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AUSTRALIA

Patents Act 1990

NOTICE OF ENTITLEMENT

We, GENERAL ELECTRIC COMPANY the applicant/nominated person in respect of Application No. 70754/91, state the following:-

The Nominated person is entitled to the grant of the patent because the Nominated person derives title to the invention from the inventors by assignment.

The Nominated person is entitled to claim priority from the application listed in the declaration under Article 8 of the PCT because the Nominated person is the assignee of the applicants in respect of the application listed in the declaration under Article 8 of the PCT.

The application listed under Article 8 of the PCT is the first application made in a Convention country in respect of the invention.

DATED this 19th day of March, 1993.

(A member of the firm of Davies Collison Cave for
and on behalf of the applicant)

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(19) AUSTRALIAN PATENT OFFICE **(10) Acceptance No. 641467**

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THERMOPLASTIC COPOLYETHERIMIDE ESTER ELASTOMER-ACRYLATE RUBBER COMPOSITIONS
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- (56) Prior Art Documents
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- (57) Claim

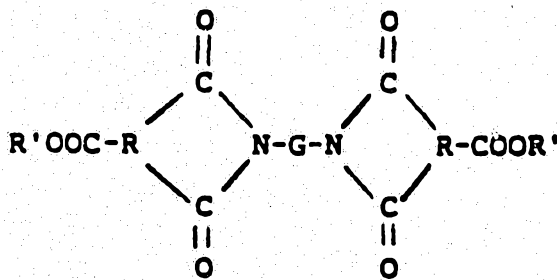
1. A thermoplastic elastomer composition comprising:

- (A) a polyetherimide ester copolymer, which comprises the reaction product of
(a) one or more low molecular weight diols; (b) one or more dicarboxylic acids;
and (c) one or more polyoxyalkylene diimide diacids,
(B) a crosslinkable rubbery alkylacrylate, and
(C) a crosslinking agent.

4. A thermoplastic elastomer composition as defined in Claim 1 wherein said polyoxyalkylene diimide diacid component (c) is derived from one or more polyoxyalkylene diamines and one or more tricarboxylic acid compounds containing two vicinal carboxyl groups or an anhydride group and an additional carboxyl group, and is characterized by the following formula:

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wherein each R is independently selected from the group consisting of C₂ to C₂₀ aliphatic and cycloaliphatic trivalent organic radicals and C₆ to C₂₀ aromatic trivalent organic radicals; each R' is independently selected from the group consisting of hydrogen, C₁ to C₆ aliphatic and cycloaliphatic monovalent organic radicals and C₆ to C₁₂ aromatic monovalent organic radicals; and G is the radical remaining after removal of the hydroxy groups of a long chain ether glycol having an average molecular weight of from about 600 to 12000.

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(21) International Application Number: PCT/US91/00430 (22) International Filing Date: 18 January 1991 (18.01.91) (30) Priority data: 467,677 19 January 1990 (19.01.90) US (71) Applicant: GENERAL ELECTRIC COMPANY [US/ US]; 1 River Road, Schenectady, NY 12345 (US). (72) Inventors: HANS, Paul, Joseph ; 1316 41st Street, Parkers- burg, WV 26104 (US). JENKINS, Kenneth, Evans ; 731 Sycamore Street, #7000, Belpre, OH 45714 (US). (74) Agents: KING, Arthur, M.; Counsel, International Patent Operation, General Electric Company, 1285 Boston Ave- nue, Bldg. 23CW, Bridgeport, CT 06602 (US) et al.		(81) Designated States: AU, CA, DE (European patent), ES (European patent), FR (European patent), GB (Euro- pean patent), IT (European patent), JP, KR, NL (Euro- pean patent). Published <i>With international search report. Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i> 641467
(54) Title: THERMOPLASTIC COPOLYETHERIMIDE ESTER ELASTOMER-ACRYLATE RUBBER COMPOSITIONS (57) Abstract Thermoplastic elastomer compositions are disclosed which comprise a thermoplastic copolyetherimide ester blended with a crosslinkable alkylacrylate rubber component. The rubber is subject to crosslinking during melt mixing resulting in a thermoplastic processable material.		

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THERMOPLASTIC COPOLYETHERIMIDE ESTER
ELASTOMER-ACRYLATE RUBBER COMPOSITIONS
CROSS REFERENCE TO RELATED APPLICATION

5 The present application is a continuation-in-part of
copending United States Patent Application Serial No.
07/467,677, filed January 19, 1990.

FIELD OF THE INVENTION

10 The present invention relates to blends of a
copolyetherimide ester elastomer and a rubbery crosslinkable
alkylacrylate elastomer.

BACKGROUND OF THE INVENTION

15 Thermoplastic elastomers of the type known as
polyetherimide esters provide a variety of unique and
excellent properties and are particularly useful in extrusion
and molding applications.

20 Polyetherimide esters prepared from diols,
dicarboxylic acids and polyoxyalkylene diimide diacids are
thermoplastic elastomers having an excellent combination of
stress-strain properties, low tensile set, high melting
temperatures and excellent strength, toughness and flexibility
properties. All of these properties are variously useful in
many elastomer applications. Copolyetherimide ester elastomers
also process well, due to their rapid crystallization rate and
excellent moldability characteristics. Elastomers with the low
25 flexural modulus of polyetherimide esters in combination with
any of the aforementioned advantageous properties have gained
wide acceptance in the field of elastomers.

30 Nonetheless, it has now been found that
polyetherimide esters can be improved or enhanced for certain
applications, especially with respect to improving the
"softness" (lower durometer) of the elastomer, while retaining
satisfactory tensile properties. The improvements are
accomplished by blending a rubbery crosslinkable alkylacrylate
elastomer with the polyetherimide ester and dynamically
35 crosslinking the former.

Work has been patented in which a rubber is mixed

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with a thermoplastic resin and subsequently crosslinked while the ingredients are being mixed. This is known in the art as "dynamic curing" or "dynamic vulcanization". Gessler et al., United States Patent No. 3,037,954 demonstrated "dynamic curing" of a butyl rubber/polypropylene blend. Fischer, United States Patent No. 3,758,643, partially cured EPDM rubber in the presence of a polyolefin. Other polyolefin/rubber vulcanizates in which the rubber is polybutadiene, natural rubber, isoprene and EPDM are disclosed in United States Patent Nos. 4,104,210; 4,130,535; and 4,311,628. In United States Patent No. 4,594,390, a continuous process for preparing EPDM/polypropylene dynamic vulcanizates is reported. The Monsanto Company commercially produces an EPDM/polypropylene dynamic vulcanizate under the trade name "SANTOPRENE", and a nitrile rubber/polyolefin dynamic vulcanizate under the trade name "GEOLAST". Further, United States Patent No. 4,801,647 discloses an EPDM/crystalline polyolefin dynamic vulcanizate.

Dynamic vulcanization has also been disclosed employing copolyesters. European patent, EP 0 293 821 A2, discloses a multiblock copolyester melt mixed with polychloroprene rubber which is then crosslinked during mixing. In United States Patent No. 4,739,012, a segmented thermoplastic copolyester is blended with a second composition which is a blend of two partially crosslinked polymers, such as PVC or PVDC and a copolymer of ethylene and one or more ethylenically unsaturated comonomers prepared by dynamic vulcanization.

Acrylic rubbers are also known to be employed in thermoplastic dynamic vulcanizates. Coran et al., Rubber Chem. and Tech., 55, 116 (1982), disclose a matrix of polymers and rubbers used in the preparation of dynamic vulcanizate compositions in which polyacrylate rubber was used; however, not with copolyetherimide esters. Wolfe, United States Patent No. 4,782,110 discloses dynamically vulcanizing ethylene-alkylacrylate copolymer rubbers with crystalline polyolefins.

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Coran et al., United States Patent No. 4,327,199, disclose employing an ethylene-acrylic type copolymer rubber containing free carboxylic moieties in blends with polyesters such as PBT. A metal oxide is used as a source of metal ions to neutralize the pendant acid groups, forming an ionomeric network structure as distinguished from covalent bond formation. In Coran et al., United States Patent Nos. 4,310,638 and 4,473,683 the ethylene acrylic copolymer is blended with a nylon resin and a metal oxide; and amorphous styrene based resins and a metal oxide, respectively.

Also to be mentioned are United States Patent Nos. 4,116,914 (ethylene vinyl acetate rubber dynamically vulcanized with polyolefins); 4,480,074 (two step process for dynamically vulcanizing an EPDM/PP vulcanizate with additional EPDM); 4,226,953 (nitrile rubber dynamically vulcanized with styrene-acrylonitrile resins); 4,207,407 (chlorinated polyethylene dynamically vulcanized with nylon resins); 4,287,324 (epichlorohydrin rubber dynamically vulcanized with polyesters such as PBT); and 4,593,062 (mixture of halogenated butyl rubber and polychloroprene dynamically vulcanized with polyolefins such as PP and PE). Special mention is made of EPO 327 010 A2, ("EP '010") which discloses blends of polyether ester copolymers with polyacrylate elastomers.

None of these however disclose a polyacrylate rubber which has been dynamically vulcanized with a thermoplastic copolyetherimide ester. It has now been discovered and is shown in the examples hereinafter that dynamically vulcanizing a polyacrylate rubber with a thermoplastic copolyetherimide ester provides an elastomer composition with improved softness while retaining excellent tensile properties. This is unexpected because the copolyether esters employed in EP '010 suffer from the loss of tensile properties to a much greater extent when mixed with crosslinkable polyacrylate rubbers and dynamically vulcanized.

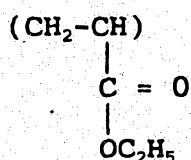
SUMMARY OF THE INVENTION

According to the present invention, there is provided a thermoplastic elastomer composition comprising:

- (A) a polyetherimide ester copolymer, which comprises the reaction product of (a) one or more low molecular weight diols; (b) one or more dicarboxylic acids; and (c) one or more polyoxyalkylene diimide diacids,
- (B) a crosslinkable rubbery alkylacrylate, and
- (C) a crosslinking agent.

Preferably the diol component (a) comprises from 60 to 100 mole percent 1,4-butanediol, the dicarboxylic acid component (b) comprises from 60 to 100 mole percent dimethyl terephthalate, and the polyoxyalkylene diimide diacid component (c) is derived from trimellitic anhydride and a polyoxyalkyl diamine selected from the group consisting of polypropylene oxide diamine and a copoly(ethylene oxide-propylene oxide) diamine having predominantly polyethylene oxide in the backbone.

The preferred rubbery alkyl acrylate is based on repeating units comprising the formula:



The preferred crosslinking agent is sodium stearate. Also contemplated are thermoplastic elastomer compositions further comprising a filler such as a silica and a plasticizer. The preferred compositions comprise component (A) in an amount ranging from 20 to 99 parts by weight and component (B) in an amount ranging from 80 to 1 part by weight based upon 100 parts by weight of (A) and (B) together.

Also according to the present invention, there is provided a process for producing a thermoplastic elastomer

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composition comprising:

(I) mixing

(i) a polyetherimide ester copolymer, and

(ii) a crosslinkable rubbery alkylacrylate; and

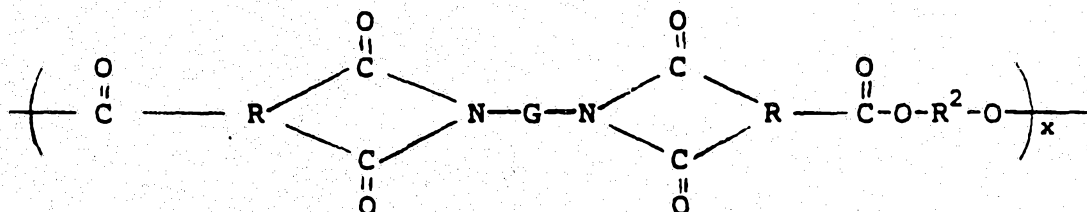
(II) curing the mixture obtained in step (I) by adding a crosslinking agent.

Preferably, step (II) of the process further comprises adding an accelerator such as sulfur, sulfur donors, magnesium oxide, tertiary amines and mixtures of any of the foregoing. Most preferred are quaternary ammonium compounds.

DETAILED DESCRIPTION OF THE INVENTION

The polyetherimide esters useful in the practice of the present invention may be prepared from one or more diols, one or more dicarboxylic acids and one or more high molecular weight polyoxyalkylene diimide diacids.

They are generally comprised of recurring polyether imide ester structural units having the general formula:



wherein G is a divalent radical remaining after removal of the amino groups of a high molecular weight polyalkylene ether diamine; R is a trivalent organic radical; R² is the divalent radical remaining after the removal of the hydroxyl groups of a diol; and x is a whole number having a value of from 2 to about 40.

Preparation of such materials is described in detail in U.S. Patent No. 4,556,705 of R.J. McCready, issued December 3, 1985 and hereby incorporated by reference.

The poly(etherimide esters) used herein may be prepared by conventional processes, such as esterification and condensation reactions for the production of polyesters, to

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provide random or block copolymers. Thus, polyetherimide esters may be generally characterized as the reaction product of the aforementioned diols and acids.

5 Preferred compositions encompassed by the present invention may be prepared from (a) one or more C_2 - C_{15} aliphatic or cycloaliphatic diols, (b) one or more C_4 - C_{16} aliphatic, cycloaliphatic or aromatic dicarboxylic acids or ester derivatives thereof and (c) one or more polyoxyalkylene diimide diacids. The amount of polyoxyalkylene diimide diacid employed is generally dependent upon the desired properties of
10 the resultant polyetherimide ester. In general, the weight ratio of polyoxyalkylene diimide diacid (c) to dicarboxylic acid (b) is from about 0.25 to 2.0, preferably from about 0.4 to about 1.4.

15 Suitable diols (a) for use in preparing the compositions of the present invention include saturated and unsaturated aliphatic and cycloaliphatic dihydroxy compounds as well as aromatic dihydroxy compounds. These diols are preferably of a low molecular weight, i.e. having a molecular weight of about 250 or less. When used herein, the term
20 "diols" and "low molecular weight diols" should be construed to include equivalent ester forming derivatives thereof, provided, however, that the molecular weight requirement pertains to the diol only and not to its derivatives. Exemplary of ester forming derivatives there may be given the
25 acetates of the diols as well as for example ethylene oxide or ethylene carbonate for ethylene glycol.

Preferred saturated and unsaturated aliphatic and cycloaliphatic diols are those having from 2 to 19 carbon
30 atoms. Exemplary of these diols there may be given ethylene glycol; propane diol; butane diol; pentane diol; 2-methyl propane diol; 2,2-dimethyl propane diol; hexane diol; decane diol; 2-octyl undecane diol; 1,2-, 1,3-, and 1,4-cyclohexane dimethanol; 1,2-, 1,3-, and 1,4-dihydroxy cyclohexane; butene
35 diol; and hexene diol. Especially preferred are 1,4-butane diol and mixtures thereof with hexane diol or butene diol, most preferably 1,4-butanediol.

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Aromatic diols suitable for use in the practice of the present invention are generally those having from 6 to about 19 carbon atoms. Included among the aromatic dihydroxy compounds are resorcinol; hydroquinone; 1,5-dihydroxy naphthalene; 4,4'-dihydroxy diphenyl bis(p-hydroxy phenyl)methane and 2,2-bis(p-hydroxy phenyl)propane.

Especially preferred diols are the saturated aliphatic diols, mixtures thereof and mixtures of a saturated diol(s) with an unsaturated diol(s), wherein each diol contains from 2 to about 8 carbon atoms. Where more than one diol is employed, it is preferred that at least about 60 mole %, based on the total diol content, be the same diol, most preferably at least 80 mole %. As mentioned above, the preferred compositions are those in which 1,4-butanediol is present in a predominant amount, most preferably when 1,4-butanediol is the only diol.

Dicarboxylic acids (b) which are suitable for use in the practice of the present invention are aliphatic, cycloaliphatic, and/or aromatic dicarboxylic acids. These acids are preferably of a low molecular weight, i.e., having a molecular weight of less than about 300; however, higher molecular weight dicarboxylic acids, especially dimer acids, which are fully described in Kirk-Othmer, Encyclopedia of Chemical Technology, 3rd Edition, vol. 7, John Wiley & Sons, N.Y., pp. 768-782, may also be used. The term "dicarboxylic acids" as used herein, includes equivalents of dicarboxylic acids having two functional carboxyl groups which perform substantially like dicarboxylic acids in reaction with glycols and diols in forming polyester polymers. These equivalents include esters and ester-forming derivatives, such as acid halides and anhydrides. The molecular weight preference, mentioned above, pertains to the acid and not to its equivalent ester or ester-forming derivative.

Aliphatic dicarboxylic acids, as the term is used herein, refers to carboxylic acids having two carboxyl groups each of which is attached to a saturated carbon atom. If the carbon atom to which the carboxyl group is attached is

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saturated and is in a ring, the acid is cycloaliphatic.

Aromatic dicarboxylic acids, as the term is used herein, are dicarboxylic acids having two carboxyl groups each of which is attached to a carbon atom in an isolated or fused benzene ring system. It is not necessary that both carboxyl groups be attached to the same aromatic ring and where more than one ring is present they can be joined by aliphatic or aromatic divalent radicals such as -O- or -SO₂-.

Representative aliphatic and cycloaliphatic acids which can be used for this invention include sebacic acid, 1,2-cyclohexane dicarboxylic acid, 1,3-cyclohexane dicarboxylic acid, 1,4-cyclohexane dicarboxylic acid, adipic acid, glutaric acid, succinic acid, oxalic acid, azelaic acid, diethylmalonic acid, allylmalonic acid, 4-cyclohexane-1,2-dicarboxylic acid, 2-ethylsuberic acid, tetramethylsuccinic acid, cyclopentanedicarboxylic acid, decahydro-1,5-naphthalene dicarboxylic acid, 4,4'-bicyclohexyl dicarboxylic acid, decahydro-2,6-naphthalene dicarboxylic acid, 4,4-methylenebis(cyclohexane carboxylic acid), 3,4-furan dicarboxylic acid, and 1,1-cyclobutane dicarboxylic acid. Preferred aliphatic acids are cyclohexane dicarboxylic acids, glutaric acid, azelaic acid and adipic acid.

Representative aromatic dicarboxylic acids which can be used include terephthalic acid, isophthalic acid, phthalic acid, bi-benzoic acid, substituted dicarboxy compounds with two benzene nuclei such as bis(p-carboxyphenyl)methane, oxybis(benzoic acid), ethylene 1,2-bis-(p-oxybenzoic acid), 1,5-naphthylene dicarboxylic acid, 2,6-naphthalene dicarboxylic acid, 2,7-naphthalene dicarboxylic acid, phenanthrene dicarboxylic acid, anthracene dicarboxylic acid, 4,4'-sulfonyl dibenzoic acid, and halo and C₁-C₁₂ alkyl, alkoxy, and aryl ring substitution derivatives thereof. Hydroxy acids such as p-(beta-hydroxyethoxy)benzoic acid can also be used provided an aromatic dicarboxylic acid is also present.

Preferred dicarboxylic acids for the preparation of the polyetherimide esters of the present invention are the

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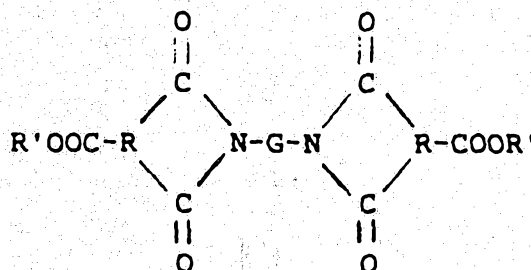
aromatic dicarboxylic acids, mixtures thereof and mixtures of one or more dicarboxylic acids with an aliphatic or cycloaliphatic dicarboxylic acid, most preferably the aromatic dicarboxylic acids. Among the aromatic acids, those with 8-16 carbon atoms are preferred, particularly the benzene dicarboxylic acids, i.e., phthalic, terephthalic and isophthalic acids and their dimethyl derivatives. Especially preferred is dimethyl terephthalate.

Finally, where mixtures of dicarboxylic acids are employed in the practice of the present invention, it is preferred that at least about 60 mole %, preferably at least about 80 mole %, based on 100 mole % of dicarboxylic acid (b) be of the same dicarboxylic acid or ester derivative thereof. As mentioned above, the preferred compositions are those in which dimethyl terephthalate is the predominant dicarboxylic acid, most preferably when dimethyl terephthalate is the only dicarboxylic acid.

Polyoxyalkylene diimide diacids (c) suitable for use herein are high molecular weight diimide diacids wherein the average molecular weight is greater than about 700, most preferably greater than about 900. They may be prepared by the imidization reaction of one or more tricarboxylic acid compounds containing two vicinal carboxyl groups or an anhydride group and an additional carboxyl group which must be esterifiable and preferably is nonimidizable with a high molecular weight polyoxyalkylene diamine. The polyoxyalkylene diimide diacids and processes for their preparation are more fully disclosed in McCready, European Patent No. 180,149, published May 7, 1986, and entitled "High Molecular Weight Diimide-Diacid Compounds Useful in the Preparation of Polyether Imide Ester(s) and Amide(s)", incorporated herein by reference.

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In general, the polyoxyalkylene diimide diacids useful herein may be characterized by the following formula:



wherein each R is independently a trivalent organic radical, preferably a C₂ to C₂₀ aliphatic, aromatic or cycloaliphatic trivalent organic radical; each R' is independently hydrogen or a monovalent organic radical preferably selected from the group consisting of C₁ to C₆ aliphatic and cycloaliphatic radicals and C₆ to C₁₂ aromatic radicals, e.g. phenyl, most preferably hydrogen; and G is the radical remaining after the removal of the terminal (or as nearly terminal as possible) hydroxy groups of a long chain ether glycol having an average molecular weight of from about 600 to about 12000, preferably from about 900 to about 4000, and a carbon-to-oxygen ratio of about 1.8 to about 4.3.

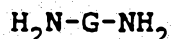
Representative long chain ether glycols from which the polyoxyalkylene diamine is prepared include poly(ethylene ether)glycol; poly(propylene ether) glycol; poly(tetramethylene ether)glycol; random or block copolymers of ethylene oxide and propylene oxide, including propylene oxide terminated poly(ethylene ether)glycol; and random or block copolymers of tetrahydrofuran with minor amounts of a second monomer such as methyl tetrahydrofuran (used in proportion such that the carbon-to-oxygen mole ratio in the glycol does not exceed about 4.3). Especially preferred poly(alkylene ether)glycols are poly(propylene ether)glycol and poly(ethylene ether)glycols end capped with poly(propylene ether)glycol or propylene oxide.

In general, the polyoxyalkyl(C₁-C₆)ene diamines

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useful within the scope of the present invention will have an average molecular weight of from about 600 to 12000, preferably from about 900 to 4000. These may be characterized by the following general formula:

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wherein G is the radical remaining after the removal of the amino groups of a long chain alkylene ether diamine. These polyether diprimary diamines are available commercially from Texaco Chemical Company under the trademark "JEFFAMINE". In general they are prepared by known processes for the amination of glycols. For example, they may be prepared by aminating the glycol in the presence of ammonia, Raney nickel catalyst, and hydrogen as set forth in Belgium Patent No. 634,741. Alternatively, they may be prepared by treating the glycol with ammonia and hydrogen over a nickel-copper-chromium catalyst as taught by United States Patent No. 3,654,370. Other methods for the production thereof include those taught in United States Patent Nos. 3,155,728 and 3,236,895 and French Patent Nos. 1,551,605 and 1,446,708. All of the foregoing patents are incorporated herein by reference.

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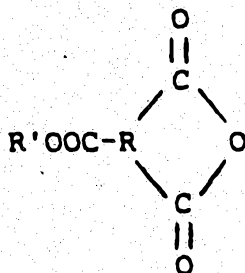
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The tricarboxylic component may be almost any carboxylic acid anhydride containing an additional carboxylic group or the corresponding acid thereof containing two imide-forming vicinal carboxyl groups in lieu of the anhydride group. Mixtures thereof are also suitable. The additional carboxylic group must be esterifiable and preferably is substantially nonimidizable.

The tricarboxylic acid materials can be characterized by the following formula:

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where R is a trivalent organic radical, preferably a C₂ to C₂₀ aliphatic, aromatic, or cycloaliphatic trivalent organic

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radical and R' is preferably hydrogen or a monovalent organic radical preferably selected from the group consisting of C₁ to C₆ aliphatic or cycloaliphatic radicals and C₆ to C₁₂ aromatic radicals, e.g. phenyl; most preferably hydrogen. A preferred tricarboxylic component is trimellitic anhydride.

Briefly, these polyoxyalkylene diimide diacids may be prepared by known imidization reactions including melt synthesis or by synthesizing in a solvent system. Such reactions will generally occur at temperatures of from 100 degrees C to 300 degrees C, preferably at from about 150 degrees C to about 250 degrees C while drawing off water or in a solvent system at the reflux temperature of the solvent or azeotropic (solvent) mixture.

Although the weight ratio of the above ingredients is not critical, it is preferred that the diol be present in at least a molar equivalent amount, preferably a molar excess, most preferably at least 150 mole % based on the moles of dicarboxylic acid (b) and polyoxyalkylene diimide diacid (c) combined. Such molar excess of diol will allow for optimal yields, based on the amount of acids, while accounting for the loss of diol during esterification/condensation.

Further, while the weight ratio of dicarboxylic acid (b) to polyoxyalkylene diimide diacid (c) is not critical to form the polyetherimide esters used in the present invention, preferred compositions are those in which the weight ratio of the polyoxyalkylene diimide diacid (c) to dicarboxylic acid (b) is from about 0.25 to about 2, preferably from about 0.4 to about 1.4. The actual weight ratio employed will be dependent upon the specific polyoxyalkylene diimide diacid used and more importantly, the desired physical and chemical properties of the resultant polyetherimide ester. In general, the lower the ratio of polyoxyalkylene diimide diester to dicarboxylic acid the better strength, crystallization and heat distortion properties of the polymer. Alternatively, the higher the ratio, the better the flexibility, tensile set and low temperature impact characteristics.

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In preferred embodiments, the polyetherimide ester product will comprise the reaction product of dimethyl terephthalate, most preferably, with up to 40 mole % of another dicarboxylic acid; 1,4-butanediol, optionally with up to 40 mole % of another saturated or unsaturated aliphatic or cycloaliphatic diol; and a polyoxyalkylene diimide diacid prepared from a polyoxyalkylene diimine of molecular weight of from about 600 to about 12000, preferably from about 900 to 4000, and trimellitic anhydride. In its most preferred embodiments, the diol will be 100 mole % 1,4-butanediol and the dicarboxylic acid 100 mole % dimethyl terephthalate.

The polyetherimide esters described herein may be prepared by conventional esterification/condensation reactions for the production of polyesters. Exemplary of the processes that may be practiced are as set forth in, for example, U.S. Pat. Nos. 3,023,192, 3,763,109, 3,651,014, 3,663,653 and 3,801,547, herein incorporated by reference. Additionally, these compositions may be prepared by such processes and other known processes to effect random copolymers, block copolymers or hybrids thereof wherein both random and block units are present.

It is customary and preferred to utilize a catalyst in the process for the production of the polyetherimide esters of the present invention. In general, any of the known ester-interchange and polycondensation catalysts may be used. Although two separate catalysts or catalyst systems may be used, one for ester interchange and one for polycondensation, it is preferred, where appropriate, to use one catalyst or catalyst system for both. In those instances where two separate catalysts are used, it is preferred and advantageous to render the ester-interchange catalyst ineffective following the completion of the precondensation reaction by means of known catalyst inhibitors or quenchers, in particular, phosphorus compounds such as phosphoric acid, phosphenic acid, phosphonic acid and the alkyl or aryl esters of salts thereof, in order to increase the thermal stability of the resultant polymer.

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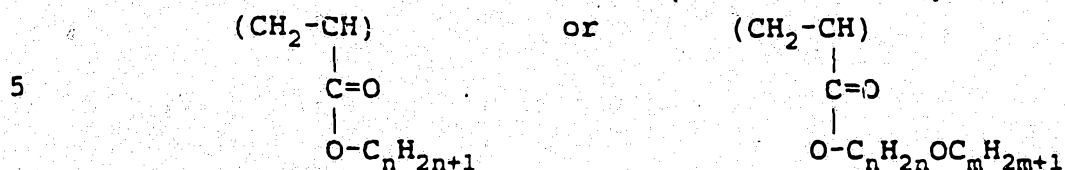
Exemplary of the suitable known catalysts there may be given the acetates, carboxylates, hydroxides, oxides, alcoholates or organic complex compounds of zinc, manganese, antimony, cobalt, lead, calcium and the alkali metals insofar
5 as these compounds are soluble in the reaction mixture. Specific examples include, zinc acetate, calcium acetate and combinations thereof with antimony tri-oxide and the like. These catalysts as well as additional useful catalysts are described in U.S. Pat. Nos. 2,465,319; 2,534,023; 2,850,483;
10 2,892,871; 2,937,160; 2,998,412; 3,047,539; 3,110,693 and 3,385,830, among others, incorporated herein by reference. Where the reactants and reactions allow, it is preferred to use the titanium catalysts including the inorganic and organic titanium containing catalysts, such as those described in, for
15 example, U.S. Pat. Nos. 2,720,502; 2,727,881; 2,729,619; 2,822,348; 2,906,737; 3,047,515; 3,056,817, 3,056,818; and 3,075,952 among others, incorporated herein by reference. Especially preferred are the organic titanates such as tetra-butyl titanate, tetra-isopropyl titanate and tetra-octyl
20 titanate and the complex titanates derived from alkali or alkaline earth metal alkoxides and titanate esters, most preferably the organic titanates. These too may be used alone or in combination with other catalysts such as for example, zinc acetate, manganese acetate or antimony trioxide, and/or
25 with a catalyst quencher as described above.

Although these polyetherimide esters possess many desirable properties, it is often preferred to stabilize the compositions to heat, oxidation, radiation by UV light and the like, as described in the aforementioned U.S. Patent No.
30 4,556,705.

The polyacrylate elastomers or rubbery alkylacrylates are generally copolymers having two major components: the backbone, comprising from about 95 to about 99 weight percent of the polymer; and the reactive cure site,
35 comprising from about 1 to about 5 weight percent of the polymer. Preferably the copolymers have high molecular weights, typically around 100,000 Mw (viscosity average

-15-

molecular weight). The backbones are made from monomeric acid esters to form repeating units of primarily two types:



where n is 2 or 4 and m is 1 or 2. The most common cure site monomers are described in the below referenced Starmer et al. article. Especially preferred are 2-chloroethyl vinyl ether and allyl glycidyl ether. Physically, polyacrylate elastomers are inherently soft and tacky. They commonly have relatively low Mooney viscosities (ML-1+4 @ 100° C) in the 25 to 60 range. These elastomers are more fully described in P.H. Starmer and F.R. Wolf, Encyclopedia of Polymer Science and Engineering, 2d Ed., 306-325 (1985), incorporated herein by reference.

The mixing of the polyetherimide esters and polyacrylate elastomers may be carried out in any device known to those skilled in the art. Preferably the components are melt mixed in compounding device such as an internal mixer (Brabender or Banbury type) and extruders (twin screw or kneading). The polyetherimide esters and polyacrylate elastomers are typically combinable in proportions ranging from about 20 to about 99 parts by weight polyetherimide ester and from about 80 to about 1 part by weight polyacrylate elastomer based upon 100 parts by weight of the two resins combined. Preferably, the polyetherimide ester is present in an amount ranging from 20 to about 80 parts by weight, most preferably from about 40 to about 60 parts by weight; and correspondingly the polyacrylate elastomer is present in an amount ranging from about 80 to about 20 parts by weight, most preferably from about 60 to about 40 parts by weight. In another preferred embodiment the compositions of the present invention comprise about 50 parts by weight polyetherimide ester and about 50 parts by weight polyacrylate elastomer.

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The mixing compositions may also contain, in addition to resin and rubber, various additives known to those skilled in the art for use in compounding of thermoplastics, rubbers, and their blends, to modify the properties thereof, such as, but not limited to fillers, stabilizers, antidegradents, processing aids, plasticizers, pigments and the like. Typical fillers would include carbon blacks, silicas, clays, minerals or mixtures thereof. Both low and high molecular weight plasticizers are contemplated.

In a typical composition, the thermoplastic copolyetherimide ester resin, rubber and additives are mixed in the appropriate device at a temperature high enough to soften and/or melt the materials such that an intimate mixture is obtained. Once an intimate mixture is obtained, the rubber material is cured by the addition of crosslinking agents, and optionally accelerators and heating, e.g., at from about 200° C to about 250° C, for from about 30 min. to about 30 sec., preferably from 5 min. to about 30 sec.

Crosslinking agents are any agents which promote vulcanization of the acrylic elastomer. The cure system employed varies with type of cure-site monomer present in the acrylic elastomer. Preferred crosslinking agents are soaps including metallic carboxylates such as sodium or potassium stearate. Optionally the cure system may also comprise an accelerator as well as a crosslinking agent. Preferred accelerators include sulfur; sulfur donors such as tetramethylthiouram; or bases such as magnesium oxide or tertiary amines. Ammonium benzoate, ammonium adipate, and soap/quaternary amine systems are also known to be effective cure systems, as are red lead/ethylene thiourea and diamines and polyamines. Most preferred is a soap/quaternary ammonium system. These and others are more extensively discussed in the above-referenced Starmer et al. article.

Mixing time is determined by the temperature and/or amounts of crosslinking agents added. The materials thus produced are processable by common thermoplastic processing techniques, such as injection and compression

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molding techniques and yield flexible, elastomeric parts.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

The following examples illustrate the present invention. They are not to be construed to limit the claims in any manner whatsoever.

EXAMPLES 1 AND 1A*

To a Banbury mixer are added a crosslinkable polyacrylate elastomer comprising alkyl halide vinyl ether cure sites ("HYTEMP" 4451CG, Zeon Chemical Co.), a polyetherimide ester resin ("LO-MOD" J-1013, GE Plastics, Pittsfield, MA, McCready, U.S. Patent No. 4,456,705), a hindered phenolic antioxidant stabilizer ("IRGANOX 1010", CIBA-Geigy), a plasticizer ("PARAPLEX" G-62, C. P. Hall Co., Chicago, IL, U.S.A.), and a phosphite/thioester secondary stabilizer ("MARK" 5117, Argus Div., Witco Co., N.Y., U.S.A.). The mixture is mixed to form an intimate blend. Once an intimate blend is obtained, a crosslinker (sodium stearate) and an accelerator (quaternary ammonium complex, "NPC-50", Zeon Chemical Co., Japan) are added. The mixture is then dynamically cured by mixing for 3 to 4 minutes at a temperature of 200° to 220° C. The composition is injection molded into test specimens and tested for tensile strength properties. For comparative purposes, a back-to-back sample is prepared, except that a copolyether ester resin similar to those described in EPO 0 327 010 A2 ("HYTREL" G-4078) is employed as the thermoplastic elastomer instead of the polyetherimide ester resin. The formulations used and the physical properties obtained are set forth in Table 1.

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TABLE 1

	<u>Example</u>	<u>1A*</u>	<u>1</u>
	<u>Ingredients(parts/wt)</u>		
5	Polyetherimide ester resin ^a	--	763.6
	Polyether ester resin ^b	763.6	--
	Polyacrylate elastomer ^c	763.6	763.6
	Sodium stearate crosslinker	30.5	30.5
	Quaternary ammonium accelerator ^d	15.3	15.3
10	Hindered phenol antioxidant ^e	3.8	3.8
	Plasticizer ^f	7.6	7.6
	Secondary antioxidant ^g	3.8	3.8
	Processing aid ^h	7.6	7.6
	<u>Properties</u>		
15	Tensile break strength, psi	889	1063
	Elongation break, %	182	156

a--"LOMOD" J1013, GE Plastics, Pittsfield, MA 01201, U.S.A.

b--"HYTREL" G-4078, DuPont Company, Wilmington, DE, U.S.A.

20 c--"HYTEMP" 4451CG, Zeon Chemical Co., Japan

d--"NPC-50", Zeon Chemical Co., Japan

e--"IRGANOX"1010, CIBA-Geigy Co., Ardsley, NY, U.S.A.

f--"PARAPLEX" G-62, C.P. HALL Co., Chicago, IL, U.S.A.

g--"MARK" 5117, Witco Chemical Co., N.Y. U.S.A.

25 h--"STRUKTOL" WS-280, Struktol Co., U.S.A.

30 The foregoing results demonstrate that a composition prepared by dynamically vulcanizing a crosslinkable polyacrylate rubber with a thermoplastic copolyetherimide ester resin unexpectedly provides better retention of tensile properties than a composition prepared by dynamically vulcanizing a polyacrylate rubber with a thermoplastic copolyether ester resin, as taught in the prior art EPO 0 327 010.

35 This is especially surprising in light of the better tensile properties of the copolyether ester alone when compared with the copolyetherimide ester alone, tensile

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breaking strengths being typically 2359 psi and 2159 psi, respectively, and elongations at break of 532 % and 215 %, respectively.

Thus, by employing a copolyetherimide ester as the thermoplastic resin dynamically vulcanized with a crosslinkable rubbery alkylacrylate there is observed a 49.2 percent retention in tensile break strength and a 72.6 percent retention in elongation at break, as compared to a 37.7 percent retention in tensile break strength and 34.2 percent retention in elongation at break with the copolyether ester of the prior art.

EXAMPLES 2-4

To a Brabender mixer are added 42.6 g of a polyacrylate elastomer (100 phr), 28.4 g of a polyetherimide ester (66.7 phr) and 0.43 g (1 phr) of an antioxidant ("IRGANOX" 1010). The mixture is mixed and heated at 220°C to form an intimate blend. Once an intimate blend is obtained, 1.70 g (4 phr) of a crosslinker (sodium stearate) and 0.85 g (2 phr) of an accelerator ("NPC-50") are added. The mixture is dynamically cured by mixing for 2 to 3 minutes at 220°C and the composition is compression molded into test specimens and tested for tensile strength properties according to ASTM D-412. For comparative purposes, tests are run without curing. The results along with compositional data are set forth below in Table 2.

TABLE 2
EFFECT OF DYNAMIC CURING

<u>Example</u>	<u>4A*</u>	<u>2</u>	<u>2A*</u>	<u>3</u>	<u>3A*</u>	<u>4</u>
Composition g (phr)						
Hytemp* 4051 EP ^a	44.1 (100)	42.6 (100)	44.1 (100)	42.6 (100)	44.1 (100)	42.6 (100)
PEIE ^b	29.4 (66.7)	-	-	-	-	28.4 (66.7)
PEIE ^c	-	28.4 (66.7)	29.4 (66.7)	-	-	-
PEIE ₁ ^d	-	-	-	28.4 (66.7)	29.4 (66.7)	-
PEIE ₂ ^e	-	0.85 (2)	-	0.85 (2)	-	0.85 (2)
NPC-50 ^f	-	1.70 (4)	-	1.70 (4)	-	1.70 (4)
NaSt	-	1.70 (4)	-	1.70 (4)	-	1.70 (4)
Irganox 1010	0.44 (1)	0.43 (1)	0.44 (1)	0.43 (1)	0.44 (1)	0.43 (1)
<u>Properties</u>						
Tensile Break, psi	475	933	402	828	512	1055
Elongation Break, %	278	276.5	515	258	567	256

* = Comparative Example

^a = Polyacrylate elastomer having alkylhalide vinyl ether cure sites (Zeon Chemicals Co.)

^b = Polyetherimide ester ("LO-MOD" J1013, General Electric Co., prepared in accordance with Example 1 of McCready, U.S. Pat. No. 4,556,705)

^c = Polyetherimide ester ("LO-MOD" JB1090, General Electric Co.)

^d = Polyetherimide ester (General Electric Co., see footnote c)

^e = Quaternary Ammonium Accelerator (Zeon Chemicals Co.)

^f = Sodium Stearate

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5 It can be seen from Table 2 above that dynamically curing the polyetherimide ester vastly improves the tensile break strength with a minimal decrease in elongation break. (Example 3 vs. 3A*). In the copolyetherimide ester examples, curing improved the tensile break strength.

EXAMPLES 5-8

10 The procedure of Examples 2-4 is repeated except employing a Banbury mixer and various other additives and the concentration of a filler is varied. The compositions are then injection molded into ASTM D-412 Type 1 bars. The results along with compositional data are set forth in Table 3 below.

TABLE 3
EFFECT OF FILLER

<u>Example</u>	<u>5</u>	<u>6</u>	<u>7</u>	<u>8</u>
<u>Composition (g)</u> ^a				
Hytemp 4051 EP ^b	900.00	875	850.00	810.00
PEIE	600.00	583.63	566.95	540.27
Struktol WS280 (processing aid)	9.00	8.75	8.50	8.10
Plastolein 9720 (plasticizer)	22.50	21.88	21.25	20.25
Hi-Sil 243LD (filler)	0.00	43.75	85.0	162.00
Mark 5117 (secondary antioxidant)	4.50	4.38	4.25	4.05
Irganox 1010 (primary antioxidant)	4.50	4.38	4.25	4.05
NPC-50 (accelerator)	18.00	17.50	17.00	16.20
Sodium Stearate (crosslinker)	36.00	35.00	34.00	32.40
<u>Properties</u>				
Tensile Break, psi	723	863	826	1001
Elongation Break, %	89	107	94	105
Young's Modulus, psi	2870	2669	2750	3113
100% modulus, psi	-	849	-	990
Strain at Yld., %	32.4	39.8	37.2	45.5
Tension Set	A/A	A/B	A/A	B/B
Shore A hardness, pts	72	72	74	76
Specific Gravity ^c (ASTM D792-A)	1.14	1.15	1.17	1.19
Compression Set ^c % at 23°C	16	16	16	16
Compression Set ^c % at 100°C	46	48	46	49

^a = Polyacrylate elastomer (Zeon Chemicals Co., footnote a Table 2)

^b = Polyetherimide ester (General Electric Co., footnote b Table 2)

A = failed during initial straining at 100%

B = failed during hold at 100% strain

^c = according to ASTM D395-B for 22 hours, average values of four runs

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EXAMPLES 9-13 .

The procedure of Examples 5-8 is repeated except various other additives are employed and two different plasticizers are employed at two concentration levels (2.5 and 5.0 phr). The results are set forth in Table 4 below.

5

TABLE 4
EFFECT OF PLASTICIZER

<u>Example</u>	<u>9</u>	<u>10</u>	<u>11</u>	<u>12</u>	<u>13</u>
<u>Composition (g)^a</u>					
Hytemp ^b 4051 EP	865.00	850.00	840.00	850.00	840.00
PEIE	576.96	566.95	560.28	566.95	560.28
Struktol WS280	8.65	8.50	8.40	8.50	8.40
Hi-Sil 243LD	86.50	85.00	84.00	85.00	84.00
Mark 5117	4.33	4.25	4.20	4.25	4.20
Irganox 1010	4.33	4.25	4.20	4.25	4.20
NPC-50	17.30	17.00	16.80	17.00	16.80
Sodium Stearate	34.60	34.00	33.60	34.00	33.60
Paraplex G-62	-	21.25	42.00	-	-
Plastolein 9720	-	-	-	21.25	42.00
<u>Properties</u>					
Tensile Break, psi	938	861	862	857	781
Elongation Break, %	96	99	94	97	107
Young's Modulus, psi	3199	2763	2730	2810	2722
100% modulus, psi	-	-	-	-	730
Strain at Yld., %	36.8	40.5	41.1	42.6	36.2
Tension Set	A/A	A/B	B/B	A/A	B/B
Shore A Hardness, pts	73	73	73	72	70
Specific Gravity ^c (ASTM D797-A)	1.17	1.17	1.16	1.17	1.16
Compression Set ^c , % at 23°C	14	14	15	13	14
Compression Set ^c , % at 100°C	46	45	45	45	45

TABLE 4 (continued)

^a = Polyacrylate elastomer (Zeon Chemicals Co., footnote a Table 2)

5 ^b = Polyetherimide ester (General Electric Co., footnote b Table 2)

A = Failed during initial straining at 100%

B = Failed during hold at 100% strain

c = according to ASTM D395-B for 22 hours, average values of four runs.

10

EXAMPLES 14-18

The procedure of Examples 5-8 is repeated except various other additives are employed and the rubber/resin weight ratio is varied at 20/80, 40/60, 50/50, 60/40 and 80/20. The results along with compositional data are set forth
15 below in Table 5.

TABLE 5
RUBBER/RESIN VARIATIONS

<u>Example</u>	<u>14</u>	<u>15</u>	<u>16</u>	<u>17</u>	<u>18</u>
<u>Composition (g)</u> ^a					
Hytemp 4051 EP ^b	314.00	615.00	760.00	900.00	1180.00
PEIE	1256.00	922.50	760.50	600.30	295.00
Struktol w5280	3.14	6.15	7.60	9.00	11.80
Paraplex G-62	6.28	12.30	15.20	18.00	23.60
Mark 5117	1.57	3.08	3.80	4.50	5.90
Irganox 1010	1.57	3.08	3.80	4.50	5.90
NPC-50	6.28	12.30	15.20	18.00	23.60
Sodium Stearate	12.56	24.60	30.40	36.00	47.20
 <u>Properties</u>					
<u>Type c bars</u> ^c					
Tensile Break, psi	2285	1935	1664	1148	317
Elongation Break, %	442	353	319	257	138
Young's Modulus, psi	9909	4932	3550	2286	385
100% Modulus, psi	1295	905	705	524	231
200% Modulus, psi	1540	1231	1053	856	-
Percent Strain at Yld., %	15	21	29	35	-
Tension Set, %	26/26	24/23	18/18	15/15	-

TABLE 5 (continued)

Example	14	15	16	17	18*
Type 1 bars					
Tensile Break, psi	1856	1304	1022	834	-
Elongation Break, %	160	120	100	92	-
Young's Modulus, psi	11,166	7066	4051	2673	-
100% Modulus, psi	1615	1251	-	-	-
200% Modulus, psi	-	-	-	-	-
Percent Strain at Yld., %	16	18	23	37	-
Hardness, Shore-A	89	84	78	72	-

^a = Polyacrylate elastomer (Zeon Chemical Co., footnote a Table 2)

^b = Polyetherimide ester (General Electric Co., footnote b Table 2)

* = Scorched in mixer, crumb was compression molded into plaque for testing

^c = Die cut from 4 in. x 6 in. compression molded plaques

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From the above Tables 1-5 it can be seen that the physical properties of a typical composition containing 60 parts by weight rubber and 40 parts by weight crystalline thermoplastic polyetherimide ester and a suitable additives package are: Tension Set (ASTM D412) typically 15 to 25 percent; Compression Set (ASTM D395 Method B-plied sample) approximately 16 percent at 23°C/22 hours and 45 percent at 100°C/22 hours. Hardness values (Shore A durometer) of 55 to 80+ points can be achieved by suitable choice of components and additives.

The compositions of the present invention are placed under Dynamic Mechanical Thermal Analysis (DMTA) to produce DMTA curves. Typical thermoplastic materials, such as the thermoplastic elastomers used herein, exhibit storage modulus versus temperature DMTA curves which can be described as possessing a glassy plateau which is generally constant in magnitude, followed by a glass transition region which is characterized by a two to three order of magnitude drop in the storage modulus to the so-called rubbery plateau. The rubbery plateau-storage modulus value is then usually observed to decrease with increasing temperature in thermoplastics (i.e. viscous flow). In semi-crystalline thermoplastics the rubbery plateau is then followed by a large drop off at the crystalline melting point of the polymer.

In the compositions of the present invention, the drop off of modulus associated with the melting of the polyetherimide ester is observed to be followed by what may be termed a second rubbery plateau which was found to be of essentially constant magnitude to the extent of the temperature tested (250° C). The presence of this second rubbery plateau was found to be dependent on the ratio of rubber to thermoplastic with compositions having below 50 weight percent rubber not exhibiting the second rubbery plateau. In a thermoset rubber material, the storage modulus would be essentially constant in the rubbery plateau region and would not drop off with increasing temperature (until

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degradation occurs), due to crosslinking of the system.

To test the thermoplasticity of the compositions a typical material which exhibits the second rubbery plateau was prepared, molded and then heated for 75 minutes at 200° C in an air circulating oven. The material is then charged into a Brabender mixer and mixed to a molten state in which the consistency was observed to be constant as a function of time over a ten minute test period.

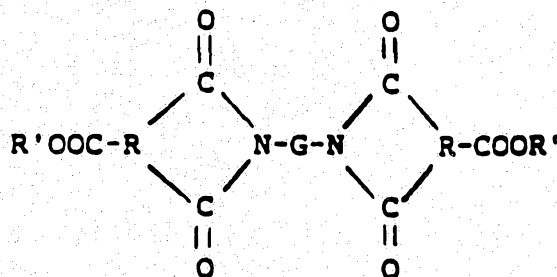
Thus, the materials are thermoplastics. If the materials were becoming thermoset above the melting point of the crystalline thermoplastic polyetherimide ester, they would have shear degraded when reprocessed as does a true thermoset material.

The above patents, patent applications, publications and test methods are hereby incorporated by reference.

Many variations of the present invention will suggest themselves to those skilled in the art in light of the above-detailed description. For example, any copolyetherimide ester resin and crosslinkable alkylacrylate elastomer may be employed. Other suitable crosslinkers and accelerators are also contemplated. Additives such as flame retardants, light stabilizers and the like may be employed in the compositions of the present invention. All such obvious modifications are within the full intended scope of the appended claims.

THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS:

1. A thermoplastic elastomer composition comprising:
 - (A) a polyetherimide ester copolymer, which comprises the reaction product of (a) one or more low molecular weight diols; (b) one or more dicarboxylic acids; and (c) one or more polyoxyalkylene diimide diacids,
 - (B) a crosslinkable rubbery alkylacrylate, and
 - (C) a crosslinking agent.
2. A thermoplastic elastomer composition as defined in Claim 1 wherein said diol component (a) comprises from 60 to 100 mole percent 1,4-butanediol.
3. A thermoplastic elastomer composition as defined in Claim 1 wherein said dicarboxylic acid component (b) comprises from 60 to 100 mole percent dimethyl terephthalate.
4. A thermoplastic elastomer composition as defined in Claim 1 wherein said polyoxyalkylene diimide diacid component (c) is derived from one or more polyoxyalkylene diamines and one or more tricarboxylic acid compounds containing two vicinal carboxyl groups or an anhydride group and an additional carboxyl group, and is characterized by the following formula:

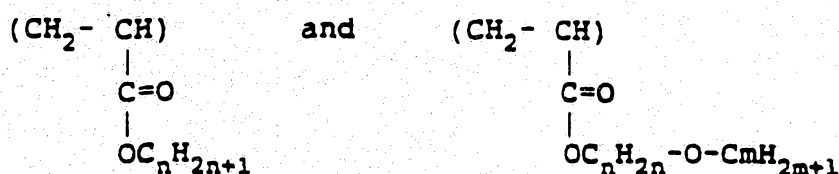


wherein each R is independently selected from the group consisting of C₂ to C₂₀ aliphatic and cycloaliphatic trivalent organic radicals and C₆ to C₂₀ aromatic

trivalent organic radicals; each R' is independently selected from the group consisting of hydrogen, C₁ to C₆ aliphatic and cycloaliphatic monovalent organic radicals and C₆ to C₁₂ aromatic monovalent organic radicals; and G is the radical remaining after removal of the hydroxy groups of a long chain ether glycol having an average molecular weight of from about 600 to 12000.

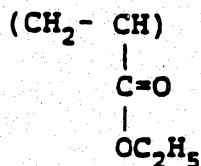
5. A thermoplastic elastomer composition as defined in Claim 4 wherein said polyoxyalkylene diimide diacid is derived from trimellitic anhydride and a polyoxyalkyl diamine selected from the group consisting of polypropylene oxide diamine and a copoly(ethylene oxide-propylene oxide) diamine having predominantly polyethylene oxide in the backbone.

6. A thermoplastic elastomer composition as defined in Claim 1 wherein said rubbery alkylacrylate consists essentially of repeating units of the formulae:



where n is 2 or 4 and m is 1 or 2 and a cure site monomer.

7. A thermoplastic elastomer composition as defined in Claim 6 wherein said rubbery alkylacrylate is based on repeating units of the formula:



8. A thermoplastic elastomer composition as defined in Claim 1 wherein said crosslinking agent is selected from the group sodium stearate, potassium stearate and a mixture thereof.

9. A thermoplastic elastomer composition as defined in Claim 1 wherein

said polyetherimide ester component (A) comprises from 20 to 99 parts by weight and said rubbery alkylacrylate component (B) comprises from 80 to 1 part by weight based upon 100 parts by weight of (A) and (B).

10. A thermoplastic elastomer composition as defined in Claim 9 wherein said component (A) comprises from 20 to 80 parts by weight and said component (B) comprises from 80 to 20 parts by weight.

11. A thermoplastic elastomer composition as defined in Claim 10 wherein said component (A) comprises from 40 to 60 parts by weight and said component (B) comprises from 60 to 40 parts by weight.

12. A thermoplastic elastomer composition as defined in Claim 9 wherein said component (A) comprises 50 parts by weight and said component (B) comprises 50 parts by weight.

13. A thermoplastic elastomer composition as defined in Claim 1 which further comprises a filler, a stabilizer, an antidegradent, a processing aid, a plasticizer, a pigment, or mixtures of any of the foregoing.

14. A thermoplastic elastomer composition as defined in Claim 13 wherein said filler is selected from the group consisting of carbon blacks, silicas, clays and minerals.

15. A thermoplastic elastomer composition as defined in Claim 13 wherein said plasticizer comprises a low molecular weight plasticizer or a high molecular weight plasticizer or both.

16. A thermoplastic elastomer composition as defined in Claim 1 which further comprises an accelerator.

17. A thermoplastic elastomer composition as defined in Claim 16 wherein

said accelerator is selected from the group consisting of sulfur, sulfur donors, magnesium oxide, tertiary amines, quaternary ammonium compounds and mixtures of any of the foregoing.

18. A thermoplastic elastomer composition as defined in Claim 17 wherein the crosslinker comprises sodium stearate and the accelerator comprises a quaternary ammonium compound.

19. A process for producing a thermoplastic elastomer composition comprising:

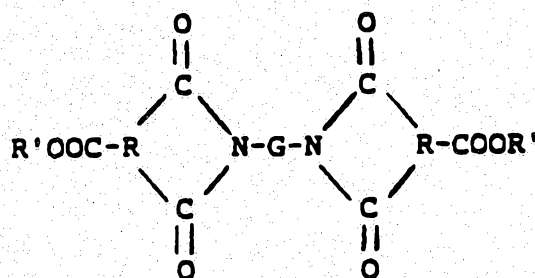
- (I) mixing
 - (i) a polyetherimide ester copolymer, and
 - (ii) a crosslinkable rubbery alkylacrylate; and
- (II) curing the mixture obtained in step (I) by adding a crosslinking agent.

20. A process as defined in Claim 19 wherein said polyetherimide ester comprises the reaction product of (a) one or more low molecular weight diols; (b) one or more dicarboxylic acids; and (c) one or more polyoxyalkylene diimide diacids.

21. A process as defined in Claim 20 wherein said diol component (a) comprises from 60 to 100 mole percent 1,4-butanediol.

22. A process as defined in Claim 20 wherein said dicarboxylic acid component (b) comprises from 60 to 100 mole percent dimethyl terephthalate.

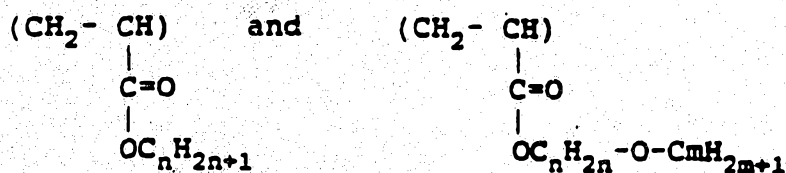
23. A process as defined in Claim 20 wherein said polyoxyalkylene diimide diacid component (c) is derived from one or more polyoxyalkylene diamines and one or more tricarboxylic acid compounds containing two vicinal carboxyl groups or an anhydride group and an additional carboxyl group, and is characterized by the following formula:



wherein each R is independently selected from the group consisting of C₂ to C₂₀ aliphatic and cycloaliphatic trivalent organic radicals and C₆ to C₂₀ aromatic trivalent organic radicals; each R' is independently selected from the group consisting of hydrogen, C₁ to C₆ aliphatic and cycloaliphatic monovalent organic radicals and C₆ to C₁₂ aromatic monovalent organic radicals; and G is the radical remaining after removal of the hydroxy groups of a long chain ether glycol having an average molecular weight of from about 600 to 12000.

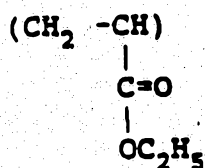
24. A process as defined in Claim 23 wherein said polyoxyalkylene diimide diacid is derived from trimellitic anhydride and a polyoxyalkyl diamine selected from the group consisting of polypropylene oxide diamine and a copoly(ethylene oxide-propylene oxide)diamine having predominantly polyethylene oxide in the backbone.

25. A process as defined in Claim 19 wherein said rubbery alkylacrylate consists essentially of repeating units of the formulae :



where n is 2 or 4 and m is 1 or 2 and a cure site monomer.

26. A process as defined in Claim 25 wherein said rubbery alkylacrylate is based on repeating units of the formula:



27. A process as defined in Claim 19 wherein said crosslinking agent is selected from the group sodium stearate, potassium stearate and a mixture thereof.

28. A process as defined in Claim 19 wherein said polyetherimide ester component (i) comprises from 20 to 99 parts by weight and said rubbery alkylacrylate component (ii) comprises from 80 to 1 part by weight based upon 100 parts by weight of (i) and (ii).

29. A process as defined in Claim 28 wherein said component (i) comprises from 20 to 80 parts by weight and said component (ii) comprises from 80 to 20 parts by weight.

30. A process as defined in Claim 29 wherein said component (i) comprises from 40 to 60 parts by weight and said component (ii) comprises from 60 to 40 parts by weight.

31. A process as defined in Claim 28 wherein said component (i) comprises 50 parts by weight and said component (ii) comprises 50 parts by weight.

32. A process as defined in Claim 29 wherein step (I) further comprises mixing (iii) a filler, a stabilizer, an antidegradant, a processing aid, a plasticizer, a pigment, or mixtures of any of the foregoing.

33. A process as defined in Claim 32 wherein said filler is selected from the group consisting of carbon blacks, silicas, clays and minerals.

34. A process as defined in Claim 32 wherein said plasticizer comprises a low molecular weight plasticizer or a high molecular weight plasticizer or both.

35. A process as defined in Claim 19 wherein said step (II) further

comprises adding an accelerator.

36. A process as defined in Claim 35 wherein said accelerator is selected from the group consisting of sulfur, sulfur donors, magnesium oxide, tertiary amines, quaternary ammonium compounds and mixtures of any of the foregoing.

37. A process as defined in Claim 36 wherein the crosslinker comprises sodium stearate and the accelerator comprises a quaternary ammonium compound.

38. A thermoplastic elastomer composition substantially as herein described with reference to Examples 1 to 18.

39. A process for producing a thermoplastic elastomer composition substantially as herein described with reference to Examples 1 to 18.

DATED this 12th day of July, 1993.

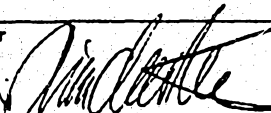
GENERAL ELECTRIC CO.

by its Patent Attorneys

DAVIES COLLISON CAVE

INTERNATIONAL SEARCH REPORT

International Application No PCT/US 91/00430

I. CLASSIFICATION OF SUBJECT MATTER (If several classification symbols apply, indicate all) ⁶		
According to International Patent Classification (IPC) or to both National Classification and IPC		
IPC5: C 08 L 79/08, C 08 L 33/08		
II. FIELDS SEARCHED		
Minimum Documentation Searched ⁷		
Classification System	Classification Symbols	
IPC5	C 08 L	
Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in Fields Searched ⁸		
III. DOCUMENTS CONSIDERED TO BE RELEVANT⁹		
Category ¹⁰	Citation of Document, ¹¹ with indication, where appropriate, of the relevant passages ¹²	Relevant to Claim No. ¹³
Y	EP, A2, 0327010 (E.I. DU PONT DE NEMOURS AND COMPANY) 9 August 1989, see page 3, line 10 - line 20; page 5, line 25 - line 55; claims 1-11	1-38
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Y	EP, A1, 0318786 (GENERAL ELECTRIC COMPANY) 7 June 1989, see page 6, line 55 - line 56; claims 1-17	1-38
	--	
Y	EP, A2, 0179470 (GENERAL ELECTRIC COMPANY) 30 April 1986, see the whole document	1-6,9-12
	--	
¹⁰ 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 document 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 "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		
IV. CERTIFICATION		
Date of the Actual Completion of the International Search	Date of Mailing of this International Search Report	
23rd April 1991	13. 05. 91	
International Searching Authority	Signature of Authorized Officer	
EUROPEAN PATENT OFFICE	Miss T. MORTENSEN 	

III. DOCUMENTS CONSIDERED TO BE RELEVANT (CONTINUED FROM THE SECOND SHEET)		
Category *	Citation of Document, with indication, where appropriate, of the relevant passages	Relevant to Claim No
A	EP, A1, 0372310 (GENERAL ELECTRIC COMPANY) 13 June 1990, see the whole document	1-38
A	EP, A2, 0273158 (GENERAL ELECTRIC COMPANY) 6 July 1988, see the whole document	1-38

**ANNEX TO THE INTERNATIONAL SEARCH REPORT.
ON INTERNATIONAL PATENT APPLICATION NO.PCT/US 91/00430**

SA 44090

This annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report.
The members are as contained in the European Patent Office EDP file on 23/03/91
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Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP-A2- 0327010	09/08/89	JP-A- 1266154 US-A- 4981908	24/10/89 01/01/91
EP-A1- 0318786	07/06/89	JP-A- 1213358 US-A- 4891397	28/08/89 02/01/90
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EP-A1- 0372310	13/06/90	JP-A- 2199153	07/08/90
EP-A2- 0273158	06/07/88	JP-A- 63199756	18/08/88

For more details about this annex : see Official Journal of the European patent Office, No. 12/82