

DESCRIPTION

RESIN COMPOSITION FOR RETARDATION THIN FILM, COLOR FILTER
SUBSTRATE FOR LIQUID CRYSTAL DISPLAY DEVICE, LIQUID CRYSTAL
5 DISPLAY DEVICE, AND METHOD FOR PRODUCTION OF COLOR FILTER
SUBSTRATE FOR LIQUID CRYSTAL DISPLAY DEVICE HAVING
RETARDATION THIN FILM ATTACHED THERETO

Technical Field

10 [0001]

The present invention relates to a resin composition for retarder, color filter substrate for LCDs (liquid crystal displays), LCD, and to a process for producing a color filter substrate for LCDs having a retarder.

Background Art

15 [0002]

LCDs are now used in a wide variety of uses such as notebook PCs, mobile terminals, desktop monitors and digital cameras, exploiting the characteristics such as light weight, thinness and low power consumption. For the LCDs, increase in the viewing-angle is demanded with the enlargement of the display size and development
20 of the monitor uses.

[0003]

The reason why the viewing-angle of the LCDs is narrower than those of the light-emitting type cathode ray tubes (CRTs) and plasma displays is that the difference in retardation of the liquid crystal layer, resulted from the difference in the
25 direction of travel of light, influence the transmission intensity since the LCDs generally have a structure in which a liquid crystal layer is sandwiched between two polarizing films. That is, in an oblique direction, retardation is large so that an

incident linear polarized light becomes an elliptically polarized light and so the light leakage in the dark condition is increased, which results in the decrease in the contrast.

[0004]

5 Therefore, in order to reduce the decrease in the contrast in oblique directions, it is effective to use a retarder for compensating the retardation of the liquid crystal. At present, in twisted nematic liquid displays, the viewing-angle is increased by attaching a viewing-angle-increasing film as a retarder, which film is made of a discotic liquid crystal.

10 [0005]

On the other hand, VA (Vertical Alignment) method and IPS (In-plane Switching) method which are new liquid crystal display methods aiming at increase in the viewing-angle have been developed.

[0006]

15 In the VA method, biaxially oriented retarder films are used for the purpose of further increasing the viewing-angle. However, production of these films is not easy, and since orientation or stretching step is indispensable, the process is complicated.

[0007]

20 To solve this problem, a method for increasing the viewing-angle of LCDs by providing a polyimide retarder which has an optically negative uniaxial anisotropy, whose optical axis is vertical or almost vertical to the surface of the retarder has been proposed (Patent Literature 1).

[0008]

25 The reason why polyimides function as a retarder is that the refractive index in the direction of the main chain is larger than that in the direction perpendicular to the main chain since they have aromatic rings, aromatic heterocycles or the like in the

direction of the main chain of the macromolecules, and so they show a large birefringence, and that since the molecular chains are likely to be oriented in the direction parallel to the substrate, difference in the refractive index (birefringence as a film) between the direction of thickness of the film and the direction parallel to the film plane is generated.

[0009]

However, the above-described polyimide retarders have a high optical absorption due to the aromatic molecular structure and so the transparency is not sufficient, so that when they are used as a retarder, the white areas in the LCDs become yellowish, which is problematic on the quality of the displayed image.

[0010]

To improve the transparency of the polyimides, it has been proposed to interfere the intramolecular conjugation and formation of charge-transfer complex by introducing into the polyimides an acid component or diamine component having a nonaromatic group such as an alicyclic group (Patent Literatures 2-4). Further, it has been proposed to apply a polyimide resin characterized by high transparency and low birefringence, which has an improved transparency and decreased orientational birefringence and stress birefringence, to optical elements (Patent Literatures 5 to 7).

[0011]

On the other hand, as a polyimide material suited for a retarder, a polyimide material having a high transparency and high birefringence is demanded.

[0012]

Patent Literature 1: JP 2001-290023 A

Patent Literature 2: JP 7-56030 A

Patent Literature 3: JP 9-73172 A

Patent Literature 4: JP 2002-161136 A

Patent Literature 5: JP 10-221549 A

Patent Literature 6: JP 11-60732 A

Patent Literature 7: JP 2005-163012 A

[Disclosure of the Invention]

Problems to be Solved by the Invention

5 [0013]

The present invention provides a resin composition for forming a retarder with which orientation and stretching steps are not necessary, and a retarder having a high transparency and high birefringence can be formed by coating the composition on a substrate; a color filter substrate having the retarder; and an LCD having a wide
10 viewing-angle and a high contrast; as well as a process for producing a color filter for LCDs with a retarder.

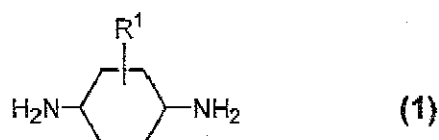
Means for Solving the Problems

[0014]

To solve the above-described problems, the present invention has the
15 following constitution:

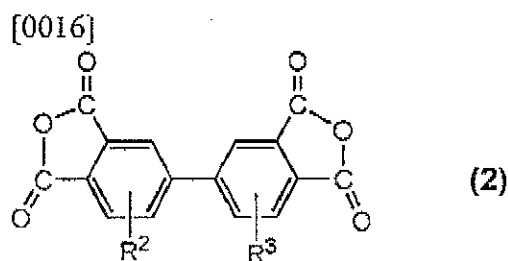
1. A resin composition for forming a retarder which is used for liquid crystal displays, which has an optically negative uniaxial anisotropy, whose optical axis is substantially vertical to the film plane of the retarder, and which has a birefringence Δn in the direction of thickness of 0.01 to 0.3, the composition comprising a
20 polyimide precursor and an organic solvent, the polyimide precursor being obtained by reacting at least one tetracarboxylic dianhydride and at least one diamine; at least one of the at least one tetracarboxylic dianhydride and the at least one diamine being an alicyclic compound.
2. The composition according to item 1, wherein the diamine which is the
25 alicyclic compound is a *trans*-1,4-diaminocyclohexane compound represented by the following General Formula (1):

[0015]



(wherein R^1 represents a monovalent organic group or a hydrogen atom).

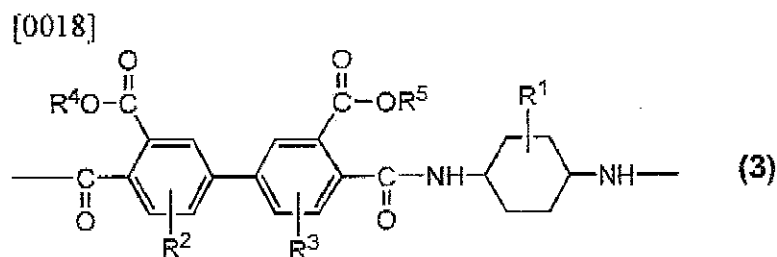
3. The composition according to item 2, wherein the tetracarboxylic dianhydride is a 3,3',4,4'-biphenyltetracarboxylic dianhydride compound represented by the following General Formula (2):



(wherein R^2 and R^3 independently represent a monovalent organic group or a hydrogen atom).

[0017]

4. The composition according to item 3, wherein the polyimide precursor has at least the structural unit represented by the following General Formula (3):



(wherein R^1 , R^2 , R^3 , R^4 and R^5 independently represent a monovalent organic group or a hydrogen atom).

[0019]

5. The composition according to item 1, wherein the tetracarboxylic dianhydride which is the alicyclic compound is 1,2,3,4-cyclobutanetetracarboxylic dianhydride.

6. The composition according to item 5, wherein the diamine is an aromatic diamine having a rigid molecular structure.

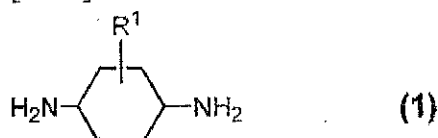
[0020]

7. The composition according to item 6, wherein the aromatic diamine having a rigid molecular structure is at least one selected from the group consisting of *p*-phenylenediamine and 4,4'-diaminobenzanilide.

- 5 8. A resin composition for forming a retarder which is used for liquid crystal displays, which has an optically negative uniaxial anisotropy, and whose optical axis is substantially vertical to the film plane of the retarder, the composition comprising a polyamic acid compound obtained by reacting a diamine component(s) containing a *trans*-1,4-diaminocyclohexane compound represented by the following General

10 Formula (1):

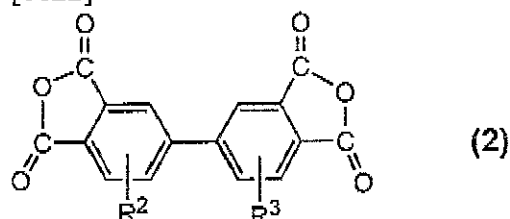
[0021]



(wherein R^1 represents a monovalent organic group or a hydrogen atom)

and a tetracarboxylic dianhydride(s) containing a 3,3',4,4'-biphenyltetracarboxylic dianhydride compound represented by the following General Formula (2):

15 [0022]

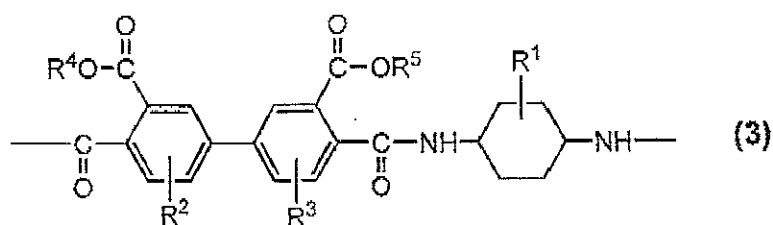


(wherein R^2 and R^3 independently represent a monovalent organic group or a hydrogen atom).

[0023]

9. The composition according to item 8, wherein the polyamic acid compound
20 has at least the structural unit represented by the following General Formula (3):

[0024]



(wherein R^1 , R^2 , R^3 , R^4 and R^5 independently represent a monovalent organic group or a hydrogen atom).

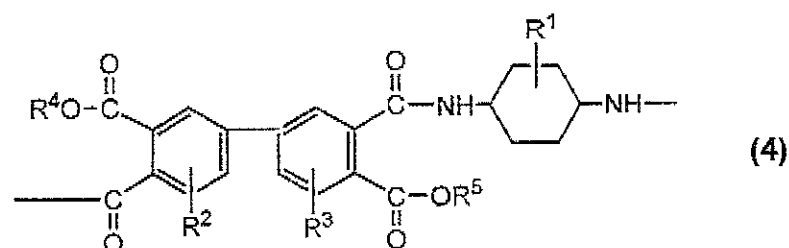
[0025]

10. The composition according to item 9, wherein the R^1 is a hydrogen atom or a C_1 - C_4 linear or branched alkyl group; and R^2 , R^3 , R^4 and R^5 are each a hydrogen atom.

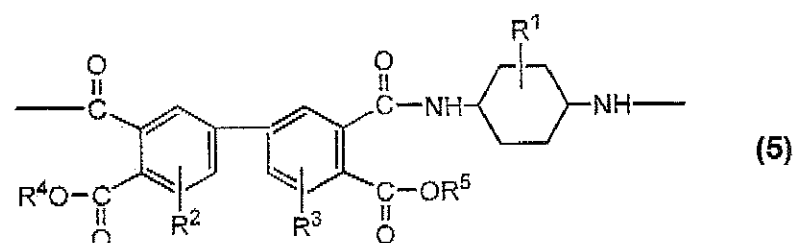
[0026]

11. The composition according to item 9 or 10, wherein the polyamic acid compound further comprises at least one structural unit represented by the following General Formulae (4) to (8):

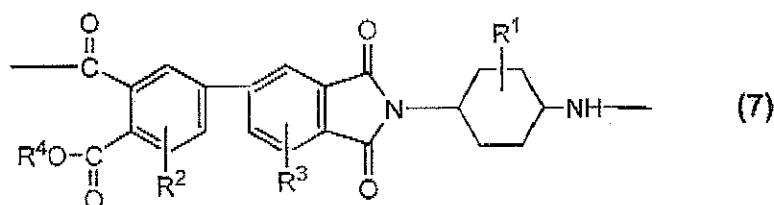
[0027]



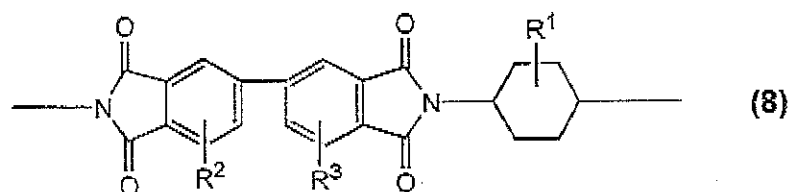
[0028]



15 [0029]



[0030]



(wherein in Formulae (4), (5), (6), (7) and (8), R¹, R², R³, R⁴ and R⁵ independently represent a monovalent organic group or a hydrogen atom).

[0031]

12. The composition according to item 10, wherein in the Formulae (4), (5), (6), (7) and (8), the R¹ is a hydrogen atom or a C₁-C₄ linear or branched alkyl group; and R², R³, R⁴ and R⁵ are each a hydrogen atom.

[0032]

13. The composition according to item 11 or 12, wherein the total amount of the structural units represented by the Formula (3), (4), (5), (6), (7) or (8) is not less than 50 mol% of the total structural units constituting the polyamic acid.

[0033]

14. The resin composition according to any one of items 1 to 13, wherein a part or all of the amine terminal groups of the polyimide precursor or the polyamic acid are capped by an amic acid-forming reaction with a dicarboxylic anhydride.

[0034]

15. The composition according to item 14, wherein the dicarboxylic anhydride is at least one selected from the group consisting of maleic anhydride, phthalic anhydride, succinic anhydride and nadic anhydride.

[0035]

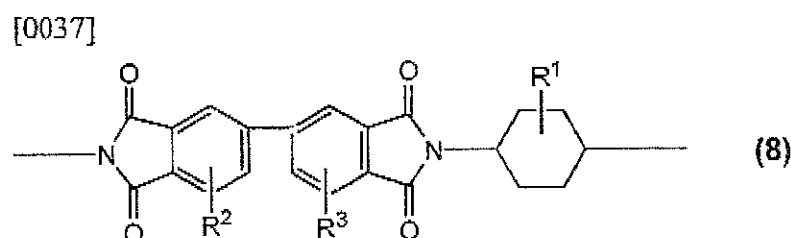
16. Use of the composition according to any one of items 1 to 15 for the

production of a resin composition for forming a retarder.

[0036]

17. A color filter substrate for liquid crystal displays, comprising a transparent substrate on which red, blue and green picture elements are two-dimensionally arrayed; and a retarder formed from the resin composition for forming a retarder according to any one of items 1 to 15.

18. The color filter substrate according to item 17, wherein the retarder comprises a polyimide containing structural units represented by the following General Formula (8):



(wherein R^1 , R^2 and R^3 independently represents a monovalent organic group or a hydrogen atom).

in an amount of not less than 50 mol%.

[0038]

19. The color filter substrate according to item 18, wherein the R^1 is a hydrogen atom or a C_1 - C_4 linear or branched alkyl group; and R^2 and R^3 are each a hydrogen atom.

[0039]

20. The color filter substrate according to any one of items 17 to 19, wherein the retarder formed from the resin composition for forming the retarder is so formed as to cover the picture elements.

[0040]

21. A liquid crystal display comprising the color filter substrate for liquid crystal displays according to any one of items 17 to 20, the liquid color display employing a

display mode wherein the liquid crystal molecules are aligned in the direction substantially vertical to the liquid crystal cell surface when no electric voltage is applied, and the liquid crystal molecules are aligned in the direction substantially parallel to the liquid crystal cell surface when an electric voltage is applied.

5 [0041]

22. A process for producing a color filter substrate for liquid crystal displays, the process comprising coating the resin composition for forming a retarder according to any one of items 1 to 15 on the surface of the color filter substrate, on which surface pixels each having red, blue and green picture elements are two-dimensionally
10 arrayed; and heating the composition.

[Effects of the Invention]

[0042]

With the resin composition for a retarder, having the above-described constitution, a retarder for LCDs can be easily formed, and improvements in the
15 viewing-angle characteristics and contrast may be attained by the retarder.

Best Mode for Carrying out the Invention

[0043]

The present invention will now be described in more detail.

The resin composition for a retarder according to the present invention is
20 characterized in that it has an optically negative uniaxial anisotropy, has an optical axis substantially vertical to the surface of the retarder, has a birefringence Δn in the direction of thickness of 0.01 to 0.3, and that it comprises a polyimide precursor and an organic solvent, wherein the polyimide precursor is obtained by reacting at least one tetracarboxylic dianhydride and at least one diamine; and at least one of the
25 tetracarboxylic dianhydride and the diamine is an alicyclic compound.

[0044]

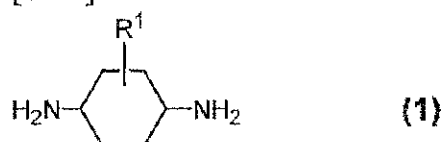
The polyimide precursor used in the present invention may have any structure

which can be converted to a polyimide by heating or chemically, such as a polyamic acid, polyamic acid ester, polyamic acid partial ester, polyamic acid silyl ester, polyamic acid salt, polyisoimide or the like.

[0045]

As the diamine which is an alicyclic compound used for obtaining the polyimide precursor, the *trans*-1,4-diaminocyclohexane compounds represented by the following General Formula (I) are preferred:

[0046]



(wherein R^1 represents a monovalent organic group or a hydrogen atom).

[0047]

Here, R^1 preferably is a C_1 - C_{30} organic group or a hydrogen atom, and more preferably C_1 - C_4 linear or branched chain alkyl group such as methyl group, ethyl group, n-propyl group, isopropyl group, n-butyl group, isobutyl, sec-butyl group or the like, or a hydrogen atom. Among these, *trans*-1,4-diaminocyclohexane, *trans*-1,4-diamino-2-methylcyclohexane and *trans*-1,4-diamino-2,5-dimethylcyclohexane are preferred, and especially, *trans*-1,4-diaminocyclohexane is preferred.

[0048]

In the 1,4-diaminocyclohexane compounds, there exist *trans* isomers wherein the configuration of the amino groups at 1- and 4-positions is in the *trans* configuration and *cis* isomers wherein the configuration thereof is in the *cis* configuration. Although *trans*-1,4-diaminocyclohexane compounds are usually obtained by hydrogenating *p*-phenylenediamine compounds which are precursors thereof, the products of the reaction are mixtures of *trans* and *cis* isomers (e.g., JP 51-48198 B). The *trans*-1,4-diaminocyclohexane compounds which may preferably be used in the present invention are those obtained by purification of the hydrogenated

compound by a known method such as distillation or recrystallization. The content of the *cis* isomer(s) is not restricted as long as the effects of the present invention are not adversely affected. Usually, the content of the *cis* isomer(s) is not more than 50% by weight, preferably not more than 30% by weight, still more preferably not more than 10% by weight. By making the content of the *cis* isomer(s) within the range mentioned above, decrease in the orientation of the polyimide molecular chains due to the bent structure of the *cis* isomer(s) is suppressed and a practically sufficient birefringence can be obtained.

[0049]

To repeat purification of the *trans*-1,4-diaminocyclohexane compound(s) by recrystallization using a solvent such as *n*-hexane so as to decrease coloring components is an effective method for promoting the transparency.

[0050]

The *trans*-1,4-diaminocyclohexane compound(s) may be used in combination with other diamine compound(s) within the range not adversely affecting the effects of the present invention. In these cases, the ratio of the *trans*-1,4-diaminocyclohexane compound(s) used among the total diamines is preferably not less than 50 mol%, more preferably not less than 70 mol%, still more preferably not less than 90 mol%. If the ratio of the *trans*-1,4-diaminocyclohexane compound(s) used is less than 50 mol%, the characteristics aimed at by the present invention may not be obtained.

[0051]

Examples of the diamine which may be used in combination with *trans*-1,4-diaminocyclohexane compound(s) include aromatic diamines such as 2,2'-bis(trifluoromethyl)benzidine, *p*-phenylenediamine, *m*-phenylenediamine, 2,4-diaminotoluene, 2,5-diaminotoluene, 2,4-diaminoxylenc, 2,4-diaminodurenc, 4,4'-diaminodiphenylmethane, 4,4'-diaminodiphenylethane, 4,4'-diaminodiphenylether,

3,4'-diaminodiphenylether, 4,4'-diaminodiphenyl sulfone, 3,3'-diaminodiphenyl sulfone, 4,4'-diaminobenzophenone, 3,3'-diaminobenzophenone, 4,4'-diaminobenzanilide, benzidine, 3,3'-dihydroxybenzidine, 3,3'-dimethoxybenzidine, *o*-tolidine, *m*-tolidine, 1,4-bis(4-aminophenoxy)benzene, 1,3-bis(4-aminophenoxy)benzene, 1,3-bis(3-aminophenoxy)benzene, 4,4'-bis(4-aminophenoxy)biphenyl, bis(4-(3-aminophenoxy)phenyl)sulfone, bis(4-(4-aminophenoxy)phenyl)sulfone, 2,2-bis(4-(4-aminophenoxy)phenyl)propane, 2,2-bis(4-(4-aminophenoxy)phenyl)hexafluoropropane, 2,2-bis(4-aminophenyl)hexafluoropropane and the like; aliphatic and alicyclic diamine such as 1,3-propanediamine, tetramethylenediamine, pentamethylenediamine, hexamethylenediamine, isophoronediamine, tetrahydrodicyclopentadienylenediamine, hexahydro-4,7-methanoindanilenedimethylene diamine, tricyclo[6.2.1.0^{2,7}]-undecylene dimethyldiamine, 4,4'-methylenebis(cyclohexylamine), 2,5-norbornanebis(methylamine), 2,6-norbornanebis(methylamine), 2,7-norbornanebis(methylamine) and the like.

[0052]

By using a siloxane diamine(s) as a part of the diamine, adhesive property with inorganic substrates and the like may be improved. Siloxane diamine(s) are usually preferably used in an amount of 1 to 20 mol% of the total amines. Specific examples of siloxane diamine include bis(3-aminopropyl)tetramethyl disiloxane.

[0053]

Examples of the tetracarboxylic dianhydride which is reacted with alicyclic diamine include aromatic tetracarboxylic dianhydrides such as 3,3',4,4'-biphenyltetracarboxylic dianhydride, 2,3,3',4'-biphenyltetracarboxylic dianhydride, 3,3',4,4'-benzophenonetetracarboxylic dianhydride, pyromellitic dianhydride, 3,4,9,10-perylenetetracarboxylic dianhydride, 3,3',4,4'-diphenylsulfonetetracarboxylic dianhydride, 4,4'-oxydiphthalic acid dianhydride,

1,2,5,6-naphthalenetetracarboxylic dianhydride, 3,3',4,4'-

paraterphenyltetracarboxylic dianhydride, 3,3',4,4'-methaterphenyltetracarboxylic dianhydride, 4,4'-(2,2-hexafluoroisopropylidene)diphthalic acid dianhydride and the like; aliphatic alicyclic tetracarboxylic dianhydride such as 1,2,3,4-

butanetetracarboxylic dianhydride, 1,3-dimethyl-1,2,3,4-cyclobutanetetracarboxylic dianhydride, 1,2,3,4-cyclopentanetetracarboxylic dianhydride, 2,3,5-

tricarboxycyclopentyl acetic acid dianhydride, 1,2,4,5-cyclohexanetetracarboxylic dianhydride, 3,3',4,4'-dicyclohexanetetracarboxylic dianhydride, 1,2,4,5-

norbornenetetracarboxylic dianhydride, 5-(2,5-dioxotetrahydrofural)-3-methyl-3-cyclohexene-1,2-dicarboxylic acid dianhydride, 1,3,3a,4,5,9b-hexahydro-5-

(tetrahydro-2,5-dioxo-3-furanyl)-naphtho[1,2-c]furan-1,3-dione, 1,3,3a,4,5,9b-hexahydro-5-methyl-5-(tetrahydro-2,5-dioxo-3-furanyl)-naphtho[1,2-c]furan-1,3-

dione, 1,3,3a,4,5,9b-hexahydro-5,8-dimethyl-5-(tetrahydro-2,5-dioxo-3-furanyl)-naphtho[1,2-c]furan-1,3-dione, bicyclo[2.2.2]-oct-7-ene-2,3:5,6-tetracarboxylic

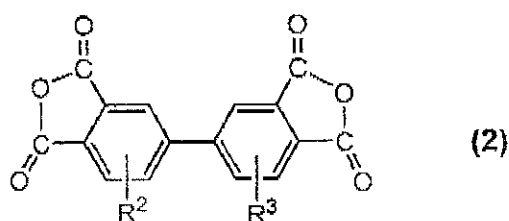
dianhydride, bicyclo[2.2.2]octane-2,3:5,6-tetracarboxylic dianhydride and the like.

[0054]

Among these tetracarboxylic dianhydrides, to use a substituted or unsubstituted 3,3',4,4'-biphenyltetracarboxylic dianhydride(s) represented by the following General Formula (2) is preferred, and to use 3,3',4,4'-

biphenyltetracarboxylic dianhydride is more preferred.

[0055]



(wherein R^2 and R^3 represent a monovalent organic group or a hydrogen atom, and may be the same or different).

Preferred examples of R^2 and R^3 in Formula (2) include hydrogen, C_1 - C_3 alkyl groups, C_1 - C_3 halogenated alkyl groups, phenyl group, substituted phenyl groups and the like, and hydrogen is especially preferred.

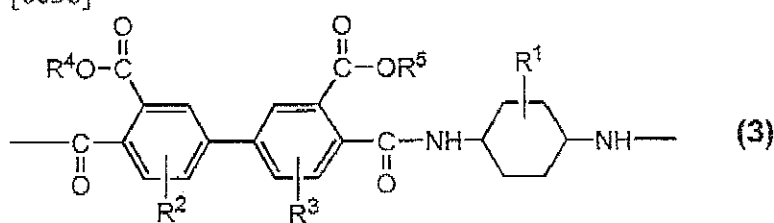
[0056]

Other tetracarboxylic dianhydride(s) may be used in combination with the 3,3',4,4'-biphenyltetracarboxylic dianhydride(s). In this case, the ratio of the 3,3',4,4'-biphenyltetracarboxylic dianhydride(s) used is preferably not less than 50 mol%, more preferably not less than 70 mol%, still more preferably not less than 90 mol%. This is because that if the ratio of the tetracarboxylic dianhydride(s) used is less than 50 mol%, the characteristics aimed at by the present invention may not be obtained in some cases.

[0057]

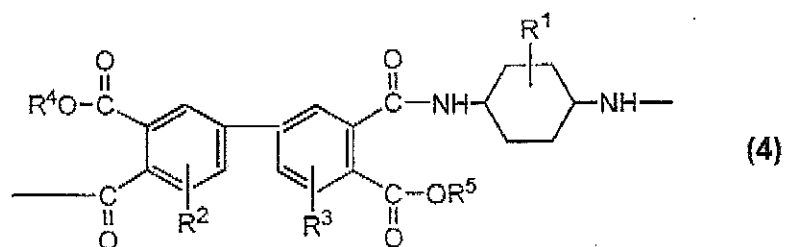
The polyimide precursor obtained by reacting the *trans*-1,4-diaminocyclohexane compound(s) represented by the General Formula (1) below and the substituted or unsubstituted 3,3',4,4'-biphenyltetracarboxylic dianhydride(s) represented by the General Formula (2) below preferably has the structural unit represented by the General Formula (3) below. Further, other than the structural unit represented by the General Formula (3), it may contain the structural unit represented by the General Formula (4) to (8). Any of these structural units are converted to the same structural unit represented by the General Formula (8) by heat or chemical imidization reaction.

[0058]

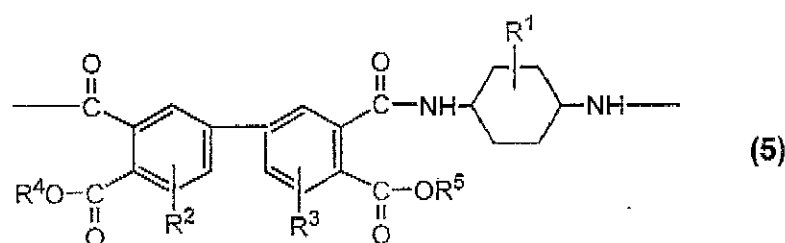


(wherein R^1 , R^2 , R^3 , R^4 and R^5 represent a monovalent organic group or a hydrogen atom, and may be the same or different).

[0059]

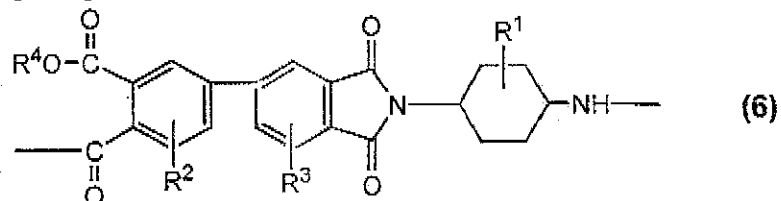


[0060]

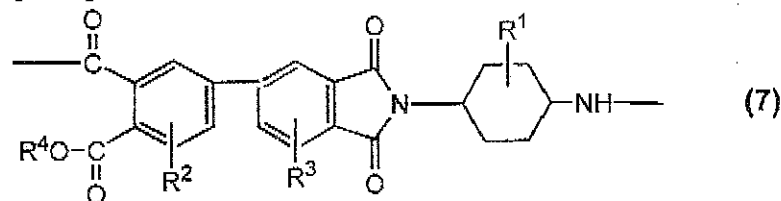


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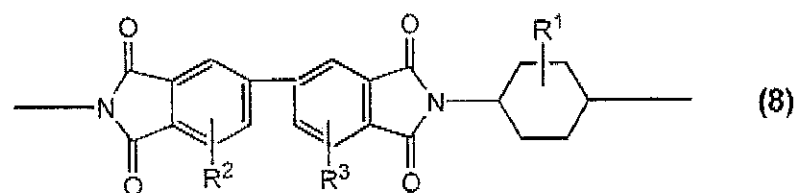
[0061]



[0062]



[0063]



10

(wherein in Formulae (3) to (8), R^1 , R^2 , R^3 , R^4 and R^5 represent a monovalent organic group or a hydrogen atom, and may be the same or different).

Preferred examples of R^1 , R^2 and R^3 in Formulae (3) to (8) are as described above in the explanation of the above-described Formulae (1) and (2); and preferred examples

15

of R^4 and R^5 include hydrogen atom, C_1 - C_{20} alkyl groups, C_1 - C_{20} halogenated alkyl

groups, phenyl group, substituted phenyl groups and the like.

[0064]

The ratio of the structural units represented by the General Formulae (3) to (8) in the polyimide precursor is preferably not less than 50 mol%, more preferably not less than 70 mol%. If the ratio of the structural units represented by the General Formulae (3) to (8) is less than 50 mol%, the characteristics aimed at by the present invention may not be obtained. As described above, since these structural units become the structural unit represented by the General Formula (8) after imidization by heat or the like, in the polyimide after imidization, the ratio of the structural units represented by the General Formula (8) in the total structural units is preferably not less than 50 mol%, more preferably not less than 70 mol%.

[0065]

The present invention also provides a resin composition for forming a retarder which is used for LCDs, which has an optically negative uniaxial anisotropy, and whose optical axis is substantially vertical to the surface of the retarder, the composition comprising a polyamic acid compound obtained by reacting a diamine component(s) containing a *trans*-1,4-diaminocyclohexane compound represented by the above-described General Formula (1) and a tetracarboxylic dianhydride(s) containing a 3,3',4,4'-biphenyltetracarboxylic dianhydride compound represented by the above-described General Formula (2). In this case, the polyamic acid compound preferably has at least the structural unit represented by the above-described General Formula (3). The formed retarder preferably has a birefringence Δn in the direction of thickness of 0.01 to 0.3. The above explanations about the *trans*-1,4-diaminocyclohexane compounds represented by the General Formula (1) and the 3,3',4,4'-biphenyltetracarboxylic dianhydrides represented by the above-described General Formula (2) as well as about the preferred compounds included therein can be equally applied here as they are.

[0066]

The polyimide precursor can be obtained by a known method by the reaction between a tetracarboxylic dianhydride(s) and a diamine(s). The polyamic acid ester can be obtained, as described in, for example, JP 8-92496 A by a method wherein a
5 tetracarboxylic dianhydride is esterified to a tetracarboxylic acid diester with an organic compound having an alcoholic hydroxyl group, converting the obtained tetracarboxylic diester to an acid chloride, and then the acid chloride is reacted with a diamine; or by a method wherein a tetracarboxylic dianhydride is esterified with an organic compound having an alcoholic hydroxyl group to a tetracarboxylic acid
10 diester which is then reacted with a carbodiimide, and then the resulting product is reacted with a diamine. The polyamic acid partial ester can be obtained, for example, by a method wherein an organic compound having a glycidyl group or isocyanate group is attached to a carboxylic group of the polyamic acid by addition reaction, which polyamic acid is obtained by reacting a tetracarboxylic dianhydride and a diamine; or by a method, as described in JP 2000-212216 A, wherein an acetal
15 compound is reacted with the carboxyl group of the polyamic acid obtained by reacting a tetracarboxylic dianhydride with a diamine. The polyamic acid silyl ester can be obtained, for example, as described in JP 64-63070 A, JP 2001-72768 and JP 2005-146073, by a method wherein a diamine is converted to a bis-silyl diamine with
20 a silylating agent, and the bis-silyl diamine is then reacted with a tetracarboxylic dianhydride.

[0067]

As the tetracarboxylic dianhydride which is an alicyclic compound used in order to obtain the polyimide precursor, 1,2,3,4-cyclobutanetetracarboxylic
25 dianhydride is preferably used. 1,2,3,4-cyclobutanetetracarboxylic dianhydride can be synthesized by a known method (for example, by the method described in JP 2-61956 B, JP 3-137125, or J. Polym. Sci.: Part A: Polymer Chemistry, Vol. 38, page

108(2000)).

[0068]

1,2,3,4-cyclobutanetetracarboxylic dianhydride may be used together with other tetracarboxylic dianhydride(s). In this case, the ratio of 1,2,3,4-cyclobutanetetracarboxylic dianhydride used in the total tetracarboxylic dianhydrides is preferably not less than 50 mol%, preferably not less than 70 mol%, still more preferably not less than 90 mol%. If the ratio of the 1,2,3,4-cyclobutanetetracarboxylic dianhydride used is less than 50 mol%, the characteristics aimed at by the present invention may not be obtained in some cases.

[0069]

Examples of tetracarboxylic dianhydride which may be used in combination with 1,2,3,4-cyclobutanetetracarboxylic dianhydride include aromatic tetracarboxylic dianhydrides such as 3,3',4,4'-biphenyltetracarboxylic dianhydride, 2,3,3',4'-biphenyltetracarboxylic dianhydride, 3,3',4,4'-benzophenonetetracarboxylic dianhydride, pyromellitic dianhydride, 3,4,9,10-perylenetetracarboxylic dianhydride, 3,3',4,4'-diphenylsulfonetetracarboxylic dianhydride, 4,4'-oxydiphthalic acid dianhydride, 1,2,5,6-naphthalenetetracarboxylic dianhydride, 3,3',4,4'-paraterphenyltetracarboxylic dianhydride, 3,3',4,4'-metaterphenyltetracarboxylic dianhydride, 4,4'-(2,2-hexafluoroisopropylidene)diphthalic acid dianhydride and the like; aliphatic and alicyclic tetracarboxylic dianhydrides such as 1,2,3,4-butanetetracarboxylic dianhydride, 1,3-dimethyl-1,2,3,4-cyclobutanetetracarboxylic dianhydride, 1,2,3,4-cyclopentanetetracarboxylic dianhydride, 2,3,5-tricarboxycyclopentyl acetic acid dianhydride, 1,2,4,5-cyclohexanetetracarboxylic dianhydride, 3,3',4,4'-dicyclohexanetetracarboxylic dianhydride, 1,2,4,5-norbornenetetracarboxylic dianhydride, 5-(2,5-dioxotetrahydrofural)-3-methyl-3-cyclohexene-1,2-dicarboxylic acid dianhydride, 1,3,3a,4,5,9b-hexahydro-5-(tetrahydro-2,5-dioxo-3-furanyl)-naphtho[1,2-c]furan 1,3-dione, 1,3,3a,4,5,9b-

hexahydro-5-methyl-5-(tetrahydro-2,5-dioxo-3-furanyl)-naphtho[1,2-c]furan 1,3-dione, 1,3,3a,4,5,9b-hexahydro-5,8-dimethyl-5-(tetrahydro-2,5-dioxo 3-furanyl)-naphtho[1,2-c]furan 1,3-dione, bicyclo[2.2.2]-oct-7-ene-2,3:5,6-tetracarboxylic dianhydride, bicyclo[2.2.2]octane-2,3:5,6-tetracarboxylic dianhydride and the like.

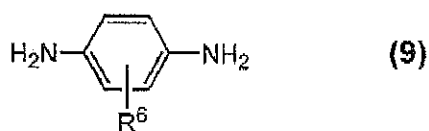
5 [0070]

As the diamine to be reacted with the tetracarboxylic dianhydride which is an alicyclic compound, an aromatic diamine having a rigid molecular structure is preferably employed for the purpose of obtaining a retarder having a high birefringence. The term "aromatic diamine having a rigid molecular structure" herein means an aromatic diamine in which the conformational change due to thermal motion of the molecules is small and the change of the relative positions of the two amino groups constituting the diamine is small. Preferably, it is (1) an aromatic diamine constituted by one selected from the group consisting of benzene ring, aromatic heterocycles and fused rings thereof, in which the two amino groups face each other, and the C-N bond of one of the amino groups and the C-N bond of the other amino group are substantially collinear or substantially parallel to each other; or (2) an aromatic diamine constituted by two or more structural units selected from the group consisting of benzene ring, aromatic heterocycles and fused rings thereof, which structural units are bound directly or through an amide bond, wherein the two amino groups face each other, and the C-N bond of one of the amino groups and the C-N bond of the other amino group are substantially collinear or substantially parallel to each other.

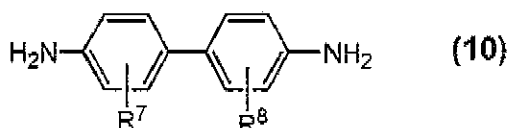
[0071]

Examples of the aromatic diamine having the rigid molecular structure include the compounds represented by the following Formulae (9) to (11).

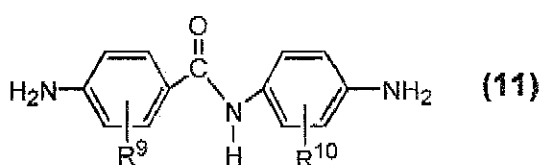
[0072]



[0073]



[0074]



(wherein R^6 , R^7 , R^8 , R^9 , and R^{10} each represents a group selected from the group consisting of -H, -CH₃, -OH, -CF₃, -SO₃H, -COOH, -CONH₂, -F, -Cl, -Br, -CF₃ and -OCF₃, which may be the same or different).

[0075]

Specific examples of aromatic diamine having rigid molecular structure include 4,4'-diaminobenzanilide, benzidine, 3,3'-dimethylbenzidine, 3,3'-dihydroxybenzidine, 3,3'-dimethoxybenzidine, 2,2'-dimethyl benzidine, 2,2'-bis(trifluoromethyl)benzidine, *p*-phenylenediamine, 2,5-diaminotoluene, 3,6-diaminodurene, *m*-phenylenediamine, 2,4-diaminotoluene, 2,4-diaminoxylene.

Among these, 4,4'-diaminobenzanilide, *p*-phenylenediamine, 2,2'-dimethyl benzidine and 2,2'-bis(trifluoromethyl)benzidine are preferably used, and especially, 4,4'-diaminobenzanilide and *p*-phenylenediamine are more preferably used. Since the retarders comprising the polyimide obtained by reacting such a diamine and 1,2,3,4-cyclobutanetetracarboxylic dianhydride has an especially high transparency and high birefringence, they are preferably used. The above-described diamine may be used individually or two or more of them may be used in combination.

[0076]

Other diamine(s) may be co-used together with the aromatic diamine having

the rigid molecular structure. In this case, the ratio of the aromatic diamine having the rigid molecular structure used in the total diamines is preferably not less than 50 mol%, more preferably not less than 70 mol%, still more preferably not less than 90 mol%. If the ratio of the aromatic diamine having the rigid molecular structure used is less than 50 mol%, the characteristics aimed at by the present invention may not be obtained in some cases.

[0077]

Examples of other diamine which may be used in combination with the aromatic diamine having the rigid molecular structure include aromatic diamines such as 4,4'-diaminodiphenylmethane, 4,4'-diaminodiphenylethane, 4,4'-diaminodiphenylether, 3,4'-diaminodiphenylether, 4,4'-diaminodiphenyl sulfone, 3,3'-diaminodiphenyl sulfone, 4,4'-diaminobenzophenone, 3,3'-diaminobenzophenone, 1,4-bis(4-aminophenoxy)benzene, 1,3-bis(4-aminophenoxy)benzene, 1,3-bis(3-aminophenoxy)benzene, 4,4'-bis(4-aminophenoxy)biphenyl, bis(4-(3-aminophenoxy)phenyl)sulfone, bis(4-(4-aminophenoxy)phenyl)sulfone, 2,2-bis(4-(4-aminophenoxy)phenyl)propane, 2,2-bis(4-(4-aminophenoxy)phenyl)hexafluoropropane, 2,2-bis(4-aminophenyl)hexafluoropropane and the like.

[0078]

Further, aliphatic and alicyclic diamines such as 1,3-propanediamine, tetramethylenediamine, pentamethylenediamine, hexamethylenediamine, trans-1,4-diaminocyclohexane, cis-1,4-diaminocyclohexane, isophoronediamine, tetrahydrodicyclopentadienylenediamine, hexahydro-4,7-methanoindanilenedimethylene diamine, tricyclo[6.2.1.0^{2,7}]-undecylene dimethyldiamine, 4,4'-methylenebis(cyclohexylamine), 2,5-norbornane bis(methylamine), 2,6-norbornane bis(methylamine), 2,7-norbornane bis(methylamine) and the like may also be used.

[0079]

By using a siloxane diamine as a part of the diamines, the adhesion with inorganic substrate and the like can be improved. It is preferred usually to use the siloxane diamine in an amount of 1 to 20 mol% in total diamines. A specific
5 example of the siloxane diamine is bis(3-aminopropyl)tetramethyldisiloxane.

[0080]

The reaction between the tetracarboxylic dianhydride and the diamine can be carried out by mixing them in a polar organic solvent. By adjusting the mixing ratio between the tetracarboxylic dianhydride and the diamine, the polymerization degree
10 of the polyamic acid can be adjusted. The ratio between the tetracarboxylic dianhydride and the diamine subjected to the synthesis reaction is preferably such that the amount of the acid anhydride groups in the tetracarboxylic dianhydride is 0.2 to 2 equivalents, more preferably 0.8 to 1.2 equivalents per 1 equivalent of the amino groups contained in the diamine. Similar to usual polycondensation reactions, the
15 closer the molar ratio of the tetracarboxylic dianhydride to the diamine to 1, the larger the polymerization degree of the polymer. If the polymerization degree is too small, the strength of the polyimide coating film is insufficient, and if the polymerization degree is too large, the ease of handling in forming the polyimide coating film is degraded. Therefore, the polymerization degree of the polyamic acid,
20 in terms of reduced viscosity (also indicated as η_{sp}/C), is preferably 0.05 to 5.0 dl/g (measured at a concentration of 0.5 g/dl in N-methylpyrrolidone at 30°C), more preferably 0.1 to 2.0 dl/g.

[0081]

For the purpose of promoting the heat resistance and processability, a
25 dicarboxylic anhydride, monoamine compound, monoisocyanate compound or the like may be added to the reaction system in order to cap all or part of the amino groups or carboxyl groups at the ends of the polyamic acid molecules. Examples of

the dicarboxylic anhydride include maleic anhydride, phthalic anhydride, 4-methyl phthalic anhydride, 4-tert-butyl phthalic anhydride, itaconic acid anhydride, anhydrous nadic acid and the like. Examples of monoamine compounds include

5 Examples of monoisocyanate compounds include phenyl isocyanate, naphthyl isocyanate and the like. It is especially preferred that a part or all of the terminal amino groups be capped by the amic acid-forming reaction with the dicarboxylic anhydride, and the dicarboxylic anhydride is preferably at least one selected from the group consisting of maleic anhydride, phthalic anhydride, succinic anhydride and
10 nadic anhydride.

[0082]

The synthesis reaction of the polyamic acid is carried out in an organic solvent at a temperature preferably from -20°C to 200°C, more preferably 0°C to 150°C. As the organic solvent, any organic solvent may be used as long as it can
15 dissolve the polyamic acid synthesized. Examples thereof include aprotic polar solvents such as N-methyl-2-pyrrolidone, N,N-dimethylacetamide, N, N-dimethylformamide, dimethyl sulfoxide, γ -butyrolactone, tetramethylurea, 1,3-dimethyl-2-imidazolidinone, hexamethylphosphoramide and the like. The amount of the organic solvent used is preferably such that concentration of the solids
20 including the tetracarboxylic dianhydride and the diamine is 0.1 to 30% by weight based on the total weight of the reaction solution.

[0083]

As the above-described organic solvent, a poor solvent of the polyamic acid, such as an alcohol, ketone, ester, halogenated hydrocarbon or hydrocarbon may be
25 used in an amount such that the polyamic acid generated does not precipitate. Specific examples of the poor solvent include methyl alcohol, ethyl alcohol, isopropyl alcohol, cyclohexanol, ethylene glycol, propylene glycol, 1,4-butanediol,

diethylene glycol, triethylene glycol, 3-methyl-3-methoxybutanol, acetone, methyl ethyl ketone, methyl isobutyl ketone, cyclohexanone, ethyl lactate, butyl lactate, methyl acetate, ethyl acetate, butyl acetate, methyl-3-methoxypropionate, 3-methyl-3-methoxybutyl acetate, ethyl ethoxypropionate, diethyl oxalic acid, diethyl malonate, diethyl ether, tetrahydrofuran, ethylene glycol methyl ether, ethylene glycol ethyl ether, ethylene glycol n-propyl ether, ethylene glycol isopropyl ether, ethylene glycol n-butyl ether, ethylene glycol dimethyl ether, ethylene glycol ethyl ether acetate, diethylene glycol monomethyl ether, diethylene glycol monoethyl ether, diethylene glycol dimethyl ether, diethylene glycol diethyl ether, diethylene glycol monomethyl ether acetate, diethylene glycol monoethyl ether acetate, propylene glycol monoethyl ether acetate, dichloromethane, 1,2-dichloroethane, 1,4-dichlorobutane, trichloroethane, chlorobenzene, *o*-dichlorobenzene, hexane, heptane, octane, benzene, toluene and xylene.

[0084]

The resin composition for the retarder can be produced by adding an organic solvent to the polyimide precursor or a solution thereof, and uniformly mixing the resulting mixture by a conventional method. The temperature during the preparation of the resin composition is preferably 0°C to 200°C, more preferably 20°C to 60°C. Examples of the organic solvent are the organic solvents exemplified as the solvent which may be used in the synthesis reaction of the polyamic acid. The poor solvents exemplified as the solvent which may be co-used in the synthesis reaction of the polyamic acid may be appropriately selected and may also be co-used.

[0085]

The solid concentration of the resin composition is selected taking the viscosity, volatility and the like into consideration, and is preferably within the range from 1 to 10% by weight. That is, the resin composition is coated on the surface of a substrate and a coating film which serves as the retarder is formed. If the solid

concentration is less than 1% by weight, the thickness of the coating film is too small so that a good retarder cannot be obtained, and if the solid concentration is more than 10% by weight, the thickness of the coating film is too large so that a good retarder cannot be obtained, and the viscosity of the resin composition is too high so that the coating characteristics are poor.

[0086]

In view of improving the adhesion with the substrate surface, the resin composition may contain a functional silane-containing compound such as 3-aminopropyl trimethoxysilane, 3-(2-aminoethyl)aminopropyl trimethoxysilane, N-phenyl-3-aminopropyl trimethoxysilane, 3-mercaptopropyltrimethoxysilane, 3-glycidoxypropyltrimethoxysilane, 2-(3,4-epoxycyclohexyl)ethyltrimethoxysilane, 3-isocyanate propyltrimethoxysilane, 3-aminopropyl triethoxysilane or the like and/or an epoxy group-containing compound such as epoxy resin class or the like; bisphenol type epoxy resin, bisphenol A novolac-type epoxy resin, biphenyl type epoxy resin, phenol novolac-type epoxy resin, alkylphenol novolac-type epoxy resin, polyglycol type epoxy resin, cyclic aliphatic epoxy resin, cresol novolac-type epoxy resin, glycidyl amine type epoxy resin, naphthalene type epoxy resin, urethane modified epoxy resin, rubber modified epoxy resin, epoxy modified polysiloxane or the like.

In view of improving the uniformity in the thickness of the coating film and surface smoothness, a nonionic surfactant such as polyoxyethylene lauryl ether, polyoxyethylene dilaurate or the like, or a surfactant such as fluorine surfactant, silane surfactant, acrylate copolymer surfactant or the like, may be contained. The contents of these additives are those which do not adversely affect the effects of the present invention, and usually not more than 20% by weight, preferably not more than 10% by weight in the whole composition.

[0087]

The resin composition is coated on a substrate by dipping method, roll coater

method, spinner method, die coating method, a method using a wire bar, or the like, and then dried by air drying, vacuum drying, drying by heating using an oven or hot plate, or the like, to form a coating film. The heating conditions vary depending on the resin and solvent used, and coating amount, and usually, heating at 50°C to 400°C for 1 to 300 minutes is preferred.

[0088]

The substrate to be coated may be the LCD substrate, that is, a color filter substrate or TFT substrate itself. Alternatively, the resin composition is once coated on a base film, and then may be attached to the LCD substrate through an adhesive layer. The coating film is formed on the surface opposite to the liquid crystal layer. Alternatively, it may be formed on the surface of the LCD substrate contacting the liquid crystal layer. For example, the resin composition may be coated on the surface of the color filter substrate for LCDs, which substrate is a transparent substrate on which red, blue and green picture elements are two-dimensionally arrayed, on which surface the picture elements are formed, such that the film covers the picture elements. The term "covers the picture elements" herein means that the film is formed closer to the liquid crystal layer than the picture elements. The picture elements and the retarder composed of the above-described composition may directly contact or may not contact each other. For example, in cases where there is an over coat layer for flattening on the color filter substrate, the retarder composed of the above-described resin composition may be formed on the side closer to the liquid crystal layer than the over coat layer, or on the side closer to the substrate than the over coat layer. Alternatively, it is possible to form the retarder composed of the above-described resin composition on the substrate, and to form a color filter for LCDs thereon in which the red, blue and green picture elements are two-dimensionally arrayed.

[0089]

Further, the above-described resin composition may contain a color component such as a pigment or dye, and the composition may be used as a varnish for forming the picture elements of the color filter, thereby giving retarder function to each picture element itself. In cases where the resin composition is used for forming the picture elements, it is preferred to adjust the retardation R with respect to the main wavelength λ in each of the red, green and blue picture elements such that the phase retardance R/λ is substantially identical, for unifying the phase retardance compensation in each color.

[0090]

The retarder for LCDs according to the present invention is formed by coating the resin composition on the substrate and heating the coated composition. The retarder has a phase retardance and has a function to compensate the birefringence generated during the passage of the light through the liquid crystal layer in LCDs. Since the molecular chains of polyimide resins are likely to be oriented in the direction parallel to the surface of the substrate, a difference in refractive indices in the direction of thickness of the film and in the direction parallel to the film plane (birefringence as a film) is generated. Further, since the orientation of the molecules in the film plane is random, there is no anisotropy of refractive index in the direction parallel to the film plane. That is, in the retarder of the present invention, taking the x-axis and y-axis in the film plane and taking the z-axis in the direction vertical to the film plane, the refractive indices are $n_x \geq n_y > n_z$, and the retarder is a retarder (negative C plate) having an optically negative uniaxial anisotropy, whose optical axis is substantially vertical to the film plane. The term "has an optically negative uniaxial anisotropy" herein means that the refractive indices along two axes are identical and the refractive index along the remaining one axis is small, and the term "optical axis is substantially vertical to the film plane" herein means that $n_x = n_y > n_z$. More specifically, this is satisfied if $0 \leq n_x - n_y \leq$

0.005, and $n_y > n_z$ are satisfied.

[0091]

The birefringence $\Delta n (= n_x - n_z)$ in the direction of thickness of the retarder is preferably 0.01 to 0.3, more preferably not less than 0.03, still more preferably not less than 0.05. If the birefringence is smaller than 0.01, the film thickness of the retarder necessary for compensating the phase retardance of the liquid crystal is so large that film-formation is difficult.

[0092]

The film thickness of the retarder is preferably 0.5 to 20 μm .

Although the retarder of the present invention is effective for general LCDs, since the optical axis is substantially vertical to the plane of the retarder, the retarder is especially preferably used for the LCDs employing a display mode wherein the liquid crystal molecules are aligned in the direction substantially vertical to the liquid crystal cell surface when no electric voltage is applied (homeotropic alignment), and the liquid crystal molecules are aligned in the direction substantially parallel to the liquid crystal cell surface when an electric voltage is applied, more specifically, MVA (Multi-domain Vertical Alignment) mode, PVA (Patterned Vertical Alignment) mode, CPA (Continuous Pinwheel Alignment) mode or the like.

[0093]

Since the optical axis of the retarder of the present invention is substantially vertical to the surfaces of the two substrates sandwiching the liquid crystal, no phase retardance-compensating effect is obtained when the display is watched in the direction vertical thereto. However, in the vertical alignment modes, since the phase retardance of the liquid crystal layer is almost zero in the vertical direction when no electric voltage is applied, there is no need to compensate the phase retardance. That is, when no electric voltage is applied, good black display is obtained even without a compensation. However, in an oblique direction, since

there is a phase retardance in the liquid crystal layer even when no electric voltage is applied, light leakage occurs if the phase retardation is not compensated and good black display is not attained, which causes decrease in the contrast. Therefore, the retarder of the present invention exhibits a prominent effect to increase the contrast in oblique directions in the vertical alignment modes, and in turn, to increase the viewing-angle.

Examples

[0094]

<Measurement of Reduced Viscosity (η_{sp}/C) of Polyimide Precursor>

The polyimide precursor was dissolved in N-methylpyrrolidone to a concentration of 0.5 g/dl, and the diluted solution was measured at 30°C using an Ubbelohde viscometer.

[0095]

<Measurement of Optical Axis>

The optical axis was measured using "OPTIPRO" produced by Shintech, Inc.

A polyimide precursor solution was coated on a glass substrate with a spinner to a final thickness of 2.0 μm , and dried at 120°C for 20 minutes. The coated solution was then heated at 240°C for 30 minutes or at 270°C for 40 minutes to obtain a polyimide resin thin film. The refractive index anisotropy in the direction parallel to the film plane of the polyimide resin thin film, and the refractive index anisotropy in the direction vertical to the film plane were measured.

<Measurement of Birefringence>

The birefringence was measured using "PRISM COUPLER 2010" produced by Metricon Company.

A polyimide precursor solution was coated on a glass substrate with a spinner to a final thickness of 2.0 μm , and dried at 120°C for 20 minutes. The coated solution was then heated at 240°C for 30 minutes or at 270°C for 40 minutes to

obtain a polyimide resin thin film. The refractive index n_1 ($= n_x$) in the direction vertical to the film plane of the polyimide resin thin film and the refractive index n_2 ($= n_z$) in the direction of thickness were measured, and the birefringence was calculated from the difference between the refractive indices according to the equation below. As the light source, HeNe laser light of 632.8 nm was used.

$$\Delta n = n_1 - n_2$$

[0096]

<Measurement Method of Coloring of Film>

The coloring of the film was measured using OTSUKA ELECTRONICS Co. Ltd. Multi Channel Photo Detector "MCPD2000". A polyimide precursor solution was coated on a glass substrate with a spinner to a final thickness of 2.0 μm , and dried at 120°C for 20 minutes. The resultant was then heated at 240°C for 30 minutes or at 270°C for 40 minutes to obtain polyimide resin thin film. In the XYZ chromaticity coordinate (CIE 1931 standard chromaticity coordinate), the difference (Δx , Δy) between the chromaticity coordinate of the standard light source C (x , y) = (0.3100, 0.3162) and the chromatic coordinate (x_1 , y_1) after the light passed through the polyimide resin thin film was determined. Here, $\Delta x = x_1 - x$, and $\Delta y = y_1 - y$. When both Δx and Δy are large, white display is yellowish and the display quality is degraded. Preferably, both Δx and Δy are not more than 0.005, more preferably, not more than 0.003.

[0097]

Synthesis Example (Synthesis of 1,2,3,4-cyclobutanetetracarboxylic Dianhydride)

To an internal radiation glass reaction flask having an inner volume of 2 liters made of Pyrex (registered trademark) equipped with a water-cooling lamp jacket made of glass, 255g (2.60 moles) of maleic anhydride and 1445 g of ethyl acetate were fed, and the atmosphere in the flask was replaced with nitrogen, followed by stirring the mixture at room temperature. While continuing the stirring, the reaction

solution was cooled to 5°C, irradiation with a 400W high pressure mercury lamp was started, and the irradiation was continued for 96 hours. During the irradiation, the temperature of the reaction solution was kept at 3°C to 5°C. After completion of the reaction, the crystals and mother liquor were separated by filtration. After washing
5 the crude crystals with ethyl acetate, the crude crystals were dried in a vacuum drier at 40°C for 10 hours to obtain 194 g of crystals of 1,2,3,4-cyclobutanetetracarboxylic dianhydride.

[0098]

Example 1

10 Under dry nitrogen gas flow, 10.96 g (0.096 moles) of *trans*-1,4-diaminocyclohexane and 0.99 g (0.004 moles) of bis(3-aminopropyl)tetramethyldisiloxane were dissolved in 177.28 g of N-methyl-2-pyrrolidone. Thereafter, 28.25 g (0.096 moles) of 3,3',4,4'-biphenyltetracarboxylic dianhydride and 40.00 g of N-methyl-2-pyrrolidone were added, and the mixture was
15 stirred at 60°C for 3 hours. Then 1.18 g (0.008 moles) of phthalic anhydride was added, and the resulting mixture was stirred at 60°C for 3 hours to obtain a transparent and viscose polyamic acid solution A (polymer concentration: 16% by weight). The viscosity of the solution A measured at 25°C was 830 mPa·s. The reduced viscosity was 0.70 dl/g.

20 [0099]

The polyamic acid solution A was coated on a glass substrate with a spinner to a final thickness of 2.0 μm, and dried at 120°C for 20 minutes, followed by further heat treatment to obtain a polyimide resin thin film. In the polyimide resin thin film heat-treated at 240°C for 30 minutes, there was no refractive index anisotropy in the
25 directions parallel to the film plane of the polyimide resin thin film, so that n_x was identical to n_y . The refractive index anisotropy in the direction vertical to the film plane was measured, and n_x was larger than n_z , so that it exhibited negative uniaxial

anisotropy. That is, the obtained polyimide resin thin film had an optically negative uniaxial anisotropy, and the optical axis thereof was substantially vertical to the film plane. The refractive index in the direction parallel to the film plane of the polyimide resin thin film n_1 was 1.683, the refractive index in the direction of the film plane n_2 was 1.592, and the birefringence Δn was 0.091. The chromaticity coordinate of the transmitted light was (0.3108, 0.3172), Δx was 0.0008 and Δy was 0.0010. Thus, both Δx and Δy were not more than 0.003, so that a retarder without coloring was obtained. In the polyimide resin thin film heat-treated at 270°C for 40 minutes, there was no refractive index anisotropy in the directions parallel to the film plane of the polyimide resin thin film, so that n_x was identical to n_y . The refractive index anisotropy in the direction vertical to the film plane was measured, and n_x was larger than n_z , so that it exhibited negative uniaxial anisotropy. That is, the obtained polyimide resin thin film had an optically negative uniaxial anisotropy, and the optical axis thereof was substantially vertical to the film plane. The refractive index in the direction parallel to the film plane of the polyimide resin thin film n_1 was 1.745, n_2 was 1.571, and the birefringence Δn was 0.174. The chromaticity coordinate of the transmitted light was (0.3116, 0.3179), Δx was 0.0016 and Δy was 0.0017. Thus, both Δx and Δy were not more than 0.003, so that a retarder without coloring was obtained.

[0100]

Example 2

Under dry nitrogen gas flow, 10.96 g (0.096 moles) of *trans*-1,4-diaminocyclohexane and 0.99 g (0.004 moles) of bis(3-aminopropyl)tetramethyldisiloxane were dissolved in 173.28 g of N-methyl-2-pyrrolidone. Thereafter, 25.30 g (0.086 moles) of 3,3',4,4'-biphenyltetracarboxylic dianhydride, 2.18 g (0.010 mole) of pyromellitic dianhydride and 40.00 g of N-methyl-2-pyrrolidone were added, and the mixture was stirred at 60°C for 3 hours.

Then 1.18 g (0.008 moles) of phthalic anhydride was added, and the resulting mixture was stirred at 60°C for 3 hours to obtain a transparent and viscose polyamic acid solution B (polymer concentration: 16% by weight). The viscosity of the solution B measured at 25°C was 690 mPa·s. The reduced viscosity was 0.67 dL/g.

[0101]

There was no refractive index anisotropy in the directions parallel to the film plane of the polyimide resin thin film prepared in the same manner as in Example 1 and heat-treated at 270°C for 40 minutes, so that n_x was identical to n_y . The refractive index anisotropy in the direction vertical to the film plane was measured, and n_x was larger than n_z , so that it exhibited negative uniaxial anisotropy. That is, the obtained polyimide resin thin film had an optically negative uniaxial anisotropy, and the optical axis thereof was substantially vertical to the film plane. The refractive index in the direction parallel to the film plane of the polyimide resin thin film n_1 was 1.698, the refractive index in the direction of thickness of the film n_2 was 1.586, and the birefringence Δn was 0.112. The chromaticity coordinate of the transmitted light was (0.3111, 0.3174), Δx was 0.0011 and Δy was 0.0012. Thus, both Δx and Δy were not more than 0.003, so that a retarder without coloring was obtained.

[0102]

Example 3

Under dry nitrogen gas flow, 10.96 g (0.096 moles) of *trans*-1,4-diaminocyclohexane and 0.99 g (0.004 moles) of bis(3-aminopropyl)tetramethyldisiloxane were dissolved in 171.06 g of N-methyl-2-pyrrolidone. Thereafter, 28.25 g (0.096 moles) of 3,3',4,4'-biphenyltetracarboxylic dianhydride and 40.00 g of N-methyl-2-pyrrolidone were added, and the mixture was stirred at 60°C for 5 hours to obtain a transparent and viscose polyamic acid solution C (polymer concentration: 16% by weight). The viscosity of the solution C

measured at 25°C was 5222 mPa·s. The reduced viscosity was 0.81 dl/g.

[0103]

The refractive index n_1 in the directions parallel to the film plane of the polyimide resin thin film prepared in the same manner as in Example 1 and heat-treated at 240°C for 30 minutes was 1.706, the refractive index n_2 in the direction of the thickness of the film was 1.592, and the birefringence Δn was 0.114. The chromaticity coordinate of the transmitted light was (0.3109, 0.3171), Δx was 0.0009 and Δy was 0.0009. Thus, both Δx and Δy were not more than 0.003, so that a retarder without coloring was obtained. In the polyimide resin thin film heat-treated at 270°C for 40 minutes, there was no refractive index anisotropy in the directions parallel to the film plane of the polyimide resin thin film, so that n_x was identical to n_y . The refractive index anisotropy in the direction vertical to the film plane was measured, and n_x was larger than n_z , so that it exhibited negative uniaxial anisotropy. That is, the obtained polyimide resin thin film had an optically negative uniaxial anisotropy, and the optical axis thereof was substantially vertical to the film plane. The refractive index in the direction parallel to the film plane of the polyimide resin thin film n_1 was 1.735, n_2 was 1.572, and the birefringence Δn was 0.163. The chromaticity coordinate of the transmitted light was (0.3116, 0.3180), Δx was 0.0016 and Δy was 0.0018. Thus, both Δx and Δy were not more than 0.003, so that a retarder without coloring was obtained.

[0104]

Example 4

Under dry nitrogen gas flow, 11.42 g (0.100 mole) of *trans*-1,4-diaminocyclohexane was dissolved in 174.42 g of N-methyl-2-pyrrolidone. Thereafter, 29.42 g (0.100 mole) of 3,3',4,4'-biphenyltetracarboxylic dianhydride and 40.00 g of N-methyl-2-pyrrolidone were added, and the mixture was stirred at 60°C for 5 hours. After cooling the mixture to room temperature, 85.07 g of N-methyl-2-

pyrrolidone was added to obtain a transparent and viscose polyamic acid solution D (polymer concentration: 12% by weight). The viscosity of the solution D measured at 25°C was 5878 mPa·s. The reduced viscosity was 1.68 dl/g.

[0105]

5 There was no refractive index anisotropy in the directions parallel to the film plane of the polyimide resin thin film prepared in the same manner as in Example 1 and heat-treated at 270°C for 40 minutes, so that n_x was identical to n_y . The refractive index anisotropy in the direction vertical to the film plane was measured, and n_x was larger than n_z , so that it exhibited negative uniaxial anisotropy. That is,
10 the obtained polyimide resin thin film had an optically negative uniaxial anisotropy, and the optical axis thereof was substantially vertical to the film plane. The refractive index in the direction parallel to the film plane of the polyimide resin thin film n_1 was 1.745, the refractive index in the direction of thickness of the film n_2 was 1.571, and the birefringence Δn was 0.174. The chromaticity coordinate of the
15 transmitted light was (0.3120, 0.3187), Δx was 0.0020 and Δy was 0.0025. Thus, both Δx and Δy were not more than 0.003, so that a retarder without coloring was obtained.

[0106]

Example 5

20 Under dry nitrogen gas flow, 11.42 g (0.100 mol) of *trans*-1,4-diaminocyclohexane was dissolved in 176.30 g of N-methyl-2-pyrrolidone. Thereafter, 29.78 g (0.096 moles) of 4,4'-oxydipthalic dianhydride and 40.00 g of N-methyl-2-pyrrolidone were added, and the mixture was stirred at 60°C for 5 hours. After cooling the mixture to room temperature, 85.83 g of N-methyl-2-pyrrolidone
25 was added to obtain a transparent and viscose polyamic acid solution E (polymer concentration: 12% by weight). The viscosity of the solution E measured at 25°C was 1139 mPa·s. The reduced viscosity was 0.96 dl/g.

[0107]

There was no refractive index anisotropy in the directions parallel to the film plane of the polyimide resin thin film prepared in the same manner as in Example 1 and heat-treated at 270°C for 40 minutes, so that n_x was identical to n_y . The refractive index anisotropy in the direction vertical to the film plane was measured, and n_x was larger than n_z , so that it exhibited negative uniaxial anisotropy. That is, the obtained polyimide resin thin film had an optically negative uniaxial anisotropy, and the optical axis thereof was substantially vertical to the film plane. The refractive index in the direction parallel to the film plane of the polyimide resin thin film n_1 was 1.631, the refractive index in the direction of thickness of the film n_2 was 1.616, and the birefringence Δn was 0.015. The chromaticity coordinate of the transmitted light was (0.3122, 0.3190), Δx was 0.0022 and Δy was 0.0028. Thus, both Δx and Δy were not more than 0.003, so that a retarder without coloring was obtained.

[0108]

Example 6

Under dry nitrogen gas flow, 10.38 g (0.096 moles) of *p*-phenylenediamine and 0.99 g (0.004 moles) of bis(3-aminopropyl)tetramethyldisiloxane were dissolved in 124.79 g of N-methyl-2-pyrrolidone. Thereafter, 18.83 g (0.096 moles) of 1,2,3,4-cyclobutanetetracarboxylic dianhydride and 40.00 g of N-methyl-2-pyrrolidone were added, and the mixture was stirred at 60°C for 3 hours. Then 1.18 g (0.008 moles) of phthalic anhydride was added, and the resulting mixture was stirred at 60°C for 3 hours to obtain a transparent and viscose polyamic acid solution F (polymer concentration: 16% by weight). The viscosity of the solution F measured at 25°C was 384 mPa·s. The reduced viscosity was 0.56 dl/g.

[0109]

The refractive index n_1 in the directions parallel to the film plane of the

polyimide resin thin film prepared in the same manner as in Example 1 and heat-treated at 240°C for 30 minutes was 1.621, the refractive index n_2 in the direction of the film plane was 1.586, and the birefringence Δn was 0.035. The chromaticity coordinate of the transmitted light was (0.3109, 0.3173), Δx was 0.0009 and Δy was 0.0011. Thus, both Δx and Δy were not more than 0.005, so that a retarder with slight coloring was obtained. In the polyimide resin thin film heat-treated at 270°C for 40 minutes, there was no refractive index anisotropy in the directions parallel to the film plane of the polyimide resin thin film, so that n_x was identical to n_y . The refractive index anisotropy in the direction vertical to the film plane was measured, and n_x was larger than n_z , so that it exhibited negative uniaxial anisotropy. That is, the obtained polyimide resin thin film had an optically negative uniaxial anisotropy, and the optical axis thereof was substantially vertical to the film plane. The refractive index in the direction parallel to the film plane of the polyimide resin thin film n_1 was 1.631, n_2 was 1.586, and the birefringence Δn was 0.045. The chromaticity coordinate of the transmitted light was (0.3127, 0.3197), Δx was 0.0027 and Δy was 0.0035. Thus, both Δx and Δy were not more than 0.005, so that a retarder with slight coloring was obtained.

[0110]

Example 7

Under dry nitrogen gas flow, 21.82 g (0.096 moles) of 4,4'-diaminobenzanilide and 0.99 g (0.004 moles) of bis(3-aminopropyl)tetramethyldisiloxane were dissolved in 112.41 g of γ -butyrolactone. Then, 72.41 g of N-methyl-2-pyrrolidone was added. Thereafter, 18.83 g (0.096 moles) of 1,2,3,4-cyclobutanetetracarboxylic dianhydride and 40.00 g of N-methyl-2-pyrrolidone were added, and the mixture was stirred at 60°C for 3 hours. Then 1.18 g (0.008 moles) of phthalic anhydride was added, and the resulting mixture was stirred at 60°C for 3 hours to obtain a transparent and viscose polyamic acid solution

G (polymer concentration: 16% by weight). The viscosity of the solution G measured at 25°C was 1080 mPa·s. The reduced viscosity was 0.71 dl/g.

[0111]

There was no refractive index anisotropy in the directions parallel to the film plane of the polyimide resin thin film prepared in the same manner as in Example 1 and heat-treated at 240°C for 30 minutes, so that n_x was identical to n_y . The refractive index anisotropy in the direction vertical to the film plane was measured, and n_x was larger than n_z , so that it exhibited negative uniaxial anisotropy. That is, the obtained polyimide resin thin film had an optically negative uniaxial anisotropy, and the optical axis thereof was substantially vertical to the film plane. The refractive index in the direction parallel to the film plane of the polyimide resin thin film n_1 was 1.669, the refractive index in the direction of thickness of the film n_2 was 1.604, and the birefringence Δn was 0.065. The chromaticity coordinate of the transmitted light was (0.3115, 0.3180), Δx was 0.0015 and Δy was 0.0018. Thus, both Δx and Δy were not more than 0.003, so that a retarder without coloring was obtained. There was no refractive index anisotropy in the directions parallel to the film plane of the polyimide resin thin film heat-treated at 270°C for 40 minutes, so that n_x was identical to n_y . The refractive index anisotropy in the direction vertical to the film plane was measured, and n_x was larger than n_z , so that it exhibited negative uniaxial anisotropy. That is, the obtained polyimide resin thin film had an optically negative uniaxial anisotropy, and the optical axis thereof was substantially vertical to the film plane. The refractive index in the direction parallel to the film plane of the polyimide resin thin film n_1 was 1.688, n_2 was 1.601, and the birefringence Δn was 0.087. The chromaticity coordinate of the transmitted light was (0.3142, 0.3212), Δx was 0.0042 and Δy was 0.0050. Thus, both Δx and Δy were not more than 0.005, so that a retarder with slight coloring was obtained.

[0112]

Example 8

Under dry nitrogen gas flow, 20.38 g (0.096 moles) of 2,2'-dimethylbenzidine and 0.99 g (0.004 moles) of bis(3-aminopropyl)tetramethyldisiloxane were dissolved in 177.28 g of N-methyl-2-pyrrolidone. Thereafter, 18.83 g (0.096 moles) of

5 1,2,3,4-cyclobutanetetracarboxylic dianhydride and 40.00 g of N-methyl-2-pyrrolidone were added, and the mixture was stirred at 60°C for 3 hours. Then 1.18 g (0.008 moles) of phthalic anhydride was added, and the resulting mixture was stirred at 60°C for 3 hours to obtain a transparent and viscose polyamic acid solution H (polymer concentration: 16% by weight). The viscosity of the solution H

10 measured at 25°C was 1055 mPa·s. The reduced viscosity was 0.78 dl/g.

[0113]

The refractive index n_1 in the directions parallel to the film plane of the polyimide resin thin film prepared in the same manner as in Example 1 and heat-treated at 240°C for 30 minutes was 1.625, the refractive index n_2 in the direction of

15 the thickness of the film was 1.587, and the birefringence Δn was 0.038. The chromaticity coordinate of the transmitted light was (0.3114, 0.3180), Δx was 0.0014 and Δy was 0.0018. Thus, both Δx and Δy were not more than 0.003, so that a retarder without coloring was obtained. In the polyimide resin thin film heat-treated at 270°C for 40 minutes, there was no refractive index anisotropy in the directions

20 parallel to the film plane of the polyimide resin thin film, so that n_x was identical to n_y . The refractive index anisotropy in the direction vertical to the film plane was measured, and n_x was larger than n_z , so that it exhibited negative uniaxial anisotropy. That is, the obtained polyimide resin thin film had an optically negative uniaxial anisotropy, and the optical axis thereof was substantially vertical to the film plane.

25 The refractive index in the direction parallel to the film plane of the polyimide resin thin film n_1 was 1.625, n_2 was 1.586, and the birefringence Δn was 0.039. The chromaticity coordinate of the transmitted light was (0.3134, 0.3210), Δx was 0.0034

and Δy was 0.0048. Thus, both Δx and Δy were not more than 0.005, so that a retarder with slight coloring was obtained.

[0114]

Example 9

Under dry nitrogen gas flow, 30.74 g (0.096 moles) of 2,2'-bis(trifluoromethyl)benzidine and 0.99 g (0.004 moles) of bis(3-aminopropyl)tetramethyldisiloxane were dissolved in 231.69 g of N-methyl-2-pyrrolidone. Thereafter, 18.83 g (0.096 moles) of 1,2,3,4-cyclobutanetetracarboxylic dianhydride and 40.00 g of N-methyl-2-pyrrolidone were added, and the mixture was stirred at 60°C for 3 hours. Then 1.18 g (0.008 moles) of phthalic anhydride was added, and the resulting mixture was stirred at 60°C for 3 hours to obtain a transparent and viscose polyamic acid solution I (polymer concentration: 16% by weight). The viscosity of the solution I measured at 25°C was 275 mPa·s. The reduced viscosity was 0.58 dl/g.

[0115]

The refractive index n_1 in the directions parallel to the film plane of the polyimide resin thin film prepared in the same manner as in Example 1 and heat-treated at 240°C for 30 minutes was 1.560, the refractive index n_2 in the direction of the thickness of the film was 1.540, and the birefringence Δn was 0.020. The chromaticity coordinate of the transmitted light was (0.3103, 0.3164), Δx was 0.0003 and Δy was 0.0002. Thus, both Δx and Δy were not more than 0.003, so that a retarder without coloring was obtained. In the polyimide resin thin film heat-treated at 270°C for 40 minutes, there was no refractive index anisotropy in the directions parallel to the film plane of the polyimide resin thin film, so that n_x was identical to n_y . The refractive index anisotropy in the direction vertical to the film plane was measured, and n_x was larger than n_z , so that it exhibited negative uniaxial anisotropy. That is, the obtained polyimide resin thin film had an optically negative uniaxial

anisotropy, and the optical axis thereof was substantially vertical to the film plane. The refractive index in the direction parallel to the film plane of the polyimide resin thin film n_1 was 1.559, n_2 was 1.541, and the birefringence Δn was 0.018. The chromaticity coordinate of the transmitted light was (0.3108, 0.3171), Δx was 0.0008 and Δy was 0.0009. Thus, both Δx and Δy were not more than 0.003, so that a retarder without coloring was obtained.

[0116]

Example 10

Under dry nitrogen gas flow, 19.22 g (0.096 moles) of 4,4'-diaminodiphenylether and 0.99 g (0.004 moles) of bis(3-aminopropyl)tetramethyldisiloxane were dissolved in 171.21 g of N-methyl-2-pyrrolidone. Thereafter, 18.83 g (0.096 moles) of 1,2,3,4-cyclobutanetetracarboxylic dianhydride and 40.00 g of N-methyl-2-pyrrolidone were added, and the mixture was stirred at 60°C for 3 hours. Then 1.18 g (0.008 moles) of phthalic anhydride was added, and the resulting mixture was stirred at 60°C for 3 hours to obtain a transparent and viscous polyamic acid solution J (polymer concentration: 16% by weight). The viscosity of the solution J measured at 25°C was 258 mPa·s. The reduced viscosity was 0.56 dl/g.

[0117]

There was no refractive index anisotropy in the directions parallel to the film plane of the polyimide resin thin film prepared in the same manner as in Example 1 and heat-treated at 240°C for 30 minutes, so that n_x was identical to n_y . The refractive index anisotropy in the direction vertical to the film plane was measured, and n_x was larger than n_z , so that it exhibited negative uniaxial anisotropy. That is, the obtained polyimide resin thin film had an optically negative uniaxial anisotropy, and the optical axis thereof was substantially vertical to the film plane. The refractive index in the direction parallel to the film plane of the polyimide resin thin

film n_1 was 1.620, the refractive index in the direction of thickness of the film n_2 was 1.608, and the birefringence Δn was 0.013. The chromaticity coordinate of the transmitted light was (0.3111, 0.3178), Δx was 0.0011 and Δy was 0.0016. Thus, both Δx and Δy were not more than 0.003, so that a retarder without coloring was obtained. There was no refractive index anisotropy in the directions parallel to the film plane of the polyimide resin thin film heat-treated at 270°C for 40 minutes, so that n_x was identical to n_y . The refractive index anisotropy in the direction vertical to the film plane was measured, and n_x was larger than n_z , so that it exhibited negative uniaxial anisotropy. That is, the obtained polyimide resin thin film had an optically negative uniaxial anisotropy, and the optical axis thereof was substantially vertical to the film plane. The refractive index in the direction parallel to the film plane of the polyimide resin thin film n_1 was 1.622, n_2 was 1.607, and the birefringence Δn was 0.015. The chromaticity coordinate of the transmitted light was (0.3127, 0.3197), Δx was 0.0027 and Δy was 0.0035. Thus, both Δx and Δy were not more than 0.005, so that a retarder with slight coloring was obtained.

[0118]

Example 11

Under dry nitrogen gas flow, 10.81 g (0.100 mole) of *p*-phenylenediamine was dissolved in 132.41 g of N-methyl-2-pyrrolidone. Thereafter, 19.61 g (0.100 mole) of 1,2,3,4-cyclobutanetetracarboxylic dianhydride and 40.00 g of N-methyl-2-pyrrolidone were added, and the mixture was stirred at 60°C for 4 hours to obtain a transparent and viscose polyamic acid solution K (polymer concentration: 15% by weight). The viscosity of the solution K measured at 25°C was 9257 mPa·s. The reduced viscosity was 1.36 dl/g.

[0119]

There was no refractive index anisotropy in the directions parallel to the film plane of the polyimide resin thin film prepared in the same manner as in Example 1

and heat-treated at 240°C for 30 minutes, so that n_x was identical to n_y . The refractive index anisotropy in the direction vertical to the film plane was measured, and n_x was larger than n_z , so that it exhibited negative uniaxial anisotropy. That is, the obtained polyimide resin thin film had an optically negative uniaxial anisotropy, and the optical axis thereof was substantially vertical to the film plane. The refractive index in the direction parallel to the film plane of the polyimide resin thin film n_1 was 1.635, the refractive index in the direction of thickness of the film n_2 was 1.584, and the birefringence Δn was 0.051. The chromaticity coordinate of the transmitted light was (0.3116, 0.3182), Δx was 0.0016 and Δy was 0.0020. Thus, both Δx and Δy were not more than 0.003, so that a retarder without coloring was obtained.

[0120]

Example 12

Under dry nitrogen gas flow, 22.73 g (0.100 mole) of 4,4'-diaminobenzanilide was dissolved in 111.14 g of γ -butyrolactone. Then, 71.14 g of N-methyl-2-pyrrolidone was added. Thereafter, 19.61 g (0.100 mole) of 1,2,3,4-cyclobutanetetracarboxylic dianhydride and 40.00 g of N-methyl-2-pyrrolidone were added, and the mixture was stirred at 60°C for 4 hours to obtain a transparent and viscose polyamic acid solution L (polymer concentration: 16% by weight). The viscosity of the solution L measured at 25°C was 31,400 mPa·s. The reduced viscosity was 1.81 dl/g.

[0121]

There was no refractive index anisotropy in the directions parallel to the film plane of the polyimide resin thin film prepared in the same manner as in Example 1 and heat-treated at 240°C for 30 minutes, so that n_x was identical to n_y . The refractive index anisotropy in the direction vertical to the film plane was measured, and n_x was larger than n_z , so that it exhibited negative uniaxial anisotropy. That is,

the obtained polyimide resin thin film had an optically negative uniaxial anisotropy, and the optical axis thereof was substantially vertical to the film plane. The refractive index in the direction parallel to the film plane of the polyimide resin thin film n_1 was 1.685, the refractive index in the direction of thickness of the film n_2 was 1.598, and the birefringence Δn was 0.087. The chromaticity coordinate of the transmitted light was (0.3131, 0.3204), Δx was 0.0031 and Δy was 0.0042. Thus, both Δx and Δy were not more than 0.005, so that a retarder with slight coloring was obtained.

[0122]

10 Comparative Example 1

Under dry nitrogen gas flow, 11.92 g (0.048 moles) of 3,3'-diaminodiphenylsulfone, 5.19 g (0.048 moles) of *p*-phenylenediamine and 0.99 g (0.004 moles) of bis(3-aminopropyl)tetramethyldisiloxane were dissolved in 209.55 g of N-methyl-2-pyrrolidone. Thereafter, 28.25 g (0.096 moles) of 3,3',4,4'-biphenyltetracarboxylic dianhydride and 40.00 g of N-methyl-2-pyrrolidone were added, and the mixture was stirred at 60°C for 3 hours. Then 1.18 g (0.008 moles) of phthalic anhydride was added, and the resulting mixture was stirred at 60°C for 3 hours to obtain a transparent and viscose polyamic acid solution M (polymer concentration: 16% by weight). The viscosity of the solution M measured at 25°C was 940 mPa·s. The reduced viscosity was 0.64 dl/g.

[0123]

There was no refractive index anisotropy in the directions parallel to the film plane of the polyimide resin thin film prepared in the same manner as in Example 1 and heat-treated at 270°C for 40 minutes, so that n_x was identical to n_y . The refractive index anisotropy in the direction vertical to the film plane was measured, and n_x was larger than n_z , so that it exhibited negative uniaxial anisotropy. That is, the obtained polyimide resin thin film had an optically negative uniaxial anisotropy,

and the optical axis thereof was substantially vertical to the film plane. The refractive index in the direction parallel to the film plane of the polyimide resin thin film n_1 was 1.754, the refractive index in the direction of thickness of the film n_2 was 1.663, and the birefringence Δn was 0.091. The chromaticity coordinate of the transmitted light was (0.3170, 0.3272), Δx was 0.0070 and Δy was 0.0110. Thus, both Δx and Δy were more than 0.005, and the retarder was colored in yellow.

[0124]

Comparative Example 2

Under dry nitrogen gas flow, 10.38 g (0.096 moles) of *p*-phenylenediamine and 0.99 g (0.004 moles) of bis(3-aminopropyl)tetramethyldisiloxane were dissolved in 174.23 g of N-methyl-2-pyrrolidone. Thereafter, 28.25 g (0.096 moles) of 3,3',4,4'-biphenyltetracarboxylic dianhydride and 40.00 g of N-methyl-2-pyrrolidone were added, and the mixture was stirred at 60°C for 3 hours. Then 1.18 g (0.008 moles) of phthalic anhydride was added, and the resulting mixture was stirred at 60°C for 3 hours to obtain a transparent and viscose polyamic acid solution N (polymer concentration: 16% by weight). The viscosity of the solution N measured at 25°C was 664 mPa·s. The reduced viscosity was 0.66 dl/g.

[0125]

There was no refractive index anisotropy in the directions parallel to the film plane of the polyimide resin thin film prepared in the same manner as in Example 1 and heat-treated at 240°C for 30 minutes, so that n_x was identical to n_y . The refractive index anisotropy in the direction vertical to the film plane was measured, and n_x was larger than n_z , so that it exhibited negative uniaxial anisotropy. That is, the obtained polyimide resin thin film had an optically negative uniaxial anisotropy, and the optical axis thereof was substantially vertical to the film plane. The refractive index in the direction parallel to the film plane of the polyimide resin thin film n_1 was 1.810, the refractive index in the direction of thickness of the film n_2

was 1.621, and the birefringence Δn was 0.189. The chromaticity coordinate of the transmitted light was (0.3195, 0.3327), Δx was 0.0095 and Δy was 0.0165. Thus, both Δx and Δy were more than 0.005, and the retarder was colored in yellow. The refractive index n_1 of the polyimide resin thin film heat-treated at 270°C for 40

5 minutes was 1.831, the refractive index n_2 was 1.616, and the birefringence Δn was 0.215. The chromaticity coordinate of the transmitted light was (0.3206, 0.3339), Δx was 0.0106 and Δy was 0.0177. Thus, both Δx and Δy were more than 0.005, and the retarder was colored in yellow.

[0126]

10 Comparative Example 3

Under dry nitrogen gas flow, 21.82 g (0.096 moles) of 4,4'-diaminobenzanilide and 0.99 g (0.004 moles) of bis(3-aminopropyl)tetramethyldisiloxane were dissolved in 137.14 g of γ -butyrolactone. Then, 97.14 g of N-methyl-2-pyrrolidone was added. Thereafter, 28.25 g (0.096

15 moles) of 3,3',4,4'-biphenyltetracarboxylic dianhydride and 40.00 g of N-methyl-2-pyrrolidone were added, and the mixture was stirred at 60°C for 3 hours. Then 1.18 g (0.008 moles) of phthalic anhydride was added, and the resulting mixture was stirred at 60°C for 3 hours to obtain a transparent and viscose polyamic acid solution O (polymer concentration: 16% by weight). The viscosity of the solution O

20 measured at 25°C was 5878 mPa·s. The reduced viscosity was 1.03 dL/g.

[0127]

There was no refractive index anisotropy in the directions parallel to the film plane of the polyimide resin thin film prepared in the same manner as in Example 1 and heat-treated at 240°C for 30 minutes, so that n_x was identical to n_y . The

25 refractive index anisotropy in the direction vertical to the film plane was measured, and n_x was larger than n_z , so that it exhibited negative uniaxial anisotropy. That is, the obtained polyimide resin thin film had an optically negative uniaxial anisotropy,

and the optical axis thereof was substantially vertical to the film plane. The refractive index in the direction parallel to the film plane of the polyimide resin thin film n_1 was 1.826, the refractive index in the direction of thickness of the film n_2 was 1.610, and the birefringence Δn was 0.216. The chromaticity coordinate of the transmitted light was (0.3260, 0.3450), Δx was 0.0160 and Δy was 0.0288. Thus, both Δx and Δy were more than 0.005, and the retarder was colored in yellow. The refractive index n_1 of the polyimide resin thin film heat-treated at 270°C for 40 minutes was 1.838, the refractive index n_2 was 1.609, and the birefringence Δn was 0.229. The chromaticity coordinate of the transmitted light was (0.3255, 0.3437), Δx was 0.0155 and Δy was 0.0275. Thus, both Δx and Δy were more than 0.005, and the retarder was colored in yellow.

[0128]

Comparative Example 4

Under dry nitrogen gas flow, 10.81 g (0.100 moles) of *p*-phenylenediamine was dissolved in 92.92 g of N-methyl-2-pyrrolidone. Thereafter, 22.42 g (0.100 mole) of 1,2,4,5-cyclohexanetetracarboxylic dianhydride and 40.00 g of N-methyl-2-pyrrolidone were added, and the mixture was stirred at 60°C for 4 hours to obtain a transparent and viscose polyamic acid solution P (polymer concentration: 20% by weight). The viscosity of the solution P measured at 25°C was 350 mPa·s. The reduced viscosity was 0.30 dl/g.

[0129]

There was no refractive index anisotropy in the directions parallel to the film plane of the polyimide resin thin film prepared in the same manner as in Example 1 and heat-treated at 240°C for 30 minutes, so that n_x was identical to n_y . The refractive index anisotropy in the direction vertical to the film plane was measured, and n_x was larger than n_z , so that it exhibited negative uniaxial anisotropy. That is, the obtained polyimide resin thin film had an optically negative uniaxial anisotropy,

and the optical axis thereof was substantially vertical to the film plane. The refractive index in the direction parallel to the film plane of the polyimide resin thin film n_1 was 1.600, the refractive index in the direction of thickness of the film n_2 was 1.598, and the birefringence Δn was 0.002. The chromaticity coordinate of the transmitted light was (0.3111, 0.3173), Δx was 0.0011 and Δy was 0.0011. Thus, both Δx and Δy were not more than 0.003, so that the retarder was not colored. However, the birefringence was not sufficient.

[0130]

Comparative Example 5

Under dry nitrogen gas flow, 22.73 g (0.100 mole) of 4,4'-diaminobenzanilide was dissolved in 90.29 g of γ -butyrolactone. Then, 50.29 g of N-methyl-2-pyrrolidone was added. Thereafter, 22.42 g (0.100 mole) of 1,2,4,5-cyclohexanetetracarboxylic dianhydride and 40.00 g of N-methyl-2-pyrrolidone were added, and the mixture was stirred at 60°C for 4 hours to obtain a transparent and viscose polyamic acid solution Q (polymer concentration: 20% by weight). The viscosity of the solution Q measured at 25°C was 209 mPa·s. The reduced viscosity was 0.24 dl/g.

[0131]

There was no refractive index anisotropy in the directions parallel to the film plane of the polyimide resin thin film prepared in the same manner as in Example 1 and heat-treated at 240°C for 30 minutes, so that n_x was identical to n_y . The refractive index anisotropy in the direction vertical to the film plane was measured, and n_x was larger than n_z , so that it exhibited negative uniaxial anisotropy. That is, the obtained polyimide resin thin film had an optically negative uniaxial anisotropy, and the optical axis thereof was substantially vertical to the film plane. The refractive index in the direction parallel to the film plane of the polyimide resin thin film n_1 was 1.638, the refractive index in the direction of thickness of the film n_2

was 1.632, and the birefringence Δn was 0.006. The chromaticity coordinate of the transmitted light was (0.3114, 0.3178), Δx was 0.0014 and Δy was 0.0016. Thus, both Δx and Δy were not more than 0.003, so that the retarder was not colored. However, the birefringence was not sufficient.

5 [0132]

Example 13

A method for producing a color filter having the retarder will now be described.

<Preparation of Black Matrix>

10 In γ -butyrolactone solvent (3825 g), pyromellitic dianhydride (149.6 g), benzophenonetetracarboxylic dianhydride (225.5 g), 3,3'-diaminodiphenylsulfone (69.5 g), 4,4'-diaminodiphenylether (210.2 g) and bis(3-aminopropyl)tetramethyldisiloxane (17.4 g) were reacted at 60°C for 3 hours. Thereafter, maleic anhydride (2.25 g) was added, and the resulting mixture was
15 allowed to react at 60°C for another 1 hour to obtain a polyamic acid solution (polymer concentration: 15% by weight).

[0133]

Using a homogenizer, 7.3 g of carbon black (MA-77 produced by Mitsubishi Chemical), 44.8g of the above-described polyamic acid solution having a polymer
20 concentration of 15% by weight, 35 g of N-methyl-2-pyrrolidone and 12.9 g of 3-methyl-3-methoxyacetate were subjected to a dispersion treatment together with 100 g of glass beads at 7000 rpm for 30 minutes, and then the glass beads were removed by filtration to obtain a pigment dispersion having a pigment concentration of 14% by weight. The primary particle size of the carbon black used was 23 nm. The
25 weight ratio of carbon black/polyamic acid compound was 52/48.

[0134]

To 57.2 g of the pigment dispersion, 36.4 g of N-methyl-2-pyrrolidone and

6.4 g of 3-methoxy-3-methyl-butyl acetate were added and admixed to prepare a black paste. After coating this paste on an alkali-free glass substrate, the paste was prebaked at 130°C to form a black polyamic acid film. Then a positive-working photoresist was applied and heat-dried at 90°C to form a photoresist coating film.

5 The photoresist coating film was exposed with a UV exposure apparatus through a photomask. Thereafter, the resultant was immersed in an alkaline developer to simultaneously carry out the development of the photoresist and etching of the black polyamic acid film to form apertural areas. After the etching, the photoresist layer no longer necessary was peeled off with ethylene glycol monomethyl acetate. The
10 etched black polyamic acid film was heated at 290°C to carry out heat curing to form a polyimide resin black matrix.

[0135]

<Preparation of Picture Elements>

In γ -butyrolactone, pyromellitic dianhydride (0.49 mol equivalent),
15 benzophenone tetracarboxylic dianhydride (0.50 mol equivalent), 4,4'-diaminodiphenylether (0.75 mol equivalent), 3,3'-diaminodiphenylsulfone (0.20 mol equivalent) and bis-(3-aminopropyl)tetramethyldisiloxane (0.05 mol equivalent) were reacted, and maleic anhydride was further reacted, to obtain a polyamic acid solution (polymer concentration: 20% by weight).

20 [0136]

Two hundred grams of this polyamic acid solution was taken, and 136 g of γ -butyrolactone and 64 g of ethylene glycol butyl ether were added to obtain a polyamic acid solution for picture elements having a polymer concentration of 10% by weight.

25 [0137]

Using a homogenizer, 4 g of Pigment Red 177 (Anthraquinone Red), 40 g of γ -butyrolactone and 6 g of ethylene glycol butyl ether were subjected to dispersion

treatment together with 100 g of glass beads at 7000 rpm for 30 minutes, and then the glass beads were removed by filtration to obtain a pigment dispersion having a pigment concentration of 8% by weight.

[0138]

- 5 To 30 g of the pigment dispersion, 30 g of the above-described polyamic acid solution for picture elements having a polymer concentration of 10% by weight was added and admixed to obtain a red color paste.

[0139]

- 10 On a substrate on which the resin black matrix was formed, the red paste was coated and prebaked to form a polyamic acid red color film. By the similar method described above, using a photoresist, red picture elements were formed and the resultant was heated at 290°C to carry out heat curing.

[0140]

- 15 Using a homogenizer, 3.6 g of Pigment Green 7 (Phtharocyanine Green), 0.4 g of Pigment Yellow 83 (Benzidine Yellow), 32 g of γ -butyrolactone and 4 g of ethylene glycol butyl ether were subjected to dispersion treatment together with 120 g of glass beads at 7000 rpm for 30 minutes, and then the glass beads were removed by filtration to obtain a pigment dispersion having a pigment concentration of 10% by weight.

20 [0141]

To 32 g of the pigment dispersion, 30 g of the above-described polyamic acid solution for picture elements having a polymer concentration of 10% by weight was added and admixed to obtain a green color paste.

[0142]

- 25 In the same manner as in the case of using the red paste, green picture elements were prepared using the green color paste, and the resultant was heated at 290°C to carry out heat curing.

[0143]

Using a homogenizer, 60 g of the above-described polyamic acid solution for picture elements having a polymer concentration of 10% by weight, 2.8 g of Pigment Blue 5 (Phtharocyanine Blue), 30 g of N-methyl-pyrrolidone and 10 g of ethylene glycol butyl ether were subjected to dispersion treatment together with 150 g of glass beads at 7000 rpm for 30 minutes, and then the glass beads were removed by filtration to obtain a blue color paste.

[0144]

In the same manner as described above, blue picture elements were prepared using the blue color paste, and the resultant was heated at 290°C to carry out heat curing.

[0145]

A color filter was prepared as described above.

To 187.5 g of the polyamic acid solution A (polymer concentration: 16% by weight) prepared in Example 1, 0.25 g of a surfactant "DISPARLON" LC951 (produced by Kusumoto Chemicals, Ltd.), 218.3 g of N-methyl-2-pyrrolidone and 94.0 g of 3-methoxy-3-methyl-1-butanol were added to prepare a coating solution having a polymer concentration of 6% by weight. The coating solution was coated on the surface of the above-described color filter on which the picture elements were formed by the slit die coating method, and dried at 120°C for 10 minutes, followed by heat treatment at 270°C for 40 minutes to form a polyimide thin film with a thickness of 1.2 μm . The birefringence Δn of this polyimide thin film was 0.174. Thus, a color filter having a retarder which had a retardation of 209 nm, whose optical axis is vertical to the thin film, and which had an optically negative refractive index anisotropy, was obtained by the method described above.

[0146]

Example 14

A polyimide thin film was formed on the color filter in the same manner as in Example 13 except that the polyamic acid solution F (polymer concentration 16% by weight) prepared in Example 6 was used. The thickness of the polyimide thin film was 4.4 μm , the birefringence Δn was 0.045. Thus, a color filter having a retarder which had a retardation of 198 nm, whose optical axis was vertical to the thin film, and which had an optically negative refractive index anisotropy, was obtained by the method described above.

[0147]

Example 15

To 200.0 g of the polyamic acid solution K (polymer concentration: 15% by weight) prepared in Example 11, 0.25 g of a surfactant "DISPARLON" LC951 (produced by Kusumoto Chemicals, Ltd.), 205.8 g of N-methyl-2-pyrrolidone and 94.0 g of 3-methoxy-3-methyl-1-butanol were added to prepare a coating solution having a polymer concentration of 6% by weight. In the same manner as in Example 13, the coating solution was coated on the surface of the above-described color filter substrate on which the picture elements were formed by the slit die coating method, and dried at 120°C for 10 minutes, followed by heat treatment at 240°C for 30 minutes to form a polyimide thin film with a thickness of 4.0 μm . The birefringence Δn of this polyimide thin film was 0.051. Thus, a color filter having a retarder which had a retardation of 204 nm, whose optical axis was vertical to the thin film, and which had an optically negative refractive index anisotropy, was obtained by the method described above.

[0148]

Example 16

<Preparation of Color LCD Element and Evaluation>

A transparent electrode film made of indium oxide was formed on the color filter with the retarder prepared in Example 13. Separately, a substrate made of an

alkali-free glass on which TFT elements, picture element electrodes, a reflecting plate and so on were formed was provided as an opposing substrate.

[0149]

Thereafter, on the transparent electrode on the respective substrates,
5 projections made of polyimide in the form of a stripe were formed by
photolithography, and then a vertical alignment layer was formed. The cross-
section of the projections was trapezoid and the height thereof was about 1.5 μm .
The stripe projections were positioned such that when the color filter substrate and
the TFT substrate are laminated together, the projections of the substrate are arranged
10 alternatively with the stripe projections on the opposing substrate. After adhering
the end portions of the two substrates with a sealing agent, the cell was filled with an
n-type liquid crystal and sealed. Polarizing plates were arranged in front of and in
the rear of the cell such that crossed Nicol is attained. By the method described
above, a test LCD element (sample A) mimicking MVA (Multi-domain Vertical
15 Alignment) mode was prepared. The interval between the electrodes of the cell was
made to be about 5 μm by using beads spacer. Further, a test LCD element (sample
B) which was the same as the above-described sample A except that the polyimide
retarder was not formed was also prepared for comparison.

[0150]

20 The ratio of intensity of the transmitted light (contrast) when a voltage of 5
volts was applied (during "on") and when no voltage was applied (during "off") in the
direction of 90° from the direction of the stripe of the projection and in the direction
of deflection angle of 70° from the normal line of the cell surface was compared.
With the sample B, the contrast was 8.3, while with the sample A, the contrast was
25 18, so that the contrast-promoting effect by the polyimide retarder was observed.
Further, with the sample A, high quality white display without yellowing was
attained.

[0151]

Example 17

Using the color filter with the retarder prepared in Example 14, a test LCD element (sample C) was prepared in the same manner as in Example 16. The ratio of the intensity of the transmitted light (contrast) was 18, so that the contrast-promoting effect by the polyimide retarder was observed. Further, with the sample A, high quality white display without yellowing was attained.

[0152]

Example 18

Using the color filter with the retarder prepared in Example 15, a test LCD element (sample D) was prepared in the same manner as in Example 16. The ratio of the intensity of the transmitted light (contrast) was 18, so that the contrast-promoting effect by the polyimide retarder was observed. Further, high quality white display without yellowing was attained.

[0153]

Comparative Example 6

A polyimide thin film was formed on the color filter in the same manner as in Example 13 except that the polyamic acid solution O (polymer concentration 16% by weight) prepared in Comparative Example 3 was used. The thickness of the polyimide thin film was 0.9 μm , the birefringence Δn was 0.229. Thus, a color filter having a retarder which had a retardation of 206 nm, whose optical axis was vertical to the thin film, and which had an optically negative refractive index anisotropy, was obtained.

[0154]

Using the color filter thus prepared, a test LCD element (sample E) was prepared in the same manner as in Example 16. The ratio of the intensity of the transmitted light (contrast) was 15. However, the white display was yellowish, so

that the image display quality was poor.

[0155]

The results of Examples 1 to 12 and Comparative Examples 1-5 are shown in Table 1. As shown in Table 1, in Examples 1 to 12, retarders with slight coloring and having good birefringence were obtained. In Table 1, the abbreviations indicate the following:

[0156]

BPDA: 3,3',4,4'-biphenyltetracarboxylic dianhydride

CBDA: 1,2,3,4-cyclobutanetetracarboxylic dianhydride

10 H-PMDA: 1,2,4,5-cyclohexanetetracarboxylic dianhydride

ODPA: 4,4'-oxydiphthalic acid dianhydride

PMDA: pyromellitic dianhydride

PA: phthalic anhydride

DABA: 4,4'-diaminobenzanilide

15 t-DACH: trans-1,4-diaminocyclohexane

DDE: 4,4'-diaminodiphenyl ether

DDS: 3,3'-diaminodiphenylsulfone

PDA: *p*-phenylenediamine

SiDA: bis-(3-aminopropyl)tetramethyldisiloxane

20 m-TB-HG: 2,2'-dimethylbenzidine

TFMB: 2,2'-bis(trifluoromethyl)benzidine

[0157]

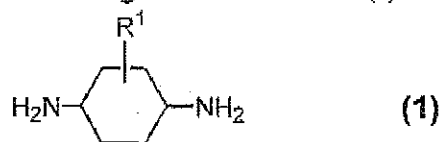
Table 1

	Acid Dianhydride (mol%)	Diamine (mol%)	End Cap Compound (mol%)	Cure Temperature	Birefringence Δn	Coloring	
						Δx	Δy
Example 1	BPDA(96)	t-DACH(96) SIDA(4)	PA(8)	240°C 270°C	0.091 0.174	0.0008 0.0016	0.0010 0.0017
Example 2	BPDA(96) PMDA(10)	t-DACH(96) SIDA(4)	PA(8)	270°C	0.112	0.0011	0.0012
Example 3	BPDA(96)	t-DACH(96) SIDA(4)	—	240°C 270°C	0.114 0.163	0.0009 0.0016	0.0009 0.0018
Example 4	BPDA(100)	t-DACH(100)	—	270°C	0.174	0.0020	0.0025
Example 5	ODPA(96)	t-DACH(100)	—	270°C	0.016	0.0022	0.0028
Example 6	CBDA(96)	PDA(96) SIDA(4)	PA(8)	240°C 270°C	0.036 0.045	0.0009 0.0027	0.0011 0.0035
Example 7	CBDA(96)	DABA(96) SIDA(4)	PA(8)	240°C 270°C	0.065 0.087	0.0015 0.0042	0.0018 0.0050
Example 8	CBDA(96)	m-TB-HG(96) SIDA(4)	PA(8)	240°C 270°C	0.038 0.039	0.0014 0.0034	0.0018 0.0048
Example 9	CBDA(96)	TFMB(96) SIDA(4)	PA(8)	240°C 270°C	0.020 0.018	0.0003 0.0008	0.0002 0.0009
Example 10	CBDA(96)	DOE(96) SIDA(4)	PA(8)	240°C 270°C	0.013 0.015	0.0011 0.0027	0.0016 0.0035
Example 11	CBDA(100)	PDA(100)	—	240°C	0.051	0.0016	0.0020
Example 12	CBDA(100)	DABA(100)	—	240°C	0.087	0.0031	0.0042
Comparative Example 1	BPDA(96)	DDS(48) PDA(48) SIDA(4)	PA(8)	270°C	0.081	0.0070	0.0110
Comparative Example 2	BPDA(96)	PDA(96) SIDA(4)	PA(8)	240°C 270°C	0.189 0.215	0.0094 0.0106	0.0185 0.0177
Comparative Example 3	BPDA(96)	DABA(96) SIDA(4)	PA(8)	240°C 270°C	0.216 0.229	0.0160 0.0155	0.0288 0.0275
Comparative Example 4	H-PMDA(100)	PDA(100)	—	240°C	0.002	0.0011	0.0011
Comparative Example 5	H-PMDA(100)	DABA(100)	—	240°C	0.006	0.0014	0.0016

CLAIMS

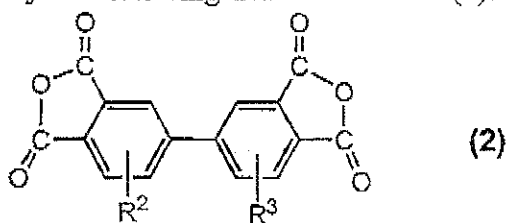
1. A resin composition for forming a retarder which is used for liquid crystal displays, which has an optically negative uniaxial anisotropy, whose optical axis is substantially vertical to the film plane of said retarder, and which has a birefringence Δn in the direction of thickness of 0.01 to 0.3, said composition comprising a polyimide precursor and an organic solvent, said polyimide precursor being obtained by reacting at least one tetracarboxylic dianhydride and at least one diamine; at least one of said at least one tetracarboxylic dianhydride and said at least one diamine being an alicyclic compound.

2. The composition according to claim 1, wherein said diamine which is said alicyclic compound is a *trans*-1,4-diaminocyclohexane compound represented by the following General Formula (1):



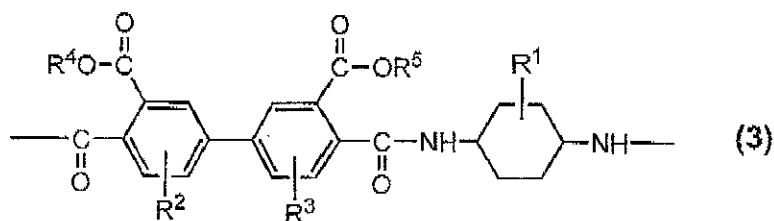
(wherein R^1 represents a monovalent organic group or a hydrogen atom).

3. The composition according to claim 2, wherein said tetracarboxylic dianhydride is a 3,3',4,4'-biphenyltetracarboxylic dianhydride compound represented by the following General Formula (2):



(wherein R^2 and R^3 independently represent a monovalent organic group or a hydrogen atom).

4. The composition according to claim 3, wherein said polyimide precursor has at least the structural unit represented by the following General Formula (3):



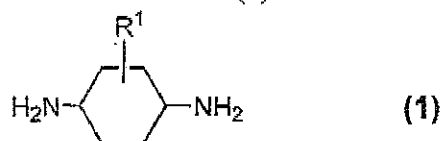
(wherein R^1 , R^2 , R^3 , R^4 and R^5 independently represent a monovalent organic group or a hydrogen atom).

5. The composition according to claim 1, wherein said tetracarboxylic dianhydride which is said alicyclic compound is 1,2,3,4-cyclobutanetetracarboxylic dianhydride.

6. The composition according to claim 5, wherein said diamine is an aromatic diamine having a rigid molecular structure.

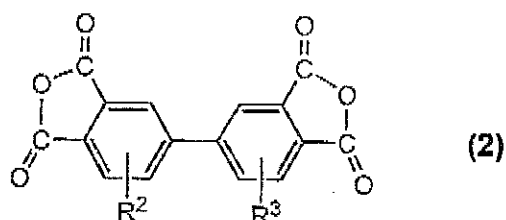
7. The composition according to claim 6, wherein said aromatic diamine having a rigid molecular structure is at least one selected from the group consisting of *p*-phenylenediamine and 4,4'-diaminobenzanilide.

8. A resin composition for forming a retarder which is used for liquid crystal displays, which has an optically negative uniaxial anisotropy, and whose optical axis is substantially vertical to the film plane of said retarder, said composition comprising a polyamic acid compound obtained by reacting a diamine component(s) containing a *trans*-1,4-diaminocyclohexane compound represented by the following General Formula (1):



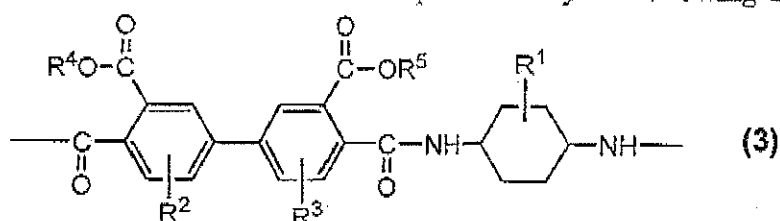
(wherein R^1 represents a monovalent organic group or a hydrogen atom)

and a tetracarboxylic dianhydride(s) containing a 3,3',4,4'-biphenyltetracarboxylic dianhydride compound represented by the following General Formula (2):



(wherein R^2 and R^3 independently represent a monovalent organic group or a hydrogen atom).

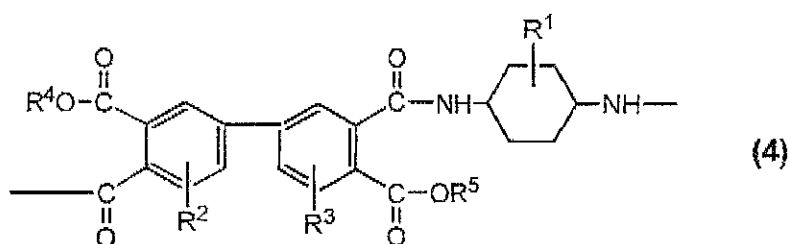
9. The composition according to claim 8, wherein said polyamic acid compound has at least the structural unit represented by the following General Formula (3):

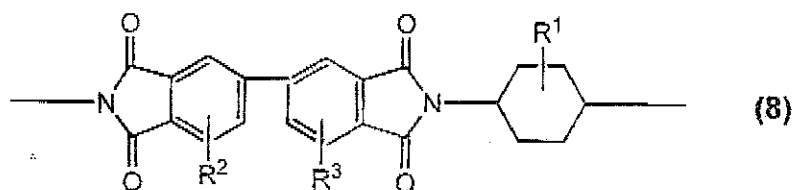
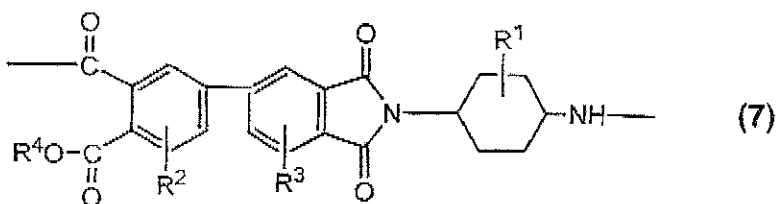
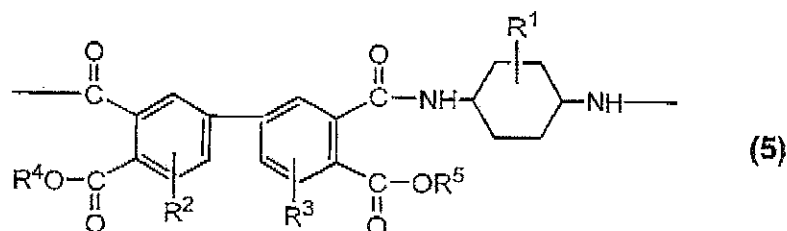


5 (wherein R^1 , R^2 , R^3 , R^4 and R^5 independently represent a monovalent organic group or a hydrogen atom).

10. The composition according to claim 9, wherein said R^1 is a hydrogen atom or a C_1 - C_4 linear or branched alkyl group; and R^2 , R^3 , R^4 and R^5 are each a hydrogen atom.

10 11. The composition according to claim 9 or 10, wherein said polyamic acid compound further comprises at least one structural unit represented by the following General Formulae (4) to (8):





(wherein in Formulae (4), (5), (6), (7) and (8), R^1 , R^2 , R^3 , R^4 and R^5 independently represent a monovalent organic group or a hydrogen atom).

12. The composition according to claim 10, wherein in said Formulae (4), (5), (6), (7) and (8), said R^1 is a hydrogen atom or a C_1 - C_4 linear or branched alkyl group; and R^2 , R^3 , R^4 and R^5 are each a hydrogen atom.

13. The composition according to claim 11 or 12, wherein the total amount of the structural units represented by the Formula (3), (4), (5), (6), (7) or (8) is not less than 50 mol% of the total structural units constituting said polyamic acid.

14. The resin composition according to any one of claims 1 to 13, wherein a part or all of the amine terminal groups of said polyimide precursor or said polyamic acid are capped by an amic acid-forming reaction with a dicarboxylic anhydride.

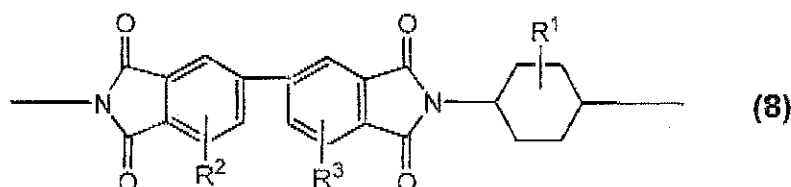
15. The composition according to claim 14, wherein said dicarboxylic anhydride is at least one selected from the group consisting of maleic anhydride, phthalic anhydride, succinic anhydride and nadic anhydride.

16. Use of said composition according to any one of claims 1 to 15 for the

production of a resin composition for forming a retarder.

17. A color filter substrate for liquid crystal displays, comprising a transparent substrate on which red, blue and green picture elements are two-dimensionally arrayed; and a retarder formed from said resin composition for forming a retarder according to any one of claims 1 to 15.

18. The color filter substrate according to claim 17, wherein said retarder comprises a polyimide containing structural units represented by the following General Formula (8):



(wherein R^1 , R^2 and R^3 independently represents a monovalent organic group or a hydrogen atom).

in an amount of not less than 50 mol%.

19. The color filter substrate according to claim 18, wherein said R^1 is a hydrogen atom or a C_1 - C_4 linear or branched alkyl group; and R^2 and R^3 are each a hydrogen atom.

20. The color filter substrate according to any one of claims 17 to 19, wherein said retarder formed from said resin composition for forming the retarder is so formed as to cover said picture elements.

21. A liquid crystal display comprising said color filter substrate for liquid crystal displays according to any one of claims 17 to 20, said liquid color display employing a display mode wherein the liquid crystal molecules are aligned in the direction substantially perpendicular to the liquid crystal cell surface when no electric voltage is applied, and the liquid crystal molecules are aligned in the direction substantially parallel to the liquid crystal cell surface when an electric voltage is applied.

22. A process for producing a color filter substrate for liquid crystal displays, said

process comprising coating said resin composition for forming a retarder according to any one of claims 1 to 15 on the surface of said color filter substrate, on which surface red, blue and green picture elements are two-dimensionally arrayed; and heating said composition.