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(54) Titre : MATIERES DE MOULAGE IGNIFUGES A BASE DE POLYCARBONATE
(54) Title: FLAME-RESISTANT POLYCARBONATE MOULDING COMPOUNDS

(57) Abrégé/Abstract:
The invention relates to the fact that compositions containing polycarbonate of an average molecular weight, a special ABS graft polymer, in addition to at least one additional component selected from oligomeric organic phosphoric acid esters, fine-particled inorganic particles, polymers containing acrylate and fluorinated polyolefins, exhibit the required characteristics.

Flame-resistant polycarbonate moulding compositions

Abstract

It was found, that compositions containing polycarbonate of average molecular weight, a special ABS graft polymer produced by mass polymerisation processes and optionally at least one further component selected from oligomeric organic phosphoric acid esters, very fine inorganic particles, acrylate-containing polymers and fluorinated polyolefins, have the desired characteristics.

Flame-resistant polycarbonate moulding compositions

The present invention relates to polycarbonate compositions modified to be impact resistant, which are characterised by a combination of improved melt flowability, resistance to chemicals and creep resistance. The compositions also have good toughness, resistance to hydrolysis and processing stability. The invention preferably relates to flame-resistant compositions.

Manufacturers are now increasing the requirements for case materials for information technology equipment, such as for example printers, copiers, Notebooks, monitors, mobile phones etc. Many of these require flame-retarding properties. The demand for materials containing halogen-free flame-retardants is met particularly by PC+ABS blends, which have flame-resistance provided by organic phosphoric acid esters. The prior art relating to such compositions is extensive. EP-A 0 345 522, EP-A 0 640 655 and EP 0 363 608 are examples. Moulded bodies produced from such compositions have a good combination of application properties, such as good toughness, high resistance to thermoforming and satisfactory processing behaviour for many applications, but generally only within a limited processing window.

More recently, the requirements for resistance to chemicals and hydrolysis, and for injection moulding behaviour and processing stability i.e. the consistency of the characteristics of such moulding compositions at high processing temperatures, have increased. These requirements are met by PC/ABS compositions containing organic phosphoric acid esters, in particular oligomeric phosphates, as a flame retardant, which contain an ABS produced by the mass polymerisation process as the ABS component. Such compositions have also already been disclosed in patent literature.

Thus EP-A 0 755 977 discloses PC+ABS blends containing ABS of a low rubber content, preferably mass ABS, and a low flame-retardant content. The required melt flowability is achieved in these moulding compositions by using polycarbonate with a relatively low molecular weight, most preferably with a weight average molecular

weight of <25,000 g/mol, which restricts the stress cracking resistance. A combination of stress cracking resistance and flame-resistance that is sufficient for many, but not all, applications is obtained only by restricting the content of flame-retardant and rubber. The current increased requirements for melt flowability, flame-resistance and stress cracking resistance are no longer met by such compositions. Although the stress cracking resistance is improved by the use of higher-molecular polycarbonate, the melt flowability is reduced still further. Using higher quantities of flame retardant meets the flowability requirements but results in a reduction in resistance to chemicals.

EP-A 0 933 396 discloses polycarbonate mixtures containing mass ABS, that are equipped with oligophosphate flame retardants and are characterised by good notched impact resistance, resistance to hydrolysis and improved processing stability. However, the moulding compositions disclosed in this application have in particular inadequate flame-resistance and joint line strength for many applications.

JP-A 11 302 523 discloses compositions of polycarbonate, mass ABS, emulsion ABS and an acrylonitrile-vinyl copolymer, which contain monophosphate and polytetrafluoroethylene (PTFE) flame-retardants. These moulding compositions have an inadequate processing window, as they have a tendency to phase agglomeration at higher processing temperatures, which impairs the mechanical properties, and bleeds out the flame-retarding additive. Furthermore, experience shows that such compositions have inadequate resistance to hydrolysis.

DE 198 53 105 A1 also discloses compositions containing polycarbonate, mass ABS, which is preferably used in combination with emulsion ABS, and phosphoric acid esters. The moulding compositions according to the compositions disclosed in DE 198 53 105 A1 have, in particular, inadequate flowability, resistance to chemicals and to hydrolysis and poor creep behaviour.

EP-A 1 026 205 discloses moulding compositions of polycarbonate, ABS graft polymers, preferably produced by the mass polymerisation process, monomeric or

oligomeric phosphoric acid esters and silicate reinforcing materials, which are characterised in that the chlorine content of the composition is no greater than 100 ppm. Compositions containing acrylate-based graft polymers with a core/shell structure as a further component are also disclosed. The moulding compositions disclosed in this application are characterised by excellent resistance to hydrolysis, but generally have poor resistance to chemicals and inadequate creep behaviour as a result of the low molecular weight of the polycarbonate used. However, the low molecular weight is necessary to achieve sufficient melt flowability of the moulding composition in the compositions disclosed here. With the mass ABS used in this application it is impossible to achieve an adequate flowability level using higher-molecular polycarbonate. The same applies also to the moulding compositions disclosed in WO 00/39210, which are characterised in that they contain a bisphenol A-bridged oligophosphate with a low acid value as the flame-retarding additive.

US-A 5,849,827 discloses flame-resistant compositions of polycarbonate and emulsion ABS, which, in addition to conventional flame-retarding additives, contain very fine inorganic particles and are characterised by improved flame resistance. Positive effects of the particles on the melt flowability and resistance to hydrolysis of the moulding compositions are not disclosed in this application.

The object of the present invention was to provide polycarbonate compositions, which are characterised in particular by a combination of excellent stress cracking resistance and creep resistance with improved melt flowability and which also have a high degree of toughness, processing stability and resistance to hydrolysis. In particular, the object of the invention was to provide flame-resistant compositions from which moulded bodies can be produced, which pass the UL94V test at 1.5 mm wall thickness with the score V-0 and in which the flame-retarding additive does not bleed out significantly under the conventional processing conditions.

It was found that compositions containing polycarbonate of average molecular weight, a special ABS graft polymer produced by mass polymerisation processes, and optionally at least one other component selected from oligomeric organic

phosphoric acid esters, very fine inorganic particles, acrylate-containing polymers and fluorinated polyolefins, have the desired characteristics.

The present invention thus provides compositions containing

A) 40 to 95 parts by weight, preferably 50 to 80 parts by weight, in particular 60 to 80 parts by weight of aromatic polycarbonate or polyestercarbonate or mixtures of these with a weight average molecular weight $\overline{M}_w$ of 25,000 to 35,000, preferably of 26,000 to 33,000, in particular of 26,000 to 31,000 g/mol,

B) 5 to 40 parts by weight, preferably 7 to 30 parts by weight, in particular 10 to 20 parts by weight of a graft polymer produced by the mass polymerisation process from at least one acrylonitrile and/or methacrylonitrile – in the following (meth)acrylonitrile-, furthermore butadiene and at least one styrene monomer with a butadiene content of 8 to 15 wt.%, preferably 10 to 15 wt.%, in particular 11 to 14 wt.% in relation to the graft polymer and a (meth)acrylonitrile content of 15 to 30 wt.%, preferably 18 to 27 wt.%, in particular 21 to 26 wt.% in relation to the sum of (meth)acrylonitrile and styrene monomer in the graft polymer, this graft polymer containing styrene-(meth)acrylonitrile copolymer with a weight average molecular weight $\overline{M}_w$ of 50,000 to 140,000, preferably of 60,000 to 130,000, in particular of 60,000 to 110,000 g/mol,

C) 0 to 20 parts by weight, preferably 2 to 18 parts by weight, in particular 8 to 16 parts by weight of a halogen-free phosphorus compound, preferably an oligomeric organic phosphoric acid ester, in particular one based on bisphenol-A and its derivatives,

D) 0 to 1 parts by weight, preferably 0.1 to 0.5 parts by weight, in particular 0.2 to 0.5 parts by weight of a fluorinated polyolefin, which is preferably

incorporated in the form of a master batch, a pre-compound or a co-precipitate of PTFE and a polymer containing vinyl monomer,

E) 0 to 5 parts by weight, preferably 0.1 to 3 parts by weight, in particular 0.5 to 2 parts by weight of a polymer containing an acrylate monomer, preferably a graft polymer of 10-90 wt.%, preferably 30-80 wt.%, in particular 50-80 wt.% in relation to the graft polymer of a polymer in particle form with a glass transition temperature below 0°C, preferably below -20°C, in particular below -40°C as a grafting base and 90 to 10 wt.%, preferably 70 to 20 wt.%, in particular 50 to 20 wt.% in relation to the graft polymer of a grafting shell of vinyl monomers, containing at least 40 wt.%, in relation to the grafting shell, preferably at least 60 wt.%, in particular at least 80 wt.% of acrylate monomers, a polybutadiene or silicon or silicon-acrylate composite rubber being preferred in particular as the grafting base,

F) 0 to 3 parts by weight, preferably 0.1 to 2 parts by weight, in particular 0.2 to 1.5 parts by weight of inorganic material with an average maximum particle diameter of no more than 1000 nm, preferably no more than 500 nm, in particular no more than 200 nm and

G) up to 10 parts by weight, preferably up to 5 parts by weight, in particular up to 2 parts by weight of commercial polymer additives.

The sum of the parts by weight of all components is standardised to 100.

The components of the compositions according to the invention are explained in more detail below.

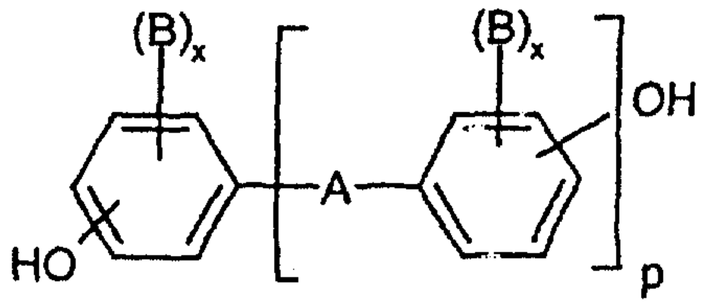
Component A

Suitable aromatic polycarbonates and/or aromatic polyestercarbonates according to Component A of the invention are known from the literature or can be produced by

processes known from the literature (for the production of aromatic polycarbonates see for example 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, DE-OS 3 832 396; for the production of aromatic polyestercarbonates e.g. DE-OS 3 077 934).

Aromatic polycarbonates are produced e.g. by the melt process or by reaction of diphenols with carbonic acid halides, preferably phosgene and/or with aromatic dicarboxylic acid dihalides, preferably benzene dicarboxylic acid dihalides, by the phase interface process, optionally using chain stoppers, for example monophenols and optionally using trifunctional or more than trifunctional branching agents, for example triphenols or tetraphenols.

Diphenols for the production of the aromatic polycarbonates and/or aromatic polyestercarbonates are preferably those of the formula (I)

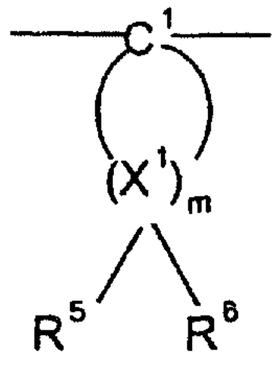


(I),

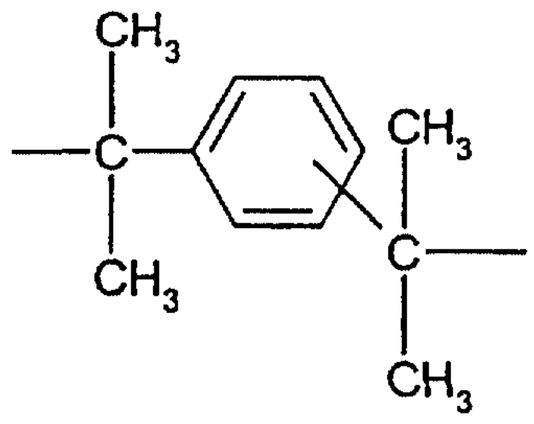
wherein

A is a single bond, C_1-C_5-alkylene, C_2-C_5-alkylidene, C_5-C_6-cycloalkylidene, -O-, -SO-, -CO-, -S-, -SO_2-, C_6-C_{12}-arylene, onto which further aromatic rings, optionally containing heteroatoms, may be condensed,

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



(II)



(III)

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

x in each case, independently of each other, is 0, 1 or 2,

p is 1 or 0, and

R^5 and R^6 can be selected individually for each X^1, independently of each other, as hydrogen or C_1-C_6-alkyl, preferably hydrogen, methyl or ethyl,

X^1 is carbon and

m is an integer from 4 to 7, preferably 4 or 5, provided that R^5 and R^6 are both alkyl on at least one X^1 atom.

Preferred diphenols are hydroquinone, resorcinol, dihydroxydiphenols, bis-(hydroxyphenyl)-C_1-C_5-alkanes, bis-(hydroxyphenyl)-C_5-C_6-cycloalkanes, bis-(hydroxyphenyl)-ethers, bis-(hydroxyphenyl)-sulfoxides, bis-(hydroxyphenyl)-ketones, bis-(hydroxyphenyl)-sulfones and α,α-bis-(hydroxyphenyl)-diisopropyl-benzenes and their ring-brominated and/or ring-chlorinated derivatives.

Particularly preferred diphenols are 4,4'-dihydroxydiphenyl, bisphenol A, 2,4-bis(4-hydroxyphenyl)-2-methylbutane, 1,1-bis-(4-hydroxyphenyl)-cyclohexane, 1,1-bis-(4-hydroxyphenyl)-3,3,5-trimethyl cyclohexane, 4,4'-dihydroxydiphenyl sulfide, 4,4'-dihydroxydiphenyl-sulfone and their di- and tetrabrominated or chlorinated derivatives 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.

2,2-bis-(4-hydroxyphenyl)-propane (bisphenol A) is preferred in particular.

The diphenols may be used individually, or in any mixture.

The diphenols are known from the literature or can be obtained by processes known from the literature.

Suitable chain stoppers for the production of the thermoplastic, aromatic polycarbonates are for example phenol, p-chlorophenol, p-tert.-butyl phenol or 2,4,6-tribromophenol, but also long chain alkyl phenols such as 4-(1,3-tetramethylbutyl)-phenol according to DE-OS 2 842 005 or monoalkylphenol or dialkyl phenols containing a total of 8 to 20 C atoms in the alkyl substituents such as 3,5-di-tert.-butyl phenol, p-iso-octyl phenol, p-tert. octyl phenol, p-dodecyl phenol and 2-(3,5-dimethylheptyl)-phenol and 4-(3,5-dimethylheptyl)-phenol. The quantity of chain stoppers to be used is generally 0.5 mol.% to 10 mol.%, in relation to the molar sum of the diphenols used in each case.

The thermoplastic, aromatic polycarbonates may be branched in the known way, and preferably by incorporating 0.05 to 2.0 mol.% in relation to the sum of the diphenols used, of trifunctional or more than trifunctional compounds, for example those having three or more phenolic groups.

Both homopolycarbonates and copolycarbonates are suitable. 1 to 25 wt.%, preferably 2.5 to 25 wt.% (in relation to the total quantity of diphenols to be used) of polydiorganosiloxanes with hydroxy-aryloxy terminal groups may also be used for the production of copolycarbonates according to Component A of the invention. These are known (see for example US-A 3 419 634) or can be produced by processes known from the literature. The production of copolycarbonates containing polydiorganosiloxanes is described e.g. in DE-A 3 334 782.

Preferred polycarbonates in addition to the bisphenol A homopolycarbonates are the copolycarbonates of bisphenol A containing up to 15 mol.% in relation to the molar sum of diphenols, of other diphenols mentioned as preferred or particularly preferred, in particular 2,2-bis(3,5-dibromo-4-hydroxyphenyl)-propane.

Aromatic dicarboxylic acid dihalides for the production of aromatic polyestercarbonates are preferably the diacid dichlorides of isophthalic acid, terephthalic acid, diphenylether-4,4'-dicarboxylic acid and naphthaline-2,6-dicarboxylic acid.

Mixtures of diacid dichlorides of isophthalic acid and terephthalic acid in a ratio of 1:20 to 20:1 are preferred in particular.

When producing polyestercarbonates, a carbonic acid halide, preferably phosgene, is also used as a bifunctional acid derivative.

In addition to the monophenols already mentioned, their chlorocarbonic acid esters and the acid chlorides of aromatic monocarboxylic acids, which may optionally be substituted by C_1-C_{22}-alkyl groups or by halogen atoms, as well as aliphatic C_2-C_{22} monocarboxylic acid chlorides are also possible chain stoppers for the production of the aromatic polyestercarbonates.

The quantity of chain stoppers is 0.1 to 10 mol.% in each case, in relation to mols of diphenols in the case of the phenolic chain stoppers, and to mols of dicarboxylic acid dichlorides in the case of the monocarboxylic acid chain stopper.

The aromatic polyestercarbonates may also have aromatic hydroxycarboxylic acids incorporated in them.

The aromatic polyestercarbonates may be either linear or branched in the known way (see also DE-A 2 940 024 and DE-A 3 007 934).

3- or polyfunctional carboxylic acid chlorides, such as trimesic acid trichloride, cyanuric acid trichloride, 3,3'-,4,4'-benzophenone-tetracarboxylic acid tetrachloride, 1,4,5,8-naphthaline tetracarboxylic acid tetrachloride or pyromellitic acid tetrachloride, in quantities of 0.01 to 1.0 mol.% (in relation to the dicarboxylic acid dichlorides used) or 3- or polyfunctional phenols, such as phloroglucinol, 4,6-dimethyl-2,4,6-tri-(4-hydroxyphenyl)-heptene-2,4,4-dimethyl-2,4-6-tri-(4-hydroxyphenyl)-heptane, 1,3,5-tri-(4-hydroxyphenyl)-benzene, 1,1,1-tri-(4-hydroxyphenyl)-ethane, tri-(4-hydroxyphenyl)-phenylmethane, 2,2-bis[4,4-bis(4-hydroxyphenyl)-cyclohexyl]-propane, 2,4-bis(4-hydroxyphenyl-isopropyl)-phenol, tetra-(4-hydroxyphenyl)-methane, 2,6-bis(2-hydroxy-5-methyl-benzyl)-4-methyl-phenol, 2-(4-hydroxyphenyl)-2-(2,4-dihydroxyphenyl)-propane, tetra-(4-[4-hydroxyphenyl-isopropyl]-phenoxy)-methane, 1,4-bis[4,4'-dihydroxytri-phenyl)-methyl]-benzene, can be used as branching agents in quantities of 0.01 to 1.0 mol.% in relation to the diphenols used. Phenolic branching agents may be added with the diphenols, acid chloride branching agents may be introduced together with the acid chlorides.

The proportion of carbonate structural units in the thermoplastic aromatic polyestercarbonates may be varied at will. The proportion of carbonate groups is preferably up to 100 mol.%, in particular up to 80 mol.%, most preferably up to 50 mol.% in relation to the sum of ester groups and carbonate groups. Both the ester

and the carbonate content of the aromatic polyestercarbonates may be present in the form of blocks or distributed at random in the polycondensate.

The thermoplastic, aromatic polycarbonates and polyestercarbonates may be used alone or in any mixture.

Component B

The ABS polymer according to Component B is a rubber-modified graft polymer in which styrene monomers according to B.1.1 and acrylonitrile and/or methacrylonitrile according to B.1.2 are polymerised in the presence of a butadiene rubber B.2, B being produced in the known way by a mass or mass suspension polymerisation process, as disclosed e.g. in US-A 3 243 481, US-A 3 509 237, US-A 3 660 535, US-A 4 221 833 and US-A 4 239 863.

Examples of B.1.1 styrene monomers are styrene, α-methylstyrene, halogen- or alkyl core-substituted styrenes such as p-methylstyrene, p-chlorostyrene. Monomers according to B.1.2 are acrylonitrile and/or methacrylonitrile.

Preferred B.1.1 monomers are styrene and α-methyl styrene, preferred B.1.2 monomer is acrylonitrile. Monomers preferred in particular are B.1.1 styrene and B.1.2 acrylonitrile.

B.2 rubbers suitable for the rubber-modified graft polymers B are butadiene rubbers. Butadiene rubbers according to the present invention are those based on butadiene, which may optionally contain up to 50 wt.%, preferably up to 30 wt.%, in particular up to 20 wt.% in relation to the rubber of copolymerisable monomers (e.g. according to B.1.1 and B.1.2), provided that the glass transition temperature of component B.2 is below 10°C, preferably below -10°C, in particular below -20°C. Pure polybutadiene rubber and butadiene/styrene copolymer rubber are preferred in particular.

The content of the grafting base (rubber) in component B is 8 to 15 wt.%, preferably 10 to 15 wt.%, in particular 11 to 14 wt.% in relation to B. If the rubber content is lower, an adequate level of toughness cannot be achieved and if it is higher, melt flowability is inadequate. The content of acrylonitrile and/or methacrylonitrile according to B.1.2 in B is 15 to 30 wt.%, preferably 18 to 27 wt.%, in particular 21 to 26 wt.% in relation to the sum of B.1.2 and styrene monomer B 1.1. in the graft polymer B.

Component B can additionally contain small quantities, normally less than 5 wt.%, preferably less than 2 wt.%, in relation to B.2, of ethylenically unsaturated monomers with a crosslinking action. Examples of such monomers with a crosslinking action are alkylene diol-di-(meth)-acrylate, polyester-di-(meth)-acrylate, divinyl benzene, trivinyl benzene, triallyl cyanurate, allyl-(meth)-acrylate, diallyl maleate and diallyl fumarate.

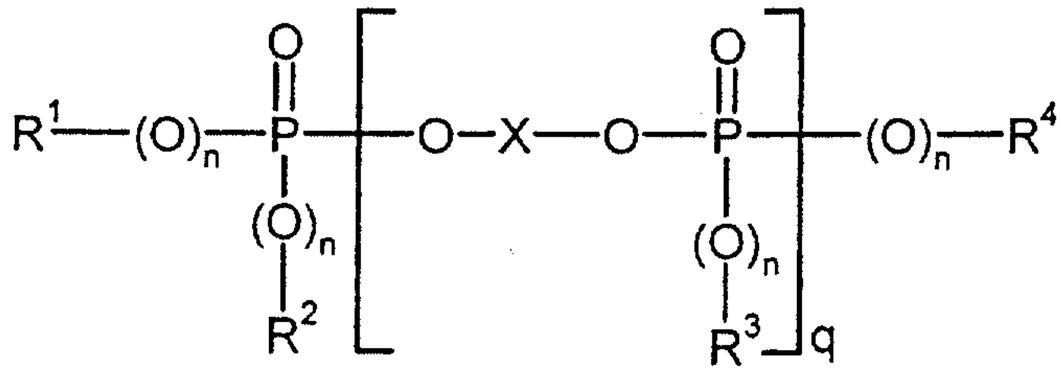
The random copolymer of styrene monomer B.1.1 and monomer B.1.2 is normally present in polymer B partially grafted onto or into rubber B.2, this graft mixed polymer forming discrete particles in the random copolymer of B.1.1 and B.1.2. The weight average molecular weight of the free, i.e. elutable random copolymer can be determined by gel permeation chromatography (GPC) and in Components B according to the invention, is $5 \cdot 10^4$ to $14 \cdot 10^4$, preferably $6 \cdot 10^4$ to $13 \cdot 10^4$, in particular $6 \cdot 10^4$ to $11 \cdot 10^4$ g/mol.

The average particle diameter of the resulting grafted rubber particles (determined by counting out from electron-microscopic recordings) is in the range 0.5 to 5 μm, preferably 0.8 to 2.5 μm.

Component C

Possible phosphorus compounds preferred as the halogen-free flame retardant are selected from the group of monomeric or oligomeric phosphoric- or phosphonic acid esters, phosphonate amines and phosphazenes.

Oligomeric phosphoric or phosphonic acid esters of the general formula (IV)



(IV)

are preferred, in which

R^1, R^2, R^3 and R^4, independently of each other are, in each case, C_1-C_8-alkyl, C_5-C_6-cycloalkyl, in each case optionally substituted by alkyl, preferably C_1 to C_4-alkyl, C_6 to C_{20}-aryl or C_7 to C_{12}-aralkyl,

n independently of each other means 0 or 1

q means 0.5 to 30 and

X means a mono- or polynuclear aromatic group having 6 to 30 C atoms, or a linear or branched aliphatic group having 2 to 30 C atoms, which may be OH substituted and may contain up to 8 ether bonds.

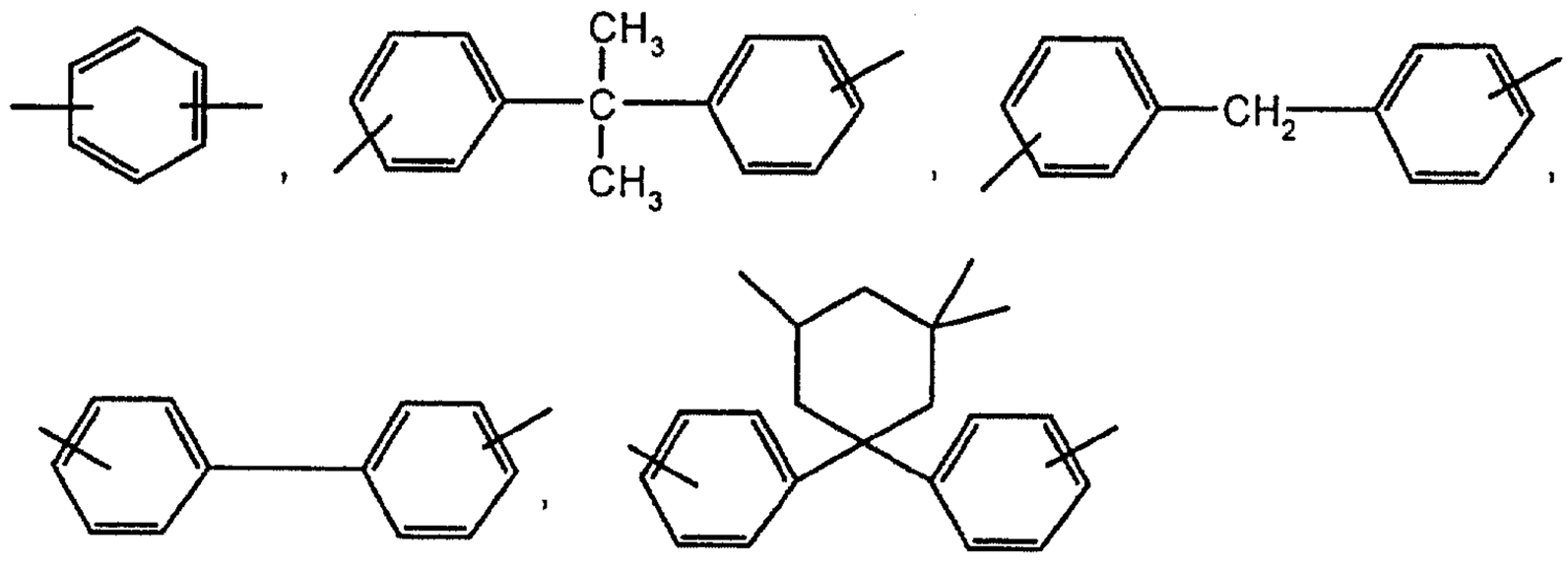
R^1, R^2, R^3 and R^4, independently of each other, preferably represent C_1 to C_4-alkyl, phenyl, naphthyl or phenyl-C_1-C_4-alkyl. The aromatic groups R^1, R^2, R^3 and R^4 may themselves be substituted with alkyl groups, preferably C_1 to C_4-alkyl. Particularly preferred aryl groups are cresyl, phenyl, xylenyl, propylphenyl or butylphenyl.

X in formula (IV) preferably means a mono- or polynuclear aromatic group having 6 to 30 C atoms. This is preferably derived from diphenols of formula (I).

n in formula (IV) may be, independently of each other, 0 or 1; n preferably equals 1.

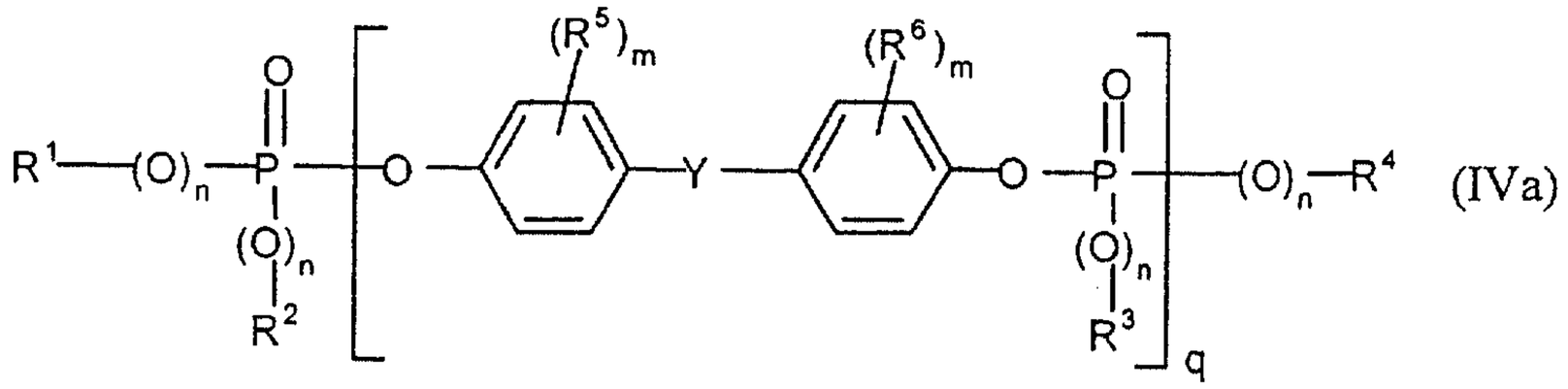
q represents values of 0.5 to 30, preferably 0.8 to 15, particularly preferably 1 to 5, in particular 0.8 to 2.

X preferably represents



in particular, X is derived from resorcinol, hydroquinone, bisphenol A or diphenylphenol. X is derived particularly preferably from bisphenol A.

Particularly preferred phosphatic compounds are compounds of formula (IVa)



(IVa)

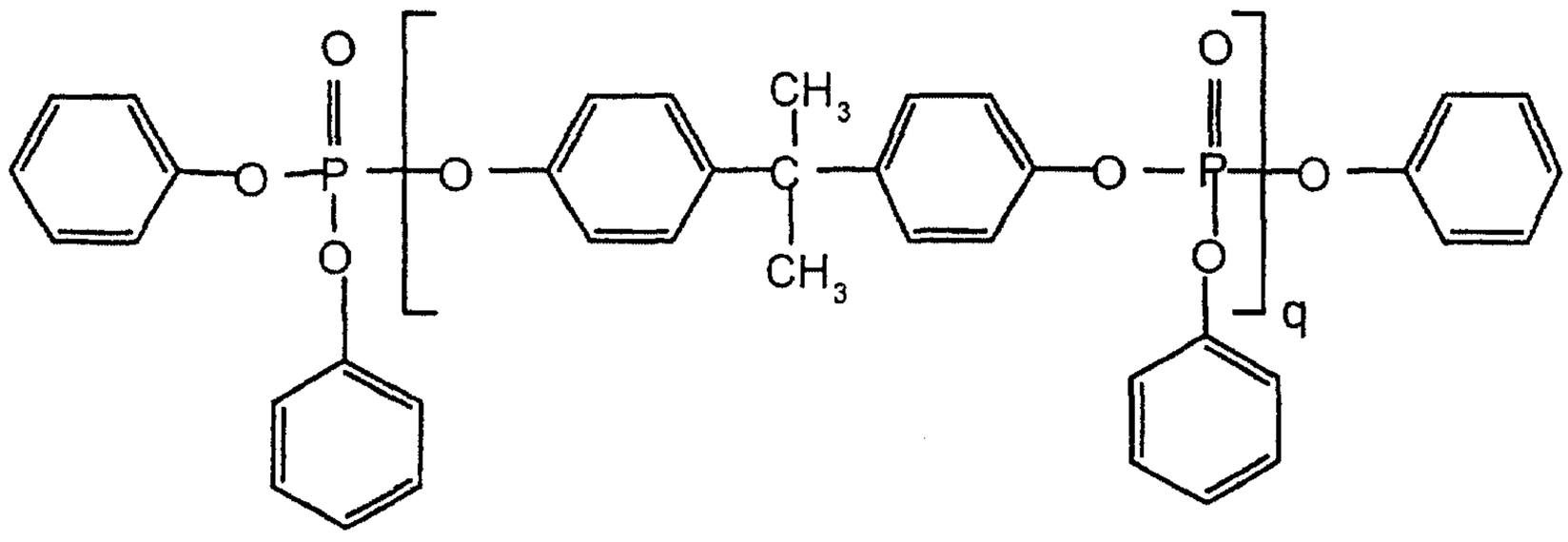
wherein

R^1, R^2, R^3, R^4, n and q have the meaning given in formula (IV),

m independently of each other means 0, 1, 2, 3 or 4

R^5 and R^6, independently of each other, mean C_1 to C_4-alkyl, preferably methyl or ethyl and

Y means C_1 to C_7-alkylidene, C_1 to C_7-alkylene, C_5 to C_{12}-cycloalkylene, C_5 to C_{12}-cycloalkylidene, -O-, -S-, $-SO_2-$ or –CO-, preferably isopropylidene or methylene.



where q = 0.8 to 1.5 is preferred in particular.

The phosphorus compounds according to Component C are known (cf. e.g. EP-A 0 363 608, EP-A 0 640 655) or can be produced in the same way 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).

The average q values can be determined by measuring the composition of the phosphate mixture (molecular weight distribution) by a suitable method (Gas Chromatography (GC), High Pressure Liquid Chromatography (HPLC), Gel Permeation Chromatography (GPC)) and calculating the mean values for q from these.

Component D

The flame retardants according to Component C are often used in combination with so-called anti-dripping agents, which reduce the tendency of the material to burning

drip-off in the event of a fire. Examples of these are compounds of the substance classes fluorinated polyolefins, silicons and aramide fibres. These may also be used in the compositions according to the invention. Fluorinated polyolefins are preferred as anti-dripping agents.

Fluorinated polyolefins are known and are disclosed for example in EP-A 0 640 655. They are marketed for example under the trademark Teflon® 30N by DuPont.

The fluorinated polyolefins can be used either in pure form or as a coagulated mixture of emulsions of fluorinated polyolefins with emulsions of vinyl monomer-based graft polymers or (co)polymers, the fluorinated polyolefin being mixed as an emulsion with an emulsion of the graft polymer or copolymer and then coagulated.

Furthermore, the fluorinated polyolefins can be used as a pre-compound with a graft polymer or copolymer, preferably vinyl monomer-based. The fluorinated polyolefins are mixed as a powder with a powder or granulate of the graft polymer or copolymer and compounded in the melt, generally at temperatures of 200 to 330°C in conventional units such as internal kneaders, extruders or twin shaft screws.

The fluorinated polyolefins can also be used in the form of a master batch, which is produced by emulsion polymerisation of at least one monoethylenically unsaturated monomer in the presence of an aqueous dispersion of the fluorinated polyolefin. Preferred monomer components are styrene, acrylonitrile, methylmethacrylate and mixtures thereof. After acid precipitation followed by drying, the polymer is used as a flowable powder.

The coagulates, pre-compounds and master batches generally have fluorinated polyolefin solids contents of 5 to 95 wt.%, preferably 7 to 80 wt.%.

The fluorinated polyolefins are used in concentrations of 0 to 1 parts by weight, preferably 0.1 to 0.5 parts by weight, in particular 0.2 to 0.5 parts by weight, these

quantities being in relation to the pure fluorinated polyolefin when using a coagulate, pre-compound or master batch.

Component E

All (co)polymers containing (meth)acrylic acid-C_1-C_8-alkylesters such as methylmethacrylate, n-butylacrylate, t-butylacrylate, which may optionally contain styrene monomers such as styrene, α-methylstyrene, halogen- or alkyl core substituted styrenes such as p-methylstyrene, p-chlorostyrene and rubbers, may be used as Component E. The proportion of (meth)acrylic acid-C_1-C_8-alkylesters in Component E is preferably greater than 10 wt.%, preferably greater than 20 wt.%.

The preferred acrylate compound in Component E is methylmethacrylate.

Graft polymers of 10-90 wt.%, preferably 30-80 wt.%, in particular 50-80 wt.% in relation to the graft polymer of a polymer in particle form with a glass transition temperature below 0°C, preferably below -20°C, in particular below -40°C as a grafting base and 90 to 10 wt.%, preferably 70 to 20 wt.%, in particular 50 to 20 wt.%, in relation to the graft polymer of a grafting shell of vinyl monomers, having a content of at least 40 wt.%, in relation to the grafting shell, preferably at least 60 wt.%, in particular at least 80 wt.% of (meth)acrylic acid-C_1-C_8-alkylesters such as methylmethacrylate, n-butylacrylate, t-butylacrylate, may be used in particular as Component E. Methylmethacrylate is particularly preferred as the (meth)acrylic acid-C_1-C_8-alkylester in the grafting shell.

Grafting bases suitable for these graft polymers are, for example, diene rubbers, EP(D)M rubbers, in other words, those based on ethylene/propylene and optionally diene compounds, also acrylate-, polyurethane-, silicon-, chloroprene- and ethylene/vinylacetate rubbers and silicon-acrylate composite rubbers.

Diene rubbers, silicon rubbers and silicon-acrylate composite rubbers are preferred. Silicon-acrylate composite rubbers are preferred in particular.

Diene rubbers according to the present invention are e.g. those based, for example, on butadiene or isoprene or copolymers of these with other copolymerisable monomers. Preferred copolymerisable monomers are vinyl monomers selected from the group of vinyl aromatics, core-substituted vinyl aromatics (such as for example styrene, α-methylstyrene, p-methylstyrene), methacrylic acid-(C_1-C_8)-alkylesters (such as methylmethacrylate, ethylmethacrylate), acrylic acid-(C_1-C_8)-alkylesters (such as n-butyl acrylate and tert.-butyl acrylate or mixtures of these), vinyl cyanides (unsaturated nitriles such as acrylonitrile and methacrylonitrile) and derivatives of unsaturated carboxylic acids such as anhydrides and imides (for example maleic acid anhydride and N-phenyl maleic imide), styrene at a preferred content of up to 30 wt.% in relation to the diene rubber being preferred in particular.

Pure polybutadiene rubber is preferred in particular.

These grafting bases generally have an average particle size (d_{50} value) of 0.05 to 5 µm, preferably 0.1 to 2 µm, in particular 0.1 to 1 µm.

The average particle size d_{50} is the diameter above and below which 50 wt.% of the particles in each case lie. It can be determined by ultracentrifugal measurement (W. Scholtan, H. Lange, Kolloid, Z. und Z. Polymere 250 (1972), 782-1796).

The gel content of these grafting bases is at least 30 wt.%, preferably at least 40 wt.% (measured in toluene).

The gel content is measured at 25°C in a suitable solvent (M. Hoffmann, H. Krömer, R. Kuhn, Polymeranalytik I and II, Georg Thieme-Verlag, Stuttgart 1977).

Acrylate rubbers, silicon rubbers or silicon-acrylate composite rubbers that contain 0 to 100 wt.%, preferably 1 to 99 wt.%, in particular 10 to 99 wt.%, particularly preferably 30 to 99 wt.% polyorganosiloxane component and 100 to 0 wt.%, preferably 99 to 1 wt.%, in particular 90 to 1 wt.%, particularly preferably 70 to

1 wt.% polyalkyl(meth)acrylate rubber component (the total quantity of the respective rubber components equalling 100 wt.%) are particularly preferred as grafting bases.

Such rubbers preferably have an average particle diameter of 0.01 to 0.6 µm.

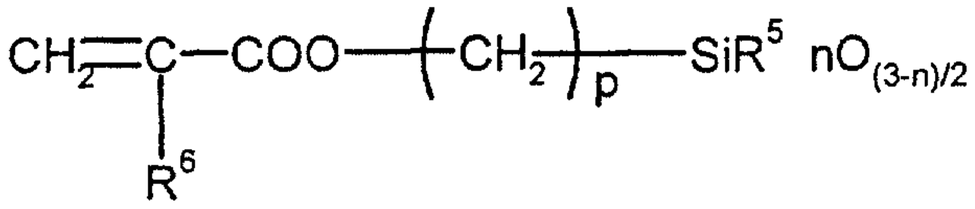
Preferred silicon-acrylate rubbers used are those for which the production process is disclosed in JP 08 259 791-A, JP 07 316 409-A and EP-A 0 315 035. The contents of these applications in this respect are hereby adopted in this application.

The polyorganosiloxane component in the silicon-acrylate composite rubber can be produced by reacting an organosiloxane with a multifunctional crosslinker in an emulsion polymerisation process. Furthermore, it is possible, by adding suitable unsaturated organosiloxanes, to insert graft-active sites into the rubber.

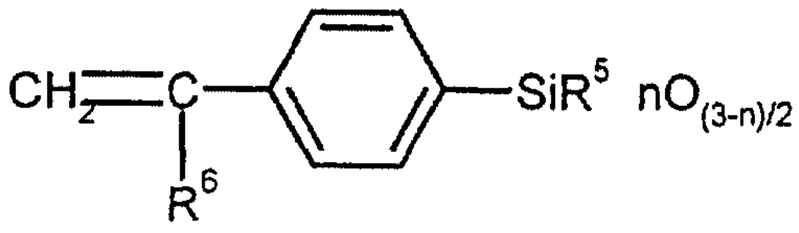
The organolsiloxane is generally cyclic, the ring structures preferably containing 3 to 6 Si atoms. Examples of these are hexamethylcyclotrisiloxane, octamethylcyclotetrasiloxane, decamethylcyclopentasiloxane, dodecamethylcyclohexasiloxane, trimethyltriphenylcyclotrisiloxane, tetramethyltetraphenylcyclotetrasiloxane, octaphenylcyclotetrasiloxane, which can be used alone or in a mixture of two or more compounds. The organosiloxane component should account for at least 50 wt.%, preferably at least 70 wt.%, in relation to the silicon content in the silicon-acrylate rubber, of the structure of the silicon content of the silicon acrylate-rubber.

3- or 4-functional silane compounds are generally used as crosslinkers. Particularly preferred examples of these are: trimethoxymethylsilane, triethoxyphenylsilane, tetramethoxysilane, tetraethoxysilane, tetra-n-propoxysilane, tetrabutoxysilane. 4-functional branching agents, in particular tetraethoxysilane. The quantity of branching agent is generally 0 to 30 wt.% (in relation to the polyorganosiloxane component of the silicon-acrylate rubber).

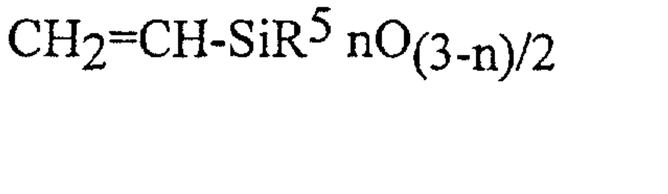
For the incorporation of graft-active sites in the polyorganosiloxane component of the silicon-acrylate rubber, compounds having one of the following structures are preferably used:



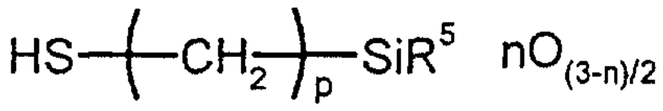
(GI-1)



(GI-2)



(GI-3)



(GI-4)

wherein

R^5 means methyl, ethyl, propyl or phenyl,

R^6 means hydrogen or methyl,

n means 0, 1 or 2 and

p means a number from 1 to 6.

(Meth)acryloyloxysilane is a preferred compound for the formation of the structure (GI 1). Preferred (meth)acryloyloxysilanes are, for example, β-methacryloyloxy-ethyl-dimethoxy-methyl-silane, γ-methacryloyl-oxy-propylmethoxy-dimethyl-

silane, γ-methacryloyloxypropyl-dimethoxy-methyl-silane, γ-methacryloyloxypropyl-trimethoxy-silane, γ-methacryloyl-propyl-ethoxy-diethyl-silane, γ-methacryloyloxypropyl-diethoxy-methyl-silane, γ-methacryloyloxy-butyl-diethoxy-methyl-silane.

Vinyl siloxanes, in particular tetramethyl-tetravinyl-cyclotetrasiloxane, are capable of forming the structure GI-2.

p-vinylphenyl-dimethoxy-methylsilane, for example, can form structure GI-3. γ-mercaptopropyldimethoxy-methylsilane, γ-mercaptopropylmethoxy-dimethylsilane, γ-mercaptopropyldiethoxymethylsilane etc can form the structure (GI-4).

The quantity of these compounds is 0 to 10, preferably 0.5 to 5 wt.% (in relation to the polyorganosiloxane component).

The acrylate component in the silicon-acrylate composite rubber may be produced from alkyl (meth)acrylates, crosslinkers and graft-active monomer units.

Examples of preferred alkyl (meth)acrylates are alkyl acrylates such as methylacrylate, ethylacrylate, n-propylacrylate, n-butylacrylate, 2-ethylhexyl acrylate and alkylmethacrylates such as hexyl methacrylate, 2-ethylhexyl methacrylate, n-lauryl methacrylate and, in particular, n-butylacrylate.

Multifunctional compounds are used as crosslinkers. Examples of these are: ethylene glycol dimethacrylate, propylene glycol dimethacrylate, 1,3-butylene glycol dimethacrylate and 1,4-butylene glycol dimethacrylate.

The following compounds are, for example, used, alone or in mixture, for the insertion of graft-active sites: allyl methacrylate, triallyl cyanurate, triallyl isocyanurate, allyl methacrylate. Allyl methacrylate can also function as a crosslinker. These compounds are used in quantities of 0.1 to 20 wt.%, in relation to the acrylate rubber component in the silicon-acrylate composite rubber.

Methods for producing the silicon-acrylate composite rubbers preferably used in the compositions according to the invention and for grafting these with monomers are disclosed, for example, in US-A 4 888 388, JP 08 259 791 A2, JP 07 316 409A and EP-A 0 315 035. Silicon-acrylate composite rubbers in which the silicon and acrylate components form a core-shell structure and those that form a network in which the acrylate and silicon components completely interpenetrate each other (interpenetrating network) are possible grafting bases for the graft polymer.

Graft polymerisation on the grafting bases described above can be carried out in suspension, dispersion or emulsion. Continuous or discontinuous emulsion polymerisation is preferred. This graft polymerisation is carried out with radical initiators (e.g. peroxides, azo compounds, hydroperoxides, persulfates, perphosphates) and optionally using anionic emulsifiers, e.g. carboxonium salts, sulfonic acid salts or organic sulfates. This forms graft polymers with high grafting yields, i.e. a large proportion of the polymer of the graft monomers is chemically bonded to the rubber. Graft polymers according to Component E, produced by emulsion polymerisation, are particularly preferred).

Component F

The very fine-particle inorganic powders F used according to the invention, preferably consist of at least one polar compound of one or more metals of the 1st to 5th main group or the 1st to 8th sub-group of the periodic system, preferably the 2nd to 5th main group or the 4th to 8th sub-group, in particular the 3rd to 5th main group or 4th to 8th sub-group, 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, hydrides, phosphites or phosphonates.

The very fine-particle inorganic powders preferably consist of oxides, phosphates, hydroxides, preferably of TiO_2, SiO_2, SnO_2, ZnO, ZnS, Boehmite, ZrO_2, Al_2O_3, aluminium phosphates, iron oxides, also TiN, WC, AlO(OH), Sb_2O_3, iron oxides, $NaSO_4$, vanadium oxides, zinc borate, silicates such as Al-silicates, Mg-silicates, one- two- and three-dimensional silicates. Mixtures and doped compounds may also be used.

Furthermore, these nanoscale particles may be surface-modified with organic molecules, to achieve better compatibility with the polymers. In this way, hydrophobic or hydrophilic surfaces can be produced.

Hydrate-containing aluminium oxides, such as Boehmite, talc or TiO_2 are particularly preferred.

The average maximum particle diameters of the fine-particle inorganic materials are less than or equal to 1000 nm, preferably less than or equal to 500 nm, in particular less than or equal to 200 nm.

The inorganic compounds may be present as powders, pastes, sols, dispersions or suspensions. Powders may be obtained by precipitation from the dispersions, sols or suspensions.

Component G

The compositions according to the invention may further contain up to 10 parts by weight, preferably up to 5 parts by weight, in particular up to 2 parts by weight of at least one conventional polymer additive such as mould lubricant and mould release agent, for example pentaerythritol tetrastearate, nucleation agent, antistatic, stabiliser, light-protection agent, hydrolysis stabiliser, filler or reinforcing agent, dye or pigment as well as a further flame retardant or a flame-retarding synergist.

All contents by weight given in this application are standardised so that the sum of the parts by weight of all components in the composition is equal to 100.

The compositions according to the invention are produced by mixing the respective components in the known way and melt compounding and melt extruding them at temperatures of 200°C to 300°C in conventional units such as internal kneaders, extruders and twin-shaft screws.

The individual components may be mixed in the known way either successively or simultaneously, and either at about 20°C (room temperature) or at a higher temperature.

The invention therefore also provides a process for the production of the compositions.

The compositions according to the invention may be used to produce moulded parts of any kind. These can be produced, for example, by injection moulding, extrusion and blow-moulding processes. A further form of processing is the production of moulded bodies by deep-drawing from previously produced sheets or films. The invention therefore also provides the moulded parts that can be obtained from the composition according to the invention.

Examples of such moulded parts are films, profiles, housing parts of all kinds e.g. for domestic appliances such as juicers, coffee machines, food mixers; for office equipment such as monitors, printers, copiers, Notebooks and mobile phones; also sheets, tubes, ducting for electrical installations, profiles for the building industry, internal fittings and external applications; parts for the electrotechnical sector such as switches and plugs and internal and external automobile components.

The compositions according to the invention can be used in particular, for example, for the production of the following moulded parts:

Internal components for rail vehicles, ships, aircraft, buses and cars, hub caps, casings for electrical equipment containing small transformers, casings for equipment used for the dissemination and transmission of information, cases and linings for medical applications, massage devices and casings for them, children's toy vehicles, wall elements in sheet form, cases for safety equipment, rear spoilers, bodywork parts and internal components for motor vehicles, heat-insulated transport containers, devices for holding and caring for small animals, moulded parts for sanitary and bathroom fittings, covering grilles for air vents, moulded parts for garden sheds and tool sheds, cases for garden tools.

The following examples explain the invention in more detail.

Examples

Component A1

Linear polycarbonate based on bisphenol A, with a weight average molecular weight $\overline{M}_w$ of 26,000 g/mol (measured by GPC).

Component A2

Linear polycarbonate based on bisphenol A with a weight average molecular weight $\overline{M}_w$ of 27,000 g/mol (measured by GPC).

Component B1

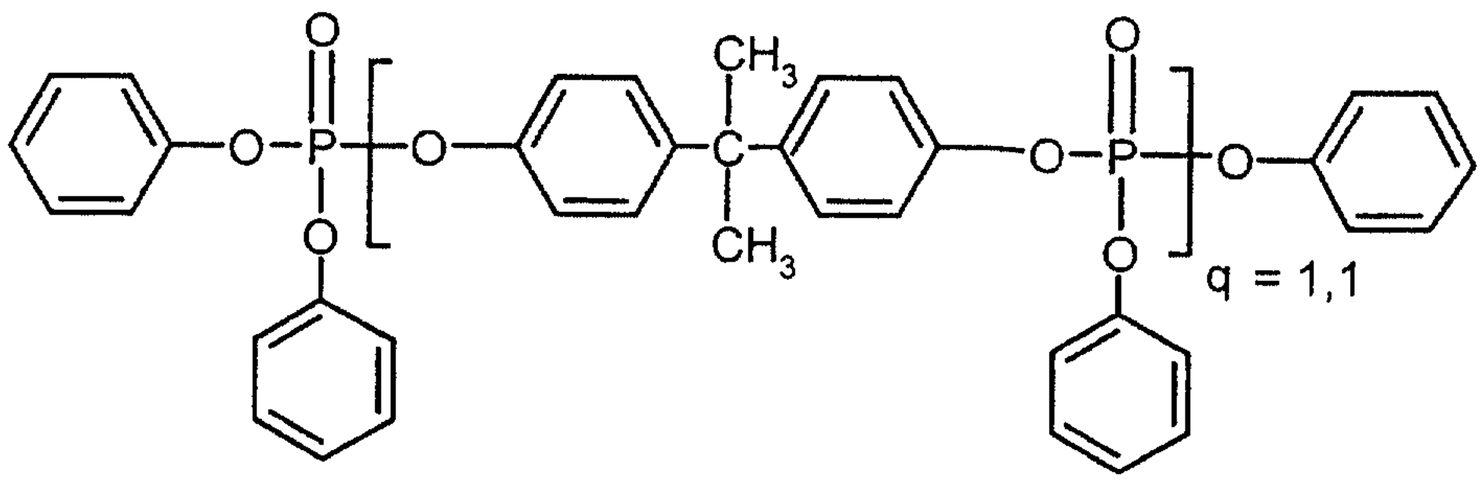
ABS graft polymer produced by mass polymerisation with an A/B/S weight ratio of 20%/13%/67% containing SAN with a weight average molecular weight $\overline{M}_w$ of 80,000 g/mol (measured by GPC).

Component B2

ABS graft polymer produced by mass polymerisation with an A/B/S weight ratio of 20%/15%/65% containing SAN with a weight average molecular weight $\overline{M}_w$ of 170,000 g/mol (measured by GPC).

Component C

Bisphenol A-bridged oligophosphate (BDP)



Component D

Blendex 449: PTFE preparation from General Electric Plastics consisting of 50 wt.% PTFE and 50 wt.% SAN copolymer.

Component E

Paraloid® EXL2600: MBS (Methyl(meth)acrylate-grafted butadiene-styrene rubber with a glass transition temperature of -80°C from Rohm & Haas, Antwerp, Belgium)

Component F

Plural® 200: nanoscale basic aluminium oxide from Condea (Hamburg, Germany)

Component G

Additive: **G1**: pentaerythritol tetrastearate (PETS)

G2: phosphite stabiliser

Production and testing of the moulding compositions according to the invention

The components are mixed in a twin-screw extruder (ZSK25) from Werner und Pfleiderer, at a mass temperature of 240°C. The moulded bodies are produced at the same mass temperature and a mould temperature of 80°C in an Arburg 270 E injection moulding machine.

The notched impact resistance a_k is measured to ISO 180/1 A.

The combustion behaviour of the samples is measured to UL-Subj. 94V on bars measuring 127 x 12.7 x 1.5 mm.

The melt viscosity is measured at 260°C and a shear rate of 1000 s^{-1} to DIN 54811.

The stress cracking behaviour under the influence of chemicals (ESC behaviour) is tested on bars measuring 80 mm x 10 mm x 4 mm. A mixture of 60 vol.% toluene and 40 vol.% isopropanol is used as the test medium. The test bodies are pre-strained using an arc-shaped jig (boundary fibre strain ϵ_x is 3.2%) and stored at 23°C in the test medium. The time to break is measured under these conditions.

The creep behaviour (creep resistance) is measured in a tensile test on shoulder bars measuring 70 mm x 40 mm x 10 mm. The bars are subjected to a constant tension of 50 MPa at 23°C and the strain as a function of the time to break is detected. The time to break is a measure of the creep resistance under these conditions.

Hydrolysis resistance is determined by means of the increase in the melt volume rate (MVR) measured at 240°C with a die load of 5 kg to ISO 1133 during storage of granulate of the compound for 100 h at 80°C and a relative atmospheric humidity of 100%. As hydrolytic degradation of the polycarbonate in the composition leads to an increase in the MVR, it is desirable to minimise the increase in MVR.

A summary of the characteristics of the moulding compositions according to the invention is given in Table 1.

The data show that using mass-ABS type 1 as opposed to the same composition with mass ABS type 2 results in clear improvements in melt flowability, creep resistance and resistance to stress cracking as under the influence of chemicals with equally good toughness and flame-resistance (cf. example 1 and reference example 1). Even when using a slightly higher-molecular polycarbonate advantages in melt flowability are still achieved with further improvements in creep resistance and resistance to stress cracking in comparison with the reference example (see example 3).

Adding nanoscale compounds increases the flowability of the composition and, in particular, improves its resistance to hydrolysis (cf. examples 1 and 2). Adding acrylate-containing graft polymers produces a considerable increase in notched impact resistance. If this is restricted to small concentrations, the other characteristics are impaired only slightly, so that the current prior art (reference example A) is still exceeded (cf. examples 1, 4, 5 and 6).

In particular, the addition of the acrylate-containing graft polymers does not have the detrimental effect on resistance to hydrolysis that is generally observed when

adding the emulsion ABS polymers used in the prior art to increase notched impact resistance.

Table 1: Moulding compositions and their characteristics

		A*	1	2	3	4	5	6
Components [parts by weight]								
A1	PC1	68.9	68.9	69.5	-	68.9	68.9	68.9
A2	PC2	-	-	-	68.9	-	-	-
B1	Mass ABS 1	-	16.4	16.6	16.4	15.9	15.4	14.4
B2	Mass ABS 2	16.4	-	-	-			
C	BDP	12.5	12.5	12.6	12.5	12.5	12.5	12.5
D	PTFE-SAN master batch	0.8	0.8	0.8	0.8	0.8	0.8	0.8
E	MBS	-	-	-	-	0.5	1.0	2.0
F	Nanoscale inorgan. material	0.9	0.9	-	0.9	0.9	0.9	0.9
G1	PETS	0.4	0.4	0.4	0.4	0.4	0.4	0.4
G2	Phosphite stabiliser	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Characteristics								
Creep resistance – time to creep failure [h]		8	13	15	15	n.m	n.m	n.m
Stress cracking resistance – time to break [min]		3	11	10	25	9	8	6
Hydrolysis resistance – MVR increase [%]		n.m	17	47	19	n.m	n.m	19
a_k [kJ/m²]		11	12	13	14	14	15	32
UL94V-score		V-0	V-0	V-0	V-0	V-0	V-0	V-0
Melt viscosity [Pas]		164	135	144	152	141	147	152

* Reference example

n.m = not measured

Claims

1. Composition containing

A) 40 to 95 parts by weight of aromatic polycarbonate or polyestercarbonate or mixtures thereof,

B) 5 to 40 parts by weight of a graft polymer produced by the mass polymerisation process from acrylonitrile and/or methacrylonitrile, butadiene and at least one styrene monomer,

C) 0 to 20 parts by weight of a halogen-free phosphorus compound,

D) 0 to 1 parts by weight fluorinated polyolefin,

E) 0 to 5 parts by weight of a polymer containing acrylate monomer and

F) 0 to 3 parts by weight of a fine-particle inorganic material with an average maximum particle diameter of no more than 1000 nm

provided that

(i) the polycarbonate or polyestercarbonate has a weight average molecular weight M_w of between 25,000 and 35,000 g/mol,

(ii) the graft polymer according to Component B) has a butadiene content of between 8 and 15 wt.% in relation to the graft polymer and an acrylonitrile and/or methacrylonitrile content of 15 to 30 wt.% in relation to the sum of acrylonitrile and/or methacrylonitrile and styrene monomer in graft polymer B and contains styrene monomer-(meth)acrylonitrile copolymer with a weight average molecular weight M_w of $5 \cdot 10^4$ to $14 \cdot 10^4$ g/mol.

2. Composition according to claim 1, in which the aromatic polycarbonate or polyestercarbonate has a weight average molecular weight M_w of between 26,000 and 31,000 g/mol.

3. Composition according to claim 1, in which Component B) is formed from acrylonitrile, butadiene and styrene.

4. Composition according to claim 1, in which Component B) has a butadiene content of 11 to 14 wt.% in relation to graft polymer B).

5. Composition according to claim 1, in which Component B) contains styrene-acrylic acid derivative copolymer with a weight average molecular weight M_w of $6 \cdot 10^4$ to $11 \cdot 10^4$ g/mol.

6. Composition according to claim 1, in which Component B) contains 21 to 26 wt.% acrylonitrile and/or methacrylonitrile in relation to the sum of the quantities of acrylonitrile and/or methacrylonitrile and styrene monomers.

7. Composition according to claim 1 having a content of aromatic poly(ester)carbonate of 60 to 80 parts by weight.

8. Composition according to claim 7 having a content of graft polymer B) produced by the mass polymerisation process of 10 to 20 parts by weight.

9. Composition according to claim 1 containing 8 to 16 parts by weight of halogen-free phosphorus compound.

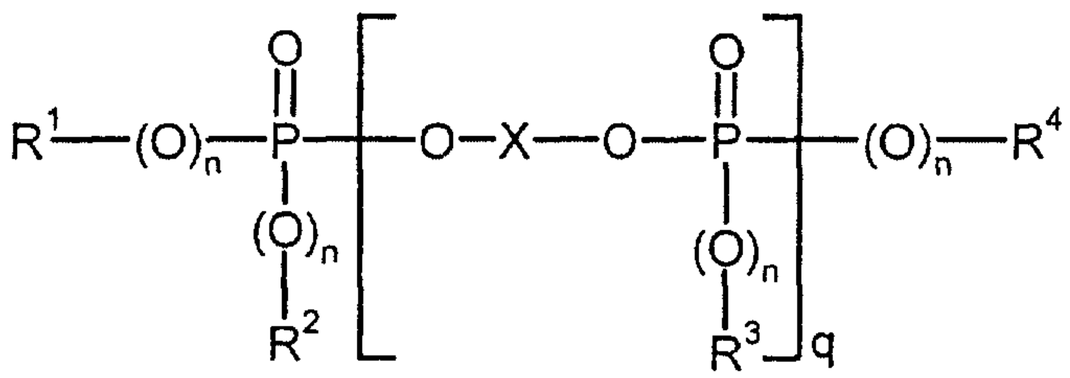
10. Composition according to claim 1 containing 0.2 to 0.5 parts by weight fluorinated polyolefin.

11. Composition according to claim 1 containing 0.5 to 2 parts by weight polymer E), which contains acrylate monomers.

12. Composition according to claim1 containing 0.2 to 1.5 parts by weight fine-particle inorganic materials.

13. Composition according to claim 1 containing at least one additive selected from the group of mould lubricants and mould release agents, nucleation agents, antistatics, stabilisers, UV-protectors, hydrolysis stabilisers, fillers and reinforcing agents, flame-retardants and synergists and also dyes and pigments.

14. Composition according to claim 1 containing as halogen-free phosphorus compound an oligomeric organic phosphoric acid ester of the general formula



in which

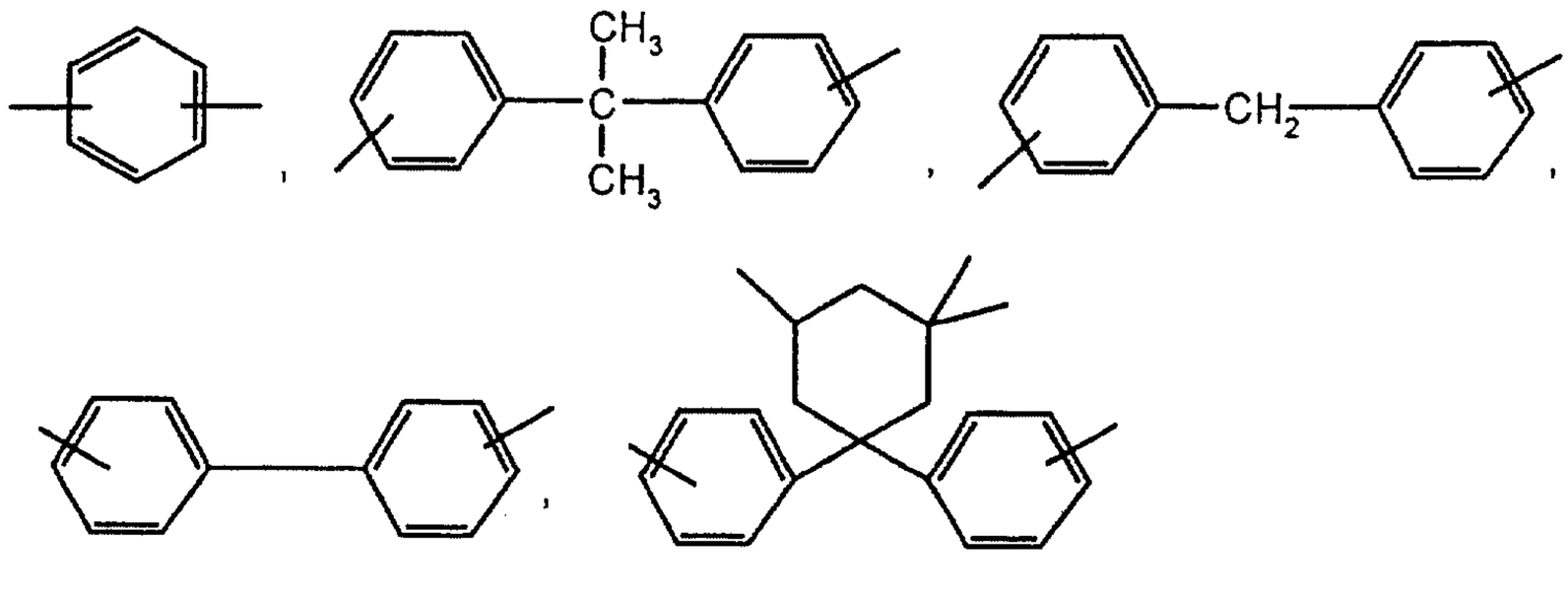
R^1, R^2, R^3 and R^4, independently of each other are, in each case, C_1 to C_8-alkyl, C_5 to C_6-cycloalkyl in each case optionally substituted by alkyl, C_6 to C_{20}-aryl or C_7 to C_{12}-aralkyl,

n independently of each other, means 0 or 1,

q means 0 to 30 and

X means a mono- or polynuclear aromatic group having 6 to 30 C atoms or a linear or branched aliphatic group having 2 to 30 C atoms, which may be OH-substituted and may contain up to 8 ether bonds.

15. Composition according to claim 14, wherein X represents



16. Composition according to claim 1, in which the fluorinated polyolefin is incorporated in the form of a master batch, a pre-compound or a co-precipitation of polytetrafluoroethylene and a vinyl monomer-containing polymer.

17. Composition according to claim 1, in which the polymer containing acrylate monomer is a graft polymer consisting of

10 to 90 wt.% of a mixture of

0 to 60 wt.% of at least one vinyl aromatic or core-substituted vinyl aromatic and

40 to 100 wt.% of at least one methacrylic acid-(C_1-C_8)-alkyl ester or acrylic acid-(C_1-C_8)-alkyl ester

on

90 to 10 wt.% of one or more rubbers with glass transition temperatures of <0°C.

18. Composition according to claim 17, in which the rubber component is selected from the group of polybutadiene rubbers, silicon rubbers and silicon-acrylate composite rubbers.

19. Composition according to claim 1, in which the fine-particle inorganic material has an average maximum particle diameter of no more than 200 nm.

20. Composition according to claim 1, which contains no acrylonitrile-butadiene-styrene copolymer produced by the emulsion-polymerisation process.

21. Moulded parts obtainable from the compositions according to claim 1.

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