PATENT SPECIFICATION

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(54) LOW WATER VAPOR TRANSMISSION RATE SILICONE COMPOSITION

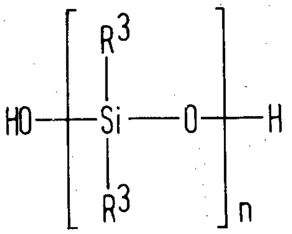
(71) We, GENERAL ELECTRIC COMPANY, a corporation organised and existing under the Laws of the State of New York, United States of America, residing at 1 River Road, Schenectady 12305, State of New York, United States of America, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention relates to a room temperature vulcanizable silicone rubber composition and more particularly the present invention relates to a room temperature vulcanizable silicone rubber composition which has a low water vapor transmission rate.

Room temperature vulcanizable silicone rubber compositions are well known, particularly, the two component or two part room temperature vulcanizable silicone rubber compositions. Such compositions generally comprise a silanol end-stopped diorganopolysiloxane as the base polymer, a reinforcing filler selected from precipitated silica and fumed silica and optionally an extending filler such as zinc oxide, iron oxide, diatomaceous earth, quartz, ground quartz, etc. Normally the silanol end-stopped diorganopolysiloxane is mixed with the filler and is packaged separately. In the other package or component there is provided a cross-linking agent which can be an alkyl silicate or a partial hydrolysis product of such a silicate. Optionally, as an alternative cross-linking agent, there can be present in the composition a silicone resin composed of monofunctional units and tetrafunctional units. There must be also present in the composition a metal salt of a carboxylic acid as a catalyst to speed up the rate of reaction of the composition to form a silicone elastomer. Usually the cross-linking agent and the catalyst are packaged in a separate package or component. Either package can have additional ingredients as long as the cross-linking agent and the catalyst are kept separate from the silanol end-stopped diorganopolysiloxane polymer. When it is desired to cure the composition, the two components are mixed together to form a homogeneous mixture which then cures in a period of time varying anywhere from a half hour to 24 hours to obtain the desired silicone elastomer. Such two component room temperature vulcanizable silicone rubber compositions are well known as encapsulant sealants. Normally such two component room temperature vulcanizable silicone rubber compositions accomplish their sealant function especially for the sealing of glass panes and partitions to masonry and other types of base structure material with desirable efficiency. However, they have one disadvantage in that as previously formulated they have a high water vapor transmission rate. Such prior art two-component room temperature vulcanizable silicone rubber compositions have a water vapor transmission rate of 40 to 50 grams per square meter in a 70 mils. thick section of silicone elastomer. As stated previously, for most applications this did not create a problem. However, the high water vapor transmission rate was exceedingly undesirable when the silicone material was used to enclose a sealed area in which water vapor accumulation was a problem. The need for such low water vapor transmission rate for silicone sealants was even more noticeable where it was desired to fabricate insulated window panes so that the glass would be insulated by the air pocket between the panes. Thermopanes are the best glass insulated glass partitions known in the industry. To produce thermopanes two glass sheets are hermetically sealed to each other with a layer of air in between the panes which is completely dry such that moisture cannot permeate and enter the air barrier between the glass panes. The manufacture of such thermopanes is expensive, which makes them unduly costly for insulation in high rise buildings. Accordingly, in a recent innovation in the industry a facsimile of thermopanes is produced by taking two clear glass panes and sealing the edges with a sealant such as a polysulfide sealant, which polysulfide sealant is again applied to the sealed glass pane so as to seal the insulated window pane to the frame of the building. Such polysulfide sealants have a water vapor transmission rate of about 15 grams per meter square of 70 mils thick sealant. Although suitable to some extent the polysulfide water vapor transmission rate as noted above is still a little too high. Accordingly, in some cases when such fabricated insulated glass panes are tested by cycling them from a low temperature of −60°F to 150°F there is invariably too much moisture that has migrated through the sealant into the air space between the glass panes and as a result the glass panes will fog up with water vapor. As an improvement there has been inserted between the two panes of the insulated glass construction a butyl rubber tape having a desiccant therein over which there is applied the polysulfide sealant so as to maintain a low water vapor transmission rate into the air space between the glass panes. The use of silica gel or micro sieves or another desiccant with the butyl rubber tape and the polysulfide sealant results in a combination that has a low water vapor transmission rate. However, such construction of insulated glass panes is very expensive. Furthermore, the construction utilizing the butyl tape with the desiccant therein is still undesirable in that polysulfides do not have as good weatherability as is experienced with silicones. Further, as can be envisioned there could be utilized a butyl rubber tape between the two glass panes with prior art silicone rubber compositions. However, in some cases too much moisture is transmitted into the air pocket between the glass panes. In addition, the silicone-butyl rubber tape combination cannot be utilized for sealing the insulated glass panes to the frame of high rise buildings. Accordingly, it would be highly desirable to have a low water vapor transmission rate silicone sealant.

According to the present invention there is provided a two-component, low water vapor transmission rate, room temperature vulcanizable silicone composition, wherein one component comprises (a) 100 parts by weight of a silanol-terminated diorganopolysiloxane polymer having a viscosity from 100 to 500,000 centipoise at 25°C wherein the organic groups are selected from monovalent hydrocarbon radicals and halogenated monovalent hydrocarbon radicals (b) from 75 to 150 parts by weight of a vapor barrier filler comprising mica; and (c) 0 to 30 parts by weight of reinforcing or extending filler; and wherein the other component comprises (d) from 1 to 15 parts by weight of a cross-linking agent which is a compound of the formula $R_a Si (OR^1)_{4-a}$ or a partial hydrolysis product thereof, where R and R^1 are selected from monovalent hydrocarbon radicals and halogenated monovalent hydrocarbon radicals, and *a* is 0 or 1, or a resinous copolymer of $R_3^2\ SiO_{0.5}$ units and SiO_2 units in a weight ratio of .5 : 1 to 1 : 1, where R^2 is selected from monovalent hydrocarbon radicals and halogenated monovalent hydrocarbon radicals or mixtures of such cross-linking agents; and (e) from .01 to 5 parts by weight of an organotin oxide or a metal salt of carboxylic acid as a catalyst for promoting cross-linking between ingredients (a) and (d), when mixed, where the metal is selected from lead to manganese in the electrochemical series. Preferably the vapor barrier filler has a particle size varying from 50 to 4000 US Standard mesh. With respect to the reinforcing and extending fillers these are added to the two component silicone sealant disclosed above solely for the purpose of giving the uncured silicone composition sag control. In addition to these ingredients, there may be added triorganosiloxy- and silanol-terminated polysiloxane polymers of low viscosities so as to increase the plasticity of the composition. There may also be added as an optional ingredient a low viscosity silanol-terminated diorganopolysiloxane polymer to slow down the rate of cure of the composition once the two components are mixed. The additive polymer not only increases the modulus of the final composition and speeds up the cure, but also facilitates the mixing of the one part to the other part. In addition, there may be added to this composition the normal two part room temperature vulcanizable silicone rubber composition additives, such as adhesion promoters. The basic aspect of the present invention lies in the utilization in the instant compositions of at least from 75 to 150 parts of the vapor barrier filler and more preferably 100 to 150 parts by weight of the vapor barrier filler, which is mica, which may be dry or wet base mica and which preferably has a particle size in the range of 50 to 4000 U.S. Standard mesh.

In one type of room temperature vulcanizable silicone rubber composition in accordance with the invention there are present 100 parts by weight of a linear diorganopolysiloxane polymer which is silanol end-stopped, having a viscosity from 100 to 500,000 centipoise at 25°C. and having organo groups selected from monovalent hydrocarbon radicals and halogenated monovalent hydrocarbon radicals. Such organo groups can be selected from alkyl radicals such as methyl, ethyl and propyl, cycloalkyl radicals such as cyclohexyl, mononuclear aryl radicals such as phenyl, methylphenyl and ethylphenyl, alkenyl radicals such as vinyl and allyl, and halogenated monovalent hydrocarbon radicals, such as fluoroalkyl radicals, such as trifluoropropyl. Preferably the organo groups are selected from alkyl radicals, alkenyl radicals, phenyl radicals and fluoroalkyl radicals of 1 to 8 carbon atoms. The preferred silanol end-stopped diorganopolysiloxanes are linear polymers that have the formula



$$HO-\left[\begin{array}{c} R^3 \\ | \\ Si-O \\ | \\ R^3 \end{array}\right]_n-H \quad (1)$$

wherein R^3 has the same definition as the foregoing organo radicals and is most preferably selected from alkyl radicals, alkenyl radicals, phenyl radicals, and fluoroalkyl radicals of 1 to 8 carbon atoms, and wherein *n* is from 150 to 1500. Linear silanol end-stopped diorganopolysiloxanes are well known in the art and they may be prepared for instance by polymerizing cyclictetrasiloxanes with the desired substituent groups in the presence of a toluene sulfonic acid catalyst or another mild acid catalyst. They can be prepared for instance by equilibrating cyclictetrasiloxanes of the desired substituting groups in the presence of an alkali metal catalyst with the appropriate chain stoppers to obtain high molecular weight linear diorganopolysiloxanes, that is polysiloxanes having a viscosity of 500,000 centipoise to 200,000,000 centipoise at 25°C. The chain stoppers in such equilibration procedures are normally selected from triorganosiloxy end-stopped low molecular weight siloxanes with the organo groups the same as defined above. The triorganosiloxy chain stopped linear diorganopolysiloxane polymer of high molecular weight can then be taken and there is added to it water and/or steam and the resulting high molecuar linear diorganopolysiloxane polymer is then broken down to a low molecular weight linear diorganopolysiloxane polymer having silanol terminal units and having a viscosity within the required 100 to 500,000 centipoise at 25°C range. A more preferred procedure for obtaining such linear silanol end-stopped diorganopolysiloxane polymers such as those in formula 1 above, is to take the hydrolyzate equilibrated cyclictetrasiloxanes containing the preferred substituent groups with a low molecular weight silanol end-stopped diorganopolysiloxane polymer to obtain the silanol terminated diorganopolysiloxane polymer of the preferred or desired viscosity within the range given above. The low molecular weight silanol-terminated diorganopolysiloxane polymer may be obtained by simply hydrolyzing diorganodichlorosilanes and refluxing the resulting hydrolyzate to separate the low molecular weight silanol end-stopped diorganopolysiloxane polymers that are formed in such a hydrolyzate. To 100 parts of this polymer there are generally added 75 to 150 parts by weight of the vapor barrier filler which is mica. Further, such silanol end-stopped polymers as discussed above are those for instance disclosed in the Specification of U.S. Patent No. 3,888,815. This is true also for many of the other ingredients of the present composition. As stated previously, the necessary filler ingredient in the present compositions is a mica filler, which can either be a wet ground mica or a dry ground mica. As stated previously generally from 75 to 150 parts by weight of mica must be used per one hundred parts of the linear diorganopolysiloxane polymer. More preferably, from 100 to 150 parts of the mica filler is utilized per 100 parts by the weight of the base silanol end-stopped linear diorganopolysiloxane polymer. It has been found that the inclusion of the mica filler in the foregoing quantities provides the present compositions with resistance to penetration by water vapor. It should be noted that if less than 75 parts by weight of mica is utilized in the present composition, the desired resistance to penetration by water vapor is not achieved. When there are utilized more than 150 parts of mica, the additional amount of over 150 parts although providing additional resistance to the composition to moisture penetration, does not provide a marked improvement over the case where 150 parts of mica filler is utilized in the composition. Along with the mica filler there may be utilized from 0 to 30 parts and preferably from 5 to 20 parts by weight per 100 parts by weight of the linear silanol end-stopped diorganopolysiloxane of a reinforcing or extending filler. It should be noted that preferably 5 to 20 parts by weight of additional filler such as the traditional reinforcing or extending fillers be utilized in the composition. The reinforcing fillers are well known in the art and constitute principally precipitated silica and fumed silica having a particle size from 50 to 400 U.S. Standard mesh in size. The extending fillers may be utilized in place of or in mixtures with the reinforcing fillers, and such extending fillers may be selected from a large class of materials including titanium dioxide, lithapone, zinc oxide, zirconium silicate, silica aero-gel, iron oxide, diatomaceous earth, calcium carbonate, glass fibers, magnesium oxide, chromic oxide, zirconium oxide, aluminium oxide, quartz, clay, asbestos, carbon, graphite, quartz carbon and synthetic fibers. In the most preferred composition some additional filler is utilized in addition to the mica filler. It is preferred that the additional filler be of the reinforcing type such as fumed silica or precipitated silica. Sufficient quantities of the reinforcing filler are utilized in the composition as to impart to it sag control properties, that is, the composition upon being applied to a vertical surface will stay there without seeping or moving to a great extent prior to cure. The mica can be untreated or can be treated with cyclicpolysiloxanes or fatty acids such as oleic acid. The reinforcing filler or extending fillers may also be treated with cyclicpolysiloxanes. The advantage of treating the fillers is that the treated fillers do not unduly increase the viscosity of the composition at the same concentrations. It should also be noted that treated fillers are also desirable in that they result in better sag control. These fillers alone with the silanol end-stopped linear diorganopolysiloxane polymer constitute the basic first component of the present two-component composition.

The second component comprises generally from 1 to 15 parts by weight based on a hundred parts of the silanol end-stopped linear diorganopolysiloxane of a cross-linking agent which is a compound of the formula

$$R_a\ Si\ (OR^1)_{4-a} \qquad (2)$$

or a partial hydrolysis product thereof, where R and R^1 are selected from monovalent hydrocarbon radicals and halogenated monovalent hydrocarbon radicals and *a* is 0 or 1. Generally, the organo substituting groups as disclosed above for R and R^1 are same as the organo groups of the silanol end-stopped linear diorganopolysiloxane polymer. In Formula (2) above *a* can be either 0 or 1. Either type of alkoxylated silane would provide the proper cross-linking in the present composition. It should also be noted as stated above that partial hydrolysis products of such compounds may also be utilized as cross-linking agents in the two component room temperature vulcanizable silicone rubber composition of the instant case. Such compounds and partial hydrolysis products of such compounds are well known in the art, as for instance set forth in the Specification of the aforesaid U.S. Patent No. 3,88,815. Such compounds may be utilized by themselves as cross-linking agents, or there may be utilized in place of them as a cross linking agent a resinous copolymer composed of $R_2^3\ SiO_{0.5}$ units and SiO_2 units in a weight ratio of 0.5 : 1 to 1 : 1 where R^2 is selected from monovalent hydrocarbon radicals and halogented monovalent hydrocarbon radicals and mixtures thereof. The R^2 radical may generally represent the same groups as those given above for the R^3 radicals, as well as the R^1 groups for the silicates. Most preferably, as stated previously, the R,R^1,R^3,R^2 substituting group or selected from alkyl radicals, phenyl radicals, alkenyl radicals, fluoroalkyl radicals such as trifluoropropyl and mixtures thereof. Such substituting groups preferably do not have more than 8 carbon atoms. Such resinous copolymers as cross-linking agents for two component room temperature vulcanizable silicone rubber compositions are described in the Specification of U.S. Patent No. 3,457,214. It should be also noted that in some cases it is possible to utilize as a cross-linking agent a mixture of two or more of the compound of formula (2), the partial hydrolysis product thereof and the resinous copolymer. Such cross-linking agents are utilized at a concentration of 1 to 15 parts by weight based on the hundred parts of the silanol end-stopped linear diorganopolysiloxane polymer. More preferably there is utilized 1 to 10 parts by weight of such cross-linking agents either alone or as mixtures thereof. Further, there must be present in the composition from .01 to 5 parts by weight based on a hundred parts of the silanol end-stopped linear diorganopolysiloxane polymer of a catalyst for promoting cross-linking between ingredients (a) and (d) selected from organotin oxides and metal salts of a carboxylic acid, which may be a dicarboxylic acid, where the metal is selected from lead to manganese in the electrochemical series. Preferred catalysts are dibutyl tin oxide and dibutyl tin dilaurate. The catalyst is mixed with the cross-linking agent and kept separate from the base polymer which usually contains the filler ingredients in it. When it is desired to cure the composition the two components are mixed immediately prior to usage and then applied or moulded to form the desired sealant bead and the mixed silicone composition is then cured to a silicone elastomer over a period of time varying from 1/2 hour to 12 hours. However, if the composition is formulated to have a high speed of cure the period during which the composition can be worked is 10 minutes after being mixed and applied to provide the proper sealant function. Other ingredients for room temperature vulcanizable silicone rubber compositions can be utilized in the present composition; for instance the nitrogen-containing silanes described in the Specification of the aforesaid U.S. Patent No. 3,888,815 can be utilized in the composition to provide a self-bonding two component room temperature vulcanizable silicone rubber composition which composition will bond in a superior manner to most substrates, e.g. to metal substrates, concrete, glass, and various plastics. In addition to the other two ingredients mentioned above in the present two component room temperature vulcanizable silicone rubber composition there may be utilized from 5 to 35 parts by weight based on a hundred parts of silanol end-stopped linear diorganopolysiloxane polymer, of an organopolysiloxane polymer having triorganosiloxy terminal units and silanol terminal units where the ratio of triorganosiloxy terminal units to silanol terminal units is from 1 to 1 to 1 to 10 and which polymer has a viscosity from 50 to 1000 centipoise at 25°C and where the organo groups are preferably selected from alkyl radicals, vinyl radicals, phenyl radicals and fluoroalkyl radicals. This ingredient, which is preferably mixed with the silanol end-stopped based polymer, slows down the cure and increases the modulus of the silicone elastomer or sealant that is formed from the composition. It should be noted that this polymer species also has some trifunctional polymer species in the polymer mixture. This ingredient does not have to be utilized with the present composition. However, it is advantageously utilized to produce a cured silicone sealant for insulated window panes with lower modulus and improved tear strength. Such polymers are well known in the silicone art and are simply prepared by hydrolyzing with water a mixture of diorganodichlorosilanes and monoorganotrichlorosilanes as is well known in the art.

Another optional and additional ingredient which facilitates the deep section curing of the present two component room temperature vulcanizable silicone rubber composition is a low molecular weight silanol-terminated diorganopolysiloxane polymer. Such low molecular weight silanol-terminated diorganopolysiloxane polymer may be utilized at a concentration of 2 to 10 parts by weight of the base polymer and has a viscosity from 100 to 500 centipoise at 25°C, the organo groups being again preferably selected from alkyl radicals, alkenyl radicals, phenyl radicals and fluoroalkyl radicals of from 1 to 8 carbon atoms. Such low molecular weight silanol-terminated diorganopolysiloxane polymers are preferably formed from the hydrolysis of diorganodichlorosilanes. The use of such polymers in room temperature vulcanizable silicone rubber compositions is well known and is described in the Specification of U.S. Patent No. 3,845,161. Such low molecular weight silanol terminated diorganopolysiloxane polymer is preferably mixed in the base polymer. If it were mixed with the cross-linking ingredient it would be polymerized and not function effectively as a deep section curing agent in the composition. Another optional ingredient that may be utilized in the present composition is for instance from 2 to 10 parts by weight of a triorganosiloxy-terminated linear diorganopolysiloxane polymer having a viscosity from 5 to 500 centipoise at 25°C wherein the organo groups are again selected from alkyl radicals, alkenyl radicals, phenyl radicals and fluoroalkyl radicals of 1 to 8 carbon atoms. Such linear diorganopolysiloxane polymer having a low molecular weight is simply utilized as a plasticizing agent in the composition. It is preferably mixed with the catalyst and the cross-linking agent to facilitate the mixing of the catalyst in the base composition so that the composition will have a uniform cure rate throughout the composition.

The composition is applied after the two components are mixed and applied to form the sealant function for forming insulated glass panes or performing any other sealant or encapsulating function.

Triorganosiloxy-terminated diorganopolysiloxane polymers of low molecular weight and a viscosity in the range specified above as well the low molecular weight silanol terminated diorganopolysiloxane polymers are well known in the art, as well as to silicone producers, and can simply be produced in one case by the hydrolysis of diorganodichlorosilanes and the other case by the equilibration of cyclictetrasiloxanes with a large amount of chain stopper composed of triorganosiloxy terminal units such as hexamethyldisiloxane, etc. The self-bonding additives disclosed in the foregoing Specifications can be utilized at a concentration of .01 to 10 parts by weight based on 100 parts of the silanol end-stopped linear diorganopolysiloxane base polymer, said self-bonding additives being nitrogen-containing silanes or siloxanes as indicated above. The preferred self-bonding additive for use in the present composition is gamma aminopropyltriethoxy silane and mixtures of the various gamma aminopropyltriethoxy silanes. The self-bonding additive is preferably mixed in the base composition but it can be mixed with the cross-linking agent and the catalyst. In addition, other ingredients may be utilized in the present composition as may be necessary to achieve the desired properties in the final composition for a specific utility. However, generally, the present invention concerns two component room temperature vulcanizable silicone rubber compositions which have in them as a basic filler, a mica filler and which result in a cured silicone elastomer with desirable resistance to water vapor transmission. Other fillers disclosed previously as reinforcing silica fillers are utilized solely for imparting to the composition consistency prior to cure and making it sag resistant. However, such reinforcing fillers perform no function in resisting the passage of moisture through the composition. It should be noted that it has been found that two part room temperature vulcanizable silicone rubber composition having in them the normal reinforcing and extending fillers disclosed above and no mica, have a water vapor transmission rate of 40 to 50 grams per meter square for test samples 69 to 70 mils thick.

In the Examples below there was used the ASTM E—96, —E—66, moisture vapor test under condition E. This ASTM Test is carried out in an outer dish which is filled to a marked degree with potassium salts and water into which there is placed a Petri dish with the sample of a uniform thickness over the Petri dish and with a desiccant inside the Petri dish such that there is 0% humidity in the atmosphere of the Petri dish which is covered by the silicone sample or other sample. There is then placed a cover over the outer dish such that the potassium salts and water at the bottom of the outer dish maintain an atmosphere at 90% relative humidity. The resulting enclosed outer dish containing the Petri dish and sample in it is then placed in an oven maintained at 100°F and a run is made for two weeks. The moisture pick up in the silicone sample or other sample is measured every 24 hours. The average reading for a 24 hour period during the second week of readings is then taken as the value for the moisture pick up of the sample. The invention will now be illustrated by the following Examples in which all parts are by weight.

EXAMPLE I.

There was prepared a base composition comprising 800 parts by weight of a silanol-terminated dimethylpolysiloxane polymer having a viscosity of 3000 centipoise at 25°C, 200 parts by weight of a trimethylsiloxy silanol end-stopped methylpolysiloxane polymer having a viscosity of 600 centipoise at 25°C, 100 parts by weight of octamethylcyclictetrasiloxane-treated fumed silica and 810 parts by weight of water-ground mica 325 U.S. Standard mesh. To 100 parts of the foregoing base composition which is hereafter referred to as component A there were mixed 10 parts of component B which was formed by mixing 33 parts by weight of a trimethylsiloxy end-stopped dimethylpolysiloxane polymer having a viscosity of 10 centipoise at 25°C, 4.0 parts by weight of gamma aminopropyltriethoxy silane, 2.04 parts of partially hydrolyzed ethyl silicate and .68 parts of dibutyl tin oxide. The resulting composition had the following properties after cure:

Tensile strength psi	400
Elongation %	50
Shore A hardness	60

The resulting composition also in the Lap Sheer Test with Tinius Olsen Apparatus had an extension rate of .5 inches per minute. Its adhesion on stainless steel screen on aluminium of 70 pounds per inch with 5% cohesive failure. Its adhesion on aluminium plus glass screen with 1/4 inch bond line was 52 lbs. per inch and 35% cohesive failure.

Further Component A was catalyzed with 10 parts per 100 parts of component A of a catalyst system prepared by mixing 22.5 parts of a gamma aminopropyltriethoxy silane, 11.26 parts of partially hydrolyzed ethyl silicate, 3.75 parts of tin oxide which was modified with a phthalate and 62.48 parts of mineral spirits. The adhesion of the cured composition was measured on an aluminium screen on aluminium which gave a value in one test of 105 pounds per inch with 50% of cohesive failure and a value of 83 pounds per inch with 50% cohesive in the second test. In another test with an aluminium with a stainless steel screen test the composition gave a result of 80 pounds per inch plus 60% cohesive failure. When the ASTM E—96—66 Test condition E was applied to a sample of this material 69 mls thick the sample had a water vapor transmission rate of 8.63 grams per square meter.

EXAMPLE II.

There were mixed wtih 480 parts of silanol end-stopped dimethylpolysiloxane having a viscosity of 4200 centipoise at 25°C, 120 parts of trimethylsiloxy end-stopped, silanol end-stopped polysiloxane oil, 18 parts of a cyclicsiloxane-treated fumed silica and 480 parts of 160 U.S. Standard mesh mica which had been treated with 4% by weight of stearic acid. To 100 parts of the above composition there were added 10 parts of a catalyst composition prepared by mixing 102 parts of a vinyl terminated dimethylpolysiloxane having a viscosity of 3,000 cps at 25°C, 120 parts of a $Ca\ Co_3$, 48 parts of gamma aminopropyltriethoxysilane, 24 parts of partially hydrolyzed ethyl silicate and 3.6 parts of dibutyl tin dilaurate.

Sample sheets of the composition which had cured for 24 hours at 25°C gave the following properties and water vapor transmission rate (WVTR):

Tensile strength psi	330
Elongation %	70
Shore A hardness	62

WVTR—8.25 grams/M^2 on sheet 74 mils thick.

EXAMPLE III.

There were mixed into 800 parts of silanol end-stopped dimethylpolysiloxane having a viscosity of 600 centipoise at 25°C, 200 parts of a trimethylsiloxy, end-stopped, silanol end-stopped methylpolysiloxane oil, 30 parts of cyclicsiloxane-treated fumed silica and 800 parts of 160 U.S. Standard mesh mica. To 100 parts of the above composition there were added 10 parts by weight of the catalyst composition of Example II. The resulting sample sheets which were cured for 24 hours at 25°C had the following properties and water vapor transmission rate:

Tensile psi	310
Elongation %	60
Shore A Hardness	60

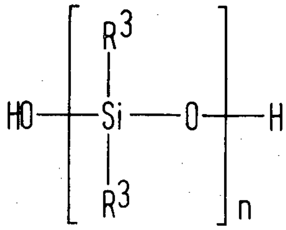
WVTR = 9.32 grams/M^2 on a sheet 76 mils thick.

Corresponding one-component compositions are the subject matter for our copending Application No. 47374/77 (Serial No. 1,590,814).

WHAT WE CLAIM IS:—

1. A two-component, low water vapor transmission rate, room temperature vulcanizable silicone composition, wherein one component comprises (a) 100 parts by weight of a silanol-terminated diorganopolysiloxane polymer having a viscosity from 100 to 500,000 centipoise at 25°C wherein the organo groups are selected from monovalent hydrocarbon radicals and halogenated monovalent hydrocarbon radicals; (b) from 75 to 150 parts by weight of a vapor barrier filler comprising mica; and (c) 0 to 30 parts by weight of a reinforcing or extending filler; and wherein the other component comprises (d) from 1 to 15 parts by weight, based on the weight of ingredient (a), of a cross-linking agent which is a compound of the formula R_a Si $(OR^1)_{4-a}$ or a partial hydrolysis product thereof, where R and R^1 are selected from monovalent hydrocarbon radicals and halogenated monovalent hydrocarbon radicals, and *a* is 0 or 1, or a resinous copolymer of R_3^2 $SiO_{0.5}$ units and SiO_2 units in a weight ratio of 0.5:1 to 1:1, where R^2 is selected from monovalent hydrocarbon radicals and halogenated monovalent hydrocarbon radicals, or mixtures of such cross-linking agents; and (e) from 0.01 to 5 parts by weight of an organotin oxide or a metal salt of a carboxylic acid as a catalyst for promoting cross-linking between ingredients (a) and (d), when mixed, where the metal is selected from lead to manganese in the electrochemical series.

2. The composition of Claim 1 wherein the diorganopolysiloxane has the formula,



$$HO\left[\begin{array}{c} R^3 \\ | \\ Si-O \\ | \\ R^3 \end{array}\right]_n H$$

wherein R^3 is selected from alkyl radicals, alkenyl radicals, phenyl radicals and fluoroalkyl radicals of 1 to 8 carbon atoms, and *n* is from 150 to 1500.

3. The composition of Claim 1 or 2 wherein said vapor barrier filler has a particle size of 50 to 4000 U.S. Standard mesh.

4. The composition of any preceding Claim wherein there is present from 5 to 20 parts by weight of a reinforcing filler selected from ground silica and precipitated silica.

5. The composition of any preceding Claim wherein there is additionally present from 5 to 35 parts by weight of a diorganopolysiloxane polymer having triorganosiloxane and silanol terminal units and a viscosity from 50 to 1000 centipoise at 25°C, the organo groups being selected from alkyl radicals, vinyl radicals, phenyl radicals and fluoroalkyl radicals.

6. The composition of any preceding Claim wherein said composition additionally comprises from 2 to 10 parts by weight of silanol terminated diorganopolysiloxane polymers having a viscosity varying from 100 to 500 centipoise at 25°C, the organo groups being selected from alkyl radicals, alkenyl radicals, phenyl radicals and fluoroalkyl radicals of 1 to 8 carbon atoms.

7. The composition of any preceding Claim wherein said composition additionally comprises from 2 to 10 parts by weight of triorganosiloxy terminated diorganopolysiloxane polymer having a viscosity from 5 to 500 centipoise at 25°C, the organo groups being selected from alkyl radicals, alkenyl radicals, phenyl radicals and fluoroalkyl radicals of 1 to 8 carbon atoms.

8. The composition of any preceding Claim wherein said composition contains from 0.01 to 10 parts by weight of a self-bonding additive.

9. The composition of Claim 8 wherein the self-bonding additive is a nitrogen-containing silane or siloxane.

10. A process for forming a low water vapor transmission rate, room temperature vulcanizable silicone composition comprising (1) mixing a first component comprising (a) 100 parts by weight of a silanol-terminated diorganopolysiloxane polymer having a viscosity from 100 to 500,000 centipoise at 25°C wherein the organo groups are selected from monovalent hydrocarbon radicals and halogenated monovalent hydrocarbon radicals; (b) from 75 to 150 parts by weight of a vapor barrier filler comprising mica; and (c) 0 to 30 parts by weight of a reinforcing or extending filler; with a second component comprising (d) from 1 to 15 parts by weight, based on the weight of ingredient (a), of a cross-linking agent consisting of R_a $Si(OR^1)_{4-a}$ or a partial hydrolysis product thereof, where R and R^1 are selected from monovalent hydrocarbon radicals and halogenated monovalent hydrocarbon radicals, and *a* is 0 or 1, or a resinous polymer of R_3^2 $SiO_{0.5}$ units and SiO_2 units, in a weight ratio of 0.5:1 to 1:1, where R^2 is selected from monovalent hydrocarbon radicals and halogenated monovalent hydrocarbon radicals or mixtures of such cross-linking agents; and (e) from 0.01 to 5 parts by weight of an organotin oxide or a metal salt of a carboxylic acid as a catalyst for promoting cross-linking between ingredients (a) and (d), when mixed, wherein the metal is selected from lead to manganese in the electrochemical series and (2) allowing the resulting mixture to cure at room temperature.

11. A two-component low water vapor transmission rate, room temperature vulcanizable silicone composition substantially as

hereinbefore described in any one of the foregoing Examples.

12. A process for forming a low water vapor transmission rate, room temperature vulcanizable silicone composition substantially as hereinbefore described in any one of the foregoing Examples.

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