EPOXY RESIN COMPOSITION, CURABLE RESIN COMPOSITION AND ELECTRONIC COMPONENT DEVICE

Technical Field

[0001] The present invention relates to an epoxy resin composition, a curable resin composition, and an electronic component device.

Background Art

[0002] As electronic devices become smaller, lighter, and more sophisticated in recent years, mounting density has been increasing. As a result, mainstream electronic component devices are changing from conventional pin insertion-type packages to surface mount-type packages such as integrated circuits (IC) and large scale integration (LSI) circuits.

[0003] Surface mount-type packages differ from conventional pin insertion-type packages in the mounting method. That is, when attaching pins to a wiring board, the conventional pin insertion-type packages are soldered from the back side of the wiring board after the pins are inserted into the wiring board, during which the packages were not directly exposed to high temperatures. On the other hand, in the surface mount-type packages, the entire electronic component device is processed with a solder bath, a reflow device or the like, and thus the packages are directly exposed to the soldering temperature (reflow temperature). As a result, in a case in which the packages absorb moisture, water due to the moisture absorption expands rapidly during the soldering, and the pressure of the generated vapor acts as peel stress, which may cause separation of inserts such as elements and lead frames from the sealing material, resulting in package cracks, poor electrical characteristics or the like. Therefore, it is desired that a sealing material having excellent adhesiveness to inserts and excellent solder heat resistance (reflow resistance) will be developed.

[0004] In order to meet such demands, various studies have been conducted on epoxy resins, which is a main material, from the viewpoint of low moisture absorption. In addition, from

the viewpoint of improving adhesiveness to inserts such as element lead frames, use of a silane coupling agents as a modifier for inorganic fillers contained in sealing materials is being studied. Specifically, use of an epoxy group-containing silane coupling agent or an amino group-containing silane coupling agent (see, for example, Patent Document 1), use of a sulfur atom-containing silane coupling agent (see, for example, Patent Document 2) and the like are being studied.

Citation List

Patent Documents

[0005] Patent Document 1: Japanese Patent Application Laid-Open (JP-A) No. H11-147939

Patent Document 2: Japanese Patent Application Laid-Open (JP-A) No. 2000-103940

SUMMARY OF INVENTION

Technical Problem

[0006] However, a curable resin composition that sufficiently satisfies reflow resistance at a practical level has not been obtained so far.

[0007] A first embodiment of the disclosure is directed to providing an epoxy resin composition having excellent reflow resistance and an electronic component device provided with an element sealed with the epoxy resin composition.

[0008] A second embodiment of the disclosure is directed to providing a curable resin composition having excellent reflow resistance and an electronic component device provided with an element sealed with the curable resin composition.

Solution to Problem

[0009] <1> An epoxy resin composition including: a monofunctional epoxy compound having one epoxy group in a molecule thereof; a polyfunctional epoxy compound having two or more epoxy groups in a molecule thereof; and a curing agent.

<2> The epoxy resin composition according to <1>, wherein the monofunctional epoxy compound includes a monofunctional epoxy compound having one alicyclic epoxy group in a molecule thereof.

<3> The epoxy resin composition according to <1>, wherein the monofunctional epoxy compound includes a monofunctional epoxy compound having one acyclic epoxy group and a phenyl group in a molecule thereof.

<4> The epoxy resin composition according to any one of <1> to <3>, wherein an amount of the monofunctional epoxy compound with respect to 100 parts by mass of the polyfunctional epoxy compound is from 1 part by mass to 30 parts by mass.

<5> The epoxy resin composition according to any one of <1> to <4>, further including a curing accelerator.

<6> The epoxy resin composition according to any one of <1> to <5>, further including an inorganic filler, wherein a content of the inorganic filler with respect to a total volume of the epoxy resin composition is from 70% by volume to 95% by volume.

<7> The epoxy resin composition according to any one of <1> to <6>, further including a silane coupling agent.

<8> An electronic component device, which is provided with an element sealed with the epoxy resin composition according to any one of <1> to <7>.

[0009a] Disclosed herein is an epoxy resin composition comprising:

a monofunctional epoxy compound having one epoxy group in a molecule thereof, the monofunctional epoxy compound having one alicyclic epoxy group in the molecule thereof;

a polyfunctional epoxy compound having two or more epoxy groups in a molecule thereof; and

a curing agent.

[0010]

<9> A curable resin composition including: a curable resin; a curing agent; and a compound having a (meth)acryloyl group.

<10> The curable resin composition according to <9>, wherein a molecular weight of the compound having a (meth)acryloyl group is from 180 to 320.

<11> The curable resin composition according to <9> or <10>, wherein the compound having a (meth)acryloyl group includes a (meth)acrylate ester compound.

<12> The curable resin composition according to any one of <9> to <11>, wherein the compound having a (meth)acryloyl group has an alicyclic structure.

<13> The curable resin composition according to any one of <9> to <12>, wherein the compound having a (meth)acryloyl group has an epoxy group.

<14> The curable resin composition according to any one of <9> to <13>, wherein the compound having a (meth)acryloyl group has an alicyclic epoxy group.

<15> The curable resin composition according to any one of <9> to <14>, further including a curing accelerator.

<16> The curable resin composition according to any one of <9> to <15>, further including an inorganic filler, wherein a content of the inorganic filler with respect to a total volume of the curable resin composition is from 70% by volume to 95% by volume.

<17> The curable resin composition according to any one of <9> to <16>, further including a coupling agent.

<18> An electronic component device, which is provided with an element sealed with the curable resin composition according to any one of <9> to <17>.

Advantageous Effects of Invention

[0011] According to the first embodiment of the disclosure, an epoxy resin composition having excellent reflow resistance and an electronic component device provided with an element sealed with the epoxy resin composition are provided.

[0012] According to the second embodiment of the disclosure, a curable resin composition having excellent reflow resistance and an electronic component device provided with an element sealed with the curable resin composition are provided.

DETAILED DESCRIPTION

[0012a] The first embodiment encompasses at least one solution to the above-described problems.

[0012b] The second embodiment encompasses at least one solution to the above-described problem.

[0013] Embodiments for carrying out the invention will be described in detail below. However, the invention is not limited to the following embodiments. In the following embodiments, components (including elements, steps, etc.) thereof are not essential unless otherwise specified. The same applies to numerical values and ranges, which do not limit the invention.

[0014] A numerical range indicated using "to" in the disclosure indicates a range including the numerical values described before and after "to" as the minimum value and the maximum value, respectively.

In the numerical range described in a stepwise manner in the disclosure, an upper limit value or a lower limit value described in one numerical range may be replaced with an upper limit value or a lower limit value of another numerical range described in a stepwise manner. In addition, in a numerical range described in the disclosure, the upper limit value or the lower limit value of the numerical range may be replaced with a value described in the Examples.

In the disclosure, a component may include a plurality of different substances corresponding thereto. In a case in which there are a plurality of substances corresponding to a component in a composition, the content or amount of the component means the total

content or amount of the plurality of substances present in the composition unless otherwise specified.

In the disclosure, particles corresponding to a component may include a plurality of different kinds of particles. In a case in which there are a plurality of different kinds of particles corresponding to a component in a composition, a particle diameter of the component means a value for a mixture of the plurality of different kinds of particles present in the composition unless otherwise specified.

In the disclosure, "(meth)acryloyl group" means at least one of acryloyl group or methacryloyl group, "(meth)acryloyloxy group" means at least one of acryloyloxy group or methacryloyloxy group, "(meth)acrylate" means at least one of acrylate or methacrylate, and "(meth)acrylate ester" means at least one of acrylate ester or methacrylate ester.

[0015] <Epoxy Resin Composition according to the First Embodiment>

An epoxy resin composition according to the first embodiment includes: (A) a monofunctional epoxy compound having one epoxy group in a molecule thereof; (B) a polyfunctional epoxy compound having two or more epoxy groups in a molecule thereof, and (C) a curing agent. The epoxy resin composition may further include other component(s) if necessary. Hereinafter, the (A) monofunctional epoxy compound having one epoxy group in a molecule thereof is also simply referred to as "monofunctional epoxy compound." In addition, the (B) polyfunctional epoxy compound having two or more epoxy groups in a molecule thereof is also simply referred to as "polyfunctional epoxy compound."

[0016] By including a monofunctional epoxy resin in an epoxy resin composition, an epoxy resin composition having excellent reflow resistance can be obtained. The detailed reason why an epoxy resin composition having excellent reflow resistance can be obtained by including a monofunctional epoxy compound therein is not necessarily clear. However, it is presumed that this is because an epoxy resin composition including a monofunctional epoxy compound has a lower crosslinking density in the cured product, and thus has a lower

high-temperature elastic modulus as compared to an epoxy resin composition that does not include a monofunctional epoxy compound as an epoxy resin. In general, from the viewpoint of peel stress during a reflow process, it is considered that the lower the high-temperature elastic modulus is, the less likely separation between inserts and a sealing material occurs, and that, therefore, the reflow resistance becomes better. It is also considered that, since the monofunctional epoxy compound, having an epoxy group, is excellent in reactivity, curability can be sufficiently maintained while crosslinking density is decreased and elastic modulus is decreased as described above.

[0017] (A) Monofunctional Epoxy Compound

The epoxy resin composition includes a monofunctional epoxy compound. For the monofunctional epoxy compound included in the epoxy resin composition, one kind of the monofunctional epoxy compound may be used singly, or two or more kinds thereof may be used in combination.

[0018] The monofunctional epoxy compound is not particularly limited as long as it is a compound having one epoxy group in a molecule thereof. The epoxy group contained in the monofunctional epoxy compound may be an alicyclic epoxy group, such as an epoxy cyclohexyl group, or an acyclic epoxy group, such as a glycidyl group. It is also possible to use a monofunctional epoxy compound having one alicyclic epoxy group in a molecule thereof (hereinafter also referred to as "monofunctional alicyclic epoxy compound") and a monofunctional epoxy compound having one acyclic epoxy group in a molecule thereof (hereinafter also referred to as "monofunctional acyclic epoxy compound") in combination. Hereinafter, (A1) a monofunctional alicyclic epoxy compound and (A2) a monofunctional acyclic epoxy compound will be respectively described.

[0019] (A1) Monofunctional Alicyclic Epoxy Compound

The monofunctional alicyclic epoxy compound is not particularly limited as long as it is a monofunctional epoxy compound having one alicyclic epoxy group in a molecule

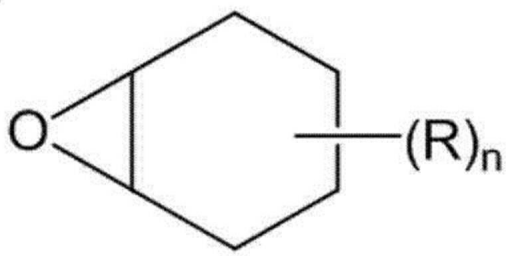
thereof. The alicyclic epoxy group is a group in which an oxygen atom is bonded to two adjacent carbon atoms which are part of a cycloaliphatic skeleton.

The number of carbon atoms of the cycloaliphatic skeleton is not particularly limited. The cycloaliphatic skeleton is, for example, preferably a 5-membered to 8-membered ring, more preferably a 5-membered or 6-membered ring, still more preferably a 6-membered ring.

The alicyclic structure may contain an unsaturated bond and may contain no unsaturated bond. The alicyclic structure may have a substituent and may have no substituent.

[0020] The monofunctional alicyclic epoxy compound is preferably a compound having an alicyclic epoxy group represented by the following Formula (a).

[0021]



(a)

[0022] In Formula (a), R represents a monovalent substituent containing no epoxy group, and n represents an integer from 0 to 10.

The monovalent substituent containing no epoxy group represented by R is not particularly limited, and examples thereof include a hydrocarbon group. Examples of the hydrocarbon group include an alkyl group, an alkenyl group, an alkynyl group, and an aryl group, which may have a substituent and may have no substituent. Of these, an alkyl group is preferable, and a substituted alkyl group is preferable. In a case in which a plurality of Rs are present, the plurality of Rs may be either the same or different.

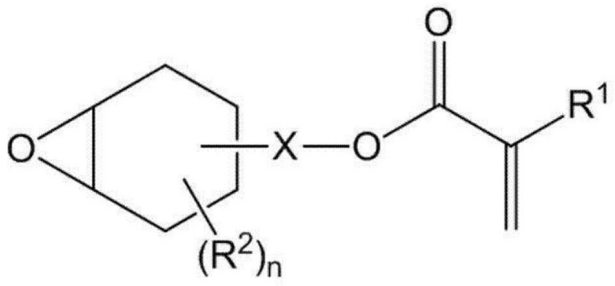
n is preferably from 1 to 4, more preferably 1.

[0023] In a case in which R is a substituted alkyl group, the substituent of the alkyl group is

preferably a group having an ethylenically unsaturated double bond, more preferably a group having a (meth)acryloyl group, still more preferably a group having a (meth)acryloyloxy group. In a case in which the monofunctional alicyclic epoxy compound has an ethylenically unsaturated double bond as the substituent of the alkyl group, the number of the ethylenically unsaturated double bond(s) in a molecule of the monofunctional alicyclic epoxy compound may be 1 or 2 or more.

[0024] Examples of the monofunctional alicyclic epoxy compound includes a compound represented by the following Formula (b).

[0025]



(b)

[0026] In Formula (b), R^1 represents a hydrogen atom or a methyl group. R^2 represents a monovalent substituent containing no epoxy group. X represents a single bond or a divalent linking group. n represents an integer from 0 to 9.

R^1 is preferably a methyl group.

The definition and preferred aspects of R^2 are the same as the definition and preferred aspects of R in Formula (a).

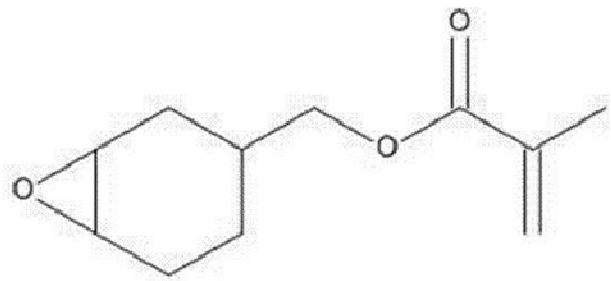
n is preferably from 0 to 3, more preferably 0.

In X, the divalent linking group is not particularly limited, and examples thereof include an aliphatic hydrocarbon. Examples of the aliphatic hydrocarbon include a linear or branched alkylene group. In a case in which X is an alkylene group, the number of carbon atom(s) contained in the alkylene group is not particularly limited, and is preferably from 1 to 10, more preferably from 1 to 3, still more preferably 1. The alkylene group may have a

substituent. Here, the number of carbon atom(s) contained in the alkylene group does not include the number of carbon atom(s) contained in a branch or a substituent.

[0027] Examples of the monofunctional alicyclic epoxy compound represented by Formula (b) include a compound represented by the following Formula (c).

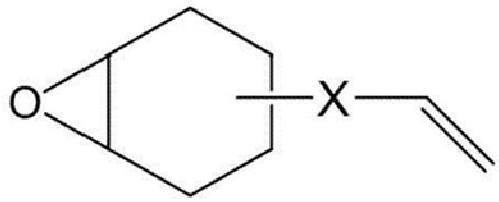
[0028]



(c)

[0029] Examples of the monofunctional alicyclic epoxy compound further includes a compound represented by the following Formula (d).

[0030]

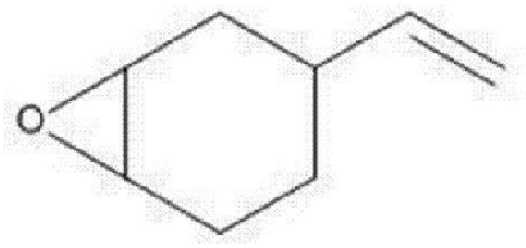


(d)

[0031] In Formula (d), X represents a single bond or a divalent linking group. In X, the divalent linking group is not particularly limited, and examples thereof include an aliphatic hydrocarbon. Examples of the aliphatic hydrocarbon include a linear or branched alkylene group. In a case in which X is an alkylene group, the number of carbon atom(s) contained in the alkylene group is not particularly limited, and may be from 1 to 10. The alkylene group may have a substituent. Here, the number of carbon atom(s) contained in the alkylene group does not include the number of carbon atom(s) contained in a branch or a substituent.

[0032] Examples of the monofunctional alicyclic epoxy compound represented by Formula (d) include a compound represented by the following Formula (e).

[0033]



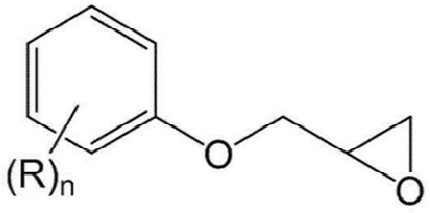
(e)

[0034] (A2) Monofunctional Acyclic Epoxy Compound

The monofunctional acyclic epoxy compound is not particularly limited as long as it is a monofunctional epoxy compound having one acyclic epoxy group in a molecule thereof. Examples of the acyclic epoxy group include a glycidyl group and a glycidyloxy group.

The monofunctional acyclic epoxy compound is preferably a monofunctional epoxy compound having one acyclic epoxy group and a phenyl group in a molecule thereof. Examples of the monofunctional epoxy compound having one acyclic epoxy group and a phenyl group in a molecule thereof include a compound having one glycidyl group and a phenyl group in a molecule thereof, and a compound having one glycidyloxy group and a phenyl group in a molecule thereof. In particular, the monofunctional acyclic epoxy compound is preferably a compound represented by the following Formula (f).

[0035]



(f)

[0036] In Formula (f), R represents a monovalent substituent containing no epoxy group, and n represents an integer from 0 to 5.

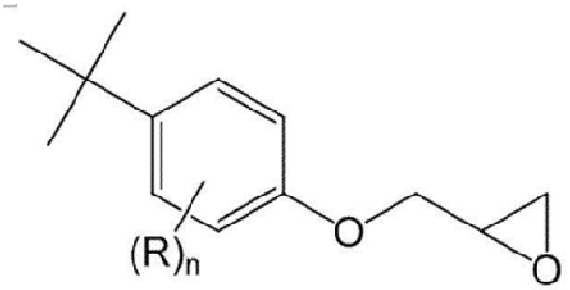
The monovalent substituent containing no epoxy group represented by R is not particularly limited, and examples thereof include a hydrocarbon group. Examples of the

hydrocarbon group include an alkyl group, an alkenyl group, an alkynyl group, and an aryl group. In a case in which R represents an alkyl group, the alkyl group may be linear or branched, may have a cyclic structure, and is preferably branched. In a case in which R represents an aryl group, the aryl group may have a substituent and may have no substituent. In a case in which a plurality of Rs are present, the plurality of Rs may be either the same or different. The substitution position of R is not particularly limited, and is preferably in the ortho or para position.

n is preferably from 0 to 3, more preferably 0.

[0037] Examples of the compound represented by Formula (f) include a compound represented by the following Formulas (g) and a compound represented by the following (h).

[0038]



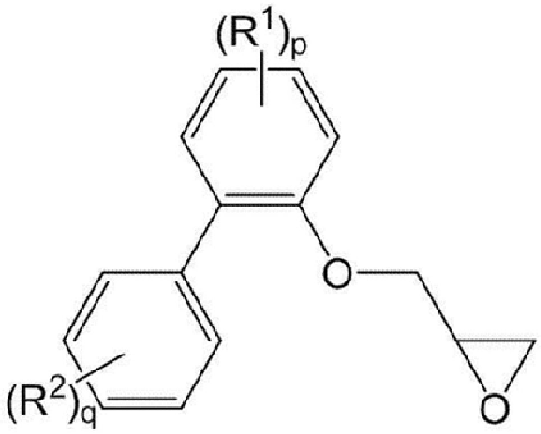
(g)

[0039] In Formula (g), R represents a monovalent substituent containing no epoxy group, and n represents an integer from 0 to 4.

The definition and preferred aspects of R in the compound represented by Formula (g) are the same as the definition and preferred aspects of R in the compound represented by Formula (f).

n is preferably from 0 to 2, more preferably 0.

[0040]



(h)

[0041] In Formula (h), R^1 and R^2 each independently represent a monovalent substituent containing no epoxy group, p represents an integer from 0 to 4, and q represents an integer from 0 to 5.

The definition and preferred aspects of the monovalent substituent containing no epoxy group represented by R^1 and R^2 are the same as the definition and preferred aspects of R in Formula (f).

p is preferably from 0 to 2, more preferably 0. q is preferably from 0 to 3, more-preferably 0.

[0042] As the monofunctional epoxy compound, a synthesized product may be used, or a commercially available product may be used. Examples of the monofunctional epoxy compound include, for example, 3,4-epoxycyclohexyl methyl methacrylate (e.g., trade name: CYCLOMER M100 manufactured by Daicel Corporation), 4-t-butylphenyl glycidyl ether (e.g., trade name: ED-509S manufactured by ADEKA), and 2-biphenylyl glycidyl ether (e.g., trade name: EX-142 manufactured by Nagase ChemteX Corporation).

[0043] The monofunctional epoxy compound may be in the solid state or in the liquid state, and preferably is in the liquid state, at room temperature (25°C).

[0044] The epoxy equivalent weight of the monofunctional epoxy compound is not particularly limited, and is preferably 500 g/eq or less, more preferably 300 g/eq or less, from the viewpoints of fluidity and reflow resistance.

[0045] The amount of the monofunctional epoxy compound with respect to 100 parts by mass of the polyfunctional epoxy compound described later is preferably from 1 part by mass to 30 parts by mass, more preferably 5 parts by mass to 30 parts by mass, still more preferably from 5 parts by mass to 25 parts by mass, particularly preferably from 10 parts by mass to 20 parts by mass. When the amount of the monofunctional epoxy compound with respect to 100 parts by mass of the polyfunctional epoxy compound is 1 part by mass or more, sufficient effects of reflow resistance tend to be achieved. When the amount is 30 parts by mass or less, there is a tendency that reduction in curability and releasability and generation of molding defects are suppressed, improving handleability.

[0046] In addition, the amount of the monofunctional epoxy compound with respect to 100 parts by mass of the epoxy resin composition is preferably from 0.05 parts by mass to 2 parts by mass, more preferably 0.1 parts by mass to 1 part by mass, from the viewpoints of curability and mold releasability.

[0047] (B) Polyfunctional Epoxy Compound

The polyfunctional epoxy compound has two or more epoxy groups in a molecule thereof. The polyfunctional epoxy compound may be one usually used for an epoxy resin composition, and is not particularly limited by type as long as it has two or more epoxy groups in a molecule thereof.

[0048] The polyfunctional epoxy compound may be an epoxy resin having two or more epoxy groups in a molecule thereof. Specific examples thereof include: a novolac-type epoxy resin (e.g., a phenol novolac-type epoxy resin or an orthocresol novolac-type epoxy resin) obtained by epoxidizing a novolac resin that is obtained by condensing or co-condensing at least one phenolic compound selected from the group consisting of phenol compounds such as phenol, cresol, xylenol, resorcin, catechol, bisphenol A, and bisphenol F and naphthol compounds such as α-naphthol, β-naphthol, and dihydroxynaphthalene, and an aliphatic aldehyde compound such as formaldehyde, acetaldehyde, or propionaldehyde, under

the presence of an acidic catalyst; a triphenylmethane-type epoxy resin obtained by epoxidizing a triphenylmethane-type phenol resin that is obtained by condensing or co-condensing the above-described phenolic compound and an aromatic aldehyde compound such as benzaldehyde or salicylaldehyde under the presence of an acidic catalyst; a copolymerized epoxy resin obtained by epoxidizing a novolac resin that is obtained by co-condensing the above-described phenol compound and a naphthol compound with an aldehyde compound under the presence of an acidic catalyst; a diphenylmethane-type epoxy resin that is a diglycidyl ether of bisphenol A, bisphenol F, or the like; a biphenyl-type epoxy resin that is a diglycidyl ether of alkyl-substituted or non-alkyl-substituted biphenol; a stilbene-type epoxy resin which is a diglycidyl ether of stilbene phenol compound; a sulfur atom-containing epoxy resin that is a diglycidyl ether of bisphenol S or the like; an epoxy resin that is a glycidyl ether of an alcohol such as butanediol, polyethylene glycol, or polypropylene glycol; a glycidyl ester-type epoxy resin that is a glycidyl ester of a polycarboxylic acid compound such as phthalic acid, isophthalic acid, and tetrahydrophthalic acid; a glycidyl amine-type epoxy resin obtained by substituting active hydrogen, bound to a nitrogen atom of aniline, diaminodiphenylmethane, isocyanuric acid or the like, with a glycidyl group; a dicyclopentadiene-type epoxy resin obtained by epoxidizing a co-condensed resin of dicyclopentadiene and a phenol compound; an alicyclic-type epoxy resin obtained by epoxidizing an olefin bond in a molecule, such as vinylcyclohexene diepoxide, 3,4-epoxy cyclohexyl methyl-3,4-epoxy cyclohexane carboxylate, and 2-(3,4-epoxy)cyclohexyl-5,5-spiro(3,4-epoxy)cyclohexane-m-dioxane; a para-xylylene-modified epoxy resin that is a glycidyl ether of a para-xylylene-modified phenol resin; a meta-xylylene-modified epoxy resin that is a glycidyl ether of a meta-xylylene-modified phenol resin; a terpene-modified epoxy resin that is a glycidyl ether of a terpene-modified phenol resin; a dicyclopentadiene-modified epoxy resin that is a glycidyl ether of a dicyclopentadiene-modified phenol resin; a cyclopentadiene-modified

epoxy resin that is a glycidyl ether of a cyclopentadiene-modified phenol resin; a polycyclic aromatic ring-modified epoxy resin that is a glycidyl ether of a polycyclic aromatic ring-modified phenol resin; a naphthalene-type epoxy resin that is a glycidyl ether of a naphthalene ring-containing phenol resin; a halogenated phenol novolac-type epoxy resin; a hydroquinone type epoxy resin; a trimethylolpropane-type epoxy resin; a linear aliphatic epoxy resin obtained by oxidizing an olefin bond with a peracid such as peracetic acid; and an aralkyl-type epoxy resin obtained by epoxidizing an aralkyl-type phenol resin such as a phenol aralkyl resin or a naphthol aralkyl resin. Further, examples of the epoxy resin also include an epoxidized product of a silicone resin, an epoxidized product of an acrylic resin, and the like. One kind of the epoxy resin may be used singly, or two or more kinds thereof may be used in combination.

[0049] Among the above-described epoxy resins, from the viewpoint of the balance between reflow resistance and fluidity, an epoxy resin selected from the group consisting of a biphenyl-type epoxy resin, a stilbene-type epoxy resin, a diphenylmethane-type epoxy resin, a sulfur atom-containing epoxy resin, a novolac-type epoxy resin, a dicyclopentadiene-type epoxy resin, a triphenylmethane-type epoxy resin, a copolymerized epoxy resin, and an aralkyl-type epoxy resin (referred to as "a specific epoxy resin") is preferable. One kind of the specific epoxy resin may be used singly, or two or more kinds thereof may be used in combination.

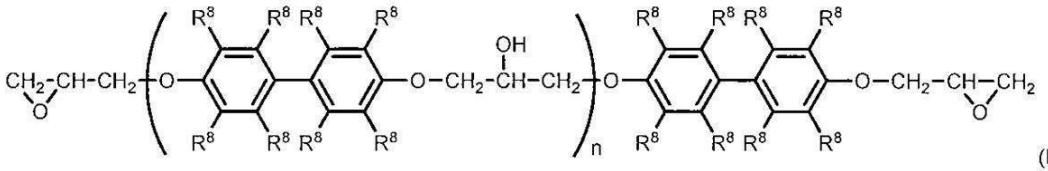
[0050] In a case in which the polyfunctional epoxy compound includes a specific epoxy resin, from the viewpoint of exerting properties of the specific epoxy resin, the content of the specific epoxy resin with respect to the entire polyfunctional epoxy compound is preferably 30% by mass or more, more preferably 50% by mass or more.

[0051] As the specific epoxy resin, from the viewpoint of fluidity, a biphenyl-type epoxy resin, a stilbene-type epoxy resin, a diphenylmethane-type epoxy resin, and a sulfur atom-containing epoxy resin are more preferable. From the viewpoint of heat resistance, a

dicyclopentadiene-type epoxy resin, a triphenylmethane-type epoxy resin, and an aralkyl-type epoxy resin are preferable. Preferred specific examples of the epoxy resin will be described below.

[0052] The biphenyl-type epoxy resin is not particularly limited as long as it is an epoxy resin having a biphenyl skeleton. For example, an epoxy resin represented by the following Formula (II) is preferable. As the epoxy resin represented by the following Formula (II), commercially available products include: YX-4000H (trade name, Mitsubishi Chemical Corporation), in which R^8s at positions 3, 3', 5, and 5' represent methyl groups, the positions substituted with the oxygen atoms being seen as positions 4 and 4', and the remaining R^8s represent hydrogen atoms; 4,4'-bis(2,3-epoxypropoxy)biphenyl, in which all R^8s represent hydrogen atoms; and YL-6121H (trade name, Mitsubishi Chemical Corporation), which is a mixture of an epoxy resin represented by Formula (II) in which all R^8s respectively represent hydrogen atoms and an epoxy resin represented by Formula (II) in which R^8s at positions 3, 3', 5, and 5' respectively represent methyl groups, the positions substituted with the oxygen atoms being seen as positions 4 and 4', and the remaining R^8s represent hydrogen atoms.

[0053]

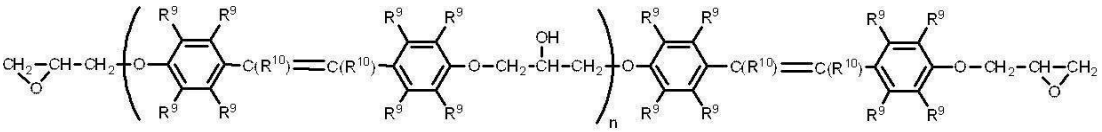


[0054] In Formula (II), R^8 represents a hydrogen atom, an alkyl group having 1 to 12 carbon atoms, or an aromatic group having from 4 to 18 carbon atoms, and respective R^8s may be either the same or different. n is an average value and represents a number from 0 to 10.

[0055] The stilbene-type epoxy resin is not particularly limited as long as it is an epoxy resin having a stilbene skeleton. For example, an epoxy resin represented by the following Formula (III) is preferable. As the epoxy resin represented by the following Formula (III),

commercially available products include ESLV-210 (trade name, Sumitomo Chemical Co., Ltd.), which is a mixture of an epoxy resin represented by Formula (III) in which R^9s at positions 3, 3', 5, and 5' represent methyl groups, the positions substituted with the oxygen atoms being seen as positions 4 and 4', the remaining R^9s represent hydrogen atoms, and all R^{10}s represent hydrogen atoms, and an epoxy resin represented by Formula (III) in which three of R^9s at positions 3, 3', 5 and 5' represent methyl groups and one represents a t-butyl group, the remaining R^9s represent hydrogen atoms, and all R^{10}s represent hydrogen atoms.

[0056]

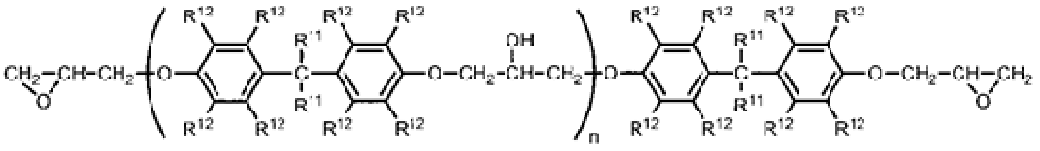


(III)

[0057] In Formula (III), R^9 and R^{10} represent a hydrogen atom or a monovalent organic group having 1 to 18 carbon atoms, and respective R^9s and R^{10}s may be either the same or different. n is an average value and represents a number from 0 to 10.

[0058] The diphenylmethane-type epoxy resin is not particularly limited as long as it is an epoxy resin having a diphenylmethane skeleton. For example, an epoxy resin represented by the following Formula (IV) is preferable. As the epoxy resin represented by the following Formula (IV), commercially available products include YSLV-80XY (trade name, NIPPON STEEL & SUMIKIN CHEMICAL CO., LTD.), in which all R^{11}s represent hydrogen atoms, R^{12}s at positions 3, 3', 5, and 5' represent methyl groups, the positions substituted with the oxygen atoms being seen as positions 4 and 4', and the remaining R^{12}s represent hydrogen atoms.

[0059]



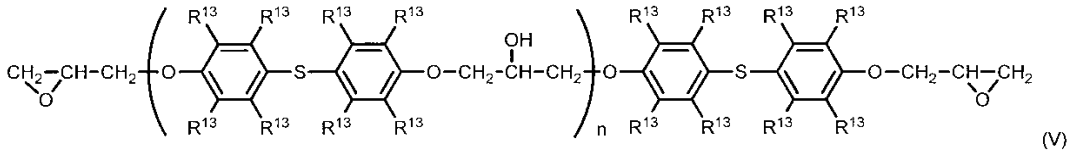
(IV)

[0060] In Formula (IV), R^{11} and R^{12} each represent a hydrogen atom or a monovalent

organic group having 1 to 18 carbon atoms, and respective R^{11}s and R^{12}s may be either the same or different. n is an average value and represents a number from 0 to 10.

[0061] The sulfur atom-containing epoxy resin is not particularly limited as long as it is an epoxy resin containing a sulfur atom. Examples include an epoxy resin represented by the following Formula (V). As the epoxy resin represented by the following Formula (V), commercially available products include YSLV-120TE (trade name, NIPPON STEEL & SUMIKIN CHEMICAL CO., LTD.), in which R^{13}s at positions 3 and 3' represent t-butyl groups and R^{13}s at positions 6 and 6' represent methyl groups, the positions substituted with the oxygen atoms being seen as positions 4 and 4', and the remaining R^{13}s represent hydrogen atoms.

[0062]

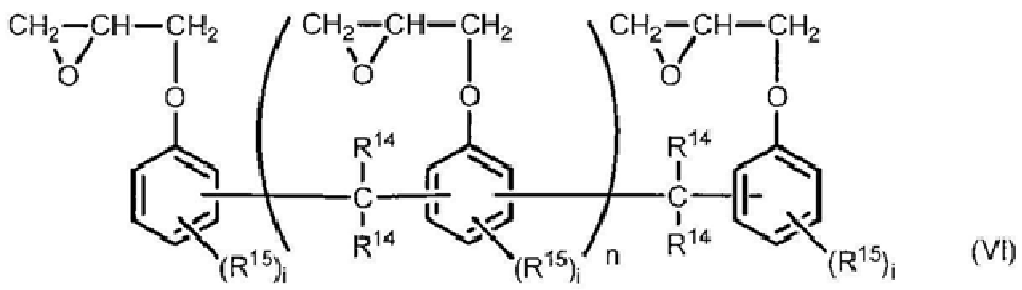


(V)

[0063] In Formula (V), R^{13} represents a hydrogen atom or a monovalent organic group having 1 to 18 carbon atoms, and respective R^{13}s may be either the same or different. n is an average value and represents a number from 0 to 10.

[0064] The novolac-type epoxy resin is not particularly limited as long as it is an epoxy resin obtained by epoxidizing a novolac-type phenol resin. For example, an epoxy resin obtained by epoxydizing a novolac-type phenol resin such as a phenol novolak resin, a cresol novolak resin, and a naphthol novolak resin, using a technique such as glycidyl etherification, is preferable, and an epoxy resin represented by the following Formula (VI) is more preferable. As the epoxy resin represented by the following Formula (VI), commercially available products include ESCN-190 and ESCN-195 (trade names, Sumitomo Chemical Co., Ltd.), in which all R^{14}s represent hydrogen atoms, R^{15}s represent a methyl group, and i = 1.

[0065]

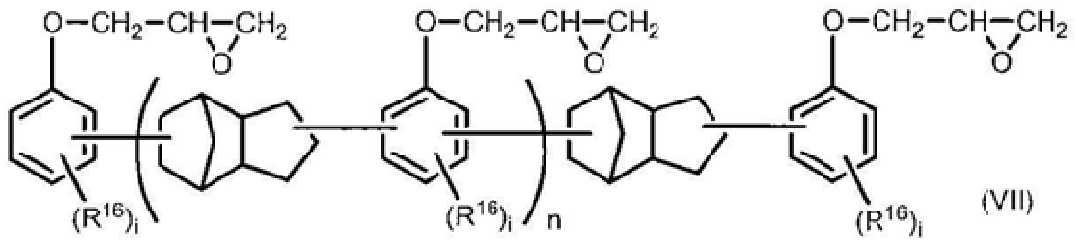


(VI)

[0066] In Formula (VI), R^{14} represents a hydrogen atom or a monovalent organic group having 1 to 18 carbon atoms, and respective R^{14}s may be either the same or different. R^{15} represents a monovalent organic group having 1 to18 carbon atoms, and respective R^{15}s may be either the same or different. i each independently represents an integer from 0 to 3. n is an average value and represents a number from 0 to 10.

[0067] The dicyclopentadiene-type epoxy resin is not particularly limited as long as it is an epoxy resin obtained by epoxydizing a compound having a dicyclopentadiene skeleton as a starting material. For example, an epoxy resin represented by the following Formula (VII) is preferable. As the epoxy resin represented by the following Formula (VII), commercially available products include HP-7200 (trade name, DIC CORPORATION), in which i = 0.

[0068]



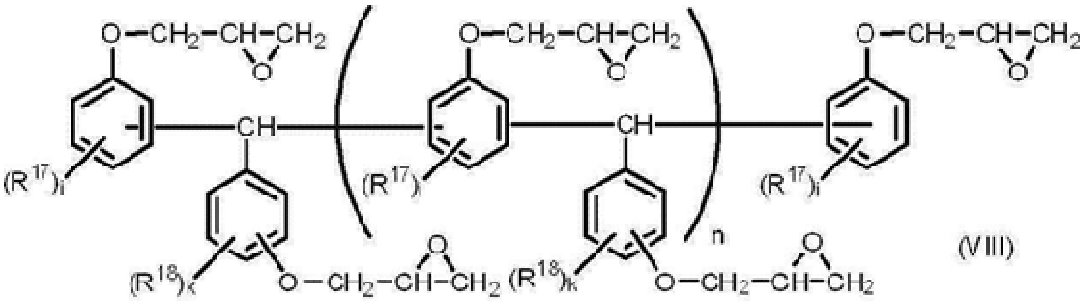
(VII)

[0069] In Formula (VII), R^{16} represents a monovalent organic group having 1 to18 carbon atoms, and respective R^{16}s may be either the same or different. i independently represents an integer of 0 to 3. n is an average value and represents a number from 0 to 10.

[0070] The triphenylmethane-type epoxy resin is not particularly limited as long as it is an epoxy resin obtained from a compound having a triphenylmethane skeleton as a starting

material. For example, an epoxy resin obtained by glycidyl etherification of a triphenylmethane-type phenol resin such as a novolac-type phenol resin of a compound having a triphenylmethane skeleton and a compound having a phenolic hydroxyl group is preferable, and an epoxy resin represented by the following Formula (VIII) is more preferable. As the epoxy resin represented by the following Formula (VIII), commercially available products include 1032H60 (trade name, Mitsubishi Chemical Corporation) and EPPN-502H (trade name, Nippon Kayaku Co., Ltd.), in which i is 0 and k is 0.

[0071]

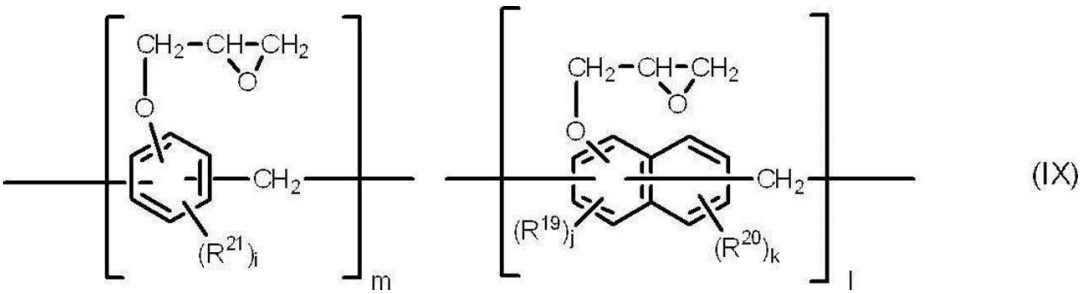


[0072] In Formula (VIII), R^{17} and R^{18} each represent a monovalent organic group having 1 to18 carbon atoms, and respective R^{17}s and R^{18}s may be either the same or different. i is independently an integer from 0 to 3, and k is independently an integer from 0 to 4. n is an average value and represents a number from 0 to 10.

[0073] A copolymerized epoxy resin, obtained by epoxidizing a novolac resin obtained from a naphthol compound and a phenol compound and an aldehyde compound, is not particularly limited as long as it is an epoxy resin starting materials of which include a compound having a naphthol skeleton and a compound having a phenol skeleton. For example, an epoxy resin obtained by glycidyl etherification of a novolac-type phenol resin obtained from a compound having a naphthol skeleton and a compound having a phenol skeleton is preferable, and an epoxy resin represented by the following Formula (IX) is more preferable. As the epoxy resin represented by the following Formula (IX), commercially available products include NC-7300 (trade name, Nippon Kayaku Co., Ltd.), in which R^{21} is a methyl group, i is 1, j is 0,

and k is 0.

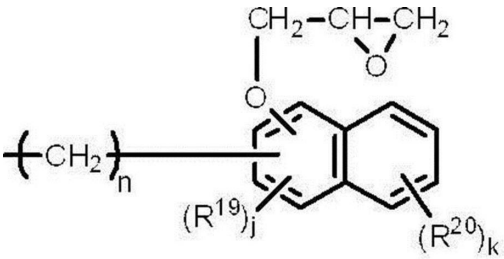
[0074]



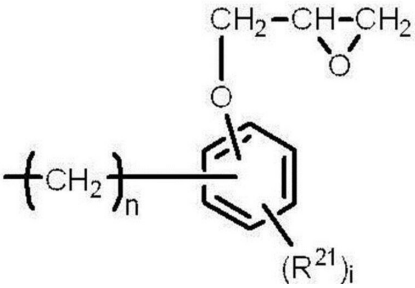
(IX)

[0075] In Formula (IX), R^{19} to R^{21} each represent a monovalent organic group having 1 to18 carbon atoms, and respective R^{19}s to R^{21}s may be either the same or different. i is each independently an integer from 0 to 3, j is each independently an integer from 0 to 2, and k is each independently an integer from 0 to 4. l and m are average values, respectively, each of which is a number from 0 to 10, and (l + m) is a number from 0 to 10. Each terminal of the epoxy resin represented by Formula (IX) is either the following Formula (IX-1) or (IX-2). In Formulas (IX-1) and (IX-2), R^{19} to R^{21}, i, j and k have the same definition of R^{19} to R^{21}, i, j, and k in Formula (IX). n is 1 (binding via a methylene group) or 0 (not binding via a methylene group).

[0076]



(IX-1)



(IX-2)

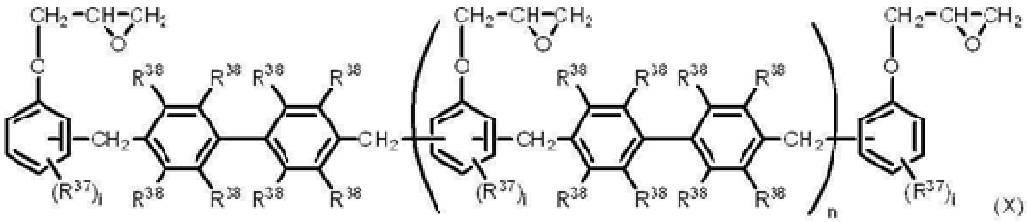
[0077] Examples of the epoxy resin represented by Formula (IX) include a random

copolymer containing l unit(s) of the constituent unit and m unit(s) of the constituent unit at random, an alternating copolymer containing l unit(s) of the constituent unit and m unit(s) of the constituent unit alternately, a copolymer containing l unit(s) of the constituent unit and m unit(s) of the constituent unit regularly, and a block copolymer containing l unit(s) of the constituent unit and m unit(s) of the constituent unit in blocks. One of these may be used singly, or two or more kinds thereof may be used in combination.

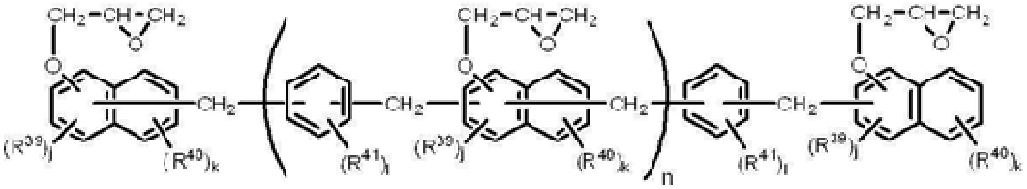
[0078] The aralkyl-type epoxy resin is not particularly limited as long as it is an epoxy resin starting materials of which include a phenol resin synthesized from: at least one selected from the group consisting of a phenol compound, such as phenol and cresol, and a naphthol compound, such as naphthol and dimethylnaphthol; and dimethoxyparaxylene, bis(methoxymethyl)biphenyl, or a derivative thereof. For example, an epoxy resin obtained by glycidyl etherification of a phenol resin synthesized from: at least one selected from the group consisting of a phenol compound, such as phenol and cresol, and a naphthol compound, such as naphthol and dimethylnaphthol; and dimethoxyparaxylene, bis(methoxymethyl)biphenyl, or a derivative thereof is preferable, and an epoxy resin represented by the following Formula (X) or (XI) is more preferable.

[0079] As the epoxy resin represented by the following Formula (X), commercially available products include NC-3000S (trade name, Nippon Kayaku Co., Ltd.), in which i is 0 and R^{38} is a hydrogen atom; and CER-3000 (trade name, Nippon Kayaku Co., Ltd.), which is a mixture of an epoxy resin in which i is 0 and R^{38} is a hydrogen atom and an epoxy resin represented by Formula (II) in which each R^{8} is a hydrogen atom, at a mass ratio of 80:20. Further, as the epoxy resin represented by the following Formula (XI), commercially available products include ESN-175 (trade name, NIPPON STEEL & SUMIKIN CHEMICAL CO., LTD.), in which i is 0, j is 0, and k is 0.

[0080]



(X)



(XI)

[0081] In Formulas (X) and (XI), R^{38} represents a hydrogen atom or a monovalent organic group having 1 to18 carbon atoms, and respective R^{38}s may be either the same or different. R^{37} and R^{39} to R^{41} each represent a monovalent organic group having 1 to18 carbon atoms, and respective R^{37}s and R^{39}s to R^{41}s may be either the same or different. i is each independently an integer from 0 to 3, j is each independently an integer from 0 to 2, k is each independently an integer from 0 to 4, and l is each independently an integer from 0 to 6. n is an average value and each independently a number from 0 to 10.

[0082] Regarding R^{8} to R^{21} and R^{37} to R^{41} in Formulas (II) to (XI) described above, the expression "respective ... may be either the same or different" means that, for example, all 8 to 88 R^{8}s in Formula (II) may be either the same or different. Similarly, regarding R^{9} to R^{21} and R^{37} to R^{41}, this expression means that the corresponding number of the respective substituent symbols in the Formulas may be either the same or different. Further, R^{8} to R^{21} and R^{37} to R^{41} may be either the same or different. For example, all of R^{9}s and R^{10}s may be the same and may be different.

Further, the organic group having 1 to18 carbon atoms in Formulas (III) to (XI) is preferably an alkyl group or an aryl group.

[0083] In Formulas (II) to (XI) described above, n is an average value and is preferably from 0 to 10 independently. When n is 10 or less, there is a tendency that the melt viscosity of the resin component does not become excessively high, and the viscosity during melt molding of

the epoxy resin composition decreases, which reduces the occurrences of insufficient filling, deformation of bonding wire (gold wire connecting elements and leads), and the like. n is preferably set in a range of from 0 to 4.

[0084] Preferable examples of the polyfunctional epoxy compound that may be used for the epoxy resin composition have been described above in reference to Formulas (II) to (XI). More specific preferable examples of the polyfunctional epoxy compound include 4,4'-bis(2,3-epoxypropoxy)-3,3',5,5'-tetramethylbiphenyl, from the viewpoint of reflow resistance, and 4,4'-bis(2,3-epoxypropoxy)-biphenyl, from the viewpoints of moldability and heat resistance.

[0085] In addition to the above, it is preferable to use a dihydroanthracene-type epoxy resin, which is a diglycidyl ether of an alkyl-substituted, aromatic ring-substituted, or unsubstituted anthracene (e.g., trade name: YX-8800 manufactured by Mitsubishi Chemical Corporation) from the viewpoint of low warpage, or a methoxynaphthalene-type epoxy resin (e.g., trade name: HP-5000 manufactured by DIC CORPORATION) from the viewpoint of the balance of reflow resistance, curability, and fluidity.

[0086] The epoxy equivalent weight of the polyfunctional epoxy compound is not particularly limited. The epoxy equivalent weight of the polyfunctional epoxy compound is preferably from 100 g/eq to 1000 g/eq, more preferably from 150 g/eq to 500 g/eq from the viewpoint of the balance of properties such as moldability, reflow resistance, and electric reliability.

[0087] The softening point or melting point of the polyfunctional epoxy compound is not particularly limited. It is preferably from 40°C to 180°C from the viewpoints of moldability and reflow resistance, more preferably from 50°C to 130°C from the viewpoint of handleability upon preparation of the epoxy resin composition.

[0088] (C) Curing Agent

The curing agent may be one commonly used for an epoxy resin composition, and

there is no particular limitation.

Examples of the curing agent include a phenol curing agent, an amine curing agent, an acid anhydride curing agent, a polymercaptan curing agent, a polyaminoamide curing agent, an isocyanate curing agent, and a blocked isocyanate curing agent. Of these, at least one selected from the group consisting of a phenol curing agent, an amine curing agent, and an acid anhydride curing agent is preferable, and a phenol curing agent is more preferable.

[0089] Examples of the phenol curing agent include, for example, a phenol resin having two or more phenolic hydroxyl groups in a molecule thereof and a polyphenol compound. Specific examples thereof include: a polyphenol compound such as resorcin, catechol, bisphenol A, bisphenol F, and substituted or unsubstituted biphenol; a novolac-type phenol resin obtained by condensing or co-condensing at least one phenolic compound selected from the group consisting of a phenol compound such as phenol, cresol, xylenol, resorcin, catechol, bisphenol A, bisphenol F, phenylphenol, and aminophenol, and a naphthol compound such as α-naphthol, β-naphthol, and dihydroxynaphthalene, with an aldehyde compound such as formaldehyde, acetaldehyde, propionaldehyde, benzaldehyde, or salicylaldehyde, under the presence of an acidic catalyst; an aralkyl-type phenol resin, such as a phenol aralkyl resin and a naphthol aralkyl resin, synthesized from the above-described phenolic compound and dimethoxyparaxylene, bis(methoxymethyl)biphenyl and the like; a para-xylylene and/or meta-xylylene-modified phenol resin; a melamine-modified phenol resin; terpene-modified phenol resin; a dicyclopentadiene-type phenol resin and a dicyclopentadiene-type naphthol resin synthesized via copolymerization of the above-described phenolic compound and dicyclopentadiene; a cyclopentadiene-modified phenol resin; a polycyclic aromatic ring-modified phenol resin; a biphenyl-type phenol resin; a triphenylmethane-type phenol resin obtained by condensing or co-condensing the above-described phenolic compound and an aromatic aldehyde compound such as benzaldehyde and salicylaldehyde under the presence of an acidic catalyst; and a phenol resin obtained by copolymerizing two or more

kinds thereof. One kind of the phenol curing agent may be used singly, or two or more kinds thereof may be used in combination.

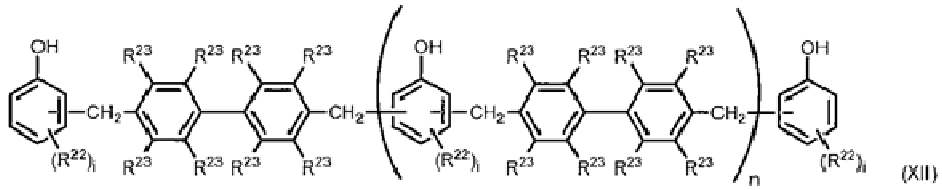
[0090] As the phenol curing agent, from the viewpoint of reflow resistance, it is preferable to use at least one selected from the group consisting of: a copolymerized phenol resin of an aralkyl-type phenol resin, a dicyclopentadiene-type phenol resin, a triphenylmethane-type phenol resin, or a benzaldehyde-type phenol resin and an aralkyl-type phenol resin; and a novolac-type phenol resin (these are referred to as "a specific phenol curing agent"). One kind of the specific phenol curing agent may be used singly, or two or more kinds thereof may be used in combination.

[0091] In a case in which the curing agent includes a specific phenol curing agent, the content of the specific phenol curing agent with respect to the entire curing agent is preferably 30% by mass or more, more preferably 50% by mass or more, from the viewpoint of exerting the properties thereof.

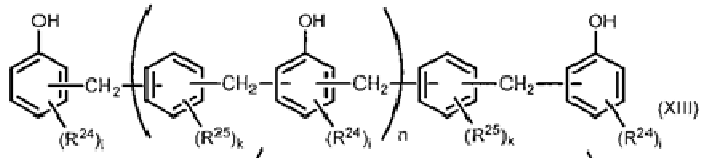
[0092] Examples of the aralkyl-type phenol resin include a phenol aralkyl resin and a naphthol aralkyl resin synthesized from a phenolic compound and dimethoxyparaxylene, bis (methoxymethyl) biphenyl, or the like. The aralkyl-type phenol resin may further be copolymerized with a different phenol resin. Examples of such a copolymerized aralkyl-type phenol resin include a copolymerized phenol resin of a benzaldehyde-type phenol resin and an aralkyl-type phenol resin, a copolymerized phenol resin of a salicylaldehyde-type phenol resin and an aralkyl-type phenol resin, and a copolymerized phenol resin of a novolac-type phenol resin and an aralkyl-type phenol resin.

[0093] The aralkyl-type phenol resin is not particularly limited as long as it is a phenol resin synthesized from: at least one selected from the group consisting of a phenol compound and a naphthol compound; and dimethoxyparaxylene, bis(methoxymethyl)biphenyl, or a derivative thereof. For example, a phenol resin such as those represented by the following Formulas (XII) to (XIV) are preferable.

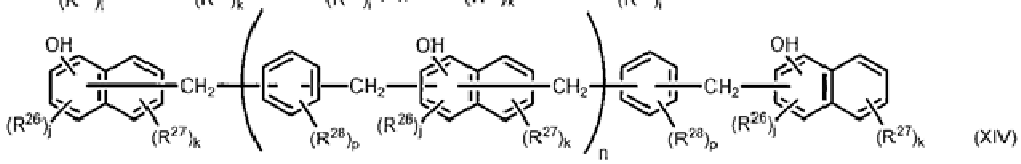
[0094]



(XII)



(XIII)



(XIV)

[0095] In Formulas (XII) and (XIV), R^{23} represents a hydrogen atom or a monovalent organic group having 1 to18 carbon atoms, and respective R^{23}s may be either the same or different. R^{22}, R^{24}, R^{25}, and R^{28} each represent a monovalent organic group having 1 to18 carbon atoms, and respective R^{22}s, R^{24}s, R^{25}s, and R^{28}s may be either the same or different. R^{26} and R^{27} each represent a hydroxyl group or a monovalent organic group having 1 to18 carbon atoms, and respective R^{26}s and R^{27}s may be either the same or different. i is each independently an integer from 0 to 3, j is each independently an integer from 0 to 2, k is each independently an integer from 0 to 4, and p is each independently an integer from 0 to 4. n is an average value and each independently a number from 0 to 10.

[0096] As the phenol resin represented by Formula (XII), commercially available products include MEH-7851 (trade name, Meiwa Plastic Industries, Ltd.) in which i is 0 and each R^{23} is a hydrogen atom.

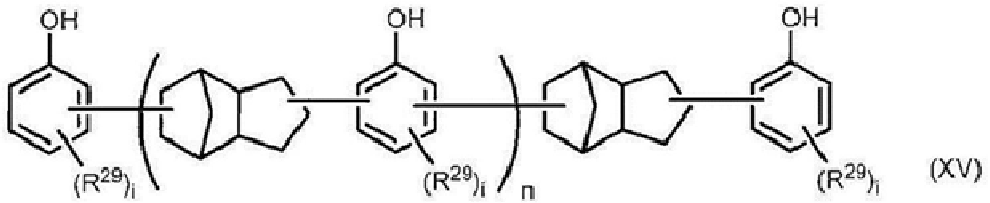
[0097] As the phenol resin represented by Formula (XIII), commercially available products include XL-225, XLC (trade names, Mitsui Chemicals, Inc.), and MEH-7800 (trade name, Meiwa Plastic Industries, Ltd.), in which i is 0 and k is 0.

[0098] As the phenol resin represented by Formula (XIV), commercially available products

include SN-170 (trade name, NIPPON STEEL & SUMIKIN CHEMICAL CO., LTD.), in which j is 0, k is 0, and p is 0, and SN-395 (trade name, NIPPON STEEL & SUMIKIN CHEMICAL CO., LTD.), in which j is 0, k is 1, R^{27} is a hydroxyl group, and p is 0.

[0099] The dicyclopentadiene-type phenol resin is not particularly limited as long as it is a phenol resin obtained by using a compound having a dicyclopentadiene skeleton as a starting material. For example, a phenol resin represented by the following Formula (XV) is preferable. As the phenol resin represented by the following Formula (XV), commercially available products include DPP (trade name, Nippon Oil Corporation), in which i is 0.

[0100]

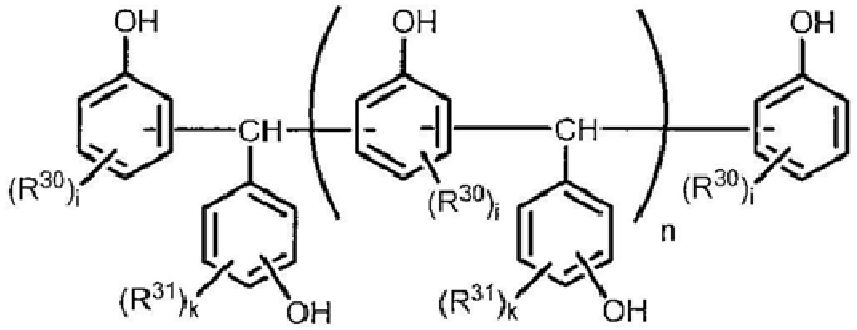


[0101] In Formula (XV), R^{29} represents a monovalent organic group having 1 to18 carbon atoms, and respective R^{29}s may be either the same or different. i independently represents an integer from 0 to 3. n is an average value and represents a number from 0 to 10.

[0102] The triphenylmethane-type phenol resin is not particularly limited as long as it is a phenol resin obtained by using a compound having a triphenylmethane skeleton as a starting material. For example, a phenol resin represented by the following Formula (XVI) are preferable.

[0103] As the phenol resin represented by Formula (XVI), commercially available products include MEH-7500 (trade name, Meiwa Plastic Industries, Ltd.) in which i is 0 and k is 0.

[0104]



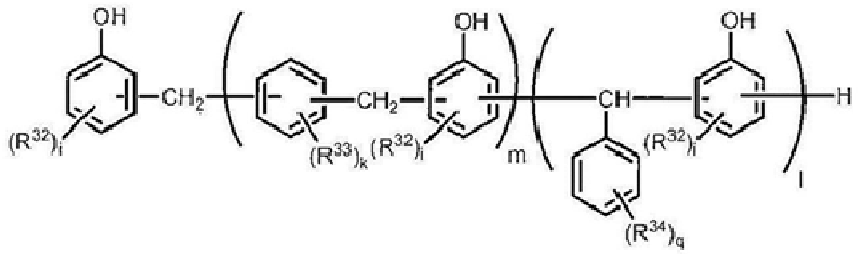
(XVI)

[0105] In Formula (XVI), R^{30} and R^{31} each represent a monovalent organic group having 1 to18 carbon atoms, and respective R^{30}s and R^{31}s may be either the same or different. i is each independently an integer from 0 to 3, and k is each independently an integer from 0 to 4. n is an average value and is a number from 0 to 10.

[0106] A copolymerized phenol resin of a benzaldehyde-type phenol resin and an aralkyl-type phenol resin is not particularly limited as long as it is a copolymerized phenol resin of: a phenol resin obtained by using a compound having a benzaldehyde skeleton as a starting material; and an aralkyl-type phenol resin. For example, a phenol resin represented by the following Formula (XVII) is preferable.

[0107] As the phenol resin represented by the following Formula (XVII), commercially available products include HE-510 (trade name, Air Water Chemical Inc.), in which i is 0, k is 0, and q is 0.

[0108]



(XVII)

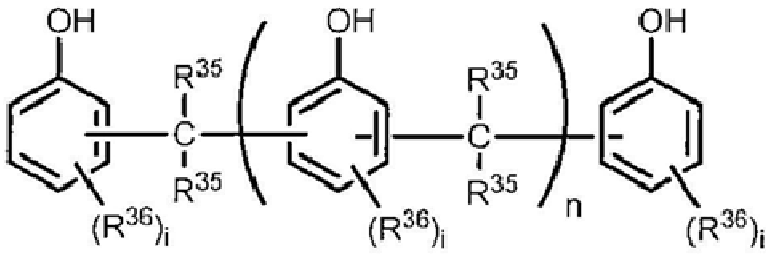
[0109] In Formula (XVII), R^{32} to R^{34} each represent a monovalent organic group having 1 to18 carbon atoms, and respective R^{32}s to R^{34}s may be either the same or different. i is each

independently an integer from 0 to 3, k is each independently an integer from 0 to 4, and q is each independently an integer from 0 to 5. l and m are each independently an average value, and are each independently a number from 0 to 11. Note that the sum of l and m is a number from 1 to11.

[0110] The novolac-type phenol resin is not particularly limited as long as it is a phenol resin obtained by condensing or co-condensing at least one phenolic compound selected from the group consisting of a phenol compound and a naphthol compound with an aldehyde compound under the presence of an acidic catalyst. For example, a phenol resin represented by the following Formula (XVIII) is preferable.

[0111] As the phenol resin represented by the following Formula (XVIII), commercially available products include TAMANOL 758 and 759 (trade name, ARAKAWA CHEMICAL INDUSTRIES, LTD.), and HP-850N (trade name, Hitachi Chemical Co., Ltd.), in which i is 0 and each R^{35} is a hydrogen atom.

[0112]



(XVIII)

[0113] In Formula (XVIII), R^{35} represents a hydrogen atom or a monovalent organic group-having 1 to18 carbon atoms, and respective R^{35}s may be either the same or different. R^{36} represents a monovalent organic group having 1 to18 carbon atoms, and respective R^{36}s may be either the same or different. i independently represents an integer from 0 to 3. n is an average value and represents a number from 0 to 10.

[0114] Regarding R^{22} to R^{36} in Formulas (XII) to (XVIII) described above, the expression "respective ... may be either the same or different" means that, for example, all R^{22}s in

Formula (XII), the number of which being i, may be either the same as or different from others. Similarly, regarding R^{23} to R^{36}, this expression means that the corresponding number of the respective substituent symbols in the Formulas may be either the same or different. In addition, R^{22} to R^{36} may be either the same or different. For example, all of R^{22}s and R^{23}s may be the same and may be different, and all of R^{30}s and R^{31}s may be the same and may be different.

[0115] In Formulas (XII) to (XVIII) described above, n is preferably from 0 to 10. When n is 10 or less, the melt viscosity of the resin component does not become excessively high, and the viscosity during melt molding of the epoxy resin composition decreases, which reduces the occurrences of insufficient filling, deformation of bonding wire (gold wire connecting elements and leads), and the like. The average n in a molecule is preferably set in a range of from 0 to 4.

[0116] The functional group equivalent weight of the curing agent (or the hydroxyl group equivalent weight in the case of a phenol curing agent) is not particularly limited, and is preferably from 70 g/eq to 1000 g/eq, more preferably from 80 g/eq to 500 g/eq, from the viewpoint of the balance of various properties such as moldability, reflow resistance, and electric reliability.

[0117] The softening point or melting point of the curing agent is not particularly limited, and is preferably from 40°C to 180°C from the viewpoints of moldability and reflow resistance, more preferably from 50°C to 130°C from the viewpoint of handleability upon production of the epoxy resin composition.

[0118] The ratio of the number of equivalent of the (C) curing agent to the total number of equivalent of the (A) monofunctional epoxy compound and the (B) polyfunctional epoxy compound, that is, the ratio of the number of functional groups of the curing agent to the number of epoxy groups of the monofunctional epoxy compound and the polyfunctional epoxy compound (the number of functional groups of the curing agent/the number of epoxy

groups of the monofunctional epoxy compound and the polyfunctional epoxy compound), is not particularly limited. In order to reduce respective unreacted components, the ratio is preferably from 0.5 to 2.0, more preferably from 0.6 to 1.3. From the viewpoints of moldability and reflow resistance, the ratio is still more preferably from 0.8 to 1.2.

[0119] (D) Curing Accelerator

The epoxy resin composition may include a curing accelerator. The curing accelerator may be one commonly used for an epoxy resin composition, and is not particularly limited. Specific examples thereof include: a cycloamidine compound such as 1,8-diazabicyclo[5.4.0]undec-7-ene, 1,5-diazabicyclo[4.3.0]non-5-en, and 5,6-dibutylamino-1,8-diazabicyclo[5.4.0]undec-7-ene; a compound that is intramolecularly polarized, which is obtained by adding a compound having a π bond such as maleic anhydride, a quinone compound such as 1,4-benzoquinone, 2,5-toluquinone, 1,4-naphthoquinone, 2,3-dimethylbenzoquinone, 2,6-dimethylbenzoquinone, 2,3-dimethoxy-5-methyl-1,4-benzoquinone, 2,3-dimethoxy-1,4-benzoquinone, or phenyl-1,4-benzoquinone, diazophenylmethane and a phenol resin, to the above cycloamidine compounds; a tertiary amine compound such as benzyldimethylamine, triethanolamine, dimethylamino ethanol, and tris(dimethylaminomethyl)phenol; a derivative of the tertiary amine compound; an imidazole compound such as 2-methylimidazole, 2-phenylimidazole, 2-phenyl-4-methylimidazole, and 2-heptadecylimidazole; a derivative of the imidazole compound; an organic phosphine compound such as tributyl phosphine, methyldiphenylphosphine, triphenylphosphine, tris(4-methylphenyl)phosphine, diphenylphosphine, and phenylphosphine; a phosphorus compound that is intramolecularly polarized, which is obtained by adding a compound having a π bond such as maleic anhydride, the above quinone compound, diazophenylmethane and a phenol resin to the phosphine compound; a tetra-substituted phosphonium-tetra-substituted borate such as tetraphenylphosphonium tetraphenyl borate, tetraphenylphosphonium ethyl triphenyl borate,

and tetrabutylphosphonium tetrabutyl borate; a tetraphenyl boron salt such as 2-ethyl-4-methylimidazole·tetraphenyl borate, N-methylmorpholine·tetraphenyl borate, and the like; and a derivative of the tetra-substituted phosphonium-tetra-substituted borate or the tetraphenyl boron salts. One kind of the curing accelerator may be used singly, or two or more kinds thereof may be used in combination.

[0120] In particular, as a curing accelerator, an adduct of a tertiary phosphine compound and a quinone compound is preferable, and an adduct of triphenylphosphine and benzoquinone or an adduct of tributylphosphine and benzoquinone is more preferable, from the viewpoints of curability and fluidity. From the viewpoint of storage stability, an adduct of a cycloamidine compound and a phenol resin is preferable, and a novolac-type phenol resin salt of diazabicycloundecene is more preferable. The content of the above-exemplified curing accelerator with respect to the total amount of the curing accelerator is preferably 60% by mass or more, more preferably 80% by mass or more.

[0121] In a case in which an adduct of a tertiary phosphine compound and a quinone compound is used as a curing accelerator, the total content of the adduct of a tertiary phosphine compound and a quinone compound with respect to the total amount of the curing accelerator is preferably 60% by mass or more, more preferably 80% by mass or more.

[0122] A tertiary phosphine used for the adduct of a tertiary phosphine and a quinone compound is not particularly limited. Specific examples thereof include a tertiary phosphine having an aryl group such as tributylphosphine, dibutylphenylphosphine, butyldiphenylphosphine, ethyldiphenylphosphine, triphenylphosphine, tris(4-methylphenyl)phosphine, tris(4-ethylphenyl)phosphine, tris(4-propylphenyl)phosphine, tris(4-butylphenyl)phosphine, tris(isopropylphenyl)phosphine, tris(t-butylphenyl)phosphine, tris(2,4-dimethylphenyl)phosphine, tris(2,6-dimethylphenyl)phosphine, tris(2,4,6-trimethylphenyl)phosphine, tris(2,6-dimethyl-4-ethoxyphenyl)phosphine, tris(4-methoxyphenyl)phosphine, and tris(4-ethoxyphenyl)phosphine. From the viewpoint

of moldability, triphenylphosphine and tributylphosphine are preferable.

[0123] Further, a quinone compound used for the adduct of a tertiary phosphine and a quinone compound is not particularly limited. Specific examples thereof include o-benzoquinone, p-benzoquinone, diphenoquinone, 1,4-naphthoquinone, and anthraquinone. From the viewpoint of moisture resistance or storage stability, p-benzoquinone is preferable.

[0124] The content of the curing accelerator is not particularly limited as long as the effect of curing acceleration can be achieved. The content of the curing accelerator with respect to the total of 100 parts by mass of the (A) monofunctional epoxy compound, the (B) polyfunctional epoxy compound, and the (C) curing agent (hereinafter also referred to as "a resin component") is preferably from 0.1 parts by mass to 10 parts by mass, more preferably 0.3 parts by mass to 5 parts by mass. When the content of the curing accelerator is 0.1 parts by mass or more, there is a tendency that the time can be shortened. When it is 10 parts by mass or less, there is a tendency that an excessive increase in the curing speed can be suppressed such that a more favorable molded product can be obtained.

[0125] (E) Inorganic Filler

The epoxy resin composition may include an inorganic filler. In particular, in a case in which the epoxy resin composition is used as a sealing material for a semiconductor package, the epoxy resin composition preferably includes an inorganic filler.

[0126] The type of the inorganic filler is not particularly limited. Specific examples include an inorganic material such as fused silica, crystalline silica, glass, alumina, calcium carbonate, zirconium silicate, calcium silicate, silicon nitride, aluminum nitride, boron nitride, beryllia, zirconia, zircon, forsterite, steatite, spinel, mullite, titania, talc, clay, and mica. An inorganic filler having flame-retardant property may also be used. Examples of the inorganic filler having flame-retardant property include a complex metal hydroxide such as aluminum hydroxide, magnesium hydroxide, and magnesium-zinc complex hydroxide, and zinc borate. In particular, fused silica is preferable from the viewpoint of reducing the

coefficient of thermal expansion, and alumina is preferable from the viewpoint of high thermal conductivity. One kind of the inorganic filler may be used singly, or two or more kinds thereof may be used in combination. Examples of the form of the inorganic filler include powder, beads that are formed in a spherical shape from powder, and fibers.

[0127] The average particle diameter of the inorganic filler is not particularly limited. In particular, from the viewpoint of moldability, the average particle diameter of the inorganic filler is preferably from 5 μm to 50 μm, more preferably from 10 μm to 30 μm. The average particle diameter of the inorganic filler is measured as a volume average particle diameter using a laser diffraction particle size distribution analyzer.

[0128] The volume average particle diameter of the inorganic filler in the epoxy resin composition or a cured product thereof can be measured by a known method. For example, an inorganic filler is extracted from an epoxy resin composition or a cured product thereof using an organic solvent, nitric acid, aqua regia, or the like, and the extract is thoroughly dispersed using an ultrasonic disperser or the like, thereby preparing a dispersion. The volume average particle diameter of the inorganic filler can be measured using the dispersion based on the volume-based particle size distribution measured by a laser diffraction particle size distribution analyzer. Alternatively, the volume average particle diameter of the inorganic filler can be measured based on the volume-based particle size distribution obtained by scanning electron microscope observation of a cross-section obtained by embedding a cured product in a transparent epoxy resin or the like and polishing the cured product. Further, the volume average particle diameter of the inorganic filler can also be measured by continuously performing two-dimensional cross-sectional observation of a cured product using an FIB device (focused ion beam SEM) and performing three-dimensional structural analysis.

[0129] From the viewpoint of fluidity of the epoxy resin composition, the particle shape of the inorganic filler is preferably spherical rather than polyhedral, and the particle size

distribution of the inorganic filler is preferably distributed in a broad range.

[0130] From the viewpoint of low warpage of the epoxy resin composition, an inorganic filler having a large coefficient of thermal expansion is preferable. For example, in a case in which the inorganic filler in the epoxy resin composition is silica, crystalline silica is more preferable than fused silica.

[0131] The specific surface area of the inorganic filler is not particularly limited. In particular, the specific surface area of the inorganic filler is preferably from 0.5 m^2/g to 12 m^2/g, more preferably from 1 m^2/g to 5 m^2/g, from the viewpoints of moldability and strength. The specific surface area of the inorganic filler is measured by nitrogen adsorption capacity at 77K according to JIS Z 8830: 2013.

[0132] The content of the inorganic filler is not particularly limited. The content of the inorganic filler in the epoxy resin composition is preferably from 70% by mass to 95% by mass from the viewpoint of improving flame retardance, moldability, hygroscopicity, and strength, and reducing the coefficient of thermal expansion, more preferably from 85% by mass to 95% by mass from the viewpoints of improving hygroscopicity and reducing the coefficient of thermal expansion. When the content of the inorganic filler is 70% by mass or more, flame retardance and reflow resistance tend to be improved. When the content of the inorganic filler is 95% by mass or less, fluidity tends to become excellent. In addition, the content of the inorganic filler with respect to the total volume of the epoxy resin composition is preferably from 70% by volume to 95% by volume, more preferably from 75% by volume to 95% by volume, and still more preferably from 80% by volume to 90% by volume. When the content of the inorganic filler is 70% by volume or more, flame retardance and reflow resistance tend to be improved. When it is 95% by volume or less, fluidity tends to become excellent.

[0133] [Various Additives]

The epoxy resin composition may include various additives such as a coupling agent,

an ion exchanger, an adhesion promoter, a mold release agent, a flame retardant, a colorant, a thermoplastic resin, and a stress relaxation agent exemplified below, in addition to the above-described components. The epoxy resin composition may include various additives known in the art as well as the additives exemplified below, as necessary.

[0134] (Coupling Agent)

In a case in which the epoxy resin composition includes an inorganic filler, the epoxy resin composition may include a coupling agent in order to improve adhesiveness between the resin component and the inorganic filler. Examples of the coupling agent include known coupling agents which include: a silane compound such as an epoxy silane, a mercapto silane, an amino silane, an alkyl silane, an ureido silane, and a vinyl silane; titanium compounds; aluminum chelate compounds; and aluminum/zirconium compounds. One kind of the coupling agent may be used singly, or two or more kinds thereof may be used in combination.

In particular, it is preferable that the coupling agent include a silane coupling agent. Types and numbers of alkoxysilyl group(s) and organic functional group(s) other than the alkoxysilyl group(s) in the silane coupling agent may be selected as necessary.

[0135] In a case in which the epoxy resin composition includes a coupling agent, the amount of the coupling agent with respect to 100 parts by mass of the inorganic filler is preferably from 0.05 parts by mass to 5 parts by mass, more preferably from 0.1 parts by mass to 2.5 parts by mass. In a case in which the amount of the coupling agent with respect to 100 parts by mass of the inorganic filler is 0.05 parts by mass or more, adhesiveness to a frame tends to be improved. In a case in which the amount of the coupling agent with respect to 100 parts by mass of the inorganic filler is 5 parts by mass or less, package moldability tends to be improved.

[0136] (Ion Exchanger)

The epoxy resin composition may include an ion exchanger. In particular, in a case in which the epoxy resin composition is used as a molding material for sealing, the epoxy

resin composition may include an ion exchanger as necessary from the viewpoint of improving moisture resistance and **high temperature endurance** of an electronic component device including an element to be sealed. The ion exchanger is not particularly limited, and a conventionally known ion exchanger can be used. Specifically, examples include a hydrotalcite compound and a hydrous oxide of at least one element selected from the group consisting of magnesium, aluminum, titanium, zirconium, and bismuth. One kind of the ion exchanger may be used singly, or two or more kinds thereof may be used in combination. Of these, a hydrotalcite represented by the following Formula (A) is preferable.

[0137] $Mg_{(1-X)}Al_X(OH)_2(CO_3)_{X/2}\cdot mH_2O$ (A)

($0 < X \leq 0.5$, and m is a positive number.)

[0138] In a case in which the epoxy resin composition includes an ion exchanger, the content thereof is not particularly limited as long as it is sufficient for capturing ions such as halogen ions. For example, the content of the ion exchanger with respect to 100 parts by mass of the resin component is preferably from 0.1 parts by mass to 30 parts by mass, more preferably from 1 part by mass to 5 parts by mass.

[0139] (Adhesion Promoter)

The epoxy resin composition may include an adhesion promoter, as necessary, in order to further improve adhesiveness. Specific examples thereof include: a derivative of imidazole, triazole, tetrazole, triazine, and the like; anthranilic acid, gallic acid, malonic acid, malic acid, maleic acid, aminophenol, quinoline, and the like and a derivative thereof; an aliphatic acid amide compound; a dithiocarbamate; and a thiadiazole derivative. One kind of the adhesion promoter may be used singly, or two or more kinds thereof may be used in combination.

[0140] (Mold Release Agent)

The epoxy resin composition may include a mold release agent from the viewpoint of achieving favorable mold releasability from a mold upon molding. The mold release agent

is not particularly limited, and a conventionally known mold release agent may be used. Specific examples thereof include a higher fatty acid such as carnauba wax, montanic acid, and stearic acid, a higher fatty acid metal salt, an ester wax such as montanic acid ester, and a polyolefin wax of oxidized polyethylene and non-oxidized polyethylene. One kind of the mold release agent may be used singly, or two or more kinds thereof may be used in combination.

[0141] In a case in which the epoxy resin composition includes a mold release agent, the amount thereof with respect to 100 parts by mass of the resin component is preferably from 0.01 parts by mass to 10 parts by mass, more preferably from 0.1 parts by mass to 5 parts by mass. When the amount of the mold release agent with respect to 100 parts by mass of the resin component is 0.01 parts by mass or more, sufficient mold releasability tends to be achieved. When the amount of the mold release agent is 10 parts by mass or less, more favorable adhesiveness tends to be achieved.

[0142] (Flame Retardant)

The epoxy resin composition may include a conventionally known flame retardant as necessary from the viewpoint of improving flame retardance of the composition. Specific examples thereof include: a brominated epoxy resin; an inorganic substance such as antimony trioxide, red phosphorus, aluminum hydroxide, magnesium hydroxide, and zinc oxide; red phosphorus coated with a heat-curable resin such as a phenol resin; a phosphorus compound such as a phosphate ester; a nitrogen-containing compound such as melamine, a melamine derivative, a melamine-modified phenol resin, a compound having a triazine ring, a cyanuric acid derivative, and an isocyanuric acid derivative; a phosphorus and nitrogen-containing compound such as cyclophosphazene; and a complex metal hydroxide represented by the following Formula (II).

[0143] $p(M1_aO_b)\cdot q(M2_cO_d)\cdot m(H_2O)$ Formula (II)

[0144] In Formula (II), M1 and M2 each represent a metal element different from each other,

and a, b, c, d, p, q, and m each represent a positive number.

[0145] M1 and M2 in Formula (II) are not particularly limited as long as they are metal elements different from each other. From the viewpoint of flame retardance, it is preferable that M1 is selected from the metal elements of period 3, the alkaline earth metal elements of Group IIA, and the metal elements belonging to Group IVB, IIB, VIII, IB, IIIA, or IVA, and M2 is selected from the transition metal elements of Group IIIB to IIB, and it is more preferable that M1 is selected from magnesium, calcium, aluminum, tin, titanium, iron, cobalt, nickel, copper, or zinc, and M2 is selected from iron, cobalt, nickel, copper, or zinc. From the viewpoint of fluidity, it is preferable that M1 is magnesium, M2 is zinc or nickel. The ratio of p and q (p/q) is not particularly limited, and the ratio is preferably from 1/99 to 1/1. The metal elements are classified based on a long-period periodic table in which representative elements fall into subgroup A and transition elements fall into subgroup B (source: Kyoritsu Shuppan Co., Ltd. "ENCYCLOPAEDIA CHIMICA 4", February 15, 1987, 30th edition (reduced-size edition)).

[0146] Examples of the flame retardant other than the above include a compound containing a metal element such as zinc oxide, zinc stannate, zinc borate, iron oxide, molybdenum oxide, zinc molybdate, and dicyclopentadienyl iron. One kind of the flame retardant may be used singly, or two or more kinds thereof may be used in combination.

[0147] In a case in which the epoxy resin composition includes a flame retardant, the amount of the flame retardant is not particularly limited. In particular, the amount of the flame retardant with respect to 100 parts by mass of the (A) epoxy resin is preferably from 1 part by mass to 30 parts by mass, more preferably from 2 parts by mass to 15 parts by mass.

[0148] (Colorant)

The epoxy resin composition may further include a colorant. Examples of the colorant include known colorants such as carbon black, organic dyes, organic pigments, titanium oxide, red lead, and colcothar. The amount of the colorant may be selected as

necessary, depending on the purposes. One kind of the colorant may be used singly, or two or more kinds thereof may be used in combination.

[0149] (Thermoplastic Resin)

The epoxy resin composition may include, as necessary, a thermoplastic resin such as: a polyphenylene ether; and an indene oligomer that is a copolymerized resin of an indene such as indene and alkylindene, a styrene such as styrene or alkylstyrene, and a phenol. One kind of the thermoplastic resin may be used singly, or two or more kinds thereof may be used in combination.

[0150] (Stress Relaxation Agent)

The epoxy resin composition may include a stress relaxation agent such as silicone oil and silicone rubber particles. By including a stress relaxation agent in the epoxy resin composition, warpage deformation of a package and occurrences of package cracking can be further reduced. Examples of the stress relaxation agent include generally used known stress relaxation agents (flexible agents). Specific examples thereof include a thermoplastic elastomer such as a silicone-based thermoplastic elastomer, a styrene-based thermoplastic elastomer, an olefin-based thermoplastic elastomer, an urethane-based thermoplastic elastomer, a polyester-based thermoplastic elastomer, a polyether-based thermoplastic elastomer, a polyamide-based thermoplastic elastomer, and a polybutadiene-based thermoplastic elastomer; rubber particles of, for example, natural rubber (NR), acrylonitrile-butadiene rubber (NBR), acrylic rubber, urethane rubber, or silicone powder; and rubber particles having a core-shell structure of, for example, methyl methacrylate-styrene-butadiene copolymer (MBS), methyl methacrylate-silicone copolymer, or methyl methacrylate-butyl acrylate copolymer. One kind of the stress relaxation agent may be used singly, or two or more kinds thereof may be used in combination. In particular, a silicone-based stress relaxation agent is preferable. Examples of the silicone-based stress relaxation agent include those having an epoxy group, those having an amino group, and a

polyether-modified product thereof.

[0151] (Method of Preparing Epoxy Resin Composition)

A method of preparing the epoxy resin composition is not particularly limited. General method thereof include a method in which components are mixed thoroughly using a mixer or the like, melt-kneaded using a mixing roll, an extruder, or the like, and then cooled, and the resulting mixture is pulverized. More specific examples include a method in which the above-described components are mixed and stirred, kneaded using a kneader, a roll, an extruder, or the like preheated to 70°C to 140°C, cooled, and pulverized.

[0152] The epoxy resin composition is preferably in the solid state at room temperature and normal pressure (e.g., 25°C, 1 atm). In a case in which the epoxy resin composition is in the solid state, the form thereof is not particularly limited, and may be in the form of powder, porticles, tablets, or the like. In a case in which the epoxy resin composition is in the form of tablets, the size and the mass thereof are preferably the size and the mass that meet the molding conditions of the package, from the viewpoint of handleability.

[0153] <Electronic Component Device according to the First Embodiment>

An electronic component device according to the first embodiment is provided with an element sealed with the above-described epoxy resin composition.

Examples of the electronic component device include an electronic component device in which an element part obtained by mounting an element (e.g., an active element such as a semiconductor chip, a transistor, a diode, or a thyristor, and a passive element such as a capacitor, a resistor, or a coil) on a support member, such as a lead frame, a pre-wired tape carrier, a wiring board, a glass, a silicon wafer, or an organic substrate, is sealed with the above-described epoxy resin composition.

More specific examples include: general resin-sealing type IC, such as DIP (Dual Inline Package), PLCC (Plastic Leaded Chip Carrier), QFP (Quad Flat Package), SOP (Small Outline Package), SOJ (Small Outline J-lead package), TSOP (Thin Small Outline Package),

and TQFP (Thin Quad Flat Package), having a structure formed by fixing the element on a lead frame, connecting the terminal part of the element, such as a bonding pad, to the lead part by wire bonding, bumping or the like, and performing a sealing process by transfer molding or the like using the epoxy resin composition; TCP (Tape Carrier Package) having a structure formed by sealing, with the epoxy resin composition, an element connected to a tape carrier with bumps; COB (Chip On Board) module, hybrid IC, multi-chip module, and the like, having a structure formed by sealing, with the epoxy resin composition, an element connected to the wiring formed on a support member by wire bonding, flip-chip bonding, soldering, or the like; and BGA (Ball Grid Array), CSP (Chip Size Package), MCP (Multi Chip Package), and the like, having a structure formed by mounting an element on a surface of a support member, at the rear surface of which terminals for connecting the wiring board have been formed, connecting the element and the wiring formed on the support member by bumping or wire bonding, and then sealing the elements with the epoxy resin composition. Further, the epoxy resin composition may be suitably used for a printed wiring board.

[0154] Examples of the method of sealing an electronic component device with the epoxy resin composition include low pressure transfer molding, injection molding, compression molding and the like. In particular, a low pressure transfer molding is common.

[0155] <Curable Resin Composition according to the Second Embodiment>

A curable resin composition according to the second embodiment includes: (A') a curable resin; (B') a curing agent; and (C') a compound having a (meth)acryloyl group. The curable resin composition may further include other component(s) as necessary. Hereinafter, (C') a compound having a (meth)acryloyl group is sometimes referred to as a "specific acrylic compound."

[0156] When the curable resin composition includes a specific acrylic compound, a curable resin composition having excellent reflow resistance can be obtained. Although the reason for this is not entirely clear, it is presumed that an acrylic resin does not have polar group(s)

after reactions and exhibits low water absorption, thereby obtaining excellent reflow resistance.

[0157] (C') Specific Acrylic Compound

The curable resin composition includes a specific acrylic compound. One kind of the specific acrylic compound may be used singly, or two or more kinds thereof may be used in combination. The specific acrylic compound is not particularly limited as long as it is a compound having a (meth)acryloyl group.

[0158] The number of (meth)acryloyl group(s) included in the specific acrylic compound is not limited, and is preferably from 1 to 10, more preferably from 1 to 3, still more preferably 1 or 2. In this study, the specific acrylic compound preferably has a (meth)acryloyl group, more preferably a (meth)acryloyloxy group. In this study, the specific acrylic compound preferably includes a (meth)acrylate ester compound.

[0159] Specific examples of the specific acrylic compound include tricyclodecane dimethanol di(meth)acrylate, dicyclopentenyloxyethyl(meth)acrylate, dicyclopentenyloxyethyl(meth)acrylate, dicyclopentanyl(meth)acrylate, dicyclopentanyl(meth)acrylate, polyethylene glycol #200 di(meth)acrylate, and 3,4-epoxycyclohexylmethyl(meth)acrylate.

[0160] The specific acrylic compound may be either in the solid state or in the liquid state at room temperature (25°C), and is preferably in the liquid state.

[0161] The molecular weight of the specific acrylic compound is not particularly limited. The molecular weight of the specific acrylic compound is preferably from 180 to 320, more preferably from 180 to 270, still more preferably from 190 to 220, from the viewpoints of fluidity and reflow resistance.

[0162] In one embodiment, the specific acrylic compound may have an alicyclic structure. The alicyclic structure may contain an unsaturated bond and may contain no unsaturated bond. The alicyclic structure may have a substituent and may have no substituent. For example,

the specific acrylic compound may have a dicyclopentadiene skeleton.

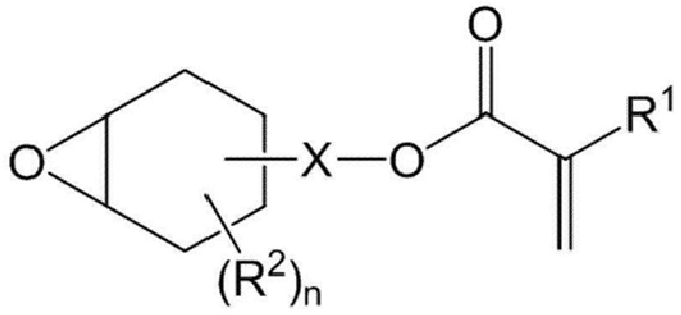
[0163] In another embodiment, the specific acrylic compound may have an epoxy group. In a case in which the specific acrylic compound has an epoxy group, curability tends to be sufficiently maintained.

In a case in which the specific acrylic compound has an epoxy group, the number of epoxy group(s) contained in the specific acrylic compound is not limited, and is preferably 1 from the viewpoint of reflow resistance.

[0164] In particular, from the viewpoints of reflow resistance, curability and the like, the specific acrylic compound preferably has an alicyclic epoxy group. In a case in which the specific acrylic compound has an alicyclic epoxy group, the alicyclic epoxy group is a group in which an oxygen atom is bound to two carbon atoms adjacent to each other that are part of a cycloaliphatic skeleton. The number of carbon atoms in the cycloaliphatic skeleton forming the alicyclic epoxy group is not particularly limited. The cycloaliphatic skeleton is, for example, preferably a 5-membered to 8-membered ring, more preferably a 5-membered or 6-membered ring, still more preferably a 6-membered ring.

[0165] Examples of the specific acrylic compound having an alicyclic epoxy group include a compound represented by the following Formula (a).

[0166]



(a)

[0167] In Formula (a), R^1 represents a hydrogen atom or a methyl group. R^2 represents a monovalent substituent. X represents a single bond or a divalent linking group. n represents an integer from 0 to 9.

R^1 is preferably a methyl group.

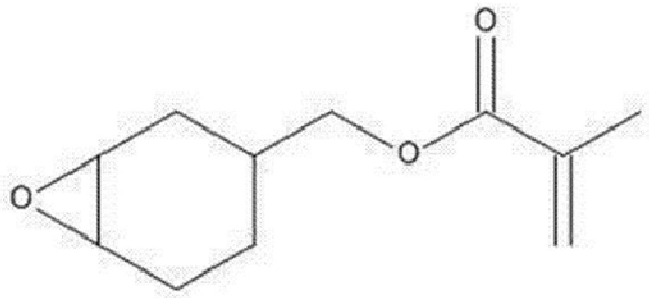
The monovalent substituent represented by R^2 is not particularly limited, and examples thereof include an alkyl group, an alkenyl group, an alkynyl group, and an aryl group. In a case in which a plurality of R^2s are present, the plurality of R^2s may be either the same or different.

The divalent linking group represented by X is not particularly limited, and examples thereof include an aliphatic hydrocarbon. Examples of the aliphatic hydrocarbon include a linear or branched alkylene group. In a case in which X is an alkylene group, the number of carbon atom(s) contained in the alkylene group is not particularly limited, and is preferably from 1 to 10, more preferably from 1 to 3, still more preferably 1. The alkylene group may have an optional substituent. Here, the number of carbon atom(s) contained in the alkylene group does not include the number of carbon atom(s) contained in a branch or a substituent.

n is preferably from 0 to 3, more preferably 0.

[0168] Examples of the specific acrylic compound having an alicyclic epoxy group represented by Formula (a) include a compound represented by the following Formula (b) (3,4-epoxycyclohexyl methyl methacrylate).

[0169]



(b)

[0170] As the specific acrylic compound, a synthesized product may be used, or a commercially available product may be used. Examples of the commercially available product include 3,4-epoxycyclohexyl methyl methacrylate (trade name: CYCLOMER M100 manufactured by Daicel Corporation)

[0171] The amount of the specific acrylic compound with respect to 100 parts by mass of the curable resin is preferably from 1 part by mass to 30 parts by mass, more preferably from 5 parts by mass to 25 parts by mass, still more preferably from 5 parts by mass to 20 parts by mass, particularly preferably from 10 parts by mass to 20 parts by mass. When the amount of the specific acrylic compound with respect to 100 parts by mass of the curable resin is 1 part by mass or more, there is a tendency that excellent fluidity can be achieved and sufficient effects of reflow resistance can be obtained. When the amount of the specific acrylic compound is 30 parts by mass or less, there is a tendency that reduction in curability and mold releasability and the generation of molding defects are suppressed, and that handleability is improved.

[0172] In addition, the amount of the specific acrylic compound with respect to 100 parts by mass of the curable resin composition is preferably from 0.05 parts by mass to 2 parts by mass, more preferably 0.1 parts by mass to 1 part by mass, from the viewpoints of curability and mold releasability.

[0173] Whether or not the specific acrylic compound is contained in the curable resin composition can be confirmed by, for example, analyzing a solvent extract with a mass spectrum.

[0174] (A') Curable Resin

The curable resin composition includes a curable resin. The curable resin is not particularly limited, and may be heat-curable and may be photo-curable. The curable resin is preferably heat-curable. The curable resin may be a resin that can be cured by self-polymerization and may be a resin that can be cured by reaction with a curing agent, a crosslinking agent, or the like.

[0175] A functional group in charge of the reaction of the curable resin is not particularly limited, and examples thereof include an acyclic epoxy group such as a glycidyl group, an alicyclic epoxy group, a hydroxyl group, a carboxy group, an amino group, an acryloyl group,

and an isocyanate group. From the viewpoint of the balance of properties for a sealing material, a curable resin having a cyclic ether group is preferable, and a curable resin having an epoxy group (epoxy resin) is more preferable.

[0176] In a case in which the curable resin is an epoxy resin, the type of the epoxy resin is not particularly limited as long as it has two or more epoxy groups in a molecule thereof. Details of the epoxy resin in a case in which the curable resin is an epoxy resin are the same as those of the (B) polyfunctional epoxy compound according to the first embodiment. Here, the term "polyfunctional epoxy compound" in the description of the (B) polyfunctional epoxy resin according to the first embodiment is replaced with "epoxy resin", and the term "epoxy resin composition" is replaced with "curable resin composition."

[0177] The functional group equivalent weight of the curable resin is not particularly limited. The functional group equivalent weight of the curable resin is preferably from 100 g/eq to 1000 g/eq, more preferably from 150 g/eq to 500 g/eq, from the viewpoint of the balance of properties such as moldability, reflow resistance, and electric reliability.

[0178] The softening point or melting point of the curable resin is not particularly limited. The softening point or melting point of the curable resin is preferably from 40°C to 180°C from the viewpoints of moldability and reflow resistance, and is more preferably from 50°C to 130°C from the viewpoint of handleability upon preparation of the curable resin composition.

[0179] (B') Curing Agent

The curable resin composition includes a curing agent. The type of the curing agent is not particularly limited, and may be selected depending on the type of the curable resin, desirable properties of the curable resin composition, and the like.

Examples of the curing agent in a case in which the curable resin is an epoxy resin include the curing agents described above in relation to the curing agent according to the first embodiment. Details of the curing agent in the case in which the curable resin is an epoxy

resin are the same as the details of the curing agent according to the first embodiment.

Here, the term "epoxy resin composition" in the description of the curing agent according to the first embodiment is replaced with "curable resin composition."

Further, the description "the ratio of the number of equivalent of the (C) curing agent to the total number of equivalent of the (A) monofunctional epoxy compound and the (B) polyfunctional epoxy compound, that is, the ratio of the number of functional groups of the curing agent to the number of epoxy groups of the monofunctional epoxy compound and the polyfunctional epoxy compound (the number of functional groups of the curing agent/the number of epoxy groups of the monofunctional epoxy compound and the polyfunctional epoxy compound)" in the description of the curing agent according to the first embodiment is replaced with "the equivalent ratio of the curing agent and the curable resin, that is, the ratio of the number of functional groups of the curing agent to the number of functional groups of the curable resin (the number of functional groups of the curing agent/the number of functional groups of the curable resin)."

[0180] (D') Curing Accelerator

The curable resin composition may include a curing accelerator. Details of the curing accelerator are the same as those of the curing accelerator according to the first embodiment.

Here, the term "epoxy resin composition" in the description of the curing accelerator according to the first embodiment is replaced with "curable resin composition."

In addition, the term "resin component" in the description of the amount of the curing accelerator according to the first embodiment refers to "both the curable resin and the curing agent."

[0181] (E') Inorganic Filler

The curable resin composition may include an inorganic filler. Details of the inorganic filler are the same as those of the inorganic filler according to the first embodiment.

Here, the term "epoxy resin composition" in the description of the inorganic filler according to the first embodiment is replaced with "curable resin composition."

[0182] [Various Additives]

The curable resin composition may include various additives such as a coupling agent, an ion exchanger, an adhesion promoter, a mold release agent, a flame retardant, a colorant, a thermoplastic resin, and a stress relaxation agent exemplified below, in addition to the above-described components. Details of the various additives are the same as those of the various additives according to the first embodiment.

Here, the term "epoxy resin composition" in the description of the various additives according to the first embodiment is replaced with "curable resin composition."

[0183] (Method of Preparing Curable Resin Composition)

A method of preparing the curable resin composition is not particularly limited. Details of the method of preparing the curable resin composition are the same as those of the method of preparing an epoxy resin composition according to the first embodiment. Here, the term "epoxy resin composition" in the description of the method of preparing an epoxy resin composition according to the first embodiment is replaced with "curable resin composition."

[0184] <Electronic Component Device according to the Second Embodiment>

An electronic component device according to the second embodiment is provided with an element sealed with the above-described curable resin composition. Details of the electronic component device are the same as those of the electronic component device according to the first embodiment. Here, the term "epoxy resin composition" in the description of the electronic component device according to the first embodiment is replaced with "curable resin composition."

Examples

[0185] The invention will be specifically described below in reference to the Examples.

However, the invention is not limited by these Examples. Unless otherwise specified, "part(s)" and "%" are based on mass.

[0186] [Examples according to the First Embodiment]

(Examples 1-1 to 1-8, Comparative Examples 1-1 to 1-5)

The following components were mixed in the amounts (unit: part by mass) shown in Tables 1 and 2 below, and roll-kneaded at a kneading temperature of 100°C for a kneading time of 10 minutes, thereby respectively preparing the epoxy resin compositions of Examples 1-1 to 1-8 and Comparative Examples 1-1 to 1-5. Note that blank columns in Tables 1 and 2 indicate that the components are not contained.

[0187] The following were used as monofunctional epoxy compounds in the Examples:

• Epoxy Compound 1: an epoxy compound having an epoxy equivalent weight of 196 g/eq represented by Formula (c) (CYCLOMER M100 manufactured by Daicel Corporation); and

• Epoxy Compound 2: 2-biphenylyl glycidyl ether having an epoxy equivalent weight of 226 g/eq.

[0188] The following were used as comparative epoxy compounds:

• Epoxy Compound 3: a bifunctional alicyclic epoxy compound having an epoxy equivalent weight of 126 g/eq (CELLOXIDE2021 manufactured by Daicel Corporation).

[0189] The following were used as polyfunctional epoxy compounds (polyfunctional epoxy compounds each having two or more epoxy groups in a molecule thereof):

• Polyfunctional Epoxy Compound 4: a methoxynaphthalene-type epoxy resin having an epoxy equivalent weight of 250 g/eq and a softening point of 58°C (HP-5000 manufactured by DIC CORPORATION);

• Polyfunctional Epoxy Compound 5: a biphenylene skeleton-containing phenol aralkyl-type epoxy resin having an epoxy equivalent weight of 241 g/eq and a softening point of 96°C (CER-3000L manufactured by Nippon Kayaku Co., Ltd.); and

• Polyfunctional Epoxy Compound 6: a biphenylene aralkyl-type epoxy resin having an epoxy equivalent weight of 282 g/eq and a softening point of 56°C (NC-3000 manufactured by Nippon Kayaku Co., Ltd.).

[0190] A xylylene-type phenol resin having a hydroxyl group equivalent weight of 175 g/eq and a softening point of 70°C (MEHC-7800SS manufactured by Meiwa Plastic Industries, Ltd.) was used as a curing agent.

A betaine-type adduct of triphenylphosphine and p-benzoquinone was used as a curing accelerator.

Spherical fused silica having an average particle diameter of 17.5 μm and a specific surface area of 3.8 m^2/g was used as an inorganic filler.

γ-Glycidoxypropyltrimethoxysilane was used as a silane coupling agent.

Carnauba wax and carbon black were used as other additive components.

[0191] [Evaluation of Epoxy Resin Composition]

Properties of the epoxy resin compositions prepared in Examples 1-1 to 1-8 and Comparative Examples 1-1 to 1-5 were evaluated by the following property tests (1) to (5). Results of the evaluation are shown in Tables 1 and 2 below. The epoxy resin compositions were molded using a transfer molding machine at a mold temperature of 175°C and a molding pressure of 6.9 MPa for a curing time of 120 seconds, unless otherwise specified. Post-curing was performed at 175°C for 5 hours where necessary.

[0192] (1) Spiral flow

The epoxy resin compositions were each molded under the above conditions using a spiral flow measurement mold according to EMMI-1-66, and the flow distance (cm) was determined.

[0193] (2) Heat hardness

The epoxy resin compositions were each molded into a disk having a diameter of 50 mm and a thickness of 3 mm under the above-described conditions. Heat hardness was

measured immediately after molding using a Shore D type hardness tester (HD-1120 (Type D) manufactured by Ueshima Seisakusho Co., Ltd.).

[0194] (3) Shear bond strength at 260°C

The epoxy resin compositions were each molded into a size of a bottom diameter of 4 mm, a top face diameter of 3 mm, and a height of 4 mm on a silver-plated copper plate under the above-described conditions, followed by post-curing. Thereafter, shear bond strength (MPa) was determined at a shear rate of 50 μm/s while maintaining the copper plate temperature at 260°C using Series 4000 manufactured by Nordson Advanced Technology K.K.

[0195] (4) Water absorption rate

The disks formed in (2) above were post-cured under the above-described conditions. Thereafter, the obtained disks were placed under the condition of 85°C, 60% RH for 168 hours, after which mass changes before and after placing the disks under the above condition was measured. Water absorption rate was calculated based on the measurement results by the following formula.

Water absorption rate (% by mass) = {(Mass of the disk after placing it in the above condition - Mass of the disk before placing it in the above condition) / Mass of the disk before placing it in the above condition} × 100

[0196] (5) Reflow resistance

80-pin flat packages (QFP), each having an external size of 20 mm × 14 mm × 2 mm mounted with a silicon chip having a size of 8 mm × 10 mm × 0.4 mm (lead frame material: copper alloy, a product with a die pad which is silver-plated at the top surface and a leads which are silver-plated at the tip thereof), were prepared by molding with the epoxy resin compositions under the above-described conditions, followed by post-curing under the above-described conditions. The obtained packages were humidified under the condition of 85°C, 85% RH for 168 hours. Thereafter, reflow processing was performed at 250°C, 260°C,

or 270°C for 10 seconds. Presence or absence of the cracks outside the packages was visually examined, and presence or absence of internal peelings was examined using an ultrasonic flaw detector (HYE-FOCUS manufactured by Hitachi Construction Machinery Co., Ltd.). Reflow resistance was evaluated by the total number of packages in which either cracking(s) or peeling(s) occurred, with respect to the number of test packages (10).

[0197]

[Table 1]

Item			Example							
			1-1	1-2	1-3	1-4	1-5	1-6	1-7	1-8
Formulation table	Epoxy compound	1	10	1	5	15	20	10	10	
		2								20
	Polyfunctional epoxy compound	4	100	100	100	100	100			100
		5						100		
		6							100	
	Curing agent		78.5	70.9	74.3	82.8	87.1	81.1	70.6	93.3
	Silane coupling agent		7.5	7.5	7.5	7.5	7.5	7.5	7.5	7.5
	Curing accelerator		3	3	3	3	3	3	3	3
	Carnauba wax		1	1	1	1	1	1	1	1
	Carbon black		3	3	3	3	3	3	3	3
	Fused silica		1562	1435	1492	1633	1705	1582	1502	1753
Evaluation items	Spiral flow (cm)		99	86	95	110	118	91	76	126
	Heat hardness		75	70	76	73	71	78	82	65
	Shear bond strength at 260°C		0.31	0.30	0.33	0.32	0.31	0.29	0.27	0.25
	Water absorption rate (%)		0.19	0.18	0.18	0.19	0.20	0.18	0.18	0.19
	Reflow resistance	250°C	0/10	0/10	0/10	0/10	0/10	0/10	0/10	0/10
		260°C	0/10	2/10	1/10	0/10	0/10	0/10	3/10	5/10
		270°C	0/10	7/10	4/10	0/10	1/10	3/10	6/10	8/10

[0198]

[Table 2]

Item			Comparative Example				
			1-1	1-2	1-3	1-4	1-5
Formulation table	Epoxy compound	3		10	20		
	Polyfunctional epoxy compound	4	100	100	100		
		5				100	
		6					100
	Curing agent		70.0	76.9	83.9	72.6	62.1
	Silane coupling agent		7.5	7.5	7.5	7.5	7.5
	Curing accelerator		3	3	3	3	3
	Carnauba wax		1	1	1	1	1
	Carbon black		3	3	3	3	3
	Fused silica		1420	1550	1680	1440	1359
Evaluation item	Spiral flow (cm)		85	96	105	81	70
	Heat hardness		82	78	77	82	85
	Shear bond strength at 260°C		0.30	0.27	0.28	0.29	0.27
	Water absorption rate (%)		0.18	0.18	0.18	0.17	0.17
	Reflow resistance	250°C	0/10	0/10	0/10	0/10	8/10
		260°C	10/10	9/10	7/10	10/10	10/10
		270°C	10/10	10/10	10/10	10/10	10/10

[0199] As shown in Tables 1 and 2, Examples 1-1 to 1-8, in which a monofunctional epoxy compound was contained, were superior in reflow resistance especially at 260°C or higher to Comparative Examples 1-1 to 1-5, in which no monofunctional epoxy compound was contained. In addition, sufficient curability was also maintained.

In addition, among Examples 1-1 to 1-5, in Examples 1-1, 1-3, 1-4, and 1-5, in which the amount of the monofunctional epoxy compound with respect to the polyfunctional epoxy compound was from 5 parts by mass to 20 parts by mass, there was a tendency that the spiral flow was excellent and favorable fluidity was observed.

[0200] [Examples according to the Second Embodiment]

(Examples 2-1 to 2-7, Comparative Examples 2-1 to 2-5)

The following components were mixed in the amounts (unit: part by mass) shown in

Tables 3 and 4 below, and roll-kneaded at a kneading temperature of 100°C for a kneading time of 10 minutes, thereby respectively preparing the curable resin compositions of Examples 2-1 to 2-7 and Comparative Examples 2-1 to 2-5. Note that the blank columns in Tables 1 and 2 indicate that the components are not contained.

[0201] The following was used as a specific acrylic compound in the Examples:

• Specific Acrylic Compound 1: an epoxy compound having an epoxy equivalent weight of 196 g/eq represented by Formula (b) (CYCLOMER M100 manufactured by Daicel Corporation).

[0202] The following was used as a comparative compound:

• Comparative Compound 1: a bifunctional alicyclic epoxy compound having an epoxy equivalent weight of 126 g/eq ((meth)acryloyl group-free CELLOXIDE2021 manufactured by Daicel Corporation).

[0203] The following were used as curable resins:

• Epoxy Resin 1: a methoxynaphthalene-type epoxy resin having an epoxy equivalent weight of 250 g/eq and a softening point of 58°C (HP-5000 manufactured by DIC CORPORATION);

• Epoxy Resin 2: a biphenylene skeleton-containing phenol aralkyl-type epoxy resin having an epoxy equivalent weight of 241 g/eq and a softening point of 96°C (CER-3000L manufactured by Nippon Kayaku Co., Ltd.); and

• Epoxy Resin 3: a biphenylene aralkyl-type epoxy resin having an epoxy equivalent weight of 282 g/eq and a softening point of 56°C (NC-3000 manufactured by Nippon Kayaku Co., Ltd.).

[0204] A xylylene-type phenol resin having a hydroxyl group equivalent weight of 175 g/eq and a softening point of 70°C (MEHC-7800SS manufactured by Meiwa Plastic Industries, Ltd.) was used as a curing agent.

A betaine-type adduct of triphenylphosphine and p-benzoquinone was used as a

curing accelerator.

Spherical fused silica having an average particle diameter of 17.5 μm and a specific surface area of 3.8 m^2/g was used as an inorganic filler.

γ-Glycidoxypropyltrimethoxysilane was used as a silane coupling agent.

Carnauba wax and carbon black were used as other additive components.

[0205] [Evaluation of Curable Resin Composition]

Properties of the curable resin compositions prepared in Examples 2-1 to 2-7 and Comparative Examples 2-1 to 2-5 were evaluated by the following property tests (1) to (5). Results of the evaluation are shown in Tables 3 and 4 below. The curable resin compositions were molded using a transfer molding machine at a mold temperature of 175°C and a molding pressure of 6.9 MPa for a curing time of 120 seconds, unless otherwise specified. Post-curing was performed at 175°C for 5 hours where necessary.

[0206] (1) Spiral flow

The curable resin compositions were each molded under the above conditions using a spiral flow measurement mold according to EMMI-1-66, and the flow distance (cm) was determined.

[0207] (2) Heat hardness

The curable resin compositions were each molded into a disk having a diameter of 50 mm and a thickness of 3 mm under the above-described conditions. Heat hardness was measured immediately after molding using a Shore D type hardness tester (HD-1120 (Type D) manufactured by Ueshima Seisakusho Co., Ltd.).

[0208] (3) Shear bond strength at 260°C

The curable resin compositions were each molded into a size of a bottom diameter of 4 mm, a top face diameter of 3 mm, and a height of 4 mm on a silver-plated copper plate under the above-described conditions, followed by post-curing. Thereafter, shear bond strength (MPa) was determined at a shear rate of 50 μm/s while maintaining the copper plate

temperature at 260°C using Series 4000 manufactured by Nordson Advanced Technology K.K.

[0209] (4) Water absorption rate

The disks formed in (2) above was post-cured under the above-described conditions. Thereafter, the obtained disks were placed under the condition of 85°C, 60% RH for 168 hours, after which mass changes before and after placing the disks under the above condition was measured. Water absorption rate was calculated based on the measurement results by the following formula.

Water absorption rate (% by mass) = {(Mass of the disk after placing it in the above condition - Mass of the disk before placing it in the above condition) / Mass of the disk before placing it in the above condition} × 100

[0210] (5) Reflow resistance

80-pin flat packages (QFP), each having an external size of 20 mm × 14 mm × 2 mm mounted with a silicon chip having a size of 8 mm × 10 mm × 0.4 mm (lead frame material: copper alloy, a product with a die pad which is silver-plated at the top surface and a leads which are silver-plated at the tip thereof), were prepared by molding with the curable resin compositions under the above-described conditions, followed by post-curing under the above-described conditions. The obtained packages were humidified under the condition of 85°C, 85% RH for 168 hours. Thereafter, reflow processing was performed at 250°C, 260°C, or 270°C for 10 seconds. Presence or absence of the cracks outside the packages was visually examined, and presence or absence of internal peelings was examined using an ultrasonic flaw detector (HYE-FOCUS manufactured by Hitachi Construction Machinery Co., Ltd.). Reflow resistance was evaluated by the total number of packages in which either cracking(s) or peeling(s) occurred, with respect to the number of test packages (10).

[0211]

[Table 3]

Item			Example						
			2-1	2-2	2-3	2-4	2-5	2-6	2-7
Formulation table	(A) Specific acrylic compound	1	10	1	5	15	20	10	10
	(B) Epoxy resin	1	100	100	100	100	100		
		2						100	
		3							100
	Curing agent		78.5	70.9	74.3	82.8	87.1	81.1	70.6
	Silane coupling agent		7.5	7.5	7.5	7.5	7.5	7.5	7.5
	Curing accelerator		3	3	3	3	3	3	3
	Carnauba wax		1	1	1	1	1	1	1
	Carbon black		3	3	3	3	3	3	3
	Fused silica		1562	1435	1492	1633	1705	1582	1502
Evaluation item	Spiral flow (cm)		99	86	95	110	118	91	76
	Heat hardness		75	70	76	73	71	78	82
	Shear bond strength at 260°C		0.31	0.30	0.33	0.32	0.31	0.29	0.27
	Water absorption rate (%)		0.19	0.18	0.18	0.19	0.20	0.18	0.18
	Reflow resistance	250°C	0/10	0/10	0/10	0/10	0/10	0/10	0/10
		260°C	0/10	2/10	1/10	0/10	0/10	0/10	3/10
		270°C	0/10	7/10	4/10	0/10	1/10	3/10	6/10

[0212]

[Table 4]

Item			Comparative Example				
			2-1	2-2	2-3	2-4	2-5
Formulation table	(A) Comparative compound	1		10	20		
	(B) Epoxy resin	1	100	100	100		
		2				100	
		3					100
	Curing agent		70.0	76.9	83.9	72.6	62.1
	Silane coupling agent		7.5	7.5	7.5	7.5	7.5
	Curing accelerator		3	3	3	3	3
	Carnauba wax		1	1	1	1	1
	Carbon black		3	3	3	3	3
	Fused silica		1420	1550	1680	1440	1359
Evaluation items	Spiral flow (cm)		85	96	105	81	70
	Heat hardness		82	78	77	82	85
	Shear bond strength at 260°C		0.30	0.27	0.28	0.29	0.27
	Water absorption rate (%)		0.18	0.18	0.18	0.17	0.17
	Reflow resistance	250°C	0/10	0/10	0/10	0/10	8/10
		260°C	10/10	9/10	7/10	10/10	10/10
		270°C	10/10	10/10	10/10	10/10	10/10

[0213] As shown in Tables 3 and 4, Examples 2-1 to 2-7, in which the specific acrylic compound was contained, were superior in reflow resistance especially at 260°C or higher to Comparative Examples 2-1 to 2-5, in which no specific acrylic compound was contained. In addition, sufficient heat hardness was also maintained.

Among Examples 2-1 to 2-5, in Examples 2-1, 2-3, 2-4, and 2-5, in which the amount of the specific acrylic compound with respect to the polyfunctional epoxy compound was from 5 parts by mass to 20 parts by mass, there was a tendency that the spiral flow was excellent and favorable fluidity was observed.

[0214] The disclosures of Japanese Patent Application No. 2017-072118, filed on March 31, 2017, and Japanese Patent Application No. 2017-072119, filed on March 31, 2017, are incorporated herein by reference in their entirety. All documents, patent applications, and

technical standards mentioned herein are incorporated herein by reference to the same extent as if individual document, patent application, and technical standard were each specifically and individually stated to be incorporated by reference.

[0215] The reference in this specification to any prior publication (or information derived from it), or to any matter which is known, is not, and should not be taken as an acknowledgment or admission or any form of suggestion that that prior publication (or information derived from it) or known matter forms part of the common general knowledge in the field of endeavour to which this specification relates.

CLAIMS

1. An epoxy resin composition comprising:

a monofunctional epoxy compound having one epoxy group in a molecule thereof, the monofunctional epoxy compound having one alicyclic epoxy group in the molecule thereof;

a polyfunctional epoxy compound having two or more epoxy groups in a molecule thereof; and

a curing agent.

2. The epoxy resin composition according to claim 1, wherein the monofunctional epoxy compound having one alicyclic epoxy group in a molecule thereof further has a (meth)acryloyl group.

3. The epoxy resin composition according to claim 1 or 2, wherein an amount of the monofunctional epoxy compound with respect to 100 parts by mass of the polyfunctional epoxy compound is from 1 part by mass to 30 parts by mass.

4. The epoxy resin composition according to any one of claims 1 to 3, wherein an amount of the monofunctional epoxy compound with respect to 100 parts by mass of the polyfunctional epoxy compound is from 10 parts by mass to 20 parts by mass.

5. The epoxy resin composition according to any one of claims 1 to 4, further comprising a curing accelerator.

6. The epoxy resin composition according to any one of claims 1 to 5, further

comprising an inorganic filler, wherein a content of the inorganic filler with respect to a total volume of the epoxy resin composition is from 70% by volume to 95% by volume.

7. The epoxy resin composition according to any one of claims 1 to 6, further comprising a silane coupling agent.

8. An electronic component device, which is provided with an element sealed with the epoxy resin composition according to any one of claims 1 to 7.