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(54) Title: DIRECTLY PAINTABLE THERMOPLASTIC OLEFIN COMPOSITION WITH IMPROVED CONDUCTIVITY

(57) Abstract: Directly paintable polymer compositions contain (1) a thermoplastic polyolefin, (2) a propylene homopolymer or propylene copolymer with ethylene or a C_{4-8} α-olefin, grafted with an anhydride of an aliphatic α,β-unsaturated dicarboxylic acid, (3) an oxidized polyethylene wax having a melting point of less than 116°C and an acid number of less than 40, (4) a functionalized polymer that reacts with the anhydride groups of the grafted polymers, (5) an epichlorohydrin rubber and, optionally, (6) a polyolefin rubber grafted with an anhydride of an aliphatic α,β-unsaturated dicarboxylic acid, (7) an ethylene polymer grafted with an anhydride of an aliphatic α,β-unsaturated dicarboxylic acid, (8) a thermoplastic resin, and (9) an organic sulfonic acid salt of a group I or II metal, or mixtures thereof; the compositions are particularly suitable for electrostatic painting.

DIRECTLY PAINTABLE THERMOPLASTIC OLEFIN COMPOSITION WITH IMPROVED CONDUCTIVITY

Field of the Invention

This invention relates to directly paintable thermoplastic olefin compositions comprising an epichlorohydrin rubber; said compositions have improved electrical conductivity and therefore are particularly suitable to electrostatic painting. The TPO compositions of the invention are useful for making injection molded parts, such as automobile bumpers, and exhibit excellent paint adhesion and durability.

Background of the Invention

Thermoplastic polyolefins (TPOs) are uncrosslinked blends of olefin polymers and polyolefin elastomers. They can be made by physically blending in an internal mixer, or by polymerizing in a reactor. These materials are not paintable or coatable, because the paints or coatings consist of polar materials like urethanes, acrylics, epoxies, or melamines that have very poor adhesion to non-polar materials like polyolefins. Typically an adhesion promoter is used as the tie layer between the TPO substrate and the paint coating. This extra step adds to the cost of the product, and the coating is not very durable.

US Patent 5,962,573 discloses a directly paintable polymer composition comprising: (1) a thermoplastic polyolefin, (2) a propylene homopolymer or a propylene copolymer with ethylene or a C_{4-8} α-olefin, grafted with an anhydride of an aliphatic α,β-unsaturated dicarboxylic acid; (3) an oxidized polyethylene wax having a melting point of less than 116°C and an acid number of less than 40; (4) a functionalized polymer that is reactive with the anhydride groups of the grafted polymers and, optionally, (5) a polyolefin rubber grafted with an anhydride of an aliphatic α,β-unsaturated dicarboxylic acid, and (6) an ethylene polymer grafted with an anhydride of an aliphatic α,β-unsaturated dicarboxylic acid.

Injections molded parts such as automobile bumpers made from this composition are directly paintable with polar paints or coatings without the need for a layer of adhesion promoter between the thermoplastic polyolefin surface and the paint, and exhibit good paint adhesion and durability.

An important parameter in the paint process is the efficiency of the paint being sprayed; in fact, since the paint is airborne, some of it does not arrive onto the part to be treated and some

is lost in the environment. Electrostatic painting of substrates tends to reduce paint waste and emissions, as compared to non-electrostatic painting techniques.

Electrostatic painting techniques require the substrate to be electrically conducting and common TPOs for injection molded articles are electrically insulating; one of the methods to solve this problem, according to the prior art, consists in applying an electrically conductive primer prior to painting, in order to display an increased paint transfer efficiency. Nevertheless, this further step is costly and time consuming.

An alternative technique is to use a grounding clip, but this causes higher film buildup near the grounding clip, with film buildup decreasing as the distance from the grounding clip increases. In addition, after several passes through the paint spraying booth, significant resistance to ground may be encountered, due to multiple paint layers on the substrate itself.

A further way of improving the electrical conductivity is to incorporate additives, such as stainless steel fibers, into the thermoplastic polymer itself; nevertheless said additives are not suitable for TPO-based compositions because they lead to a degradation of their desirable mechanical properties, such as impact strength and tensile elongation. Moreover, said additives significantly increase the brittleness of such TPO compositions.

US 5,484,838 describes thermoplastic olefin compositions having improved electrical conductivity due to the addition of electrically conductive carbon black; in these compositions, at least a portion of the carbon black has to be dispersed within the crystalline polymer component of the TPO. Nevertheless, carbon black or additives of similar nature are difficult to handle and, as a consequence, their exact dispersion and the morphology needed for the TPO matrix are very difficult to control.

In addition, conductive carbon black may cause a reduction in the effectiveness of the directly paintable functionalized polymers, presumably by absorbing the materials onto the surface of the carbon black itself.

Thus, there is still a need for a TPO composition having an improved electrical conductivity and therefore allowing a better paint efficiency, at the same time meeting the stringent requirements for paint adhesion and durability of today's marketplace, particularly in the automotive industry.

Summary of the Invention

The composition of the present invention comprises, by weight:

(1) 100 parts of a thermoplastic polyolefin comprising an olefin polymer having an isotactic index of at least 80 and an olefin polymer rubber, the thermoplastic polyolefin having a rubber content of at least 20%;

(2) 5 to 20 parts per hundred parts of the thermoplastic polyolefin of a propylene homopolymer or propylene copolymer with ethylene or a C_{4-8} α-olefin having an ethylene or α-olefin content of 0.5% to 20%, grafted with an anhydride of an aliphatic α,β-unsaturated dicarboxylic acid, and having an anhydride content of 2% to 5%;

(3) 3 to 20 parts per hundred parts of the thermoplastic polyolefin of an oxidized polyethylene wax having a melting point of less than 116°C and an acid number of less than 40;

(4) a functionalized polymer that is reactive with the anhydride groups of the grafted polymer, selected from the group consisting of:

(a) 2 to 6 parts per hundred parts of the thermoplastic polyolefin of an amine-terminated polyalkylene glycol;

(b) 2 to 6 parts per hundred parts of the thermoplastic polyolefin of a hydroxy-terminated polyolefin;

(c) 2 to 6 parts per hundred parts of the thermoplastic polyolefin of a hydroxy-terminated polybutadiene;

(d) 2 to 8 parts per hundred parts of the thermoplastic polyolefin of a hydroxy-terminated olefin/alkylene oxide copolymer;

(e) 2 to 8 parts per hundred parts of the thermoplastic polyolefin of a hydroxy-terminated polyalkylene oxide;

(f) 2 to 8 parts per hundred parts of the thermoplastic polyolefin of a methoxy-terminated polyalkylene oxide;

(g) 2 to 8 parts per hundred parts of the thermoplastic polyolefin of an amine-terminated olefin/alkylene oxide copolymer, and

(h) mixtures thereof;

(5) 2 to 20 parts per hundred parts of the thermoplastic polyolefin of an epichlorohydrin rubber;

(6) optionally, 5 to 30 parts per hundred parts of the thermoplastic polyolefin of a polyolefin rubber grafted with an anhydride of an aliphatic α,β-unsaturated

dicarboxylic acid, having an anhydride content of at least 0.3% but less than 3%, and comprising a polymer of ethylene and a C_{3-8} α-olefin, optionally containing 0.5% to 10% of a diene, having an ethylene content of 30% to 70%;

(7) optionally, 5 to 20 parts per hundred parts of the thermoplastic polyolefin of an ethylene polymer grafted with an anhydride of an aliphatic α,β-unsaturated dicarboxylic acid, having an anhydride content of 1% to 16% and a number average molecular weight M_n of 500 to 5000, provided that at least 5 parts of anhydride-grafted polypropylene or propylene copolymer and 3 parts of oxidized polyethylene wax per hundred parts of the thermoplastic elastomer are also present;

(8) optionally, 5 to 20 parts per hundred parts of the thermoplastic polyolefin of a thermoplastic resin selected from the group consisting of:

(a) a hydrogenated polymer consisting essentially of terpene hydrocarbons, having a drop softening point greater than 70°C, and a number average molecular weight M_n greater than 500; and

(b) a hydrogenated polymer of styrene or alkyl-substituted styrene, having a number average molecular weight M_n ranging between 600 and 20,000; and

(9) optionally, 0.1 to 5 parts per hundred parts of the thermoplastic polyolefin of an organic sulfonic acid salt of a group I or II metal of the Periodic Table of the Elements (IUPAC version).

Injection molded parts, such as automobile bumpers, made from this composition have an increased conductivity and are particularly suitable for electrostatic painting. Such parts are directly paintable with polar paints or coatings with much higher paint efficiency with respect to the prior art compositions; molded parts do not require the use of a layer of adhesion promoter between the thermoplastic polyolefin surface and the paint, and exhibit excellent paint adhesion and durability.

Moreover, with respect to the compositions of the prior art, as described in US Patent 5,484,838, the composition of the invention does not display the problems associated with the use of carbon black, since epichlorohydrin rubbers are easy to handle and can be readily dispersed in the TPO matrix.

Finally, the addition of an epichlorohydrin rubber to the TPO composition does not alter the favorable mechanical properties of the composition itself.

A further object of the present invention is a molded thermoplastic article comprising a composition as described above, and more particularly a molded thermoplastic automotive article comprising a composition as described above.

Detailed Description of the Invention

Component (1) of the composition of this invention is a thermoplastic polyolefin comprising a crystalline olefin polymer and an olefin polymer rubber, the thermoplastic polyolefin having a rubber content of at least 20%. Suitable thermoplastic polyolefins include, for example,

(a) a composition comprising, by weight,

(i) 10% to 60%, preferably 20% to 50%, of a propylene homopolymer having an isotactic index greater than 90, preferably between 95 and 98, or a crystalline propylene copolymer with ethylene and/or a C_{4-8} α-olefin having a propylene content greater than 85% and an isotactic index of greater than 85;

(ii) 30% to 60%, preferably 30% to 50%, of an amorphous ethylene-propylene or ethylene-butene copolymer, optionally containing 1% to 10% of a diene, which is xylene soluble at room temperature and has an ethylene content of 30% to 70%;

(iii) 2% to 20%, preferably 7% to 15%, of a semi-crystalline ethylene-propylene or ethylene-butene copolymer that is xylene insoluble at room temperature and has an ethylene content greater than 75% but less than 92%; and

(iv) 5% to 20%, preferably 7% to 15%, of an ethylene polymer having a density of 0.91 to 0.96 g/cm^3 and a melt index of 0.1 to 100 g/10 min, preferably 15 to 50 g/10 min. Ethylene homopolymer is preferred. However, copolymers containing 8% or less of an α-olefin comonomer can also be used.

(b) a composition comprising, by weight:

(i) 20% to 70%, preferably 50% to 70%, of a crystalline propylene homopolymer having an isotactic index greater than 90, preferably between 95 and 98, or a crystalline propylene copolymer with ethylene and/or a C_{4-8} α-olefin having a propylene content greater than 85% and an isotactic index of greater than 85;

(ii) 20% to 75%, preferably 30% to 50%, most preferably 30% to 35%, of an amorphous copolymer of ethylene selected from the group consisting of (1) ethylene/propylene, (2) ethylene/butene-1, (3) ethylene/octene-1, and (4) mixtures thereof, optionally containing 1% to 10%, preferably 1% to 4%, of a diene, which is

xylene soluble at room temperature and has an ethylene content of 30% to 70%, preferably 40% to 60%; and

(iii) 2% to 30%, preferably 2% to 10%, most preferably 2% to 5%, of a semi-crystalline copolymer of ethylene selected from the group consisting of (1) ethylene/propylene, (2) ethylene/butene-1, (3) ethylene/octene-1, and (4) mixtures thereof, which is xylene insoluble at room temperature and has an ethylene content greater than 90%;

(c) a composition comprising, by weight,

(i) at least one heterophasic polyolefin composition comprising:

(1) 90% to 55% of a propylene polymer material selected from the group consisting of a propylene homopolymer having an isotactic index greater than 90, and a crystalline copolymer of propylene and an α-olefin of the formula CH_2=CHR, where R is H or C_2-C_6 alkyl, the α-olefin being less than 10% of the copolymer, and

(2) 10% to 45% of an elastomeric copolymer of propylene and an α-olefin of the formula CH_2=CHR, where R is H or C_2-C_6 alkyl, the α-olefin being 50% to 70% of the elastomeric copolymer, and 10% to 40% of the elastomeric copolymer being insoluble in xylene at ambient temperature, and

(ii) 5 to 50 parts, preferably 10 to 30 parts, and most preferably 10 to 25 parts, per hundred parts of (c)(i) of an elastomeric copolymer of ethylene and a C_{3-8} α-olefin made with a metallocene catalyst. If more than one heterophasic polyolefin (c)(i) is present, the heterophasic polyolefins can be combined in any proportion.

(d) a composition comprising, by weight:

(i) 30% to 50%, preferably 35% to 45%, of a propylene homopolymer having an isotactic index greater than 90, and

(ii) 70% to 50%, preferably 65% to 55%, of an olefin polymer composition comprising:

(1) 25% to 50% of a crystalline propylene homopolymer with a solubility in xylene at room temperature of less than or equal to 4%, or a crystalline copolymer of propylene with ethylene or a C_{4-8} α-olefin having an ethylene

or α-olefin content of 0.5% to 3%, and a solubility in xylene at room temperature of less than or equal to 4%, and

(2) 50% to 75% of an amorphous copolymer of ethylene and a C_{4-8} α-olefin, wherein the α-olefin content is 10% to 20%, and the copolymer is 10% to 40% soluble in xylene at room temperature; and

(e) a composition comprising, by weight:

(i) 80% to 30%, preferably 70% to 50%, of a propylene homopolymer having an isotactic index greater than 90, and

(ii) 20% to 70%, preferably 30% to 50%, of an elastomeric copolymer of ethylene and a C_{3-8} α-olefin, optionally containing 1% to 10%, preferably 1% to 4%, of a diene, and having an ethylene content of 30% to 70%, preferably 40% to 60%.

Thermoplastic polyolefins (a) and (c) are preferred.

Thermoplastic polyolefins (a) and (b) and compositions (c)(i) and (d)(ii) are typically prepared by sequential polymerization in at least two stages. Alternatively, the components can be prepared separately and then blended together by melt-kneading or melt blending. The polymerization conditions and the polymerization catalyst are described in more detail in U.S. Patents 5,143,978; 5,302,454; 5,360,868; and 5,486,419. Sequential polymerization is preferred.

For TPO (a), (i) can be made in the first reactor, (ii) and (iii) in the second reactor, and (iv) in the third reactor. Alternatively, (iv) can be made in the second reactor and (ii) and (iii) in the third reactor.

For TPO (b), (i), (ii), and (iii) are preferably formed in a reactor or series of reactors in at least two stages by first polymerizing propylene to form (i) and then polymerizing ethylene and propylene, butene-1, or octene-1, or mixtures thereof, in the presence of (i) and the catalyst used in the first stage to form (ii) and (iii). The polymerization can be conducted in the liquid or gas phase or in liquid-gas phase.

For TPO (b), (i) can be prepared using a Ziegler-Natta catalyst or a mixture of Ziegler-Natta and metallocene catalysts. Components (ii) and (iii) can be prepared using Ziegler-Natta or metallocene catalysts or a combination of the two, with one type of catalyst being used for one stage and the other type of catalyst being used for the next stage when the TPO is made by sequential polymerization.

The C_{4-8} α-olefins useful in the preparation of the thermoplastic polyolefins include, for example, butene-1; pentene-1; hexene-1; 4-methylpentene-1, and octene-1.

The diene, when present, is typically a butadiene; 1,4-hexadiene; 1,5-hexadiene, or ethylidenenorbornene.

Component (2) is a propylene homopolymer or propylene copolymer with ethylene or a C_{4-8} α-olefin, grafted with an anhydride of an aliphatic α,β-unsaturated dicarboxylic acid and having an ethylene or α-olefin content of 0.5% to 20%, preferably 1% to 10%, and most preferably 1% to 5%. The polymer has an anhydride content of 2% to 5%, preferably 3% to 4%, and preferably has a number average molecular weight M_n of 2500 to 25,000, more preferably 3000 to 10,000. Maleic anhydride is the preferred anhydride. Component (2) is present in an amount of 5 to 20 parts, preferably 8 to 16 parts, most preferably 10 to 14 parts, per hundred parts of the thermoplastic polyolefin.

Component (3) is an oxidized polyethylene wax having a melting point of less than 116°C and an acid number of less than 40. The oxidized wax is present in an amount of 3 to 20 parts, preferably 5 to 15 parts, most preferably 5 to 10 parts, per hundred parts of the thermoplastic polyolefin, whether or not the anhydride-grafted olefin polymer rubber (6) is present.

Component (4) is a functionalized polymer that is reactive with the anhydride groups of the grafted polymers (2) and, when present, components (6) and (7), selected from the group consisting of (a) an amine-terminated polyalkylene glycol, (b) a hydroxy-terminated polyolefin, (c) a hydroxy-terminated polybutadiene, (d) hydroxy-terminated olefin/alkylene oxide copolymers, (e) hydroxy-terminated polyalkylene oxides, (f) methoxy-terminated polyalkylene oxides, (g) amine-terminated olefin/alkylene oxide copolymers, and (h) mixtures thereof.

Component (4)(a), when present, is used in an amount of 2 to 6 parts, preferably 2 to 4 parts, per hundred parts of the thermoplastic polyolefin. The polyalkylene glycol can be, for example, polyethylene glycol, polypropylene glycol, copolymers of ethylene glycol and propylene glycol, poly(1,2-butylene glycol), and poly(tetramethylene glycol).

Component (4)(b), when present, is used in an amount of 2 to 6 parts, preferably 2 to 4 parts, per hundred parts of the thermoplastic polyolefin. Polyethylene is the preferred polyolefin,

although polypropylene, polybutene, and copolymers of ethylene and another α-olefin can also be used.

Component (4)(c), when present, is used in an amount of 2 to 6 parts, preferably 2 to 4 parts, per hundred parts of the thermoplastic polyolefin.

Component (4)(d), when present, is used in an amount of 2 to 8 parts, preferably 2 to 6 parts, per hundred parts of the thermoplastic polyolefin. An ethylene/ethylene oxide copolymer is preferred, although other copolymers such as ethylene/propylene oxide, propylene/ethylene oxide, butene/ethylene oxide, and butene/propylene oxide copolymers can also be used. The amount of alkylene oxide can be from 10% to 99.9%, preferably 50% to 98%, and most preferably 75% to 95%, based on 100% of the copolymer.

Component (4)(e), when present, is used in an amount of 2 to 8 parts, preferably 2 to 6 parts, per hundred parts of the thermoplastic polyolefin. Polyethylene oxide is preferred; however, polypropylene oxide, copolymers of ethylene oxide and propylene oxide, poly(1,2-butylene oxide), and poly(tetramethylene oxide) can also be used.

Component (4)(f), when present, is used in an amount of 2 to 8 parts, preferably 2 to 6 parts, per hundred parts of the thermoplastic polyolefin. Suitable polyalkylene oxides are those described for component (4)(e).

Component (4)(g), when present, is used in an amount of 2 parts to 8 parts, preferably 2 to 6 parts, per hundred parts of the thermoplastic polyolefin. The amount of alkylene oxide can be 10% to 99.9%, preferably 50% to 98%, and most preferably 75% to 95%. Examples of suitable olefin/alkylene oxide copolymers are described for component (4)(d).

When using a combination of functionalized polymers, the amount of each component can vary widely from 0.1% to 99.9% of each, based on the total amount of functionalized polymers. It is preferred that one component be present in an amount of >50%, preferably >60%, based on the total amount of functionalized polymers.

Instead of adding the functionalized polymer directly to the thermoplastic polyolefin, an adduct of the functionalized polymer and the anhydride-grafted polypropylene or ethylene/propylene copolymer can be prepared separately, then blended with the thermoplastic polyolefin.

Component (5) is an epichlorohydrin rubber; suitable epichlorohydrin rubbers include:

(5)(a) homopolymers of epichlorohydrin;

(5)(b) copolymers of epichlorohydrin with from 1% to 30% by mole of a saturated epoxy monomer or of an unsaturated epoxy monomer; and

(5)(c) terpolymers of epichlorohydrin with from 1% to 30% by mole of a saturated epoxy monomer and an unsaturated epoxy monomer; more preferably, the amount of such saturated epoxy monomer ranges from 1 to 10% by mole.

Said epichlorohydrin rubbers generally have high molecular weights, density values ranging from 1.35 g/cm^3 to 1.38 g/cm^3, and a Mooney viscosity after 4 minutes at 212°F of from 40 to 80 ML. These rubbers can evidence certain degrees of crystallinity.

The epichlorohydrin rubbers may be prepared according to the methods known in the state of the art, for instance by polymerizing monomeric epichlorohydrin alone or together with one or more of the above-mentioned epoxy monomers in the presence of suitable catalysts, such as organometallic catalysts.

Typical saturated epoxy monomers include alkylene oxides, such ethylene oxide and propylene oxide, and typical unsaturated epoxy monomers include allylglycidyl ether.

The epichlorohydrin rubber is present in an amount of 2 to 20 parts, preferably 5 to 15 parts, most preferably 10 parts, per hundred parts of the thermoplastic polyolefin.

Optional component (6) is an olefin polymer rubber grafted with an anhydride of an aliphatic α,β-unsaturated dicarboxylic acid and comprising a polymer of ethylene and a C_{3-8} α-olefin, optionally containing 0.5% to 10% of a diene, preferably 2% to 6%. The anhydride-grafted polyolefin rubber has an ethylene content of 30% to 70%, preferably 40% to 60%, and has an anhydride content of at least 0.3% but less than 3%. Maleic anhydride is the preferred anhydride. When present, the anhydride-grafted rubber is used in an amount of 5 to 30 parts, preferably 5 to 15 parts, most preferably 5 to 12 parts, per hundred parts of the thermoplastic polyolefin.

Optional component (7) is an ethylene polymer grafted with an anhydride of an aliphatic α,β-unsaturated dicarboxylic acid and having an anhydride content of 1% to 16% by weight, preferably 2% to 13%, most preferably 3% to 13%. Maleic anhydride is the preferred anhydride. Ethylene homopolymer is preferred. However, copolymers containing 10% or less of an α-olefin comonomer can also be used. The ethylene polymer preferably has a number average molecular weight M_n of 500 to 5000, preferably 600 to 2000, most preferably 600 to 1000. When component (6) is used, it is present in an amount of 5 to 20 parts, preferably 5 to

10 parts, per hundred parts of the thermoplastic polyolefin, provided that at least 5 parts of anhydride-grafted polypropylene or propylene copolymer and 3 parts of oxidized polyethylene wax per hundred parts of the thermoplastic polyolefin are also present.

Optional component (8) is a thermoplastic resin selected from the group consisting of:

(a) a hydrogenated polymer consisting essentially of terpene hydrocarbons, having a drop softening point greater than 70°C, and a number average molecular weight greater than 500; and

(b) a hydrogenated polymer of styrene or alkyl-substituted styrene, having a number average molecular weight M_n ranging between 600 and 20,000.

Components (8)(a) are amorphous terpene hydrocarbons polymers, preferably having a drop softening point greater than 100°C. The softening point of the polymers is the temperature (°C) at which the polymer changes from a rigid to a soft state, as determined by the Hercules drop method (Hercules Method R 25-3: Softening Point - Thermometer Drop Method, 1993).

Components (8)(a) have preferably a number average molecular weight M_n greater than 600 and a iodine value of less than 50, and even more preferably a iodine value of less than 15. The iodine value of the hydrocarbon polymers was determined in accordance with Method No. L 8a-57 of the American Oil Chemistry Society.

These amorphous terpene hydrocarbons polymers include the polymers produced by the hydrogenation of the resinous polymerization products obtained by the catalytic polymerization of mixed unsaturated monomers derived from the deep cracking of petroleum, as well as higher polymers obtained by polymerization and/or copolymerization of terpene hydrocarbons, followed by hydrogenation under pressure. Suitable components (8)(a) and the process for their preparation are described in U.S. Patent 3,361,849.

Component (8)(b) is an hydrogenated polymer of styrene or alkyl-substituted styrene, having a number average molecular weight M_n ranging between 600 and 20,000.

By "hydrogenated polymer of styrene or alkyl-substituted styrene" is meant a polymer selected from the group consisting of homopolymers of styrene and alkyl-substituted styrenes, copolymers of styrene and alkyl-substituted styrenes with each other, and copolymers of styrene and alkyl-substituted styrenes with other hydrocarbons having non-aromatic carbon-to-carbon unsaturations, said polymer being characterized by having at least 50% of its aromatic unsaturations hydrogenated, preferably at least 70% of its aromatic unsaturations

hydrogenated, and a number average molecular weight M_n ranging from 600 to 20,000; said polymers have preferably a drop softening point between 70° and 170°C.

Suitable components (8)(b) and the process for their preparation are described in U.S. Patent 3,666,836.

When present, the thermoplastic resin (8) is used in an amount of 5 to 20 parts, preferably 8 to 6, most preferably 10 to 14 per hundred parts of the thermoplastic polyolefin.

Optional component (9) is an organic sulfonic acid salt of a group I or II metal of the Periodic Table of the Elements (IUPAC version).

Suitable organic sulfonic acids are sulfonic acids bearing a radical selected from saturated or unsaturated, linear or cyclic C_1-C_{20} alkyl, C_3-C_{20} cycloalkyl, C_6-C_{20} aryl, C_7-C_{20} alkylaryl and C_7-C_{20} arylalkyl radicals, optionally containing one or more atoms belonging to groups 13-17 of the Periodic Table of the Elements (new IUPAC notation), such as B, N, P, Al, Si, Ge, O, S, Cl and F; more preferably, said radical is selected from the group consisting of trifluoromethyl, benzene, C_1-C_5 alkyl-benzene, naphthalene and C_1-C_5 alkyl-naphthalene.

Preferred metals belong to group I, and more preferably are selected from the group consisting of Li, Na and K.

When present, the organic sulfonic acid salt is used in an amount of 0.1 to 5 parts, preferably 0.5 to 1.5 per hundred parts of the thermoplastic polyolefin.

According to a preferred embodiment, the directly paintable polymer compositions of the present invention contain at least one of the components selected from the components (6), (7), (8) and (9) described above.

The composition of the present invention can also contain other conventional additives, for example, antioxidants; stabilizers; extender oils such as paraffinic and naphthenic oils; fillers such as $CaCO_3$, talc, Al_2O_3, carbon black, and zinc oxide; or flame retardants.

If non-polymeric additives such as conductive or non-conductive carbon black are used, they are preferably added after the functionalized polymer has reacted with the anhydride-grafted polymers. The additives can also be added as a dispersion in a polymer, preferably an olefin polymer.

The compounding or melt blending of the components of the composition can be carried out on an open roll, in an internal mixer (Banbury or Haake mixers), or in single-screw or twin screw extruders.

The compositions of this invention can be formed in any way, such as by extrusion, compression molding, and thermoforming; injection molding is preferred. They can also be co-extruded or co-injection molded with other polyolefin materials such as propylene homopolymers, copolymers, and graft copolymers, ethylene homopolymers and copolymers, or thermoplastic polyolefins such as those described previously. They can also be co-extruded or co-injection molded with olefin-based dynamically vulcanized elastomers or olefin-compatible thermoplastic elastomers such as styrene/butadiene copolymers.

The compositions of the present invention show an increased electrical conductivity; in fact, the volume resistivity, determined by ASTM D257 method, is preferably lower than $1 \cdot 10^{15}$ ohm·cm, and more preferably lower than $5 \cdot 10^{14}$ ohm·cm. As described above, the epichlorohydrin rubber component in the compositions of the invention leads to a significant electrical conductivity increase, without detrimental impact on the chemical/physical properties of the thermoplastic compositions; this significant increase is critical for reaching electrical dissipating molded articles.

The compositions according to the present invention can be used to make a variety of articles, such as automotive fascia, rocker panels, spoilers, bumpers and other vehicle exterior or interior trim components. Since the compositions of the present invention have an increased electrical conductivity, they are particularly suitable for articles which are subject to painting.

General Procedures and Characterizations

Paint Adhesion Test and Durability Test

The specimens for testing were prepared using a pin-gated mold rather than the fan-gated mold typically used for molding thermoplastic polyolefins. Durability depends upon the paint thickness; the thicker the paint or film, the better the durability. In the following examples and comparative examples only one coat of paint was used with an approximately 1.2 mil ($3.048 \cdot 10^{-5}$ m) film thickness, which is a very severe test. A typical durability test used in the automotive industry also employs a top coat that has a low coefficient of friction, which reduces the severity of the test; no top coat was used in the following examples and comparative examples.

The samples for testing were prepared by dry blending the ingredients and reactive mixing in a twin screw extruder at a temperature of 450°F (232°C) and pelletizing the resultant material. The pellets were injection molded into disks that were painted with a 1.2 to 2 mil ($3.048 \cdot 10^{-5}$

to $5.0 \cdot 10^{-5}$ m) thick coating using DuPont 872 white paint and cured at 250°F (121°C) for thirty minutes. A lattice pattern of squares with each square about ¼ inch in size was scribed on the painted disk at the end opposite the gate area of the disk. Adhesive tape (3M 898) was pressed onto the paint and pulled off to test the amount of paint removed or the paint adhesion. The % failure was recorded as the % of the squares removed by the tape after one pull. The durability was determined by using a Taber abrader with a type C scuffing head assembly and a one pound load. The painted disk was placed in an oven at 70°C for one hour, removed and placed on the platform of the abrader. The scuffing head was placed in contact with the painted surface and the disk was rotated for a specified number of cycles. The amount of paint removed from the complete circumference subtended by the scuffing head was recorded as the % failure.

The criteria set for acceptable paint adhesion were <50% failure in the gate area of the disk and <10% in the area opposite the gate area after the first pull, and <85% failure in the gate area and <50% in the area opposite the gate after the fifth pull.

The criteria for satisfactory durability was <50% failure after 100 cycles.

Volume Resistivity Test

Volume Resistivity was measured according to ASTM D257 method, with a time of electrification equal to 5 seconds; acceptable values for volume resistivity are lower than $1 \cdot 10^{15}$ ohm·cm.

In this specification all parts and percentages are by weight unless otherwise noted. The following examples are given for illustrative and not limiting purposes.

Examples 1-2 and Comparative Examples 1-2

Examples 1 and 2 show the paint adhesion, durability and volume resistivity of compositions containing a thermoplastic polyolefin (TPO), a maleic anhydride-grafted polypropylene (MA-g-PP), an oxidized polyethylene wax, an amine-terminated polyethylene oxide (ATPEO) or an hydroxy-terminated ethylene/ethylene oxide copolymer (HO-E/EO), an epichlorohydrin rubber and a maleic anhydride-grafted polyolefin rubber (MA-g-rubber),.

The effect of the presence of the epichlorohydrin rubber, according to the composition of the invention, on the electrical conductivity of the compositions is demonstrated by the results shown in Table 1. Moreover, these results demonstrate that good levels of paint adhesion and durability are obtained with the compositions of the invention.

In Table 1, the TPO contained 55% propylene homopolymer, 3% semi-crystalline ethylene/propylene copolymer that had a propylene content of ~10% and was insoluble in xylene at room temperature (~ 23°C), 30% amorphous ethylene/propylene copolymer rubber that had an ethylene content of 50% and was soluble in xylene at room temperature, and 12% ethylene homopolymer having a melt index of about 50 g/10 min (ASTM-D1238, at 190°C and 2.16 Kg).

MA-g-PP was Epolene E-43 maleic anhydride-modified polypropylene wax, commercially available from Eastman Chemical Company.

The oxidized polyethylene wax was Petrolite C-3500 wax, commercially available from Petrolite Corporation and has the following properties: acid no.= 24, Mn = 1,500, viscosity = 30 cp at 149°C, Melt Index = 5,000 g/10 min, and Melting Point = 96°C.

The ATPEO was XTJ-418 monoamine-terminated polyethylene oxide, commercially available from Huntsman Corporation.

The HO-E/EO was Unithox 480 hydroxy-terminated ethylene/ethylene oxide copolymer, having M_n of 2250 and a hydroxyl number of 22, and is commercially available from Petrolite Corporation.

The epichlorohydrin rubber was Hydrin T, epichlorohydrin terpolymer with ethyleneoxide and allyglycidyl ether, commercially available from Zeon Chemicals, Inc.

The MA-g-rubber was Exxelor VA-1803, ethylene/propylene rubber containing 0.7% grafted maleic anhydride, commercially available from Exxon Chemical Company.

The antioxidant was Irganox B 225, a blend of 1 part Irganox 1010 tetrakis [methylene(3,5-di-tert-butyl-4-hydroxyhydrocinnamate)methane and 1 part Irgafos 168 tris(2,4-di-tert-butylphenyl) phosphite antioxidant, commercially available from CIBA Specialty Chemicals Company.

Table 1

	Ex. 1	**Ex. 2**	**Comp. Ex. 1**	**Comp. Ex. 2**
TPO	100	100	100	100
MA-g-PP	10	10	--	10
Oxidized PE wax	10	10	--	10
ATPEO	3	--	--	3
OH-E/EO	--	3	--	--
Epichlorohydrin rubber	10	10	--	--
MA-g-rubber	10	10	--	10
Antioxidant	0.2	0.2	--	0.2
Paint Adhesion (% Failure)	(g/op)	(g/op)	(g/op)	(g/op)
1st pull	0/0	0/0	100	0/0
3rd pull	0/0	0/0	n.d.	0/0
5th pull	4/0	0/0	n.d.	0/0
Durability (% Failure)				
25 cycles	0	0	100	0
100 cycles	0	0	n.d.	0
Volume Resistivity (ohm·cm)	$6.0 \cdot 10^{13}$	$2.0 \cdot 10^{14}$	$2.0 \cdot 10^{16}$	$1.2 \cdot 10^{16}$

n.d. = not determined

Examples 1 and 2 show the improvement in electrical conductivity, as shown by the volume resistivity values, when an epichlorohydrin rubber is used along with the TPO, MA-g-PP, oxidized polyethylene wax, ATPEO, hydroxy-terminated E/EO and MA-g-rubber. Moreover, the presence of an epichlorohydrin rubber does not affect paint adhesion and durability.

In Comparative Example 1, the composition comprises the same TPO as used in Examples 1 and 2, but does not contain an epichlorohydrin rubber, while Comparative Example 2 comprises a prior art TPO composition, as described in US 5,962,573, which does not contain an epichlorohydrin rubber; as evident from results reported in Table 1, both these compositions do not show acceptable electrical conductivity properties.

Examples 3-5

These Examples show the paint adhesion, durability and volume resistivity of compositions containing a thermoplastic polyolefin (TPO), a maleic anhydride-grafted polypropylene (MA-g-PP), an oxidized polyethylene wax, an amine-terminated polyethylene oxide (ATPEO) or an hydroxy-terminated ethylene/ethyleneoxide copolymer (HO-E/EO), an epichlorohydrin rubber, a maleic anhydride-grafted polyethylene (MA-g-PE), and a thermoplastic resin.

The effect of the presence of the epichlorohydrin rubber, according to the composition of the invention, on the volume resistivity of the compositions is demonstrated by the results shown in Table 2. Moreover, these results demonstrate that good levels of paint adhesion and durability are obtained with the compositions of the invention.

In Table 2, the TPO, the MA-g-PP, the oxidized PE wax, the ATPEO, the HO-E/EO, the epichlorohydrin rubber and the antioxidant were the same as in Examples 1-2 and Comparative Examples 1-2.

The MA-g-PE was Ceramer 1608, maleic anhydride-grafted polyethylene, having M_n of 700, maleic anhydride content of 12.7% by weight, and melting point of 121°C, commercially available from Petrolite Corporation.

The acid resin was Foral AX, thermoplastic acid resin produced by hydrogenating wood rosin to a high degree, having a weight average molecular weight M_w of 300 and an acid number of at least 158, commercially available from Hercules Incorporated.

Table 2

	Ex. 3	**Ex. 4**	**Ex. 5**
TPO	100	100	100
MA-g-PP	10	10	10
Oxidized PE wax	10	10	10
ATPEO	4	--	--
OH-E/EO	--	4	4
Epichlorohydrin rubber	10	10	10
Ma-g-PE	10	10	--
Acid resin	--	--	10
Antioxidant	0.2	0.2	0.2
Paint Adhesion (% Failure)	(g/op)	(g/op)	(g/op)
1st pull	0/0	0/0	0/0
3rd pull	0/0	12/0	18/0
5th pull	0/0	36/0	30/0
Durability (% Failure)			
50 cycles	15	20	5
100 cycles	30	38	35
Volume Resistivity (ohm·cm)	$6.0 \cdot 10^{13}$	$3.0 \cdot 10^{13}$	$2.0 \cdot 10^{14}$

Examples 3 to 5 show that a good volume resistivity is obtained by using an epichlorohydrin rubber along with the TPO, MA-g-PP, oxidized polyethylene wax, ATPEO, hydroxy-terminated E/EO and MA-g-PE. Moreover, the presence of an epichlorohydrin rubber does not affect paint adhesion and durability.

Examples 6-7 and Comparative Example 3

Examples 6 and 7 show the paint adhesion, durability and volume resistivity of compositions containing a thermoplastic polyolefin (TPO), a maleic anhydride-grafted polypropylene (MA-g-PP), an oxidized polyethylene wax, an hydroxy-terminated ethylene/ethyleneoxide copolymer (HO-E/EO), an epichlorohydrin rubber, and a Li or Na sulfonic acid salt.

Comparative Example 3 shows the use of a Na sulfonic acid salt along with a TPO composition of the prior art, in the absence of an epichlorohydrin rubber, in order to enhance the electrical conductivity of the compositions.

The TPO, the MA-g-PP, the oxidized PE wax, the ATPEO, the HO-E/EO, the MA-g-rubber, and the antioxidant were the same as in the preceding Examples.

The Na sulfonic acid salt was 2-naphthalenesulfonic acid sodium salt (90% purity), commercially available from Aldrich (catalogue no. 10,967-3, 1996-1997), while the Li sulfonic acid salt was lithium trifluoromethanesulfonate (96% purity), also commercially available from Aldrich (catalogue no. 28,266-9, 1996-1997).

The results are shown in Table 3.

Table 3

	Ex. 6	**Ex. 7**	**Comp. Ex. 3**
TPO	100	100	100
MA-g-PP	10	10	10
Oxidized PE wax	10	10	10
ATPEO	--	--	3
HO-E/EO	4	4	--
Epichlorohydrin rubber	10	10	--
MA-g-rubber	--	--	10
Antioxidant	0.2	0.2	0.2
Na sulfonic acid salt	1	--	1
Li sulfonic acid salt	--	1	--
Paint Adhesion (% Failure)	(g/op)	(g/op)	(g/op)
1st pull	0/0	0/0	2/0
3rd pull	0/0	6/0	12/0
5th pull	12/0	42/0	22/6
Durability (% Failure)			
50 cycles	5	5	0*
100 cycles	15	5	18
Volume Resistivity (ohm·cm)	$2 \cdot 10^{12}$	$3 \cdot 10^{13}$	$5 \cdot 10^{15}$

* Measured after 25 cycles.

The results reported in Table 3 show that a good volume resistivity is obtained by using an epichlorohydrin rubber and a Na or Li sulfonic acid salt along with the TPO, MA-g-PP, oxidized polyethylene wax, ATPEO and hydroxy-terminated E/EO; moreover, the presence of an epichlorohydrin rubber does not affect paint adhesion and durability.

Furthermore, the addition of salt in a TPO composition of the prior art, in the absence of an epichlorohydrin rubber (as in Comp. Ex. 3), slightly improves the electrical conductivity of the composition, but does not give adequate paint adhesion and durability.

Other features, advantages and embodiments of the invention disclosed herein will be readily apparent to those exercising ordinary skill after reading the foregoing disclosure. In this regard, while specific embodiments of the invention have been described in considerable detail, variations and modifications of these embodiments can be effected without departing from the spirit and scope of the invention as described and claimed.

Claims

1. A composition comprising, by weight:

(1) 100 parts of a thermoplastic polyolefin comprising an olefin polymer having an isotactic index of at least 80 and an olefin polymer rubber, the thermoplastic polyolefin having a rubber content of at least 20%;

(2) 5 to 20 parts per hundred parts of the thermoplastic polyolefin of a propylene homopolymer or propylene copolymer with ethylene or a C_{4-8} α-olefin having an ethylene or α-olefin content of 0.5% to 20%, grafted with an anhydride of an aliphatic α,β-unsaturated dicarboxylic acid and having an anhydride content of 2% to 5%;

(3) 3 to 20 parts per hundred parts of the thermoplastic polyolefin of an oxidized polyethylene wax having a melting point of less than 116°C and an acid number of less than 40;

(4) a functionalized polymer that is reactive with the anhydride groups of the grafted polymers, selected from the group consisting of:

(a) 2 to 6 parts per hundred parts of the thermoplastic polyolefin of an amine-terminated polyalkylene glycol;

(b) 2 to 6 parts per hundred parts of the thermoplastic polyolefin of a hydroxy-terminated polyolefin,

(c) 2 to 6 parts per hundred parts of the thermoplastic polyolefin of a hydroxy-terminated polybutadiene;

(d) 2 to 8 parts per hundred parts of the thermoplastic polyolefin of a hydroxy-terminated olefin/alkylene oxide copolymer;

(e) 2 to 8 parts per hundred parts of the thermoplastic polyolefin of a hydroxy-terminated polyalkylene oxide,

(f) 2 to 8 parts per hundred parts of the thermoplastic polyolefin of a methoxy-terminated polyalkylene oxide,

(g) 2 to 8 parts per hundred parts of the thermoplastic polyolefin of an amine-terminated olefin/alkylene oxide copolymer, and

(h) mixtures thereof;

(5) 2 to 20 parts per hundred parts of the thermoplastic polyolefin of an epichlorohydrin rubber;

(6) optionally, 5 to 30 parts per hundred parts of the thermoplastic polyolefin of an olefin polymer rubber grafted with an anhydride of an aliphatic α,β-unsaturated dicarboxylic acid, having an anhydride content of at least 0.3% but less than 3%, and comprising a polymer of ethylene and a C_{3-8} α-olefin, optionally containing 0.5% to 10% of a diene, which contains 30% to 70% ethylene;

(7) optionally 5 to 20 parts per hundred parts of the thermoplastic polyolefin of an ethylene polymer grafted with an anhydride of an aliphatic α,β-unsaturated dicarboxylic acid, having an anhydride content of 1% to 16% and a number average molecular weight M_n of 500 to 5000, provided that at least 5 parts of anhydride-grafted polypropylene or propylene copolymer and 3 parts of oxidized polyethylene wax per hundred parts of the thermoplastic polyolefin are also present;

(8) optionally 5 to 20 parts per hundred parts of the thermoplastic polyolefin of a thermoplastic resin selected from the group consisting of:

(c) a hydrogenated polymer consisting essentially of terpene hydrocarbons, having a drop softening point greater than 70°C, and a number average molecular weight M_n greater than 500; and

(d) a hydrogenated polymer of styrene or alkyl-substituted styrene, having a number average molecular weight M_n ranging between 600 and 20,000; and

(9) optionally 0.1 to 5 parts per hundred parts of the thermoplastic polyolefin of an organic sulfonic acid salt of a group I or II metal.

2. The composition of claim 1, wherein the thermoplastic polyolefin (1) is a composition comprising, by weight:

(a) 10% to 60% of a propylene homopolymer having an isotactic index greater than 90, or a crystalline propylene copolymer with ethylene and/or a C_{4-8} α-olefin having a propylene content greater than 85% and an isotactic index of greater than 85;

(b) 30% to 60% of an amorphous ethylene-propylene or ethylene-butene copolymer, optionally containing 1% to 10% of a diene, which is xylene soluble at room temperature and having an ethylene content of 30% to 70%;

(c) 2% to 20% of a semi-crystalline ethylene-propylene or ethylene-butene copolymer that is xylene insoluble at room temperature and having an ethylene content of greater than 75% but less than 92%; and

(d) 5% to 20% of an ethylene polymer having a density of 0.91 to 0.96 g/cm^3 and a melt index of 0.1 to 100 g/10 min.

3. The composition of claim 1, wherein the thermoplastic polyolefin (1) is a composition comprising, by weight:

(a) 20% to 70% of a crystalline propylene homopolymer having an isotactic index greater than 90, or a crystalline propylene copolymer with ethylene and/or a C_{4-8} α-olefin having a propylene content greater than 85% and an isotactic index of greater than 85;

(b) 20% to 75% of an amorphous copolymer of ethylene selected from the group consisting of (i) ethylene/propylene, (ii) ethylene/butene-1, (iii) ethylene/octene-1, and (iv) mixtures thereof, optionally containing 1% to 10% of a diene, which is xylene soluble at room temperature and has an ethylene content of 30% to 70%; and

(c) 2% to 30% of a semi-crystalline copolymer of ethylene selected from the group consisting of (i) ethylene/propylene, (ii) ethylene/butene-1, (iii) ethylene/octene-1, and (iv) mixtures thereof, which is xylene insoluble at room temperature and has an ethylene content of greater than 90%.

4. The composition of claim 1, wherein the thermoplastic polyolefin (1) is a composition comprising, by weight:

(a) at least one heterophasic polyolefin composition comprising:

(i) 90% to 55% of a propylene polymer material selected from the group consisting of a propylene homopolymer having an isotactic index greater than 90, and a crystalline copolymer of propylene and an α-olefin of the formula CH_2=CHR, where R is H or C_2-C_6 alkyl, the α-olefin being less than 10% of the copolymer, and

(ii) 10% to 45% of an elastomeric copolymer of propylene and an α-olefin of the formula CH_2=CHR, where R is H or C_2-C_6 alkyl, the α-olefin being 50% to 70% of the elastomeric copolymer, and 10% to 40% of the elastomeric copolymer being insoluble in xylene at ambient temperature, and

(b) 5 to 50 parts per hundred parts of (a) of an elastomeric copolymer of ethylene and a C_{3-8} α-olefin made with a metallocene catalyst.

5. The composition of claim 1, wherein the thermoplastic polyolefin (1) is a composition comprising, by weight:

(a) 30% to 50% of a propylene homopolymer having an isotactic index greater than 90, and

(b) 70% to 50% of an olefin polymer composition comprising:

(i) 25% to 50% of a crystalline propylene homopolymer with a solubility in xylene at room temperature of less than or equal to 4%, or a crystalline copolymer of propylene with ethylene or a C_{4-8} α-olefin having an ethylene or α-olefin content of 0.5% to 3%, and a solubility in xylene at room temperature of less than or equal to 4%, and

(ii) 50% to 75% of an amorphous copolymer of ethylene and a C_{4-8} α-olefin, wherein the α-olefin content is 10% to 20%, and the copolymer is 10% to 40% soluble in xylene at room temperature.

6. The composition of claim 1, wherein the thermoplastic polyolefin (1) is a composition comprising, by weight:

(a) 80% to 30% of a propylene homopolymer having an isotactic index greater than 90, and

(b) 20% to 70% of an elastomeric copolymer of ethylene and a C_{3-8} α-olefin, optionally containing 1% to 10% of a diene, and having an ethylene content of 30% to 70%.

7. The composition of claim 1, wherein the anhydride of the aliphatic α,β-unsaturated dicarboxylic acid is maleic anhydride.

8. The composition of claim 1, wherein the functionalized polymer (4) is selected from the group consisting of (a) amine-terminated polyethylene oxide, (b) hydroxy-terminated polyethylene oxide, and (c) a hydroxy-terminated ethylene/ethylene oxide copolymer.

9. The composition of claim 1, wherein the epichlorohydrin rubber (5) is selected from the group consisting of (a) homopolymers of epichlorohydrin, (b) copolymers of epichlorohydrin with from 1% to 30% by mole of a saturated epoxy monomer or of an unsaturated epoxy monomer, and (c) terpolymers of epichlorohydrin with from 1% to 30% by mole of a saturated epoxy monomer and an unsaturated epoxy monomer.

10. A molded thermoplastic article comprising a composition as described in claim 1.

11. A molded thermoplastic automotive article comprising a composition as described in claim 1.