# Trial Decision

# Invalidation No. 2012-800199

Yamaguchi, Japan	
Demandant	UBE INDUSTRIES LTD.
Tokyo, Japan	
Patent Attorney	ITO, Katsuhiro
Tokyo, Japan	
Patent Attorney	ONO, Akiko
Tokyo, Japan	
Attorney	OZAKI, Hideo
Tokwo Japan	
1 okyo, Japan	
Auomey	HINO, EIICHIKO
Tokyo, Japan	
Attorney	UENO, JUNICHI
Tokyo, Japan	
Attorney	IMADA, HITOMI
Tokvo, Japan	
Demandee	DU PONT-TORAY COMPANY LTD.
Tokyo, Japan	
Attorney	MASUI, Kazuo
Teluce Japan	
Tokyo, Japan	HASHICICHI Neovalti
Auomey	HASHIOUCIII, IvaUyuki
Tokyo, Japan	
Attorney	SAITO, Seijiro

The patent invalidation trial case regarding Patent No. 4777471 entitled "POLYIMIDE FILM AND COPPER CLAD LAMINATE USING THE FILM AS BASE MATERIAL" between the parties above has resulted in the following trial decision:

#### Conclusion

The trial of the case was groundless.

The costs in connection with the trial shall be borne by Demandant.

#### Reason

No. 1 History of the procedures

The patent according to the trial (hereinafter referred to as "the Patent") was registered on July 8, 2011 (Patent No. 4777471, the title of the invention: "polyimide film and copper clad laminate using the film as base material", the number of the claims: 11). Hereinafter the specification of the Patent is referred to as "the patent specification." The Patent is based on a divisional application filed on August 11, 2010 (Patent Application No. 2010-180128 (hereinafter referred to as "the present application")), which was originally filed on March 25, 2005 as Patent Application No. 2005-88334 (claiming priority benefit of March 30, 2004).

Further, Ube Industries, Ltd. (hereinafter referred to as "Demandant") made a demand for trial to invalidate the Patent.

History of procedure in the Trial of the case is as follows.

November 30, 2012	Written demand for trial, Evidence A No. 1 to A No. 6
	submitted
February 15, 2013	Written reply of trial case, Evidence B No. 1 to B No. 5
	submitted
March 13, 2013	Notification of matters to be examined
April 10, 2013	Oral proceedings statement brief (Demandant) Evidence
	A No. 7 to A No. 18 submitted
April 10, 2013	Oral proceedings statement brief (Demandee) Evidence
	B No. 6 to B No. 7 submitted

April 22, 2013	Written statement (Demandee) Evidence B No. 8 to B
	No. 10 submitted
April 24, 2013	Oral proceeding
May 24, 2013	Written statement (Demandant) submitted
May 24, 2013	Written statement (Demandee) submitted, Evidence B
	No. 11 and Reference Documents 1 and 2 submitted

No. 2 The inventions relating to the Patent

The inventions relating to the Patent are as recited in the following Claims 1 to 11 of the Claims (hereinafter referred to as "the patent invention 1" to "the patent invention 11," and also collectively referred to as "the patent invention").

#### "[Claim 1]

A substrate for COF having a copper clad laminate comprising: a base material of a polyimide film into which a fine silica is uniformly dispersed; and a copper formed thereon with a thickness of 1 to 10  $\mu$ m, wherein the polyimide film is produced by using one or more aromatic diamine components selected from the group consisting of paraphenylenediamine, 4,4'-diaminodiphenyl ether, and 3,4'-diaminodiphenyl ether, and one or more acidic anhydride components selected from the group consisting of pyromellitic acid dianhydride and 3,3'-4,4'-diphenyltetracarboxylic acid dianhydride, wherein said polyimide film comprises the fine silica with a particle diameter of 0.07 to 2.0  $\mu$ m, and wherein a thermal expansion coefficient  $\alpha$ MD in a machine direction (MD) of the film measured at a temperature range of 50 to 200°C and a heat elevation rate of 10°C/min by TMA-50 manufactured by Shimadzu Corporation falls within a range of 10 ppm/°C or more to 20 ppm/°C or less, and wherein a thermal expansion coefficient  $\alpha$ TD in a traverse direction (TD) measured under said conditions falls within a range of 3 ppm/°C or more to 7 ppm/°C or less. [Claim 2]

The substrate for COF of Claim 1, wherein a thermal expansion coefficient  $\alpha$ MD in a machine direction (MD) of film measured at a temperature range of 50 to 200°C and a heat elevation rate of 10°C/min by TMA-50 manufactured by Shimadzu Corporation falls within a range of 14 ppm/°C or more to 18 ppm/°C or less, and wherein a thermal expansion coefficient  $\alpha$ TD in a traverse direction (TD) measured under said conditions falls within a range of 3 ppm/°C or more to 7 ppm/°C or less. [Claim 3]

The substrate for COF of Claim 1 or Claim 2, wherein said fine silica is uniformly dispersed into the film in a proportion of 0.03 to 0.30 weight% on the film resin weight basis, and fine protrusions are formed on a surface. [Claim 4]

The substrate for COF of Claim 3, wherein said fine silica has an average particle size of 0.10  $\mu m$  or more to 0.90  $\mu m$  or less.

# [Claim 5]

The substrate for COF of Claim 3, wherein said fine silica has an average particle size of 0.10  $\mu$ m or more to 0.30  $\mu$ m or less. [Claim 6]

The substrate for COF of any of Claim 3 to Claim 5, wherein the number of the protrusions formed by said fine silica is from  $1*10^3$  to  $1*10^8$  per 1 mm<sup>2</sup>. [Claim 7]

A copper clad laminate comprising: a base material of a polyimide film into which a fine silica is uniformly dispersed; and a copper formed thereon with a thickness of 1 to 10  $\mu$ m, wherein the polyimide film is produced by using one or more selected aromatic diamine components from the group consisting of paraphenylenediamine, 4,4'-diaminodiphenyl ether, and 3,4'-diaminodiphenyl ether, and one or more acidic anhydride components selected from the group consisting of pyromellitic acid dianhydride and 3,3'-4,4'-diphenyltetracarboxylic acid dianhydride, wherein said polyimide film comprises the fine silica with a particle diameter of 0.07 to 2.0  $\mu$ m, and wherein a thermal expansion coefficient  $\alpha$ MD in a machine direction (MD) of film measured at a temperature range of 50 to 200°C and a heat elevation rate of 10°C/min by TMA-50 manufactured by Shimadzu Corporation falls within a range of 10 ppm/°C or more to 20 ppm/°C or less, and wherein a thermal expansion coefficient aTD in a traverse direction (TD) measured under said conditions falls within a range of 3 ppm/°C or more to 7 ppm/°C or less. [Claim 8]

The copper clad laminate of Claim 7, wherein a thermal expansion coefficient  $\alpha$ MD in a machine direction (MD) of film measured at a temperature range of 50 to 200°C and a heat elevation rate of 10°C/min by TMA-50 manufactured by Shimadzu Corporation falls within a range of 14 ppm/°C or more to 18 ppm/°C or less, and wherein a thermal expansion coefficient  $\alpha$ TD in a traverse direction (TD) measured under said conditions falls within a range of 3 ppm/°C or more to 7 ppm/°C or less. [Claim 9]

A polyimide film into which a fine silica is uniformly dispersed, wherein the

polyimide film is produced by using one or more aromatic diamine components selected from the group consisting of paraphenylenediamine, 4,4'-diaminodiphenyl ether, and 3,4'-diaminodiphenyl ether, and one or more acidic anhydride components selected from the group consisting of pyromellitic acid dianhydride and 3,3'-4,4'- diphenyltetracarboxylic acid dianhydride, wherein said polyimide film comprises the fine silica with a particle diameter of 0.07 to 2.0  $\mu$ m, and wherein a thermal expansion coefficient  $\alpha$ MD in a machine direction (MD) of film measured at a temperature range of 50 to 200°C and a heat elevation rate of 10°C/min by TMA-50 manufactured by Shimadzu Corporation falls within a range of 10 ppm/°C or more to 20 ppm/°C or less, and wherein a thermal expansion coefficient  $\alpha$ TD in a traverse direction (TD) measured under said conditions falls within a range of 3 ppm/°C or more to 7 ppm/°C or less.

# [Claim 10]

A method of producing a substrate for COF according to any one of Claims 1 to 6, comprising the steps of: subjecting a film to mechanical direction stretching with a draw ratio (MDX) of 1.05 to 1.6 times at a temperature of 140°C or less, and subjecting to traverse direction stretching with a draw ratio (TDX) of 1.1 to 1.5 times the draw ratio of said mechanical direction stretching, wherein said film is obtained by using one or more aromatic diamine components selected from the group consisting of paraphenylenediamine, 4,4'-diaminodiphenyl ether, and 3,4'-diaminodiphenyl ether, and one or more acidic anhydride components selected from the group consisting of pyromellitic acid dianhydride and 3,3'-4,4'-diphenyltetracarboxylic acid dianhydride. [Claim 11]

A method of producing a polyimide film of Claim 9, comprising the steps of: subjecting a film to mechanical direction stretching with a draw ratio (MDX) of 1.05 to 1.6 times at a temperature of 140°C or less, and subjecting to traverse direction stretching with a draw ratio (TDX) of 1.1 to 1.5 times the draw ratio of said mechanical direction stretching, wherein said film is obtained by using one or more aromatic diamine components selected from the group consisting of paraphenylenediamine, 4,4'-diaminodiphenyl ether, and 3,4'-diaminodiphenyl ether, and one or more acidic anhydride components selected from the group consisting of pyromellitic acid dianhydride and 3,3'-4,4'-diphenyltetracarboxylic acid dianhydride."

3 The outline of the reasons for invalidation alleged by Demandant and the means of proof submitted by Demandant

#### 1. Gist of reasons for invalidation

Demandant claims for a decision to the effect that "Patents regarding the inventions according to Claims 1 to 11 of Japanese Patent No. 4777471 shall be invalidated, and the costs in connection with the trial shall be borne by Demandee," and argues based on the following Reasons for invalidation.

### [Reasons for invalidation 1]

The Detailed Description of the Invention of the patent specification fails to disclose definitely and sufficiently to the extent that allows a person skilled in the art to implement the invention directed to a polyimide film other than four-component polyimide film disclosed in the examples, e.g. two-component polyimide film, whereas the recitation of the claims encompasses such two-component polyimide film.

Therefore, with respect to the patent inventions 1 to 11, the Detailed Description of the Invention fails to conform to the requirement of Article 36(4)(i) of the Patent Act, and the recitation of the Claims according to the patent inventions 1 to 11 fails to conform to Article 36(6)(i) of the Patent Act; i.e., Article 36(6) of the Patent Act. Therefore, the patents should be invalidated under the provision of Article 123(1)(iv) of the Patent Act.

#### [Reasons for invalidation 2]

It cannot be seen from the Detailed Description of the Invention of the patent specification as to whether or not a substrate for COF according to the patent invention 1 may be obtained by a method of producing a substrate for COF according to the patent invention 10, nor whether or not a polyimide film according to the patent invention 9 may be obtained by a method of producing a polyimide film according to the patent invention 11. Therefore, it fails to disclose definitely and sufficiently to the extent that allows a person skilled in the art to implement the patent inventions 10 and 11.

Therefore, the patent specification fails to conform to the requirement of Article 36(4)(i) of the Patent Act, and the recitation of the Claims according to the patent inventions 10 and 11 fails to conform to Article 36(6)(i) of the Patent Act; i.e., Article 36(6) of the Patent Act. Therefore, the patents should be invalidated under the provision of Article 123(1)(iv) of the Patent Act.

In addition, Demandant has withdrawn the Reasons for invalidation under the

provision of Article 17-2(3) and Article 29(1)(iii) of the Patent Act in the oral proceeding.

2. Means of proof submitted by Demandant

Demandant submitted the following means of proof.

[Means of Proof]

Evidence A No. 1:	Japanese Unexamined Patent Application Publication No. 2001-72781
Evidence A No. 2:	Japanese Unexamined Patent Application Publication No. 2004-122372
Evidence A No. 3:	Notice of reasons for refusal dated July 12, 2012 of Japanese Patent Application No. 2005-88334 (the patent application of the patent application)
Evidence A No. 4:	Japanese Unexamined Patent Application Publication No. 2003-335874
Evidence A No. 5:	Explanation of circumstances concerning accelerated examination on December 10, 2010 in the prosecution of the Patent
Evidence A No. 6:	Japanese Unexamined Patent Application Publication No. 2005-314669 (a publication of unexamined application of the patent application of the Patent)
Evidence A No. 7:	POLYMER, 1989, Vol. 30, June (Conference issue page (only a part of abstract)) pages 1170 to 1173
Evidence A No. 8:	<ul> <li>Planned and Edited by Kotaro Senba, "Top-edge 'materials' and 'technology' revealed by top companies' practicians,</li> <li>Complete collection of latest electronics mounting <first volume="">", page 136 (published date: June 29, 2007, published by technical information institute Co., Ltd)</first></li> </ul>
Evidence A No. 9:	Thin Solid Films 339 (1999), pages 68 to 73 (only a part of abstract)
Evidence A No. 10:	POLYMER, 1987, Vol. 28, December, pages 2282 to 2288 (only a part of abstract)

Evidence A No. 11: Macromolecules, 1996, 29, pages 7897 to 7909 (only a part of

abstract)

Evidence A No. 12:	Mat. Res. Soc. Symp. Proc. Vol. 381 1995, pages 19 to 29			
	(only a part of abstract)			
Evidence A No. 13:	Japanese Unexamined Patent Application Publication No. H5- 237928			
Evidence A No. 14:	Japanese Unexamined Patent Application Publication No. 2004-255845			
Evidence A No. 15:	Japanese Unexamined Patent Application Publication No. 2003-109989			
Evidence A No. 16:	Japanese Unexamined Patent Application Publication No. H11-80390			
Evidence A No. 17:	KANEKA CORPORATION APICAL catalog (published date unknown)			
Evidence A No. 18:	DU PONT-TORAY CO., LTD. "Capton" catalog (published			
	date: October 1999)			

No. 4 The Demandee's allegation

1. Outline of the Demandee's allegation

Demandee claimed in a written reply for a trial decision to the effect that the demand for trial should be rejected and the cost for trial should be borne by Demandant with an argument that none of the Reasons for invalidation 1 to 4 argued by Demandant has a point.

Means of proof submitted by Demandee
 Demandee submitted the following means of proof and reference documents.

[Means of Proof]

- Evidence B No. 1:Low-thermal expansion polyimide film Capton 150EN-A (A<br/>catalog of a Demandee's product as an implemented product<br/>of the patent invention)Evidence B No. 2:"The latent trend of experime rederined with the latent trend of experimental product
- Evidence B No. 2: "The latest trend of growing polyimide II" Cover page, Contents, pages 1 to 8, and 17 to 20, colophon (published on December 2000 by S.B. TECHNO RESEARCH CO., LTD.)

Evidence B No. 3: Polyimide film "Upirex-S" manufactured by Ube Industries, Ltd. (a printout of Demandant's website introducing Upirex-S, URL:

http://www.ube-ind.co.jp/japanese/products/fine/\_01\_01.htm)

- Evidence B No. 4: Analysis result report on August 31, 2012, prepared by Koichi
   Sawazaki, Product Technology Director, Technical
   Development Department of Capton, DU PONT-TORAY
   CO., LTD. (Analysis result of heat expansion coefficient of
   conventional product of Upirex S series)
- Evidence B No. 5: Analysis result report on March 30, 2012, prepared by Koichi
   Sawazaki, Product Technology Director, Technical
   Development Department of Capton, DU PONT-TORAY
   CO., LTD. (Analysis result of heat expansion coefficient of
   conventional product of Upirex 35SGV1)
- Evidence B No. 6: Iwanami Rikagaku Jiten, Fifth edition, cover letter, page 405, page 944, colophon (published on February 20, 1998, published by Iwanami Shoten, Publishers)
- Evidence B No. 7: Asahi Glass Research Report No. 57 (2007) pages 37 to 44
- Evidence B No. 8: Japanese Patent No. 3085529
- Evidence B No. 9: Japanese Patent No. 3994946
- Evidence B No. 10: Japanese Patent No. 3355986
- Evidence B No. 11: Japanese Patent Publication No. H04-6213

[Reference Documents]

Reference Document 1: Japanese Unexamined Patent Application Publication No. S63-297029

Reference Document 2: Japanese Patent No. 3346228

No. 5 Judgment by the body

The body determines that none of Demandant's allegations of reasons for the above Reasons for invalidation 1 to 2 has a point. The reason is set forth below.

1. Described matter of Detailed Description of the Invention of the patent specification

The above Reasons for invalidation 1 and 2 both relate to the description requirement of the Detailed Description of the Invention of the patent specification. In view of this, the description of the Detailed Description of the Invention relevant to either of these Reasons for invalidation is pointed out at the offset in the following.

# A "[Technical Field]

The present invention relates to a polyimide film excellent in dimension stability and suitable for a substrate for fine pitch circuit, in particular COF (Chip on Film) wired at a narrow pitch in a film width direction and a copper clad laminate using the same as a base material." (paragraph [0001])

В "Recently, by the way, a bilayer type without adhesives (a copper layer is directly formed on a polyimide film) is used for copper clad laminate to address wire miniaturization. This includes a method of forming a copper layer on a film by a plating method and a method of casting polyamic acid on a copper foil and then subjecting to imidization, neither of which is a heat compression process like a lamination method, and thus it is unnecessary to make a thermal expansion coefficient of the film in MD smaller than that in TD. Furthermore, in COF use where a bilayer type prevails, the pattern wired in a narrow pitch in TD of film is common. Conversely, if thermal expansion coefficient in TD is large, the dimensional change between wires becomes large in the chip mounting bonding, etc., which makes it difficult to measure for the requirement of fine pitch wiring. In order to address this problem, it is ideal to decrease thermal expansion coefficient of film as small as to make it comparable to silicon; however, there is a problem of generation of strain by a heating process including bonding of chip mounting due to difference in thermal expansion coefficient from copper." (paragraph [0005])

# C "[Summary of Invention]

# [Problem to be solved by the invention]

The invention has been made as a result of investigating the solution of problems in the aforementioned conventional technique, and has an object to provide a polyimide film suitable for a substrate for fine pitch circuit including for COF capable of decreasing dimensional change in TD of a film, while maintaining thermal expansion coefficient close to that of metal, and a copper clad laminate using the same as a base material." (paragraphs [0006] to [0007])

#### D "[Means for solving the problem]

In order to achieve the above object, the polyimide film of the present invention has a thermal expansion coefficient  $\alpha$ MD in a machine direction (MD) of a film of 10 to 20 ppm/°C, and a thermal expansion coefficient  $\alpha$ TD in a traverse direction (TD) of 3 to 10 ppm/°C, wherein  $\alpha$ MD is preferably 14 to 18 ppm/°C and  $\alpha$ TD is preferably 3 to 7 ppm/°C." (paragraphs [0007] to [0008])

#### E "[Advantage of the Invention]

The polyimide film of the invention proceeds with orientation of the film to TD, and thus allows for suppressing the thermal expansion coefficient in this direction, and the thermal expansion coefficient in MD has a value close to that of metal, and furthermore, the heat shrinkage rate is low, while maintaining a high elongation elastic modulus." (paragraphs [0010] to [0011])

#### F "[Best Mode for Carrying Out the Invention]

In producing a polyimide film of the present invention, an aromatic diamine component and an acidic anhydride component are polymerized in an organic solvent to obtain a polyamic acid solution.

Specific examples aromatic diamines of the above include paraphenylenediamine, metaphenylenediamine, benzidine, paraxylylenediamine, 4,4'diaminodiphenyl ether, 3,4'-diaminodiphenyl ether, 4,4'-diaminodiphenylmethane, 4,4'-diaminodiphenylsulfone, 3,3'-dimethyl-4,4'-diaminodiphenylmethane, 1.5diaminonaphthalene, 3,3'-dimethoxybenzidine, 1,4-bis(3-methyl-5aminophenyl)benzene, and amido-forming derivatives thereof. It is preferable for the use in a fine pitch base material to adjust the amount of diamine such as paraphenylenediamine, benzidine, or 3,4'-diaminodiphenyl ether among them so that a polyimide film finally obtained may have an elongation elastic modulus of 4.0 GPa or more, since they have the effects of increasing elongation elastic modulus of a film.

Specific examples of the above acidic anhydride components include pyromellitic acid, 3,3'4,4'-biphenyltetracarboxylic acid, 2,3',3,4'biphenyltetracarboxylic acid, 3,3'4,4'-benzophenonetetracarboxylic acid, 2,3,6,7naphthalenedicarboxylic acid, 2,2-bis(3,4-dicarboxyphenyl)ether, pyridine-2,3,5,6tetracarboxylic acid, and acidic anhydride of these amido-forming derivatives.

Further, specific examples of organic solvent used for the formation of polyamic acid solution in the present invention include, for example, sulfoxide-based

solvents such as dimethylsulfoxide and diethylsulfoxide, formamide-based solvents such as N,N-dimethylformamide and N,N-diethylformamide, acetamide-based solvents such as N,N-dimethylacetamide and N,N-diethylacetamide, pyrolidone-based solvents such as N-methyl-2-pyrolidone and N-vinyl-2-pyrolidone, phenol-based solvents such as phenol, o-, m- or p-cresol, xylenol, halogenated phenol and catechol, or aprotic polar solvents such as hexamethylphosphoramide and gamma-butyrolactone. These compounds may be preferably used solely or as a mixture. Further, aromatic hydrocarbons such as xylylene and toluene may be used." (paragraphs [0011] to [0015])

G "Subsequently, the method of producing a polyimide film of the present invention is explained.

The method of forming a polyimide film includes a method of casting a polyamic acid solution into a film and subjecting to thermal ring-opening and desolvation to obtain a polyimide film, and a method of mixing cyclization catalyst and dehydrate with a polyamic acid solution and subjecting to chemical ring-opening to form a gel film and subjecting the film to heating and desolvation to obtain a polyimide film. The latter is preferable, in view that the obtained polyimide film may have a lower thermal expansion coefficient.

In addition, this polyamic acid solution may contain chemically an inert organic filler or inorganic filler such as titanium oxide, fine silica, calcium carbonate, calcium phosphate, calcium hydrogen phosphate, or polyimide filler as necessary to obtain smoothness of a film. Among them, it is particularly preferable to form fine protrusions on a surface by uniformly dispersing fine silica having a particle diameter of 0.07 to 2.0 µm into a film in a proportion of 0.03 to 0.30 weight% on the basis of film resin weight. If the particle diameter falls within a range of 0.07 to 2.0 µm, the inspection by the automatic engineering inspection system may be applied to polyimide films without trouble, which is preferable. If the additive amount exceeds 0.30 weight%, the mechanical strength decreases, whereas if the additive amount falls below 0.03 weight% or less, it fails to show sufficient surface smoothing effects and is thus not preferable. Further, the average particle size is preferably 0.10 µm or more to 0.90 µm or less, more preferably 0.10 µm or more to 0.30 µm or less. If the average particle size is 0.10 µm or less, the surface smoothing effect of film may decrease and thus it is not preferable, whereas if it is 0.90 µm or more, particles may be locally present as coarse particles and thus it is not preferable.

The above polyamic acid solution may contain a cyclization catalyst

(imidization catalyst), a dehydrating agent, a gelation retardant, etc.

Specific examples of cyclic catalyst used herein include aliphatic tertiary amines such as trimethylamine and triethylenediamine, aromatic tertiary amines such as dimethylaniline, and heterocyclic tertiary amines such as isoquinoline, pyridine, and beta-picoline. It is preferable to use at least one kind of amine selected from heterocyclic tertiary amine.

Specific examples of dehydrating agent used herein include aliphatic carboxylic anhydrides such as acetic anhydride, propionic anhydrides and lactic anhydride, and aromatic carboxylic anhydrides such as benzoic anhydride. Among them, acetic anhydride and/or benzoic anhydride is preferable. ...

A method of manufacturing a polyimide film from a polyamic acid solution comprises the steps of casting a polyamic acid solution containing a cyclization catalyst and a dehydrating agent from a slit die on a support to form a film, and partially proceeding with the imidization on the support to form a gel film having selfsupporting property, followed by peeling from the support, heat drying/imidization, and heat treatment.

The above polyamic acid solution passes through a slit die to form a film shape, which is casted on a heated support, and undergoes heat cyclization reaction on the support to form a gel film having self-supporting property, followed by peeling from the support.

The support is a rotary drum made of metal and an endless belt. The temperature is controlled by a heat carrier in liquid or gas form, and/or by radiation heat by an electric heater and a heat carrier in liquid or gas form, and/or by radiation heat of an electric heater.

The gel film is subjected to heating at 30 to 200°C, preferably 40 to 150°C, by receiving heat from the support and/or from a heat source such as heated air or an electric heater to conduct cyclization reaction, and drying volatile portions including free organic solvent to impart self-supporting property, followed by peeling from the support.

The gel film peeled from the above support is drawn in a running direction, while regulating a running speed, usually by a rotary roll. The draw ratio in a machine direction (MDX) is 1.01 to 1.9 times at a temperature of 140°C or less, preferably 1.05 to 1.6 times, further preferably 1.05 to 1.4 times. The gel film drawn in a transporting direction is introduced into a tenter device with both edges in a width direction being gripped by tenter clips, and running together with the tenter clips, and drawn in a width direction. Higher draw ratio in the traverse direction (TD) relative

to the draw ratio in the machine direction (MD) of film, specifically the draw ratio in the traverse direction, is set to 1.1 to 1.5 times as high as the draw ratio in the machine direction, may provide a film preferentially oriented in TD of a film; i.e., having a low thermal expansion coefficient in TD of the film, while maintaining a thermal expansion coefficient in MD of the film close to that of metal. The draw ratios in both directions are adjusted within these ranges so as to have a thermal expansion coefficient  $\alpha$ MD of film of preferably 3 to 10 ppm/°C, more preferably 3 to 7 ppm/°C, a thermal expansion coefficient  $\alpha$ TD of TD of film of preferably 10 to 20 ppm/°C, more preferably 14 to 18 ppm/°C." (Paragraphs [0023] and [0033])

"The polyimide film thus obtained is preferably further subjected to annealing Η treatment at a temperature of 200 to 500°C. This may cause heat relaxation of the film and suppress heat shrinkage rate. The method of producing a polyimide film of the present invention may provide strong orientation in TD of film, which tends to cause high heat shrinkage rate in this direction. Heat relaxation due to annealing treatment may suppress heat shrinkage rate at 200°C to 0.05% or less in both MD and TD of the film, which further increases dimension accuracy and is thus preferable. Specifically, the annealing treatment is implemented by running a film in a furnace at 200 to 500°C under low tensile condition. The film residence time in a furnace is a treatment time, which can be controlled by changing a running speed. The treatment time is preferably from 30 seconds to 5 minutes. A shorter treatment time may result in insufficient heat transfer to a film, whereas a longer treatment time may result in overheating of the film and the loss of smoothness, which is not preferable. Further, the film tension in running is preferably 10 to 50 N/m, further preferably 20 to 30 N/m. A tension lower than this range may impair the running of the film, whereas a higher tension may increase a heat shrinkage rate in a running direction of the obtained film, and is thus undesirable." (paragraph [0036])

I "[Examples]

Hereinafter, the present invention will be specifically discussed by reference to examples.

In addition, PPD in the examples represents paraphenylenediamine, 4,4'-ODA represents 4,4'-diaminodiphenyl ether, 3,4'-ODA represents 3,4'-diaminodiphenyl ether, PMDA represents pyromellitic acid dianhydride, BPDA represents 3,3'-4,4'-diphenyltetracarboxylic acid dianhydride, and DMAc represents N,N-dimethylacetamide.

•••

(7) Dimension change ratio before and after soldering treatment and curl of a copper wired film

(i) Copper layer formation...(ii) Photoresist pattern formation

...

(iii) Copper etching

•••

(iv) Removal of photoresist

•••

(v) Tin plating

•••

(vi) Dimension change ratio and Curl measurement

After tin plating, a dimension in the TD direction was measured (L3). Subsequently, the sample was immersed into a soldering bath at 250°C for 30 seconds, and thereafter a dimension in the TD direction was measured (L4) again. Dimension change ratio before and after the treatment in a solder bath was calculated by the following formula:

Dimension change ratio (%) = {(L4 - L3)/L3}\*100

Further, curl was evaluated by stationary placing a sample in a flat site after treating in a solder bath, and measuring a degree of curling of the edge of the sample from a floor.

[Example 1]

While feeding 239.1 g of DMAc in a 500 ml separable flask, 4.53 g of PPD (0.042 mol), 21.53 g of 4,4'-ODA (0.108 mol), 8.79 g of BPDA (0.030 mol), 26.06 g of PMDA (0.119 mol) were fed thereto and subjected to reaction for one hour at ambient temperature and ambient pressure with the mixture being stirred into a uniform state to obtain a polyamic acid solution.

Subsequently, N,N-dimethylacetamide slurry of silica with an average diameter of 0.30  $\mu$ m in which particles having diameters of less than 0.08  $\mu$ m and 2  $\mu$ m or more were excluded was added by 0.03 weight% to said polyamic acid solution per resin weight, after which the solution was stirred well and dispersed.

Subsequently, this polyamic acid solution was cooled at minus 5°C, and mixed with 15 weight% of acidic anhydride and 15 weight% of  $\beta$ -picoline on the basis of 100 weight% of polyamic acid solution to undergo imidization of polyamic acid.

A polyimide polymer thus obtained was casted for 30 seconds in a rotary drum at 90°C, and the obtained gel film was drawn by 1.1 times in a running direction while being heated for five minutes at 100°C. Subsequently, gripping both edges in a traverse direction, the film was drawn in a traverse direction by 1.5 times while heating for two minutes at 270°C, followed by heating at 380°C for five minutes to obtain a 38  $\mu$ m-thick polyimide film. This polyimide film was subjected to annealing treatment for one minute with a tension of 20 N/m in a furnace where a temperature was set to 220°C, and then each property was evaluated.

Thermal expansion coefficient $\alpha MD$ of Film MD	: 15.8 ppm/°C
Thermal expansion coefficient $\alpha TD$ of Film TD	: 4.8 ppm/°C
200 °C Heat Shrinkage rate (MD)	: 0.02%
200 °C Heat Shrinkage rate (TD)	: 0.02%
Elongation elastic modulus (MD)	: 6.0 GPa
Elongation elastic modulus (TD)	: 6.6 GPa
Additive amounts of Silica	: 0.03 weight%
Particle size distribution	: 0.08 to 2.0 $\mu m$
Average particle diameter	: 0.30 µm
Number of projection	$: 3.2*10^5 /\text{mm}^2$
Dimension change ratio	: 0.02%
Curl	: 2.5 mm
Coefficient of friction	: 0.90

[Examples 2 to 15]

In a similar manner to Example 1, a polyamic acid solution was obtained with respective materials and proportions of aromatic diamine component and aromatic tetracarboxylic acid component, the additive amount of silica, and the average particle size as shown in Tables 1, 2, and 3, followed by drawing with draw ratios in the traverse and longitudinal directions as shown in Tables 1, 2, and 3 to evaluate each property of polyimide film obtained in a similar way to Example 1. The results are shown in Tables 1, 2, and 3.

[Table 1]

実施例		1	2	3	4	5
各原料の比率		PPD 28			199	增加值的
(2,0,12, + )		BPDA 20 PMDA 80			o- ang Britis-poor, Talet	
延伸倍率(MDX)	1	1.1	1.1	1. 1	1. 1	1.1
(TDX)		1.5	1. 5	1.5	1. 5	1.5
熱膨張係数(MD)	ppm/°C	15.8	15.8	15. 8	15. 8	15. 8
(TD)	16	4.8	4.8	4. 8	4. 8	4. 8
加熱収縮率(MD)	%	0. 02	0. 02	0. 02	0. 02	0. 02
(TD)	16	0. 02	0. 02	0. 02	0. 02	0. 02
引張弾性率(MD)	GPa	6.0	6.0	6. 0	6. 0	6. 0
(TD)	<i></i>	6.6	6.6	6.6	6. 6	6. 6
シリカ添加量	重量%	0. 03	0.10	0.10	0. 15	0. 10
流度分布	μm	0. 08~2. 0	0. 08~2. 0	0. 08~2. 0	0. 08~2. 0	0. 08~2. 0
平均粒径	μm	0. 30	0.30	0. 50	0. 50	0. 70
突起数	個/mm <sup>2</sup>	3. 2×10 <sup>5</sup>	9. 3×10 <sup>5</sup>	7.7×10 <sup>5</sup>	1. 2×10 <sup>6</sup>	5.8×10 <sup>5</sup>
寸法変化率	%	0. 02	0. 02	0. 02	0. 02	0. 02
カール	mm	2.5	2.5	2.5	2. 5	2. 5
摩擦係数	28	0. 90	0.72	0.75	0. 42	0. 51

# 実施例 Example

各原料の比率 Ratio of each raw material (molar ratio\*)

モル比 Molar ratio

延伸倍率	Draw ratio
熱膨張係数	Thermal expansion coefficient
加熱収縮率	Heat Shrinkage rate
引張弾性率	Elongation elastic modulus
シリカ添加量	Additive amounts of Silica
重量% Weight%	
流度分布	Particle size distribution
平均粒径	Average particle diameter
突起数 Number of	of projections
個/mm <sup>2</sup>	/mm <sup>2</sup>
寸法変化率	Dimension change ratio
カール Curl	
摩擦係数	Coefficient of friction

[Table 2]

実施例		6	7	8	9	10	
各原料の比率		PPD 25		. 26 (	144. 1 .	- P:100	
(モル比*)		4,4'-ODA 75			0-4 A		
		BPDA 20 PMDA 80	BPDA 20 PMDA 80				
延伸倍率(MDX)		1.1	1.1	1. 1	1.1	1.1	
(TDX)		1.5	1.5	1.5	1.5	1.5	
熱膨張係数(MD)	ppm/°C	16.4	16. 4	16.4	16. 4	16.4	
(TD)	8	6.0	6.0	6. 0	6. 0	6. 0	
加熱収縮率(MD)	%	0.01	0. 01	0. 01	0. 01	0. 01	
(TD)	92	0. 01	0. 01	0. 01	0. 01	0. 01	
引張弾性率(MD)	GPa	5. 9	5. 9	5.9	5.9	5.9	
(TD)	-0	6.4	6.4	6.4	6.4	6.4	
シリカ添加量	<b>重量%</b>	0. 03	0.10	0.10	0. 15	0.10	
流度分布	μm	0. 08~2. 0	0. 08~2. 0	0. 08~2. 0	0. 08~2. 0	0. 08~2. 0	
平均粒径	μm	0. 30	0.30	0.50	0. 50	0. 70	
突起数	個/mm <sup>2</sup>	4. 1 × 10 <sup>5</sup>	1. 2×10 <sup>6</sup>	7.3×10 <sup>5</sup>	1.3×10 <sup>6</sup>	6.4×10 <sup>5</sup>	
寸法変化率	%	0. 03	0. 02	0. 02	0. 02	0. 02	
カール	mm	3. 3	3. 3	3. 3	3. 3	3. 3	
摩擦係数	-5. <b>4</b>	0.95	0.75	0.71	0.39	0.49	

# 実施例 Example

各原料の比率 Ratio of each raw material (molar ratio\*)

モル比 Molar ratio

延伸倍率 Draw ratio

熱膨張係数 Thermal expansion coefficient

加熱収縮率 Heat Shrinkage rate

引張弾性率 Elongation elastic modulus

シリカ添加量 Additive amounts of Silica

重量% Weight%

- 流度分布 Particle size distribution
- 平均粒径 Average particle diameter

# 突起数 Number of projections

個/mm<sup>2</sup> /mm<sup>2</sup>

寸法変化率 Dimension change ratio

カール Curl

摩擦係数 Coefficient of friction

[Table 3	3]
----------	----

実施例		0 11	12	13	14	15
各原料の比率		PPD 20	M . Dida	PPD '70		库田内科
(モル比*)		3,4'-ODA 80 BPDA 20 PMDA 80	ALIO-TAA SI ALIO-TAA			(*1). (*1).
延伸倍率(MDX)		1. 1	1.1	1.1	1.1	1. 1
(TDX)		1. 5	1.5	1.5	. 1. 5	1.5
熱膨張係数(MD)	ppm/°C	14. 1	14.1	14. 1	14. 1	X14. 1
(TD)		4.5	4.5	4. 5	4.5	4. 5
加熱収縮率(MD)	%	0. 02	0. 02	0. 02	0. 02	0. 02
(TD)		0. 01	0.01	0. 01	0. 01	0. 01
引張弾性率(MD)	GPa	6. 7	6.7	6. 7	6. 7	6. 7
(TD)		7.5	7.5	7.5	7. 5	7.5
シリカ添加量	重量%	0. 03	0.10	0. 10	0. 15	0. 10
流度分布	μm	0. 08~2. 0	0. 08~2. 0	0. 08~2. 0	0. 08~2. 0	0. 08~2. 0
平均粒径	μm	0.30	0.30	0. 50	0. 50	0. 70
突起数	個/mm <sup>2</sup>	5. 5×10 <sup>5</sup>	1. 2×10 <sup>8</sup>	8. 2 × 10 <sup>5</sup>	1.4×10 <sup>6</sup>	7.6×10⁵
寸法変化率	%	0. 01	0. 01	0. 01	0. 01	0. 01
カール	mm	3.6	3.6	3.6	3.6	3. 6
摩擦係数	1	0.88	0.71	0.72	0.35	0. 54

実施例 Example

各原料の比率	Ratio of each raw m	naterial (molar ratio*)
--------	---------------------	-------------------------

モル比 Molar ratio

延伸倍率 Draw ratio

熱膨張係数 Thermal expansion coefficient

- 加熱収縮率 Heat Shrinkage rate
- 引張弾性率 Elongation elastic modulus
- シリカ添加量 Additive amounts of Silica

重量% Weight%

- 流度分布 Particle size distribution
- 平均粒径 Average particle diameter
- 突起数 Number of projections
- 個/mm<sup>2</sup> /mm<sup>2</sup>
- 寸法変化率 Dimension change ratio

カール Curl

# 摩擦係数 Coefficient of friction

\* Molar ratio in Table respectively shows mol% in all aromatic diamine components and mol% in all aromatic tetracarboxylic components.

[Comparative Examples 1 to 4]

In a similar manner to Example 1, a polyamic acid solution was obtained with respective proportions of aromatic diamine component and aromatic tetracarboxylic acid component, and the additive amount of silica and the average particle size as shown in Table 4, followed by drawing with draw ratios in the traverse and longitudinal directions as shown in Table 4 to evaluate each property of polyimide film obtained in a similar way to Example 1. The results are shown in Table 4.

[Table 4]

比較例		1	2	3	4
各原料の比率		PPD 70	PPD 25	2 099	PPD 28
(モル比*)		4,4'-ODA 304,4'-ODA 75		4,4'-ODA 72	
		BPDA 25	BPDA 20		BPDA 20
		PMDA 75	PMDA 80		PMDA 80
延伸倍率(MDX)		1.2	1.2	1.1	1. 1
(TDX)		1.3	1.3	1.5	1.5
熱膨張係数(MD)	ppm/°C	7.5	12. 3	16.4	15.8
(TD)	0.02	6. 0	13. 2	6. 0	4.8
加熱収縮率(MD)	%	0. 03	0. 02	0. 01	0. 02
(TD)	- S A8	0. 02	0. 01	0. 01	0. 02
引張弾性率(MD) GPa (TD)		7.7	6.2	5.9	6. 0
		8.0	6.3	6.4	6.6
シリカ添加量	重量%	0. 03	0. 03	無し	無し
流度分布	μm	0. 08~2. 0	0. 08~2. 0	a x a 🐻 🗄	
平均粒径	μm	0. 30	0. 30	10.0	64 - 8-30 S
突起数	個/mm <sup>2</sup>	2. 5×10 <sup>5</sup>	3. $5 \times 10^5$	0	0
寸法変化率	%	0. 02	0.06	0. 02	0.02
カール	mm	9.7	4.7	3. 3	2. 5
摩擦係数		0. 85	0.80	2. 24	2.08

比較例 Comparative Example

各原料の比率 Ratio of each raw material (molar ratio\*)

モル比 Molar rat	io
延伸倍率	Draw ratio
熱膨張係数	Thermal expansion coefficient
加熱収縮率	Heat Shrinkage rate
引張弾性率	Elongation elastic modulus
シリカ添加量	Additive amounts of Silica
重量% Weight%	
無し -	
流度分布	Particle size distribution
平均粒径	Average particle diameter
突起数 Number of	of projections
個/mm <sup>2</sup>	/mm <sup>2</sup>
寸法変化率	Dimension change ratio
カール Curl	
摩擦係数	Coefficient of friction

\* Molar ratio in Table respectively shows mol% in all aromatic diamine components and mol% in all aromatic tetracarboxylic components." (paragraphs [0039] to [0069])

# J "[Industrial applicability]

The polyimide film of the present invention is preferably used for a substrate for fine pitch circuit, in particular COF (Chip on Film) wired at a narrow pitch in TD of film." (paragraphs [0069] to [0070])

2. Regarding Reasons for invalidation 1

(1) Regarding the patent invention

The patent invention (see the aforesaid "No. 2 The invention relating to the Patent"), in order to solve a conventional problem below: "Recently, by the way, a bilayer type without adhesives (a copper layer is directly formed on a polyimide film) is used for copper clad laminate to address wire miniaturization. This includes a method of forming a copper layer on a film by a plating method and a method of casting polyamic acid on a copper foil and then subjecting to imidization, neither of which is a heat compression process like the lamination method, and thus it is

unnecessary to make thermal expansion coefficient of film in MD smaller than that in TD. Furthermore, in COF use where bilayer type prevails, the pattern wired in a narrow pitch in TD of film is common. Conversely, if thermal expansion coefficient in TD is large, the dimensional change between wires becomes large in chip mounting bonding, etc., which makes it difficult to measure for the requirement of fine pitch wiring. In order to address this problem, it is ideal to decrease thermal expansion coefficient of film as small as it is comparable to silicon, however, there is a problem of generating strain by a heating process including bonding of chip mounting due to difference in thermal expansion coefficient from copper" (point B) has "an object to provide a polyimide film suitable for a base material for fine pitch circuit including for COF capable of decreasing dimensional change in TD of a film, while maintaining thermal expansion coefficient close to that of metal, and a copper clad laminate using the same as a substrate" (point C). The object to be solved by the invention may be solved by a substrate for COF to be specified by the patent inventions 1 to 6 of the claims of the Patent, a copper clad laminate to be specified by the patent inventions 7 and 8, and a polyimide film to be specified by the patent invention 9. The method of producing a substrate for COF and a polyimide film capable of solving the problem is the patent inventions 10 and 11.

Further, in order to obtain a polyimide film "wherein a coefficient of thermal expansion  $\alpha$ MD in a machine direction (MD) of film measured in a temperature range of 50 to 200°C and a heat elevation rate of 10°C/min by TMA-50 manufactured by Shimadzu Corporation falls within a range of 10 ppm/°C or more to 20 ppm/°C or less, and wherein a coefficient of thermal expansion  $\alpha TD$  in a traverse direction (TD) measured in said condition falls within a range of 3 ppm/°C or more to 7 ppm/°C or less" of the patented invention, "In producing a polyimide film of the present invention, an aromatic diamine component and an acidic anhydride component are polymerized in an organic solvent to obtain a polyamic acid solution" (point F), "The method of forming a polyimide film includes a method of casting a polyamic acid solution in the form of a film and subjecting to thermal ring-opening and desolvation to obtain a polyimide film, and a method of mixing cyclization catalyst and dehydrate with a polyamic acid solution and subjecting to chemical ring-opening to form a gel film and subjecting the film to heating and desolvation to obtain a polyimide film" (point G), and regarding a method of obtaining a desired gel film, "The latter is preferable, since the obtained polyimide film may have a lower thermal expansion coefficient." (point G), and "The gel film peeled from the above support may be drawn in a running

direction, while regulating a running speed, usually by a rotary roll. The draw ratio in the machine direction (MDX) is 1.01 to 1.9 times at a temperature of 140°C or less, preferably 1.05 to 1.6 times, further preferably 1.05 to 1.4 times. The gel film drawn in a transporting direction is introduced into a tenter device with both edges in a width direction being held by tenter clips, and running together with the tenter clips, and drawn in a width direction. Higher draw ratio in the traverse direction (TD) relative to the draw ratio in the machine direction (MD) of film, specifically the draw ratio in the traverse direction, is set to 1.1 to 1.5 times as high as the draw ratio in the machine direction, may provide a film preferentially oriented in TD of film; i.e., having a low thermal expansion coefficient in TD of film, while maintaining a thermal expansion coefficient similar to metal in MD of film." (point G).

Further, the manufacturing process of four-component polyimide film according to the patent invention is specifically described in Example 1, and the specific method of manufacturing a substrate for COF from the polyimide film and the physical properties of the substrate for COF manufactured from the polyimide film are described in Tables 1 to 3 as Examples 1 to 15 (point I).

# (2) Article 36(4)(i) of the Patent Act according to Reasons for invalidation 1

As described in the aforesaid "(1) Regarding the patent invention," the patent invention specifies "a polyimide film produced by using one or more aromatic diamine components selected from the group consisting of paraphenylenediamine, 4,4'diaminodiphenyl ether, and 3,4'-diaminodiphenyl ether, and one or more acidic anhydride components selected from the group consisting of pyromellitic acid dianhydride and 3,3'-4,4'-diphenyltetracarboxylic acid dianhydride" as a matter for specifying the invention (hereinafter referred to as "matters for specifying the invention"). A four-component polyimide film produced by using aromatic diamine components of paraphenylenediamine and 4,4'-diaminodiphenyl ether, and acidic anhydride components of pyromellitic acid dianhydride and 3,3'-4,4'diphenyltetracarboxylic acid dianhydride, and a four-component polyimide film produced by using aromatic diamine components consisting of paraphenylenediamine and 3,4'-diaminodiphenyl ether, and acidic anhydride components consisting of pyromellitic acid dianhydride and 3,3'-4,4'-diphenyltetracarboxylic acid dianhydride are described as the specific examples (the former four-component system is described in Examples 1 to 10, the latter four-component system is described in Examples 11 to 15).

Consequently, the Detailed Description of the Invention of the patent specification clearly describes the technical significance of the patent invention (points A to C), and clearly describes the general means for obtaining a polyimide film according to the patent invention (point D), and the examples are specifically disclosed for the above four-component system.

Consequently, it is obvious that the Detailed Description of the Invention of the patent specification conforms to the requirement of Article 36(4)(i) of the Patent Act; i.e., the enablement requirement, with respect to the invention of four-component polyimide film as one of the alternatives in the patent inventions.

Further, Demandant does not present an argument to the effect that a person skilled in the art cannot implement the examples.

Accordingly, it is considered hereinafter as to whether or not a person skilled in the art fails to make one of the alternatives of the patent invention of two-component polyimide film on the basis of the descriptions of the patent specification and the common technical knowledge when the patent application was filed.

Evidence A No. 1, Evidence A No. 2, and Evidence A No. 3 to A No. 17 submitted by Demandant fail to disclose a two-component polyimide film having a thermal expansion coefficient in the TD direction  $\alpha$ TD of 3 ppm/°C or more to 7 ppm/°C or less, and a thermal expansion coefficient in the MD direction  $\alpha$ MD of 10 ppm/°C or more to 20 ppm/°C or less, let alone a two-component polyimide film having a thermal expansion coefficient both in the TD and MD directions  $\alpha$ TD and  $\alpha$ MD of 10 ppm/°C or more to 20 ppm/°C or less. Further, the aforesaid respective items of Evidence A fail to disclose a method of obtaining a two-component polyimide film having a thermal expansion coefficient in the TD direction  $\alpha$ TD of 3 ppm/°C or more to 7 ppm/°C or less, and a thermal expansion coefficient in the TD direction  $\alpha$ TD of 3 ppm/°C or more to 7 ppm/°C or less, and a thermal expansion coefficient in the TD direction  $\alpha$ TD of 3 ppm/°C or more to 7 ppm/°C or less, and a thermal expansion coefficient in the TD direction  $\alpha$ TD of 3 ppm/°C or more to 7 ppm/°C or less, and a thermal expansion coefficient in the TD direction  $\alpha$ TD of 3 ppm/°C or more to 7 ppm/°C or less, and a thermal expansion coefficient in the MD direction  $\alpha$ TD of 10 ppm/°C or more to 20 ppm/°C or less.

Evidence B No. 11 discloses in the claims, however, that "1: A dimensionally stable film made of aromatic polyimide obtained from a polymer solution produced by polymerizing 1-biphenyltetracarboxylic acid and phenylenediamines, wherein said polyimide film has an average linear expansion coefficient of about 0.1\*10<sup>-5</sup> to 2.5\*10<sup>-5</sup> cm/cm\*°C in a range of about 50°C to 300°C, and a ratio of linear expansion coefficients in a longitudinal direction of a film (MD direction) and in a traverse direction of a film (TD direction) (MD/TD) of about 1/5 to 4, and when a temperature is elevated from an ambient temperature to  $400^{\circ}$ C, a heat dimension stability shown by a dimensional change of a film at an ambient temperature before and after heating at 400°C for two hours of about 0.3% or less.

2: A method of manufacturing a polyimide film with dimension stability, comprising the steps of: preparing a polymer solution obtained by polymerizing biphenyltetracarboxylic acid and phenylenediamines in an organic polar solvent;

subsequently forming a thin film of the solution on a surface of a support by use of the polymer solution; drying the thin film to form a solidified film body with about 27 to 60 weight% of said solvent and produced moisture remaining;

further peeling the solidified film body from the surface of the support;

drying at a temperature of about 80 to 250°C under low tension of 100 g/mm<sup>2</sup> or less to form a solidified film containing about 5 to 25 weight% of said solvent and produced moisture; and finally subjecting to drying and heat treatment with at least one pair of both edges of said solidified film being fixed at a temperature of 200 to 500°C higher than said drying temperature.",

and the Detailed Description of the Invention discloses a specific manufacturing method as follows:

"Subsequent to the preparation of aforesaid doping solution, the method of the invention comprises the steps of:

(a) preferably continuously or intermittently forming a thin film of the aforesaid doping solution with a uniform thickness on a surface of the support such as drum and a belt made of metal with a smooth surface by a publicly-known solution casting method;

(i) drying the thin film on the support by gradually evaporating a solvent of thin film or produced moisture at a drying temperature of preferably about 40 to 180°C, particularly preferably 50 to 150°C

to form a solidified film body with about 27 to 60 weight%, preferably 30 to 50 weight% of said solvent and produced moisture remaining;

(b) subsequently peeling the solidified film body from the surface of the support;

(i) under "substantially free state or low tension up to said upper limit" of 100 g/mm<sup>2</sup> or less, preferably 80 g/mm<sup>2</sup> or less, and

(ii) drying at a drying temperature of about 80 to 250°C, preferably 100 to 230°C, preferably for about 1 to 200 minutes, particularly for 2 to 100 minutes,

to form preferably continuously or intermittently a solidified film body with about 5 to 25 weight%, preferably 10 to 23 weight% of said solvent and produced moisture being contained.

When elevating temperature from a first stage drying temperature to a second stage drying temperature, it is preferable to elevate temperature in a relatively short time, e.g., a temperature elevating speed of  $10^{\circ}$ C/minutes or more.

In this method of the invention, the increase of tension applied to a solidified film body in drying after peeling said solidified film body from the support may decrease an average linear expansion coefficient of polyimide film finally obtained, thereby adjusting the average linear expansion coefficient to a desired value within the aforesaid range." (publication, page 4, column 7, line 28 to column 8, line 24). Similarly, the Detailed description of the invention discloses in the examples a twocomponent polyimide film (3,3'-4,4'-diphenyltetracarboxylic acid dianhydride and paraphenylenediamine) having a thermal expansion coefficient in the TD direction  $\alpha$ TD of 12 ppm/°C and a thermal expansion coefficient in the MD direction  $\alpha$ MD of 14 ppm/°C (Example 5), a two-component polyimide film (3,3'-4,4'diphenyltetracarboxylic acid dianhydride and paraphenylenediamine) having a thermal expansion coefficient in the TD direction  $\alpha$ TD of 13 ppm/°C and a thermal expansion coefficient in the MD direction aMD of 11 ppm/°C (Example 4), and a method of adjusting a ratio of linear expansion coefficients in the MD direction and the TD direction. Therefore, it is natural to think that a person skilled in the art, who could have known of the patent publication disclosing a polyimide film with  $\alpha$ TD and  $\alpha$ MD that overlapped those of the patent invention, could have obtained a two-component polyimide film of PPD/BPDA of the patent invention.

On the other hand, although it cannot be said that the remaining two-component polyimide films of the patent invention can be obtained on the basis of two-component polyimide films described in the evidences submitted, it cannot be said that only the two-component polyimide films described in the evidences exited as a two-component polyimide film and there was no other two-component polyimide films when the original patent application was filed. Therefore, it cannot be said that a person skilled in the art who had common technical knowledge that thermal expansion coefficient is decreased as a thickness of a film gets thinner, the thermal expansion coefficient is changed by heat treatment of the film, etc. and had known the contents of the Japanese Patent Publication No. H4-6213 could not have obtained a polyimide film having specific linear expansion coefficients in the TD direction and the MD direction according to the patent invention when the original patent application was filed.

Consequently, it cannot be said that a person skilled in the art fails to make a two-component polyimide film of the patent invention on the basis of the descriptions of the patent specification and the common technical knowledge when the patent application was filed.

As seen above, the Detailed Description of the Invention of the patent specification discloses a technical significance of the patent invention (points A to C), a general means for obtaining a polyimide film of the patent invention (point D), a specific example of four-component polyimide film (point I), respectively. It cannot be said that a person skilled in the art fails to make a two-component polyimide film of the patent invention on the basis of the descriptions of the Detailed Description of the Invention and the common technical knowledge when the patent application was filed. Therefore, it can be said that the Detailed Description of the Invention discloses definitely and sufficiently to the extent that allows a person skilled in the art to understand and implement the patent invention.

Consequently, the Detailed Description of the Invention in the patent specification conforms to the requirement of Article 36(4)(i) of the Patent Act.

(3) Nonconformance to Article 36(6) of the Patent Act according to Reasons for invalidation 1

The determination of whether or not the recitation of the Claims might comply with the requirement of Article 36(6)(i) of the Patent Act (hereinafter referred to as "the support requirement") should follow the following steps of: comparing the recitation of the Claims with the description of the Detailed Description of the Invention; and considering whether or not the Invention recited in the Claims might be the invention described in the Detailed Description of the Invention and fall within the range in which a person skilled in the art could recognize that the problem to be solved by the Invention might be solved by the Detailed Description of the Invention.

The patent invention specifies "a polyimide film produced by using one or more selected aromatic diamine components from the group consisting of paraphenylenediamine, 4,4'-diaminodiphenyl ether, and 3,4'-diaminodiphenyl ether, and one or more acidic anhydride components selected from the group consisting of pyromellitic acid dianhydride and 3,3'-4,4'-diphenyltetracarboxylic acid dianhydride" as a matter specifying the invention. Therefore, it is confirmed hereinafter as to whether or not there is any description that allows a person skilled in the art to recognize that the patent invention with regard to various polyimide films including two-component films contained in the description may solve the problem to be solved by the patent invention.

The description of the patent specification with regard to the patent invention is as mentioned in the above item (1). It can be seen from the description that specific values of thermal expansion coefficients in the TD direction and the MD direction of polyimide film may solve the problem to be solved by the patent invention without relation to the resin composition constituting a polyimide film, in view of the common technical knowledge of a person skilled in the art when the patent application was filed.

Consequently, the patent invention directed to a two-component system of "a polyimide film produced by using one or more aromatic diamine components selected from the group consisting of paraphenylenediamine, 4,4'-diaminodiphenyl ether, and 3,4'-diaminodiphenyl ether, and one or more acidic anhydride components selected from the group consisting of pyromellitic acid dianhydride and 3,3'-4,4'-diphenyltetracarboxylic acid dianhydride" is described to the extent that allows a person skilled in the art to recognize that the problem to be solved by the patent invention may be solved. Therefore, the patent inventions of the claims are the inventions described in the Detailed Description of the Invention, and thus conform to Article 36(6)(i) of the Patent Act.

# (4) Demandant's allegation

Demandant argues about the following matters with respect to the Reasons for invalidation 1 in the written demand for trial, the oral proceedings statement brief on April 10, 2013, and the written statement on May 24, 2013.

(A) In drawing technique of polyimide film, in order to adjust a thermal expansion coefficient in the MD direction and a thermal expansion coefficient in the TD direction to specific numerical ranges, it is necessary to precisely control operation condition such as the selection of raw materials of polyimide, and drawing temperature and draw ratio of the film. In drawing control, a thermal expansion coefficient in the MD direction is decreased by drawing in the MD direction, whereas a thermal expansion coefficient in the TD direction gets elevated. Thus extensive trial and error is required to adjust thermal expansion coefficients in both the MD direction and the TD direction to specific ranges, which can be achieved only by a range satisfying a specific condition.

(B) A massive majority of two-component polyimide films of PPD (paraphenylenediamine) and BPDA (3,3',4,4'-diphenyltetracarboxylic anhydride) have a thermal expansion coefficient of 5 ppm or less, and thus too low a thermal expansion coefficient as compared to the thermal expansion coefficient of the patent invention to be adjustable to a thermal expansion coefficient of the patent invention by drawing.

(C) Two-component polyimide film of ODA (diaminodiphenyl ether) and PMDA (pyromellitic acid dianhydride) has too high a thermal expansion coefficient to be adjusted to a thermal expansion coefficient of the patent invention by drawing.

(D) A two-component polyimide film of ODA and BPDA has too high a thermal expansion coefficient to be adjusted to a thermal expansion coefficient of the patent invention by drawing.

(E) The whole range of the claims must satisfy the enablement requirement, whereas two-component polyimide film is infeasible as discussed in the above items (B), (C), and (D), and thus the description does not conform to the requirement of Article 36(4)(i) of the Patent Act.

(F) Demandee submits Evidence B No. 8 to B No. 11 that disclose a manufacturing method of two-component polyimide film which is not a manufacturing method by the chemical cyclization method described in the examples of the patent specification, but a manufacturing method by the heat cyclization method. Therefore, a manufacturing method of the patent specification cannot be applied, and a person skilled in the art cannot obtain a film with specific linear expansion coefficients in the MD direction and the TD direction of the patent invention when the patent application was filed.

(G) Demandee submits Evidence B No. 8 to B No. 11 that disclose a two-component polyimide film PPD/BPDA having a thermal expansion coefficient of 10 to 20 ppm/°C. A person skilled in the art cannot manufacture a two-component polyimide film of PPD/BPDA having a desired thermal expansion coefficient (10 to 20 ppm/°C).

(H) The polyimide film of Evidence B No. 8 to B No. 11 is inappropriate for drawing. This is also described in Evidence A No. 13. In a two-component polyimide film of PPD/BPDA, thermal expansion coefficient gets smaller than a numerical range of the patent invention by chemical imidization described in the patent specification, which

makes it impossible to realize the numerical value by drawing described in the patent specification. In the case of the polyimide film to be produced by heat imidization, the drawing is difficult. A person skilled in the art cannot manufacture a film of the patent invention.

(I) Demandant may realize a thermal expansion coefficient falling within a numerical range of the patent invention not only by drawing but also his own unique knowhow. It is obvious, however, that the description of the patent specification is infeasible.

(J) Regarding two component system of ODA and PMDA as well as two-component system of ODA and BPDA, there is totally no evidence showing the existence of a polyimide film having 12 to 20 ppm/°C in an unstretched state where Demandee believes this one is enough for feasibility. Further, there is no argument to the effect that a predetermined thermal expansion coefficient of the patent invention is achieved with regard to a two-component system of ODA and BPDA or even a two-component system of ODA and PMDA, which is a composition of Demandee's polyimide film product, whose thermal expansion coefficient is thus well-known to Demandee. Therefore, it is evidenced that a polyimide film having a predetermined thermal expansion coefficient of the patent invention is obtained in neither a two-component system of ODA and PMDA, nor a two-component system of ODA and BPDA.

Hereinafter, these arguments are considered.

# [Regarding allegation (A)]

It was common technical knowledge for a person skilled in the art when the patent application was filed that, in drawing technique of polyimide film, it is necessary to precisely control operation conditions such as the selection of raw materials of polyimide, and the drawing temperature and the draw ratio of the film, and in drawing control, a thermal expansion coefficient in one direction is decreased by drawing in the one direction, whereas a thermal expansion coefficient in another direction is elevated. In view of the description of the patent specification and the common technical knowledge when the patent application was filed, it cannot be said that undue trial and error is required for the implementation of the patent invention. Thus the Demandant's allegation (A) is not acceptable.

[Regarding allegations (B) to (D)]

The arguments (B) to (D) are as considered in the above item 2.(2).

[Regarding allegation (E)]

Article 36(4)(i) of the Patent Act specifies that "the Detailed Description of the Invention should ... disclose definitely and sufficiently to the extent that allows a person who has ordinary knowledge in the technical field to which the invention belongs to implement the invention." This cannot be construed as being required to show the embodiments for all the options of the invention recited in the claim.

Further, as is discussed above, it cannot be said that a person skilled in the art fails to make two-component polyimide as an alternative of the patent invention on the basis of the descriptions of the patent specification and the common technical knowledge when the patent application was filed. Therefore, the Demandant's allegation (E) is not acceptable.

#### [Regarding allegations (F) to (I)]

First, a manufacturing method specified in the Inventions 10 and 11 fails to describe a curing method of polyimide. The Inventions 10 and 11 are not limited to the heat cyclization method. Therefore, the Demandant's allegation is based on an erroneous premise.

Subsequently, a manufacturing method of polyimide film described in Evidence B No. 8 to B No. 11 is a manufacturing method by heat cyclization, which is distinct from a manufacturing method by chemical cyclization described in the examples of the patent specification. The patent specification does not totally exclude manufacturing by the heat cyclization method; however, in view of the following description of heat cyclization method for a manufacturing method of polyimide film according to the patent invention: "The method of forming a polyimide film includes a method of casting a polyamic acid solution in the form of a film and subjecting to thermal ring-opening and desolvation to obtain a polyimide film, and a method of mixing cyclization catalyst and dehydrating with a polyamic acid solution and subjecting to chemical ring-opening to form a gel film and subjecting the film to heating and desolvation to obtain a polyimide film." (point G).

Further, taking into consideration the common technical knowledge of a person skilled in the art who could know of Evidence B No. 11 when the patent application

was filed, at least two-component polyimide film of PPD and BPDA described in Evidence B No. 11 could be obtained by use of heat cyclization, as is considered in the above item 2.(2).

Further, regarding the argument to the effect that a person skilled in the art cannot manufacture a polyimide film described in the examples of the patent publication of Evidence B No. 11, its specific reason is indefinite, and thus is not acceptable.

Consequently, the Demandant's allegation is not reasonable, and thus not acceptable.

#### [Regarding allegation (J)]

As is confirmed in the consideration of the supporting requirement of the above item (3), the patent invention may provide a substrate for fine pitch excellent in dimensional stability by adjusting thermal expansion coefficients in the MD direction and the TD direction of a polyimide film for COF to a specific range. The patent invention directed to a two-component system of "a polyimide film produced by using one or more aromatic diamine components selected from the group consisting of paraphenylenediamine, 4,4'-diaminodiphenyl ether, and 3,4'-diaminodiphenyl ether, and one or more acidic anhydride components selected from the group consisting of pyromellitic acid dianhydride and 3,3'-4,4'-diphenyltetracarboxylic acid dianhydride" is described to the extent that allows a person skilled in the art to recognize that the problem to be solved by the patent invention may be solved.

Further, in the patent invention having a plurality of options as a composition, even if the patent specification only discloses a four-component polyimide film, it cannot be said that two-component polyimide film could not be implemented in view of the description of the patent specification and the common technical knowledge of a person skilled in the art when the patent application was filed, as is discussed in the above item 2.(2). Therefore, Demandee's failure to submit evidence of feasibility does not lead to a conclusion that the Detailed Description of the Invention of the patent specification violates the provision of Article 36(4)(i) of the Patent Act. Thus the Demandant's allegation is not acceptable.

(5) Summary of Reasons for invalidation 1

As seen above, the Demandant's allegation of Reasons for invalidation 1 may not be the grounds for invalidating the Patent.

3. Regarding Reasons for invalidation 2

(1) Nonconformance to Article 36(4)(i) of the Patent Act according to Reasons for invalidation 2

The patent inventions 10 and 11 are inventions of process of manufacturing the product. Thus the feasibility of the method is usability of the method. Thus the mode for carrying out the invention is also necessary to be described so as to allow for this. There are the following two requirements as a prerequisite for this:

- Part 1: The invention of process should be definitely described.
- Part 2: A product can be made through the process.

Accordingly, when the above two requirements are confirmed with respect to the description of the Detailed Descriptions of the Invention of the Patent, "Part 1" is definitely described with respect to the patent inventions 10 and 11 on the basis of the point G.

Regarding "Part 2," it can be seen from the point I that a polyimide film of the patent invention 9 and a substrate for COF of the patent invention 1 are manufactured as well as the specific manufacturing process of four-component polyimide film. Therefore, a part of the embodiment included in alternatives of polyimide film of the patent inventions 10 and 11 is specifically disclosed.

Consequently, it is obvious that the Detailed Description of the Invention of the patent specification conforms to the requirement of Article 36(4)(i) of the Patent Act; i.e., the enablement requirement, with regard to the patent invention 11 which specifies a method of manufacturing a four-component polyimide film of the patent invention 9 as one of the alternatives in the patent inventions 10 and 11, and with regard to the patent invention 10 which specifies a method of producing a substrate for COF of the patent invention 1.

Further, Demandant does not present an argument to the effect that a person skilled in the art cannot implement the examples.

Accordingly, it is considered hereinafter as to whether or not a person skilled in the art fails to make the product with regard to the patent invention 11 which specifies a method of producing a two-component polyimide film of the patent invention 9 as one of the alternatives in the patent inventions 10 and 11, and with regard to the patent invention 10 which specifies a method of producing a substrate for COF of the patent invention 1 on the basis of the descriptions of the patent specification and the common technical knowledge when the patent application was filed.

As is discussed in the above item 2.(2), Evidence B No. 11 discloses in the examples a two-component polyimide film (3,3'-4,4'-diphenyltetracarboxylic acid dianhydride and paraphenylenediamine) having a thermal expansion coefficient in the TD direction  $\alpha$ TD of 12 ppm/°C and a thermal expansion coefficient in the MD direction  $\alpha$ MD of 14 ppm/°C (Example 5), a two-component polyimide film (3,3'-4,4'-diphenyltetracarboxylic acid dianhydride and paraphenylenediamine) having a thermal expansion coefficient in the TD direction  $\alpha$ TD of 13 ppm/°C and a thermal expansion coefficient in the TD direction  $\alpha$ MD of 11 ppm/°C (Example 4), and a method of adjusting a ratio of linear expansion coefficients in the MD direction add of the patent publication disclosing a polyimide film with  $\alpha$ TD and  $\alpha$ MD that overlapped those of the patent invention, could have obtained a two-component polyimide film of PPD/BPDA of the patent invention by subjecting the polyimide film to the drawing treatment of the Invention 10 or 11.

Further, Demandant alleges that a film would break if it were drawn by 1.1 times or more in a condition of Evidence B No. 8 to B No. 11; however, whether or not a drawing is appropriate may vary depending on a temperature condition during the drawing. Thus the argument without defining a temperature condition is unreasonable and unacceptable.

On the other hand, although it cannot be said that the remaining two-component polyimide films of the patent invention can be obtained on the basis of two-component polyimide films described in the evidences submitted, it cannot be said that only the two-component polyimide films described in the evidences exited as a two-component polyimide film and there was no other two-component polyimide films when the original patent application was filed. Therefore, it cannot be said that a person skilled in the art who had common technical knowledge that thermal expansion coefficient is decreased as a thickness of a film gets thinner, the thermal expansion coefficient is changed by heat treatment of the film, etc. and had known the contents of the Japanese Patent Publication No. H4-6213 could not have obtained a polyimide film having specific linear expansion coefficients in the TD direction and the MD direction according to the patent invention based on the manufacturing method of the patent inventions 10 and 11 when the original patent application was filed.

Consequently, it cannot be said that a person skilled in the art fails to make a two-component polyimide film of the patent invention on the basis of the descriptions of the patent specification and the common technical knowledge when the patent application was filed with respect to a manufacturing method of two-component polyimide film in the patent inventions 10 and 11.

As seen above, the Detailed Description of the Invention of the patent specification discloses definitely and sufficiently to the extent that allows a person skilled in the art to implement the patent inventions 10 and 11, thus conforming to Article 36(4)(i) of the Patent Act.

(2) Nonconformance to Article 36(6) of the Patent Act according to Reasons for invalidation 2

For a reason similar to that discussed in the above item 2.(3), the patent inventions 10 and 11 of the claims are the inventions described in the Detailed Description of the Invention, and thus conform to Article 36(6)(i) of the Patent Act.

#### No. 6 Closing

As seen above, even if the Demandant's allegation in the written demand for trial, the oral proceedings statement brief on April 10, 2013, and the written statement on May 24, 2013 as well as means of proof were taken into account, the Reasons for invalidation 1 and 2 as Demandant argues may not be the grounds for invalidating the Patent.

The costs in connection with the trial shall be borne by Demandant under the provisions of Article 61 of the Code of Civil Procedure as applied mutatis mutandis to the provision of Article 169(2) of the Patent Act.

Therefore, the trial decision shall be made as described in the Conclusion.

July 30, 2013

Chief administrative judge:	TAGUCHI, Masahiro
Administrative judge:	ONODERA, Tsutomu
Administrative judge:	OSHIMA, Shogo