

DESCRIPTION

TITLE OF INVENTION: COMPOUND, METHOD FOR PRODUCING SAME, CURABLE RESIN COMPOSITION, AND CURED PRODUCT OF SAME

TECHNICAL FIELD

5 [0001] The present invention relates to a compound having a specific structure, a curable resin composition, and a cured product thereof, which are suitable for use in electrical and electronic components such as semiconductor encapsulants, printed wiring boards, and build-up laminates, lightweight, high-strength materials such as carbon fiber reinforced plastics and glass fiber reinforced plastics, and 3D printing applications.

10 BACKGROUND ART

[0002] In recent years, the required characteristics of laminates for mounting electrical and electronic components have become more widespread and sophisticated due to the expansion of their application fields. Conventional semiconductor chips were mainly mounted on metal lead frames, but semiconductor chips with high processing power, such as central processing
15 units (hereinafter referred to as CPUs), are increasingly being mounted on laminates made of polymer materials.

[0003] The fifth-generation communication system (5G), currently undergoing accelerated development, is expected to further increase capacity and speed. 5G will use increasingly higher frequencies. Reducing transmission loss is crucial to achieving high-speed
20 communication using high frequencies, requiring substrate materials with even lower dielectric properties. Transmission loss on printed circuit boards is due to conductor loss and dielectric loss. As described in Non-Patent Document 1, dielectric loss α_D is proportional to the square root of the dielectric constant ϵ_r and the dielectric loss tangent $\tan\delta$ of the dielectric. Therefore, improving the dielectric loss tangent $\tan\delta$, which contributes more than
25 the dielectric constant ϵ_r , is effective in reducing transmission loss. Low-loss dielectric materials include thermoplastic materials such as PTFE (polytetrafluoroethylene) and LCP (liquid

crystal polymer), but they have poor moldability compared to thermosetting resins. Given this, the development of thermosetting resins with excellent low dielectric properties is desired.

[0004] Against this background, polymeric materials with excellent low dielectric properties

5 have been investigated. For example, Patent Document 1 proposes a thermosetting resin composition containing an imide compound having a maleimide group and a phenol aralkyl resin having an aliphatic unsaturated bond. However, due to the presence of phenolic hydroxy groups that do not participate in the curing reaction, the electrical properties are not sufficient. Patent Document 2 also discloses an allyl ether-modified biphenyl aralkyl novolac resin to
10 which an allyl group is added along with the phenolic hydroxy group. However, it has been shown that the allyl ether-modified biphenyl aralkyl novolac resin undergoes Claisen rearrangement at 190°C. At 200°C, the molding temperature for typical substrates, phenolic hydroxy groups that do not participate in the curing reaction are generated, resulting in unsatisfactory electrical properties.

15 CITATION LIST

NON-PATENT LITERATURE

[0005]

NON-PATENT LITERATURE 1: "Consideration of signal loss factor in the high-speed Signal transmission on a printed Circuit board" 29th Spring Conference of the Japan Institute of
20 Electronics Packaging, Session ID: 16P1-17, 2015

PATENT LITERATURE

[0006]

PATENT LITERATURE 1: Japanese Patent Application Laid-Open No. H04-359911

PATENT LITERATURE 2: International Publication No. 2016/002704

25 SUMMARY OF INVENTION

TECHNICAL PROBLEM

[0007] The present invention has been made in view of the above circumstances, and an object of the present invention is to provide a compound having excellent low dielectric properties, a method for producing the same, a curable resin composition, and a cured product thereof.

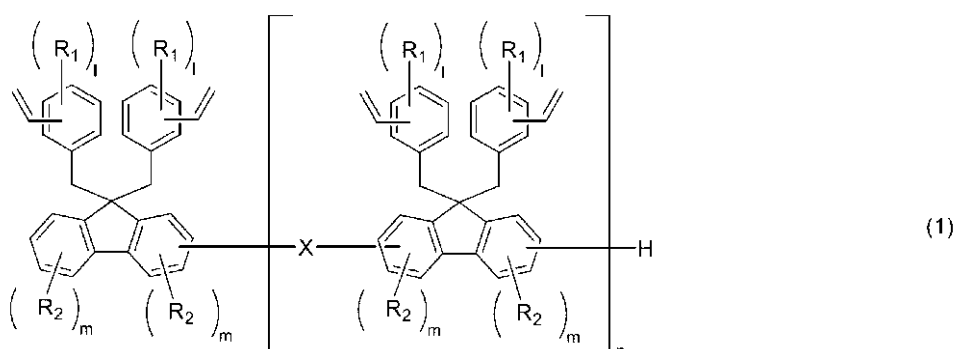
5 SOLUTION TO PROBLEM

[0008] That is, the present invention relates to the following [1] to [7]. In the present invention, "(numerical value 1) to (numerical value 2)" indicates that the upper and lower limits are included.

[1]

10 A compound represented by the following formula (1):

[0009] [Chemical Formula 1]



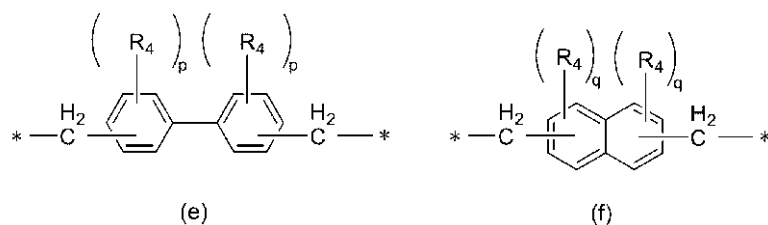
[0010] In the above formula (1), each of the multiple R_1 's independently represents a hydrogen atom or a hydrocarbon group having 1 to 5 carbon atoms. Each of the multiple R_2 's independently represents a hydrocarbon group represented by the following formula (a). X is a structure represented by any of the following formulas (c) to (f). Each of the multiple 1s independently represents an integer of 1 to 4. Each of the multiple ms independently represents an integer of 0 to 3, and the average value m_{ave} of m is $0 < m_{ave} \leq 3$. n is the number of repetitions, and the average value n_{ave} of n is $0.05 \leq n_{ave} \leq 20$.

20 [0011] [Chemical Formula 2]



(c)

(d)

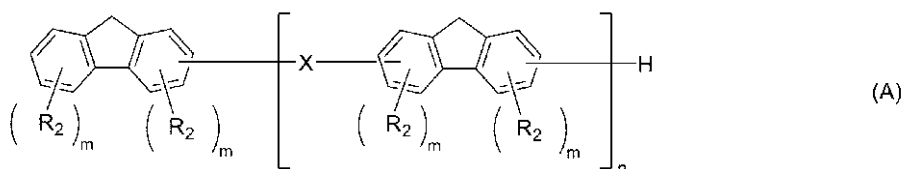


[0014] In the above formulas (c) to (f), * represents the bonding position to the fluorene structure in formula (1). Multiple R₄'s each independently represent a hydrogen atom or a hydrocarbon group having 1 to 5 carbon atoms. Multiple p's each independently represent an integer of 1 to 4, and multiple q's each independently represent an integer of 1 to 3.

A compound obtained by reacting a compound represented by the following formula

(A) with a compound represented by the following formula (B):

[0015] [Chemical Formula 4]



[0016] In the above formula (A), each of the multiple R₂'s independently represents a hydrocarbon group represented by the following formula (a). X has a structure represented by the following formulas (c) to (f). Each of the multiple m's independently represents an integer

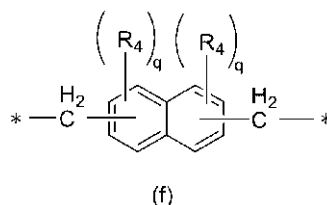
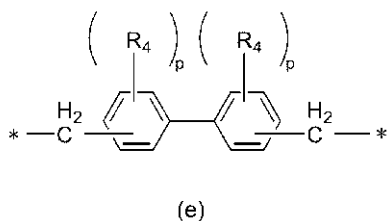
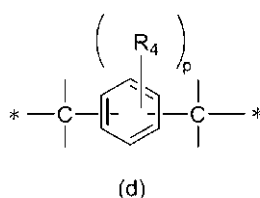
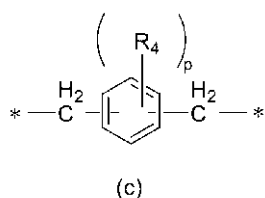
of 0 to 3, and the average value m_{ave} of m is $0 < m_{ave} \leq 3$. n is the number of repetitions, and the average value n_{ave} of n is $0.05 \leq n_{ave} \leq 20$.

[0017] [Chemical Formula 5]



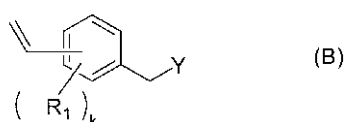
- 5 [0018] In the above formula (a), * represents the bonding position to the fluorene structure of formula (A). R_3 represents a hydrogen atom or a hydrocarbon group having 1 to 5 carbon atoms. k represents an integer of 1 to 5.

[0019] [Chemical Formula 6]



- 10 [0020] In the above formulas (c) to (f), * represents the bonding position to the fluorene structure in formula (A). Multiple R_4 's each independently represent a hydrogen atom or a hydrocarbon group having 1 to 5 carbon atoms. Multiple p 's each independently represent an integer of 1 to 4, and multiple q 's each independently represent an integer of 1 to 3.

[0021] [Chemical Formula 7]



15

- [0022] In the above formula (B), R_1 represents a hydrogen atom or a hydrocarbon group having 1 to 5 carbon atoms. k represents an integer of 1 to 4. Y represents a halogen atom.

[3]

A curable resin composition containing the compound described in the above item

[1] or [2].

[4]

5 A cured product obtained by curing the compound described in the above item [1] or [2].

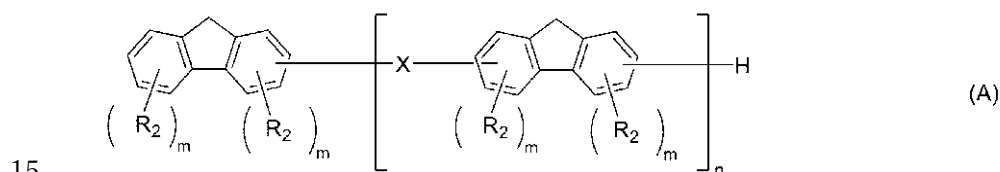
[5]

A cured product obtained by curing the curable resin composition described in the above item [3].

10 [6]

A method for producing a compound obtained by reacting a compound represented by the following formula (A) with a compound represented by the following formula (B) in an aprotic polar solvent in the presence of a basic catalyst.

[0023] [Chemical Formula 8]



[0024] In the above formula (A), each of the multiple R_2 's independently represents a hydrocarbon group represented by the following formula (a). X has a structure represented by the following formulas (c) to (f). Each of the multiple m 's independently represents an integer of 0 to 3, and the average value m_{ave} of m is $0 < m_{ave} \leq 3$. n is the number of repetitions, and the average value n_{ave} of n is $0.05 \leq n_{ave} \leq 20$.

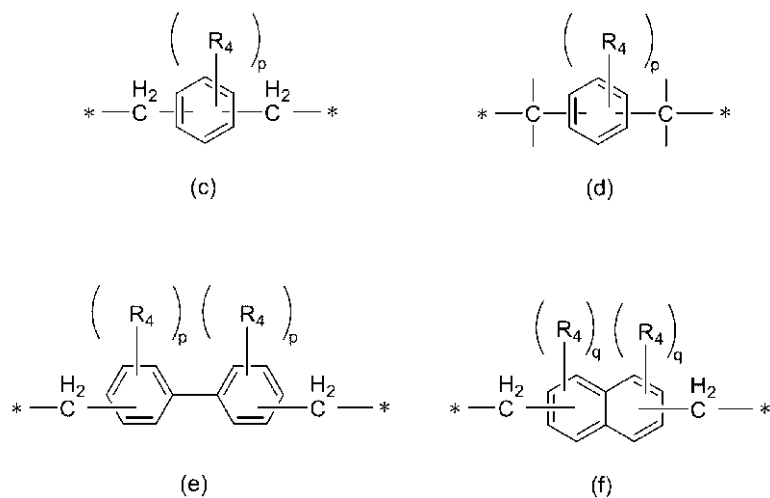
[0025] [Chemical Formula 9]



[0026] In the above formula (a), * represents the bonding position to the fluorene structure in formula (A). R_3 represents a hydrogen atom or a hydrocarbon group having 1 to 5 carbon

atoms. k represents an integer of 1 to 5.

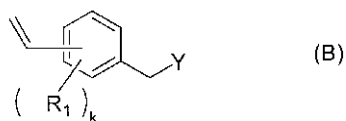
[0027] [Chemical Formula 10]



[0028] In the above formulas (c) to (f), * represents the bonding position to the fluorene

5 structure in formula (A). Multiple R₄'s each independently represent a hydrogen atom or a hydrocarbon group having 1 to 5 carbon atoms. Multiple p's each independently represent an integer of 1 to 4, and multiple q's each independently represent an integer of 1 to 3.

[0029] [Chemical Formula 11]

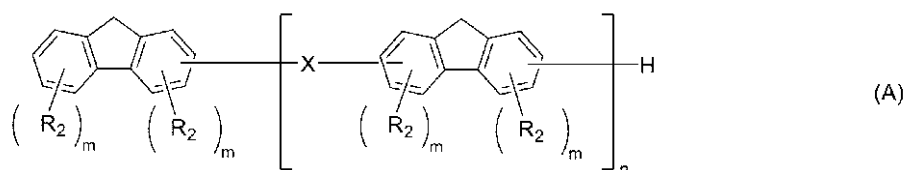


10 [0030] In the above formula (B), R₁ represents a hydrogen atom or a hydrocarbon group having 1 to 5 carbon atoms, k represents an integer of 1 to 4, and Y represents a halogen atom.

[7]

A compound represented by the following formula (A):

[0031] [Chemical Formula 12]



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In the above formula (A), multiple R₂'s each independently represent a hydrocarbon

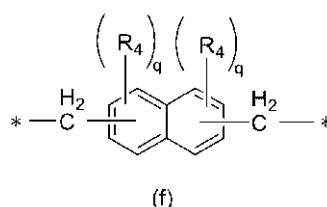
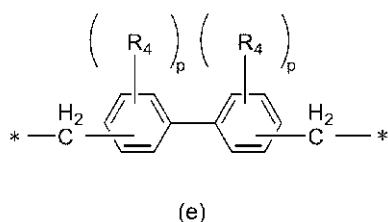
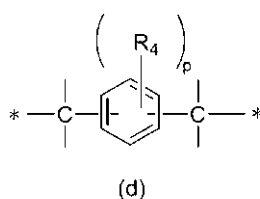
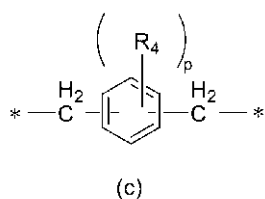
group represented by the following formula (a). X has a structure represented by the following formulas (c) to (f). Multiple m's each independently represent an integer of 0 to 3, and the average value m_{ave} of m is $0 < m_{ave} \leq 3$. n is the number of repetitions, and the average value n_{ave} of n is $0.05 \leq n_{ave} \leq 20$.

5 [0032] [Chemical Formula 13]



[0033] In the above formula (a), * represents the bonding position to the fluorene structure in formula (A). R_3 represents a hydrogen atom or a hydrocarbon group having 1 to 5 carbon atoms. k represents an integer of 1 to 5.

10 [0034] [Chemical Formula 14]



[0035] In the above formulas (c) to (f), * represents the bonding position to the fluorene structure in formula (A). Multiple R_4 's each independently represent a hydrogen atom or a hydrocarbon group having 1 to 5 carbon atoms. Multiple p's each independently represent an integer of 1 to 4, and multiple q's each independently represent an integer of 1 to 3.

ADVANTAGEOUS EFFECTS OF INVENTION

[0036] According to the present invention, it is possible to provide a compound and a curable resin composition having excellent low dielectric properties.

BRIEF DESCRIPTION OF DRAWINGS

[0037]

[Fig. 1] Fig. 1 shows an HP-LC chart of Example 1.

[Fig. 2] Fig. 2 shows a GPC chart of Example 1.

5 [Fig. 3] Fig. 3 shows a ^1H -NMR chart of Example 1.

[Fig. 4] Fig. 4 shows an HP-LC chart of Comparative Synthesis Example 1.

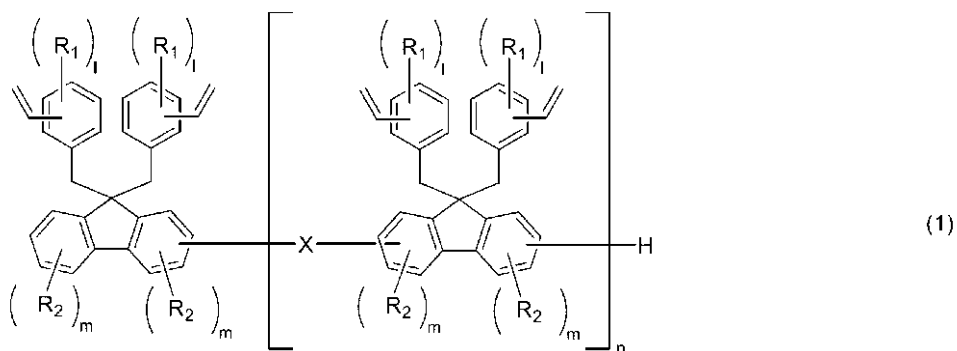
[Fig. 5] Fig. 5 shows a ^1H -NMR chart of Comparative Synthesis Example 1 .

DESCRIPTION OF EMBODIMENTS

[0038] Hereinafter, an embodiment of the present invention (hereinafter also referred to as
 10 "the present embodiment") will be described in further detail.

[0039] The compound of the present embodiment is represented by the following formula
 (1).

[0040] [Chemical Formula 15]



15 [0041] In the above formula (1), each of the multiple R_1 's independently represents a hydrogen atom or a hydrocarbon group having 1 to 5 carbon atoms, preferably a hydrogen atom or a hydrocarbon group having 1 to 3 carbon atoms, and more preferably a hydrogen atom. When the number of carbon atoms is 5 or less, the polymer is less likely to undergo molecular vibration when exposed to high frequency waves, resulting in excellent electrical
 20 properties. Furthermore, when the R_1 is a hydrogen atom, deterioration of the dielectric properties and water absorption properties associated with the generation of polar groups

resulting from the oxidation reaction of the alkyl group during a high-temperature storage test can be suppressed. Each of the multiple R_2 's independently represents a hydrocarbon group represented by the following formula (a). X has a structure represented by any of the following formulas (c) to (f). Each of the multiple l's independently represents an integer of 1 to 4, preferably 1. Each of the multiple m's independently represents an integer of 0 to 3. The average value m_{ave} of m is $0 < m_{ave} \leq 3$, and from the viewpoint of solvent solubility and heat resistance, it is more preferable that $0 < m_{ave} \leq 2.2$. The m_{ave} may be calculated from the charging ratio of the raw materials or from the results of NMR analysis. n is the number of repetitions, and the average value n_{ave} of n is $0.05 \leq n_{ave} \leq 20$. From the viewpoint of heat resistance and solvent solubility, the average value n_{ave} of n is preferably 0.05 to 15, more preferably 0.1 to 12.5, and even more preferably 0.1 to 10. The n_{ave} may be calculated from the charging ratio of the raw materials or from the results of NMR analysis. That is, the compound of the present embodiment is an aggregate of compounds in which n in formula (1) is an integer of 0 or more (compounds where $n=0$, compounds where $n=1$, compounds where $n=2$, compounds where $n=3$, ...). The molecular weight of the compound represented by formula (1) (the average molecular weight of the aggregate of compounds represented by formula (1)) is preferably 200 to 5,000, more preferably 300 to 3,000, and even more preferably 400 to 2,000, as determined by gel permeation chromatography (GPC) analysis using a differential refractive index detector. If the number average molecular weight is less than 200, there is a risk of reduced heat resistance due to residual raw materials, and tackiness may occur during B-staging. If the number average molecular weight exceeds 5,000, there is a risk of the increase in viscosity of the compound, which may impair circuit embedding properties or reduce solubility in solvents. The weight average molecular weight is preferably 200 to 10,000, more preferably 300 to 7,500, and even more preferably 400 to 5,000. If the weight average molecular weight is less than 200, there is a risk of reduced heat resistance due to residual raw materials, and tackiness may occur during B-staging. If the weight

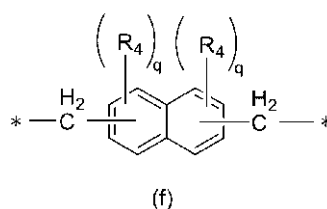
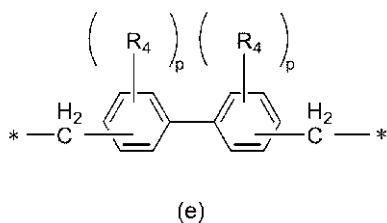
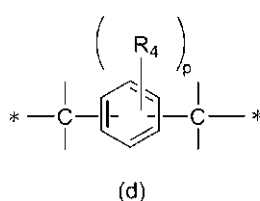
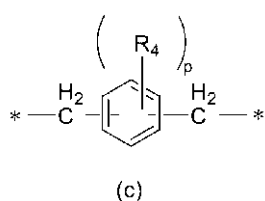
average molecular weight exceeds 10,000, the viscosity of the compound increases, which may impair the circuit embedding properties or reduce the solubility in solvents.

[0042] [Chemical Formula 16]



- 5 [0043] In the above formula (a), * represents the bonding position to the fluorene structure in formula (1). Multiple R₃'s each independently represent a hydrogen atom or a hydrocarbon group having 1 to 5 carbon atoms, preferably a hydrogen atom or a hydrocarbon group having 1 to 3 carbon atoms, and more preferably a hydrogen atom. When the number of carbon atoms is 5 or less, the molecular vibration is unlikely to occur when exposed to high
- 10 frequency waves, resulting in excellent electrical properties. Furthermore, when R₃ is a hydrogen atom, deterioration of the dielectric properties and water absorption properties associated with the generation of polar groups resulting from the oxidation reaction of the alkyl group during high-temperature storage tests can be suppressed. k represents an integer of 1 to 5, preferably 1.

15 [0044] [Chemical Formula 17]



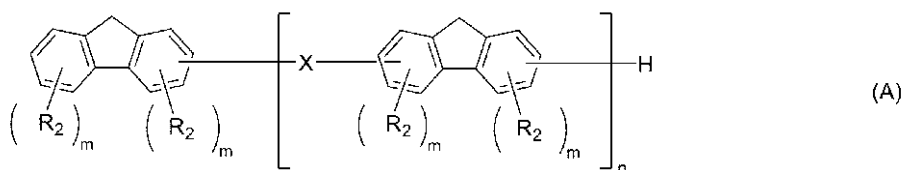
[0045] In the above formulas (c) to (f), * represents the bonding position to the fluorene structure in formula (1). From the perspective of exhibiting low water absorption and low

dielectric properties, any of the structures of formulas (c) to (e) is preferred, any of the structures of formulas (c) or (e) is more preferred, and the structure of formula (c) is particularly preferred. Multiple R₄'s each independently represent a hydrogen atom or a hydrocarbon group having 1 to 5 carbon atoms, preferably a hydrogen atom or a hydrocarbon group having 1 to 3 carbon atoms, and even more preferably a hydrogen atom. When the number of carbon atoms is 5 or less, molecular vibration is unlikely to occur when exposed to high frequency waves, resulting in excellent electrical properties. Furthermore, when the R₄ is a hydrogen atom, deterioration of dielectric properties and water absorption properties associated with the generation of polar groups resulting from the oxidation reaction of alkyl groups during high-temperature storage tests can be suppressed. Multiple p's each independently represent an integer of 1 to 4, preferably 1 to 3, more preferably 1 to 2, and most preferably 1. Multiple q's each independently represent an integer of 1 to 3, more preferably 1 to 2, and most preferably 1.

[0046] The method for producing the compound represented by formula (1) is not

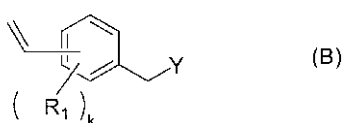
particularly limited, but the compound can be obtained by reacting a compound represented by the following formula (A) with a compound represented by the following formula (B).

[0047] [Chemical Formula 18]



[0048] In the above formula (A), the values and preferred ranges of R₂, X, m, and n are the same as those in the above formula (1).

[0049] [Chemical Formula 19]



[0050] In the above formula (B), the values and preferred ranges of R₁ and k are the same as

those in the above formula (1). Y represents a halogen atom, and from the viewpoints of reactivity and suppression of waste generation, it is preferably a bromine atom or a chlorine atom, and more preferably a chlorine atom.

[0051] When synthesizing the compound represented by formula (1), when the molar

5 number of the compound represented by formula (A) is α and the molar number of the compound represented by formula (B) is β , β/α is preferably 1.8 or more and 2.1 or less, more preferably 1.8 or more and 2.0 or less, and particularly preferably 1.8 or more and 1.95 or less. If β/α is less than 1.8, the compound represented by formula (A) remains unreacted, so that the toughness of the cured film may decrease and the dielectric properties may
10 deteriorate. This is because the unreacted compound represented by formula (A) does not have a structure that can be crosslinked, and the reaction of oxygen with the methylene structure at the 9-position of the compound represented by formula (A) produces a ketone, which increases polarity. If the β/α ratio is greater than 2.1, the halogen element in the compound represented by formula (B) that has not been completely removed by purification
15 may be eliminated during curing (e.g., at temperatures of 175°C or higher) or during high-temperature, high-humidity testing (e.g., at 85°C, 85% humidity, or 120°C, 100% humidity), potentially resulting in corrosion of the copper wiring. The amount of residual halogen contained in the reaction product containing the compound represented by formula (1) is preferably 1 to 10,000 ppm, more preferably 1 to 3,000 ppm, and even more preferably 1 to
20 2,000 ppm. Here, the "residual halogen" is derived from unreacted raw material compounds and impurities contained in the reaction product obtained by reacting the compound represented by formula (A) with the compound represented by formula (B) to obtain the compound represented by formula (1).

[0052] The reaction between the compound represented by formula (A) and the compound
25 represented by formula (B) will be described in detail. The compound represented by formula (1) can be obtained by reacting the compound represented by formula (A) with the compound

represented by formula (B) in an aprotic polar solvent in the presence of a basic catalyst.

Examples of aprotic polar solvents include dimethyl sulfone, dimethyl sulfoxide, dimethylformamide, dimethylacetamide, 1,3-dimethyl-2-imidazolidinone, and N-methylpyrrolidone, and two or more of these may be used in combination. Furthermore, if

5 necessary, a non-aqueous solvent may be used in combination. Examples of non-aqueous solvents include: aromatic solvents such as toluene and xylene; aliphatic solvents such as cyclohexane and n-hexane; ethers such as diethyl ether and diisopropyl ether; ester solvents such as ethyl acetate and butyl acetate; and ketone solvents such as methyl isobutyl ketone and cyclopentanone, but are not limited thereto, and two or more of these may be used in
10 combination. The basic catalyst is not particularly limited, and examples include basic catalysts such as sodium hydroxide, potassium hydroxide, and potassium carbonate. The order of adding the compound represented by formula (A), the compound represented by formula (B), and the basic catalyst can be changed as needed. However, a preferred method is to add the compound represented by formula (A), an aprotic polar solvent, and a basic catalyst,
15 sufficiently ionize the compound represented by formula (A), and then add the compound represented by formula (B). When the reaction is carried out without an aprotic polar solvent, the reaction rate significantly decreases. When an aprotic polar solvent is not used, the reaction is generally carried out using a phase transfer catalyst. In this case, the raw materials are dissolved in a non-aqueous solvent such as toluene, and the compound represented by
20 formula (A) and the compound represented by formula (B) are reacted in the presence of a basic catalyst such as aqueous sodium hydroxide solution and a phase transfer catalyst such as tetrabutylammonium bromide. In this case, it is difficult to completely remove the phase transfer catalyst such as tetrabutylammonium bromide, making it difficult to achieve low dielectric properties (low dielectric constant and low dielectric loss tangent). Furthermore, the
25 remaining phase transfer catalyst may cause problems such as ion migration when a substrate material using the compound of the present embodiment is subjected to a long-term wet heat

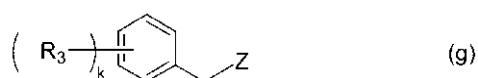
reliability test. The reaction temperature is preferably 0 to 120°C, more preferably 0 to 100°C, and even more preferably 0 to 80°C. If the temperature is higher than the upper limit, the compound of the present embodiment may undergo self-polymerization, resulting in gelation. If the temperature is lower than the lower limit, the reaction may not proceed sufficiently.

5 Neutralization with an optional acid compound may be performed as post-reaction treatment.

If necessary, an alcohol compound, water, or the like may be added to the reaction solution to recover the target product as crystals of the compound represented by formula (1). The resulting reaction solution or crystals may be redissolved in an optional solvent and subjected to an extraction step. For the extraction step, aromatic hydrocarbon solvents such as toluene and xylene may be used alone, or non-aromatic hydrocarbons such as cyclohexane may be used in combination. After extraction, the organic layer is washed with water until the wastewater becomes neutral, and the solvent is removed using an evaporator or the like to obtain the compound represented by formula (1).

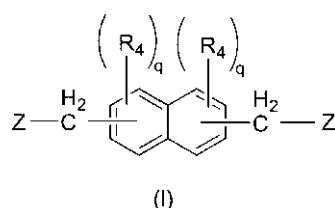
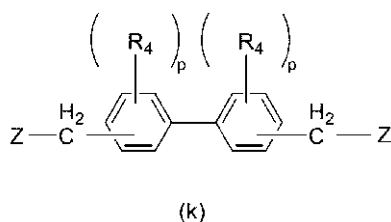
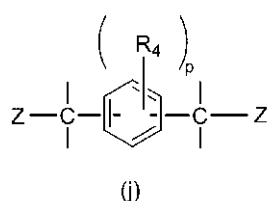
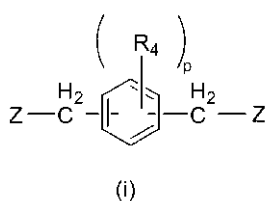
[0053] The compound represented by formula (A) is derived from fluorene and one or more of a compound represented by formula (g) below and compounds represented by formulas (i) to (l) below:

[0054] [Chemical Formula 20]



[0055] In the above formula (g), the values and preferred ranges of R₃ and k are the same as those in the above formula (a). Z represents a hydroxy group or a halogen atom, and from the viewpoints of reactivity and suppression of waste generation, Z is preferably a hydroxy group, a bromine atom, or a chlorine atom, and more preferably a hydroxy group or a chlorine atom.

[0056] [Chemical Formula 21]



[0057] In the above formulas (i) to (l), the values and preferred ranges of R_4 , p , and q are the same as those in the above formulas (c) to (f). Z represents a hydroxy group or a halogen atom, and from the viewpoint of reactivity and suppression of waste generation, Z is

5 preferably a hydroxy group, a bromine atom, or a chlorine atom, and more preferably a hydroxy group or a chlorine atom.

[0058] When reacting fluorene with a compound represented by formula (g) and a compound selected from one or more of the compounds represented by formulas (i) to (l), the molar ratio of the compound represented by formula (g) to 1 mol of fluorene is preferably

10 0.01 to 10 mol, more preferably 0.1 to 7.5 mol, even more preferably 0.2 to 5 mol, and most preferably 0.3 to 2.5 mol. If the molar ratio of the compound represented by formula (g) to be reacted with fluorene is less than 0.01 mol, there is a concern that the storage stability after dissolving in a solvent may be deteriorated, such as crystal precipitation when the compound represented by formula (1) is dissolved in a solvent and left for a long time. If the molar ratio

15 is greater than 10 mol, there is a risk of reduced heat resistance, or that the compound represented by formula (g) may become excessive and remain in the reaction product, impairing the low dielectric properties. Furthermore, the total number of moles of compounds selected from one or more of the compounds represented by (i) to (l) reacted with 1 mol of fluorene is preferably 0.01 to 0.99 mol, more preferably 0.02 to 0.9 mol, and even more

preferably 0.03 to 0.8 mol. If the total number of moles of the compounds represented by formulas (i) to (l) reacted with 1 mol of fluorene is less than 0.01 mol, there is a concern that the storage stability after dissolving in a solvent may be deteriorated, such as crystal precipitation when the compound represented by formula (1) is dissolved in a solvent and left for a long period of time. If the total number of moles is greater than 0.99 mol, there is a risk that the compound will not dissolve in the solvent for reaction in the next step, or that the compound represented by formulas (i) to (l) will be in excess and remain in the reaction product containing the compound represented by formula (1), impairing the low dielectric properties.

[0059] When reacting fluorene with a compound represented by formula (g) and one or more compounds selected from the group consisting of compounds represented by formulas (i) to (l), an acidic catalyst may be used, if necessary. Examples of such catalysts include: hydrochloric acid, phosphoric acid, sulfuric acid, formic acid, p-toluenesulfonic acid, and methanesulfonic acid, as well as Lewis acids such as aluminum chloride and zinc chloride; solid acids such as activated clay, acid clay, white carbon, zeolite, silica, and alumina; and acidic ion exchange resins. These may be used alone or in combination. The amount of acidic catalyst used is preferably 0.1 to 40 wt %, preferably 0.1 to 20 wt %, based on the total weight of the fluorene, the compound represented by formula (g), and one or more compounds selected from the group consisting of compounds represented by formulas (i) to (l). If the amount of acidic catalyst used is too large, the viscosity of the reaction solution may become too high, making stirring difficult, while if the amount is too small, the reaction may proceed slowly. The reaction may be carried out using an organic solvent such as hexane, cyclohexane, octane, toluene, or xylene, as needed, or without a solvent.

For example, an acidic catalyst is added to a mixed solution of fluorene, a compound represented by formula (g), and one or more compounds represented by formulas (i) to (l), and an organic solvent (or no solvent), followed by reaction. If the reaction product contains

generated water, the water is removed from the system by azeotropy or other methods. The reaction is then carried out at 40 to 180°C, preferably 50 to 170°C, for 0.5 to 20 hours. After completion of the reaction, the acidic catalyst may be neutralized with an aqueous alkali solution, but the reaction can proceed to a water-washing step without neutralization. For the water-washing step, a water-insoluble organic solvent is added to the oil layer, and washing is repeated until the wastewater becomes neutral.

[0060] Furthermore, the compound represented by formula (A) obtained by the above reaction may be continuously converted into the compound represented by formula (1) by adding a neutralizing agent such as an alkali and an aprotic polar solvent to the solution after the reaction, and then adding a basic catalyst and the compound represented by formula (B).

[0061] The softening point of the compound represented by formula (A) is preferably 180°C or lower, more preferably 150°C or lower. When the softening point of the compound represented by formula (A) is 180°C or lower, the viscosity when it is converted into the compound represented by formula (1) is low. This makes it easier to ensure fluidity, does not impair the impregnation ability into fibrous materials such as glass cloth and carbon fiber, and facilitates B-staging, such as prepreg formation. If the viscosity is reduced by increasing the dilution solvent, there is a possibility that the resin containing the compound represented by formula (1) will not sufficiently adhere to the fibrous material in the impregnation step.

[0062] A resin containing a compound represented by the formula (1) (an aggregate of compounds in which n in formula (1) is an integer of 0 or more) can be cured alone by heating or the like, but performance can also be improved by adding various materials to form a curable resin composition.

[0063] [Curing Accelerator]

The curability of the curable resin composition of the present embodiment can be improved by adding a curing accelerator. As the curing accelerator, an anionic curing accelerator that accelerates the curing reaction by generating anions upon irradiation with

ultraviolet light or visible light or heating, or a cationic curing accelerator that accelerates the curing reaction by generating cations upon irradiation with ultraviolet light or visible light or heating, is preferred.

[0064] Examples of anionic curing accelerators include imidazoles such as 2-

5 methylimidazole, 2-ethylimidazole, and 2-ethyl-4-methylimidazole; trialkylamines such as triethylamine and tributylamine; 4-dimethylaminopyridine, benzyldimethylamine, 2,4,6-tris(dimethylaminomethyl)phenol, and 1,8-diazabicyclo(5,4,0)-undecene, with 4-dimethylaminopyridine and 1,8-diazabicyclo(5,4,0)-undecene being preferred. Other examples include phosphines such as triphenylphosphine; and quaternary ammonium salts
10 such as tetrabutylammonium salt, triisopropylmethylammonium salt, trimethyldecanylammonium salt, cetyltrimethylammonium salt, and hexadecyltrimethylammonium hydroxide, but are not limited thereto. These may be used alone or in combination.

[0065] Examples of cationic curing accelerators include quaternary phosphonium salts such

15 as triphenylbenzylphosphonium salt, triphenylethylphosphonium salt, and tetrabutylphosphonium salt (the counter ion of the quaternary salt may be a halogen, an organic acid ion, a hydroxide ion, or the like, and is not particularly specified, but an organic acid ion or a hydroxide ion is particularly preferred), and transition metal compounds (transition metal salts) such as tin octylate, zinc carboxylate (zinc 2-ethylhexanoate, zinc
20 stearate, zinc behenate, zinc myristate), and zinc phosphate ester (zinc octylphosphate, zinc stearylphosphate), but are not limited thereto. These may be used alone or in combination.

[0066] The curing accelerator is used in an amount of 0.01 to 5.0 parts by mass based on 100 parts by mass of the curable resin composition, as needed.

[0067] [Inorganic Filler]

25 The curable resin composition of the present embodiment may contain an inorganic filler. Examples of inorganic fillers include, but are not limited to, powders such as fused

silica, crystalline silica, porous silica, alumina, zircon, calcium silicate, calcium carbonate, quartz powder, silicon carbide, silicon nitride, boron nitride, zirconia, aluminum nitride, graphite, forsterite, steatite, spinel, mullite, titania, talc, clay, iron oxide, asbestos, and glass powder, as well as inorganic fillers obtained by shaping these into spherical or crushed forms.

5 These fillers may be used alone or in combination.

[0068] When a curable resin composition for semiconductor encapsulation is obtained, the amount of inorganic filler used is preferably 80 to 92 parts by mass, and more preferably 83 to 90 parts by mass, per 100 parts by mass of the curable resin composition. When a curable resin composition for interlayer insulating layer formation, copper-clad laminates, prepregs, 10 RCC (Resin Coated Copper), and other substrate materials is obtained, the amount of inorganic filler used is preferably 5 to 80 parts by mass, and more preferably 10 to 60 parts by mass, per 100 parts by mass of the curable resin composition.

[0069] [Polymerization Initiator]

The curability of the curable resin composition of the present embodiment can be 15 improved by adding a polymerization initiator. The polymerization initiator is a compound capable of polymerizing olefin functional groups such as ethylenically unsaturated bonds, and examples thereof include olefin metathesis polymerization initiators, anionic polymerization initiators, cationic polymerization initiators, and radical polymerization initiators. Among these, it is preferable to use a radical polymerization initiator that has curability and 20 appropriate stability. A radical polymerization initiator is a compound that generates radicals upon irradiation with ultraviolet or visible light or heating, thereby initiating a chain polymerization reaction. Usable radical polymerization initiators include organic peroxides, azo compounds, and benzopinacols. Organic peroxides are preferred because they are effective in controlling the curing temperature, suppress outgassing, and minimize the impact 25 of decomposition products on electrical properties.

[0070] Examples of the organic peroxides include, but not limited to: ketone peroxides such

as methyl ethyl ketone peroxide and acetylacetone peroxide; diacyl peroxides such as benzoyl peroxide; dialkyl peroxides such as dicumyl peroxide and 1,3-bis-(t-butylperoxyisopropyl)-benzene; peroxyketals such as t-butyl peroxybenzoate and 1,1-di-t-butylperoxycyclohexane; alkyl peresters such as α -cumylperoxyneodecanoate, t-butylperoxyneodecanoate, t-butylperoxypivalate, 1,1,3,3-tetramethylbutylperoxy-2-ethylhexanoate, t-amylperoxy-2-ethylhexanoate, t-butylperoxy-2-ethylhexanoate, t-amylperoxy-3,5,5-trimethylhexanoate, t-butylperoxy-3,5,5-trimethylhexanoate, and t-amylperoxybenzoate; peroxycarbonates such as di-2-ethylhexyl peroxydicarbonate, bis(4-t-butylcyclohexyl)peroxydicarbonate, t-butylperoxyisopropyl carbonate, and 1,6-bis(t-butylperoxycarbonyloxy)hexane; t-butyl hydroperoxide, cumene hydroperoxide, t-butyl peroxyoctoate, and lauroyl peroxide. These peroxycarbonates may be used alone or in combination. Among the above organic peroxides, ketone peroxides, diacyl peroxides, hydroperoxides, dialkyl peroxides, peroxyketals, alkyl peresters, peroxycarbonates, etc. are preferred, with dialkyl peroxides being more preferred.

[0071] Examples of the azo compounds include, but are not limited to,

azobisisobutyronitrile, 4,4'-azobis(4-cyanovaleric acid), 2,2'-azobis(2,4-dimethylvaleronitrile), etc. These compounds may be used alone or in combination.

[0072] The amount of polymerization initiator added is preferably 0.01 to 5 parts by mass, and particularly preferably 0.01 to 3 parts by mass, relative to 100 parts by mass of the curable resin composition. If the amount of polymerization initiator used is less than 0.01 part by mass, the molecular weight may not be sufficiently elongated during the polymerization reaction, while if it is more than 5 parts by mass, the dielectric properties such as the dielectric constant and dielectric loss tangent may be impaired.

[0073] [Polymerization inhibitor]

The curable resin composition of the present embodiment may contain a polymerization inhibitor. The inclusion of a polymerization inhibitor improves storage

stability and allows the reaction initiation temperature to be controlled. Controlling the reaction initiation temperature makes it easier to ensure fluidity, prevents impregnation into glass cloth and the like from being impaired, and facilitates B-staging, such as prepreg formation. If the polymerization reaction proceeds too much during prepreg formation, problems such as difficulty in lamination during the lamination process are likely to occur.

[0074] The polymerization inhibitor may be added during or after the synthesis of the compound of the present embodiment. The amount of the polymerization inhibitor used is 0.008 to 1 part by weight, preferably 0.01 to 0.5 parts by weight, based on 100 parts by weight of the compound of the present embodiment.

[0075] Examples of the polymerization inhibitor include phenol-based, sulfur-based, phosphorus-based, hindered amine-based, nitroso-based, and nitroxyl radical-based. One type of polymerization inhibitor may be used alone, or multiple types may be used in combination. Among them, in the present embodiment, phenol-based, hindered amine-based, nitroso-based, and nitroxyl radical-based inhibitors are preferred.

[0076] Examples of the phenol-based polymerization inhibitor include, but are not limited to: monophenols such as 2,6-di-*t*-butyl-*p*-cresol, butylated hydroxyanisole, 2,6-di-*t*-butyl-*p*-ethylphenol, stearyl- β -(3,5-di-*t*-butyl-4-hydroxyphenyl)propionate, isooctyl-3-(3,5-di-*t*-butyl-4-hydroxyphenyl)propionate, 2,4-bis-(*n*-octylthio)-6-(4-hydroxy-3,5-di-*t*-butylanilino)-1,3,5-triazine, and 2,4-bis[(octylthio)methyl]-*o*-cresol; bisphenols such as 2,2'-methylenebis(4-methyl-6-*t*-butylphenol), 2,2'-methylenebis(4-ethyl-6-*t*-butylphenol), 4,4'-thiobis(3-methyl-6-*t*-butylphenol), 4,4'-butylidenebis(3-methyl-6-*t*-butylphenol), triethylene glycol-bis[3-(3-*t*-butyl-5-methyl-4-hydroxyphenyl)propionate], 1,6-hexanediol-bis[3-(3,5-di-*t*-butyl-4-hydroxyphenyl)propionate], *N,N'*-hexamethylenebis(3,5-di-*t*-butyl-4-hydroxyhydrocinnamamide), 2,2'-thio-diethylenebis[3-(3,5-di-*t*-butyl-4-hydroxyphenyl)propionate], 3,5-di-*t*-butyl-4-hydroxybenzylphosphonate-diethyl ester, 3,9-bis[1,1-dimethyl-2- $\{\beta$ -(3-*t*-butyl-4-hydroxy-5-methylphenyl)propionyloxy $\}$ ethyl]2,4,8,10-tetraoxaspiro[5,5]undecane,

bis(3,5-di-t-butyl-4-hydroxybenzylsulfonate ethyl)calcium; polymeric phenols such as 1,1,3-tris(2-methyl-4-hydroxy-5-t-butylphenyl)butane, 1,3,5-trimethyl-2,4,6-tris(3,5-di-t-butyl-4-hydroxybenzyl)benzene, tetrakis-[methylene-3-(3',5'-di-t-butyl-4'-hydroxyphenyl)propionate]methane, bis[3,3'-bis-(4'-hydroxy-3'-t-butylphenyl)butyric acid]glycol ester, tris-(3,5-di-t-butyl-4-hydroxybenzyl)isocyanurate, 1,3,5-tris(3',5'-di-t-butyl-4'-hydroxybenzyl)-S-triazine-2,4,6-(1H,3H,5H)trione, tocopherol.

[0077] Examples of the sulfur-based polymerization inhibitor include, but are not limited to, dilauryl-3,3'-thiodipropionate, dimyristyl-3,3'-thiodipropionate, and distearyl-3,3'-thiodipropionate.

[0078] Examples of the phosphorus-based polymerization inhibitor include, but are not limited to: phosphites such as triphenyl phosphite, diphenyl isodecyl phosphite, phenyl diisodecyl phosphite, tris(nonylphenyl) phosphite, diisodecyl pentaerythritol phosphite, tris(2,4-di-t-butylphenyl) phosphite, cyclic neopentane tetrayl bis(octadecyl) phosphite, cyclic neopentane tetrayl bis(2,4-di-t-butylphenyl) phosphite, cyclic neopentane tetrayl bis(2,4-di-t-butyl-4-methylphenyl) phosphite, bis[2-t-butyl-6-methyl-4-{2-(octadecyloxycarbonyl)ethyl}phenyl]hydrogen phosphite; and oxaphosphaphenanthrene oxides such as 9,10-dihydro-9-oxa-10-phosphaphenanthrene-10-oxide, 10-(3,5-di-t-butyl-4-hydroxybenzyl)-9,10-dihydro-9-oxa-10-phosphaphenanthrene-10-oxide, and 10-decyloxy-9,10-dihydro-9-oxa-10-phosphaphenanthrene-10-oxide.

[0079] Examples of the hindered amine polymerization inhibitor include, but are not limited to: ADK STAB (registered trademark) LA-40MP, ADK STAB LA-40Si, ADK STAB LA-402AF, ADK STAB LA-87, ADK STAB LA-82, ADK STAB LA-81, ADK STAB LA-77Y, ADK STAB LA-77G, ADK STAB LA-72, ADK STAB LA-68, ADK STAB LA-63P, ADK STAB LA-57, and ADK STAB LA-52 (all manufactured by ADE Corporation); and Chimassorb (registered trademark) 2020FDL, Chimassorb 944FDL, Chimassorb 944LD, Tinuvin (registered trademark) 622SF, Tinuvin PA144, Tinuvin 765, Tinuvin 770DF, Tinuvin

XT55FB, Tinuvin 111FDL, Tinuvin 783FDL, Tinuvin 791FB (all manufactured by BASF).

[0080] Examples of the nitroso-based polymerization inhibitor include, but are not limited to, p-nitrosophenol, N-nitrosodiphenylamine, and N-nitrosophenylhydroxyamine ammonium salt (cupferron). Among them, N-nitrosophenylhydroxyamine ammonium salt (cupferron) is preferred.

[0081] Examples of the nitroxyl radical polymerization inhibitor include, but are not limited to: di-tert-butyl nitroxide, 2,2,6,6-tetramethylpiperidine-1-oxyl, 4-hydroxy-2,2,6,6-tetramethylpiperidine-1-oxyl, 4-oxo-2,2,6,6-tetramethylpiperidine-1-oxyl, 4-amino-2,2,6,6-tetramethylpiperidine-1-oxyl, 4-methoxy-2,2,6,6-tetramethylpiperidine-1-oxyl, 4-acetoxy-2,2,6,6-tetramethylpiperidine-1-oxyl, and 4-benzoyloxy-2,2,6,6-tetramethylpiperidine-1-oxyl.

[0082] [Flame Retardant]

The curable resin composition of the present embodiment may contain a flame retardant. Examples of the flame retardant include halogen-based flame retardants, inorganic flame retardants (antimony compounds, metal hydroxides, nitrogen compounds, boron compounds, etc.), and phosphorus-based flame retardants. From the viewpoint of achieving halogen-free flame retardancy, phosphorus-based flame retardants are preferred.

[0083] The phosphorus-based flame retardant may be either a reactive type or an additive type. Specific examples include, but are not limited to: phosphate esters such as trimethyl phosphate, triethyl phosphate, tricresyl phosphate, trixylylene phosphate, cresyl diphenyl phosphate, cresyl-2,6-dixylylene phosphate, 1,3-phenylenebis(dixylylene phosphate), 1,4-phenylenebis(dixylylene phosphate), and 4,4'-biphenyl(dixylylene phosphate); phosphanes such as 9,10-dihydro-9-oxa-10-phosphaphenanthrene-10-oxide and 10-(2,5-dihydroxyphenyl)-10H-9-oxa-10-phosphaphenanthrene-10-oxide; phosphorus-containing epoxy compounds obtained by reacting an epoxy resin with the active hydrogen of the phosphanes; and red phosphorus. These may be used alone or in combination. Of the above-listed substances, phosphate esters, phosphanes, and phosphorus-containing epoxy

compounds are preferred, with 1,3-phenylenebis(dixylilanyl phosphate), 1,4-phenylenebis(dixylilene phosphate), 4,4'-biphenyl(dixylilene phosphate), and phosphorus-containing epoxy compounds being particularly preferred.

[0084] The content of the flame retardant is preferably in the range of 0.1 to 0.6 parts by mass per 100 parts by mass of the curable resin composition. If the content is less than 0.1 part by mass, the flame retardancy may be insufficient, and if the content is more than 0.6 part by mass, the moisture absorption and dielectric properties of the cured product may be adversely affected.

[0085] [Light Stabilizer]

The curable resin composition of the present embodiment may contain a light stabilizer. As the light stabilizer, a hindered amine light stabilizer (Hindered Amine Light Stabilizers, HALS) or the like is suitable. Examples of HALS include, but are not limited to: reaction products of dibutylamine-1,3,5-triazine-N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexamethylenediamine and N-(2,2,6,6-tetramethyl-4-piperidyl)butylamine, reaction products of dimethyl succinate-1-(2-hydroxyethyl)-4-hydroxy-2,2,6,6-tetramethylpiperidine, poly[{6-(1,1,3,3-tetramethylbutyl)amino-1,3,5-triazine-2,4-diyl} {(2,2,6,6-tetramethyl-4-piperidyl)imino} hexamethylene {(2,2,6,6-tetramethyl-4-piperidyl)imino}], bis(1,2,2,6,6-pentamethyl-4-piperidyl)[[3,5-bis(1,1-dimethylethyl)-4-hydroxyphenyl]methyl]butyl malonate, bis(2,2,6,6-tetramethyl-4-piperidyl)sebacate, bis(1,2,2,6,6-pentamethyl-4-piperidyl)sebacate, bis(1-octyloxy-2,2,6,6-tetramethyl-4-piperidyl)sebacate, 2-(3,5-di-t-butyl-4-hydroxybenzyl)-2-n-butylmalonate bis(1,2,2,6,6-pentamethyl-4-piperidyl), and the like. These may be used alone or in combination.

[0086] The content of the light stabilizer is preferably in the range of 0.001 to 0.1 parts by mass relative to 100 parts by mass of the curable resin composition. If the content is less than 0.001 part by mass, the light stabilizing effect may be insufficient, and if the content is more than 0.1 part by mass, the moisture absorption and dielectric properties of the cured product

may be adversely affected.

[0087] [Binder Resin]

The curable resin composition of the present embodiment may use a binder resin.

Examples of binder resins include, but are not limited to: butyral resins, acetal resins, acrylic

5 resins, epoxy-nylon resins, NBR-phenolic resins, epoxy-NBR resins, and silicone resins.

These may be used alone or in combination.

[0088] The blending amount of the binder resin is preferably within a range that does not impair the flame retardancy and heat resistance of the cured product, and is preferably 0.05 to

10 50 parts by mass, and more preferably 0.05 to 20 parts by mass, per 100 parts by mass of the curable resin composition, as needed.

[0089] [Additives]

The curable resin composition of the present embodiment may contain additives.

Example of additives include: modified acrylonitrile copolymers, polyethylene, fluororesins, silicone gels, silicone oils; surface treatment agents for fillers such as silane coupling agents; 15 release agents; and colorants such as carbon black, phthalocyanine blue, and phthalocyanine green.

[0090] The amount of the additive to be added is preferably 30 parts by mass or less, more preferably 20 parts by mass or less, and particularly preferably 10 parts by mass or less, per 100 parts by mass of the curable resin composition.

20 [0091] The curable resin composition of the present embodiment may further contain epoxy resins, active ester compounds, phenolic resins, polyphenylene ether compounds, amine resins, compounds having ethylenically unsaturated bonds, isocyanate resins, polyamide resins, maleimide compounds, cyanate ester resins, polyimide resins, polybutadiene and modified products thereof, polystyrene and modified products thereof, polyethylene and 25 modified products thereof, etc., and these may be used alone or in combination. Among these compounds, polyphenylene ether compounds, compounds having ethylenically unsaturated

bonds, cyanate ester resins, polybutadiene and modified products thereof, and polystyrene and modified products thereof are preferred in terms of the balance of heat resistance, adhesion, and dielectric properties. The inclusion of these compounds can improve the brittleness of the cured product and adhesion to metals, thereby suppressing package cracking during solder reflow and reliability tests such as thermal cycling. Unless otherwise specified, the total amount of the above compounds used is preferably 10 times or less by weight, more preferably 5 times or less by weight, and particularly preferably 3 times or less by weight, relative to the compound of the present embodiment. The lower limit is preferably 0.1 times by mass or more, more preferably 0.25 times by mass or more, and even more preferably 0.5 times by mass or more. By being within the above range, the effect of the low dielectric properties of the compound of the present embodiment can be utilized while the effects of each compound added can be enhanced. As these components, the following examples can be used.

[0092] [Epoxy Resin]

Preferred examples of epoxy resins are listed below, but are not limited thereto. The epoxy resin may be liquid or solid, and may be used alone or in combination.

[0093] Examples of liquid epoxy resins include bisphenol A type epoxy resins, bisphenol F type epoxy resins, bisphenol AF type epoxy resins, naphthalene type epoxy resins, glycidyl ester type epoxy resins, glycidyl amine type epoxy resins, phenol novolac type epoxy resins, alicyclic epoxy resins having an ester skeleton, cyclohexane type epoxy resins, cyclohexanedimethanol type epoxy resins, and epoxy resins having a butadiene structure. Specific examples include "RE310S" and "RE410S" (all manufactured by Nippon Kayaku Co., Ltd., bisphenol A type epoxy resins), "RE303S", "RE304S", "RE403S", and "RE404S" (all manufactured by Nippon Kayaku Co., Ltd., bisphenol F type epoxy resins), "HP-4032", "HP-4032D", and "HP-4032SS" (all manufactured by DIC Corporation, naphthalene type epoxy resins), "jER (registered trademark) 828US", "jER828EL", and "jER825" (all

manufactured by Mitsubishi Chemical Corporation, bisphenol A type epoxy resins), "jER807", and "jER1750" (all manufactured by Mitsubishi Chemical Corporation, bisphenol F type epoxy resins), and "jER152" (manufactured by Mitsubishi Chemical Corporation, phenol novolac type epoxy resin), "jER630", "jER630LSD" (all manufactured by Mitsubishi Chemical Corporation, glycidylamine type epoxy resin), "ZX1059" (manufactured by Nippon Steel & Sumitomo Metal Chemical Co., Ltd., a mixture of bisphenol A type epoxy resin and bisphenol F type epoxy resin), "EX-721" (manufactured by Nagase ChemteX Corporation, glycidyl ester type epoxy resin), "Celloxide (registered trademark) 2021P" (manufactured by Daicel Corporation, alicyclic epoxy resin having an ester skeleton), "PB-3600" (manufactured by Daicel Corporation, epoxy resin having a butadiene structure), "ZX1658", "ZX1658GS" (all manufactured by Nippon Steel & Sumitomo Metal Chemical Co., Ltd., liquid 1,4-glycidylcyclohexane type epoxy resin). These may be used alone or in combination of two or more.

[0094] Examples of solid epoxy resins include bixylenol-type epoxy resins, naphthalene-type epoxy resins, naphthalene-type tetrafunctional epoxy resins, cresol novolac-type epoxy resins, dicyclopentadiene-type epoxy resins, trisphenol-type epoxy resins, naphthol-type epoxy resins, biphenyl-type epoxy resins, naphthylene ether-type epoxy resins, anthracene-type epoxy resins, bisphenol A-type epoxy resins, bisphenol AF-type epoxy resins, and tetraphenylethane-type epoxy resins, and preferred examples of such solid epoxy resins include naphthol-type epoxy resins, bisphenol AF-type epoxy resins, naphthalene-type epoxy resins, and biphenyl-type epoxy resins. Specific examples include "HP4032H" (manufactured by DIC Corporation, naphthalene-type epoxy resin), "HP-4700", and "HP-4710" (all manufactured by DIC Corporation, naphthalene-type tetrafunctional epoxy resin), "N-690" (manufactured by DIC Corporation, cresol novolac-type epoxy resin), "N-695" (manufactured by DIC Corporation, cresol novolac-type epoxy resin), "HP-7200", "HP-7200HH", and "HP-7200H" (all manufactured by DIC Corporation, dicyclopentadiene-type epoxy resin). epoxy

resin), "EXA-7311", "EXA-7311-G3", "EXA-7311-G4", "EXA-7311-G4S", "HP-6000" (all manufactured by DIC Corporation, naphthylene ether type epoxy resin), "EPPN-502H" (manufactured by Nippon Kayaku Co., Ltd., trisphenol type epoxy resin), "NC-7000L", "NC-7300" (all manufactured by Nippon Kayaku Co., Ltd., naphthol-cresol novolac type epoxy resin), "NC-3000H", "NC-3000", "NC-3000L", "NC-3100" (all manufactured by Nippon Kayaku Co., Ltd., biphenyl aralkyl type epoxy resin), "XD-1000-2L", "XD-1000-L", "XD-1000-H" (all manufactured by Nippon Kayaku Co., Ltd., dicyclopentadiene-type epoxy resin), "ESN475V" (manufactured by Nippon Steel & Sumikin Chemical Co., Ltd., naphthol-type epoxy resin), "ESN485" (manufactured by Nippon Steel & Sumikin Chemical Co., Ltd., naphthol novolac-type epoxy resin), "YX-4000H", "YX-4000", "YL6121" (all manufactured by Mitsubishi Chemical Corporation, biphenyl-type epoxy resin), "YX-4000HK" (manufactured by Mitsubishi Chemical Corporation, bixylenol-type epoxy resin), "YX-8800" (manufactured by Mitsubishi Chemical Corporation, anthracene-type epoxy resin), "PG-100" and "CG-500" (manufactured by Osaka Gas Chemicals Co., Ltd., fluorene-based epoxy resin), "YL-7760" (manufactured by Mitsubishi Chemical Corporation, bisphenol AF-type epoxy resin), "YL-7800" (manufactured by Mitsubishi Chemical Corporation, fluorene-type epoxy resin), "jER1010" (manufactured by Mitsubishi Chemical Corporation, solid bisphenol A-type epoxy resin), and "jER1031S" (manufactured by Mitsubishi Chemical Corporation, tetraphenylethane-type epoxy resin). These may be used alone or in combination of two or more.

[0095] [Active Ester Compound]

An active ester compound refers to a compound containing at least one ester bond in its structure, with an aliphatic chain, an aliphatic ring, or an aromatic ring bonded to both sides of the ester bond. Examples of active ester compounds include: phenol esters, thiophenol esters, N-hydroxyamine esters, and esters of heterocyclic hydroxy compounds, which have two or more highly reactive ester groups per molecule. These active ester

compounds are obtained by a condensation reaction between at least one of a carboxylic acid compound, an acid chloride, or a thiocarboxylic acid compound and at least one of a hydroxy compound or a thiol compound. From the viewpoint of improving heat resistance, active ester compounds obtained from a reaction of a carboxylic acid compound or an acid chloride and a hydroxy compound are particularly preferred, with phenol compounds or naphthol compounds being preferred as the hydroxy compound. Active ester compounds may be used alone or in combination of two or more.

[0096] Examples of the carboxylic acid compound include benzoic acid, acetic acid, succinic acid, maleic acid, itaconic acid, phthalic acid, isophthalic acid, terephthalic acid, and pyromellitic acid.

[0097] Examples of the acid chloride include acetyl chloride, acrylic acid chloride, methacrylic acid chloride, malonyl chloride, succinic acid dichloride, diglycolyl chloride, glutaric acid dichloride, suberic acid dichloride, sebacic acid dichloride, adipic acid dichloride, dodecandioyl dichloride, azelaoyl chloride, 2,5-furandicarbonyl dichloride, phthaloyl chloride, isophthaloyl chloride, terephthaloyl chloride, trimesic acid chloride, bis(4-chlorocarbonylphenyl) ether, 4,4'-diphenyldicarbonyl chloride, and 4,4'-azodibenzoyl dichloride.

[0098] Examples of the phenol compound and naphthol compound include hydroquinone, resorcinol, bisphenol A, bisphenol F, bisphenol S, phenolphthalene, methylated bisphenol A, methylated bisphenol F, methylated bisphenol S, phenol, o-cresol, m-cresol, p-cresol, catechol, α -naphthol, β -naphthol, 1,5-dihydroxynaphthalene, 1,6-dihydroxynaphthalene, 2,6-dihydroxynaphthalene, dihydroxybenzophenone, trihydroxybenzophenone, tetrahydroxybenzophenone, phloroglucinol, benzenetriol, dicyclopentadiene-type diphenol compounds, phenol novolac, and the phenolic resins described below. Here, the term "dicyclopentadiene-type diphenol compound" refers to a diphenol compound obtained by condensing one molecule of dicyclopentadiene with two molecules of phenol.

[0099] Preferred specific examples of the active ester compound include active ester compounds containing a dicyclopentadiene-type diphenol structure, active ester compounds containing a naphthalene structure, active ester compounds containing an acetylated phenol novolac, active ester compounds containing a benzoylated phenol novolac, the compound described in Example 2 of WO 2020/095829, and the compounds disclosed in WO 2020/059625. Among them, active ester compounds containing a naphthalene structure and active ester compounds containing a dicyclopentadiene-type diphenol structure are more preferred. The dicyclopentadiene-type diphenol structure refers to a divalent structural unit consisting of phenylene-dicyclopentylene-phenylene.

[0100] Commercially available active ester compounds include, for example: active ester compounds containing a dicyclopentadiene-type diphenol structure such as "EXB9451," "EXB9460," "EXB9460S," "HPC-8000-65T," "HPC-8000H-65TM," "EXB-8000L-65TM," and "EXB-8150-65T" (manufactured by DIC Corporation); an active ester compound containing a naphthalene structure such as "EXB9416-70BK" (manufactured by DIC Corporation); an active ester compound containing an acetylated phenol novolac such as "DC808" (manufactured by Mitsubishi Chemical Corporation); an active ester compound containing a benzoylated phenol novolac such as "YLH1026," "YLH1030," and "YLH1048" (manufactured by Mitsubishi Chemical Corporation); and a phosphorus atom-containing active ester curing agent such as "EXB-9050L-62M" (manufactured by DIC Corporation).

[0101] Regarding the compounding ratio of the active ester compound and the epoxy resin, the ratio (α/β) of the active ester equivalent (α) to the epoxy equivalent (β) is preferably 0.5 to 1.5, more preferably 0.8 to 1.2, and even more preferably 0.90 to 1.10. If the ratio is outside the above range, excess epoxy groups or active ester groups may remain in the system, which may deteriorate the properties in a high-temperature storage test (e.g., 150°C, 1000 hours) or a long-term reliability test under high-temperature and high-humidity conditions (e.g., temperature: 85°C, humidity: 85%).

[0102] [Phenol Resin]

A phenol resin is a compound having two or more phenolic hydroxy groups in the molecule. Examples of phenol resins include, but are not limited to, reaction products of phenols and aldehydes, reaction products of phenols and diene compounds, reaction products of phenols and ketones, reaction products of phenols and substituted biphenyls, reaction products of phenols and substituted phenyls, and reaction products of bisphenols and aldehydes. These may be used alone or in combination.

Specific examples of the above raw materials are listed below, but are not limited thereto.

10 <Phenols>

Phenol, alkyl-substituted phenol, aromatic-substituted phenol, hydroquinone, resorcinol, naphthol, alkyl-substituted naphthol, dihydroxybenzene, alkyl-substituted dihydroxybenzene, dihydroxynaphthalene, etc.

<Aldehydes>

15 Formaldehyde, acetaldehyde, alkyl aldehyde, benzaldehyde, alkyl-substituted benzaldehyde, hydroxybenzaldehyde, naphthaldehyde, glutaraldehyde, phthalaldehyde, crotonaldehyde, cinnamaldehyde, furfural, etc.

<Diene compounds>

20 Dicyclopentadiene, terpenes, vinylcyclohexene, norbornadiene, vinylnorbornene, tetrahydroindene, divinylbenzene, divinylbiphenyl, diisopropenylbiphenyl, butadiene, isoprene, etc.

<Ketones>

Acetone, methyl ethyl ketone, methyl isobutyl ketone, acetophenone, benzophenone, fluorenone, etc.

25 <Substituted biphenyls>

4,4'-bis(chloromethyl)-1,1'-biphenyl, 4,4'-bis(methoxymethyl)-1,1'-biphenyl, 4,4'-

bis(hydroxymethyl)-1,1'-biphenyl, etc.

<Substituted phenyls>

1,4-bis(chloromethyl)benzene, 1,4-bis(methoxymethyl)benzene, 1,4-bis(hydroxymethyl)benzene, etc.

5 [0103] [Polyphenylene Ether Compound]

From the viewpoint of heat resistance and electrical properties, the polyphenylene ether compound is preferably a polyphenylene ether compound having an ethylenically unsaturated bond, and more preferably a polyphenylene ether compound having an acrylic group, a methacrylic group, or a styrene structure. Commercially available products include
 10 SA-9000 (manufactured by SABIC Corporation, a polyphenylene ether compound having a methacrylic group) and OPE-2St 1200 (manufactured by Mitsubishi Gas Chemical Company, a polyphenylene ether compound having a styrene structure).

The number average molecular weight (M_n) of the polyphenylene ether compound is preferably 500 to 5,000, more preferably 2,000 to 5,000, and even more preferably 2,000 to
 15 4,000. If the number average molecular weight is less than 500, the heat resistance of the cured product tends to be insufficient. Furthermore, if the number average molecular weight is greater than 5,000, the melt viscosity increases, and sufficient fluidity cannot be obtained, which tends to result in molding defects. Furthermore, the reactivity decreases, the curing reaction takes a long time, and the amount of unreacted polyphenylene ether not incorporated
 20 into the curing system increases, which lowers the glass transition temperature of the cured product and reduces the heat resistance of the cured product.

If the number average molecular weight of the polyphenylene ether compound is 500 to 5,000, it is possible to exhibit excellent heat resistance and moldability while maintaining excellent dielectric properties. The number average molecular weight here can be measured
 25 specifically using gel permeation chromatography or the like.

[0104] The polyphenylene ether compound may be one obtained by a polymerization

reaction or one obtained by a redistribution reaction of a high-molecular-weight polyphenylene ether compound having a number-average molecular weight of approximately 10,000 to 30,000. Furthermore, these compounds may be used as raw materials and reacted with a compound having an ethylenically unsaturated bond, such as methacrylic acid chloride, acrylic acid chloride, or chloromethylstyrene, to impart radical polymerizability. A polyphenylene ether compound obtained by a redistribution reaction may be obtained, for example, by heating a high-molecular-weight polyphenylene ether compound in a solvent such as toluene in the presence of a phenolic compound and a radical initiator to cause a redistribution reaction. Such polyphenylene ether compounds obtained by a redistribution reaction are preferred because they have hydroxy groups derived from the phenolic compound at both ends of the molecular chain that contribute to curing, thereby maintaining even higher heat resistance. Furthermore, functional groups can be introduced at both ends of the molecular chain even after modification with a compound having an ethylenically unsaturated bond. Furthermore, polyphenylene ether compounds obtained by a polymerization reaction are preferred because they exhibit excellent fluidity.

[0105] In the case of polyphenylene ether compounds obtained by polymerization, the molecular weight of the polyphenylene ether compound can be adjusted by adjusting the polymerization conditions, etc. In the case of polyphenylene ether compounds obtained by redistribution, the molecular weight of the resulting polyphenylene ether compound can be adjusted by adjusting the conditions, etc. of the redistribution reaction. More specifically, adjusting the amount of the phenolic compound used in the redistribution reaction can be considered. That is, the greater the amount of the phenolic compound, the lower the molecular weight of the resulting polyphenylene ether compound. In this case, poly(2,6-dimethyl-1,4-phenylene ether) or the like can be used as the high-molecular-weight polyphenylene ether compound that undergoes the redistribution reaction. Furthermore, the phenolic compound used in the redistribution reaction is not particularly limited, but preferred are, for example,

polyfunctional phenolic compounds having two or more phenolic hydroxy groups per molecule, such as bisphenol A, phenol novolac, and cresol novolac. These compounds may be used alone or in combination of two or more.

[0106] The content of the polyphenylene ether compound is not particularly limited, but is preferably 5 to 1,000 parts by mass, and more preferably 10 to 750 parts by mass, relative to 100 parts by mass of the curable resin composition. A content of the polyphenylene ether compound within the above range is preferable in that it not only has excellent heat resistance and the like, but also allows a cured product to be obtained that fully exhibits the excellent dielectric properties of the polyphenylene ether compound.

[0107] [Amine Resin]

Amine resin is a compound having two or more amino groups in the molecule.

Examples of amine resins include, but are not limited to: diaminodiphenylmethane, diaminodiphenylsulfone, isophoronediamine, naphthalenediamine, aniline novolac (a reaction product of aniline and formalin), N-methylaniline novolac (a reaction product of N-methylaniline and formalin), orthoethylaniline novolac (a reaction product of orthoethylaniline and formalin), a reaction product of 2-methylaniline and formalin, a reaction product of 2,6-diisopropylaniline and formalin, a reaction product of 2,6-diethylaniline and formalin, a reaction product of 2-ethyl-6-ethylaniline and formalin, a reaction product of 2,6-dimethylaniline and formalin, and aniline resin obtained by the reaction of aniline and xylylene chloride, reaction products of aniline and substituted biphenyls (such as 4,4'-bis(chloromethyl)-1,1'-biphenyl and 4,4'-bis(methoxymethyl)-1,1'-biphenyl) disclosed in Japanese Patent No. 6,429,862, reaction products of aniline and substituted phenyls (such as 1,4-bis(chloromethyl)benzene, 1,4-bis(methoxymethyl)benzene and 1,4-bis(hydroxymethyl)benzene), 4,4'-(1,3-phenylenediisopropylidene)bisaniline, 4,4'-(1,4-phenylenediisopropylidene)bisaniline, reaction products of aniline and diisopropenylbenzene, and dimer diamine. These may be used alone or in combination.

[0108] [Compound containing an ethylenically unsaturated bond]

The compound containing an ethylenically unsaturated bond is a compound having one or more ethylenically unsaturated bonds in the molecule that can be polymerized by heat or light, regardless of whether a polymerization initiator is used.

5 Examples of compounds containing an ethylenically unsaturated bond include, but are not limited to: reaction products of the phenol resins with ethylenically unsaturated bond-containing halogen-based compounds (chloromethylstyrene, allyl chloride, methallyl chloride, acrylic acid chloride, methacrylic acid chloride, etc.); reaction products of ethylenically unsaturated bond-containing phenols (2-allylphenol, 2-propenylphenol, 4-allylphenol, 4-
10 propenylphenol, eugenol, isoeugenol, etc.) with halogen-based compounds (1,4-bis(chloromethyl)benzene, 4,4'-bis(chloromethyl)biphenyl, 4,4'-difluorobenzophenone, 4,4'-dichlorobenzophenone, 4,4'-dibromobenzophenone, cyanuric chloride, etc.); reaction products of epoxy resins or alcohols with (meth)acrylic acids (acrylic acid, methacrylic acid, etc.), and acid-modified products thereof. These compounds may be used alone or in combination.

15 [0109] [Isocyanate Resin]

An isocyanate resin is a compound having two or more isocyanate groups in the molecule. Examples of the isocyanate resin include, but are not limited to: aromatic diisocyanates such as p-phenylene diisocyanate, m-phenylene diisocyanate, p-xylene diisocyanate, m-xylene diisocyanate, 2,4-tolylene diisocyanate, 2,6-tolylene diisocyanate,
20 4,4'-diphenylmethane diisocyanate, and naphthalene diisocyanate; aliphatic or alicyclic diisocyanates such as isophorone diisocyanate, hexamethylene diisocyanate, 4,4'-dicyclohexylmethane diisocyanate, hydrogenated xylene diisocyanate, norbornene diisocyanate, and lysine diisocyanate; polyisocyanates such as one or more biuret compounds of isocyanate monomers or isocyanate compounds obtained by trimerizing the above
25 diisocyanate compounds; and polyisocyanates obtained by a urethanization reaction between the above isocyanate compounds and polyol compounds. These may be used alone or in

combination.

[0110] [Polyamide Resin]

Examples of polyamide resins include reaction products of one or more of diamines, diisocyanates, and oxazolines with dicarboxylic acids, reaction products of diamines with acid chlorides, and ring-opening polymerization products of lactam compounds. These may be used alone or in combination.

Specific examples of the above raw materials are listed below, but are not limited thereto.

<Diamines>

Ethylenediamine, trimethylenediamine, tetramethylenediamine, pentamethylenediamine, hexamethylenediamine, heptamethylenediamine, octamethylenediamine, nonamethylenediamine, decanediamine, undecanediamine, dodecanediamine, tridecanediamine, tetradecanediamine, pentadecanediamine, hexadecanediamine, heptadecanediamine, octadecanediamine, nonadecanediamine, eicosanediamine, 2-methyl-1,5-diaminopentane, 2-methyl-1,8-diaminooctane, dimer diamine, cyclohexanediamine, bis-(4-aminocyclohexyl)methane, bis(3-methyl-4-aminocyclohexyl)methane, xylylenediamine, norbornanediamine, isophoronediamine, bisaminomethyltricyclodecane, phenylenediamine, diethyltoluenediamine, naphthalenediamine, diaminodiphenylmethane, bis(4-amino-3,5-dimethylphenyl)methane, bis(4-amino-3,5-diethylphenyl)methane) methane, 4,4'-methylenebis-o-toluidine, 4,4'-methylenebis-o-ethylaniline, 4,4'-methylenebis-2-ethyl-6-methylaniline, 4,4'-methylenebis-2,6-diisopropylaniline, 4,4'-ethylenedianiline, diaminodiphenyl sulfone, diaminodiphenyl ether, 1,3-bis(3-aminophenoxy)benzene, 1,3-bis(4-aminophenoxy)benzene, 4,4-bis(4-aminophenoxy)biphenyl, 2,2-bis[4-(4-aminophenoxy)phenyl]propane, bis[4-(4-aminophenoxy)phenyl]sulfone, 2,2-bis(3-amino-4-hydroxyphenyl)propane, 2,2-bis(4-aminophenyl)hexafluoropropane, 4,4'-(1,3-phenylenediisopropylidene)bisaniline, 4,4'-(1,4-

phenylenediisopropylidene)bisaniline, 9,9-bis(4-aminophenyl)fluorene, 2,7-diaminofluorene, aminobenzylamine, diaminobenzophenone, and the like.

<Diisocyanates>

benzene diisocyanate, toluene diisocyanate, 1,3-bis(isocyanatomethyl)benzene, 1,3-bis(isocyanatomethyl)cyclohexane, bis(4-isocyanatophenyl)methane, isophorone diisocyanate, 1,3-bis(2-isocyanato-2-propyl)benzene, 2,2-bis(4-isocyanatophenyl)hexafluoropropane, dicyclohexylmethane-4,4'-diisocyanate, and the like.

<Dicarboxylic acids>

oxalic acid, malonic acid, succinic acid, glutaric acid, adipic acid, suberic acid, azelaic acid, sebacic acid, undecanedioic acid, dodecanedioic acid, terephthalic acid, isophthalic acid, 5-hydroxyisophthalic acid, 2-chloroterephthalic acid, 2-methylterephthalic acid, 5-methylisophthalic acid, 5-sodiumsulfoisophthalic acid, hexahydroterephthalic acid, hexahydroisophthalic acid, cyclohexanedicarboxylic acid, biphenyldicarboxylic acid, naphthalenedicarboxylic acid, benzophenonedicarboxylic acid, furandicarboxylic acid, 4,4'-dicarboxydiphenyl ether, 4,4'-dicarboxydiphenyl sulfide, and the like.

<Acid chlorides>

acetyl chloride, acrylic acid chloride, methacrylic acid chloride, malonyl chloride, succinic acid dichloride, diglycolyl chloride, glutaric acid dichloride, suberic acid dichloride, sebacic acid dichloride, adipic acid dichloride, dodecandioyl dichloride, azelayl chloride, 2,5-furandicarbonyl dichloride, phthaloyl chloride, isophthaloyl chloride, terephthaloyl chloride, trimesic acid chloride, bis(4-chlorocarbonylphenyl) ether, 4,4'-diphenyldicarbonyl chloride, 4,4'-azodibenzoyl dichloride, etc.

<Lactams>

ϵ -caprolactam, ω -undecanelactam, ω -laurolactam, etc.

[0111] [Polyimide Resin]

Examples of polyimide resins include, but are not limited to, reaction products of the

diamines described above with the tetracarboxylic dianhydrides listed below. These may be used alone or in combination.

<Tetracarboxylic Dianhydrides>

- 4,4'-(hexafluoroisopropylidene)diphthalic anhydride, 5-(2,5-dioxotetrahydro-3-furanyl)-3-methyl-cyclohexene-1,2dicarboxylic anhydride, pyromellitic dianhydride, 1,2,3,4-benzenetetracarboxylic dianhydride, 3,3',4,4'-benzophenonetetracarboxylic dianhydride, 2,2',3,3'-benzophenonetetracarboxylic dianhydride, 3,3',4,4'-biphenyltetracarboxylic dianhydride, 3,3',4,4'-diphenyltetracarboxylic dianhydride, 2,2',3,3'-biphenyltetracarboxylic dianhydride, methylene-4,4'-diphthalic dianhydride, 1,1-ethylidene-4,4'-diphthalic dianhydride, 2,2'-propylidene-4,4'-diphthalic dianhydride, 1,2-ethylene-4,4'-diphthalic dianhydride, 1,3-trimethylene-4,4'-diphthalic dianhydride, 1,4-tetramethylene-4,4'-diphthalic dianhydride, 1,5-pentamethylene-4,4'-diphthalic dianhydride, 4,4'-oxydiphthalic dianhydride, thio-4,4'-diphthalic dianhydride, sulfonyl-4,4'-diphthalic dianhydride, 1,3-bis(3,4-dicarboxyphenyl)benzene dianhydride, 1,3-bis(3,4-dicarboxyphenoxy)benzene dianhydride, 1,4-bis(3,4-dicarboxyphenoxy)benzene dianhydride, 1,3-bis[2-(3,4-dicarboxyphenyl)-2-propyl]benzene dianhydride, 1,4-bis[2-(3,4-dicarboxyphenyl)-2-propyl]benzene dianhydride, bis[3-(3,4-dicarboxyphenoxy)phenyl]methane dianhydride, bis[4-(3,4-dicarboxyphenoxy)phenyl]methane dianhydride, 2,2-bis[3-(3,4-dicarboxyphenoxy)phenyl]propane dianhydride, 2,2-bis[4-(3,4-dicarboxyphenoxy)phenyl]propane dianhydride, bis(3,4-dicarboxyphenoxy)dimethylsilane dianhydride, 1,3-bis(3,4-dicarboxyphenyl)-1,1,3,3-tetramethyldisiloxane dianhydride, 2,3,6,7-naphthalene tetracarboxylic dianhydride, 1,4,5,8-naphthalene tetracarboxylic dianhydride, 1,2,5,6-naphthalene tetracarboxylic dianhydride, 3,4,9,10-perylene tetracarboxylic dianhydride, 2,3,6,7-anthracene tetracarboxylic dianhydride, 1,2,7,8-phenanthrene tetracarboxylic dianhydride, ethylene tetracarboxylic dianhydride, 1,2,3,4-butane tetracarboxylic dianhydride, 1,2,3,4-cyclobutane tetracarboxylic dianhydride, cyclopentane

tetracarboxylic dianhydride, cyclohexane-1,2,3,4-tetracarboxylic acid dianhydride, cyclohexane-1,2,4,5-tetracarboxylic acid dianhydride, 3,3',4,4'-bicyclohexyltetracarboxylic acid dianhydride, carbonyl-4,4'-bis(cyclohexane-1,2-dicarboxylic acid) dianhydride, methylene-4,4'-bis(cyclohexane-1,2-dicarboxylic acid) dianhydride, 1,2-ethylene-4,4'-bis(cyclohexane-1,2-dicarboxylic acid) dianhydride, 1,1-ethylidene-4,4'-bis(cyclohexane-1,2-dicarboxylic acid) dianhydride, 2,2-propylidene-4,4'-bis(cyclohexane-1,2-dicarboxylic acid) dianhydride, oxy-4,4'-bis(cyclohexane-1,2-dicarboxylic acid) dianhydride, thio-4,4'-bis(cyclohexane-1,2-dicarboxylic acid) dianhydride, sulfonyl-4,4'-bis(cyclohexane-1,2-dicarboxylic acid) dianhydride, bicyclo[2,2,2]oct-7-ene-2,3,5,6-tetracarboxylic acid dianhydride, rel-[1S,5 R,6R]-3-oxabicyclo[3,2,1]octane-2,4-dione-6-spiro-3'-(tetrahydrofuran-2',5'-dione), 4-(2,5-dioxotetrahydrofuran-3-yl)-1,2,3,4-tetrahydronaphthalene-1,2-dicarboxylic anhydride, ethylene glycol bis-(3,4-dicarboxylic anhydride phenyl) ether, 4,4'-biphenyl bis(trimellitic acid monoester acid anhydride), 9,9'-bis(3,4-dicarboxyphenyl)fluorene dianhydride, and the like.

[0112] [Maleimide Compound]

The curable resin composition of the present embodiment may contain a maleimide compound. A maleimide compound is a compound having one or more maleimide groups in the molecule. Examples of the maleimide compound include, but are not limited to: 4,4'-diphenylmethane bismaleimide, polyphenylmethane maleimide, m-phenylene bismaleimide, 2,2'-bis[4-(4-maleimidophenoxy)phenyl]propane, 3,3'-dimethyl-5,5'-diethyl-4,4'-diphenylmethane bismaleimide, 4-methyl-1,3-phenylene bismaleimide, 4,4'-diphenylether bismaleimide, 4,4'-diphenylsulfone bismaleimide, 1,3-bis(3-maleimidophenoxy)benzene, 1,3-bis(4-maleimidophenoxy)benzene, and Xylok-type maleimide compounds (anilix maleimide, manufactured by Mitsui Chemicals Fine Co., Ltd.), biphenylaralkyl-type maleimide compounds (solidified by distilling off the solvent under reduced pressure from a resin solution containing the maleimide compound (M2) described in Example 4 of JP 2009-

001783 A), bisaminocumylbenzene-type maleimide (maleimide compounds described in WO 2020/054601 A), maleimide compounds having an indane structure described in JP 6629692B or WO 2020/217679, maleimide compounds described in MATERIAL STAGE, Vol. 18, No. 12, 2019, "Sequel Epoxy Resin CAS Number Story - Hardener CAS Number Memorandum No. 31 Bismaleimide (1)" and MATERIAL STAGE, Vol. 19, No. 12, 2019, "Sequel Epoxy Resin CAS Number Story - Hardener CAS Number Memorandum No. 32, Bismaleimide (2)". These compounds may be used alone or in combination.

[0113] [Cyanate ester resin]

A cyanate ester resin is a cyanate ester compound obtained by reacting a phenolic resin with a cyanogen halide. Specific examples include dicyanatobenzene, tricyanatobenzene, dicyanatonaphthalene, dicyanatobiphenyl, 2,2'-bis(4-cyanatophenyl)propane, bis(4-cyanatophenyl)methane, bis(3,5-dimethyl-4-cyanatophenyl)methane, 2,2'-bis(3,5-dimethyl-4-cyanatophenyl)propane, 2,2'-bis(4-cyanatophenyl)ethane, 2,2'-bis(4-cyanatophenyl)hexafluoropropane, bis(4-cyanatophenyl)sulfone, bis(4-cyanatophenyl)thioether, phenol novolac cyanate, and phenol-dicyclopentadiene co-condensates in which the hydroxy groups have been converted to cyanate groups, but are not limited thereto. These compounds may be used alone or in combination.

Furthermore, the cyanate ester compounds whose synthesis methods are described in Japanese Patent Laid-Open Publication No. 2005-264154 are particularly preferred as cyanate ester compounds because they have low moisture absorption, excellent flame retardancy, and excellent dielectric properties.

The cyanate ester resin may also contain a catalyst such as zinc naphthenate, cobalt naphthenate, copper naphthenate, lead naphthenate, zinc octoate, tin octoate, lead acetylacetonate, or dibutyltin maleate, in order to trimerize the cyanate group to form a sym-triazine ring, as necessary.

[0114] The catalyst is preferably used in an amount of 0.0001 to 0.10 parts by mass, and more preferably 0.00015 to 0.0015 parts by mass, per 100 parts by mass of the cyanate ester resin and the curable resin composition.

[0115] [Polybutadiene and its modified products]

5 Polybutadiene and its modified products are compounds having polybutadiene or a structure derived from polybutadiene in the molecule. The unsaturated bonds in the polybutadiene-derived structure may be partially or entirely converted to single bonds by hydrogenation.

Examples of polybutadiene and its modified products include, but are not limited to, 10 polybutadiene, hydroxy-terminated polybutadiene, (meth)acrylate-terminated polybutadiene, carboxylic acid-terminated polybutadiene, amine-terminated polybutadiene, and styrene-butadiene rubber. These may be used alone or in combination. Of these, polybutadiene or styrene-butadiene rubber is preferred from the viewpoint of dielectric properties. Examples of styrene-butadiene rubber (SBR) include RICON-100, RICON-181, and RICON-184 (all 15 manufactured by Cray Valley Chemical Industries, Ltd.) and 1,2-SBS (manufactured by Nippon Soda Co., Ltd.). Examples of polybutadienes include B-1000, B-2000, and B-3000 (all manufactured by Nippon Soda Co., Ltd.). The molecular weight of polybutadiene and styrene-butadiene rubber is preferably a weight-average molecular weight of 500 to 10,000, more preferably 750 to 7,500, and even more preferably 1,000 to 5,000. Below the lower limit 20 of the above range, the amount of volatilization is high, making it difficult to adjust the solids content during prepreg production. Above the upper limit of the above range, compatibility with other curable resins deteriorates. In general, in the case of compounds containing heteroatoms such as oxygen and nitrogen, such as bismaleimides and polymaleimides, their polarity makes it difficult to ensure compatibility with low-polarity compounds, such as 25 compounds composed mainly of hydrocarbons or compounds composed only of hydrocarbons. On the other hand, the compound of the present embodiment does not have a

skeleton design in which heteroatoms such as oxygen and nitrogen are actively introduced, and therefore has excellent compatibility with materials having low polarity and low dielectric properties and compounds composed only of hydrocarbons.

[0116] [Polystyrene and its modified products]

5 Polystyrene and its modified products are polystyrene or compounds having a structure derived from polystyrene in the molecule.

Examples of polystyrene and modified products thereof include polystyrene, styrene-2-isopropenyl-2-oxazoline copolymers (Epocross RPS-1005, RP-61, both manufactured by Nippon Shokubai Co., Ltd.), SEP (styrene-ethylene-propylene copolymer: Septon (registered trademark) 1020, manufactured by Kuraray Co., Ltd.), SEPS (styrene-ethylene-propylene-styrene copolymer: Septon 2002, Septon 2004F, Septon 2005, Septon 2006, Septon 2063, Septon 2104, all manufactured by Kuraray Co., Ltd.), SEEPS (styrene-ethylene-ethylene-propylene-styrene block copolymer: Septon 4003, Septon 4044, Septon 4055, Septon 4077, Septon 4099, all manufactured by Kuraray Co., Ltd.), SEBS (styrene-ethylene-butylene-styrene copolymer: SEPTON 8004, SEPTON 8006, and SEPTON 8007L, all manufactured by Kuraray Co., Ltd.), SEEPS-OH (a styrene-ethylene-ethylene-propylene-styrene block copolymer having a hydroxy group at its terminal: SEPTON HG252, manufactured by Kuraray Co., Ltd.), SIS (styrene-isoprene-styrene block copolymer: SEPTON 5125 and SEPTON 5127, both manufactured by Kuraray Co., Ltd.), hydrogenated SIS (hydrogenated styrene-isoprene-styrene block copolymer: HYBRAR (registered trademark) 7125F and HYBRAR 7311F, both manufactured by Kuraray Co., Ltd.), and SIBS (styrene-isobutylene-styrene block copolymer: SIBSTAR (registered trademark) 073T, SIBSTAR 102T, and SIBSTAR 103T (all manufactured by Kaneka Corporation), and SEPTON V9827 (manufactured by Kuraray Co., Ltd.)), but are not limited thereto. These may be used alone or in combination. Polystyrene and modified products thereof are preferably free of unsaturated bonds because they have higher heat resistance and are less susceptible to oxidative

degradation. The weight-average molecular weight of polystyrene and modified products thereof is not particularly limited as long as it is 10,000 or more. However, if it is too large, compatibility with polyphenylene ether compounds, low-molecular-weight components with a weight-average molecular weight of about 50 to 1,000, and oligomer components with a weight-average molecular weight of about 1,000 to 5,000 deteriorates, making it difficult to ensure mixing and solvent stability. Therefore, a weight-average molecular weight of about 10,000 to 300,000 is preferred.

[0117] [Polyethylene and Modified Polyethylenes]

Polyethylene and modified polyethylenes are polyethylene or compounds having a polyethylene-derived structure in the molecule. Examples of polyethylene and modified polyethylenes include, but are not limited to, ethylene-propylene copolymers, ethylene-styrene copolymers, ethylene-propylene-ethylidene norbornene copolymers (EBT: K-8370EM, K-9330M, etc., manufactured by Mitsui Chemicals, Inc.), ethylene-propylene-vinyl norbornene copolymers (VNB-EPT: PX-006M, PX-008M, PX-009M, etc., manufactured by Mitsui Chemicals, Inc.), ethylene-vinyl alcohol copolymers, and ethylene-vinyl acetate copolymers. From the viewpoint of improving heat resistance, it is preferable to use ethylene-propylene-ethylidene norbornene copolymers or ethylene-propylene-vinyl norbornene copolymers containing a crosslinkable structure. These may be used alone or in combination. There are no particular restrictions on the weight-average molecular weight of polyethylene and modified polyethylenes as long as it is 10,000 or more. If it is too large, compatibility with not only the polyphenylene ether compound but also low-molecular-weight components having a weight-average molecular weight of about 50 to 1,000 and oligomer components having a weight-average molecular weight of about 1,000 to 5,000 deteriorates, making it difficult to ensure mixing and solvent stability. Therefore, it is preferably about 10,000 to 300,000.

[0118] The curable resin composition of the present embodiment can be obtained by

preparing the above components in a predetermined ratio, pre-curing the composition at 130 to 180°C for 30 to 500 seconds, and then post-curing the composition at 150 to 200°C for 2 to 15 hours, thereby allowing the curing reaction to proceed sufficiently and producing a cured product of the present embodiment. Alternatively, the components of the curable resin composition can be uniformly dispersed or dissolved in a solvent or the like, and the solvent can be removed before curing.

[0119] The method for preparing the curable resin composition of the present embodiment is not particularly limited, and the components may be simply mixed uniformly, or may be prepolymerized. For example, a mixture containing the compound of the present embodiment is heated in the presence or absence of a curing accelerator or a polymerization initiator, and in the presence or absence of a solvent, to form a prepolymer. Similarly, compounds such as amine compounds, compounds having ethylenically unsaturated bonds, maleimide compounds, cyanate ester compounds, polybutadiene and its modified products, polystyrene and its modified products, inorganic fillers, and other additives may be added to form a prepolymer. The components may be mixed or prepolymerized using, for example, an extruder, kneader, or rolls in the absence of a solvent, or a reaction kettle equipped with a stirrer in the presence of a solvent.

[0120] The uniform mixing method involves kneading the components using a device such as a kneader, roll, or planetary mixer at a temperature within the range of 50 to 100°C to obtain a uniform resin composition. The resulting resin composition can be pulverized and then molded into cylindrical tablets using a molding machine such as a tablet machine, or into a granular powder or powder-like molded body. Alternatively, these compositions can be melted on a surface support and molded into a sheet with a thickness of 0.05 mm to 10 mm to obtain a molded curable resin composition. The resulting molded body is non-sticky at 0 to 20°C, and exhibits little loss of fluidity or curability even when stored at -25 to 0°C for one week or more.

The resulting molded body can be molded into a cured product using a transfer molding machine or compression molding machine.

[0121] The curable resin composition of the present embodiment can also be made into a varnish-like composition (hereinafter simply referred to as varnish) by adding an organic

5 solvent. The curable resin composition of the present embodiment can be dissolved in a solvent such as toluene, xylene, acetone, methyl ethyl ketone, methyl isobutyl ketone, dimethylformamide, dimethylacetamide, or N-methylpyrrolidone, as needed, to form a varnish. This varnish is then impregnated into a substrate such as glass fiber, carbon fiber, polyester fiber, polyamide fiber, alumina fiber, or paper, and heated and dried to obtain a
10 prepreg. This prepreg can then be hot-press molded to form a cured product of the curable resin composition of the present embodiment. The solvent used in this case accounts for 10 to 70 wt %, preferably 15 to 70 wt %, of the mixture of the curable resin composition of the present embodiment and the solvent. Furthermore, if the composition is in liquid form, a cured curable resin containing carbon fiber can also be obtained directly, for example, by the
15 RTM method.

[0122] The curable resin composition of the present embodiment can also be used as a modifier for film-type compositions. Specifically, it can be used to improve flexibility and the like in the B-stage. Such a film-type resin composition can be obtained as a sheet-like adhesive by applying the curable resin composition of the present embodiment as a varnish
20 onto a release film, removing the solvent under heating, and then performing B-staging. This sheet-like adhesive can be used as an interlayer insulating layer in a multilayer substrate or the like.

[0123] A prepreg can be obtained by heating and melting the curable resin composition of the present embodiment to reduce its viscosity and impregnating it into reinforcing fibers such
25 as glass fiber, carbon fiber, polyester fiber, polyamide fiber, and alumina fiber. Specific examples include glass fibers such as E-glass cloth, D-glass cloth, S-glass cloth, Q-glass

cloth, spherical glass cloth, NE-glass cloth, and T-glass cloth; inorganic fibers other than glass; and organic fibers such as polyparaphenylene terephthalamide (Kevlar(registered trademark), manufactured by DuPont), wholly aromatic polyamide, polyester, polyparaphenylene benzoxazole, polyimide, and carbon fiber, but are not limited thereto. The shape of the substrate is not particularly limited, but examples include woven fabric, nonwoven fabric, roving, and chopped strand mat. Known weaving methods for woven fabrics include plain weave, saddle-weave, and twill weave, and these known methods can be appropriately selected and used depending on the intended application and performance. Also preferred are woven fabrics that have been opened or glass woven fabrics that have been surface-treated with a silane coupling agent or the like. The thickness of the substrate is not particularly limited, but is preferably about 0.01 to 0.4 mm. Alternatively, a prepreg can be obtained by impregnating reinforcing fibers with the varnish and drying the varnish by heating.

[0124] A laminate can also be manufactured using the prepreg. The laminate is not

particularly limited as long as it comprises one or more preregs, and may also have any other layers. The method for manufacturing the laminate can be any generally known method, and is not particularly limited. For example, when molding a metal foil-clad laminate, a multi-stage press, a multi-stage vacuum press, a continuous molding machine, an autoclave molding machine, or the like can be used. The preregs are laminated together and then heated and pressure molded to obtain a laminate. The heating temperature is not particularly limited, but is preferably 65 to 300°C, and more preferably 120 to 270°C. The pressure applied is also not particularly limited, but if the pressure is too high, it becomes difficult to adjust the solid content of the resin in the laminate, resulting in unstable quality. If the pressure is too low, air bubbles will form and adhesion between the laminate layers will be poor. Therefore, a pressure of 2.0 to 5.0 MPa is preferred, and 2.5 to 4.0 MPa is more preferred. The laminate of the present embodiment, having a layer made of metal foil, can be suitably used as a metal

foil-clad laminate, as described below.

The prepreg is cut into a desired shape and laminated with copper foil or the like as needed. The laminate is then heated and cured while applying pressure to the laminate by press molding, autoclave molding, sheet winding molding, or the like, to obtain an electrical and electronic laminate (printed wiring board) or a carbon fiber reinforced material.

[0125] The curable resin composition of the present embodiment can also be made into a resin sheet. A method for obtaining a resin sheet from the curable resin composition of the present embodiment includes, for example, applying the curable resin composition to a support film (support), drying the composition, and forming a resin composition layer on the support film. When the curable resin composition of the present embodiment is used for a resin sheet, it is essential that the film softens under the lamination temperature conditions (70°C to 140°C) in the vacuum lamination method and exhibits fluidity (resin flow) that allows resin filling of via holes or through holes present in the circuit board simultaneously with lamination of the circuit board. It is preferable to blend the above-mentioned components so as to exhibit such properties. The resulting resin sheet or circuit board (such as a copper-clad laminate) requires a uniform appearance in order to exhibit consistent performance at any location without causing a phenomenon in which locally different characteristic values are exhibited due to phase separation or the like.

[0126] Here, the diameter of the through-holes in the circuit board is 0.1 to 0.5 mm, and the depth is 0.1 to 1.2 mm, and it is preferable to make it possible to fill the resin within this range. When laminating both sides of the circuit board, it is desirable to fill about half of the through-holes.

[0127] A specific method for producing the resin sheet includes preparing a resin composition varnished by blending an organic solvent therewith, applying the varnished resin composition to the surface of a support film (Y), and then drying the organic solvent by heating or blowing hot air or the like to form a resin composition layer (X).

[0128] The organic solvent used here preferably includes, for example: ketones such as acetone, methyl ethyl ketone, cyclohexanone, etc.; acetate esters such as ethyl acetate, butyl acetate, cellosolve acetate, propylene glycol monomethyl ether acetate, carbitol acetate, etc.; carbitols such as cellosolve, butyl carbitol, etc.; aromatic hydrocarbons such as toluene, xylene, etc.; dimethylformamide, dimethylacetamide, N-methylpyrrolidone, etc. It is also preferable to use the organic solvent in such a proportion that the nonvolatile content of the organic solvent is 30 to 60% by mass of the total.

[0129] The thickness of the resin composition layer (X) formed must be equal to or greater than the thickness of the conductor layer of the circuit board to which the resin composition layer (X) is laminated. Since the thickness of the conductor layer of the circuit board is in the range of 5 to 70 μm , the thickness of the resin composition layer (X) is preferably 10 to 100 μm . The resin composition layer (X) in the present embodiment may be protected with a protective film described below. Protection with a protective film can prevent the adhesion of dust and scratches to the surface of the resin composition layer (X).

[0130] Examples of the support film and protective film include: polyolefins such as polyethylene, polypropylene, and polyvinyl chloride; polyesters such as polyethylene terephthalate (PET) and polyethylene naphthalate; polycarbonate; polyimide; and release paper and metal foils such as copper foil and aluminum foil. The support film and protective film may be subjected to a matte treatment, corona treatment, or release treatment. The thickness of the support film is not particularly limited, but is generally in the range of 10 to 150 μm , preferably 25 to 50 μm . The thickness of the protective film is preferably 1 to 40 μm .

[0131] The support film (Y) is peeled off after laminating the resin composition layer (X) onto a circuit board, or after forming an insulating layer by heat-curing the resin composition layer (X). Peeling off the support film (Y) after the resin composition layer (X) constituting the resin sheet has been heat-cured can prevent the adhesion of dust and the like during the curing process. When peeling off the support film (Y) after the resin composition layer (X)

has been cured, the support film (Y) is previously subjected to a release treatment.

[0132] A multilayer printed circuit board can be produced from the resin sheet obtained as

described above. For example, when the resin composition layer (X) is protected by a

protective film, the protective film is peeled off from the resin composition layer (X), and

5 then the resin composition layer (X) is laminated to one or both sides of the circuit board so as to be in direct contact with the circuit board, for example, by a vacuum lamination method.

The lamination method may be a batch method or a continuous method using a roll. If

necessary, the resin sheet and circuit board may be heated (preheated) before lamination.

Regarding lamination conditions, a pressure bonding temperature (lamination temperature) of

10 70 to 140°C is preferred, a pressure bonding pressure of 1 to 11 kgf/cm² (9.8×10^4 to 107.9×10^4 N/m²) is preferred, and lamination is preferably performed under reduced pressure of 20 mmHg (26.7 hPa) or less.

[0133] The curable resin composition of the present embodiment can be used to manufacture semiconductor devices, such as dual in-line packages (DIPs), quad flat packages (QFPs), ball

15 grid arrays (BGAs), chip size packages (CSPs), small outline packages (SOPs), thin small outline packages (TSOPs), and thin quad flat packages (TQFPs).

[0134] The curable resin composition and its cured product according to the present

embodiment can be used in a wide range of fields. Specifically, they can be used in various

applications such as molding materials, adhesives, composite materials, and paints. The cured

20 product of the curable resin composition according to the present embodiment exhibits

excellent heat resistance and dielectric properties, and is therefore suitable for use in electrical

and electronic components such as encapsulants for semiconductor elements, encapsulants for

liquid crystal display elements, encapsulants for organic EL elements, laminates (printed

wiring boards, BGA (ball grid array) substrates, build-up substrates, etc.), lightweight, high-

25 strength structural composite materials such as carbon fiber reinforced plastics and glass fiber reinforced plastics, 3D printing, etc.

Example

[0135] The present invention will now be described in more detail with reference to examples. Unless otherwise specified, all parts are by weight. However, the present invention is not limited to these examples.

5 [0136] The various analytical methods used in the examples are described below.

<GPC (gel permeation chromatography) analysis>

Apparatus: online degassing unit (DGU-20A), liquid delivery unit (LC-20AD), autosampler (SIL-20A), photodiode array detector (SPD-M40), column oven (CTO-20A), system controller (CBM-20A), all manufactured by Shimadzu Corporation

10 Columns: SHODEX GPC KF-601 (2 columns), KF-602, KF-602.5, KF-603

Flow rate: 1.5 ml/min.

Column temperature: 40°C.

Solvent used: THF (tetrahydrofuran).

Detector: differential refractive index detector (RID-20A, manufactured by Shimadzu

15 Corporation).

<High-performance liquid chromatography (HP-LC)>

HP-LC: solvent delivery unit (LC-20AB), online degasser (DGU-20A3), autosampler (SIL-20A), column oven (CTO-20A), system controller (CBM-20A), absorbance detector (SPD-M20A) (all manufactured by Shimadzu Corporation).

20 Column: ODS-2 (manufactured by GL Sciences)

Eluent: tetrahydrofuran:water = 3:1 (no gradient)

Flow rate: 0.5 ml/min.

Column temperature: 40°C.

Detection: PDA (photodiode array detector).

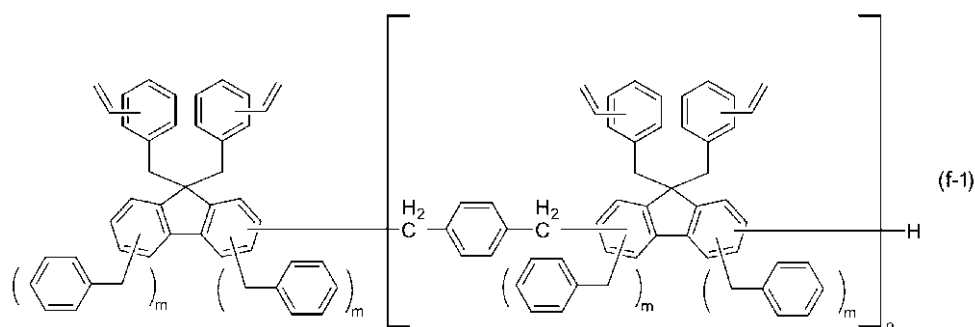
25 [0137] [Example 1]

While nitrogen purging into a flask equipped with thermometer, condenser, stirrer,

and Dean-Stark tube, 16.6 parts of fluorene, 3.5 parts of 1,4-benzenedimethanol, 21.6 parts of benzyl alcohol, 2.1 parts of methanesulfonic acid were added, and the temperature was raised to 130 °C. The reaction was continued for 5 hours while removing the generated water from the system using a Dean-Stark tube. After cooling to 70 °C, 1.0 parts of sodium hydroxide was added and stirred for 5 minutes, and then 100 parts of dimethyl sulfoxide and 12 parts of sodium hydroxide were added and stirred at 60 °C for 30 minutes. Thereafter, while maintaining the internal temperature at 65 °C or less, 29.0 parts of CMS-14 (AGC Seimi Chemical Co., Ltd., a mixture of 4-chloromethylstyrene and 3-chloromethylstyrene, 4-chloromethylstyrene: 3-chloromethylstyrene = 95:5 (mol ratio), purity 96.87 wt%) was added dropwise over 2 hours, and the reaction was continued at 65 °C for 3 hours. 200 parts of toluene was added, and the organic layer was washed five times with 100 parts of water. The obtained organic layer was concentrated to obtain 42.6 parts of compound (F1) represented by the following formula (f-1). The HP-LC chart of the obtained compound (F1) is shown in FIG. 1, and the GPC chart is shown in FIG. 2. The ¹H-NMR chart (deuterated chloroform) of the obtained compound is shown in FIG. 3. The number average molecular weight (M_n) of F1 by GPC analysis was 806, and the weight average molecular weight (M_w) was 2,276. In the ¹H-NMR chart, a signal derived from the terminal hydrogen of the vinyl group (two hydrogens per vinyl group) was observed at 4.95-5.65 ppm, a signal derived from a structure modified with 1,4-benzenedimethanol was observed at 3.05-3.45 ppm, and a signal derived from a structure modified with benzyl alcohol was observed at 3.75-4.25 ppm. When the integral value of the peak derived from the terminal hydrogen of the vinyl group was taken as 4.00, the integral value of the peak derived from the methylene hydrogen of the benzyl group modified with benzyl alcohol was 4.05. The average value (m_{ave}) of m in the following formula (f-1) calculated from this was 2.02. Furthermore, the integral value of the peak derived from the methylene hydrogen derived from the structure modified with 1,4-benzenedimethanol at this time was 3.40. The average value (n_{ave}) of n in the following

formula (f-1) calculated from this was 0.85.

[0138] [Chemical Formula 22]



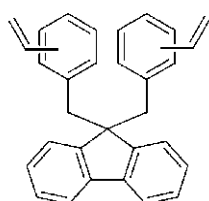
[0139] [Comparative Synthesis Example 1]

5 While nitrogen purging into a flask equipped with a thermometer, a condenser, and a stirrer, 133 parts of methyl isobutyl ketone (hereinafter also referred to as MIBK), 33.3 parts of fluorene, 1.9 parts of tetrabutylammonium bromide, 0.49 parts of hydroquinone, and 64 parts of 50 wt% aqueous sodium hydroxide solution were added, and the internal temperature was raised to 60 °C. Then, 71.2 parts of CMS-P (AGC Seimi Chemical Co., Ltd., a mixture of

10 4-chloromethylstyrene and 3-chloromethylstyrene, 4-chloromethylstyrene:3-chloromethylstyrene = 1:1 (molar ratio), purity 95.59 wt%) was added dropwise over 1 hour, and the reaction was carried out at 60 °C for 9 hours. The mixture was neutralized with 41.6 parts of a 35 wt% aqueous hydrochloric acid solution, and the organic layer was washed three times with 100 parts of water. Recrystallization was carried out with toluene and methanol to

15 obtain 35.6 parts of compound (F2) represented by the following formula (f-2). The HP-LC chart of the obtained compound (F2) is shown in FIG. 4, and the ¹H-NMR data (deuterated chloroform) is shown in FIG. 5.

[0140] [Chemical Formula 23]



(f-2)

[0141] [Example 2, Comparative Examples 1 and 2]

The compounds (F1, F2) obtained in Example 1 and Comparative Synthesis Example 1, and OPE-2St (a polyphenylene ether compound manufactured by Mitsubishi Gas Chemical Co., Ltd.) were used in the amounts shown in Table 1, and vacuum press-molded while sandwiched between mirror-finished copper foils (T4X: manufactured by Fukuda Metal Copper Foil Co., Ltd.), and cured at 220°C for 2 hours. A 250 μ m-thick piece of cushion paper with a 150 mm x 150 mm hole cut out from the center was used as a spacer. For evaluation, test pieces were cut to the desired size using a laser cutter as needed, and evaluation was performed.

[0142] <Dielectric constant test and dielectric loss tangent test>

Tests were performed at 25°C using a 10 GHz cavity resonator manufactured by AET Corporation using a cavity resonator perturbation method. The test was performed on a sample with a width of 1.7 mm, a length of 100 mm, and a thickness of 0.1 mm. The evaluation results are shown in Table 1.

[0143] [Table 1]

		Example 2	Comparative Example 1	Comparative Example 2
Compound	F1	100		
	F2		100	
	OPE-2St			100
Evaluation Results				
Dielectric Property	Dielectric Constant	2.51	2.51	2.51
	Dielectric Tangent	0.00106	0.00410	0.00395

[0144] From the results in Table 1, it was confirmed that the compounds of the present invention have a smaller dielectric loss tangent than those of Comparative Examples 1 and 2, and are excellent in low dielectric properties.

INDUSTRIAL APPLICABILITY

[0145] The compound of the present invention is suitable for use in electrical and electronic components such as semiconductor encapsulants, printed wiring boards, build-up laminates, etc.

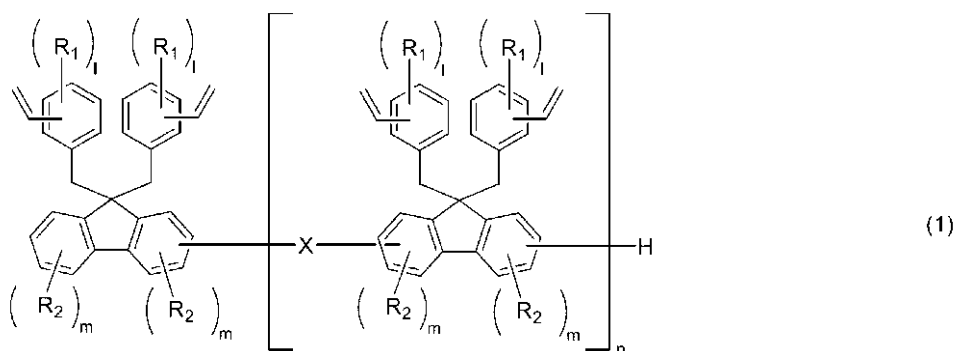
This application claims priority to Japanese Patent Application No. 2023-156530,

5 filed on September 21, 2023.

CLAIMS

[Claim 1] A compound represented by the following formula (1):

[Chemical Formula 1]



5 in the above formula (1), each of the multiple R_1 's independently represents a hydrogen atom or a hydrocarbon group having 1 to 5 carbon atoms;

each of the multiple R_2 's independently represents a hydrocarbon group represented by the following formula (a);

X has a structure represented by the following formulas (c) to (f);

10 each of the multiple l 's independently represents an integer of 1 to 4;

each of the multiple m 's independently represents an integer of 0 to 3;

the average value m_{ave} of m is $0 < m_{ave} \leq 3$;

n is the number of repetitions, and

the average value n_{ave} of n is $0.05 \leq n_{ave} \leq 20$,

15 [Chemical Formula 2]



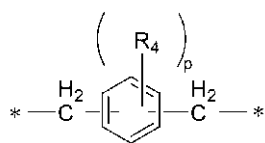
in the above formula (a),

* represents a bonding position to the fluorene structure in formula (1); R_3

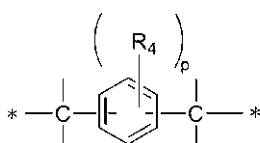
represents a hydrogen atom or a hydrocarbon group having 1 to 5 carbon atoms; and

20 k represents an integer of 1 to 5,

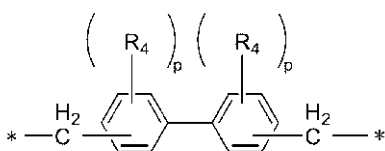
[Chemical Formula 3]



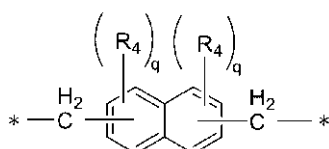
(c)



(d)



(e)



(f)

in the above formulas (c) to (f),

* represents a bonding position to the fluorene structure in formula (1);

multiple R₄'s each independently represent a hydrogen atom or a hydrocarbon group

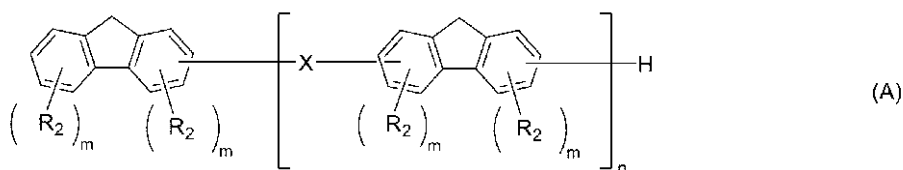
5 having 1 to 5 carbon atoms;

multiple p's each independently represent an integer of 1 to 4, and

multiple q's each independently represent an integer of 1 to 3.

[Claim 2] A compound obtained by reacting a compound represented by the following formula (A) with a compound represented by the following formula (B):

10 [Chemical Formula 4]



(A)

in the above formula (A), multiple R₂'s each independently represent a hydrocarbon group represented by the following formula (a);

X has a structure represented by the following formulas (c) to (f);

15 multiple m's each independently represent an integer of 0 to 3,

the average value m_{ave} of m is $0 < m_{ave} \leq 3$;

n is the number of repetitions; and

the average value n_{ave} of n is $0.05 \leq n_{ave} \leq 20$,

[Chemical Formula 5]



in the above formula (a),

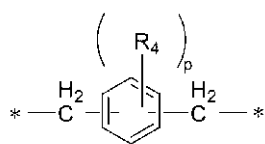
5 * represents the bonding position to the fluorene structure in formula (A);

R_3 represents a hydrogen atom or a hydrocarbon group having 1 to 5 carbon atoms;

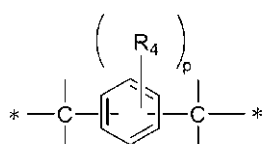
and

k represents an integer of 1 to 5,

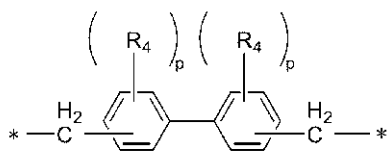
[Chemical Formula 6]



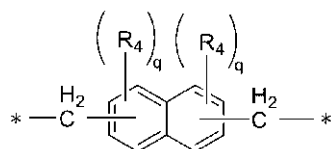
(c)



(d)



(e)



(f)

10

in the above formulas (c) to (f),

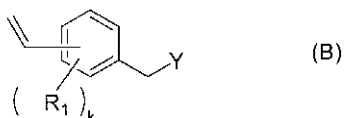
* represents the bonding position to the fluorene structure in formula (1);

each of the multiple R_4 's independently represents a hydrogen atom or a hydrocarbon group having 1 to 5 carbon atoms;

15 each of the multiple p 's independently represents an integer of 1 to 4; and

each of the multiple q 's independently represents an integer of 1 to 3,

[Chemical Formula 7]



in the above formula (B),

R_1 represents a hydrogen atom or a hydrocarbon group having 1 to 5 carbon atoms;

k represents an integer of 1 to 4; and

5 Y represents a halogen atom.

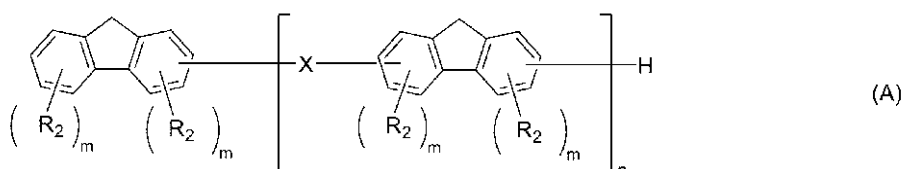
[Claim 3] A curable resin composition containing the compound according to claim 1 or 2.

[Claim 4] A cured product obtained by curing the compound according to claim 1 or 2.

[Claim 5] A cured product obtained by curing the curable resin composition according
10 to claim 3.

[Claim 6] A method for producing a compound obtained by reacting a compound represented by the following formula (A) with a compound represented by the following formula (B) in an aprotic polar solvent in the presence of a basic catalyst:

[Chemical Formula 8]



15 in the above formula (A), multiple R_2 's each independently represent a hydrocarbon group represented by the following formula (a);

X has a structure represented by the following formulas (c) to (f);

multiple m 's each independently represent an integer of 0 to 3;

20 the average value m_{ave} of m is $0 < m_{ave} \leq 3$;

n is the number of repetitions; and

the average value n_{ave} of n is $0.05 \leq n_{ave} \leq 20$,

[Chemical Formula 9]



in the above formula (a),

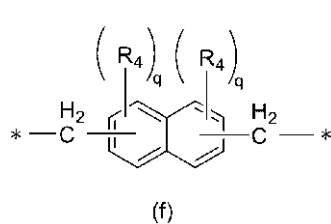
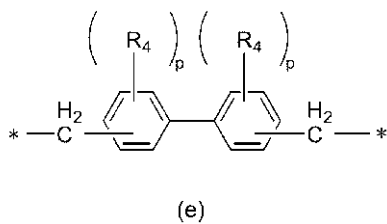
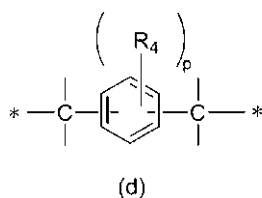
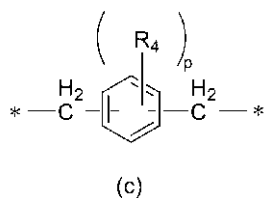
* represents the bonding position to the fluorene structure in formula (A);

R₃ represents a hydrogen atom or a hydrocarbon group having 1 to 5 carbon atoms;

5 and

k represents an integer of 1 to 5,

[Chemical Formula 10]



in the above formulas (c) to (f),

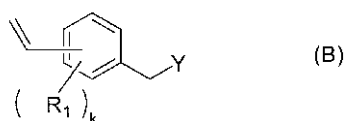
10 * represents the bonding position to the fluorene structure in formula (A);

each of the multiple R₄'s independently represents a hydrogen atom or a hydrocarbon group having 1 to 5 carbon atoms;

each of the multiple p's independently represents an integer of 1 to 4; and

each of the multiple q's independently represents an integer of 1 to 3,

15 [Chemical Formula 11]



in the above formula (B),

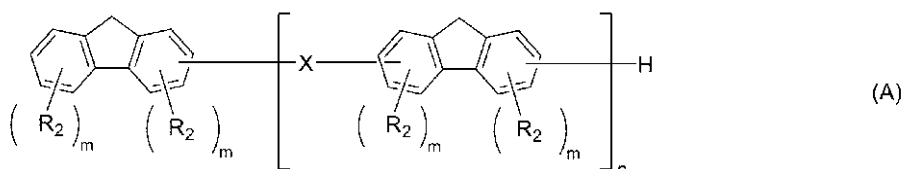
R_1 represents a hydrogen atom or a hydrocarbon group having 1 to 5 carbon atoms;

k represents an integer of 1 to 4; and

Y represents a halogen atom.

[Claim 7] A compound represented by the following formula (A):

5 [Chemical Formula 12]



in the above formula (A),

multiple R_2 's each independently represent a hydrocarbon group represented by the following formula (a);

10 X has a structure represented by the following formulas (c) to (f);

multiple m 's each independently represent an integer of 0 to 3;

the average value m_{ave} of m is $0 < m_{ave} \leq 3$;

n is the number of repetitions; and

the average value n_{ave} of n is $0.05 \leq n_{ave} \leq 20$,

15 [Chemical Formula 13]



in the above formula (a),

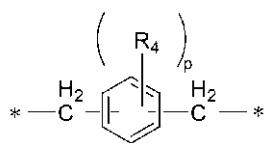
* represents the bonding position to the fluorene structure in formula (A);

R_3 represents a hydrogen atom or a hydrocarbon group having 1 to 5 carbon atoms;

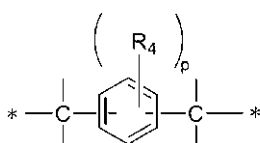
20 and

k represents an integer of 1 to 5,

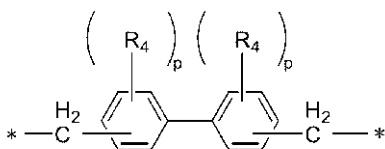
[Chemical Formula 14]



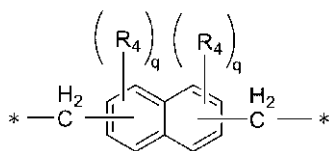
(c)



(d)



(e)



(f)

in the above formulas (c) to (f),

* represents the bonding position to the fluorene structure in formula (A);

multiple R₄'s each independently represent a hydrogen atom or a hydrocarbon group

5 having 1 to 5 carbon atoms;

multiple p's each independently represents an integer of 1 to 4; and

multiple q's each independently represents an integer of 1 to 3.