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(54) **THERMALLY CONDUCTIVE SILICONE GEL COMPOSITION, THERMALLY CONDUCTIVE SILICONE SHEET, AND PRODUCTION METHODS THEREFOR**

WÄRMELEITENDE SILIKONGELZUSAMMENSETZUNG, WÄRMELEITENDE SILIKONFOLIE UND VERFAHREN ZU IHRER HERSTELLUNG

COMPOSITION DE GEL DE SILICONE THERMOCONDUCTRICE, FEUILLE DE SILICONE THERMOCONDUCTRICE, ET LEURS PROCÉDÉS DE PRODUCTION

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Description

Technical Field

[0001] The present invention relates to a thermally conductive silicone gel composition that is suitable to be interposed between a heat generating member and a heat dissipating material of electrical and electronic components or the like. More preferably, the present invention relates to a thermally conductive silicone gel composition and a thermally conductive silicone sheet having a low dielectric constant, and a production method for the same.

BackgroundArt

[0002] With the significant improvement in performance of semiconductors such as CPUs in recent years, the amount of heat generated by them has become extremely large. For this reason, heat dissipating materials are attached to electronic components that may generate heat, and a thermally conductive silicone gel is used between heat generating members such as semiconductors and the heat dissipating materials. The thermally conductive silicone gel is required to have thermal conductive properties, electromagnetic wave absorbing properties, and noise prevention properties as devices become smaller in size, more sophisticated, and more highly integrated At the same time, to address global environmental issues, substances to be used need to comply with the Globally Harmonized System of Classification and Labelling of Chemicals (hereinafter, also referred to as "GHS"), which specifies criteria for classifying hazardous substances and labelling methods therefor (e.g., safety data sheets). Alumina, crystalline silica, and magnesium oxide, which are thermally conductive fillers, are classified as hazardous under GHS criteria and are not preferred Patent Document 1 proposes adding crystalline silica particles and aluminum hydroxide particles to acrylic resin to form a low dielectric constant sheet. Patent Documents 2 to 4 propose adding alumina particles and plural kinds of aluminum hydroxide particles to organopolysiloxane to prepare a thermally conductive silicone composition.

Prior Art Documents

Patent Documents

[0003]

Patent Document 1: JP 2020-077777 A
Patent Document 2: JP 5304588 B2
Patent Document 3: JP 2020-002236 A
Patent Document 4: JP 2020-066713 A

Disclosure of Invention

Problem to be Solved by the Invention

[0004] Crystalline silica contained in a composition of Patent Document 1 is classified as hazardous under GHS criteria. Alumina contained in the compositions of Patent Documents 2 to 4 is classified as hazardous under GHS criteria, and these documents are silent as to making low dielectric constant sheets.

[0005] To solve the above conventional problems, the present invention provides a thermally conductive silicone gel composition and a thermally conductive silicone sheet having a low dielectric constant that are composed of substances not classified as hazardous under GHS criteria and thus highly safe while having flexibility, and a production method for the same.

Means for Solving Problem

[0006] A thermally conductive silicone gel composition according to a first aspect of the present invention contains:

(A) an organopolysiloxane having two or more alkenyl groups per molecule;
(B) an organohydrogenpolysiloxane having two or more hydrogen atoms bonded directly to silicon atoms;
(C) a platinum group metal-containing curing catalyst in a catalytic amount; and
(D) aluminum hydroxide particles as thermally conductive particles in an amount of 200 to 600 parts by mass with respect to 100 parts by mass of a total amount of the components (A) to (C), the aluminum hydroxide particles containing, based on 100% by mass of the aluminum hydroxide particles, aluminum hydroxide particles (D-1) having

a median diameter (D50) of 30 μm or more in an amount of more than 45% by mass and less than 75% by mass; and aluminum hydroxide particles (D-2) having a median diameter (D50) of less than 30 μm in an amount of 25% by mass or more and 55% by mass or less, wherein the thermally conductive silicone gel composition is free from alumina particles, crystalline silica particles and magnesium particles, and wherein the thermally conductive silicone gel composition has a relative dielectric constant of 5.0 or less at a frequency of 1 MHz according to a mutual induction bridge method of JIS K 6911: 2006 and has a Shore OO hardness of 5 to 60.

[0007] A thermally conductive silicone sheet according to a second aspect of the present invention is a sheet of the thermally conductive silicone gel composition.

[0008] A method for producing the thermally conductive silicone sheet according to a third aspect of the present invention includes: mixing the following components; sheeting the mixture; and curing the sheet, the components of the mixture comprising:

(A) an organopolysiloxane having two or more alkenyl groups per molecule;
(B) an organohydrogenpolysiloxane having two or more hydrogen atoms bonded directly to silicon atoms;
(C) a platinum group metal-containing curing catalyst in a catalytic amount; and
(D) aluminum hydroxide particles as thermally conductive particles in an amount of 200 to 600 parts by mass with respect to 100 parts by mass of a total amount of the components (A) to (C), the aluminum hydroxide particles containing, based on 100% by mass of the aluminum hydroxide particles, aluminum hydroxide particles (D-1) having a median diameter (D50) of 30 μm or more in an amount of more than 45% by mass and less than 75% by mass; and aluminum hydroxide particles (D-2) having a median diameter (D50) of less than 30 μm in an amount of 25% by mass or more and 55% by mass or less, wherein the thermally conductive silicone gel composition is free from alumina particles, crystalline silica particles and magnesium particles, and

wherein the thermally conductive silicone sheet has a relative dielectric constant of 5.0 or less at a frequency of 1 MHz according to a mutual induction bridge method of JIS K 6911: 2006 and has a Shore OO hardness of 5 to 60.

Effect of the Invention

[0009] The thermally conductive silicone gel composition and the thermally conductive silicone sheet of the present invention have high safety, flexibility, and a low dielectric constant by containing the components (A) to (D) that are not classified as hazardous under GHS criteria and satisfying a relative dielectric constant of 5.0 or less at a frequency of 1 MHz according to the mutual induction bridge method of JIS K 6911: 2006 and a Shore OO hardness of 5 to 60. The present invention further provides the production method for the same.

Brief Description of Drawings

[0010] [FIG. 1] FIGS. 1A and 1B are diagrams illustrating a method for measuring the thermal conductivity of a sample in an example of the present invention.

Description of the Invention

[0011] The component (A) of the present invention is an organopolysiloxane having two or more alkenyl groups per molecule. The component (A) is a base polymer component of the silicone gel composition. Preferably, the organopolysiloxane has two or more alkenyl groups per molecule, and the viscosity is 100 to 10000 Pa·s. The viscosity is measured with a B-type rotational viscometer at 23°C at a rotational speed of 20 rpm 30 seconds after start of rotation of a spindle.

[0012] The component (B) of the present invention is an organohydrogenpolysiloxane having two or more hydrogen atoms bonded directly to silicon atoms. The component (B) is a crosslinker component of the silicone gel composition. The number of moles of the hydrogen atoms bonded directly to silicon atoms is preferably 0.1 to 5.0 times the number of moles of the alkenyl groups of the component (A). The number of moles is more preferably 0.1 to 4.0 times, and further preferably 0.1 to 3.0 times the number of moles of the alkenyl groups of the component (A).

[0013] The component (C) of the present invention is a platinum group metal-containing curing catalyst. The catalyst is an addition reaction curing catalyst. The content of the component (C) is not limited particularly as long as it is a catalytic amount, and the content is preferably 0.1 to 1000 ppm with respect to the component A on a mass basis of the platinum group metal element.

[0014] The component (D) of the present invention is aluminum hydroxide particles as thermally conductive particles. The content of the component (D) is 200 to 600 parts by mass with respect to 100 parts by mass of a total amount of the components (A) to (C). The component (D) includes aluminum hydroxide particles with different median diameters

(D50). The aluminum hydroxide particles contain, based on 100% by mass of a total amount of the component (D), aluminum hydroxide particles (D-1) having a median diameter (D50) of 30 μm or more in an amount of more than 45% by mass and less than 75% by mass, and the content of the component (D-2) is more than 25% by mass and less than 55% by mass. By using the aluminum hydroxide particles with different median diameters (D50), small-size particles fill the spaces between large-size particles, which can provide nearly the closest packing and improve the thermal conductivity. The particle diameter as used herein refers to D50 (median diameter) at 50% in a volume-based cumulative particle size distribution, which is determined by a particle size distribution measurement with a laser diffraction scattering method. The measuring device may be, e.g., a laser diffraction/scattering particle size distribution analyzer LA-950 S2 manufactured by HORIBA, Ltd.

[0015] The thermally conductive silicone gel composition of the present invention has a relative dielectric constant of 5.0 or less, and preferably 4.8 or less, at a frequency of 1 MHz according to the mutual induction bridge method of JIS K 6911: 2006. This improves the electromagnetic wave absorbing properties and noise prevention properties of the thermally conductive silicone gel composition. The lower limit of the relative dielectric constant is 2.6 or more, and preferably 3.0 or more.

[0016] The thermally conductive silicone gel composition of the present invention has a Shore OO hardness of 5 to 60, and preferably 10 to 55. Thus, the thermally conductive silicone gel composition can exhibit flexibility. The thermally conductive silicone gel composition having flexibility can enhance adhesion between a heat generating member such as a semiconductor and a heat dissipating material and is suitably used as a thermal interface material (TIM).

[0017] The thermally conductive silicone gel composition of the present invention may further contain a surface treatment agent for the thermally conductive particles, as a component (E). Examples of the surface treatment agent include a titanate coupling agent, an aluminate coupling agent, a stearate coupling agent, an epoxy silane coupling agent, and an alkyl silane coupling agent. Among these, the alkyl silane coupling agent is preferred, and specifically, the alkyl silane coupling agent preferably contains at least one selected from an alkoxysilane compound expressed by $R_aSi(OR')_{4-a}$ (where R represents a substituted or unsubstituted organic group having 6 to 12 carbon atoms, R' represents an alkyl group having 1 to 4 carbon atoms, and a is 0 or 1), a partial hydrolysate of the alkoxysilane compound, and an alkoxy group-containing silicone having a substituted or unsubstituted organic group having 6 to 12 carbon atoms. Examples of the silane compound include hexyltrimethoxysilane, hexyltriethoxysilane, octyltrimethoxysilane, octyltriethoxysilane, decyltrimethoxysilane, decyltriethoxysilane, dodecyltrimethoxysilane, and dodecyltriethoxysilane. The silane compounds may be used individually or in combinations of two or more. The alkoxysilane and one-end silanol siloxane may be used together as the surface treatment agent. In this case, the surface treatment may include adsorption in addition to a covalent bond

[0018] The surface treatment agent may be previously mixed with the thermally conductive particles in a pretreatment (i.e., a pretreatment method) or may be added when the matrix resin is mixed with the thermally conductive particles (ie., an integral blend method). In these methods, the surface treatment agent is preferably added in an amount of 0.01 to 10 parts by mass with respect to 100 parts by mass of the thermally conductive particles. The surface treatment allows the thermally conductive particles to be easily blended with the matrix resin.

[0019] The thermally conductive silicone gel composition of the present invention may contain components other than the above as needed For example, an addition reaction controlling agent such as 1-ethynyl-1-cyclohexanol, a heat resistance improver, a flame retardant, and a flame retardant auxiliary such as colcothar, titanium oxide, and cerium oxide may be added Moreover, an organic or inorganic particle pigment may be added for the purpose of coloring and toning. Alkoxy group-containing silicone may be added as a material, e.g., for the surface treatment of a filler.

[0020] The thermally conductive silicone gel composition of the present invention may contain thermally conductive particles other than the component (D) that are not classified as hazardous under GHS criteria, as a component (F). For example, hexagonal boron nitride, aluminum nitride, amorphous silica, and calcium carbonate are preferably used Alumina particles, crystalline silica particles, and magnesium oxide particles are classified as hazardous under GHS criteria and not preferred.

[0021] The thermal conductivity of the thermally conductive silicone gel composition is preferably 2.0 W/m-K or less, more preferably 0.5 to 2.0 W/m·K, and fiirther preferably 0.7 to 1.8 W/m-K Thus, it is possible to provide a silicone gel composition composed of substances not classified as hazardous under GHS criteria and thus being highly safe while having flexibility, a low dielectric constant, and well-balanced thermal conductivity.

[0022] The specific gravity of the thermally conductive silicone gel composition is preferably 2.2 or less, and more preferably 2.0 or less. This makes the thermally conductive silicone gel composition a lightweight thermal interface material (TIM). The lower limit of the specific gravity of the composition is preferably 1.2 or more, and more preferably 1.4 or more.

[0023] The thermally conductive silicone gel composition of the present invention is preferably in the form of a sheet, i.e., a thermally conductive silicone sheet. The sheet is suitable as a thermal interface material.

[0024] The method for producing the thermally conductive silicone sheet of the present invention includes: mixing the components (A), (B), (C), (D), and other components as needed sheeting the mixture; and curing the sheet, wherein the thermally conductive silicone sheet has a relative dielectric constant of 5.0 or less at a frequency of 1 MHz according to the mutual induction bridge method of JIS K 6911: 2006 and has a Shore OO hardness of 5 to 60.

[0025] The sheet is preferably produced by mixing the above material components, sandwiching the mixture between polyethylene terephthalate (PET) films, rolling the sandwiched mixture into a sheet, and curing the sheet at 80 to 120°C for 5 to 40 minutes.

Examples

[0026] Hereinafter, the present invention will be described by way of examples. However, the present invention is not limited to the following examples. Various parameters were measured in the following manner.

<Thermal Conductivity>

[0027] The thermal conductivity of a thermally conductive grease was measured by a hot disk (in accordance with ISO/CD 22007-2). As shown in FIG. 1A, in a thermal conductivity measuring apparatus 1, a polyimide film sensor 2 was sandwiched between two samples 3a, 3b, and constant power was applied to the sensor 2 to generate a certain amount of heat. Then, the thermal characteristics were analyzed from the value of a temperature rise of the sensor 2. The sensor 2 has a tip 4 with a diameter of 7 mm. As shown in FIG. 1B, the tip 4 has a double spiral structure of electrodes. An electrode 5 for an applied current and an electrode 6 for a resistance value (temperature measurement electrode) are located on the lower portion of the sensor 2. The thermal conductivity was calculated by the following formula (1).

[Formula 1]

$$\lambda = \frac{Po \cdot D(\tau)}{\pi^{3/2} \cdot r} \cdot \frac{D(\tau)}{\Delta T(\tau)}$$

λ:	Thermal conductivity (W/m-K)
P_0:	Constant power (W)
r:	Radius of sensor (m)
τ:	$\sqrt{\alpha \cdot t / r^2}$
α:	Thermal diffusivity of sample (m^2/s)
t:	Measuring time (s)
$D(\tau)$:	Dimensionless function of τ
$\Delta T(\tau)$:	Temperature rise of sensor (K)

<Relative Dielectric Constants

[0028] The relative dielectric constant at a frequency of 1 MHz was measured according to the mutual induction bridge method of JIS K 6911: 2006 using a 2 -mm-thick sheet.

<Shore OO Hardness>

[0029] The Shore OO hardness was measured by an automatic durometer stand according to ASTM D2240 using a stack of four 3-mm-thick sheets.

(Example 1)

1. Material Components

[0030] As the components (A) to (C), a two-part room temperature curing silicone polymer was used The two-part room temperature curing silicone polymer was composed of a solution A containing the components (A) and (C) and a solution B containing the components (A) and (B).

Two-part room temperature curing silicone polymer: 100 parts by mass in total

Viscosity of solution A: 3000 mPa·s
Viscosity of solution B: 1000 mPa·s
Component (D):

(D-1) Aluminum hydroxide particles having a median diameter (D50) of 55 pm (Higilite H-10 manufactured by SHOWADENKO KK, not surface treated): 240 parts by weight
(D-2a) Aluminum hydroxide particles having a median diameter (D50) of 4 μm (CL-303 manufactured by Sumitomo Chemical Co., Ltd, surface treated with an epoxy silane coupling agent): 100 parts by weight

2. Mixing and Sheet Formation

[0031] Thermally conductive silicone sheets were produced by mixing the above material components, sandwiching the mixture between PET films, rolling the sandwiched mixture into a sheet, and curing the sheet at 100°C for 10 minutes. The thermally conductive silicone sheets were either 2 mm or 3 mm thick.

[0032] The thermally conductive silicone sheets thus obtained were evaluated Table 1 summarizes the conditions and results.

(Example 2)

[0033] Thermally conductive silicone sheets of Example 2 were producedin the same manner as in Example 1 except for the following.

(D-1): 150 parts by mass
(D-2b) Aluminum hydroxide particles having a median diameter (D50) of 10 μm (Higilite H-32 manufactured by SHOWADENKO KK, not surface treated): 100 parts by mass
(F) Amorphous silica having a median diameter (D50) of 7 μm: 100 parts by mass

(Example 3)

[0034] Thermally conductive silicone sheets of Example 3 were producedin the same manner as in Example 1 except for the following.

(D-1): 92 parts by mass
(D-2a): 118 parts by mass

(Example 4)

[0035] Thermally conductive silicone sheets of Example 4 were produced in the same manner as in Example 1 except for the following.

[0036] As the components (A) to (C), a two-part room temperature curing silicone polymer was used

Two-part room temperature curing silicone polymer: 100 parts by mass in total
Viscosity of solution A: 300 mPa·s
Viscosity of solution B: 300 mPa·s
(D-1): 250 parts by mass
(D-2a): 130 parts by mass
(D-2c) Aluminum hydroxide particles having a median diameter (D50) of 18 μm (trade name "Higilite H-31T" manufactured by SHOWADENKO KK, surface treated with a titanate coupling agent): 70 parts by mass
(E) Decyltrimethoxysilane: 1 part by mass

[0037] Table 1 summarizes the above conditions and results.

[Table 1]

	Example 1	Example 2	Example 3	Example 4
(A)+(B)+(C)mass(g)	100	100	100	100
(D-1)mass(g)	240	150	92	250

(continued)

		Example 1	Example 2	Example 3	Example 4
(D-2) total mass (g)		100	100	118	200
(D-2a)		100	-	118	130
(D-2b)		-	100	-	-
(D-2c)		-	-	-	70
(F) Amorphous silica, mass (g)		-	100	-	-
Relative dielectric constant	50 Hz	5.0	4.6	4.3	5.4
	1 kHz	4.4	4.1	3.8	4.8
	1 MHz	4.2	3.9	3.7	4.6
Hardness (Shore OO)		35	51	20	10
Thermal conductivity (W/m·K), hot disk method		1.4	1.4	0.9	1.8
Specific gravity		1.81	1.87	1.60	1.88
GHS labelling		No	No	No	No

(Comparative Examples 1 to 5)

[0038] Thermally conductive silicone sheets of Comparative Examples 1 to 5 were produced in the same manner as in Example 1 except for those indicated in Table 2.

[Table 2]

		Comparative Example 1	Comparative Example 2	Comparative Example 3	Comparative Example 4	Comparative Example 5
(A)+(B)+(C)mass(g)		100	100	100	100	100
(D-1) mass (g)		350	0	0	240	0
(D-2) total mass (g)		0	350	20	0	0
(D-2a)		-	350	-	-	-
(D-2b)		-	-	20	-	-
(D-2c)		-	-	-	-	-
Alumina particles, (D50) 70 μm, mass (g)		0	0	-	96	-
Alumina particles, (D50) 35 μm, mass (g)		-	-	150	-	400
Alumina particles, (D50) 3 μm, mass (g)		-	-	300	432	300
Relative dielectric constant	50 Hz	-	-	5.7	6.1	6.0
	1 kHz	-	-	5.6	5.7	5.9
	1 Mhz	-	-	5.6	5.5	5.8
Hardness (Shore OO)		-	-	50	48	41

(continued)

			Comparative Example 1	Comparative Example 2	Comparative Example 3	Comparative Example 4	Comparative Example 5
Thermal conductivity (W/m·K), hot disk method	-	-	1.5	2.5	2.0		
Specific gravity	-	-	2.56	2.58	2.90		
GHS labelling	-	-	Yes	Yes	Yes		
(Note: In Comparative Examples 1 and 2, the particles (D-1) and (D-2) could not be blended, and sheets could not be formed)							

[0039] The above results indicate that the silicone gel compositions of Examples 1 to 4 composed of substances not classified as hazardous under GHS criteria had high safety, flexibility, a low dielectric constant, and well-balanced thermal conductivity.

Industrial Applicability

[0040] The thermally conductive silicone gel composition of the present invention is suitable to be interposed between a heat generating member and a heat dissipating material of electrical and electronic components or the like.

Description of Reference Numerals

[0041]

1 Thermal conductivity measuring apparatus
2 Sensor
3a, 3b Sample
4 Tip of the sensor
5 Electrode for applied current
6 Electrode for resistance value (temperature measurement electrode)

Claims

1. A thermally conductive silicone gel composition, comprising:

an organopolysiloxane, A, having two or more alkenyl groups per molecule;
an organohydrogenpolysiloxane, B, having two or more hydrogen atoms bonded directly to silicon atoms;
a platinum group metal-containing curing catalyst, C, in a catalytic amount; and
aluminum hydroxide particles, D, as thermally conductive particles in an amount of 200 to 600 parts by mass with respect to 100 parts by mass of a total amount of the components A to C, the aluminum hydroxide particles comprising, based on 100% by mass of the aluminum hydroxide particles,
aluminum hydroxide particles, D-1, having a median diameter, D50, of 30 μm or more in an amount of more than 45% by mass and less than 75% by mass;
and aluminum hydroxide particles, D-2, having a median diameter, D50, of less than 30 μm in an amount of 25% by mass or more and 55% by mass or less, the median diameter, D50, being a particle diameter at 50% in a volume based cumulative particle size distribution, which is determined by a particle size distribution measurement according to a laser diffraction scattering method, wherein the thermally conductive silicone gel composition is free from alumina particles, crystalline silica particles, and magnesium oxide particles, and the thermally conductive silicone gel composition has a relative dielectric constant of 5.0 or less at a frequency of 1 MHz according to a mutual induction bridge method of JIS K 6911: 2006 and has a Shore OO hardness of 5 to 60 according to ASTM D2240 measured by an automatic durometer stand using a stack of four 3 mm thick sheets of the thermally conductive silicone gel composition.

2. The thermally conductive silicone gel composition according to claim 1, wherein the organopolysiloxane as the component A has a viscosity of 100 to 10000 Pa·s measured at 23°C with a B type rotational viscometer at a rotational speed of 20 rpm 30 seconds after start of rotation of a spindle of the viscometer.

3. The thermally conductive silicone gel composition according to claim 1 or 2, wherein in the organohydrogenpolysiloxane as the component B, the number of moles of the hydrogen atoms bonded directly to silicon atoms is 0.1 to 5.0 times the number of moles of the alkenyl groups of the component A.

4. The thermally conductive silicone gel composition according to any one of claims 1 to 3, further comprising a surface treatment agent for thermally conductive particles, as a component E.

5. The thermally conductive silicone gel composition according to claim 4, wherein the surface treatment agent comprises at least one coupling agent selected from the group consisting of a titanate coupling agent, an aluminate coupling agent, a stearate coupling agent, an epoxy silane coupling agent, and an alkyl silane coupling agent.

6. The thermally conductive silicone gel composition according to claim 5, wherein the alkyl silane coupling agent comprises at least one selected from the group consisting of an alkoxysilane compound expressed by $R_aSi(OR')_{4-a}$, where R represents a substituted or unsubstituted organic group having 6 to 12 carbon atoms, R' represents an alkyl group having 1 to 4 carbon atoms, and a is 0 or 1, a partial hydrolysate of the alkoxysilane compound, and an alkoxy group-containing silicone having a substituted or unsubstituted organic group having 6 to 12 carbon atoms.

7. The thermally conductive silicone gel composition according to any one of claims 4 to 6, wherein the thermally conductive silicone gel composition comprises the surface treatment agent in an amount of 0.01 to 10 parts by mass with respect to 100 parts by mass of the thermally conductive particles.

8. The thermally conductive silicone gel composition according to any one of claims 1 to 7, further comprising at least one thermally conductive particles selected from the group consisting of hexagonal boron nitride, aluminum nitride, amorphous silica, and calcium carbonate, as a component F.

9. The thermally conductive silicone gel composition according to any one of claims 1 to 8, wherein the thermally conductive silicone gel composition has a thermal conductivity of 2.0 W/m·K or less measured according to ISO/CD 22007-2 by a hot disk in accordance with the description.

10. The thermally conductive silicone gel composition according to any one of claims 1 to 9, wherein the thermally conductive silicone gel composition has a specific gravity of 2.2 or less.

11. A thermally conductive silicone sheet that is a sheet of the thermally conductive silicone gel composition according to any one of claims 1 to 10.

12. A method for producing the thermally conductive silicone sheet according to claim 11, comprising: mixing the following components of A to D; sheeting the mixture; and curing the sheet, the components of the mixture comprising:

an organopolysiloxane, A, having two or more alkenyl groups per molecule;
an organohydrogenpolysiloxane, B, having two or more hydrogen atoms bonded directly to silicon atoms;
a platinum group metal-containing curing catalyst, C, in a catalytic amount; and
aluminum hydroxide particles, D, as thermally conductive particles in an amount of 200 to 600 parts by mass with respect to 100 parts by mass of a total amount of the components A to C, the aluminum hydroxide particles comprising, based on 100% by mass of the aluminum hydroxide particles, aluminum hydroxide particles, D-1, having a median diameter, D50, of 30 μm or more in an amount of more than 45% by mass and less than 75% by mass;
and aluminum hydroxide particles, D-2, having a median diameter, D50, of less than 30 μm in an amount of 25% by mass or more and 55% by mass or less, the median diameter, D50, being a particle diameter at 50% in a volume based cumulative particle size distribution, which is determined by a particle size distribution measurement according to a laser diffraction scattering method, wherein the thermally conductive silicone gel composition is free from alumina particles, crystalline silica particles, and magnesium oxide particles, and the ther-

mally conductive silicone sheet has a relative dielectric constant of 5.0 or less at a frequency of 1 MHz according to a mutual induction bridge method of JIS K 6911: 2006 and has a Shore OO hardness of 5 to 60 according to ASTM D2240 measured by an automatic durometer stand using a stack of four 3 mm thick sheets of the thermally conductive silicone gel composition.

Patentansprüche

1. Wärmeleitfähige Silicongel-Zusammensetzung, beinhaltend:

   ein Organopolysiloxan, A, mit zwei oder mehr Alkenylgruppen pro Molekül;
   ein Organohydrogenpolysiloxan, B, mit zwei oder mehr direkt an Siliciumatome gebundenen Wasserstoffatomen;
   einen Platingruppenmetall-enthaltenden Härtungskatalysator, C, in einer katalytischen Menge; und
   Aluminiumhydroxid-Partikel, D, als wärmeleitfähige Partikel in einer Menge von 200 bis 600 Massenteilen bezogen auf 100 Massenteile der Komponenten A bis C, wobei die Aluminiumhydroxid-Partikel, bezogen auf 100% Massenteile der Aluminiumhydroxid-Partikel, Aluminiumhydroxid-Partikel, D-1, mit einem Mediandurchmesser, D50, von 30 μm oder mehr in einer Menge von mehr als 45 Masse% und weniger als 75 Masse%; und Aluminiumhydroxid-Partikel, D-2, mit einem Mediandurchmesser, D50, von weniger als 30 μm in einer Menge von 25 Masse% oder mehr und 55 Masse% oder weniger beinhalten, wobei der Mediandurchmesser, D50, ein Partikeldurchmesser bei 50% in einer Volumenbasierten kumulativen Partikelgrößenverteilung ist, die durch eine Partikelgrößenverteilungs-Messung gemäß einem Laser-Beugungs-Streuverfahren bestimmt wird, wobei die wärmeleitfähige Silicongel-Zusammensetzung frei von Alumina-Partikeln, kristallinen Silica-Partikeln und Magnesiumoxid-Partikeln ist, und die wärmeleitfähige Silicongel-Zusammensetzung eine relative Dielektrizitätskonstante von 5,0 oder weniger bei einer Frequenz von 1 MHz gemäß einem Mutual-Induction-Bridge-Verfahren von JIS K 6911: 2006 und eine Shore-OO-Härte von 5 bis 60 gemäß ASTM D2240 aufweist, gemessen durch ein automatisches Durometer-Gestell mittels eines Stapels von vier 3 mm dicken Bögen der wärmeleitfähigen Silicongel-Zusammensetzung.

2. Wärmeleitfähige Silicongel-Zusammensetzung gemäß Anspruch 1, wobei das Organopolysiloxan als die Komponente A eine Viskosität von 100 bis 10000 Pa·s aufweist, gemessen bei 23°C mit einem B-Typ-Rotations-Viscometer bei einer Rotationsgeschwindigkeit von 20 UpM 30 Sekunden nach dem Start der Rotation einer Spindel des Viscometers.

3. Wärmeleitfähige Silicongel-Zusammensetzung gemäß Anspruch 1 oder 2, wobei bei dem Organohydrogenpolysiloxan als die Komponente B, die Anzahl an Molen der direkt an Siliciumatome gebundenen Wasserstoffatome das 0,1- bis 5,0-fache der Anzahl der Alkenylgruppen Mole der Alkenylgruppen der Komponente A beträgt.

4. Wärmeleitfähige Silicongel-Zusammensetzung gemäß einem der Ansprüche 1 bis 3, ferner aufweisend ein Oberflächenbehandlungs Agens für wärmeleitfähige Partikel als eine Komponente E.

5. Wärmeleitfähige Silicongel-Zusammensetzung gemäß Anspruch 4, wobei das Oberflächenbehandlungs-Agens wenigstens ein Kopplungs-Agens aufweist ausgewählt aus der Gruppe, die aus einem Titanat-Kopplungs-Agens, einem Aluminat-Kopplungs-Agens, einem Stearat-Kopplungs-Agens, einem Epoxysilan-Kopplungs-Agens und einem Alkylsilan-Kopplungs-Agens besteht.

6. Wärmeleitfähige Silicongel-Zusammensetzung gemäß Anspruch 5, wobei das Alkylsilan-Kopplungs-Agens wenigstens eines aufweist ausgewählt aus der Gruppe, die aus einer Alkoxysilanverbindung, ausgedrückt durch R_a-Si(OR')$_{4-a}$, wobei R eine substituierte oder unsubstituierte organische Gruppe mit 6 bis 12 Kohlenstoffatomen darstellt, R' eine Alkylgruppe mit 1 bis 4 Kohlenstoffatomen darstellt und a 0 oder 1 ist, einem partiellen Hydrolysat der Alkoxysilanverbindung und einem Alkoxygruppen-enthaltenden Silicon mit einer substituierten oder unsubstituierten organischen Gruppe mit 6 bis 12 Kohlenstoffatomen besteht.

7. Wärmeleitfähige Silicongel-Zusammensetzung gemäß einem der Ansprüche 4 bis 6, wobei die wärmeleitfähige Silicongel-Zusammensetzung das Oberflächenbehandlungs-Agens in einer Menge von 0,01 bis 10 Massenteilen bezogen auf 100 Massenteile der wärmeleitfähigen Partikel enthält.

8. Wärmeleitfähige Silicongel-Zusammensetzung gemäß einem der Ansprüche 1 bis 7, ferner beinhaltend wenigstens

eine wärmeleitfähige Partikel ausgewählt aus der Gruppe, die aus hexagonalem Bornitrid, Aluminiumnitrid, amorphem Silica und Calciumcarbonat besteht, als eine Komponente F.

9. Wärmeleitfähige Silicongel-Zusammensetzung gemäß einem der Ansprüche 1 bis 8, wobei die wärmeleitfähige Silicongel-Zusammensetzung eine Wärmeleitfähigkeit von 2,0 W/m·K oder weniger, gemessen gemäß ISO/CD 22007-2 mittels einer heißen Scheibe gemäß der Beschreibung, aufweist.

10. Wärmeleitfähige Silicongel-Zusammensetzung gemäß einem der Ansprüche 1 bis 9, wobei die wärmeleitfähige Silicongel-Zusammensetzung ein spezifisches Gewicht von 2,2 oder weniger aufweist.

11. Wärmeleitfähiger Silicongel-Bogen, der ein Bogen aus der wärmeleitfähigen Silicongel-Zusammensetzung gemäß einem der Ansprüche 1 bis 10 ist.

12. Verfahren zum Herstellen des wärmeleitfähigen Silicongel-Bogens gemäß Anspruch 11, beinhaltend: Mischen der folgenden Komponenten von A bis D; Ausbreiten der Mischung; und Aushärten des Bogens, wobei die Komponenten der Mischung beinhalten:

ein Organopolysiloxan, A, mit zwei oder mehr Alkenylgruppen pro Molekül;
ein Organohydrogenpolysiloxan, B, mit zwei oder mehr direkt an Siliciumatome gebundenen Wasserstoffatomen;
einen Platingruppenmetall-enthaltenden Härtungskatalysator, C, in einer katalytischen Menge; und
Aluminiumhydroxid-Partikel, D, als wärmeleitfähige Partikel in einer Menge von 200 bis 600 Massenteilen bezogen auf 100 Massenteile der Komponenten A bis C, wobei die Aluminiumhydroxid-Partikel, bezogen auf 100% Massenteile der Aluminiumhydroxid-Partikel, Aluminiumhydroxid-Partikel, D-1, mit einem Mediandurchmesser, D50, von 30 μm oder mehr in einer Menge von mehr als 45 Masse% und weniger als 75 Masse%; und Aluminiumhydroxid-Partikel, D-2, mit einem Mediandurchmesser, D50, von weniger als 30 μm in einer Menge von 25 Masse% oder mehr und 55 Masse% oder weniger beinhalten, wobei der Mediandurchmesser, D50, ein Partikeldurchmesser bei 50% in einer Volumenbasierten kumulativen Partikelgrößenverteilung ist, die durch eine Partikelgrößenverteilungs-Messung gemäß einem Laser-Beugungs-Streuverfahren bestimmt wird, wobei die wärmeleitfähige Silicongel-Zusammensetzung frei von Alumina-Partikeln, kristallinen Silica-Partikeln und Magnesiumoxid-Partikeln ist, und die wärmeleitfähige Silicongel-Zusammensetzung eine relative Dielektrizitätskonstante von 5,0 oder weniger bei einer Frequenz von 1 MHz gemäß einem Mutual-Induction-Bridge-Verfahren von JIS K 6911: 2006 und eine Shore-OO-Härte von 5 bis 60 gemäß ASTM D2240 aufweist, gemessen durch ein automatisches Durometer-Gestell mittels eines Stapels von vier 3 mm dicken Bögen der wärmeleitfähigen Silicongel-Zusammensetzung.

Revendications

1. Composition de gel de silicone thermoconductrice, comprenant:

un organopolysiloxane, A, ayant deux groupes alcényle ou plus par molécule;
un organohydrogénopolysiloxane, B, ayant deux atomes d'hydrogène ou plus liés directement à des atomes de silicium;
un catalyseur de durcissement contenant un métal du groupe du platine, C, en une quantité catalytique; et
des particules d'hydroxyde d'aluminium, D, en tant que particules thermoconductrices en une quantité de 200 à 600 parties en masse par rapport à 100 parties en masse d'une quantité totale des composants A à C, les particules d'hydroxyde d'aluminium comprenant, sur la base de 100% en masse des particules d'hydroxyde d'aluminium, des particules d'hydroxyde d'aluminium, D-1, ayant un diamètre médian, D50, de 30μm ou plus en une quantité supérieure à 45% en masse et inférieure à 75% en masse; et des particules d'hydroxyde d'aluminium, D-2, ayant un diamètre médian, D50, inférieur à 30μm en une quantité de 25% en masse ou plus et 55% en masse ou moins, le diamètre médian, D50, étant un diamètre de particule à 50% dans une distribution granulométrique cumulative basée sur un volume, qui est déterminé par une mesure de la distribution granulométrique selon un procédé de diffusion par diffraction laser, dans lequel la composition de gel de silicone thermoconductrice est exempte de particules d'alumine, de particules de silice cristalline et de particules d'oxyde de magnésium, et la composition de gel de silicone thermoconductrice a une constante diélectrique relative de 5,0 ou moins à une fréquence de 1 MHz selon un procédé de pont à induction mutuelle de JIS K 6911:2006 et a une dureté Shore OO de 5 à 60 selon la norme ASTM D2240 mesurée par un banc de duromètre automatique

à l'aide d'un empilement de quatre feuilles d'une épaisseur de 3mm de la composition de gel de silicone thermoconductrice.

2. Composition de gel de silicone thermoconductrice selon la revendication 1, dans laquelle l'organopolysiloxane en tant que composant A a une viscosité de 100 à 10 000 Pa.s mesurée à 23°C avec un viscosimètre rotatif de type B à une vitesse de rotation de 20 tr/min 30 secondes après le début de rotation d'une broche du viscosimètre.

3. Composition de gel de silicone thermoconductrice selon la revendication 1 ou 2, dans laquelle dans l'organohydrogénopolysiloxane en tant que composant B, le nombre de moles des atomes d'hydrogène liés directement aux atomes de silicium est de 0,1 à 5,0 fois le nombre de moles des groupes alcényle du composant A.

4. Composition de gel de silicone thermoconductrice selon l'une quelconque des revendications 1 à 3, comprenant en outre un agent de traitement de surface pour des particules thermiquement conductrices, en tant que composant E.

5. Composition de gel de silicone thermoconductrice selon la revendication 4, dans laquelle l'agent de traitement de surface comprend au moins un agent de couplage choisi dans le groupe constitué d'un agent de couplage titanate, d'un agent de couplage aluminate, d'un agent de couplage stéarate, d'un agent de couplage époxy silane et d'un agent de couplage alkylsilane.

6. Composition de gel de silicone thermoconductrice selon la revendication 5, dans laquelle l'agent de couplage alkylsilane comprend au moins un composé choisi dans le groupe constitué d'un composé alcoxysilane exprimé par RaSi(OR')4-a, où R représente un groupe organique substitué ou non substitué ayant 6 à 12 atomes de carbone, R' représente un groupe alkyle ayant 1 à 4 atomes de carbone, et a vaut 0 ou 1, un hydrolysat partiel du composé alcoxysilane, et une silicone contenant un groupe alcoxy ayant un groupe organique substitué ou non substitué ayant 6 à 12 atomes de carbone.

7. Composition de gel de silicone thermoconductrice selon l'une quelconque des revendications 4 à 6, dans laquelle la composition de gel de silicone thermoconductrice comprend l'agent de traitement de surface en une quantité de 0,01 à 10 parties en masse par rapport à 100 parties en masse des particules thermoconductrices.

8. Composition de gel de silicone thermoconductrice selon l'une quelconque des revendications 1 à 7, comprenant en outre au moins une particule thermoconductrice choisie dans le groupe constitué par le nitrure de bore hexagonal, le nitrure d'aluminium, la silice amorphe et le carbonate de calcium, en tant que composant F.

9. Composition de gel de silicone thermoconductrice selon l'une quelconque des revendications 1 à 8, dans laquelle la composition de gel de silicone thermoconductrice a une conductivité thermique de 2,0 W/m·K ou moins mesurée selon la norme ISO/CD 22007-2 par un disque chaud conformément à la description.

10. Composition de gel de silicone thermoconductrice selon l'une quelconque des revendications 1 à 9, dans laquelle la composition de gel de silicone thermoconductrice a une densité spécifique de 2,2 ou moins.

11. Feuille de silicone thermoconductrice qui est une feuille de la composition de gel de silicone thermoconductrice selon l'une quelconque des revendications 1 à 10.

12. Procédé de production de la feuille de silicone thermoconductrice selon la revendication 11, comprenant: le mélange des composants suivants de A à D; la mise en feuille du mélange; et le durcissement de la feuille, les composants du mélange comprenant: un organopolysiloxane, A, ayant deux groupes alcényle ou plus par molécule; un organohydrogénopolysiloxane, B, ayant deux atomes d'hydrogène ou plus liés directement à des atomes de silicium;

un catalyseur de durcissement contenant un métal du groupe du platine, C, en une quantité catalytique; et des particules d'hydroxyde d'aluminium, D, en tant que particules thermoconductrices en une quantité de 200 à 600 parties en masse par rapport à 100 parties en masse d'une quantité totale des composants A à C, les particules d'hydroxyde d'aluminium comprenant, sur la base de 100% en masse des particules d'hydroxyde d'aluminium, des particules d'hydroxyde d'aluminium, D-1, ayant un diamètre médian, D50, de 30μm ou plus en une quantité supérieure à 45% en masse et inférieure à 75% en masse; et des particules d'hydroxyde d'aluminium, D-2, ayant un diamètre médian, D50, inférieur à 3μm en une quantité de 25% en masse ou plus et 55% en masse ou moins, le diamètre médian, D50, étant un diamètre de particule à 50% dans une distribution granulométrique cumulative basée sur un volume, qui est déterminé par une mesure de la distribution granulométrique selon un procédé de diffusion par diffraction laser, dans lequel la composition de gel de silicone thermoconductrice est exempte de particules d'alumine, de particules de silice cristalline et de particules d'oxyde de magnésium, et la feuille de silicone thermoconductrice a une constante diélectrique relative de 5,0 ou moins à une fréquence de 1 MHz selon un procédé de pont à induction mutuelle de JIS K 6911:2006 et a une dureté Shore OO de 5 à 60 selon la norme ASTM D2240 mesurée par un banc de duromètre automatique à l'aide d'un empilement de quatre feuilles d'une épaisseur de 3mm de la composition de gel de silicone thermoconductrice.

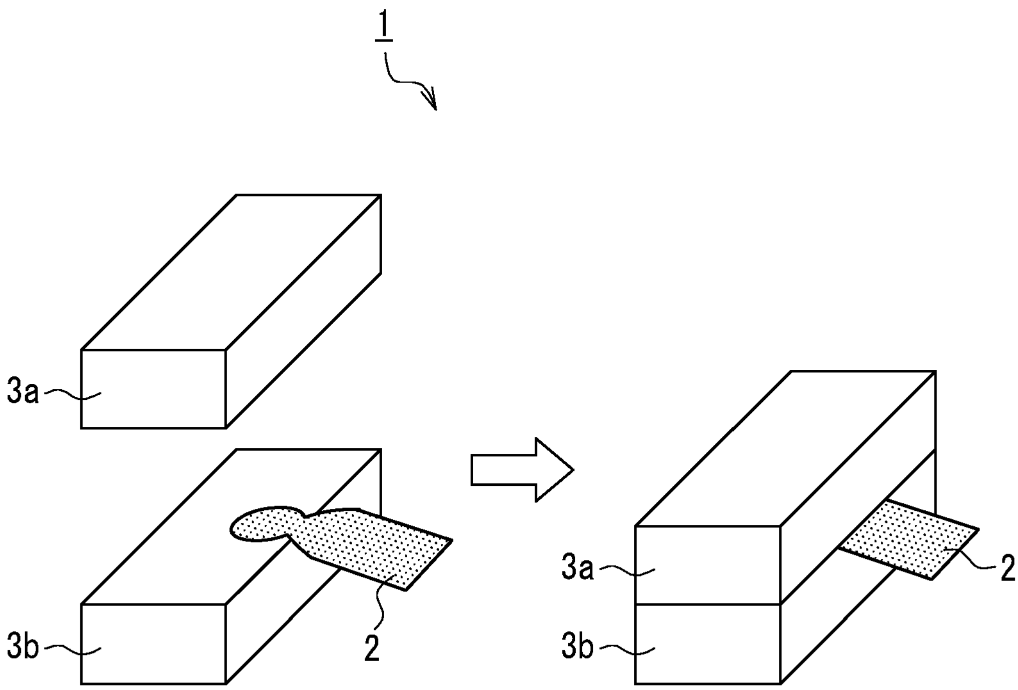

FIG. 1A

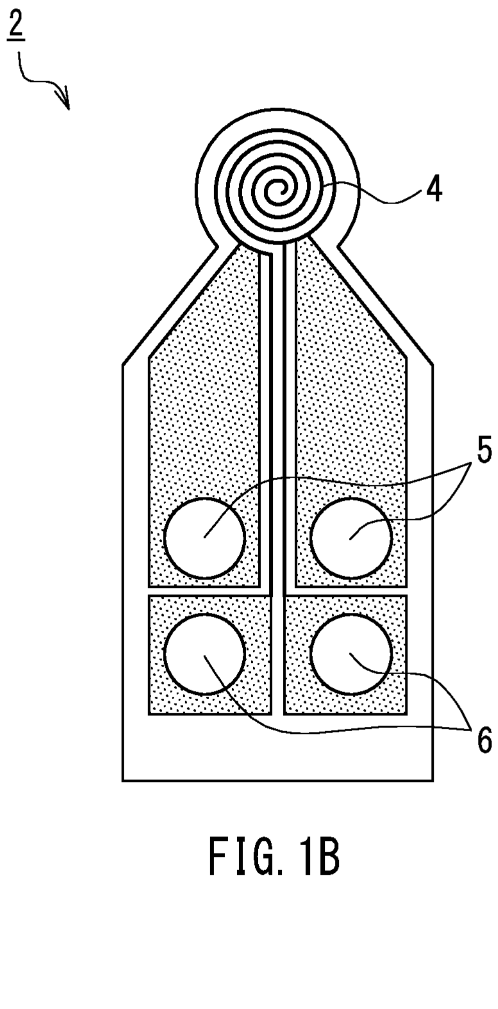

FIG. 1B

REFERENCES CITED IN THE DESCRIPTION

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