

Polycarbonate Having a Low Shear Thinning Behaviour

The present invention relates to a novel melt-polymerised polycarbonate that has a flow behaviour similar to that of a linear polycarbonate that has been produced by interface polymerisation, compositions containing this polycarbonate, as well as
5 moulded articles produced therefrom. The invention also relates to a process for the production of this polycarbonate.

Polycarbonate is produced industrially by means of the interface process or by
10 transesterification in the melt (melt polymerisation process). The melt polymerisation process is becoming increasingly important since it can be carried out without the use of phosgene or chlorinated solvents. Polycarbonates produced by the interface process that is mainly used at the present time are linear, and such
15 polymers do not contain any multifunctional structural units. This type of linear polycarbonate, which lacks the multifunctional structural units capable of undergoing branching, has only a slight non-Newtonian flow behaviour.

Polycarbonates produced using the melt polymerisation process contain multifunctional structural units, are highly branched, and have an excellent non-
20 Newtonian flow behaviour. The unavoidable production of multifunctional branching structural units during the polymerisation is known and is described for example in *Angewandte Chemie* 20, pp. 633-660, 1956. On account of their different flow properties it is difficult to use these two types of polycarbonates interchangeably in for example injection moulding processes or extrusion processes.

25

Typical melt-polymerised polycarbonate contains branching units that lead to an excellent, non-Newtonian flow behaviour that is usually characterised by the shear thinning ratio. The branching unfortunately also leads to an unacceptable yellow discolouration.

30

DE 42 38 123 A describes a process for the production of melt-polymerised polycarbonate having a low proportion of branching units, though no details of the flow behaviour of the resultant polycarbonate are given.

US 5 932 683 discloses a typical melt polymerisation process in which a typical branched polycarbonate is produced having a pronounced non-Newtonian flow behaviour. In addition special branching structural units that are necessary for the pronounced non-Newtonian flow behaviour are disclosed, as well as a special relationship between the reaction time and reaction temperature, a sufficient number of branchings being produced within these defined parameters so as to achieve the pronounced non-Newtonian flow behaviour while retaining the other good properties of the polycarbonate.

10 The object of the present invention is to provide a melt-polymerised polycarbonate having rheological properties that are comparable to those of an interface-polymerised polycarbonate, wherein these polycarbonates should have an excellent colour and colour stability.

15 This object is achieved by a melt-polymerised polycarbonate with a shear thinning ratio (y) that is defined by the limiting values of the following equation

$$(Q) \quad c + ax^b > y > -c + ax^b$$

20 - for all $y \geq 1$,

- wherein a , b and c are constants, and

- $c = 0.3$ to 0.1 , preferably 0.2 to 0.1 , and most preferably about 0.1 ,

- $b = 14.831 \pm 0.05$ to 0.02 , preferably ± 0.04 to 0.02 , and most preferably $\pm$ about 0.02 , and

25 - $a = 0.1262 \pm 0.005$ to 0.003 , preferably ± 0.004 to 0.003 , and most preferably $\pm$ about 0.003 , and

x is the relative viscosity of the polycarbonate.

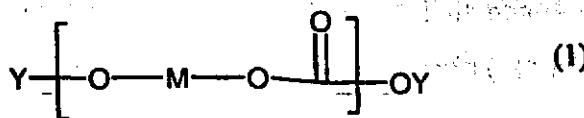
The shear thinning ratio serves to quantify the flow behaviour. The shear thinning ratio is defined as the ratio of the viscosity at a low shear rate to the viscosity at a high shear rate. According to the invention the shear thinning ratio is the ratio of the

- 3 -

viscosity at a shear rate of 50s^{-1} to the viscosity at a shear rate of $5,000\text{s}^{-1}$ measured at 280°C .

It was surprisingly found that the polycarbonate produced according to the invention
 5 by melt polymerisation (transesterification process) contains multifunctional structural units capable of branching but nevertheless has a slight non-Newtonian flow behaviour that is equivalent to that of linear polycarbonates produced by interface polymerisation. This is surprising since all previously known melt-polymerised polycarbonates exhibit a very much more pronounced non-Newtonian
 10 flow behaviour than linear polycarbonates. The polycarbonates according to the invention exhibit an excellent colour stability during injection moulding.

The melt-polymerised polycarbonates according to the invention correspond to the general formula (1)



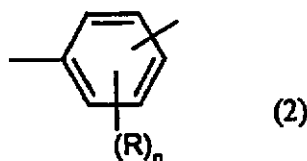
15

wherein the square brackets denote repeating structural units,

M may denote Ar or a multifunctional compound A, B, C as well as compound
 D,

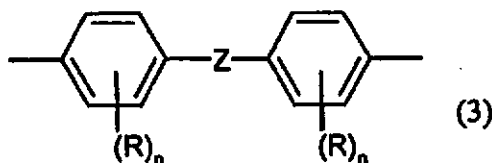
20

wherein Ar may be a compound that is represented by Formula (2)



or particularly preferably a compound that is represented by Formula (3)

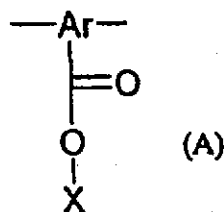
- 4 -



wherein

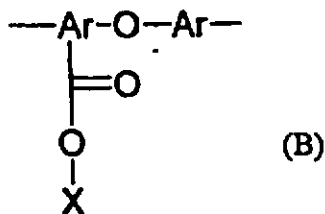
- Z denotes C₁-C₈ alkylidene or C₅-C₁₂ cycloalkylidene, S, SO₂ or a single bond,
 5 R denotes a substituted or unsubstituted phenyl, methyl, propyl, ethyl, butyl,
 Cl or Br, and n denotes 0, 1 or 2,

wherein the multifunctional compound A is a compound of the formula



- 10 and may be contained in an amount of less than 1,500 ppm, preferably less than
 1,000 ppm, most particularly preferably less than about 500 ppm in the
 polycarbonate,

wherein the multifunctional compound B is a compound of the formula

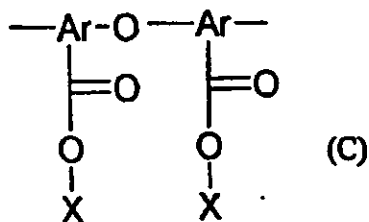


15

- and may be contained in an amount of less than 350 ppm, preferably less than
 250 ppm, most particularly preferably less than about 100 ppm in the polycarbonate,

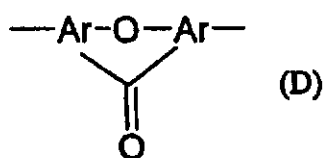
- 20 wherein the multifunctional compound C is a compound of the formula

- 5 -



and may be contained in an amount of less than 200 ppm, preferably less than 150 ppm, most particularly preferably less than about 100 ppm in the polycarbonate,

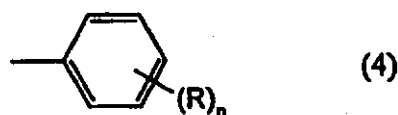
5 wherein compound D is a compound of the formula



and may be contained in an amount of less than 750 ppm, preferably less than 500 ppm, most particularly preferably less than about 300 ppm in the polycarbonate,

10 wherein

Y is H or a compound of the formula (4)



wherein

15

R may be identical or different and denotes H, C₁-C₂₀ alkyl, C₆H₅ or C(CH₃)₂C₆H₅, and

n may be 0, 1, 2 or 3,

20

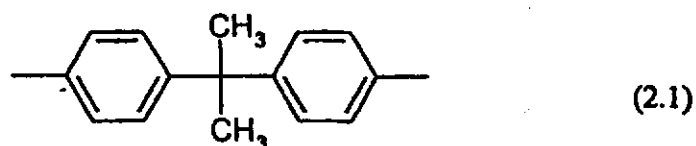
wherein X = Y or -(MOCOO)Y,

- 6 -

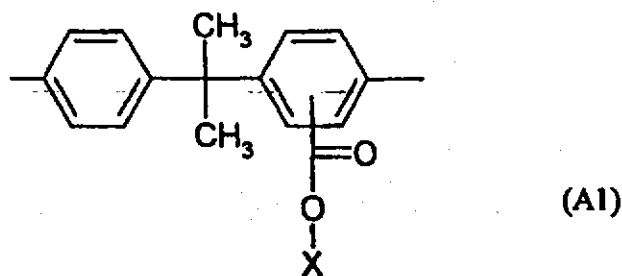
wherein M and Y have the meanings given above and these definitions also apply to the compounds mentioned hereinafter.

The polycarbonate according to the invention has a weight average molecular weight as determined by gel permeation chromatography of 5,000 to 80,000, preferably 10,000 to 60,000, and most particularly preferably 15,000 to 40,000.

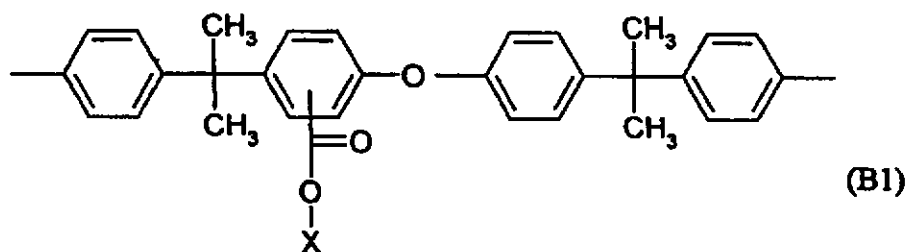
Ar preferably has the following meaning:



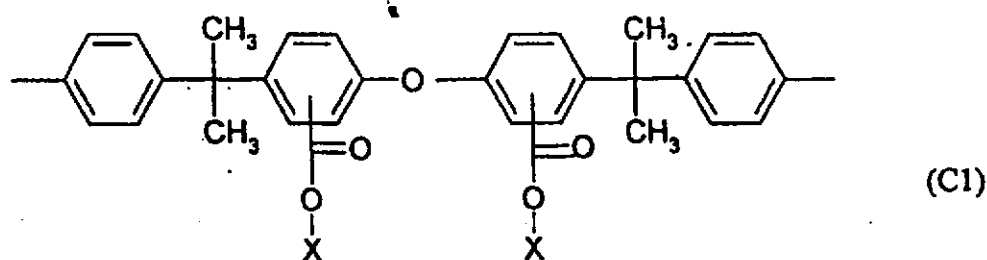
10 Preferably the multifunctional compound A corresponds to the following formula:



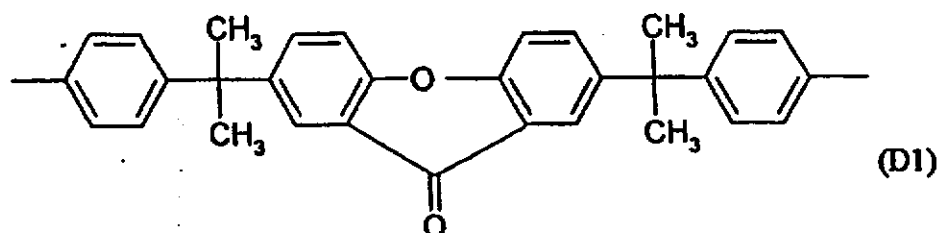
Preferably the multifunctional compound B corresponds to the following formula:



Preferably the multifunctional compound C corresponds to the following formula:



Preferably the multifunctional compound D corresponds to the following formula:



- The production of aromatic polycarbonates according to the melt transesterification process is known and is described for example in "Schnell", Chemistry and Physics of Polycarbonates, Polymer Reviews, Vol. 9, Interscience Publishers, New York, London, Sydney 1964, in D.C. Prevorsek, B.T. Debona and Y. Kesten, Corporate Research Center, Allied Chemical Corporation, Moristown, New Jersey 07960, "Synthesis of Poly(ester)carbonate Copolymers" in Journal of Polymer Science, Polymer Chemistry Edition, Vol. 19, 75-90 (1980), in D. Freitag, U. Grigo, P.R. Müller, N. Nouvertne, Bayer AG, "Polycarbonates" in Encyclopedia of Polymer Science and Engineering, Vol. 11, Second Edition, 1988, pp. 648-718 and finally in U. Grigo, K. Kircher and P.R. Müller "Polycarbonate" in Becker/Braun, Kunststoff-Handbuch, Vol. 3/1, Polycarbonates, Polyacetals, Polyesters, Cellulose esters, Carl Hanser Verlag Munich, Vienna, 1992, pp. 117-299.

The polycarbonate according to the invention, which may also be a polyester carbonate, is produced by the melt transesterification reaction of suitable diphenol and carbonic acid diaryl esters in the presence of a suitable catalyst. The polycarbonate may also be obtained by the condensation of carbonate oligomers containing hydroxy terminal groups or carbonate terminal groups, and suitable diphenols as well as carbonic acid diaryl esters.

Suitable carbonic acid diaryl esters in the context of the invention are di-C₆ to C₁₄ aryl esters, preferably the diesters of phenol or of alkyl-substituted phenols, i.e. diphenyl carbonate, dicresyl carbonate and di-4-tert.-butylphenyl carbonate. Diphenyl carbonate is particularly preferred.

5

Suitable carbonate oligomers are described by the above Formula (1) with molecular weights of 220 to 15,000.

Suitable di-C₆-C₁₄ aryl esters also include unsymmetrical diaryl esters that contain a mixture of aryl substituents. Most particularly preferred are phenylcresyl carbonate and 4-tert.-butylphenyl carbonate.

10

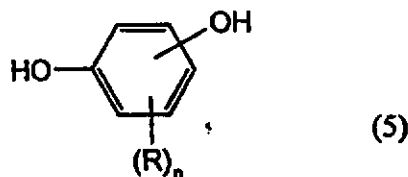
Suitable diaryl esters also include mixtures of more than one di-C₆-C₁₄ aryl ester. Most particularly preferred are mixtures of diphenyl carbonate, dicresyl carbonate and di-4-ter.-butylphenyl carbonate.

15

Referred to 1 mole of diphenol, the carbonic acid diaryl esters may be used in amounts of 1.00 to 1.30 mole, particularly preferably in amounts of 1.02 to 1.20 mole, and most particularly preferably in amounts of 1.05 to 1.15 mole.

20

Suitable dihydroxybenzene compounds in connection with the invention are those that correspond to Formula (5):



wherein

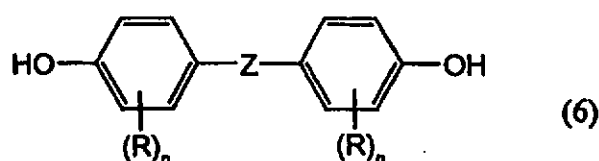
25

R denotes a substituted or unsubstituted phenyl, methyl, propyl, ethyl, butyl, Cl or Br, and n denotes 0, 1 or 2.

Most particularly preferred dihydroxybenzene compounds are 1,3-dihydroxybenzene, 1,4-dihydroxybenzene and 1,2-dihydroxybenzene.

Suitable diphenols in connection with the invention are those that correspond to

5 Formula (6):



wherein

10 Z denotes C₁-C₈ alkylidene or C₅-C₁₂ cycloalkylidene, S, SO₂ or a single bond,

R denotes a substituted or unsubstituted phenyl, methyl, propyl, ethyl, butyl, Cl or Br, and

n denotes 0, 1 or 2.

15

Preferred diphenols are 4,4'-dihydroxydiphenyl, 4,4'-dihydroxydiphenyl sulfide, 1,1-bis(4-hydroxyphenyl)cyclohexane, 1,2-bis(4-hydroxyphenyl)benzene, 1,3-bis(4-hydroxyphenyl)benzene, 1,4-bis(4-hydroxyphenyl)benzene, bis(4-hydroxyphenyl)methane, 2,2-bis(4-hydroxyphenyl)propane, 2,4-bis(4-hydroxyphenyl)-2-methylbutane, 2,2-bis(3-methyl-4-hydroxyphenyl)propane, 2,2-bis(3-chloro-4-hydroxyphenyl)-2-propane, bis(3,5-dimethyl-4-hydroxyphenyl)methane, 2,2-bis(3,5-dimethyl-4-hydroxyphenyl)propane, bis(3,5-dimethyl-4-hydroxyphenyl)sulfone, bis(4-hydroxyphenyl)sulfone, 1,2-bis[2-(4-hydroxyphenyl)isopropyl]benzene, 1,3-bis[2-(4-hydroxyphenyl)isopropyl]benzene, 1,4-bis[2-(4-hydroxyphenyl)isopropyl]benzene, 1,1-bis(4-hydroxyphenyl)-1-phenylethane, 2,4-bis(4-hydroxyphenyl)-2-methylbutane, 2,2-bis(3,5-dichloro-4-hydroxyphenyl)propane, 2,2-bis(3,5-dibromo-4-hydroxyphenyl)propane, and 1,1-bis(4-hydroxyphenyl)-3,3,5-trimethylcyclohexane.

20

25

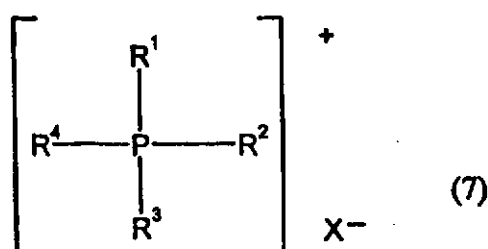
The most particularly preferred diphenols are 1,1-bis(4-hydroxyphenyl)-1-phenylethane, 2,2-bis(4-hydroxyphenyl)propane, 4,4'-dihydroxydiphenyl, 1,1-bis(4-hydroxyphenyl)-3,3,5-trimethylcyclohexane, 2,2-bis(3,5-dibromo-4-hydroxyphenyl)propane and 1,3-bis[2-(4-hydroxyphenyl)isopropyl]benzene.

5

Suitable diphenols also include mixtures of more than one diphenol, a copolycarbonate being formed if a mixture were used. The most particularly preferred mixtures are 1,3-bis[2-(4-hydroxyphenyl)isopropyl]benzene, 1,1-bis(4-hydroxyphenyl)-3,3,5-trimethylcyclohexane, 2,2-bis(4-hydroxyphenyl)propane,
10 4,4'-dihydroxydiphenyl and 2,2-bis(3,5-dibromo-4-hydroxyphenyl)propane.

In addition a branching agent may be added, such as compounds that contain three functional phenolic OH groups. This would obviously branch the polymer and would increase the non-Newtonian flow behaviour of the polymer. Suitable
15 branching agents include phloroglucinol, 3,3-bis(3-methyl-4-hydroxyphenyl)-2-oxo-2,3-dihydroindole, 4,6-dimethyl-2,4,6-tris(4-hydroxyphenyl)heptene-2, 4,6-dimethyl-2,4,6-tris(4-hydroxyphenyl)heptane, 1,3,5-tris(4-hydroxyphenyl)benzene, 1,1,1-tris(4-hydroxyphenyl)ethane, tris(4-hydroxyphenyl)phenylmethane, 2,2-bis-[4,4-bis(4-hydroxyphenyl)cyclohexyl]propane, 2,4-bis(4-hydroxyphenylisopropyl)-
20 phenol, 2,6-bis(2-hydroxy-5'-methylbenzyl)-4-methylphenol, 2-(4-hydroxyphenyl)-2-(2,4-dihydroxyphenyl)propane, hexakis(4-(4-hydroxyphenylisopropyl)phenyl)-orthoterephthalate, tetrakis(4-hydroxyphenyl)methane, tetrakis(4-(4-hydroxyphenylisopropyl)phenoxy)methane and 1,4-bis((4',4''-dihydroxytriphenyl)methyl)benzene, isatin biscreosol, pentaerythritol, 2,4-dihydroxybenzoic acid, trimesic acid and
25 cyanuric acid.

Suitable catalysts for the production of the polycarbonates according to the invention are for example those of the general formula (7)



wherein

5 R^1 , R^2 , R^3 and R^4 , which are identical or different, may denote C_1 to C_{18} alkyls, C_6 to C_{10} aryls or C_5 to C_6 cycloalkyls, and

X^- may denote an anion in which the corresponding acid-base pair $H^+ + X^- \leftrightarrow HX$ has a pK_b of <11 .

- 10 Preferred catalysts are tetraphenylphosphonium fluoride, tetraphenylphosphonium tetraphenyl borate and tetraphenylphosphonium phenolate. Most particularly preferred is tetraphenylphosphonium phenolate. Preferred amounts of phosphonium salt catalysts are 10^{-2} to 10^{-8} mole per mole of diphenol, the most particularly preferred amounts of catalysts being 10^{-4} to 10^{-6} mole per mole of diphenol. Further
- 15 co-catalysts may optionally also be used in addition to the phosphonium salt in order to increase the rate of the polymerisation. Such co-catalysts include salts of alkali metals and alkaline earth metals, such as hydroxides, alkoxides and aryl oxides of lithium, sodium and potassium, preferably hydroxide, alkoxide or aryl oxide salts of sodium. Most particularly preferred are sodium hydroxide and sodium phenolate.
- 20 The amounts of the co-catalyst may be in the range from 1 to 200 ppb, preferably 5 to 150 ppb, and most particularly preferably 10 to 125 ppb, in each case calculated as sodium.

The polycarbonates may be produced under reaction conditions that are carried out

25 in temperature stages from 150° to 400°C , the residence time in each stage may be 15 minutes to 5 hours, and the pressures are 1000 to 0.01 mbar.

The present invention also provides polycarbonate compositions that contain the polycarbonates according to the invention, as well as moulded articles containing such compositions.

- 5 The compositions according to the invention may contain, in addition to the polycarbonates or polyester carbonates, further polymeric constituents and conventional additives. Possible polymeric constituents are for example styrene/acrylic/nitrile terpolymers (SAN), acrylic/butadiene/styrene terpolymers (ABS), poly(methyl methacrylate) (PMMA), fluorinated polyolefins (PTFE),
10 polyphenylene sulfide (PPS), polyolefins such as polyethylene, polypropylene and ethylene/propylene rubbers, epoxy resins, polyesters such as polybutylene terephthalate (PBT), polyethylene terephthalate (PET), polycyclohexene dimethanol (PCT), copolyesters of ethylene glycol and cyclohexene dimethanol terephthalic acid in a ratio of the two constituents of 1:4 (PCTG), copolyesters of ethylene glycol
15 and cyclohexene dimethanol terephthalic acid in a ratio of the two constituents of 4:1 (PETG), as well as other polycarbonates produced by the interface process and also mixtures of the aforementioned components. Further additives include mould release agents, stabilisers, antioxidants, flame retardants, colouring agents and pigments, antistatics, nucleating agents, anti-trickle agents, as well as organic and
20 further inorganic fillers and reinforcing agents.

- The polycarbonate according to the invention may be contained in the composition according to the invention in an amount of preferably 5 to 95 wt.%, particularly preferably 10 to 90 wt.%, and most particularly preferably 20 to 80 wt.%, referred to
25 the weight of the composition. The further polymeric constituents of the composition according to the invention may be contained in an amount of preferably 1 to 60 wt.%, particularly preferably 1 to 40 wt.%, and most particularly preferably 2 to 30 wt.%, referred to the weight of the composition.

- 30 The compositions may contain up to 60 wt.%, preferably 10 to 40 wt.%, referred to the filled or reinforced moulding composition, of inorganic materials such as fillers and/or reinforcing agents. Flame retardants may be contained in the composition

according to the invention in an amount of up to 35 wt.%, preferably 10 to 25 wt.%, referred to the mass of the composition.

5 These and further constituents or additives that may possibly be contained in the compositions according to the invention, in addition to the polycarbonates/polyester carbonates, are illustrated hereinafter by means of examples.

10 These substances can be found in many publications, such as for example in the Additives for Plastics Handbook, John Murphy, 1999 and are commercially available.

1. Suitable antioxidants are for example:

15 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 that may be linear or branched in the side chain, for example 2,6-dinonyl-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.

25 1.2. Alkylthiomethylphenols, for example 2,4-dioctylthiomethyl-6-tert.-butylphenol, 2,4-dioctylthiomethyl-6-methylphenol, 2,4-dioctylthiomethyl-6-ethylphenol, 2,6-didodecylthiomethyl-4-nonylphenol.

30 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 and bis(3,5-di-tert.-butyl-4-hydroxyphenyl)adipate.

1.4. Tocopherols, for example α -tocopherol, β -tocopherol, γ -tocopherol, 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-hydroxy-phenol)disulfide.

1.6 Alkylidene bisphenols, 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-methylphenyl)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-dimethylbenzyl mercaptoacetate, tridecyl-4-hydroxy-3,5-di-tert.-butylbenzyl mercaptoacetate, tris(3,5-di-tert.-butyl-4-hydroxybenzyl)amine, bis(4-tert.-butyl-3-hydroxy-2,6-dimethylbenzyl)dithio-

terephthalate, bis(3,5-di-tert.-butyl-4-hydroxybenzyl)sulfide, isooctyl-3,5-di-tert.-butyl-4-hydroxybenzyl mercaptoacetate.

- 1.8. Hydroxybenzylated malonates, for example dioctadecyl-2,2-bis(3,5-di-tert.-butyl-2-hydroxybenzyl)malonate, dioctadecyl-2-(3-tert.-butyl-4-hydroxy-5-methylbenzyl)malonate, didodecylmercaptoethyl-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-hydroxyphenylpropioyl)-hexahydro-1,3,5-triazine, 1,3,5-tris(3,5-dicyclohexyl-4-hydroxybenzyl)isocyanurate.
- 1.11. Acylaminophenols, for example 4-hydroxylauranilide, 4-hydroxystearanilide, octyl-N-(3,5-di-tert.-butyl-4-hydroxyphenyl)carbamate.

- 1.12. Esters of β -(3,5-di-tert.-butyl-4-hydroxyphenyl)propionic acid with monohydric or polyhydric alcohols, for example 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.

- 1.13 Esters of β -(5-tert.-butyl-4-hydroxy-3-methylphenyl)propionic acid with
5 monohydric or polyhydric alcohols, for example 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,
10 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.

- 1.14 Esters of β -(3,5-dicyclohexyl-4-hydroxyphenyl)propionic acid with
monohydric or polyhydric alcohols, for example with methanol, ethanol, octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol,
15 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.

- 20 1.15. Esters of 3,5-di-tert.-butyl-4-hydroxyphenylacetic acid with monohydric or polyhydric alcohols, for example 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,
25 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.

- 1.16. Amides of β -(3,5-di-tert.-butyl-4-hydroxyphenyl)propionic acid, for example
N,N'-bis(3,5-di-tert.-butyl-4-hydroxyphenylpropionyl)hexamethylenediamide, N,N'-
30 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 from Uniroyal).

1.17 Ascorbic acid (vitamin C)

5

1.18. Amine antioxidants, for example N,N'-diisopropyl-p-phenylenediamine, N,N'-di-sec-butyl-p-phenylenediamine, N,N'-bis(1,4-dimethylpentyl)-p-phenylenediamine, N,N'-bis(1-ethyl-3-methylpentyl)-p-phenylenediamine, N,N'-bis(1-methylheptyl)-p-phenylenediamine, N,N'-dicyclohexyl-p-phenylenediamine, N,N'-diphenyl-p-phenylenediamine, N,N'-bis(2-naphthyl)-p-phenylenediamine, N-isopropyl-N'-phenyl-p-phenylenediamine, N-(1,3-dimethylbutyl)-N'-phenyl-p-phenylenediamine, N-(1-methylheptyl)-N'-phenyl-p-phenylenediamine, N-cyclohexyl-N'-phenyl-p-phenylenediamine, 4-(p-toluenesulfamoyl)diphenylamine, N,N'-dimethyl-N,N'-di-sec-butyl-p-phenylenediamine, diphenylamine, N-allyl-
 10 diphenylamine, 4-isopropoxydiphenylamine, N-phenyl-1-naphthylamine, N-(4-tert.-octylphenyl)-1-naphthylamine, N-phenyl-2-naphthylamine, octylated diphenylamine, for example p,p'-di-tert.-octyldiphenylamine, 4-n-butylaminophenol, 4-n-butyrylaminophenol, 4-nonanoylaminophenol, 4-dodecanoylaminophenol, 4-octadecanoylaminophenol, bis(4-methoxyphenol)amine, 2,6-di-tert.-butyl-4-
 20 dimethylaminomethylphenol, 2,4'-diaminodiphenylmethane, 4,4'-diaminodiphenylmethane, N,N,N',N'-tetramethyl-4,4'-diaminodiphenylmethane, 1,2-bis[(2-methylphenyl)amino]ethane, 1,2-bis(phenylamino)propane, (o-tolyl)biguanide, bis[4-(1',3'-dimethylbutyl)phenyl]amine, tert.-octylated N-phenyl-1-naphthylamine, a mixture of monoalkylated and dialkylated tert.-butyl/tert.-octyldiphenylamines, a
 25 mixture of monoalkylated and dialkylated nonyldiphenylamines, a mixture of monoalkylated and dialkylated dodecyldiphenylamines, a mixture of monoalkylated and dialkylated isopropyl/isohexyldiphenylamines, a mixture of monoalkylated and dialkylated tert.-butyldiphenylamines, 2,3-dihydro-3,3-dimethyl-4H-1,4-benzothiazine, phenothiazine, a mixture of monoalkylated and dialkylated tert.-butyl/tert.-
 30 octyl phenothiazines, a mixture of monoalkylated and dialkylated tert.-octyl phenothiazines, N-allylphenothiazine, N,N,N',N'-tetraphenyl-1,4-diaminobut-2-ene, N,N-bis(2,2,6,6-tetramethylpiperid-4-yl)-hexamethylenediamine, bis(2,2,6,6-tetra-

methylpiperid-4-yl)sebacate, 2,2,6,6-tetramethylpiperidin-4-one, 2,2,6,6-tetramethylpiperidin-4-ol. Individual compounds or mixtures thereof may be used.

1.19. Suitable thiosynergists are for example dilauryl thiodipropionate and/or distearyl thiodipropionate.

2. UV-absorbers and light stabilisers may be used in the compositions according to the invention in amounts of 0.1 to 15 wt.%, preferably 3 to 8 wt.%, referred to the mass of the composition. Suitable UV-absorbers and light stabilisers are for example:

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-tetramethylbutylphenyl)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-octyloxycarbonyl)ethyl)phenyl)-5-chlorobenzotriazole, 2-(3'-tert.-butyl-5'-[2-(2-ethylhexyloxy)carbonyl]ethyl)-2'-hydroxyphenyl)-5-chlorobenzotriazole, 2-(3'-tert.-butyl-2'-hydroxy-5'-(2-methoxycarbonyl)ethyl)phenyl)-5-chlorobenzotriazole, 2-(3'-tert.-butyl-2'-hydroxy-5'-(2-methoxycarbonyl)ethyl)phenyl)benzotriazole, 2-(3'-tert.-butyl-2'-hydroxy-5'-(2-octyloxycarbonyl)ethyl)phenyl)benzotriazole, 2-(3'-tert.-butyl-5'-[2-(2-ethylhexyloxy)carbonyl]ethyl)-2'-hydroxyphenyl)benzotriazole, 2-(3'-dodecyl-2'-hydroxy-5'-methylphenyl)benzotriazole, 2-(3'-tert.-butyl-2'-hydroxy-5'-(2-isooctyloxycarbonyl)ethyl)phenyl)benzotriazole, 2,2'-methylenebis[4-(1,1,3,3-tetramethylbutyl)-6-benzotriazol-2-ylphenol]; the transesterification product of 2-[3'-tert.-butyl-5'-(2-methoxycarbonyl)ethyl)-2'-hydroxyphenyl]-2H-benzotriazole with polyethylene glycol 300; [R-CH₂CH₂-COO-CH₂CH₂]₂, wherein R = 3'-tert.-butyl-4'-hydroxy-5'-2H-benzo-

triazol-2-yl-phenyl, 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.

- 5 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.

- 10 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.

15

- 15 2.4. Acrylates, for example ethyl- α -cyan- β,β -diphenyl acrylate, isooctyl- α -cyan- β,β -diphenyl acrylate, methyl- α -carbomethoxy cinnamate, methyl- α -cyan- β -methyl-p-methoxy cinnamate, butyl- α -cyan- β -methyl-p-methoxy cinnamate, methyl- α -carbomethoxy-p-methoxy cinnamate and N-(β -carbomethoxy- β -cyanovinyl)-2-methylindoline.

20

- 25 2.5. Nickel compounds, for example nickel complexes of 2,2'-thiobis[4-(1,1,3,3-tetramethylbutyl)phenol], such as the 1:1 complex or 1:2 complex, with or without additional ligands, such as n-butylamine, triethanolamine or N-cyclohexyldiethanolamine, nickel dibutyl dithiocarbamate, nickel salts of monoalkyl esters, for example of the methyl or ethyl ester, of 4-hydroxy-3,5-di-tert.-butylbenzylphosphonic acid, nickel complexes of ketoximes, for example of 2-hydroxy-4-methylphenylundecyl ketoxime, nickel complexes of 1-phenyl-4-lauroyl-5-hydroxypyrazole, with or without additional ligands.

30

- 2.6 Sterically hindered amines, for example bis(2,2,6,6-tetramethyl-4-piperidyl)sebacate, bis(2,2,6,6-tetramethyl-4-piperidyl)succinate, bis(1,2,2,6,6-

pentamethyl-4-piperidyl)sebacate, bis(1-octyloxy-2,2,6,6-tetramethyl-4-piperidyl)sebacate, bis(1,2,2,6,6-pentamethyl-4-piperidyl), n-butyl-3,5-di-tert.-butyl-4-hydroxybenzyl malonate, the condensate of 1-(2-hydroxyethyl)-2,2,6,6-tetramethyl-4-hydroxypiperidine and succinic acid, linear or cyclic condensates of

5 N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine and 4-tert.-octylamino-2,6-dichloro-1,3,5-triazine, tris(2,2,6,6-tetramethyl-4-piperidyl)-nitrilotriacetate, tetrakis(2,2,6,6-tetramethyl-4-piperidyl)-1,2,3,4-butanetetracarboxylate, 1,1'-(1,2-ethanediyl)bis(3,3,5,5-tetramethylpiperazinone), 4-benzoyl-2,2,6,6-tetramethylpiperidine, 4-stearyloxy-2,2,6,6-tetramethylpiperidine,

10 bis(1,2,2,6,6-pentamethylpiperidyl)-2-n-butyl-2-(2-hydroxy-3,5-di-tert.-butylbenzyl)malonate, 3-n-octyl-7,7,9,9-tetramethyl-1,3,8-triazaspiro[4.5]decane-2,4-dione, bis(1-octyloxy-2,2,6,6-tetramethylpiperidyl)sebacate, bis(1-octyloxy-2,2,6,6-tetramethylpiperidyl)succinate, linear or cyclic condensates of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine and 4-morpholino-2,6-dichloro-

15 1,3,5-triazine, the condensate of 2-chloro-4,6-bis(4-n-butylamino-2,2,6,6-tetramethylpiperidyl)-1,3,5-triazine and 1,2-bis(3-aminopropylamino)ethane, the condensate of 2-chloro-4,6-bis(4-n-butylamino-1,2,2,6,6-pentamethylpiperidyl)-1,3,5-triazine and 1,2-bis(3-aminopropylamino)ethane, 8-acetyl-3-dodecyl-7,7,9,9-tetramethyl-1,3,8-triazaspiro[4.5]decane-2,4-dione, 3-dodecyl-1-(2,2,6,6-tetra-

20 methyl-4-piperidyl)pyrrolidine-2,5-dione, 3-dodecyl-1-(1,2,2,6,6-pentamethyl-4-piperidyl)pyrrolidine-2,5-dione, a mixture of 4-hexadecyloxy- and 4-stearyloxy-2,2,6,6-tetramethylpiperidine, a condensation product of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine and 4-cyclohexylamino-2,6-dichloro-1,3,5-triazine, a condensation product of 1,2-bis(3-aminopropyl-

25 amino)ethane and 2,4,6-trichloro-1,3,5-triazine as well as 4-butylamino-2,2,6,6-tetramethylpiperidine (CAS Reg. No. [136504-96-6]); N-(2,2,6,6-tetramethyl-4-piperidyl)-n-dodecylsuccinimide, N-(1,2,2,6,6-pentamethyl-4-piperidyl)-n-dodecylsuccinimide, 2-undecyl-7,7,9,9-tetramethyl-1-oxa-3,8-diaza-4-oxospiro[4.5]decane, a reaction product of 7,7,9,9-tetramethyl-2-cycloundecyl-1-oxa-3,8-diaza-4-

30 oxospiro[4.5]decane and epichlorohydrin, 1,1-bis(1,2,2,6,6-pentamethyl-4-piperidyl-oxocarbonyl)-2-(4-methoxyphenyl)ethene, N,N'-bis(formyl)-N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine, diesters of 4-methoxymethylene-

malonic acid with 1,2,2,6,6-pentamethyl-4-hydroxypiperidine, poly[methylpropyl-3-oxy-4-(2,2,6,6-tetramethyl-4-piperidyl)]siloxane, reaction product of maleic anhydride/ α -olefin copolymer with 2,2,6,6-tetramethyl-4-aminopiperidine or 1,2,2,6,6-pentamethyl-4-aminopiperidine.

5

2.7. Oxamides, 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'-ethoxyanilide, N,N'-bis(3-dimethylaminopropyl)oxamide, 2-ethoxy-5-tert.-butyl-2'-ethoxyanilide 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.

10

2.8. 2-(2-hydroxyphenyl)-1,3,5-triazines, 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.

30

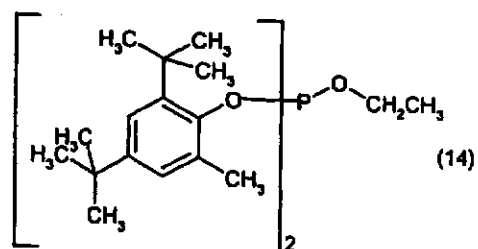
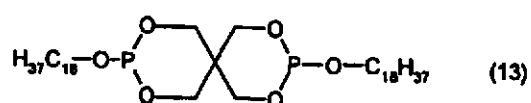
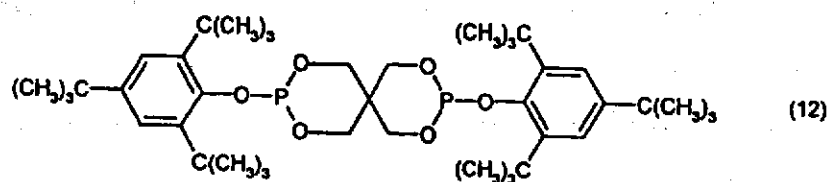
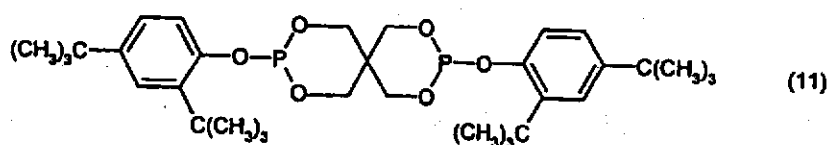
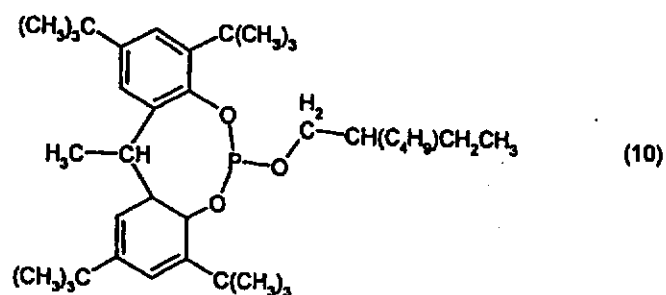
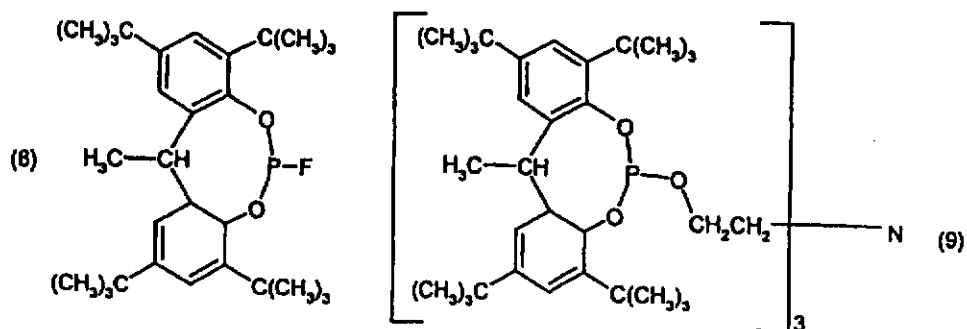
Individual compounds or mixtures thereof may be used.

3. Suitable metal deactivators are for example N,N'-diphenyloxamide, N-salicylal-N'-salicyloylhydrazine, 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, 5 sebacoylbisphenyl hydrazide, N,N'-diacetyl adipoyl dihydrazide, N,N'-bis(salicyloyl)oxalyl dihydrazide, N,N'-bis(salicyloyl)thiopropionyl dihydrazide. Individual compounds or mixtures thereof may be used.

4. Suitable phosphites and phosphonites that may be used as heat stabilisers include for example triphenyl phosphite, diphenylalkyl phosphites, phenyldialkyl phosphites, tri(nonylphenyl)phosphite, trilauryl phosphite, trioctadecyl phosphite, distearyl pentaerythritol diphosphite, tris(2,4-di-tert.-butylphenyl)phosphite, diisodecyl pentaerythritol diphosphite, bis(2,4-di-tert.-butylphenyl) pentaerythritol diphosphite, bis(2,6-di-tert.-butyl-4-methylphenyl) pentaerythritol diphosphite, 15 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-dioxaphosphocine, 6-fluoro-2,4,8,10-tetra-tert.-butyl-12-methyldibenz-[d,g]-1,3,2-dioxaphosphocine, 20 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-methyldibenz[d,g]-1,3,2-dioxaphosphocine, 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-2-25 (2,4,6-tri-tert.-butylphenoxy)-1,3,2-dioxaphosphirane. Individual compounds or mixtures thereof may be used.

Particularly preferred are tris(2,4-di-tert.-butylphenyl)phosphite (Irgafos® 168, Ciba-Geigy), triphenyl phosphine, or (2,4,6-tri-tert.-butylphenyl)-(2-butyl-2-ethylpropane-30 1,3-diyl)phosphite (Ultrinox 641®, GE Specialty Chemicals).

Examples of suitable phosphites and phosphonite are also:



5. Suitable peroxide traps are for example esters of β -thiodipropionic acid, for example the lauryl, stearyl, myristyl or tridecyl esters, mercaptobenzimidazole or the zinc salt of 2-mercaptobenzimidazole, zincdibutyl dithiocarbamate, dioctadecyl disulfide, pentaerythritol tetrakis(dodecylmercapto)propionate. Individual compounds or mixtures thereof may be used.

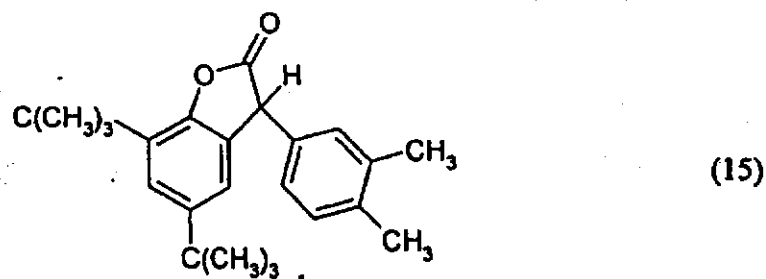
6. Suitable basic co-stabilisers are for example melamine, poly(vinylpyrrolidone), dicyandiamide, triallyl cyanurate, urea derivatives, hydrazine derivatives, amines, polyamides, polyurethanes, alkali metal salts and alkaline earth metal salts of higher fatty acids, for example calcium stearate, zinc stearate, magnesium behenate, magnesium stearate, sodium ricinoleate and potassium palmitate, antimony pyrocatecholate or zinc pyrocatecholate. Individual compounds or mixtures thereof may be used.

7. Suitable nucleating agents are for example inorganic substances such as talcum, metal oxides such as titanium dioxide or magnesium oxide, phosphates, carbonates or sulfates, preferably of alkaline earth metals; organic compounds such as monocarboxylic acids or polycarboxylic acids and their salts, e.g. 4-tert.-butylbenzoic acid, adipic acid, diphenylacetic acid, sodium succinate or sodium benzoate; polymeric compounds such as ionic copolymers (ionomers). Particularly preferred are 1,3:2,4-bis(3',4'-dimethylbenzylidene)sorbitol, 1,3:2,4-di(paramethyl-dibenzylidene)sorbitol and 1,3:2,4-di(benzylidene)sorbitol. Individual compounds or mixtures thereof may be used.

8. Suitable fillers and reinforcing agents are for example calcium carbonate, silicates, glass fibres, glass balls, asbestos, talcum, kaolin, mica, barium sulfate, metal oxides and hydroxides, carbon black, graphite, wollastonite, sawdust and flour or fibres of other natural products, synthetic fibres. Individual compounds or mixtures thereof may be used.

9. Other suitable additives are for example plasticisers, lubricants, emulsifiers, pigments, viscosity modifiers, catalysts, flow-control agents, optical brighteners, flame retardants, antistatic agents and blowing agents.

- 5 10. Suitable benzofuranonones and indolinones are for example those that are disclosed in US 4,325,863; US 4,338,244; US 5,175,312; US 5,216,052; US 5,252,643; DE-A-43 16 611; DE-A-43 16 622; DE-A-43 16 876; EP-A-0 589 839 or EP-A-0 591 102, 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-
10 [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, lactone antioxidants
15 such as



These compounds act for example as antioxidants. Individual compounds or mixtures thereof may be used.

- 20 11. Suitable fluorescing plasticisers are listed in "Plastics Additives Handbook", Eds. R. Gächter and H. Müller, Hanser Verlag, 3rd Edition, 1990, pp. 775-789.

12. Suitable mould release agents are esters of aliphatic acids and alcohols, for example pentaerythritol tetrastearate and glycerol monostearate, which are used
25 alone or as a mixture, preferably in an amount of 0.02 to 1 wt.% referred to the mass of the composition.

13. Suitable flame-inhibiting additives are phosphate esters, i.e. triphenyl phosphate, resorcinol diphosphoric acid esters, bromine-containing compounds such as brominated phosphoric acid esters, brominated oligocarbonates and polycarbonates, as well as salts such as $C_4F_9SO_3 Na^+$.

5

14. Suitable impact modifiers are butadiene rubber with grafted-on styrene-acrylonitrile or methyl methacrylate, ethylene-propylene rubbers with grafted-on maleic anhydride, ethyl acrylate and butyl acrylate rubbers with grafted-on methyl methacrylate or styrene-acrylonitrile, interpenetrating siloxane and acrylate networks with grafted-on methyl methacrylate or styrene-acrylonitrile.

10

15. Suitable polymers are SAN, ABS, PMMA, PTFE, PSU, PPS, polyolefins such as polyethylene, polypropylene and ethylene-propylene rubbers, epoxy resins, polyesters such as PBT, PET, PCT, PCTG and PETG as well as other polycarbonates produced in the interface process.

15

16. Suitable antistatic agents are sulfonate salts, for example tetraethylammonium salts of $C_{12}H_{25}SO_3^-$ or $C_8F_{17}SO_3^-$.

20 17. Suitable colouring agents are pigments as well as organic and inorganic dyes.

18. Compounds that contain epoxy groups, such as 3,4-epoxycyclohexylmethyl-3,4-epoxycyclohexylcarboxylate, copolymers of glycidyl methacrylate and epoxysilanes.

25

19. Compounds that contain anhydride groups, such as maleic anhydride, succinic anhydride, benzoacetic anhydride and phthalic anhydride.

The compounds of groups 18 and 19 act as melt stabilisers. They may be used individually or as mixtures.

30

The compositions (moulding compounds) according to the invention are produced by mixing the respective constituents in a known manner and melt compounding and

- 27 -

melt extruding the resultant mixtures at temperatures of about 200°C and for example at temperatures of 200°C to 300°C in conventional equipment such as internal kneaders, extruders and double-shaft screw extruders. The mixing of the individual constituents may take place successively as well as simultaneously, and
5 more specifically at about 20°C (room temperature) as well as at higher temperatures.

The moulding compositions according to the invention may be used to produce all types of moulded articles. These articles may be produced by injection moulding,
10 extrusion and blow moulding processes. A further type of processing is the production of moulded articles by thermoforming of previously produced sheets or films.

Examples of the moulded articles according to the invention include profiled
15 sections; sheets; all types of housing parts, e.g. for domestic appliances such as juice presses, coffee-making machines, mixers; for office equipment such as monitors, printers, copiers; for sheets, tubes, pipes, electrical installation ducting, windows, doors and profiled sections for the building and construction sector, interior fittings and external applications; in the electrical technology field, such as for switches and
20 plugs. In addition the moulded articles according to the invention may be used for internal fittings and structural parts of tracked vehicles, boats, aircraft, buses and other vehicles, as well as for vehicle body parts.

The moulded articles according to the invention may be transparent or opaque.
25 Further moulded articles include in particular optical and magneto-optical data storage media such as minidisks, compact discs (CDs) or digital versatile discs (DVDs), packagings for foodstuffs and beverages, optical lenses and prisms, lenses for illumination purposes, vehicle headlamp lenses, glazing for building and construction vehicles and road vehicles, other types of transparent panels, for
30 example for greenhouses, so-called reinforced sandwich panels, or hollow cavity plates.

The following examples serve to illustrate the invention in more detail.

Examples

Examples 1 to 8 and comparison examples 1 to 6

- 5 The polymers of Examples 1 to 8 were produced by melt polymerisation of diphenyl carbonate and bisphenol A using tetraphenylphosphonium phenolate as catalyst, sodium phenolate being used in addition in Examples 7 and 8. The monomer and catalyst amounts are shown in Table 1. The reaction conditions are given in Table 2. The relative viscosity of the solution, the yellowing index (YI), the melt viscosity, 10 the shear thinning ratio and the amounts of the multifunctional compounds are given in Table 3.

- The comparison examples 1, 2, 3 and 4 involve linear polycarbonates that were produced according to a conventional interface polymerisation process. The 15 polycarbonates are marketed under the trade names Makrolon® 2808, 2408, KU1-1266 and 3108 by Bayer AG. The polymers of comparison examples 5 and 6 are branched polycarbonates that were produced by the interface polymerisation process. Corresponding products are marketed by Bayer AG under the trade names Makrolon® 1243 and 1239. The relative viscosity was measured in a 0.5% solution 20 of methylene chloride at 20°C.

- The yellowing index (YI) was determined on 4 mm thick injection moulded samples in accordance with CIELAB methods. The injection moulding was carried out at 270°C and 330°C. 25

- The melt viscosity was determined at 280°C using the test method according to ISO 11443. The non-Newtonian flow behaviour was quantified by calculating the shear thinning ratio. The shear thinning ratio was calculated as the melt viscosity at a shear rate of 50 s^{-1} , divided by the melt viscosity at a shear rate of $5,000 \text{ s}^{-1}$. 30

The amount of the multifunctional compounds was determined by separating the monomeric compounds using HPLC (high pressure liquid chromatography) after

complete hydrolysis of the polycarbonate. These compounds were characterised by magnetic resonance spectroscopy techniques.

Table 1

5

Example	1	2	3	4	5	6	7	8
DPC, mole	1.234	1.178	1.173	1.198	1.168	1.187	1.190	1.180
BPA, mole	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
Sodium phenolate (ppb), calculated as sodium	0.0	0.0	0.0	0.0	0.0	0.0	125	100
Tetraphenyl- phosphonium phenolate, mole	0.00004	0.00004	0.00004	0.00004	0.00004	0.00004	0.00004	0.00004

DPC: diphenyl carbonate; BPA: bisphenol A

Table 2

Reaction conditions for Examples 1-8	Time (min.)	Temperature (°C)	Pressure (mbar)
Precondensation	45	190	1000
Reactor I	20	190	200
Reactor II	20	230	80
Reactor III	10	250	50
Reactor IV	10	270	25
Reaction conditions for Example 1			
Evaporator I	45	275	5.3
Evaporator II	130	312	0.55
Reaction conditions for Example 2			
Evaporator I	45	275	7.3
Evaporator II	130	312	1.35
Reaction conditions for Example 3			
Evaporator I	45	275	7.3
Evaporator II	130	312	1.00
Reaction conditions for Example 4			
Evaporator I	45	275	7.3
Evaporator II	130	312	1.85
Reaction conditions for Example 5			
Evaporator I	45	275	7.3
Evaporator II	130	312	0.98
Reaction conditions for Example 6			
Evaporator I	45	275	5.5
Evaporator II	130	312	0.50
Reaction conditions for Example 7			
Evaporator I	45	275	7.0
Evaporator II	130	310	1.85
Reaction conditions for Example 8			
Evaporator I	45	275	7.4
Evaporator II	130	310	1.40

Table 3

Example	V1	V2	V3	V4	V5	V6	1	2	3	4	5	6	7	8
Relative viscosity	1.294	1.259	1.195	1.322	1.287	1.318	1,201	1.253	1.277	1,218	1.287	1.283	1.249	1.292
YI (moulding at 270°C)	2.0	1.8	1.8				3.7	3.0	2.6	2.6	2.6	2.8	3.5	3.7
YI (moulding at 330°C)	2.4	2.0	1.9				3.7	2.7	2.5	2.6	2.6	2.8		3.8
Melt viscosity at 50 s ⁻¹ and 280°C	1002	593	129	1685	1272	2075	139	447	770	218	904	782	438	1071
Melt viscosity at 5,000 s ⁻¹ and 280°C	186	148	74	214	167	205	74	130	160	94	170	159	117	188
Shear thinning Ratio	5.4	4.0	1.7	7.9	7.6	10.1	1.9	3.4	4.8	2.3	5.3	4.9	3.7	5.7
A	ppm	< 5	< 5	< 5	< 5	< 5	253	272	226	268	256	231	1387	1475
B	ppm	< 5	< 5	< 5	< 5	< 5	26	16	6	16	21	10	289	337
C	ppm	< 5	< 5	< 5	< 5	< 5	< 5	57	< 5	< 5	59	53	197	178
D	ppm	< 5	< 5	< 5	< 5	< 5	157	117	138	188	175	144	720	692

Although these new melt-polymerised polycarbonates contain multifunctional structural units, they exhibit the same flow behaviour as characterised by the shear thinning ratio, as the interface-polymerised linear polycarbonates of the same relative viscosity. The polycarbonates are also characterised by a surprisingly good
5 colour stability during injection moulding. In comparison, the branched polycarbonates 5 and 6 exhibit a high degree of non-Newtonian flow behaviour, which is typical of a branched polymer. The comparison examples 5 and 6 contain 3,000 ppm of branching agent.

Patent Claims

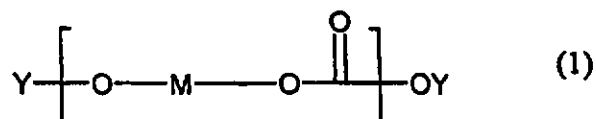
1. Melt-polymerised polycarbonate with a shear thinning ratio (y) that is defined by the limiting values of the following equation

$$(Q) \quad c + ax^b > y > -c + ax^b$$

- for all $y \geq 1$, calculated as the melt viscosity according to ISO 11443 at a shear rate of 50 s^{-1} divided by the melt viscosity at a shear rate of 5000 s^{-1} .
- wherein a , b and c are constants, and
- $c = 0.3$ to 0.1 ,
- $b = 14.831 \pm 0.05$
- $a = 0.1262 \pm 0.005$, and

x is the relative viscosity of the polycarbonate, measured in a 0.5% solution of methylene chloride at 20°C .

2. Melt-polymerised polycarbonate according to claim 1, wherein $c = 0.2$ to 0.1 .
3. Melt-polymerised polycarbonate according to claim 1 or 2, wherein $b = 14.831 \pm 0.04$.
4. Melt-polymerised polycarbonate according to one of claims 1 to 3, wherein $a = 0.1262 \pm 0.004$.
5. Melt-polymerised polycarbonate of the general formula (1)



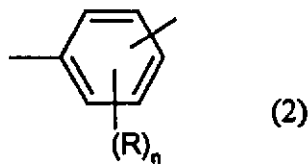
containing multifunctional structural units capable of branching

wherein in formula (1) the square brackets denote repeating structural units,

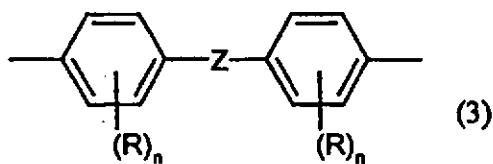
M may denote Ar or a multifunctional compound A, B, C as well as compound D,

wherein

Ar may be a compound that is represented by Formula (2)



or a compound that is represented by Formula (3)



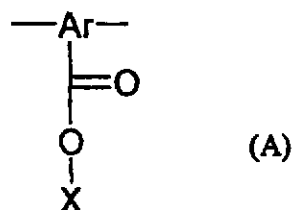
wherein

Z denotes C₁-C₈ alkylidene or C₅-C₁₂ cycloalkylidene, S, SO₂ or a single bond,

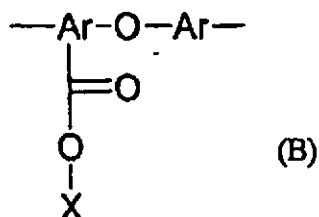
R denotes a substituted or unsubstituted phenyl, methyl, propyl, ethyl, butyl, Cl or Br, and

n denotes 0, 1 or 2,

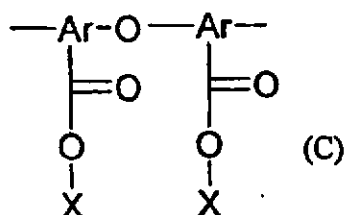
wherein the multifunctional compound A is a compound of the formula



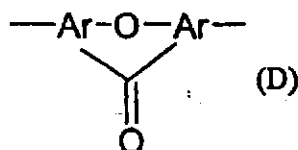
and is contained in amounts of <1,500 ppm, wherein the multifunctional compound B is a compound of the formula



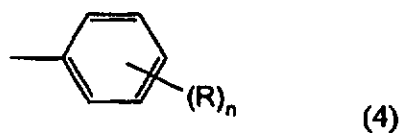
and is contained in amounts of <350 ppm, wherein the multifunctional compound C is a compound of the formula



and is contained in amounts of <200 ppm, wherein compound D is a compound of the formula



and is contained in amounts of <750 ppm, wherein Y is H or a compound of the formula (4)

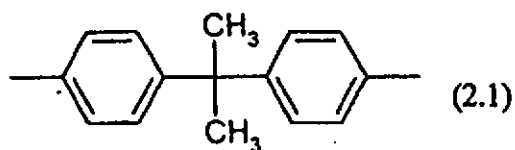


wherein

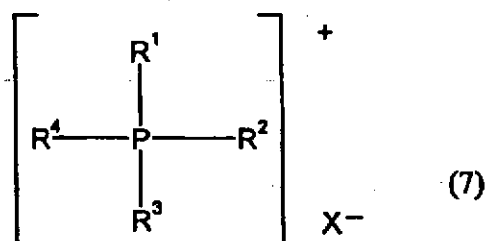
R is H, C₁-C₂₀ alkyl, C₆H₅ or C(CH₃)₂C₆H₅, and may be identical or different,

X is Y or -(MOCOO)Y, and M and Y have the meanings given above.

6. Melt-polymerised polycarbonate according to claim 5, wherein Ar is a compound of the formula



7. Composition containing a melt-polymerised polycarbonate according to one of claims 1 to 6.
8. Moulded article containing a composition according to claim 7.
9. Melt polymerisation process for the production of polycarbonates, in which diphenols and/or carbonate oligomers with hydroxy terminal groups or carbonate terminal groups are produced in the presence of a catalyst at a temperature of 150° to 400°C and a pressure of 1,000 mbar to 0.01 mbar, and as catalyst a compound of the formula (7)



is used,

wherein

R^1 , R^2 , R^3 and R^4 , which are identical or different, may denote C_1 to C_{18} alkyls, C_6 to C_{10} aryl compounds or C_5 to C_6 cycloalkyl compounds, and

X^- may denote an anion, in which the corresponding acid-base pair $H^+ + X^- \leftrightarrow HX$ has a pK_b of <11 .

10. Melt polymerisation process according to claim 9, wherein the catalyst is a mixture of tetraphenylphosphonium phenolate and 1-200 ppm, referred to sodium, of sodium phenolate.