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(54) Title: REINFORCED POLYPHTHALAMIDE/POLY(PHENYLENE ETHER) COMPOSITION

(57) Abstract: Disclosed herein is a reinforced composition comprising: 55 to 80 wt% of a compatibilized blend of a polyphthalamide and a poly(phenylene ether) wherein said compatibilized blend is formed from a mixture of polyphthalamide and poly(phenylene ether) in a ratio of between 1.5:1 and 7.0:1, and a functionalizing agent in an amount sufficient to effect compatibilization; and 20 to 45 wt% of a nylon glass fiber, wherein the composition contains less than 0.1 wt% of phosphinates and less than 0.1 wt% of impact modifiers and weight percent is based on the total weight of the composition.



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REINFORCED POLYPHTHALAMIDE/POLY(PHENYLENE ETHER) COMPOSITION

BACKGROUND OF INVENTION

[0001] This application relates to polyphthalamide/poly(phenylene ether) blends and more particularly to reinforced polyphthalamide/poly(phenylene ether) blends.

[0002] Glass filled poly(phenylene ether)/polyamide 66 (GF-PPE/PA66) blends have been used in applications such as water pumps and water meters for long time. The morphology of PPE/PA66 blends can be defined as “islands-and-sea” where PPE particles are the dispersed phase (as islands) and PA66 is the continuous phase (as sea). As a result, PPE/PA66 blends are commonly recognized in industry as modified nylons with superior performance such as higher heat resistance, better dimension stability, less warpage and sagging, and less moisture absorption. Yet due to the inherent nature of PA66, PPE/PA66 blends are also susceptible to moisture induced property changes such as reduction of flexural modulus and tensile strength as the moisture content in the nylon matrix increases. In some cases when elevated temperatures are employed and/or aggressive chemicals are added such as chlorine in order to purify water, neat GF PA66 and GF PPE/PA66 blends may result in earlier part failure caused by severe nylon hydrolysis.

[0003] There is an ongoing need in the art for reinforced polyamide (nylon) blends with improved high temperature hydrolytic stability.

BRIEF DESCRIPTION OF THE INVENTION

[0004] The above-described and other drawbacks are alleviated by a reinforced composition comprising

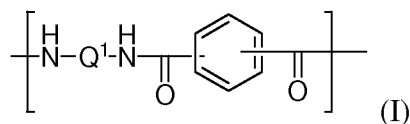
55 to 80 weight percent (wt%) of a compatibilized blend of a polyphthalamide and a poly(phenylene ether) wherein said compatibilized blend is formed from a mixture of polyphthalamide and poly(phenylene ether) in a ratio of between 1.5: 1 and 7.0:1, and a functionalizing agent in an amount sufficient to effect compatibilization; and

20 to 45 wt% of a nylon glass fiber,

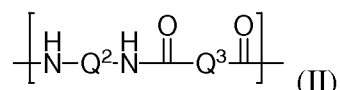
wherein the composition contains less than 0.1 wt% of phosphinates and less than 0.1 wt% of impact modifiers and weight percent is based on the total weight of the composition.

[0005] In another embodiment a reinforced composition comprises

55 to 80 wt% of a compatibilized blend of a polyphthalamide comprising (a) 60-70 mol% of units of formula (I)



wherein Q^1 is a 1,6-hexyl group and the aromatic portion of the repeating unit is derived from terephthalic acid, (b) 20-30 mol% of units of formula (I) wherein Q^1 is a 1,6-hexyl group and the aromatic portion of the repeating unit is derived from isophthalic acid, and (c) 5-15 mol% of units of formula (II)



wherein Q^2 is a 1,4-butyl group and Q^3 is a 1,6-hexyl group, and a poly(phenylene ether) comprising 2,6-dimethyl-1,4-phenylene ether units wherein said compatibilized blend is formed from a mixture of said polyphthalamide and poly(phenylene ether) in a ratio of between 1.5: 1 and 7.0:1, and 0.2 to 0.9 weight percent citric acid; and

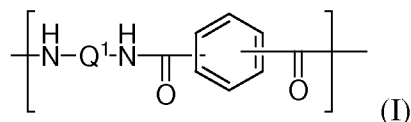
20 to 45 wt% of a nylon glass fiber, wherein the composition contains less than 0.1 wt% of phosphinates and less than 0.1 wt% of impact modifiers and weight percent is based on the total weight of the composition.

[0006] In another embodiment an article comprises a reinforced composition comprising

55 to 80 wt% of a compatibilized blend of a polyphthalamide and a poly(phenylene ether) wherein said compatibilized blend is formed from a mixture of polyphthalamide and poly(phenylene ether) in a ratio of between 1.5: 1 and 7.0:1, and a functionalizing agent in an amount sufficient to effect compatibilization; and

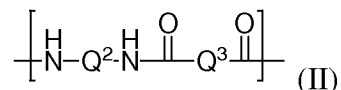
20 to 45 wt% of a nylon glass fiber, wherein the composition contains less than 0.1 wt% of phosphinates and less than 0.1 wt% of impact modifiers and weight percent is based on the total weight of the composition.

[0007] In another embodiment an article comprises 55 to 80 weight percent (wt%) of a compatibilized blend of a polyphthalamide comprising (a) 60-70 mol% of units of formula (I)



wherein Q^1 is a 1,6-hexyl group and the aromatic portion of the repeating unit is derived from terephthalic acid, (b) 20-30 mol% of units of formula (I) wherein Q^1 is a 1,6-

hexyl group and the aromatic portion of the repeating unit is derived from isophthalic acid, and (c) 5-15 mol% of units of formula (II)



wherein Q² is a 1,4-butyl group and Q³ is a 1,6-hexyl group, and a poly(phenylene ether) comprising 2,6-dimethyl-1,4-phenylene ether units wherein said compatibilized blend is formed from a mixture of said polyphthalamide and poly(phenylene ether) in a ratio of between 1.5: 1 and 7.0:1, and 0.2 to 0.9 weight percent citric acid; and

20 to 45 wt% of a nylon glass fiber, wherein the composition contains less than 0.1 wt% of phosphinates and less than 0.1 wt% of impact modifiers and weight percent is based on the total weight of the composition.

[0008] Also disclosed herein is a method of making the reinforced composition.

DETAILED DESCRIPTION OF THE INVENTION

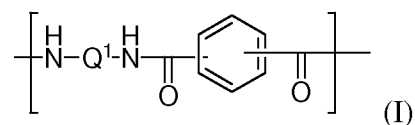
[0009] Polyphthalamides (PPA) are semi-aromatic, high temperature nylons that typically have melting points higher than 290°C and glass transitions temperatures greater than 80°C. PPAs are generally based on polyamide 6T. However polyamide 6T has a melting point of 370°C, and therefore is difficult to process and/or blend with other polymers, such as polyphenylene ether (PPE). PPAs generally are copolymers that have various ratios of polyamide 6T, polyamide 6I, and polyamide 6/6 to control properties such as melt flow, melting point, and glass transition temperature. PPAs are distinct from high temperature nylons (HTN) in that PPAs comprise aromatic repeating units in an amount greater than or equal to 55 weight percent. The glass transitions temperature and the melting point are indicative of the aromatic content of the PPA.

[0010] Reinforced PPA has better hydrolytic stability than reinforced PA66. However, it was surprising that reinforced PPE/PPA blends as described herein have better hydrolytic stability than reinforced PPA.

[0011] In some embodiments the compositions described herein can be substantially free of added aliphatic polyamides. Substantially free of added aliphatic polyamide is defined as containing less than or equal to 2 weight percent, or, more specifically less than or equal to 1 weight percent, or, more specifically, less than or equal to 0.5 weight percent of an added aliphatic polyamide, based on the total weight of the composition. An added liphatic polyamides is defined as an aliphatic polyamide added in addition to the polyphthalamide. It

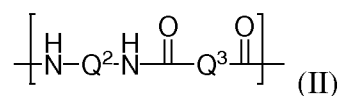
is understood that polyphthalamides may contain residual aliphatic polyamides resulting from synthesis of the polyphthalamide. Aliphatic polyamides are distinguished from polyphthalamides in that aliphatic polyamides have no repeating units that comprise aromatic moieties. Aliphatic polyamides include both homopolymers and copolymers.

[0012] Polyphthalamides comprise repeating units having formula (I)



wherein Q^1 is independently at each occurrence a branched or unbranched alicyclic alkyl group having 4 to 8 carbons. In some embodiments, Q^1 is independently at each occurrence a 1,6-hexyl group. Polyamide resins, in general characterized by the presence of an amide group ($-\text{C}(\text{O})\text{NH}-$) which is the condensation product of a carboxylic acid and an amine. Polyphthalamides are the condensation product of terephthalic acid and an amine, isophthalic acid and an amine or a combination of terephthalic acid, isophthalic acid and an amine. When employing more than one diamine the ratio of the diamines can affect some of the physical properties of the resulting polymer such as the melt temperature. When employing more than one acid, the ratio of the acids can affect some of the physical properties of the resulting polymer as well. The ratio of diamine to dicarboxylic acid is typically equimolar although excesses of one or the other may be used to determine the end group functionality. In addition the reaction can further include monoamines and monocarboxylic acids which function as chain stoppers and determine, at least in part, the end group functionality. In some embodiments it is preferable to have an amine end group content of greater than or equal to about 30 milliequivalents per gram (meq/g), or, more specifically, greater than or equal to about 40meq/g.

[0013] In some embodiments the polyphthalamide is a block copolymer or a random copolymer comprising the units of formula (I) and units of formula (II)



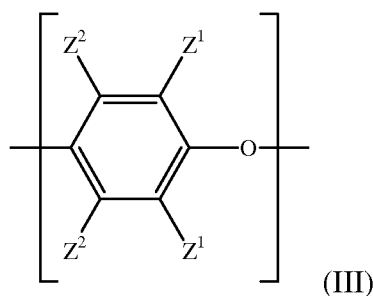
wherein Q^2 and Q^3 are independently at each occurrence a branched or unbranched alicyclic alkyl group having 4 to 12 carbons. Q^2 and Q^3 can be the same or different alicyclic alkyl group.

[0014] The polythalamide has a glass transition temperature (T_g) greater than or equal to 80°C , or, greater than or equal to 100°C , or, greater than or equal to 120°C . The polythalamide also has melting temperature (T_m) of 290 to 330°C . Within this range the T_m

may be greater than or equal to 300°C. Also within this range the T_m may be less than or equal to 325°C.

[0015] The polyphthalamide is present in an amount of 30 to 70 weight percent based on the total weight of the composition. Within this range the amount of polyphthalamide can be greater than or equal to 35 weight percent, or, more specifically, greater than or equal to 40 weight percent. Also within this range the amount of polyphthalamide can be less than or equal to 60 weight percent, or, more specifically, less than or equal to 55 weight percent.

[0016] In addition to polyphthalamide, the composition comprises a poly(phenylene ether). Poly(phenylene ether) comprises repeating structural units of formula (III)



wherein for each structural unit, each Z¹ is independently halogen, unsubstituted or substituted C₁-C₁₂ hydrocarbyl with the proviso that the hydrocarbyl group is not tertiary hydrocarbyl, C₁-C₁₂ hydrocarbylthio, C₁-C₁₂ hydrocarbyloxy, or C₂-C₁₂ halohydrocarbyloxy wherein at least two carbon atoms separate the halogen and oxygen atoms; and each Z² is independently hydrogen, halogen, unsubstituted or substituted C₁-C₁₂ hydrocarbyl with the proviso that the hydrocarbyl group is not tertiary hydrocarbyl, C₁-C₁₂ hydrocarbylthio, C₁-C₁₂ hydrocarbyloxy, or C₂-C₁₂ halohydrocarbyloxy wherein at least two carbon atoms separate the halogen and oxygen atoms.

[0017] As used herein, the term “hydrocarbyl”, whether used by itself, or as a prefix, suffix, or fragment of another term, refers to a residue that contains only carbon and hydrogen. The residue can be aliphatic or aromatic, straight-chain, cyclic, bicyclic, branched, saturated, or unsaturated. It can also contain combinations of aliphatic, aromatic, straight chain, cyclic, bicyclic, branched, saturated, and unsaturated hydrocarbon moieties. However, when the hydrocarbyl residue is described as “substituted”, it can contain heteroatoms over and above the carbon and hydrogen members of the substituent residue. Thus, when specifically described as substituted, the hydrocarbyl residue can also contain halogen atoms, nitro groups, cyano groups, carbonyl groups, carboxylic acid groups, ester groups, amino groups, amide groups, sulfonyl groups, sulfoxyl groups, sulfonamide groups,

sulfamoyl groups, hydroxyl groups, alkoxyl groups, or the like, and it can contain heteroatoms within the backbone of the hydrocarbyl residue.

[0018] The poly(phenylene ether) can comprise molecules having aminoalkyl-containing end group(s), typically located in an ortho position to the hydroxy group. Also frequently present are tetramethyl diphenylquinone (TMDQ) end groups, typically obtained from reaction mixtures in which tetramethyl diphenylquinone by-product is present.

[0019] The poly(phenylene ether) can be in the form of a homopolymer; a copolymer; a graft copolymer; an ionomer; or a block copolymer; as well as combinations comprising two or more of the foregoing polymers. Poly(phenylene ether) includes polyphenylene ether comprising 2,6-dimethyl-1,4-phenylene ether units optionally in combination with 2,3,6-trimethyl-1,4-phenylene ether units.

[0020] The poly(phenylene ether) can be prepared by the oxidative coupling of monohydroxyaromatic compound(s) such as 2,6-xyleneol and/or 2,3,6-trimethylphenol. Catalyst systems are generally employed for such coupling; they can contain heavy metal compound(s) such as a copper, manganese or cobalt compound, usually in combination with various other materials such as a secondary amine, tertiary amine, halide or combination of two or more of the foregoing.

[0021] A portion of the poly(phenylene ether) can be functionalized with a polyfunctional compound (functionalizing agent) as described below. The poly(phenylene ether) can be functionalized prior to making the composition or can be functionalized as part of making the composition. Furthermore, prior to functionalization the poly(phenylene ether) can be extruded, for example to be formed into pellets. It is also possible for the poly(phenylene ether) to be melt mixed with other additives that do not interfere with functionalization. Exemplary additives of this type include s, flow promoters, and the like.

[0022] In some embodiments the poly(phenylene ether) can comprise 0.1 weight percent weight percent to 90 weight percent of structural units derived from a functionalizing agent, based on the total weight of the poly(phenylene ether). Within this range, the poly(phenylene ether) can comprise less than or equal to 80 weight percent, or, more specifically, less than or equal to 70 weight percent of structural units derived from functionalizing agent, based on the total weight of the poly(phenylene ether).

[0023] The poly(phenylene ether) can have a number average molecular weight of 3,000 to 40,000 grams per mole (g/mol) and a weight average molecular weight of 5,000 to 80,000 g/mol, as determined by gel permeation chromatography using monodisperse polystyrene standards, a styrene divinyl benzene gel at 40°C and samples having a

concentration of 1 milligram per milliliter of chloroform. The poly(phenylene ether) or combination of poly(phenylene ether)s has an initial intrinsic viscosity of 0.1 to 0.60 deciliters per gram (dl/g), as measured in chloroform at 25°C. Initial intrinsic viscosity is defined as the intrinsic viscosity of the poly(phenylene ether) prior to melt mixing with the other components of the composition and final intrinsic viscosity is defined as the intrinsic viscosity of the poly(phenylene ether) after melt mixing with the other components of the composition. As understood by one of ordinary skill in the art the viscosity of the poly(phenylene ether) may be up to 30% higher after melt mixing. The percentage of increase can be calculated by $(\text{final intrinsic viscosity} - \text{initial intrinsic viscosity}) / \text{initial intrinsic viscosity}$. Determining an exact ratio, when two initial intrinsic viscosities are used, will depend somewhat on the exact intrinsic viscosities of the poly(phenylene ether) used and the ultimate physical properties that are desired.

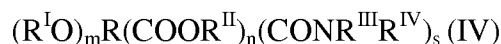
[0024] The poly(phenylene ether) is present in an amount of 10 to 30 weight percent based on the total weight of the composition. Within this range the poly(phenylene ether) may be present in an amount greater than or equal to 15 weight percent, or, more specifically, greater than or equal to 18 weight percent. Also within this range the poly(phenylene ether) can be present in an amount of less than or equal to 25 weight percent, or, more specifically, less than or equal to 20 weight percent.

[0025] The compatibilized polyphthalamide/poly(phenylene ether) blend is formed using a functionalizing agent. When used herein, the expression "functionalizing agent" refers to polyfunctional compounds which interact with the poly(phenylene ether), the polyamide resin, or both. This interaction may be chemical (e.g., grafting) and/or physical (e.g., affecting the surface characteristics of the dispersed phases). In either instance the resulting compatibilized polyphthalamide/poly(phenylene ether) composition appears to exhibit improved compatibility, particularly as evidenced by enhanced impact strength, mold knit line strength and/or elongation. As used herein, the expression "compatibilized polyphthalamide/poly(phenylene ether) blend" refers to those compositions which have been physically and/or chemically compatibilized with a functionalizing agent.

[0026] The functionalizing agent comprises a polyfunctional compound that is one of two types. The first type has in the molecule both (a) a carbon-carbon double bond and (b) at least one carboxylic acid, anhydride, epoxy, imide, amide, ester group or functional equivalent thereof. Examples of such polyfunctional compounds include maleic acid; maleic anhydride; fumaric acid; maleic hydrazide; dichloro maleic anhydride; and unsaturated dicarboxylic acids (e.g. acrylic acid, butenoic acid, methacrylic acid, t-ethylacrylic acid,

pentenoic acid). In some embodiments, the functionalizing agent comprises maleic anhydride and/or fumaric acid.

[0027] The second type of polyfunctional functionalizing agent compounds are characterized as having both (a) a group represented by the formula (OR) wherein R is hydrogen or an alkyl, aryl, acyl or carbonyl dioxy group and (b) at least two groups each of which may be the same or different selected from carboxylic acid, acid halide, anhydride, acid halide anhydride, ester, orthoester, amide, imido, amino, and salts thereof. Typical of this type of functionalizing agents are the aliphatic polycarboxylic acids, acid esters and acid amides represented by the formula (IV):



wherein R is a linear or branched chain saturated aliphatic hydrocarbon having 2 to 20, or, more specifically, 2 to 10 carbon atoms; R^I is hydrogen or an alkyl, aryl, acyl or carbonyl dioxy group having 1 to 10, or, more specifically, 1 to 6, or, even more specifically, 1 to 4 carbon atoms; each R^{II} is independently hydrogen or an alkyl or aryl group having 1 to 20, or, more specifically, 1 to 10 carbon atoms; each R^{III} and R^{IV} are independently hydrogen or an alkyl or aryl group having 1 to 10, or, more specifically 1 to 6, or, even more specifically, 1 to 4, carbon atoms; m is equal to 1 and (n + s) is greater than or equal to 2, or, more specifically, equal to 2 or 3, and n and s are each greater than or equal to zero and wherein (OR^I) is alpha or beta to a carbonyl group and at least two carbonyl groups are separated by 2 to 6 carbon atoms. Obviously, R^I , R^{II} , R^{III} and R^{IV} cannot be aryl when the respective substituent has less than 6 carbon atoms.

[0028] Suitable polycarboxylic acids include, for example, citric acid, malic acid, agaricic acid; including the various commercial forms thereof, such as for example, the anhydrous and hydrated acids; and combinations comprising one or more of the foregoing. In some embodiments, the functionalizing agent comprises citric acid. Illustrative of esters useful herein include, for example, acetyl citrate and mono- and/or distearyl citrates and the like. Suitable amides useful herein include, for example, N,N'-diethyl citric acid amide; N-phenyl citric acid amide; N-dodecyl citric acid amide; N,N'-didodecyl citric acid amide and N-dodecyl malic acid. Derivates include the salts thereof, including the salts with amines and the alkali and alkaline metal salts. Exemplary suitable salts include calcium malate, calcium citrate, potassium malate, and potassium citrate.

[0029] The foregoing functionalizing agents may be added directly to the melt blend or pre-reacted with either or both the poly(phenylene ether) and polyamide. In some embodiments, at least a portion of the functionalizing agent is pre-reacted, either in the melt

or in a solution of a suitable solvent, with all or a part of the poly(phenylene ether). It is believed that such pre-reacting may cause the functionalizing agent to react with the polymer and, consequently, functionalize the poly(phenylene ether). For example, the poly(phenylene ether) may be pre-reacted with maleic anhydride, fumaric acid and/or citric acid to form an anhydride and/or acid functionalized poly(phenylene ether) which has improved compatibility with the polyamide compared to a non-functionalized poly(phenylene ether).

[0030] The amount of the functionalizing agent used will be dependent upon the specific functionalizing agent chosen and the specific polymeric system to which it is added.

[0031] In some embodiments, the functionalizing agent is employed in an amount of 0.05 to 2.0 weight percent, based on the total weight of the composition. Within this range the amount of functionalizing agent may be greater than or equal to 0.1, or, more specifically, greater than or equal to 0.2, or, more specifically, greater than or equal to 0.5 weight percent. Also within this range the amount of functionalizing agent may be less than or equal to 1.75, or, more specifically, less than or equal to 1.5 weight percent, or, more specifically less than or equal to 0.9 weight percent.

[0032] The composition comprises a nylon glass fiber. A nylon glass fiber is defined as a glass fiber surface treated with silanes to improve adhesion and dispersion with polyamide as are commonly known in the art. The glass fiber may have an average length of 2.8 to 3.6 millimeters. The glass fiber may have an average diameter of 8 to 16 micrometers.

[0033] The nylon glass fiber is incorporated in an amount of 20 to 45 weight percent based on the total weight of the composition. Within this range the amount of reinforcing filler can be greater than or equal to 25 weight percent, or, more specifically, greater than or equal to 28 weight percent. Also within this range the amount of reinforcing filler can be less than or equal to 40, or, more specifically, less than or equal to 37 weight percent.

[0034] The composition can, optionally, further comprise one or more other additives known in the thermoplastics arts. Useful additives include, for example, stabilizers, mold release agents, processing aids, drip retardants, nucleating agents, dyes, pigments, colorants, crystallization nucleators, metal salts, antioxidants, anti-static agents, plasticizers, lubricants, blowing agents, metal deactivators, antiblocking agents, nanoclays, fragrances (including fragrance-encapsulated polymers), and the like, and combinations thereof. Additives can be added in amounts that do not unacceptably detract from the desired performance and physical properties of the composition. Such amounts can be determined by a skilled artisan without undue experimentation. Generally, the total amount of additives will be less than or equal to 5 weight percent based on the total weight of the composition.

[0035] In one embodiment, the composition comprises a black pigment. In one embodiment the black pigment comprises carbon black in an amount up to 0.5 weight percent, based on the total weight of the composition.

[0036] The composition can, optionally, exclude any polymer other than those taught herein as required.

[0037] Specific embodiments of the compositions are shown in Table 1.

Table 1

	Embodiment A	Embodiment B	Embodiment C
PPE (as described in Table 2)	19.00	19.00	24.00
Citric Acid	0.49	0.49	0.49
Carbon black	0.2	0.2	0.2
Antioxidant	0.3	0.3	0.3
Potassium Iodide solution, 33%	0.2	0.2	0.2
Cuprous Iodide	0.01	0.01	0.01
PPA (as described in Table 2)	45.00	50.00	45.00
Glass fiber (as described in Table 2)	35.00	30.00	30.00

[0038] The composition can be prepared using various techniques, including batch or continuous techniques that employ kneaders, extruders, mixers, and the like. For example, the composition can be formed as a melt blend employing a twin-screw extruder. In some embodiments at least some of the components are added sequentially. For example, the poly(phenylene ether) and functionalizing agent may be added to the extruder at the feed throat or in feeding sections adjacent to the feed throat, while the polyphthalamide may be added to the extruder in a subsequent feeding section downstream. When a functionalized poly(phenylene ether) is used the functionalized poly(phenylene ether) may be added to the extruder at the feed throat or in feeding sections adjacent to the feed throat, while the polyphthalamide may be added to the extruder in a subsequent feeding section downstream. A vacuum system may be applied to the extruder, prior to the second sequential addition, to generate a sufficient vacuum to lower the residual levels of non-reacted functionalizing agent and any other volatile materials. In an alternative embodiment, the sequential addition of the components may be accomplished through multiple extrusions. A composition may be made by preextrusion of selected components, such as the poly(phenylene ether) and the functionalizing agent to produce a pelletized mixture. A second extrusion may then be

employed to combine the preextruded components with the remaining components. The extruder may be a two lobe or three lobe twin screw extruder.

[0039] The composition can be used to make articles such as engine fluid handling components, engine cooling components such as hoses, pumps, manifolds, turbo outlets, and fluid meters.

[0040] Embodiment 1: A reinforced composition comprising 55 to 80 wt% of a compatibilized blend of a polyphthalamide and a poly(phenylene ether) wherein said compatibilized blend is formed from a mixture of polyphthalamide and poly(phenylene ether) in a ratio of between 1.5: 1 and 7.0:1, and a functionalizing agent in an amount sufficient to effect compatibilization; and 20 to 45 wt% of a nylon glass fiber, wherein the composition contains less than 0.1 wt% of phosphinates and less than 0.1 wt% of impact modifiers and weight percent is based on the total weight of the composition.

[0041] Embodiment 2: The reinforced composition of Embodiment 1, wherein the polyphthalamide is present in an amount of 30 to 70 weight percent and the poly(phenylene ether) is present in an amount of 10 to 30 weight percent, and weight percent is based on the total weight of the composition.

[0042] Embodiment 3: The reinforced composition of Embodiment 1 or 2, wherein the polyphthalamide has a melting point greater than 290°C and a glass transition temperature greater than or equal to 80°C.

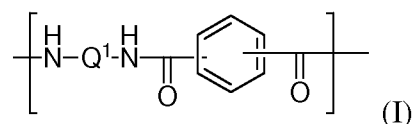
[0043] Embodiment 4: The reinforced composition of any of Embodiments 1-3, wherein the composition is substantially free of added aliphatic polyamides.

[0044] Embodiment 5: The reinforced composition of any of Embodiments 1-4, wherein the glass fiber has an average diameter of 8 to 16 micrometers.

[0045] Embodiment 6: The reinforced composition of Embodiments 1-5, wherein the glass fiber has an average length of 2.8 to 3.6 millimeters.

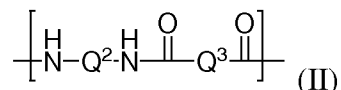
[0046] Embodiment 7: The reinforced composition of any of Embodiments 1-6, wherein the poly(phenylene ether) comprises 2,6-dimethyl-1,4-phenylene ether units.

[0047] Embodiment 8: The reinforced composition of any of Embodiments 1-7, wherein the polyphthalamide comprises (a) 60-70 mol% of units of formula (I)



wherein Q¹ is a 1,6-hexyl group and the aromatic portion of the repeating unit is derived from terephthalic acid, (b) 20-30 mol% of units of formula (I) wherein Q¹ is a 1,6-

hexyl group and the aromatic portion of the repeating unit is derived from isophthalic acid, and (c) 5-15 mol% of units of formula (II)



wherein Q^2 is a 1,4-butyl group and Q^3 is a 1,6-hexyl group.

[0048] Embodiment 9: The reinforced composition of any of Embodiments 1-8, wherein the functionalizing agent comprises maleic anhydride, fumaric acid, or citric acid.

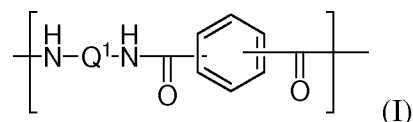
[0049] Embodiment 10: The reinforced composition of any of Embodiments 1-9, wherein the functionalizing agent is citric acid.

[0050] Embodiment 11: The reinforced composition of any of Embodiments 1-10, wherein the citric acid is used in an amount of 0.2 to 0.9 weight percent based on the total weight of the composition.

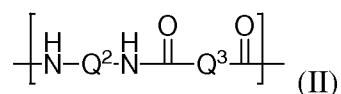
[0051] Embodiment 12: The reinforced composition of any of Embodiments 1-11, further comprising a black pigment.

[0052] Embodiment 13: The reinforced composition of Embodiment 12, wherein the black pigment is carbon black and is present in an amount up to 0.5 weight percent, based on the total weight of the composition..

[0053] Embodiment 14: A reinforced composition comprising 55 to 80 wt% of a compatibilized blend of a polyphthalamide comprising (a) 60-70 mol% of units of formula (I)



wherein Q^1 is a 1,6-hexyl group and the aromatic portion of the repeating unit is derived from terephthalic acid, (b) 20-30 mol% of units of formula (I) wherein Q^1 is a 1,6-hexyl group and the aromatic portion of the repeating unit is derived from isophthalic acid, and (c) 5-15 mol% of units of formula (II)



wherein Q^2 is a 1,4-butyl group and Q^3 is a 1,6-hexyl group, and a poly(phenylene ether) comprising 2,6-dimethyl-1,4-phenylene ether units wherein said compatibilized blend is formed from a mixture of said polyphthalamide and poly(phenylene ether) in a ratio of between 1.5: 1 and 7.0:1, and 0.2 to 0.9 weight percent citric acid; and 20 to 45 wt% of a nylon glass fiber, wherein the composition contains less than 0.1 wt% of phosphinates and

less than 0.1 wt% of impact modifiers and weight percent is based on the total weight of the composition.

[0054] Embodiment 15: The composition of Embodiment 14, wherein the polyphthalamide is present in an amount of 30 to 70 weight percent, the poly(phenylene ether) is present in an amount of 10 to 30 weight percent, and weight percent is based on the total weight of the composition.

[0055] Embodiment 16: The reinforced composition of any of Embodiments 14-15, wherein the polyphthalamide has a melting point greater than 290°C and a glass transition temperature greater than or equal to 80°C.

[0056] Embodiment 17: The reinforced composition of any of Embodiments 14-16, wherein the composition is substantially free of added aliphatic polyamides.

[0057] Embodiment 18: The reinforced composition of any of Embodiments 14-17, wherein the glass fiber has an average diameter of 8 to 16 micrometers.

[0058] Embodiment 19: The reinforced composition of any of Embodiments 14-18, wherein the glass fiber has an average length of 2.8 to 3.6 millimeters.

[0059] Embodiment 20: The reinforced composition of any of Embodiments 14-19, further comprising a black pigment.

[0060] Embodiment 21: The reinforced composition of Embodiment 20, wherein the black pigment is carbon black and is present in an amount up to 0.5 weight percent, based on the total weight of the composition.

[0061] Embodiment 22: An article comprises a reinforced composition comprising 55 to 80 wt% of a compatibilized blend of a polyphthalamide and a poly(phenylene ether) wherein said compatibilized blend is formed from a mixture of polyphthalamide and poly(phenylene ether) in a ratio of between 1.5:1 and 7.0:1, and a functionalizing agent in an amount sufficient to effect compatibilization; and 20 to 45 wt% of a nylon glass fiber, wherein the composition contains less than 0.1 wt% of phosphinates and less than 0.1 wt% of impact modifiers and weight percent is based on the total weight of the composition.

[0062] Embodiment 23: The article of Embodiment 22, wherein the polyphthalamide is present in an amount of 30 to 70 weight percent and the poly(phenylene ether) is present in an amount of 10 to 30 weight percent, and weight percent is based on the total weight of the composition.

[0063] Embodiment 24: The article of any of Embodiments 22-23, wherein the polyphthalamide has a melting point greater than 290°C and a glass transition temperature greater than or equal to 80°C.

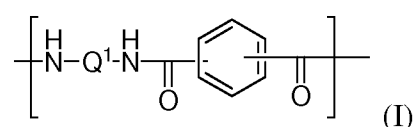
[0064] Embodiment 25: The article of any of Embodiments 22-24, wherein the composition is substantially free of added aliphatic polyamides.

[0065] Embodiment 26: The article of any of Embodiments 22-25, wherein the glass fiber has an average diameter of 8 to 16 micrometers.

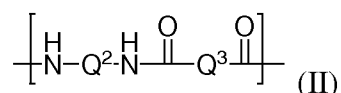
[0066] Embodiment 27: The article of any of Embodiments 22-26, wherein the glass fiber has an average length of 2.8 to 3.6 millimeters.

[0067] Embodiment 28: The article of any of Embodiments 22-27, wherein the poly(phenylene ether) comprises 2,6-dimethyl-1,4-phenylene ether units.

[0068] Embodiment 29: The article of any of Embodiments 22-28, wherein the polyphthalamide comprises (a) 60-70 mol% of units of formula (I)



wherein Q^1 is a 1,6-hexyl group and the aromatic portion of the repeating unit is derived from terephthalic acid, (b) 20-30 mol% of units of formula (I) wherein Q^1 is a 1,6-hexyl group and the aromatic portion of the repeating unit is derived from isophthalic acid, and (c) 5-15 mol% of units of formula (II)



wherein Q^2 is a 1,4-butyl group and Q^3 is a 1,6-hexyl group.

[0069] Embodiment 30: The article of any of Embodiments 22-29, wherein the functionalizing agent comprises maleic anhydride, fumaric acid, or citric acid.

[0070] Embodiment 31: The article of any of Embodiments 22-30, wherein the functionalizing agent is citric acid.

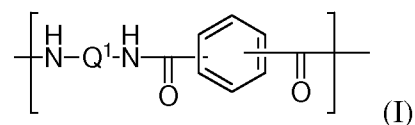
[0071] Embodiment 32: The article of any of Embodiments 22-31, wherein the citric acid is used in an amount of 0.2 to 0.9 weight percent based on the total weight of the composition.

[0072] Embodiment 33: The article of any of Embodiments 22-32, wherein the composition further comprises a black pigment.

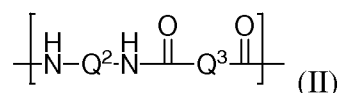
[0073] Embodiment 34: The article of Embodiment 33, wherein the black pigment is carbon black and is present in an amount up to 0.5 weight percent based on the total weight of the composition.

[0074] Embodiment 35: The article of any of Embodiments 22-34, wherein the article is selected from the group consisting of engine cooling components, engine fluid handling components, hoses, pumps, manifolds, turbo outlets, and fluid meters.

[0075] Embodiment 36: An article comprises 55 to 80 weight percent (wt%) of a compatibilized blend of a polyphthalamide comprising (a) 60-70 mol% of units of formula (I)



wherein Q^1 is a 1,6-hexyl group and the aromatic portion of the repeating unit is derived from terephthalic acid, (b) 20-30 mol% of units of formula (I) wherein Q^1 is a 1,6-hexyl group and the aromatic portion of the repeating unit is derived from isophthalic acid, and (c) 5-15 mol% of units of formula (II)



wherein Q^2 is a 1,4-butyl group and Q^3 is a 1,6-hexyl group, and a poly(phenylene ether) comprising 2,6-dimethyl-1,4-phenylene ether units wherein said compatibilized blend is formed from a mixture of said polyphthalamide and poly(phenylene ether) in a ratio of between 1.5: 1 and 7.0:1, and 0.2 to 0.9 weight percent citric acid; and 20 to 45 wt% of a nylon glass fiber, wherein the composition contains less than 0.1 wt% of phosphinates and less than 0.1 wt% of impact modifiers and weight percent is based on the total weight of the composition.

[0076] Embodiment 37: The article of Embodiment 36, wherein the polyphthalamide is present in an amount of 30 to 70 weight percent, the poly(phenylene ether) is present in an amount of 10 to 30 weight percent, and weight percent is based on the total weight of the composition.

[0077] Embodiment 38: The article of any of Embodiments 36-37, wherein the polyphthalamide has a melting point greater than 290°C and a glass transition temperature greater than or equal to 80°C.

[0078] Embodiment 39: The article of any of Embodiments 36-38, wherein the composition is substantially free of added aliphatic polyamides.

[0079] Embodiment 40: The article of any of Embodiments 36-39, wherein the glass fiber has an average diameter of 8 to 16 micrometers.

[0080] Embodiment 41: The article of any of Embodiments 36-40, wherein the glass fiber has an average length of 2.8 to 3.6 millimeters.

[0081] Embodiment 42: The article of any of Embodiments 36-41, wherein the composition further comprises a black pigment.

[0082] Embodiment 43: The article of Embodiment 42, wherein the black pigment is carbon black and is present in an amount up to 0.5 weight percent, based on the total weight of the composition..

[0083] Embodiment 44: The article of any of Embodiments 36-43, wherein the article wherein is selected from the group consisting of engine cooling components, engine fluid handling components, hoses, pumps, manifolds, turbo outlets, and fluid meters

EXAMPLES

[0084] The examples used the materials described in Table 2 and in the following paragraphs.

Table 2.

Component	Grade	Supplier	Description
PPE	PPO® (0.40IV)	Sabic Innovative Plastics, US LLC	Poly(2,6-dimethyl-1,4-phenylene)ether with an intrinsic viscosity of 0.40 dl/g.
Citric Acid	Citric Acid	International Chemical Incorporated	Citric Acid – functionalizing agent
Anti-oxidant	Irganox 1010	Great Lakes Chemical Corporation	Heat Stabilizer
Liquid potassium iodide	Liquid KI	Ajay North America Incorporated	Heat Stabilizer
Cuprous Iodide	Cuprous Iodide	Ajay North America Incorporated	Heat Stabilizer
PA 6/6	Vydyne 21Z	Ascend Inc.	Polyamide 6/6
PA 6/6 ground	Vydyne 21Z	Ground version of PA 6/6; all particles passed through a 20 mesh screen	
Polyphthalamide	Amodel A-1006C	Solvay Advanced Polymers	Semi-aromatic nylon
Polyphthalamide ground	Amodel A-1006C	Ground version of polyphthalamide, all particles passed through a 20 mesh screen	
Glass Fibers	ChopVantage® HP 3540	PPG	Chopped glass fibers with average length of 3.2 millimeters and an average diameter of 10 micrometers

[0085] Polyphthalamide, Amodel A-1006C, is a copolymer comprising three different repeating units in the following amounts: (a) 60-70 mol% of units of formula (I) wherein Q^1 is a 1,6-hexyl group and the aromatic portion of the repeating unit is derived from terephthalic acid, (b) 20-30 mol% of units of formula (I) wherein Q^1 is a 1,6-hexyl group and the aromatic portion of the repeating unit is derived from isophthalic acid, and (c) 5-15 mol% of units of formula (II) wherein Q^2 is a 1,4-butyl group and Q^3 is a 1,6-hexyl group. The

amounts and structure of the repeating units was determined using nuclear magnetic resonance spectroscopy.

[0086] The Examples also contained 0.7 weight percent of a combination of additives (Irganox 1010, potassium iodide, copper iodide, and carbon black).

[0087] The examples were made by melt blending the poly(phenylene ether), citric acid, additives, polyphthalamide or polyamide to form a first melt mixture and melt mixing the masterbatch and glass fibers with the first melt mixture in a 30 millimeter Werner Pfleider twin screw extruder. The extruder for the polyamide 66 examples was set with barrel temperatures of 249-288°C and a die temperature of 288°C, with the screw rotating at 240 rotations per minute (rpm) and a rate of about 18 kilograms per hour. The extruder for the polyphthalamide examples was set with barrel temperatures of 260-307°C and a die temperature of 343°C, with the screw rotating at 240 rotations per minute (rpm) and a rate of about 18 kilograms per hour. The amounts of the components of the compositions are shown in Table 3. Amounts are in weight percent based on the total weight of the compositions.

Table 3.

	1*	2*	*3	4*	5	6
PPE	-	10	20	-	10	20
CA	-	0.7	0.7	-	0.7	0.7
PA 6/6 ground	5	-	-	-	-	-
Polyphthalamide ground	-	-	-	5	-	-
Polyphthalamide	-	-	-	60	55	45
PA 6/6	60	55	45	-	-	-
Glass Fibers	35	35	35	35	35	35

*Comparative Example

[0088] The compositions were molded and tested. Testing methods are shown in Table 4. The compositions were tested after three time period/condition combinations – after molding, after immersion in 23C water for 13 and 31 days. The change in physical properties after immersion in 23C water is shown in Table 5.

Table 4

Test Name	Method
Notched Izod	ISO 180
Dynatup total energy	ASTM D3763
Chord Modulus	ISO 527
Stress @ Break	ISO 527
Strain @ Break	ISO 527

Table 5

	1*	2*	*3	4*	5	6
Moisture content (%)						
0 days immersion	0.1	0.1	0.1	0.1	0.1	0.1
13 days immersion	1.6	1.3	1.2	0.6	0.6	0.5
30 days immersion	2.5	2.1	1.9	0.8	0.9	0.9
Notched Izod @23C (kJ/m ²)						
0 days immersion	9.6	9.4	9.5	8.5	8.1	7.0
13 days immersion	11.6	10.5	10.5	8.1	7.8	7.4
30 days immersion	13.2	11.8	11.8	7.6	7.8	7.3
Dynatup (J)						
0 days immersion	8.9	9.0	9.1	7.9	9.8	9.8
13 days immersion	15.2	12.5	14.1	8.4	10.6	10.1
30 days immersion	21.7	20.2	15.8	8.5	10.6	11.8
Chord Modulus (MPa)						
0 days immersion	11784	11776	11854	12718	12344	12228
13 days immersion	9188	8966	9126	12600	12827	12160
30 days immersion	8372	8044	8343	12793	12620	12143
Stress @ Break (MPa)						
0 days immersion	191	185	182	182	186	187
13 days immersion	139	133	139	170	190	175
30 days immersion	127	121	128	169	184	170
Strain @ Break (%)						
0 days immersion	2.6	2.4	2.4	1.7	1.9	2.1
13 days immersion	2.8	2.5	3.3	1.6	2.0	2.0
30 days immersion	3.0	2.6	3.6	1.6	2.0	2.0

*Comparative example

[0089] The reinforced PPE/PPA examples (Examples 5 & 6) showed remarkable retention of stress @ break after immersion in water, especially when compared to reinforced

PPA (Example 4), reinforced PA 6/6 (Example 1), and reinforced PPE/PA66 (Examples 2 &3)

[0090] The compositions were also tested for stress@break both initially and after immersion in 80 °C water for 20 weeks. Results (in MPa) are shown in Table 6.

Table 6

	Ex. 3*	% retention	Ex. 4*	% retention	Ex. 6	% retention
Initial	155		187		172	
1 week	105	68	162	87	144	84
4 weeks	100	65	133	71	134	78
8 weeks	90	58	116	62	135	78
16 weeks	84	54	114	61	133	77
20 weeks	79	51	109	58	124	72

*Comparative example

[0091] The reinforced PPE/PPA composition (Example 6) showed much better retention of tensile strength compared to the reinforced PPE/PA 66 (Example 3) and reinforced PPA (example 4).

[0092] Examples 7-11 were made as described above. The compositions are shown in Table 7. Similar to Examples 1-6 the Examples also contained 0.7 weight percent of a combination of additives (Irganox 1010, potassium iodide, copper iodide, and carbon black).

Table 7

	Ex. 7*	Ex. 8*	Ex. 9	Ex. 10	Ex. 11
PPE	-	-	19.00	19.00	24.00
CA	-	-	0.49	0.49	0.49
Polyphthalamide ground	5.00	5.00	-	-	-
Polyphthalamide	59.49	64.49	45.00	50.00	45.00
Glass Fibers	35.00	30.00	35.00	30.00	30.00

*Comparative Example

[0093] The compositions were molded and tested for notched Izod (see Table 4 for the testing method). The compositions were tested after molding and after exposure to heat aging for the time periods shown in Table 8.

Table 8

Aging Temp (degree C)	Aging time (hr)	Ex. 7*	% retention	Ex. 8*	% retention	Ex. 9	% retention	Ex. 10	% retention	Ex. 11	% retention
Initial	Initial	9.0	-	8.4		9.7		8.3		8.7	
190	250	8.9	99	8.5	101	9.9	103	8.7	104	8.8	102
190	500	9.2	102	8.5	101	9.7	101	8.5	102	8.8	101
210	250	9.2	102	8.4	100	9.7	100	8.4	101	8.5	98
210	500	9.1	101	8.4	101	10.0	103	8.4	101	8.3	96
230	250	8.8	98	8.1	97	9.9	103	8.4	101	8.7	100
230	500	8.8	98	8.0	95	9.5	99	8.1	98	8.3	95
250	250	7.9	88	7.9	95	9.4	97	8.3	100	8.3	95
250	500	5.8	64	5.2	62	9.6	99	8.2	99	8.0	92

*Comparative Example

[0094] As can be seen from the extended aging at high temperatures, Examples 9-11 retain much more notched Izod impact strength than the comparative Examples 7-8.

[0095] In the specification and the claims, reference is made to a number of terms, which shall be defined to have the following meanings. The terms “first,” “second,” and the like, “primary,” “secondary,” and the like, “(a),” “(b)” and the like, as used herein do not denote any order, quantity, or importance, but rather are used to distinguish one element from another. The endpoints of all ranges directed to the same component or property are inclusive of the endpoint and independently combinable. Reference throughout the specification to “one embodiment,” “another embodiment,” “an embodiment,” “some embodiments,” and so forth, means that a particular element (e.g., feature, structure, property, and/or characteristic) described in connection with the embodiment is included in at least one embodiment described herein, and may or may not be present in other embodiments. In addition, it is to be understood that the described element(s) may be combined in any suitable manner in the various embodiments. The singular forms “a,” “an” and “the” include plural referents unless the context clearly dictates otherwise. “Optional” or “optionally” means that the subsequently described event or circumstance may or may not occur, and that the description includes instances where the event occurs and instances where it does not.

[0096] While the invention has been illustrated and described in typical embodiments, it is not intended to be limited to the details shown, since various modifications and substitutions can be made without departing in any way from the spirit of the present invention. As such, further modifications and equivalents of the invention herein disclosed may occur to persons skilled in the art using no more than routine experimentation, and all such modifications and equivalents are believed to be within the spirit and scope of the

invention as defined by the following claims. All patents and published articles cited herein are incorporated herein by reference.

CLAIMS:

1: A reinforced composition comprising 55 to 80 wt% of a compatibilized blend of a polyphthalamide and a poly(phenylene ether) wherein said compatibilized blend is formed from a mixture of polyphthalamide and poly(phenylene ether) in a ratio of between 1.5: 1 and 7.0:1, and a functionalizing agent in an amount sufficient to effect compatibilization; and 20 to 45 wt% of a nylon glass fiber, wherein the composition contains less than 0.1 wt% of phosphinates and less than 0.1 wt% of impact modifiers and weight percent is based on the total weight of the composition.

2: The reinforced composition of claim 1, wherein the polyphthalamide is present in an amount of 30 to 70 weight percent and the poly(phenylene ether) is present in an amount of 10 to 30 weight percent, and weight percent is based on the total weight of the composition.

3: The reinforced composition of claim 1 or 2, wherein the polyphthalamide has a melting point greater than 290°C and a glass transition temperature greater than or equal to 80°C.

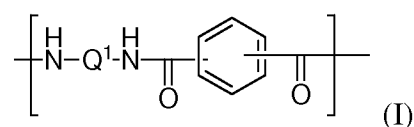
4: The reinforced composition of any of claims 1-3, wherein the composition is substantially free of added aliphatic polyamides.

5: The reinforced composition of any of claims 1-4, wherein the glass fiber has an average diameter of 8 to 16 micrometers.

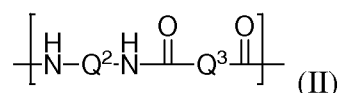
6: The reinforced composition of claims 1-5, wherein the glass fiber has an average length of 2.8 to 3.6 millimeters.

7: The reinforced composition of any of claims 1-6, wherein the poly(phenylene ether) comprises 2,6-dimethyl-1,4-phenylene ether units.

8: The reinforced composition of any of claims 1-7, wherein the polyphthalamide comprises (a) 60-70 mol% of units of formula (I)



wherein Q¹ is a 1,6-hexyl group and the aromatic portion of the repeating unit is derived from terephthalic acid, (b) 20-30 mol% of units of formula (I) wherein Q¹ is a 1,6-hexyl group and the aromatic portion of the repeating unit is derived from isophthalic acid, and (c) 5-15 mol% of units of formula (II)



wherein Q² is a 1,4-butyl group and Q³ is a 1,6-hexyl group.

9: The reinforced composition of any of claims 1-8, wherein the functionalizing agent comprises maleic anhydride, fumaric acid, or citric acid.

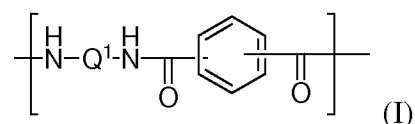
10: The reinforced composition of any of claims 1-9, wherein the functionalizing agent is citric acid.

11: The reinforced composition of any of claims 1-10, wherein the citric acid is used in an amount of 0.2 to 0.9 weight percent based on the total weight of the composition.

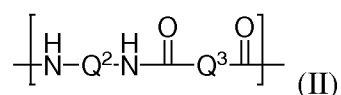
12: The reinforced composition of any of claims 1-11, further comprising a black pigment.

13: The reinforced composition of claim 12, wherein the black pigment is carbon black and is present in an amount up to 0.5 weight percent, based on the total weight of the composition..

14: A reinforced composition comprising 55 to 80 wt% of a compatibilized blend of a polyphthalamide comprising (a) 60-70 mol% of units of formula (I)



wherein Q¹ is a 1,6-hexyl group and the aromatic portion of the repeating unit is derived from terephthalic acid, (b) 20-30 mol% of units of formula (I) wherein Q¹ is a 1,6-hexyl group and the aromatic portion of the repeating unit is derived from isophthalic acid, and (c) 5-15 mol% of units of formula (II)



wherein Q² is a 1,4-butyl group and Q³ is a 1,6-hexyl group, and a poly(phenylene ether) comprising 2,6-dimethyl-1,4-phenylene ether units wherein said compatibilized blend is formed from a mixture of said polyphthalamide and poly(phenylene ether) in a ratio of between 1.5: 1 and 7.0:1, and 0.2 to 0.9 weight percent citric acid; and 20 to 45 wt% of a nylon glass fiber, wherein the composition contains less than 0.1 wt% of phosphinates and less than 0.1 wt% of impact modifiers and weight percent is based on the total weight of the composition.

15: The composition of claim 14, wherein the polyphthalamide is present in an amount of 30 to 70 weight percent, the poly(phenylene ether) is present in an amount of 10 to 30 weight percent, and weight percent is based on the total weight of the composition.

16: The reinforced composition of any of Embodiments 14-15, wherein the polyphthalamide has a melting point greater than 290°C and a glass transition temperature greater than or equal to 80°C.

17: The reinforced composition of any of claims 14-16, wherein the composition is substantially free of added aliphatic polyamides.

18: The reinforced composition of any of claims 14-17, wherein the glass fiber has an average diameter of 8 to 16 micrometers.

19: The reinforced composition of any of claims 14-18, wherein the glass fiber has an average length of 2.8 to 3.6 millimeters.

20: The reinforced composition of any of claims 14-19, further comprising a black pigment.

21: The reinforced composition of claim 20, wherein the black pigment is carbon black and is present in an amount up to 0.5 weight percent, based on the total weight of the composition.

22: An article comprising the reinforced composition of any of claims 1 to 13.

23: The article of claim 22, wherein the article is selected from the group consisting of engine cooling components, engine fluid handling components, hoses, pumps, manifolds, turbo outlets, and fluid meters.

24: An article comprising the reinforced composition of any of claims 14-21.

25: The article of claim 24, wherein the article wherein is selected from the group consisting of engine cooling components, engine fluid handling components, hoses, pumps, manifolds, turbo outlets, and fluid meters

INTERNATIONAL SEARCH REPORTInternational application No.
PCT/US2014/020236**Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)**

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☒ Claims Nos.: 13, 21, 23 and 25
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:
Claims 13, 21, 23 and 25 are unclear because they refer to a multiple dependent claim which does not comply with PCT Rule 6.4(a).
3. ☒ Claims Nos.: 4-12, 17-20, 22 and 24
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying an additional fees, this Authority did not invite payment of any additional fees.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest and, where applicable, the payment of a protest fee.
- ☐ The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.
- ☐ No protest accompanied the payment of additional search fees.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US2014/020236**A. CLASSIFICATION OF SUBJECT MATTER****C08L 77/10(2006.01)i, C08L 71/12(2006.01)i, C08G 73/10(2006.01)i, C08G 65/34(2006.01)i**

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 77/10; H01B 1/12; C08L 71/12; C08L 77/00; C08K 3/32; C08K 3/34; C08K 5/53; C08K 5/49; C08G 73/10; C08G 65/34

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

Korean utility models and applications for utility models

Japanese utility models and applications for utility models

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

eKOMPASS(KIPO internal) & Keywords:polyphthalamide, phenylene ether, reinforce, fiber

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2009-0242844 A1 (ELKOVITCH, MARK) 01 October 2009 See abstract; claims 1, 4-7, 13, 14; and paragraphs [0003], [0016], [0017], [0022], [0031], [0034].	1-3, 14-16
A	US 5916970 A (LEE JR, GIM FUN et al.) 29 June 1999 See abstract; and claims 1, 4.	1-3, 14-16
A	US 2006-0111484 A1 (FISHBURN, JAMES ROSS) 25 May 2006 See abstract; and claims 1, 31, 32.	1-3, 14-16
A	US 2006-0041046 A1 (MHETAR, VIJAY R. et al.) 23 February 2006 See abstract; and claims 1, 17, 18.	1-3, 14-16
A	US 2009-0292051 A1 (LI, YANJUN FRANK et al.) 26 November 2009 See abstract; and claims 1, 2.	1-3, 14-16



Further documents are listed in the continuation of Box C.



See patent family annex.

* Special categories of cited documents:

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

"E" earlier 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

"&" document member of the same patent family

Date of the actual completion of the international search

18 June 2014 (18.06.2014)

Date of mailing of the international search report

18 June 2014 (18.06.2014)

Name and mailing address of the ISA/KR

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

PCT/US2014/020236

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

PCT/US2014/020236Patent document
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date

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WO 2009-141800 A3	04/11/2010