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(54) **Title:** POLYSILOXANE-POLYCARBONATE BLOCK COCONDENSATES

(57) **Abstract:** The present invention relates to polysiloxane-polycarbonate block cocondensates comprising particular rearrange-  
ment structures and having improved rheological properties and melt stability as well as to mouldings and extrudates made from  
these polysiloxane-polycarbonate block cocondensates.



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**Polysiloxane-polycarbonate block cocondensates**

The present invention relates to polysiloxane-polycarbonate block cocondensates comprising particular rearrangement structures and having improved rheological properties and melt stability as well as to mouldings and extrudates made from these polysiloxane-polycarbonate block cocondensates.

It is known that polysiloxane-polycarbonate block cocondensates have good properties with regard to low-temperature impact strength or low-temperature notched impact strength, chemical resistance and outdoor weathering resistance, and to ageing properties and flame retardancy. In terms of these properties, they are in some cases superior to the conventional polycarbonates (homopolycarbonate based on bisphenol A).

The industrial preparation of these cocondensates proceeds from the monomers, usually via the interfacial process with phosgene. Also known is the preparation of these siloxane cocondensates via the melt transesterification process using diphenyl carbonate. However, these processes have the disadvantage that the industrial plants used therefor are used for preparation of standard polycarbonate and therefore have a high plant size. The preparation of specific block cocondensates in these plants is often economically unviable because of the smaller volume of these products. Moreover, the feedstocks required for preparation of the cocondensates, for example polydimethylsiloxanes, impair the plant, since they can lead to soiling of the plant or of the solvent circuits. In addition, toxic feedstocks such as phosgene are required for the preparation, or these processes entail a high energy demand.

The preparation of polysiloxane-polycarbonate block copolymers via the interfacial process is known from the literature and is described, for example, in US-A 3 189 662, US-A 3 419 634, DE-B 3 34 782 and EP 122 535.

The preparation of polysiloxane carbonate block copolymers by the melt transesterification process from bisphenol, diaryl carbonate and silanol end-terminated polysiloxanes in the presence of a catalyst is described in US 5 227 449. The siloxane compounds used are polydiphenyl- or polydimethylsiloxane telomers with silanol end groups. It is known, however, that such dimethylsiloxanes having silanol end groups, in contrast to diphenylsiloxane with silanol end groups, have an increasing tendency to self-condensation with decreasing chain length in an acidic or basic medium, such that incorporation into the copolymer as it forms is made more difficult as a result. Cyclic siloxanes formed in this process remain in the polymer and have an exceptionally disruptive effect in applications in the electrical/electronics sector.

US 5 504 177 describes the preparation of a block copolysiloxane carbonate via melt transesterification from a carbonate-terminated silicone with bisphenol and diaryl carbonate. Because of the great incompatibility of the siloxanes with bisphenol and diaryl carbonate, homogeneous incorporation of the siloxanes into the polycarbonate matrix can be achieved only with very great difficulty, if at all, via the melt transesterification process. Furthermore, the preparation of the block cocondensates proceeding from the monomers is very demanding.

Disadvantages of all these processes are the use of organic solvents in at least one step of the synthesis of the silicone-polycarbonate block copolymers, the use of phosgene as a feedstock and/or the inadequate quality of the cocondensate. More particularly, the synthesis of the cocondensates proceeding from the monomers is very demanding, both in the interfacial process and particularly in the melt transesterification process. For example, in the case of the melt process, a small relative underpressure and low temperatures have to be employed, in order to prevent vaporization and hence removal of the monomers. Only in later reaction stages in which oligomers with higher molar mass have formed can lower pressures and higher temperatures be employed. This means that the reaction has to be conducted over several stages and that the reaction times are accordingly long. Furthermore, there is a risk that production process residues such as low-molecular siloxane components remain in the co-condensate.

It is known that branching can have an effect on the rheological properties of polycarbonates. For instance, branched polycarbonates exhibit high structural viscosity. This is used in extrusion processes, e.g. production of water bottles or multi-skin sheets. The preparation of such branched polycarbonates is well known and described in EP 649724. In addition to the manufacturing of branched polycarbonates via the interfacial process with phosgene, the preparation of branched polycarbonates via the melt transesterification process is also known and described in EP 2374829. In these processes, branching is achieved by the use of multifunctional compounds, such as 1,1,1-tris-(4-hydroxyphenyl)ethane. Interestingly, these defective structures also lead to branching, but they have a disadvantageous effect on the rheology of the resultant polycarbonates during processing thereof (EP 2374829).

DE 19710081 describes a process for preparing the cocondensates mentioned in a melt transesterification process proceeding from an oligocarbonate and a specific hydroxyarylsiloxane. The preparation of the oligocarbonate is also described in this application. However, the industrial scale preparation of oligocarbonates for preparation of relatively small-volume specific cocondensates is very costly and inconvenient. Furthermore, the resulting material is unsuitable for the preparation of cocondensates, since the high concentration of OH end groups and other impurities, for example catalyst residue constituents, lead to a poor colour in the end product.

Nowadays, polycarbonates are industrially manufactured from the monomers, i.e. from low molecular weight bisphenols and organic carbonates such as diphenyl carbonate, which is very demanding and requires a costly standard polycarbonate synthesis or copolycarbonate synthesis in a corresponding industrial scale plant

5 None of the abovementioned applications describes polysiloxane-polycarbonate block cocondensates comprising particular rearrangement structures. Further, siloxane-containing block condensates derived from commercially available polycarbonates and siloxane-components having a molecular weight of more than 3000 g/mol and exhibiting high flowability under shear have not yet been described in the art. These products are advantageous because their preparation requires neither large industrial  
10 facilities, such as those for the interfacial polycondensation process, nor toxic feedstocks, such as phosgene.

Proceeding from the prior art outlined, the problem addressed was therefore that of providing siloxane-containing polycarbonate block co-condensates with high flowability and high melt stability in a cost-efficient manner.

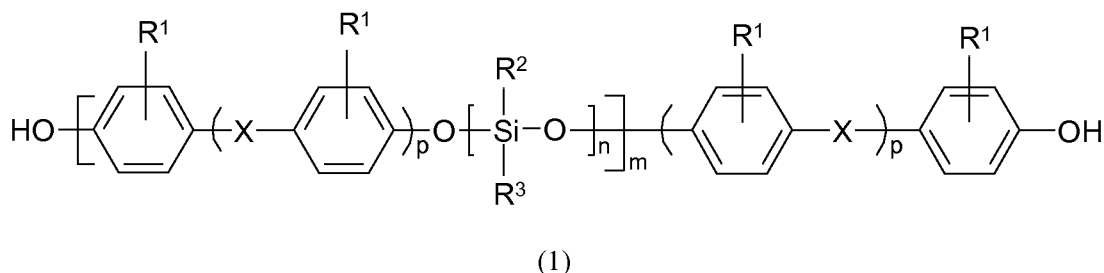
15 It has been found, surprisingly, that particular polysiloxane-polycarbonate block cocondensates containing particular rearrangement structures exhibit the above described positive features. Further, it has been found that these co-condensates can be prepared from commercially available polycarbonates without using phosgene, but nonetheless exhibits similar positive features as siloxane-containing block co-condensates prepared by the known interfacial process with phosgene.

20 This is particularly surprising because the siloxane-containing polycarbonate cocondensates that are commercially available nowadays are produced via the interfacial process with phosgene, which is distinguished by a small portion of non-reacted starting material, such as monomers and siloxane components. Due to the high reactivity of phosgene, the starting materials react virtually completely to yield the condensation product. Remaining by-products, such as salts, are almost completely removed  
25 by washing, so that the co-condensate products exhibit excellent heat stability. In contrast thereto, the polysiloxane-polycarbonate block cocondensates according to the invention are prepared via a melt transesterification process. Due to the lower reactivity of the reagents, there is a higher risk that extractable residues remain in the co-condensate product. Therefore, it is particularly surprising, that the polysiloxane-polycarbonate block cocondensates according to the invention have high melt  
30 stability and contain only traces of extractable siloxane-components. Such extractable siloxane components favour the formation of mold contaminants and thus have a negative influence on the electrical properties and/or surface properties of mouldings and extrudates made from such polysiloxane-polycarbonate block cocondensates. Therefore, it is desirable to provide polysiloxane-polycarbonate block cocondensates containing a minimum amount of extractable siloxane-components.

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The present invention therefore provides a polysiloxane-polycarbonate block cocondensate derived from

(A) at least one hydroxyaryl-terminated siloxane of the formula (1) (siloxane component)



in which

$\text{R}^1$  is H, Cl, Br or  $\text{C}_1$  to  $\text{C}_4$ -alkyl, preferably H or methyl, and especially preferably H,

$\text{R}^2$  and  $\text{R}^3$  are the same or different and each independently from one another selected from aryl,  $\text{C}_1$  to  $\text{C}_{10}$ -alkyl and  $\text{C}_1$  to  $\text{C}_{10}$ -alkylaryl, preferably  $\text{R}^2$  and  $\text{R}^3$  are methyl,

X is a single bond,  $-\text{CO}-$ ,  $-\text{O}-$ ,  $\text{C}_1$ - to  $\text{C}_6$ -alkylene,  $\text{C}_2$  to  $\text{C}_5$ -alkylidene,  $\text{C}_5$  to  $\text{C}_{12}$ -cycloalkylidene or  $\text{C}_6$  to  $\text{C}_{12}$ -arylene which may optionally be fused to further aromatic rings containing heteroatoms, X preferably being a single bond,  $\text{C}_1$  to  $\text{C}_5$ -alkylene,  $\text{C}_2$  to  $\text{C}_5$ -alkylidene,  $\text{C}_5$  to  $\text{C}_{12}$ -cycloalkylidene,  $-\text{O}-$  or  $-\text{CO}-$ , X more preferably being a single bond, isopropylidene,  $\text{C}_5$ - to  $\text{C}_{12}$ -cycloalkylidene or oxygen, and most preferably isopropylidene,

n is a number from 1 to 500, preferably from 10 to 400, especially preferably from 10 to 100, most preferably from 20 to 60,

m is a number from 1 to 10, preferably from 1 to 6, especially preferably from 2 to 5, and

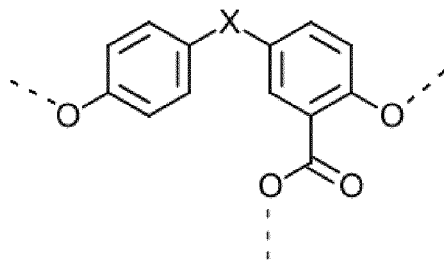
p is 0 or 1, preferably 0,

and the value of n times m is preferably between 12 and 400, more preferably between 15 and 200;

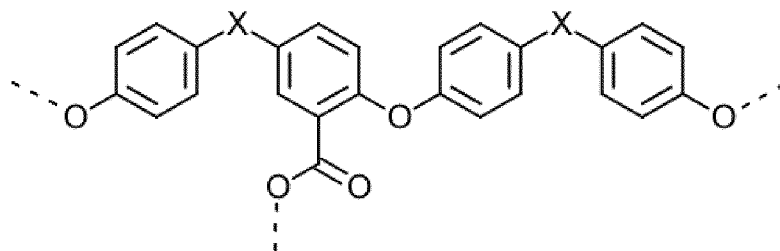
(B) and at least one polycarbonate

which polysiloxane-polycarbonate block cocondensate contains at least one, preferably more than one, of the following structures (I) to (IV):

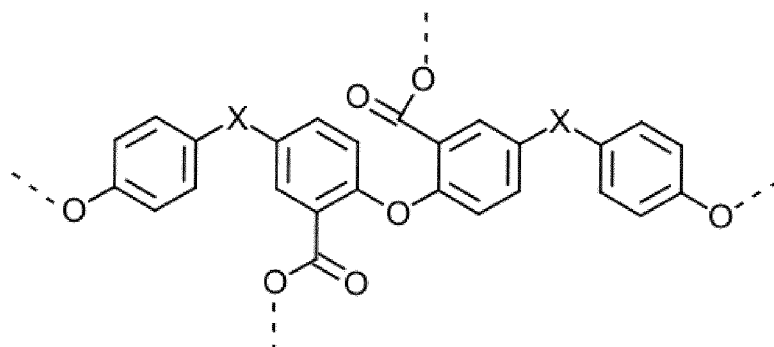
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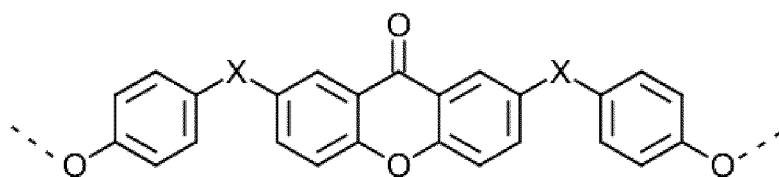
(I)



(II)



(III)



(IV)

5

in which

the phenyl rings are unsubstituted or independently mono- or disubstituted by C<sub>1</sub> to C<sub>8</sub>-alkyl and/or halogen, preferably C<sub>1</sub> to C<sub>4</sub>-alkyl, more preferably methyl,

X is a single bond, C<sub>1</sub> to C<sub>6</sub>-alkylene, C<sub>2</sub> to C<sub>5</sub>-alkylidene or C<sub>5</sub> to C<sub>6</sub>-cycloalkylidene, preferably a single bond or C<sub>1</sub> to C<sub>4</sub>-alkylene, and especially preferably isopropylidene,

the linkages indicated by --- in the structural units (I) to (IV) are each part of a carboxylate group;

5 and wherein the amount of the structural units (I) to (IV) totals 60 to 1500, preferably 70 to 1200 ppm, and most preferably 80 to 850 ppm (determined after hydrolysis, based on the polysiloxane-polycarbonate block cocondensate).

10 The structural units (I), (II), (III) and/or (IV) are built into the polymer chain of the polysiloxane-polycarbonate block cocondensate, preferably into those parts of the polymer chain that are derived from the polycarbonate (component B).

Preferably, the polysiloxane-polycarbonate block cocondensate according to the invention has a relative solution viscosity of 1.26 to 1.40, more preferably of 1.27 to 1.38, and especially preferably of 1.28 to 1.35, determined in dichloromethane at a concentration of 5 g/l at 25°C using a Ubbelohde viscosimeter.

15 Preferably, the polysiloxane-polycarbonate block cocondensate according to the invention has a weight average molecular weight of 26,000 to 40,000 g/mol, more preferably 27,000 to 38,000 g/mol, and most preferably 28,000 to 35,000 g/mol, determined by measuring the relative solution viscosity in dichloromethane at a concentration of 5 g/l at 25°C using a Ubbelohde viscosimeter.

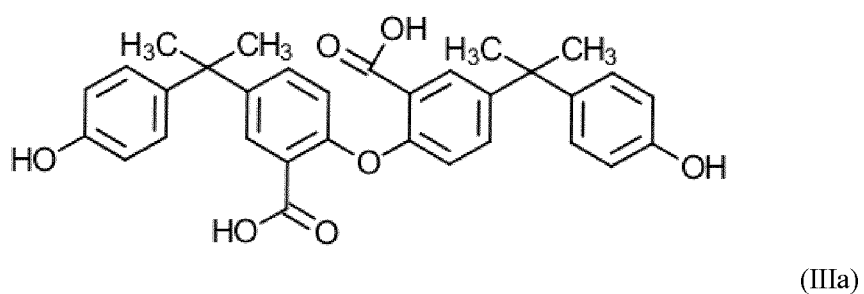
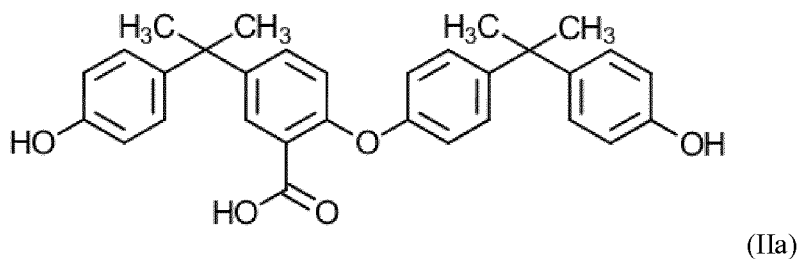
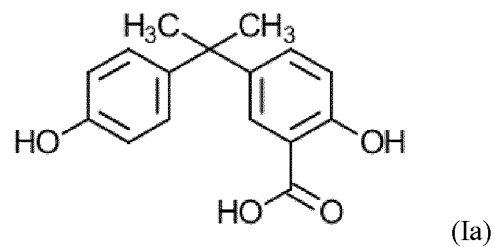
20 In a preferred embodiment, the polysiloxane-polycarbonate block cocondensate according to the invention has a sodium content of at least 50 ppb, preferably at least 80 ppb, more preferably at least 100 ppb, and in particular at least 150 ppb, based on the total weight of the polysiloxane-polycarbonate block cocondensate. In a preferred embodiment, the polysiloxane-polycarbonate block cocondensate according to the invention has a sodium content within the range of from 0.1 ppm to 1000 ppm, preferably 0.2 to 100 ppm, more preferably 0.3 to 10 ppm and in particular 0.4 to 5 ppm, based on the  
25 total weight of the polysiloxane-polycarbonate block cocondensate. The sodium content of the cocondensate can be determined, for example, by atomic absorption spectroscopy with flame atomization.

The rearrangement structures mentioned occur in different amounts and ratios relative to one another. The amounts thereof can be determined by total hydrolysis of the polysiloxane-polycarbonate block  
30 cocondensate.

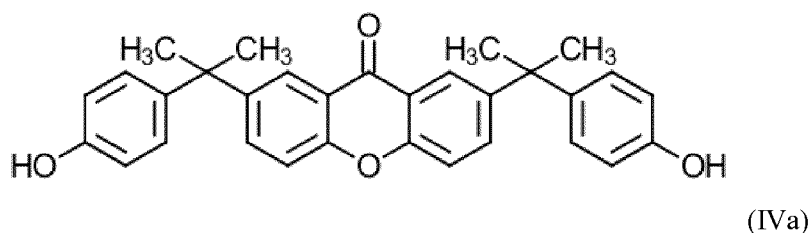
In order to determine the amount of the rearrangement structures, the particular polysiloxane-polycarbonate block cocondensate is subjected to a total hydrolysis and the corresponding degradation products of the formulae (Ia) to (IVa) are thus formed, the amount of which is determined by HPLC.

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(This can be done, for example, as follows: the polycarbonate sample is hydrolysed under reflux by means of sodium methoxide. The corresponding solution is acidified and concentrated to dryness. The drying residue is dissolved in acetonitrile and the phenolic compounds of the formula (Ia) to (IVa) are determined by means of HPLC with UV detection.







Preferably, the amount of the compound of the formula (Ia) released is 20 to 800 ppm, more preferably 25 to 700 ppm and especially preferably 30 to 500 ppm, based on the polysiloxane-polycarbonate block cocondensate.

Preferably, the amount of the compound of the formula (IIa) released is 0 (i.e. below the detection limit of 10 ppm) to 100 ppm, more preferably 0 to 80 ppm and especially preferably 0 to 50 ppm, based on the polysiloxane-polycarbonate block cocondensate.

Preferably, the amount of the compound of the formula (IIIa) released is 10 to 800 ppm, further preferably 10 to 700 ppm and more preferably 20 to 600 ppm, and most preferably 30 to 350 ppm, based on the polysiloxane-polycarbonate block cocondensate.

Preferably, the amount of the compound of the formula (IVa) released is 0 (i.e. below the detection limit of 10 ppm) to 300 ppm, preferably 10 to 250 ppm and most preferably 20 to 200 ppm, based on the polysiloxane-polycarbonate block cocondensate.

For reasons of simplification, the amount of the structures of the formula (I) to (IV) is equated to the respective amount of the compounds of the formula (Ia) to (IVa) released.

“C<sub>1</sub>-C<sub>4</sub>-alkyl” in the context of the invention is, for example, methyl, ethyl, n-propyl, isopropyl, n-butyl, sec-butyl, tert-butyl; “C<sub>1</sub>-C<sub>6</sub>-alkyl” is additionally, for example, n-pentyl, 1-methylbutyl, 2-methylbutyl, 3-methylbutyl, neopentyl, 1-ethylpropyl, cyclohexyl, cyclopentyl, n-hexyl, 1,1-dimethylpropyl, 1,2-dimethylpropyl, 1-methylpentyl, 2-methylpentyl, 3-methylpentyl, 4-methylpentyl, 1,1-dimethylbutyl, 1,2-dimethylbutyl, 1,3-dimethylbutyl, 2,2-dimethylbutyl, 2,3-dimethylbutyl, 3,3-dimethylbutyl, 1-ethylbutyl, 2-ethylbutyl, 1,1,2-trimethylpropyl, 1,2,2-trimethylpropyl, 1-ethyl-1-methylpropyl or 1-ethyl-2-methylpropyl; “C<sub>1</sub>-C<sub>10</sub>-alkyl” is additionally, for example, n-heptyl and n-octyl, pinacyl, adamantyl, the isomeric menthyls, n-nonyl, n-decyl; C<sub>1</sub>-C<sub>34</sub>-alkyl is additionally for example, n-dodecyl, n-tridecyl, n-tetradecyl, n-hexadecyl or n-octadecyl. The same applies to the corresponding alkyl radical, for example, in aralkyl or alkylaryl, alkylphenyl or alkylcarbonyl radicals. Alkylene radicals in the corresponding hydroxyalkyl or aralkyl or alkylaryl radicals are, for example, alkylene radicals corresponding to the above alkyl radicals.

“Aryl” is a carbocyclic aromatic radical having 6 to 34 skeletal carbon atoms. The same applies to the aromatic moiety of an arylalkyl radical, also called aralkyl radical, and to aryl constituents of more complex groups, for example arylcarbonyl radicals.

5 Examples of “C<sub>6</sub>-C<sub>34</sub>-aryl” are phenyl, o-, p-, m-tolyl, naphthyl, phenanthrenyl, anthracenyl or fluorenyl.

“Arylalkyl” or “aralkyl” is in each case independently a straight-chain, cyclic, branched or unbranched alkyl radical as defined above, which may be singly, multiply or fully substituted by aryl radicals as defined above.

The above enumerations should be understood by way of example and not as a limitation.

10 In the context of the present invention, ppm and ppb – unless stated otherwise – are understood to mean parts by weight.

The preparation of polycarbonates containing the structural elements (I) to (IV) on the industrial scale is known in principle and is described, for example, in DE 102008019503.

15 Polycarbonates in the context of the present invention are both homopolycarbonates and copolycarbonates.

Preferably, the polycarbonate has a weight-average molecular weight of 16 000 to 28 000 g/mol, preferably of 17 000 to 27 000 g/mol, and especially preferably of 18 000 to 26 500 g/mol, determined by means of gel permeation chromatography and BPA (bisphenol A) standard, and phenolic OH groups in an amount of 250 ppm to 1000 ppm, preferably 300 to 900 ppm and especially preferably of  
20 350 to 800 ppm.

In a preferred embodiment, the polycarbonate has a relative solution viscosity ( $\eta_{rel}$ ) of 1.16 to 1.30, preferably 1.17 to 1.28, and more preferably 1.18 to 1.27, determined in dichloromethane at a concentration of 5 g/l at 25°C using a Ubbelohde viscosimeter.

25 Preferred modes of preparation of the polycarbonates, including the polyesterarbonates, proceed by the known melt transesterification process.

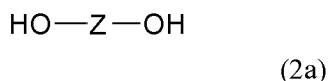
Some, preferably up to 80 mol%, more preferably from 20 mol% up to 50 mol%, of the carbonate groups in the polycarbonates suitable in accordance with the invention may be replaced by aromatic dicarboxylic ester groups. Such polycarbonates, which contain both acid radicals of the carbonic acid and acid radicals of aromatic dicarboxylic acids incorporated into the molecule chain, are, to be exact,  
30 aromatic polyesterarbonates. For the sake of simplicity, they are to be covered in the present application by the umbrella term of thermoplastic aromatic polycarbonates.

Polycarbonates are prepared in a known manner from diphenols, carbonic acid derivatives, optionally chain terminators and optionally branching agents, with replacement of a portion of the carbonic acid derivatives by aromatic dicarboxylic acids or derivatives of the dicarboxylic acids for preparation of the polyestercarbonates, specifically by aromatic dicarboxylic ester structural units according to the carbonate structural units to be replaced in the aromatic polycarbonates.

By way of example for the preparation of polycarbonates, reference is made here to Schnell, "Chemistry and Physics of Polycarbonates", Polymer Reviews, Volume 9, Interscience Publishers, New York, London, Sydney 1964.

Diphenols suitable for preparation of polycarbonates have been described many times in the prior art.

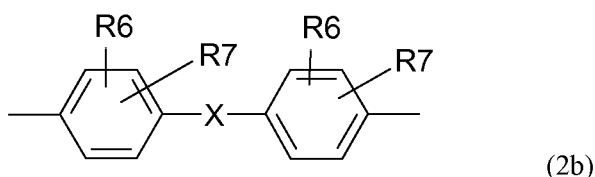
Preferably, the polycarbonate, and thereby also the polysiloxane-polycarbonate block cocondensate according to the invention, contains structures derived from a diphenol according to general formula (2a):



wherein

Z is an aromatic residue having 6 to 30 carbon atoms, which contains one or more optionally substituted aromatic rings, wherein the aromatic rings may be connected by an aliphatic residue or alkylaryl group or hetero atom bridge.

In a preferred embodiment, Z is a residue according to formula (2b)



wherein

$R^6$  and  $R^7$  are independently of one another H,  $C_1$ - $C_{18}$ -alkyl-,  $C_1$ - $C_{18}$ -alkoxy, halogen such as Cl or Br or an optionally substituted aryl- or aralkyl, preferably H or  $C_1$ - $C_{12}$ -alkyl, more preferably H or  $C_1$ - $C_8$ -alkyl, and most preferably H or methyl, and

X is a single bond, -CO-, -O-, C<sub>1</sub>- to C<sub>6</sub>-alkylene, C<sub>2</sub> to C<sub>5</sub>-alkylidene, C<sub>5</sub> to C<sub>12</sub>-cycloalkylidene or C<sub>6</sub> to C<sub>12</sub>-arylene which may optionally be fused to further aromatic rings containing heteroatoms, X preferably being a single bond, C<sub>1</sub> to C<sub>5</sub>-alkylene, C<sub>2</sub> to C<sub>5</sub>-alkylidene, C<sub>5</sub> to C<sub>12</sub>-cycloalkylidene, -O- or -CO-, X more preferably being a single bond, isopropylidene, C<sub>5</sub>- to C<sub>12</sub>-cycloalkylidene or oxygen, and most preferably isopropylidene.

Suitable diphenols according to formula (2a) are, for example, hydroquinone, resorcinol, dihydroxydiphenyl, bis(hydroxylphenyl)alkanes, bis(hydroxyphenyl) sulphides, bis(hydroxyphenyl) ethers, bis(hydroxyphenyl) ketones, bis(hydroxyphenyl) sulphones, bis(hydroxyphenyl) sulfoxides, α,α'-bis(hydroxylphenyl)diisopropylbenzenes, and the alkylated, ring-alkylated and ring-halogenated compounds thereof.

Preferred diphenols according to formula (2a) are 4,4'-dihydroxydiphenyl, 2,2-bis(4-hydroxyphenyl)1-phenylpropane, 1,1-bis(4-hydroxyphenyl)phenylethane, 2,2-bis(4-hydroxyphenyl)propane, 2,4-bis(4-hydroxyphenyl)2-methylbutane, 1,3-bis[2-(4-hydroxyphenyl)2-propyl]benzene (bisphenol M), 2,2-bis(3-methyl-4-hydroxyphenyl)propane, bis(3,5-dimethyl-4-hydroxyphenyl)methane, 2,2-bis(3,5-dimethyl-4-hydroxyphenyl)propane, bis(3,5-dimethyl-4-hydroxyphenyl) sulphone, 2,4-bis(3,5-dimethyl-4-hydroxyphenyl)2-methylbutane and 1,3-bis[2-(3,5-dimethyl-4-hydroxyphenyl)2-propyl]benzene.

Particularly preferred diphenols according to formula (2a) are 4,4'-dihydroxydiphenyl, 1,1-bis(4-hydroxyphenyl)phenylethane and 2,2-bis(4-hydroxyphenyl)propane, 2,2-bis(3,5-dimethyl-4-hydroxyphenyl)propane. These and further suitable other dihydroxyaryl compounds are described, for example, in DE-A 3 832 396, FR-A 1 561 518, in H. Schnell, Chemistry and Physics of Polycarbonates, Interscience Publishers, New York 1964, p. 28 ff.; p.102 ff. and in D.G. Legrand, J.T. Bendler, Handbook of Polycarbonate Science and Technology, Marcel Dekker New York 2000, p. 72 ff..

In the case of the homopolycarbonates, only one diphenol is used; in the case of the copolycarbonates, a plurality of diphenols are used; it will be appreciated that the diphenols used, and also all the other chemicals and assistants added to the synthesis, may be contaminated with the impurities originating from their own synthesis, handling and storage, although it is desirable to work with raw materials of maximum cleanliness.

The polycarbonates can be modified in a conscious and controlled manner by the use of small amounts of chain terminators and branching agents. Suitable chain terminators and branching agents are known from the literature. Some are described, for example, in DE-A 38 33 953. Preferably used chain terminators are phenol or alkylphenols, in particular phenol, p-tert-butylphenol, isooctylphenol, cumylphenol, the chlorocarbonic acid esters thereof or acid chlorides of monocarboxylic acids or

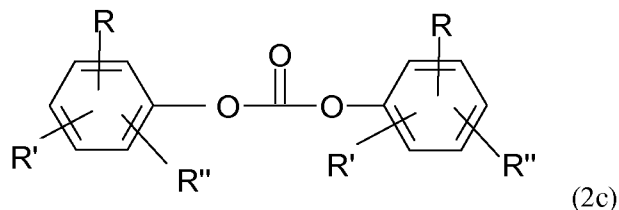
mixtures of these chain terminators. Preferred chain terminators are phenol, cumylphenol, isooctylphenol, para-tert-butylphenol, and in particular phenol.

Examples of compounds suitable as branching agents are aromatic or aliphatic compounds having at least three, preferably three or four, hydroxyl groups. Particularly suitable examples having three or more than three phenolic hydroxyl groups are phloroglucinol, 4,6-dimethyl-2,4,6-tri(4-hydroxyphenyl)hept-2-ene, 4,6-dimethyl-2,4,6-tri(4-hydroxyphenyl)heptane, 1,3,5-tri(4-hydroxyphenyl)-benzene, 1,1,1-tri(4-hydroxyphenyl)ethane, tri(4-hydroxyphenyl)phenylmethane, 2,2-bis(4,4-bis(4-hydroxyphenyl)cyclohexyl)propane, 2,4-bis(4-hydroxyphenylisopropyl)phenol, tetra(4-hydroxyphenyl)methane.

Examples of other trifunctional compounds suitable as branching agents are 2,4-dihydroxybenzoic acid, trimesic acid, cyanuric chloride and 3,3-bis(3-methyl-4-hydroxyphenyl)-2-oxo-2,3-dihydroindole.

Particularly preferred branching agents are 3,3-bis(3-methyl-4-hydroxyphenyl)-2-oxo-2,3-dihydroindole and 1,1,1-tri(4-hydroxyphenyl)ethane.

The diaryl carbonates suitable for the reaction with the dihydroxyaryl compounds in the melt transesterification are those of the general formula (2c)



in which

R, R' and R'' are the same or different and are each independently hydrogen, linear or branched C<sub>1</sub>-C<sub>34</sub>-alkyl, C<sub>7</sub>-C<sub>34</sub>-alkylaryl or C<sub>6</sub>-C<sub>34</sub>-aryl,

R may additionally also be -COO-R''' where R''' is hydrogen, linear or branched C<sub>1</sub>-C<sub>34</sub>-alkyl, C<sub>7</sub>-C<sub>34</sub>-alkylaryl or C<sub>6</sub>-C<sub>34</sub>-aryl.

Preferred diaryl carbonates are, for example, diphenyl carbonate, methylphenyl phenyl carbonates and di(methylphenyl) carbonates, 4-ethylphenyl phenyl carbonate, di(4-ethylphenyl) carbonate, 4-n-propylphenyl phenyl carbonate, di(4-n-propylphenyl) carbonate, 4-isopropylphenyl phenyl carbonate, di(4-isopropylphenyl) carbonate, 4-n-butylphenyl phenyl carbonate, di(4-n-butylphenyl) carbonate, 4-isobutylphenyl phenyl carbonate, di(4-isobutylphenyl) carbonate, 4-tert-butylphenyl phenyl carbonate, di(4-tert-butylphenyl) carbonate, 4-n-pentylphenyl phenyl carbonate, di(4-n-pentylphenyl) carbonate, 4-n-hexylphenyl phenyl carbonate, di(4-n-hexylphenyl) carbonate,

4-isooctylphenyl phenyl carbonate, di(4-isooctylphenyl) carbonate, 4-n-nonylphenyl phenyl carbonate, di(4-n-nonylphenyl) carbonate, 4-cyclohexylphenyl phenyl carbonate, di(4-cyclohexylphenyl) carbonate, 4-(1-methyl-1-phenylethyl)phenyl phenyl carbonate, di[4-(1-methyl-1-phenylethyl)phenyl] carbonate, biphenyl-4-yl phenyl carbonate, di(biphenyl-4-yl) carbonate, 4-(1-naphthyl)phenyl phenyl carbonate, 4-(2-naphthyl)phenyl phenyl carbonate, di[4-(1-naphthyl)phenyl] carbonate, di[4-(2-naphthyl)phenyl] carbonate, 4-phenoxyphenyl phenyl carbonate, di(4-phenoxyphenyl) carbonate, 3-pentadecylphenyl phenyl carbonate, di(3-pentadecylphenyl) carbonate, 4-tritylphenyl phenyl carbonate, di(4-tritylphenyl) carbonate, (methyl salicylate) phenyl carbonate, di(methyl salicylate) carbonate, (ethyl salicylate) phenyl carbonate, di(ethyl salicylate) carbonate, (n-propyl salicylate) phenyl carbonate, di(n-propyl salicylate) carbonate, (isopropyl salicylate) phenyl carbonate, di(isopropyl salicylate) carbonate, (n-butyl salicylate) phenyl carbonate, di(n-butyl salicylate) carbonate, (isobutyl salicylate) phenyl carbonate, di(isobutyl salicylate) carbonate, (tert-butyl salicylate) phenyl carbonate, di(tert-butyl salicylate) carbonate, di(phenyl salicylate) carbonate and di(benzyl salicylate) carbonate.

Particularly preferred diaryl compounds are diphenyl carbonate, 4-tert-butylphenyl phenyl carbonate, di(4-tert-butylphenyl) carbonate, biphenyl-4-yl-phenyl carbonate, di(biphenyl-4-yl) carbonate, 4-(1-methyl-1-phenylethyl)phenyl phenyl carbonate, di[4-(1-methyl-1-phenylethyl)phenyl] carbonate and di(methyl salicylate) carbonate.

Very particular preference is given to diphenyl carbonate.

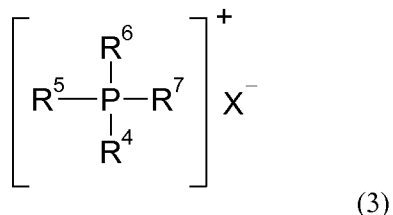
It is possible to use either one diaryl carbonate or else various diaryl carbonates.

The diaryl carbonates can also be used with residual contents of the monohydroxyaryl compounds from which they have been prepared. The residual contents of the monohydroxyaryl compounds may be up to 20% by weight, preferably up to 10% by weight, more preferably up to 5% by weight and most preferably up to 2% by weight.

Based on the dihydroxyaryl compound(s), generally 1.02 to 1.30 mol of the diaryl carbonate(s), preferably 1.04 to 1.25 mol, more preferably 1.045 to 1.22 mol and most preferably 1.05 to 1.20 mol per mole of dihydroxyaryl compound are used. It is also possible to use mixtures of the abovementioned diaryl carbonates, in which case the above-stated molar figures per mole of dihydroxyaryl compound relate to the total amount of the mixture of the diaryl carbonates.

The catalysts used in the melt transesterification process for preparation of polycarbonates may be the basic catalysts known in the literature, for example alkali metal and alkaline earth metal hydroxides and oxides and/or onium salts, for example ammonium or phosphonium salts. Preference is given to using onium salts in the synthesis, more preferably phosphonium salts. Such phosphonium salts are, for example, those of the general formula (3)

-14-



in which

- 5             $\text{R}^{4-7}$  are identical or different, optionally substituted  $\text{C}_1\text{-C}_{10}$ -alkyl,  $\text{C}_6\text{-C}_{14}$ -aryl,  $\text{C}_7\text{-C}_{15}$ -arylalkyl or  $\text{C}_5\text{-C}_6$ -cycloalkyl radicals, preferably methyl or  $\text{C}_6\text{-C}_{14}$ -aryl, more preferably methyl or phenyl, and
- $\text{X}^-$  is an anion selected from the group of hydroxide, sulphate, hydrogensulphate, hydrogencarbonate, carbonate, halide, preferably chloride, and alkoxide or aroxide of
- 10            the formula  $-\text{OR}^8$  where  $\text{R}^8$  is an optionally substituted  $\text{C}_6\text{-C}_{14}$ -aryl,  $\text{C}_7\text{-C}_{15}$ -arylalkyl,  $\text{C}_5\text{-C}_6$ -cycloalkyl or  $\text{C}_1\text{-C}_{20}$ -alkyl radical, preferably phenyl.

Particularly preferred catalysts are tetraphenylphosphonium chloride, tetraphenylphosphonium hydroxide and tetraphenylphosphonium phenoxide, very particular preference being given to tetraphenylphosphonium phenoxide.

- 15            The catalysts are used preferably in amounts of  $10^{-8}$  to  $10^{-3}$  mol, more preferably in amounts of  $10^{-7}$  to  $10^{-4}$  mol, based on one mole of dihydroxyaryl compound.

It is optionally also possible to use cocatalysts in order to increase the rate of polycondensation.

- These may, for example, be alkaline salts of alkali metals and alkaline earth metals, such as hydroxides, optionally substituted  $\text{C}_1\text{-C}_{10}$ -alkoxides and  $\text{C}_6\text{-C}_{14}$ -aroxides of lithium, sodium and
- 20            potassium, preferably hydroxides, optionally substituted  $\text{C}_1\text{-C}_{10}$ -alkoxides or  $\text{C}_6\text{-C}_{14}$ -aroxides of sodium. Preference is given to sodium hydroxide, sodium phenoxide or the disodium salt of 2,2-bis(4-hydroxyphenyl)propane.

- If alkali metal or alkaline earth metal ions are supplied in the form of their salts, the amounts of alkali metal or alkaline earth metal ions, determined, for example, by atomic absorption spectroscopy, is 1 to
- 25            500 ppb, preferably 5 to 300 ppb and most preferably 5 to 200 ppb, based on polycarbonate to be formed. In preferred embodiments of the process according to the invention, however, no alkali metal salts are used.

The performance of the polycarbonate synthesis may be continuous or batchwise.

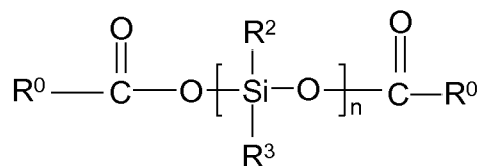
Preference is additionally given to polycarbonates which bear phenol as end groups (phenyl-terminated polycarbonate).

In a particular embodiment, polycarbonates having a water content of 0.01 to 0.40 and preferably 0.05 to 0.35% by weight are used.

- 5 The weight-average molecular weight of the siloxane component is preferably 3000 to 20 000 g/mol, determined by means of gel permeation chromatography and BPA (bisphenol A) Standard, and especially preferably 3500-15 000 g/mol.

Very particular preference is given to using, as the siloxane component, hydroxyaryl-terminated siloxanes of the formula (1) where the  $R^2$  and  $R^3$  radicals are both methyl and the  $R^1$  radical is  
10 hydrogen, and p is 0.

The siloxanes of the formula (1) can be prepared by a process including the step of reacting a linear  $\alpha,\omega$ -bisacyloxy-polydialkylsiloxane of the general formula (4) with at least one aromatic compound having at least two phenolic hydroxy groups, wherein the general formula (4) is



15 (4)

in which

$R^0$  is aryl,  $C_1$  to  $C_{10}$ -alkyl or  $C_1$  to  $C_{10}$ -alkylaryl,

$R^2$  and  $R^3$  are the same or different and each independently from one another selected from  
20 aryl,  $C_1$  to  $C_{10}$ -alkyl and  $C_1$  to  $C_{10}$ -alkylaryl, preferably  $R^2$  and  $R^3$  are both methyl, and

n is a number from 1 to 500, preferably from 10 to 400, especially preferably from 10 to 100, most preferably from 20 to 60,

and wherein the compound of formula (4) and the aromatic compound are reacted in such a molar ratio that the ratio of the phenolic hydroxyl groups in the aromatic compound to the acyloxy groups in the  
25 compound of formula (4) is less than 2.0.

In a particularly preferred embodiment, the aromatic compound having at least two phenolic hydroxy groups is a bisphenolic compound or a hydroxyl-functional oligomer thereof.



The preparation of the siloxane of formula (1) is preferably performed in an inert solvent, preferably selected from aromatic hydrocarbons such as toluene, xylenes, chlorobenzene and the like, and polar organic acids, such as acetic acid and other C3 to C6 organic carboxylic acids. The reaction can be performed in the presence of a catalyst, which is preferably selected from the metal salts of organic acids, such as sodium or potassium acetate. Other catalysts known in the art to catalysed siloxane condensation reactions can also be used.

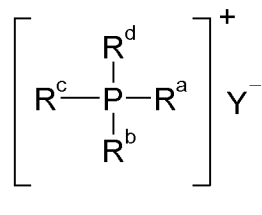
Preferably, the polysiloxane-polycarbonate block cocondensate according to the invention is derived from a composition comprising the siloxane component of the formula (1) and the polycarbonate and which contains the siloxane component in an amount of 0.5 to 50% by weight, more preferably of 1 to 40% by weight, especially preferably of 2 to 20%, most preferably of 2.5 to 10% by weight, and in particular 2.5% by weight to 7.5% by weight, based in each case on the polycarbonate used.

Preferably, the polysiloxane-polycarbonate block cocondensate according to the invention can be obtained by reacting the component (A) with the component (B) in the melt at temperatures of 280°C to 400°C, preferably of 300°C to 390°C, more preferably of 320°C to 380°C and most preferably of 330°C to 370°C, and pressures of 0.001 mbar to 50 mbar, preferably 0.005 mbar to 40 mbar, especially preferably 0.02 to 30 mbar, and most preferably 0.03 to 5 mbar, preferably in the presence of a catalyst.

Preference is given to reacting the polycarbonate and the siloxane by means of catalysts. It is also possible in principle to conduct the reaction without catalyst, but in that case it may be necessary to accept higher temperatures and longer residence times.

Suitable catalysts are, for example, tetra alkyl ammonium catalysts, for example tetramethylammonium hydroxide, tetramethylammonium acetate, tetramethylammonium fluoride, tetramethylammonium tetraphenylboranate, dimethyldiphenylammonium hydroxide, tetraethylammonium hydroxide, cetyltrimethylammonium tetraphenylboranate and cetyltrimethylammonium phenoxide.

Especially suitable catalysts are phosphonium catalysts of the formula (5):



where R<sup>a</sup>, R<sup>b</sup>, R<sup>c</sup> and R<sup>d</sup> may be identical or different C<sub>1</sub>-C<sub>10</sub>-alkyls, C<sub>6</sub>-C<sub>14</sub>-aryls, C<sub>7</sub>-C<sub>15</sub>-arylalkyls or C<sub>5</sub>-C<sub>6</sub>-cycloalkyls, preferably methyl or C<sub>6</sub>-C<sub>14</sub>-aryls, more preferably methyl or phenyl, and Y<sup>-</sup> may be an anion such as hydroxide, sulphate, hydrogensulphate, hydrogencarbonate, carbonate or a halide,

preferably chloride, or an alkoxide or aroxide of the formula  $-OR^e$  where  $R^e$  may be a  $C_6-C_{14}$ -aryl,  $C_7-C_{15}$ -arylalkyl or  $C_5-C_6$ -cycloalkyl, preferably phenyl.

Particularly preferred catalysts are tetraphenylphosphonium chloride, tetraphenylphosphonium hydroxide and tetraphenylphosphonium phenoxide; very particular preference is given to tetraphenylphosphonium phenoxide.

The catalyst is used preferably in amounts of 0.0001 to 1.0% by weight, preferably from 0.001 to 0.5% by weight, especially preferably from 0.005 to 0.3% by weight and most preferably from 0.01 to 0.15% by weight, based on the overall composition.

The catalyst can be used alone or as a catalyst mixture and be added in substance or as a solution, for example in water or in phenol (for example as a cocrystal with phenol).

Catalysts suitable for preparing the co-condensate according to the invention are those mentioned above, which are introduced into the reaction either by means of a masterbatch with a suitable polycarbonate, especially the above-described inventive polycarbonate, or can be added separately therefrom or in addition thereto.

The catalysts can be used alone or in a mixture and be added in substance or as a solution, for example in water or in phenol.

In a preferred embodiment, reaction of the siloxane of the formula (1) and the polycarbonate is performed in presence of at least one organic or inorganic salt of an acid having a  $pK_A$  value within the range of from 3 to 7 (25 °C). Suitable acids include carboxylic acids, preferably  $C_2-C_{22}$  carboxylic acids, such as acetic acid, propionic acid, oleic acid, stearic acid, lauric acid, benzoic acid, 4-methoxybenzoic acid, 3-methylbenzoic acid, 4-tert-butylbenzoic acid, p-tolylacetic acid, 4-hydroxybenzoic acid and salicylic acid, partial esters of polycarboxylic acids, such as monoesters of succinic acid, partial esters of phosphoric acid, such as mono- or diorgano phosphoric acid esters, branched aliphatic carboxylic acids, such as 2,2-dimethylpropanoic acid, 2,2-dimethylbutanoic acid, 2,2-dimethylpentanoic acid and 2-ethylhexanoic acid.

The organic or inorganic salt is preferably used in addition to the catalyst of formula (5); and in this embodiment the organic or inorganic salt acts as co-catalyst.

Preferably, the organic or inorganic salt is selected from the group consisting of alkali metal salts, earth alkaline metal salts, quaternary ammonium salts and quaternary phosphonium salts. Useful quaternary ammonium salts are selected from tetra-(n-butyl)-ammonium, tetraphenylammonium, tetrabenzylammonium and cetyltrimethylammonium salts. Useful quaternary phosphonium salts are selected from tetra-(n-butyl)-phosphonium, tetraphenylphosphonium, tetrabenzylphosphonium and

cetyltrimethylphosphonium salts. Especially preferred are alkali metal salts and earth alkaline metal salts.

Useful organic and inorganic salts are or are derived from sodium hydrogencarbonate, potassium hydrogencarbonate, lithium hydrogencarbonate, sodium carbonate, potassium carbonate, lithium carbonate, sodium acetate, potassium acetate, lithium acetate, sodium stearate, potassium stearate, lithium stearate, sodium oleate, lithium oleate, potassium oleate, sodium benzoate, potassium benzoate, lithium benzoate, disodium, dipotassium and dilithium salts of bisphenol A. Furthermore the salts may include calcium hydrogencarbonate, barium hydrogencarbonate, magnesium hydrogencarbonate, strontium hydrogencarbonate, calcium carbonate, barium carbonate, magnesium carbonate, strontium carbonate, calcium acetate, barium acetate, magnesium acetate, strontium acetate, calcium stearate, barium stearate, magnesium stearate, strontium stearate and the respective oleates. These salts may be used singly or in combination.

In a particular preferred embodiment, the salt is selected from the group consisting of alkali metal salts and phosphonium salts of carboxylic acids.

In a preferred embodiment, the salt is derived from a carboxylic acid.

The organic or inorganic salts are used preferably in amounts of 0.5 to 1000 ppm, more preferably 1 to 100 ppm, and most preferably 1 to 10 ppm, based on the total weight of the siloxane and the organic or inorganic salt. Preferably, the organic or inorganic salts are used in amounts of 0.0005 to 5 mmol/kg, more preferably 0.001 to 1 mmol/kg, and most preferably 0.001 to 0.5 mmol/kg, based on the total weight of the siloxane, the polycarbonate and the organic or inorganic salt.

In a preferred embodiment, the organic or inorganic salt is a sodium salt, preferably a sodium salt of a carboxylic acid and is preferably used in such an amount that the sodium content in the resulting polysiloxane-polycarbonate block cocondensate is at least 50 ppb, preferably at least 80 ppb, more preferably at least 100 ppb, and in particular at least 150 ppb, based on the total weight of the polysiloxane-polycarbonate block cocondensate.

In a preferred embodiment, the organic or inorganic salt is a sodium salt, preferably a sodium salt of a carboxylic acid and is preferably used in such an amount that the sodium content in the resulting polysiloxane-polycarbonate block cocondensate is within the range of from 0.1 ppm to 1000 ppm, preferably 0.2 to 100 ppm, more preferably 0.3 to 10 ppm and in particular 0.4 to 5 ppm, based on the total weight of the polysiloxane-polycarbonate block cocondensate. The sodium content of the cocondensate can be determined, for example, by atomic absorption spectroscopy with flame atomization.

The organic or inorganic salt can be used alone or as a mixture and be added in substance or as a solution. In a preferred embodiment, the inorganic or organic salt is added in form of a mixture containing the siloxane and the organic or inorganic salt. Preferably, the mixture is obtained by mixing the siloxane and the organic or inorganic salt and optionally one or more polar organic compounds having up to 30, preferably up to 20 carbon atoms, and at least one heteroatom, preferably selected from O, N and S, and optionally heating the mixture, for example to a temperature of 50°C to 300°C, until it becomes clear and then cooling to room temperature. The polar organic compound can be removed before adding the mixture to the polycarbonate or thereafter, preferably by distillation.

Suitable polar organic compounds are selected from the group consisting of organic ketones, esters and alcohols. Alcohols, especially primary alcohols having up to 20 carbon atoms, such as 1-octanol, 1-decanol, 2-ethylhexanol, 1-dodecanol, 1,2-octanediol, benzyl alcohol, ethylhexylglycerin and oleoyl alcohol are particularly preferred. Preferably, the polar organic compound has a boiling point of less than 300°C (at 1.013 bar).

The process for preparing the block copolycarbonate according to the invention can be performed continuously or batchwise, for example in stirred tanks, thin-film evaporators, stirred tank cascades, extruders, kneaders and simple disc reactors. The feedstocks may be blended together and melted from the start. In addition, the feedstocks may also be added separately from one another. For instance, the polycarbonate for use in accordance with the invention can first be melted and the siloxane component for use in accordance with the invention can be added at a later time. This can be done, for example, by means of liquid metering with an appropriate pump or via granules sprinkled on to polycarbonate. The catalyst can be added at any time, preferably at the start of the reaction or after the melting, in free form or in the form of a masterbatch. The melting can be effected under air, but preferably under a protective gas atmosphere such as nitrogen or argon, or likewise preferably under reduced pressure.

The reaction is effected under the above-specified temperatures and pressures. Preference is given to shearing the reaction mixture. This can be done by rapid stirring in a tank or by means of appropriate mixing elements such as static mixers, mixing elements on an extruder screw etc. Higher mixing is preferable over low mixing. The reaction is conducted in such a way that low molecular weight constituents such as water, phenol, linear and cyclic low molecular weight siloxane, diphenyl carbonate, bisphenol A and bisphenol A oligomers (oligocarbonates) are removed effectively.

The reactants are preferably melted under reduced pressure. According to the plant construction, during the melting phase, atmospheric pressure, preferably gentle vacuum, i.e. absolute pressures of lower than 200 mbar, especially preferably 100-200 mbar and most preferably less than 100 mbar can be applied. However, the reactants can also be melted under standard pressure, preferably under protective gas atmosphere, for example nitrogen. The melting is preferably effected at a temperature in the range

from 250 to 400°C, more preferably in the range from 280 to 380°C, most preferably in the range from 300 to 360°C. For the reaction or condensation phase, the temperatures and pressures mentioned above apply.

Granules are obtained, if possible, by direct spinning of the melt and subsequent granulation, or else through use of discharge extruders or gear pumps, by which spinning is effected in air or under liquid, usually water. If extruders are utilized, additives can be added to the melt upstream of this extruder, optionally with use of static mixers or by means of side extruders in the extruder.

It is possible to add additives and/or fillers and reinforcers to the polysiloxane-polycarbonate block cocondensate according to the invention. Additives are preferably used in amounts of 0% by weight to 5.0% by weight, more preferably 0% by weight to 2.0% by weight, and most preferably 0% by weight to 1.0% by weight. The additives are standard polymer additives, for example the following which are described in EP-A 0 839 623, WO-A 96/15102, EP A 0 500 496 or "Plastics Additives Handbook", Hans Zweifel, 5th Edition 2000, Hanser Verlag, Munich: flame retardants, UV stabilizers, gamma stabilizers, antistats, optical brighteners, flow improvers, thermal stabilizers, inorganic pigments, demoulding agents or processing aids. Fillers and/or reinforcers can be used in amounts of 0% by weight to 50% by weight, preferably 0% by weight to 20% by weight, more preferably 0% by weight to 12% by weight, and in particular 0% by weight to 9% by weight.

These additives, fillers and/or reinforcers can be added to the polymer melt individually or in any desired mixtures or a plurality of different mixtures, and additives can specifically be supplied directly in the course of insulation of the polymer (for example via a side unit such as a side extruder) as a pure substance or as a masterbatch in polycarbonate, or else after melting of granules in a compounding step. The additives or mixtures thereof can be added to the polymer melt in solid form, i.e. as a powder, or as a melt. Another method of metered addition is the use of masterbatches or mixtures of masterbatches of the additives or additive mixtures.

Moreover, the polysiloxane-polycarbonate block cocondensate according to the invention can be provided with thermal stabilizers or processing stabilizers. Preferentially suitable are phosphites and phosphonites, and also phosphines. Examples are triphenyl phosphite, diphenyl alkyl phosphite, phenyl dialkyl phosphite, tris(nonylphenyl) phosphite, trilauryl phosphite, trioctadecyl phosphite, distearyl pentaerythrityl diphosphite, tris(2,4-di-tert-butylphenyl) phosphite, diisodecyl pentaerythrityl diphosphite, bis(2,4-di-tert-butylphenyl) pentaerythrityl diphosphite, bis(2,4-di-cumylphenyl) pentaerythrityl diphosphite, bis(2,6-di-tert-butyl-4-methylphenyl) pentaerythrityl diphosphite, diisodecyloxy pentaerythrityl diphosphite, bis(2,4-di-tert-butyl-6-methylphenyl) pentaerythrityl diphosphite, bis(2,4,6-tris(tert-butylphenyl)) pentaerythrityl diphosphite, tristearylsorbitol triphosphite, tetrakis(2,4-di-tert-butylphenyl)4,4'-biphenylene diphosphonite, 6-isooctyloxy-2,4,8,10-tetra-tert-butyl-

12H-dibenzo[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-methyldibenzo[d,g]-1,3,2-dioxaphosphocin, 2,2',2''-nitrido-[triethyl tris(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, bis(2,6-di-tert-butyl-4-methylphenyl) pentaerythrityl diphosphite, triphenylphosphine (TPP), trialkylphenylphosphine, bisdiphenylphosphinoethane or a trinaphthylphosphine. Especially preferred are triphenylphosphine (TPP), Irgafos® 168 (tris(2,4-di-tert-butylphenyl) phosphite) and tris(nonylphenyl) phosphite, or mixtures thereof.

It is additionally possible to use phenolic antioxidants such as alkylated monophenols, alkylated thioalkylphenols, hydroquinones and alkylated hydroquinones. Particular preference is given to using Irganox® 1010 (pentaerythrityl 3-(4-hydroxy-3,5-di-tert-butylphenyl)propionate; CAS: 6683-19-8) and Irganox 1076® (2,6-di-tert-butyl-4-(octadecanoxycarbonyl)phenol).

Suitable UV absorbers are described, for example, in EP 1 308 084 A1, in DE 102007011069 A1, and in DE 10311063 A1.

Particularly suitable ultraviolet absorbers are hydroxybenzotriazols such as 2-(3',5'-bis(1,1-dimethylbenzyl)2'-hydroxyphenyl)benzotriazole (Tinuvin® 234, BASF SE, Ludwigshafen), 2-(2'-hydroxy-5'-(tert-octyl)phenyl)benzotriazole (Tinuvin® 329, BASF SE, Ludwigshafen), 2-(2'-hydroxy-3'-(2-butyl)5'-(tert-butyl)phenyl)benzotriazole (Tinuvin® 350, BASF SE, Ludwigshafen), bis(3-(2H-benzotriazolyl)2-hydroxy-5-tert-octyl)methane, (Tinuvin® 360, BASF SE, Ludwigshafen), 2-(4,6-diphenyl-1,3,5-triazin-2-yl)5-(hexyloxy)phenol (Tinuvin® 1577, BASF SE, Ludwigshafen), and the benzophenones 2,4-dihydroxybenzophenone (Chimasorb® 22, BASF SE, Ludwigshafen) and 2-hydroxy-4-(octyloxy)benzophenone (Chimassorb® 81, BASF SE, Ludwigshafen), 2-cyano-3,3-diphenyl-2-propenoic acid, 2,2-bis[[(2-cyano-1-oxo-3,3-diphenyl-2-propenyl)oxy]methyl]1,3-propanediyl ester (9CI) (Uvinul® 3030, BASF AG Ludwigshafen), 2-[2-hydroxy-4-(2-ethylhexyl)oxy]phenyl-4,6-di(4-phenyl)phenyl-1,3,5-triazine (Tinuvin® 1600, BASF SE, Ludwigshafen) or tetra ethyl-2,2'-(1,4-phenylenedimethylidene)bismalonate (Hostavin® B-Cap, Clariant AG).

It is also possible to use mixtures of these ultraviolet absorbers.

Moreover, the polysiloxane-polycarbonate block cocondensate according to the invention can be provided with demoulding agents. Particularly suitable demoulding agents for the inventive composition are pentaerythrityl tetrastearate (PETS) or glyceryl monostearate (GMS).

In addition, it is also possible to add other polymers to the block cocondensate according to the invention, for example polycarbonate, polyester carbonate, polystyrene, styrene copolymers, aromatic

polyesters such as polyethylene terephthalate (PET), PET-cyclohexanedimethanol copolymer (PETG), polyethylene naphthalate (PEN), polybutylene terephthalate (PBT), cyclic polyolefin, poly- or copolyacrylates and poly- or copolymethacrylate, for example poly- or copolymethylmethacrylates (such as PMMA), and copolymers with styrene, for example transparent polystyrene-acrylonitrile (PSAN), rubber-modified vinyl (co-)polymers, such as acrylonitrile butadiene styrene copolymer, thermoplastic polyurethanes, polymers based on cyclic olefins (e.g. TOPAS®, a commercial product from Ticona).

The block cocondensates according to the invention can be processed in a manner known for thermoplastic polycarbonates to give any desired mouldings.

In this context, the inventive co-condensates can be converted, for example, by hot pressing, spinning, blow-moulding, thermoforming, extrusion or injection moulding to products, mouldings or shaped articles. Also of interest is the use of multilayer systems. The application may coincide with or immediately follow the shaping of the base structure, for example by coextrusion or multicomponent injection moulding. However, application may also be on to the ready-shaped base structure, for example by lamination with a film or by coating with a solution.

Sheets or mouldings composed of base layer and optional top layer/optional top layers (multilayer systems) can be produced by (co)extrusion, direct skinning, direct coating, insert moulding, film insert moulding, or other suitable processes known to those skilled in the art.

Injection moulding processes are known to those skilled in the art and are described, for example, in “Handbuch Spritzgiessen”, Friedrich Johannaber/Walter Michaeli, Munich; Vienna: Hanser, 2001. ISBN 3-446-15632-1 or “Anleitung zum Bau von Spritzgiesswerkzeugen”, Menges/Michaeli/Mohren, Munich; Vienna: Hanser, 1999, ISBN 3-446-21258-2.

Extrusion processes are known to those skilled in the art and are described, for example, for coextrusion, inter alia, in EP-A 0 110 221, EP-A 0 110 238 and EP-A 0 716 919. For details of the adapter and nozzle process, see Johannaber/Ast: “Kunststoff- Maschinenführer”, Hanser Verlag, 2000 and in Gesellschaft Kunststofftechnik: “Coextrudierte Folien und Platten: Zukunftsperspektiven, Anforderungen, Anlagen und Herstellung, Qualitätssicherung”, VDI-Verlag, 1990.

The polysiloxane-polycarbonate block cocondensates according to the invention are usable wherever the known aromatic polycarbonates have been used to date, and wherever good flowability coupled with improved demoulding characteristics and high toughness at low temperatures and improved chemical resistance are additionally required, for example for production of large external motor vehicle parts and switch boxes for exterior use, and of sheets, cavity sheets, electric and electronic parts, and optical memory. For example, the block cocondensates can be used in the IT sector for

computer housings and multimedia housings, mobile phone cases, and in the domestic sector, such as in washing machines, and in the sports sector, for example as a material for helmets.



### Examples

The invention is described in detail hereinafter by working examples, the determination methods described here being employed for all corresponding parameters in the present invention, in the absence of any descriptions to the contrary.

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#### Determination of melt volume flow rate (MVR):

The melt volume flow rate (MVR) is determined to ISO 1133 (at 300°C; 1.2 kg), unless any other conditions have been described.

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#### Determination of solution viscosity ( $\eta_{rel}$ ):

The relative solution viscosity ( $\eta_{rel}$ ; also referred to as  $\eta_{rel}$ ) was determined in dichloromethane at a concentration of 5 g/l at 25°C with an Ubbelohde viscometer.

#### Determination of rearrangement structures (Ia) to (IVa):

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The sample is hydrolysed by means of sodium methoxide under reflux. The corresponding solution is acidified and concentrated to dryness. The drying residue is dissolved in acetonitrile and the phenolic compounds of the formulae (Ia) to (IVa) are determined by means of HPLC with UV detection. The structures (Ia) to (IVa) are unambiguously characterized by means of nuclear magnetic resonance spectroscopy (NMR).

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#### Determination of flowability:

The flowability was determined by measuring the melt viscosity at low shear rate and high shear rate by means of a cone-plate viscosimeter.

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The melt viscosities were determined using a Physica UDS 200 rotation-/oscillation rheometer and a cone-plate geometry. The conical angle was 2° and the cone diameter was 25 mm (MK 216). When evaporation residues were used, samples were first dried in a vacuum drying oven and then compressed by means of a hot press at 230°C to obtain thin films.

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Isothermal frequency spectra of the complex shear modulus  $G^* = G' + iG''$  were recorded at a temperature range of from 300°C to 260°C at intervals of 10 K. Accordingly, the temperature was decreases stepwise by 10 K. The deformation was measured at 10%. In each case a spectrum in the range 75 to 0.08 Hz (20 measure points) was recorded.

Materials used:

PC 1: linear bisphenol A polycarbonate having end groups based on phenol with a solution viscosity of 1.205 and a melt volume flow rate MVR of 59 cm<sup>3</sup>/10 min (measured at 300°C and load 1.2 kg to ISO 1033). This polycarbonate does not contain any additives such as UV stabilizers, demoulding agents or thermal stabilizers. The polycarbonate was prepared by means of a melt transesterification process as described in DE 102008019503.

PC 2 – Oligocarbonate according to DE 19710081

Even though what is used in DE 19710081 is not a polycarbonate prepared on the industrial scale but a specifically prepared oligocarbonate, the block cocondensate is prepared for the comparative examples based on the conditions which were described in DE 19710081.

In a three-way glass flask with stirrer and short-path separator, 150.0 g (0.65 mol) of bisphenol A, 146.6 g (0.68 mol) of diphenyl carbonate are admixed with 0.027 g (0.00004 mol) of tetraphenylphosphonium tetraphenylborate. The mixture is melted at 150°C. At a vacuum of about 100 mbar and 180°C, phenol is distilled off. The vacuum is improved stepwise down to 10 mbar over 90 minutes and the temperature is increased to 250°C. Phenol which forms is distilled off at 250°C and 10 mbar for another 30 minutes. This gives 150 g of an oligocarbonate having a solution viscosity of 1.069.

PC 3: linear bisphenol A polycarbonate having end groups based on phenol with a melt volume flow rate (MVR) of 9.5 cm<sup>3</sup>/10 min (measured at 300°C and load 1.2 kg according to ISO 1033). The polycarbonate does not contain any additives, such as UV-stabilizers, lubricants or thermostabilizers. The polycarbonate was prepared via the interfacial process. Makrolon® 2808 from Bayer MaterialScience is used.

Siloxane component:

The siloxane used is hydroquinone-terminated polydimethylsiloxane of the formula (1) (i.e. R<sup>1</sup> = H, R<sup>2</sup>, R<sup>3</sup> = methyl, p = 0), in which n = 33.7 and m = 3.7, having a hydroxy content of 11.9 mg KOH/g and a viscosity of 358 mPa·s (23°C).

The weight-average molecular weight is M<sub>w</sub> = 9100 g/mol, determined by means of gel permeation chromatography (GPC) with bisphenol A standard; detection was effected by means of an IR detector at 1050 cm<sup>-1</sup>.

The siloxane component can be prepared according to the following procedure:

In a reaction flask equipped with a thermostat heater, stirrer, thermometer, and reflux condenser, 250 g of an  $\alpha,\omega$ -bisacyloxypolydimethylsiloxane, with an average chain length of 31.8 dimethylsiloxy units as determined by  $^{29}\text{Si}$  NMR and 230 mmol of acyloxy terminal groups, is added dropwise over 4 hours to a solution of 35.1 g (150 mmol) bisphenol-A in 50 g xylenes, 25 g acetic acid and 0.5 g of sodium acetate, while heating to a mild reflux at 105 °C. After complete addition the clear solution is stirred for an additional hour. Then the solvents and volatiles are removed by vacuum distillation to 160 °C and 3 mbar pressure. After cooling the crude product is filtered over a 3 micron filter (Seitz K300) to give 236 g (83% theory) of a clear, colorless liquid.

Catalyst:

The catalyst used is tetraphenylphosphonium phenoxide from Rhein Chemie Rheinau GmbH (Mannheim, Germany). The substance is used in the form of a cocrystal with phenol and contains about 70% tetraphenylphosphonium phenoxide. The amounts which follow are based on the substance obtained from Rhein Chemie (as a cocrystal with phenol).

#### **Example 1 (inventive example)**

142.5 g of polycarbonate granules (PC 1), 7.5 g of siloxane (5% by weight) and 0.021 g of tetraphenylphosphonium phenoxide cocrystal (0.1% by weight) are weighed into a 250 ml glass flask with stirrer and short-path separator. The apparatus is evacuated and vented with nitrogen (3x each). The mixture is melted by means of a metal bath preheated at 350°C under standard pressure (under nitrogen) within 10 minutes. A reduced pressure is then applied. The pressure in the apparatus is about 100 mbar. The reaction mixture is kept at this reduced pressure while stirring for 30 minutes. This is followed by venting with nitrogen and removal of the polymer melt. This gives an opaque white powder. The solution viscosity is reported in Table 1.

#### **Example 2 (comparative example)**

The cocondensate is prepared as described in Example 1. In a departure from Example 1, PC 2 is used as the reactant.

**Example 3 (comparative example)**

The cocodensate is prepared from the monomers via the interfacial process with phosgene:

To a nitrogen-purged solution of 4566 g (20 mol) of bisphenol A and 3520 g (88 mol) of sodium hydroxide in 40 liters of water are added 40 l of methylene chloride. Bei einem pH-Wert von 12.5–13.5 und 20°C leitet man 3556 g (40 mol) Phosgen ein. At a pH of 12.5-13.5 and 20 ° C is passed a 3556 g (40 mol) of phosgene. Um den pH-Wert nicht unter 12,5 fallen zu lassen, wurde während der Phosgenierung 30%-ige Natronlauge zugegeben (ca. 7000 g). In order to prevent the pH from falling below 12.5, 30% sodium hydroxide solution was added during the phosgenation (approx. 2500 g). Nach beendeter Phosgenierung und Spülung mit Stickstoff gibt man 258 g (0.85 mol) m-Pentadecylphenol (technische Qualität von Sigma-Aldrich, USA) gelöst in 1 l Dichlormethan hinzu. After the phosgenation and flushing with nitrogen, 65,4 g (0.43 mol) of tert-butylphenol dissolved in 2 l of dichloromethane added. Man rührt für weitere 10 Minuten und gibt 22.6 g (0.2 mol) N-Ethylpiperidin zu und lässt 1 Stunde nachrühren. The mixture is stirred for 10 minutes and 10.96 g (0.10 mol) of N-ethyl piperidine is added. The mixture is stirred for another 1 hour. Die organische Phase wird nach Abtrennen der wässrigen Phase mit Phosphorsäure angesäuert und mit destilliertem Wasser neutral und salzfrei gewaschen. The organic phase is acidified with phosphoric acid after separating off the aqueous phase, and washed to neutral and salt-free with distilled water. Nach Lösungsmittelaustausch gegen Chlorbenzol wird das Produkt bei 290°C und 80 Umdrehungen/min bei 0.1 mbar über einen Ausdampfextruder extrudiert und über einen Granulator granuliert. After replacing the solvent by chlorobenzene, the product at 290 ° C and 80 rev / min is extruded at 0.1 mbar over a vented extruder and pelletized with a pelletizer.

Table 1

	Example 1 (inventive)	Example 2 (comparative)	Example 3 (comparative)
Polycarbonat (reactant)	PC 1	PC 2	-
Structures (Ia) to (IVa) concentration in the polycarbonate (reactant) in ppm <sup>1)</sup>	(Ia) 39ppm (IIa) <10ppm (IIIa) 51 ppm (IVa) 28ppm	(Ia) 26ppm (IIa) <10ppm (IIIa) <10ppm (IVa) <10ppm	(Ia) <10ppm (IIa) <10ppm (IIIa) <10ppm (IVa) <10ppm
Solution viscosity of product	1.315	1.23	1.260
Colour of product	White/light- coloured	Yellow/dark-coloured	White/light-coloured

Inventive example 1 shows that a high molecular weight siloxane-containing block cocondensate simultaneously having a good colour is obtained. The appearance of the product is similar to that of the

product obtained by the interfacial process with phosgene (example 3). In contrast thereto, the product made from the oligocarbonate according to the procedure described in DE 19710081 shows a very poor colour and has relatively a low molecular weight (example 2).

The melt stability of the products was studied by determining the melt volume flow rate (MVR) according to ISO 1133 of the products at various temperatures and after various residence times. The results are summarized in table 2 here below.

Table 2:

Example	300 °C (5 min)	300 °C (20 min)	$\Delta$ MVR (300 °C)	320 °C (5 min)	320 °C (20 min)	$\Delta$ MVR (320 °C)
1 (inventive)	5.9	6.3	0.4	11.7	13.0	1.3
2 (comparative)	20.4	19.5	0.9	32.0	43.3	11.3
3 (comparative)	13.3	14.2	0.9	24.1	28.6	4.5

At 300 °C all products exhibit high melt stability. However, the melt stability of comparative examples 2 and 3 decreases significantly at higher temperatures (320 °C). Surprisingly, the polysiloxane-polycarbonate block cocondensate according to the invention which has been obtained via a melt transesterification process is significantly more stable at higher temperatures compared to the polysiloxane-polycarbonate block cocondensate which has been obtained by the interfacial process with phosgene. Even when compared to the polysiloxane-polycarbonate block cocondensate made from a oligocarbonate according to the procedure described in DE 19710081, the polysiloxane-polycarbonate block cocondensate according to the invention exhibits a high melt stability.

The melt viscosity and flowability under high shear were determined by measuring the melt viscosity using a cone-plate viscosimeter. The samples were measured at 300 °C, 280 °C and 260 °C. Table 3 shows the results at low shear rate (0.08 Hz; corresponding approximately to the zero viscosity) and high shear rate (75 Hz). Due to its low molecular weight, the sample of example 2 was not measured.

Table 3:

Example	300°C	280°C	260°C	Comments
	[Pa·s]	[Pa·s]	[Pa·s]	
1	618	1160	2500	Low shear
	308	451	677	High shear
3	264	427	897	Low shear
	209	281	469	High shear
PC 3	545	975	2070	Low shear
	381	420*	805*	High shear

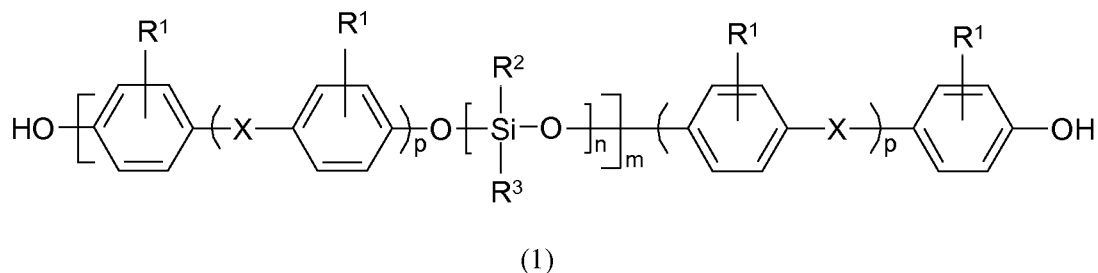
\*extrapolated values

5 It becomes evident from table 3 that the polysiloxane-polycarbonate block cocondensate according to the invention exhibits high flowability under high shear. This can be of advantage for the production of thin-walled components by injection moulding. In contrast thereto, conventional linear polycarbonate shows a higher viscosity under shear. This is surprising because the zero viscosity is lower than in inventive example 1. Comparative example 2 exhibits a lower molecular weight compared to inventive example 1 but under shear the viscosity decrease, in relative terms, is less pronounced than in inventive
   
 10 example 1. Consequently, the polysiloxane-polycarbonate block cocondensate according to the invention which can be produced in a cost-efficient manner shows better properties than the conventional polysiloxane-polycarbonate block cocondensate of example 3.

**Claims**

1. A polysiloxane-polycarbonate block cocondensate derived from

(A) at least one hydroxyaryl-terminated siloxane of the formula (1) (siloxane component)



in which

R<sup>1</sup> is H, Cl, Br or C<sub>1</sub> to C<sub>4</sub>-alkyl,

R<sup>2</sup> and R<sup>3</sup> are the same or different and each independently from one another selected from aryl, C<sub>1</sub> to C<sub>10</sub>-alkyl and C<sub>1</sub> to C<sub>10</sub>-alkylaryl,

X is a single bond, -CO-, -O-, C<sub>1</sub>- to C<sub>6</sub>-alkylene, C<sub>2</sub> to C<sub>5</sub>-alkylidene, C<sub>5</sub> to C<sub>12</sub>-cycloalkylidene or C<sub>6</sub> to C<sub>12</sub>-arylene which may optionally be fused to further aromatic rings containing heteroatoms,

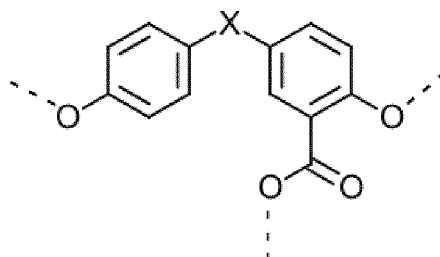
n is a number from 1 to 500,

m is a number from 1 to 10, and

p is 0 or 1;

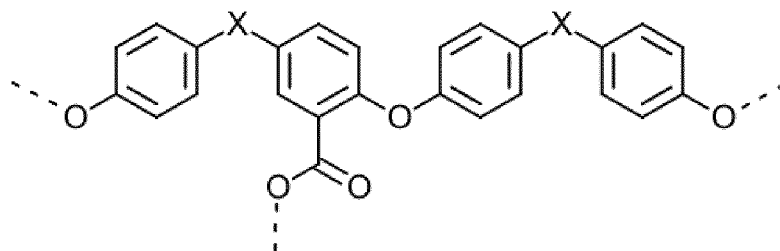
(B) and at least one polycarbonate

wherein the polysiloxane-polycarbonate block cocondensate contains at least one, preferably more than one, of the following structures (I) to (IV):

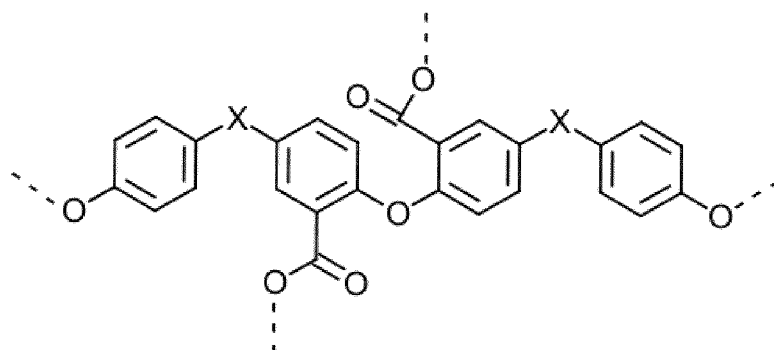


(I)

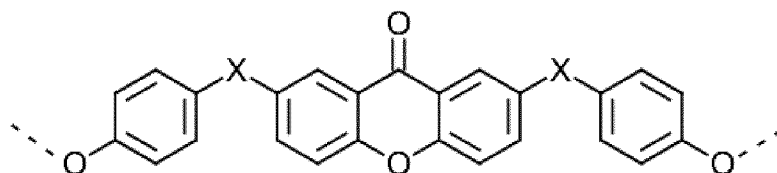
-31-



(II)



(III)



(IV)

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in which

the phenyl rings are unsubstituted or independently mono- or disubstituted by C<sub>1</sub> to C<sub>8</sub>-alkyl and/or halogen,

X is a single bond, C<sub>1</sub> to C<sub>6</sub>-alkylene, C<sub>2</sub> to C<sub>5</sub>-alkylidene or C<sub>5</sub> to C<sub>6</sub>-cycloalkylidene,

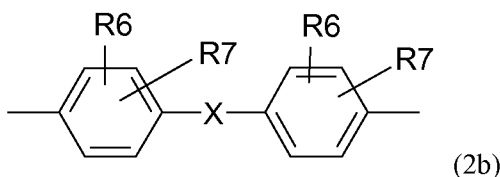
10

the linkages indicated by --- in the structural units (I) to (IV) are each part of a carboxylate group;

and wherein the amount of the structural units (I) to (IV) totals 60 to 1500 ppm, preferably 80 to 850 ppm (determined after hydrolysis, based on the polysiloxane-polycarbonate block cocondensate).



2. The polysiloxane-polycarbonate block cocondensate according to claim 1, characterized in that the amount of the structural units (I) to (IV) totals 80 to 850 ppm (based on the polysiloxane-polycarbonate block cocondensate and determined after hydrolysis).
3. The polysiloxane-polycarbonate block cocondensate according to claim 1 or 2, which has a relative solution viscosity of 1.26 to 1.40, determined in dichloromethane at a concentration of 5 g/l at 25°C using a Ubbelohde viscosimeter.
4. The polysiloxane-polycarbonate block cocondensate according to any of the preceding claims, characterized in that it is derived from a composition comprising the siloxane component of the formula (1) and the polycarbonate, which composition contains the siloxane component in an amount of 2 to 20%, based on the polycarbonate.
5. The polysiloxane-polycarbonate block cocondensate according to any of the preceding claims, which contains structures derived from a diphenol according to general formula (2b):



wherein

R6 and R7 are independently of one another H, C<sub>1</sub>-C<sub>18</sub>-alkyl-, C<sub>1</sub>-C<sub>18</sub>-alkoxy, halogen or an optionally substituted aryl- or aralkyl, and

X is a single bond, -CO-, -O-, C<sub>1</sub>- to C<sub>6</sub>-alkylene, C<sub>2</sub> to C<sub>5</sub>-alkylidene, C<sub>5</sub> to C<sub>12</sub>-cycloalkylidene or C<sub>6</sub> to C<sub>12</sub>-arylene which may optionally be fused to further aromatic rings containing heteroatoms.

6. The polysiloxane-polycarbonate block cocondensate according to any of the preceding claims, characterized in that the hydroxyaryl-terminated siloxane has a weight-average molecular weight of 3000-20 000 g/mol.
7. The polysiloxane-polycarbonate block cocondensate according to any of the preceding claims, characterized in that R<sup>1</sup> is H, p is 1 and X is isopropylidene.
8. The polysiloxane-polycarbonate block cocondensate according to any of the preceding claims, characterized in that R<sup>2</sup> and R<sup>3</sup> are methyl.

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9. The polysiloxane-polycarbonate block cocondensate according to any of the preceding claims, characterized in that n is a number from 10 to 100 and m is a number from 2 to 5.
10. The polysiloxane-polycarbonate block cocondensate according to any of claims 1 to 9 which has a sodium content within the range of from 0.1 ppm to 1000 ppm.
- 5 11. Use of the polysiloxane-polycarbonate block cocondensate according to any of claims 1 to 10 for the production of mouldings.
12. A moulding or extrudate comprising the polysiloxane-polycarbonate block cocondensate according to any of claims 1 to 10.

# INTERNATIONAL SEARCH REPORT

International application No

PCT/EP2014/071299

## A. CLASSIFICATION OF SUBJECT MATTER

INV. C08G64/18 C08G77/448  
ADD.

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)  
C08G

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 practicable, search terms used)

EPO-Internal, WPI Data

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	DE 197 10 081 A1 (BAYER AG [DE]) 17 September 1998 (1998-09-17) cited in the application page 4, line 19 - page 4, line 27; examples 1-9; tables 1, 2, 3 -----	1-12
Y	WO 02/077067 A2 (BAYER AG [DE]; KRATSCHEMER SILKE [DE]; HUCKS UWE [DE]; PREIN MICHAEL [B]) 3 October 2002 (2002-10-03) page 10, line 13 - page 13, line 1; figure 2; examples 2-8; table 1 -----	1-12
A	US 2011/040066 A1 (WEHRMANN ROLF [DE] ET AL) 17 February 2011 (2011-02-17) paragraph [0129] - paragraph [0145]; examples PC1, PC3, PC5, PC7; table 1 ----- -/-	1-12



Further documents are listed in the continuation of Box C.



See patent family annex.

\* Special categories of cited documents :

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"E" earlier application or patent 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

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Date of the actual completion of the international search

18 December 2014

Date of mailing of the international search report

08/01/2015

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## INTERNATIONAL SEARCH REPORT

International application No

PCT/EP2014/071299

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
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A	EP 0 414 071 A2 (BAYER AG [DE]) 27 February 1991 (1991-02-27) examples 1-3; table 1 -----	1-12

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Information on patent family members

International application No

PCT/EP2014/071299

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