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(21) International Application Number: PCT/US99/05149 (22) International Filing Date: 8 March 1999 (08.03.99) (30) Priority Data: 09/045,300 20 March 1998 (20.03.98) US (71) Applicant: THE DOW CHEMICAL COMPANY [US/US]; 2030 Dow Center, Midland, MI 48674 (US). (72) Inventors: CHEUNG, C., Alice; 510 That Way #810, Lake Jackson, TX 77566 (US). POLANSKY, Christine, A.; Route 1, Box 1722, Brazoria, TX 77422 (US). WHITE, Jerry, E.; 418 Huckleberry Drive, Lake Jackson, TX 77566 (US). (74) Agent: DAMOCLES, Nemia, C.; Patent Dept., P.O. Box 1967, Midland, MI 48641-1967 (US).		(81) Designated States: AU, BR, CA, CN, CZ, IN, JP, KR, MX, PL, RU, European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE). Published <i>With international search report.</i>
(54) Title: POLYMER COMPOSITE COMPRISING A HYDROXY-FUNCTIONALIZED POLYETHER OR POLYESTER AND AN INORGANIC FILLER AND METHOD FOR PREPARING THE SAME (57) Abstract A polymer composite comprising a hydroxy-phenoxyether or hydroxy-phenoxy ester polymer matrix and an inorganic filler is prepared by melt-blending the polymer and the inorganic filler. The filler may be an inorganic metal oxide, metal hydroxide, metal carbonate, metal nitride, metal carbide or metal boride. Methods of polymer composite preparation involving <i>in situ</i> type polymerization may also be used. The polymer composites have increased barrier capabilities compared to the unmodified polymers.		

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POLYMER COMPOSITE COMPRISING A HYDROXY-FUNCTIONALIZED
POLYETHER OR POLYESTER AND AN INORGANIC FILLER
AND METHOD FOR PREPARING THE SAME

5 The present invention relates to a polymer composite comprising a polymer
and an inorganic additive, and to a method for preparing the polymer composite.

Polymer composites comprising a polymer matrix having one or more additives, such as a particulate or fiber material dispersed throughout the continuous polymer matrix, are well known. The additive is often added to enhance one or more properties of the polymer.

10 In one aspect, the present invention is a polymer composite comprising a hydroxy-functionalized polyether or polyester and an inorganic filler.

In a second aspect, the present invention is a method for forming a composite which comprises contacting a hydroxy-functionalized polyether or polyester or a precursor to the polyether or polyester with an inorganic filler.

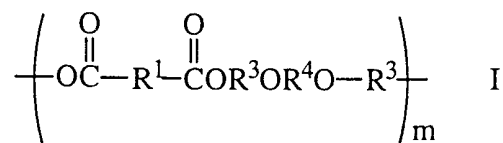
15 In a preferred embodiment, the polymer is a melt processible, thermoplastic hydroxy-functionalized polyether or polyester and the method comprises melt-blending the polymer and the inorganic filler.

20 The polymer composites of this invention can exhibit an excellent balance of properties and can exhibit one or more superior properties such as improved heat or chemical resistance, ignition resistance, superior resistance to diffusion of polar liquids and of gases, yield strength in the presence of polar solvents such as water, methanol, or ethanol, or enhanced stiffness and dimensional stability, as compared to polymers which do not contain an inorganic filler.

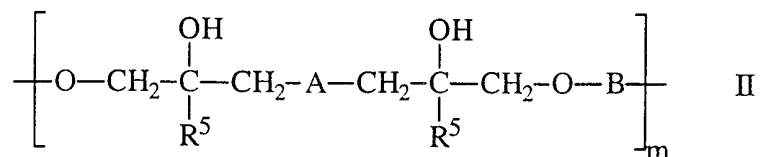
25 The polymer composites of the present invention are useful as barrier films, barrier foams, or other molded or extruded thermoplastic articles using any conventional thermoplastic fabrication methods. The articles can be used in a wide variety of applications including transportation (for example, automotive and aircraft) parts, electronics, business equipment such as computer housings, building and construction materials, and packaging material.

30 Preferably, the polymer matrix of the polymer composite comprises the following hydroxy-functionalized polyether or polyester:

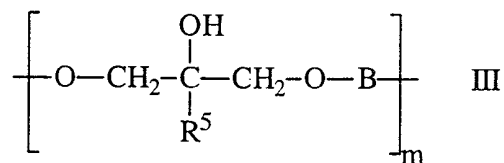
(1) poly(hydroxy ester ethers) having repeating units represented by the formula:



(2) polyetheramines having repeating units represented by the formula:

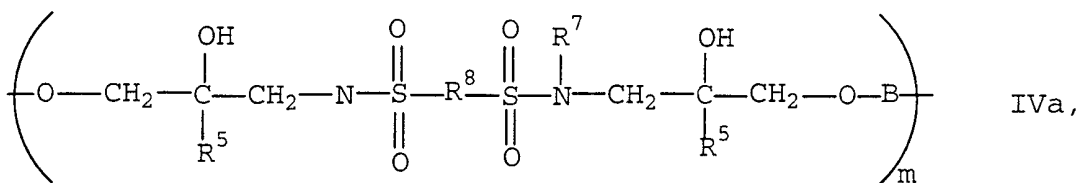


5 (3) hydroxy-phenoxyether polymers having repeating units represented by the formula:

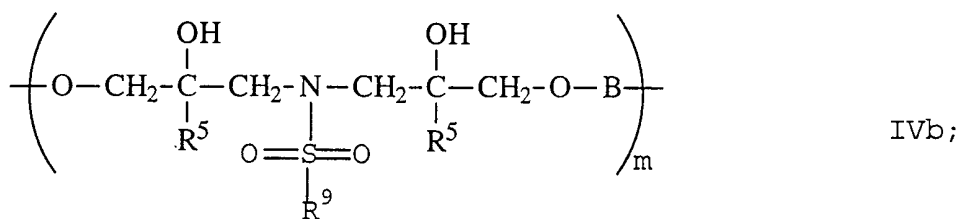


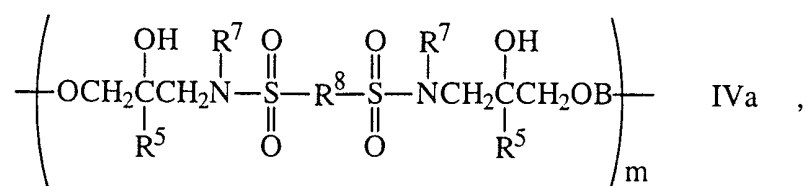
or

(4) hydroxy-functional poly(ether sulfonamides) having repeating units represented by the formula:

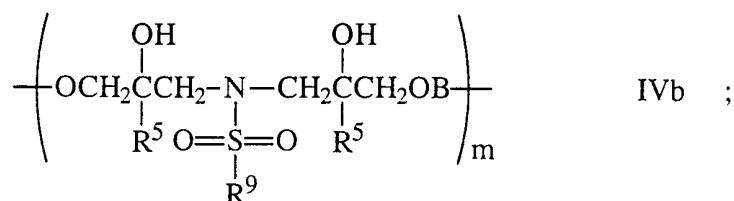


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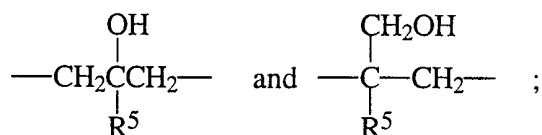




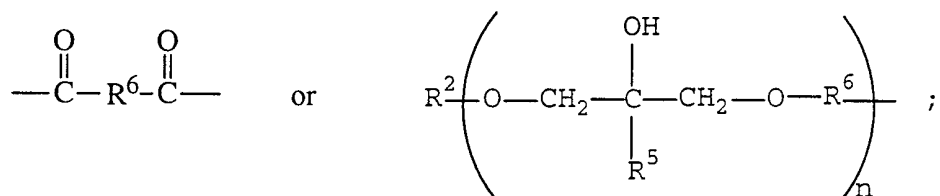
or



wherein R¹ is a divalent organic moiety which is primarily hydrocarbon; R² is independently a divalent organic moiety which is primarily hydrocarbon; R³ is



5 R⁴ is



R⁵ is hydrogen or alkyl; R⁶ is a divalent organic moiety which is primarily hydrocarbon; R⁷ and R⁹ are independently alkyl, substituted alkyl, aryl, substituted aryl; R⁸ is a divalent organic moiety which is primarily hydrocarbon; A is an amine moiety or a combination of different amine moieties; B is a divalent organic moiety which is primarily hydrocarbon; m is an integer from 5 to 1000; and n is an integer from 0 to 100.

In the preferred embodiment of the present invention, A is 2-hydroxyethylimino-, 2-hydroxypropylimino-, piperazenyl, N,N'-bis(2-hydroxyethyl)-1,2-ethylenediimino; and B and R¹ are independently 1,3-phenylene, 1,4-phenylene; 10 sulfonyldiphenylene, oxydiphenylene, thiodiphenylene or isopropylidene-diphenylene; R⁵ is hydrogen; R⁷ and R⁹ are independently methyl, ethyl, propyl, butyl, 2-hydroxyethyl or phenyl; and B and R⁸ are independently 1,3-phenylene, 1,4-phenylene, sulfonyldiphenylene, 15 oxydiphenylene, thiodiphenylene or isopropylidenediphenylene.

The poly(hydroxy ester ethers) represented by Formula I are prepared by reacting diglycidyl ethers of aliphatic or aromatic diacids, such as diglycidyl terephthalate, or diglycidyl ethers of dihydric phenols with, aliphatic or aromatic diacids such as adipic acid or isophthalic acid. These polyesters are described in U.S. Patent 5,171,820. Alternatively, the poly(hydroxy ester ethers) are prepared by reacting a diglycidyl ester with a bisphenol or by reacting a diglycidyl ester or an epihalohydrin with a dicarboxylic acid.

The polyetheramines represented by Formula II are prepared by contacting one or more of the diglycidyl ethers of a dihydric phenol with an amine having two amine hydrogens under conditions sufficient to cause the amine moieties to react with epoxy moieties to form a polymer backbone having amine linkages, ether linkages and pendant hydroxyl moieties. These polyetheramines are described in U.S. Patent 5,275,853. The polyetheramines can also be prepared by contacting a diglycidyl ether or an epihalohydrin with a difunctional amine.

The hydroxy-phenoxyether polymers represented by Formula III are prepared, for example, by contacting an epihalohydrin or a diglycidyl ether with a bisphenol. These polymers are described in U.S. Patent 5,496,910.

The hydroxy-functional poly(ether sulfonamides) represented by Formulae IVa and IVb are prepared, for example, by polymerizing an N,N'-dialkyl or N,N'-diaryldisulfonamide with a diglycidyl ether as described in U.S. Patent 5,149,768.

The hydroxy-phenoxyether polymers commercially available from Phenoxy Associates, Inc. are also suitable for use in the present invention. These hydroxy-phenoxyether polymers are the condensation reaction products of a dihydric polynuclear phenol, such as bisphenol A, and an epihalohydrin and have the repeating units represented by Formula I wherein Ar is an isopropylidene diphenylene moiety.

The hydroxy-phenoxyether polymers available from Phenoxy Associates, Inc. and the process for preparing them are described in U.S. Patent 3,305,528. U.S. Patent 5,401,814 also describes a process for preparing these hydroxy-phenoxyether polymers.

Inorganic fillers which can be employed in the practice of the present invention for preparing the polymer composite include talc, mica and additional members of the clay mineral family such as montmorillonite, hectorite, kaolinite, dickite, nacrite, halloysite, saponite, nontronite, beidellite, volhonskoite, sauconite, magadiite, medmontite, kenyaite, vermiculite, serpentines, chlorites, palygorskite, kulkeite, aliettite, sepiolite, allophane and imogolite. In the practice of the present invention, naturally occurring

members of the clay mineral family or synthetic members of the clay mineral family may be used. Mixtures of one or more such materials may also be employed.

Metal oxide, metal carbonate or metal hydroxide materials can also be used as fillers in the practice of the present invention. Such materials include calcium oxide,
5 magnesium oxide, zirconium oxide, titanium oxide, manganese oxide, iron oxide, aluminum oxide, calcium hydroxide, magnesium hydroxide, zirconium hydroxide, aluminum hydroxide, manganese hydroxide, iron hydroxide, calcium carbonate, magnesium carbonate, manganese carbonate, iron carbonate or zirconium carbonate.

Metal nitride, metal carbide and metal boride materials such as aluminum
10 nitride, silicon nitride, iron nitride, silicon carbide, manganese carbide, iron carbide, iron boride, aluminum boride, manganese boride or other materials used in the preparation of ceramic materials may also be used in the practice of the present invention for preparing the polymer composite. Aluminum oxide or aluminum hydroxide such as gibbsite, bayerite, nordstrandite, boehmite, diaspore and corundum may also be used as inorganic fillers in the
15 practice of the present invention. Mixtures of one or more such materials may also be employed.

Preferred inorganic fillers are talc, mica, calcium carbonate and silica coated aluminum nitride (SCAN). Most preferred inorganic fillers are talc and mica.

In general, the composite of the present invention can be prepared by
20 dispersing the inorganic filler in the monomer(s) which form the polymer matrix and the monomer(s) polymerized *in situ* or alternatively, can be dispersed in the hydroxy-phenoxyether or hydroxy-phenoxyester polymer, in melted or liquid form.

Melt-blending is one method for preparing the composites of the present invention. Techniques for melt-blending of a polymer with additives of all types are known in
25 the art and can typically be used in the practice of this invention. Typically, in a melt-blending operation useful in the practice of the present invention, the hydroxy-phenoxy ether or hydroxy-phenoxy ester polymer is heated to a temperature sufficient to form a polymer melt and combined with the desired amount of the inorganic filler material in a suitable mixer, such as an extruder, a Banbury mixer, a Brabender mixer, or a continuous mixer. A physical
30 mixture of the different components may also be heated simultaneously and blended using one of the previously mentioned methods.

In the practice of the present invention, the melt-blending is preferably carried out in the absence of air, as for example, in the presence of an inert gas, such as argon,

neon, or nitrogen. However, the present invention may be practiced in the presence of air. The melt-blending operation can be conducted in a batch or discontinuous fashion but is more preferably conducted in a continuous fashion in one or more processing zones such as in an extruder from which air is largely or completely excluded. The extrusion can be
5 conducted in one zone, or in a plurality of reaction zones which are in series or parallel.

A hydroxy-functionalized polyether or hydroxy-functionalized polyester melt containing the inorganic filler may also be formed by reactive melt processing in which the inorganic filler is initially dispersed in a liquid or solid monomer or cross-linking agent which will form or be used to form the polymer matrix of the composite. This dispersion can be
10 injected into a polymer melt containing one or more polymers in an extruder or other mixing device. The injected liquid may result in new polymer or in chain extension, grafting or even cross-linking of the polymer initially in the melt.

Methods for preparing a polymer composite using *in situ* type polymerization are also known in the art and reference is made thereto for the purposes of this invention. In
15 applying this technique to the practice of the present invention, the composite is formed by mixing monomers and/or oligomers with the inorganic filler in the presence or absence of a solvent and subsequently polymerizing the monomer and/or oligomers to form the hydroxy-phenoxyether polymer matrix of the composite. After polymerization, any solvent that is used is removed by conventional means.

20 Alternatively, the polymer may be granulated and dry-mixed with the inorganic filler, and thereafter, the composition heated in a mixer until the hydroxy-phenoxyether polymer is melted to form a flowable mixture. This flowable mixture can then be subjected to a shear in a mixer sufficient to form the desired composite. The polymer may also be heated in the mixer to form a flowable mixture prior to the addition of the inorganic filler. The
25 inorganic filler and polymer are then subjected to a shear sufficient to form the desired composite.

The amount of the inorganic filler most advantageously incorporated into the hydroxy-functionalized polyether or hydroxy-functionalized polyester is dependent on a variety of factors including the specific inorganic material and polymer used to form the
30 composite as well as its desired properties. Typical amounts can range from 0.001 to 90 weight percent of the inorganic filler based on the weight of the total composite. Generally, the composite comprises at least about 0.1, preferably about 1, more preferably about 2, and most preferably about 4 weight percent and less than about 80, preferably about 60, more

preferably about 50 weight percent of the inorganic filler based on the total weight of the composite.

Optionally, the inorganic fillers used in the practice of this invention may contain various other additives such as dispersing agents, antistatic agents, colorants, mold
5 release agents or pigments. The optional additives and their amount employed are dependent on a variety of factors including the desired end use properties.

Optionally, the polymer composites of the present invention may contain various other additives such as nucleating agents, lubricants, plasticizers, chain extenders, colorants, mold release agents, antistatic agents, pigments, or fire retardants. The optional
10 additives and their amounts employed are dependent on a variety of factors including the desired end-use properties.

The polymer composites of this invention exhibit useful properties, such as increased barrier properties to oxygen, water vapor and carbon dioxide. Increases in tensile strength are also observed. Improvements in one or more properties can be obtained even
15 though small amounts of inorganic fillers are employed.

The properties of the polymer composites of the present invention may be further enhanced by post-treatment such as by heat-treating, orienting or annealing the composite at an elevated temperature, conventionally from 80°C to 230°C. Generally, the annealing temperatures will be more than 100°C, preferably more than 110°C, and more
20 preferably more than 120°C, to less than 250°C, preferably less than 220°C, and more preferably less than 180°C.

The polymer composites of the present invention can be molded by conventional shaping processes such as melt-spinning, casting, vacuum molding, sheet molding, injection molding and extruding, melt-blowing, spun-bonding, blow-molding, and co-
25 or multilayer extrusion. Examples of such molded articles include components for technical equipment, apparatus castings, household equipment, sports equipment, bottles, containers, components for the electrical and electronics industries, car components, and fibers. The composites may also be used for coating articles by means of powder coating processes or as hot-melt adhesives.

30 The polymer composites of the present invention may be directly molded by injection molding or heat pressure molding, or mixed with other polymers. Alternatively, it is also possible to obtain molded products by performing the *in situ* polymerization reaction in a mold.

The polymer composites according to the invention are also suitable for the production of sheets and panels using conventional processes such as vacuum or hot-pressing. The sheets and panels can be laminated to materials such as wood, glass, ceramic, metal or other plastics, and outstanding strengths can be achieved using
5 conventional adhesion promoters, for example, those based on vinyl resins. The sheets and panels can also be laminated with other plastic films by coextrusion, the sheets being bonded in the molten state. The surfaces of the sheets and panels, can be finished by conventional methods, for example, by lacquering or by the application of protective films.

The polymer composites of this invention are also useful for fabrication of
10 extruded films and film laminates, as for example, films for use in food packaging. Such films can be fabricated using conventional film extrusion techniques. The films are preferably from 10 to 100, more preferably from 20 to 100, and most preferably from 25 to 75, microns thick.

The polymer composites of the present invention may also be useful in
15 preparing fiber-reinforced composites in which a resin matrix polymer is reinforced with one or more reinforcing materials such as a reinforcing fiber or mat. Fibers which can be employed in the process of the present invention are described in numerous references, such as, for example, U.S. Patent 4,533,693; Kirk-Othmer Ency. Chem. Tech., Aramid Fibers, 213 (J. Wiley & Sons 1978); Kirk-Othmer Ency. Chem., Tech. - Supp., Composites,
20 High Performance, pages 261-263; Ency. Poly. Sci. & Eng. The fibers can be of varying composition, provided that they do not melt as a composite is made therewith. In general, the fibers are chosen so that they provide improvements in physical properties, such as tensile strength, flexural modulus, and electrical conductivity. Thus, high flexural modulus organic polymers such as polyamides, polyimides, aramids, metals, glass and other
25 ceramics, carbon fibers, and graphite fibers, are suitable fiber materials. Examples of glass fibers, include E-glass and S-glass. E-glass is a low alkali, aluminum-borosilicate composition with excellent electrical properties and good strength and modulus. S-glass is a magnesium-aluminosilicate composition with considerably higher strength and modulus. Fiber rovings are also useful. A roving consists of a number of continuous yarns, strands, or
30 tows collected into a parallel bundle with little or no twist.

The following working examples are given to illustrate the invention and should not be construed as limiting its scope. Unless otherwise indicated, all parts and percentages are by weight.

Example 1

Talc (purchased from the Aldrich Chemical Company) and poly(hydroxy amino ether) derived from bisphenol A diglycidyl ether and monoethanolamine henceforth referred to as PHAE resin were combined to yield varying volume percent talc/PHAE composites. The talc and PHAE resin were slowly added to a preheated Haake torque rheometer at low rpm to allow the resin to melt and equilibrate. After complete addition of the sample, the mixer was ramped to 120 rpm. The sample was melt-blended between 100°C and 250°C, between 5 and 60 minutes, between 20 and 200 rpm, more preferably 170°C and 120 rpm, for approximately 10 minutes. After blending, the sample was removed and pressed into films using compression molding.

The samples were then tested for oxygen barrier properties according to ASTM D3985-81. Oxygen concentration was 100 percent.

The oxygen barrier properties of the samples containing the talc filler were vastly improved over that of the pure PHAE resin under the same test conditions. The values are listed in Table I for the blank PHAE resin and four different volume percent loading of talc. The test conditions were 23.7°C, relative humidity of 52 percent at an oxygen concentration of 100 percent.

Table I

<u>Volume % Talc</u>	<u>Oxygen Transmission Rate (cc-mil/100 in²-day-atm O₂)</u>
0	0.773, 0.771
5	0.441, 0.461
10	0.260, 0.256
15	0.114, 0.146
20	0.103, 0.101

Example 2

The composites described in Example 1 of varying volume percent talc were tested to determine the water vapor transmission rate, (gm-mil/100in²-day), using ASTM F1249-90 at 37.9°C and 100 percent relative humidity. A significant improvement was obtained compared to the pure PHAE resin. The results are shown in Table II.

Table II

<u>Volume %</u> <u>Talc</u>	<u>Water Vapor</u> <u>Transmission Rate (gm-mil/100in²-day).</u>
0	5.77, 6.05
5	5.04, 4.86
10	3.39, 3.58
15	2.96, 2.47
20	3.56, 3.57

Example 3

Composites were prepared as described in Example 1 using talc obtained
 5 from Specialty Minerals, Inc. of Barretts, Montana. The composites of varying volume
 percent were prepared using the talc. The oxygen transmission rates are shown in
 Table III.

Table III

<u>Volume %</u> <u>Talc</u>	<u>Oxygen Transmission Rate</u> <u>(cc-mil/100 in²-day-atm O₂)</u>
10%	0.191, 0.257
15%	0.073, 0.051
20%	0.052, 0.025
10%	0.187, 0.147
15%	0.010, 0.055

10 The talc/PHAE resin composites were also tested for oxygen transmission
 rate at high relative humidity and compared to the blank PHAE resin. The results are shown
 in Table IV.

Table IV

<u>Volume % Talc</u>	<u>% Relative Humidity</u>	<u>Oxygen Transmission Rate</u> <u>(cc-mil/100 in²-day-atm O₂)</u>
10%	86	0.460, 0.465
15%	84	0.341, 0.354
0%	91	0.938, 0.982

Example 4

PHAE resin was blended with mica (obtained from Franklin Industrial Minerals) as described in Example 1, at 10 and 20 volume percent. The oxygen transmission rates obtained at 23°C and 60 percent relative humidity are listed in Table V. Additional data were obtained at high relative humidity for the 10 and 15 volume percent mica/PHAE composites.

Table V

<u>Volume %</u> <u>Mica</u>	<u>% Relative</u> <u>Humidity</u>	<u>Oxygen Transmission</u> <u>(cc-mil/100 in²-day-atm O₂)</u>
10	60	0.139, 0.146
20	60	0.110, 0.104
10	85	0.291, 0.274
15	84	0.167, 0.147

10

Example 5

SCAN (silica coated aluminum nitride) provided by The Dow Chemical Company and calcium carbonate OMYACARB 5 provided by Omya Inc. were blended as described in Example 1 with PHAE resin at varying volume percent resulting in different PHAE composites. Table VI contains the oxygen transmission rate data for the different composites.

15

Table VI

<u>Volume % Filler</u>	<u>Oxygen Transmission</u> (cc-mil/100 in ² -day-atm O ₂)
5% SCAN	0.710, 0.691
10% SCAN	0.631, 0.662
20% SCAN	0.460, 0.538
10% CaCO ₃	0.759, 0.767
20% CaCO ₃	0.589, 0.608

The microtensile properties of the 20 volume percent composites were tested. The results are shown in Table VII.

5

Table VII

<u>Composite</u>	<u>Tensile Modulus</u>	<u>% Strain at Break</u>	<u>Tensile Break</u>	<u>Tensile Yield</u>
20% SCAN	768.5 kpsi	10.25	6.382 kpsi	8.472 kpsi
20% CaCO ₃	896.4 kpsi	4.07	7.914 kpsi	*

* not determined

Example 6

Talc (purchased from the Aldrich Chemical Company) and hydroxy-functionalized polyether, PHE, (formed by the reaction of an epihalohydrin or a diglycidyl ether with a bisphenol) were combined to yield varying volume percent talc/PHE composites. The PHE was provided by PAPHEN® Phenoxy Resins as PKHH®. The talc and PKHH resin were blended in the manner described in Example 1. After complete addition of the sample, the mixer was ramped to 120 rpm. The sample was melt-blended at approximately 170°C, 120 rpm for approximately 10 minutes. After blending, the sample was removed and pressed into films using compression molding.

The samples were tested for oxygen barrier properties according to ASTM D3985-81. Oxygen concentration was 100 percent.

The oxygen barrier property of the samples containing the talc filler were vastly improved over that of the pure PKHH resin under the same test conditions. Calcium

carbonate and SCAN composites were prepared and tested for oxygen barrier properties. The results are shown in Table VIII.

Table VIII

<u>Volume % Filler</u>	<u>Oxygen Transmission Rate (cc-mil/100 in²-day-atm O₂)</u>
0% Filler	6.094, 6.015
10% Talc	3.743, 3.755
20% Talc	2.356, 2.401
10% CaCO ₃	5.890, 5.840
20% CaCO ₃	4.337*
10% SCAN	5.770, 5.816

*Only one sample tested.

5 The microtensile properties of the 20 volume percent filler/PKHH composites were tested. The results are shown in Table IX.

Table IX

<u>Composite</u>	<u>Tensile Modulus</u>	<u>% Strain at Break</u>	<u>Tensile Break</u>	<u>Tensile Yield</u>
20% Talc	1,080.0 kpsi	3.32`	9.394 kpsi	*
20% SCAN	781.3 kpsi	12.75	7.148 kpsi	9.138 kpsi
20% CaCO ₃	731.2 kpsi	2.65	8.886 kpsi	*

* not determined

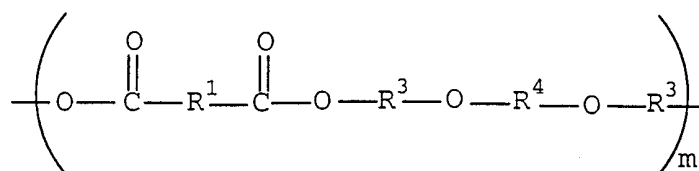
CLAIMS:

1. A polymer composite comprising a hydroxy-functionalized polyether or polyester and an inorganic filler.

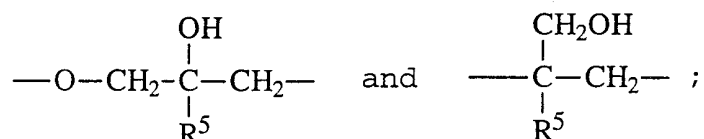
2. The polymer of Claim 1 wherein the inorganic filler is an inorganic oxide,
5 hydroxide, carbonate, nitride, carbide, boride or mixture thereof.

3. The polymer composite of Claim 1 wherein the hydroxy-functionalized polyether or polyester is formed by the reaction of a dinucleophile and a monomer containing at least one epoxy moiety.

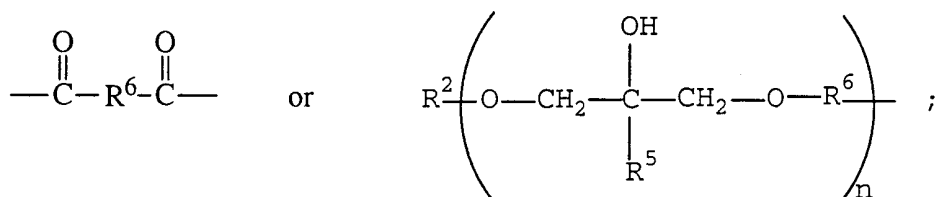
4. The polymer composite of Claim 1 wherein the hydroxy-functionalized
10 polyether or polyester has repeating units represented by the formula:



wherein m is an integer from 5 to 1000, R¹ is a divalent organic moiety which is primarily hydrocarbon; R³ is:



15 R⁴ is



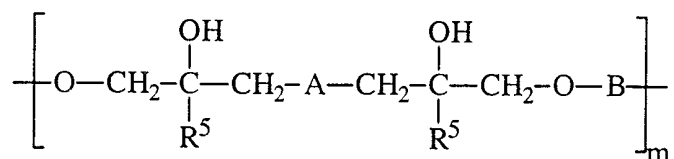
wherein R² and R⁶ are independently divalent organic moieties which are primarily hydrocarbon; R⁵ is hydrogen or alkyl, m is an integer from 5 to 1000 and n is an integer from 0 to 100.

5. The polymer composite of Claim 4 wherein the hydroxy-functionalized
20 polyether is formed by the reaction of a diglycidyl ether with a dicarboxylic acid.

6. The polymer composite of Claim 4 wherein the hydroxy-functionalized polyether is formed by the reaction of a diglycidyl ester with a bisphenol.

7. The polymer composite of Claim 4 wherein the hydroxy-functionalized polyester is formed by the reaction of a diglycidyl ester or an epihalohydrin with a dicarboxylic acid.

8. The polymer composite of Claim 1 wherein the hydroxy-functionalized polyether is represented by the formula:

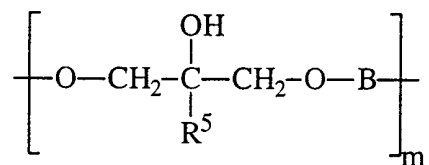


wherein A is an amine moiety or a combination of different amine moieties; B is a divalent organic moiety which is predominantly hydrocarbylene; R is alkyl or hydrogen; and m is an integer from 5 to 1000.

9. The polymer composite of Claim 8 wherein A is 2-hydroxyethylimino-, 2-hydroxypropylimino-, piperazeryl, N,N'-bis(2-hydroxyethyl)-1,2-ethylenedimino-, and B is isopropylidenediphenylene, 1,3-phenylene, or 1,4-phenylene, and R⁵ is hydrogen.

10. The polymer composite of Claim 8 wherein the hydroxy-functionalized polyether is formed by the reaction of a diglycidyl ether or an epihalohydrin with a difunctional amine.

11. The polymer composite of Claim 1 wherein the hydroxy-functionalized polyether is represented by the formula:

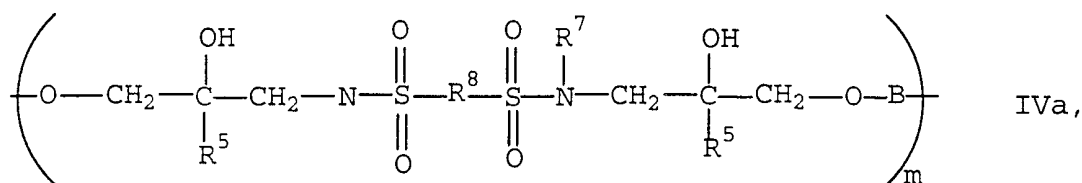


wherein B is a divalent organic moiety which is primarily hydrocarbon, and R⁵ is alkyl or hydrogen, and m is an integer from 5 to 1000.

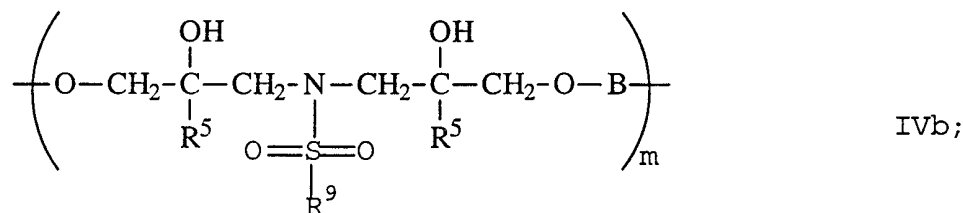
12. The polymer composite of Claim 11 wherein B is 1,3-phenylene, 1,4-phenylene, sulfonyldiphenylene, oxydiphenylene, thiodiphenylene or isopropylidenediphenylene and R⁵ is hydrogen.

13. The polymer composite of Claim 11 wherein the hydroxy-functionalized polyether is formed by the reaction of an epihalohydrin or a diglycidyl ether with a bisphenol.

14. The polymer composite of Claim 1 wherein the hydroxy-functionalized polyether is represented by the formulae:



or



wherein R⁵ is hydrogen or alkyl, R⁷ and R⁹ are independently alkyl, substituted alkyl, aryl or substituted aryl, B and R⁸ are independently a divalent organic moiety that is substantially hydrocarbon, and m is an integer from 5 to 1000.

15. The polymer composite of Claim 14 wherein R⁵ is hydrogen, R⁷ and R⁹ are independently methyl, ethyl, propyl, butyl, 2-hydroxyethyl or phenyl and B and R⁸ are independently 1,3-phenylene, 1,4-phenylene, sulfonyldiphenylene, oxydiphenylene, thiodiphenylene or isopropylidenediphenylene.

16. The polymer composite of Claim 14 wherein the hydroxy-functionalized polyether is formed by the reaction of an unsubstituted monosulfonamide or an N,N'-disubstituted disulfonamide with a diglycidyl ether.

17. The polymer composite of Claim 1 wherein the inorganic filler is talc, mica, montmorillonite, hectorite, kaolinite, dickite, nacrite, halloysite, saponite, nontronite, beidellite, volhonskoite, sauconite, magadiite, medmontite, kenyaite, vermiculite, serpentine, chlorite, palygorskite, kulkeite, aliettite, sepiolite, allophane, imogolite or a mixture thereof.

18. The polymer composite of Claim 17 wherein the inorganic filler is talc, mica, montmorillonite, hectorite or a mixture thereof.

19. The polymer composite of Claim 1 wherein the inorganic filler is a metal oxide, metal hydroxide, metal carbonate, mixed metal oxide, mixed metal hydroxide, mixed
5 metal carbonate or a mixture thereof.

20. The polymer composite of Claim 1 wherein the inorganic filler is calcium oxide, magnesium oxide, zirconium oxide, titanium oxide, manganese oxide, iron oxide, aluminum oxide, calcium hydroxide, magnesium hydroxide, zirconium hydroxide, aluminum hydroxide, manganese hydroxide, iron hydroxide, calcium carbonate, magnesium carbonate,
10 manganese carbonate, iron carbonate or zirconium carbonate.

21. The polymer composite of Claim 19 wherein the inorganic filler is calcium carbonate, calcium oxide, calcium hydroxide or a mixture thereof.

22. The polymer composite of Claim 1 wherein the inorganic filler is a metal nitride, metal carbide, or metal boride or a mixture thereof.

15 23. The polymer composite of Claim 1 wherein the inorganic filler is aluminum nitride, silicon nitride, iron nitride, silicon carbide, manganese carbide, iron carbide, iron boride, aluminum boride or manganese boride or a mixture thereof.

24. The polymer composite of Claim 21 wherein the aluminum nitride is silica-coated aluminum nitride.

20 25. The polymer composite of Claim 1 wherein the inorganic filler is at least 0.1 weight percent and not more than 90 weight percent of the final composite.

26. The polymer composite of Claim 1 formed by adding the inorganic filler to one or more of the monomers forming the hydroxy-functionalized polyether or polyester matrix and then polymerizing the monomers.

25 27. A method for forming the polymer composite of Claim 26 wherein the composite is made by reactive extrusion.

28. The composite of Claim 1 in the form of a coating, film, foam, laminate, fiber, hot-melt adhesive or molded article.

29. A method for forming a composite which comprises contacting a hydroxy-functionalized polyether or polyester, or a precursor to a hydroxy-functionalized polyether or polyester with an inorganic filler.

5 30. A method for forming a composite which comprises contacting a hydroxy-functionalized polyether or polyester, or a precursor to a hydroxy-functionalized polyether or polyester with an inorganic filler using compounding extrusion.

31. A fiber-reinforced composite comprising a resin matrix polymer reinforced with one or more reinforcing fiber or mat, wherein the resin matrix polymer is the composite of Claim 1.

INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 99/05149

A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C08K3/00 C08L67/04 C08L71/00

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)

IPC 6 C08K

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

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

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DATABASE WPI Section Ch, Week 8907 Derwent Publications Ltd., London, GB; Class A12, AN 89-050185 XP002107196 & JP 01 006061 A (TOTO KASEI KK) , 10 January 1989 see abstract ---	1,2,19, 29
X	DATABASE WPI Section Ch, Week 9427 Derwent Publications Ltd., London, GB; Class A23, AN 94-222161 XP002107197 & JP 06 157878 A (MITSUBISHI KASEI CORP) , 7 June 1994 see abstract --- -/---	1,2,22, 25,29

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents:

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

24 June 1999

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INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 99/05149

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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A	US 5 149 768 A (WHITE JERRY E ET AL) 22 September 1992 cited in the application see table 1 ----	1-31
A	EP 0 513 679 A (DOW CHEMICAL CO) 19 November 1992 see tables I,,II see claims 1-16 -----	1-31

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International Application No

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