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(72) **Inventeurs/Inventors:**
 BUCHHOLZ, VERA, DE;
 WENZ, ECKHARD, DE;
 ECKEL, THOMAS, DE;
 THUERMER, BURKHARD, DE;
 WITTMANN, DIETER, DE

(73) **Propriétaire/Owner:**
 BAYER MATERIALSCIENCE AG, DE

(74) **Agent:** FETHERSTONHAUGH & CO.

(54) **Titre : COMPOSITIONS DE POLYCARBONATE IGNIFUGÉES A RESILIENCE MODIFIÉE**
 (54) **Title: FLAMEPROOF IMPACT-RESISTANCE MODIFIED POLYCARBONATE COMPOSITIONS**

(57) **Abrégé/Abstract:**

The present invention relates to impact-strength-modified polycarbonate compositions comprising: (A) 60 - 85 parts by weight of a branched aromatic polycarbonate, a branched aromatic polyester carbonate or a combination thereof; (B) 1 - 25 parts by weight of a graft polymer comprising at least one graft base selected from the group consisting of a silicone rubber and a silicone-acrylate rubber; (C) 9 - 18 parts by weight of talc; (D) 0.4 - 20 parts by weight of a phosphorus-containing flameproofing agent; (E) 1 - 6 parts by weight of at least one inorganic boron compound selected from the group consisting of $Zn_4B_2O_7 \cdot H_2O$, $Zn_2B_6O_{11} \cdot 3.5H_2O$ and $ZnB_4O_7 \cdot 4H_2O$ and; (F) 0 - 3 parts by weight of an anti-dripping agent, wherein the sum of the parts by weight of components (A), (B), (C), (D), (E) and (F) are normalised to 100. The compositions satisfy enhanced fire-protection requirements when used for producing moulded articles or thermoformed moulded articles.



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Abstract

The present invention relates to impact-strength-modified polycarbonate compositions comprising: (A) 60 - 85 parts by weight of a branched aromatic polycarbonate, a branched aromatic polyester carbonate or a combination thereof; (B) 1 - 25 parts by weight of a graft polymer comprising at least one graft base selected from the group consisting of a
5 silicone rubber and a silicone-acrylate rubber; (C) 9 - 18 parts by weight of talc; (D) 0.4 - 20 parts by weight of a phosphorus-containing flameproofing agent; (E) 1 - 6 parts by weight of at least one inorganic boron compound selected from the group consisting of $Zn_4B_2O_7 \cdot H_2O$, $Zn_2B_6O_{11} \cdot 3.5H_2O$ and $ZnB_4O_7 \cdot 4H_2O$ and; (F) 0 - 3 parts by weight of an anti-dripping
10 agent, wherein the sum of the parts by weight of components (A), (B), (C), (D), (E) and (F) are normalised to 100. The compositions satisfy enhanced fire-protection requirements when used for producing moulded articles or thermoformed moulded articles.

Flameproof impact-resistance modified polycarbonate compositions

The present invention relates to impact-strength-modified polycarbonate compositions that satisfy enhanced fire-protection requirements, to a process for their production, and
5 to their use for producing moulded articles.

PC/ABS blends are described in JP-A 11-199768 that have been made flame-retardant with monomeric and oligomeric phosphoric esters, the flame resistance being distinctly improved by addition of an inorganic filler such as talc, for example. However, the
10 reduction in the phosphate content that can be accomplished by this means, with unchanged flame resistance, is insufficient to obtain the melt viscosities that are necessary for extrusion applications. Furthermore, the inorganic filler generally has an adverse effect on the mechanical properties, in particular on the toughness of the polycarbonate blend.

15

US-A 5,849,827 and WO 99/07782 describe PC/ABS moulding compositions that have been made flame-retardant with resorcinol-based or bisphenol-A-based oligophosphate, the burning-times being distinctly reduced by addition of nanoscale inorganic materials in low concentrations. But the moulding compositions that are described herein also
20 have a melt stability that is inadequate for extrusion applications.

WO 99/57198 describes PC/ABS moulding compositions that have been made flame-retardant with a resorcinol-derived oligophosphate and that are distinguished by a very low TeflonTM content of only 0.1 wt.%, corresponding to a fluorine content of 0.076 %.
25 Linear and branched polycarbonates with a high molecular weight (31,000 g/mol or 32,000 g/mol) find application in the moulding compositions. The rheological properties of the moulding compositions that are described (MVR) permit processing in the extrusion process. However, the moulding compositions are distinguished by a weakness in their ESC behaviour and in their thermostability, particularly when sufficient
30 flameproofing agent is employed to achieve a sufficient flame resistance also in the case of thin wall thicknesses.

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US 2002/0077417 A1 discloses flame-retardant polycarbonate resin compositions consisting of branched polycarbonate, a silicone/acrylate composite graft polymer, oligomeric phosphoric ester, polytetrafluoroethylene and, optionally, talc. Oligomeric phosphoric esters of the BDP type are not disclosed.

5

WO 02/100948 A1 discloses thermoplastic moulding compositions containing branched polycarbonate, graft polymer, talc with a mean particle size below 1000 nm and also, optionally, oligophosphates, vinyl copolymers and anti-dripping agents.

10 WO 01/48074 A1 discloses thermoplastic moulding compositions containing branched polycarbonate, graft polymer, talc having a special purity and also, optionally, oligophosphates, vinyl copolymers and anti-dripping agents.

EP 0 675 001 A1 discloses compositions containing a thermoplastic resin (such as polycarbonate, for example) and water-eliminating compounds, wherein phosphorus-
15 containing flameproofing agents, talc, acrylonitrile-butadiene-styrene copolymer (ABS) and styrene-acrylonitrile copolymer (SAN) may optionally be included as further components. These resin compositions are suitable for marking by laser. Corresponding compositions containing silicone(-acrylate) graft polymers are not disclosed.

20 The present invention relates to a chlorine-free and bromine-free moulding composition that satisfies both particularly high flame-resistance requirements and the requirements placed on materials in American rail vehicles (Docket 90-A), and that, by reason of a high melt stability, can be processed in the extrusion process. In particular, the moulding composition according to Docket 90-A in ASTM E 162 must
25 not display any burning dripping and must exhibit a flame-spread index I_s of less than 35 and also according to ASTM E 662 must exhibit a low smoke density (D_s 1.5 min < 100 and D_s 4 min < 200). At the same time, the moulding compositions are to exhibit a tensile modulus of elasticity of at least 3500 N/mm^2 , in order to guarantee sufficient mechanical strength.

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Surprisingly, it has been found that compositions containing

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- A) 40 - 95 parts by weight, preferably 60 - 85 parts by weight, particularly preferably 65 - 78 parts by weight, of branched aromatic polycarbonate and/or branched aromatic polyester carbonate,
- B) 1 - 25 parts by weight, preferably 2 - 9 parts by weight, particularly preferably 4 - 8 parts
5 by weight, quite particularly preferably 4.7 - 6.6 parts by weight, of a graft polymer including one or more graft bases (B.2) selected from the group comprising the silicone rubbers (B.2.1) and silicone-acrylate rubbers (B.2.2),
- C) 9 - 18 parts by weight, preferably 10 - 15 parts by weight, particularly preferably 10 - 12 parts by weight, of talc,
- 10 D) 0.4 - 20 parts by weight, preferably 6 - 17 parts by weight, particularly preferably 8 - 12 parts by weight, of phosphorus-containing flameproofing agents,
- E) 0.5 - 20 parts by weight, preferably 1 - 10 parts by weight, particularly preferably 1 - 6 parts by weight, quite particularly preferably 2 - 4 parts by weight, of one or more inorganic boron compounds,
- 15 F) 0 - 3 parts by weight, preferably 0.01 - 1 parts by weight, particularly preferably 0.1 - 0.6 parts by weight, of anti-dripping agents,

where all the parts-by-weight data in the present application are normalised in such a way that the sum of the parts by weight of all the components in the composition is 100, have the desired range of properties.

20 In one composition aspect, the invention relates to a composition comprising:

(A) 60 - 85 parts by weight of a branched aromatic polycarbonate, a branched aromatic polyester carbonate or a combination thereof;

(B) 1 - 25 parts by weight of a graft polymer comprising at least one graft base selected from the group consisting of a silicone rubber and a silicone-acrylate rubber;

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(C) 9 - 18 parts by weight of talc;

(D) 0.4 - 20 parts by weight of a phosphorus-containing flameproofing agent;

(E) 1 - 6 parts by weight of at least one inorganic boron compound selected from the group consisting of $Zn_4B_2O_7 \cdot H_2O$, $Zn_2B_6O_{11} \cdot 3.5H_2O$ and $ZnB_4O_7 \cdot 4H_2O$; and

5 (F) 0 - 3 parts by weight of an anti-dripping agent, wherein the sum of the parts by weight of components (A), (B), (C), (D), (E) and (F) are normalised to 100.

Component A

10 Branched aromatic polycarbonates and/or branched aromatic polyester carbonates according to component A that are suitable in accordance with the invention are known from the literature or can be produced by processes known from the literature (on the production of aromatic polycarbonates, see, for example, Schnell, "Chemistry and Physics of Polycarbonates", Interscience Publishers, 1964, and also DE-AS 1 495 626, DE-A 2 232 877, DE-A 2 703 376, DE-A 2 714 544, DE-A 3 000 610, DE-A 3 832 396; on the production of aromatic polyester carbonates, see, for example, DE-A 3 077 934).

The production of aromatic poly(ester) carbonates is undertaken, for example, by conversion of diphenols with carbonic acid halides, preferentially phosgene, and/or with aromatic dicarboxylic acid dihalides, preferentially benzenedicarboxylic acid dihalides, by the interphase process, optionally using chain stoppers, for example monophenols, and using trifunctional or tetrafunctional phenolic branching agents that may also include amine functionalities by way of active functional groups, the branching coming about in this case by virtue of amide linkages. Suitable as branching agents are, for example, triphenols or tetraphenols, and in preferred manner also those phenolic branching agents having at least three functional groups with graduated reactivity that are suitable for a condensation reaction. Also suitable as branching agent is 1,1,1-tris(p-hydroxyphenyl)ethane.

Isatin bicresol finds application in particularly preferred manner by way of branching agent.

15

The branching agents are employed in a quantity from 0.01 mol.% to 5 mol.%, preferably from 0.02 mol.% to 2 mol.%, in particular from 0.05 mol.% to 1 mol.%, particularly preferably from 0.1 mol.% to 0.5 mol.%, relative to the sum of diphenol and branching agent in the poly(ester) carbonate.

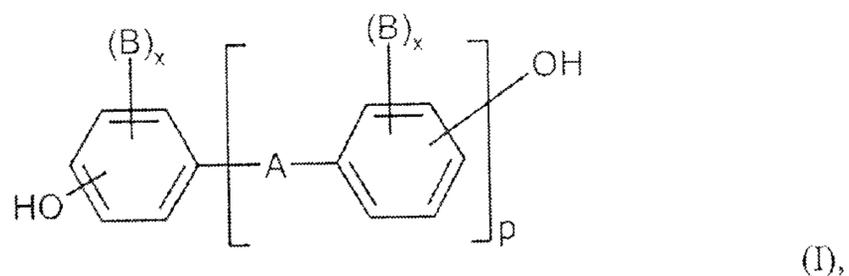
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Branched polycarbonates that are suitable in accordance with the invention may also be produced by the known melt-polymerisation process, by conversion of diphenolic compounds with diphenyl carbonate using the aforementioned branching agents and chain stoppers.

25

Diphenols for producing the branched aromatic polycarbonates and/or aromatic polyester carbonates are preferentially those of the formula (I)

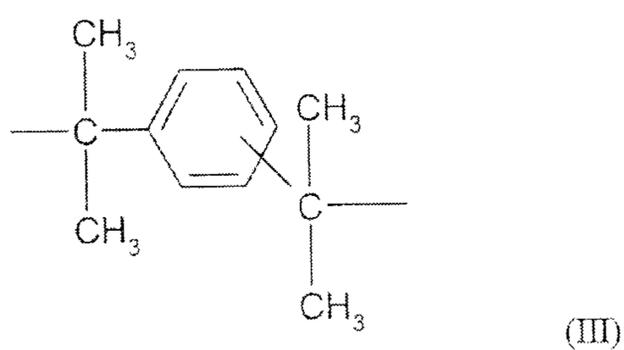
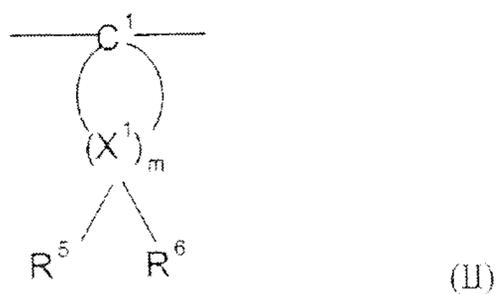
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wherein

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

or a residue of the formula (II) or (III)



10

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

15 x are, in each instance, independently of one another, 0, 1 or 2,

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p is 1 or 0, and

R⁵ and R⁶ are individually selectable for each X¹ and are, independently of one another,
5 hydrogen or C₁ to C₆ alkyl, preferentially hydrogen, methyl or ethyl,

X¹ is carbon and

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

Preferred diphenols are hydroquinone, resorcinol, dihydroxydiphenols,
bis(hydroxyphenyl)-C₁-C₅-alkanes, bis(hydroxyphenyl)-C₅-C₆-cycloalkanes,
bis(hydroxyphenyl)ethers, bis(hydroxyphenyl)sulfoxides, bis(hydroxyphenyl)ketones,
15 bis(hydroxyphenyl)sulfones and α,α -bis(hydroxyphenyl)diisopropylbenzenes and also
the ring-brominated and/or ring-chlorinated derivatives thereof.

Particularly preferred diphenols are 4,4'-dihydroxydiphenyl, bisphenol A, 2,4-bis(4-
hydroxyphenyl)-2-methylbutane, 1,1-bis(4-hydroxyphenyl)cyclohexane, 1,1-bis(4-
20 hydroxyphenyl)-3,3,5-trimethylcyclohexane, 4,4'-dihydroxydiphenylsulfide, 4,4'-
dihydroxydiphenylsulfone and also the dibrominated and tetrabrominated or chlorinated
derivatives thereof, such as, for example, 2,2-bis(3-chloro-4-hydroxyphenyl)propane,
2,2-bis(3,5-dichloro-4-hydroxyphenyl)propane or 2,2-bis(3,5-dibromo-4-
hydroxyphenyl)propane. Particularly preferred is 2,2-bis(4-hydroxyphenyl)propane
25 (bisphenol A).

The diphenols may be employed individually or in the form of arbitrary mixtures. The
diphenols are known from the literature or can be obtained by processes known from the
literature.

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Suitable chain stoppers for the production of the thermoplastic, aromatic branched polycarbonates are, for example, phenol, p-chlorophenol, p-tert-butylphenol or 2,4,6-tribromophenol, but also long-chain alkylphenols, such as 4-(1,1,3,3-tetramethylbutyl)phenol according to DE-A 2 842 005 or monoalkylphenol or dialkylphenols with a total of 8 to 5 20 C atoms in the alkyl substituents, such as 3,5-di-tert-butylphenol, p-iso-octylphenol, p-tert-octylphenol, p-dodecylphenol and 2-(3,5-dimethylheptyl)phenol and 4-(3,5-dimethylheptyl)phenol. The quantity of chain stoppers to be employed amounts generally to between 0.5 mol.% and 10 mol.%, relative to the molar sum of the diphenols to be employed in the given case.

10

Besides the monophenols already named, the chlorocarbonic esters thereof and also the acid chlorides of aromatic monocarboxylic acids, which may optionally be substituted by C₁ to C₂₂ alkyl groups or by halogen atoms, and also aliphatic C₂ to C₂₂ monocarboxylic acid chlorides, also come into consideration as chain stoppers for the production of the 15 aromatic polyester carbonates.

The quantity of chain stoppers amounts in each instance to 0.1 mol.% to 10 mol.%, relative, in the case of the phenolic chain stoppers, to moles of diphenols and, in the case of monocarboxylic acid chlorides, to moles of dicarboxylic acid dichlorides.

20

The aromatic polyester carbonates may also include, in integrated manner, aromatic hydroxycarboxylic acids.

The proportion of carbonate structural units in the thermoplastic, aromatic polyester 25 carbonates may vary arbitrarily. The proportion of carbonate groups amounts preferentially to up to 100 mol.%, in particular up to 80 mol.%, particularly preferably up to 50 mol.%, relative to the sum of ester groups and carbonate groups. Both the ester portion and the carbonate portion of the aromatic polyester carbonates may be present in the polycondensate in the form of blocks or in randomly distributed manner.

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The thermoplastic, aromatic branched polycarbonates and polyester carbonates may be employed on their own or in an arbitrary mixture. Preferred compositions according to the invention are free from linear polycarbonates and polyester carbonates.

- 5 The relative solution viscosities of the poly(ester)carbonates that are suitable in accordance with the invention lie within the range from 1.20 to 1.50, preferably from 1.24 to 1.40, in particular from 1.25 to 1.35, measured in CH_2Cl_2 as solvent at 25 °C and at a concentration of 0.5 g/100 ml.

10 Component B

Component B comprises one or more graft polymers of

- 15 B.1 5 wt.% to 95 wt.%, preferentially 10 wt.% to 90 wt.%, of one or more vinyl monomers on

- B.2 95 wt.% to 5 wt.%, preferentially 90 wt.% to 10 wt.%, of one or more graft bases selected from the group comprising the silicone rubbers (B.2.1) and silicone-acrylate rubbers (B.2.2).

20

The graft polymers B are produced by radical polymerisation, for example by emulsion polymerisation, suspension polymerisation, solution polymerisation or bulk polymerisation, preferentially by emulsion polymerisation or bulk polymerisation.

- 25 Suitable monomers B.1 are vinyl monomers such as vinyl aromatics and/or ring-substituted vinyl aromatics (such as styrene, α -methylstyrene, p-methylstyrene, p-chlorostyrene), (C₁-C₈)-alkyl methacrylates (such as methyl methacrylate, ethyl methacrylate, 2-ethylhexyl methacrylate, allyl methacrylate), (C₁-C₈)-alkyl acrylates (such as methyl acrylate, ethyl acrylate, n-butyl acrylate, t-butyl acrylate), organic acids
30 (such as acrylic acid, methacrylic acid), and/or vinyl cyanides (such as acrylonitrile and methacrylonitrile), and/or derivatives (such as anhydrides and imides) of unsaturated

carboxylic acids (for example, maleic anhydride and N-phenyl maleimide). These vinyl monomers may be used on their own or in mixtures of at least two monomers.

Preferred monomers B.1 are selected from at least one of the monomers styrene, α -methylstyrene, methyl methacrylate, n-butyl acrylate and acrylonitrile. Methyl methacrylate is employed in particularly preferred manner as monomer B.1.

The glass transition temperature of the graft base B.2 amounts to $< 10\text{ }^{\circ}\text{C}$, preferentially $< 0\text{ }^{\circ}\text{C}$, particularly preferably $< -20\text{ }^{\circ}\text{C}$. The graft base B.2 generally has a mean particle size (d_{50} value) from $0.05\text{ }\mu\text{m}$ to $10\text{ }\mu\text{m}$, preferentially $0.06\text{ }\mu\text{m}$ to $5\text{ }\mu\text{m}$, particularly preferably $0.08\text{ }\mu\text{m}$ to $1\text{ }\mu\text{m}$.

The mean particle size d_{50} is that diameter, above and below which 50 wt.%, respectively, of the particles lie; it can be determined by means of ultracentrifuge measurement (W. Scholtan, H. Lange, *Kolloid-Z. und Z. Polymere* 250 (1972), 782-796).

Suitable silicone rubbers according to B.2.1 are silicone rubbers with graft-active sites, the method of production of which is described, for example, in US 2,891,920, US 3,294,725, DE-OS 3 631 540, EP 249 964, EP 430 134 and US 4,888,388.

20

The silicone rubber according to B.2.1 is preferably produced by emulsion polymerisation, wherein siloxane monomer units, cross-linking or branching agents (IV) and optionally grafting agents (V) are employed.

25 Dimethylsiloxane or cyclic organosiloxanes with at least 3 ring members, preferentially 3 to 6 ring members, are employed, for example, and preferably, as siloxane-monomer structural units, such as, for example, and preferably, hexamethyl cyclotrisiloxane, octamethyl cyclotetrasiloxane, decamethyl cyclopentasiloxane, dodecamethyl cyclohexasiloxane, trimethyltriphenyl cyclotrisiloxanes, tetramethyltetraphenyl
30 cyclotetrasiloxanes, octaphenyl cyclotetrasiloxane.

The organosiloxane monomers may be employed on their own or in the form of mixtures with 2 or more monomers. The silicone rubber preferentially contains not less than 50 wt.%, and particularly preferably not less than 60 wt.%, organosiloxane, relative to
5 the total weight of the silicone-rubber component.

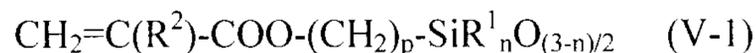
Use is preferentially made of silane-based cross-linking agents with a functionality of 3 or 4, particularly preferably 4, by way of cross-linking or branching agents (IV). The following may be named in exemplary and preferential manner: trimethoxymethylsilane,
10 triethoxyphenylsilane, tetramethoxysilane, tetraethoxysilane, tetra-n-propoxysilane and tetrabutoxysilane. The cross-linking agent may be employed on its own or in a mixture of two or more. Tetraethoxysilane is particularly preferred.

The cross-linking agent is employed in a quantitative range between 0.1 wt.% and
15 40 wt.%, relative to the total weight of the silicone-rubber component. The quantity of cross-linking agent is chosen in such a way that the degree of swelling of the silicone rubber, measured in toluene, lies between 3 and 30, preferably between 3 and 25, and particularly preferably between 3 and 15. The degree of swelling is defined as the weight ratio of the quantity of toluene that is absorbed by the silicone rubber when it is saturated
20 with toluene at 25 °C to the quantity of silicone rubber in the dried state. The ascertainment of the degree of swelling is described in detail in EP 249 964.

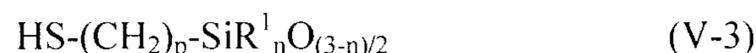
If the degree of swelling is less than 3, i.e. if the content of cross-linking agent is too high, the silicone rubber does not display adequate rubber-like elasticity. If the swelling
25 index is greater than 30, the silicone rubber is unable to form a domain structure in the matrix polymer and is therefore also unable to enhance impact strength; the effect would then be similar to a simple addition of polydimethylsiloxane.

Tetrafunctional cross-linking agents are preferred over trifunctional cross-linking agents,
30 because the degree of swelling is then easier to control within the limits described above.

Suitable as grafting agents (V) are compounds that are capable of forming structures having the following formulae:



5



10 wherein

R^1 stands for C_1 - C_4 -alkyl, preferentially methyl, ethyl or propyl, or phenyl,

R^2 stands for hydrogen or methyl,

15 n signifies 0, 1 or 2 and

p signifies an integer from 1 to 6.

20 Acryloyloxysilanes or methacryloyloxysilanes are particularly suitable for forming the aforementioned structure (V-1), and have a high grafting efficiency. As a result, an effective formation of the graft chains is guaranteed, and consequently the impact strength of the resulting resin composition is favoured.

The following may be named in exemplary and preferred manner: β -methacryloyloxy-
25 ethyldimethoxymethyl silane, γ -methacryloyloxy-propylmethoxydimethyl silane, γ -
methacryloyloxy-propyldimethoxymethyl silane, γ -methacryloyloxy-
propyltrimethoxymethyl silane, γ -methacryloyloxy-propylethoxydiethyl silane, γ -
methacryloyloxy-propyldiethoxymethyl silane, δ -methacryloyloxy-butyldiethoxymethyl
silane or mixtures thereof.

30

Preferably 0 wt.% to 20 wt.% grafting agent are employed, relative to the total weight of the silicone rubber.

The silicone rubber may be produced by emulsion polymerisation, as described in
5 US 2,891,920 and US 3,294,725, for example. In this case the silicone rubber is obtained
in the form of an aqueous latex. For this, a mixture containing organosiloxane, cross-
linking agent and optionally grafting agent is mixed, subject to shear, with water, for
example by means of a homogeniser, in the presence of an emulsifier based on sulfonic
acid, such as, for example, alkylbenzenesulfonic acid or alkylsulfonic acid, whereby the
10 mixture polymerises completely to form silicone-rubber latex. Particularly suitable is an
alkylbenzenesulfonic acid, since it acts not only as an emulsifier but also as a
polymerisation initiator. In this case a combination of the sulfonic acid with a metal salt
of an alkylbenzenesulfonic acid or with a metal salt of an alkylsulfonic acid is
favourable, because the polymer is stabilised by this means during the later graft
15 polymerisation.

After the polymerisation the reaction is terminated by the reaction mixture being
neutralised by addition of an aqueous alkaline solution, for example by addition of an
aqueous solution of sodium hydroxide, potassium hydroxide or sodium carbonate.

20

Also suitable as graft bases B.2 in accordance with the invention are silicone-acrylate
rubbers (B.2.2). These silicone-acrylate rubbers are composite rubbers with graft-active
sites containing 10 - 90 wt.% silicone-rubber portion and 90 wt.% to 10 wt.% polyalkyl-
(meth)acrylate-rubber portion, the two named rubber components permeating each other
25 in the composite rubber, so that they cannot be substantially separated from one another.

If the proportion of the silicone-rubber component in the composite rubber is too high,
the finished resin compositions have adverse surface properties and impaired
pigmentability. If, on the other hand, the proportion of the polyalkyl-(meth)acrylate-
30 rubber component in the composite rubber is too high, the impact strength of the finished
resin composition is adversely influenced.

Silicone-acrylate rubbers are known and are described, for example, in US 5,807,914, EP 430 134 and US 4,888,388.

- 5 Suitable silicone-rubber components of the silicone-acrylate rubbers according to B.2.2 are those as already described under B.2.1.

Suitable polyalkyl-(meth)acrylate-rubber components of the silicone-acrylate rubbers according to B.2.2 may be produced from alkyl methacrylates and/or alkyl acrylates, a
10 cross-linking agent (VI) and a grafting agent (VII). Exemplary and preferred alkyl methacrylates and/or alkyl acrylates in this connection are the C₁ to C₈ alkyl esters, for example methyl, ethyl, n-butyl, t-butyl, n-propyl, n-hexyl, n-octyl, n-lauryl and 2-ethylhexyl esters; halogen alkyl esters, preferentially halogen C₁-C₈-alkyl esters, such as chloroethyl acrylate, and also mixtures of these monomers. Particularly preferred is n-
15 butyl acrylate.

Monomers with more than one polymerisable double bond may be employed as cross-linking agents (VI) for the polyalkyl-(meth)acrylate-rubber component of the silicone-acrylate rubber. Preferred examples of cross-linking monomers are esters of unsaturated
20 monocarboxylic acids with 3 to 8 C atoms and of unsaturated monohydric alcohols with 3 to 12 C atoms, or of saturated polyols with 2 to 4 OH groups and 2 to 20 C atoms, such as ethylene glycol dimethacrylate, propylene glycol dimethacrylate, 1,3-butylene glycol dimethacrylate and 1,4-butylene glycol dimethacrylate. The cross-linking agents may be used on their own or in mixtures of at least two cross-linking agents.

25

Exemplary and preferred grafting agents (VII) are allyl methacrylate, triallyl cyanurate, triallyl isocyanurate or mixtures thereof. Allyl methacrylate may also be employed as cross-linking agent (VI). The grafting agents may be used on their own or in mixtures of at least two grafting agents.

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The quantity of cross-linking agent (VI) and grafting agent (VII) amounts to 0.1 wt.% to 20 wt.%, relative to the total weight of the polyalkyl-(meth)acrylate-rubber component of the silicone-acrylate rubber.

5 The silicone-acrylate rubber is produced by the silicone rubber according to B.2.1 firstly being produced in the form of aqueous latex. This latex is subsequently enriched with the alkyl methacrylates and/or alkyl acrylates, with the cross-linking agent (VI) and with the grafting agent (VII) to be used, and a polymerisation is carried out. Preferred is a radically initiated emulsion polymerisation, initiated for example by a peroxide initiator,
10 an azo initiator or a redox initiator. Particularly preferred is the use of a redox initiator system, especially a sulfoxylate initiator system produced by combination of iron sulfate, disodium ethylenediamine tetraacetate, rongalite and hydroperoxide.

The grafting agent (V) which is used in the production of the silicone rubber has the
15 result that the polyalkyl-(meth)acrylate-rubber portion is covalently bonded to the silicone-rubber portion. In the course of polymerisation, the two rubber components permeate each other and in this way form the composite rubber which after polymerisation can no longer be separated into its constituents consisting of silicone-rubber component and polyalkyl-(meth)acrylate-rubber component.

20

With a view to producing the silicone(-acrylate) graft rubbers B named as component B), the monomers B.1 are grafted onto the rubber base B.2.

In this connection the polymerisation methods that are described, for example, in
25 EP 249 964, EP 430 134 and US 4,888,388 may be employed.

For example, the graft polymerisation is undertaken in accordance with the following polymerisation method. In a single-stage or multi-stage radically initiated emulsion polymerisation the desired vinyl monomers B.1 are grafted onto the graft base which is
30 present in the form of aqueous latex. The grafting efficiency here should be as high as possible, and is preferably greater than or equal to 10 %. The grafting efficiency depends

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crucially on the grafting agent (V) or (VII) being used. After the polymerisation to form the silicone(-acrylate) graft rubber, the aqueous latex is passed into hot water in which metal salts, such as calcium chloride or magnesium sulfate, for example, have previously been dissolved. In the process the silicone(-acrylate) graft rubber coagulates and can
5 subsequently be separated.

The alkyl-methacrylate graft rubbers and alkyl-acrylate graft rubbers named as component B) are commercially available. The following may be named in exemplary manner: Metablen[®] SX 005 and Metablen[®] SRK 200 produced by Mitsubishi Rayon Co.
10 Ltd.

Component C

The term 'talc' is to be understood to mean a naturally occurring or synthetically produced
15 talc.

Pure talc has the chemical composition $3\text{MgO}_4 \cdot 4\text{SiO}_2 \cdot \text{H}_2\text{O}$ and consequently has an MgO content of 31.9 wt.%, an SiO_2 content of 63.4 wt.% and a content of chemically bound water of 4.8 wt.%; it is a silicate with a layered structure.
20

Naturally occurring talc materials generally do not have the ideal composition specified above, since they are contaminated as a result of partial replacement of the magnesium by other elements, by partial replacement of silicon, by, for example, aluminium, and/or as a result of intergrowths with other minerals such as, for example, dolomite, magnesite
25 and chlorite.

The special grades of talc in the sense according to the invention are distinguished by a particularly high purity, characterised by an MgO content from 28 wt.% to 35 wt.%, preferably 30 wt.% to 33 wt.%, particularly preferably 30.5 wt.% to 32 wt.%, and an
30 SiO_2 content from 55 wt.% to 65 wt.%, preferably 58 wt.% to 64 wt.%, particularly preferably 60 wt.% to 62.5 wt.%. Preferred types of talc are distinguished furthermore

by an Al₂O₃ content of less than 5 wt.%, particularly preferably less than 1 wt.%, in particular less than 0.7 wt.%.

A commercially available type of talc that conforms to this definition is, for example,
5 Luzenac[®] A3 produced by Luzenac Naintsch Mineralwerke GmbH (Graz, Austria).

Talc types not in the sense according to the invention are, for example, Luzenac SE-Standard, Luzenac SE-Super, Luzenac SE-Micro and also Luzenac ST 10, 15, 20, 30 and 60, all of which are marketed by Luzenac Naintsch Mineralwerke GmbH.

10

Advantageous, in particular, is the use of the talc according to the invention in the form of finely ground types with a mean particle size d₅₀ from 0.1 µm to 20 µm, preferably 0.2 µm to 10 µm, particularly preferably 1.1 µm to 5 µm, quite particularly preferably 1.15 µm to 2.5 µm.

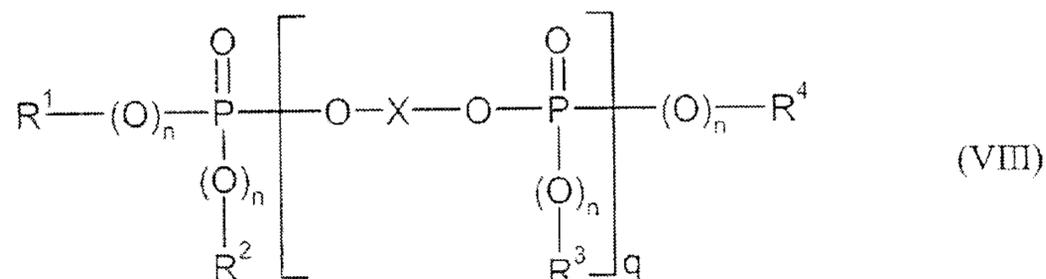
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The talc may have been surface-treated, for example silanised, in order to guarantee better compatibility with the polymer. With regard to the processing and production of the moulding compositions, the use of compacted talcs is also advantageous.

20 Component D

Phosphorus-containing flameproofing agents (D) in the sense according to the invention are preferably selected from the groups comprising the monomeric and oligomeric phosphoric and phosphonic esters, phosphonate amines and phosphazenes, in which case
25 mixtures of several components selected from one or various of these groups may also find application as flameproofing agents. Other halogen-free phosphorus compounds, not mentioned especially herein, may also be employed on their own or in arbitrary combination with other halogen-free phosphorus compounds.

Preferred monomeric and oligomeric phosphoric and phosphonic esters are phosphorus compounds of the general formula (VIII)



5 wherein

R^1 , R^2 , R^3 and R^4 signify in each instance, independently of one another, C_1 to C_8 alkyl, C_5 to C_6 cycloalkyl, C_6 to C_{20} aryl or C_7 to C_{12} aralkyl, which may be optionally substituted in each instance by alkyl, preferentially C_1 to C_4 alkyl, and/or halogen, preferentially chlorine, bromine,

10

n signify, independently of one another, 0 or 1,

q signifies 0 to 30 and

15

X signifies a single-ring or multi-ring aromatic residue with 6 to 30 C atoms, or a linear or branched aliphatic residue with 2 to 30 C atoms, which may have been OH-substituted and may include up to 8 ether linkages.

20 R^1 , R^2 , R^3 and R^4 preferably stand, independently of one another, for C_1 to C_4 alkyl, phenyl, naphthyl or phenyl- C_1 - C_4 -alkyl. The aromatic groups R^1 , R^2 , R^3 and R^4 may, in turn, have been substituted with halogen groups and/or alkyl groups, preferably chlorine, bromine and/or C_1 to C_4 alkyl. Particularly preferred aryl residues are cresyl, phenyl, xylenyl, propylphenyl or butylphenyl, as well as the corresponding brominated and

25 chlorinated derivatives thereof.

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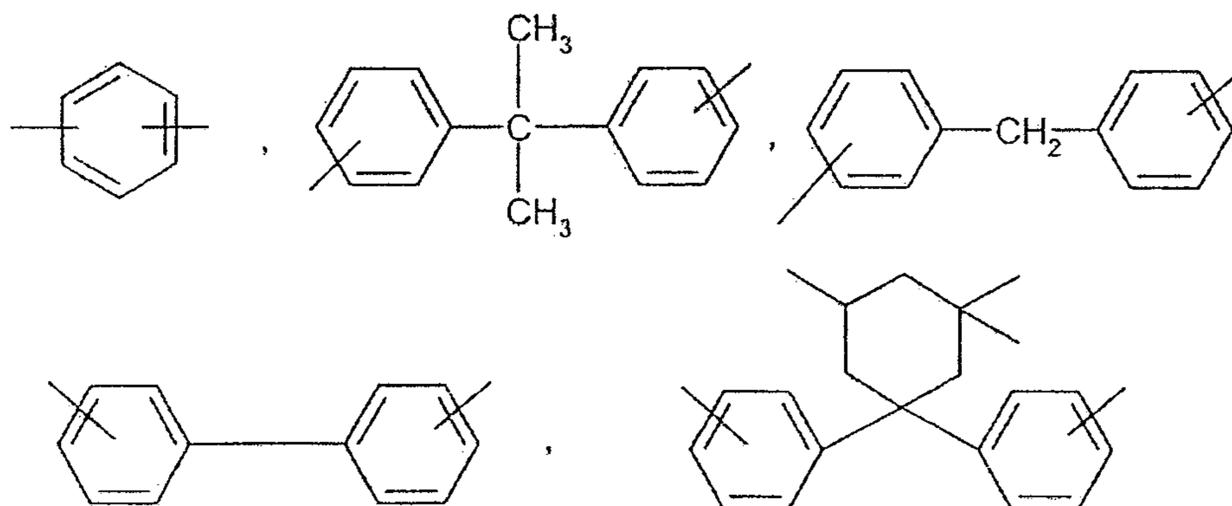
X in formula (VIII) preferably signifies a single-ring or multi-ring aromatic residue with 6 to 30 C atoms. Said residue is preferably derived from diphenols of the formula (I).

5 n in formula (VIII) may be, independently of one another, 0 or 1; n is preferentially equal to 1.

q stands for values from 0 to 30, preferably 0.3 to 20, particularly preferably 0.5 to 10, in particular 0.5 to 6, quite particularly preferably 1.1 to 1.6.

10

X stands in particularly preferred manner for



15 or the chlorinated or brominated derivatives thereof; in particular, X is derived from resorcinol, hydroquinone, bisphenol A or diphenylphenol. In particularly preferred manner X is derived from bisphenol A.

20 Mixtures of various phosphates may also be employed by way of component D according to the invention.

Phosphorus compounds of the formula (VIII) are, in particular, tributyl phosphate, triphenyl phosphate, tricresyl phosphate, diphenylcresyl phosphate, diphenyloctyl

phosphate, diphenyl-2-ethylcresyl phosphate, tri(isopropylphenyl)phosphate, resorcinol-bridged diphosphate and bisphenol-A-bridged diphosphate. The use of oligomeric phosphoric esters of the formula (VIII) that are derived from bisphenol A is particularly preferred.

5

The phosphorus compounds according to component D are known (cf., for example, EP-A 0 363 608, EP-A 0 640 655) or may be produced in analogous manner by known methods (e.g. *Ullmanns Enzyklopädie der technischen Chemie*, Vol. 18, p 301 ff. 1979; Houben-Weyl, *Methoden der organischen Chemie*, Vol. 12/1, p 43; *Beilstein*, Vol. 6, p 177).

10

The mean q-values can be determined by the composition of the phosphate mixture (molecular-weight distribution) being determined by means of suitable methods (gas chromatography (GC), high-pressure liquid chromatography (HPLC), gel permeation chromatography (GPC)), and by the mean values of q being calculated therefrom.

15

Furthermore, phosphonate amines and phosphazenes, such as are described in WO 00/00541 and WO 01/18105, may be employed as flameproofing agents.

20 The flameproofing agents may be employed on their own or in an arbitrary mixture with one another or in a mixture with other flameproofing agents.

Component E

25 Component E is constituted by inorganic boron compounds with elements of the 6th Main Group of the Periodic Table, preferably with oxygen. Preferred oxygen-containing boron compounds are metal salts of the borates, in which case the oxygen-containing boron compound may be present as orthoborate, metaborate, hydroxoborate or polyborate. Metals of the 1st to 5th Main Groups or of the 1st to 8th Subgroups of the
30 Periodic Table, preferably metals of the 1st and 2nd Main Groups or of the 1st and 2nd Subgroups of the Periodic Table, act as counterions of the borates; for example and

preferably, compounds such as $\text{Li}_3[\text{BO}_3]$, $\text{Li}[\text{BO}_2]$, $\text{Li}[\text{B}(\text{OH})_4]$, $\text{Na}_3[\text{B}_3\text{O}_6]$, $\text{Na}_2\text{B}_4\text{O}_7 \cdot 4\text{H}_2\text{O}$, $\text{Na}_2\text{B}_4\text{O}_7 \cdot 10\text{H}_2\text{O}$, $\text{NaCaB}_5\text{O}_9 \cdot 6\text{H}_2\text{O}$, $\text{K}_3[\text{B}_3\text{O}_6]$, $\text{KB}_5\text{O}_8 \cdot 4\text{H}_2\text{O}$, $\text{Mg}_3[\text{BO}_3]_2$, $\text{Ca}[\text{BO}_3]_2$, $\text{Ca}[\text{BO}_2]_2$, $\text{CaB}_4\text{O}_7 \cdot 4\text{H}_2\text{O}$, $\text{Ca}_2\text{B}_6\text{O}_{11} \cdot 5\text{H}_2\text{O}$, $\text{Ca}_2\text{B}_6\text{O}_{11} \cdot 7\text{H}_2\text{O}$, $\text{Ca}_4\text{B}_{10}\text{O}_{19} \cdot 7\text{H}_2\text{O}$, $\text{Ca}_5\text{B}_{12}\text{O}_{23} \cdot 9\text{H}_2\text{O}$, $\text{Sr}[\text{BO}_2]_2$, $\text{Ba}_3[\text{B}_3\text{O}_6]_2$, $\text{Cu}_3[\text{BO}_3]_2$,
 5 $\text{Na}_2\text{B}_4\text{O}_7 \cdot 5\text{H}_2\text{O}$, $\text{Na}_2\text{B}_8\text{O}_{13} \cdot 4\text{H}_2\text{O}$, $\text{BaB}_2\text{O}_4 \cdot \text{H}_2\text{O}$, $\text{ZnB}_2\text{O}_4 \cdot 2\text{H}_2\text{O}$, $\text{Zn}_2\text{B}_4\text{O}_8 \cdot 3\text{H}_2\text{O}$, $\text{Zn}_2\text{B}_6\text{O}_{11} \cdot 7\text{H}_2\text{O}$, $\text{Zn}_2\text{B}_6\text{O}_{11} \cdot 9\text{H}_2\text{O}$, $\text{Zn}_3\text{B}_4\text{O}_9 \cdot 5\text{H}_2\text{O}$, $\text{Zn}[\text{B}_3\text{O}_3(\text{OH})_5] \cdot \text{H}_2\text{O}$, $\text{Zn}_3[\text{BO}_3]_2$, $\text{Zn}_2\text{B}_6\text{O}_{11}$, $\text{Zn}_4\text{B}_2\text{O}_7 \cdot \text{H}_2\text{O}$, $\text{Zn}_2\text{B}_6\text{O}_{11} \cdot 3.5\text{H}_2\text{O}$ and $\text{ZnB}_4\text{O}_7 \cdot 4\text{H}_2\text{O}$. The inorganic boron compounds may be employed either on their own or in mixtures.

10 Particularly preferred are those inorganic boron compounds which at temperatures from 200 °C and 1000 °C, in particular from 300 °C to 600 °C, eliminate water that is bound in the form of water of crystallisation. Quite particularly preferred is zinc borate hydrate (e.g. $\text{Zn}_4\text{B}_2\text{O}_7 \cdot \text{H}_2\text{O}$, $\text{Zn}_2\text{B}_6\text{O}_{11} \cdot 3.5\text{H}_2\text{O}$ and $\text{ZnB}_4\text{O}_7 \cdot 4\text{H}_2\text{O}$), in particular $\text{Zn}_2\text{B}_6\text{O}_{11} \cdot 3.5\text{H}_2\text{O}$.

15

The mean particle diameter of the inorganic boron compounds amounts to 1 nm to 20 μm , preferably 0.1 μm to 15 μm , and particularly preferably 0.5 μm - 12 μm .

The terms 'particle size' and 'particle diameter' always signify the mean particle diameter
 20 d_{50} , ascertained by ultracentrifuge measurements according to W. Scholtan et al., *Kolloid-Z. und Z. Polymere* 250 (1972), pp 782 to 796.

Anti-dripping agents F

25 The compositions according to the invention may preferentially include fluorinated polyolefins F as anti-dripping agents. Fluorinated polyolefins are generally known (cf., for example, EP-A 640 655). A commercial product is, for example, Teflon[®] 30 N produced by DuPont.

30 The fluorinated polyolefins may also be employed in the form of a coagulated mixture of emulsions of the fluorinated polyolefins with emulsions of the graft polymers B) or with

an emulsion of a copolymer G.1) preferentially based on styrene/acrylonitrile, the fluorinated polyolefin being mixed as emulsion with an emulsion of the graft polymer or copolymer and being subsequently coagulated.

- 5 Furthermore, the fluorinated polyolefins may be employed as a precompound with the graft polymer B) or with a copolymer G.1) preferentially based on styrene/acrylonitrile. The fluorinated polyolefins are mixed as powder with a powder or granulate of the graft polymer or copolymer and are compounded in the melt generally at temperatures from 200 °C to 330 °C in conventional units such as internal mixers, extruders or double-shaft
10 screws.

- The fluorinated polyolefins may also be employed in the form of a master batch that is produced by emulsion polymerisation of at least one monoethylenically unsaturated monomer in the presence of an aqueous dispersion of the fluorinated polyolefin.
15 Preferred monomer components are styrene, acrylonitrile and mixtures thereof. The polymer is employed as a free-flowing powder after acidic precipitation and subsequent drying.

- The coagulates, precompounds or master batches ordinarily have solids contents of
20 fluorinated polyolefin from 5 wt.% to 95 wt.%, preferentially 7 wt.% to 60 wt.%.

Component G

- The composition according to the invention may include as further component G one or
25 more thermoplastic vinyl (co)polymers G.1 and/or polyalkylene terephthalates G.2 in quantities of 0 - 1.5 parts by weight, preferably 0 - 1 parts by weight. In particularly preferred manner the composition is free from thermoplastic vinyl (co)polymers G.1 and polyalkylene terephthalates G.2.

- 30 Suitable as vinyl (co)polymers G.1 are polymers of at least one monomer from the group comprising the vinyl aromatics, vinyl cyanides (unsaturated nitriles), (C₁-C₈)-alkyl

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(meth)acrylates, unsaturated carboxylic acids and also derivatives (such as anhydrides and imides) of unsaturated carboxylic acids. Particularly suitable are (co)polymers formed from

5 G.1.1 50 parts by weight to 99 parts by weight, preferentially 60 parts by weight to 80 parts by weight, of vinyl aromatics and/or ring-substituted vinyl aromatics such as styrene, α -methylstyrene, p-methylstyrene, p-chlorostyrene and/or (C₁-C₈)-alkyl methacrylates such as methyl methacrylate, ethyl methacrylate, and

10

G.1.2 1 part by weight to 50 parts by weight, preferentially 20 parts by weight to 40 parts by weight, of vinyl cyanides (unsaturated nitriles) such as acrylonitrile and methacrylonitrile and/or (C₁-C₈)-alkyl (meth)acrylates, such as 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-phenyl maleimide.

15

The vinyl (co)polymers G.1 are resinous, thermoplastic and rubber-free. In particularly preferred manner the copolymer is formed from G.1.1 styrene and G.1.2 acrylonitrile.

20

The (co)polymers according to G.1 are known and may be produced by radical polymerisation, in particular by emulsion polymerisation, suspension polymerisation, solution polymerisation or bulk polymerisation. The (co)polymers preferentially have average molecular weights Mw (weight-average, ascertained by light scattering or sedimentation) between 15,000 and 200,000.

25

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

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Preferred polyalkylene terephthalates include at least 80 wt.%, preferentially at least 90 wt.%, relative to the dicarboxylic-acid component, terephthalic-acid residues and at least 80 wt.%, preferentially at least 90 wt.%, relative to the diol component, ethylene-
5 glycol residues and/or butane-1,4-diol residues.

The preferred polyalkylene terephthalates may include, in addition to terephthalic-acid residues, up to 20 mol.%, preferentially up to 10 mol.%, residues of other aromatic or cycloaliphatic dicarboxylic acids with 8 to 14 C atoms, or of aliphatic dicarboxylic acids
10 with 4 to 12 C atoms, such as, for example, residues of phthalic acid, isophthalic acid, naphthalene-2,6-dicarboxylic acid, 4,4'-diphenyldicarboxylic acid, succinic acid, adipic acid, sebacic acid, azelaic acid, cyclohexanediacetic acid.

The preferred polyalkylene terephthalates may include, in addition to ethylene-glycol
15 residues or butane-1,4-diol residues, up to 20 mol.%, preferentially up to 10 mol.%, other aliphatic diols with 3 to 12 C atoms or cycloaliphatic diols with 6 to 21 C atoms, for example residues of propane-1,3-diol, 2-ethylpropane-1,3-diol, neopentyl glycol, pentane-1,5-diol, hexane-1,6-diol, cyclohexanedimethanol-1,4, 3-ethylpentane-2,4-diol, 2-methylpentane-2,4-diol, 2,2,4-trimethylpentane-1,3-diol, 2-ethylhexane-1,3-diol, 2,2-
20 diethylpropane-1,3-diol, hexane-2,5-diol, 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-A-2 407 674, DE-A-2 407 776, DE-A-2 715 932).

25 The polyalkylene terephthalates may be branched by incorporation of relatively small quantities of trihydric or tetrahydric alcohols or of tribasic or tetrabasic carboxylic acids, for example according to DE-A 1 900 270 and US-PS 3,692,744. Examples of preferred branching agents are trimesic acid, trimellitic acid, trimethylolethane, trimethylolpropane and pentaerythritol.

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Particularly preferred are polyalkylene terephthalates that have been produced solely from terephthalic acid and the reactive derivatives thereof (e.g. the dialkyl esters thereof) and ethylene glycol and/or butane-1,4-diol, and mixtures of these polyalkylene terephthalates.

5

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

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

The polyalkylene terephthalates may be produced by known methods (see, for example,
15 *Kunststoff-Handbuch*, Volume VIII, p 695 ff., Carl-Hanser-Verlag, Munich 1973).

Further added substances H

The moulding compositions according to the invention may contain at least one other of
20 the conventional additives, such as, for example, lubricants, mould-release agents, nucleating agents, antistatic agents, stabilisers, dyestuffs and pigments and also fillers and reinforcing materials different from talc and from component E.

Component H also comprises extremely fine-particle inorganic compounds that are
25 distinguished by an average particle diameter less than or equal to 200 nm, preferably less than or equal to 150 nm, in particular 1 nm to 100 nm.

Suitable extremely fine-particle inorganic compounds preferentially consist of at least one polar compound of one or more metals of the 1st to 5th Main Groups or 1st to 8th
30 Subgroups of the Periodic Table, preferably of the 2nd to 5th Main Groups or 4th to 8th Subgroups, particularly preferably of the 3rd to 5th Main Groups or 4th to 8th

Subgroups, or of compounds of these metals with at least one element selected from oxygen, hydrogen, sulfur, phosphorus, boron, carbon, nitrogen or silicon. Preferred compounds are, for example, oxides, hydroxides, hydrous oxides, sulfates, sulfites, sulfides, carbonates, carbides, nitrates, nitrites, nitrides, borates, silicates, phosphates, 5 hydrides, phosphites or phosphonates.

The extremely fine-particle inorganic compounds preferably consist of oxides, phosphates, hydroxides, preferentially of TiO_2 , SiO_2 , SnO_2 , ZnO , ZnS , boehmite, ZrO_2 , Al_2O_3 , aluminium phosphates, iron oxides, furthermore TiN , WC , $\text{AlO}(\text{OH})$, Fe_2O_3 iron 10 oxides, Na_2SO_4 , vanadium oxides, zinc borate, silicates such as Al silicate, Mg silicate, one-dimensional, two-dimensional, three-dimensional silicates. Mixtures and doped compounds may also be used. Furthermore, these extremely fine-particle inorganic compounds may be surface-modified with organic molecules, in order to achieve better compatibility with the polymers. It is possible for hydrophobic or hydrophilic surfaces to 15 be generated in this manner.

Particularly preferred are hydrated aluminium oxides (e.g. boehmite) or TiO_2 .

The terms 'particle size' and 'particle diameter' always signify the mean particle diameter 20 d_{50} , ascertained by ultracentrifuge measurements according to W. Scholtan et al., *Kolloid-Z. und Z. Polymere* 250 (1972), pp 782-796).

The inorganic compounds may be present in the form of powders, pastes, sols, dispersions or suspensions. Powders can be obtained from dispersions, sols or 25 suspensions by precipitation.

The extremely fine-particle inorganic compounds may be worked into the thermoplastic moulding compositions by conventional processes, for example by direct kneading or extruding of moulding compositions and the extremely fine-particle inorganic 30 compounds. Preferred processes are constituted by the production of a master batch, for example in flameproofing additives, and of at least one component of the moulding

compositions according to the invention in monomers or solvents, or the co-precipitation of a thermoplastic component and the extremely fine-particle inorganic compounds, for example by co-precipitation of an aqueous emulsion and the extremely fine-particle inorganic compounds, optionally in the form of dispersions, suspensions, pastes or sols
5 of the extremely fine-particle inorganic materials.

The compositions are produced by the respective constituents being mixed in known manner and being melt-compounded and melt-extruded at temperatures from 200 °C to 300 °C in conventional units such as internal mixers, extruders and double-shaft screws.

10

Mixing of the individual constituents may be undertaken in known manner both successively and simultaneously, to be specific both at about 20 °C (room temperature) and at higher temperature.

15 The thermoplastic moulding compositions are suitable, by reason of their excellent flame resistance and their high thermostability, for producing moulded articles of any type. By reason of the thermostability and rheological properties, processing temperatures of over 240 °C are preferred.

20 The invention also provides processes for producing the moulding compositions, and the use of the moulding compositions for producing moulded articles.

The moulding compositions may be processed into moulded articles by injection moulding, or the moulding compositions may preferentially be extruded into sheets or
25 films, particularly preferably into sheets.

The invention further provides the production of moulded articles from previously produced sheets or films by thermoforming.

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Thermoforming processes are described, for example, by G. Burkhardt et al. ("Plastics, Processing", in *Ullmann's Encyclopedia of Industrial Chemistry*, Wiley-VCH Verlag GmbH & Co. KGaA, 2002), or in the *Römpp Lexikon Chemie*, Georg Thieme Verlag Stuttgart, 1999. Thermoforming processes generally describe procedures in which
5 semifinished plastic products are heated and are shaped into three-dimensional objects under the influence of external forces (heat, pressure or vacuum).

Whereas, in the case of drawing (hot forming), a preheated plastic sheet is inserted between the two parts of the tool, the positive and the negative, and these parts are then
10 pressed together, as a result of which the plastic part is given its shape, draw forming operates with spring-loaded hold-down devices. The procedure without a negative tool is designated as deep drawing; deformation by means of a vacuum (vacuum forming) is also possible.

15 The extruded planar moulded articles described herein can be processed, for example, in the deep drawing process at surface temperatures from 150 °C to 220 °C, in particularly preferred manner at surface temperatures from 160 °C to 215 °C.

The invention consequently also provides a process for producing the thermoformed
20 moulded articles according to the invention, wherein

- (i) in a first step the components of the polycarbonate composition are melted and mixed,
- 25 (ii) in a second step the resulting melt is cooled and granulated,
- (iii) in a third step the granulate is melted and is extruded into sheets, and
- (iv) in a fourth step the sheets are shaped into a three-dimensional object, preferentially
30 by means of hot forming, draw forming, deep drawing or vacuum forming under the influence of external forces, for example by means of a one-part or two-part

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tool and/or by means of vacuum, the three-dimensional object preferentially being shaped in the deep drawing process and preferentially at a surface temperature of the sheet of 150 °C to 220 °C, particularly preferably at surface temperatures from 160 °C to 215 °C.

5

The moulded articles are suitable for the following applications: vehicle parts or interior finishing parts for motor vehicles, buses, lorries, motor caravans, rail vehicles, aircraft, watercraft or other vehicles, cover plates for the construction industry, planar wall elements, partition walls, wall-protection and edge-protection strips, profiles for electrical-installation channels, cable conductors, conductor-rail covers, window and door profiles, furniture parts and traffic signs. The moulded articles are particularly suitable for the following applications: vehicle parts or interior finishing parts for cars, buses, lorries, motor caravans, rail vehicles and aircraft.

15 The moulded articles are suitable in particularly preferred manner for producing covers, ceiling linings and side panellings, luggage flaps and similar interior panellings for rail vehicles and aircraft.

The following Examples serve for further elucidation of the invention.

Examples**Component A1**

- 5 Branched polycarbonate based on bisphenol A with a relative solution viscosity of $\eta_{\text{rel}} = 1.34$, measured in CH_2Cl_2 as solvent at 25 °C and at a concentration of 0.5 g/100 ml, which was branched by insertion of 0.3 mol.% isatin bicresol, relative to the sum of bisphenol A and isatin bicresol.

10 **Component B1**

Impact modifier, methyl-methacrylate-modified silicone-acrylate rubber, Metablen[®] SX 005 produced by Mitsubishi Rayon Co. Ltd., CAS 143106-82-5.

15 **Component B2**

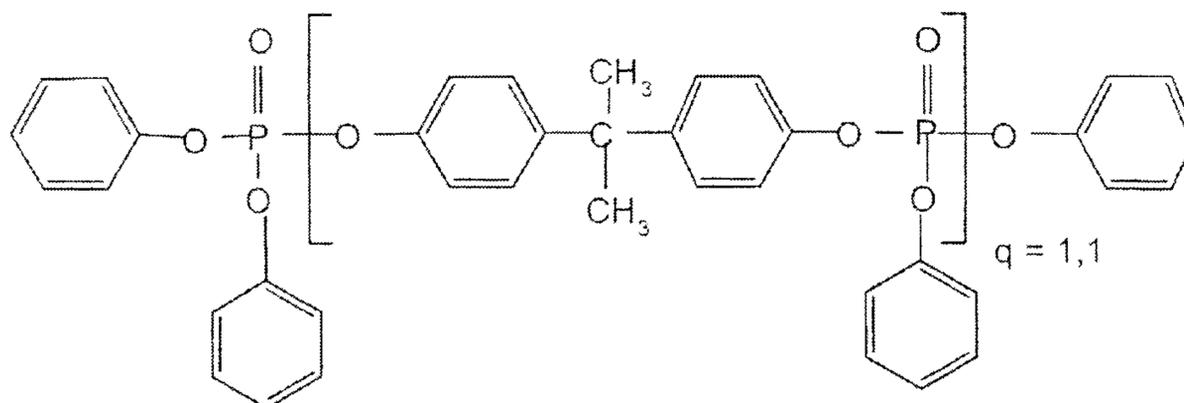
Impact modifier, styrene-acrylonitrile-modified silicone-acrylate rubber, Metablen[®] SRK 200 produced by Mitsubishi Rayon Co. Ltd., CAS 178462-89-0.

20 **Component C1**

Talc, Luzenac[®] A3C produced by Luzenac Naintsch Mineralwerke GmbH with an MgO content of 32 wt.%, an SiO_2 content of 61 wt.% and an Al_2O_3 content of 0.3 wt.%.

25 **Component D**

Bisphenol-A-based oligophosphate

**Component E**

- 5 Zinc borate hydrate ($Zn_2B_6O_{11} \cdot 3.5H_2O$, CAS No. 138265-88-0).

Component F

Polytetrafluoroethylene powder, CFP 6000 N, DuPont.

10

Component H

Mixture of 0.2 parts by weight of pentaerythritol tetrastearate as lubricant or mould-release agent and 0.1 parts by weight of phosphite stabiliser, Irganox[®] B 900, Ciba

- 15 Speciality Chemicals.

Production and testing of the moulding compositions

- 20 In a twin-screw extruder (ZSK-25) (Werner und Pfleiderer) the charging materials listed in Table 1 are compounded and granulated at a rotary speed of 225 rpm and with a throughput of 20 kg/h at a machine temperature of 260 °C.

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The finished granulates are processed in an injection-moulding machine into the appropriate test specimens (stock temperature 260 °C, tool temperature 80 °C, flow-front velocity 240 mm/s). Characterisation is undertaken in accordance with DIN EN ISO 180/1A (Izod notched impact strength), DIN EN ISO 527 (tensile modulus of elasticity), DIN ISO 306 (Vicat softening temperature, process B with 50 N loading and with a heating-rate of 120 K/h), ISO 11443 (melt viscosity), DIN EN ISO 1133 (melt volume-flow rate MVR) and UL 94 V.

In addition, sheets with a thickness of 3 mm were extruded (degassing extruder Breyer 60 without predrying of granulate, three-roll smoothing mill, two-roll take-off, radiometric thickness measurement) at a melt temperature of 270 °C in a sheet-and-film plant produced by Breyer, Singen.

The appropriate test-specimen geometries for ASTM E 162 and ASTM E 662 were cut out of the extruded sheets. Determination of the flame-spread index (Is) and of the dripping behaviour is undertaken in accordance with ASTM E 162 (with aluminium backing, $d = 3$ mm). Determination of the smoke density is undertaken in accordance with ASTM E 662 (with igniting flame, $d = 3$ mm).

The requirements placed on materials for American rail vehicles are laid down in the so-called Docket 90-A (Recommended Fire Safety Practices for Transit Bus and Van Materials Selection - published by the Department of Transportation, Federal Transit Administration, Federal Register, Vol. 58, No. 201). Accordingly, materials for interior panellings in ASTM E 162 must not display any burning dripping and must have a flame-spread index Is of less than 35; in addition, according to ASTM E 662 they must have a low smoke density ($Ds_{1.5 \text{ min}} < 100$ and $Ds_{4 \text{ min}} < 200$).

The suitability for thermoforming can be demonstrated by producing so-called deep drawn pyramids, the extruded sheets being deep drawn at 200 °C to a depth of 20 cm into a stepped pyramid with six elements. The surface quality of the deep drawn pyramids is assessed visually. The assessment "good" means that no edge

cracks and no stress whitenings occur at the corners. The assessment "poor" means that edge cracks and/or stress whitenings occur at the edges.

From Table I it is evident that only the compositions of Examples 3-6 - with the
5 combination of branched polycarbonate, silicone impact modifier, BDP, talc and zinc borate hydrate - achieve the object according to the invention and satisfy the requirements according to the American regulation for rail vehicles (Docket 90-A), i.e. according to ASTM E 162 have a flame-spread index I_s of less than 35, exhibit no burning dripping in the course of the test according to ASTM E 162, and satisfy the
10 requirements with respect to smoke density according to ASTM E 662 (D_s 1.5 min < 100 and D_s 4 min < 200). In addition, the tensile modulus of elasticity in Examples 3-6 according to the invention is clearly above 3500 N/mm^2 . The Comparative Examples V1 and V2, on the other hand, do not satisfy at least one of the aforementioned requirements.

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Table 1 Composition and Properties of the Moulding Compositions

Components (wt.%)	V1	V2	3	4	5	6
A1	74.5	77.6	72.6	70.6	72.6	70.5
B1	4.7	4.7	4.7	4.7	4.7	4.7
B2						
C1	10		10	10	10	10
D	10.1	13	10	10	10	10.1
E		4	2	4	2	4
F	0.4	0.4	0.4	0.4	0.4	0.4
H	0.3	0.3	0.3	0.3	0.3	0.3
Properties						
Izod notched impact strength / RT (DIN EN ISO 180/1A)	13.8	10.6	10.4	9.9	8.0	7.6
Tensile modulus of elasticity (DIN EN ISO 527)	3813	2664	3784	3871	4076	4082
Vicat B 120 (DIN ISO 306)	112	104	110	110	111	111
Melt viscosity (260 °C) [100 s ⁻¹] (ISO 11443)	1161	653	914	992	881	1001
Melt viscosity (260 °C) [1000 s ⁻¹] (ISO 11443)	406	291	349	377	345	372
Melt viscosity (260 °C) [1500 s ⁻¹] (ISO 11443)	315	236	274	293	274	289
MVR 260 °C / 5 kg (DIN EN ISO 1133)	7.5	18.8	9.8	9	11.0	9.3
UL 94 V (d = 1.5 mm); classification	V0	V0	V0	V0	V0	V0
UL 94 V (d = 1.5 mm); total burning time	10	19	10	8	5	7
Flame-spread index Is (ASTM E 162 (d = 3 mm))	11	28	6	7	0	5
Burning dripping? (ASTM E 162 (d = 3 mm))	yes	yes	no	no	no	no
Smoke density Ds after 1.5 min (ASTM E 662 (d = 3 mm))	n.d.	9	2	2	2	2
Smoke density Ds after 4 min (ASTM E 662 (d = 3 mm))	n.d.	143	68	69	100	79
Test acc. to Docket 90-A (d = 3 mm) passed?	no	no	yes	yes	yes	yes
Visual assessment of the deep drawpyramids	n.d.	n.d.	good	good	good	good

n.d. = not determined

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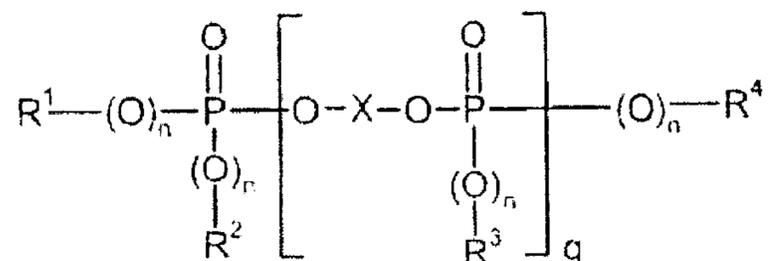
CLAIMS:

1. A composition comprising:
 - (A) 60 - 85 parts by weight of a branched aromatic polycarbonate, a branched aromatic polyester carbonate or a combination thereof;
- 5 (B) 1 - 25 parts by weight of a graft polymer comprising at least one graft base selected from the group consisting of a silicone rubber and a silicone-acrylate rubber;
 - (C) 9 - 18 parts by weight of talc;
 - (D) 0.4 - 20 parts by weight of a phosphorus-containing flameproofing agent;
 - (E) 1 - 6 parts by weight of at least one inorganic boron compound selected
- 10 from the group consisting of $Zn_4B_2O_7 \cdot H_2O$, $Zn_2B_6O_{11} \cdot 3.5H_2O$ and $ZnB_4O_7 \cdot 4H_2O$; and
 - (F) 0 - 3 parts by weight of an anti-dripping agent,wherein the sum of the parts by weight of components (A), (B), (C), (D), (E) and (F) are normalised to 100.
2. The composition according to claim 1, wherein the branched aromatic
- 15 polycarbonate or the branched aromatic polyester carbonate comprises at least one amine functionality.
3. The composition according to claim 1 or 2, wherein the graft polymer (B) is synthesised from:
 - (B.1) 5 wt.% to 95 wt.% of one or more vinyl monomers, on
- 20 (B.2) 95 wt.% to 5 wt.% of one or more graft bases selected from the group consisting of a silicone rubber (B.2.1) and a silicone-acrylate rubber (B.2.2), wherein the graft base has a glass transition temperature $< 10^\circ C$.

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4. The composition according to any one of claims 1 to 3, wherein the phosphorus-containing flameproofing agent (D) is of the general formula (VIII):



(VIII)

wherein:

- 5 R^1 , R^2 , R^3 and R^4 , independently of one another, represent: (i) optionally halogenated C_1 to C_8 alkyl, or (ii) C_5 to C_6 cycloalkyl, C_6 to C_{20} aryl or C_7 to C_{12} aralkyl, optionally substituted by alkyl and/or a halogen atom,
- n independently of one another, is 0 or 1;
- q is 0 to 30; and
- 10 X represents: (i) a single-ring or a multi-ring aromatic residue with 6 to 30 C atoms, or (ii) a linear or branched aliphatic residue with 2 to 30 C atoms, optionally OH-substituted and optionally containing up to 8 ether linkages.
5. The composition according to claim 4, wherein R^1 , R^2 , R^3 and R^4 , independently of one another, represent optionally halogenated C_1 to C_4 alkyl.
- 15 6. The composition according to claim 4 or 5, wherein the optional halogeno substituent is Cl or Br.
7. The composition according to any one of claims 4 to 6, wherein X in formula (VIII) is bisphenol A.
8. The composition according to any one of claims 1 to 7, comprising 4.7 parts by
20 weight to 6.6 parts by weight of component (B).

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9. The composition according to any one of claims 1 to 8, comprising 10 parts by weight to 12 parts by weight of component (C).
10. The composition according to any one of claims 1 to 9, wherein the talc of component (C) has a mean particle size (d_{50}) from 1.1 μm to 5 μm .
- 5 11. The composition according to any one of claims 1 to 10, comprising 2 parts by weight to 4 parts by weight of component (E).
12. The composition according to any one of claims 1 to 11, wherein component (E) is $\text{Zn}_2\text{B}_6\text{O}_{11} \cdot 3.5\text{H}_2\text{O}$.
13. The composition according to any one of claims 1 to 12, which is free from a
10 thermoplastic vinyl (co)polymer (G.1) and a polyalkylene terephthalate (G.2).
14. The composition according to any one of claims 1 to 13, further comprising a lubricant, a mould-release agent, a nucleating agent, an antistatic agent, a stabiliser, a dyestuff, a pigment, a filler or a reinforcing material different from talc.
15. The composition according to any one of claims 1 to 14, further comprising an
15 inorganic compound with an average particle diameter less than or equal to 200 nm.
16. Use of the composition according to any one of claims 1 to 15, for producing an injection-moulded or a thermoformed moulded article.