

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property
Organization

International Bureau

(43) International Publication Date
03 October 2024 (03.10.2024)



(10) International Publication Number
WO 2024/206154 A1

(51) International Patent Classification:
C08L 69/00 (2006.01)

LU, LV, MC, ME, MK, MT, NL, NO, PL, PT, RO, RS, SE,
SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN,
GQ, GW, KM, ML, MR, NE, SN, TD, TG).

(21) International Application Number:
PCT/US2024/021159

Published:

— with international search report (Art. 21(3))

(22) International Filing Date:
22 March 2024 (22.03.2024)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
63/492,058 24 March 2023 (24.03.2023) US

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(81) Designated States (unless otherwise indicated, for every kind of national protection available): AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CV, CZ, DE, DJ, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IQ, IR, IS, IT, JM, JO, JP, KE, KG, KH, KN, KP, KR, KW, KZ, LA, LC, LK, LR, LS, LU, LY, MA, MD, MG, MK, MN, MU, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SA, SC, SD, SE, SG, SK, SL, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, WS, ZA, ZM, ZW.

(84) Designated States (unless otherwise indicated, for every kind of regional protection available): ARIPO (BW, CV, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SC, SD, SL, ST, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT,

(54) Title: THERMAL STABILIZATION PACKAGE FOR POLYCARBONATE/ACRYLONITRILE-BUTADIENE-STYRENE BLENDS

(57) Abstract: Provided is a composition comprising: A) an aromatic polycarbonate or an aromatic polyester carbonate, B) an acrylonitrile/butadiene/styrene polymer, C) a stabilization package comprising C.1) a phosphate ester included in an amount greater than zero but less than 0.1 wt.%, C.2) a phenolic antioxidant included in an amount greater than zero but less than 0.3 wt.%, and optionally C.3) a UV light absorber, and optionally D) one or more components selected from the group consisting of polymer additives, polymeric blending partners different from component B, and optionally E) colorants. The composition provides good thermal stabilization of polycarbonate/acrylonitrile/butadiene/styrene (PC/ABS) blends with a surprisingly low variation in the yellowness index (YI) value and reduces residual BPA content while retaining mechanical and thermal properties and providing potentially good hydrolytic stability.



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**THERMAL STABILIZATION PACKAGE FOR
POLYCARBONATE/ACRYLONITRILE-BUTADIENE-STYRENE BLENDS**

FIELD OF THE INVENTION

[0001] The present invention relates in general to, plastics and more specifically to, a thermal stabilization package for polycarbonate/acrylonitrile/butadiene/styrene (PC/ABS) blends.

BACKGROUND OF THE INVENTION

[0002] Many polycarbonate/acrylonitrile/butadiene/styrene (PC/ABS) + flame retardant patents and applications provide PC/ABS blends containing organophosphate flame retardants and hindered alkylated phenols as a thermostabilizer. These patents disclose that, when used as flame retardants, the organophosphates are typically used in concentrations >2 wt%, in formulations with higher ABS contents typically even in concentrations of 10 wt% or higher. The patents further disclose that for flame retardation of blends of aromatic polycarbonates with ABS aromatic organophosphate flame retardants are typically used as aromatic (in contrast to aliphatic) organophosphates will better disperse and even dissolve in both the aromatic polycarbonate and the ABS phases which is a prerequisite to achieve the desired material performance.

[0003] Also, many patents and applications provide non-flame-retardant PC/ABS blends that do not contain organophosphate flame retardants. WO/2022/058222 for example, teaches a multi-layer body comprising, in this order, (I) a substrate layer made of a specific thermoplastic polycarbonate molding compound, (II) at least one layer made of a colorant or a colorant composition, and (III) a film. The disclosure also relates to a lighting unit, comprising the multi-layer body and a light source, to a method for producing the multi-layer body, and to the use of the specific thermoplastic polycarbonate molding compound as a substrate layer of a multi-layer body of this type. Backlighted translucent composite parts containing a carrier and a decorative ink-printed film in which the carrier is made by injection molding of a thermoplastic composition with improved transmittance of visible light containing polycarbonate and bulk polymerized acrylonitrile/butadiene/styrene (ABS), the composition being essentially free of emulsion polymerized graft polymer and containing a specific low amount of polybutadiene rubber.

[0004] WO/2008/005655 discloses a method of making polycarbonate including the steps of forming polycarbonate by a melt transesterification method using an activated diaryl carbonate and compounding the polycarbonate with a phosphorus-containing compound that

has an abstractable proton or hydrolyzable group. The phosphorous-containing compound is compounded with the polycarbonate in an amount said to be sufficient to result in an improvement in the color properties of the polycarbonate as compared to pellets formed from the same polycarbonate without addition of the phosphorus-containing compound.

[0005] EP2935436B1 is directed to a colored infrared-reflective polycarbonate molding compound that has high melt stability containing at least one infrared (IR) reflective pigment and at least one phosphate stabilizer, preferably TOF (triisooctyl phosphate (tris-2-ethylhexyl phosphate)) was used as stabilizer of the composition, and products made thereof, in particular multilayered bodies. The disclosure also mentions IRGANOX 1076 and TINUVIN 329/360 as additional preferable additives.

[0006] EP3802121B1 is directed to a multilayer composite part comprising a substrate layer with a light transmission in the wavelength area of 380 to 780 nm of less than 0.5%, measured at a thickness of 4 mm according to DIN ISO 13468-2:2006 (D65, 10°) which consists of a thermoplastic composition based on polycarbonate containing titanium dioxide as a white pigment with a specified particle size and at least two colorants, which are either one organic and one inorganic or two organic colorants, whereby the inorganic colorant is not titanium dioxide or carbon black. This patent also discloses that the substrate layer compositions can optionally contain at least one additional component such as e.g., UV stabilizers (benzotriazoles and TINUVIN 329 and TINUVIN 360 are explicitly mentioned) and favorably at least one thermal stabilizer in amounts of favorably up to 0.20 wt. As suitable thermal stabilizers, are mentioned among others, alkylphosphates, especially triisooctylphosphate (tris-2-ethyl-hexyl-phosphate). The patent further states that phenolic antioxidants, such as alkylated phenols, can be used in the thermoplastic compositions, most preferably IRGANOX 1076 (2,6-di-tert-butyl-4-(octadecanoxycarbonyl)phenol). Also, pentaerythritol tetrastearate (PETS) is said to be potentially useful as a mold release agent.

[0007] A need continues to exist in the art for improved thermal stabilization packages for PC/ABS blends with low variation in yellowness index values.

SUMMARY OF THE INVENTION

[0008] Accordingly, the present invention obviates problems inherent in the art by providing a stabilization package based on a synergistic combination of a phosphate flame retardant and an antioxidant which provides good thermal stabilization of polycarbonate/acrylonitrile/butadiene/styrene (PC/ABS) blends with a surprisingly low

variation in the yellowness index (YI) value. The inventive stabilization package may be combined with a color matching package to provide a new resin, which preferably is a PC/mass-ABS blend. The inventive stabilization package provides better thermal stability of PC/mass-ABS blends, especially in processes using an extended time in the melt and elevated melt temperature during injection molding. The stabilization package also reduces residual in such PC/mass-ABS blends the PC molecular weight degradation and BPA formation upon thermal exposure of an injection molding process and therefore the retention of the mechanical as well as thermal properties, and potentially hydrolytic stability.

[0009] These and other advantages and benefits of the present invention will be apparent from the Detailed Description of the Invention herein below.

BRIEF DESCRIPTION OF THE FIGURE

[0010] The present invention will now be described for purposes of illustration and not limitation in conjunction with the figures, wherein:

[0011] FIG. 1 is a chromatogram of time vs. MV.

DETAILED DESCRIPTION OF THE INVENTION

[0012] The present invention will now be described for purposes of illustration and not limitation. Except in the operating examples, or where otherwise indicated, all numbers expressing quantities, percentages, and so forth in the specification are to be understood as being modified in all instances by the term “about.”

[0013] Any numerical range recited in this specification is intended to include all sub-ranges of the same numerical precision subsumed within the recited range. For example, a range of “1.0 to 10.0” is intended to include all sub-ranges between (and including) the recited minimum value of 1.0 and the recited maximum value of 10.0, that is, having a minimum value equal to or greater than 1.0 and a maximum value equal to or less than 10.0, such as, for example, 2.4 to 7.6. Any maximum numerical limitation recited in this specification is intended to include all lower numerical limitations subsumed therein and any minimum numerical limitation recited in this specification is intended to include all higher numerical limitations subsumed therein. Accordingly, Applicant reserves the right to amend this specification, including the claims, to expressly recite any sub-range subsumed within the ranges expressly recited herein. All such ranges are intended to be inherently described in this specification such that amending to expressly recite any such sub-ranges would comply with the requirements of 35 U.S.C. §112(a), and 35 U.S.C. §132(a). The various embodiments

disclosed and described in this specification can comprise, consist of, or consist essentially of the features and characteristics as variously described herein.

[0014] Any patent, publication, or other disclosure material identified herein is incorporated by reference into this specification in its entirety unless otherwise indicated, but only to the extent that the incorporated material does not conflict with existing definitions, statements, or other disclosure material expressly set forth in this specification. As such, and to the extent necessary, the express disclosure as set forth in this specification supersedes any conflicting material incorporated by reference herein. Any material, or portion thereof, that is said to be incorporated by reference into this specification, but which conflicts with existing definitions, statements, or other disclosure material set forth herein, is only incorporated to the extent that no conflict arises between that incorporated material and the existing disclosure material. Applicant reserves the right to amend this specification to expressly recite any subject matter, or portion thereof, incorporated by reference herein.

[0015] Reference throughout this specification to “various non-limiting embodiments,” “certain embodiments,” or the like, means that a particular feature or characteristic may be included in an embodiment. Thus, use of the phrase “in various non-limiting embodiments,” “in certain embodiments,” or the like, in this specification does not necessarily refer to a common embodiment, and may refer to different embodiments. Further, the particular features or characteristics may be combined in any suitable manner in one or more embodiments. Thus, the particular features or characteristics illustrated or described in connection with various or certain embodiments may be combined, in whole or in part, with the features or characteristics of one or more other embodiments without limitation. Such modifications and variations are intended to be included within the scope of the present specification.

[0016] The grammatical articles “a”, “an”, and “the”, as used herein, are intended to include “at least one” or “one or more”, unless otherwise indicated, even if “at least one” or “one or more” is expressly used in certain instances. Thus, these articles are used in this specification to refer to one or more than one (i.e., to “at least one”) of the grammatical objects of the article. By way of example, and without limitation, “a component” means one or more components, and thus, possibly, more than one component is contemplated and may be employed or used in an implementation of the described embodiments. Further, the use of a singular noun includes the plural, and the use of a plural noun includes the singular, unless the context of the usage requires otherwise.

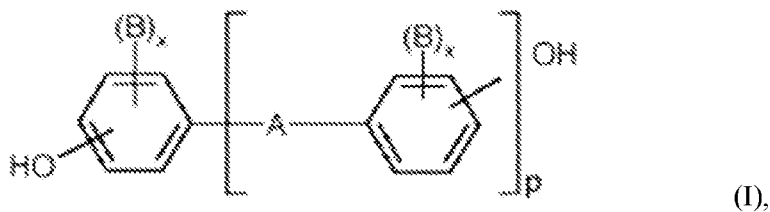
[0017] To prevent a substantial increase in the yellowness index (YI) value in PC/ABS blends, in particular PC/mass-ABS blends during extended molding cycle, various thermal stabilization packages have been evaluated using extended time in the melt and elevated melt temperature during injection molding. The inventive stabilization package with surprisingly low variation in the YI value has been discovered. This stabilization package together with a color matching package have been combined to produce a new resin. The inventive stabilization package also can help to minimize the M_w reduction, reduce residual bisphenol A (BPA) content, and potentially improves the hydrolysis resistance of polycarbonate during molding processes, especially those processes relying on extended time in the melt and elevated melt temperature during injection.

[0018] In a first embodiment, the present invention is directed to a composition comprising: A) an aromatic polycarbonate or an aromatic polyester carbonate, B) an acrylonitrile/butadiene/styrene polymer, C) a stabilization package comprising C.1) a phosphate ester, preferably an aliphatic phosphate ester included in an amount greater than zero but less than 0.1 wt%, C.2) a phenolic antioxidant included in an amount greater than zero but less than 0.3 wt%, and optionally C.3) a UV light absorber, and optionally D) one or more selected from the group consisting of polymer additives, polymeric blending partners different from component B, and optionally E) colorants.

Component A

[0019] Aromatic polycarbonates and / or aromatic polyester carbonates suitable for use as component A are known in the literature or can be prepared by methods in the literature (for the preparation of aromatic polycarbonates see, for example, Schnell, "Chemistry and Physics of Polycarbonates", Interscience Publishers, 1964 and U.S. Pat. No. 3,553,167, DE-A 2,232,877, U.S. Pat. No. 4,075,173, GB1552558, U.S. Pat. No. 4,311,823, DE-A 3,832,396; for the preparation of aromatic polyester carbonates, e.g., CA1173998). The preparation of aromatic polycarbonates is carried out, for example, by reacting diphenols with carbonic acid halides, preferably phosgene and / or with aromatic dicarboxylic acid dihalides, preferably benzenedicarboxylic acid dihalides, by the phase boundary method, optionally using chain terminators, for example monophenols and optionally using trifunctional or more than trifunctional branches, for example triphenols or tetraphenols. Likewise, a preparation via a melt polymerization process by reaction of diphenols with, for example, diphenyl carbonate is possible.

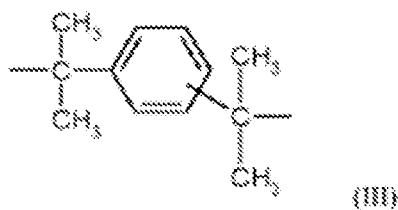
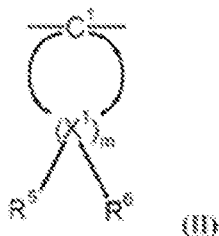
[0020] Diphenols for the preparation of the aromatic polycarbonates and / or aromatic polyester carbonates are preferably those of formula (I)



wherein

A is a single bond, C₁ to C₅-alkylene, C₂ to C₅-alkylidene, C₅ to C₆-cycloalkylidene, -O-, -SO-, -CO-, -S-, -SO₂-, C₆ to C₁₂ arylene, to which further aromatic optionally heteroatom-containing rings may be condensed,

or a remainder of formula (II) or (III)



B in each case is C₁ to C₁₂ alkyl, preferably methyl, halogen, preferably chlorine and / or bromine

x independently is 0, 1 or 2,

p is 1 or 0, and

R⁵ and R⁶ for each X¹ is independently hydrogen or C₁ to C₆-alkyl, preferably hydrogen, methyl, or ethyl,

X¹ is carbon and

m is an integer from 4 to 7, preferably 4 or 5, with the proviso that at least one atom X¹, R⁵, and R⁶ is alkyl.

[0021] Preferred diphenols are hydroquinone, resorcinol, dihydroxydiphenols, bis (hydroxyphenyl)-C₁-C₆-alkanes, bis (hydroxyphenyl) -C₅-C₆ cycloalkanes, bis (hydroxyphenyl) ether, bis (hydroxyphenyl) sulfoxides, bis (hydroxyphenyl) ketones, bis (hy-

droxyphenyl) sulfones and α,α -bis (hydroxyphenyl) diisopropylbenzenes and ring-brominated and / or ring-chlorinated derivatives thereof.

[0022] Particularly preferred diphenols are 4,4'-dihydroxydiphenyl, bisphenol-A, 2,4-bis (4-hydroxyphenyl) -2-methylbutane, 1, 1-bis (4-hydroxyphenyl) cyclohexane, 1, 1-bis (4-hydroxyphenyl) -3,3, 5-trimethylcyclohexane, 4,4'-dihydroxydiphenyl sulfide, 4,4'-dihydroxydiphenyl sulfone and di- and tetrabrominated or chlorinated derivatives thereof, such as 2,2-bis (3-chloro-4-hydroxyphenyl) propane, 2,2-bis (3,5-dichloro-4-hydroxyphenyl)-propane or 2,2-bis-(3,5-dibromo-4-hydroxyphenyl)-propane. Particularly preferred is 2,2-bis (4-hydroxyphenyl) propane (bisphenol-A).

[0023] The diphenols can be used individually or as any mixture. The diphenols are known to literature or available by methods known to the literature.

[0024] For the preparation of thermoplastic, aromatic polycarbonates suitable chain terminators are, for example, phenol, p-chlorophenol, p-tert-butylphenol or 2,4,6-tribromophenol, but also long-chain alkylphenols such as 4- [2- (2,4,4-trimethylpentyl)] -phenol, 4-(1, 3-tetramethylbutyl) phenol according to U.S. Pat. No. 4,269,964 or monoalkylphenol or dialkylphenols having a total of 8 to 20 carbon atoms in the alkyl substituents, such as 3,5-di-tert-butylphenol, p-iso-octylphenol, p-tert-octylphenol, p-dodecylphenol and 2- (3,5-dimethylheptyl) phenol and 4- (3,5-dimethylheptyl) phenol. The amount of chain terminators to be used is preferably between 0.5 mol% and 10 mol%, based on the molar sum of the diphenols used.

[0025] The thermoplastic, aromatic polycarbonates have average molecular weights (weight average M_w , measured by GPC (gel permeation chromatography) with polycarbonate standard based on bisphenol A) of preferably 20000 to 40000 g/mol, more preferably 24000 to 32000 g/mol, more preferably 26000 to 30000 g/mol. By the preferred areas is achieved in the inventive compositions a particularly advantageous balance of mechanical and rheological properties.

[0026] The thermoplastic, aromatic polycarbonates may be branched in a known manner, preferably by the incorporation of 0.05 to 2.0 mol%, based on the sum of the diphenols used, on three-fictional or more than three-factor compounds, for example those having three and more phenolic groups. Linear polycarbonates, more preferably based on bisphenol A, are preferably used.

[0027] Both homopolycarbonates and copolycarbonates are suitable. For the preparation of inventive copolycarbonates according to component A, 1 to 25 wt.%, preferably 2.5 to 25 wt.%, Based on the total amount of diphenols to be used, polydiorganosiloxanes having hydroxyaryloxy end groups can be used. These are known (U.S. Pat. No. 3,419,634) and can be prepared by methods known in the literature. Also suitable are polydiorganosiloxane-containing copolycarbonates; the preparation of the polydiorganosiloxaneiger copolycarbonates is described, for example, in U.S. Pat. No. 4,584,360.

[0028] Aromatic dicarboxylic acid dihalogenides for the preparation of aromatic polyester carbonates are preferably the diacid dichlorides of isophthalic acid, terephthalic acid, diphenyl ether-4,4'-dicarboxylic acid and naphthalene-2,6-dicarboxylic acid.

[0029] Particularly preferred are mixtures of the diacid dichlorides of isophthalic acid and terephthalic acid in the ratio between 1:20 and 20:1.

[0030] In the preparation of polyester carbonates, a carbonic acid halide, preferably phosgene, is additionally used as a bifunctional acid derivative.

[0031] As chain terminators for the preparation of the aromatic polyester carbonates, in addition to the monophenols already mentioned, their chlorinated carbon dioxide esters and the acid chlorides of aromatic monocarboxylic acids, which may optionally be substituted by C₁ to C₂₂ alkyl groups or by halogen atoms, and aliphatic C₂ to C₂₂ monocarboxylic acid chlorides into consideration.

[0032] The amount of chain terminators is in each case 0.1 to 10 mol%, based in the case of phenolic chain terminators on mole diphenol and in the case of monocarboxylic acid chloride chain terminators on moles of dicarboxylic acid dichloride.

[0033] In the preparation of aromatic polyester carbonates, one or more aromatic hydroxycarboxylic acid can also be used.

[0034] The aromatic polyester carbonates may be branched both linearly and in a known manner (see U.S. Pat. No. 4,334,053 and CA 1173998), wherein linear polyester carbonates are preferred.

[0035] Suitable branching agents can, for example, three- or multifunctional carboxylic acid chlorides such as trimesic acid trichloride, cyanuric acid trichloride, 3,3', 4,4'-benzophenone-tetracarboxylic acid tetrachloride, 1,4,5,8-naphthalenetetracarboxylic acid tetrachloride or pyromellitic acid tetrachloride, in amounts of 0.01 to 1.0 mol% (based on

dicarboxylic acid dichlorides used) or three- or multifunctional phenols such as phloroglucinol, 4,6-dimethyl-2,4,6-tri- (4-hydroxyphenyl) -hept-2-ene, 4,6-dimethyl-2,4,6-tri- (4-hydroxyphenyl) heptane, 1,3,5-tri- (4-hydroxyphenyl) benzene, 1,1, 1-tri- (4-hydroxyphenyl) ethane, tri- (4-hydroxyphenyl)-phenylmethane, 2,2-bis [4, 4-bis (4-hydroxyphenyl) -cyclohexyl] -propane, 2,4-bis (4-hydroxyphenyl-isopropyl) phenol, tetra- (4-hydroxyphenyl) -methane, 2,6-bis (2-hydroxy-5-methyl-benzyl) -4-methyl-phenol, 2- (4-hydroxyphenyl)-2-(2,4-dihydroxyphenyl) -propane, tetra- (4-[4-hydroxyphenyl-isopropyl]-phenoxy)-methane, 1, 4-bis [4,4'-dihydroxytri-phenyl) -methyl] benzene, in amounts of 0.01 to 1.0 mol% based on diphenols used. Phenolic branching agents can be used with the diphenols; acid chloride branching agents can be used together with the acid dichlorides.

[0036] In the thermoplastic, aromatic polyester carbonates, the proportion of carbonate structural units can vary arbitrarily. Preferably, the proportion of carbonate groups is up to 100 mol%, in particular up to 80 mol%, particularly preferably up to 50 mol%, based on the sum of ester groups and carbonate groups. Both the ester and the carbonate content of the aromatic polyester carbonates can be in the form of blocks or statistically distributed in the polycondensate.

[0037] The thermoplastic, aromatic polycarbonates and polyester carbonates can be used alone or in any mixture.

[0038] Linear polycarbonate based exclusively on bisphenol A is preferably used as component A.

Component B

[0039] Component B is a rubber-modified vinyl (co) polymer of

[0040] B. 1) 80 to 95 wt.%, preferably 83 to 93 wt.%, more preferably 85 to 92 wt.%, based on the rubber-modified vinyl (co) polymer B, structural units derived from at least one vinyl monomer and

[0041] B.2) 5 to 20 wt.%, preferably 7 to 17 wt.%, more preferably 8 to 15 wt.%, based on the rubber-modified vinyl (co) polymer B, one or more rubber-elastic graft bases having glass transition temperatures $T_g < -50^\circ \text{C}$, preferably from $< -60^\circ \text{C}$, particularly preferably $< -70^\circ \text{C}$ containing at least 50 wt.%, preferably at least 70 wt.%, particularly preferably 100 wt.%, based on B.2, derived from 1,3-butadiene structural units,

wherein the rubber-modified vinyl (co) polymer B is

(i) a disperse phase consisting of

(i. 1) with vinyl (co) polymer of structural units according to B.1 grafted rubber particles and

(i.2) vinyl (co)polymer enclosed in the rubber particles as a separated disperse phase also from structural units according to B.1

and

(ii) a rubber-free vinyl (co)polymer matrix not bound to the rubber particles and not enclosed in these rubber particles consisting of structural units according to B.1

and wherein the disperse phase according to (I) has an average diameter D50 measured by ultracentrifugation of 0.7 to 2.0 μm , preferably from 0.7 to 1.5 μm , in particular from 0.7 to 1.2 μm .

[0042] The glass transition temperature T_g is determined, unless expressly described otherwise in the present invention, for all components by means of differential scanning calorimetry (DSC) according to DIN EN 61006 (version of 1994) at a heating rate of 10 K / min with determination of the T_g as the center temperature (tangent method).

[0043] The rubber-modified vinyl (co) polymers according to component B have a melt flow rate (MVR), measured according to ISO 1133 (version of 2012) at 220 ° C with a stamp load of 10 kg, of preferably 2 to 20 ml/10min, more preferably 3 to 15 ml / 10min, in particular 4 to 8 ml/10min. If mixtures of several rubber-modified vinyl (co)polymers are used as component B, the preferred MVR ranges are the mean value of the MVR of the individual components weighted over the mass fractions of the components in the mixture.

[0044] Such rubber-modified vinyl (co) polymers B are prepared, for example, by radical polymerization, preferably in the mass polymerization process, of

B. 1 80 to 95 wt.%, preferably 83 to 93 wt.%, particularly preferably 85 to 92 wt.%, based on the rubber-modified vinyl (co) polymer B, at least one vinyl monomer in the presence of

B.2 5 to 20 wt.%, preferably 7 to 17 wt.%, particularly preferably 8 to 15 wt.%, based on the rubber-modified vinyl (co) polymer B, one or more rubber-elastic graft bases with glass transition temperatures < -50 ° C, preferably < -60 ° C, particularly preferably < -70 ° C, containing at least 50 wt.%, preferably at least 70 wt.%, particularly preferably 100 wt.% based on B.2, of structural units derived from 1,3-butadiene.

[0045] In the mass polymerization process used for the preparation of the rubber-modified vinyl (co) polymer B, both the polymerization of the vinyl monomers according to B.1 and a grafting of the resulting vinyl (co) polymer onto the rubber-elastic graft base according to B.2. Furthermore, in this reaction formation by self-organization (phase separation) the formation of a disperse phase (i) consisting of

(1.1) rubber particles grafted with vinyl(co)polymer from structural units according to B.1 and

(1.2) vinyl (co)polymer enclosed in the rubber particles as a separate disperse phase also from structural units according to B.1,

wherein this rubber-containing phase (i) is dispersed in a rubber-free vinyl (co) polymer matrix (ii) consisting of structural units according to B.1 not bound to the rubber particles and not enclosed in these rubber particles.

[0046] The rubber-free vinyl (co) polymer (ii) can be dissolved in contrast to the other vinyl (co) polymer proportions in component B by suitable solvents such as acetone.

[0047] The size of the disperse phase (i) in the rubber-modified vinyl (co) polymers B thus prepared is adjusted via the conditions of reaction formation such as temperature and resulting viscosity of the polymer and shear by, for example, stirring.

[0048] The average particle size D_{50} is the diameter above and below which 50 wt.% of the particles lie. It is, unless expressly described otherwise in the present invention, for all components by means of ultracentrifuge measurement (W. Scholtan, H. Lange, Colloid and Polymer Science, 250 (1972), 782-796).

[0049] The monomers B.1 are preferably mixtures consisting of

B.1.1 60 to 85 wt. preferably, 65 to 80 wt., more preferably, 70 to 78 wt., parts, each based on the sum of B.1.1 and B.1.2, styrene and

B.1.2 15 to 40 wt. - preferably 20 to 35 wt. more preferably 22 to 30 wt. parts, each based on the sum of B.1.1 and B.1.2, acrylonitrile, and optionally.

B.1.3 0-10 wt., preferably 0-7 wt., more preferably 0-5 wt. parts methyl methacrylate or n-butyl acrylate, each based on 100 wt.-parts as the sum of B.1.1 and B.1.2.

[0050] Preferred graft bases B.2 are diene rubbers containing butadiene, or mixtures of diene rubbers containing butadiene or copolymers of diene rubbers containing butadiene or mixtures thereof with other copolymerizable monomers (e.g., according to B.1.1 and B.1.2).

Particularly preferred graft bases B.2 are produced by anionic polymerization using a lithium compound as polymerization catalyst.

[0051] Particularly preferred as graft base B.2 is pure polybutadiene rubber. In a further preferred embodiment, B.2 is styrene-butadiene-block copolymer rubber.

[0052] The component B preferably has a polybutadiene content of 5 to 18 wt.%, more preferably from 7 to 15 wt.%, in particular from 8 to 13 wt.%.

[0053] Particularly preferred rubber-modified vinyl (co) polymers according to component B are mass ABS polymers such as those described for example in U.S. Pat. No. 3,644,574 (=DE-OS 2,035,390) or in DE-OS 2,248,242 (= GB-PS 1,409,275) or in Ullmann's Encyclopedia of Chemical Technology, Vol. 19 (1980), p. 280 ff. are described.

[0054] The not chemically bound to the rubber base (s) B.2 and not enclosed in the rubber particles vinyl (co) polymer (ii) can be formed as shown above due to the production during the polymerization of graft polymers B. It is also possible that a part of this not chemically bound to the rubber base (s) B.2 and not included in the rubber particles vinyl (co) polymer (ii) in the rubber-modified vinyl (co) polymer according to component B is formed due to production during its production in the mass polymerization process and another part is polymerized separately and added to component B as part of component B. The proportion of vinyl (co) polymer (ii), regardless of its origin, measured as an acetone-soluble portion, is in component B, based on component B, preferably at least 50 wt.%, particularly preferably at least 60 wt.%, more preferably at least 70 wt.%.

[0055] This vinyl (co) polymer (ii) has in the rubber-modified vinyl (co) polymers according to component B a weight-averaged molecular weight M_w from 70 to 250 kg / mol, preferably from 130 to 200 kg/mol, in particular from 150 to 180 kg/mol.

[0056] The weight-averaged molecular weight M_w of vinyl (co) polymer (ii) in component B is measured in the present invention by gel permeation chromatography (GPC) in tetrahydrofuran against polystyrene as standard.

[0057] Component B is preferably free of emulsifiers that are typically used in emulsion polymerization processes such as for example saturated fatty acids having 8 to 22 carbon atoms, resin acids, alkyl and alkylarylsulfonic acids, and fatty alcohol sulfates.

[0058] The component B preferably contains less than 100 ppm, more preferably less than 50 ppm, most preferably less than 20 ppm ions of alkali metals and alkaline earth metals.

The component B preferably contains an amount greater than zero but less than 10 ppm, more preferably 0.01 – 5 ppm, most preferably 0.1 – 2 ppm of lithium.

[0059] Suitable as component B rubber-modified vinyl (co) polymers are, for example, MAGNUM 3404, MAGNUM 3504, and MAGNUM 3904 from Trinseo S.A. (Luxembourg).

Component C

[0060] Component C is a stabilization package based on a synergistic combination of an organophosphate C.1, preferably an aliphatic organophosphate, included in an amount greater than zero but less than 0.1 wt.%, preferably in an amount of from 0.005 wt.% to 0.08 wt.%, more preferably in an amount of from 0.01 wt.% to 0.03 wt.% and a, preferably sterically hindered, phenolic antioxidant C.2 included in an amount greater than zero but less than 0.3 wt.%. Component C.1 preferably is tris (2-ethylhexyl) phosphate. Component C.2 preferably is octadecyl-3-[3,5-di-tert-butyl-4-hydroxyphenyl]propionate]. Component C optionally contains in addition a UV light absorber C.3, preferably a benzotriazole, more preferably 2-phenol, 2-(2H-benzotriazol-2-yl)-4-(1, 1, 3, 3-tetramethylbutyl). The UV light absorber C.3 is preferably used in an amount greater than zero but less than 1 wt.%. The stabilization package Component C provides good thermal stabilization of polycarbonate/acrylonitrile/butadiene/styrene (PC/ABS) blends with a surprisingly low variation in the yellowness index (YI) value.

[0061] Particularly preferred as organophosphate C.1 is tris (2-ethylhexyl) phosphate commercially available from Lanxess as DISFLAMOLL TOF.

[0062] Particularly preferred as phenolic antioxidant is octadecyl-3-[3,5-di-tert-butyl-4-hydroxyphenyl]propionate], commercially available from BASF as IRGANOX 1076.

Component D

[0063] As component D, one or more representatives selected from the group consisting of polymer additives and polymeric blending partners different from component B may optionally be included.

[0064] The polymer additives or polymeric blend partners different from component B are preferably selected from the group consisting of lubricants and demolding agents, other stabilizers, colorants, compatibility mediation, further of component B different impact resistance modifiers, further from components A and B different polymeric components (for example, functional blending partners or graft polymers with core-shell structure prepared in the emulsion polymerization process) and fillers and fillers and reinforcing materials.

[0065] In the preferred embodiment, no fillers or reinforcing materials are contained in component D. More preferably, no polymeric blend partners different from component B are included. More preferably, no polymeric components different from components A and B are included. In a particularly preferred embodiment, neither fillers or reinforcing agents, nor polymeric blend partners different from component B are included. Most preferably, neither fillers or reinforcing agents, nor polymeric components different from component A and B are included.

[0066] In a preferred embodiment, at least one polymer additive selected from the group consisting of lubricants and molding agents is used as component D.

[0067] As lubricants and demolding agents, fatty acid esters, particularly preferably fatty acid esters of pentaerythritol or glycerol, are used in a preferred embodiment.

[0068] In a preferred embodiment, at least one polymer additive selected from the group of UV stabilizers is used as component D.

[0069] As UV stabilizer particularly preferably benzotriazoles are used in a preferred embodiment.

Component E (Colorant composition)

[0070] Optional colorant compositions E, useful in the invention are organic and inorganic pigments and dyes, including soluble dyes. In addition, one or more other components, which can be divided into volatile and non-volatile components, may be included.

[0071] Non-volatile components include binders, fillers, and auxiliaries. These are usually only needed in very small quantities but are often indispensable for problem-free processing.

[0072] The volatile components are essentially liquids in which the colorants and possible other components are dissolved or dispersed. It can be organic, inorganic solvents or mixtures of several solvents. Often the solvent is water or a mixture of water and another solvent.

[0073] Suitable organic solvents are, for example, ketones, esters, alcohols and aromatic or aliphatic hydrocarbons. It can also be used mixtures of several organic or inorganic solvents.

[0074] The non-volatile binders ensure that the colorants are anchored to the substrate so that the finished print resists stresses caused by abrasion, heat, and mechanical bending.

[0075] Suitable binders for the colorants are, for example, nitrocellulose in combination with plasticizers, thermoplastic polyurethanes, thermoplastic polyesters, thermoplastic polycarbonates, and thermoplastic poly (meth) acrylates. It is also possible that different binders are combined, as described in DE 198 32 570 A1. It is also possible to form the binder (e.g., a polyurethane or an epoxy resin) during the application of the colorant composition by chemical reaction in situ.

[0076] The selection of suitable colorants is practically unrestricted, provided that sufficient temperature resistance is guaranteed. Suitable organic colorants include, for example, colorants from the azo, anthraquinone, azoporphine, thioindigo, dioxazine, naphthalenetetracarboxylic acid or perylenetetracarboxylic acid series and phthalocyanine compounds. Suitable inorganic colorants include, for example, iron oxides, ultramarines, zinc sulfides, silicon dioxides, aluminum oxides, titanium oxides, nickel and chromium compounds, phosphorus-tungsten-molybdenum acid bronzes and carbon blacks. Colorants with special effects, such as metal oxide-coated mica pigments and metallic aluminum pigments, can also be used.

[0077] Particularly suitable colorants are those based on anthraquinone, on perinone, or on phthalocyanine, or those derived from such structures. Particularly preferred colorants are described in WO 2012/080395 A1. It is also possible to use the following as colorants: MACROLEX VIOLET 3R (CAS 61951-89-1; SOLVENT VIOLET 36), MACROLEX GREEN 5B (CAS 128-80-3; SOLVENT GREEN 3; C.I. 61565), AMAPLAST YELLOW GHS (CAS 13676-91-0; SOLVENT YELLOW 163; C.I. 58840), MACROLEX ORANGE 3G (CAS 6925-69-5; SOLVENT ORANGE 60; C.I. 564100), MACROLEX BLUE RR (CAS 32724-62-2; SOLVENT BLUE 97; C.I. 615290); KEYPLAST BLUE KR (CAS 116-75-6; SOLVENT BLUE 104; C.I. 61568), HELIOGEN BLUE types (e.g., HELIOGEN BLUE K 6911; CAS 147-14-8; PIGMENT BLUE 15:1; C.I. 74160), HELIOGEN GREEN types (e.g., HELIOGEN GREEN K 8730; CAS 1328-53-6; PIGMENT GREEN 7; C.I. 74260), and also MACROLEX GREEN G (CAS 28198-05-2; SOLVENT GREEN 28; C.I. 625580). If necessary, a filler can also be incorporated into the colorant composition.

[0078] Suitable fillers include carbonates, sulfates, silicates, and oxides. For example, magnesium, calcium and barium carbonate, calcium and barium sulfate, silicates and

aluminosilicates and aluminum, titanium, and silicon oxides are well suited. Mixtures of these compounds can also be used.

Preparation of the compositions of the invention

[0079] Thermoplastic compositions are prepared from the components A, B, C, and optionally D, and E of the invention.

[0080] The thermoplastic molding compositions of the invention can be prepared, for example, by mixing the respective components of the compositions in a known manner and melt-compounded at temperatures of preferably 200 ° C to 320 ° C, more preferably at 240 to 310 ° C, most preferably at 260 ° C to 300 ° C in conventional aggregates such as internal kneaders, extruders and twin-shaft screws melt compounded and melt extruded. This process is commonly referred to as compounding in the context of this application.

[0081] The mixing of the individual components of the compositions can be carried out in a known manner both successively and simultaneously, both at about 20 ° C (room temperature) and at higher temperature. This means, for example, that some of the components can be dosed via the main feed of an extruder and the remaining components can later be fed via a side extruder in the compounding process.

EXAMPLES

[0082] The non-limiting and non-exhaustive examples that follow are intended to further describe various non-limiting and non-exhaustive embodiments without restricting the scope of the embodiments described in this specification. All quantities given in “parts” and “percents” are understood to be by weight, unless otherwise indicated.

[0083] The following materials were used in preparation of the Examples:

Component	Description
ABS-A	acrylonitrile-butadiene-styrene commercially available from Trinseo PLC as MAGNUM 3404;
ADDITIVE A	octadecyl-3-[3,5-di-tert-butyl-4-hydroxyphenyl]propionate], commercially available from BASF as IRGANOX 1076 (Component C.2);
ADDITIVE B	a blend of 80% IRGAFOS 168 and 20% IRGANOX 1076 (Component C.2) commercially available from BASF as IRGANOX B 900;
ADDITIVE C	tris (2-ethylhexyl) phosphate, commercially available from Lanxess as DISFLAMOLL TOF (Component C.1);
MOLD RELEASE AGENT	a long-chain ester of pentaerythritol, available from Emery Oleochemicals as LOXIOL HOB-7119;

DYE-A	an anthraquinone dyestuff, blue with a red cast powder, commercially available from Lanxess as MACROLEX BLUE RR;
DYE-B	an anthraquinone dyestuff, violet with a red cast powder, commercially available from Lanxess as MACROLEX VIOLET 3R;
PC-A	a linear polycarbonate based on bisphenol A with a melt volume-flow rate MVR according to ISO 1133-1:2011 of 12 cm ³ /(10 min), measured at 300 °C under a 1.2 kg load, commercially available from Covestro AG as MAKROLON 2608-1000; and
UV LIGHT ABSORBER	2-phenol, 2-(2H-benzotriazol-2-yl)-4-(1, 1, 3, 3-tetramethylbutyl), commercially available from BASF as TINUVIN 329 (Component C.3).

[0084] Production of samples for the PC/ABS blends was carried out using a ZSK25 twin-screw extruder from Coperion, Werner & Pfleiderer (Stuttgart, Germany) at a melt temperature range of 260°C/280°C and an extrusion rate of 16 -32 Kgs/hr.

[0085] Molded articles (plaques of dimension 4 in x 6 in) were produced at a melt temperature of 296 °C and a mold temperature of 88°C) in a MILACRON ROBOSHOT α -S110 iA or MILACRON ROBOSHOT SiB 110 injection molding machine with the residence time (hold time) in the melt at the melt temperature varied between instantaneous (< 1 min) to 10 min, using the following hold times in the melt <1 min, 2.5 min, 5 min, 7 min, and 10 min.

[0086] The Yellowness Index (YI) was measured in reflection on the molded plaques according to ASTM E313.

GPC analysis

[0087] The samples were prepared in THF at 5 mg/mL and analyzed via Waters 2690 Separations Module equipped with a Waters 2414 RI Detector. The samples were processed to evaluate the molecular weight distribution (MWD) based on a universal calibration curve from poly(styrene) standards.

Instrument Conditions

HPLC: Waters 2695 Alliance (GPC Mode)

Solvent: THF (BHT-stabilized)

Temperature (Column and Detector): 30 °C

Flow Standard: Toluene

Detection: Waters 2414 Refractive Index Detector

Operating Pressure: 780 psi

Injection Volume: 75 μ L

Table I

	EX. 1	EX. 2	EX. 3	EX. 4	EX. 5	EX.C6
PC-A	68.92	68.88	69.00	68.88	68.96	68.93
MOLD RELEASE AGENT	0.50	0.50	0.50	0.50	0.50	0.50
ADDITIVE A	0.20	0.20	0.10	0.20	0.10	0.20
ABS-A	29.54	29.53	29.56	29.52	29.55	29.54
ADDITIVE B	0.10	0.10	0.10	0.10	0.10	0.10
UV LIGHT ABSORBER	0.75	0.75	0.75	0.75	0.75	0.75
ADDITIVE C	0.01	0.03	0.01	0.05	0.05	0.00
DYE-B	0.000071	0.000071	0.000071	0.000071	0.000071	0.000071
DYE-A	0.000079	0.000079	0.000079	0.000079	0.000079	0.000079
Time in the melt (minutes)	YI value					
<1 min	-6.86	-3.49	-4.1	-2.19	-3.48	-4.52
2.5	-4.16	-1.28	-2.53	0.74	-0.81	5.54
5	-3.13	-0.1	-0.5	3.47	3.37	16.78
7	-1.1	2.09	1.06	5.52	6.11	23.07
10	0.87	4.7	3.04	7.8	8.83	23.81

Table II

Sample without stabilization package (EX.C6)	Time in the melt	YI value	Mn	MP	Mw	Mz
Cell 1	5 min	43.35	7930 (30)	18720 (10)	35210 (20)	133210 (70)
Cell 4	INST (0 min)	8.32	8520 (10)	20870 (20)	38530 (60)	143050 (240)
Pellets			9760 (10)	27760 (30)	43930 (10)	138070 (130)

Molding was done using production tool for automotive part.

Table III

Sample	Mn	Mp	Mw	Mz	Mw/Mn	Mn retention (%)	Mw retention (%)
Pellets from material of EX C6	9760 (10)	27760 (30)	43930 (10)	138070 (130)	4.50		
Molded part from material of C6, INST (<1 min in melt).	8520 (10)	20870 (20)	38530 (60)	143050 (240)	4.52	87.3	87.7
Molded part from material of C6, (5 min in the melt)	7930 (30)	18720 (10)	35210 (20)	133210 (70)	4.44	81.3	80.2
Pellets from material of EX. 1	8609 (13)	28809 (19)	43629 (3)	131175 (12)	5.07 (0.01)		

Molded part from material of EX. 1, INST (<1 min in melt)	8508 (6)	28465 (33)	43080 (24)	130209 (3)	5.06 (0.01)	98.8	98.7
Molded part from material of EX. 1, (10 min in the melt)	8356 (2)	27661 (25)	42829 (32)	130322 (260)	5.13 (0)	97.1	98.2

*Standard deviations, provided in (), are calculated from the processing of two separate injections.

Table IV

Sample	Mn	Mp	Mw	Mz	Mw/ Mn	Mn retention (%)	Mw retention (%)
Pellets from material of EX. 1	10020 (5)	28809 (19)	43876 (2)	131181 (12)	4.38 (0)		
Molded part INST (<1 min in melt)	9909 (4)	28465 (33)	43326 (25)	130215 (3)	4.37 (0)	98.9	98.7
Molded part from material of EX. 1, INST (10 min in melt)	9719 (37)	27661 (25)	43075 (25)	130328 (260)	4.43 (0.02)	97.0	98.2

[0088] To provide a viable comparison of the samples containing the new stabilization package (pellets, dotted line) to samples that do not contain the new stabilization package (pellets, dashed line), an integration line was drawn to remove the peak associated with the stabilization package (portion shown between the triangles). This allows the comparison of the poly(carbonate) portion, without skewing MWD information by including the stabilization package peak.

[0089] A low absolute value coupled with a small increase in the YI (Yellowness Index) as a function of time in the melt is an indication of good thermal stability. Also included are the results of YI value increase and corresponding Mw reduction for the same formulation without stabilization package.

[0090] This specification has been written with reference to various non-limiting and non-exhaustive embodiments. However, it will be recognized by persons having ordinary skill in the art that various substitutions, modifications, or combinations of any of the disclosed embodiments (or portions thereof) may be made within the scope of this specification. Thus, it is contemplated and understood that this specification supports additional embodiments not expressly set forth herein. Such embodiments may be obtained,

for example, by combining, modifying, or reorganizing any of the disclosed steps, components, elements, features, aspects, characteristics, limitations, and the like, of the various non-limiting embodiments described in this specification. In this manner, Applicant reserves the right to amend the claims during prosecution to add features as variously described in this specification, and such amendments comply with the requirements of 35 U.S.C. §112(a), and 35 U.S.C. §132(a).

[0091] Various aspects of the subject matter described herein are set out in the following numbered clauses:

[0092] Clause 1. A composition comprising: A) an aromatic polycarbonate or an aromatic polyester carbonate, B) an acrylonitrile/butadiene/styrene polymer, C) a stabilization package comprising C.1) a phosphate ester, preferably an aliphatic phosphate ester included in an amount greater than zero but less than 0.1 wt.%, C.2) a phenolic antioxidant included in an amount greater than zero but less than 0.3 wt.%, and optionally C.3) a UV light absorber, and optionally D) one or more selected from the group consisting of polymer additives, polymeric blending partners different from component B, and optionally E) colorants.

[0093] Clause 2. The composition according to Clause 1, wherein the phosphate ester C.1 is tris (2-ethylhexyl) phosphate.

[0094] Clause 3. The composition according to one of Clauses 1 and 2, wherein the UV light absorber C.3 is a benzotriazole.

[0095] Clause 4. The composition according to Clause 3, wherein the benzotriazole is 2-phenol, 2-(2H-benzotriazol-2-yl)-4-(1, 1, 3, 3-tetramethylbutyl).

[0096] Clause 5. The composition according to any one of Clauses 1 to 4, wherein the phenolic antioxidant C.2 is octadecyl-3-[3,5-di-tert-butyl-4-hydroxyphenyl]propionate].

[0097] Clause 6. The composition according to any one of Clauses 1 to 5, wherein at least one blue or violet dyestuff is included in an amount effective to compensate for a light-yellow natural tint the composition displays in the absence of such dyestuff.

[0098] Clause 7. The composition according to Clause 6, wherein the at least one blue or violet dyestuff is at least one anthraquinone dyestuff.

[0099] Clause 8. The composition according to any one of Clauses 1 to 7, wherein the preferably aliphatic phosphate ester C.1 is included in an amount of from 0.005 wt.% to 0.08 wt.%.

[0100] Clause 9. The composition according to any one of Clauses 1 to 8, wherein the preferably aliphatic phosphate ester C.1 is included in an amount of from 0.01 wt.% to 0.03 wt.%.

[0101] Clause 10. The composition according to any one of Clauses 1 to 9, wherein the benzotriazole UV absorber C.3 is included in an amount greater than zero but less than 1 wt.%.

[0102] Clause 11. The composition according to any one of Clauses 1 to 10, wherein the acrylonitrile/butadiene/styrene polymer is produced in a bulk polymerization process.

[0103] Clause 12. The composition according to any one of Clauses 1 to 11, wherein the acrylonitrile/butadiene/styrene polymer is an ABS polymer that is free of emulsifiers selected from the group consisting of saturated fatty acids having 8 to 22 carbon atoms, resin acids, alkyl and alkylarylsulfonic acids, and fatty alcohol sulfates.

[0104] Clause 13: The composition according to any one of Clauses 1 to 12, wherein the acrylonitrile/butadiene/styrene polymer contains less than 100 ppm of alkali metals and alkaline earth metals.

[0105] Clause 14: The composition according to any one of Clauses 1 to 13, wherein the acrylonitrile/butadiene/styrene polymer contains greater than zero but less than 10 ppm of lithium.

[0106] Clause 15: The composition according to any one of Clauses 1 to 14, wherein C.1 is included in an amount of from 0.01 wt.% to 0.03 wt.%.

What is claimed is:

1. A composition comprising:
 - A) an aromatic polycarbonate or an aromatic polyester carbonate,
 - B) an acrylonitrile/butadiene/styrene polymer,
 - C) a stabilization package comprising
 - C.1) a phosphate ester, preferably an aliphatic phosphate ester included in an amount greater than zero but less than 0.1 wt.%,
 - C.2) a phenolic antioxidant included in an amount greater than zero but less than 0.3 wt.%, and optionally
 - C.3) a UV light absorber, and optionally
 - D) one or more selected from the group consisting of polymer additives, polymeric blending partners different from component B, and optionally
 - E) colorants.
2. The composition according to claim 1, wherein the phosphate ester C.1 is tris (2-ethylhexyl) phosphate.
3. The composition according to one of claims 1 and 2, wherein the UV light absorber C.3 is a benzotriazole.
4. The composition according to claim 3, wherein the benzotriazole is 2-phenol, 2-(2H-benzotriazol-2-yl)-4-(1, 1, 3, 3-tetramethylbutyl).
5. The composition according to any one of claims 1 to 4, wherein the phenolic antioxidant C.2 is octadecyl-3-[3,5-di-tert-butyl-4-hydroxyphenyl]propionate].
6. The composition according to any one of claims 1 to 5, wherein at least one blue or violet dyestuff is included in an amount effective to compensate for a light-yellow natural tint the composition displays in the absence of such dyestuff.
7. The composition according to claim 6, wherein the at least one blue or violet dyestuff is at least one anthraquinone dyestuff.

8. The composition according to any one of claims 1 to 7, wherein the preferably aliphatic phosphate ester C.1 is included in an amount of from 0.005 wt.% to 0.08 wt.%.
9. The composition according to any one of claims 1 to 8, wherein the preferably aliphatic phosphate ester C.1 is included in an amount of from 0.01 wt.% to 0.03 wt.%.
10. The composition according to any one of claims 1 to 9, wherein the benzotriazole UV absorber C.3 is included in an amount greater than zero but less than 1 wt.%.
11. The composition according to any one of claims 1 to 10, wherein the acrylonitrile/butadiene/styrene polymer is produced in a bulk polymerization process.
12. The composition according to any one of claims 1 to 11, wherein the acrylonitrile/butadiene/styrene polymer is an ABS polymer that is free of emulsifiers selected from the group consisting of saturated fatty acids having 8 to 22 carbon atoms, resin acids, alkyl and alkylarylsulfonic acids, and fatty alcohol sulfates.
13. The composition according to any one of claims 1 to 12, wherein the acrylonitrile/butadiene/styrene polymer contains less than 100 ppm of alkali metals and alkaline earth metals.
14. The composition according to any one of claims 1 to 13, wherein the acrylonitrile/butadiene/styrene polymer contains greater than zero but less than 10 ppm of lithium.
15. The composition according to any one of claims 1 to 14, wherein C.1 is included in an amount of from 0.01 wt.% to 0.03 wt.%.

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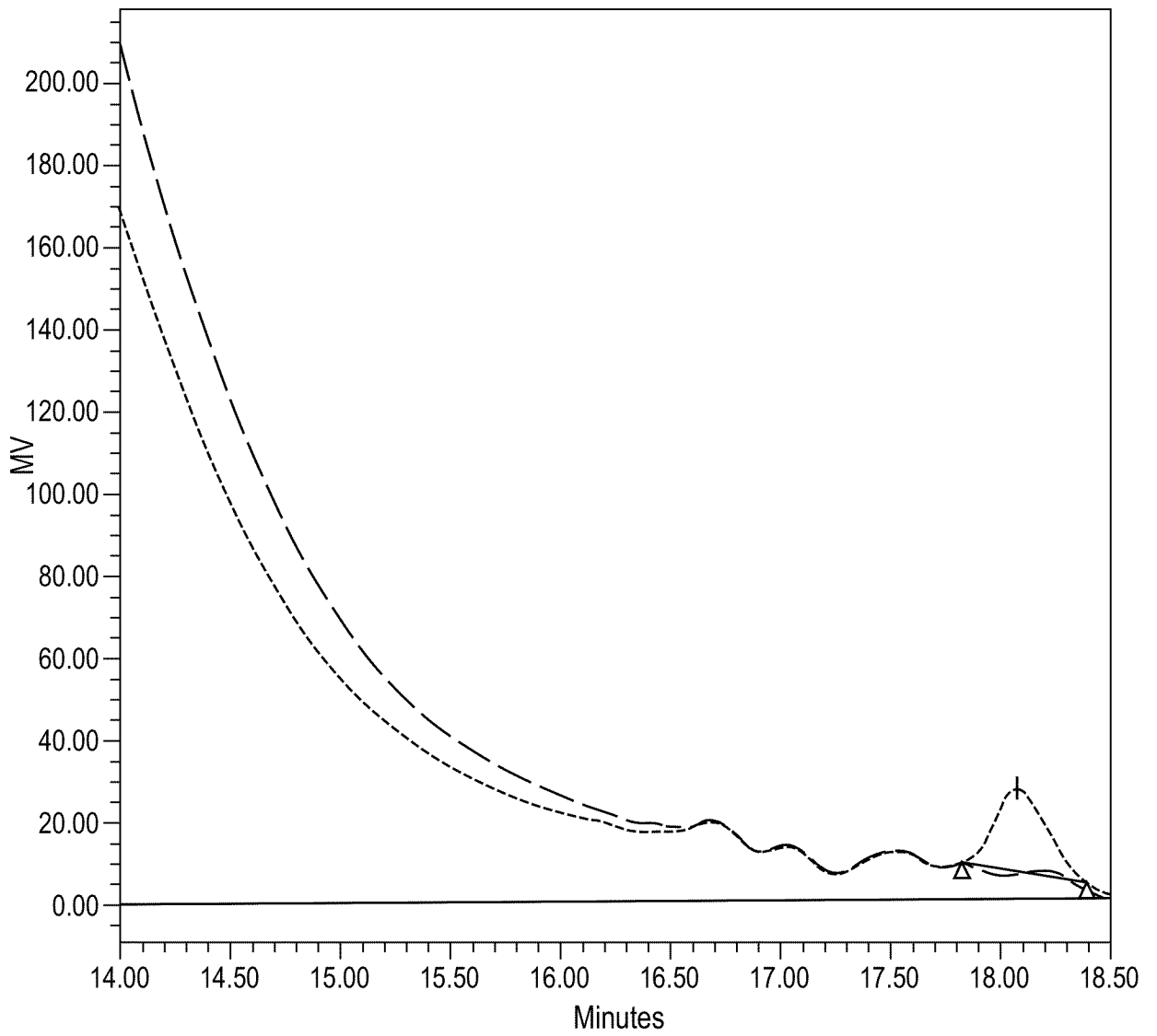


FIG. 1

INTERNATIONAL SEARCH REPORT

International application No
PCT/US2024/021159

A. CLASSIFICATION OF SUBJECT MATTER
INV. C08L69/00
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)
C09J 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 practicable, search terms used)

EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2014/115475 A1 (MITSUBISHI ENG PLASTICS CORP [JP]) 31 July 2014 (2014-07-31) table 2 -----	1, 5, 12-14
X	US 2009/281216 A1 (AVTOMONOV EVGUENI [DE] ET AL) 12 November 2009 (2009-11-12) table 3 -----	1, 5, 12-14
X	EP 2 657 298 A1 (BAYER MATERIALSCIENCE AG [DE]) 30 October 2013 (2013-10-30) table 2 -----	1, 5, 12-14
X	JP 2014 184720 A (MITSUBISHI ENG PLASTICS CORP) 2 October 2014 (2014-10-02) paragraphs [0162], [0173], [0278]; example 5 and 6 -----	1-15

Further documents are listed in the continuation of Box C.

See patent family annex.

* Special categories of cited documents :

- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier application or patent but published on or after the international filing date
- "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

Date of mailing of the international search report

13 June 2024

28/06/2024

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/US2024/021159

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