

[Name of Document] Description
[Title of Invention] RELEASE FILM FOR CERAMIC GREEN
SHEET PRODUCTION PROCESS

[Technical Field]

5 [0001]

The present invention relates to a release film that is used in a process for producing a ceramic green sheet.

[Background Art]

[0002]

10 To produce laminated ceramic products such as laminated ceramic capacitors and multilayer ceramic substrates, it has conventionally been performed to form ceramic green sheets, laminate the obtained ceramic green sheets, and fire them.

15 [0003]

Each ceramic green sheet may be formed by coating a release film with a ceramic slurry that contains ceramic materials such as barium titanate and titanium oxide. The release film is required to have releasability such that
20 the thin ceramic green sheet formed on the release film can be released from the release film by moderate release force without deformation, breakage, and other troubles in the ceramic green sheet. The release film is also required to have smoothness of the surface to be coated with the
25 ceramic slurry (the surface to be in contact with the ceramic slurry/ceramic green sheet, which may be referred to as a "release surface," hereinafter) so that defects such as pinholes and thickness irregularity do not occur when the release film is coated with the ceramic slurry and
30 the ceramic slurry is dried.

[0004]

Patent Documents 1 and 2 disclose examples of such release films. In the release film disclosed in Patent

Document 1, a layer formed of an ultraviolet curable resin is provided on a base material and a release agent layer is provided on the layer. The release agent layer is a silicone resin layer formed by an addition reaction of organopolysiloxane. The release film disclosed in Patent Document 2 is produced through coating a base material with a coating liquid that contains a (meth)acrylate component and a modified silicone oil modified with a (meth)acryloyl group and/or a vinyl group and curing the coating film. Through this process, a layer that contains a cured product of the (meth)acrylate component is formed on the base material and a release agent layer is formed on the layer. The release agent layer is a layer that contains a silicone polymer component.

[Prior Art Document]

[Patent Document]

[0005]

[Patent Document 1] JP5675246B

[Patent Document 2] JP5423975B

[Summary of the Invention]

[Problems to be solved by the Invention]

[0006]

The silicone component in the release agent composition may readily migrate to the surface of the ceramic green sheet in contact with the release agent layer. The surface to which silicone migrates may have slippery properties to deteriorate the adhesiveness. If ceramic green sheets are produced with such release sheets to which the silicone component readily migrates and a laminated ceramic product is produced using the ceramic green sheets, interlaminar misalignment may occur in the plane direction in the laminated ceramic product when pressure is applied between the laminated ceramic green sheets. The occurrence

of such misalignment may deteriorate the positional accuracy of components such as electrodes in the obtained laminated ceramic product, and the product performance of the laminated ceramic product cannot be obtained. Thus,
5 there is a demand for release films from which the silicone component is less likely to migrate to the ceramic green sheets.

[0007]

When the release films as described in Patent
10 Documents 1 and 2 are used, however, such migration of the silicone component cannot be sufficiently suppressed.

[0008]

The present invention has been made in consideration of such actual circumstances and an object of the present
15 invention is to provide a release film for a ceramic green sheet production process with which the migration of polyorganosiloxane from the release agent layer to the ceramic green sheet is suppressed.

[Means for solving the Problems]

20 [0009]

To achieve the above object, first, the present invention provides a release film for a ceramic green sheet production process, the release film comprising a base material and a release agent layer provided on one side of
25 the base material, the release agent layer being formed by curing a release agent composition that contains a melamine resin and a polyorganosiloxane, the release film having a coating elastic modulus of 3.5 to 7.0 GPa as measured by a nanoindentation test from a surface of the release agent
30 layer opposite to the base material (Invention 1).

[0010]

In the above invention (Invention 1), the release agent layer is formed of a release agent composition that

contains a polyorganosiloxane, thereby to moderately reduce the surface free energy, and the releasability is excellent when the release film is released from the ceramic green sheet. Moreover, the melamine resin is sufficiently cured
5 in the release agent layer so that the coating elastic modulus falls within the above-described range, and the polyorganosiloxane confined in the mesh-like structure is thereby tightly restrained. Thus, the migration of the polyorganosiloxane from the release agent layer to the
10 ceramic green sheet is suppressed.

[0011]

In general, the "melamine resin" refers to a mixture that contains two or more types of melamine compounds and/or a polynuclear substance obtained by condensing such
15 melamine compounds. In the present description, the term "melamine resin" shall refer to the above mixture or an aggregate of one type of a melamine compound. In the present description, a product obtained by curing the melamine resin shall be referred to as a "melamine cured
20 product."

[0012]

In the above invention (Invention 1), the polyorganosiloxane may preferably have a mass-average molecular weight of 500 to 20,000 (Invention 2).

25 [0013]

In the above invention (Invention 1, 2), the content of the polyorganosiloxane in the release agent composition may preferably be 0.1 to 30 mass parts with respect to 100 mass parts of the melamine resin (Invention 3).

30 [0014]

In the above invention (Invention 1 to 3), the melamine resin contained in the release agent composition may preferably contain methylated melamine and/or

iminomethylol melamine (Invention 4).

[0015]

In the above invention (Invention 1 to 4), the release film may preferably further comprise a resin layer between
5 the base material and the release agent layer (Invention 5).

[0016]

In the above invention (Invention 5), the resin layer may preferably be formed by curing a resin composition that contains an active energy ray-curable component or a
10 thermoset component (Invention 6).

[0017]

In the above invention (Invention 6), the active energy ray-curable component may preferably be a polyfunctional acrylate (Invention 7).

15 [0018]

In the above invention (Invention 1 to 7), the release film may further comprise a second resin layer provided on a side of the base material opposite to the release agent layer (Invention 8).

20 [Brief Description of Drawings]

[0019]

FIG. 1 is a cross-sectional view of a release film for a ceramic green sheet production process according to a first embodiment of the present invention.

25 FIG. 2 is a cross-sectional view of a release film for a ceramic green sheet production process according to a second embodiment of the present invention.

FIG. 3 is a cross-sectional view of a release film for a ceramic green sheet production process according to a
30 third embodiment of the present invention.

[Embodiments for Carrying out the Invention]

[0020]

Hereinafter, embodiments of the present invention will

be described.

<Release Film for Ceramic Green Sheet Production Process>

As illustrated in FIG. 1, a release film for a ceramic green sheet production process 1A according to a first
5 embodiment (which may be simply referred to as a "release film 1A," hereinafter) is configured to include a base material 11 and a release agent layer 12.

[0021]

As illustrated in FIG. 2, a release film for a ceramic
10 green sheet production process 1B according to a second embodiment (which may be simply referred to as a "release film 1B," hereinafter) is configured to include a base material 11, a resin layer 13 laminated on one surface (upper surface in FIG. 2) of the base material 11, and a
15 release agent layer 12 laminated on a surface of the resin layer 13 opposite to the base material 11.

[0022]

As illustrated in FIG. 3, a release film for a ceramic green sheet production process 1C according to a third
20 embodiment (which may be simply referred to as a "release film 1C," hereinafter) is configured to include a base material 11, a resin layer 13 laminated on one surface (upper surface in FIG. 3) of the base material 11, a release agent layer 12 laminated on a surface of the resin
25 layer 13 opposite to the base material 11, and a second resin layer 14 laminated on the other surface (lower surface in FIG. 3) of the base material 11.

[0023]

In the release film 1A, 1B, 1C, the release agent
30 layer 12 is formed of a release agent composition that contains a melamine resin and a polyorganosiloxane. The coating elastic modulus as measured by a nanoindentation test from a surface of the release agent layer 12 opposite

to the resin layer 13 (or the base material 11) is 3.5 to 7.0 GPa.

[0024]

In the release film 1A, 1B, 1C, the release agent
5 layer 12 is formed using the release agent composition
which contains polyorganosiloxane, thereby to moderately
reduce the surface free energy of the release surface of
the release agent layer 12. Moreover, the release film 1A,
1B, 1C has the above-described coating elastic modulus.
10 Owing to these features, when the release film 1A, 1B, 1C
is released from the ceramic green sheet formed on the
release surface of the release film 1A, 1B, 1C, the release
force is moderately reduced and excellent releasability is
exhibited.

15 [0025]

Furthermore, in the release film 1A, 1B, 1C, the
polyorganosiloxane is confined in a mesh-like structure
that is formed by curing the melamine resin. Here, the
melamine resin is sufficiently cured so that the coating
20 elastic modulus falls within the above-described range, and
the polyorganosiloxane confined in the above mesh-like
structure is thereby tightly restrained. This restricts
the polyorganosiloxane from freely moving in the cured
melamine resin. When a ceramic green sheet is formed on
25 the release surface of the release film 1A, 1B, 1C,
therefore, migration of the polyorganosiloxane from the
release agent layer 12 to the ceramic green sheet is
suppressed. This can enhance the interfacial adhesion
between ceramic green sheets each formed using the release
30 film 1A, 1B, 1C when they are laminated on one another.
Thus, in the production of laminated ceramic products, the
occurrence of misalignment is suppressed between the
ceramic green sheets or between the electrode printing

surface and the surface of a ceramic green sheet in contact with the electrode printing surface.

[0026]

In the release film 1A, 1B, 1C, the release agent
5 layer 12 is formed of a release agent composition that contains a melamine resin. The surface free energy of the release agent layer 12 is therefore avoided from being unduly low as compared with the case in which the release agent layer is formed using only the polyorganosiloxane.
10 Moreover, unlike an active energy ray-curable resin in which curing of the surface layer is insufficient due to oxygen inhibition, the melamine resin is cured by thermal curing; therefore, in the release agent layer 12, not only the entire coating elastic modulus is improved, but also
15 the elastic modulus is sufficiently improved in the layer and in the surface layer. This allows the polyorganosiloxane, which exists in the vicinity of the surface layer of the release agent layer 12, to be sufficiently fixed there. Furthermore, the melamine resin
20 has high affinity with the resin contained in the resin layer 13 which will be described later. In the release film 1B, 1C provided with the resin layer 13, therefore, the interfacial adhesion between the release agent layer 12 and the resin layer 13 is excellent, and the occurrence of
25 delamination of the release agent layer 12 from the resin layer 13 is suppressed.

[0027]

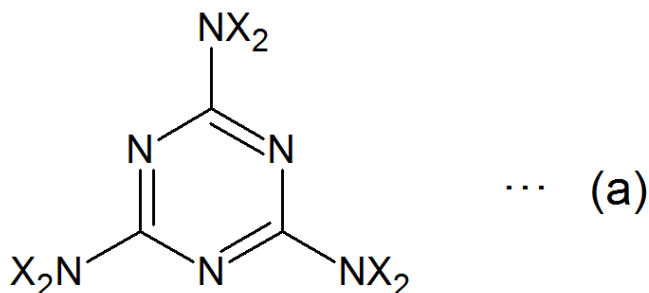
1. Release Agent Layer

(1) Melamine Resin

30 The melamine resin contained in the release agent composition may contain a melamine compound as represented by general formula (a) below or a polynuclear substance obtained by condensing two or more molecules of the

melamine compound.

[Chemical Formula 1]



5 [0028]

In the formula (a), each X represents -H, -CH₂-OH, or -CH₂-O-R. These groups may constitute reactive groups in the condensation reaction between molecules of the above melamine compound. Specifically, an -NH group in which X
 10 is H can undergo condensation reactions with an -N-CH₂-OH group and an -N-CH₂-R group. An -N-CH₂-OH group in which X is -CH₂-OH and an -N-CH₂-O-R group in which X is -CH₂-O-R can both undergo condensation reactions with an -NH group, an -N-CH₂-OH group, and an -N-CH₂-O-R group.

15 [0029]

In the above -CH₂-O-R group, R represents an alkyl group having a carbon number of 1 to 8. The carbon number may be preferably 1 to 6 and particularly preferably 1 to 3. Examples of the alkyl group having a carbon number of 1 to
 20 8 include a methyl group, ethyl group, propyl group, butyl group, pentyl group, hexyl group, and octyl group, among which the methyl group may be particularly preferred.

[0030]

In the above, each X may be the same or different and
 25 each R may be the same or different.

[0031]

In general, melamine compounds may be classified into those of a full ether type in which all of Xs are -CH₂-O-R,

those of an imino-methylol type in which at least one X is $-\text{CH}_2-\text{OH}$ and at least one X is H, those of a methylol type in which at least one X is $-\text{CH}_2-\text{OH}$ and no X exists as H, and those of an imino type in which at least one X is H and
5 no X exists as $-\text{CH}_2-\text{OH}$. Examples preferably used as the melamine compound contained in the above melamine resin include methylated melamine (full ether type in which R is a methyl group), iminomethylol melamine (imino-methylol type), methylol melamine (methylol type), and butylated
10 melamine (full ether type in which R is a butyl group. It may be preferred to use methylated melamine or iminomethylol melamine from the viewpoint of being readily dissolved in an organic solvent and readily cured at low temperatures. In particular, it may be preferred to use
15 iminomethylol melamine from the viewpoint that a reaction for removing protective groups is unnecessary and the reaction rate is further improved.

[0032]

The above melamine resin may contain a polynuclear
20 substance in which 2 to 50 molecules of the compound represented by the above formula (a) are condensed in an embodiment, contain a polynuclear substance in which 2 to 30 molecules of the compound are condensed in another embodiment, contain a polynuclear substance in which 2 to
25 10 molecules of the compound are condensed in a particular embodiment, and contain a polynuclear substance in which 2 to 5 molecules of the compound are condensed in a further embodiment.

[0033]

30 In the release agent composition for forming the release agent layer 12, the mass-average molecular weight of the melamine resin may be preferably 120 to 10,000, particularly preferably 200 to 5,000, and further

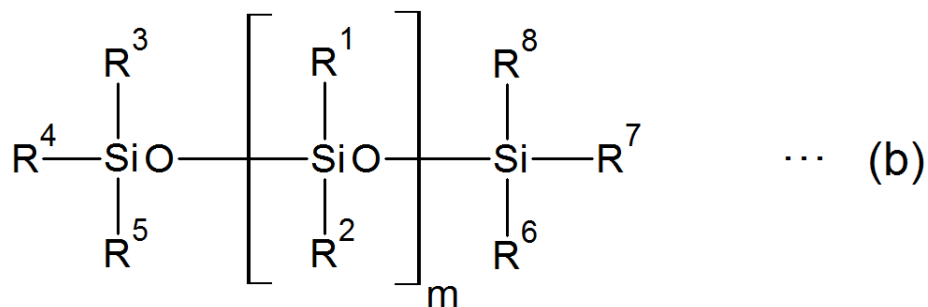
preferably 1,000 to 4,000. The mass-average molecular weight being 120 or more allows the melamine resin to be stably crosslinked, and a smoother release surface can be formed. The mass-average molecular weight being 10,000 or less can suppress the viscosity of the release agent composition from being excessively high, and good coating properties can be obtained when the base material 11 is coated with the release agent composition. As used in the present description, the mass-average molecular weight refers to a value that is measured as a standard polystyrene equivalent value using the gel permeation chromatography (GPC) method.

[0034]

(2) Polyorganosiloxane

A polymer of a silicon-containing compound represented by general formula (b) below can be used as the polyorganosiloxane contained in the release agent composition.

[Chemical Formula 2]



[0035]

In the formula (b), m is an integer not smaller than one. In the formula (b), each of R^1 to R^8 may be preferably an alkyl group or an aryl group and particularly preferably an alkyl group having a carbon number of 1 to 8 or an aryl group having a carbon number of 6 to 8.

Examples of the alkyl group having a carbon number of 1 to 8 include a methyl group, ethyl group, propyl group, butyl group, pentyl group, hexyl group, and octyl group. Examples of the aryl group having a carbon number of 6 to 8
5 include a phenyl group and methylphenyl group. Among these, the methyl group may be particularly preferred.

[0036]

In the above, R^1 to R^8 may be the same or different. When two or more R^1 and two or more R^2 are present, R^1 and
10 R^2 may be the same or different between repeating units.

[0037]

The polyorganosiloxane contained in the release agent composition may preferably have an organic group at an end or at a side chain. That is, at least one of R^1 to R^8 in
15 the formula (b) may preferably be an organic group. As used in the present description, the "organic group" refers to a group that does not contain the above-described alkyl group and aryl group. Examples of such an organic group include an organic group having a repeating structure, such
20 as polyether, polyester, or polyurethane. In such an organic group, the atom at one end of the organic group, such as polyether, polyester, or polyurethane, is bonded to the silicon atom at an end or side chain of the polyorganosiloxane.

25 [0038]

The polyorganosiloxane may preferably contain a functional group capable of a crosslinking reaction. The functional group capable of a crosslinking reaction may be directly bonded to the silicon atom of the
30 polyorganosiloxane or may also be bonded to the polyorganosiloxane via the previously-described organic group. That is, at least one of R^1 to R^8 in the formula (b) may preferably be a crosslinkable functional group or

an organic group that has a crosslinkable functional group. Examples of the crosslinkable functional group directly bonded to the silicon atom include an alkenyl group, hydrogen group (hydrosilyl group), and hydroxyl group
5 (hydroxysilyl group). Examples of the functional group bonded to the polyorganosiloxane via an organic group include a hydroxyl group, carboxyl group, glycidyl group, and (meth)acryloyl group. Among these functional groups, the hydroxyl group directly bonded to the silicon atom
10 (hydroxysilyl group) or the hydroxyl group bonded to the silicon atom via an organic group may be particularly preferred. The "(meth)acryloyl group" as used in the present description refers to both an acryloyl group and a methacryloyl group. The same applies to other similar
15 terms.

[0039]

When the release agent layer 12 is formed by curing the release agent composition which contains the polyorganosiloxane having a hydroxyl group, the melamine
20 resin and the polyorganosiloxane are chemically bonded via the hydroxyl group, and the polyorganosiloxane is fixed to the melamine cured product. This can effectively restrict the polyorganosiloxane from freely moving in the release agent layer 12, and the migration of the polyorganosiloxane
25 from the release agent layer 12 to the ceramic green sheet is effectively suppressed. As a result, when ceramic green sheets are laminated, the occurrence of misalignment is effectively suppressed between the ceramic green sheets or between the electrode printing surface and the surface of a
30 ceramic green sheet in contact with the electrode printing surface. This can improve the positional accuracy of electrodes and the like.

[0040]

The mass-average molecular weight of the polyorganosiloxane may be preferably 500 to 20,000, particularly preferably 1,000 to 10,000, and further preferably 3,000 to 8,000. When the mass-average molecular weight of the polyorganosiloxane is 500 or more, the surface free energy of the release surface of the release agent layer 12 moderately decreases to allow the release force to be effectively reduced when the release film 1A, 1B, 1C is released from the ceramic green sheet. The mass-average molecular weight of the polyorganosiloxane being 20,000 or less can suppress the viscosity of the release agent composition from being excessively high, and the coating with the release agent composition may be made easy.

[0041]

The content of the polyorganosiloxane in the release agent composition may be preferably 0.1 to 30 mass parts, particularly preferably 0.3 to 25 mass parts, and further preferably 0.5 to 20 mass parts, to 100 mass parts of the melamine resin. The content of the polyorganosiloxane in the release agent composition being 0.1 mass parts or more can sufficiently reduce the surface free energy of the release surface of the release agent layer 12 to achieve the moderate release force. The content of the polyorganosiloxane in the release agent composition being 30 mass parts or less can suppress the above-described migration of the polyorganosiloxane. This can prevent the surface free energy of the release agent composition from being excessively reduced, and the occurrence of crawling can be suppressed when the resin layer 13 is coated with a coating liquid of the release agent composition. The release film 1A, 1B, 1C can thus produced well.

[0042]

(3) Other Components

The release agent composition may preferably further contain an acid catalyst. Preferred examples of the acid catalyst include hydrochloric acid, sulfuric acid, nitric acid, phosphoric acid, phosphorus acid, and p-toluenesulfonic acid, among which the p-toluenesulfonic acid may be particularly preferred. The release agent composition containing an acid catalyst allows the condensation reaction to efficiently progress in the above melamine resin.

10 [0043]

The content of the acid catalyst in the release agent composition may be preferably 0.1 to 30 mass parts, particularly preferably 0.5 to 20 mass parts, and further preferably 1 to 15 mass parts, to 100 mass parts of the
15 melamine resin.

[0044]

The release agent composition may contain other additives, such as a crosslinker and reaction inhibitor, in addition to the above components.

20 [0045]

(4) Thickness of Release Agent Layer

The thickness of the release agent layer 12 may be preferably 5 to 2,000 nm. In particular, in the release film 1A comprising the base material 11 and the release
25 agent layer 12, the thickness of the release agent layer 12 may be preferably 100 to 2,000 nm, particularly preferably 150 to 1,000 nm, and further preferably 200 to 600 nm. In the release film 1B or 1C further comprising the resin layer 13 between the base material 11 and the release agent
30 layer 12, the thickness of the release agent layer 12 may be preferably 5 to 300 nm, particularly preferably 10 to 250 nm, and further preferably 15 to 200 nm. When the thickness of the release agent layer 12 is 5 nm or more,

the release agent layer 12 can effectively exert its functions. When the thickness of the release agent layer 12 is 2,000 nm or less, the occurrence of curling can be suppressed.

5 [0046]

2. Base Material

The base material 11 of the release film 1A, 1B, 1C is not particularly limited, provided that the resin layer 13 (or the release agent layer 12) can be laminated on the
10 base material 11. Examples of the base material 11 include films formed of plastics, such as polyethylene terephthalate, polyethylene naphthalate and other polyesters, polypropylene, polymethylpentene and other polyolefins, polycarbonate, and polyvinyl acetate. The
15 base material 11 may be a single layer or may also be a multilayer of two or more layers of the same type or different types. Among these, a polyester film may be preferred, a polyethylene terephthalate film may be particularly preferred, and a biaxially stretched
20 polyethylene terephthalate film may be further preferred. The polyethylene terephthalate film is less likely to generate dust and the like during the work, use, and other occasions and can therefore effectively prevent troubles, such as ceramic slurry coating failure, due to dust and the
25 like, for example. The polyethylene terephthalate film may be treated with an antistatic thereby to prevent firing due to static electrical charge at the time of coating with the ceramic slurry, which uses an organic solvent, and to enhance the effect of preventing the coating failure and
30 the like.

[0047]

For the purpose of improving the interfacial adhesion of the base material 11 with the release agent layer 12 or

resin layer 13 provided on the surface of the base material 11, one surface or both surfaces of the base material 11 may be subjected to surface treatment, such as using an oxidation method or roughening method, or primer treatment, as necessary. Examples of the above oxidation method include corona discharge treatment, plasma discharge treatment, chromium oxidation treatment (wet type), flame treatment, hot-air treatment, ozone exposure treatment, and ultraviolet irradiation treatment. Examples of the roughening method include a sandblast method and thermal spraying method. These surface treatment methods may be appropriately selected depending on the type of the base material film, and the corona discharge treatment method may preferably be used in view of the effect and the operability in general.

[0048]

The thickness of the base material 11 may be ordinarily 10 to 300 μm , preferably 15 to 200 μm , and particularly preferably 20 to 125 μm .

[0049]

3. Resin Layer

The release film according to one or more embodiments of the present invention may be provided with the resin layer 13 between the base material 11 and the release agent layer 12. FIGS. 2 and 3 illustrate the release films 1B and 1C each provided with the resin layer 13. In the release film 1B, 1C, the resin layer 13 provided between the base material 11 and the release agent layer 12 absorbs irregularities on the surface of the base material 11 on the side of the resin layer 13. This allows the surface of the resin layer 13 opposite to the base material 11 to have high smoothness. Thus, when the release agent layer 12 is provided on that surface of the resin layer 13, the release

surface of the release agent layer 12 can have excellent smoothness.

[0050]

The resin which forms the resin layer 13 is not particularly limited, provided that the resin can give smoothness to the release surface without impeding the effects of the present invention. It may be preferred to use a resin that allows the release film 1B, 1C to readily achieve a coating elastic modulus of 3.5 to 7.0 GPa, as the resin which forms the resin layer 13. In particular, the resin layer 13 may preferably be formed of a resin composition that contains a curable component. The curable component may be an active energy ray-curable component or may also be a thermoset component.

[0051]

(1) Active Energy Ray-Curable Component

The active energy ray-curable component is not particularly limited, provided that the component can be cured by irradiation with active energy rays without impeding the effects of the present invention, and may be any of a monomer, oligomer, and polymer or may also be a mixture thereof. In particular, it may be preferred to use a component that constitutes an acrylic resin as the active energy ray-curable component, and it is particularly preferred to use a polyfunctional acrylate.

[0052]

Examples of the polyfunctional acrylate include bifunctional ones such as 1,4-butanediol di(meth)acrylate, 1,6-hexanediol di(meth)acrylate, neopentyl glycol di(meth)acrylate, polyethylene glycol di(meth)acrylate, neopentyl glycol adipate di(meth)acrylate, hydroxypivalic acid neopentyl glycol di(meth)acrylate, dicyclopentanyldi(meth)acrylate, caprolactone-modified dicyclopentenyl

di(meth)acrylate, ethylene oxide-modified phosphoric acid
 di(meth)acrylate, di(acryloxyethyl) isocyanurate, and
 allylated cyclohexyl di(meth)acrylate; trifunctional ones
 such as trimethylolpropane tri(meth)acrylate,
 5 dipentaerythritol tri(meth)acrylate, propionic acid-
 modified dipentaerythritol tri(meth)acrylate,
 pentaerythritol tri(meth)acrylate, propylene oxide-modified
 trimethylolpropane tri(meth)acrylate, tris(acryloxyethyl)
 isocyanurate, and ϵ -caprolactone-modified tris-(2-
 10 (meth)acryloxyethyl) isocyanurate; tetrafunctional ones
 such as diglycerin tetra(meth)acrylate and pentaerythritol
 tetra(meth)acrylate; pentafunctional ones such as propionic
 acid-modified dipentaerythritol penta(meth)acrylate; and
 hexafunctional ones such as dipentaerythritol
 15 hexa(meth)acrylate and caprolactone-modified
 dipentaerythritol hexa(meth)acrylate. One type thereof may
 be used alone, or two or more types may also be used in
 combination. The number of functional groups of the
 polyfunctional acrylate may be preferably 3 to 15 and
 20 particularly preferably 3 to 6 from the viewpoints that a
 crosslinked structure can be readily formed and the coating
 elastic modulus of the release film 1B, 1C can be readily
 set to a value as described later. Among the above-
 described polyfunctional acrylates, the pentaerythritol
 25 triacrylate or dipentaerythritol hexaacrylate may be
 preferably used from the viewpoints that irregularities on
 the surface of the base material can be effectively
 absorbed and excellent smoothness of the release surface
 can be achieved.

30 [0053]

When the resin layer 13 is formed of a resin
 composition that contains an active energy ray-curable
 component, the resin composition may preferably further

contain a photopolymerization initiator. By containing the photopolymerization initiator, the active energy ray-curable component can be efficiently cured, and the polymerization curing time and the irradiance level of
5 active energy rays can be reduced.

[0054]

The photopolymerization initiator is not particularly limited and known ones can be used. When a polyfunctional acrylate is used as the active energy ray-curable component,
10 it may be preferred to use an α -aminoalkylphenone-based compound from the viewpoints of promoting the polymerization reaction and improving the curability. Examples of the α -aminoalkylphenone-based compound include 2-methyl-1[4-(methylthio)phenyl]-2-morpholinopropan-1-one,
15 2-benzyl-2-dimethylamino-1-(4-morpholinophenyl)-butanone-1, and 2-dimethylamino-2-[(4-methylphenyl)methyl]-1-[4-(4-morpholinyl)phenyl]-1-butanone. The content of the α -aminoalkylphenone-based compound in the resin composition may be preferably 1 to 20 mass parts, particularly
20 preferably 3 to 15 mass parts, and further preferably 5 to 10 mass parts, with respect to 100 mass parts of the polyfunctional acrylate.

[0055]

(2) Thermoset Component

25 The thermoset component is not particularly limited, provided that the component is cured by heating without impeding the effects of the present invention. In particular, it may be preferred to use a melamine resin, alkyd resin, epoxy resin, phenol resin, urea resin,
30 polyester resin, urethane resin, polyimide resin, benzoxazine resin, or acrylic resin as the thermoset component and it is particularly preferred to use a melamine resin. The use of a melamine resin can improve

the interfacial adhesion between the resin layer 13 and the release agent layer 12 to effectively suppress the delamination of the release agent layer 12 from the resin layer 13. The use of a melamine resin can also enhance the
5 coating elastic modulus of the release film 1B, 1C to improve the releasability when releasing the release film 1B, 1C from the ceramic green sheet.

[0056]

The melamine resin for forming the resin layer 13 is
10 not particularly limited, but it may be preferred to use the same type of melamine resin as that used for forming the release agent layer 12. In this case, the melamine resin of the resin layer 13 and the melamine resin of the release agent layer 12 exhibit high affinity, and high
15 interfacial adhesion can thereby be obtained between the resin layer 13 and the release agent layer 12; therefore, the delamination of the release agent layer 12 can be more effectively suppressed.

[0057]

20 (3) Other Components

The resin composition may contain additives, such as a crosslinker, reaction inhibitor, antistatic, and agent for improving the interfacial adhesion, in addition to the above-described components.

25 [0058]

(4) Thickness of Resin Layer

The thickness of the resin layer 13 may be preferably 100 to 3,000 nm, particularly preferably 300 to 2,000 nm, and further preferably 500 to 1,000 nm. When the thickness
30 of the resin layer 13 is 100 nm or more, irregularities on the surface of the base material 11 can be effectively absorbed, and excellent smoothness of the release surface can be achieved. When the thickness of the resin layer 13

is 3,000 nm or less, the occurrence of curling of the release film 1B, 1C can be effectively suppressed.

[0059]

4. Second Resin Layer

5 In the release film 1C, the second resin layer 14 is provided on the surface of the base material 11 opposite to the resin layer 13 (this surface may be referred to as a "base material back surface," hereinafter). The smoothness of the surface of the second resin layer 14 opposite to the
10 base material 11 is higher than the smoothness of the base material back surface because the second resin layer 14 absorbs the irregularities existing on the base material back surface. Moreover, when the release film 1C formed with a ceramic green sheet is wound into a roll-like shape,
15 the ceramic green sheet comes into contact with the surface of the second resin layer 14 having high smoothness, and the ceramic green sheet having higher smoothness can therefore be provided. Furthermore, the existence of the second resin layer 14 allows the curing shrinkage of the
20 resin layer 13 and/or the release agent layer 12 to be canceled with the curing shrinkage of the second resin layer 14, and the occurrence of curling of the release film 1C can thus be suppressed.

[0060]

25 The resin for forming the second resin layer 14 is not particularly limited. The previously-described resin used for forming the resin layer 13 can be used as the resin for the second resin layer 14. In the release film 1C, the resin layer 13 and the second resin layer 14 may be formed
30 of the same type of resin or may also be formed of different types of resins. However, from the viewpoint of suppressing the curling, the resin layer 13 and the second resin layer 14 may preferably be formed of the same type of

resin.

[0061]

The thickness of the second resin layer 14 can be set similarly to the resin layer 13. However, from the
5 viewpoint of effectively suppressing the occurrence of curling, the ratio of the thickness of the second resin layer 14 to the total thickness of the resin layer 13 and the release agent layer 12 may be preferably 0.2 to 2, particularly preferably 0.7 to 1.5, and further preferably
10 0.8 to 1.2.

[0062]

5. Physical Properties of Release Film for Ceramic Green Sheet Production Process

In the release film 1A, 1B, 1C, the coating elastic
15 modulus as measured by a nanoindentation test from the surface of the release agent layer 12 opposite to the resin layer 13 (or the base material 11) is 3.5 to 7.0 GPa and may be preferably 4.0 to 6.5 GPa and particularly preferably 4.5 to 6.3 GPa. When the coating elastic
20 modulus of the release film 1A, 1B, 1C is 3.5 GPa or more, good releasability can be effectively achieved when the release film 1A, 1B, 1C is released from the ceramic green sheet. When the coating elastic modulus of the release film 1A, 1B, 1C is 7.0 GPa or less, the release film 1A, 1B,
25 1C can be readily wound up.

[0063]

Measurement of the coating elastic modulus in the present description is performed by a nanoindentation test. Specifically, the release film 1A, 1B, 1C cut into a size
30 of 10 mm×10 mm is fixed to a glass plate, which adheres to an aluminum stage, using a two-component-type epoxy adhesive. In this operation for the release film 1A, 1B, the two-component-type epoxy adhesive is applied to the

surface of the base material 11 opposite to the release agent layer 12, and this surface is fixed to the glass plate. For the release film 1C, the two-component-type epoxy adhesive is applied to the surface of the second
5 resin layer 14 opposite to the base material 11, and this surface is fixed to the glass plate. Thereafter, the coating elastic modulus is measured from the surface of the release agent layer 12 opposite to the base material 11 using a microhardness evaluation apparatus (in the
10 exemplary test, using "Nano Indenter SA2" available from MTS Systems Corporation).

[0064]

The release force required when releasing the release film 1A, 1B, 1C from the ceramic green sheet formed on the
15 release surface of the release film 1A, 1B, 1C can be appropriately set, but may be preferably 5 to 100 mN/40 mm, particularly preferably 7 to 50 mN/40 mm, and further preferably 10 to 30 mN/40 mm. The release agent layer 12 of the release film 1A, 1B, 1C is formed using the release
20 agent composition which contains the polyorganosiloxane, and the release film 1A, 1B, 1C has moderate elasticity. It is therefore possible to appropriately set the release force, such as within a range of 5 to 100 mN/40 mm.

[0065]

25 When a ceramic green sheet is formed using the release film 1A, 1B, 1C, migration of the polyorganosiloxane from the release agent layer 12 to the ceramic green sheet is suppressed. Thus, when the ceramic green sheet is formed on the release surface of the release film 1A, 1B, 1C and
30 the ceramic green sheet is thereafter released from the release film 1A, 1B, 1C, the amount of migration of the polyorganosiloxane small at the surface of the ceramic green sheet which has been in contact with the release

surface. Specifically, when the ceramic green sheet is formed using the release film 1A, 1B, 1C, the silicon atomic ratio obtained by measuring the surface of the ceramic green sheet having been in contact with the release surface may be preferably less than 1.0 at%, particularly preferably less than 0.5 at%, and further preferably less than 0.3 at%. The silicon atomic ratio may be calculated in accordance with the equation below on the basis of the amounts (XPS counts) of silicon atoms (Si), carbon atoms (C), and oxygen atoms (O) which are measured, for example, by X-ray photoelectron spectroscopy (XPS).

$$\text{Silicon atomic ratio (at\%)} = \left[\frac{\text{(Si element amount)}}{\{(\text{C element amount}) + (\text{O element amount}) + (\text{Si element amount})\}} \right] \times 100$$

[0066]

A ceramic green sheet from which silicon is not detected by XPS (i.e. which does not contain a silicon compound) may be appropriately selected from ceramic green sheets for measurement, and the silicon atomic ratio thereof may be utilized as a reference for evaluating the amount of migration of the polyorganosiloxane from the release agent layer 12.

[0067]

The maximum peak height R_p of the release surface of the release film 1A, 1B, 1C may be preferably 5 to 800 nm, particularly preferably 10 to 400 nm, and further preferably 20 to 200 nm. In particular, in the release film 1B, 1C provided with the resin layer 13, the maximum peak height R_p of the release surface may be preferably 5 to 300 nm, particularly preferably 10 to 150 nm, and further preferably 20 to 75 nm. When the maximum peak height R_p of the release surface is within the above-described range, the ceramic green sheet formed using the

release film 1A, 1B, 1C can exhibit excellent smoothness, and a laminated ceramic product exhibiting excellent performance can be produced. Particularly in the release film 1B, 1C, the resin layer 13 is provided thereby to
5 allow the maximum peak height R_p to be a very small value as described above, and the ceramic green sheet having more excellent smoothness can be formed. The method of measuring the maximum peak height R_p of the release surface is as described in the exemplary test which will be
10 described later.

[0068]

6. Production Method for Release Film for Ceramic Green Sheet Production Process

When the release film 1A is produced, one surface of
15 the base material 11 may be coated with a coating liquid that contains the release agent composition and may further contain an organic solvent, if desired, and the coating film may then be dried and cured to form the release agent layer 12. The release film 1A can thus be obtained.

20 [0069]

When the release film 1B is produced, one surface of the base material 11 may be coated with a coating liquid that contains the resin composition and may further contain an organic solvent, if desired, and the coating film may
25 then be dried and cured to form the resin layer 13. Further, the surface of the resin layer 13 opposite to the base material 11 may be coated with a coating liquid that contains the release agent composition and may further contain an organic solvent, if desired, and the release
30 agent composition may then be cured by drying and heating to form the release agent layer 12. The release film 1B can thus be obtained.

[0070]

When the release film 1C is produced, one surface of the base material 11 may be coated with a coating liquid that contains the resin composition for the resin layer 13 and may further contain an organic solvent, if desired, and
5 the coating film may then be dried and cured to form the resin layer 13. Further, the other surface of the base material 11 may be coated with a coating liquid that contains the resin composition for the second resin layer 14 and may further contain an organic solvent, if desired,
10 and the coating film may then be dried and cured to form the second resin layer 14. Subsequently, the surface of the resin layer 13 opposite to the base material 11 may be coated with a coating liquid that contains the release agent composition and may further contain an organic
15 solvent, if desired, and the release agent composition may then be cured by drying and heating to form the release agent layer 12. The release film 1C can thus be obtained. The second resin layer 14 may be formed prior to the formation of the resin layer 13 or may also be formed
20 subsequent to the formation of the release agent layer 12.

[0071]

In the above-described methods, when the resin composition contains an active energy ray-curable component, the coating film formed of the resin composition may be
25 cured by irradiation with active energy rays. As the active energy rays, for example, electromagnetic wave or charged particle radiation having an energy quantum can be used and, specifically, ultraviolet rays, electron rays or the like can be used. In particular, ultraviolet rays may
30 be preferred because of easy management. Irradiation with ultraviolet rays can be performed using a high pressure mercury lamp, xenon lamp or the like, and the irradiance level of ultraviolet rays may be preferably about 50 to

1,000 mW/cm² as the illuminance. The light amount may be preferably 50 to 10,000 mJ/cm², more preferably 80 to 5,000 mJ/cm², and particularly preferably 200 to 2,000 mJ/cm². On the other hand, irradiation with electron rays can be performed using an electron ray accelerator or the like, and the irradiance level of electron rays may be preferably about 10 to 1,000 krad. Irradiation of the coating film with the active energy rays may be performed under an atmosphere of inert gas such as nitrogen.

10 [0072]

When the resin composition contains a thermoset component, the coating film formed of the resin composition may be cured by heating. The heating temperature in this case may be preferably 90°C to 140°C and particularly preferably 110°C to 130°C. The heating time may be preferably about 10 to 120 seconds and particularly preferably about 50 to 70 seconds.

[0073]

As the method of coating with the coating liquid in the above-described methods, for example, a gravure-coating method, bar-coating method, spray-coating method, spin-coating method, knife-coating method, roll-coating method, die-coating method, or other appropriate method can be used.

[0074]

25 The above-described organic solvent is not particularly limited and various types can be used. Examples of the organic solvent to be used include hydrocarbon compounds, such as toluene, hexane and heptane, isopropyl alcohol, isobutyl alcohol, acetone, ethyl acetate, methyl ethyl ketone, methyl isobutyl ketone, and a mixture thereof.

30 [0075]

7. Method of using Release Film for Ceramic Green Sheet

Production Process

The release film 1A, 1B, 1C can be used for producing a ceramic green sheet. Specifically, the release surface of the release agent layer 12 may be coated with a ceramic
5 slurry that contains a ceramic material, such as barium titanate or titanium oxide, and the ceramic slurry can then be dried to obtain a ceramic green sheet. The coating can be performed, for example, using an appropriate scheme, such as a slot die coating scheme or doctor blade scheme.

10 [0076]

Examples of binder components contained in the ceramic slurry include a butyral-based resin and acrylic-based resin. Examples of a solvent contained in the ceramic slurry include an organic solvent and water-based solvent.

15 [0077]

The release film 1A, 1B, 1C exhibits excellent releasability because the release agent layer 12 is formed of the release agent composition which contains the polyorganosiloxane and the release film 1A, 1B, 1C has
20 moderate elasticity. Moreover, in the release agent layer 12, the polyorganosiloxane is confined in a mesh-like structure formed by curing the melamine resin and therefore restricted from freely moving, and the migration of the polyorganosiloxane from the release agent layer 12 to the
25 ceramic green sheet is suppressed. This can enhance the interfacial adhesion between ceramic green sheets and suppress the occurrence of misalignment between the ceramic green sheets when they are laminated on one another. Furthermore, in the release film 1B, 1C provided with the
30 resin layer 13, the resin layer 13 absorbs the irregularities on the surface of the base material 11 thereby to achieve excellent smoothness on the release surface of the release agent layer 12. In addition, the

interfacial adhesion between the release agent layer 12 and the resin layer 13 is excellent because the release agent layer 12 is formed of the release agent composition which contains the melamine resin, and the occurrence of
5 delamination of the release agent layer 12 from the resin layer 13 is suppressed.

[0078]

It should be appreciated that the embodiments heretofore explained are described to facilitate
10 understanding of the present invention and are not described to limit the present invention. It is therefore intended that the elements disclosed in the above embodiments include all design changes and equivalents to fall within the technical scope of the present invention.

15 [0079]

For example, one or more other layers such as an antistatic layer may be provided between the base material 11 and the release agent layer 12 in the release film 1A, between the base material 11 and the resin layer 13 or
20 between the resin layer 13 and the release agent layer 12 in the release film 1B, 1C, or between the base material 11 and the second resin layer 14 in the release film 1C.

[Examples]

[0080]

25 Hereinafter, the present invention will be described further specifically with reference to examples, etc., but the scope of the present invention is not limited to these examples, etc.

[0081]

30 <Example 1>

(1) Formation of Resin Layer

A coating liquid of a resin composition was obtained through mixing 100 mass parts (equivalent amount to the

solid content, here and hereinafter) of pentaerythritol triacrylate (trade name: A-TMM-3L, available from Shin Nakamura Chemical Co., Ltd.) as an active energy ray-curable component and 10 mass parts of 2-methyl-1[4-
5 (methylthio)phenyl]-2-morpholinopropan-1-one (trade name: IRGACURE907, available from BASF Japan Ltd., α -aminoalkylphenone-based compound) as a photopolymerization initiator using toluene as a solvent. The solid concentration of the coating liquid was 15 mass%.

10 [0082]

A surface of a biaxially stretched polyethylene terephthalate film (thickness: 31 μm) as a base material was uniformly coated with the obtained coating liquid of the resin composition using a Meyer bar #6. The arithmetic
15 average roughness R_a of the surface was 10 nm and the maximum peak height R_p of the surface was 80 nm. Then, the coating film was dried at 80°C for 60 seconds and irradiated with ultraviolet rays (illuminance: 150 mW/cm^2 , light amount: about 350 mJ/cm^2) using an electrodeless lamp
20 under a nitrogen atmosphere (oxygen concentration of 1% or less) to cure the resin composition, and a laminate was obtained in which a resin layer having a thickness of 500 nm was laminated on the base material.

[0083]

25 (2) Formation of Release Agent Layer

A coating liquid of a release agent composition was obtained through mixing 100 mass parts of an imino-type methylated melamine resin (trade name: MX730, available from NIPPON CARBIDE INDUSTRIES CO., INC., mass-average
30 molecular weight: 1,508), 10 mass parts of polydimethylsiloxane (trade name: X-62-1387, available from Shin-Etsu Chemical Co., Ltd., mass-average molecular weight: 2,000), and 8 mass parts of p-toluenesulfonic acid

(trade name: Dryer 900, available from Hitachi Kasei Polymer Co., Ltd.) as an acid catalyst using toluene as a solvent. The solid concentration of the coating liquid was 2 mass%.

5 [0084]

The surface of the resin layer of the above laminate opposite to the base material was uniformly coated with the obtained coating liquid of the release agent composition using a Meyer bar #6. Then, the coating film was heated
10 and dried at 120°C for 60 seconds to cure the release agent composition, and a release agent layer having a thickness of 100 nm was formed on the above laminate. The release film was thus obtained.

[0085]

15 <Example 2>

A release film was obtained in the same manner as in Example 1 except that another type of coating liquid of a release agent composition was used and its coating film was heated and dried at 120°C for 60 seconds to form the
20 release agent layer. The coating liquid was prepared through mixing 100 mass parts of an imino-type methylated melamine resin (trade name: MX730, available from NIPPON CARBIDE INDUSTRIES CO., INC., mass-average molecular weight: 1,508), 0.1 mass parts of polyester-modified
25 hydroxyl group-containing polydimethylsiloxane (trade name: BYK-370, available from BYK Japan KK, mass-average molecular weight: 5,000), and 8 mass parts of p-toluenesulfonic acid (trade name: Dryer 900, available from Hitachi Kasei Polymer Co., Ltd.) as an acid catalyst using
30 toluene as a solvent. The solid concentration of the coating liquid was 15 mass%.

[0086]

<Example 3>

A release film was obtained in the same manner as in Example 1 except that the content of the polydimethylsiloxane in the release agent composition was 30 mass parts.

5 [0087]

<Example 4>

A release film was obtained in the same manner as in Example 1 except that the content of the polydimethylsiloxane in the release agent composition was
10 40 mass parts.

[0088]

<Example 5>

A release film was obtained in the same manner as in Example 1 except that the 10 mass parts of the
15 polydimethylsiloxane in the release agent composition were substituted by 30 mass parts of polyester-modified hydroxyl group-containing polydimethylsiloxane (trade name: BYK-370, available from BYK Japan KK, mass-average molecular weight: 5,000).

20 [0089]

<Comparative Example 1>

A coating liquid was obtained through mixing 100 mass parts of pentaerythritol triacrylate (trade name: A-TMM-3L, available from Shin Nakamura Chemical Co., Ltd.), 10 mass
25 parts of polydimethylsiloxane (trade name: X-62-1387, available from Shin-Etsu Chemical Co., Ltd., mass-average molecular weight: 2,000), and 10 mass parts of 2-methyl-1[4-(methylthio)phenyl]-2-morpholinopropan-1-one (trade name: IRGACURE907, available from BASF Japan Ltd., α -aminoalkylphenone-based compound) as a photopolymerization
30 initiator using toluene as a solvent. The solid concentration of the coating liquid was 15 mass%.

[0090]

A surface of a biaxially stretched polyethylene terephthalate film (thickness: 31 μm) as a base material was uniformly coated with the obtained coating liquid using a Meyer bar #6. The arithmetic average roughness R_a of the surface was 10 nm and the maximum peak height R_p of the surface was 80 nm. Then, the coating film was dried at 80°C for 60 seconds and irradiated with ultraviolet rays (illuminance: 150 mW/cm^2 , light amount: about 350 mJ/cm^2) using an electrodeless lamp under a nitrogen atmosphere (oxygen concentration of 1% or less) to cure the resin composition, and a release film was obtained in which a release agent layer having a thickness of 500 nm was laminated on the base material.

[0091]

15 <Comparative Example 2>

A release film was obtained in the same manner as in Example 1 except that another type of coating liquid of a release agent composition was used. The coating liquid was prepared through mixing 100 mass parts of an addition-type silicone-based release agent (trade name: CF-2172, available from Dow Corning Toray Co., Ltd., mass-average molecular weight: 300,000) and 2 mass parts of a platinum-based catalyst (BY24-835 available from Dow Corning Toray Co., Ltd.) using toluene as a solvent. The solid concentration of the coating liquid was 1.5 mass%.

[0092]

<Exemplary Test 1> (Measurement of Coating Elastic Modulus)

The release film obtained in each of the examples and comparative examples was cut into a size of 10 mm×10 mm, and the base material back surface of the cut release film was then fixed to a glass plate, which adhered to an aluminum stage, using a two-component-type epoxy adhesive. Subsequently, nanoindentation test was performed to measure

the coating elastic modulus of the above release film using a microhardness evaluation apparatus (Nano Indenter SA2 available from MTS Systems Corporation) under a condition of a maximum indenting depth of 100 nm, a strain speed of 5 0.05 sec⁻¹, a displacement amplitude of 2 nm, and a vibration frequency of 45 Hz. Results are listed in Table 1.

[0093]

<Exemplary Test 2> (Evaluation of Migration Property of 10 Polyorganosiloxane)

For the release film stored under an ordinary temperature for 48 hours after produced in each of the examples and comparative examples, an acrylic pressure sensitive adhesive tape (trade name: 31B Tape, available 15 from Nitto Denko Corporation) was attached to the release surface of the release agent layer and stored for 24 hours.

[0094]

Thereafter, the above pressure sensitive adhesive tape was released from the release film. For the surface of the 20 pressure sensitive adhesive tape which had been in contact with the release agent layer surface, the silicon atomic ratio (at%) was calculated in accordance with the equation below on the basis of the amounts (XPS counts) of silicon atoms (Si), carbon atoms (C), and oxygen atoms (O) which 25 were measured by X-ray photoelectron spectroscopy (XPS).

Silicon atomic ratio (at%)=[(Si element amount)/{(C element amount)+(O element amount)+(Si element amount)}]×100

[0095]

30 ESCA 5600 available from PerkinElmer was used as the measurement apparatus and the measurement condition was as follows.

X-ray source: Mg standard (15 kV, 400 W)

Takeoff angle: 45°

Measurement time: 3 minutes

Elements measured: silicon atoms (Si), carbon atoms (C), oxygen atoms (O)

5 [0096]

Then, the migration property of polyorganosiloxane was evaluated in accordance with the criteria below. Results of the silicon atomic ratio and evaluation are listed in Table 1.

10 A; The silicon atomic ratio was less than 0.35 at%.

B; The silicon atomic ratio was 0.35 at% or more and less than 0.5 at%.

C; The silicon atomic ratio was 0.5 at% or more and less than 1.0 at%.

15 D; The silicon atomic ratio was 1.0 at% or more.

[0097]

<Exemplary Test 3> (Evaluation of Interfacial Adhesion between Layers constituting Release Film)

The surface of the resin layer opposite to the base material (this surface may be referred to as a "resin layer top surface," hereinafter) before the release agent layer was formed in each of Examples 1 to 5 and Comparative Example 2, and the release surface of the release film obtained in each of the examples and comparative examples, 25 were rubbed ten times with a finger and the presence or absence of occurrence of smear (tarnish) and rub-off (drop-off) was visually determined under a fluorescent lamp. Then, the interfacial adhesion between layers constituting the release film was evaluated in accordance with the 30 criteria below. Results are listed in Table 1.

A; Smear and rub-off did not occur both when the resin layer top surface was rubbed and when the release surface was rubbed, and the interfacial adhesion was good.

B; At least one of smear and rub-off occurred when at least one of the resin layer top surface and the release surface was rubbed, and the interfacial adhesion was not good.

5 [0098]

<Exemplary Test 4> (Measurement of Release Force)

Ceramic slurry was prepared through adding 69 mass parts of toluene and 46 mass parts of ethanol to 100 mass parts of barium titanate (BaTiO_3 , trade name: BT-03, available from SAKAI CHEMICAL INDUSTRY CO., LTD), 8 mass parts of polyvinyl butyral (trade name: S-LEC B·K BM-2, available from SEKISUI CHEMICAL CO., LTD.), and 1.4 mass parts of dioctyl phthalate (trade name: dioctyl phthalate Cica first grade, available from KANTO CHEMICAL CO., INC.) and mixing and dispersing them in a ball mill.

[0099]

In the release film stored under an ordinary temperature for 48 hours after produced in each of the examples and comparative examples, the release surface of the release agent layer was uniformly coated with the above ceramic slurry using an applicator and thereafter the ceramic slurry was dried at 80°C for one minute using a dryer. Through this operation, a ceramic green sheet having a thickness of 3 μm was obtained on the release film. Thus, the release film with the ceramic green sheet was produced.

[0100]

The release film with the ceramic green sheet was stationarily placed under an atmosphere of a room temperature of 23°C and relative humidity of 50% for 24 hours. Then, an acrylic pressure sensitive adhesive tape (trade name: 31B Tape, available from Nitto Denko Corporation) was attached to the surface of the ceramic

green sheet opposite to the release film and, in this state, they were cut at a width of 20 mm. A measurement sample was thus obtained.

[0101]

- 5 The pressure sensitive adhesive tape side of the measurement sample was fixed to a flat plate and the release film was peeled off from the ceramic green sheet at a release angle of 180° and a release speed of 100 mm/min using a tensile tester (product name: AG-IS500N, available
10 from SHIMADZU CORPORATION) to measure the force necessary for the peeling (release force; mN/20 mm). Results are listed in Table 1.

[0102]

<Exemplary Test 5> (Measurement of Surface Roughness)

- 15 A double-sided tape was attached to a glass plate, and the release film obtained in each of the examples and comparative examples was fixed to the glass plate via the above double-sided tape so that the surface opposite to the release agent layer was on the side of the glass plate.
20 The release surface of the release film was observed at ×50 magnification in the PSI mode using an optical interferometric surface profile observation apparatus (product name: WYKO-1100, available from Bruker AXS), and the maximum peak height (R_p ; nm) was measured on the basis
25 of a range of $91.2 \times 119.8 \mu\text{m}$ in the obtained surface profile image. Results are listed in Table 1.

[0103]

[Table 1]

	Composition of release agent layer				Composition of resin layer	Coating elastic modulus (GPa)	Evaluation of migration property of polyorganosiloxane	Evaluation of interfacial adhesion	Release force (mN/40mm)	Maximum peak height Rp (nm)	
Example 1	Imino-type methylated melamine resin	100 mass parts	Polydimethylsiloxane	10 mass parts	Pentaerythritol triacrylate	100 mass parts	6.0	B	A	10	30
Example 2			Polyester-modified hydroxyl group-containing polydimethylsiloxane	0.1 mass parts			6.0	A	A	13	30
Example 3			Polydimethylsiloxane	30 mass parts			5.1	B	A	10	30
Example 4			Polydimethylsiloxane	40 mass parts			3.5	C	A	11	30
Example 5			Polyester-modified hydroxyl group-containing polydimethylsiloxane	30 mass parts			5.5	A	A	12	30
Comparative Example 1	Pentaerythritol triacrylate	100 mass parts	Polydimethylsiloxane		10 mass parts	6.1	D	A	18	50	
Comparative Example 2	Addition-type silicone-based release agent	100 mass parts			Pentaerythritol triacrylate	100 mass parts	2.8	D	B	12	40

[0104]

As apparent from Table 1, when the release film obtained in each of the examples was released from the ceramic green sheet, the release was possible with moderate
5 release force. Moreover, it has been found that the silicon atomic ratio is relatively small on the contact surface of the acrylic pressure sensitive adhesive tape which was in contact with the release film obtained in each of the examples. That is, it has been found that the
10 migration of polyorganosiloxane from the release agent layer to the ceramic green sheet formed using the release film of each of the examples is effectively suppressed. This suggests that, when a laminated ceramic product is produced, misalignment in the plane direction of laminated
15 layers of the ceramic green sheet due to migration of the polyorganosiloxane is suppressed. It has also been found that, in the release film obtained in each of the examples, the interfacial adhesion between the layers is sufficiently high and the delamination of the release agent layer from
20 the resin layer is less likely to occur. Furthermore, it has been confirmed that, in the release film obtained in each of the examples provided with the resin layer, the maximum peak height R_p of the release surface is relatively low and the release film is excellent also in the
25 smoothness as compared with the release film of Comparative Example 1 provided with no resin layer.

[0105]

On the other hand, it has been found that the silicon atomic ratio is relatively large on the contact surface of
30 the acrylic pressure sensitive adhesive tape which was in contact with the release film obtained in each of the comparative examples and the migration of polyorganosiloxane to the ceramic green sheet formed using

the release film is not effectively suppressed. It has also been found that, in the release film obtained in Comparative Example 2, the interfacial adhesion between the layers is low and the delamination is likely to occur.

5 Furthermore, it has been confirmed that, in the release film obtained in Comparative Example 1 provided with no resin layer, the maximum peak height R_p of the release surface is high and the smoothness is poor as compared with the release film of each of the examples.

10 [Industrial Applicability]

[0106]

The release film for a ceramic green sheet production process of the present invention is suitable for forming a ceramic green sheet to which polyorganosiloxane is less
15 likely to migrate.

[Description of Reference Numerals]

[0107]

1A, 1B, 1C... Release film for a ceramic green sheet production process

20 11... Base material

12... Release agent layer

13... Resin layer

14... Second resin layer

[Name of Document] Claims

1. A release film for a ceramic green sheet production process, the release film comprising a base material and a
5 release agent layer provided on one side of the base material,
 the release agent layer being formed by curing a release agent composition that contains a melamine resin and a polyorganosiloxane,
10 the release film having a coating elastic modulus of 3.5 to 7.0 GPa as measured by a nanoindentation test from a surface of the release agent layer opposite to the base material.
- 15 2. The release film for a ceramic green sheet production process as recited in claim 1, wherein the polyorganosiloxane has a mass-average molecular weight of 500 to 20,000.
- 20 3. The release film for a ceramic green sheet production process as recited in claim 1 or 2, wherein a content of the polyorganosiloxane in the release agent composition is 0.1 to 30 mass parts with respect to 100 mass parts of the melamine resin.
- 25 4. The release film for a ceramic green sheet production process as recited in any one of claims 1 to 3, wherein the melamine resin contained in the release agent composition contains methylated melamine and/or iminomethylol melamine.
- 30 5. The release film for a ceramic green sheet production process as recited in any one of claims 1 to 4, further comprising a resin layer between the base material and the

release agent layer.

6. The release film for a ceramic green sheet production process as recited in claim 5, wherein the resin layer is
5 formed by curing a resin composition that contains an active energy ray-curable component or a thermoset component.

7. The release film for a ceramic green sheet production
10 process as recited in claim 6, wherein the active energy ray-curable component is a polyfunctional acrylate.

8. The release film for a ceramic green sheet production process as recited in any one of claims 1 to 7, further
15 comprising a second resin layer provided on a side of the base material opposite to the release agent layer.