DESCRIPTION

Mould Release Film

Technical Field

This invention relates to a release film. More specifically, it relates to a release film that can fully satisfy the performances required of a release film for use in the production of a ceramic sheet.

Background Art

A release film using a polyester film as a base material is used as a carrier film for producing a ceramic sheet (green sheet), etc., used in the production of various ceramic electronic parts such as a stacked ceramic condenser, a ceramic substrate, etc.

A ceramic sheet to be used for producing a stacked ceramic condenser is produced, for example, by applying a ceramic slurry which is a dispersion of a ceramic powder, a binder, etc. in a solvent on a carrier film by reverse roll method, removing the solvent by drying under heat to form a ceramic layer, then, forming a metal film to constitute an internal electrode on the above ceramic layer by vapor deposition, printing, etc., thereby forming a metal film/ceramic layer/carrier film composite material, and removing the carrier film from the composite material by peeling it off.

A stacked ceramic condenser can be obtained by stacking the thus-produced metal film/ceramic layer composite materials in desirable dimensions, hot-pressing the stacked composite materials, then, cutting the hot-pressed product to have the form of a rectangle, thereby obtaining a chip-shaped stacked material, calcining the chip-shaped stacked material and forming

an external electrode on a predetermined surface of the calcined product.

Meanwhile, in recent years, it is demanded in the field of condensers such as stacked ceramic condensers to increase the density of circuit parts as they are to be downsized and greatly increased in capacity. Therefore, there arises a need to further and further decrease the thickness of a ceramic sheet used and to further increase the number of layers of the internal electrodes.

However, when the thickness of a ceramic sheet is decreased or when the number of stacked sheets is increased, even a slight non-uniformity in the thickness of a ceramic sheet causes the positional deviation of an internal electrode.

It is hence proposed that the thickness non-uniformity of a ceramic sheet to be produced should be decreased by decreasing the thermal deformation of a carrier film in the production of the ceramic sheet (see Patent Document 1). Patent Document 1 describes that since the thermal deformation under heat treatment is very small so long as the absolute values of dimensional change ratios under a stress of 1.47 MPa at 120°C both in the longitudinal direction and the width direction are 0.3 % or less, the non-uniformity in the thickness of a ceramic sheet to be obtained can be suppressed.

Generally, however, a ceramic sheet is dried at a temperature around 100°C without being held in the width direction. A carrier film used in the production of the ceramic sheet therefore shrinks in the state of being almost without any tension in the width direction when the ceramic sheet is dried. Therefore, the use of a release film having low thermal shrinkage factors in the longitudinal direction and in the width direction in the

state of being tensioned as described in Patent Document 1 can not fully overcome the shrinkage non-uniformity of a carrier film in all the steps up to the drying of a ceramic film. The thickness non-uniformity still takes place in a ceramic sheet produced, which leaves a problem that positional deviations of internal electrodes occur during their stacking.

Further, when a ceramic sheet has a small thickness, the high surface roughness of a carrier film causes a defect brought by the occurrence of a pin hole or the breaking of a ceramic sheet when it is peeled off, which leads to a decrease in productivity. That is, when a ceramic sheet has a small thickness, even impalpable surface defects such as scratches, foreign substances, etc., on a carrier film, which have been conventionally negligible problems, may be apparent as a cause for pin holes, etc., of a ceramic sheet to be obtained.

As the downsizing and capacity increase proceed recently in the field of condensers, therefore, a carrier film for producing a ceramic sheet is required to have higher-accuracy dimensional stability and higher-level smoothening of surface roughness.

On the other hand, when the surface of a carrier film is smoothened, peel charging tends to increase. For example, when the application speed of a ceramic slurry is increased for improving productivity, there is involved a problem that a spark is liable to take place in the step of taking out a carrier film, leading fire to easily break out. Further, a ceramic sheet is charged with electricity when the ceramic sheet is separated from a carrier film, and when the ceramic sheet is sacked in the subsequent step, the charge deviates the position of an internal electrode. In a carrier film

having a smoother surface, it is strenuously required to inhibit the peel charge.

In particular, in the production of a ceramic sheet having a small thickness (1 μm or less), it is essential not only to attain the above higher-accuracy dimensional stability and high-level smoothening (so-called surface roughness) but also to highly control the flatness of a film. That is, when a carrier film has a poor flatness, a coating of a ceramic slurry thereon comes to defectively have a thickness non-uniformity, and a ceramic sheet hence comes to defectively have a thickness non-uniformity, so that stacked ceramic condensers come to be non-uniform in capacity.

Meanwhile, the above carrier film is generally used in a state where it is taken up in the form of a roll. Since, however, it has a release layer on the surface, it is slippery and liable to cause a problem of the deformation of a roll form, etc., while it is taken up or carried. It is hence general practice to employ conditions under which the hardness of a roll is increased high so that the roll is not deformed in form when the above carrier film is taken up in the form of a roll. However, when the roll hardness is too high, the carrier film is liable to track the form of a roll, and it is prolonged due to an impalpable defect of the form of a roll and results in poor flatness.

In particular, in the production of a ceramic sheet having a small thickness (1 μm or less), it is essential to highly control the non-uniformity of the film. That is, when a carrier film defectively has a thickness non-uniformity, a coating of a ceramic slurry thereon comes to defectively have a thickness non-uniformity, and a ceramic sheet hence comes to defectively have a thickness non-uniformity, so that stacked ceramic

condensers come to be non-uniform in capacity.

(Patent Document 1) JP-A 2000-343663

Summary

In a first aspect, there is provided a release film having a release layer on at least one surface of a polyester film, wherein the release layer contains 0.5 mass% or more but 10 mass% or less, based on weight of the release layer, of a surfactant, wherein: the elongation ratio (S_{MD}) of the release film in the longitudinal direction at 100°C when a tension of 0.2 MPa or more but 4.0 MPa or less is exerted on the release film in the longitudinal direction of the release film satisfies the following expression (1), the elongation ratio (S_{TD}) of the release film in the direction perpendicular to the longitudinal direction at 100°C when a tension of 0.01 MPa is exerted on the release film in the direction perpendicular to the longitudinal direction of the release film satisfies the following expression (2), the thermal elongation ratio (HS_{MD}) of the release film in the longitudinal direction at 100°C when no load is exerted on the release film satisfies the following expression (3), the thermal elongation ratio (HS_{TD}) of the release film in the direction perpendicular to the longitudinal direction at 100°C when no load is exerted on the release film satisfies the following expression (4), and the thermal elongation ratio (HS_{MD}) in the longitudinal direction and the thermal elongation ratio (HS_{TD}) in the direction perpendicular to the longitudinal direction satisfy the following expression (5),

$$0.0961X - 0.45 \leq S_{MD} \leq 0.0961X - 0.25 \qquad (1)$$

wherein X is a tension (MPa) exerted on a film unit area, and X is a value of 0.2 MPa or more but 4.0 MPa or less,

$$-0.6 \leq S_{TD} \leq -0.2 \qquad (2)$$

$$-0.4 \leq HS_{MD} \leq -0.1 \qquad (3)$$

$$-0.6 \leq HS_{TD} \leq -0.2 \qquad (4)$$

$$HS_{MD} > HS_{TD} \qquad (5).$$

In a second aspect, there is provided a film roll that is a film roll obtained by taking up the release film as defined in the disclosure in the form of a roll, the roll having a surface layer having a Vicker's hardness (Hv) of 0 or more but 450 or less.

Disclosure of the Invention

This invention has been made in the light of the above prior art, and it is an object of this invention to provide a release film that has a appropriate dimensional change ratio under tension under heat when a ceramic sheet is produced, that is excellent in thermal shrinkage balance when a ceramic slurry is dried, and that fully satisfies the performances required for a release film used for the production of a ceramic sheet.

The present inventor has made diligent studies for overcoming the above problems. As a result, it has been found that a release film having a specific elongation ratio under a specific load and a thermal elongation ratio under no load can satisfy the performances required for a release film used for the production of a ceramic sheet, and this invention has been accordingly completed.

That is, this invention provides a release film having a release layer on at least one surface of a polyester film,

wherein:

the elongation ratio (S_{MD}) of the release film in the longitudinal direction at 100°C when a tension of 0.2 MPa or more but 4.0 MPa or less is exerted on the release film in the longitudinal direction of the release film satisfies the following expression (1),

the elongation ratio (S_{TD}) of the release film in the direction perpendicular to the longitudinal direction at 100°C when a tension of 0.01 MPa is exerted on the release film in the direction perpendicular to the longitudinal direction of the release film satisfies

the following expression (2),

the thermal elongation ratio (HS_{MD}) of the release film in the longitudinal direction at 100°C when no load is exerted on the release film satisfies the following expression (3),

the thermal elongation ratio (HS_{TD}) of the release film in the direction perpendicular to the longitudinal direction at 100°C when no load is exerted on the release film satisfies the following expression (4), and

the thermal elongation ratio (HS_{MD}) in the longitudinal direction and the thermal elongation ratio (HS_{TD}) in the direction perpendicular to the longitudinal direction satisfy the following expression (5),

$$0.0961X - 0.45 \leq S_{MD} \leq 0.0961X - 0.25 \quad (1)$$

wherein X is a tension (MPa) exerted on a film unit area, and X is a value of 0.2 MPa or more but 4.0 MPa or less,

$$-0.6 \leq S_{TD} \leq -0.2 \quad (2)$$

$$-0.4 \leq HS_{MD} \leq -0.1 \quad (3)$$

$$-0.6 \leq HS_{TD} \leq -0.2 \quad (4)$$

$$HS_{MD} > HS_{TD} \quad (5).$$

Further, the release film of this invention preferably has a maximum height (Rmax) in the range of 100 nm or more but 600 nm or less when its release layer surface is measured with a stylus three-dimensional surface roughness measuring apparatus. Further, preferably, the release layer surface and the surface on the side having no release layer have a maximum height (Rmax) of 100 nm or more but 600 nm or less, respectively, when measured with a stylus three-dimensional surface roughness measuring apparatus. Further, the release layer preferably has a surfactant content of 0.5 mass% or more but 10 mass% or less based on the weight of the release layer. Further, preferably,

the thickness non-uniformity in the longitudinal direction is 3.0 % or less, and that in the lateral direction is 3.0 % or less. Further, preferably, the release layer is formed by applying a release-film-forming composition to a monoaxially stretched polyester film.

The release film of this invention is preferably a release film for producing a ceramic sheet. In particular, it is preferably a release film for producing a ceramic condenser.

This invention includes a film roll that is obtained by taking up the above release film in the form of a roll and whose roll surface layer has a Vickers hardness (Hv) of 0 or more but 450 or less. This invention includes a process of using, as a release film for a ceramic sheet, a film having the properties of the above expressions (1) to (5).

Best Mode for Carrying Out the Invention

In this invention, the machine direction in the formation of a film will be sometimes referred to as the longitudinal direction or the length direction, and the direction perpendicular to the longitudinal direction, sometimes as the width direction or lateral direction.

<Release film>

The release film of this invention is a release film that has a release layer on at least one surface of a polyester film and that has a specific elongation ratio under a specific load and has a specific thermal elongation ratio under no load. The release film of this invention will be explained below with regard to its properties and constitution.

[Elongation ratio (S_{MD}) in the longitudinal direction]

In the release film of this invention, when a

tension of 0.2 MPa or more but 4.0 MPa or less is exerted thereon in the machine direction of the release film, the elongation ratio (S_{MD}) thereof in the machine direction at 100°C satisfies the following expression (1). A ceramic layer/release film composite material is generally carried in a state where a tension is exerted thereon at a temperature around 100°C. When 100°C is selected as a temperature condition, therefore, a situation closer to an actual step can be estimated.

$$0.0961X - 0.45 \leq S_{MD} \leq 0.0961X - 0.25 \qquad (1)$$

In the expression (1), X is a tension (MPa) exerted on a film unit area, and X is a value of 0.2 MPa or more but 4.0 MPa or less.

When the elongation ratio (S_{MD}) in the longitudinal direction is smaller than the value of left-hand side of the above expression (1), the shrinking stress of the film in the longitudinal direction is greater than the carrying tension of a ceramic layer/release film composite material, and as a result, the film shrinks non-uniformly in the longitudinal direction to cause the thickness non-uniformity of a ceramic sheet. When the elongation ratio (S_{MD}) in the longitudinal direction is greater than the value of right-hand side of the expression (1), the shrinking stress of a film in the longitudinal direction is smaller than the carrying tension of a ceramic layer/release film composite material, so that the flatness of the film comes to be poor to cause the thickness non-uniformity of a ceramic sheet. When the elongation ratio (S_{MD}) is in the range satisfying the above expression (1), the shrinking stress in the longitudinal direction has a proper balance with the carrying tension of a ceramic layer/release film composite material, so that the thickness non-uniformity of a ceramic sheet to be

obtained in the longitudinal direction can be suppressed. From the above point of view, the release film of this invention is preferably an embodiment in which the elongation ratio (S_{MD}) in the longitudinal direction at 100°C when a tension of 0.3 MPa or more but 2.5 MPa or less is exerted thereon in the longitudinal direction of the release film satisfies the above expression (1).

The elongation ratio (S_{MD}) in the longitudinal direction and the elongation ratio (S_{TD}) in the width direction to be described later are determined on the basis of the following expression. In the expression, M_0 is a length of the film in the longitudinal direction or in the width direction before the elevation temperature is started and M is a length of the film in such a direction when a temperature of 100°C is reached after the elevation of the temperature is started. That is, the elongation ratio (S_{MD}) and the elongation ratio (S_{TD}) represent that the film is shrunken when they are minus, and they represent that the film is extended when they are plus.

$$\text{Elongation ratio } (S_{MD},\ S_{TD}) = (\Delta M/M_0) \times 100\ (\%)$$

$$\Delta M = M - M_0$$

[Elongation ratio (S_{TD}) in the direction perpendicular to the longitudinal direction (in the width direction)]

In the release film of this invention, the elongation ratio (S_{TD}) thereof in the width direction at 100°C when a tension of 0.01 MPa is exerted thereon in the width direction of the release film satisfies the following expression (2). When 100°C is selected as a temperature condition, a situation closer to an actual step can be estimated like the above case. The elongation ratio (S_{TD}) is determined according to the above expression on the basis of lengths of the film

before and after the heat treatment.

$$-0.6 \leq S_{TD} \leq -0.2 \qquad (2)$$

When the elongation ratio (S_{TD}) in the width direction is smaller than -0.6 %, the film shrinks to a great extent in the width direction in the carrying step of a ceramic layer/release film composite material and causes the thickness non-uniformity of a ceramic sheet. On the other hand, when the elongation ratio (S_{TD}) in the width direction is larger than -0.2 % and when the elongation ratio (S_{TD}) is larger than 0 (the film extends), the flatness of the film disintegrates due to the elongation of the film in the width direction while a ceramic layer/release film composite material is carried, and the thickness non-uniformity of a ceramic sheet is caused. Further, due to the poor balance between the shrinkage of a ceramic layer and the elongation of the release film, there is caused a problem that a ceramic layer is partly peeled off and separated. When the elongation ratio (S_{TD}) is over -0.2 % but 0 % or less, the balance between the shrinkage of a ceramic layer and the shrinkage of the release film is poor, and there is caused a problem that a ceramic layer is partly peeled off the release film and separated.

[Thermal elongation ratio (HS_{MD}) in the longitudinal direction and thermal elongation ratio (HS_{TD}) in the direction perpendicular to the longitudinal direction (in the width direction)]

In the release film of this invention, the thermal elongation ratio (HS_{MD}) in the longitudinal direction at 100°C under no load satisfies the following expression (3), the thermal elongation ratio (HS_{TD}) in the width direction at 100°C under no load satisfies the following expression (4) at the same time, and the thermal elongation ratio (HS_{MD}) in the longitudinal direction and

the thermal elongation ratio (HS_{TD}) in the width direction satisfy the following expression (5).

$$-0.4 \leq HS_{MD} \leq -0.1 \quad (3)$$

$$-0.6 \leq HS_{TD} \leq -0.2 \quad (4)$$

$$HS_{MD} > HS_{TD} \quad (5).$$

When the thermal elongation ratio (HS_{MD}) of the film in the longitudinal direction and the thermal elongation ratio (HS_{TD}) thereof in the width direction are in the above ranges, respectively, and the thermal elongation ratio (HS_{MD}) thereof in the longitudinal direction is made greater than the thermal elongation ratio (HS_{TD}) thereof in the width direction, the shrinkage of the release film in the longitudinal direction and the shrinkage thereof in the width direction are well balanced during the removal of a solvent contained in a ceramic layer by drying under heat after a ceramic slurry is applied, and as a result, the thickness non-uniformity of a dry ceramic layer to be obtained can be reduced.

The thermal elongation ratio (HS_{MD}) in the longitudinal direction and the thermal elongation ratio (HS_{TD}) in the width direction are determined on the basis of the following expression. In the expression, L_0 is a length of a film in the longitudinal or width direction before heat treatment, and L is a length of the film in the same direction above after the heat treatment. That is, the thermal elongation ratio (HS_{MD}) and the thermal elongation ratio (HS_{TD}) represent that a film is shrunken when they are minus, and they represent that the film is extended when they are plus.

Thermal elongation ratio (HS_{MD}, HS_{TD}) = $(\Delta L/L_0) \times 100$ (%)

$\Delta L = L - L_0$

[Maximum height (Rmax)]

The release film of this invention preferably has a maximum height (Rmax) of 100 nm or more but 600 nm or less when the release layer surface is measured with a stylus three-dimensional surface roughness measuring instrument. The maximum height (Rmax) is more preferably 200 nm or more but 550 nm or less, particularly preferably 300 nm or more but 500 nm or less.

The maximum height (Rmax) refers to a maximum height of portions obtained by subtracting a datum length from a roughness curve determined by means of a three-dimensional surface roughness meter on the basis of JIS Standard (B0601-2001: Surface roughness - Definition and Expression, B0651 - 2001: Stylus surface roughness measuring instrument). The maximum height (Rmax) is a maximum height of a profile and is a sum of a maximum value of peak heights of the profile and a maximum value of valley depths of the profile in the datum length. The stylus surface roughness measuring instrument (stylus instrument) described in B0651-2001 is a measuring instrument by which a stylus moves on a surface to measure deviations of profile of the surface, a parameter is calculated and a profile curve is recorded.

JIS B0601-2001 corresponds to ISO 4287:1997, and JIS B0651-2001 corresponds to ISO 3274: 1996.

The maximum height (Rmax) represents a maximum projection height and is useful as an indicator for a pinhole defect of a ceramic sheet. Specifically, when the maximum height (Rmax) of a release layer surface exceeds 600 nm, a ceramic sheet formed on a portion where the above maximum height (Rmax) exceeds 600 nm comes to have a small thickness, and as result, a pinhole defect is easily caused. On the other hand, when the Rmax of a release layer surface is less than 100 nm,

no lubricity is obtained, a taking-up property is poor, and productivity tends to be poor.

That is, when the maximum height (Rmax) of release layer surface of the release film of this invention is in the above numerical range, the release film is excellent in surface flatness and lubricity, and the concavo-convex shapes can be hence suppressed, so that a ceramic sheet having a thickness non-uniformity suppressed can be obtained. As a result, when the thus-obtained ceramic sheet is used to produce a ceramic condenser, there can be obtained a condenser in which the positional deviation of an internal electrode is more suppressed.

In the release film of this invention, preferably, its surface on the side having no release layer preferably has a maximum height (Rmax), measured with a stylus three-dimensional surface roughness meter, of 100 nm or more but 600 nm or less simultaneously with having the above described release layer surface. The maximum height (Rmax) is more preferably 200 nm or more but 550 nm or less, particularly preferably 300 nm or more but 500 nm or less.

The maximum height (Rmax) represents a maximum projection height on the surface on the side having no release layer, and is useful as an indicator for the pinhole defect of a ceramic sheet. Specifically, when the maximum height (Rmax) of the surface on the side having no release layer exceeds 600 nm, a portion where the above height (Rmax) exceeds 600 nm is pressed on a ceramic sheet when the ceramic sheet is taken up after the application and drying of a ceramic slurry, a concave portion is formed on a ceramic sheet surface, the concave portion has a decreased thickness, and as a result, a pinhole defect is liable to be caused.

Further, even if it does not lead to the pinhole defect, a ceramic sheet eventually has an extremely thin portion formed therein, which leads to a ceramic condenser defect.

That is, in the release film of this invention, the maximum height (Rmax) of the release layer surface and the maximum height (Rmax) of the surface on the side having no release layer are simultaneously adjusted to be in the above numerical range, whereby the release film is excellent in surface flatness and lubricity, so that the concavo-convex shapes of a ceramic sheet to be obtained can be more suppressed and that a ceramic sheet having a thickness non-uniformity more suppressed can be obtained. As a result, when the thus-obtained ceramic sheet is used to produce a ceramic condenser, the positional deviation of an internal electrode can be further suppressed. Further, the occurrence of pinholes in a ceramic sheet can be suppressed. Furthermore, the peeling property of a ceramic sheet is improved.

The maximum height (Rmax) can be attained by adjusting the condition for filtering a molten polymer and the embodiment of particles to be contained in a polyester film.

[Thickness non-uniformity in the longitudinal direction and thickness non-uniformity in the lateral direction]

The thickness non-uniformity of the release film of this invention in the longitudinal direction is preferably 3.0 % or less. Further, preferably, the thickness non-uniformity of the release film of this invention in the lateral direction is 3.0 % or less. In the release film of this invention preferably, the thickness non-uniformity in the longitudinal direction and the thickness non-uniformity in the lateral direction are simultaneously in the above numerical

range. In particular, when a release film excellent in freedom from thickness non-uniformity is used, the thickness non-uniformity of a ceramic sheet to be obtained can be suppressed to a higher degree. When such a release film is used to produce a ceramic sheet and when thus-obtained ceramic sheet is used to produce ceramic condensers, there can be obtained ceramic condensers having a more uniform capacity. From the above viewpoint, the thickness non-uniformity in the longitudinal direction is preferably 2.9 % or less, more preferably 2.5 % or less, particularly preferably 2.0 %. The thickness non-uniformity in the lateral direction is preferably 2.8 % or less, more preferably 2.6 % or less, particularly preferably 2.5 % or less.

The thickness non-uniformity in the longitudinal direction can be adjusted on the basis of a stretch ratio in the longitudinal direction. Further, it is also important to adjust an auxiliary heating temperature and a stretching temperature in the step of stretching in the longitudinal direction. The thickness non-uniformity in the lateral direction can be adjusted on the basis of stretch ratios in the longitudinal direction and in the lateral direction. Further, it is also important to adjust an auxiliary heating temperature and a stretching temperature in the step of stretching in the lateral direction.

<Method for producing release film>

The method for producing the release film of this invention having the above properties will be explained below. The release film of this invention is produced through the step of forming an unstretched polyester film, a primary stretching step, an in-line application step, a secondary stretching step and a thermal setting step as will be described below.

[Step of forming an unstretched polyester film]

Before obtaining the release film of this invention, first, a polyester material to be described later is extrusion-molded to form an unstretched polyester film in the step of forming an unstretched polyester film.

In the extrusion-molding, a molten sheet extruded through a die with an extruder is cooled to solidness on a cooling roll to obtain an unstretched polyester film. In this case, for decreasing coarse particles in a polymer, preferably, a molten polymer is filtered, prior to its melt-extrusion with a non-woven fabric filter formed of stainless small-gage wires having a wire diameter of 15 μm or less and an average opening of 10 μm or more but 30 μm or less, preferably 10 μm or more but 20 μm or less. When the number of coarse particles in the polymer is described in the above manner, the maximum height (Rmax) of each of the release layer surface and the surface on the side having no release layer in the release film can be brought into the numerical range of 100 nm or more but 600 nm or less.

After the above filtering treatment, it is preferred to filter the molten polymer through a filter having an average opening of 10 μm or more but 50 μm or less, more preferably 15 μm or more, but 30 μm or less, just before a die opening from the viewpoint that a thermally deteriorated substance can be further removed to a higher degree, and the maximum height (Rmax) can be brought into a more preferable numerical range.

Further, from the viewpoint that the unstretched polyester film is improved in the property of being planar, it is preferred to improve the intimate contact between a molten sheet extruded from a die and the cooling roll, and for example, an electrostatically

contacting method and/or a liquid application contacting method are/is preferably employed.

[Primary stretching step]

In a primary stretching step, the unstretched polyester film obtained in the above step of forming an unstretched polyester film is stretched in the longitudinal direction (to be sometimes referred to as "longitudinal stretching" hereinafter) to obtain a monoaxially stretched polyester film in the longitudinal direction.

In this case, prior to the primary stretching step, it is preferred to pre-heat the unstretched film under a temperature condition of (Tg-10)°C or higher but (Tg-2)°C or lower beforehand, for obtaining a release film having a uniform thickness and having a desired elongation ratio (S_{MD}) and thermal elongation ratio (HS_{MD}) in the longitudinal direction. The above Tg stands for a glass transition temperature (unit: °C) of an unstretched polyester film.

In the primary stretching step, the unstretched polyester film that is pre-heated as required is stretched at a stretch ratio in the range of 3.3 times or more but 4.0 times or less in the longitudinal direction under a temperature condition of (Tg+2)°C or higher but (Tg+40)°C or lower.

When the stretch ratio is smaller than 3.3 times, undesirably, the thermal elongation ratio HS_{MD} in the longitudinal direction tends to be a plus value, that is, the film tends to be elongated. On the other hand, when the stretch ratio is greater than 4.0 times, undesirably, the elongation ratio (S_{MD}) in the longitudinal direction tends to be too small. Further, undesirably, the thermal elongation ratio (HS_{MD}) in the longitudinal direction tends to be too small. It is also

essential to employ a stretch ratio of 3.3 times or more but 4.0 times or less in order to adjust the elongation ratio (S_{MD}) and the thermal elongation ratio (HS_{MD}) to the desired numerical ranges.

Further, when the unstretched polyester film is stretched at a stretch ratio in the range of 3.7 times or more but 4.0 times or less in the longitudinal direction under a temperature condition of (Tg+2)°C or higher but (Tg+40)°C or lower, the thickness non-uniformity in the longitudinal direction can be adjusted to 3.0 % or less. When the stretch ratio in the longitudinal direction is too low, the thickness non-uniformity in the longitudinal direction tends to be deteriorated. From this point of view, the lower limit of the stretch ratio in the longitudinal direction is preferably 3.8 times or more. When the stretch ratio in the longitudinal direction is too high, the monoaxially stretched film obtained by monoaxial stretching in the longitudinal direction in the step of stretching in the longitudinal direction tends to be poor in thickness non-uniformity in the lateral direction, and oriented crystallization portions are partly formed, so that it is liable to be difficult to improve the thickness non-uniformity of the release film in the lateral direction.

[In-line application step]

In an in-line application step, a release-layer-forming composition (to be sometimes referred to as "coating agent" hereinafter) is applied to at least one surface of the polyester film monoaxially stretched in the longitudinal direction in an in-line application manner, whereby a polyester film having a coating film is obtained. That is, the release layer is formed by applying the release-layer-forming composition to the monoaxially stretched film.

As a coating agent for use in the in-line application step, it is preferred to use an aqueous coating liquid containing an aqueous thermosetting silicone resin composition to be described layer.

The method for the application is not specially limited, and any application method known as a method for applying an aqueous emulsion can be used. The coating agent can be applied onto the polyester film monoaxially stretched in the longitudinal direction obtained in the monoaxial stretching step, by a method of roller coating, spray coating, gravure coating, reverse gravure coating, slot coating, or the like.

The release film of this invention has a feature in that the coating agent is applied in an in-line application manner. In the release film of this invention, the thermal treatment to the release film is completed by applying the coating agent in an in-line application manner, then carrying out stretching in the second axial direction and further carrying out thermal setting. Thereafter, it is treated in an off-line manner, and no heat is applied. Therefore, the thus-obtained film can be used for the actual use purpose in a state in which those properties determined as target values for the production of the release film, in particular, the elongation ratio (S_{MD}) in the longitudinal direction, the elongation ratio (S_{TD}) in the width direction, the thermal elongation ratio (HS_{MD}) in the longitudinal direction and the thermal elongation ratio (HS_{MD}) in the width direction are maintained as they are. That is, the release film of this invention is a release film excellent in dimensional stability since the target values of properties in the production of a release film by using the in-line application of a coating agent constitute the target values of final

properties of a release film.

On the other hand, in a method in which a coating agent is applied onto a once produced polyester film in an off-line application manner to form a release layer, there is required the step of removing a solvent contained in the coating agent by drying to cure a resin that forms a release layer. In the step of curing a resin that forms a release layer, it is required to apply a temperature around 150°C, so that a release film having a release layer formed by an off-line application manner has increased elongation ratios both in the longitudinal direction and the width direction, and undesirably, the film is elongated in the step of carrying a ceramic layer/release film composite material.

In the release film of this invention, the coating agent is applied in an in-line application manner to obtain a coating film. Thereafter, a solvent can be removed from the coating film by drying only by heat applied in each step of pre-heating after the application, stretching in the second axial direction and thermally setting, with result that the resin contained in the applied coating agent can be cured to be formed to the release layer and can be closely adhered to the polyester film. Therefore, no special step of curing the coating film by drying is required, so that a release film excellent in dimensional stability can be obtained without the cumbersome steps for obtaining the release film.

The release film of this invention has a feature in that the coating agent is applied onto the monoaxially stretched film in an in-line application manner. When this embodiment is employed, the adhesion between the release layer and the polyester film can be improved.

Further, the surfactant to be contained in the release layer can be easily adjusted with regard to its kind and amount, and there can be produced a higher effect on the improvement of prevention of peel charging.

The solid content in the coating agent is preferably 0.5 mass% or more but 30 mass% or less, more preferably 1 mass% or more but 15 mass% or less, particularly preferably 1.5 mass% or more but 10 mass% or less. When the solid content in the coating agent is less than 0.5 mass%, no uniform application is liable to be carried out since the coating agent is repelled on a polyester film surface during the application of the coating agent onto the polyester film surface. When it exceeds 30 mass%, there is sometimes caused a problem that fogging is caused in the release layer obtained, that the coating agent is liable to undergo gelling, or that an effect produced does not pay for an increase in the coating cost.

[Secondary stretching step]

In the secondary stretching step, a biaxially stretched film is obtained by stretching the polyester film having the coating film obtained in the in-line application step in the width direction (to be sometimes referred to as "in the lateral stretching" hereinafter).

In this case, prior to the secondary stretching step, preferably, auxiliary heating is carried out under a temperature condition of (Tg+10)°C or higher but (Tg+30°C) or lower, so that the solvent contained in the applied coating agent can be fully dried, and that the subsequent secondary stretching step enables uniform stretching.

In the secondary stretching step, the stretching is carried out under a temperature condition of (Tg+10)°C or higher but (Tg+80°C) or lower, preferably an auxiliary

heating temperature or higher but (Tg+70)°C or lower in the width direction at a stretch ratio in the range of 3.0 times or more but 5.0 times or less. When the stretch ratio in the width direction is in the above numerical range, the thus-obtained polyester film is excellent in freedom from thickness non-uniformity in the lateral direction. When the stretch ratio in the width direction is too low, the thickness non-uniformity in the lateral direction tends to be deteriorated. On the other hand, when the stretch ratio in the width direction is too high, the film is liable to break during the production thereof. From the above point of view, the stretch ratio in the width direction is more preferably 3.5 times or more but 4.5 times or less, still more preferably 3.9 times or more but 4.3 times or less, particularly preferably 4.0 times or more but 4.2 times or less.

In this invention, further, the planar stretch ratio (longitudinal stretch ratio x lateral stretch ratio) is preferably 15 or more, and the freedom from the thickness non-uniformity in each of the longitudinal direction and the lateral direction can be more improved. The planar stretch ratio is more preferably 16 or more.

In this invention, preferably, the stretch ratio (to be sometimes referred to as "longitudinal stretch ratio" hereinafter) in the primary stretching step and the stretch ratio (to be sometimes referred to as "lateral stretch ratio" hereinafter) in the secondary stretching step satisfies the relationship of longitudinal stretch ratio ≤ lateral stretch ratio. When the relationship of longitudinal stretch ratio ≤ lateral stretch ratio is satisfied, the elongation ratio (S_{MD}) in the longitudinal direction, the elongation ratio (S_{TD}) in

the width direction, the thermal elongation ratio (HS_{MD}) in the longitudinal direction and the thermal elongation ratio (HS_{TD}) in the width direction can be easily controlled such that they come to be desired values.

[Thermal setting step]

In the thermal setting step, the biaxially stretched polyester film obtained in the secondary stretching step is thermally set to obtain a release film.

Although differing depending upon the kind of the polyester constituting the polyester film, the thermally setting temperature condition is preferably in the range of (Tg+70)°C or higher but (Tm)°C or lower. For example, when the polyester is polyethylene terephthalate, it is preferably thermally set in the temperature range of 180°C or higher but 235°C or lower. Further, when the polyester is polyethylene-2,6-naphthalate, it is preferably thermally set in the temperature range of 185°C or higher but 240°C or lower. When the thermal setting is carried out in the above temperature range, the desired elongation ratios (S_{MD} and S_{TD}) and thermal elongation ratios (HS_{MD} and HS_{TD}) can be obtained. The above Tm refers to a melting point (unit: °C) of the polyester.

Further, desirably, the thermal setting is carried out not in one zone alone but in a plurality of separate zones, and it is preferably carried out in three or more zones under temperature control. For example, when the thermal setting is carried out in three zones, the temperature is controlled such that the temperature in the first zone is 180°C or higher but 210°C or lower, and that the temperature in the second zone is higher than that in the first zone and the highest in the three zones. And, the temperature in the third zone is lower

than that in the second zone and is set at 180°C or higher but 200°C or lower. When the thermal setting is carried out at the highest temperature in the second zone and at a lower temperature than that of the second zone in the third zone, the planeness of the release film to be obtained can be maintained well, and the thickness non-uniformity of a ceramic sheet can be reduced. The time period for the thermal setting is not specially limited, and it is preferred to carry out the thermal setting, for example, for 1 second or more but 60 seconds or less.

Further, for adjusting the thermal elongation ratio (HS_{TD}) of the release film in the with direction into the desired value, the film is preferably relaxed in the final zone of the thermal setting step while the rail width is narrowed by 2 % or more but 5 % or less.

[Cooling step] (Step as required)

The release film of this invention may be subjected to an optional cooling step after the thermal setting step. When the cooling step is provided, the release film to be obtained can maintain planeness well and the thickness non-uniformity of the ceramic sheet can be reduced.

In the cooling step, the cooling is preferably carried out at a temperature in the range of (Tg-30)°C or higher but (Tg+20)°C or lower, and it is preferably carried out in a plurality of separate zones like the above thermal setting step. When the cooling temperature is lower than the above numerical range, the thermal elongation ratios (HS_{MD}, HS_{TD}) sometimes tend to be too small. On the other hand, when the cooling temperature is higher than the above numerical range, undesirably, there is caused a phenomenon in which oblique orientation is intensified in side circumferential

portions even if physical properties in the center line portion of the film along the longitudinal direction are equivalent in all directions. The phenomenon in which oblique orientation is intensified in side circumferential portions along the longitudinal direction may take place when the above thermally setting temperature is on the lower limit side of the preferred temperature range, while its degree is relatively small.

<Polyester film>

[Polyester]

The polyester for forming the polyester film used in this invention may be a homopolyester or may be a copolyester.

When the polyester film used in this invention is formed of a homopolyester, it is preferably a homopolyester obtained by polycondensation of an aromatic dicarboxylic acid and an aliphatic glycol. The above aromatic dicarboxylic acid includes terephthalic acid, 2,6-naphthalenedicarboxylic acid, etc. Further, the aliphatic glycol includes ethylene glycol, diethylene glycol, 1,4-cyclohexanedimethanol, etc. The homopolyester for the polyester film used in this invention typically includes polyethylene terephthalate (PET), polyethylene-2,6-naphthalate (PEN), etc.

When the polyester to form the polyester film used in this invention is a copolyester, it is preferably a copolymer obtained by copolymerizing 20 mol% or less, based on the total acid components, of a dicarboxylic acid and/or a glycol as a third component.

Examples of the dicarboxylic acid that is a monomer component for the copolyester include isophthalic acid, terephthalic acid, 2,6-naphthalenedicarboxylic acid, adipic acid, sebacic acid, oxycarboxylic acid (e.g., p-

oxybenzoic acid, etc.), etc. These can be used singly or in combination of two or more of them. Further, the glycol to form a monomer component for the copolyester includes ethylene glycol, diethylene glycol, propylene glycol, butanediol, 1,4-cyclohexanedimethanol, neopentyl glycol, etc., and these can be used singly or in combination of two or more of them.

Of these, the material for the polyester film used in this invention is preferably a polyethylene terephthalate having ethylene terephthalate units amount of 80 mol% or more, preferably 90 mol% or more, or polyethylene-2,6-naphthalate having ethylene-2,6-naphthalate units amount of 80 mol% or more, preferably 90 mol% or more, and the polyethylene terephthalate is particularly preferred. Generally, the Tg of polyethylene terephthalate is a low temperature of around 78°C, and when a polyethylene terephthalate film has been used as a carrier film for the production of a ceramic sheet to be treated at a temperature around 100°C, in particular, its dimensional stability during the step has become problematic. In contrast thereto, the release film of this invention is excellent in dimensional stability, so that it fully exhibits performances as a release film even when it is used at a temperature by far over the Tg.

[Additive]

The polyester that is a raw material for the polyester film used in this invention may contain particles mainly for imparting lubricity when a film is formed. The kind of the particles to be incorporated is not specially limited so long as they can impart lubricity. Specifically, they can be particles, for example, of silica, calcium carbonate, magnesium carbonate, barium carbonate, calcium sulfate, calcium

phosphate, magnesium phosphate, kaolin, aluminum oxide, titanium oxide, or the like.

Further, such heat-resistant organic particles as described in JP-B 59-5216 and JP-A 59-217755 can be also used. As other heat-resistant organic particles, for example, there can be particles of a silicone resin, a thermosetting urea resin, a thermosetting phenolic resin, a thermosetting epoxy resin, a benzoguanamine resin, or the like. Further, there can be also used precipitated particles obtained by precipitating part of a metal compound such as a catalyst and finely dispersing the thus-precipitated particles during the production of a polyester.

The form of the particles for imparting the above lubricity are not specially limited, and it may be any one of the forms of a sphere, a mass, a rod, flatness, etc. Further, its hardness, specific gravity, color, etc., are not specially limited. Further, two or more kinds of these particles can be used in combination as required.

The average particle diameter of the above particles is preferably 0.1 μm or more but 1 μm or less, more preferably 0.2 μm or more but 0.5 μm or less. When the average particle diameter is less than 0.1 μm, the particles are liable to aggregate, and the dispersibility thereof is sometimes insufficient. When it exceeds 1 μm, the surface roughness of a polyester film obtained is too high, and it is difficult to adjust the maximum height (Rmax) of the release layer surface of the release film and the maximum height (Rmax) on the side having no release layer into 100 nm or more but 600 nm or less.

The content of the particles incorporated for imparting lubricity in the polyester film is preferably

0.01 mass% or more but 2 mass% or less, more preferably 0.01 mass% or more but 1 mass% or less. When the content of the particles is less than 0.01 mass%, the lubricity of the film tends to be insufficient. When it exceeds 2 mass%, the film surface is liable to be insufficient in flatness and smoothness.

The method of incorporating the particles into the polyester film is not specially limited, and conventionally known methods can be employed. For example, intended particles can be added at any stage of producing the polyester, while it is preferred to incorporate the particles at the stage of esterification or after the completion of an ester exchange reaction, and then to proceed with a reaction for polycondensation. Further, there may be also employed a method in which a slurry of the particles dispersed in ethylene glycol, water, etc., and polyester materials are blended by means of a vented kneading extruder, or a method in which dry particles and polyester materials are blended by means of a kneading extruder.

[Layer structure]

The layer structure of the polyester film in this invention is not specially limited, and it may be a single layer structure or a laminated layer structure. When it is a laminated layer structure, it may be a structure of four layers or more in addition to structures of two layers and three layers as far as it exceeds the scope of gist of the present invention. The laminated layer structure is not specially limited, either, and it includes laminated layer structures, for example, of A/B, A/B/A, A/B/C, A/B/A′, etc. The above A, B and C stand for layers formed of the above polyester, and they may be the same or different. A′ stands for a layer formed of a polyester which is structurally

similar in particular to A.

[Thickness of polyester film]

The thickness of the entire polyester film in this invention is not specially limited, while it is preferably 9 μm or more but 50 μm or less, more preferably 15 μm or more but 38 μm or less, particularly preferably 25 μm or more but 31 μm or less.

<Release layer>

[Silicone resin composition]

Preferably, the release layer in this invention is mainly formed from a silicone resin composition since it is stable as an emulsion. The silicone resin composition contains, as constituent components, a main agent composed of a polysiloxane having at least two unsaturated groups or at least two hydroxyl groups, or both of these per molecule and a crosslinking agent composed of a hydrogen polysiloxane having at least two hydrogen atoms bonding directly to a silicon atom per molecule. In this invention, an aqueous coating liquid (coating agent) containing the above silicone resin composition is applied to form a coating film, and the coating film is cured to form a release layer.

The silicone resin composition in this invention is preferably an aqueous silicone resin composition. The aqueous silicone resin composition is excellent in stability when it is prepared in the form of an emulsion, with result that the stability of the coating agent can be increased. Further, the surfactant to be described later can be selected from a wider choice of surfactants, for example, a surfactant containing more hydroxyl groups can be used, and there can be produced a higher effect on the improvement of prevention of peel charging. It is also preferred from the viewpoint of environmental protection to employ an aqueous coating

agent.

In this invention, the method for curing the silicone resin composition can be selected from thermal curing, ultraviolet curing, electron radiation curing, etc, and thermal curing is preferred above all. That is, a thermosetting silicone resin composition is preferred as a silicone resin composition. In this invention, a polyester film having a coating film is obtained in the in-line application step as described above, and then heat-treated in the thermal setting step. When the silicone resin composition is a thermosetting silicone resin composition, a crosslinking reaction is promoted by heat treatment in the thermal setting step, and the curing of the silicone resin composition can be fully proceeded with, so that a release layer having excellent properties such as excellent releasability can be obtained.

For the above reason, an aqueous thermosetting silicone resin composition is particularly preferred as the silicone resin composition in this invention. The above "mainly" is to mean that 75 mass% or more of the release layer, based on the release layer weight, is formed from the silicone resin composition.

The silicone resin composition may be any one of an addition polymerization type (the main agent is a polysiloxane having at least two unsaturated group per molecule) and a condensation type (the main agent is a polysiloxane having at least two hydroxyl groups per molecule). As an embodiment of the addition polymerization type, an embodiment in which the coating agent contains platinum as a catalyst is preferred, and as an embodiment of the condensation type, an embodiment in which the coating agent contains tin as a catalyst is preferred. Of these, an addition polymerization type is

preferred from the viewpoint of excellence in peel property. Further, as the crosslinking agent, there can be preferably used those recommended by a manufacturer of the main agent to be used at the same time.

Specific examples of the aqueous thermosetting silicone resin composition suitable for use in this invention will be described below.

1) An aqueous 400E silicone resin composition by Wacker Silicone (Adrian, Michigan). It contains polysiloxane, a platinum catalyst and a V20 crosslinking agent type comprising methyl hydrogen polysiloxane.

2) An aqueous X2-7720 silicone resin composition by Dow Corning (Midland, Michigan). It contains methyl vinyl polysiloxane and methyl hydrogen polysiloxane containing an X2-7721 crosslinking agent type comprising platinum polysiloxane.

3) Aqueous PC-105 silicone resin composition by PCL (Rhone-Poulenc Inc., Rock Hill, South Carolina). It contains methyl vinyl polysiloxane and methyl hydrogen polysiloxane comprising a PC-95 catalyst component containing platinum polysiloxane.

4) An aqueous PC-107 silicone resin composition by PCL (similar to PC-105). It contains the above PC-95 crosslinking agent.

5) An aqueous PC-188 silicone resin composition by PCL (similar to PC-105). It contains the above PC-95 crosslinking agent.

In addition, these aqueous thermosetting silicone resin compositions can be used as coating agents after their solid contents are properly adjusted by adding deionized water, etc.

[Silane coupling agent]

Further, the release layer in this invention preferably contains a silane coupling agent. The silane

coupling agent is preferably an organosilicon low-molecular weight compound having a reacting group that bonds to one or both of a polyester resin and a silicone resin composition. It is preferably an organosilicon low-molecular weight compound having, as the above reacting group, at least one of methoxy, ethoxy, silanol, vinyl, epoxy, (meth)acryl, amino, mercapto, chloro, hydroxyl and carboxyl groups, etc.

The content of the above silane coupling agent based on the solid content weight of the release layer is preferably 0.1 mass% or more but 20 mass% or less, more preferably 1 mass% or more but 10 mass% or less, particularly preferably 3 mass% or more but 7 mass% or less. When the above content is adjusted to the above numerical range, the adhesion of the release layer can be increased high.

[Surfactant]

Preferably, the release layer in this invention contains 0.5 mass% or more but 10 mass% or less, based on the solid content weight of the release layer, of a surfactant. When the release layer contains a surfactant in the above numerical range, the peel charge that occurs when the release film in the form of a roll is unrolled and the peel charge that occurs when the ceramic sheet is peeled off the release film can be suppressed. Further, when a stacked ceramic condenser is produced, the positional deviation of an internal electrode can be suppressed. Further, when a surfactant is added, the coating agent comes to have excellent wettability to the polyester film surface. As a result, the defect of local repellence to the coating agent is suppressed, and not only a uniform coating film can be obtained, but the pinhole defect that occurs when the ceramic sheet is peeled off can be suppressed.

From the above point of view, the content of the surfactant based on the total dry weight of the release layer is more preferably 1.0 mass% or more but 7.0 mass% or less, particularly preferably 2.0 mass% or more but 5.0 mass% or less. When the above content is less than 0.5 mass%, the peel charge increases. Further, the wettability of the coating agent to the polyester film surface tends to be insufficient. When it exceeds 10 mass%, undesirably, the peeling force from the ceramic sheet tends to be heavy peeling force.

The surfactant includes ionic surfactants (anionic surfactant, cationic surfactant and amphoteric surfactant) and ion-free surfactants (nonionic surfactant). Of these, a nonionic surfactant is preferred. When ionic surfactants such as an anionic surfactant, a cationic surfactant, an amphoteric surfactant, etc., are used, these surfactants are poison to the silicone resin composition that forms the release layer, and in some cases, the silicone resin composition are not fully cured.

Examples of the nonionic surfactant include surfactants of a polyoxyethylene type, a polyhydric alcohol fatty acid ester type, a polyhydric alcohol alkyl ether type, a nitrogen-containing type, etc., a nonionic silicone-containing surfactant, a nonionic fluorine-containing surfactant, etc.

Examples of the polyoxyethylene type surfactant include poly(oxyethylene)alkyl ether, poly(oxyethylene)alkyl phenyl ether, poly(oxyethylene) poly(oxypropylene)alkyl ether, poly(oxyethylene) fatty acid ester, poly(oxyethylene) sorbitan fatty acid ester, etc. Of these, the poly(oxyethylene)alkyl ether preferably includes poly(oxyethylene) alkyl ethers having an alkyl group having 12 or more carbon atoms,

such as polyoxyethylene lauryl ether, polyoxyethylene myristyl ether, polyoxyethylene cetyl ether, polyoxyethylene stearyl ether, polyoxyethylene oleyl ether, etc. The above alkyl group may be linear, or may be branched. Examples of the polyhydric alcohol fatty acid ester type surfactant include propylene glycol fatty acid ester, glycerin fatty acid ester, polyglycerin fatty acid ester, sorbitan fatty acid ester, cane sugar fatty acid ester, etc. Examples of the polyhydric alcohol alkyl ether type surfactant include alkyl polyglycoxide, etc. Examples of the nitrogen-containing type surfactant include alkyl diethanolamide, alkylamine oxide, etc.

Examples of the silicone-containing surfactant include polyether-modified silicone, polyglycerin-modified silicone, etc. Further, these modified silicone structure is classified into a side-chain modification type, a two-terminal modification type (ABA type), a one-terminal modification type (AB type), a two-terminal-side-chain modification type, a linear block type (ABn type) and a branched type, while a modified silicone of any type can be used.

Above all, as a surfactant in this invention, a polyoxyethylene surfactant and a silicone-containing surfactant are preferred. These are less likely to be poison to the silicone resin composition, and they can exhibit sufficient wettability. The silicone-containing surfactants are particularly preferred and are by far less likely to be poison, and they can attain better releasability and can produce a higher effect on the improvement of prevention of peel charge.

In this invention, the thickness of the release layer (i.e., the thickness of a dried release layer) is not specially limited, while it is preferably 20 nm or

more but 90 nm or less. Generally, when it is less than 20 nm, the exhibition of effects as the release layer such as small peeling strength is difficult. When it exceeds 90 nm, an effect produced does not pay for a cost required.

<Use of release film>

The release film of this invention can be used as a film for forming a resin sheet, a ceramic sheet, etc., or as a separator for an adhesive film for labels, medical use, office stationery supplies, etc.

Particulary, the release film of this invention can be suitably used as a carrier film for producing a ceramic sheet since it has satisfactory performances required of a release film to be used for the production of a ceramic sheet. When the release film of this invention is used to produce a ceramic sheet, a thin-film ceramic sheet with excellent accuracy is obtained. Further, the thus-obtained thin-film ceramic sheet can be suitably used for a stacked ceramic condenser in which increasing of the number of internal electrode layers is required along with downsizing and an increase in capacity.

When the ceramic sheet is produced, it is sufficient to apply a prepared ceramic slurry to the release layer surface of the release film of this invention and remove a solvent contained in the applied ceramic slurry by drying. The method for applying the ceramic slurry is not specially limited, and there can be used conventionally known application methods. For example, there can be employed a method in which a ceramic slurry prepared by dispersing a ceramic powder, a binder agent, etc., in a solvent is applied by a reverse roll method and removing the solvent by drying under heat. The binder agent for use is not specially

limited, and there can be used, for example, polyvinyl butyral. Further, the solvent for use is not specially limited, and there can be used ethanol, toluene, etc.

<Film roll>

The film roll of this invention is obtained by taking up the above-obtained release film in the form of a roll. The release film roll may be a master roll obtained directly from the step of producing the above release film, or may be a slit roll obtained by slitting the above master roll to a width and a length demanded by a customer.

In the film roll of this invention, the roll surface layer has a Vicker's hardness (Hv) of 0 or more but 450 or less. When the Vicker's hardness (Hv) of the roll surface layer is adjusted to the above numerical range, there can be obtained a release film excellent in flatness. Further, the release film roll is hardly deformed in form or unlikely to get wrinkled.

The Vicker's hardness refers to a quotient determined by dividing a load used to form a pyramid-shaped dent in a test surface with a four-sided pyramid presser of diamond having an angle of 136 degrees between opposite faces with a surface area calculated from a diagonal line of a permanent dent, and it is calculated on the following expression.

$$Hv = (2 \cdot P \cdot \sin(\alpha/2))/d^2$$

P: load (kg), d: average lengths of diagonal lines of dents (mm), and α: angle between opposite faces

Since α = 136 degrees, the above expression is rewritten as follows.

$$Hv = 1.854 \times p/d^2$$

Generally, when a film is taken up in the form of a roll, a thick portion of the film is tightly taken up as the roll diameter increases, and it sometimes has a so-

called band-like defect. In such a portion, the film is extended to impair the flatness of the film. This tendency becomes more conspicuous with an increase in the Vicker's hardness (Hv) of the roll surface layer. That is, when the Vicker's hardness (Hv) of the roll surface layer exceeds 450, the film is too tightly taken up, and the film is extended to excess, so that the film has a poor flatness. From the above point of view, the upper limit of the Vicker's hardness (Hv) of the roll surface layer is preferably 430 or less, more preferably 420 or less, particularly preferably 410 or less. On the other hand, when the Vicker's hardness (Hv) of the roll surface layer is too low, the roll form of the film is liable to be deformed, while a lower Vicker's hardness is preferred so long as such a problem does not take place. From the above point of view, the lower limit of the Vicker's hardness (Hv) of the roll surface layer is preferably 340 or more, more preferably 360 or more, particularly preferably 380 or more.

The above Vicker's hardness (Hv) of the roll surface layer can be attained by adjusting the take-up tension, nip pressure, etc., when the release film is taken up.

With regard to the take-up tension, it is required to set an initial tension at 49 N/m or less. When the initial tension is adjusted to the above numerical range, the roll surface layer can be imparted with a Vicker's hardness (Hv) of 450 or less. Further, the amount of air caught in the roll during the taking-up film comes to be proper, the influence that the thickness non-uniformity of the film has can be decreased, that is, the tightening of a take-up roll can be suppressed, and the roll is excellent in flatness. Further, the deformation of a roll form can be inhibited. When the initial

tension is too high, the Vicker's hardness (Hv) of the roll surface layer tends to become too high. Further, the amount of air caught during the taking-up tends to become small, and the film tends to be poor in flatness. From the above point of view, the upper limit of the initial tension is preferably 48 N/m or less, more preferably 47 N/m or less. When the initial tension is too low, the taking-up is destabilized, the film is liable to meander, so that the end faces of a roll tends to be uneven, that the film tends to get wrinkled or that a slitting failure tends to take place. Preferably, the initial tension is as low as possible so long as these problems do not take place. From the above point of view, the lower limit of the initial tension is preferably 30 N/m or more. The above initial tension represents a tension at a time when the taking up of the release film, which is substantially a commercial product, in the form of a roll is started, and it does not necessarily mean a tension immediately after the taking-up of the release film around a core is started. That is, for the purpose of decreasing those influences which are caused by foreign matter, damage, etc., on the core surface, there is generally employed a method in which the take-up tension is set in particular high immediately after the taking-up film is started until a several meters or several tens meters long film is taken up, and it does not show the take-up tension for such a portion.

Further, it is required to taper the tension so that the final tension is lower than the initial tension. Specifically, the ratio of the final tension to the initial tension (tension taper ratio) is required to be 80 % or less. When the tension taper ratio is adjusted to the above numerical range, the deformation of the

roll form can be suppressed while the Vicker's hardness (Hv) of the roll surface layer is maintained low. When the tension taper ratio is too high, the Vicker's hardness (Hv) of the roll surface layer tends to become too high. From the above point of view, the upper limit of the tension taper ratio is preferably 70 % or less, more preferably 60 % or less. On the other hand, although the tension taper ratio is preferably low from the viewpoint of the roll hardness, the final tension becomes too low when the tension taper ratio is too low, and the roll shows a tendency to the easy occurrence of problems such as uneven roll end faces, etc. From the above point of view, the lower limit of the tension taper ratio is preferably 30 % or more. In this invention, the take-up tension may be increased in some portions from the initial tension to the final tension so long as the take-up tension does not exceed 49 N/m. In the main portion of the roll, however, an embodiment in which the take-up tension is continuously gradually decreased is preferred, and an embodiment in which it is continuously gradually decreased at a constant rate is more preferred. When the above embodiment is employed for the tension taper, the internal stress in the take-up direction in the roll can be brought into 0 or more, and failures such as a crease defect in the lateral direction (T bar), air gap between films, etc., can be suppressed. The above main portion of a roll refers to a portion that is 5 mm or more outwardly away from the core surface layer and that is 5 mm or more inwardly apart from the roll surface layer.

On the basis of the above initial tension and tension taper ratio, it is required to adjust the final tension to 39 N/m or less. When the final tension is adjusted to the above numerical range, the roll surface

layer can be imparted with a Vicker's hardness (Hv) of 450 or less. When the final tension is too high, the Vicker's hardness (Hv) of the roll surface layer tends to become too high. From the above point of view, the upper limit of the final tension is preferably 38 N/m or less, more preferably 30 N/m or less. On the other hand, although the final tension is preferably low from the viewpoint of the roll hardness, the taking-up is destabilized and the film tends to easily meander when the final tension is too low, and the roll shows a tendency to the easy occurrence of problems such as uneven roll end faces, etc. From the above point of view, the lower limit of the final tension is preferably 10 N/m or more, more preferably 15 N/m or more.

As a nip pressure, it is required to adjust the initial nip pressure to 200 N/m or less. When the initial nip pressure is adjusted to the above numerical range, the roll surface layer can be imparted with a Vicker's hardness (Hv) of 450 or less. Further, the amount of air caught in the roll during the taking-up film comes to be proper, the influence by the thickness non-uniformity of the film can be decreased, that is, the tightening of a take-up roll can be suppressed, and the roll is excellent in flatness. Further, the formation of uneven end face of a roll can be inhibited. When the initial nip pressure is too high, the Vicker's hardness (Hv) of the roll surface layer tends to become too high. Further, the amount of air caught in the roll during the take-up tends to be small, and the film tends to be poor in flatness. From the above point of view, the upper limit of the initial nip pressure is preferably 180 N/m or less, more preferably 160 N/m or less, particularly preferably 120 N/m or less. When the initial nip pressure is too low, the taking-up tends to

be destabilized, and the deformation of the roll form is, or wrinkles are, liable to take place. From the above point of view, the lower limit of the initial nip pressure is preferably 50 N/m or more, more preferably 80 N/m or more.

The nip pressure may not be tapered, while the occurrence of winkles or pimples or the uneven end faces of the roll can be suppressed by tapering the nip pressure at a nip pressure taper ratio of -10 % or more but 10 % or less.

When the above initial nip pressure and the nip pressure taper are employed, the final nip pressure can be 220 N/m or less. When the final nip pressure is too high, however, the Vicker's hardness (Hv) of the roll surface layer tends to become too high. From the above point of view, the upper limit of the final nip pressure is preferably 170 N/m or less, more preferably 150 N/m or less, particularly preferably 140 N/m or less. When the final nip pressure is too low, the taking-up film tends to be destabilized. From the above point of view, the lower limit of the final nip pressure is preferably 50 N/m or more, more preferably 70 N/m or more, particularly preferably 90 N/m or more.

Examples

This invention will be further specifically explained hereinafter with reference to Examples and Comparative Examples, while this invention shall not be limited thereto unless it exceeds the scope of gist of the present invention.

<Methods for measurements and evaluations>

In Examples and Comparative Examples, the following items were measured and evaluated by the following methods.

(1) Average particle diameter of particles

Measured with a CP-50 centrifugal particle size analyzer (trade name) supplied by Shimadzu Corporation. On the basis of the cumulative curve of particles having particles diameters calculated from a centrifugal sedimentation curve obtained by the measurement and the amount of the existed particles, "an equivalent sphere diameter" that was a particle diameter corresponding to 50 mass percent was taken out and used as an average particle diameter (unit: μm) of the particles (see a book "Particle Size Measuring Technology" issued by Nikkan Kogyo Shinbunsha, 1975, pages 242-247).

(2) Maximum height (Rmax)

According to JIS Standard (B0601: Surface roughness - Definition and Presentation, B0651: Stylus surface roughness measuring instrument), a maximum height in an area was determined with a three-dimensional surface roughness tester (trade name: SE-3AK, supplied by Kosaka Laboratories, Inc.) under conditions of a magnification: 20,000 times, a scanning pitch: 2 μm, a scanning length: 1 mm, a scan line number: 100 and a cutoff: 0.25 mm, and an average of results of the above measurements of 10 areas was taken as Rmax (unit: nm).

(3) Elongation ratios under load (S_{MD}, S_{TD})

With TMA (trade name: SS6000, supplied by Seiko Instruments Inc.), a sample film having a width of 4 mm was treated by temperature-increasing at a humidity of 50 %RH at a temperature elevation rate of 10°C/minute from a start temperature of 30°C under a load of 0.3 MPa, 1.0 MPa or 2.5 MPa per unit area in the longitudinal direction at an inter-chuck distance of 20 mm.
An elongation ratio (S_{MD}) (unit: %) under a load condition of each of 0.3 MPa, 1.0 MPa or 2.5 MPa was determined from the elongation/shrink behavior of the

film when reached at 100°C and on the basis of the following expression. Similarly, the measurement was carried out by exerting a load of 0.01 MPa in the width direction, and an elongation ratio (S_{TD}) (unit: %) under this load condition was determined. The elongation ratios (S_{MD}, S_{TD}) were taken with regard to 10 samples each, and an average of them was determined for each.

Elongation ratio (S_{MD}, S_{TD}) = $(\Delta M/M_0) \times 100$ (%)

$\Delta M = M - M_0$

In the above expression, M_0 represents a length of a film in the longitudinal or width direction before the heat treatment, and M represents a length of the film in the same direction after the heat treatment. That is, when the elongation ratios (S_{MD}, S_{TD}) are minus, they show the film is shrunken, and when they are plus, they show that the film is extended.

(4) Thermal elongation ratios under no load (HS_{MD}, HS_{TD})

A film sample having a square area of approximately 30 cm^2, which had been accurately measured for a side length beforehand, was suspended in an oven set a temperature of 100°C, and held therein for 30 minutes under no load. After the 30 minutes, the film sample was taken out of the oven, and after it had room temperature, it was measured for a length or width. Thermal elongation ratios (HS_{MD}, HS_{TD}) (unit: %) were determined on the basis of the following expression. Elongation ratios (HS_{MD}, HS_{TD}) were taken with regard to 10 samples each, and an average of them was determined for each.

Thermal elongation ratio (HS_{MD}, HS_{TD})

$= (\Delta L/L_0) \times 100$ (%)

$\Delta L = L - L_0$

In the above expression, L_0 represents a length of a film in the longitudinal or width direction before the

heat treatment, and L represents a length of the film in the same direction after the heat treatment. That is, when the thermal elongation ratios (HS_{MD}, HS_{TD}) are minus, they show that the film is shrunken, and when they are plus, they show that the film is extended.

(5) Evaluation of surface flatness/smoothness of ceramic sheet (Alternative test for evaluation of practical properties)

(Condition 1) A ceramic slurry having the following composition was applied to a surface on the release layer side of a release film with a die coater, a ceramic layer to have a thickness after drying of 5 μm was formed, and 200 m of a ceramic layer/release film composite material was taken up.

(Condition 2) A roll of a release film having a width of 450 mm and a length of 2,000 m was prepared. A ceramic slurry having the following composition was applied to a surface on the release layer side of the release film with a die coater at a film carrying speed of 60 m/minute, a ceramic layer to have a thickness after drying of 5 μm was formed to give a ceramic layer/release film composite material having a length of 1,900 mm, and the composite material was taken up in the form of a roll.

Then, the ceramic layers were separated from the ceramic layer/release film composite materials obtained under Conditions 1 and 2 to give ceramic sheets. Both surfaces of each of the thus-obtained ceramic sheets (measurement object area: 1 m^2) were observed through a scanning laser microscope (supplied by Lasertec Corporation), and they were evaluated for flatness/smoothness properties on the basis of the following evaluation ratings.

[Composition for ceramic slurry]

·Barium titanate (average particle diameter: 0.7 μm, supplied by Fuji Titanium Industry Co., Ltd.): 100 parts

·Polyvinyl butyral resin (trade name: S-LEC BM-S, supplied by Sekisui Chemical Co., Ltd.)] 30 parts

·Plasticizer (dioctyl phthalate): 5 parts

·Toluene/ethanol solvent mixture (mixing ratio: 6:4): 200 parts

[Evaluation ratings for surface flatness/smoothness]

○: 2 or less craters (concave portions) per m^2, each crater having a depth of 0.5 μm or more (level having no problem in practical use)

△: over 2 but less than 6 craters (concave portions) per m^2, each crater having a depth of 0.5 μm or more (level sometimes having a problem in practical use)

×: 6 or more craters (concave portions) per m^2, each crater having a depth of 0.5 μm or more (level having a problem in practical use)

(6) Evaluation of peel charge of release film

When the evaluation in the above (5) (Condition 2) was carried out, an amount of peel charge caused by unrolling the release film was measured. A centralized potential measuring apparatus (trade name: static charge measuring instrument SV-10, supplied by Kasuga Electric Works Corp.) was set in a position vertically above at a distance of 5 cm from the surface of the release film immediately after it was unrolled (the inside surface of a film unrolled), and the amount of peel charge was measured in an atmosphere having a temperature of 22°C and a humidity of 44 %RH. While the release film having a total length of 2,000 was unrolled, peel charge amounts were measured at least 15 points every 100 meters, and an average of them was taken as a peel charge amount (unit: kV) of the release film. The

evaluation was made on the basis of the following evaluation ratings.

[Evaluation ratings for evaluation of peel charge of release film]

○: A peel charge amount of 2.0 kV or less (excellent in peel charge)

x: A peel charge amount of over 2.0 kV (defective in peel charge)

(7) Evaluation of peel charge of ceramic sheet

A ceramic sheet/release film composite material was obtained in the same manner as in the above (5) (Condition 2). On the surface of ceramic layer of the thus-obtained ceramic layer/release film composite material, a patterned Ni electrode print layer to have a thickness after drying of 3 μm was formed as a metal film by a screen printing method. Then, the thus-obtained metal film/ceramic layer/release film composite material was cut to a size of 300 mm x 300 mm to give a sheet sample. With regard to the thus-obtained sheet sample, the metal film/ceramic layer composite material was separated from the release film at a peel speed of 20 m/minute, and an amount of peel charge caused by this separation was measured. A centralized potential measuring apparatus (trade name: static charge measuring instrument SV-10, supplied by Kasuga Electric Works Corp.) was set in a position at a distance of 5 cm from the surface of the ceramic layer of the separated metal film/ceramic layer composite material, and the amount of peel charge was measured in an atmosphere having a temperature of 22°C and a humidity of 44 %RH. The measurement was made with regard to 100 sheet samples, and an average of them was taken as a peel charge amount (unit: kV). The evaluation was made on the following evaluation ratings.

[Evaluation ratings for evaluation of peel charge of ceramic sheet]

○: A peel charge amount of 20 kV or less (excellent in peel charge)

x: A peel charge amount of over 20 kV (defective in peel charge)

(8) Evaluation of peel of ceramic sheet

With regard to the sheet sample in the above (7), a peeling state when the metal film/ceramic layer composite material was separated from the release film was evaluated on the basis of the following evaluation ratings.

[Peeling evaluation ratings]

◎: Peeling force is to a proper degree, the metal film/ceramic layer does not break, and there is found no residual ceramic layer in the release film (level having no problem in practical use at all).

○: Slightly heavy separation force is required, slight breaking of the meal film/ceramic layer is found, or a slight residual ceramic layer in the release film is found (level of having no problem in practical use at all).

×: Heavy separation force is required, breaking of the metal film/ceramic layer is found, or a residual ceramic layer in the release film is found (level having a problem in practical use).

(9-1) Evaluation 1 of stacking of ceramic sheet (Alternative test for evaluation of practical properties)

A pattern-printed Ni electrode printed layer (having a thickness after drying of 3 μm) was stacked on the ceramic sheet obtained in the above (5) (Condition 1). Ten ceramic sheet/electrode stacked material layers obtained in the above manner were stacked with ends

portions on one side being a positioning reference, and a degree of positional deviation of the electrode printed layers was evaluated on the basis of the following evaluation ratings.

[Evaluation ratings for positional deviation]

◎: The positional deviation is less than 200 μm (level having no problem in practical use at all)

○: The positional deviation is 200 μm or more but less than 400 μm (level not having a problem in practical use)

×: The positional deviation is 400 μm or more (level having a problem in practical use)

(9-2) Evaluation 2 of stacking of ceramic sheet (Alternative test for evaluation of practical properties)

Ten cut and separated metal film/ceramic layer composite material layers obtained in the same manner as in the above (7) each were stacked by means of a stacking machine according to a method of detecting a position with a CCD camera, to give a stacked material. With regard to the thus-obtained stacked material, the amount of deviation of each layer was measured with a microscope using the first metal film/ceramic composite material layer as a reference, and the thus-obtained value was used as a positional deviation (unit: μm). The evaluation was made on the basis of the following evaluation ratings. The stacking was carried out immediately after each release film was separated.

[Ratings for evaluation of positional deviation]

◎: The positional deviation is less than 200 μm (level having no problem in practical use at all).

○: The positional deviation is 200 μm or more but less than 400 μm (level not having a problem in practical use).

×: The positional deviation is more than 400 μm (level of having a problem in practical use).

(10) Vicker's hardness (Hv) of roll surface layer

Measurements were made by the following method according to the procedures in JIS Z2244 (1961). The surface layer of each of release film rolls obtained in Examples was measured in 10 points along the width direction excluding a 5 mm portion from each end face, and a largest value was used as a Vicker's hardness (Hv).

(11) Uneven end face

Each of release film rolls obtained in Examples was evaluated for an uneven end face state on the basis of the following evaluation ratings.

[Evaluation ratings for uneven end face]

◎: A deviation in an uneven end face is 1 mm or less (level being suitably used without any problem)

○: A deviation in an uneven end face is over 1 mm but 2 mm or less (level being used without a problem)

△: A deviation in an uneven end face is over 2 mm but 3 mm or less (level having a slight problem but being usable)

×: A deviation in an end face is over 3 mm (level having a problem and being unusable)

(12) Flatness

A 2-m long film sample is taken from a release film roll, and spread on a horizontal and flat rest such that its surface which has been on the side of a roll surface faces upward. After it is left for 10 minutes, the entire surface of the film surface was observed to measure lengths (unit: cm) of remaining wrinkles (flutes, portions floating from the above rest) on the surface, and a total thereof is divided by a measurement area (unit: m^2) to calculate a flatness (unit: cm/m^2). The flatness was evaluated on the basis of the following

evaluation ratings.

[Evaluation ratings for flatness]

◎: The flatness is 28 cm/m^2 or less (level having no problem at all in practical used).

○: The flatness is over 28 cm/m^2 but 33 cm/m^2 or less (level not having a problem in practical use).

×: The flatness is over 33 cm/m^2 (level having a problem in practical use).

(13) Thickness non-uniformity of film

A film was measured for a thickness with a micrometer (trade name "K-402B" model supplied by Anritsu Corporation) in a total of 100 points consisting of selecting 10 points at intervals of 10 cm in the longitudinal direction and measuring at the above 10 points at each 10 lines at intervals of 10 cm in the lateral direction. An average of the thus-obtained film thicknesses of the 100 points was calculated and used as a film thickness (unit: μm).

Then, the film was measured with an electronic micrometer (trade name "K312A" model supplied by Anritsu Corporation) under a stylus pressure of 30 g, at a running speed of 25 mm/second, along a length of 1 m in the longitudinal direction of the film and along a width of 450 mm in the lateral direction of the film, to obtain a continuous thickness chart. A maximum thickness (unit: μm) and a minimum thickness (unit: μm) in each of the longitudinal direction and the lateral direction were taken from the thus-obtained chart, and the thickness non-uniformity (unit: %) was determined on the basis of the following expression using the above film thickness in combination.

Thickness non-uniformity (%) = ((maximum thickness − minimum thickness)/film thickness) x 100

(14) Thickness non-uniformity of ceramic sheet

The ceramic layer/release film composite material obtained in the above (5) (Condition 2) was measured for a thickness with a micrometer (trade name "K-402B" model supplied by Anritsu Corporation). Then, the ceramic layer in a thickness-measured portion was completely separated, and the same portion was measured for a thickness again. A difference between them was taken as a thickness of the ceramic sheet. These procedures were repeated in a total of 100 points consisting of selecting 10 points at intervals of 1 m in the longitudinal direction and measuring at the above 10 points at each 10 lines at intervals of 1 cm in the lateral direction. An average of ceramic sheet thicknesses obtained in the 100 points was taken as a ceramic sheet thickness (unit: μm). Then, of the measurement values in the above 100 points, the largest was used as a maximum thickness (unit: μm), the smallest was used as a minimum thickness, and the thickness non-uniformity (%) was determined on the basis of the following expression.

Thickness non-uniformity (%) = ((maximum thickness - minimum thickness)/(ceramic sheet thickness) x 100

The evaluation was made on the basis of the following evaluation ratings.

[Evaluation ratings for thickness non-uniformity]

◎: The thickness non-uniformity is 2.0 % or less (level being remarkably excellent free from thickness non-uniformity and having no problem in practical use)

○: The thickness non-uniformity is over 2.0 % but 3.0 % or less (level being excellent free from thickness non-uniformity and not having a problem in practical use)

×: The thickness non-uniformity is over 3.0 % (level being poor free from thickness non-uniformity and

having a problem in practical use)

Example 1

[Preparation of polyester]

A manganese acetate-tetrahydrate as an ester exchange catalyst was added to a mixture containing 100 parts by weight of dimethyl terephthalate and 70 parts of ethylene glycol in such an amount that the content of manganese element in a polyester to be obtained was to be 80 ppm, and an ester exchange reaction was carried out while the internal temperature was gradually increased from 150°C. When 95 % of the ester exchange reaction was completed, 0.01 part of phosphorous acid as a stabilizer was added, and after the mixture was fully stirred, 0.03 part of antimony trioxide was added. Thereafter, water included in the system was fully distilled off, and then synthetic calcium carbonate particles having an average particle diameter of 0.6 μm as an internal additive filler (lubricant) were added in such an amount that the content thereof based on the mass of a polyester to be obtained was to be 0.2 mass%, followed by fully stirring. Then, a reaction product was transferred into a polymerizer, and subjected to polycondensation under high-temperature vacuum (final internal temperature 295°C) to give a polyethylene terephthalate composition having an intrinsic viscosity of 0.65 (in o-chlorophenol at 35°C).

[Preparation of coating agent]

A coating agent was obtained by adding 0.5 part of a silane coupling agent (trade name: KBM-403, supplied by Shin-Etsu Silicone K.K.) to a mixture of 88.5 parts of deionized water, 10 parts of a silicone emulsion 400E (silicone supplied by Wacker Silicone AG: methylpolysiloxane having a vinyl group, and optionally

containing in combination an inhibitor for preventing a premature reaction with a platinum catalyst when a crosslinking agent is added) and 1 part of a crosslinking agent V72 (supplied by Wacker Silicones AG, an emulsion of methyl hydrogen polysiloxane, which reacts with a double bond in methyl siloxane). It had a solid content of 5 mass%.

[Step of forming unstretched polyester film]

The above-obtained polyethylene terephthalate composition was dried at 170°C for 5 hours until a polymer had a water content of 0.05 mass%. Then, the dry polyethylene terephthalate composition was fed to an extruder, melted at a melting temperature of 280 to 300°C, highly accurately filtered with a steel wire filter having an average mesh opening of 11 μm, and then extruded through a T-die, and the extrudate was rapidly cooled by bringing it into contact with a cooling drum by an electrostatically contacting method to give an unstretched polyester film having a thickness of 450 μm.

[Primary stretching step]

The thus-obtained unstretched polyester film was pre-heated at 75°C, and then stretched 3.6 times in the longitudinal direction between low-speed and high-speed rolls at a film temperature of 105°C, and then the stretched film was rapidly cooled to give a polyester film stretched in the longitudinal direction (length direction).

[In-line application step]

Then, the above-prepared coating agent was applied to the thus-obtained stretched polyester film stretched in the longitudinal direction, such that a dry coating had a thickness of 40 nm, to give a polyester film having a coating film.

[Secondary stretching step]

The polyester film having a thus-obtained coating film was supplied to a stenter and pre-heated in two zones at 105°C and 115°C for 2 seconds in each zone. Then, the pre-heated polyester film was uniformly stretched in four zones at 120°C, 130°C, 145°C and 155°C for 2 seconds in each zone such that its stretch ratio (lateral stretch ratio) in total became 4.1 times in the direction perpendicular to the longitudinal direction (width direction) to obtain a biaxially stretched polyester film.

[Thermally setting step]

The thus-obtained biaxially stretched polyester film was thermally set in three zones at 210°C, 225°C and 195°C for a total of 6 seconds, 2 seconds in each zone, and in the last thermally setting zone at 195°C, it was relaxed by 2.5 % in the direction perpendicular to the longitudinal direction (width direction), to give a release film having a total thickness of 31 μm. The thus-obtained release film was used for various measurements and evaluations. Table 1 shows the results.

Example 2

A release film was obtained in the same manner as in Example 1 except that the stretch ratio in the direction perpendicular to the longitudinal direction (width direction) in the secondary stretching step was changed to 4.5 times and that the amount of relaxation in the third zone in the thermally setting step was changed to 4.0 %. Table 1 shows the results of various measurements and evaluations made with regard to the thus-obtained release film.

Comparative Example 1

A biaxially stretched polyester film having no

release layer was obtained in the same manner as in Example 1 except that the release coating liquid containing the aqueous thermosetting silicone composition in an in-line step was not applied the production of a release film.

Further, to a solution of an addition polymerization type silicone compound (trade name: TPR-6721, supplied by Toshiba Silicone K.K.) in toluene (solid content: 3 mass%) was added a Pt catalyst (trade name: CM670, supplied by Toshiba Silicone K.K.) in such an amount that its content came to be 1 part by mass per 100 parts by mass of the addition polymerization type silicone compound, whereby a release agent coating liquid was prepared.

Thereafter, the above-obtained biaxially stretched polyester film roll having no release layer was unrolled, and the above-prepared release agent coating liquid was applied to the unrolled biaxially stretched polyester film in its center portion in the direction perpendicular to the longitudinal direction (width direction) such that the application amount (wet) came to be 6 g/m^2. A release layer was formed by drying the applied coating liquid with an air floating and carrying type drying apparatus having lower and upper air-blowing ports at intervals of 38 cm each at a carrying tension of 2,000 kPa at a drying temperature of 160°C for 16 seconds, to give a release film having a dry cured release layer having a weight of 0.2 g/m^2. Table 1 shows the results of various measurements and evaluations made with regard to the thus-obtained release film.

Comparative Example 2

A release film was obtained in the same manner as in Example 1 except that the stretch ratio in the

longitudinal direction (length direction) in the primary stretching step was changed to 3.0 times. Table 1 shows the results of various measurements and evaluations made with regard to the thus-obtained release film.

Comparative Example 3

A release film was obtained in the same manner as in Example 1 except that the stretch ratio in the longitudinal direction (length direction) in the primary stretching step was changed to 4.8 times and that the stretch ratio in the direction perpendicular to the longitudinal direction (width direction) was changed to 3.0 times. Table 1 shows the results of various measurements and evaluations made with regard to the thus-obtained release film.

Table 1

			Example 1	Example 2	Comparative Example 1	Comparative Example 2	Comparative Example 3
Elongation ratio (S_{MD})	Load 0.3MPa	%	-0.31	-0.28	-0.05	-0.2	-0.48
	Load 1.0MPa	%	-0.24	-0.19	0.02	-0.14	-0.38
	Load 2.5MPa	%	-0.1	-0.08	0.13	0.05	-0.3
Elongation ratio(S_{TD})	Load 0.01MPa	%	-0.39	-0.43	0	-0.62	-0.1
Thermal elongation ratio (HS_{MD})	No load	%	-0.2	-0.18	-0.05	-0.1	-0.5
Thermal elongation ratio (HS_{TD})	No load	%	-0.4	-0.5	0	-0.7	-0.1
HS_{MD}-HS_{TD}	MD-TD		0.2	0.32	-0.05	0.6	-0.4
Rmax	Release layer surface	nm	450	448	452	440	455
	Side having no release layer	nm	460	455	464	450	465
Evaluations of ceramic sheet	Positional deviation (Evaluation 1 of stacking)		◎	◎	×	×	×
	Surface smoothness and flatness (Condition 1)		○	○	○	○	○

Example 3

[Preparation of polyester]

A polyethylene terephthalate composition having an intrinsic viscosity of 0.65 (in o-chlorophenol at 35°C) was obtained in the same manner as in Example 1.

[Preparation of coating agent]

A coating agent was obtained by adding 10 parts of a silicone emulsion 400E (silicone supplied by Wacker Silicones AG: methylpolysiloxane having a vinyl group, and optionally containing in combination an inhibitor for preventing a premature reaction with a platinum catalyst when a crosslinking agent is added, solid content 50 mass%) as a main component, 1 part of a crosslinking agent V72 (supplied by Wacker Silicones AG, an emulsion of methyl hydrogen polysiloxane, which reacts with a double bond in methyl siloxane, solid content 50 mass%), 0.3 part of a silane coupling agent (trade name: KBM-403, supplied by Shin-Etsu Silicone K.K.) and 0.15 part of polyoxyethylene oleyl ether (trade name: Emulgen 404, supplied by Kao Corporation) (S1 component) as a nonionic surfactant to 87 parts of deionized water with stirring. The coating agent had a solid content of 6.0 mass%. The solid content ratios of components based on 100 mass% of a release layer to be obtained from the above coating agent were to be as follows.

Main component: 84.1 mass%

Crosslinking agent: 8.4 mass%

Silane coupling agent: 5.0 mass%

Surfactant: 2.5 mass%

[Step of forming unstretched polyester film]

The above-obtained polyethylene terephthalate composition was dried at 170°C for 5 hours until the polyethylene terephthalate composition had a water

content of 0.05 mass% or less. Then, the dry polyethylene terephthalate composition was fed to an extruder, melted at a melting temperature of 280 to 300°C, highly accurately filtered with a steel wire filter having an average mesh opening of 11 μm, and then extruded through a T-die to form a molten sheet, and the molten sheet was rapidly cooled by bringing it into contact with a cooling drum by an electrostatically contacting method to give an unstretched polyester film having a thickness of 450 μm.

[Primary stretching step]

A monoaxially stretched polyester film stretched in the longitudinal direction was obtained from the thus-obtained unstretched polyester film in the same manner as in Example 1.

[In-line application step]

Then, the above-prepared coating agent was applied to the thus-obtained monoaxially stretched polyester film stretched in the longitudinal direction, such that the resultant release film had a release layer having a thickness of 40 nm, to give a polyester film having a coating film. The above application of the coating agent was carried out on the side of the polyester film which had not been brought into contact with the cooling drum in the step of forming an unstretched polyester film.

[Secondary stretching step]

Then, the thus-obtained polyester film having a coating film was supplied to stenter and stretched in the same manner as in Example 1 to give a biaxially stretched polyester film.

[Thermally setting step]

The thus-obtained biaxially stretched polyester film was treated in the same manner as in Example 1 to give a release film having a total thickness of 31 μm.

The thus-obtained release film was subjected to various measurements and evaluations. Table 2 shows the results.

Example 4

A release film was obtained in the same manner as in Example 3 except that the conditions in some steps were changed as follows. The thus-obtained release film was subjected to various measurements and evaluations. Table 2 shows the results.

[Preparation of coating agent]

As a surfactant, 0.06 part of a polyoxyethylene-methylpolysiloxane copolymer (trade name: EMALEX SS-5051, supplied by Nihon Emulsion Co., Ltd.) (S2 component) that was a nonionic silicone-containing surfactant was used in place of the polyoxyethylene oleyl ether. The coating agent had a solid content of 6.0 mass%. Further, the solid content ratios of components based on 100 mass% of a release layer to be obtained from the above coating agent were as follows.

Main component: 85.4 mass%

Crosslinking agent: 8.5 mass%

Silane coupling agent: 5.1 mass%

Surfactant: 1.0 mass%

[Secondary stretching step]

The stretch ratio in the lateral direction was changed to 4.5 times.

[Thermally setting step]

The amount of relaxation in the relaxation treatment was changed to 4.0 %.

Example 5

A release film was obtained in the same manner as in Example 4 except that the conditions in the corresponding step were changed as follows. The thus-

obtained release film was subjected to various measurements and evaluations. Table 2 shows the results.

[Preparation of coating agent]

As a surfactant, 0.15 part of a polyoxyethylene-methylpolysiloxane copolymer (trade name: EMALEX SS-5051, supplied by Nihon Emulsion Co., Ltd.) (S2 component) that was a nonionic silicone-containing surfactant was used. The coating agent had a solid content of 6.0 mass%. Further, the solid content ratios of components based on 100 mass% of a release layer to be obtained from the above coating agent were as follows.

Main component: 84.1 mass%

Crosslinking agent: 8.4 mass%

Silane coupling agent: 5.0 mass%

Surfactant: 2.5 mass%

Example 6

A release film was obtained in the same manner as in Example 4 except that the conditions in the corresponding step were changed as follows. The thus-obtained release film was subjected to various measurements and evaluations. Table 2 shows the results.

[Preparation of coating agent]

As a surfactant, 0.3 part of a polyoxyethylene-methylpolysiloxane copolymer (trade name: EMALEX SS-5051, supplied by Nihon Emulsion Co., Ltd.) (S2 component) that was a nonionic silicone-containing surfactant was used. The coating agent had a solid content of 6.2 mass%. Further, the solid content ratios of components based on 100 mass% of a release layer obtained from the above coating agent were as follows.

Main component: 82.0 mass%

Crosslinking agent: 8.2 mass%

Silane coupling agent: 4.9 mass%

Surfactant: 4.9 mass%

Example 7

A release film was obtained in the same manner as in Example 4 except that the conditions in the corresponding step were changed as follows. The thus-obtained release film was subjected to various measurements and evaluations. Table 2 shows the results.

[Preparation of coating agent]

As a surfactant, 0.6 part of a polyoxyethylene-methylpolysiloxane copolymer (trade name: EMALEX SS-5051, supplied by Nihon Emulsion Co., Ltd.) (S2 component) that was a nonionic silicone-containing surfactant was used. The coating agent had a solid content of 6.5 mass%. Further, the solid content ratios of components based on 100 mass% of a release layer to be obtained from the above coating agent were as follows.

Main component: 78.1 mass%

Crosslinking agent: 7.8 mass%

Silane coupling agent: 4.7 mass%

Surfactant: 9.4 mass%

Comparative Example 4

A release film was obtained in the same manner as in Example 4 except that the conditions in the corresponding steps were changed as follows. The thus-obtained release film was subjected to various measurements and evaluations. Table 2 shows the results.

[Preparation of coating agent]

No surfactant was used. The coating agent had a solid content of 5.9 mass%. Further, the solid content ratios of components based on 100 mass% of a release

layer obtained from the above coating agent were as follows.

Main component: 86.2 mass%

Crosslinking agent: 8.6 mass%

Silane coupling agent: 5.2 mass%

Surfactant: 0 mass%

[Primary stretching step]

The stretch ratio in the longitudinal direction was changed to 3.0 times.

Comparative Example 5

A release film was obtained in the same manner as in Comparative Example 4 except that the conditions in the corresponding steps were changed as follows. The thus-obtained release film was subjected to various measurements and evaluations. Table 2 shows the results.

[Primary stretching step]

The stretch ratio in the longitudinal direction was changed to 4.8 times.

[Secondary stretching step]

The stretch ratio in the lateral direction was changed to 3.0 times.

Comparative Example 6

A biaxially stretched polyester film having no release layer was obtained in the same manner as in Example 3 except that no coating agent was applied.

Then, to a solution of an addition polymerization type silicone compound (trade name: TPR-6721, supplied by Toshiba Silicone K.K.) in toluene (solid content: 3 mass%) was added a Pt catalyst (trade name: CM670, supplied by Toshiba Silicone K.K.) in such an amount that its content came to be 1 part by mass per 100 parts by mass of the addition polymerization type silicone

compound, whereby a release agent coating liquid was prepared. This release agent coating liquid contained no surfactant.

Thereafter, the above-obtained biaxially stretched polyester film having no release layer was unrolled, and the above-prepared release agent coating liquid was applied to the unrolled biaxially stretched polyester film in its center portion in the width direction such that the application amount (wet) came to be 6 g/m^2. A release layer was formed by drying the applied coating liquid with an air floating and carrying type drying apparatus having lower and upper air-blowing ports at intervals of 38 cm each at a carrying tension of 2,000 kPa at a drying temperature of 160°C for 16 seconds, to give a release film having a dry cured release layer having a weight of 0.2 g/m^2. Table 2 shows the results of various measurements and evaluations made with regard to the thus-obtained release film. The above application of the coating agent was carried out on the side of the polyester film which had not contacted with the cooling drum in the step of forming an unstretched polyester film.

Table 2

Measurement and evaluation			Unit	Example 3	Example 4	Example 5	Example 6	Example 7
Surfactant contained in release layer		Kind	-	S1	S2	S2	S2	S2
		Content	mass%	2.5	1.0	2.5	4.9	9.4
Properties of release film	Elongation ratio (S_{MD}) in longitudinal direction	Load 0.3MPa	%	-0.31	-0.28	-0.28	-0.28	-0.28
		Load 1.0 MPa	%	-0.24	-0.19	-0.19	-0.19	-0.19
		Load 2.5 MPa	%	-0.10	-0.08	-0.08	-0.08	-0.08
	Elongation ratio (S_{TD}) in width direction	Load 0.01 MPa	%	-0.39	-0.43	-0.43	-0.43	-0.43
	Thermal elongation ratio (HS_{MD}) In longitudinal direction	No load	%	-0.20	-0.18	-0.18	-0.18	-0.18
	Thermal elongation ratio (HS_{TD}) In width direction	No load	%	-0.40	-0.50	-0.50	-0.50	-0.50
	HS_{MD}-HS_{TD}	—	%	0.20	0.32	0.32	0.32	0.32
	Maximum height (Rmax)	Release layer surface	nm	450	448	448	448	448
		Surface on side having no release layer	nm	460	455	455	455	455
Evaluation of peel		—	—	◎	◎	◎	◎	○
Evaluation of peel charge		Release film	kV	1.0	1.5	1.2	0.9	0.8
		Ceramic sheet	kV	10	14	11	9	8
Evaluations of ceramic sheet (Alternative evaluation for practical use)		Surface smoothness (Condition 2)	—	○	○	○	○	○
		Evaluation 2 of stacking (Positional deviation evaluation)	—	◎	○	◎	◎	◎

Table 2 (continued)

Measurement and evaluation			Unit	C.Ex. 4	C.Ex.5	C.Ex.6
Surfactant contained in release layer		Kind	—	None	None	None
		Content	mass%	—	—	—
Properties of release film	Elongation ratio (S_{MD}) in longitudinal direction	Load0.3MPa	%	-0.20	-0.48	-0.05
		Load1.0MPa	%	-0.14	-0.38	0.02
		Load2.5MPa	%	0.05	-0.30	0.13
	Elongation ratio (D_{TD}) in width direction	Load0.01MPa	%	-0.62	-0.10	0.00
	Thermal elongation ratio (HS_{MD}) in longitudinal direction	No load	%	-0.10	-0.50	-0.05
	Thermal elongation ratio (HS_{TD}) in width direction	No load	%	-0.70	-0.10	0.00
	HS_{MD}-HS_{TDD}	—	%	0.60	-0.40	-0.05
	Maximum height (Rmax)	Release layer surface	nm	440	455	452
		Surface on side having no release layer	nm	450	465	464
Evaluation of peel		—	—	◎	◎	◎
Evaluation of peel charge		Release film	kV	7.5	7.8	8
		Ceramic sheet	kV	25	23	24
Evaluations of ceramic sheet (Alternative evaluation for practical use)		Surface smoothness (Condition 2)	—	○	○	○
		Evaluation 2 of stacking (Positional deviation evaluation)	—	×	×	×

C.Ex.: Comparative Example

Example 8

[Preparation of polyester]

A polyethylene terephthalate composition having an intrinsic viscosity of 0.65 (in o-chlorophenol at 35°C) was obtained in the same manner as in Example 1.

[Preparation of coating agent]

A coating agent was obtained in the same manner as in Example 3. The coating agent had a solid content of 6.0 mass%. Further, the solid content ratios of components constituting 100 mass% of a release layer obtained from the above coating agent were as follows.

Main component: 84.1 mass%

Crosslinking agent: 8.4 mass%

Silane coupling agent: 5.0 mass%

Surfactant: 2.5 mass%

[Step of forming unstretched polyester film]

An unstretched polyester film having a thickness of 450 μm was obtained from the above-obtained polyethylene terephthalate composition in the same manner as in Example 3.

[Primary stretching step]

A monoaxially stretched polyester film stretched in the longitudinal direction was obtained from the thus-obtained unstretched polyester film in the same manner as in Example 1.

[In-line application step]

Then, the above-prepared coating agent was applied to the thus-obtained monoaxially stretched film stretched in the longitudinal direction such that the resultant release film had a release layer having a thickness of 40 nm, to give a polyester film having a coating film. The above application of the coating agent was carried out on the side of the polyester film which had not contacted with the cooling drum in the step of

forming an unstretched polyester film.

[Secondary stretching step]

Thereafter, the thus-obtained polyester film having a coating film was supplied to a stenter, and treated in the same manner as in Example 1 to give a biaxially stretched polyester film.

[Thermally setting step]

The thus-obtained biaxially stretched film was treated in the same manner as in Example 1 to give a release film having a total thickness of 31 μm. The thus-obtained release film was used as a pre-slitting release film and subjected to various measurements and evaluations. Table 3 shows the results.

[Slitting step]

As take-up conditions, an initial tension of 47 N/m, a tension taper ratio of 60 % (constant), a nip pressure of 150 N/m, a nip pressure taper ratio of 100 % and a speed of 180 m/minute were set, and a roll of a release film having a width of 450 mm and a length of 2,000 m was obtained. The thus-obtained release film roll was subjected to various measurements and evaluations, and Table 3 shows the results.

Further, a release film was sampled in a position 500 m away from the surface layer of the thus-obtained release film roll. The thus-obtained release film was used as a post-slitting release film and subjected to various measurements and evaluations, and Table 3 shows the results.

Example 9

A pre-slitting release film, a release film roll and a post-slitting release film were obtained in the same manner as in Example 8 except that the conditions in the steps were changed as follows. These were

subjected to various measurements and evaluations, and Table 3 shows the results.

[Preparation of coating agent]

No surfactant was used. The coating agent had a solid content of 5.9 mass%. Further, the solid content ratios of components based on 100 mass% of a release layer obtained from the above coating agent were as follows.

Main component: 86.2 mass%

Crosslinking agent: 8.6 mass%

Silane coupling agent: 5.2 mass%

Surfactant: 0 mass%

[Secondary stretching step]

The stretch ratio in the lateral direction was changed to 4.5 times.

[Thermally setting step]

The amount of relaxation in the relaxation treatment was changed to 4.0 %.

Example 10

A pre-slitting release film, a release film roll and a post-slitting release film were obtained in the same manner as in Example 9 except that the conditions in the corresponding step were changed as follows. These were subjected to various measurements and evaluations, and Table 3 shows the results.

[Preparation of coating agent]

As a surfactant, 0.06 part of a polyoxyethylene-methylpolysiloxane copolymer (trade name: EMALEX SS-5051, supplied by Nihon Emulsion Co., Ltd.) (S2 component) that was a nonionic silicone-containing surfactant was used. The coating agent had a solid content of 6.0 mass%. Further, the solid content ratios of components based on 100 mass% of a release layer

obtained from the above coating agent were as follows.

Main component: 85.4 mass%

Crosslinking agent: 8.5 mass%

Silane coupling agent: 5.1 mass%

Surfactant: 1.0 mass%

Example 11

A pre-slitting release film, a release film roll and a post-slitting release film were obtained in the same manner as in Example 10 except that the conditions in the corresponding steps were changed as follows. These were subjected to various measurements and evaluations, and Table 3 shows the results.

[Preparation of coating agent]

The amount of the surfactant (S2 component) to be added was changed to 0.15 part. The coating agent had a solid content of 6.0 mass%. Further, the solid content ratios of components based on 100 mass% of a release layer obtained from the above coating agent were as follows.

Main component: 84.1 mass%

Crosslinking agent: 8.4 mass%

Silane coupling agent: 5.0 mass%

Surfactant: 2.5 mass%

[Slitting step]

The take-up conditions in the slitting step were changed as shown in Table 3.

Examples 12 to 15

Pre-slitting release films, release film rolls and post-slitting release films were obtained in the same manner as in Example 11 except that the take-up conditions in the slitting steps were changed as shown in Table 3. These were subjected to various measurements

and evaluations, and Table 3 shows the results.

Example 16

A pre-slitting release film, a release film roll and a post-slitting release film were obtained in the same manner as in Example 10 except that the conditions in the corresponding step were changed as follows. These were subjected to various measurements and evaluations, and Table 3 shows the results.

[Preparation of coating agent]

The amount of the surfactant (S2 component) to be added was changed to 0.3 part. The coating agent had a solid content of 6.2 mass%. Further, the solid content ratios of components based on 100 mass% of a release layer obtained from the above coating agent were as follows.

Main component: 82.0 mass%

Crosslinking agent: 8.2 mass%

Silane coupling agent: 4.9 mass%

Surfactant: 4.9 mass%

Example 17

A pre-slitting release film, a release film roll and a post-slitting release film were obtained in the same manner as in Example 10 except that the conditions in the corresponding step were changed as follows. These were subjected to various measurements and evaluations, and Table 3 shows the results.

[Preparation of coating agent]

The amount of the surfactant (S2 component) added was changed to 0.6 part. The coating agent had a solid content of 6.5 mass%. Further, the solid content ratios of components based on 100 mass% of a release layer obtained from the above coating agent were as follows.

Main component: 78.1 mass%

Crosslinking agent: 7.8 mass%

Silane coupling agent: 4.7 mass%

Surfactant: 9.4 mass%

Comparative Example 7

A pre-slitting release film, a release film roll and a post-slitting release film were obtained in the same manner as in Example 9 except that the conditions in the corresponding step were changed as follows. These were subjected to various measurements and evaluations, and Table 3 shows the results.

[Primary stretching step]

The stretch ratio in the longitudinal direction was changed to 3.0 times.

Comparative Example 8

A pre-slitting release film, a release film roll and a post-slitting release film were obtained in the same manner as in Example 9 except that the conditions in the corresponding steps were changed as follows. These were subjected to various measurements and evaluations, and Table 3 shows the results.

[Primary stretching step]

The stretch ratio in the longitudinal direction was changed to 4.8 times.

[Secondary stretching step]

The stretch ratio in the lateral direction was changed to 3.0 times.

Comparative Example 9

A biaxially stretched film having no release layer was obtained in the same manner as in Example 8 except that no coating agent was applied.

Then, to a solution of an addition polymerization type silicone compound (trade name: TPR-6721, supplied by Toshiba Silicone K.K.) in toluene (solid content: 3 mass%) was added a Pt catalyst (trade name: CM670, supplied by Toshiba Silicone K.K.) in such an amount that its content came to be 1 part by mass per 100 parts by mass of the addition polymerization type silicone compound, whereby a release agent coating liquid was prepared. This release agent coating liquid contained no surfactant.

Thereafter, the above-obtained biaxially stretched polyester film having no release layer was unrolled, and the above-prepared release agent coating liquid was applied to the unrolled biaxially stretched polyester film in its center portion in the width direction such that the application amount (wet) came to be 6 g/m^2. A release layer was formed by drying the applied coating liquid with an air floating and carrying type drying apparatus having lower and upper air-blowing ports at intervals of 38 cm each at a carrying tension of 2,000 kPa at a drying temperature of 160°C for 16 seconds, to give a release film having a dry cured release layer having a weight of 0.2 g/m^2. Table 3 shows the results of various measurements and evaluations made with regard to the thus-obtained release film as a pre-slitting film. The above application of the coating agent was carried out on that side of the polyester film which had not been brought into contact with the cooling drum in the step of forming an unstretched polyester film.

[Slitting step]

A release film roll and a post-slitting release film were obtained under the take-up conditions in the slitting step that were set as shown in Table 3. These were subjected to various measurements and evaluations,

and Table 3 shows the results.

Referential Example 1

A pre-slitting release film, a release film roll and a post-slitting release film were obtained in the same manner as in Example 11 except that the conditions in the slitting step were changed as shown in Table 3. These were subjected to various measurements and evaluations, and Table 3 shows the results.

Table 3

Items measured, evaluated			Unit	Ex.8	Ex.9	Ex.10	Ex.11	Ex.12	Ex.13	Ex.14
Release layer	Surfactant	Kind	-	S1	nil	S2	S2	S2	S2	S2
		Content	mass%	2.5	-	1.0	2.5	2.5	2.5	2.5
Pre-slitting release film	Elongation ratio in longitudinal direction (S_{MD})	Load 0.3MPa	%	-0.31	-0.28	-0.28	-0.28	-0.28	-0.28	-0.28
		Load 1.0MPa	%	-0.24	-0.19	-0.19	-0.19	-0.19	-0.19	-0.19
		Load 2.5MPa	%	-0.10	-0.08	-0.08	-0.08	-0.08	-0.08	-0.08
	Elongation ratio in width direction (S_{TD})	Load 0.01MPa	%	-0.39	-0.43	-0.43	-0.43	-0.43	-0.43	-0.43
	Thermal elongation ratio in longitudinal direction (HS_{MD})	No load	%	-0.20	-0.18	-0.18	-0.18	-0.18	-0.18	-0.18
	Thermal elongation ratio in width direction (HS_{TD})	No load	%	-0.40	-0.50	-0.50	-0.50	-0.50	-0.50	-0.50
	HS_{MD}-HS_{TD}	-	%	0.20	0.32	0.32	0.32	0.32	0.32	0.32
	Maximum height (Rmax)	Release layer surface	nm	450	448	448	448	448	448	448
		Surface on side having no release layer	nm	460	455	455	455	455	455	455
	Evaluation of peel	-	-	◎	◎	◎	◎	◎	◎	◎
	Evaluation of peel charge	Release film	kV	1.0	7.5	1.5	1.2	1.2	1.2	1.2
		Ceramic sheet	kV	10	25	14	11	11	11	11
	Evaluations of ceramic sheet (Alternative evaluation for practical use)	Surface smoothness (Condition 2)	-	○	○	○	○	○	○	○
		Evaluation 2 of stacking (Evaluation of positional deviation)	-	◎	-	○	◎	◎	◎	◎

Ex.: Example

Table 3 (continued)

Items measured, evaluated			Unit	Ex.8	Ex.9	Ex.10	Ex.11	Ex.12	Ex.13	Ex.14
Take-up conditions		Initial tension	N/m	47	47	47	47	47	40	40
		Tension taper ratio	%	60	60	60	80	60	40	60
		Final tension	N/m	28	28	28	38	28	16	24
		Initial nip pressure	N/m	150	150	150	180	150	100	100
		Nip pressure taper ratio	%	100	100	90	90	90	100	90
		Final nip pressure	N/m	150	150	135	162	135	100	90
Release film roll	Vicker's hardness (Hv) of roll surface layer	(Maximum value)	—	410	410	400	420	400	380	360
	Uneven end face	-		◎	◎	◎	◎	◎	◎	○
Post-slitting release film	Flatness	-		◎	◎	◎	○	◎	◎	◎

Ex.: Example

Table 3 (continued)

Items measured, evaluated			Unit	Ex.15	Ex.16	Ex.17	C.Ex.7	C.Ex.8	C.Ex.9	R.Ex.1
Release layer	Surfactant	Kind	-	S2	S2	S2	Nil	Nil	Nil	S2
		Content	mass%	2.5	4.9	9.4	-	-	-	2.5
Pre-slitting release film	Elongation ratio in longitudinal direction (S_{MD})	Load 0.3MPa	%	-0.28	-0.28	-0.28	-0.20	-0.48	-0.05	-0.28
		Load 1.0MPa	%	-0.19	-0.19	-0.19	-0.14	-0.38	0.02	-0.19
		Load 2.5MPa	%	-0.08	-0.08	-0.08	0.05	-0.30	0.13	-0.08
	Elongation ratio in width direction (S_{TD})	Load 0.01MPa	%	-0.43	-0.43	-0.43	-0.62	-0.10	0.00	-0.43
	Thermal elongation ratio in longitudinal direction (HS_{MD})	No load	%	-0.18	-0.18	-0.18	-0.10	-0.50	-0.05	-0.18
	Thermal elongation ratio in width direction (HS_{TD})	No load	%	-0.50	-0.50	-0.50	-0.70	-0.10	0.00	-0.50
	HS_{MD}-HS_{TD}	-	%	0.32	0.32	0.32	0.60	-0.40	-0.05	0.32
	Maximum height (Rmax)	Release layer surface	Nm	448	448	448	440	455	452	448
		Surface on side having no release layer	nm	455	455	455	450	465	464	455
	Evaluation of peel	-	-	◎	◎	○	◎	◎	◎	◎
	Evaluation of peel charge	Release film	kV	1.2	0.9	0.8	7.5	7.8	8	1.2
		Ceramic sheet	kV	11	9	8	25	23	24	11
	Evaluations of ceramic sheet (Alternative evaluation for practical use)	Surface smoothness (Condition 2)	-	○	○	○	○	○	○	○
		Evaluation 2 of stacking (Evaluation of positional deviation)	-	◎	◎	◎	×	×	×	◎

Ex.: Example C.Ex.: Comparative Example R.Ex.: Reference Example

Table 3 (continued)

Items measured, evaluated			Unit	Ex.15	Ex.16	Ex.17	C.Ex.7	C.Ex.8	C.Ex.9	R.Ex.1
Take-up conditions		Initial tension	N/m	30	47	47	47	47	47	55
		Tension taper ratio	%	40	60	60	60	60	60	40
		Final tension	N/m	12	28	28	28	28	28	22
		Initial nip pressure	N/m	100	150	150	150	150	150	180
		Nip pressure taper ratio	%	90	100	100	90	90	90	100
		Final nip pressure	N/m	90	150	150	135	135	135	180
Release film roll	Vicker's hardness (Hv) of roll surface layer	(Maximum value)	–	350	410	410	400	400	400	460
	Uneven end face	–		△	◎	◎	◎	◎	◎	◎
Post-slitting release film	Flatness	–		◎	◎	◎	◎	◎	◎	×

Ex.: Example C.Ex.: Comparative Example R.Ex.: Reference Example

Example 18

[Preparation of polyester]

A polyethylene terephthalate composition having an intrinsic viscosity of 0.65 (in o-chlorophenol at 35°C) was obtained in the same manner as in Example 1.

[Preparation of coating agent]

A coating agent was obtained in the same manner as in Example 3. The coating agent had a solid content of 6.0 mass%. Further, the solid content ratios of components based on 100 mass% of a release layer obtained from the above coating agent were as follows.

Main component: 84.1 mass%

Crosslinking agent: 8.4 mass%

Silane coupling agent: 5.0 mass%

Surfactant: 2.5 mass%

[Step of forming unstretched polyester film]

The above-obtained polyethylene terephthalate composition was dried at 170°C for 5 hours until it had a water content of 0.05 mass% or less. Then, the dry polyethylene terephthalate composition was fed to an extruder, melted at a melting temperature of 280 to 300°C, highly accurately filtered with a steel wire filter having an average mesh opening of 11 μm, and then extruded through a T-die to form a molten sheet, and the molten sheet was rapidly cooled by bringing it into contact with a cooling drum by an electrostatically contacting method to give an unstretched polyester film having a thickness of 480 μm.

[Primary stretching step]

The thus-obtained unstretched polyester film was pre-heated at 75°C, and then stretched 3.8 times in the longitudinal direction between low-speed and high-speed rolls at a film temperature of 105°C, and then the stretched film was rapidly cooled to give a polyester

film stretched in the longitudinal direction.

[In-line application step]

Then, the above-prepared coating agent was applied to the thus-obtained stretched polyester film stretched in the longitudinal direction, such that the release layer of a release film to be obtained was to have a thickness of 40 nm, to give a polyester film having a coating film. The above application of the coating agent was carried out on the side of the polyester film which had not been brought into contact with the cooling drum in the step of forming an unstretched polyester film.

[Secondary stretching step]

The polyester film having a thus-obtained coating film was supplied to a stenter and treated in the same manner as in Example 1 to obtain a biaxially stretched polyester film.

[Thermally setting step]

The thus-obtained biaxially stretched polyester film was treated in the same manner as in Example 1 to give a release film having a total thickness of 31 μm. The thus-obtained release film was subjected to various measurements and evaluations. Table 4 shows the results.

Example 19

A release film having a total thickness of 31 μm was obtained in the same manner as in Example 18 except that the conditions in the corresponding steps were changed as follows. The thus-obtained release film was subjected to various measurements and evaluations. Table 4 shows the results.

[Preparation of coating agent]

As a surfactant, 0.06 part of a polyoxyethylene-methylpolysiloxane copolymer (trade name: EMALEX SS-5051, supplied by Nihon Emulsion Co., Ltd.) (S2

component) that was a nonionic silicone-containing surfactant was used in place of the polyoxyethylene oleyl ether. The coating agent had a solid content of 6.0 mass%. Further, the solid content ratios of components based on 100 mass% of a release layer obtained from the above coating agent were as follows.

Main component: 84.1 mass%

Crosslinking agent: 8.4 mass%

Silane coupling agent: 5.0 mass%

Surfactant: 2.5 mass%

[Primary stretching step]

The stretch ratio in the longitudinal direction was changed to 4.0 times.

[Thermally setting step]

The value of relaxing treatment was changed to 4.0 %.

Table 4

Items measured, evaluated			Unit	Example 18	Example 19
Surfactant contained in release layer		Kind	-	S1	S2
		Content	mass%	2.5	2.5
Stretch ratio		Longitudinal direction	times	3.8	4.0
		Width direction	times	4.1	4.1
		Plane	times	15.6	16.4
Film thickness		—	μm	31	31
Properties of release film	Elongation ratio in longitudinal direction (S_{MD})	Load 0.3MPa	%	-0.33	-0.35
		Load 1.0MPa	%	-0.26	-0.29
		Load 2.5MPa	%	-0.12	-0.15
	Elongation ratio in width direction (S_{TD})	Load 0.01MPa	%	-0.38	-0.37
	Thermal elongation ratio in longitudinal direction (HS_{MD})	No load	%	-0.23	-0.25
	Thermal elongation ratio in width direction (HS_{TD})	No load	%	-0.40	-0.39
	$HS_{MD}-HS_{TD}$	—	%	0.17	0.14
	Thickness non-uniformity	Longitudinal direction	%	2.9	1.9
		Width direction	%	2.5	2.5
	Maximum height (Rmax)	Release layer surface	Nm	452	454
		Surface on side having no release layer	Nm	458	460
	Evaluation of peel	—	—	◎	◎
	Evaluation of peel charge	Release film	—	○	○
		Ceramic sheet	—	○	○
	Evaluation of ceramic sheet (Alternative evaluation for practical use)	Surface smoothness (Condition 2)	—	○	○
		Thickness non-uniformity	—	◎	◎

Effect of the Invention

The release film of this invention has a proper dimensional change ratio under tension under heat when a ceramic sheet is produced, and it is excellent in thermal shrinkage balance when applied ceramic slurry is dried. That is, it fully satisfies the performances required for a release film that is used in the production of a ceramic sheet. When the release film of this invention is used as a carrier film in the production of a ceramic sheet, therefore, it is excellent in a thermal shrinkage balance as a carrier film not only in a carrying step but also in a drying step, so that the thickness non-uniformity of a ceramic sheet to be obtained can be suppressed to a high degree.

Further, the release film having a preferred embodiment in this invention is excellent in surface smoothness, the occurrence of a pin hole in a ceramic sheet to be obtained can be suppressed. According to the release film having the preferred embodiment in this invention, therefore, the productivity of a ceramic sheet and a ceramic condenser can be improved.

In the release film having a preferred embodiment in this invention, further, it is possible to highly suppress a peel charge that occurs in the step of unrolling a carrier film or the step of separating a ceramic sheet from a carrier film. As a result, when a ceramic sheet produced from the release film of this invention is used to produce a ceramic condenser, the positional deviation of an internal electrode of the condenser to be obtained can be suppressed to a high degree.

The film roll of this invention can provide a release film excellent in flatness. When a ceramic sheet produced from the above release film is used to produce

a ceramic condenser, there can be obtained ceramic condensers having uniform capacity.

CLAIMS

1. A release film having a release layer on at least one surface of a polyester film, wherein the release layer contains 0.5 mass% or more but 10 mass% or less, based on weight of the release layer, of a surfactant,

wherein:

the elongation ratio (S_{MD}) of the release film in the longitudinal direction at 100°C when a tension of 0.2 MPa or more but 4.0 MPa or less is exerted on the release film in the longitudinal direction of the release film satisfies the following expression (1),

the elongation ratio (S_{TD}) of the release film in the direction perpendicular to the longitudinal direction at 100°C when a tension of 0.01 MPa is exerted on the release film in the direction perpendicular to the longitudinal direction of the release film satisfies the following expression (2),

the thermal elongation ratio (HS_{MD}) of the release film in the longitudinal direction at 100°C when no load is exerted on the release film satisfies the following expression (3),

the thermal elongation ratio (HS_{TD}) of the release film in the direction perpendicular to the longitudinal direction at 100°C when no load is exerted on the release film satisfies the following expression (4), and

the thermal elongation ratio (HS_{MD}) in the longitudinal direction and the thermal elongation ratio (HS_{TD}) in the direction perpendicular to the longitudinal direction satisfy the following expression (5),

$$0.0961X - 0.45 \leq S_{MD} \leq 0.0961X - 0.25 \quad (1)$$

wherein X is a tension (MPa) exerted on a film unit area, and X is a value of 0.2 MPa or more but 4.0 MPa or less,

$$-0.6 \leq S_{TD} \leq -0.2 \quad (2)$$

$$-0.4 \leq HS_{MD} \leq -0.1 \quad (3)$$

$$-0.6 \leq HS_{TD} \leq -0.2 \quad (4)$$

$$HS_{MD} > HS_{TD} \quad (5) .$$

2. The release film of claim 1, which has a release layer whose surface has a maximum height (Rmax) of 100 nm or more but 600 nm or less when measured with a stylus three-dimensional surface roughness meter.

3. The release film of claim 1 or 2, wherein each of the surface of the release layer and a surface on the side having no release layer has a maximum height (Rmax) of 100 nm or more but 600 nm or less when measured with a stylus three-dimensional surface roughness meter.

4. The release film of any one of claim 1 to 3, which has a thickness non-uniformity of 3.0 % or less in the longitudinal direction and a thickness non-uniformity of 3.0 % or less in the lateral direction.

5. The release film of any one of claims 1 to 4, wherein the release layer is a layer formed by applying a release-layer-forming composition to a monoaxially stretched polyester film.

6. The release film of any one of claims 1 to 5, which is used for producing a ceramic sheet.

7. The release film of claim 6, wherein the ceramic sheet is used for producing a ceramic condenser.

8. A film roll that is a film roll obtained by taking up the release film of any one of claims 1 to 5 in the form of a roll, the roll having a surface layer having a Vicker's hardness (Hv) of 0 or more but 450 or less.

ABSTRACT

This invention is to provide a release film that has a dimensional change ratio suitable under tension under heat in the production of a ceramic sheet, that is excellent in thermal shrinkage balance in the drying of ceramic slurry, and that fully satisfies the performances required for a release film used in the production of a ceramic sheet.

This invention is a release film having specified elongation ratios under specific loads and having specified thermal elongation ratios under no load.