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(54) Title: SILICONE-POLYOLEFIN HYBRID ELASTOMERS

(57) Abstract: A silicone-polyolefin composition is disclosed. The silicone-polyolefin composition comprises (A) a polysiloxane, (B) a functionalized polyolefin, (C) a polyolefin that is not reactable with components (A) or (B), and (D) a curable silicone comprising at least one of an elastomer component or a liquid rubber. The polysiloxane (A) comprises an average of at least one functional group X per molecule, and the functionalized polyolefin (B) comprises an average of at least one functional group Y per molecule, where the functional group X and functional group Y are reactable to form a bond therebetween. A silicone-polyolefin blend, a curable composition comprising the silicone-polyolefin blend, a cured product of the curable composition, and methods of preparing the silicone-polyolefin blend, curable composition, and cured product are also disclosed.

SILICONE-POLYOLEFIN HYBRID ELASTOMERS

CROSS REFERENCE TO RELATED APPLICATIONS

[0001] This application claims priority to and all advantages of U.S. Provisional Patent Application No. 63/249,755 filed on 29 September 2021, the content of which is incorporated herein by reference.

FIELD OF THE INVENTION

[0002] The present disclosure relates generally to silicone compositions and, more specifically, to a hybrid silicone-polyolefin composition, related curable compositions, and silicone-polyolefin hybrid elastomers formed therewith.

DESCRIPTION OF THE RELATED ART

[0003] Silicones are polymeric materials used in numerous commercial applications, primarily due to significant advantages they possess over their carbon-based analogues. More particularly referred to as polymerized siloxanes or polysiloxanes, silicones include an inorganic silicon-oxygen backbone chain (···–Si–O–Si–O–Si–O–···) having organic side groups attached to the silicon atoms. Organic side groups may be used to link two or more of these backbones together. By varying the –Si–O– chain lengths, side groups, and cross-linking, silicones can be synthesized with a wide variety of properties and compositions, with silicone networks varying in consistency from liquid to gel to rubber to hard plastic. Silicone and siloxane-based materials are utilized in myriad end use applications and environments, including as components in a wide variety of industrial, home care, and personal care formulations.

[0004] The most common silicone materials are based on the linear organopolysiloxane polydimethylsiloxane (PDMS), a silicone oil, followed by those based on silicone resins formed with branched and cage-like oligosiloxanes. Many of these materials enable unique technologies by providing enhanced performance and benefits due to inherent attributes of organopolysiloxane, including low-loss and stable optical transmission capabilities, high thermal and oxidative stabilities, and biocompatibility.

[0005] Unfortunately, despite widespread success in numerous technologies, the use of silicone materials in certain applications remains limited, if even practicable, due to less-desirable attributes of many conventional siloxanes, such as their weak mechanical properties, which may manifest in materials with poor or unsuitable characteristics such as low tensile strength, low tear strength, etc. As such, carbon-based polymers, such as those based on polyolefin, polyacrylate, and polyurethane resins, are frequently employed in applications that could otherwise benefit from particular inherent attributes of silicones. Complicating matters further, conventional siloxanes are incompatible with most carbon-based polymers, typically due to immiscibility and/or exhibiting antagonistic properties with respect to one another.

BRIEF SUMMARY OF THE INVENTION

[0006] A silicone-polyolefin composition (the "composition") is provided. The composition comprises (A) a polysiloxane comprising an average of at least one functional group X per molecule, (B) a functionalized polyolefin comprising an average of at least one functional group Y per molecule, (C) a polyolefin that is not reactable with components (A) or (B), and (D) a curable silicone comprising at least one of an elastomer component or a liquid rubber. The functional group X of the polysiloxane (A) and the functional Y of the functionalized polyolefin (B) are reactable to form a bond therebetween.

[0007] A method of preparing a silicone-polyolefin blend (the "preparation method") is also provided. The preparation method comprises combining the polysiloxane (A), the functionalized polyolefin (B), the polyolefin (C), and the curable silicone (D) to prepare the silicone-polyolefin composition, and reacting the polysiloxane (A) and the anhydride-functional polyolefin (B) in the presence of the polyolefin (C) and curable silicone (D), thereby preparing the silicone-polyolefin blend.

[0008] A silicone-polyolefin blend prepared according to the preparation method is also provided.

[0009] A curable composition is further provided. The curable composition comprises the silicone-polyolefin blend and a curing agent.

[0010] A cured product of the curable composition, and a method of preparing the cured product, are also provided.

DETAILED DESCRIPTION OF THE INVENTION

[0011] A silicone-polyolefin composition (the "composition") is provided herein. As will be understood from the description herein, the composition provides a hybrid composition that contains both silicone and polyolefin components, and may be prepared and used without the aid of solvent. As silicone and polyolefin components are generally understood to be immiscible or otherwise incompatible with one another, the particular compounds and conditions utilized provide unique hybrid materials that exhibit desirable characteristics and properties that may be unobtainable with conventional methods and materials. Likewise, the composition may be exploited as a platform for efficient and economical preparation of numerous functional compositions and components thereof, including in some applications that are poorly suited, if even practicable, for use with conventional materials. Indeed, a compatibilized silicone-polyolefin blend, a curable composition prepared therewith, and a cured product prepared therefrom, as well as methods of making each of the same, are also provided herein and illustrated in the examples below.

[0012] In view of this disclosure, one of skill in the art will readily appreciate that the unique structural and physical features of the inventive compositions are compatible with useful production techniques, such as melt-blending and reactive extrusion, without certain drawbacks inherent to using such techniques with traditional materials. Additionally, as also described and

illustrated herein, the inventive compositions enable the preparation of products with enhanced performance characteristics, including injection moldable articles with improved toughness (e.g. increased tear strength) and chemical resistance (e.g. increased solvent swell resistance), satisfactory elongation and tensile strength, and desirable haptics. Such articles may be specifically employed in consumer products, where specific performance characteristics provided by conventional materials are often mutually exclusive, and enhancing any one property may decrease the positive tactual experience of a user.

[0013] An "ethylene-based polymer" is a polymer that contains more than 50 weight percent (wt.%) polymerized ethylene monomer (based on the total amount of polymerizable monomers) and, optionally, may contain at least one comonomer. Ethylene-based polymer includes ethylene homopolymer, and ethylene copolymer (meaning units derived from ethylene and one or more comonomers). The terms "ethylene-based polymer" and "polyethylene" may be used interchangeably. Non-limiting examples of ethylene-based polymer (polyethylene) include low density polyethylene (LDPE) and linear polyethylene. Non-limiting examples of linear polyethylene include linear low density polyethylene (LLDPE), ultra low density polyethylene (ULDPE), very low density polyethylene (VLDPE), multi- component ethylene-based copolymer (EPE), ethylene/α-olefin multi-block copolymers (also known as olefin block copolymer (OBC)), substantially linear, or linear, plastomers/elastomers, and high density polyethylene (HDPE).

[0014] High density polyethylene (or "HDPE") is an ethylene homopolymer or an ethylene/α-olefin copolymer with at least one C4-C10 α-olefin comonomer, or C4-C8 α-olefin comonomer and a density of at least 0.940 g/cm^3, or at least 0.945 g/cm^3, or at least 0.950 g/cm^3, for example from 0.953 g/cm^{3}to 0.955 g/cm^3, or at least 0.960 g/cm^3, or at least 0.965 g/cm^3, or at least 0.970 g/cm^3, or at least 0.975 g/cm^3, or at least 0.980 g/cm^3. HDPE can be a monomodal copolymer or a multimodal copolymer. A "monomodal ethylene copolymer" is an ethylene/C4-C10 α-olefin copolymer that has one distinct peak in a gel permeation chromatography (GPC) showing the molecular weight distribution. A "multimodal ethylene copolymer" is an ethylene/C4-C10 α-olefin copolymer that has at least two distinct peaks in a GPC showing the molecular weight distribution. Multimodal includes copolymer having two peaks (bimodal) as well as copolymer having more than two peaks.

[0015] "Low density polyethylene" (or "LDPE") consists of ethylene homopolymer, or ethylene/ α-olefin copolymer comprising at least one C3-C10 α-olefin that has a density from 0.915 g/cm^3 to 0.940 g/cm^3 and contains long chain branching with broad MWD.

[0016] "Linear low density polyethylene" (or "LLDPE") is a linear ethylene/ α-olefin copolymer containing heterogeneous short-chain branching distribution comprising units derived from ethylene and units derived from at least one C3-C10 α-olefin comonomer. LLDPE is

characterized by little, if any, long chain branching, in contrast to conventional LDPE. LLDPE has a density from 0.910 g/cm^3 to less than 0.940 g/cm^3.

[0017] An "olefin-based polymer" or "polyolefin" is a polymer that contains a majority amount, or greater than 50 wt.%, of polymerized olefin monomer, for example, ethylene or propylene, (based on the weight of the polymer), and optionally, may contain at least one comonomer. A nonlimiting example of an olefin-based polymer is an ethylene-based polymer and propylene-based polymer.

[0018] A "propylene-based polymer" is a polymer that contains more than 50 wt.% polymerized propylene monomer (based on the total amount of polymerizable monomers) and, optionally, may contain at least one comonomer. The terms "propylene- based polymer" and "polypropylene" may be used interchangeably.

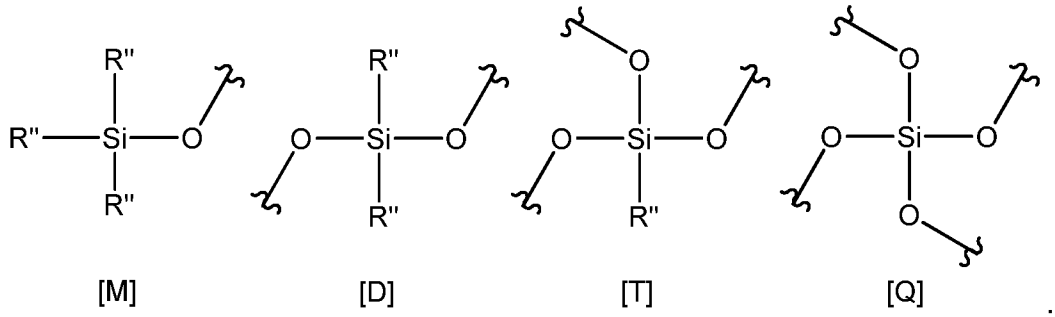
[0019] "Ultra low density polyethylene" (or "ULDPE") and "very low density polyethylene" (or "VLDPE") each is a linear ethylene/ α-olefin copolymer containing heterogeneous short-chain branching distribution comprising units derived from ethylene and units derived from at least one C3-C10 α-olefin comonomer. ULDPE and VLDPE each has a density from 0.885 g/cm^3 to 0.915 g/cm^3.

[0020] The silicone-polyolefin composition generally includes (A) a polysiloxane, (B) a functionalized polyolefin, (C) a polyolefin that is not reactable with components (A) or (B), and (D) a curable silicone, which comprises at least one of an elastomer component or a liquid rubber. The polysiloxane (A) comprises an average, per molecule, of at least one functional group X, and the functionalized polyolefin (B) comprises an average, per molecule, of at least one functional group Y. In some embodiments, the polysiloxane (A) comprises an average of at least two functional groups X per molecule. In these or other embodiments, the functionalized polyolefin (B) comprises an average of at least two functional groups Y per molecule. As will be understood in view of the description and examples herein, the functional group X of the polysiloxane (A) and the functional group Y of the functionalized polyolefin (B) are reactable (e.g. via additive functional reaction) to form a bond therebetween, i.e., to couple the polysiloxane (A) and the functionalized polyolefin (B).

[0021] The polysiloxane (A), functionalized polyolefin (B), polyolefin (C), and curable silicone (D) are described in turn below, along with additional compounds that may be present in the silicone-polyolefin composition, which may be collectively referred to herein as the “components” of the silicone-polyolefin composition (i.e., “component (A)”, “component (B)”, “component (C)”, “component (D)”, etc., respectively.) or, likewise, as “compound(s),” and/or “reagent(s)” (A), (B), (C), and/or (D), etc.

[0022] As introduced above, the silicone-polyolefin composition comprises the polysiloxane (A). As understood by those of skill in the art, polysiloxanes are silicon-based compounds comprising a siloxane backbone, i.e., an at least semi-contiguous chain composed of inorganic silicon-

oxygen-silicon groups (i.e., -Si-O-Si-), with organosilicon and/or organic side groups attached to the silicon atoms. Such siloxanes are typically characterized in terms of the number, type, and/or proportion of [M], [D], [T], and/or [Q] units/siloxy groups, which each represent structural units of individual functionality present in polysiloxanes, such as organosiloxanes and organopolysiloxanes. In particular, [M] represents a monofunctional unit of general formula $R''_3SiO_{1/2}$; [D] represents a difunctional unit of general formula $R''_2SiO_{2/2}$; [T] represents a trifunctional unit of general formula $R''SiO_{3/2}$; and [Q] represents a tetrafunctional unit of general formula $SiO_{4/2}$, as shown by the general structural moieties below:



[0023] In these general structural moieties, each R" is independently a monovalent or polyvalent substituent. As understood in the art, specific substituents suitable for each R" are not particularly limited (e.g. may be monoatomic or polyatomic, organic or inorganic, linear or branched, substituted or unsubstituted, aromatic, aliphatic, saturated or unsaturated, etc., as well as various combinations thereof). In typical examples, each R" is independently selected from hydrocarbyl groups, alkoxy and/or aryloxy groups, and siloxy groups. With regard to hydrocarbyl groups suitable for R", examples generally include monovalent hydrocarbon moieties, as well as derivatives and modifications thereof, which may independently be substituted or unsubstituted, linear, branched, cyclic, or combinations thereof, and saturated or unsaturated. With regard to such hydrocarbyl groups, the term “unsubstituted” describes hydrocarbon moieties composed of carbon and hydrogen atoms, i.e., without heteroatom substituents. The term “substituted” describes hydrocarbon moieties where either at least one hydrogen atom is replaced with an atom or group other than hydrogen (e.g. a halogen atom, an alkoxy group, an amine group, etc.) (i.e., as a pendant or terminal substituent), a carbon atom within a chain/backbone of the hydrocarbon is replaced with an atom other than carbon (e.g. a heteroatom, such as oxygen, sulfur, nitrogen, etc.) (i.e., as a part of the chain/backbone), or both. As such, suitable hydrocarbyl groups may comprise, or be, a hydrocarbon moiety having one or more substituents in and/or on (i.e., appended to and/or integral with) a carbon chain/backbone thereof, such that the hydrocarbon moiety may comprise, or otherwise be referred to as, an ether, an ester, etc. Linear and branched hydrocarbyl groups may independently be saturated or unsaturated and, when unsaturated, may be conjugated or nonconjugated. Cyclic hydrocarbyl groups may independently

be monocyclic or polycyclic, and encompass cycloalkyl groups, aryl groups, and heterocycles, which may be aromatic, saturated and nonaromatic and/or non-conjugated, etc. Examples of combinations of linear and cyclic hydrocarbyl groups include alkaryl groups, aralkyl groups, etc. General examples of hydrocarbon moieties suitably for use in or as the hydrocarbyl group include alkyl groups, aryl groups, alkenyl groups, alkynyl groups, halocarbon groups, and the like, as well as derivatives, modifications, and combinations thereof. Examples of alkyl groups include methyl, ethyl, propyl (e.g. iso-propyl and/or n-propyl), butyl (e.g. isobutyl, n-butyl, tert-butyl, and/or sec-butyl), pentyl (e.g. isopentyl, neopentyl, and/or tert-pentyl), hexyl, and the like (i.e., other linear or branched saturated hydrocarbon groups, e.g. having greater than 6 carbon atoms). Examples of aryl groups include phenyl, tolyl, xylyl, naphthyl, benzyl, dimethyl phenyl, and the like, as well as derivatives and modifications thereof, which may overlap with alkaryl groups (e.g. benzyl) and aralkyl groups (e.g. tolyl, dimethyl phenyl, etc.). Examples of alkenyl groups include vinyl, allyl, propenyl, isopropenyl, butenyl, isobutenyl, pentenyl, heptenyl, hexenyl, cyclohexenyl groups, and the like, as well as derivatives and modifications thereof. General examples of halocarbon groups include halogenated derivatives of the hydrocarbon moieties above, such as halogenated alkyl groups (e.g. any of the alkyl groups described above, where one or more hydrogen atoms is replaced with a halogen atom such as F or Cl), aryl groups (e.g. any of the aryl groups described above, where one or more hydrogen atoms is replaced with a halogen atom such as F or Cl), and combinations thereof. Examples of halogenated alkyl groups include 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, 2,2-difluorocyclopropyl, 2,3-difluorocyclobutyl, 3,4-difluorocyclohexyl, 3,4-difluoro-5-methylcycloheptyl, chloromethyl, chloropropyl, 2-dichlorocyclopropyl, 2,3-dichlorocyclopentyl, and the like, as well as derivatives and modifications thereof. Examples of halogenated aryl groups include chlorobenzyl, pentafluorophenyl, fluorobenzyl groups, and the like, as well as derivatives and modifications thereof. With regard to alkoxy and/or aryloxy groups suitable for R", examples generally include hydrocarbyl (e.g. alkyl, aryl, etc.) groups bonded to the silicon atom via an oxygen atom (i.e., forming a silyl ether). The hydrocarbyl groups in these examples may include any of the hydrocarbyl groups described above. With regard to siloxy groups suitable for R", examples generally include siloxy groups represented by any one, or combination, of [M], [D], [T], and/or [Q] units described above.

[0024] One of skill in the art understands how [M], [D], [T] and [Q] units, and their relative proportions (i.e., molar fractions) influence and control the structure of siloxanes, and that polysiloxanes in general may be monomeric, polymeric, oligomeric, linear, branched, cyclic, and/or resinous depending on the selection of [M], [D], [T] and/or [Q] units therein. For example, [T] units and/or [Q] units are typically present in siloxane resins, whereas siloxane polymers (e.g.

silicones) are typically free from such [T] units and/or [Q] units. [D] units are typically present in both siloxane resins and polymers. One of skill in the art will also appreciate that siloxanes may be named based on the type and proportion of such siloxy units. For example, the siloxane resins referenced above may be characterized as DT resins, MQ resins, MDQ resins, etc. Likewise, siloxanes that are substantially free from branching attributable to [T] units and/or [Q] units are typically referred to as "linear". However, it will be appreciated that a linear (i.e., MDM-type) siloxane may comprise individual molecules having T and/or Q units and still be considered "linear" based on the average unit formula of the siloxane as a whole.

[0025] In general, the polysiloxane (A) comprises a polydiorganosiloxane-containing backbone having at least one functional group X per molecule. Typically, the polysiloxane (A) is substantially linear, alternatively is linear. In such instances, one of skill in the art will appreciate that the polysiloxane (A) is typically free from [T] siloxy units and/or [Q] siloxy units, as described above.

[0026] In some embodiments, the polysiloxane (A) has the following general average unit formula:

$$[X_mR^1_{3-m}SiO_{1/2}]_a[X_nR^1_{2-n}SiO_{2/2}]_b,$$

where X is the functional group defined above, each R^1 is an independently selected hydrocarbyl group, subscript m is independently 1 or 0 in each moiety indicated by subscript a, subscript n is independently 1 or 0 in each moiety indicated by subscript b, and subscripts a and b are each mole fractions such that $a+b=1$, with the provisos that $0<a<1$, $0<b<1$, and that the polysiloxane (A) comprises at least one functional group X.

[0027] With reference to the general unit formula of the polysiloxane (A) above, hydrocarbyl groups suitable for R^1 are generally exemplified by those described above. Typically, each R^1 is a substituted or unsubstituted hydrocarbyl group having from 1 to 30 carbon atoms. For example, in some embodiments, each R^1 is an independently selected hydrocarbyl group having from 1 to 12, alternatively from 1 to 8, alternatively from 1 to 6, carbon atoms. In some such embodiments, each R^1 is further defined as an alkyl group, aryl group, or combination thereof. For example, in some embodiments, R^1 represents an independently selected substituted or unsubstituted alkyl group. Specific examples of such alkyl groups include methyl groups, ethyl groups, propyl groups (e.g. n-propyl and iso-propyl groups), butyl groups (e.g. n-butyl, sec-butyl, iso-butyl, and tert-butyl groups), pentyl groups, hexyl groups, etc., and the like, as well as derivatives and/or modifications thereof. Examples of derivatives and/or modifications of such alkyl groups include substituted versions thereof, e.g. where a hydroxyl ethyl group will be understood to be a derivative and/or a modification of the ethyl groups described above.

[0028] Each R^1 may be the same as or different from any other R^1 of the polysiloxane (A). In certain embodiments, each R^1 is the same as each other R^1 of the polysiloxane (A). For example,

in some such embodiments, each R^1 is methyl. In other embodiments, at least one R^1 is different from at least one other R^1 of the polysiloxane (A). For example, in certain embodiments, R^1 is predominantly methyl throughout the polysiloxane (A), with one or more other groups pending from the polydiorganosiloxane backbone in minor amounts (e.g. from the preparation of the polysiloxane (A), environmental reactions or impurities, etc.). In some embodiments, each R^1 is a fluoroalkyl group, i.e. such that the polysiloxane (A) may be further defined or referred to as a fluorosilicone or fluoropolysiloxane.

[0029] As introduced above, the polysiloxane (A) comprises, on average, at least one functional group per molecule, as represented by moiety X in the general formula of the polysiloxane (A) above. In some embodiments, however, the polysiloxane (A) comprise an average of at least two functional groups X per molecule.

[0030] As described in herein, the functional groups X of the polysiloxane (A) are reactable with the functional groups Y of the functionalized polyolefin (B) to form a bond therebetween. In other words, one functional group X and one functional group Y are capable of reacting together (i.e., via a coupling reaction, cross-linking reaction, etc.), to covalently bond together the polysiloxane (A) and the functionalized polyolefin (B). It is to be understood that terms such as "coupling," "coupleable," "reactable," "cross-linking," and "cross-linkable" used herein are not intended to imply any directionality to a reaction, but instead will be understood in the customary sense to refer to the coupling facilitated by groups X and Y without inference as to particular reactivity or role in the reaction therebetween. As described herein, in some embodiments, the average molecules of components (A) and/or (B) have at least two groups capable of participating in the coupling reaction, such that a single molecule of the polysiloxane (A) may be, on average, capable of being coupled at least once to two or more molecules of the functionalized polyolefin (B) or, likewise, at least twice to a single molecule of the functionalized polyolefin (B).

[0031] In general, each functional group X comprises, alternatively is, a functional group that may participate in the coupling/cross-linking reaction described above. Examples of such functional groups are typically reactive via substitution reaction, addition reaction, coupling reaction, or combinations thereof. Specific examples of such reactions include nucleophilic substitutions, ring-opening additions, alkoxylations and/or transalkoxylations, hydrosilylations, olefin metatheses, condensations, radical couplings and/or polymerizations, and the like, as well as combinations thereof. Accordingly, functional groups X may comprise, or be, a functional group that is hydrosilylatable (e.g. a silicon-bonded hydrogen atom, an olefinically (i.e., ethylenically) unsaturated group, such as an alkenyl group, alkynyl group, etc.), condensable (e.g. a hydroxyl group, a carboxyl group, a carbinol group, an alkoxysilyl group, a silanol group, an amide group, an anhydride group, etc., or a group that may be hydrolyzable and subsequently condensable), displaceable (e.g. a "leaving group" as understood in the art, such as a halogen

atom, or other group stable in an ionic form once displaced, or a functional group comprising such a leaving group, such as esters, anhydrides, amides, epoxides, etc.), nucleophilic (e.g. a heteroatom with lone pairs, an anionic or anionizable group, etc., such as a hydroxyl group (e.g. of a carbinol), an amine group, a thiol group, a silanol group, a carboxylic acid group, etc.), electrophilic (e.g. isocyanates, epoxides, etc.), or various combinations thereof.

[0032] In some embodiments, at least one, alternatively at least two, alternatively each functional group X is a hydrosilylatable group, and thus selected from olefinically-unsaturated groups (e.g. ethylenically unsaturated groups) and H. In some such embodiments, each hydrosilylatable group represented by X is H, such that the polysiloxane (A) is silicon hydride-functional. In other of such embodiments, each hydrosilylatable group represented by X is an ethylenically unsaturated group.

[0033] Examples of ethylenically unsaturated groups generally include substituted or unsubstituted hydrocarbon groups having at least one alkene or alkyne functional group. For example, in certain embodiments, each functional group X comprises, alternatively is, an alkenyl group or an alkynyl group. Specific examples thereof include $H_2C=CH–$, $H_2C=CHCH_2–$, $H_2C=CHCH_2CH_2–$, $H_2C=CH(CH_2)_3–$, $H_2C=CH(CH_2)_4–$, $H_2C=C(CH_3)–$, $H_2C=C(CH_3)CH_2–$, $H_2C=C(CH_3)CH_2CH_2–$, $H_2C=C(CH_3)CH_2CH(CH_3)–$, $H_2C=C(CH_3)CH(CH_3)CH_2–$, $H_2C=C(CH_3)C(CH_3)_2–$, $HC\equiv C–$, $HC\equiv CCH_2–$, $HC\equiv CCH(CH_3)–$, $HC\equiv CC(CH_3)_2–$, and $HC\equiv CC(CH_3)_2CH_2–$. In specific embodiments, each functional group X comprises, alternatively is, a vinyl group.

[0034] In embodiments where the functional group X comprises one of the ethylenically unsaturated groups above, it will be appreciated that the functional group X may also comprise a divalent linking group between the ethylenically unsaturated group and a silicon atom of the polysiloxane (A). Examples of such divalent linking groups include divalent versions of the hydrocarbyl groups described above, such as alkyl groups. For example, the functional group X may have the formula $H_2C=CH–(CH_2)_5–$, which may be considered to represent the alkenyl group $H_2C=CHCH_2–$ with a butylene linking group, the alkenyl group $H_2C=CH(CH_2)_4–$ with a methylene linking group, etc. In certain embodiments, each functional group X comprises, alternatively is a methacryloxy group, such as a silicon-bonded methacryloxyalkyl group.

[0035] In some embodiments, at least one, alternatively at least two, alternatively each functional group X comprises, alternatively is, a condensable group, i.e., is capable of participating in a condensation reaction. In specific embodiments, each functional group X comprises a condensable group selected from anhydride groups, amine groups, silanol groups, carbinol groups, and alkoxysilyl groups.

[0036] Examples of suitable anhydrides for functional group X generally include anhydrides of monocarboxylic acids (e.g. acetic acid, lactic acid, propanoic acid, pentanoic acid, methacrylic acid, etc.), which may be homoanydrides or mixed anhydrides, as well as polycarboxylic acids such as succinates (i.e., succinic anhydrides), maleates (i.e., maleic anhydrides), phthalates, etc. One of skill in the art will appreciate that various substitution patterns are possible with such anhydrides in terms of linking the same to the silicon atom of the polysiloxane (A). Typically, such anhydrides may be grafted onto a siloxane polymer to prepare the polysiloxane (A), and thus one of skill in the art will understand the applicability of other anhydrides, and carboxylic acids/carboxylates that may also be utilized, e.g. via grafting directly to the polysiloxane (A) or instead via an initial grafting and subsequent reaction to prepare the anhydride. For example, anhydrides containing at least one olefinically unsaturated group, such as alkenylsuccinic anhydrides, bromomaleic anhydride, chloromaleic anhydride, citraconic anhydride, methylnadic anhydride, nadic anhydride, tetrahydrophthalic anhydride, and the like, may be grafted onto a siloxane (e.g. via hydrosilylation). Free radical based grafting schemes may also be used to produce anhydride functional siloxanes from reagents such as maleic anhydride and vinylsiloxanes.

[0037] Examples of suitable amines for functional group X generally include primary amino-substituted derivatives of the hydrocarbyl groups described above. For example, functional group X may comprise, alternatively may be, an aminoalkyl group, such as an amino-substituted alkyl group having from 1 to 20 carbon atoms (e.g. aminomethyl, 2-aminoethyl, 3-aminopropyl, 6-aminohexyl, an aminoaryl group (e.g. 4-aminophenyl, 3-(4-aminophenyl) propyl, etc.), or an aminoalkylamino group (e.g. N-(2-aminoethyl)-3-aminopropyl, N-(2-aminoethyl)-3-aminoisobutyl, etc.). In specific embodiments, the polysiloxane (A) includes only amino functionality as the functional group X.

[0038] In some embodiments, at least one, alternatively at least two, alternatively each functional group X comprises, alternatively is, a silanol group. In certain embodiments where at least one functional group X comprises a silanol group, the silicon atom of the silanol group is a silicon atom of the backbone of the polysiloxane (A). In other embodiments, at least one X comprises, alternatively is, a moiety of formula $–D\text{-}SiR^1_{3-c}(OH)_c$, where each D is a covalent bond, an oxygen atom, or a divalent hydrocarbon group, R^1 is independently selected and defined above, and subscript c is 1, 2, or 3. When subscript c is 3, the silicon atom of the silanol group includes three silicon-bonded hydroxyl groups; when subscript c is 2, the silicon atom of the silanol group includes two silicon-bonded hydroxyl groups; when subscript c is 1, the silicon atom of the silanol group includes two silicon-bonded hydroxyl groups. In certain embodiments, D is an oxygen atom. In other embodiments, D is a divalent hydrocarbon group having from 2 to 18, alternatively from 2 to 16, alternatively from 2 to 14, alternatively from 2 to 16, alternatively from 2 to 12,

alternatively from 2 to 10, alternatively from 2 to 8, alternatively from 2 to 6, alternatively from 2 to 4, carbon atoms.

[0039] In some embodiments, each functional group X comprises, alternatively is a silicon-bonded hydroxyl group.

[0040] In some embodiments, at least one, alternatively at least two, alternatively each functional group X comprises, alternatively is, a carbinol group. Carbinol functional groups bonded to silicon atoms in organopolysiloxanes are distinguished from silanol groups. Specifically, carbinol functional groups include a carbon-bonded hydroxyl group, and silanol functional groups include a silicon-bonded hydroxyl group. Said differently, carbinol functional groups include at least one moiety of formula –COH, whereas silanol functional groups are of formula –SiOH. These functional groups perform differently; for example, silanol functional groups can readily condense, which generally does not occur with carbinol functional groups (at least under the same catalysis of hydrolysis/condensation of silanol functional groups). The carbinol functional groups can be the same as or different from one another.

[0041] In certain embodiments, the carbinol functional groups independently include a moiety having the general formula $–D^1–O_d–(C_eH_{2e}O)_f–H$, where D^1 is a covalent bond or a divalent hydrocarbon linking group having from 2 to 18 carbon atoms, subscript d is 0 or 1, subscript e is independently selected from 2 to 4 in each moiety indicated by subscript f, and subscript f is from 0 to 500, with the proviso that subscripts d and f are not simultaneously 0.

[0042] In some such embodiments, subscript f is at least one, such that at least one of the carbinol functional groups includes a moiety having the general formula:

$$–D^1–O_d–[C_2H_4O]_g[C_3H_6O]_h[C_4H_8O]_i–H;$$

where D^1 and subscript d are defined above, $0≤g≤500$, $0≤h≤500$, and $0≤i≤500$, with the proviso that $1≤g+h+i≤500$. In these embodiments, the carbinol functional group may alternatively be referred to as a polyether group or moiety, although the polyether group or moiety terminates with –COH, rather than –COR, where R is a monovalent hydrocarbon group, which is the case with certain conventional polyether groups or moieties. As understood in the art, moieties indicated by subscript g are ethylene oxide (EO) units, moieties indicated by subscript h are propylene oxide (PO) units, and moieties indicated by subscript i are butylene oxide (BO) units. The EO, PO, and BO units, if present, may be in block or randomized form in the polyether group or moiety. The relative amounts of EO, PO, and BO units, if present, can be selectively controlled based on desired properties, e.g. hydrophilicity and other properties.

[0043] In other embodiments, subscript f is 0 and subscript d is 1 such that at least one of the carbinol functional groups includes a moiety having the general formula: $–D^1$-OH, where D^1 is

described above. In these embodiments, the carbinol functional groups having this general formula are not polyether groups or moieties.

[0044] In some embodiments, at least one, alternatively at least two, alternatively each functional group X comprises, alternatively is, an alkoxysilyl group.

[0045] In embodiments where at least one X is an alkoxysilyl group, each alkoxysilyl group may independently comprise or be a monoalkoxysilyl group, dialkoxysilyl group, or trialkoxysilyl group, respectively. In particular embodiments, the alkoxysilyl group comprises, alternatively is, a monoalkoxysilyl group. In other embodiments, the alkoxy group comprise, alternatively is, a dialkoxysilyl group. In yet other embodiments, the alkoxysilyl group comprises, alternatively is, a trialkoxysilyl group.

[0046] In certain embodiments, the silicon atom of the alkoxysilyl group is a silicon atom of the backbone of the polysiloxane (A). In other embodiments, at least one X comprises, alternatively is, a moiety of formula $–D^2\text{-}SiR^1_{3-j}(OR^6)_j$, where each D^2 is a covalent bond, an oxygen atom, or a divalent hydrocarbon group, R^1 is independently selected and defined above, subscript j is 1, 2, or 3, and each R^6 is an independently selected alkyl group having from 1 to 12 carbon atoms. Typically, each R^6 is an independently selected alkyl group having from 1 to 10, alternatively from 1 to 8, alternatively from 1 to 6, alternatively from 1 to 4, alternatively 1 to 3, alternatively 1 or 2, alternatively 1, carbon atoms.

[0047] Specific examples of alkoxysilyl groups include those comprising trimethoxysilyl groups, triethoxysilyl groups, dimethoxyethoxysilyl groups, dimethoxymethyl groups, diethoxymethyl groups, methoxyethoxymethyl groups, dimethylmethoxy groups, dimethylethoxy groups, etc.

[0048] In some embodiments, at least one, alternatively at least two, alternatively each functional group X comprises, alternatively is, an epoxy group. Examples of suitable epoxy groups include a 3-glycidoxypropyl group, a 4-glycidoxybutyl group, or similar glycidoxyalkyl (i.e., glycidyloxyalkyl) groups; a 2-(3,4-epoxycyclohexyl)ethyl group, a 3-(3,4-epoxycyclohexyl)propyl group, or similar epoxycyclohexylalkyl groups; a 4-oxiranylbutyl group, and an 8-oxiranyloctyl group. Such epoxy groups may be bonded directly to the polysiloxane (A). Alternatively, a divalent hydrocarbon group may be present in X between the epoxide group and the silicon atom to which X is bonded. In some of these embodiments, the divalent hydrocarbon group comprises, alternatively is, an alkylene group having the general formula $–(CH_2)_k–$, where subscript k is from 2 to 10. In other embodiments, the divalent hydrocarbon group comprises from 2 to 10 carbon atoms and includes at least one ether moiety, i.e., at least one oxygen heteroatom. Such divalent hydrocarbon groups may also be suitable for use as the divalent linking groups described herein (e.g. D, D^1, D^2, etc.).

[0049] With reference to the general unit formula of the polysiloxane (A) above, subscript m is independently 1 or 0 in each moiety indicated by subscript a, subscript n is independently 1 or 0 in each moiety indicated by subscript b. As such, subscripts m and n merely indicate the presence of the functional group X in any particular [M] unit (i.e., as indicated by subscript a) or [D] unit (i.e., as indicated by subscript b). When each subscript m is 0 (i.e., subscript m is 0 in each moiety indicated by subscript a), the polysiloxane (A) comprises at least one pendant functional X (i.e., bonded to a [D] unit). When each subscript n is 0 (i.e., subscript n is 0 in each moiety indicated by subscript b), the polysiloxane (A) comprises at least one terminal functional X (i.e., bonded to an [M] unit). In some embodiments, the polysiloxane (A) is only terminally-functional with respect to the functional groups X, such that subscript n is 0 in each moiety indicated by subscript b. In some such embodiments, the polysiloxane (A) comprises at least two moieties indicated by subscript a, and subscript m is 1 in at least two moieties indicated by subscript a. In other embodiments, the polysiloxane (A) comprises at least one terminal functional group X. In yet other embodiments, the polysiloxane (A) comprises only pendant functionality with respect to the functional groups X, such that subscript m is 0 in each moiety indicated by subscript a, the polysiloxane (A) comprises at least two moieties indicated by subscript b, and subscript n is 1 in at least two moieties indicated by subscript b.

[0050] With continued reference to the general unit formula of the polysiloxane (A) above, subscripts a and b are each mole fractions such that $a+b=1$, with the provisos that $0<a<1$ and $0<b<1$. It will be appreciated that the moieties indicated by subscripts a and b are generally [M] and [D] siloxy units, respectively. As such, in certain embodiments, the polysiloxane (A) may be defined as an MDM-type polysiloxane. Accordingly, in such embodiments, the polysiloxane (A) may be defined as a linear polysiloxane (or, more simply, "linear siloxane"). Nonetheless, it is to be appreciated that the general formula above may be an average unit formula, i.e., the average formula based on all molecules in the polysiloxane (A). As such, as described above with regard to siloxanes generally, the polysiloxane (A) may comprise a limited amount of branching (e.g. attributable to [T] and/or [Q] units) without departing from the scope of linearity understood by those of skill in the art, even though such units are not included in the general unit formula above. Typically, the polysiloxane (A) is substantially free from, alternatively free from, [T] and/or [Q] units.

[0051] In some embodiments, each of the units represented by subscripts a and b are independently selected, and at least two units of the polysiloxane (A) comprise the functional group X. In such embodiments, the preceding general formula for the polysiloxane (A) may be rewritten as the following expanded average unit formula:

$$[XR^1{}_2SiO_{1/2}]_{a'}[XR^1SiO_{2/2}]_{b'}[R^1{}_2SiO_{2/2}]_{b''}[R^1{}_3SiO_{1/2}]_{a''},$$

where each X and R^1 are as defined above, subscripts a', a'', b', and b'' each indicate the number of corresponding moieties are present in the polysiloxane (A). In this fashion, a'+a'' is equal to number of [M] siloxy units present in the mole fraction represented by subscript a in the general formula above, and b'+b'' is equal to number of [D] siloxy units present in the mole fraction represented by subscript b in the general formula above. For example, in general, a'+a''≥2, a'+b' ≥ 2, and b'+b''≥ 1. In the specific embodiments described above, where the polysiloxane (A) is only terminally functional, a' = 2, b' = 0, b'' ≥ 1, and a'' = 0.

[0052] In general, the polysiloxane (A) may have a number average degree of polymerization (DP) of from 10 to 10,000. As such, with reference to the preceding expanded formula of the polysiloxane (A), a'+a''+b'+b'' is generally from 10 to 10,000. In some embodiments, the polysiloxane (A) has a DP of from 10 to 1200, alternatively from 50 to 1200. Likewise, in these embodiments, a'+a''+b'+b'' is generally from 10 to 1200, alternatively from 20 to 1200, alternatively from 50 to 1200. For example, in some embodiments, the polysiloxane (A) has a DP of from 50 to 1100, alternatively from 50 to 1000, alternatively from 100 to 1000. In specific embodiments, a' and a'' are each from 0 to 2, typically with a'+a''=2. Subscript b'' may be from 0 to 10,000, such as from 5 to 5,000, alternatively from 50 to 1200, alternatively from 50 to 1100, alternatively from 50 to 1000, alternatively from 100 to 1000. In these or other embodiments, subscript b' is from 0 to 200, such as from 0 to 10, alternatively from 1 to 10, alternatively from 1 to 8.

[0053] In certain embodiments, the polysiloxane (A) has a degree of substitution (DS) of from 1 to 200. It will be appreciated that the DS of the polysiloxane (A) may be represented by the sum of subscripts a' and b' in the expanded formula above, i.e., which indicates the number of functional groups X. In some embodiments, the polysiloxane (A) has a DS of from 1 to 100, alternatively from 1 to 50, alternatively from 1 to 20, alternatively from 1 to 10, alternatively from 2 to 10.

[0054] In some embodiments, the polysiloxane (A) comprises a molecular weight distribution, as represented by polydispersity index (PDI) (i.e., the weight average molecular weight/number average molecular weight (Mw/Mn), of less than 3, alternatively less than 2.5, alternatively less than 2.25, and at the same time greater than or equal to 1. For example, the polysiloxane (A) may comprise a PDI of from 1 to 3, such as from 1 to 2.5, alternatively from 1.5 to 2.5, alternatively from 1.5 to 2.2, alternatively from 1.8 to 2.2, alternatively of about 2. Methods of determining the PDI for the polysiloxane (A) are known in the art, and generally include weight determinations via rheology, solution viscosity, gel permeation chromatography (GPC), etc., with standards and procedures readily understood and available.

[0055] Typically, the polysiloxane (A) utilized in the silicone-polyolefin composition is flowable, i.e., comprises a viscosity low enough to exhibit flow under ambient conditions (e.g. at 25 °C). In

some embodiments, the polysiloxane (A) is a liquid at room temperature. In certain embodiments, the polysiloxane (A) exhibits a viscosity of at least 1000 cP, alternatively of at least 3500 cP, at 25 °C (e.g. as determined via viscometer, such as a Brookfield LV DV-E viscometer, equipped with an appropriate spindle).

[0056] In certain embodiments, the polysiloxane (A) is further defined as a functionalized polydimethylsiloxane (PDMS), i.e., where each R^1 is methyl. In some such embodiments, the polysiloxane (A) is selected from amine-functional PDMS (i.e., where each functional group X comprises an amine, such as a primary aminoalkyl group) and vinyl-functional PDMS (i.e., where each functional group X comprises, alternatively is, a vinyl group).

[0057] In view of the description above, examples of such amine-functionalized PDMS suitable for use in, or as, the polysiloxane (A) will be understood to include terminal and/or pendant amine-functional PDMS oligomers and polymers. However, it will also be understood that, in certain embodiments, the polysiloxane (A) may comprise, alternatively may be, a terminal and/or pendant amine-functional random, graft, or block copolymer or co-oligomer of PDMS and a non-reactive siloxane (e.g. a polyphenylmethylsiloxane, a tris(trifluoropropyl)methylsiloxane, etc.). In this same fashion, examples of vinyl-functionalized PDMS suitable for use in, or as, the polysiloxane (A) include terminal and/or pendant vinyl-functional PDMS oligomers and polymers, as well as random, graft, or block copolymer or co-oligomer of PDMS and a non-reactive. In specific embodiments, the polysiloxane (A) comprises, alternatively is, an aminoalkyl-terminated PDMS, such as an α,ω-aminopropyl-terminated PDMS. In certain embodiments, the polysiloxane (A) comprises, alternatively is, a vinyl-terminated PDMS, such as an α,ω-vinyl-terminated PDMS. In some embodiments, the polysiloxane (A) comprises, alternatively is, is a methacryloylpropyl-terminal PDMS, a silanol-terminal PDMS, a succinic anhydride-terminal PDMS, a SiH-terminal PDMS, a vinyl-terminated PDMS, a mono carbinol-functional PDMS, or an aminopropyl-terminated PDMS.

[0058]

[0059] As introduced above, the silicone-polyolefin composition also comprises the functionalized polyolefin (B). The functionalized polyolefin (B) comprises an average of at least one functional group Y per molecule, e.g. as a substituent of a polyolefin backbone. In some embodiments, the functionalized polyolefin (B) comprises an average of at least two functional group Y per molecule. As described herein, the functional groups Y are reactable with the functional groups X of the polysiloxane (A) to form a bond therebetween. As such, it will be understood that component (B) of the silicone-polyolefin composition generally comprises a polyolefin that is prepared with, obtained with, or otherwise functionalized to include the functional groups Y as substituents. Accordingly, the functionalized polyolefin (B) may comprise,

alternatively may be, a terminally-substituted (i.e., a functional group-terminated) polyolefin, a pendantly-substituted polyolefin, or a combination thereof.

[0060] In general, polyolefins suitable for the functionalized polyolefin (B) are exemplified by polymers prepared from olefinic monomers, olefinic macromonomer and oligomers, and combinations thereof. Regardless of the actual synthetic route by which the functionalized polyolefin (B) is prepared, one of skill in the art will readily appreciate the scope of the polyolefin component of the functionalized polyolefin (B) in terms of its constituent parts (or theoretical constituent parts), i.e., the olefinic base monomers polymerized to prepare the polyolefin. The term "olefinic" used in the context of the base monomers composing the functionalized polyolefin (B) refers to the presence of an ethylenically unsaturated end group, i.e., which is polymerizable with an ethylenically unsaturated group of other olefinic monomer to provide a polyolefin. In this fashion, it will be understood that a "polyethylene" is a polyolefin derived, or theoretically derivable, from the monomer ethene (ethylene), which is the smallest ethylenically unsaturated compound. Likewise, a polyethylene-methacrylate copolymer is a polyolefin derived, or theoretically derivable, from the comonomers ethylene and methacrylate, with the latter monomer comprising a terminal ethylenically unsaturated group, i.e., an alpha-olefin (e.g. ~$C=CH_2$). As such, it will be appreciated that, in typical embodiments, the functionalized polyolefin (B) comprises a poly-alpha-olefin backbone.

[0061] The poly-alpha-olefin backbone of the functionalized polyolefin (B) is not particularly limited, and generally comprises monomeric units derived, or at least theoretically derivable from, an alpha olefin having the general formula $R^2{}_2C=CH_2$, where each R^2 is hydrogen or a hydrocarbyl group (i.e., a substituted or unsubstituted hydrocarbyl group), such as any of those described above. For example, in certain embodiments one R^2 is methyl and the other R^2 is an esteric carbon, such that the alpha olefin is a methacrylate (e.g. a methyl or ethyl methacrylate, where the other R^2 is a methyl ester or ethyl ester, respectively). In some embodiments, at least one R^2 is hydrogen and the alpha olefin has the general formula $R^2CH=CH_2$, where R^2 is selected from hydrogen and linear or branched hydrocarbyl groups having from 1 to 12, alternatively from 1 to 8, alternatively from 1 to 6, alternatively 1 or 2, carbon atoms. Such hydrocarbyl groups may be substituted or unsubstituted, and are exemplified above with regard to the appropriate descriptions of hydrocarbyl groups for R" and R^1. It will be appreciated, however, that oligomers of such alpha olefins may also be utilized in the preparation of the poly-alpha-olefin backbone. For example, polyethylene (PE) oligomers may be utilized to prepare a polyethylene polymer, which may also be prepared using ethene as the sole monomer. Likewise, polyethylene (PE) and polypropylene (PP) oligomers may be copolymerized to prepare a polyethylene-polypropylene (PE-PP) copolymer, such as a PE-PP block copolymer. Examples of

other oligomers that may be used to prepare poly-alpha-olefin backbone of the functionalized polyolefin (B) include polypropylene oligomers, polybutylene oligomers, polyisobutylene oligomers, polyisoprene oligomers, polybutadiene oligomers, as well as combinations thereof, such as polyethylene/polypropylene oligomers and copolymers, polyethylene/polybutylene oligomers and copolymers, poly(ethylene/butylene)-polyisoprene oligomers and copolymers, etc.

[0062] In certain embodiments, the functionalized polyolefin (B) comprises a poly-alpha-olefin backbone comprising monomeric units selected from ethylene, propylene, butylene, and 2-methyl-propylene (i.e., isobutylene). In these or other embodiments, the poly-alpha-olefin backbone comprises monomeric units derived (or theoretically derivable) from alpha-olefins exemplified by hexene, heptene, octene, styrene, an acrylate or methacrylate compound (e.g. acrylic acid, methacrylic acid, acrylonitrile, methacrylonitrile, an acrylic or methacrylic ester such as a C_1-C_{12} alkyl ester of acrylic or methacrylic acid, etc.), dienes such as butadiene, etc., or combinations thereof. Accordingly, it will be appreciated that the functionalized polyolefin (B) may comprise, alternatively may be, a homopolymer (i.e., having but one type of monomeric unit, or prepared from but one monomer or oligomers of but one monomer) or an interpolymer (i.e., having at least two different monomeric subunits, typically prepared from at least two monomers or oligomers comprising two or more monomeric subunits). It will be understood that the term "interpolymer" encompasses copolymers and terpolymers, i.e., polymers comprising two, or three, different monomeric units, respectively, as well as polymers prepared from four, five, six, or more monomers.

[0063] In particular embodiments, the functionalized polyolefin (B) comprises a functionalized polyethylene, polypropylene, or polyethylene-alpha olefin copolymer. In some such embodiments, the polyethylene-alpha olefin copolymer is selected from copolymers and terpolymers comprising polyethylene and at least one of polypropylene and polybutylene. Various forms of such polyolefins may also be utilized. For example, polyethylenes may be high density (HDPE, $\rho \geq 0.941$ g/cm^3), medium density (MDPE, $\rho = 0.926$-0.940 g/cm^3), low density, (LDPE, $\rho = 0.910$-0.940 g/cm^3) or ultra-low density (ULDPE, $\rho \leq 0.880$ g/cm^3), and variations thereof, such as linear-low density polyethylene (LLDPE, $\rho = 0.915$-0.925 g/cm^3). While such polyethylenes are distinguished from one another by density, one of skill in the art will appreciate that various other physical characteristics of such variants differ as well, and may be selected to impart particular properties on the silicone-polyolefin composition, as well as the curable composition and cured products that may be prepared therefrom.

[0064] As introduced above, the functionalized polyolefin (B) comprises an average of at least one, alternatively at least two functional groups Y per molecule. For purposes of illustration, the functionalized polyolefin (B) may be represented by the general formula $L(\text{-}Y)_l$, where L is the

polyolefin backbone, each Y is a functional group as introduced above, and subscript l≥1. It will be appreciated that each functional group Y may be independently selected in each moiety indicated by subscript l, which is at least one, alternatively at least two, but may theoretically be much larger, as will be understood in view of the description of the degree of substitution of the functionalized polyolefin (B. Moreover, the location of each functional group Y along the polyolefin backbone L is not particularly limited, such that any functional group Y may represent a terminal or pendant group.

[0065] In some embodiments, the functionalized polyolefin (B) comprises the following general unit formula:

$R^4[CH_2C(R^3)(Y)]_o[CH_2CH(R^3)]_pR^4$

where each Y is an independently selected functional group as introduced above, each R^3 is independently selected from H and substituted and unsubstituted hydrocarbyl groups, each R^4 is an independently selected terminal group, and subscripts o and p are each mole fractions such that o+p=1, with the provisos that 0<o<1, 0<p<1, the functionalized polyolefin (B) comprises at least one, alternatively at least two functional groups Y, and the moieties indicated by subscripts o and p may be in any order in the functionalized polyolefin (B).

[0066] With reference to the general unit formula of the functionalized polyolefin (B) above, one of skill in the art will appreciate that the group R^3 is generally selected or otherwise controlled based on the particular alpha-olefin monomers used to prepare the functionalized polyolefin (B), or at least the backbone thereof. For example, where the functionalized polyolefin (B) is a functionalized polypropylene, R^3 is methyl in each moiety indicated by subscript k. In such instances, the nature of R^3 in the moieties indicated by subscript j will depend on how the functionalized polyolefin (B) was prepared. In particular, where such a functionalized polyethylene is a copolymer prepared from polypropylene and an alpha olefin comprising group Y, R^3 will be H in each moiety indicated by subscript o (as opposed to the methyl groups R^3 in the moieties indicated by subscript p). On the other hand, where such a functionalized polypropylene is a polypropylene homopolymer grafted with functional groups Y (e.g. via radial mediated grafting), R^3 will typically be a methyl group throughout the functionalized polyolefin (B). As such, it will be appreciated that any R^3 may be selected such that any one moiety indicated by subscript p may reflect a polymerization product of any of the alpha-olefin monomers described herein or, alternatively, a grafting-functionalization onto a polymer prepared from such alpha-olefins.

[0067] In view of the above, it will be appreciated that each R^3 may be the same as or different from any other R^3 of the functionalized polyolefin (B). In certain embodiments, each R^3 is the

same as each other R^3 of the functionalized polyolefin (B). For example, in some such embodiments, each R^3 is methyl. In other embodiments, at least one R^3 is different from at least one other R^3 of the functionalized polyolefin (B). For example, in certain embodiments, R^3 is predominantly hydrogen throughout the functionalized polyolefin (B) (i.e., from ethene monomer), with a minor proportion of R^3 being selected from alkyl groups (i.e., from propene or higher-order alpha-olefin monomer).

[0068] As introduced above, each R^4 is an independently selected terminal group. More specifically, each R^4 generally represents a terminally reacted monomer from the polymerization of the functionalized polyolefin (B), the byproduct of polymerization (i.e., from a radial initiation, propagation, and/or termination step, etc.), or simply a hydrogen atom. One of skill in the art will appreciate that the R^4 is thus not particularly limited, will generally be selected by virtue of the route by which the functionalized polyolefin (B), and is typically present in the functionalized polyolefin (B) in such minor amounts as to not substantively impact the average unit formula indicated by subscripts o and p. As such, it is to be understood that R^4 generally represents an unreactive group with regard to the compositions and methods provided herein.

[0069] As introduced above, the functionalized polyolefin (B) comprises at least one, alternatively at least two functional groups per molecule, which are represented by moiety Y in the general unit formula of the functionalized polyolefin (B) above. In general, the functional groups Y are selected based on the functional group X of the polysiloxane (A), such that the functionalized polyolefin (B) is reactive with the polysiloxane (A) in a coupling reaction involving functional group X and functional group Y. More specifically, as introduced above, the functional groups Y of the functionalized polyolefin (B) is reactable with the functional group X of the polysiloxane (A) to form a bond therebetween. In other words, one functional group Y and one functional group X are capable of reacting together (i.e., via additive coupling/cross-linking reaction), to covalently bond together the functionalized polyolefin (B) and the polysiloxane (A).

[0070] Accordingly, each functional group Y comprises a functional group that may participate in the coupling/cross-linking reaction described above, such as a functional group reactive via substitution reaction, addition reaction, coupling reaction, or combinations thereof, as well as any of the specific variants described above with respect to the functional groups X. Specific examples of such reactions include nucleophilic substitutions, ring-opening additions, alkoxylations and/or transalkoxylations, hydrosilylations, olefin metatheses, condensations, radical couplings and/or polymerizations, and the like, as well as combinations thereof.

[0071] Accordingly, functional group Y may comprise, or be, a functional group that is hydrosilylatable, condensable, displaceable, nucleophilic, or otherwise reactable (e.g. graftable, linkable, etc.) with the functional group X, or various combinations thereof. As such, functional

group Y may comprise, or be, a functional group that is hydrosilylatable (e.g. a silicon-bonded hydrogen atom, an olefinically (i.e., ethylenically) unsaturated group, such as an alkenyl group, alkynyl group, etc.), condensable (e.g. a hydroxyl group, a carboxyl group, a carbinol group, an alkoxysilyl group, a silanol group, an amide group, an anhydride group, etc., or a group that may be hydrolyzable and subsequently condensable), displaceable (e.g. a "leaving group" as understood in the art, such as a halogen atom, or other group stable in an ionic form once displaced, or a functional group comprising such a leaving group, such as esters, anhydrides, amides, epoxides, etc.), nucleophilic (e.g. a heteroatom with lone pairs, an anionic or anionizable group, etc., such as a hydroxyl group (e.g. of a carbinol), an amine group, a thiol group, a silanol group, a carboxylic acid group, group, etc.), electrophilic (e.g. isocyanates, epoxides, etc.), or various combinations thereof.

[0072] In some embodiments, each functional group Y is a hydrosilylatable group. Examples of such hydrosilylatable groups include the olefinically-unsaturated groups (e.g. ethylenically unsaturated groups) described above with respect to the hydrosilylatable groups suitable for functional group X. In specific embodiments, each functional group Y comprises, alternatively is, a vinyl-substituted organosilicon group (e.g. comprises a vinylsilyl group).

[0073] In embodiments where the functional group Y comprises one of the ethylenically unsaturated groups above, it will be appreciated that the functional group Y may also comprise a divalent linking group between the ethylenically unsaturated group and a carbon atom of the polyolefin backbone of the functionalized polyolefin (B). In certain embodiments, each functional group Y comprises, alternatively is, a methacryloyl group, methacryloxy group, or a methacrylate group.

[0074] Other examples of hydrosilylatable groups suitable for functional group Y include hydridosilyl groups. Examples of such hydridosilyl groups may be generally represented by the subformula $–[D^3]_q–Si(R^5)_2H$, where D^3 is a divalent linking group, subscript q is 0 or 1, and each R^5 is independently H or a hydrocarbyl group. Such moieties may be selected, or otherwise provided, based on a particular alpha-olefin-functional organosilicon compound polymerized in the preparation of the functionalized polyolefin (B). For example, in some embodiments, the functionalized polyolefin (B) comprises a copolymer of ethylene and 7-octenyldimethylsilane, such that, with regard to the preceding general unit formula of the functionalized polyolefin (B) and subformula of the functional group Y, each R^3 is H, each subscript q is 1, each linking group D^3 is $–(CH_2)_6–$, and each R^5 is methyl. In specific embodiments, the functionalized polyolefin (B) comprises the polymerization reaction product of ethylene, an alkenyl-functional silane compound, and optionally one or more additional alpha-olefins (e.g. propene, butene, etc.). In such embodiments, examples of suitable alkenyl-functional silane compounds include 7-

octentyldimethylsilane (ODMS), 5-hexenyldimethylsilane (HDMS), allyldimethylsilane (ADMS), and the like, as well as combinations thereof. It will be appreciated that such alkenyl-functional silane compounds may also be grafted onto a polyolefin polymer to prepare the functionalized polyolefin (B). The particular method used to prepare the functionalized polyolefin (B) is not particularly limited, and numerous examples of such methods are known in the art.

[0075] In addition to the preceding examples, one of skill in the art will appreciate that other alpha-olefin-functional organosilicon compounds may likewise be used to prepare the functionalized polyolefin (B) to give hydridosilyl groups suitable for functional group Y. More specifically, such organosilicon compounds may be represented by the formula $H_2C=C(H)–[D^3]_q–Si(R^5)_2H$, where each R^5, D^3, and subscript q are as defined above. In some instances, divalent linking group D^3 may be silyl or siloxy group.

[0076] In some embodiments, each functional group Y is a condensable group, i.e., capable of participating in a condensation reaction. In specific embodiments, each functional group Y comprises condensable group selected from anhydride groups, amine groups, silanol groups, carbinol groups, and alkoxysilyl groups.

[0077] Examples of suitable anhydrides and amines for functional group Y generally include those described above with respect to condensable groups suitable for functional group X. However, in view of the preceding examples and descriptions of the functionalized polyolefin (B), one of skill in the art will appreciate that anhydrides available from anhydride-functional compounds with olefinic unsaturation will be particularly suitable for use in some embodiments, such as where the anhydride-functional compound can be readily copolymerized with an alpha-olefin monomer (e.g. ethene), or grafted onto an alpha-olefin homopolymer (e.g. via radical grafting, metathesis, etc.).

[0078] Examples of suitable amines for functional group Y generally include primary amino-substituted derivatives of the hydrocarbyl groups described above, as the aminoalkyl groups described above with respect to the functional group X.

[0079] In some embodiments, at least one, alternatively at least two, alternatively functional group Y comprises, alternatively is, a silanol group. In certain embodiments, at least one functional group Y comprises, alternatively is, a moiety of formula $–D\text{-}SiR^1_{3-c}(OH)_c$, where each D, R^1, and subscript c is independently selected and defined above. In certain embodiments, D is an oxygen atom. In other embodiments, D is a divalent hydrocarbon group having from 2 to 18, alternatively from 2 to 16, alternatively from 2 to 14, alternatively from 2 to 16, alternatively from 2 to 12, alternatively from 2 to 10, alternatively from 2 to 8, alternatively from 2 to 6, alternatively from 2 to 4, carbon atoms.

[0080] In some embodiments, at least one, alternatively at least two, alternatively each functional group Y comprises, alternatively is, a carbinol group. The carbinol functional groups can be the same as or different from one another.

[0081] In certain embodiments, the carbinol functional groups independently include a moiety having the general formula $–D^1–O_d–(C_eH_{2e}O)_f–H$, where D^1 and subscripts d, e, and f are independently selected and defined above. For example, in some embodiments, subscript f is at least 1, such that at least one of the carbinol functional groups includes a moiety having the general formula:

$–D^1–O_d–[C_2H_4O]_g[C_3H_6O]_h[C_4H_8O]_i–H;$

where D^1 and subscripts d, g, h, and i are independently selected and defined above.

[0082] In another embodiment, subscript f is 0 and subscript d is 1 such that at least one of the carbinol functional groups includes a moiety having the general formula: $–D^1$-OH, where D^1 is described above. In these embodiments, the carbinol functional groups having this general formula are not polyether groups or moieties.

[0083] D^1 may be a covalent bond when functional group Y is bonded directly to a carbon atom in the functionalized polyolefin (B).

[0084] In some embodiments, at least one, alternatively at least two, alternatively each functional group Y comprises, alternatively is, an alkoxysilyl group.

[0085] In embodiments where at least one functional group Y is an alkoxysilyl group, each alkoxysilyl group may independently comprise or be a monoalkoxysilyl group, dialkoxysilyl group, or trialkoxysilyl group, respectively. In particular embodiments, the alkoxysilyl group comprises, alternatively is, a monoalkoxysilyl group. In other embodiments, the alkoxy group comprise, alternatively is, a dialkoxysilyl group. In yet other embodiments, the alkoxysilyl group comprises, alternatively is, a trialkoxysilyl group.

[0086] In certain embodiments, at least one functional group Y comprises, alternatively is, a moiety of formula $–D^2$-$SiR^1_{3-j}(OR^6)_j$, where each D^2, R^1, R^6, and subscript q are independently selected and defined above. Typically, each R^6 is an independently selected alkyl group having from 1 to 10, alternatively from 1 to 8, alternatively from 1 to 6, alternatively from 1 to 4, alternatively 1 to 3, alternatively 1 or 2, alternatively 1, carbon atoms.

[0087] Specific examples of suitable alkoxysilyl groups include those comprising trimethoxysilyl groups, triethoxysilyl groups, dimethoxyethoxysilyl groups, dimethoxymethyl groups, diethoxymethyl groups, methoxyethoxymethyl groups, dimethylmethoxy groups, dimethylethoxy groups, etc.

[0088] In some embodiments, at least one, alternatively at least two, alternatively each functional group Y comprises, alternatively is, an epoxy group. Examples of suitable epoxy groups include a 3-glycidoxypropyl group, a 4-glycidoxybutyl group, or similar glycidoxyalkyl (i.e., glycidyloxyalkyl) groups; a 2-(3,4-epoxycyclohexyl)ethyl group, a 3-(3,4-epoxycyclohexyl)propyl group, or similar epoxycyclohexylalkyl groups; a 4-oxiranylbutyl group, and an 8-oxiranyloctyl group. Such epoxy groups may be bonded directly to the functionalized polyolefin (B). Alternatively, a divalent hydrocarbon group may be present in the functional group Y between the epoxide group and the atom to which the functional group Y is bonded. In some of these embodiments, the divalent hydrocarbon group comprises, alternatively is, an alkylene group having the general formula $-(CH_2)_k-$, where subscript k is as defined above. In other embodiments, the divalent hydrocarbon group comprises from 2 to 10 carbon atoms and includes at least one ether moiety, i.e., at least one oxygen heteroatom.

[0089] With further reference to the general unit formula of the functionalized polyolefin (B), as introduced above subscripts o and p are each mole fractions such that $o+p=1$. In general, $0<o<1$ and $0<p<1$, such that the functionalized polyolefin (B) may comprise at least one functional group Y, and theoretically many of such groups, but is not fully-substituted in terms of each olefin subunit present in the functionalized polyolefin (B) (e.g. as indicated by subscript $p>0$. In some embodiments, the moieties indicated by subscript o comprise from 0.01 to 5%, alternatively from 0.01 to 2.5% of the total number of olefin subunits in the functionalized polyolefin (B) (e.g. o+p). In these of other embodiments, the moieties indicated by subscript o may comprise from 0.05 to 10 wt.% of the functionalized polyolefin (B) (e.g. by total weight).

[0090] The particular properties and physical characteristics of the functionalized polyolefin (B) may be varied. In some embodiments, the functionalized polyolefin (B) comprises a number average molecular weight of from 10 to 100 kDa, such as from 10 to 90, alternatively from 15 to 90, alternatively from 15 to 80, alternatively from 20 to 80, alternatively from 20 to 70, alternatively from 20 to 65 kDa. In these or other embodiments, the functionalized polyolefin (B) comprises a molecular weight distribution, as represented by the polydispersity index (PDI) (e.g. as determined by gel permeation chromatography (GPC)), of from 1 to 12, such as from 1 to 10. In some embodiments, the functionalized polyolefin (B) exhibits a PDI of from 1 to 5, such as from 1 to 4, alternatively from 1.5 to 3.5, from 1.75 to 3.25, alternatively from 2 to 3. In some embodiments, the functionalized polyolefin (B) exhibits a PDI of from 3 to 6, such as from 3.5 to 5.5, alternatively from 4 to 5.

[0091] In certain embodiments, the functionalized polyolefin (B) is anhydride functional. In these or other embodiments, the functionalized polyolefin (B) comprises functional group Y in an amount of from 0.5 to 2.0 wt.%.

[0092] The functionalized polyolefin (B) typically has a melt flow index (MFI) of from 1 to 49 g/10 min. MFI can be measured in accordance with ASTM D1238–86.

[0093] As introduced above, the silicone-polyolefin composition also comprises the polyolefin (C). The polyolefin (C) is not reactable with components (A) or (B). Said differently, the polyolefin (C) does not include a functional group that is reactable with functional group X of component (A) or functional group Y of component (B). In certain embodiments, the polyolefin (C) can include a functional group so long as the functional group is not reactable with functional group X of component (A) or functional group Y of component (B). One of skill in the art readily understands such functional groups based on the selection of functional groups X and Y. Typically, however, the polyolefin (C) is free from any functional groups, i.e., groups that are reactable with other functional groups.

[0094] The polyolefin (C) may be selected from any of those described above for the functionalized polyolefin (B), with the only difference being that the polyolefin (C) does not include the functional group Y present in component (B).

[0095] In one embodiment, the polyolefin (C) is a post-consumer recycle resin. In another embodiment, the polyolefin (C) is virgin material. The polyolefin (C) can also comprise a combination of post-consumer recycle resin and virgin material.

[0096] The term "post-consumer recycle resin" (or "PCR") is a polymeric material that has been previously used as consumer packaging or industrial packaging. In other words, PCR is waste plastic. PCR is typically collected from recycling programs and recycling plants. PCR typically requires additional cleaning and processing before it can be re-introduced into a manufacturing line. The PCR is the PCR multilayer film after the PCR multilayer film has completed a first use; i.e. having already served its first purpose. It is understood PCR includes postindustrial recycle (PIR) resin. In an embodiment, the PCR multilayer film is waste barrier film that was used to hold, or otherwise to store, consumer-edible oil.

[0097] PCR is distinct from virgin polymeric material. Since PCR has gone through an initial heat and molding process; PCR is not "virgin" polymeric material. A "virgin polymeric material" is a polymeric material that has not undergone, or otherwise has not been subject to, a heat process or a molding process. The physical, chemical and flow properties PCR resin differ when compared to virgin polymeric resin.

[0098] PCR can be considered waste plastic. PCR can include, for example, HDPE packaging such as bottles (milk jugs, juice containers), LDPE/LLDPE packaging such as films. PCR can also include residue from its original use, residue such as paper, adhesive, ink, colorants, dyes, nylon, ethylene vinyl alcohol (EVOH), polyethylene terephthalate (PET), and other odor causing agents. When the PCR comprises HDPE, the PCR can comprise up to 40% polypropylene contamination.

[0099] Non-limiting examples commercially available PCR include PCR sold by Envision Plastics, North Carloina, USA, under the tradenames EcoPrime™, PRISMA™, Natural HDPE PCR Resins, Mixed Color and Black HDPE PCR Resins; PCR sold by KW Plastics, Alabama, USA under the following tradenames KWR101-150, KWR101-150-M5-BLK, KWR101-150-M10 BLK, KWR102- 8812 BLK, KWR102, KWR102LVW, KWR105, KW620, KWR102-M4, KWR-105M2, KWR105M4, KWR621 FDA, KWR621-20-FDA, KW308A, KW621, KW621-T10, KW621-T20, KW622-20, KW622-35, KW627C, KW1250G, and KWBK10-NB.

[00100] In an embodiment, the polyolefin (C) comprises PCR blended with an olefin-based polymer that is not a PCR. In other words, in this embodiment, PCR is blended with a "virgin olefin-based polymer" to give the polyolefin (C).

[00101] The polyolefin (C) typically has a melt flow index (MFI) of from 0.5 to 35 g/10 min. MFI can be measured in accordance with ASTM D1238–86.

[00102] As introduced above, the silicone-polyolefin composition also comprises the curable silicone (D). The curable silicone (D) comprises at least one of an elastomer component and a liquid rubber. In one embodiment, the curable silicone (D) comprises, alternatively is, the elastomer component. In another embodiment, the curable silicone (D) comprises, alternatively is, the liquid rubber. In yet another embodiment, the curable silicone (D) comprises a combination of the elastomer component and the liquid rubber. The elastomer component and liquid rubber are described below.

[00103] As will be understood by those of skill in the art in view of the description and examples herein, the curable silicone (D) typically represents the predominant component of the silicone-polyolefin composition, and is adapted for facilitating the formation of curable and cured compositions therefrom.

[00104] In general, the elastomer component of the curable silicone (D) comprises a functional or curable organosiloxane, i.e., a reaction-curable silicone, optionally with additional components (e.g. filler, etc., as described below). In this fashion, it will be understood by those of skill in the art that component (D) may be referred to or otherwise described in these embodiments as a “silicone elastomer base,” a “curable silicone base,” a “curable silicone elastomer base,” and/or other such terms known in the art. As such, it will be appreciated that component (D) is generally capable of being cured to prepare a silicone elastomer, and does not typically itself comprise an elastomer until such curing.

[00105] Functional organosiloxanes suitable for use in, or as, the elastomer component of the curable silicone (D) may be described in terms of the [M], [D], [T], and/or [Q] units/siloxy groups of the polymers therein. More specifically, as set forth above, the elastomer component of the curable silicone (D) typically comprises a functional organosiloxane, which may comprise any number and/or combination of M, D, T and/or Q siloxy units combined in various manners to form

cyclic, linear, branched and/or resinous structures as described above. Typically, however, the functional organosiloxane of component (D) is substantially free from, alternatively is free from, resinous segments. In these or other embodiments, the organosiloxane of component (D) is substantially free from, alternatively is free from, [T] and/or [Q] units.

[00106] Specific compounds and compositions suitable for use in, or as, the elastomer component of the curable silicone (D) are commercially available, and may be selected based on particular properties thereof. For example, in some embodiments, the elastomer component of the curable silicone (D) comprises, alternatively is, a silicone high-consistency rubber (HCR) base. In specific embodiments, component (D) may be characterized as a 40 durometer silicone rubber base.

[00107] In specific embodiments, the functional organosiloxane of the elastomer component of the curable silicone (D) comprises, alternatively is, a polydiorganosiloxane gum, i.e., an organopolysiloxane comprising predominately D siloxy units and having a molecular weight sufficiently high enough to not be considered a fluid or liquid at room temperature. In some embodiments, the polydiorganosiloxane gum may exhibit a Williams plasticity number of at least 40 (e.g. as determined by the American Society for Testing and Materials (ASTM) test method 926), such as a plasticity number of from 40 to 200, although other or narrower ranges (e.g. 50 to 150) may be selected based on a particular desired use of the silicone-polyolefin composition. In these or other embodiments, the polydiorganosiloxanes gum has a viscosity of at least 1,000,000 mPa·s at 25 °C. In some embodiments, the polydiorganosiloxane gum comprises a number average molecular weight of at least 600,000 Daltons, such as a molecular weight of 1,000,000 or more, alternatively of, 2,000,000 or more, Daltons.

[00108] The functional organosiloxane of the elastomer component of the curable silicone (D) is typically reaction-curable. In such embodiments, the organosiloxane typically comprises an average of at least two functional groups per molecule, where the functional groups are reactive with one or more other components during a curing process, as described in further detail below. Typically, the particular functionality of such functional organosiloxanes is selected based on the other components of the silicone-polyolefin composition and/or compositions to be prepared therewith. For example, in some embodiments, the functional organosiloxane comprises functional groups complementary to and reactive with the functional groups X of the polysiloxane (A), e.g. to form a bond therebetween (i.e., via additive cross-linking reaction). In specific embodiments, component (D) comprises a functional organosiloxane comprising hydrosilylatable or a condensable functional groups. In some such embodiments, component (D) comprises olefinic unsaturation, e.g. in the form of ethylenically unsaturated groups that may be exploited in a hydrosilylation reaction. In other embodiments, the functional organosiloxane of component (D) comprises cross-linkable functionality different than that of functional groups X and Y of

components (A) and (B), respectively. For example, in some embodiments, functional groups X and Y are addition-reactable, and component (D) comprises a radically-curable organosiloxane.

[00109] In some embodiments, the elastomer component of the curable silicone (D) comprises a functional organosiloxane comprising one or more functional groups reactive with a filler, e.g. directly or at a surface treatment/modification thereof. For example, in some embodiments, component (D) further comprises a silica (e.g. a treated, fumed silica), which may be mutually selected with the functional organosiloxane to influence the reaction-curable nature of the silicone-polyolefin composition and/or curable compositions formed therewith. The elastomer component of the curable silicone (D) may comprise other components in addition to or, with regard to certain components like the silica, in place of those described above. For example, the elastomer component of the curable silicone (D) may comprise one or more fillers or additives, such as those described herein.

[00110] In some embodiments, the elastomer component of the curable silicone (D) may be further defined as a fluorosilicone base. For example, in specific embodiments, the functional organosiloxane of component (D) may comprise a fluoroalkyl silicone, such as a trifluoromethyl silicone.

[00111] In these or other embodiments, component (D) comprises a liquid rubber. Liquid rubbers are known in the art and are typically liquid or flowable at 25 °C. generally, liquid rubbers are liquid or flowable at 25 °C in the absence of any solvents (e.g. organic solvents) or carrier vehicles. By "flowable", it is meant that the liquid rubber is flowable at 25 °C and/or has a viscosity that is measurable at 25 °C. Liquid rubbers are commercially available from various suppliers. Liquid rubbers are curable silicone compositions which, once cured, give silicone rubbers as cured products. Liquid rubbers can be one part, two part, or multi-part compositions depending on their selection and cure mechanism.

[00112] In certain embodiments, the liquid rubber is selected from: (a) hydrosilylation-curable silicone compositions; (b) condensation-curable silicone compositions; (c) thiol-ene reaction-curable silicone compositions; (d) free-radical-curable silicone compositions; and (e) ring-opening reaction-curable silicone compositions. The liquid rubber is not limited to hydrosilylation or addition-curable liquid rubbers. As understood in the art, these liquid rubbers may be cured via different curing conditions, such as exposure to moisture, exposure to heat, exposure to irradiation, etc., but can all cure to five silicone rubbers. Moreover, these liquid rubbers may be curable upon exposure to different types of curing conditions, e.g. both heat and irradiation, which may be utilized together or as only one. In addition, a specific curing condition, e.g. heat, may be utilized to cure or initiate cure of condensation-curable silicone compositions, hydrosilylation-curable silicone compositions, and free radical-curable silicone compositions.

[00113] In certain embodiments, the liquid rubber comprises a hydrosilylation-curable silicone composition. In these embodiments, the hydrosilylation-curable silicone composition typically comprises: an organopolysiloxane having an average of at least two silicon-bonded alkenyl groups or silicon-bonded hydrogen atoms per molecule; an organosilicon compound having an average of at least two silicon-bonded hydrogen atoms or silicon-bonded alkenyl groups per molecule capable of reacting with the silicon-bonded alkenyl groups or silicon-bonded hydrogen atoms in the organopolysiloxane; and a hydrosilylation catalyst. When the organopolysiloxane includes silicon-bonded alkenyl groups, the organosilicon compound includes at least two silicon-bonded hydrogen atoms per molecule, and when the organopolysiloxane includes silicon-bonded hydrogen atoms, the organosilicon compound includes at least two silicon-bonded alkenyl groups per molecule. The organosilicon compound may be referred to as a cross-linker or cross-linking agent.

[00114] The organopolysiloxane and the organosilicon compound may independently be linear, branched, cyclic, or resinous, but are typically independently linear or partly branched. For example, the organopolysiloxane and/or the organosilicon compound may be an organopolysiloxane comprising repeating D units. Such organopolysiloxanes are substantially linear but may include some branching attributable to T and/or Q units. Alternatively, such organopolysiloxanes are linear.

[00115] The silicon-bonded alkenyl groups and silicon-bonded hydrogen atoms of the organopolysiloxane and the organosilicon compound, respectively, may independently be pendent, terminal, or in both positions. Suitable alkenyl groups are described above with regard to the polysiloxane (A).

[00116] The hydrosilylation catalyst can be any of the well-known hydrosilylation catalysts comprising a platinum group metal (i.e., platinum, rhodium, ruthenium, palladium, osmium and iridium) or a compound containing a platinum group metal. Typically, the platinum group metal is platinum, based on its high activity in hydrosilylation reactions. Examples of hydrosilylation catalysts are below.

[00117] The hydrosilylation catalyst may also, or alternatively, be a photoactivatable hydrosilylation catalyst, which may initiate curing via irradiation and/or heat. The photoactivatable hydrosilylation catalyst can be any hydrosilylation catalyst capable of catalyzing the hydrosilylation reaction, particularly upon exposure to radiation having a wavelength of from 150 to 800 nanometers (nm).

[00118] The concentration of the hydrosilylation catalyst is sufficient to catalyze the addition reaction between the organopolysiloxane and the organosilicon compound. In certain embodiments, the concentration of the hydrosilylation catalyst is sufficient to provide typically from 0.1 to 1000 ppm of platinum group metal, alternatively from 0.5 to 100 ppm of platinum

group metal, alternatively from 1 to 25 ppm of platinum group metal, based on the combined weight of the organopolysiloxane and the organosilicon compound.

[00119] The hydrosilylation-curable silicone composition may be a two-part composition where the organopolysiloxane and organosilicon compound are in separate parts. In these embodiments, the hydrosilylation catalyst may be present along with either or both of the organopolysiloxane and organosilicon compound. Alternatively still, the hydrosilylation catalyst may be separate from the organopolysiloxane and organosilicon compound in a third part such that the hydrosilylation reaction-curable silicone composition is a three-part composition.

[00120] Hydrosilylation-curable silicone compositions may be solidified or cured upon exposure to irradiation and/or heat, generally as a function of the hydrosilylation catalyst utilized. One of skill in the art understands how selection of the hydrosilylation catalyst impacts techniques for solidification and curing. In particular, photoactivatable hydrosilylation catalysts are typically utilized when curing via irradiation is desired.

[00121] In certain embodiments, the liquid rubber comprises a condensation-curable silicone composition. In these embodiments, the condensation-curable silicone composition typically comprises an organopolysiloxane having an average of at least two silicon-bonded hydroxyl or hydrolysable groups per molecule; optionally an organosilicon compound having an average of at least two silicon-bonded hydrogen atoms, hydroxyl groups, or hydrolysable groups per molecule; and a condensation catalyst. Although any parameter or condition may be selectively controlled during the inventive method or any individual step thereof, relative humidity and/or moisture content of ambient conditions may be selectively controlled to further impact a cure rate of condensation-curable silicone compositions.

[00122] The organopolysiloxane and the organosilicon compound may independently be linear, branched, cyclic, or resinous, but are typically independently linear or partly branched. For example, the organopolysiloxane and/or the organosilicon compound can be an organopolysiloxane comprising repeating D units. Such organopolysiloxanes are substantially linear but may include some branching attributable to T and/or Q units. Alternatively, such organopolysiloxanes are linear.

[00123] The silicon-bonded hydroxyl groups and silicon-bonded hydrogen atoms, hydroxyl groups, or hydrolysable groups of the organopolysiloxane and the organosilicon compound, respectively, may independently be pendent, terminal, or in both positions.

[00124] As known in the art, silicon-bonded hydroxyl groups result from hydrolyzing silicon-bonded hydrolysable groups. These silicon-bonded hydroxyl groups may condense to form siloxane bonds with water as a byproduct.

[00125] Examples of hydrolysable groups include the following silicon-bonded groups: H, a halide group, an alkoxy group, an alkylamino group, a carboxy group, an alkyliminoxy group, an alkenyloxy group, or an N-alkylamido group. Alkylamino groups may be cyclic amino groups.

[00126] As set forth above, the condensation-curable silicone composition further comprises the organosilicon compound. Specific examples of organosilicon compounds include alkoxy silanes such as $MeSi(OCH_3)_3$, $CH_3Si(OCH_2CH_3)_3$, $CH_3Si(OCH_2CH_2CH_3)_3$, $CH_3Si[O(CH_2)_3CH_3]_3$, $CH_3CH_2Si(OCH_2CH_3)_3$, $C_6H_5Si(OCH_3)_3$, $C_6H_5CH_2Si(OCH_3)_3$, $C_6H_5Si(OCH_2CH_3)_3$, $CH_2=CHSi(OCH_3)_3$, $CH_2=CHCH_2Si(OCH_3)_3$, $CF_3CH_2CH_2Si(OCH_3)_3$, $CH_3Si(OCH_2CH_2OCH_3)_3$, $CF_3CH_2CH_2Si(OCH_2CH_2OCH_3)_3$, $CH_2=CHSi(OCH_2CH_2OCH_3)_3$, $CH_2=CHCH_2Si(OCH_2CH_2OCH_3)_3$, $C_6H_5Si(OCH_2CH_2OCH_3)_3$, $Si(OCH_3)_4$, $Si(OC_2H_5)_4$, and $Si(OC_3H_7)_4$; organoacetoxysilanes such as $CH_3Si(OCOCH_3)_3$, $CH_3CH_2Si(OCOCH_3)_3$, and $CH_2=CHSi(OCOCH_3)_3$; organoiminooxysilanes such as $CH_3Si[O\text{-}N=C(CH_3)CH_2CH_3]_3$, $Si[O\text{-}N=C(CH_3)CH_2CH_3]_4$, and $CH_2=CHSi[O\text{-}N=C(CH_3)CH_2CH_3]_3$; organoacetamidosilanes such as $CH_3Si[NHC(=O)CH_3]_3$ and $C_6H_5Si[NHC(=O)CH_3]_3$; amino silanes such as $CH_3Si[NH(s\text{-}C_4H_9)]_3$ and $CH_3Si(NHC_6H_{11})_3$; and organoaminooxysilanes.

[00127] The organosilicon compound can be a single silane or a mixture of two or more different silanes, each as described above. Also, methods of preparing tri- and tetra-functional silanes are well known in the art; many of these silanes are commercially available.

[00128] When present, the concentration of the organosilicon compound in the condensation-curable silicone composition is sufficient to cure (cross-link) the organopolysiloxane. The particular amount of the organosilicon compound utilized depends on the desired extent of cure, which generally increases as the ratio of the number of moles of silicon-bonded hydrolysable groups in the organosilicon compound to the number of moles of silicon-bonded hydroxy groups in the organopolysiloxane increases. The optimum amount of the organosilicon compound can be readily determined by routine experimentation.

[00129] The condensation catalyst can be any condensation catalyst typically used to promote condensation of silicon-bonded hydroxy (silanol) groups to form Si-O-Si linkages. Examples of condensation catalysts include, but are not limited to, amines; and complexes of lead, tin, zinc, and iron with carboxylic acids. In particular, the condensation catalyst (C^1) can be selected from tin(II) and tin(IV) compounds such as tin dilaurate, tin dioctoate, and tetrabutyl tin; and titanium compounds such as titanium tetrabutoxide.

[00130] When present, the concentration of the condensation catalyst is typically from 0.1 to 10% (w/w), alternatively from 0.5 to 5% (w/w), alternatively from 1 to 3% (w/w), based on the total weight of the organopolysiloxane in the condensation-curable silicone composition.

[00131] When the condensation-curable silicone composition includes the condensation catalyst, the condensation-curable silicone composition is typically a two-part composition where the organopolysiloxane and condensation catalyst are in separate parts. In this embodiment, the organosilicon compound is typically present along with the condensation catalyst. Alternatively still, the condensation-curable silicone composition may be a three-part composition, where the organopolysiloxane, the organosilicon compound and condensation catalyst are in separate parts.

[00132] In certain embodiments, the liquid rubber comprises a free radical-curable silicone composition. In these embodiments, the free radical-curable silicone composition can comprise an organopolysiloxane having an average of at least two silicon-bonded unsaturated groups and an organic peroxide.

[00133] The organopolysiloxane may be linear, branched, cyclic, or resinous, but is typically linear or partly branched. For example, the organopolysiloxane may comprise repeating D units. Such organopolysiloxanes are substantially linear but may include some branching attributable to T and/or Q units. Alternatively, such organopolysiloxanes are linear.

[00134] The silicon-bonded unsaturated groups of the organopolysiloxane may be pendent, terminal, or in both positions. The silicon-bonded unsaturated groups may include ethylenic unsaturation in the form of double bonds and/or triple bonds. Exemplary examples of silicon-bonded unsaturated groups include silicon-bonded alkenyl groups and silicon-bonded alkynyl groups, examples of which are disclosed above with regard to the polysiloxane (A).

[00135] The free radical-curable silicone composition can further comprise an unsaturated compound selected from (i) at least one organosilicon compound having at least one silicon-bonded alkenyl group per molecule, (ii) at least one organic compound having at least one aliphatic carbon-carbon double bond per molecule, (iii) at least one organosilicon compound having at least one silicon-bonded acryloyl group per molecule; (iv) at least one organic compound having at least one acryloyl group per molecule; and (v) mixtures comprising (i), (ii), (iii) and (iv). The unsaturated compound can have a linear, branched, or cyclic structure.

[00136] The organosilicon compound (i) can be an organosilane or an organosiloxane. The organosilane can be a monosilane, disilane, trisilane, or polysilane. Similarly, the organosiloxane can be a disiloxane, trisiloxane, or polysiloxane. Cyclosilanes and cyclosiloxanes typically have from 3 to 12 silicon atoms, alternatively from 3 to 10 silicon atoms, alternatively from 3 to 4 silicon atoms. In acyclic polysilanes and polysiloxanes, the silicon-bonded alkenyl group(s) can be located at terminal, pendant, or at both terminal and pendant positions.

[00137] Specific examples of organosilanes include, but are not limited to, silanes having the following formulae: Vi_4Si, $PhSiVi_3$, $MeSiVi_3$, $PhMeSiVi_2$, Ph_2SiVi_2, and $PhSi(CH_2CH{=}CH_2)_3$, wherein Me is methyl, Ph is phenyl, and Vi is vinyl.

[00138] Specific examples of organosiloxanes include, but are not limited to, siloxanes having the following formulae: $PhSi(OSiMe_2Vi)_3$, $Si(OSiMe_2Vi)_4$, $MeSi(OSiMe_2Vi)_3$, and $Ph_2Si(OSiMe_2Vi)_2$, wherein Me is methyl, Vi is vinyl, and Ph is phenyl.

[00139] The organic compound can be any organic compound containing at least one aliphatic carbon-carbon double bond per molecule, provided the compound does not prevent the organopolysiloxane from curing to form a silicone resin film. The organic compound can be an alkene, a diene, a triene, or a polyene. Further, in acyclic organic compounds, the carbon-carbon double bond(s) can be located at terminal, pendant, or at both terminal and pendant positions.

[00140] The organic compound can contain one or more functional groups other than the aliphatic carbon-carbon double bond. Examples of suitable functional groups include, but are not limited to, -O-, >C=O, -CHO, $-CO_2-$, -C≡N, $-NO_2$, >C=C<, -C≡-, -F, -Cl, -Br, and –I. The suitability of a particular unsaturated organic compound for use in the free-radical curable silicone composition of the present invention can be readily determined by routine experimentation.

[00141] Examples of organic compounds containing aliphatic carbon-carbon double bonds include, but are not limited to, 1,4-divinylbenzene, 1,3-hexadienylbenzene, and 1,2-diethenylcyclobutane.

[00142] The unsaturated compound can be a single unsaturated compound or a mixture comprising two or more different unsaturated compounds, each as described above. For example, the unsaturated compound can be a single organosilane, a mixture of two different organosilanes, a single organosiloxane, a mixture of two different organosiloxanes, a mixture of an organosilane and an organosiloxane, a single organic compound, a mixture of two different organic compounds, a mixture of an organosilane and an organic compound, or a mixture of an organosiloxane and an organic compound.

[00143] The organic peroxide is utilized as a free radical initiator to initiate polymerization of the organopolysiloxane. Examples of organic peroxides include, diaroyl peroxides such as dibenzoyl peroxide, di-p-chlorobenzoyl peroxide, and bis-2,4-dichlorobenzoyl peroxide; dialkyl peroxides such as di-t-butyl peroxide and 2,5-dimethyl-2,5-di-(t-butylperoxy)hexane; diaralkyl peroxides such as dicumyl peroxide; alkyl aralkyl peroxides such as t-butyl cumyl peroxide and 1,4-bis(t-butylperoxyisopropyl)benzene; and alkyl aryl peroxides such as t-butyl perbenzoate, t-butyl peracetate, and t-butyl peroctoate.

[00144] The organic peroxide can be a single peroxide or a mixture comprising two or more different organic peroxides. The concentration of the organic peroxide is typically from 0.1 to 5% (w/w), alternatively from 0.2 to 2% (w/w), based on the weight of the organopolysiloxane.

[00145] The free radical-curable silicone composition may be a two-part composition where the organopolysiloxane and organic peroxide are in separate parts.

[00146] In certain embodiments, the liquid rubber comprises a ring opening reaction-curable silicone composition. In these embodiments, the ring opening reaction-curable silicone composition can comprise an organopolysiloxane having an average of at least two epoxy-substituted groups per molecule and a curing agent. However, the ring opening reaction-curable silicone composition is not limited specifically to epoxy-functional organopolysiloxanes. Other examples of ring opening reaction-curable silicone compositions include those comprising silacyclobutane and/or benzocyclobutene.

[00147] The organopolysiloxane may be linear, branched, cyclic, or resinous, but is typically linear or partly branched.

[00148] The epoxy-substituted groups of the organopolysiloxane may be pendent, terminal, or in both positions. “Epoxy-substituted groups” are generally monovalent organic groups in which an oxygen atom, the epoxy substituent, is directly attached to two adjacent carbon atoms of a carbon chain or ring system. Examples of epoxy-substituted organic groups include, but are not limited to, 2,3-epoxypropyl, 3,4-epoxybutyl, 4,5-epoxypentyl, 2-glycidoxyethyl, 3-glycidoxypropyl, 4-glycidoxybutyl, 2-(3,4-epoxycylohexyl)ethyl, 3-(3,4-epoxycylohexyl)propyl, 2-(3,4-epoxy-3-methylcylohexyl)-2-methylethyl, 2-(2,3-epoxycylopentyl)ethyl, and 3-(2,3 epoxycylopentyl)propyl.

[00149] The curing agent can be any curing agent suitable for curing the organopolysiloxane. Examples of curing agents suitable for that purpose include phenolic compounds, carboxylic acid compounds, acid anhydrides, amine compounds, compounds containing alkoxy groups, compounds containing hydroxyl groups, or mixtures thereof or partial reaction products thereof. More specifically, examples of curing agents include tertiary amine compounds, such as imidazole; quaternary amine compounds; phosphorus compounds, such as phosphine; aluminum compounds, such as organic aluminum compounds; and zirconium compounds, such as organic zirconium compounds. Furthermore, either a curing agent or curing catalyst or a combination of a curing agent and a curing catalyst can be used as the curing agent. The curing agent can also be a photoacid or photoacid generating compound.

[00150] The ratio of the curing agent to the organopolysiloxane is not limited. In certain embodiments, this ratio is from 0.1-500 parts by weight of the curing agent per 100 parts by weight of the organopolysiloxane.

[00151] In certain embodiments, the liquid rubber comprises a thiolene curable silicone composition. In these embodiments, the thiol-ene curable silicone composition typically comprises: an organopolysiloxane having an average of at least two silicon-bonded alkenyl groups or silicon-bonded mercapto-alkyl groups per molecule; an organosilicon compound having an average of at least two silicon-bonded mercapto-alkyl groups or silicon-bonded alkenyl groups per molecule capable of reacting with the silicon-bonded alkenyl groups or silicon-bonded mercapto-alkyl groups in the organopolysiloxane; a catalyst; and, optionally, an organic

compound containing two or more mercapto groups. When the organopolysiloxane includes silicon-bonded alkenyl groups, the organosilicon compound and/or the organic compound include at least two mercapto groups per molecule bonded to the silicon and/or in the organic compound, and when the organopolysiloxane includes silicon-bonded mercapto groups, the organosilicon compound includes at least two silicon-bonded alkenyl groups per molecule. The organosilicon compound and/or the organic compound may be referred to as a cross-linker or cross-linking agent.

[00152] The catalyst can be any catalyst suitable for catalyzing a reaction between the organopolysiloxane and the organosilicon compound and/or the organic compound. Typically, the catalyst is selected from: i) a free radical catalyst; ii) a nucleophilic reagent; and iii) a combination of i) and ii). Suitable free radical catalysts for use as the catalyst include photo-activated free radical catalysts, heat-activated free radical catalysts, room temperature free radical catalysts such as redox catalysts and alkylborane catalysts, and combinations thereof. Suitable nucleophilic reagents for use as the catalyst include amines, phosphines, and combinations thereof.

[00153] Any of the liquid rubbers may optionally and independently further comprise additional ingredients or components, especially if the ingredient or component does not prevent any particular component of the composition from curing. Examples of additional ingredients include, but are not limited to, fillers; inhibitors; adhesion promoters; dyes; pigments; anti-oxidants; carrier vehicles; heat stabilizers; flame retardants; thixotroping agents; flow control additives; fillers, including extending and reinforcing fillers; and cross-linking agents. In various embodiments, the composition further comprises ceramic powder. The amount of ceramic powder can vary and may depend on the 3D printing process being utilized.

[00154] One or more of the additives can be present as any suitable wt.% of the particular composition, such as about 0.1 wt.% to about 15 wt.%, about 0.5 wt.% to about 5 wt.%, or about 0.1 wt.% or less, about 1 wt.%, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, or about 15 wt.% or more of the composition.

[00155] Any of the compositions described above may be a single part or a multi-part composition, as described above with reference to certain compositions. Certain compositions are highly reactive such that multi-part compositions prevent premature mixing and curing of the components. The multi-part composition may be, for example, a two-part system, a three-part system, etc. contingent on the selection of the composition and the components thereof. Any component of the composition may be separate from and individually controlled with respect to the remaining components.

[00156] The liquid rubber can be of various viscosities. In certain embodiments, the liquid rubber has a viscosity less than 500, less than 250, or less than 100, centistokes at 25°C, alternatively

a viscosity of from 1 to 1,000,000 centistokes at 25°C, alternatively from 1 to 100,000 centistokes at 25°C, alternatively from 1 to 10,000 centistokes at 25°C. As readily understood in the art, kinematic viscosity may be measured in accordance with ASTM D-445 (2011), entitled "Standard Test Method for Kinematic Viscosity of Transparent and Opaque Liquids (and Calculation of Dynamic Viscosity)."

[00157] The amount of components (A), (B), (C), and (D) in the silicone-polyolefin composition may vary, as will be best understood in view of the additional description below.

[00158] Typically, the polysiloxane (A) is present in the silicone-polyolefin composition in an amount of from 0.5 to 15 wt.%, such as from 1 to 12, alternatively from 1.5 to 10, alternatively from 2 to 7 wt.%, based on the total weight of the silicone-polyolefin composition. The functionalized polyolefin (B) is typically present in the silicone-polyolefin composition in an amount of from 1 to 40 wt.%, alternatively from 1 to 30 wt.%, based on the total weight of the silicone-polyolefin composition. The polyolefin (C) is typically present in the silicone-polyolefin composition in an amount of from 1 to 40, alternatively from 1 to 35, alternatively from 2 to 30, alternatively from 3 to 25, alternatively from 4 to 20, alternatively from 5 to 25, wt.% based on the total weight of the silicone-polyolefin composition. Component (D) is typically present in the silicone-polyolefin composition an amount of from 55 to 99, alternatively from 60 to 99, alternatively from 70 to 98, wt.%, based on the total weight of the silicone-polyolefin composition.

[00159] In some embodiments, the amount of component (A), (B), (C), and/or (D) corresponds to the ranges above, where the wt.% is determined on a basis of the combined weight of components (A), (B), (C), and (D) (i.e., rather than the silicone-polyolefin composition as a whole, should additional components be present).

[00160] With regard to the preceding embodiments and component amounts, it is to be understood that the balance of the silicone-polyolefin composition, if any, may comprise one or more additional components of the silicone-polyolefin composition. For example, as will be better understood in view of the additional description and methods described herein, the silicone-polyolefin composition may comprise a catalyst, a solvent or carrier vehicle, or a reaction promotor. In some embodiments, however, the silicone-polyolefin composition is free from, alternatively substantially free from, a reaction catalyst or promotor. In these or other embodiments, the silicone-polyolefin composition comprises less than 1 wt.% solvent, based on the total weight of the silicone-polyolefin composition. In other embodiments, the silicone-polyolefin composition is substantially free from, alternatively is free from, solvents or carrier vehicles.

[00161] It will also be appreciated that amounts outside of the ranges and ratios above may also be utilized. However, as will also be better understood in view of the additional description and methods described herein, decreased loadings of the polysiloxane (A) and/or increased loadings

of the functionalized polyolefin (B) may result in macrophase separation due to poor compatibilization.

[00162] In some embodiments, the silicone-polyolefin composition comprises a (E) catalyst adapted to facilitate a coupling reaction between the functional groups X of the polysiloxane (A) and functional groups Y of the functionalized polyolefin (B).

[00163] The use of the catalyst (E), as well as the particular type or specific compound(s) selected for use in or as the catalyst (E), will be selected by those of skill in the art based on the particular polysiloxane (A) and functionalized polyolefin (B) selected. More specifically, the catalyst (E) is not particularly limited, but is instead selected to catalyze coupling of components (A) and (B), and may thus comprise or be any compound suitable for facilitating the reaction of the polysiloxane (A) and the functionalized polyolefin (B) (e.g. via reaction of/including functional groups X and functional groups Y), as will be understood by one of skill in the art in view of the description herein. For example, in certain embodiments, the catalyst (E) is selected from those facilitating reactions including hydrosilylation, condensation, displacement, ring-opening, nucleophilic substitution, and the like, as well as combinations of such reactions. It is to be appreciated that the catalyst (E) may itself comprise more than one type of catalyst and/or the reaction may utilize more than one type of catalyst (E), such as two, three, or more different catalysts (E).

[00164] In specific embodiments, the catalyst (E) comprises, alternatively is, a hydrosilylation catalyst. In such embodiments, functional groups X and functional groups Y are complementary coupleable hydrosilylatable groups.

[00165] Hydrosilylation catalysts suitable for use in the silicone-polyolefin composition are not particularly limited and may be any known catalyst for catalyzing hydrosilylation reactions. Combinations of different hydrosilylation catalysts may also be utilized.

[00166] In certain embodiments, the hydrosilylation catalyst comprises a Group VIII to Group XI transition metal. Group VIII to Group XI transition metals refer to the modern IUPAC nomenclature. Group VIII transition metals are iron (Fe), ruthenium (Ru), osmium (Os), and hassium (Hs); Group IX transition metals are cobalt (Co), rhodium (Rh), and iridium (Ir); Group X transition metals are nickel (Ni), palladium (Pd), and platinum (Pt); and Group XI transition metals are copper (Cu), silver (Ag), and gold (Au). Additional examples of catalysts suitable for the hydrosilylation catalyst include rhenium (Re), molybdenum (Mo), Group IV transition metals (i.e., titanium (Ti), zirconium (Zr), and/or hafnium (Hf)), lanthanides, actinides, and Group I and II metal complexes (e.g. those comprising calcium (Ca), potassium (K), strontium (Sr), etc.). Combinations, complexes (e.g. organometallic complexes), and other forms of such metals may also be utilized as the hydrosilylation catalyst.

[00167] The hydrosilylation catalyst may be in any suitable form. For example, the hydrosilylation catalyst may be a solid, or instead by may be disposed in or on a solid carrier. Examples of solid catalysts typically include platinum-based catalysts, palladium-based catalysts, and similar noble metal-based catalysts, and also nickel-based catalysts. Specific examples thereof include elemental nickel, palladium, platinum, rhodium, cobalt, and similar metals, and also elemental mixtures/combinations such as platinum-palladium, nickel-copper-chromium, nickel-copper-zinc, nickel-tungsten, nickel-molybdenum, and the like, as well as Cu-Cr, Cu-Zn, Cu-Si, Cu-Fe-Al, Cu-Zn-Ti, and similar copper-containing catalysts. Examples of carriers include activated carbons, silicas, silica aluminas, aluminas, zeolites and other inorganic powders/particles (e.g. sodium sulphate), and the like. The hydrosilylation catalyst may also be disposed in a vehicle, e.g. a solvent which solubilizes the hydrosilylation catalyst, alternatively a vehicle which merely carries, but does not solubilize, the hydrosilylation catalyst. Such vehicles are known in the art.

[00168] In specific embodiments, the hydrosilylation catalyst comprises platinum. In these embodiments, the hydrosilylation catalyst is exemplified by platinum black, chloroplatinic acids (e.g. chloroplatinic acid hexahydrate, reaction products of chloroplatinic acid and a monohydric alcohol, or aliphatically-unsaturated organosilicon compound such as divinyltetramethyldisiloxane, etc.), platinum bis(ethylacetoacetate), platinum bis(acetylacetonate), platinum chloride, complexes of any such compounds with olefins or organopolysiloxanes, and microencapsulated platinum compounds (e.g. matrix or core-shell type). For example, suitable complexes of platinum with organopolysiloxanes, such as 1,3-diethenyl-1,1,3,3-tetramethyldisiloxane complexes with platinum, may be used directly or, alternatively, in microencapsulated form (e.g. in a resin matrix).

[00169] The hydrosilylation catalyst may be a photoactivatable hydrosilylation catalyst, which may initiate curing via irradiation (e.g. upon exposure to radiation having a wavelength of from 150 to 800 nm) and/or heat.

[00170] Specific examples of photoactivatable hydrosilylation-reaction catalysts suitable for the catalyst (E) include platinum(II) β-diketonate complexes such as platinum(II) bis(2,4-pentanedioate), platinum(II) bis(2,4-hexanedioate), platinum(II) bis(2,4-heptanedioate), platinum(II) bis(1-phenyl-1,3-butanedioate, platinum(II) bis(1,3-diphenyl-1,3-propanedioate), platinum(II) bis(1,1,1,5,5,5-hexafluoro-2,4-pentanedioate); (η-cyclopentadienyl)trialkylplatinum complexes, such as (Cp)trimethylplatinum, (Cp)ethyldimethylplatinum, (Cp)triethylplatinum, (chloro-Cp)trimethylplatinum, and (trimethylsilyl-Cp)trimethylplatinum, where Cp represents cyclopentadienyl; triazene oxide-transition metal complexes, such as $Pt[C_6H_5NNNOCH_3]_4$, $Pt[p\text{-}CN\text{-}C_6H_4NNNOC_6H_{11}]_4$, $Pt[p\text{-}H_3COC_6H_4NNNOC_6H_{11}]_4$, $Pt[p\text{-}CH_3(CH_2)_x\text{-}$

$C_6H_4NNNOCH_3]_4$, 1,5-cyclooctadiene.Pt[p-CN-$C_6H_4NNNOC_6H_{11}]_2$, 1,5-cyclooctadiene.Pt[p-CH_3O-$C_6H_4NNNOCH_3]_2$, $[(C_6H_5)_3P]_3Rh$[p-CN-$C_6H_4NNNOC_6H_{11}$], and Pd[p-$CH_3(CH_2)_x$—$C_6H_4NNNOCH_3]_2$, where x is 1, 3, 5, 11, or 17; (η-diolefin)(σ-aryl)platinum complexes, such as (η^4-1,5-cyclooctadienyl)diphenylplatinum, η^4-1,3,5,7-cyclooctatetraenyl)diphenylplatinum, (η^4-2,5-norboradienyl)diphenylplatinum, (η^4-1,5-cyclooctadienyl)bis-(4-dimethylaminophenyl)platinum, (η^4-1,5-cyclooctadienyl)bis-(4-acetylphenyl)platinum, and (η^4-1,5-cyclooctadienyl)bis-(4-trifluormethylphenyl)platinum. In specific embodiments, the photoactivatable hydrosilylation-reaction catalyst of the catalyst (E) is a Pt(II) β-diketonate complex, such as platinum(II) bis(2,4-pentanedioate).

[00171] It will be appreciated that the compounds described above with regard to the catalyst (E) above, i.e., when comprising or being the hydrosilylation catalyst, may generally promote rapid reaction (i.e., coupling, cross-linking, etc.) of components (A) and (B) of the silicone-polyolefin composition (i.e., when reactable via hydrosilylation, as described above), even at room temperature. As such, in certain embodiments, the silicone-polyolefin composition further comprises a reaction inhibitor, which may be further defined as a cross-linking reaction inhibitor, a hydrosilylation reaction inhibitor, or a stabilizer. Suitable inhibitors are known in the art, and generally include compounds that stop catalysis, yet are volatile or readily decomposed by heat or light (e.g. UV). Examples of such inhibitors typically include relatively low-boiling alkyne- and alkene- based compounds with electron-withdrawing groups, which complex with catalytic metals and thereby block activity thereof until heat is applied. General examples of inhibitors include acetylenic alcohols having boiling points of less than 250 °C (e.g. 2-methyl-3-butyn-2-ol, 1-ethynylcyclohexanol (ETCH)), as well as various fumarates, maleates (e.g. diallyl maleate), small vinyl-functional siloxanes (e.g. tetravinyl-tetramethyltetrasiloxane), ethynylalkenes (e.g. 3-methyl-3-penten-1-yne, 3-methyl-3-hexen-1-yne, 3,5-dimethyl-3-hexen-1-yne, 3-ethyl-3-buten-1-yne, 3-phenyl-3-buten-1-yne, etc.), dialkyl esters of acetylenedicarboxylic acids (e.g. dimethyl acetylenedicarboxylate (DMAD)), and the like, as well as various derivatives thereof. One of skill in the art will selected such an inhibitor for use in or as the reaction inhibitor in view of the particular other components utilized for the silicone-polyolefin composition, as well as the intended use thereof, including the uses and processes disclosed herein. For example, it is well understood that particular hydrosilylation reaction inhibitors/stabilizers exhibit different compatibilities and operating windows (e.g. in terms of light and/or heat sensitivity, require loading amounts, etc.), such that a person of skill in the art will readily select an appropriate inhibitor for use as the reaction inhibitor, if needed, based on the description of the compositions and methods provided herein.

[00172] In specific embodiments, the catalyst (E) comprises, alternatively is, a condensation catalyst. In such embodiments, functional groups X and functional groups Y are complementary condensable groups (e.g. amine (X) + anhydride (Y)).

[00173] Condensation catalysts suitable for use in the silicone-polyolefin composition may be any known compound (or combination) compatible with components (A) and (B) and capable of catalyzing or otherwise facilitating a condensation reaction of functional groups X and Y. Combinations of different hydrosilylation catalysts may also be utilized.

[00174] Examples of condensation catalysts generally inorganic and organic bases and acids (i.e., an acid-type or base-type catalyst), which may comprise metal atoms or, alternatively, may be substantially free from, alternatively free from metal atoms. Examples of such catalysts generally include mineral acids and bases (e.g. H_2SO_4, LiOH, NaOH, KOH, CsOH, etc.), organic bases and amines (e.g. tetramethylammonium hydroxide ($(CH_3)_4NOH$), 1,8-diazabicyclo [5.4.0]undec-7-ene (DBU)), organic and inorganic acids (e.g. carboxylic acids, sulfamic acids, etc.), and particular metal complexes such as titanium alkoxides (e.g. $Ti(OiPr)_2(acac)_2$), and the like, as well as derivatives, modifications, and combinations thereof. Such condensation catalysts are well known in the art and commercially available.

[00175] When utilized, particular condensation catalysts will be selected by those of skill in the art in view of the particular components and conditions utilized in the silicone-polyolefin composition, and processes involving the same. Particular limitations will be understood in view of the description below. For example, in some embodiments, residual amounts of catalyst may be carried forward from the silicone-polyolefin composition to other compositions and/or processes that may be incompatible with certain types of condensation catalysts. As such, in certain embodiments, the catalyst (E), optionally the silicone-polyolefin composition as a whole, is substantially free from, alternatively free from, one or more metal-based condensation catalysts, strong acids and bases, oxidizing compounds, and the like.

[00176] It is to be appreciated that other catalysts may also be utilized, regardless of falling under one of the general descriptions herein. For example, in will be understood that certain coupling reactions described herein may be facilitated by various Lewis acid catalysts, such as those based on boron, aluminum, iron, tin, and titanium, such as tri- or tetravalent halides and alkoxides thereof. Likewise, in certain embodiments, the catalyst (E) comprises, alternatively is, a Piers-Rubinsztajn-type catalysts (e.g. tris(pentafluorophenyl)borane), which may be utilized to facilitate the coupling between one of the hydridosilyl groups (e.g. functional group X) and an alkoxy- or hydroxy-silyl groups (e.g. functional groups Y) described herein.

[00177] The catalyst (E) may be utilized in any amount, which will be selected by one of skill in the art, e.g. dependent upon the particular catalyst (E) selected (e.g. the concentration/amount of active components thereof, the type of catalyst being utilized, the type of coupling reaction

being performed, etc.), the scale being utilized (e.g. total amounts of components (A) and (B), relative amounts of functional groups X and functional groups Y, etc.), etc. As will be best understood in view of the description below, the molar ratio of the catalyst (E) to components (A) and/or (B) utilized in the reaction may influence the rate and/or amount of coupling/crosslinking, when desired. Thus, the amount of the catalyst (E) as compared to components (A) and/or (B), as well as the molar ratios therebetween, may vary. Typically, these relative amounts and the molar ratio are selected to maximize the reaction of components (A) and (B) (e.g. via functional groups X an Y) while minimizing the loading of the catalyst (E) (e.g. for increased economic efficiency of the reaction, increased ease of purification of the reaction product formed, etc.).

[00178] For example, it will be appreciated that, beyond a minimum threshold value, decreasing catalyst loadings may result in poor reactivity, and thus may not be able to achieve the properties and effects demonstrated and described herein. With regard to the present embodiments, such a minimum threshold is generally applicable to the hydrosilylation-mediated coupling reaction of components (A) and (B) described herein. As such, in typical embodiments where the functional groups X and functional groups Y are complementary coupleable hydrosilylatable groups, the silicone-polyolefin composition comprises the hydrosilylation catalyst of catalyst (E) in an amount of from 1 to 100 ppm, alternatively from 5 to 100 ppm. However, it will also be appreciated that loadings outside these ranges may be utilized to realize the benefits described herein, and one of skill in the art will be able to determine proper catalyst loadings (e.g. based on reactivity rates, compatibilization of components (A) and (B), etc.) in view of the description and examples herein. For example, such minimum threshold amounts of catalyst (E) may not be needed in other embodiments, such as where the epoxy-amine coupling reaction of components (A) and (B) is utilized.

[00179] Likewise, in yet other embodiments, the silicone-polyolefin composition is substantially free, alternatively is free from a reaction catalyst or promotor, including those described above with regard to the catalyst (E). As will be understood in view of the additional description herein, certain features of the silicone-polyolefin composition and related methods and compositions may be realized and/or achieved without the use of a catalyst or promotor. For example, in specific embodiments, the functional groups X and functional groups Y are complementary condensable groups (e.g. amine (X) + anhydride (Y)), and the silicone-polyolefin composition is substantially free, alternatively is free from a condensation catalyst or promotor. Other cross-linking chemistries may also be used absent a discreet catalyst loading, such as the amine-methacrylate, silanol-alkoxysilane, and carbinol-anhydride couplings provided herein.

[00180] In certain embodiments, the catalyst (E) is utilized in the silicone-polyolefin composition in an amount of from 0.000001 to 5 wt.%, based on the total amount of component (A) utilized (i.e., wt./wt.). For example, the catalyst (E) may be used in an amount of from 0.00001 to 5 wt.%,

such as from 0.00001 to 4, alternatively from 0.00001 to 3, alternatively from 0.00001 to 2, alternatively from 0.0001 to 2, alternatively from 0.0001 to1, alternatively from 0.0001 to 0.5, alternatively from 0.001 to 0.5, alternatively of from 0.005 to 0.5, wt.%, based on the total amount of component (A) utilized. Likewise, or alternatively, the catalyst (E) may be utilized in the silicone-polyolefin composition in an amount of from 0.000001 to 5 wt.%, based on the total amount of component (B) utilized (i.e., wt./wt.). For example, the catalyst (E) may be used in an amount of from 0.00001 to 5 wt.%, such as from 0.00001 to 4, alternatively from 0.00001 to 3, alternatively from 0.00001 to 2, alternatively from 0.0001 to 2, alternatively from 0.0001 to1, alternatively from 0.0001 to 0.5, alternatively from 0.001 to 0.5, alternatively of from 0.005 to 0.5, wt.%, based on the total amount of component (B) utilized.

[00181] In some embodiments (e.g. when the type of reaction dictates a stoichiometric loading), the amount of the catalyst (E) utilized may be selected and/or determined on a molar ratio based on one or more components of the silicone-polyolefin composition, as will be understood by those of skill in the art. In such embodiments, the catalyst (E) may be utilized in the silicone-polyolefin composition in an amount of from 0.0001 to 5 mol %, based on the total amount of component (A) and/or component (B) utilized. For example, the catalyst (E) may be used in an amount of from 0.0005 to 5, alternatively of from 0.0005 to 3, alternatively of from 0.0005 to 1, alternatively of from 0.001 to 1 mol %, based on the total amount of component (A) utilized.

[00182] The functionalized polyolefin (B) and the polyolefin (C) are typically immiscible, or substantially immiscible, with the curable silicone elastomer component (D). As such, as introduced above, a method of preparing a hybrid silicone-polyolefin blend from the silicone-polyolefin composition (the “preparation method”) is also provided, and generally comprises compatibilizing the functionalized polyolefin (B) and the polyolefin (C) with the polysiloxane (A) within the silicone-polyolefin composition.

[00183] More specifically, the preparation method allows for the preparation of a uniform dispersion of the functionalized polyolefin (B) and the polyolefin (C) in component (D) (i.e., as a polyolefin-in-silicone dispersion), which components are based on otherwise non-compatible chemistries (i.e., polysiloxanes and polyolefins). Moreover, the preparation method provides the silicone-polyolefin blend as a uniform composition having small domains (e.g. polyolefin domains) and a useful characteristics supporting application of the silicone-polyolefin blend to curable compositions.

[00184] The polysiloxane (A) is capable of compatibilizing the functionalized polyolefin (B), e.g. via additive coupling/cross-linking reaction between the functional groups X and functional groups Y, respectively. As such, the preparation method comprises reacting the polysiloxane (A) and the functionalized polyolefin (B). Typically, components (A) and (B) are reacted in the presence of components (C) and (D). As such, it will be understood that the preparation method may be

carried out on the silicone-polyolefin composition in a pre-made form, or may include preparing the silicone-polyolefin composition (e.g. by combining the polysiloxane (A), the functionalized polyolefin (B), the polyolefin (C), and component (D), optionally with any of the additional and/or optional components described here, such as the catalyst (E)). In this fashion, the silicone-polyolefin composition may be referred to or characterized as a reactive pre-mix, or a compatibilization mixture, comprising at least components (A), (B), (C), and (D) or the silicone-polyolefin composition.

[00185] With regard to the method components, the polysiloxane (A), functionalized polyolefin (B), the polyolefin (C), and the curable silicone (D) may each be prepared (i.e., as part of the preparation method) or otherwise obtained, e.g. as prepared reagents/feedstocks. Methods of preparing each of these components (e.g. reactable/cross-linkable polydiorganosiloxanes, functionalized poly-alpha olefin-alkenyl copolymers, reaction-curable polydiorganosiloxane gums, etc.) are known in the art, with suitable precursors and starting materials commercially available from various suppliers.

[00186] Components (A), (B), (C), and (D) may be utilized in any amount, bearing in mind the upper limit/ratio of the functionalized polyolefin (B) with regard to compatibilization by the polysiloxane (A) described above. As such, suitable amounts which will be selected by one of skill in the art, e.g. dependent upon the particular components selected, the reaction parameters employed for the compatibilization, the scale of the compatibilization (e.g. total amounts of components (A), (B), (C), and (D) in the silicone-polyolefin composition being utilized, etc.

[00187] In general, components ((A), (B), (C), and (D) may independently be utilized in any form, such as neat (i.e., absent solvents, carrier vehicles, diluents, etc.), or disposed in a carrier vehicle, such as a solvent or dispersant, as described in further detail below. However, in certain embodiments, each of (A), (B), (C), and (D) is utilized neat, or otherwise in a form substantially free from, alternatively free from a carrier vehicle. When substantially free from carrier vehicles, components ((A), (B), (C), and (D) will typically be free from (or at substantially free from), water and carrier vehicles/volatiles reactive with any components to participate in the compatibilization, or post-compatibilization reaction, as described below. As such, in some embodiments, the compatibilization, or the preparation method as a whole, is carried out in the absence of carrier vehicles/volatiles that are reactive with the polysiloxane (A) (i.e., at the polysiloxane backbone or the functional groups X), the functionalized polyolefin (B) (e.g. at the functional groups Y, or any one or more other components being utilized to prepare the silicone-polyolefin composition (e.g. the catalyst (E), if present). For example, in certain embodiments, the preparation method may comprise stripping a mixture comprising one or more of the components (A), (B), (C), and (D), and optionally (E) (i.e., the “compatibilization components”) of volatiles, solvents, etc., e.g., prior to combining the same with any one or other components being utilized in the preparation

method. Techniques for stripping such siloxanes, polyolefins, and reaction catalysts are generally known in the art, and may include heating, drying, applying reduced pressure/vacuum, azeotroping with solvents, utilizing drying agents such as molecular sieves, etc., and combinations thereof, with care take to not initiate premature reaction with the functional groups X or functional groups Y.

[00188] In some embodiments, one or more of the compatibilization components may be combined with a carrier vehicle, e.g. prior to and/or during the compatibilization of component (B). When combined prior to the compatibilization, e.g. as part of the preparation of such component, or to otherwise facilitate providing and/or metering out a component to the compatibilization mixture, the stripping process above may be carried out. When the carrier vehicle is to be present during the compatibilization, it will be appreciated that suitable choices will be limited based on compatible with the components and conditions of the compatibilization process.

[00189] With regard to carrier vehicles generally, examples typically include oils (e.g. organic oils and/or a silicone oils), fluids, solvents, etc., and combinations thereof. For example, in some embodiments, one or more of the components of the compatibilization disposed in a carrier fluid prior to being combined with the other components of the compatibilization. In some such embodiments, the carrier fluid comprises, alternatively consists essentially of, a silicone fluid. The silicone fluid is typically a low viscosity and/or volatile siloxane. In some embodiments, the silicone fluid is a low viscosity organopolysiloxane, a volatile methyl siloxane, a volatile ethyl siloxane, a volatile methyl ethyl siloxane, or the like, or combinations thereof. Typically, the silicone fluid has a viscosity at 25 °C in the range of 1 to 1,000 mPa-s. Specific examples of suitable silicone fluids include hexamethylcyclotrisiloxane, octamethylcyclotetrasiloxane, decamethylcyclopentasiloxane, dodecamethylcyclohexasiloxane, octamethyltrisiloxane, decamethyltetrasiloxane, dodecamethylpentasiloxane, tetradecamethylhexasiloxane, hexadeamethylheptasiloxane, heptamethyl-3-{(trimethylsilyl)oxy)}trisiloxane, hexamethyl-3,3, bis{(trimethylsilyl)oxy}trisiloxane pentamethyl{(trimethylsilyl)oxy}cyclotrisiloxane as well as polydimethylsiloxanes, polyethylsiloxanes, polymethylethylsiloxanes, polymethylphenylsiloxanes, polydiphenylsiloxanes, caprylyl methicone, hexamethyldisiloxane, heptamethyloctyltrisiloxane, hexyltrimethicone, and the like, as well as derivatives, modifications, and combinations thereof. Additional examples of suitable silicone fluids include polyorganosiloxanes with suitable vapor pressures, such as from 5×10^{-7} to 1.5×10^{-6} m^2/s, such as DOWSIL™ 200 Fluids and DOWSIL™ OS FLUIDS, which are commercially available from Dow Silicones Corporation of Midland, Mich., U.S.A.

[00190] In other such embodiments, the carrier fluid comprises, alternatively consists essentially of, an organic fluid, which typically comprises an organic oil including a volatile and/or semi-

volatile hydrocarbon, ester, and/or ether. General examples of such organic fluids include volatile hydrocarbon oils, such as C_6-C_{16} alkanes, C_8-C_{16} isoalkanes (e.g. isodecane, isododecane, isohexadecane, etc.) C_8-C_{16} branched esters (e.g. isohexyl neopentanoate, isodecyl neopentanoate, etc.), and the like, as well as derivatives, modifications, and combinations thereof. Additional examples of suitable organic fluids include aromatic hydrocarbons and aliphatic hydrocarbons. Hydrocarbons include isododecane, isohexadecane, Isopar L (C_{11}-C_{13}), Isopar H (C_{11}-C_{12}), hydrogentated polydecene. Ethers and esters include isodecyl neopentanoate, neopentylglycol heptanoate, glycol distearate, dicaprylyl carbonate, diethylhexyl carbonate, propylene glycol n-butyl ether, ethyl-3 ethoxypropionate, propylene glycol methyl ether acetate, tridecyl neopentanoate, propylene glycol methylether acetate (PGMEA), propylene glycol methylether (PGME), octyldodecyl neopentanoate, diisobutyl adipate, diisopropyl adipate, propylene glycol dicaprylate/dicaprate, octyl ether, octyl palmitate, and combinations thereof.

[00191] In yet other such embodiments, the carrier fluid comprises, alternatively consists essentially of, an organic solvent. Examples of the organic solvents include those comprising a ketone, such as acetone, methylethyl ketone, and methyl isobutyl ketone; an aromatic hydrocarbon, such as benzene, toluene, and xylene; an aliphatic hydrocarbon, such as heptane, hexane, and octane; a glycol ether, such as propylene glycol methyl ether, dipropylene glycol methyl ether, propylene glycol n-butyl ether, propylene glycol n-propyl ether, and ethylene glycol n-butyl ether; a halogenated hydrocarbon, such as dichloromethane, 1,1,1-trichloroethane and methylene chloride; chloroform; dimethyl sulfoxide; dimethyl formamide, acetonitrile; tetrahydrofuran; white spirits; mineral spirits; naphtha; n-methylpyrrolidone; and the like, as well as derivatives, modifications, and combination thereof.

[00192] As introduced above, and notwithstanding the preceding section, the compatibilization reaction (i.e., the reaction of components (A) and (B) in the presence of components (C) and (D)) may be carried out under conditions substantially free from carrier vehicle or solvent. Indeed, as will be appreciated from the examples and additional description herein, certain features of the preparation method enable solventless preparation of the compatibilized blends of the silicone components (e.g. components (A) and (D)) and the functionalized polyolefin (B) and polyolefin (C).

[00193] As also introduced above, the compatibilization mixture may comprise components other than components (A) and (B). For example, in certain embodiments, the reaction (i.e., the compatibilization) is further defined as the hydrosilylation reaction, where the functional groups (X) and functional groups (Y) are selected from the complementary hydrosilylatable groups (e.g. where each X is the silicon-bonded olefinically-ethylenically unsaturated group, and each Y comprises the hydridosilyl group). In such embodiments, the compatibilization mixture will

typically include the hydrosilylation catalyst (E). In other embodiments, however, the compatibilization is further defined as the condensation reaction, where the functional groups (X) and functional groups (Y) are selected from the complementary condensable groups (e.g. where each X is the aminoalkyl group and each Y comprises the anhydride group). In some such embodiments, the compatibilization is free from catalyst (e.g. condensation catalyst (E)).

[00194] The components of the silicone-polyolefin composition are typically combined in a vessel or reactor to carry out the compatibilization and disperse components (B) and (C) in component (D), and thereby prepare the silicone-polyolefin blend. The compatibilization components may be fed together or separately to the vessel, or may be disposed in the vessel in any order of addition, and in any combination, to prepare the compatibilization mixture (i.e., the silicone-polyolefin composition). Likewise, the compatibilization mixture may be prepared in batch, semi-batch, semi-continuous, or continuous processes, unless otherwise noted herein.

[00195] Typically, the silicone-polyolefin blend is prepared via dynamic reaction/crosslinking of the silicone-polyolefin blend i.e., via mixing together the components (A), (B), (D), and optionally (E), as the coupling/crosslinking is carried out. In this sense, the term "dynamic" indicates the silicone-polyolefin composition is subjected to shear forces during the crosslinking/ compatibilization step, in contrast to "static crosslinking" whereby a polymer is relatively immobile during such crosslinking.

[00196] The compatibilization is typically conducted at an elevated temperature with mixing (e.g. under shear). As such, the vessel or reactor is typically heated, e.g. via a jacket, mantle, exchanger, bath, coils, etc., and equipped with mixing means for blending and/or shearing the compatibilization mixture. In general, the elevated temperature for compatibilization is from 100 to 200 °C. In specific embodiments, the elevated temperature is from 100 to 180, alternatively from 100 to 170, alternatively from 100 to 160, alternatively from 100 to 150, alternatively from 110 to 150, alternatively from 110 to 140, °C.

[00197] With regard to the mixing/shear, the silicone-polyolefin composition is typically homogenized via melt blending or extruding the silicone-polyolefin composition at the elevated temperature to prepare the silicone-polyolefin blend. As such, while other reactors and mixing/blending techniques (e.g. using twin-rotor mixers, ribbon blenders, solution blenders, co-kneaders, screw extruders, static mixers, Banbury-type mixers, etc.) may be utilized, in certain embodiments the reactor/vessel is an extruder or melt blender. However, one of skill in the art will understand that other mixers may also be utilized. The process of mixing is also a function of the selection of component (D). For example, when component (D) is the liquid rubber, mixing may include any method typically utilized with liquids or viscous materials.

[00198] As the compatibilization progresses and the components are mixed and reacted, the functionalized polyolefin (B) and polyolefin (C) are taken into and dispersed uniformly throughout

component (D), thereby preparing the silicone-polyolefin blend as a polyolefin-in-silicone dispersion.

[00199] As introduced above, the silicone-polyolefin blend comprises the functionalized polyolefin (B) and polyolefin (C) dispersed in a combination of the polysiloxane (A) and the curable silicone elastomer component (D). The silicone-polyolefin blend presents as a thick but spreadable mixture comprising the discontinuous phase comprising the functionalized polyolefin (B) and the polyolefin (C), uniformly dispersed in the continuous phase comprising component (D) and polysiloxane (A). Said differently, the silicone-polyolefin blend comprises domains of the functionalized polyolefin (B) and polyolefin (C) (i.e., "polyolefin domains") dispersed in the silicone matrix presented by component (D). In certain embodiments, the polyolefin domains are uniformly dispersed through the continuous phase comprising component (D).

[00200] A curable composition is also provided herein. In the same nature as the silicone-polyolefin compositions and blend described above, the curable composition does not require a carrier vehicle or solvent under typical circumstances. As such, the curable composition may be solvent-free (i.e., free from, alternatively substantially free from, a carrier vehicle or solvent).

[00201] The curable composition comprises the silicone-polyolefin blend and a curing agent, although other components may also be utilized. The curing agent typically comprises, alternatively is, a free radical initiator and/or a photoinitiator.

[00202] Examples of suitable free radical initiators typically include benzoyl peroxide, tert-butyl peroxide, dicumyl peroxide, lauroyl peroxide, peracetic acid, cyclohexanone peroxide, cumene hydroperoxide, tert-butyl peroxide, tert-butyl hydroperoxide, 2,2'-azobisisobutyronitril (AIBN), 2,2'-azodi(2-methylbutyronitrile) (AMBN), tert-amyl peroxybenzoate, tert-butyl peracetate, tert-butyl peroxybenzoate, tert-butylperoxy isopropyl carbonate, cumene hydroperoxide, and potassium persulfate. Any amount of the free radical initiator may be used in the curable composition, although only catalytic amounts are typically necessary. For examples, in certain embodiments, the curable composition comprises the free radical initiator in an amount of from 0.01 to 10 wt.%, based on the total weight of the curable composition. In some embodiments, the free radical initiator is employed in an amount of at least 0.15, alternatively at least 0.2 wt.%, while at the same time 10, alternatively 7, alternatively 5, alternatively 3 wt.% or less, preferably 5 wt.% or less and can be 3 wt.% or less, based on the total weight of the curable composition. Amounts outside of these ranges may also be utilized, recognizing that excess free radical initiator may not significantly increase the time or efficiency of the curing process, which is typically complete in less than 30, alternatively less than 20, alternatively less than 15, alternatively less than 15 minutes, while at the same time typically greater than 1, alternatively greater than 5 minutes.

[00203] Examples of suitable photoinitiators include onium salts, nitrobenzyl sulfonate esters, diaryliodonium salts of sulfonic acids, triarylsulfonium salts of sulfonic acids, diaryliodonium salts of boronic acids, triarylsulfonium salts of boronic acids, bis-diaryl iodonium salts (such as bis(dodecyl phenyl) iodonium hexafluoroarsenate and bis(dodecylphenyl) iodonium hexafluoroantimonate), dialkylphenyl iodonium hexafluoroantimonate, diaryliodonium salts of sulfonic acids, triarylsulfonium salts of sulfonic acids, diaryliodonium salts of boronic acids, and triarylsulfonium salts of boronic acids.

[00204] Examples of suitable diaryioadonium salts of sulfonic acid include diaryliodonium salts of perfluoroalkylsulfonic acids and diaryliodonium salts of aryl sulfonic acids. Examples of suitable diaryliodonium salts of perfluoroalkylsulfonic acids include diaryliodonium salts of perfluorobutanesulfonic acid, diaryliodonium salts of perfluoroethanesulfonic acid, diaryliodonium salts of perfluoro-octanesulfonic acid, and diaryliodonium salts of trifluoromethane sulfonic acid. Examples of suitable diaryliodonium salts of aryl sulfonic acids include diaryliodonium salts of para-toluene sulfonic acid, diaryliodonium salts of dodecylbenzene sulfonic acid, diaryliodonium salts of benzene sulfonic acid, and diaryliodonium salts of 3-nitrobenzene sulfonic acid. Examples of suitable triarylsulfonium salts of sulfonic acid include triarylsulfonium salts of perfluoroalkylsulfonic acids and triarylsulfonium salts of aryl sulfonic acids. Examples of suitable triarylsulfonium salts of perfluoroalkylsulfonic acids include triarylsulfonium salts of perfluorobutanesulfonic acid, triarylsulfonium salts of perfluoroethanesulfonic acid, triarylsulfonium salts of perfluoro-octanesulfonic acid, and triarylsulfonium salts of trifluoromethane sulfonic acid. Examples of suitable triarylsulfonium salts of aryl sulfonic acids include triarylsulfonium salts of para-toluene sulfonic acid, triarylsulfonium salts of dodecylbenzene sulfonic acid, triarylsulfonium salts of benzene sulfonic acid, and triarylsulfonium salts of 3-nitrobenzene sulfonic acid. Examples of suitable diaryliodonium salts of boronic acids include diaryliodonium salts of perhaloarylboronic acids and preferred triarylsulfonium salts of boronic acids are triarylsulfonium salts of perhaloarylboronic acid.

[00205] The photoinitiator is typically used in the range of from 0.001 to 5 wt.%, based on the total weight of the curable composition. For example, the photoinitiator may be employed in an amount of 0.01 wt.% or more, such as an amount of 0.1 wt.% or more, alternatively 0.15 wt.% or more, alternatively 0.2 wt.% or more, alternatively 0.4 wt.% or more, alternatively 0.6 wt.% or more, alternatively 0.8 wt.% or more, alternatively even 1.0 wt.% or more, while at the same time typically 5 wt.% or less, alternatively 4 wt.% or less based on the total weight of the curable composition.

[00206] The curable composition is typically prepared by combining the curing agent and the silicone-polyolefin blend. The process for combining is not particularly limited, and may be performed using any of the mixing devices described above. Likewise, the curable composition

may be prepared in sequence with the silicone-polyolefin blend (e.g. by adding the curing agent to the silicone-polyolefin blend upon, or soon after, formation thereof). However, it will be appreciated that a separate and/or different mixer, or mixing process, may be used to prepare the curable composition. For example, in some embodiments, the silicone-polyolefin blend is prepared as described above by dynamic crosslinking of the silicone-polyolefin composition via hot-melt extrusion, and the curing agent is then milled into the extruded silicone-polyolefin blend using a roller mill, thereby preparing the curable composition. One of skill in the art will appreciate that various mixing processes and equipment, including any of those described herein, and combinations thereof, may be utilized to combine the curing agent and the silicone-polyolefin and prepare the curable composition.

[00207] In certain embodiments, the curable composition further comprises one or more additional components, such as one or more additives. For example, in certain embodiments, the curable composition may comprise one or more additives comprising, alternatively consisting essentially of, alternatively consisting of: a filler treating agent; a binder; a thickener; a tackifying agent; an adhesion promotor; a compatibilizer; an extender; a plasticizer; an end-blocker; a drying agent; a colorant (e.g. a pigment, dye, etc.); an anti-aging additive; a biocide; a flame retardant; a corrosion inhibitor; a UV absorber; an anti-oxidant; a light-stabilizer; a catalyst (e.g. other than the catalyst (D)), procatalyst, or catalyst generator; an initiator (e.g. a heat activated initiator, an electromagnetically activated initiator, etc.); a photoacid generator; a heat stabilizer; and the like, as well as derivatives, modifications, and combinations thereof. It is to be appreciated that such additives may be classified under different terms of art and, just because an additive is classified under a specific term and/or characterized according to a particular function does not mean that it is thusly limited to that function. Moreover, some additives may be present in a particular component of the curable composition, or instead may be incorporated when forming the curable. Theoretically, the curable composition may comprise any number of additional components and additives, e.g. depending on the particular type and/or function of the same in the curable composition.

[00208] When present, the one or more additives may be combined with the curing agent or the silicone-polyolefin blend before, during, or after combining the curing agent and the silicone-polyolefin blend. Said differently, one or more of the additives may be combined with the silicone-polyolefin blend (or the curing agent) to form an intermediate composition, which is then combined with the curing agent (or the silicone-polyolefin blend) to give the curable composition. Alternatively, the silicone-polyolefin blend, the curing agent, and many of the suitable additives may be combined together in a concerted step. One of skill in the art will readily appreciate that the particular order of addition and/or combination suitable for a given additive will depend on the nature of the additive and the other components of the curable composition, and thus will be

independently selected based on the particular components and parameters being employed. Likewise, it will be appreciated that certain additives (e.g. those unreactive with components (A), (B), (C), and (D) during the preparation of the curable composition) may be introduced into the silicone-polyolefin blend during, or prior to, the compatibilization of components (B) and (C).

[00209] A cured product of the curable composition, and a method of preparing the cured product, are also provided. Specifically, the curable composition may be cured to give the cured product. As will be understood by those of skill in the art, such curing typically comprises activate curing agent, e.g. via heating the composition to a temperature sufficient to activate the radical initiator (e.g. via thermal decomposition), irradiating the photoinitiator, etc. Such activation processes are known in the art, and will be selected based on the particular curing agent utilized.

[00210] The cured product is formed via radical cure of the curable composition, i.e., upon activation of the radical initiator. It will be understood that curing the curable composition generally comprises crosslinking components thereof, such as the functional organosiloxane of component (D). The cured product can be referred to as a silicone-polyolefin elastomer, or a hybrid elastomer.

[00211] As such, the method of preparing the cured product generally comprises heating the curable composition to an elevated temperature, such as a temperature of from 90 to 300, alternatively from 100 to 300, alternatively from 100 to 250, alternatively from 100 to 200 °C, for a time sufficient to cure the curable composition. In some embodiments, the curable composition is cured at a temperature of from 150 to 220 °C for a time of from 1 to 20, alternatively from 5 to 20, alternatively from 5 to 15 minutes.

[00212] In view of the description above and examples below, it will be appreciated that the methods and compositions provided herein provide a cost-effective route to obtaining unique silicone-polyolefin hybrid materials due to inexpensive precursors and the solvent-less preparations. Moreover, the reactive compatibilization of the functionalized polyolefin (B) generates compatibilized graft copolymers in situ, which localizes the compatibilizer at the interface and results in an increased solvent swell resistance and tear strength. Moreover, the cured products maintain satisfactory elongation and tensile strength, exhibit improved handing and appearance characteristics, and present desirable haptics. These and other features of the cured product will be understood in view of the example below.

[00213] The inventive compositions enable the preparation of products and articles with enhanced performance characteristics, including as compared to products and articles formed via organic compositions, silicone compositions, or conventional hybrid silicone-organic compositions. For example, injection moldable articles and compression moldable articles may be made from the inventive compositions having improved toughness (e.g., increased tear strength), chemical resistance (e.g., increased solvent swell resistance), excellent elongation and

tensile strength, and delayed elastic recovery when compared to typical silicone rubber elastomers. Articles made with the inventive compositions also have a higher modulus at comparable hardness values when compared to typical silicone rubber elastomers, and have desirable haptics. For example, haptic properties of articles can be improved by forming a layer on a surface of an article with the inventive compositions.

[00214] Such products and articles may be employed in or for a wide range of applications and in the production of a wide range of consumer products and articles. Examples include products and articles in or for consumer wearable electronics; consumer packaging and dispensing, such as for food, personal care, and beauty care articles and/or products; vibration isolation components; electrical protection in wire & cable applications and coating or co-molding on substrates such as buttons, knobs and user interface controls or components.

[00215] The following examples, illustrating embodiments of this disclosure, are intended to illustrate and not to limit the invention. Unless otherwise noted, all reactions are carried out under air, and all solvents, substrates, and reagents are purchased or otherwise obtained from various commercial suppliers (e.g. Gelest, Acros Organics, Sigma-Aldrich, etc.) and utilized as received.

Equipment and Characterization Parameters

[00216] The following equipment and characterization procedures/parameters are used to evaluate various physical properties of the compounds prepared in the examples below.

[00217] Cure Property: Cure kinetic parameters (S'max, TC90, TS2) were measured using a Moving Die Rheometer for 10 minutes at 115 °C using methods specified in ASTM D5289.

[00218] Plasticity: Plasticity of uncured compositions were measured at 3-hour rest after milling according to ASTM D926.

[00219] Hardness: Durometer hardness of cured products was measured according to ASTM D2240.

[00220] Viscosity: Viscosity was measured with a Brookfield LV DV-E viscometer, equipped with an appropriate spindle, in accordance with ASTM E3116.

[00221] Elastomer Properties: Tensile strength, elongation, and modulus were measured using methods described in ASTM D412; and the toughness values calculated from the same stress-strain curves thus prepared. Tear strengths were measured according to ASTM D624, Die B.

[00222] Solvent Resistance: Solvent immersion samples were produced by fully immersing die cut tensile specimen (described in ASTM D412) into specified solvent at room temperature for 72 hours. The volume changes were calculated from specific gravity measurement as described in ASTM D792 before and after the solvent immersion. Hardness, tensile strength, and elongation of samples after solvent immersion were measured using the same ASTM methods described above.

[00223] Melt Flow Index (MFI): MFI was measured in accordance with ASTM D1238–86, with values typically reported as g / 10 min at 190 °C and 21.6 kg.

Materials

[00224] A brief summary is provided in Table 1 below, setting forth information as to certain abbreviations, shorthand notations, and components utilized in the Examples. Viscosity is typically reported as zero-shear viscosities measured at 25 °C. Degree of polymerization (DP) is typically reported as number average DP, e.g. from NMR, IR, and/or GPC (e.g. relative to standards, such as polystyrene).

Table 1: Materials Utilized

Component	Description
Polysiloxane (A)-1	Diamino-functional PDMS (with amino functionality being pendant), having an amine neutral equivalent of 6500-9000, viscosity (25 °C) of 3000-8000 cP, and a spec. gravity of 0.98
Polysiloxane (A)-2	α,ω-aminopropyl-terminated PDMS having a zero shear viscosity (25 °C) of ~10000 cP and a polydispersity index of ~2
Polyolefin (B)-1	Maleic anhydride-grafted high density polyethylene, having a melt flow index of ~12 g/10 min, and an anhydride content of ~1.2 wt.%
Polyolefin (B)-2	Maleic anhydride-grafted high density polyethylene, having a melt flow index of ~2 g/10 min, and an anhydride content of ~1.2 wt.%
Polyolefin (B)-3	Maleic anhydride-grafted linear low density polyethylene, having a melt flow index of ~1.3 g/10 min, and an anhydride content of ~0.5-1 wt.%
Polyolefin (B)-4	Maleic anhydride-grafted polypropylene, having a melt flow index of ~49 g/10 min, and an anhydride content of ~0.5 to 1.0 wt.%
Polyolefin (B)-5	Maleic anhydride-grafted polypropylene, having a melt flow index of ~22 g/10 min, and an anhydride content of ~0.5 to 1.0 wt.%
Polyolefin (C)-1	Post-consumer high density polyethylene (blow molding/extrusion/film grade) having a melt flow index of 0.6 g/10 min and a specific gravity of 0.96 g/mL
Polyolefin (C)-2	Post-consumer high density polyethylene resin (mixed color) having a melt flow index of 0.5 g/10 min, a specific gravity of 0.95 g/mL, and comprising ~7% polypropylene (as measured in accordance with ASTM D5576)
Polyolefin (C)-3	Post-consumer grade polypropylene resin having a melt flow index of 20 g/10 min and a specific gravity of 0.91 g/mL
Polyolefin (C)-4	Post-consumer grade low density/linear low density polyethylene having a melt flow index of ~1 g/10 min
Polyolefin (C)-5	Polypropylene having a melt flow index of 35 g/10 min and a density of 0.9 g/mL as measured in accordance with ASTM D792
Polyolefin (C)-6	Polypropylene having a melt flow index of 4 g/10 min and a density of 0.9 g/mL as measured in accordance with ASTM D792
Polyolefin (C)-7	High density polyethylene having a melt flow index of 0.8 g/10 min and a density of 0.96 g/mL as measured in accordance with ASTM D792
Silicone Base (D)-1	40 durometer general purpose silicone rubber (HCR) base
Silicone Base (D)-2	30 durometer general purpose silicone rubber (HCR) base
Silicone Base (D)-3	50 durometer general purpose silicone rubber (HCR) base
Silicone Base (D)-4	70 durometer general purpose silicone rubber (HCR) base

Silicone Base (D)-5	30 durometer high strength silicone rubber (HCR) base
Silicone Base (D)-6	50 durometer high strength silicone rubber (HCR) base
Silicone Base (D)-7	18 durometer general purpose silicone rubber (HCR) base
Silicone Base (D)-8	40 durometer general purpose silicone rubber (HCR) base containing 22.7 wt.% precipitated silica as reinforcing filler
Organopolysiloxane 1	Vinyl-terminal PDMS, having a DP of ~800-1000
Organopolysiloxane 2	Vinyl-terminal PDMS, having a DP of ~150
Catalyst (E)-1	A Karstedt's Catalyst complex, comprising a mixture of a platinum(IV) complex of 1,3-diethenyl-1,1,3,3-tetramethyldisiloxane (1 wt.%), a dimethylvinylsiloxy-terminated polydimethylsiloxane having a viscosity of ~0.45 Pa-s at 25 °C (92 wt.%), and tetramethyldivinyldisiloxane (7 wt.%)
Cure Agent	50 wt.% bis-(2,4-dichlorobenzoyl)peroxide in silicone gum
Inhibitor	3 wt.% 1-ethynylcyclohexanol in vinyl-terminated PDMS
Crosslinker	Branched oligomeric trimethylsilyl-terminated polydimethylsiloxane-polyhydridomethylsiloxanecopolymer with an average of one T branched unit per chain with viscosity (25 °C) of ~ 15 cP and including ~ 0.8 wt.% H in the form of SiH
Filler	Dispersion of 64 wt.% of dimethylvinylsiloxy-terminated polydimethylsiloxane having a viscosity of ~55,000 cP at 25 °C with a total vinyl content of 0.088 wt.% and 36 wt.% of dimethylvinylated and trimethylated treated fumed silica
Solvent 1	Love Beauty & Planet hand cream
Solvent 2	Cetaphil SPF 50 water-in-oil based sunscreen

General Procedure for Preparing Hybrid Elastomer Base

[00225] Equipment: Hybrid elastomer bases (or silicone-polyolefin compositions) were prepared in a 50 cc Haake Rheomix OS system (Haake Bowl) with roller rotors driven and controlled by a PolyoLab OS system.

[00226] Synthesis: The bowl was heated to 180 °C and the rotors were set to 75 rpm. A Silicone Base (D) was added to the bowl in portions under a flow of N_2, followed by a Polyolefin (B) and a Polyolefin (C). After approximately 1 minute of blending a smooth white mixture was obtained. A Polysiloxane (A) was added and blending continued for 5 minutes and then the product was emptied onto a steel tray and allowed to cool.

[00227] Curing Elastomers: A polished stainless steel mold with internal cavity dimensions of 6” by 6” by 2 mm was conditioned by covering the interior surfaces with a Teflon release coating spray. A curable composition was prepared by blending the hybrid elastomer base with the Cure Agent (1 part per one hundred (pph) parts of hybrid elastomer base, by weight) directly on the two-roll mill until the resulting mass was visually homogeneous, which was then divided up into portions to fill 110% of a mold with internal cavity dimension of 6” by 6” by 2 mm. The individual

portion of formulated compound was preformed into a flat sheet that is slightly smaller than 6" by 6", placed in the mold at room temperature. The compounds were compression molded in a heated hydraulic press at 115 °C and 1500 psi for 10 minutes and removed from the mold immediately after cure. The cured hybrid elastomer sheets were rested at room temperature for 24 hours before characterization.

Examples 1-6 and Comparative Examples 1-4

[00228] Preparation of Examples 1-6 and Comparative Example 1: Synthesis of hybrid elastomer bases was performed according to the general procedure above. The procedure was repeated to produce sufficient material for a given test. Each hybrid elastomer base was then cured in accordance with the procedure described above, and physical properties were measured. The particular components, parameters, and properties of material obtained for Examples 1-6 and Comparative Examples 1-4 are shown in Tables 2-3 below, respectively.

Table 2: Components and Properties of Examples 1-6

Example:	1	2	3	4	5	6
Silicone Base (D)-1 [g]:	36	36	36	36	36	36
Polyolefin (B)-2 [g]:	6.75	4.50	2.25	4.50	6.75	2.25
Polysiloxane (A)-1 [g]:	2.3	2.3	2.3	2.3	2.3	2.3
Polyolefin (C)-7 [g]	2.25	4.50	6.75	--	--	--
Polyolefin (C)-1 [g]	--	--	--	4.5	2.25	6.75
S'max [lb-in]:	11.47	11.25	11.79	11.62	11.26	11.58
TC90 [s]:	78.96	72.6	63.14	76.96	72.1	72.78
TS2 [s]:	25.8	25.2	25.8	24.0	25.2	25.37
Hardness [Shore A]:	52	53	50	53	54	57
Tensile Strength [MPa]:	8.13	8.28	5.77	6.62	7.61	5.32
Elongation [%]:	392	470	377	384	415	408
100% Modulus [MPa]:	1.92	1.97	1.73	1.87	1.88	2.12
Tear B [ppi]:	91	112	87	91	88	87
Plasticity [3hr]:	100	96	116	103	96	129

Table 3: Components and Properties of Comparative Examples 1-4

Comparative Example:	1	2	3	4
Silicone Base (D)-1 [g]:	36	36	36	36
Polyolefin (B)-2 [g]:	9	9	--	--
Polysiloxane (A)-1 [g]:	--	2.3	2.3	2.3
Polyolefin (C)-7 [g]	--	--	9	--
Polyolefin (C)-1 [g]	--	--	--	9
S'max [lb-in]:	12.37	11.10	9.89	9.15
TC90 [s]:	79.83	74.84	77.24	73.09
TS2 [s]:	26.40	25.20	32.40	27.59
Hardness [Shore A]:	56	53	45	51

Tensile Strength [MPa]:	3.29	8.16	3.44	4.01
Elongation [%]:	316	438	592	622
100% Modulus [MPa]:	2.06	1.95	1.17	1.59
Tear B [ppi]:	61	86	72	78
Plasticity [3hr]:	75	98	135	116

Examples 7-15 and Comparative Examples 5-8

[00229] Preparation of Examples 7-15 and Comparative Examples 5-8: Synthesis of hybrid elastomer bases was performed in accordance with the general procedure above. In addition, curing of each hybrid elastomer base was performed in accordance with the general procedure above.

[00230] The particular components, parameters, and properties of material obtained for Examples 7-15 and Comparative Examples 5-8 are shown in Tables 4 and 5 below, respectively.

Table 4: Components and Properties of Examples 7-12

Example:	**7**	**8**	**9**	**10**	**11**	**12**
Silicone Base (D)-1 [g]:	36.0	36.0	31.5	36.0	36.0	36.0
Polyolefin (B)-1 [g]:	4.50	--	--	--	--	--
Polyolefin (B)-2 [g]:	--	4.50	6.75	4.50	4.50	--
Polyolefin (B)-3 [g]:	--	--	--	--	--	4.5
Polysiloxane (A)-1 [g]:	2.3	--	2.3	2.3	2.3	2.3
Polysiloxane (A)-2 [g]:	--	2.3	--	--	--	--
Polyolefin (C)-1 [g]	4.50	4.50	6.75	--	2.70	--
Polyolefin (C)-2 [g]	--	--	--	4.50	--	--
Polyolefin (C)-4 [g]	--	--	--	--	--	4.5
Polyolefin (C)-6 [g]	--	--	--	--	1.8	--
S'max [lb-in]:	--	--	--	11.71	11.96	7.82
TC90 [s]:	--	--	--	78.48	66.04	81.21
TS2 [s]:	--	--	--	23.40	24.00	32.40
Hardness [Shore A]:	52	56	64	54	55	45
Tensile Strength [MPa]:	6.33	5.51	6.03	7.27	4.98	6.35
Elongation [%]:	341	293	223	379	307	488
100% Modulus [MPa]:	1.89	2.23	3.23	1.99	1.86	1.22
Tear B [ppi]:	66	75	61	75	96	164
Plasticity [3hr]:	113	115	154	107	123	108

Table 5: Components and Properties of Examples 13-15 and Comparative Examples 5-8

Example:	**13**	**14**	**15**	**CE5**	**CE6**	**CE7**	**CE8**
Silicone Base (D)-1 [g]:	36	36	36	45	145.5	33.8	35.6
Polyolefin (B)-1 [g]:	--	--	--	--	--	8.45	8.90
Polyolefin (B)-2 [g]:	--	4.5	4.5	--	--	--	--
Polyolefin (B)-4 [g]:	4.5	--	--	--	--	--	--
Polysiloxane (A)-1 [g]:	2.30	0.46	4.60	--	4.50	2.22	--

Polyolefin (C)-1 [g]	--	4.5	4.5	--	--	--	--
Polyolefin (C)-5 [g]	4.5	--	--	--	--	--	--
S'max [lb-in]:	0.32	--	--	10.1	9.2	11.0	11.7
TC90 [s]:	55.8	--	--	61.2	65.6	98.8	96.8
TS2 [s]:	22.8	--	--	27.0	28.8	28.2	30.0
Hardness [Shore A]:	47	59	51	40	35	55	55
Tensile Strength [MPa]:	4.61	4.21	5.96	5.98	4.78	7.65	3.56
Elongation [%]:	472	220	444	419	458	384	363
100% Modulus [MPa]:	1.30	2.57	1.82	1.00	0.74	2.28	1.48
Tear B [ppi]:	111	76	163	46	105	87	53
Plasticity [3hr]:	130	110	113	59	77	102	72

Examples 16-19 and Comparative Examples 9-23

[00231] Preparation of Examples 16-19 and Comparative Examples 9-23: Synthesis of hybrid elastomer bases was performed in accordance with the general procedure above. In addition, curing of each hybrid elastomer base was performed in accordance with the general procedure above.

[00232] The particular components, parameters, and properties of material obtained for Examples 16-19 and Comparative Examples 9-23 are shown in Tables 6-8 below, respectively.

Table 6: Components and Properties of Examples 16-19 and Comparative Examples 9-11

Example:	**16**	**17**	**18**	**19**	**CE9**	**CE10**	**CE11**
Silicone Base (D)-2 [g]:	36.0	--	--	--	36.0	--	--
Silicone Base (D)-3 [g]:	--	36.0	--	--	--	--	36.0
Silicone Base (D)-5 [g]:	--	--	36.0	--	--	36.0	--
Silicone Base (D)-6 [g]:	--	--	--	36.0	--	--	--
Polyolefin (B)-1 [g]:	--	--	--	--	--	--	--
Polyolefin (B)-2 [g]:	4.5	4.5	4.5	4.5	--	9.0	--
Polysiloxane (A)-1 [g]:	2.3	2.3	2.3	2.3	--	2.3	--
Polyolefin (C)-1 [g]	4.5	4.5	4.5	4.5	--	--	--
S'max [lb-in]:	--	--	--	--	10.13	--	7.10
TC90 [s]:	--	--	--	--	68.40	--	52.20
TS2 [s]:	--	--	--	--	25.80	--	22.20
Hardness [Shore A]:	57	66	53	62	38	56	47
Tensile Strength [MPa]:	6.22	8.11	6.56	6.88	5.46	6.95	7.94
Elongation [%]:	382	335	460	396	514	435	501
100% Modulus [MPa]:	2.10	3.19	2.07	3.46	0.66	2.35	0/94
Tear B [ppi]:	69	102	234	256	33	264	54
Plasticity [3hr]:	110	110	158	264	58	154	83

Table 7: Components and Properties of Comparative Examples 12-18

Example:	**CE12**	**CE13**	**CE14**	**CE15**	**CE16**	**CE17**	**CE18**
Silicone Base (D)-2 [g]	--	--	--	--	--	33.8	--
Silicone Base (D)-3 [g]	--	--	--	--	--	--	33.8

Silicone Base (D)-4 [g]:	36.0	--	--	--	--	--	--
Silicone Base (D)-5 [g]:	--	36.0	--	--	--	--	--
Silicone Base (D)-6 [g]:	--	--	36.0	--	--	--	--
Silicone Base (D)-7 [g]:	--	--	--	36.0		--	--
Silicone Base (D)-8 [g]					36.0	--	--
Polyolefin (B)-1 [g]:	--	--	--	--	--	8.44	8.44
Polysiloxane (A)-1 [g]:	--	--	--	--	--	2.11	2.11
S'max [lb-in]:	12.27	7.86	6.87	8.12	9.51	9.80	11.18
TC90 [s]:	63.00	59.40	45.00	175.80	61.20	105.00	95.40
TS2 [s]:	19.60	25.80	21.60	105.00	24.00	24.60	24.00
Hardness [Shore A]:	58	36	46	25	43	51	60
Tensile Strength [MPa]:	7.22	10.09	9.12	3.46	5.09	6.44	7.85
Elongation [%]:	434	888	768	777	491	364	377
100% Modulus [MPa]:	1.38	0.69	1.01	0.50	1.17	1.57	2.28
Tear B [ppi]:	112	146	200	69	51	68	116
Plasticity [3hr]:	121	88	98	47	60	110	158

Table 8: Components and Properties of Comparative Examples 19-23

Example:	**CE19**	**CE20**	**CE21**	**CE22**	**CE23**
Silicone Base (D)-4 [g]:	33.8	--	--	--	--
Silicone Base (D)-5 [g]:	--	33.8	--	--	--
Silicone Base (D)-6 [g]:	--	--	33.8	--	--
Silicone Base (D)-7 [g]:	--	--	--	33.8	--
Silicone Base (D)-8 [g]:	--	--	--	--	33.8
Polyolefin (B)-1 [g]	8.44	8.44	8.44	8.44	8.44
Polysiloxane (A)-1 [g]	2.11	2.11	2.11	2.11	2.11
S'max [lb-in]:	--	10.82	11.69	11.04	10.82
TC90 [s]:	--	76.80	82.20	373.20	87.60
TS2 [s]:	--	23.40	23.40	170.40	24.60
Hardness [Shore A]:	71	55	63	41	54
Tensile Strength [MPa]:	6.75	6.24	7.21	3.45	6.21
Elongation [%]:	271	443	446	443	316
100% Modulus [MPa]:	3.58	2.06	2.71	1.17	2.10
Tear B [ppi]:	110	249	276	65	85
Plasticity [3hr]:	225	146	225	80	104

Example 20

[00233] Preparation of Example 20:

[00234] In Example 20, a masterbatch was prepared in a 50 cc Haake Rheomix OS system (Haake Bowl) with roller rotors driven and controlled by a PolyoLab OS system. The bowl was heated to 180 °C and the rotors were set to 100 rpm. In Example 20, a liquid rubber was utilized, and an organopolysiloxane of the liquid rubber was added to the bowl in portions under a flow of N_2, followed by a Polyolefin (B) and a Polyolefin (C). After approximately 5 minute of blending a

smooth white mixture was obtained. A Polysiloxane (A) was added and blending continued for 10 minutes and then the product was emptied onto a steel tray and allowed to cool.

[00235] The particular components utilized in the hybrid masterbatch of Example 20 are shown in Table 9 below.

Table 9: Components and Parameters of Hybrid Masterbatch of Example 20

Component	**Mass [g]**
Organopolysiloxane 1 [g]:	25.3
Polyolefin (B)-5 [g]:	9.89
Polysiloxane (A)-2[g]:	4.95
Polyolefin (C)-3 [g]	9.89

[00236] The hybrid masterbatch was then utilized in a silicone-polyolefin composition in the form of a two-part formulation. The components and their amounts for each part of the two-part formulation are below in Table 10. Each part of the two-part formulation is prepared by mixing the components.

Table 10: Components of Two-Part Formulation of Example 20

Part A Components	**Wt.%**
Hybrid Masterbatch of Table 9:	97.63
Catalyst (E)-1	0.56
Inhibitor	1.81
Part B Components	**Wt.%**
Filler	95.04
Crosslinker	1.21
Inhibitor	1.874
Organopolysiloxane 2	1.874

[00237] Part A was prepared by combining and mixing the components of Part A in a speed mixer (3 times for 20s @ 1800rpm, scraped down between mixes). Part B was prepared by incorporating the Inhibitor in Silicone Base (D)-10 to give a premix and combining the premix with the other components of Part B and mixing in a speed mixer (3 times for 20s @ 1800rpm, scraped down between mixes).

[00238] Part A and Part B were mixed at 1:1.1 ratio by weight using a speed mixer (3 times for 20 seconds at 1800 rpm, scraped down between mixes) to give a visually homogenous mixture. The visually homogeneous mixture was loaded in a 10”x10”x2mm mold and press cured at 1500 psi and 115 °C for 10 minutes. The mold was cooled in a room temperature hydraulic press until it reached room temperature, after which a cured rubber plaque was removed for property testing. The results are below in Table 11.

Table 11: Properties of Example 20

Example:	**20**

Hardness [Shore A]:	48
Tensile Strength [MPa]:	4.28
Elongation [%]:	326
100% Modulus [MPa]:	1.47
Tear B [ppi]:	50

[00239] In Example 20, the curable silicone (D) is a liquid rubber. In contrast, in Examples 1-19, the curable silicone (D) is an elastomer component.

[00240] Certain silicone-polyolefin hybrid elastomers are subjected to a solvent resistance test as described above. The results of the solvent resistance test for these certain elastomers are below in Table 12. In table 12, h/RT indicates hours at room temperature.

Table 12: Solvent Resistance

Example	Unaged tensile strength (MPa)	Tensile strength (MPa) after 72 h/RT, Solvent 1	Tensile strength (MPa) after 72 h/RT, Solvent 2
CE10	6.44	4.43	6.18
18	6.11	4.72	6.62
CE13	10.10	3.73	8.49
CE7	8.14	5.69	8.03
4	6.15	5.99	6.06
CE5	7.58	3.28	6.76

[00241] The invention has been described in an illustrative manner, and it is to be understood that the terminology which has been used is intended to be in the nature of words of description rather than of limitation. Obviously, many modifications and variations of the present invention are possible in light of the above teachings. The invention may be practiced otherwise than as specifically described.

CLAIMS

What is claimed is:

1. A silicone-polyolefin composition, comprising:

(A) a polysiloxane comprising an average of at least one functional group X per molecule;

(B) a functionalized polyolefin comprising an average of at least one functional group Y per molecule, where the functional group Y is reactable with the functional group X of the polysiloxane (A) to form a bond therebetween;

(C) a polyolefin that is not reactable with components (A) or (B); and

(D) a curable silicone comprising at least one of an elastomer component or a liquid rubber.

2. The silicone-polyolefin composition of claim 1, wherein the polysiloxane (A) has the following average unit formula:

$$[X_mR^1_{3-m}SiO_{1/2}]_a[X_nR^1_{2-n}SiO_{2/2}]_b,$$

where X is the functional group defined above, each R^1 is an independently selected hydrocarbyl group, subscript m is independently 1 or 0 in each moiety indicated by subscript a, subscript n is independently 1 or 0 in each moiety indicated by subscript b, and subscripts a and b are each mole fractions such that $a+b=1$, with the provisos that $0<a<1$, $0<b<1$, and the polysiloxane (A) comprises at least one functional group X.

3. The silicone-polyolefin composition of claim 1 or 2, wherein: (i) each functional group X comprises an aminoalkyl group, a methacryloxy group, a silicon-bonded hydroxyl group, an anhydride group, a silicon-bonded hydrogen atom, an olefinically-unsaturated group, or a carbinol group; (ii) the polysiloxane (A) has a viscosity of at least 3,000 cP at 25 °C; (iv) the polysiloxane (A) has a degree of polymerization (DP) of from 20 to 1,200; (iv) the polysiloxane (A) is present in the silicone-polyolefin composition in an amount of from 0.5 to 15 wt.%, based on the total weight of the silicone-polyolefin composition; or (v) any combination of (i)-(iv).

4. The silicone-polyolefin composition of any one of claims 1-3, wherein: (i) the functionalized polyolefin (B) comprises a functionalized polyethylene (PE), polypropylene (PP), or polyethylene-alpha olefin copolymer; (ii) each functional group Y comprises an epoxide group, an aminoalkyl group, a trimethoxysilyl group, a hydridosilyl group, or an anhydride group; (iii) the functionalized polyolefin (B) has a density of from 0.86-0.96 g/cm^3; (iv) the functionalized polyolefin (B) is functionalized with from 0.05 to 10 wt.% of functional moieties comprising the functional group Y, based on the total weight of the functionalized polyolefin (B); (v) the functionalized polyolefin (B)

is present in the silicone-polyolefin composition in an amount of from 1 to 40 wt.%, based on the total weight of the silicone-polyolefin composition; or (vi) any combination of (i)-(v).

5. The silicone-polyolefin composition of any one of claims 1-4, wherein the curable silicone (D) comprises the elastomer component, and wherein the elastomer component: (i) comprises a reaction-curable organosiloxane; (ii) comprises a filler; (iii) is further defined as a polydiorganosiloxane gum; (iv) comprises olefinic unsaturation; (v) is present in the silicone-polyolefin composition in an amount of from 55 to 99 wt.%, based on the total weight of the silicone-polyolefin composition; (vi) is immiscible with the functionalized polyolefin (B); or (vii) any combination of (i) to (vi).

6. The silicone-polyolefin composition of any one of claims 1-5, wherein the polyolefin (C): (i) comprises a virgin polyolefin; (ii) comprises a recycled polyolefin; (iii) comprises a polyethylene (PE), polypropylene (PP), or polyethylene-alpha olefin copolymer; (iv) is present in the silicone-polyolefin composition in an amount of from 1 to 40 wt.%, based on the total weight of the silicone-polyolefin composition; or (v) any combination of (i) to (iv).

7. The silicone-polyolefin composition of any one of claims 1-6, further comprising: (i) (E) a catalyst capable of facilitating a coupling reaction between the functional group X of the polysiloxane (A) and the functional group Y of the functionalized polyolefin (B); (ii) a reaction inhibitor; or (iii) both (i) and (ii).

8. The silicone-polyolefin composition of any one of claims 1-7, wherein the silicone-polyolefin composition: (i) is free from a carrier vehicle; (ii) is free from a reaction catalyst or promotor; (iii) is further defined as a polyolefin-in-silicone dispersion; or (iv) any combination of (i)-(iii).

9. A method of preparing a silicone-polyolefin blend, said method comprising:

combining the polysiloxane (A), the functionalized polyolefin (B), the polyolefin (C), and the curable silicone (D) to prepare the silicone-polyolefin composition of any one of claims 1-8; and

reacting the polysiloxane (A) and the functionalized polyolefin (B) in the presence of the polyolefin (C) and the curable silicone (D), thereby preparing the silicone-polyolefin blend.

10. The method of claim 9, wherein reacting the polysiloxane (A) and the functionalized polyolefin (B) in the presence of the polyolefin (C) and the curable silicone (D): (i) comprises melt blending the silicone-polyolefin composition at a temperature greater than the melting point of the

functionalized polyolefin (B) and greater than the melting point of the polyolefin (C); (ii) comprises extruding the silicone-polyolefin composition; (iii) is carried out under substantially solvent-free conditions; (iv) is carried out in the presence of (E) a catalyst capable of facilitating a coupling reaction between the functional group X of the polysiloxane (A) and functional group Y of the functionalized polyolefin (B); or (v) any combination of (i)-(iv).

11. A silicone-polyolefin blend prepared according to the method of claim 9 or 10.

12. A curable composition, comprising the silicone-polyolefin blend of claim 11 and a curing agent.

13. The curable composition of claim 12, wherein: (i) the curing agent comprises a radical initiator and/or a photoinitiator; (ii) the curable composition is free from a carrier vehicle; or (iii) both (i) and (ii).

14. A method of preparing a cured product, said method comprising curing the curable composition of claim 12 or 13, thereby preparing the cured product.

15. A cured product of the curable composition of claim 12 or 13 or prepared according to the method of claim 14.

16. Use of the silicone-polyolefin composition of any one of claims 1-8, the silicone-polyolefin blend of claim 11, the curable composition of claim 12 or 13, or the cured product of claim 15 in consumer wearable electronics; consumer packaging or dispensing articles and/or products; vibration isolation components; electrical protection in wire and cable applications; and/or coating or co-molding on substrates.

17. A method of improving haptic properties of an article having a surface, said method comprising forming a layer on the surface of the article with the silicone-polyolefin composition of any one of claims 1-8, the silicone-polyolefin blend of claim 11, the curable composition of claim 12 or 13, or the cured product of claim 15.

18. An article comprising the cured product of claim 15, wherein the article is further defined as a consumer wearable electronic, a consumer packaging or dispensing article and/or product, a vibration isolation component, a wire or cable, or a consumer interface component.

INTERNATIONAL SEARCH REPORT

International application No
PCT/US2022/045139

A. CLASSIFICATION OF SUBJECT MATTER
INV. C08L83/04
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)
C08L C08G

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

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2015/196459 A1 (DOW GLOBAL TECHNOLOGIES LLC [US]; YU HAIYANG [CN]; HUO YANLI [BE]) 30 December 2015 (2015-12-30)	1-16,18
A	abstract Inventive example 3; paragraphs [0058] - [0060]	17
A	EP 3 050 911 A1 (POLYMERES TECH SOC D [FR]) 3 August 2016 (2016-08-03) paragraphs [0029] - [0062]	1-18

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

☒ See patent family annex.

* Special categories of cited documents :

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"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

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"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
18 January 2023	30/01/2023

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/US2022/045139

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