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(54) **Title**: FLAME-RETARDANT POLYESTER RESIN COMPOSITION AND MOLDED ARTICLE THEREOF

(57) **Abstract**: The present invention is a flame-retardant polyester resin composition comprising (A) a polyester resin with (B) a salt of a triazine-based compound with cyanuric acid or isocyanuric acid. The composition comprises the component (B) in an amount of 1 to 70 parts by weight based on 100 parts by weight of the component (A), and, in the composition, the number of particles of the component (B) with a maximum size of more than 15 pm is 1 or less per 100 μm x 100 μm cross section. The present invention is also a molded article made of the flame-retardant polyester resin composition. The present invention provides a flame-retardant polyester resin composition excellent in the balance of flame retardancy and mechanical properties without using a halogen-based flame retardant; and also a molded article.

FLAME-RETARDANT POLYESTER RESIN COMPOSITION AND MOLDED ARTICLE THEREOF

TECHNICAL FIELD

[0001]

The present invention relates to a flame-retardant resin composition comprising a non-halogen-based flame retardant incorporated into a polyester resin, and also to a molded article thereof.

BACKGROUND ART

[0002]

Polyester resins have been used in a wide range of fields, such as machine mechanism parts, electrical/electronic parts, and automobile parts, taking advantage of their excellent characteristics including injection moldability, mechanical properties, and the like.

[0003]

Polyester resins are essentially flammable. Therefore, when they are used as industrial materials for machine mechanism parts, electrical/electronic parts, automobile parts, and the like, safety against fire, i.e., flame retardancy, is required in addition to the balance of general chemical and physical characteristics. In such cases, high-level flame retardancy showing V0 of UL-94 standard is often required.

[0004]

As a method for imparting flame retardancy to a polyester resin, a method in which a halogen-based organic compound as a flame retardant and further an antimony compound as a flame-retardant synergist are compounded to a resin is common. However, this method tends to cause the generation of a large amount of smoke during burning. Further, with an increase in environmental awareness, there are growing concerns about the effects of halogen-based flame-retardant materials on the environment. Accordingly, in recent years, there has been a strong demand for the use of a flame retardant that does not contain halogen at all.

[0005]

In the past, as a method for flame-retarding a thermoplastic resin without using a halogen-based flame retardant, the addition of a hydrated metal compound such as aluminum hydroxide or magnesium hydroxide has been widely known. However, there has been a problem in that a large amount of the hydrated metal compound has to be added in order to acquire sufficient flame retardancy, whereby the original characteristics of the resin are lost.

[0006]

Meanwhile, as a method for flame-retarding a thermoplastic resin without using such a hydrated metal compound, Patent Documents 1 to 8 and the like disclose the

addition of red phosphorus. These are useful flame-retardant resin materials without using halogen-based flame retardant. However, there has been a problem in that specific coloring is caused, and, due to the color tone of the product, the application thereof is limited.

[0007]

Further, Patent Documents 9 to 13 disclose the incorporation of a styrene-based resin into a composition containing a resin, such as PBT or polyphenylene ether, and a phosphate ester. These are useful flame-retardant resin materials without using halogen-based flame retardant. However, the incorporation of a resin such as polyphenylene ether has caused problems, including reduction of mechanical strength, reduction of fluidity during injection molding, yellow coloring of molded articles, limited application due to inferior hydrolysis resistance and metal contamination resistance, etc. Further, Patent Document 14 discloses a composition comprising an organic phosphorus-based flame retardant that contains a polyester and a phosphate ester, glass fibers, and a vinyl-based resin. These are useful flame-retardant resin materials without using halogen-based flame retardant. However, there have been problems of reduction of hydrolysis resistance and metal contamination properties caused by the organic phosphorus-based flame retardant.

[0008]

Further, Patent Document 15 discloses the incorporation of a flame retardant made of a phosphorus-containing compound containing a phosphate ester, a specific aromatic resin, aromatic nylon, polycarbonate resin, polyarylate resin, polyepoxy resin, and polyphenylene ether resin into a resin component containing a polyester-based resin and a styrene-based resin. The resin component mentioned above containing a polyester-based resin and a styrene-based resin is a resin component useful in the improvement of moldability, such as warpage, and the flame retardant mentioned above is a useful flame retardant without using halogen-based flame retardant. However, there have been problems of reduction of hydrolysis resistance and metal contamination properties caused by the phosphorus-containing compound.

[0009]

Further, Patent Documents 16 to 21 disclose the incorporation of an aromatic phosphate ester and melamine cyanurate. These are useful flame-retardant resin materials without using halogen-based flame retardant, but there has been a problem in that it is difficult to balance of mechanical properties and flame retardancy.

PRIOR ART DOCUMENTS

PATENT DOCUMENTS

[0010]

[Patent Document 1] JP-A-51-150553

[Patent Document 2] JP-A-58-108248

[Patent Document 3] JP-A-59-81351

[Patent Document 4] JP-A-5-78560

[Patent Document 5] JP-A-5-287119

[Patent Document 6] JP-A-5-295164

[Patent Document 7] JP-A-5-320486

[Patent Document 8] JP-A-5-339417

[Patent Document 9] JP-A-10-77396

[Patent Document 10] JP-A-10-147699

[Patent Document 11] JP-A-10-182955

[Patent Document 12] JP-A-10-182956

[Patent Document 13] JP-A-2000-26710

[Patent Document 14] JP-A-2000-212412

[Patent Document 15] JP-A-2001-49096

[Patent Document 16] JP-A-3-281652

[Patent Document 17] JP-A-5-70671

[Patent Document 18] JP-A-7-233311

[Patent Document 19] JP-A-8-73713

[Patent Document 20] JP-A-10-120881

[Patent Document 21] WO2004/029154

SUMMARY OF THE INVENTION

PROBLEMS TO BE SOLVED BY THE INVENTION

[0011]

An object of the present invention is to provide a flame-retardant polyester resin composition which is excellent in the balance of flame retardancy and mechanical properties without using a halogen-based flame retardant, and also a molded article.

MEANS FOR SOLVING THE PROBLEMS

[0012]

That is, the present invention is a flame-retardant polyester resin composition comprising (A) a polyester resin and (B) a salt of a triazine-based compound with cyanuric acid or isocyanuric acid. The composition includes the component (B) in an amount of 1 to 70 parts by weight based on 100 parts by weight of the component (A), and, in the composition, the number of particles of the component (B) with a maximum size of more than 15 μm is 1 or less per 100 μm x 100 μm cross section. The present invention is also a molded article made of the flame-retardant polyester resin composition.

ADVANTAGES OF THE INVENTION

[0013]

The molded article made of the flame-retardant polyester resin composition of the present invention uses a non-halogen-based flame retardant that is believed to have less

impact on the environment, and has safety against electrical fire in the equipment and also has high flame retardancy to fire on the molded article itself. The molded article is thus useful for electrical/electronic parts, machine mechanism parts, and automobile parts. Specifically, breakers, electromagnetic switches, focus cases, flyback transformers, molded articles for fusers for copiers and printers, general household electrical appliances, housings for office-automation equipments, coil bobbins, connectors, relays, disk drive chassis, transformers, switch parts, receptacle parts, motor parts, sockets, plugs, condensers, various cases, resistors, electrical/electronic parts into which metal terminals or conducting wires are incorporated, computer-associated parts, audio parts, laser discs and like sound parts, illumination parts, parts associated with telegraph/telephone devices, air conditioner parts, parts for household electrical appliances such as VTRs and televisions, parts for copiers, parts for fax machines, parts for optical devices, automobile ignition parts, connectors for automobiles, various electrical equipment parts for automobiles, and the like can be mentioned.

[0014]

In the present invention, a highly reliable molded article having excellent performance regarding, particularly, flame retardancy, hydrolysis resistance, and metal contamination can be obtained. Accordingly, it is especially

useful for electrical/electronic parts, such as relays, breakers, electromagnetic switches, focus cases, flyback transformers, and molded articles for fusers for copiers or printers.

MODE FOR CARRYING OUT THE INVENTION

[0015]

In the resin composition of the present invention, the polyester resin to serve as the component (A) is a polymer or copolymer having, as a main structural unit, one or more kinds of structural units selected from dicarboxylic acids or ester-forming derivatives thereof, diols or ester-forming derivatives thereof, hydroxycarboxylic acids or ester-forming derivatives thereof, and lactone. The polyester resin may also be a commercial product. Among them, aromatic polyester resins are preferable, such as polyethylene terephthalate (PET), polypropylene terephthalate, polybutylene terephthalate (PBT), polycyclohexane dimethylene terephthalate, polyethylene naphthalate, polypropylene naphthalate, polybutylene naphthalate, polyethylene isophthalate/terephthalate, polypropylene isophthalate/terephthalate, polybutylene isophthalate/terephthalate, polyethylene terephthalate/naphthalate, polypropylene terephthalate/naphthalate, and polybutylene terephthalate/naphthalate. Further, in terms of the balance

of mechanical properties, flame retardancy, and cost, a polyester resin selected from polyethylene terephthalate, polypropylene terephthalate, polybutylene terephthalate, and polycyclohexane dimethylene terephthalate is preferable, and a polyester resin selected from polyethylene terephthalate and polybutylene terephthalate is particularly preferable. It is also preferable to use two or more kinds of these aromatic polyester resins mixed in arbitrary amounts.

[0016]

Further, for the excellent balance of the moldability, flame retardancy, and mechanical properties of the resin composition, it is preferable to use PET and PBT together. In particular, in terms of the balance of moldability and flame retardancy, it is preferable that PET/PBT = 0/100 to 80/20% by weight, and more preferably 0/100 to 60/40% by weight.

[0017]

In the resin composition of the present invention, the salt of a triazine-based compound and cyanuric acid or isocyanuric acid to serve as the component (B) is preferably an adduct of cyanuric acid or isocyanuric acid and a triazine-based compound, and it is usually an adduct having composition of 1:1 (molar ratio) or, in some cases, 1:2 (molar ratio). In the triazine-based compounds, those that do not form a salt with cyanuric acid or isocyanuric acid are excluded. Further, In the salts of a triazine-based compound and cyanuric

acid or isocyanuric acid, in particular, salts of melamine, benzoguanamine, acetoguanamine, 2-amido-4,6-diamino-1,3,5-triazine, mono(hydroxymethyl)melamine, di(hydroxymethyl)melamine, and tri(hydroxymethyl)melamine are preferable, and salts of melamine, benzoguanamine, and acetoguanamine are particularly preferable. These salts are produced by a known method. For example, a mixture of a triazine-based compound and cyanuric acid or isocyanuric acid is formed into an aqueous slurry and thoroughly mixed to produce a salt thereof in particulate form, and then the slurry is filtered and dried, whereby a salt in powder form can be generally obtained. Further, the above salt does not have to be completely pure, and there may be some unreacted triazine-based compound, cyanuric acid, or isocyanuric acid remaining. Further, in terms of the flame retardancy, mechanical strength, wet heat resistance, retention stability, and surface properties of the molded article, the average particle size of the salt before incorporating into a resin is preferably 0.01 to 100 μm, and more preferably 1 to 80 μm. Further, in the case where the dispersibility of the above salt is poor, a dispersant such as tris(β-hydroxyethyl)isocyanurate, a known surface treating agent, or the like may also be used. Further, in terms of flame retardancy and mechanical properties, the amount of the salt of a triazine-based compound and cyanuric acid or isocyanuric

acid incorporated is preferably 1 to 70 parts by weight, more preferably 35 to 70 parts by weight, based on 100 parts by weight of the polyester resin. When the salt content is more than 70 parts by weight, the flame retardancy and mechanical properties of the resin composition are insufficient. Such a content is thus undesirable.

[0018]

In the resin composition of the present invention, it is necessary that the number of particles of the salt of a triazine-based compound and cyanuric acid or isocyanuric acid with a maximum size of more than 15 μm is 1 or less on average per 100 μm x 100 μm cross section. The smaller the number of particles of the salts, the more excellent the mechanical properties and flame retardancy of the resin composition and a molded article thereof; therefore, the number is preferably 0.5 or less on average, and still more preferably 0.2 or less on average. Average herein means the average of the results of cross-sectional observation of at least 5 points per kind of resin composition. The maximum size of the salt means the greatest length of a particle of the salt observed in a cross-sectional photograph. For example, when the particle is elliptical, it means the major axis, when the particle is quadrangle, it means the long diagonal line. The cross section is observed under an optical microscope or transmission electron microscope. The optical microscope or transmission

electron microscope apparatus used for observation is not particularly limited, and may be an apparatus usually used. The pretreatment on a sample for the cross-sectional photograph is not particularly limited, and staining or the like may be performed for facilitating the observation. Optical microscope or transmission electron microscope observation requires the preparation of an ultrathin section. However, depending on the conditions of the sample, embedding in a resin is also possible. With respect to observation magnification of the optical microscope or transmission electron microscope, when the magnification is too small, it is difficult to observe particles, when it is too large, the observation is localized. Therefore, observation is performed at a magnification of 200 to 1,000 times. Further, in order to increase the accuracy of observation results, it is also preferable to combine the observation results at low magnification and at high magnification from the same observation site.

[0019]

In the X-ray diffraction spectrum of the salt of a triazine-based compound and cyanuric acid or isocyanuric acid, a high-intensity diffraction line exists at $2\theta = 11.0 \pm 0.5°$. The present inventors have found that this diffraction line greatly affects the dispersion of the salt of a triazine-based compound and cyanuric acid or isocyanuric acid in a resin. The full width at half maximum of the peak is preferably 0.5° or

less, more preferably 0.4° or less, and still more preferably 0.35° or less. When the full width at half maximum of the peak of the diffraction line is small, this indicates that the crystal structure of the salt is uniform. The present inventors have found that although the reason for this is unclear, when the full width at half maximum of the peak is 0.4° or less, the salt is particularly unlikely to aggregate and is easily dispersible in a resin. A full width at half maximum of more than 0.4° causes uneven distribution of the salt in a resin due to aggregation, resulting in insufficient mechanical properties, and thus is undesirable.

[0020]

Further, from the salt of a triazine-based compound and cyanuric acid or isocyanuric acid, low-molecular-weight components and impurities are extracted by an organic solvent (methanol, benzene, chloroform, etc.). In order to suppress uneven distribution of the salt in a resin due to aggregation, it is preferable that the amount of such extract is small. The amount of the extract which is extracted from the component (B) by a 80% by volume methanol/20% by volume benzene mixed solvent is preferably 0.1% by weight or less, more preferably 0.06% by weight or less, and still more preferably 0.05% by weight or less. When the amount extracted is more than 0.1% by weight, uneven distribution of the salt in a resin is caused by aggregation, resulting in insufficient mechanical properties.

Such an amount is thus undesirable.

[0021]

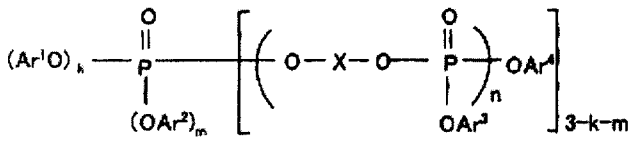
In the resin composition of the present invention, one or more kinds of non-halogen flame retardants other than the component (B) can be further added as a component (C), and this may be expected to shorten the burning time during burning or reduce gas generation during burning. Examples of such other non-halogen flame retardants include, but are not limited to, phosphate esters, phosphinate salts, aluminum hydroxide, hydrotalcite, boric acid, calcium borate, calcium borate hydrate, zinc borate, zinc borate hydrate, zinc hydroxide, zinc hydroxide hydrate, zinc tin hydroxide, zinc tin hydroxide hydrate, red phosphorus, heat-expandable graphite, and dawsonite. Further, a thermosetting resin, such as a thermosetting melamine resin, a thermosetting phenolic resin, or a thermosetting epoxy resin, may also be mixed or applied to cover the surface. Further, a coupling agent, an epoxy compound, or fats and oils such as stearic acid may also be mixed or applied to cover the surface. Among them, a compound selected from phosphate esters and phosphinate salts is favorable in the balance of mechanical properties and flame retardancy, and thus is preferably used. Further, it is preferable to use a phosphate ester and a phosphinate salt together, because this further enhances the effectiveness. Considering flame retardancy, hydrolysis resistance, and metal

contamination properties, the content of the component (C) is preferably 1 to 100 parts by weight, more preferably 10 to 40 parts by weight, based on 100 parts by weight of the polyester. A content of more than 100 parts by weight causes mold deposit, and thus is undesirable.

[0022]

The phosphate ester is not particularly limited, and generally commercially available phosphate esters and synthesized phosphate esters are usable. Specific examples thereof include tricresyl phosphate, trixylenyl phosphate, cresyl diphenyl phosphate, triphenyl phosphate, tris-isopropylbiphenyl phosphate, trimethyl phosphate, triethyl phosphate, tributyl phosphate, trioctyl phosphate, tributoxyethyl phosphate, octyl diphenyl phosphate, orthophenylphenol-based phosphate esters, pentaerythritol-based phosphate esters, neopentyl-glycol-based phosphate esters, substituted neopentyl-glycol phosphonates, nitrogen-containing phosphate esters, and aromatic phosphate esters of the following formula (1). Among them, aromatic phosphate esters are preferable, and an aromatic phosphate ester of the following formula (1) is particularly preferable:

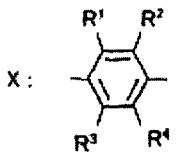
[Chemical Formula 1]



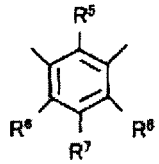
(1).

In the above formula, Ar^1, Ar^2, Ar^3, and Ar^4 represent the same or different non-halogen-containing aromatic groups. Further, X represents a structure selected from the formulae (2) to (4) below. In the formulae (2) to (4), R^1 to R^8 represent the same or different hydrogen atoms or C_{1-5} alkyl groups, Y represents a direct bond, or a group selected from O, S, SO_2, $C(CH_3)_2$, CH_2, and CHPh, and Ph represents a phenyl group. Further, n in the formula (1) is an integer of 0 or more. Further, k and m in the formula (1) are each an integer of 0 or more and 2 or less, and (k + m) is an integer of 0 or more and 2 or less. Besides, the aromatic phosphate ester may be a mixture of aromatic phosphate esters having different ns or different structures.

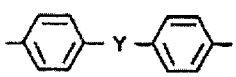
[Chemical Formula 2]



(2)



(3)



(4)

[0023]

More specifically, n in the above formula (1) is an integer of 0 or more, and the upper limit is preferably 40 or less in terms of flame retardancy. n is preferably 0 to 10, and particularly preferably 0 to 5.

[0024]

Further, k and m are each an integer of 0 or more and 2 or less, and k + m is an integer of 0 or more and 2 or less. It is preferable that k and m are each an integer of 0 or more and 1 or less, and it is particularly preferable that k and m are each 1.

[0025]

Further, in the above formulae (2) to (4), R^1 to R^8 represent the same or different hydrogen or C_{1-5} alkyl groups. Specific examples of the C_{1-5} alkyl groups herein include a methyl group, an ethyl group, a n-propyl group, an isopropyl group, a n-butyl group, a sec-butyl group, a tert-butyl group, n-isopropyl, neopentyl, a tert-pentyl group, 2-isopropyl, neopentyl, a tert-pentyl group, 3-isopropyl, neopentyl, a tert-pentyl group, neoisopropyl, neopentyl, and a tert-pentyl group. Among them, a group selected from hydrogen, a methyl group, and an ethyl group is preferable, and hydrogen is particularly preferable.

[0026]

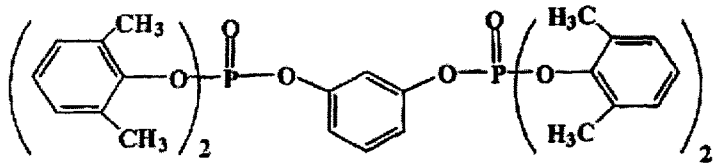
Further, Ar^1, Ar^2, Ar^3, and Ar^4 represent the same or

different non-halogen-containing aromatic groups. Examples of such aromatic groups include aromatic groups having a skeleton selected from a benzene skeleton, a naphthalene skeleton, an indene skeleton, and an anthracene skeleton. Among them, those having a benzene skeleton or a naphthalene skeleton are preferable. They may be substituted with a non-halogen-containing organic residue, and preferably with a C_{1-8} organic residue. When they have a substituent, the number of substituents is not limited either, but is preferably 1 to 3. Specific examples of Ar^1, Ar^2, Ar^3, and Ar^4 include aromatic groups such as a phenyl group, a tolyl group, a xylyl group, a cumenyl group, a mesityl group, a naphthyl group, an indenyl group, and an anthryl group. Among them, a group selected from a phenyl group, a tolyl group, a xylyl group, a cumenyl group, and a naphthyl group is preferable, and a group selected from a phenyl group, a tolyl group, and a xylyl group is particularly preferable.

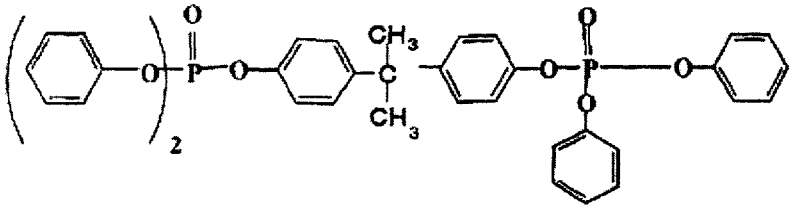
[0027]

As a specific example of an aromatic phosphate ester, the following compound (6) or (7) is preferable, and (6) is particularly preferable.

[Chemical Formula 3]



(6)



(7)

[0028]

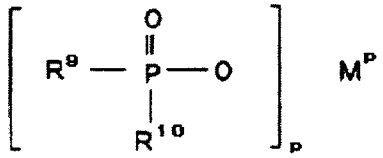
As commercially available phosphate esters, one kind or two or more kinds selected from PX-200, PX-201, PX-130, CR-733S, TPP, CR-741, CR747, TCP, TXP, and CDP manufactured by Daihachi Chemical Industry Co., Ltd. are usable. It is preferable to use one kind or two or more kinds selected from PX-200, TPP, CR-733S, CR-741, and CR747, and it is particularly preferable to use one selected from PX-200, CR-733S, and CR-741. Among them, PX-200 is particularly effective in improving hydrolysis resistance and contact contamination resistance when used together with a vinyl-based resin and an alkaline earth metal compound, and thus is most preferable.

[0029]

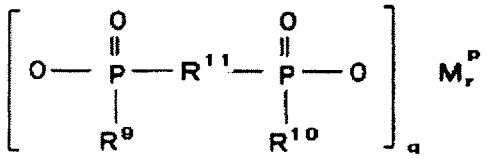
As the phosphinate salt in the present invention, a flame retardant including at least one kind selected from phosphinate salts, diphosphinate salts, and polymers thereof is preferable. The phosphinate salt is preferably a salt of a metal selected from calcium, aluminum, and zinc. A flame retardant including

one or more kinds of phosphinate salts of the following formula (8), diphosphinate salts of the following formula (9), and polymers thereof is particularly preferable:

[Chemical Formula 4]



(8)



(9).

R^9 and R^{10} in the formula may be the same or different and are C_{1-6} alkyl groups or aryl groups. R^9 and R^{10} may be straight or branched, examples thereof including a methyl group, an ethyl group, a propyl group, an isopropyl group, a n-butyl group, a tert-butyl group, a pentyl group, and a phenyl group. Further, R^{11} is a straight or branched C_{1-10} alkylene group or C_{6-10} arylene group, alkylarylene group, or arylalkylene group. Examples thereof include a methylene group, an ethylene group, a propylene group, an isopropylene group, a n-butylene group, a tert-butylene group, a pentylene group, an octylene group, a dodecylene group, a phenyl group, a naphthylene group, a methylphenylene group, an ethylphenylene group, a methylnaphthylene group, an ethylnaphthylene group, a tert-butylphenylene group, and a tert-butylnaphthylene group.

M is calcium, aluminum, or zinc. M is preferably aluminum. Further, p is 1 to 4, q is 1 to 4, and r is 1 to 4.

[0030]

Examples of commercial products include "Exolit" (registered trademark) OP1230 and OP1240 manufactured by Clariant Japan. Further, mixtures of a flame retardant including at least one kind selected from phosphinate salts, diphosphinate salts, and polymers thereof with a nitrogen-containing compound and/or a boron-containing compound are also commercially available from Clariant Japan. As an example of the commercial product, OP1312 can be mentioned. OP1240 is preferably used.

[0031]

An epoxy compound may be further incorporated into the polyester resin composition of the present invention. Examples of such epoxy compounds include glycidyl ester compounds, glycidyl ether compounds, and glycidyl ester ether compounds. One or more kinds thereof can be used. In order to exert excellent improving effects on hydrolysis resistance, an epoxy compound having an epoxy equivalent of less than 500 is preferable. Further, an epoxy compound having an epoxy equivalent of less than 400 is particularly preferable. The epoxy equivalent herein means the number of grams of an epoxy compound containing one gram equivalent of an epoxy group. The epoxy equivalent can be determined by a method in which an epoxy

compound is dissolved in pyridine, then 0.05 N hydrochloric acid is added thereto, followed by heating at 45°C, and back titration is performed with 0.05 N caustic soda using a mixture of thymol blue and cresol red as an indicator. Further, as the above epoxy compound, an epoxy compound using a monofunctional glycidyl ester compound together with a glycidyl ether compound, or alternatively a monofunctional glycidyl ester compound, is preferably used. In particular, in terms of the balance of the viscosity stability and hydrolysis resistance of the resulting composition, a monofunctional glycidyl ester compound is more preferable. Specific examples of the glycidyl ester compounds include, but are not limited to, glycidyl benzoate, glycidyl t-butylbenzoate, glycidyl p-toluate, glycidyl cyclohexanecarboxylate, glycidyl pelargonate, glycidyl stearate, glycidyl laurate, glycidyl palmitate, glycidyl behenate, glycidyl versatate, glycidyl oleate, glycidyl linoleate, glycidyl linolenate, glycidyl behenolate, glycidyl stearolate, diglycidyl terephthalate, diglycidyl isophthalate, diglycidyl phthalate, diglycidyl naphthalenedicarboxylate, diglycidyl bibenzoate, diglycidyl methyl terephthalate, diglycidyl hexahydrophthalate, diglycidyl tetrahydrophtalate, diglycidyl cyclohexanedicarboxylate, diglycidyl adipate, diglycidyl succinate, diglycidyl sebacate, diglycidyl dodecanedioate, diglycidyl octadecanedicarboxylate, triglycidyl trimellitate, and tetraglycidyl pyromellitate.

One kind or two or more kinds thereof can be used. Further, specific examples of the glycidyl ether compounds include, but are not limited to, diglycidyl ethers obtained by allowing a bisphenol to react with epichlorohydrin, such as phenyl glycidyl ether, o-phenyl phenyl glycidyl ether, 1,4-bis(β,γ-epoxypropoxy)butane, 1,6-bis(β,γ-epoxy propoxy)hexane, 1,4-bis(β,γ-epoxypropoxy)benzene, 1-(β,γ-epoxypropoxy)-2-ethoxyethane, 1-(β,γ-epoxypropoxy)-2-benzyloxyethane, 2,2-bis-[p-(β,γ-epoxypropoxy)phenyl]propane, and bis-(4-hydroxyphenyl)methane, and one kind or two or more kinds thereof can be used. Further, in terms of mechanical properties and hydrolysis resistance, the amount of the epoxy compound incorporated is preferably 0.1 to 5 parts by weight, particularly preferably 0.3 to 4 parts by weight, based on 100 parts by weight of the polyester.

[0032]

In order to improve mechanical strength, a fiber-reinforced material may be incorporated into the polyester resin composition of the present invention. Examples of such fiber-reinforced materials include glass fibers, aramid fibers, and carbon fibers. As the glass fibers, it is preferable to use chopped-strand-type or roving-type glass fibers used as an ordinary reinforcement material for polyester resins, which are glass fibers treated with a silane

coupling agent, such as an amino silane compound or an epoxy silane compound, and/or a sizing agent including urethane, vinyl acetate, bisphenol A diglycidyl ether, one or more kinds of epoxy compounds such as novolac-type epoxy compounds, etc. Further, the silane coupling agent and/or sizing agent may be used in an emulsion liquid. Further, when the fiber-reinforced material is incorporated, in terms of the fluidity of the polyester resin composition during molding, the durability of the molding machine and die, and the mechanical properties of the composition, the amount incorporated is preferably 0 to 100 parts by weight, more preferably 25 to 90 parts by weight, based on 100 parts by weight of the polyester.

[0033]

In order to improve some of crystallization characteristics, arc resistance, anisotropy, mechanical strength, flame retardancy, heat distortion temperature, and the like, inorganic fillers other than the fiber-reinforced material may be further incorporated into the polyester resin composition of the present invention. Examples of such inorganic fillers other than the fiber-reinforced material include, but are not limited to, acicular, granular, powdery, and laminated inorganic fillers. Specific examples thereof include glass beads, milled fibers, glass flakes, potassium titanate whisker, calcium sulfate whisker, wollastonite, silica, kaolin, talc, smectite-based clay minerals

(montmorillonite, hectorite), vermiculite, mica, fluor-taeniolite, zirconium phosphate, titanium phosphate, and dolomite, and one or more kinds are used. Further, the inorganic fillers other than the fiber-reinforced material may have been subjected to a surface treatment, such as coupling agent treatment, an epoxy compound, or ionization treatment. Further, in terms of impact strength, the granular, powdery, and laminated inorganic fillers preferably have an average particle size of 0.1 to 20 μm, and particularly preferably 0.2 to 10 μm. Further, in terms of the fluidity of the composition during molding and the durability of a molding machine or die, the amount of the inorganic filler incorporated other than the fiber-reinforced material is, as a total with the amount of the fiber-reinforced agent incorporated, preferably 0 to 100 parts by weight based on 100 parts by weight of the polyester.

[0034]

A fluorine-based resin can be further incorporated into the polyester resin composition of the present invention to suppress the meltdown of the flame-retardant resin composition during burning and also to improve flame retardancy. The fluorine-based resin is a resin containing fluorine in the molecule. Specific examples thereof include polytetrafluoroethylene, polyhexafluoropropylene, a (tetrafluoroethylene/hexafluoropropylene)copolymer, a (tetrafluoroethylene/perfluoroalkyl vinyl ether)copolymer, a

(tetrafluoroethylene/ethylene)copolymer, a (hexafluoropropylene/propylene)copolymer, polyvinylidene fluoride, and a (vinylidene fluoride/ethylene)copolymer. Among them, polytetrafluoroethylene, a (tetrafluoroethylene/perfluoroalkyl vinyl ether)copolymer, a (tetrafluoroethylene/hexafluoropropylene)copolymer, a (tetrafluoroethylene/ethylene)copolymer, and polyvinylidene fluoride are preferable, and polytetrafluoroethylene and a (tetrafluoroethylene/ethylene)copolymer are particularly preferable. Further, when the fluorine-based resin is incorporated, in terms of flame retardancy and mechanical properties, the amount incorporated is preferably 0.02 to 5 parts by weight, more preferably 0.2 to 2 parts by weight, based on 100 parts by weight of the polyester.

[0035]

A polycarbonate resin can be further incorporated into the polyester resin composition of the present invention to further improve flame retardancy. Examples of the polycarbonate resins include aromatic homo- or copolycarbonates obtained by allowing an aromatic dihydric phenol-based compound to react with phosgene or a carbonic acid diester. Such an aromatic homo- or copolycarbonate resin preferably has a weight-average molecular weight within a range of 10,000 to 1,100,000, and more preferably has a glass-transition temperature of about 150°C and a

weight-average molecular weight within a range of 10,000 to 1,000,000. It is also possible to use polycarbonate resins with different weight-average molecular weights together. A polycarbonate resin having a weight-average molecular weight within a range 60,000 to 1,100,000 is particularly preferable. Weight-average molecular weight is obtained from measurement in terms of polystyrene by gel permeation chromatography using tetrahydrofuran as the solvent. A weight-average molecular weight of less than 10,000 causes a decrease in the mechanical properties of the polyester resin composition, and thus is undesirable. A weight-average molecular weight of more than 110,000 causes a loss of the fluidity of the composition during molding, and thus is undesirable. Further, a polycarbonate resin having a melt viscosity index (melt flow index) within a range of 1 to 100 g/10 minutes as measured by a melt-indexer under conditions where the temperature is 300°C and the load is 1.2 kg according to ASTM D1238 is preferable, and particularly preferably 1 to 15 g /10 minutes in terms of mechanical properties.

[0036]

Further, examples of the aromatic dihydric phenol-based compounds include 2,2-bis(4-hydroxyphenyl)propane, 2,2-bis(4-hydroxy-3,5-dimethylphenyl)propane, bis(4-hydroxyphenyl)methane, 1,1-bis(4-hydroxyphenyl)ethane, 2,2-bis(4-hydroxyphenyl)butane,

2,2-bis(4-hydroxy-3,5-diphenyl)butane,

2,2-bis(4-hydroxy-3,5-diethylphenyl)propane,

2,2-bis(4-hydroxy-3,5-diethylphenyl)propane,

1,1-bis(4-hydroxyphenyl)cyclohexane, and

1-phenyl-1,1-bis(4-hydroxyphenyl)ethane, and they may be used singly or as a mixture. When the polycarbonate resin is incorporated in an amount of more than 20 parts by weight based on 100 parts by weight of the polyester, hydrolysis resistance may be greatly reduced, and therefore caution is necessary. In terms of hydrolysis resistance and flame retardancy, the amount of the polycarbonate resin incorporated is preferably 0.1 to 10 parts by weight based on 100 parts by weight of the polyester. Further, it is also possible to incorporate a polycarbonate resin oligomer in an amount within a range where the characteristics of the present invention are not impaired.

[0037]

Further, when the polycarbonate resin is incorporated, the incorporation of a small amount of an acid phosphate ester is useful as an ester interchange inhibitor for the polyester and the polycarbonate resin. In particular, a decrease in the heat distortion temperature, for example, is inhibited. The acid phosphate ester is preferably an alkyl acid phosphate compound of a partial ester of an alcohol and phosphoric acid. A low-molecular-weight one is a colorless liquid, while a high-molecular-weight one is a solid in the form of white wax

or flakes.

[0038]

Specific examples of the acid phosphate esters include, but are not limited to, monomethyl acid phosphate, monoethyl acid phosphate, monoisopropyl acid phosphate, monobutyl acid phosphate, monolauryl acid phosphate, monostearyl acid phosphate, monododecyl acid phosphate, monobehenyl acid phosphate, dimethyl acid phosphate, diethyl acid phosphate, diisopropyl acid phosphate, dibutyl acid phosphate, lauryl acid phosphate, distearyl acid phosphate, didodecyl acid phosphate, dibehenyl acid phosphate, trimethyl acid phosphate, triethyl acid phosphate, a mixture of the above mono- and di- acid phosphate esters, a mixture of mono-, di-, and tri- acid phosphate esters, and a mixture of one or more kinds of the above compounds. Preferred examples of the acid phosphate esters include long-chain alkyl acid phosphate compounds, such as a mixture of mono- and distearyl acid phosphates. Such a compound is commercially available from Asahi Denka Kogyo Co., Ltd. under the name of "ADEKASTAB (registered trademark)" AX-71, and is a flaky solid having a melting point. Further, in terms of heat distortion temperature and mechanical properties, the amount of the acid phosphate ester incorporated is preferably 0.01 to 0.5 parts by weight, more preferably 0.02 to 0.3 parts by weight, based on 100 parts by weight of the polyester.

[0039]

A flame-retardant synergist that supports flame retardancy, such as a silicone compound, a phenol resin, a phosphonitrile compound, ammonium polyphosphate, or melamine phosphate, can be further incorporated into the polyester resin composition of the present invention, and one or more kinds are used. Further, when such a flame-retardant synergist is incorporated, in terms of flame retardancy and mechanical properties, the amount incorporated is preferably 0.1 to 10 parts by weight, more preferably 0.2 to 7.5 parts by weight, based on 100 parts by weight of the polyester.

[0040]

Examples of the silicone compounds include silicone resins, silicone oils, and silicone powders. Further, examples of the silicone resins include polyorganosiloxanes having a group and siloxane chemically bonded together, the group being selected from a saturated or unsaturated monovalent hydrocarbon group, a hydrogen atom, a hydroxyl group, an alkoxyl group, an aryl group, a vinyl group, or an allyl group. One having a viscosity of about 200 to 300,000,000 cP at room temperature is preferable, but the silicone resin is not limited thereto, and, with respect to the product form, it may be in the form of oil, powder, or gum. It may have, as functional groups, an epoxy group, a methacrylic group, and an amino group introduced, and may also be a mixture of two or more kinds of the silicone resins. Further, examples of the silicone oils

include polyorganosiloxanes having a group and siloxane chemically bonded together, the group being selected from a saturated or unsaturated monovalent hydrocarbon group, a hydrogen atom, a hydroxyl group, an alkoxyl group, an aryl group, a vinyl group, or an allyl group. One having a viscosity of about 0.65 to 100,000 cSt at room temperature is preferable, but the silicone oil is not limited thereto, and, with respect to the product form, it may be in the form of oil, powder, or gum. It may have, as functional groups, an epoxy group, a methacrylic group, and an amino group introduced. It may also be a mixture of two or more kinds of the silicone oils or with the silicone resin. Further, examples of the silicone powders include those obtained by incorporating an inorganic filler into the silicone resin and/or silicone oil mentioned above. As the inorganic filler, silica or the like is preferably used.

[0041]

Further, the phenol resin may be any phenol resin as long as it is a polymer having a plurality of phenolic hydroxy groups. Examples thereof include novolac-type, resol-type, and heat-reactive resins, as well as resins obtained by modification thereof. They may also be uncured resins having no curing agent added, semi-cured resins, or cured resins. Among them, in terms of flame retardancy, mechanical properties, and economical efficiency, a novolac-type phenol resin that has no curing agent added and is not heat-reactive or a

melamine-modified, novolac-type phenol resin is preferable. Further, the form is not particularly limited, any of those in pulverized form, granular form, flaky form, powdery form, acicular form, liquid form, and the like are usable, and one kind or two or more kinds thereof can be used as required. Further, commercially available phenolic resins are also usable. For example, in the case of the novolac-type phenol resin, a reaction vessel is prepared in such a manner that the molar ratio between phenols and aldehydes is 1:0.7 to 1:0.9, then a catalyst such as oxalic acid, hydrochloric acid, sulfuric acid, or toluenesulfonic acid is further added thereto followed by heating, and a reflux reaction is performed for a predetermined period of time. Vacuum dehydration or standing dehydration is then performed to remove produced water, and still remaining water and unreacted phenols are removed. By such a method, a novolac-type phenol resin can be obtained. Co-condensed phenol resins obtained by using these resins or a plurality of raw material components can be used alone, or it is also possible to use two or more kinds thereof. Further, in the case of the resol-type phenol resin, a reaction vessel is prepared in such a manner that the molar ratio between phenols and aldehydes is 1:1 to 1:2, and a catalyst such as sodium hydroxide, aqueous ammonia, or other basic substances is added thereto, followed by the same reaction and treatment as in the case of the novolac-type phenol resin; a resol-type phenol resin can thus

be obtained. Here, examples of the phenols include phenol, o-cresol, m-cresol, p-cresol, thymol, p-tert-butylphenol, tert-butylcatechol, catechol, isoeugenol, o-methoxyphenol, 4,4'-dihydroxyphenyl-2,2-propane, isoamyl salicylate, benzyl salicylate, methyl salicylate, and 2,6-di-tert-butyl-p-cresol. One kind or two or more kinds of these phenols can be used. Meanwhile, examples of the aldehydes include formaldehyde, paraformaldehyde, polyoxymethylene, and trioxane. One kind or two or more kinds of these aldehydes can be used as required. The molecular weight of the phenolic resin is not particularly limited, but is preferably 200 to 2,000 in number-average molecular weight. It is particularly preferably within a range of 400 to 1,500, because this leads to excellent mechanical properties, fluidity, and economical efficiency. Besides, the molecular weight of a phenolic resin can be measured, as a tetrahydrofuran solution, using a polystyrene standard sample by gel permeation chromatography.

[0042]

Further, examples of the phosphonitrile compounds include phosphonitrile compounds having as a main component a phosphonitrile linear polymer and/or cyclic polymer, which may be linear, cyclic, or a mixture thereof. The phosphonitrile linear polymer and/or cyclic polymer can be synthesized by a known method described in "*Hosfazen-kagobutsu no Gosei to Oyo* (Synthesis and Application of Phosphazene Compound)" written

by Kajiwara, etc. For example, phosphorus pentachloride or phosphorus trichloride as a phosphorus source is allowed to react with ammonium chloride or ammonia gas as a nitrogen source (it is also possible to purify a cyclic product) by a known method, and the obtained substance is substituted with alcohols, phenols, or amines, whereby synthesis can be achieved.

[0043]

Further, examples of the ammonium polyphosphates include ammonium polyphosphates, melamine-modified ammonium polyphosphates, and carbamyl ammonium polyphosphates. The ammonium polyphosphate may be coated with a thermosetting resin, such as a phenol resin, urethane resin, melamine resin, urea resin, epoxy resin, or urea resin having thermosetting properties, and one kind or two or more kinds may be used. Further, examples of the melamine polyphosphates include melamine polyphosphates such as melamine phosphates and melamine pyrophosphates, and one kind or two or more kinds may be used.

[0044]

In the present invention, an ethylene (co)polymer may be further incorporated for the purpose of improving the toughness of the composition, such as impact strength. Examples of such ethylene (co)polymers include ethylene polymers and ethylene copolymers, such as high-density polyethylene, low-density polyethylene, and ultra-low-density polyethylene. An

ethylene copolymer can be obtained by copolymerizing ethylene and a monomer copolymerizable therewith. Examples of the copolymerizable monomers include propylene, butene-1, vinyl acetate, isoprene, butadiene, monocarboxylic acids such as acrylic acid and methacrylic acid, ester acids thereof, and dicarboxylic acids such as maleic acid, fumaric acid, and itaconic acid. An ethylene copolymer can be produced by a known method. Specific examples of the ethylene copolymers include ethylene/propylene, ethylene/butene-1, ethylene/vinyl acetate, ethylene/ethyl acrylate, ethylene/methyl acrylate, and ethylene/ethyl methacrylate acrylate. It is also preferable to use a copolymer obtained by grafting or copolymerization of an anhydride or glycidyl methacrylate with the ethylene (co)polymer. One kind or two or more kinds thereof are used. In the ethylene (co)polymers, a copolymer obtained by grafting or copolymerization of an anhydride or glycidyl methacrylate with polyethylene has excellent compatibility with a polyester component and thus is preferably used.

[0045]

Further, when the ethylene (co)polymer is incorporated, in terms of the flame retardancy and impact strength of the resulting composition, the amount incorporated is preferably 1 to 10 parts by weight, more preferably 2 to 7.5 parts by weight, based on 100 parts by weight of the polyester.

[0046]

Phenoxy resins, oxazoline compounds, carbodiimide compounds, and the like, which are materials for improving hydrolysis resistance, can be further incorporated into the polyester resin composition of the present invention. In particular, a phenoxy resin is preferably used. When the material for improving hydrolysis resistance is incorporated, in terms of the hydrolysis resistance and flame retardancy of the resulting composition, the amount incorporated is preferably 0.1 to 7.5 parts by weight, more preferably 0.2 to 5 parts by weight, based on 100 parts by weight of the polyester.

[0047]

Examples of the phenoxy resins include phenoxy resins obtained by allowing an aromatic dihydric phenol-based compound to react with epichlorohydrin in various proportions. The molecular weight of the phenoxy resin is not particularly limited, but those having a viscosity-average molecular weight within a range of 1,000 to 100,000 are preferable. Here, as examples of the aromatic dihydric phenol-based compounds, 2,2-bis(4-hydroxyphenyl)propane, 2,2-bis(4-hydroxy-3,5-dimethylphenyl)propane, bis(4-hydroxyphenyl)methane, 1,1-bis(4-hydroxyphenyl)ethane, 2,2-bis(4-hydroxy-3,5-diethylphenyl)propane, 1,1-bis(4-hydroxyphenyl)cyclohexane, 1-phenyl-1,1-bis(4-hydroxyphenyl)ethane, 2,2-bis(4-hydroxyphenyl)methane, and the like are usable.

They may be used alone or as a mixture. Further, the form of the phenoxy resin is not particularly limited, and any of those in pulverized form, granular form, flaky form, powdery form, liquid form, and the like are usable. One kind or two or more kinds of these phenoxy resins can be used as required.

[0048]

In order to achieve extremely excellent heat aging resistance even when the composition of the present invention is exposed to high temperatures for a long period of time, a hindered-phenol-based antioxidant and/or a phosphite-based antioxidant can be further incorporated as a stabilizer into the polyester resin composition of the present invention. When the hindered-phenol-based antioxidant and/or the phosphite-based antioxidant is incorporated, in terms of heat aging resistance and flame retardancy, the amount incorporated is preferably 0.1 to 1 part by weight, more preferably 0.2 to 0.5 parts by weight, based on 100 parts by weight of the polyester. When the amount incorporated is less than 0.1 parts by weight, heat aging resistance is not sufficient, while when it is more than 1 part by weight, mechanical properties and flame retardancy are impaired. Such an amount is thus undesirable.

[0049]

Specific examples of the hindered-phenol-based antioxidants include triethylene glycol-bis[3-(3-t-butyl-5-methyl-4-hydroxyphenyl)propionate

],

1,6-hexanediol-bis[3-(3,5-di-t-butyl-4-hydroxyphenyl)propionate],

pentaerythrityl-tetrakis[3-(3,5-di-t-butyl-4-hydroxyphenyl)propionate],

2,2-thio-diethylenebis[3-(3,5-di-t-butyl-4-hydroxyphenyl)propionate],

octadecyl-3-(3,5-di-t-butyl-4-hydroxyphenyl)propionate,

3,5-di-t-butyl-4-hydroxybenzyl phosphonate diethyl ester,

1,3,5-trimethyl-2,4,6-tris(3,5-di-t-butyl-4-hydroxybenzyl)benzene, bis- or

tris(3-t-butyl-6-methyl-4-hydroxyphenyl)propane,

N,N'-hexamethylenebis(3,5-di-t-butyl-4-hydroxy-hydrocinnamamide), and

N,N'-trimethylenebis(3,5-di-t-butyl-4-hydroxy-hydrocinnamamide).

[0050]

Further, examples of the phosphite-based stabilizers include tris(2,4-di-t-butylphenyl)phosphite,

2,2-methylenebis(4,6-di-t-butylphenyl)octylphosphite,

trisnonylphenyl phosphite, alkyl-allyl-based phosphite,

trialkyl phosphite, triallyl phosphite, and

pentaerythritol-based phosphite compounds.

[0051]

One or more kinds of lubricants can be further added to

the polyester resin composition of the present invention to improve fluidity and mold release properties during molding. Examples of such lubricants include, but are not limited to, metallic soaps such as calcium stearate and barium stearate; fatty acid esters; salts of fatty acid esters (including those partially converted into a salt); fatty acid amides such as ethylene bis stearamide; fatty acid amides made of a polycondensate of ethylenediamine, stearic acid, and sebacic acid or a polycondensate of phenylenediamine, stearic acid, and sebacic acid; polyalkylene wax; anhydride-modified polyalkylene wax; and mixtures of the above lubricants with fluorine-based resins or fluorine-based compounds. When the lubricant is incorporated, the amount added is preferably 0.05 to 2 parts by weight, more preferably 0.1 to 1 part by weight, based on 100 parts by weight of the polyester.

[0052]

Further, one or more kinds of carbon black, titanium oxide, and pigments and dyes of various colors can be incorporated into the polyester resin composition of the present invention to prepare the resin in various colors and also to improve weather (light) resistance and electrical conductivity. When the pigment or the dye is incorporated, in terms of the mechanical properties of the resulting composition, the amount incorporated is preferably 0.1 to 10 parts by weight, more preferably 0.1 to 5 parts by weight, based on 100 parts by weight

of the polyester.

[0053]

Examples of the carbon black include, but are not limited to, channel black, furnace black, acetylene black, anthracene black, soot, turpentine soot, and graphite. Carbon black having an average particle size of 500 nm or less and a dibutyl phthalate oil absorption of 50 to 400 $cm^3/100$ g is preferably used. It may have been treated with, as a treatment agent, aluminum oxide, silicon oxide, zinc oxide, zirconium oxide, polyol, a silane coupling agent, or the like. Further, as the titanium oxide, titanium oxide that is in a crystal form such as rutile form or anatase form and has an average particle size of 5 μm or less is preferably used. It may have been treated with, as a treatment agent, aluminum oxide, silicon oxide, zinc oxide, zirconium oxide, polyol, a silane coupling agent, or the like. Further, for the purpose of improving dispersibility with the flame-retardant resin composition of the present invention or improving handleability during production, the carbon black, titanium oxide, and pigments and dyes of various colors may be melt-blended or simply blended with various thermoplastic resins and used as a mixed material. In particular, as a thermoplastic resin, polyalkylene terephthalate is preferably used.

[0054]

Further, for the flame-retardant polyester resin

composition of the present invention and a molded article, a material including one or more kinds of known additives, such as sulfur-based antioxidants, ultraviolet absorbers, plasticizers, and antistatic agents, can also be used to the extent that they do not interfere with an object of the present invention.

[0055]

Besides, in the present invention, although the addition of a polyphenylene ether resin or polyphenylene sulfide resin further improves flame retardancy, it may reduce tracking resistance, hydrolysis resistance, and metal contamination properties. Therefore, such a resin is not preferably incorporated. When it is incorporated, the amount is preferably not more than 5% by weight.

[0056]

The flame-retardant polyester resin composition of the present invention and a molded article are produced by a known method. For example, a polyester resin, a vinyl-based resin, a phosphate ester, a salt of a triazine-based compound with cyanuric acid or isocyanuric acid, and an alkaline earth metal compound, if necessary as well as an epoxy compound and a fiber-reinforced material such as glass fibers, further if necessary as well as an inorganic filler other than the fiber-reinforced material, a fluorine-based compound, a polycarbonate resin, various flame-retardant synergists, an

ethylene (co)polymer, a material for improving hydrolysis resistance, a hindered-phenol-based antioxidant, and a phosphite-based antioxidant, together with other necessary additives and coloring agents such as pigments and dyes, are fed to a kneading machine after premixing or without premixing, and thoroughly melt-kneaded to thereby prepare a flame-retardant polybutylene terephthalate resin composition. Examples of the premixing include dry blending and mixing using a mechanical mixing apparatus, such as a tumbler, a ribbon mixer, or a Henschel mixer. Further, the method may also be such that the fiber-reinforced material is added from a side feeder installed between a main loading part and a vent part of a multi-screw extruder such as a twin-screw extruder. Further, in the case where the additive is liquid, the method may be such that a liquid addition nozzle is installed between a main loading part and a vent part of a multi-screw extruder such as a twin-screw extruder, and the additive is added using a plunger pump, or that the additive is supplied from the main loading part or the like using a metering pump.

[0057]

Further, the kneading machine may be, but is not limited to, for example, a single-screw extruder, twin-screw extruder, or triple-screw extruder equipped with a "uni-melt" or "dulmadge" type screw or a kneader-type kneading machine.

[0058]

The flame-retardant polyester resin composition thus obtained can be molded by a known method. It can be formed into a molded article of any shape by, for example, injection molding, extrusion molding, compression molding, sheet forming, film forming, etc. In particular, injection molding is preferable, and it is also possible to perform insert molding where a part of a metal component is directly integrated with the molded article.

EXAMPLES

[0059]

Hereinafter, the advantages of the present invention will be described in more detail through examples. As used herein, "%" and "part" all represent "% by weight" and "part by weight", and "/" in reference examples means copolymerization.

[0060]

Reference Example 1: Polybutylene Terephthalate Resin (hereinafter abbreviated as PBT)

1100S (manufactured by Toray Industries Inc.), PBT having an intrinsic viscosity of 0.85 (25°C, orthochlorophenol solvent) was used.

[0061]

Reference Example 2: Polyethylene Terephthalate Resin (hereinafter abbreviated as PET)

Mitsui PET j005 (manufactured by Mitsui PET Resin), PET having an intrinsic viscosity of 0.65 (25°C, 1:1 mixed solvent

of phenol/tetrachloroethane) was used.

[0062]

Reference Example 3: Salt of Triazine-based Compound with Cyanuric Acid or Isocyanuric Acid

<B-1> MCA-12 (manufactured by Sichuan Institute of Fine Chemical Industry Research and Design)

<B-2> MC-15 (manufactured by Sichuan Institute of Fine Chemical Industry Research and Design)

<B-3> MC4000 (manufactured by Nissan Chemical Industries, Ltd.)

<B-4> MC4500 (manufactured by Nissan Chemical Industries, Ltd.)

<B-5> MC6000 (manufactured by Nissan Chemical Industries, Ltd.)

The X-ray diffraction spectra of these salts were measured using an X-ray diffraction apparatus (manufactured by Rigaku Corporation, Multiflex). In the X ray diffraction spectra obtained, among the diffraction lines at $2\theta = 11.0 \pm 0.5°$, the full width at half maximum of the diffraction line with the highest peak intensity was measured. Further, in methanol/benzene extraction, 5 g of a sample was placed in 200 ml of a 80% by volume methanol/20% by volume benzene mixed solvent, and then subjected to extraction by stirring and heating at 70°C for 4 hours, followed by cooling and filtration. The filtrate was dried, and the weight thereof was used for

determination. The full width at half maximum of the maximum peak of the diffraction line at 2θ = 11.0 ± 0.5° in the X-ray diffraction spectrum and the methanol/benzene extraction amount are shown in Table 1.

[0063]

[Table 1]

		Full width at half maximum* (°)	Methanol/Benzene Extraction Amount (%)
B-1	MCA-12	0.31	0.05
B-2	MC-15	0.30	0.05
B-3	MC4000	0.57	0.16
B-4	MC4500	0.44	0.09
B-5	MC6000	0.66	0.07

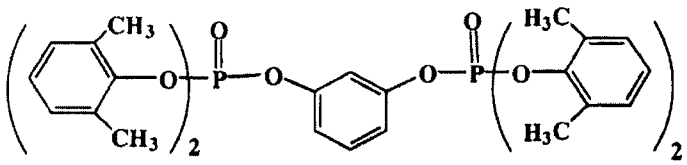
*Maximum peak of diffraction line at 2° = 11.0 ± 0.5°

[0064]

Reference Example 4: Phosphate Ester

An aromatic phosphate ester of the following formula (10) (manufactured by Daihachi Chemical Industry Co., Ltd., PX-200) was used.

[Chemical Formula 5]



(10)

[0065]

Reference Example 5: Phosphinate Salt

As a metal salt of phosphinic acid, "Exolit OP1240" manufactured by Clariant was used.

[0066]

Reference Example 6: A polycarbonate resin (hereinafter abbreviated as PC) "Iupilon" S3000 (manufactured by Mitsubishi Engineering-Plastics Corporation) was used.

[0067]

Reference Example 7: A polyphenylene sulfide "Torelina" M2588 (manufactured by Toray Industries Inc.) (hereinafter abbreviated as PPS) was used.

[0068]

Reference Example 8: Fiber-Reinforced Material

Glass fibers having a fiber diameter of about 13 μm (hereinafter abbreviated as GF) "ECS T-187" (manufactured by Nippon Electric Glass Co., Ltd.) were used.

[0069]

Reference Example 9: Fluorine-Based Compound

Polytetrafluoroethylene "Teflon (registered trademark) 6-J" (manufactured by Dupont-Mitsui Fluorochemicals Co., Ltd.) was used (hereinafter abbreviated as Teflon). To the resin materials of all the examples, 0.2 parts by weight of the polytetrafluoroethylene was added.

[0070]

Reference Example 10: Silicone Compound

A silicone powder "DC4-7105" (manufactured by Dow Corning Toray Silicone Co., Ltd.) was used. To the resin materials of all the examples, 0.2 parts by weight of the silicon powder was

added.

[0071]

Reference Example 11: Alkaline Earth Metal Compound

To the resin materials of all the examples, 0.2 parts by weight of calcium carbonate "KSS1000" (manufactured by Dowa Calfine Co., Ltd.) was added.

[0072]

Reference Example 12: Hindered-Phenol-Based Antioxidant

A hindered-phenol-based antioxidant "IR-1010" (manufactured by Ciba Specialty Chemicals Inc.) of pentaerythrityl-tetrakis[3-(3,5-di-t-butyl-4-hydroxyphenyl) propionate] was used. To the resin materials of all the examples, 0.2 parts by weight of the hindered-phenol-based antioxidant was added.

[0073]

Examples 1 to 11 and Comparative Examples 1 to 4

Using a twin-screw extruder having a screw diameter of 30 mm and L/D of 35 equipped with a co-rotating vent (manufactured by Japan Steel Works, Ltd., TEX-30α), the raw materials were mixed according to the incorporation composition shown in Tables 2 to 4, and added from the main loading part. Further, in the examples where GF was incorporated, a side feeder was installed between the main loading part and the vent part to add GF, and the raw materials other than GF were added from the main loading part. Melt-mixing was performed under

extrusion conditions where the kneading temperature was 270°C and the screw rotation was at 150 rpm. The melt-mixture was discharged in strand form, passed through a cooling bus, and pelletized using a strand cutter.

[0074]

The obtained pellets were used, or, alternatively, the obtained pellets were dried and then molded to prepare each specimen, and the following physical properties were measured. The results are shown in Tables 2 to 4. The measurement method for each characteristic is as follows.

[0075]

(1) Flame Retardancy

Using an injection molding machine IS55EPN manufactured by Toshiba Machine Co., Ltd., a specimen for the evaluation of flame retardancy was formed by injection molding under the conditions where the molding temperature was 270°C and the die temperature was 70°C. Flame retardancy was evaluated in accordance with the evaluation criteria defined in the UL94 vertical test. Flame retardancy decreases in the following order: V-0 > V-1 > V-2. Further, the thickness of the specimens was 1/32 inch, 1/16 inch, and 1/8 inch. Generally, even in the specimens of the same V-0 rank, a specimen that is thinner but satisfies the evaluation criteria has higher flame retardancy of the raw material.

[0076]

(2) Tensile Test

Using an injection molding machine IS55EPN manufactured by Toshiba Machine Co., Ltd., an ISO527 dumbbell specimen having a thickness of 4 mm was formed by injection molding under the conditions where the molding temperature was 270°C and the die temperature was 70°C. The tensile strength and tensile elongation were measured according to ISO527-1, 2.

[0077]

(3) Flexural Test

Using an injection molding machine IS55EPN manufactured by Toshiba Machine Co., Ltd., an ISO527 dumbbell specimen having a thickness of 4 mm was formed by injection molding under the conditions where the molding temperature was 270°C and the die temperature was 70°C. The flexural strength was measured according to ISO527-1, 2.

[0078]

(4) Charpy Impact Test

Using an injection molding machine IS55EPN manufactured by Toshiba Machine Co., Ltd., a rectangular molded article having a thickness of 4 mm was formed by injection molding under the conditions where the molding temperature was 260°C and the die temperature was 70°C. The charpy impact strength (notched) was measured according to ISO179.

[0079]

(5) Cross-Sectional Observation of Specimen

An ultrathin section was cut from the above ISO527 dumbbell specimen, and then, at a magnification of 400 times using an optical microscope (ECLIPSE LV100 manufactured by Nikon Corporation), the cross section was observed about arbitrary 300 μm x 300 μm. The number of particles with a maximum particle size of more than 15 μm was confirmed at each observation site, and converted into the number of particles per 100 μm x 100 μm cross section.

[0080]

(6) Moldability

Using an injection molding machine IS55EPN manufactured by Toshiba Machine Co., Ltd., a box-shaped molded article with 30 mm in each side length and a thickness of 1 mm was formed under the conditions where the molding temperature was 270°C and the die temperature was 70°C, and runner release and mold-release properties were confirmed.

[0081]

(7) Mold deposit

Using an injection molding machine IS55EPN manufactured by Toshiba Machine Co., Ltd., a box-shaped molded article with 30 mm in each side length and a thickness of 1 mm was formed for 1000 shots under the conditions where the molding temperature was 270°C and the die temperature was 70°C, and then the level of mold deposit was confirmed.

[0082]
[Table 2]

			Example 1	Example 2	Example 3	Example 4	Example 5
A	PBT	wt%	20	20		25	20
	PET	wt%	20	20	40	20	20
	Other Resins	wt%					
B	MC Salt (kind)		B-1	B-2	B-1	B-1	B-1
	MC Salt (amount incorporated)	wt%	25	25	25	15	25
C	PX200	wt%	10	10	10	15	5
	OP1240	wt%					5
D	T-187	wt%	25	25	25	25	25
Physical Properties	Flame Retardancy (UL94)		V0(1/32'')	V0(1/32'')	V0(1/32'')	V0(1/16'')	V-0(1/32'')
	Tensile Strength	MPa	109	103	110	115	112
	Flexural Strength	MPa	166	159	165	170	169
	Impact Strength	kJ/m^2	8.4	8.3	9.0	9.2	9.4
	Number of Particles	-	0.1	0.7	0.1	0.1	0.2
Moldability			No problem	No problem	Mold-release properties, slightly poor	No problem	No problem
Mold Deposit			No problem	No problem	No problem	No problem	No problem

[0083]

[Table 3]

			Example 6	Example 7	Example 8	Example 9	Example 10	Example 11
A	PBT	wt%	20	40	40	25	20	60
	PET	wt%	20			20	20	
	Other Resins	wt%		PC, 5	PPS, 5			
B	MC Salt (kind)		B-1	B-1	B-1	B-1	B-1	B-1
	MC Salt (amount incorporated)	wt%	15	20	20	25	5	15
C	PX200	wt%	10	10	10	15	20	10
	OP1240	wt%	10				10	15
D	T-187	wt%	25	25	25	15	25	
Physical Properties	Flame Retardancy (UL94)		V-0(1/32'')	V0(1/32'')	V0(1/32'')	V0(1/32'')	V0(1/32'')	V0(1/16'')
	Tensile Strength	MPa	114	120	112	95	110	50
	Flexural Strength	MPa	169	173	167	150	171	85
	Impact Strength	kJ/m^2	8.5	10.5	8.7	6.4	8.8	3.0
	Number of Particles	-	0.2	0.1	0.1	0.3	0.1	0.2
Moldability			No problem	No problem	No problem	No problem	No problem	No problem
Mold Deposit			Mold deposit, slightly present	No problem	No problem	No problem	Presence of mold deposit	No problem

[0084]
[Table 4]

			Comparative Example 1	Comparative Example 2	Comparative Example 3	Comparative Example 4
A	PBT	wt%	20	20	20	20
	PET	wt%	20	20	20	15
	Other Resins	wt%				
B	MC Salt (kind)		B-3	B-4	B-5	B-1
	MC Salt (amount incorporated)	wt%	25	25	25	35
C	PX200	wt%	10	10	10	5
	OP1240	wt%				
D	T-187	wt%	25	25	25	25
Physical Properties	Flame Retardancy (UL94)		V0(1/32'')	V0(1/32'')	V0(1/32'')	V0(1/8'')
	Tensile Strength	MPa	90	90	81	104
	Flexural Strength	MPa	144	143	127	162
	Impact Strength	kJ/m^2	7.4	7.3	6.4	7.9
	Number of Particles	-	8	2	3	0.2
Moldability			No problem	No problem	Mold-release properties, slightly poor	No problem
Mold Deposit			No problem	No problem	No problem	No problem

INDUSTRIAL APPLICABILITY

[0085]

The molded article made of the flame-retardant polyester resin composition of the present invention uses a non-halogen-based flame retardant that is believed to have less impact on the environment, and has safety against electrical fire in the equipment and also has high flame retardancy to fire on the molded article itself. The molded article is thus useful for electrical/electronic parts, machine mechanism parts, and automobile parts. Specifically, breakers, electromagnetic switches, focus cases, flyback transformers, molded articles for fusers for copiers and printers, general household electrical appliances, housings for office-automation equipments, coil bobbins, connectors, relays, disk drive chassis, transformers, switch parts, receptacle parts, motor parts, sockets, plugs, condensers, various cases, resistors, electrical/electronic parts into which metal terminals or conducting wires are incorporated, computer-associated parts, audio parts, laser discs and like sound parts, illumination parts, parts associated with telegraph/telephone devices, air conditioner parts, parts for household electrical appliances such as VTRs and televisions, parts for copiers, parts for fax machines, parts for optical devices, automobile ignition parts, connectors for automobiles, various electrical equipment parts for automobiles, and the like can be mentioned.

[0086]

In the present invention, a highly reliable molded article having excellent performance regarding, particularly, flame retardancy, hydrolysis resistance, and metal contamination can be obtained. Accordingly, it is especially useful for electrical/electronic parts, such as relays, breakers, electromagnetic switches, focus cases, flyback transformers, and molded articles for fusers for copiers or printers.

CLAIMS

1. A flame-retardant polyester resin composition comprising (A) a polyester resin and (B) a salt of a triazine-based compound with cyanuric acid or isocyanuric acid, wherein

the composition comprises the component (B) in an amount of 1 to 70 parts by weight based on 100 parts by weight of the component (A), and

in the composition, the number of particles of the component (B) with a maximum size of more than 15 μm is 1 or less per 100 μm x 100 μm cross section.

2. The flame-retardant polyester resin composition according to claim 1, wherein among the diffraction lines at $2\theta = 11.0 \pm 0.5°$ in the X ray diffraction spectrum of the component (B), the diffraction line with the highest peak intensity has a full width at half maximum of 0.4° or less.

3. The flame-retardant polyester resin composition according to claim 1 or 2, wherein the amount of an extract which is extracted from the component (B) by a 80% by volume methanol/20 % by volume benzene mixed solvent is 0.06% by weight or less.

4. The flame-retardant polyester resin composition

according to claim 1 or 2, further comprising (C) a compound selected from phosphate esters and phosphinate salts, in an amount of 1 to 100 parts by weight based on 100 parts by weight of the component (A).

5. The flame-retardant polyester resin composition according to claim 1 or 2, wherein in the composition, the number of particles of the component (B) with a maximum size of more than 15 μm is 0.5 or less per 100 μm x 100 μm cross section.

6. The flame-retardant polyester resin composition according to claim 1 or 2, wherein the component (A) is a polyester selected from polyethylene terephthalate and polybutylene terephthalate.

7. The flame-retardant polyester resin composition according to claim 4, wherein the component (C) comprises a phosphate ester and a phosphinate salt.

8. The flame-retardant polyester resin composition according to claim 4, wherein the phosphate ester in the component (C) comprises at least one kind of aromatic phosphate ester.

9. The flame-retardant polyester resin composition

according to claim 4, wherein the phosphinate salt in the component (C) comprises at least one kind of a salt of a metal selected from calcium, aluminum, and zinc.

10. A molded article comprising the flame-retardant polyester resin composition according to any one of claims 1 to 9.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/CN2011/000972

A. CLASSIFICATION OF SUBJECT MATTER

See extra sheet

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC: C08L67/-

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

WPI, EPODOC, CNKI,CNPAT polyester, polyethylene terephthalate, polybutylene terephthalate, polyalkylene terephthalate, PET, PBT, +triazine, +melamine, +guanamine, cyanuric, cyanurate, isocyanuric, isocyanurate, phosphate, phosphin+, diphosphin+, light diffraction, extract, diameter, size, flam+

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO9967326A1(BASF AG) 29 December 1999 (29.12.1999) Examples 5,7	1-10
X	CN101684192A(TORAY FIBER RES INST CHINA CO LTD) 31 March 2010(31.03 2010) Example 1 and Page 6, Lines 10-15	1-6, 8, 10
X	JP2010024272A (MITSUBISHI ENG PLASTICS KK) 04 February 2010(04.02.2010) Example 1	1-6, 9, 10
X	JP2007039853A (MITSUBISHI ENG PLASTICS KK) 15 February 2007(15.02.2007) Examples	1-6, 8, 10
X	JP2009227750A(TORAY IND INC) 08 October 2009(08.10.2009) Examples	1-6, 8, 10
X	JP2005325216A(TORAY IND INC) 24 November 2005(24.11.2005) Examples	1-6, 8, 10
X	JP6157880A(AKZO KASHIMA KK) 07 June 1994(07.06.1994) Examples	1-6, 8, 10

☒ Further documents are listed in the continuation of Box C. ☒ See patent family annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier application or patent but published on or after the international filing date

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&"document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
28 August 2011(28.08.2011)	**08 Sep. 2011 (08.09.2011)**
Name and mailing address of the ISA/CN The State Intellectual Property Office, the P.R.China 6 Xitucheng Rd., Jimen Bridge, Haidian District, Beijing, China 100088 Facsimile No. 86-10-62019451	Authorized officer TANG, Dongmei Telephone No. (86-10)62084436

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INTERNATIONAL SEARCH REPORT

International application No.
PCT/CN2011/000972

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	CN101346429A(WINTECH POLYMER LTD) 14 January 2009(14.01.2009) Examples	1-6, 9, 10
X	CN1894319A(GENERAL ELECTRIC CO) 10 January 2007(10.01.2007) Examples	1-6, 9, 10
X	US2009036578A1(SABIC INNOVATIVE PLASTICS IP BV) 05 February 2009(05.02 2009) Examples	1-6, 8-10

INTERNATIONAL SEARCH REPORT
Information on patent family members

International application No.
PCT/CN2011/000972

Patent Documents referred in the Report	Publication Date	Patent Family	Publication Date
WO9967326A1	29.12.1999	DE19827845A1	30.12.1999
		AU4771399A	10.01.2000
CN101684192A	31.03.2010	None	
JP2010024272A	04.02.2010	None	
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JP2009227750A	08.10.2009	None	
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US2009036578A1	05.02.2009	WO2009009249A2	15.01.2009
		WO2009009249A3	30.04.2009

INTERNATIONAL SEARCH REPORT

International application No.
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A. CLASSIFICATION OF SUBJECT MATTER

C08L67/00 (2006.01) i

C08L67/02 (2006.01) i

C08K5/3492 (2006.01) i

C08K5/521 (2006.01) i

C08K5/523 (2006.01) i

C08K5/5313 (2006.01) i