SPECIFICATION

TITLE OF THE INVENTION: SEALABLE SHEET AND ELECTRONIC ELEMENT DEVICE

BACKGROUND ART

[0001]

The present invention relates to a sealable sheet and an electronic element device.

[0002]

It has conventionally known to produce an electronic element device by sealing an electronic element with a sealable sheet (for example, see Patent Document 1 below).

[0003]

The sealable sheet described in Patent Document 1 contains an epoxy resin, a phenol resin (a curing agent), and an imidazole-based curing accelerator. The sealable sheet forms a sealing body by heat-curing after the electronic element is embedded therein.

Citation List

Patent Document

[0004]

Patent Document 1: Japanese Unexamined Patent Publication No. 2017-92103

SUMMARY OF THE INVENTION

PROBLEM TO BE SOLVED BY THE INVENTION

[0005]

The sealable sheet may be adhered to the one-side surface of the electronic element and left to stand in the adhered state for a long period of time (for example, for about 24 hours). However, the adhesive force of the sealable sheet described in Patent Document 1 drastically decreases during the long standing. This drastic

decrease in the adhesive force adversely reduces the positional accuracy of the sealable sheet relative to the electronic element. There is a need to suppress the percentage of the change in the adhesive force of the sealable sheet.

[0006]

It is also necessary that the amount of the unreacted material is small in the sealing body sheet that is a cured product of the sealable sheet.

[0007]

The present invention provides a sealable sheet and an electronic element device, both of which can suppress the percentage of the change in the adhesive force and reduce the unreacted material in the sealing body sheet that is a cured product.

MEANS FOR SOLVING THE PROBLEM

[0008]

The present invention [1] includes a sealable sheet comprising: an epoxy resin; a phenol resin; a urea-based curing accelerator; and an inorganic filler, wherein a percentage ([A0-A1]/A0×100) of a change (A0-A1) from an adhesive force A0 that the sealable sheet has immediately after the sealable sheet is produced to an adhesive force A1 that the sealable sheet has after the sealable sheet is left to stand at 23℃ and 50% RH for 24 hours over the adhesive force A0 is 10% or less, and wherein, with respect to a calorific value of the sealable sheet that is obtained by differential scanning calorimetry immediately after the sealable sheet is produced, a ratio of a calorific value of the sealable sheet that is obtained by differential scanning calorimetry after the sealable sheet is heated at 150℃ for 20 minutes is 0.10 or less.

[0009]

Of the sealable sheet, the percentage ([A0-A1]/A0×100) of the change (A0-A1) from the adhesive force A0 to the adhesive force A1 after the sealable sheet is left to stand at 23℃ and 50% RH for 24 hours over the adhesive force A0 is 10% or less. Even when the sealable sheet is left to stand in a low-temperature region for a long period of time, the percentage of the change in the adhesive force is suppressed.

[0010]

Further, the ratio of the calorific value of the sealable sheet obtained by differential scanning calorimetry after the sealable sheet is heated at 150℃ for 20 minutes to the calorific value of the sealable sheet obtained by differential scanning calorimetry immediately after the sealable sheet is produced is 0.10 or less. Thus, the amount of the unreacted material in the sealing body sheet is small.

[0011]

The present invention [2] includes the sealable sheet described in [1], wherein the urea-based curing accelerator is an aliphatic urea compound.

[0012]

An aliphatic urea compound exhibits a low reactivity in the low-temperature region and thus is highly effective in reducing the percentage of the change in the entering length at the gap between the mounting board and the electronic element.

[0013]

The present invention [3] includes the sealable sheet described in the above [1] or [2], wherein a percentage ([L1-L2]/L1×100) of a change (L1-L2) from a first entering length L1 obtained by Test X to a second entering length L2 obtained by Test Y over the first entering length L1 is 90% or less.

[0014]

<Test X>

The following first to fifth steps are carried out in numerical order.

[0015]

The first step: A sample with a thickness of 200 μm is prepared from a sealable sheet immediately after the sealable sheet is produced. Separately, a mounting board including a glass board and nine dummy chips is prepared. The dummy chips are mounted on an upper surface of the glass board through bumps, and each have a size of 1 mm × 1 mm × a thickness of 200 μm, and are disposed in a row at 300 μm intervals.

[0016]

The second step: The sample is pressed toward the glass board by vacuum

flat-plate press at a temperature of 65℃, a degree of vacuum of 1.6 kPa or less, a pressure of 0.2 MPa for 60 seconds.

[0017]

The third step: The sample is brought into contact with upper surfaces of the dummy chips and left to stand under the conditions of 23℃ and 50% RH for 30 minutes.

[0018]

The fourth step: The sample is heat-cured by heating the sample at 150℃ under atmospheric pressure for one hour.

[0019]

The fifth step: An entering length L1 in which the sample enters a void defined between a lower surface of a dummy chip that is located at the center of the glass board and the upper surface of the glass board is measured.

[0020]

<Test Y>

The first to fifth steps are carried out. However, the time of standing in the third step is changed from 30 minutes to 24 hours. In the fifth step, a second entering length L2 of the sample is measured.

[0021]

Of the sealable sheet, the percentage of the change from the first entering length L1 to the second entering length L2 over the first entering length L1 is 90% or less. Thus, even when a long period of time has elapsed in the low-temperature region, the change in the entering length of the sealing body sheet is suppressed. Consequently, excellent sealing stability is achieved.

[0022]

The present invention [4] includes an electronic element device comprising: a sealing body sheet formed from the sealable sheet described in any one of the above [1] to [3]; and an electronic element sealed with the sealing body sheet.

[0023]

The electronic element device includes the sealing body sheet formed from

the sealable sheet and thus achieves excellent sealing reliability and makes the amount of the unreacted material small.

EFFECTS OF THE INVENTION

[0024]

The sealable sheet and electronic element device of the present invention can suppress the percentage of the change in the adhesive force and reduce the unreacted material in the sealing body sheet that is a cured product.

BRIEF DESCRIPTION OF THE DRAWINGS

[0025]

FIG. 1A illustrates the first step of each of Test X and Test Y. FIG. 1B illustrates the second to fifth steps of each of Test X and Test Y.

FIG. 2 is a cross-sectional view of an electronic element device.

DESCRIPTION OF THE EMBODIMENT

[0026]

1. Sealable Sheet

One embodiment of the sealable sheet of the present invention is described.

[0027]

The sealable sheet is used for sealing an electronic element. The sealable sheet has a thickness. The sealable sheet extends in a surface direction. The surface direction is orthogonal to a thickness direction. The sealable sheet has an approximately plate shape (film shape).

[0028]

1.1 Components of Sealable Sheet

The sealable sheet contains an epoxy resin, a phenol resin, a urea-based curing accelerator, and an inorganic filler. Specifically, the sealable sheet is formed into a sheet shape from a thermosetting composition containing the above-described components. Further, the thermosetting composition contains a resin component and a filler component. The blending ratio of the resin component in the thermosetting composition is, for example, 1 mass% or more, preferably 5 mass% or more, and, for example, 25 mass% or less, preferably 15 mass% or less.

The blending ratio of the filler component in the thermosetting composition is, for example, 75 mass% or more, preferably 85 mass% or more, and, for example, 99 mass% or less, preferably 90 mass% or less. The blending ratio of the filler component to 100 parts by mass of the resin component is, for example, 300 parts by mass or more, preferably 700 parts by mass or more, and, for example, 3000 parts by mass or less, preferably 1500 parts by mass or less.

[0029]

The resin component contains the above-described epoxy resin, phenol resin, and urea-based curing accelerator. The filler component contains the inorganic filler.

[0030]

1.1.1 Epoxy Resin

The epoxy resin is the main agent in the thermosetting composition. The type of the epoxy resin is not limited. Examples of the epoxy resin include bifunctional epoxy resins and trifunctional or more of multifunctional epoxy resins. Examples of the bifunctional epoxy resin include bisphenol A epoxy resin, bisphenol F epoxy resin, modified bisphenol A epoxy resin, modified bisphenol F epoxy resin, and biphenyl epoxy resin. Examples of the multifunctional epoxy resin include 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 epoxy resin, a bifunctional epoxy resin is preferable. More preferred examples of the epoxy resin include bisphenol F epoxy resin and modified bisphenol A epoxy resin. In terms of the suppression of the percentage of the change in the adhesive force (described below) and in terms of the suppression of the percentage of the reduction in the entering length (described below), bisphenol F epoxy resin is preferable. On the other hand, in terms of the reduction in the ratio of the calorific value (described below), modified bisphenol A epoxy resin is preferable.

[0031]

The blending ratio of the epoxy resin in the sealable sheet is not limited.

The blending ratio of the epoxy resin in the sealable sheet is, for example, 1 mass% or more, preferably 2 mass% or more, and, for example, 25 mass% or less, preferably 10 mass% or less. The blending ratio of the epoxy resin in the resin component is, for example, 5 mass% or more, preferably 15 mass% or more, and, for example, 50 mass% or less, preferably 25 mass% or less.

[0032]

1.1.2 Phenol Resin

The phenol resin is a latent phenol resin that cures the epoxy resin. The type of the phenol resin is not limited. Examples of the phenol resin include novolak phenol resin and phenol-aralkyl resin. These epoxy resins can be used alone or in combination of two or more. As the phenol resin, in terms of the reduction in the percentage of the change in the adhesive force (described below), phenol-aralkyl resin is preferable. As the phenol resin, in terms of the reduction in the ratio in DSC (described below), novolak phenol resin is preferable.

[0033]

The blending ratio of the phenol resin to 100 parts by mass of the epoxy resin is, for example, 10 parts by mass or more and 150 parts by mass or less. The blending ratio of the phenol resin in the sealable sheet is, for example, 0.5 mass% or more, preferably 2 mass% or more, and, for example, 5 mass% or less, preferably 2.5 mass% or less. The blending ratio of the phenol resin in the resin component is, for example, 3 mass% or more, preferably 10 mass% or more, and, for example, 40 mass% or less, preferably 15 mass% or less.

[0034]

1.1.3 Urea-based Curing Accelerator

While not accelerating the curing reaction of the sealable sheet in the low-temperature region, a urea-based curing accelerator accelerates the above-described curing reaction in the high-temperature region. Specifically, the urea-based curing accelerator does not enhance thermal dissociation in the low-temperature region (because the isocyanate group blocks the dissociation of the amino group). On the other hand, in the high-temperature region, thermal dissociation progresses and

generates an isocyanate compound and an amine compound, and then the above-described amine compound accelerates the curing reaction. The low-temperature region is, for example, 100℃ or less, further 90℃ or less, and the lower limit of the low temperature is not limited. The high-temperature region is, for example, more than 100℃, further 110℃ or more, 120℃ or more, 130℃ or more, 140℃ or more, and the high-temperature region is, for example, 210℃ or less, further 200℃ or less, 190℃ or less, 180℃ or less.

[0035]

Examples of the urea-based curing accelerator include a urea compound containing a dialkylamino group. Examples of the dialkylamino group include dimethylamino. Because the urea-based curing accelerator includes the above-described dimethylamino, dimethylamine is produced by thermal dissociation.

[0036]

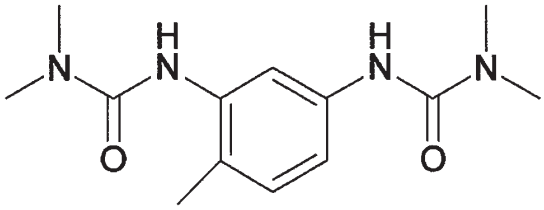
Further, examples of the urea-based curing accelerator include aromatic urea compounds and aliphatic urea compounds. These can be used alone or in combination of two or more.

[0037]

The aromatic urea compound contains, for example, the above-described dialkylamino group and an aromatic group. Examples of the aromatic urea compound include a dimethylamino group-containing aromatic urea compound represented by the following formula (1).

[0038]

Chemical Formula 1



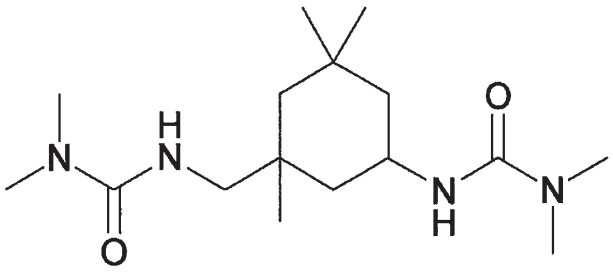
(1)

[0039]

The aliphatic urea compound contains, for example, the above-described dialkylamino group and an aliphatic group. Examples of the aliphatic urea compound include a dimethylamino group-containing aliphatic urea compound represented by the following formula (2).

[0040]

Chemical Formula 2



(2)

[0041]

As the urea-based curing accelerator, an aliphatic urea compound is preferable. As compared with an aromatic urea compound, an aliphatic urea

compound has a low reactivity in the low-temperature region and thus is highly effective in reducing the percentage of the change in the entering length (described below). On the other hand, an aliphatic urea compound exhibits the same curing acceleration effect as an aromatic urea compound in the high-temperature region.

[0042]

The blending ratio of the urea-based curing accelerator to 100 parts by mass of the epoxy resin is, for example, 0.1 parts by mass or more, preferably 1 parts by mass or more, more preferably 2 parts by mass or more, and, for example, 10 parts by mass or less, preferably 5 parts by mass or less, preferably 3 parts by mass or less. The blending ratio of the urea-based curing accelerator in the sealable sheet is, for example, 0.01 mass% or more, preferably 0.02 mass% or more, and, for example, 1 mass% or less, preferably 0.2 mass% or less. The blending ratio of the urea-based curing accelerator in the resin component is, for example, 0.1 mass% or more, preferably 0.2 mass% or more, more preferably 0.3 mass% or more, and, for example, 1.5 mass% or less, preferably 1.0 mass% or less, preferably 0.7 mass% or less. When the blending ratio of the urea-based curing accelerator is the above-described lower limit or more and the upper limit or less, the percentage of the change in the entering length can be reduced while a high curing acceleration effect is achieved.

[0043]

1.1.4 Inorganic Filler

Examples of the inorganic filler include silicate compounds and silicon compounds. The silicate compound and silicon compound are described in Japanese Unexamined Patent Publication No. 2021-097206.

[0044]

The inorganic filler contains a layered silicate compound. The layered silicate compound is a flow reducer at the curing used to reduce the fluidity of the cured body sheet when the sealable sheet is heated to form the cured body sheet. Examples of the layered silicate compound include montmorillonite. The layered silicate compound may have a surface modified with an organic component. Examples of the organic component include alkylammonium. Examples of the

alkylammonium include dimethyl distearyl ammonium. The above-described layered silicate compound is described, for example, in Japanese Unexamined Patent Publication No. 2021-097206.

[0045]

The blending ratio of the inorganic filler in the sealable sheet (the thermosetting resin composition) is, for example, 50 mass% or more, preferably 60 mass% or more, more preferably 65 mass% or more, and, 80 mass% or less, preferably 70 mass% or less. The blending ratio of the inorganic filler to 100 parts by mass of the resin component is, for example, 200 parts by mass or more, preferably 300 parts by mass or more, and, for example, 1500 parts by mass or less, preferably 500 parts by mass or less.

The details of the blending ratio of the inorganic filler are described, for example, in Japanese Unexamined Patent Publication No. 2021-097206.

[0046]

In the present embodiment, preferably, the resin component does not contain a curing accelerator other than the above-described urea-based curing accelerator. Examples of the curing accelerator other than the urea-based curing accelerator include imidazole-based curing accelerators and phosphorus curing accelerators. In particular, more preferably, the resin component does not contain an imidazole-based curing accelerator. Because its amino group easily enhances the curing reaction in the low-temperature region, the imidazole-based curing accelerator is not appropriate for the present invention. On the other hand, the phosphorus curing accelerator has low curing acceleration properties, and it takes time to cause the curing reaction. This reduces the manufacturing efficiency of the electronic element device to be described below (see FIG. 2). However, the resin component is allowed to contain a small amount of the phosphorus curing accelerator.

[0047]

1.1.5 Additive Agent

For example, an additive agent can be added to the thermosetting resin composition. Examples of the additive agent include a thermoplastic resin as the

resin component, a silane coupling agent, and a pigment as the filler component. The types and blinding ratios of other additive agents are not limited. The types and blending ratios of the other additive agents are described in Japanese Unexamined Patent Publication No. 2021-097206.

[0048]

1.1.6 Production of Sealable Sheet

To produce the sealable sheet, the thermosetting resin composition is prepared by blending the above-described components in the above-described ratios. Preferably, the above-described components are sufficiently stirred. As necessary, a solvent (for example, a ketone-based solvent such as methyl ethyl ketone) is further blended, thereby preparing a varnish. Thereafter, the varnish is applied to a release liner not illustrated and then dried by heating, thereby producing a sealable sheet having a sheet shape as a film. Alternatively, without preparing a varnish, the sealable sheet (film) can be formed from the thermosetting resin composition by kneading extrusion.

[0049]

A plurality of the above-described films can be laminated (adhered to each other) to produce a thick sealable sheet.

[0050]

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

[0051]

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

[0052]

1.2 Percentage of Change in Adhesive Force

The sealable sheet has the adhesive force A0 immediately after the sealable sheet is produced and the adhesive force A1 after the sealable sheet is left to stand at 23℃ and 50% RH for 24 hours. The percentage ([A0-A1]/A0×100) of the change (A0-A1) from the adhesive force A0 of the sealable to the adhesive force A1 of the sealable sheet over the adhesive force A0 is 10% or less.

[0053]

Where the percentage of the change from the adhesive force A0 to the adhesive force A1 over the adhesive force A0 is more than 10%, the adhesive force drastically reduces when the sealable sheet is left to stand in the low-temperature region for a long period of time.

[0054]

On the other hand, the above-described percentage of the change is 10% or less in the present sealable sheet. Thus, even when the sealable sheet is left to stand in the low-temperature region for a long period of time, the percentage of the change in the adhesive force is suppressed as much as possible.

[0055]

The percentage ([A0-A1]/A0×100) of the change (A0-A1) from the adhesive force A0 of the sealable sheet immediately after the production of the sealable sheet to the adhesive force A1 of the sealable sheet after the freshly-produced sealable sheet is left to stand at 23℃ and 50% RH for 24 hours over the adhesive force A0 is preferably 9% or less, more preferably 7% or less, even more preferably 5% or less, particularly preferably 3% or less. The above-described percentage of the change in the adhesive force is, for example, 0.001% or more, further 0.01% or more, 0.1% or more.

[0056]

The “immediately after the production” includes, for example, in 10 minutes after the production, further in five minutes, and further in three minutes.

[0057]

The “long period of time” is, for example, one hour or more and 50 hours or

less.

[0058]

To obtain the adhesive forces A0 and A1, two plates facing each other up and down are prepared. The sealable sheet is fixed to the lower plate, and subsequently the upper plate is pressed against the sealable sheet. Thereafter, the upper plate is lifted at 5 μm/sec to obtain the necessary load for peeling the upper plate from the sealable sheet as "the adhesive force of the sealable sheet immediately after the production of the sealable sheet".

[0059]

1.3 Ratio of Calorific Value by DSC

The calorific value of the sealable sheet is obtained by differential scanning calorimetry (DSC) immediately after the production of the sealable sheet. Further, the calorific value of the sealable sheet is obtained by differential scanning calorimetry after the freshly-produced sealable sheet is heated at 150℃ for 20 minutes. The ratio of the calorific value of the heated sealable sheet to the calorific value of the freshly-produced sealable sheet is, 0.10 or less, preferably 0.09 or less, more preferably 0.07 or less, even more preferably 0.05 or less, particularly preferably 0.03 or less.

[0060]

The calorific value of the sealable sheet is obtained based on the reaction of the unreacted material in the sealable sheet. Further, the heating at 150℃ is assumed as the heating for thermally curing the sealable sheet after the electronic element is sealed therein. It is preferable that the amount of unreacted material is small in a sealing body sheet (described below) after the above-described thermal curing. Thus, it is preferable that the ratio of the calorific value of the sealable sheet after the freshly-produced sealable sheet is heated at 150℃ for 20 minutes to the calorific value of the sealable sheet immediately after the production of the sealable sheet is low.

[0061]

In the present embodiment, the above-described ratio of the calorific value is

0.10 or less, and thus the amount of the unreacted material in the sealing body sheet is small. Further, when the above-described ratio is the above-described preferred upper limit or less, the amount of the unreacted material in the sealing body sheet becomes smaller. This is preferable.

[0062]

For the calculation of the calorific value of the sealable sheet, the DSC curve of the sealable sheet is obtained. The calorific value (mJ/mg) is obtained from the value of integral of the peak area. Differential scanning calorimetry (DSC) is described in Examples below.

[0063]

1.4 Percentage of Change in Entering Length

The percentage ([L1-L2]/L1×100) of the change (L1-L2) from a first entering length L1 obtained by Test X to a second entering length L2 obtained by Test Y over the above-described first entering length L1 is, for example, 90% or less, preferably 70% or less, more preferably 65% or less, even more preferably 59% or less, and, for example, 10% or more, further 25% or more, 45% or more.

[0064]

The percentage of the change from the first entering length L1 to the second entering length L2 means the degree to which the change in the entering length is effectively reduced when a long period of time has elapsed in the low-temperature region. Hence, a low percentage is preferable and achieves excellent sealing stability.

[0065]

<Test X>

In Test X, the following first to fifth steps are carried out in numerical order. By that, the entering length L1 is measured.

[0066]

The first step: As illustrated in FIG. 1A, a sample 11 with a thickness of 200 μm is prepared from a sealable sheet 1 immediately after the sealable sheet 1 is produced. Separately, a glass board 2 and nine dummy chips 4 disposed on an

upper surface of the glass board 2 through bumps 3 are prepared. Each of the nine dummy chips 4 has a size of 1 mm × 1 mm × a thickness of 200 μm. The nine dummy chips 4 are disposed in three columns × three rows at 300 μm intervals.

[0067]

The second step: As illustrated in FIG. 1B, the sample 11 is pressed toward the glass board 2 by vacuum flat-plate press at temperature of 65℃, a degree of vacuum of 1.6 kPa or less, a pressure of 0.2 MPa for 60 seconds.

[0068]

The third step: The sample 11 is left to stand at 23℃ and 50% RH for 30 minutes.

[0069]

The fourth step: The sample 11 is thermally cured by heating the sample 11 at 150℃ under atmospheric pressure for one hour.

[0070]

The fifth step: An entering length L1 in which the sample enters a void 10 defined between a lower surface of a dummy chip 4 that is located in the center of the glass board 2 and the upper surface of the glass board 2.

[0071]

Although not illustrated, if an unfilled space is formed outside the above-described void 10, a length L1 of the unfilled space is obtained as a negative value.

[0072]

<Test Y>

Separately, in Test Y, the first to fifth steps are carried out. However, the time of standing in the third step is changed from 30 minutes to 24 hours. In the fifth step, the second entering length L2 of the sample 11 is measured.

[0073]

The first entering length L1 is obtained by Test X. The second entering length L2 is obtained by Test Y. The percentage ([L1-L2]/L1×100) of the change (L1-L2) from the above-described first entering length L1 to the second entering length L2 over the above-described first entering length L1 is obtained.

[0074]

2. Electronic Element Device

With reference to FIG. 2, one embodiment of the electronic element device including the sealing body sheet formed from the above-described sealable sheet is described.

[0075]

An electronic element device 5 has a thickness. The electronic element device 5 extends in the surface direction. The electronic element device 5 includes a sealing body sheet 6 and an electronic element 7. The electronic element device 5 further includes a bump 8 and a mounting board 9.

[0076]

2.1 Sealing Body Sheet 6

The sealing body sheet 6 seals the electronic element 7. The sealing body sheet 6 is disposed at a one-end portion of the electronic element device 5 in the thickness direction. The sealing body sheet 6 forms the one-side surface of the electronic element device 5 in the thickness direction. The sealing body sheet 6 is a cured body of the above-described sealable sheet 1 (see FIG. 1A and FIG. 1B).

[0077]

2.2 Electronic Element 7

In the present embodiment, a plurality of the electronic elements 7 is included in the electronic element device 5. The electronic elements 7 each have an approximately rectangular shape in the cross-sectional view. The electronic elements 7 are disposed in the surface direction and separated from each other by an interval. An electrode (not illustrated) is provided on the other-side surface of each of the electronic elements 7 in the thickness direction. In the present invention, a plurality of electrodes is provided to each of the electronic elements 7. Examples of the electronic element 7 include a semiconductor device.

[0078]

2.3 Bump 8

The bumps 8 electrically connect the electrodes (not illustrated) of the

electronic elements 7 and the electrodes of the mounting board 9. The bumps 8 are disposed between the electrodes of the electronic elements 7 and the electrodes of the mounting board 9. The bumps 8 each have a shape extending in the thickness direction.

[0079]

2.4 Mounting Board 9

The mounting board 9 is disposed at the other-end portion of the electronic element device 5 in the thickness direction. The mounting board 9 forms the other-side surface of the electronic element device 5 in the thickness direction. The mounting board 9 extends in the surface direction. The mounting board 9 has a sheet shape. A one-side surface of the mounting board 9 in the thickness direction faces the other-side surface of the electronic element 7 in the thickness direction. The one-side surface of the above-described mounting board 9 is provided with the electrodes (not illustrated).

[0080]

A gap 10 is defined between a region of the one-side surface of the mounting board 9 facing the above-described electronic element 7 and the other-side surface of the electronic element 7. The entry of a part of the sealing body sheet 6 to a peripheral end of the gap 10 is allowed.

[0081]

3. Operations and Effects of One Embodiment

In one embodiment, the percentage ([A0-A1]/A0×100) of the change (A0-A1) from the adhesive force A0 of the freshly-produced sealable sheet to the adhesive force A1 of the sealable sheet after the sealable sheet is left to stand at 23℃ and 50% RH for 24 hours over the adhesive force A0 is 10% or less. Thus, even when the sealable sheet is left to stand in the low-temperature region for a long period of time, the percentage of the change in the adhesive force can be suppressed as much as possible.

[0082]

The calorific value of the sealable sheet is obtained by differential scanning

calorimetry immediately after the sealable sheet is produced and after the sealable sheet is heated at 150℃ for 20 minute. The ratio of the calorific value of the heated sealable sheet to the calorific value of the freshly-produced sealable sheet is 0.10 or less, and thus the amount of the unreacted material in the sealing body sheet 6 is small.

[0083]

Because an aliphatic urea compound exhibits a low reactivity in the low temperature region, the sealable sheet including the urea-based curing accelerator that is an aliphatic urea compound is highly effective in reducing the percentage of the change in the entering length at the gap between the mounting board and the electronic element.

[0084]

In the sealable sheet, the percentage ([L1-L2]/L1×100) of the change (L1-L2) from the first entering length L1 obtained by Test X to the second entering length L2 obtained by Test Y over the first entering length L1 is 90% or less, and thus excellent sealing stability is achieved even after a long period of time has elapsed in the low-temperature region.

[0085]

The electronic element device 5 includes the sealing body sheet 6 formed from the above-described sealable sheet. Thus excellent sealing stability is achieved, and the amount of the unreacted material is small.

[Examples]

[0086]

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

[0087]

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

Epoxy resin A: YSLV-80XY manufactured by the new Nippon Steel Chemical Co., Ltd., bisphenol F type epoxy resin

Epoxy resin B: EPICLON EXA-4850-150 manufactured by DIC Corporation, modified bisphenol A type epoxy resin

Phenol resin A: LVR- 8210DL manufactured by Gunei Chemical Industry Co., Ltd. novolac-type phenol resin

Phenol resin B: HME-7851SS manufactured by Meiwa Plastic Industries, Ltd., phenol-aralkyl resin

[0088]

Thermoplastic resin: HME-2006M manufactured by Negami Chemical Industrial Co,.Ltd., carboxyl group-containing acrylic acid ester copolymer, a methyl ethyl ketone solution with a 80 mass% solid content concentration

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

[0089]

Curing accelerator A: U-CAT 3513N manufactured by San-Apro Ltd., the urea-based curing accelerator represented by the above-described formula (2), an aliphatic urea compound

Curing accelerator B: U-CAT 3512T manufactured by San-Apro Ltd., the urea-based curing accelerator represented by the above-described formula (1), an aromatic urea compound

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

Curing accelerator D: TPP-K manufactured by HOKKO CHEMICAL INDUSTRY CO., LTD., tetraphenylphosphonium tetraphenylborate

[0090]

Inorganic filler A: FB-8SMFD manufactured by Denka Company Limited., spherical molten silica powder, average particle size of 7.0 μm

Inorganic filler B: 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 of which surfaces are finished with 1 part by mass of a silane coupling agent with respect to 100 parts by mass of the inorganic filler.

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

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

Solvent: methyl ethyl ketone

[0091]

Example 1

A film was formed by applying a varnish to a surface of a release liner consisting of a polyethylene terephthalate film (PET film) having a surface finished by silicone mold release. The varnish was prepared by blending and mixing the components according to the formulations shown in Table 1. Next, the film was heated and dried at 120℃ for 1.5 minutes, thereby producing a film with a thickness of 50 μm on the surface of the release liner. The film was in Stage B. Next, four films were bonded to each other at 90℃, thereby producing a sample 11 with a thickness of 200 μm. The sample 11 was also in B stage.

[0092]

Examples 2 to 5 and Comparative Examples 1 and 2

Samples 11 of the sealable sheet were produced in the same manner as Example 1 except that the blending formulations were changed according to Table 1.

[0093]

<<Evaluations>>

Of each of Examples 1 to 5 and Comparative Examples 1 and 2, the following items were evaluated. The results are shown in Table 1.

[0094]

<Percentage of Change of Adhesive Force A0-A1>

Two plates with a diameter of 15 mm were mounted on a viscoelasticity measuring instrument (RSA-G2 manufactured by TA instruments). Each of the upper and lower plates had a circular disk shape. A sample 11 was fixed to the lower plate with a double-sided tape. Thereafter, the upper plate (probe) was put down under 25℃ atmosphere and pressed against the sample 11 with a load of 1.74 N for 60 seconds. Thereafter, the upper plate was lifted at 5 μm/sec to measure the necessary load for peeling the upper plate from the sample 11 as "the adhesive force A0 of the freshly-produced sealable sheet".

[0095]

Separately, after the freshly-produced sample 11 was left to stand at 23℃ and 50% RH for 24 hours, the load necessary for peeling the upper plate from the sample 11 was measured as "the adhesive force A1 of the sealable sheet that was left to stand at 23℃ and 50% RH for 24 hours" in the same manner as described above.

[0096]

Then, the percentage ([A0-A1]/A0×100) of the change (A0-A1) from the adhesive force A0 of the freshly-produced sample 11 to the adhesive force A1 of the sample 11 left to stand at 23℃ and 50% RH for 24 hours over the adhesive force A0 was obtained.

[0097]

<Ratio of Calorific Value>

The calorific value of the freshly-produced sample 11 was measured by differential scanning calorimetry (DSC). Specifically, the calorific value was obtained from the DSC curve drawn by differential scanning calorimetry under the following conditions.

[0098]

Differential scanning calorimeter: Q2000 manufactured by TA instruments

Measurement temperature: -10℃ to 300℃

Rate of temperature increase: 10℃/min

Sample pan: aluminum pan

[0099]

Separately, after the freshly-produced sample 11 was heated at 150℃ for 20 minutes, the calorific value of the heated sample 11 was measured by differential scanning calorimetry (DSC).

[0100]

Then, the ratio of the calorific value of the sample 11, which is obtained by differential scanning calorimetry after the sample 11 was heated at 150℃ for 20 minutes, to the calorific value of the sample 11, which is obtained by differential scanning calorimetry immediately after the sample 11 was produced, was obtained.

[0101]

<Percentage of Change of Entering length L1-L2>

<Test X>

In Test X, the following first to fifth steps were carried out in this order. By that, the entering length L1 was measured.

[0102]

The first step: As illustrated in FIG. 1A, a sample 11 with a thickness of 200 μm was prepared from a freshly-produced sample 11. Separately, a glass board 2 and nine dummy chips 4 mounted on an upper surface of the glass board 2 through bumps 3 were prepared. Each of the nine dummy chips 4 had a size of 1 mm × 1 mm × a thickness of 200 μm. The nine dummy chips 4 were disposed at 300 μm intervals. The nine dummy chips 4 were disposed in three columns and three rows.

[0103]

The second step: As illustrated in FIG. 1B, the sample 11 was pressed toward the glass board 2 by vacuum flat-plate press at a temperature of 65℃, a degree of vacuum of 1.6 kPa or less, and a pressure of 0.2 MPa for 60 seconds.

[0104]

The third step: The sample 11 was brought into contact with the upper

surfaces of the dummy chips 4 and left to stand under the conditions of 23℃ and 50% RH for 30 minutes.

[0105]

The fourth step: The sample 11 was heat-cured by heating the sample 11 at 150℃ under atmospheric pressure for one hour.

[0106]

The fifth step: The entering length L1 in which the sample 11 enters the void 10 defined between the lower surface of a dummy chip 7 located at the center of the glass board 2 and the upper surface of the glass board 2.

[0107]

<Test Y>

Separately, in Test Y, the first to fifth steps were carried out. The time of standing in the third step was changed from 30 minutes to 24 hours. In the fifth step, the second entering length L2 of the sample 11 was measured.

[0108]

The percentage ([L1-L2]/L1×100) of the change (L1-L2) from the first entering length L1 obtained by Test X to the second entering length L2 obtained by Test Y over the first entering length L1 was obtained.

[0109]

Table 1

				Example 1	Example 2	Example 3	Example 4	Example 5	Comp. Ex. 1	Comp. Ex. 2
Thermosetting resin composition	Resin component	Epoxy resin A	YSLV-80XY	4.23	-	3.18	5.21	4.23	4.23	4.23
		Epoxy resin B	EXA-4850-150	-	5.31	-	-	-	-	-
		Phenol resin A	LVR8210DL	2.31	1.23	-	2.84	2.31	2.31	2.31
		Phenol resin B	MEHC-7851SS	-	-	3.36	-	-	-	-
		Thermoplastic resin (80% MEK content)	HME-2006M	11.46	11.46	11.46	13.66	11.46	11.46	11.46
		Silane coupling agent	KBM-403	1.03	1.03	1.03	0.98	1.03	1.03	1.03
		Curing accelerator A (aliphatic urea-based)	U-CAT3513N	0.10	0.10	0.10	0.12	-	-	-
		Curing accelerator B (aromatic urea-based)	U-CAT3512T	-	-	-	-	0.10	-	-
		Curing accelerator C (imidazole-based)	2PHZ-PW	-	-	-	-	-	0.10	-
		Curing accelerator D (phosphorus-based)	TPP-K	-	-	-	-	-	-	0.10
	Filler component	Inorganic filler A	FB-8SMFD	44.67	44.67	44.67	42.62	44.67	44.67	44.67
		Inorganic filler B	SC220G-SMJ	24.06	24.06	24.06	22.95	24.06	24.06	24.06
		Layered silicate compound	S-BEN NX	-	-	-	1.19	-	-	-
		Pigment	Carbon black#20	0.31	0.31	0.31	0.37	0.31	0.31	0.31
	Solvent		MEK	11.84	11.84	11.84	10.07	11.84	11.84	11.84
Adhesive force			A0 [N] immediately after production	16.1	19.4	14.1	13.8	15.9	13.3	15.8
			A1 [N] after 24 hours have elapsed at 23°C and 50% RH	15.3	18.8	13.8	13.1	15.1	11.4	14.4
			Percentage of Change [%] (A0-A1)/A0×100	5	3	2	5	5	14	8
Calorific value in DSC			Immediately after production [mJ/mg]	19.82	21.5	19.1	23.8	21.3	23.48	23.53
			After heating at 150°C for 20 minutes [mJ/mg]	0.68	1.15	0.72	0.61	0.4	1	13.87
			Ratio	0.03	0.05	0.04	0.03	0.02	0.04	0.59
Entering length [μm]			3rd step: After 30 minutes have elapsed at 23°C and 50% RH (1st Entering length L1)	99	140	75	105	74	51	116
			3rd step: After 24 hours have elapsed at 23°C and 50% RH (2nd Entering length L2)	40	60	25	30	17	-60	66
			Percentage of Change [%] (L1-L2)/L1×100	60	57	67	71	77	218	43

[0110]

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.

Description of Reference Numerals

[0110]

1 sealable sheet

2 glass board

3, 8 bump

4 dummy chip

5 electronic element device

6 sealing body sheet

7 electronic element

9 mounting board

11 sample

CLAIMS

1. A sealable sheet comprising: an epoxy resin; a phenol resin; a urea-based curing accelerator; and an inorganic filler,

wherein a percentage ([A0-A1]/A0×100) of a change (A0-A1) from an adhesive force A0 that the sealable sheet has immediately after the sealable sheet is produced to an adhesive force A1 that the sealable sheet has after the sealable sheet is left to stand at 23℃ and 50% RH for 24 hours over the adhesive force A0 is 10% or less, and

wherein, with respect to a calorific value of the sealable sheet that is obtained by differential scanning calorimetry immediately after the sealable sheet is produced, a ratio of a calorific value of the sealable sheet that is obtained by differential scanning calorimetry after the sealable sheet is heated at 150℃ for 20 minutes is 0.10 or less.

2. The sealable sheet according to claim 1, wherein the urea-based curing accelerator is an aliphatic urea compound.

3. The sealable sheet according to claim 1 or 2, wherein a percentage ([L1-L2]/L1×100) of a change (L1-L2) from a first entering length L1 obtained by Test X to a second entering length L2 obtained by Test Y over the first entering length L1 is 90% or less.

<Test X>

The following first to fifth steps are carried out in this order.

The first step: A sample with a thickness of 200 μm is prepared from a sealable sheet immediately after the sealable sheet is produced. Separately, a mounting board including a glass board and nine dummy chips is prepared. The dummy chips are mounted on an upper surface of the glass board through bumps, have a size of 1 mm × 1 mm × a thickness of 200 μm, and are disposed in a row at

300 μm intervals.

The second step: The sample is pressed toward the glass board by vacuum flat-plate press at a temperature of 65℃, a degree of vacuum of 1.6 kPa or less, and a pressure of 0.2 MPa for 60 seconds.

The third step: The sample is brought into contact with upper surfaces of the dummy chips and left to stand under the conditions of 23℃ and 50% RH for 30 minutes.

The fourth step: The sample is heat-cured by heating the sample at 150℃ under atmospheric pressure for one hour.

The fifth step: The entering length L1 in which the sample enters a void defined between a lower surface of a dummy chip located at the center of the glass board and the upper surface of the glass board is measured.

<Test Y>

The above-described first to fifth steps are carried out. However, the time of the standing in the third step is changed from 30 minutes to 24 hours. In the fifth step, the second entering length L2 of the sample is measured.

4. An electronic element device comprising:

a sealing body sheet formed from the sealable sheet according to any one of claims 1 to 3; and

an electronic element sealed with the sealing body sheet.