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(54) Title: POLYMER IMPACT MODIFIER BLEND CONCENTRATE

(57) Abstract: Polymer impact modifier blend concentrates are provided. The impact modifier blend concentrates are useful to form objects that resist fracturing upon impact, in particular materials that require toughness at low temperatures, such as sports equipment and dual ovenable trays. The impact modifier blend concentrates are polymeric compositions that include a polyester and an ethylene copolymer blend.



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TITLE

POLYMER IMPACT MODIFIER BLEND CONCENTRATE

FIELD OF THE INVENTION

5 The present invention is directed to improving the impact resistance, and in particular the low temperature impact resistance, of certain polymers such as polyesters. Specifically, an impact modifier blend concentrate that can be dried with a bulk polyester is provided.

BACKGROUND OF THE INVENTION

10 Polyesters offer many advantageous properties, such as very low water absorption, particularly in comparison to polyamides such as nylons; good dimensional stability; good electrical properties; good resistance to chemical attack and high environmental stress; crack resistance, particularly in comparison to polycarbonates; good heat resistance and
15 heat aging resistance; low creep, even at elevated temperatures; good color stability; and excellent wear properties. Crystalline polyesters offer further advantages, including higher melting temperatures, uniform melting properties, and insolubility in organic solvents.

20 Accordingly, polyesters are the material of choice for many applications, including packaging, such as beverage containers and packaging for baked goods, frozen foods, and meats; engineering resins; monofilaments; fibers and nonwoven materials; films, including medical imaging and videotape; adhesives; and as a thick sheet for signage and medical packages, for example.

25 Many polyester applications require superior toughness, that is, the property of resisting fracturing upon impact. Improving the toughness of various polymer compositions is therefore a perennial goal in certain industries, such as the manufacture of sports equipment, dual ovenable trays, automotive parts, and the like. It is therefore known to use an
30 impact modifying additive to provide adequate toughness to polyester resins. Commonly used impact modifiers include styrene block copolymers, acrylic core/shell particles, and ethylene copolymers such as

Elvaloy® PTW and Elvaloy® AC, available from E. I. du Pont de Nemours and Company of Wilmington, DE (hereinafter "DuPont").

5 Improved toughness is also important to polyester recycling processes. It is well known that polyesters may decrease in molecular weight due to hydrolysis during melt processing. The decrease in molecular weight causes degradation of certain mechanical properties, for example toughness. The addition of an impact modifier to recycled polyester during the recycling re-extrusion process counteracts the observed loss in toughness, however.

10 Disadvantageously, though, many traditional impact modifiers melt at temperatures lower than those at which polyesters are dried prior to melting and extrusion (typically about 150°C). Thus, pellets of the impact modifier quickly fuse with the polyester pellets at higher temperatures, resulting in clumping in the drying apparatus. Consequently, the use of
15 the traditional impact modifiers requires additional resin handling systems to co-feed the hot dry polyester pellets and the impact modifier pellets at the throat of the sheet extruder, so that the hot polyester pellets are separated from the impact modifier pellets until both types of pellets are at the extruder feed screw. This specialized co-feed arrangement detracts
20 from the economic efficiency of the manufacturing process.

Another additive commonly used in crystalline polyester formulations is the crystallization nucleant. When the polyester is in the amorphous or glassy form, the crystallization nucleant reduces the temperature at which the amorphous polyester will crystallize.
25 Crystallization nucleants generally used in the art, such as polyethylene, waxes, and inorganic particles, including talc, silica (SiO₂), titania (TiO₂), and the like, are disadvantageously prone to create deposits in molds, however.

30 One use for polyesters that presents particular challenges is the fabrication of dual-ovenable trays. These trays, which are typically formed principally of crystalline polyethylene terephthalate (CPET), are used to freeze, transport, and store food items. The food items, still in the dual-

ovenable trays, are then heated in a microwave or conventional oven to a temperature that is safe for consumption. Thus, a dual-ovenable tray must resist shattering on impact over a broad range of temperatures, generally about -40°C to about 200°C . The materials in a dual-ovenable tray must be safe for direct contact with food, and they must maintain dimensional stability on heating. In addition, both the materials and the processes used to fabricate dual-ovenable trays must be economically favorable.

Injection molding and thermoforming are widely known methods for forming thermoplastic polyester articles. In injection molding, the polyester is heated above its melting point and injected under sufficient pressure to force the molten polyester to fill the mold cavity. The molten polyester is cooled in the mold until it is rigid enough to be removed. The injection molding method is generally not satisfactory, however, for the production of thin walled articles, such as dual-ovenable trays, due to flow lines and layering that may develop during the filling of the mold and that may lead to non-uniform properties, surface irregularities, and warping of the finished article. Moreover, very high filling pressures are typically required in the injection molding of thin walled articles, due to high melt viscosities.

Thermoforming is a particularly valuable technique for producing thin walled articles, such as dual ovenable food trays. In thermoforming, cast polyester sheet is preheated to a temperature sufficient to allow the deformation of the sheet. This temperature may be well below the melting point of the polyester. The sheet is then conformed to the contours of a mold by such means as vacuum assist, air pressure assist, or matched mold assist. The thermoformed article thus produced is normally heat treated in the mold in order to increase the crystallinity of the polyester.

In this connection, *Thermoforming* by J. L. Throne, Hansler (New York, 1986) provides a comprehensive overview of techniques including CPET thermoforming. *The Encyclopedia of Polymer Science and Engineering*, Wiley Interscience (New York, 1989), in Vol. 16 at page 823, describes the CPET thermoforming process from the glassy state. In addition, U.S. Pat. No. 6,576,309, issued to Dalgewicz, III et al., describes

thermoplastic compositions that are useful in food grade applications such as dual ovenable containers. Also, U.S. Pat. No. 5,747,127, issued to Prince, describes a thermoplastic resin composition that is suitable for use in thermoforming thin walled articles such as dual ovenable trays. Finally, GB Pat. No. 1 222 223, issued to Farbwerke Hoechst Aktiengesellschaft, describes methods to improve the toughness and crystallization rate of aromatic polyester based molding compositions. Each of these patents also includes a description of one or more of the "melt to mold" process or CPET thermoforming from the amorphous or glassy state.

In summary, there are many useful applications that benefit from the availability of tougher polymers, and in particular polymers with superior low temperature impact resistance. Consequently, it will be appreciated that an ongoing need exists for improved means of imparting impact resistance to polymeric materials.

SUMMARY OF THE INVENTION

A new impact modifier blend concentrate composition that can be dried with a bulk polyester has now been found in the present invention.

Accordingly, in a first aspect, the invention provides an impact modifier blend concentrate composition comprising a polyester and an ethylene copolymer blend.

In a second aspect, the invention provides a method of making an impact resistant polyester. The method includes providing an impact modifier blend concentrate of the invention, and blending the impact modifier blend concentrate with at least one bulk polyester. In some embodiments, the impact modifier blend concentrate is blended with the bulk polyester, and the resulting pellet blend is dried. The bulk polyesters may be new or recycled materials.

In accordance with another aspect of the invention, a method of improving the impact resistance of an article is provided. The method includes incorporating the impact modifier blend concentrate of the invention into the article.

In accordance with another aspect of the invention, an object comprising the impact modifier blend concentrate of the invention is provided.

5 Advantageously, the impact modifier blend concentrate of the invention provides a convenient and economical means to improve the impact resistance of polyesters and articles fabricated with polyesters, in particular their low temperature impact resistance. Also, the use of the impact modifier blend concentrate provides certain advantages in the process of making impact resistant polyesters by melt extrusion.

10 First, in general, no clumping results when the impact modifier blend concentrate is blended with one or more polyesters in the desiccant hopper of an extruder under typical polyester drying conditions. In addition, the impact modifier blend concentrate promotes crystallization of the impact resistant polyester due to the nucleating effect of the ionomer
15 compatibilizer.

DETAILED DESCRIPTION OF THE INVENTION

The definitions herein apply to the terms as used throughout this specification, unless otherwise limited in specific instances.

20 The terms "finite amount" and "finite value" refer to an amount that is not equal to zero.

The term "about" means that amounts, sizes, formulations, parameters, and other quantities and characteristics are not and need not be exact, but may be approximate and/or larger or smaller, as desired, reflecting tolerances, conversion factors, rounding off, measurement error
25 and the like, and other factors known to those of skill in the art. In general, an amount, size, formulation, parameter or other quantity or characteristic is "about" or "approximate" whether or not expressly stated to be such.

In one embodiment, the present invention provides an impact modifier blend concentrate composition comprising a polyester and an
30 ethylene copolymer blend.

Suitable polyesters for use in the present invention include, without limitation, poly(ethylene terephthalate) (PET); PET modified by

incorporating diacids other than terephthalic acid, such as isophthalic acid, or glycols other than ethylene glycol, such as 1,3-propanediol, diethylene glycol or cyclohexane dimethanol; copolymers containing terephthalic acid, cyclohexane dimethanol, 1,3-propanediol, diethylene glycol and other dibasic acids, such as isophthalic acid; poly(ethylene naphthalate); copolyesters thereof; combinations, e.g., blends, thereof; and derivatives thereof. Among the preferred polyesters are PET, blends thereof, and derivatives thereof.

The polyester may be recycled or virgin material. The term "recycled polyester", as used herein, refers to any polyester resin that has been previously extruded, for example into a sheet, or molded, for example by injection molding. Recycled polyester may be collected from any source, such as, for example, post consumer recycle streams, in-house operations at a manufacturing facility, and the like. Examples of recycled polyester include, without limitation, trim scrap, edge trim, chopped up polyester bottles and trays, and the like.

The preferred polyesters are also thermoplastic and crystallizable, having a glass transition temperature (T_g) of at least 50°C, and a melting point of at least 150°C. More preferred polyesters have a melting point of at least 200°C, and still more preferably at least 250°C. Typically, for packaging applications, the color of the crystallized polyester should be very white and opaque.

Preferred polyesters are also characterized by a molecular weight adequate to obtain desired flexural characteristics. In particular, the intrinsic viscosity (IV) is preferably at least about 0.5 as measured in a 1:1 weight:weight solution of methylene chloride and trifluoroacetic acid. Preferably, the intrinsic viscosity is at least about 0.65, and more preferably at least about 0.85 or about 0.95.

Methods of preparing polyesters are well known in the art. See, for example, the *Modern Plastics Encyclopedia*, McGraw Hill, (New York, 1994); and the *Wiley Encyclopedia of Packaging Technology*, 2d edition, A.L. Brody and K.S. Marsh, Eds., Wiley-Interscience (Hoboken, 1997).

Briefly, polyesters are generally synthesized from aromatic dicarboxylic acids, and preferably the lower alkyl esters thereof, such as the dimethyl ester of terephthalic acid. The aromatic dicarboxylic acid or its ester or anhydride may be esterified or trans-esterified and polycondensed with a saturated diol such as ethylene glycol. Typically used saturated diols include saturated aliphatic or cycloaliphatic diols, preferably the lower alkane diols, such as ethylene glycol. Mixtures of aliphatic carboxylic acids and saturated diols may also be used, provided that the above-described physical properties, i.e., melting point and T_g ranges, are generally obtained.

Several preferred grades of polyester for use in the present invention are commercially available. These include high molecular weight PET homopolymers with intrinsic viscosities of 0.85 or higher, such as Futura™ NP 107, available from Futura Polymers Ltd. of Chennai, Tamil Nadu, India, and Crystar® 5005, available from DuPont.

The amount of polyester in the impact modifier blend concentrate preferably ranges up to about 75 wt%, more preferably from about 40 to about 75 wt%, and still more preferably from about 55 to about 70 wt%, based on the total weight of the impact modifier blend concentrate.

The impact modifier blend concentrate of the invention also comprises an ethylene copolymer blend. Preferably, the amount of ethylene copolymer blend in the impact modifier blend concentrate ranges from greater than about 25 wt%, more preferably from about 25 to about 60 wt%, and still more preferably from about 30 to about 45 wt%, based on the total weight of the impact modifier blend concentrate.

It is generally preferable to maximize the loading of the ethylene copolymer blend in the impact modifier blend concentrate while maintaining the polyester as the continuous phase. Maximizing the loading of ethylene copolymer blend minimizes the amount of concentrate required to achieve the desired toughness in the finished polyester or article. Maintaining the polyester as the continuous phase ensures that the impact modifier blend concentrate does not agglomerate during the

high temperature processing steps, such as drying the concentrate with a bulk polyester prior to extrusion. This is believed to be the result of the morphology of the impact modifier blend concentrate blend, specifically, the lower melting ethylene copolymer phase being dispersed in the continuous polyester phase.

In this connection, it has now surprisingly been found that the ethylene copolymer phase of the impact modifier blend concentrate need not have a small particle size or even particulate morphology in order to provide a polyester blend whose impact modifier has the preferred particulate morphology. Accordingly, it is preferred but not necessary that the ethylene copolymer phase have particulate morphology.

The ethylene copolymer blend comprises a copolymer of ethylene and an alkyl acrylate, a compatibilizer comprising an ionomer, and, optionally, a crosslinking agent. Any known ethylene alkyl acrylate copolymer, whether prepared by an autoclave or tubular process, is suitable for use in the ethylene copolymer blend.

The alkyl group of the acrylate ester may be a straight chain or branched group having from one to ten carbon atoms. Preferred alkyl groups include methyl, ethyl, and butyl groups, and combinations of two or more of these groups are also preferred. Combinations including methyl groups are more preferred.

Still more preferably, the alkyl acrylate consists essentially of methyl acrylate. U.S. Patent Appln. Pub. No. 2004/0006180 by Trouilhet et al. describes various copolymers of ethylene and methyl acrylate, and uses thereof. In the present invention, the methyl acrylate comonomer content is preferably greater than about 10 wt%, based on the weight of the ethylene methyl acrylate (EMA) copolymer. More preferably, the methyl acrylate comonomer content is greater than about 15 wt%, and, still more preferably, the methyl acrylate comonomer content is greater than about 25 wt%, based on the weight of the EMA copolymer. For uses that require direct contact with food, however, such as the dual ovenable

tray, applicable food packaging guidelines may limit the total fraction of methyl acrylate in the EMA copolymer to less than 25 wt%.

The melt flow index (MFI or I2) of the ethylene alkyl acrylate copolymers, measured according to ASTM D1238 at conditions of 190°C and under a weight of 2.16 kg, may range up to about 10 g/10 min; however, as those of skill in the art are aware, an MFI of the EMA copolymer that corresponds to the viscosity of the polyester will be preferred. It is well known that a dispersed phase is more easily developed in a continuous phase when the viscosity ratio of the dispersed and continuous phases is near unity. See Dealy J.M., Wissburn K.F. *Melt Rheology and its Role in Plastic Processing*, Van Nostrand Reinhold (New York, 1990). Accordingly, the MFI of the ethylene alkyl acrylate copolymer is preferably less than about 4.0 g/10 min, 2.0 g/10 min, 1.0 g/10 min, 0.5 g/10 min, or 0.3 g/10 min, with increasing preference in the order given.

In general, tubular grades of ethylene acrylate copolymers have higher melting temperatures at the same weight fraction of acrylate, compared to ethylene acrylate copolymers prepared by the autoclave process. As noted above, higher melting ethylene acrylate copolymers are preferred. Therefore, ethylene alkyl acrylate copolymers that are prepared in a tubular reactor are also preferred.

Several preferred ethylene alkyl acrylate copolymers for use in the present invention are commercially available. These include Elvaloy® AC tubular grade EMA copolymers, available from DuPont. Autoclave produced grades of ethylene acrylate copolymers are sold by the ExxonMobil Chemical Company of Houston, TX, under the Optema™ brand name.

Methods of preparing EMA copolymers are well known in the art. See, for example, the *Modern Plastics Encyclopedia* and the *Wiley Encyclopedia of Packaging Technology*.

The amount of ethylene alkyl acrylate copolymer in the ethylene copolymer blend ranges from about 20 to about 60 wt%, more preferably

from about 20 to about 50 wt%, and still more preferably from about 20 to about 36 wt%, based on the total weight of the impact modifier blend concentrate.

The ethylene copolymer blend also includes a compatibilizer that comprises at least one ionomer. The term "ionomer" as used herein, alone or in derivative form, such as "ionomeric", e.g., refers to an acid copolymer that has been at least partially neutralized with a neutralizing agent such as an inorganic base, and that comprises carboxylate salts with counterions derived from the neutralizing agent.

Suitable ethylene acid copolymers that are precursors for ionomers may be selected from a wide variety of polymers that incorporate ethylene as the basic monomeric unit, and that incorporate at least one comonomer selected from vinyl acetate, acrylic acid, and methacrylic acid. Additional comonomers may also form part of the polymeric structure, and these may be selected from esters such as alkyl acrylates and the like. The alkyl acrylates may be selected from methyl acrylate, ethyl acrylate, n-butyl acrylate, and iso-butyl acrylate. The use of copolymers including acrylate esters of two or more alkyl groups is specifically contemplated.

A common feature of the non-ethylene monomers is the presence of a carboxylic acid moiety or a precursor to such moiety. Additional acid containing comonomers may be selected from ethylenically unsaturated diacids, such as maleic and fumaric acids and their anhydrides. The acid containing comonomer may comprise up to fifty percent of the copolymer. Methods of preparing suitable ethylene acid copolymers are disclosed in, for example, U.S. Pat. Nos. 3,264,272; 3,404,134; 3,355,319 and 4,321,337.

Preferred thermoplastic ethylene acid copolymers are selected from copolymers of ethylene and about 2 to about 50%, more preferably about 5 to about 25%, by weight of acrylic or methacrylic acid. The melt index of the copolymer is preferably about 0.1 to 30, more preferably about 0.5 to 20 dg/min, according to ASTM standard D1238.

Ionomers suitable for use in the present invention may comprise any feasible cation or combination of cations. Preferred cations may be monovalent or divalent. When the cations are metal cations, they are preferably selected from among alkali metals (Group 1), alkaline earth
5 metals (Group 2), transition metals (Groups 3 through 12), lanthanides, and actinides. Preferred cations include lithium, sodium, potassium, magnesium, calcium, barium, copper, zinc, ammonium, or a combination of two or more of these cations. Sodium, potassium, and zinc are more preferred cations for use in the present invention. The sodium cation is
10 particularly preferred. The extent of neutralization of the acid groups by the cation is greater than 36 mol% and preferably greater than 50 mol%.

Also preferably, the molecular weight of the ionomer is sufficiently high so that the viscosity of the ionomer (that is, the viscosity after neutralization) is approximately equal to the viscosity of the polyester. For
15 the same reason, the ionomer preferably has a melt flow index (MFI) measured at 190°C and 2.16 kg wt that is less than 2, and more preferably the MFI is less than 1.

Several preferred ionomers and acid precursors to preferred ionomers can be obtained commercially from DuPont. For example,
20 BYNEL® and NUCREL® are ethylene acrylic or methacrylic acid copolymers optionally containing an additional softening monomer such as an alkyl acrylate; and the SURLYN® line includes ionomers of certain of the above ethylene acid copolymers or terpolymers. In addition, Himilan™ ionomers are available from Mitsui-DuPont Polychemicals Co., Ltd., of
25 Tokyo, Japan.

Methods of preparing ionomers are described in U.S. Pat. No. 3,344,014, issued to Rees, for example.

The amount of ionomer in the ethylene copolymer blend is preferably a finite amount of from about 0 to about 30 wt%, more
30 preferably from about 2 to about 20 wt%, and still more preferably from about 5 to about 10 wt%, based on the total weight of the impact modifier blend concentrate.

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The ionomer, especially a sodium or potassium containing ionomer, is also believed to function as a crystallization nucleant in the impact modifier blend concentrate. See, e.g., U.S. Pat. No. 4,753,980, issued to Deyrup. That is, because of the ionomer, the blend of impact modifier
5 blend concentrate and bulk polyester typically crystallizes more rapidly from the melt or from the glassy state. Moreover, certain blends according to the invention have been measured to crystallize from the glassy or amorphous state at temperatures about 10°C lower than unmodified polyesters.

10 The impact modifier blend concentrate of the invention may optionally include one or more crosslinking agents. Any agent known to crosslink an ethylene alkyl acrylate copolymer is suitable for use in the present invention. Preferably, the crosslinking agent is an organic peroxide compound or an organic compound with two or more double
15 bonds. Examples of preferred crosslinking agents include, without limitation, diethylene glycol diacrylate, diethylene glycol dimethacrylate, ethylene glycol dimethacrylate, N,N'-m-phenylene dimaleimide, polyethylene glycol dimethacrylate, triallyl cyanurate, triallylisocyanurate, trimethylolpropane trimethacrylate, tetraallyloxyethane, tetramethylene
20 diacrylate, 2,5-dimethyl-2,5-di-(*t*-butylperoxy) hexyne-3, *t*-butyl peroxybenzoate, 2,5-dimethyl-2,5-di-(*t*-butylperoxy)-2,5-dimethylhexane, dicumyl peroxide, α,α -bis(*t*-butylperoxy)-2,5-dimethylhexane, and the like.

Particularly preferred crosslinking agents include diethylene glycol diacrylate; diethylene glycol dimethacrylate; ethylene glycol
25 dimethacrylate; 2,5-dimethyl-2,5-di-(*t*-butylperoxy) hexyne-3; and 2,5-dimethyl-2,5-di-(*t*-butylperoxy)hexane.

In certain preferred embodiments, the crosslinking agent includes both an organic peroxide compound and an organic compound with two or more double bonds. Of note are ethylene copolymer blends including both
30 diethylene glycol dimethacrylate and 2,5-dimethyl-2,5-di-(*t*-butylperoxy) hexane.

Several preferred crosslinking agents for use in the present invention are commercially available. These include Sartomer™ 231 (diethylene glycol dimethacrylate), available from Sartomer Company, Inc., of Exton, PA, and Luperox™ 101 (2,5-dimethyl-2,5-di(tert-butylperoxy)hexane), available from Atofina Chemicals, Inc., of Philadelphia, PA.

The crosslinking agent(s), when used, are preferably present in a finite amount of up to about 2%, more preferably a finite amount up to about 1%, and still more preferably in a finite amount up to about 0.4 wt%, based on the total weight of the impact modifier blend concentrate.

Preferably, the impact modifier blend concentrate also includes a stabilizer, such as, e.g., pentaerythritol tetrakis(3-(3,5-di-tert-butyl-1,4-hydroxyphenyl)propionate, which is available under the trade name Irganox™ 1010 from Ciba Specialty Chemicals of Basel, Switzerland. The stabilizer may be present at a level of from 0.0001 to about 5 wt%, about 0.001 to about 4 wt%, about 0.01 to about 3 wt%, or about 0.1 to 2 wt%. The stabilizer is preferably present at a level of about 1% based on the total weight of the impact modifier blend concentrate.

The impact modifier blend concentrate may also include such additives as are conventional in polymer compositions, for example, antioxidants, UV stabilizers, flame retardants, plasticizers, pigments, fillers, reinforcements, processing aids, and the like. Suitable levels of these additives and methods of incorporating these additives into polymer compositions are known to those of skill in the art. See, e.g., *Modern Plastics Encyclopedia*.

The impact modifier blend concentrate composition of the invention may be made by blending the individual components by any suitable means known in the art. For example, the individual materials can be mixed with each other in molten form, such as by melt blending in an extruder. Alternatively, the individual materials can be blended with each other in a high shear mixing device, such as a two-roll mill or a Banbury mixer.

In some preferred embodiments, the ethylene alkyl acrylate copolymer is crosslinked *in situ* in an extruder. Crosslinking increases the molecular weight and thereby decreases the MFI of the ethylene alkyl acrylate. Preferred crosslinking agents and preferred levels of crosslinking agents are as set forth above. Preferred conditions for the *in situ* crosslinking include polymer melt temperatures not exceeding 265°C and more preferably not exceeding 260°C. Higher melt temperatures tend to promote chain scission of the ethylene acrylate copolymer rather than the intended crosslinking reaction.

In one preferred method, an impact modifier blend concentrate is made by first extruding the ethylene alkyl acrylate copolymer with a small amount of crosslinking agent in a twin-screw extruder to reduce the melt index, preferably to about 0.3MI. In a second extrusion step, dried polyester resin is co-fed to the throat of a twin-screw extruder along with an ionomer, the crosslinked ethylene alkyl acrylate copolymer, and an antioxidant. These raw materials are processed in the extruder using high screw speed to prepare the impact modifier blend concentrate.

Also provided by the invention are methods of making impact resistant polyesters. In these methods, an impact modifier blend concentrate of the invention is provided, and the impact modifier blend concentrate is combined with at least one bulk polyester. The resulting polymer blend will have an impact resistance that is greater than that of the unmodified bulk polyester.

The preferred bulk polyesters are those described above as suitable for use in the impact modifier blend concentrate. The bulk polyester may be the same as or different from the polyester in the impact modifier blend concentrate. The impact modifier blend concentrate may be combined with a combination of two or more bulk polyesters. Moreover, the bulk polyester may include recycled polyester.

Preferably, the impact modifier blend concentrate and the bulk polyester are dried prior to being combined. The drying may be carried out in a desiccant hopper dryer. In certain preferred embodiments, the

FIG. 1 is a schematic diagram of a melt mixing device.

impact modifier blend concentrate and bulk polyester can be blended as pellets prior to drying or co-fed into the melt mixing device after drying.

The dried impact modifier blend concentrate and the dried bulk polyester may be combined by any means known in the art. Preferably, they are combined by a melt mixing process such as, e.g., single screw extrusion, twin-screw extrusion or Banbury batch mixing. Single screw extrusion is a preferred method of combining the impact modifier blend concentrate and the bulk polyester.

Preferably, the impact resistant polyester includes at least 10 wt% of the impact modifier blend concentrate, based on the total weight of the impact resistant composition. More preferably, the impact resistant polyester includes about 20 wt% to about 30 wt% of the impact modifier blend concentrate, based on the total weight of the impact resistant composition. Also preferably, the impact modifier in the impact resistant polyester has a particulate morphology. More preferably, the size of the dispersed impact modifier particles is less than about 2 μm , and still more preferably less than or equal to about 1 μm .

Also provided by the present invention are articles comprising the impact modifier blend concentrate of the invention. Such articles may be made according to methods that are well known in the art. For example, after mixing the components of an impact resistant composition, including the impact modifier blend concentrate, in an extruder, the impact resistant composition extruded through a flat die and cast onto a forming drum to form a tray. Alternatively, after mixing the components of an impact resistant composition, including the impact modifier blend concentrate, in an extruder, the impact resistant composition is extruded through a flat die and cast onto a chilled roll to rapidly quench the sheet to prepare amorphous impact resistant sheet that can subsequently be thermoformed to make various articles. Thermoforming is the preferred method of forming articles according to the invention.

The impact resistant composition may be used alone, or it may be used in combination with other materials, such as reinforcing fillers or

blowing agents, to form impact resistant articles. Replacing even a portion of material in a conventional article with an impact resistant composition is expected to enhance the impact resistance of the article. Preferably, the impact resistant composition includes at least 10 wt% of the impact
5 modifier blend concentrate, based on the total weight of the impact resistant composition. More preferably, the impact resistant composition includes about 20 wt% to about 30 wt% of the impact modifier blend concentrate, based on the total weight of the impact resistant composition. Also preferably, the impact modifier in the impact resistant article has a
10 particulate morphology. More preferably, the size of the dispersed impact modifier particles is less than about 2 μm , and still more preferably less than or equal to about 1 μm .

Examples of suitable articles whose impact resistance may be increased by at least partial fabrication from an impact resistant
15 composition comprising the impact modifier blend concentrate of the invention include, without limitation, sports equipment, dual ovenable trays, automotive parts, shipping containers, construction materials, furniture, cabinetry, decorative objects, cups, bowls, bakery trays and lids, and the like. Dual ovenable trays are preferred articles according to the
20 invention.

The present invention also provides a method of improving the impact resistance of an article, and an article obtainable by this method. In this method, an impact modifier blend concentrate of the invention is combined with a bulk polyester. The resulting blend is formed into an
25 article by any suitable method known in the art, such as, for example, those described above with respect to other articles according to the invention. The resulting article has an impact resistance that is greater than that of an article fabricated from an unmodified bulk polyester. Replacing even a portion of the polyester in a conventional article with an
30 impact resistant composition of the invention is expected to enhance the impact resistance of the article.

The suitable and preferred bulk polyesters are those described above for use in the impact modifier blend concentrate. The suitable and preferred means and conditions of combining the impact modifier blend concentrate and the bulk polyester are as set forth above with respect to the methods of making impact resistant polyesters. The suitable and preferred articles, the suitable and preferred means of fabricating the articles, and the suitable and preferred levels of impact modifier blend concentrate in the articles, are as set forth above with respect to articles comprising the impact modifier blend concentrate of the invention.

The following examples are provided to describe the invention in further detail. These examples, which set forth a preferred mode presently contemplated for carrying out the invention, are intended to illustrate and not to limit the invention.

EXAMPLES

Comparative Examples

Using the conditions and equipment outlined in Appendix 4, below, a sheet (nominally 0.8mm thick) was cast from NP 107, a PET homopolymer with 0.95IV available from Futura Polymers. This sheet is Comparative Example 1. Using the same conditions and equipment, a sheet (nominally 0.8mm thick) was cast from a blend comprising 90 wt% dried NP 107 and 10 wt% Elvaloy® CT-2. Unlike the conditions described in Appendix 4, the Elvaloy® CT-2 was not dried prior to extrusion with the PET. Before blending the Elvaloy® CT-2 with the PET it was necessary to allow the PET to cool to less than the melting point of the ethylene copolymer to prevent clumping during pellet blending or clumping in the extruder hopper. Elvaloy® CT-2 is a pre-extruded blend of Elvaloy® 1224AC and Surlyn® 8920 in a 5:1 wt:wt ratio. This sheet is Comparative Example 2.

The sheet samples were annealed at 140°C for three hours to promote crystallization, then conditioned overnight at -40°C prior to dart drop testing according to ASTM D5628 geometry FA. The average F50/25µm normalized failure weight for sheets of Comparative Example 1

It is to be understood that the present invention is not limited to the specific details of the above description.

was 1 gm/25 μ m at -40°C . The average F50/25 μ m normalized failure weight for sheets of Comparative Example 2 was 15 g/25 μ m.

5 Scanning electron microscopy (SEM) analysis of the sheet samples reveal that the unmodified polyester of Comparative Example 1 is a single phase, and that the ethylene copolymers in Comparative Example 2 form a relatively uniformly dispersed particulate phase with particle sizes ranging from less than 0.5 μ m to about 2 μ m, and with occasional particles as large as about 4 μ m.

10 These data demonstrate that polyester, without an impact modifier, is very brittle. Moreover, the impact resistance of polyester can be significantly improved by the addition of 10 wt% ethylene copolymer.

Example Set 1

15 Blends of a polyester and various ethylene copolymers and ionomers were prepared as described below in Appendix 1. The compositions of the blends are set forth in Table 1.

Table 1

Impact Modifier Blend Concentrate Compositions

Example No.	Poly-ester	Terpolymer	Ionomer	EMA 1	EMA 2	AO	Processing Notes
1	84%	0%	0%	15%	0%	1%	poor
2	84%	0%	0%	0%	15%	1%	poor
3	80%	4%	0%	15%	0%	1%	poor
4	76%	4%	4%	0%	15%	1%	acceptable
5	72%	4%	0%	0%	23%	1%	acceptable
6	80%	4%	0%	15%	0%	1%	poor
7	72%	0%	4%	0%	23%	1%	acceptable
8	68%	4%	4%	23%	0%	1%	poor
9	72%	0%	4%	0%	23%	1%	acceptable
10	68%	4%	4%	0%	23%	1%	acceptable
11	80%	0%	4%	15%	0%	1%	acceptable
12	76%	4%	4%	0%	15%	1%	acceptable
13	76%	0%	0%	23%	0%	1%	poor
14	76%	2%	2%	9.5%	9.5%	1%	acceptable
15	72%	4%	0%	23%	0%	1%	poor
16	80%	4%	0%	0%	15%	1%	acceptable
17	80%	0%	4%	0%	15%	1%	acceptable
18	72%	0%	4%	23%	0%	1%	poor
19	89.7%	1.7%	0%	8.3%	0%	0.3%	acceptable

- 5 Notes for Table 1: The polyester is NP 107, a PET homopolymer whose intrinsic viscosity is nominally 0.95, supplied by Futura Polymers. The terpolymer is Bynel® 2002 with MI = 10. The ionomer is Surlyn® 8920, 0.9 MI, sodium counterion. EMA 1 is Elvaloy® 1224AC, an ethylene methyl acrylate copolymer (24 wt% MA) with MI = 2. EMA 2 is Elvaloy®
- 10 1125AC, an ethylene methyl acrylate copolymer (25 wt% MA) with MI = 0.4. The antioxidant (AO) is Irganox™ 1010.

The Processing Notes in Table 1 refer to the presence or absence of surging or strand breaks at the die. Strand breaks and surging are unacceptable because they lead to material losses and non-uniform pellet cutting. It can be concluded from the data in Table 1 that grades of EMA with lower melt indices (MI) produce more uniform pellets and minimize strand breaks. Furthermore, even at the lowest melt index (0.4 MI), an ionomer or terpolymer compatibilizer is required to prevent strand breaks or surging.

Example Set 2

The melt viscosity of the Futura NP107 polyester and the EMA copolymers were measured at 280°C according to ASTM D3835 using a GALAXY V Model 8052 melt rheometer, available from Kayeness, Inc., of Morgantown, PA. The capillary die geometry was 1.52mm in diameter and 23mm in length. The NP 107 resin was dried overnight at 125°C in a stream of dry nitrogen gas prior to measuring its viscosity. Upon removing the sample from the drier, the sample was immediately charged to the rheometer. There the sample was allowed to melt in the rheometer barrel for five minutes under a preload force of 1100N before initiating a series of ram extrusion rates that developed shear rates from 10.3 to 1006.7(1/s). In addition to the NP 107 resin, samples of EMA were also processed through the rheometer, without drying. The melt viscosity values the NP 107 resin and the EMA copolymers are included in Table 2.

Table 2

Viscosity (PaS) at 280°C

	1006.7	501.6	250.8	51.5	20.6	10.3
Futura NP 107	503	726	964	1431	1659	1738
EMA 1	78	107	148	271	342	429
EMA 2	123	174	246	516	783	956
EMA 3	83	140	170	321	463	609

5 Note: The column headings are the shear rates (1/s).

From the data in Table 3, it is apparent that reducing the melt index of the EMA from 2 to 0.8 to 0.4 brings the viscosity of EMA closer to that of the PET. As is noted above, a viscosity ratio close to unity favors the formation of a dispersed phase.

10 Example Set 3

A second series of impact modifier blend concentrates were prepared on a 43mm 48/1 L/D twin-screw extruder. The melt exiting the die of the extruder was cut into pellets using a Gala Industries underwater pelletizer. The polyester was dried overnight at 130°C in a desiccant
 15 hopper drier prior to extrusion. The compositions of these impact modifier blend concentrates are listed in Table 3, below. The extrusion conditions are set forth in detail in Appendix 3, below.

To increase the viscosity and reduce the melt index of the ethylene copolymer, a blend of Elvaloy® AC1224 and an organic peroxide
 20 (Luperox™ 101) was also extruded using the 43mm twin screw extruder. It was found the addition of nominally 0.35wt% Luperox to Elvaloy® AC1224 reduced the MI of the EMA from 2 g/10min to 0.8 g/10min. The resulting crosslinked EMA is designated "EMA 3" in Table 3 below. The process conditions used to produce EMA 3 are set forth in detail in
 25 Appendix 2, below.

Table 3
Impact Modifier Blend Concentrate Compositions

Example No.	Polyester	EMA 2	EMA 3	Ionomer	Terpolymer	AO	Ratio of EMA/Compa
20	61%		32%	6.4%		0.7%	5.0
21	55%		37%	7.4%		0.9%	5.0
22	50%		41%	8.2%		1.0%	5.0
23	67%		27%	5.4%		0.6%	5.0
24	66%	24%		4.2%	4.2%	1.1%	2.9
25	52%	34%		6.0%	6.0%	1.5%	2.9
26	40%	43%		7.5%	7.5%	1.9%	2.9
27	67%	27%		5.4%		1.0%	5.0
28	52%	39%		7.8%		1.4%	5.0
29	68%*	23%		4.0%	4.0%	1.0%	2.9

Notes for Table 3: The polyester is Futura NP 107, except for Example 29, in which the polyester is Crystar® 5005, available from DuPont, a PET homopolymer with IV = 0.85. The ionomer, terpolymer, EMA 2, and antioxidant (AO) are as defined above. In the last column of Table 3 the ratio of EMA to Compatibilizer is calculated. In the case of example 24, the EMA fraction of 24wt% while the sum of Ionomer and Terpolymer (compatibilizers) was 8.4%. The fraction EMA divided by the sum of compatibilizers equals 2.9.

All of the example compositions in Table 3 were extruded without pressure surges and produced pellets of uniform dimensions.

To verify that the samples would not lump together under polyester drying conditions, samples of the impact modifier blend concentrates (27g) were placed in a small aluminum pan and heated in air at 150°C for two hours. Because the ethylene copolymers all have melting points of 90°C or less, the pellets would be expected to fuse together at this temperature if the polyester were not the continuous phase, or at least a co-continuous

phase. In all cases, however, it was possible to break the lumps into individual pellets after the heating cycle. In the case of Examples 22 and 26, it was more difficult to break up the pellets compared to the other examples.

5 Example Set 4

Toughened polyester sheets were prepared from selected impact modifier blend concentrates. The proportions of concentrate and polyester were selected to produce sheet samples that contained 10 wt% of ethylene copolymer. Typical operating conditions for the sheet casting
10 process are provided in Appendix 4. The sheet compositions are set forth in Table 4, below.

Table 4
Sheet Compositions

Sheet example	IMBC* Example No.	Amount of IMBC, wt%	Amount of Polyester, Wt%
1	20	25.5	74.5
2	22	20.0	80.0
3	24	29.5	70.5
4	25	20.9	79.1
5	26	16.7	83.3
6	27	30.3	69.7
7	28	20.8	79.2

15 * "IMBC" is an abbreviation of "impact modifier blend concentrate."

To estimate the size of the dispersed phase particle size in the sheet, sheet samples were notched in the machine direction with a razor knife at room temperature. The sheet was subsequently annealed at
20 150°C for three hours to crystallize the polyester fraction of the sheet. The notched and annealed sheet samples were then conditioned at -40°C

overnight. The sheet samples were removed from the freezer and flexed along the notched line to fracture the sheet into two pieces. One of these pieces was incubated in xylene for three hours at 50°C to extract the ethylene copolymer phase from the polyester matrix. The extracted fracture surfaces were examined by scanning electron microscopy (SEM) at 500 and 5000 times magnification. This procedure is outlined in the Proceedings of the Antec conference of the Society of Plastics Engineers in a paper titled IMPACT MODIFICATION OF POLY(ETHYLENE TEREPHTHALATE) by N. Chapleau and M. A. Huneault (Antec 2001, Abstract #702).

The SEM images at 5000X magnification of sheet samples 1 to 7 reveal that sheet samples 1 and 2 have round or slightly oval holes left behind by the extraction of the dispersed phase. The size of the holes indicates an average particle diameter of 1µm, with a few particles as large as 2µ. In sheet samples 6 and 7, the holes are round and less than 1µm in diameter. In sheet samples 3, 4 and 5, the holes are elongated. The image from sheet sample 3 shows holes that are slightly elongated, generally small ovals uniformly less than 1µm in longest axis. The image from sheet sample 4 shows holes where the elongation of the long axis is more pronounced. The average long axis length is over 1µm and there is evidence of larger domains of ethylene copolymer up to 5µ in diameter. The holes in sheet sample 5 are very elongated, and some elongated particles are over 2µ in length. There is also evidence of large (5µm diameter) holes in the sample. Table 5 includes a summary of the SEM analysis.

Sheet samples 1 to 7 were cut into 10cm by 10cm squares and annealed for three hours at 140°C to crystallize the polyester component of the sheet. The squares were then conditioned at -40°C overnight prior to dart drop testing according to ASTM D5628 geometry FA. From each sample sheet, approximately 25 squares were tested to arrive at an estimate of how much dart weight would be required for 50% of the sheet samples to fail when impacted at -40°C from a drop height of 66cm.

Because sheet thickness influences the outcome, we have reported a normalized F50 in grams/25 μ m. Table 5 summarizes the results of the dart drop testing of the annealed sheet samples.

5

Table 5
SEM Analysis and
Results of ASTM D5628 Geometry FA

	Impact	SEM sheet	Dispersed particle
sheet	F50 (g/25 μ m)	average (μ m)	Shape
1	23	1	Round
2	28	1	Round
3	21	1	Oval
4	20	1.5	Oval
5	13	1.5-2	Oval
6	33	<1	Round
7	24	<1	Round

10

The results in Table 5 demonstrate that small particle size and a tendency towards spherical morphology correlate with superior toughness in polyester blends including the impact modifier blend concentrates of the invention.

APPENDICES

Appendix 1

15

20

The polyester resins were dried overnight at 130°C in a desiccant hopper dryer system. The dryer was built by the Conair Group, Inc. of Pittsburgh, PA, and the hoppers were built by Thoreson McCosh, Inc., of Troy, MI. The temperature set point of the dryer was lowered in the morning to 65°C, and after one hour the ethylene copolymer and compatibilizers were added to separate compartments of the dryer and dried for at least two hours. After drying, weighed amounts of

compatibilizer, ethylene copolymer, polyester, and antioxidant were blended by shaking in a bag for 30 seconds. The resulting 3.5kg batches of pellet blends were fed to the throat (Barrel 1) of a 25mm 38/1 L/D ZSK-25 World Lab twin-screw extruder manufactured by Krupp Werner & Pfleiderer Lebensmitteltechnik GmbH of Dinkelsbuehl, Germany.

The melt blend exiting the extruder through a two-hole die was quenched in water. The quenched strand was cut into pellets using a pelletizer available from C.F. Scheer of Stuttgart, Germany. Typical operating conditions (actual conditions for Example 4) are shown in Table A1.

Table A1
Operating Conditions During Extrusion Compounding

	Temperature Set Point, °C	Actual Temperature, °C
Temperature Control Zone 1 (Barrel Zones 2 and 3)	280	280
Temperature Control Zone 2 (Barrel Zones 4 and 5)	280	280
Temperature Control Zone 3 (Barrel Zones 6 and 7)	270	270
Temperature Control Zone 4 (Barrel Zones 8 and 9)	260	262
Temperature Control Zone 5 (Die)	260	260
Screw Speed (rpm)		350
Torque (%)		39
Die Pressure (kPa)		883
Melt Temperature (°C)		284
Total Throughput (g/min)		160
Shear Cutter Set-point		16
Vacuum Zone 4 (mmHg)		29

5 Appendix 2

10 The Elvaloy® 1224AC (EMA 1) and the organic peroxide, 2,5-dimethyl-2,5-Di-(t-butylperoxy)hexane, were fed into the throat (Barrel 1) of the extruder. Because the peroxide is a liquid at room temperature, it was first coated onto the surface of a portion of the EMA using a drum blender, resulting in a concentrate of peroxide on EMA with a nominal peroxide loading of 1.5wt%. Two K-Tron loss in weight feeders, available from the K-Tron Feeder Group of Pitman, NJ, were used to meter the

peroxide concentrate and EMA 1 into the throat of the extruder. The peroxide concentrate was fed at 326g/min and EMA 1 was fed at 1091g/min.

5 Extruder operating conditions for the preparation of EMA 3 are set forth in detail in Table A2.

Table A2
Extruder Operating Conditions

	Temperature Set Point, °C	Actual Temperature, °C
Barrel 2	120	119
Barrel 3	150	150
Barrel 4	200	199
Barrel 5	210	208
Barrel 6	210	210
Barrel 7	210	209
Barrel 8	210	225
Barrel 9	210	209
Barrel 10	210	209
Barrel 11	210	210
Barrel 12	210	209
Die	210	211
Melt Temp at Gala		240
Die Pressure at Gala (kPa)		5330
Screw Speed (rpm)		350
Extruder Torque (amps)		30
Torque (% of maximum)		60
Melt Temperature Barrel 7 (°C)		242
Total Throughput (g/min)		1409
Gala Cutter Open Holes		11
Gala Speed (rpm)		13468
Vacuum Barrel 11 (mmHg)		27

Appendix 3

The polyester resins were dried overnight at 130°C in a desiccant hopper dryer system using the equipment described above at Appendix 1.

5 The compatibilizer and the ethylene copolymer were not dried prior to extrusion. In this case the compatibilizer or blend of compatibilizers was preblended with the antioxidant in a large drum blender, typically in 35 kg batches. The resulting pellet blend was fed into Barrel 1 of the twin-screw extruder using a K-Tron loss in weight feeder. A second loss in weight

10 feeder metered the dried polyester resin, also into Barrel 1. Typical operating conditions (actual conditions for fabrication of Example 28) are shown in Table A3.

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Table A3
Extruder Operating Conditions

	Temperature Set Point, °C	Actual Temperature, °C
Barrel 2	250	243
Barrel 3	250	249
Barrel 4	280	281
Barrel 5	280	280
Barrel 6	280	274
Barrel 7	280	274
Barrel 8	280	317
Barrel 9	280	311
Barrel 10	265	264
Barrel 11	265	266
Barrel 12	265	266
Die	285	282
Melt Temp at Gala		302
Feeder (0.95V PET) g/min		420
Feeder (Blend of Ethylene Copolymers and AO)		388
Die Pressure at Gala (kPa)		4275
Screw Speed (rpm)		350
Extruder Torque (amps)		27
Torque (% of maximum)		48
Melt Temperature Barrel 7 (°C)		315
Total Throughput (g/min)		806
Gala Cutter Open Holes (diameter of hole mm)		11 (3.8)
Gala Speed (rpm)		14737
Vacuum Barrel 11 (mmHg)		27.4

Appendix 4

The polyester resin and example concentrates were dried separately overnight at 130°C in a desiccant hopper dryer system using the equipment described above at appendix 1. The polyester and example concentrate were removed from the drier and blended by hand in the proportions outlined in Table 4 to make up a 3500g batch of pellet blend that was immediately charged to the throat (Barrel 1) of the single screw extruder through a nitrogen swept hopper. The extruder was a 31.75mm diameter 30/1 L/D model fitted with a 3/1 compression ratio, single flighted screw with 5 L/D of a melt mixing section. The extruder die was a 203mm wide coat hanger type flat film die with a 0.8mm die gap. The extruder was built by Wayne Machine of Totowa, NJ. The molten polymer film exiting from the die drawn down to nominally 0.7mm thick as it was cast onto a 203mm wide by 203mm diameter double shell spiral baffle casting roll fitted with controlled temperature cooling water. The casting roll and 203mm wide die were built by Killion Extruders – Davis Standard of Cedar Grove, NJ. Typical operating conditions for sheet extrusion (actual conditions for fabrication of Sheet 7) are shown in Table A4.

Table A4
Extruder Operating Conditions

	Temperature Set Point, °C	Actual Temperature, °C
Barrel Zone 1	295	295
Barrel Zone 2	295	295
Barrel Zone 3	280	279
Barrel Zone 4	280	280
Filter Flange	280	280
Adapter	280	280
End Plates on Die	280	281
Die Face	290	291
Filter Melt		276
Adapter Melt		290
Filter Pressure (kPa)		7171
Adapter Pressure (kPa)		3930
Screw R.P.M.		100
Screw Torque (Amps)		3
CASTING UNIT		
Casting Roll – meters per minute		1.62
Haul Off Roll ratio (speed relative to casting roll)		1.01
Casting Roll Nip – (kPa)		414
Film Gauge (mm)		0.711
Layflat (cm)		18.5
Throughput (gm/min)		264
H ₂ O Recirculation Unit (water to Casting and Nip Rolls)		
Temperature Set/Actual T (°C)	49	47

FIG. 10 is a schematic diagram of a system for processing data.

While certain of the preferred embodiments of the present invention have been described and specifically exemplified above, it is not intended that the invention be limited to such embodiments. Various modifications
5 may be made without departing from the scope and spirit of the present invention, as set forth in the following claims.

WHAT IS CLAIMED IS:

1. An impact modifier blend concentrate composition comprising
 - a. a polyester;
 - 5 b. an ethylene copolymer blend comprising an ethylene alkyl acrylate copolymer; and
 - c. a compatibilizer, said compatibilizer comprising an ionomer;wherein the polyester is present in an amount less than about 75 wt% and the ethylene copolymer blend is present in an amount
10 greater than about 25 wt%, based on the total weight of the impact modifier blend concentrate; and wherein the ratio of the weight of the ethylene alkyl acrylate copolymer to the weight of the compatibilizer preferably ranges from about 3:1 to about 6:1.
2. The impact modifier blend concentrate composition of claim 1,
15 wherein the amount of polyester ranges from about 40 to about 75 wt % and the amount of ethylene copolymer blend ranges from about 25 to about 60 wt%; and preferably the amount of polyester ranges from about 55 to about 70 wt % and the amount of ethylene copolymer blend ranges from about 30 to about 45 wt%, based on
20 the total weight of the impact modifier blend concentrate.
3. The impact modifier blend concentrate composition of claim 1,
wherein the polyester is selected from the group consisting of polyethylene terephthalate; polyethylene terephthalate that
25 incorporates isophthalic acid or cyclohexane dimethanol; copolymers of terephthalic acid, cyclohexane dimethanol and isophthalic acid; polyethylene naphthalate; copolyesters thereof; combinations thereof; and derivatives thereof; and preferably the polyester comprises polyethylene terephthalate; optionally the polyester comprises recycled polyester; and optionally the polyester
30 forms a continuous phase.
4. The impact modifier blend concentrate composition of claim 1,
wherein the ethylene alkyl acrylate copolymer has a melt flow index

less than 1 g/10 min; and preferably the ethylene alkyl acrylate copolymer has a melt flow index less than 0.5 g/10 min, when measured at 190°C with 2.16 kg weight.

5. The impact modifier blend concentrate composition of claim 1,
5 wherein the ionomer comprises a copolymer of ethylene and methacrylic acid or a copolymer of ethylene, methacrylic acid, and an alkyl methacrylate, and wherein the ionomer is at least partially neutralized with sodium ions.
6. The impact modifier blend concentrate composition of claim 1,
10 further comprising a crosslinking agent; preferably the crosslinking agent comprises one or more of diethylene glycol diacrylate, diethylene glycol dimethacrylate, ethylene glycol dimethacrylate, N,N'-m-phenylene dimaleimide, polyethylene glycol dimethacrylate, triallyl cyanurate, triallylisocyanurate, trimethylolpropane
15 trimethacrylate, tetraallyloxyethane, tetramethylene diacrylate, 2,5-dimethyl-2,5-di-(*t*-butylperoxy) hexyne-3, *t*-butyl peroxybenzoate, 2,5-dimethyl-2,5-di-(*t*-butylperoxy)-2,5-dimethylhexane, dicumyl peroxide, copolymers of ethylene with maleic acid or anhydride, and α,α -bis(*t*-butylperoxy)-2,5-dimethylhexane; and more preferably
20 the crosslinking agent comprises diethylene glycol dimethacrylate, 2,5-dimethyl-2,5-di-(*t*-butylperoxy) hexane, or a combination of both diethylene glycol dimethacrylate and 2,5-dimethyl-2,5-di-(*t*-butylperoxy) hexane.
7. An article comprising a bulk polyester and the impact modifier blend
25 concentrate composition of any of claims 1 through 6, wherein the article is preferably a dual-ovenable tray; and also preferably the amount of the impact modifier blend concentrate composition ranges from about 10 wt% to about 40 wt%, based on the combined weight of the bulk polyester and the impact modifier
30 blend concentrate composition; and also preferably the ethylene copolymer blend forms a dispersed phase that is particulate and the particle size is less than or equal to about 2 μm .

8. The article of claim 7, wherein the bulk polyester is selected from the group consisting of polyethylene terephthalate; polyethylene terephthalate that incorporates isophthalic acid or cyclohexane dimethanol; copolymers of terephthalic acid, cyclohexane dimethanol and isophthalic acid; polyethylene naphthalate; copolyesters thereof; combinations thereof; and derivatives thereof; and preferably the bulk polyester comprises polyethylene terephthalate; and optionally the polyester or the bulk polyester comprises recycled polyester.
9. A method of making an impact modifier blend concentrate composition comprising the steps of reacting an ethylene alkyl acrylate and a crosslinking agent, optionally in an extruder, to produce a higher molecular weight ethylene alkyl acrylate; combining the higher molecular weight ethylene alkyl acrylate with a compatibilizer comprising an ionomer to produce an ethylene copolymer blend; and combining the ethylene copolymer blend with a polyester; wherein the higher molecular weight ethylene alkyl acrylate preferably has a melt index less than or equal to 0.3 g/10 min when measured according to ASTM D1238; and wherein the crosslinking agent comprises one or more of diethylene glycol diacrylate, diethylene glycol dimethacrylate, ethylene glycol dimethacrylate, N,N'-m-phenylene dimaleimide, polyethylene glycol dimethacrylate, triallyl cyanurate, triallylisocyanurate, trimethylolpropane trimethacrylate, tetraallyloxyethane, tetramethylene diacrylate, 2,5-dimethyl-2,5-di-(*t*-butylperoxy) hexyne-3, *t*-butyl peroxybenzoate, 2,5-dimethyl-2,5-di-(*t*-butylperoxy)-2,5-dimethylhexane, dicumyl peroxide, copolymers of ethylene with maleic acid or anhydride, and $\alpha\alpha$ -bis(*t*-butylperoxy)-2,5-dimethylhexane, and more preferably the crosslinking agent comprises diethylene glycol dimethacrylate, 2,5-dimethyl-2,5-di-(*t*-butylperoxy) hexane, or a combination of both

1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23 24 25 26 27 28 29 30 31 32 33 34 35 36 37 38 39 40 41 42 43 44 45 46 47 48 49 50

diethylene glycol dimethacrylate and 2,5-dimethyl-2,5-di-(*t*-butylperoxy) hexane.

10. A method of making an impact resistant polyester, said method comprising the steps of

- 5 a. providing an impact modifier blend concentrate comprising
- i. a polyester;
 - ii. an ethylene copolymer blend comprising an ethylene alkyl acrylate copolymer;
 - 10 iii. a compatibilizer, said compatibilizer comprising an ionomer; and, optionally,
 - iv. a crosslinking agent, said crosslinking agent preferably comprising one or more of diethylene glycol diacrylate, diethylene glycol dimethacrylate, ethylene glycol dimethacrylate, N,N'-m-phenylene dimaleimide, polyethylene glycol dimethacrylate, triallyl cyanurate, triallylisocyanurate, trimethylolpropane trimethacrylate, tetraallyloxyethane, tetramethylene diacrylate, 2,5-dimethyl-2,5-di-(*t*-butylperoxy) hexyne-3, *t*-butyl peroxybenzoate, 2,5-dimethyl-2,5-di-(*t*-butylperoxy)-2,5-dimethylhexane, dicumyl peroxide, copolymers of ethylene with maleic acid or anhydride, and $\alpha\alpha$ -bis(*t*-butylperoxy)-2,5-dimethylhexane; and more preferably comprising diethylene glycol dimethacrylate, 2,5-dimethyl-2,5-di-(*t*-butylperoxy) hexane, or a combination of both diethylene glycol dimethacrylate and 2,5-dimethyl-2,5-di-(*t*-butylperoxy) hexane;
- 15
- wherein the polyester is present in an amount less than about 75 wt% and the ethylene copolymer blend is present in an amount greater than about 25 wt%, based on the total weight of the impact modifier blend concentrate; and
- 20
- 25
- 30 b. combining the impact modifier blend concentrate with a bulk polyester, optionally by extrusion; and

- 5 c. optionally, when the impact modifier blend concentrate comprises a crosslinking agent, reacting the ethylene alkyl acrylate and the crosslinking agent to produce a higher molecular weight ethylene alkyl acrylate; combining the higher molecular weight ethylene alkyl acrylate with a compatibilizer comprising an ionomer to produce an ethylene copolymer blend; and combining the ethylene copolymer blend with a polyester to produce the impact modifier blend concentrate; and,
- 10 d. also optionally, comprising the step of drying the bulk polyester, the impact modifier blend concentrate composition, or both the bulk polyester and the impact modifier blend concentrate composition prior to combining the impact modifier blend concentrate composition with the
- 15 bulk polyester or prior to extruding the impact modifier blend concentrate composition with the bulk polyester;
- wherein preferably the polyester or the bulk polyester is selected from the group consisting of polyethylene terephthalate; polyethylene terephthalate that incorporates isophthalic acid or
- 20 cyclohexane dimethanol; copolymers of terephthalic acid, cyclohexane dimethanol and isophthalic acid; polyethylene naphthalate; copolyesters thereof; combinations thereof; and derivatives thereof; more preferably the polyester or the bulk polyester comprises polyethylene terephthalate; and optionally the
- 25 polyester or the bulk polyester comprises recycled polyester.
11. An impact resistant polyester obtainable by the method of claim 10, wherein the ethylene copolymer blend preferably forms a dispersed phase in the impact resistant polyester that is particulate, and also preferably the particle size is less than or equal to about 2 μm .
- 30 12. A method of making an impact resistant article, said method comprising the steps of

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- 10
- 15
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- 30
- a. providing an impact modifier blend concentrate composition comprising
 - i. a polyester;
 - ii. an ethylene copolymer blend comprising an ethylene alkyl acrylate copolymer;
 - iii. a compatibilizer, said compatibilizer comprising an ionomer; and, optionally,
 - iv. a crosslinking agent, said crosslinking agent preferably comprising one or more of diethylene glycol diacrylate, diethylene glycol dimethacrylate, ethylene glycol dimethacrylate, N,N'-m-phenylene dimaleimide, polyethylene glycol dimethacrylate, triallyl cyanurate, triallylisocyanurate, trimethylolpropane trimethacrylate, tetraallyloxyethane, tetramethylene diacrylate, 2,5-dimethyl-2,5-di-(*t*-butylperoxy) hexyne-3, *t*-butyl peroxybenzoate, 2,5-dimethyl-2,5-di-(*t*-butylperoxy)-2,5-dimethylhexane, dicumyl peroxide, copolymers of ethylene with maleic acid or anhydride, and $\alpha\alpha$ -bis(*t*-butylperoxy)-2,5-dimethylhexane; and more preferably comprising diethylene glycol dimethacrylate, 2,5-dimethyl-2,5-di-(*t*-butylperoxy) hexane, or a combination of both diethylene glycol dimethacrylate and 2,5-dimethyl-2,5-di-(*t*-butylperoxy) hexane;wherein the polyester is present in an amount less than about 75 wt% and the ethylene copolymer blend is present in an amount greater than about 25 wt%, based on the total weight of the impact modifier blend concentrate; and
 - b. combining the impact modifier blend concentrate composition with a bulk polyester, preferably by extrusion; and
 - c. forming the impact resistant article from the impact modifier blend concentrate composition and the bulk polyester;

- 5 d. optionally, when the impact modifier blend concentrate comprises a crosslinking agent, reacting the ethylene alkyl acrylate and the crosslinking agent to produce a higher molecular weight ethylene alkyl acrylate; combining the higher molecular weight ethylene alkyl acrylate with a compatibilizer comprising an ionomer to produce an ethylene copolymer blend; and combining the ethylene copolymer blend with a polyester to produce the impact modifier blend concentrate; and,
- 10 e. also optionally, comprising the step of drying the bulk polyester, the impact modifier blend concentrate composition, or both the bulk polyester and the impact modifier blend concentrate composition prior to combining the impact modifier blend concentrate composition with the
- 15 bulk polyester or prior to extruding the impact modifier blend concentrate composition with the bulk polyester;
- wherein preferably the polyester or the bulk polyester is selected from the group consisting of polyethylene terephthalate; polyethylene terephthalate that incorporates isophthalic acid or
- 20 cyclohexane dimethanol; copolymers of terephthalic acid, cyclohexane dimethanol and isophthalic acid; polyethylene naphthalate; copolyesters thereof; combinations thereof; and derivatives thereof; more preferably the polyester or the bulk polyester comprises polyethylene terephthalate; and optionally the
- 25 bulk polyester comprises recycled polyester.
13. An article obtainable by the method of claim 12, wherein the article is preferably a dual-ovenable tray, and wherein the ethylene copolymer blend preferably forms a dispersed phase in the impact resistant polyester that is particulate, and also preferably the
- 30 particle size is less than or equal to about 2 μm .

INTERNATIONAL SEARCH REPORT

International application No

PCT/US2006/004226

A. CLASSIFICATION OF SUBJECT MATTER
INV. C08L67/02

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)

C08L

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 practical, search terms used)

EPO-Internal, WPI Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 2002/055586 A1 (DALGEWICZ EDWARD J ET AL) 9 May 2002 (2002-05-09) claim 1	1-13
Y	US 5 277 864 A (BLATZ ET AL) 11 January 1994 (1994-01-11) column 3, line 10 - line 12; claim 1	1-13

☐ 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 document but published on or after the international filing date

L document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

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

28 June 2006

Date of mailing of the International search report

05/07/2006

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/US2006/004226

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