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Compositions based on shock resistance-modified polyethyleneterephthalate/polycarbonate blends

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Zur Erklärung der Zweibuchstaben-Codes, und der anderen Abkürzungen wird auf die Erklärungen ("Guidance Notes on Codes and Abbreviations") am Anfang jeder regulären Ausgabe der PCT-Gazette verwiesen.

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(54) **Title:** COMPOSITIONS BASED ON SHOCK RESISTANCE-MODIFIED POLYETHYLENETEREPHTHALATE/POLYCARBONATE BLENDS

(54) **Bezeichnung:** ZUSAMMENSETZUNGEN AUF BASIS SCHLAGZÄHMODIFIZIERTEN POLYETHYLENTEREPHTHALAT/POLYCARBONAT BLENDS

(57) **Abstract:** The invention relates to compositions which are based on shock-resistance-modified polyethyleneterephthalate/polycarbonate blends and which contain a mineral filler in the form of talc. The invention also relates to the use of thermoplastic moulding materials for producing semi-finished products and moulded parts and to semi-finished products and moulded parts produced from these thermoplastic moulding materials.

(57) **Zusammenfassung:** Die vorliegende Erfindung betrifft Zusammensetzungen auf Basis schlagzähmodifizierten Polyethylenterephthalat/Polycarbonat-Blends die als mineralischen Füllstoff Talk enthalten. Darüber hinaus betrifft die vorliegende Erfindung die Verwendung der thermoplastischen Formmassen zur Herstellung von Halbzeugen und Formteilen, sowie die aus den thermoplastischen Formmassen erhältlichen Halbzeuge und Formteile.

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Compositions based on impact-resistant modified polyethylene terephthalate/polycarbonate blends

The present invention relates to compositions based on impact-resistant modified polyethylene terephthalate/polycarbonate blends which contain talc as a mineral filler. The present invention further relates to the use of these thermoplastic moulding compositions for the production of semi-finished products and mouldings, and also relates to the semi-finished products and mouldings which can be obtained from said thermoplastic moulding compositions.

Polycarbonate moulding compositions are known which contain fillers and which contain partially crystalline polyesters, graft copolymers and mineral fillers. Moulding compositions of this type are used in the automobile sector, for example.

DE-A 19 753 541 discloses polycarbonate moulding compositions which contain partially aromatic polyesters, graft copolymers and mineral fillers, and which exhibit a toughness which is satisfactory for exterior bodywork parts. However, these claimed moulding compositions exhibit unsatisfactory dimensional stability when hot.

EP-A 135 904 describes polycarbonate moulding compositions which contain polyethylene terephthalate, graft copolymers based on polybutadiene, and talc in amounts of up to 4 % by weight. The advantage thereof is stated to be a favourable combination of properties comprising low warpage and good toughness.

JP-A 08 176 339 describes polycarbonate moulding compositions which contain talc as a mineral filler. ABS resins, polyethylene terephthalate and polybutylene terephthalate can be used as other components of the blend. The advantages of these moulding compositions which are emphasised are their good impact toughness and surface quality.

JP-A 07 025 241 describes polycarbonate moulding compositions which exhibit high stiffness and good surface quality. These moulding compositions contain 60 to 70 % by weight polycarbonate, 20 to 30 % by weight polyester, 5 to 10 % by weight acrylate rubber and 5 to 10 % by weight talc, as well as 0.1 to 1 parts by weight of antioxidant (with respect to 100 parts of polymer components).

JP-A 63 132 961 discloses polycarbonate moulding compositions, which contain polybutylene terephthalate, polyesters, graft copolymers and mineral fillers, for use in the automobile sector.

Exterior bodywork parts made of plastics generally have to be lacquer-coated. In the case of plastics which are pigmented with the colour of the vehicle, bodywork parts produced therefrom are generally overcoated with one or more coats of transparent lacquer. In the case of plastics which are not pigmented with the colour of the vehicle, bodywork parts produced therefrom are generally coated with a plurality of lacquer coats, wherein at least one of these coats is a colour-imparting coat. The lacquer coats applied generally have to be stoved at an elevated temperature, which can range up to 200°C, and cured. The temperature and duration of stoving depend on the lacquer system used. The plastics material used for bodywork components has to undergo as few changes as possible, such as irreversible deformations for example, during the curing or stoving processes. It is therefore necessary to provide thermoplastic polycarbonate moulding compositions which exhibit improved dimensional stability when hot.

Other requirements which are imposed on bodywork components made of plastics include good toughness properties under impact and tensile loading, particularly at low temperatures also, satisfactory stiffness, low thermal expansion, good surface quality, a good capacity for being lacquer-coated with good lacquer adhesion, and good resistance to chemicals and fuels. In addition, moulding compositions used for the production of exterior bodywork parts have to exhibit good flowability in the molten state.

Practical experience has shown that, depending on the specific area of use, materials which are used for bodywork components can exhibit large variations in the properties listed above. The factor which is ultimately decisive and which is very important for all materials, however, is satisfactory dimensional stability when hot, in order to enable lacquer coating to be effected without problems.

The object of the present invention was therefore to provide polycarbonate moulding compositions which exhibit excellent dimensional stability when hot and an excellent capacity for being lacquer-coated. In addition, the compositions according to the invention unexpectedly exhibit only a slight decrease in impact toughness at low temperatures. Moreover, the polycarbonate moulding compositions should exhibit an excellent surface property profile for bodywork components made of plastics with respect to the requirements mentioned above. Furthermore, the polycarbonate moulding compositions should readily be capable of being processed to form bulky mouldings which are suitable for use as bodywork components.

It has now been found that compositions containing polyethylene terephthalate in combination with polycarbonate, impact toughness modifiers and talc as a mineral filler, possess the requisite properties.

The present invention relates to compositions containing

A) 4 to 80, preferably more than 10 to 60, most preferably 12 to 40, particularly 19 to 29 parts by weight, of at least one polyethylene terephthalate,

B) 10 to 90, preferably 20 to 80, most preferably 25 to 55, particularly 30 to 50 parts by weight, of at least one aromatic polycarbonate,

C) 1.5 to 30, preferably 3 to 25, most preferably 6 to 20, particularly 8 to 17 parts by weight, of at least one graft polymer comprising a graft base based on acrylates with a glass transition temperature less than -5°C or at least one elastomeric block copolymer, particularly two- or three block copolymers based on aromatic vinyl compounds and dienes or mixtures thereof, and block copolymers, and

D) 1.5 to 54, preferably 3 to 34, most preferably 6 to 25, particularly 8 to 21 parts by weight, of at least one mineral filler based on talc,

wherein the sum of the parts by weight of all the components is 100.

As now claimed, according to one aspect, the present invention provides a composition comprising

A) 4 to 80 parts by weight of at least one polyethylene terephthalate,

B) 10 to 90 parts by weight of at least one aromatic polycarbonate,

C) 1.5 to 30 parts by weight of a mixture of at least one graft polymer comprising a graft base based on acrylates with a glass transition temperature of less than –5°C and or at least one elastomeric block copolymer or mixtures of graft copolymers and block copolymers, and

D) 1.5 to 54 parts by weight at least one mineral filler based on talc,

wherein the sum of the parts by weight of all the components is 100 and wherein component D) has an upper particle size d_{97} less than 50μm.

According to the invention, the compositions contain, as **component A**, one or a mixture of two or more different polyethylene terephthalates. Polyethylene terephthalates in the sense of the invention are polyalkylene terephthalates which are derived from terephthalic acid (or reactive derivatives thereof) and alkanediols based on ethylene glycol.

The preferred polyethylene terephthalates (hereinafter also abbreviated to PETs) can be produced by known methods from terephthalic acid (or reactive derivatives thereof) and aliphatic or cycloaliphatic diols comprising an ethylene glycol unit (Kunststoff-Handbuch, Volume VIII, pages 695 et seq., Karl-Hanser-Verlag, Munich 1973).

The preferred polyethylene terephthalates contain at least 80, preferably 90 mol %, with respect to the dicarboxylic acid, of terephthalic acid radicals and at least 80, preferably at least 90 mol % with respect to the diol component, of ethylene glycol radicals.

In addition to terephthalic acid radicals, the preferred polyethylene terephthalates can contain up to 20 mol % of radicals of other aromatic dicarboxylic acids comprising 8 to 14 C atoms or aliphatic dicarboxylic acids comprising 4 to 12 C atoms, preferably phthalic acid, isophthalic acid, naphthalene-2,6-dicarboxylic acid, 4,4'-diphenyldicarboxylic acid, succinic, adipic, sebacic or azelaic acids, or cyclohexanediacetic acid.

In addition to ethylene glycol, the preferred polyethylene terephthalates can contain up to 20 mol % of other aliphatic diols comprising 3 to 12 C atoms or cycloaliphatic diols comprising 6 to 21 C atoms, e.g. 1,3-propanediol, 2-ethylpropandiol-1,3, neopentyl glycol, 1,5-pentanediol, 1,6-hexanediol, cyclohexane-dimethanol-1,4, 3-methylpentanediol-2,4, 2-methylpentanediol-2,4, 2,2,4-trimethylpentanediol-1,3 and - 1,6,2-ethylhexanediol-1,3 2,2-diethylpropanediol-1,3, 2,5-hexanediol, 1,4-di-(β-hydroxyethoxy)-benzene, 2,2-bis-(4-hydroxycyclohexyl)-propane 2,4-dihydroxy-1,1,3,3-tetramethyl-cyclobutane, 2,2-bis-3-β-hydroxyethoxyphenyl)-propane and 2,2-bis-(4-hydroxypropoxyphenyl)-propane (DE-A 24 07 674, 24 07 776, 27 15 932). Moreover, the polyethylene terephthalates can also contain up to 20 mol % of ether or polyether structures.

The polyethylene terephthalates can be branched by the incorporation of relatively small amounts of tri- or tetrahydric alcohols or of a tri- or tetrabasic carboxylic acid, such as those which are described in DE-A 19 00 270 and in US-A 3 692 744, for example. Examples of preferred branching agents include trimesic acid, trimellitic acid, trimethylolethane and -propane and pentaerythritol. It is advisable not to use more than 1 mol % of the branching agent with respect to the acid component.

Preferred polyethylene terephthalates also include copolyesters which are produced from at least two acid components and/or from at least two alcohol components. The most preferred copolyesters are poly-(ethylene glycol/1,4-butanediol) terephthalates.

Polyethylene terephthalates which have been produced solely from terephthalic acid or reactive derivatives thereof (e.g. dialkyl esters thereof) and ethylene glycol are particularly preferred.

The polyethylene terephthalates generally have an intrinsic viscosity of about 0.3 to 1.5 dl/g, preferably 0.4 to 1,3 dl/g, most preferably 0.5 to 0.8 dl/g, as measured in phenol/o-dichlorbenzene (1:1 parts by weight) at 25°C in each case.

Polyethylene terephthalates which crystallise rapidly are particularly preferred, i.e. polyethylene terephthalates which exhibit times of crystallisation, as measured by the DSC method for isothermal crystallisation at 215°C, which are generally less than 15 minutes, preferably less than 10 minutes and most preferably less than 5 minutes.

Rapid crystallisation of the polyethylene terephthalates according to the invention is preferably achieved by the addition of crystallisation agents to the polyethylene terephthalates during or following the production thereof, e.g. by admixing them with the polyethylene terephthalate melt. Metal salts of organic carboxylic acids are preferably used as crystallisation agents, such as alkali or alkaline earth metal salts of benzoic acid or of substituted benzoic acids.

Part of the polyethylene terephthalate can be replaced by other thermoplastic polyesters, preferably by polybutylene terephthalates. In general, up to 50 % by weight, preferably up to 10 % by weight (with respect to the polyethylene terephthalate) of the polyethylene terephthalate can be replaced by other thermoplastic polyesters, preferably by polyalkylene terephthalates.

Thermoplastic polyesters are the reaction products of aromatic dicarboxylic acids or reactive derivatives thereof (e.g. dimethyl esters or anhydrides) with aliphatic, cycloaliphatic or araliphatic diols, and mixtures of these reaction products.

Other thermoplastic polyesters which are preferred include polyalkylene terephthalates which can be produced by known methods from terephthalic acid (or reactive derivatives thereof) and aliphatic or cycloaliphatic diols comprising 3 to 10 C atoms (Kunststoff-Handbuch, Volume VIII, page 695 et seq., Karl-HanserVerlag, Munich 1973).

The preferred polyalkylene terephthalates contain at least 80, preferably 90 mol % with respect to the dicarboxylic acid, of terephthalic acid radicals and at least 80, preferably at least 90 mol % with respect to the diol component, of 1,4-butanediol radicals.

In addition to terephthalic acid radicals, the preferred polyalkylene terephthalates can contain up to 20 mol % of radicals of other aromatic dicarboxylic acids comprising 8 to 14 C atoms or aliphatic dicarboxylic acids comprising 4 to 12 C atoms, such as radicals of phthalic acid, isophthalic acid, naphthalene-2,6-dicarboxylic acid, 4,4'-diphenyldicarboxylic acid, succinic, adipic, sebacic acid or azelaic acid, and cyclohexane-diacetic acid.

In addition to 1,4-butanediol glycol radicals, the preferred polyalkylene terephthalates can contain up to 20 mol % of other aliphatic diols comprising 3 to 12 C atoms or cycloaliphatic diols comprising 6 to 21 C atoms, e.g. radicals of 1,3-propanediol, 2-ethylpropanediol-1,3, neopentyl glycol, 1,5-pentanediol, 1,6-hexanediol, cyclohexane-dimethanol-1,4, 3-methylpentanediol-2,4, 2-methylpentanediol-2,4, 2,2,4-tri-methylpentanediol-1,3 and -1,6,2-ethylhexanediol-1,3, 2,2-diethylpropanediol-1,3, 2,5-hexanediol, 1,4-di-(β-hydroxyethoxy)-benzol, 2,2-bis-(4-hydroxycyclohexyl)-propane 2,4-dihydroxy-1,1,3,3-tetramethylcyclobutane, 2,2-bis-(3-β-hydroxyethoxy-phenyl)-propane and 2,2-bis-(4-hydroxypropoxyphenyl)-propane (DE-A 24 07 674, 24 07 776, 27 15 932).

As described above, the polyalkylene terephthalates can also be branched by the incorporation of relatively small amounts of tri- or tetrahydric alcohols or tri- or tetrabasic carboxylic acids.

Polyethylene terephthalates which have been produced solely from terephthalic acid or reactive derivatives thereof (e.g. dialkyl esters thereof) and 1,4-butanediol are particularly preferred (polybutylene terephthalates).

The preferred polyalkylene terephthalates also include copolyesters which are produced from at least two of the aforementioned acid components and/or from at least two of the aforementioned alcohol components.

The polyethylene terephthalates generally have an intrinsic viscosity of about 0.3 to 1.5 dl/g, preferably 0.4 to 1.3 dl/g, as measured in phenol/o-dichlorobenzene (1:1 parts by weight) at 25°C in each case.

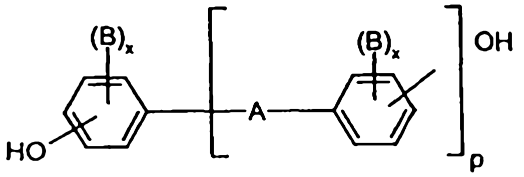
According to the invention, the compositions according to the invention contain a polycarbonate or a mixture of polycarbonates as **component B**.

The preferred polycarbonates are those homopolycarbonates and copolycarbonates which are based on bisphenols of general formula (I):

$$HO\text{-}Z\text{-}OH \qquad (I)$$

wherein Z is a divalent organic radical which comprises 6 to 30 C atoms and which contains one or more aromatic groups.

Bisphenols of formula (Ia) are preferred

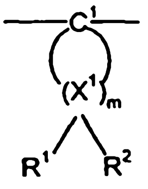


(Ia),

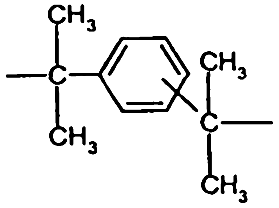
wherein

A denotes a single bond, a C_1-C_5-alkylene, a C_2-C_5-alkylidene or a C_5-C_6-cycloalkylidene group, or -O-, -SO-, -CO-, -S-, $-SO_2-$, or a C_6-C_{12}-arylene group, on to which other aromatic rings, which optionally contain hetero atoms, can be condensed,

or denotes a radical of formulae (II) or (III)



(II)



(III)

B in each case denotes a C_1-C_{12}-alkyl, preferably methyl, or a halogen, preferably chlorine and/or bromine

x denotes 0, 1 or 2, which are independent of each other in each case,

p is 1 or 0, and

R^1 and R^2 can be selected individually for each X^1, are independent of each other, and denote hydrogen or a C_1-C_6-alkyl, preferably hydrogen, methyl or ethyl,

X^1 denotes carbon, and

m denotes an integer from 4 to 7, preferably 4 or 5, with the proviso that on at least one X^1 atom R^1 and R^2 simultaneously denote an alkyl group.

Examples of bisphenols corresponding to general formula (I) include bisphenols which belong to the following groups: dihydroxydiphenyls, bis-(hydroxyphenyl)-alkanes, bis-(hydroxyphenyl)-cycloalkanes, indane bisphenols, bis-(hydroxyphenyl) sulphides, bis-(hydroxyphenyl) ethers, bis-(hydroxyphenyl)-ketones, bis-(hydroxyphenyl) sulphones, bis-(hydroxyphenyl) sulphoxides and α,α'-bis-(hydroxyphenyl)-diisopropylbenzenes.

Examples of bisphenols corresponding to general formula (I) also include derivatives of said bisphenols, which can be obtained, for example, by alkylation or halogenation of the aromatic rings of said bisphenols.

Examples of bisphenols corresponding to general formula (I) include the following compounds in particular: hydroquinone, resorcinol, 4,4'-dihydroxydiphenyl, bis-(4 hydroxyphenyl) sulphide, bis-(4-hydroxyphenyl) sulphone, bis-(3,5-dimethyl-4-hydroxy-phenyl)-methane, bis-(3,5-dimethyl-4-hydroxyphenyl) sulphone, 1,1-bis-(3,5-dimethyl-4-hydroxyphenyl)-p/m-diisopropylbenzene, 1,1-bis-(4-hydroxyphenyl)-1-phenylethane, 1,1-bis-(3,5-dimethyl-4-hydroxyphenyl)-cyclohexane, 1,1-bis-(4-hydroxy-phenyl)-3-methylcyclohexane, 1,1-bis-(4-hydroxyphenyl)4-methylcyclohexane, 1,1-bis-(-hydroxyphenyl)-cyclohexane, 1,1-bis-(4-hydroxyphenyl)-3,3,5-trimethylcyclohexane, 2,2-bis-(3,5-dichloro-hydroxyphenyl)-propane, 2,2-bis-(3-methyl-4-hydroxyphenyl)-propane, 2,2-bis-(3,5-dimethyl-4-hydroxyphenyl)-propane, 2,2-bis-(4-hydroxyphenyl)-propane (i.e. bisphenol A), 2,2-bis-(3-chloro-4-hydroxy-phenyl)-propane, 2,2-bis-(3,5-dibromo-4-hydroxyphenyl)-propane, 2,4-bis-(4-hydroxyphenyl)-2-methylbutane, 2,4-bis-(3,5-dimethyl-4-hydroxyphenyl)-2-methyl-butane, α,α′-bis-(4-hydroxyphenyl)-m-diisopropylbenzene, (i.e. bisphenol M), α,α′-bis-(4-hydroxyphenyl)-p-diisopropylbenzene, and indane-bisphenol.

Polycarbonates which are particularly preferred are the homopolycarbonate based on bisphenol A, the homopolycarbonate based on 1,1-bis-(4-hydroxyphenyl)-3,3,5-trimethylcyclohexane and the copolycarbonates based on the two monomers bisphenol A and 1,1-bis-(4-hydroxyphenyl)-3,3,5-trimethylcyclohexane.

The bisphenols corresponding to general formula (I) which were described above can be produced by known methods, e.g. from the corresponding phenols and ketones.

The aforementioned bisphenols and methods of producing them are described, for example, in the monograph by H. Schnell, "Chemistry and Physics of Polycarbonates", Polymer Reviews, Volume 9, pages 77-98, Interscience Publishers, New York, London, Sydney, 1964, and in US-A 3 028 635, in US-A 3 062 781, in US-A 2 999 835, in US-A 3 148 172, in US-A 2 991 273, in US-A 3 271 367, in US-A 4 982 014, in US-A 2 999 846, in DE-A 1 570 703, in DE-A 2 063 050, in DE-A 2 036 052, in DE-A 2 211 956, in DE-A 3 832 396, and in FR-A 1 561 518, as well as in Japanese Patents laid open to public inspection with the Application Numbers 62039/1986, 62040/1986 and 105550/1986.

1,1-bis-(4-hydroxyphenyl)-3,3,5-trimethylcyclohexane and the production thereof is described, for example, in US-A 4 982 014.

Indane-bisphenols and the production thereof are described, for example, in US-A 3 288 864, in JP-A 60 035 150 and in US-A 4 334 106. Indane-bisphenols can be produced, for example, from isopropenylphenol or from derivatives thereof, or from dimers of isopropenylphenol or derivatives thereof, in the presence of a Friedel-Craft catalyst in organic solvents.

Polycarbonates can be produced by known methods. Examples of suitable methods for the production of polycarbonates include the production thereof from bisphenols with phosgene by the phase boundary method or the production thereof from bisphenols with phosgene by the homogeneous phase method, which is termed the pyridine method, or the production thereof from bisphenols with esters of carbonic acid by the transesterification method in the melt. These methods of production are described by H. Schnell, "Chemistry and Physics of Polycarbonates", Polymer Reviews, Volume 9, pages 31-76, Interscience Publishers, New York, London, Sydney, 1964. The aforementioned methods of production are also described by D. Freitag, U. Grigo, P. R. Müller, H. Nouvertne in "Polycarbonates" in the Encyclopedia of Polymer Science and Engineering, Volume 11, Second Edition, 1988, pages 648 to 718, and by U. Grigo, K. Kircher and P.R. Müller in "Polycarbonates" in Becker, Braun, Kunststoff-Handbuch, Volume 3/1, Polycarbonates, Polyacetals, Polyesters, Cellulose Esters, Carl Hanser Verlag, Munich, Vienna 1992, pages 117 to 299, and by D.C. Prevorsek, B.T. Debona and Y. Kesten, Corporate Research Center, Allied Chemical Corporation, Morristown, New Jersey 07960, in "Synthesis of Poly(ester carbonate) Copolymers" in Journal of Polymer Science, Polymer Chemistry Edition, Vol. 19, 75-90 (1980).

The transesterification method in the melt is described by H. Schnell, "Chemistry and Physics of Polycarbonates", Polymer Reviews, Volume 9, pages 44 to 51, Interscience Publishers, New York, London, Sydney, 1964, and is also described in DE-A 1 031 512, in US-A 3 022 272, in US-A 5 340 905 and in US-A 5 399 659.

Raw materials and adjuvant materials which contain only small amounts of impurities are preferably used for the production of polycarbonates. For the production thereof by the transesterification method in the melt in particular, the bisphenols and carbonic acid derivatives which are used should be as free as possible from alkali ions and alkaline earth ions. Pure raw materials of this kind can be obtained, for example, by recrystallising, washing or distilling the bisphenols and carbonic acid derivatives, such as esters of carbonic acid for example.

Polycarbonates which are suitable according to the invention have a weight average molecular weight (M_w), which can be determined, for example, by ultracentrifugation or by the measurement of scattered light, from 10,000 to 200,000 g/mol. They more preferably have a weight average molecular weight from 12,000 to 80,000 g/mol, most preferably from 20,000 to 35,000 g/mol.

The average molecular weight of the polycarbonates according to the invention can be adjusted in the known manner, for example, by adding a corresponding amount of chain terminators.

The chain terminators can be used individually or as a mixture of different chain terminators.

Suitable chain terminators include both monophenols and monocarboxylic acids. Examples of suitable monophenols include phenol, p-chlorophenol, p-tert.-butyl phenol, cumylphenol or 2,4,6-tribromophenol, as well as long chain alkylphenols such as e.g. 4-(1,1,3,3-tetramethylbutyl)-phenol or monoalkylphenols or dialkylphenols containing a total of 8 to 20 C atoms in their alkyl substituents, such as 3,5-di-tert.-butyl-phenol, p-tert.-octylphenol, p-dodecylphenol, 2-(3,5-dimethyl-heptyl)-phenol or 4-(3,5-dimethyl-heptyl)-phenol. Suitable monocarboxylic acids include benzoic acid, alkylbenzoic acids and halogenobenzoic acids.

The preferred chain terminators are phenol, p-tert.-butyl phenol, 4-(1,1,3,3-tetramethylbutyl)-phenol and cumylphenol.

Thee amount of chain terminators preferably ranges between 0.25 and 10 mol % with respect to the sum of the bisphenols which are used in each case.

Polycarbonates which are suitable according to the invention can be branched in the known manner, preferably by the incorporation of trifunctional branching agents or of branching agents with a functionality greater than three. Examples of suitable branching agents are those which contain three or more than three phenolic groups or those with three or more than three carboxylic acid groups.

Examples of suitable branching agents include phloroglucinol, 4,6-dimethyl-2,4,6-tri-(4-hydroxyphenyl)-heptene-2, 4,6-dimethyl-2,4,6-tri-(4-hydroxyphenyl)-heptane, 1,3,5-tri-(4-hydroxyphenyl)-benzene, 1,1,1-tris-(4-hydroxyphenyl)-ethane, tri-(4-hydroxy-phenyl)-phenylmethane, 2,2-bis-[4,4-bis-(4-hydroxyphenyl)-cyclohexyl]-propane, 2,4-bis-(4-hydroxyphenyl)-isopropyl)-phenol, 2,6-bis-2-hydroxy-5'-methyl-benzyl)-4-methylphenol, 2-(4-hydroxyphenyl)-2-(2,4-dihydroxyphenyl)-propane, hexa-(4-(4-hydroxyphenyl-isopropyl)-phenyl)-terephthalic acid esters, tetra-(4-hydroxyphenyl)-methane, tetra-(4-(-hydroxyphenylisopropyl)-phenoxy)-methane and 1,4-bis-(4',4"-dihydroxytriphenyl)-methylbenzene, as well as 2,4-dihydroxybenzoic acid, trimesic acid, cyanuric chloride, trimesic acid trichloride and α,α',α''-tris-(4-hydroxyphenol)-1,3,5-triisopropylbenzene.

The preferred branching agents are 1,1,1-tris-(4-hydroxyphenyl)-ethane and 3,3-bis-(3-methyl-4-hydroxyphenyl)-2-oxo-2,3-dihydroindole.

The amount of branching agents which are optionally used preferably ranges from 0.05 mol % to 2 mol % with respect to the moles of bisphenols used.

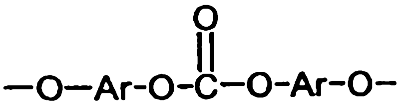
During the production of polycarbonates by the phase boundary method, for example, the branching agents can be introduced into the aqueous alkaline phase with the bisphenols and the chain terminators, or can be added dissolved in an organic solvent together with the acid derivatives. In the transesterification method, the branching agents are preferably added together with the dihydroxy aromatic compounds or bisphenols.

The catalysts which are preferably used for the production of polycarbonates by the melt transesterification method are the ammonium salts and phosphonium salts which are known from the literature (see, for example, US-A 3 442 864, JP-A-14742/72, US-A 5 399 659 and DE-A 19 539 290).

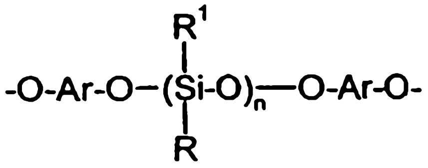
Copolycarbonates can also be used. In particular, copolycarbonates in the sense of the invention include polydiorganosiloxane-polycarbonate block copolymers, the weight average molecular weight (M_w) of which preferably ranges from 10,000 to 200,000 g/mol, most preferably from 20,000 to 80,000 g/mol (as determined by gel chromatography after prior calibration by the measurement of light scattering or by ultracentrifugation). The content of aromatic carbonate structural units in these polydiorganosiloxane-polycarbonate block copolymers is preferably 75 to 97.5 % by weight, most preferably 85 to 97 % by weight. The content of polydiorganosiloxane structural units in the polydiorganosiloxane-polycarbonate block copolymers is preferably 25 to 2.5 % by weight, most preferably 15 to 3 % by weight. These polydiorganosiloxane–polycarbonate block copolymers can be produced, for example, from polydiorganosiloxanes which contain α,ω-bis-hydroxyaryloxy terminal groups and which preferably have an average degree of polymerisation corresponding to P_n = 5 to 100, most preferably P_n = 20 to 80.

These polydiorganosiloxane-polycarbonate block polymers can also be a mixture of polydiorganosiloxane-polycarbonate block copolymers with customary polysiloxane-free, thermoplastic polycarbonates, wherein the total content of polydiorganosiloxane structural units in said mixture is preferably 2.5 to 25 % by weight.

Polydiorganosiloxane-polycarbonate block copolymers such as these are characterised in that they firstly contain aromatic carbonate structural units (1) in their polymer chain and secondly contain polydiorganosiloxanes (2) which comprise aryloxy terminal groups:



$$-O-Ar-O-\overset{\overset{\displaystyle O}{\|}}{C}-O-Ar-O- \qquad (1),$$



$$-O-Ar-O-(\underset{\underset{\displaystyle R}{|}}{\overset{\overset{\displaystyle R^1}{|}}{Si}}-O)_n-O-Ar-O- \qquad (2)$$

wherein

Ar	represents identical or different difunctional aromatic radicals, and
R and R^1	are identical or different and denote a linear alkyl, a branched alkyl, an alkenyl, a halogenated linear alkyl, a halogenated branched alkyl or aryl or a halogenated aryl group, preferably methyl, and
n	denotes the average degree of polymerisation, which is preferably 5 to 100, most preferably 20 to 80.

The alkyl in formula (2) above is preferably a C_1-C_{20}-alkyl, the alkenyl in formula (2) above is preferably a C_2-C_6-alkenyl; the aryl in formula (2) above is preferably a C_6-C_{14}-aryl. In the above formula, the term "halogenated" means partially or completely chlorinated, brominated or fluorinated.

Examples of alkyl, alkenyl, aryl, halogenated alkyl and halogenated aryl groups include methyl, ethyl, propyl, n-butyl, tert.-butyl, vinyl, phenyl, naphthyl, chloromethyl, perfluorobutyl, perfluorooctyl and chlorophenyl.

Polydiorganosiloxane-polycarbonate block copolymers of this type and the production thereof are described, for example, in US-A 3 189 662, US-A 3 821 325 and US-A 3 832 419.

The preferred polydiorganosiloxane-polycarbonate block copolymers can be produced, for example, by the reaction of polydiorganosiloxanes, which contain α,ω-bishydroxyaryloxy terminal groups, with other bisphenols, optionally with the use in conjunction of branching agents in the usual amounts, e.g. by the two-phase boundary method (as described, for example, by H. Schnell in "Chemistry and Physics of Polycarbonates", Polymer Reviews, Volume 9, pages 31-76, Interscience Publishers, New York, London, Sydney, 1964). The polydiorganosiloxanes which contain α,ω-bishydroxyaryloxy terminal groups and which are used as starting materials for this synthesis and the production thereof are described in US-A 3 419 634, for example.

Customary additives such as demoulding agents can be added to the polycarbonates or can be provided on the surface thereof. The polycarbonates which are used preferably already contain demoulding agents before they are compounded with the other components of the moulding compositions according to the invention.

According to the invention , the compositions contain, as **component C)**, one or a mixture of two or more graft polymers comprising a graft base based on acrylates with a glass transition temperature of less than -5°C (graft polymers such as these are generally termed acrylate rubbers and are known to one skilled in the art) or onc or a mixture of two or more different elastic block polymers, particularly two- or three-block copolymers based on aromatic vinyl compounds and dienes, or mixtures of graft polymers and elastic block polymers.

Component C) according to the invention comprises graft copolymers with rubber-like elastic properties, which can essentially be obtained from at least 2 of the following monomers: esters of (meth)acrylic acid comprising 1 to 18 C atoms in their alcohol component, chloroprene, 1,3-butadiene, isopropene, styrene, acrylonitrile, ethylene, propylene and vinyl acetate, wherein the graft base contains at least one ester of (meth)acrylic acid, namely polymers such as those described, for example, in "Methoden der Organischen Chemie" (Houben-Weyl), Volume 1411, Georg Thieme-Verlag, Stuttgart 1961, pages 393-406, and by C.B. Bucknall in "Toughened Plastics", Appl. Science Publishers, London 1977.

The preferred polymers C) are partially crosslinked and have gel contents of more than 5 % by weight, preferably more than 20 % by weight, most preferably more than 40 % by weight, particularly more than 60 % by weight.

The preferred polymers C) are graft polymers which contain

C.1) 95 to 5, preferably 10 to 80 % by weight with respect to component C, of a graft layer based on at least one polymerisable, ethylenically unsaturated monomer as graft monomers, and

C.2) 5 to 95, preferably 20 to 90 % by weight with respect to component C, of an acrylate rubber with a glass transition temperature <-10°C, preferably <-20°C as a graft base.

The acrylate rubbers specified in C.2 are preferably polymers of acrylic acid alkyl esters or methacrylic acid alkyl esters, optionally comprising up to 40 % by weight with respect to C.2 of other polymerisable, ethylenically unsaturated monomers. The preferred acrylic acid esters or methacrylic acid esters comprise C_1-C_8-alkyl esters, particularly methyl, ethyl, butyl, n-octyl and 2-ethylhexyl esters; as well as halogenoalkyl esters, preferably halogeno-C_1-C_8-alkyl esters such as chloroethyl acrylates, as well as mixtures of these monomers.

The acrylic acid alkyl esters and methacrylic acid esters are preferably esters of acrylic acid or methacrylic acid with monohydric alcohols comprising 1 to 18 C atoms. Methyl, ethyl and propyl methacrylates, n-butyl acrylate, t-butyl acrylate and t-butyl methacrylate are particularly preferred.

The graft monomers of graft layer C.l. are preferably selected from at least one monomer, preferably 2 or 3 monomers, from the group consisting of styrene, α-methylstyrene, styrenes comprising halogeno- or methyl-substituted nuclei, (meth)acrylic acid C_1-C_8-alkyl esters, acrylonitrile, methacrylonitrile, maleic anhydride, C_1-C_4-alkyl- or phenyl-N-substituted maleinimides or mixtures thereof.

Graft polymers C) which are particularly preferred comprise graft polymers of:

C.1) 5 to 95, preferably 10 to 80, particularly 30 to 80 parts by weight of a mixture of

C.1.1 50 to 99, preferably 65 to 90 % by weight of methyl methacrylate, styrene, α-methylstyrene, styrenes comprising halogeno- or methyl-substituted nuclei, or mixtures of these compounds, and

C 1.2 1 to 50, preferably 35 to 10 % by weight of methyl methacrylate, acrylonitrile, methacrylonitrile, maleic anhydride, C_1-C_4-alkyl- or -phenyl-N-substituted maleinimides or mixtures of these compounds, on

C.2) 5 to 95, preferably 20 to 90, particularly 20 to 70 parts by weight of polymers which are based on alkyl acrylates and which have a glass transition temperature below -10°C, preferably below -20°C,

wherein the sum of the parts by weight of C.1) and C.2) is 100.

Graft polymers C) which are particularly preferred are those which are obtainable by the graft reaction of

α 10 to 70, preferably 15 to 50, particularly 20 to 40 % by weight, with respect to graft polymer C, of at least one ester of (meth)acrylic acid, or 10 to 70, preferably 15 to 50, particularly 20 to 40 % by weight of a mixture comprising 10 to 50, preferably 20 to 35 % by weight with respect to the mixture, of acrylonitrile or esters of (meth)acrylic acid and 50 to 90, preferably 65 to 80 % by weight with respect to the mixture, of styrene, as graft layer C. 1, on

β 30 to 90, preferably 50 to 85, particularly 60 to 80 % by weight with respect to graft polymer C), of a graft base C.2) which contains 70 to 100 % by weight of at least one alkyl acrylate comprising 1 to 8 C atoms in its alkyl radical, preferably n-butyl acrylate and/or methyl-n-butyl acrylate and/or 2-ethylhexylacrylate, particularly n-butyl acrylate as the sole alkyl acrylate, 0 to 30, preferably 0 to 15 % by weight of a further copolymerisable monoethylenically unsaturated monomer, such as butadiene, isoprene, styrene, acrylonitrile, methyl methacrylate or vinyl ethyl ether or mixtures thereof, 0 to 5 % by weight of a copolymerisable, polyfunctional, preferably bi- or trifunctional monomer which gives rise to crosslinking wherein the percentages by weight are given with respect to the total weight of the graft base.

Examples of preferred graft polymers C) include graft bases C.2) which are grafted with (meth)acrylic acid alkyl esters and/or styrene and/or acrylonitrile. Acrylate rubbers based on n-butyl acrylate are most preferably used as graft base C.2).

Graft polymers C) which are particularly preferred are those which contain less than 5 % by weight of polystyrene units, preferably less than 1 % by weight of polystyrene units with respect to the total weight of the graft, and which most preferably contain no polystyrene units.

Component C) can also be a mixture of different graft copolymers.

The gel content of graft base β is generally at least 20 % by weight, preferably 40 % by weight (as measured in toluene) and the degree of grafting G is generally 0.15 to 0.55.

The average particle diameter of graft polymer C) is preferably 0.01 to 2 μm, most preferably 0.1 to 0.6, particularly 0.2 to 0.4 μm.

The average particle diameter is determined, for example, from electron microscope photographs (TEM) of ultra-thin sections of the moulding compositions according to the invention which have been treated with OsO_4 and RuO_4, by measuring a representative quantity of particles (about 50).

The average particle size d_{50} as determined by ultracentrifugation (W. Scholtan, H. Lange, Kolloid, Z. und Z. Polymere 250 (1972), 782-796) is the diameter above which and below which there is 50 % by weight of the particles. The average particle size d_{50} of graft polymer C) is preferably 0.1 to 0.6 μm.

The gel content of graft base C.2 is determined at 25°C in dimethylformamide (M. Hoffmann, H. Krömer, R. Kuhn, Polymeranalytik I and II, Georg Thieme-Verlag, Stuttgart 1977).

The degree of grafting G denotes the ratio by weight of grafted-on graft monomers to the graft base, and is dimensionless.

Monomers containing more than one polymerisable double bond can be copolymerised to effect crosslinking. Preferred examples of crosslinking monomers include esters of unsaturated monocarboxylic acids comprising 3 to 8 C atoms and unsaturated monohydric alcohols comprising 3 to 12 C atoms, or saturated polyols comprising 2 to 4 OH groups and 2 to 20 C atoms, such as ethylene glycol dimethacrylate, allyl methacrylate; multiply-unsaturated heterocyclic compounds such as e.g. trivinyl and triallyl cyanurates; polyfunctional vinyl compounds such as di- and trivinylbenzenes; and also triallyl phosphate and diallyl phthalate. The preferred crosslinking monomers are allyl methacrylate, ethylene glycol dimethylacrylate, diallyl phthalate, and heterocyclic compounds which comprise at least 3 ethylenically unsaturated groups. Crosslinking monomers which are particularly preferred are the cyclic monomers triallyl cyanurate, triallyl isocyanurate, trivinyl cyanurate, triacryloyl hexahydro-s-triazine, triallylbenzenes, and acrylic acid esters of tricyclodecenyl alcohol.

The amount of crosslinking monomers is preferably 0.02 to 5, particularly 0.05 to 2 % by weight with respect to graft base C.2.

When using cyclic crosslinking monomers comprising at least 3 ethylenically unsaturated groups, it is advantageous to limit the amount thereof to less than 1 % by weight of graft base C.2.

Graft polymers C) can be produced by known methods, such as bulk, suspension, emulsion or bulk suspension methods.

Since, as is known, the graft monomers are not absolutely completely grafted on to the graft base during the graft reaction, graft polymers C) are also to be understood according to the invention as those products which are obtained by the polymerisation the graft monomers in the presence of the graft base.

Graft polymers C) are preferably used in compacted form.

Component CV) according to the invention further comprises block polymers with rubber-like elastic properties, particularly for example two- (A-B) and three- (A-B-A) block copolymers. Block copolymers of the A-B and A-B-A type may exhibit the typical behaviour of thermoplastic elastomers. The preferred block copolymers of the A-B and A-B-A type contain one or two aromatic vinyl blocks (preferably based on styrene) and a rubber block (preferably a diene rubber block, particularly a polybutadiene block or an isoprene block) which may optionally be partially or completely hydrogenated.

Suitable block copolymers of the A-B and A-B-A type are described, for example, in US-A 3 078 254, 3 402 159, 3 297 793, 3 265 765 and 3 594 452 and in GB-A 1 264 741. Examples of typical block copolymers of the A-B and A-B-A type include: polystyrene-polybutadiene (SBR), polystyrene-poly(ethylene-propylene), polystyrene-polyisoprene, poly(α-methylstyrene)-polybutadiene, polystyrene-polybutadiene-polystyrene (SBR), polystyrene-poly(ethylene-propylene)-polystyrene, polystyrene-polyisoprene-polystyrene and poly(α-methylstyrene)-polybutadiene-poly(α-methyl-styrene), as well as hydrogenated versions thereof, such as, for example and preferably, hydrogenated polystyrene-polybutadiene-polystyrene (SEBS) and hydrogenated polystyrene-polyisoprene (SEP). The use of corresponding hydrogenated block copolymers, optionally in admixture with the non-hydrogenated precursor thereof as an impact toughness modifier, is described, for example, in DE-A 2 750 515, DE-A 2 434 848, DE-A 038 551, EP-A 0 080 666 and WO 83/01254. Reference is hereby expressly made to the disclosures of the documents cited above.

Mixtures of said block polymers can also be used.

Partially or completely hydrogenated block copolymers are particularly preferred; hydrogenated polystyrene-polybutadiene-polystyrene (SEBS) and hydrogenated polystyrene-polyisoprene (SEP) are most particularly preferred.

Block polymers such as these, of the A-B and A-B-A type, are commercially available from a variety of sources, e.g. from Phillips Petroleum under the trade name

SOLPRENE, from Shell Chemical Co. under the trade name KRATON, from Dexco under the trade name VECTOR and from Kuraray under the trade name SEPTON.

According to the invention, the thermoplastic moulding compositions contain, as **component D**, one or a mixture of two or more different mineral fillers based on talc. If component C is a block copolymer, the blends contain the mineral filler in particular in an amount ranging from 1.5 to 34, most preferably in an amount ranging from 1.5 to 25 parts by weight.

All particulate fillers which one skilled in the art associates with talc or french chalk, which are commercially available, and the product descriptions of which contain the terms talc or french chalk as characterising features, are suitable as mineral fillers based on talc in the sense of the invention.

The preferred mineral fillers have a content of talc according to DIN 55920 of more than 50 % by weight, preferably more than 80 % by weight, more preferably more than 95 % by weight and most preferably more than 98 % by weight with respect to the total weight of filler.

The mineral fillers can also be surface-treated. They can be coated with a suitable size system, for example one which contains a bonding agent or bonding agent system, e.g. a silane-based material.

The particulate fillers preferably have an upper particle size or grain size d_{97} of less than 50 μm, preferably less than 25, more preferably less than 10 and most preferably less than 6 μm. A value less than 10, preferably less than 6, more preferably less than 2 and most preferably less than 1 μm is selected as the average particle size d_{50}. The (d_{97}- and d_{50} values of the fillers D are determined by sedimentation analysis using a SEDIGRAPH D 5000 or by sieve analysis according to DIN 66 165.

The average aspect ratio (diameter to thickness) of the particulate fillers preferably falls within the range from 1 to 100, more preferably from 2 to 25 and most preferably from 5 to 25, as determined on electron microscope photographs of ultra-thin sections of the finished products by measuring a representative quantity (about 50) of filler particles.

Due to the processing conditions employed to form the moulding composition or moulding, the particles in the moulding composition or moulding may have a d_{97} or d_{50} value which is less than that of the fillers originally used.

The particle diameter in the finished product can be determined, for example, by taking electron microscope photographs of thin sections of the polymer mixture and employing at least 25, preferably at least 50 filler particles for the evaluation.

In addition, the compositions according to the invention can contain customary additives. These can generally be added in an amount of up to 15 % by weight, preferably in an amount from 0.01 to 10, more preferably from 0.05 to 5, most preferably from 0.1 to 3 % by weight, with respect to the total weight of the moulding compositions.

All customary additives are suitable, such as stabilisers (for example UV stabilisers, thermal stabilisers), antistatic agents, flow enhancers, demoulding agents, flame retardant additives, emulsifiers, nucleating agents, plasticisers, internal lubricants, additives which lower the pH (e.g. compounds which contain carboxyl groups), additives for increasing the conductivity, colorants and pigments. These and other suitable additives are described, for example, by Gächter and Müller in Kunststoff-Additive, 3rd Edition, Hanser-Verlag, Munich, Vienna, 1989. The additives can be used on their own or in the form of master batches. The additives can be admixed and/or can be applied to the surface.

Examples of substances which can be used as stabilisers include sterically hindered phenols and/or phosphites, hydroquinones, aromatic secondary amines such as diphenylamines, substituted resorcinols, salicylates, benzotriazoles and benzophenones, as well as various substituted representatives of these groups and mixtures thereof.

Examples of pigments which can be used include titanium dioxide, ultramarine blue, iron oxide, carbon black, phthalocyanins, quinacridones, perylenes, nigrosin and anthraquinones.

Examples of nucleating agents which can be used include sodium phenylphosphinate, alumina, silica, as well as french chalk, which is preferred, and the other nucleating agents described above.

Examples of internal lubricants and demoulding agents which can be used include ester waxes, pentaerythritol stearate (PETS), long chain fatty acids (e.g. stearic acid or behenic acid) and salts thereof (e.g. Ca or Zn stearates), as well as amide derivatives (e.g. ethylene-bis-stearylamide) or montan waxes (mixtures of straight chain, saturated carboxylic acids with chain lengths ranging from 28 to 32 C atoms), and also low molecular weight polyethylene or polypropylene waxes.

Examples of plasticisers which can be used include dioctyl phthalate, dibenzyl phthalate, butyl benzyl phthalate, hydrocarbon oils, and N-(n-butyl)benzenesulphonamide.

In order to obtain conductive moulding compositions, carbon blacks, conductivity blacks, carbon fibres, nano-scale graphite fibres (nanotubes), graphite, conductive polymers, metal fibres or other customary additives can be used in order to increase the conductivity.

Examples of flame retardants which can be used include commercially available organic halogen compounds comprising synergists or commercially available organic nitrogen compounds or organic/inorganic phosphorus compounds; these can be used individual or in admixture. Mineral flame retardant additives can also be used, such as magnesium hydroxide or hydrated Ca-Mg carbonates (e.g. as described in DE-OS 4

236 122). Example of compounds which contain halogens, particularly brominated and chlorinated compounds, include: ethylene-1,2-bistetrabromophthalimide, epoxidised tetrabromobisphenol A resin, tetrabromobisphenol A oligocarbonate, tetrachloro-bisphenol A oligocarbonate, pentabromopolyacrylate, and brominated polystyrene. Suitable organic phosphorus compounds include the phosphorus compounds according to W098/17720 (PCTIEP/05705), e.g. triphenyl phosphate (TPP), resorcinol-bis-(diphenylphosphate) including oligomers thereof, and bisphenol A-bis-diphenylphosphate including oligomers thereof (see EP-A 363 608 and EP-A 640 655, for example), melamine phosphate, melamine pyrophosphate, melamine polyphosphate and mixtures thereof. Nitrogen compounds which are particularly suitable include melamine and melamine cyanurate. Examples of synergists include antimony compounds, particularly antimony trioxide and antimony pentoxide, zinc compounds, and tin compounds such as tin stannate and borate. Carbon-forming substances and tetrafluorethylene polymers can also be added. Flame retardants, optionally with a synergist such as antimony compounds, and with antidripping agents, are generally used in amounts up to 30 % by weight, preferably up to 20 % by weight, with respect to the overall composition.

Reinforcing materials, e.g. in the form of glass fibres, can also be used as additives.

The present invention further relates to a process for producing said compositions, to the use of the compositions according to the invention for the production of semi-finished products and mouldings, and to semi-finished products and mouldings produced therefrom.

The compositions according to the invention are produced by mixing the components by processes known in the art. It may be advantageous to premix individual components. Mixing of components A to D, as well as other constituents, is preferably effected at temperatures from 220 to 330°C by jointly kneading, extruding or rolling the components.

The compositions according to the invention can be processed by customary methods to form semi-finished products or mouldings of all types. Examples of processing methods include extrusion methods and injection moulding methods. Examples of semi-finished products include sheeting and panels.

The mouldings can be of small or large dimensions and can be used for exterior. Bulky mouldings for vehicle construction, particularly for the automobile sector, are preferred. In particular, exterior bodywork parts can be produced from the moulding compositions according to the invention, such as wings, tailgates, bonnets, bumpers, loading areas, covers for loading areas, automobile roofs, or other bodywork components.

Mouldings or semi-finished products formed from the moulding materials/compositions according to the invention can also be used in combination with other materials, such as metals or plastics for example. After the lacquer-coating of exterior bodywork parts, for example, lacquer coats can be applied directly to the moulding compositions according to the invention and/or to the materials which are used in combination therewith. The moulding compositions according to the invention or the mouldings/semi-finished products which are produced from the moulding compositions according to the invention can be used themselves for the production of finished parts such as exterior bodywork parts, or can be used therefor in combination with other materials by means of customary joining and assembly techniques such as coextrusion, injection moulding behind sheeting, injection moulding around inserted parts, adhesive bonding, welding, screwed connections or flanged connections.

The moulding compositions according to the invention can also be used for numerous other applications. Examples thereof include their use in electrical engineering, in the building sector or in data storage. Examples of the use of moulding compositions according to the invention in the aforementioned areas of use include lamp covers, coil bodies, safety glass panes, housing materials for electronic devices, housing materials for domestic appliances, and as panels for the production of covers.

The compositions according to the invention are distinguished by their excellent resistance to thermal deformation and dimensional stability when hot. In addition, the compositions according to the invention exhibit an unexpectedly slight extent of decrease in their impact toughness at low temperatures. Furthermore, they comply with stringent requirements as regards stability during processing, flowability in the melt, toughness, drawing capacity at low temperatures, stiffness, thermal expansion, surface quality, capacity for being lacquer-coated, and resistance to chemicals and fuels.

Examples

Component A

Polyethylene terephthalate Type 1: this was a polyethylene terephthalate with an intrinsic viscosity IV of about 0.69 cm^3/g and an isothermal time of crystallisation at 215°C of about 7 minutes.

Polyethylene terephthalate Type 2: this was a polyethylene terephthalate with an intrinsic viscosity IV of about 0.73 cm^3/g and an isothermal time of crystallisation at 215°C of about 3 minutes.

The intrinsic viscosity was measured in phenol/o-dichlorobenzene (1:1 parts by weight) at 25°C.

The isothermal time of crystallisation of PET was determined by the DSC method (differential scanning calorimetry) using a PERKIN ELMER DSC 7 differential scanning calorimeter (sample weight about 10 mg, perforated Al capsule) with the following temperature programme:

1. Heating from 30°C to 290°C at 40°C/min,
2. 5 minute isotherm at 290°C,
3. Cooling from 290°C to 215°C at 160°C/min,
4. 30 min isotherm at 215°C (crystallisation temperature).

The evaluation software used was PE Thermal Analysis 4.00.

Component B

A linear polycarbonate (Makrolon 2805 from Bayer AG, Leverkusen, Germany) based on bisphenol A with a viscosity $\eta_{rel.}$ of about 1.29 (measurement conditions: 5 g polycarbonate per litre of methylene chloride, 2°C) and a molecular weight Mw of about 29,000 g/mol as determined by GPC methods against a polycarbonate standard).

Component C

The acrylate graft polymer which was used was Paraloid EXL 2300 from Rohm und Haas Deutschland GmbH, Frankfurt.

The block copolymers used were Kraton G 1651 (SEBS, Type 1) and Kraton G 1702 (SEP, Type 2) from Shell Chemical.

Component D

Talc Type 1: This was Tital 5 from Incemin AG (Switzerland) with a d_{50} value of about 0.9 μm and a d_{97} value of less than 5 μm.

Talc Type 2: This was Naintsch A 10 from Naintsch Mineralwerke (Austria) with a d_{50} value of about 2.2 μm and a d_{97} value of less than 10 μm.

Talc Type 3: This was Naintsch A 60 from Naintsch Mineralwerke (Austria) with a d_{50} value of about 12 μm and a d_{97} value of less than 80 μm.

The d_{50} and d_{97} values were determined particle size distribution measurements using a Sedigraph 5000 D or by sieve analysis according to DIN 66 165.

Customary stabilisers, nucleating agents and demoulding agents were used as additives.

Compounding was effected in a Type ZSK32 twin-shaft extruder (Werner und Pfleiderer) at material temperatures from 250 to 290°C.

The test specimens were injection moulded in an Arburg 320-210-500 injection moulding machine at material temperatures from 260 to 280°C and tooling temperatures from 70 to 90°C.

The moulding compositions according to the invention were tested by the following methods:

Vicat B: dimensional stability when hot or resistance to thermal deformation according to DIN ISO 306/B 120 in silicone oil.

HDT A: dimensional stability when hot or resistance to thermal deformation according to DIN ISO 75-2, method Af.

Izod impact toughness: toughness according to ISO 180, method 1 U.

Tensile modulus: stiffness according to DIN / EN / ISO 527-2/1A.

Elongation at break: extensibility determined according to DIN/EN/ISO 527-2/1A.

Coefficient of linear thermal expansion: determined according to DIN 53 752IB over the temperature range from 0 to 55°C.

MVR: Flowability according to DIN/ISO 1133 at 280°C and 2.16 kg.

Surface quality: determination of surface quality by expert visual observation. ++ denotes a responsive, smooth surface with a very good capacity for being lacquer-coated.

The compositions and properties of the thermoplastic moulding compositions according to the invention are given in Tables 1 to 5.

It follows from Tables 1 to 5 that the moulding compositions according to the invention exhibit excellent resistance to thermal deformation/dimensional stability when hot (Vicat B, HDT A), and exhibit an unexpectedly slight decrease in impact toughness at low temperatures (Izod impact toughness).

Moreover, they comply with the requirements imposed on thermoplastic moulding compositions for exterior bodywork parts of large surface area with regard to stiffness (tensile modulus), extensibility (elongation at break), thermal expansion (coefficient of linear thermal expansion), flowability in the melt (MVR) and capacity for being lacquer-coated (surface quality).

Table 1

Example		1	2	3
Polycarbonate	[%]	40	40	40
Polyethylene terephthalate, Type 2	[%]	29.2	24.2	19.2
Graft rubber	[%]	10	15	20
Talc, Type 1	[%]	20	20	20
Additive	[%]	0.8	0.8	0.8
Vicat B	[°C]	143	140	134
HDTA	[°C]	111	111	109
Izod impact toughness 23°C	[kJ/m^2]	77	116	119
Izod impact toughness -10°C	[kJ/m^2]	80	67	123
Izod impact toughness -20°C	[kJ/m^2]	70	69	92
Tensile modulus	[Mpa]	4500	3940	3560
Elongation at break	[%]		14	13
Coefficient of linear thermal expansion (l/tr)	[10^{-6} / K]	54/84	44/76	41/66
MVR (280°C/2.16 k	[cm^3/10min]	9	4	2
Surface quality		++	++	++

l/tr = longitudinal/transverse n.b. = not broken

Table 2

Example		4	5	6	7
Polycarbonate	[%]	50	50	50	50
Polyethylene terephthalate, Type 1	[%]	27.1	22.1	17.1	12.1
Graft rubber	[%]	12	12	12	12
Talc, Type 1	[%]	10	15	20	25
Additive	[%]	0.9	0.9	0.9	0.9
Vicat B	[°C]	140	142	140	138
HDTA	[°C]	101	106	118	120
Izod impact toughness 23°C	[kJ/m^2]	n.b.	49	62	81
Izod impact toughness -10°C	[kJ/m^2]		59	59	87
Izod impact toughness -20°C	[kJ/m^2]	n.b.	53	62	55
Tensile modulus	[Mpa]	3000	3630	4040	4190
Elongation at break	[%]	47	6	8	7
Coefficient of linear thermal expansion (l/tr)	[10^{-6} / K]	59/85	54/76	48/81	47/72
MVR (280°C/2.16 k	[cm^3/10min]	12	IS	5	3
Surface quality		++	++	++	++

l/tr = longitudinal/transverse n.b. = not broken

Table 3

Example		8	9	10
Polycarbonate	[%]	40	40	40
Polyethylene terephthalate, Type 1	[%]	37.1	32.1	27.1
Graft rubber	[%]	12	12	12
Talc, Type 1	[%]	10	15	20
Additive	[%]	0.9	0.9	0.9
Vicat B	[°C]	140	141	140
HDTA	[°C]	98	103	111
Izod impact toughness 23°C	[kJ/m^2]	n.b.	174	58
Izod impact toughness -10°C	[kJ/m^2]	167	87	56
Izod impact toughness -20°C	[kJ/m^2]	156	93	62
Tensile modulus	[Mpa]	3090	3610	4140
Elongation at break	[%]	22	20	8
Coefficient of linear thermal expansion (l/tr)	[10^{-6} / K]	73/81	69/78	50/71
MVR (280°C/2.16 k	[cm^3/10min]	16	12	9
Surface quality		++	++	++

l/tr = longitudinal/transverse n.b. = not broken

Table 4

Example		11	12	13
Polycarbonate	[%]	30	30	30
Polyethylene terephthalate, Type 1	[%]	47.1	42.1	37.1
Graft rubber	[%]	12	12	12
Talc, Type 1	[%]	10	15	20
Additive	[%]	0.9	0.9	0.9
Vicat B	[°C]	142	142	141
HDTA	[°C]	90	93	106
Izod impact toughness 23°C	[kJ/m^2]	73	67	24
Izod impact toughness -10°C	[kJ/m^2]	66	63	32
Izod impact toughness -20°C	[kJ/m^2]	63	57	33
Tensile modulus	[Mpa]	3140	3680	4300
Elongation at break	[%]	8	5	3
Coefficient of linear thermal expansion (l/tr)	[10^{-6} / K]	66/90	62/60	48/72
MVR (280°C/2.16 k	[cm^3/10min]	22	17	12
Surface quality		++	++	++

l/tr = longitudinal/transverse n.b. = not broken

Table 5

Example		14	15	16	17	18	19
Polycarbonate	[%]	50	50	50	50	50	50
Polyethylene terephthalate, Type 1	[%]	27.2	27.2	27.2	--	--	--
Polyethylene terephthalate, Type 2	[%]	--	--	--	27.2	27.2	27.2
Graft rubber	[%]	12	12	12	12	12	12
Talc, Type 1	[%]	10	--	--	10	--	--
Talc, Type 2	[%]	--	10	--	--	10	--
Talc, Type 3	[%]	--	--	10	--	--	10
Additive	[%]	0.8	0.8	0.8	0.8	0.8	0.8
Vicat B	[°C]	140	140	138	139	139	138
HDTA	[°C]	107			105		
Izod impact toughness 23°C	[kJ/m^2]	n.b.	n.b.	166	n.b.	n.b.	n.b.
Izod impact toughness -10°C	[kJ/m^2]	--	286	--	--	--	--
Izod impact toughness -20°C	[kJ/m^2]	n.b.	198	122	n.b.	n.b.	127
Tensile modulus	[Mpa]	3020	3040	3040	3020	2990	2990
Elongation at break	[%]	43	33	15	38	44	19
MVR (280°C/2.16 kg)	[cm^3/10min]	11	10	11	7	8	8
Surface quality		++	++	++	++	++	++

n.b. = not broken

Table 6

Example		20	21	22	23	24
Polycarbonate	[%]	47	47	50	50	43
Polyethylene-	[%]	34.2	34.2	27.2	26.9	21.3
block polymer,Type 1	[%]	12	0	12	0	15
block polymer,Type 2	[%]	0	12	0	12	0
Talc,Type 1	[%]	3	3	10	10	20
Additive	[%]	0.8	0.8	0.8	1.1	0.7
Vicat B	[°C]	136	133	140	138	140
HDTA	[°C]	90	95	107	108	112
Izod impact	[kJ/m^2]	n.b.	n.b.	n.b.	n.b.	92
Izod impact	[kJ/m^2]	n.b.	n.b.	n.b.	n.b.	79
Izod impact	[kJ/m^2]	n.b.	n.b.	n.d.	n.b.	76
Tensile modulus	[Mpa]	2200	2190	2900	2900	3940
Elongation at break	[%]	110	120	50	54	9
Coefft. of linear ...	10^{-6} / K	-	-	71/82	-	53/65
MVR (280°C/2.16 kg)	[cm^3/10min]	24	27	21	24	11
Surface quality		++	++	++	++	++

l/tr = longitudinal/transverse n.b. = not broken

Throughout this specification and the claims which follow, unless the context requires otherwise, the word "comprise", or variations such as "comprises" or "comprising", will be understood to imply the inclusion of a stated integer or step or group of integers or steps but not the exclusion of any other integer or step or group of integers or steps.

The reference to any prior art in this specification is not, and should not be taken as, an acknowledgment or any form of suggestion that the prior art forms part of the common general knowledge in Australia.

THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS:

1. A composition comprising
   A) 4 to 80 parts by weight of at least one polyethylene terephthalate,
   B) 10 to 90 parts by weight of at least one aromatic polycarbonate,
   C) 1.5 to 30 parts by weight of a mixture of at least one graft polymer comprising a graft base based on acrylates with a glass transition temperature of less than –5°C and or at least one elastomeric block copolymer or mixtures of graft copolymers and block copolymers, and
   D) 1.5 to 54 parts by weight at least one mineral filler based on talc,

wherein the sum of the parts by weight of all the components is 100 and wherein component D) has an upper particle size d_{97} less than 50μm.

2. A composition according to Claim 1, wherein the mineral filler D) has a content of talc greater than 50 % by weight with respect to the total amount of filler D.

3. A composition according to Claim 1, wherein component D) has an average particle size d_{50} less than 10 μm.

4. A composition according to Claim 1, comprising from 10 to 60 parts by weight of at least one polyethylene terephthalate.

5. A composition according to Claim 1, comprising 12 to 40 parts by weight of at least one polyethylene terephthalate.

6. A composition according to Claim 5, comprising 19 to 29 parts by weight of at least one polyethylene terephthalate

7. A composition according to Claim 1, comprising 20 to 80 parts by weight of at least one aromatic polycarbonate.

8. A composition according to Claim 1, comprising 3 to 25 parts by weight of the mixture of at least one graft polymer comprising a graft base based on acrylates with a glass transition temperature of less than –5°C and at least one elastomeric block copolymergraft copolymer.

9. A composition according to Claim 1, comprising 3 to 34 parts by weight of at least one filler corresponding to component D).

10. A composition according to Claim 1, comprising as component C), graft polymers comprising

- A) 4 to 80 parts by weight of at least one polyethylene terephthalate,
- B) 10 to 90 parts by weight of at least one aromatic polycarbonate,
- C) 1.5 to 30 parts by weight of
  - C.1) 95 to 5 % by weight, with respect to component C), of a graft layer based on at least one polymerizable, ethylenically unsaturated monomer as graft monomers, and
  - C.2) 5 to 95 % by weight, with respect to component C), of an acrylate rubber with a glass transition temperature <–10°C as a graft base, and
- D) 1.5 to 54 parts by weight at least one mineral filler based on talc,

wherein the sum of the parts by weight of all the components is 100.

11. A composition according to Claim 10, wherein C.2) comprises polymers of acrylic acid esters or methacrylic acid esters, which comprise up to 40 % by weight, with respect to C.2), of other ethylenically unsaturated monomers.

12. A composition according to Claim 1, comprising, as a component,

- A) 4 to 80 parts by weight of at least one polyethylene terephthalate,
- B) 10 to 90 parts by weight of at least one aromatic polycarbonate,
- C) 1.5 to 30 parts by weight of one or more block polymers comprising two (A-B) blocks or three (A-B-A) blocks, and
- D) 1.5 to 54 parts by weight at least one mineral filler based on talc,

wherein the sum of the parts by weight of all the components is 100.

13. A composition according to Claim 12, comprising block polymers comprising one or two aromatic vinyl blocks and one diene rubber block.

14. A composition according to Claim 12, wherein one or two blocks are polystyrene and one block is polybutadiene or polyisoprene.

15. A composition according to Claim 12 wherein one or more blocks are partially or completely hydrogenated.

16. A molding or semi-finished product comprising the composition according to Claim 1 comprising

A) 4 to 80 parts by weight of at least one polyethylene terephthalate,

B) 10 to 90 parts by weight of at least one aromatic polycarbonate,

C) 1.5 to 30 parts by weight of at least one graft polymer comprising a graft base based on acrylates with a glass transition temperature of less than –5°C or at least one elastomeric block copolymer or mixtures of graft copolymers and block copolymers, and

D) 1.5 to 54 parts by weight at least one mineral filler based on talc,

wherein the sum of the parts by weight of all the components is 100.

17. A molding or semi-finished product comprising the composition according to Claim 10.

18. A composition substantially as hereinbefore described with reference to the examples.

DATED THIS 26th day of October, 2004.

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