

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 mounted on a board with a sealable sheet containing thermosetting resin and spherical silica particles, and thereafter curing the thermosetting resin by heating (for example, see Patent document 1 below).

Citation List

Patent Document

[0003]

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

SUMMARY OF THE INVENTION

PROBLEM TO BE SOLVED BY THE INVENTION

[0004]

As electronic devices have been getting more sophisticated in functionality, there is a need to downsize the semiconductor elements or electronic components included therein. With the need, there is another need to increase the dimensional accuracy at the curing of the resin (cured product) that protects the semiconductor elements or electronic components.

[0005]

Specifically, there is a need to reduce an entering amount of the cured product that enters between the semiconductor element or electronic component and the board from a lateral edge of the semiconductor element or electronic component. 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, all the elements may not uniformly be sealed.

[0006]

However, the sealable sheet described in Patent Document 1 includes the same spherical silica particles. The same spherical silica particles tend to smoothly roll with the flow of the thermosetting resin at the thermal curing of the thermosetting resin. Thus, such a sealable sheet cannot meet the above-described needs.

[0007]

The present invention provides a sealable resin sheet that can reduce an entering amount of a cured product that enters between an element and the board.

MEANS FOR SOLVING THE PROBLEM

[0008]

The present invention [1] includes A sealable resin sheet for sealing an element, the sealable resin sheet containing: a matrix formed from thermosetting resin; first domains formed from a non-layered filler and dispersed in the matrix; and second domains formed from a layered filler and dispersed in the matrix, wherein the sealable resin sheet includes a codispersion region in which the second domain is located among the first domains.

[0009]

The present invention [2] includes the sealable resin sheet described in [1], wherein an average value a of maximum lengths A of the second domain and an average value b of maximum lengths B of the first domain are obtained by first to fourth steps described below, and satisfy a formula (1) described below.

[0010]

$$0.2 < a/b < 5 \quad (1)$$

The first step of obtaining one field-of-view image by a TEM observation of the sealable resin sheet

The second step of observing the one field-of-view image by ten observers to measure the maximum lengths A of the largest second domain among the second domains and the maximum lengths B of the largest first domain among the first domains

The third step of obtaining the average value a of the maximum lengths A measured by the ten observers, and the average value b of the maximum lengths B measured by the ten observers

The fourth step of substituting the average value a and the average value b for the formula (1)

[0011]

The present invention [3] includes the sealable resin sheet described in [2] above, wherein the average value a of the maximum lengths A of the second domain is larger than the average value b of the maximum lengths B of the first domain.

[0012]

The present invention [4] includes the sealable resin sheet described in [2] or [3] above, wherein the average value a of the maximum lengths A of the second domain is 1 μm or more and 10 μm or less.

[0013]

The present invention [5] includes the sealable resin sheet described any one of the above-described [1] to [4], wherein one field-of-view image is obtained by a TEM observation of the sealable resin sheet, and the second domain recognized in the one field-of-view image has an aspect ratio of 2 or more.

[0014]

The present invention [6] includes the sealable resin sheet described any one of the above-described [1] to [5], wherein one field-of-view image is obtained by a TEM observation of the sealable resin sheet, and the first domains recognized in the one field-of-view image have an approximately circular shape.

EFFECTS OF THE INVENTION

[0015]

The sealable resin sheet of the present invention includes the first domains formed from the non-layered filler and the second domains formed from the layered filler, and codispersion regions in which the second domains are located among the first domains. Thus, even when the thermosetting resin is temporarily softened by heating and thus the matrix easily flows, the codispersion regions can suppress the flow of the material of the sealable resin sheet as much as possible. Accordingly, the entering amount of the cured product between the electronic element and board can be reduced when the cured product is formed.

[0016]

Moreover, when the sealable resin sheet of the present invention is disposed on a plurality of elements varying in size or design or a plurality of elements connected to terminals varying in size or design and the sealable resin sheet is heated to form a cured product, the

entering amount of the cured product that enters the gaps is controlled. Thus, the plurality of the elements can uniformly be sealed.

BRIEF DESCRIPTION OF THE DRAWINGS

[0017]

[FIG. 1] FIG. 1 is a view of a processed image of a TEM picture of an embodiment of the sealable resin sheet of the present invention. The lower image is one field-of-view image. The upper image is an enlarged view of the region surrounded by the square frame in the lower image.

[FIG. 2] FIG. 2 is a view of a processed image of the one field-of-view image of FIG. 1, illustrating the first domains by white lines and the second domains by black lines.

[FIG. 3] FIG. 3A to FIG. 3D 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. 3A depicts the step of preparing the sealable resin sheet. FIG. 3B depicts the step of preparing the electronic elements. FIG. 3C depicts the step of forming a sealing body by pressing the sealable resin sheet. FIG. 3D depicts the step of forming a cured product by heating the sealing body.

[FIG. 4] FIG. 4A to FIG. 4D are cross-sectional views illustrating the step of producing an electronic element package by sealing a plurality of electronic elements with a sealing multi-layer resin sheet including the sealable resin sheet and second sealable resin sheet illustrated in FIG. 3A. FIG. 4A illustrates the step of preparing the sealing multi-layer resin sheet. FIG. 4B illustrates the step of preparing the electronic elements. FIG. 4C illustrates the step of forming a sealing body by pressing the sealing multi-layer resin sheet. FIG. 4D illustrates the step of forming a cured product by heating the sealing body.

[FIG. 5] FIG. 5A to FIG. 5D illustrate a method of measuring a cured product entering length of Examples. FIG. 5A illustrates the step of preparing a sealing multi-layer resin sheet (step A). FIG. 5B illustrates the step of preparing an electronic element (dummy element (step B)). FIG. 5C illustrates the step of forming a sealing body by pressing the sealing multi-layer resin sheet (step C). FIG. 5D illustrates the step of forming a cured product by heating the sealing body (step D).

DESCRIPTION OF THE EMBODIMENTS

[0018]

An embodiment of the sealable resin sheet of the present invention is described with appropriate reference to FIG. 1 and FIG. 2.

[0019]

FIG. 1 and FIG. 2 are views of processed images of a TEM picture of the sealable resin sheet of Example 3 described below.

[0020]

A sealable resin sheet 1 is a resin sheet used to seal an element and has an approximately plate shape (film shape) (see FIG. 3A) extending in a surface direction orthogonal to a thickness direction.

[0021]

As illustrated in FIG. 1, the sealable resin sheet 1 includes a matrix region 2 and codispersion regions 3. In detail, the sealable resin sheet 1 includes the matrix region 2 and the codispersion regions 3 in the one field-of-view image (described below) obtained by a TEM observation of the sealable resin sheet 1.

[0022]

<Matrix Region>

The matrix region 2 is the region other than the codispersion regions 3 in the sealable resin sheet 1. The matrix region 2 consists of a matrix 4 for the codispersion regions 3.

[0023]

The matrix 4 is a resin matrix for making up the sealable resin sheet 1.

[0024]

The matrix 4 is formed from thermosetting resin.

[0025]

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.

[0026]

As the thermosetting resin, preferably epoxy resin is used. The epoxy resin is described in detail in the description of a preparation method below.

[0027]

The thermosetting resin may be in a state before completely being cured (including B stage or A stage) or in a state after completely being cured (C stage). The thermosetting resin, however, is preferably in a state after completely being cured (C stage) in a TEM observation described below.

[0028]

<Codispersion regions>

The codispersion regions 3 are particle domains dispersed in the matrix 4.

[0029]

The codispersion regions 3 include first domains 5 and second domains 6 and the first domains 5 have a shape different from that of the second domains 6. The first domains 5 and second domains 6 are dispersed (scattered) together in the codispersion regions 3.

[0030]

<First Domains>

The first domains 5 are cured-product-reinforcement regions to enhance the strength of the cured product when the cured product is obtained by pressing the sealable resin sheet 1 to form a sealing body and subsequently thermally curing the sealing body.

[0031]

The first domains 5 are formed from a non-layered filler.

[0032]

The shape of the non-layered filler is not especially limited as long as the shape is other than that of the layered filler of the second domains 6 described next. When being recognized in the one field-of-view image (described below) obtained by the TEM observation of the sealable resin sheet 1, the non-layered filler has, for example, an approximately circular shape (see FIG. 1) (including an approximate ellipse), or a multangular shape such as an approximate square, or an approximate triangle (however, excluding an approximate flat rectangle).

[0033]

When being recognized in one field-of-view image, the non-layered filler preferably has an approximately circular shape as illustrated in FIG. 1. When the non-layered filler recognized in the one field-of-view image has an approximately circular shape, the non-layered

filler has an excellent dispersibility and can uniformly suppress the flow of the sealable resin sheet.

[0034]

An average value b of maximum lengths B of the first domain 5 in the one field-of-view image is described in detail below.

[0035]

Specific examples of the material of the non-layered filler include: silicate compounds (silicon compounds) such as quartz (silicic acid), and silica (anhydrous silicic acid) (however, excluding the silicate salt compounds described below); and aluminum compounds such as aluminum oxide (alumina), and aluminum nitride. These can be used alone or in combination of two or more. Preferably, a silicate compound, more preferably, silica is used as the material of the non-layered filler. In other words, preferably, a silicate compound filler, more preferably, a silica filler is used as the material of the non-layered filler.

[0036]

<Second domains>

The second domains 6 are flow adjustment regions when a sealing body and a cured product (describe below) are formed from the sealable resin sheet 1. Specifically, the second domains 6 are flow reduction regions at the curing used to reduce the fluidity of the cured product when the sealable resin sheet 1 is heated to form the cured product. In detail, when the sealable resin sheet 1 is pressed to form a sealing body and the sealing body is subsequently thermally cured, the second domains 6 work as flow-improving-at-sealing-and-reducing-at-curing regions in order to first increase the fluidity of the sealing body and thereafter reduce it by heating.

[0037]

The second domains 6 are formed from a layered filler.

[0038]

The layered filler is, for example, a filler having a structure (three-dimensional structure) in which layers each two-dimensionally extending (in the surface direction) are laminated in the thickness direction. The layered filler has a shape in which a plurality of thin layers 7 is laminated in the thickness direction.

[0039]

Specifically, as illustrated in the upper image of FIG. 1, when one field-of-view image (described below) is obtained by a TEM observation of the sealable resin sheet 1, examples of the shape of the layered filler recognized in the one field-of-view image include a flat rectangular shape (board shape) (including a shape having an arc part on its peripheral edge).

[0040]

The average value a of the maximum lengths A of the second domain 6 in the one field-of-view image (see FIG. 2) is described in detail below.

[0041]

Specifically, examples of the material of the layered filler include silicate salt compounds, boron compounds such as boron nitride, and carbon-based materials such as carbon (graphite). Preferably, a silicate salt compound is used.

[0042]

The silicate salt compound is referred to as phyllosilicate.

[0043]

Specifically, examples of the silicate salt 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 silicate salt compound. More preferably, montmorillonite is used.

[0044]

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

[0045]

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

[0046]

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.

[0047]

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.

[0048]

<One Field-of-view Image obtained by TEM Observation of Sealable Resin Sheet 1>

The matrix region 2 and codispersion regions 3 (additionally, the matrix 4, first domains 5, and second domains 6, which make up the matrix region 2 and codispersion regions 3) in the sealable resin sheet 1 are observed in one field-of-view image (FIG. 1 and FIG. 2) obtained by a TEM (transmission electron microscope) observation of the sealable resin sheet 1.

[0049]

A method for making a sample for the TEM observation and the conditions for the TEM are described in Examples below.

[0050]

The matrix region 2 observed in the one field-of-view image is a continuous region (continuous phase). The matrix 4 is formed as a homogeneous region in the one field-of-view image. Thus, in the above-described one field-of-view image, the matrix 4 is recognized as a uniform gray region (substantially without contrast in color).

[0051]

The codispersion regions 3 observed in the one field-of-view image are dispersed (scattered) in the matrix region 2.

[0052]

The first domains 5 and the second domains 6 are dispersed together in the codispersion regions 3.

[0053]

The first domains 5 include a solo domain 8 that solely exists and/or a chain domain 9 formed from the solo domains 8 coupled to each other in one direction. The chain domain 9 may partially branch.

[0054]

Preferably, the first domains 5 include both the solo domain 8 and the chain domain 9.

[0055]

In the one field-of-view image, the lower limit of the ratio of the chain domains 9 to the total of the solo domains 8 and the chain domains 9 in number is, for example, 0.01, preferably, 0.1. The upper limit thereof is, for example, 0.8, preferably, 0.7.

[0056]

As for the number of the chain domains 9, when adjacent first domains 5 are coupled, the coupled first domains 5 are counted as one chain domain 9.

[0057]

The second domains 6 are located among the adjacent first domains 5 in the codispersion regions 3. Specifically, for example, the second domains 6 include a second domain 6 located between the adjacent solo domains 8, a second domain 6 between the adjacent solo domain 8 and chain domain 9, and a second domain 6 between the adjacent chain domains 9. Among them, the second domains 6 between the chain domains 9 exist at the highest rate (for example, in an area ratio).

[0058]

When it is difficult to detect the presence of the second domains 6 in the one field-of-view image, the presence of the layered filler is detected by enlarging only the above-described part as illustrated in the upper image of FIG. 1 and determining the presence of the thin layers 7.

[0059]

The average value a of the maximum lengths A of the second domain 6 and average value b of the maximum lengths B of the first domain 5 obtained by the following first to fourth steps satisfy the following formula (1).

[0060]

$$0.2 < a/b < 5 \quad (1)$$

The first step: one field-of-view image is obtained by a TEM observation of the sealable resin sheet 1.

[0061]

The second step: ten observers observe the one field-of-view image to measure the maximum length A of the largest second domain among the second domains and the maximum length B of the largest first domain among the first domains.

[0062]

The third step: the average value a of the maximum lengths A measured by the ten observers and the average value b of the maximum lengths B measured by the ten observers are obtained.

[0063]

The fourth step: the average value a and the average value b are substituted for the formula (1).

[0064]

The one field-of-view image obtained by the TEM observation in the first step is illustrated in FIG. 1 and FIG. 2.

[0065]

In the second step, each of the ten observers measure the maximum length A of the largest second domain among the second domains 6 as illustrated in FIG. 2. In consideration of the slight difference depending on the observer, the measurement of the maximum length A of the second domain 6 is carried out by the ten observers. When the presence of the second domains 6 is difficult to detect as described above, the maximum lengths A may be different from each other depending on the observer. Accordingly, ten observers measure the maximum

length and find their average value a to reduce the difference (error) and increase the measurement accuracy.

[0066]

Specifically, a maximum length A_1 of the second domain 6 measured by the first observer, a maximum length A_2 (that may differ from A_1) of the second domain 6 measured by the second observer, ... and a maximum length A_{10} (that may differ from A_1 and A_2) of the second domain 6 measured by the tenth observer are obtained.

[0067]

The average value b of the maximum lengths B of the first domain 5 is obtained in the same manner as described above. The maximum length B of the first domain 5 corresponds to the maximum length B of the solo domain 8. When a chain domain 9 includes the maximum length B , the maximum length B of the solo domain 8 making up the chain domain 9 is regarded as the maximum length B of the first domain 5.

[0068]

In the third step, the average value a is obtained as the calculation result of $(A_1 + A_2 + \dots + A_{10})/10$. The average value b is obtained in the same manner as described above.

[0069]

When the first domains 5 include a first non-layered filler and a second non-layered filler (described below) and the first non-layered filler has a different median size from that of the second non-layered filler, the average value b of the maximum lengths B of the first domain 5 depends on the median size of the first non-layered filler larger than the second non-layered filler in size.

[0070]

When the above-described formula (1) is satisfied, the cured product surely covers the peripheral side surface of the element and maintains its excellent sealability while the entering amount of the cured product between the element and board can be reduced.

[0071]

When the solution of the formula (1) is less than the lower limit, the layered filler is excessively dispersed with respect to the non-layered filler in size. Thus, the flow of the resin may not be suppressed as much as possible. When the solution of the formula (1) is more than

the upper limit, the layered filler is insufficiently dispersed with respect to the non-layered filler in size in the resin sheet. Thus, the cured product may not surely cover the peripheral side surface of the element uniformly.

[0072]

Preferably, the average value a of the maximum lengths A of the second domain is larger than the average value b of the maximum lengths B of the first domain. This increases the contribution of the second domain including the layered filler to the flow of the resin, the entering amount of the cured product between the element and board can further be reduced.

[0073]

In other words, the average value a of the maximum lengths A of the second domain and the average value b of the maximum lengths B of the first domain preferably satisfy the following formula (2). The average value a and the average value b satisfy, more preferably, the following formula (3), even more preferably, the following formula (4).

[0074]

$$1 < a/b \quad (2)$$

$$1.3 < a/b \quad (3)$$

$$1.5 < a/b \quad (4)$$

The average value a of the maximum lengths A of the second domain and the average value b of the maximum lengths B of the first domain satisfy, preferably, the following formula (5), more preferably, the following formula (6).

[0075]

$$a/b < 3 \quad (5)$$

$$a/b < 2 \quad (6)$$

Specifically, the lower limit of the average value a of the maximum lengths A of the second domain 6 is, for example, $0.1 \mu\text{m}$, preferably, $1 \mu\text{m}$. The upper limit of the average value a of the maximum lengths A of the second domain 6 is, for example, $30 \mu\text{m}$, preferably, $10 \mu\text{m}$. When the average value a of the maximum lengths A of the second domain 6 is less than the above-described lower limit, the layered filler is excessively dispersed. Thus, the flow may not be suppressed as much as possible. When the average value a of the maximum lengths A of the second domain 6 is more than the above-described upper limit, the layered filler is

insufficiently dispersed in the resin sheet. Thus, the cured product may not surely cover the peripheral side surface of the element uniformly.

[0076]

The lower limit of the aspect ratio of the second domain 6 (layered filler) recognized in the one field-of-view image (described below) is, for example, 1.5, preferably, 2, more preferably, 3. The upper limit of the aspect ratio of the second domain 6 is, for example, 100 or less.

[0077]

The aspect ratio of the second domain 6 is the average value x of the maximum lengths (widths) X in a direction (width direction) orthogonal to the maximum lengths A . The average value x is obtained in conformity with the above-described method of measuring the average value a .

[0078]

Where the aspect ratio of the second domain 6 is the above-described lower limit or more, the second domains of the layered filler can further suppress the flow of the first domains of the non-layered filler when the resin flows. Thus, the flow of the material of the sealable resin sheet can be suppressed as much as possible.

[0079]

In the observation of the one field-of-view image, the lower limit of the area ratio of the matrix region 2 in the sealable resin sheet 1 is, for example, 5area%, preferably, 10area%. The upper limit of the area ratio of the matrix region 2 in the sealable resin sheet 1 is, for example, 50area%, preferably, 40area%.

[0080]

In the observation of the one field-of-view image, the lower limit of the area ratio of the codispersion regions 3 in the sealable resin sheet 1 is, for example, 50area%, preferably, 60area%, more preferably, 70area%. The upper limit of the area ratio of the codispersion regions 3 in the sealable resin sheet 1 is, for example, 95area%, preferably, 90area%, more preferably, 85area%.

[0081]

In the one field-of-view image, the lower limit of the ratio of the second domains 6 to the total of the first domains 5 and the second domains 6 in number is, for example, 1%, preferably, 5%. The upper limit of the ratio of the second domains 6 to the total of the first domains 5 and the second domains 6 in number is, for example, 50%, preferably, 30%.

[0082]

In the one field-of-view image, the lower limit of the ratio of the second domains 6 to the first domains 5 in number is, for example, 0.01, preferably, 0.05. The upper limit of the ratio of the second domains 6 to the first domains 5 in number is, for example, 1, preferably, 0.5.

[0083]

When the above-described ratios fall within the above-described ranges, respectively, the flow of the material of the sealable resin sheet can be suppressed as much as possible. Thus, the entering amount of the cured product between the element and board can be reduced.

[0084]

<Production of Sealable Resin Sheet>

To produce the sealable resin sheet 1, the above-described thermosetting resin, non-layered filler, and layered filler are blended, thereby preparing a mixture (composition).

[0085]

The epoxy resin that is a preferred example of the thermosetting resin is prepared as an epoxy resin composition containing a main agent, a curing agent, and a curing accelerator.

[0086]

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 main agents 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.

[0087]

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..

[0088]

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.

[0089]

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. 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 sealable resin sheet 1 can be flattened in the step illustrated in FIG. 3C.

[0090]

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 high heat resistance and high chemical resistance. Accordingly, the cured product has excellent reliability for sealing.

[0091]

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.

[0092]

The lower limit of the content ratio of the thermosetting resin composition (preferably, the epoxy resin composition) in the mixture (solid content) (corresponding to the sealable resin sheet 1) is, for example, 5 mass%, preferably, 15 mass%, more preferably, 17 mass%. The upper limit of the content ratio of the thermosetting resin composition (preferably, the epoxy resin composition) in the mixture (solid content) (corresponding to the sealable resin sheet 1) is,

for example, 30 mass%, preferably, 25 mass%, more preferably, 20 mass%, even more preferably, 18 mass%.

[0093]

The lower limit of the ratio of the main agent in the mixture is, for example, 1 mass%, preferably, 3 mass%, more preferably, 10 mass%. The upper limit of the ratio of the main agent in the mixture is, for example, 30 mass%, preferably, 15 mass%, more preferably, 12.5 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%.

[0094]

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.

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.

[0095]

Example of the shape of the non-layered filler particles include shapes (other than a layered shape) such as an approximately spherical shape, and an approximately aciculate shape. Preferably, an approximately spherical shape is used.

[0096]

The lower limit of the content ratio of the non-layered filler in the mixture 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 non-layered filler in the mixture is, for example, 90 mass%, preferably, 85 mass%, more preferably, 80 mass%, even more preferably, 75 mass%.

[0097]

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

[0098]

The non-layered filler can include the first non-layered filler, and the second non-layered filler having a median size smaller than the median size of the first non-layered filler.

[0099]

The lower limit of the median size of the first non-layered filler is, for example, 1 μm , preferably, 3 μm . The upper limit of the median size of the first non-layered filler is, for example, 50 μm , preferably, 30 μm .

[0100]

The upper limit of the median size of the second non-layered filler is, for example, 0.9 μm , preferably, 0.8 μm . The lower limit of the median size of the second non-layered filler is, for example, 0.01 μm , preferably, 0.1 μm .

[0101]

The lower limit of the ratio of the median size of the first non-layered filler to the median size of the second non-layered filler is, for example, 2, preferably, 5. The upper limit of the ratio of the median size of the first non-layered filler to the median size of the second non-layered filler is, for example, 50, preferably, 20.

[0102]

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

[0103]

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

[0104]

When the non-layered filler includes the first non-layered filler and second non-layered filler, the lower limit of the content ratio of the first non-layered filler in the mixture is, for example, 30 mass%, preferably, 40 mass%. The upper limit of the content ratio of the first non-layered filler in the mixture is, for example, 60 mass%, preferably, 50 mass%. The lower limit of the contained amount of the second non-layered filler to 100 parts by mass of the first non-layered 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 non-layered filler to 100 parts by mass of the first non-layered filler is, for example, 70 parts by mass, preferably, 60 parts by mass, more preferably, 55 parts by mass.

[0105]

The layered filler has a layered shape such as a plate shape, or a thin slice shape. The layered filler has cleavage properties.

[0106]

The lower limit of the content ratio of the layered filler in the mixture is, for example, 1mass%, preferably, 2mass%, more preferably, 3mass%, even more preferably, 3.5mass%, particularly preferably, 4mass%, most preferably, 4.5mass%. The upper limit of the content ratio of the layered filler in the mixture is, for example, 25mass%, preferably, 15mass%, more preferably, 10mass%, even more preferably, 9mass%, particularly preferably, 8mass%, most preferably, 7mass%.

[0107]

The lower limit of the amount of the layered filler with respect to 100 parts by mass of the non-layered 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 amount of the layered filler with respect to 100 parts by mass of the non-layered 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, particularly preferably, 8 parts by mass.

[0108]

The lower limit of the median size of the layered filler is, for example, 1 μm , preferably, 5 μm , more preferably, 10 μm . The upper limit of the median size of the layered filler is, for example, 100 μm , preferably, 50 μm , more preferably, 10 μm .

[0109]

The median size of the layered filler 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.

[0110]

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

[0111]

Another additive agent such as thermoplastic resin, a pigment, or a silane coupling agent can be added to the mixture.

[0112]

Among the above-described additives, the thermoplastic resin and the silane coupling agent can make up the matrix 4 together with the thermosetting resin. The pigment can make up the first domains 5 together with the non-layered filler.

[0113]

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.

[0114]

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

[0115]

Examples of the acrylic resin include a carboxyl group-containing (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).

[0116]

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.

[0117]

Examples of the other monomer include carboxyl group-containing monomers such as acrylic acid, methacrylic acid, carboxyethyl acrylate, carboxypentyl acrylate, itaconic acid, maleic acid, fumaric acid, and crotonic acid.

[0118]

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, 1,000,000, preferably 900,000. The weight average molecular weight is calibrated with standard polystyrene based on gel permeation chromatography (GPC).

[0119]

The ratio (solid content ratio) of the thermoplastic resin is adjusted so as not to block the thermal curing of the thermosetting resin. Specifically, the lower limit of the ratio (solid content ratio) of the thermoplastic resin in the mixture is, for example, 1 mass% or more, preferably 2 mass% or more. The upper limit of the ratio (solid content ratio) of the thermoplastic resin in the mixture is, for example, 10 mass%, preferably, 5 mass%.

[0120]

The thermoplastic resin may be prepared by being diluted with an appropriate solvent.

[0121]

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 μm . The upper limit of the particle size of the pigment is, for example, 1 μm . The lower limit of the ratio of the pigment to the mixture is, for example, 0.1 mass%. The upper limit thereof is, for example, 2 mass%.

The particle size of the pigment is an arithmetic average particle size obtained by observing the pigment with an electron microscope.

[0122]

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 mixture is, for example, 0.1 mass%, preferably, 1 mass%. The upper limit of the content ratio of the silane coupling agent in the mixture is, for example, 10 mass%, preferably, 5 mass%.

[0123]

As necessary, a solvent is further blended, thereby preparing a varnish of the mixture. The solvent is not especially limited. Examples thereof include ketone-based solvents such as acetone, and alcohol-based solvents such as methanol. Preferably, a ketone-based solvent is used.

[0124]

The lower limit of the solid content concentration in the varnish is, for example, 40mass%, preferably, 60mass%. The upper limit of the solid content concentration in the varnish is, for example, 99mass%, preferably, 90mass%.

[0125]

The above-described components (including the additives and solvents) are blended and mixed.

[0126]

For example, conditions under which the non-layered filler and the layered filler are sufficiently dispersed in the thermosetting resin are selected as the conditions for the mixing.

[0127]

A mixer such as a planetary centrifugal mixer, a disperser, a bead mill, a homo mixer, or a planetary mixer is used to stir and mix the above-described components. Preferably, a planetary centrifugal mixer is used to stir and mix the above-described components.

[0128]

A planetary centrifugal mixer is a device that simultaneously carries out a rotation (autorotation) around the center of rotation (the first center) located in the vessel, which is a closed (tubular) container encapsulating the mixture, and a rotation (revolution) around the center of rotation (the second center) located outside the vessel. The axis of the first center and the axis of the second center may be parallel or orthogonal to each other. Preferably, the axes intersect with each other (at an inclination angle of, for example, 30 degrees or more and 60 degrees or less.).

[0129]

A planetary centrifugal mixer can simultaneously give the mixture a shearing force by the autorotation and a centrifugal force by the revolution. In this manner, the non-layered filler and the layered filler can sufficiently be dispersed in the thermosetting resin.

[0130]

A commercialized product can be used as the planetary centrifugal mixer. As an exemplary commercialized product of the planetary centrifugal mixer, the Awatori Rentaro (THINKY mixer) series (manufactured by THINKY CORPORATION) is used.

[0131]

Specifically, when a planetary centrifugal mixer is used as the mixer, the lower limit of the autorotation speed is, for example, 1,000 rpm and the upper limit of the autorotation speed is 10,000 rpm. The lower limit of the revolution speed is, for example, 1,000 rpm and the upper limit of the revolution speed is 10,000 rpm. The lower limit of the time for the mixing is, for example, 1 minute, preferably, 3 minutes. The upper limit thereof is, for example, 30 minutes, preferably, 10 minutes.

[0132]

When a planetary centrifugal mixer is used as the mixer, the lower limit of the centrifugal force on the vessel is, for example, 10 MPa, preferably, 100 MPa. The upper limit thereof is, for example, 10,000 MPa.

[0133]

The mixing may be carried out during or after the blending of the components. Preferably, the mixing is carried out both during and after the blending. Specifically, the thermosetting resin, the non-layered filler, and the layered filler (additionally, the additive) are blended, and subsequently stirred with the above-described mixer (the first mixing). Thereafter, a solvent is added thereto. Then, the mixture is stirred with the above-described mixer (the second mixing).

[0134]

The conditions for the second mixing may be, for example, different from or the same as those of the first mixing. Preferably, the conditions are the same.

[0135]

Thereafter, the varnish is applied to a peeling sheet not illustrated and dried by heating, thereby producing a sealable resin sheet having a sheet shape.

[0136]

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

[0137]

The lower limit of the thickness of the sealable resin sheet is, for example, 10 μm , preferably, 25 μm , more preferably, 50 μm . The upper limit of the thickness of the sealable resin sheet is, for example, 3000 μm , preferably, 1000 μm , more preferably, 500 μm , even more preferably, 300 μm , particularly preferably, 100 μm .

[0138]

To surely produce a thin slice-shaped sample for the TEM observation, the sealable resin sheet 1 in B stage is heated (at normal pressure without pressing) to get into C stage. By that, the sealable resin sheet 1 in C stage can be prepared. The one field-of-view image obtained by the TEM observation of the sealable resin sheet 1 in B stage is practically the same as the one field-of-view image obtained by the TEM observation of the sealable resin sheet 1 in C stage. In other words, the codispersion regions 3 in the sealable resin sheet 1 in B stage are

organized in practically the same manner as the codispersion regions 3 in the sealable resin sheet 1 in C stage are.

[0139]

<Sealing Multi-layer resin Sheet>

The sealable resin sheet may be included together with the second sealable resin sheet in the sealing multi-layer resin sheet.

[0140]

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

[0141]

The sealable multi-layer resin sheet is, for example, the same as the above-described sealable resin sheet 1 except that the codispersion regions 3 including the non-layered filler (the second domains 6) are not observed in the one field-of-view image obtained by the TEM observation of the second sealable resin sheet.

[0142]

The lower limit of the thickness of the second sealable resin sheet is, for example, 25 μm , preferably, 50 μm , more preferably, 100 μm , even more preferably, 150 μm . The upper limit of the thickness of the second sealable resin sheet is, for example, 1000 μm , preferably, 500 μm , more preferably, 300 μm .

[0143]

The sealable multi-layer resin sheet is prepared by bonding the sealable resin sheet and the second sealable resin sheet to each other.

[0144]

<Production of Electronic Element Package>

A method of producing an electronic element package by sealing electronic elements as exemplary elements with the sealable resin sheet is described with reference to FIG. 3A to FIG. 3D.

[0145]

(Example Using Sealable Resin Sheet)

In this method, as illustrated in FIG. 3A, 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.

[0146]

As illustrated in FIG. 3B, an electronic element 21 is separately be prepared.

[0147]

For example, a plurality of electronic elements 21 is mounted on a board 22. The plurality of electronic elements 21 and board 22 are included together with bumps 23 on an element mounting board 24. In short, the element mounting board 24 includes the plurality of electronic elements 21, the board 22, and the bumps 23.

[0148]

The board 22 has an approximately flat plate shape extending in the surface direction. A one-side surface 25 in the thickness direction of the board 22 is provided with terminals (not illustrated) electrically connected to electrodes (not illustrated) of the electronic elements 21.

[0149]

Each of the electronic elements 21 has an approximately flat board shape (chip shape) extending in the surface direction. The electronic elements 21 are disposed with a space therebetween in the surface direction. The other-side surfaces 28 in the thickness direction of the electronic elements 2 are parallel to the one-side surface 25 in the thickness direction of the board 22. The other-side surface 28 in the thickness direction of each of the electronic elements 21 is provided with an electrode (not illustrated). The electrodes of the electronic elements 21 are electrically connected to the terminals of the board 22 via the bumps 23 described next. The other-side surfaces 28 in the thickness direction of the electronic elements 21 are separated from the one-side surface 25 in the thickness direction of the board 22 with gaps (spaces) 26 therebetween.

[0150]

The bumps 23 electrically connect the electrodes (not illustrated) of the electronic elements 21 to the terminals on the board 22, respectively. The bumps 23 are disposed between the electrodes of the electronic elements 21 and the terminals on the board 22. Examples of the material of the bumps 23 include metals such as a solder and gold. The bump 23 has a thickness corresponding to the thickness (height) of the gap 26. The thickness of the bump 23 is appropriately set, depending on the use or purpose of the element mounting board 24.

[0151]

Next, as illustrated in FIG. 3B, the sealable resin sheet 1 is disposed on the plurality of electronic elements 21. Specifically, 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.

[0152]

Next, as illustrated in FIG. 3C, the sealable resin sheet 1 and the element mounting board 24 are pressed. Preferably, the sealable resin sheet 1 and element mounting board 24 are heat-pressed at a low temperature.

[0153]

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.

[0154]

The conditions for the press (for example, pressure, time, and temperature) are not especially limited. The conditions allowing the sealable resin sheet 1 to enter between the electronic elements 21 and preventing the element mounting board 24 from being damaged are selected. More specifically, the conditions for the press are set so that a sufficient shear stress is given to the sealable resin sheet 1, and thus the sealable resin sheet 1 can flow and enter between the adjacent electronic elements 21, cover the side peripheral surface of each of the electronic elements 21, and be in contact with the one-side surface 25 in the thickness direction of the board 22 that does not overlap the electronic elements 21 in the plan view.

[0155]

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.

[0156]

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.

[0157]

When the sealable resin sheet 1 is pressed (is given the shear stress), the sealable resin sheet 1 plastically deforms corresponding in the outer shape of the electronic elements 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 surfaces and side peripheral surfaces of the electronic elements 21.

[0158]

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

[0159]

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 elements 21 in the plan view.

[0160]

In this manner, a sealing body 31 sealing the electronic elements 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.

[0161]

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

[0162]

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.

[0163]

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.

[0164]

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.

[0165]

An edge of the sealing body 31 allowed to slightly enter the gap is further allowed to slightly enter the inside of the gap 26 and become the cured product 41 therein. The degree of the entry is suppressed as much as possible.

[0166]

As illustrated in FIG. 1 and FIG. 2, the sealable resin sheet 1 includes the first domains 5 formed from the non-layered filler and the second domains 6 formed from the layered filler, and has the codispersion regions 3 in which the second domains 6 are located among the first domains 5. Thus, even when the thermosetting resin is temporarily softened by heating and thus the matrix 4 easily flows, the codispersion regions 3 can suppress the flow of the material of the sealable resin sheet 1 (the mixture) as much as possible. Accordingly, as illustrated in FIG. 3D, the entering amount of the cured product 41 between the electronic elements 21 and board 22 can be reduced when the cured product 41 is formed.

[0167]

When the sealable resin sheet 1 is disposed on a plurality of electronic elements 21 varying in size or design or on a plurality of electronic elements 21 connected to terminals varying in size and design and the sealable resin sheet 1 is heated to form a cured product 41, the

entering amount of the cured product 41 that enters the gap therebetween can be controlled. Thus, the plurality of electronic elements 21 can uniformly be sealed.

[0168]

Where the above-described formula (1) ($0.2 < a/b < 5$) is satisfied, the cured product 41 surely covers the peripheral side surfaces of the elements 21 and maintains its excellent sealability while the entering amount of the cured product 41 between the electronic elements 21 and the board 22 can be reduced.

[0169]

When the average value a of the maximum lengths A of the second domain is larger than the average value b of the maximum lengths B of the first domain, the entering amount of the cured product 41 between the electronic elements 21 and the board 22 can further be reduced.

[0170]

When the average value a of the maximum lengths A of the second domain 6 is $1\ \mu\text{m}$ or more, the flow of the material of the sealable resin sheet 1 can be suppressed as much as possible. When the average value a of the maximum lengths A of the second domain 6 is $10\ \mu\text{m}$ or less, the cured product 41 can surely cover the peripheral side surfaces of the electronic elements 21.

[0171]

When the second domain 6 recognized in the one field-of-view image has an aspect ratio of 2 or more, the flow of the material of the sealable resin sheet 1 can be suppressed as much as possible.

[0172]

When the first domains 5 recognized in the one field-of-view image have an approximately circular shape, a cured product 41 with an excellent strength can be formed.

[0173]

(Example Using Sealable Multi-layer Resin Sheet)

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. 4A to FIG. 4D.

[0174]

As illustrated in FIG. 4A, a sealable multi-layer resin sheet 11 is prepared. Specifically, a sealable resin sheet 1 and a second sealable resin sheet 12 are bonded to each other.

[0175]

As illustrated in FIG. 4B, the plurality of electronic elements 21 mounted on the board 22 is prepared.

[0176]

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.

[0177]

As illustrated in FIG. 4C, thereafter, the sealable resin sheet 1 and the element mounting board 24 are pressed.

[0178]

The press makes the sealable resin sheet 1 flow and enter between the adjacent electronic elements 21.

[0179]

On the other hand, the second sealable resin sheet 12 does not contain a codispersion region 3. Thus, when the second sealable resin sheet 12 is pressed, the fluidity of the second sealable resin sheet 12 is not improved. Hence, the entry of the second sealable resin sheet 12 between the adjacent electronic elements 21 is suppressed.

[0180]

In this manner, the sealing body 31 sealing the electronic elements 21 is formed from the sealable multi-layer resin sheet 11.

[0181]

When the sealable resin sheet 1 contains the non-layered filler in a ratio of the above-described lower limit or more and the second sealable resin sheet 12 contains the non-layered filler in a ratio of the above-described lower limit or more, the press illustrated in FIG. 4C can make both the sealable resin sheet 1 and second sealable resin sheet 12 flow.

[0182]

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 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.

[0183]

Thereafter, as illustrated in FIG. 4D, the sealing body 31 is heated, thereby forming the cured product 41 from the sealing body 31.

[0184]

This sealable multi-layer resin sheet 11 can also have the same operations and effects as those of the above-described sealable resin sheet 1.

[0185]

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. 4C. Thus, the time for the step illustrated in FIG. 4C 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. 4C.

[0186]

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.

[0187]

In the step illustrated in FIG. 4C, 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. 4C, 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

resin sheet 1 deforms, following the outer shapes of the electronic elements 21. In the step illustrated in FIG. 4C, the sealable resin sheet 1 is allowed to slightly enter the gap 26.

[0188]

In the step illustrated in FIG. 4D, the flow of the sealable resin sheet 1 is suppressed based on the reduction in complex viscosity η^* with the rise in temperature. Thus, an excessive entry of the sealable resin sheet 1 to the gap 26 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 length Y.

[0189]

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.

[0190]

Although not illustrated, the sealable resin sheet 1 can have a monodispersion region in which the second domains 6 are not located among the first domains 5, and surrounded only by the matrix 4 in the one field-of-view image. Simultaneously, the sealable resin sheet 1 can have a monodispersion region in which the first domains 5 surrounded only by the matrix 4 in the one field-of-view image.

[0191]

To prepare the mixture, the thermosetting resin, the non-layered filler, and the layered filler can be mixed without using the solvent. In such a case, a known kneader is used to knead the thermosetting resin, the non-layered filler, and the layered filler.

[0192]

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

[0193]

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.

[0194]

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

Examples

[0195]

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

[0196]

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

[0197]

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

Main agent: YSLV-80XY manufactured by the new Nippon Steel Chemical Co., Ltd. (bisphenol F type 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: 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 (T_g): -35°C, methyl ethyl ketone solution with a 20 mass% solid content concentration

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

First non-layered filler: FB-8SM (spherical molten silica powder (inorganic filler), average particle size of 7.0 μm)

Second non-layered filler: an inorganic filler made of SC220G-SMJ manufactured by Admatechs Company Limited (average particle size of 0.5 μm) 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.

[0198]

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

[0199]

Preparation Example 1 to Preparation Example 6 and Comparative Preparation Example 1

The components were blended in conformity with Table 1 to prepare the mixture in a vessel. Subsequently, the vessel was set in a planetary centrifugal mixer, and the mixture was stirred (the first mixing). Thereafter, an appropriate amount of a solvent (methyl ethyl ketone) was added to the vessel. Furthermore, the vessel was set in the planetary centrifugal mixer, and the mixture including the solvent was stirred (the second mixing).

[0200]

The type of the planetary centrifugal mixer and the conditions for the mixings are described below.

Planetary centrifugal mixer: Awatori Rentaro (THINKY mixer) model “AR-100”

(First Mixing)

Autorotation speed: 2000 rpm

Revolution speed: 2000 rpm

Mixing time: 5 minutes

(Second Mixing)

Autorotation speed: 2000 rpm

Revolution speed: 2000 rpm

Mixing time: 5 minutes

In this manner, a varnish of the mixture is prepared.

[0201]

Next, 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 μm . The sealable resin sheet 1 was in B stage.

[0202]

Preparation Example 7

A varnish of the mixture was prepared in conformity with the blending formula described in Table 2 in the same manner as in Preparation Example 1. 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 μm . The second sealable resin sheet 12 was in B stage.

[0203]

Examples 1 to 6 and Comparative Example 1

In each of the combinations of Preparation Examples shown in Table 3, 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 μm .

[0204]

Evaluation

The following items were evaluated. The results are shown in Table 3

<TEM Observation>

(1) Presence or Absence of Codispersion region

The sealing multi-layer resin sheet of each of Examples 1 to 6 and Comparative Example 1 was heated at 150°C for 1 hour to produce a sheet in C stage. A sample (thin slice) was produced by sampling the C-stage sheet. Specifically, the C-stage sheet was set on an FIB processing system (FB2200 manufactured by Hitachi High-Tech Corporation) and cut in the thickness direction with an ion beam with an accelerating voltage of 10 to 40kV and a microprobe to obtain the sample.

[0205]

Thereafter, using a field emission scanning electron microscopy (FE-TEM) (JEM-2800 manufactured by JEOL Ltd.), the cross section of the C-stage sheet was observed at an accelerating voltage of 200 kV.

[0206]

The presence or absence of a codispersion region 3 in which the second domains 6 were located among the first domains 5 was detected from the one field-of-view image of the TEM observation.

[0207]

FIG. 1 and FIG. 2 show the one field-of-view image (processed image) of a TEM picture of the C-stage sheet in Example 3.

[0208]

(2) Average Value b of Maximum Lengths B of First Domain, Average value a of Maximum Lengths A of Second Domain 6, and a/b

The above-described first to fourth steps were carried out to calculate each of the average value b of the maximum lengths B of the first domain and the average value a of the maximum lengths A of the second domain 6. Subsequently, a/b was obtained.

[0209]

(3) Area Ratio of Matrix Region, and Ratio of Number of First Domains

Ratios including the area ratio of the matrix region, and the ratio of the number of the first domains were obtained.

[0210]

<Evaluation of Cured Product>

(1) Cured Product Entering Length

The following Steps A to E were carried out to measure the cured product entering length Y.

[0211]

Step A: As illustrated in FIG. 5A, a sample sheet 61 with a longitudinal length of 10 mm, a lateral length of 10 mm, and a thickness of 260 μm was prepared from the sealing multi-layer resin sheet 11 of each of Examples and Comparative Example.

[0212]

Step B: As illustrated in FIG. 5B, 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 μm on a glass board 72 through bumps 23 with a thickness of 20 μm .

[0213]

Step C: As illustrated in FIG. 5C, 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.

[0214]

Step D: As illustrated in FIG. 5D, 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.

[0215]

Step E: As illustrated in an enlarged drawing of FIG. 5D, the cured product entering length Y in which the cured product 41 entered the gap 26 between the dummy element 71 and the glass board 72 from a side edge 75 was measured based on the side edge 75 of the dummy element 71.

[0216]

By the following criteria, the cured product entering length Y was evaluated.

Good: The cured product entering length Y was 0 μm or more and 20 μm or less.

Fair: The cured product entering length Y was more than 20 μm and 30 μm or less, or less than 0 μm and -5 μm or more.

Bad: The cured product entering length Y was more than 30 μm , or, less than -5 μm .

[0217]

In the evaluation, the “minus” means that a space protruding outward from the side edge 75 of the dummy element 71 was formed (see the thick dashed line in FIG. 5D). The absolute value of the “minus” corresponds to a length of the protruding space.

[0218]

[Table 1]

Formula for mixture										
Mixture			Preparation Example 1	Preparation Example 2	Preparation Example 3	Preparation Example 4	Preparation Example 5	Preparation Example 6	Comparative Preparation Example 1	
Layered filler	Organic bentonite (modified by dimethyl distearyl ammonium)	S-Ben NX	(part(s))	0.43	0.57	0.71	0.86	1	1.43	0
	Content in mixture		[%]	3	4	5	6	7	10	0
	Parts to 100 parts of non-layered filler		(part(s))	4	6	7	9	10	14	0
Thermosetting resin (epoxy resin composition)	Main agent	Epoxy resin	(part(s))	1.8	1.73	1.66	1.59	1.52	1.31	2.01
	Curing agent	Phenol resin	(part(s))	0.98	0.94	0.9	0.87	0.83	0.712	1.01
	Curing accelerator	Imidazole-type	(part(s))	0.026	0.025	0.024	0.023	0.022	0.02	0.029
Ratio of main agent in mixture			[%]	13	12	12	11	11	9	14
Ratio of main agent in thermosetting resin			[%]	64	64	64	64	64	64	66
Parts of curing agent to 100 parts of main agent			(part(s))	54	54	54	55	55	54	50
Parts of curing accelerator to 100 parts of main agent			(part(s))	1.4	1.4	1.4	1.4	1.4	1.5	1.4
Content of thermosetting resin in mixture			[%]	20	19	18	17	17	14	21
Thermoplastic resin	Acrylic resin (solid content only)	HME-2006M	(part(s))	0.79	0.76	0.73	0.7	0.67	0.58	0.87
Ratio of thermoplastic resin in mixture			[%]	6	5	5	5	5	4	6
Silane coupling agent	Glycidyl group-containing	KBM-403	(part(s))	0.15	0.15	0.15	0.15	0.15	0.15	0.15
Content of silane coupling agent in mixture			[%]	1	1	1	1	1	1	1
Non-layered filler	First non-layered filler	FB-8SM	(part(s))	6.5	6.5	6.5	6.5	6.5	6.5	6.5
	Second non-layered filler	SC220G-SMJ	(part(s))	3.5	3.5	3.5	3.5	3.5	3.5	3.5
	Content		[%]	70	70	70	70	70	70	70
	Content of first non-layered filler in mixture		[%]	45	46	46	45	45	45	46
Parts of second non-layered filler to 100 parts of first non-layered filler			(part(s))	54	54	54	54	54	54	54
Pigment	Carbon black	#20	(part(s))	0.12	0.11	0.11	0.1	0.099	0.086	0.13
Ratio of pigment to mixture			[%]	1	1	1	1	1	1	1

Formula for mixture

[Table 2]

Mixture						Preparation Example 7	
Formula for mixture	Thermosetting resin (epoxy resin composition)	Main agent	Epoxy resin	YSLV-80XY	(part(s))	1.84	
		Curing agent	Phenol resin	LVR-8210DL	(part(s))	1	
		Curing accelerator	Imidazole-type	2PHZ-PW	(part(s))	0.02	
	Ratio of main agent in mixture					[%]	7
	Ratio of main agent in thermosetting resin					[%]	64
	Parts of curing agent to 100 parts of main agent					(part(s))	54
	Parts of curing accelerator to 100 parts of main agent					(part(s))	1.1
	Content of thermosetting resin in mixture					[%]	11
	Thermoplastic resin	Acrylic resin (solid content only)		HME-2006M	(part(s))	0.17	
	Ratio of thermoplastic resin in mixture					[%]	1
	Silane coupling agent	Glycidyl group-containing		KBM-403	(part(s))	0.35	
	Content of silane coupling agent in mixture					[%]	1
	Non-layered filler	First non-layered filler		FB-8SM	(part(s))	15.15	
		Second non-layered filler		SC220G-SMJ	(part(s))	8.16	
		Content			[%]	87	
		Content of first non-layered filler in mixture			[%]	57	
		Parts of second non-layered filler to 100 parts of first non-layered filler			(part(s))	54	
	Pigment	Carbon black		#20	(part(s))	0.1	
	Ratio of pigment to mixture					[%]	0

[0220]

[Table 3]

Sealable multi-layer resin sheet	Example 1	Example 2	Example 3	Example 4	Example 5	Example 6	Comparative Example 1
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
Sealable resin sheet	Preparation Example 1	Preparation Example 2	Preparation Example 3	Preparation Example 4	Preparation Example 5	Preparation Example 6	Comparative Preparation Example 1
Properties of sealable resin sheet	Presence or absence of covariance region		Presence	Presence	Presence	Presence	Absence
	First domain	Shape	Circular shape	Circular shape	Circular shape	Circular shape	Circular shape
	Second domain	Average value b of the maximum lengths B	1.1	0.7	1.1	0.6	2.5
		Shape	Layered shape (board shape)	Layered shape (board shape)	Layered shape (board shape)	Layered shape (board shape)	-
	Average value a of the maximum lengths A		1.3	2.1	1.8	1.5	2.0
Evaluation of sealable multi-layer resin sheet	a/b		1.2	3.0	1.7	2.5	0.8
	Cured product entering length Y in gap		Good	Good	Good	Good	Bad

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

[0221]

The sealable resin sheet is used to seal an element.

Description of Reference Numerals

[0222]

- 1 sealable resin sheet
- 3 codispersion region
- 4 matrix
- 5 first domain
- 6 second domain
- 21 electronic element
- A maximum length of the largest second domain
- B maximum length of the largest first domain
- a average value of the maximum lengths of the second domain
- b average value of the maximum lengths of the first domain

Claims

1. A sealable resin sheet for sealing an element, the sealable resin sheet containing:
a matrix formed from thermosetting resin;
5 first domains formed from a non-layered filler and dispersed in the matrix,
wherein a lower limit of a content ratio of the non-layered filler in the sealable resin sheet
is 50 mass%; and
second domains formed from a layered filler and dispersed in the matrix,
wherein the sealable resin sheet includes a codispersion region in which the
10 second domain is located among the first domains.

2. The sealable resin sheet according to Claim 1, wherein an average value a of
maximum lengths A of the second domain and an average value b of maximum lengths
B of the first domain are obtained by first to fourth steps described below, and satisfy a
15 formula (1) described below:

$$0.2 < a/b < 5 \quad (1),$$
wherein the first step comprises obtaining one field-of-view image by a TEM
observation of the sealable resin sheet,
wherein the second step comprises observing the one field-of-view image by ten
20 observers to measure the maximum lengths A of the largest second domain among the
second domains and the maximum lengths B of the largest first domain among the first
domains,
wherein the third step comprises obtaining the average value a of the maximum
lengths A measured by the ten observers, and the average value b of the maximum lengths
25 B measured by the ten observers, and
wherein the fourth step comprises substituting the average value a and the
average value b for the formula (1)

3. The sealable resin sheet according to Claim 2, wherein the average value a of the
30 maximum lengths A of the second domain is larger than the average value b of the
maximum lengths B of the first domain.

4. The sealable resin sheet according to Claim 2, wherein the average value a of the maximum lengths A of the second domain is $1\text{ }\mu\text{m}$ or more and $10\text{ }\mu\text{m}$ or less.
- 5 5. The sealable resin sheet according to Claim 1, wherein one field-of-view image is obtained by a TEM observation of the sealable resin sheet, and the second domain recognized in the one field-of-view image has an aspect ratio of 2 or more.
6. The sealable resin sheet according to Claim 1, wherein one field-of-view image
10 is obtained by a TEM observation of the sealable resin sheet, and the first domains recognized in the one field-of-view image have an approximately circular shape.