

DESCRIPTION

TITLE OF THE INVENTION: ADHESIVE COMPOSITION COMPRISING
AROMATIC POLYETHER DERIVATIVE

5

TECHNICAL FIELD

[0001] The present invention relates to an adhesive composition. More particularly, the present invention relates to an adhesive composition adhering between
10 layers of a laminate in a process of forming the laminate such as a semiconductor including an IC chip and an optical system product.

BACKGROUND ART

15 [0002] Recently, higher integration of a semiconductor device has been required in association with development of electronic devices such as mobile phones and IC cards having higher performance and smaller size. As a method for the higher integration, a semiconductor itself having a finer structure or a stacked structure made by stacking semiconductor elements in a longitudinal direction has been studied.

20 In production of the stacked structure, an adhesive is used for bonding between the semiconductor elements.

However, an acrylic resin, an epoxy resin, or a silicone resin publicly known as an adhesive only has a thermostability of about 250°C. Therefore, these adhesives cannot be used in processes requiring a high temperature of 250°C or more such as an electrode
25 bonding process of a metal bump and an ion diffusion process.

[0003] On the other hand, a substrate for thick film technique of forming an undercoating film containing 30% by volume to 45% by volume of polyetheretherketone resin powder and a thermosetting resin on an inorganic insulating substrate made by impregnating a silicone resin to a glass substrate is disclosed (Patent Document 1).

Related Art Documents

Patent Document

- [0004] Patent Document 1: Japanese Patent Application Publication No.
5 2009-070875 (JP 2009-070875 A)

SUMMARY OF THE INVENTION

Problem to be Solved by the Invention

- 10 [0005] The substrate on which the undercoating film is formed described above has dimensions that hardly change with moisture and heat, and thus, has excellent moisture resistance. However, polyetheretherketone contained in the undercoating film is not dissolved in a solvent or a resin solution. Therefore, polyetheretherketone cannot form a coating-type adhesive, and thus, adopted is a method for coating a substrate by a
15 screen printing method using polyetheretherketone in the form of paste-type ink. In this composition, screen printable ink cannot be formed when content of the polyetheretherketone is too high. As a result, the resin can be contained in a ratio of only 30% by volume to 45% by volume, and the composition has a problem of thermostability.

- 20 As described above, the acrylic resin, the epoxy resin, or the silicone resin publicly known as an adhesive only has a thermostability of about 250°C, and cannot be used under a high temperature of 250°C or more.

- On the other hand, an adhesive made by wholly aromatic polyether such as polyetheretherketone is exemplified in Patent Document 1. However,
25 polyetheretherketone has extremely low solvent solubility and is not soluble, so that the adhesive has poor solvent selectivity and is difficult to form a thick film. The adhesive also has a problem of an insufficient coating property.

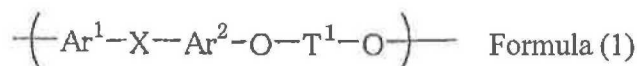
As a method for increasing the solvent solubility, introduction of a flexible structure represented by long-chain alkyl is suggested. However, this method generally causes

deterioration in thermostability.

[0006] The present invention was devised in view of the problems described above, and an object of the present invention is to provide a highly thermostable adhesive composition that dissolves easily into various organic solvents and that can form an adhesive layer with sufficient thickness and an excellent coating property, in which the adhesive layer has an extremely small thermogravimetry loss in thermal processes such as metal bump bonding, CVD, ion diffusion process, and the like and has excellent adhesion.

Means for Solving the Problem

10 [0007] The present invention relates to:
as a first aspect, an adhesive composition including a polymer that contains a unit structure of Formula (1):



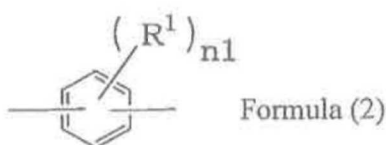
(where X is a sulfonyl group or a carbonyl group; Ar¹ and Ar² each are a C₆₋₃₀ arylene group; and T¹ is a fluoroalkylene group, a cyclic alkylene group, an arylene group having a substituent, or a combination of an arylene group optionally having a substituent and a fluoroalkylene group or a cyclic alkylene group);

as a second aspect, the adhesive composition according to the first aspect, in which the arylene group is a phenylene group, a naphthylene group, or an anthrylene group;

20 as a third aspect, the adhesive composition according to the first aspect or the second aspect, in which the polymer is a homopolymer containing one kind of unit structure;

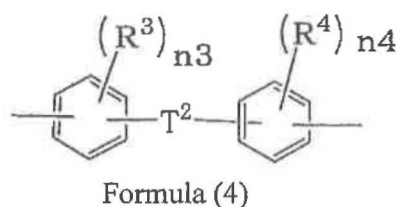
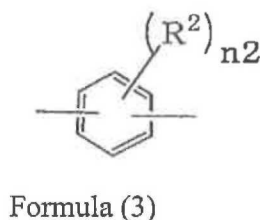
as a fourth aspect, the adhesive composition according to the first aspect or the second aspect, in which the polymer is a copolymer containing at least two kinds of unit structures;

25 as a fifth aspect, the adhesive composition according to any one of the first aspect to the fourth aspect including a polymer that contains the unit structure of Formula (1) with Ar¹ and Ar² each being a group of Formula (2):



(where R^1 is a C_{1-10} alkyl group, a C_{1-4} fluoroalkyl group, a hydroxy group, an allyl group, an allyloxy group, an amino group, a cyano group, a nitro group, an acyl group, an acyloxy group, a carboxy group, a group containing a tertiary-carbon structure, a cyclic alkyl group, or a combination of these, and n_1 is an integer of 0 to 4);

as a sixth aspect, the adhesive composition according to any one of the first aspect to the fifth aspect including a polymer that contains the unit structure of Formula (1) with T^1 being a group of Formula (3), the unit structure of Formula (1) with T^1 being a group of Formula (4), or a combination of the unit structures:



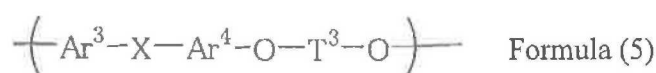
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(where R^2 , R^3 , and R^4 each are a C_{1-10} alkyl group, a C_{1-4} fluoroalkyl group, a hydroxy group, an allyl group, an allyloxy group, an amino group, a cyano group, a nitro group, an acyl group, an acyloxy group, a carboxy group, a group containing a tertiary-carbon structure, a cyclic alkyl group, or a combination of these; n_2 , n_3 , and n_4 each are an integer of 0 to 4; and T^2 is a fluoroalkylene group, a cyclic alkylene group, or a combination of these);

as a seventh aspect, the adhesive composition according to the sixth aspect, in which, in Formula (3), R^2 includes at least a group containing a tertiary-carbon structure, and n_2 is an integer of 1 to 4;

as an eighth aspect, the adhesive composition according to any one of the first aspect, the second aspect, and the fourth aspect to the seventh aspect further including a polymer that contains a unit structure of Formula (5):

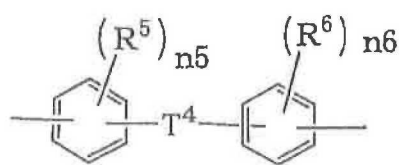
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(where X is a sulfonyl group or a carbonyl group; Ar³ and Ar⁴ each are a C₆₋₃₀ arylene group; and T³ is an alkylene group, a sulfonyl group, a carbonyl group, a C₆₋₃₀ arylene group, or a combination of these);

5 as a ninth aspect, the adhesive composition according to the eighth aspect, in which the arylene group is a phenylene group, a naphthylene group, or an anthrylene group;

as a tenth aspect, the adhesive composition according to the eighth aspect or the ninth aspect, in which T³ is a group of Formula (6):



Formula (6)

10 (where R⁵ and R⁶ each are a C₁₋₁₀ alkyl group, a C₁₋₄ fluoroalkyl group, a hydroxy group, an allyl group, an allyloxy group, an amino group, a cyano group, a nitro group, an acyl group, an acyloxy group, a carboxy group, a group containing a tertiary-carbon structure, a cyclic alkyl group, or a combination of these; n₅ and n₆ each are an integer of 0 to 4; T⁴ is an alkylene group, a sulfonyl group, a carbonyl group, a C₆₋₃₀ arylene group, or a combination of these);

15 as an eleventh aspect, the adhesive composition according to any one of the first aspect to the tenth aspect, in which the group containing a tertiary-carbon structure is a tertiary butyl group;

20 as a twelfth aspect, the adhesive composition according to any one of the first aspect to the eleventh aspect, in which the polymer has a weight average molecular weight of 500 to 5,000,000;

as a thirteenth aspect, the adhesive composition according to any one of the first aspect to the twelfth aspect further including a cross-linker;

as a fourteenth aspect, the adhesive composition according to any one of first aspect

to the thirteenth aspect further including a solvent, in which the adhesive composition has a viscosity of 0.001 to 5,000 Pa·s;

as a fifteenth aspect, a laminate including: at least two adhered articles; and an adhesive layer that is provided between the adhered articles, is formed from the adhesive composition according to any one of the first aspect to the fourteenth aspect, and has a thickness of 0.1 μm to 200 μm ; and

as a sixteenth aspect, the laminate according to the fifteenth aspect, in which the adhered articles each are selected from the group consisting of a silicon substrate, a glass substrate, a resin substrate, or a ceramic substrate.

10 Effects of the Invention

[0008] According to the present invention, the adhesive composition of the present invention includes a polymer that contains an aromatic polyether structure such as aromatic polyetheretherketone and aromatic polyetherethersulfone. Therefore, the adhesive composition can be easily dissolved into various organic solvents, and an excellent coating property can be obtained. Also, the adhesive composition of the present invention can form an adhesive layer with sufficient thickness for adhesion, and can form the adhesive layer, which has an extremely small thermogravimetry loss in thermal processes such as metal bump bonding, CVD, ion diffusion process, and the like, and that can retain high adhesion even after the thermal processes.

20 The adhesive composition of the present invention containing the polymer that contains the polyether structure is therefore effective for adhesion of a processed article (for example, a layered substance processed through heating treatment, pressure treatment, lithography, or the like) such as an electronic substrate used for a semiconductor device and the like that requires processing in the thermal processes.

25 The laminate of the present invention has an adhesive layer having high thermostability. Therefore, the adhered article can be processed in the thermal processes and the laminate can be used under high temperature conditions.

MODES FOR CARRYING OUT THE INVENTION

[0009] The present invention includes the adhesive composition including the polymer that contains the unit structure of Formula (1).

The adhesive composition includes the polymer that contains the unit structure of
5 Formula (1) and a solvent. Furthermore, as optional components, the adhesive composition may include an additional resin, a tackifier, a plasticizer, an adhesion improving agent, a stabilizer, a colorant, a surfactant, and the like for improving performance of the adhesive.

The adhesive composition of the present invention has a solid content of 0.1% by
10 mass to 80% by mass, and preferably 1% by mass to 60% by mass. The solid content is represented as a ratio of residual part formed by removing the solvent from the adhesive composition, to the adhesive composition. A ratio of the polymer that contains the unit structure of Formula (1) in the solid content can be set to 30% by mass to 100% by mass, and preferably 50% by mass to 100% by mass.

15 [0010] A preferable polymer used in the present invention is a thermoplastic resin.

A molecular weight of the polymer used in the present invention is, as weight average molecular weight, 500 to 5,000,000, preferably 1,000 to 1,000,000, and 1,000 to 100,000.

20 [0011] In Formula (1), X is a sulfonyl group or a carbonyl group; Ar¹ and Ar² each are a C₆₋₃₀ arylene group; and T¹ is a fluoroalkylene group, a cyclic alkylene group, an arylene group having a substituent or a combination of an arylene group optionally having a substituent and a fluoroalkylene group or a cyclic alkylene group.

The arylene group having a substituent is an arylene group having the substituents
25 described below.

The arylene group optionally having a substituent is a substituted or an unsubstituted arylene group, and is a combination of an arylene group and a fluoroalkylene group or a cyclic alkylene group.

The arylene group may be a phenylene group, a naphthyl group, an anthryl group,

or the like, and is preferably the phenylene group or the naphthyl group.

The fluoroalkylene group described above includes a C_{1-10} fluoroalkylene group. Specific examples thereof include a fluoromethylene group, a fluoroethylene group, a fluoro-n-propylene group, a fluoroisopropylene group, a fluoropropane-2,2-diyl group, a
 5 fluoro-n-butylene group, a fluoroisobutylene group, a fluoro-s-butylene group, and a fluoro-t-butylene group. Among them, the specific examples include C_{1-4} fluoroalkylene groups such as the perfluoropropane-2,2-diyl group. Also, a completely fluorinated (perfluorinated) alkylene group and a partially fluorinated (monofluorinated, difluorinated, and the like) alkylene group can be used as these fluoroalkylene groups.

10 The cyclic alkylene group may be a C_{3-30} cyclic alkylene group.

Examples of the cyclic alkylene group include a cyclopropylene group, a cyclobutylene group, a cyclohexylene group, a 1-methyl-cyclopentylene group, a 2-methyl-cyclopentylene group, a 3-methyl-cyclopentylene group, a 1-ethyl-cyclobutylene group, a 2-ethyl-cyclobutylene group, a 3-ethyl-cyclobutylene
 15 group, a 1,2-dimethyl-cyclobutylene group, a 1,3-dimethyl-cyclobutylene group, a 2,2-dimethyl-cyclobutylene group, a 2,3-dimethyl-cyclobutylene group, a 2,4-dimethyl-cyclobutylene group, a 3,3-dimethyl-cyclobutylene group, a 1-n-propyl-cyclopropylene group, a 2-n-propyl-cyclopropylene group, a 1-i-propyl-cyclopropylene group, a 2-i-propyl-cyclopropylene group, a
 20 1,2,2-trimethyl-cyclopropylene group, a 1,2,3-trimethyl-cyclopropylene group, a 2,2,3-trimethyl-cyclopropylene group, a 1-ethyl-2-methyl-cyclopropylene group, a 2-ethyl-1-methyl-cyclopropylene group, a 2-ethyl-2-methyl-cyclopropylene group, and a 2-ethyl-3-methyl-cyclopropylene group, and the like. Bivalent organic groups derived from adamantane and norbornene can also be used.

25 [0012] In the polymer that contains the unit structure of Formula (1) used in the present invention, these functional groups can be used, as T^1 , singly or in combination.

The polymer can be used as a homopolymer containing one type of repeating unit structure or a copolymer containing two or more types of repeating unit structures.

[0013] The present invention may include the polymer that contains the unit

structure of Formula (1) with Ar^1 and Ar^2 each being a group of Formula (2).

In Formula (2), R^1 is a C_{1-10} alkyl group, a C_{1-4} fluoroalkyl group, a hydroxy group, an allyl group, an allyloxy group, an amino group, a cyano group, a nitro group, an acyl group, an acyloxy group, a carboxy group, a group containing a tertiary-carbon structure, a cyclic alkyl group, or a combination of these, and n_1 is an integer of 0 to 4

In Formula (2), the alkyl group of R^1 is the C_{1-10} alkyl group. Examples of R^1 include a methyl group, an ethyl group, an n-propyl group, an i-propyl group, an n-butyl group, an i-butyl group, an s-butyl group, a t-butyl group, an n-pentyl group, a 1-methyl-n-butyl group, a 2-methyl-n-butyl group, a 3-methyl-n-butyl group, a 1,1-dimethyl-n-propyl group, a 1,2-dimethyl-n-propyl group, a 2,2-dimethyl-n-propyl group, a 1-ethyl-n-propyl group, an n-hexyl group, a 1-methyl-n-pentyl group, a 2-methyl-n-pentyl group, a 3-methyl-n-pentyl group, a 4-methyl-n-pentyl group, a 1,1-dimethyl-n-butyl group, a 1,2-dimethyl-n-butyl group, a 1,3-dimethyl-n-butyl group, a 2,2-dimethyl-n-butyl group, a 2,3-dimethyl-n-butyl group, a 3,3-dimethyl-n-butyl group, a 1-ethyl-n-butyl group, a 2-ethyl-n-butyl group, a 1,1,2-trimethyl-n-propyl group, a 1,2,2-trimethyl-n-propyl group, a 1-ethyl-1-methyl-n-propyl group, and a 1-ethyl-2-methyl-n-propyl group,

The fluoroalkyl group may be a C_{1-10} fluoroalkyl group, or may be a C_{1-4} fluoroalkyl group. Further, examples of the fluoroalkyl group include a fluoromethyl group, a fluoroethyl group, a fluoro-n-propyl group, a fluoroisopropyl group, a fluoro-n-butyl group, a fluoroisobutyl group, a fluoro-s-butyl group, and a fluoro-t-butyl group. A completely fluorinated (perfluorinated) alkyl group and a partially fluorinated alkyl group can be used as these fluoroalkyl groups.

The acyl group may be a C_{2-10} acyl group. Examples of the acyl group include a methylcarbonyl group, an ethylcarbonyl group, an n-propylcarbonyl group, an i-propylcarbonyl group, a cyclopropylcarbonyl group, an n-butylcarbonyl group, an i-butylcarbonyl group, an s-butylcarbonyl group, a t-butylcarbonyl group, a cyclobutylcarbonyl group, a 1-methyl-cyclopropylcarbonyl group, a 2-methylcyclopropylcarbonyl group, an n-pentylcarbonyl group, a

- 1-methyl-n-butylcarbonyl group, a 2-methyl-n-butylcarbonyl group, a
 3-methyl-n-butylcarbonyl group, a 1,1-dimethyl-n-propylcarbonyl group, a
 1,2-dimethyl-n-propylcarbonyl group, a 2,2-dimethyl-n-propylcarbonyl group, a
 1-ethyl-n-propylcarbonyl group, a cyclopentylcarbonyl group, a
 5 1-methyl-cyclobutylcarbonyl group, a 2-methyl-cyclobutylcarbonyl group, a
 3-methyl-cyclobutylcarbonyl group, a 1,2-dimethyl-cyclopropylcarbonyl group, a
 2,3-dimethyl-cyclopropylcarbonyl group, a 1-ethyl-cyclopropylcarbonyl group, a
 2-ethyl-cyclopropylcarbonyl group, an n-hexylcarbonyl group, a
 1-methyl-n-pentylcarbonyl group, a 2-methyl-n-pentylcarbonyl group, a
 10 3-methyl-n-pentylcarbonyl group, a 4-methyl-n-pentylcarbonyl group, a
 1,1-dimethyl-n-butylcarbonyl group, a 1,2-dimethyl-n-butylcarbonyl group, a
 1,3-dimethyl-n-butylcarbonyl group, a 2,2-dimethyl-n-butylcarbonyl group, a
 2,3-dimethyl-n-butylcarbonyl group, a 3,3-dimethyl-n-butylcarbonyl group, a
 1-ethyl-n-butylcarbonyl group, a 2-ethyl-n-butylcarbonyl group, and a
 15 1,1,2-trimethyl-n-propylcarbonyl group.

- The acyloxy group may be a C₂₋₁₀ acyloxy group. Examples of the acyloxy group
 include a methylcarbonyloxy group, an ethylcarbonyloxy group, an n-propylcarbonyloxy
 group, an i-propylcarbonyloxy group, a cyclopropylcarbonyloxy group, an
 n-butylcarbonyloxy group, an i-butylcarbonyloxy group, an s-butylcarbonyloxy group, a
 20 t-butylcarbonyloxy group, a cyclobutylcarbonyloxy group, a
 1-methyl-cyclopropylcarbonyloxy group, a 2-methyl-cyclopropylcarbonyloxy group, an
 n-pentylcarbonyloxy group, a 1-methyl-n-butylcarbonyloxy group, a
 2-methyl-n-butylcarbonyloxy group, a 3-methyl-n-butylcarbonyloxy group, a
 1,1-dimethyl-n-propylcarbonyloxy group, a 1,2-dimethyl-n-propylcarbonyloxy group, a
 25 2,2-dimethyl-n-propylcarbonyloxy group, a 1-ethyl-n-propylcarbonyloxy group, a
 cyclopentylcarbonyloxy group, a 1-methyl-cyclobutylcarbonyloxy group, a
 2-methyl-cyclobutylcarbonyloxy group, a 3-methyl-cyclobutylcarbonyloxy group, a
 1,2-dimethyl-cyclopropylcarbonyloxy group, a 2,3-dimethyl-cyclopropylcarbonyloxy
 group, a 1-ethyl-cyclopropylcarbonyloxy group, a 2-ethyl-cyclopropylcarbonyloxy group,

an n-hexylcarbonyloxy group, a 1-methyl-n-pentylcarbonyloxy group, a
 2-methyl-n-pentylcarbonyloxy group, a 3-methyl-n-pentylcarbonyloxy group, a
 4-methyl-n-pentylcarbonyloxy group, a 1,1-dimethyl-n-butylcarbonyloxy group, a
 1,2-dimethyl-n-butylcarbonyloxy group, a 1,3-dimethyl-n-butylcarbonyloxy group, a
 5 2,2-dimethyl-n-butylcarbonyloxy group, a 2,3-dimethyl-n-butylcarbonyloxy group, a
 3,3-dimethyl-n-butylcarbonyloxy group, a 1-ethyl-n-butylcarbonyloxy group, a
 2-ethyl-n-butylcarbonyloxy group, and a 1,1,2-trimethyl-n-propylcarbonyloxy group.

A tertiary-butyl group can be preferably used as the group containing a
 tertiary-carbon structure.

10 The cyclic alkyl group may be a C₃₋₃₀ cyclic alkyl group. Examples of the cyclic
 alkyl group include a cyclopropyl group, a cyclobutyl group, a cyclohexyl group, a
 1-methyl-cyclopentyl group, a 2-methyl-cyclopentyl group, a 3-methyl-cyclopentyl group,
 a 1-ethyl-cyclobutyl group, a 2-ethyl-cyclobutyl group, a 3-ethyl-cyclobutyl group, a
 1,2-dimethyl-cyclobutyl group, a 1,3-dimethyl-cyclobutyl group, a
 15 2,2-dimethyl-cyclobutyl group, a 2,3-dimethyl-cyclobutyl group, a
 2,4-dimethyl-cyclobutyl group, a 3,3-dimethyl-cyclobutyl group, a
 1-n-propyl-cyclopropyl group, a 2-n-propyl-cyclopropyl group, a 1-i-propyl-cyclopropyl
 group, a 2-i-propyl-cyclopropyl group, a 1,2,2-trimethyl-cyclopropyl group, a
 1,2,3-trimethyl-cyclopropyl group, a 2,2,3-trimethyl-cyclopropyl group, a
 20 1-ethyl-2-methyl-cyclopropyl group, a 2-ethyl-1-methyl-cyclopropyl group, a
 2-ethyl-2-methyl-cyclopropyl group, and a 2-ethyl-3-methyl-cyclopropyl group.

Monovalent organic groups derived from adamantane and norbornene can also be used.

[0014] The polymer used in the present invention may include a polymer that
 contains the unit structure of Formula (1) with T¹ being a group of Formula (3), the unit
 25 structure of Formula (1) with T¹ being a group of Formula (4), or a combination of these
 unit structures, other than the selection of Ar¹ and Ar² described above or with the
 selection of Ar¹ and Ar² described above.

In other words, the polymer can be used that contains the unit structure of Formula
 (1) with Ar¹ and Ar² being groups of Formula (2), the unit structure of Formula (1) with

T¹ being a group of Formula (3), the unit structure of Formula (1) with T¹ being a group of Formula (4), the unit structure of Formula (1) with Ar¹ and Ar² being groups of Formula (2) and T¹ being a group of Formula (3), the unit structure of Formula (1) with Ar¹ and Ar² being groups of Formula (2) and T¹ being a group of Formula (4), or the
 5 combination of these.

[0015] In Formula (3) and Formula (4), R², R³, and R⁴ are a C₁₋₁₀ alkyl group, a C₁₋₄ fluoroalkyl group, a hydroxy group, an allyl group, an allyloxy group, an amino group, a cyano group, a nitro group, an acyl group, an acyloxy group, a carboxy group, a group containing a tertiary-carbon structure, a cyclic alkyl group, or a combination of
 10 these; and n₂, n₃, and n₄ each are an integer of 0 to 4. T² is a fluoroalkylene group, a cyclic alkylene group, or a combination of these.

For the C₁₋₁₀ alkyl group, the C₁₋₄ fluoroalkyl group, the group containing a tertiary-carbon structure, the cyclic alkyl group, the acyl group, and the acyloxy group, the examples described above can be used.

15 For the fluoroalkylene group and the cyclic alkylene group, the examples described above can be used. These examples can be used singly or in combination.

In Formula (3), a unit structure in which R² includes at least a group containing a tertiary-carbon structure, and n₂ is an integer of 1 to 4 can be used.

[0016] In the present invention, in addition to the unit structure of Formula (1),
 20 a copolymer containing the unit structure of Formula (5) can be used. In Formula (5), X is a sulfonyl group or a carbonyl group; Ar³ and Ar⁴ each are a C₆₋₃₀ arylene group; and T³ is an alkylene group, sulfonyl group, a carbonyl group, a C₆₋₃₀ arylene group, or a combination of these. For the arylene group and the alkylene group, the examples described above can be used.

25 In Formula (5), a phenylene group, a naphthylene group, or an anthrylene group that are substituted or unsubstituted can be used.

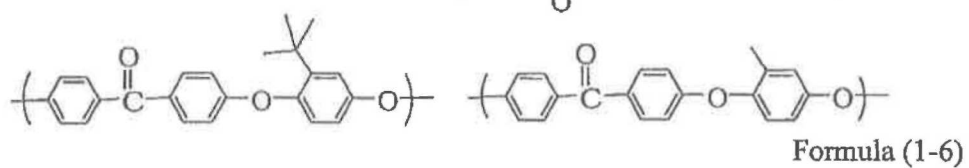
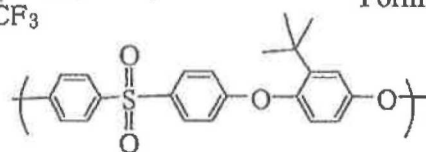
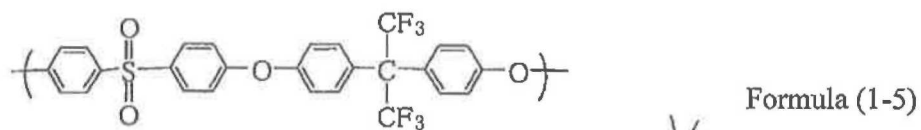
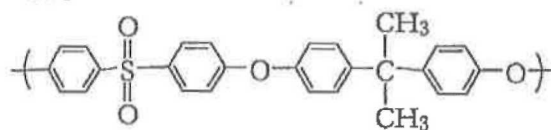
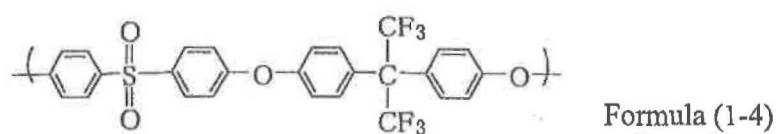
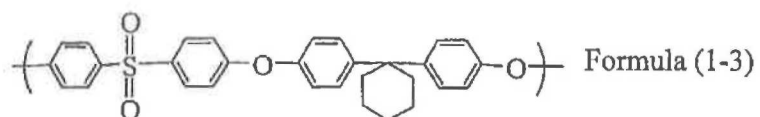
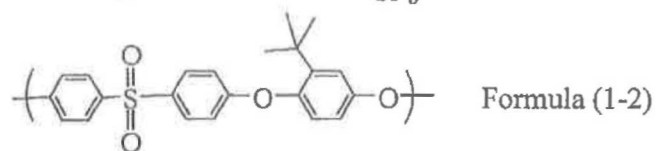
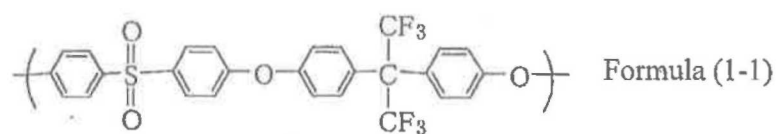
In Formula (5), the alkylene group may be a C₁₋₁₀ alkylene group. Examples of the alkylene group include a methylene group, an ethylene group, a propylene group, an isopropylene group, a propane-2,2-diyl group, an n-butylene group, an isobutylene group,

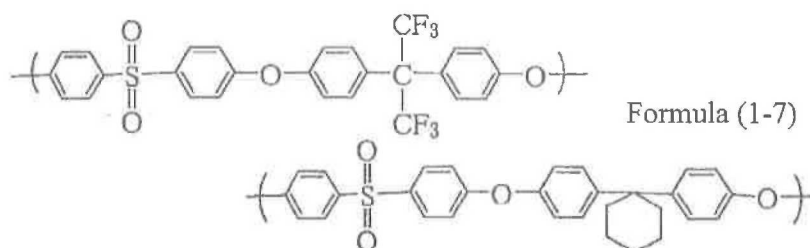
an s-butylene group, and a t-butylene group. Among them, the examples include C₁₋₄ alkylene groups such as the propane-2,2-diyl group.

In Formula (5), a group of Formula (6) can be used as T³. In Formula (6), R⁵ and R⁶ each are a C₁₋₁₀ alkyl group, a C₁₋₄ fluoroalkyl group, a hydroxy group, an allyl group, an allyloxy group, an amino group, a cyano group, a nitro group, an acyl group, an acyloxy group, a carboxy group, a group containing a tertiary-carbon structure, a cyclic alkyl group, or a combination of these; n₅ and n₆ each are an integer of 0 to 4. T⁴ is an alkylene group, a sulfonyl group, a carbonyl group, a C₆₋₃₀ arylene group, or a combination of these. For the alkyl group, the C₁₋₄ fluoroalkyl group, the acyl group, the acyloxy group, the group containing a tertiary-carbon structure, the cyclic alkyl group, the alkylene group, and the arylene group, the examples described above can be used.

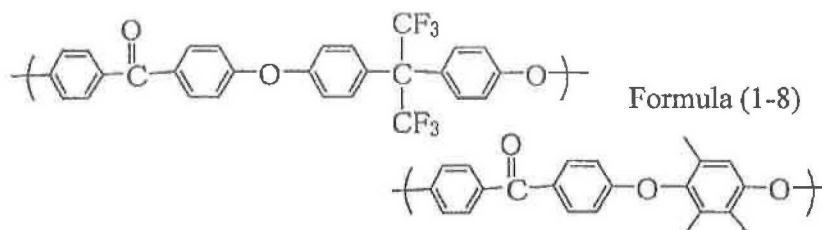
In the present invention, the group containing a tertiary-carbon structure is a functional group containing a tertiary-carbon structure. This functional group substitutes for a hydrogen atom on the carbon atom to form a quaternary carbon. Examples of an organic group bonding to the tertiary-carbon atom in the tertiary-carbon structure include alkyl groups such as a methyl group, an ethyl group, and a propyl group, and aryl groups such as a phenyl group. Among them, the methyl group is preferably used. A tertiarybutyl group having three methyl groups can be preferably used as a group containing a tertiary-carbon structure.

[0017] The unit structure of Formula (1) used in the present invention can be exemplified as follows:

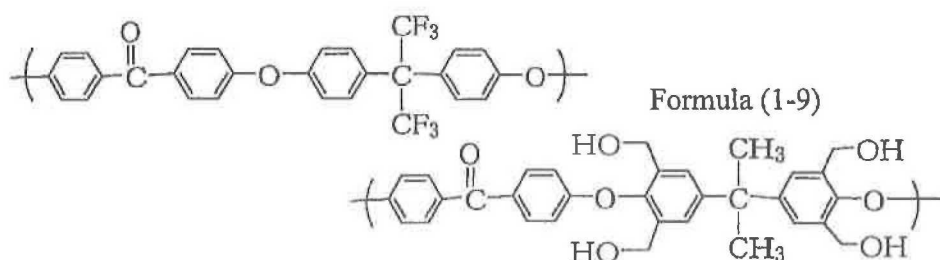




Formula (1-7)



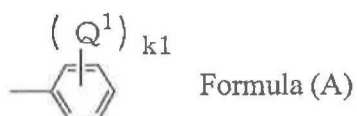
Formula (1-8)



Formula (1-9)

[0018] The polymer that contains the unit structure of Formula (1) may be a polymer that contains a hydroxy group or a halogen group as a terminal group, or a polymer that is capped by substituting the hydrogen atom of the hydroxy group or the halogen group with a phenyl group or the like. The capping group is a group of

5 Formula (A):



Formula (A)

where Q¹ is the alkyl group, or the alkyl group substituted with a halogen atom (fluorine, chlorine, bromine, and iodine), and k1 is an integer of 0 to 5.

10 [0019] In the present invention, a cross-linker may be used. The cross-linker has no limitation. However, a cross-linker having 2 or more, for example, 2 to 6, or 2 to 4 substituents that can react with the functional group in the polymer is preferable. A ratio of the cross-linker in the solid content can be 0% by mass to 50% by mass, and preferably 5% by mass to 40% by mass.

[0020] Examples of the cross-linker used in the present invention include a nitrogen-containing compound having a nitrogen atom substituted with a hydroxymethyl group or an alkoxymethyl group. Examples of the cross-linker include a nitrogen-containing compound having a nitrogen atom substituted by a hydroxymethyl group, a methoxymethyl group, an ethoxymethyl group, a butoxymethyl group, and a hexyloxymethyl group. An epoxy group-containing compound, an epoxy group-containing polymer, an allyl group-containing compound, an allyl group-containing polymer, an isocyanate group-containing compound, and an isocyanate group-containing polymer can be used as the cross-linker.

[0021] Specific examples of the nitrogen-containing compound include nitrogen-containing compounds such as hexamethoxymethylmelamine, tetramethoxymethylbenzoguanamine, 1,3,4,6-tetrakis(butoxymethyl)glycoluril, 1,3,4,6-tetrakis(hydroxymethyl)glycoluril, 1,3-bis(hydroxymethyl)urea, 1,1,3,3-tetrakis(butoxymethyl)urea, 1,1,3,3-tetrakis(methoxymethyl)urea, 1,3-bis(hydroxymethyl)-4,5-dihydroxy-2-imidazolinone, and 1,3-bis(methoxymethyl)-4,5-dimethoxy-2-imidazolinone.

Examples of the cross-linker also include commercially available compounds such as methoxymethyl-type melamine compounds (trade name: Cymel 300, Cymel 301, Cymel 303, and Cymel 350), butoxy methyl-type melamine compounds (trade name: Mycoat 506, and Mycoat 508), glycoluril compounds (trade name: Cymel 1170 and Powderlink 1174), a methylated urea resin (trade name: UFR 65), butylated urea resins (trade name: UFR 330, U-VAN 10S60, U-VAN 10R, and U-VAN 11HV), which are manufactured by Mitsui Cytec Ltd.; and urea/ formaldehyde-based resin (high condensation type, trade name: Beckamine J-300S, Beckamine P-955, and Beckamine N) manufactured by DIC Corporation, and the like.

The cross-linker may be a compound obtained by condensing the melamine compound, the urea compound, the glycoluril compound, and the benzoguanamine compound in which a hydrogen atom in an amino group is substituted with the hydroxymethyl group or the alkoxymethyl group as described above. For example, a

high molecular weight compound manufactured from the melamine compound (trade name: Cymel 303) and the benzoguanamine compound (trade name: Cymel 1123) described in U.S. Patent No. 6323310 can be used as a crosslinkable compound.

As the cross-linker, a polymer compound manufactured by using an acrylamide compound or a methacrylamide compound that are substituted by a hydroxymethyl group or an alkoxymethyl group such as N-hydroxymethylacrylamide, N-methoxymethylmethacrylamide, N-ethoxymethylacrylamide, and N-butoxymethylmethacrylamide can be used. Examples of such a polymer compound may include poly(N-butoxymethylacrylamide), a copolymer of N-butoxymethylacrylamide and styrene, a copolymer of N-hydroxymethylmethacrylamide and methyl methacrylate, a copolymer of N-ethoxymethylmethacrylamide and benzyl methacrylate, and a copolymer of N-butoxymethylacrylamide, benzyl methacrylate, and 2-hydroxypropyl methacrylate.

[0022] A compound having 1 to 6, or 2 to 4 epoxy ring(s) can be used as the cross-linker containing epoxy group(s). Examples of the compound having the epoxy ring(s) may include a compound having two or more hydroxy groups or carboxy groups such as a diol compound, a triol compound, a dicarboxylic acid compound, and a tricarboxylic acid, and a compound having two or more glycidyl ether structures or glycidyl ester structures manufactured from a glycidyl compound such as epichlorohydrin. Examples of the compound having the epoxy ring(s) include 1,4-butanediol diglycidyl ether, 1,2-epoxy-4-(epoxyethyl)cyclohexane, glycerol triglycidyl ether, diethylene glycol diglycidyl ether, 2,6-diglycidylphenyl glycidyl ether, 1,1,3-tris[p-(2,3-epoxypropoxy)phenyl]propane, 1,2-cyclohexanedicarboxylic acid diglycidyl ester, 4,4'-methylenebis(N,N-diglycidylaniline), 3,4-epoxycyclohexylmethyl-3,4-epoxycyclohexanecarboxylate, trimethylolethane triglycidyl ether, triglycidyl-p-aminophenol, tetraglycidyl meta-xylenediamine, tetraglycidyl diaminodiphenyl methane, tetraglycidyl-1,3-bisaminomethylcyclohexane, bisphenol-A-diglycidyl ether, bisphenol-S-diglycidyl ether, pentaerythritol tetraglycidyl ether, resorcinol diglycidyl ether, phthalic acid diglycidyl ester, neopentyl glycol

- diglycidyl ether, polypropylene glycol diglycidyl ether,
 tetrabromobisphenol-A-diglycidyl ether, bisphenolhexafluoroacetone diglycidyl ether,
 pentaerythritol diglycidyl ether, tris-(2,3-epoxypropyl)isocyanurate, monoallyl diglycidyl
 isocyanurate, diglycerol polydiglycidyl ether, pentaerythritol polyglycidyl ether,
 5 1,4-bis(2,3-epoxypropoxyperfluoroisopropyl)cyclohexane, sorbitol polyglycidyl ether,
 trimethylolpropane polyglycidyl ether, resorcin diglycidyl ether, 1,6-hexanediol
 diglycidyl ether, polyethylene glycol diglycidyl ether, phenyl glycidyl ether,
 p-tertiary-butylphenyl glycidyl ether, adipic acid diglycidyl ether, o-phthalic acid glycidyl
 ether, dibromophenyl glycidyl ether, 1,2,7,8-diepoxyoctane,
 10 1,6-dimethylolperfluorohexane diglycidyl ether,
 4,4'-bis(2,3-epoxypropoxyperfluoroisopropyl)diphenyl ether,
 2,2-bis(4-glycidyloxyphenyl)propane,
 3,4-epoxycyclohexylmethyl-3',4'-epoxycyclohexane carboxylate,
 3,4-epoxycyclohexyloxirane,
 15 2-(3,4-epoxycyclohexyl)-3',4'-epoxy-1,3-dioxane-5-spiro-cyclohexane,
 1,2-ethylenedioxy-bis(3,4-epoxycyclohexylmethane),
 4',5'-epoxy-2'-methylcyclohexylmethyl-4,5-epoxy-2-methylcyclohexane carboxylate,
 ethylene glycol-bis(3,4-epoxycyclohexane carboxylate),
 bis-(3,4-epoxycyclohexylmethyl) adipate, and bis(2,3-epoxycyclopentyl) ether.
 20 As the cross-linker, only one-type compound can be used, or a combination of two
 or more types of compounds can also be used.

[0023] The adhesive composition of the present invention may include a
 crosslinking catalyst. Reaction of a crosslinkable compound is accelerated by using the
 crosslinking catalyst.

- 25 An acid compound such as p-toluenesulfonic acid, trifluoromethanesulfonic acid,
 methanesulfonic acid, pyridinium-p-toluenesulfonic acid, salicylic acid, camphorsulfonic
 acid, sulfosalicylic acid, citric acid, benzoic acid, and hydroxybenzoic acid can be used as
 the crosslinking catalyst.

An aromatic sulfonic acid compound can be used as the crosslinking catalyst.

Specific examples of the aromatic sulfonic acid compound include p-toluenesulfonic acid, pyridinium-p-toluenesulfonic acid, sulfosalicylic acid, 4-chlorobenzenesulfonic acid, 4-hydroxybenzenesulfonic acid, benzenedisulfonic acid, 1-naphthalenesulfonic acid, and pyridinium-1-naphthalenesulfonic acid.

- 5 [0024] When the cross-linker having epoxy groups is used, a phenol resin, amines, a polyamide resin, imidazoles, polymercaptan, and an acid anhydride can be used as the crosslinking catalyst.

Examples of the phenol resin include a phenol-novolac resin and cresol-novolac resin.

- 10 Examples of the amines include piperidine, N,N-dimethylpiperadine, triethylenediamine, 2,4,6-tris(dimethylaminomethyl)phenol, benzyldimethylamine, 2-(dimethylaminomethyl)phenol, diethylenetriamine, triethylenetetramine, tetraethylenepentamine, diethylaminopropylamine, N-aminoethylpiperadine, di(1-methyl-2-aminocyclohexyl)methane, menthane diamine, isophoronediamine, 15 diaminodicyclohexylmethane, 1,3-diaminomethylcyclohexane, xylenediamine, meta-phenylenediamine, diaminodiphenylmethane, and diaminodiphenylsulfone. Among them, diethylenetriamine, triethylenetetramine, tetraethylenepentamine, diethylaminopropylamine, N-aminoethylpiperadine, di(1-methyl-2-aminocyclohexyl)methane, menthane diamine, isophoronediamine, 20 diaminodicyclohexylmethane, and the like can be preferably used.

Examples of the polyamide resin include a polyamidoamine having primary amines and secondary amines in its molecule, which is generated by condensing a dimer acid and a polyamine.

- 25 Examples of the imidazoles include 2-methylimidazole, 2-ethyl-4-methylimidazole, 1-cyanoethyl-2-undecylimidazolium trimellitate, and an epoxyimidazole adduct.

Examples of the polymercaptan include a polymercaptan having a mercaptan group at the terminal of a polypropylene glycol chain and a polymercaptan having a mercaptan group at the terminal of a polyethylene glycol chain.

As the acid anhydride, an anhydride of a compound having a plurality of carboxy

groups in its one molecule is preferable. Examples of the acid anhydride include phthalic anhydride, trimellitic anhydride, pyromellitic dianhydride, benzophenonetetracarboxylic dianhydride, ethylene glycol bistrimellitate, glycerol tristrimellitate, maleic anhydride, tetrahydrophthalic anhydride, methyltetrahydrophthalic anhydride, endo-methylenetetrahydrophthalic anhydride, methyl-endo-methylenetetrahydrophthalic anhydride, methylbutenyltetrahydrophthalic anhydride, dodecenylsuccinic anhydride, hexahydrophthalic anhydride, methylhexahydrophthalic anhydride, succinic anhydride, methylcyclohexenedicarboxylic anhydride, and chlorendic anhydride.

- 10 Examples of other crosslinking catalysts include an organic phosphorus compound such as triphenylphosphine and tributylphosphine; quaternary phosphonium salts such as ethyl triphenylphosphonium bromide and tetrabutylphosphonium diethyl dithiophosphate; and quaternary ammonium salts such as 1,8-diazabicyclo(5,4,0)undecane-7-ene, a salt of 1,8-diazabicyclo(5,4,0)undecane-7-ene and octylic acid, zinc octylate, and tetrabutylammonium bromide.

[0025] When a cross-linker having an allyl group is used, for example, a radical polymerization initiator and a cation polymerization initiator can be used as a curing catalyst.

- 20 Examples of the radical polymerization initiator include an imidazole compound, a diazo compound, a bisimidazole compound, an N-arylglycine compound, an organic azide compound, a titanocene compound, an alminate compound, an organic peroxide, an N-alkoxypyridinium chloride compound, and a thioxanthone compound.

- 25 Examples of the organic azide compound include p-azide benzaldehyde, p-azide acetophenone, p-azide benzoic acid, p-azidebenzalacetophenone, 4,4'-diazide chalcone, 4,4'-diazide diphenyl sulfide, and 2,6-bis(4'-azide benzal)-4-methylcyclohexanone.

Examples of the diazo compound include 1-diazo-2,5-diethoxy-4-p-tolylmercaptobenzene borofluoride, 1-diazo-4-N,N-dimethylaminobenzene chloride, and 1-diazo-4-N,N-diethylaminobenzene borofluoride.

Examples of the bisimidazole compound include

2,2'-bis(o-chlorophenyl)-4,5,4',5'-tetrakis(3,4,5-trimethoxyphenyl) 1,2'-bisimidazole and 2,2'-bis(o-chlorophenyl)-4,5,4',5'-tetraphenyl-1,2'-bisimidazole.

Examples of the titanocene compound include

- 5 dicyclopentadienyl-titanium-dichloride, dicyclopentadienyl-titanium-bisphenyl, dicyclopentadienyl-titanium-bis(2,3,4,5,6-pentafluorophenyl), dicyclopentadienyl-titanium-bis(2,3,5,6-tetrafluorophenyl), dicyclopentadienyl-titanium-bis(2,4,6-trifluorophenyl), dicyclopentadienyl-titanium-bis(2,6-difluorophenyl),
- 10 dicyclopentadienyl-titanium-bis(2,4-difluorophenyl), bis(methylcyclopentadienyl)-titanium-bis(2,3,4,5,6-pentafluorophenyl), bis(methylcyclopentadienyl)-titanium-bis(2,3,5,6-tetrafluorophenyl), bis(methylcyclopentadienyl)-titanium-bis(2,6-difluorophenyl) and dicyclopentadienyl-titanium-bis(2,6-difluoro-3-(1H-pyrrol-1-yl)-phenyl).

- 15 Examples of the radical polymerization initiator also include

1,3-di(tert-butyldioxy carbonyl)benzophenone, 3,3',4,4'-tetrakis(tert-butyldioxy carbonyl)benzophenone, 3-phenyl-5-isooxazolone, 2-mercaptobenzimidazole, 2,2-dimethoxy-1,2-diphenylethane-1-one, 1-hydroxy-cyclohexyl-phenyl-ketone, and

- 20 2-benzyl-2-dimethylamino-1-(4-morpholinophenyl)-butanone.

[0026] Examples of the cationic polymerization initiator include a sulfonic acid ester, a sulfonimide compound, a disulfonyl diazomethane compound, a dialkyl-4-hydroxysulfonium salt, arylsulfonic acid p-nitrobenzyl ester, a silanol-aluminum complex, and (η 6-benzene)(η 5-cyclopentadienyl) iron (II).

- 25 Examples of the sulfonimide compound include

N-(trifluoromethanesulfonyloxy)succinimide, N-(nonafluoro-normalbutanesulfonyloxy)succinimide, N-(camphorsulfonyloxy)succinimide, and N-(trifluoromethanesulfonyloxy)naphthalimide.

Examples of the disulfonyl diazomethane compound include
 bis(trifluoromethylsulfonyl)diazomethane, bis(cyclohexylsulfonyl)diazomethane,
 bis(phenylsulfonyl)diazomethane, bis(p-toluenesulfonyl)diazomethane,
 bis(2,4-dimethylbenzenesulfonyl)diazomethane, and
 5 methylsulfonyl-p-toluenesulfonyldiazomethane.

Examples of the cationic polymerization initiator also include
 2-methyl-1-(4-methylthiophenyl)-2-morpholinopropane-1-one.

An aromatic iodonium salt compound, an aromatic sulfonium salt compound, an
 aromatic diazonium salt compound, an aromatic phosphonium salt compound, a triazine
 10 compound, an iron-arene complex compound, and the like can be used as both of the
 radical polymerization initiator and the cationic polymerization initiator.

Examples of the aromatic iodonium salt compound include
 diphenyliodoniumhexafluorophosphate, diphenyliodoniumtrifluoromethanesulfonate,
 diphenyliodoniumnonafluoro-normalbutanesulfonate,
 15 diphenyliodoniumperfluoro-normaloctanesulfonate, diphenyliodoniumcamphorsulfonate,
 bis(4-tert-butylphenyl)iodoniumcamphorsulfonate, and bis(4-tert-butylphenyl)
 iodoniumtrifluoromethanesulfonate.

Examples of the aromatic sulfonium salt compound include
 triphenylsulfoniumhexafluoroantimonate,
 20 triphenylsulfoniumnonafluoro-normalbutanesulfonate,
 triphenylsulfoniumcamphorsulfonate, and triphenylsulfoniumtrifluoromethanesulfonate.

The crosslinking catalyst can be used singly or in combination of two or more of the
 crosslinking catalysts.

[0027] The crosslinking catalyst of 0 parts by mass to 10 parts by mass, or 0.01
 25 parts by mass to 10 parts by mass, or 0.05 parts by mass to 8 parts by mass, or 0.1 parts
 by mass to 5 parts by mass, or 0.3 parts by mass to 3 parts by mass, or 0.5 parts by mass
 to 1 part by mass to the polymer can be used.

[0028] A silicon substrate, a glass substrate, a resin substrate, and a ceramic
 substrate can be exemplified as an adhered article used in the present invention. These

substrates can be used as a supporting body of the laminate of the present invention.

[0029] Examples of the adhered article used in the present invention include inorganic materials such as silicon, silicon oxide, glass, and silicon nitride; metals such as aluminum and copper; and resin materials such as a polyamide resin, a polyimide resin,
5 an epoxy resin, a polyacrylic acid resin, a polymethacrylic acid resin, a polybenzoxazole resin, a polyhydroxystyrene resin, and a benzocyclobutene resin.

[0030] The adhesive composition can be dissolved using an organic solvent for spin coating. A polymer solution in which the polymer is dissolved in an organic solvent and its solution viscosity is in a range of 0.001 Pa·s to 5000 Pa·s can be prepared
10 as a coating solution having a spin coating property.

The organic solvent is not particularly limited as long as the solvent can be used in other semiconductor processes. However, ketones such as cyclohexanone, methyl isoamyl ketone, and 2-heptanone; polyvalent alcohols, and derivatives thereof, such as ethylene glycol, ethylene glycol monoacetate, diethylene glycol, diethylene glycol
15 monoacetate, propylene glycol, propylene glycol monoacetate, monomethyl ether, monoethyl ether, monopropyl ether, monobutyl ether, and monophenyl ether of dipropylene glycol or dipropylene glycol monoacetate; cyclic ethers such as dioxane, and esters such as methyl lactate, ethyl lactate, methyl acetate, ethyl acetate, butyl acetate, methyl pyruvate, ethyl pyruvate, methyl methoxypropionate, and ethyl ethoxypropionate
20 are preferably used. These solvents can be used singly or in combination of two or more solvents.

[0031] Commonly used additives such as an additional resin, a tackifier, a plasticizer, an adhesion improving agent, a stabilizer, a colorant, a surfactant, and the like for improving performance of a miscible additive such as an adhesive can be further
25 added to the adhesive composition according to this embodiment within a range that does not impair the substantial characteristics in the present invention.

Addition polymerization polymers and polycondensation polymers such as polyester, polystyrene, polyimide, an acrylic polymer, a methacrylic polymer, polyvinyl ether, phenol-novolac, naphthol-novolac, polyether, polyamide, and polycarbonate can be

used as the additional resin (polymer) for improving performance of the adhesive. A polymer that contains an aromatic ring structures such as a benzene ring, a naphthalene ring, an anthracene ring, a triazine ring, a quinoline ring, and a quinoxaline ring are preferably used.

- 5 Examples of such an additional resin (polymer) include addition polymerization polymers including addition-polymerizable monomers, such as benzyl acrylate, benzyl methacrylate, phenyl acrylate, naphthyl acrylate, anthryl methacrylate, anthryl methylmethacrylate, styrene, hydroxystyrene, benzyl vinyl ether, and N-phenylmaleimide, as their structural units; and polycondensation polymers such as phenol-novolac and
10 naphthol-novolac.

- A polymer that contains no aromatic ring can also be used as the additional resin (polymer). Examples of such a polymer include addition polymerization polymers including, as their structural units, only addition-polymerizable monomers having no aromatic rings, such as an alkyl acrylate, an alkyl methacrylate, a vinyl ether, an alkyl
15 vinyl ether, acrylonitrile, maleimide, an N-alkylmaleimide, and maleic anhydride.

- When the addition polymerization polymer is used as the additional resin (polymer), the polymer may be a homopolymer or a copolymer. The addition-polymerizable monomer is used for manufacturing the addition polymerization-type polymer. Examples of such an addition-polymerizable monomer include acrylic acid, methacrylic
20 acid, an acrylic acid ester compound, a methacrylic acid ester compound, an acrylamide compound, a methacrylamide compound, a vinyl compound, a styrene compound, a maleimide compound, maleic anhydride, and acrylonitrile.

- Examples of the acrylic acid ester compound include methyl acrylate, ethyl acrylate, normalhexyl acrylate, isopropyl acrylate, cyclohexyl acrylate, benzyl acrylate, phenyl
25 acrylate, anthrylmethyl acrylate, 2-hydroxyethyl acrylate, 3-chloro-2-hydroxypropyl acrylate, 2-hydroxypropyl acrylate, 2,2,2-trifluoroethyl acrylate, 2,2,2-trichloroethyl acrylate, 2-bromoethyl acrylate, 4-hydroxybutyl acrylate, 2-methoxyethyl acrylate, tetrahydrofurfuryl acrylate, 2-methyl-2-adamantyl acrylate, 5-acryloyloxy-6-hydroxynorbornene-2-carboxylic-6-lactone, 3-acryloxypropyl triethoxy

silane, and glycidyl acrylate.

Examples of the methacrylic acid ester compound include methyl methacrylate, ethyl methacrylate, normalhexyl methacrylate, isopropyl methacrylate, cyclohexyl methacrylate, benzyl methacrylate, phenyl methacrylate, anthrylmethyl methacrylate,
 5 2-hydroxyethyl methacrylate, 2-hydroxypropyl methacrylate, 2,2,2-trifluoroethyl methacrylate, 2,2,2-trichloroethyl methacrylate, 2-bromoethyl methacrylate, 4-hydroxybutyl methacrylate, 2-methoxyethyl methacrylate, tetrahydrofurfuryl methacrylate, 2-methyl-2-adamantyl methacrylate, 5-methacryloyloxy-6-hydroxynorbornene-2-carboxylic-6-lactone, 3-methacryloxypropyl
 10 triethoxy silane, glycidyl methacrylate, 2-phenylethyl methacrylate, hydroxyphenyl methacrylate, and bromophenyl methacrylate.

Examples of the acrylamide compound include acrylamide, N-methylacrylamide, N-ethylacrylamide, N-benzylacrylamide, N-phenylacrylamide, N,N-dimethylacrylamide, and N-anthrylacrylamide.

15 Examples of the methacrylamide compound include methacrylamide, N-methylmethacrylamide, N-ethylmethacrylamide, N-benzylmethacrylamide, N-phenylmethacrylamide, N,N-dimethylmethacrylamide, and N-anthrylacrylamide.

Examples of the vinyl compound include vinyl alcohol, 2-hydroxyethyl vinyl ether, methyl vinyl ether, ethyl vinyl ether, benzyl vinyl ether, vinylacetic acid, vinyl trimethoxy
 20 silane, 2-chloroethyl vinyl ether, 2-methoxyethyl vinyl ether, vinyl naphthalene, and vinyl anthracene.

Examples of the styrene compound include styrene, hydroxystyrene, chlorostyrene, bromostyrene, methoxystyrene, cyanostyrene, and acetylstyrene.

Examples of the maleimide compound include maleimide, N-methylmaleimide,
 25 N-phenylmaleimide, N-cyclohexylmaleimide, N-benzylmaleimide, and N-hydroxyethylmaleimide.

[0032] A molecular weight of the additional resin (polymer) used in the adhesive composition of the present invention is, as weight average molecular weight, 1,000 to 1,00,0000, or 3,000 to 300,000, or 5,000 to 200,000, or 10,000 to 100,000.

When the additional resin (polymer) is included in the adhesive composition of the present invention, the content of the additional resin (polymer) in the solid content is 0% by mass to 40% by mass, or 0% by mass to 20% by mass, or 1% by mass to 19% by mass.

5 [0033] The tackifier is added for controlling an elastic modulus, viscosity, and surface conditions of an adhesive layer. The type of such a tackifier is preferably determined in consideration of the viscosity of the adhesive layer. Specific examples of the tackifier include a single or a combination of an aliphatic petroleum resin, an aromatic petroleum resin, an aliphatic/aromatic copolymer-based petroleum resin, an
10 aliphatic hydrogenated petroleum resin, an alkylphenol resin, a xylene resin, a coumarone-indene resin, a terpene resin, a terpene-phenol resin, an aromatic modified terpene resin, a hydrogenated terpene resin, a rosin resin, a hydrogenated rosin resin, a disproportionated rosin resin, a dimerized rosin resin, and an esterified rosin resin. The tackifier can be included in a ratio of 0 parts by weight to 100 parts by weight per 100
15 parts by weight of the polyether that is the main component of the adhesive composition.

Therefore, an amount of the added tackifier is preferably a value within a range of 0 parts by weight to 100 parts by weight, and further preferably a value within a range of 0 parts by weight to 50 parts by weight.

 [0034] The present invention also relates to a laminate including at least two
20 adhered articles and an adhesive layer that is formed from the adhesive composition of the present invention and provided between the adhered articles.

In the present invention, the laminate of the adhered articles can be obtained by a method including applying the adhesive composition of the present invention on a first adhered article by spin coating, baking at a temperature from 50°C to 300°C to form an
25 adhesive layer, and adhering a second adhered article to the first adhered article through the formed adhesive layer. Processing the adhered article layers in the laminate can be further included.

A laminate made of a plurality of adhered articles and adhesive layers formed therebetween can also be formed by repeating methods including further forming an

adhesive layer on the second adhered article, adhering a third adhered article to the second adhered article through the formed adhesive layer, and optionally processing the adhered article layers obtained by adhesion.

In the present invention, a thickness of the adhesive applied to a semiconductor wafer by the spin coating can be set to 0.1 μm or more and 200 μm or less. When the thickness is thinner than 0.1 μm , void may be generated at the time of adhesion because the adhesive layer cannot follow the irregularities of the surface, while when the thickness is thicker than 200 μm , the adhesive layer may generate cracks. More preferably, the thickness of the adhesive layer is 1 μm to 50 μm .

Examples

[0035] Hereinafter, Examples in which characteristics of the adhesive composition according to the present invention are determined are described.

However, the present invention is not limited to the Examples.

GPC analysis of macromolecular compounds obtained in Synthesis Examples described below is performed using the following device and the following measurement conditions.

Device: Integrated high-speed GPC system HLC-8220GPC, manufactured by Tosoh Corporation

Column: KF-G, KF804L

Column temperature: 40°C

Solvent: THF

Flow rate: 1.0 mL/minute

Standard sample: Polystyrene

Detector: R1

[0036] Synthesis Example 1

To a flask equipped with a stirring device, a reflux condenser, a thermometer, and a dropping tank, 20.10 g of 4,4'-dichlorodiphenyl-sulfone, 24.77 g of 2,2-bis(4-hydroxyphenyl)hexafluoropropane, 29.02 g of potassium carbonate, and 369.50

g of N-methyl-2-pyrrolidinone were added. Thereafter, the air in the flask was replaced with nitrogen, and then the flask was heated to 160°C and the reaction was performed for 20 hours. The synthesized aromatic polyetherethersulfone was cooled to room temperature, and then filtrated to recover the filtrate. The filtrate was mixed with 30 ml of a mixed solution in which a volume ratio of N-methyl-2-pyrrolidinone and 1 mol/l hydrochloric acid was 90:10. Thereafter, the mixed solution was poured into methanol to perform reprecipitation purification.

Thereafter, the obtained precipitate was washed with methanol and water and dried under vacuum at 85 °C for 1 day. Thus, the aromatic polyetherethersulfone (corresponding to formula (1-1)) used in the present invention was obtained. When GPC analysis of the aromatic polyether was performed, the weight average molecular weight in terms of standard polystyrene was 18,600.

[0037] Synthesis Example 2

To a flask equipped with a stirring device, a reflux condenser, a thermometer, and a dropping tank, 68.20 g of 4,4'-dichlorodiphenyl-sulfone, 84.06 g of 2,2-bis(4-hydroxyphenyl)hexafluoropropane, 103.68 g of potassium carbonate, and 456.98 g of N-methyl-2-pyrrolidinone were added. Thereafter, the air in the flask was replaced with nitrogen, and then the flask was heated to 160 °C and the reaction was performed for 20 hours. Thereafter, a sample made by dissolving 32.83 g of 4-fluorobenzenetrifluoride in 160.48 g of N-methyl-2-pyrrolidinone was added dropwise as a capping agent, and the mixture was further stirred for 20 hours. The synthesized aromatic polyetherethersulfone was cooled to room temperature, and then filtrated to recover the filtrate. The filtrate was mixed with 100 ml of a mixed solution in which a volume ratio of N-methyl-2-pyrrolidinone and 2 mol/l hydrochloric acid was 90:10. Thereafter, the mixed solution was poured into methanol to perform reprecipitation purification.

Thereafter, the obtained precipitate was washed with methanol and water and dried under vacuum at 85 °C for 1 day. Thus, the aromatic polyether capped with the substituent, which was used in the present invention, was obtained. When GPC analysis

of the obtained aromatic polyetherethersulfone (corresponding to formula (1-1)) was performed, the weight average molecular weight in terms of standard polystyrene was 17,000.

[0038] Synthesis Example 3

5 To a flask equipped with a stirring device, a reflux condenser, a thermometer, and a dropping tank, 16.37 g of 4,4'-dichlorodiphenyl-sulfone, 9.97 g of tert-butylhydroquinone, 24.90 g of potassium carbonate, and 79.04 g of N-methyl-2-pyrrolidinone were added. Thereafter, the air in the flask was replaced with nitrogen, and then the flask was heated to 160°C and the reaction was performed for 20
10 hours. Thereafter, a sample made by dissolving 7.90 g of 4-fluorobenzenetrifluoride in 28.94 g of N-methyl-2-pyrrolidinone was added dropwise as a capping agent, and the mixture was further stirred for 20 hours. The synthesized aromatic polyetherethersulfone was cooled to room temperature, and then filtrated to recover the filtrate. The filtrate was mixed with 30 ml of a mixed solution in which a volume ratio
15 of N-methyl-2-pyrrolidinone and 2 mol/l hydrochloric acid was 90:10. Thereafter, the mixed solution was poured into methanol to perform reprecipitation purification.

Thereafter, the obtained precipitate was washed with methanol and water, and dried under vacuum at 85 °C for 1 day. Thus, the aromatic polyether capped with the substituent, which was used in the present invention, was obtained. When GPC analysis
20 of the obtained aromatic polyetherethersulfone (corresponding to formula (1-2)) was performed, the weight average molecular weight in terms of standard polystyrene was 14,800.

[0039] Synthesis Example 4

25 To a flask equipped with a stirring device, a reflux condenser, a thermometer, and a dropping tank, 13.64 g of 4,4'-dichlorodiphenyl-sulfone, 13.42 g of 1,1-bis(4-hydroxyphenyl)cyclohexane, 20.78 g of potassium carbonate, and 81.23 g of N-methyl-2-pyrrolidinone were added. Thereafter, the air in the flask was replaced with nitrogen, and then the flask was heated to 160°C and the reaction was performed for 20 hours. Thereafter, a sample made by dissolving 6.55 g of 4-fluorobenzenetrifluoride in

28.75 g of N-methyl-2-pyrrolidinone was added dropwise, and the mixture was further stirred for 20 hours. The synthesized aromatic polyetherethersulfone was cooled to room temperature, and then filtrated to recover the filtrate. The filtrate was mixed with 30 ml of a mixed solution in which a volume ratio of N-methyl-2-pyrrolidinone and 2 mol/l hydrochloric acid was 90:10. Thercafter, the mixed solution was poured into methanol to perform reprecipitation purification.

Thereafter, the obtained precipitate was washed with methanol and water, and dried under vacuum at 85°C for 1 day. Thus, the aromatic polyether capped with the substituent, which was used in the present invention, was obtained. When GPC analysis of the obtained aromatic polyetherethersulfone (corresponding to formula (1-3)) was performed, the weight average molecular weight in terms of standard polystyrene was 14,600.

[0040] Synthesis Example 5

To a flask equipped with a stirring device, a reflux condenser, a thermometer, and a dropping tank, 13.64 g of 4,4'-dichlorodiphenyl-sulfone, 8.41 g of 2,2-bis(4-hydroxyphenyl)hexafluoropropane, 5.71 g of 2,2-bis(4-hydroxyphenyl)propane, 34.56 g of potassium carbonate, and 83.44 g of N-methyl-2-pyrrolidinone were added. Thereafter, the air in the flask was replaced with nitrogen, and then the flask was heated to 160°C and the reaction was performed for 20 hours. Thereafter, a sample made by dissolving 6.56 g of 4-fluorobenzenetrifluoride in 29.56 g of N-methyl-2-pyrrolidinone was added dropwise, and the mixture was further stirred for 20 hours. The synthesized aromatic polyetherethersulfone was cooled to room temperature, and then filtrated to recover the filtrate. The filtrate was mixed with 30 ml of a mixed solution in which a volume ratio of N-methyl-2-pyrrolidinone and 2 mol/l hydrochloric acid was 90:10. Thereafter, the mixed solution was poured into methanol to perform reprecipitation purification.

Thereafter, the obtained precipitate was washed with methanol and water, and dried under vacuum at 85°C for 1 day. Thus, the aromatic polyether capped with the substituent, which was used in the present invention, was obtained. When GPC analysis

of the obtained aromatic polyetherethersulfone (corresponding to formula (1-4)) was performed, the weight average molecular weight in terms of standard polystyrene was 17,700.

[0041] Synthesis Example 6

5 To a flask equipped with a stirring device, a reflux condenser, a thermometer, and a dropping tank, 13.64 g of 4,4'-dichlorodiphenyl-sulfone, 8.41 g of 2,2-bis(4-hydroxyphenyl)hexafluoropropane, 4.16 g of tert-butylhydroquinone, 34.55 g of potassium carbonate, and 78.95 g of N-methyl-2-pyrrolidinone were added. Thereafter, the air in the flask was replaced with nitrogen, and then the flask was heated
10 to 160°C and the reaction was performed for 20 hours. Thereafter, a sample made by dissolving 6.56 g of 4-fluorobenzenetrifluoride in 28.06 g of N-methyl-2-pyrrolidinone was added dropwise, and the mixture was further stirred for 20 hours. The synthesized aromatic polyetherethersulfone was cooled to room temperature, and then filtrated to recover the filtrate. The filtrate was mixed with 30 ml of a mixed solution in which a
15 volume ratio of N-methyl-2-pyrrolidinone and 2 mol/l hydrochloric acid was 90:10. Thereafter, the mixed solution was poured into methanol to perform reprecipitation purification.

Thereafter, the obtained precipitate was washed with methanol and water, and dried under vacuum at 85°C for 1 day. Thus, the aromatic polyether capped with the
20 substituent, which was used in the present invention, was obtained. When GPC analysis of the obtained aromatic polyetherethersulfone (corresponding to formula (1-5)) was performed, the weight average molecular weight in terms of standard polystyrene was 17,300.

[0042] Synthesis Example 7

25 To a flask equipped with a stirring device, a reflux condenser, a thermometer, and a dropping tank, 94.26 g of 4,4'-difluorobenzophenone, 44.88 g of tert-butylhydroquinone, 33.52 g of methylhydroquinone, 82.10 g of potassium carbonate, and 764.27 g of N-methyl-2-pyrrolidinone were added. Thereafter, the air in the flask was replaced with nitrogen, and then the flask was heated to 140°C and the reaction was performed for 20

hours. The synthesized aromatic polyetheretherketone was cooled to room temperature, and then filtrated to recover the filtrate. The filtrate was mixed with 30 ml of a mixed solution in which a volume ratio of N-methyl-2-pyrrolidinone and 2 mol/l hydrochloric acid was 90:10. Thereafter, the mixed solution was poured into methanol to perform
5 reprecipitation purification.

Thereafter, the obtained precipitate was washed with methanol and water and dried under vacuum at 85°C for 1 day. Thus, the aromatic polyether capped with the substituent, which was used in the present invention, was obtained. When GPC analysis of the obtained aromatic polyetheretherketone (corresponding to formula (1-6)) was
10 performed, the weight average molecular weight in terms of standard polystyrene was 15,200.

[0043] Synthesis Example 8

To a flask equipped with a stirring device, a reflux condenser, a thermometer, and a dropping tank, 14.07 g of 4,4'-dichlorodiphenylsulfone, 8.40 g of
15 1,1-bis(4-hydroxyphenyl)hexafluoropropane, 6.71 g of 1,1-bis(4-hydroxyphenyl)cyclohexane, 20.75 g of potassium carbonate, and 87.68 g of N-methyl-2-pyrrolidinone were added. Thereafter, the air in the flask was replaced with nitrogen, and then the flask was heated to 160°C and the reaction was performed for 20 hours. The synthesized aromatic polyetherethersulfone was cooled to room temperature,
20 and then filtrated to recover the filtrate. The filtrate was mixed with 30 ml of a mixed solution in which a volume ratio of N-methyl-2-pyrrolidinone and 2 mol/l hydrochloric acid was 90:10. Thereafter, the mixed solution was poured into methanol to perform reprecipitation purification.

Thereafter, the obtained precipitate was washed with methanol and water and dried
25 under vacuum at 85°C for 1 day. Thus, the aromatic polyetherethersulfone (corresponding to formula (1-7)) used in the present invention was obtained. When GPC analysis of the aromatic polyether was performed, the weight average molecular weight in terms of standard polystyrene was 43,000.

[0044] Synthesis Example 9

To a flask equipped with a stirring device, a reflux condenser, a thermometer, and a dropping tank, 74.19 g of 4,4'-difluorobenzophenone, 94.14 g of 2,2-bis(4-hydroxyphenyl)hexafluoropropane, 18.26 g of trimethylhydroquinone, 60.81 g of potassium carbonate, and 742.22 g of N-methyl-2-pyrrolidinone were added.

- 5 Thereafter, the air in the flask was replaced with nitrogen, and then the flask was heated to 140°C and the reaction was performed for 20 hours. The synthesized aromatic polyetheretherketone was cooled to room temperature, and then filtrated to recover the filtrate. The filtrate was mixed with 30 ml of a mixed solution in which a volume ratio of N-methyl-2-pyrrolidinone and 2 mol/l hydrochloric acid was 90:10. Thereafter, the
10 mixed solution was poured into methanol to perform reprecipitation purification.

- Thereafter, the obtained precipitate was washed with methanol and water and dried under vacuum at 85° for 1 day. Thus, the aromatic polyetheretherketone (corresponding to formula (1-8)) used in the present invention was obtained. When GPC analysis of the aromatic polyether was performed, the weight average molecular weight in terms of
15 standard polystyrene was 24,700.

[0045] Synthesis Example 10

- To a flask equipped with a stirring device, a reflux condenser, a thermometer, and a dropping tank, 40.42 g of 4,4'-difluorobenzophenone, 73.97 g of 2,2-bis(4-hydroxyphenyl)hexafluoropropane, 4.03 g of
20 2,2-bis(4-hydroxy-3,5-dihydroxymethylphenyl)propane, 32.01 g of potassium carbonate, and 752.16 g of N-methyl-2-pyrrolidinone. Thereafter, the air in the flask was replaced with nitrogen, and then the flask was heated to 140°C and the reaction was performed for 20 hours. The synthesized aromatic polyetheretherketone was cooled to room temperature, and then filtrated to recover the filtrate. The filtrate was mixed with 30 ml
25 of a mixed solution in which a volume ratio of N-methyl-2-pyrrolidinone and 2 mol/l hydrochloric acid was 90:10. Thereafter, the mixed solution was poured into methanol to perform reprecipitation purification.

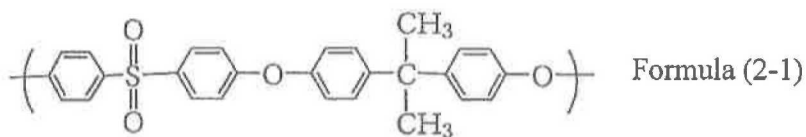
Thereafter, the obtained precipitate was washed with methanol and water and dried under vacuum at 85°C for 1 day. Thus, the aromatic polyetheretherketone

(corresponding to formula (1-9)) used in the present invention was obtained. When GPC analysis of the aromatic polyether was performed, the weight average molecular weight in terms of standard polystyrene was 19,000.

[0046] Comparative Synthesis Example 1

5 To a flask equipped with a stirring device, a reflux condenser, a thermometer, and a dropping tank, 15.00 g of 4,4'-dichlorodiphenyl-sulfone, 12.56 g of 2,2-bis(4-hydroxyphenyl)propane, 8.37 g of potassium carbonate, and 82.61 g of N-methyl-2-pyrrolidinone were added. Thereafter, the air in the flask was replaced with nitrogen, and then the flask was heated to 160° and the reaction was performed for 20
10 hours. The synthesized aromatic polyetherethersulfone was cooled to room temperature, and then filtrated to recover the filtrate. The filtrate was mixed with 30 ml of a mixed solution in which a volume ratio of N-methyl-2-pyrrolidinone and 2 mol/l hydrochloric acid was 90:10. Thereafter, the mixed solution was poured into methanol to perform reprecipitation purification.

15 Thereafter, the obtained precipitate was washed with methanol and water and dried under vacuum at 85° for 1 day. Thus, the aromatic polyetherethersulfone (corresponding to formula (2-1) used in Comparative Example 1 was obtained. When GPC analysis of the aromatic polyether was performed, the weight average molecular weight in terms of standard polystyrene was 16,700.



[0047] Comparative Synthesis Example 2

Polymethylmethacrylate that is a commercially available product and a synthesized acrylic resin (manufactured by Wako Pure Chemical Industries, Ltd.) was prepared.

[0048] (Evaluation of solvent solubility)

25 To 5 parts by mass of the aromatic polyethers obtained in Synthesis Examples 1 to 10 and Comparative Synthesis Example 1 and the acrylic resin in Comparative Synthesis Example 2, each of 95 parts by mass of (1) propylene glycol monomethyl ether (PGME),

- (2) propylene glycol monomethyl ether acetate (PGMEA), (3) ethyl lactate (EL), (4) cyclohexanone (Cy), (5) N-methyl-2-pyrrolidinone (NMP), (6) dimethylacetamide (DMAC), (7) 4-methyl-2-pentanone (MIBK), (8) 5-methyl-2-hexanone (MIAK), (9) ethyl acetoacetate (EAA), (10) gamma-butyrolactone (GBL), and (11) cyclopentanone (Cp) was added to evaluate the solubility.

The results are shown in Table 1. ○ indicates dissolved; △ indicates slightly dissolved; and × indicates hardly dissolved.

[0049] Table 1

	(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)	(9)	(10)	(11)
Synthesis Example 1	△	○	△	○	○	○	△	△	○	○	○
Synthesis Example 2	△	○	△	○	○	○	△	△	○	○	○
Synthesis Example 3	△	○	△	○	○	○	△	△	○	○	○
Synthesis Example 4	×	×	×	○	○	○	×	×	×	△	○
Synthesis Example 5	×	○	×	○	○	○	△	△	○	○	○
Synthesis Example 6	△	○	△	○	○	○	○	○	○	○	○
Synthesis Example 7	×	×	×	○	○	○	×	×	×	×	○
Synthesis Example 8	×	×	×	○	○	○	×	×	×	○	○
Synthesis Example 9	×	○	×	○	○	○	○	○	×	○	○
Synthesis Example 10	○	○	○	○	○	○	○	○	○	○	○
Comparative Synthesis Example 1	×	×	×	△	○	△	×	×	×	×	×
Comparative Synthesis Example 2	○	○	○	○	○	○	○	○	○	○	○

- [0050] The aromatic polyethers obtained in Synthesis Examples 1 to 10 had solubility similar to that of the acrylic resin used as Comparative Synthesis Example 2 and had solubility higher than that of the polysulfone obtained in Comparative Synthesis Example 1.

[0051] (Evaluation of thermostability)

- Thermostability of the aromatic polyethers obtained in Synthesis Examples 1 to 10 and Comparative Synthesis Example 1 and the polymethylmethacrylate used as

Comparative Synthesis Example 2 was evaluated based on a temperature generating 5% mass reduction determined by using TG-DTA (manufactured by Bruker AXS, TG/DTA2010SR) in a temperature rising of 10°C/minute. The results are shown in Table 2.

5 [0052] Table 2

	5% mass reduction temperature
Synthesis Example 1	500°C
Synthesis Example 2	512°C
Synthesis Example 3	459°C
Synthesis Example 4	470°C
Synthesis Example 5	486°C
Synthesis Example 6	487°C
Synthesis Example 7	477°C
Synthesis Example 8	491°C
Synthesis Example 9	493°C
Synthesis Example 10	500°C
Comparative Synthesis Example 1	500°C
Comparative Synthesis Example 2	323°C

[0053] Similar to the aromatic polyether in Comparative Synthesis Example 1, the aromatic polyethers obtained in Synthesis Examples 1 to 10 used in the present invention had 5% mass reduction temperature exceeding 450°C, and indicated higher thermostability compared to the acrylic resin in Comparative Synthesis Example 2.

10 [0054] (Preparation of adhesive composition)

Each adhesive composition was prepared in the following compositions using the aromatic polyethers obtained in Synthesis Examples 1 to 10 and Comparative Synthesis Example 1 and the acrylic resin in Comparative Synthesis Example 2.

[0055] Example 1

15 The aromatic polyether obtained in Synthesis Example 1 was dissolved into cyclohexanone to prepare an adhesive composition containing 20% by mass of solid content.

[0056] Example 2

20 The aromatic polyether obtained in Synthesis Example 2 was dissolved into cyclohexanone to prepare an adhesive composition containing 20% by mass of solid content.

[0057] Example 3

The aromatic polyether obtained in Synthesis Example 3 was dissolved into cyclohexanone to prepare an adhesive composition containing 20% by mass of solid content.

[0058] Example 4

5 The aromatic polyether obtained in Synthesis Example 4 was dissolved into cyclohexanone to prepare an adhesive composition containing 20% by mass of solid content.

[0059] Example 5

10 The aromatic polyether obtained in Synthesis Example 5 was dissolved into cyclohexanone to prepare an adhesive composition containing 20% by mass of solid content.

[0060] Example 6

15 The aromatic polyether obtained in Synthesis Example 6 was dissolved into cyclohexanone to prepare an adhesive composition containing 20% by mass of solid content.

[0061] Example 7

The aromatic polyether obtained in Synthesis Example 7 was dissolved into cyclohexanone to prepare an adhesive composition containing 20% by mass of solid content.

20 [0062] Example 8

The aromatic polyether obtained in Synthesis Example 8 was dissolved into cyclohexanone to prepare an adhesive composition containing 20% by mass of solid content.

[0063] Example 9

25 The aromatic polyether obtained in Synthesis Example 9 was dissolved into cyclohexanone to prepare an adhesive composition containing 20% by mass of solid content.

[0064] Example 10

The aromatic polyether obtained in Synthesis Example 10 was dissolved into

cyclohexanone to prepare an adhesive composition containing 20% by mass of solid content.

[0065] Example 11

To the aromatic polyether obtained in Synthesis Example 10, 5% by mass of Cymel 303 (a cross-linker) was dissolved in cyclohexanone to prepare an adhesive composition containing 20% by mass of solid content.

[0066] Example 12

To the aromatic polyether obtained in Synthesis Example 7, 5% by mass of Pinecrystal KE100 (a plasticizer, manufactured by ARAKAWA CHEMICAL INDUSTRIES, LTD., trade name, its component is an esterified rosin resin) was dissolved in cyclohexanone to prepare an adhesive composition containing 20% by mass of solid content.

[0067] Comparative Example 1

The aromatic polyether obtained in Comparative Synthesis Example 1 was dissolved into N-methyl-2-pyrrolidinone to prepare an adhesive composition containing 10% by mass of solid content.

[0068] Comparative Example 2

The polymethylmethacrylate in Comparative Synthesis Example 2 was dissolved into cyclohexanone to prepare an adhesive composition containing 20% by mass of solid content.

[0069] (Evaluation of coating property)

Films were formed in such a manner that each of the adhesive compositions obtained in Examples 1 to 12 and Comparative Example 1 and Comparative Example 2 was coated by spin coating on a silicon wafer under coating conditions of 1,000 rpm and 30 seconds, and then the coated wafer was baked at 100°C and 250°C each for 2 minutes. Then, the films were evaluated. The results are shown in Table 3.

[0070] Table 3

	Coating state	Film thickness
Example 1	Good	3.3 μm
Example 2	Good	3.3 μm

Example 3	Good	4.8 μm
Example 4	Good	5.4 μm
Example 5	Good	4.3 μm
Example 6	Good	4.1 μm
Example 7	Good	3.2 μm
Example 8	Good	4.7 μm
Example 9	Good	2.8 μm
Example 10	Good	2.0 μm
Example 11	Good	1.8 μm
Example 12	Good	2.9 μm
Comparative Example 1	Poor	-
Comparative Example 2	Good	6.5 μm

[0071] While the adhesive composition prepared in Comparative Example 1 was not able to form a uniform film because of generated coating failure, any of the adhesive compositions of the aromatic polyethers prepared in Examples 1 to 12 of the present invention and the acrylic resin used in Comparative Synthesis Example 2 were able to form uniform films having a thickness of 1 μm or more.

[0072] (Evaluation of adhesion property)

A coating film was formed on a silicon wafer in such a manner that each of the adhesive compositions obtained in Examples 1 to 12 and Comparative Example 1 and Comparative Example 2 was coated on the silicon wafer under coating conditions of 1,000 rpm and 30 seconds, and then the coated wafer was baked at each 100°C and 250°C each for 2 minutes. A 2-cm-square piece was cut out from the silicon wafer, and the cutout silicon wafer was placed on a glass wafer, which had been placed in advance on a hot plate of 270°C, such that the coated surface faced downward. The placed silicon wafer was pressurized from above to be bonded and an adhesion property of the bonded silicon wafer was evaluated. The results are shown in Table 4. The case that the wafer is adhered with no voids (holes) formed seen from the glass wafer surface is determined as good.

[0073] Table 4

	Adhesion state
Example 1	Good
Example 2	Good
Example 3	Good
Example 4	Good
Example 5	Good
Example 6	Good
Example 7	Good

Example 8	Good
Example 9	Good
Example 10	Good
Example 11	Good
Example 12	Good
Comparative Example 1	Poor
Comparative Example 2	Good

[0074] While the adhesive composition obtained in Comparative Example 1 generated adhesion failure and was not able to uniformly adhere, the adhesive compositions of the present invention in Examples 1 to 12 and the adhesive composition in Comparative Example 2 showed the excellent adhesion property.

5 [0075] (Preparation of sample for evaluating adhesion force)

Samples for evaluating adhesion force of the aromatic polyether prepared in Synthesis Example 7 and Synthesis Example 9 and the acrylic resin in Comparative Synthesis Example 2 were prepared by the following method.

[0076] Example 13

10 The aromatic polyether obtained in Synthesis Example 7 was dissolved into cyclohexanone to prepare an adhesive composition containing 27% by mass of solid content. A film having a thickness of 5.5 μm was formed in such a manner that the obtained adhesive composition was coated by spin coating on a silicon wafer of 4 inches under coating conditions of 1,500 rpm and 60 seconds, and then the coated wafer was
 15 baked at 100° C and 230°C each for 2 minutes. Thereafter, the silicon wafer was adhered to a glass wafer of 4 inches through the film under conditions of a degree of vacuum of 10 Pa or less, a temperature of 250°C, and a lamination pressure of 300 Kg using a lamination device (VJ-300, manufactured by Ayumi Industries Company Limited). The wafer was cut into 1-cm-square pieces to prepare samples using a dicing device
 20 (DAD 321, manufactured by DISCO Corporation).

[0077] Example 14

The aromatic polyether obtained in Synthesis Example 9 was dissolved into cyclohexanone to prepare an adhesive composition containing 27% by mass of solid content. A film having a thickness of 4.9 μm was formed in such a manner that the
 25 obtained adhesive composition was coated by spin coating on a silicon wafer of 4 inches

under coating conditions of 1,500 rpm and 60 seconds, and then the coated wafer was baked at 100°C and 230°C each for 2 minutes. Thereafter, the silicon wafer was adhered to a glass wafer of 4 inches through the film under conditions of a degree of vacuum of 10 Pa or less, a temperature of 270°C, and a lamination pressure of 300 Kg using a lamination device (VJ-300, manufactured by Ayumi Industries Company Limited). The wafer was cut into 1-cm-square pieces to prepare samples using a dicing device (DAD 321, manufactured by DISCO Corporation).

[0078] Comparative Example 3

The acrylic resin in Comparative Synthesis Example 2 was dissolved into cyclohexanone to prepare an adhesive composition containing 20% by mass of solid content. A film having a thickness of 5.0 μm was formed in such a manner that the obtained adhesive composition was coated by spin coating on a silicon wafer of 4 inches under coating conditions of 1,300 rpm and 60 seconds, and then the coated wafer was baked at 100°C and 200°C each for 2 minutes. Thereafter, the silicon wafer is adhered to a glass wafer of 4 inches through the film under conditions of a degree of vacuum of 10 Pa or less, a temperature of 270°C, and a lamination pressure of 400 Kg using a lamination device (VJ-300, manufactured by Ayumi Industries Company Limited). The wafer was cut into 1-cm square pieces to prepare samples using a dicing saw (DAD 321, manufactured by DISCO Corporation).

[0079] (Evaluation of adhesion force 1)

Araldite 2014 (trade name, manufactured by Huntsman Advanced Materials) was applied to both surfaces of the samples for evaluating adhesion force, which were obtained in Examples 13 and 14 and Comparative Example 3. After both surfaces of each sample were adhered to a dedicated jig for measuring adhesion force (shear), the adhesion force (shear) was evaluated using Autograph (Autograph AGS-100NX, manufactured by Shimadzu Corporation). The adhesion force was measured at a tensile speed of 1 mm/minute. The results are shown in Table 5.

[0080] Table 5

	Adhesion force (shear) N/cm^2
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Example 13	1,000 N or more
Example 14	1,000 N or more
Comparative Example 3	708 N

[0081] In Table 5, a value of the adhesion force being 1,000 N or more means that the adhesion force is equal to or more than measurement limit of the adhesion force measurement device.

[0082] The samples obtained in Example 13 and Example 14 indicated adhesion
5 force higher than that of the sample obtained in Comparative Example 3.

[0083] (Evaluation of adhesion force 2)

The samples for evaluating adhesion force, which were obtained in Examples 13 and 14 and Comparative Example 3, were placed on a hot plate heated at 350°C for 1 hour, and then Araldite 2014 (trade name, manufactured by Huntsman Advanced
10 Materials) was applied to the samples. After both surfaces of each sample were adhered to a dedicated jig for measuring adhesion shear force, the adhesion force (shear) was evaluated using Autograph (Autograph AGS-100NX, manufactured by Shimadzu Corporation). The adhesion force was measured at a tensile speed of 1 mm/minute. The results are shown in Table 6.

15 [0084] Table 6

	Adhesion force (shear)
Example 13	1000 N or more
Example 14	1000 N or more
Comparative Example 3	409 N

[0085] In Table 6, a value of the adhesion force being 1,000 N or more means that the adhesion force is equal to or more than measurement limit of the adhesion force measurement device.

[0086] While the adhesion forces of the samples obtained in Example 13 and
20 Example 14 remained high, the adhesion force of the sample obtained in Comparative Example 3 decreased.

INDUSTRIAL APPLICABILITY

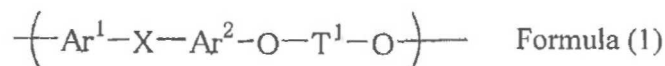
25 [0087] A highly thermostable adhesive composition can be provided that

dissolves easily into various organic solvents and that can form an adhesive layer with an excellent coating property and sufficient thickness, in which the adhesive layer has an extremely small thermogravimetry loss in thermal processes such as metal bump bonding, CVD, ion diffusion process, and the like and has excellent adhesion.

44 CLAIMS

1. An adhesive composition comprising:

a polymer that contains a unit structure of Formula (1):



(where X is a sulfonyl group or a carbonyl group; Ar¹ and Ar² each are a C₆₋₃₀ arylene group; and T¹ is a fluoroalkylene group, a cyclic alkylene group, an arylene group having a C₁₋₁₀ alkyl group, or a combination of an arylene group optionally having a substituent and a fluoroalkylene group or a cyclic alkylene group), and

a solvent, wherein

a solid content represented as a ratio of residual part formed by removing the solvent from the adhesive composition is 0.1% by mass to 80% by mass.

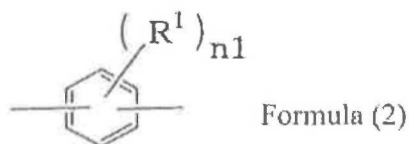
2. The adhesive composition according to claim 1, wherein the arylene group is a phenylene group, a naphthylene group, or an anthrylene group.

3. The adhesive composition according to claim 1 or 2, wherein the polymer is a homopolymer containing one unit structure.

4. The adhesive composition according to claim 1 or 2, wherein the polymer is a copolymer containing at least two unit structures.

5. The adhesive composition according to any one of claims 1 to 4, comprising:

a polymer that contains the unit structure of Formula (1) with Ar¹ and Ar² each being a group of Formula (2):

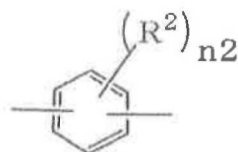


(where R¹ is a C₁₋₁₀ alkyl group, a C₁₋₄ fluoroalkyl group, a hydroxy group, an allyl group, an allyloxy group, an amino group, a cyano group, a nitro group, an acyl group, an acyloxy group, a carboxy group, a group containing a tertiary-carbon structure, a cyclic alkyl group, or a combination of these, and n₁ is an integer of 0 to 4).

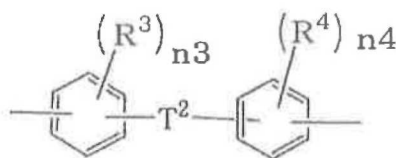
6. The adhesive composition according to any one of claims 1 to 5, comprising:

a polymer that contains the unit structure of Formula (1) with T¹ being a group of

Formula (3), the unit structure of Formula (1) with T^1 being a group of Formula (4), or a combination of the unit structure of Formula (1) with T^1 being a group of Formula (3), and the unit structure of Formula (1) with T^1 being a group of Formula (4):



Formula (3)

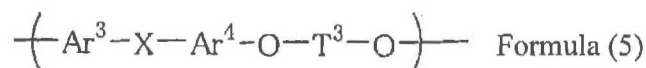


Formula (4)

(where R^2 , R^3 , and R^4 each are a C_{1-10} alkyl group, a C_{1-4} fluoroalkyl group, a hydroxy group, an allyl group, an allyloxy group, an amino group, a cyano group, a nitro group, an acyl group, an acyloxy group, a carboxy group, a group containing a tertiary-carbon structure, a cyclic alkyl group, or a combination of these; n_2 , n_3 , and n_4 each are an integer of 0 to 4; and T^2 is a fluoroalkylene group, a cyclic alkylene group, or a combination of these).

7. The adhesive composition according to claim 6, wherein, in Formula (3), R^2 is tertiary butyl, and n_2 is an integer of 1 to 4.

8. The adhesive composition according to any one of claims 1, 2, and 4 to 7, comprising: a copolymer that, in addition to a unit structure of Formula (1), further contains a unit structure of Formula (5):

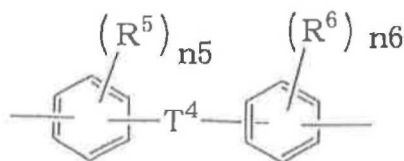


Formula (5)

(where X is a sulfonyl group or a carbonyl group; Ar^3 and Ar^4 each are C_{6-30} arylene group; and T^3 is an alkylene group, a sulfonyl group, a carbonyl group, a C_{6-30} arylene group, or a combination of these).

9. The adhesive composition according to claim 8, wherein the arylene group is a phenylene group, a naphthylene group, or an anthrylene group.

10. The adhesive composition according to claim 8 or 9, wherein T^3 is a group of Formula (6):



Formula (6)

(where R^5 and R^6 each are a C_{1-10} alkyl group, a C_{1-4} fluoroalkyl group, a hydroxy group,

an allyl group, an allyloxy group, an amino group, a cyano group, a nitro group, an acyl group, an acyloxy group, a carboxy group, a group containing a tertiary-carbon structure, a cyclic alkyl group, or a combination of these; n_5 and n_6 each are an integer of 0 to 4; T^4 is alkylene group, a sulfonyl group, a carbonyl group, a C_{6-30} arylene group, or a combination of these).

11. The adhesive composition according to any one of claims 1 to 10, wherein the group containing a tertiary-carbon structure is a tertiary butyl group.

12. The adhesive composition according to any one of claims 1 to 11, wherein the polymer has a weight average molecular weight of 500 to 5,000,000.

13. The adhesive composition according to any one of claims 1 to 12, further comprising a cross-linker.

14. The adhesive composition according to any one of claims 1 to 13, further comprising:

solvent, wherein

the adhesive composition has a viscosity of 0.001 to 5,000 Pa·s.

15. A laminate comprising:

at least two adhered articles, and

an adhesive layer that is provided between the adhered articles, is formed from the adhesive composition according to any one of claims 1 to 14, and has a thickness of 0.1 μm to 200 μm .

16. The laminate according to claim 15, wherein the adhered articles each are selected from the group consisting of a silicon substrate, a glass substrate, a resin substrate, or a ceramic substrate.