conductive particles that are not surface treated and used for the heat-resistant thermally conductive composition of the present invention. The surface treated thermally conductive particles are easily incorporated into the matrix resin, prevent the curing catalyst from being adsorbed on the thermally conductive particles, and thus have the effects of preventing cure inhibition. This is useful for storage stability.

[0013] The benzimidazolone compound used in the present invention is added preferably in an amount of 0.001 to 5 parts by mass, more preferably in an amount of 0.001 to 3 parts by mass, and further preferably in an amount of 0.001 parts by mass or more and 0.5 parts by mass or less with respect to 100 parts by mass of the heat-resistant thermally conductive composition. If the amount is less than 0.001 parts by mass, the benzimidazolone compound has few effects of improving heat resistance. If the amount is more than 5 parts by mass, the benzimidazolone compound has heat resistance effects, but is disadvantageous in terms of cost.

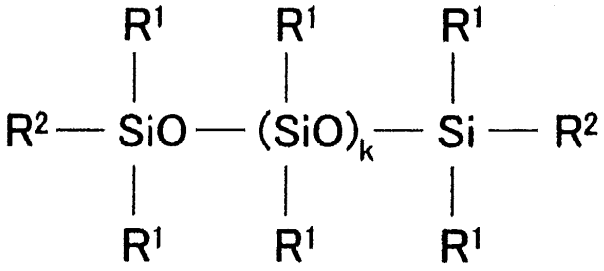
[0014] The benzimidazolone compound has a benzimidazolone moiety in its skeleton and includes a benzimidazolone pigment. Examples of the benzimidazolone pigment include: Pigment Yellow 120 (hereinafter, PY), PY 151, PY 154, PY 175, PY 180, PY 181, and PY 194; Pigment Orange 36 (hereinafter, PO), PO 60, PO 62, and PO 72; Pigment Red 171 (hereinafter, PR), PR 175, PR 176, PR 185, and PR 208; Pigment Violet 32 (hereinafter, PV 32); and Pigment Brown 25 (hereinafter, PBr 25). The pigments are indicated by color index (C.I.) names and numbers jointly established by the Society of Dyers and Colourists and the American Association of Textile Chemists and Colorists. The structure of Pigment Yellow 181 as an example of the benzimidazolone pigment is expressed by the following formula (Chemical Formula 1). The compound is expressed as 4'-carbamoyl-4-[1-(2,3-dihydro-2-oxo-1H-benzimidazol-5-ylcarbamoyl)acetonylazo]benzanilide. Although a mechanism in which the addition of the benzimidazolone compound improves heat resistance is not clear, it is considered that the compound absorbs or suppresses substances that may cause the thermal decomposition such as thermal radicals produced at high temperature.

[0015] [Chemical Formula 1]

containing two or more alkenyl groups is the base resin (base polymer component) of a silicone gel composition of the present invention. In the organopolysiloxane, two or more alkenyl groups having 2 to 8 carbon atoms, and preferably 2 to 6 carbon atoms such as vinyl groups or allyl groups are bonded to the silicon atoms per molecule. The viscosity of the organopolysiloxane is preferably 10 to 100000 mPa·s, and more preferably 100 to 10000 mPa·s at 25°C in terms of workability and curability.

[0023] Specifically, an organopolysiloxane expressed by the following general formula (Chemical Formula 2) is used. The organopolysiloxane has an average of two or more alkenyl groups per molecule, in which the alkenyl groups are bonded to silicon atoms at both ends of the molecular chain. The organopolysiloxane is a linear organopolysiloxane whose side chains are blocked with alkyl groups. The viscosity of the linear organopolysiloxane is preferably 10 to 100000 mPa·s at 25°C in terms of workability and curability. Moreover, the linear organopolysiloxane may include a small amount of branched structure (trifunctional siloxane units) in the molecular chain.

[0024] [Chemical Formula 2]



[0025] In the general formula, R^1 represents substituted or unsubstituted monovalent hydrocarbon groups that are the same as or different from each other and have no aliphatic unsaturated bond, R^2 represents alkenyl groups, and k represents 0 or a positive integer. The monovalent hydrocarbon groups represented by R^1 preferably have 1 to 10 carbon atoms, and more preferably 1 to 6 carbon atoms. Specific examples of the monovalent hydrocarbon groups include the following: alkyl groups such as methyl, ethyl, propyl, isopropyl, butyl, isobutyl, tert-butyl, pentyl, neopentyl, hexyl, cyclohexyl, octyl, nonyl, and decyl groups; aryl groups such as phenyl, tolyl, xylyl, and naphthyl groups; aralkyl groups such as benzyl, phenylethyl, and

sheet was measured in accordance with the Society of Rubber Industry, Japan Standard (SRIS0101).

<Heat resistance>

The heat-resistant thermally conductive silicone gel sheets were stacked to prepare a test block having a thickness of 10 mm or more. The test block was placed on a wire mesh. In this state, the test block was placed in a hot air circulating oven at 150°C or 220°C. The test block was taken out from the oven after a predetermined time and cooled to a room temperature. Then, the Asker C hardness of the test block was measured in accordance with the Society of Rubber Industry, Japan Standard (SRIS0101).

The heat resistance was evaluated with a rate of increase in the Asker C hardness after exposure at 150°C or 220°C.

The rate of increase was calculated by the following formula (Formula 2).

[Formula 2]

$$\text{Rate of increase (\%)} = [\ |B' - B|/|A' - A|\ ] \times 100$$

A: Initial hardness of sample without heat resistance improver

A′: Hardness of sample without heat resistance improver after exposure at 150°C or 220°C

B: Initial hardness of sample containing heat resistance improver

B′: Hardness of sample containing heat resistance improver after exposure at 150°C or 220°C for same period of time as that of A′

*The composition and the processing method of the sample of A were the same as those of the sample of B except for the presence or absence of the heat resistance improver.

[0043] (Examples 1 and 2, Comparative Example 1)

1. Material components

(1) Matrix resin

A two-part addition-curable silicone polymer that becomes a silicone gel after curing was used. One solution (solution A) contained a base polymer component (component A) and a platinum group metal catalyst (component C). The other solution (solution B) contained a base polymer component (component A) and an organohydrogenpolysiloxane that is a crosslinking agent component (component B).

[0061] [TABLE 9]

	220°C Exposure time (hours)	0	100	250	500	1000
Hardness [Asker C]	Ex. 5	32	31	31	41	58
	Ex. 6	33	32	31	30	36
	Ex. 7	33	30	29	37	55
	Ex. 8	33	30	26	26	28
	Ex. 9	33	30	27	25	27
	Comp. Ex. 5	13	13	20	30	48
	Comp. Ex. 6	33	22	55	77	86
	Comp. Ex. 7	25	19	92	94	95
Rate of increase (%) with respect to rate of increase of Comp. Ex. 7	Ex. 5	-	17	1	13	37
	Ex. 6	-	17	3	4	4
	Ex. 7	-	50	6	6	31
	Ex. 8	-	50	10	10	7
	Ex. 9	-	50	9	12	9
	Comp. Ex. 5	-	0	10	25	50
	Comp. Ex. 6	-	183	33	64	76

*-: Calculation impossible, Ex.: Example, Comp. Ex.: Comparative Example

[0062] (Examples 10-14, Comparative Example 7)

Examples 10-14 and Comparative Example 7 were performed in the same manner as Example 1 except for the amounts of the thermally conductive particles and the heat resistance improver that were added shown in Table 7. The heat resistances thereof were evaluated at a temperature of 220°C. Table 10 shows the conditions, and Table 11 shows the results.

[0063] [TABLE 10]

			Ex. 10	Ex. 11	Ex. 12	Ex. 13	Comp. Ex. 7
Matrix	Two-part addition-curable silicone polymer: Solution A		50	50	50	50	50
	Two-part addition-curable silicone polymer: Solution B		50	50	50	50	50
Thermally conductive particles	Alumina Average particle size: 35 μm BET specific surface area: 0.2 m^2/g		600	600	600	600	600
Heat resistance improver	Benzimidazolone compound	PR 176	0.05	0.10	0.50	5.0	-
Result	Thermal conductivity (W/m·K) [Hot disk]		1.7	1.7	1.7	1.7	1.7
	Hardness [Asker C] after curing		30	29	29	29	25

*-: Not added, Ex.: Example, Comp. Ex.: Comparative Example

[0064] [TABLE 11]

CLAIMS

1. A heat-resistant thermally conductive composition comprising:

a matrix resin; and

thermally conductive particles,

wherein the heat-resistant thermally conductive composition contains a benzimidazolone compound as a heat resistance improver, and

if the matrix resin is 100 parts by mass, the thermally conductive particles are added in an amount of 100 to 4000 parts by mass.

2. The heat-resistant thermally conductive composition according to claim 1, wherein the benzimidazolone compound is added in an amount of 0.001 to 5 parts by mass with respect to 100 parts by mass of the heat-resistant thermally conductive composition.

3. The heat-resistant thermally conductive composition according to claim 1 or 2, wherein the matrix resin is a silicone polymer.

4. The heat-resistant thermally conductive composition according to any of claims 1 to 3, wherein the benzimidazolone compound is a benzimidazolone pigment.

5. The heat-resistant thermally conductive composition according to any of claims 1 to 4, further comprising a curing catalyst,

wherein the matrix resin is an addition-curable silicone polymer.

6. The heat-resistant thermally conductive composition according to any of claims 1 to 5, wherein the heat-resistant thermally conductive composition has an Asker C hardness of 70 or less after curing.

7. The heat-resistant thermally conductive composition according to any of claims 1 to 6, wherein the thermally conductive particles are at least one selected from alumina, zinc oxide, magnesium oxide, aluminum nitride, boron nitride, aluminum hydroxide, and silica.

8. The heat-resistant thermally conductive composition according to any of claims 1 to 7, wherein part or all of the thermally conductive particles are surface treated with a silane coupling agent.

9. The heat-resistant thermally conductive composition according to claim 8, wherein the silane coupling agent is added in an amount of 0.01 to 10 parts by mass with respect to 100 parts by mass of the thermally conductive particles.

10. The heat-resistant thermally conductive composition according to any of claims 1 to 9, wherein the heat-resistant thermally conductive composition comprises the following components (A) to (E), and is cured:

(A) a base polymer component: an organopolysiloxane having an average of two or more silicon atoms bonded to alkenyl groups per molecule;

(B) a crosslinking component: an organopolysiloxane having an average of two or more silicon atoms bonded to hydrogen atoms per molecule, in which an amount of the organopolysiloxane is 0.01 to 3 mol with respect to 1 mol of the alkenyl groups bonded to the silicon atoms in the component A;

(C) a catalyst component: a platinum group metal catalyst, in which an amount of the platinum group metal catalyst is 0.01 to 1000 ppm in terms of a weight unit of metal atoms with respect to a total amount of the component A and the platinum group metal catalyst;

(D) thermally conductive particles: 100 to 4000 parts by mass with respect to 100 parts by mass of an addition-curable silicone polymer component (the component A + the component B); and

(E) a benzimidazolone compound: 0.001 to 5 parts by mass with respect to 100 parts by mass of the heat-resistant thermally conductive composition.

11. The heat-resistant thermally conductive composition according to any of claims 1 to 10, wherein the heat resistance improver contains no metal atom.

12. A heat-resistant thermally conductive sheet comprising the heat-resistant thermally conductive composition, according to any of claims 1 to 11, in the form of a sheet.

13. The heat-resistant thermally conductive sheet according to claim 12, wherein the heat-resistant thermally conductive sheet has a thermal conductivity of 0.8 W/m·K or more.

14. The heat-resistant thermally conductive sheet according to claim 12 or 13, wherein the heat-resistant thermally conductive sheet has a thickness of 0.2 to 10 mm.