

TITLE OF THE INVENTION

THERMOPLASTIC POLYESTER RESIN COMPOSITION

BACKGROUND OF THE INVENTION

5 Field of the Invention

The present invention relates to a thermoplastic polyester resin composition that offers high rigidity, high impact strength and high heat resistance and is also excellent in flowability and productivity for suitable application to various types of structural members for
10 electrical components and OA components.

Description of the Related Art

Thermoplastic polyester resins are widely used in many fields as fibers, films, sheets, structural members, etc., for their excellent
15 mechanical properties (e.g., rigidity and impact strength) as well as for their relatively low prices. In particular, structural members for electrical components and OA components require high heat resistance in addition to high rigidity and high impact strength. For this reason, for example, thermoplastic polyester resins have been
20 extensively blended with inorganic fillers such as glass fiber to form thermoplastic polyester resin compositions.

To improve the mechanical properties and heat resistance of such thermoplastic polyester resin compositions, it is required that a large amount of inorganic filler such as glass fiber be contained, but
25 this undesirably reduces the flowability of the thermoplastic polyester

resin compositions and affects their moldability. Although the addition of a nucleating agent, a crystallization accelerator, and/or a plasticizer together with inorganic filler can increase flowability, it results in a problem that mechanical properties – particularly impact strength – deteriorate.

To solve these problems, methods have been proposed that allow a thermoplastic polyester resin composition to contain a compound that is reactive with thermoplastic polyester resins. Examples of thermoplastic polyester resin compositions containing such a reactive compound include a thermoplastic polyester resin composition containing a carbodiimide compound (see Japanese Patent Application Laid-Open (JP-A) No. 11-49939); a thermoplastic polyester resin composition containing brominated epoxy (see JP-A No. 09-241488); a thermoplastic polyester resin composition containing a compound selected from glycidyl ether, carbodiimide, ketene imine, aziridine, and isocyanate (see JP-A No. 10-130482); and a thermoplastic polyester resin composition containing a multifunctional compound (see JP-A No. 10-110090).

These reactive compounds, however, tend to undergo gelation to produce solidified material in dies upon kneading and extrusion, resulting in reduced productivity due to the occurrence of strand break and/or surging. In addition, the thermoplastic polyester resin composition obtained after kneading has a problem that it shows reduced flowability and/or suffers from unstable mechanical properties (e.g., rigidity and impact strength) and unstable heat

resistance. As described above, thermoplastic polyester resin compositions have not yet been provided in which a good balance is struck among mechanical properties, heat resistance, flowability, and productivity.

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SUMMARY OF THE INVENTION

It is an object of the present invention to overcome the foregoing problems and to achieve the object described below. Namely, it is an object of the present invention to provide a
10 thermoplastic polyester resin composition that offers high rigidity, high impact strength and high heat resistance and is also excellent in flowability and productivity.

The present inventors have diligently conducted studies to solve the foregoing problems. As a result, they have established that
15 this is made possible by allowing a thermoplastic polyester resin to contain a specific polyalkylene glycol derivative and a specific reactive compound, thereby completing the present invention.

The present invention has been accomplished based on the above-described finding, and the means to solve the foregoing
20 problems are as follows: a thermoplastic polyester resin composition including (A) a thermoplastic polyester resin; (B) a polyalkylene glycol or derivative thereof, having at least one of a carboxylic acid ester structure and an alkylether structure at least at one terminal thereof; (C) a reactive compound which is a polymerization product of raw
25 monomers containing a vinyl monomer having a functional group that

is reactive with the thermoplastic polyester resin; and (D) an inorganic filler.

Preferably, in the polyalkylene glycol or derivative thereof, at least one of the carboxylic acid ester structure and the alkylether
5 structure has an alkyl group, and the number of carbon atoms in the alkyl group is 5 to 12.

Preferably, the raw monomers further contain styrene.

Preferably, in the reactive compound, the functional group that is reactive with the thermoplastic polyester resin is an epoxy group.

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DESCRIPTION OF THE PREFERRED EMBODIMENTS

(Thermoplastic Polyester Resin Composition)

The thermoplastic polyester resin composition of the present invention contains (A) a thermoplastic polyester resin, (B) a
15 polyalkylene glycol or derivative thereof, (C) a reactive compound, and (D) an inorganic filler, and optionally contains additional ingredient(s) appropriately selected.

– (A) Thermoplastic Polyester Resin –

The thermoplastic polyester resin is not particularly limited and
20 can be appropriately selected depending on the intended purpose; for example, it is preferably selected from those in which its dicarboxylic acid component is mainly consisting of an aromatic dicarboxylic acid and its glycol component is mainly consisting of an alkylene glycol.

Examples of the aromatic dicarboxylic acid include terephthalic
25 acid, isophthalic acid, naphthalene dicarboxylic acid, diphenyl

dicarboxylic acid, diphenylether dicarboxylic acid, diphenylketone dicarboxylic acid, and diphenoxyethane dicarboxylic acid. These aromatic dicarboxylic acids may be used singly or in combination.

Examples of the alkylene glycol include ethylene glycol, trimethylene glycol, tetramethylene glycol, pentamethylene glycol, and hexamethylene glycol. These alkylene glycols may be used singly or in combination.

The limiting viscosity number of the thermoplastic polyester resin is not particularly limited and can be appropriately determined depending on the intended purpose; however, it is preferably 0.4 to 1.5 and, more preferably, 0.5 to 1.2. If the limiting viscosity number is less than 0.4, such mechanical strength as rigidity and impact strength may be reduced, whereas if it exceeds 1.5, flowability may be reduced to result in poor moldability. On the other hand, setting the limiting viscosity number to within the foregoing more preferable range is advantageous because a better balance is struck between mechanical strength and flowability.

The limiting viscosity number is a value measured in accordance with ISO1628 using an Ostwald viscometer in o-chlorophenol at 35°C; the unit is 100mL/g.

Suitable examples of commercial products of the thermoplastic polyester resin include DIANITE MA-521-R25, MA-521-D25, and PA-200-D25 (all produced by Mitsubishi Rayon Co., Ltd.).

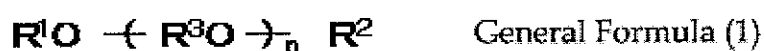
The foregoing thermoplastic polyester resins may be used singly or in combination.

The content of the thermoplastic polyester resin in the thermoplastic polyester resin composition is not particularly limited and can be appropriately determined depending on the intended purpose; however, it is preferably 60% by mass to 90% by mass and, more preferably, 70% by mass to 80% by mass. If less than 60% by mass is used, it may result in poor moldability due to reduced flowability. If greater than 90% by mass is used, such mechanical strength as rigidity and impact strength may be reduced. The use of the thermoplastic polyester resin in an amount falling within the foregoing more preferable content range is advantageous because a better balance is struck between mechanical strength and flowability.

– (B) Polyalkylene Glycol or Derivative Thereof –

The polyalkylene glycol or derivative thereof is used to increase the flowability and heat resistance of the thermoplastic polyester resin composition.

Such a polyalkylene glycol or derivative thereof is not particularly limited as long as it has at least one of a carboxylic acid ester structure and an alkyl ether structure at least at one terminal thereof, and can be appropriately selected; examples include polyalkylene glycol carboxylic acid esters and polyalkylene glycol alkyl ethers, represented by the following General Formula (1):



where R¹ and R² each represents a hydrogen atom, alkyl group or acyl group, R³ represents an alkylene group of 2 to 4 carbon atoms, and “n”

represents any positive number (note here that R¹ and R² do not represent a hydrogen atom at the same time).

The degree of polymerization of the polyalkylene glycol unit – the number of “n” in the General Formula (1) shown above – in the polyalkylene glycol or derivative thereof is not particularly limited and can be appropriately determined depending on the intended purpose; however it is preferably 5 to 25 in view of its compatibility with the thermoplastic polyester resin. If the degree of polymerization is less than 5, the problem that the polyalkylene glycol component bleeds out of the molded article is likely to occur, whereas if it exceeds 25, the ability of the polyalkylene glycol component to be dispersed into the thermoplastic polyester resin decreases, which may result in reduced capability of increasing flowability and heat resistance. The weight-average molecular weight of the polyalkylene glycol or derivative thereof is preferably 300 to 3,000 and, more preferably, 350 to 2,500.

At least one of the carboxylic ester structure and alkyl ether structure, both of which form a terminal of the polyalkylene glycol or derivative thereof, preferably has an alkyl group. The number of carbon atoms present in the alkyl group is not particularly limited and can be appropriately determined depending on the intended purpose; however, it is preferably 5 to 12 in view of its compatibility with the thermoplastic polyester resin. If the number of carbon atoms is less than 5, the problem that the polyalkylene glycol component bleeds out of the molded article is likely to occur, whereas if it exceeds 12, the

ability of the polyalkylene glycol component to be dispersed into the thermoplastic polyester resin decreases, which may reduce its capability to increase flowability and heat resistance. The weight-average molecular weight can be determined by gel permeation chromatography (GPC) using polystyrene as a standard and tetrahydrofuran (THF) as an eluent and by using a refractive index (RI) detector. The structure and the number of carbon atoms in $-R^1O$ and R^2 in the General Formula (1) can be determined by 1H -NMR. The degree of polymerization of the polyalkylene glycol component can be calculated by subtracting the total molecular weight of R^1O and R^2 from the polyalkylene glycol component's weight-average molecular weight and dividing the resultant value by a per unit molecular weight of polyalkylene glycol (i.e., the molecular weight of $-R^3O$ in the General Formula (1)).

Suitable examples of commercial products of the polyalkylene glycol or derivative thereof include LIONON DEH-40 and LEOFAT LA90-92 (both produced by LION Corporation).

The foregoing polyalkylene glycols or derivatives thereof may be used singly or in combination.

The content of the polyalkylene glycol or derivative thereof in the thermoplastic polyester resin composition is not particularly limited and can be appropriately determined depending on the intended purpose; however, it is preferably 0.5% by mass to 10% by mass and, more preferably, 1% by mass to 8% by mass. If less than 0.5% by mass is used, it may result in reduced capability of increasing

flowability and heat resistance. If greater than 10% by mass is used, such mechanical strength as rigidity and impact strength may be reduced. The use of the polyalkylene glycol or derivative thereof in an amount falling within the foregoing more preferable content range
5 is advantageous because a better balance is struck among flowability, heat resistance, and mechanical strength.

- (C) Reactive Compound -

The reactive compound is used to increase the rigidity and impact strength of the thermoplastic polyester resin composition.

10 The reactive compound is not particularly limited as long as it is a polymerization product of raw monomers containing a vinyl monomer having a functional group that is reactive with the thermoplastic polyester resin, and can be appropriately selected from those containing such a vinyl monomer. Among these, reactive
15 compounds that further contain styrene as a raw monomer are preferable in view of their compatibility with the thermoplastic polyester resin and thermal stability upon kneading, as well as of their handleability upon kneading because they solidify due to increased glass transition temperatures.

20 Examples of "the functional group that is reactive with the thermoplastic polyester resin" include functional groups that are reactive with hydroxyl group and/or carboxylic group bonded to a terminal of the thermoplastic polyester resin. Specific examples of such functional groups include epoxy, oxazoline, carbodiimide, and
25 isocyanate groups. Among these, epoxy group is preferable in view

of its high reactivity with functional groups bonded to a terminal of the thermoplastic polyester resin.

In addition, "the vinyl monomer having a functional group that is reactive with the thermoplastic polyester resin" is not particularly limited and can be appropriately selected; examples include
5 unsaturated carboxylic acid monomers such as acrylic acid and methacrylic acid, epoxy group-containing unsaturated monomers such as glycidyl methacrylate, and oxazoline group-containing unsaturated monomers such as isopropenyl oxazoline.

10 Note that "a functional group that is reactive with the thermoplastic polyester resin" may be contained either in the main chain or side chain of the reactive compound.

The weight-average molecular weight of the reactive compound, as determined by gel permeation chromatography (GPC) in terms of
15 the equivalent polystyrene molecular weight, is preferably 2,000 to 30,000, more preferably 4,000 to 20,000, still more preferably, 6,000 to 15,000 and, most preferably, 8,000 to 13,000. If less than 2,000 is used, the reactive compound is susceptible to gelation, and if greater than 30,000 is used, its ability to be dispersed into the thermoplastic
20 polyester resin decreases, resulting in reduced capability of increasing such mechanical strength as rigidity and impact strength. The use of a reactive compound whose weight-average molecular weight falls within the most preferable range is advantageous because the reactive compound is not gelled and mechanical strength can be further
25 increased.

The epoxy value of the reactive compound as determined in accordance with ASTM D-1652-73 is preferably 0.8 meq/g to 3.0 meq/g, more preferably 1.2 meq/g to 2.6 meq/g and, most preferably, 1.6 meq/g to 2.2 meq/g. If less than 0.8 meq/g is used, its reactivity with a functional group bonded to the terminal of the thermoplastic polyester resin is reduced, resulting in reduced capability of increasing such mechanical strength as rigidity and impact strength in some instances. If greater than 3.0 meq/g is used, the reactive compound tends to undergo gelation. The use of a reactive compound whose epoxy value falls within the most preferable range is advantageous because its capability of increasing mechanical strength becomes more stable without regard to the kneading conditions.

The process for producing the reactive compound is not particularly limited and can be appropriately selected from those known in the art; for example, processes described in JP-A Nos. 59-6207 and 60-215007 can be employed.

Examples of commercial products of the reactive compound include ARUFON UG-4030, UG-4040 and UG-4070 (reactive compounds prepared from raw monomers containing epoxy group-containing monomers, all produced by TOAGOSEI Co., Ltd.); ARUFON UFX-5010 (reactive compound prepared from raw monomers containing carboxylic group-containing monomers, produced by TOAGOSEI Co., Ltd.); and RPS-1005 (reactive compound prepared from raw monomers containing oxazoline group-containing monomers, produced by Nippon Shokubai Co., Ltd.).

Among these, ARUFON UG-4030, which contains epoxy group-containing monomers as a copolymerization ingredient, is most preferable.

5 The foregoing reactive compounds may be used singly or in combination.

The content of the reactive compound in the thermoplastic polyester resin composition is not particularly limited and can be appropriately determined depending on the intended purpose; however, it is preferably 0.5% by mass to 10% by mass and, more preferably, 1% by mass to 5% by mass. If less than 0.5% by mass is used, it may result in reduced capability of increasing such mechanical strength as rigidity and impact strength. If greater than 10% by mass is used, mechanical strength increases but flowability decreases, resulting in poor moldability in some cases. The use of the reactive compound in an amount falling within the foregoing more preferable content range is advantageous because a better balance is struck between mechanical strength and flowability.

– (D) Inorganic Filler –

20 The inorganic filler is used to increase the heat resistance, rigidity, and impact strength of the thermoplastic polyester resin composition.

The inorganic filler is not particularly limited and can be appropriately selected; examples include glass fiber, carbon fiber, potassium titanate fiber, glass beads, glass flake, mica, talc, clay, kaolin, calcium silicate, calcium carbonate, and calcium sulfate. Among

these, fibrous fillers such as glass fiber and carbon fiber are preferable.

The glass fiber is not particularly limited and can be appropriately selected; examples include chopped strands of glass fiber. In view of increased handleability, it is preferable that the glass
5 fiber be 3 mm to 6 mm in length and have a fiber density of 10k to 20k, which the density achieved using a sizing agent. The diameter of the glass fiber is preferably 5 μm to 15 μm and, more preferably, 7 μm to 12 μm .

The carbon fiber is not particularly limited and can be
10 appropriately selected; examples include chopped strands of pitch carbon fiber and chopped strand of PAN carbon fiber. In view of increased handleability, it is preferable that the carbon fiber be 3 mm to 6 mm in length and have a fiber density of 10k to 20k, which the density achieved using a sizing agent. The diameter of the carbon
15 fiber is preferably 5 μm to 15 μm and, more preferably 7 μm to 12 μm .

The foregoing inorganic fillers may be used singly or in combination.

The content of the inorganic filler in the thermoplastic polyester resin composition is preferably 1% by mass to 40% by mass and, more
20 preferably, 5% by mass to 30% by mass. If less than 1% by mass is used, it may result in reduced capability of increasing such mechanical strength as rigidity and impact strength and heat resistance. If greater than 40% by mass is used, flowability decreases to result in poor moldability in some cases. The use of the inorganic filler in an
25 amount falling within the foregoing more preferable content range is

advantageous because a better balance is struck among mechanical strength, heat resistance and flowability.

- Additional Ingredient -

The additional ingredient is not particularly limited and can be
5 appropriately selected; examples include known additives such as
conducting agents, plasticizers, thermal stabilizers, processing aids,
various types of fire retardants, lubricants, colorants, flowability
improvers, antistatic agents, compatibilizing agents, weatherization
agents, nucleating agents, and laser marking agents. Note that the
10 laser marking agents are not particularly limited and can be
appropriately selected depending on the print color or the like.

The content of the additional ingredient in the thermoplastic
polyester resin composition is not particularly limited either, and can
be appropriately determined depending on the intended purpose that
15 does not impair the effect of the present invention.

- Production Process -

The process for producing the thermoplastic polyester resin
composition is not particularly limited and can be selected from
processes known in the art. For example, the following processes are
20 exemplified: a process in which the foregoing ingredients are placed
into a pre-mixer (e.g., a Henschel Mixer or a tumbler) at the same time
and mixed until homogenous; and a process in which specific
ingredients are individually placed into a kneader using a volumetric
feeder.

25 In addition, the apparatus used to produce the thermoplastic

polyester resin composition is not particularly limited and can be appropriately selected depending on the intended purpose; examples include twin screw extruders, single screw extruders, rolls, kneaders, and Banbury mixers. Among these, twin screw extruders provided
5 with an upstream supply port and one or more downstream supply ports are preferable because (A) a thermoplastic polyester resin, (B) a polyalkylene glycol or derivative thereof, and (C) a reactive compound can be kneaded before addition of (D) an inorganic filler.

- Applications -

10 Since the thermoplastic polyester resin composition offers high rigidity, high impact strength and high heat resistance and is excellent in flowability and productivity, it can be suitably applied for instance to structural members for electrical components, OA components and the like by molding it using an existing molding machine like an
15 injection molding machine or an extrusion molding machine. Such structural members are not particularly limited; examples include molded articles for packing electrical components (e.g., magazines and trays), and molded articles for OA components (e.g., structural members for printers, scanners and facsimiles, and cartridges).

20

Examples

Hereinafter, Examples of the present invention will be described, which however shall not be construed as limiting the invention thereto.

25

The production process for thermoplastic polyester resin

compositions of Examples will be first described.

As a production apparatus, a twin screw extruder (NR-II57mm, manufactured by Nakatani Kikai Co., Ltd.) was employed. A mixture of (A) a thermoplastic polyester resin, (B) a polyalkylene glycol derivative, (C) a reactive compound and an additional ingredient was introduced into the extruder through its upstream supply port using a volumetric feeder. After the mixture was completely melted, (D) an inorganic filler was introduced into the extruder through the downstream supply port using the volumetric feeder, followed by kneading (cylinder and die temperatures were set to 290°C to 270°C). After kneading, a strand-shaped compound ejected out of the dies was cooled and then cut into cylindrical pellets using a pelletizer.

The evaluation method for the resultant thermoplastic polyester resin composition will be described below.

(Heat Resistance)

The obtained pellets were dried by hot air at 140°C for 3 hours, and dumbbell test pieces were fabricated using an injection molding machine (FD120S5ASE, manufactured by Nissei Plastic Industrial Co., Ltd.) where the cylinder temperature was set to 280°C and mold temperature to 80°C). The deflection temperature under load was measured for each test piece in accordance with ISO75-2Af. In addition, the coefficient of linear expansion was determined in the lengthwise direction in accordance with ASTM E831 using test machine TMA/SS6000 (manufactured by Seiko Instruments Inc.,

measurement temperature range = 60°C to 138°C, heating rate = 5°C/min, test piece dimension = 5 mm (width) x 10 mm (length) x 3.2 mm (thickness).

In Examples of the present invention a test piece that showed a
5 deflection temperature under load of 130°C or more and coefficient of linear expansion of $1.0 (10^{-5} \times ^\circ\text{C}^{-1})$ was considered of high heat resistance.

(Rigidity)

The obtained pellets were dried by hot air at 140°C for 3 hours,
10 and dumbbell test pieces were fabricated using an injection molding machine where the cylinder temperature was set to 280°C and mold temperature was set to 80°C). The tensile strength was measured for each test piece in accordance with ISO527-2/1A/5.

In Examples of the present invention a test piece that showed a
15 tensile strength of 90Mpa or more was considered of high rigidity.

(Impact Strength)

The obtained pellets were dried by hot air at 140°C for 3 hours, and dumbbell test pieces were fabricated using an injection molding machine where the cylinder temperature was set to 280°C and mold
20 temperature was set to 80°C. Charpy impact strength was then measured for each test piece in accordance with ISO179/1eA.

In Examples of the present invention a test piece that showed Charpy impact strength of 4.0 kJ/m² or more was considered of high impact strength.

25 (Flowability)

The obtained pellets were dried by hot air at 140°C for 3 hours and subjected to measurement of melt viscosity in accordance with ISO11443 using test machine 140SAS2002 (manufactured by Yasuda Seiki Seisakusyo Ltd., temperature = 280°C, shear rate = 1000 sec⁻¹, die dimension = 1 mm in diameter and 10 mm in length).

In Examples of the present invention a test piece that showed a melt viscosity of 180 Pa·s or less was considered of high flowability.
(Productivity)

As a measure for the productivity of the thermoplastic polyester resin composition, the number of times a resin composition strand continuously extruding out of the extruder was broken in two was counted (per hour). Strand break occurs when a gelled reactive compound accumulated inside the extruder dies.

In Examples of the present invention a test piece that showed no strand break, i.e., the fact that the number of times the strand was broken in two was 0 was considered of high productivity.
(Example 1)

A thermoplastic polyester resin composition of Example 1 was obtained through the foregoing production process using the following ingredients: as (A) a thermoplastic polyester resin, (A-1) 78% by mass of polyethylene terephthalate resin with a limiting viscosity number of 0.78 (100mL/g) (product name: MA521-D25, produced by Mitsubishi Rayon Co., Ltd.); as (B) a polyalkylene glycol derivative, (B-1) 3.0% by mass of polyethylene glycol-di-2-ethylhexoate obtained by esterifying both terminals of polyethylene glycol (product name:

LIONON DEH-40, produced by LION Corporation); as (C) a reactive compound, (C-1) 2.0% by mass of styrene-methymethacrylate-glycidylmethacrylate copolymer with a weight-average molecular weight of 11,000 and an epoxy value of 1.8 meq/g (product name: ARUFON UG-4030, produced by TOAGOSEI Co., Ltd.); as (D) an inorganic filler, (D-1) 15.0% by mass of chopped strands of glass fiber of 10 μ m in diameter and 3 mm in length (product name: RES03-TP78, produced by Nippon Sheet Glass Co., Ltd.); as a colorant, 1.5% by mass of carbon black with a DBP oil absorption of 157 mL/100g as measured using a DBP absorptometer in accordance with ASTM D2414 (product name: ASAHI F-200, produced by Asahi Carbon Co., Ltd.); and as an antioxidant, 0.5% by mass of a hindered phenol antioxidant (product name: AO-60, produced by ADEKA Corporation).

The evaluation results are shown in Table 1.

(Examples 2 to 10)

As in Example 1, thermoplastic polyester resin compositions of Examples 2 to 10 were prepared using the ingredients shown in Table 1.

As (B) a polyalkylene glycol derivative, (B-2) methoxypolyethylene glycol laurate obtained by esterifying one terminal of polyethylene glycol and etherifying the other terminal was employed (product name: LEOFAT LA90-92, produced by LION Corporation). As (C) reactive compounds, the following were employed: (C-2) styrene-methylmethacrylate-glycidylmethacrylate

copolymer with a weight-average molecular weight of 9,700 and an epoxy value of 1.4 meq/g (product name: ARUFON UG-4070, produced by TOAGOSEI Co., Ltd.); (C-3) styrene-free methylmethacrylate-glycidylmethacrylate copolymer with a weight-average molecular weight of 2,900 and an epoxy value of 1.4 meq/g (product name: ARUFON XG-4010, produced by TOAGOSEI Co., Ltd.); and (C-4) styrene-acrylic acid-tetradecylacrylate copolymer with a weight-average molecular weight of 15,000 and having carboxylic groups as functional groups that are reactive with thermoplastic polyester resins (product name: ARUFON UFX-5010, produced by TOAGOSEI Co., Ltd.). The evaluation results are shown in Table 1.

Table 1

Ingredient(mass%)	Ex. 1	Ex. 2	Ex. 3	Ex. 4	Ex. 5	Ex. 6	Ex. 7	Ex. 8	Ex. 9	Ex. 10
(A-1) MA521-D25	78.0	73.0	78.0	71.0	75.0	70.0	60.0	78.0	78.0	80.0
(B-1) LIONON DEH-40	3.0	8.0	—	10.0	3.0	3.0	—	3.0	3.0	3.0
(B-2) LEOFATLA-90-92	—	—	3.0	—	—	—	6.0	—	—	—
(C-1) ARUFON UG4030	2.0	—	2.0	2.0	5.0	—	2.0	—	—	2.0
(C-2) ARUFON UG4070	—	2.0	—	—	—	10.0	—	—	—	—
(C-3) ARUFON XG4010	—	—	—	—	—	—	—	2.0	—	—
(C-4) ARUFON UFX5010	—	—	—	—	—	—	—	—	2.0	—
(D-1) RES03-TP78	15.0	15.0	15.0	15.0	15.0	15.0	30.0	15.0	15.0	15.0
Colorant: ASAHI F-200	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	—
Antioxidant AO60	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	—
Deflection temperature under load (°C)	154	176	135	185	157	162	186	149	148	152
Coefficient of linear expansion ($10^{-5} \times ^\circ\text{C}^{-1}$)	2.2	2.5	1.5	2.6	2.1	2.4	2.2	2.0	1.9	2.2
Tensile strength(MPa)	115	108	110	106	126	136	145	104	101	116
Charpy impact strength (kJ/m ²)	5.0	4.8	5.1	4.6	5.2	5.8	6.3	4.6	4.4	4.9
Melt viscosity (Pa·s)	120	107	115	101	135	172	176	119	123	122
Productivity (the number of times strand break occurred)	0	0	0	0	0	0	0	0	0	0

The results shown in Table 1 show that the thermoplastic

polyester resin compositions of Examples 1 to 10 are all of high rigidity, high impact strength and high heat resistance and are excellent in flowability and productivity.

(Comparative Examples 1 to 7)

5 As in Example 1, thermoplastic polyester resin compositions of Comparative Examples 1 to 7 were prepared using the ingredients shown in Table 2.

As a comparison for (B) the alkylene glycol derivative, (B'-1) polyethylene glycol whose terminals are neither esterified nor
10 etherified was employed (product name: PEG#400, produced by LION Corporation). As a comparison for (C) the reactive compound, the following were employed: (C'-1) styrene-methylmethacrylate copolymer with a weight-average molecular weight of 8,000 and is free from functional groups that are reactive with polyesters (product
15 name: ARUFON UP-1100, produced by TOAGOSEI Co., Ltd.); (C'-2) bisphenol-A epoxy resin (product name: EPIKOTE 828, produced by Japan Epoxy Resin Co., Ltd.); and (C'-3) aromatic polycarbodiimide (product name: STABACSOL P, produced by Bayer A.G.). The evaluation results are shown in Table 2.

Table 2

Ingredient(mass%)	Comp. Ex. 1	Comp. Ex. 2	Comp. Ex. 3	Comp. Ex. 4	Comp. Ex. 5	Comp. Ex. 6	Comp. Ex. 7
(A-1) MA521-D25	81.0	78.0	80.0	78.0	78.0	78.0	93.0
(B-1) LIONON DEH-40	—	—	3.0	3.0	3.0	3.0	3.0
(B'-1) PEG#400	—	3.0	—	—	—	—	—
(C-1) ARUFON UG4030	2.0	2.0	—	—	—	—	2.0
(C'-1) ARUFON UP1100	—	—	—	2.0	—	—	—
(C'-2) EPIKOTE 828	—	—	—	—	2.0	—	—
(C'-3) STABACSOL P	—	—	—	—	—	2.0	—
(D-1) RES03-TP78	15.0	15.0	15.0	15.0	15.0	15.0	—
Colorant: ASAHI F-200	1.5	1.5	1.5	1.5	1.5	1.5	1.5
Antioxidant: AO60	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Deflection temperature under load (°C)	87	95	149	143	152	150	91
Coefficient of linear expansion ($10^{-5} \times ^\circ\text{C}^{-1}$)	-1.3	-0.7	1.8	1.7	1.9	1.6	-0.9
Tensile strength (MPa)	117	112	82	80	95	98	65
Charpy impact strength (kJ/m ²)	5.2	4.7	3.7	3.6	4.4	4.5	3.5
Melt viscosity (Pa·s)	220	202	98	82	205	210	72
Productivity (the number of times strand break occurred)	0	0	0	0	10	13	0

The results shown in Table 2 reveal the facts described below.

The thermoplastic polyester resin composition prepared in Comparative Example 1 is not blended with (B) a polyalkylene glycol derivative described in the present invention and thus offered low flowability and low heat resistance. The composition prepared in Comparative Example 2 is blended with (B) a polyalkylene glycol that lies outside of the scope of the present invention, i.e., that includes neither a carboxylic acid ester terminal nor an alkylether terminal, thus offering low flowability and low heat resistance. The composition prepared in Comparative Example 3 is not blended with (C) a reactive compound described in the present invention and thus offered low rigidity and low strength impact. The composition prepared in Comparative Example 4 is blended with (C) a reactive compound that lies outside of the scope of the present invention, i.e., that does not

bear functional groups that are reactive with thermoplastic polyester resins, thus offering low rigidity and low strength impact. The compositions prepared in Comparative Examples 5 and 6 are each blended with (C) a reactive compound that lies outside of the scope of the present invention and thus produced solidified material accumulated inside the dies upon kneading, resulting in a high likelihood of strand break and significantly reducing productivity. The composition prepared in Comparative Example 7 is not blended with (D) an inorganic filler and thus offered low heat resistance, low rigidity and low strength impact.

(Examples 11 to 17)

To each of the pellets obtained in Example 1 was added (E) a laser marking agent, and the pellets were kneaded again in a twin screw extruder to prepare pellets containing different laser marking agents shown in Table 3 (Examples 11 to 17). Note in Table 3 that the added amount of each laser marking agent is expressed in terms of mass percent based on the total mass of the composition of Example 1.

The following laser marking agents were employed: (E-1) mica with a particle diameter of 15 μm or less, which is coated with black oxide of iron (Fe_3O_4) (Laserflair LF835: LF835, produced by Merk Ltd.); (E-2) carbon black for pigment (#2600B, produced by Mitsubishi Chemical Corporation, arithmetic average particle diameter as measured using an electronic microscope = 13 μm , nitrogen adsorption specific area as measured in accordance with JIS K6217 = 370 m^2/g , DBP absorption as measured in accordance with JIS K6217 = 60

cm³/100g, the proportion of the volatilized portion after heated at 950°C for 7 hours = 1.8%, pH = 6.5); (E-3) exfoliated graphite (BSP60AS, produced by Chuetsu Graphite Works Co., Ltd., average particle diameter as measured by laser diffraction = 60 μm, ash content = 0.3%, volatile content = 0.7%); and (E-4) titanium dioxide (CR-EL, produced by Ishihara Sangyo Kaisha Ltd., specific surface area as measured by BET = 6.3 m²/g, rutilation percentage as determined by X ray diffraction = 99.9%). Each pellet was evaluated for its laser marking property as described below.

(Laser Marking Property)

The obtained pellets were dried by hot air at 140°C for 3 hours, and plate-shaped test pieces (10 cm (length) x 10 cm (wide) x 3.2 mm (thickness)) were fabricated using the injection molding machine where the cylinder temperature was set to 280°C and mold temperature to 80°C). Using a YAG laser, a mark was printed on the surface of the pellet under the marking condition described below. The evaluation results are shown in Table 3.

YAG laser: ML-7110B (manufactured by Miyachi Corporation)

Laser diode current: 18.0A

Marking speed: 1000 mm/s

Q switch frequency: 30.0 kHz

Table 3

	Ex. 11	Ex. 12	Ex. 13	Ex. 14	Ex. 15	Ex. 16	Ex. 17	Ex. 1
(E-1) LF835	0.1	0.3	0.5	1.0	—	—	—	—
(E-2) #2600B	—	—	—	—	0.5	—	—	—
(E-3) BSP60AS	—	—	—	—	—	0.5	—	—
(E-4) CR-EL	—	—	—	—	—	—	2.0	—
Laser marking property	Excellent	Excellent	Excellent	Excellent	Excellent	Excellent	Excellent	Excellent

mass% based on the total mass of the composition of Example 1

The thermoplastic polyester resin compositions prepared in Examples 11 to 17 were all capable of being printed with a vivid mark with any laser marking agent, offering excellent laser marking property. In addition, since the composition of Example 1 contains carbon black as a colorant, printing of a vivid mark was possible without having to add a laser marking agent, thereby offering excellent laser marking property.

As a result, it was established that the thermoplastic polyester resin composition of the present invention is also suitable for receiving marks by laser marking.

The thermoplastic polyester resin composition of the present invention offers high rigidity, high impact strength and high heat resistance and is also excellent in flowability and productivity; therefore, it can be suitably applied to structural members for electrical components such as gears, ink cartridges and paper feed guides, which are components of electric devices including personal computers and printers, to structural members for printers, scanners, and facsimiles, and to structural members for OA components such as cartridges.

What is claimed is:

1. A thermoplastic polyester resin composition comprising:

(A) a thermoplastic polyester resin;

(B) a polyalkylene glycol or derivative thereof, having at least one of a carboxylic acid ester structure and an alkylether structure at least at one terminal thereof;

(C) a reactive compound which is a polymerization product of raw monomers containing a vinyl monomer having a functional group that is reactive with the thermoplastic polyester resin; and

(D) an inorganic filler wherein at least one of the carboxylic acid ester structure and the alkylether structure in the polyalkylene glycol or derivative thereof has an alkyl group, and the number of carbon atoms in the alkyl group is 5 to 12.

2. The thermoplastic polyester resin composition according to one of claim 1, wherein the raw monomers further comprise styrene.

3. The thermoplastic polyester resin composition according to any one of claims 1 or 2, wherein in the reactive compound, the functional group that is reactive with the thermoplastic polyester resin is an epoxy group.