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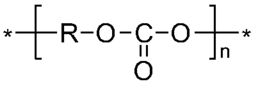
(54) **Title:** AROMATIC PHOSPHATE ACID ESTER FLAME RETARDANT COMPOSITIONS

(57) **Abstract:** The invention relates to flame retardant compositions, wherein at least one salt of an aromatic phosphoric acid ester is present in a polycarbonate substrate. The compositions attain the desirable V-0 rating, according to UL-94 (Underwriter's Laboratories Subject 94) and other excellent ratings in related test methods while preserving the excellent mechanical, chemical and thermal properties of polycarbonates, such as light transparency.

Aromatic Phosphate Acid Ester Flame Retardant Compositions

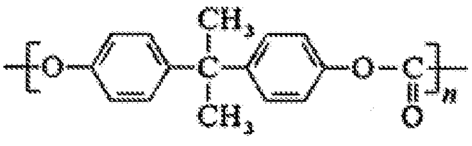
The invention relates to flame retardant compositions comprising at least one salt of an aromatic phosphoric acid ester and polycarbonates and to a process for imparting flame retardancy to a polymer substrate comprising polycarbonates and at least one salt of an aromatic phosphoric acid ester.

Polycarbonates are thermoplastic polymers of high toughness, outstanding transparency, excellent compatibility with several polymers, and high heat distortion resistance. Polycarbonates correspond to the general formula



$$*-\left[R-O-\underset{\underset{O}{\|}}{C}-O\right]_n-*$$

The economically most important polycarbonate is 2,2-bis(4-hydroxyphenyl)propane polycarbonate (1), also termed bisphenol A polycarbonate [24936-68-3] (BPA-PC):



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cf. *Ullmann's Encyclopaedia of Industrial Chemistry, On-Line Edition, Wiley-VCH, DOI: 10.1002/14356007.a21_207*, and entry *Polycarbonate, Roempp On-line, www.roempp.com*.

Various additives for improving the mechanical, chemical and thermal properties of polycarbonates are known. Fluorocarbon terminated poly-carbonates are useful for various technical applications, such as reducers of surface energy, "surface modifiers", for organic materials, preferably polycarbonates, polyesters, polyacrylates or polymethacrylates or their mixtures, blends or alloys. Polymers with such a reduced surface energy possess desirable properties, such as "easy to clean", "self-cleaning", "antisoiling", "soil-release", "antigraffiti", "oil resistance", "solvent resistance", "chemical resistance", "self lubricating", "scratch resistance", "low moisture absorption" and "hydrophobic" surface. The preparation of particularly useful fluorocarbon terminated polycarbonates is described in the *International Patent Application No. PCT/EP2004/053331*.

Flame retardants are added to polymeric materials (synthetic or natural) to enhance the flame retardant properties of the polymers. Depending on their composition, flame retardants may act in the solid, liquid or gas phase either chemically, e.g. as a spumescent by liberation of nitrogen, and/or physically, e.g. by producing a foam coverage. Flame retardants interfere during a particular stage of the combustion process, e.g. during heating, decomposition, ignition or flame spread.

The addition of flame retardants to polycarbonates is known, cf. *J. Troitzsch, Plastics Flammability Handbook, 3rd edition, Hanser Publishers, Munich 2004, pp. 158-172 (ISBN 3-446-21308-2).*

The addition of phosphate esters to polycarbonate flame retardant compositions is known, cf. *U.S. Patent Specification No. 6,756,433.*

In applications where a sample thickness smaller or equal than 1.6 mm is required, a flame retardancy of V-0, according to UL-94 (Underwriter's Laboratories Subject 94), is obtained by the addition of a so-called anti-dripping agent, such as polytetrafluoroethylene. The addition of aromatic phosphoric ester salts to polycarbonate flame retardant compositions containing tetrafluoroethylene polymers has been described in *U.S. Patent Specification No. 4,649,168*. Other co-additives for flame retardants have been proposed, such as haloarylphosphates, cf. *U.S. Patent Specification No. 5,478,874* or guanidine salts; cf. *U.S. Patent Specification No. 6,518,340*.

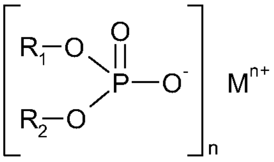
The addition of polysiloxanes of different structures has been proposed in various references; cf. *U.S. Patent Specification Nos. 6,660,787*, *6,727,302* or *6,730,720*. A problem of these additives is seen in the fact that the concentration of the flame retardant must be increased to arrive at the V-0 classification, which is detrimental to the mechanical, chemical and thermal properties of polycarbonates.

Therefore, the present invention relates to finding suitable additives, which are applicable in low concentrations as flame retardants in polycarbonates. It has surprisingly been found that salts of selected aromatic phosphoric acid esters, which are applicable as so-called nucleating agents, are particularly suitable as flame retardant additives, even at low concentrations. Nucleating agents are polymer additives that control the crystallization properties of crystalline polymers by increasing the crystallization temperature and/or accelerating the crystallization rate and/or decreasing the spherulite diameter and narrowing the size distribution.

Like other flame retardants of first choice, the salts of selected aromatic phosphoric acid esters are present in small quantities in the polycarbonates and, but due to the low dosing levels, have no significant negative effect on polymer mechanics and other properties.

The present invention relates to a composition, particularly a flame retardant composition, which comprises

a) At least one salt of an aromatic phosphoric acid ester of the formula



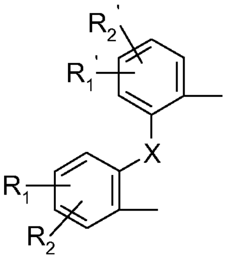
(I),

Wherein

One of R_1 and R_2 represents phenyl or phenyl substituted by one or two C_1-C_8alkyl; and the other one represents C_1-C_8alkyl; or

R_1 and R_2 both represent phenyl or phenyl substituted by one or two C_1-C_8alkyl; or

R_1 and R_2 together represent a group of the partial formula



(A),

Wherein

X represents C_1-C_4alkylene;

One of R_1 and R_2 represents hydrogen or C_1-C_8alkyl and the other one represents C_4-C_8alkyl; and

R_1' and R_2' are as defined as R_1 and R_2;

n represents 1 or 2; and

M^{n+} represents a cation that bears one positive charge if n represents 1; or

M^{n+} represents a cation that bears two positive charges if n represents 2; and

b) A polymer substrate comprising polycarbonate or polycarbonate blends.

A preferred embodiment of the invention relates to a composition, particularly a flame retardant composition, which comprises as component a) at least one salt of an aromatic phosphoric acid ester (I), wherein

One of R_1 and R_2 represents phenyl or phenyl substituted by one or two C_1-C_4alkyl; and the other one represents C_1-C_4alkyl; or

R_1 and R_2 both represent phenyl or phenyl substituted by one or two C_1-C_4alkyl; or

R_1 and R_2 together represent the group (A), wherein

X represents C_1-C_4alkylene;

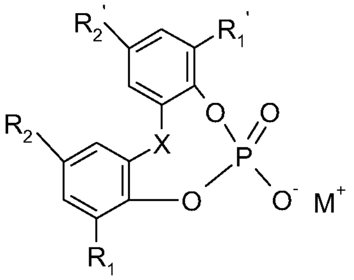
One of R_1 and R_2 represents hydrogen, methyl or tert-butyl and the other one represents tert-butyl; and

R_1' and R_2' are as defined as R_1 and R_2; and

n represents 1;

M^{n+} represents a cation that bears one positive charge.

A particularly preferred embodiment relates to a composition, particularly a flame retardant composition, which comprises as component a) at least one salt of an aromatic phosphoric acid ester of the formula



(I'),

Wherein

X represents C_1-C_4alkylene;

One of R_1 and R_2 represents hydrogen, methyl or tert-butyl and the other one represents tert-butyl; and

R_1' and R_2' are as defined as R_1 and R_2; and

M^+ represents a cation that bears one positive charge.

A highly preferred embodiment relates to a composition, particularly a flame retardant composition, which comprises as component a) at least one salt of an aromatic phosphoric acid ester (I'), wherein

X represents methylene;

One of R_1 and R_2 represents methyl or tert-butyl and the other one represents tert-butyl; and

R_1' and R_2' are as defined as R_1 and R_2; and

M^{n+} represents a cation that bears one positive charge.

Highest preference is given to a composition which comprises as component a) at least one salt of an aromatic phosphoric acid ester (I'), wherein

X represents methylene;

R_1 and R_2 and R_1' and R_2' represent tert-butyl; and

M^{n+} represents sodium or potassium.

The compositions according to the invention attain the desirable V-0 rating, according to UL-94 (Underwriter's Laboratories Subject 94) and other excellent ratings in related test methods while preserving the excellent mechanical, chemical and thermal properties of polycarbonates.

The composition, as defined above, comprises the following components:

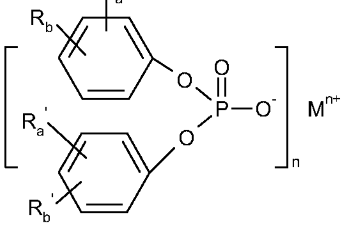
Component a)

The term salt of an aromatic phosphoric acid ester comprises within its scope preferably a metal salt, for example an alkali metal (n = 1) or alkaline earth metal salt (n = 2), e.g. the sodium, potassium, calcium salt.

According to an alternative embodiment, the term salts comprises non-metallic salts (n = 1), e.g. the ammonium, (C_1-C_{22}alkyl)$_{1\text{-}4}$ammonium or (2-hydroxyethyl)$_{1\text{-}4}$ammonium, e.g. tetramethylammonium, tetraethylammonium or the 2-hydroxyethyltrimethylammonium salt.

Therefore the definition of the cation M^{n+} in the formula above comprises within its scope an alkali metal atom, e.g. the sodium or potassium ion, the ammonium ion or a cation formed from an amine, e.g. (C_1-C_{22}alkyl)$_{1\text{-}4}$ammonium or (2-hydroxyethyl)$_{1\text{-}4}$ammonium, e.g. the tetramethylammonium, tetraethylammonium or the 2-hydroxyethyltrimethylammonium ions. In the aromatic phosphoric acid ester (I) C_1-C_8alkyl is present in R_1 and R_2 and, correspondingly in R_1' and R_2', is preferably methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl or tert-butyl.

Compounds (I) are exemplified by the following structural formulae:



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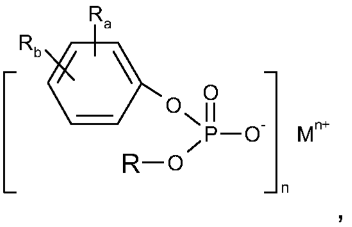
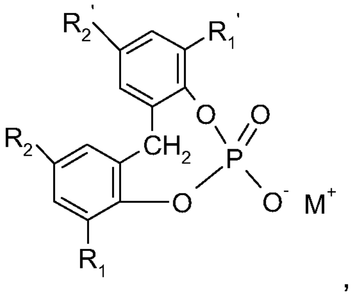
Wherein R_a represents hydrogen or C_1-C_8alkyl, R_b represents C_1-C_8alkyl and R_a' and R_b' are as defined as R_a and R_b, n represents 1 or 2, preferably 1, and M^{n+} represents a cation that bears one positive charge if n represents 1 or a cation that bears two positive charges if n represents 2;



Wherein R represents C_1-C_8alkyl, R_a represents hydrogen or C_1-C_8alkyl, R_b represents C_1-C_8alkyl, n represents 1 or 2, preferably 1, and M^{n+} represents a cation that bears one positive charge if n represents 1 or a cation that bears two positive charges if n represents 2;



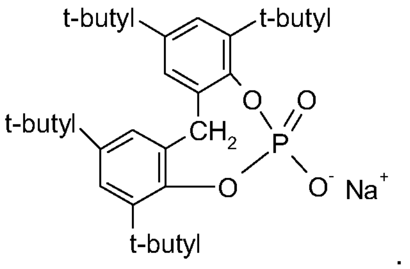
Wherein

One of R_1 and R_2 represents hydrogen, methyl or tert-butyl and the other one represents tert-butyl; and

R_1' and R_2' are as defined as R_1 and R_2; and

M^+ represents a cation that bears one positive charge, e.g. the sodium or potassium ion;

Particularly the compound:

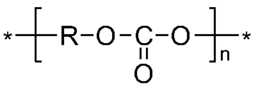


The aromatic phosphoric acid esters and their salts are known compounds. They are available by known methods. The preparation of aromatic phosphoric esters of the formula I' is described in the published *European Patent Application No. 1 209 190.*

Component b)

The polymer substrate comprising polycarbonates or polycarbonate blends may be of any grade and prepared by any known method. The term polymer substrate comprises within its scope any polycarbonate homopolymers or copolymers thereof, such as copolymers with polyesters.

Polycarbonates are thermoplastic polymers that correspond to the general formula:

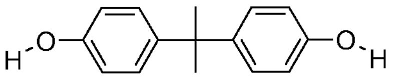


$$*\!\!-\!\!\left[R-O-\underset{\displaystyle O}{\underset{\|}{C}}-O\right]_n\!\!-\!\!*$$

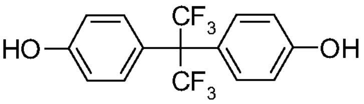
Polycarbonates are obtainable by interfacial processes or by melt processes (catalytic transesterification). The polycarbonate may be either branched or linear in structure and may include any functional substituents. Polycarbonate copolymers and polycarbonate blends are also within the scope of the invention. The term polycarbonate should be interpreted as inclusive of copolymers and blends with other thermoplastics. Methods for the manufacture of polycarbonates are known, for example, from *U.S. Patent Specification Nos. 3,030,331; 3,169,121; 4,130,458; 4,263,201; 4,286,083; 4,552,704; 5,210,268; and 5,606,007*. A combination of two or more polycarbonates of different molecular weights may be used.

Preferred are polycarbonates obtainable by reaction of a diphenol, such as bisphenol A, with a carbonate source. Examples of suitable diphenols are:

Bisphenol A:



, bisphenol AF:



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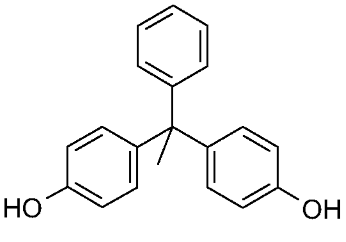
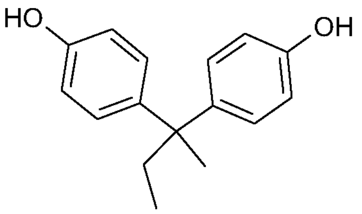
bisphenol AP:



, bisphenol B:



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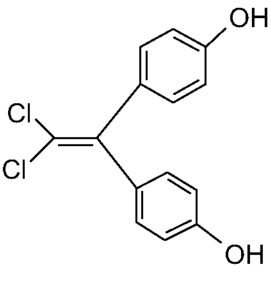
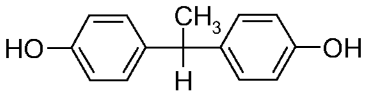
bisphenol C:



, bisphenol E:



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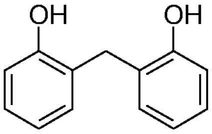
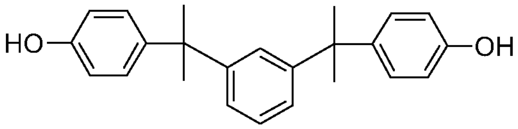
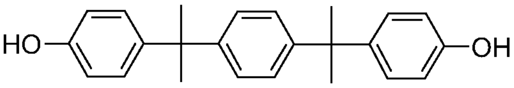
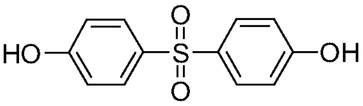
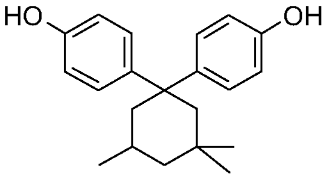
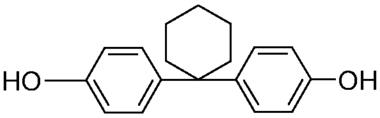
bisphenol F:  , bisphenol M:  ,

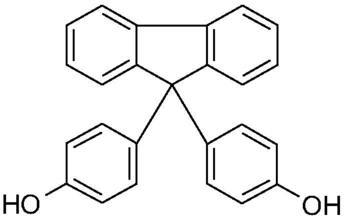
bisphenol P:  ,

bisphenol S:  , bisphenol TMC:  ,

bisphenol Z:  ,

4,4'-(2-norbornylidene)bis(2,6-dichlorophenol); or

fluorene-9-bisphenol:



The carbonate source may be either a carbonyl halide, a carbonate ester or a haloformate. Suitable carbonate halides are phosgene or carbonylbromide. Suitable carbonate esters are dialkylcarbonates, such as dimethyl- or diethylcarbonate, diphenyl carbonate, phenyl-alkylphenylcarbonate, such as phenyl-tolylcarbonate, dialkylcarbonates, such as dimethyl- or diethylcarbonate, di-(halophenyl)carbonates, such as di-(chlorophenyl)carbonate, di-(bromophenyl)carbonate, di-(trichlorophenyl)carbonate or di-(trichlorophenyl)carbonate, di-(alkylphenyl)carbonates, such as di-tolylcarbonate, naphthylcarbonate, dichloronaphthylcarbonate and others.

Other process details, such as the addition of molecular weight regulators, acid acceptors, catalysts are disclosed in the references mentioned above.

According to an additional embodiment, the polymer substrate comprising polycarbonates or polycarbonate blends is a polycarbonate-copolymer, wherein isophthalate/terephthalate-resorcinol segments are present. Such polycarbonates are commercially available, e.g.

Lexan® SLX (General Electrics Co. USA). Other polymeric substrates of component b) may additionally contain in the form as admixtures or as copolymers a wide variety of synthetic polymers including polyolefins, polystyrenes, polyesters, polyethers, polyamides, poly(meth)acrylates, thermoplastic polyurethanes, polysulphones, polyacetals and PVC, including suitable compatibilizing agents. For example, the polymer substrate may additionally contain thermoplastic polymers selected from the group of resins consisting of polyolefins, thermoplastic polyurethanes, styrene polymers and copolymers thereof. Specific embodiments include polypropylene (PP), polyethylene (PE), polyamide (PA), polybutylene terephthalate (PBT), polyethylene terephthalate (PET), glycol-modified polycyclohexylenemethylene terephthalate (PCTG), polysulphone (PSU), polymethylmethacrylate (PMMA), thermoplastic polyurethane (TPU), acrylonitrile-butadiene-styrene (ABS), acrylonitrile-styrene-acrylic ester (ASA), acrylonitrile-ethylene-propylene-styrene (AES), styrene-maleic anhydride (SMA) or high impact polystyrene (HIPS).

A list of suitable synthetic polymers is given below:

1. Polymers of monoolefins and diolefins, for example polypropylene, polyisobutylene, polybut-1-ene, poly-4-methylpent-1-ene, polyvinylcyclohexane, polyisoprene or polybutadiene, as well as polymers of cycloolefins, for instance of cyclopentene or norbornene, polyethylene (which optionally can be crosslinked), for example high density polyethylene (HDPE), high density and high molecular weight polyethylene (HDPE-HMW), high density and ultrahigh molecular weight polyethylene (HDPE-UHMW), medium density polyethylene (MDPE), low density polyethylene (LDPE), linear low density polyethylene (LLDPE), (VLDPE) and (ULDPE).

   Polyolefins, i.e. the polymers of monoolefins exemplified in the preceding paragraph, preferably polyethylene and polypropylene, can be prepared by different and especially by the following methods:

   a) Radical polymerisation (normally under high pressure and at elevated temperature).

   b) Catalytic polymerisation using a catalyst that normally contains one or more than one metal of groups IVb, Vb, VIb or VIII of the Periodic Table. These metals usually have one or more than one ligand, typically oxides, halides, alcoholates, esters, ethers, amines, alkyls, alkenyls and/or aryls that may be either π- or σ-coordinated. These metal complexes may be in the free form or fixed on substrates, typically on activated magnesium chloride, titanium(III) chloride, alumina or silicon oxide. These catalysts may be soluble or insoluble in the polymerisation medium. The catalysts can be used by themselves in the polymerisation or further activators may be used, typically metal alkyls, metal hydrides, metal alkyl halides, metal alkyl oxides or metal alkyloxanes, said metals being elements of groups Ia, IIa and/or IIIa of the Periodic Table. The activators may be modified conveniently with further ester, ether, and amine or silyl ether groups. These catalyst systems are usually termed Phillips, Standard Oil Indiana, Ziegler-Natta), TNZ (DuPont), metallocene or single site catalysts (SSC).

2. Mixtures of the polymers mentioned under 1), for example mixtures of polypropylene with polyisobutylene, polypropylene with polyethylene (for example PP/HDPE, PP/LDPE) and mixtures of different types of polyethylene (for example LDPE/HDPE).

3. Copolymers of monoolefins and diolefins with each other or with other vinyl monomers, for example ethylene/propylene copolymers, linear low density polyethylene (LLDPE) and mixtures thereof with low density polyethylene (LDPE), propylene/but-1-ene copolymers, propylene/isobutylene copolymers, ethylene/but-1-ene copolymers, ethylene/hexene copolymers, ethylene/methylpentene copolymers, ethylene/heptene copolymers, ethylene/octene copolymers, ethylene/vinylcyclohexane copolymers, ethylene/cycloolefin copolymers (e.g. ethylene/norbornene like COC), ethylene/1-olefins copolymers, where the 1-olefin is generated in-situ; propylene/butadiene copolymers, isobutylene/isoprene copolymers, ethylene/vinylcyclohexene copolymers, ethylene/alkyl acrylate copolymers, ethylene/alkyl methacrylate copolymers, ethylene/vinyl acetate copolymers or ethylene/acrylic acid copolymers and their salts (ionomers) as well as terpolymers of ethylene with propylene and a diene such as hexadiene, dicyclopentadiene or ethylidene-norbornene; and mixtures of such copolymers with one another and with polymers mentioned in 1) above, for example polypropylene/ethylene-propylene copolymers, LDPE/ethylene-vinyl acetate copolymers (EVA), LDPE/ethylene-acrylic acid copolymers (EAA), LLDPE/EVA, LLDPE/EAA and alternating or random polyalkylene/carbon monoxide copolymers and mixtures thereof with other polymers, for example polyamides.

4. Hydrocarbon resins (for example C_5-C_9) including hydrogenated modifications thereof (e.g. tackifiers) and mixtures of polyalkylenes and starch;

   The homopolymers and copolymers mentioned above may have a stereo structure including syndiotactic, isotactic, hemi-isotactic or atactic; where atactic polymers are preferred. Stereo block polymers are also included.

5. Polystyrene, poly (p-methylstyrene), poly(α-methylstyrene).

6. Aromatic homopolymers and copolymers derived from vinyl aromatic monomers including styrene, α-methylstyrene, all isomers of vinyl toluene, especially p-vinyl toluene, all isomers of ethyl styrene, propyl styrene, vinyl biphenyl, vinyl naphthalene, and vinyl anthra-

cene, and mixtures thereof. Homopolymers and copolymers may have a stereo structure including syndiotactic, isotactic, hemi-isotactic or atactic; where atactic polymers are preferred. Stereo block polymers are also included;

a) Copolymers including aforementioned vinyl aromatic monomers and comonomers selected from ethylene, propylene, dienes, nitriles, acids, maleic anhydrides, maleimides, vinyl acetate and vinyl chloride or acrylic derivatives and mixtures thereof, for example styrene/butadiene, styrene/acrylonitrile, styrene/ethylene (interpolymers), styrene/alkyl methacrylate, styrene/butadiene/alkyl acrylate, styrene/butadiene/alkyl methacrylate, styrene/maleic anhydride, styrene/acrylonitrile/methyl acrylate; mixtures of high impact strength of styrene copolymers and another polymer, for example a polyacrylate, a diene polymer or an ethylene/propylene/diene terpolymer; and block copolymers of styrene such as styrene/butadiene/styrene, styrene/isoprene/styrene, styrene/ethylene/butylene/styrene or styrene/ethylene/propylene/styrene.

b) Hydrogenated aromatic polymers derived from hydrogenation of polymers mentioned under 6.), especially including polycyclohexylethylene (PCHE) prepared by hydrogenating atactic polystyrene, often referred to as polyvinylcyclohexane (PVCH).

c) Hydrogenated aromatic polymers derived from hydrogenation of polymers mentioned under 6a). Homopolymers and copolymers may have a stereo structure including syndiotactic, isotactic, hemi-isotactic or atactic; where atactic polymers are preferred. Stereo block polymers are also included.

7. Graft copolymers of vinyl aromatic monomers such as styrene or α-methylstyrene, for example styrene on polybutadiene, styrene on polybutadiene-styrene or polybutadiene-acrylonitrile copolymers; styrene and acrylonitrile (or methacrylonitrile) on polybutadiene; styrene, acrylonitrile and methyl methacrylate on polybutadiene; styrene and maleic anhydride on polybutadiene; styrene, acrylonitrile and maleic anhydride or maleimide on polybutadiene; styrene and maleimide on polybutadiene; styrene and alkyl acrylates or methacrylates on polybutadiene; styrene and acrylonitrile on ethylene/propylene/diene terpolymers; styrene and acrylonitrile on polyalkyl acrylates or polyalkyl methacrylates, styrene and acrylonitrile on acrylate/butadiene copolymers, as well as mixtures thereof with the copolymers listed under 6), for example the copolymer mixtures known as ABS, MBS, ASA or AES polymers.

8. Halogen-containing polymers such as polychloroprene, chlorinated rubbers, chlorinated and brominated copolymer of isobutylene-isoprene (halobutyl rubber), chlorinated or sulphochlorinated polyethylene, copolymers of ethylene and chlorinated ethylene, epichlorohydrin homo- and copolymers, especially polymers of halogen-containing vinyl compounds, for example polyvinyl chloride, polyvinylidene chloride, polyvinyl fluoride, polyvinylidene fluoride, as well as copolymers thereof such as vinyl chloride/vinylidene chloride, vinyl chloride/vinyl acetate or vinylidene chloride/vinyl acetate copolymers.
9. Polymers derived from α,β-unsaturated acids and derivatives thereof such as polyacrylates and polymethacrylates; polymethyl methacrylates, polyacrylamides and polyacrylonitriles, impact-modified with butyl acrylate.
10. Copolymers of the monomers mentioned under 9) with each other or with other unsaturated monomers, for example acrylonitrile/ butadiene copolymers, acrylonitrile/alkyl acrylate copolymers, acrylonitrile/alkoxyalkyl acrylate or acrylonitrile/vinyl halide copolymers or acrylonitrile/ alkyl methacrylate/butadiene terpolymers.
11. Polymers derived from unsaturated alcohols and amines or the acyl derivatives or acetals thereof, for example polyvinyl alcohol, polyvinyl acetate, polyvinyl stearate, polyvinyl benzoate, polyvinyl maleate, polyvinyl butyral, polyallyl phthalate or polyallyl melamine; as well as their copolymers with olefins mentioned in 1. above.
12. Homopolymers and copolymers of cyclic ethers such as polyalkylene glycols, polyethylene oxide, polypropylene oxide or copolymers thereof with bisglycidyl ethers.
13. Polyacetals such as polyoxymethylene and those polyoxymethylenes, which contain ethylene oxide as a comonomer; polyacetals modified with thermoplastic polyurethanes, acrylates or MBS.
14. Polyphenylene oxides and sulphides, and mixtures of polyphenylene oxides with styrene polymers or polyamides.
15. Polyurethanes derived from hydroxyl-terminated polyethers, polyesters or polybutadienes on the one hand and aliphatic or aromatic polyisocyanates on the other, as well as precursors thereof.
16. Polyamides and copolyamides derived from diamines and dicarboxylic acids and/or from aminocarboxylic acids or the corresponding lactams, for example polyamide 4, polyamide 6, polyamide 6/6, 6/10, 6/9, 6/12, 4/6, 12/12, polyamide 11, polyamide 12, aromatic polyamides starting from m-xylene diamine and adipic acid; polyamides prepared from hexamethylenediamine and isophthalic or/and terephthalic acid and with or without an

elastomer as modifier, for example poly-2,4,4,-trimethylhexamethylene terephthalamide or poly-m-phenylene isophthalamide; and also block copolymers of the aforementioned polyamides with polyolefins, olefin copolymers, ionomers or chemically bonded or grafted elastomers; or with polyethers, e.g. with polyethylene glycol, polypropylene glycol or polytetramethylene glycol; as well as polyamides or copolyamides modified with EPDM or ABS; and polyamides condensed during processing (RIM polyamide systems).

17. Polyureas, polyimides, polyamide imides, polyether imides, polyester imides, polyhydantoins and polybenzimidazoles.

18. Polyesters derived from dicarboxylic acids and diols and/or from hydroxycarboxylic acids or the corresponding lactones, for example polyethylene terephthalate, polybutylene terephthalate, poly-1,4-dimethylolcyclohexane terephthalate, polyalkylene naphthalate (PAN) and polyhydroxybenzoates, as well as block copolyether esters derived from hydroxyl-terminated polyethers; and also polyesters modified with polycarbonates or MBS.

19. Polyketones.

20. Polysulphones, polyether sulphones and polyether ketones.

21. Blends of the aforementioned polymers (polyblends), for example PP/EPDM, Polyamide/EPDM or ABS, PVC/EVA, PVC/ABS, PVC/MBS, PC/ABS, PBTP/ABS, PC/ASA, PC/PBT, PVC/CPE, PVC/acrylates, POM/thermoplastic PUR, PC/thermoplastic PUR, POM/acrylate, POM/MBS, PPO/HIPS, PPO/PA 6.6 and copolymers, PA/HDPE, PA/PP, PA/PPO, PBT/PC/ABS or PBT/PET/PC.

Component a) is added to the substrate of component b) in an amount of about 0.001 to 5.0 weight%, preferably 0.01 to 2.0 weight% and most preferably 0.04 to 0.5 weight%.

A further embodiment of the invention relates to a process for imparting flame retardancy to a polymer substrate comprising polycarbonates or polycarbonate blends, which process comprises adding to said polymer substrate at least one salt of an aromatic phosphoric acid ester of the formula (I) as defined above.

Additional Components

The instant invention further pertains to a composition, which comprises, in addition to the components a) and b), as defined above, further additives selected from the group consisting of so-called anti-dripping agents, polymer stabilizers and additional flame-retardants, such as phosphorus containing flame-retardants, nitrogen containing flame-retardants, halogenated flame-retardants and inorganic flame-retardants.

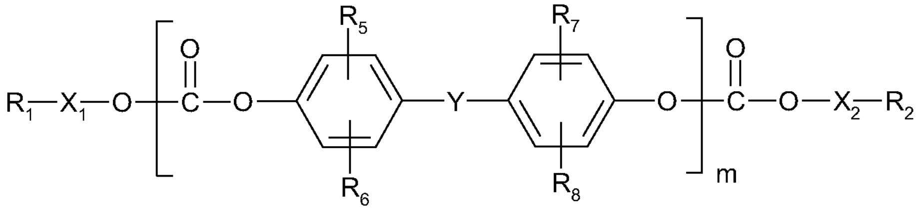
According to a preferred embodiment, the invention relates to a composition which additionally comprises as additional component so-called anti-dripping agents.

These anti-dripping agents reduce the melt flow of the polycarbonate composition and inhibit the formation of drops at high temperatures. Various references, such as *U.S. Patent Specification No. 4,263,201*, describe the addition of anti-dripping agents to polycarbonate flame retardant compositions.

Suitable additives that inhibit the formation of drops at high temperatures include glass fibers, polytetrafluoroethylene (PTFE), high temperature elastomers, carbon fibers, glass spheres and the like.

The addition of polysiloxanes of different structures has been proposed in various references; cf. *U.S. Patent Specification Nos. 6,660,787*, *6,727,302* or *6,730,720*.

According to a specific embodiment of the invention a (poly)carbonate compound of the formula



(1),

Wherein

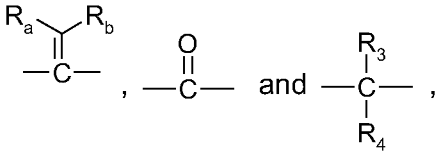
R_1 and R_2 independently of one another represent an aliphatic group substituted by fluorine;

X_1 and X_2 independently of one another represent the direct bond or C_1-C_{12}alkylene;

m represents a numeral from 1 to 1000;

R_5, R_6, R_7 and R_8 independently of one another represent hydrogen, C_1-C_{12}alkyl or C_3-C_{12}alkenyl; and

Y represents the direct bond or a bivalent group selected from the group consisting of -O-, -S-, -SO-, -SO$_2$-,



$-\overset{O}{\overset{\|}{C}}-$ and $-\underset{R_4}{\overset{R_3}{C}}-$,

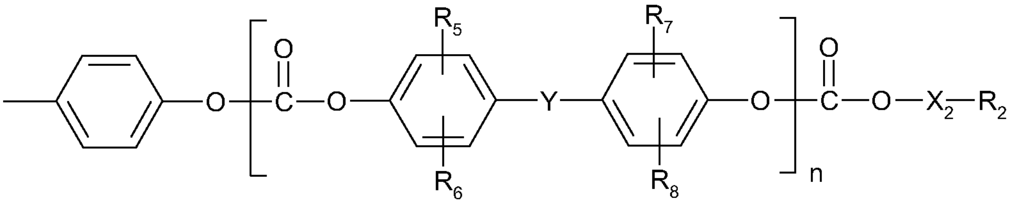
Wherein

Both R_a and R_b represent hydrogen or halogen; or

One of R_a and R_b represents hydrogen and the other one represents halogen;

R_3 and R_4, together with the carbon atom to which they are bonded, form a C_5-C_8-cycloalkylidene group with 1 to 3 C_1-C_4alkyl groups as optional substituents; or

R_3 and R_4 independently of one another represent hydrogen, an aliphatic group substituted by fluorine, C_1-C_{12}alkyl, C_1-C_{12}alkyl substituted by carboxy, C_2-C_{12}alkenyl, aryl, or the group of the partial formula


(A),

Wherein

n represents a numeral from 0-10 000; and

X_2, Y, R_2, R_5, R_6, R_7 and R_8 are as defined above.

In the (poly)carbonate compound (1), as defined above, the substituents are defined as follows:

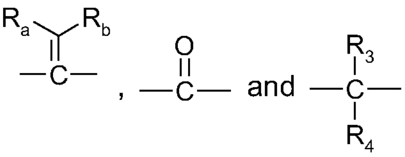
R_1 and R_2 independently of one another represent an aliphatic group substituted by fluorine;

X_1 and X_2 independently of one another represent the direct bond or C_1-C_{12}alkylene;

m represents a numeral from 1 to 1000;

R_5, R_6, R_7 and R_8 independently of one another represent hydrogen, C_1-C_{12}alkyl or C_3-C_{12}alkenyl; and

Y represents the direct bond or a bivalent group selected from the group consisting of

-O-, -S-, -SO-, $-SO_2-$,

$-C(=CR_aR_b)-$, $-C(=O)-$ and $-C(R_3)(R_4)-$, wherein

Both R_a and R_b represent hydrogen or halogen; or

One of R_a and R_b represents hydrogen and the other one represents halogen;

R_3 and R_4, together with the carbon atom to which they are bonded, form a C_5-C_8-cycloalkylidene group with 1 to 3 C_1-C_4alkyl groups as optional substituents; or

R_3 and R_4 independently of one another represent hydrogen, an aliphatic group substituted by fluorine, C_1-C_{12}alkyl, C_1-C_{12}alkyl substituted by carboxy, C_2-C_{12}alkenyl, aryl, or the group (A), as defined above, wherein n represents a numeral from 0-10 000 and X_2, Y, R_2, R_5, R_6, R_7 and R_8 are as defined above.

R_1 and R_2 defined as an aliphatic group substituted by fluorine is preferably a straight chain or branched hydrocarbon group, which contains at least one fluoro atom, for example fluoro-C_1-C_{25}alkyl, or is a perfluoroalkyl group of the partial formula

$$-(CF_2)_pF \quad (B),$$

wherein p is a numeral from 1 to 100.

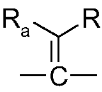
Fluoro-C_1-C_{25}alkyl is for example, mono-, difluoromethyl, 2-fluoroethyl, 3-fluoropropyl, 4-fluorobutyl, 5-fluoropentyl, 6-fluorohexyl, 7-fluoroheptyl or pentafluorobutyl.

Perfluoroalkyl is a group (B) derived from the perfluoro alcohol $F(CF_2)_p$-OH wherein p is 1 to 50, for example trifluoromethyl (p = 1) or pentafluoroethyl (p = 2). Preferred perfluoroalkyl groups are derived form perfluoro alcohols wherein p is 5, 8, 9 or 11.

X_1 and X_2 defined as C_1-C_{12}alkylene is a branched or unbranched bivalent group, for example methylene, ethylene, propylene, trimethylene, tetramethylene, pentamethylene, hexamethylene, heptamethylene, octamethylene, decamethylene or dodecamethylene. One of the preferred definitions for X_1 and X_2 is C_1-C_8alkylene, for example C_2-C_8alkylene. An especially preferred definition for X_1 and X_2 is C_2-C_4alkylene, for example ethylene.

R_5, R_6, R_7 and R_8 defined as C_1-C_{12}alkyl is a straight chain or, where possible, branched radical, for example methyl, ethyl, propyl, isopropyl, n-butyl, sec-butyl, isobutyl, tert-butyl, 2-ethylbutyl, n-pentyl, isopentyl, 1-methylpentyl, 1,3-dimethylbutyl, n-hexyl, 1-methylhexyl, n-heptyl, isoheptyl, 1,1,3,3-tetramethylbutyl, 1-methylheptyl, 3-methylheptyl, n-octyl, 2-ethylhexyl, 1,1,3-trimethylhexyl, 1,1,3,3-tetramethylpentyl, n-nonyl, n-decyl, n-undecyl, 1-methylundecyl or n-dodecyl. One of the preferred definitions is, for example, C_1-C_8alkyl, for example C_1-C_4alkyl, such as methyl.

R_5, R_6, R_7 and R_8 defined as C_3-C_{12}alkenyl is a straight chain or, where possible, branched radical, for example allyl, 2-butenyl, 3-butenyl, isobutenyl, n-2,4-pentadienyl, 3-methyl-2-butenyl, n-2-octenyl, n-2-dodecenyl or iso-dodecenyl.

Y defined as a bivalent group of the partial formula  $-C(=CR_aR_b)-$ is preferably methylene (CH_2=, R_a and R_b = H). According to alternative embodiments, both R_a and R_b represent hydrogen or halogen, for example chlorine or bromine, or one of R_a and R_b represents hydrogen and the other one represents halogen.

R_3 and R_4 defined as a C_5-C_8-cycloalkylidene group with 1 to 3 C_1-C_4alkyl groups as optional substituents is, for example, cyclopentylidene, methylcyclopentylidene, dimethylcyclopentylidene, cyclohexylidene, methylcyclohexylidene, dimethylcyclohexylidene, trimethylcyclohexylidene, tert-butylcyclohexylidene, cycloheptylidene or cyclooctylidene. Preference is given to cyclohexylidene.

R_3 and R_4 defined as an aliphatic group substituted by fluorine is for example fluoro-C_1-C_{25}alkyl, as defined above, or is the above-mentioned perfluoroalkyl group (B), wherein p is 1 to 50.

R_3 and R_4 defined as C_1-C_{12}alkyl is as defined above with regard to R_5, R_6, R_7 and R_8.

R_3 and R_4 defined as C_1-C_{12}alkyl substituted by carboxy is, for example, carboxymethyl or 1- or 2-carboxyethyl.

R_3 and R_4 defined as aryl is preferably phenyl or 1- or 2-naphthyl.

In the group (A) the index n represents a numeral from 0-10 000 and X_2, Y, R_2, R_5, R_6, R_7 and R_8 are as defined above.

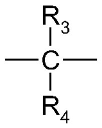
Of particular interest are (poly)carbonate compounds (1), wherein

R_1 and R_2 independently of one another represent an aliphatic group substituted by fluorine;

X_1 and X_2 independently of one another represent C_1-C_{12}alkylene;

m represents a numeral from 1 to 1 000;

R_5, R_6, R_7 and R_8 represent hydrogen;

Y represents the bivalent group  $-\overset{R_3}{\underset{R_4}{\overset{|}{\underset{|}{C}}}}-$, wherein independently of one another R_3 and R_4 represent hydrogen, $-CF_3$, C_1-C_{12}alkyl, phenyl or the group (A), wherein n represents a numeral from 0 to 10 000 and X_2, Y, R_2, R_5, R_6, R_7 and R_8 are as defined above or R_3 and R_4, together with the carbon atom to which they are bonded, form the cyclohexylidene group with 1 to 3 C_1-C_4alkyl groups as optional substituents.

Of particular interest are also (poly)carbonate compounds (1), wherein R_1 and R_2 independently of one another represent groups (B), wherein p is a numeral from 1 to 50.

Of special interest are (poly)carbonate compounds (1), wherein p is a numeral from 4 to 15.

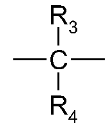
Of very special interest are (poly)carbonate compounds (1), wherein

R_1 and R_2 independently of one another represent groups (B), wherein p is a numeral from 1 to 50;

X_1 and X_2 independently of one another represent C_2-C_8alkylene;

m represents a numeral from 1 to 1 000;

R_5, R_6, R_7 and R_8 represent hydrogen; and

Y represents the bivalent group  $-\overset{R_3}{\underset{R_4}{C}}-$, wherein

R_3 represents hydrogen, $-CF_3$, C_1-C_{12}alkyl, phenyl or the group (A), wherein the numeral n represents a numeral from 0 to 10 000 and X_2, Y, R_2, R_5, R_6, R_7 and R_8 are as defined above or R_3 and R_4, together with the carbon atom to which they are bonded, form the cyclohexylidene group with 1 to 3 C_1-C_4alkyl groups as optional substituents.

Of high interest are (poly)carbonate compounds (1), wherein R_3 and R_4 independently of one another represent hydrogen or C_1-C_4alkyl; or R_3 and R_4, together with the carbon atom to which they are bonded, form the cyclohexylidene group.

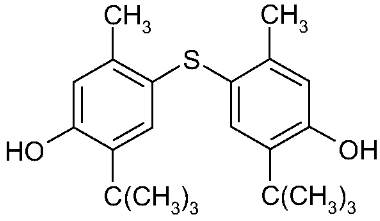
Particularly preferred are also (poly)carbonate compounds (1), wherein m is a numeral from 1 to 50, and n is a numeral from 0 to 50.

The (poly)carbonate compounds (1) are prepared by known methods. A fluoro alcohol is treated with bis(2,4-dinitrophenyl)carbonate (DNPC) to give the 2,4-dinitrophenyl carbonate of the fluoro alcohol *in situ*. This derivative can be isolated and treated separately, for example by hydroxy terminated bisphenol A oligomers of various molecular weights.

Brunelle et al., Macromolecules 1991, 24, 3035-3044, discloses the use of bis(2,4-dinitrophenyl)carbonate for preparation of dimer and cyclic oligomers of bisphenol A. The coupling reactions can also be carried out by carbonate linkage forming reagents, such as phosgene or carbonyl diimidazole (CDI).

Preferred fluoro alcohols are, for example, so-called fluorotelomer alcohols. These are, for example, commercially available from DuPont or Aldrich as Zonyl® BA-L.

Preferred bisphenol starting materials are, for example, bisphenol A and the compounds of the formulae:



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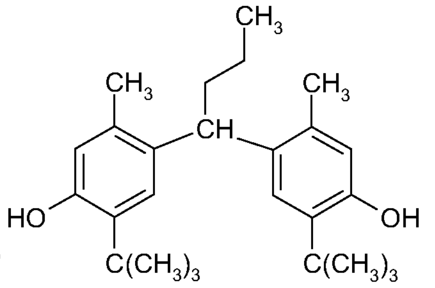

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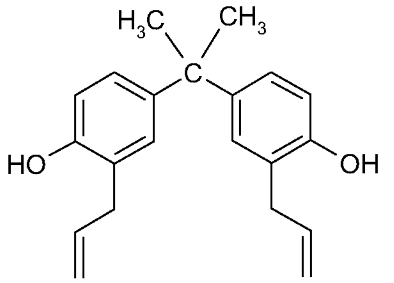

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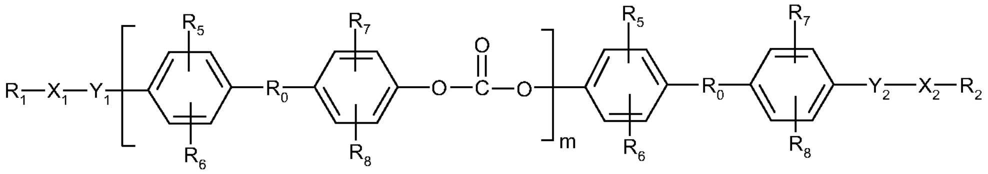
According to an alternative embodiment a (poly)carbonate compound of the formula



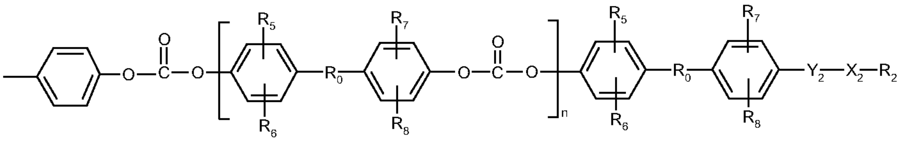
(2),

is added, wherein

R_0 represents the direct bond or a bivalent group selected from the group consisting of $-\overset{R_3}{\underset{R_4}{C}}-$, -O-, -S-, -SO-, -SO_2- and $-\overset{O}{\overset{\|}{C}}-$;

R_1 and R_2 independently of one another represent a silicon containing group;

R_3 and R_4 independently of one another represent hydrogen, an aliphatic group substituted by fluorine, a silicon containing group, C_1-C_{12}alkyl, C_1-C_{12}alkyl substituted by carboxy, C_2-C_{12}alkenyl, aryl, or a group of the partial formula



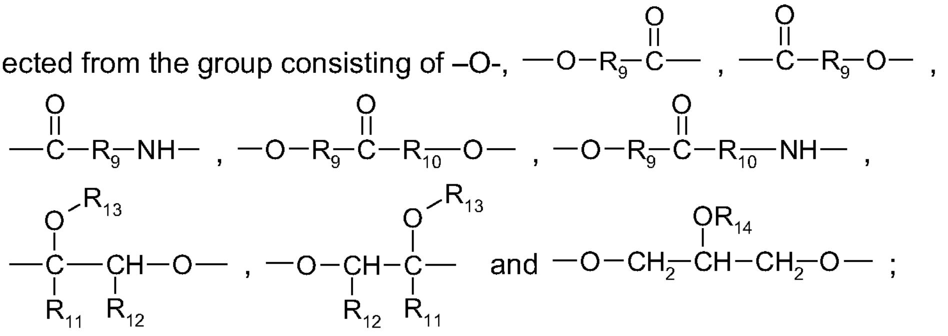
(C),

or R_3 and R_4, together with the carbon atom to which they are bonded represent C_5-C_8-cycloalkylidene or C_5-C_8-cycloalkylidene that is substituted by from 1 to 3 C_1-C_4alkyl groups;

R_5, R_6, R_7 and R_8 independently of one another represent hydrogen, C_1-C_{12}alkyl or C_3-C_{12}alkenyl;

X_1 and X_2 independently of one another represent the direct bond, C_1-C_{12}alkylene or C_4-C_{25}alkylene interrupted by –O-;

Y_1 and Y_2 independently of one another represent the direct bond or a bivalent group selected from the group consisting of –O-,



$-O-R_9-C(=O)-$, $-C(=O)-R_9-O-$, $-C(=O)-R_9-NH-$, $-O-R_9-C(=O)-R_{10}-O-$, $-O-R_9-C(=O)-R_{10}-NH-$, $-C(OR_{13})(R_{11})-CH(R_{12})-O-$, $-O-CH(R_{12})-C(OR_{13})(R_{11})-$ and $-O-CH_2-CH(OR_{14})-CH_2-O-$;

R_9 and R_{10} independently of one another represent the direct bond or C_1-C_4alkylene;

R_{11}, R_{12} and R_{13} independently of one another represent hydrogen, C_1-C_{12}alkyl or C_3-C_{12}alkenyl;

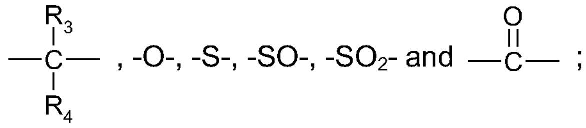
R_{14} represents hydrogen, C_1-C_{12}alkyl or a silicon containing group,

m represents a numeral from 0 to 10 000; and

n represents a numeral from 0 to 10 000.

In the (poly)carbonate compound (2), as defined above, the substituents are defined as follows:

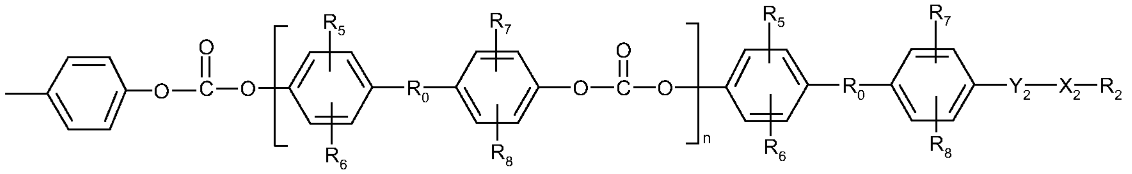
R_0 represents the direct bond or a bivalent group selected from the group consisting of



$-C(R_3)(R_4)-$, -O-, -S-, -SO-, $-SO_2-$ and $-C(=O)-$;

R_1 and R_2 independently of one another represent a silicon containing group;

R_3 and R_4 independently of one another represent hydrogen, an aliphatic group substituted by fluorine, a silicon containing group, C_1-C_{12}alkyl, C_1-C_{12}alkyl substituted by carboxy, C_2-C_{12}alkenyl, aryl, or a group of the partial formula



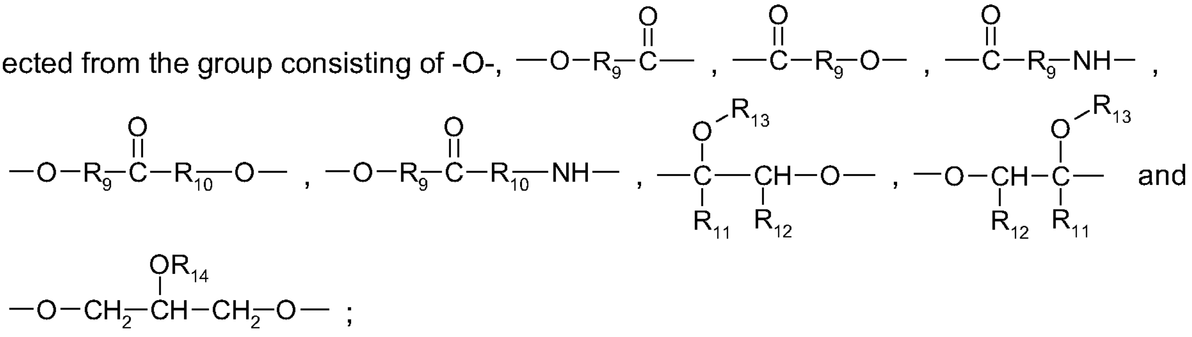
(C)

or R_3 and R_4, together with the carbon atom to which they are bonded represent C_5-C_8-cycloalkylidene or C_5-C_8-cycloalkylidene that is substituted by 1 to 3 C_1-C_4alkyl groups;

R_5, R_6, R_7 and R_8 independently of one another represent hydrogen, C_1-C_{12}alkyl or C_3-C_{12}alkenyl;

X_1 and X_2 independently of one another represent the direct bond, C_1-C_{12}alkylene or C_4-C_{25}alkylene interrupted by –O-;

Y_1 and Y_2 independently of one another represent the direct bond or a bivalent group selected from the group consisting of -O-,



$-O-R_9-\overset{O}{\overset{\|}{C}}-$, $-\overset{O}{\overset{\|}{C}}-R_9-O-$, $-\overset{O}{\overset{\|}{C}}-R_9-NH-$, $-O-R_9-\overset{O}{\overset{\|}{C}}-R_{10}-O-$, $-O-R_9-\overset{O}{\overset{\|}{C}}-R_{10}-NH-$, $-\underset{R_{11}}{\overset{OR_{13}}{C}}-\underset{R_{12}}{CH}-O-$, $-O-\underset{R_{12}}{CH}-\underset{R_{11}}{\overset{OR_{13}}{C}}-$ and $-O-CH_2-\overset{OR_{14}}{CH}-CH_2-O-$;

R_9 and R_{10} independently of one another represent the direct bond or C_1-C_4alkylene;

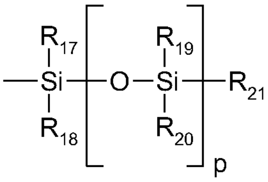
R_{11}, R_{12} and R_{13} independently of one another represent hydrogen, C_1-C_{12}alkyl or C_3-C_{12}alkenyl;

R_{14} represents hydrogen, C_1-C_{12}alkyl or a silicon containing group;

m represents a numeral from 0 to 10 000; and

n represents a numeral from 0 to 10 000.

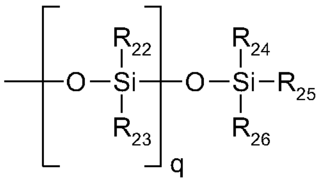
A silicon containing group preferably represents a group of the partial formula



$$-\underset{R_{18}}{\overset{R_{17}}{Si}}-\left[O-\underset{R_{20}}{\overset{R_{19}}{Si}}\right]_p-R_{21} \quad (D),$$

wherein

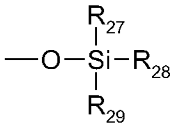
R_{17}, R_{18}, R_{19} and R_{20} independently of one another represent C_1-C_{12}alkyl, C_1-C_{12}alkyl substituted with hydroxy or amino; hydroxyC_4-C_{12}alkyl interrupted with –O-; or represents a group of the partial formula



$$-\left[O-\overset{R_{22}}{\underset{R_{23}}{Si}}\right]_q-O-\overset{R_{24}}{\underset{R_{26}}{Si}}-R_{25}$$ (E),

wherein

R_{21} represents C_1-C_{12}alkyl or a group of the partial formula



$$-O-\overset{R_{27}}{\underset{R_{29}}{Si}}-R_{28}$$ (F),

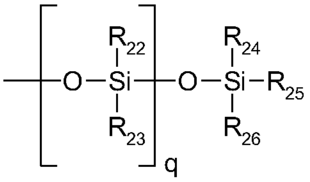
R_{22}, R_{23}, R_{24}, R_{25}, R_{26}, R_{27}, R_{28} and R_{29} independently of one another represent C_1-C_{12}alkyl or C_1-C_{12}-alkyl substituted with hydroxy or amino;

p represents 0 to 200; and

q represent 0 to 200.

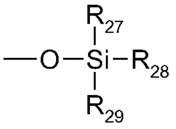
Of special interest as a silicon containing group is a group of the partial formula (D), wherein

R_{17}, R_{18}, R_{19} and R_{20} independently of one another represent methyl or a group of the partial formula



$$-\left[O-\overset{R_{22}}{\underset{R_{23}}{Si}}\right]_q-O-\overset{R_{24}}{\underset{R_{26}}{Si}}-R_{25}$$ (G),

R_{21} represents methyl or a group of the partial formula

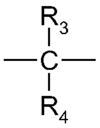


$$-O-\overset{R_{27}}{\underset{R_{29}}{Si}}-R_{28}$$ (H),

R_{22}, R_{23}, R_{24}, R_{25}, R_{26}, R_{27}, R_{28} and R_{29} are methyl; and

p and q independently of one another represent 0 to 100.

Of particular interest are (poly)carbonate compounds (2), wherein

R_0 represents the bivalent group  $-\overset{R_3}{\underset{R_4}{C}}-$;

R_1 and R_2 independently of one another represent a silicon containing group;

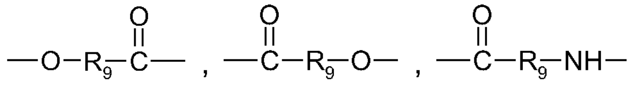
R_3 and R_4 independently of one another represent hydrogen, trifluoromethyl, a silicon containing group, C_1-C_{12}alkyl, phenyl or the group (C); or

R_3 and R_4, together with the carbon atom to which they are bonded represent C_5-C_8-cycloalkylidene or C_5-C_8-cycloalkylidene that is substituted by 1 to 3 C_1-C_4alkyl groups;

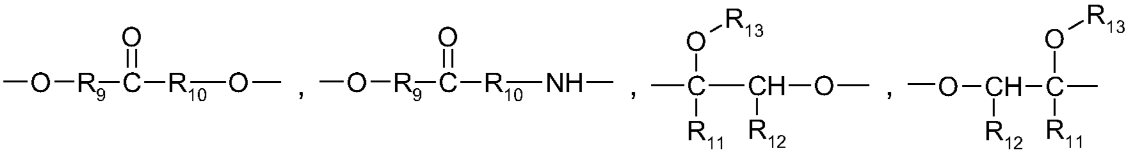
R_5, R_6, R_7 and R_8 are hydrogen;

X_1 and X_2 independently of one another represent C_1-C_{12}alkylene or C_4-C_{25}alkylene interrupted by –O-;

Y_1 and Y_2 independently of one another represent the direct bond or a bivalent group selected from the group consisting of –O-,

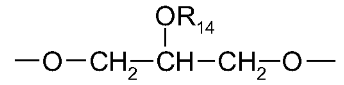


$$-O-R_9-\overset{O}{\overset{\|}{C}}-\ ,\quad -\overset{O}{\overset{\|}{C}}-R_9-O-\ ,\quad -\overset{O}{\overset{\|}{C}}-R_9-NH-\ ,$$



$$-O-R_9-\overset{O}{\overset{\|}{C}}-R_{10}-O-\ ,\quad -O-R_9-\overset{O}{\overset{\|}{C}}-R_{10}-NH-\ ,\quad -\underset{R_{11}}{\overset{OR_{13}}{C}}-\underset{R_{12}}{CH}-O-\ ,\quad -O-\underset{R_{12}}{CH}-\underset{R_{11}}{\overset{OR_{13}}{C}}-$$

and



$$-O-CH_2-\overset{OR_{14}}{CH}-CH_2-O-\ ;$$

R_9 and R_{10} independently of one another represent the direct bond or methylene;

R_{11}, R_{12} and R_{13} independently of one another represent hydrogen, C_1-C_4alkyl or C_3-C_4alkenyl;

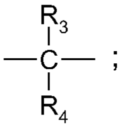
R_{14} represents hydrogen or C_1-C_{12}alkyl;

m represents 0 to 10 000; and

n represents 0 to 10 000.

Of very special interest are (poly)carbonate compounds (2), wherein

R_0 represents the bivalent group



$$-\underset{R_4}{\overset{R_3}{C}}-\ ;$$

R_3 represents hydrogen, $-CF_3$, C_1-C_{12}alkyl, phenyl or the group (C);

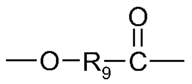
R_4 represents $-CF_3$, C_1-C_{12}alkyl or phenyl; or

R_3 and R_4, together with the carbon atom to which they are bonded, form a C_5-C_8-cycloalkylidene group or C_5-C_8-cycloalkylidene that is substituted by 1 to 3 C_1-C_4alkyl groups;

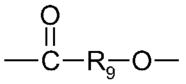
R_5, R_6, R_7 and R_8 represent hydrogen;

X_1 and X_2 are each independently of the one another represent C_1-C_{12}alkylene or C_4-C_{25}alkylene interrupted by –O-;

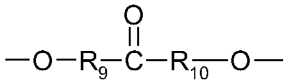
Y_1 and Y_2 independently of one another represent the direct bond or a bivalent group selected from the group consisting of –O-,



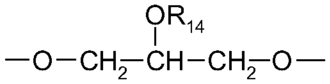
$-O-R_9-\overset{O}{\overset{\|}{C}}-$,



$-\overset{O}{\overset{\|}{C}}-R_9-O-$,



$-O-R_9-\overset{O}{\overset{\|}{C}}-R_{10}-O-$ and



$-O-CH_2-\overset{OR_{14}}{\overset{|}{CH}}-CH_2-O-$;

R_9 and R_{10} independently of one another represent the direct bond or methylene;

R_{14} represents hydrogen or C_1-C_{12}alkyl;

m represents 0 to 10 000; and

n represents 0 to 10 000.

Of interest are also (poly)carbonate compounds (2), wherein

R_3 and R_4 independently of one another represent hydrogen or C_1-C_4alkyl; or

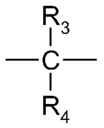
R_3 and R_4, together with the carbon atom to which they are bonded, form the cyclohexylidene group.

Preferred are (poly)carbonate compounds (2), wherein X_1 and X_2 independently of one another represent C_2-C_8alkylene or C_4-C_{25}alkylene interrupted with –O-.

Also preferred are (poly)carbonate compounds (2), wherein m represents 0 to 100, and n represents 0 to 100.

Of very special interest are (poly)carbonate compounds (2), wherein

R_0 represents the bivalent group



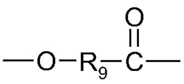
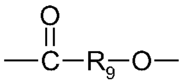
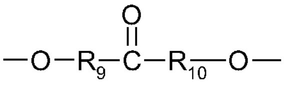
$-\overset{R_3}{\overset{|}{\underset{\underset{R_4}{|}}{C}}}-$;

R_3 and R_4 independently of one another represent C_1-C_4alkyl; or

R_3 and R_4, together with the carbon atom to which they are bonded, form the cyclohexylidene group;

R_5, R_6, R_7 and R_8 represent hydrogen;

X_1 and X_2 independently of one another represent C_2-C_4alkylene or C_4-C_{25}alkylene interrupted with –O-;

Y_1 and Y_2 independently of one another represent the direct bond or a bivalent group selected from the group consisting of –O-,  $-O-R_9-\overset{\overset{O}{\|}}{C}-$,  $-\overset{\overset{O}{\|}}{C}-R_9-O-$ and  $-O-R_9-\overset{\overset{O}{\|}}{C}-R_{10}-O-$;

R_9 and R_{10} independently of one another represent the direct bond or methylene;

m represents 0 to 100, and

n represents 0 to 100.

In a (poly)carbonate compound (2) C_1-C_{12}alkyl is a straight chain or, where possible, branched alkyl group, which is the same as defined above with regard to (poly)carbonate compounds (1).

R_3 and R_4 defined as C_1-C_{12}alkyl substituted by carboxy is preferably carboxymethyl or 1- or 2-carboxyethyl.

R_3 and R_4 defined as aryl preferably represent phenyl or phenyl substituted by 1-3 C_1-C_4alkyl groups, e.g. methyl.

R_3, R_4, R_5, R_6, R_7 and R_8 defined as C_2-C_{12}alkenyl represent a straight chain or, where possible, branched alkenyl group, which is the same as defined above with regard to (poly)carbonate compounds (1).

R_3 and R_4 defined as C_5-C_8-cycloalkylidene or C_5-C_8-cycloalkylidene that is substituted by from 1 to 3 C_1-C_4alkyl groups are as defined above with regard to (poly)carbonate compounds (1).

X_1, and X_2, defined as C_1-C_{12}alkylene and R_9 and R_{10} defined as C_1-C_4alkylene represent straight chain or, where possible, branched alkylene groups as defined above with regard to (poly)carbonate compounds (1).

X_1, and X_2, defined as C_4-C_{25}alkylene interrupted with –O- is straight chain or, where possible, branched, for example $-CH_2CH_2-O-CH_2CH_2-$, $-CH_2CH_2CH_2-O-CH_2CH_2-$, $-CH_2CH_2CH_2-O-CH_2CH_2CH_2-$ or $-CH_2CH_2-O-CH_2CH_2-O-CH_2CH_2-$.

C_1-C_{12}Alkyl substituted with hydroxy or amino is, for example, hydroxymethyl, 1- or 2-hydroxyethyl or aminomethyl, 1- or 2-aminoethyl.

Hydroxy-C_4-C_{12}alkyl interrupted with –O- is for example $-CH_2CH_2-O-CH_2CH_2OH$ or $-CH_2CH_2-O-CH_2CH_2-O-CH_2CH_2OH$.

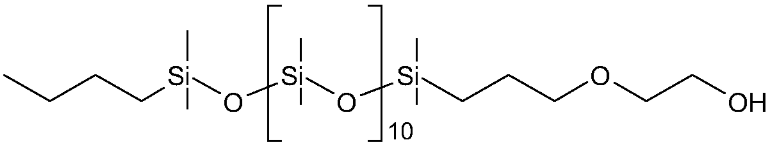
A fluorine containing group is a branched or unbranched radical, which contains at least one fluoro atom, for example fluoro-C_1-C_{25}alkyl; or is the group (B), wherein p is 1 to 50.

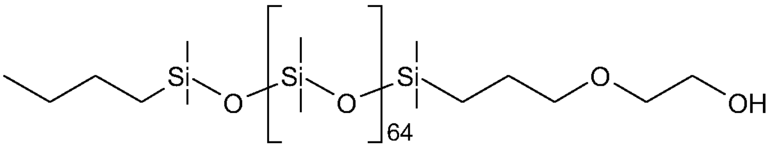
Fluoro-C_1-C_{25}alkyl is for example fluoromethyl, 2-fluoroethyl, 3-fluoropropyl, 4-fluorobutyl, pentafluorobutyl, 5-fluoropentyl, 6-fluorohexyl, 7-fluoroheptyl, difluoromethyl or pentafluorobutyl.

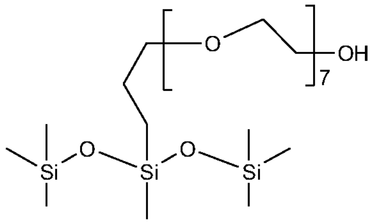
The group (B), wherein p is 1 to 50, is, for example trifluoromethyl or pentafluoromethyl.

The (poly)carbonate compounds (2) are obtainable by known methods. A silicon alcohol is treated with bis(2,4-dinitrophenyl)carbonate (DNPC) to give the 2,4-dinitrophenyl carbonate of the silicon alcohol *in situ*. This derivative can be isolated and treated separately, for example by hydroxy terminated bisphenol A oligomers of various molecular weights. *Brunelle et al., Macromolecules 1991, 24, 3035-3044*, discloses the use of bis(2,4-dinitrophenyl)carbonate for preparation of dimer and cyclic oligomers of bisphenol A. The coupling reactions can also be carried out by carbonate linkage forming reagents, such as phosgene or carbonyl diimidazole (CDI).

Especially preferred silicon containing groups are derived from mono hydroxypolysiloxanes, wherein p=10; polysiloxanes, wherein p=64; polyalkylene oxides modified heptamethyltrisiloxanes; or 3-(polyoxyethylene)propylheptamethyltrisiloxane. Representative structural formulae are shown below:







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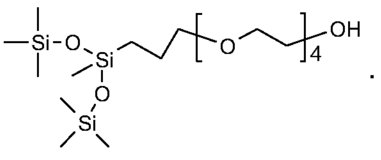

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Preferred bisphenol starting materials are the same as the ones mentioned above with regard to the preparation of the (poly)carbonate compounds (1).

(poly)carbonate compounds (1) or (2) are added to the substrate of component b) in an amount of about 0.01 to 5.0 weight% and preferably 0.25 to 1.0 weight%. The addition of mixtures of (poly)carbonate compounds (1) or (2) is suggested.

According to another embodiment the invention relates to a composition which additionally comprises further additives in selected from the group consisting of polymer stabilizers and additional flame-retardants.

Stabilizers are preferably halogen-free and selected from nitroxyl stabilizers, nitrone stabilizers, amine oxide stabilizers, benzofuranone stabilizers, phosphite and phosphonite stabilizers, quinone methide stabilizers and monoacrylate esters of 2,2'-alkylidenebisphenol stabilizers.

Additional flame-retardants as of present component are known components, items of commerce or can be obtained by known methods.

Representative phosphorus containing flame-retardants, in addition to the ones defined above with regard to component b), are for example:

Tetraphenyl resorcinol diphosphite (FYROLFLEX® RDP, Akzo Nobel), tetrakis(hydroxymethyl)phosphonium sulphide, triphenyl phosphate, diethyl-N,N-bis(2-hydroxyethyl)-aminomethyl phosphonate, hydroxyalkyl esters of phosphorus acids, ammonium polyphosphate (APP) or (HOSTAFLAM® AP750), resorcinol diphosphate oligomer (RDP), phosphazene flame-retardants and ethylenediamine diphosphate (EDAP).

Nitrogen containing flame-retardants are, for example, isocyanurate flame-retardants, such as polyisocyanurate, esters of isocyanuric acid or isocyanurates. Representative examples are hydroxyalkyl isocyanurates, such as tris-(2-hydroxyethyl)isocyanurate, tris(hydroxymethyl)isocyanurate, tris(3-hydroxy-n-proyl)isocyanurate or triglycidyl isocyanurate.

Nitrogen containing flame-retardants include melamine-based flame-retardants. Representative examples are: melamine cyanurate, melamine borate, melamine phosphates, melamine polyphosphate, melamine pyrophosphate, melamine ammonium polyphosphate and melamine ammonium pyrophosphate.

Further examples are: benzoguanamine, tris(hydroxyethyl) isocyanurate, allantoin, glycoluril, melamine cyanurate, melamine phosphate, dimelamine phosphate, melamine pyrophosphate, urea cyanurate, melamine polyphosphate, melamine borate, ammonium polyphosphate, melamine ammonium polyphosphate or melamine ammonium pyrophosphate, a condensation product of melamine from the series melem, melam, melon and/or a higher condensed compound or a reaction product of melamine with phosphoric acid and/or a reaction product of condensation products of melamine with phosphoric acid or a mixture thereof.

Special emphasis should be given to: dimelamine pyrophosphate, melamine polyphosphate, melem polyphosphate, melam polyphosphate, and/or a mixed polysalt of such a type, more especially melamine polyphosphate.

Representative organohalogen flame-retardants are, for example:

Polybrominated diphenyl oxide (DE-60F, Great Lakes Corp.), decabromodiphenyl oxide (DBDPO; SAYTEX® 102E), tris[3-bromo-2,2-bis(bromomethyl)propyl] phosphate (PB 370®, FMC Corp.), tris(2,3-dibromopropyl)phosphate, tris(2,3-dichloropropyl)phosphate, chlorendic acid, tetrachlorophthalic acid, tetrabromophthalic acid, poly-β-chloroethyl triphosphonate mixture, tetrabromobisphenol A bis(2,3-dibromopropyl ether) (PE68), brominated epoxy resin, ethylene-bis(tetrabromophthalimide) (SAYTEX® BT-93), bis(hexachlorocyclopentadieno)-cyclooctane (DECLORANE PLUS®), chlorinated paraffins, octabromodiphenyl ether, hexachlorocyclopentadiene derivatives, 1,2-bis(tribromophenoxy)ethane (FF680), tetrabromobisphenol A (SAYTEX® RB100), ethylene bis-(dibromo-norbornanedicarboximide) (SAYTEX® BN-451), bis-(hexachlorocycloentadeno) cyclooctane, PTFE, tris-(2,3-dibromopropyl)-isocyanurate, and ethylene-bis-tetrabromophthalimide.

The flame-retardant mentioned above routinely combined with an inorganic oxide synergist. Most common for this use are zinc or antimony oxides, e.g. Sb_2O_3 or Sb_2O_5. Boron compounds are suitable, too.

The above-mentioned additional flame-retardant classes are advantageously contained in the composition of the invention in an amount from about 0.5% to about 45.0% by weight of the organic polymer substrate; for instance about 1.0% to about 40.0%; for example about 5.0% to about 35.0% by weight of the polymer.

As mentioned above, the composition according to the invention may additionally contain one or more conventional additives, for example selected from pigments, dyes, plasticizers, antioxidants, thixotropic agents, levelling assistants, basic co-stabilizers, metal passivators, metal oxides, organophosphorus compounds, further light stabilizers and mixtures thereof, especially pigments, phenolic antioxidants, calcium stearate, zinc stearate, UV absorbers of the 2-hydroxy-benzophenone, 2-(2'-hydroxyphenyl)benzotriazole and/or 2-(2-hydroxyphenyl)-1,3,5-triazine groups. More specific examples are the following components:

1. Antioxidants

1.1. Alkylated monophenols, for example 2,6-di-tert-butyl-4-methylphenol, 2-tert-butyl-4,6-dimethylphenol, 2,6-di-tert-butyl-4-ethylphenol, 2,6-di-tert-butyl-4-n-butylphenol, 2,6-di-tert-butyl-4-isobutylphenol, 2,6-dicyclopentyl-4-methylphenol, 2-(α-methylcyclohexyl)-4,6-dimethylphenol, 2,6-dioctadecyl-4-methylphenol, 2,4,6-tricyclohexylphenol, 2,6-di-tert-butyl-4-methoxymethylphenol, nonylphenols which are linear or branched in the side chains, for example 2,6-di-nonyl-4-methylphenol, 2,4-dimethyl-6-(1'-methylundec-1'-yl)phenol, 2,4-dimethyl-6-(1'-methylheptadec-1'-yl)phenol, 2,4-dimethyl-6-(1'-methyltridec-1'-yl)phenol and mixtures thereof.

1.2. Alkylthiomethylphenols, for example 2,4-dioctylthiomethyl-6-tert-butylphenol, 2,4-dioctylthiomethyl-6-methylphenol, 2,4-dioctylthiomethyl-6-ethylphenol, 2,6-di-dodecylthiomethyl-4-nonylphenol.

1.3. Hydroquinones and alkylated hydroquinones, for example 2,6-di-tert-butyl-4-methoxyphenol, 2,5-di-tert-butylhydroquinone, 2,5-di-tert-amylhydroquinone, 2,6-diphenyl-4-octadecyloxyphenol, 2,6-di-tert-butylhydroquinone, 2,5-di-tert-butyl-4-hydroxyanisole, 3,5-di-tert-butyl-4-hydroxyanisole, 3,5-di-tert-butyl-4-hydroxyphenyl stearate, bis(3,5-di-tert-butyl-4-hydroxyphenyl) adipate.

1.4. Tocopherols, for example α-, β-, γ-, δ-tocopherol and mixtures thereof (vitamin E).

1.5. Hydroxylated thiodiphenyl ethers, for example 2,2'-thiobis(6-tert-butyl-4-methylphenol), 2,2'-thiobis(4-octylphenol), 4,4'-thiobis(6-tert-butyl-3-methylphenol), 4,4'-thiobis(6-tert-butyl-2-methylphenol), 4,4'-thiobis(3,6-di-sec-amylphenol), 4,4'-bis(2,6-dimethyl-4-hydroxyphenyl)disulphide.

1.6. Alkylidenebisphenols, for example 2,2'-methylenebis(6-tert-butyl-4-methylphenol), 2,2'-methylenebis(6-tert-butyl-4-ethylphenol), 2,2'-methylenebis[4-methyl-6-(α-methylcyclohexyl)phenol], 2,2'-methylenebis(4-methyl-6-cyclohexylphenol), 2,2'-methylenebis(6-nonyl-4-methylphenol), 2,2'-methylenebis(4,6-di-tert-butylphenol), 2,2'-ethylidenebis(4,6-di-tert-butylphenol), 2,2'-ethylidenebis(6-tert-butyl-4-isobutylphenol), 2,2'-methylenebis[6-(α-methylbenzyl)-4-nonylphenol], 2,2'-methylenebis[6-(α,α-dimethylbenzyl)-4-nonylphenol], 4,4'-methylenebis(2,6-di-tert-butylphenol), 4,4'-methylenebis(6-tert-butyl-2-methylphenol), 1,1-bis(5-tert-butyl-4-hydroxy-2-methylphenyl)butane, 2,6-bis(3-tert-butyl-5-methyl-2-hydroxybenzyl)-4-methylphenol, 1,1,3-tris(5-tert-butyl-4-hydroxy-2-methylphenyl)butane, 1,1-bis(5-tert-butyl-4-hydroxy-2-methylphenyl)-3-n-dodecylmercaptobutane, ethylene glycol bis[3,3-bis(3'-tert-butyl-4'-hydroxyphenyl)butyrate], bis(3-tert-butyl-4-hydroxy-5-methyl-phenyl)dicyclopentadiene, bis[2-(3'-tert-butyl-2'-hydroxy-5'-methylbenzyl)-6-tert-butyl-4-methylphenyl]-terephthalate, 1,1-bis-(3,5-dimethyl-2-hydroxyphenyl)butane, 2,2-bis(3,5-di-tert-butyl-4-hydroxyphenyl)propane, 2,2-bis-(5-tert-butyl-4-hydroxy-2-methylphenyl)-4-n-dodecylmercaptobutane, 1,1,5,5-tetra(5-tert-butyl-4-hydroxy-2-methylphenyl)pentane.

1.7. O-, N- and S-benzyl compounds, for example 3,5,3',5'-tetra-tert-butyl-4,4'-dihydroxydibenzyl ether, octadecyl-4-hydroxy-3,5-dimethylbenzylmercaptoacetate, tridecyl-4-hydroxy-3,5-di-tert-butylbenzylmercaptoacetate, tris(3,5-di-tert-butyl-4-hydroxybenzyl)amine, bis(4-tert-butyl-3-hydroxy-2,6-dimethylbenzyl)dithioterephthalate, bis(3,5-di-tert-butyl-4-hydroxybenzyl)sulphide, isooctyl-3,5-di-tert-butyl-4-hydroxybenzylmercaptoacetate.

1.8. Hydroxybenzylated malonates, for example dioctadecyl-2,2-bis(3,5-di-tert-butyl-2-hydroxybenzyl)malonate, di-octadecyl-2-(3-tert-butyl-4-hydroxy-5-methylbenzyl)malonate, di-dodecylmercaptoethyl-2,2-bis(3,5-di-tert-butyl-4-hydroxybenzyl)malonate, bis[4-(1,1,3,3-tetramethylbutyl)phenyl]-2,2-bis(3,5-di-tert-butyl-4-hydroxybenzyl)malonate.

1.9. Aromatic hydroxybenzyl compounds, for example 1,3,5-tris(3,5-di-tert-butyl-4-hydroxybenzyl)-2,4,6-trimethylbenzene, 1,4-bis(3,5-di-tert-butyl-4-hydroxybenzyl)-2,3,5,6-tetramethylbenzene, 2,4,6-tris(3,5-di-tert-butyl-4-hydroxybenzyl)phenol.

1.10. Triazine compounds, for example 2,4-bis(octylmercapto)-6-(3,5-di-tert-butyl-4-hydroxyanilino)-1,3,5-triazine, 2-octylmercapto-4,6-bis(3,5-di-tert-butyl-4-hydroxyanilino)-1,3,5-triazine, 2-octylmercapto-4,6-bis(3,5-di-tert-butyl-4-hydroxyphenoxy)-1,3,5-triazine, 2,4,6-tris(3,5-di-tert-butyl-4-hydroxyphenoxy)-1,2,3-triazine, 1,3,5-tris(3,5-di-tert-butyl-4-hydroxybenzyl)isocyanurate, 1,3,5-tris(4-tert-butyl-3-hydroxy-2,6-dimethylbenzyl)isocyanurate, 2,4,6-tris(3,5-di-tert-butyl-4-hydroxyphenylethyl)-1,3,5-triazine, 1,3,5-tris(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)-hexahydro-1,3,5-triazine, 1,3,5-tris(3,5-dicyclohexyl-4-hydroxybenzyl)isocyanurate.

1.11. Benzylphosphonates, for example dimethyl-2,5-di-tert-butyl-4-hydroxybenzylphosphonate, diethyl-3,5-di-tert-butyl-4-hydroxybenzylphosphonate, dioctadecyl3,5-di-tert-butyl-4-hydroxybenzylphosphonate, dioctadecyl-5-tert-butyl-4-hydroxy-3-methylbenzylphosphonate, the calcium salt of the monoethyl ester of 3,5-di-tert-butyl-4-hydroxybenzylphosphonic acid.

1.12. Acylaminophenols, for example 4-hydroxylauranilide, 4-hydroxystearanilide, octyl N-(3,5-di-tert-butyl-4-hydroxyphenyl)carbamate.

1.13. Esters of β-(3,5-di-tert-butyl-4-hydroxyphenyl)propionic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, n-octanol, i-octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl)isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane, such as commercially available products like Irganox® 1076

1.14. Esters of β-(5-tert-butyl-4-hydroxy-3-methylphenyl)propionic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, n-octanol, i-octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl)-isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane; 3,9-bis[2-{3-(3-tert-butyl-4-hydroxy-5-methylphenyl)propionyloxy}-1,1-dimethylethyl]-2,4,8,10-tetraoxaspiro[5.5]undecane.

1.15. Esters of β-(3,5-dicyclohexyl-4-hydroxyphenyl)propionic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl)isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo-[2.2.2]octane.

1.16. Esters of 3,5-di-tert-butyl-4-hydroxyphenyl acetic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl)isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.

1.17. Amides of β-(3,5-di-tert-butyl-4-hydroxyphenyl)propionic acid e.g. N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)hexamethylenediamide, N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)trimethylenediamide, N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)hydrazide, N,N'-bis[2-(3-[3,5-di-tert-butyl-4-hydroxyphenyl]propionyloxy)ethyl]oxamide (Naugard®XL-1, supplied by Uniroyal).

1.18. Ascorbic acid (vitamin C)

2. Light stabilisers

2.1. 2-(2'-Hydroxyphenyl)benzotriazoles, for example 2-(2'-hydroxy-5'-methylphenyl)benzotriazole, 2-(3',5'-di-tert-butyl-2'-hydroxyphenyl)benzotriazole, 2-(5'-tert-butyl-2'-hydroxyphenyl)benzotriazole, 2-(2'-hydroxy-5'-(1,1,3,3-tetramethylbutyl)phenyl)benzotriazole, 2-(3',5'-di-tert-butyl-2'-hydroxyphenyl)-5-chlorobenzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-methylphenyl)-5-chlorobenzotriazole, 2-(3'-sec-butyl-5'-tert-butyl-2'-hydroxyphenyl)benzotriazole, 2-(2'-hydroxy-4'-octyloxyphenyl)benzotriazole, 2-(3',5'-di-tert-amyl-2'-hydroxyphenyl)benzotriazole, 2-(3',5'-bis(α,α-dimethylbenzyl)-2'-hydroxyphenyl)benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-octyloxycarbonylethyl)phenyl)-5-chlorobenzotriazole, 2-(3'-tert-butyl-5'-[2-(2-ethylhexyloxy)carbonylethyl]-2'-hydroxyphenyl)-5-chlorobenzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-methoxycarbonylethyl)phenyl)-5-chlorobenzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-methoxycarbonylethyl)phenyl)benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-octyloxycarbonylethyl)phenyl)benzotriazole, 2-(3'-tert-butyl-5'-[2-(2-ethylhexyloxy)carbonylethyl]-2'-hydroxyphenyl)benzotriazole, 2-(3'-dodecyl-2'-hydroxy-5'-methylphenyl)benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-isooctyloxycarbonylethyl)phenylbenzotriazole, 2,2'-methylenebis[4-(1,1,3,3-tetramethylbutyl)-6-benzotriazole-2-ylphenol]; the transesterification product of 2-[3'-tert-butyl-5'-(2-methoxycarbonylethyl)-2'-hydroxyphenyl]-2H-benzotriazole with polyethylene glycol 300; $\left[R-CH_2CH_2-COO-CH_2CH_2\right]_2$,

where R = 3'-tert-butyl-4'-hydroxy-5'-2H-benzotriazol-2-ylphenyl, 2-[2'-hydroxy-3'-(α,α-dimethylbenzyl)-5'-(1,1,3,3-tetramethylbutyl)phenyl]benzotriazole; 2-[2'-hydroxy-3'-(1,1,3,3-tetramethylbutyl)-5'-(α,α-dimethylbenzyl)phenyl]benzotriazole, such as commercially available light stabilisers from the Tinuvin® series, such as TINUVIN 234, 326, 329, 350, 360 or TINUVIN 1577.

2.2. 2-Hydroxybenzophenones, for example the 4-hydroxy, 4-methoxy, 4-octyloxy, 4-decyloxy, 4-dodecyloxy, 4-benzyloxy, 4,2',4'-trihydroxy and 2'-hydroxy-4,4'-dimethoxy derivatives.

2.3. Esters of substituted and unsubstituted benzoic acids, for example 4-tert-butylphenyl salicylate, phenyl salicylate, octylphenyl salicylate, dibenzoyl resorcinol, bis(4-tert-butylbenzoyl)resorcinol, benzoyl resorcinol, 2,4-di-tert-butylphenyl 3,5-di-tert-butyl-4-hydroxybenzoate, hexadecyl 3,5-di-tert-butyl-4-hydroxybenzoate, octadecyl 3,5-di-tert-butyl-4-hydroxybenzoate, 2-methyl-4,6-di-tert-butylphenyl 3,5-di-tert-butyl-4-hydroxybenzoate.

2.4. <u>Acrylates</u>, for example ethyl α-cyano-β,β-diphenylacrylate, isooctyl α-cyano-β,β-diphenylacrylate, methyl α-carbomethoxycinnamate, methyl α-cyano-β-methyl-p-methoxycinnamate, butyl α-cyano-β-methyl-p-methoxycinnamate, methyl α-carbomethoxy-p-methoxycinnamate and N-(β-carbomethoxy-β-cyanovinyl)-2-methylindoline.

2.5. <u>Nickel compounds</u>, for example nickel complexes of 2,2'-thiobis[4-(1,1,3,3-tetramethylbutyl)phenol], such as the 1:1 or 1:2 complex, with or without additional ligands such as n-butylamine, triethanolamine or N-cyclohexyldiethanolamine, nickel dibutyldithiocarbamate, nickel salts of the monoalkyl esters, e.g. the methyl or ethyl ester, of 4-hydroxy-3,5-di-tert-butylbenzylphosphonic acid, nickel complexes of ketoximes, e.g. of 2-hydroxy-4-methylphenylundecylketoxime, nickel complexes of 1-phenyl-4-lauroyl-5-hydroxypyrazole, with or without additional ligands.

2.6. <u>Oxamides</u>, for example 4,4'-dioctyloxyoxanilide, 2,2'-diethoxyoxanilide, 2,2'-dioctyloxy-5,5'-di-tert-butoxanilide, 2,2'-didodecyloxy-5,5'-di-tert-butoxanilide, 2-ethoxy-2'-ethyloxanilide, N,N'-bis(3-dimethylaminopropyl)oxamide, 2-ethoxy-5-tert-butyl-2'-ethoxanilide and its mixture with 2-ethoxy-2'-ethyl-5,4'-di-tert-butoxanilide, mixtures of o- and p-methoxy-disubstituted oxanilides and mixtures of o- and p-ethoxy-disubstituted oxanilides.

2.7. <u>2-(2-Hydroxyphenyl)-1,3,5-triazines</u>, for example 2,4,6-tris(2-hydroxy-4-octyloxyphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-octyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2,4-dihydroxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2,4-bis(2-hydroxy-4-propyloxyphenyl)-6-(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-octyloxyphenyl)-4,6-bis(4-methylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-dodecyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-tridecyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-[2-hydroxy-4-(2-hydroxy-3-butyloxypropoxy)phenyl]-4,6-bis(2,4-dimethyl)-1,3,5-triazine, 2-[2-hydroxy-4-(2-hydroxy-3-octyloxypropyloxy)phenyl]-4,6-bis(2,4-dimethyl)-1,3,5-triazine, 2-[4-(dodecyloxy/tridecyloxy-2-hydroxypropoxy)-2-hydroxyphenyl]-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-[2-hydroxy-4-(2-hydroxy-3-dodecyloxypropoxy)phenyl]-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-hexyloxy)phenyl-4,6-diphenyl-1,3,5-triazine, 2-(2-hydroxy-4-methoxyphenyl)-4,6-diphenyl-1,3,5-triazine, 2,4,6-tris[2-hydroxy-4-(3-butoxy-2-hydroxypropoxy)phenyl]-1,3,5-triazine, 2-(2-hydroxyphenyl)-4-(4-methoxyphenyl)-6-phenyl-1,3,5-triazine, 2-{2-hydroxy-4-[3-(2-ethylhexyl-1-oxy)-2-hydroxypropyloxy]-phenyl}-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine.

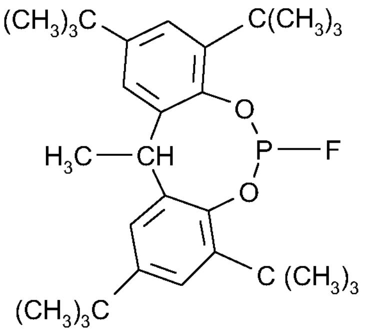
3. Metal deactivators, for example N,N'-diphenyloxamide, N-salicylal-N'-salicyloyl hydrazine, N,N'-bis(salicyloyl)hydrazine, N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)hydrazine, 3-salicyloylamino-1,2,4-triazole, bis(benzylidene)oxalyl dihydrazide, oxanilide, isophthaloyl dihydrazide, sebacoyl bisphenylhydrazide, N,N'-diacetyladipoyl dihydrazide, N,N'-bis(salicyloyl)oxalyl dihydrazide, N,N'-bis(salicyloyl)thiopropionyl dihydrazide.

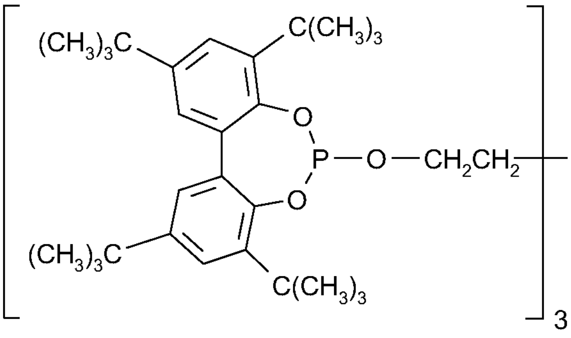
4. Further phosphites and phosphonites, for example triphenyl phosphite, diphenylalkyl phosphites, phenyldialkyl phosphites, tris(nonylphenyl) phosphite, trilauryl phosphite, trioctadecyl phosphite, distearylpentaerythritol diphosphite, tris(2,4-di-tert-butylphenyl) phosphite, diisodecyl pentaerythritol diphosphite, bis(2,4-di-tert-butylphenyl)pentaerythritol diphosphite, bis(2,4-di-cumylphenyl)pentaerythritol diphosphite, bis(2,6-di-tert-butyl-4-methylphenyl)pentaerythritol diphosphite, diisodecyloxypentaerythritol diphosphite, bis(2,4-di-tert-butyl-6-methylphenyl)pentaerythritol diphosphite, bis(2,4,6-tris(tert-butylphenyl)pentaerythritol diphosphite, tristearyl sorbitol triphosphite, tetrakis(2,4-di-tert-butylphenyl) 4,4'-biphenylene diphosphonite, 6-isooctyloxy-2,4,8,10-tetra-tert-butyl-12H-dibenz[d,g]-1,3,2-dioxaphosphocin, bis(2,4-di-tert-butyl-6-methylphenyl)methyl phosphite, bis(2,4-di-tert-butyl-6-methylphenyl)ethyl phosphite, 6-fluoro-2,4,8,10-tetra-tert-butyl-12-methyl-dibenz[d,g]-1,3,2-dioxaphosphocin, 2,2',2"-nitrilo[triethyltris(3,3",5,5'-tetra-tert-butyl-1,1'-biphenyl-2,2'-diyl)phosphite], 2-ethylhexyl(3,3',5,5'-tetra-tert-butyl-1,1'-biphenyl-2,2'-diyl)phosphite, 5-butyl-5-ethyl-2-(2,4,6-tri-tert-butylphenoxy)-1,3,2-dioxaphosphirane.

The following phosphites are especially preferred:

Tris(2,4-di-tert-butylphenyl) phosphite (Irgafos®168, Ciba Specialty Chemicals), tris(nonylphenyl) phosphite,

(A)





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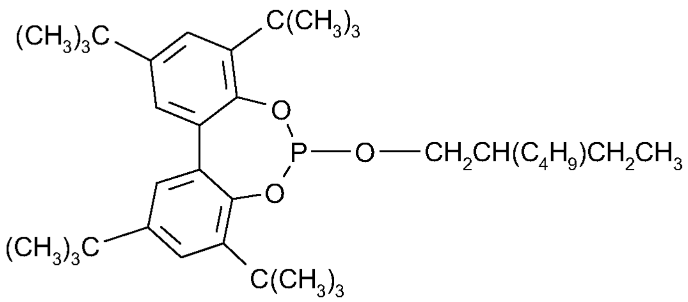

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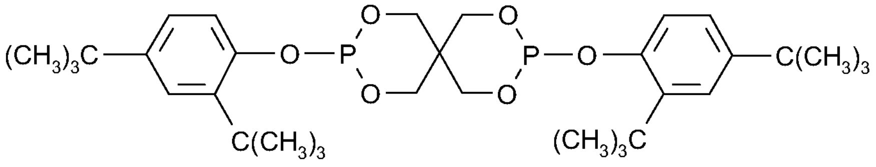

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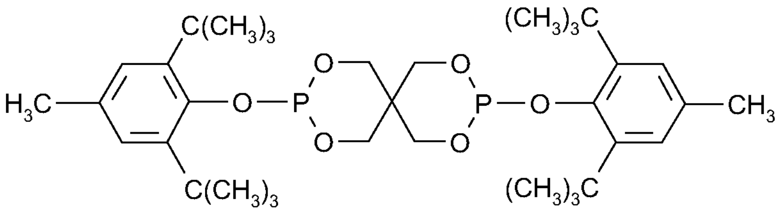

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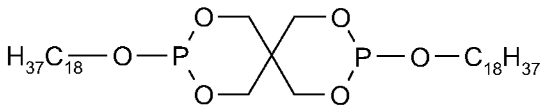

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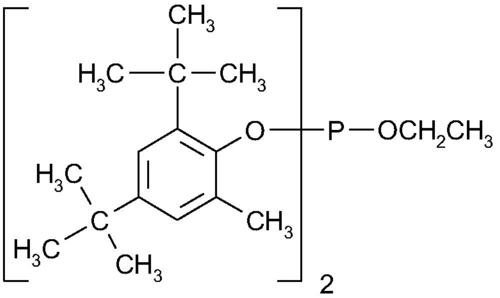


(G)

5. Further nitrones, for example N-benzyl-alpha-phenylnitrone, N-ethyl-alpha-methylnitrone, N-octyl-alpha-heptylnitrone, N-lauryl-alpha-undecylnitrone, N-tetradecyl-alpha-tridecylnitrone, N-hexadecyl-alpha-pentadecylnitrone, N-octadecyl-alpha-heptadecylnitrone, N-hexadecyl-alpha-heptadecylnitrone, N-ocatadecyl-alpha-pentadecylnitrone, N-heptadecyl-alpha-heptadecylnitrone, N-octadecyl-alpha-hexadecylnitrone, nitrone derived from N,N-dialkylhydroxylamine derived from hydrogenated tallow amine.

6. Thiosynergists, for example dilauryl thiodipropionate or distearyl thiodipropionate.

7. Peroxide scavengers, for example esters of β-thiodipropionic acid, for example the lauryl, stearyl, myristyl or tridecyl esters, mercaptobenzimidazole or the zinc salt of 2-mercaptobenzimidazole, zinc dibutyldithiocarbamate, dioctadecyl disulphide, pentaerythritol tetrakis(β-dodecylmercapto)propionate.

8. Polyamide stabilisers, for example copper salts in combination with iodides and/or phosphorus compounds and salts of divalent manganese.

9. Basic co-stabilisers, for example melamine, polyvinylpyrrolidone, dicyandiamide, triallyl cyanurate, urea derivatives, polyamides, polyurethanes, alkali metal salts and alkaline earth metal salts of higher fatty acids, for example calcium stearate, zinc stearate, magnesium behenate, magnesium stearate, sodium ricinoleate and potassium palmitate, antimony pyrocatecholate or zinc pyrocatecholate.

10. Nucleating agents, for example inorganic substances, such as talcum, metal oxides, such as titanium dioxide or magnesium oxide, phosphates, carbonates or sulphates of, preferably, alkaline earth metals; organic compounds, such as mono- or polycarboxylic acids and the salts thereof, e.g. 4-tert-butylbenzoic acid, adipic acid, diphenylacetic acid, sodium succinate or sodium benzoate; polymeric compounds, such as ionic copolymers (ionomers). Especially preferred are 1,3:2,4-bis(3',4'-dimethylbenzylidene)sorbitol, 1,3:2,4-di(paramethyldibenzylidene)sorbitol, and 1,3:2,4-di(benzylidene)sorbitol.

11. Further fillers and reinforcing agents, for example calcium carbonate, silicates, glass fibres, glass bulbs, stainless steel fibres, aramide fibers, asbestos, talc, kaolin, mica, barium sulphate, metal oxides and hydroxides, carbon black, graphite, wood flour and flours or fibers of other natural products, synthetic fibres.

12. Other additives, for example blend compatibilizing agents, plasticisers, lubricants, emulsifiers, pigments, rheology additives, catalysts, flow-control agents, optical brighteners, flame proofing agents, antistatic agents and blowing agents.

13. Additional benzofuranones and indolinones, for example those disclosed in *U.S. Patent Specification Nos. 4,325,863; 4,338,244; 5,175,312; 5,216,052; or 5,252,643; DE-A-4316611; DE-A-4316622; DE-A-4316876; EP-A-0589839* or *EP-A-0591102* or 3-[4-(2-acetoxyethoxy)phenyl]-5,7-di-tert-butylbenzofuran-2-one, 5,7-di-tert-butyl-3-[4-(2-stearoyloxyethoxy)phenyl]benzofuran-2-one, 3,3'-bis[5,7-di-tert-butyl-3-(4-[2-hydroxyethoxy]phenyl)benzofuran-2-one], 5,7-di-tert-butyl-3-(4-ethoxyphenyl)benzofuran-2-one, 3-(4-acetoxy-3,5-dimethylphenyl)-5,7-di-tert-butylbenzofuran-2-one, 3-(3,5-dimethyl-4-pivaloyloxyphenyl)-5,7-di-tert-butylbenzofuran-2-one, 3-(3,4-dimethylphenyl)-5,7-di-tert-butylbenzofuran-2-one, 3-(2,3-dimethylphenyl)-5,7-di-tert-butylbenzofuran-2-one.

Preferred additional additives for the compositions as defined above are processing stabilizers, such as the above-mentioned phosphites and phenolic antioxidants, and light stabilizers, such as benzotriazoles. Preferred specific antioxidants include octadecyl 3-(3,5-di-tert-butyl-4-hydroxyphenyl) propionate (IRGANOX 1076). Specific processing stabilizers include tris-(2,4-di-tert-butylphenyl) phosphite (IRGAFOS 168) and tetrakis(2,4-di-tert-butylphenyl)[1,1-biphenyl]-4,4'-diylbisphosphonite (IRGAFOS P-EPQ). Specific light stabilizers include 2-(2H-benzotriazole-2-yl)-4,6-bis(1-methyl-1-phenylethyl)phenol (TINUVIN 234), 2-(5-chloro(2H)-benzotriazole-2-yl)-4-(methyl)-6-(tert-butyl)phenol (TINUVIN 326), 2-(2H-benzotriazole-2-yl)-4-(1,1,3,3-tetramethylbutyl)phenol (TINUVIN 329), 2-(2H-benzotriazole-2-yl)-4-(tert-butyl)-6-(sec-butyl)phenol (TINUVIN 350), 2,2'-Methylenebis(6-(2H-benzotriazol-2-yl)-4-(1,1,3,3-tetram ethylbutyl)phenol) (TINUVIN 360), and 2-(4,6-Diphenyl-1,3,5-triazin-2-yl)-5-[(hexyl)-oxy]-phenol (TINUVIN 1577).

The additives mentioned above are preferably contained in an amount of 0.01 to 10.0%, especially 0.05 to 5.0%, relative to the weight of the polymer component b).

The incorporation of the additive component a) and optional further components into the polymer component b) is carried out by known methods such as dry blending in the form of a powder, or wet mixing in the form of solutions, dispersions or suspensions for example in an inert solvent, water or oil. The additive components a) and optional further additives may be incorporated, for example, before or after molding or also by applying the dissolved or dispersed additive or additive mixture to the polymer material, with or without subsequent evaporation of the solvent or the suspension/dispersion agent. They may be added directly into the processing apparatus (e.g. extruders, internal mixers, etc.), e.g. as a dry mixture or powder, or as a solution or dispersion or suspension or melt.

The addition of the additive components to the polymer substrate b) can be carried out in customary mixing machines in which the polymer is melted and mixed with the additives. Suitable machines are known to those skilled in the art. They are predominantly mixers, kneaders and extruders.

The process is preferably carried out in an extruder by introducing the additive during processing.

Particularly preferred processing machines are single-screw extruders, contra rotating and co-rotating twin-screw extruders, planetary-gear extruders, ring extruders or co kneaders. It is also possible to use processing machines provided with at least one gas removal compartment to which a vacuum can be applied.

Suitable extruders and kneaders are described, for example, in *Handbuch der Kunststoffextrusion, Vol. 1 Grundlagen, Editors F. Hensen, W. Knappe, H. Potente, 1989, pp. 3-7, ISBN:3-446-14339-4 (Vol. 2 Extrusionsanlagen 1986, ISBN 3-446-14329-7)*.

For example, the screw length is 1 - 60 screw diameters, preferably 35-48 screw diameters. The rotational speed of the screw is preferably 10 - 600 rotations per minute (rpm), preferably 25 - 300 rpm.

The maximum throughput is dependent on the screw diameter, the rotational speed and the driving force. The process of the present invention can also be carried out at a level lower than maximum throughput by varying the parameters mentioned or employing weighing machines delivering dosage amounts.

If a plurality of components is added, these can be premixed or added individually.

The additives component a) and optional further additives can also be sprayed onto the polymer substrate b). The additive mixture dilutes other additives, for example the conventional additives indicated above, or their melts so that they can be sprayed also together with these additives onto the polymer substrate. Addition by spraying during the deactivation of the polymerisation catalysts is particularly advantageous; in this case, the steam evolved may be used for deactivation of the catalyst. In the case of spherically polymerised polyolefins it may, for example, be advantageous to apply the additives of the invention, optionally together with other additives, by spraying.

The additive component a) and optional further additives can also be added to the polymer in the form of a master batch (“concentrate”) which contains the components in a concentration of, for example, about 1.0% to about 40.0% and preferably 2.0% to about 20.0% by weight incorporated in a polymer. The polymer is not necessarily of identical structure than the polymer where the additives are added finally. In such operations, the polymer can be used in the form of powder, granules, solutions, and suspensions or in the form of lattices.

Incorporation can take place prior to or during the shaping operation. The materials containing the additives of the invention described herein preferably are used for the production of molded articles, for example roto-molded articles, injection molded articles, profiles and the like, and especially a fiber, spun melt non-woven, film or foam.

Thus, present invention further pertains to a molded or extruded article, a fiber, spun melt non-woven or a foam comprising the composition of the invention.

The following examples illustrate the invention:

Examples

Materials and Methods

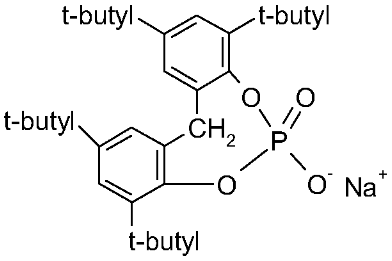
PC 145 Resin (GE Plastics) is vacuum-dried for 8 h at 120 °C and stabilized with IRGAFOS® P-EPQ (Ciba Specialty Chemicals). Dyneon PA5931(=PTFE) is used as an anti-dripping agent.

The polycarbonate compositions shown in Tab. 1 are extruded on a Haake TW-100 at 280°C and pelletized by strand granulation. After drying at 120°C for 12 h, the granulated compositions are injection molded at 290°C into plaques of 1.6 mm or 3.2 mm thickness according to Underwriter's Laboratories flame retardancy standard UL-94.

Flame retardancy is tested according to UL-94 in the vertical mode.

Example	PC 145 Resin [wt.-%]	IRGAFOS [wt.-%]	FR [wt.-%]	PTFE [wt.-%]	UL-94
1	99.57	0.08	0.1	0.25	V-0 (1.6 mm)
2	99.82	0.08	0.1	0	V-0 (3.2 mm)
Comp. Ex.	99.92	0.08	--	--	V-2 (1.6 mm)

Flame Retardant (FR) present in composition:

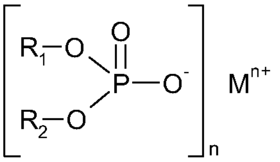


(Irgastab® NA 11)

Claims

1. A composition which comprises

a) At least one salt of an aromatic phosphoric acid ester of the formula



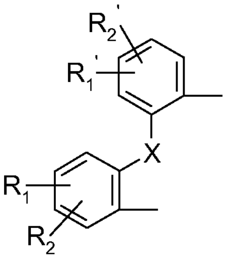
(I),

Wherein

One of R_1 and R_2 represents phenyl or phenyl substituted by one or two C_1-C_8alkyl; and the other one represents C_1-C_8alkyl; or

R_1 and R_2 both represent phenyl or phenyl substituted by one or two C_1-C_8alkyl; or

R_1 and R_2 together represent a group of the partial formula



(A),

Wherein

X represents C_1-C_4alkylene;

One of R_1 and R_2 represents hydrogen or C_1-C_8alkyl and the other one represents C_4-C_8alkyl; and

R_1' and R_2' are as defined as R_1 and R_2;

n represents 1 or 2; and

M^{n+} represents a cation that bears one positive charge if n represents 1; or

M^{n+} represents a cation that bears two positive charges if n represents 2; and

b) A polymer substrate comprising polycarbonate or polycarbonate blends.

2. A composition according to claim 1 which comprises as component a) at least one salt of an aromatic phosphoric acid ester (I), wherein

One of R_1 and R_2 represents phenyl or phenyl substituted by one or two C_1-C_4alkyl; and the other one represents C_1-C_4alkyl; or

R_1 and R_2 both represent phenyl or phenyl substituted by one or two C_1-C_4alkyl; or

R_1 and R_2 together represent the group (A), wherein

X represents C_1-C_4alkylene;

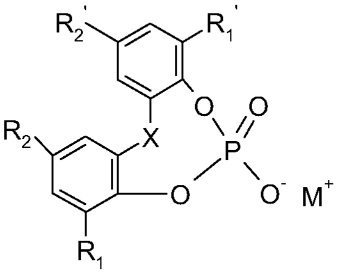
One of R_1 and R_2 represents hydrogen, methyl or tert-butyl and the other one represents tert-butyl; and

R_1' and R_2' are as defined as R_1 and R_2; and

n represents 1;

M^{n+} represents a cation that bears one positive charge.

3. A composition according to claim 1 which comprises as component a) at least one salt of an aromatic phosphoric acid ester of the formula



(I'),

Wherein

X represents C_1-C_4alkylene;

One of R_1 and R_2 represents hydrogen, methyl or tert-butyl and the other one represents tert-butyl; and

R_1' and R_2' are as defined as R_1 and R_2; and

M^{n+} represents a cation that bears one positive charge.

4. A composition according to claim 1 which comprises as component a) at least one salt of an aromatic phosphoric acid ester (I'), wherein

X represents methylene;

One of R_1 and R_2 represents methyl or tert-butyl and the other one represents tert-butyl; and

R_1' and R_2' are as defined as R_1 and R_2; and

M^{n+} represents a cation that bears one positive charge.

5. A composition according to claim 1 which comprises as component a) at least one salt of an aromatic phosphoric acid ester (I'), wherein

   X represents methylene;

   R_1 and R_2 and R_1' and R_2' represent tert-butyl; and

   M^{n+} represents sodium or potassium.

6. A composition according to claim 1 which additionally comprises as additional component anti-dripping agents.

7. A composition according to claim 1 which additionally comprises further additives selected from the group consisting of polymer stabilizers and additional flame-retardants.

8. A process for imparting flame retardancy to a polymer substrate comprising polycarbonates or polycarbonate blends, which process comprises adding to said polymer substrate at least one salt of an aromatic phosphoric acid ester of the formula (I) according to claim 1.

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2007/051315

A. CLASSIFICATION OF SUBJECT MATTER
INV. C08K5/523 C08L69/00 C08K5/521

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
C08K C08L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 342 696 A2 (TEIJIN CHEMICALS LTD [JP]) 23 November 1989 (1989-11-23) page 3, line 47 - page 4, line 13; claims 1,2; examples 2,11; compound II	1-8
X	EP 0 522 432 A2 (TEIJIN CHEMICALS LTD [JP]) 13 January 1993 (1993-01-13) page 4, lines 11-14; claims 1-5	1-8
X	DE 198 28 535 A1 (BAYER AG [DE]) 30 December 1999 (1999-12-30) page 9, lines 42-58; claims 1-15; example 7	1,2,6-8

☐ Further documents are listed in the continuation of Box C. ☒ See patent family annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance
"E" earlier document but published on or after the international filing date
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
"O" document referring to an oral disclosure, use, exhibition or other means
"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
"&" document member of the same patent family

Date of the actual completion of the international search 1 June 2007	Date of mailing of the international search report 12/06/2007
Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Authorized officer Sperry, Pascal

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/EP2007/051315

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