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

Tokyo, Japan
Attorney HINO, EIICHIRO

Tokyo, Japan
Attorney UENO, JUNICHI

Tokyo, Japan
Attorney IMADA, HITOMI

Tokyo, Japan
Demandee DU PONT-TORAY COMPANY LTD.

Tokyo, Japan
Attorney MASUI, Kazuo

Tokyo, Japan
Attorney HASHIGUCHI, Naoyuki

Tokyo, Japan
Attorney SAITO, Seijiro

Regarding the trial decision dated July 30, 2013 for the case of patent invalidation trial of Patent No. 4777471, titled "polyimide film and copper clad laminate using the film as base material" between the parties, the IP High Court has made a court decision to cancel the trial decision (2013 (Gyo-Ke) No. 10250, rendered on April 28, 2015), which has been made final and binding. In response, as a result of further proceeding, a decision is made as follows:

Conclusion

The patents regarding the inventions according to Claims 1 to 11 of Patent No. 4777471 shall be invalidated.

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

Reason

No.1 Demandant's demand

The gist is the same as the above conclusion. Specifically, the demandant seeks a trial decision to the effect that the patents for the inventions according to Claims 1 to 11 of Patent No. 4777471 (the number of claims as of the registration was 11; hereinafter, the Patent is referred to as "the Patent") shall be invalidated, and the costs in connection with the trial shall be borne by the demandee.

No. 2 History of the procedures, etc.

1(1) The demandee is the Patentee of the Patent titled "polyimide film and copper clad laminate using the film as base material."

(2) The Patent is derived from a divisional patent application based on a patent application filed March 25, 2005 (Japanese Patent Application No. 2005-88334; priority claimed on the basis of earlier application: March 30, 2004, hereinafter referred to as "the original application") and registered on July 8, 2011.

2(1) The demandant made a demand for the trial seeking for a trial decision to invalidate the patents for the inventions according to Claims 1 to 11 on November 30, 2012. In response, the demandee submitted a written reply on February 15, 2013.

(2) The chief administrative judge notified both parties of matters to be examined in the oral proceeding on March 13, 2013 (Notification of trial examination), and in response, the demandant and the demandee respectively submitted oral proceedings statement briefs on April 10, 2013. Further, the demandee submitted a written statement on April 22, 2013.

(3) On April 24, 2013, in the presence of the demandant's representative and the demandee's representative, the first oral proceeding was held, and the parties were advised of the case being transferred to the examination by documentary proceeding.

(4) The demandant and the demandee respectively submitted written statements on May 24, 2013.

3 The Japan Patent Office made a trial decision on July 30, 2013 to the effect that "the demand for the trial of the case was groundless." (hereinafter simply referred to as "first trial decision"), copies of which were sent to both parties on August 8.

4 The demandant filed a suit on September 4, 2013 with the IP High Court against the first trial decision to complain about the first trial decision (2013 (Gyo-Ke) No. 10250). The Court rendered a judgment on April 28, 2015 to the effect that the first trial decision should be canceled, and the judgment (hereinafter referred to as "cancellation judgment") was made final and binding.

5 The binding of the above cancellation judgment leads to a further proceeding pursuant to the provision of Article 181(2) of the Patent Act. (Further, the demandee did not file a petition pursuant to Article 134-3 of the Patent Act.)

6 The Japan Patent Office sent an advance notice of a trial decision on July 21, 2015 to give an opportunity for the demandee to request for correction within a designated period, but there was no response from the demandee.

No. 3 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"). Further, the specification of the invention is referred to as "the patent specification."

"[Claim 1]

A base material 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 μm, wherein a polyimide film is produced by using one or more aromatic diamine components selected from the group consisting of paraphenylenediamine (Trial Decision's note: hereinafter sometimes referred to as "PPD"), 4,4'-diaminodiphenyl ether (Trial Decision's note: hereinafter sometimes referred to as "4,4'-ODA"), and 3,4'-diaminodiphenyl ether (Trial Decision's note: hereinafter sometimes referred to as "3,4'-ODA"), and one or more acidic anhydride components selected from the group consisting of pyromellitic acid dianhydride (Trial Decision's note: hereinafter sometimes referred to as "PMDA") and 3,3',4,4'-diphenyltetracarboxylic dianhydride (Trial Decision's note: hereinafter sometimes referred to as "BPDA"), wherein said polyimide film comprises fine silica with a particle diameter of 0.07 to 2.0 μm, and wherein a thermal expansion coefficient αMD in a machine direction (MD) of the film measured within 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 αTD in a transverse direction (TD) measured in said condition falls within a range of 3 ppm/°C or more to 7 ppm/°C or less.

[Claim 2]

The base material for COF of Claim 1, wherein a thermal expansion coefficient αMD in a machine direction (MD) of the film measured within 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 αTD in a transverse direction (TD) measured in said condition falls within a range of 3 ppm/°C or more to 7 ppm/°C or less.

[Claim 3]

The base material 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 a fine protrusion is formed on a surface.

[Claim 4]

The base material for COF of Claim 3, wherein said fine silica has an average particle size of 0.10 μm or more to 0.90 μm or less.

[Claim 5]

The base material for COF of Claim 3, wherein said fine silica has an average particle size of 0.10 μm or more to 0.30 μm or less.

[Claim 6]

The base material 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^2.

[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 μm, 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 μm, and wherein a thermal expansion coefficient αMD 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 αTD in a transverse direction (TD) measured in said condition 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 αMD 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 αTD in a transverse direction (TD) measured in said condition 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 μm, and wherein a thermal expansion coefficient αMD in a machine direction (MD) of film measured within 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 αTD in a transverse direction (TD) measured in said condition falls within a range of 3 ppm/°C or more to 7 ppm/°C or less.

[Claim 10]

A method of producing a base material 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 transverse 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 transverse 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."

No. 4 Allegations by the parties

1 Demandant's allegation relating to the reasons for invalidation

According to the following grounds (1) and (2) for invalidation regarding the patent inventions, the patents with respect to the inventions according to Claims 1 to 11 correspond to the provision of Article 123(1)(iv) of the Patent Act and should be invalidated (First oral proceeding record and overall purport of allegation). In addition, the 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.

Further, the documents (A1 to A18) as documentary evidence were submitted as listed in the following item (3).

(1) Ground 1 for invalidation

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 a four-component polyimide film disclosed in the examples, e.g. the two-component polyimide film, whereas the recitation of the claims encompasses such two-component polyimide film.

Therefore, with respect to 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., fails to satisfy the requirement of Article 36(6)(i) of the Patent Act. Therefore, the patents should be invalidated under the provision of Article 123(1)(iv) of the Patent Act.

(2) Ground 2 for invalidation

It cannot be seen from the Detailed Description of the Invention of the patent specification as to whether or not a base material for COF according to patent invention 1 may be obtained by a method of producing a base material for COF according to patent invention 10, nor whether or not a polyimide film according to patent invention 9 may be obtained by a method of producing a polyimide film according to patent invention 11. Therefore, it fails to disclose definitely and sufficiently to the extent that allows a person skilled in the art to implement 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 patent inventions 10 and 11 fails to conform to Article 36(6)(i) of the Patent Act; i.e., fails to satisfy the requirement provided in Article 36(6)(i) of the Patent Act. Therefore, the patents should be invalidated under the provision of Article 123(1)(iv) of the Patent Act.

(Trial Decision's note: Regarding the above items (1) and (2), for the convenience of trial decision, the reason according to the violation of Article 36(4)(i) of the Patent Act (so-called the violation of enablement requirement) with respect to the invalidation reasons 1 and 2 is referred to as "invalidation ground A," and the reason according to the violation of Article 36(6)(i) of the Patent Act (so-called the violation of supporting requirement) is referred to as "invalidation ground B.")

(3) 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 parent 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 parent application of the Patent)
Evidence A No. 7: POLYMER, 1989, Vol. 30, June (Conference issue), pages 1170 to 1173 (only a part of abstract)
Evidence A No. 8: Planned and Edited by Kotaro Senba, "Top-edge 'materials' and 'technology' revealed by top companies' practicians, Complete collection of latest electronics mounting <first volume>", page 136 (published date: June 29, 2007, published by technical information institute Co., Ltd)
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)

2 The demandee's allegation

(1) Object of the reply and outline of argument

The demandee seeks for a trial decision to the effect that the demand for trial regarding the invalidation is groundless, and the costs in connection with the trial shall be borne by the demandant since none of the demandant's allegations of reasons for invalidation 1 to 2 has a point.

Further, the documents (B No. 1 to B No. 11) as documentary evidence were submitted as listed in the following item (2).

(2) Means of proof

Evidence B No. 1: A low-thermal expansion polyimide film, Capton 150EN-A (A catalog of the demandee's products as an implemented product of the patent invention)

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: Patent No. 3085529

Evidence B No. 9: Patent No. 3994946

Evidence B No. 10: Patent No. 3355986

Evidence B No. 11: Japanese Patent Publication No. H4-6213

No. 5 Judgment

As mentioned below, the collegial body construes that the reasons A and B for invalidation have a point, and thus the patents for the inventions according to Claims 1 to 11 of the Patent should be invalidated.

1 Regarding the present invention

The Claims of the patent invention is set forth as in the foregoing Part 2. The Detailed Description of the Invention of the patent specification generally has the following descriptions:

"[Technical field]

[0001]

The present invention relates to a polyimide film excellent in dimension stability and suitable for a base material 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.

[Background Art]

[0002]

In accordance with higher integration requirement of flexible print substrate and semiconductor package, the requirements for polyimide used for the same are increasing, e.g. the decrease in the dimensional change and curl due to the lamination with metal, and high handleability. Polyimide film is required to have physical properties of thermal expansion coefficient equivalent to metals and high elastic modulus, and further small dimensional change by moisture absorption. Accordingly, a polyimide film has been developed.

[0005]

Incidentally, in recent years, 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 polyamide 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 a thermal expansion coefficient in TD is large, the change of dimension 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 a thermal expansion coefficient of film as small as to be comparable to silicon; however, there is a problem of generating strain by a heating process including a bonding process of chip mounting, due to a difference in thermal expansion from copper.

[Problem to be Solved by the Invention]

[0007]

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

[Means for Solving the Problem]

[0008]

In order to achieve the above goal, the polyimide film of the present invention has a thermal expansion coefficient αMD in machine direction (MD) of a film of 10 to 20 ppm/°C, and a thermal expansion coefficient αTD in transverse direction (TD) of 3 to 10 ppm/°C, wherein αMD is preferably 14 to 18 ppm/°C and αTD is preferably 3 to 7 ppm/°C.

[0010]

Further, the copper clad laminate of the present invention comprises a polyimide film having any of the above features as a base material, and a copper with a thickness of 1 to 10 μm formed thereon.

[Advantage of the Invention]

[0011]

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.

[Description of Embodiments]

[0012]

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

[0013]

Specific examples of the above aromatic diamines 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, 1,5-diaminonaphthalene, 3,3'-dimethoxybenzidine, 1,4-bis(3-methyl-5-aminophenyl)benzene, and amido-forming derivatives thereof. It is preferable for the use in a fine pitch base material to adjust an 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.

[0014]

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, 2,3,6,7-naphthalenedicarboxylic acid, 2,2-bis(3,4-dicarboxyphenyl)ether, pyridine-2,3,5,6-tetracarboxylic acid, and acidic anhydride of these amido-forming derivatives.

[0022]

Polyamide acid solution thus obtained comprises 5 to 40 weight%, preferably 10 to 30 weight% of a solid content, and the viscosity is 10 to 2000 Pa*s, preferably 100 to 1000 Pa*s as a value measured by a Brookfield viscometer for stable liquid supply. Further, a polyamide acid solution in an organic solvent solution may partially undergo imidization.

[0023]

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

[0024]

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

[0025]

In addition, this polyamide acid solution may contain a chemically inert organic filler or an inorganic filler such as titanium oxide, fine silica, calcium carbonate, calcium phosphate, calcium hydrogen phosphate, or polyimide filler as necessary to obtain the smoothness of a film. Among them, it is particularly preferable to form fine protrusions by uniformly dispersing a 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 a film resin weight basis....

[0029]

A method of manufacturing a polyimide film from a polyamide acid solution comprises the steps of casting a polyamide 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 self-supporting property, followed by peeling from the support, heat drying/imidization, and heat treatment.

[0030]

The above polyamide 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.

[0031]

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

[0032]

The above 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 perform cyclization reaction, and drying volatile portions including a free organic solvent to impart self-supporting property, after which the gel film is peeled from the support.

[0033]

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 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 transverse direction (TD) relative to the draw ratio in the machine direction (MD) of film, specifically the draw ratio in the transverse direction, is set to 1.1 to 1.5 times 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 αTD of TD of film of preferably 3 to 10 ppm/°C, more preferably 3 to 7 ppm/°C, and a thermal expansion coefficient αMD of MD of film of preferably 10 to 20 ppm/°C, more preferably 14 to 18 ppm/°C.

[0034]

The film dried in the above drying zone is heated for 15 seconds to 10 minutes with hot air, an infrared heater, etc. Subsequently, the film is subjected to heat treatment at a temperature of 250 to 500 for 15 seconds to 20 minutes with hot air and/or an electric heater.

[0035]

Further, the thickness of polyimide film is adjusted by adjusting a running speed, and preferably is 3 to 250 μm. Thinner or thicker thickness significantly deteriorates the film-forming ability, and thus is not preferable.

[0036]

The polyimide film thus obtained is preferably further subjected to annealing treatment at a temperature of 200 to 500°C. This may cause heat relaxation of the film and suppress heat shrinkage rate. ...

[0039]

The polyimide film thus obtained and copper clad laminate using the film as a base material proceed with orientation of the film to TD, and thus 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. Therefore, it is suitable for a base material for fine pitch circuit, in particular COF (Chip on Film) wired at a narrow pitch in TD of film.

[Examples]

[0040]

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

[0041]

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.

[0042]

Further, properties in the examples were measured as in the following.

[0043]

(1) Thermal expansion coefficient:

measured by TMA-50, manufactured by Shimadzu Corporation, in a measurement temperature range of 50 to 200°C and a temperature elevation rate of 10°C/min.

[0044]

(2) Heat Shrinkage rate:

a film with a size of 20 cm*20 cm was prepared. A film dimension (L1) after leaving for 2 days in a room adjusted to 25°C and 60% RH was measured, and subsequently the film was heated to 200°C for 60 minutes, and left again for 2 days in a room adjusted to 25°C and 60% RH to measure a film dimension (L2), and evaluated by calculation with the following formula:

Heat Shrinkage rate = - (L2 - L1)/L1*100

[0045]

(3) Elongation elastic modulus...

[0046]

(4) Particle size distribution

[0047]

(5) Number of projections

[0048]

(6) Coefficient of friction (coefficient of static friction) ...

[0049]

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

(i) Copper layer formation...

[0050]

(ii) Photoresist pattern formation...

[0053]

(iii) Copper etching

After forming a photoresist in a trace pattern ... was subjected to etching treatment to form a patterning of a copper layer at 100 μm pitch (wiring width: 50 μm/ wiring gap: 50 μm). After copper etching, the film as immersed at 25°C for 5 minutes 2 times + water washing with swinging, followed by natural drying.

[0054]

(iv) Removal of photoresist...

[0055]

(v) Tin plating...

[0056]

(vi) Dimension change ratio and Curl measurement

After tin plating, a dimension in TD direction (L3) was measured. Subsequently, the film was immersed into a soldering bath at 250°C for 30 seconds, and thereafter a dimension in TD direction (L4) was measured 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

[0057]

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

[0058]

[Example 1]

After feeding of 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 uniformly stirred to obtain a polyamide acid solution.

[0059]

Subsequently, N,N-dimethylacetamide slurry of silica with an average diameter of 0.30 μm in which particle diameters of less than 0.08 μm and 2 μm or more was excluded was added by 0.03 weight% to said polyamide acid solution on a resin weight basis, and stirred well and dispersed.

[0060]

Subsequently, this polyamide acid solution was cooled at minus 5°C, and mixed with 15 weight % of acidic anhydride and 15 weight% of β-picoline on the basis of 100 weight % of polyamide acid solution.

[0061]

This mixture fluid 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, with both edges being gripped in a transverse direction, the film was stretched in a transverse direction by 1.5 times while heating for two minutes at 270°C, and heating at 380°C for five minutes to obtain a 38 μm-thick polyimide film. This polyimide film was subjected to annealing treatment for one minute with a tension of 20 N/m being applied in a furnace where a temperature was set to 220°C, and then each property was evaluated.

Thermal expansion coefficient αMD of Film MD: 15.8 ppm/°C

Thermal expansion coefficient αTD of Film TD: 4.8 ppm/°C

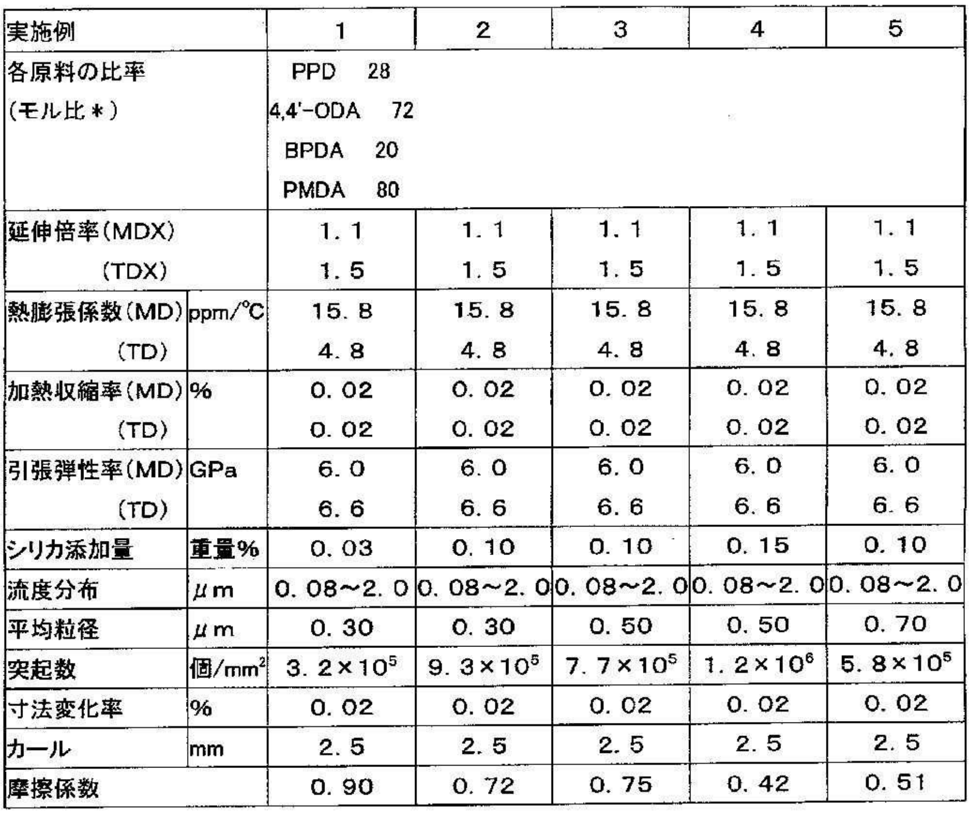
[0062]

[Examples 2 to 15]

In a similar manner to Example 1, a polyamide acid solution was obtained with 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, and followed by drawing with draw ratios in transverse 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.

[0063]

[Table 1]

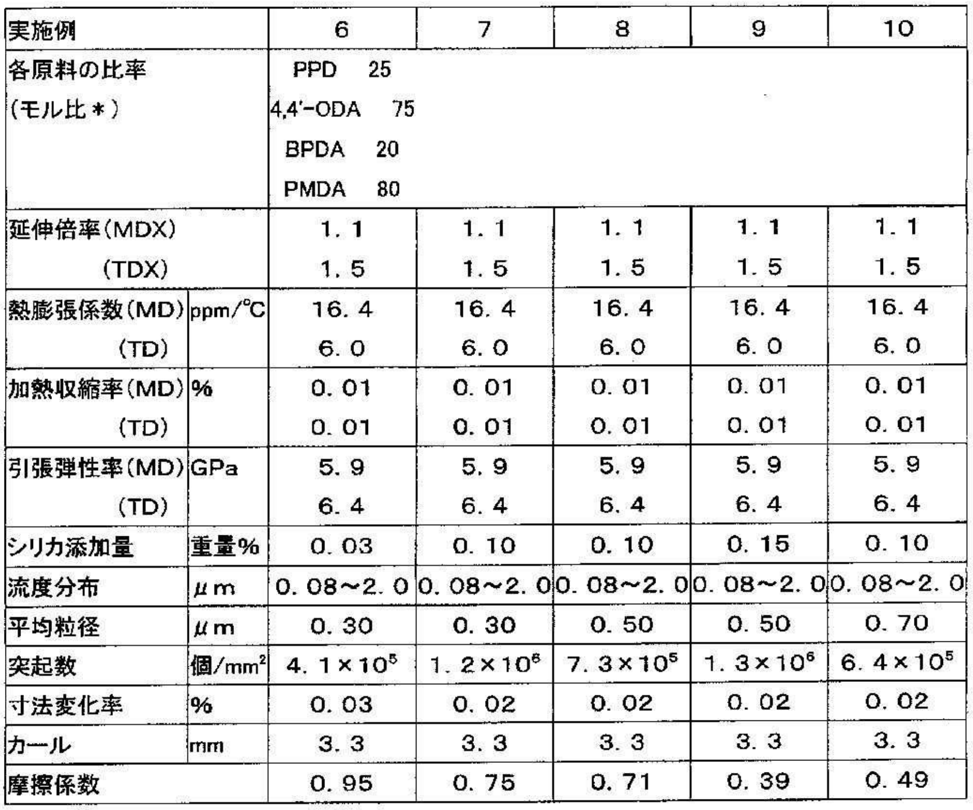


実施例		1	2	3	4	5
各原料の比率 （モル比＊）		PPD 28 4,4'-ODA 72 BPDA 20 PMDA 80				
延伸倍率(MDX)		1. 1	1. 1	1. 1	1. 1	1. 1
(TDX)		1. 5	1. 5	1. 5	1. 5	1. 5
熱膨張係数(MD)	ppm/℃	15. 8	15. 8	15. 8	15. 8	15. 8
(TD)		4. 8	4. 8	4. 8	4. 8	4. 8
加熱収縮率(MD)	%	0. 02	0. 02	0. 02	0. 02	0. 02
(TD)		0. 02	0. 02	0. 02	0. 02	0. 02
引張弾性率(MD)	GPa	6. 0	6. 0	6. 0	6. 0	6. 0
(TD)		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^2	3.2×10^5	9.3×10^5	7.7×10^5	1.2×10^6	5.8×10^5
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		0. 90	0. 72	0. 75	0. 42	0. 51

実施例	Example
各原料の比率（モル比＊）	Ratio of each raw material (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 amount of Silica
流度分布	Particle size distribution
平均粒径	Average particle diameter
突起数	Number of projections
寸法変化率	Dimension change ratio
カール	Curl
摩擦係数	Coefficient of friction
重量％	Percent by weight
個	Number

[0064]

[Table 2]

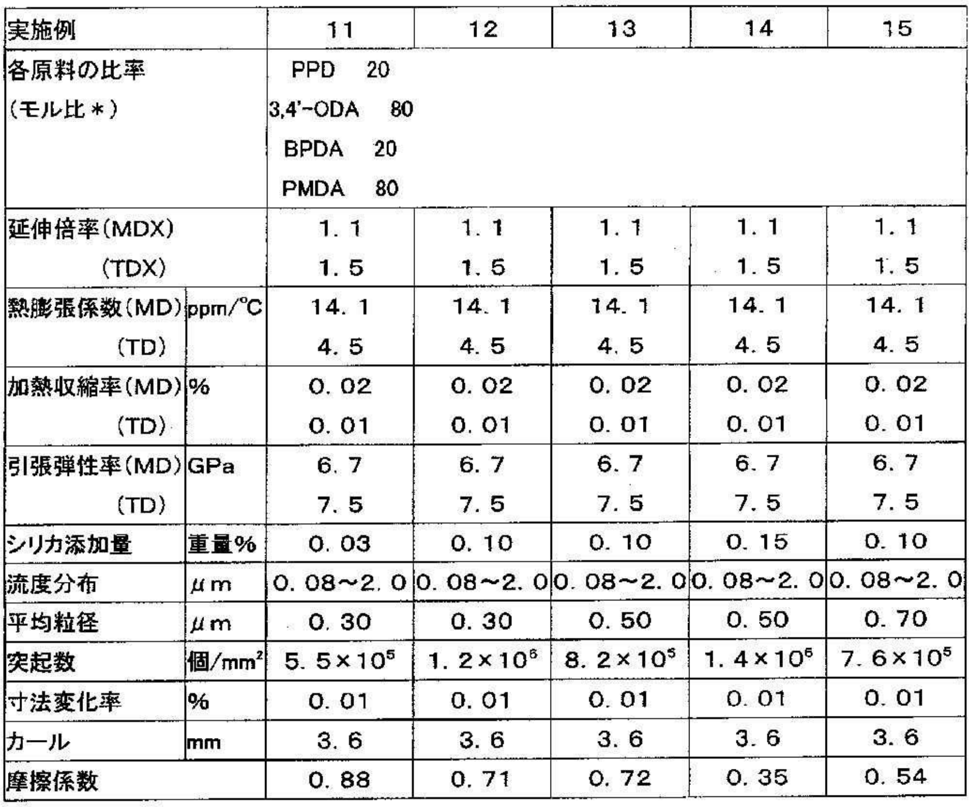


実施例		6	7	8	9	10
各原料の比率 （モル比＊）		PPD 25 4,4'-ODA 75 BPDA 20 PMDA 80				
延伸倍率（MDX） （TDX）		1.1 1.5	1.1 1.5	1.1 1.5	1.1 1.5	1.1 1.5
熱膨張係数（MD） （TD）	ppm/℃	16.4 6.0	16.4 6.0	16.4 6.0	16.4 6.0	16.4 6.0
加熱収縮率（MD） （TD）	%	0.01 0.01	0.01 0.01	0.01 0.01	0.01 0.01	0.01 0.01
引張弾性率（MD） （TD）	GPa	5.9 6.4	5.9 6.4	5.9 6.4	5.9 6.4	5.9 6.4
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^2	4.1×10^5	1.2×10^6	7.3×10^5	1.3×10^6	6.4×10^5
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		0.95	0.75	0.71	0.39	0.49

実施例	Example
各原料の比率（モル比＊）	Ratio of each raw material (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 amount of Silica
流度分布	Particle size distribution
平均粒径	Average particle diameter
突起数	Number of projections
寸法変化率	Dimension change ratio
カール	Curl
摩擦係数	Coefficient of friction
重量％	Percent by weight
個	Number

[0065]

[Table 3]



実施例		11	12	13	14	15
各原料の比率 （モル比＊）		PPD 20 3,4'-ODA 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/℃	14. 1	14. 1	14. 1	14. 1	14. 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^2	5. 5×10^{5}	1. 2×10^{6}	8. 2×10^{5}	1. 4×10^{6}	7. 6×10^{5}
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		0. 88	0. 71	0. 72	0. 35	0. 54

実施例	Example
各原料の比率（モル比＊）	Ratio of each raw material (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 amount of Silica
流度分布	Particle size distribution
平均粒径	Average particle diameter
突起数	Number of projections
寸法変化率	Dimension change ratio
カール	Curl
摩擦係数	Coefficient of friction
重量％	Percent by weight
個	Number

[0066]

* The molar ratios in Table show mol % in all aromatic diamine components and mol % in all aromatic tetracarboxylic components."

[0067]

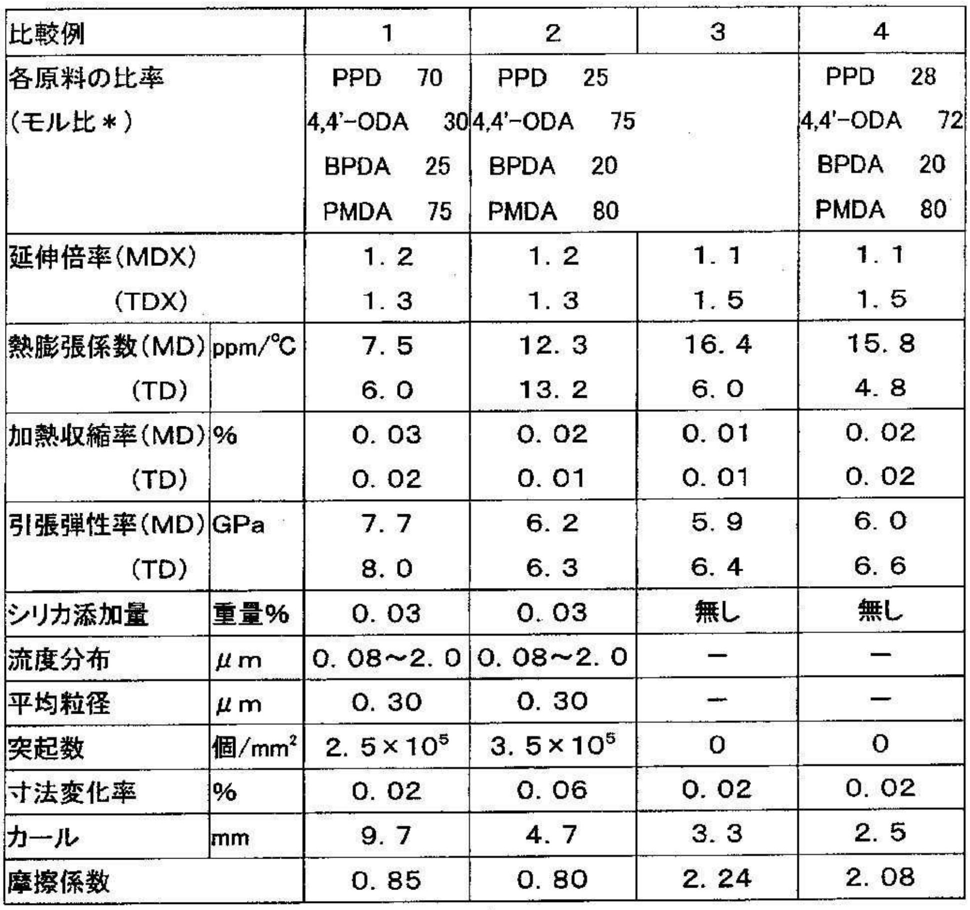
[Comparative Examples 1 to 4]

In a similar procedure to Example 1, a polyamide acid solution was obtained with 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 stretching with draw ratios in transverse and longitudinal directions as shown in Table 4 to evaluate each property of polyimide film obtained in a similar manner to Example 1.　The results are shown in Table 4.

[0068]

[Table 4]



比較例		1	2	3	4
各原料の比率 （モル比＊）		PPD 70 4,4'-ODA 30 BPDA 25 PMDA 75	PPD 25 4,4'-ODA 75 BPDA 20 PMDA 80		PPD 28 4,4'-ODA 72 BPDA 20 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/℃	7. 5	12. 3	16. 4	15. 8
（TD）		6. 0	13. 2	6. 0	4. 8
加熱収縮率（MD）	%	0. 03	0. 02	0. 01	0. 02
（TD）		0. 02	0. 01	0. 01	0. 02
引張弾性率（MD）	GPa	7. 7	6. 2	5. 9	6. 0
（TD）		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	－	－
平均粒径	μm	0. 30	0. 30	－	－
突起数	個/mm^2	2. 5×10^5	3. 5×10^5	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（モル比＊）	Ratio of each raw material (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 amount of Silica
流度分布	Particle size distribution
平均粒径	Average particle diameter
突起数	Number of projections
寸法変化率	Dimension change ratio
カール	Curl
摩擦係数	Coefficient of friction
重量%	Percent by weight
個	Number

無し None
[0069]

* The molar ratios in Table respectively shows mol% in all aromatic diamine components and mol% in all aromatic tetracarboxylic components."

2 Ground A for Invalidation (Noncompliance to Enablement Requirement regarding the patent invention)

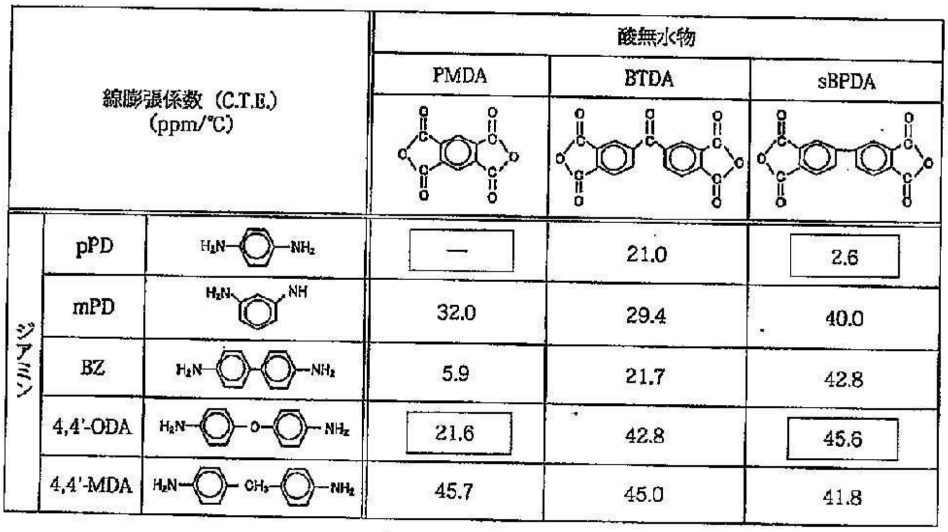
(1) Article 36(4)(i) of the Patent Act (enablement requirement) specifies that the Detailed Description of the Invention should "disclose definitely and sufficiently to the extent that allowed those who have expert knowledge in the technical field to implement the invention." Further, an implementation of an invention in a product invention includes an action of production, use, etc. of the product (Article 2(3)(i) of the Patent Act). Therefore, the product invention requires a specific disclosure in the specification showing that the product can be manufactured; however, even in the absence of such description, it can be said to conform to the enablement requirement if a person skilled in the art could make the product on the basis of the description of the specification and drawings as well as the common technical knowledge as of the filing.

Incidentally, the patent invention includes Claim 1 (a base material for COF), Claim 7 (a copper clad laminate), and Claim 9 (polyimide film) as independent claims. Both Claims 1 and 7 encompass the constituent element of polyimide film specified in Claim 9. Thus all the remaining Claims also encompass the matters specifying the invention specified in Claim 9. Further, in view of the nature of the case, there is considered the enablement requirement of the patent invention 9, which is the matters specifying the invention common to the patent invention.

(2) Common technical knowledge as to the relationship between a constituent component of polyimide and a thermal expansion coefficient

A Evidence A No. 8 has the following description:

"For such improvement on dimension stability, the optimization of CTE (linear expansion coefficient; Trial Decision's note: means thermal expansion coefficient) of the polyimide layer is considered. FIG. 6 shows an example of the relationship between polyimide skeleton (the combination of acidic anhydride and diamines) and CTE. CTE of copper foil is usually 17 to 18 ppm/°C or so, and thus a CTE design adapted to this property is important. CTE depends on the drying method in a cast type even for the same polyimide skeleton, and CTE uses TPI of 40 to 60 ppm/°C in a lamination type. Therefore, a design calculating a final dimension stability is essential to improve the final dimension stability.



線膨張係数（C.T.E.） (ppm/°C)			酸無水物		
			PMDA	BTDA	sBPDA
ジアミン	pPD		—	21.0	2.6
	mPD		32.0	29.4	40.0
	BZ		5.9	21.7	42.8
	4,4'-ODA		21.6	42.8	45.6
	4,4'-MDA		45.7	45.0	41.8

図6　ポリイミド骨格のCTE依存性

線膨張係数	Linear expansion coefficient
酸無水物	Acidic anhydride
ジアミン	Diamine
図６　ポリイミド骨格のＣＴＥ依存性	FIG. 6 CTE dependency of polyimide skeleton

"

B　Evidence A10 has the following description:

"varnish was coated on a glass plate, and dried at 100°C for 1 hour. Subsequently, the obtained film was peeled, and heated at 200°C for 1 hour and at 400°C for 1 hour in nitrogen gas. This resulted in a polyimide film. In the case of polyimide having a methyl or methylene group, the final heating was set to 350°C for 1 hour, instead of 400°C. When a polyamide acid film was heated after peeling from a glass plate, the film was securely fixed with an iron frame so as to prevent shrinkage due to solvent evaporation and imidization. (bidirectionally fixed cure) or held with a spring so as not to prevent curing shrinkage (free cure)." (translation, page 1)

Further, Table 1 titled "thermal expansion coefficient of aromatic polyimide (10^{-5}*unit K^{-1})" (page 2284), which can be recognized as data of thermal expansion coefficient of FIG. 6 of Evidence A No. 8 of the aforesaid item A, discloses that PPD/BPDA has a Bifix (bidirectional fixed cure; Trial Decision's note: when the film is fixed for preventing shrinkage) of 2.6 ppm/°C, and a free (free cure;

Trial Decision's note: when the film was not held by a spring to prevent shrinkage) of 19.0 ppm/°C, and ODA/BPDA has a Bifix of 45.6 ppm/°C and a free of 52.0 ppm/°C, ODA/PMDA has a Bifix of 21.6 ppm/°C and a free of 47.8 ppm/°C.

C　According to the aforesaid items A and B, in the technical field of polyimide film, it is widely known that the use of a compound with a linear structure (e.g. PPD) as a component of polyimide film may result in a small thermal expansion coefficient, whereas the use of a compound with a flexible structure or a degree of rotational freedom (e.g. ODA) may result in a large thermal expansion coefficient. It was thus recognized as a matter of common technical knowledge in the technical field to adjust a thermal expansion coefficient of polyimide film in a wide range depending on a combination of compounds as a diamine component and an acidic anhydride component composed of polyimide.

(3) Characteristics of polyimide film of patent invention 9

A According to the Detailed Description of the Invention of the patent specification of the aforesaid item 1, patent invention 9 relates to a polyimide film excellent in dimension stability and suitable for a base material for a fine pitch circuit, in particular COF wired at a narrow pitch in a film width direction.

When wired at a narrow pitch in a TD direction of a film, greater thermal expansion coefficient of TD results in larger dimensional change between wires in the process of chip mounting bonding, etc. In order to address this problem, when decreasing a thermal expansion coefficient of film as small as to be comparable to silicon, there is a problem of generating strain by a heating process including a bonding process of chip mounting, due to difference in thermal expansion coefficient from copper. In order to address this problem, patent invention 9 has an objective to provide a polyimide film suitable for a base material for a 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 (paragraphs [0002], [0005], and [0007]).

As a means for solving the problem, patent invention 9 allows for suppressing the thermal expansion coefficient in TD direction by proceeding with the orientation of a film toward TD, and has a thermal expansion coefficient in MD close to that of metal, and causes an advantageous effect of providing a polyimide film suitable for a base material for fine pitch circuit such as COF by adopting the constitution of "a thermal expansion coefficient αMD in a machine direction (MD) falling within a range of 10 ppm/°C or more to 20 ppm/°C or less" and "a thermal expansion coefficient αTD in a transverse direction (TD) falling within a range of 3 ppm/°C or more to 7 ppm/°C or less" (paragraphs [0008], [0011], and [0039]). As seen above, it can be said that the feature of patent invention 9 is to adjust the thermal expansion coefficient αMD and the thermal expansion coefficient αTD respectively to different specific ranges.

B Subsequently, the Detailed Description of the Invention of the patent specification discloses that a method of producing polyimide film includes: "thermal imidization," a method of causing a polyamide acid solution obtained by polymerizing an aromatic diamine component and an acidic anhydride component in an organic solvent to be subjected to thermally ring-opening and desolvation to obtain a polyimide film; and "chemical imidization," a method of mixing a cyclization catalyst and a dehydrate with a polyamide 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 chemical imidization is preferable, since the obtained polyimide film may have a lower thermal expansion coefficient (paragraphs [0012] and [0024]).

Further, when a film is manufactured by "chemical imidization," a film with a low thermal expansion coefficient in TD and a thermal expansion coefficient close to that of metal in MD being retained may be manufactured by casting a polyamide acid solution containing a cyclization catalyst and dehydrate on a heated support via a slit die, subjecting to heat ring-closing reaction on the support, peeling from the support as a gel film with self-supporting property, stretching the gel film by 1.01 to 1.9 times at a temperature of 140°C or less in MD, and stretching in TD by 1.1 to 1.5 times the draw ratio in MD (paragraphs [0029] to [0036]).

Further, Examples made a gel film containing four components of PPD/4,4'-ODA and BPDA /PMDA (Examples 1 to 10) or four components of PPD/3,4'-ODA and BPDA/PMDA (Examples 11 to 15), and drew the gel film by 1.1 times in MD and 1.5 times in TD to produce a polyimide film. The obtained film satisfied the thermal expansion coefficient of Invention 9, and had a low "dimension change ratio in TD of a film" and less "curl (a degree of curling of the edge of the sample from a floor when stationary placing a sample in a flat site)" (paragraphs [0058] to [0066]).

Comparative Examples 1 and 2 used the same four components as in Examples 1 to 10 and manufactured a polyimide film with varied ratios of each component and varied draw ratios. The obtained films did not satisfy the thermal expansion coefficient of Invention 9, and as a result, had a low dimension change ratio in TD of a film and a large curl (paragraphs [0067] to [0069]).

C It can be seen from the above description that it is necessary to produce a polyimide film with "a thermal expansion coefficient αMD falling within a range of 10 ppm/°C or more to 20 ppm/°C or less" and "a thermal expansion coefficient αTD falling within a range of 3 ppm/°C or more to 7 ppm/°C or less" in order to achieve the effects of patent invention 9, and patent invention 9 specifies the alternatives of five components used in the examples of "one or more aromatic diamine components selected from the group consisting of PPD, 4,4'-ODA and 3,4'-ODA and one or more acidic anhydride components selected from the group consisting of PMDA and BPDA" as raw materials for achieving the above thermal expansion coefficient.

Consequently, in order to find that the polyimide film of patent invention 9 may be manufactured by a person skilled in the art, it should require that polyimide film having a thermal expansion coefficient specified in invention 9 may be manufactured by combining one or more aromatic diamine components and one or more acidic anhydrides specified in patent invention 9 on the basis of the description of the patent specification and the common technical knowledge of the aforesaid item (2)C.

(4) Two-component polyimide film of PPD/BPDA

A The manufacturing method by thermal imidization

(A) As in the aforesaid items (2)A and B, FIG. 6 of Evidence A No. 8 and Table 1 of Evidence A No. 10 disclose that a thermal expansion coefficient of a two-component polyimide film of PPD/BPDA manufactured by thermal imidization was 2.6 ppm/°C when the film was fixed to prevent shrinkage in heating self-supporting film (Bifix), and 19.0 ppm/°C when the film was held by a spring so as not to prevent shrinkage (Free).

Therefore, it can be understood that a two-component film of PPD/BPDA by thermal imidization may be adjusted to a range of 2.6 to 19.0 ppm/°C depending on whether or not to fix in heating after forming into a self-supporting film.

(B) Description of Evidence B No. 8 to No. 11 and documents discovered by ex officio search

a Evidence B No. 8 generally has the following descriptions, and discloses that a polyimide film of a two-component film of PPD/BPDA by thermal imidization with a thermal expansion coefficient of 11 ppm/°C (Example 1) was obtained.

"[Claim 1] A method of forming a metal thin film on a polyimide film, comprising the steps of: heating a polyimide film with a linear expansion coefficient at

30 to 300°C of $0.4*10^{-5}$ to $2.0*10^{-5}$ cm/cm/°C obtained from any aromatic tetracarboxylic acid component selected from 3,3'4,4'-biphenyltetracarboxylic acid, pyromellitic acid, 3,3'4,4'-benzophenonetetracarboxylic acid, 3,3'4,4'-diphenylethertetracarboxylic acid, and dianhydrides thereof, and an aromatic diamine component selected from a mixture system of p-henylenediamine and diaminodiphenylether, diaminodiphenylmethane, or 2,2-bis(aminophenyl)propane; subjecting the polyimide film to (i) vacuum deposition by heating at 200 to 600°C, or (ii) sputtering by heating at 200 to 450°C in forming a metal thin film to form a metal thin film with a thickness of 200 to 20000 angstroms."

"[0014]

[Examples] The Examples are elaborated hereinafter.

Example 1

Equimolar amounts of 3,3'4,4'-biphenyltetracarboxylic acid dianhydride and p-phenylenediamine were polymerized for 2 hours at 30°C in N-methyl-2-pyrolidone to produce an aromatic polyamide acid with a logarithmic viscosity of 3.5 (concentration: 0.5 g/100 ml solvent; solvent: N-methyl-2-pyrrolidone; measurement temperature: 30°C). The polymerization liquid had a polymer concentration of 25 weight%, and a solution viscosity at 100°C of about 3000 poise. By the use of this polyamide acid solution, a thin film of the aforesaid solution was formed on a support by a solution casting method (film-forming temperature: 90°C), and a solvent was gradually evaporated and removed at a temperature of about 110°C to form a solidified film of aromatic polyamide acid containing about 15 weight% of solvent, and the solidified film was peeled from the support, and subjected to heating at a temperature of about 200°C for 30 minutes, at about 300°C for 15 minutes, at about 450°C for 15 minutes to undergo imidization to form an aromatic polyimide film with a thickness of about 50 μm.

[0015] This aromatic polyimide film has the following physical property values:

Linear expansion coefficient (30 to 300°C): $1.1*10^{-5}$ cm/cm/°C"

b Evidence B No. 9 discloses that the obtained polyimide films had a thermal expansion coefficient of two-component film of PPD/BPDA by thermal imidization of 22.5 ppm/°C (Example 1), 22.3 ppm/°C (Example 2), and 15.4 ppm/°C (Comparative Example 1).

c Evidence B No. 10 discloses that the obtained polyimide films respectively had a thermal expansion coefficient of two-component film of PPD/BPDA by thermal imidization of 12.0 ppm/°C (Example 1), 11.4 ppm/°C (Example 2), 12.3 ppm/°C (Example 3), 21.0 ppm/°C (Example 4), and 19.5 ppm/°C (Example 5).

d Evidence B No. 11 generally has the following descriptions, and discloses that the obtained polyimide films had a thermal expansion coefficient of two-component film of PPD/BPDA by thermal imidization of 18 ppm/°C (MD) and 20 ppm/°C (TD) (Example 1), 16 ppm/°C (MD) and 17 ppm/°C (TD) (Example 2), 10 ppm/°C (MD) and 18 ppm/°C (TD) (Example 3), 11 ppm/°C (MD) and 13 ppm/°C (TD) (Example 4), and 14 ppm/°C (MD) and 12 ppm/°C (TD) (Example 5).

"The scope of claims

1 A dimensionally stable polyimide film made of aromatic polyimide obtained from a polymer solution produced by polymerizing biphenyltetracarboxylic acid and phenylenediamines, wherein said polyimide film has an average linear expansion

coefficient of about $0.1*10^{-5}$ to $2.5*10^{-5}$ cm/cm/°C in a temperature range of about 50°C to 300°C, a ratio of linear expansion coefficients in a longitudinal direction of a film (MD direction) and in a transverse direction of a film (TD direction) (MD/TD) of about 1/5 to 4, and a heat dimension stability shown by a dimensional change of a film at 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^2 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." (page 1, column 1, line 1 to column 2, line 8)

"[Requirement of the present invention and the function and effect]

... found that implementing the drying of a thin film formed from the polymer solution on a surface of a support by two stages: on the support; and under low tension of solidified film after peeling off the support may result in an excellent aromatic polyimide film with a specific low linear expansion coefficient and thermal dimension stability, and thereby completed the Invention." (page 2, column 3, lines 20 to 35)

"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 belt made of metal with smooth surface by a publicly-known solution casting method;

(i) drying the thin film on the support by gradually evaporating a solvent and a produced moisture of thin film 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^2 or less, preferably 80 g/mm^2 or less, and

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

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

When elevating temperature from the aforesaid first stage drying temperature to the aforesaid second stage drying temperature, it is preferable to elevate temperature in a relatively short time, e.g. a temperature elevating speed of 10°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." (page 4, column 7, line 28 to column 8, line 24)
"Example 1
(Preparation of doping liquid)
To a 50 L-volume cylindrical polymerization tank, there were gradually added 3,3'4,4'-biphenyltetracarboxylic acid dianhydride ... paraphenylenediamine to undergo polymerization of both components to produce a polyamide acid.
(Film formation)
This polyamide acid was used for a doping liquid for film formation, and the doping liquid was extruded from ... a slit of T-die mold into a thin film at about 30°C, and the thin film was continuously laid on a smooth metal belt, and ... followed by the first-stage drying, a solidified film body was formed. Subsequently, the solidified film body was peeled from the belt, and cut into a 200 mm square, and one end of the square piece of the solidified film was fixed with a frame by a pin sheet, and uniformly subjected to a load of about 35 g by a dancer across the whole width of the opposite end of the square piece to be held under a low tension condition, and heated to 190°C in about 30 seconds, and subjected to the second-stage drying for 5 minutes at 190°C to form a solidified film, and finally heated at the temperature elevation speed of about 10°C/min with four sides of the square of the solidified film being fixed and maintained with a pin tenter and subjected to drying and heat treatment (imidization) for 30 minutes at 450°C to manufacture a 25 μm-thick aromatic polyimide film." (page 5, column 10, line 16 to page 6, column 11, line 6)
"Example 2

Polyamide acid solution manufactured in a similar manner to Example 1 was used for a doping liquid for film formation, and the doping liquid was extruded into a thin film at about 30°C from a slit (gap between lips: 0.5 mm, lip width: 650 mm) of a T-die mold, and a thin film of the doping liquid was continuously laid on a smooth metal belt, and subjected to the first-stage drying with hot air at about 120°C on the belt to continuously form a solidified film body, which was subsequently peeled off the belt and fed to vertical-type furnace (drying temperature: 180°C) under a low tension (shown in Table 1) by a dancer to pass through in about 4 minutes, and subjected to the second-stage drying to form a solidified film, which was then fed to a high-temperature heating furnace, and subjected to drying, heat treatment and imidization by hot air while gradually elevating temperature of about 250 to 450°C, while holding both ends of the film in a longitudinal direction with a horizontal tenter and moving to continuously form an aromatic polyimide film, and finally wound up into a roll by cooling." (page 6, column 11, lines 19 to 38)
"Example 3

A 25 μm-thick aromatic polyimide film was manufactured in a similar manner to Example 2, except that the tension of solidified film body was set to 40 g/mm^2 in the second-stage drying of a film-forming process." (page 6, column 12, lines 14 to 18)
"Example 4

A 25 μm-thick aromatic polyimide film was manufactured in a similar manner to Example 2, except that the tension was set to about 80 g/mm^2 while holding both ends of a solidified film body in a longitudinal direction with a certain gap in the second-stage drying of a film-forming process." (page 6, column 12, lines 33 to 38)

"Example 5

An aromatic polyimide film was manufactured in a similar manner to Example 2, except for gradually broadening a width of vertical pin tenter in a high-temperature heat treatment furnace up to about 1.04 times a width of entry of the furnace in a high-temperature zone of the furnace." (page 7, column 14, lines 1 to 6)

Further, Comparative Examples 1 and 2 manufactured a film while varying a film-forming condition using the same polyamide acid as Example 1. Table 1 describes a thermal expansion coefficient of the films of Examples 1 to 5 and Comparative Examples 1 to 2.

"

第 1 表

	揮発性成分の含有率(重量%)			第2乾燥工程での固化フイルム状体の張力(g/㎟)		線膨張係数(㎝/㎝/℃)($\times 10^{-5}$)		熱寸法安定性	
	第1乾燥後固化フイルム状体	第2乾燥後固化フイルム	熱処理後芳香族ポリイミドフイルム	MD	TD	MD	TD	MD	TD
実施例1	35	13.0	0.01以下	70	フリー	1.8	2.0	0.18	0.27
比較例1	35	—	0.01以下	—	—	1.1	1.3	0.50	0.47
実施例2	33	17.0	0.01以下	10	フリー	1.6	1.7	0.15	0.09
比較例2	33	—	0.01以下	—	—	0.5	0.8	0.52	0.42
実施例3	33	17.5	0.01以下	40	フリー	1.0	1.8	0.21	0.10
実施例4	33	18.0	0.01以下	80	80	1.1	1.3	0.23	0.18
実施例5	33	18.0	0.01以下	10	フリー	1.4	1.2	0.18	0.47

第１表	Table 1
揮発性成分の含有率（重量%）	Content of volatile components (weight%)
第１乾燥後固化フィルム状態	Solidified film after first drying
第２乾燥後固化フィルム	Solidified film after second drying
熱処理後芳香族ポリイミドフィルム	Aromatic polyimide film after heat treatment
第２乾燥工程での固化フィルム状態の張力	Tension of solidified film after second drying process
線膨張係数	Linear expansion coefficient
熱寸法安定性	Thermal dimension stability
実施例	Example
比較例	Comparative Example

" (page 7, Table 1)

e Further, according to the ex officio investigation, the publication of Patent No. 3346228 (Issued date: Heisei 11 (1999), February 2, hereinafter referred to as "Patent Gazette e") discloses that the obtained polyimide films respectively had thermal expansion coefficients of two-component film of PPD/BPDA by thermal imidization of 9.8 ppm/°C (Example 1), 12.3 ppm/°C (Example 3), and 13.1 ppm/°C (Example 5).

f Similarly, the publication of Patent No. 2573595 (Issued date: Showa 63 (1988), September 14, hereinafter referred to as "Patent Gazette f") generally has the following descriptions, and discloses that a two-component film of PPD/BPDA by thermal imidization with a thermal expansion coefficient of 23 ppm/°C (Comparative Example 1) was obtained:

"Comparative Example 1 In a four-necked flask with 500 ml volume, there was sampled 26.78 g of paraphenylenediamine, followed by addition of 245.00 g of N,N-dimethylacetamide to dissolve. Meanwhile, in a 100 ml eggplant flask, there was sampled 18.22 g of 3,3'4,4'-biphenyltetracarboxylic acid anhydride, which was added in a solid form to the aforesaid paraphenylenediamine solution. Furthermore, 10.00 g of 3,3'4,4'-biphenyltetracarboxylic acid dianhydride attached and remaining in the eggplant flask was poured into a reaction system (four-necked flask) with N,N-dimethylacetamide. After continuous stirring for 3 hours, 15 weight% polyamide acid solution was obtained. The reaction temperature was kept at 5 to 10°C. Note that, in the above operation, 3,3'4,4'-biphenyltetracarboxylic acid dianhydride and the reaction system were placed under dried nitrogen gas flow. Subsequently, this polyamide acid solution was subjected to cast coating on a glass plate, and dried at 100°C for 10 minutes, and a polyamide acid coating was then peeled from the glass plate, and the coating was fixed with a support frame, followed by heating at about 100°C for about 30 minutes, at about 200°C for about 60 minutes, and at about 300°C for about 60 minutes to undergo dehydration and ring-closing, thereby obtaining a 25 micron-thick polyimide film. The properties of this film are shown in Table 1.

Comparative Example 2 To a polyamide acid solution obtained in a similar manner to Comparative Example 1, there were added 4 mol of acetic acid anhydride and 0.5 mol of isoquinoline on the basis of 1 mol amide bonding of polyamide acid solution, and after further stirring, the solution was cast coated on a glass plate and heated at about 100°C for about 10 minutes, at about 250°C for about 10 minutes and at about 350°C for about 5 minutes, followed by peeling from the glass plate to obtain a polyimide film having a thickness of about 25 microns. The properties of this film are shown in Table 1."

(page 4, column 8, lines 4 to 31)

Further, Table 1 respectively describes a linear expansion coefficient of Comparative Example 1 (thermal imidization) of $2.3*10^{-5}$ (23 ppm/°C), and a linear expansion coefficient of Comparative Example 2 (chemical imidization) of $2.0*10^{-5}$ (20 ppm/°C).

(C) According to the aforesaid item (B), two-component film of PPD/BPDA by thermal imidization has a thermal expansion coefficient of approximately 10 to 20 ppm/°C in Evidence B Nos. 8 to 11 and Patent Gazette e, and 23 ppm/°C in Comparative Example 1 of Patent Gazette f.

In particular, as aforesaid (B)d, Evidence B No. 11 discloses that the tension in MD and TD applied to a solidified film body in drying is adjusted to control a thermal expansion coefficient to 1 to 25 ppm/°C, and to control a ratio of thermal expansion coefficient (MD/TD) to about one-fifth to four (the Claims), and discloses in Example 3 the manufacture of a film having isotropic thermal expansion coefficients of 10 ppm/°C (MD) and 18 ppm/°C (TD). Further, Example 5 manufactured a film in a similar manner to Example 2, except that Example 5 gradually broadened a width of a horizontal pin tenter in a high-temperature heat treatment furnace so that the width may become about 1.04 times a width of the entrance of the furnace in a high-temperature zone of the furnace. As a result, Example 5 yielded smaller thermal expansion coefficients in both directions of 14 ppm/°C (MD) and 12 ppm/°C (TD), as compared to 16 ppm/°C (MD) and 17 ppm/°C (TD) of Example 2.

Consequently, it can be said that it was a well-known art as of the filing to adjust a thermal expansion coefficient to 2.6 to 20 ppm/°C or about 23 ppm/°C by the adjustment of "apparent stretching operation" of fixing a film (Bifix) or holding a film with a spring (Free) during heating as described in Evidence A No. 8 and No. 10 in manufacturing a two-component film of PPD/BPDA by thermal imidization, or by the adjustment of tension applied to the film when drying in the respective MD and TD directions as described in Evidence B No. 11, or by broadening a width of a lateral pin tenter, etc. during heat treatment.

Therefore, it can be said that it was possible for a person skilled in the art to adjust "a thermal expansion coefficient αMD falling within a range of 10 ppm/°C or more to 20 ppm/°C or less" and "a thermal expansion coefficient αTD falling within a range of 3 ppm/°C or more to 7 ppm/°C or less," which falls within a range of thermal expansion coefficient of 2.6 to 20 ppm/°C for two-component polyimide film of PPD/BPDA.

B Manufacturing method by chemical imidization

As aforesaid item A(B)f, Patent Gazette f discloses in Comparative Example 2 that the two-component polyimide film of PPD/BPDA obtained by chemical imidization has a thermal expansion coefficient of 20 ppm/°C. Therefore, it is recognized that a polyimide film obtained by chemical imidization may also have a thermal expansion coefficient approximately comparable to that of the film obtained by thermal imidization.

Further, as aforesaid item 1, the patent specification specifically discloses that a film obtained by chemical imidization may decrease thermal expansion coefficient by subjecting to drawing. Thus it was feasible for a person skilled in the art to manufacture a two-component polyimide film of PPD/BPDA obtained by chemical imidization and subject the film to appropriate stretching in MD and TD to adjust a range of thermal expansion coefficient of patent invention 9.

C Demandant's allegation

(A) The demandant argues that Evidence B No. 2 to No. 5 (Demandant's product) and Evidence B No. 8 to No. 11, on which the demandee relies as a ground of the fact that it is common technical knowledge to realize a thermal expansion coefficient of the two-component film of PPD/BPDA of 10 to 20 ppm/°C, are manufactured by thermal imidization that Demandant has originally developed, and manufacturing such a film requires much know-how that is not described in the patent application filed by Demandant, and thus the thermal expansion coefficient of the demandant's product and the patent application cannot be a ground of the common technical knowledge as of the filing.

Demandant only argues, however, that Demandee could not manufacture a polyimide film having a thermal expansion coefficient on the basis of the description of each of Evidence B No. 8 to No. 11, because manufacturing such a film requires much know-how that is not described in the patent application filed by Demandant, and argues or demonstrate no specific contents thereof. Further, each publication of the above Evidence B No. 8 to No. 11 describes a detailed manufacturing condition to manufacture a polyimide film having a thermal expansion coefficient of 10 to 20 ppm/°C, and thus there is no obstacle to see the contents of these publications as common technical knowledge as of the filing.

Therefore, the above argument presented by Demandant is not acceptable.

(B) Demandant argues that thermally imidized polyimide film has a property not suitable for stretching as compared to a chemically imidized polyimide film according to Evidence A No. 13 (paragraphs [0018] and [0019]), and stretching becomes difficult if the solid content concentration becomes 60 weight% or more due to the progress of the drying of gel film obtained by thermal imidization, and if stretching is performed by 1.05 times in a running direction, the subsequent stretching in a width direction becomes impossible due to gel film breakage (paragraph [0026]), and all of Evidence B Nos. 8 to 11 that the demandee cites are imidized by thermal imidization, and further the solid content is 60% or more, and thus the application of stretching ratio of 1.1 to 1.5 times to MD in the examples of the patent specification may not result in a range of thermal expansion coefficient of patent invention 9.

Further, consideration is given to the demandant's argument; Evidence B No. 13 has the following description: "[0018] The thermal ring-closing method has an advantage of not requiring a ring-closing catalyst and a desiccant, but requires long-term heat drying on a support to make a self-supporting gel film, and the solid content of the gel film peeled from the support is too high for implementing stable stretching. Thus it is not suitable for the use in the present invention."
"[0019] On the other hand, a chemical ring-closing method requires the facility to cause a ring-closing catalyst and a desiccant to include, but allows for the manufacture of a self-supporting gel film in a short time, and furthermore the solid content of the gel film peeled from the support may be maintained low, and thus allows for a high degree of stretching, and therefore the method is a manufacturing method of polyimide film suitable for the present invention. A manufacturing method of polyimide film close to the heat ring-closing method with lower contents of ring-closing catalyst and less desiccant can be said to be a chemical ring-closing method since it contains a ring-closing catalyst and a desiccant." Subsequent description relates to a manufacturing method on the basis of the chemical ring-closing method. Therefore, it is obvious that the description of paragraph [0026] "the stretching is greatly affected by the solid content concentration, and the stretching becomes difficult if the solid content becomes 60 weight% or more due to the progress of the drying of gel film. The stretching in a subsequent width direction becomes impossible due to the breakage of gel film if stretched in a running direction by 1.05 times. Therefore, it is necessary to adjust the solid content of the gel film molded and peeled from the support to 50 weight% or less, ..." is also a description of the manufacturing method of polyimide film by chemical imidization of the invention of Evidence A No. 13.

Therefore, the description of this paragraph [0026] is not applied to the corresponding parts of Evidence B No. 8 to No. 11 regarding the manufacturing method of polyimide film by thermal imidization as explained in the aforesaid item A(B), and further Evidence B No. 8 to No. 11 realize a thermal expansion coefficient of two-component polyimide film of PPD/BPDA of thermal imidization of 10 to 20 ppm/°C. Therefore, the above argument by the demandant is not acceptable.

(5) Three-component polyimide film of PPD/PMDA/BPDA

As aforesaid item (2)C, it was recognized as a matter of common technical knowledge in the technical field to adjust a thermal expansion coefficient of polyimide film in a wide range depending on a combination of compounds serving as a diamine component and an acidic anhydride component composed of polyimide. As aforesaid item (4), a person skilled in the art could adjust "a thermal expansion coefficient αMD falling within a range of 10 ppm/°C or more to 20 ppm/°C or less" and "a thermal expansion coefficient αTD falling within a range of 3 ppm/°C or more to 7 ppm/°C or less" for a two-component polyimide film of PPD/BPDA. Therefore, it can be said that it was feasible for a person skilled in the art to adjust "a thermal expansion coefficient αMD falling within a range of 10 ppm/°C or more to 20 ppm/°C or less" and "a thermal expansion coefficient αTD falling within a range of 3 ppm/°C or more to 7 ppm/°C or less" for a three-component polyimide film of PPD/PMDA/BPDA using PMDA and BPDA as an acidic anhydride component, by adjusting, e.g., a mixing ratio of PMDA and BPDA.

(6) Three-component polyimide film of PPD/4,4'-ODA/BPDA

A As aforesaid item (2)C, it was recognized as a matter of common technical knowledge in the technical field to adjust a thermal expansion coefficient of polyimide film in a wide range depending on a combination of compounds as a diamine component and an acidic anhydride component composed of polyimide, and it is widely known that the use of a compound with a linear structure (e.g. PPD) may result in a small thermal expansion coefficient, whereas the use of a compound with a flexible structure or a degree of rotational freedom (e.g. ODA) may result in a large thermal expansion coefficient. Evidence A No. 8 and No. 10 disclose those having a thermal expansion coefficient of a two-component polyimide film of PPD/BPDA obtained by thermal imidization of 2.6 ppm/°C as well as those having a thermal expansion coefficient of a two-component polyimide film of 4,4'-ODA/BPDA obtained by thermal imidization of 45.6 ppm/°C.

B Consequently, it can be said that it was feasible for a person skilled in the art to adjust "a thermal expansion coefficient αMD falling within a range of 10 ppm/°C or more to 20 ppm/°C or less" and "a thermal expansion coefficient αTD falling within a range of 3 ppm/°C or more to 7 ppm/°C or less," which falls within a range of thermal expansion coefficient of 2.6 to 45.6 ppm/°C for a three-component polyimide film of PPD/4,4'-ODA/BPDA in which PPD and 4,4'-ODA were used in combination as a diamine component, for example, by adjusting a mixing ratio of PPD to 4,4'-ODA.

(7) Four-component polyimide film of PPD/4,4'-ODA/PMDA/BPDA, four component polyimide film of PPD/4,4'-ODA/3,4'-ODA/BPDA and five-component polyimide film of PPD/4,4'-ODA/3,4'-ODA/PMDA/BPDA

As aforesaid item (2)C, it was recognized as a matter of common technical knowledge in the technical field to adjust a thermal expansion coefficient of a polyimide film in a wide range depending on a combination of compounds as a diamine component and an acidic anhydride component composed of polyimide. As aforesaid item (6), a person skilled in the art could adjust "a thermal expansion coefficient αMD falling within a range of 10 ppm/°C or more to 20 ppm/°C or less" and "a thermal expansion coefficient αTD falling within a range of 3 ppm/°C or more to 7 ppm/°C or less" for a three-component polyimide film of PPD/4,4'-ODA/BPDA. Therefore, it can further be said that, regarding a four-component polyimide film of PPD/4,4'-ODA/PMDA/BPDA in which PMDA and BPDA are used in combination as an acidic anhydride component, a four-component polyimide film of PPD/4,4'-ODA/3,4'-

ODA/BPDA in which PPD, 4,4'-ODA, and 3,4'-ODA are used in combination as a diamine component, or a five-component polyimide film of PPD/4,4'-ODA/3,4'-ODA/PMDA/BPDA in which PMDA and BPDA are used in combination as an acidic anhydride component, while PPD, 4,4'-ODA, and 3,4'-ODA are used in combination as a diamine component, for example, the adjustment of the mixing ratio of PPD to 4,4'-ODA and the adjustment of the mixing ratio of PMDA to BPDA or the mixing ratio of 3,4'-ODA would allow a person skilled in the art to adjust "a thermal expansion coefficient αMD falling within a range of 10 ppm/°C or more to 20 ppm/°C or less" and "a thermal expansion coefficient αTD falling within a range of 3 ppm/°C or more to 7 ppm/°C or less."

Actually, the patent specification specifically discloses in Examples 1 to 10 a manufacturing condition such as a ratio of these raw materials and a draw ratio for a four-component polyimide film of PPD/4,4'-ODA/PMDA/BPDA, and thus it can be said that at least a part in the vicinity of these examples was feasible for a person skilled in the art.

(8) Three-component polyimide film of PPD/3,4'-ODA/BPDA and four-component polyimide film of PPD/3,4'-ODA/PMDA/BPDA

Considering the three-component polyimide film of PPD/3,4'-ODA/BPDA, as in the aforesaid item (4), it was feasible for a person skilled in the art to adjust "a thermal expansion coefficient αMD falling within a range of 10 ppm/°C or more to 20 ppm/°C or less" and "a thermal expansion coefficient αTD falling within a range of 3 ppm/°C or more to 7 ppm/°C or less" for a two-component polyimide film of PPD/BPDA. Therefore, it can be said that it was feasible for a person skilled in the art to adjust "a thermal expansion coefficient αMD falling within a range of 10 ppm/°C or more to 20 ppm/°C or less" and "a thermal expansion coefficient αTD falling within a range of 3 ppm/°C or more to 7 ppm/°C or less" for a three-component polyimide film of PPD/3,4'-ODA/BPDA in which PPD and 3,4'-ODA were used in combination as a diamine component, for example, by adjusting a mixing ratio of PPD to 3,4'-ODA.

Further, considering the four-component polyimide film of PPD/3,4'-ODA/PMDA/BPDA, as aforementioned, it was feasible for a person skilled in the art to adjust "a thermal expansion coefficient αMD falling within a range of 10 ppm/°C or more to 20 ppm/°C or less" and "a thermal expansion coefficient αTD falling within a range of 3 ppm/°C or more to 7 ppm/°C or less" for a three-component polyimide film of PPD/3,4'-ODA/BPDA. Therefore, it can further be said that it was feasible for a person skilled in the art to adjust "a thermal expansion coefficient αMD falling within a range of 10 ppm/°C or more to 20 ppm/°C or less" and "a thermal expansion coefficient αTD falling within a range of 3 ppm/°C or more to 7 ppm/°C or less" for a four-component polyimide film of PPD/3,4'-ODA/PMDA/BPDA using PMDA and BPDA in combination as an acidic anhydride component, by adjusting, for example, a mixing ratio of PPD and 3,4'-ODA, and further adjusting a mixing ratio of PMDA and BPDA. Further, as in the aforesaid item (5), a person skilled in the art could adjust "a thermal expansion coefficient αMD falling within a range of 10 ppm/°C or more to 20 ppm/°C or less" and "a thermal expansion coefficient αTD falling within a range of 3 ppm/°C or more to 7 ppm/°C or less" for a three-component polyimide film of PPD/ODA/BPDA. Therefore, it can also be said that it was feasible for a person skilled in the art to adjust "a thermal expansion coefficient αMD falling within a range of 10 ppm/°C or more to 20 ppm/°C or less" and "a thermal expansion coefficient αTD falling within a range of 3 ppm/°C or more to 7 ppm/°C or less" for a four-component polyimide film of PPD/3,4'-ODA/PMDA/BPDA using PPD and 3,4'-ODA in combination as a diamine component, by adjusting, for example, a mixing ratio of PMDA and BPDA, and further adjusting a mixing ratio of PPD and 3,4'-ODA.

Actually, the patent specification specifically discloses in Examples 11 to 15 a manufacturing condition such as the ratio of these raw materials and draw ratio for a four-component polyimide film of PPD/3,4'-ODA/PMDA/BPDA, and thus it can be said that at least a part in the vicinity of these examples was feasible for a person skilled in the art.

(9) Two-component polyimide film of 4,4'-ODA/BPDA

A Evidence A No. 8 and No. 10 disclose that a polyimide film manufactured by 4,4'-ODA/BPDA by thermal imidization has a thermal expansion coefficient of 45.6 ppm/°C even in a condition of Bifix where a thermal expansion coefficient gets lowered.

The numerical values of thermal expansion coefficient described in each of the above documents are quite beyond the range of thermal expansion coefficient of patent invention 9.

B In general, it is known that a thinner film thickness may result in a smaller thermal expansion coefficient (Evidence A No. 9, translation, page 1). It can be said that a thinner film thickness may further decrease thermal expansion coefficient in a polyimide film obtained by thermal imidization like Evidence A No. 8 and No. 10, but the patent specification fails to describe to what extent it may be decreased.

Further, in the case of a polyimide film obtained by thermal imidization, the solid content becomes too large, and the stretching becomes difficult (Evidence A No. 13, paragraph [0018]). Further, even if it is possible to stretch by about 1.04 times like Example 5 of Evidence B No. 11, there is no reason to find it possible to decrease the thermal expansion coefficient of 45.6 ppm/°C to a level of 3 to 7 ppm/°C, and the patent specification fails to describe any specific reason.

Furthermore, even if it is possible to manufacture two-component polyimide film of 4,4'-ODA/BPDA by chemical imidization and adjust a film thickness and a draw ratio, etc., there is no reason to find it possible to decrease the thermal expansion coefficient to a level of 3 to 7 ppm/°C, and the patent specification fails to describe any specific reason.

In this regard, the demandee argues that Evidence A No. 9 discloses that a thermal expansion coefficient finally obtained for a polyimide film is greatly affected by many conditions such as solvent content, temperature condition, and draw rate in stretching as well as draw ratio, and is also affected by a film thickness, and thus it cannot be asserted that the numerical range of thermal expansion coefficient of patent invention 9 may be realized for a two-component system of 4,4'-ODA/BPDA only on the basis of data of Evidence A No. 8.

The patent specification fails to provide any guidance, however, as to how to control solvent content, temperature condition, draw rate, etc. to decrease a thermal expansion coefficient to a level of patent invention 9.

Regardless of the fact that, inherently, the demandee or the applicant has burden of proof about establishment of the enablement requirement, the demandee fails to show any concrete manufacturability of a two-component polyimide film of 4,4'-ODA/BPDA satisfying the range of thermal expansion coefficient of patent invention 9.

Therefore, it cannot be said that it was possible for a person skilled in the art to adjust thermal expansion coefficient to a range of patent invention 9 for a two-component film of 4,4'-ODA/BPDA in view of the description of the patent specification and the common technical knowledge as of the filing. (binding effects of the above court decision of cancellation)

(10) Two-component polyimide film of 4,4'-ODA/PMDA

A Evidence A No. 8 and No. 10 disclose that a polyimide film manufactured from 4,4'-ODA/PMDA by thermal imidization has a thermal expansion coefficient of 21.6 ppm/°C even in a condition of Bifix where a thermal expansion coefficient gets lowered.

Further, Evidence A No. 13 discloses that Examples 1 to 3 and Comparative Examples 1 to 3 had an average thermal expansion coefficient of 27.5 to 40.0 ppm/°C as shown in Table 1 of Evidence A No. 13 by changing the conditions such as draw ratio and the presence or the absence of nip roll in manufacturing a polyimide film by chemical imidization from 4,4'-ODA/PMDA (paragraphs [0044], [0047] to [0059], [Table 1]).

The numerical values of thermal expansion coefficient described in each of the above documents are quite beyond the range of thermal expansion coefficient of patent invention 9.

B As in the aforesaid item (9), in general, it is known that a thinner film thickness may result in a smaller thermal expansion coefficient. It can be said that a thinner film thickness may further decrease thermal expansion coefficient in a polyimide film obtained by thermal imidization, but the patent specification fails to describe to what extent it may be decreased.

Further, in the case of polyimide film obtained by thermal imidization, the solid content becomes too large, and the stretching becomes difficult. Further, there is no reason to find it possible to decrease the thermal expansion coefficient of 21.6 ppm/°C to a level of 3 to 7 ppm/°C, and the patent specification fails to describe any specific reason.

Furthermore, even if two-component polyimide film of 4,4'-ODA/PMDA were manufactured by chemical imidization and the film thickness, draw ratio, etc. should be adjusted, there is no reason to find it possible to decrease the thermal expansion coefficient to a level of 3 to 7 ppm/°C, and the patent specification fails to describe any specific reason.

Regardless of the fact that, inherently, the demandee or the applicant has burden of proof about establishment of the enablement requirement, the demandee fails to show any concrete proof about manufacturability of a two-component polyimide film of 4,4'-ODA/PMDA satisfying the range of thermal expansion coefficient of patent invention 9.

Therefore, it cannot be said that it was possible for a person skilled in the art to adjust thermal expansion coefficient to a range of patent invention 9 for a two-component film of 4,4'-ODA/PMDA in view of the description of the patent specification and the common technical knowledge as of the filing.

(11) Three-component polyimide film of 4,4'-ODA/PMDA/BPDA

As stated in the aforesaid items (9) to (10), since thermal expansion coefficient values of the two-component film of 4,4'-ODA/BPDA and the two-component film of 4,4'-ODA/PMDA is quite beyond the range of that of patent invention 9, it cannot be said that it was possible for a person skilled in the art to adjust the thermal expansion coefficient to the range of patent invention 9 on the basis of those films. Consequently, it cannot be said that it was possible for a person skilled in the art to adjust "a thermal expansion coefficient αMD falling within a range of 10 ppm/°C or more to 20 ppm/°C or less" and "a thermal expansion coefficient αTD falling within a range of 3 ppm/°C or more to 7 ppm/°C or less" for a three-component polyimide film of 4,4'-ODA/PMDA/BPDA using PMDA and BPDA in combination as an acidic anhydride component, no matter how, e.g., a mixing ratio of PMDA and BPDA is adjusted.

(12) Polyimide film where the other diamine component and acidic anhydride component are combined with each other

Considering a polyimide film where a diamine component and an acidic anhydride component other than the aforesaid items (4) to (11) are combined with each other (hereinafter simply referred to as "the other polyimide film") regarding the other polyimide film (e.g. a two-component polyimide film of PPD/PMDA, a two-component polyimide film of 3,4'-ODA/PMDA, a two-component polyimide film of 3,4'-ODA/BPDA, a three-component polyimide film of PPD/4,4'-ODA/PMDA, a three-component polyimide film of PPD/3,4'-ODA/PMDA, a three-component polyimide film of 4,4'-ODA/3,4'-ODA/BPDA, or a three-component polyimide film of 4,4'-ODA/3,4'-ODA, etc.), the patent specification fails to disclose the numerical ranges of thermal expansion coefficient of these polyimide films. First of all, these values are unknown even in view of the submitted evidences.

Consequently, it cannot be said that it was possible for a person skilled in the art to adjust "a thermal expansion coefficient αMD falling within a range of 10 ppm/°C or more to 20 ppm/°C or less" and "a thermal expansion coefficient αTD falling within a range of 3 ppm/°C or more to 7 ppm/°C or less" on the basis of those polyimide films, even if they are stretched or thinned, or the mixing ratio of a diamine component and an acidic anhydride component used is adjusted, since their thermal expansion coefficient is unknown.

Regardless of the fact that, inherently, the demandee as an applicant should establish the enablement requirement, the demandee fails to specifically establish the possibility of the manufacture of another polyimide film satisfying the range of thermal expansion coefficient of patent invention 9.

Therefore, it cannot be said that it was possible for a person skilled in the art to adjust thermal expansion coefficient to a range of patent invention 9 for another polyimide film in view of the description of the patent specification and the common technical knowledge as of the filing.

(13) Summary

As seen above, it cannot be said that a person skilled in the art could manufacture the polyimide film of the aforesaid items (9) to (12) on the basis of the patent specification and the common general knowledge as of the filing, and thus the polyimide film of patent invention 9 encompasses ones that do not conform to the enablement requirement. Consequently, patent inventions 1 to 8, 10, and 11 also encompass ones that were not feasible, and thus it cannot be said that the Detailed Description of the Invention is described definitely and sufficiently to the extent that allows a person skilled in the art to implement the patent invention.

Therefore, it cannot be said that the patent invention conforms to the enablement requirement, and thus the Ground A for invalidation presented by the demandant has a point.

3 Ground B for Invalidation (Noncompliance to Supporting Requirement regarding the Invention)

(1) Article 36(6)(i) of the Patent Act requires that an invention for which a patent is sought should be described in the Detailed Description of the Invention with regard to the scope of the claims, and eliminates the grant of exclusive right with overly broad scope as compared to the technical matter disclosed in the Detailed Description of the Invention. Therefore, the determination of whether or not the recitation of the Claims might comply with the support requirement should follow the steps of: comparing the recitation of the Claims and the descriptions of the Detailed Description of the Invention; and considering whether or not the invention recited in the Claims might fall within the scope in which a person skilled in the art could recognize that a problem to be solved by the invention might be solved by the description of the Detailed Description of the Invention, or considering whether or not the invention recited in the Claims might fall within the scope in which a person skilled in the art could recognize without such description or suggestion in view of common technical knowledge as of the filing that the problem to be solved by the invention might be solved.

(2) Accordingly, comparing the recitation of the Claims and the descriptions of the Detailed Description of the Invention, the recitation of the Claims of patent invention 9 is as described in the aforesaid [Claim 9] of No. 2. Further, as in the aforesaid item 2(3), the Detailed Description of the Invention of the patent specification discloses that patent invention 9 has an object to provide a polyimide film suitable for a base material for a 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 (paragraphs [0002], [0005], and [0007]); as a means for solving the problem, patent invention 9 allows for suppressing the thermal expansion coefficient in TD direction by proceeding with the orientation of a film toward TD, and has a thermal expansion coefficient in MD close to that of metal, and causes an advantageous effect of providing a polyimide film suitable for a base material for a fine pitch circuit such as COF by adopting the constitution of "a thermal expansion coefficient αMD falling within a range of 10 ppm/°C or more to 20 ppm/°C or less" and "a thermal expansion coefficient αTD falling within a range of 3 ppm/°C or more to 7 ppm/°C or less" (paragraphs [0008], [0011], and [0039]), Examples made a gel film containing four components of PPD/4,4'-ODA and BPDA /PMDA (Examples 1 to 10) or four components of PPD/3,4'-ODA and BPDA/PMDA (Examples 11 to 15) by chemical imidization, and drew the gel film by 1.1 times in MD and 1.5 times in TD to produce a polyimide film. The obtained film satisfied the thermal expansion coefficient of Invention 9, and had a low "dimension change ratio in TD of a film" and less "curl" (paragraphs [0058] to [0066]), and Comparative Examples 1 and 2 used the same four components as Examples 1 to 10 and manufactured a polyimide film by chemical

imidization with varied ratios of each component and varied draw ratios. The obtained film did not satisfy the thermal expansion coefficient of patent invention 9, and as a result, had a large dimension change ratio in TD of a film and a large curl (paragraphs [0067] to [0069]).

Further, a person skilled in the art may manufacture the polyimide films of the aforesaid items 2(4) to (8) on the basis of the descriptions of the patent specification and the common technical knowledge as of the priority date. Consequently, patent invention 9 according to the constitution of polyimide film of the aforesaid items 2(4) to (8) falls within a range that allows a person skilled in the art to recognize from the description of the patent specification and the common technical knowledge as of the filing that the above problem may be solved. Thus it conforms to the supporting requirement.

However, a person skilled in the art fails to have a specific reason to manufacture a polyimide film of the aforesaid items 2(9) to (12) on the basis of the descriptions of the patent specification and the common technical knowledge as of the filing. Consequently, it cannot be said that patent invention 9 according to the constitution of polyimide film of the aforesaid items 2(9) to (12) falls within a range that allows a person skilled in the art to recognize from the description of the patent specification and the common technical knowledge as of the filing that the above problem may be solved. Thus it does not conform to the supporting requirement.

(3) Summary

As seen above, patent invention 9 according to the constitution of polyimide film of the aforesaid items 2(9) to (12) should not conform to the supporting requirement, and thus the polyimide film of patent invention 9 encompasses those that do not conform to the supporting requirement. Consequently, patent inventions 1 to 8, 10, and 11 also encompass those that do not conform to the supporting requirement, and thus it cannot be said that an invention for which a patent is sought is described in the Detailed Description of the Invention with regard to the patent invention.

Therefore, it cannot be said that the patent invention conforms to the supporting requirement, and thus the Ground B for invalidation presented by the demandant has a point.

No. 6 Closing

For the above reasons, patent inventions 1 to 11 should be invalidated.

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

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

November 4, 2015

Chief administrative judge: SUTO, Yasuhiro
Administrative judge: ONODERA, Tsutomu
Administrative judge: MAEDA, Hiroyuki