

DESCRIPTION**Title of Invention**

STYRENE RESIN COMPOSITION, MOLDED ARTICLE, AND LIGHT GUIDE PLATE

Technical Field

[0001]

The present invention relates to a styrene-based resin composition, a molded article, and a light guide plate.

Background Art

[0002]

There are two types of backlights for liquid crystal displays: the direct-light type, where the light source is placed in front of the display unit, and the edge-light type, where the light source is placed on the side of the display unit. The edge-light type backlight uses a component called a light guide plate which guides the light from the light source placed on the side to the front of the display unit.

Acrylic resin, represented by polymethyl methacrylate (PMMA), is used as the material for light guide plates. However, since PMMA has high water absorption, the light guide plate may warp or change in size due to moisture absorption. In addition, since it is easily thermally decomposed during molding, there is a problem that the molded article tends to have a poor appearance when molded at a high temperature. To improve these problems, it has been proposed to use styrene-(meth)acrylic acid methyl copolymer as a material for a light guide plate (see, for example, Patent Literature 1).

Citation List**Patent Literature**

[0003]

Patent Literature 1 JP2003-075648

Summary of Invention**Technical Problem**

[0004]

An object of the present invention is to provide a styrene-based resin composition a molded article thereof having excellent moisture and heat resistance, and a light guide plate.

Solution to Problem

[0005]

The present invention is as follows.

(1) A styrene-based resin composition, wherein when an oxidation induction time of the styrene-based resin composition measured in an oxygen atmosphere of 200°C is t₁ and an oxidation induction time of the styrene-based resin composition measured in an oxygen atmosphere of 200°C after wet heat treatment for 500 hours in an air atmosphere of 80°C and 90% humidity is t₂, t₁-t₂ is 20 minutes or less.

(2) The styrene-based resin composition of (1), wherein t₁ is 50 minutes or more.

(3) The styrene-based resin composition of (1) or (2), comprising a hindered phenol-based antioxidant (B) and a phosphorus-based antioxidant (C2) which does not have a phenolic hydroxyl group.

(4) The styrene-based resin composition of any one of (1) to (3), comprising a styrene-based resin (A) having a styrene-based monomer unit and a (meth)acrylic ester-based monomer unit.

(5) The styrene-based resin composition of any one of (1) to (4), comprising a phosphorus-based antioxidant (C1) having a phenolic hydroxyl group.

(6) The styrene-based resin composition of (3), wherein the hindered -based phenol antioxidant (B) is at least one compound selected from a group consisting of octadecyl-3-(3,5-di-tert-butyl-4 hydroxyphenyl)propionate, ethylene bis(oxyethylene) bis [3- (5-tert-butyl-4-hydroxy-m-tolyl) propionate], and pentaerythritol tetrakis [3-(3,5-di-tert-butyl-4-hydroxyphenyl) propionate].

(7) The styrene-based resin composition of (3), wherein the phosphorus -based antioxidant (C2) is at least one compound selected from a group consisting of 2,2'-methylenebis(4,6-di-tert-butylphenyl)2-ethylhexylphosphite and tris(2,4-di-tert-butylphenyl) phosphite.

(8) The styrene-based resin composition of (5), wherein the phosphorus-based antioxidant (C1) is 6- [3-(3-tert-butyl-4-hydroxy-5-methylphenyl) propoxy] -2,4,8,10-tetra-tert-butyl-dibenzo [d, f] [1,3,2] dioxaphosphine.

(9) The styrene-based resin composition of any one of (1) to (8), wherein an initial YI value in an optical path length of 115 mm is 2.5 or less.

(10) A molded article comprising the styrene-based resin composition of any one of (1) to (9),

(11) A light guide plate comprising the molded article of (10).

Advantageous Effects of Invention

[0006]

According to the present invention, a styrene-based resin composition and a molded article thereof having excellent moisture and heat resistance, and a light guide plate can be obtained.

Brief Description of the Drawings

[0007]

Fig.1 is a conceptual diagram of the method of measuring the oxidation induction time by a chemiluminescence method (chemical luminescent method).

Description of Embodiments

[0008]

In the present specification, for example, the expression "A to B" means A or more and B or less.

[0009]

When an oxidation induction time of the styrene-based resin composition measured in an oxygen atmosphere of 200°C is t1 and an oxidation induction time of the styrene-based resin composition measured in an oxygen atmosphere of 200°C after wet heat treatment for 500 hours in an air atmosphere of 80°C and 90% humidity is t2, t1-t2 is 20 minutes or less, preferably 15 minutes or less, and more preferably 10 minutes or less. By setting t1-t2 in such a range, deterioration of hue when stored in a high-temperature and high-humidity environment can be suppressed. Further, although an increase in the amount of the antioxidant added contributes to the maintenance of the oxidation induction time, on the other hand, it may adversely affect the hue. Therefore, when considering the balance, t1-t2 is preferably 1 minute or more, more preferably 2 minutes or more, and even more preferably 4 minutes or more. Specifically, t1-t2 is, for example, 0, 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14,

15, 16, 17, 18, 19, 20 minutes, and may be within the range between any two of the numerical values exemplified here.

[0010]

t1 is preferably 50 minutes or longer, more preferably 70 minutes or longer, and even more preferably 90 minutes or longer. When t1 is in such a range, yellowing in a high temperature environment can be suppressed. Further, although an increase in the amount of the antioxidant added contributes to prolonging the oxidation induction time, on the other hand, it may adversely affect the hue. Therefore, when considering the balance, t1 is preferably 500 minutes or less and more preferably 200 minutes or less.

[0011]

The oxidation induction time is a value measured by the chemiluminescence method (chemical luminescent method). Changes in the luminescence amount of the styrene-based resin composition over time were measured under the following conditions. From the relationship between the obtained measurement time and the amount of light emitted, the time of the intersection of the straight line before the amount of luminescence changed and the straight line after the amount of luminescence increased was calculated as shown in Fig. 1. The styrene-based resin composition is gradually oxidized when heated in the presence of oxygen. When an antioxidant is present in the sample, the antioxidant is gradually consumed by oxidation but a constant amount of luminescence is observed during the presence of the antioxidant. That is, the oxidation induction time represents the time at which the antioxidant is consumed and the resin composition is rapidly oxidized.

Chemiluminescence measuring instrument: CLA-FS4 (manufactured by Tohoku Denshi Sangyo Co., Ltd.)

Measurement temperature: 200°C

Oxygen flow rate: 100 mL / min

[0012]

The styrene-based resin composition of the present invention preferably comprises a styrene-based resin (A) which is a copolymer of a styrene-based monomer and a (meth) acrylic acid ester-based monomer.

[0013]

The styrene-based monomer is an aromatic vinyl-based monomer. Examples of the styrene-based monomer include styrene, α -methylstyrene, o-methylstyrene, m-methylstyrene, ethylstyrene, and p-tert-butylstyrene. The styrene-based monomer may be used alone or a mixture of two or more may be used. Among these monomers, styrene is preferably used from the viewpoint of a good hue.

[0014]

Examples of the (meth) acrylic acid ester-based monomer include (meth) acrylic ester such as methyl (meth) acrylate, ethyl (meth) acrylate, butyl (meth) acrylate, 2-ethylhexyl (meth) acrylate, methyl acrylate, ethyl acrylate, n.-butyl acrylate, 2-methylhexyl acrylate, 2-ethylhexyl acrylate, decyl acrylate. The (meth) acrylic acid ester-based monomer alone or a mixture of two or more may be used. From the viewpoint of excellent hue and heat resistance, methyl (meth) acrylate is preferably used.

[0015]

The styrene-based resin (A) preferably has a content of a styrene-based monomer unit of 20 to 80% by

mass and a content of a (meth) acrylic acid ester-based monomer unit of 80 to 20% by mass. Preferably, the content of the styrene-based monomer unit is 30 to 60% by mass, and the content of the (meth) acrylic acid ester-based monomer unit is 70 to 40% by mass. When the content of the monomer units is in such a range, the water absorption rate and the deformation rate (water absorption capacity) are low, an increase of warp or change in size due to moisture absorption can be suppressed, and the deterioration of the hue and the scratches due to a decrease in surface hardness can be suppressed.

[0016]

The styrene-based resin (A) may be copolymerized with a monomer other than the (meth) acrylic acid ester-based monomer in a range of 5% by mass or less. Examples of the monomer to be copolymerized include a vinyl monomer copolymerizable with a styrene-based monomer and a (meth) acrylic acid ester-based monomer and include acrylonitrile, methacrylic acid, acrylic acid, and maleic anhydride.

[0017]

The content of the styrene-based monomer unit and the (meth) acrylic acid ester-based monomer unit in the styrene-based resin can be measured by pyrolysis gas chromatography under the following conditions.

Pyrolysis furnace: PYR-2A (manufactured by Shimadzu Corporation)

Pyrolysis furnace set temperature: 525°C

Gas chromatograph: GC-14A (manufactured by Shimadzu Corporation)

Column: 3 mm diameter × 3 m, made of glass

Filler: FFAP Chromsorb WAW

Column temperature: 120°C

Carrier gas: Nitrogen

[0018]

As a method for producing the styrene-based resin, a known method such as bulk polymerization, a solution polymerization, a suspension polymerization, and an emulsion polymerization can be used. As an operation method of the reactor, any of a continuous type, a batch type and a semi-batch type can be applied. From the viewpoint of quality such as transparency and productivity, the bulk polymerization or the solution polymerization is preferable, and the continuous type is preferable. Examples of the solvent for the bulk polymerization or the solution polymerization include alkylbenzenes such as benzene, toluene, ethylbenzene and xylene, ketones such as acetone and methyl ethyl ketone, and aliphatic hydrocarbons such as hexane and cyclohexane.

[0019]

A known method can be adopted as the polymerization method of the styrene-based resin. A radical polymerization method is preferable because it is a simple process and has excellent productivity.

[0020]

In the bulk polymerization or the solution polymerization of the styrene-based resin, a polymerization initiator and a chain transfer agent can be used, and the polymerization temperature is preferably in the range of 110 to 170°C. When continuous type bulk polymerization or continuous type solution polymerization is carried out, the conversion rate of the styrene-based monomer and the (meth) acrylic acid ester-based monomer is preferably

60% or more at the outlet of the polymerization step from the viewpoint of productivity.

[0021]

Examples of the polymerization initiators include organic peroxides such as benzoyl peroxide, tert-butylperoxybenzoate, 1,1-di (t-butylperoxy) cyclohexane, 1,1-bis (t-butylperoxy) -3,3,5-trimethylcyclohexane, 1,1-bis (t-butylperoxy) -3,3,5-trimethylcyclohexane, 2,2-bis (4,4-di-tert-butylperoxycyclohexyl) propane, tert-butylperoxy isopropyl carbonate, dicumyl peroxide, tert-butyl cumyl peroxide, tert-butyl peroxyacetate, tert-butyl peroxy-2-ethylhexanoate, polyether tetrakis (tert-butyl peroxy carbonate), ethyl-3, 3-di (tert-butylperoxy) butyrate, tert-butylperoxyisobutyrate and the like.

[0022]

The amount of the polymerization initiator added is preferably 0.001 to 0.2% by mass, more preferably 0.001 to 0.05% by mass, with respect to 100% by mass of the total amount of the monomers. When the amount of the polymerization initiator added is too large, the hue may deteriorate.

[0023]

Examples of the chain transfer agent include aliphatic mercaptans, aromatic mercaptans, pentaphenylethane, α -methylstyrene dimer, terpinolene and the like.

[0024]

The amount of the chain transfer agent added is preferably 0.001 to 0.5% by mass, more preferably 0.005 to 0.2% by mass, with respect to 100% by mass of the total amount of the monomers. When the amount of the chain transfer agent added is 0.001 to 0.5% by mass, the thermal stability is good.

[0025]

A known method can be used as a devolatilization method for removing volatile components such as the unreacted monomer and solvent used for the solution polymerization from the solution after completion of polymerization of the styrene-based resin. For example, a vacuum devolatilizing tank equipped with a preheater or a devolatilizing extruder with a vent can be used. The temperature of the styrene-based resin in the devolatilization step is preferably 200°C to 300°C, more preferably 220°C to 260°C. When the temperature of the styrene-based resin in the devolatilization step is too high, the hue may deteriorate. The devolatilized molten styrene-based resin is transferred to a granulation process, extruded from a porous die in a strand form, and processed into a pellet shape by a cold cut method, an air hot cut method, or an underwater hot cut method.

[0026]

The unreacted monomer and the solvent used for solution polymerization removed in the devolatilization step are preferably recovered, purified to remove impurities such as a polymerization inhibitor, mixed with a fresh raw material and used as a recovery raw material. By using the recovered raw material in combination with a fresh raw material which does not contain a polymerization inhibitor, it is possible to reduce the content of the polymerization inhibitor in the raw material supplied to the polymerization step. The content of the polymerization inhibitor in the raw material supplied to the polymerization step is preferably less than 12 ppm, more preferably less than 9 ppm, further preferably less than 6 ppm, and most preferably less than 4 ppm. When the content of the polymerization inhibitor in the raw material supplied to the polymerization step is less than 12 ppm, the transmittance and transparency are good. It is difficult to remove all the polymerization inhibitors, and

the content of the polymerization inhibitor may be 0.01 ppm or more. In order to distinguish the raw material newly supplied to the production process of the styrene- (meth) acrylic acid ester-based copolymer from the recovered raw material, the raw material newly supplied is referred to as a fresh raw material.

[0027]

As a method for recovering and purifying the unreacted monomer and the solvent used for solution polymerization removed in the devolatilization step, a known method can be adopted. Examples of the method include a method of condensing the gas of the unreacted monomer and solvent, removed in the volatilization step, with a condenser, liquefying it, and purifying it with a flash distillation column to separate and remove high boiling point components. In addition, they include a method of condensing and separating only the high boiling point components from the gas of the unreacted monomer and solvent removed in the volatilization step with a condenser or spray tower, and then condensing the remaining gas with a condenser. The polymerization inhibitor 4-tert-butylcatechol has a boiling point of 285°C and 6-tert-butyl-2,4-xylenol has a boiling point of 249°C. They can be separated and removed as high boiling point components from the monomer and solvent. (The boiling point of styrene is 145°C, the boiling point of methyl (meth) acrylate is 101°C, and the boiling point of ethylbenzene is 136°C).

[0028]

The weight average molecular weight (Mw) of the styrene-based resin is preferably 50,000 to 450,000, more preferably 70,000 to 300,000, and even more preferably 70,000 to 200,000. When the weight average molecular weight (Mw) is less than 50,000, the strength of the light guide plate may decrease. When the weight average molecular weight (Mw) exceeds 200,000, the fluidity may decrease and the molding processability may deteriorate. The weight average molecular weight (Mw) can be controlled by the reaction temperature, the residence time in the polymerization step, the type and addition amount of the polymerization initiator, the type and addition amount of the chain transfer agent, and the type and amount of the solvent used in the polymerization step, and the like.

[0029]

The weight average molecular weight (Mw) can be measured by gel permeation chromatography (GPC) under the following conditions.

PC model: Shodex GPC-101 manufactured by Showa Denko K.K.

Column: PLgel 10 μ m MIXED-B manufactured by Polymer Laboratories

Mobile phase: tetrahydrofuran

Sample concentration: 0.2% by mass

Temperature: Oven 40°C, inlet 35°C, detector 35°C

Detector: differential refractometer

The molecular weight of the present invention was calculated as the polystyrene-equivalent molecular weight by calculating the molecular weight at each elution time from the elution curve of monodisperse polystyrene.

[0030]

The total amount of the residual monomer and the residual polymerization solvent in the styrene-based

resin is preferably 0.5% by mass or less, more preferably 0.2% by mass. When the total amount of the residual monomer and the polymerization solvent exceeds 0.5% by mass, the heat resistance may be insufficient.

[0031]

The residual monomer and the residual polymerization solvent are the amounts of the monomer and the polymerization solvent remaining in the styrene-based resin and include styrene, methyl (meth) acrylate, and ethylbenzene. The amount of the residual monomer and the residual polymerization solvent can be adjusted by the constituents of the volatilization step and the conditions of the volatilization step.

[0032]

The amount of the residual monomer and the residual polymerization solvent were measured using a capillary gas chromatograph under the following conditions. 0.2 g of styrene-based resin was precisely weighed and dissolved in 10 mL of tetrahydrofuran containing p-diethylbenzene as an internal standard substance to prepare a sample.

Capillary gas chromatograph: GC-4000 (manufactured by GL Sciences Inc.)

Column: InertCap WAX, inner diameter 0.25 mm, length 30 m, film thickness 50 μ m manufactured by GL Sciences Inc.

Injection temperature: 180°C

Column temperature: 60°C to 170°C

Detector temperature: 210°C

Split ratio: 5/1

[0033]

The total amount of the dimer or trimer (hereinafter referred to as oligomer) of the styrene-based monomer and the (meth) acrylic acid ester-based monomer in the styrene-based resin is preferably 2% by mass or less. It is more preferably 1% by mass or less. When the total amount of oligomers exceeds 1% by mass, the heat resistance of the light guide plate may be insufficient.

[0034]

The oligomer was measured using a gas chromatograph under the following conditions. 200 mg of a styrene-based resin was dissolved in 2 mL of 1,2-dichloromethane, 2 mL of methanol was added to the solution to precipitate a polymer, and the mixture was allowed to stand, and then the supernatant obtained was used as a sample.

Gas chromatograph: HP-5890 (manufactured by Hewlett-Packard Company)

Column: DB-1 (ht), 0.25 mm $\times$ 30 m, Film thickness 0.1 μ m

Injection temperature: 250°C

Column temperature: 100 -300°C

Detector temperature: 300°C

Split ratio: 50/1

Internal standard substance: n-eicosane

Carrier gas: Nitrogen

[0035]

The styrene-based resin composition of the present invention preferably comprises a hindered phenol-based antioxidant (B). Examples of the hindered phenol-based antioxidant include octadecyl-3-(3,5-di-tert-butyl-4-hydroxyphenyl) propionate, ethylene bis (oxyethylene) bis [3-(5-tert-butyl-4-hydroxy-m-tolyl) propionate], 3,9-bis [2-[3-(3-tert-butyl-4-hydroxy-5-methylphenyl) propionyloxy]-1,1-dimethylethyl]-2,4,8,10-tetraoxaspiro [5.5] undecane, pentaerythritol tetrakis [3-(3,5-di-tert-butyl-4-hydroxyphenyl) propionate], 4,6-bis (octylthiomethyl)-o-cresol, 4,6-bis[(dodecylthio) methyl]-o-cresol, 2,4-dimethyl-6-(1-methylpentadecyl) phenol, tetrakis [methylene-3-(3,5-di-tert-butyl-4-hydroxyphenyl) propionate] methane, 4,4'-thiobis (6-tert-butyl-3-methylphenol), 1,1,3-tris (2-methyl-4-hydroxy-5-tert-butylphenyl) butane, 4,4'-butylidenebis (3-methyl-6-tert-butylphenol), bis-[3,3-bis-(4'-hydroxy-3'-tert-butylphenyl)-butanoic acid]-glycol ester, 2-tert-butyl-6-(3-tert-butyl-2-hydroxy-5-methylbenzyl)-4-methylphenyl acrylate, 2-[1-(2-hydroxy-3,5-di-tert-pentylphenyl) ethyl]-4,6-di-tert-pentylphenyl acrylate and the like. The hindered phenol-based antioxidant may be used alone, or two or more kinds thereof may be used in combination.

[0036]

The content of the hindered phenol-based antioxidant (B) is preferably 0.001 to 0.3 parts by mass, and more preferably 0.03 to 0.09 parts by mass with respect to 100 parts by mass of the styrene-based resin (A). By adjusting the content of the hindered phenol-based antioxidant (B) within this range, a styrene-based resin composition having an excellent hue can be obtained.

[0037]

The styrene-based resin composition of the present invention preferably comprises a phosphorus-based antioxidant (C1) having a phenolic hydroxyl group. The phosphorus-based antioxidant (C1) contained in the styrene-based resin composition is a trivalent phosphorus compound having a phenolic hydroxyl group in the basic skeleton. The phosphorus-based antioxidant (C1) is easily hydrolyzed as compared with other phosphorus-based antioxidants and has a high effect of improving the hue of the obtained styrene-based resin composition. The phosphorus-based antioxidant (C1) is 6- [3- (3-tert-butyl-4-hydroxy-5-methylphenyl) propoxy] -2,4,8,10-tetra-tert-butylidibenzo [d, f] [1,3,2] dioxaphosphine and the like.

[0038]

The content of the phosphorus-based antioxidant (C1) is preferably 0.001 to 0.3 parts by mass, more preferably 0.03 to 0.09, and more preferably 0.05 to 0.08 parts by mass, with respect to 100 parts by mass of the styrene-based resin (A). By adjusting the content of the phosphorus-based antioxidant (C1) within this range, a styrene-based resin composition having an excellent hue can be obtained.

[0039]

The styrene-based resin composition of the present invention preferably contains a phosphorus-based antioxidant (C2) other than the phosphorus-based antioxidant (C1) having a phenolic hydroxyl group. The phosphorus-based antioxidant (C2) contained in the styrene-based resin composition is a trivalent phosphorus compound having no phenolic hydroxyl group in the basic skeleton. The phosphorus-based antioxidant (C2) is less likely to be hydrolyzed than the phosphorus-based antioxidant (C1), but the effect of improving the hue of the

styrene-based resin composition is long-lasting. That is, it can particularly contribute to moisture and heat resistance. Examples of the phosphorus antioxidant (C2) is 3,9-bis (2,6-di-tert-butyl-4-methylphenoxy) -2,4,8,10-tetraoxa-3,9-diphosphaspiro [5. 5] undecane, bis (2,4-dicumylphenyl) pentaerythritol diphosphite, 2,2'-methylenebis (4,6-di-tert-butylphenyl) 2-ethylhexyl phosphite, tris (2,4-di-tert-butylphenyl) phosphite, bis [2,4-bis (1,1-dimethylethyl) -6-methylphenyl] ethyl ester phosphorous acid, bis (2,4-di-tert-butylphenyl) pentaerythritol diphosphite, cyclic neopentetetraylbis (octadecylphosphite), bis (nonylphenyl) pentaerythritol diphosphite, tetrakis(2,4-di-tert-butylphenyl) 4,4'-biphenylenediphosphinate, 9,10-dihydro-9-oxa-10-phosphaphenanthrene-10-oxide, tetrakis (2,4-di-tert-butyl-5-methylphenyl) -4,4'-biphenylenediphosphonite, and the like. From the viewpoint of sustaining the effect of improving hue, 2,2'-methylenebis (4,6-di-tert-butylphenyl) 2-ethylhexyl phosphite or tris (2,4-di-tert-butylphenyl) phosphite is preferable. The phosphorus-based antioxidant (C2) may be used alone, or two or more of these can be used in combination.

[0040]

The content of the phosphorus-based antioxidant (C2) is preferably 0.001 to 0.3 parts by mass, more preferably 0.03 to 0.09, and more preferably 0.05 to 0.08 parts by mass with respect to 100 parts by mass of the styrene-based resin (A). By adjusting the content of the phosphorus-based antioxidant (C2) within this range, the styrene-based resin composition having an excellent initial hue and an excellent hue over a long period of time can be obtained.

[0041]

The mixing ratio of the phosphorus-based antioxidant (C1) and the phosphorus-based antioxidant (C2) is preferably in the range of 3:1 to 1:3, and more preferably 2:1 to 1:2 by mass. By adjusting to this range, the balance between the hue and the durability of the obtained styrene-based resin composition is improved.

[0042]

As a method for producing the styrene-based resin composition, a known method can be adopted. Examples of the method include adding the hindered phenol-based antioxidant (B), the phosphorus-based antioxidant (C1), and the phosphorus-based antioxidant (C2) in the production process such as a polymerization step of styrene-based resin, a volatilization step, and a granulation step, and adding them after the unreacted monomer and the solvent are removed in the devolatilization step is preferable. When a vacuum devolatilizing tank is used, the method of adding molten hindered phenol-based antioxidant (B), a phosphorus-based antioxidant (C1), and a phosphorus-based antioxidant (C2) to the styrene-based resin extracted from the devolatilizing tank and mixing the mixture with a static mixer may be used. When using a devolatilizing extruder with a vent, the method of adding hindered phenol-based antioxidant (B), a phosphorus-based antioxidant (C1), and a phosphorus-based antioxidant (C2) and mixing after the vent zone may be used. The method of using an extruder and melt-kneading the hindered phenol-based antioxidant (B), the phosphorus-based antioxidant (C), and the phosphorus-based antioxidant (C2) with the styrene-based resin (A) after granulation may also be used.

[0043]

Mineral oil may be added to the styrene-based resin composition as long as the transparency is not impaired. In addition, an internal lubricant such as stearic acid and ethylene bisstearyl amide, a sulfur-based

antioxidant, a lactone-based antioxidant, an ultraviolet absorber, a hindered amine-based stabilizer, an antistatic agent, and an external lubricant such as ethylene bisstearyl amide may be added.

[0044]

The ultraviolet absorber has a function of suppressing deterioration and coloring due to ultraviolet rays, and Examples of them include benzophenone-based, benzotriazole-based, triazine-based, benzoate-based, salicylate-based, cyanoacrylate-based, malonic acid ester-based, formamidine-based ultraviolet absorber. These can be used alone or in combination of two or more, and a light stabilizer such as hindered amine may be used in combination.

[0045]

The Vicat softening temperature of the styrene-based resin is preferably 95°C or higher, more preferably 98°C or higher. When the Vicat softening temperature is less than 95°C, the heat resistance is insufficient, and the molded article may be deformed depending on the usage environment. (The Vicat softening temperature was measured based on JIS K 7206 at a heating rate of 50°C/hr and a test load of 50 N.)

[0046]

The styrene-based resin composition can be used to produce a plate-shaped molded article by a known method such as extrusion molding, injection molding, compression molding, blow molding, and the like and it can be used as a light guide plate or the like.

[0047]

Since the styrene-based resin composition of the present invention has excellent thermal stability, non-product parts such as sheet scraps generated in an extrusion molding and spools and runners generated in an injection molding can be recovered and crushed and mixed with a virgin raw material and the mixture can be used.

[0048]

The light guide plate is a member which has a function of guiding the light incident from the end face of the plate-shaped molded article to the surface side of the plate-shaped molded article and emitting the light from there by the reflection pattern formed on one surface of the plate-shaped molded article. The reflection pattern can be formed by a method such as a screen printing method, a laser processing method, or an inkjet method. Further, a prism pattern or the like can be provided on the surface (luminescence surface) opposite to the surface on which the reflection pattern is formed. The reflection pattern and the prism pattern of the plate-shaped molded article can be formed at the same time as the molding of the plate-shaped molded article and can be formed by a mold shape in the injection molding and a roll transfer in the extrusion molding.

[0049]

An initial YI value of the styrene-based resin composition measured in the optical path length of 115 mm is preferably 2.5 or less, more preferably 2.0 or less. Here, the "initial" means before the wet heat treatment for 500 hours in an air atmosphere of 80°C and 90% humidity. In the measurement, the spectral transmittance at a wavelength of 350 nm to 800 nm in an optical path length of 115 mm was measured, and the YI value at a field of view of 2° with a C light source was calculated according to JIS K7373. When the YI value measured in an optical path length of 115 mm is higher than 2.5, since the color changes with the optical path length, when used as a backlight, color unevenness may occur on the surface of the liquid crystal display device. Further, the average

value of the spectral transmittance at a wavelength of 350 nm to 800 nm measured in an optical path length of 115 mm is preferably 87% or more, more preferably 88% or more, and further preferably 89% or more.

EXAMPLES

[0050]

The present invention is described in further details below with Examples. The present embodiments are not limited to them.

[0051]

<Production Example of styrene-based resin A-1>

The styrene-based resin was produced by a continuous type solution polymerization by the radical polymerization method. A complete mixing tank type stirring tank was used as the first reactor, and a plug flow type reactor with a static mixer was used as the second reactor, and the first and the second reactor were connected in series to form a polymerization step. The capacity of the first reactor was 30 L, and the capacity of the second reactor was 12 L. Styrene (hereinafter referred to as fresh Sty), which is industrially used, was prepared as the styrene-based monomer and the concentration of 4-tert-butylcatechol (hereinafter referred to as TBC) in the styrene was 10.2 ppm. Methyl (meth) acrylate (hereinafter referred to as fresh MMA), which is industrially used, was prepared as a (meth) acrylic acid ester-based monomer, and the concentration of 6-tert-butyl-2,4-xyleneol (hereinafter referred to as TBX) in methyl (meth) acrylate was 4.9 ppm. Ethylbenzene (hereinafter referred to as fresh EB) used industrially was prepared as a polymerization solvent. Further, the gas such as the monomer and the polymerization solvent separated from the vacuum devolatilizing tank vacuum devolatilizing tank described later was condensed by a condenser and purified by a flash distillation column, which was used as a recovery raw material. The concentrations of TBX and TBC in the recovered raw material were below the lower limit of detection. Using fresh Sty, fresh MMA, and recovered raw materials, a raw material solution was prepared with a composition of Sty: 49% by mass, MMA: 41% by mass, and EB: 10% by mass and the raw material solution was continuously supplied at a flow rate of 8.0 kg/h in the polymerization step. The ratio of the recovered raw material used in the raw material solution was 33% by mass. Further, t-butylperoxyisopropyl monocarbonate as a polymerization initiator was continuously added to the raw material solution supply line so as to have a concentration of 150 ppm, and n-dodecyl mercaptan as a chain transfer agent was continuously added to the raw material solution supply line so as to have a concentration of 500 ppm. The temperature of the first reactor was adjusted to 135°C, the temperature of the second reactor was adjusted to have a temperature gradient along the direction of flow, the temperature at the intermediate part of the second reactor was adjusted to 130°C, and the temperature at the outlet part of the second reactor to be 145°C. The polymer concentration at the outlet of the polymerization step was 65%, and the conversion rate of styrene and methyl (meth) acrylate was 72%. The polymer solution continuously taken out from the reactor was supplied to a vacuum devolatilizing tank equipped with a preheater to separate unreacted styrene, methyl (meth) acrylate, ethylbenzene, and the like. The temperature of the preheater was adjusted so that the polymer temperature in the devolatilizing tank was 240°C, and the pressure in the devolatilizing tank was 1 kPa. The polymer was extracted from the vacuum devolatilizing tank by a gear pump, extruded into a strand shape, cooled with cooling water, and then cut to obtain a pellet-shaped styrene-based resin A-1. The composition of A-1 was Sty: 50% by mass and

MMA: 50% by mass. The weight average molecular weight of A-1 was 145,000, the total amount of the residual monomer and the residual polymerization solvent was 0.07% by mass, and the total amount of the residual oligomer was 0.35% by mass.

[0052]

<Production Example of styrene-based resin A-2>

The procedure was carried out in the same manner as in A-1 except that the raw material composition was changed to Sty: 77% by mass, MMA: 13% by mass, and EB: 10% by mass, the addition of n-dodecyl mercaptan was stopped, the temperature of the first reactor was set to 140°C, the temperature at the intermediate part of the second reactor was set to 140°C and the temperature at the outlet part of the second reactor was 160°C. The ratio of the recovered raw material used in the raw material solution was 33% by mass. The composition of A-2 was Sty: 82% by mass and MMA: 18% by mass. The weight average molecular weight of A-2 was 240,000, the total amount of the residual monomer and the residual polymerization solvent was 0.06% by mass, and the total amount of the residual oligomer was 0.33% by mass.

[0053]

<Production example of styrene-based resin A-3>

The procedure was carried out in the same manner as in A-1 except that the raw material composition was changed to Sty: 8% by mass, MMA: 79% by mass, and EB: 13% by mass, the feed flow rate was 5.7 kg/h, the concentration of tert-butylperoxyisopropyl monocarbonate was 100 ppm, the concentration of n-dodecyl mercaptan was 3000 ppm, the temperature of the first reactor was 122°C, the temperature at the intermediate part of the second reactor was 140°C, and the temperature at the outlet part of the second reactor was 150°C. The ratio of the recovered raw material used in the raw material solution was 34% by mass. The composition of A-3 was Sty: 10% by mass and MMA: 90% by mass. The weight average molecular weight of A-3 was 80,000, the total amount of the residual monomer and the residual polymerization solvent was 0.06% by mass, and the total amount of the residual oligomer was 0.34% by mass.

[0054]

<Examples 1 to 10, Comparative Example 1, Reference Examples 1 to 2>

To the styrene-based resins A-1 to A-3 obtained in the Production Example, the following hindered phenol-based antioxidants (B), phosphorus-based antioxidants (C1) and phosphorus-based antioxidants (C2-1), (C2-2) are added at the contents shown in Table 1, and, using a sheet extruder manufactured by Leader Extrusion Machinery Company Ltd., the mixture was melt-kneaded to obtain a sheet molded article having a size of 450 mm × 500 mm × 2 mm. The sheet extruder was composed of a 50 mmφ single-screw extruder, a T-die, and three mirror rolls, and the sheet was extruded at a cylinder temperature of 225°C and a screw rotation speed of 120 rpm of the single-screw extruder. The width of the T-die was 450 mm and the opening was 3 mm.

(B) Octadecyl-3- (3,5-di-tert-butyl-4-hydroxyphenyl) propionate (Irganox 1076, manufactured by BASF Japan Ltd.)

(C1) 6- [3- (3-tert-Butyl-4-hydroxy-5-methylphenyl) propoxy] -2,4,8,10-tetra-tert-butyl-dibenzo [d, f] [1,3, 2] dioxaphosphine (Sumilizer GP manufactured by SUMITOMO CHEMICAL COMPANY, LIMITED)

(C2-1) 2,2'-methylenebis (4,6-di-tert-butylphenyl) 2-ethylhexyl phosphite (ADEKA Corporation ADEKA

STAB HP-10 manufactured by ADEKA Corporation)

(C2-2) tris (2,4-di-tert-butylphenyl) phosphite (Irgafos 168, manufactured by BASF Japan Ltd.)

[0055]

(Oxidation induction time)

A test piece having a thickness of 30 mm × 30 mm × 2 mm was cut out from the obtained sheet molded article, and the amount of light emitted was measured at 200°C and an oxygen flow rate of 100 mL/min using a Chemiluminescence analyzer CLA-FS4 manufactured by Tohoku Denshi Sangyo Co., Ltd. The oxidation induction time was determined by the method shown in Fig. 1 from the relationship between the measurement time and the amount of light emitted. Table 1 shows the measurement results of the oxidation induction time (t1) of the sheet molded article immediately after extrusion and the oxidation induction time (t2) of the sheet molded article after wet heat treatment for 500 hours in an air atmosphere of 80°C and 90% humidity. In Reference Example 1 and Reference Example 2, the amount of light emitted continued to increase immediately after the start of measurement, and the oxidation induction time could not be measured.

[0056]

(Optical property in an optical path length of 115 mm)

A test piece having a size of 115 mm × 85 mm and thickness of 2 mm was cut out from the obtained sheet molded article, and the end face was polished to prepare a plate-shaped molded article having a mirror surface on the end face. Using an ultraviolet visible spectrophotometer V-670 manufactured by JASCO Corporation, the spectral transmittance of 350 nm to 800 nm of the plate-shaped molded article after polishing in an optical path length of 115 mm was measured with incident light having a size of 20 × 1.6 mm and a spreading angle of 0° and the YI value at a field of view of 2° with a C light source was calculated according to JIS K7373. The transmittance shown in Table 1 indicates an average transmittance of a wavelength of 380 nm to 780 nm. Table 1 shows the measurement results of the sheet molded article immediately after extrusion (initial stage), the sheet molded article after wet heat treatment for 500 hours in an air atmosphere of 80°C and 90% humidity, and the sheet molded article which was stored for 1000 hours in an environment of 80°C.

[0057]

(Water absorption capacity)

The obtained sheet molded article was cut to obtain a molded article having a size of 200 mm × 300 mm. This molded article is stored for 500 hours under the conditions of temperature 60°C and 90% humidity, and the change in mass and length of the long side before and after storage were measured. The water absorption rate and deformation rate were calculated by the following formula as an index of water absorption capacity.

(Water absorption rate) = ((Mass after storage) - (Mass before storage)) ÷ (Mass before storage) × 100 (%)

(Deformation rate) = ((Long side length after storage) - (Long side length before storage)) ÷ (Long side length before storage) × 100 (%)

Table 1 shows the evaluation results. When the water absorption rate was 1.0 or less, it was judged to be good, and when the deformation rate was 0.30 or less, it was judged to be good.

Table 1			Example											Comparative Example	Reference Example	
			1	2	3	4	5	6	7	8	9	10	1	2		
resin composition	styrene-based resin A-1		part	100	100	100	100	100	100	100	100	-	-	100	100	100
	styrene-based resin A-2		part	-	-	-	-	-	-	-	-	100	-	-	-	-
	styrene-based resin A-3		part	-	-	-	-	-	-	-	-	-	100	-	-	-
	phenol-based antioxidant B		part	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.01	0.05	0.05	0.05	0.05	-
	phosphorus-based antioxidant C1		part	0.03	0.03	0.06	0.02	0.03	-	-	-	0.03	0.03	0.06	-	-
	phosphorus-based antioxidant C2-1		part	0.03	0.06	0.06	0.02	-	0.06	-	-	0.03	0.03	-	-	-
	phosphorus-based antioxidant C2-2		part	-	-	-	-	0.03	-	0.06	0.01	-	-	-	-	0.06
properties	oxidation induction time (200°C in oxygen atmosphere)	t1	minute	90	102	121	68	73	72	52	33	47	106	79	-	-
		t2	minute	82	93	104	61	67	66	48	28	31	99	53	-	-
		t1-t2	minute	8	9	17	7	6	6	4	5	16	7	26	-	-
	optical properties in optical path length of 115 mm (initial stage)	transmission	%	89	89	88	88	89	88	88	88	88	89	89	86	86
		YI value	-	1.7	1.9	2.3	2.1	1.8	2.9	3.1	3.3	2.6	1.3	1.7	4.5	4.7
	optical properties in optical path length of 115 mm (After 80°C, 90% RH 500hr treatment)	transmission	%	88	88	88	88	88	88	87	87	87	88	86	85	85
		YI value	-	2.3	2.4	3.2	2.7	2.4	3.6	3.8	3.9	3.9	2.0	4.1	7.5	7.2
	change in hue after 80°C, 1000hr	ΔYI	-	1.9	1.8	1.9	2.3	1.9	1.9	1.8	2.3	2.5	1.7	1.7	3.3	3.1
	water absorption capacity	water absorption rate	%	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.2	1.2	0.5	0.5	0.5
		deformation rate	%	0.12	0.12	0.12	0.12	0.12	0.12	0.12	0.12	0.05	0.36	0.12	0.12	0.12

Industrial Applicability

[0059]

Since the styrene-based resin composition of the present invention and its molded article are excellent in moisture and heat resistance, the hue and transparency are not easily deteriorated. Thus, they can be suitably used for light guide plate applications such as and a television, a desktop personal computer, a notebook personal computer, a mobile phone, a car navigation system, and an interior lighting.

CLAIMS

1. A styrene-based resin composition, wherein when an oxidation induction time of the styrene-based resin composition measured in an oxygen atmosphere of 200°C is t₁ and an oxidation induction time of the styrene-based resin composition measured in an oxygen atmosphere of 200°C after wet heat treatment for 500 hours in an air atmosphere of 80°C and 90% humidity is t₂, t₁-t₂ is 15 minutes or less, the styrene-based resin composition comprises a styrene-based resin (A), a hindered phenol-based antioxidant (B), a phosphorus-based antioxidant (C1) having a phenolic hydroxyl group, and a phosphorus-based antioxidant (C2) which does not have a phenolic hydroxyl group, a content of the phosphorus-based antioxidant (C1) is 0.001 to 0.3 parts by mass with respect to 100 parts by mass of the styrene-based resin (A) a content of the phosphorus-based antioxidant (C2) is 0.001 to 0.3 parts by mass with respect to 100 parts by mass of the styrene-based resin (A).
2. The styrene-based resin composition of Claim 1, wherein t₁ is 50 minutes or more.
3. The styrene-based resin composition of Claims 1 or 2, wherein the styrene-based resin (A) has a styrene-based monomer unit and a (meth)acrylic ester-based monomer unit.
4. The styrene-based resin composition of Claim 1, wherein the hindered -based phenol antioxidant (B) is at least one compound selected from a group consisting of octadecyl-3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate, ethylene bis(oxyethylene) bis [3- (5-tert-butyl-4-hydroxy-m-tolyl) propionate], and pentaerythritol tetrakis [3- (3,5-di-tert-butyl-4-hydroxyphenyl) propionate].
5. The styrene-based resin composition of Claim 1, wherein the phosphorus-based antioxidant (C2) is at least one compound selected from a group consisting of 2,2'-methylenebis(4,6-di-tert-butylphenyl)2-ethylhexylphosphite and tris(2,4-di-tert-butylphenyl) phosphite.
6. The styrene-based resin composition of Claim 1, wherein the phosphorus-based antioxidant (C1) is 6-[3- (3-tert-butyl-4-hydroxy-5-methylphenyl) propoxy] -2,4,8,10-tetra-tert-butyl-dibenzo [d, f] [1,3,2] dioxaphosphine.
7. The styrene-based resin composition of any one of Claims 1 to 6, wherein an initial YI value in an optical path length of 115 mm is 2.5 or less.
8. A molded article comprising the styrene-based resin composition of any one of Claims 1 to 7.
9. A light guide plate comprising the molded article of Claim 8.