

RESIN COMPOSITION

BACKGROUND OF THE INVENTION

1. Field of the Invention

The present invention relates to a novel resin composition. More particularly, the present invention relates to a resin composition suitable for use in an insulating layer of a multilayer printed circuit board.

2. Description of the Related Art

As a technique of manufacturing a multilayer printed circuit board, a manufacturing method by a build-up system is known. In the method, an insulating layer and a conductive layer are alternately stacked on a core substrate. In the manufacturing method by a build-up system, the insulating layer is generally formed by curing a resin composition. It is known that an epoxy resin composition is used as such a resin composition (Patent Literature 1).

Also, in recent years, when manufacturing a multilayer printed circuit board, inorganic fillers such as silica particles tend to be highly formulated in the resin composition to inhibit cracks and circuit distortion which are attributable to a difference in thermal expansion

between the insulating layer and the conductive layer
(Patent Literature 2).

Prior Art Literature

Patent Literature

Patent Literature 1: Japanese Unexamined Patent
Publication No. 2007-254709

Patent Literature 2: Japanese Unexamined Patent
Publication No. 2010-202865

Since further decrease in thickness of the multilayer printed circuit board is demanded, the thickness of the core substrate has a tendency to gradually decrease. However, the use of the thin core substrate causes insufficient stiffness of the core substrate. Accordingly, there can arise a problem that when the resin composition layer is laminated and cured on only one surface of the core substrate, the whole layered substrate is warped (hereinafter, also referred to as a "warpage problem").

Such a warpage problem is expected to be possibly mitigated by reducing the thermal expansion coefficient of the insulating layer while suppressing the elastic modulus at a low level. That is, the warpage problem is expected to be alleviated by suppressing the thermal expansion itself of the insulating layer and mitigating the stress

caused by the thermal expansion. Here, according to the recent tendency of using a resin composition having a high content of inorganic fillers, the thermal expansion of the obtained insulating layer is generally low, but the elastic modulus is extremely high. Therefore, the warpage problem is not alleviated at all, and it is desired to take measures. It is also considered that a flexible component can be added to the resin composition in order to reduce the elastic modulus of the insulating layer. However, in such a case, the thermal expansion of the insulating layer increases. Therefore, it is inferred that the warpage problem cannot be alleviated, either.

SUMMARY OF THE INVENTION

Embodiments of the present invention provide a resin composition that does not cause the warpage problem even when the thin core substrate is used.

The present inventors have found that the above-described problem can be solved by using a specific resin composition described below, and thus completed the present invention.

That is, embodiments of the present invention include the following contents.

According to a first aspect of the present invention, there is provided a resin composition including:

(A) an epoxy resin that is solid at 25°C;

(B) an inorganic filler; and

(C) at least one resin selected from the group consisting of a functional group-containing saturated butadiene resin that is liquid at 25°C, a functional group-containing unsaturated butadiene resin that is liquid at 25°C, and a functional group-containing acrylic resin having a glass transition temperature of 25°C or lower, wherein

when a content of a non-volatile component in the resin composition is assumed to be 100% by mass, a content of the (B) component is 40% by mass or more,

when a content of a resin component is assumed to be 100% by mass, a content of the (C) component is 10 to 40% by mass, and

a linear thermal expansion coefficient (α :ppm/°C) of a cured body obtained by curing the resin composition is 25 ppm or less.

In an embodiment of the above resin composition, the (C) component has at least one functional group selected from the group consisting of an acid anhydride group, a phenolic hydroxy group, an epoxy group, an isocyanate group, and a urethane group.

In an embodiment of the above resin composition, the (A) component is a multifunctional naphthalene skeleton epoxy resin.

In an embodiment, the above resin composition further comprises a curing agent.

In an embodiment of the above resin composition, the resin composition is a resin composition for embedding parts for the circuit board with built-in parts.

In an embodiment of the above resin composition, the inorganic filler has an average particle diameter of 10 μm or less, and is classified to remove a particle of 20 μm or more.

According to a second aspect of the present invention, there is provided a cured body obtained by curing the above resin composition.

In an embodiment, the above cured body has a glass transition temperature of 170°C or higher.

In an embodiment of the above cured body, a product $\alpha \times E$ of a linear thermal expansion coefficient (α :ppm/°C) and an elastic modulus (E:GPa) is 150 or less.

According to a third aspect of the present invention, there is provided a circuit board with built-in parts prepared using the above cured body.

According to embodiments of the present invention, there can be provided a resin composition that does not cause the warpage problem even when the thin core substrate is used.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

The present invention will be described in detail below in conformity with preferred embodiments thereof.

[Resin Composition]

The resin composition according to the present invention includes (A) an epoxy resin that is solid at 25°C, (B) an inorganic filler, and (C) at least one resin selected from the group consisting of a functional group-containing saturated butadiene resin that is liquid at 25°C, a functional group-containing unsaturated butadiene resin that is liquid at 25°C, and a functional group-containing acrylic resin having a glass transition temperature of 25°C or lower, wherein when the content of a non-volatile component in the resin composition is assumed to be 100% by mass, the content of the (B) component is 40% by mass or more, and when the content of a resin component is assumed to be 100% by mass, the content of the (C) component is 10 to 40% by mass.

The present inventors have found that the warpage problem can be solved by using the resin composition

containing the above-described specific components of (A) to (C) in the insulating layer of the multilayer printed circuit board in the above-described specific amount ratios.

Here, the index for the warpage of a composite material having two layers laminated to each other may include the thermal expansion coefficient (α_{com}) of the whole composite material calculated by the Turner's formula below. The lower the value of α_{com} is, the more the warpage of the composite material is suppressed. The Turner's formula is also described in, for example, Japanese Patent No. 3260340 and Japanese Unexamined Patent Publication No. 2008-186905.

[Mathematical Formula 1]

$$\alpha_{\text{com}} = (\alpha_1 E_1 t_1 + \alpha_2 E_2 t_2) / (E_1 t_1 + E_2 t_2)$$

α_1 : Thermal expansion coefficient of first layer [ppm]

α_2 : Thermal expansion coefficient of second layer [ppm]

E_1 : Elastic modulus of first layer [GPa]

E_2 : Elastic modulus of second layer [GPa]

t_1 : Film thickness of first layer [μm]

t_2 : Film thickness of second layer [μm]

For example, it is assumed that the first layer is an insulating layer, and the second layer is a core substrate. In this case, α_2 and E_2 are intrinsic values of the core

substrate used in the manufacture of the multilayer printed circuit board. Therefore, once the thicknesses of the insulating layer and the core substrate are determined, the above α_{com} can be substantially treated as a function of α_1 and E_1 . Also, since the elastic modulus of the core substrate is sufficiently higher than the elastic modulus of the insulating layer in general ($E_2 \gg E_1$), the contribution of the denominator is limited when the above α_{com} is considered as a function of α_1 and E_1 , and thus the α_{com} comes to exclusively depend on the product of α_1 and E_1 .

Although described in detail later, a cured product (the insulating layer) of the resin composition according to the present invention is low in both the thermal expansion coefficient (α_1) and the elastic modulus (E_1), and the product of α_1 and E_1 is extraordinarily low. According to the conventionally known technical common knowledge, the addition of a flexible component such as the (C) component causes the thermal expansion coefficient to increase, and therefore should inhibit desired effects from being achieved. Such knowledge found by the present inventors could not be anticipated from the conventionally known technical common knowledge.

The (A) to (C) components, which are contained in the resin composition according to the present invention, will be described below.

<(A) Component>

The (A) component is an epoxy resin that is solid at 25°C (hereinafter, also referred to as a "solid epoxy resin").

Examples of the solid epoxy resin may include, but not limited to, naphthalene-type epoxy resins, cresol novolac-type epoxy resins, dicyclopentadiene-type epoxy resins, trisphenol epoxy resins, naphthol novolac epoxy resins, biphenyl-type epoxy resins, and naphthylene ether-type epoxy resins. Among these, naphthalene-type epoxy resins, biphenyl-type epoxy resins or naphthylene ether-type epoxy resins are preferable, naphthalene-type epoxy resins are more preferable, and multifunctional naphthalene skeleton epoxy resins are further preferable. Specific examples of the solid epoxy resin may include "HP-4700," "HP-4710," and "EXA-7311G4S" (naphthalene-type epoxy resins), "N-690" (cresol novolac-type epoxy resin), "N-695" (cresol novolac-type epoxy resin), "HP-7200" (dicyclopentadiene-type epoxy resin), and "EXA7311," "EXA7311-G3," and "HP6000" (naphthylene ether-type epoxy resins) manufactured by DIC Corporation; "EPPN-502H" (trisphenol epoxy resin), "NC7000L" (naphthol novolac epoxy resin), and "NC3000H,"

"NC3000," "NC3000L," and "NC3100" (biphenyl-type epoxy resins) manufactured by Nippon Kayaku Co., Ltd.; "ESN475" (naphthol novolac-type epoxy resin) and "ESN485" (naphthol novolac-type epoxy resin) manufactured by Nippon Steel Chemical Co., Ltd.; and "YX4000H" and "YL6121" (biphenyl-type epoxy resins), and "YX4000HK" (bixylenol-type epoxy resin) manufactured by Mitsubishi Chemical Corporation. These substances may be used either singly or in any combination of two or more. Among these, a combination of two or more of the multifunctional naphthalene-type epoxy resins is preferably used.

The epoxy equivalent of the (A) component is preferably 50 to 3000, more preferably 80 to 2000, and further preferably 110 to 1000. Within this range, the crosslink density of the cured product becomes sufficient. Here, the epoxy equivalent can be measured in accordance with JIS K7236, and is the mass of a resin containing one equivalent of the epoxy group.

The number average molecular weight (M_n) of the (A) component, an epoxy resin that is solid at 25°C, is preferably 1000-5000, more preferably 250-3000, and further preferably 400-2000. Here, the number average molecular weight (M_n) of the resin is a polystyrene equivalent number average molecular weight measured by GPC (gel permeation chromatography).

The content of the (A) component in the resin composition is preferably 5% by mass to 50% by mass, more preferably 10% by mass to 40% by mass, and further preferably 15% by mass to 30% by mass.

<(B) Component>

The (B) component is an inorganic filler.

Examples of the inorganic filler may include silica, alumina, barium sulfate, talc, clay, mica, aluminum hydroxide, magnesium hydroxide, calcium carbonate, magnesium carbonate, magnesium oxide, boron nitride, aluminum borate, barium titanate, strontium titanate, calcium titanate, magnesium titanate, bismuth titanate, titanium oxide, barium zirconate, and calcium zirconate. Among these, silica such as amorphous silica, fused silica, crystalline silica, synthetic silica, hollow silica, and spherical silica is preferable, and spherical silica is particularly preferable. The inorganic fillers may be used either singly or in any combination of two or more. Preferred examples of the spherical silica may include "SO-C2" manufactured by Admatechs Company Limited and "FB-5SDC" manufactured by Denki Kagaku Kogyo Kabushiki Kaisha.

From the viewpoint of fluidity and circuit embedding properties, the average particle diameter of the inorganic filler is preferably 10 μm or less, more preferably 9 μm or less, further preferably 8 μm or less, further more

preferably 7 μm or less, and particularly preferably 6 μm or less or 5 μm or less. Although the lower limit of the average particle diameter of the inorganic filler is not particularly limited, it is preferably 0.05 μm or more, and more preferably 0.1 μm or more. Furthermore, the inorganic filler is subjected to classification to preferably remove a particle equal to or more than 20 μm , and to more preferably remove a particle equal to or more than 15 μm . Accordingly, when a circuit board with built-in parts is manufactured as a multilayer printed circuit board, the cavity filling properties and the parts embedding properties of the core substrate become favorable. In a preferred embodiment, the inorganic filler has an average particle diameter of 10 μm or less, and is subjected to classification to remove a particle of 20 μm or more. The average particle diameter of the inorganic filler can be measured by the laser diffraction and scattering method based on the Mie scattering theory. Specifically, the measurement is performed by preparing the volume-based particle size distribution of the inorganic filler using a laser diffraction and scattering-type particle size distribution measuring device to obtain the median diameter thereof as the average particle diameter. The inorganic filler dispersed in water with ultrasonic waves can be

preferably used as a measurement sample. As the laser diffraction and scattering-type particle size distribution measuring device, LA-500 manufactured by Horiba, Ltd., for example, may be used.

From the viewpoint of improving moisture resistance and dispersibility, the inorganic filler is preferably treated with at least one surface treatment agent such as an aminosilane-based coupling agent, an epoxysilane-based coupling agent, a mercaptosilane-based coupling agent, a silane-based coupling agent, an organosilazane compound, and a titanate-based coupling agent. Examples of the surface treatment agent as a commercially available product may include "KBM403" (3-glycidoxypropyltrimethoxysilane) manufactured by Shin-Etsu Chemical Co., Ltd., "KBM803" (3-mercaptopropyltrimethoxysilane) manufactured by Shin-Etsu Chemical Co., Ltd., "KBE903" (3-aminopropyltriethoxysilane) manufactured by Shin-Etsu Chemical Co., Ltd., "KBM573" (N-phenyl-3-aminopropyltrimethoxysilane) manufactured by Shin-Etsu Chemical Co., Ltd. and "SZ-31" (hexamethyldisilazane) manufactured by Shin-Etsu Chemical Co., Ltd.

From the viewpoint of improving dispersibility of the inorganic filler, amount of carbon per unit surface area of the inorganic filler is preferably 0.02 mg/m^2 or more, more preferably 0.1 mg/m^2 or more, and further preferably 0.2 mg/m^2 or more. On the other hand, from the viewpoint of

inhibiting the increase in melt viscosity of a thermosetting resin composition layer, amount of carbon per unit surface area of the inorganic filler is preferably 1 mg/m² or less, more preferably 0.8 mg/m² or less, and further preferably 0.5 mg/m² or less.

The inorganic filler subjected to surface treatment with the surface treatment agent is washed with a solvent (for example, methyl ethyl ketone (MEK)), and then amount of carbon per unit surface area of the inorganic filler can be measured. Specifically, MEK in an amount sufficient as a solvent is added to the inorganic filler subjected to surface treatment with the surface treatment agent, and the inorganic filler is washed with ultrasonic waves at 25°C for 5 minutes. The supernatant liquid is removed, and the solid content is dried. Thereafter, using a carbon analyser, amount of carbon per unit surface area of the inorganic filler can be measured. As the carbon analyser, "EMIA-320V" manufactured by Horiba, Ltd., for example, can be used.

From the viewpoint of sufficiently reducing the thermal expansion coefficient of the obtained cured product (the insulating layer), when the content of the non-volatile component in the resin composition is assumed to be 100% by mass, the content of the (B) component in the resin composition according to the present invention is 40%

by mass or more, preferably 45% by mass or more, and more preferably 50% by mass or more. In the present invention, while the warpage problem is suppressed, the content of the (B) component can be further increased. For example, the content of the (B) component in the resin composition may be increased to 55% by mass or more, 60% by mass or more, 65% by mass or more, 70% by mass or more, or 75% by mass or more.

From the viewpoint of the mechanical strength of the obtained cured product (the insulating layer), when the content of the non-volatile component in the resin composition is assumed to be 100% by mass, the upper limit of the content of the (B) component in the resin composition is preferably 95% by mass or less, more preferably 90% by mass or less, and further preferably 85% by mass or less.

<(C) Component>

The (C) component is at least one resin selected from the group consisting of a functional group-containing saturated and/or unsaturated butadiene resin that is liquid at 25°C and a functional group-containing acrylic resin having a glass transition temperature of 25°C or lower.

The glass transition temperature (T_g) of the (C) component is 25°C or lower. It is difficult to measure T_g

of the functional group-containing saturated and/or unsaturated butadiene resin. However, as understood from the fact of being liquid at 25°C, when the functional group-containing saturated and/or unsaturated butadiene resin is liquid at 25°C, it is naturally considered that Tg thereof is 25°C or lower. When such a resin having a low glass transition temperature is used in the insulating layer of the multilayer printed circuit board, the low elastic modulus of the (C) component tends to cause the elastic modulus of the insulating layer to decrease. On the other hand, it is known that such a resin having a low glass transition temperature usually increases the thermal expansion coefficient of the insulating layer. The present inventors have found that a specific resin having a Tg of 25°C or lower or being liquid at 25°C and having a specific functional group not only inhibits the thermal expansion coefficient of the insulating layer from increasing but also rather exerts an effect of reducing the thermal expansion coefficient of the insulating layer, while maintaining the effect of reducing the elastic modulus of the insulating layer. Thus, the present invention has been completed.

As the functional group contained in the resin of the (C) component, a functional group that can react with the

(A) component is preferable. In a preferred embodiment, the functional group contained in the resin of the (C) component is at least one functional group selected from the group consisting of an acid anhydride group, a phenolic hydroxy group, an epoxy group, an isocyanate group, and a urethane group. Among these, as the functional group, an epoxy group and a phenolic hydroxy group are preferable, and an epoxy group is more preferable.

From the viewpoint of obtaining the insulating layer having a low elastic modulus, the glass transition temperature (T_g) of the (C) component is 25°C or lower, preferably 20°C or lower, and more preferably 15°C or lower. The lower limit of the glass transition temperature of the (C) component can be, but not particularly limited to, usually -15°C or higher.

(Functional group-containing saturated and/or unsaturated butadiene resin that is liquid at 25°C)

The functional group-containing saturated and/or unsaturated butadiene resin that is liquid at 25°C is preferably at least one resin selected from the group consisting of an acid anhydride group-containing saturated and/or unsaturated butadiene resin that is liquid at 25°C, a phenolic hydroxy group-containing saturated and/or unsaturated butadiene resin that is liquid at 25°C, an

epoxy group-containing saturated and/or unsaturated butadiene resin that is liquid at 25°C, an isocyanate group-containing saturated and/or unsaturated butadiene resin that is liquid at 25°C, and an urethane group-containing saturated and/or unsaturated butadiene resin that is liquid at 25°C. As described herein, the "saturated and/or unsaturated butadiene resin" refers to a resin that contains a saturated butadiene skeleton and/or an unsaturated butadiene resin skeleton. In the resin, the saturated butadiene skeleton and/or the unsaturated butadiene skeleton may be contained either in the main chain or in the side chain.

The number average molecular weight (M_n) of the functional group-containing saturated and/or unsaturated butadiene resin that is liquid at 25°C is preferably 500 to 50000, more preferably 1000 to 10000, further preferably 2000 to 10000, and further more preferably 2300 to 10000. As described herein, the number average molecular weight (M_n) of the resin is a polystyrene equivalent number average molecular weight measured using GPC (gel permeation chromatography).

The functional group equivalent of the functional group-containing saturated and/or unsaturated butadiene resin that is liquid at 25°C is preferably 100 to 10000,

more preferably 150 to 5000, further preferably 190 to 3000, and further more preferably 200 to 2000. As described herein, the functional group equivalent is the mass of a resin containing one equivalent of the functional group. For example, the epoxy equivalent of a resin can be measured in accordance with Japanese Industrial Standards JIS K7236.

Especially, from the viewpoint of further reducing the thermal expansion coefficient and the elastic modulus of the insulating layer to further mitigate the warpage problem, the (C) component is preferably an epoxy group-containing saturated and/or unsaturated butadiene resin that is liquid at 25°C.

The epoxy group-containing saturated and/or unsaturated butadiene resin that is liquid at 25°C is preferably a saturated and/or unsaturated butadiene skeleton-containing epoxy resin that is liquid at 25°C; more preferably a polybutadiene skeleton-containing epoxy resin that is liquid at 25°C and a hydrogenated polybutadiene skeleton-containing epoxy resin that is liquid at 25°C. As described herein, the "hydrogenated polybutadiene skeleton-containing epoxy resin" refers to an epoxy resin including a polybutadiene skeleton that is at least partly hydrogenated, and is not necessarily an epoxy

resin including a polybutadiene skeleton that is completely hydrogenated. Specific examples of the polybutadiene skeleton-containing resin that is liquid at 25°C and the hydrogenated polybutadiene skeleton-containing resin that is liquid at 25°C may include PB3600 (a polybutadiene skeleton epoxy resin) manufactured by Daicel Chemical Industries, Ltd. and FCA-061L (a hydrogenated polybutadiene skeleton epoxy resin) manufactured by Nagase Chemtex Corporation.

The acid anhydride group-containing saturated and/or unsaturated butadiene resin that is liquid at 25°C is preferably a saturated and/or unsaturated butadiene skeleton-containing acid anhydride resin that is liquid at 25°C.

The phenolic hydroxy group-containing saturated and/or unsaturated butadiene resin that is liquid at 25°C is preferably a saturated and/or unsaturated butadiene skeleton-containing phenol resin that is liquid at 25°C.

The isocyanate group-containing saturated and/or unsaturated butadiene resin that is liquid at 25°C is preferably a saturated and/or unsaturated butadiene skeleton-containing isocyanate resin that is liquid at 25°C.

The urethane group-containing saturated and/or unsaturated butadiene resin that is liquid at 25°C is preferably a saturated and/or unsaturated butadiene skeleton-containing urethane resin that is liquid at 25°C. (Functional Group-Containing Acrylic Resin Having a Tg of 25°C or Lower)

The functional group-containing acrylic resin having a Tg of 25°C or lower is preferably at least one selected from the group consisting of an acid anhydride group-containing acrylic resin having a Tg of 25°C or lower, a phenolic hydroxy group-containing acrylic resin having a Tg of 25°C or lower, an isocyanate group-containing acrylic resin having a Tg of 25°C or lower, an urethane group-containing acrylic resin having a Tg of 25°C or lower, and an epoxy group-containing acrylic resin having a Tg of 25°C or lower.

The number average molecular weight (Mn) of the functional group-containing acrylic resin having a Tg of 25°C or lower is preferably 10000 to 1000000, more preferably 30000 to 900000, and further preferably 300000 to 900000.

The functional group equivalent of the functional group-containing acrylic resin having a Tg of 25°C or lower

is preferably 1000 to 50000, and more preferably 2500 to 30000.

Especially, from the viewpoint of further reducing the thermal expansion coefficient and the elastic modulus of the insulating layer to further mitigate the warpage problem, the epoxy group-containing acrylic resin having a Tg of 25°C or lower is preferable.

The epoxy group-containing acrylic resin having a Tg of 25°C or lower is preferably an epoxy group-containing acrylic ester copolymer resin having a Tg of 25°C or lower. Specific examples thereof may include "SG-80H" (an epoxy group-containing acrylic ester copolymer resin (number average molecular weight Mn: 350000 g/mol, epoxy value 0.07 eq/kg, Tg 11°C) manufactured by Nagase Chemtex Corporation), and "SG-P3" (an epoxy group-containing acrylic ester copolymer resin (number average molecular weight Mn: 850000 g/mol, epoxy value 0.21 eq/kg, Tg 12°C)) manufactured by Nagase Chemtex Corporation.

The acid anhydride group-containing acrylic resin having a Tg of 25°C or lower is preferably an acid anhydride group-containing acrylic ester copolymer resin having a Tg of 25°C or lower.

The phenolic hydroxy group-containing acrylic resin having a Tg of 25°C or lower is preferably a phenolic

hydroxy group-containing acrylic ester copolymer resin having a Tg of 25°C or lower. Specific examples thereof may include "SG-790" (an epoxy group-containing acrylic ester copolymer resin (number average molecular weight Mn: 500000 g/mol, hydroxy group value of 40 mg KOH/kg, Tg - 32°C)) manufactured by Nagase Chemtex Corporation.

Among these, the (C) component is preferably at least one selected from the group consisting of a polybutadiene skeleton epoxy resin that is liquid at 25°C, a hydrogenated polybutadiene skeleton epoxy resin that is liquid at 25°C, and an epoxy group-containing acrylic ester copolymer having a Tg of 25°C or lower.

From the viewpoint of reducing the thermal expansion coefficient and the elastic modulus of the insulating layer to mitigate the warpage problem, when the content of the resin component is assumed to be 100% by mass, the content of the (C) component in the resin composition according to the present invention is 10 to 40% by mass, preferably 12% by mass to 35% by mass, and more preferably 15% by mass to 32% by mass. As described herein, the "resin component" is a component other than the (B) inorganic filler in the resin composition. That is, other than the (A) component and the (C) component, a resin contained as a later-described curing agent and the like is also contained.

In particular, when the mass of the (A) component is assumed to be 100% by mass, the content of the (C) component in the resin composition is preferably 15% by mass to 80% by mass, and more preferably 20% by mass to 70% by mass.

Although the content of the (C) component in the resin composition according to the present invention is not particularly limited as long as the relative ratio to the above-described resin component is satisfied, the lower limit thereof is preferably not less than 3% by mass and not less than 5% by mass, and the upper limit thereof is preferably not more than 20% by mass.

From the viewpoint of reducing the thermal expansion coefficient and the elastic modulus of the insulating layer to mitigate the warpage problem, the ratio by mass between the (B) component and the (C) component ((B) component/(C) component) is preferably 2 or more, and more preferably 3 or more. Also, from the viewpoint of mitigating the warpage problem, the upper limit of the ratio by mass of (B) component/(C) component is preferably 25 or less, more preferably 20 or less, and further preferably 15 or less.

The resin composition according to the present invention may include a curing agent in addition to the above-described components of (A) to (C).

Although the curing agent is not particularly limited as long as it has a function of curing the resin used in the present invention, examples thereof may include a phenol-based curing agent, a naphthol-based curing agent, an active ester-based curing agent, a benzoxazine-based curing agent and a cyanate ester-based curing agent. The curing agents may be used singly or in any combination of two or more. Among these, the curing agent in the present invention is preferably at least one selected from the group consisting of a phenol-based curing agent, a naphthol-based curing agent and an active ester-based curing agent, and is more preferably any combination of two or more elected from the group consisting of a phenol-based curing agent, a naphthol-based curing agent and an active ester-based curing agent.

From the viewpoint of thermal resistance and water resistance, the phenol-based curing agent and the naphthol-based curing agent are preferably a phenol-based curing agent having a novolac structure, or a naphthol-based curing agent having a novolac structure. Also, from the viewpoint of adhesive properties to the conductive layer (circuit wiring), a nitrogen-containing phenol-based curing agent is preferable; and a triazine skeleton-containing phenol-based curing agent is more preferable. Among these, from the viewpoint of highly satisfying the thermal

resistance, the water resistance, and the adhesive properties (peel strength) to the conductive layer, it is preferable that a triazine skeleton-containing phenol novolac resin be used as a curing agent.

Specific examples of the phenol-based curing agent and the naphthol-based curing agent may include "MEH-7700," "MEH-7810," and "MEH-7851" manufactured by Meiwa Plastic Industries, Ltd.; "NHN," "CBN," and "GPH" manufactured by Nippon Kayaku Co., Ltd.; "SN170," "SN180," "SN190," "SN475," "SN485," "SN495," "SN375," and "SN395" manufactured by Tohto Kasei Co., Ltd.; and "LA7052," "LA7054," "LA3018," and "HPC-9500" manufactured by DIC Corporation.

Although the active ester-based curing agent is not particularly limited, a compound that contains in one molecule two or more ester groups having a high reaction activity is preferably used in general. Examples of such compounds may include phenol esters, thiophenol esters, N-hydroxyamine esters, and esters of heterocyclic hydroxy compounds. The active ester-based curing agent is preferably obtained by a condensation reaction between a carboxylic acid compound and/or a thiocarboxylic acid compound with a hydroxy compound and/or a thiol compound. Particularly, from the viewpoint of improving thermal resistance, an active ester-based curing agent obtained

from a carboxylic acid compound and a hydroxy compound is preferable, and an active ester-based curing agent obtained from a carboxylic acid compound and a phenol compound and/or a naphthol compound is more preferable. Examples of the carboxylic acid compound may include benzoic acid, acetic acid, succinic acid, maleic acid, itaconic acid, phthalic acid, isophthalic acid, terephthalic acid, and pyromellitic acid. Examples of the phenol compound or the naphthol compound may include hydroquinone, resorcin, bisphenol A, bisphenol F, bisphenol S, phenolphthalin, methylated bisphenol A, methylated bisphenol F, methylated bisphenol S, phenol, o-cresol, m-cresol, p-cresol, catechol, α -naphthol, β -naphthol, 1,5-dihydroxy naphthalene, 1,6-dihydroxy naphthalene, 2,6-dihydroxy naphthalene, dihydroxybenzophenone, trihydroxybenzophenone, tetrahydroxybenzophenone, phloroglucin, benzenetriol, a dicyclopentadiene-type diphenol compound (a polycyclopentadiene-type diphenol compound), and phenol novolac.

Specifically, an active ester compound containing a dicyclopentadiene-type diphenol condensation structure, an active ester compound containing a naphthalene structure, an active ester compound containing an acetylated product of phenol novolac, and an active ester compound containing a benzoylated product of phenol novolac are preferable.

Among these, the active ester compound containing a naphthalene structure and the active ester compound containing a dicyclopentadiene-type diphenol condensation structure are more preferable.

Examples of a commercially available active ester-based curing agent may include "EXB9451," "EXB9460," "EXB9460S," and "HPC-8000-65T" (manufactured by DIC Corporation) as the active ester compound containing a dicyclopentadiene-type diphenol condensation structure; "EXB9416-70BK" (manufactured by DIC Corporation) as the active ester compound containing a naphthalene structure; "DC808" (manufactured by Mitsubishi Chemical Corporation) as the active ester compound containing an acetylated product of phenol novolac; and "YLH1026" (manufactured by Mitsubishi Chemical Corporation) as the active ester compound containing a benzoylated product of phenol novolac. Specific examples of the benzoxazine-based curing agent may include "HFB2006M" manufactured by Showa Highpolymer Co., Ltd., and "P-d" and "F-a" manufactured by Shikoku Chemicals Corporation.

Examples of the cyanate ester-based curing agent may include bifunctional cyanate resins such as bisphenol A dicyanate, polyphenol cyanate (oligo(3-methylene-1,5-phenylene cyanate)), 4,4'-methylenebis(2,6-dimethylphenyl cyanate), 4,4'-ethyldiene diphenyl dicyanate,

hexafluorobisphenol A dicyanate, 2,2-bis(4-cyanate)phenylpropane, 1,1-bis(4-cyanatephenyl methane), bis(4-cyanate-3,5-dimethylphenyl)methane, 1,3-bis(4-cyanatephenyl-1-(methylethylidene))benzene, bis(4-cyanatephenyl)thioether and bis(4-cyanatephenyl)ether; multifunctional cyanate resins derived from phenol novolac, cresol novolac and the like; and a prepolymer obtained by partially triazinating these cyanate resins. Specific examples of the cyanate ester-based curing agent may include "PT30" and "PT60" (both are a phenol novolac-type multifunctional cyanate ester resin) and "BA230" (a prepolymer that is prepared as a trimer by partially or wholly triazinating bisphenol A dicyanate) manufactured by Lonza Japan Ltd.

The content of the curing agent in the resin composition according to the present invention may be adjusted such that the ratio of [total number of epoxy groups in the (A) component]:[total number of reactive groups in the curing agent] falls preferably within a range of 1:0.2 to 1:2, more preferably a range of 1:0.3 to 1:1.5, and further preferably a range of 1:0.4 to 1:1. As described herein, the reactive group in the curing agent is an active hydroxy group, an active ester group and the like, and varies depending on the type of the curing agent. Also, the total number of epoxy groups in the (A) component

is a sum of values for all epoxy resins used as the (A) component, wherein each of the values is obtained by dividing the mass of the solid content for each epoxy resin by the epoxy equivalent. The total number of reactive groups in the curing agent is a sum of values for all curing agents, wherein each of the values is obtained by dividing the mass of the solid content for each curing agent by the reactive group equivalent.

The resin composition according to the present invention may further include, as necessary, additives such as a curing accelerator, a thermoplastic resin, and a flame retardant.

Examples of the curing accelerator may include an organic phosphine compound, an imidazole compound, an amine adduct compound, and a tertiary amine compound. When the total of the non-volatile components of the epoxy resin and the curing agent in the resin composition is assumed to be 100% by mass, the content of the curing accelerator to be used is preferably 0.05% by mass to 3% by mass. The curing accelerators may be used singly or in any combination of two or more.

Examples of the flame retardant may include an organic phosphorus-based flame retardant, an organic nitrogen-containing phosphorus compound, a nitrogen compound, a silicone-based flame retardant, and a metal hydroxide. The

flame retardants may be used singly or in any combination of two or more. The content of the flame retardant in the resin composition is, but not particularly limited to, preferably 0.5% by mass to 10% by mass, more preferably 1% by mass to 9% by mass, and further preferably 1% by mass to 8% by mass.

Examples of the thermoplastic resin may include phenoxy resins, polyvinyl acetal resins, polyimide resins, polyamide imide resins, polyethersulfone resins, and polysulfone resins. The thermoplastic resins may be used singly or in any combination of two or more.

The polystyrene equivalent weight average molecular weight of the thermoplastic resin is preferably 8,000 to 70,000, more preferably 10,000 to 60,000, and further preferably 20,000 to 60,000. The polystyrene equivalent weight average molecular weight of the thermoplastic resin is measured by gel permeation chromatography (GPC). Specifically, the polystyrene equivalent weight average molecular weight of the thermoplastic resin can be measured at a column temperature of 40°C, using LC-9A/RID-6A manufactured by Shimadzu Corporation as a measuring device, Shodex K-800P/K-804L/K-804L manufactured by Showa Denko K.K. as a column, and chloroform and the like as a mobile phase; and can be calculated using a standard polystyrene calibration curve.

Examples of the phenoxy resins may include a phenoxy resin having at least one skeleton selected from the group consisting of a bisphenol A skeleton, a bisphenol F skeleton, a bisphenol S skeleton, a bisphenol acetophenone skeleton, a novolac skeleton, a biphenyl skeleton, a fluorene skeleton, a dicyclopentadiene skeleton, a norbornene skeleton, a naphthalene skeleton, an anthracene skeleton, an adamantane skeleton, a terpene skeleton and a trimethyl cyclohexane skeleton. The terminal of the phenoxy resin may be any functional group such as a phenolic hydroxy group and an epoxy group. The phenoxy resins may be used singly or in any combination of two or more. Specific examples of the phenoxy resins may include "1256" and "4250" (both are bisphenol A skeleton-containing phenoxy resins), "YX8100" (a bisphenol S skeleton-containing phenoxy resin), and "YX6954" (a bisphenol acetophenone skeleton-containing phenoxy resin) manufactured by Mitsubishi Chemical Corporation. Other than these, "FX280" and "FX293" manufactured by Tohto Kasei Co., Ltd.; "YL7553," "YL6794," "YL7213," "YL7290," and "YL7482" manufactured by Mitsubishi Chemical Corporation; and the like may be included.

Specific examples of the polyvinyl acetal resins may include Denka Butyral 4000-2, 5000-A, 6000-C, and 6000-EP manufactured by Denki Kagaku Kogyo Kabushiki Kaisha; and

Eslec BH series, BX series, KS series, BL series, and BM series manufactured by Sekisui Chemical Co., Ltd.

Specific examples of the polyimide resins may include "Rikacoat SN20" and "Rikacoat PN20" manufactured by New Japan Chemical Co., Ltd. Specific examples of the polyimide resins may also include a modified polyimide such as a linear polyimide obtained by reaction among a bifunctional hydroxy group-terminated polybutadiene, a diisocyanate compound, and a tetrabasic acid anhydride (disclosed in Japanese Patent Application Laid-Open No. 2006-37083), and a polysiloxane skeleton-containing polyimide (disclosed in Japanese Patent Application Laid-Open No. 2002-12667 and Japanese Patent Application Laid-Open No. 2000-319386).

Specific examples of the polyamide imide resins may include "Vylomax HR11NN" and "Vylomax HR16NN" manufactured by Toyo Boseki Kabushiki Kaisha. Specific examples of the polyamide imide resins may also include a modified polyamide imide such as polysiloxane skeleton-containing polyamide imides "KS9100" and "KS9300" manufactured by Hitachi Chemical Co., Ltd.

Specific examples of the polyethersulfone resins may include "PES5003P" manufactured by Sumitomo Chemical Company, Limited.

Specific examples of the polysulfone resins may include polysulfones "P1700" and "P3500" manufactured by Solvay Advanced Polymers, L.L.C.

The content of the thermoplastic resin in the resin composition is preferably 0.1% by mass to 10% by mass.

The resin composition according to the present invention may include, as necessary, other additives. Examples of such additives may include organometallic compounds such as an organic copper compound, an organic zinc compound and an organic cobalt compound, as well as organic fillers, thickening agents, antifoaming agents, leveling agents, adhesion imparting agents and coloring agents.

The resin composition according to the present invention may be impregnated in a sheet-like fibrous base material to obtain a prepreg. The prepreg is obtained by impregnating the resin composition according to the present invention in the sheet-like fibrous base material.

The sheet-like fibrous base material used in the prepreg is not particularly limited, and may be a material that is usually used as a base material for a prepreg. Examples of such a base material for a prepreg may include glass cloth, aramid nonwoven fabric and liquid crystal polymer nonwoven fabric. When used for the formation of the insulating layer of the multilayer printed circuit

board, a thin sheet-like fibrous base material having a thickness of 50 μm or less is suitably used. Particularly, a sheet-like fibrous base material having a thickness of 10 μm to 40 μm is preferable; a sheet-like fibrous base material having a thickness of 10 μm to 30 μm is more preferable; and a sheet-like fibrous base material having a thickness of 10 to 20 μm is further preferable. Specific examples of the glass cloth base material used as the sheet-like fibrous base material may include "Style 1027MS" (warp density of 75 yarns/25 mm, weft density of 75 yarns/25 mm, cloth weight of 20 g/m^2 , and thickness of 19 μm) manufactured by Asahi-Schwebel Co., Ltd., "Style 1037MS" (warp density of 70 yarns/25 mm, weft density of 73 yarns/25 mm, cloth weight of 24 g/m^2 , and thickness of 28 μm) manufactured by Asahi-Schwebel Co., Ltd., "1078" (warp density of 54 yarns/25 mm, weft density of 54 yarns/25 mm, cloth weight of 48 g/m^2 , and thickness of 43 μm) manufactured by Arisawa Mfg. Co., Ltd., "1037NS" (warp density of 72 yarns/25 mm, weft density of 69 yarns/25 mm, cloth weight of 23 g/m^2 , and thickness of 21 μm) manufactured by Arisawa Mfg. Co., Ltd., "1027NS" (warp density of 75 yarns/25 mm, weft density of 75 yarns/25 mm, cloth weight of 19.5 g/m^2 , and thickness of 16 μm) manufactured by Arisawa Mfg. Co., Ltd., "1015NS" (warp

density of 95 yarns/25 mm, weft density of 95 yarns/25 mm, cloth weight of 17.5 g/m², and thickness of 15 μm) manufactured by Arisawa Mfg. Co., Ltd., and "1000NS" (warp density of 85 yarns/25 mm, weft density of 85 yarns/25 mm, cloth weight of 11 g/m², and thickness of 10 μm) manufactured by Arisawa Mfg. Co., Ltd. Also, specific examples of the liquid crystal polymer nonwoven fabric may include "Vecrus" (basis weight of 6 to 15 g/m²) and "Vectran," manufactured by Kuraray Co., Ltd. and produced from aromatic polyester nonwoven fabric by a melt-blow method.

The prepreg may be manufactured by known methods such as a hot melt method and a solvent method.

The resin composition according to the present invention can be suitably used as a resin composition for forming an insulating layer of a multilayer printed circuit board (a resin composition for an insulating layer of a multilayer printed circuit board). By forming the insulating layer of the multilayer printed circuit board with the resin composition according to the present invention, there can be realized an insulating layer that is low in both thermal expansion coefficient and elastic modulus. Thus, the warpage problem can be significantly improved. Especially, in the manufacture of the multilayer

printed circuit board by the build-up system, the resin composition according to the present invention can be suitably used as a resin composition for forming an insulating layer (a resin composition for a build-up insulating layer of a multilayer printed circuit board), and further suitably used as a resin composition for forming an insulating layer on which a conductive layer is formed by plating (a resin composition for a build-up insulating layer of a multilayer printed circuit board in which a conductive layer is plated). Furthermore, since the resin composition according to the present invention is excellent in fluidity and component embedding properties, it can be suitably used even when the multilayer printed circuit board is a circuit board with built-in parts. That is, the resin composition according to the present invention can be suitably used as a resin composition for embedding parts for the circuit board with built-in parts (a resin composition for embedding parts). For the core substrate used in the manufacture of the circuit board with built-in parts, the density of the cavities tends to become higher since the core substrate has cavities for embedding the parts and there is a demand for reducing in size of the circuit board with built-in parts itself. Thus, the warpage problem attributable to insufficient stiffness of the core substrate tends to be more serious. However, by

using the resin composition according to the present invention as the resin composition for embedding parts, the warpage problem can be significantly mitigated even when a core substrate that has a high cavity density and is thin is used.

[Adhesive Film]

The resin composition according to the present invention can be used to form an adhesive film.

In an embodiment, the adhesive film according to the present invention contains a support and a resin composition layer to be adhered to the support. The resin composition layer includes the resin composition according to the present invention.

The adhesive film can be formed by, for example, preparing a resin varnish in which the resin composition according to the present invention is dissolved in an organic solvent, coating the support with the resin varnish using a die coater or the like, and drying the resin varnish.

Examples of the organic solvent may include ketones such as acetone, methyl ethyl ketone (hereinafter also referred to as "MEK"), and cyclohexanone; acetate esters such as ethyl acetate, butyl acetate, Cellosolve acetate, propylene glycol monomethyl ether acetate, and carbitol acetate; carbitols such as Cellosolve and butyl carbitol;

aromatic hydrocarbons such as toluene and xylene; and amide-based solvents such as dimethylformamide, dimethylacetamide, and N-methylpyrrolidone. The organic solvents may be used singly or in any combination of two or more.

The resin varnish may be dried by known drying methods such as heating and hot air blowing. When using the resin varnish containing the organic solvent in an amount of 30% by mass to 60% by mass, for example, the adhesive film can be formed by performing drying at 50°C to 150°C for 3 minutes to 10 minutes, which varies depending on the boiling point of the organic solvent in the resin varnish.

Examples of the support used in the formation of the adhesive film may include a film made of plastic materials, metal foil (such as copper foil and aluminum foil), and mold release paper. The film made of plastic materials is suitably used. Examples of the plastic materials may include polyester such as polyethylene terephthalate (hereinafter, sometimes abbreviated as "PET") and polyethylene naphthalate (hereinafter, sometimes abbreviated as "PEN"); polycarbonate (hereinafter, sometimes abbreviated as "PC"); acryl such as polymethyl methacrylate (PMMA); cyclic polyolefin, triacetyl cellulose (TAC), polyether sulfide (PES), polyetherketone, and polyimide. Among these, polyethylene terephthalate and

polyethylene naphthalate are preferable; and inexpensive polyethylene naphthalate is particularly preferable. In a preferred embodiment, the support is a polyethylene terephthalate film.

The support surface to which the resin composition layer is adhered may be subjected to mat treatment and corona treatment.

Also, the support to be used may include a support with a mold release layer. The support with a mold release layer has the mold release layer on the surface to be adhered to the resin composition layer. An example of a mold release agent used in the mold release layer of the support with a mold release layer may be at least one mold release agent selected from the group consisting of alkyd resins, polyolefin resins, urethane resins, and silicone resins.

In the present invention, as the support with a mold release layer, a commercially available product may be used. Examples of the commercially available product may include "SK-1," "AL-5," and "AL-7" manufactured by Lintec Corporation. These products are a PET film having a mold release layer that contains an alkyd resin-based mold release agent as a main component.

The thickness of the support is, but not particularly limited to, preferably 5 μm to 75 μm , and more preferably

10 μm to 60 μm . Here, when the support is the support with a mold release layer, it is preferable that the thickness of the whole support with a mold release layer be within the above-described range.

In the adhesive film according to the present invention, the thickness of the resin composition layer is, but not particularly limited to, preferably 100 μm or less, more preferably 80 μm or less, further preferably 60 μm or less, and further more preferably 50 μm or less, from the viewpoint of achieving a thinner multilayer printed circuit board. The lower limit of the thickness of the resin composition layer is usually 15 μm or more.

In the adhesive film according to the present invention, a protective film conforming to the support can be further laminated on the resin composition layer surface which the support is not in contact with (that is, the surface opposite to the support). The thickness of the protective film is, but not particularly limited to, for example, 1 μm to 40 μm . By laminating the protective film, the surface of the resin composition layer is protected from deposition of dust or the like and scratches. The adhesive film can be wound into a roll shape and stored. Thus, when forming the insulating layer in the manufacture

of the multilayer printed circuit board, the protective film is peeled off to allow the adhesive film to be used.

[Cured body]

The resin composition according to the present invention can be thermally cured into a cured body.

The condition for thermal curing is not particularly limited, and the condition normally employed when forming an insulating layer in the manufacture of the multilayer printed circuit board may be used.

For example, although the condition for thermal curing also varies depending on the formulation of the resin composition or the like, the curing temperature may be in the range of 120°C to 240°C (preferably in the range of 150°C to 210°C, and more preferably in the range of 170°C to 190°C); and the curing time may be in the range of 5 minutes to 90 minutes (preferably 10 minutes to 75 minutes, and more preferably 15 minutes to 60 minutes).

Prior to the thermal curing, the resin composition may be preheated at a temperature lower than the curing temperature. For example, prior to the thermal curing, the resin composition may be preheated at a temperature 50°C or higher and lower than 120°C (preferably 60°C or higher and 110°C or lower, and more preferably 70°C or higher and 100°C or lower), for 5 minutes or longer (preferably 5 minutes to

150 minutes, and more preferably 15 minutes to 120 minutes).

The thermal curing is preferably performed under atmospheric pressure (under normal pressure).

The glass transition temperature (T_g) of the cured body according to the present invention is preferably 170°C or higher, more preferably 180°C or higher, further preferably 190°C or higher, further more preferably 200°C or higher, and particularly preferably 210°C or higher or 220°C or higher.

The linear thermal expansion coefficient (α) of the cured body according to the present invention is preferably 25 ppm or less, more preferably 20 ppm or less, further preferably 15 ppm or less, and further more preferably 10 ppm or less. Although the lower limit of the linear thermal expansion coefficient (α) is not particularly limited, it is usually 1 ppm or more. In the present invention, the linear thermal expansion coefficient (α) of the cured body is a linear thermal expansion coefficient (α) at 25 to 150°C in a plane direction measured under a tensile mode in a thermomechanical analysis (TMA). Examples of a thermomechanical analyzer that can be used for measuring the linear thermal expansion coefficient (α)

of the cured body may include "Thermo Plus TMA8310" manufactured by Rigaku Corporation.

The elastic modulus (E) of the cured body according to the present invention is preferably 14 GPa or less, more preferably 12 GPa or less, further preferably 9 GPa or less, further more preferably 5 GPa or less, and particularly preferably 4 GPa or less. Although the lower limit of the elastic modulus (E) is not particularly limited, it is usually 1 GPa or more. In the present invention, the elastic modulus (E) of the cured body is an elastic modulus (E) measured at room temperature (20 to 30°C) using a universal testing machine manufactured by Orientec Co., Ltd.

The value of the product $\alpha \times E$ of the linear thermal expansion coefficient (α) and the elastic modulus (E:GPa) of the cured body according to the present invention is preferably 150 or less, more preferably 125 or less, further preferably 100 or less, further more preferably 95 or less, particularly preferably 90 or less, 85 or less, 80 or less, 75 or less, 70 or less, 65 or less, 60 or less, or 55 or less. Also, although the lower limit of the product is not particularly limited, it is normally 10 or more.

In a preferred embodiment, the linear thermal expansion coefficient (α) of the cured body according to

the present invention is 25 ppm or more, and the product $\alpha \times E$ of the linear thermal expansion coefficient (α) and the elastic modulus (E:GPa) is 150 or less.

As described above, the cured body according to the present invention, which is obtained by thermally curing the resin composition that contains the above-described specific components of (A) to (C) in the specific amount ratios, is low in both the thermal expansion coefficient (α) and the elastic modulus (E), and extraordinarily low in the value of the product $\alpha \times E$ thereof. Accordingly, the use of the cured body according to the present invention as the insulating layer of the multilayer printed circuit board or the substrate with built-in parts enables the warpage problem to be advantageously mitigated.

Examples

Hereinafter, the present invention will now be described in detail using Examples and Comparative Examples. However, the present invention is not limited by these Examples and Comparative Examples in any sense. In the description below, "parts" means "parts by mass".

<Measurement Method and Evaluation Method>

First, various measurement methods and evaluation methods are described.

<Evaluation of Warpage>

(1) Laminate of Adhesive Film

An adhesive film prepared in Examples and Comparative Examples was cut out into a piece having a size of 9.5 cm square. The cut out adhesive film was laminated to a coarsened surface of a 10 cm square piece cut out from a copper foil (3EC-III, thickness 35 μm) manufactured by Mitsui Mining & Smelting Co., Ltd., using a batch-type vacuum pressure laminator ("MVLP-500" manufactured by Meiki Co., Ltd.).

The pressure was reduced for 30 seconds so that the pressure was adjusted to 13 hPa or lower. Then, the laminate was pressure-bonded at 120°C for 30 seconds under a pressure of 0.74 MPa. Thus, a metal foil with a resin composition layer was prepared. Thereafter, a PET film was peeled off.

(2) Curing of Resin Composition Layer

The four sides of the metal foil with the resin composition layer obtained in the above (1) were bonded to SUS boards having a thickness of 1 mm with polyimide tapes such that the resin composition layer faced upward. Then, the resin composition layer was cured under the curing condition of 180°C and 30 minutes.

(3) Measurement of Warpage

The polyimide tapes on three of the four sides of the metal foil with the resin composition layer obtained in the above (2) were peeled off, and the highest point from the SUS board was measured to obtain the value of warpage. Then, when the magnitude of warpage was less than 1 cm, "o" was assigned; when it was 1 cm or more and less than 3 cm, "Δ" was assigned; and when it was 3 cm or more, "x" was assigned.

<Measurement and Evaluation of Linear Thermal Expansion Coefficient>

An adhesive film prepared in Examples and Comparative Examples was laminated to a polyimide film (Upilex S, manufactured by Ube Industries, Ltd.), using a batch-type vacuum pressure laminator ("MVLP-500" manufactured by Meiki Co., Ltd.). The pressure was reduced for 30 seconds so that the pressure was adjusted to 13 hPa or lower. Then, the laminate was pressure-bonded at 120°C for 30 seconds under a pressure of 0.74 MPa. Then, a PET film was peeled off, and the resin composition was cured under the condition of 190°C and 90 minutes. Thereafter, the polyimide film was peeled off to obtain the cured product.

The obtained cured product sample was cut into a specimen having a width of 5 mm and a length of 15 mm. The specimen was subjected to a thermomechanical analysis by a

tensile loading method, using a thermomechanical analyzer ("Thermo Plus TMA8310" manufactured by Rigaku Corporation). The sample was mounted in the analyzer, and then consecutively measured twice under the measurement condition of a load of 1 g and a temperature increasing rate of 5°C/minute. An average linear thermal expansion coefficient (ppm) from 25°C to 150°C in the second measurement was calculated. The value of the linear thermal expansion coefficient of less than 20 ppm was evaluated as "o"; the value of 20 ppm or more and less than 25 ppm was evaluated as "Δ"; and the value of 25 ppm or more was evaluated as "x".

<Measurement of Elastic Modulus>

The above-described adhesive film was thermally cured at 180°C for 90 minutes to obtain a sheet-like cured product. Next, a PET film was peeled off, and the cured product was subjected to a tensile test in accordance with Japanese Industrial Standard (JIS K7127) using a Tensilon universal testing machine (manufactured by A&D Company, Limited) to measure the elastic modulus.

<Measurement of Glass Transition Temperature>

The above cured product sample was cut into a specimen having a width of 5 mm and a length of 15 mm. The specimen was subjected to a thermomechanical analysis by a tensile

loading method using a dynamic viscoelasticity measuring device ("EXSTAR6000" manufactured by SII NanoTechnology Inc.). The sample was mounted in the measuring device, and then consecutively measured twice under the measurement condition of a load of 200 mN and a temperature increasing rate of 2°C/minute. A glass transition temperature (°C) was calculated from a point at which the slope of the dimension change signal in the second measurement changed. (Example 1)

<Preparation of Resin Varnish>

To 15 parts of methyl ethyl ketone (MEK) and 15 parts of cyclohexanone, 25 parts of a naphthalene-type tetrafunctional epoxy resin (number average molecular weight Mn: 700 g/mol, epoxy equivalent of 163, "HP4710" manufactured by DIC Corporation), 25 parts of a naphthalene-type epoxy resin (number average molecular weight Mn: 490 g/mol, epoxy equivalent of 186, "EXA7311-G4S" manufactured by DIC Corporation), and a polybutadiene skeleton-containing epoxy resin (number average molecular weight Mn: 5900 g/mol, epoxy equivalent of 190, "PB3600" manufactured by Daicel Chemical Industries, Ltd.) were added, and heated while stirring to be dissolved therein. To the obtained solution, 15 parts of an MEK solution of a triazine-containing phenol novolac resin with 60% by weight of the solid content (hydroxy group equivalent of 125,

"LA7054" manufactured by DIC Corporation, nitrogen content of approximately 12% by weight), 25 parts of an MEK solution of a naphthol-based curing agent with 60% by weight of the solid content (hydroxy group equivalent of 153, "HPC-9500" manufactured by DIC Corporation), and 160 parts of a spherical silica (average particle size of 4.0 μm , specific surface area of 2.4m²/g, "FB-5SDC" manufactured by Denki Kagaku Kogyo Kabushiki Kaisha) were mixed. The mixture was uniformly dispersed with a high speed rotary mixer to prepare a resin varnish.

<Preparation of Adhesive Film>

The resin varnish was uniformly applied using a die coater on a mold release treated surface of a PET film (thickness of 38 μm) treated with an alkyd-based mold release agent so that the thickness of the dried resin composition layer becomes 40 μm . Thereafter, the applied resin varnish was dried at 80 to 120°C (average 100°C) for 6 minutes, to obtain an adhesive film.

(Example 2)

An adhesive film was prepared in the same manner as in Example 1, except that the polybutadiene skeleton-containing epoxy resin was replaced with a hydrogenated polybutadiene skeleton-containing epoxy resin (number average molecular weight Mn: 2650 g/mol, epoxy equivalent

of 1500, "FCA-061L" manufactured by Nagase ChemteX Corporation).

(Example 3)

An adhesive film was prepared in the same manner as in Example 1, except that 25 parts of the polybutadiene skeleton-containing epoxy resin was replaced with 24.9 parts of an epoxy group-containing acrylic ester copolymer (number average molecular weight Mn: 850000 g/mol, epoxy value of 0.21 eq/kg, Tg of 12°C, "SG-P3" manufactured by Nagase ChemteX Corporation).

(Example 4)

An adhesive film was prepared in the same manner as in Example 1, except that the content of the spherical silica was changed to 70 parts.

(Example 5)

An adhesive film was prepared in the same manner as in Example 1, except that the content of the spherical silica was changed to 350 parts.

(Example 6)

An adhesive film was prepared in the same manner as in Example 1, except that the content of the polybutadiene skeleton-containing epoxy resin was changed to 10 parts.

(Example 7)

An adhesive film was prepared in the same manner as in Example 1, except that the content of the polybutadiene skeleton-containing epoxy resin was changed to 35 parts.

(Example 8)

An adhesive film was prepared in the same manner as in Example 1, except that 350 parts of a spherical silica (average particle size of 0.5 μm , specific surface area of 6.8 m^2/g , "SO-C2" manufactured by Admatechs Company Limited, treated with an aminosilane-based coupling agent) was used as the spherical silica.

(Example 9)

An adhesive film was prepared in the same manner as in Example 1, except that the resin varnish was prepared according to the following.

<Preparation of Resin Varnish>

To 15 parts of methyl ethyl ketone (MEK) and 15 parts of cyclohexanone, 50 parts of a naphthalene-type tetrafunctional epoxy resin (number average molecular weight M_n : 700 g/mol, epoxy equivalent of 163, "HP4710" manufactured by DIC Corporation) and 24 parts of an 80% by weight of MEK solution of a polybutadiene skeleton-containing epoxy resin (number average molecular weight M_n : 5900 g/mol, epoxy equivalent of 190, "PB3600" manufactured by Daicel Chemical Industries, Ltd.) were added, and heated while stirring to be dissolved therein. To the obtained

solution, 18 parts of an MEK solution of a triazine-containing phenol novolac resin with 60% by weight of the solid content (hydroxy group equivalent of 151, "LA3018" manufactured by DIC Corporation, nitrogen content of approximately 18% by weight), 20 parts of an MEK solution of an active ester-based curing agent with 65% by weight of the solid content (functional group equivalent of 223, "HPC-8000-65T" manufactured by DIC Corporation), and 140 parts of a spherical silica (average particle size of 4.0 μm , specific surface area of 2.4 m^2/g , "FB-5SDC" manufactured by Denki Kagaku Kogyo Kabushiki Kaisha) were mixed. The mixture was uniformly dispersed with a high speed rotary mixer to prepare a resin varnish.

(Comparative Example 1)

An adhesive film was prepared in the same manner as in Example 1, except that the polybutadiene skeleton-containing epoxy resin was not used and that the content of the spherical silica was changed to 120 parts.

(Comparative Example 2)

An adhesive film was prepared in the same manner as in Example 1, except that the content of the polybutadiene skeleton-containing epoxy resin was changed to 5.5 parts.

(Comparative Example 3)

An adhesive film was prepared in the same manner as in Example 1, except that the filling amount of the spherical silica was changed to 40 parts.

(Comparative Example 4)

An adhesive film was prepared in the same manner as in Example 1, except that the polybutadiene skeleton-containing epoxy resin was not used, and that the content of the spherical silica was changed to 300 parts.

Results are shown below.

[Table 1]

In terms of non-volatile component	Ex. 1	Ex. 2	Ex. 3	Ex. 4	Ex. 5	Ex. 6	Ex. 7	Ex. 8	Ex. 9	Comparative Ex. 1	Comparative Ex. 2	Comparative Ex. 3	Comparative Ex. 4
	Ex. 1	Ex. 2	Ex. 3	Ex. 4	Ex. 5	Ex. 6	Ex. 7	Ex. 8	Ex. 9	Comparative Ex. 1	Comparative Ex. 2	Comparative Ex. 3	Comparative Ex. 4
Epoxy resin that is solid at 25°C (A)		25	25	25	25	25	25	25	50	25	25	25	25
	Naphthalene-type epoxy resin	HP4710											
	Naphthalene-type epoxy resin	EXA-7311-G4S											
Functional Group-containing saturated and/or unsaturated butadiene skeleton epoxy resin that is liquid at 25°C, or functional group-containing acrylic resin having a Tg of 25°C or lower (C)		25		25	25	10	35	25	19.2		5.5	25	
	Polybutadiene skeleton epoxy resin	PB3600											
	Hydrogenated polybutadiene skeleton epoxy resin	FCA-061L	25										
	Acrylic ester copolymer resin	SG-P3	24.9										
	Triazine skeleton-containing phenol novolac resin	LA7054	9	9	15	15	15	9		9	9	9	9
	Naphthol-based curing agent	HPC-9500	15	15	15	15	15	15	10.8				
	Active ester-based curing agent	HPC-8000-65T							13				
	Spherical silica filler (B)	SO-C2	160	160	160	160	160	350	140				
	Filler amount (wt%) when a content of a non-volatile component in the resin composition is assumed to be 100% by mass	62	62	62	41	78	66	59	78	62	67	29	80
	Content (wt%) of reactive flexible component resin when a content of a resin component is assumed to be 100% by mass	25	25	25	25	12	32	25	21	0	7	25	0
	Linear thermal expansion coefficient (30-150°C; ppm)	O(18)	O(19)	O(18)	Δ(24)	O(8)	Δ(24)	Δ(23)	O(7)	O(18)	X(29)	X(45)	O(11)
	Elastic modulus (30°C; Gpa)	5	5	3	4	8	6	4	8	4	7	3.5	15
	Product of linear thermal expansion coefficient and elastic modulus	O(90)	O(95)	O(54)	O(96)	O(64)	O(144)	O(92)	O(56)	X(203)	X(189)	X(157.5)	X(165)
	Warpage (cm)	O(0.2)	O(0.3)	O(0.2)	Δ(1.5)	O(0.2)	Δ(2.1)	Δ(1.2)	O(0.2)	X(4.8)	X(4.3)	X(3.3)	X(3.8)
	Glass transition temperature (°C)	220	220	220	220	220	220	220	221	220	220	220	220

As described above, according to the resin composition of the present invention, warpage can be sufficiently suppressed.

CLAIMS

1. A resin composition comprising:
 - (A) an epoxy resin that is solid at 25°C;
 - (B) an inorganic filler; and
 - (C) at least one resin selected from the group consisting of a functional group-containing saturated butadiene resin that is liquid at 25°C, a functional group-containing unsaturated butadiene resin that is liquid at 25°C, and a functional group-containing acrylic resin having a glass transition temperature of 25°C or lower, wherein
 - when a content of a non-volatile component in the resin composition is assumed to be 100% by mass, a content of the (B) component is 40% by mass or more,
 - when a content of a resin component is assumed to be 100% by mass, a content of the (C) component is 10 to 40% by mass, and
 - a linear thermal expansion coefficient (α :ppm/°C) of a cured body obtained by curing the resin composition is 25 ppm or less.
2. The resin composition according to claim 1, wherein the (C) component has at least one functional group selected from the group consisting of an acid anhydride group, a phenolic hydroxy group, an epoxy group, an isocyanate group, and a urethane group.
3. The resin composition according to claim 1 or 2, wherein the (A) component is a multifunctional naphthalene skeleton epoxy resin.
4. The resin composition according to any one of claims 1 to 3, further comprising a curing agent.

5. The resin composition according to any one of claims 1 to 4, wherein the resin composition is a resin composition for embedding parts for the circuit board with built-in parts.

6. The resin composition according to any one of claims 1 to 5, wherein the inorganic filler has an average particle diameter of 10 μm or less, and is classified to remove a particle of 20 μm or more.

7. A cured body obtained by curing the resin composition according to any one of claim 1 to 6.

8. The cured body according to claim 7, having a glass transition temperature of 170°C or higher.

9. The cured body according to claim 7 or 8, wherein a product $\alpha \times E$ of a linear thermal expansion coefficient (α :ppm/°C) and an elastic modulus (E:GPa) is 150 or less.

10. A circuit board with built-in parts prepared using the cured body according to any one of claims 7 to 9.

RESIN COMPOSITION

ABSTRACT

A resin composition includes:

(A) an epoxy resin that is solid at 25°C;

(B) an inorganic filler; and

(C) at least one resin selected from the group consisting of a functional group-containing saturated butadiene resin that is liquid at 25°C, a functional group-containing unsaturated butadiene resin that is liquid at 25°C, and a functional group-containing acrylic resin having a glass transition temperature of 25°C or lower, wherein

when the content of a non-volatile component in the resin composition is assumed to be 100% by mass, the content of the (B) component is 40% by mass or more, and

when the content of a resin component is assumed to be 100% by mass, the content of the (C) component is 10 to 40% by mass.

[No Suitable Figure]