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(54) Title: POLYSULFONE COMPOSITIONS INCLUDING A POLYCARBONATE-POLYSILOXANE COPOLYMER

(57) Abstract: A polymer composition includes greater than 30 wt.% of a polysulfone (PSU), based on the total weight of the polymer composition, at least one polycarbonate-polysiloxane copolymer (SiPC), optionally, at least one poly(ether imide) (PEI), and optionally, at least one reinforcing filler. Methods of making the polymer composition and shaped articles including the polymer composition are also described.



Polysulfone Compositions Including
a Polycarbonate-Polysiloxane Copolymer

CROSS REFERENCE TO RELATED APPLICATIONS

This application claims priority to copending U.S. provisional application 62/585,004 filed on November 13, 2017 to Xia et al., entitled "Polysulfone Compositions Including a Polycarbonate-Polysiloxane Copolymer," incorporated
5 herein by reference.

FIELD OF THE INVENTION

The present invention relates to polymer compositions including greater than 30 wt.% of at least one polysulfone (PSU), based on the total weight of the polymer composition, at least one polycarbonate-polysiloxane copolymer (SiPC),
10 optionally, at least one poly(ether imide) (PEI), and optionally, at least one reinforcing filler, methods of making the polymer composition, and shaped articles including the polymer composition. The polymer composition is particularly suitable for use in parts for mobile electronic devices.

BACKGROUND

15 Mobile electronic devices are getting smaller and lighter for portability and convenience; however, they still need to possess a certain structural strength, so that they are not damaged by normal handling and occasional drops. Thus, usually built into such devices are structural parts whose primary function is to provide strength and rigidity to the device, and possibly also mounting places for
20 various internal components of the device and/or part or all of the mobile electronic device case (outer housing). While in the past, low density metals such as magnesium or aluminum were the materials of choice for such structural parts, synthetic resins have progressively at least partially replaced such metals for reasons of cost reduction, design flexibility, weight reduction, and aesthetics.
25 Plastic parts of electronic devices are hence made of materials that are easy to process into various and complex shapes, are able to withstand the rigors of frequent use, including outstanding mechanical properties, and which can meet challenging aesthetic demands while not interfering with their intended operability.

30 Another important requirement for plastic materials used as metal replacements in applications such as mobile electronics is flame resistance. Demanding industry legislation and recent incidences of mobile electronic

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device overheating has increased the need for halogen-free materials with inherent flame resistance. Specifically, there is a need for high performance plastic materials which, in addition to possessing good mechanical properties, exhibit excellent flame resistance.

5 DETAILED DESCRIPTION OF PREFERRED EMBODIMENTS

The present invention is generally directed to a polymer composition comprising greater than 30 wt.% of at least one polysulfone (PSU), based on the total weight of the polymer composition, at least one polycarbonate-polysiloxane copolymer (SiPC), optionally, at least one poly(ether imide) (PEI), and
10 optionally, at least one reinforcing filler.

The polymer compositions described herein exhibit significantly improved flame resistance. In particular, the present polymer compositions unexpectedly achieve “V-0” UL94 flame resistance ratings. In addition, in some cases, the polymer compositions also exhibited markedly improved toughness as compared
15 with, for example, the toughness observed for compositions of PSU or SiPC copolymer alone.

Accordingly, in some embodiments, the polymer composition exhibits a UL94 flame resistance rating of “V-0” using a 1.6 mm-thick sample.

In some embodiments, the polymer composition exhibits a notched Izod
20 impact resistance of 750 to 1000 J/M, preferably 800 to 1000 J/M, as measured according to ASTM D256.

In certain embodiments, the polymer composition exhibits a UL94 flame resistance rating of “V-0” using a 1.6 mm-thick sample and a notched Izod impact resistance of 750 to 1000 J/M, preferably 800 to 1000 J/M, as measured
25 according to ASTM D256.

In some embodiments, the polymer composition does not comprise a polymer other than the at least one PSU, the at least one SiPC copolymer, and the optional PEI or comprise such a polymer in an amount not exceeding 1 wt.%, for example not exceeding 0.5 wt.% or 0.3 wt.%.

30 In some aspects, the polymer composition comprises less than 0.5 wt.% of a poly(carbonate-arylate ester), a poly(carbonate-arylate ester-siloxane), or a combination thereof. Preferably the polymer composition does not comprise any of a poly(carbonate-arylate ester), a poly(carbonate-arylate ester-siloxane), or a combination thereof.

35 In some aspects, the polymer composition includes less than 10 wt.%, preferably less than 5 wt.%, preferably less than 1 wt.% of polyphenylsulfone

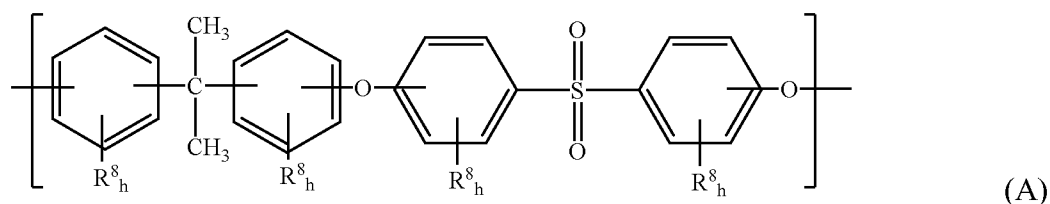
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(PPSU) polymer. Most preferably, the polymer composition does not comprise any PPSU polymer.

In some embodiments, the amount of the polysulfone (PSU) ranges from 35 wt. % to 95 wt. %, preferably 40 wt.% to 90 wt.%, and the amount of the at least one polycarbonate-polysiloxane copolymer (SiPC) ranges from 5 wt.% to 65 wt.%, preferably 10 wt.% to 60 wt.%, each based on the total weight of the polymer composition.

Polysulfone (PSU)

As used herein, a “polysulfone (PSU)” any polymer of which more than 50 mol % of the recurring units are recurring units of formula (A) :



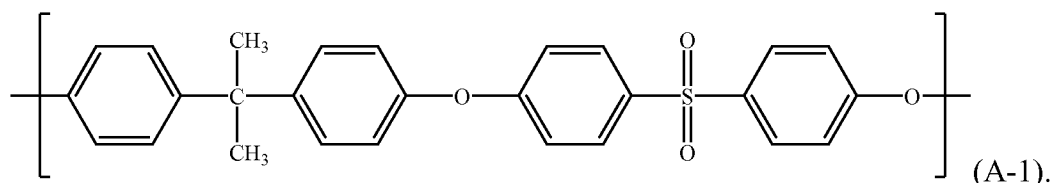
where :

each R^8 is independently selected from the group consisting of a halogen, an alkyl, an alkenyl, an alkynyl, an aryl, an ether, a thioether, a carboxylic acid, an ester, an amide, an imide, an alkali or alkaline earth metal sulfonate, an alkyl sulfonate, an alkali or alkaline earth metal phosphonate, an alkyl phosphonate, an amine, and a quaternary ammonium; and

each h is independently selected from the group consisting of integers ranging from 0 to 4.

Preferably at least 60 mol %, 70 mol %, 80 mol %, 90 mol %, 95 mol %, 99 mol %, of recurring units in the PSU are recurring units of formula (A).

Preferably, the PSU includes more than 50 mol % of recurring units of formula (A-1) :



Preferably at least 60 mol %, 70 mol %, 80 mol %, 90 mol %, 95 mol %, 99 mol %, of the recurring units in the PSU are recurring units of formula (A-1).

PSU can be prepared by known methods and is available as UDEL[®] PSU from Solvay Specialty Polymers USA, L.L.C.

The polymer composition includes greater than 30 wt. % and less than 100 wt.% of the PSU, based on the total weight of the polymer composition. In

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some embodiments, the polymer composition includes at least 35 wt.%, 40 wt.%, 45 wt.%, 50 wt.%, 55 wt.% of the PSU, based on the total weight of the polymer composition. In some embodiments, the amount of PSU in the polymer composition ranges from 35 wt.% to 95 wt.%, preferably 35 wt.% to 90 wt.%, 40 wt.% to 90 wt.%, 45 wt.% to 90 wt.%, 50 wt.% to 90 wt.%, 55 wt.% to 90 wt.% of the polymer composition based on the total weight of the polymer composition. In some aspects, the amount of PSU in the polymer composition ranges from greater than 30 wt.% to 55 wt.%, preferably 35 wt.% to 55 wt.%, 40 wt.% to 55 wt.%, of the polymer composition based on the total weight of the polymer composition.

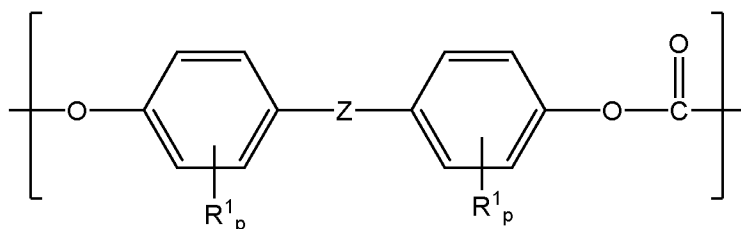
In some embodiments, the PSU is the only poly(aryl ether sulfone) (PAES) polymer in the polymer composition.

Polycarbonate-Polysiloxane Copolymer (SiPC copolymer)

The polycarbonate-polysiloxane copolymer ("SiPC copolymer") is a copolymer comprising polycarbonate repeat units and polysiloxane repeat units, where the mol% of polycarbonate repeat units and polysiloxane repeat units together represents more than 50 mol % of the repeat units of the copolymer.

Preferably, the polycarbonate repeat units are repeat units of formula (B) :

i) polycarbonate repeat units of formula (B) :



(B)

where :

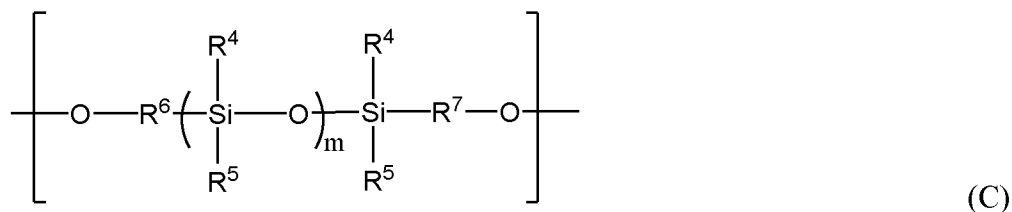
Z is selected from the group consisting of a bond, an alkylene group having 1 to 8 carbon atoms, an alkylidene group having 2 to 8 carbon atoms, a cycloalkylene group having 5 to 15 carbon atoms, a cycloalkylidene group having 5 to 15 carbon atoms, -S-, -SO-, -SO₂-, -O- and -CO-. Preferably Z is a group -C(R²)(R³)-, where R² and R³ are independently selected from a hydrogen and an alkyl group having 1-4 carbon atoms. Most preferably, R² and R³ are methyl groups;

each R¹ is independently selected from the group consisting of a halogen, an alkyl group having 1-20 carbon atoms, and an aryl group; and

each p is independently selected from the group consisting of integers ranging from 0 to 4; and

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ii) polysiloxane repeat units of formula (C) :



where :

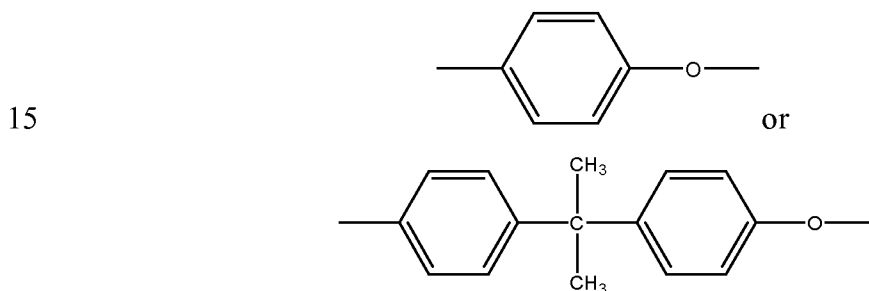
each R^4 and R^5 are independently selected from the group consisting of a hydrogen, a halogen, a hydrocarbyl, and a halogen-substituted hydrocarbyl.

Preferably, R^4 is a methyl group and R^5 is a methyl or phenyl group;

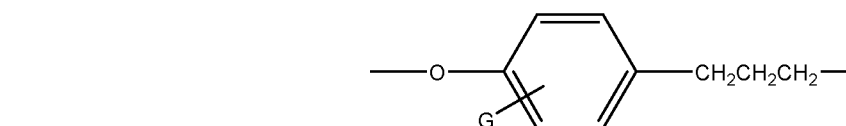
R^6 and R^7 are independently selected from the group consisting of organic residues having an aromatic nucleus,

m is selected from integers ranging from 5 to more than 100. Preferably, m is selected from the group consisting of integers ranging from 5 to 1000, preferably from 5 to 200.

In some embodiments, R^6 and R^7 are independently selected from the group consisting of a 3-(o-hydroxyphenyl)propylene group, a 2-(p-hydroxyphenyl)ethylene group, and groups represented by the formulas :

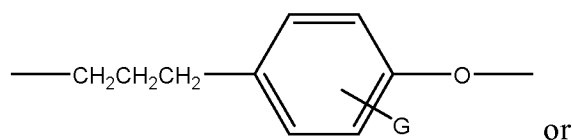


In some embodiments, the moiety $-\text{O}-\text{R}^6-$ in Formula (C) is a group of formula :

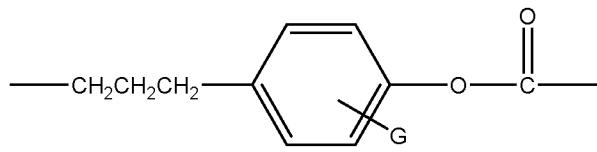


where G is selected from the group consisting of a hydrogen, a hydrocarbyl, and a halogen-substituted hydrocarbyl (preferably methoxy), and

the moiety $-\text{R}^7-\text{O}-$ in formula (C) is a group of formula :



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where G is defined as above.

Preferably at least 75 mol %, 85 mol %, 95 mol %, 99 mol %, of repeat units in the SiPC copolymer are the polycarbonate and polysiloxane repeat units.

5 The polysiloxane repeat unit is preferably a polydimethylsiloxane repeat unit.

In some embodiments, the ratio of the weight of the polycarbonate repeat units to the weight of the polysiloxane repeat units ranges from 0.05 to 3. The SiPC copolymer preferably includes 0.5 % to 30 %, preferably 1 % to 30 %, preferably 4 % to 8 % by weight, of the polysiloxane repeat units.

10 SiPC copolymers, their precursors, and methods for their preparation are described, for example, in U.S. Patent Nos. 3,189,662, 5,502,134, 6,072,011, and 8,981,017, each of which is incorporated by reference herein in its entirety.

The SiPC copolymer may be a random or a block copolymer, preferably a block copolymer.

15 SiPC copolymers are available, for example, as TARFLON[®] NEO from Idemitsu Kosan Co., Ltd, and as LEXAN[®] EXL from Saudi Basic Industries Corporation (SABIC).

The polymer composition includes less than 70 wt.% of the SiPC copolymer, based on the total weight of the polymer composition.

20 In some embodiments, the polymer composition includes 65 wt.% or less, 60 wt.% or less, 55 wt.% or less, 50 wt.% or less, 45 wt.% or less, 40 wt.% or less, 35 wt.% or less of the SiPC copolymer, based on the total weight of the polymer composition. In some embodiments, the polymer composition includes less than 35 wt.% of the SiPC copolymer, based on the total weight of the polymer composition. In some embodiments, the amount of the SiPC copolymer in the polymer composition ranges from 10 wt.% to 65 wt.%, preferably 10 wt.% to 60 wt.%, 10 wt.% to 55 wt.%, 10 wt.% to 50 wt.%, 10 wt.% to 45 wt.% of the polymer composition based on the total weight of the polymer composition.

30 In some embodiments, the amount of the SiPC copolymer ranges from 10 wt.% to 65 wt.%, preferably 10 wt.% to 60 wt.%, 10 wt.% to 55 wt.%, 10 wt.% to 50 wt.%, 10 wt.% to 45 wt.%, based on the combined weight of PSU and the SiPC copolymer. In some aspects, the amount of the SiPC copolymer ranges from 45 wt.% to less than 70 wt.%, preferably from 45 wt.% to 65 wt.%,

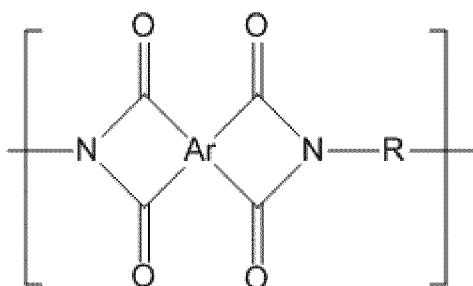
45 wt.% to 60 wt.%, based on the combined weight of PSU and the SiPC copolymer.

Optional Polyetherimide (PEI)

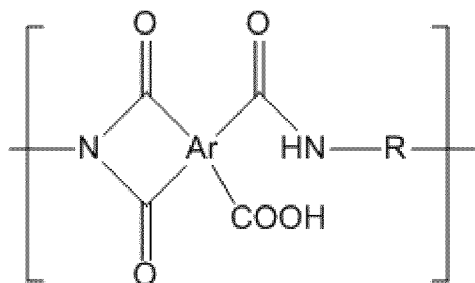
In some embodiments, the polymer composition includes at least one poly(ether imide) (PEI), preferably in an amount ranging from 1 wt.% to 25 wt.%, preferably from 1 wt.% to 20 wt.%, preferably from 1 wt.% to 15 wt.% of the polymer composition.

As used herein, a poly(ether imide) (PEI) denotes any polymer comprising at least 50 mol %, based on the total number of moles in the polymer, of recurring units (R_{PEI}) comprising at least one aromatic ring, at least one imide group, as such and/or in its amic acid form, and at least one ether group. Recurring units (R_{PEI}) may optionally further comprise at least one amide group which is not included in the amic acid form of an imide group.

According to an embodiment, the recurring units (R_{PEI}) are selected from the group consisting of following formulas (I), (II), (III), (IV), (V) and mixtures thereof :

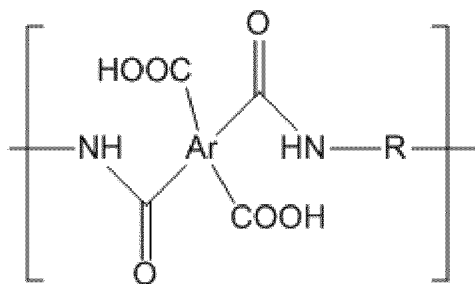


(I)

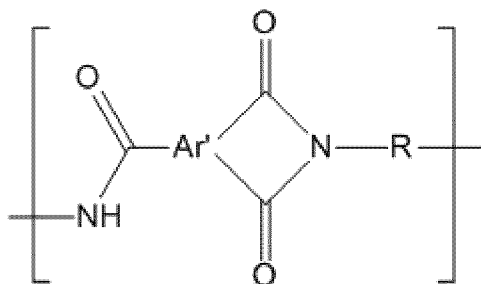


(II)

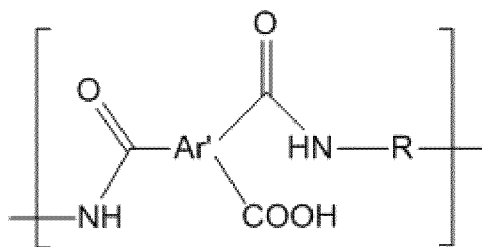
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(III)



(IV)

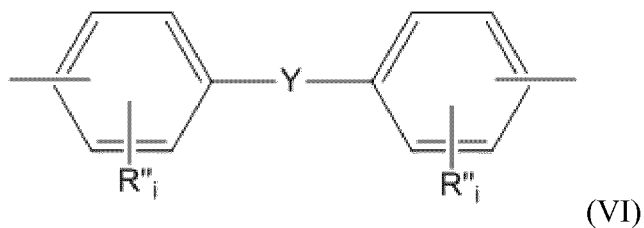


(V)

where

- 5 - Ar is a tetravalent aromatic moiety and is selected from the group consisting of a substituted or unsubstituted, saturated, unsaturated or aromatic monocyclic and polycyclic group having 5 to 50 carbon atoms;
- Ar' is a trivalent aromatic moiety and is selected from the group consisting of a substituted, unsubstituted, saturated, unsaturated, aromatic
- 10 monocyclic and aromatic polycyclic group having from 5 to 50 C atoms; and
- R is selected from the group consisting of substituted and unsubstituted divalent organic radicals, for example selected from the group consisting of
 - (a) aromatic hydrocarbon radicals having 6 to 20 carbon atoms and halogenated derivatives thereof ;
 - 15 (b) straight or branched chain alkylene radicals having 2 to 20 carbon atoms ;
 - (c) cycloalkylene radicals having 3 to 20 carbon atoms, and
 - (d) divalent radicals of formula (VI) :

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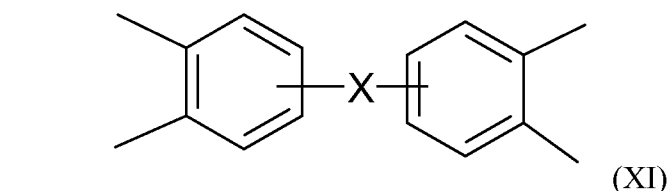
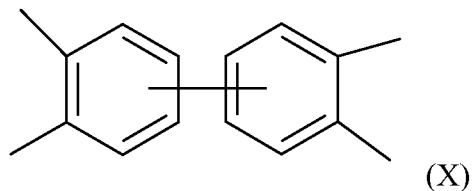
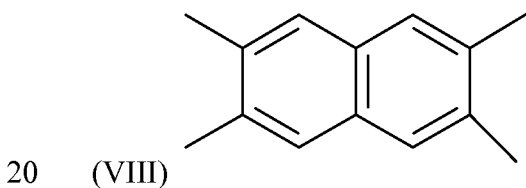
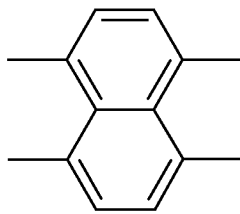
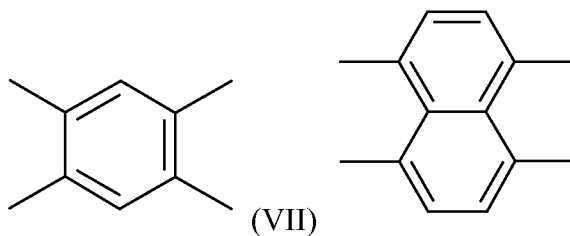
where

- Y is selected from the group consisting of alkylenes of 1 to 6 carbon atoms, for example $-\text{C}(\text{CH}_3)_2$ and $-\text{C}_n\text{H}_{2n}-$ (n being an integer from 1 to 6);
 5 perfluoroalkylenes of 1 to 6 carbon atoms, for example $-\text{C}(\text{CF}_3)_2$ and $-\text{C}_n\text{F}_{2n}-$ (n being an integer from 1 to 6); cycloalkylenes of 4 to 8 carbon atoms; alkylenes of 1 to 6 carbon atoms; cycloalkylidenes of 4 to 8 carbon atoms; $-\text{O}-$; $-\text{S}-$; $-\text{C}(\text{O})-$; $-\text{SO}_2-$; $-\text{SO}-$, and

- R'' is selected from the group consisting of hydrogen, halogen, alkyl,
 10 alkenyl, alkynyl, aryl, ether, thioether, carboxylic acid, ester, amide, imide, alkali earth metal sulfonate, alkaline earth metal sulfonate, alkyl sulfonate, alkali earth metal phosphonate, alkaline earth metal phosphonate, alkyl phosphonate, amine and quaternary ammonium and

- i, for each R'' , is independently zero or an integer ranging from 1 to 4,
 15 with the proviso that at least one of Ar, Ar' and R comprise at least one ether group and that the ether group is present in the polymer chain backbone.

According to an embodiment, Ar is selected from the group consisting of formulas :

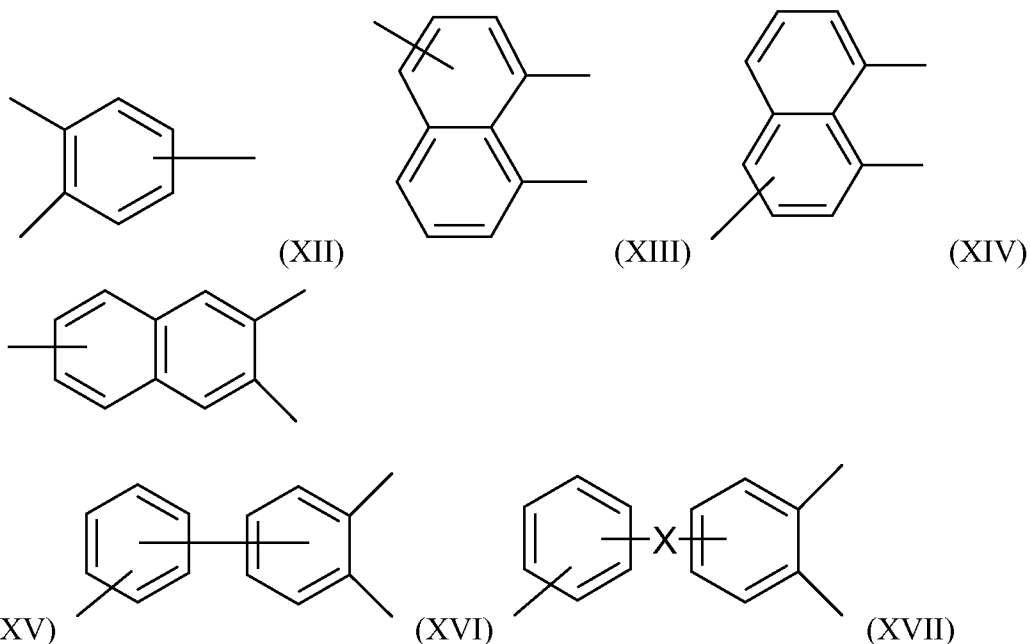


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where

X is a divalent moiety, having divalent bonds in the 3,3', 3,4', 4,3'' or the 4,4' positions and is selected from the group consisting of alkylenes of 1 to 6 carbon atoms, for example $-\text{C}(\text{CH}_3)_2$ and $-\text{C}_n\text{H}_{2n}-$ (n being an integer from 1 to 6); perfluoroalkylenes of 1 to 6 carbon atoms, for example $-\text{C}(\text{CF}_3)_2$ and $-\text{C}_n\text{F}_{2n}-$ (n being an integer from 1 to 6); cycloalkylenes of 4 to 8 carbon atoms; alkylidenes of 1 to 6 carbon atoms; cycloalkylidenes of 4 to 8 carbon atoms; $-\text{O}-$; $-\text{S}-$; $-\text{C}(\text{O})-$; $-\text{SO}_2-$; $-\text{SO}-$; or X is a group of the formula $-\text{O}-\text{Ar}''-\text{O}-$, wherein Ar'' is an aromatic moiety selected from the group consisting of a substituted or unsubstituted, saturated, unsaturated or aromatic monocyclic and polycyclic group having 5 to 50 carbon atoms.

According to an embodiment, Ar' is selected from the group consisting of formulas :



where

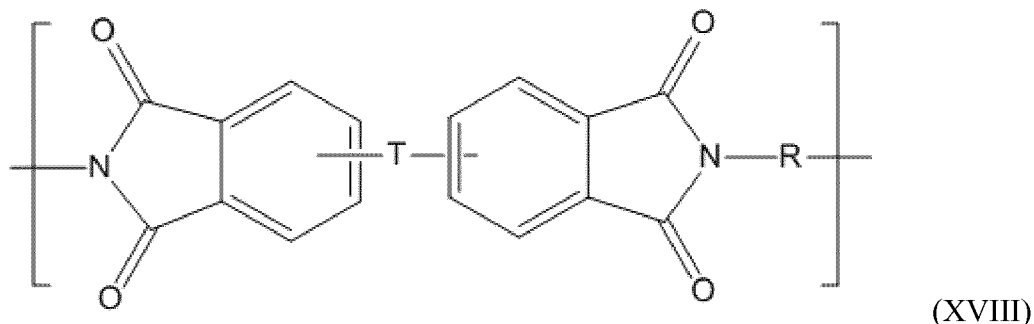
X is a divalent moiety, having divalent bonds in the 3,3', 3,4', 4,3'' or the 4,4' positions and is selected from the group consisting of alkylenes of 1 to 6 carbon atoms, for example $-\text{C}(\text{CH}_3)_2$ and $-\text{C}_n\text{H}_{2n}-$ (n being an integer from 1 to 6); perfluoroalkylenes of 1 to 6 carbon atoms, for example $-\text{C}(\text{CF}_3)_2$ and $-\text{C}_n\text{F}_{2n}-$ (n being an integer from 1 to 6); cycloalkylenes of 4 to 8 carbon atoms; alkylidenes of 1 to 6 carbon atoms; cycloalkylidenes of 4 to 8 carbon atoms; $-\text{O}-$; $-\text{S}-$; $-\text{C}(\text{O})-$; $-\text{SO}_2-$; $-\text{SO}-$; or X is a group of the formula $-\text{O}-\text{Ar}''-\text{O}-$, wherein Ar'' is an aromatic moiety selected from the group consisting of a

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substituted or unsubstituted, saturated, unsaturated or aromatic monocyclic and polycyclic group having 5 to 50 carbon atoms.

According to an embodiment of the present disclosure, at least 50 mol. %, at least 60 mol. %, at least 70 mol. %, at least 80 mol. %, at least 90 mol. %, at least 95 mol. %, at least 99 mol. % or all of the recurring units in the PEI are recurring units (R_{PEI}) of formulas (I), (II), (III), (IV), (V) and/or mixtures thereof, as defined above.

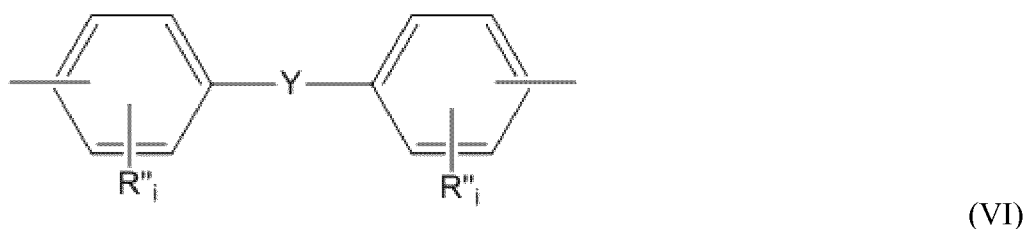
According to an embodiment, a poly(ether imide) (PEI) denotes any polymer comprising at least 50 mol.%, based on the total number of moles in the polymer, of recurring units (R_{PEI}) of formula (VII) :



where

- R is selected from the group consisting of substituted and unsubstituted divalent organic radicals, for example selected from the group consisting of

- (a) aromatic hydrocarbon radicals having 6 to 20 carbon atoms and halogenated derivatives thereof ;
- (b) straight or branched chain alkylene radicals having 2 to 20 carbon atoms ;
- (c) cycloalkylene radicals having 3 to 20 carbon atoms, and
- (d) divalent radicals of formula (VI) :



where

- Y is selected from the group consisting of alkylenes of 1 to 6 carbon atoms, for example $-C(CH_3)_2$ and $-C_nH_{2n}-$ (n being an integer from 1 to 6);
- perfluoroalkylenes of 1 to 6 carbon atoms, for example $-C(CF_3)_2$ and $-C_nF_{2n}-$

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(n being an integer from 1 to 6) ; cycloalkylenes of 4 to 8 carbon atoms ;
alkylidenes of 1 to 6 carbon atoms ; cycloalkylidenes of 4 to 8 carbon atoms ;
-O- ; -S- ; -C(O)- ; -SO₂- ; -SO-, and

- R'' is selected from the group consisting of hydrogen, halogen, alkyl,
5 alkenyl, alkynyl, aryl, ether, thioether, carboxylic acid, ester, amide, imide, alkali
earth metal sulfonate, alkaline earth metal sulfonate, alkyl sulfonate, alkali earth
metal phosphonate, alkaline earth metal phosphonate, alkyl phosphonate, amine
and quaternary ammonium and

- i, for each R'', is independently zero or an integer ranging from 1 to 4,
10 with the proviso that at least one of Ar, Ar' and R comprise at least one
ether group and that the ether group is present in the polymer chain backbone.

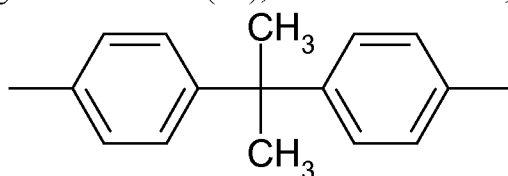
- T can either be

- O - or - O - Ar'' - O -

wherein the divalent bonds of the - O - or the - O - Ar'' - O - group are in the
15 3,3', 3,4', 4,3', or the 4,4' positions,

wherein Ar'' is a aromatic moiety selected from the group consisting of a
substituted or unsubstituted, saturated, unsaturated or aromatic monocyclic and
polycyclic group having 5 to 50 carbon atoms, for example a substituted or
unsubstituted phenylene, a substituted or unsubstituted biphenyl group, a
20 substituted or unsubstituted naphthalene group or a moiety comprising two
substituted or unsubstituted phenylene.

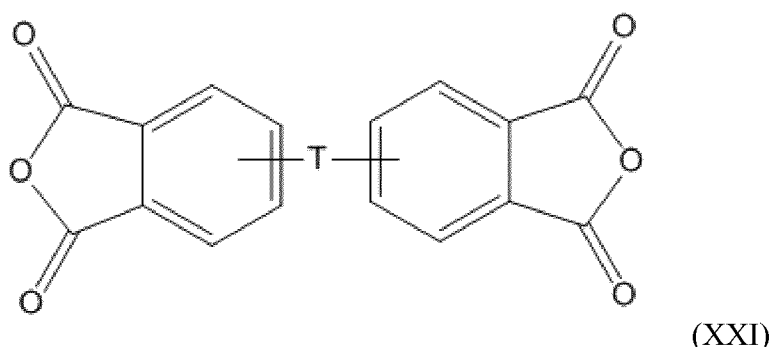
According to an embodiment of the present disclosure, Ar'' is of the
general formula (VI), as detailed above ; for example, Ar'' is of formula (XIX) :



(XIX).

25 The polyetherimides (PEI) of the present invention may be prepared by any
of the methods well-known to those skilled in the art including the reaction of a
diamino compound of the formula H₂N-R-NH₂ (XX), where R is as defined
before, with any aromatic bis(ether anhydride)s of the formula (XXI) :

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where T as defined before.

In general, the preparation can be carried out in solvents, e.g.,
 o-dichlorobenzene, m-cresol/toluene, N,N-dimethylacetamide, at temperatures
 5 ranging from 20°C to 250°C.

Alternatively, these polyetherimides can be prepared by melt
 polymerization of any dianhydrides of formula (XXI) with any diamino
 compound of formula (XX) while heating the mixture of the ingredients at
 elevated temperatures with concurrent intermixing.

10 The aromatic bis(ether anhydride)s of formula (XXI) include, for example :

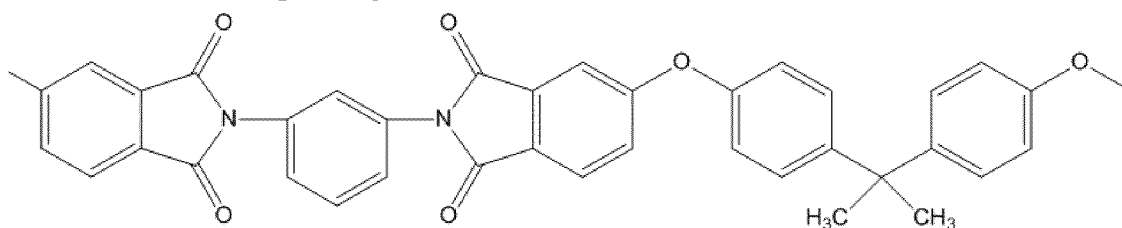
2,2-bis[4-(2,3-dicarboxyphenoxy)phenyl]propane dianhydride ;
 4,4'-bis(2,3-dicarboxyphenoxy)diphenyl ether dianhydride ;
 1,3-bis(2,3-dicarboxyphenoxy)benzene dianhydride ;
 4,4'-bis(2,3-dicarboxyphenoxy)diphenyl sulfide dianhydride ;
 15 1,4-bis(2,3-dicarboxyphenoxy)benzene dianhydride ;
 4,4'-bis(2,3-dicarboxyphenoxy)benzophenone dianhydride ;
 4,4'-bis(2,3-dicarboxyphenoxy)diphenyl sulfone dianhydride ;
 2,2-bis[4 (3,4-dicarboxyphenoxy)phenyl]propane dianhydride ;
 4,4'-bis(3,4-dicarboxyphenoxy)diphenyl ether dianhydride ;
 20 4,4'-bis(3,4-dicarboxyphenoxy)diphenyl sulfide dianhydride ;
 1,3-bis(3,4-dicarboxyphenoxy)benzene dianhydride ;
 1,4-bis(3,4-dicarboxyphenoxy)benzene dianhydride ;
 4,4'-bis(3,4-dicarboxyphenoxy)benzophenone dianhydride ;
 4-(2,3-dicarboxyphenoxy)-4'-(3,4-dicarboxyphenoxy)diphenyl-2,2-propane
 25 dianhydride ; and mixtures of such dianhydrides.

The organic diamines of formula (XX) are chosen from the group
 consisting of m-phenylenediamine, p-phenylenediamine, 2,2-bis(p-
 aminophenyl)propane, 4,4'-diaminodiphenyl-methane, 4,4'-diaminodiphenyl
 sulfide, 4,4'-diamino diphenyl sulfone, 4,4'-diaminodiphenyl ether,

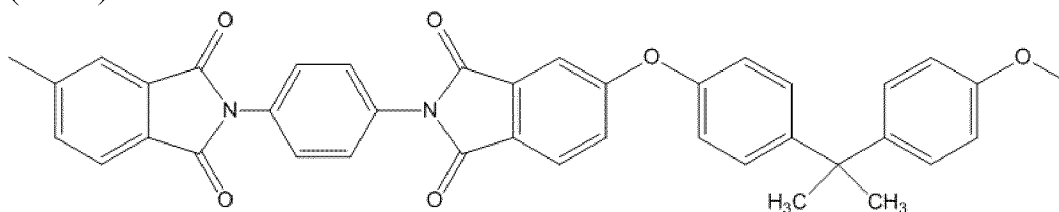
- 14 -

1,5-diaminonaphthalene, 3,3'-dimethylbenzidine, 3,3'-dimethoxybenzidine, and mixtures thereof; preferably, the organic diamines of formula (XX) are chosen from the group consisting of m-phenylenediamine and p-phenylenediamine and mixture thereof.

- 5 According to an embodiment, a poly(ether imide) (PEI) denotes any polymer comprising at least 50 mol.%, based on the total number of moles in the polymer, of recurring units (R_{PEI}) of formulas (XXIII) or (XXIV), in imide forms, or their corresponding amic acid forms and mixtures thereof :



10 (XXIII)



(XXIV).

- Preferably at least 60 mol. %, at least 70 mol. %, at least 80 mol. %, at least 90 mol. %, at least 95 mol. %, at least 99 mol. % of the recurring units in the PEI are recurring units (R_{PEI}) of formulas (XXIII) or (XXIV), in imide forms, or their corresponding amic acid forms and mixtures thereof.
- 15

Such aromatic polyimides are notably commercially available from Sabic Innovative Plastics as ULTEM[®] polyetherimides.

Optional Reinforcing Fillers

The polymer composition optionally includes at least one reinforcing filler such as a fibrous or particulate filler. A fibrous reinforcing filler is a material having length, width and thickness, wherein the average length is significantly
5 larger than both the width and thickness. Preferably, such a material has an aspect ratio, defined as the average ratio between the length and the smallest of the width and thickness of at least 5. Preferably, the aspect ratio of the reinforcing fibers is at least 10, more preferably at least 20, still more preferably at least 50. The particulate fillers have an aspect ratio of at most 5, preferably at
10 most 2.

Preferably, the reinforcing filler is selected from mineral fillers, such as talc, mica, kaolin, calcium carbonate, calcium silicate, magnesium carbonate, boron nitride; glass fibers; carbon fibers, boron carbide fibers; wollastonite; silicon carbide fibers; boron fibers, graphene, carbon nanotubes (CNT), and the
15 like. Most preferably, the reinforcing filler is glass fiber, preferably chopped glass fiber, or carbon fiber, preferably chopped carbon fibers.

Glass fibers are silica-based glass compounds that contain several metal oxides which can be tailored to create different types of glass. The main oxide is silica in the form of silica sand; the other oxides such as calcium, sodium and
20 aluminum are incorporated to reduce the melting temperature and impede crystallization. Glass fibers have a round cross-section or a non-circular cross-section (so called "flat glass fibers"), including oval, elliptical or rectangular. The glass fibers may be added as endless fibers or as chopped glass fibers. The glass fibers have generally an equivalent diameter of 5 to 20 preferably of 5 to
25 15 μm and more preferably of 5 to 10 μm . All glass fiber types, such as A, C, D, E, M, S, R, T glass fibers (as described in chapter 5.2.3, pages 43-48 of *Additives for Plastics Handbook*, 2nd ed, John Murphy, which is incorporated herein by reference), or any mixtures thereof or mixtures thereof may be used. For
30 example, R, S and T glass fibers are high modulus glass fibers that have typically an elastic modulus of at least 76, preferably at least 78, more preferably at least 80, and most preferably at least 82 GPa as measured according to ASTM D2343.

E, R, S and T glass fibers are well known in the art. They are notably described in *Fiberglass and Glass Technology*, Wallenberger, Frederick T.; Bingham, Paul A. (Eds.), 2010, XIV, chapter 5, pages 197 – 225, which is
35 incorporated herein by reference. R, S and T glass fibers are composed essentially of oxides of silicon, aluminum and magnesium. In particular, those

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glass fibers comprise typically from 62-75 wt. % of SiO_2 , from 16-28 wt. % of Al_2O_3 and from 5-14 wt. % of MgO . Different than the regular E-glass fibers widely used in polymer compositions, R, S and T glass fibers comprise less than 10 wt. % of CaO .

5 The fibrous filler, in particular the glass fibers, may have a cross-sectional longest diameter of at least 15 μm , preferably at least 20 μm , more preferably at least 22 μm , still more preferably at least 25 μm . It is advantageously of at most 40 μm , preferably at most 35 μm , more preferably at most 32 μm , still more preferably at most 30 μm . Excellent results were obtained when the cross-
10 sectional longest diameter was in the range of 15 to 35 preferably of 20 to 30 μm and more preferably of 25 to 29 μm .

 The fibrous filler, in particular the glass fibers may have a cross-sectional shortest diameter of at least 4 μm , preferably at least 5 μm , more preferably at least 6 μm , still more preferably at least 7 μm . It is advantageously of at most
15 25 μm , preferably at most 20 μm , more preferably at most 17 μm , still more preferably at most 15 μm . Excellent results were obtained when the cross-sectional shortest diameter was in the range of 5 to 20 preferably of 5 to 15 μm and more preferably of 7 to 11 μm .

 The fibrous filler, in particular the glass fibers, may have an aspect ratio of
20 at least 2, preferably at least 2.2, more preferably at least 2.4, still more preferably at least 3. The aspect ratio is defined as a ratio of the longest diameter in the cross-section of the glass fiber to the shortest diameter thereof. Also, the aspect ratio of the glass fibers is of at most 8, preferably at most 6, more preferably of at most 4. Excellent results were obtained when said ratio was of
25 from about 2 to about 6, and preferably, from about 2.2 to about 4.

 The shape of the cross-section of the glass fiber, its length, its cross-sectional diameter and its aspect ratio can be easily determined using optical microscopy.

 The amount of the reinforcing filler may range in the case of particulate
30 fillers, from 1 wt. % to 40 wt. %, preferably from 5 wt. % to 35 wt. % and most preferably from 10 wt. % to 30 wt. %, and in the case of fibrous fillers from 5 wt. % to 50 wt. %, preferably from 10 wt. % to 40 wt. %, and most preferably from 15 wt. % to 30 wt. % based on the total weight of the polymer composition. Preferably, the polymer composition includes about 25 to about 35 wt. %, most
35 preferably about 30 wt. %, of glass or carbon fiber, most preferably glass fiber. In some embodiments, the polymer composition includes less than 10 wt.%, less

than 5 wt.%, less than 1 wt.% of a fibrous filler, a particulate filler, or both. In some aspects, the polymer composition is free of a fibrous filler, a particulate filler, or both.

Optional Additives

5 The polymer composition may further include optional additives such as titanium dioxide, zinc sulfide, zinc oxide, ultraviolet light stabilizers, heat stabilizers, antioxidants such as organic phosphites and phosphonites, acid scavengers, processing aids, nucleating agents, lubricants, flame retardants, smoke-suppressing agents, anti-static agents, anti-blocking agents, and
10 conductivity additives such as carbon black.

 When one or more optional additives are present, their total concentration is preferably less than 10 wt. %, more preferably less than 5 wt. %, and most preferably less than 2 wt. %, based on the total weight of polymer composition.

 In some embodiments, the polymer composition includes less than
15 0.5 wt.% of a flame retardant. Preferably, the polymer composition is free of a flame retardant.

Method of Making the Polymer Composition

 The polymer composition can be prepared by melt-mixing a the PSU, the SiPC, the optional PEI, the optional reinforcing filler, and any optional additives,
20 to provide a molten mixture, followed by extrusion and cooling of the molten mixture.

 Exemplary embodiments also include a method of increasing the flame resistance of a composition comprising greater than 30 wt.% of a PSU, based on the total weight of the polymer composition, by adding a SiPC as described
25 herein to the polymer composition.

 The compositions described herein are advantageously provided in the form of pellets, which may be used in injection molding or extrusion processes known in the art.

 The preparation of the polymer composition can be carried out by any
30 known melt-mixing process that is suitable for preparing thermoplastic molding compositions. Such a process is typically carried out by heating the thermoplastic polymer above the melting temperature of the thermoplastic polymer thereby forming a melt of the thermoplastic polymer. The process for the preparation of the composition can be carried out in a melt-mixing apparatus,
35 for which any melt-mixing apparatus known to the one skilled in the art of preparing polymer compositions by melt mixing can be used. Suitable melt-

mixing apparatus are, for example, kneaders, Banbury mixers, single-screw extruders, and twin-screw extruders. Preferably, use is made of an extruder fitted with means for dosing all the desired components to the extruder, either to the extruder's throat or to the melt. In the process for the preparation of the polymer composition the constituting components for forming the composition are fed to the melt-mixing apparatus and melt-mixed in that apparatus. The components may be fed simultaneously as a powder mixture or granule mixture, also known as dry-blend, or may be fed separately.

Shaped Articles Comprising the Polymer Composition and Methods of Making

Exemplary embodiments also include shaped articles comprising the polymer-metal junction and methods of making the shaped articles.

The polymer composition may be well suited for the manufacture of articles useful in a wide variety of applications. For example, the excellent flame resistance of the polymer compositions makes them ideal for use in applications such as medical, health care, electrical, electronic, and smart device applications

The polymer compositions described herein can be used for the manufacture of shaped articles, in particular, parts of electronic devices, more particularly parts of portable or mobile electronic devices.

The term "mobile electronic device" is intended to denote any electronic device that is designed to be conveniently transported and used in various locations while exchanging/providing access to data, e.g. through wireless connections or mobile network connection. Representative examples of mobile electronic devices include mobile phones, personal digital assistants, laptop computers, tablet computers, radios, cameras and camera accessories, watches, calculators, music players, global positioning system receivers, portable games, hard drives and other electronic storage devices, and the like. The at least one part of the mobile electronic device according to the present invention may be selected from a large list of articles such as fitting parts, snap fit parts, mutually moveable parts, functional elements, operating elements, tracking elements, adjustment elements, carrier elements, frame elements, switches, connectors and (internal and external) components of housing, which can be notably produced by injection molding, extrusion or other shaping technologies.

An aspect of the present invention is directed to mobile electronic devices comprising at least one structural part made of a polymer composition described herein, and in particular to a laptop, a mobile phone, a GPS, a tablet, personal

digital assistants, portable recording devices, portable reproducing devices and portable radio receives.

In particular, the polymer compositions described herein are very well suited for the production of housing components of mobile electronic devices.

5 Therefore, the at least one part of the mobile electronic device according to the present invention is advantageously a component of a mobile electronic device housing. By "mobile electronic device housing" is meant one or more of the back cover, front cover, antenna housing, frame and/or backbone of a mobile electronic device. The housing may be a single component-article or, more often,
10 may comprise two or more components. By "backbone" is meant a structural component onto which other components of the device, such as electronics, microprocessors, screens, keyboards and keypads, antennas, battery sockets, and the like are mounted. The backbone may be an interior component that is not visible or only partially visible from the exterior of the mobile electronic device.
15 The housing may provide protection for internal components of the device from impact and contamination and/or damage from environmental agents (such as liquids, dust, and the like). Housing components such as covers may also provide substantial or primary structural support for and protection against impact of certain components having exposure to the exterior of the device such
20 as screens and/or antennas. Housing components may also be designed for their aesthetic appearance and touch.

In a preferred embodiment, the mobile electronic device housing is selected from the group consisting of a mobile phone housing, a tablet housing, a laptop computer housing and a tablet computer housing. Excellent results were
25 obtained when the part of the mobile electronic device according to the present invention was a mobile phone housing.

In some aspects, the shaped articles may be made from the polymer composition using any suitable melt-processing method such as injection molding, extrusion molding, roto-molding, or blow-molding.

30 Exemplary embodiments are also directed to methods of making shaped articles by additive manufacturing, where the shaped article is printed from the polymer composition. The methods include printing layers of the shaped article from the polymer composition as described below.

Additive manufacturing systems are used to print or otherwise build a
35 shaped object from a digital representation of the shaped object by one or more additive manufacturing techniques. Examples of commercially available

additive manufacturing techniques include extrusion-based techniques, selective laser sintering, powder/binder jetting, electron-beam melting, and stereolithography processes. For each of these techniques, the digital representation of the shaped object is initially sliced into multiple horizontal layers. For each layer, a tool path is then generated, which provides instructions for the particular additive manufacturing system to print the given layer.

Accordingly, some embodiments include a method of making a shaped article comprising printing layers of the polymer composition to form the shaped article by an extrusion-based additive manufacturing system (for example FFF), a powder-based additive manufacturing system (for example SLS), or a continuous Fiber-Reinforced Thermoplastic (FRTP) printing method.

EXAMPLES

The invention will be now described in more details with reference to the following examples, whose purpose is merely illustrative and not intended to limit the scope of the invention.

Materials

The following materials were used to prepare the Examples and Comparative Examples :

Udel® PSU P-1700 NT 11 and Udel® P-3703 NT 11, available from Solvay Specialty Polymers USA, LLC.

Tarflon® AG1760 SiPC copolymer available from Idemitsu Chemical Co.

Veradel® PESU 3300, available from Solvay Specialty Polymers USA, LLC.

Makrolon® 3108 PC available from Bayer Materials Science, Inc.

Preparation of Polymer Compositions

The polymer compositions of the Examples and Comparative Examples shown in Table 1 were prepared by first drying the polymer ingredients for at least 16 hours in desiccated ovens. The PSU was dried at a temperature of 300°C, while the PC and SiPC were dried at a temperature of 175 °F. Following drying, the ingredients of each example were tumble-blended for about 20 minutes. Each formulation was then melt compounded using a 26 mm diameter Coperion® ZSK-26 co-rotating partially intermeshing twin screw extruder. In each case, the resins were fed at barrel section 1 using a gravimetric feeder. The compositions were extruded, cooled, and then cut in a pelletizer to form pellets approximately 3.0 mm in length by 2.7 mm in diameter.

Preparation of Test Specimens

The example and comparative polymer compositions were injection molded to produce the ASTM tensile and flexural specimens and UL94 flame resistance specimens described below.

5 **Mechanical Testing**

Mechanical properties were evaluated for all the formulations using injection molded 0.125 inch thick ASTM test specimens which consisted of Type I tensile bars and 5 in x 0.5 in x 0.125 in flexural bars. The following ASTM test methods were employed in evaluating the polymer compositions :

- 10 D-638 : Tensile properties : tensile strength at break, tensile strength at yield, tensile elongation at break, tensile elongation at yield, and modulus of elasticity.

D-256 : Notched Izod impact resistance.

Flame Resistance Testing

- 15 The flame resistance properties of the polymer compositions were evaluated according to UL94 using specimens measuring 12.7 mm x 127 mm with thicknesses of 1.6 mm.

The following UL94 ratings, listed in increasing order of flame resistance, were assigned to each of the polymer compositions in Table 1 :

- 20 **N.R.** : No rating—the composition failed the test.

V-2 : burning stops within 30 seconds on a vertical specimen; drips of flaming particles are allowed.

V-1 : burning stops within 30 seconds on a vertical specimen; drips of particles allowed as long as they are not inflamed.

- 25 **V-0** : burning stops within 10 seconds on a vertical specimen; drips of particles allowed as long as they are not inflamed.

Table 1

[illegible]

- 23 -

Referring to Table 1, Comparative Examples C1 and C14 show that compositions of 100 wt.% SiPC copolymer and 100 wt.% PSU, respectively, either failed the flame resistance test (NR) or achieved a low score of “V-2”.

5 The compositions comprising 30 wt.% or less of PSU (Comparative Examples C2 to C4) lead to a V-2 or lower flame resistance score and therefore were not considered satisfactory.

Comparative Example C15 including 50 wt.% of poly(ether sulfone) (PES) and 50 wt.% of SiPC copolymer achieved a V-2 flame resistance rating in each of the flame resistance tests. This low rating was exhibited despite the fact that
10 the 100 wt.% PES composition of Comparative Example C16 showed a V-0 rating.

Comparative Example C17 shows that a composition including PSU and a polycarbonate achieved a V-2 flame resistance rating, thereby not leading to a satisfactory flame resistance.

15 The polymer compositions of Examples 5 to 13 comprising more than 30 wt.% of PSU and a SiPC copolymer unexpectedly achieved a V-0 UL94 flame rating.

Finally, the polymer compositions of Examples 5 to 8 also unexpectedly exhibited markedly improved impact resistance as compared with the impact
20 resistance observed for the comparative examples.

Should the disclosure of any patents, patent applications, and publications which are incorporated herein by reference conflict with the description of the present application to the extent that it may render a term unclear, the present description shall take precedence.

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CLAIMS

1. A polymer composition comprising :

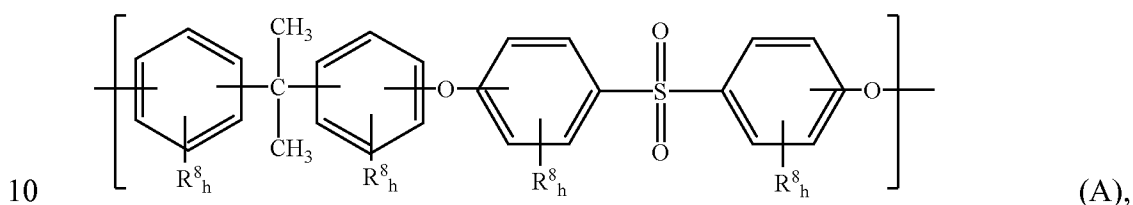
- greater than 30 wt.% of at least one polysulfone (PSU), based on the total weight of the polymer composition,

5 - at least one polycarbonate-polysiloxane copolymer (SiPC),

- optionally, at least one poly(ether imide) (PEI), and

- optionally, at least one reinforcing filler.

2. The polymer composition of claim 1, wherein the polysulfone (PSU) comprises more than 50 mol % of recurring units of formula (A-1) :



wherein :

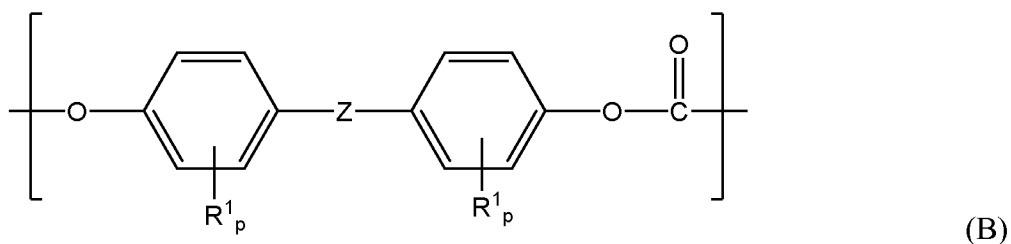
each R^8 is independently selected from the group consisting of a halogen, an alkyl, an alkenyl, an alkynyl, an aryl, an ether, a thioether, a carboxylic acid, an ester, an amide, an imide, an alkali or alkaline earth metal sulfonate, an alkyl
15 sulfonate, an alkali or alkaline earth metal phosphonate, an alkyl phosphonate, an amine, and a quaternary ammonium; and

each h is independently selected from the group consisting of integers ranging from 0 to 4.

3. The polymer composition of any one of claims 1 and 2, wherein the at
20 least one polycarbonate-polysiloxane copolymer (SiPC) comprises :

i) polycarbonate repeat units of formula (B) :

- 25 -



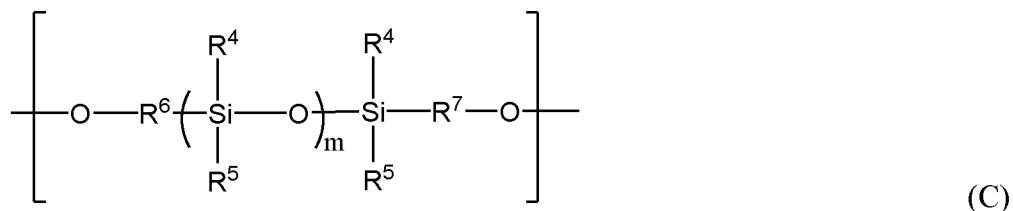
wherein :

Z is selected from the group consisting of a bond, an alkylene group having 1 to 8 carbon atoms, an alkylidene group having 2 to 8 carbon atoms, a
 5 cycloalkylene group having 5 to 15 carbon atoms, a cycloalkylidene group having 5 to 15 carbon atoms, -S-, -SO-, -SO₂-, -O- and -CO-,

each R¹ is independently selected from the group consisting of a halogen, an alkyl group having 1-20 carbon atoms, and an aryl group; and

each p is independently selected from integers ranging from 0 to 4; and

10 ii) polysiloxane repeat units of formula (C) :



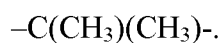
wherein :

each R⁴ and R⁵ are independently selected from the group consisting of a hydrogen, a halogen, a hydrocarbyl, and a halogen-substituted hydrocarbyl,

15 R⁶ and R⁷ are independently selected from the group consisting of organic residues having an aromatic nucleus, and

m is selected from integers ranging from 5 to more than 100.

4. The polymer composition of claim 3, wherein Z is a group

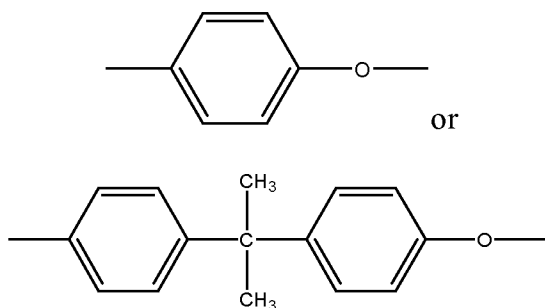


20 5. The polymer composition of any one of claims 3 and 4, wherein each R⁴ and R⁵ is a methyl group.

- 26 -

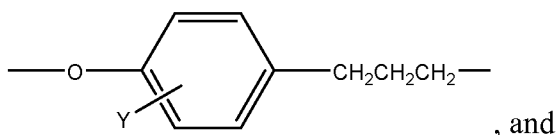
6. The polymer composition of any one of claims 3 to 5, wherein R^6 and R^7 are independently selected from the group consisting of a 3-(o-hydroxyphenyl)propylene group, a 2-(p-hydroxyphenyl)ethylene group, and groups of formulas :

5

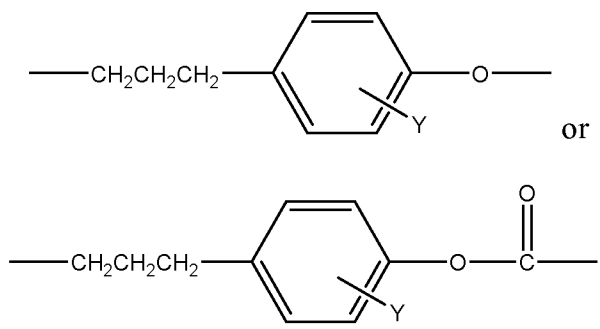


7. The polymer composition of any one of claims 3 to 6, wherein :

- the moiety $-O-R^6-$ in Formula (C) is a group of formula :



10 - the moiety $-R^7-O-$ in Formula (C) is a group of formula :



wherein Y is a hydrogen, a hydrocarbyl, or a halogen-substituted hydrocarbyl.

8. The polymer composition of any one of claims 1 to 7, wherein the polymer composition comprises from 1 wt.% to 25 wt.% of at least one poly(ether imide) (PEI), based on the total weight of the polymer composition.

9. The polymer composition of any one of claims 1 to 8, wherein the polymer composition further comprises at least one reinforcing filler, preferably glass fiber.

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10. The polymer composition of any one of claims 1 to 9, wherein the polymer composition comprises from 35 wt. % to 95 wt. % of polysulfone (PSU), based on the total weight of the polymer composition.

5 11. The polymer composition of any one of claims 1 to 9, wherein the polymer composition comprises less than 70 wt.% of the at least one polycarbonate-polysiloxane copolymer (SiPC), based on the total weight of the polymer composition.

10 12. The polymer composition of any one of claims 1 to 11, wherein the polymer composition comprises from 40 wt. % to 90 wt. % of at least one polysulfone (PSU), and from 10 wt. % to 60 wt. % of at least one polycarbonate-polysiloxane copolymer (SiPC), based on the total weight of the polymer composition.

13. The polymer composition of any one of claims 1 to 12, wherein the polymer composition exhibits a UL94 1.6 mm flame rating of V-0.

15 14. A shaped article comprising the polymer composition of any one of claims 1 to 13.

15. The shaped article of claim 14, wherein the shaped article is a part of a mobile electronic device.

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2018/080862

A. CLASSIFICATION OF SUBJECT MATTER
INV. C08L81/06
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)
C08L

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

Electronic data base consulted during the international search (name of data base and, where 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.
X	US 8 017 699 B1 (SANNER MARK [US] ET AL) 13 September 2011 (2011-09-13) column 1, lines 7-11,40-67; claims 1,32,37; examples A16,A17,A18; tables 1,5A -----	1-15
A	WO 2017/003843 A1 (SABIC GLOBAL TECHNOLOGIES BV [NL]; ZHOU HAO [US]) 5 January 2017 (2017-01-05) claim 1 -----	1-15
A	WO 2014/202673 A1 (SOLVAY SPECIALTY POLYMERS USA [US]) 24 December 2014 (2014-12-24) page 1, lines 7-9; examples E5,E6,E8,E12,E16,E17,E18,E20,E21,E25,E28,E 31,E34 ----- -/--	1-15



Further documents are listed in the continuation of Box C.



See patent family annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

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"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search

18 January 2019

Date of mailing of the international search report

30/01/2019

Name and mailing address of the ISA/

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Authorized officer

Bezard, Stéphane

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2018/080862

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 2014/371360 A1 (ZHENG YUN [CN] ET AL) 18 December 2014 (2014-12-18) paragraphs [0002], [0003], [0004]; claim 1 -----	1-15

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/EP2018/080862

Patent document cited in search report		Publication date		Patent family member(s)		Publication date
US 8017699	B1	13-09-2011	CN	103180389 A		26-06-2013
			EP	2630192 A1		28-08-2013
			EP	3246363 A1		22-11-2017
			KR	20130127982 A		25-11-2013
			US	8017699 B1		13-09-2011
			US	2012100324 A1		26-04-2012
			WO	2012054595 A1		26-04-2012

WO 2017003843	A1	05-01-2017	CN	107922616 A		17-04-2018
			EP	3317325 A1		09-05-2018
			US	2018215872 A1		02-08-2018
			WO	2017003843 A1		05-01-2017

WO 2014202673	A1	24-12-2014	CN	105473662 A		06-04-2016
			EP	3010976 A1		27-04-2016
			JP	2016522300 A		28-07-2016
			US	2016130441 A1		12-05-2016
			WO	2014202673 A1		24-12-2014

US 2014371360	A1	18-12-2014	NONE			
