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(54) Title: CURABLE SILICONE COMPOSITION OR SILICONE COMPOSITION KIT INCLUDING BLOCKED POLYISOCYANATE COMPOSITION, PRODUCTION METHOD FOR OBTAINING INTEGRALLY MOLDED PRODUCT WITH BASE MATERIAL, AND RESULTING INTEGRALLY MOLDED PRODUCT

(57) Abstract: The present invention provides a silicone composition being a one-liquid curable composition including a blocked polyisocyanate composition, or a silicone composition kit in which the curable silicone compositions including the blocked polyisocyanate compositions are distributed at least in two liquids as different components and preserved, which shows no increase in the viscosity after mixing the blocked polyisocyanate into the silicone composition, has no blocking agent being released from isocyanate during storage, and is capable of promoting the thermal dissociation during curing and expressing the adhesion properties simultaneously with the completion of the curing. In the silicone composition being a one-liquid curable composition including the blocked polyisocyanate composition or the silicone composition kit in which the curable silicone compositions including the blocked polyisocyanate compositions are distributed at least in two liquids as different components and preserved, a thermally dissociating blocking agent component included in the blocked polyisocyanate composition has a vapor pressure of not less than a predetermined lower limit value and not more than a predetermined upper limit value.



CURABLE SILICONE COMPOSITION OR SILICONE COMPOSITION KIT
INCLUDING BLOCKED POLYISOCYANATE COMPOSITION, PRODUCTION
METHOD FOR OBTAINING INTEGRALLY MOLDED PRODUCT WITH BASE
MATERIAL, AND RESULTING INTEGRALLY MOLDED PRODUCT

5

TECHNICAL FIELD

[0001]

The present invention relates to a one-liquid curable
silicone composition including a blocked polyisocyanate
composition or a silicone composition kit in which the curable
10 silicone compositions including the blocked polyisocyanate
compositions are distributed at least in two liquids as
different components and preserved, both of which being
characterized in that a thermally dissociating blocking agent
component included in the blocked polyisocyanate composition
15 has the vapor pressure of not less than the predetermined
lower limit value and not more than the predetermined upper
limit value.

BACKGROUND ART

[0002]

20 In order to bond a silicone composition to metal, a resin,
a fiber, or the like, a so-called self-adhesive silicone
composition having adhesion properties has been developed and
directly bonded to an adherend serving as a base material.
Further, the silicone composition is conventionally mixed with
25 various additives for imparting self-adhesion properties to
the silicone composition. For example, Patent Literature 1
discloses a curable organopolysiloxane compound mixed with an
organosilicon compound having an epoxy group and an organic
titanium compound. Patent Literature 2 discloses a method for
30 performing adhesion to a polycarbonate resin, a polybutylene
terephthalate resin, a polyphenylene sulfide resin, or the
like in combination with an organosilicon compound and an
organometallic compound. Further, Patent Literature 3
discloses a method for performing strong adhesion also to
35 metal by mixing an organic compound having an isocyanate group.
[0003]

However, the isocyanate group, which is highly active and thus has a high adhesion effect on various base materials, has the disadvantage of being easily inactivated. Further, the isocyanate group is inactivated by reacting with water, various substances having active hydrogen, and the like present in the system, and thus the usable environment or situation has been limited. Further, the isocyanate group sometimes inactivates a curing catalyst included in the curable silicone composition or reacts with a silicone composition component. This makes it difficult to mix the organic compound including the isocyanate group into the silicone composition in advance and store it as a silicone composition kit for a long period of time. Thus, when the silicone compositions are mixed in the actual process, the organic compound including the isocyanate group needs to be separately measured and mixed in a total of three liquid preparation lines, resulting in disadvantages in the process in terms of labor and cost. Further, an organic solvent is used to prevent difficulty in mixing, and viscosity rise, poor dispersion, and separation after mixing, thereby causing an environmentally unfavorable situation.

[0004]

In order to solve the aforementioned problems relating to reactivity caused by the use of the organic compound including the isocyanate group, there is proposed a blocked isocyanate composition, that is, a composition in which the isocyanate compound is reacted or associated with a blocking agent that is an organic substance. In such a composition, the isocyanate group is covered and protected by the blocking agent at a certain temperature or lower to control the reactivity of the isocyanate group and the blocking agent is dissociated under a certain temperature to expose the isocyanate group and express the intrinsic adhesion properties of the isocyanate group.

[0005]

Specifically, for example, Patent Literature 4 proposes a

blocked polyisocyanate. The blocking agent is a thermally dissociating compound, and examples thereof include an alkylphenol-based compound, an oxime-based compound, an alcohol-based compound, an acid amide-based compound, an acid imide-based compound, a phenol-based compound, an amine-based compound, an imine-based compound, an active methylene-based compound, an active ethylene-based compound, a mercaptan-based compound, a urea-based compound, an imidazole-based compound, and a pyrazole-based compound. These compounds suppress the reaction by blocking the isocyanate group under the room temperature but are dissociated from the isocyanate groups by heating to expose the isocyanate groups and promote the reaction. However, Patent Literature 4 never shows findings on the preparation of a silicone composition kit and the stability of the blocked isocyanate in the silicone composition. Many of these blocking agents have insufficient compatibility with silicone.

[0006]

When the aforementioned blocked polyisocyanate is used in the silicone composition, there is no index for determining the timing of thermal dissociation of the blocking agent and the timing of dissipation of the thermally dissociated blocking agent, making it practically difficult to maintain the balance between storage stability, curing properties, and self-adhesion properties.

[0007]

That is, up to this time, there is no one-liquid curable silicone composition including the blocked polyisocyanate composition, or the silicone composition kit in which the curable silicone compositions including the blocked polyisocyanate compositions are distributed at least in two liquids as different components and preserved, which shows no increase in the viscosity after the mixing into the silicone composition, has no blocking agent being released from isocyanate during storage, and is capable of promoting the thermal dissociation during curing and expressing the adhesion

properties simultaneously with the completion of the curing.

CITATION LIST

PATENT LITERATURE

[0008]

5 Patent Literature 1: Japanese Patent Application Laid-
Open No. Hei. 6-145525

 Patent Literature 2: Japanese Patent Application Laid-
Open No. 2005-290312

 Patent Literature 3: Japanese Patent Application Laid-
10 Open No. 2011-99090

 Patent Literature 4: Japanese Patent Application Laid-
Open No. 2016-216557

SUMMARY OF INVENTION

TECHNICAL PROBLEM

15 [0009]

 The present invention has been made in view of the above
circumstances, and an object of the present invention is to
provide a silicone composition being a one-liquid curable
composition including a blocked polyisocyanate composition, or
20 a silicone composition kit in which the curable silicone
compositions including the blocked polyisocyanate compositions
are distributed at least in two liquids as different
components and preserved, which shows no increase in the
viscosity after mixing the blocked polyisocyanate into the
25 silicone composition, has no blocking agent being released
from isocyanate during storage, and is capable of promoting
the thermal dissociation during curing and expressing the
adhesion properties simultaneously with the completion of the
curing.

30 SOLUTION TO PROBLEM

[0010]

 The present inventors have intensively conducted studies.
As a result, they have found that the afore mentioned problems
can be solved by using a blocked polyisocyanate composition in
35 which a thermally dissociating blocking agent having a vapor
pressure of not less than a predetermined value and not more

than a predetermined value is selected, thereby completing the present invention.

[0011]

That is, the present invention provides a silicone
5 composition being a one-liquid curable composition including a blocked polyisocyanate composition or a silicone composition kit in which the curable silicone compositions including the blocked polyisocyanate compositions are distributed at least
10 in two liquids as different components and preserved, both of which being characterized in that a thermally dissociating blocking agent component included in the blocked polyisocyanate composition has a vapor pressure of not less than a predetermined lower limit value and not more than a predetermined upper limit value.

15 [0012]

It is preferable that the lower limit value of the vapor pressure at 20°C of the aforementioned blocking agent is 0.05 mmHg and the upper limit value thereof is 40 mmHg.

[0013]

20 It is preferable that the aforementioned blocked polyisocyanate composition is substantially free from an organic solvent and has a viscosity at 60°C of 100,000 mPa·s or lower, and that the blocked polyisocyanate composition includes a polyisocyanate obtained from at least one kind of
25 diisocyanate selected from the group consisting of an aliphatic diisocyanate and an alicyclic diisocyanate, and the thermally dissociating blocking agent.

[0014]

The aforementioned curable silicone composition is
30 preferably an addition-curable silicone composition.

[0015]

Further, the present invention provides a production method for obtaining an integrally molded product with a base material, wherein the integrally molded product is obtained by
35 curing the silicone composition or the silicone composition kit, and the resulting integrally molded product.

[0016]

Further, the present invention provides a method for producing an integrated product utilizing self-adhesion of a silicone composition, the method including: a step of
5 selecting a blocking agent in accordance with a curing method and a curing temperature for curing a curable silicone composition including a blocked polyisocyanate composition such that a viscosity of a blocked polyisocyanate composition and a thermal dissociation temperature and a vapor pressure of
10 a blocking agent fall within respective predetermined ranges; a step of preserving a one-liquid composition or a silicone composition kit having at least two separated liquids by including the blocked polyisocyanate composition in which the selected blocking agent is used; and a step of curing the
15 silicone composition while thermal dissociation of the blocking agent is initiated by mixing and heating the silicone composition kit.

ADVANTAGEOUS EFFECTS OF INVENTION

[0017]

20 In the curable silicone composition or the silicone composition kit of the present invention, the timing of the thermal dissociation and dissipation of the blocking agent can be determined by using the vapor pressure of the blocking agent as an index, and thus the object of the present
25 invention is to provide the silicone composition or the silicone composition kit, which shows no increase in the viscosity after mixing the blocked polyisocyanate into the silicone composition, has no blocking agent being released from isocyanate during storage, and is capable of promoting
30 the thermal dissociation during curing and expressing the adhesion properties simultaneously with the completion of the curing.

[0018]

Further, the curable silicone composition or the silicone
35 composition kit of the present invention has the low viscosity due to easy dispersion of the blocked polyisocyanate

composition, shows no increase in the viscosity after the mixing, and is free from the solvent, thereby making it possible to take advantage of the intrinsic characteristics of the blocked polyisocyanate. As a result, it can be easily
5 produced, used in an automated facility, and processed in a safe work environment. Further, since the blocking agent is not released during storage, there is little variation in the adhesion properties at the time of use, which can significantly improve the productivity and also the product
10 quality.

DESCRIPTION OF EMBODIMENTS

[0019]

The curable silicone composition or the silicone composition kit including the blocked polyisocyanate
15 composition according to the present invention will be described in detail below.

[0020]

The curable silicone composition of the present invention may have any form and composition, as long as it has at least
20 one liquid or more and can be cured by reaction to obtain a cured product of the silicone composition as a final product. Specific examples of the curing method may include peroxide curing, condensation curing, addition curing, ultraviolet curing, and electron beam curing. Examples of the preferable
25 curing method of the present invention may include a method of crosslinking an Si-H group of an organohydrogenpolysiloxane to an organopolysiloxane having an alkenyl group bonded to a silicon atom in one molecule by an addition reaction catalyst, or by an organic peroxide, and a method of crosslinking an Si-
30 H group of an organohydrogenpolysiloxane to an organopolysiloxane having a hydroxy group or an alkoxy group bonded to a silicon atom at both terminals of the molecular chain by a condensation reaction catalyst. The addition curing is preferable from the viewpoints of productivity and
35 easy handleability.

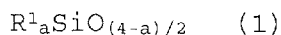
[0021]

As specific component constituents of addition curing, an organopolysiloxane containing an alkenyl group bonded to a silicon atom in one molecule as a component (A) which is a main agent, an organohydrogenpolysiloxane containing a
5 hydrogen atom bonded to a silicon atom as a component (B) which is a crosslinking agent, a component (C) as a curing catalyst, and a blocked polyisocyanate composition as a component (D) for imparting adhesion properties thereto are essential components.

10 [0022]

(Component (A))

A more specific component (A) is an organopolysiloxane containing, on average, 1.8 or more alkenyl groups bonded to a silicon atom in one molecule, and the average composition
15 formula is usually represented by the following general formula (1).



20 (In the formula (1), R^1 's which are the same as or different from each other are each an unsubstituted or substituted monovalent hydrocarbon group having 1 to 18 carbon atoms; and a is 1.7 to 2.1.)

[0023]

25 Here, at least two or more of the monovalent hydrocarbon groups represented by the afore mentioned R^1 are selected from an alkenyl group such as a vinyl group, an allyl group, a propenyl group, an isopropenyl group, a butenyl group, an isobutenyl group, a hexenyl group, and a cyclohexenyl group.
30 Other groups are a substituted or unsubstituted monovalent hydrocarbon group having 1 to 18 carbon atoms, and are specifically selected from an alkyl group such as a methyl group, an ethyl group, a propyl group, an isopropyl group, a butyl group, an isobutyl group, a tert-butyl group, a pentyl
35 group, a neopentyl group, a hexyl group, a 2-ethylhexyl group, a heptyl group, an octyl group, a nonyl group, a decyl group,

and a dodecyl group; a cycloalkyl group such as a cyclopentyl group, a cyclohexyl group, and a cycloheptyl group; an aryl group such as a phenyl group, a tolyl group, a xylyl group, a biphenyl group, and a naphthyl group; an aralkyl group such as
5 a benzyl group, a phenylethyl group, a phenylpropyl group, and a methylbenzyl group; and a halogen-substituted alkyl group and a cyano-substituted alkyl group in which a portion or the entirety of hydrogen atoms in each of these hydrocarbon groups is substituted with a halogen atom, a cyano group, and the
10 like, such as a chloromethyl group, a 2-bromoethyl group, a 3,3,3-trifluoropropyl group, a 3-chloropropyl group, and a cyanoethyl group.

[0024]

In selection of R^1 , a vinyl group is preferable as two or
15 more alkenyl groups required, and a methyl group, a phenyl group, and a 3,3,3-trifluoropropyl group are preferable as the other groups. In addition, 70 mol% or more of all R^1 's is preferably a methyl group from the viewpoint of properties of a cured product and economic efficiency. Usually, those having
20 a methyl group in an amount of 80 mol% or more of all R^1 's are used.

[0025]

The organopolysiloxane of the component (A) may be linear or branched. Examples of the molecular structure may include a
25 dimethylpolysiloxane capped at both molecular terminals with dimethylvinylsiloxyl groups, a dimethylsiloxane-methylphenylsiloxane copolymer capped at both molecular terminals with dimethylvinylsiloxyl groups, a dimethylsiloxane-methylvinylsiloxane copolymer capped at both molecular
30 terminals with dimethylvinylsiloxyl groups, a dimethylsiloxane-methylvinylsiloxane-methylphenylsiloxane copolymer capped at both molecular terminals with dimethylvinylsiloxyl groups, a dimethylsiloxane-methylvinylsiloxane copolymer capped at both molecular terminals with trimethylsiloxyl groups, an
35 organopolysiloxane obtained by substituting a portion or the entirety of methyl groups of each of these organopolysiloxanes

with an alkyl group such as an ethyl group or a propyl group; an aryl group such as a phenyl group or a tolyl group; and a halogenated alkyl group such as a 3,3,3-trifluoropropyl group, and mixtures of two or more of these organopolysiloxanes. It is preferable to use a linear organopolysiloxane capped at both molecular terminals with vinyl groups from the viewpoint of availability.

[0026]

In order to improve compatibility with the blocked polyisocyanate composition and additives, those in which a main skeleton of the organopolysiloxane contains a phenyl skeleton or a phenylene skeleton may be used, and it is preferable that two or more of these skeleton components are contained in one molecule.

[0027]

The organopolysiloxane of the component (A) is produced by a method known to a person skilled in the art, and the viscosity at 25°C is preferably 50 to 1,000,000 mPa·s, more preferably 200 to 500,000 mPa·s, and in particular, the use of two or more kinds of organopolysiloxans having different viscosities is preferable because the viscosity of the final silicone composition is easily adjusted. The viscosity may be measured by a rotary viscometer or the like.

[0028]

(Component (B))

A more specific component (B) is an organohydrogenpolysiloxane having, on average, two or more hydrogen atoms bonded to a silicon atom in one molecule. Specific examples thereof may include a methylhydrogenpolysiloxane, a dimethylsiloxane-methylhydrogenpolysiloxane copolymer, a methylphenylsiloxane-methylhydrogenpolysiloxane copolymer, a cyclic methylhydrogenpolysiloxane, and a copolymer composed of a dimethylhydrogensiloxy unit and an $\text{SiO}_{4/2}$ unit. The mixed amount of the organohydrogenpolysiloxane is preferably an

amount in which the hydrogen atom bonded to the silicon atom is 0.5 to 20 mol with respect to the total of the alkenyl groups of the component (A). If the amount is less than 0.5 mol, the hardness of the cured product is remarkably lowered, and if the amount is more than 20 mol, the cured product becomes too hard, and a cured coating film is easily cracked or peeled off, which is not preferable.

[0029]

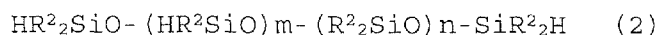
The viscosity of the organohydrogenpolysiloxane of the component (B) at 25°C is preferably 1 to 100,000 mPa·s, and more preferably 2 to 5,000 mPa·s. The component (B) may be used alone, or two or more kinds thereof may also be used in combination.

[0030]

In order to improve the elongation of the cured product, an organohydrogenpolysiloxane having a hydrogen atom bonded to a silicon atom only at both terminals of the molecular chain may be used. These are preferably linear, and the molecular chain length of the organopolysiloxane having the alkenyl group of the component (A) can be relatively easily increased by the curing reaction to give a high elongation to the cured product.

[0031]

In addition, from the viewpoint of elongation and adhesion properties, the component (B) may include those containing a hydrogen atom at both terminals of the molecular chain and those containing a hydrogen atom other than both terminals, and specifically, a linear organohydrogenpolysiloxane as represented by the following general formula (2) is exemplified.



In the formula (2), R²'s, which are the same as or different from each other and do not independently have an aliphatic unsaturated bond, are each an unsubstituted or

halogen-substituted monovalent hydrocarbon group having 1 to 10 carbon atoms, m is a positive number of 1 to 50, n is 0 or a positive number of 1 to 150, and t represented by the formula $t = m/(m + n)$ satisfies $0.01 \leq t \leq 1.0$. In the formula (2), m is more preferably 1 to 20, n is more preferably 10 to 100, t is $0.02 \leq t \leq 1.0$, and more preferably t is $0.02 \leq t \leq 0.2$. When m is 50 or more, the elongation at the time of fracture does not increase, and when n is 150 or more, the hardness of the cured product decreases, which is not preferable. When t is 0.01 or less, there is no addition effect, and when t is 0.1 or more, elongation at the time of fracture of the cured product does not increase, which is not preferable.

[0032]

Alternatively, a compound having a functional group imparting adhesion properties to an organohydrogenpolysiloxane may be used as the component (B). Specific examples of the functional group may include an epoxy group, a glycidoxy group, a (meth)acryloxy group, a methacrylic group, and an acrylic group.

[0033]

From the viewpoint of adhesion properties and heat resistance, the compound (B) may contain an organohydrogenpolysiloxane having a trimethylsiloxy group at both terminals of the molecular chain thereof and at least one aromatic group in the molecule, and for economic reasons, the aromatic group is preferably a phenyl group. Further, it is preferable to use them in combination with an organohydrogenpolysiloxane having a trimethylsiloxy group at both terminals of the molecular chain thereof and no aromatic group in the molecule, and having a hydrogen content of 5 mmol/g or more, because the adhesion properties are further enhanced.

[0034]

(Component (C))

The addition curing catalyst of the component (C) may be

any of those known in the art. Such catalysts are those known to a skilled person in the art which accelerate the addition curing reaction of an alkenyl group with a hydrogen atom bonded to a silicon atom. Specific examples of the catalysts may include a platinum group metal such as platinum, rhodium, palladium, osmium, iridium, and ruthenium, and particulate carrier materials (for example, activated carbon, aluminum oxide, and silicon oxide) to which the platinum group metal is fixed. Furthermore, examples of platinum compounds may include a platinum halide, a platinum-olefin complex, a platinum-alcohol complex, a platinum-alcoholate complex, a platinum-vinylsiloxane complex, dicyclopentadiene-platinum dichloride, cyclooctadiene-platinum dichloride, and cyclopentadiene-platinum dichloride.

[0035]

Metal compound catalysts other than noble metals may be used for economic reasons. Specific examples of iron catalysts for hydrosilylation may include an iron-carbonyl complex catalyst, an iron catalyst having a cyclopentadienyl group as a ligand, an iron catalyst having a terpyridine-based ligand, an iron catalyst having a terpyridine-based ligand and a bistrimethylsilyl methyl group, an iron catalyst having a bisiminopyridine ligand, an iron catalyst having a bisiminoquinoline ligand, an iron catalyst having an aryl group as a ligand, an iron catalyst having a cyclic or acyclic olefin group having an unsaturated group, and an iron catalyst having a cyclic or acyclic olefinyl group having an unsaturated group. Other examples of the metal compound catalysts for hydrosilylation may include a cobalt catalyst, a vanadium catalyst, a ruthenium catalyst, an iridium catalyst, a samarium catalyst, a nickel catalyst, and a manganese catalyst.

[0036]

The catalyst may also be used in a microencapsulated form, such as in a particulate solid. In this case, the particulate solid containing the catalyst and insoluble in the

organopolysiloxane is, for example, a thermoplastic resin (such as a polyester resin or a silicone resin). It is also possible to use the catalyst in the form of an inclusion compound, for example, in a cyclodextrin.

5 [0037]

Although the catalyst used is mixed in an effective amount corresponding to a curing temperature and a curing time desired in the present application, the concentration of the catalyst metal element relative to the total mass of the
10 silicone composition may usually be in the range of 0.5 to 1,000 ppm, more preferably in the range of 1 to 500 ppm, and even more preferably in the range of 1 to 100 ppm. If the mixed amount is less than 0.5 ppm, the curing may be remarkably slow or may not be cured. On the other hand, if it
15 exceeds 1,000 ppm, the cost rises, which is not economically preferable.

[0038]

A curing inhibitor may be used for the above-mentioned catalyst, and any conventionally known compound having a
20 curing inhibitory effect may be used. Examples thereof may include hydrazines, triazoles, phosphines, mercaptans, a phosphorus-containing compound such as triphenylphosphine, a nitrogen-containing compound such as tributylamine, tetramethylethylenediamine, and benzotriazole, a sulfur-
25 containing compound, an acetylene-based compound, a compound containing two or more alkenyl groups, a hydroperoxy compound, a maleic acid derivative, and a silane and a silicone compound having an amino group.

[0039]

30 More specific examples thereof may include various "ene-in" systems such as 3-methyl-3-pentene-1-in and 3,5-dimethyl-3-hexene-1-in; an acetylenic alcohol such as 3,5-dimethyl-1-hexin-3-ol, 1-ethynyl-1-cyclohexanol, and 2-phenyl-3-butyne-2-ol; a maleate and a fumarate such as known dialkyl, dialkenyl,
35 and dialkoxyalkyl fumarates and maleates; and a cyclovinylosiloxane.

[0040]

The vapor pressure at 20°C of these components is preferably 30 mmHg or less, more preferably 25 mmHg or less, and further preferably 20 mmHg or less. If the vapor pressure exceeds 30 mmHg, the amount evaporated during storage of the silicone composition or the silicone composition kit is increased, and the curing rate is increased when the composition/kit is used, or addition curing is locally progressed and hardness is likely to be varied, which is not preferable.

[0041]

(Component (D))

The blocked polyisocyanate composition of the component (D) is an essential component of the present invention for imparting adhesion properties. Conventional blocked polyisocyanate compounds are incompatible with the silicone composition, and therefore, it is necessary to mix an organic solvent to prevent separation and poor dispersion. However, the presence of an organic solvent is not preferable because it promotes evaporation of the blocking agent. In addition, the viscosity is increased due to strong intramolecular hydrogen bond of the urethane group and the urea group generated when the polyisocyanate as a raw material is reacted with the blocking agent, and therefore, the conventional blocked polyisocyanate compounds are difficult to handle.

[0042]

The blocked polyisocyanate composition is preferably a polyisocyanate obtained from at least one diisocyanate selected from the group consisting of an aliphatic diisocyanate and a cycloaliphatic diisocyanate because of the reduced yellowing of the final silicone cured product. Examples of the aliphatic diisocyanate may include butane diisocyanate, pentane diisocyanate, hexamethylene diisocyanate, trimethylhexamethylene diisocyanate, and lysine diisocyanate. Among these, hexamethylene diisocyanate is preferable from the viewpoint of easy industrial availability. These aliphatic

diisocyanates may be used alone, or two or more kinds thereof may also be used in combination. Examples of the alicyclic diisocyanate may include isophorone diisocyanate, hydrogenated xylylene diisocyanate, hydrogenated diphenylmethane

5 diisocyanate, and 1,4-cyclohexane diisocyanate. Among these, isophorone diisocyanate is preferable from the viewpoint of easy industrial availability. These alicyclic diisocyanates may be used alone, or two or more kinds thereof may also be used in combination.

10 [0043]

Either the aliphatic diisocyanate or the alicyclic diisocyanate may be used alone, or two or more aliphatic diisocyanate and alicyclic diisocyanate may be used in combination.

15 [0044]

The mixed amount of the blocked polyisocyanate composition is preferably 0.01 parts by mass to 5 parts by mass, and more preferably 0.02 parts by mass to 3 parts by mass, relative to 100 parts by mass of the organopolysiloxane
20 containing an alkenyl group bonded to a silicon atom in one molecule, which is the component (A). When the amount is 0.01 parts by mass or less, it is difficult to obtain an adhesive effect, and when the amount is 5 parts by mass or more, curing becomes slow, which is not preferable.

25 [0045]

The blocking agent is preferably thermally dissociative and is not particularly limited as long as it meets the object of the present invention. From the viewpoint of easy industrial availability, it is preferable to use one or a
30 plurality of compounds selected from an oxime-based compound, an alcohol-based compound, an acid amide-based compound, an acid imide-based compound, a phenol-based compound, an amine-based compound, an active methylene-based compound, an imidazole-based compound, and a pyrazole-based compound.

35 Specific examples thereof may include formaldoxime, acetaldoxime, acetoxime, methylethylketoxime, cyclohexanone

oxime, 2-propanol, n-butanol, sec-butanol, 2-ethyl-1-hexanol, 2-methoxyethanol, 2-ethoxyethanol, 2-butoxyethanol, acetamide, succinimide, maleimide, phenol, cresol, ethylphenol, diphenylamine, aniline, di-n-propylamine, isopropylethylamine, dimethyl malonate, diethyl malonate, methyl acetoacetate, ethyl acetoacetate, acetylacetone, 2-methylimidazole, pyrazole, 3-methylpyrazole, and 3,5-dimethylpyrazole.

[0046]

The thermal dissociation of the thermally dissociating blocking agent means that the blocking agent bound to the isocyanate group is dissociated by heat. The dissociation temperature differs depending on the structure of the blocking agent and is ranging, for example, from 40°C to 300°C. On the other hand, the curing temperature of the silicone composition is ranging from the room temperature to about 250°C, and thus the thermal dissociation temperature of the blocking agent is preferably within the range of the curing temperature of the silicone composition. In particular, when the addition curing is performed, a reaction inhibitor of the addition curing catalyst is also dissociated by heat. Thus, the thermal dissociation temperature of the blocking agent is preferably not lower than the thermal dissociation temperature of the reaction inhibitor so as not to inhibit the action of the addition curing catalyst.

[0047]

Further, the timing of the thermal dissociation can be adjusted in accordance with the application. For example, the timing may be designed to obtain powerful adhesion properties concurrently with curing by thermally dissociating the blocking agent from the isocyanate group approximately simultaneously with the curing of the silicone composition. Alternatively, the timing may be designed so as to preserve the blocked isocyanate group in the cured film by the blocking agent having the high thermal dissociation temperature even after the curing of the silicone composition proceeds and then express or promote the adhesion properties by thermally

dissociating the blocking agent at the time of reheating for performing secondary curing of the silicone cured product or the like.

[0048]

5 In order to adjust the timing of the thermal dissociation described above, in principle, it can be expected that the proper blocking agent is selected by judging the thermal dissociation temperature as the characteristics of the blocking agent. However, in practice, such data is not usually
10 available up to now. In particular, the curable silicone composition includes various additives for imparting functionality, making it difficult to accurately measure the thermal dissociation temperature in the actual reaction system. Furthermore, the thermal dissociation temperature is affected
15 by a difference in condition and thus cannot be used as an independent index.

 In order to solve this problem, the present inventors have found that the vapor pressure of the blocking agent component included in the blocking agent composition is useful
20 for designing the timing of the thermal dissociation.

 In order to achieve this object, the molecular weight, the viscosity, or the like of the blocking agent may be used as the index other than the vapor pressure. However, it has been found that the vapor pressure is the optimal index as the
25 vapor pressure has a strong correspondence relation with the thermal dissociation temperature, thereby serving as a rough index indicating how easily the blocking agent is dissipated to the outside of the system after dissociation, a universal value can be obtained regardless of the conditions, and the
30 wealth of data is available. Further, the temperature conditions for transportation and storage differ between using countries and regions, thus the vapor pressure is the useful index in designing.

[0049]

35 In the present invention, it has been found that the vapor pressure of the blocking agent component only needs be

not less than the predetermined lower limit value and not more than the predetermined upper limit value in order to achieve the aforementioned object by mixing the blocked polyisocyanate composition into the curable silicone composition.

5 If the vapor pressure is greater than the predetermined value, the amount of the blocking agent evaporated during storage of the silicone composition kit increases as well as the thermal dissociation of the blocking agent proceeds, and the isocyanate group is inactivated before being used for
10 adhesion, resulting in a decrease in the adhesion properties. Thus, it is not preferable. Further, if the vapor pressure is less than the predetermined value, the thermal dissociation of the blocking agent does not sufficiently proceed during curing by heating and the blocking agent is not sufficiently
15 dissipated to the outside of the system, resulting in insufficient adhesion.

[0050]

The predetermined lower limit value and predetermined upper limit value of the aforementioned vapor pressure is
20 preferably 0.05 mmHg and 40 mmHg at 20°C, respectively. This is because, when the vapor pressure is greater than 40 mmHg, the amount of the blocking agent evaporated during storage of the silicone composition kit increases as well as the thermal dissociation of the blocking agent proceeds, and the
25 isocyanate group is inactivated before being used for adhesion, resulting in a notable tendency of decreasing the adhesion properties. Further, when the vapor pressure is less than 0.05 mmHg, the thermal dissociation of the blocking agent does not sufficiently proceed during curing by heating and the blocking
30 agent is not sufficiently dissipated to the outside of the system, resulting in a notable tendency of not obtaining sufficient adhesion.

More preferably, the vapor pressure at 20°C is 0.07 mmHg or more and 30 mmHg or less, and further preferably 0.1 mmHg
35 or more and 20 mmHg or less.

[0051]

Even if the temperature of the thermal dissociation of the blocking agent is shifted more or less from the target temperature, a fine control of storage properties, curing properties, and self-adhesion properties can be practically performed by setting the vapor pressure of the blocking agent. Further, even if the thermal dissociation occurs earlier than intended, the isocyanate group can be protected to some extent if the blocking agent is not quickly dissipated and remains inside the system.

10 [0052]

The amount of the thermally dissociating blocking agent is preferably 1 mol or more relative to one mol of the isocyanate group in the polyisocyanate. In this manner, almost all of the isocyanate groups are blocked and stabilized in the silicone composition kit. On the other hand, if the amount of the thermally dissociating blocking agent becomes excessive, it inhibits the action of the catalyst used in addition curing and reduces the curing rate. Thus, the amount of the thermally dissociating blocking agent is preferably 5 mol or less, further preferably 3 mol or less, relative to one mol of the isocyanate group in the polyisocyanate. Further, in addition curing, the amount of the thermally dissociating blocking agent is preferably 10 mol or less, further preferably 5 mol or less, relative to one mol of metal atom in the addition curing catalyst. The amount exceeding 10 mol inhibits the addition curing reaction and is thus not preferable.

[0053]

The blocked polyisocyanate composition of the present invention does not substantially contain an organic solvent; however, the term "does not substantially contain" means that the content of the organic solvent in the blocked polyisocyanate composition is 5% by mass or less, and from the viewpoint of reducing the environmental load, 3% by mass or less is preferable, and 1% by mass or less is more preferable. In addition, the lower the viscosity, the better the compatibility with the silicone composition, and from the

viewpoint of ease of handling, the viscosity is preferably 100,000 mPa·s or less at 60°C, and more preferably 30,000 mPa·s or less at 60°C from the viewpoint of ease of mixing.

The viscosity may be measured using a Type E viscometer up to
5 25,600 mPa·s and using a Leometer (RS-1, Haake Technik) for the viscosity higher than 25,600 mPa·s.

Also, the rotor used may appropriately be selected according to viscosity.

[0054]

10 To further promote adhesion properties of the blocked polyisocyanate composition, an organosilicon compound having an epoxy group and a silicon atom-bonded alkoxy group in one molecule may be included. The epoxy group is preferably bonded to a silicon atom in the form of a glycidoxyalkyl group such
15 as a glycidoxypropyl group, and an epoxy-containing cyclohexylalkyl group such as a 2,3-epoxycyclohexylethyl group and a 3,4-epoxycyclohexylethyl group, and two to three epoxy groups may be contained in one molecule. Preferable examples of the silicon atom-bonded alkoxy group may include a
20 trimethylsilyl group, a triethylsilyl group, a methyldimethoxysilyl group, an ethyldimethoxysilyl group, a methyldiethoxysilyl group, a trialkylsilyl group such as an ethyldiethoxysilyl group, and an alkyl dialkoxysilyl group. As a functional group other than those described above, a
25 functional group selected from an alkenyl group such as a vinyl group, a (meth)acryloxy group, and a hydrosilyl group (SiH group) may be used.

[0055]

For the organosilicon compound having an epoxy group and
30 a silicon atom-bonded alkoxy group in one molecule described above, a silane coupling agent containing no epoxy group may be used in combination. As the organic functional group, one or a plurality of groups selected from a vinyl group, a methacrylic group, an acrylic group, and an isocyanate group
35 is preferable. Examples thereof may include a methacryloxysilane such as 3-

methacryloxypropyltrimethoxysilane and 3-methacryloxypropyltriethoxysilane, 3-trimethoxysilylpropylsuccinic anhydride, and a furandion such as dihydro-3-(3-(triethoxysilyl)propyl)-2,5-furandion. The
5 organic functional group may be bonded to the silicon atom via another group such as an alkylene group.

[0056]

The silicone composition of the present invention may further contain a component having a function as a
10 condensation promoter catalyst, and examples thereof may include one or more compounds selected from the group consisting of a metal alkoxide, a metal acid salt, and a metal chelate, wherein an element selected from B, Al, Ti, and Zr is adopted as a metal atom. Examples of such organometallic
15 compounds may include: a boron-based condensation catalyst such as boron isopropoxide; a titanium-based condensation promoter catalyst such as an organic titanium alkoxide such as tetraisopropyl titanate, tetranormalbutyl titanate, tetratertiarybutyl titanate, tetraoctyl titanate, and
20 tetrastearyl titanate, an organic titanium acylate such as titanium isostearate, and an organic titanium chelating compound such as diisopropoxy(acetylacetonate)titanium, diisopropoxy(ethylacetoacetate)titanium, tetraacetylacetonatetitanium, and titanium-1,3-
25 propanedioxybis(ethylacetoacetate); a zirconium-based condensation promoter catalyst such as an organic zirconium alkoxide such as tetraisopropyl zirconate, tetranormalbutyl zirconate, tetratertiarybutyl zirconate, tetraoctyl zirconate, and tetrastearyl zirconate, an organic zirconium acylate such
30 as zirconium isostearate, an organic zirconium chelating compound such as zirconium diisopropoxy(acetylacetonate), zirconium diisopropoxy (ethylacetoacetate), zirconium tetraacetylacetonate, zirconium tributoxyacetylacetonate, and zirconium butoxyacetylacetonate, and an oxozirconium compound
35 such as zirconium bis(2-ethylhexanoate) oxide and zirconium acetylacetonate(2-ethylhexanoate) oxide; and an aluminum-based

condensation catalyst such as an aluminum alkoxide such as aluminum triethylate, aluminum triisopropylate, and aluminum tri(sec-butylate), an aluminum chelating compound such as diisopropoxyaluminum (ethylacetoacetate) aluminum
5 tris(ethylacetoacetate) and aluminum tris(acetylacetonate), and an aluminum acyloxy compound such as hydroxyaluminum bis(2-ethylhexanoate).
[0057]

The above-mentioned component is preferable when used in
10 combination with the organosilicon compound because the effect is further promoted. Specific examples may include combinations of a titanium chelating compound and/or a zirconium chelating compound and/or an aluminum chelating compound for a methacryloxy group-containing organoalkoxysilane, and
15 combinations of a titanium chelating compound and/or a zirconium chelating compound and/or an aluminum chelating compound for dihydro-3-(3-(triethoxysilyl)propyl)-2,5-furandion.
[0058]

20 In the present invention, any conventionally known additive for silicone or silicone rubber may be used as a further optional component to the extent that the purpose is not impaired. Specific examples thereof may include a reinforcing filler, a non-reinforcing filler, a dispersant, a
25 viscosity adjusting material, a pigment, a dye, a heat resistance-imparting agent, a flame retardant, an antistatic agent, a conductivity-imparting agent, an airtightness improver, a radiation shielding agent, an electromagnetic wave shielding agent, a preservative, a stabilizer, an organic
30 solvent, a plasticizer, a fungicide, an organopolysiloxane containing one silicon atom-bonded hydrogen atom or one alkenyl group in one molecule and not containing any other functional group, and an organopolysiloxane and an organopolysiloxane resin containing neither silicon atom-
35 bonded hydrogen atom nor alkenyl group. One of these may be used alone, or two or more thereof may also be used in

combination.

[0059]

Examples of the reinforcing filler may include fumed silica, silica fume, precipitated silica, fired silica, colloidal silica, and diatom earth, which are hydrophilic or hydrophobic, and particularly finely powdered ones. The particle diameter thereof is preferably 100 μm or less, and the specific surface area thereof is preferably 50 m^2/g or more, and even more preferably 150 m^2/g or more. Silica which has been surface-treated with organosilane, organosilazane, organocyclopolydimethylsiloxane, or the like in advance may also be suitably used. The amounts thereof to be added are usually in the range of 0.5 to 50 parts by mass, and preferably in the range of 1 to 30 parts by mass, relative to 100 parts by mass of the component (A). These may be used alone or in combination of a plurality of types thereof. If the mixed amount is small, it is difficult to obtain sufficient physical properties such as tear strength, and if it is too large, the fluidity of the silicone composition is lowered to reduce the workability.

[0060]

In the case of using a hydrophilic fine powder silica, it is preferable to use the silica after hydrophobizing the surface thereof with a hydrophobizing treatment agent, as necessary.

Examples of the hydrophobizing treatment agent may include an organosilazane such as hexamethyldisilazane, a halogenated silane such as methyltrichlorosilane, dimethyldichlorosilane, and trimethylchlorosilane, an organoalkoxysilane in which halogen atoms of the above compounds are substituted with an alkoxy group such as a methoxy group or an ethoxy group, and a dimethyl silicone oil, among which hexamethyldisilazane is preferable.

[0061]

The dispersant is a component which imparts a function of improving the dispersion of the blocked polyisocyanate

composition, and the powder dispersant having a density of 2.0 g/cm³ or more is preferable. Since a greater oil absorption amount further improves the dispersion of the blocked polyisocyanate composition, it is preferably 15 ml/100 g or
5 more. In terms of facilitating mixing, the average particle size of the dispersant is preferably 100 μm or less. Specific examples thereof may include a powder such as precipitated barium sulfate and talc.

[0062]

10 The viscosity adjusting material may be any material as long as it can impart a function of controlling the viscosity change of the silicone composition and the silicone composition kit, and specifically, a silane containing at least one silanol group (i.e., a hydroxyl group bonded to a
15 silicon atom) in one molecule is preferable. These are used alone or in plural. Specific examples thereof may include trimethylsilanol, triethylsilanol, triisopropylsilanol, triphenylsilanol, dimethylphenylsilanol, vinylphenylmethysilanol, and dimethylvinylsilanol.

20 Trimethylsilanol, triethylsilanol, triisopropylsilanol, and triphenylsilanol are preferable from the viewpoint of easy industrial availability.

[0063]

Examples of the pigment may include titanium oxide,
25 alumina silicic acid, iron oxide, zinc oxide, calcium carbonate, carbon black, a rare earth oxide, chromium oxide, a cobalt pigment, ultramarine, cerium silanolate, aluminum oxide, aluminum hydroxide, titanium yellow, carbon black, phthalocyanine blue, and mixtures thereof.

30 [0064]

Examples of the heat resistance-imparting agent may include cerium hydroxide, cerium oxide, iron oxide, fume titanium dioxide, and mixtures thereof.

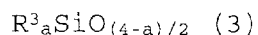
[0065]

35 Examples of the flame retardant may include a metal hydroxide such as aluminum hydroxide, magnesium hydroxide, and

calcium hydroxide, huntite, hydromagnesite, halloysite, kaolin, hydrous aluminum silicate, diatom earth, and calcium carbonate.
[0066]

The airtightness improving agent may be any agent as long
5 as it has an effect of lowering the air permeability of the
cured product, and may be an organic substance or an inorganic
substance. Specific examples thereof may include polyvinyl
alcohol, polyisobutylene, an isobutylene-isoprene copolymer,
mica, glass flakes, boehmite, powders having a flat shape such
10 as various metal foils and metal oxides, silicone rubber
powders, silicone resin powders, and mixtures thereof.
[0067]

In order to suppress the stickiness of the cured product
of the present invention, an organopolysiloxane in the form of
15 raw rubber at room temperature (25°C) may be further added, and
the average composition formula represented by the following
general formula (3) may be used.



20

(In the formula (3), R^3 's, which are the same as or different
from each other, are each an unsubstituted or substituted
monovalent hydrocarbon group having 1 to 18 carbon atoms; and
a is 1.7 to 2.1.)

25 [0068]

Here, at least two or more of the monovalent hydrocarbon
groups represented by the aforementioned R^3 are selected from
an alkenyl group such as a vinyl group, an allyl group, a
propenyl group, an isopropenyl group, a butenyl group, an
30 isobutenyl group, a hexenyl group, and a cyclohexenyl group.
Other groups are substituted or unsubstituted monovalent
hydrocarbon groups having 1 to 18 carbon atoms. More
specifically, the groups may be selected from an alkyl group
such as a methyl group, an ethyl group, a propyl group, an
35 isopropyl group, a butyl group, an isobutyl group, a tert-
butyl group, a pentyl group, a neopentyl group, a hexyl group,

a 2-ethylhexyl group, a heptyl group, an octyl group, a nonyl group, a decyl group, and a dodecyl group, a cycloalkyl group such as a cyclopentyl group, a cyclohexyl group, and a cycloheptyl group, an aryl group such as a phenyl group, a tolyl group, a xylyl group, a biphenyl group, and a naphthyl group, an aralkyl group such as a benzyl group, a phenylethyl group, a phenylpropyl group, and a methylbenzyl group, and a halogen-substituted alkyl group and a cyano-substituted alkyl group in which a portion or the entirety of hydrogen atoms in each of these hydrocarbon groups is substituted with a halogen atom, a cyano group, and the like, such as a chloromethyl group, a 2-bromoethyl group, a 3,3,3-trifluoropropyl group, a 3-chloropropyl group, and a cyanoethyl group.

[0069]

In selection of R^3 , a vinyl group is preferable as two or more alkenyl groups required, and a methyl group, a phenyl group, and a 3,3,3-trifluoropropyl group are preferable as the other groups. In addition, 70 mol% or more of all R^3 's is preferably a methyl group from the viewpoint of properties of a cured product and economic efficiency. Usually, those having a methyl group in an amount of 80 mol% or more of all R^3 's are used.

[0070]

Any methods known to those skilled in the art can be used to produce the silicone composition kit of the present invention, without limitation. Specifically, a silicone base may be prepared by mixing with a stirrer, or by uniformly kneading with a high shear type mixer such as a two-roll kneader mixer, a pressure kneader mixer, a loss mixer, a continuous extruder, or the like, and then any of the optional components may be mixed into the silicone base to produce the silicone composition kit. Each raw material may be produced by a known method using an emulsifying agent in an emulsifying machine.

[0071]

The method for producing an integrally molded product

using the silicone composition kit of the present invention can be freely selected depending on the viscosity of the mixture, and methods such as compression molding, injection molding, extrusion molding, transfer molding, and the like, and coating methods may be adopted. It is preferable to perform metering and mixing by an automatic apparatus from the viewpoint of production efficiency.

[0072]

As the base material to be bonded, an organic resin, a metal, or the like is suitably used. Examples of the organic resin may include an ordinary organic resin such as an olefin polymer-based organic resin and a condensation polymer-based organic resin. Specific examples thereof may include an acrylonitrile-butadiene-styrene (ABS) resin, a styrene resin, a polyethylene resin, a polypropylene resin, an acrylic resin, a PC resin, a PC/ABS resin, a polyethylene terephthalate resin, a PBT resin, a polyphenylene oxide resin, a PPS resin, a polyetheretherketone (PEEK) resin, a polyethersulfone (PES) resin, a polyetherimide (PEI) resin, a polysulfone resin, a nylon resin, a polyamide resin, a polyimide resin, a liquid crystal resin, and mixtures thereof. Examples of the shape thereof may include a plate shape, a film shape, a nonwoven fabric, and a woven fabric made of yarn using these resins as a raw material. Examples of the metal may include aluminum, iron, copper, zinc, lead, nickel, molybdenum, tungsten, manganese, chromium, cobalt, silicon, titanium, stainless steel, and brass.

[0073]

As a method for integrally molding the silicone composition kit of the present invention on the aforementioned organic resin or the like, may be mentioned a method for depositing an uncured silicone composition in a desired shape on a preformed organic resin and heating the same at a temperature equal to or lower than a melting temperature of the organic resin, a method for depositing an uncured silicone composition on a preformed organic resin and compressing the

same at or below the melting temperature of the organic resin,
and a method for injection molding an organic resin into a
mold by an injection molding machine, followed by heat-
injection of a silicone composition into the mold. As an

5 example of the method for producing the molded article of the
organic resin, for example, a known method of injecting the
organic resin into a mold heated to a softening point thereof
or higher, and then cooling the mold to a softening point or
lower of the organic resin is convenient. In this case, it is
10 possible to use equipment commonly referred to as an injection
molding machine and a transfer molding machine.

[0074]

The silicone composition kit of the present invention may
be in a liquid state, a putty state, or a paste state, but is
15 preferably in a liquid state or a paste state from the
viewpoint of ease of molding. The curing conditions of the
silicone composition are preferably at a temperature and for a
curing time at and during which the organic resin is not
deformed, melted or denatured. Although it depends on the type
20 of the resin, it is possible to obtain an integrally molded
product at 80 to 150°C for about 0.2 to 30 minutes, in
particular, for about 0.4 to 10 minutes.

[0075]

The method for producing the integrated product by the
25 self-adhesion of the silicone composition of the present
invention may be any production method without limitation as
long as the blocked polyisocyanate is dissociated by the heat
during curing and the self-adhesion of the isocyanate group is
expressed.

30 The following production method is preferable for
effectively expressing the stability during storage, the
simplicity and efficiency in processing, and the adhesion
properties.

That is, the method for producing the integrated product
35 by the self-adhesion of the silicone composition,
characterized by including:

a step of selecting the blocking agent in accordance with the curing method and the curing temperature for curing the curable silicone composition including the blocked polyisocyanate composition such that the viscosity of the blocked polyisocyanate composition and the vapor pressure of the blocking agent fall within the corresponding predetermined ranges;

a step of preserving the one-liquid composition or the silicone composition kit having at least two separated liquids by including the blocked polyisocyanate composition in which the selected blocking agent is used; and

a step of curing the silicone composition while initiating the thermal dissociation of the blocking agent by mixing and heating the silicone composition kit.

[0076]

In a case where the integrally molded product is produced by injection molding or coating of the curable silicone composition, it is preferable that the thermal dissociation takes place in a stage of the low viscosity before the curing proceeds and the activated isocyanate group is oriented to metal, an organic resin, or a fiber, which serves as an adherend, to express the self-adhesion function, while the curing of the silicone composition also proceeds to a certain degree in a concerted manner, thereby causing a state in which the isocyanate group is sufficiently exerting the self-adhesion function while being encapsulated in the cured silicone crosslinked network.

Further, a decision of whether the functional expression of the self-adhesion properties should be terminated at the time when the curing is completed or continued after the completion of the curing differs depending on a difference in the molding method, a difference in the adherend, or a difference in the application. However, in any case, at least, it is not preferable that the thermal dissociation of the blocked isocyanate composition is initiated after the completion of the curing of the silicone composition.

[0077]

After the blocked isocyanate composition is thermally dissociated, the released blocking agent is dissipated to the outside of the system by evaporation. If the blocking agent stays in the vicinity of the isocyanate group after being thermally dissociated, the reactivity of the isocyanate group is still slightly suppressed. If the dissociation of the blocking agent is too vigorous, the blocking agent is dissipated during storage of the silicone composition kit or before initiation of the curing, thereby causing inactivation of the isocyanate group or an undesired reaction. Further, if the dissipation is too slow, the curing is prematurely completed before the isocyanate group is sufficiently exposed. Thus, the blocking agent needs to be properly dissipated.

Further, in some cases, the blocking agent contributes, for example, to prevent the deterioration of the adhesion properties or the like of the integrally molded product with time, and thus the moderately remaining blocking agent due to slow dissipation sometimes acts advantageously.

From the above reasons, a decision of how the dissipation of the blocking agent should be differs depending on a difference in the molding method, a difference in the adherend, or a difference in the application, however, it can be designed to some extent.

[0078]

The relationship between the curing temperature of the silicone composition and the thermal dissociation temperature of the blocking agent varies depending various factors such as the curing method, the curing condition, the device, the curing time, the temperature rising mode, the application, and a kind of base material or base fabric, and cannot be unconditionally specified. However, in general, the thermal dissociation temperature of the blocking agent is preferably higher than the curing temperature by about 10°C. This is because, in many cases, it is preferable that the dissociation of the blocking agent is prevented until shortly before the

curing in order to prevent the inactivation of the isocyanate group and poisoning of the catalyst and to express the adhesion properties immediately after the initiation of the curing. The blocking agent is preferably selected on the basis
5 of the vapor pressure with an emphasis on this point.

[0079]

In a case of performing coating, the base material preferably has a plate-like shape, a film-like shape, or a fabric-like shape. The coating is performed by using, for
10 example, immersion and padding, brush coating, flow coating, spray, roller coating, gravure coating, comma coater, printing, knife coating, wire bar, air brush, sloop padding, and roll coating. These are used individually or in combination in accordance with the situation. Further, it is not necessary to
15 perform the coating at once and the coating may be performed multiple times until the desired coating condition is obtained. Thus, the cured film after the coating is not necessarily formed in a single layer and may be constituted by a plurality of the cured films. Further, for the purpose of imparting
20 other effects such as, for example, antifouling, destaticizing, imparting slidability, and preventing blocking, the surface of the cured film after the coating may be added with a desired component for the silicone composition, subjected to surface processing after the coating or curing, or further provided
25 with the cured layer having such functions.

[0080]

The drying and curing after carrying out the coating is normally performed in a heating device capable of heating by hot air, steam, infrared ray, near infrared ray, a gas burner,
30 a heat exchanger, or other energy sources. Note that, for the drying and curing, any device capable of achieving the intended purpose may be used in addition to the commonly used heating device. Examples thereof may include a heating calendar roll, a heatable bonding press, a heatable stage
35 press or a high-temperature contact roll, a hot air dryer, and a microwave dryer.

[0081]

In order to avoid the formation of air bubbles in the cured film during the curing, a plurality of temperature zones having different temperatures are preferably arranged in the heating device. For example, the preliminary drying can be performed at the temperature of 60 to 150°C, preferably 80 to 130°C, further preferably 90 to 120°C, in a first temperature zone and the curing can be performed at the temperature of 300°C or lower in the subsequent second temperature zone onward. However, the curing is preferably performed at 250°C or lower as many films and fabrics have heat resistance limits in processing.

[0082]

Note that, even if it is difficult to arrange the plurality of the temperature zones due to a restriction on processing, the preliminary drying is preferably performed such that the temperature of the base material to be cured reaches at least 170°C or higher at least once. Further, although the retention time required for the curing varies depending on the weight of the coating, the thermal conductivity of the fabric, and the thermal conductivity of the coated film or fabric, it is preferably about 0.5 to 30 minutes. The retention may be performed by leaving the cured product for 10 minutes to several hours at room temperature.

[0083]

Specific examples of the application of the coating may include a tent material and an air bag base fabric. The air bag, which is made by sewing the air bag base fabrics together in a bag shape, is a device that is mainly equipped in an automobile to secure the safety of the driver and passenger by expanding the bag upon collision. The air bag base fabric is usually a fabric interwoven with a synthetic fiber such as polyimide and polyethylene terephthalate. The silicone composition kit of the present invention is coated on these synthetic fiber woven fabrics. Specifically, a polyamide fiber woven fabric such as nylon 6, nylon 66, and nylon 46, an

aramid woven fabric, a polyester fiber woven fabric represented by polyalkylene terephthalate, a polyetherimide fiber woven fabric, a sulfone-based fiber woven fabric, a carbon fiber woven fabric, or a mixture thereof may be used.

5 The coating is performed on a plain fabric, a bag-like fabric, a hose-like fabric, or the like using a yarn with a thickness of 10 to 5,000 dtex. From the viewpoints of processability and economic efficiency, the fabric using a yarn of 50 to 1,000dtex is preferably used.

10 [0084]

As the fabric for the coating, both an unrefined fabric and a refined fabric can be used. A refining step may be omitted by performing the coating directly on the unrefined fabric. In a case where the refining is performed, a control
15 value of the amount of residual oil may be more relaxed than the conventional value. The amount of residual oil is most preferably 0.01% by mass or more relative to the total weight of the fabric from the viewpoint of adhesiveness, however, the fabric of 0.1% by mass or more may be used from the viewpoint
20 of economic efficiency.

[0085]

In a case where the silicone composition kit of the present invention is prepared in two liquids, a ratio between them is preferably 1:1, although the ratio is not particularly
25 limited. Further, an automated device is preferably used for measurement, mixture, and supply from the viewpoint of labor saving, automatization, and improving productivity. In this case, it is preferable that two liquids have a viscosity difference as small as possible as they can be precisely
30 measured. The viscosity difference is preferably 100 Pa·s or less, and more preferably 50 Pa·s or less.

EXAMPLES

[0086]

Hereinafter, the present invention will be specifically
35 described with reference to examples and comparative examples. However, the present invention is not limited to the following

examples. It is noted that "part(s)" in each example means "part(s) by mass". The results of the examples and comparative examples are shown in Tables 1 and 2.

[0087]

5 <Property Measuring Method of Silicone Composition>

The silicone composition kit was mixed and press-cured at a temperature of 170°C for a curing time of 5 minutes to produce a cured sheet having a thickness of 2 mm. The hardness of the resulting sheet was measured in accordance with JIS K
10 6249 (a type A durometer).

[0088]

<Adhesiveness Measuring Method of Silicone Composition>

The silicone composition kit was mixed, coated on a plain polyester fabric woven with a yarn of 470 dtex using a knife
15 coater, and cured at a temperature of 170°C for a curing time of 60 seconds in a drying furnace. Further, in a scratch test, the surface of the cured product was scratched back and forth 20 times with a metal plate made of stainless steel (a thickness of 1.5 mm, a 3cm square) and the number of
20 occurrences of tear or peeling was measured by visually observing the peeling condition of the cured film.

[0089]

<Example 1>

<Preparation of Silicone Composition Kit Agent A>

25 To 73.5 parts by mass of a dimethylpolysiloxane having vinyl groups at both terminals with the viscosity of about 20,000 mPa·s as a component (A-1), 5.7 parts by mass of a dimethylpolysiloxane having vinyl groups at both terminals with the viscosity of about 100,000 mPa·s as a component (A-2),
30 and 1.0 part by mass of a dimethylpolysiloxane (having the vinyl group content of 3 mmol/g) having vinyl groups at both terminals and in the molecule with the viscosity of about 10 mPa·s as a component (A-4) were added and mixed. To this mixture, 0.6 parts by mass of Duranate X2252 (manufactured by
35 Asahi Kasei Corp.), in which acetylacetone (having the vapor pressure of 6.9 mmHg at 20°C) as the blocking agent was

adjusted to 2 relative to the mole number of the isocyanate groups, with the viscosity of about 20,000 mPa·s at 60°C as a component (B-1), 4.9 parts by mass of fumed silica having the specific surface area measured by the BET method of 300 m²/g, 12.1 parts by mass of an organopolysiloxane resin having a vinyl group unit and a Q siloxane unit in the molecule, and 0.6 parts by mass of precipitated barium sulfate powders (having the density of 4.0 g/cm³, the oil absorption amount of 18 ml/100g, and the average particle diameter of 0.3 μm) were added, and the resulting mixture was sufficiently mixed with a stirring and mixing machine. Further, to this mixture, 0.6 parts by mass of a dimethylpolysiloxane solution including 1% by mass of a platinum-divinyltetramethyldisiloxane complex in terms of platinum content as a component (C), and 1.0 part by mass of diisopropoxy(ethylacetoacetate)titanium were added, and the resulting mixture was sufficiently mixed with a stirring and mixing device to prepare a silicone composition kit agent A of Example 1.

[0090]

<Preparation of silicone composition kit agent B>

To 47.3 parts by mass of a dimethylpolysiloxane having vinyl groups at both terminals with the viscosity of about 20,000 mPa·s as a component (A-1), 10.4 parts by mass of a dimethylpolysiloxane having vinyl groups at both terminals with the viscosity of about 100,000 mPa·s as a component (A-2), and 15.7 parts by mass of a dimethylpolysiloxane having vinyl groups at both terminals with the viscosity of about 1,000 mPa·s as a component (A-3) were added and mixed. To this mixture, 1.2 parts by mass of a methylhydrogenpolysiloxane capped at both terminals with trimethylsiloxy groups (having the hydrogen content of 1.6% by mass) with the viscosity of 30 mPa·s at 25°C as a component (D-1), 6.1 parts by mass of a methylhydrogenpolysiloxane capped at both terminals with trimethylsiloxy groups (having the hydrogen content of 0.8% by mass) with the viscosity of 70 mPa·s at 25°C as a component (D-2), 0.4 parts by mass of a methylphenylhydrogenpolysiloxane

capped at both terminals with trimethylsiloxy groups (having the hydrogen content of 0.8% by mass) with a viscosity of 40 mPa·s at 25°C as a component (D-3), and 10.1 parts by mass of an organohydrogenpolysiloxane (having the hydrogen content of 0.05% by mass) having hydrogen atoms bonded to silicon atoms only at both terminals of the molecule chain with the viscosity of 50 mPa·s at 25°C as a component (D-4) were added. To this mixture, 7.0 parts by mass of fumed silica having the specific surface area measured by the BET method of 300 m²/g, 1.6 parts by mass of 3-glycidoxypropyltrimethoxysilane, 0.2 parts by mass of diallyl maleate, and 0.3 parts by mass of trimethylsilanol were further added, and the resulting mixture was sufficiently mixed with a stirring and mixing device to prepare a silicone composition kit agent B of Example 1.

[0091]

The silicone composition kit agent A and agent B of Example 1 were allowed to stand for one day after production and mixed together at a ratio of 1:1 to produce a silicone composition of Example 1. The resulting silicone composition was cured to produce a sheet and the hardness of the sheet was measured to be 22. Further, the silicone composition was coated on a polyester fabric at about 35 g/m² and cured. After leaving it for 24 hours, the scratch test was performed. There was no tear or peeling in the cured film after 20 times of scratching, demonstrating the excellent adhesion properties. On the other hand, a cured product, which was produced by mixing the agents stored for 3 months at 40°C after production at a ratio of 1:1, had the hardness of 22 and showed no tear or peeling in the cured film after 20 times of scratching in the scratch test.

[0092]

<Example 2>

Instead of 0.6 parts by mass of Duranate X2252 (manufactured by Asahi Kasei Corp.) as the component (B) of the silicone composition kit agent A of Example 1, 0.6 parts by mass of Duranate X2252 (manufactured by Asahi Kasei Corp.),

in which methylethylketoxime (having the vapor pressure of 8 mmHg at 20°C) as the blocking agent was adjusted to 2 relative to the mole number of the isocyanate groups, with the viscosity of about 30,000 mPa·s at 60°C as a component (B-2) was used to produce a silicone composition kit agent A of Example 2. Further, instead of 0.2 parts by mass of diallyl maleate of the silicone composition kit agent B of Example 1, 0.2 parts by mass of 2-methyl-3-butyn-2-ol was mixed to produce a silicone composition kit agent B of Example 2.

10 [0093]

The silicone composition kit agent A and agent B of Example 2 were allowed to stand for one day after production and mixed together at a ratio of 1:1 to produce a silicone composition of Example 2. The resulting silicone composition was cured to produce a sheet and the hardness of the sheet was measured to be 22. Further, the silicone composition was coated on a polyester fabric at about 35 g/m² and cured. After leaving it for 24 hours, the scratch test was performed. There was no tear or peeling in the cured film after 20 times of scratching, demonstrating the excellent adhesion properties. On the other hand, a cured product, which was produced by mixing the agents stored for 3 months at 40°C after production at a ratio of 1:1, had the hardness of 22 and showed no tear or peeling in the cured film after 20 times of scratching in the scratch test.

25 [0094]

<Comparative Example 1>

Instead of using 0.6 parts by mass of Duranate X2252 (manufactured by Asahi Kasei Corp.) as the component (B) of the silicone composition kit agent A produced in Example 1, 0.6 parts by mass of Duranate X2252 (manufactured by Asahi Kasei Corp.), in which diisopropylamine (having the vapor pressure of 60 mmHg at 20°C) as the blocking agent was adjusted to 2 relative to the mole number of the isocyanate groups, with the viscosity of 30,000 mPa·s at 60°C as a component (B-3) was used to produce a silicone composition kit agent A of

Comparative Example 1. Further, instead of using 0.2 parts by mass of diallyl maleate in the silicone composition kit agent B of Example 1, 0.2 parts by mass of 2-methyl-3-butyn-2-ol was mixed to produce a silicone composition kit agent B of

5 Comparative Example 1.

[0095]

The silicone composition kit agent A and agent B of Comparative example 1 were allowed to stand for one day after production and mixed together at a ratio of 1:1 to produce a
10 silicone composition of Comparative Example 1. The resulting silicone composition was cured to produce a sheet and the hardness of the sheet was measured to be 22. Further, the silicone composition was coated on a polyester fabric at about 36 g/m² and cured. After leaving it for 24 hours, the scratch
15 test was performed. The cured film was torn and peeled after 15 times of scratching. On the other hand, a cured product, which was produced by mixing the materials stored for 3 months at 40°C after production at a ratio of 1:1, had the hardness of 20 and showed peeling in the cured film after 13 times of
20 scratching in the scratch test.

[0096]

<Comparative Example 2>

Instead of using 0.6 parts by mass of Duranate X2252 (manufactured by Asahi Kasei Corp.) as the component (B) of
25 the silicone composition kit agent A produced in Example 1, 0.6 parts by mass of Duranate X2252 (manufactured by Asahi Kasei Corp.), in which diisopropylamine (having the vapor pressure of 60 mmHg at 20°C) as the blocking agent was adjusted to 2 relative to the mole number of the isocyanate groups,
30 with the viscosity of 120,000 mPa·s at 60°C as a component (B-4) was used to produce a silicone composition kit agent A of Comparative Example 2. Further, instead of using 0.2 parts by mass of diallyl maleate in the silicone composition kit agent B of Example 1, 0.2 parts by mass of 2-methyl-3-butyn-2-ol was
35 mixed to produce a silicone composition kit agent B of Comparative Example 2.

[0097]

The silicone composition kit agent A and agent B of Comparative Example 2 were allowed to stand for one day after production and mixed together at a ratio of 1:1 to produce a
5 silicone composition of Comparative Example 2. The resulting silicone composition was cured to produce a sheet and the surface of the cured sheet showed stickiness. The hardness of the sheet was measured to be 18. Further, the silicone composition was coated on a polyester fabric at about 35 g/m²
10 and cured. The cured product showed stickiness. The cured product was left for 24 hours and subjected to the scratch test. As a result, the cured film was peeled after 8 times of scratching. On the other hand, a cured product, which was produced by mixing the materials stored for 3 months at 40°C
15 after production at a ratio of 1:1 also had stickiness. The cured product had the hardness of 16 and showed peeling in the cured film after 4 times of scratching in the scratch test.

[0098]

[Table 1]

	UNIT: part(s) by mass			
	EXAMPL E 1	EXAMPL E 2	COMPARATIV E EXAMPLE 1	COMPARATIV E EXAMPLE 2
SILICONE COMPOSITION KIT AGENT A				
COMPONENT (A)				
(A-1) VISCOSITY 20,000 mPa·s	73.5	73.5	73.5	73.5
(A-2) VISCOSITY 100,000 mPa·s	5.7	5.7	5.7	5.7
(A-3) VISCOSITY 1,000 mPa·s	-	-	-	-
(A-4) VISCOSITY 10 mPa·s (VINYL GROUP CONTENT 3 mmol/g)	1.0	1.0	1.0	1.0
COMPONENT (B)				
(B-1) DURANATE X 2252 (VISCOSITY 20,000 mPa·s/60°C) ACETYLACETONE (VAPOR PRESSURE 6.9 mmHg/20°C)	0.6	-	-	-
(B-2) DURANATE X2252 (VISCOSITY 30,000 mPa·s/60°C) METHYLETHYL KETONE OXIME (VAPOR PRESSURE 8 mmHg/20°C)	-	0.6	-	-
(B-3) DURANATE X2252 (VISCOSITY 30,000 mPa·s/60°C) DIISOPROPYLAMINE (VAPOR PRESSURE 60 mmHg/20°C)	-	-	0.6	-
(B-4) DURANATE X2252 (VISCOSITY 120,000 mPa·s/60°C) DIISOPROPYLAMINE (VAPOR PRESSURE 60 mmHg/20°C)	-	-	-	0.6
COMPONENT (C) CURING CATALYST	0.6	0.6	0.6	0.6
FUMED SILICA (SPECIFIC SURFACE AREA 300 m ² /g)	4.9	4.9	4.9	4.9
ORGANOPOLYSILOXANE RESIN	12.1	12.1	12.1	12.1
DIISOPROPOXY(ETHYLACETOACETATE)TITANIUM	1.0	1.0	1.0	1.0
PRECIPITATED BARIUM SULFATE	0.6	0.6	0.6	0.6
VISCOSITY OF AGENT A (Pa·s)	57	57	57	57

RATIO OF MOLE NUMBER OF BLOCKING AGENTS/MOLE NUMBER OF ISOCYANATE GROUPS	2	2	2	2
SILICONE COMPOSITION KIT AGENT B				
COMPONENT (A)				
(A-1) VISCOSITY 20,000 mPa·s	47.3	47.3	47.3	47.3
(A-2) VISCOSITY 100,000 mPa·s	10.4	10.4	10.4	10.4
(A-3) VISCOSITY 1,000 mPa·s	15.7	15.7	15.7	15.7
(A-4) VISCOSITY 10 mPa·s(VINYL GROUP CONTENT 3 mmol/g)	-	-	-	-
COMPONENT (D) CROSSLINKING AGENT				
(D-1) VISCOSITY 30 mPa·s (HYDROGEN CONTENT 1.6% by mass)	1.2	1.2	1.2	1.2
(D-2) VISCOSITY 70 mPa·s (HYDROGEN CONTENT 0.8% by mass)	6.1	6.1	6.1	6.1
(D-3) VISCOSITY 40 mPa·s (HYDROGEN CONTENT 0.8% by mass)	0.4	0.4	0.4	0.4
(D-4) VISCOSITY 50 mPa·s (HYDROGEN CONTENT 0.05% by mass)	10.1	10.1	10.1	10.1
FUMED SILICA(SPECIFIC SURFACE AREA 300 m ² /g)	7.0	7.0	7.0	7.1
3-GLYCIDOXYPROPYLTRIMETHOXYSILANE	1.6	1.6	1.6	1.6
DIALLYL MALEATE (VAPOR PRESSURE 4 mmHg/20°C)	0.2	-	-	-
2-METHYL-3-BUTYN-2-OL (VAPOR PRESSURE 15 mmHg/20°C)	-	0.2	0.2	0.2
TRIMETHYLSILANOL	0.3	0.3	0.3	0.3
VISCOSITY OF AGENT B (Pa·s)	53	53	53	53
RATIO OF MOLE NUMBER OF HYDROGEN ATOMS IN COMPONENT (D) RELATIVE TO VINYL GROUPS IN TOTAL COMPONENTS (A) (TOTAL Si-H GROUPS/TOTAL VINYL GROUPS)	3.3	3.3	3.3	3.3

[0099]

[Table 2]

TEST RESULTS	EXAMPLE 1	EXAMPLE 2	COMPARATIVE EXAMPLE 1	COMPARATIVE EXAMPLE 2
CURING PROPERTIES (CURING CONDITIONS: 170°C × 5 min)				
IMMEDIATELY AFTER PRODUCTION	CURED	CURED	CURED	CURED (WITH STICKINESS)
40°C × AFTER 3 MONTH STORAGE	CURED	CURED	CURED	CURED (WITH STICKINESS)
HARDNESS (TYPE A DUROMETER)				
IMMEDIATELY AFTER PRODUCTION	22	22	22	18
40°C × AFTER 3 MONTH STORAGE	22	22	20	16
SCRATCH TEST				
COATING AMOUNT ON POLYESTER FABRIC (m ² /g)	35	35	36	35
CURING CONDITIONS 170°C × 1 min				
NUMBER OF SCRATCHING (IMMEDIATELY AFTER PRODUCTION)	20 OR MORE	20 OR MORE	15	8
NUMBER OF SCRATCHING (40°C × AFTER 3 MONTH STORAGE)	20 OR MORE	20 OR MORE	13	4

INDUSTRIAL APPLICABILITY

5 [0100]

Since the curable silicone composition or the silicone composition kit of the present invention does not contain an organic solvent, the hazardous nature is lowered, and it is safe and easy to manage. Furthermore, the adhesion properties and the curability after storage are sufficient and less fluctuated, thereby improving the quality thereof as greatly improving the productivity.

CLAIMS

1. A silicone composition being a one-liquid curable composition including a blocked polyisocyanate composition, or a silicone composition kit in which the curable silicone compositions including the blocked polyisocyanate compositions are distributed at least in two liquids as different components and preserved, both of which being characterized in that a thermally dissociating blocking agent component included in the blocked polyisocyanate composition has a vapor pressure of not less than a predetermined lower limit value and not more than a predetermined upper limit value.
2. The silicone composition or the silicone composition kit according to claim 1, wherein the lower limit value of the vapor pressure at 20°C is 0.05 mmHg and the upper limit value thereof is 40 mmHg.
3. The silicone composition or the silicone composition kit according to claim 1 or 2, wherein the blocked polyisocyanate composition is substantially free from an organic solvent and has a viscosity at 60°C of 100,000 mPa·s or lower, and the blocked polyisocyanate composition includes a polyisocyanate obtained from at least one kind of diisocyanate selected from the group consisting of an aliphatic diisocyanate and an alicyclic diisocyanate, and the thermally dissociating blocking agent.
4. The silicone composition or the silicone composition kit according to any one of claims 1 to 3, wherein the curable silicone composition is an addition-curable silicone composition.
5. A production method for obtaining an integrally molded product with a base material, and the resulting integrally molded product, wherein the integrally molded product is obtained by curing the silicone composition or the silicone

composition kit according to any one of claims 1 to 4.

6. A method for producing an integrated product utilizing self-adhesion of a silicone composition, the method

5 comprising:

a step of selecting a blocking agent in accordance with a curing method and a curing temperature for curing a curable silicone composition including a blocked polyisocyanate composition such that a viscosity of a blocked polyisocyanate
10 composition and a vapor pressure of a blocking agent fall within respective predetermined ranges;

a step of preserving a one-liquid composition or a silicone composition kit having at least two separated liquids by including the blocked polyisocyanate composition in which
15 the selected blocking agent is used; and

a step of curing the silicone composition while thermal dissociation of the blocking agent is initiated by mixing and heating the silicone composition kit.

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2019/053842

A. CLASSIFICATION OF SUBJECT MATTER
INV. C09J183/04 C08L83/04 C08G18/80
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)
C09J 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, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2017/029555 A1 (HAMAJIMA YUTA [JP] ET AL) 2 February 2017 (2017-02-02)	1-3
Y	Synthesis example 3; claim 1; example 4 -----	5,6
X	DATABASE WPI Week 201128 Thomson Scientific, London, GB; AN 2011-D87791 XP002790600, -& WO 2011/043471 A1 (DEXERIALS CORP) 14 April 2011 (2011-04-14)	1,3,4
Y	abstract; example 1 -----	5,6
X,P	WO 2019/001901 A1 (WACKER CHEMIE AG [DE]) 3 January 2019 (2019-01-03) abstract; claims 1-2,4-6; examples 1-2 -----	1-4



Further documents are listed in the continuation of Box C.



See patent family annex.

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

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