(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau

(43) International Publication Date
9 July 2009 (09.07.2009)

PCT

(10) International Publication Number
WO 2009/085183 A2

(51) International Patent Classification:
C08L 83/04 (2006.01)

(21) International Application Number:
PCT/US2008/013847

(22) International Filing Date:
17 December 2008 (17.12.2008)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
61/015,412 20 December 2007 (20.12.2007) US

(71) Applicant *(for all designated States except US)*: **HENKEL CORPORATION** [US/US]; 1001 Trout Brook Crossing, Rocky Hill, Connecticut 06067 (US).

(72) Inventors; and

(75) Inventors/Applicants *(for US only)*: **CHU, Hsien-Kun** [US/US]; 6 Harvest Hill, Wethersfield, Connecticut 06109 (US). **LIM, Thomas, Fay-Oy,** [US/US]; 128 Route 148, Killingworth, Connecticut 06419 (US). **DECATO, Alfred, A.,** [US/US]; 86 Colonial Street, Oakville, Connecticut 06779 (US). **COMO, Paige** [US/US]; 35 Garland Drive, Glastonbury, Connecticut 06033 (US). **LIISTRO JR.,, Mathias, E.,** [US/US]; 32 Parkside Drive, Plainville, Connecticut 06062 (US). **CROSS, Robert, P.,** [US/US]; 3290 Main Street, Rocky Hill, Connecticut 06067 (US).

(74) Agent: **CUNNINGHAM, Marina, F.,**; McCormick, Paulding & Huber LLP, 185 Asylum Street, CityPlace II, Hartford, Connecticut 06103-3410 (US).

(81) Designated States *(unless otherwise indicated, for every kind of national protection available)*: AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BR, BW, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DE, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IS, JP, KE, KG, KM, KN, KP, KR, KZ, LA, LC, LK, LR, LS, LT, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PG, PH, PL, PT, RO, RS, RU, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.

(84) Designated States *(unless otherwise indicated, for every kind of regional protection available)*: ARIPO (BW, GH, GM, KE, LS, MW, MZ, NA, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MT, NL, NO, PL, PT, RO, SE, SI, SK, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

Declaration under Rule 4.17:
— *as to the identity of the inventor (Rule 4.17(i))*

Published:
— *without international search report and to be republished upon receipt of that report*

WO 2009/085183 A2

(54) Title: METHOD OF POLYMER BODYING TO OBTAIN INSTANT SEALING OF FORM-IN-PLACE ENGINE GASKETING SEALANTS

(57) Abstract: The present invention provides methods of preparing reactive compositions that have instant sealability. More specifically, in the methods of the present invention, a hydroxy-functionalized polydiorganosiloxane is reacted with a C1 tri-functional silane to form a chain-extended polyorganosiloxane. The chain-extended polyorganosiloxane then is reacted with a moisture-curable silane to end-cap the polyorganosiloxane with moisture curing groups. The present invention also provides methods of forming gasket assemblies and methods of sealing substrates together using the reactive compositions.

METHOD OF POLYMER BODYING TO OBTAIN INSTANT SEALING OF FORM-IN-PLACE ENGINE GASKETING SEALANTS

FIELD OF THE INVENTION

[0001] The present invention provides methods of preparing reactive compositions with instant sealing capabilities. In particular, the methods build the molecular weight of reactive polymers, which produces instant seal compositions for use in a variety of applications, such as engine gasketing sealants.

BRIEF DESCRIPTION OF RELATED TECHNOLOGY

[0002] Curable silicone compositions are used in a broad range of applications including automotive, construction, highway, electronic device and package assembly, appliance assembly and consumer uses. Silicone compositions are used as sealants, conformal coatings, potting compounds, and the like. Typically, silicone compositions used in these applications have been tailored to provide the strength and toughness required for the application at hand. In addition to these properties, rapid cure speeds and product stability are often desired.

[0003] In some applications, it is desirable to use a sealant that provides instant sealability upon application. Such compositions would provide resistance to blow-out. For instance, when preparing form-in-place gasket assemblies, such as engine gaskets, it is desirable to use compositions that are both dispensable and provide an instant seal upon assembly of the gasket. This allows the seal to be tested immediately after dispensing the composition, thereby improving the efficiency of defect detection.

[0004] Therefore, it is desirable to provide methods for preparing reactive compositions, useful as sealants, that exhibit instant sealability upon application. Specifically, it is desirable to provide methods that build the molecular weight of reactive polymers to form instant seal compositions.

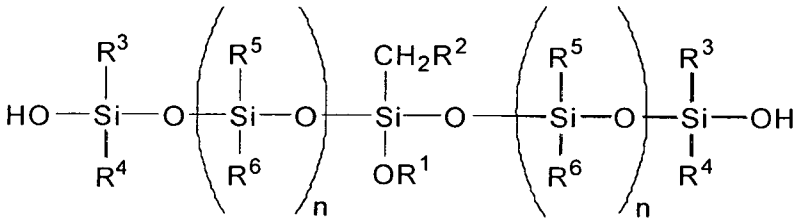
SUMMARY OF THE INVENTION

[0005] The present invention provides methods of preparing reactive compositions, which build the molecular weight of reactive polymers to provide instant sealability upon

application to substrates. The present invention also provides methods of forming gasket assemblies and methods of sealing substrates together with the reactive compositions described herein.

[0006] In one aspect of the present invention, there is provided a method of preparing a reactive composition, which includes the steps of:

(a) reacting a hydroxy-terminated polydiorganosiloxane with a C_1 tri-functional silane, where the ratio of the hydroxy-terminated polydiorganosiloxane to the C_1 tri-functional silane is about 2:1 to about 1:0.8, in the presence of a catalyst to form a chain-extended polyorganosiloxane having the formula II:

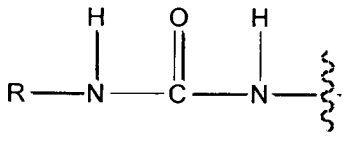
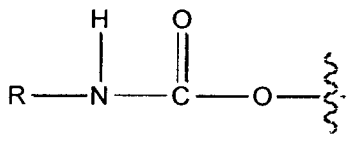


II

where:

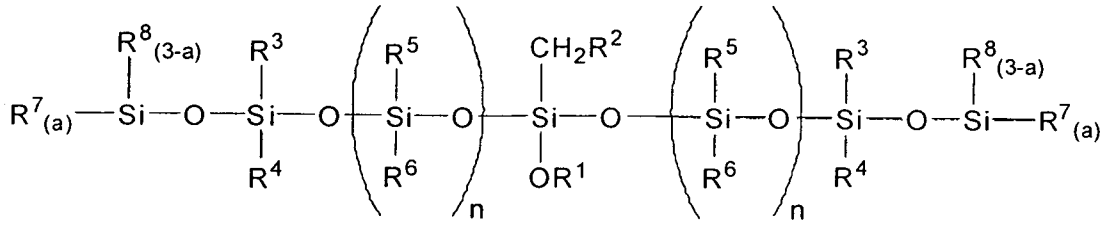
R^1, R^3, R^4, R^5 and R^6 are independently selected from C_{1-4} alkyl;

R^2 is selected from (meth)acryloxy, acetoxy, alkylcarbamato, amino, acetate,

 and  ; and

n is from 10 to 3500; and

(b) reacting the chain-extended polyorganosiloxane of formula II with a moisture curable silane to form a reactive polymer having the formula III:



III

where:

R^7 is selected from alkyl, alkenyl, alkynyl and methacryloxy;

R^8 is selected from aminoxy, oximino, alkoxy, enoxy, acetoxy; and

a is 0 or 1.

[0007] In another aspect of the present invention, there is provided a method of forming a gasket assembly, which includes the steps of:

(a) applying the curable instant seal composition described herein to at least one of two substrate surfaces;

(b) exposing the composition to moisture to effectuate at least partial cure; and

(c) mating the substrate surfaces in abutting relationship to form the gasket assembly.

[0008] In yet another aspect of the present invention, there is provided a method for sealing together two substrates, which includes the steps of:

(a) applying the curable instant seal composition described herein to at least one of two substrate surfaces;

(b) mating the substrate surfaces in abutting relationship to form an assembly;

(c) exposing the composition to moisture; and

(d) maintaining the abutting relationship for a time sufficient to allow the composition to cure.

[0009] In another aspect of the present invention, there is provided a curable instant seal composition including:

(a) a reactive composition including: at least one hydroxy-terminated polydimethylsiloxane; ethyl silicate; methacryloxymethyltrimethoxysilane; 1,8-diazabicyclo[5.4.0]undec-7-ene; vinyl trimethoxysilane; amorphous hydrophobic silica; calcium carbonate; polybutene; hexamethyldisilazane; tris copoly(oxy-1-methylethylene) oxyethylene; iron aluminum silicate; carbon black; 3-aminopropyl trimethoxysilane; and

(b) a cure system including dioctyltindicarboxylate.

DETAILED DESCRIPTION OF THE INVENTION

[0010] The present invention provides methods of polymer bodying to provide instant sealability to reactive compositions. Specifically, the methods are directed to preparing reactive compositions by building the molecular weight of the reactive polymers contained therein. These compositions may be used as sealants, such as, for example, in gasket assemblies. More specifically, the reactive compositions are prepared by first reacting a hydroxy-functionalized polydiorganosiloxane with a C_1 tri-functional silane in the presence of a catalyst to form a chain-extended polyorganosiloxane. This chain-extended polyorganosiloxane is an intermediate in the process described herein. The chain-extended polyorganosiloxane then is reacted with a moisture-curable silane in a second reaction step to form a final reactive polymer. The final reactive polymer is curable upon exposure to moisture.

[0011] The reactive compositions formed in accordance with the methods described herein are useful in a variety of end-use applications, such as adhesives and sealants, particularly as sealants for form-in-place gaskets, as well as other uses in the electronic, automotive and consumer markets.

[0012] The term "instant sealability," as used herein, refers to the blow-out resistance of the reactive compositions prepared as described herein. Blow-out resistance is typically referred to as the ability of a sealant composition to form a liquid engine gasket without failing during a pressurized leak check. Blow-out resistance is tested by simulating a typical engine leak check. For instance, a controlled gap is filled with the sealant composition and subjected to air pressure before it is cured. The consistency of the composition must be such as to contain this pressure for a specified time. Failure is determined by a sudden loss of pressure within the fixture. In

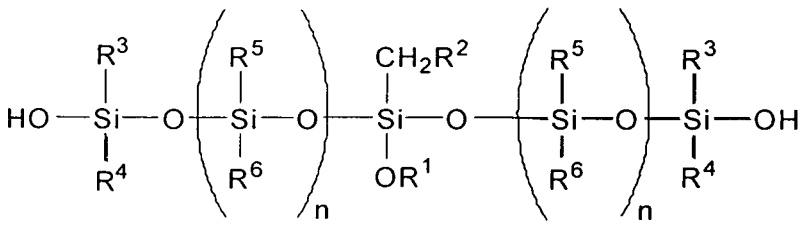
accordance therewith, the reactive compositions described herein have a blow-out resistance of about 62 seconds or greater at 2 psi when tested with a gap of 1.0 mm. In some embodiments, the compositions have a blow-out resistance of greater than about 30 seconds at 4 psi when tested with a gap of 1.5 mm.

[0013] The term "cure" or "curing," as used herein, refers to a change in state, condition, and/or structure in a material that is usually, but not necessarily, induced by at least one variable, such as time, temperature, moisture, radiation, presence and quantity in such material of a curing catalyst or accelerator, or the like. The terms cover partial as well as complete curing.

First Reaction Step

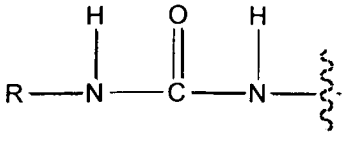
[0014] As mentioned above, in accordance with the methods of the present invention, in the first reaction step, a hydroxy-functionalized polydiorganosiloxane is reacted with a C_1 tri-functional silane. This reaction step produces a chain-extended polyorganosiloxane, which is an intermediate reaction product. The chain-extended polyorganosiloxane is subsequently reacted with a moisture-curable silane in a second reaction step, which will be described in more detail below.

[0015] For instance, in the first reaction step, the hydroxy-functionalized polydiorganosiloxane starting material may be hydroxy-terminated. Desirably, the ratio of the hydroxy-functionalized polydiorganosiloxane to the C_1 tri-functional silane starting material is about 2:1 to about 1:0.8. Because a stoichiometric shortage of the C_1 tri-functional silane is used in this first reaction step, the C_1 tri-functional silane acts to chain-extend the hydroxy-functionalized polydiorganosiloxane. The resultant chain-extended polyorganosiloxane has the following general formula II:

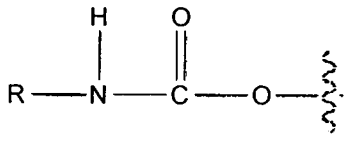


II

[0016] In formula II, desirably, R^1, R^3, R^4, R^5 and R^6 are independently selected from C_{1-4} alkyl; R^2 is selected from (meth)acryloxy, acetoxy, alkylcarbamato, amino, acetate,

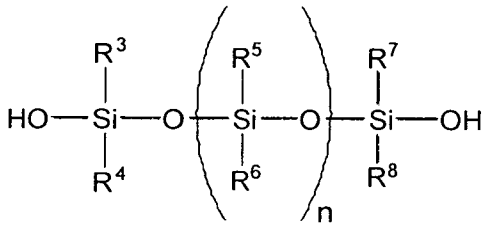


and



; and n is from 10 to 3500.

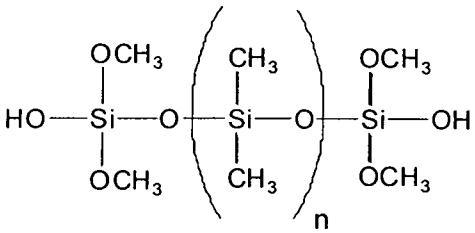
[0017] Hydroxy-functionalized polydiorganosiloxanes suitable for use as starting materials in this first reaction step include those having the following general formula I:



I

[0018] In formula I above, desirably, R^3, R^4, R^5, R^6, R^7 and R^8 may be independently selected from C_{1-4} alkyl.

[0019] An example of a commercially available hydroxy-functionalized polydiorganosiloxane suitable for reaction with a C_1 tri-functional silane is hydroxy-terminated dimethoxy polydimethylsiloxane ("PDMS"), as represented by Formula I(a):



I(a)

[0020] The number of repeating units, "n" plays a role in determining the molecular weight and hence the viscosity of the reactive compositions of the invention, particularly because the end product of the reaction oftentimes has substantially the same viscosity as the silanol-terminated reactant. Thus, n may be, for example, an integer from about 10 to about 3500. The viscosity may be readily chosen for a particular product application. Viscosities of such hydroxy-functionalized polydiorganosiloxanes are often within the range of from about 10cps to about 300,000 cps. Desirably, the viscosity range for those siloxanes used in the present invention may be from about 40 cps to about 100,000 cps. A particularly suitable viscosity for those siloxanes used in the present invention is about 6,000 cps and having a molecular weight of about 50,000 AMU.

[0021] C_1 tri-functional silanes suitable for use as starting materials in the first reaction step described above include, but are not limited to, methacryloxymethyltrimethoxysilane, methacryloxymethyltriethoxysilane, acetoxymethyltrimethoxysilane, acetoxymethyltriethoxysilane, methylcarbamatomethyltrimethoxysilane and methylcarbamatomethyltriethoxysilane.

[0022] As described above, the reaction of the hydroxy-functionalized polydiorganosiloxane with the C_1 tri-functional silane in the first step occurs in the presence of a catalyst. Suitable catalysts include organo-lithium compounds, amines and combinations thereof. Examples of suitable organo-lithium compounds include, but are not limited to: methyl lithium; n-butyl lithium; sec-butyl lithium; t-butyl lithium; n-hexyl lithium; 2-ethylhexyl lithium; n-octyl lithium; phenyl lithium; vinyl lithium; lithium phenylacetylide; lithium (trimethylsilyl) acetylide; lithium dimethylamide; lithium diethylamide; lithium diisopropylamide; lithium dicyclohexylamide; lithium silanolate; lithium siloxanolate; and combinations thereof. Organo-lithium compounds are described in more detail in U.S. Patent Nos. 5,300,608, 5,498,642, 5,516,812 and 5,663,269 (assigned to Henkel Corporation), which are incorporated herein by reference in their entirety.

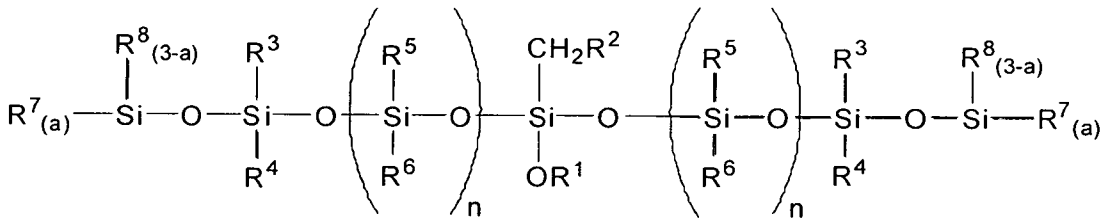
[0023] Examples of suitable amine catalysts include, but are not limited to: tetramethylguandineamines, diazabicyclo[5.4.0]undec-7-ene (DBU) and triethylamine and the like and combinations thereof.

[0024] Any of the catalysts provided herein may be present in amounts of about 0.0001% to about 0.5% by weight of the composition.

[0025] This first reaction step proceeds at a temperature of about 20°C to about 80°C, desirably about 60°C, with dynamic vacuum.

Second Reaction Step

[0026] In accordance with the methods of the present invention, the chain-extended polyorganosiloxane of formula II, which was produced in the first reaction step, then is reacted with a moisture curable silane in a second reaction step to form a final reactive polymer having the following general formula III:



III

[0027] In formula III, desirably, R^7 is selected from alkyl, alkenyl, alkynyl and methacryloxy; R^8 is selected from amino, oximino, alkoxy, enoxy, acetoxy; and a is 0 or 1. All remaining groups are as defined above.

[0028] Moisture curable silanes suitable for use in this second reaction step include any silanes containing moisture curing groups. Particularly suitable moisture curable silanes include tri-functional and tetra-functional silanes. Examples of suitable silanes include, but are not limited to: alkoxy silanes; acetoxy silanes; enoxy silanes; oximino silanes; amino silanes; and

combinations thereof. One particularly suitable example of a moisture curable silane is vinyl trimethoxysilane.

[0029] In this second reaction step, desirably, a molar excess of the moisture curable silane is used. Specifically, the ratio of the moisture curable silane to the chain-extended polyorganosiloxane of formula II may be about 2:1 to about 100:1. Desirably, the ratio of the moisture curable silane to the chain-extended polyorganosiloxane of formula II may be about 10:1. In some embodiments, the moisture curable silane may be present in amounts of about 0.01% to about 10% by weight of the reactive composition. Accordingly, the moisture curable silane acts to end-cap the chain-extended polyorganosiloxane of formula II to produce the final reactive polymer of formula III. The final reactive polymer of formula III contains moisture-curing groups and thereby is curable upon exposure to moisture at ambient conditions. Additionally, the molecular weight of the final reactive polymer of formula III has been increased by the reaction steps described herein.

[0030] The second reaction step proceeds at a temperature of about 15°C to about 120°C, desirably about 20 to 50°C, with dynamic vacuum.

[0031] The methods of the present invention also may include the step of adding a cure system to the reactive polymer of formula III to form a curable instant seal composition. The cure system added in this method step includes, but is not limited to, catalysts or other reagents which act to accelerate or otherwise promote the curing of the composition of the invention. Due to the presence of moisture curing groups, compounds of formula III have the capability of curing by moisture curing mechanisms. Accordingly, the cure system in the compositions of the present invention may include a moisture curing, or condensation, catalyst. Any suitable moisture curing catalyst may be used, such as, organic titanium compounds, organic tin compounds, organic amines, combinations thereof, or any other known catalyst for moisture-curing silicones.

[0032] More specifically, suitable moisture curing catalysts include, but are not limited to, tin IV salts of carboxylic acids, such as dibutyltin dilaurate, organotitanium compounds such

as tetrabutyl titanate, and partially chelated derivatives of these salts with chelating agents such as acetoacetic acid esters and β-di-ketones. Desirably, titanium alkoxide, dibutyl tin laurate or alkyl tin carboxylate are used. Additionally, organic amines such as tetramethylguandinemaines, diazabicyclo[5.4.0]undec-7-ene (DBU) and triethylamine and the like may be used.

[0033] Moisture curing catalysts may be present in amounts of about 10% or less by weight of the composition, more desirably about 0.01% to about 1.0% by weight of the composition and most desirably about 0.05% to about 0.5% by weight of the composition.

[0034] A variety of optional components also may be added in preparing the reactive compositions. For instance, fillers optionally may be added to the compositions in accordance with the present invention. Generally, any suitable mineral, carbonaceous, glass, or ceramic filler may be used, including, but not limited to: fumed silica; clay; metal salts of carbonates, such as calcium carbonate; sulfates; phosphates; carbon black; metal oxides; titanium dioxide; ferric oxide; aluminum oxide; zinc oxide; quartz; zirconium silicate; gypsum; silicium nitride; boron nitride; zeolite; glass; plastic powder; stearic acid; and combinations thereof. The filler may be present in the composition in any suitable concentration in the curable composition. Generally, concentrations of from about 5% to about 80 % by weight of the composition are sufficient. However, a more desirable range would be 20-60%.

[0035] Among the more desirable fillers are reinforcing silicas. The silica may be a fumed silica, which may be untreated (hydrophilic) or treated with an adjuvant so as to render it hydrophobic. The fumed silica should be present at a level of at least about 5% by weight of the composition in order to obtain any substantial reinforcing effect. Although optimal silica levels vary depending on the characteristics of the particular silica, it has generally been observed that the thixotropic effects of the silica produce compositions of impractically high viscosity before maximum reinforcing effect is reached. Hydrophobic silicas tend to display lower thixotropic ratios and therefore greater amounts can be included in a composition of desired consistency. In choosing the silica level, therefore, desired reinforcement and practical viscosities must be balanced. A hexamethyldisilazane treated silica is particularly desirable (HDK2000 by Wacker-Chemie, Burghausen, Germany).

[0036] Adhesion promoters also may be added to the reactive compositions. An adhesion promoter may act to enhance the adhesive character of the curable composition for a specific substrate (i.e., metal, glass, plastics, ceramic, and blends thereof). Any suitable adhesion promoter may be employed for such purpose, depending on the specific substrate elements employed in a given application. Various organosilane compounds may be desired.

[0037] Suitable organosilane adhesion promoters include, for example, 3-aminopropyltriethoxysilane, 3-aminopropyltrimethoxysilane, 3-aminopropylmethyldiethoxysilane, 3-aminopropylmethyldimethoxysilane, methylaminopropyltrimethoxysilane, 1,3,5-tris(trimethylsilylpropyl)isocyanurate, 3-glycidoxypropyltrimethoxysilane, 3-glycidoxypropylethyldimethoxysilane, 2-glycidoxyethyltrimethoxysilane, 2-cyanoethyltrimethoxysilane, 3-cyanopropyltriethoxysilane, isocyanatopropyltriethoxysilane, isocyanatopropyltrimethoxysilane, gamma-ureidopropyltrimethoxysilane, tris(3-(trimethoxysilyl) propyl) isocyanurate (commercially available under the trade name SILQUEST A-LINK 597 from General Electric Company), and combinations thereof.

[0038] Adhesion promoters, when present, may be used in amounts of about 0.1% to about 10% by weight of the composition. Desirably, the adhesion promoter is present from about 0.2% to about 2.0% by weight of the composition.

[0039] Alcohol scavengers also may be added to the reactive compositions. Examples of suitable alcohol scavengers for use in the methods of the present invention include, but are not limited to, hexamethyldisilazane and vinyltrienoxysilane.

[0040] Condensation catalysts also may be added to the reactive compositions. Examples of suitable condensation catalysts for use in the methods of the present invention include, but are not limited to, dialkyltindicarboxylates, dibutyltindilaurate, dimethyltindicarboxylate and dioctyltindicarboxylate.

[0041] Any number of other optional additives also may be added to the compositions, such as pigments or dyes, plasticizers, stabilizers, anti-oxidants, flame retardants, UV-stabilizers, biocides, fungicides, thermal stabilizing agents, rheological additives, tackifiers, thickeners, such as tris copoly(oxy-1-methylene) oxyethylene, hydrogen bonding aids, moisture scavengers, and the like or combinations thereof.

[0042] The reactive compositions prepared as described herein, including a base-bodied reactive polymer of formula III, a cure system, and any of the optional additives, exhibit characteristics that provide instant sealability when applied to substrates. In particular, as set forth above, the compositions have a blow-out resistance of greater than about 30 seconds at 4 psi. The compositions also have a high thixotropic ratio (viscosity at $0.5s^{-1}$/viscosity $5.0s^{-1}$) of greater than 4.0. The compositions have a viscosity at $5s^{-1}$ of less than 1,500,000 cps. The extrusion rate of the compositions is greater than 15 grams/minute. In view thereof, the compositions are particularly suitable for form-in-place gaskets in automotive applications, such as, for example, automotive powertrain applications.

[0043] The present invention also is directed to methods of forming gasket assemblies. In accordance with such methods, a reactive composition, as described above, is applied onto at least one of two substrate surfaces. The reactive composition includes a reactive polymer of formula III, as described above, and a cure system. Any of the optional additives set forth herein also may be included in the reactive composition. Once the reactive composition is applied onto the substrate surface(s), it is exposed to moisture to effectuate at least partial cure. The substrate surface(s) then are mated together in an abutting relationship to form the gasket assembly. Due to the increased molecular weight of the reactive polymer of formula III, the composition provides instant sealability upon application to the substrate surfaces(s). As mentioned above, such instant sealability may be particularly desirable in certain gasketing applications, such as, for example, automotive applications.

[0044] The present invention additionally is directed to methods for sealing together two substrates. In accordance therewith, a reactive composition, as described above, is applied onto at least one of two substrate surfaces. The reactive composition includes a final reactive polymer

of formula III, as described above, and a cure system. Any of the optional additives set forth herein also may be included in the reactive composition. The two substrate surfaces subsequently are mated in an abutting relationship to form an assembly. Thereafter, the reactive composition is exposed to moisture at ambient conditions. The abutting relationship of the surfaces is maintained for a time sufficient to allow the reactive composition to cure.

EXAMPLES

Example 1:

[0045] A reactive composition was prepared in accordance with the methods of the present invention. The components listed in Table 1 below were combined in the indicated amounts as described in more detail below.

TABLE 1

Component	Weight %
Hydroxy-terminated PDMS[1]	42.0
Ethyl silicate	0.5
Methacryloxymethyltrimethoxysilane	0.7
1,8-diazabicyclo[5.4.0]undec-7-ene (DBU)	0.05
Vinyl trimethoxysilane	3.0
Amorphous hydrophobic silica	3.45
Polymer additive filler[2]	20.0
Tris copoly(oxy-1-methylethylene) oxyethylene	1.0
Iron aluminum silicate	18.0
Carbon black	1.0
Calcium carbonate	10.0
3-aminopropyl trimethoxysilane	0.3

[1] Includes two different hydroxy-terminated PDMS materials
[2] Contains calcium carbonate, polybutene and hexamethyldisilazane

[0046] In accordance with the first reaction step described herein, two hydroxy-terminated PDMS materials were added to a mixing vessel and mixed for 15 minutes under a nitrogen atmosphere while heating to a temperature of 60°C. At temperature, a mixture of ethyl silicate and methacryloxymethyltrimethoxysilane, which is a C_1 tri-functional silane, was added. The mixture was mixed for 5 minutes under a nitrogen atmosphere and then DBU catalyst was added. The mixture was mixed for 30 minutes under a nitrogen atmosphere with medium shear

at 60 ±5°C. Methacryloxymethyltrimethoxysilane chain-extended the hydroxy-terminated PDMS as described in the first reaction step of the present invention.

[0047] Subsequently, vinyl trimethoxysilane, which is a moisture curable silane, was added under a nitrogen atmosphere. The mixture was mixed for 15 minutes at medium shear under vacuum while cooling. In accordance with the second reaction step described herein, vinyl trimethoxysilane end-capped the chain-extended polyorganosiloxane, thereby forming a final reactive polymer having moisture curable groups.

[0048] Amorphous hydrophobic silica then was added, mixed at low shear until wetted in and then mixed for 15 minutes at high shear under a nitrogen atmosphere. Tris copoly(oxy-1-methylethylene) oxyethylene, which functions as a thickener, and the polymer additive filler were added to the mixture. The polymer additive filler contained calcium carbonate, which is a filler, polybutene, which is a rubber/elastomer, and hexamethyldisilazane, which is a moisture scavenger. The mixture then was mixed for 10 minutes at medium shear under a nitrogen atmosphere. Iron aluminum silicate and carbon black were added and mixed with low shear under a nitrogen atmosphere until wetted in. The mixture then was mixed at medium shear for 15 minutes under dynamic vacuum. Calcium carbonate, which is a filler, was added and mixed at low shear under a nitrogen atmosphere until wetted in. The mixture then was mixed at high shear under dynamic vacuum for 30 minutes and cooled if necessary to maintain a temperature less than 50°C. 3-aminopropyl trimethoxysilane, which functions as an adhesion promoter, was added and mixed for 15 minutes at medium shear under dynamic vacuum while cooling.

[0049] A cure system, including a moisture curing catalyst, was added to the composition. The cure system included 0.2% by weight of dioctyltindicarboxylate (available from Witco Polymer Additives Fomrez UL-38). The composition was subjected to moisture-curing conditions (7 day cure at 100°C and 50% relative humidity) and the following properties were measured: hardness; tensile strength; elongation; and blow out resistance. The initial and post-cure results are provided in Table 2 below. With respect to blow-out resistance, only initial results are provided in Table 2, as the standard testing method for blow-out resistance is only

performed prior to cure. Blow-out resistance was measured in accordance with the standard testing method described above using a 1.0 mm gap.

TABLE 2

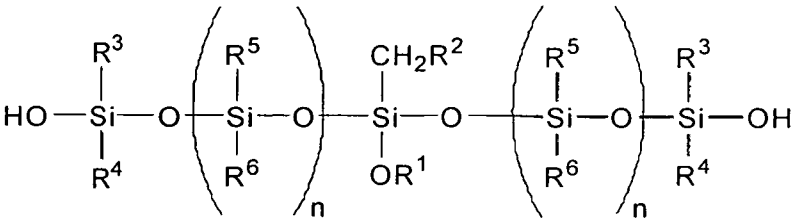
Test	Initial	After 7 day cure at 100°C and 50% RH
Shore A hardness	40	4
Tensile strength (psi)	217 ± 9	33 ± 8
at 100%	167	25
Elongation (%)	158 ± 3	173 ± 31
Blow-out resistance	62 sec at 2 psi	-

[0050] A comparative composition also was prepared and tested for blow-out resistance. The comparative composition was essentially the same as the inventive composition, except it was not prepared in accordance with the method described herein. Specifically, the comparative composition did not include the DBU catalyst, and thus, did not include the two-step reaction process described herein. The comparative composition had a blow-out resistance of 0 seconds when tested in accordance with the standard testing method described herein using the same parameters as the inventive composition (i.e., at 2 psi and a 1.0 mm gap). These results evidence that the reactive composition prepared in accordance with the methods of the present invention was strong, had high elongation and a high resistance to blow-out. Therefore, the composition would provide instant sealability when applied to seal together two substrates.

WHAT IS CLAIMED IS:

1. A method of preparing a reactive composition, comprising the steps of:

(a) reacting a hydroxy-terminated polydiorganosiloxane with a C_1 tri-functional silane, wherein the ratio of said hydroxy-terminated polydiorganosiloxane to said C_1 tri-functional silane is about 2:1 to about 1:0.8, in the presence of a catalyst to form a chain-extended polyorganosiloxane having the formula II:

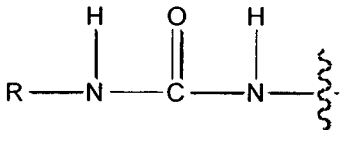
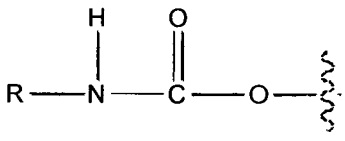


II

wherein :

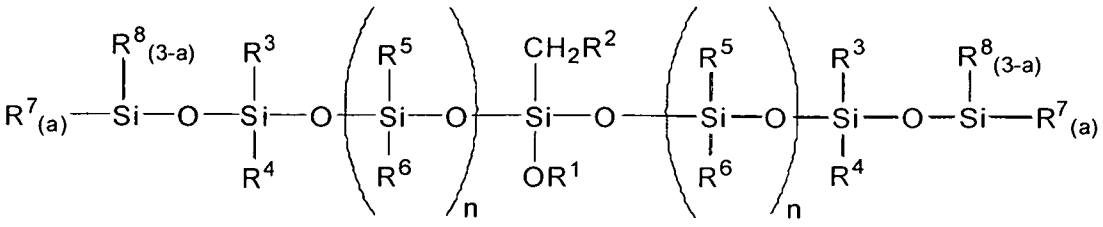
R^1, R^3, R^4, R^5 and R^6 are independently selected from C_{1-4} alkyl;

R^2 is selected from (meth)acryloxy, acetoxy, alkylcarbamato, amino, acetate,


and

; and

n is from 10 to 3500; and

(b) reacting the chain-extended polyorganosiloxane of formula II with a moisture curable silane to form a reactive polymer having the formula III:



III

wherein:

R^7 is selected from alkyl, alkenyl, alkynyl and methacryloxy;

R^8 is selected from aminoxy, oximino, alkoxy, enoxy, acetoxy; and

a is 0 or 1.

2. The method of claim 1, wherein said hydroxy-terminated polydiorganosiloxane comprises hydroxy-terminated polydimethylsiloxane.

3. The method of claim 1, wherein said C_1 tri-functional silane is selected from the group consisting of methacryloxymethyltrimethoxysilane, methacryloxymethyltriethoxysilane, acetoxymethyltrimethoxysilane, acetoxymethyltriethoxysilane, methylcarbamatomethyltrimethoxysilane and methylcarbamatomethyltriethoxysilane.

4. The method of claim 1, wherein said catalyst is selected from the group consisting of an organo-lithium compound, an amine and combinations thereof.

5. The method of claim 1, wherein step (b) comprises a molar excess of said moisture curable silane.

6. The method of claim 5, wherein the ratio of said moisture curable silane to said polyorganosiloxane of formula I is about 2:1 to about 100:1.

7. The method of claim 5, wherein the ratio of said moisture curable silane to said polyorganosiloxane of formula I is about 10:1.

8. The method of claim 1, wherein said moisture curable silane of step (b) is present in amounts of about 0.01% to about 10% by weight of said reactive composition.

9. The method of claim 1, wherein said moisture curable silane is selected from the group consisting of tri-functional silanes and tetra-functional silanes.

10. The method of claim 1, wherein said moisture curable silane is selected from the group consisting of: alkoxy silanes; acetoxy silanes; enoxy silanes; oximino silanes; amino silanes; and combinations thereof.

11. The method of claim 1, wherein said moisture curable silane comprises vinyl trimethoxysilane.

12. The method of claim 1, further comprising the step of:

(c) adding a cure system to form a curable instant seal composition.

13. The method of claim 12, wherein said cure system comprises a moisture curing catalyst selected from the group consisting of: organic titanium compounds; organic tin compounds; organic amines; and combinations thereof.

14. A method of forming a gasket assembly, comprising the steps of:

(a) applying the curable instant seal composition of claim 12 to at least one of two substrate surfaces;

(b) exposing the composition to moisture to effectuate at least partial cure; and

(c) mating the substrate surfaces in abutting relationship to form the gasket assembly.

15. A method for sealing together two substrates, comprising the steps of:

(a) applying the curable instant seal composition of claim 12 to at least one of two substrate surfaces;

(b) mating the substrate surfaces in abutting relationship to form an assembly;

(c) exposing the composition to moisture; and

(d) maintaining the abutting relationship for a time sufficient to allow the composition to cure.

16. A curable instant seal composition comprising:

(a) a reactive composition comprising: at least one hydroxy-terminated polydimethylsiloxane; ethyl silicate; methacryloxymethyltrimethoxysilane; 1,8-

diazabicyclo[5.4.0]undec-7-ene; vinyl trimethoxysilane; amorphous hydrophobic silica; calcium carbonate; polybutene; hexamethyldisilazane; tris copoly(oxy-1-methylethylene) oxyethylene; iron aluminum silicate; carbon black; 3-aminopropyl trimethoxysilane; and

(b) a cure system comprising dioctyltindicarboxylate.