxylic acid groups; a di-epoxy compound can be used in the coupling of the first and second reactive functional groups when they both are carboxylic acid groups; a di-amine compound can be used in the coupling of the first and second reactive functional groups when they both are aldehyde groups. A person skilled in the art knows well how to select one or more coupling agents, based on selectivity and/or differential reactivity of a given reactive functional group, to link the first and second reactive functional groups to form a photochromic polydiorganosiloxane vinylic crosslinker of the invention.

Examples of commercially available di-carboxylic acid anhydrides include without limitation succinic acid anhydride, methylsuccinic anhydride, 2,2-dimethylsuccinic anhydride, 2,3-dimethylsuccinic acid, glutaric acid anhydride, 3,3-dimethylglutaric anhydride, 2,2-dimethylglutaric anhydride, 3-methylglutaric anhydride, 3,3-tetramethylglutaric anhydride, diglycolic anhydride, adipic anhydride, etc.

Any suitable C₃-C₂₄ di-carboxylic acid compounds can be used in the invention. Examples of preferred di-carboxylic acid compounds include without limitation a linear or branched C₃-C₂₄ aliphatic dicarboxylic acid, a C₅-C₂₄ cycloaliphatic or aliphatic-cycloaliphatic dicarboxylic acid, a C₆-C₂₄ aromatic or araliphatic dicarboxylic acid, a dicarboxylic acid which contains amino or imido groups or N-heterocyclic rings, and combinations thereof. Examples of suitable aliphatic dicarboxylic acids are: oxalic acid, malonic acid, succinic acid, glutaric acid, adipic acid, pimelic acid, suberic acid, azelaic acid, sebacic acid, undecanedioic acid, dodecanedioic acid, dimethylmalonic acid, octadecylsuccinic acid, trimethyladipic acid, and dimeric acids (dimerisation products of unsaturated aliphatic carboxylic acids, such as oleic acid). Examples of suitable cycloaliphatic dicarboxylic acids are: 1,3-cyclobutanedicarboxylic acid, 1,3-cyclopentanedicarboxylic acid, 1,3- and 1,4-cyclohexanedicarboxylic acid, 1,3- and 1,4-dicarboxylmethylcyclohexane, 4,4'-dicyclohexyldicarboxylic acid. Examples of suitable aromatic dicarboxylic acids are: terephthalic acid, isophthalic acid, o-phthalic acid, 1,3-, 1,4-, 2,6- or 2,7-naphthalenedicarboxylic acids, 4,4'-biphenyldicarboxylic acid, 2,2'-biphenyldicarboxylic acid, 4,4'-diphenylsulphone-dicarboxylic acid, 1,1,3-trimethyl-5-carboxyl-3-(p-

carboxyphenyl)-indane, 4,4'-diphenyl ether-dicarboxylic acid, bis-p-(carboxylphenyl)methane.

Any suitable diacid halides can be used in the invention. Examples of preferred diacid halide include without limitations fumaryl chloride, suberoyl chloride, succinyl chloride, phthaloyl chloride, isophthaloyl chloride, terephthaloyl chloride, sebacoyl chloride, adipoyl chloride, trimethyladipoyl chloride, azelaoyl chloride, dodecanedioic acid chloride, succinic chloride, glutaric chloride, oxalyl chloride, dimer acid chloride, and combinations thereof.

Any suitable diamines can be used in the invention. An organic diamine can be a linear or branched C₂-C₂₄ aliphatic diamine, a C₅-C₂₄ cycloaliphatic or aliphatic-cycloaliphatic diamine, or a C₆-C₂₄ aromatic or alkyl-aromatic diamine. A preferred organic diamine is N,N'-bis(hydroxyethyl)ethylenediamine, N,N'-dimethylethylenediamine, ethylenediamine, N,N'-dimethyl-1,3-propanediamine, N,N'-diethyl-1,3-propanediamine, N-methyl-1,3-diaminopropane, N-methylethylenediamine, N-ethylethylenediamine, N-isopropyl-1,3-propanediamine, N-propyl-1,3-propanediamine, N-butylethylenediamine, 2,2-dimethyl-1,3-propanediamine, 1,4-butanediamine, 1,5-pentanediamine, hexamethylenediamine, 2-Methyl-1,5-pentanediamine, 1,6-hexamethylenediamine, N,N'-dimethyl-1,6-hexamethylenediamine, 2,2,4(2,4,4)-trimethyl-1,6-hexanediamine, 1,3-diamino-2-propanol, 1,2-diaminoethane-1,2-diol, 1,1-diaminoethane-1,2-diol, 1,4-diamino-2,3-butanediol, 1,3-cyclopentanediamine, 1,4-diaminocyclohexane, 1,3-Bis(aminomethyl)cyclohexane, 4,4'-diaminodicyclohexylmethane, 4,4'-methylenebis(2-methyl-cyclohexylamine), isophorone diamine (3-aminomethyl-3,5,5-trimethylcyclohexylamine), m-xylylene diamine, p-xylylene diamine, piperazine, 1-(2-aminoethyl)piperazine, 1,4-bis(3-amino-propyl)piperazine, 2-piperazinoethylamine, 1-Boc-piperazine, 4-(2-aminoethyl)-1-Boc-piperazine, 1-(2-N-Boc-aminoethyl)piperazine, 4-(2-aminoethyl)-1-Boc-piperazine, 4-aminopiperidine, 3-aminopiperidine, 4-aminomethylpiperidine, 2-aminomethylpiperidine, 1-Boc-piperidine-4-carboxaldehyde, 1-Boc-piperidine-4-acetaldehyde, etc.

Any suitable C₄-C₂₄ diisocyanates can be used in the invention. Examples of preferred diisocyanates include without limitation tetramethylene diisocyanate, pentamethylene diisocyanate, hexamethylene diisocyanate, heptamethylene diisocyanate, 1,6-diisocyanato-2,2,4-trimethylhexane, octamethylene diisocyanate, dodecamethylene diisocyanate, cyclohexane diisocyanate, 1,3-bis-(4,4'-isocyanatomethyl)cyclohexane, isophorone diisocyanate, 4,4'-dicyclohexylmethane diisocyanate, toluene diisocyanate, 4,4'-diphenyl diisocyanate, 4,4'-diphenylmethane diisocyanate, p-phenylene diisocyanate, etc.

Any suitable di-epoxy compounds can be used in the invention. Examples of preferred di-epoxy compounds are neopentyl glycol diglycidyl ether, 1,3-butadiene diepoxide, 1,4-butanediol diglycidyl ether, 1,2,7,8-diepoxyoctane, 3,4-epoxycyclohexylmethyl-3,4-epoxycyclohexane, 3,4-epoxycyclohexylmethyl-3,4-

epoxycyclohexane carboxylate, vinylcyclohexene dioxide, 1,6-hexanediol diglycidyl ether, glycerol diglycidyl ether, ethylene glycol diglycidyl ether, diethylene glycol diglycidyl ether, polyethylene glycol diglycidyl ether, propylene glycol diglycidyl ether, dipropylene glycol diglycidyl ether, polypropylene glycol diglycidyl ether, bis[4-(glycidyloxy)phenyl]methane, bisphenol A diglycidyl ether (2,2-bis[4-(glycidyloxy)phenyl]propane), bisphenol A propoxylate diglycidyl ether, etc.

Any suitable C₂-C₂₄ diols (i.e., compounds with two hydroxyl groups) can be used in the invention. Examples of preferred diols include without limitation ethylene glycol, diethylene glycol, triethylene glycol, tetraethylene glycol, polyethylene glycol, propylene glycol, 1,4-butanediol, various pentanediols, various hexanediols, various cyclohexanediols, 1,4-bis(2-hydroxyethyl)piperazine, bisphenol A, bisphenol F, 4,4'-methylenediphenol, etc.

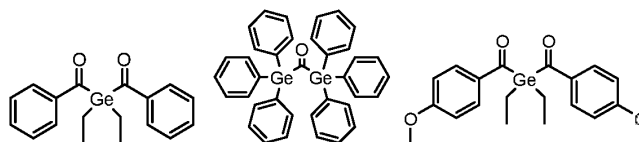
Any amino acids can be used in the invention. Examples of amino acids include without limitation glycine, proline, alanine, valine, isoleucine, leucine, 2-aminoisobutyric acid, 4-aminobutyric acid, 3-aminoisobutyric acid, 3-amino-butyric acid, β -alanine, 1-amino-3-cyclopentane carboxylic acid, 3-aminocyclohexanecarboxylic acid, pyrrolidine-3-carboxylic acid, 4-piperidinecarboxylic acid, 3-piperidinecarboxylic acid, 1-piperazineacetic acid, etc.

Examples of compounds having one amino group and one hydroxyl or aldehyde group (or ketone group) include without limitation 1-piperazinepropanol, 2-[2-(1-piperazinyl)-ethoxy]ethanol, 4-amino-1-piperazineethanol, 4-piperidinemethanol, 1-Boc-piperidine-4-carboxaldehyde, 4-formylpiperidine, N-Boc-4-piperidineacetaldehyde, etc.

Preferred reactive (meth)acrylamido or (meth)acryloxy monomers without limitation include those described later in this application.

Preferred photochromic compounds without limitation include without limitation those compounds that are described later in this application and can be obtained from commercial suppliers or prepared according to procedures described in the patents discussed above and in the texts described above.

Any acylgermanium photoinitiators can be used in this invention, so long as they are capable of initiating a free-radical polymerization under irradiation with a light source including a light in the region of about 420 to about 500 nm. Examples of acylgermanium photoinitiators are acylgermanium compounds described in U.S. Pat. No. 7605190. Preferably, the monomer of the polymerizable composition comprises at least one of the following acylgermanium compounds.



In a preferred embodiment, a polymerizable composition further comprises at least

one UV-absorbing vinylic monomer and optionally (but preferably) one or more UV/HEVL-absorbing vinylic monomers. The term "UV/HEVL-absorbing vinylic monomer" refers to a vinylic monomer that can absorb UV light and high-energy-violet-light (i.e., light having wavelength between 380 nm and 440 nm).

Any suitable UV-absorbing vinylic monomers and UV/HEVL-absorbing vinylic monomers can be used in a polymerizable composition for preparing a preformed SiHy contact lens of the invention. Examples of preferred UV-absorbing and UV/HEVL-absorbing vinylic monomers include without limitation: 2-(2-hydroxy-5-vinylphenyl)-2H-benzotriazole, 2-(2-hydroxy-5-acryloyloxyphenyl)-2H-benzotriazole, 2-(2-hydroxy-3-methacrylamido methyl-5-tert octylphenyl) benzotriazole, 2-(2'-hydroxy-5'-methacrylamidophenyl)-5-chlorobenzotriazole, 2-(2'-hydroxy-5'-methacrylamidophenyl)-5-methoxybenzotriazole, 2-(2'-hydroxy-5'-methacryloxy-propyl-3'-t-butyl-phenyl)-5-chlorobenzotriazole, 2-(2'-hydroxy-5'-methacryloxypropylphenyl) benzotriazole, 2-hydroxy-5-methoxy-3-(5-(trifluoromethyl)-2H-benzo[d][1,2,3]triazol-2-yl)benzyl methacrylate (WL-1), 2-hydroxy-5-methoxy-3-(5-methoxy-2H-benzo[d][1,2,3]triazol-2-yl)benzyl methacrylate (WL-5), 3-(5-fluoro-2H-benzo[d][1,2,3]triazol-2-yl)-2-hydroxy-5-methoxybenzyl methacrylate (WL-2), 3-(2H-benzo[d][1,2,3]triazol-2-yl)-2-hydroxy-5-methoxybenzyl methacrylate (WL-3), 3-(5-chloro-2H-benzo[d][1,2,3]triazol-2-yl)-2-hydroxy-5-methoxybenzyl methacrylate (WL-4), 2-hydroxy-5-methoxy-3-(5-methyl-2H-benzo[d][1,2,3]triazol-2-yl)benzyl methacrylate (WL-6), 2-hydroxy-5-methyl-3-(5-(trifluoromethyl)-2H-benzo[d][1,2,3]triazol-2-yl)benzyl methacrylate (WL-7), 4-allyl-2-(5-chloro-2H-benzo[d][1,2,3]triazol-2-yl)-6-methoxy-phenol (WL-8), 2-{2'-Hydroxy-3'-tert-5'-'[3''-(4''-vinylbenzyloxy)propoxy]phenyl}-5-methoxy-2H-benzotriazole, phenol, 2-(5-chloro-2H-benzotriazol-2-yl)-6-(1,1-dimethylethyl)-4-ethenyl- (UVAM), 2-[2'-hydroxy-5'-(2-methacryloxyethyl)phenyl]-2H-benzotriazole (2-Propenoic acid, 2-methyl-, 2-[3-(2H-benzotriazol-2-yl)-4-hydroxyphenyl]ethyl ester, Norbloc), 2-{2'-Hydroxy-3'-tert-butyl-5'-[3'-methacryloyloxypropoxy]phenyl}-2H-benzotriazole, 2-[2'-Hydroxy-3'-tert-butyl-5'-[3'-methacryloyloxypropoxy]phenyl]-5-methoxy-2H-benzotriazole (UV13), 2-{2'-Hydroxy-3'-tert-butyl-5'-[3'-methacryloyloxypropoxy]phenyl}-5-chloro-2H-benzotriazole (UV28), 2-[2'-Hydroxy-3'-tert-butyl-5'-(3'-acryloyloxypropoxy)phenyl]-5-trifluoromethyl-2H-benzotriazole (UV23), 2-(2'-hydroxy-5-methacrylamidophenyl)-5-methoxybenzotriazole (UV6), 2-(3-allyl-2-hydroxy-5-methylphenyl)-2H-benzotriazole (UV9), 2-(2-Hydroxy-3-methallyl-5-methylphenyl)-2H-benzotriazole (UV12), 2-3'-t-butyl-2'-hydroxy-5'-(3''-dimethylvinylsilylpropoxy)-2'-hydroxyphenyl)-5-methoxybenzotriazole (UV15), 2-(2'-hydroxy-5'-methacryloylpropyl-3'-tert-butylphenyl)-5-methoxy-2H-benzotriazole (UV16), 2-(2'-hydroxy-5'-acryloylpropyl-3'-tert-butylphenyl)-5-methoxy-2H-benzotriazole (UV16A), 2-Methylacrylic acid 3-[3-tert-butyl-5-(5-chlorobenzotriazol-2-yl)-4-hydroxyphenyl]-propyl ester (16-100, CAS#96478-15-8), 2-(3-(tert-butyl)-4-hydroxy-5-(5-methoxy-2H-benzo[d][1,2,3]triazol-2-yl)phenoxy)ethyl methacrylate

(16-102); Phenol, 2-(5-chloro-2*H*-benzotriazol-2-yl)-6-methoxy-4-(2-propen-1-yl) (CAS#1260141-20-5); 2-[2-Hydroxy-5-[3-(methacryloyloxy)propyl]-3-*tert*-butylphenyl]-5-chloro-2*H*-benzotriazole; Phenol, 2-(5-ethenyl-2*H*-benzotriazol-2-yl)-4-methyl-, homopolymer (9CI) (CAS#83063-87-0). In accordance with the invention, the polymerizable composition comprises about 0.1% to about 2.5%, preferably about 0.2% to about 20%, more preferably about 0.3% to about 1.5%, by weight of one or more UV-absorbing vinylic monomers, related to the amount of all polymerizable components in the polymerizable composition.

It is understood that the amount of at least one UV-absorbing vinylic monomer in the polymerizable composition is sufficient to render a contact lens, which is obtained from the curing of the polymerizable composition, an ability of blocking or absorbing (i.e., the inverse of transmittance) at least 90% (preferably at least about 95%, more preferably at least about 97.5%, even more preferably at least about 99%) of UVB (between 280 and 315 nanometers), at least 70% (preferably at least about 80%, more preferably at least about 90%, even more preferably at least about 95%) of UVA transmittance (between 315 and 380 nanometers), and optionally (but preferably) at least 30% (preferably at least about 40%, more preferably at least about 50%, even more preferably at least about 60%) of violet light between 380 nm and 440 nm, which impinge on the lens.

In accordance with the present invention, the polymerizable composition can also comprise additional polymerizable components selected from the group consisting of one or more secondary hydrophilic vinylic monomers (i.e., hydrophilic vinylic monomers other than hydrophilic (meth)acrylamido monomer), one or more secondary siloxane-containing vinylic monomers (i.e., siloxane-containing vinylic monomers other than siloxane-containing acrylamido monomer), one or more hydrophobic vinylic monomers, one or more non-silicone vinylic crossliners, and combinations thereof. It is understood that if the other polymerizable components are present in the polymerizable composition, their total amount is about 10% or less by weight (preferably about 7% or less by weight, more preferably about 4% or less by weight, even more preferably about 2% or less by weight) relative the total amount of all polymerizable component in the polymerizable composition.

Any hydrophilic vinylic monomers other than hydrophilic (meth)acrylamido monomer can be used as secondary hydrophilic vinylic monomers in the invention. Examples of such secondary hydrophilic vinylic monomers are hydroxyl-containing (meth)acryloxy monomers (as described below), amino-containing (meth)acryloxy monomers (as described later in this application), carboxyl-containing (meth)acryloxy monomers (as described later in this application), N-vinyl amide monomers (as described later in this application), methylene-containing pyrrolidone monomers (i.e., pyrrolidone derivatives each having a methylene group connected to the pyrrolidone ring at 3- or 5- position) (as described later in this application), (meth)acryloxy monomers having a C₁-C₄ alkoxyethoxy group (as described

later in this application), vinyl ether monomers (as described later in this application), allyl ether monomers (as described later in this application), phosphorylcholine-containing vinylic monomers (as described later in this application), N-2-hydroxyethyl vinyl carbamate, N-carboxyvinyl- β -alanine (VINAL), N-carboxyvinyl- α -alanine, and combinations thereof. Among the preferred hydrophilic vinylic monomers are 2-hydroxyethyl (meth)acrylate aminopropyl (meth)acrylate hydrochloride, dimethylaminoethyl (meth)acrylate, glycerol methacrylate (GMA), ethylene glycol methyl ether (meth)acrylate, N-vinyl-2-pyrrolidone (NVP), allyl alcohol, (meth)acrylic acid, a C₁-C₄-alkoxy polyethylene glycol (meth)acrylate having a weight average molecular weight of from 200 to 1500, N-vinyl acetamide, N-vinyl-N-methyl acetamide, allyl alcohol, and combinations thereof.

Any siloxane-containing vinylic monomer (other than siloxane-containing (meth)acrylamido monomers) known to a person skilled in the art can be used as secondary siloxane-containing vinylic monomer in the invention. Examples of such siloxane-containing vinylic monomers include without limitation siloxane-containing (meth)acryloxy monomers each having a bis(trialkylsilyloxy)alkylsilyl group or a tris(trialkylsilyloxy)silyl group, mono-(meth)acryloxy-terminated mono-alkyl-terminated polysiloxanes, 3-methacryloxy propylpentamethyldisiloxane, t-butyl dimethyl-siloxyethyl vinyl carbonate, trimethylsilylethyl vinyl carbonate, and trimethylsilylmethyl vinyl carbonate, and combinations thereof.

Preferred polysiloxanes vinylic monomers including those of formula (M1) are described later in this application and can be obtained from commercial suppliers (e.g., Shin-Etsu, Gelest, etc.); prepared according to procedures described in patents, e.g., U.S. Pat. Nos. 5070215, 6166236, 6867245, 8415405, 8475529, 8614261, and 9217813; prepared by reacting a hydroxyalkyl (meth)acrylate or (meth)acrylamide or a (meth)acryloxypolyethylene glycol with a mono-epoxypropyloxypropyl-terminated polydimethylsiloxane; prepared by reacting glycidyl (meth)acrylate with a mono-carbinol-terminated polydimethylsiloxane, a mono-aminopropyl-terminated polydimethylsiloxane, or a mono-ethylaminopropyl-terminated polydimethylsiloxane; or prepared by reacting isocyanatoethyl (meth)acrylate with a mono-carbinol-terminated polydimethylsiloxane according to coupling reactions well known to a person skilled in the art.

Preferred siloxane-containing (meth)acryloxy monomers each having a bis(trialkylsilyloxy)alkylsilyl group or a tris(trialkylsilyloxy)silyl group, including those of formula (M2), are described later in this application and can be obtained from commercial suppliers (e.g., Shin-Etsu, Gelest, etc.) or can be prepared according to procedures described in U.S. Pat. Nos. 5070215, 6166236, 7214809, 8475529, 8658748, 9097840, 9103965, and 9475827.

Any hydrophobic vinylic monomers can be in this invention. Examples of preferred hydrophobic vinylic monomers include methyl (meth)acrylate, ethyl (meth)acrylate, propyl

(meth)acrylate, isopropyl (meth)acrylate, cyclohexyl (meth)acrylate, 2-ethylhexyl (meth)acrylate, vinyl acetate, vinyl propionate, vinyl butyrate, vinyl valerate, styrene, chloroprene, vinyl chloride, vinylidene chloride, (meth)acrylonitrile, 1-butene, butadiene, vinyl toluene, vinyl ethyl ether, perfluorohexylethyl-thio-carbonyl-aminoethyl-methacrylate, isobornyl (meth)acrylate, trifluoroethyl (meth)acrylate, hexafluoro-isopropyl (meth)acrylate, hexafluorobutyl (meth)acrylate, and combinations thereof.

In accordance with the invention, any non-silicone vinylic crosslinkers can be in this invention. Examples of preferred non-silicone vinylic cross-linking agents are described later in this application.

A polymerizable composition of the invention can further comprise visibility tinting agents (e.g., D&C Blue No. 6, D&C Green No. 6, D&C Violet No. 2, carbazole violet, certain copper complexes, certain chromium oxides, various iron oxides, phthalocyanine green, phthalocyanine blue, titanium dioxides, or mixtures thereof), antimicrobial agents (e.g., silver nanoparticles), a bioactive agent (e.g., a drug, an amino acid, a polypeptide, a protein, a nucleic acid, 2-pyrrolidone-5-carboxylic acid (PCA), an alpha hydroxyl acid, linoleic and gamma linoleic acids, vitamins, or any combination thereof), leachable lubricants (e.g., a non-crosslinkable hydrophilic polymer having an average molecular weight from 5,000 to 500,000, preferably from 10,000 to 300,000, more preferably from 20,000 to 100,000 Daltons), leachable tear-stabilizing agents (e.g., a phospholipid, a monoglyceride, a diglyceride, a triglyceride, a glycolipid, a glyceroglycolipid, a sphingolipid, a sphingo-glycolipid, a fatty acid having 8 to 36 carbon atoms, a fatty alcohol having 8 to 36 carbon atoms, or a mixture thereof), and the like, as known to a person skilled in the art.

In accordance with the invention, a polymerizable composition of the invention is a fluid composition, which can be a solution, a solventless blend (i.e., a fluid composition free of any non-reactive diluent - organic solvent).

Where a polymerizable composition of the invention is a solution. It can be prepared by dissolving all of the desirable components in any suitable solvent known to a person skilled in the art. Example of suitable solvents includes without limitation, water, tetrahydrofuran, tripropylene glycol methyl ether, dipropylene glycol methyl ether, ethylene glycol n-butyl ether, ketones (e.g., acetone, methyl ethyl ketone, etc.), diethylene glycol n-butyl ether, diethylene glycol methyl ether, ethylene glycol phenyl ether, propylene glycol methyl ether, propylene glycol methyl ether acetate, dipropylene glycol methyl ether acetate, propylene glycol n-propyl ether, dipropylene glycol n-propyl ether, tripropylene glycol n-butyl ether, propylene glycol n-butyl ether, dipropylene glycol n-butyl ether, tripropylene glycol n-butyl ether, propylene glycol phenyl ether dipropylene glycol dimethyl ether, polyethylene glycols, polypropylene glycols, ethyl acetate, butyl acetate, amyl acetate, methyl lactate, ethyl lactate, i-propyl lactate, methylene chloride, 2-butanol, 1-propanol, 2-propanol,

menthol, cyclohexanol, cyclopentanol and exonorborneol, 2-pentanol, 3-pentanol, 2-hexanol, 3-hexanol, 3-methyl-2-butanol, 2-heptanol, 2-octanol, 2-nonanol, 2-decanol, 3-octanol, norborneol, tert-butanol, tert-amyl, alcohol, 2-methyl-2-pentanol, 2,3-dimethyl-2-butanol, 3-methyl-3-pentanol, 1-methylcyclohexanol, 2-methyl-2-hexanol, 3,7-dimethyl-3-octanol, 1-chloro-2-methyl-2-propanol, 2-methyl-2-heptanol, 2-methyl-2-octanol, 2-2-methyl-2-nonanol, 2-methyl-2-decanol, 3-methyl-3-hexanol, 3-methyl-3-heptanol, 4-methyl-4-heptanol, 3-methyl-3-octanol, 4-methyl-4-octanol, 3-methyl-3-nonanol, 4-methyl-4-nonanol, 3-methyl-3-octanol, 3-ethyl-3-hexanol, 3-methyl-3-heptanol, 4-ethyl-4-heptanol, 4-propyl-4-heptanol, 4-isopropyl-4-heptanol, 2,4-dimethyl-2-pentanol, 1-methylcyclopentanol, 1-ethylcyclopentanol, 1-ethylcyclopentanol, 3-hydroxy-3-methyl-1-butene, 4-hydroxy-4-methyl-1-cyclopentanol, 2-phenyl-2-propanol, 2-methoxy-2-methyl-2-propanol, 2,3,4-trimethyl-3-pentanol, 3,7-dimethyl-3-octanol, 2-phenyl-2-butanol, 2-methyl-1-phenyl-2-propanol and 3-ethyl-3-pentanol, 1-ethoxy-2-propanol, 1-methyl-2-propanol, t-amyl alcohol, isopropanol, 1-methyl-2-pyrrolidone, N,N-dimethylpropionamide, dimethyl formamide, dimethyl acetamide, dimethyl propionamide, N-methyl pyrrolidinone, and mixtures thereof. Preferably, a polymerizable composition is a solution of all the desirable components in water, 1,2-propylene glycol, a polyethyleneglycol having a molecular weight of about 400 Daltons or less, or a mixture thereof.

Where a polymerizable composition of the invention is a solventless blend, it can be prepared by mixing all polymerizable components and other necessary component. A solventless polymerizable composition typically comprises at least one blending vinylic monomer as a reactive solvent for dissolving all other polymerizable components of the solventless polymerizable composition. Examples of preferred blending vinylic monomers are described later in this application. Preferably, methyl methacrylate is used as a blending vinylic monomer in preparing a solventless polymerizable composition.

In a preferred embodiment, a polymerizable composition of the invention comprises: (1) from about 5% to about 60% by weight, preferably from about 10% to about 50% by weight, even more preferably from about 15% to about 45% by weight, even more preferably from about 20% to about 40% by weight, of at least one hydrophilic (meth)acrylamide-type monomer; (2) from about 5% to about 50% by weight, preferably from about 10% to about 40% by weight, more preferably from about 15% to about 30% by weight, of at least one siloxane-containing (meth)acrylamido monomer; (3) from about 5% to about 50%, preferably from about 10% to about 40% by weight, more preferably from about 15% to about 35% by weight, of at least one polysiloxane vinylic crosslinker (preferably at least one α,ω -dimethacrylamido-terminated polysiloxane crosslinker, more preferably at least one α,ω -dimethacrylamido-terminated polysiloxane crosslinker of formula (V)); (4) from about 0.1% to about 2.5% by weight, preferably from about 0.2% to about 2.0% by weight, more preferably

from about 0.3% to about 1.5% by weight, of at least one UV-absorbing vinylic monomer; (5) from about 0.005% to about 1.0% by weight (preferably from about 0.01% to about 0.8% by weight, more preferably from about 0.02% to about 0.6% by weight, even more preferably from about 0.05% to about 0.5% by weight) of a photochromic compound, and (6) from about 0.05% to about 1.5% by weight, preferably from about 0.1% to 1.3% by weight, more preferably from about 0.5% to about 1.1% by weight, of at least one acygermanium photoinitiator. The foregoing range combinations are presented with the proviso that the listed components and any additional components add up to 100% by weight.

In accordance with the invention, the polymerizable composition of the invention can be introduced (dispensed) into a cavity formed by a mold according to any known methods.

After the polymerizable composition is dispensed into the mold, it is polymerized to produce a photochromic SiHy contact lens. Crosslinking is initiated upon exposure to a visible light in a region between 420 nm to 500 nm, preferably under a spatial limitation of actinic radiation, to crosslink the polymerizable components in the polymerizable composition.

In accordance with the invention, light source can be any ones emitting light in the 420-500 nm range sufficient to activate acylgermanium photoinitiators. Blue-light sources are commercially available and include: the Palatray CU blue-light unit (available from Heraeus Kulzer, Inc., Irvine, Calif.), the Fusion F450 blue light system (available from TEAMCO, Richardson, Tex.), Dymax Blue Wave 200, LED light sources from Opsytec (435 nm, 445 nm, 460 nm), and the GE 24" blue fluorescent lamp (available from General Electric Company, U.S.). A preferred blue-light source is the LED from Opsytec (those described above).

The crosslinking according to the invention may be effected in a very short time, e.g. in $\leq$ about 80 seconds, preferably in $\leq$ about 65 seconds, more preferably in $\leq$ 50 about seconds, even more preferably in $\leq$ about 35 seconds, and most preferably in 5 to 30 seconds.

Opening of the mold so that the molded photochromic SiHy contact lens can be removed from the mold may take place in a manner known *per se*.

The molded photochromic SiHy contact lenses can be subject to lens extraction with a liquid extraction medium to remove unpolymerized polymerizable components and formed and oligomers. In accordance with the invention, the extraction liquid medium is any solvent capable of dissolving the organic solvent, unpolymerized polymerizable materials, and oligomers in the dry contact lens. Water, any organic solvents known to a person skilled in the art, or a mixture thereof can be used in the invention. Preferably, the organic solvents used extraction liquid medium are water, a buffered saline, a C₁-C₃ alkyl alcohol, 1,2-

propylene glycol, a polyethyleneglycol having a number average molecular weight of about 400 Daltons or less, a C₁-C₆ alkylalcohol, or combinations thereof.

The molded photochromic SiHy contact lenses can also subject to further processes, such as, for example, surface treatment (for example, such as, plasma treatment, chemical treatments, the grafting of hydrophilic monomers or macromers onto the surface of a lens, Layer-by-layer coating, in-package crosslinking of a thermally-reactive hydrophilic polymeric material, etc.); packaging in lens packages with a packaging solution which can contain about 0.005% to about 5% by weight of a wetting agent (e.g., a hydrophilic polymer), a viscosity-enhancing agent (e.g., methyl cellulose (MC), ethyl cellulose, hydroxymethylcellulose, hydroxyethyl cellulose (HEC), hydroxypropylcellulose (HPC), hydroxypropylmethyl cellulose (HPMC), or a mixture thereof), or an in-package-coating material; sterilization such as autoclave at from 118 to 124°C for at least about 30 minutes; and the like.

Preferred surfaces treatments are LbL coating such as those described in US Patent Ser. No. 6,451,871, 6,719,929, 6,793,973, 6,811,805, 6,896,926, plasma treatment, in-package-coating such as those disclosed in US Pat. Nos. 8557334, 8529057 and 9505184. A preferred plasma treatment is those processes in which an ionized gas is applied to the surface of an article as described in U.S. Pat. Nos. 4,312,575 and 4,632,844.

The molded photochromic SiHy contact lens is hydrated in water or an aqueous solution to replace the liquid extraction medium, according to any method known to a person skilled in the art.

The hydrated and/or surface-treated photochromic SiHy contact lens can further subject to further processes, such as, for example, packaging in lens packages with a packaging solution which is well known to a person skilled in the art; sterilization such as autoclave at from 118 to 124°C for at least about 30 minutes; and the like.

Lens packages (or containers) are well known to a person skilled in the art for autoclaving and storing a soft contact lens. Any lens packages can be used in the invention. Preferably, a lens package is a blister package which comprises a base and a cover, wherein the cover is detachably sealed to the base, wherein the base includes a cavity for receiving a sterile packaging solution and the contact lens.

Lenses are packaged in individual packages, sealed, and sterilized (e.g., by autoclave at about 120°C or higher for at least 30 minutes under pressure) prior to dispensing to users. A person skilled in the art will understand well how to seal and sterilize lens packages.

A contact lens of the invention has an oxygen permeability of preferably at least about 40 barrers, more preferably at least about 60 barrers, even more preferably at least about 80 barrers (at about 35°C).

A contact lens of the invention has an elastic modulus of about 1.5 MPa or less, preferably about 1.2 MPa or less, more preferably from about 0.3 MPa to about 1.0 MPa (at a temperature of from about 22°C to 28°C).

A contact lens of the invention further has an equilibrium water content of from about 15% to about 75%, more preferably from about 20% to about 70% by weight, even more preferably from about 25% to about 65% by weight (at room temperature) when fully hydrated. The equilibrium water content of a photochromic SiHy contact lens can be measured according to the procedure disclosed in Example 1.

In a further aspect, the invention provides a photochromic SiHy contact lens obtained by the method of the invention.

All of the various embodiments of the molds, polymerizable composition, and spatial limitation of radiation, and contact lens of the invention described above can be used in this aspect of the invention.

Although various embodiments of the invention have been described using specific terms, devices, and methods, such description is for illustrative purposes only. The words used are words of description rather than of limitation. It is to be understood that changes and variations may be made by those skilled in the art without departing from the spirit or scope of the present invention, which is set forth in the following claims. In addition, it should be understood that aspects of the various embodiments may be interchanged either in whole or in part or can be combined in any manner and/or used together, as illustrated below:

1. A polymerizable composition for actinically cast-molding photochromic silicone hydrogel contact lenses (preferably under a spatial limitation of actinic radiation), wherein the polymerizable composition is a fluid composition which comprises:
 - (a) at least one hydrophilic (meth)acrylamido monomer,
 - (b) at least one siloxane-containing (meth)acrylamido monomer,
 - (c) at least one polysiloxane vinylic crosslinker,
 - (d) at least one photochromic compound, and
 - (e) from about 0.05% to about 1.5% by weight of at least one an acyl germanium photoinitiator relative to the total weight of the polymerizable composition, wherein the acyl germanium photoinitiator is capable of initiating a free-radical polymerization under irradiation with a light source including a light in the region of about 420 to about 500 nm,wherein the sum of the amounts of components (a) to (e) is at least 90% by weight relative to total amount of all polymerizable components in the polymerizable composition, wherein the polymerizable composition has a curing time of about 80 seconds or less as determined in photo-rheology study using a LED light source

which provide a visible light with a spectral centroid of about 452nm and a total irradiance of about 54 mW/cm².

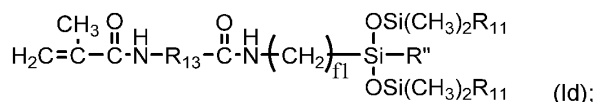
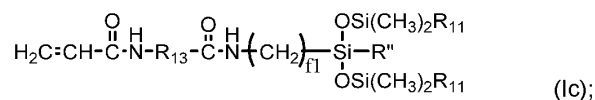
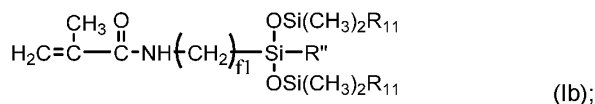
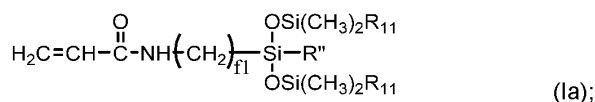
2. The polymerizable composition of embodiment 1, wherein the sum of the amounts of components (a) to (e) is at least 93% by weight relative to total amount of all polymerizable components in the polymerizable composition.
3. The polymerizable composition of embodiment 1, wherein the sum of the amounts of components (a) to (e) is at least 96% by weight relative to total amount of all polymerizable components in the polymerizable composition.
4. The polymerizable composition of embodiment 1, wherein the sum of the amounts of components (a) to (e) is at least 98% by weight relative to total amount of all polymerizable components in the polymerizable composition.
5. The polymerizable composition of any one of embodiments 1 to 4, wherein the polymerizable composition has a curing time of about 65 seconds or less as determined in photo-rheology study using a LED light source which provide a visible light with a spectral centroid of about 452nm and a total irradiance of about 54 mW/cm².
6. The polymerizable composition of any one of embodiments 1 to 4, wherein the polymerizable composition has a curing time of about 50 seconds or less as determined in photo-rheology study using a LED light source which provide a visible light with a spectral centroid of about 452nm and a total irradiance of about 54 mW/cm².
7. The polymerizable composition of any one of embodiments 1 to 4, wherein the polymerizable composition has a curing time of about 35 seconds or less as determined in photo-rheology study using a LED light source which provide a visible light with a spectral centroid of about 452nm and a total irradiance of about 54 mW/cm².
8. The polymerizable composition of any one of embodiments 1 to 7, wherein the polymerizable composition comprises at least one photochromic compound in an amount of from about 0.005% to about 1.0% by weight, relative to the total weight of the polymerizable composition.
9. The polymerizable composition of any one of embodiments 1 to 7, wherein the polymerizable composition comprises at least one photochromic compound in an amount of from about 0.01% to about 0.8% by weight, relative to the total weight of the polymerizable composition.
10. The polymerizable composition of any one of embodiments 1 to 7, wherein the polymerizable composition comprises at least one photochromic compound in an amount of from about 0.02% to about 0.6% by weight, relative to the total weight of

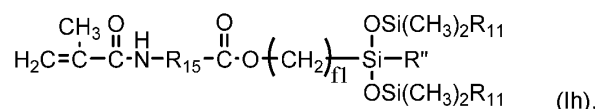
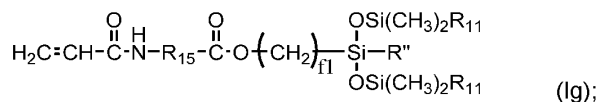
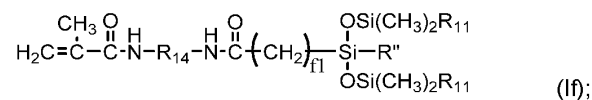
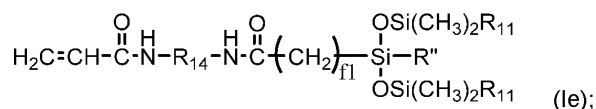
the polymerizable composition.

11. The polymerizable composition of any one of embodiments 1 to 7, wherein the polymerizable composition comprises at least one photochromic compound in an amount of from about 0.05% to about 0.5% by weight, relative to the total weight of the polymerizable composition.
12. The polymerizable composition of any one of embodiments 1 to 11, wherein said at least one hydrophilic (meth)acrylamido monomer comprises (meth)acrylamide, N,N-dimethyl (meth)acrylamide, N-ethyl (meth)acrylamide, N,N-diethyl (meth)acrylamide, N-propyl (meth)acrylamide, N-isopropyl (meth)acrylamide, N-3-methoxypropyl (meth)acrylamide, N-2-hydroxyethyl (meth)acrylamide, N,N-bis(hydroxyethyl) (meth)acrylamide, N-3-hydroxypropyl (meth)acrylamide, N-2-hydroxypropyl (meth)acrylamide, N-2,3-dihydroxypropyl (meth)acrylamide, N-4-hydroxybutyl (meth)acrylamide, N,N-bis(2-hydroxyethyl) (meth)acrylamide, *N*-tris(hydroxymethyl) methyl (meth)acrylamide, 2-(meth)acrylamidoglycolic acid, 3-(meth)acrylamido-propionic acid, 4-(meth)acrylamido-butanoic acid, 3-(meth)acrylamido-2-methylbutanoic acid, 3-(meth)acrylamido-3-methylbutanoic acid, 2-(meth)acrylamido-2-methyl-3,3-dimethyl butanoic acid, 5-(meth)acrylamidopentanoic acid, 3-(meth)acrylamidohexanoic acid, 4-(meth)acrylamido-3,3-dimethylhexanoic acid, (3-(meth)acrylamidophenyl)boronic acid, 3-((3-methacrylamidopropyl)dimethylammonio)-propane-1-sulfonate; 3-((3-acrylamidopropyl)dimethylammonio)propane-1-sulfonate, N-2-aminoethyl (meth)acrylamide, N-2-methylaminoethyl (meth)acrylamide, N-2-ethylaminoethyl (meth)acrylamide, N-2-dimethylaminoethyl (meth)acrylamide, N-3-aminopropyl (meth)acrylamide, N-3-methylaminopropyl (meth)acrylamide, N-3-dimethylaminopropyl (meth)acrylamide, poly(ethylene glycol)ethyl (meth)acrylamide having a number average molecular weight of up to 700, methoxy-poly(ethylene glycol)ethyl (meth)acrylamide having a number average molecular weight of up to 700, or combination thereof.
13. The polymerizable composition of any one of embodiments 1 to 11, wherein said at least one hydrophilic (meth)acrylamido monomer comprises N,N-dimethyl (meth)acrylamide, N-ethyl (meth)acrylamide, N-2-hydroxyethyl (meth)acrylamide, N-3-hydroxypropyl (meth)acrylamide, N-2-hydroxypropyl (meth)acrylamide, N-2,3-dihydroxypropyl (meth)acrylamide, (meth)acrylamide, N-(2-aminoethyl)(meth)acrylamide, N-(3-aminopropyl)(meth)acrylamide, or combinations thereof, more preferably is N,N-dimethylacrylamide.
14. The polymerizable composition of any one of embodiments 1 to 11, wherein said at least one hydrophilic (meth)acrylamido monomer comprises N,N-dimethyl

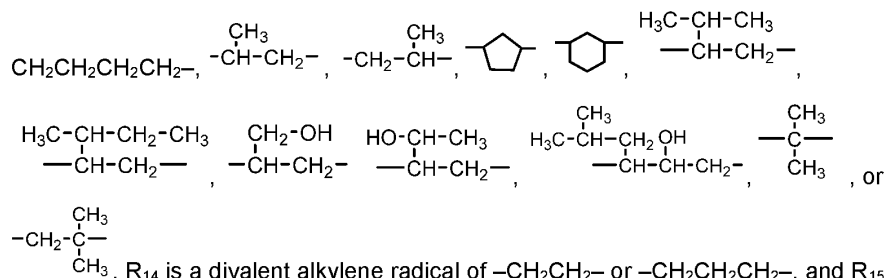
(meth)acrylamide.

15. The polymerizable composition of any one of embodiments 1 to 14, wherein said at least one siloxane-containing (meth)acrylamido monomer comprises a (meth)acrylamido monomer containing a tris(trialkylsiloxy)silyl group.
16. The polymerizable composition of embodiment 15, wherein the (meth)acrylamido monomer containing a tris(trialkylsiloxy)silyl group is N-[tris(trimethylsiloxy)silylpropyl] (meth)acrylamide, N-[tris(dimethylethylsiloxy)-silylpropyl] (meth)acrylamide, N-[tris(dimethylpropylsiloxy)silylpropyl]acrylamide, N-[tris(dimethylphenylsiloxy)silylpropyl] (meth)acrylamide, N-(2-hydroxy-3-(3-(tris(trimethylsiloxy)silyl)propyloxy)propyl)-2-methyl acrylamide, N-(2-hydroxy-3-(3-(tris(trimethylsiloxy)silyl)propyloxy)propyl) acrylamide, N,N-bis[2-hydroxy-3-(3-(tris(trimethylsiloxy)silyl)propyloxy)propyl]-2-methyl acrylamide, N,N-bis[2-hydroxy-3-(3-(tris(trimethylsiloxy)silyl)propyloxy)propyl] acrylamide, or a combination thereof.
17. The polymerizable composition of any one of embodiments 1 to 16, wherein said at least one siloxane-containing (meth)acrylamido monomer comprises a (meth)acrylamido monomer containing a bis(trialkylsiloxy)alkylsilyl group.
18. The polymerizable composition of embodiment 17, wherein the (meth)acrylamido monomer containing a bis(trialkylsiloxy)alkylsilyl group is N-(2-hydroxy-3-(bis(trimethylsiloxy)methylsilyl)propyloxy)propyl)-2-methyl acrylamide, N-(2-hydroxy-3-(3-(bis(trimethylsiloxy)methylsilyl)propyloxy)propyl) acrylamide, N,N-bis[2-hydroxy-3-(3-(bis(trimethylsiloxy)methylsilyl)propyloxy)propyl]-2-methyl acrylamide, N,N-bis[2-hydroxy-3-(3-(bis(trimethylsiloxy)methylsilyl)propyloxy)propyl] acrylamide, N-[2-hydroxy-3-(3-(t-butylidimethylsilyl)propyloxy)propyl]-2-methyl acrylamide, a siloxane-containing (meth)acrylamido monomer of one of formula (Ia) to (Ih)



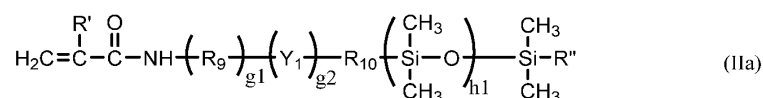


in which R₁₃ is a divalent alkylene radical of -CH₂CH₂-, -CH₂CH₂CH₂-, -



is a divalent alkylene radical of $\begin{array}{c} \text{CH}_3 \\ | \\ -\text{C}- \\ | \\ \text{CH}_3 \end{array}$ or $\begin{array}{c} \text{CH}_3 \\ | \\ -\text{CH}_2-\text{C}- \\ | \\ \text{CH}_3 \end{array}$, R'' and R₁₁ independent of each other is C₁-C₄ alkyl, and f₁ is an integer of from 3 to 5.

19. The polymerizable composition of any one of embodiments 1 to 18, wherein said at least one siloxane-containing (meth)acrylamido monomer comprises a mono-(meth)acrylamido-terminated oligo- or polysiloxane of formula (IIa)

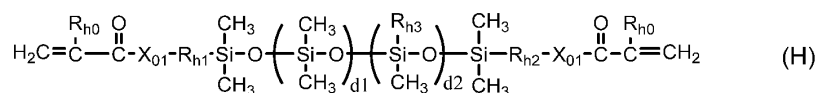


in which R' is hydrogen or methyl, R'' independent of each other is C₁-C₆ alkyl, g₁ and g₂ independent of each other are integer of 0 or 1, h₁ is an integer of 2 to 25, R₉ and R₁₀ independent of each other are a substituted or unsubstituted C₁-C₁₀ alkylene

divalent radical, Y₁ is a linkage of $-\text{NH}-\overset{\text{O}}{\text{C}}-$, $-\overset{\text{O}}{\text{C}}-\text{NH}-$, or $-\overset{\text{R}_{12}'}{\text{C}}-\overset{\text{O}}{\text{C}}-\text{O}-$ in which R₁₂ and R₁₂' independent of each other are C₁-C₆ alkyl.

20. The polymerizable composition of embodiment 19, wherein in formula (IIa) h₁ is an integer of 3 to 20 (more preferably 3 to 15)

21. The polymerizable composition of embodiment 19 or 20, wherein in formula (IIa) R_{12} and R_{12}' independent of each other are methyl.
22. The polymerizable composition of any one of embodiments 1 to 21, wherein said at least one polysiloxane vinylic crosslinker comprises: an α,ω -(meth)acryloxy-terminated polydimethylsiloxane; an α,ω -(meth)acrylamido-terminated polydimethylsiloxane; an α,ω -vinyl carbonate-terminated polydimethylsiloxane; an α,ω -vinyl carbamate-terminated polydimethylsiloxane; a bis-3-methacryloxy-2-hydroxypropyloxypropyl polydimethylsiloxane; N,N,N',N'-tetrakis(3-methacryloxy-2-hydroxypropyl)- α,ω -bis-3-aminopropyl-polydimethylsiloxane of various molecular weight; a reaction product of glycidyl methacrylate with a di-amino-functionalized polydimethylsiloxane; a reaction product of an azlactone-containing vinylic monomer with a d-hydroxyl-functionalized polydimethylsiloxane; or combinations thereof.
23. The polymerizable composition of any one of embodiments 1 to 22, wherein said at least one polysiloxane vinylic crosslinker comprises a vinylic crosslinker of formula (H)



in which:

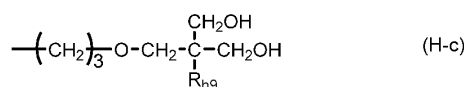
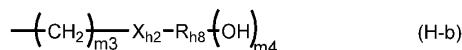
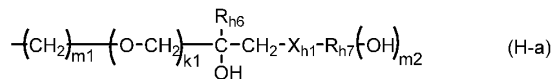
d_1 is an integer of from 30 to 500 and d_2 is an integer of from 1 to 75, provided that d_2/d_1 is from about 0.035 to about 0.15 (preferably from about 0.040 to about 0.12, even more preferably from about 0.045 to about 0.10);

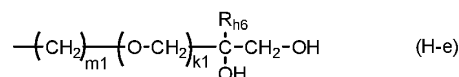
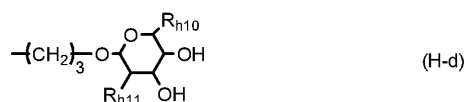
X_{01} is O or NR_{hN} in which R_{hN} is hydrogen or $\text{C}_1\text{-C}_{10}$ -alkyl;

R_{h0} is hydrogen or methyl;

R_{h1} and R_{h2} independently of each other are a substituted or unsubstituted $\text{C}_1\text{-C}_{10}$ alkylene divalent radical or a divalent radical of $-\text{R}_{\text{h4}}-\text{O}-\text{R}_{\text{h5}}-$ in which R_{h4} and R_{h5} independently of each other are a substituted or unsubstituted $\text{C}_1\text{-C}_{10}$ alkylene divalent radical;

R_{h3} is a monovalent radical of any one of formula (H-a) to (H-e)





k1 is zero or 1; m1 is an integer of 2 to 4; m2 is an integer of 1 to 5; m3 is an integer of 3 to 6; m4 is an integer of 2 to 5;

R_{h6} is hydrogen or methyl;

R_{h7} is a C₂-C₆ hydrocarbon radical having (m2+1) valencies;

R_{h8} is a C₂-C₆ hydrocarbon radical having (m4+1) valencies;

R_{h9} is ethyl or hydroxymethyl;

R_{h10} is methyl or hydromethyl;

R_{h11} is hydroxyl or methoxy;

X_{h1} is a sulfur linkage of ---S--- or a tertiary amino linkage of $\text{---NR}_{h12}\text{---}$ in which R_{h12} is C₁-C₁ alkyl, hydroxyethyl, hydroxypropyl, or 2,3-dihydroxypropyl; and

X_{h2} is a linkage of $\text{---NR}_{h13}\text{---}\overset{\text{O}}{\underset{\text{O}}{\text{C}}}\text{---}$ or $\text{---}\overset{\text{O}}{\underset{\text{O}}{\text{C}}}\text{---NR}_{h13}\text{---}$ in which R_{h13} is hydrogen or C₁-C₁₀ alkyl.

24. The polymerizable composition of any one of embodiments 1 to 23, wherein said at least one polysiloxane vinylic crosslinker comprises: (1) a vinylic crosslinker which comprises one sole polydiorganosiloxane segment and two terminal ethylenically-unsaturated groups selected from the group consisting of (meth)acryloyloxy groups, (meth)acryloylamino groups, vinyl carbonate groups, vinylcarbamate groups; and/or (2) a chain-extended polysiloxane vinylic crosslinker which comprises at least two polydiorganosiloxane segment and a covalent linker between each pair of polydiorganosiloxane segments and two two terminal ethylenically-unsaturated groups selected from the group consisting of (meth)acryloyloxy groups, (meth)acryloylamino groups, vinyl carbonate groups, vinylcarbamate groups.
25. The polymerizable composition of any one of embodiments 1 to 24, wherein said at least one polysiloxane vinylic crosslinker comprises α,ω -bis[3-(meth)acrylamidopropyl]-terminated polydimethylsiloxane, α,ω -bis[3-(meth)acryloxypropyl]-terminated polydimethylsiloxane, α,ω -bis[3-(meth)acryloxy-2-hydroxypropyloxypropyl]-terminated polydimethylsiloxane, α,ω -bis[3-(meth)acryloxyethoxy-2-hydroxypropyloxypropyl]-terminated polydimethylsiloxane, α,ω -bis[3-(meth)acryloxypropyloxy-2-hydroxypropyloxypropyl]-terminated polydimethylsiloxane, α,ω -bis[3-(meth)acryloxy-isopropyloxy-2-hydroxypropyloxypropyl]-terminated polydimethylsiloxane, α,ω -bis[3-(meth)acryloxybutyloxy-2-hydroxypropyloxypropyl]-terminated polydimethylsiloxane,

α,ω -bis[3-(meth)acrylamidoethoxy-2-hydroxypropyloxypropyl]-terminated polydimethylsiloxane, α,ω -bis[3-(meth)acrylamidopropoxy-2-hydroxypropyloxypropyl]-terminated polydimethylsiloxane, α,ω -bis[3-(meth)acrylamidoisopropoxy-2-hydroxypropyloxypropyl]-terminated polydimethylsiloxane, α,ω -bis[3-(meth)acrylamidobutoxy-2-hydroxypropyloxypropyl]-terminated polydimethylsiloxane, α,ω -bis[3-(meth)acryloxyethylamino-2-hydroxypropyloxypropyl]-terminated polydimethylsiloxane, α,ω -bis[3-(meth)acryloxypropylamino-2-hydroxypropyloxypropyl]-terminated polydimethylsiloxane, α,ω -bis[3-(meth)acryloxybutylamino-2-hydroxypropyloxypropyl]-terminated polydimethylsiloxane, α,ω -bis[(meth)acrylamidoethylamino-2-hydroxypropyloxypropyl]-terminated polydimethylsiloxane, α,ω -bis[3-(meth)acrylamidopropylamino-2-hydroxypropyloxypropyl]-terminated polydimethylsiloxane, α,ω -bis[3-(meth)acrylamide-butylamino-2-hydroxypropyloxypropyl]-terminated polydimethylsiloxane, α,ω -bis[(meth)acryloxy-2-hydroxypropyloxy-ethoxypropyl]-terminated polydimethylsiloxane, α,ω -bis[(meth)acryloxy-2-hydroxypropyl-N-ethylaminopropyl]-terminated polydimethylsiloxane, α,ω -bis[(meth)acryloxy-2-hydroxypropyl-aminopropyl]-polydimethylsiloxane, α,ω -bis[(meth)acryloxy-2-hydroxypropyloxy-(polyethylenoxy)propyl]-terminated polydimethylsiloxane, α,ω -bis[(meth)acryloxyethylamino-carbonyloxy-ethoxypropyl]-terminated polydimethylsiloxane, α,ω -bis[(meth)acryloxyethylamino-carbonyloxy-(polyethylenoxy)propyl]-terminated polydimethylsiloxane, or combinations thereof.

26. The polymerizable composition of any one of embodiments 1 to 25, wherein said at least one photochromic compound comprises a naphthopyran, an indeno-fused naphthopyran (i.e., indeno-naphthopyran), a heterocyclic ring-fused naphthopyran, a benzopyran, a phenanthropyran, a quinopyran, a quinolinopyran, a fluoroanthropyran, an anthracene-fused pyran, a tetracene-fused pyran, a spiro(benzindoline) naphthopyran, a spiro(indoline)naphthopyran, a spiro(indoline)benzopyran, a spiro(indoline)quinopyran, a spiro(indoline)pyran, a naphthoxazine, a spirobenzopyran, a spirobenzothiopyran, a naphthacenedione, a benzoxazine, a spirooxazine, a naphthoxazine, a spiro(benzindoline)naphthoxazine, a spiro(indoline)naphthoxazine, a spiro(indoline)pyrido-benzoxazine, a spiro(indoline)benzoxazine, a spiro(benzindoline) benzoxazine, a spiro(benzindoline)pyridobenzoxazine, a spiro(indoline) fluoranthoxazine, a spiro(indoline)-quinoxazine, a spiro(piperidine)-naphthopyran, a piro(indoline)pyronobenzoxazinone, a benzospiropyran, a naphthospiropyran, a spirobenzoxazine-pyrrolopyridine, a spironaphthoxazine-pyrrolopyridine, a spiro-

oxazepin-benzoxazine, a spiro-oxazepin-naphthoxazine, a spiro(indoline)benzothiazoloxazine, a spiro(indoline)benzopyrroloxazine, a spiro(indoline)quinazolino-oxazine, a spiro(indoline)-anthracenobenzoxazine, a benzofurobenzopyran, a benzothienobenzopyran, a naphthofurobenzopyran, a benzopyrano-fused naphthopyran, a spiro(isoindoline)-naphthoxazine, a spiro(isoindoline)benzoxazine, or a mixture thereof.

27. The polymerizable composition of embodiment 26, wherein said at least one photochromic compound is polymerizable (i.e., comprising an ethylenically-unsaturated group).
28. The polymerizable composition of embodiment 27, wherein said at least one photochromic compound is a coupling reaction product of a reactive (meth)acrylamido or (meth)acryloxy monomer having a first reactive functional group and a reactive photochromic compound have a second reactive functional group in the absence or presence of a coupling agent under coupling reaction, wherein the first reactive functional group is selected from the group consisting of $-\text{COCl}$, $-\text{COBr}$, $-\text{COOH}$, $-\text{NHR}_{\text{N}_2}$, $-\text{NCO}$, $-\text{OH}$, $-\text{CHO}$, $\text{---}\text{O}$, $\text{---}\text{N}^{\text{R}_0}$, or $\text{---}\text{N}^{\text{R}_0}$, wherein the second reactive functional group is selected from the group consisting of $-\text{COOH}$, $-\text{NHR}_{\text{N}_2}$, $\text{---}\text{N}^{\text{R}_0}$, $\text{---}\text{NH}$, $\text{---}\text{NH}$, $-\text{NCO}$, $-\text{OH}$, $-\text{SH}$, $-\text{CHO}$, $\text{---}\text{O}$, $\text{---}\text{N}^{\text{R}_0}$, or $\text{---}\text{N}^{\text{R}_0}$, wherein R_0 is hydrogen or methyl and R_{N_2} is hydrogen, a linear or branched C_1 - C_{15} alkyl, cyclohexyl, cyclopentyl, a substituted or unsubstituted phenyl, or a substituted- or unsubstituted-phenyl- C_1 - C_6 alkyl.
29. The polymerizable composition of embodiment 27, wherein reactive (meth)acrylamido or (meth)acryloxy monomer is selected from the group consisting of (meth)acryloyl halides ($\text{CH}_2=\text{CH}-\text{COX}$ or $\text{CH}_2=\text{C}(\text{CH}_3)-\text{COX}$, $\text{X}=\text{Cl}$ or Br), N-hydroxysuccinimide ester of (meth)acrylic acid, glycidyl (meth)acrylate, isocyanato- C_2 - C_6 alkyl (meth)acrylate, amino- C_2 - C_6 alkyl (meth)acrylamide, C_1 - C_6 alkylamino- C_2 - C_6 alkyl (meth)acrylamide, (meth)acrylic acid, C_2 - C_4 alkylacrylic acid (e.g., ethylacrylic acid, propylacrylic acid, butylacrylic acid), N-2-acrylamidoglycolic acid, 3-(acryloylxy)propanoic acid, C_2 - C_6 hydroxylalkyl (meth)acrylate, C_2 - C_6 hydroxyalkyl (meth)acrylamide, azlactone-containing vinylic monomers (e.g., 2-vinyl-4,4-dimethyl-1,3-oxazolin-5-one, 2-isopropenyl-4,4-dimethyl-1,3-oxazolin-5-one, 2-vinyl-4-methyl-4-ethyl-1,3-oxazolin-5-one, 2-isopropenyl-4-methyl-4-butyl-1,3-oxazolin-5-one, 2-vinyl-4,4-dibutyl-1,3-oxazolin-5-one, 2-isopropenyl-4-methyl-4-dodecyl-1,3-oxazolin-5-one, 2-isopropenyl-4,4-diphenyl-1,3-oxazolin-5-one, 2-isopropenyl-4,4-pentamethylene-1,3-oxazolin-5-one, 2-isopropenyl-4,4-tetramethylene-1,3-oxazolin-

5-one, 2-vinyl-4,4-diethyl-1,3-oxazolin-5-one, 2-vinyl-4-methyl-4-nonyl-1,3-oxazolin-5-one, 2-isopropenyl-4-methyl-4-phenyl-1,3-oxazolin-5-one, 2-isopropenyl-4-methyl-4-benzyl-1,3-oxazolin-5-one, 2-vinyl-4,4-pentamethylene-1,3-oxazolin-5-one, and 2-vinyl-4,4-dimethyl-1,3-oxazolin-6-one, with 2-vinyl-4,4-dimethyl-1,3-oxazolin-5-one (VDMO) and 2-isopropenyl-4,4-dimethyl-1,3-oxazolin-5-one (IPDMO) as preferred azlactone-containing vinylic monomers), aziridinyl C₁-C₁₂ alkyl (meth)acrylate (e.g., 2-(1-aziridinyl) ethyl (meth)acrylate, 3-(1-aziridinyl) propyl (meth)acrylate, 4-(1-aziridinyl) butyl (meth)acrylate, 6-(1-aziridinyl) hexyl (meth)acrylate, or 8-(1-aziridinyl) octyl (meth)acrylate), acrolein, methacrolein, crotonaldehyde, and combinations thereof.

30. The polymerizable composition of embodiment 28 or 29, wherein the reactive photochromic compound comprises 3,3-bis(4-methoxyphenyl)-6,11-dimethyl-13-hydroxyethoxy-13-phenyl-3H,13H-indeno [2',3':3,4]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-6,11-dimethyl-13-(2-hydroxycarbonylethyl)carboxyethoxy-13-phenyl-3H,13H-indeno [2',3':3,4]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-6,11-dimethyl-13-hydroxy-13-phenyl-3H,13H-indeno [2',3':3,4]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-6,11-dimethyl-13-(2-hydroxycarbonylethyl)carboxy-13-phenyl-3H,13H-indeno [2',3':3,4] naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-6,11-dimethyl-13-hydroxy-13-propyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-6,11-dimethyl-13-(2-hydroxycarbonylethyl)carboxy-13-propyl-3H,13H-indeno[2',3':3,4]-naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-10,11-dimethoxy-13-hydroxy-13-ethyl-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-10,11-dimethoxy-13-(2-hydroxycarbonylethyl)carboxy-13-ethyl-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-7-methoxy-11-phenyl-13-hydroxymethyl-13-methyl-3H,13H-indeno[2',3':3,4] naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-7-methoxy-11-phenyl-13-(2-hydroxycarbonylethyl)carboxymethyl-13-methyl-3H,13H-indeno[2',3':3,4]-naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-7-methoxy-11-phenyl-13-hydroxy-13-methyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-7-methoxy-11-phenyl-13-(2-hydroxycarbonylethyl)carboxy-13-methyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-11-phenyl-13-hydroxy-13-methyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-11-phenyl-13-(2-hydroxycarbonylethyl)carboxy-13-methyl-3H,13H-indeno[2',3':3,4]-naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-6,7,10,11-tetramethoxy-13-hydroxy-13-ethyl-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-6,7,10,11-tetramethyl-13-(2-hydroxycarbonylethyl)carboxy-13-methyl-3H,13H-indeno[2',3':3,4]-naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-6,7-dimethoxy-13-

hydroxy-13-ethyl-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-11-(4-(4,5-diphenyl-1H-imidazol-2-yl)phenyl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-6-methoxy-7-(3-hydroxymethylenepiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-6-methoxy-7-(3-(2-hydroxycarbonylethyl)-carboxymethylenepiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-6,7-dimethoxy-13-hydroxy-13-butyl-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-6,11,13-trimethyl-13-(2-(2-hydroxyethoxy)-ethoxy)-indeno[2,1-f]naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-6,11,13-trimethyl-13-(2-(2-(2-(2-hydroxyethoxy)ethoxy)ethoxy)ethoxy)-indeno[2,1-f]naphtho[1,2-b]pyran; 3,3-diphenyl-13-hydroxy-13-(2-oxo-2-ethoxyethyl)-1H-indeno[2,1-f]naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-6,11-dimethyl-13-hydroxy-13-(2-oxo-2-ethoxyethyl)-1H-indeno[2,1-f]naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-6,11-dimethyl-13-butyl-13-hydroxy-indeno[2,1-f]naphtho[1,2-b]pyran; 3,3-bis(4-hydroxyphenyl)-7-methoxy-11-phenyl-13,13-diethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-bis(4-hydroxyphenyl)-7-methoxy-11-phenyl-13,13-dipropyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-bis(4-hydroxyphenyl)-7-methoxy-11-phenyl-13-carbomethoxy-13-methyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-diphenyl-6,7-dimethoxy-13-methyl-13-methyl-13-hydroxyethoxy-ethoxyethoxy-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-diphenyl-6,7,10,11-tetramethoxy-13-ethyl-13-methyl-13-hydroxyethoxyethoxy-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 6,7-dimethoxy-2,2-diphenyl-13-hydroxy-13-butyl-2H,13H-indeno[1',2':4,3]naphtho[1,2-b]pyran; 6,7-dimethoxy-2,2-diphenyl-13-hydroxy-13-methyl-2H,13H-indeno[1',2':4,3]naphtho[1,2-b]pyran; 6,7-dimethoxy-2,2-diphenyl-13-hydroxy-2H,13H-indeno[1',2':4,3]naphtho[1,2-b]pyran; 3-(4-allyloxyphenyl)-3-(4-morpholinophenyl)-7-methoxy-11-phenyl-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-fluorophenyl)-3-(4-butoxyphenyl)-10-(piperazin-1-yl)-6-trifluoromethyl-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-fluorophenyl)-3-(4-butoxyphenyl)-10-(4-hydroxybenzamido)-6-trifluoromethyl-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-fluorophenyl)-3-(4-butoxyphenyl)-10-vinyl-6-trifluoromethyl-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-fluorophenyl)-3-(4-butoxyphenyl)-10-(carboxylic acid)-6-trifluoromethyl-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-fluorophenyl)-3-(4-methoxyphenyl)-6-methoxy-7-(4-hydroxypiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-fluorophenyl)-3-(4-methoxyphenyl)-6-methoxy-7-(4-(2-hydroxycarbonylethyl) carboxypiperidin-1-yl)-13,13-dimethyl-

3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-fluorophenyl)-3-(4-morpholinophenyl)-6-methoxy-7-(4-hydroxypiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-fluorophenyl)-3-(4-morpholinophenyl)-6-methoxy-7-(4-(2-hydroxycarbonylethyl) carboxypiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4] naphtho[1,2-b]pyran; 3-(4-hydroxyethoxyphenyl)-3-(4-methoxyphenyl)-11-(4-(N,N-dimethylamino)phenyl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-methoxyphenyl)-3-(2,4-dimethoxyphenyl)-phenyl-6,11-dimethoxy-13-methyl-13-methyl-13-hydroxyethoxyethoxyethoxy-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-methoxyphenyl)-3-(3-methyl-4-methoxyphenyl)-13-hydroxy-indeno[2,1-f]naphtho[1,2-b]pyran; 3-(4-methoxyphenyl)-3-(4-morpholinophenyl)-6,7,10,11-tetramethoxy-13-hydroxy-13-ethyl-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3-(4-methoxyphenyl)-3-(4-morpholinophenyl)-6,7,10,11-tetramethoxy-13-hydroxy-13-butyl-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3-(4-methoxyphenyl)-3-(4-morpholinophenyl)-6,7-dimethoxy-13-hydroxy-13-ethyl-3H,13H-indeno [2,1-f]naphtho[1,2-b]pyran; 3-(4-methoxyphenyl)-3-(4-morpholinophenyl)-6,7-dimethoxy-13-hydroxy-13-methyl-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3-(4-methoxyphenyl)-3-(4-morpholinophenyl)-13-oxo-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3-(4-methoxyphenyl)-3-phenyl-6-methoxy-7-mprphlino-13-methyl-13-methyl-13-hydroxyethoxyethoxyethoxy-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-methoxyphenyl)-3-phenyl-6,11-dimethoxy-13-methyl-13-methyl-13-hydroxyethoxy-ethoxy-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 2-(4-methoxyphenyl)-2-(4-morpholinophenyl)-5-hydroxy-6-carboethoxy-2H-naphtho[1,2-b] pyran; 3-(4-morpholinophenyl)-3-phenyl-6,7-dimethoxy-13-hydroxy-13-ethyl-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3-(4-morpholinophenyl)-3-phenyl-6,7-dimethoxy-13-hydroxy-13-butyl-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3-(4-morpholinophenyl)-3-phenyl-6,7-dimethoxy-13-oxo-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3-phenyl-3-(4-hydroxyethoxyphenyl)-6-methoxy-7-morpholino-13,13-dimethyl-3H,13H-indeno-[2',3':3,4] naphtho[1,2-b]pyran; 3-phenyl-3-(4-(2-hydroxycarbonylethyl)carboxyethoxy-phenyl)-6-methoxy-7-morpholino-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-(2-hydroxyethoxy)phenyl)-6-methoxy-7-piperidino-13,13-dimethy-3H,13H-indene[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-(2-(2-hydroxycarbonylethyl)-carboxyethoxy)phenyl)-6-methoxy-7-piperidino-13,13-dimethy-3H,13H-indene[2',3':3,4] naphtho[1,2-b]pyran; 3-phenyl-3-(4-hydroxyphenyl)-6,7-dimethoxy-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[l,2-b]pyran; 3-phenyl-3-(4-hydroxyethoxy)phenyl)-6,7-dimethoxy-13,13-dimethyl-3H,13H-indeno[2',3':3,4]-naphtho[1,2-b]pyran; 3-phenyl-3-(4-(2-hydroxycarbonylethyl)carboxyethoxy)phenyl)-

6,7-dimethoxy-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-(2-hydroxycarbonylethyl)carboxy-phenyl)-6,7-dimethoxy-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-(2-hydroxyethyl)piperazin-1-yl)phenyl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-(2-(2-hydroxycarbonylethyl)-carboxyethyl)piperazin-1-yl)phenyl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-methoxyphenyl)-6-methoxy-7-piperazinyl-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-methoxyphenyl)-6-methoxy-7-(3-hydroxymethylenepiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-methoxyphenyl)-6-methoxy-7-(3-(2-hydroxycarbonylethyl) carboxymethylenepiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-methoxyphenyl)-6-methoxy-7-(4-hydroxypiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-methoxyphenyl)-6-methoxy-7-(4-(2-hydroxycarbonylethyl)-carboxypiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-(4-methoxyphenyl-piperazin-1-yl)phenyl)-13,13-dimethyl-6-methoxy-7-hydroxy-indeno [2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-(4-methoxyphenyl-piperazin-1-yl)phenyl)-13,13-dimethyl-6-methoxy-7-(2-hydroxy-2-methyl-3-butyn-4-yl)-indeno[2',3':3,4]naphtho [1,2-b]pyran; 3-phenyl-3-(4-morpholinophenyl)-6-methoxy-7-(3-hydroxymethylene-piperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-morpholinophenyl)-6-methoxy-7-(3-(2-hydroxycarbonyl-ethyl)carboxymethylene-piperidin-1-yl)-13,13-dimethyl-3H,13H-indeno [2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-morpholinophenyl)-6-methoxy-7-(3-(2-hydroxycarbonylethyl)carboxymethylene-piperidin-1-yl)-13,13-dimethyl-3H,13H-indeno [2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-morpholinophenyl)-6-methoxy-7-(4-hydroxypiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-morpholinophenyl)-6-methoxy-7-(4-(2-hydroxycarbonylethyl)-carboxypiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4] naphtho[1,2-b]pyran; 3-phenyl-3-(4-morpholinophenyl)-6-methoxy-7-piperazinyl-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho [1,2-b]pyran; 3-phenyl-3-(4-morpholinophenyl)-6,11-dimethyl-13-hydroxy-13-(1-oxo-methoxyprop-2-yl)-1H-indeno[2,1-f]naphtho[1,2-b]pyran; 3-phenyl-3-(4-(4-phenylpiperazino)-phenyl)-6-methoxy-7-(4-hydroxypiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4] naphtho[1,2-b]pyran; 3-phenyl-3-(4-(4-phenylpiperazino)phenyl)-6-methoxy-7-(4-(2-hydroxycarbonylethyl) carboxypiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4] naphtho[1,2-b]pyran; 3-phenyl-3-(4-piperazinylphenyl)-6,11-dimethoxy-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho-[1,2-b]pyran; 3-phenyl-3-(4-

piperazinylphenyl)-6-methoxy-7-piperidinyl-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-(3-methylpiperazin-1-yl)phenyl)-6-methoxy-7-(decahydroisoquinolin-2-yl)-11-*tert*-butyl-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-methoxyphenyl)-3-(4-morpholinophenyl)-16-(ethoxycarbonyl)methyl-16-hydroxy-3,16-dihydrobenzofuro-[2',3':7,8]indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-methoxyphenyl)-16-(ethoxycarbonyl)-methyl-16-hydroxy-3,16-dihydrobenzofuro [2'',3'':6',7']indeno-[3',2':4,3]naphtho[1,2-b]pyran; 3-phenyl-3-(4-morpholinophenyl)-16-(ethoxycarbonyl)-methyl-16-hydroxy-3,16-dihydrobenzofuro[2'',3'':6',7']indeno[3',2':4,3] naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-16-(ethoxycarbonyl)methyl-16-hydroxy-3,16-dihydro-benzofuro[2',3':7,8]indeno[2',3':3,4] naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-16-hydroxy-16-ethyl-16H-benzofuro[2',3':7,8]indeno[2',3':3,4]naphtho [1,2-b]pyran; 3,3-di(4-methoxyphenyl)-16-hydroxy-16H-benzofuro [2'',3'':6,7]indeno [3',2':4,3]naphtho-[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-16-hydroxy-16-ethyl-16H-benzofuro[2'',3'':6,7]indeno[3',2':4,3]naphtho[1,2-b]pyran; 3-(4-methoxyphenyl)-3-(2,3-dihydrobenzofur-5-yl)-13-hydroxy-13-methyl-indeno[2,1-f]naphtho[1,2-b]pyran; 3-(4-methoxyphenyl)-3-(2,3-dihydrobenzofur-5-yl)-6,11-difluoro-13-hydroxy-13-(1-oxo-methoxyprop-2-yl)-1H-indeno[2,1-f]naphtho[1,2-b]pyran, or a mixture thereof.

31. The polymerizable composition of embodiment 28 or 29, wherein the reactive photochromic compound is 2,2-di(4-fluorophenyl)-5-hydroxycarbonyl-6-phenyl-9-methoxy-2H-naphtho[1,2-b]pyran; 2,2-bis(4-methylphenyl)-5-methoxycarbonyl-6-hydroxy-2H-naphtho[1,2-b]pyran; 2,2-bis(4-methoxyphenyl)-5-methoxycarbonyl-6-hydroxy-[2H]-naphtho[1,2-b]pyran; 2,2-bis(4-methoxyphenyl)-5-methoxyethoxycarbonyl-6-methyl-8-vinyl-2H-naphtho[1,2-b]pyran; 2,2-bis(4-methoxyphenyl)-5-methoxyethoxycarbonyl-6-methyl-8-hydroxycarbonyl-2H-naphtho [1,2-b]pyran; 2,2-bis(4-methoxyphenyl)-5-methoxycarbonyl-6-(2-hydroxyethoxy)ethoxy-[2H]-naphtho[1,2-b]pyran; 2,2-bis(4-methoxyphenyl)-5-methoxycarbonyl-6-phenyl-9-(2-hydroxyethoxy)-[2H]-naphtho[1,2-b]pyran; 2,2-bis(4-methoxyphenyl)-5-methoxy-carbonyl-6-(4-(2-hydroxyethoxy)phenyl)-[2H]-naphtho[1,2-b]pyran; 2,2-bis(4-methoxyphenyl)-5-diphenylmethylol-6-hydroxy-2H-naphtho[1,2-b]pyran; 2,2-bis(4-methoxyphenyl)-5-diphenylmethylol-6-methoxy-2H-naphtho[1,2-b]pyran; 2,2-bis(4-methoxyphenyl)-5-(2-hydroxyethoxycarbonyl)-6-phenyl-[2H]-naphtho[1,2-b]pyran; 2,2-bis(4-methoxyphenyl)-5-(2-(2-hydroxyethoxy)-ethoxycarbonyl)-6-phenyl-[2H]-naphtho[1,2-b]pyran; 2,2-bis(4-methoxyphenyl)-5-(2-(2-(2-hydroxyethoxy)ethoxy)-ethoxycarbonyl)-6-phenyl-[2H]-naphtho[1,2-b]pyran; 2,2-bis(4-methoxyphenyl)-5-(2-(2-(2-(2-hydroxyethoxy)ethoxy)ethoxy)-ethoxy)-ethoxy)-ethoxy)-6-phenyl-[2H]-naphtho[1,2-b]pyran; or a mixture thereof.

ethoxycarbonyl)-6-phenyl-[2H]-naphtho[1,2-b]pyran; 2,2-bis(4-methoxyphenyl)-5-hydroxymethyl-6-methyl-9-methoxy-2H-naphtho[1,2-b]pyran; 2,2-bis(4-methoxyphenyl)-5-hydroxymethyl-6-phenyl-2H-naphtho[1,2-b]pyran; 2,2-bis(4-methoxyphenyl)-5-hydroxymethyl-6-methoxy-2H-naphtho[1,2-b]pyran; 2,2-bis(4-methoxyphenyl)-5-methoxycarbonyl-6-(3-aminophenyl)-2H-naphtho[1,2-b]pyran; 2,2-bis(4-methoxyphenyl)-2,5,7-trihydro-7-methylidene-5-oxo-furo-[3',4':3,4]naphtho[1,2-b]pyran; 2,2-bis(4-methoxyphenyl)-5-phenylthio-6-hydroxy-2H-naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-13-hydroxy-13-methyl-indeno[2,1-f]naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-9-methoxycarbonyl-8-(2-hydroxyethoxy)ethoxy-[3H]-naphtho[1,2-b]pyran; 2,2-diphenyl-5-hydroxycarbonyl-9-methoxy-2H-naphtho[1,2-b]pyran; 2,2-diphenyl-5-hydroxycarbonyl-8,9-dimethoxy-2H-naphtho[1,2-b]pyran; 2,2-diphenyl-5-hydroxy-6-carboethoxy-2H-naphtho[1,2-b]pyran; 2,2-diphenyl-5-hydroxy-6-morpholinocarbonyl-2H-naphtho[1,2-b]pyran; 2,2-diphenyl-5-hydroxy-6-carboethoxy-2H-naphtho[1,2-b]pyran; 2,2-diphenyl-5-hydroxy-6-carbomethoxy-9-methoxy-2H-naphtho[1,2-b]pyran; 2,2-diphenyl-5-hydroxy-6-morpholinocarbonyl-2H-naphtho[1,2-b]pyran; 2,2-diphenyl-5-methylol-6-(3-dimethylaminopropyl)methylamino-2H-naphtho[1,2-b]pyran; 2,2-diphenyl-2,5,7-trihydro-7-methylidene-5-oxofuro-[3',4':3,4]naphtho[1,2-b]pyran; 2,2-spiroadamantylene-5-phenylthio-6-hydroxy-2H-naphtho[1,2-b]pyran; 2,2-diphenyl-5-methoxycarbonyl-6-hydroxy-2H-naphtho[1,2-b]pyran; 2,2-spiro-admantylene-5-methoxycarbonyl-6-hydroxy-2H-naphtho[1,2-b]pyran; 3,3-diphenyl-8-hydroxy-9-carbopropoxy-3H-naphtho[2,1-b]pyran; 2,2,5-triphenyl-6-carboethoxy-2H-naphtho[1,2-b]pyran; 2,2,6-triphenyl-5-(2-(2-(2-hydroxyethoxy)ethoxy)-ethoxy)ethoxy)carbonyl-[2H]-naphtho[1,2-b]pyran; 2,2,6-triphenyl-5-(2-(2-(2-oxiran-2-ylmethoxy)ethoxy)ethoxy)-ethoxycarbonyl-[2H]-naphtho[1,2-b]pyran; 2-(4-methoxyphenyl)-2-(4-morpholinophenyl)-5-hydroxy-6-carboethoxy-2H-naphtho[1,2-b]pyran; 2-(4-methoxyphenyl)-2-tert-butyl-5-methoxy-carbonyl-6-hydroxy-2H-naphtho[1,2-b]pyran; 2-(4-methoxyphenyl)-2-phenyl-5-methoxycarbonyl-6-hydroxy-2H-naphtho[1,2-b]pyran; 2-(4-(2-(2-hydroxyethoxy)ethoxy)-ethoxyphenyl)-2-phenyl-5-methoxycarbonyl-6-methyl-9-methoxy-[2H]-naphtho[1,2-b]pyran; 2-(4-(2-(2-hydroxyethoxy)ethoxy)phenyl)-2-phenyl-5-methoxycarbonyl-6-(2-(2-methylprop-2-enoxyl)ethoxy)-[2H]-naphtho[1,2-b]pyran; 3-(4-(2-(2-hydroxyethoxy)-ethoxy)ethoxyphenyl)-3-phenyl-9-methoxycarbonyl-8-methoxy-[3H]-naphtho[1,2-b]pyran; 2-(4-morpholinophenyl)-2-phenyl-5-methoxycarbonyl-6-hydroxy-2H-naphtho[1,2-b]pyran; 2-(9-ethylcarbazol-2-yl)-2-phenyl-5-methoxycarbonyl-6-hydroxy-2H-naphtho[1,2-b]pyran; 2-(9-phenylcarbazol-2-yl)-2-phenyl-5-methoxycarbonyl-6-hydroxy-2H-naphtho[1,2-b]pyran; 2-(4-dimethylaminophenyl)-2-

phenyl-5-methoxycarbonyl-6-hydroxy-2H-naphtho[1,2-b]pyran; 3-(2-fluorophenyl)-3-(4-methoxyphenyl-8-hydroxy-9-carbopropoxy-3H-naphtho[2,1-b]pyran; 3-(2-fluorophenyl)-3-(4-methoxyphenyl-8-hydroxy-9-carbomethoxy-3H-naphtho[2,1-b]pyran; 3-(2,4-dimethoxyphenyl)-3-(4-methoxyphenyl-8-hydroxy-9-carbomethoxy-3H-naphtho[2,1-b]pyran; 3-(2,4,6-trifluorophenyl)-3-(2,4,6-trimethoxy-1-naphthyl)-8-acetyl-9-carbonyl-3H-naphtho[2,1-b]pyran; 3-(4-methoxyphenyl)-3-(2-methyl-2,3-dihydrobenzofur-5-yl)-6-methoxy-12-hydroxymethyl-11-phenyl-3H-phenanthro[1,2-b]pyran; 5,5-bis(4-methoxyphenyl)-8-methylol-5H-fluorantheno[3,2-b]pyran; or a mixture thereof.

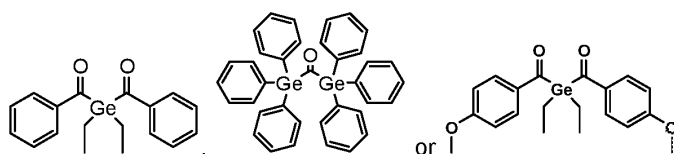
32. The polymerizable composition of embodiment 28 or 29, wherein the reactive photochromic compound is 1-hydroxyethyl-3,3-dimethylspiro[indoline-2,3'-[3H]-naphtho[2,1-b][1,4]oxazine]; 1-hydroxypropyl-3,3-dimethylspiro[indoline-2,3'-[3H]-naphtho[2,1-b][1,4]oxazine]; 1-aminoethyl-3,3-dimethyl-5-chlorospiro[indoline-2,3'-[3H]-naphtho[2,1-b][1,4]oxazine]; 1-hydroxyethyl-3,3-dimethyl-8'-methoxyspiro[indoline-2,3'-[3H]-naphtho[2,1-b][1,4]oxazine]; 1-(p-vinylphenyl)-3,3-dimethyl-5,6-dichlorospiro[indoline-2,3'-[3H]-naphtho[2,1-b][1,4]oxazine]; 1,3,3-trimethyl-9'-hydroxyspiro[indoline-2,3'-[3H]-naphtho[2,1-b][1,4]oxazine]; 1,3,3-trimethyl-5'-hydroxymethylspiro[indoline-2,3'-[3H]-naphtho[2,1-b][1,4]oxazine]; 1,3,3-trimethyl-9'-aminospiro[indoline-2,3'-[3H]-naphtho[2,1-b][1,4]oxazine]; 1,3,3-trimethyl-5-chloro-8'-hydroxyspiro[indoline-2,3'-[3H]-naphtho[2,1-b][1,4]oxazine]; 1-benzyl-3,3-dimethyl-9'-vinylbenzoyloxyspiro[indoline-2,3'-[3H]-naphtho[2,1-b][1,4]oxazine]; 1-benzyl-3,3-dimethyl-5'-hydroxymethylpiperidinospironaphthooxazine; 1,3,3-trimethyl-9'-hydroxyspiro[indoline-2,3'-[3H]-pyrido-[2,1-b][1,4]benzooxazine]; 5-hydroxy-6'-cyano-1,3,3-trimethyl-spiro-[indoline-2,3'-[3H]-naphtho[2,1-b][1,4]oxazine]; 5-hydroxy-6'-phenylsulfonyl-1,3,3-trimethyl-spiro-[indoline-2,3'-[3H]-naphtho[2,1-b][1,4]oxazine]; 5'-formyl-1,3,3-trimethylspiro[indoline-2,3'-[3H]-naphtho[2,1-b][1,4]oxazine]; 1,3-dihydro-6'-piperazino-1,3,3-trimethyl spiro [2H indole-2,3'-[3H]-naphtho[2,1-b][1,4]oxazine]; 1,3-dihydro-6'-(4-hydroxyethyl)piperazino-1,3,3-trimethyl spiro [2H indole-2,3'-[3H]-naphtho[2,1-b][1,4]oxazine]; 1,3-dihydro-9'-hydroxy-1,3,3-trimethyl spiro 92H indole-2,3'-[3H]-naphtho[2,1-b][1,4] oxazine; 1,3,3,4,5-pentamethyl-9'-methoxycarbonyl-8'-hydroxy-spiro[indoline-2,3'-[3H]naphth[2,1-b][1,4]oxazine; 1,3,3,5,6-pentamethyl-9'-methoxycarbonyl-8'-hydroxy-spiro[indoline-2,3'-[3H]naphth[2,1-b][1,4]oxazine; 1-propyl-3,3,4,5-tetramethyl-9'-methoxycarbonyl-8'-hydroxy-spiro[indoline-2,3'-[3H]naphth[2,1-b][1,4]oxazine; 1-propyl-3,3,5,6-tetramethyl-9'-methoxycarbonyl-8'-hydroxy-spiro[indoline-2,3'-[3H]naphth[2,1-b][1,4]oxazine; 1-methoxyethyl-3,3-dimethyl-9'-allyloxycarbonyl-8'-chloroactoxy-spiro[indoline-2,3'-[3H]naphth[2,1-b][1,4]oxazine; 1-allyl-3,3-spirocyclohexyl-9'-

benzyloxycarbonyl-8'-chloroacetoxy-spiro[indoline-2,3'-[3H]naphtho[2,1-b][1,4]oxazine;
 5-amino-1,3,3-trimethyl-spiro[indoline-2,3'-[3H]-pyrido[3,4-f][1,4]benzoxazine; 6-
 hydroxy-1',3',3',4',5'-pentamethylspiro[2H-1,4-benzoxazine-2,2'-indoline]; 6-hydroxy-
 1',3',3',5',6'-pentamethylspiro[2H-1,4-benzoxazine-2,2'-indoline]; 5,7-dimethoxy-1'-
 hydroxy-carbonylethyl-3',3'-dimethylspiro[2H-1,4-benzoxazine-2,2'-indoline]; 7-
 methoxy-1'-hydroxy-ethyl-3',3'-dimethylspiro[2H-1,4-benzoxazine-2,2'-indoline]; 9'-
 hydroxy-1-methylspiro[indoline-2,3'-(3H)-naphtho(2,1-b)-1,4-oxazine]; 5-chloro-9'-
 hydroxy-1-methylspiro[indoline-2,3'-(3H)-naphtho(2,1-b)-1,4-oxazine]; 8'-hydroxy-1-
 methylspiro[indoline-2,3'-(3H)-naphtho(2,1-b)-1,4-oxazine]; 8'-hydroxy-5-methoxy-1-
 methylspiro[indoline-2,3'-(3H)-naphtho(2,1-b)-1,4-oxazine]; 8'-hydroxy-1,4,5-
 trimethylspiro[indoline-2,3'-(3H)-naphtho(2,1-b)-1,4-oxazine]; 9'-hydroxy-1-
 isopropylspiro[indoline-2,3'-(3H)-naphtho(2,1-b)-1,4-oxazine]; 9'-hydroxy-1-
 hexadecyl-spiro[indoline-2,3'-(3H)-naphtho(2,1-b)-1,4-oxazine]; 9'-hydroxy-1-
 octadecylspiro[indoline-2,3'-(3H)-naphtho(2,1-b)-1,4-oxazine]; 1,1''-(1,5-
 pentanediyl)bis[3,3-dimethyl]-9'-hydroxy-spiro[indoline-2,3'-[3H]-naphtho[2,1-b]
 [1,4]oxazine]; 1,1''-[1,4-phenylenebis(methylene)] bis[3,3-dimethyl]-5'-
 hydroxymethylspiro[indoline-2,3'-[3H]-naphtho[2,1-b] [1,4]oxazine]; 1,1''-(1,4-
 butanediyl)bis[5,6-dichloro-3,3-dimethyl]-8'-hydroxyspiro[indoline-2,3'-[3H]-
 naphtho[2,1-b] [1,4]oxazine]; 1,3,3-trimethyl-9'-
 hydroxypiperidinospironaphthooxazine; 3-carboxyethyl-1,1-dimethyl-9'-
 methoxyspiro[benz[e]-indoline-2,3'-[3H]-naphtho[2,1-b][1,4]oxazine]; 1-carboxyethyl-
 3,3-dimethyl-9'-methoxyspiro[benz[g]-indoline-2,3'-[3H]-naphtho[2,1-b][1,4]oxazine];
 1,1-dimethyl-3-hydroxyethyl-9'-methoxyspiro[benz[e]-indoline-2,3'-[3H]-naphtho[2,1-
 b][1,4]oxazine]; 3,3-dimethyl-1-hydroxyethyl-9'-methoxyspiro[benz[g]-indoline-2,3'-
 [3H]-naphtho[2,1-b][1,4]oxazine]; 3-carboxyethyl-1,1-dimethyl-9'-
 methoxyspiro[benz[e]-indoline-2,3'-[3H]-pyrido[2,1-b][1,4]benzoxazine]; 1-
 carboxyethyl-3,3-dimethyl-9'-methoxyspiro[benz[g]-indoline-2,3'-[3H]-pyrido[2,1-
 b][1,4]benzoxazine]; 1,1-dimethyl-3-hydroxyethyl-9'-methoxyspiro[benz[e]-indoline-
 2,3'-[3H]-pyrido[2,1-b][1,4]benzoxazine]; 3,3-dimethyl-1-hydroxyethyl-9'-
 methoxyspiro[benz[g]-indoline-2,3'-[3H]-pyrido[2,1-b][1,4]benzoxazine]; 5-amino-5'-
 (2-benzthiazolyl)-1,3,3-trimethylspiro[indoline-2,3'-[3H]-naphtho[2,1-b][1,4]oxazine];
 5-amino-5'-(5'-(2-hydroxyphenyl)-2-oxadiazolyl)-1-isopropyl-3,3-
 dimethylspiro[indoline-2,3'-[3H]-naphtho[2,1-b][1,4]oxazine]; 5-amino-5'-(5'-phenyl-2-
 oxadiazolyl)-1,3,3-trimethylspiro[indoline-2,3'-[3H]-naphtho[2,1-b][1,4]oxazine]; or a
 mixture thereof.

33. The polymerizable composition of embodiment 28 or 29, wherein the reactive
 photochromic compound is 3,3-bis(4-methoxyphenyl)-6,11-dimethyl-13-

hydroxyethoxy-13-phenyl-3H,13H-indeno [2',3':3,4]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-6,11-dimethyl-13-(2-hydroxycarbonylethyl)carboxyethoxy-13-phenyl-3H,13H-indeno [2',3':3,4]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-6,11-dimethyl-13-hydroxy-13-phenyl-3H,13H-indeno [2',3':3,4]naphtho[1,2-b]pyran; or a mixture thereof.

34. The polymerizable composition of any one of embodiments 1 to 33, wherein said at least one acylgermanium photoinitiator comprises any one of

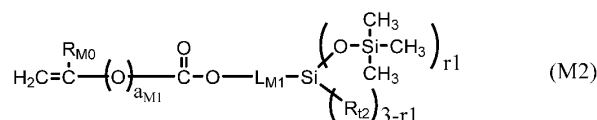
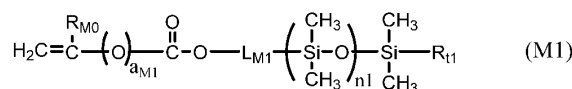


35. The polymerizable composition of any one of embodiments 1 to 34, wherein the polymerizable composition comprises at least one UV-absorbing vinylic monomer and/or at least one UV/HEVL-Absorbing vinylic monomer.
36. The polymerizable composition of embodiment 35, wherein the polymerizable composition comprises 2-[2'-hydroxy-5'-(2-methacryloxyethyl)phenyl]-2H-benzotriazole (Norbloc).
37. The polymerizable composition of any one of embodiments 1 to 36, wherein the polymerizable composition further comprise about 10% by weight or less of additional polymerizable components selected from the group consisting of one or more secondary hydrophilic vinylic monomers (i.e., hydrophilic vinylic monomers other than hydrophilic (meth)acrylamido monomer), one or more secondary siloxane-containing vinylic monomers (i.e., siloxane-containing vinylic monomers other than siloxane-containing acrylamido monomer), one or more hydrophobic vinylic monomers, one or more non-silicone vinylic crossliners, and combinations thereof.
38. The polymerizable composition of embodiment 37, wherein the polymerizable composition further comprise at least one secondary hydrophilic vinylic monomer which comprises:
- (1) a hydroxyl-containing (meth)acryloxy monomer selected from the group consisting of 2-hydroxyethyl (meth)acrylate, 3-hydroxypropyl (meth)acrylate, 2-hydroxypropyl (meth)acrylate, glycerol methacrylate (GMA), di(ethylene glycol) (meth)acrylate, tri(ethylene glycol) (meth)acrylate, tetra(ethylene glycol) (meth)acrylate, poly(ethylene glycol) (meth)acrylate having a number average molecular weight of up to 1500, and combinations thereof;
 - (2) a carboxyl-containing (meth)acryloxy monomer selected from the group consisting of (meth)acrylic acid, ethylacrylic acid, and combinations thereof;
 - (3) an amino-containing (meth)acryloxy monomer selected from the group consisting of

- 2-aminoethyl (meth)acrylate, 2-methylaminoethyl (meth)acrylate, 2-ethylaminoethyl (meth)acrylate, 3-aminopropyl (meth)acrylate, 3-methylaminopropyl (meth)acrylate, 3-ethylaminopropyl (meth)acrylate, 3-amino-2-hydroxypropyl (meth)acrylate, trimethylammonium 2-hydroxy propyl (meth)acrylate hydrochloride, dimethylaminoethyl (meth)acrylate, and combinations thereof;
- (4) an N-vinyl amide monomer selected from the group consisting of N-vinylpyrrolidone, N-vinyl-3-methyl-2-pyrrolidone, N-vinyl-4-methyl-2-pyrrolidone, N-vinyl-5-methyl-2-pyrrolidone, N-vinyl-6-methyl-2-pyrrolidone, N-vinyl-3-ethyl-2-pyrrolidone, N-vinyl-4,5-dimethyl-2-pyrrolidone, N-vinyl-5,5-dimethyl-2-pyrrolidone, N-vinyl-3,3,5-trimethyl-2-pyrrolidone, N-vinyl piperidone (*aka*, N-vinyl-2-piperidone), N-vinyl-3-methyl-2-piperidone, N-vinyl-4-methyl-2-piperidone, N-vinyl-5-methyl-2-piperidone, N-vinyl-6-methyl-2-piperidone, N-vinyl-6-ethyl-2-piperidone, N-vinyl-3,5-dimethyl-2-piperidone, N-vinyl-4,4-dimethyl-2-piperidone, N-vinyl caprolactam (*aka*, N-vinyl-2-caprolactam), N-vinyl-3-methyl-2-caprolactam, N-vinyl-4-methyl-2-caprolactam, N-vinyl-7-methyl-2-caprolactam, N-vinyl-7-ethyl-2-caprolactam, N-vinyl-3,5-dimethyl-2-caprolactam, N-vinyl-4,6-dimethyl-2-caprolactam, N-vinyl-3,5,7-trimethyl-2-caprolactam, N-vinyl-N-methyl acetamide, N-vinyl formamide, N-vinyl acetamide, N-vinyl isopropylamide, N-vinyl-N-ethyl acetamide, N-vinyl-N-ethyl formamide, and mixtures thereof;
- (5) a methylene-containing pyrrolidone monomer selected from the group consisting of 1-methyl-3-methylene-2-pyrrolidone, 1-ethyl-3-methylene-2-pyrrolidone, 1-methyl-5-methylene-2-pyrrolidone, 1-ethyl-5-methylene-2-pyrrolidone, 5-methyl-3-methylene-2-pyrrolidone, 5-ethyl-3-methylene-2-pyrrolidone, 1-n-propyl-3-methylene-2-pyrrolidone, 1-n-propyl-5-methylene-2-pyrrolidone, 1-isopropyl-3-methylene-2-pyrrolidone, 1-isopropyl-5-methylene-2-pyrrolidone, 1-n-butyl-3-methylene-2-pyrrolidone, 1-tert-butyl-3-methylene-2-pyrrolidone, and combinations thereof;
- (6) a (meth)acryloxy monomer having a C₁-C₄ alkoxyethoxy group and selected from the group consisting of ethylene glycol methyl ether (meth)acrylate, di(ethylene glycol) methyl ether (meth)acrylate, tri(ethylene glycol) methyl ether (meth)acrylate, tetra(ethylene glycol) methyl ether (meth)acrylate, C₁-C₄-alkoxy poly(ethylene glycol) (meth)acrylate having a number average molecular weight of up to 1500, and combinations thereof;
- (7) a vinyl ether monomer selected from the group consisting of ethylene glycol monovinyl ether, di(ethylene glycol) monovinyl ether, tri(ethylene glycol) monovinyl ether, tetra(ethylene glycol) monovinyl ether, poly(ethylene glycol) monovinyl ether, ethylene glycol methyl vinyl ether, di(ethylene glycol) methyl vinyl ether, tri(ethylene

- glycol) methyl vinyl ether, tetra(ethylene glycol) methyl vinyl ether, poly(ethylene glycol) methyl vinyl ether, and combinations thereof;
- (8) an allyl ether monomer selected from the group consisting of ethylene glycol monoallyl ether, di(ethylene glycol) monoallyl ether, tri(ethylene glycol) monoallyl ether, tetra(ethylene glycol) monoallyl ether, poly(ethylene glycol) monoallyl ether, ethylene glycol methyl allyl ether, di(ethylene glycol) methyl allyl ether, tri(ethylene glycol) methyl allyl ether, tetra(ethylene glycol) methyl allyl ether, poly(ethylene glycol) methyl allyl ether, and combinations thereof;
- (9) a phosphorylcholine-containing vinylic monomer selected from the group consisting of (meth)acryloyloxyethyl phosphorylcholine, (meth)acryloyloxypropyl phosphorylcholine, 4-((meth)acryloyloxy)butyl-2'-(trimethylammonio)ethylphosphate, 2-[(meth)acryloylamino]ethyl-2'-(trimethylammonio)-ethylphosphate, 3-[(meth)acryloylamino]propyl-2'-(trimethylammonio)ethylphosphate, 4-[(meth)acryloyl-amino]butyl-2'-(trimethylammonio)ethylphosphate, 5-((meth)acryloyloxy)pentyl-2'-(trimethylammonio)ethyl phosphate, 6-((meth)acryloyloxy)hexyl-2'-(trimethylammonio)ethylphosphate, 2-((meth)acryloyloxy)ethyl-2'-(triethylammonio)-ethylphosphate, 2-((meth)acryloyloxy)ethyl-2'-(tripropylammonio)ethylphosphate, 2-((meth)acryloyloxy)ethyl-2'-(tributylammonio)ethyl phosphate, 2-((meth)acryloyloxy)-propyl-2'-(trimethylammonio)-ethylphosphate, 2-((meth)acryloyloxy)butyl-2'-(trimethylammonio)ethylphosphate, 2-((meth)acryloyloxy)pentyl-2'-(trimethylammonio)-ethylphosphate, 2-((meth)acryloyloxy)hexyl-2'-(trimethylammonio)ethyl phosphate, 2-(vinylloxy)ethyl-2'-(trimethylammonio)ethylphosphate, 2-(allyloxy)ethyl-2'-(trimethylammonio)ethylphosphate, 2-(vinylloxycarbonyl)ethyl-2'-(trimethylammonio)ethyl phosphate, 2-(allyloxycarbonyl)ethyl-2'-(trimethylammonio)-ethylphosphate, 2-(vinylcarbonylamino)ethyl-2'-(trimethylammonio)ethylphosphate, 2-(allyloxycarbonyl-amino)ethyl-2'-(trimethylammonio)ethyl phosphate, 2-(butenoyloxy)ethyl-2'-(trimethylammonio)ethylphosphate, and combinations thereof;
- (10) allyl alcohol;
- (11) N-2-hydroxyethyl vinyl carbamate;
- (12) N-vinyloxycarbonyl- β -alanine (VINAL);
- (13) N-vinyloxycarbonyl- α -alanine; or
- (14) combinations thereof.
39. The polymerizable composition of embodiment 37 or 38, wherein the polymerizable composition further comprise at least one secondary siloxane-containing vinylic monomer which comprises at least one siloxane-containing vinylic monomer of formula

(M1) or (M2):



in which: $\text{a}_{\text{M}1}$ is zero or 1; $\text{R}_{\text{M}0}$ is H or methyl; $\text{X}_{\text{M}0}$ is O or $\text{NR}_{\text{M}1}$; $\text{L}_{\text{M}1}$ is a C_2 - C_8 alkylene divalent radical or a divalent radical of $-\text{L}_{\text{M}1}'-\text{X}_{\text{M}1}-\text{L}_{\text{M}1}''-$, $-(\text{C}_2\text{H}_4\text{O})_{\text{v}1}-\text{CONH}-\text{L}_{\text{M}1}''-$, $-(\text{C}_2\text{H}_4\text{O})_{\text{v}1}-\text{L}_{\text{M}1}''-$, $-\text{L}_{\text{M}1}'-\text{NHCOO}-(\text{C}_2\text{H}_4\text{O})_{\text{v}1}-\text{L}_{\text{M}1}''-$, $-\text{CH}_2-\text{CH}(\text{OH})-\text{CH}_2-\text{X}_{\text{M}1}'-(\text{C}_2\text{H}_4\text{O})_{\text{v}2}-\text{L}_{\text{M}1}''-$, $-\text{L}_{\text{M}1}'-\text{X}_{\text{M}1}'-\text{CH}_2-\text{CH}(\text{OH})-\text{CH}_2-\text{O}-\text{L}_{\text{M}1}''-$, or $-(\text{C}_2\text{H}_4\text{O})_{\text{v}1}-\text{CH}_2-\text{CH}(\text{OH})-\text{CH}_2-\text{O}-\text{L}_{\text{M}1}''-$; $\text{L}_{\text{M}1}'$ is a C_2 - C_8 alkylene divalent radical which has zero or one hydroxyl group; $\text{L}_{\text{M}1}''$ is C_3 - C_8 alkylene divalent radical which has zero or one hydroxyl group; $\text{X}_{\text{M}1}$ is O, $\text{NR}_{\text{M}1}$, NHCOO , OCONH , $\text{CONR}_{\text{M}1}$, or $\text{NR}_{\text{M}1}\text{CO}$; $\text{R}_{\text{M}1}$ is H or a C_1 - C_4 alkyl having 0 to 2 hydroxyl group; $\text{R}_{\text{T}1}$ and $\text{R}_{\text{T}2}$ independent of each other are a C_1 - C_6 alkyl; $\text{X}_{\text{M}1}'$ is O or NR_1 ; $\text{v}1$ is an integer of 1 to 30; $\text{v}2$ is an integer of 0 to 30; $\text{n}1$ is an integer of 3 to 40; and $\text{r}1$ is an integer of 2 or 3.

40. The polymerizable composition of any one of embodiments 37 to 39, wherein the polymerizable composition further comprise at least one secondary siloxane-containing vinylic monomer which comprises tris(trimethylsilyloxy)silylpropyl (meth)acrylate, [3-(meth)acryloxy-2-hydroxypropyloxy]propylbis(trimethylsiloxy)methylsilane, [3-(meth)acryloxy-2-hydroxypropyloxy]propylbis(trimethylsiloxy)butylsilane, 3-(meth)acryloxy-2-(2-hydroxyethoxy)-propyloxypropylbis(trimethylsiloxy)methylsilane, 3-(meth)acryloxy-2-hydroxypropyloxypropyltris(trimethylsiloxy)silane, N-2-(meth)acryloxyethyl-O-(methyl-bis-trimethylsiloxy-3-propyl)silyl carbamate, 3-(trimethylsilyl)propylvinyl carbonate, 3-(vinylloxycarbonylthio)-propyl-tris(trimethylsiloxy)silane, 3-[tris(trimethylsiloxy)silyl]propylvinyl carbamate, 3-[tris(trimethylsiloxy)silyl] propyl allyl carbamate, 3-[tris(trimethylsiloxy)silyl]propyl vinyl carbonate, or a combination thereof.
41. The polymerizable composition of any one of embodiments 37 to 40, wherein the polymerizable composition further comprise at least one non-silicone vinylic crosslinking agent selected from the group consisting of ethyleneglycol di-(meth)acrylate, diethyleneglycol di-(meth)acrylate, triethyleneglycol di-(meth)acrylate, tetraethyleneglycol di-(meth)acrylate, glycerol di-(meth)acrylate, 1,3-propanediol di-(meth)acrylate, 1,3-butanediol di-(meth)acrylate, 1,4-butanediol di-(meth)acrylate,

glycerol 1,3-diglycerolate di-(meth)acrylate, ethylenebis[oxy(2-hydroxypropane-1,3-diyl)] di-(meth)acrylate, bis[2-(meth)acryloxyethyl] phosphate, trimethylolpropane di-(meth)acrylate, 3,4-bis[(meth)acryloyl]tetrahydrofuran, diacrylamide, dimethacrylamide, N,N-di(meth)acryloyl-N-methylamine, N,N-di(meth)acryloyl-N-ethylamine, N,N'-methylene bis(meth)acrylamide, N,N'-ethylene bis(meth)acrylamide, N,N'-dihydroxyethylene bis(meth)acrylamide, N,N'-propylene bis(meth)acrylamide, N,N'-2-hydroxypropylene bis(meth)acrylamide, N,N'-2,3-dihydroxybutylene bis(meth)acrylamide, 1,3-bis(meth)acrylamidepropane-2-yl dihydrogen phosphate, piperazine diacrylamide, tetraethyleneglycol divinyl ether, triethyleneglycol divinyl ether, diethyleneglycol divinyl ether, ethyleneglycol divinyl ether, triallyl isocyanurate, triallyl cyanurate, trimethylolpropane trimethacrylate, pentaerythritol tetramethacrylate, bisphenol A dimethacrylate, allylmethacrylate, allylacrylate, N-allyl-(meth)acrylamide, and combinations thereof.

42. The polymerizable composition of any one of embodiments 1 to 41, wherein the polymerizable composition is free of organic solvent and comprises at least one blending vinylic monomer.
43. The polymerizable composition of embodiment 42, wherein said at least one blending vinylic monomer comprises C₁-C₁₀ alkyl (meth)acrylate, cyclopentylacrylate, cyclohexylmethacrylate, cyclohexylacrylate, isobornyl (meth)acrylate, styrene, 4,6-trimethylstyrene (TMS), t-butyl styrene (TBS), trifluoroethyl (meth)acrylate, hexafluoroisopropyl (meth)acrylate, hexafluorobutyl (meth)acrylate, or combinations thereof.
44. The polymerizable composition of any one of embodiments 1 to 43, wherein the polymerizable composition comprises:
 - (1) from about 5% to about 60% by weight of said at least one hydrophilic (meth)acrylamide-type monomer;
 - (2) from about 5% to about 50% by weight of said at least one siloxane-containing (meth)acrylamido monomer;
 - (3) from about 5% to about 50% by weight of said at least one polysiloxane vinylic crosslinker; and
 - (4) from about 0.1% to about 2.5% by weight of at least one UV-absorbing vinylic monomer,
 with the proviso that the listed components and any additional components add up to 100% by weight.
45. The polymerizable composition of embodiment 44, wherein the polymerizable composition comprises from about 10% to about 50% by weight of said at least one hydrophilic (meth)acrylamide-type monomer.
46. The polymerizable composition of embodiment 44, wherein the polymerizable

composition comprises from about 15% to about 45% by weight of said at least one hydrophilic (meth)acrylamide-type monomer.

47. The polymerizable composition of embodiment 44, wherein the polymerizable composition comprises from about 20% to about 40% by weight of said at least one hydrophilic (meth)acrylamide-type monomer.
48. The polymerizable composition of any one of embodiments 44 to 47, wherein the polymerizable composition comprises from about 10% to about 40% by weight of said at least one siloxane-containing (meth)acrylamido monomer.
49. The polymerizable composition of any one of embodiments 44 to 47, wherein the polymerizable composition comprises from about 15% to about 30% by weight of said at least one siloxane-containing (meth)acrylamido monomer.
50. The polymerizable composition of any one of embodiments 44 to 49, wherein the polymerizable composition comprises from about 10% to about 40% by weight of said at least one polysiloxane vinylic crosslinker.
51. The polymerizable composition of any one of embodiments 44 to 49, wherein the polymerizable composition comprises from about 15% to about 35% by weight of said at least one polysiloxane vinylic crosslinker.
52. The polymerizable composition of any one of embodiments 44 to 51, wherein the polymerizable composition comprises from about 0.2% to about 2.0% by weight of said at least one UV-absorbing vinylic monomer.
53. The polymerizable composition of any one of embodiments 44 to 51, wherein the polymerizable composition comprises from about 0.3% to about 1.5% by weight of said at least one UV-absorbing vinylic monomer.
54. The polymerizable composition of any one of embodiments 44 to 53, wherein the polymerizable composition comprises from about 0.1% to 1.3% by weight of said at least one acygermanium photoinitiator.
55. The polymerizable composition of any one of embodiments 44 to 53, wherein the polymerizable composition comprises from about 0.5% to about 1.1% by weight of said at least one acygermanium photoinitiator.
56. A method for producing photochromic silicone hydrogel contact lenses, the method comprising the steps of:
 - (1) obtaining a mold, wherein the mold has a first mold half with a first molding surface defining the anterior surface of a contact lens and a second mold half with a second molding surface defining the posterior surface of the contact lens, wherein said first and second mold halves are configured to receive each other such that a cavity is formed between said first and second molding surfaces;
 - (2) introducing a polymerizable composition of any one of embodiments 1 to 55 into the

cavity; and

(3) irradiating the polymerizable composition in the mold for about 80 seconds or less with a visible light in a region of from 420 to 500 nm, so as to form the photochromic silicone hydrogel contact lens,

wherein the formed photochromic silicone hydrogel contact lens comprises an anterior surface defined by the first molding surface and an opposite posterior surface defined by the second molding surface and is capable of undergoing reversible change from a colorless or light-colored state to a colored state upon exposure to UV or HEVL irradiation.

57. The method of embodiment 56, wherein the mold is a reusable mold, wherein the step of irradiating is performed under a spatial limitation of actinic radiation, wherein the formed UV-absorbing silicone hydrogel contact lens comprises a lens edge defined by the spatial limitation of actinic radiation.
58. The method of embodiment 56 or 57, wherein the polymerizable composition in the mold is irradiated for about 65 seconds or less.
59. The method of embodiment 56 or 57, wherein the polymerizable composition in the mold is irradiated for about 50 seconds or less.
60. The method of embodiment 56 or 57, wherein the polymerizable composition in the mold is irradiated for about 35 seconds or less.
61. A photochromic silicone hydrogel contact lens obtained according to a method of any one of embodiments 56 to 60.
62. The photochromic silicone hydrogel contact lens of embodiment 61, having: an oxygen permeability of at least about 40 barrers (preferably at least about 60 barrers, more preferably at least about 80 barrers) (at about 35°C); an elastic modulus of about 2.0 MPa or less (preferably about 1.5 MPa or less, more preferably about 1.2 or less, even more preferably from about 0.4 MPa to about 1.0 MPa) (at a temperature of from 22°C to 28°C); and/or a water content of from about 15% to about 70% (preferably from about 20% to about 50% by weight) (at a temperature of from 22°C to 28°C) when fully hydrated.

The previous disclosure will enable one having ordinary skill in the art to practice the invention. Various modifications, variations, and combinations can be made to the various embodiment described herein. In order to better enable the reader to understand specific embodiments and the advantages thereof, reference to the following examples is suggested. It is intended that the specification and examples be considered as exemplary.

Example 1

Oxygen Permeability Measurements

Unless specified, the oxygen transmissibility (Dk/t), the intrinsic (or edge-corrected) oxygen permeability (Dk_i or Dk_e) of a lens and a lens material are determined according to procedures described in ISO 18369-4.

Equilibrium Water Content

The equilibrium water content (EWC) of contact lenses are determined as follows.

Amount of water (expressed as percent by weight) present in a hydrated hydrogel contact lens, which is fully equilibrated in saline solution, is determined at room temperature. Quickly stack the lenses, and transfer the lens stack to the aluminum pan on the analytical balance after blotting lens in a cloth. The number of lenses for each sample pan is typically five (5). Record the pan plus hydrated weight of the lenses. Cover the pan with aluminum foil. Place pans in a laboratory oven at $100 \pm 2^\circ\text{C}$ to dry for 16-18 hours. Remove pan plus lenses from the oven and cool in a desiccator for at least 30 minutes. Remove a single pan from the desiccator, and discard the aluminum foil. Weigh the pan plus dried lens sample on an analytical balance. Repeat for all pans. The wet and dry weight of the lens samples can be calculated by subtracting the weight of the empty weigh pan.

Elastic Modulus

The elastic modulus of a contact lens is determined using a MTS insight instrument. The contact lens is first cut into a 3.12 mm wide strip using Precision Concept two stage cutter. Five thickness values are measured within 6.5mm gauge length. The strip is mounted on the instrument grips and submerged in PBS (phosphate buffered saline) with the temperature controlled at $21 \pm 2^\circ\text{C}$. Typically 5N Load cell is used for the test. Constant force and speed is applied to the sample until the sample breaks. Force and displacement data are collected by the TestWorks software. The elastic modulus value is calculated by the TestWorks software which is the slope or tangent of the stress vs. strain curve near zero elongation, in the elastic deformation region.

Transmittance

Contact lenses are manually placed into a specially fabricated sample holder or the like which can maintain the shape of the lens as it would be when placing onto eye. This holder is then submerged into a 1 cm path-length quartz cell containing phosphate buffered saline (PBS, pH $\sim 7.0 - 7.4$) as the reference. A UV/visible spectrophotometer, such as, Varian Cary 3E UV-Visible Spectrophotometer with a LabSphere DRA-CA-302 beam splitter or the like, can be used in this measurement. Percent transmission spectra are collected at a wavelength range of 250-800 nm with %T values collected at 0.5 nm intervals. This data is transposed onto an Excel spreadsheet and used to determine if the lenses conform to Class

1 UV absorbance. Transmittance is calculated using the following equations:

$$\text{UVA \%T} = \text{Average \% Transmission between 315 nm and 380 nm} \times 100$$

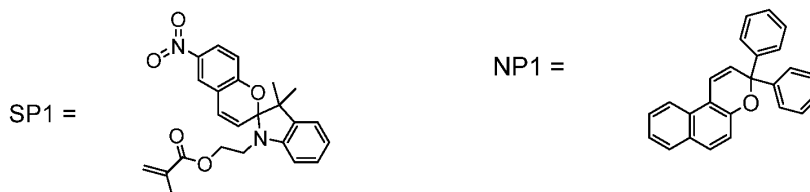
$$\text{UVB \%T} = \text{Average \% Transmission between 280 nm and 315 nm} \times 100$$

$$\text{Violet \%T} = \text{Average \% Transmission between 380 nm and 440 nm} \times 100.$$

Photo-rheology: The photo-rheology experiment measures the elastic (G') and viscous modulus (G'') as a function of time during curing. The experiment is conducted by using an appropriate light source, optionally cutoff filters to select wavelengths of interest, and a rheometer. The light source is a LED that delivers a visible light having a spectral centroid of about 452 nm and a total irradiance of about 54 mW/cm² (OPSYTEC DR. GROEBEL). The sample (i.e., a polymerizable composition) is placed between a quartz plate that allows the actinic radiation (i.e., a visible light with a spectral centroid of about 452nm and a total irradiance of about 54 mW/cm²) to pass through and the rheometer. The curing time of a polymerizable composition is determined when the elastic modulus (G') reaches a plateau.

Chemicals

DMAC represents N,N-dimethyl acetamide; PrOH represents 1-propanol; CE-PDMS represents a polysiloxane vinylic crosslinker which has three polydimethylsiloxane (PDMS) segments linked via diurethane linkages between two PDMS segments and two urethane linkages each located between one terminal methacrylate group and one PDMS segment and is prepared according to method similar to what described in Example 2 of U.S. Pat. No. 9315669; TRIS-Am represents N-[tris(trimethylsiloxy)-silylpropyl]acrylamide; DMA represent N,N-dimethylacrylamide; H-TEMPO represents 4-hydroxy-2,2,6,6-tetramethylpiperidin-1-oxyl; L-PEG 2000 represents N-(carbonyl-methoxypolyethylene glycol-2000)-1,2-disteaoyl-sn-glycero-3-phosphoethanolamin, sodium salt; DMPC represents 1,2-dimyristoyl-sn-glycero-3-phosphocholine; Ge PI represent Bis(4-methoxybenzoyl)diethylgermanium; SP1 represents 2-(3',3'-Dimethyl-6-nitrospiro[chromene-2,2'-indolin]-1'-yl)ethyl methacrylate (an acylgermanium photoinitiator from BLD pharma) of structure formula shown below; NP1 represents 3,3-Diphenyl-3H-naphtho[2,1-b]pyran (from TCI America) of the structure formula shown below; NPD-blue represent a polymerizable naphthopyran photochromic dye having a blue color in activated state (from Vivimed).



Example 2

The solution of SP1 is prepared by dissolving SP1 in PrOH in a desired amount to

make a 20 ppm solution. Its UV-Vis spectrum is acquired on a Lambda 850 spectrometer. A black light lamp is used to activate the SP1 solution ($5\text{mW}/\text{cm}^2$ for 2 minutes), and the UV-Vis spectrum is collected immediately after UV activation. As shown in Figure 1 on the spectral comparison, the SP1 (at “off” state, without UV activation) has no significant absorption around 450nm, and therefore does not interfere with Ge-photoinitiator’s curing condition. On the other hand, the dye has significant absorption in the range of 300-400nm, and may pose a challenge to UV-cure lens formulations that contain UV-photoinitiator, e.g., Darocur 1173.

Example 3

The solution of NP1 is prepared by dissolving NP1 in PrOH in a desired amount to make a 20 ppm solution. Its UV-Vis spectrum is acquired on a Lambda 850 spectrometer. A black light lamp is used to activate the dye solution ($5\text{mW}/\text{cm}^2$ for 2 minutes), and the UV-Vis spectrum is collected immediately after UV activation. As shown in Figure 2 on the spectral comparison, the NP1 (at “off” state, without UV activation) has no significant absorption around 450nm, and therefore does not interfere with Ge-photoinitiator’s curing condition. On the other hand, the dye has significant absorption in the range of 300-400nm, and may pose a challenge to UV-cure lens formulations that contain a UV-photoinitiator, e.g., Darocur 1173.

Example 4

Preparation of SiHy Lens Formulations

Control lens formulation and photochromic dye containing lens formulations are prepared to have the composition shown in Table 1. In order to better dissolve the photochromic dye, about 4.5 parts of DMAC is used to replace 4.5 parts of PrOH in the photochromic dye containing lens formulations.

Table 1

Components	Lens Formulation (weight part units)		
	control	SP1 _{0.5%}	NP1 _{0.5%}
CE-PDMS	31.63	31.60	31.67
TRIS-Am	19.77	19.76	19.80
DMA	23.27	23.20	23.15
PrOH	23.33	18.35	18.39
DMAC	0	4.58	4.47
H-TEMPO	0.04	0.04	0.04
L-PEG 2000	0.61	0.61	0.61
DMPC	0.76	0.76	0.76
Ge-PI	0.60	0.60	0.60
SP1	0	0.51	0
NP1	0	0	0.50

Example 5

Photo-rheology data are acquired by curing a lens formulation (Control, SP1_{0.5%}, or NP1_{0.5%}) prepared in Example 4 by using 452nm LED light with a total irradiance of 54 mW/cm². The results are shown in Figure 3 and Table 2. With the addition of photochromic dyes in the lens formulation, the curing time is slightly slower and the G' is almost the same as the control lens formulation. Therefore, a comparable lens property is expected for photochromic dye containing lens formulation under the same curing profile.

Table 2

Lens Formulation	Curing time (s)	G' (kPa)
control	12	131
SP1 _{0.5%}	16	124
NP1 _{0.5%}	16	128

Example 6**Photochromic SiHy Lens fabrication**

Photochromic SiHy contact lenses are prepared by cast-molding from a lens formulation prepared in Example 4 in a reusable mold (quartz female mold half and glass male mold half, optical power -3.00), similar to the mold shown in Figs. 1-6 in U.S. patent Nos. 7,384,590 and 7,387,759 (Figs. 1-6). A lens formulation prepared in Example 4 in the molds is cured under a controlled LED light source (curing time 30 seconds, irradiation peak at 452 nm, total irradiation 54 mW/cm²). After demolding and delensing, the lenses are hydrated in deionized water.

Example 7

Photochromic SiHy contact lenses (SP1_{0.5%}) prepared in Example 6 are activated by using a black light lamp for 2 minutes (peaks at 365nm, total irradiance of 5mW/cm²). Figure 4 shows the UV spectra of the photochromic SiHy contact lens before and after the activation. Upon removing the UV light, the photochromic SiHy contact lenses gradually faded back to their "off" state. Figure 5 shows a detailed fading kinetics at room temperature that is tracked by the UV-Vis spectrometer set at 550nm.

Example 8

Poly(acrylamide-co-acrylic acid) partial sodium salt ("poly(AAm-co-AA) 90/10", Mw 200,000) is purchased from Polysciences, Inc. and used as received; polyamidoamine-epichlorohydrin ("PAE") (Kymene) is obtained from Ashland as an aqueous solution and used as received; polyacrylic acid ("PAA", Mn: ~450 kDa) is obtained from Polysciences, Inc. and used as received.

PAA Solution

This solution is prepared as follows. An amount of PAA powder is dissolved in a

desired volume of PrOH to prepare a PAA solution. After PAA is fully dissolved, the pH is adjusted by adding ~3.8 wt.% formic acid to the PAA PrOH solution to about 2. The target concentration of PAA is about 0.44 wt.% by weight. The prepared PAA solution is filtered to remove any particulate or foreign matter.

Phosphate Buffered Saline (PBS)

A phosphate buffered saline is prepared by dissolving $\text{NaH}_2\text{PO}_4 \cdot \text{H}_2\text{O}$, $\text{Na}_2\text{HPO}_4 \cdot 2\text{H}_2\text{O}$, and in a given volume of purified water (distilled or deionized) to have the following composition: *ca.* 0.044 wt.% $\text{NaH}_2\text{PO}_4 \cdot \text{H}_2\text{O}$, *ca.* 0.388 wt.% $\text{Na}_2\text{HPO}_4 \cdot 2\text{H}_2\text{O}$, and *ca.* 0.79 wt.% NaCl.

IPC saline

The IPC saline is prepared by dissolving/mixing appropriate amounts of Poly(AAm-co-AA) (90/10), PAE, $\text{NaH}_2\text{PO}_4 \cdot \text{H}_2\text{O}$ and $\text{Na}_2\text{HPO}_4 \cdot 2\text{H}_2\text{O}$ in DI (de-ionized) water to have the following concentrations: about 0.28 wt.% of poly(AAm-co-AA); about 0.025 wt.% PAE; about 0.028 wt.% $\text{NaH}_2\text{PO}_4 \cdot \text{H}_2\text{O}$, about 0.231 wt.% $\text{Na}_2\text{HPO}_4 \cdot 2\text{H}_2\text{O}$ and then by adjusting pH to about 7.3. The prepared solution is pre-treated at 60°C for about 6 hours. After the heat pre-treatment, the IPC saline is cooled down back to room temperature. The IPC saline is filtered using a 0.22 micron membrane filter.

Preparation of SiHy Lens Formulations

A solution of NPD-blue (2% by weight) is prepared by dissolving NPD-blue in DMAc.

Two lens formulations including a polymerizable naphthopyran photochromic dye (NPD-blue) are prepared by mixing the control lens formulation prepared in Example 4 with the NPD-blue solution to have the composition shown in Table 3.

Table 3

Components	Lens Formulation (weight part units)	
	Ge-PC _{0.1%}	Ge-PC _{0.2%}
Control lens formulation	90	90
NPD-blue Solution (2wt%)	5	10
DMAc	5	0

Photorheology Study

Photo-rheology data are acquired by curing control formulation or formulation Ge-PC_{0.2%} prepared above by using 452nm LED light with a total irradiance of 54 mW/cm². The results are shown in Figure 6 (1 – Control Formulation; 2 – Formulation Ge-PC_{0.2%} (1st run); 3 – Formulation Ge-PC_{0.2%} (2nd run)) and Table 4. With the addition of photochromic dyes in the lens formulation, the curing time is slightly slower and the G' is almost the same as the control lens formulation. Therefore, a comparable lens property is expected for photochromic dye containing lens formulation under the same curing profile.

Table 4

Formulation	Curing time (s)	G' (kPa)
control	15	110
Ge-PC _{0.2%} (1 st run)	27	75
Ge-PC _{0.2%} (2 nd run)	28	78

Fabrication of Photochromic SiHy Contact Lenses

Two different types of photochromic SiHy contact lenses are cast-molded as described below.

The first type of photochromic SiHy contact lenses is the uniform distribution of photochromic dye in the lens body from edge to edge. This type of photochromic SiHy contact lenses are prepared by cast-molding from a lens formulation (control, Ge-PC_{0.1%}, or Ge-PC_{0.2%}) prepared above in a reusable mold (quartz female mold half and glass male mold half, optical power -3.00), similar to the mold shown in Figs. 1-6 in U.S. patent Nos. 7,384,590 and 7,387,759 (Figs. 1-6). ~50mg of a formulation (control, Ge-PC_{0.1%}, or Ge-PC_{0.2%}) is dosed into a reusable mold and cured under a controlled LED light source (curing time 30 seconds, irradiation peak at 452 nm, total irradiation 54 mW/cm²).

The second type of photochromic SiHy contact lenses is the lens central portion comprising a photochromic dye distributed therein. This type of photochromic SiHy contact lenses are prepared according to 2-steps of cast-molding from two lens formulations (control + Ge-PC_{0.1%}; or Control + Ge-PC_{0.2%}) prepared above in a reusable mold (quartz female mold half and glass male mold half, optical power -3.00), similar to the mold shown in Figs. 1-6 in U.S. patent Nos. 7,384,590 and 7,387,759 (Figs. 1-6). At the first step, ~3mg of a formulation (Ge-PC_{0.1%} or Ge-PC_{0.2%}) is dosed into the central portion of the quartz female mold half, closed with the glass male mold half and cured under a controlled LED light source (curing time 30 seconds, irradiation peak at 452 nm, total irradiation 54 mW/cm²). At the second step, the mold is opened, then ~50mg of the control formulation is dosed into the female mold half with a cured disk located in the central portion of the molding surface of the female mold half to immerse the cured disk, and then closed with the male mold half. The control formulation in the closed mold is cured under a controlled LED light source (curing time 30 seconds, irradiation peak at 452 nm, total irradiation 54 mW/cm²).

After demolding and delensing, the resultant photochromic SiHy contact lenses are placed in lens trays. Then the lens trays with lenses are immersed in water for about 1 minutes, followed by in MEK for about 5 minutes, then in water for about 52 seconds, then in 0.44 wt.% PAA Solution #1 (in PrOH) for about 44 seconds, followed by about 52 seconds in PrOH/H₂O (50/50 wt./wt.) mixture, then in DI water for about 3.5 minutes, then in phosphate buffer for about 52 seconds.

Photochromic SiHy contact lenses with a PAA base coating thereon prepared above

are placed in polypropylene lens packaging shells (one lens per shell) containing about 0.65 mL of the IPC saline prepared above. The blisters (packaging shells) are then sealed with foil and autoclaved for about 45 minutes at about 121°C, forming photochromic SiHy contact lenses with a cross-linked hydrophilic coating (i.e., a hydrogel coating) thereon.

Characterization of Photochromic SiHy Contact lenses

The photochromic SiHy contact lenses are activated by using a black light lamp (peaks at 365nm, total irradiance of 5mW/cm², 2 minutes), and upon removing the UV light, the lenses gradually fades back to their off state.

The fading kinetics at room temperature is quantified by using UV-Vis data. Figure 7 shows the temporally evolving spectra of a photochromic contact lens made from formulation Ge-PC_{0.2%} during the fading at room temperature. Figure 8 shows a detailed fading kinetics of the same photochromic contact lens as measured by the UV-Vis spectrometer set at 570nm.

Photochromic fatigue test of a photochromic contact lens made from formulation Ge-PC_{0.2%} is also carried out. In each cycle the lens is activated by a black light (5 mW/cm², 2 minutes), followed by a 5-minute fading. As shown in Figure 9, the fatigue test of the photochromic SiHy contact lens over 10 cycles shows no significant change in photochromic properties.

Example 9

Preparation of SiHy Lens Formulations

Lens formulations are prepared to have the compositions shown in Table 5.

Table 5

			(weight part units)		
	Control	Ge-PC _{1%}	Ge-PC _{0.75%}	Ge-PC _{0.5%}	Ge-PC _{0.35%}
H-TEMPO	0.04	0.04	0.04	0.04	0.04
L-PEG 2000	0.61	0.61	0.61	0.61	0.61
DMPC	0.76	0.76	0.76	0.76	0.76
CE-PDMS	31.67	31.67	31.67	31.67	31.67
TRIS	19.81	19.81	19.81	19.81	19.81
PrOH	22.11	18.05	18.05	18.05	18.05
DMA	23.12	23.12	23.12	23.12	23.12
Ge-PI	0.60	0.60	0.60	0.60	0.60
NPD-blue	-	1.00	0.75	0.50	0.25
DMAc	-	4.44	4.69	4.94	5.19

Photorheology Studies of Lens Formulations

The date of the photo-rheology studies of the lens formulations prepared above are shown in Table 6. The results show that the photocuring profiles of lens formulations are not changed significantly when the concentration of the NPD-blue is changed from zero to 1wt%.

Table 6

formulation ID	Run	cure time (s)	avg cure time (s)	G' at cure time (Pa)	G' avg. (Pa)
Control	1	19.5	21	106300	108050
	2	21.6		109800	
Ge-PC _{1%}	1	22.3	23	103900	108750
	2	24.4		113600	
Ge-PC _{0.75%}	1	20	20	111700	112950
	2	20.2		114200	
Ge-PC _{0.5%}	1	19.4	18	114000	108250
	2	15.9		102500	
Ge-PC _{0.25%}	1	18.7	18	109700	111600
	2	18		113500	

Lens fabrication

Two types of photochromic SiHy contact lenses are prepared from the lens formulations prepared above according to the procedures described in Example 8.

Characterization of Photochromic SiHy Contact lenses

The photochromic SiHy contact lenses are activated by using a black light lamp (peaks at 365nm, total irradiance of 5mW/cm², 2 minutes), and upon removing the UV light, the lenses gradually fades back to their off state over about 5 ~ 6 minutes.

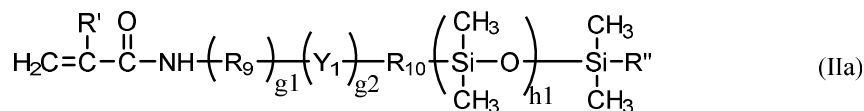
All the publications and patents which have been cited herein above are hereby incorporated by reference in their entireties.

Claims:

1. A method for producing photochromic silicone hydrogel contact lenses, the method comprising the steps of:
 - (1) obtaining a mold, wherein the mold has a first mold half with a first molding surface defining the anterior surface of a contact lens and a second mold half with a second molding surface defining the posterior surface of the contact lens, wherein said first and second mold halves are configured to receive each other such that a cavity is formed between said first and second molding surfaces;
 - (2) introducing a polymerizable composition into the cavity, wherein the polymerizable composition comprises
 - (a) from 20% to 40% by weight of at least one hydrophilic (meth)acrylamido monomer
 - (b) from 15% to 30% by weight of at least one siloxane-containing (meth)acrylamido monomer,
 - (c) from 15% to 35% by weight of at least one polysiloxane vinylic crosslinker,
 - (d) from 0.005% to 1.0% by weight of at least one photochromic compound,
 - (e) from 0.3% to 1.5% by weight of at least one UV-absorbing vinylic monomer, and
 - (f) from 0.05% to 1.5% by weight of at least one an acyl germanium photoinitiator,relative to the total weight of the polymerizable composition, wherein said at least one acyl germanium photoinitiator is capable of initiating a free-radical polymerization under irradiation with a light source including a light in the region of 420 to 500 nm, wherein the sum of the amounts of components (a) to (e) is at least 96% by weight relative to total amount of all polymerizable components in the polymerizable composition, wherein the polymerizable composition has a curing time of 50 seconds or less as determined in photo-rheology study using a LED light source which provide a visible light with a spectral centroid of 452 nm and a total irradiance of 54 mW/cm², wherein the listed components and any additional components add up to 100% by weight; and
 - (3) irradiating the polymerizable composition in the mold for 50 seconds or less with a visible light in a region of from 420 to 500 nm, so as to form the photochromic silicone hydrogel contact lens,wherein the formed photochromic silicone hydrogel contact lens comprises an anterior surface defined by the first molding surface and an opposite posterior surface defined by the second molding surface and is capable of undergoing reversible change from a colorless or light-colored state to a colored state upon exposure to UV or HEVL

- irradiation.
2. The method of claim 1, wherein the mold is a reusable mold, wherein the step of irradiating is performed under a spatial limitation of actinic radiation, wherein the formed UV-absorbing silicone hydrogel contact lens comprises a lens edge defined by the spatial limitation of actinic radiation.
 3. The method of claim 1 or 2, wherein the polymerizable composition has a curing time of 35 second or less as determined in photo-rheology study using a LED light source which provide a visible light with a spectral centroid of 452nm and a total irradiance of 54 mW/cm², wherein the polymerizable composition in the mold is irradiated for 35 seconds or less.
 4. The method of any one of claims 1 to 3, wherein said at least one hydrophilic (meth)acrylamido monomer comprises (meth)acrylamide, N,N-dimethyl (meth)acrylamide, N-ethyl (meth)acrylamide, N,N-diethyl (meth)acrylamide, N-propyl (meth)acrylamide, N-isopropyl (meth)acrylamide, N-3-methoxypropyl (meth)acrylamide, N-2-hydroxyethyl (meth)acrylamide, N,N-bis(hydroxyethyl) (meth)acrylamide, N-3-hydroxypropyl (meth)acrylamide, N-2-hydroxypropyl (meth)acrylamide, N-2,3-dihydroxypropyl (meth)acrylamide, N-4-hydroxybutyl (meth)acrylamide, N,N-bis(2-hydroxyethyl) (meth)acrylamide, N-tris(hydroxymethyl) methyl (meth)acrylamide, 2-(meth)acrylamidoglycolic acid, 3-(meth)acrylamido-propionic acid, 4-(meth)acrylamido-butanoic acid, 3-(meth)acrylamido-2-methylbutanoic acid, 3-(meth)acrylamido-3-methylbutanoic acid, 2-(meth)acrylamido-2-methyl-3,3-dimethyl butanoic acid, 5-(meth)acrylamidopentanoic acid, 3-(meth)acrylamidohexanoic acid, 4-(meth)acrylamido-3,3-dimethylhexanoic acid, (3-(meth)acrylamidophenyl)boronic acid, 3-((3-methacrylamidopropyl)dimethylammonio)propane-1-sulfonate; 3-((3-acrylamidopropyl)dimethylammonio)propane-1-sulfonate, N-2-aminoethyl (meth)acrylamide, N-2-methylaminoethyl (meth)acrylamide, N-2-ethylaminoethyl (meth)acrylamide, N-2-dimethylaminoethyl (meth)acrylamide, N-3-aminopropyl (meth)acrylamide, N-3-methylaminopropyl (meth)acrylamide, N-3-dimethylaminopropyl (meth)acrylamide, poly(ethylene glycol)ethyl (meth)acrylamide having a number average molecular weight of up to 700, methoxy-poly(ethylene glycol)ethyl (meth)acrylamide having a number average molecular weight of up to 700, or combination thereof.
 5. The method of any one of claims 1 to 4, wherein said at least one siloxane-containing (meth)acrylamido monomer comprises a (meth)acrylamido monomer containing a tris(trialkylsiloxy)silyl group or a bis(trialkylsilyloxy)alkylsilyl group and/or a mono-

(meth)acrylamido-terminated oligo- or polysiloxane of formula (IIa)



in which R' is hydrogen or methyl, R'' independent of each other is C₁-C₆ alkyl, g1 and g2 independent of each other are integer of 0 or 1, h1 is an integer of 2 to 25, R₉ and R₁₀ independent of each other are a substituted or unsubstituted C₁-C₁₀ alkylene

divalent radical, Y₁ is a linkage of $-\text{NH}-\overset{\text{O}}{\underset{||}{\text{C}}}-$, $-\overset{\text{O}}{\underset{||}{\text{C}}}-\text{NH}-$, or $-\overset{\text{R}_{12}'}{\underset{|}{\text{C}}}-\overset{\text{O}}{\underset{||}{\text{C}}}-\text{O}-$ in which R₁₂ and R₁₂' independent of each other are C₁-C₆ alkyl.

6. The method of any one of claims 1 to 5, wherein said at least one polysiloxane vinylic crosslinker comprises: (1) a vinylic crosslinker which comprises one sole polydiorganosiloxane segment and two terminal ethylenically-unsaturated groups selected from the group consisting of (meth)acryloyloxy groups, (meth)acryloylamino groups, vinyl carbonate groups, vinylcarbamate groups; and/or (2) a chain-extended polysiloxane vinylic crosslinker which comprises at least two polydiorganosiloxane segment and a covalent linker between each pair of polydiorganosiloxane segments and two two terminal ethylenically-unsaturated groups selected from the group consisting of (meth)acryloyloxy groups, (meth)acryloylamino groups, vinyl carbonate groups, vinylcarbamate groups.
7. The method of any one of claims 1 to 6, wherein said at least one photochromic compound comprises a naphthopyran, an indeno-fused naphthopyran (i.e., indeno-naphthopyran), a heterocyclic ring-fused naphthopyran, a benzopyran, a phenanthropyran, a quinopyran, a quinolinopyran, a fluoroanthropyran, an anthracene-fused pyran, a tetracene-fused pyran, a spiro(benzindoline) naphthopyran, a spiro(indoline)naphthopyran, a spiro(indoline)benzopyran, a spiro(indoline)quinopyran, a spiro(indoline)pyran, a naphthoxazine, a spirobenzopyran, a spirobenzothiopyran, a naphthacenedione, a benzoxazine, a spirooxazine, a naphthoxazine, a spiro(benzindoline)naphthoxazine, a spiro(indoline)naphthoxazine, a spiro(indoline)pyrido-benzoxazine, a spiro(indoline)benzoxazine, a spiro(benzindoline)benzoxazine, a spiro(benzindoline)pyridobenzoxazine, a spiro(indoline)fluoranthoxazine, a spiro(indoline)-quinoxazine, a spiropiperidine-naphthopyran, a piro(indoline)pyronobenzoxazinone, a benzospiropyran, a naphthospiropyran, a spirobenzoxazine-pyrrolopyridine, a spironaphthoxazine-pyrrolopyridine, a spiro-oxazepin-benzoxazine, a spiro-oxazepin-naphthoxazine, a spiro(indoline)

benzothiazoloxazine, a spiro(indoline)benzopyrroloxazine, a spiro(indoline)quinazolino-oxazine, a spiro(indoline)-anthracenobenzoxazine, a benzofurobenzopyran, a benzothienobenzopyran, a naphthofurobenzopyran, a benzopyrano-fused naphthopyran, a spiro(isoindoline)-naphthoxazine, a spiro(isoindoline)benzoxazine, or a mixture thereof.

8. The method of claim 7, wherein said at least one photochromic compound is polymerizable and comprises an ethylenically-unsaturated group.
9. The method of claim 8, wherein said at least one photochromic compound is a coupling reaction product of a reactive (meth)acrylamido or (meth)acryloxy monomer having a first reactive functional group and a reactive photochromic compound have a second reactive functional group in the absence or presence of a coupling agent under coupling reaction, wherein the first reactive functional group is selected from the group consisting of $-\text{COCl}$, $-\text{COBr}$, $-\text{COOH}$, $-\text{NHR}_{\text{N}_2}$, $-\text{NCO}$, $-\text{OH}$, $-\text{CHO}$, , , or $-\text{N} \diagup \text{R}_0$, wherein the second reactive functional group is selected from the group consisting of $-\text{COOH}$, $-\text{NHR}_{\text{N}_2}$, , , , $-\text{NCO}$, $-\text{OH}$, $-\text{SH}$, $-\text{CHO}$, , , or $-\text{N} \diagup \text{R}_0$, wherein R_0 is hydrogen or methyl and R_{N_2} is hydrogen, a linear or branched $\text{C}_1\text{-C}_{15}$ alkyl, cyclohexyl, cyclopentyl, a substituted or unsubstituted phenyl, or a substituted- or unsubstituted-phenyl- $\text{C}_1\text{-C}_6$ alkyl.
10. The method of claim 8, wherein reactive (meth)acrylamido or (meth)acryloxy monomer is selected from the group consisting of (meth)acryloyl halides of $\text{CH}_2=\text{CH}-\text{COX}$ or $\text{CH}_2=\text{CCH}_3-\text{COX}$ in which $\text{X} = \text{Cl}$ or Br , N-hydroxysuccinimide ester of (meth)acrylic acid, glycidyl (meth)acrylate, isocyanato- $\text{C}_2\text{-C}_6$ alkyl (meth)acrylate, amino- $\text{C}_2\text{-C}_6$ alkyl (meth)acrylamide, $\text{C}_1\text{-C}_6$ alkylamino- $\text{C}_2\text{-C}_6$ alkyl (meth)acrylamide, (meth)acrylic acid, a $\text{C}_2\text{-C}_4$ alkylacrylic acid, N-2-acrylamidoglycolic acid, 3-(acryloylxy)propanoic acid, $\text{C}_2\text{-C}_6$ hydroxylalkyl (meth)acrylate, a $\text{C}_2\text{-C}_6$ hydroxyalkyl (meth)acrylamide, an azlactone-containing vinylic monomer, an aziridinyl $\text{C}_1\text{-C}_{12}$ alkyl (meth)acrylate, acrolein, methacrolein, crotonaldehyde, and combinations thereof.
11. The method of claim 9 or 10, wherein the reactive photochromic compound comprises 3,3-bis(4-methoxyphenyl)-6,11-dimethyl-13-hydroxyethoxy-13-phenyl-3H,13H-indeno [2',3':3,4]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-6,11-dimethyl-13-(2-hydroxycarbonyl)ethyl)carboxyethoxy-13-phenyl-3H,13H-indeno [2',3':3,4]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-6,11-dimethyl-13-hydroxy-13-phenyl-3H,13H-indeno [2',3':3,4]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-6,11-dimethyl-13-(2-hydroxycarbonyl)ethyl)carboxy-13-phenyl-3H,13H-indeno [2',3':3,4] naphtho[1,2-

b]pyran; 3,3-bis(4-methoxyphenyl)-6,11-dimethyl-13-hydroxy-13-propyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-6,11-dimethyl-13-(2-hydroxycarbonylethyl)carboxy-13-propyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-10,11-dimethoxy-13-hydroxy-13-ethyl-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-10,11-dimethoxy-13-(2-hydroxycarbonylethyl)carboxy-13-ethyl-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-7-methoxy-11-phenyl-13-hydroxymethyl-13-methyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-7-methoxy-11-phenyl-13-(2-hydroxycarbonylethyl)carboxymethyl-13-methyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-7-methoxy-11-phenyl-13-hydroxy-13-methyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-7-methoxy-11-phenyl-13-(2-hydroxycarbonylethyl)carboxy-13-methyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-11-phenyl-13-hydroxy-13-methyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-11-phenyl-13-(2-hydroxycarbonylethyl)carboxy-13-methyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-6,7,10,11-tetramethoxy-13-hydroxy-13-ethyl-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-6,7,10,11-tetramethyl-13-(2-hydroxycarbonylethyl)carboxy-13-methyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-6,7-dimethoxy-13-hydroxy-13-ethyl-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3,3-bis(4-methoxyphenyl)-11-(4-(4,5-diphenyl-1H-imidazol-2-yl)phenyl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-6-methoxy-7-(3-hydroxymethylenepiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-6-methoxy-7-(3-(2-hydroxycarbonylethyl)-carboxymethylenepiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-6,7-dimethoxy-13-hydroxy-13-butyl-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-6,11,13-trimethyl-13-(2-(2-hydroxyethoxy)-ethoxy)-indeno[2,1-f]naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-6,11,13-trimethyl-13-(2-(2-(2-(2-hydroxyethoxy)ethoxy)ethoxy)ethoxy)-indeno[2,1-f]naphtho[1,2-b]pyran; 3,3-diphenyl-13-hydroxy-13-(2-oxo-2-ethoxyethyl)-1H-indeno[2,1-f]naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-6,11-dimethyl-13-hydroxy-13-(2-oxo-2-ethoxyethyl)-1H-indeno[2,1-f]naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-6,11-dimethyl-13-butyl-13-hydroxy-indeno[2,1-f]naphtho[1,2-b]pyran; 3,3-bis(4-hydroxyphenyl)-7-methoxy-11-phenyl-13,13-diethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-bis(4-hydroxyphenyl)-7-methoxy-11-phenyl-13,13-dipropyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-

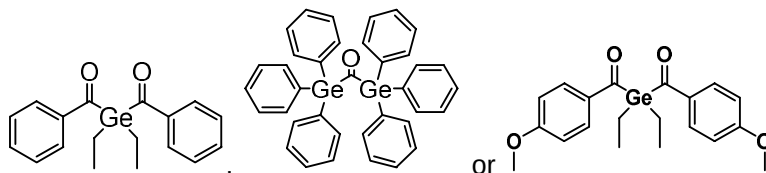
bis(4-hydroxyphenyl)-7-methoxy-11-phenyl-13-carbomethoxy-13-methyl-3H,13H-indeno[2',3':3,4] naphtho[1,2-b]pyran; 3,3-diphenyl-6,7-dimethoxy-13-methyl-13-methyl-13-hydroxyethoxy-ethoxyethoxy-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3,3-diphenyl-6,7,10,11-tetramethoxy-13-ethyl-13-methyl-13-hydroxyethoxyethoxy-3H,13H-indeno[2',3':3,4]naphtho [1,2-b]pyran; 6,7-dimthoxy-2,2-diphenyl-13-hydroxy-13-butyl-2H,13H-indeno[1',2':4,3] naphtho[1,2-b]pyran; 6,7-dimthoxy-2,2-diphenyl-13-hydroxy-13-methyl-2H,13H-indeno[1',2':4,3]naphtho[1,2-b]pyran; 6,7-dimthoxy-2,2-diphenyl-13-hydroxy-2H,13H-indeno[1',2':4,3]naphtho[1,2-b]pyran; 3-(4-allyloxyphenyl)-3-(4-morpholinophenyl)-7-methoxy-11-phenyl-13,13-dimethy-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-fluorophenyl)-3-(4-butoxyphenyl)-10-(piperazin-1-yl)-6-trifluoromethyl-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-fluorophenyl)-3-(4-butoxyphenyl)-10-(4-hydroxybenzamido)-6-trifluoromethyl-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-fluorophenyl)-3-(4-butoxyphenyl)-10-vinyl-6-trifluoromethyl-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-fluorophenyl)-3-(4-butoxyphenyl)-10-(carboxylic acid)-6-trifluoromethyl-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-fluorophenyl)-3-(4-methoxyphenyl)-6-methoxy-7-(4-hydroxypiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-fluorophenyl)-3-(4-methoxyphenyl)-6-methoxy-7-(4-(2-hydroxycarbonyl)ethyl) carboxypiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-fluorophenyl)-3-(4-morpholinophenyl)-6-methoxy-7-(4-hydroxypiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-fluorophenyl)-3-(4-morpholinophenyl)-6-methoxy-7-(4-(2-hydroxycarbonyl)ethyl) carboxypiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4] naphtho[1,2-b]pyran; 3-(4-hydroxyethoxyphenyl)-3-(4-methoxyphenyl)-11-(4-(N,N-dimethylamino)phenyl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-methoxyphenyl)-3-(2,4-dimethoxyphenyl)-phenyl-6,11-dimethoxy-13-methyl-13-methyl-13-hydroxyethoxyethoxyethoxy-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-methoxyphenyl)-3-(3-methyl-4-methoxyphenyl)-13-hydroxy-indeno[2,1-f]naphtho[1,2-b]pyran; 3-(4-methoxyphenyl)-3-(4-morpholinophenyl)-6,7,10,11-tetramethoxy-13-hydroxy-13-ethyl-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3-(4-methoxyphenyl)-3-(4-morpholinophenyl)-6,7,10,11-tetramethoxy-13-hydroxy-13-butyl-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3-(4-methoxyphenyl)-3-(4-morpholinophenyl)-6,7-dimethoxy-13-hydroxy-13-ethyl-3H,13H-indeno [2,1-f]naphtho[1,2-b]pyran; 3-(4-methoxyphenyl)-3-(4-morpholinophenyl)-6,7-dimethoxy-13-hydroxy-13-methyl-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3-(4-

methoxyphenyl)-3-(4-morpholinophenyl)-13-oxo-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3-(4-methoxyphenyl)-3-phenyl-6-methoxy-7-morpholino-13-methyl-13-methyl-13-hydroxyethoxyethoxyethoxy-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-methoxyphenyl)-3-phenyl-6,11-dimethoxy-13-methyl-13-methyl-13-hydroxyethoxyethoxy-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 2-(4-methoxyphenyl)-2-(4-morpholinophenyl)-5-hydroxy-6-carboethoxy-2H-naphtho[1,2-b] pyran; 3-(4-morpholinophenyl)-3-phenyl-6,7-dimethoxy-13-hydroxy-13-ethyl-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3-(4-morpholinophenyl)-3-phenyl-6,7-dimethoxy-13-hydroxy-13-butyl-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3-(4-morpholinophenyl)-3-phenyl-6,7-dimethoxy-13-oxo-3H,13H-indeno[2,1-f]naphtho[1,2-b]pyran; 3-phenyl-3-(4-hydroxyethoxyphenyl)-6-methoxy-7-morpholino-13,13-dimethyl-3H,13H-indeno[2',3':3,4] naphtho[1,2-b]pyran; 3-phenyl-3-(4-(2-hydroxycarbonylethyl)carboxyethoxyphenyl)-6-methoxy-7-morpholino-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-(2-hydroxyethoxy)phenyl)-6-methoxy-7-piperidino-13,13-dimethyl-3H,13H-indene[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-(2-(2-hydroxycarbonylethyl)-carboxyethoxy)phenyl)-6-methoxy-7-piperidino-13,13-dimethyl-3H,13H-indene[2',3':3,4] naphtho[1,2-b]pyran; 3-phenyl-3-(4-hydroxyphenyl)-6,7-dimethoxy-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-hydroxyethoxy)phenyl)-6,7-dimethoxy-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-(2-hydroxycarbonylethyl)carboxyethoxy)phenyl)-6,7-dimethoxy-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-(2-hydroxycarbonylethyl)carboxy-phenyl)-6,7-dimethoxy-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-(4-(2-hydroxyethyl)piperazin-1-yl)phenyl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-(4-(2-(2-hydroxycarbonylethyl)-carboxyethyl)piperazin-1-yl)phenyl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho-[1,2-b]pyran; 3-phenyl-3-(4-methoxyphenyl)-6-methoxy-7-piperazinyl-13,13-dimethyl-3H,13H-indene[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-methoxyphenyl)-6-methoxy-7-(3-hydroxymethylenepiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno [2',3":3,4]naphtho[1.2-b]pyran; 3-phenyl-3-(4-methoxyphenyl)-6-methoxy-7-(3-(2-hydroxycarbonylethyl) carboxymethylenepiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3":3,4]naphtho[1.2-b]pyran; 3-phenyl-3-(4-methoxyphenyl)-6-methoxy-7-(4-hydroxypipendin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-methoxyphenyl)-6-methoxy-7-(4-(2-hydroxycarbonylethyl)-carboxypipendin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-(4-methoxyphenyl-piperazin-1-yl)phenyl)-13,13-dimethyl-6-methoxy-7-

hydroxy-indeno [2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-(4-methoxyphenyl)-piperazin-1-yl)phenyl)-13,13-dimethyl-6-methoxy-7-(2-hydroxy-2-methyl-3-butyn-4-yl)-indeno[2',3':3,4]naphtho [1,2-b]pyran; 3-phenyl-3-(4-morpholinophenyl)-6-methoxy-7-(3-hydroxymethylene-piperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-morpholinophenyl)-6-methoxy-7-(3-(2-hydroxycarbonyl-ethyl)carboxymethylene-piperidin-1-yl)-13,13-dimethyl-3H,13H-indeno [2',3':3,4]-naphtho[1,2-b]pyran; 3-phenyl-3-(4-morpholinophenyl)-6-methoxy-7-(3-(2-hydroxycarbonyl-ethyl)carboxymethylene-piperidin-1-yl)-13,13-dimethyl-3H,13H-indeno [2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-morpholinophenyl)-6-methoxy-7-(4-hydroxypiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-morpholinophenyl)-6-methoxy-7-(4-(2-hydroxycarbonyl-ethyl)-carboxypiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4] naphtho[1,2-b]pyran; 3-phenyl-3-(4-morpholinophenyl)-6-methoxy-7-piperazinyl-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho [1,2-b]pyran; 3-phenyl-3-(4-morpholinophenyl)-6,11-dimethyl-13-hydroxy-13-(1-oxo-methoxyprop-2-yl)-1H-indeno[2,1-f]naphtho[1,2-b]pyran; 3-phenyl-3-(4-(4-phenylpiperazino)-phenyl)-6-methoxy-7-(4-hydroxypiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4] naphtho[1,2-b]pyran; 3-phenyl-3-(4-(4-phenylpiperazino)phenyl)-6-methoxy-7-(4-(2-hydroxycarbonyl-ethyl) carboxypiperidin-1-yl)-13,13-dimethyl-3H,13H-indeno[2',3':3,4] naphtho[1,2-b]pyran; 3-phenyl-3-(4-piperazinylphenyl)-6,11-dimethoxy-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-piperazinylphenyl)-6-methoxy-7-piperidinyl-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-(3-methylpiperazin-1-yl)phenyl)-6-methoxy-7-(decahydroisoquinolin-2-yl)-11-*tert*-butyl-13,13-dimethyl-3H,13H-indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-(4-methoxyphenyl)-3-(4-morpholinophenyl)-16-(ethoxycarbonyl)methyl-16-hydroxy-3,16-dihydrobenzofuro[2',3':7,8] indeno[2',3':3,4]naphtho[1,2-b]pyran; 3-phenyl-3-(4-methoxyphenyl)-16-(ethoxycarbonyl)-methyl-16-hydroxy-3,16-dihydrobenzofuro [2'',3'':6',7']indeno[3',2':4,3]naphtho[1,2-b]pyran; 3-phenyl-3-(4-morpholinophenyl)-16-(ethoxycarbonyl)-methyl-16-hydroxy-3,16-dihydrobenzofuro[2'',3'':6',7']indeno[3',2':4,3] naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-16-(ethoxycarbonyl)methyl-16-hydroxy-3,16-dihydrobenzofuro[2',3':7,8]indeno[2',3':3,4] naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-16-hydroxy-16-ethyl-16H-benzofuro[2',3':7,8] indeno[2',3':3,4]naphtho [1,2-b]pyran; 3,3-di(4-methoxyphenyl)-16-hydroxy-16H-benzofuro [2'',3'':6,7]indeno [3',2':4,3]naphtho[1,2-b]pyran; 3,3-di(4-methoxyphenyl)-16-hydroxy-16-ethyl-16H-benzofuro[2'',3'':6,7]-indeno[3',2':4,3]naphtho[1,2-b]pyran; 3-(4-methoxyphenyl)-3-(2,3-dihydrobenzofur-5-yl)-

13-hydroxy-13-methyl-indeno[2,1-f]naphtho[1,2-b]pyran; 3-(4-methoxyphenyl)-3-(2,3-dihydrobenzofur-5-yl)-6,11-difluoro-13-hydroxy-13-(1-oxo-methoxyprop-2-yl)-1H-indeno[2,1-f]naphtho[1,2-b]pyran, or a mixture thereof.

12. The method of any one of claims 1 to 11, wherein said at least one acylgermanium photoinitiator comprises any one of



13. The method of any one of claims 1 to 12, wherein the polymerizable composition comprises from 0.2% to 2.0% by weight of said at least one UV-absorbing vinylic monomer and/or said at least one UV/HEVL-absorbing vinylic monomer.
14. A photochromic silicone hydrogel contact lens obtained according to a method of any one of claims 1 to 13.
15. The photochromic silicone hydrogel contact lens of claim 14, having: an oxygen permeability of at least 40 barrers at 35°C; an elastic modulus of 2.0 MPa or less at a temperature of from 22°C to 28°C; and/or a water content of from 15% to 70% by weight at a temperature of from 22°C to 28°C when fully hydrated.