

300°C or higher. In addition, silicone-based adhesives have a problem in that they contain a low-molecular-weight silicone component which adversely affects electronic parts.

[0004]

5 Examples of resins having a heat resistance of 250°C or higher include polyimide resins. Although polyimide resins have a problem in that the handling thereof is difficult due to their high processing temperatures, it has been proposed to use a polyimide resin in the form of a siloxane-based polyimide resin copolymerized with siloxane diamine for the application as a bonding agent or an adhesive (see, for
10 example, Patent Documents 1 and 2). However, since the copolymerized siloxane diamine has a low molecular weight or is contained in a small amount, although these siloxane-based polyimide resins can be used as bonding agents, they are insufficient for the use as an adhesive that is required to have a press-bonding property at room temperature. Accordingly, polysiloxane-based polyimide resins in
15 which a relatively high-molecular-weight polysiloxane diamine is copolymerized in a large amount have been proposed (see, for example, Patent Documents 3 and 4).

PRIOR ART REFERENCES

PATENT DOCUMENTS

[0005]

20 [Patent Document 1] Japanese Patent Application Laid-Open Publication (JP-A) No. H5-200946

 [Patent Document 2] JP-A No. 2004-277619

 [Patent Document 3] WO 2013/039029

 [Patent Document 4] JP-A No. H9-183850

25 SUMMARY

PROBLEMS TO BE SOLVED

[0006]

However, even these polysiloxane-based polyimide resins are still insufficient in terms of adhesive strength and have a problem in performing uniform adhesion over a large area. In view of this, an object of the embodiments disclosed herein is to provide: a polyimide resin which generates no volatile component due to decomposition or the like even at a high temperature of 250°C or higher and exhibits sufficiently high adhesive strength in a uniform manner over a large area when used for adhesion at room temperature; a resin composition comprising the polyimide resin; and a laminated film.

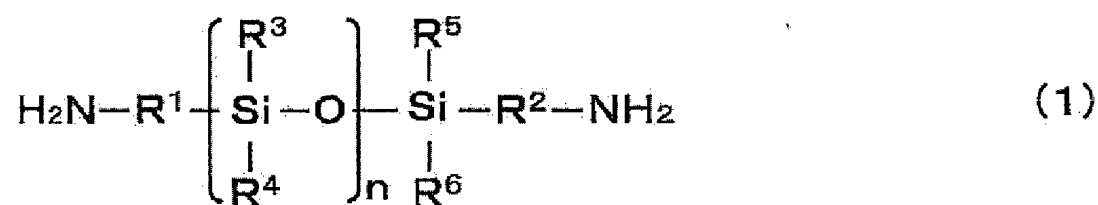
MEANS FOR SOLVING THE PROBLEMS

[0007]

That is, the present disclosure is a polyimide resin comprising at least an acid anhydride residue and a diamine residue, the polyimide resin being characterized by comprising a residue of a polysiloxane diamine represented by the following Formula (1) in an amount of not less than 60% by mole in the total amount of the diamine residue:

[0008]

[C1]



[0009]

(wherein, n is a natural number and the average value thereof calculated from the average molecular weight of the polysiloxane diamine is 45 to 200; R¹ and R², the same or different, each represent an alkylene group having 1 to 30 carbon atoms or a phenylene group; and R³ to R⁶, the same or different, each represent an alkyl group having 1 to 30 carbon atoms, a phenyl group or a phenoxy group).

It is noted here that the term "residue" means a chemical structure that exists in a chemical substance such as a polymer and is originated from a raw material, and each residue is indicated with the name or abbreviation of the raw material.

[0009a]

5 In an aspect, there is provided a polyimide resin comprising at least:

an acid anhydride residue; and

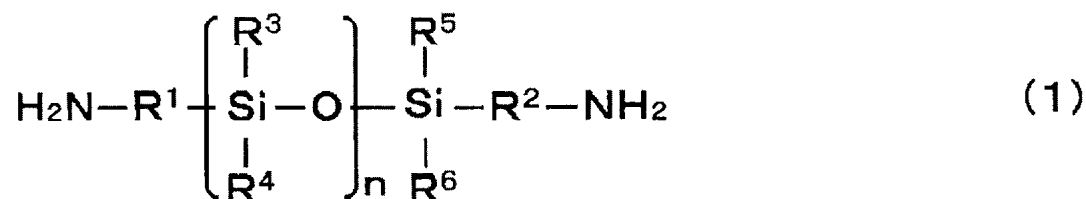
a diamine residue,

said polyimide resin comprising a residue of a polysiloxane diamine

represented by the following Formula (1) in an amount of not less than 60% by mole

10 in the total amount of said diamine residue:

[C1]



wherein, n is a natural number and the average value thereof calculated from the average molecular weight of said polysiloxane diamine is 45 to 200; R¹ and R², the same or different, each represent an ethylene group, trimethylene group, tetramethylene group or pentamethylene group; and R³ to R⁶, the same or different, each represent a methyl group, ethyl group, propyl group or butyl group.

[0009b]

In another aspect, there is provided a resin composition comprising the polyimide

20 resin as defined above or a polyamic acid resin which is a precursor of said polyimide resin.

[0009c]

In another aspect, there is provided a cured film obtained by curing the resin composition as defined above.

[0009d]

In another aspect, there is provided a laminated film in which the resin composition as defined above is laminated on at least one side of a heat-resistant insulation film.

[0009e]

- 5 In another aspect, there is provided a method of producing a semiconductor device using the resin composition as defined above.

[0009f]

In another aspect, there is provided a method of producing a semiconductor device using the laminated film as defined above.

10 EFFECTS

[0010]

- According to the present disclosure, adhesion can be performed uniformly over a large area even at room temperature with expression of good adhesiveness. Further, a polyimide resin having excellent heat resistance which generates no
15 volatile component due to decomposition or the like even at a high temperature of 250°C or higher; a resin composition comprising the polyimide resin; and a laminated film can be provided.

MODE FOR CARRYING OUT THE INVENTION

[0011]

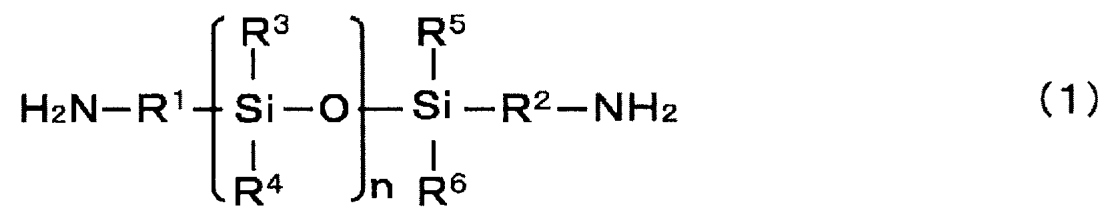
- 20 The polyimide resin of the present invention can be laminated on a heat-resistant insulation film, a glass substrate, a silicon wafer or the like to be used as an adhesive layer-laminated film or adhesive layer-laminated substrate.

[0012]

- The polyimide resin of the present invention comprises at least an acid
25 dianhydride residue and a diamine residue, and the diamine residue contains a residue of a polysiloxane diamine represented by the Formula (1):

[0013]

[C2]



[0014]

CLAIMS

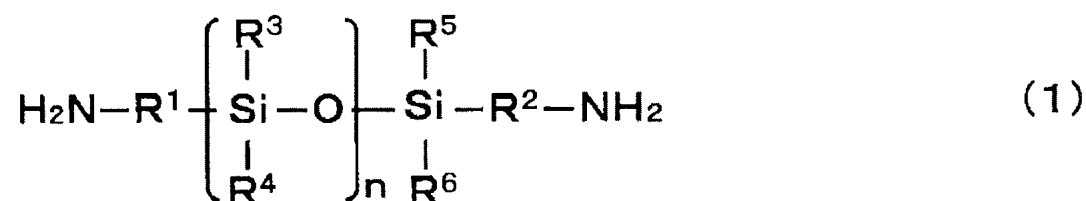
1. A polyimide resin comprising at least:

an acid anhydride residue; and

a diamine residue,

said polyimide resin comprising a residue of a polysiloxane diamine represented by the following Formula (1) in an amount of not less than 60% by mole in the total amount of said diamine residue:

[C1]



wherein, n is a natural number and the average value thereof calculated from the average molecular weight of said polysiloxane diamine is 45 to 200; R¹ and R², the same or different, each represent an ethylene group, trimethylene group, tetramethylene group or pentamethylene group; and R³ to R⁶, the same or different, each represent a methyl group, ethyl group, propyl group or butyl group.

2. The polyimide resin according to claim 1, comprising a residue of a hydroxyl group-containing diamine in said diamine residue.

3. The polyimide resin according to claim 2, comprising said residue of said hydroxyl group-containing diamine in an amount of 0.1 to 40% by mole in the total amount of said diamine residue.

4. The polyimide resin according to claim 1, having a glass transition temperature of 30°C or lower.

5. A resin composition comprising the polyimide resin according to any one of claims 1

to 4 or a polyamic acid resin which is a precursor of said polyimide resin.

6. The resin composition according to claim 5, comprising a methylol compound.

7. The resin composition according to claim 6, wherein said methylol compound is a compound having two or more groups represented by the following Formula (2):

[C2]



wherein, when plural R⁷s exist in said compound, said R⁷s, the same or different, each represent hydrogen or an alkyl group having 1 to 10 carbon atoms.

8. The resin composition according to claim 6 or 7, wherein the content of said methylol compound is 0.1 to 20% by mass with respect to said polyimide resin or said polyamic acid resin which is a precursor of said polyimide resin.

9. A cured film obtained by curing the resin composition according to any one of claims 5 to 8.

10. A laminated film in which the resin composition according to any one of claims 5 to 8 is laminated on at least one side of a heat-resistant insulation film.

11. The laminated film according to claim 10, wherein a release treatment is performed on the surface of said heat-resistant insulation film on which said resin composition is laminated.

12. The laminated film according to claim 10 or 11, wherein a release-treated film is further laminated on the surface of said resin composition laminated on said heat-resistant insulation film.

13. A method of producing a semiconductor device using the resin composition according to any one of claims 5 to 8.

14. The method of producing a semiconductor device according to claim 13, wherein a substrate for semiconductor circuit formation and a support substrate are joined