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(54) Title: HYDROSILYLATION REACTION CURABLE COMPOSITIONS AND METHODS FOR THEIR PREPARATION AND USE

(57) Abstract: A composition contains (A) a hydrosilylation reaction catalyst and (B) an aliphatically unsaturated compound having an average, per molecule, of one or more aliphatically unsaturated organic groups capable of undergoing hydrosilylation reaction. The composition is capable of reacting via hydrosilylation reaction to form a reaction product, such as a silane, a gum, a gel, a rubber, or a resin. Ingredient (A) contains a platinum-ligand complex that can be prepared by reacting a platinum precursor and a ligand.



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HYDROSILYLATION REACTION CURABLE COMPOSITIONS AND METHODS FOR THEIR PREPARATION AND USE

CROSS-REFERENCE TO RELATED APPLICATIONS and STATEMENT REGARDING FEDERALLY SPONSORED RESEARCH

[0001] None.

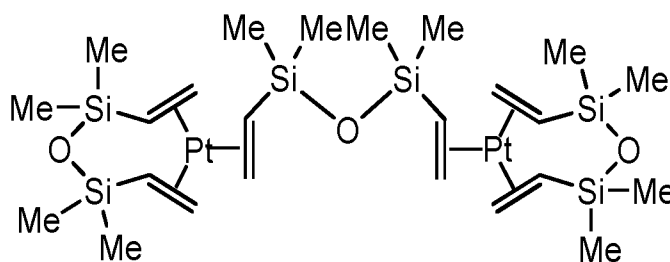
TECHNICAL FIELD

[0002] Hydrosilylation reaction curable compositions contain new catalysts. The new catalysts are capable of catalyzing hydrosilylation reaction of the compositions without conventional platinum group metal hydrosilylation catalysts.

BACKGROUND

[0003] Catalysts for catalyzing hydrosilylation reaction are platinum group metal catalysts, which known in the art and are commercially available. Such hydrosilylation catalysts can be a metal selected from platinum, rhodium, ruthenium, palladium, osmium, and iridium.

Alternatively, the hydrosilylation catalyst may be a compound of such a metal, for example, 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 coresheath type structure. Complexes of platinum with low molecular weight organopolysiloxanes include 1,3-diethenyl-1,1,3,3 -tetramethyldisiloxane complexes with platinum as described in US Patent 3,775,452. This provides tris(divinyldisiloxane) diplatinum catalyst $\text{Pt}_2[(\text{ViSiMe}_2)_2\text{O}]_3$ with the following structure;



Other exemplary hydrosilylation catalysts are described in U.S. Patents 3,159,601; 3,220,972; 3,296,291; 3,419,593; 3,516,946; 3,814,730; 4,784,879; and 5,175,325 and EP 0 347 895 B.

[0004] These hydrosilylation catalysts suffer from the drawback of being extremely costly. Some of the metals in these hydrosilylation catalysts may also be difficult to obtain, and some of these hydrosilylation catalysts may be difficult to prepare. There is a need in industry to replace the conventional hydrosilylation catalysts described above with a less expensive and/or more readily available alternative. There is further a need to provide efficient catalysts, giving high yields, fast cure or good selectivity in the hydrosilylation reaction.

BRIEF SUMMARY OF THE INVENTION

[0005] A hydrosilylation catalyst comprises a platinum-ligand complex. The complex can be formed of $\text{Pt}_3[\text{O}(\text{SiMe}_2\text{C}\equiv\text{C-R})_2]_2$. R can be H (I), Ph (II), t-Bu (III) or SiMe₃ (IV).

[0006] The hydrosilylation catalyst is useful in a composition comprising:

- (A) the platinum-ligand complex, and
- (B) an aliphatically unsaturated compound having an average, per molecule, of one or more aliphatically unsaturated organic groups capable of undergoing hydrosilylation reaction. When ingredient (B) does not contain silicon bonded hydrogen atoms, then the composition further comprises
- (C) an SiH functional compound having an average, per molecule, of one or more silicon bonded hydrogen atoms.

[0007] Ingredient (A) is capable of catalyzing hydrosilylation reaction of the composition to form a reaction product. The reaction product may be a silane, gel, rubber, or resin.

DETAILED DESCRIPTION OF THE INVENTION

Definitions And Usage Of Terms

[0008] All amounts, ratios, and percentages are by weight unless otherwise indicated. The articles 'a', 'an', and 'the' each refer to one or more, unless otherwise indicated by the context of specification. The disclosure of ranges includes the range itself and also anything subsumed therein, as well as endpoints. For example, disclosure of a range of 2.0 to 4.0 includes not only the range of 2.0 to 4.0, but also 2.1, 2.3, 3.4, 3.5, and 4.0 individually, as well as any other number subsumed in the range. Furthermore, disclosure of a range of, for

example, 2.0 to 4.0 includes the subsets of, for example, 2.1 to 3.5, 2.3 to 3.4, 2.6 to 3.7, and 3.8 to 4.0, as well as any other subset subsumed in the range. Similarly, the disclosure of Markush groups includes the entire group and also any individual members and subgroups subsumed therein. For example, disclosure of the Markush group a hydrogen atom, an alkyl group, an aryl group, an aralkyl group, or an alkaryl group includes the member alkyl individually; the subgroup alkyl and aryl; and any other individual member and subgroup subsumed therein.

[0009] The abbreviation "cSt" means centiStokes. The abbreviation "Et" means ethyl group. The abbreviation "Hex" means hexyl group. The abbreviation "Me" means methyl group. The abbreviation "Ph" means phenyl group. The abbreviation "t-Bu" means tert-butyl group. The abbreviation "Vi" means vinyl group. The abbreviation "Pa·s" means Pascal seconds. The abbreviation "ppm" means parts per million. The abbreviation "cod" means 1,5-Cyclooctadiene.

[0010] The designation "M unit" means a siloxane unit having formula $R_3SiO_{1/2}$, where each R independently represents a monovalent atom or organic group. The designation "D unit" means a siloxane unit having formula $R_2SiO_{2/2}$, where each R independently represents a monovalent atom or group. The designation "T unit" means a siloxane unit having formula $RSiO_{3/2}$, where each R independently represents a monovalent atom or group. The designation "Q unit" means a siloxane unit having formula $SiO_{4/2}$.

[0011] "Non-functional" means that the ingredient does not participate in a hydrosilylation reaction.

[0012] "Aralkyl" refers to an alkyl group having a pendant and/or terminal aryl group or an aryl group having a pendant an alkyl group. Exemplary aralkyl groups include benzyl, phenylethyl, phenyl propyl, and phenyl butyl.

[0013] "Carbocycle" and "carbocyclic" refer to a hydrocarbon ring. Carbocycles may be monocyclic or alternatively may be fused, bridged, or spiro polycyclic rings. Monocyclic carbocycles may have 3 to 9 carbon atoms, alternatively 4 to 7 carbon atoms, and alternatively 5 to 6 carbon atoms. Polycyclic carbocycles may have 7 to 17 carbon atoms, alternatively 7 to 14 carbon atoms, and alternatively 9 to 10 carbon atoms. Carbocycles may be saturated or unsaturated. Carbocycles may be saturated or unsaturated.

[0014] "Cycloalkyl" refers to a saturated carbocycle. Cycloalkyl groups are exemplified by cyclobutyl, cyclopentyl, and cyclohexyl.

- 5 [0015] "Heterocycle" and "heterocyclic" refer to a ring group comprised of carbon atoms and one or more heteroatoms in the ring. The heteroatom may be N, O, P, S, or a combination thereof. Heterocycles may be monocyclic or alternatively may be fused, bridged, or spiro polycyclic rings. Monocyclic heterocycles may have 3 to 9 member atoms in the ring, alternatively 4 to 7 member atoms, and alternatively 5 to 6 member atoms.
- 10 Polycyclic heterocycles may have 7 to 17 member atoms, alternatively 7 to 14 member atoms, and alternatively 9 to 10 member atoms. Heterocycles may be saturated or unsaturated.

- [0016] The conventional platinum group metal catalysts are those described above in the
- 15 BACKGROUND section. Alternatively, the composition described herein may be free of hydrosilylation reaction catalysts (*i.e.*, other than ingredient (A) described herein).

Composition

- 20 [0017] A composition comprises:
- (A) a hydrosilylation reaction catalyst, and
 - (B) an aliphatically unsaturated compound having an average, per molecule, of one or more aliphatically unsaturated organic groups capable of undergoing hydrosilylation reaction.

25

- [0018] The composition is capable of reacting via hydrosilylation reaction to form a reaction product. When ingredient (B) does not contain a silicon bonded hydrogen atom, then the composition further comprises ingredient (C), an SiH functional compound having an average, per molecule, of one or more silicon bonded hydrogen atoms, which is distinct from
- 30 ingredients (A) and (B).

- [0019] The composition may optionally further comprise one or more additional ingredients, which are distinct from ingredients (A), (B), and (C) described above. Suitable additional ingredients are exemplified by (D) a spacer; (E) an extender, a plasticizer, or a
- 35 combination thereof; (F) a filler; (G) a filler treating agent; (H) a biocide; (I) an inhibitor, (J) a

flame retardant; (K) a surface modifier; (L) a chain lengthener; (M) an endblocker; (N) a flux agent; (O) an anti-aging additive; (P) a pigment; (Q) an acid acceptor (R) a rheological additive; (S) a solvent; (T) a surfactant; and a combination thereof.

- 5 **[0020]** The composition reacts via hydrosilylation reaction to form a reaction product. The reaction product may have various forms, such as a silane, a gum, a gel, a rubber, or a resin.

Platinum Compound

10

The invention provides a platinum compound containing

- i) a Pt atom and
 - ii) a ligand having at least one atom of Si, at least one atom of O, and at least one alkyne group. This platinum compound can be used as Ingredient (A) of the
- 15 composition.

Ingredient (A) Hydrosilylation Catalyst

- 20 **[0021]** Ingredient (A) comprises a Pt containing hydrosilylation reaction catalyst. The Pt containing hydrosilylation reaction catalyst comprises a Pt-ligand complex. The Pt-ligand complex can be prepared by reacting a Pt precursor and a ligand. The Pt precursor is an organic compound of Pt having general formula (i): $\text{Pt}(\text{A})_a$, where

- Pt is a platinum atom;
 - each A is independently a monovalent organic group or a complexing group or
- 25 compound; and
- subscript a has a value of 2.

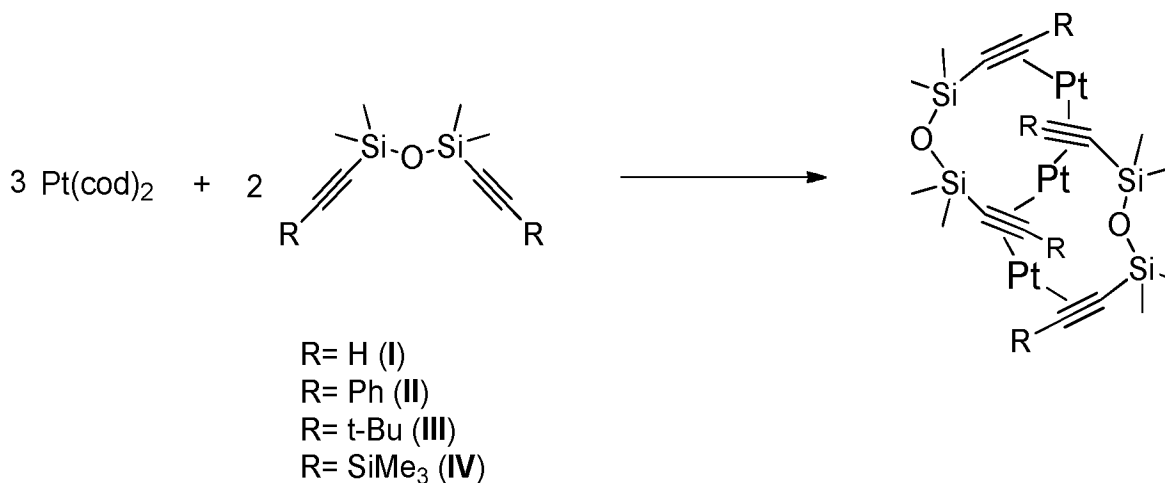
- 30 **[0022]** Examples of monovalent hydrocarbon groups for A include, but are not limited to, alkyl such as methyl, ethyl, propyl, pentyl, hexyl, heptyl, ethylhexyl, octyl, decyl, dodecyl, undecyl, and octadecyl; alkenyl such as vinyl, allyl, propenyl, and hexenyl; cycloalkyl such as cyclopentyl and cyclohexyl; aryl such as phenyl, tolyl, xylyl, and naphthyl; and aralkyl benzyl and 2-phenylethyl. The complexing group or compound can be ethyl, benzyl, mesityl, phenyl, $-\text{NEt}_2$, cyclooctadiene, ethoxide, iso-propoxide, butoxide, 2-ethylhexanoate, neodecanoate, octanoate, and stearate. Subscript a may have a value ranging from 0 to 2.
- 35 Example of preferred complexing group is 1,5 cyclooctadiene (cod). Ingredient (A) may thus

comprise *cod*. The ligand is an organic compound that coordinates with Pt. The ligand has at least one Si atom, one O atom and one alkyne group. The ligand may have general formula (I):

O(SiMe₂C≡C-R). R can be H (I), Ph (II), t-Bu (III) or SiMe₃ (IV).

5 and the Pt hydrosilylation catalyst may be:

[0023] Ingredient (A) may be prepared by a method comprising reacting a ligand and a Pt precursor, described above, thereby forming a reaction product comprising a Pt-ligand complex. The reaction is for example:



Combining the Pt precursor and the ligand may be performed by any convenient means, such as mixing or shaking the container.

[0024] Reacting the Pt precursor and ligand may be performed by under any convenient conditions such as allowing the Pt precursor and ligand prepared as described above to react at room temperature (RT) of 25 °C for a period of time, such as 24 hours, or by heating. Heating may be performed by any convenient means, such as placing the container in an oven. The reaction temperature depends on various factors including the reactivities of the specific Pt precursor and ligand selected and the Ligand:Metal Ratio, however, temperature may range from 25 °C to 200 °C, alternatively 25 °C to 75 °C. Reaction time depends on various factors including the reaction temperature selected, however, reaction time may range from 1 minute to 48 hours, alternatively 10 hours to 30 hours. The ligand and Pt precursor may be combined and heated sequentially. Alternatively, the ligand and Pt precursor may be combined and heated concurrently.

[0025] By-products and/or solvent may be removed by any convenient means, such as stripping or distillation, with heating or under vacuum, or a combination thereof. The resulting isolated Pt-ligand complex may be used as ingredient (A).

5 [0026] Alternatively, the reaction by-products are not removed before using the reaction product as ingredient (A). For example, the ligand and Pt precursor may be reacted as described above, with or without solvent removal, and the resulting reaction product (comprising the Pt-ligand complex and the reaction by-product and optionally a solvent) may be used as ingredient (A). Without wishing to be bound by theory, it is thought that a by-
10 product may act as a hydrosilylation reaction catalyst, or as a co-catalyst or an activator, in addition to the Pt-ligand complex. Therefore, the reaction product may catalyze a hydrosilylation reaction.

[0027] The composition may contain one single catalyst. Alternatively, the composition
15 may comprise two or more catalysts described above as ingredient (A), wherein the two or more catalysts differ in at least one property such as selection of ligand, selection of precursor, Ligand: Platinum Ratio, and definitions for group A in formula (i). The composition may be free of hydrosilylation reaction catalysts other than ingredient (A).

20 **Ingredient (B) Aliphatically Unsaturated Compound**

[0028] Ingredient (B) is an aliphatically unsaturated compound having an average, per molecule, of one or more aliphatically unsaturated organic groups capable of undergoing hydrosilylation reaction. Alternatively, ingredient (B) may have an average of two or more
25 aliphatically unsaturated organic groups per molecule. The aliphatically unsaturated organic groups may be alkenyl exemplified by, but not limited to, vinyl, allyl, propenyl, butenyl, and hexenyl. The unsaturated organic groups may be alkynyl groups exemplified by, but not limited to, ethynyl, propynyl, and butynyl.

30 [0029] Ingredient (B) of the composition may be an unsaturated hydrocarbon, where the unsaturated group is capable of reacting via hydrosilylation reaction. Ingredient (B) may be monomeric. For example, suitable aliphatically unsaturated organic compounds for ingredient (B) include, but are not limited to alkenes such as 1-pentene, 1-hexene, 1-heptene; and halogenated alkenes, such as allyl chloride.

[0030] Alternatively, ingredient (B) of the composition may be polymeric. Ingredient (B) may comprise a base polymer having an average of one or more aliphatically unsaturated organic groups per molecule. The base polymer for ingredient (B) may be a silicon containing base polymer having a linear, branched, cyclic, or resinous structure.

5 Alternatively, the base polymer may have a linear and/or branched structure. Alternatively, the base polymer may have a resinous structure. The base polymer may be a homopolymer or a copolymer. Ingredient (B) may be one base polymer. Alternatively, ingredient (B) may comprise two or more base polymers differing in at least one of the following properties: structure, viscosity, average molecular weight, siloxane units, and sequence. The
10 aliphatically unsaturated organic groups in the base polymer may be located at terminal, pendant, or both terminal and pendant positions.

[0031] The remaining silicon-bonded organic groups in the base polymer for ingredient (B) may be monovalent organic groups free of aliphatic unsaturation. Examples of monovalent
15 hydrocarbon groups include, but are not limited to, alkyl such as methyl, ethyl, propyl, butyl, pentyl, hexyl, heptyl, octyl, decyl, dodecyl, undecyl, and octadecyl; cycloalkyl such as cyclopentyl and cyclohexyl; aryl such as phenyl, tolyl, xylyl, and naphthyl; alkaryl such as benzyl; and aralkyl such as 1-phenylethyl and 2-phenylethyl. Examples of monovalent halogenated hydrocarbon groups include, but are not limited to, chlorinated alkyl groups
20 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
25 other monovalent organic groups include, but are not limited to, hydrocarbon groups substituted with oxygen atoms such as glycidoxyalkyl, and hydrocarbon groups substituted with nitrogen atoms such as aminoalkyl and cyano-functional groups such as cyanoethyl and cyanopropyl.

30

[0032] Ingredient (B) may comprise a polydiorganosiloxane of

- Formula (I): $R^1_2R^2SiO(R^1_2SiO)_a(R^1R^2SiO)_bSiR^1_2R^2$,
 - Formula (II): $R^1_3SiO(R^1_2SiO)_c(R^1R^2SiO)_dSiR^1_3$,
- or a combination thereof.

35

[0033] In formulae (I) and (II), each R¹ is independently a hydrogen atom or a monovalent organic group free of aliphatic unsaturation and each R² is independently an aliphatically unsaturated organic group, exemplified by those described above. Subscript a may be 0 or a positive number. Alternatively, subscript a has an average value of at least 2.

5 Alternatively subscript a may have a value ranging from 2 to 2000. Subscript b may be 0 or a positive number. Alternatively, subscript b may have an average value ranging from 0 to 2000. Subscript c may be 0 or a positive number. Alternatively, subscript c may have an average value ranging from 0 to 2000. Subscript d has an average value of at least 2. Alternatively subscript d may have an average value ranging from 2 to 2000. Suitable
10 monovalent organic groups for R¹ are as described above for ingredient (B). Alternatively, each R¹ is a monovalent hydrocarbon group exemplified by alkyl such as methyl and aryl such as phenyl. Each R² is independently an aliphatically unsaturated monovalent organic group as described above for ingredient (B). Alternatively, R² is exemplified by alkenyl groups such as vinyl, allyl, butenyl, and hexenyl; and alkynyl groups such as ethynyl and
15 propynyl.

[0034] Ingredient (B) may comprise a polydiorganosiloxane such as

- i) dimethylvinylsiloxyl-terminated polydimethylsiloxane,
- ii) dimethylvinylsiloxyl-terminated poly(dimethylsiloxane/methylvinylsiloxane),
- 20 iii) dimethylvinylsiloxyl-terminated polymethylvinylsiloxane,
- iv) trimethylsiloxyl-terminated poly(dimethylsiloxane/methylvinylsiloxane),
- v) trimethylsiloxyl-terminated polymethylvinylsiloxane,
- vi) dimethylvinylsiloxyl-terminated poly(dimethylsiloxane/methylphenylsiloxane),
- vii) dimethylvinylsiloxyl-terminated poly(dimethylsiloxane/diphenylsiloxane),
- 25 viii) phenyl,methyl,vinyl-siloxyl-terminated polydimethylsiloxane,
- ix) dimethylhexenylsiloxyl-terminated polydimethylsiloxane,
- x) dimethylhexenylsiloxyl-terminated poly(dimethylsiloxane/methylhexenylsiloxane),
- xi) dimethylhexenylsiloxyl-terminated polymethylhexenylsiloxane,
- xii) trimethylsiloxyl-terminated poly(dimethylsiloxane/methylhexenylsiloxane),
- 30 xiii) a combination thereof.

[0035] Methods of preparing polydiorganosiloxane fluids suitable for use as ingredient (B), such as hydrolysis and condensation of the corresponding organohalosilanes or equilibration of cyclic polydiorganosiloxanes, are well known in the art.

[0036] In addition to, or instead of, the polydiorganosiloxane described above, ingredient (B) may further comprise a resin such as an MQ resin consisting essentially of $R^3_3SiO_{1/2}$ units and $SiO_{4/2}$ units, a TD resin consisting essentially of $R^3SiO_{3/2}$ units and $R^3_2SiO_{2/2}$ units, an MT resin consisting essentially of $R^3_3SiO_{1/2}$ units and $R^3SiO_{3/2}$ units, an MTD resin consisting essentially of $R^3_3SiO_{1/2}$ units, $R^3SiO_{3/2}$ units, and $R^3_2SiO_{2/2}$ units, or a combination thereof.

[0037] Each R^3 is a monovalent organic group exemplified by those described above for ingredient (B). Alternatively, the monovalent organic groups represented by R^3 may have 1 to 20 carbon atoms. Alternatively, examples of monovalent organic groups for R^3 include, but are not limited to, monovalent hydrocarbon groups and monovalent halogenated hydrocarbon groups.

[0038] 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.

[0039] 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.

[0040] 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.

[0041] The resin, which typically contains less than 2 % 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.

Ingredient (C) SiH Functional Compound

[0042] Ingredient (C) in the composition may be a silane or an

organohydrogenpolysiloxane having an average, per molecule, of one or more silicon bonded hydrogen atoms. Alternatively, ingredient (C) may have an average, per molecule, of at least two silicon-bonded hydrogen atoms. The amount of ingredient (C) in the composition depends on various factors including the SiH content of ingredient (C), the unsaturated group content of ingredient (B), and the properties of the reaction product of the composition desired, however, the amount of ingredient (C) may be sufficient to provide a molar ratio of SiH groups in ingredient (C) to aliphatically unsaturated organic groups in ingredient (B) (commonly referred to as the SiH:Vi ratio) ranging from 0.3:1 to 5:1.

Ingredient (C) can have a monomeric or polymeric structure. When ingredient (C) has a polymeric structure, the polymeric structure may be linear, branched, cyclic, or resinous structure. When ingredient (C) is polymeric, then ingredient (C) can be a homopolymer or a copolymer. The silicon-bonded hydrogen atoms in ingredient (C) can be located at terminal, pendant, or at both terminal and pendant positions. Ingredient (C) may be one SiH functional compound. Alternatively, ingredient (C) may comprise a combination of two or more SiH functional compounds. Ingredient (C) may be two or more

organohydrogenpolysiloxanes that differ in at least one of the following properties: structure, average molecular weight, viscosity, siloxane units, and sequence.

[0043] Ingredient (C) may comprise a silane of formula $R^4_eSiH_f$, where subscript e is 0, 1, 2, or 3; subscript f is 1, 2, 3, or 4, with the proviso that a sum of (e + f) is 4. Each R^4 is

independently a halogen atom or a monovalent organic group. Suitable halogen atoms for R^4 are exemplified by chlorine, fluorine, bromine, and iodine; alternatively chlorine. Suitable monovalent organic groups for R^4 include, but are not limited to, monovalent hydrocarbon and monovalent halogenated hydrocarbon groups. Monovalent hydrocarbon groups include, but are not limited to, alkyl such as methyl, ethyl, propyl, butyl, pentyl, hexyl, heptyl, octyl, decyl, dodecyl, undecyl, and octadecyl; cycloalkyl such as cyclopentyl and cyclohexyl; aryl

such as phenyl, tolyl, xylol, and naphthyl; alkaryl such as benzyl; and aralkyl such as 1-phenylethyl and 2-phenylethyl. Examples of monovalent halogenated hydrocarbon groups include, but are not limited to, 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 other monovalent organic groups include, but are not limited to, hydrocarbon groups substituted with oxygen atoms such as glycidoxyalkyl, and alkoxy groups such as methoxy, ethoxy, propoxy, and butoxy; and hydrocarbon groups substituted with nitrogen atoms such as aminoalkyl and cyano-functional groups such as cyanoethyl and cyanopropyl. Examples of suitable silanes for ingredient (C) are exemplified by trichlorosilane (HSiCl_3), Me_2HSiCl , or MeHSi(OMe)_2 .

[0044] Alternatively, ingredient (C) may be a polyorganohydrogensiloxane comprising siloxane units including, but not limited to, $\text{HR}^5_2\text{SiO}_{1/2}$, $\text{R}^5_3\text{SiO}_{1/2}$, $\text{HR}^5\text{SiO}_{2/2}$, $\text{R}^5_2\text{SiO}_{2/2}$, $\text{R}^5\text{SiO}_{3/2}$, and $\text{SiO}_{4/2}$ units. In the preceding formulae, each R^5 is independently selected from the monovalent organic groups free of aliphatic unsaturation described above.

[0045] Ingredient (C) may comprise a polyorganohydrogensiloxane of

- Formula (III): $\text{R}^5_3\text{SiO}(\text{R}^5_2\text{SiO})_g(\text{R}^5\text{HSiO})_h\text{SiR}^5_3$,
- Formula (IV): $\text{R}^5_2\text{HSiO}(\text{R}^5_2\text{SiO})_i(\text{R}^5\text{HSiO})_j\text{SiR}^5_2\text{H}$, or

a combination thereof.

[0046] In formulae (III) and (IV) above, subscript g has an average value ranging from 0 to 2000, subscript h has an average value ranging from 2 to 2000, subscript i has an average value ranging from 0 to 2000, and subscript j has an average value ranging from 0 to 2000. Each R^5 is independently a monovalent organic group, as described above.

[0047] Polyorganohydrogensiloxanes for ingredient (C) are exemplified by:

- a) dimethylhydrogensiloxyl-terminated polydimethylsiloxane,
- b) dimethylhydrogensiloxyl-terminated poly(dimethylsiloxane/methylhydrogensiloxane),

- c) dimethylhydrogensiloxy-terminated polymethylhydrogensiloxane,
- d) trimethylsiloxy-terminated poly(dimethylsiloxane/methylhydrogensiloxane),
- e) trimethylsiloxy-terminated polymethylhydrogensiloxane,
- f) a resin consisting essentially of $\text{H}(\text{CH}_3)_2\text{SiO}_{1/2}$ units and $\text{SiO}_{4/2}$ units, and
- 5 g) a combination thereof.

[0048] Methods of preparing linear, branched, and cyclic organohydrogenpolysiloxanes suitable for use as ingredient (C), such as hydrolysis and condensation of organohalosilanes, are well known in the art. Methods of preparing
10 organohydrogenpolysiloxane resins suitable for use as ingredient (C) are also well known as exemplified in U.S. Patents 5,310,843; 4,370,358; and 4,707,531.

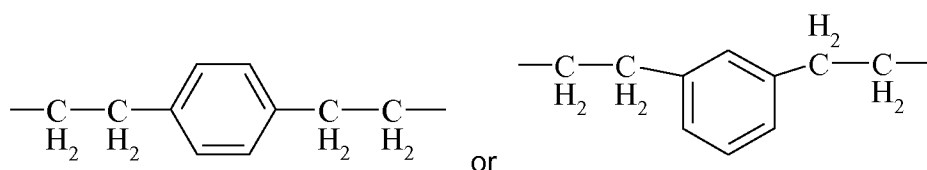
[0049] The composition may optionally further comprise one or more additional ingredients, *i.e.*, in addition ingredients (A) and (B), and optionally (C). The one or more
15 additional ingredients are distinct from ingredients (A), (B) and (C). Suitable additional ingredients are exemplified by (D) a spacer; (E) an extender, a plasticizer, or a combination thereof; (F) a filler; (G) a filler treating agent; (H) a biocide; (I) an inhibitor, (J) a flame retardant; (K) a surface modifier; (L) a chain lengthener; (M) an endblocker; (N) a flux agent; (O) an anti-aging additive; (P) a pigment; (Q) an acid acceptor (R) a rheological additive; (S)
20 a solvent; (T) a surfactant; and a combination thereof.

Ingredient (D) Spacer

[0050] Ingredient (D) is a spacer. Spacers can comprise organic particles, inorganic
25 particles, or a combination thereof. Spacers can be thermally conductive, electrically conductive, or both. Spacers can have a desired particle size, for example, particle size may range from 25 micrometers to 125 micrometers. Spacers can comprise monodisperse beads, such as glass or polymer (*e.g.*, polystyrene) beads. Spacers can comprise thermally conductive fillers such as alumina, aluminum nitride, atomized metal powders, boron nitride,
30 copper, and silver. The amount of ingredient (D) depends on various factors including the particle size distribution, pressure to be applied during use of the composition or the cured product prepared therefrom, temperature during use, and desired thickness of the composition or the cured product prepared therefrom. However, the composition may contain an amount of ingredient (D) ranging from 0.05 % to 2 %, alternatively 0.1 % to 1 %.

Ingredient (E) Extender

[0051] Ingredient (E) is an extender and/or a plasticizer. An extender comprising a non-functional polyorganosiloxane may be used in the composition. For example, the non-functional polyorganosiloxane may comprise difunctional units of the formula $R^6_2SiO_{2/2}$ and terminal units of the formula $R^7_3SiR^{28}$, where each R^6 and each R^7 are independently a monovalent organic group such as a monovalent hydrocarbon group exemplified by alkyl such as methyl, ethyl, propyl, and butyl; alkenyl such as vinyl, allyl, and hexenyl; aryl such as phenyl, tolyl, xylyl, and naphthyl; and aralkyl groups such as phenylethyl; and R^{28} is an oxygen atom or a divalent group linking the silicon atom of the terminal unit with another silicon atom. The divalent linking group for R^{28} may be a divalent organic group, a silicone organic group, or a combination of a divalent hydrocarbon group and a divalent siloxane group. Alternatively, each R^{28} may be independently selected from an oxygen atom and a divalent hydrocarbon group. Alternatively, each R^{28} may be an oxygen atom. Alternatively, each R^{28} may be a divalent hydrocarbon group exemplified by an alkylene group such as ethylene, propylene, butylene, or hexylene; an arylene group such as phenylene, or an alkylarylene group such as:

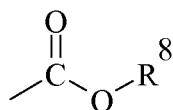


[0052] Alternatively, an instance of R^{28} may be an oxygen atom while a different instance of R^{28} is a divalent hydrocarbon group. Non-functional polyorganosiloxanes are known in the art and are commercially available. Suitable non-functional polyorganosiloxanes are exemplified by, but not limited to, polydimethylsiloxanes. Such polydimethylsiloxanes include DOW CORNING® 200 Fluids, which are commercially available from Dow Corning Corporation of Midland, Michigan, U.S.A. and may have viscosity ranging from 50 cSt to 100,000 cSt, alternatively 50 cSt to 50,000 cSt, and alternatively 12,500 cSt to 60,000 cSt.

[0053] An organic plasticizer may be used in addition to, or instead of, the non-functional polyorganosiloxane extender described above. Organic plasticizers are known in the art and are commercially available. The organic plasticizer may comprise a phthalate, a carboxylate, a carboxylic acid ester, an adipate or a combination thereof. The organic plasticizer may be selected from the group consisting of: bis(2-ethylhexyl) terephthalate; bis(2-ethylhexyl)-1,4-benzenedicarboxylate; 2-ethylhexyl methyl-1,4-benzenedicarboxylate;

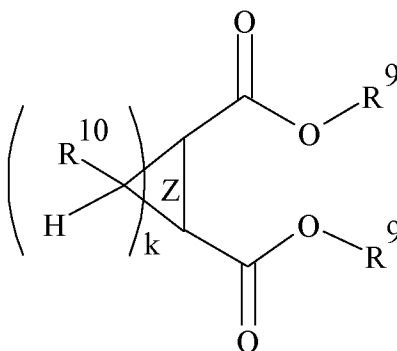
1,2 cyclohexanedicarboxylic acid, dinonyl ester, branched and linear; bis(2-propylheptyl) phthalate; diisononyl adipate; and a combination thereof.

[0054] The organic plasticizer may have an average, per molecule, of at least one group of formula



where R^8 represents a hydrogen atom or a monovalent organic group. Alternatively, R^8 may represent a branched or linear monovalent hydrocarbon group. The monovalent organic group may be a branched or linear monovalent hydrocarbon group such as an alkyl group of 4 to 15 carbon atoms, alternatively 9 to 12 carbon atoms. Suitable plasticizers may be selected from the group consisting of adipates, carboxylates, phthalates, and a combination thereof.

[0055] Alternatively, the organic plasticizer may have an average, per molecule, of at least two groups of the formula above bonded to carbon atoms in a cyclic hydrocarbon. The organic plasticizer may have general formula:



In this formula, group Z represents a cyclic hydrocarbon group having 3 or more carbon atoms, alternatively 3 to 15 carbon atoms. Subscript k may have a value ranging from 1 to 12. Group Z may be saturated or aromatic. Each R^{10} is independently a hydrogen atom or a branched or linear monovalent organic group. The monovalent organic group for R^9 may be an alkyl group such as methyl, ethyl, or butyl. Alternatively, the monovalent organic group for R^{10} may be an ester functional group. Each R^9 is independently a branched or linear monovalent hydrocarbon group, such as an alkyl group of 4 to 15 carbon atoms.

[0056] Suitable organic plasticizers are known in the art and are commercially available. The plasticizer may comprise a phthalate, such as: a dialkyl phthalate such as dibutyl

phthalate, diheptyl phthalate, di(2-ethylhexyl) phthalate, or diisodecyl phthalate (DIDP), bis(2-propylheptyl) phthalate, di(2-ethylhexyl) phthalate, dimethyl phthalate; diethyl phthalate; butyl benzyl phthalate, and bis(2-ethylhexyl) terephthalate; a dicarboxylate such as 1,2,4-benzenetricarboxylic acid, bis(2-ethylhexyl)-1,4-benzenedicarboxylate; 2-ethylhexyl methyl-1,4-benzenedicarboxylate; 1,2 cyclohexanedicarboxylic acid, dinonyl ester, branched and linear; diisononyl adipate; trimellitates such as trioctyl trimellitate; triethylene glycol bis(2-ethylhexanoate); triacetin; nonaromatic dibasic acid esters such as dioctyl adipate, bis(2-ethylhexyl) adipate, di-2-ethylhexyladipate, dioctyl sebacate, dibutyl sebacate and diisodecyl succinate; aliphatic esters such as butyl oleate and methyl acetyl recinolate; phosphates such as tricresyl phosphate and tributyl phosphate; chlorinated paraffins; hydrocarbon oils such as alkylidiphenyls and partially hydrogenated terphenyls; process oils; epoxy plasticizers such as epoxidized soybean oil and benzyl epoxystearate; tris(2-ethylhexyl) ester; a fatty acid ester; and a combination thereof. Examples of suitable plasticizers and their commercial sources include those listed below in the table below.

Table of Exemplary Organic Plasticizers and Commercial Sources

Product Name	%	Component
Eastman(TM) 425 Plasticizer	75 %	bis(2-ethylhexyl) terephthalate
Eastman(TM) 168 Plasticizer	> 98 %	bis(2-ethylhexyl)-1,4-benzenedicarboxylate
	< 2 %	2-ethylhexyl methyl-1,4-benzenedicarboxylate
Eastman(TM) 168-CA Plasticizer	> 97 %	bis(2-ethylhexyl)-1,4-benzenedicarboxylate
	< 2 %	2-ethylhexyl methyl-1,4-benzenedicarboxylate
BASF Hexamoll *DINCH	> 99.5 %	1,2 cyclohexanedicarboxylic acid, dinonyl ester, branched and linear
BASF Palatinol® DPHP	99.9 %	bis(2-propylheptyl) phthalate or Di-(2-Propyl Heptyl) Phthalate
BASF Palamoll® 652	96.0 %	PMN00-0611
	4.0 %	diisononyl adipate
Eastman 168 Xtreme (TM) Plasticizer	100 %	Plasticizer
Eastman(TM) TOTM Plasticize	> 99.9 %	trioctyl trimellitate
Eastman(TM) TEG-EH Plasticizer	100 %	triethylene glycol bis(2-ethylhexanoate)
Eastman(TM) DOP Plasticizer	100 %	di(2-ethylhexyl) phthalate
Eastman(TM) Triacetin	100 %	Triacetin
Eastman(TM) DOA Plasticizer	100 %	bis(2-ethylhexyl) adipate
Eastman(TM) DOA Plasticizer, Kosher	100 %	bis(2-ethylhexyl) adipate
Eastman(TM) DMP Plasticizer	100 %	dimethyl phthalate
Eastman(TM) DEP Plasticizer	100 %	diethyl phthalate
Eastman(TM) DBP Plasticizer	100 %	dibutyl phthalate
BASF Plastomoll® DOA	> 99.5 %	Di-2-ethylhexyladipate
BASF Palatinol® TOTM-I	> 99 %	1,2, 4-Benzenetricarboxylic acid, tris(2-ethylhexyl) ester
Ferro SANTICIZER® 261A	> 99.5 %	Benzyl, C7-C9 linear and branched alkyl esters, 1, 2, benzene dicarboxylic acid

[0057] Alternatively, a polymer plasticizer can be used. Examples of the polymer

- 5 plasticizer include alkenyl polymers obtained by polymerizing vinyl or allyl monomers by means of various methods; polyalkylene glycol esters such as diethylene glycol dibenzoate, triethylene glycol dibenzoate and pentaerythritol ester; polyester plasticizers obtained from dibasic acids such as sebacic acid, adipic acid, azelaic acid and phthalic acid and dihydric
- 10 alcohols such as ethylene glycol, diethylene glycol, triethylene glycol, propylene glycol and dipropylene glycol; polyethers including polyether polyols each having a molecular weight of not less than 500 such as polyethylene glycol, polypropylene glycol and polytetramethylene

glycol, polystyrenes such as polystyrene and poly-alpha-methylstyrene; and polybutadiene, polybutene, polyisobutylene, butadiene acrylonitrile, and polychloroprene.

[0058] The polyorganosiloxane extenders and organic plasticizers described above for ingredient (E) may be used either each alone or in combinations of two or more thereof. A low molecular weight organic plasticizer and a higher molecular weight polymer plasticizer may be used in combination. The exact amount of ingredient (E) used in the composition will depend on various factors including the desired end use of the composition and the cured product thereof. However, the amount of ingredient (E) may range from 0.1 % to 10 % based on the combined weights of all ingredients in the composition.

Ingredient (F) Filler

[0059] Ingredient (F) is a filler. The filler may comprise a reinforcing filler, an extending filler, a conductive filler, or a combination thereof. For example, the composition may optionally further comprise ingredient (f1), a reinforcing filler, which when present may be added in an amount ranging from 0.1 % to 95 %, alternatively 1 % to 60 %, based on the weight of the composition. The exact amount of ingredient (f1) depends on various factors including the form of the reaction product of the composition (e.g., gel or rubber) and whether any other fillers are added. Examples of suitable reinforcing fillers include chopped fibre such as chopped KEVLAR®, and/or reinforcing silica fillers such as fume silica, silica aerogel, silica xerogel, and precipitated silica. Fumed silicas are known in the art and commercially available; e.g., fumed silica sold under the name CAB-O-SIL by Cabot Corporation of Massachusetts, U.S.A.

[0060] The composition may optionally further comprise ingredient (f2) an extending filler in an amount ranging from 0.1 % to 95 %, alternatively 1 % to 60 %, and alternatively 1 % to 20 %, based on the weight of the composition. Examples of extending fillers include crushed quartz, aluminum oxide, magnesium oxide, calcium carbonate such as precipitated calcium carbonate, zinc oxide, talc, diatomaceous earth, iron oxide, clays, mica, 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. Suitable precipitated calcium carbonates included Winnofil® SPM from Solvay and Ultrapflex® and Ultrapflex® 100 from SMI.

[0061] The composition may optionally further comprise ingredient (f3) a conductive filler. Ingredient (F) may be both thermally conductive and electrically conductive. Alternatively, ingredient (F) may be thermally conductive and electrically insulating. Ingredient (F) may be selected from the group consisting of aluminum nitride, aluminum oxide, aluminum trihydrate, barium titanate, beryllium oxide, boron nitride, carbon fibres, diamond, graphite, magnesium hydroxide, magnesium oxide, metal particulate, onyx, silicon carbide, tungsten carbide, zinc oxide, and a combination thereof. Ingredient (F) may comprise a metallic filler, an inorganic filler, a meltable filler, or a combination thereof. Metallic fillers include particles of metals and particles of metals having layers on the surfaces of the particles. These layers may be, for example, metal nitride layers or metal oxide layers on the surfaces of the particles. Suitable metallic fillers are exemplified by particles of metals selected from the group consisting of aluminum, copper, gold, nickel, silver, and combinations thereof, and alternatively aluminum. Suitable metallic fillers are further exemplified by particles of the metals listed above having layers on their surfaces selected from the group consisting of aluminum nitride, aluminum oxide, copper oxide, nickel oxide, silver oxide, and combinations thereof. For example, the metallic filler may comprise aluminum particles having aluminum oxide layers on their surfaces.

[0062] Inorganic conductive fillers are exemplified by onyx; aluminum trihydrate, metal oxides such as aluminum oxide, beryllium oxide, magnesium oxide, and zinc oxide; nitrides such as aluminum nitride and boron nitride; carbides such as silicon carbide and tungsten carbide; and combinations thereof. Alternatively, inorganic conductive fillers are exemplified by aluminum oxide, zinc oxide, and combinations thereof. Meltable fillers may comprise Bi, Ga, In, Sn, or an alloy thereof. The meltable filler may optionally further comprise Ag, Au, Cd, Cu, Pb, Sb, Zn, or a combination thereof. Examples of suitable meltable fillers include Ga, In-Bi-Sn alloys, Sn-In-Zn alloys, Sn-In-Ag alloys, Sn-Ag-Bi alloys, Sn-Bi-Cu-Ag alloys, Sn-Ag-Cu-Sb alloys, Sn-Ag-Cu alloys, Sn-Ag alloys, Sn-Ag-Cu-Zn alloys, and combinations thereof. The meltable filler may have a melting point ranging from 50 °C to 250 °C, alternatively 150 °C to 225 °C. The meltable filler may be a eutectic alloy, a non-eutectic alloy, or a pure metal. Meltable fillers are commercially available.

[0063] For example, meltable fillers may be obtained from Indium Corporation of America, Utica, N.Y., U.S.A.; Arconium, Providence, R.I., U.S.A.; and AIM Solder, Cranston, R.I., U.S.A. Aluminum fillers are commercially available, for example, from Toyal America, Inc. of Naperville, Illinois, U.S.A. and Valimet Inc., of Stockton, California, U.S.A. Silver filler is

commercially available from Metalor Technologies U.S.A. Corp. of Attleboro, Massachusetts, U.S.A.

[0064] Thermally conductive fillers are known in the art and commercially available. For example, CB-A20S and Al-43-Me are aluminum oxide fillers of differing particle sizes commercially available from Showa-Denko, and AA-04, AA-2, and AA18 are aluminum oxide fillers commercially available from Sumitomo Chemical Company. Zinc oxides, such as zinc oxides having trademarks KADOX® and XX®, are commercially available from Zinc Corporation of America of Monaca, Pennsylvania, U.S.A.

[0065] The shape of the filler particles is not specifically restricted, however, rounded or spherical particles may prevent viscosity increase to an undesirable level upon high loading of the filler in the composition.

[0066] Ingredient (F) may be a single filler or a combination of two or more fillers that differ in at least one property such as particle shape, average particle size, particle size distribution, and type of filler. For example, it may be desirable to use a combination of fillers, such as a first filler having a larger average particle size and a second filler having a smaller average particle size. Use of a first filler having a larger average particle size and a second filler having a smaller average particle size than the first filler may improve packing efficiency and/or may reduce viscosity of the composition as compared to a composition without such a combination of fillers.

[0067] The average particle size of the filler will depend on various factors including the type of the filler selected for ingredient (F) and the exact amount added to the composition, as well as the end use for the reaction product of the composition. However, the filler may have an average particle size ranging from 0.1 micrometer to 80 micrometers, alternatively 0.1 micrometer to 50 micrometers, and alternatively 0.1 micrometer to 10 micrometers.

[0068] The amount of ingredient (F) in the composition depends on various factors including the end use selected for the composition and the reaction product of the composition, the type and amount of ingredient (B), and the type and amount of the filler selected for ingredient (F). However, the amount of ingredient (F) may range from 0 vol % to 80 vol %, alternatively 50 vol % to 75 vol %, and alternatively 30 % to 80 %, by volume of the composition. Without wishing to be bound by theory, it is thought that when the amount of

filler is greater than 80 vol %, the composition may react to form a reaction product with insufficient dimensional integrity for some applications.

Ingredient (G) Filler Treating Agent

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[0069] The composition may optionally further comprise ingredient (G) a treating agent. The amount of ingredient (G) will vary depending on factors such as the type of treating agent selected and the type and amount of particulates (such as ingredients (F) and/or (D)) to be treated, and whether the particulates are treated before being added to the composition, or whether the particulates are treated *in situ*. However, ingredient (G) may be used in an amount ranging from 0.01 % to 20 %, alternatively 0.1 % to 15 %, and alternatively 0.5 % to 5 %, based on the weight of all ingredients in the composition. Particulates, such as the filler, the physical drying agent, certain flame retardants, and/or certain pigments, when present, may optionally be surface treated with ingredient (G).

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Particulates may be treated with ingredient (G) before being added to the composition, or *in situ*. Ingredient (G) 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 fatty acids include stearates such as calcium stearate.

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[0070] Some representative organosilicon filler treating agents that can be used as ingredient (G) include compositions normally used to treat silica fillers such as organochlorosilanes, organosiloxanes, organodisilazanes such as hexaalkyl disilazane, and organoalkoxysilanes such as $C_6H_{13}Si(OCH_3)_3$, $C_8H_{17}Si(OC_2H_5)_3$, $C_{10}H_{21}Si(OCH_3)_3$, $C_{12}H_{25}Si(OCH_3)_3$, $C_{14}H_{29}Si(OC_2H_5)_3$, and $C_6H_5CH_2CH_2Si(OCH_3)_3$. Other treating agents that can be used include alkylthiols, fatty acids, titanates, titanate coupling agents, zirconate coupling agents, and combinations thereof.

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[0071] Alternatively, ingredient (G) may comprise an alkoxysilane having the formula:

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$R^{11}_mSi(OR^{12})_{(4-m)}$, where subscript m may have a value ranging from 1 to 3, alternatively subscript m is 3. Each R^{11} is independently a monovalent organic group, such as a monovalent hydrocarbon group of 1 to 50 carbon atoms, alternatively 8 to 30 carbon atoms, alternatively 8 to 18 carbon atoms. R^{11} is exemplified by alkyl groups such as hexyl, octyl, dodecyl, tetradecyl, hexadecyl, and octadecyl; and aromatic groups such as benzyl and

phenylethyl. R^{11} may be saturated or unsaturated, and branched or unbranched. Alternatively, R^{11} may be saturated and unbranched.

[0072] Each R^{12} is independently a saturated hydrocarbon group of 1 to 4 carbon atoms, alternatively 1 to 2 carbon atoms. Alkoxysilanes suitable for use as ingredient (G) are exemplified by hexyltrimethoxysilane, octyltriethoxysilane, decyltrimethoxysilane, dodecyltrimethoxysilane, tetradecyltrimethoxysilane, phenylethyltrimethoxysilane, octadecyltrimethoxysilane, octadecyltriethoxysilane, and combinations thereof.

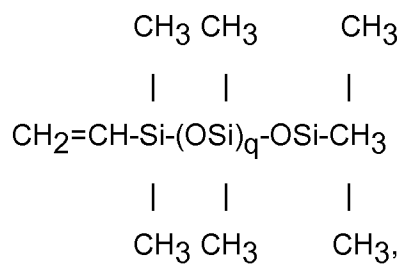
[0073] Alkoxy-functional oligosiloxanes may also be used as treating agents. For example, suitable alkoxy-functional oligosiloxanes include those of the formula (V):

$(R^{13}O)_nSi(OSiR^{14}_2R^{15})_{(4-n)}$. In this formula, subscript n is 1, 2 or 3, alternatively subscript n is 3. Each R^{13} may be an alkyl group. Each R^{14} may be an unsaturated monovalent hydrocarbon group of 1 to 10 carbon atoms. Each R^{15} may be an unsaturated monovalent hydrocarbon group having at least 10 carbon atoms.

[0074] Certain particulates, such as metal fillers may be treated with alkylthiols such as octadecyl mercaptan; fatty acids such as oleic acid and stearic acid; and a combination thereof.

[0075] Treatment agents for alumina or passivated aluminum nitride may include alkoxysilyl functional alkylmethyl polysiloxanes (e.g., partial hydrolysis condensate of $R^{16}_oR^{17}_pSi(OR^{18})_{(4-o-p)}$ or cohydrolysis condensates or mixtures), or similar materials where the hydrolyzable group may comprise silazane, acyloxy or oximo. In all of these, a group tethered to Si, such as R^{16} in the formula above, is a long chain unsaturated monovalent hydrocarbon or monovalent aromatic-functional hydrocarbon. Each R^{17} is independently a monovalent hydrocarbon group, and each R^{18} is independently a monovalent hydrocarbon group of 1 to 4 carbon atoms. In the formula above, subscript o is 1, 2, or 3 and subscript p is 0, 1, or 2, with the proviso that a sum ($o + p$) is 1, 2, or 3.

[0076] Other treating agents include alkenyl functional polyorganosiloxanes. Suitable alkenyl functional polyorganosiloxanes include, but are not limited to:



where subscript q has a value up to 1,500. Other treating agents include mono-endcapped alkoxy functional polydiorganosiloxanes, *i.e.*, polydiorganosiloxanes having an alkoxy group at one end. Such treating agents are exemplified by the formula:

10 $\text{R}^{25}\text{R}^{26}_2\text{SiO}(\text{R}^{26}_2\text{SiO})_u\text{Si}(\text{OR}^{27})_3$, where subscript u has a value of 0 to 100, alternatively 1 to 50, alternatively 1 to 10, and alternatively 3 to 6. Each R^{25} is independently selected from an alkyl group, such as methyl, ethyl, propyl, butyl, hexyl, and octyl; and an alkenyl group, such as vinyl, allyl, butenyl, and hexenyl. Each R^{26} is independently an alkyl group such as methyl, ethyl, propyl, butyl, hexyl, and octyl. Each R^{27} is independently an alkyl group such as methyl, ethyl, propyl, and butyl. Alternatively, each R^{25} , each R^{26} , and each R^{27} is a methyl group. Alternatively, each R^{25} is a vinyl group. Alternatively, each R^{26} and each R^{27} is a methyl group.

[0077] Alternative, a polyorganosiloxane capable of hydrogen bonding is useful as a treating agent. This strategy to treating surface of a filler takes advantage of multiple hydrogen bonds, either clustered or dispersed or both, as the means to tether the compatibilization moiety to the filler surface. The polyorganosiloxane capable of hydrogen bonding has an average, per molecule, of at least one silicon-bonded group capable of hydrogen bonding. The group may be selected from: an organic group having multiple hydroxyl functionalities or an organic group having at least one amino functional group. The polyorganosiloxane capable of hydrogen bonding means that hydrogen bonding is the primary mode of attachment for the polyorganosiloxane to a filler. The polyorganosiloxane may be incapable of forming covalent bonds with the filler. The polyorganosiloxane capable of hydrogen bonding may be selected from the group consisting of a saccharide-siloxane polymer, an amino-functional polyorganosiloxane, and a combination thereof. Alternatively, the polyorganosiloxane capable of hydrogen bonding may be a saccharide-siloxane polymer.

Ingredient (H) Biocide

[0078] Ingredient (H) is a biocide. The amount of ingredient (H) will vary depending on factors including the type of biocide selected and the benefit desired. However, the amount of ingredient (H) may range from greater than 0 % to 5 % based on the weight of all ingredients in the composition. Ingredient (H) is exemplified by (h1) a fungicide, (h2) an herbicide, (h3) a pesticide, or a combination thereof.

[0079] Ingredient (h1) is a fungicide, for example, these include N-substituted benzimidazole carbamate, benzimidazolyl carbamate such as methyl 2-benzimidazolylcarbamate, ethyl 2-benzimidazolylcarbamate, isopropyl 2-benzimidazolylcarbamate, methyl N-{2-[1-(N,N-dimethylcarbamoyl)benzimidazolyl]}carbamate, methyl N-{2-[1-(N,N-dimethylcarbamoyl)-6-methylbenzimidazolyl]}carbamate, methyl N-{2-[1-(N,N-dimethylcarbamoyl)-5-methylbenzimidazolyl]}carbamate, methyl N-{2-[1-(N-methylcarbamoyl)benzimidazolyl]}carbamate, methyl N-{2-[1-(N-methylcarbamoyl)-6-methylbenzimidazolyl]}carbamate, methyl N-{2-[1-(N-methylcarbamoyl)-5-methylbenzimidazolyl]}carbamate, ethyl N-{2-[1-(N,N-dimethylcarbamoyl)benzimidazolyl]}carbamate, ethyl N-{2-[2-(N-methylcarbamoyl)benzimidazolyl]}carbamate, ethyl N-{2-[1-(N,N-dimethylcarbamoyl)-6-methylbenzimidazolyl]}carbamate, ethyl N-{2-[1-(N-methylcarbamoyl)-6-methylbenzimidazolyl]}carbamate, isopropyl N-{2-[1-(N,N-dimethylcarbamoyl)benzimidazolyl]}carbamate, isopropyl N-{2-[1-(N-methylcarbamoyl)benzimidazolyl]}carbamate, methyl N-{2-[1-(N-propylcarbamoyl)benzimidazolyl]}carbamate, methyl N-{2-[1-(N-butylcarbamoyl)benzimidazolyl]}carbamate, methoxyethyl N-{2-[1-(N-propylcarbamoyl)benzimidazolyl]}carbamate, methoxyethyl N-{2-[1-(N-butylcarbamoyl)benzimidazolyl]}carbamate, ethoxyethyl N-{2-[1-(N-propylcarbamoyl)benzimidazolyl]}carbamate, ethoxyethyl N-{2-[1-(N-butylcarbamoyl)benzimidazolyl]}carbamate, methyl N-{1-(N,N-dimethylcarbamoyloxy)benzimidazolyl]}carbamate, methyl N-{2-[N-methylcarbamoyloxy)benzimidazolyl]}carbamate, methyl N-{2-[1-(N-butylcarbamoyloxy)benzimidazolyl]}carbamate, ethoxyethyl N-{2-[1-(N-propylcarbamoyl)benzimidazolyl]}carbamate, ethoxyethyl N-{2-[1-(N-butylcarbamoyloxy)benzimidazolyl]}carbamate, methyl N-{2-[1-(N,N-dimethylcarbamoyl)-6-chlorobenzimidazolyl]}carbamate, and methyl N-{2-[1-(N,N-dimethylcarbamoyl)-6-

nitrobenzimidazolyl}}carbamate; 10, 10'-oxybisphenoxarsine (which has trade name Vinyzene, OBPA), di-iodomethyl-para-tolylsulfone, benzothiophene-2-cyclohexylcarboxamide-S,S-dioxide, N-(fluordichloridemethylthio)phthalimide (which has trade names Fluor-Folper, and Preventol A3); methyl-benzimideazol-2-ylcarbamate (which has trade names Carbendazim, and Preventol BCM), zinc-bis(2-pyridylthio-1-oxide) (zinc pyrrithion) 2-(4-thiazolyl)-benzimidazol, N-phenyl-iodpropargylcarbamate, N-octyl-4-isothiazolin-3-on, 4,5-dichloride-2-n-octyl-4-isothiazolin-3-on, N-butyl-1,2-benzisothiazolin-3-on and/or triazolyl-compounds, such as tebuconazol in combination with zeolites containing silver.

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[0080] Ingredient (h2) is an herbicide, for example, suitable herbicides include amide herbicides such as allidochlor *N,N*-diallyl-2-chloroacetamide; CDEA 2-chloro-*N,N*-diethylacetamide; etniproimid (*RS*)-2-[5-(2,4-dichlorophenoxy)-2-nitrophenoxy]-*N*-ethylpropionamide; anilide herbicides such as cisanilide *cis*-2,5-dimethylpyrrolidine-1-carboxanilide; flufenacet 4'-fluoro-*N*-isopropyl-2-[5-(trifluoromethyl)-1,3,4-thiadiazol-2-yloxy]acetanilide; naproanilide (*RS*)- α -2-naphthoxypropionanilide; arylalanine herbicides such as benzoylprop *N*-benzoyl-*N*-(3,4-dichlorophenyl)-DL-alanine; flamprop-M *N*-benzoyl-*N*-(3-chloro-4-fluorophenyl)-D-alanine; chloroacetanilide herbicides such as butachlor *N*-butoxymethyl-2-chloro-2',6'-diethylacetanilide; metazachlor 2-chloro-*N*-(pyrazol-1-ylmethyl)acet-2',6'-xylidide; prynachlor (*RS*)-2-chloro-*N*-(1-methylprop-2-ynyl)acetanilide; sulphonanilide herbicides such as cloransulam 3-chloro-2-(5-ethoxy-7-fluoro[1,2,4]triazolo[1,5-*c*]pyrimidin-2-ylsulphonamido)benzoic acid; metosulam 2',6'-dichloro-5,7-dimethoxy-3'-methyl[1,2,4]triazolo[1,5-*a*]pyrimidine-2-sulphonanilide; antibiotic herbicides such as bilanafos 4-[hydroxy(methyl)phosphinoyl]-L-homoalanyl-L-alanyl-L-alanine; benzoic acid herbicides such as chloramben 3-amino-2,5-dichlorobenzoic acid; 2,3,6-TBA 2,3,6-trichlorobenzoic acid; pyrimidinyloxybenzoic acid herbicides such as bispyribac 2,6-bis(4,6-dimethoxypyrimidin-2-yloxy)benzoic acid; pyrimidinylthiobenzoic acid herbicides such as pyrrithiobac 2-chloro-6-(4,6-dimethoxypyrimidin-2-ylthio)benzoic acid; phthalic acid herbicides such as chlorthal tetrachloroterephthalic acid; picolinic acid herbicides such as aminopyralid 4-amino-3,6-dichloropyridine-2-carboxylic acid; quinolinecarboxylic acid herbicides such as quinclorac 3,7-dichloroquinoline-8-carboxylic acid; arsenical herbicides such as CMA calcium bis(hydrogen methylarsonate); MAMA ammonium hydrogen methylarsonate; sodium arsenite; benzoylcyclohexanedione herbicides such as mesotrione 2-(4-mesyl-2-nitrobenzoyl)cyclohexane-1,3-dione; benzofuranyl alkylsulphonate herbicides such as benfuresate 2,3-dihydro-3,3-dimethylbenzofuran-5-yl

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ethanesulphonate; carbamate herbicides such as carboxazole methyl 5-*tert*-butyl-1,2-oxazol-3-ylcarbamate; fenasulam methyl 4-[2-(4-chloro-*o*-tolylloxy)acetamido]phenylsulphonylcarbamate; carbanilate herbicides such as BCPC (*RS*)-*sec*-butyl 3-chlorocarbanilate; desmedipham ethyl 3-phenylcarbamoxyloxyphenylcarbamate;

5 swep methyl 3,4-dichlorocarbanilate; cyclohexene oxime herbicides such as butroxydim (*RS*)-(E)-5-(3-butyryl-2,4,6-trimethylphenyl)-2-(1-ethoxyiminopropyl)-3-hydroxycyclohex-2-en-1-one; tepraloxym (*RS*)-(E)-2-{1-[(2E)-3-chloroallyloxyimino]propyl}-3-hydroxy-5-perhydropyran-4-ylcyclohex-2-en-1-one; cyclopropylisoxazole herbicides such as isoxachlortole 4-chloro-2-mesylphenyl 5-cyclopropyl-1,2-oxazol-4-yl ketone; dicarboximide

10 herbicides such as flumezin 2-methyl-4-(α,α,α -trifluoro-*m*-tolyl)-1,2,4-oxadiazinane-3,5-dione; dinitroaniline herbicides such as ethalfluralin *N*-ethyl- α,α,α -trifluoro-*N*-(2-methylallyl)-2,6-dinitro-*p*-toluidine; prodiamine 5-dipropylamino- α,α,α -trifluoro-4,6-dinitro-*o*-toluidine; dinitrophenol herbicides such as dinoprop 4,6-dinitro-*o*-cymen-3-ol; etinofen α -ethoxy-4,6-dinitro-*o*-cresol; diphenyl ether herbicides such as ethoxyfen O-[2-chloro-5-(2-chloro- α,α,α -trifluoro-*p*-tolylloxy)benzoyl]-L-lactic acid; nitrophenyl ether herbicides such as aclofen 2-

15 chloro-6-nitro-3-phenoxyaniline; nitrofen 2,4-dichlorophenyl 4-nitrophenyl ether; dithiocarbamate herbicides such as dazomet 3,5-dimethyl-1,3,5-thiadiazinane-2-thione; halogenated aliphatic herbicides such as dalapon 2,2-dichloropropionic acid; chloroacetic acid; imidazolinone herbicides such as imazapyr (*RS*)-2-(4-isopropyl-4-methyl-5-oxo-2-

20 imidazolin-2-yl)nicotinic acid; inorganic herbicides such as disodium tetraborate decahydrate; sodium azide; nitrile herbicides such as chloroxynil 3,5-dichloro-4-hydroxybenzonitrile; ioxynil 4-hydroxy-3,5-di-iodobenzonitrile; organophosphorus herbicides such as anilofos S-4-chloro-*N*-isopropylcarbaniloylmethyl O,O-dimethyl phosphorodithioate; glufosinate 4-[hydroxy(methyl)phosphinoyl]-DL-homoalanine; phenoxy herbicides such as

25 clomeprop (*RS*)-2-(2,4-dichloro-*m*-tolylloxy)propionanilide; fenteracol 2-(2,4,5-trichlorophenoxy)ethanol; phenoxyacetic herbicides such as MCPA (4-chloro-2-methylphenoxy)acetic acid; phenoxybutyric herbicides such as MCPB 4-(4-chloro-*o*-tolylloxy)butyric acid; phenoxypropionic herbicides such as fenoprop (*RS*)-2-(2,4,5-trichlorophenoxy)propionic acid; aryloxyphenoxypropionic herbicides such as isoxapyrifop

30 (*RS*)-2-[2-[4-(3,5-dichloro-2-pyridyloxy)phenoxy]propionyl]isoxazolidine; phenylenediamine herbicides such as dinitramine *N*¹,*N*¹-diethyl-2,6-dinitro-4-trifluoromethyl-*m*-phenylenediamine, pyrazolyloxyacetophenone herbicides such as pyrazoxyfen 2-[4-(2,4-dichlorobenzoyl)-1,3-dimethylpyrazol-5-yloxy]acetophenone; pyrazolylphenyl herbicides such as pyraflufen 2-chloro-5-(4-chloro-5-difluoromethoxy-1-methylpyrazol-3-yl)-4-

35 fluorophenoxyacetic acid; pyridazine herbicides such as pyridafol 6-chloro-3-

phenylpyridazin-4-ol; pyridazinone herbicides such as chloridazon 5-amino-4-chloro-2-phenylpyridazin-3(2*H*)-one; oxapyrazon 5-bromo-1,6-dihydro-6-oxo-1-phenylpyridazin-4-yloxamic acid; pyridine herbicides such as fluroxypyr 4-amino-3,5-dichloro-6-fluoro-2-pyridyloxyacetic acid; thiazopyr methyl 2-difluoromethyl-5-(4,5-dihydro-1,3-thiazol-2-yl)-4-isobutyl-6-trifluoromethylnicotinate; pyrimidinediamine herbicides such as iprymidam 6-chloro-*N*⁴-isopropylpyrimidine-2,4-diamine; quaternary ammonium herbicides such as diethamquat 1,1'-bis(diethylcarbamoylmethyl)-4,4'-bipyridinium; paraquat 1,1'-dimethyl-4,4'-bipyridinium; thiocarbamate herbicides such as cycloate *S*-ethyl cyclohexyl(ethyl)thiocarbamate; tiocarbazil *S*-benzyl di-*sec*-butylthiocarbamate;

10 thiocarbonate herbicides such as EXD *O,O*-diethyl dithiobis(thioformate); thiourea herbicides such as methiuron 1,1-dimethyl-3-*m*-tolyl-2-thiourea; triazine herbicides such as triaziflam (*RS*)-*N*-[2-(3,5-dimethylphenoxy)-1-methylethyl]-6-(1-fluoro-1-methylethyl)-1,3,5-triazine-2,4-diamine; chlorotriazine herbicides such as cyprazine 6-chloro-*N*²-cyclopropyl-*N*⁴-isopropyl-1,3,5-triazine-2,4-diamine; propazine 6-chloro-*N*²,*N*⁴-di-isopropyl-1,3,5-triazine-2,4-diamine;

15 methoxytriazine herbicides such as prometon *N*²,*N*⁴-di-isopropyl-6-methoxy-1,3,5-triazine-2,4-diamine; methylthiotriazine herbicides such as cyanatryn 2-(4-ethylamino-6-methylthio-1,3,5-triazin-2-ylamino)-2-methylpropionitrile; triazinone herbicides such as hexazinone 3-cyclohexyl-6-dimethylamino-1-methyl-1,3,5-triazine-2,4(1*H*,3*H*)-dione; triazole herbicides such as epronaz *N*-ethyl-*N*-propyl-3-propylsulphonyl-1*H*-1,2,4-triazole-1-carboxamide;

20 triazolone herbicides such as carfentrazone (*RS*)-2-chloro-3-{2-chloro-5-[4-(difluoromethyl)-4,5-dihydro-3-methyl-5-oxo-1*H*-1,2,4-triazol-1-yl]-4-fluorophenyl}propionic acid; triazolopyrimidine herbicides such as florasulam 2',6',8-trifluoro-5-methoxy[1,2,4]triazolo[1,5-*c*]pyrimidine-2-sulphonanilide; uracil herbicides such as flupropacil isopropyl 2-chloro-5-(1,2,3,6-tetrahydro-3-methyl-2,6-dioxo-4-trifluoromethylpyrimidin-1-yl)benzoate; urea

25 herbicides such as cycluron 3-cyclo-octyl-1,1-dimethylurea; monisouron 1-(5-*tert*-butyl-1,2-oxazol-3-yl)-3-methylurea; phenylurea herbicides such as chloroxuron 3-[4-(4-chlorophenoxy)phenyl]-1,1-dimethylurea; siduron 1-(2-methylcyclohexyl)-3-phenylurea; pyrimidinylsulphonylurea herbicides such as flazasulphuron 1-(4,6-dimethoxypyrimidin-2-yl)-3-(3-trifluoromethyl-2-pyridylsulphonyl)urea; pyrazosulphuron 5-[(4,6-dimethoxypyrimidin-2-ylcarbamoyl)sulphamoyl]-1-methylpyrazole-4-carboxylic acid; triazinylsulphonylurea

30 herbicides such as thifensulphuron 3-(4-methoxy-6-methyl-1,3,5-triazin-2-ylcarbamoylsulphamoyl)thiophene-2-carboxylic acid; thiadiazolylurea herbicides such as tebuthiuron 1-(5-*tert*-butyl-1,3,4-thiadiazol-2-yl)-1,3-dimethylurea; and/or unclassified herbicides such as chlorfenac (2,3,6-trichlorophenyl)acetic acid; methazole 2-(3,4-

35 dichlorophenyl)-4-methyl-1,2,4-oxadiazolidine-3,5-dione; tritac (*RS*)-1-(2,3,6-

trichlorobenzyloxy)propan-2-ol; 2,4-D, chlorimuron, and fenoxaprop; and combinations thereof.

[0081] Ingredient (h3) is a pesticide. Suitable pesticides are exemplified by atrazine, diazinon, and chlorpyrifos. For purposes of this application, pesticide includes insect repellents such as N,N-diethyl-meta-toluamide and pyrethroids such as pyrethrin.

[0082] Ingredient (h4) is an antimicrobial agent. Suitable antimicrobials are commercially available, such as DOW CORNING® 5700 and DOW CORNING® 5772, which are from Dow Corning Corporation of Midland, Michigan, U.S.A.

[0083] Alternatively, ingredient (H) may comprise a boron containing material, e.g., boric anhydride, borax, or disodium octaborate tetrahydrate; which may function as a pesticide, fungicide, and/or flame retardant.

Ingredient (I) Stabilizer

[0084] Ingredient (I) is a stabilizer. Stabilizers for hydrosilylation curable compositions are exemplified by acetylenic alcohols such as methyl butynol, ethynyl cyclohexanol, dimethyl hexynol, and 3,5-dimethyl-1-hexyn-3-ol, 1-butyn-3-ol, 1-propyn-3-ol, 2-methyl-3-butyn-2-ol, 3-methyl-1-butyn-3-ol, 3-methyl-1-pentyn-3-ol, 3-phenyl-1-butyn-3-ol, 4-ethyl-1-octyn-3-ol, 3,5-dimethyl-1-hexyn-3-ol, and 1-ethynyl-1-cyclohexanol, 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.

[0085] Alternatively, ingredient (I) in the composition may be a silylated acetylenic compound. Without wishing to be bound by theory, it is thought that adding a silylated acetylenic compound reduces yellowing of the reaction product prepared from hydrosilylation reaction of the composition as compared to a reaction product from hydrosilylation of a

composition that does not contain a silylated acetylenic compound or that contains an organic acetylenic alcohol stabilizer, such as those described above.

[0086] The silylated acetylenic compound is exemplified by (3-methyl-1-butyn-3-oxy)trimethylsilane, ((1,1-dimethyl-2-propynyl)oxy)trimethylsilane, bis(3-methyl-1-butyn-3-oxy)dimethylsilane, bis(3-methyl-1-butyn-3-oxy)silanemethylvinylsilane, bis((1,1-dimethyl-2-propynyl)oxy)dimethylsilane, methyl(tris(1,1-dimethyl-2-propynyloxy))silane, methyl(tris(3-methyl-1-butyn-3-oxy))silane, (3-methyl-1-butyn-3-oxy)dimethylphenylsilane, (3-methyl-1-butyn-3-oxy)dimethylhexenylsilane, (3-methyl-1-butyn-3-oxy)triethylsilane, bis(3-methyl-1-butyn-3-oxy)methyltrifluoropropylsilane, (3,5-dimethyl-1-hexyn-3-oxy)trimethylsilane, (3-phenyl-1-butyn-3-oxy)diphenylmethylsilane, (3-phenyl-1-butyn-3-oxy)dimethylphenylsilane, (3-phenyl-1-butyn-3-oxy)dimethylvinylsilane, (3-phenyl-1-butyn-3-oxy)dimethylhexenylsilane, (cyclohexyl-1-ethyn-1-oxy)dimethylhexenylsilane, (cyclohexyl-1-ethyn-1-oxy)dimethylvinylsilane, (cyclohexyl-1-ethyn-1-oxy)diphenylmethylsilane, (cyclohexyl-1-ethyn-1-oxy)trimethylsilane, and combinations thereof. Alternatively, ingredient (I) is exemplified by methyl(tris(1,1-dimethyl-2-propynyloxy))silane, ((1,1-dimethyl-2-propynyl)oxy)trimethylsilane, or a combination thereof. The silylated acetylenic compound useful as ingredient (I) may be prepared by methods known in the art, such as silylating an acetylenic alcohol described above by reacting it with a chlorosilane in the presence of an acid receptor.

[0087] The amount of stabilizer added to the composition will depend on various factors including the desired pot life of the composition, whether the composition will be a one part composition or a multiple part composition, the particular stabilizer used, and the selection and amount of ingredient (C), if present. However, when present, the amount of stabilizer may range from 0 % to 1 %, alternatively 0.001 % to 1 %, alternatively 0.01 % to 0.5 %, and alternatively 0.0025 % to 0.025 %, based on the weight of all ingredients in the composition.

Ingredient (J) Flame Retardant

[0088] Ingredient (J) is a flame retardant. Suitable flame retardants may include, for example, carbon black, hydrated aluminum hydroxide, and silicates such as wollastonite, platinum and platinum compounds. Alternatively, the flame retardant may be selected from halogen based flame-retardants such as decabromodiphenyloxide, octabromodiphenyl oxide, hexabromocyclododecane, decabromobiphenyl oxide, diphenyloxybenzene, ethylene

bis- tetrabromophthalamide, pentabromoethyl benzene, pentabromobenzyl acrylate, tribromophenyl maleic imide, tetrabromobisphenyl A, bis-(tribromophenoxy) ethane, bis-(pentabromophenoxy) ethane, polydibromophenylene oxide, tribromophenylallyl ether, bis-dibromopropyl ether, tetrabromophthalic anhydride, dibromoneopentyl glycol, dibromoethyl
 5 dibromocyclohexane, pentabromodiphenyl oxide, tribromostyrene, pentabromochlorocyclohexane, tetrabromoxylene, hexabromocyclododecane, brominated polystyrene, tetradecabromodiphenoxybenzene, trifluoropropene and PVC. Alternatively, the flame retardant may be selected from phosphorus based flame-retardants such as (2,3-dibromopropyl)-phosphate, phosphorus, cyclic phosphates, triaryl phosphate, bis-
 10 melaminium pentate, pentaerythritol bicyclic phosphate, dimethyl methyl phosphate, phosphine oxide diol, triphenyl phosphate, tris- (2-chloroethyl) phosphate, phosphate esters such as tricreyl, trixylenyl, isodecyl diphenyl, ethylhexyl diphenyl, phosphate salts of various amines such as ammonium phosphate, trioctyl, tributyl or tris-butoxyethyl phosphate ester. Other flame retardants may include tetraalkyl lead compounds such as tetraethyl lead, iron
 15 pentacarbonyl, manganese methyl cyclopentadienyl tricarbonyl, melamine and derivatives such as melamine salts, guanidine, dicyandiamide, ammonium sulphamate, alumina trihydrate, and magnesium hydroxide alumina trihydrate.

[0089] The amount of flame retardant will vary depending on factors such as the flame
 20 retardant selected and whether solvent is present. However, the amount of flame retardant in the composition may range from greater than 0 % to 10 % based on the weight of all ingredients in the composition.

Ingredient (K) Surface Modifier

[0090] Ingredient (K) is a surface modifier. Suitable surface modifiers are exemplified by
 (k1) an adhesion promoter and (k2) a release agent. Suitable adhesion promoters for
 ingredient (k1) may comprise a transition metal chelate, a hydrocarbonoxysilane such as an
 alkoxysilane, a combination of an alkoxysilane and a hydroxy-functional polyorganosiloxane,
 30 an aminofunctional silane, or a combination thereof. Adhesion promoters are known in the art and may comprise silanes having the formula $R^{19}_r R^{20}_s Si(OR^{21})_{4-(r+s)}$ where each R^{19} is independently a monovalent organic group having at least 3 carbon atoms; R^{20} contains at least one SiC bonded substituent having an adhesion-promoting group, such as amino, epoxy, mercapto or acrylate groups; subscript r has a value ranging from 0 to 2; subscript s
 35 is either 1 or 2; and the sum of (r + s) is not greater than 3. Alternatively, the adhesion

promoter may comprise a partial condensate of the above silane. Alternatively, the adhesion promoter may comprise a combination of an alkoxy silane and a hydroxy-functional polyorganosiloxane.

- 5 **[0091]** Alternatively, the adhesion promoter may comprise an unsaturated or epoxy-functional compound. The adhesion promoter may comprise an unsaturated or epoxy-functional alkoxy silane. For example, the functional alkoxy silane can have the formula $R^{22}_tSi(OR^{23})_{(4-t)}$, where subscript t is 1, 2, or 3, alternatively subscript t is 1. Each R^{22} is independently a monovalent organic group with the proviso that at least one R^{22} is an
- 10 unsaturated organic group or an epoxy-functional organic group. Epoxy-functional organic groups for R^{22} are exemplified by 3-glycidoxypropyl and (epoxycyclohexyl)ethyl. Unsaturated organic groups for R^{22} are exemplified by 3-methacryloyloxypropyl, 3-acryloyloxypropyl, and unsaturated monovalent hydrocarbon groups such as vinyl, allyl, hexenyl, undecenyl. Each R^{23} is independently a saturated hydrocarbon group of 1 to 4
- 15 carbon atoms, alternatively 1 to 2 carbon atoms. R^{23} is exemplified by methyl, ethyl, propyl, and butyl.

- [0092]** Examples of suitable epoxy-functional alkoxy silanes include 3-glycidoxypropyltrimethoxysilane, 3-glycidoxypropyltriethoxysilane,
- 20 (epoxycyclohexyl)ethyldimethoxysilane, (epoxycyclohexyl)ethyldiethoxysilane and combinations thereof. Examples of suitable unsaturated alkoxy silanes include vinyltrimethoxysilane, allyltrimethoxysilane, allyltriethoxysilane, hexenyltrimethoxysilane, undecenyltrimethoxysilane, 3-methacryloyloxypropyl trimethoxysilane, 3-methacryloyloxypropyl triethoxysilane, 3-acryloyloxypropyl trimethoxysilane, 3-
- 25 acryloyloxypropyl triethoxysilane, and combinations thereof.

- [0093]** Alternatively, the adhesion promoter may comprise an epoxy-functional siloxane such as a reaction product of a hydroxy-terminated polyorganosiloxane with an epoxy-functional alkoxy silane, as described above, or a physical blend of the hydroxy-terminated
- 30 polyorganosiloxane with the epoxy-functional alkoxy silane. The adhesion promoter may comprise a combination of an epoxy-functional alkoxy silane and an epoxy-functional siloxane. For example, the adhesion promoter is exemplified by a mixture of 3-glycidoxypropyltrimethoxysilane and a reaction product of hydroxy-terminated methylvinylsiloxane with 3-glycidoxypropyltrimethoxysilane, or a mixture of 3-

glycidoxypyrroltrimethoxysilane and a hydroxy-terminated methylvinylsiloxane, or a mixture of 3-glycidoxypyrroltrimethoxysilane and a hydroxy-terminated methylvinyl/dimethylsiloxane copolymer.

- 5 **[0094]** Alternatively, the adhesion promoter may comprise an aminofunctional silane, such as an aminofunctional alkoxysilane exemplified by $\text{H}_2\text{N}(\text{CH}_2)_2\text{Si}(\text{OCH}_3)_3$, $\text{H}_2\text{N}(\text{CH}_2)_2\text{Si}(\text{OCH}_2\text{CH}_3)_3$, $\text{H}_2\text{N}(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3$, $\text{H}_2\text{N}(\text{CH}_2)_3\text{Si}(\text{OCH}_2\text{CH}_3)_3$, $\text{CH}_3\text{NH}(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3$, $\text{CH}_3\text{NH}(\text{CH}_2)_3\text{Si}(\text{OCH}_2\text{CH}_3)_3$, $\text{CH}_3\text{NH}(\text{CH}_2)_5\text{Si}(\text{OCH}_3)_3$, $\text{CH}_3\text{NH}(\text{CH}_2)_5\text{Si}(\text{OCH}_2\text{CH}_3)_3$, $\text{H}_2\text{N}(\text{CH}_2)_2\text{NH}(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3$,
 10 $\text{H}_2\text{N}(\text{CH}_2)_2\text{NH}(\text{CH}_2)_3\text{Si}(\text{OCH}_2\text{CH}_3)_3$, $\text{CH}_3\text{NH}(\text{CH}_2)_2\text{NH}(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3$, $\text{CH}_3\text{NH}(\text{CH}_2)_2\text{NH}(\text{CH}_2)_3\text{Si}(\text{OCH}_2\text{CH}_3)_3$, $\text{C}_4\text{H}_9\text{NH}(\text{CH}_2)_2\text{NH}(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3$, $\text{C}_4\text{H}_9\text{NH}(\text{CH}_2)_2\text{NH}(\text{CH}_2)_3\text{Si}(\text{OCH}_2\text{CH}_3)_3$, $\text{H}_2\text{N}(\text{CH}_2)_2\text{SiCH}_3(\text{OCH}_3)_2$, $\text{H}_2\text{N}(\text{CH}_2)_2\text{SiCH}_3(\text{OCH}_2\text{CH}_3)_2$, $\text{H}_2\text{N}(\text{CH}_2)_3\text{SiCH}_3(\text{OCH}_3)_2$, $\text{H}_2\text{N}(\text{CH}_2)_3\text{SiCH}_3(\text{OCH}_2\text{CH}_3)_2$, $\text{CH}_3\text{NH}(\text{CH}_2)_3\text{SiCH}_3(\text{OCH}_3)_2$, $\text{CH}_3\text{NH}(\text{CH}_2)_3\text{SiCH}_3(\text{OCH}_2\text{CH}_3)_2$, $\text{CH}_3\text{NH}(\text{CH}_2)_5\text{SiCH}_3(\text{OCH}_3)_2$,
 15 $\text{CH}_3\text{NH}(\text{CH}_2)_5\text{SiCH}_3(\text{OCH}_2\text{CH}_3)_2$, $\text{H}_2\text{N}(\text{CH}_2)_2\text{NH}(\text{CH}_2)_3\text{SiCH}_3(\text{OCH}_3)_2$, $\text{H}_2\text{N}(\text{CH}_2)_2\text{NH}(\text{CH}_2)_3\text{SiCH}_3(\text{OCH}_2\text{CH}_3)_2$, $\text{CH}_3\text{NH}(\text{CH}_2)_2\text{NH}(\text{CH}_2)_3\text{SiCH}_3(\text{OCH}_3)_2$, $\text{CH}_3\text{NH}(\text{CH}_2)_2\text{NH}(\text{CH}_2)_3\text{SiCH}_3(\text{OCH}_2\text{CH}_3)_2$, $\text{C}_4\text{H}_9\text{NH}(\text{CH}_2)_2\text{NH}(\text{CH}_2)_3\text{SiCH}_3(\text{OCH}_3)_2$, $\text{C}_4\text{H}_9\text{NH}(\text{CH}_2)_2\text{NH}(\text{CH}_2)_3\text{SiCH}_3(\text{OCH}_2\text{CH}_3)_2$, and a combination thereof.
- 20 **[0095]** Alternatively, the adhesion promoter may comprise a transition metal chelate. Suitable transition metal chelates include titanates, zirconates such as zirconium acetylacetonate, aluminum chelates such as aluminum acetylacetonate, and combinations thereof. Alternatively, the adhesion promoter may comprise a combination of a transition metal chelate with an alkoxysilane, such as a combination of
 25 glycidoxypyrroltrimethoxysilane with an aluminum chelate or a zirconium chelate.

[0096] Ingredient (k2) is a release agent. Suitable release agents are exemplified by fluorinated compounds, such as fluoro-functional silicones, or fluoro-functional organic compounds.

30

[0097] Alternatively, the surface modifier for ingredient (K) may be used to change the appearance of the surface of a reaction product of the composition. For example, surface modifier may be used to increase gloss of the surface of a reaction product of the composition. Such a surface modifier may comprise a polydiorganosiloxane with alkyl and
 35 aryl groups. For example, DOW CORNING® 550 Fluid is a trimethylsiloxy-terminated

poly(dimethyl/methylphenyl)siloxane with a viscosity of 125 cSt that is commercially available from Dow Corning Corporation of Midland, Michigan, U.S.A.

[0098] Alternatively, ingredient (K) may be a natural oil obtained from a plant or animal source, such as linseed oil, tung oil, soybean oil, castor oil, fish oil, hempseed oil, cottonseed oil, oiticica oil, or rapeseed oil.

[0099] The exact amount of ingredient (K) depends on various factors including the type of surface modifier selected as ingredient (K) and the end use of the composition and its reaction product. However, ingredient (K), when present, may be added to the composition in an amount ranging from 0.01 to 50 weight parts based on the weight of the composition, alternatively 0.01 to 10 weight parts, and alternatively 0.01 to 5 weight parts. Ingredient (K) may be one adhesion promoter. Alternatively, ingredient (K) may comprise two or more different surface modifiers that differ in at least one of the following properties: structure, viscosity, average molecular weight, polymer units, and sequence.

Ingredient (L) Chain Lengthener

[0100] Chain lengtheners may include difunctional silanes and difunctional siloxanes, which extend the length of polyorganosiloxane chains before crosslinking occurs. Chain lengtheners may be used to reduce the modulus of elongation of the cured product. Chain lengtheners compete in their reactions with aliphatically unsaturated groups and/or silicon bonded hydrogen atoms in other ingredients of the composition, e.g., ingredients (B) and/or ingredient (C), when present. Dimethylhydrogensiloxy-terminated polydimethylsiloxanes having relatively low degrees of polymerization (e.g., DP ranging from 3 to 50) may be used as ingredient (L). Ingredient (L) may be one chain lengthener. Alternatively, ingredient (L) may comprise two or more different chain lengtheners that differ in at least one of the following properties: structure, viscosity, average molecular weight, polymer units, and sequence

Ingredient (M) Endblocker

[0101] Ingredient (M) is an endblocker comprising an M unit, i.e., a siloxane unit of formula $R^{24}_3SiO_{1/2}$, where each R^{24} independently represents a monovalent, non-functional, organic group, such as a monovalent hydrocarbon group free of aliphatic unsaturation.

Ingredient (M) may comprise polyorganosiloxanes endblocked on one terminal end by a triorganosilyl group, e.g., $(\text{CH}_3)_3\text{SiO}-$, and on the other end by a silicon bonded hydrogen atom and/or an aliphatically unsaturated organic group. Ingredient (M) may be a polydiorganosiloxane such as a polydimethylsiloxane. The polydiorganosiloxanes having both silicon bonded hydrogen terminals and triorganosilyl end groups, may have more than 50 %, alternatively more than 75 %, of the total end groups as silicon bonded hydrogen atoms. The amount of triorganosilyl group in the polydimethylsiloxane may be used to regulate the modulus of a cured product prepared by curing the composition. Without wishing to be bound by theory, it is thought that higher concentrations of triorganosilyl end groups may provide a lower modulus in cured products. Ingredient (M) may be one endblocker. Alternatively, ingredient (M) may comprise two or more different endblockers that differ in at least one of the following properties: structure, viscosity, average molecular weight, polymer units, and sequence.

Ingredient (N) Flux Agent

[0102] Ingredient (N) is a flux agent. The composition may comprise 0 % to 2 % of the flux agent based on the weight of all ingredients in the composition. Molecules containing chemically active functional groups such as carboxylic acid and amines can be used as flux agents. Such flux agents can include aliphatic acids such as succinic acid, abietic acid, oleic acid, and adipic acid; aromatic acids such as benzoic acids; aliphatic amines and their derivatives, such as triethanolamine, hydrochloride salts of amines, and hydrobromide salts of amines. Flux agents are known in the art and are commercially available.

Ingredient (O) Anti-Aging Additive

[0103] Ingredient (O) is an anti-aging additive. The anti-aging additive may comprise an antioxidant, a UV absorber, a UV stabilizer, a heat stabilizer, or a combination thereof. Suitable antioxidants are known in the art and are commercially available. Suitable antioxidants include phenolic antioxidants and combinations of phenolic antioxidants with stabilizers. Phenolic antioxidants include fully sterically hindered phenols and partially hindered phenols; and sterically hindered amines such as tetramethyl-piperidine derivatives. Suitable phenolic antioxidants include vitamin E and IRGANOX® 1010 from Ciba Specialty Chemicals, U.S.A. IRGANOX® 1010 comprises pentaerythritol tetrakis(3-(3,5-di-t-butyl-4-hydroxyphenyl)propionate). Examples of UV absorbers include phenol, 2-(2H-benzotriazol-

2-yl)-6-dodecyl-4-methyl-, branched and linear (TINUVIN® 571). Examples of UV stabilizers include bis(1,2,2,6,6-pentamethyl-4-piperidyl) sebacate; methyl 1,2,2,6,6-pentamethyl-4-piperidyl/sebacate; and a combination thereof (TINUVIN® 272). These and other TINUVIN® additives, such as TINUVIN® 765 are commercially available from Ciba Specialty Chemicals of Tarrytown, NY, U.S.A. Other UV and light stabilizers are commercially available, and are exemplified by LowLite from Chemtura, OnCap from PolyOne, and Light Stabilizer 210 from E. I. du Pont de Nemours and Company of Delaware, U.S.A. Oligomeric (higher molecular weight) stabilizers may alternatively be used, for example, to minimize potential for migration of the stabilizer out of the composition or the cured product thereof. An example of an oligomeric antioxidant stabilizer (specifically, hindered amine light stabilizer (HALS)) is Ciba TINUVIN® 622, which is a dimethylester of butanedioic acid copolymerized with 4-hydroxy-2,2,6,6-tetramethyl-1-piperidine ethanol. Heat stabilizers may include iron oxides and carbon blacks, iron carboxylate salts, cerium hydrate, barium zirconate, cerium and zirconium octoates, and porphyrins.

[0104] The amount of ingredient (O) depends on various factors including the specific anti-aging additive selected and the anti-aging benefit desired. However, the amount of ingredient (O) may range from 0 to 5 weight %, alternatively 0.1 % to 4 %, and alternatively 0.5 to 3 weight %, based on the weight of all ingredients in the composition. Ingredient (O) may be one anti-aging additive. Alternatively, ingredient (O) may comprise two or more different anti-aging additives.

Ingredient (P) Pigment

[0105] Ingredient (P) is a pigment. For purposes of this application, the term 'pigment' includes any ingredient used to impart color to a reaction product of a composition described herein. The amount of pigment depends on various factors including the type of pigment selected and the desired degree of coloration of the product. For example, the composition may comprise 0 to 20 %, alternatively 0.001 % to 5 %, of a pigment based on the weight of all ingredients in the composition.

[0106] Examples of suitable pigments include indigo, titanium dioxide Stan-Tone 50SP01 Green (which is commercially available from PolyOne) and carbon black. Representative, non-limiting examples of carbon black include Shawinigan Acetylene black, which is commercially available from Chevron Phillips Chemical Company LP; SUPERJET® Carbon

Black (LB-1011) supplied by Elementis Pigments Inc., of Fairview Heights, IL U.S.A.; SR 511 supplied by Sid Richardson Carbon Co, of Akron, OH U.S.A.; and N330, N550, N762, N990 (from Degussa Engineered Carbons of Parsippany, NJ, U.S.A.).

5 **Ingredient (Q) Acid Acceptor**

[0107] Ingredient (Q) is an acid acceptor. Suitable acid acceptors include magnesium oxide, calcium oxide, and combinations thereof. The composition may comprise 0 % to 2 % of ingredient (Q) based on the weight of the composition.

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Ingredient (R) Rheological Additive

[0108] The composition may optionally further comprise up to 5 %, alternatively 1 % to 2 % based on the weight of the composition of ingredient (R) a rheological additive for modifying
15 rheology of the composition. Rheological additives are known in the art and are commercially available. Examples include polyamides, Polyvest, which is commercially available from Evonk, Disparlon from King Industries, Kevlar Fibre Pulp from Du Pont, Rheospan from Nanocor, and Ircogel from Lubrizol. Other suitable rheological additives include polyamide waxes; hydrogenated castor oil derivatives; and metal soaps such as
20 calcium stearate, aluminum stearate and barium stearate, and combinations thereof.

[0109] Alternatively, ingredient (R) may comprise a microcrystalline wax that is a solid at 25 °C (wax). The melting point may be selected such that the wax has a melting point at the low end of the desired application temperature range. Without wishing to be bound by
25 theory, it is thought that ingredient (R) acts as a process aid that improves flow properties of the composition. Without wishing to be bound by theory, it is thought that incorporation of wax may also facilitate incorporation of fillers, compounding and de-airing (during production of the composition), and mixing (static or dynamic mixing during application of parts of a multiple part composition). It is thought that the wax, when molten, serves as a process aid,
30 substantially easing the incorporation of filler in the composition during compounding, the compounding process itself, as well as in during a de-airing step, if used. The wax, with a melt temperature below 100 °C, may facilitate mixing of the parts of a multiple part composition before application, even in a simple static mixer.

[0110] Waxes suitable for use as ingredient (R) may be non-polar hydrocarbons. The waxes may have branched structures, cyclic structures, or combinations thereof. For example, petroleum microcrystalline waxes are available from Strahl & Pitsch, Inc., of West Babylon, NY, U.S.A. and include SP 96 (melting point ranging from 62 °C to 69 °C), SP 18 (melting point ranging from 73 °C to 80 °C), SP 19 (melting point ranging from 76 °C to 83 °C), SP 26 (melting point ranging from 76 °C to 83 °C), SP 60 (melting point ranging from 79 °C to 85 °C), SP 617 (melting point ranging from 88 °C to 93 °C), SP 89 (melting point ranging from 90 °C to 95 °C), and SP 624 (melting point ranging from 90 °C to 95 °C). Other petroleum microcrystalline waxes include waxes marketed under the trademark Multiwax® by Crompton Corporation of Petrolia, Pennsylvania, U.S.A. These waxes include 180-W, which comprises saturated branched and cyclic non-polar hydrocarbons and has melting point ranging from 79 °C to 87 °C; Multiwax® W-445, which comprises saturated branched and cyclic non-polar hydrocarbons, and has melting point ranging from 76 °C to 83 °C; and Multiwax® W-835, which comprises saturated branched and cyclic non-polar hydrocarbons, and has melting point ranging from 73 °C to 80 °C.

[0111] The amount of ingredient (R) depends on various factors including the specific rheological additive selected and the selections of the other ingredients of the composition. However, the amount of ingredient (R) may range from 0 parts to 20 parts, alternatively 1 parts to 15 parts, and alternatively 1 part to 5 parts based on the weight of all ingredients in the composition. Ingredient (R) may be one rheological additive. Alternatively, ingredient (R) may comprise two or more different rheological additives.

Ingredient (S) Solvent

[0112] Solvent may be used in the composition. Solvent may facilitate flow of the composition and introduction of certain ingredients, such as silicone resin. Solvents used herein are those that help fluidize the ingredients of the composition but essentially do not react with the ingredients. Solvent may be selected based on solubility the ingredients in the composition and volatility. The solubility refers to the solvent being sufficient to dissolve and/or disperse ingredients of the composition. Volatility refers to vapor pressure of the solvent. If the solvent is too volatile (having too high vapor pressure) bubbles may form in the composition during hydrosilylation reaction, and the bubbles may cause cracks or otherwise weaken or detrimentally affect properties of the reaction product. However, if the

solvent is not volatile enough (too low vapor pressure) the solvent may remain as a plasticizer in the reaction product of the composition.

[0113] Suitable solvents include polyorganosiloxanes with suitable vapor pressures, such as hexamethyldisiloxane, octamethyltrisiloxane, hexamethylcyclotrisiloxane and other low molecular weight polyorganosiloxanes, such as 0.5 to 1.5 cSt Dow Corning® 200 Fluids and Dow Corning® OS FLUIDS, which are commercially available from Dow Corning Corporation of Midland, Michigan, U.S.A.

[0114] Alternatively, the solvent may be an organic solvent. The organic solvent can be an alcohol such as methanol, ethanol, isopropanol, butanol, or n-propanol; a ketone such as acetone, methylethyl ketone, or methyl isobutyl ketone; an aromatic hydrocarbon such as benzene, toluene, or xylene; an aliphatic hydrocarbon such as heptane, hexane, or octane; a glycol ether such as propylene glycol methyl ether, dipropylene glycol methyl ether, propylene glycol n-butyl ether, propylene glycol n-propyl ether, or ethylene glycol n-butyl ether, a halogenated hydrocarbon such as dichloromethane, 1,1,1-trichloroethane or methylene chloride; chloroform; dimethyl sulfoxide; dimethyl formamide, acetonitrile; tetrahydrofuran; white spirits; mineral spirits; naphtha; n-methyl pyrrolidone; or a combination thereof.

[0115] The amount of solvent will depend on various factors including the type of solvent selected and the amount and type of other ingredients selected for the composition. However, the amount of solvent may range from 1 % to 99%, alternatively 2 % to 50 %, based on the weight of all ingredients in the composition. Ingredient (S) can be added during preparation of the composition, for example, to aid mixing and delivery. All or a portion of ingredient (S) may optionally be removed after the composition is prepared.

(T) Surfactant

[0116] Ingredient (T) is a surfactant. Suitable surfactants include silicone polyethers, ethylene oxide polymers, propylene oxide polymers, copolymers of ethylene oxide and propylene oxide, other non-ionic surfactants, and combinations thereof. The composition may comprise 0 % to 0.05 % of the surfactant based on the weight of all ingredients in the composition.

[0117] When selecting ingredients for the composition described above, there may be overlap between types of ingredients because certain ingredients described herein may have more than one function. For example, certain alkoxysilanes may be useful as filler treating agents and as adhesion promoters, and certain plasticizers such as fatty acid esters may also be useful as filler treating agents. Certain particulates may be useful as fillers and as pigments, and even as flame retardants, *e.g.*, carbon black. When adding additional ingredients to the composition, the additional ingredients are distinct from one another.

Method of Preparation of the Composition

[0118] The composition can be prepared by a method comprising combining all ingredients by any convenient means such as mixing at ambient or elevated temperature. When the composition is prepared at elevated temperature, the temperature during preparation is less than the hydrosilylation reaction temperature of the composition. Ingredient (I), when present, may be added before ingredient (A), for example, when the composition will be prepared at elevated temperature and/or the composition will be prepared as a one part composition.

[0119] When ingredient (G) is present, the composition may optionally be prepared by surface treating a particulate ingredient (*e.g.*, filler and/or spacer, if present) with ingredient (G), and thereafter mixing the product thereof with the other ingredients of the composition.

[0120] Alternatively, the composition may be prepared as a multiple part composition, for example, when ingredient (I) is absent, or when the composition will be stored for a long period of time before use. In the multiple part composition, ingredient (A) is stored in a separate part from any ingredient having a silicon bonded hydrogen atom, for example ingredient (C), and the parts are combined shortly before use of the composition. For example, a two part composition may be prepared by combining ingredients comprising (B), (A), (F), and optionally one or more other additional ingredients described above to form a base by any convenient means such as mixing. A curing agent may be prepared by combining ingredients comprising (B), (C), and optionally one or more other additional ingredients described above by any convenient means such as mixing. The ingredients may be combined at ambient or elevated temperature. When a two part composition is used, the weight ratio of amounts of base to curing agent may range from 1:1 to 10:1. The

composition will react via hydrosilylation reaction to form a reaction product. The reaction product may have various forms, such as a silane, a gum, a gel, a rubber, or a resin.

EXAMPLES

[0121] These examples are intended to illustrate the invention and should not be interpreted as limiting the scope of the invention set forth in the claims. All syntheses and manipulations were carried out under argon using standard Schlenk and vacuum techniques. ^1H , ^{13}C , and ^{29}Si NMR spectra were recorded on a Varian Gemini 300 VT and Varian Mercury 300 VT spectrometers in C_6D_6 . The mass spectra of the products were determined by GC-MS (Varian Saturn 2100T equipped with a DB-1, 30m capillary column). GC analyses were carried out on a Varian 3400 CX series gas chromatograph with a capillary column DB-1, 30m and TC detector. The chemicals were obtained from the following sources: benzene- d_6 , 1,3-dichlorotetramethyldisiloxane from ABCR, phenylacetylene, *n*-butyl lithium from Sigma Aldrich Co., heptamethyltrisiloxane and vinylheptamethyltrisiloxane from Gelest, THF, hexane, toluene, benzene from POCH Gliwice (Poland). $\text{Pt}(\text{cod})_2$ was prepared according to the well-known procedures. All solvents and liquid reagents were dried and distilled under argon prior to use.

Example 1 Synthesis of platinum(0) complexes

Synthesis of Pt complex with 1,3-diethynyl-1,1,3,3-tetramethyldisiloxane ligand (I)

To a Schlenk's tube containing 0.5g (1.22 mmol) $[\text{Pt}(\text{cod})_2]$, at room temperature, in argon atmosphere, 5 mL of benzene and 2.22 g (12.2 mmol) of 1,3-diethynyltetramethyldisiloxane were added. The reaction was conducted for 24 hours at room temperature on stirring the reaction mixture with a magnetic stirrer. After this time the dark-brown solution was filtered off by cannula system and the solvent was evaporated under reduced pressure. The obtained precipitate was dried under vacuum for 16h. The complex was obtained with a yield of 96 %.

^1H NMR (300 MHz, C_6D_6 , 300 K) $\delta(\text{ppm})$ = 1.58, 1.40 (s, 2H, $\text{HC}\equiv$); 0.29 (s, 12H, $-\text{CH}_3$)

^{13}C NMR (75.42 MHz, C_6D_6 , 300 K) $\delta(\text{ppm})$ = 30.47, 30.22 ($\text{HC}\equiv\text{C}-$); 2.18 (bs), 1.40 (bs) ($-\text{CH}_3$)

Example 2: Synthesis of Pt complex with 1,3-(diphenylethynyl)-1,1,3,3-tetramethyldisiloxane ligand (II)

To a Schlenk's tube containing 0.5g (1.22 mmol) [Pt(cod)₂], at room temperature, in argon atmosphere, 5 mL of benzene and 2.04 g (6.1 mmol) of 1,3-(diphenylethynyl)-1,1,3,3-tetramethyldisiloxane were added. The reaction was conducted for 24 hours at room temperature on stirring the reaction mixture with a magnetic stirrer. After this time dark-brown solution was filtered off by cannula system and the solvent was evaporated under reduced pressure. The obtained precipitate was dried under vacuum for 16h. The complex was obtained with a yield of 92 %.

¹H NMR (300 MHz, C₆D₆, 300 K) δ(ppm) = 7.55 (d,); 7.38 (m,); 7.28 (m,); 0.51 (s, 12H, -CH₃)

¹³C NMR (75.42 MHz, C₆D₆, 300 K) δ(ppm) = 131.99, 129.01, 128.66, 128.21, 128.17, 93.27, 2.18

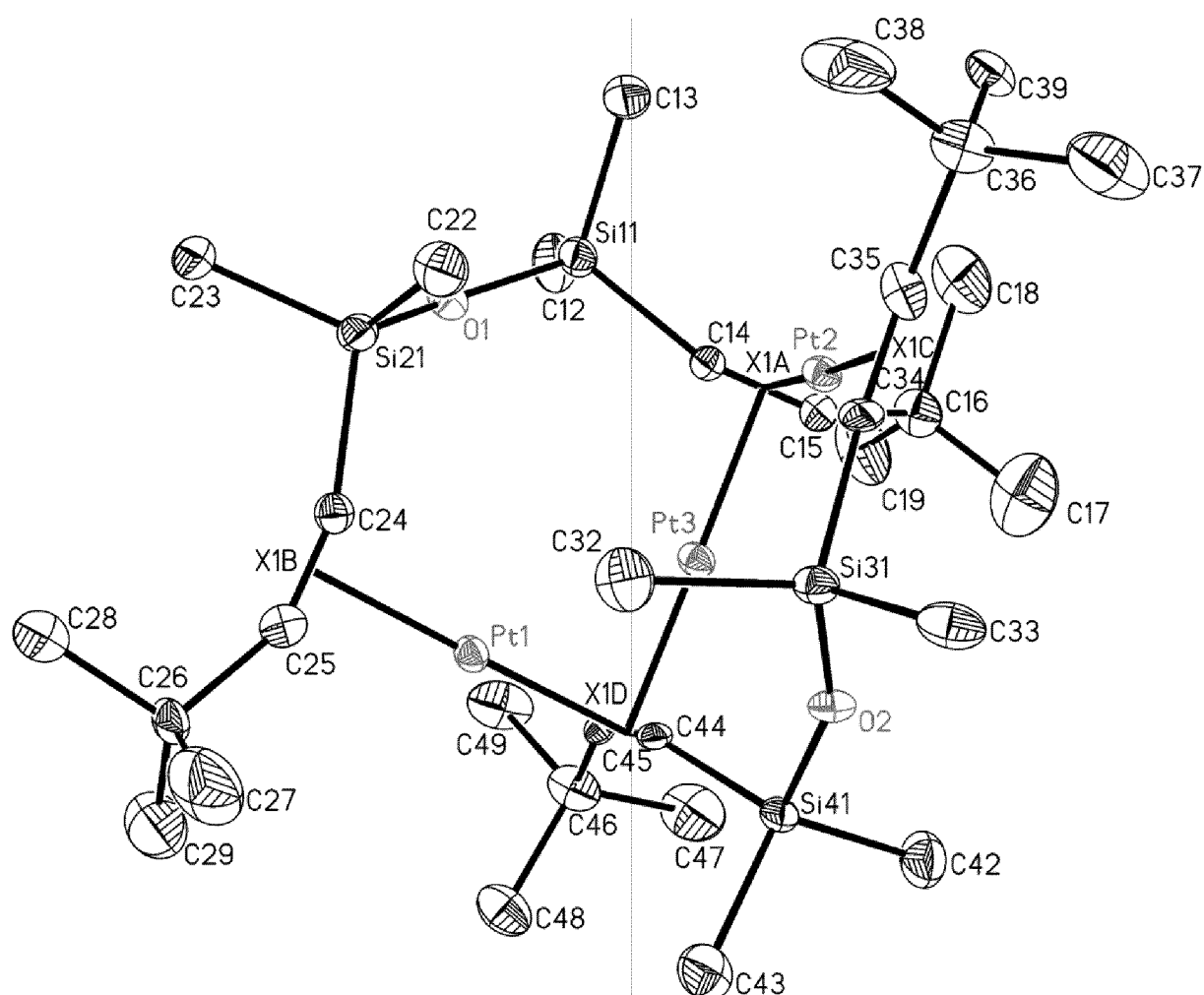
Example 3: Synthesis of Pt complex with 1,3-bis(tert-buthylethynyl)-1,1,3,3-tetramethyldisiloxane ligand (III)

To a Schlenk's tube containing 0.5g (1.22 mmol) [Pt(cod)₂], at room temperature, in argon atmosphere, 5 mL of benzene and 1.79g (6.1 mmol) of 1,3-bis(tert-buthylethynyl)-1,1,3,3-tetramethyldisiloxane were added. The reaction was conducted for 24 hours at room temperature on stirring the reaction mixture with a magnetic stirrer. After this time light-brown/yellow solution was filtered off by cannula system and the solvent was evaporated under reduced pressure. The obtained precipitate was dried under vacuum for 16h. The complex was obtained with a yield of 93 %.

¹H NMR (300 MHz, C₆D₆, 300 K) δ(ppm) = 1.46 (s, 9H, t-Bu); 1.30 (s, 9H, t-Bu); 0.49 (s, 3H, -CH₃); 0.47 (s, 3H, -CH₃); 0.44 (s, 3H, -CH₃); 0.40 (s, 3H, -CH₃)

¹³C NMR (75.42 MHz, C₆D₆, 300 K) δ(ppm) = 156.73, 137.10, 115.06, 110.72, 92.83, 82.53, 32.90, 31.84, 30.86, 30.22, 3.73, 3.37, 2.78, 2.37

The structure of the isolated Pt-ligand complex was analysed by X-ray crystallography:



A perspective view of the molecule with atom labeling. Ellipsoids are drawn at the 50% probability level, hydrogen atoms are shown as spheres of arbitrary radii. X1A, X1B, X1C and X1D are the middlepoints of the double bonds. Selected geometrical parameters: Pt1-X1B 1.932Å, Pt1-X1D 1.943Å, Pt2-X1C 1.936Å, Pt2-X1A 1.960Å, Pt3-X1A 1.998Å, Pt3-X1D 1.983Å; Pt1...Pt3 2.7455(8)Å, Pt2...Pt3 2.7470(97)Å, C14-C15 1.324(11)Å, C24-C25 1.260(11)Å, C34-C35 1.281(12)Å, C44-C45 1.344(12)Å, X1B-Pt1-X1D 176.5°, X1C-Pt2-X1A 178.0°, X1A-Pt3-X1D 178.4°.

10 **Example 4: Synthesis of Pt complex with 1,3-bis(trimethylsilyl)ethynyl)-1,1,3,3-tetramethyldisiloxane ligand (IV)**

To a Schlenk's tube containing 0.5g (1.22 mmol) [Pt(cod)₂], at room temperature, in argon atmosphere, 5 mL of benzene and 1.99g (6.1 mmol) of 1,3- *bis(trimethylsilyl)ethynyl)-1,1,3,3-tetramethyldisiloxane* were added. The reaction was conducted for 24 hours at room temperature on stirring the reaction mixture with a magnetic stirrer. After this time dark-

brown solution was filtered off by cannula system and the solvent was evaporated under reduced pressure. The obtained precipitate was dried under vacuum for 16h. The complex was obtained with a yield of 97%.

5 ^1H NMR (300 MHz, C_6D_6 , 300 K) $\delta(\text{ppm}) = 0.39$ (s, 12H, Me); 0.31 (s, 18H, Me).

^{13}C NMR (75.42 MHz, C_6D_6 , 300 K) $\delta(\text{ppm}) = 113.69$; 113.53 , 2.53 , 1.65 .

^{29}C NMR (119.26 MHz, C_6D_6 , 300 K) $\delta(\text{ppm}) = -17.62$; -18.44 .

Comparative

10

As comparative, Karstedt catalyst was prepared to provide tris(divinylsiloxane) diplatinum catalyst $\text{Pt}_2[(\text{ViSiMe}_2)_2\text{O}]_3$ (Karstedt).

II. Hydrosilylation reactions

15

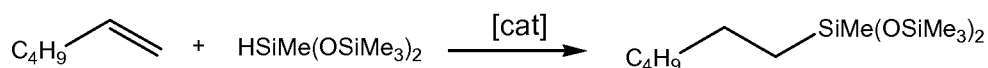
Compounds (**I - IV**) were used as catalysts in the hydrosilylation of 1-hexene (examples 5-16 and comparative), styrene (examples 29 to 32 and comparative), as well as small vinylsiloxane vinylmethylbis(trimethylsiloxy)silane (examples 17 to 28 and comparative) as a model for curing of silicone rubbers.

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Examples 5 to 16

The hydrosilylation of 1-hexene with heptamethyltrisiloxane occurred according to the following equation:

25



The reaction was conducted for 3 hours for examples 5 to 12 and 16 hours for examples 13 to 16. Because of the low boiling point of olefin, the reaction in an opened system was conducted in 50°C with doubled molar excess of olefin to the starting silane.

30

Molar ratio [Pt]:[\equiv SiH] = 10^{-6} : 1. 3 hours

Example	Compound	Yield [%]
5	I	66
6	II	59
7	III	77
8	IV	63
Comparative	Karstedt	64

Molar ratio [Pt]:[\equiv SiH] 10×10^{-7} . 3 hours

5

Example	Compound	Yield [%]
9	I	45
10	II	34
11	III	55
12	IV	43
Comparative	Karstedt	44

Molar ratio [Pt]:[\equiv SiH] 10×10^{-7} . 16 hours

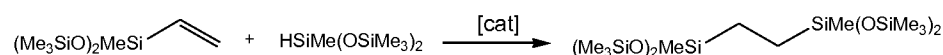
Example	Compound	Yield [%]
13	I	82
14	II	79
15	III	100
16	IV	80
Comparative	Karstedt	81

- 10 The data collected indicated that all Pt precursors were very active in this reaction, but complex III seems to be the best. Due to a lower temperature of the process the total consumption of silane was observed after 16-24 hours for low concentrated catalyst solutions.

Examples 17 to 28

The hydrosilylation of vinylmethylbis(trimethylsiloxy)silane with heptamethyltrisiloxane was conducted and occurs according to the following equation:

5



This reaction was performed at 120°C when the molar ratio of reagents was 1:1. The Molar ratio [Pt]:[≡SiH] was 10x-7.

10 Time of reaction 1h

Example	Compound	Yield [%]
17	I	46
18	II	58
19	III	62
20	IV	27
Comparative	Karstedt	36

Time of reaction 2h

Example	Compound	Yield [%]
21	I	66
22	II	70
23	III	79
24	IV	48
Comparative	Karstedt	64

15

Time of reaction 3h

Example	Compound	Yield [%]
25	I	77
26	II	87
27	III	87
28	IV	65
Comparative	Karstedt	77

The results on the catalytic activity of new Pt-compounds indicated that under identical conditions the complex (III) was a little bit more active than Karstedt. After 3 – 12h we observed total conversion of both substrates in reactions catalyzed by compound III, whereas by using Karstedt system 16 – 24h were required to complete the reaction. High catalytic activity of the new compounds showed in the reaction between simple siloxanes suggested their application for catalyzing hydrosilylation of big molecules like cross-linking of silicon rubbers.

Examples 29 to 32

The hydrosilylation of styrene with heptamethyltrisiloxane was conducted at 100°C for 24 hours. The Molar ratio of reagents was 1:1. The molar ratio [Pt]:[≡SiH] was 10^{-6} : 1

Example	Compound	Yield [%]
29	I	87
30	II	84
31	III	98
32	IV	82
Comparative	Karstedt	96

CLAIMS

1. A composition comprising:
 - (A) a complex platinum compound containing
 - i) a Pt atom and
 - ii) a ligand having at least one atom of Si, at least one atom of O, and at least one alkyne group; and
 - (B) an aliphatically unsaturated compound having an average, per molecule, of one or more aliphatically unsaturated organic groups capable of undergoing hydrosilylation reaction; with the proviso that when ingredient (B) does not contain a silicon bonded hydrogen atom, then the composition further comprises
 - (C) an SiH functional compound having an average, per molecule, of one or more silicon bonded hydrogen atoms; which is distinct from ingredients (A) and (B).
2. The composition of claim 1, where the ligand has the general formula:
$$[\text{O}(\text{SiMe}_2\text{C}\equiv\text{C-R})_2]$$
with R being selected from H (I), Ph (II), t-Bu (III) or SiMe₃ (IV).
3. The composition of claim 1, where the composition further comprises one or more additional ingredients, which are distinct from ingredients (A), (B), and (C), and which are selected from the group consisting of (D) a spacer; (E) an extender, a plasticizer, or a combination thereof; (F) a filler; (G) a filler treating agent; (H) a biocide; (I) an inhibitor, (J) a flame retardant; (K) a surface modifier; (L) a chain lengthener; (M) an endblocker; (N) a flux agent; (O) an anti-aging additive; (P) a pigment; (Q) an acid acceptor (R) a rheological additive; (S) a solvent; (T) a surfactant; and a combination thereof.
4. A method for making the composition of claim 1, comprising mixing ingredients comprising ingredient (A), ingredient (B), optionally ingredient (C), and optionally one or more additional ingredients, which are distinct from ingredients (A), (B), and (C), and which are selected from the group consisting of (D) a spacer; (E) an extender, a plasticizer, or a combination thereof; (F) a filler; (G) a filler treating agent; (H) a biocide; (I) an inhibitor, (J) a flame retardant; (K) a surface modifier; (L)

a chain lengthener; (M) an endblocker; (N) a flux agent; (O) an anti-aging additive; (P) a pigment; (Q) an acid acceptor (R) a rheological additive; (S) a solvent; (T) a surfactant; and a combination thereof.

5. A method comprising: reacting
 - i) a Pt precursor having general formula (i) Pt-A_a , where
 - Pt is a platinum atom;
 - each A is independently a monovalent organic group or complexing compound; and
 - subscript a has a value of 2; and
 - ii) a ligand selected from the group consisting of a compound containing at least one atom of Si, at least one atom of O and at least one alkyne group.
6. A platinum compound containing
 - i) a Pt atom and
 - ii) a ligand having at least one atom of Si, at least one atom of O, and at least one alkyne group.
7. The complex platinum compound according to claim 6, wherein the ligand has the formula $[\text{O}(\text{SiMe}_2\text{C}\equiv\text{C-R})_2]$ with R being selected from H (I), Ph (II), t-Bu (III) or SiMe₃ (IV).
8. The platinum compound according to claim 7, wherein the formula is $\text{Pt}_3[\text{O}(\text{SiMe}_2\text{C}\equiv\text{C-t-Bu})_2]_2$.
9. Use of a platinum compound as defined in claim 6 as hydrosilylation catalyst.

INTERNATIONAL SEARCH REPORT

International application No

PCT/EP2012/061826

A. CLASSIFICATION OF SUBJECT MATTER
INV. C07F17/02
ADD.

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)
C07F

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 practicable, search terms used)

EPO-Internal, WPI Data, 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	US 6 359 098 B1 (FEHN ARMIN [DE] ET AL) 19 March 2002 (2002-03-19) catalyst 6 column 13-14; column 2 lines 22-31, column 1 lines 5-10 -----	1-9
X	DE 199 43 102 A1 (WACKER CHEMIE GMBH [DE]) 15 March 2001 (2001-03-15) column 9 example 7 -----	6
X	OZAWA, FUMIYUKI ET AL: "Preparation and C-Si Reductive Elimination Behavior of cis-Alkynyl(silyl)platinum(II) Complexes", ORGANOMETALLICS, 22(17), 3593-3599 CODEN: ORGND7; ISSN: 0276-7333, vol. 22, 25 July 2003 (2003-07-25), pages 3593-3599, XP002684926, page 3595; table 2; compound 11 -----	6



Further documents are listed in the continuation of Box C.



See patent family annex.

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"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/EP2012/061826

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