

## TITLE OF THE INVENTION

### SEALABLE RESIN SHEET

## TECHNICAL FIELD

[0001]

The present invention relates to a sealable resin sheet, in particular, to a sealable resin sheet for sealing an element.

## BACKGROUND ART

[0002]

It has been known to form a cured product from a sealable sheet by press sealing an element connected to a terminal of a board with a sealable sheet containing thermosetting resin, and thereafter curing the thermosetting resin by heating (for example, see Patent document 1 below).

[0003]

Patent Document 1 discloses an embodiment in which an entering amount of the cured product that enters between the chip and board from a lateral edge of the chip is 27  $\mu\text{m}$ .

## Citation List

### Patent Document

[0004]

Patent Document 1: Japanese Unexamined Patent Publication No. 2016-162909

## SUMMARY OF THE INVENTION

## PROBLEM TO BE SOLVED BY THE INVENTION

[0005]

However, when a plurality of elements varying in size or design or a plurality of elements connected to terminals varying in size or design is sealed, such variations make the fluidity of the thermosetting resin different. Thus, all the elements may not uniformly be sealed.

[0006]

Meanwhile, there is a need to ensure excellent sealing properties to seal the elements. In other words, to ensure excellent sealing properties, it is necessary to cover the side peripheral

surfaces of the elements and surely block the peripheral edges of the elements in gaps varying in thickness.

[0007]

The present invention provides a sealable resin sheet that has excellent sealing properties to seal elements varying in size or design and can reduce an entering amount of the cured product that enters two gaps varying in thickness.

#### MEANS FOR SOLVING THE PROBLEM

[0008]

The present invention [1] includes a sealable resin sheet containing thermosetting resin, a layered silicate compound, and thermoplastic resin, and used to seal an element, wherein a ratio ( $A/\alpha$ ) of a mass A of the layered silicate compound to a mass  $\alpha$  of the thermoplastic resin is 0.3 or more and 2.0 or less.

[0009]

The present invention [2] includes the sealable resin sheet described in [1] above, wherein a content ratio B of the layered silicate compound is 3 mass% or more and 7 mass% or less.

[0010]

The present invention [3] includes the sealable resin sheet described in [1] or [2] above, wherein the thermoplastic resin has a glass transition temperature Tg of -30°C or more and -10°C or less.

[0011]

The present invention [4] includes the sealable resin sheet described in any one of the above-described [1] to [3], wherein the layered silicate compound has a surface modified by an organic component.

[0012]

The present invention [5] includes the sealable resin sheet described in any one of the above-described [1] to [4], wherein the thermoplastic resin contains a carboxyl group.

#### EFFECTS OF THE INVENTION

[0013]

The sealable resin sheet of the present invention contains thermosetting resin, a layered silicate compound, and thermoplastic resin. The ratio of the mass of the layered silicate compound to the mass of the thermoplastic resin is 0.3 or more. Thus, when the sealable resin sheet is disposed on a plurality of elements varying in size or design or on a plurality of elements connected to a plurality of terminals varying in size or design and heated to form a cured product, an entering amount of the cured product that enters the gaps of the elements is controlled and the plurality of elements can uniformly be sealed.

[0014]

The ratio of the mass of the layered silicate compound to the mass of the thermoplastic resin is 2.0 or less. Thus, the side peripheral surfaces of the elements are surely covered and the peripheral edges in the gaps varying in thickness can surely be sealed. Thus, the sealable resin sheet has excellent sealing properties to seal elements.

#### BRIEF DESCRIPTION OF THE DRAWINGS

[0015]

[FIG. 1] FIG. 1A to FIG. 1D are cross-sectional views illustrating the steps of producing an electronic element package by sealing a plurality of electronic elements with an embodiment of the sealable resin sheet of the present invention. FIG. 1A depicts the step of preparing the sealable resin sheet. FIG. 1B depicts the step of preparing the electronic elements. FIG. 1C depicts the step of forming a sealing body by pressing the sealable resin sheet. FIG. 1D depicts the step of forming a cured product by heating the sealing body.

[FIG. 2] FIG. 2A to FIG. 2B are cross-sectional views illustrating an element mounting board including the electronic elements illustrated in FIG. 1B. FIG. 2A is a plan view thereof. FIG. 2B is a cross-sectional view taken along line X-X of FIG. 2A.

[FIG. 3] FIG. 3A to FIG. 3D are cross-sectional views illustrating the step of producing an electronic element package by sealing a plurality of electronic elements with a sealable multi-layer resin sheet including the sealable resin sheet illustrated in FIG. 1A and a second sealable resin sheet. FIG. 3A illustrates the step of preparing a sealable multi-layer resin sheet. FIG. 3B illustrates the step of preparing an electronic element. FIG. 3C illustrates the step of forming a sealing body by pressing the sealable multi-layer resin sheet. FIG. 3D illustrates the step of forming a cured product by heating the sealing body.

## DESCRIPTION OF THE EMBODIMENTS

[0016]

An embodiment of the sealable resin sheet of the present invention is described.

[0017]

The sealable resin sheet is a resin sheet used to seal an element and has an approximately plate shape (film shape) extending in a surface direction orthogonal to a thickness direction.

[0018]

The material of the sealable resin sheet contains thermosetting resin, a layered silicate compound, and thermoplastic resin. In other words, the material of the sealable resin sheet is a silicate-resin composition containing thermosetting resin, a layered silicate compound, and thermoplastic resin.

[0019]

Examples of the thermosetting resin include epoxy resin, silicone resin, urethane resin, polyimide resin, urea resin, melamine resin, and unsaturated polyester resin. These can be used alone or in combination of two or more.

[0020]

As the thermosetting resin, preferably, epoxy resin is used. The epoxy resin is prepared as an epoxy resin composition containing a main agent, a curing agent, and a curing accelerator.

[0021]

Examples of the main agent include: bifunctional epoxy resins such as bisphenol A epoxy resin, bisphenol F epoxy resin, modified bisphenol A epoxy resin, modified bisphenol F epoxy resin, and biphenyl epoxy resin; and trifunctional or more of multifunctional epoxy resins such as phenol novolak epoxy resin, cresol novolak epoxy resin, trishydroxyphenyl methane epoxy resin, tetraphenylol ethane epoxy resin, and dicyclopentadiene epoxy resin. These epoxy resins can be used alone or in combination of two or more. As the main agent, preferably, a bifunctional epoxy resin, more preferably, a bisphenol F epoxy resin is used.

[0022]

The lower limit of the epoxy equivalent of the main agent is, for example, 10 g/eq., preferably, 100 g/eq.. The upper limit of the epoxy equivalent of the main agent is, for example, 300 g/eq., preferably, 250 g/eq..

[0023]

The lower limit of the softening point of the main agent is, for example, 50°C, preferably, 70°C, more preferably, 72°C, even more preferably, 75°C. The upper limit of the softening point of the main agent is, for example, 130°C, preferably, 110°C, more preferably, 90°C.

[0024]

When the softening point of the main agent is the above-described lower limit or more, a sealable resin sheet 1 can flow in the step illustrated in FIG. 1C. Thus, the time for the step illustrated in FIG. 1C can be shortened, and a one-side surface in the thickness direction of the sealable resin sheet 1 can be flattened in the step illustrated in FIG. 1C.

[0025]

The lower limit of the ratio of the main agent in the thermosetting resin composition is, for example, 1 mass%, preferably, 2 mass%. The upper limit of the ratio of the main agent in the thermosetting resin composition is, for example, 30 mass%, preferably, 15 mass%. The lower limit of the ratio of the main agent in the epoxy resin composition is, for example, 30 mass%, preferably, 50 mass%. The upper limit of the ratio of the main agent in the epoxy resin composition is, for example, 80 mass%, preferably, 70 mass%.

[0026]

The curing agent is a latent curing agent that cures the main agent by heating. Examples of the curing agent include phenol resins such as phenol novolak resin. When the curing agent is a phenol resin, a cured product of the phenol resin and main agent has a high heat resistance and a high chemical resistance. Accordingly, the cured product has excellent reliability for sealing.

[0027]

The ratio of the curing agent is set so that the following equivalent ratio holds. Specifically, the lower limit of the total amount of the hydroxyl group in the phenol resin with respect to 1 equivalent of the epoxy group in the main agent is, for example, 0.7 equivalent,

preferably, 0.9 equivalent. The upper limit of the total amount of the hydroxyl group in the phenol resin with respect to 1 equivalent of the epoxy group in the main agent is, for example, 1.5 equivalent, preferably, 1.2 equivalent. Specifically, the lower limit of the contained amount of the curing agent with respect to 100 parts by mass of the main agent is, for example, 20 parts by mass, preferably, 40 parts by mass. The upper limit of the contained amount of the curing agent with respect to 100 parts by mass of the main agent is, for example, 80 parts by mass, preferably, 60 parts by mass.

[0028]

The curing accelerator is a catalyst (thermosetting catalyst) that promotes curing of the main agent by heating. Examples of the curing accelerator include organic phosphorus compounds and imidazole compounds such as 2-phenyl-4, 5-dihydroxy methyl imidazole (2PHZ-PW). Preferably, an imidazole compound is used. The lower limit of the contained amount of the curing accelerator with respect to 100 parts by mass of the main agent is, for example, 0.05 parts by mass. The upper limit of the contained amount of the curing accelerator with respect to 100 parts by mass of the main agent is, for example, 5 parts by mass.

[0029]

The content ratio of the material of the thermosetting resin is described below.

[0030]

The layered silicate compound is dispersed into the thermosetting resin and thermoplastic resin (resin matrix) in the material (sealable resin sheet). The layered silicate compound works as a flow adjuster when a sealing body and a cured product (describe below) are formed from the sealable resin sheet. Specifically, the layered silicate compound is a flow reducer at the curing used to reduce the fluidity of the cured product when the sealable resin sheet is heated and cured to form the cured product.

[0031]

The layered silicate compound is, for example, silicate salt having a structure (three-dimensional structure) in which layers each two-dimensionally extending (in the surface direction) are laminated in the thickness direction, and referred to as phyllosilicate.

[0032]

Specifically, examples of the layered silicate compound include: smectite such as montmorillonite, beidellite, non-tronite, saponite, hectorite, sauconite, and stevensite; kaolinite; halloysite; talc; and mica. To improve the mixability with the thermosetting resin, preferably, smectite is used as the layered silicate compound. More preferably, montmorillonite is used.

[0033]

The layered silicate compound may be an unmodified product of which surface is not modified. Alternatively, the layered silicate compound may be a modified product of which surface is modified by an organic component. To achieve excellent affinity with the thermosetting resin and thermoplastic resin, preferably, the layered silicate compound has a surface modified by an organic component. Specifically, as the layered silicate compound, organic smectite having a surface modified by an organic component is used. More preferably, organic bentonite having a surface modified by an organic component is used.

[0034]

Examples of the organic component include organic cation (onium ion) such as ammonium, imidazolium, pyridinium, and phosphonium.

[0035]

Examples of the ammonium include dimethyl distearyl ammonium, distearyl ammonium, octadecylammonium, hexylammonium, octylammonium, 2-hexylammonium, dodecylammonium, and trioctylammonium. Examples of the imidazolium include methyl stearyl imidazolium, distearyl imidazolium, methylhexylimidazolium, dihexylimidazolium, methyloctylimidazolium, dioctylimidazolium, methyldodecylimidazolium, and didodecylimidazolium. Examples of the pyridinium include stearyl pyridinium, hexylpyridinium, octylpyridinium, and dodecylpyridinium. Examples of the phosphonium include dimethyl distearyl phosphonium, distearyl phosphonium, octadecylphosphonium, hexylphosphonium, octylphosphonium, 2-hexylphosphonium, dodecylphosphonium, and trioctylphosphonium. These organic cations can be used alone or in combination of two or more. Preferably, ammonium, more preferably, dimethyl distearyl ammonium is used.

[0036]

As the organic layered silicate compound, preferably, organic smectite having a surface modified by ammonium is used. More preferably, organic bentonite having a surface modified by dimethyl distearyl ammonium is used.

[0037]

The lower limit of the average particle size of the layered silicate compound is, for example, 1 nm, preferably, 5 nm, more preferably, 10 nm. The upper limit of the average particle size of the layered silicate compound is, for example, 100  $\mu\text{m}$ , preferably, 50  $\mu\text{m}$ , more preferably, 10  $\mu\text{m}$ . The average particle size of the layered silicate compound is, for example, obtained as a D50 value (50% accumulation in median diameter) based on the particle size distribution obtained by a particle size distribution measurement method in a laser scattering method.

[0038]

A commercialized product can be used as the layered silicate compound. As an exemplary commercialized product of organic bentonite, the S-Ben series (manufactured by HOJUN.,Co.Ltd.) is used.

[0039]

The content ratio of the layered silicate compound is described below.

[0040]

The thermoplastic resin is a second flow reducer at curing that reduces the fluidity of the cured product, working together with the layered silicate compound. The thermoplastic resin is also a second flow improver at sealing and a second flow reducer at curing.

[0041]

Examples of the thermoplastic resin include natural rubber, butyl rubber, isoprene rubber, chloroprene rubber, ethylene-vinyl acetate copolymer, ethylene-acrylic acid copolymer, ethylene-acrylate copolymer, polybutadiene resin, polycarbonate resin, thermoplastic polyimide resin, polyamide resin (6-nylon, 6,6-nylon, or the like), phenoxy resin, acrylic resin, saturated polyester resin (PET or the like), polyamide imide resin, fluorine resin, and styrene-isobutylene-styrene block copolymer. These thermoplastic resins can be used alone or in combination of two or more.

[0042]

To improve the dispersibility with thermosetting resin, preferably, an acrylic resin is used as the thermoplastic resin.

[0043]

Examples of the acrylic resin include a (meth)acrylate copolymer (preferably, carboxyl group-containing acrylate copolymer) obtained by polymerizing a monomer component containing an alkyl (meth)acrylate having a straight-chain or branched alkyl group and another monomer (copolymerizable monomer).

[0044]

Examples of the alkyl group include an alkyl group having a carbon number of 1 to 6 such as methyl, ethyl, propyl, isopropyl, n-butyl, t-butyl, isobutyl, pentyl, and hexyl.

[0045]

Examples of the other monomer include: carboxyl group-containing monomers such as acrylic acid, methacrylic acid, carboxyethyl acrylate, carboxy pentyl acrylate, itaconic acid, maleic acid, fumaric acid, and crotonic acid; glycidyl group-containing monomers such as glycidyl acrylate, and glycidyl methacrylate; acid anhydride monomers maleic acid anhydride, and itaconic acid anhydride; hydroxy group-containing monomers such as 2-hydroxyethyl (meth)acrylate, 2-hydroxypropyl (meth)acrylate, 4-hydroxybutyl (meth)acrylate, 6-hydroxyhexyl (meth)acrylate, 8-hydroxyoctyl (meth)acrylate, 10-hydroxydecyl (meth)acrylate, and 12-hydroxylauryl (meth)acrylate or (4-hydroxymethylcyclohexyl)-methyl acrylate; sulfonic acid group-containing monomers such as styrenesulfonic acid, arylsulfonic acid, 2-(meth)acrylamide-2-methylpropanesulfonic acid, (meth)acrylamide propanesulfonic acid, sulfopropyl (meth)acrylate, and (meth)acryloyl oxynaphthalenesulfonic acid; phosphoric acid group-containing monomers such as 2-hydroxyethyl acryloyl phosphate; styrene monomers; and acrylonitriles. These can be used alone or in combination of two or more.

Preferably, a carboxyl group-containing monomer and a hydroxy group-containing monomer, more preferably, a carboxyl group-containing monomer is used.

[0046]

Preferable examples of the thermoplastic resin include a thermoplastic resin containing a carboxyl group, and a thermoplastic resin containing a carboxyl group and a hydroxy group.

[0047]

The lower limit of the glass transition temperature  $T_g$  of the thermoplastic resin is -70°C, preferably, -50°C, more preferably, -30°C. The upper limit of the glass transition temperature  $T_g$  of the thermoplastic resin is, for example, 0°C, preferably, -5°C, more preferably, -10°C. The glass transition temperature  $T_g$  is a theoretical value obtained by, for example, the Fox equation. A specific calculation method is described, for example, in Japanese Unexamined Patent Publication No. 2016-175976.

[0048]

When the glass transition temperature  $T_g$  of the thermoplastic resin is the above-described lower limit or more, the entering amount of the cured product to both the gaps varying in thickness can surely be reduced. When the glass transition temperature  $T_g$  of the thermoplastic resin is the above-described upper limit or less, formation of an unfilled portion outside the gaps can be suppressed.

[0049]

The lower limit of the weight average molecular weight of the thermoplastic resin is, for example, 100,000, preferably, 300,000. The upper limit of the weight average molecular weight of the thermoplastic resin is, for example, 1,200,000, preferably, 1,000,000. The weight average molecular weight is calibrated with standard polystyrene based on gel permeation chromatography (GPC).

[0050]

The content ratio of the thermoplastic resin is described next. The content ratio of the thermoplastic resin is adjusted to be a degree to produce a cured product from the sealable resin sheet.

[0051]

<Content Ratios of Thermosetting Resin, Layered Silicate Compound, and Thermoplastic Resin>

The lower limit of the ratio of a mass  $A$  of the layered silicate compound to a mass  $\alpha$  of the thermoplastic resin ( $A/\alpha$ ) is, 0.3, preferably, 0.5, more preferably, 0.6, even more preferably, 0.7, particularly preferably, 0.9.

[0052]

The upper limit of the ratio of the mass A of the layered silicate compound to the mass  $\alpha$  of the thermoplastic resin ( $A/\alpha$ ) is, 2.0, preferably, 1.4, more preferably, 1.2, even more preferably, 1.1, particularly preferably, 1.0.

[0053]

When the ratio of the mass A of the layered silicate compound to the mass  $\alpha$  of the thermoplastic resin ( $A/\alpha$ ) is less than the above-described lower limit, an unfilled portion, which is not filled with the cured product (where the cured product is not formed), is formed. Thus, reliability reduces.

[0054]

When the ratio of the mass A of the layered silicate compound to the mass  $\alpha$  of the thermoplastic resin ( $A/\alpha$ ) is more than the above-described upper limit, the entering amount of the cured product that enters the two gaps (a first gap 26 and a second gap 36, which are described below) varying in thickness cannot be reduced.

[0055]

The lower limit of a content ratio B of the layered silicate compound in the material (sealable resin sheet) is, for example, 0.1 mass%, preferably, 1 mass%, more preferably, 2 mass%, even more preferably, 3 mass%, particularly preferably, 4 mass%. The upper limit of the content ratio B of the layered silicate compound in the material (sealable resin sheet) is, for example, 25 mass%, preferably, 15 mass%, more preferably, 10 mass%, even more preferably, 8 mass%.

[0056]

When the content ratio B of the layered silicate compound is the above-described lower limit or more, the entering amount of the cured product that enters the gaps can be reduced by reducing the fluidity of the sealable resin sheet. When the content ratio B of the layered silicate compound is the above-described upper limit or less, a sealable resin sheet with a uniform fluidity can be produced by sufficiently dispersing the layered silicate compound in the sealable resin sheet.

[0057]

The lower limit of the ratio of the thermoplastic resin in the material (sealable resin sheet) is, for example, 1 mass%, preferably, 2 mass%. The upper limit of the ratio of the thermoplastic resin in the material is, for example, 10 mass%, preferably, 5 mass%.

[0058]

The lower limit of the ratio of the thermoplastic resin to the total amount of the thermoplastic resin and thermosetting resin is, for example, 5 mass%, preferably, 15 mass%. The upper limit of the ratio of the thermoplastic resin to the total amount of the thermoplastic resin and thermosetting resin is, for example, 40 mass%, preferably, 25 mass%.

[0059]

The lower limit of the content ratio of the thermosetting resin in the material (sealable resin sheet) is, for example, 5 mass%, preferably, 15 mass%, more preferably, 17 mass%, even more preferably, 18 mass%. The upper limit of the content ratio of the thermosetting resin in the material (sealable resin sheet) is, for example, 30 mass%, preferably, 25 mass%, more preferably, 20 mass%.

[0060]

<Other components>

A component other than the layered silicate compound, such as an inorganic filler, a pigment, a silane coupling agent, or another additive can further be added to the material.

[0061]

Examples of the inorganic filler include: a silicate salt compound other than the layered silicate compound such as orthosilicate salt, sorosilicate salt, and inosilicate salt; and a silicon compound (other than the layered silicate compound) such as quartz (silicic acid), silica (anhydrous silicic acid), and silicon nitride. Examples of the inorganic filler further include alumina, aluminum nitride, and boron nitride. These can be used alone or in combination of two or more. Preferably, a silicate salt compound other than the layered silicate compound, more preferably, silica is used.

[0062]

The shape of the inorganic filler particles is not especially limited. Example thereof include an approximately spherical shape, an approximately plate shape, an approximately aciculate shape, and an amorphous shape. Preferably, an approximately spherical shape is used.

[0063]

The upper limit of the average value (average particle size in the case of an approximately spherical shape) of the maximum lengths of the inorganic filler particles is, for example, 50  $\mu\text{m}$ , preferably, 20  $\mu\text{m}$ , more preferably, 10  $\mu\text{m}$ . The lower limit of the average value of the maximum lengths of the inorganic filler particles is, for example, 0.1  $\mu\text{m}$ , preferably, 0.5  $\mu\text{m}$ . The average particle size is, for example, obtained as a D50 value (50% accumulation in median diameter) based on the particle size distribution obtained by a particle size distribution measurement method in a laser scattering method.

[0064]

The inorganic filler can include a first filler and a second filler having an average value of the maximum lengths smaller than the average value of the maximum lengths of the first filler particles.

[0065]

The lower limit of the average value of the maximum lengths of the first filler particles is, for example, 1  $\mu\text{m}$ , preferably, 3  $\mu\text{m}$ . The upper limit of the average value of the maximum lengths of the first filler particles is, for example, 50  $\mu\text{m}$ , preferably, 30  $\mu\text{m}$ .

[0066]

The upper limit of the average value of the maximum lengths of the second filler particles is, for example, 0.9  $\mu\text{m}$ , preferably, 0.8  $\mu\text{m}$ . The lower limit of the average value of the maximum lengths of the second filler particles is, for example, 0.01  $\mu\text{m}$ , preferably, 0.1  $\mu\text{m}$ .

[0067]

The lower limit of the ratio of the average value of the maximum lengths of the first filler particles to the average value of the maximum lengths of the second filler particles is, for example, 2, preferably, 5. The upper limit of the ratio of the average value of the maximum lengths of the first filler particles to the average value of the maximum lengths of the second filler particles is, for example, 50, preferably, 20.

[0068]

The material of the first filler may be the same as or different from that of the second filler.

[0069]

In addition, the inorganic filler may have a surface partially or wholly treated with, for example, a silane coupling agent.

[0070]

The lower limit of the content ratio of the inorganic filler in the material (sealable resin sheet) is, for example, 50 mass%, preferably, 55 mass%, more preferably, 60 mass%, even more preferably, 65 mass%. The upper limit of the content ratio of the inorganic filler in the material (sealable resin sheet) is, for example, 90 mass%, preferably, 85 mass%, more preferably, 80 mass%, even more preferably, 75 mass%.

[0071]

The lower limit of the contained amount of the layered silicate compound with respect to 100 parts by mass of the inorganic filler is, for example, 1 part by mass, preferably, 2 parts by mass, more preferably, 3 parts by mass, even more preferably, 5 parts by mass. The upper limit of the contained amount of the layered silicate compound with respect to 100 parts by mass of the inorganic filler is, for example, 25 parts by mass, preferably, 20 parts by mass, more preferably, 15 parts by mass, even more preferably, 10 parts by mass.

[0072]

When the content ratio and/or contained amount of the inorganic filler is the above-described lower limit or more, the sealable resin sheet 1 can flow in the step illustrated in FIG. 1C.

[0073]

When the inorganic filler includes the first filler and second filler, the lower limit of the content ratio of the first filler in the material (sealable resin sheet) is, for example, 30 mass%, preferably, 40 mass% in the material. The upper limit of the content ratio of the first filler in the material (sealable resin sheet) is, for example, 60 mass%, preferably, 50 mass% in the material. The lower limit of the contained amount of the second filler to 100 parts by mass of the first filler is, for example, 30 parts by mass, preferably, 40 parts by mass, more preferably, 50 parts by mass. The upper limit of the contained amount of the second filler to 100 parts by mass of the first filler is, for example, 70 parts by mass, preferably, 60 parts by mass, more preferably, 55 parts by mass.

[0074]

Examples of the pigment include black pigments such as carbon black. The lower limit of the particle size of the pigment is, for example, 0.001  $\mu\text{m}$ . The upper limit of the particle size of the pigment is, for example, 1  $\mu\text{m}$ . The lower limit of the ratio of the pigment to the thermosetting resin composition is, for example, 0.1 mass%. The particle size of the pigment is an arithmetic average particle size obtained by observing the pigment with an electron microscope. The upper limit of the ratio of the pigment to the thermosetting resin composition is, for example, 2 mass%.

[0075]

Examples of the silane coupling agent include a silane coupling agent containing an epoxy group. Examples of the silane coupling agent containing an epoxy group include: 3-glycidoxy dialkyl dialkoxy silanes such as 3-glycidoxy propyl methyl dimethoxy silane and 3-glycidoxy propyl methyl diethoxy silane; and 3-glycidoxy alkyl trialkoxy silanes such as 3-glycidoxy propyl trimethoxy silane and 3-glycidoxy propyl triethoxy silane. Preferably, a 3-glycidoxy alkyl trialkoxy silane is used. The lower limit of the content ratio of the silane coupling agent in the thermosetting resin composition is, for example, 0.1 mass%, preferably, 1 mass%. The upper limit of the content ratio of the silane coupling agent in the thermosetting resin composition is, for example, 10 mass%, preferably, 5 mass%.

[0076]

To produce the sealable resin sheet, the material is prepared by blending the above-described components in the above-described ratios. Preferably, the above-described components are sufficiently stirred to uniformly disperse the layered silicate compound into the thermosetting resin and thermoplastic resin.

[0077]

As necessary, a solvent (for example, ketone such as methyl ethyl ketone) is further blended, thereby preparing a varnish. Thereafter, the varnish is applied to a peeling sheet not illustrated and then dried by heating, thereby producing a sealable resin sheet having a sheet shape. Alternatively, without preparing a varnish, the sealable resin sheet can be formed from the thermosetting resin composition by kneading extrusion.

[0078]

The formed sealable resin sheet is B stage (a semi-cured state), and specifically in a state before C stage. In short, the formed sealable resin sheet is in a state before being completely cured. The sealable resin sheet is formed as a B-stage sheet from the material in A stage by heating in the above-described drying or by heating in the kneading extrusion.

[0079]

The lower limit of the thickness of the sealable resin sheet is, for example, 10  $\mu\text{m}$ , preferably, 25  $\mu\text{m}$ , more preferably, 50  $\mu\text{m}$ . The upper limit of the thickness of the sealable resin sheet is, for example, 3000  $\mu\text{m}$ , preferably, 1000  $\mu\text{m}$ , more preferably, 500  $\mu\text{m}$ , even more preferably, 300  $\mu\text{m}$ .

[0080]

Next, a method of producing an electronic element package 50 by sealing electronic elements as exemplary elements with the sealable resin sheet is described with reference to FIG. 1A to FIG. 2B.

[0081]

In FIG. 2B, the members viewed on the line A-A of FIG. 2A are hatched with lines while the members viewed away from the line A-A are not hatched with lines.

[0082]

In this method, as illustrated in FIG. 1A, first, a sealable resin sheet 1 is prepared. The sealable resin sheet 1 has a one-side surface and the other-side surface in thickness direction, which face each other in the thickness direction.

[0083]

As illustrated in FIG. 1B, FIG. 2A, and FIG. 2B, an electronic element 21 is separately be prepared.

[0084]

As illustrated in FIG. 2A, and FIG. 2B, the electronic element 21 includes an electronic component. The electronic element 21 has an approximately flat board shape (chip shape) extending in the surface direction. The other-side surface 28 in the thickness direction of the electronic element 21 is provided with electrodes (not illustrated). The electrodes are disposed at a peripheral edge of the electronic element 21 with a space therebetween. The electronic element 21 contains, for example, a known electronic material.

[0085]

The electronic element 21 is mounted on a board 22 via bumps 23 so that the two gaps (first gap 26 and second gap 36) varying in thickness are formed. The electronic element 21 and board 22 are included in the element mounting board 24. The element mounting board 24 includes the first gap 26 and second gap 36.

[0086]

The board 22 has an approximately flat plate shape extending in a surface direction. A one-side surface 25 in the thickness direction of the board 22 is, for example, parallel to the other-side surface 28 in the thickness direction of the electronic element 21.

[0087]

A length T2 between the one-side surface 25 in the thickness direction of the board 22 and the other-side surface 28 in the thickness direction of the electronic element 21 corresponds to a thickness T2 of the second gap 36 between the board 22 and electronic element 21. The lower limit of the thickness T2 of the second gap 36 is, for example, 10  $\mu\text{m}$ , preferably, 30  $\mu\text{m}$ . The upper limit of the thickness T2 of the second gap 36 is, for example, 200  $\mu\text{m}$ , preferably, 100  $\mu\text{m}$ .

[0088]

A plurality of terminals 29 electrically connected to the electrodes (not illustrated) of the electronic element 21 is provided on the one-side surface 25 in the thickness direction of the board 22.

[0089]

The terminals 29 extend along the peripheral edge of the electronic element 21 in the plan view. The terminals 29 have a predetermined thickness T0. The terminals 29 are disposed with a space therebetween in the surface direction. The terminals 29 are provided corresponding to the electrodes (not illustrated) of the electronic element 21, respectively.

[0090]

The thickness T0 of the terminal 29 is a length between the one-side surface 25 in the thickness direction of the board 22 and a one-side surface 30 in the thickness direction of the terminal 29. The thickness T0 of the terminal 29 is smaller than the thickness T2 of the second gap 36. Specifically, the lower limit of the thickness T0 of the terminal 29 is, for example, 5

μm, preferably, 15 μm. The upper limit of the thickness T0 of the terminal 29 is, for example, 100 μm, preferably, 50 μm.

[0091]

The terminals 29 are electrically connected to the electrodes (not illustrated) of the electronic element 21 via the bumps 23.

[0092]

The plurality of bumps 23 is provided corresponding to the plurality of terminals 29. The bumps 23 are disposed between the electrodes 29 of the electronic element 21 and the terminals (not illustrated) of the board 22, respectively. Examples of the material of the bumps 23 include metals such as a solder and gold. A thickness T1 of the bump 23 corresponds to a thickness T1 of the first gap 26 described below. The thickness T1 of the bump 23 corresponds to the difference of the subtraction of the thickness T0 of the terminal 29 from the thickness T2 of the second gap 36 ( $T2 - T0$ ) when the one-side surface 25 in the thickness direction of the board 22 is parallel to the other-side surface 28 in the thickness direction of the electronic element 21. Specifically, the lower limit of the thickness T1 of the bump 23 is, for example, 5 μm, preferably, 15 μm. The upper limit of the thickness T1 of the bumps 23 is, for example, 100 μm, preferably, 50 μm.

[0093]

The first gap 26 is a gap (void or hollow) formed by the bumps 23 intervening the electronic element 21 and terminals 29. Thus, the thickness T1 of the first gap 26 equals the thickness T1 of the bumps 23. The first gap 26 is a space between the other-side surface 28 in the thickness direction of the electronic element 21 and the one-side surface 30 in the thickness direction of the terminal 29. The first gap 26 partially overlaps a lateral edge 75 of the electronic element 21 in the plan view. In detail, the first gap 26 is a region excluding the bump 23 in a region where the electronic element 21 overlaps the terminals 29 when being projected in the thickness direction.

[0094]

The second gap 36 has the thickness T2 larger than the thickness T1 of the first gap 26. The second gap 36 communicates with the first gap 26. Specifically, the thickness T2 of the second gap 36 is a total of the thickness T1 of the first gap 26 and the thickness T0 of the

terminal 29. The second gap 36 is a gap (void or hollow) between the other-side surface 28 in the thickness direction of the electronic element 21 and the one-side surface 25 in the thickness direction of the board 22. The second gap 36 is a region excluding the terminals 29 in a region where the electronic element 21 overlaps the board 22 when being projected in the thickness direction. The second gap 36 partially overlaps the lateral edge 75 of the electronic element 21.

[0095]

The lower limit of the ratio of the thickness T1 of the first gap 26 to the thickness T2 of the second gap 36 ( $T1/T2$ ) is, for example, 0.1, preferably, 0.2. The upper limit of the ratio of the thickness T1 of the bump 23 to the thickness T2 of the gap between the board 22 and electronic element 21 ( $T1/T2$ ) is, for example, 0.6, preferably, 0.5.

[0096]

The first gap 26 and second gap 36 overlap the whole of the lateral edge 75 of the electronic element 21.

[0097]

Next, as illustrated in FIG. 1B, the sealable resin sheet 1 is disposed on the electronic element 21. Specifically, the other-side surface in the thickness direction of the sealable resin sheet 1 is brought into contact with a one-side surface in the thickness direction of the electronic element 21.

[0098]

Next, as illustrated in FIG. 1C, the sealable resin sheet 1 and the element mounting board 24 are pressed. Specifically, the sealable resin sheet 1 and element mounting board 24 are heat-pressed.

[0099]

For example, a press 27 having two flat plates presses the sealable resin sheet 1 and element mounting board 24, sandwiching them in the thickness direction. The flat plates of the press 27 include, for example, a heat source not illustrated.

[0100]

The conditions for the press (for example, pressure, time, and temperature) are not especially limited. The conditions allowing the sealable resin sheet 1 to cover the side peripheral surfaces of the electronic element 21 and preventing the element mounting board 24

from being damaged are selected. More specifically, the conditions for the press are set so that the sealable resin sheet 1 can flow the outside of the side peripheral surfaces of the electronic element 21 and cover the side peripheral surfaces of the electronic element 21, and be in contact with both of the one-side surface 25 in the thickness direction of the board 22 and the one-side surfaces 30 in the thickness direction of the terminals 29 that do not overlap the electronic element 21 in the plan view.

[0101]

Specifically, the lower limit of the pressure of the press is, for example, 0.05 MPa, preferably, 0.1 MPa. The upper limit of the pressure of the press is, for example, 10 MPa, preferably, 5 MPa. The lower limit of the time of the press is, for example, 0.3 minutes, preferably, 0.5 minutes. The upper limit of the time of the press is, for example, 10 minutes, preferably, 5 minutes.

[0102]

Specifically, the lower limit of the heating temperature is, for example, 40°C, preferably, 60°C. The upper limit of the heating temperature is, for example, 100°C, preferably, 95°C.

[0103]

When the sealable resin sheet 1 is pressed, the sealable resin sheet 1 plastically deforms corresponding in the outer shape of the electronic element 21. The other-side surface in the thickness direction of the sealable resin sheet 1 deforms into a shape corresponding to the thickness-direction one-side surface and side peripheral surfaces of the electronic element 21, and the one-side surface 25 in the thickness direction of the board 22 that does not overlap the electronic element 21 in the plan view and the one-side surfaces 30 in the thickness direction of the terminals 29.

[0104]

In this manner, the sealable resin sheet 1 blocks the peripheral edges of the first gap 26 and second gap 36. In this manner, the electronic element 21 is sealed with the excellent sealing properties.

[0105]

The sealable resin sheet 1 plastically deforms while maintaining B stage.

[0106]

In this manner, the sealable resin sheet 1 covers the side peripheral surface of each of the electronic elements 21, and is in contact with the one-side surface 25 in the thickness direction of the board 22 that does not overlap the electronic element 21 in the plan view.

[0107]

In this manner, a sealing body 31 sealing the electronic element 21 is formed (produced) from the sealable resin sheet 1. A one-side surface in the thickness direction of the sealing body 31 is a flat surface.

[0108]

At that time, the sealing body 31 is allowed to slightly enter the first gap 26 and second gap 36.

[0109]

Thereafter, as illustrated in FIG. 1D, the sealing body 31 is heated, thereby forming a cured product 41 from the sealing body 31.

[0110]

Specifically, the sealing body 31 and element mounting board 24 are taken out of the press 27. Subsequently, the sealing body 31 and element mounting board 24 are heated under atmospheric pressure in a dryer.

[0111]

The lower limit of the heating temperature (curing temperature) is, for example, 100°C, preferably, 120°C. The upper limit of the heating temperature (curing temperature) is, for example, 200°C, preferably, 180°C. The lower limit of the heating time is, for example, 10 minutes, preferably, 30 minutes. The upper limit of the heating time is, for example, 180 minutes, preferably, 120 minutes.

[0112]

The above-described heating of the sealing body 31 forms the cured product 41 in C stage (completely cured) from the sealing body 31. A one-side surface in the thickness direction of the cured product 41 is an exposed surface.

[0113]

An edge of the sealing body 31 allowed to slightly enter the first gap 26 and second gap 36 is further allowed to slightly enter the inside of the first gap 26 and second gap 36 and become the cured product 41 therein.

[0114]

The sealable resin sheet 1 contains the layered silicate compound, and the layered silicate compound and thermoplastic resin. The ratio ( $A/\alpha$ ) of the Mass A of the layered silicate compound to the mass  $\alpha$  of the thermoplastic resin is 0.3 or more. Thus, when the sealable resin sheet 1 is disposed on a plurality of electronic elements 21 varying in size or design or a plurality of electronic elements 21 connected to the terminals 29 varying in size or design, and heated to form the cured product 41, the entering amount of the cured product 41 that enters the first gap 26 and second gap 36 is controlled. Thus, the electronic elements 21 are uniformly sealed.

[0115]

For example, when the sealable resin sheet 1 is disposed on the electronic element 21 forming the first gap 26 and second gap 36 varying in thickness depending on the board 22 and terminals 29 and the sealable resin sheet 1 (sealing body 31) is heated to form the cured product 41 as illustrated in FIG. 1D, the entering amount of the cured product 41 that enters both of the first gap 26 and second gap 36 can be reduced. Specifically, both of a first cured product entering length Y1 and a second cured product entering length Y2, which are described in detail in Examples (see FIG. 1D and FIG. 3D), can be reduced.

[0116]

On the other hand, the ratio ( $A/\alpha$ ) of the mass A of the layered silicate compound to the mass  $\alpha$  of the thermoplastic resin is 2.0 or less. Thus, the side peripheral surfaces of the electronic element 21 are surely covered and the peripheral edges of the first gap 26 and second gap 36 are surely blocked. In other words, formation of an unfilled portion is suppressed and excellent sealing properties for the electronic element 21 are achieved.

[0117]

When the lower limit of the content ratio B of the layered silicate compound in the sealable resin sheet 1 is 3 mass% or more, both the first cured product entering length Y1 and second cured product entering length Y2 can be reduced. When the lower limit of the content

ratio B of the layered silicate compound is 7 mass% or less, the edges of the first gap 26 and second gap 36 can surely be blocked.

[0118]

When the glass transition temperature  $T_g$  of the thermoplastic resin is  $-30^{\circ}\text{C}$  or more, the layered silicate compound can sufficiently be dispersed in the sealable resin sheet, and thus a sealable resin sheet with a uniform fluidity can be produced. When the glass transition temperature  $T_g$  of the thermoplastic resin is  $-10^{\circ}\text{C}$  or less, the fluidity of the sealable resin sheet can be reduced, and thus the entering amount of the cured product that enters the first gap 26 and second gap 36 can be reduced.

[0119]

When the layered silicate compound has a surface modified by an organic component, the layered silicate compound and the thermosetting resin and thermoplastic resin have an adequate affinity for each other. Thus, both of the first cured product entering length Y1 and the second cured product entering length Y2 can surely be reduced.

[0120]

<Sealable Multi-layer Resin Sheet>

As illustrated in FIG. 3A to FIG. 3D, a sealable multi-layer resin sheet 11, which includes the sealable resin sheet 1 and a second sealable resin sheet 12 sequentially at one side in the thickness direction, seals the electronic element 21. Subsequently, the cured product 41 can be formed.

[0121]

The sealable multi-layer resin sheet 11 includes the sealable resin sheet 1, and the second sealable resin sheet 12 disposed on the whole of the one-side surface in the thickness direction of the sealable resin sheet 1. Preferably, the sealable multi-layer resin sheet 11 includes only the sealable resin sheet 1 and the second sealable resin sheet 12.

[0122]

The material of the second sealable resin sheet 12 is the same as the material of the sealable resin sheet 1, except that the material of the second sealable resin sheet 12 does not contain a layered silicate compound. The lower limit of the ratio of the thickness of the second sealable resin sheet 12 to the thickness of the sealable resin sheet 1 is, for example, 0.5,

preferably, 1, more preferably, 2. The upper limit of the ratio of the thickness of the second sealable resin sheet 12 to the thickness of the sealable resin sheet 1 is, for example, 10, preferably, 5.

[0123]

The lower limit of the thickness of the sealable multi-layer resin sheet 11 is, for example, 50  $\mu\text{m}$ , preferably, 100  $\mu\text{m}$ , more preferably, 150  $\mu\text{m}$ . The upper limit of the thickness of the sealable multi-layer resin sheet 11 is, for example, 3000  $\mu\text{m}$ , preferably, 1000  $\mu\text{m}$ , more preferably, 500  $\mu\text{m}$ , even more preferably, 300  $\mu\text{m}$ .

[0124]

A method of producing an electronic element cured product package 50 by sealing the plurality of electronic elements 21 with the sealable multi-layer resin sheet 11 and subsequently forming the cured product 41 is described with reference to FIG. 3A to FIG. 3D.

[0125]

As illustrated in FIG. 3A, the sealable multi-layer resin sheet 11 is prepared. Specifically, the sealable resin sheet 1 and the second sealable resin sheet 12 are bonded to each other.

[0126]

As illustrated in FIG. 3B, the plurality of electronic elements 21 mounted on the board 22 is prepared.

[0127]

Subsequently, the sealable multi-layer resin sheet 11 is disposed on the electronic elements 21 so that the other-side surface in the thickness direction of the sealable resin sheet 1 is brought into contact with the one-side surfaces in the thickness direction of the electronic elements 21.

[0128]

As illustrated in FIG. 3C, thereafter, the sealable resin sheet 1 and the element mounting board 24 are pressed.

[0129]

The press makes the sealable resin sheet 1 flow and enter between the adjacent electronic elements 21. On the other hand, the second sealable resin sheet 12 does not contain a

layered silicate compound. Thus, when the second sealable resin sheet 12 is pressed, the fluidity of the second sealable resin sheet 12 is not improved and maintained low. Hence, the entry of the second sealable resin sheet 12 between the adjacent electronic elements 21 is suppressed.

[0130]

In this manner, the sealing body 341 sealing the electronic elements 21 is formed from the sealable multi-layer resin sheet 11.

[0131]

Thereafter, as illustrated in FIG. 3D, the sealing body 31 is heated, thereby forming the cured product 41 from the sealing body 31.

[0132]

When the sealable resin sheet 1 contains the inorganic filler in a ratio of the above-described lower limit or more and the second sealable resin sheet 12 contains the inorganic filler in a ratio of the above-described lower limit or more, the press illustrated in FIG. 3C can make both the sealable resin sheet 1 and second sealable resin sheet 12 flow.

[0133]

At that time, the sealable resin sheet 1 is in contact with the electronic elements 21 while the second sealable resin sheet 12 is located at a side opposite to the electronic elements 21 relative to the sealable resin sheet 1. In other words, an edge of the sealing body 31 that faces the gap 26 and second gap 36 is formed from the sealable resin sheet 1. On the other hand, the one-side surface in the thickness direction of the sealing body 31 is formed from the second sealable resin sheet 12.

[0134]

The sealable multi-layer resin sheet 11 includes the above-described sealable resin sheet 1. Thus, the entering amount of the cured product 41 that enters the gap 26 and second gap 36 can be reduced.

[0135]

In particular, when the sealable resin sheet 1 and the second sealable resin sheet 12 contain a main agent of epoxy resin having a softening point of 50°C or more and 130°C or less, the sealable resin sheet 1 and the second sealable resin sheet 12 can flow in the step illustrated in

FIG. 3C. Thus, the time for the step illustrated in FIG. 3C can be shortened, and a one-side surface in the thickness direction of the second sealable resin sheet 12 can be flattened in the step illustrated in FIG. 3C.

[0136]

Furthermore, when the sealable resin sheet 1 and the second sealable resin sheet 12 contain phenol resin as a curing agent together with the main agent of epoxy resin, the cured product 41 has a high heat resistance and a high chemical resistance. Accordingly, the cured product 41 has excellent reliability for sealing.

[0137]

In the step illustrated in FIG. 3C, the second sealable resin sheet 12 receives the pressing force and is fluidized. Thus, the one-side surface in the thickness direction of the second sealable resin sheet 12 is flattened. As described above, in the step illustrated in FIG. 3C, the sealable resin sheet 1 receives the pressing force, is softened and flows together with the second sealable resin sheet 12 in the sealable multi-layer resin sheet 11. Then, the sealable multi-layer resin sheet 11 deforms, following the outer shapes of the electronic elements 21. In the step illustrated in FIG. 3C, the sealable resin sheet 1 is allowed to slightly enter the gap 26 and second gap 36.

[0138]

In the step illustrated in FIG. 3D, the flow of the sealable resin sheet 1 is suppressed based on the reduction in complex viscosity  $\eta^*$  with the rise in temperature. Thus, an excessive entry of the sealable resin sheet 1 to the first gap 26 and second 36 is suppressed. In other words, the cured product 41 that is the cured sealable multi-layer resin sheet 11 including the sealable resin sheet 1 can reduce the cured product entering lengths Y.

[0139]

#### Variations

In each of the variations described below, the same members and steps as the above-described embodiment are given the same numerical references and the descriptions thereof are omitted. Further, each of the variations has the same operations and effects as those of the embodiment unless especially described otherwise. The embodiment and variations can appropriately be combined.

[0140]

As the gaps varying in thickness, the first gap 26 and second gap 36 are exemplified. The gaps varying in thickness may be three or more.

[0141]

The plurality of gaps varying in thickness is formed on one electronic element 21. For example, however, the plurality of gaps varying in thickness may be formed on a plurality of electronic elements 21, respectively. Specifically, the first gap 26 may be formed on one electronic element 21 while the second gap 36 may be formed on another electronic element 21.

[0142]

In the embodiment, the one-layer sealable resin sheet 1 seals the electronic elements 21. Although not illustrated, however, (a laminate sheet of) a plurality of the sealable resin sheets 1 can seal the electronic elements 21.

[0143]

Alternatively, the second sealable resin sheet 12 in the sealable multi-layer resin sheet 11 can have a plurality of layers.

[0144]

As an example of the element, the electronic elements 21 separated from the one-side surface 25 in the thickness direction of the board 22 with the gap 26 therebetween is described. Then, the electronic elements 21 are sealed with the sealable resin sheet 1. However, although not illustrated, for example, electronic elements 21 in contact with the one-side surface 25 in the thickness direction of the board 22 can be used. Then, the electronic elements 21 can be sealed with the sealable resin sheet 1 as well.

[0145]

As an example of the element, the electronic elements 21 are described. However, a semiconductor element can be used as well.

Examples

[0146]

Hereinafter, with reference to Preparation Examples, Comparative Preparation Example, Examples, and Comparative Examples, the present invention is more specifically described. The present invention is not limited to Preparation Examples, Comparative Preparation Example,

Examples, and Comparative Examples in any manner. The specific numeral values used in the description below, such as mixing ratios (contents), physical property values, and parameters can be replaced with the corresponding mixing ratios (contents), physical property values, parameters in the above-described "DESCRIPTION OF EMBODIMENTS", including the upper limit values (numeral values defined with "or less", and "less than") or the lower limit values (numeral values defined with "or more", and "more than").

[0147]

The components used in Preparation Examples and Comparative Preparation Example are described as follows.

[0148]

Layered silicate compound: S-BEN NX manufactured by HOJUN Co., Ltd. (organic bentonite having a surface modified by dimethyl distearyl ammonium)

Main agent: YSLV-80XY manufactured by the new Nippon Steel Chemical Co., Ltd. (bisphenol F type epoxy resin, high-molecular-weight epoxy resin, epoxy equivalent of 200 g/eq., softening point of 80°C)

Curing agent: LVR- 8210DL manufactured by Gunei Chemical Industry Co., Ltd. (novolac-type phenol resin, latent curing agent, hydroxyl group equivalent: 104 g/eq., softening point: 60°C)

Curing accelerator: 2PHZ-PW manufactured by SHIKOKU CHEMICALS CORPORATION (2-phenyl-4,5-dihydroxymethylimidazole)

Acryl resin 1: HME-2006M manufactured by Negami Chemical Industrial Co.,Ltd., a carboxyl group-containing acrylic acid ester copolymer (acrylic polymer), weight-average molecular weight: 600,000, glass-transition temperature (Tg): -30°C, methyl ethyl ketone solution with a 20 mass% solid content concentration

Acryl resin 2: TEISANRESIN SG-70L manufactured by Nagase ChemteX Corporation, a carboxyl group and hydroxy group-containing acrylic acid ester copolymer (acrylic polymer), weight-average molecular weight: 900,000, glass-transition temperature (Tg): -10°C, methyl ethyl ketone solution with a 12.5 mass% solid content concentration

Silane coupling agent: KBM-403 manufactured by Shin-Etsu Chemical Co., Ltd. (3-glycidoxypentyltrimethoxysilane)

First filler: FB-8SM (spherical molten silica powder (inorganic filler), average particle size of 7.0  $\mu\text{m}$ )

Second filler: an inorganic filler made of SC220G-SMJ manufactured by Admatechs Company Limited (average particle size of 0.5  $\mu\text{m}$ ) having a surface finished with 3-methacryloxypropyl trimethoxysilane (manufactured by Shin-Etsu Chemical Co., Ltd. product name: KBM- 503). Inorganic particles having a surface finished with 1 part by mass of a silane coupling agent with respect to 100 parts by mass of the inorganic filler.

[0149]

Carbon black: #20 manufactured by Mitsubishi Chemical Corporation., particle size of 50 nm

[0150]

Preparation Examples 1-6 and Comparative Preparation Examples 1-4

A varnish of the material was prepared in conformity with the blending formula described in Tables 1 to 3. The varnish was applied on a surface of a peeling sheet, and then dried at 120°C for 2 minutes, thereby producing a sealable resin sheet 1 with a thickness of 65  $\mu\text{m}$ . The sealable resin sheet 1 was in B stage.

[0151]

Preparation Example 7

A varnish of the material was prepared in conformity with the blending formula described in Table 4. The varnish was applied on a surface of a peeling sheet, and then dried at 120°C for 2 minutes, thereby producing a second sealable resin sheet 12 with a thickness of 195  $\mu\text{m}$ . The second sealable resin sheet 12 was in B stage.

[0152]

Examples 1 to 6 and Comparative Examples 1 and 2

In each of the combinations of Preparation Examples shown in Table 5, the sealable resin sheet and the second sealable resin sheet were adhered to each other, thereby producing a sealable multi-layer resin sheet with a thickness of 260  $\mu\text{m}$ .

[0153]

Evaluation

Steps A to E were carried out. Then, the first cured product entering length Y1 and second cured product entering length Y2 were measured.

[0154]

Step A: As illustrated in FIG. 3A, a sample sheet 61 with a longitudinal length of 10 mm, a lateral length of 10 mm, and a thickness of 260  $\mu\text{m}$  was prepared from the sealable multi-layer resin sheet 11 of each of Examples and Comparative Examples.

[0155]

Step B: As illustrated in FIG. 3B, a dummy element mounting board 74 was prepared by mounting a dummy element 71 with a longitudinal length of 3 mm, a lateral length of 3 mm, and a thickness of 200  $\mu\text{m}$  on a glass board 72 through bumps 23.

The bumps 23 were mounted on one-side surfaces in the thickness direction of dummy terminals 79 with a thickness T0 of 30  $\mu\text{m}$ , respectively. The dummy element mounting board 74 included a first gap 26 with a thickness T1 of 20  $\mu\text{m}$  and a second gap 36 with a thickness T2 of 50  $\mu\text{m}$ .

[0156]

Step C: As illustrated in FIG. 3C, the sample sheet 61 sealed the dummy element 71 on the dummy element mounting board 74 by vacuum flat-plate press at a temperature of 65°C, a pressure of 0.1 MPa, a degree of vacuum of 1.6 kPa, and for a press time of 1 minute, thereby forming a sealing body 31 from the sample sheet 61.

[0157]

Step D: As illustrated in FIG. 3D, the sealing body 31 was heat-cured by heating at 150°C under atmospheric pressure for 1 hour, thereby forming a cured product 41 from the sealing body 31.

[0158]

Step E: As illustrated in an enlarged drawing of FIG. 3D, the first cured product entering length Y1 in which the cured product 41 entered the first gap 26 from a side edge 75 and the second cured product entering length Y2 in which the cured product 41 entered the second gap 26 from the side edge 75 were measured based on the side edge 75 of the dummy element 71.

[0159]

By the following criteria, the first cured product entering length Y1 and the second cured product entering length Y2 were evaluated. The results are shown in Table 1.

Good: Both of the first cured product entering length Y1 and the second cured product entering length Y2 were 0  $\mu\text{m}$  or more and 20  $\mu\text{m}$  or less.

Bad: Both of the first cured product entering length Y1 and the second cured product entering length Y2 were more than 30  $\mu\text{m}$ , or, less than 0  $\mu\text{m}$ .

[0160]

In the evaluation, the “minus” means that a space (unfilled portion) protruding outward from the side edge 75 of the dummy element 71 was formed (see the thick dashed line in FIG. 3D). The absolute value of the “minus” corresponds to a length of the protruding space.

[0161]

[Table 1]

| Material of sealable resin sheet |                                                                                                |                                                                |              |                    | Preparation<br>Ex. 1 | Preparation<br>Ex. 2 | Comp. Preparation Ex.<br>1 |      |
|----------------------------------|------------------------------------------------------------------------------------------------|----------------------------------------------------------------|--------------|--------------------|----------------------|----------------------|----------------------------|------|
| Formulas of materials            | Layered<br>silicate<br>compound                                                                | Organic bentonite (modified by<br>dimethyl distearyl ammonium) |              | NX S-BEN NX (part) | 0.71                 | 0.71                 | 0.71                       |      |
|                                  | Content ratio B of layered silicate compound in sealable resin sheet                           |                                                                |              | [%]                | 5                    | 5                    | 5                          |      |
|                                  | Parts of layered silicate compound with respect<br>to 100 parts by mass of inorganic filler    |                                                                |              | (part)             | 7                    | 7                    | 7                          |      |
|                                  | Epoxy resin<br>composition                                                                     | Main agent                                                     | Epoxy resin  | YSLV-80XY (part)   | 1.66                 | 1.66                 | 1.91                       |      |
|                                  |                                                                                                | Curing agent                                                   | Phenol resin | LVR-8210DL (part)  | 0.9                  | 0.9                  | 1.04                       |      |
|                                  |                                                                                                | Curing<br>accelerator                                          | Imidazoles   | 2PHZ-PW (part)     | 0.024                | 0.024                | 0.02                       |      |
|                                  | Ratio of main agent in material                                                                |                                                                |              | [%]                | 12                   | 12                   | 13                         |      |
|                                  | Ratio of main agent in epoxy resin composition                                                 |                                                                |              | [%]                | 64                   | 64                   | 64                         |      |
|                                  | Parts of curing agent with respect<br>to 100 parts by mass of main agent                       |                                                                |              | (part)             | 54                   | 54                   | 54                         |      |
|                                  | Parts of curing accelerator with respect<br>to 100 parts by mass of main agent                 |                                                                |              | (part)             | 1.4                  | 1.4                  | 1.3                        |      |
|                                  | Content ratio of epoxy resin composition in material                                           |                                                                |              | [%]                | 18                   | 19                   | 21                         |      |
|                                  | Thermoplastic<br>resin                                                                         | Acrylic resin 1<br>(solid content only)                        |              | HME-2006M (part)   | 0.73                 | –                    | 0.35                       |      |
|                                  |                                                                                                | Acrylic resin 2<br>(solid content only)                        |              | SG-70L (part)      | –                    | 0.73                 | –                          |      |
|                                  | Ratio of thermoplastic resin in material                                                       |                                                                |              | [%]                | 5                    | 5                    | 2                          |      |
|                                  | Ratio of thermoplastic resin in total amount of<br>thermosetting resin and thermoplastic resin |                                                                |              | [%]                | 22                   | 22                   | 10                         |      |
|                                  | Ratio (A/ α) of Mass A of layered silicate compound<br>to mass α of thermoplastic resin        |                                                                |              |                    |                      | 0.97                 | 0.97                       | 2.05 |
|                                  | Silane<br>coupling agent                                                                       | Glycidyl group-containing                                      |              | KBM-403 (part)     | 0.15                 | 0.15                 | 0.15                       |      |
|                                  | Content ratio of silane coupling agent in material                                             |                                                                |              | [%]                | 1                    | 1                    | 1                          |      |
|                                  | Inorganic filler                                                                               | First filler                                                   |              | FB-8SM (part)      | 6.5                  | 6.5                  | 6.50                       |      |
|                                  |                                                                                                | Second filer                                                   |              | SC220G-SMJ (part)  | 3.5                  | 3.5                  | 3.50                       |      |
|                                  | Content ratio of inorganic filler                                                              |                                                                |              | [%]                | 70                   | 70                   | 70                         |      |
|                                  | Content ratio of first filler in material                                                      |                                                                |              | [%]                | 46                   | 48                   | 46                         |      |
|                                  | Parts of second filler with respect<br>to 100 parts by mass of first filler                    |                                                                |              | (part)             | 54                   | 54                   | 54                         |      |
|                                  | Pigment                                                                                        | Carbon black                                                   |              | #20 (part)         | 0.11                 | 0.11                 | 0.11                       |      |
|                                  | Ratio of pigment in material                                                                   |                                                                |              | [%]                | 0.8                  | 0.8                  | 0.8                        |      |

[0162]

[Table 2]

Table 2

| Material of sealable resin sheet |                                                                                             |                                                             |              |             | Preparation<br>Ex. 3 | Preparation<br>Ex. 4 | Comp. Preparation<br>Ex. 2 |      |      |
|----------------------------------|---------------------------------------------------------------------------------------------|-------------------------------------------------------------|--------------|-------------|----------------------|----------------------|----------------------------|------|------|
| Formulas of materials            | Layered silicate compound                                                                   | Organic bentonite (modified by dimethyl distearyl ammonium) |              | NX S-BEN NX | (part)               | 0.43                 | 0.43                       | 0.43 |      |
|                                  | Content ratio B of layered silicate compound in sealable resin sheet                        |                                                             |              |             | [%]                  | 3                    | 3                          | 3    |      |
|                                  | Parts of layered silicate compound with respect to 100 parts by mass of inorganic filler    |                                                             |              |             | (part)               | 4                    | 4                          | 4    |      |
|                                  | Epoxy resin composition                                                                     | Main agent                                                  | Epoxy resin  | YSLV-80XY   | (part)               | 1.8                  | 1.8                        | 1.22 |      |
|                                  |                                                                                             | Curing agent                                                | Phenol resin | LVR-8210DL  | (part)               | 0.98                 | 0.98                       | 0.66 |      |
|                                  |                                                                                             | Curing accelerator                                          | Imidazoles   | 2PHZ-PW     | (part)               | 0.026                | 0.026                      | 0.03 |      |
|                                  | Ratio of main agent in material                                                             |                                                             |              |             | [%]                  | 13                   | 13                         | 9    |      |
|                                  | Ratio of main agent in epoxy resin composition                                              |                                                             |              |             | [%]                  | 64                   | 64                         | 64   |      |
|                                  | Parts of curing agent with respect to 100 parts by mass of main agent                       |                                                             |              |             | (part)               | 54                   | 54                         | 54   |      |
|                                  | Parts of curing accelerator with respect to 100 parts by mass of main agent                 |                                                             |              |             | (part)               | 1.4                  | 1.4                        | 2.1  |      |
|                                  | Content ratio of epoxy resin composition in material                                        |                                                             |              |             | [%]                  | 20                   | 21                         | 13   |      |
|                                  | Thermoplastic resin                                                                         | Acrylic resin 1 (solid content only)                        |              | HME-2006M   | (part)               | 0.79                 | —                          | 1.68 |      |
|                                  |                                                                                             | Acrylic resin 2 (solid content only)                        |              | SG-70L      | (part)               | —                    | 0.79                       | —    |      |
|                                  | Ratio of thermoplastic resin in material                                                    |                                                             |              |             | [%]                  | 6                    | 6                          | 12   |      |
|                                  | Ratio of thermoplastic resin in total amount of thermosetting resin and thermoplastic resin |                                                             |              |             | [%]                  | 22                   | 22                         | 47   |      |
|                                  | Ratio (A/α) of Mass A of layered silicate compound to mass α of thermoplastic resin         |                                                             |              |             |                      |                      | 0.46                       | 0.46 | 0.27 |
|                                  | Silane coupling agent                                                                       | Glycidyl group-containing                                   |              | KBM-403     | (part)               | 0.15                 | 0.15                       | 0.15 |      |
|                                  | Content ratio of silane coupling agent in material                                          |                                                             |              |             | [%]                  | 1                    | 1                          | 1    |      |
|                                  | Inorganic filler                                                                            | First filler                                                |              | FB-8SM      | (part)               | 6.5                  | 6.5                        | 6.50 |      |
|                                  |                                                                                             | Second filler                                               |              | SC220G-SMJ  | (part)               | 3.5                  | 3.5                        | 3.50 |      |
|                                  | Content ratio of inorganic filler                                                           |                                                             |              |             | [%]                  | 70                   | 70                         | 70   |      |
|                                  | Content ratio of first filler in material                                                   |                                                             |              |             | [%]                  | 45                   | 48                         | 46   |      |
|                                  | Parts of second filler with respect to 100 parts by mass of first filler                    |                                                             |              |             | (part)               | 54                   | 54                         | 54   |      |
|                                  | Pigment                                                                                     | Carbon black                                                |              | #20         | (part)               | 0.12                 | 0.12                       | 0.12 |      |
|                                  | Ratio of pigment in material                                                                |                                                             |              |             | [%]                  | 0.8                  | 0.9                        | 0.8  |      |

[0163]

[Table 3]

| Material of sealable resin sheet |                                                                                             |                                                             |              |            | Preparation<br>Ex. 5 | Preparation<br>Ex. 6 |
|----------------------------------|---------------------------------------------------------------------------------------------|-------------------------------------------------------------|--------------|------------|----------------------|----------------------|
| Formulas of materials            | Layered silicate compound                                                                   | Organic bentonite (modified by dimethyl distearyl ammonium) | NX S-BEN NX  | (part)     | 1                    | 1                    |
|                                  | Content ratio B of layered silicate compound in sealable resin sheet                        |                                                             |              | [%]        | 7                    | 7                    |
|                                  | Parts of layered silicate compound with respect to 100 parts by mass of inorganic filler    |                                                             |              | (part)     | 10                   | 10                   |
|                                  | Epoxy resin composition                                                                     | Main agent                                                  | Epoxy resin  | YSLV-80XY  | (part)               | 1.52                 |
|                                  |                                                                                             | Curing agent                                                | Phenol resin | LVR-8210DL | (part)               | 0.83                 |
|                                  |                                                                                             | Curing accelerator                                          | Imidazoles   | 2PHZ-PW    | (part)               | 0.22                 |
|                                  | Ratio of main agent in material                                                             |                                                             |              | [%]        | 10                   | 11                   |
|                                  | Ratio of main agent in epoxy resin composition                                              |                                                             |              | [%]        | 59                   | 59                   |
|                                  | Parts of curing agent with respect to 100 parts by mass of main agent                       |                                                             |              | (part)     | 55                   | 55                   |
|                                  | Parts of curing accelerator with respect to 100 parts by mass of main agent                 |                                                             |              | (part)     | 14.5                 | 14.5                 |
|                                  | Content ratio of epoxy resin composition in material                                        |                                                             |              | [%]        | 18                   | 19                   |
|                                  | Thermoplastic resin                                                                         | Acrylic resin 1 (solid content only)                        | HME-2006M    | (part)     | 0.67                 | —                    |
|                                  |                                                                                             | Acrylic resin 2 (solid content only)                        | SG-70L       | (part)     | —                    | 0.67                 |
|                                  | Ratio of thermoplastic resin in material                                                    |                                                             |              | [%]        | 5                    | 5                    |
|                                  | Ratio of thermoplastic resin in total amount of thermosetting resin and thermoplastic resin |                                                             |              | [%]        | 21                   | 21                   |
|                                  | Ratio (A/α) of Mass A of layered silicate compound to mass α of thermoplastic resin         |                                                             |              |            | 1.49                 | 1.49                 |
|                                  | Silane coupling agent                                                                       | Glycidyl group-containing                                   | KBM-403      | (part)     | 0.15                 | 0.15                 |
|                                  | Content ratio of silane coupling agent in material                                          |                                                             |              | [%]        | 1                    | 1                    |
|                                  | Inorganic filler                                                                            | First filler                                                | FB-8SM       | (part)     | 6.5                  | 6.5                  |
|                                  |                                                                                             | Second filler                                               | SC220G-SMJ   | (part)     | 3.5                  | 3.5                  |
|                                  | Content ratio of inorganic filler                                                           |                                                             |              | [%]        | 69                   | 69                   |
|                                  | Content ratio of first filler in material                                                   |                                                             |              | [%]        | 45                   | 47                   |
|                                  | Parts of second filler with respect to 100 parts by mass of first filler                    |                                                             |              | (part)     | 54                   | 54                   |
|                                  | Pigment                                                                                     | Carbon black                                                | #20          | (part)     | 0.1                  | 0.1                  |
|                                  | Ratio of pigment in material                                                                |                                                             |              | [%]        | 0.7                  | 0.7                  |

[0164]

[Table 4]

| Material of second sealable resin sheet                                  |                                                                             |                                      |              |            | Preparation Ex. 7 |      |
|--------------------------------------------------------------------------|-----------------------------------------------------------------------------|--------------------------------------|--------------|------------|-------------------|------|
| Formulas of materials                                                    | Epoxy resin composition                                                     | Main agent                           | Epoxy resin  | YSLV-80XY  | (part)            |      |
|                                                                          |                                                                             | Curing agent                         | Phenol resin | LVR-8210DL | (part)            |      |
|                                                                          |                                                                             | Curing accelerator                   | Imidazoles   | 2PHZ-PW    | (part)            |      |
|                                                                          | Ratio of main agent in material                                             |                                      |              |            | [%]               | 7    |
|                                                                          | Ratio of main agent in epoxy resin composition                              |                                      |              |            | [%]               | 64   |
|                                                                          | Parts of curing agent with respect to 100 parts by mass of main agent       |                                      |              |            | (part)            | 54   |
|                                                                          | Parts of curing accelerator with respect to 100 parts by mass of main agent |                                      |              |            | (part)            | 1.1  |
|                                                                          | Content ratio of epoxy resin composition in material                        |                                      |              |            | [%]               | 11   |
|                                                                          | Thermoplastic resin                                                         | Acrylic polymer (solid content only) |              | HME-2006M  | (part)            | 0.17 |
|                                                                          | Ratio of thermoplastic resin in material                                    |                                      |              |            | [%]               | 1    |
|                                                                          | Silane coupling agent                                                       | Glycidyl group-containing            |              | KBM-403    | (part)            | 0.35 |
|                                                                          | Content ratio of silane coupling agent in material                          |                                      |              |            | [%]               | 1    |
|                                                                          | Inorganic filler                                                            | First filler                         | FB-8SM       | (part)     | 15.15             |      |
|                                                                          |                                                                             | Second filler                        | SC220G-SMJ   | (part)     | 8.16              |      |
|                                                                          | Content ratio of inorganic filler                                           |                                      |              |            | [%]               | 87   |
|                                                                          | Content ratio of first filler in material                                   |                                      |              |            | [%]               | 57   |
| Parts of second filler with respect to 100 parts by mass of first filler |                                                                             |                                      |              | (part)     | 54                |      |
| Pigment                                                                  | Carbon black                                                                |                                      | #20          | (part)     | 0.1               |      |
| Ratio of pigment in material                                             |                                                                             |                                      |              | [%]        | 0.4               |      |

[0165]

[Table 5]

|                                                                          |                       |                       |                                   |                       |                       |                                   |                       |                       |
|--------------------------------------------------------------------------|-----------------------|-----------------------|-----------------------------------|-----------------------|-----------------------|-----------------------------------|-----------------------|-----------------------|
| Sealable multi-layer resin sheet                                         | Example 1             | Example 2             | Comparative Example 1             | Example 3             | Example 4             | Comparative Example 2             | Example 5             | Example 6             |
| Second sealable resin sheet                                              | Preparation Example 7 | Preparation Example 7 | Preparation Example 7             | Preparation Example 7 | Preparation Example 7 | Preparation Example 7             | Preparation Example 7 | Preparation Example 7 |
| Sealable resin sheet                                                     | Preparation Example 1 | Preparation Example 2 | Comparative Preparation Example 1 | Preparation Example 3 | Preparation Example 4 | Comparative Preparation Example 2 | Preparation Example 5 | Preparation Example 6 |
| Entry of cured product into first gap (thickness T1: 20 $\mu\text{m}$ )  | Good                  | Good                  | Bad                               | Good                  | Good                  | Bad                               | Good                  | Good                  |
| Entry of cured product into second gap (thickness T2: 50 $\mu\text{m}$ ) | Good                  | Good                  | Bad                               | Good                  | Good                  | Bad                               | Good                  | Good                  |
| Evaluation of entering amount (20 $\mu\text{m}$ )                        | 8                     | 2                     | 104                               | 23                    | 20                    | -97                               | 5                     | 2                     |
| Entry to gaps varying in thickness (50 $\mu\text{m}$ )                   | 12                    | 10                    | 163                               | 49                    | 41                    | -76                               | 9                     | 6                     |

[0166]

While the illustrative embodiments of the present invention are provided in the above description, such is for illustrative purpose only and it is not to be construed as limiting in any manner. Modification and variation of the present invention that will be obvious to those skilled in the art is to be covered by the following claims.

#### Industrial Applicability

[0167]

The sealable resin sheet is used to seal an element.

#### Description of Reference Numerals

[0168]

- 1 sealable resin sheet
- 21 electronic element 21

## Claims

1. A sealable resin sheet for sealing an element, the sealable resin sheet containing thermosetting resin, a layered silicate compound, and thermoplastic resin,  
5            wherein a ratio ( $A/\alpha$ ) of a mass A of the layered silicate compound to a mass  $\alpha$  of the thermoplastic resin is 0.3 or more and 2.0 or less,  
             wherein an upper limit of a ratio of the thermoplastic resin to a total amount of the thermoplastic resin and the thermosetting resin is 40 mass%, and  
             wherein an upper limit of a glass transition temperature Tg of the thermoplastic  
10 resin is -10°C, the glass transition temperature Tg being a theoretical value obtained by the Fox equation.
2. The sealable resin sheet according to Claim 1, wherein a content ratio B of the layered  
silicate compound is 3 mass% or more and 7 mass% or less.
- 15 3. The sealable resin sheet according to Claim 1, wherein the glass transition temperature Tg of the thermoplastic resin is -30°C or more.
4. The sealable resin sheet according to Claim 2, wherein the glass transition  
20 temperature Tg of the thermoplastic resin is -30°C or more.
5. The sealable resin sheet according to Claim 1, wherein the layered silicate compound has a surface modified by an organic component.
- 25 6. The sealable resin sheet according to Claim 2, wherein the layered silicate compound has a surface modified by an organic component.
7. The sealable resin sheet according to Claim 3, wherein the layered silicate compound has a surface modified by an organic component.

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8. The sealable resin sheet according to Claim 4, wherein the layered silicate compound has a surface modified by an organic component.