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(54) Title: CURABLE SILICONE COMPOSITIONS CONTAINING SILICA DERIVED FROM BIOGENIC MATTER

(57) Abstract: A curable silicone composition cures to form a translucent or transparent cured silicone. The curable silicone composition comprises: (I) 1% to 50% based on the weight of the composition of a non-colorant silica derived from biogenic matter and (II) 10% to 99% based on the weight of the composition of a silicone cure package.

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CURABLE SILICONE COMPOSITIONS CONTAINING SILICA DERIVED FROM BIOGENIC MATTER

CROSS-REFERENCE TO RELATED APPLICATIONS

- 5 **[0001]** This application claims the benefit of U.S. Provisional Patent Application Serial No. 60/756,409 filed on 5 January 2006. U.S. Provisional Patent Application Serial No. 60/756,409 is hereby incorporated by reference.

BACKGROUND OF THE INVENTION

Technical Field

- 10 **[0002]** This invention relates to curable silicone compositions and methods for their preparation and use. The compositions contain silica filler derived from biogenic matter. Cured products of the compositions are capable of transmitting light or capable of being pigmented to a light color, or both.

Background

- 15 **[0003]** Silica fillers derived from biogenic matter are known in the art. Previous attempts to incorporate them into curable silicone compositions have been unsuccessful because silica fillers derived from biogenic matter frequently impart black color to curable silicone compositions, and cured products thereof.

Problem to be Solved

- 20 **[0004]** There is a need to reduce the cost of producing curable silicone compositions as compared to curable silicone compositions prepared using fumed and/or precipitated silica derived from crystalline silica, which may be mined or dredged. There is a need to produce curable silicone compositions containing silica derived from biogenic matter that cure to form cured products that are capable of transmitting light or capable of being pigmented to a light
25 color, or both.

BRIEF SUMMARY OF THE INVENTION

- 30 **[0005]** This invention relates to a curable silicone composition. The curable silicone composition comprises: (I) 1% to 50% based on the weight of the composition of a non-colorant silica derived from biogenic matter and (II) 10% to 99% based on the weight of the composition of a silicone cure package. The inventors surprisingly found a non-colorant silica derived from biogenic matter allows the cured product of the curable silicone

composition to remain capable of transmitting light or capable of being pigmented to a light color, or both.

DETAILED DESCRIPTION OF THE INVENTION

[0006] All amounts, ratios, and percentages are by weight unless otherwise indicated. The articles “a”, “an”, and “the” each mean one or more. “Colorant” means any substance that imparts color to another material or mixture. The term colorant applies to black and white, as well as actual colors. “Non-colorant” means a substance that imparts low or no color to a curable composition such that the cured product thereof is either capable of transmitting light or capable of being pigmented to a light color, or both. For example, “non-colorant” includes a substance that imparts some gray color to a transparent or translucent curable composition as long as the cured product thereof is also transparent or translucent when evaluated as in example 1, described below. “Translucent” means capable of transmitting light but causing sufficient diffusion to eliminate perception of distinct images. “Transparent” means capable of transmitting light so that objects or images can be perceived. “Capable of being pigmented to a light color” means that the cured product may be a color other than black or gray, such as white or almond, when the composition contains no more than 10% of a pigment, such as titanium dioxide.

Ingredient (I) Non-colorant Silica Derived from Biogenic Matter

[0007] The curable silicone composition of this invention comprises (I) a non-colorant silica derived from biogenic matter in an amount sufficient to reduce the cost of production of the curable silicone composition (as compared to a comparable composition, which contains silica not derived from biogenic matter instead of the non-colorant silica derived from biogenic matter) but insufficient to render a cured product of the composition incapable of transmitting light or incapable of being pigmented to a light color, or both. The exact amount depends on various factors including the ingredients and amounts selected in the composition, however, the curable silicone composition of this invention may comprise an amount ranging from 1% to 50%, alternatively 1% to 40%, alternatively 1% to 35%, alternatively 1% to 20%, alternatively 5% to 20%, alternatively 5% to 30%, and alternatively 10% to 20% of ingredient (I). A cured product prepared from this composition including an amount ranging from 1 to 20% of ingredient (I) is capable of transmitting light. A cured product prepared from the composition including an amount of ingredient (I) ranging from

1% to 50% or 1% to 35% is capable of being pigmented to a light color. Without wishing to be bound by theory, it is thought that using less than 1% of the non-colorant silica derived from biogenic matter provides insufficient economic benefit, using more than 50% of a non-colorant silica derived from biogenic matter may impart a too high viscosity to the curable silicone composition, and using more than 20% of a non-colorant silica derived from biogenic matter may render a cured product of the curable silicone composition incapable of transmitting light. The cured product of the composition of this invention may be translucent or transparent.

[0008] One skilled in the art would be able to optimize the amount of ingredient (I) for a given curable silicone composition without undue experimentation. For example, amounts of ingredient (I) ranging from 1% to 50% may be used in curable silicone rubber compositions, such as high consistency rubber compositions, as exemplified below. Amounts of ingredient (I) ranging from 1% to 35% may be used in sealant compositions, such as acetoxycurable silicone sealant compositions, as exemplified below, and other moisture curable silicone sealant compositions.

[0009] Ingredient (I) may be prepared by processes known in the art, for example, the process disclosed in U.S. Patent 6,406,678. This process involves soaking a biogenic material containing amorphous silica, such as rice hulls, in an oxidizing agent and thereafter burning the biogenic material at elevated temperature (above 500 °C) but below the melting point of silica. Thereafter, the silica may be rinsed with water or mild acid solution. Before soaking, the biogenic material may be cleaned, softened, or both. The resulting silica may be white. One silica suitable for use as ingredient (I) is StratoSil™, which is commercially available from International Silica Technologies, LLC of The Woodlands, Texas, U.S.A.

[0010] Non-colorant silica suitable for use as ingredient (I) may comprise rice hull ash, wheat chaff ash, oat bran ash, or a combination thereof; alternatively rice hull ash. Ingredient (I) may be amorphous. Ingredient (I) may have a BET surface area ranging from 150 to 350 m²/g, alternatively 250 to 400 m²/g. Ingredient (I) may be white. Ingredient (I) has high purity. For example, ingredient (I) may have purity greater than or equal to 99.5% SiO₂, alternatively greater than or equal to 99.8%. Sodium content may be less than 0.03%. Iron content may be less than 0.02%. Potassium content may be less than 0.01%. Calcium

content may be less than 0.01%. Water content may be 0.5% to 1.5%, alternatively 0.88% to 1.19%.

Ingredient (II) Silicone Cure Package

5 **[0011]** The curable silicone composition of this invention cures to form a cured product that is capable of transmitting light. The curing mechanism of the silicone cure package is not critical, for example, the composition may be moisture curable such as a one part or two part room temperature vulcanizing (RTV) composition, hydrosilylation curable such as a thermally curable one part composition or a two part composition that cures at ambient or
10 elevated temperature, a peroxide curable composition, a radiation curable composition, or a combination thereof.

Moisture Cure Packages

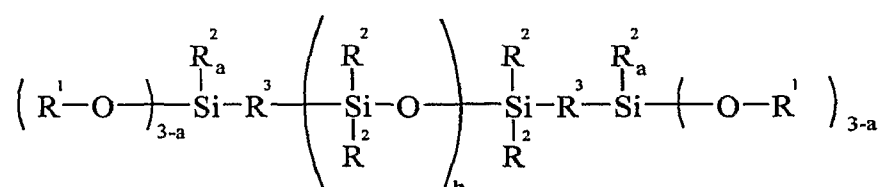
[0012] Ingredient (II) may comprise a moisture cure package. The moisture cure package may comprise: 100 parts by weight of (A) a base polymer, an amount sufficient to cure the
15 composition of (B) a crosslinking agent, and optionally an amount sufficient to accelerate curing of the composition of (C) a catalyst, where the ingredients and amounts are selected such that a cured product of the composition is capable of transmitting light or is capable of being pigmented to a light color, or both.

Ingredient (A) Base Polymer

20 **[0013]** Ingredient (A) in the moisture cure package of this invention is a polyorganosiloxane having an average per molecule of at least two hydrolyzable substituents, such as halogen atoms, acetamido groups, acyloxy groups such as acetoxy, alkoxy groups, amido groups, amino groups, aminoxy groups, hydroxyl groups, oximo groups, ketoximo groups, methylacetamido groups, alkoxysilylhydrocarbylene groups, or a combination
25 thereof. The hydrolyzable substituents in ingredient (A) may be located at terminal, pendant, or both terminal and pendant positions. Ingredient (A) may have a linear or branched structure. Ingredient (A) may be a homopolymer or a copolymer.

[0014] Ingredient (A) may comprise an alkoxy-endblocked polydiorganosiloxane, a alkoxysilylhydrocarbylene-endblocked polydiorganosiloxane, a hydroxyl-endblocked
30 polydiorganosiloxane, or a combination thereof.

[0015] Ingredient (A) may comprise a polydiorganosiloxane of Formula (I):



where each R^1 is independently a hydrolyzable substituent, each R^2 is independently a monovalent organic group, each R^3 is independently an oxygen atom or a divalent hydrocarbon group, each a is independently 0, 1, or 2, and b is an integer having a value sufficient to provide the polydiorganosiloxane with a viscosity of at least 100 mPa·s at 25 °C.

[0016] Suitable hydrolyzable substituents for R^1 include, but are not limited to, a halogen atom, an acetamido group, an acetoxy group, an acyloxy group, an alkoxy group, an amido group, an amino group, an aminoxy group, a hydroxyl group, an oximo group, a ketoximo group, and a methylacetamido group.

[0017] Suitable organic groups for R^2 include, but are not limited to, monovalent substituted and unsubstituted hydrocarbon groups. Examples of monovalent unsubstituted hydrocarbon groups for R^2 include, but are not limited to, alkyl such as methyl, ethyl, propyl, pentyl, octyl, undecyl, and octadecyl; cycloalkyl such as cyclohexyl; aryl such as phenyl, tolyl, xylyl, benzyl, and 2-phenylethyl. Examples of monovalent substituted hydrocarbon groups for R^2 include, but are not limited to, monovalent halogenated hydrocarbon groups such as chlorinated alkyl groups such as chloromethyl and chloropropyl groups; fluorinated alkyl groups such as fluoromethyl, 2-fluoropropyl, 3,3,3-trifluoropropyl, 4,4,4-trifluorobutyl, 4,4,4,3,3-pentafluorobutyl, 5,5,5,4,4,3,3-heptafluoropentyl, 6,6,6,5,5,4,4,3,3-nonafluorohexyl, and 8,8,8,7,7-pentafluorooctyl; chlorinated cycloalkyl groups such as 2,2-dichlorocyclopropyl, 2,3-dichlorocyclopentyl; and fluorinated cycloalkyl groups such as 2,2-difluorocyclopropyl, 2,3-difluorocyclobutyl, 3,4-difluorocyclohexyl, and 3,4-difluoro-5-methylcycloheptyl. Examples of monovalent substituted hydrocarbon groups for R^2 include, but are not limited to, hydrocarbon groups substituted with oxygen atoms such as glycidioxyalkyl, and hydrocarbon groups substituted with nitrogen atoms such as aminoalkyl and cyano-functional groups such as cyanoethyl and cyanopropyl. Alternatively, each R^2 may be an alkyl group.

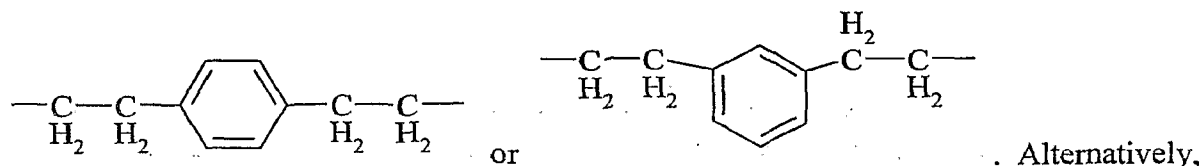
[0018] Ingredient (A) may comprise an α,ω -difunctional-polydiorganosiloxane when, in Formula (I) above, each a is 2 and each R^3 is an oxygen atom. For example, ingredient (A) may have Formula (II): $R^1R^2_2SiO-(R^2_2SiO)_b-SiR^2_2R^1$, where R^1 and R^2 are as described above and b is an integer having a value ranging from 50 to 1,000, alternatively 200 to 700.

[0019] Ingredient (A) may comprise a hydroxyl-functional polydiorganosiloxane of the formula described above, in which each R^1 may be a hydroxyl group, each R^2 may be an alkyl group such as methyl, and b may have a value such that the hydroxyl functional polydiorganosiloxane has a viscosity of at least 100 mPa·s at 25 °C. Alternatively, b may have a value ranging from 50 to 700. Exemplary hydroxyl-endblocked

polydiorganosiloxanes are hydroxyl-endblocked polydimethylsiloxanes. Hydroxyl-endblocked polydiorganosiloxanes suitable for use as ingredient (A) may be prepared by methods known in the art, such as hydrolysis and condensation of the corresponding organohalosilanes or equilibration of cyclic polydiorganosiloxanes.

[0020] Ingredient (A) may further comprise polydimethylsiloxanes endblocked on one terminal end by a triorganosilyl group, *e.g.*, $(CH_3)_3Si-$, and on the other end by a hydroxyl group. The polydiorganosiloxanes having both hydroxyl end groups and triorganosilyl end groups, may have more than 50%, alternatively more than 75%, of the total end groups as hydroxyl groups. The amount of triorganosilyl group in the polymer may be used to regulate the modulus of the resulting cured sealant. Without wishing to be bound by theory, it is thought that higher concentrations of triorganosilyl end groups provide a lower modulus in cured sealants.

[0021] Alternatively, ingredient (A) may comprise an alkoxysilylhydrocarbylene-endblocked polydiorganosiloxane, for example when in Formula (I) above each R^3 is divalent hydrocarbon group or a combination of a divalent hydrocarbon group and a divalent siloxane group. R^3 may be an alkylene group such as ethylene, propylene, or hexylene; an arylene group such as phenylene, or an alkylarylene group such as:



Alternatively, each R^1 and each R^2 may be alkyl, each R^3 may be ethylene, and a may be 0.

[0022] Ingredient (A) can be a single base polymer or a combination comprising two or more base polymers that differ in at least one of the following properties: average molecular weight, siloxane units, sequence, and viscosity.

[0023] Alkoxysilylhydrocarbylene-endblocked polydiorganosiloxanes may be prepared by reacting a vinyl-terminated, polydimethylsiloxane with (alkoxysilylhydrocarbyl)tetramethyldisiloxane. Alkoxysilylhydrocarbylene-endblocked polydiorganosiloxanes are known in the art and are disclosed in U.S. Patents 4,962,076; 5,051,455; and 5,053,442.

[0024] Ingredient (A) may further comprise an MQ resin, which comprises siloxane units of the formulae $R^1_x R^2_{3-x} SiO_{1/2}$ and $SiO_{4/2}$, where R^1 and R^2 are as described above and each instance of x is 0, 1, or 2. The MQ resin may have a ratio of M units to Q units (M:Q) of 0.5 to 1.2. Methods of preparing MQ resins are known in the art. For example, a MQ resin may be prepared by treating a product produced by the silica hydrosol capping process of Daudt, *et al.* disclosed in U.S. Patent 2,676,182. Briefly stated, the method of Daudt, *et al.* involves reacting a silica hydrosol under acidic conditions with a hydrolyzable triorganosilane such as trimethylchlorosilane, a siloxane such as hexamethyldisiloxane, or a combination thereof, and recovering a product comprising M and Q units (MQ resin). The resulting MQ resins may contain from 2 to 5 percent by weight of silicon-bonded hydroxyl groups.

Ingredient (B) Crosslinker

[0025] Ingredient (B) in the moisture cure package is a crosslinker added to the curable silicone composition in an amount sufficient to cure the curable silicone composition without too much weight loss from condensation products of the curing reaction and insufficient to allow curing of the curable silicone composition during storage for several months in a moisture impermeable package. The exact amount of ingredient (B) depends on various factors including the hydrolyzable substituents of ingredients (A) and (B), however, the amount of ingredient (B) may range from 0.5 to 15 parts based on 100 parts by weight of ingredient (A). Ingredient (B) may comprise a silane crosslinker having hydrolyzable groups or partial or full hydrolysis products thereof. Examples of suitable silane crosslinkers may have the general formula (III) $R^4_c Si(R^5)_{4-c}$, where each R^4 is independently a monovalent hydrocarbon group such as an alkyl group; each R^5 is a hydrolyzable substituent, for example a halogen atom, an acetamido group, an acyloxy group such as acetoxy, an alkoxy group, an

amido group, an amino group, an aminoxy group, a hydroxyl group, an oximo group, a ketoximo group, or a methylacetamido group; and c is 0, 1, 2, or 3. Alternatively, each R⁵ may be independently selected from hydroxyl, alkoxy, acetoxy, amide, or oxime.

Alternatively, ingredient (B) may be selected from an acyloxysilane, an alkoxysilane, a ketoximosilane, and an oximosilane.

[0026] Ingredient (B) may comprise an alkoxysilane exemplified by a dialkoxysilane, such as a dialkyldialkoxysilane; a trialkoxysilane, such as an alkyltrialkoxysilane; a tetraalkoxysilane; or partial or full hydrolysis products thereof, or another combination thereof. Examples of suitable trialkoxysilanes include methyltrimethoxysilane, methyltriethoxysilane, ethyltrimethoxysilane, ethyltriethoxysilane, and a combination thereof, and alternatively methyltrimethoxysilane. Examples of suitable tetraalkoxysilanes include tetraethoxysilane. The amount of the alkoxysilane that is used in the curable silicone composition may range from 0.5 to 15 parts by weight per 100 parts by weight of ingredient (A). Examples of alkoxysilane crosslinkers are disclosed in U.S. Patents 4,962,076; 5,051,455; and 5,053,442.

[0027] Ingredient (B) may comprise an acyloxysilane, such as an acetoxysilane. Acetoxysilanes include a tetraacetoxysilane, an organotriacetoxysilane, a diorganodiacetoxysilane, or a combination thereof. The acetoxysilane may contain alkyl groups such as methyl, ethyl, propyl, isopropyl, butyl, and tertiary butyl; alkenyl groups such as vinyl, allyl, or hexenyl; aryl groups such as phenyl, tolyl, or xylyl; aralkyl groups such as benzyl or 2-phenylethyl; and fluorinated alkyl groups such as 3,3,3-trifluoropropyl. Alternatively, ingredient (B) may comprise organotriacetoxysilanes, for example mixtures containing methyltriacetoxysilane and ethyltriacetoxysilane. The amount of the acetoxysilane that is used in the curable silicone composition may range from 0.5 to 15 parts by weight per 100 parts by weight of ingredient (A); alternatively 3 to 10 parts by weight of acetoxysilane per 100 parts by weight of ingredient (A).

Ingredient (C) Catalyst

[0028] Ingredient (C) is a catalyst that may optionally be added to the silicone cure package to accelerate curing. Ingredient (C) in the moisture cure package may comprise a carboxylic acid salt of metal, a tin compound, a titanium compound, or a zirconium compound.

[0029] Ingredient (C) may comprise carboxylic acid salts of metals, ranging from lead to manganese inclusive, in the electromotive series of metals. Alternatively, ingredient (C) may comprise a chelated titanium compound, a titanate such as a tetraalkoxytitanate, or a combination thereof. Examples of suitable titanium compounds include, but are not limited to, diisopropoxytitanium bis(ethylacetoacetate), tetrabutoxy titanate, tetrabutyltitanate, tetraisopropyltitanate, and bis-(ethoxyacetoacetate)diisopropoxy titanium (IV), and a combination thereof. Alternatively ingredient (C) may comprise a tin compound such as dibutyltin diacetate, dibutyltin dilaurate, dibutyl tin oxide, stannous octoate tin oxide, or a combination thereof. Examples of catalysts are disclosed in U.S. Patents 4,962,076; 5,051,455; and 5,053,442.

[0030] These moisture cure packages are stable when the curable silicone compositions containing them are stored in containers that protect them from exposure to moisture, but the compositions cure rapidly when exposed to moisture.

Hydrosilylation Cure Packages

[0031] Alternatively, ingredient (II) in the curable silicone composition may comprise a hydrosilylation cure package. The hydrosilylation cure package may comprise: 100 parts by weight of (A') a base polymer, an amount sufficient to cure the composition of (B') a crosslinking agent, and an amount sufficient to initiate curing of the composition of (C') a catalyst, where the ingredients and amounts are selected such that a cured product of the composition is capable of transmitting light or capable of being pigmented to a light color, or both.

Ingredient (A') Base Polymer

[0032] Ingredient (A') of the hydrosilylation cure package may comprise a polyorganosiloxane having an average of at least two aliphatically unsaturated organic groups per molecule. Ingredient (A') may have a linear or branched structure. Ingredient (A') may be a homopolymer or a copolymer. The aliphatically unsaturated organic groups may be alkenyl exemplified by, but not limited to, vinyl, allyl, butenyl, and hexenyl. The unsaturated organic groups may be alkynyl groups exemplified by, but not limited to, ethynyl, propynyl, and butynyl. The aliphatically unsaturated organic groups in ingredient (A') may be located at terminal, pendant, or both terminal and pendant positions.

[0033] The remaining silicon-bonded organic groups in ingredient (A') may be monovalent organic groups free of aliphatic unsaturation. These monovalent organic groups may have 1 to 20 carbon atoms, alternatively 1 to 10 carbon atoms, and are exemplified by, but not limited to alkyl groups such as methyl, ethyl, propyl, pentyl, octyl, undecyl, and octadecyl; cycloalkyl groups such as cyclohexyl; and aromatic groups such as phenyl, tolyl, xylyl, benzyl, and 2-phenylethyl.

[0034] Ingredient (A') may comprise a polyorganosiloxane of

Formula (IV): $R^6_2R^7SiO(R^6_2SiO)_d(R^6R^7SiO)_eSiR^6_2R^7$,

Formula (V): $R^6_3SiO(R^6_2SiO)_f(R^6R^7SiO)_gSiR^6_3$,

or a combination thereof.

[0035] In Formulae (IV) and (V), each R^6 is independently a monovalent organic group free of aliphatic unsaturation and each R^7 is independently an aliphatically unsaturated organic group, d has an average value ranging from 2 to 2000, e has an average value ranging from 0 to 2000, f has an average value ranging from 0 to 2000, and g has an average value ranging from 2 to 2000. Suitable monovalent organic groups for R^6 include, but are not limited to, alkyl such as methyl, ethyl, propyl, pentyl, octyl, undecyl, and octadecyl; cycloalkyl such as cyclohexyl; and aryl such as phenyl, tolyl, xylyl, benzyl, and 2-phenylethyl. Each R^7 is independently an aliphatically unsaturated monovalent organic group. R^7 is exemplified by alkenyl groups such as vinyl, allyl, and butenyl and alkynyl groups such as ethynyl and propynyl.

[0036] Ingredient (A') may comprise polydiorganosiloxanes such as

- i) dimethylvinylsiloxo-terminated polydimethylsiloxane,
- ii) dimethylvinylsiloxo-terminated poly(dimethylsiloxane/methylvinylsiloxane),
- iii) dimethylvinylsiloxo-terminated polymethylvinylsiloxane,
- iv) trimethylsiloxo-terminated poly(dimethylsiloxane/methylvinylsiloxane),
- v) trimethylsiloxo-terminated polymethylvinylsiloxane,
- vi) dimethylvinylsiloxo-terminated poly(dimethylsiloxane/methylphenylsiloxane),
- vii) dimethylvinylsiloxo-terminated poly(dimethylsiloxane/diphenylsiloxane),
- viii) phenyl,methyl,vinyl-siloxo-terminated polydimethylsiloxane,
- ix) dimethylhexenylsiloxo-terminated polydimethylsiloxane,
- x) dimethylhexenylsiloxo-terminated poly(dimethylsiloxane/methylhexenylsiloxane),

- xi) dimethylhexenylsiloxyl-terminated polymethylhexenylsiloxane,
- xii) trimethylsiloxyl-terminated poly(dimethylsiloxane/methylhexenylsiloxane),
- xiii) a combination thereof.

[0037] Methods of preparing polydiorganosiloxane fluids suitable for use as ingredient (A'), such as hydrolysis and condensation of the corresponding organohalosilanes or equilibration of cyclic polydiorganosiloxanes, are well known in the art.

[0038] Ingredient (A') may further comprise a resin such as an MQ resin consisting essentially of $R^8_3SiO_{1/2}$ units and $SiO_{4/2}$ units, a TD resin consisting essentially of $R^8SiO_{3/2}$ units and $R^8_2SiO_{2/2}$ units, an MT resin consisting essentially of $R^8_3SiO_{1/2}$ units and $R^8SiO_{3/2}$ units, an MTD resin consisting essentially of $R^8_3SiO_{1/2}$ units, $R^8SiO_{3/2}$ units, and $R^8_2SiO_{2/2}$ units, or a combination thereof.

[0039] Each R^8 is a monovalent organic group. The monovalent organic groups represented by R^8 may have 1 to 20 carbon atoms. Examples of monovalent organic groups include, but are not limited to, monovalent hydrocarbon groups and monovalent halogenated hydrocarbon groups. Monovalent hydrocarbon groups include, but are not limited to, alkyl such as methyl, ethyl, propyl, pentyl, octyl, undecyl, and octadecyl; cycloalkyl such as cyclohexyl; alkenyl such as vinyl, allyl, butenyl, and hexenyl; alkynyl such as ethynyl, propynyl, and butynyl; and aryl such as phenyl, tolyl, xylyl, benzyl, and 2-phenylethyl.

[0040] The resin may contain an average of 3 to 30 mole percent of aliphatically unsaturated organic groups. The aliphatically unsaturated organic groups may be alkenyl groups, alkynyl groups, or a combination thereof. The mole percent of aliphatically unsaturated organic groups in the resin is the ratio of the number of moles of unsaturated group-containing siloxane units in the resin to the total number of moles of siloxane units in the resin, multiplied by 100.

[0041] Methods of preparing resins are well known in the art. For example, resin may be prepared by treating a resin copolymer produced by the silica hydrosol capping process of Daudt, *et al.* with at least an alkenyl-containing endblocking reagent. The method of Daudt *et al.*, is disclosed in U.S. Patent 2,676,182.

[0042] Briefly stated, the method of Daudt, *et al.* involves reacting a silica hydrosol under acidic conditions with a hydrolyzable triorganosilane such as trimethylchlorosilane, a siloxane such as hexamethyldisiloxane, or mixtures thereof, and recovering a copolymer

having M and Q units. The resulting copolymers generally contain from 2 to 5 percent by weight of hydroxyl groups.

[0043] The resin, which typically contains less than 2 percent by weight of silicon-bonded hydroxyl groups, may be prepared by reacting the product of Daudt, *et al.* with an unsaturated organic group-containing endblocking agent and an endblocking agent free of aliphatic unsaturation, in an amount sufficient to provide from 3 to 30 mole percent of unsaturated organic groups in the final product. Examples of endblocking agents include, but are not limited to, silazanes, siloxanes, and silanes. Suitable endblocking agents are known in the art and exemplified in U.S. Patents 4,584,355; 4,591,622; and 4,585,836. A single endblocking agent or a mixture of such agents may be used to prepare the resin.

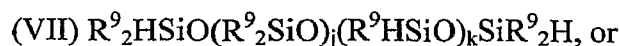
[0044] Ingredient (A') can be a single base polymer or a combination comprising two or more base polymers that differ in at least one of the following properties: structure, viscosity, average molecular weight, siloxane units, and sequence.

Ingredient (B') Organohydrogenpolysiloxane

[0045] Ingredient (B') in the hydrosilylation cure package is an organohydrogenpolysiloxane having an average of at least two silicon-bonded hydrogen atoms per molecule. The amount of ingredient (B') in the hydrosilylation cure package may range from 0.5 to 15 parts per 100 parts by weight of ingredient (A'). Ingredient (B') can be a homopolymer or a copolymer. Ingredient (B') can have a linear, branched, cyclic, or resinous structure. The silicon-bonded hydrogen atoms in ingredient (B') can be located at terminal, pendant, or at both terminal and pendant positions.

[0046] Ingredient (B') may comprise siloxane units including, but not limited to, $\text{HR}^9_2\text{SiO}_{1/2}$, $\text{R}^9_3\text{SiO}_{1/2}$, $\text{HR}^9\text{SiO}_{2/2}$, $\text{R}^9_2\text{SiO}_{2/2}$, $\text{R}^9\text{SiO}_{3/2}$, and $\text{SiO}_{4/2}$ units. In the preceding formulae, each R^9 is independently selected from monovalent organic groups free of aliphatic unsaturation.

[0047] Ingredient (B') may comprise a compound of the formula



(VIII) a combination thereof.

[0048] In formulae above, h has an average value ranging from 0 to 2000, i has an average value ranging from 2 to 2000, j has an average value ranging from 0 to 2000, and k has an

average value ranging from 0 to 2000. Each R⁹ is independently a monovalent organic group. Suitable monovalent organic groups include alkyl such as methyl, ethyl, propyl, pentyl, octyl, undecyl, and octadecyl; cycloalkyl such as cyclohexyl; alkenyl such as vinyl, allyl, butenyl, and hexenyl; alkynyl such as ethynyl, propynyl, and butynyl; and aryl such as phenyl, tolyl, xylyl, benzyl, and 2-phenylethyl.

[0049] Ingredient (B') is exemplified by

- a) dimethylhydrogensiloxy-terminated polydimethylsiloxane,
- b) dimethylhydrogensiloxy-terminated poly(dimethylsiloxane/methylhydrogensiloxane),
- c) dimethylhydrogensiloxy-terminated polymethylhydrogensiloxane,
- d) trimethylsiloxy-terminated poly(dimethylsiloxane/methylhydrogensiloxane),
- e) trimethylsiloxy-terminated polymethylhydrogensiloxane,
- f) a resin consisting essentially of H(CH₃)₂SiO_{1/2} units and SiO_{4/2} units, and
- g) a combination thereof.

[0050] Ingredient (B') may be a combination of two or more organohydrogenpolysiloxanes that differ in at least one of the following properties: structure, average molecular weight, viscosity, siloxane units, and sequence. Ingredient (B') may be added in an amount ranging from 0.4 to 20% based on the weight of the hydrosilylation cure package.

[0051] Methods of preparing linear, branched, and cyclic organohydrogenpolysiloxanes suitable for use as ingredient (B'), such as hydrolysis and condensation of organohalosilanes, are well known in the art. Methods of preparing organohydrogenpolysiloxane resins suitable for use as ingredient (B') are also well known as exemplified in U.S. Patents 5,310,843; 4,370,358; and 4,707,531.

Ingredient (C') Hydrosilylation Catalyst

[0052] Ingredient (C') of the hydrosilylation cure package is a hydrosilylation catalyst.

Ingredient (C') is added to the hydrosilylation cure package in an amount of 0.1 to 1000 ppm of platinum group metal, alternatively 1 to 500 ppm, alternatively 2 to 200, alternatively 5 to 150 ppm, based on the weight of the curable silicone composition.

[0053] Suitable hydrosilylation catalysts are known in the art and commercially available.

Ingredient (C') may comprise a platinum group metal selected from platinum, rhodium, ruthenium, palladium, osmium or iridium metal or organometallic compound thereof, or a combination thereof. Ingredient (C') is exemplified by compounds such as chloroplatinic

acid, chloroplatinic acid hexahydrate, platinum dichloride, and complexes of said compounds with low molecular weight organopolysiloxanes or platinum compounds microencapsulated in a matrix or coreshell type structure. Complexes of platinum with low molecular weight organopolysiloxanes include 1,3-diethenyl-1,1,3,3 -tetramethyldisiloxane complexes with platinum. These complexes may be microencapsulated in a resin matrix. Alternatively, the catalyst may comprise 1,3-diethenyl-1,1,3,3 -tetramethyldisiloxane complex with platinum. When the catalyst is a platinum complex with a low molecular weight organopolysiloxane, the amount of catalyst may range from 0.04 to 0.4% based on the weight of the curable silicone composition.

[0054] Suitable hydrosilylation catalysts for ingredient (C') are described in, for example, U.S. Patents 3,159,601; 3,220,972; 3,296,291; 3,419,593; 3,516,946; 3,814,730; 3,989,668; 4,784,879; 5,036,117; and 5,175,325 and EP 0 347 895 B. Microencapsulated hydrosilylation catalysts and methods of preparing them are known in the art, as exemplified in U.S. Patent No. 4,766,176; and U.S. Patent No. 5,017,654.

Peroxide Cure Packages

[0055] Alternatively, ingredient (II) may comprise a peroxide cure package. The peroxide cure package may comprise: 100 parts by weight of (A'') a base polymer, optionally an amount sufficient to cure the composition of (B'') a crosslinking agent, and an amount sufficient to accelerate curing of the composition of (C'') a catalyst, where the ingredients and amounts are selected such that a cured product of the composition is capable of transmitting light or capable of being pigmented to a light color, or both.

Ingredient (A'') Base Polymer

[0056] Ingredient (A'') of the peroxide cure package comprises a polydiorganosiloxane having an average of at least two aliphatically unsaturated organic groups per molecule. The polydiorganosiloxane may have a viscosity of at least 100 mPa·s at 25 °C. The polydiorganosiloxane may comprise a polydiorganosiloxane gum having a viscosity, measured as Williams Plasticity Number in accordance with ASTM D-962 of greater than 100, which is equivalent to 4×10^6 mPa·s at 25 °C. Alternatively, viscosity may range from 125 to 200 Williams Plasticity number (corresponding to 10×10^6 to 80×10^6 mPa·s at 25 °C).

[0057] The aliphatically unsaturated organic groups may be alkenyl exemplified by, but not limited to, vinyl, allyl, butenyl, and hexenyl. The aliphatically unsaturated organic groups may be alkynyl groups exemplified by, but not limited to, ethynyl, propynyl, and butynyl. The unsaturated organic groups in ingredient (A'') may be located at terminal, pendant, or both terminal and pendant positions.

[0058] The remaining silicon-bonded organic groups in ingredient (A''') may be monovalent organic groups free of aliphatic unsaturation. These monovalent organic groups are exemplified by, but not limited to alkyl groups such as methyl, ethyl, propyl, pentyl, octyl, undecyl, and octadecyl; cycloalkyl groups such as cyclohexyl; and aromatic groups such as phenyl, tolyl, xylyl, benzyl, and 2-phenylethyl.

[0059] Ingredient (A'') may comprise a polydiorganosiloxane of

Formula (IX): $R^{10}_2R^{11}SiO(R^9_2SiO)_m(R^9R^{11}SiO)_nSiR^9_2R^{11}$,

Formula (X): $R^{10}_3SiO(R^{10}_2SiO)_o(R^{10}R^{11}SiO)_pSiR^{10}_3$,

or a combination thereof.

[0060] In formulae (IX) and (X), each R^{10} is independently a monovalent organic group free of aliphatic unsaturation, each R^{11} is independently an aliphatically unsaturated organic group, m has an average value of at least 2, n may be 0 or a positive number, o may be 0 or a positive number, and p has an average value of at least 2, with the proviso that the subscripts have values sufficient to give the polydiorganosiloxanes of formulae (IX) and (X) Williams Plasticity Numbers greater than 100. Suitable monovalent organic groups for R^{10} include, but are not limited to, alkyl such as methyl, ethyl, propyl, pentyl, octyl, undecyl, and octadecyl; cycloalkyl such as cyclohexyl; and aryl such as phenyl, tolyl, xylyl, benzyl, and 2-phenylethyl. Each R^{11} is independently an aliphatically unsaturated monovalent organic group. R^{11} is exemplified by alkenyl groups such as vinyl, allyl, and butenyl and alkynyl groups such as ethynyl and propynyl.

[0061] Ingredient (A'') may be a combination of two or more polydiorganosiloxanes that differ in at least one of the following properties: structure, average molecular weight, viscosity, siloxane units, and sequence.

Optional Ingredient (B'') Crosslinking Agent

[0062] Ingredient (B'') a crosslinking agent may optionally be added to the peroxide cure package to improve compression set of a silicone elastomer prepared by curing this

composition. The amount of ingredient (B'') in the composition may range from 0 to 15 parts per 100 parts by weight of ingredient (A''). Ingredient (B'') may comprise a polydiorganohydrogensiloxane having an average of at least two silicon-bonded hydrogen atoms per molecule.

- 5 [0063] Ingredient (B'') may comprise a polydiorganohydrogensiloxane of the formula
 (XI) $R^{12}_3SiO(R^{12}_2SiO)_q(R^{12}HSiO)_rSiR^{12}_3$,
 (XII) $R^{12}_2HSiO(R^{12}_2SiO)_s(R^{12}HSiO)_tSiR^{12}_2H$, or
 (XIII) a combination thereof.

10 [0064] In formulae above, q has an average value ranging from 0 to 2000, r has an average value ranging from 2 to 2000, s has an average value ranging from 0 to 2000, and t has an average value ranging from 0 to 2000, with the provisos that $(q + r) < 2000$ and $(s + t) < 2000$. Each R^{12} is independently a monovalent organic group. Suitable monovalent organic groups include alkyl such as methyl, ethyl, propyl, pentyl, octyl, undecyl, and octadecyl; cycloalkyl such as cyclohexyl; alkenyl such as vinyl, allyl, butenyl, and hexenyl; alkynyl
 15 such as ethynyl, propynyl, and butynyl; and aryl such as phenyl, tolyl, xylyl, benzyl, and 2-phenylethyl.

[0065] Ingredient (B'') is exemplified by

- i) dimethylhydrogensiloxy-terminated polydimethylsiloxane,
 ii) dimethylhydrogensiloxy-terminated poly(dimethylsiloxane/methylhydrogensiloxane),
 20 iii) dimethylhydrogensiloxy-terminated polymethylhydrogensiloxane,
 iv) trimethylsiloxy-terminated poly(dimethylsiloxane/methylhydrogensiloxane),
 v) trimethylsiloxy-terminated polymethylhydrogensiloxane,
 vi) a combination thereof.

[0066] Ingredient (B'') may be a combination of two or more
 25 polydiorganohydrogensiloxanes that differ in at least one of the following properties: structure, average molecular weight, viscosity, siloxane units, and sequence.

Ingredient (C'') Catalyst

[0067] Ingredient (C'') in the peroxide cure package comprises a peroxide compound. The amount of ingredient (C'') added to the composition depends on the specific peroxide
 30 compound selected for ingredient (C''), however, the amount may range from 0.2 to 5 parts per 100 parts by weight of ingredient (A''). Examples of peroxide compounds suitable for

ingredient (C") include, but are not limited to 2,4-dichlorobenzoyl peroxide, dicumyl peroxide, and a combination thereof; as well as combinations of such a peroxide with a benzoate compound such as tertiary-butyl perbenzoate.

[0068] Suitable peroxide cure packages are known in the art, and are disclosed in, for example, U.S. Patent 4,774,281. The fume silica filler in the composition of U.S. Patent 4,774,281 may be partially replaced by ingredient (I).

Optional Ingredients

[0069] The curable silicone compositions of this invention may further comprise one or more additional ingredients in addition to ingredients (I) and (II), provided that the ingredient and amount thereof does not render a cured product of the composition incapable of transmitting light. The composition may further comprise an additional ingredient selected from the group consisting of (III) a reinforcing filler, (IV) an extending filler, (V) a conductive filler (*e.g.*, electrically conductive, thermally conductive, or both), (VI) a filler treating agent, (VII) a stabilizer (*e.g.*, a hydrosilylation cure stabilizer, a heat stabilizer, or a UV stabilizer), (VIII) a plasticizer, (IX), an extender (sometimes referred to as a secondary plasticizer or processing aid), (X) an adhesion promoter, (XI) a fungicide, (XII) a rheological additive, (XIII) a flame retardant, (XIV) a pigment, and a combination thereof.

[0070] For example, ingredient (III), a reinforcing filler, may be added in addition to ingredient (I) in an amount ranging from 1% to 35%, alternatively 1% to 15%, based on the weight of the composition when the composition will be formulated such that it cures to form a high consistency rubber. Examples of suitable reinforcing fillers include reinforcing silica fillers not derived from biogenic matter such as fume silica, silica aerogel, silica xerogel, and precipitated silica. Fumed silicas not derived from biogenic matter are known in the art and commercially available; fumed silica sold under the name CAB-O-SIL by Cabot Corporation of Massachusetts.

[0071] The composition may optionally further comprise ingredient (IV) an extending filler in an amount ranging from 1% to 60%, alternatively 1% to 20%, based on the weight of the composition. Examples of extending fillers include crushed quartz, aluminum oxide, magnesium oxide, calcium carbonate, zinc oxide, talc, diatomaceous earth, iron oxide, clays, titanium dioxide, zirconia, sand, carbon black, graphite, or a combination thereof. Extending

fillers are known in the art and commercially available; such as a ground silica sold under the name MIN-U-SIL by U.S. Silica of Berkeley Springs, WV.

[0072] The composition may optionally further comprise ingredient (VI), a filler treating agent in an amount ranging from 0.1% to 15%, alternatively 0.5% to 5%, based on the weight of the composition. Ingredient (I), and when present ingredients (III), (IV), and (V) may optionally be surface treated with ingredient (VI). Ingredients (I), (III), (IV), and (V) may be treated with ingredient (VI) before being added to the composition, or *in situ*. Ingredient (VI) may comprise an alkoxysilane, an alkoxy-functional oligosiloxane, a cyclic polyorganosiloxane, a hydroxyl-functional oligosiloxane such as a dimethyl siloxane or methyl phenyl siloxane, or a fatty acid. Examples of stearates include calcium stearate. Examples of filler treating agents and methods for their use are disclosed in, for example, EP 1 101 167 A2 and U.S. Patents 5,051,455, 5,053,442, and 6,169,142 (col. 4, line 42 to col. 5, line 2).

[0073] Ingredient (VII) is a stabilizer. Stabilizers for hydrosilylation cure packages are exemplified by acetylenic alcohols such as methyl butynol, ethynyl cyclohexanol, dimethyl hexynol, 1,1-dimethyl-2-propynyl)oxy)trimethylsilane, methyl(tris(1,1-dimethyl-2-propynyloxy))silane, and a combination thereof; cycloalkenylsiloxanes such as methylvinylcyclotetrasiloxanes exemplified by 1,3,5,7-tetramethyl-1,3,5,7-tetravinylcyclotetrasiloxane, 1,3,5,7-tetramethyl-1,3,5,7-tetrahexenylcyclotetrasiloxane, and a combination thereof; ene-yne compounds such as 3-methyl-3-penten-1-yne, 3,5-dimethyl-3-hexen-1-yne; triazoles such as benzotriazole; phosphines; mercaptans; hydrazines; amines such as tetramethyl ethylenediamine, dialkyl fumarates, dialkenyl fumarates, dialkoxyalkyl fumarates, maleates such as diallyl maleate, and a combination thereof. Alternatively, the stabilizer may comprise phenyl butynol. Suitable hydrosilylation cure package stabilizers are disclosed by, for example, U.S. Patents 3,445,420; 3,989,667; 4,584,361; and 5,036,117.

[0074] The amount of stabilizer added to the curable silicone composition will depend on the particular stabilizer used and the composition and amount of ingredient (II). However, the amount of hydrosilylation cure package stabilizer may range from 0.0025% to 0.025% based on the weight of the curable silicone composition.

[0075] One skilled in the art would recognize that the curable silicone composition may comprise more than one cure package. For example a dual cure package that is both moisture

curable and hydrosilylation curable is within the scope of this invention. One skilled in the art would be able to select ingredients and amounts thereof in each cure package described above to prepare a cured product that has a desired consistency, such as a rubber or gel.

Method of Making the Curable Silicone Composition

5 [0076] The curable silicone composition may be prepared as a one part composition, for example, by combining all ingredients by any convenient means, such as mixing. Alternatively, the curable silicone composition may be prepared as a multiple part composition in which the crosslinking agent and catalyst are stored in separate parts, and the parts are combined shortly before use of the composition. For example, a two part curable
10 silicone composition may be prepared by combining ingredients comprising (A), (C), and any optional ingredients in a first part by any convenient means such as mixing. A second part may be prepared by combining ingredients comprising (A), (B), and any optional ingredients by any convenient means such as mixing. The non-colorant silica derived from biogenic matter may be added to the first part, the second part, or both. The ingredients may be
15 combined at ambient or elevated temperature and under ambient or anhydrous conditions, depending on the silicone cure package selected. One skilled in the art would be able to prepare a curable silicone composition without undue experimentation.

Method of Using the Curable Silicone Composition

[0077] Curable silicone compositions prepared according to this invention have various
20 uses. One skilled in the art would be able to select ingredients to formulate a composition, for example, for use as a sealant or a curable silicone rubber composition, based on the description herein without undue experimentation.

EXAMPLES

[0078] These examples are included to illustrate the invention to one of ordinary skill in the
25 art. However, those of skill in the art should, in light of the present disclosure, appreciate that many changes can be made in the specific embodiments disclosed and still obtain a like or similar result without departing from the spirit and scope of the invention set forth in the claims. The following ingredients are used in the examples.

[0079] Base Polymer 1 was α,ω -dihydroxy-polydimethylsiloxane.

30 [0080] Base Polymer 2 was dimethylvinylsiloxyl-terminated poly(dimethylsiloxane/methylvinylsiloxane).

[0081] Base Polymer 3 was dimethylvinylsiloxo-terminated polydimethylsiloxane.

[0082] Base Polymer 4 is dimethylvinylsiloxo-terminated polydimethylsiloxane.

[0083] Base Polymer 5 is dimethylvinyl-siloxo terminated,
poly(dimethylsiloxane/methylvinylsiloxane).

5 [0084] Treating Agent 1 is hexamethyldisilazane.

[0085] Crosslinker 1 was a mixture of methyltriacetoxysilane and ethyltriacetoxysilane.

[0086] Crosslinker 2 is poly(dimethylsiloxane/methylhydrogensiloxane) with methyl
silsesquioxane.

[0087] Catalyst 1 was dibutyltin diacetate.

10 [0088] Catalyst 2 is 1,3-diethenyl-1,1,3,3 -tetramethyldisiloxane complex with platinum.

[0089] Silica was StratoSil™, which is commercially available from International Silica
Technologies, LLC of The Woodlands, Texas, U.S.A.

[0090] Plasticizer 1 was hydroxy-terminated poly(dimethylsiloxane/methylvinylsiloxane).

[0091] Plasticizer 2 was hydroxy-terminated polydimethylsiloxane.

15 [0092] Stabilizer 1 was ethynyl cyclohexanol.

Example 1 Acetoxy Curable Silicone Sealant Compositions

[0093] Curable silicone sealant compositions were prepared by mixing the ingredients in
Table 1 for 1 minute using a whip mixer and de-airing the resulting mixtures under vacuum
for 1 minute. The samples were packaged in plastic screw cap containers.

20 Table 1

	Base Polymer 1	Crosslinker 1	Catalyst 1	Silica
Sample 1	85.5%	4.5%	0.015%	10%
Sample 2	80.5%	4.5%	0.015%	15%
Sample 3	74.5%	5.5%	0.015%	20%

[0094] Each sample was spread out on white paper and allowed to cure. Upon visual
observation, appearance was grainy. Sample 1 appeared white and translucent. Samples 2
and 3 appeared gray and translucent. All cured samples had low elongation.

25 [0095] Example 1 shows that the Silica appears to be a semi-reinforcing filler in this
formulation. The Silica is non-colorant in this formulation.

Example 2 – High Consistency Rubber Composition

[0096] Samples were prepared by mixing the ingredients in Table 2 under a nitrogen atmosphere with a small amount of ammonium carbonate (to accelerate silanol condensation between silanol groups on the plasticizers and the Silica) until the Silica was incorporated to form a base. Thereafter 1.2 parts of a 2,4-dichlorobenzoylperoxide catalyst per hundred parts of the base were mixed together to form a curable silicone composition.

Table 2

Sample	Base Polymer 2	Base Polymer 3	Plasticizer 1	Plasticizer 2	Silica
Sample 4	50 parts	50 parts	2 parts	6 parts	51 parts

[0097] Sample 4 was cured and evaluated for tensile strength, % elongation, specific gravity, durometer, tear strength, and compression set at 22 hours and 177 °C. The results are in Table 3.

Table 3

Cure Conditions	Median Tensile Strength (MPa)	Median % Elongation	Specific Gravity	Durometer	Tear Strength (kN/m)	Compression Set
5 min at 116 °C	2.8	321	1.2	32.8	5.06	70.6
with 4 hour 200 °C Post Cure	2.9	288	1.2	32.2	5.08	21.4

Example 3 – Low Consistency Silicone Rubber Composition

[0098] A composition was prepared by mixing 571.9 parts Base Polymer 4, 26 parts water, 0.9 parts Plasticizer 1, and 39.4 parts Treating Agent 1 for 5 minutes. Thereafter, 131.5 parts Silica was added and mixed until incorporated. Thereafter, 23.7 parts Treating Agent 1 and 131.5 parts Silica were added and mixed until the Silica is incorporated. Thereafter, another 131.5 parts Silica were added and mixed until incorporated. Thereafter 13.2 parts Treating

Agent 1 were added and mixed. The resulting mixture is heated at 160 °C for 1 hour and 45 minutes. Thereafter, 200.1 parts Base Polymer 4, 105.2 parts Base Polymer 5, 0.5 parts Stabilizer 1, and 14.9 parts Crosslinker 2 were added. The resulting mixture was mixed for 15 minutes and stored in a metal can. 100 parts of this mixture were taken from the metal can and mixed with 0.15 parts Catalyst 2. The resulting curable silicone composition was heated at 150 °C for 5 minutes and then post cured by heating at 200 °C for 4 hours. Sample 5 was evaluated for tensile strength, % elongation, specific gravity, durometer, tear strength, and compression set at 22 hours and 177 °C. The results are in Table 4.

Table 4

Cure Conditions	Median Tensile Strength (MPa)	Median % Elongation	Specific Gravity	Durometer	Tear Strength (kN/m)	Compression Set
5 min at 150 °C	2.8	198	1.2	44	4.6	9.4
with 4 hour 200 °C Post Cure	2.6	225	1.2	43	4.9	11

Example 4 – Water Content of Rice Hull Ash

[0099] Three particle sizes of Silica were dried in an oven at 150 °C for 12 hours. The weight loss (water content) is shown in Table 5.

Table 5

Silica sample particle size (microns)	Weight Loss (%)
4	1.19
15	0.88
50	1.13

Industrial Applicability

[0100] Using a non-colorant silica derived from biogenic matter in a curable silicone composition may provide the benefits of reduced cost (as compared to a curable silicone composition containing only fillers derived from non-biogenic matter) while maintaining
5 translucency or transparency of the curable silicone composition and cured products thereof. The non-colorant silica derived from biogenic matter may be used in combination with silica derived from other sources to reduce cost. The non-colorant silica derived from biogenic matter may be used to replace all or a portion of silica in commercially available products. For example, such products include moisture curable products such as DOW CORNING®
10 732, 3330, 3-7132 Premium, 3440, 1199, and 899; and hydrosilylation curable products such as DOW CORNING® SYLGARD® 182, 184, and 186, all of which are available from Dow Corning Corporation of Midland, Michigan, U.S.A. to reduce cost without impairing capability of transmitting light.

CLAIMS:

1. A composition comprising:

- (I) 1% to 50% based on the weight of the composition of a non-colorant silica derived from biogenic matter, and
(II) a silicone cure package.

2. The composition of claim 1, where the composition contains 1% to 20% based on the weight of the composition of ingredient (I) and the composition is curable to form a cured product capable of transmitting light.

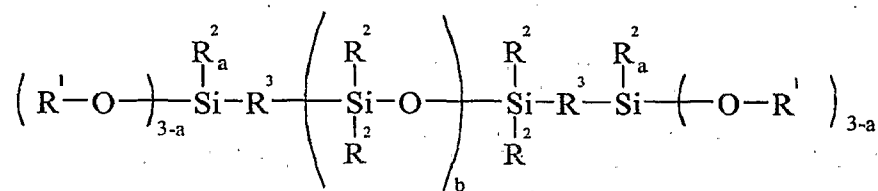
3. The composition of claim 1, where ingredient (I) is rice hull ash.

4. The composition of claim 1, where the silicone cure package comprises:

- 100 parts by weight of (A) a base polymer,
optionally an amount sufficient to cure the composition of (B) a crosslinking agent, and
optionally an amount sufficient to initiate curing of the composition of (C) a catalyst.

5. The composition of claim 4, where ingredient (A) comprises a polyorganosiloxane having an average per molecule of at least two hydrolyzable substituents, and ingredient (B) comprises a silane crosslinker having hydrolyzable groups or partial or full hydrolysis products thereof.

6. The composition of claim 5, where ingredient (A) comprises a polydiorganosiloxane of Formula (I):



where each R^1 is independently a hydrolyzable substituent, each R^2 is independently a monovalent organic group, each R^3 is independently an oxygen atom or a divalent

hydrocarbon group, each a is independently 0, 1, or 2, and b is an integer having a value sufficient to provide the polydiorganosiloxane with a viscosity of at least 100 mPa·s at 25 °C; and ingredient (B) is present, and ingredient (B) comprises a silane of formula $R^4_cSi(R^5)_{4-c}$, where each R^4 is independently a monovalent hydrocarbon group, each R^5 is independently a hydrolyzable substituent, and c is 0, 1, 2, or 3.

7. The composition of claim 6, where ingredient (C) is present, and ingredient (C) is selected from the group consisting of a carboxylate, a carboxylic acid salt of metal, a titanate, and a zirconate.

8. The composition of claim 4, where ingredient (A) is a polyorganosiloxane having an average of at least two aliphatically unsaturated organic groups per molecule, ingredient (B) is present, ingredient (B) is a polyorganohydrogensiloxane having an average of at least two silicon bonded hydrogen atoms per molecule, ingredient (C) is present, and ingredient (C) is a hydrosilylation catalyst.

9. The composition of claim 4, where ingredient (A) is a polydiorganosiloxane having an average of at least two aliphatically unsaturated organic groups per molecule, ingredient (C) is present, and ingredient (C) is a peroxide compound.

10. The composition of claim 1, further comprising an additional ingredient selected from the group consisting of (III) a reinforcing filler, (IV) an extending filler, (V) a conductive filler, (VI) a filler treating agent, (VII) a stabilizer, (VIII) a plasticizer, (IX), an extender, (X) an adhesion promoter, (XI) a fungicide, (XII) a rheological additive, (XIII) a flame retardant, (XIV) a pigment, and a combination thereof.

11. A method of making the composition of any one of claims 1-10 comprising mixing all the ingredients.

12. Use of the composition of any one of claims 1-10 as a sealant.

13. Use of the composition of any one of claims 1-10 as a curable silicone rubber composition.

INTERNATIONAL SEARCH REPORT

International application No

PCT/US2006/044053

A. CLASSIFICATION OF SUBJECT MATTER
 INV. C08K3/36 C08L83/04

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
 C08K C08L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, COMPENDEX, CHEM ABS Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	V. P. E SILVA; I. V. P. YOSHIDA; M. DO C. GONÇALVES; M. RODRIGUES: "Poster E-P8: BIOGENIC SILICA MICROFIBERS / SILICONE COMPOSITE" SBPMAT BRAZIL-MRS. 2ND BRAZILIAN MRS MEETING SYMPOSIUM E: PROCESSING/PROPERTIES STRUCTURAL COMPOSITE MATERIALS, 26 October 2003 (2003-10-26), - 29 October 2003 (2003-10-29) XP002426984 abstract	1-3,11, 13
X	US 4 382 057 A (TOLENTINO LUISITO A) 3 May 1983 (1983-05-03) examples claims 1,6 ----- -/--	1-11,13

☒ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

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Date of the actual completion of the international search

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INTERNATIONAL SEARCH REPORT

International application No

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