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(71) Applicant (for all designated States except US): **DOW CORNING TORAY CO., LTD.** [JP/JP]; 1-3, Marunouchi 1-chome, Chiyoda-ku, Tokyo 1000005 (JP).

(72) Inventor; and

(75) Inventor/Applicant (for US only): **SEKIBA, Kazuhiro** [JP/JP]; c/o Dow Corning Toray Co., Ltd., 2-2 Chigusakaigan, Ichihara-shi, Chiba 2990108 (JP).

(74) Agent: **KUBOTA, Yoshitaka**; c/o Asa International Patent Firm Chiba Office, Takada Building 2F, 2-15, Yayoicho, Inage-ku, Chiba-shi, Chiba 2630022 (JP).

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(54) Title: THERMOCONDUCTIVE SILICONE ELASTOMER, THERMOCONDUCTIVE SILICONE ELASTOMER COMPOSITION AND THERMOCONDUCTIVE MEDIUM

(57) Abstract: A thermoconductive silicone elastomer comprising a silicone elastomer being a cured body of a hydrosilation-curable organopolysiloxane composition, a reinforcement fine silica powder, a thermoconductive inorganic powder, and an alkylphenylpolysiloxane that is liquid at room temperature. A hydrosilation-curable thermoconductive silicone elastomer composition comprising: a hydrosilation-curable organopolysiloxane composition, a reinforcement fine silica powder, a thermoconductive inorganic powder, and an alkylphenylpolysiloxane that is liquid at room temperature. A thermoconductive medium comprising the aforementioned thermoconductive silicone elastomer.

WO 2006/025552 A1

## DESCRIPTION

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THERMOCONDUCTIVE SILICONE ELASTOMER,  
THERMOCONDUCTIVE SILICONE ELASTOMER COMPOSITION AND  
THERMOCONDUCTIVE MEDIUM

## Technical Field

[0001] The present invention relates to a thermoconductive silicone elastomer that, when it is sandwiched in an adhered state between a heat-generating member and a heat-radiating member and is maintained in such a state over a long period of time, can be peeled from the aforementioned members at their respective interfaces with this elastomer, and to a thermoconductive medium comprising the aforementioned thermoconductive silicone elastomer. The present invention also relates to a thermoconductive silicone elastomer composition that is composed of a hydrosilation-curable organopolysiloxane composition, a fine reinforcement silica powder, a thermoconductive inorganic powder, and an alkylphenylsiloxane that is liquid at room temperature.

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## Background Art

[0002] In view of recent trends towards miniaturization, increase in density, and improvement in properties of electronic parts that incorporate heat-generating elements and are intended for use in electronic devices, methods of transmitting and dissipating heat from such elements become an important issue. Heretofore, it has been known to fill the space between the aforementioned heat-generating elements and heat-radiating fins or a similar metal chassis with a molded body of a thermoconductive rubber filled with a large amount of alumina or a similar thermoconductive inorganic powder. For example, Japanese Unexamined Patent Application Publication (hereinafter JP Kokai) S56-2349 describes a molded rubber article for heat dissipation.

[0003] Since such a molded rubber article is not tacky and has a high hardness of about 54 to 73 measured by a Type A durometer according to JIS K6253, it cannot be easily fixed between a heat-generating element and heat-radiating fins, or a similar metal chassis, and, therefore, assembling of 5 such units is associated with difficulties. Fixation of the unit in an assembled state requires the use of screws and bolt, and this creates problems in case of disassembling and repair. With regard to plasma display panels that recently have found use in personal computers, TV sets, etc., it was proposed to place a grooved heat-radiating adhesive sheet between the plasma display and the 10 heat-generating sheet (see JP Kokai 2001-11402). An adhesive agent mentioned in the above application is a silicone-type adhesive agent, but the adhesive connection between the plasma display and the heat-radiating sheet is permanent, and the unit cannot be disassembled for repair or discarding without breaking it apart, which is inconvenient.

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#### **Summary of the Invention**

[0004] It is an object of the present invention to provide a thermoconductive elastomer that, when sandwiched between a heat-generating member and a heat-radiating member can be fixed between these members without the use 20 of screws, bolts or other fasteners, can be easily peeled from the aforementioned members, if necessary for repair or disassembling, after being maintained in the sandwiched state over a long period of time. It is another object to provide a thermoconductive silicone elastomer composition for manufacturing the aforementioned silicone elastomer.

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[0005] The above object is accomplished by means of

[1] A thermoconductive silicone elastomer that has a hardness of 5 to 70

measured by a spring-type hardness tester in accordance with the provisions of SRIS 0101-1968, a tensile strength of no less than 0.2 MPa in 30 accordance with JIS K 6251, a elongation of no less than 300%, and is

capable of peeling at interfaces from a heat-generating member and a heat-radiating member after being sandwiched in an adhered state between said members and held in this state for 48 hours at 120°C, the aforementioned thermoconductive silicone elastomer comprising 90 to 10  
5 wt.% of a silicone elastomer being a cured body of a hydrosilation-curable organopolysiloxane composition consisting of (A) an alkylalkenylpolysiloxane having at least two alkenyl groups in one molecule, (B) an alkylhydrogenpolysiloxane having at least two hydrogen atoms in one molecule and (C) a platinum metal type catalyst; (D) 0.2 to 5.0 wt.% of a  
10 finely powdered reinforcement silica; (E) 10 to 90 parts by weight of a conductive inorganic powder ; and (F) 0.1 to 10 wt.% of an alkylphenylpolysiloxane which is liquid at room temperature (100 wt.% total), where components (D), (E) and (F) are dispersed in said silicone elastomer.

15 [2] The thermoconductive silicone elastomer of Item [1], wherein component (A) has a linear molecular structure.

[3] The thermoconductive silicone elastomer of Item [1], wherein component (A) is a methylvinylpolysiloxane of the following formula:  $(R^1)_3SiO(R_2SiO)_mSi(R^2)_3$  (where R, R<sup>1</sup>, R<sup>2</sup> are methyl or vinyl groups; at least two vinyl groups  
20 being present in one molecule; when all R's in one molecule are methyl group, at least one R<sup>1</sup> and at least one R<sup>2</sup> are vinyl groups; when one R in one molecule is a methyl group, at least one of R<sup>1</sup> and R<sup>2</sup> is a vinyl group; 0.1 to 2% of the total number of methyl groups and vinyl groups of one molecule are vinyl groups; and "m" is a number within the range of 50 to 1,000); the alkylhydrogenpolysiloxane is a methylhydrogenpolysiloxane; component (D) is a fumed silica; component (E) is an alumina powder or a crystalline silica powder; and the alkylphenylpolysiloxane is a methylphenylpolysiloxane of the following formula:  $(R^4)_3SiO[(R^3)_2SiO]_nSi(R^4)_3$  (where R<sup>3</sup> and R<sup>4</sup> are methyl or phenyl groups; 1 to 50 % of the total number of methyl groups and  
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phenyl groups in one molecule are phenyl groups; «n» is a number within the range of 1 to 100).

[0006]

5 [4] A hydrosilation-curable thermoconductive silicone elastomer composition comprising: 90 to 10 wt.% of a hydrosilation-curable organopolysiloxane composition consisting of (A) an alkylalkenylpolysiloxane having at least two alkenyl groups in one molecule, (B) an alkylhydrogenpolysiloxane having at least two hydrogen atoms in one molecule, and (C) a platinum metal type catalyst; (D) 0.2 to 5.0 wt.% of a finely powdered reinforcement silica; (E) 10 to 90 parts by weight of a thermoconductive inorganic powder with an average particle size of 1 to 50  $\mu\text{m}$ ; (F) 0.1 to 10 wt.% of an alkylphenylpolysiloxane which is liquid at room temperature (100 wt.% total); said composition, when cured, forming a thermoconductive silicone elastomer that has a hardness of 5 to 70 measured by a spring-type hardness tester in accordance with the provisions of SRIS 0101-1968, a tensile strength of no less than 0.2 MPa in accordance with JIS K 6251, an elongation of no less than 300%, and is capable of peeling at interfaces from a heat-generating member and a heat-radiating member after being sandwiched in an adhered state between said members and held in this state for 48 hours at 120°C.

20 [5] The hydrosilation-curable thermoconductive silicone elastomer composition of Item [4], wherein component (A) has a linear molecular structure.

25 [6] The hydrosilation-curable thermoconductive silicone elastomer composition of Item [4], wherein component (A) is a methylvinylpolysiloxane of the following formula:  $(\text{R}^1)_3\text{SiO}(\text{R}_2\text{SiO})_m\text{Si}(\text{R}^2)_3$  (where R, R<sup>1</sup>, R<sup>2</sup> are methyl or vinyl groups; at least two vinyl groups being present in one molecule; when all R's in one molecule are methyl group, at least one R<sup>1</sup> and at least one R<sup>2</sup> are vinyl groups; when one R in one

molecule is a methyl group, at least one of R<sup>1</sup> and R<sup>2</sup> is a vinyl group; 0.1 to 2% of the total number of methyl groups and vinyl groups of one molecule are vinyl groups; and "m" is a number within the range of 50 to 1,000); the alkylhydrogenpolysiloxane is a methylhydrogenpolysiloxane; component (D) 5 is a fumed silica; component (E) is an alumina powder or a crystalline silica powder; and the alkylphenylpolysiloxane is a methylphenylpolysiloxane of the following formula: (R<sup>4</sup>)<sub>3</sub> SiO[(R<sup>3</sup>)<sub>2</sub> SiO]<sub>n</sub> Si(R<sup>4</sup>)<sub>3</sub> (where R<sup>3</sup> and R<sup>4</sup> are methyl or phenyl groups; 1 to 50 % of the total number of methyl groups and phenyl groups in one molecule are phenyl groups; "n" is a number within 10 the range of 1 to 100).

[7] A thermoconductive medium comprising the thermoconductive silicone elastomer of one of Items [1] to [3].

[0007] Since the thermoconductive silicone elastomer and the 15 thermoconductive medium of the present invention can be easily peeled from a heat-generating member and a heat-radiating member between which it is sandwiched without the use of screws, bolts or other fasteners and kept in such a sandwiched state over a long period of time, the heat-generating member and heat-radiating member that are adhered to each other via the 20 aforementioned thermoconductive silicone elastomer can be easily disassembled and repaired. The effect of the thermoconductive hydrosilation-curable silicone elastomer composition of the present invention consists in that it can be cured, for obtaining the aforementioned thermoconductive silicone elastomer that can be easily peeled from a 25 heat-generating and heat-radiating members between which it is sandwiched without the use of screws, bolts or other fasteners and kept in such a sandwiched state over a long period of time, so that the heat-generating member and heat-radiating member that are adhered to each other via the aforementioned thermoconductive silicone elastomer can be easily 30 disassembled and repaired.

### Best Mode for Carrying Out the Invention

[0008] Alkenyl groups contained in the alkylalkenylpolysiloxane that has in one molecule at least two alkenyl groups and that constitutes component (A) of the hydrosilation-curable organopolysiloxane composition participate in the 5 hydrosilation reaction and are cross-linked with silicon-bonded hydrogen atoms of component (B). The alkylalkenylpolysiloxane may have a linear molecular structure or, to some extent, branched linear molecular structure. It may be in the form of a single polymer, copolymer, or a mixture of two or more polymers.

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[0009] The silicon-bonded alkyl groups of component (A) may be represented by methyl, ethyl, propyl, butyl, octyl, cyclopentyl, and cyclohexyl groups; and the alkenyl groups can be represented by vinyl, allyl, butenyl, pentenyl, hexenyl, and heptenyl groups. Component (A) may also contain 15 phenyl, toyl, xylyl, or similar aryl group; benzyl, phenylethyl, or similar aralkyl groups; and 3,3,3,-trifluoropropyl, or similar halogenated alkyl groups, if the content of them in one molecule is less than 5 mole %. The terminal groups are represented by alkyl or aryl groups, but may be represented by methoxy, ethoxy, or similar alkoxy groups and hydroxyl groups. It is 20 recommended that 90 mole % or more in one molecule are methyl groups, and more preferably all silicon-bonded alkyl groups in one molecule are methyl groups. It is recommended that the silicon-bonded alkenyl groups of a molecule are vinyl groups. From the point of view of the silicon elastomer hardness, it is recommended to have in one molecule five or more 25 silicon-bonded alkenyl groups.

[0010] A typical example of an alkylalkenylpolysiloxane that contains in one molecule at least two alkenyl groups is a methylvinylpolysiloxane of the following formula:  $(R^1)_3 SiO (R_2SiO)_m Si(R^2)_3$  (where R, R<sup>1</sup>, and R<sup>2</sup> are methyl 30 or vinyl groups; at least two vinyl groups are present in one molecule; when

all R's in one molecule are methyl groups, then at least one R<sup>1</sup> and at least one R<sup>2</sup> are vinyl groups; when one R in one molecule is a methyl group, at least one of R<sup>1</sup> and R<sup>2</sup> is a vinyl group; 0.1 to 2% of all methyl and vinyl groups of one molecule are vinyl groups; and "m" is a number within the range of 50 to 5 1,000).

[0011] There are no special restrictions with regard to the viscosity of component (A) at 25°C, provided that it is liquid at room temperature, but from the point of view of better conditions for curing the composition and 10 improved physical properties of a cured body, it is recommended to have the viscosity within the range of 50 to 1,000,000 mPa·s, preferably within the range of 200 to 500,000, and, even more preferably, within the range of 1,000 to 100,000 mPa·s.

15 [0012] Specific examples of aforementioned component (A) are the following: a dimethylpolysiloxane having both molecular terminals capped with dimethylvinylsiloxy groups, a dimethylsiloxane·methylvinylsiloxane copolymer capped at both molecular terminals with trimethylsiloxy groups, a dimethylsiloxane·methylvinylsiloxane copolymer capped at both molecular 20 terminals with dimethylvinylsiloxy groups, and a copolymer of a dimethylsiloxane·methylvinylsiloxane copolymer capped at both molecular terminals with a dimethylethoxysiloxy groups.

[0013] Silicon-bonded hydrogen atoms of the alkylhydrogenpolysiloxane (B) 25 that has more than two silicon-bonded hydrogen atoms in one molecule participate in a hydrosilation reaction with alkenyl groups of component (A) and, as a result, cross-links and cures component (A).

[0014] It is recommended that component (B) is liquid at room temperature. 30 It may have a linear, branched, or cyclic molecular structure. Examples of the

silicon-bonded alkyl groups are the same alkyl groups as given above for component (A), of which methyl groups are preferable. When a molecule of component (A) contains two alkenyl groups, it is recommended to have three or more silicon-bonded hydrogen atoms. When a molecule of component (A)

5 contains three alkenyl groups, it is necessary to have two or more silicon-bonded hydrogen atoms. From the view point of better hardness of the silicone elastomer, it is recommended that the number of the silicon-bonded hydrogen atoms in one molecule does not exceed 5.

From the point of view of tackiness of the silicone elastomer and peelability

10 thereof from heat-generating and heat-radiating members, it is recommended that the mole ratio of silicon-bonded hydrogen atoms of component (B) to alkenyl groups of component (A) is within the range of 3.0 to 0.5, preferably, 2.0 to 0.6, and, even more preferably, 1.5 to 0.7.

15 [0015] The following are specific examples of component (B): a methylhydrogenpolysiloxane capped at both molecular terminals with trimethylsiloxy groups, a methylhydrogensiloxane-dimethylsiloxane copolymer capped at both molecular terminals with trimethylsiloxy groups, a dimethylpolysiloxane capped at both molecular terminals with 20 dimethylhydrogensiloxy groups; a methylhydrogenpolysiloxane capped at both molecular terminals with dimethylhydrogensiloxy groups, a methylhydrogensiloxane-dimethylsiloxane copolymer capped at both molecular terminals with dimethylhydrogensiloxy groups, a cyclic methylhydrogenpolysiloxane, tetra(dimethylhydrogensiloxy)silane, and 25 methyltri(dimethylhydrogensiloxy)silane.

[0016] The platinum metal type catalyst (C) is used for accelerating the hydrosilation reaction between alkenyl groups of component (A) and silicon-bonded hydrogen atoms of component (B). Catalyst (C) may be 30 exemplified by a fine platinum metal powder, chloroplatinic acid, a complex of

chloroplatinic acid with  $\beta$ -diketone, a complex a chloroplatinic acid with olefin, a complex of a chloroplatinic acid with 1,3-divinyltetramethyldisiloxane, a complex of platinum with 1,3-divinyltetramethyldisiloxane, a thermoplastic silicone resin powder that contains aforementioned catalysts, a rhodium

5 compound, such as those expressed by formulae:  $\text{RhCl}(\text{Ph}_3\text{P})_3$ ,  $\text{RhCl}_3[\text{S}(\text{C}_4\text{H}_9)_2]_3$ , etc.; tetrakis(triphenyl)palladium, a mixture of palladium black and triphenylphosphine, etc.

According to the present invention, the catalyst has to be added in the amount sufficient for cross-linking components (A) and (B). For example, it can be 10 added in the amount of 0.01 to 500 parts by weight, preferably, 0.1 to 100 parts by weight per  $10^6$  parts by weight of the sum of components (A) and (B).

[0017] The silicone elastomer being a cured body of a hydrosilation-curable organopolysiloxane composition consisting of (A) an alkylalkenylpolysiloxane

15 having at least two alkenyl groups in one molecule, (B) an alkylhydrogenpolysiloxane having at least two hydrogen atoms in one molecule, and (C) a platinum metal type catalyst, constitutes 90 to 10 wt.%, preferably, 80 to 20 wt.%, of the thermoconductive silicone elastomer of the present invention. The hydrosilation-curable organopolysiloxane composition

20 that consists of (A) an alkylalkenylpolysiloxane having at least two alkenyl groups in one molecule, (B) an alkylhydrogenpolysiloxane having at least two hydrogen atoms in one molecule, and (C) a platinum metal type catalyst constitutes 90 to 10 wt.%, preferably, 80 to 20 wt.% of the aforementioned hydrosilation-curable thermoconductive silicone elastomer composition.

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[0018] Fine reinforcement silica powder (D) is used for improving viscosity of the (A) and (B) mixture, as well as for improving mechanical strength of the thermoconductive silicone elastomer. This typically may be fumed silica (dry-process silica or aerosol silica) or precipitated silica (precipitation-process silica). In view of their thickening action, the fumed silica and precipitated

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silica, and, especially, fumed silica, can be hydrophobized with an organic silicon compound (such as a trimethylchlorosilane, dimethyldichlorosilane, hexamethyldisiloxane, and octamethylcyclotetrasiloxane).

Component (D) is dispersed in the thermoconductive silicone elastomer in an 5 amount of 0.2 to 5 wt.%, and is contained in the hydrosilation-curable thermoconductive silicone elastomer composition, also in a dispersed form, in an amount of 0.2 to 5.0 wt.%, preferably, 0.5 to 3 wt.%.

[0019] The thermoconductive inorganic powder (E) imparts

10 thermoconductive properties to the silicone elastomer. This component may be exemplified by an alumina powder, crystalline silica powder, zinc oxide powder, magnesium oxide powder, aluminum nitride, etc. Component (E) may have an average particle size within the range of 1 to 50  $\mu\text{m}$ .

15 Typical representatives of the alumina powder are an amorphous alumina powder and spherical alumina. The crystalline silica powder can be exemplified by an amorphous crystalline silica powder and a spherical crystalline silica powder.

[0020] The amorphous alumina powder is mainly an  $\alpha$ -alumina powder

20 obtained by crushing. The spherical alumina powder has an average diameter of particles within the range of 1 to 50  $\mu\text{m}$ . This alumina powder is used mainly in the form of an  $\alpha$ -alumina powder obtained by a high-temperature melt spray method or by hydrothermal treatment of an alumina hydrate. In the context of the present patent application, the term "spherical" covers not 25 only strictly spherical shapes, but also substantially spherical particles, i.e., 90% or more of the particles have a shaped factor of 1.0 to 1.4. The aforementioned shape factor is determined according to JIS R 6002 by measuring diameters of 200 particles under a microscope and then calculating an average ratio of the larger diameter to the perpendicular smaller diameter 30 of the particle. In other words, an ideally spherical particle will have the ratio

equal to 1.0, and the aforementioned shape factor shows a degree of deviation from 1.0.

[0021] Component (E) is contained in the thermoconductive silicone

5 elastomer and in the hydrosilation-curable thermoconductive silicone  
elastomer composition in a dispersed state. If the content of component (E) in  
the hydrosilation-curable thermoconductive silicone elastomer composition is  
too high, the composition becomes very viscous, and if the content of  
component (E) is too low, it is difficult to obtain a cured body of high thermal  
10 conductivity. Therefore, it is recommended to add component (E) to the  
composition in the amount of 10 to 90 wt.%, preferably 30 to 80 wt.% per total  
weight of the thermoconductive silicone elastomer composition.

[0022] Component (E) may be surface-treated with methyltrimethoxysilane,

15 vinyltrimethoxysilane, dimethyldimethoxysilane,  
3-glycidoxypolytrimethoxysilane, 3-methacryloxypropyltrimethoxysilane,  
hexamethyldisilazane, or a similar silane-type surface-active agent or an  
organosiloxane-type surface treatment agent.

20 [0023] The alkylphenylpolysiloxane (F) that is liquid at room temperature

is an agent that facilitates separation of the thermoconductive silicone  
elastomer from the heat-generating member and heat-radiating member  
between which the elastomer was sandwiched and maintained in a  
sandwiched state over a long period of time. Alkyl groups of this

25 alkylphenylpolysiloxane are the same as alkyl groups of component (A).

Component (F) can typically be represented by the following formula:

$(R^4)_3SiO[(R^3)_2SiO]_nSi(R^4)_3$  (where  $R^3$  and  $R^4$  are methyl or phenyl groups; 1  
to 50 % of the total number of methyl groups and phenyl groups in one  
molecule are phenyl groups; "n" is a number within the range of 1 to 100).

30 However, component (F) may also be represented by a branched

methylphenylpolysiloxane of the following formula:  $(R^3)Si[OSi(R^4)_3]_3$  (where  $R^3$  and  $R^4$  designate methyl group or phenyl group and where one molecule contains at least one methyl group and at least one phenyl group), or a cyclic methylphenylpolysiloxane.

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[0024] The following are specific examples of component (F): a methylphenylpolysiloxane capped at both molecular chains with trimethylsiloxy groups, a dimethylsiloxane-methylphenylsiloxane copolymer capped at both molecular terminals with trimethylsiloxy groups, a

10 dimethylpolysiloxane capped at both molecular terminals with dimethylphenylsiloxy groups, a methylphenylpolysiloxane capped at both molecular terminals with dimethylphenylsiloxy groups, a dimethylsiloxane-methylphenylsiloxane copolymer capped at both molecular terminals with dimethylphenylsiloxy groups, a phenyltris(dimethylphenylsiloxy)silane, and  
15 methyltri(dimethylphenylsiloxy)silane.

[0025] In the hydrosilation-curable thermoconductive silicone elastomer, component (F) should be used in a dispersed state in the amount of 0.1 to 10 wt.%, preferably, 1 to 6 wt.%. In the hydrosilation-curable thermoconductive  
20 silicone elastomer composition, component (F) also should be used in the amount of 0.1 to 10 wt.%, preferably, 1 to 6 wt.%.

[0026] The hydrosilation-curable thermoconductive silicone elastomer composition of the present invention comprises aforementioned components

25 (A) through (F), but in order to limit thickening at room temperature and to prolong the storage time, the composition may contain hydrosilation-reaction inhibitors, such an alkyne alcohol, enyne compound, benzotriazole, etc. The following are specific examples of such compounds: 3-methyl-1-butyn-3-ol, 3,5-dimethyl-1-hexyn-3-ol, 3-phenyl-1-butyn-3-ol, or similar acetylene-type  
30 compounds; 3-methyl-3-penten-1-yne, 3,5-dimethyl-3-hexen-1-yne, or similar

enyne compounds; 1,3,5,7-tetramethyl-1,3,5,7-tetravinyl cyclotetrasiloxane, 1,3,5,7-tetramethyl-1,3,5,7-tetrahexenyl cyclotetrasiloxane, benzotriazole, or similar triazoles, phosphines, mercaptanes, and hydrazines. The content of such inhibitors in the hydrosilation-curable thermoconductive silicone elastomer 5 composition may be within the range of 0.001 to 5 wt.%.

[0027] Within the limits not detrimental to the objects of the present invention, the thermoconductive silicone elastomer and hydrosilation-curable thermoconductive silicone elastomer composition of the present invention may 10 also incorporate, if necessary, various additives, such as iron oxide, cerium oxide, or similar heat-resistance imparting agents; silane coupling agents, their derivative, or similar adhesion promoters; iron oxide red, carbon black, or other pigments; fumed titanium dioxide, zinc carbonate, aluminum hydroxide, or other flame retardants.

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[0028] The hydrosilation-curable thermoconductive silicone elastomer composition of the present invention can be easily prepared by uniformly mixing all aforementioned components (A) through (F), if necessary, with other optional components, in a mixer with stirring blades. Alternatively, a 20 mixture can first be prepared by mixing components (A) and (D) with heating, then adding and mixing the product with components (E) and (F), and then compounding the mixture with components (B) and (C). If component (D) is not hydrophobized, in case of heating and premixing this component with component (A), it is recommended to subject component (D) to hydrophobic 25 surface treatment with hydrophobic treatment agents, such as a hexamethyldisiloxane, silanol-end-capped dimethylsiloxane oligomer, silanol-end-capped methylphenylsiloxane oligomer, trimethylsilanol, or the like.

[0029] The thermoconductive silicone elastomer of the present invention can be easily produced by holding the aforementioned hydrosilation-curable thermoconductive silicone elastomer composition at room temperature or with heating. Forming may be carried out by pressure molding, extruding, injection 5 molding of the liquid composition, or casting. There are no special restrictions with regard to the shape of the silicone elastomer product that can be made in the form of sheets, tapes, bars, cylinders, blocks, or irregular bodies.

[0030] The thermoconductive silicone elastomer of the invention should has 10 a hardness of 5 to 70 measured by a spring-type hardness tester in accordance with the provisions of SRIS 0101-1968, a tensile strength of no less than 0.2 MPa in accordance with JIS K 6251, a elongation of no less than 300%, and should be capable of peeling at interfaces from a heat-generating member and a heat-radiating member after being sandwiched in an adhered state between 15 these members and held in this state for 48 hours at 120°C.

[0031] It is recommended that the thermoconductive silicone elastomer of the present invention has a hardness of 5 to 70, preferably, 10 to 65, measured by a spring-type hardness tester which is Asukar-C hardness tester, available 20 from Kobushikeiki Co., Ltd., in accordance with the provisions of SRIS 0101-1968. Since such hardness 70 corresponds to 45 units of a durometer type A hardness according to JIS K6253, the elastomer of the present invention is softer than a conventional silicone rubber, and its hardness holds an intermediate position between silicone rubber and silicone gel.

25 [0032] Furthermore, since the thermoconductive silicone elastomer of the present invention has a tensile strength of no less than 0.2 MPa in accordance with JIS K 6251 and a elongation of no less than 300%, it can be easily formed into a 0.5 to 3 mm thick sheet, tape, bar, cylinder, ring, etc. The 30 thermoconductive elastomer of the present invention is tacky and has

shearing adhesive strength measured in accordance with JIS K 6850 with regard to a flat sheet, e.g., an aluminum plate and a glass plate, within the range of 5 to 30 N/cm<sup>2</sup>, preferably, 7 to 25 N/cm<sup>2</sup>. Therefore, even though the elastomer can be adhered to a heat-generating member and a heat-radiating member, it also can be easily disconnected from these members. The elastomer of the present invention has thermal conductivity within the range of 0.5 to 3.0 W/m·K.

[0033] The thermoconductive silicone elastomer of the present invention can be used as a thermoconductive medium between a semiconductor chip and heat-radiating plate, as a thermoconductive medium between a heat-radiating plate and a printed circuit board that may supports transistors, IC, hybrid IC's, memory elements, or similar heat-generating electronic elements, as a thermoconductive medium between a plasma display and a heat-radiating plate, or as a thermoconductive element of a DC-DC converter. The aforementioned heat-radiating plate may be made from aluminum, duralumin, stainless steel, magnesium alloy, copper, etc. Furthermore, the hydrosilation-curable thermoconductive elastomer composition of the present invention is suitable for the manufacture of the aforementioned thermoconductive silicone elastomer. Such an elastomer can also be formed by curing the composition between heat-generating and heat-radiating members.

### Examples

[0034] The present invention is further described in more detail with reference to practical and comparative examples. All characteristics given in the subsequent practical and comparative examples were obtained at 25°C. Tackiness, peelability, hardness, elongation, and thermal conductivity of the thermoconductive silicone elastomers were evaluated by the methods described below.

[0035] [Tackiness and Peelability of the Thermoconductive Silicone Elastomer]

Tackiness was evaluated by placing a 1 mm-thick layer of the hydrosilation-curable thermoconductive silicone elastomer composition

5 between a float plate glass and an aluminum plate (A1050P), and then curing the composition by heating for 30 min. at 100°C. Tackiness of the obtained thermoconductive silicone elastomer was measured in accordance with JIS K 6850 by means of a universal tester Tensilon (RTC-1325A, made by Orientech Co.).

10 Peelability was evaluated visually by observing whether the thermoconductive silicone elastomer left some material on the surfaces of float plate glass and aluminum plate (A1050P) used in the tackiness evaluation, or if it was completely peeled off from the aforementioned surfaces.

15 [0036] [Hardness of the Thermoconductive Silicone Elastomer]

The hydrosilation-curable thermoconductive silicone elastomer composition was subjected to pressure vulcanization for 30 min. at 100°C. Two 6-mm thick sheets of the obtained thermoconductive silicone elastomer were stacked, one onto the other, and used for measuring hardness at 1 kg load on a spring-type 20 hardness tester which is Asukar-C hardness tester, available from Kobushikeiki Co., Ltd., in accordance with the procedure of SRIS 0101-1968 for testing physical properties of expanded rubbers.

25 [0037] [Tensile Strength and Elongation of the Thermoconductive Silicone Elastomer]

The hydrosilation-curable thermoconductive silicone elastomer composition was subjected to pressure vulcanization for 15 min. at 100°C. The obtained 2 mm-thick sheet of the thermoconductive silicone elastomer was used for measuring a tensile strength and elongation on dumbbell No. 4 specimens in

accordance with JIS K 6251 on a tensile strength tester ST102-1, available from Ueshima Seisakusho Co., Ltd..

[0038] [Coefficient of Thermal Conductivity of the Thermoconductive

5 Silicone Elastomer]

The hydrosilation-curable thermoconductive silicone elastomer composition was subjected to pressure vulcanization for 30 min. at 100°C. The obtained 15 mm-thick sheet of the thermoconductive silicone elastomer was used for measuring a coefficient of thermal conductivity by means of a Quick Thermal 10 Conductivity Meter, Model QTM-500, available from Kyoto Electronics Manufacturing Co., Ltd.

[0039] [Practical Example 1]

A mixer with stirring blades was loaded with 26.97 wt.% of a

15 dimethylvinylsiloxy-capped dimethylpolysiloxane having a viscosity of 40,000 mPa·s (vinyl group content = 0.09 wt.%), 0.98 wt.% of a trimethylsiloxy-capped dimethylsiloxane-methylhydrogensiloxane copolymer having a viscosity of 25 mPa·s and containing on average 3 silicon-bonded hydrogen atoms in the side molecular chains (the content of silicon-bonded 20 hydrogen atoms = 0.13 wt.%), 0.15 wt.% of a solution of a platinum-1,3-divinyl-1,1,3,3-tetramethyldisiloxane complex with 0.5 wt.% content of metallic platinum, 0.05 wt.% of an addition reaction inhibitor in the form of 2-phenyl-3-butyn-2-ol, 1.4 wt.% of a fumed silica surface-treated with hexamethyldisilazane having a specific surface area of 200 m<sup>2</sup>/g, 67.5 wt.% of 25 an irregular shaped alumina powder with a 11 µm average particle size, and 3.0 wt.% of a trimethylsiloxy-capped methylphenylsiloxane-dimethylsiloxane copolymer having a viscosity of 500 mPa·s (the content of phenyl groups = 9.4 wt.%; (phenyl group number is 5% of the total sum of the methyl and phenyl groups). The components were uniformly mixed to produce a 30 hydrosilation-curable thermoconductive silicone elastomer composition. The

composition was evaluated with regard to tackiness, peelability, hardness, tensile strength, elongation, and coefficient of thermal conductivity. The results of measurement are shown in Table 1.

5 [0040] [Practical Example 2]

A mixer with stirring blades was loaded with 28.46 wt.% of a dimethylvinylsiloxy-capped dimethylpolysiloxane having a viscosity of 40,000 mPa·s (vinyl group content = 0.09 wt.%), 1.04 wt.% of a trimethylsiloxy-capped dimethylsiloxane-methylhydrogensiloxane copolymer 10 having a viscosity of 25 mPa·s and containing on average 3 silicon-bonded hydrogen atoms in the side molecular chains (the content of silicon-bonded hydrogen atoms = 0.13 wt.%), 0.15 wt.% of a solution of a platinum-1,3-divinyl-1,1,3,3-tetramethyldisiloxane complex with 0.5 wt.% content of metallic platinum, 0.05 wt.% of an addition reaction inhibitor in the 15 form of 2-phenyl-3-butyn-2-ol, 1.4 wt.% of a fumed silica surface-treated with hexamethyldisilazane having a specific surface area of 200 m<sup>2</sup>/g, 67.5 wt.% of an irregular shaped alumina powder with a 11 µm average particle size, and 1.0 wt.% of a trimethylsiloxy-capped copolymer of a methylphenylsiloxane-dimethylsiloxane copolymer having a viscosity of a 125 mPa·s (the content of 20 phenyl groups = 34.2 wt.%; (phenyl group number is 25% of the total sum of the methyl and phenyl groups). The components were uniformly mixed to produce a hydrosilation-curable thermoconductive silicone elastomer composition. The composition was evaluated with regard to tackiness, peelability, hardness, tensile strength, elongation, and coefficient of thermal 25 conductivity. The results of measurement are shown in Table 1.

[0041] [Practical Example 3]

A mixer with stirring blades was loaded with 26.97 wt.% of a dimethylvinylsiloxy-capped dimethylpolysiloxane having a viscosity of 40,000 mPa·s (vinyl group content = 0.09 wt/%), 0.98 wt.% of a 30

trimethylsiloxy-capped dimethylsiloxane-methylhydrogensiloxane copolymer having a viscosity of 25 mPa·s and containing on average 3 silicon-bonded hydrogen atoms in the side molecular chains (the content of silicon-bonded hydrogen atoms = 0.13 wt.%), 0.15 wt.% of a solution of a

5 platinum-1,3-divinyl-1,1,3,3-tetramethyldisiloxane complex with 0.5 wt.% content of metallic platinum, 0.05 wt.% of an addition reaction inhibitor in the form of 2-phenyl-3-butyn-2-ol, 1.4 wt.% of a fumed silica surface-treated with hexamethyldisilazane having a specific-surface area of 200 m<sup>2</sup>/g, 67.5 wt.% of an irregular shaped alumina powder with a 11 µm average particle size, and

10 3.0 wt.% of a trimethylsiloxy-capped methylphenylpolysiloxane having a viscosity of 500 mPa·s (the content of phenyl groups = 55.7 wt.%; (phenyl group number is 50% of the total sum of the methyl and phenyl groups). The components were uniformly mixed to produce a hydrosilation-curable thermoconductive silicone elastomer composition. The composition was

15 evaluated with regard to tackiness, peelability, hardness, tensile strength, elongation, and coefficient of thermal conductivity. The results of measurement are shown in Table 1.

[0042] [Practical Example 4]

20 A mixer with stirring blades was loaded with 26.97 wt.% of a dimethylvinylsiloxy-capped dimethylpolysiloxane having a viscosity of 40000 mPa·s (vinyl group content = 0.09 wt.%), 0.98 wt.% of a trimethylsiloxy-capped dimethylsiloxane-methylhydrogensiloxane copolymer having a viscosity of 25 mPa·s and containing on average 3 silicon-bonded

25 hydrogen atoms in the side molecular chains (the content of silicon-bonded hydrogen atoms = 0.13 wt.%), 0.15 wt.% of a solution of a platinum-1,3-divinyl-1,1,3,3-tetramethyldisiloxane complex with 0.5 wt.% content of metallic platinum, 0.05 wt.% of an addition reaction inhibitor in the form of 2-phenyl-3-butyn-2-ol, 1.4 wt.% of a fumed silica surface-treated with

30 hexamethyldisilazane having a specific-surface area of 200 m<sup>2</sup>/g, 67.5 wt.% of

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an irregular shaped crystalline silica powder with a 18  $\mu\text{m}$  average particle size, and 3.0 wt.% of a trimethylsiloxy-capped methylphenylsiloxane-dimethylsiloxane copolymer having a viscosity of a 125 mPa·s (the content of phenyl groups = 9.4 wt.%; (phenyl group number is 5% of the total sum of the 5 methyl and phenyl groups). The components were uniformly mixed to produce a hydrosilation-curable thermoconductive silicone elastomer composition. The composition was evaluated with regard to tackiness, peelability, hardness, tensile strength, elongation, and coefficient of thermal conductivity. The results of measurement are shown in Table 1.

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[0043]

[Table 1]

Items and Characteristics				Pr. Ex. 1	Pr. Ex. 2	Pr. Ex. 3	Pr. Ex. 4	
Tackiness	Glass	Initial	N/cm <sup>2</sup>	17	15	25	20	
	Aluminum	Initial	N/cm <sup>2</sup>	12	7	13	15	
Peelability	Glass	Initial	-	Interfacial separation	Interfacial separation	Interfacial separation	Interfacial separation	
		After 48 hours at 120°C	-	Interfacial separation	Interfacial separation	Interfacial separation	Interfacial separation	
	Aluminum	Initial	-	Interfacial separation	Interfacial separation	Interfacial separation	Interfacial separation	
		After 48 hours at 120°C	-	Interfacial separation	Interfacial separation	Interfacial separation	Interfacial separation	
Hardness			-	52	50	53	60	
Tensile Strength			MPa	0.9	1.0	0.9	0.5	
Elongation			%	850	650	740	350	
Coefficient of Thermal Conductivity			0.7	0.7	0.7	0.7	0.7	

[0044] [Comparative Example 1]

5 A mixer with stirring blades was loaded with 28.40 wt.% of a dimethylvinylsiloxy-capped methylvinylsiloxane·dimethylsiloxane copolymer having a viscosity of 40,000 mPa·s (vinyl group content = 0.13 wt.%), 18.91

wt.% of a dimethylvinylsiloxy-capped dimethylpolysiloxane having a viscosity of 10,000 mPa·s (vinyl group content = 0.13 wt.%),

1.94 wt.% of a trimethylsiloxy-capped methylhydrogenpolysiloxane having a viscosity of 20 mPa·s (the main molecular chain consists of 23

5 methylhydrogensiloxane units on average, and the content of silicon-bonded hydrogen atoms = 1.55 wt.%), 0.15 wt.% of a solution of a platinum-1,3-divinyl-1,1,3,3-tetramethyldisiloxane complex with 0.5 wt.% content of metallic platinum, 0.03 wt.% of an addition reaction inhibitor in the form of 2-phenyl-3-butyn-2-ol, 11.80 wt.% of a fumed silica surface-treated with hexamethyldisilazane having a specific-surface area of 200 m<sup>2</sup>/g, 35.50 wt.% of an irregular shaped crystalline silica powder with a 5 µm average particle size, 2.30 wt.% of an irregular shaped alumina powder with a 11 µm average particle size, and 0.97 wt.% of a trimethylsiloxy-capped methyldodecylsiloxane-methyltetradecansiloxane copolymer having a viscosity of 900 mPa·s (the content of dodecyl groups = 33.9 wt%; the content of tetradecan groups = 39.5 wt%; dodecyl group number is 25% of the total sum of the methyl, dodecyl, and tetradecan groups; tetradecan group number is 25% of the total sum of the methyl, dodecyl, and tetradecan groups). The components were uniformly mixed to produce a hydrosilation-curable 10 15 20 25 thermoconductive silicone elastomer composition. The composition was evaluated with regard to tackiness, initial peelability, hardness, tensile strength, elongation, and coefficient of thermal conductivity. The results of measurement are shown in Table 2. This comparative example is based on Practical Example 2 of JP Kokai S56-2349.

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[0045] [Comparative Example 2]

A mixer with stirring blades was loaded with 43.75 wt.% of a dimethylvinylsiloxy-capped dimethylpolysiloxane having a viscosity of 3,000 mPa·s (vinyl group content = 0.16 wt.%), 1.96 wt.% of a

30 trimethylsiloxy-capped methylhydrogenpolysiloxane having a viscosity of 20

mPa·s (the main molecular chain consists of 23 methylhydrogensiloxane units on average, and the content of silicon-bonded hydrogen atoms = 1.55 wt.%), 0.15 wt.% of a solution of a platinum-1,3-divinyl-1,1,3,3-tetramethyldisiloxane

complex with 0.5 wt.% content of metallic platinum, 0.03 wt.% of an addition

5 reaction inhibitor in the form of 2-phenyl-3-butyn-2-ol, 1.4 wt.% of a fumed  
silica surface-treated with hexamethyldisilazane having a specific-surface  
area of 200 m<sup>2</sup>/g, 52.33 wt.% of an irregular shaped crystalline silica powder  
with a 5 µm average particle size, 1.00 wt.% of a titanium oxide powder with a  
1 µm average particle size, and 0.78 wt.% of a trimethylsiloxy-capped  
10 methylphenylpolysiloxane having a viscosity of 500 mPa·s (the content of  
phenyl groups = 55.7 wt.%; (phenyl group number is 50% of the total sum of  
the methyl and phenyl groups). The components were uniformly mixed to  
produce a hydrosilation-curable thermoconductive silicone elastomer  
composition. The composition was evaluated with regard to tackiness, initial  
15 peelability, hardness, tensile strength, elongation, and coefficient of thermal  
conductivity. The results of measurement are shown in Table 2. This  
comparative example is based on Practical Example 3 of JP Kokai S56-2349.

[0046] [Comparative Example 3]

20 A mixer with stirring blades was loaded with 43.75 wt.% of a  
dimethylvinylsiloxy-capped dimethylpolysiloxane having a viscosity of 3,000  
mPa·s (vinyl group content = 0.16 wt.%), 1.96 wt.% of a  
trimethylsiloxy-capped methylhydrogenpolysiloxane having a viscosity of 20  
mPa·s (the main molecular chain consists of 23 methylhydrogensiloxane units  
25 on average, and the content of silicon-bonded hydrogen atoms = 1.55 wt.%),  
0.15 wt.% of a solution of a platinum-1,3-divinyl-1,1,3,3-tetramethyldisiloxane  
complex with 0.5 wt.% content of metallic platinum, 0.03 wt.% of an addition  
reaction inhibitor in the form of 2-phenyl-3-butyn-2-ol, 52.33 wt.% of an  
irregular shaped crystalline silica powder with a 5 µm average particle size,

1.00 wt.% of a titanium oxide powder with a 1  $\mu\text{m}$  average particle size, and 0.97 wt.% of a

trimethylsiloxy-capped dimethylsiloxane-methyloctylsiloxane copolymer

having a viscosity of 20 mPa·s (the content of octyl groups = 28.3 wt.%; (octyl

5 group number is 15% of the total sum of the methyl and octyl groups). The components were uniformly mixed to produce a hydrosilation-curable thermoconductive silicone elastomer composition. The composition was evaluated with regard to tackiness, initial peelability, hardness, tensile strength, elongation, and coefficient of thermal conductivity. The results of 10 measurement are shown in Table 2. This comparative example is based on Practical Example 3 of JP Kokai S56-2349.

[0047]

[Table 2]

Items and Characteristics			Co. Ex. 1	Co. Ex. 2	Co. Ex. 3
Tackiness	Glass	N/cm <sup>2</sup>	Fall	40	40
	Aluminum	N/cm <sup>2</sup>	Fall	98	155
Peelability	Glass	-	Interfacial separation	Cohesive failure	Cohesive failure
	Aluminum	-	Interfacial separation	Interfacial separation	Interfacial separation
Hardness		-	74	64	62
Tensile Strength		MPa	4.8	3.3	3.1
Elongation		%	111	176	190
Coefficient of Thermal Conductivity		W/m·K	0.2	0.4	0.4

### Industrial Applicability

[0044] The thermoconductive silicone elastomer of the present invention can be used as a thermoconductive medium between a semiconductor chip and

heat-radiating plate, as a thermoconductive medium between a heat-radiating plate and a printed circuit board that may supports transistors, IC, hybrid IC's, memory elements, or similar heat-generating electronic elements, as a thermoconductive medium between a plasma display and a heat-radiating plate, or as a thermoconductive element of a DC-DC converter. Furthermore, the hydrosilation-curable thermoconductive elastomer composition of the present invention is suitable for the manufacture of the aforementioned thermoconductive silicone elastomer and thermoconductive medium. The thermoconductive medium of the present invention is useful for heat-releasing or heat-sinking.

## CLAIMS

1. A thermoconductive silicone elastomer that has a hardness of 5 to 70 measured by a spring-type hardness tester in accordance with the provisions of SRIS 0101-1968, a tensile strength of no less than 0.2 MPa in accordance with JIS K 6251, a elongation of no less than 300%, and is capable of peeling at interfaces from a heat-generating member and a heat-radiating member after being sandwiched in an adhered state between said members and held in this state for 48 hours at 120°C, said thermoconductive silicone elastomer comprising: 90 to 10 wt.% of a silicone elastomer being a cured body of a hydrosilation-curable organopolysiloxane composition consisting of (A) an alkylalkenylpolysiloxane having at least two alkenyl groups in one molecule and (B) an alkylhydrogenpolysiloxane having at least two hydrogen atoms in one molecule and (C) a platinum metal type catalyst; (D) 0.2 to 5.0 wt.% of a finely powdered reinforcement silica; (E) 10 to 90 parts by weight of a conductive inorganic powder; and (F) 0.1 to 10 wt.% of an alkylphenylpolysiloxane which is liquid at room temperature (100 wt.% total), where components (D), (E) and (F) are dispersed in said silicone elastomer.
- 20 2. The thermoconductive silicone elastomer of Claim 1, wherein component (A) has a linear molecular structure.
- 25 3. The thermoconductive silicone elastomer of Claim 1, wherein component (A) is a methylvinylpolysiloxane of the following formula:  $(R^1)_3SiO(R_2SiO)_mSi(R^2)_3$  (where R, R<sup>1</sup>, R<sup>2</sup> are methyl or vinyl groups; at least two vinyl groups being present in one molecule; when all R's in one molecule are methyl group, at least one R<sup>1</sup> and at least one R<sup>2</sup> are vinyl groups; when one R in one molecule is a methyl group, at least one of R<sup>1</sup> and R<sup>2</sup> is a vinyl group; 0.1 to 2% of the total number of methyl groups and vinyl groups of

one molecule are vinyl groups; and "m" is a number within the range of 50 to 1,000); component (B) is a methylhydrogenpolysiloxane; component (D) is a fumed silica; component (E) is an alumina powder or a crystalline silica powder; and component (F) is a methylphenylpolysiloxane of the following formula:  $(R^4)_3SiO[(R^3)_2SiO]_nSi(R^4)_3$  (where  $R^3$  and  $R^4$  are methyl or phenyl groups; 1 to 50 % of the total number of methyl groups and phenyl groups in one molecule are phenyl groups; and «n» is a number within the range of 1 to 100).

10 4. A hydrosilation-curable thermoconductive silicone elastomer composition comprising: 90 to 10 wt.% of a hydrosilation-curable organopolysiloxane composition consisting of (A) an alkylalkenylpolysiloxane having at least two alkenyl groups in one molecule, (B) an alkylhydrogenpolysiloxane having at least two hydrogen atoms in one molecule, and (C) a platinum metal type catalyst; (D) 0.2 to 5.0 wt.% of a finely powdered reinforcement; (E) 10 to 90 parts by weight of a conductive inorganic powder; (F) 0.1 to 10 wt.% of an alkylphenylpolysiloxane which is liquid at room temperature (100 wt.% total); said composition, when cured, forming a thermoconductive silicone elastomer that has a hardness of 5 to 70 measured by a spring-type hardness tester in accordance with the provisions of SRIS 0101-1968, a tensile strength of no less than 0.2 MPa in accordance with JIS K 6251, a elongation of no less than 300%, and is capable of peeling at interfaces from a heat-generating member and a heat-radiating member after being sandwiched in an adhered state between said members and held in this state for 48 hours at 120°C.

25 5. The hydrosilation-curable thermoconductive silicone elastomer composition of Claim 4, wherein component (A) has a linear molecular structure.

6. The hydrosilation-curable thermoconductive elastomer composition of Claim 4, wherein component (A) is a methylvinylpolysiloxane of the following formula:

(R<sup>1</sup>)<sub>3</sub>SiO (R<sub>2</sub>SiO)<sub>m</sub> Si(R<sup>2</sup>)<sub>3</sub> (where R, R<sup>1</sup>, R<sup>2</sup> are methyl or vinyl groups; at least two vinyl groups being present in one molecule; when all R's in one molecule are methyl group, at least one R<sup>1</sup> and at least one R<sup>2</sup> are vinyl groups; when one R in one molecule is a methyl group, at least one of R<sup>1</sup> and R<sup>2</sup> is a vinyl group; 0.1 to 2% of the total number of methyl groups and vinyl groups of one molecule are vinyl groups; and "m" is a number within the range of 50 to 1,000); component (B) is a methylhydrogenpolysiloxane; component (D) is a fumed silica; component (E) is an alumina powder or a crystalline silica powder; and component (F) is a methylphenylpolysiloxane of the following formula: (R<sup>4</sup>)<sub>3</sub>SiO[(R<sup>3</sup>)<sub>2</sub>SiO]<sub>n</sub> Si(R<sup>4</sup>)<sub>3</sub> (where R<sup>3</sup> and R<sup>4</sup> are methyl or phenyl groups; 1 to 50 % of the total number of methyl groups and phenyl groups in one molecule are phenyl groups; "n" is a number within the range of 1 to 100).

7. A thermoconductive medium comprising the thermoconductive silicone elastomer of one of Claim 1 to Claim 3.

# INTERNATIONAL SEARCH REPORT

Intel ~~l~~ Application No  
PCT/JP2005/016170

A. CLASSIFICATION OF SUBJECT MATTER  
C08L83/04 H01B3/46

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)  
C08L H01B

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 practical, search terms used)

EPO-Internal, PAJ, 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	JP 56 002349 A (TOSHIBA SILICONE CO LTD) 12 January 1981 (1981-01-12) example 3 ----- X PATENT ABSTRACTS OF JAPAN vol. 005, no. 049 (C-049), 8 April 1981 (1981-04-08) & JP 56 002349 A (TOSHIBA SILICONE CO LTD), 12 January 1981 (1981-01-12) abstract -----	1-7
A	US 4 444 944 A (MATSUSHITA ET AL) 24 April 1984 (1984-04-24) tables 1,2 -----	1-7
A	US 5 008 307 A (INOMATA ET AL) 16 April 1991 (1991-04-16) column 7, lines 14-17; table 1 -----	1-7

Further documents are listed in the continuation of box C.

Patent family members are listed in annex.

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Date of the actual completion of the international search

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Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,  
Fax: (+31-70) 340-3016

Authorized officer

Buestrich, R

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/JP2005/016170

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