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CA 2494635 A1 2004/02/19

(21) 2 494 635

(12) DEMANDE DE BREVET CANADIEN CANADIAN PATENT APPLICATION

(13) A1

(86) Date de dépôt PCT/PCT Filing Date: 2003/07/23

(87) Date publication PCT/PCT Publication Date: 2004/02/19

(85) Entrée phase nationale/National Entry: 2005/02/02

(86) N° demande PCT/PCT Application No.: EP 2003/008034

(87) N° publication PCT/PCT Publication No.: 2004/015001

(30) Priorité/Priority: 2002/08/05 (102 35 754.4) DE

(51) Cl.Int.⁷/Int.Cl.⁷ C08K 5/521, C08L 69/00

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(54) Titre: MATIERES A MOULER POLYCARBONATE IGNIFUGES ET MODIFIEES AVEC UN POLYMERE GREFFE (54) Title: FLAME-RETARDANT POLYCARBONATE MOULDED MASSES MODIFIED WITH A GRAFT POLYMER

$$R^{1} = (O)_{n} = (O)_{n$$

(57) Abrégé/Abstract:

Polycarbonate moulded masses modified with a graft polymer, comprising phosphorous compounds of formula (1), where R^1 , R^2 , R^3 and R^4 , independently = optionally halo-substituted C_1 - C_8 alkyl, optionally halo- or alkyl-substituted C_5 - C_6 cycloalkyl, C_6 - C_{10} aryl or C_7 - C_{12} aralkyl, n independently = 0 or 1, q independently = 0, 1, 2, 3 or 4, N = 0 to 10, R^5 and R^6 independently = C_1 - C_4 alkyl or halogen, $Y = C_1$ - C_7 alkylidene, C_1 - C_7 alkylene, C_5 - C_{12} cycloalkylene, C_5 - C_{12} cycloalkylidene, -O-, -S-, -SO-, -SO₂- or -CO- and fluorinated polyolefins in the form of a mixture with polyalkyl(meth)arylates.





Flame-resistant polycarbonate moulding compounds modified with a graft polymer

Abstract

Polycarbonate moulding compounds modified with a graft polymer and containing phosphorus compounds of formula (I):

$$R^{1} = (O)_{n} = (O)_{n$$

wherein

- R¹, R², R³ and R⁴ independently of one another are optionally halogen-substituted C₁-C₈-alkyl or optionally halogen-substituted and/or alkyl-substituted C₅-C₆-cycloalkyl, C₆-C₁₀-aryl or C₇-C₁₂-aralkyl,
- n independently of one another are 0 or 1,
- q independently of one another are 0, 1, 2, 3 or 4,
- N is 0 to 10,

R⁵ and R⁶ independently of one another are C₁-C₄-alkyl or halogen, and

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Y is C_1 - C_7 -alkylidene, C_1 - C_7 -alkylene, C_5 - C_{12} -cycloalkylene, C_5 - C_{12} -cycloalkylidene, C_5 - C_7

and fluorinated polyolefins in the form of a mixture with polyalkyl (meth)acrylates.

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_ 1 _

Flame-resistant polycarbonate moulding compounds modified with a graft polymer

The present invention relates to flame-resistant polycarbonate moulding compounds modified with a graft polymer and containing phosphorus compounds and specially formulated fluorinated polyolefins, said moulding compounds having an excellent fire resistance and very good mechanical properties such as notched impact strength, elongation at break, modulus of elasticity and weld strength, together with a very good stress cracking behaviour.

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Diphosphates are known as flameproofing additives. JP 59 202 240 describes the preparation of such a product from phosphorus oxychloride, diphenols like hydroquinone or bisphenol A, and monophenols like phenol or cresol. These diphosphates can be used in polyamide or polycarbonate as flameproofing agents. However, said patent contains no indication of an improved stress cracking resistance due to the addition of the oligomeric phosphate to polycarbonate moulding compounds.

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EP-A 363 608 describes flame-resistant polymer mixtures of aromatic polycarbonate, styrene-containing copolymer or graft copolymer, and oligomeric phosphates as flameproofing agents. It is mentioned in general that tetrafluoroethylene polymers can be added.

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EP-A 0 767 204 describes flame-resistant polyphenylene oxide (PPO) or polycarbonate mixtures that contain a mixture of oligophosphates (bisphenol A (BPA) oligophosphate type) and monophosphates as flameproofing agents. High contents of flameproofing agents lead to disadvantageous mechanical properties (notched impact strength, stress cracking behaviour) and a reduced dimensional stability under heat.

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EP-A 0 611 798 and WO 96/27600 describe moulding compounds containing polycarbonate and also oligomeric, terminally alkylated phosphoric acid esters of the BPA type. Because of the alkylation, the attainment of effective flameproofing requires high proportions, which is very disadvantageous for many application engineering properties (mechanics, dimensional stability under heat).

EP-A 0 754 531 describes reinforced PC/ABS moulding compounds suitable for precision parts. The flameproofing agents used are oligophosphates of the BPA type, inter alia. The high proportions of filler have a very disadvantageous effect on mechanical properties such as elongation at break or notched impact strength.

EP-A 771 851 describes moulding compounds containing aromatic polycarbonate, graft polymer based on diene rubber, SAN copolymer, a phosphate and tetra-fluoroethylene polymers, the polycarbonate having various molecular weights. Stability to loss of impact strength, heat stability and moisture resistance are disclosed as advantages.

To achieve an adequate level of flameproofing, the above-mentioned polymer mixtures usually require the presence of small proportions of PTFE as an antidripping agent. In the event of a fire, this prevents the polymer mixture from tending to produce burning drips and igniting materials situated underneath. In certain fire tests, e.g. the UL 94 V test, the avoidance of burning drips after ignition is assessed as particularly favourable and is stipulated as essential for achieving the best assessment, V-0.

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US-A 5,804,654 describes PTFE powders which are partially encapsulated with a styrene-containing polymer or copolymer and which are very suitable as additives in polyester blends such as PC / ABS. US-A 6, 040, 300 describes processes for the preparation of such special PTFE-containing powders.

The object of the present invention is to provide flameproof PC moulding compounds modified with a graft polymer which are distinguished by very good mechanical properties, a high degree of fire resistance and a very good stress cracking behaviour. In particular, these advantages shall also be observed at elevated processing temperatures. These moulding compounds are thus particularly suitable for applications where contact with special media, e.g. solvent, lubricant, cleaning agent, etc., can occur.

It has now been found that the compositions according to the invention exhibit the above-described advantages in terms of properties when the PTFE additive is used as a mixture with polyalkyl (meth)acrylates.

The invention therefore provides polycarbonate moulding compounds modified with a graft polymer and containing phosphorus compounds of formula (I):

$$R^{1} = (O)_{n} = (P^{5})_{q} = (P^{5})_{q} = (Q)_{n} = (Q)_{n}$$

wherein

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- 20 R¹, R², R³ and R⁴ independently of one another are optionally halogen-substituted C₁-C₈-alkyl or optionally halogen-substituted and/or alkyl-substituted C₅-C₆-cycloalkyl, C₆-C₁₀-aryl or C₇-C₁₂-aralkyl,
 - n independently of one another are 0 or 1,
 - q independently of one another are 0, 1, 2, 3 or 4,

- N is 0 to 10, preferably 0.5 to 5 and particularly 0.9 to 3,
- R⁵ and R⁶ independently of one another are C₁-C₄-alkyl, preferably methyl, or halogen, preferably chlorine and/or bromine, and
 - Y is C_1 - C_7 -alkylidene, C_1 - C_7 -alkylene, C_5 - C_{12} -cycloalkylene, C_5 - C_{12} -cycloalkylidene, -O-, -S-, -SO-, -SO₂- or -CO-,
- and fluorinated polyolefins in the form of a mixture with polyalkyl (meth)acrylates.

The thermoplastic moulding compounds contain preferably 0.5 to 20, particularly preferably 1 to 18 and particularly 2 to 16 parts by weight of phosphorus compound (I) or a mixture of phosphate compounds (I).

The thermoplastic moulding compounds contain preferably 0.01 to 3, particularly 0.05 to 2 and very particularly preferably 0.1 to 0.8 parts by weight of fluorinated polyolefins in the form of special formulations with polyalkyl (meth)acrylates.

- 20 Preferred thermoplastic moulding compounds are those containing
 - A) 40 to 99 and preferably 60 to 98.5 parts by weight of aromatic polycarbonate and/or polyestercarbonate,
- 25 B) 0.5 to 60, preferably 1 to 40 and particularly 2 to 25 parts by weight of a graft polymer consisting of
 - B.1) 5 to 95 and preferably 30 to 80 wt.% of one or more vinyl monomers to

- B.2) 95 to 5 and preferably 20 to 70 wt.% of one or more graft bases with a glass transition temperature of <10°C, preferably of <0°C and particularly preferably of <-20°C,
- C) 0 to 45, preferably 0 to 30 and particularly preferably 0 to 25 parts by weight of at least one thermoplastic polymer selected from the group comprising vinyl (co)polymers and polyalkylene terephthalates,
- D) 0.5 to 20 parts by weight, preferably 1 to 18 parts by weight and particularly preferably 2 to 16 parts by weight of a phosphorus compound of formula (I):

wherein R¹ to R⁶, Y, n, N and q are as defined above, and

E) 0.01 to 3, preferably 0.05 to 2 and particularly preferably 0.1 to 0.8 parts by weight of fluorinated polyolefins in the form of a mixture with polyalkyl (meth)acrylates.

20 <u>Component A</u>

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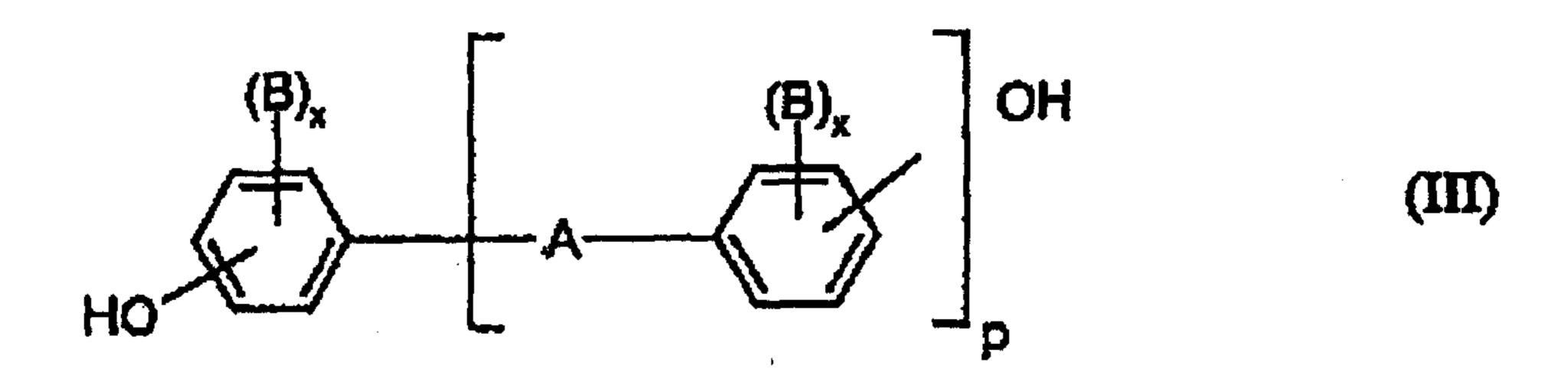
Aromatic polycarbonates and/or aromatic polyestercarbonates that are suitable according to the invention as the component A are known in the literature or can be prepared by processes known in the literature (for the preparation of aromatic polycarbonates see e.g. Schnell, "Chemistry and Physics of Polycarbonates", Interscience Publishers, 1964, and DE-AS 1 495 626, DE-OS 2 232 877, DE-OS

2 703 376, DE-OS 2 714 544, DE-OS 3 000 610 and DE-OS 3 832 396; for the preparation of aromatic polyestercarbonates see e.g. DE-OS 3 077 934).

Aromatic polycarbonates are prepared e.g. by reacting diphenols with carbonic acid halides, preferably phosgene, and/or with aromatic dicarboxylic acid dihalides, preferably benzenedicarboxylic acid dihalides, by the phase interface process, optionally using chain terminators, for example monophenols, and optionally using trifunctional or more than trifunctional branching agents, for example triphenols or tetraphenols.

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Diphenols for the preparation of the aromatic polycarbonates and/or aromatic polyestercarbonates are preferably those of formula (III):



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wherein

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

or a radical of formula (TV) or (V):

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$$\begin{array}{c}
C^{1} \\
(X^{1})_{m} \\
R^{7} \\
R^{8}
\end{array}$$

$$\begin{array}{c}
CH_{3} \\
-C \\
-C \\
-C \\
-C
\end{array}$$

$$\begin{array}{c}
CH_{3} \\
-C \\
-C \\
-C
\end{array}$$

- B are in each case C_1 - C_{12} -alkyl, preferably methyl, or halogen, preferably chlorine and/or bromine,
 - x independently of one another are in each case 0, 1 or 2,
 - p is 1 or 0, and

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- R^7 and R^8 can be individually chosen for each X^1 and independently of one another are hydrogen or C_1 - C_6 -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 R^7 and R^8 are simultaneously alkyl on at least one atom X^1 .

Preferred diphenols are hydroquinone, resorcinol, dihydroxydiphenols, bis(hydroxy-phenyl)-C₁-C₅-alkanes, bis(hydroxyphenyl)-C₅-C₆-cycloalkanes, bis(hydroxyphenyl) ethers, bis(hydroxyphenyl) sulfoxides, bis(hydroxyphenyl) ketones, bis(hydroxy-

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

Particularly preferred diphenols are 4,4'-dihydroxybiphenyl, 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 sulfide and their di- and tetrabrominated or chlorinated derivatives, for example 2,2-bis(3-chloro-4-hydroxyphenyl)propane, 2,2-bis(3,5-dibromo-4-hydroxyphenyl)-propane.

2,2-Bis(4-hydroxyphenyl)propane (bisphenol A) is particularly preferred.

The diphenols can be used individually or as any desired mixtures.

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The diphenols are known in the literature or obtainable by processes known in the literature.

Examples of suitable chain terminators for the preparation of the thermoplastic aromatic polycarbonates are phenol, p-chlorophenol, p-tert-butylphenol or 2,4,6-tribromophenol, as well as long-chain alkylphenols, such as 4-(1,3-tetramethylbutyl)phenol according to DE-OS 2 842 005, or monoalkylphenols or dialkylphenols having a total of 8 to 20 C atoms in the alkyl substituents, such as 3,5-ditert-butylphenol, p-isooctylphenol, p-tert-octylphenol, p-dodecylphenol, 2-(3,5-dimethylheptyl)phenol and 4-(3,5-dimethylheptyl)phenol. The amount of chain terminators to be used is generally between 0.5 mol% and 10 mol%, based on the molar sum of the particular diphenols used.

The thermoplastic aromatic polycarbonates have mean weight-average molecular weights (M_w, measured e.g. by ultracentrifugation or light scattering) of 10,000 to 200,000 and preferably of 20,000 to 80,000.

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The thermoplastic aromatic polycarbonates can be branched in known manner, preferably by the incorporation of 0.05 to 2.0 mol%, based on the sum of the diphenols used, of trifunctional or more than trifunctional compounds, for example those with three or more phenolic groups.

Both homopolycarbonates and copolycarbonates are suitable. Copolycarbonates according to the invention as component A can also be prepared using 1 to 25 wt.% and preferably 2.5 to 25 wt.% (based on the total amount of diphenols to be used) of polydiorganosiloxanes with hydroxyaryloxy end groups. These are known (cf. for example US patent 3 419 634) or can be prepared by processes known in the literature. The preparation of copolycarbonates containing polydiorganosiloxanes is described e.g. in DE-OS 3 334 782.

- Apart from bisphenol A homopolycarbonates, preferred polycarbonates are copolycarbonates of bisphenol A with up to 15 mol%, based on the molar sum of diphenols, of other diphenols mentioned as being preferred or particularly preferred, especially 2,2-bis(3,5-dibromo-4-hydroxyphenyl) propane.
- Aromatic dicarboxylic acid dihalides for the preparation of aromatic polyestercarbonates are preferably the diacid dichlorides of isophthalic acid, terephthalic acid, diphenyl ether 4,4'-dicarboxylic acid and naphthalene-2,6-dicarboxylic acid.
- Mixtures of the diacid dichlorides of isophthalic acid and terephthalic acid in a ratio of between 1:20 and 20:1 are particularly preferred.

A carbonic acid halide, preferably phosgene, is additionally used as a bifunctional acid derivative in the preparation of polyestercarbonates.

Suitable chain terminators for the preparation of the aromatic polyestercarbonates, apart from the monophenols already mentioned, are their chlorocarbonic acid esters

and the acid chlorides of aromatic monocarboxylic acids which can optionally be substituted by C₁-C₂₂-alkyl groups or halogen atoms, as well as aliphatic C₂-C₂₂-monocarboxylic acid chlorides.

The amount of chain terminators is 0.1 to 10 mol% in each case, based on moles of diphenols for phenolic chain terminators and on moles of dicarboxylic acid dichlorides for monocarboxylic acid chloride chain terminators.

Aromatic hydroxycarboxylic acids can also be incorporated in the aromatic polyestercarbonates.

The aromatic polyestercarbonates can be both linear and branched in known manner (cf. also DE-OS 2 940 024 and DE-OS 3 007 934 in this connection).

Examples of branching agents which can be used are trifunctional or more than trifunctional carboxylic acid chlorides, such as trimesic acid trichloride, cyanuric trichloride, benzophenone-3,3',4,4'-tetracarboxylic acid tetrachloride. acid naphthalene-1,4,5,8-tetracarboxylic acid tetrachloride or pyromellitic acid tetrachloride, in amounts of 0.01 to 1.0 mol% (based on the dicarboxylic acid dichlorides used), or trifunctional or more than trifunctional phenols, such as 20 phloroglucinol, 4,6-dimethyl-2,4,6-tri(4-hydroxyphenyl)-2-heptene, 4,4-dimethyl-2,4,6-tri(4-hydroxyphenyl)heptane, 1,3,5-tri(4-hydroxyphenyl)benzene, 1,1,1-tri(4hydroxyphenyl)ethane, tri(4-hydroxyphenyl)phenylmethane, 2,2-bis[4,4-bis(4-2,4-bis(4-hydroxyphenylisopropyl)phenol, hydroxyphenyl)cyclohexyl]propane, 2,6-bis(2-hydroxy-5-methylbenzyl)-4tetra(4-hydroxyphenyl)methane, methylphenol, 2-(4-hydroxyphenyl)-2-(2,4-dihydroxyphenyl)propane, tetra(4-[4-1,4-bis[4,4'hydroxyphenylisopropyl]phenoxy)methane or (dihydroxytriphenyl)methyl]benzene, in amounts of 0.01 to 1.0 mol%, based on the diphenols used. Phenolic branching agents can be used with the diphenols, and acid chloride branching agents can be introduced together with the acid dichlorides. 30

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The proportion of carbonate structural units in the thermoplastic aromatic polyestercarbonates can vary freely. The proportion of carbonate groups is preferably up to 100 mol%, particularly up to 80 mol% and particularly preferably up to 50 mol%, based on the sum of the ester groups and carbonate groups. Both the ester part and the carbonate part of the aromatic polyestercarbonates can be present in the polycondensation product in the form of blocks or as a random distribution.

The relative solution viscosity (η_{rel}) of the aromatic polycarbonates and polyestercarbonates ranges from 1.18 to 1.4 and preferably from 1.22 to 1.3 (measured on solutions of 0.5 g of polycarbonate or polyestercarbonate in 100 ml of methylene chloride solution at 25°C).

The thermoplastic aromatic polycarbonates and polyestercarbonates can be used on their own or in any desired mixture with one another.

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Component B

The component B comprises one or more graft polymers consisting of

- B.1 5 to 95 and preferably 30 to 80 wt.% of at least one vinyl monomer to
 - B.2 95 to 5 and preferably 70 to 20 wt.% of one or more graft bases with glass transition temperatures of < 10°C, preferably of < 0°C and particularly preferably of < -20°C.

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In general the graft base B.2 has a mean particle size (d_{50} value) of 0.05 to 5 μm , preferably of 0.10 to 0.5 μm and particularly preferably of 0.20 to 0.40 μm .

Monomers B.1 are preferably mixtures of

B.1.1 50 to 99 parts by weight of vinylaromatics and/or ring-substituted vinylaromatics (for example styrene, α-methylstyrene, p-methylstyrene, p-chlorostyrene) and/or C₁-C₈-alkyl (meth)acrylates (e.g. methyl methacrylate, ethyl methacrylate), and

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B.1.2 1 to 50 parts by weight of vinyl cyanides (unsaturated nitriles such as acrylonitrile and methacrylonitrile) and/or C₁-C₈-alkyl (meth)acrylates (e.g. methyl methacrylate, n-butyl acrylate, t-butyl acrylate) and/or derivatives (such as anhydrides and imides) of unsaturated carboxylic acids (for example maleic anhydride and N-phenylmaleimide).

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Preferred monomers B.1.1 are selected from at least one of the monomers styrene, α -methylstyrene and methyl methacrylate, and preferred monomers B.1.2 are selected from at least one of the monomers acrylonitrile, maleic anhydride and methyl methacrylate.

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Particularly preferred monomers B.1.1 and B.1.2 are styrene and acrylonitrile respectively.

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Examples of suitable graft bases B.2 for the graft polymers B are diene rubbers, EP(D)M rubbers, i.e. those based on ethylene/propylene and optionally diene, and acrylate, polyurethane, silicone, chloroprene and ethylene/vinyl acetate rubbers.

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Preferred graft bases B.2 are diene rubbers (e.g. those based on butadiene, isoprene, etc.), or mixtures of diene rubbers, or copolymers of diene rubbers or mixtures thereof with other copolymerizable monomers (e.g. according to B.1.1 and B.1.2), preferably butadiene/styrene copolymers, with the proviso that the glass transition temperature of the component B.2 is <10°C, preferably <0°C and particularly preferably <-10°C.

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Pure polybutadiene rubber is particularly preferred.

Examples of particularly preferred polymers B are ABS polymers (emulsion, bulk and suspension ABS) such as those described e.g. in DE-OS 2 035 390 (= US-PS 3 644 574), in DE-OS 2 248 242 (= GB-PS 1 409 275) or in Ullmann, Enzyklopädie der Technischen Chemie, vol. 19 (1980), p. 280 et seq. The gel content of the graft base B.2 is at least 30 wt.% and preferably at least 40 wt.% (measured in toluene).

The graft copolymers B are prepared by free-radical polymerization, e.g. by emulsion, suspension, solution or bulk polymerization, preferably by emulsion polymerization or bulk polymerization.

Other particularly suitable graft rubbers are ABS polymers prepared by redox initiation with an initiator system consisting of organic hydroperoxide and ascorbic acid according to US-P 4 937 285.

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It is known that the graft monomers are not necessarily completely grafted onto the graft base in the grafting reaction, so graft polymers B are also understood according to the invention as meaning products which are obtained by (co)polymerization of the graft monomers in the presence of the graft base and appear therewith in the work-up.

Suitable acrylate rubbers B.2 of the polymers B are preferably polymers of alkyl acrylates, optionally with up to 40 wt.%, based on B.2, of other polymerizable, ethylenically unsaturated monomers. The preferred polymerizable acrylic acid esters include C₁-C₈-alkyl esters, for example methyl, ethyl, butyl, n-octyl and 2-ethylhexyl esters; halogenoalkyl esters, preferably halogeno-C₁-C₈-alkyl esters such as chloroethyl acrylate; and mixtures of these monomers.

Monomers having more than one polymerizable double bond can be copolymerized for the purpose of crosslinking. Preferred examples of crosslinking monomers are esters of unsaturated monocarboxylic acids having 3 to 8 C atoms and unsaturated

monohydric alcohols having 3 to 12 C atoms or saturated polyols having 2 to 4 OH groups and 2 to 20 C atoms, e.g. ethylene glycol dimethacrylate and allyl methacrylate; polyunsaturated heterocyclic compounds, e.g. trivinyl and triallyl cyanurate; polyfunctional vinyl compounds such as di- and trivinylbenzenes; and also triallyl phosphate and diallyl phthalate.

Preferred crosslinking monomers are allyl methacrylate, ethylene glycol dimethacrylate, diallyl phthalate and heterocyclic compounds having at least 3 ethylenically unsaturated groups.

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Particularly preferred crosslinking monomers are the cyclic monomers triallyl cyanurate, triallyl isocyanurate, triacryloylhexahydro-s-triazine and triallylbenzenes. The amount of crosslinked monomers is preferably 0.02 to 5 and particularly 0.05 to 2 wt.%, based on the graft base B.2.

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In the case of cyclic crosslinking monomers having at least 3 ethylenically unsaturated groups, it is advantageous to restrict their amount to below 1 wt.% of the graft base B.2.

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Preferred examples of "other" polymerizable, ethylenically unsaturated monomers that can optionally be used, apart from the acrylic acid esters, for the preparation of the graft base B.2 are acrylonitrile, styrene, α -methylstyrene, acrylamides, vinyl C_1 - C_6 -alkyl ethers, methyl methacrylate and butadiene. Preferred acrylate rubbers as the graft base B.2 are emulsion polymers having a gel content of at least 60 wt.%.

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Other suitable graft bases B.2 are silicone rubbers with graft-active sites, such as those described in DE-OS 3 704 657, DE-OS 3 704 655, DE-OS 3 631 540 and DE-OS 3 631 539.

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

The mean particle size d₅₀ is the diameter above which 50 wt.% of the particles fall and below which 50 wt.% of the particles fall. It can be measured by ultracentrifugation (W. Scholtan, H. Lange, Kolloid, Z. und Z. Polymere 250 (1972), 782-1796).

10 Component C

The component C comprises one or more thermoplastic vinyl (co)polymers C.1 and/or polyalkylene terephthalates C.2.

Suitable vinyl (co)polymers C.1 are polymers of at least one monomer from the group comprising vinylaromatics, vinyl cyanides (unsaturated nitriles), C₁-C₈-alkyl (meth)acrylates, unsaturated carboxylic acids and derivatives (such as anhydrides and imides) of unsaturated carboxylic acids. Particularly suitable (co)polymers are those consisting of

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C.1.1 50 to 99 and preferably 60 to 80 parts by weight of vinylaromatics and/or ring-substituted vinylaromatics (for example styrene, α-methylstyrene, p-methylstyrene, p-chlorostyrene) and/or C₁-C₈-alkyl (meth)acrylates (e.g. methyl methacrylate, ethyl methacrylate), and

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C.1.2 1 to 50 and preferably 20 to 40 parts by weight of vinyl cyanides (unsaturated nitriles) (such as acrylonitrile and methacrylonitrile) and/or C₁-C₈-alkyl (meth)acrylates (e.g. methyl methacrylate, n-butyl acrylate, t-butyl acrylate) and/or unsaturated carboxylic acids (such as maleic acid) and/or derivatives (such as anhydrides and imides) of unsaturated carboxylic acids (for example maleic anhydride and N-phenylmaleimide).

The (co)polymers C.1 are resinous, thermoplastic and rubber-free.

The copolymer of styrene as C.1.1 and acrylonitrile as C.1.2 is particularly preferred.

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The (co)polymers C.1 are known and can be prepared by free-radical polymerization, especially by emulsion, suspension, solution or bulk polymerization. The (co)-polymers preferably have molecular weights M_w (weight-average, determined by light scattering or sedimentation) of between 15,000 and 200,000.

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The polyalkylene terephthalates of the component C.2 are products resulting from the reaction of aromatic dicarboxylic acids or their reactive derivatives, such as dimethyl esters or anhydrides, with aliphatic, cycloaliphatic or araliphatic diols, and mixtures of these reaction products.

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Preferred polyalkylene terephthalates contain at least 80 wt.% and preferably at least 90 wt.%, based on the dicarboxylic acid component, of terephthalic acid radicals and at least 80 wt.% and preferably at least 90 mol%, based on the diol component, of ethylene glycol and/or 1,4-butanediol radicals.

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Apart from terephthalic acid radicals, the preferred polyalkylene terephthalates can contain up to 20 mol% and preferably up to 10 mol% of radicals of other aromatic or cycloaliphatic dicarboxylic acids having 8 to 14 C atoms or of aliphatic dicarboxylic acids having 4 to 12 C atoms, e.g. radicals of phthalic acid, isophthalic acid, naphthalene-2,6-dicarboxylic acid, biphenyl-4,4'-dicarboxylic acid, succinic acid, adipic acid, sebacic acid, azelaic acid and cyclohexanediacetic acid.

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Apart from ethylene glycol or 1,4-butanediol radicals, the preferred polyalkylene terephthalates can contain up to 20 mol% and preferably up to 10 mol% of other aliphatic diols having 3 to 12 C atoms or cycloaliphatic diols having 6 to 21 C atoms, e.g. radicals of 1,3-propanediol, 2-ethyl-1,3-propanediol, neopentyl glycol,

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1,5-pentanediol, 1,6-hexanediol, cyclohexane-1,4-dimethanol, 3-ethyl-2,4-pentanediol, 2-methyl-2,4-pentanediol, 2,2,4-trimethyl-1,3-pentanediol, 2-ethyl-1,3-hexanediol, 2,2-diethyl-1,3-propanediol, 2,5-hexanediol, 1,4-di(β-hydroxyethoxy)benzene, 2,2-bis(4-hydroxycyclohexyl)propane, 2,4-dihydroxy-1,1,3,3-tetramethylcyclobutane, 2,2-bis(4-β-hydroxyethoxyphenyl)propane and 2,2-bis(4-hydroxypropoxyphenyl)propane (DE-OS 2 407 674, 2 404 776, 2 715 932).

The polyalkylene terephthalates can be branched by the incorporation of relatively small amounts of trihydric or tetrahydric alcohols or tribasic or tetrabasic carboxylic acids, e.g. according to DE-OS 1 900 270 and US-PS 3 692 744. Examples of preferred branching agents are trimesic acid, trimellitic acid, trimethylolethane, trimethylolpropane and pentaerythritol.

Particularly preferred polyalkylene terephthalates are those prepared only from terephthalic acid and its reactive derivatives (e.g. its dialkyl esters) and ethylene glycol and/or 1,4-butanediol, and mixtures of these polyalkylene terephthalates.

Mixtures of polyalkylene terephthalates contain 1 to 50 wt.% and preferably 1 to 30 wt.% of polyethylene terephthalate and 50 to 99 wt.% and preferably 70 to 99 wt.% of polybutylene terephthalate.

The polyalkylene terephthalates that are preferably used generally have an intrinsic viscosity of 0.4 to 1.5 dl/g and preferably of 0.5 to 1.2 dl/g, measured in phenol/o-dichlorobenzene (1:1 parts by weight) at 25°C in an Ubbelohde viscometer.

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The polyalkylene terephthalates can be prepared by known methods (cf. for example Kunststoff-Handbuch, volume VIII, p. 695 et seq., Carl-Hanser-Verlag, Munich 1973).

Component D

The moulding compounds according to the invention contain, as flameproofing agents, phosphorus compounds of formula (I):

in which the radicals are as defined above.

The phosphorus compounds of the component D that are suitable according to the invention are generally known (cf. for example Ullmanns Encyklopädie der Technischen Chemie, vol. 18, p. 301 et seq., 1979; Houben-Weyl, Methoden der Organischen Chemie, vol. 12/1, p. 43; Beilstein, vol. 6, p. 177).

Preferred substituents R¹ to R⁴ include methyl, butyl, octyl, chloroethyl, 2-chloropropyl, 2,3-dibromopropyl, phenyl, cresyl, cumyl, naphthyl, chlorophenyl, bromophenyl, pentachlorophenyl and pentabromophenyl. Methyl, ethyl, butyl, phenyl and naphthyl are particularly preferred.

The aromatic groups R^1 , R^2 , R^3 and R^4 can be substituted by halogen and/or C_1 - C_4 -alkyl. Particularly preferred aryl radicals are cresyl, phenyl, xylenyl, propylphenyl or butylphenyl, as well as the brominated and chlorinated derivatives thereof.

R⁵ and R⁶ independently of one another are preferably methyl or bromine.

Y is preferably C₁-C₇-alkylene and particularly isopropylidene or methylene.

n in formula (I) independently of one another can be 0 or 1 and are preferably 1.

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q can be 0, 1, 2, 3 or 4, preferably 0, 1 or 2 and particularly preferably 0.

N can assume values of 0 to 10, preferably of 0.5 to 5 and particularly of 0.9 to 3. Mixtures of different phosphates can also be used as the component D according to the invention. In this case N has an average value. This mixture can also contain monophosphorus compounds (N = 0).

The mean values of N can be determined by using a suitable method [gas chromatography (GC), high pressure liquid chromatography (HPLC), gas permeation chromatography (GPC)] to determine the composition (molecular weight distribution) of the phosphate mixture and calculating the mean values of N from this.

15 Component E

Fluorinated polyolefins are added as a further component.

The fluorinated polyolefins E are high-molecular and have glass transition temperatures above -30°C and normally above 100°C, fluorine contents preferably 20 of 65 to 76 and particularly of 70 to 76 wt.% and mean particle diameters d₅₀ of 0.05 to 1000 and preferably of 0.08 to 20 µm. The fluorinated polyolefins E generally have a density of 1.2 to 2.3 g/cm³. Preferred fluorinated polyolefins E are polyvinylidene polytetrafluoroethylene, fluoride and tetrafluoroethylene/hexafluoropropylene and ethylene/tetrafluoroethylene The fluorinated polyolefins are known (cf. "Vinyl and Related Polymers" by Schildknecht, John Wiley & Sons, Inc., New York, 1962, pages 484-494; "Fluoropolymers" by Wall, Wiley-Interscience, John Wiley & Sons, Inc., New York, volume 13, 1970, pages 623-654; "Modern Plastics Encyclopedia", 1970-1971, volume 47, no. 10 A, October 1970, McGraw-Hill, Inc., New York, pages 134 30 and 774; "Modern Plastics Encyclopedia", 1975-1976, October 1975, volume 52, no.

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10 A, McGraw-Hill, Inc., New York, pages 27, 28 and 472; and US-PS 3 671 487, 3 723 373 and 3 838 092).

They can be prepared by known processes, for example by the polymerization of tetrafluoroethylene in an aqueous medium with a catalyst that forms free radicals, for example sodium, potassium or ammonium peroxydisulfate, at pressures of 7 to 71 kg/cm² and at temperatures of 0 to 200°C and preferably of 20 to 100°C. (For further details see e.g. US patent 2 393 967.) Depending on the form in which they are used, the density of these materials can be between 1.2 and 2.3 g/cm³ and the mean particle size can be between 0.5 and 1000 µm.

Fluorinated polyolefins E which are preferred according to the invention are used in the form of an emulsion with a mean particle diameter of 0.05 to 20 μm and preferably of 0.08 to 10 μm and a density of 1.2 to 1.9 g/cm³, or in the form of a powder with a mean particle diameter of 100 to 1000 μm and densities of 2.0 g/cm³ to 2.3 g/cm³.

According to the invention, the fluorinated polyolefins E are preferably used in the form of special formulations.

According to the invention, the mixtures of fluorinated polyolefin and polyalkyl (meth)acrylates, preferably poly-C₁-C₈-alkyl (meth)acrylates, can be obtained in the following ways:

as a coagulated mixture of an emulsion of poly-C₁-C₈-alkyl (meth)acrylates with an emulsion of the fluorinated polyolefin. Special embodiments are also suitable here, for example polymerization of the polyalkyl (meth)acrylate in the presence of an emulsion of the fluorinated polyolefin, followed by joint precipitation.

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as a preliminary compound of a fluorinated polyolefin and a polyalkyl (meth)acrylate, the components being compounded in the melt, generally at temperatures of 200°C to 330°C, in the conventional mechanical units such as internal kneaders, extruders or double-shaft screws.

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In terms of the present invention, polyalkyl (meth)acrylates are preferably synthesized from at least one monomer selected from C₁-C₈-alkyl methacrylates, preferably C₁-C₄-alkyl methacrylates and particularly methyl methacrylate, and C₁-C₈-alkyl acrylates, preferably C₁-C₄-alkyl acrylates and particularly ethyl acrylate. Polyalkyl (meth)acrylate can take the form of a homopolymer or a copolymer. Copolymers preferably contain methyl methacrylate and up to approx. 30 wt.% and preferably 3 to 30 wt.% of one or more C₁-C₄-alkyl acrylates as comonomers.

Polymethyl methacrylate is particularly preferred. The polyalkyl (meth)acrylates are generally known (e.g. US-A 5,292,786).

In the mixture the ratio of polyalkyl (meth)acrylate to fluorinated polyolefin E is 95:5 to 5:95 and preferably 90:10 to 40:60. The emulsion mixture is then coagulated in known manner, for example by spray drying, freeze drying or coagulation by the addition of inorganic or organic salts, acids or bases or organic water-miscible solvents such as alcohols or ketones, at temperatures preferably of 20 to 150°C and particularly of 50 to 100°C. If necessary, the ingredients can be dried at 50 to 200°C and preferably at 70 to 100°C.

Suitable tetrafluoroethylene polymer emulsions are commercially available products and are sold for example by DuPont (Wilmington, Delaware, USA) as Teflon® 30 N or by Dyneon GmbH (Burgkichen, Germany) as Hostaflon®.

Suitable fluorinated polyolefin powders are commercially available products and are sold for example by DuPont as Teflon[®] CFP 6000 N or by Dyneon GmbH (Burgkichen, Germany) as Hostaflon[®] TF 2071.

5 Component F

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The moulding compounds according to the invention can contain at least one of the conventional additives, such as lubricants and demoulding agents, for example pentaerythritol tetrastearate, nucleating agents, antistatic agents, stabilizers, fillers and reinforcing agents, as well as dyes and pigments.

The filled or reinforced moulding compounds can contain up to 60 and preferably 1 to 40 wt.%, based on the filled or reinforced moulding compound, of fillers and/or reinforcing agents. Preferred reinforcing agents are glass fibres. Preferred fillers, which can also have a reinforcing effect, are glass spheres, mica, silicates, quartz, talcum, titanium dioxide and wollastonite.

Preferred fillers and reinforcing agents are very finely divided mineral particles with an anisotropic particle geometry.

Mineral particles with an anisotropic particle geometry are understood as meaning particles whose so-called aspect ratio - the ratio of the largest to the smallest particle dimension - is greater than 1, preferably greater than 2 and particularly preferably greater than about 5. Such particles are platelet-like or fibrous, at least in the broadest sense. Such materials include e.g. certain talcums and certain (alumino)-silicates with a sheet or fibre geometry, such as bentonite, wollastonite, mica, kaolin, hydrotalcite, hectorite or montmorillonite.

Preferably, it is also possible to use inorganic materials of scale-like or platelet-like character, such as talcum, mica/clay sheet minerals, montmorillonite (the latter also in an organophilic form modified by ion exchange), kaolin and vermiculite.

Furthermore, the mineral particles can be surface-modified, for example silanized, with organic molecules in order to improve compatibility with the polymers. Hydrophobic or hydrophilic surfaces can be produced in this way.

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The moulding compounds according to the invention can contain up to 35 wt.%, based on the total moulding compound, of another flameproofing agent optionally having a synergistic action. Examples of other flameproofing agents which may be mentioned are organic halogen compounds such as decabromodiphenyl ether and tetrabromobisphenol, inorganic halogen compounds such as ammonium bromide, nitrogen compounds such as melamine and melamine/formaldehyde resins, inorganic hydroxide compounds such as Mg and Al hydroxide, inorganic compounds such as antimony oxides, barium metaborate, hydroxoantimonate, zirconium oxide, zirconium hydroxide, molybdenum oxide, armmonium molybdate, zinc borate, ammonium borate, talcum, silicate, silicon oxide and tin oxide, as well as siloxane compounds. Other flameproofing agents which can also be used are monophosphate compounds, oligomeric phosphate compounds or mixtures thereof. Such phosphorus compounds are described in EP-A 363 608, EP-A 345 522 and DE-OS 197 21 628.

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The parts by weight in the present patent application are standardized so that the sum of the parts by weight of the components A to F in the composition is 100.

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The moulding compounds according to the invention, containing the components A to E and optionally other known additives such as stabilizers, dyes, pigments, lubricants and demoulding agents, nucleating agents and antistatic agents, fillers and reinforcing agents, are prepared by mixing the appropriate constituents in known manner and compounding and extruding the mixture in the melt at temperatures of 200°C to 300°C in conventional mechanical units such as internal kneaders, extruders and double-shaft screws.

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The individual constituents can be mixed in known manner, either successively or simultaneously and either at about 20°C (room temperature) or at elevated temperature.

The invention therefore also provides a process for the preparation of the moulding compounds.

The moulding compounds of the present invention can be used for the production of all kinds of moulded articles. Moulded articles can be produced in particular by injection moulding. Examples of moulded articles which can be produced are all kinds of housing parts, e.g. for domestic appliances such as juice presses, coffee machines and mixers, or for office machines such as monitors, printers and copiers, or cover plates for the building sector and parts for the motor vehicle sector. They can also be used in the field of electrotechnology because they have very good electrical properties.

The moulding compounds according to the invention can also be used for example for the production of the following moulded articles or moulded parts:

interior trim for rail vehicles, hub caps, housings for electrical equipment containing small transformers, housings for equipment for the dissemination and communication of information, housings and clothing for medical purposes, massage equipment and its housings, children's toy vehicles, wall panels, housings for safety devices, rear spoilers, thermally insulated transport containers, devices for keeping or caring for small animals, moulded parts for sanitary and bath fittings, covering grids for ventilation apertures, moulded parts for summer houses and garden sheds, and housings for garden tools.

Another form of processing is the production of moulded articles by deep drawing from previously produced slabs or sheets.

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The present invention therefore also provides the use of the moulding compounds according to the invention for the production of all kinds of moulded articles, preferably those mentioned above, and the moulded articles made from the moulding compounds according to the invention.

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Examples

Component A1

Polycarbonate based on bisphenol A with a relative solution viscosity of 1.280, measured in methylene chloride at 25°C and at a concentration of 0.5 g/100 ml.

Component A2

Polycarbonate based on bisphenol A with a relative solution viscosity of 1.255, measured in methylene chloride at 25°C and at a concentration of 0.5 g/100 ml.

Component B

Graft polymer consisting of 40 parts by weight of styrene and acrylonitrile in a ratio of 73:27 to 60 parts by weight of particulate crosslinked polybutadiene rubber (mean particle diameter $d_{50} = 0.33 \mu m$), prepared by emulsion polymerization.

Component C

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Styrene/acrylonitrile copolymer with a styrene/acrylonitrile ratio of 72:28 and an intrinsic viscosity of 0.55 dl/g (measured in dimethylformamide at 20°C).

Component D1

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A mixture of BDP and TPP in a weight ratio of 75:25.

a) Bisphenol diphosphate (BDP):

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The mean value of N is determined by first using HPLC to measure the proportions of the monomeric and oligomeric phosphates:

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Column type:

LiChrosorb RP-8

Eluents in gradient:

acetonitrile/water 50:50 to 100:0

Concentration:

5 mg/ml

- The number-weighted means are then calculated by known methods from the proportions of the individual components (monophosphates and oligophosphates).
 - b) Triphenyl phosphate (TPP): Disflamoll® TP, Bayer AG, Leverkusen, Germany

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Component D2

BDP of component D1 a).

20 Component E

E.1

Coagulated mixture of 50 parts by weight of SAN copolymer and 50 parts by weight of PTFE; Blendex 449, General Electric Plastics.

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E.2

Coagulated mixture of 50 parts by weight of polymethyl methacrylate and 50 parts by weight of PTFE; Metablen A-3800, Mitsubishi Rayon.

5 E.3

Coagulated mixture of 90 parts by weight of graft polymer (consisting of 40 parts by weight of styrene and acrylonitrile in a ratio of 73:27 to 60 parts by weight of particulate crosslinked polybutadiene rubber with a mean particle diameter d₅₀ of 0.28 µm) and 10 parts by weight of polytetrafluoroethylene polymer (Teflon[®] 30 N, DuPont, Wilmington, Delaware, USA).

Component F

HTP Ultra 5 talcum from HiTak.

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Preparation and testing of the moulding compounds according to the invention

The components are mixed with the conventional processing aids on a ZSK 25 twin-screw extruder. The moulded articles are produced on an Arburg 270E injection moulding machine at 240°C or 280°C.

The weld strength is determined at room temperature by the ISO 179 1eU method on 80 x 10 x 4 mm bars injection-moulded from both sides.

The Vicat B dimensional stability under heat is determined according to DIN 53 460 on 80 x 10 x 4 mm bars.

The modulus of elasticity and elongation at break are determined according to ISO 527/DIN 53 457.

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The fire behaviour of the specimens was measured according to UL-Subj. 94 V on 127 x 12.7 x 1.6 mm bars produced on an injection moulding machine at 260°C.

The UL 94 V test is carried out as follows:

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Specimens of substance are shaped to $127 \times 12.7 \times 1.6$ mm bars. The bars are mounted vertically so that the underside of the test piece is located 305 mm above a strip of surgical dressing. Each test bar is ignited individually by means of two consecutive ignition processes lasting 10 seconds, the burning properties are observed after each ignition process and the specimen is then assessed. The specimen is ignited using a Bunsen burner with a 10 mm (3.8 inch) high blue flame of natural gas with a calorific value of $3.73 \times 10^4 \text{ kJ/m}^3$ (1000 BTU per cubic foot).

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The UL 94 V-0 classification includes the material properties described below, tested according to UL 94 V instructions. The specimens of moulding compounds in this class may not burn for longer than 10 seconds after each application of the test

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flame; the total flaming combustion time may not exceed 50 seconds for the two flame applications for each set of specimens; the specimens may not burn away completely up to the holding clamp fixed to the upper end of the specimen; the specimens may not produce flaming drips or particles that ignite the surgical cotton located below the specimen; the specimens may not exhibit glowing combustion that persists for more than 30 seconds after removal of the test flame.

Other UL 94 classifications denote specimens which are less flame-resistant or less self-extinguishing because they produce flaming drips or particles. These classifications are designated by UL 94 V-1 and V-2. F stands for "failed" and is the classification for specimens which exhibit an afterglow time of ≥ 30 seconds.

The stress cracking behaviour (ESC behaviour) is examined on $80 \times 10 \times 4$ mm bars injection-moulded at a temperature of 260° C. The test medium used is a mixture of 60 vol.% of toluene and 40 vol.% of isopropanol. The test pieces are pre-extended by means of a circular arc template (pre-extension ϵ_x in percent) and stored in the test medium at room temperature. The stress cracking behaviour is assessed via the cracking or rupture as a function of the pre-extension in the test medium.

Table 1
Composition and properties of the moulding compounds

Based on the total composition, the PTFE content in Examples 1 to 4 is the same at 0.4 wt.% of PTFE.

Component (parts by weight)	1 (Comp.)	2	3 (Comp.)	4
A1 (PC)	68.3	68.3		-
A2 (PC)	-		70.0	70.0
B (ABS)	9.8	9.8	5.0	8.6
C (SAN)	7.1	7.1	5.0	4.6
D1 (BDP/TPP)	14.5	14.5	_	_
D2 (BDP)	-	_	12.5	12.5
E1 (SAN/PTFE)	0.8		_	-
E2 (PMMA/PTFE)		0.8	_	0.8
E3 (ABS/PTFE)	-	_	4.0	-
F (talcum)	-		3.0	3.0
Demoulding agent	0.4	0.4	0.4	0.4
Property				
Vicat B 120 [°C]	95	95	102	103
UL 94 V, 3.0 mm	V-0	V-0	-	-
afterglow time [s]	15	9	-	-
UL 94 V, 1.5 mm	V-0	V-0	-	_
afterglow time [s]	39	29		
UL 94 V, 1.2 mm	_] -	V-1	V-0
afterglow time [s]			103	36
UL 94 V, 1.0 mm		_	F	V-0
afterglow time [s]			152	34
ESC behaviour				·
rupture at ε _x [%]	1.8	2.0	2.0	2.4
Modulus of elasticity [N/mm ²].			2861	2978
Elongation at break [%]		_	33	49
a _n F weld strength ¹⁾ [kJ/m ²]	8.8	9.6	7.2	7.7

¹⁾ test bars injection-moulded at 280°C

F = failed

Claims

1. Polycarbonate moulding compounds modified with a graft polymer and containing phosphorus compounds of formula (I):

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wherein

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R¹, R², R³ and R⁴ independently of one another are optionally halogen-substituted C₁-C₈-alkyl or optionally halogen-substituted and/or alkyl-substituted C₅-C₆-cycloalkyl, C₆-C₁₀-aryl or C₇-C₁₂-aralkyl,

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n independently of one another are 0 or 1,

q

independently of one another are 0, 1, 2, 3 or 4,

N

is 0 to 10,

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R⁵ and R⁶ independently of one another are C₁-C₄-alkyl or halogen, and

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is C_1 - C_7 -alkylidene, C_1 - C_7 -alkylene, C_5 - C_{12} -cycloalkylene, C_5 - C_{12} -cycloalkylidene, -O-, -S-, -SO-, -SO₂- or -CO-,

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and fluorinated polyolefins in the form of a mixture with polyalkyl (meth)acrylates.

- 2. Moulding compounds according to Claim 1 containing the fluorinated polyolefin or polyolefin mixture as
 - 1) a coagulated mixture of an emulsion of polyalkyl (meth)acrylates with an emulsion of the fluorinated polyolefin

or

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- 2) a preliminary compound of a fluorinated polyolefin and a polyalkyl (meth)acrylate.
- 3. Moulding compounds according to Claim 1 containing 0.5 to 20 parts by weight (based on 100 parts by weight of the total composition) of a phosphorus compound of formula (I) or mixtures of such compounds.

4. Moulding compounds according to Claims 1 to 3 containing 0.01 to 3 parts by weight (based on 100 parts by weight of the total composition) of fluorinated polyolefin in the form of a coagulated mixture or a preliminary compound.

5. Moulding compounds according to Claim 4 containing 0.05 to 2 parts by weight (based on 100 parts by weight of the total composition) of fluorinated polyolefin.

- 5 6. Moulding compounds according to Claim 1 containing
 - A) 40 to 99 parts by weight of aromatic polycarbonate and/or polyester-carbonate,
 - B) 0.5 to 60 parts by weight of graft polymer consisting of

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- B.1) 5 to 95 wt.% of one or more vinyl monomers to
- B.2) 95 to 5 wt.% of one or more graft bases with a glass transition temperature of <10°C,

C) 0 to 45 parts by weight of at least one thermoplastic polymer selected from the group comprising vinyl (co)polymers and polyalkylene terephthalates,

D) 0.5 to 20 parts by weight of a phosphorus compound of formula (I):

$$R^{1} = (O)_{n} = (P^{5})_{q} = (P^{6})_{q} = (Q)_{n} = (Q)_{n}$$

wherein R¹ to R⁶, Y, n, N and q are as defined in Claim 1, and

- E) 0.01 to 3 parts by weight of fluorinated polyolefins according to Claim 1.
- 7. Moulding compounds according to one of the preceding claims wherein N in formula (I) has an average value of 0.5 to 5.
 - 8. Moulding compounds according to one of the preceding claims wherein N in formula (I) has an average value of 0.9 to 3.
- 9. Moulding compounds according to one of the preceding claims containing one or more graft polymers consisting of

- B.1 5 to 95 wt.% of at least one vinyl monomer to
- B.2 95 to 5 wt.% of one or more graft bases with glass transition temperatures of < 10°C.
- 10. Moulding compounds according to Claim 9 containing, as the vinyl monomers B.1, mixtures of
- B.1.1 50 to 99 parts by weight of vinylaromatics and/or ring-substituted vinylaromatics and/or C₁-C₈-alkyl (meth)acrylates, and
 - B.1.2 1 to 50 parts by weight of vinyl cyanides and/or C₁-C₈-alkyl (meth)acrylates and/or derivatives of unsaturated carboxylic acids.
 - 11. Moulding compounds according to one of the preceding claims containing, as the graft base B.2, a diene rubber, acrylate rubber, silicone rubber or ethylene/propylene/diene rubber, or mixtures thereof.
- 20 12. Moulding compounds according to one of the preceding claims wherein Y in formula (I) is isopropylidene or methylene.
 - 13. Moulding compounds according to Claim 12 wherein Y in formula (I) is isopropylidene.
 - 14. Moulding compounds according to Claim 1 containing polymethyl methacrylate as the polyalkyl (meth)acrylate.
- 15. Moulding compounds according to one of the preceding claims containing vinyl (co)polymers of at least one monomer from the group comprising vinylaromatics, vinyl cyanides, C₁-C₈-alkyl (meth)acrylates, unsaturated

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carboxylic acids and derivatives (such as anhydrides and imides) of unsaturated carboxylic acids.

- Moulding compounds according to one of the preceding claims containing at least one additive from the group comprising stabilizers, pigments, demoulding agents, flow aids and/or antistatic agents, fillers and reinforcing agents.
- 17. Use of the moulding compounds according to one of the preceding claims for the production of moulded articles.
 - 18. Moulded articles obtainable from moulding compounds according to one of the preceding claims.

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