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(54) Title: POLYESTER RESIN COMPOSITION

(57) Abstract: The present invention relates to polyester resin compositions comprising (i) 25 to 39 wt. % of a polyester resin comprising at least a polytrimethylene terephthalate (PTT) resin; and (ii) 61 to 75 wt. % of a glass fiber, wherein the glass fiber comprises at least a flat glass fiber having a cross section with aspect ratio (long diameter/short diameter) of 2 to 10; (iii) 0 to 10 wt. % of at least one polymer additive; wherein the weight % of (i), (ii), and (iii) total 100 wt % of the polyester resin composition.



TITLE**POLYESTER RESIN COMPOSITION****FIELD OF THE INVENTION**

The invention relates to a molded article, more specifically a polyester

5 resin pellet and a polyester resin composition to form the molded article.

TECHNICAL BACKGROUND OF THE INVENTION

A molded article of polyester resin is often required to suffice strength and surface appearance.

10 US5059638 discloses a glass fiber-filled polyester resin composition. A fiber-filled polyester resin comprises a polyetherester, a poly(etherimide)ester or a blend of both, or a blend of one or both of a polyetherester or a poly(etherimide)ester with a poly(butylene terephthalate) or poly(ethylene terephthalate) other than a blend having a matrix phase of poly(butylene

15 terephthalate), poly(ethylene terephthalate) or both; and glass or mineral fibers that do not substantially adhere to the resin, the fiber-filled resin having an elongation at break when injection molded substantially greater than that of the same resin when filled with reinforcing glass or mineral fiber that adhere to the resin..

SUMMARY OF THE INVENTION

An objective is to provide a polyester resin composition to form a molded article having sufficient strength and appearance.

5 An aspect of the invention relates to a polyester resin composition comprising (i) 25 to 39 wt. % of polytrimethylene terephthalate (PTT) resin; and (ii) 61 to 75 wt. % of flat glass fibers having a cross section with aspect ratio (long diameter/short diameter) of 2 to 10; (iii) 0 to 10 wt. % of at least one polymer additive; wherein the weight % of (i), (ii), and (iii) total 100 wt % of the
10 polyester resin composition.

Another aspect of the invention relates to a polyester resin composition comprising: (i) 25 to 39 wt. % of a mixture of polytrimethylene terephthalate (PTT) resin and a second polyester resin selected from polyethylene terephthalate (PET), polybutylene terephthalate (PBT), polyethylene
15 2,6-naphthalate (PEN), polybutylene naphthalate (PBN), polycyclohexane dimethylene terephthalate (PCT), and mixtures of these polyester resins; and (ii) 61 to 75 wt. % of flat glass fibers having a cross section with aspect ratio (long diameter/short diameter) of 2 to 10; (iii) 0 to 10 wt. % of at least one polymer additive; wherein the weight % of (i), (ii), and (iii) total 100 wt % of the polyester

resin composition.

5 Another aspect of the invention relates to a polyester resin pellet comprising: (i) 25 to 39 wt. % of a polyester resin; and (ii) 61 to 75 wt. % of a glass fiber, wherein the glass fiber comprises at least a flat glass fiber having a cross section with aspect ratio (long diameter/short diameter) of 2 to 10, wherein the wt. % is based on the weight of the polyester resin pellet.

10 Another aspect of the invention relates to a molded article comprising: (i) 25 to 39 wt. % of a polyester resin; and (ii) 61 to 75 wt. % of a glass fiber, wherein the glass fiber comprises at least a flat glass fiber having a cross section with aspect ratio (long diameter/short diameter) of 2 to 10, wherein the wt. % is based on the weight of the molded article.

15 A polyester resin composition to form a molded article having sufficient strength and appearance can be provided by the present invention.

DETAILED DESCRIPTION OF THE INVENTION

The polyester resin composition comprises (i) 25 to 39 wt. % of a

polyester resin selected from polytrimethylene terephthalate (PTT), polyethylene terephthalate (PET), polybutylene terephthalate (PBT), polyethylene 2,6-naphthalate (PEN), polybutylene naphthalate (PBN), polycyclohexane dimethylene terephthalate (PCT), and mixtures of these polyester resins; (ii) 61 to 75 wt. % of glass fibers selected from flat glass fibers, or a combination of flat glass fibers and circular glass fibers wherein the weight ratio of flat glass fibers to circular glass fibers (flat glass fiber:circular glass fiber) is 1:0.01 to 1:10; and (iii) 0 to 10 wt. % of at least one polymer additive; wherein the weight % of (i), (ii), and (iii) total 100 wt % of the polyester resin composition.

10 (i) Polyester Resin

The polyester resin is condensation products of dicarboxylic acids and diols. The polyester resin comprises at least a polytrimethylene terephthalate (PTT) resin. The polyester resin can be a mixture of PTT resin and a second polyester resin selected from the group consisting of polyethylene terephthalate (PET), polybutylene terephthalate (PBT), polyethylene 2,6-naphthalate (PEN), polybutylene naphthalate (PBN), polycyclohexane dimethylene terephthalate (PCT), a copolymer thereof and a mixture thereof in an embodiment.

The weight ratio of the PTT resin and the second polyester resin (PTT:second polyester resin) ranges from about 1:1 to 5:1 in an embodiment,

1.5:1 to 4:1 in another embodiment, 2:1 to 3:1 in another embodiment. The second polyester resin is preferably selected from the group consisting of PET resin, PBT resin, and mixtures thereof in another embodiment. The polyester resin can be a mixture of PTT resin and PET resin in another embodiment.

5 As the examples of copolymers, some of the dicarboxylic acids or some of the diols can be added to the condensation products. The polyester resins may comprise small amounts of additional acids such as trimesic acid, trimellitic acid, pyromellitic acid, glycerol, and pentaerythritol which have more than three functional groups in another embodiment.

10 The polyester resin is 25 to 39 wt. %, 30 to 38 wt. % in another embodiment, 32 to 37 wt. % in another embodiment, based on the weight of the polyester resin composition.

(ii) Glass Fibers

The glass fibers may comprise at least flat glass fibers having a cross
15 section with aspect ratio (long diameter/short diameter) of 2 to 10. The flat glass fibers may have a cross section which is an ellipse, oval, or rectangle. Cross section of the flat glass fibers has the long diameter and the short diameter. The long diameter is the length of the longer side of a circumscribed rectangular of the flat glass fiber cross section. The short diameter is the length

of the shorter side of a circumscribed rectangular.

Aspect ratio is the ratio of the long diameter to the short diameter (long diameter/short diameter). The aspect ratio of the flat glass fibers is 2.5 to 8 in another embodiment, 3 to 6 in another embodiment. The short diameter of the

5 flat glass fiber is 1 to 20 μm in an embodiment, 3 to 16 μm in another embodiment, 5 to 13 μm in another embodiment.

Cross-sectional area of the flat glass fibers is 0.5×10^{-6} to $5.0 \times 10^{-6} \text{ cm}^2$ in an embodiment, 0.8×10^{-6} to $3.5 \times 10^{-6} \text{ cm}^2$ in another embodiment, 1.0×10^{-6} to $2.8 \times 10^{-6} \text{ cm}^2$ in another embodiment.

10 The flat glass fibers are 100 wt. % based on the weight of the glass fibers in an embodiment.

The glass fibers further comprise circular glass fibers in another embodiment. The circular glass fibers have a circle cross section with an aspect ratio of about 1. The diameter of the circular glass fiber may range from
15 15 to 20 μm in an embodiment, 16 to 19 μm in another embodiment, 16 to 18 μm in another embodiment.

The weight ratio of flat glass fibers and circular glass fibers (flat glass fiber:circular glass fiber) may range from about 1:0.01 to 1:10 in an embodiment, 1:0.1 to 1:10 in another embodiment, 1:0.4 to 1:6 in another embodiment, 1:0.7

to 1:2 in another embodiment.

The length of glass fibers may be from about 1 to 10 mm in an embodiment, 1.5 to 7 mm in another embodiment, 2 to 5 mm in another embodiment.

5 The glass fibers comprise 61 to 75 wt. %, 61.5 to 70 wt. % in another embodiment, 62 to 66 wt. % in another embodiment, based on the weight of the polyester resin composition.

(iii) Polymer Additive

10 The polyester resin composition can further comprise typical polymer additives in an embodiment. The polymer additives are compounds such as a flame retardant, an impact modifier, a viscosity modifier, a heat resistance improver, a lubricant, an antioxidant and a UV-stabilizer. The polymer additive can be tailored in accordance with required properties of the molded article. The polymer additives may comprise from 0 to 10 wt. % of the total weight of the
15 polyester resin composition.

The polyester resin compositions preferably exhibit a combination of tensile strengths of at least 22 GPa and surface gloss values of at least 40.

EXAMPLES

The present invention is illustrated by, but is not limited to, the following

examples.

The polyester resin compositions used for testing were prepared with the following procedure. PTT resin pellets, PET (IV = 0.66) resin pellets and/or PBT (IV = 0.88) resin pellets were used as obtained from the supplier. PTT was
5 obtained from E.I. DuPont de Nemours and Company, Wilmington, Delaware, USA as Sorona® Bright. Blends of polyester resins were obtained by mixing in a stainless tumble mixer with stirring for one hour. The following glass fibers were used:

Circular glass fiber: a chop strand glass fiber having a cross-section with
10 diameter (ϕ) of 10 μm , and length of 3 mm (NEG 187H available from Nippon Electric Glass Co., Ltd.).

Flat glass fiber: a chop strand glass fiber (CSG 3PA-830, available from Nitto Boseki Co., Ltd.) having an aspect ratio (long diameter/short diameter) of 4, short diameter of 9 μm , fiber length of 3 mm and cross-sectional area of 1.9 x
15 10^{-6} cm^2 .

The polyester resin(s) and the glass fibers were mixed in a twin-screw extruder to make a polyester resin composition. Amounts of materials are shown in Table 1. The extruder was fitted with a vacuum vent port in a barrel. The barrel temperature was set at approximately 260 deg. C. The polyester resin

composition was extruded in the form of strands, cooled, chopped into granules to form polyester resin composition pellets.

Test Methods

Compounding processability of the polyester resin composition was evaluated by visually observing the polyester resin composition pellets. The surface of polyester resin composition pellets was evaluated to determine if the pellet exhibited a rough surface and/or non-uniform shape or had a smooth surface and/or uniform thickness. The evaluation was "NG" when the polyester resin composition pellets had glass fiber fuzz on its surface or non-uniform shape. The evaluation was "OK" when the polyester resin composition pellets had smooth surface and uniform shape.

Tensile Modulus (GPa) was measured in accordance with ISO 527. The polyester resin composition pellets to be tested were injection molded to form ISO dumbbell bars (10 mm wide, 170 mm long, 4 mm thick) by a 100t-hydraulic injection molding machine. The cylinder temperature was set to 260 deg. C. The mold temperature was 130 deg. C.

The polyester resin composition pellets were also injection molded to form a plate (100 mm wide, 100 mm long, 2 mm thick) using 180t-electric injection molding machine to evaluate surface appearance of molded articles

obtained from the polyester resin compositions. The cylinder temperature was set to 260 deg. C. The mold temperature was 130 deg. C. Molded plates having glass fibers on its surface was evaluated “NG” as a defect. The molded plates having smooth surface was evaluated as “OK”. Gloss value of the

5 molded plate was also measured to quantitatively evaluate the surface appearance. Molded plates having a smooth surface exhibit gloss values greater than 40. The gloss value of the surface of the molded plates was measured with a gloss meter (VG-2000, Nippon Denshoku Industries Co., Ltd.) at 60 degree of lighting angle. The molded plates were set in the gloss meter

10 while keeping the direction of resin flow constant during injection molding.

Examples (Ex.) 1 and 2 show that desirable compounding processability, tensile modules over 22 GPa, and a surface gloss value over 40 can be obtained with the polyester resin compositions herein. Comparative Example (Com. Ex.) 1 and 2 show that the use of pure PBT or circular glass fibers having a diameter of

15 10 μm are unable to obtain the desired properties.

Table 1

		Ex. 1	Ex. 2	Com. Ex. 1	Com. Ex. 2
Polyester resin	PTT	35	25	38	0
	PET	0	10	15	0
	PBT	0	0	0	35
Glass fiber	Circular (ϕ 10 μm)	0	0	45	0
	Flat	63	63	0	63

Compounding processability	OK	OK	OK	OK
Tensile Modulus (GPa)	24.7	24.4	15.8	22.9
Surface appearance	OK	OK	OK	NG
Gloss value*	45.5	47.6	45.8	35.6

*Relative value

All values are in weight percent

Polyester resin compositions used in table 2 were prepared in the same manner as those in table 1 except for the amount of the materials as shown in the table. The circular glass fiber was a chop strand glass fiber having a cross-section with diameter (ϕ) of 10 μm , 13 μm or 17 μm , and length of 3 mm (NEG 187H, 187, 187N, respectively from Nippon Electric Glass Co., Ltd.) and the flat glass fibers were the same as used in table 1.

Example 3 in table 2, which is a blend of PTT and PET, shows that the use of circular glass fibers having a diameter of at least 15 microns in combination with flat glass fibers provides articles having a tensile modulus over 22 GPa and desirable surface gloss. Comparative examples 4-6, which use various combinations of circular glass fibers having diameters of less than 15 microns and flat glass fibers show that such compositions do not provide the desired combination of surface gloss and tensile modulus. Comparative (Com.) Example 3 and 7, using only circular glass fibers, also failed to provide the desired combination of physical properties.

Table 2

		Com. Ex. 3	Com. Ex. 4	Com. Ex. 5	Com. Ex. 6	Ex. 3	Com. Ex. 7
Polyester resin	PTT	27	27	25	25	25	25
	PET	11	11	10	10	10	10

Glass fiber	Circular ($\varnothing$ 10 μm)	60	30	30	0	0	0
	Circular ($\varnothing$ 13 μm)	0	0	0	30	0	0
	Circular ($\varnothing$ 17 μm)	0	0	0	0	30	63
	Flat	0	30	33	33	33	0
Compounding processability		NG	NG	OK	NG	OK	NG
Tensile Modulus (GPa)		22.2	24.8	23.0	24.0	24.5	22.4
Surface appearance		NG	NG	NG	NG	OK	NG

All values are in weight percent

CLAIMS

What is claimed is:

1. A polyester resin composition comprising:

(i) 25 to 39 wt. % of polytrimethylene terephthalate (PTT) resin; and

5 (ii) 61 to 75 wt. % of flat glass fibers having a cross section with aspect ratio

(long diameter/short diameter) of 2 to 10;

(iii) 0 to 10 wt. % of at least one polymer additive;

wherein the weight % of (i), (ii), and (iii) total 100 wt % of the polyester resin composition.

10 2. A polyester resin composition comprising:

(i) 25 to 39 wt. % of a mixture of polytrimethylene terephthalate (PTT) resin and a

second polyester resin selected from polyethylene terephthalate (PET),

polybutylene terephthalate (PBT), polyethylene 2,6-naphthalate (PEN),

polybutylene naphthalate (PBN), polycyclohexane dimethylene terephthalate

15 (PCT), and mixtures of these polyester resins; and

(ii) 61 to 75 wt. % of flat glass fibers having a cross section with aspect ratio

(long diameter/short diameter) of 2 to 10;

wherein the weight % of (i) and (ii) total 100 wt % of the polyester resin composition.

3. The polyester resin composition of claim 2, wherein weight ratio of the polytrimethylene terephthalate (PTT) resin and the second polyester resin (PTT:second polyester resin) is 1:1 to 5:1.

4. The polyester resin composition of claim 3 wherein the second polyester resin is polyethylene terephthalate (PET).

5. The polyester resin composition of claims 1 or 2, wherein length of the glass fibers ranges from 1 to 10 mm.

6. The polyester resin composition of claims 1 or 2, wherein cross-sectional area of the flat glass fiber is 0.5×10^{-6} to $5.0 \times 10^{-6} \text{ cm}^2$.

7. The polyester resin composition of claims 1 or 2, wherein the short diameter of the flat glass fiber is 1 to 20 μm .

8. The polyester resin composition of claims 1 or 2 having a surface gloss of at least 40 and a tensile modulus of at least 22 GPa.

9. The polyester resin composition of claims 1 or 2 additionally comprising circular glass fibers.

10. The polyester resin composition of claim 9 wherein the weight ratio of flat glass fibers to circular glass fibers (flat glass fiber:circular glass fiber) is 1:0.01 to 1:10.

11. The polyester resin composition of claim 10 wherein the weight ratio of flat

glass fibers to circular glass fibers is 1:0.1 to 1:1.

12. The polyester resin composition of claim 1 or 2 wherein polymer additive (iii)

is selected from the group consisting of flame retardants, impact modifiers,

viscosity modifiers, heat resistance improvers, lubricants, antioxidants,

5 UV-stabilizers, and combinations of these.

13. A polyester resin composition pellet comprising:

(i) 25 to 39 wt. % of a polyester resin; and

(ii) 61 to 75 wt. % of a glass fiber, wherein the glass fiber comprises at least a flat

glass fiber having a cross section with aspect ratio (long diameter/short

10 diameter) of 2 to 10,

wherein the wt. % is based on the weight of the polyester resin composition

pellet.

14. A molded article prepared from the polyester resin composition of any

preceding claim.

15

INTERNATIONAL SEARCH REPORT

International application No
PCT/US2018/016970A. CLASSIFICATION OF SUBJECT MATTER
INV. C08K7/14 C08L67/02
ADD.

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)
C08K

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)

EPO-Internal, WPI Data, CHEM ABS Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 2 020 282 A1 (MITSUBISHI ENG PLASTICS CORP [JP]) 4 February 2009 (2009-02-04)	13,14
A	paragraph [0093]; claims 1, 2, 7, 10, 12, 13; example 2 page 22, lines 15-16	1-12
A	----- WO 2014/104011 A1 (WINTECH POLYMER LTD [JP]) 3 July 2014 (2014-07-03)	1-14
A	----- JP H10 219026 A (NITTO BOSEKI CO LTD) 18 August 1998 (1998-08-18)	1-14



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

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/US2018/016970

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