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(54) Title: IMPACT-RESISTANT POLYOLEFIN COMPOSITIONS

(57) Abstract: A polyolefin composition comprising, all percentages being by weight: A) from 40 to 75% of a polypropylene component having a solubility in xylene at room temperature lower than 20% by weight; B) from 3 to 20% of a copolymer of ethylene with 15-35% by weight of C_4-C_{10} α-olefin(s) having solubility in xylene at room temperature greater than 50% by weight, the intrinsic viscosity of the xylene soluble fraction being from 2.5 to 4 dl/g; C) from 10 to 35% of one or more elastomeric or plastomeric polymers different from B), having a Shore A hardness value equal to or lower than 90 points; D) from 5 to 25% of a mineral filler; and, optionally E) from 0.5 to 5% of a copolymer, different from C), of ethylene with propylene having an ethylene content from 20 to 70% by weight and a solubility in xylene at room temperature greater than 50% by weight, the intrinsic viscosity of the xylene soluble fraction being from 4 to 9 dl/g. wherein the amounts of A), B), C) and E) are referred to the total weight of A) + B) + C) + D) and optionally E).

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“IMPACT-RESISTANT POLYOLEFIN COMPOSITIONS”

The present invention relates to an impact resistant thermoplastic polyolefin composition. In particular, the present invention relates to a composition containing a propylene polymer component, a copolymer of ethylene with C_4-C_{10} α-olefins, other polyolefin elastomeric or plastomeric materials and a filler material.

It is known in the art of thermoplastic polymer compositions to combine propylene polymers, generally homopolymers or copolymers with minor amounts of comonomers, with elastomeric ethylene copolymers, to achieve a useful balance of rigidity (high flexural modulus) and impact resistance. For certain applications it is also desirable to achieve low values of thermal shrinkage. In fact, said property imparts a high dimensional stability to the final articles obtained from the said compositions.

In particular, in WO03/076511 and WO2005/121240 polyolefin compositions with low values of thermal shrinkage and good mechanical properties are described, comprising a propylene polymer component, a copolymer of ethylene with one or more C_4-C_{10} α-olefins, other elastomeric or plastomeric polyolefins and mineral fillers. As shown in the examples of the said documents, by using the therein disclosed technical solutions, flexural modulus values significantly higher than 1000 MPa are obtained only by adding around 20% by weight of mineral fillers. Moreover, the melt flow rate (MFR) of the compositions with high values of flexural modulus is relatively low.

It has now been found that by selecting specific propylene polymers and ethylene/α-olefin(s) copolymers, in combination with other features relating to the composition and the proportions of the various components, it is possible to obtain a desirable balance of mechanical properties (in particular flexural modulus and Izod impact strength), combined with a good rheology in the molten state and low values of thermal shrinkage.

Due to its mechanical and physical properties, the polyolefin composition of the present invention finds application above all in the automotive field, in particular in the preparation of door trims.

Thus the present invention relates to a composition comprising, all percentages being by weight:

A) from 40 to 75%, preferably from 45 to 65%, of a polypropylene component comprising a propylene homopolymer or a propylene copolymer with another α-olefin or combinations thereof, said polypropylene component containing at least 85% by weight of propylene and having a MFRL value equal to or higher than 90 g/10 min., preferably equal to or higher than 100 g/10 min., the upper limit being preferably of 2500 g/10 min., and a solubility in xylene at room temperature lower than 20% by weight;

B) from 3 to 20% of a copolymer of ethylene and one or more C_4-C_{10} α-olefin(s), containing 15-35% by weight of C_4-C_{10} α-olefin(s) and having a solubility in xylene at room temperature greater than 50% by weight, the intrinsic viscosity of the xylene soluble fraction being from 2.5 to 4 dl/g;

C) from 10 to 35%, preferably from 10 to 30, of one or more elastomeric or plastomeric polymers different from B), having a Shore A hardness value equal to or lower than 90 points, preferably lower than 80 points;

D) from 5 to 25%, preferably from 6 to 20%, of a mineral filler, preferably talc; and, optionally

E) from 0.5 to 5% of a copolymer, different from C), of ethylene with propylene optionally containing 0.5 to 5% by weight of a diene, said copolymer having an ethylene content from 20 to 70% by weight and a solubility in xylene at room temperature greater than 50% by weight, the intrinsic viscosity of the xylene soluble fraction being from 4 to 9 dl/g.

wherein the amounts of A), B), C), D) and E) are referred to the total weight of A) + B) + C) + D) and optionally E).

From the above definitions it is evident that the term “copolymer” includes polymers containing more than one kind of comonomers.

The composition of the present invention can also optionally comprise a nucleating agent F), in preferred amounts of 0.01 to 0.5 parts by weight with respect to 100 parts by weight of A) + B) + C) + D) and optionally E).

The composition of the present invention has preferably a MFR value of 20 g/10 min. or higher, or even of 23 g/10 min. or higher, for example in the range from 20 to 60 g/10 min., in particular from 23 to 60 g/10 min.

Thus the composition of the present invention can be easily converted into various kinds of finished or semi-finished articles, in particular by using injection-molding techniques, due to its relatively high values of MFR, associated with the said high balance of properties.

The MFRL value of component A) can result from mixing various propylene homoplymers and/or copolymers with different MFRL values.

In such a case the MFRL value for A) can be easily determined, on the basis of the amounts and MFRL values of the single polymers, by means of the known correlation between the

MFR of a polyolefin composition and the MFR of the separate components, which, for instance, in the case of two polymer components A^1 and A^2, can be expressed as follows:
$\ln MFR^A = [W_A{}^1/(W_A{}^1 + W_A{}^2)] \times \ln MFR^1 + [W_A{}^2/(W_A{}^1 + W_A{}^2)] \times \ln MFR^2$
wherein $W_A{}^1$ and $W_A{}^2$ represent the weight of components A^1) and A^2) respectively, while MFR^A represent the calculated value of MFR for A) and MFR^1 and MFR^2 represent the MFR of components A^1) and A^2) respectively.

The MFRL value of a single polymer can therefore be even lower than 90g/10 min.

Thus the definition of MFRL value of component A) includes also the said calculated value.

The amount of fraction (XI) insoluble in xylene at room temperature of component B) preferably satisfies the following equation:

$(XI) < 1.14 \times B_2 - 34$

where B_2 is the amount of ethylene in component B), expressed as percent by weight with respect to the weight of B).

The amount of component (A) which is soluble in xylene at room temperature is, as previously said, equal to or lower than 20%, preferably equal to or lower than 10% by weight.

Typically component (A) is selected from (i) a propylene homopolymer having a preferred solubility in xylene at room temperature lower than 10% by weight, more preferably lower than 5%, and even more preferably lower than 3%, or (ii) a copolymer of propylene with at least one α-olefin of formula $H_2CH=CHR$, where R is H or a C_{2-8} linear or branched alkyl, or a combination of (i) and (ii).

Preferably the copolymer of propylene (ii) contains at least 90% propylene, and has a preferred solubility in xylene at room temperature of lower than 15% by weight, more preferably lower than 10%, and even more preferably lower than 8%. Said α-olefin is preferably ethylene, butene-1, pentene-1, 4-methylpentene-1, hexene-1, octene-1 or any combinations thereof, and even more preferably the copolymer of propylene (ii) is a copolymer of propylene and ethylene.

As previously said, the components (B) and (E) are partially soluble in xylene at room temperature. The content of fraction of component (B) or (E) which is soluble in xylene at room temperature is preferably of about 50-95% by weight, more preferably 55-95% by weight.

The C_4-C_{10} α-olefins in component (B) are generally selected from olefins of formula $H_2CH=CHR$, where R is a C_{2-8} linear or branched alkyl.

Illustrative C_4-C_{10} α-olefins for component (B) include 1-butene, 1-pentene, 1-hexene, 4-methyl-1-pentene and 1-octene, with 1-butene being particularly preferred.

The composition of the present invention can be prepared by melt-blending components (A), (B), (C) and (D) and optionally (E) and/or (F).

The composition of the present invention can also be prepared by subjecting to melt-blending with the other polyolefin components, the component (D) and optionally (F), a masterbatch composition (I) comprising, all percentages being by weight:

A^I) from 60 to 85% of a polypropylene component comprising a propylene homopolymer or a propylene copolymer with another α-olefin or combinations thereof, said polypropylene component containing at least 85% by weight of propylene, and having a MFRL value equal to or higher than 20 g/10 min. and a solubility in xylene at room temperature lower than 20% by weight;

B^I) from 15 to 40% of a copolymer of ethylene and one or more C_4-C_{10} α-olefin(s), containing 15-35% by weight of C_4-C_{10} α-olefin(s) and having a solubility in xylene at room temperature greater than 50% by weight, the intrinsic viscosity of the xylene soluble fraction being from 2.5 to 4 dl/g.

Kinds of preferred polymers, kinds and amounts of comonomers and amounts of fraction soluble in xylene at room temperature for the masterbatch component (A^I) are the same as described above for component (A).

Kinds and amounts of comonomers and amounts of fraction soluble in xylene at room temperature for the masterbatch component (B^I) are the same as described above for component (B).

Preferably the masterbatch composition (I) has a value of melting enthalpy ΔHm of the DSC melting peak detectable at a temperature between 100 and 130°C of 1 J/g or more.

The elastomeric or plastomeric polymer or polymers C) can be any elastomeric or plastomeric polymer or polymer composition commonly used to modify the mechanical properties of polyolefins.

The term “plastomeric” is used to include the particular class of materials having properties intermediate to those of thermoplastic and elastomeric materials, generally called “plastomers”. Said plastomers can have a broad range of densities (up to about 0.925 g/cm^3) and a higher crystallinity than the traditional elastomers.

Be it an elastomeric or plastomeric material, the said component C) preferably has at least one of the following features, in addition to the said Shore A hardness value equal to or lower than 90 points:

- Flexural modulus (ISO 178A) equal to or less than 200 MPa, preferably equal to or less than 170 MPa, most preferably equal to or less than 100 MPa;

- Shore D hardness equal to or less than 50 points, more preferably equal to or less than 45 points and most preferably equal to or less than 32 points;
- X-ray crystallinity from 0 to 40%, more preferably from 0 to 30%.

Preferred examples of C) are:

1) ethylene copolymers containing up to 45% by weight, in particular from 10 to 42% by weight, of an olefin comonomer, preferably a C_3-C_{10} α-olefin, in particular butene-1 or octene-1, and having Shore A hardness of 90 points or less.
2) propylene copolymers containing up to 40% by weight of an olefin comonomer, preferably ethylene or a C_4-C_{10} α-olefin, and having Shore A hardness of 90 points or less.

The said ethylene copolymers 1) have typically a density from 0.86 to 0.925 g/cm^3.

A specific example of copolymers 1) is a copolymer containing 63.5 wt% of ethylene and 36.5% wt% of octene-1 (^{13}C-NMR analysis), having a hardness of 20 Shore D points and 72 Shore A points, a melting point of 60°C, a MFR of 30 g/10 min. (ASTM D 1238, 190°C/2.16 kg), an intrinsic viscosity of 0.9 dl/g and a density of 0.87 g/ml according to method ASTM D 792.

It is marketed by Dow Chemical with the trademark Engage 8407.

Another specific example of copolymers 1) is a copolymer containing 62 wt% of ethylene and 38 wt% of octene-1 (IR analysis), having a hardness of 21 Shore D points and 75 Shore A points, a melting point of 60°C, a MFR of 5 g/10 min. (ASTM D 1238, 190°C/2.16 kg) and a density of 0.87 g/ml according to method ASTM D 792.

It is marketed by Dow Chemical with the trademark Engage 8200.

Other polymer materials of this kind, sold by Dow under the trademark Affinity can also be conveniently used.

Another specific example of copolymers 1) is a copolymer containing 61 wt% of ethylene and 39 wt% of octene-1 (IR analysis), having a hardness of 20 Shore D points and 75 Shore A points, a melting point of 55°C, a density of 0.868 g/cm^3 according to method ASTM D 792 and MFR of 0.5 g/10 min (ASTM D 1238, 190°C/2.16 kg).

It is marketed by Dow Chemical with the trademark Engage 8150.

Another specific example of copolymers 1) is a copolymer of ethylene and butene-1, having a hardness of 56 Shore A points, melting point of 36°C, a density of 0.86 g/cm^3 according to method ASTM D 792, a MFR of 1.2 g/10 min (ASTM D 1238, 190°C/2.16 kg) and an intrinsic viscosity of 1.5 dl/g.

It is marketed by Dow Chemical with the trademark Engage ENR 7467.

Another specific example of copolymers 1) is a copolymer of ethylene and butene-1, having a hardness of 30 Shore D points, 85 Shore A points, flexural modulus of 22.8 MPa (ASTM D 790), melting point of 60°C, a density of 0.88 g/cm^3 according to method ASTM D 792 and an intrinsic viscosity of 1.6 dl/g.

It is marketed by Exxon Chemical with the trademark Exact 4033.

Preferred examples of propylene copolymers 2) are the propylene copolymers containing from 0.1 to 40% by weight, more preferably from 0.1 to 25% by weight of olefin comonomers, in particular ethylene.

The said propylene copolymers 2) have typically a density from 0.850 to 0.890 g/cm^3, in particular from 0.855 to 0.885 g/cm^3. They generally display no or relatively low degree of crystallinity, indicatively from 0 to 25% when measured as X- ray crystallinity.

Other typical properties of propylene copolymers 2) are:

- Shore A hardness preferably equal to or less than 88 points, more preferably equal to or less than 75 points;
- melting point, measured with differential scanning calorimetry (DSC) at a heating /cooling rate of 10-20°C, of 105°C or less, preferably of 90°C or less;
- heat of fusion, measured with DSC under the said conditions, of 75 J/g or less;
- molecular weight distribution, in terms of Mw/Mn (Mw = weight average molecular weight and Mn = number average molecular weight, both measured by gel permeation chromatography in trichlorobenzene at 135 °C) from 1.5 to 5, more preferably from 1.5 to 3.5.

Suitable propylene copolymers 2) are the plastomers Vistamaxx® and Versify® made available on the market by ExxonMobil Chemical and Dow Chemical, and the Notio® polymers, made available on the market by Mitsui Petrochemical.

The said copolymers 1) and 2) are typically produced by polymerization in the presence of single site, in particular metallocene and constrained geometry catalysts.

Particularly preferred as component C) are the said copolymers 1). Such copolymers have typically intrinsic viscosity values of 2 dl/g or less. Such intrinsic viscosity values substantially correspond to the intrinsic viscosity values of the fraction soluble in xylene at room temperature, due to the very high solubility of said copolymers.

In general, the components (A), (B), (C) and (E) can be prepared separately by using known polymerization processes.

The said masterbatch composition (I) can advantageously be prepared by a sequential polymerization, comprising at least two sequential steps, wherein components (A^I) and (B^I) are prepared in separate subsequent steps, operating in each step, except the first step, in the presence of the polymer formed and the catalyst used in the preceding step. The catalyst is added only in the first step, however its activity is such that it is still active for all the subsequent steps.

The polymerization, which can be continuous or batch, is carried out following known techniques and operating in liquid phase, in the presence or not of inert diluent, or in gas phase, or by mixed liquid-gas techniques.

Reaction time, pressure and temperature relative to the polymerization steps are not critical, however it is best if the temperature is from 50 to 100 °C. The pressure can be atmospheric or higher.

The regulation of the molecular weight is carried out by using known regulators, hydrogen in particular.

In order to prepare components (A), (B) and (E), the said polymerizations are preferably carried out in the presence of stereospecific Ziegler-Natta catalysts.

Thus component B) is preferably prepared by using a Ziegler-Natta catalyst in polymerization, while component C) is preferably prepared by using a single site catalyst in polymerization.

The said catalysts are known in the art.

Conventional molecular weight regulators known in the art, such as chain transfer agents (e.g. hydrogen or $ZnEt_2$), may be used.

Preferred examples of Ziegler-Natta catalysts are the supported catalyst systems comprising a trialkylaluminium compound, optionally an electron donor, and a solid catalyst component comprising a halide or halogen-alcoholate of Ti and optionally an electron-donor compound supported on anhydrous magnesium chloride. Catalysts having the above-mentioned characteristics and polymerization processes employing such catalysts are well known in the patent literature; particularly advantageous are the catalysts and polymerization processes described in USP 4,399,054 and EP-A-45 977. Other examples can be found in USP 4,472,524.

The catalysts can be pre-contacted with small amounts of olefins (prepolymerization).

Mineral fillers (D) present in the composition of the present invention include talc, $CaCO_3$, silica, mica, wollastonite ($CaSiO_3$), clays, diatomaceaous earth, titanium oxide and zeolites. Talc is preferred. Typically the mineral filler is in particle form having an average diameter ranging form 0.1 to 5 micrometers.

Useful nucleating agents (F) include, for example, metal salts of carboxylic acids, dibenzylsorbitol derivatives, alkali metal salts of phosphate and the like.

Specific examples of the nucleating agents include sodium benzoate, aluminum adipate, aluminum p-t-butylbenzoate, 1,3,2,4-dibenzylidenesorbitol, 1,3,2,4-bis(p-methylbenzylidene)sorbitol, 1,3,2,4-bis(p-ethylbenzylidene)sorbitol, 1,3-p-chlorobenzylidene-2,4-p-methylbenzylidene)sorbitol, sodium bis(4-t-butylphenyl) phosphate, sodium bis(4-t-methylphenyl) phosphate, potassium bis(4,6-di-t-butylphenyl) phosphate, sodium 2,2'-methylene-bis(4,6-di-t-butylphenyl) phosphate, sodium 2,2'-ethylidene-bis(4,6-di-t-butylphenyl) phosphate.

The composition of the present invention can also contain additives commonly employed in the art, such as antioxidants, light stabilizers, heat stabilizers and colorants.

As previously said, the composition of the present invention can be prepared by melt-blending the components (A), (B), (C), (D) and optionally (E) and/or (F) or by melt-blending the matsterbatch composition (I) with the other polyolefin components, and with the component (D) and optionally (F). Any mixing apparatus equipped with mixing elements and known in the art can be used, such as an internal mixer or extruder. For example one can use a Banbury mixer or single-screw Buss extruder or twin-screw Maris or Werner type extruder.

The present invention also provides final articles, in particular door trims, made of the said polyolefin composition.

The practice and advantages of the present invention are disclosed below in the following examples. These Examples are illustrative only, and are not intended to limit the scope of the invention in any manner whatsoever.

The following analytical methods are used to characterize the polymer compositions.

Melt Flow Rate

Determined according to ASTM-D 1238, condition L (i.e. 230°C with 2.16 kg load), unless differently specified.

[η] intrinsic viscosity: determined in tetrahydronaphtalene at 135°C.

Ethylene and butene content: I.R. Spectroscopy.

Flexural Modulus: ISO 178.

Tensile strength at break: ISO 527.

Elongation at break: ISO 527.

Notched IZOD impact test: ISO 180/1A.

Shore A and D: ASTM D2240.

Gloss

10 rectangular specimens (55x60x1 mm) for each polymer to be tested are prepared by injection molding using an injection press Battenfeld BA500CD operated under the following conditions:

Screw speed: 120 rpm
Back pressure: 10 bar
Mould temperature: 40°C
Melt temperature: 260°C
Injection time: 3 sec
First holding time: 5 sec
Second holding time: 5 sec
Cooling time (after second holding): 10 sec

The value of the injection pressure should be sufficient to completely fill the mould in the above mentioned indicated time span.

By a glossmeter the fraction of luminous flow reflected by the examined specimens surface is measured, under an incident angle of 60°. The value reported in Table III corresponds to the mean gloss value over 10 specimens for each tested polymer.

The glossmeter used is a photometer Zehntner model ZGM 1020 or 1022 set with an incident angle of 60°. The measurement principle is given in the Norm ASTM D2457. The apparatus calibration is done with a sample having a known gloss value.

Xylene soluble and isoluble fractions

2.5 g of polymer and 250 cm^3 of xylene are introduced in a glass flask equipped with a refrigerator and a magnetical stirrer. The temperature is raised in 30 minutes up to the boiling point of the solvent. The so obtained clear solution is then kept under reflux and stirring for further 30 minutes. The closed flask is then kept for 30 minutes in a bath of ice and water and in thermostatic water bath at 25 °C for 30 minutes as well. The so formed solid is filtered on quick filtering paper. 100 cm^3 of the filtered liquid is poured in a previously weighed aluminum container which is heated on a heating plate under nitrogen flow, to remove the solvent by evaporation. The container is then kept in an oven at 80 °C under vacuum until constant weight is obtained. The weight percentage of polymer soluble in xylene at room temperature is then calculated.

The percent by weight of polymer insoluble in xylene at room temperature is considered the crystallisable portion of the polymer. This value also corresponds substantially to the isotacticity index determined by extraction with boiling n-heptane, which by definition constitutes the isotacticity index of polypropylene.

Longitudinal and transversal thermal shrinkage

A plaque of 100 x 200 x 2.5 mm is moulded in an injection moulding machine "SANDRETTO serie 7 190" (where 190 stands for 190 tons of clamping force).

The injection conditions are:

melt temperature = 250°C;

mould temperature = 40°C;

injection time = 8 seconds;

holding time = 22 seconds;

screw diameter = 55 mm.

The plaque is measured 48 hours after moulding, through callipers, and the shrinkage is given by:

$$\text{Longitudinal shrinkage} = \frac{200 - read\_value}{200} \times 100$$

$$\text{Transversal shrinkage} = \frac{100 - read\_value}{100} \times 100$$

wherein 200 is the length (in mm) of the plaque along the flow direction, measured immediately after moulding;

100 is the length (in mm) of the plaque crosswise the flow direction, measured immediately after moulding;

the *read_value* is the plaque length in the relevant direction.

Determination of melting peaks and melting enthalpy (ΔHm) by differential scanning calorimetry (DSC)

The instrument used is a PerkinElmer Diamond DSC. The polymer sample is heated to 230°C at a rate of 10°C/min and kept at 230°C for 5 minutes in nitrogen stream and it is thereafter cooled at a rate of 10°C/min to 20°C, thereby kept at this temperature for 5 min to crystallise the sample. Then, the sample is again fused at a temperature rise rate of 10°C/min up to 230°C. The melting scan is recorded, a thermogram is obtained, and, from this, the temperature and fusion enthalpy value corresponding to the most intense peak between 100 and 130°C are read.

Preparation of the masterbatch composition (I)

Two masterbatch compositions are prepared. The solid catalyst component used in polymerization is a highly stereospecific Ziegler-Natta catalyst component supported on magnesium chloride, prepared according to the Example 5, lines 48-55 of the European Patent

EP728769. Triethylaluminium (TEAL) is used as co-catalyst and dicyclopentyldimethoxysilane (DCPMS) as external donor.

The solid catalyst component is contacted at 12° C for 24 minutes with TEAL and DCPMS. The weight ratio between TEAL and the solid catalyst component and the weight ratio between TEAL and DCPMS are of 20 and 10 respectively.

The catalyst system is then subjected to prepolymerization by maintaining it in suspension in liquid propylene at 20° C for about 5 minutes before introducing it into the first polymerization reactor.

POLYMERIZATION

The polymerisation run is conducted in continuous in a series of two reactors equipped with devices to transfer the product from one reactor to the one immediately next to it. The first reactor is a liquid phase reactor, and the second reactor is a fluid bed gas phase reactor. Component (A^I) is prepared in the first reactor, while component (B^I) is prepared in the second reactor, respectively.

Component (A^I) is a propylene homopolymer, while component (B^I) is an ethylene/butene-1 copolymer.

Temperature and pressure are maintained constant throughout the course of the reaction. Hydrogen is used as molecular weight regulator.

The gas phase (propylene, ethylene, butene and hydrogen) is continuously analysed via gas-chromatography.

Polymerization conditions, molar ratio of the reactants and composition of the polymer components obtained are shown in Table I.

The polymer particles exiting the second reactor are subjected to a steam treatment to remove the reactive monomers and volatile substances, and then dried.

Then the polymer particles are introduced in a rotating drum, where they are mixed with 0.05% by weight of paraffin oil ROL/OB 30 (having a density of 0.842 kg/l at 20 °C according to ASTM D 1298 and flowing point of –10 °C according to ASTM D 97), 0.15% by weight of Irganox® B 215 (made of about 34% Irganox® 1010 and 66% Irgafos® 168) and 0.04% by weight of DHT-4A (hydrotalcite).

The said Irganox 1010 is 2,2-bis[3-[,5-bis(1,1-dimethylethyl)-4-hydroxyphenyl)-1-oxopropoxy]methyl]-1,3-propanediyl-3,5-bis(1,1-dimethylethyl)-4-hydroxybenzene-propanoate, while Irgafos 168 is tris(2,4-di-tert.-butylphenyl)phosphite.

Then, the polymer particles are extruded under nitrogen in a screw extruder with a melt temperature of 200-250 °C.

The characteristics relating to the polymer compositions, reported in Table II, are obtained from measurements carried out on the so extruded polymer.

Table I

Masterbatch	1	2
1° Reactor (component (A^I))		
Temperature (° C)	70	70
Amount produced (wt%)	76.7	77.6
MFR L (g/10 min.)	44	68
Xylene soluble (wt%)	1.8	1.9
2° Reactor (component (B^I))		
Temperature (° C)	85	85
Pressure (MPa)	2	2
Amount produced (wt%)	23.3	22.4
C4/(C2+C4) mol	0.44	0.48
C4 in (B^I) (wt%)	26	29.5
Xylene soluble in (B^I) (wt%)	58.4	55.6
I.V. of xylene soluble of (B^I) (dl/g)	3.2	3.3
ΔHm (100-135°C peak) (J/g)	2.8	2.1

Notes: C2 = ethylene; C4 = butene; I.V. = Intrinsic Viscosity

Table II

Masterbatch	1	2
MFR L (g/10 min)	13.0	16.1
Xylene soluble (wt%)	15	14
Ethylene content (wt%)	17.2	15.8
Butene content (wt%)	6.1	6.6

Examples 1 and 2 and Comparative 1

The masterbatch compositions 1 to 3 prepared as described above, are mechanically mixed with the other components by extrusion under the previously described conditions. The proportions of the polyolefin components used in these examples are reported in Table III, together with the amounts of components A), B), C) and E) of the final composition, obtained by aggregating the contributions of said polyolefin components, and with the calculated value of MFR L of A), based on the previously reported correlation of MFR logarithms.

The following polyolefin components are used.

PP:	Propylene homopolymer having a MFRL value of 2000 g/10 min. and solubility in xylene at room temperature of 2.3% by weight;
EG8407	Engage 8407 as described above;
ENR7467	Engage ENR 7467 as described above;
Heco:	Polyolefin composition (heterophasic blend) comprising 67% by weight of propylene homopolymer having a MFRL value of 9 g/10 min. and solubility in xylene at room temperature of 2% by weight, and 33% by weight of ethylene/propylene copolymer containing 38% by weight of ethylene and having a solubility in xylene at room temperature of 84% by weight and an intrinsic viscosity of the xylene soluble fraction of 7.5 dl/g.

EG8407 and ENR7467 together constitute the component C) of the composition of the present invention.

In addition to the said polyolefin components, also talc and a nucleating agent, corresponding respectively to components D) and F), are added.

The talc D) used is Neotalc UNI manufactured by Neotalc Industries, Ltd . The amount of D) is reported in Table III as well.

Sodium-2,2'-methylene-bis(4,6-di-t-butylphenyl)-phosphate (trade name NA11, manufactured by Asahi Denka K. K.) is used as nucleating agent F). The amount of F) in the compositions of all the examples is 0.1 parts by weight with respect to 100 parts by weight of A) + B) + C) + D) + E).

The properties of the so obtained final compositions are reported in Table IV.

For comparison purpose, In Table III and Table IV are reported the data of a composition obtained by blending the same components as in Examples 1 and 2, but using a masterbatch composition identified as Comparison 1, having MFRL of 15 g/10 min. and comprising (percentages by weight):

A^I) 74% of a propylene homopolymer having a MFRL value of 20 g/10 min. and a solubility in xylene at room temperature of 1.7%;

B^I) 26% of a copolymer of ethylene and butene-1, containing 21% of butene-1 and having solubility in xylene at room temperature of 51%, the intrinsic viscosity of the xylene soluble fraction being of 1.2 dl/g.

Table III

EXAMPLE	1	2	Comp. 1
Masterbatch composition No.	1	2	Comp. 1
Amount of Masterbatch composition (wt%)	46	48	44
PP (wt%)	19	17	21
EG8407 (wt%)	5	5	5
ENR7467 (wt%)	15	15	15
Heco (wt%)	5	5	5
A) (wt%)	57.65	57.55	56.91
B) (wt%)	10.7	10.8	11.44
C) (wt%)	20	20	20
D) (wt%)	10	10	10
E) (wt%)	1.65	1.65	1.65
MFR L of A) (g/10 min)	136	169	102

Table IV

EXAMPLE	1	2	Comp.1
MFR L (g/10 min)	25	28	36
Flexural modulus (MPa)	1347	1326	1363
Tensile strength at break (MPa)	18	17	19
Elongation at break (%a)	501	499	504
IZOD resilience at 23° C (KJ/m^2)	57	57	46
IZOD resilience at -15° C (KJ/m^2)	10	11	5
IZOD resilience at -30° C (KJ/m^2)	5	5	3.7
Longitudinal shrinkage (%)	0.88	0.9	0.63
Transversal shrinkage (%)	1.28	1.31	1.05
Gloss (%)	69	68	77

CLAIMS

1. A polyolefin composition comprising, all percentages being by weight:
   A) from 40 to 75% of a polypropylene component comprising a propylene homopolymer or a propylene copolymer with another α-olefin or combinations thereof, said polypropylene component containing at least 85% by weight of propylene and having a MFRL value equal to or higher than 90 g/10 min. and a solubility in xylene at room temperature lower than 20% by weight;
   B) from 3 to 20% of a copolymer of ethylene and one or more C_4-C_{10} α-olefin(s), containing 15-35% by weight of C_4-C_{10} α-olefin(s) and having solubility in xylene at room temperature greater than 50% by weight, the intrinsic viscosity of the xylene soluble fraction being from 2.5 to 4 dl/g;
   C) from 10 to 35% of one or more elastomeric or plastomeric polymers different from B), having a Shore A hardness value equal to or lower than 90 points;
   D) from 5 to 25%, preferably from 6 to 20%, of a mineral filler, preferably talc; and, optionally
   E) from 0.5 to 5% of a copolymer, different from C), of ethylene with propylene optionally containing 0.5 to 5% by weight of a diene, said copolymer having an ethylene content from 20 to 70% by weight and a solubility in xylene at room temperature greater than 50% by weight, the intrinsic viscosity of the xylene soluble fraction being from 4 to 9 dl/g;

   wherein the amounts of A), B), C) and E) are referred to the total weight of A) + B) + C) + D) and optionally E).
2. The polyolefin composition of claim 1, further comprising 0.01 to 0.5 parts by weight of a nucleating agent F) with respect to 100 parts by weight of A) + B) + C) + D) and optionally E).
3. The polyolefin composition of claim 1, having a MFRL value equal to or higher than 10 g/10 min., preferably equal to or higher than 15 g/10 min., more preferably equal to or higher than 20 g/10 min.
4. The polyolefin composition of claim 1, wherein the amount of fraction (XI) insoluble in xylene at room temperature of component B) satisfies the following equation:

   $(XI) < 1.14 \times B_2 - 34$

   where B_2 is the amount of ethylene in component B), expressed as percent by weight with respect to the weight of B).

5. A process for preparing the polyolefin composition of claim 1 by subjecting to melt-blending with the other polyolefin components a masterbatch composition comprising, all percentages being by weight:
   - A^I) from 60 to 85% of a polypropylene component comprising a propylene homopolymer or a propylene copolymer with another α-olefin or combinations thereof, said polypropylene component containing at least 85% by weight of propylene, and having a MFRL value equal to or higher than 20 g/10 min. and a solubility in xylene at room temperature lower than 20% by weight;
   - B^I) from 15 to 40% of a copolymer of ethylene and one or more C_4-C_{10} α-olefin(s), containing 15-35% by weight of C_4-C_{10} α-olefin(s) and having solubility in xylene at room temperature greater than 50% by weight, the intrinsic viscosity of the xylene soluble fraction being from 2.5 to 4 dl/g.
6. The process of claim 5, wherein the masterbatch composition has a MFRL value equal to or higher than 2 g/10 min.
7. The process of claim 5, wherein the masterbatch composition has a ΔHm of the DSC melting peak detectable at a temperature between 100 and 130°C of 1 J/g or more.
8. The process of claim 5, wherein the masterbatch composition has is obtainable by polymerization in the presence of a $MgCl_2$ supported Ziegler-Natta catalyst.
9. Manufactured articles comprising the polyolefin composition of claim 1.
10. The manufactured articles of claim 9 in form of door trims.

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2010/069021

A. CLASSIFICATION OF SUBJECT MATTER
INV. C08L23/12
ADD.

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
C08L

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

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

EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO 2005/121240 A1 (BASELL POLIOLEFINE SRL [IT]; PELLICONI ANTEO [IT]; GARAGNANI ENEA [IT]) 22 December 2005 (2005-12-22) cited in the application page 13; examples 1,2 claims 1,4 -----	1-10
A	WO 02/28958 A2 (BASELL TECHNOLOGY CO BV [NL]) 11 April 2002 (2002-04-11) the whole document -----	1-10
A	WO 2007/060114 A1 (BASELL POLIOLEFINE SRL [IT]; MASSARI PAOLA [IT]; NEWS JEAN [IT]; CIARA) 31 May 2007 (2007-05-31) the whole document -----	1-10
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☒ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

* Special categories of cited documents :

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

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

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

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

"&" document member of the same patent family

Date of the actual completion of the international search 4 April 2011	Date of mailing of the international search report 11/04/2011
Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Authorized officer Lippert, Sigrid

International application No
PCT/EP2010/069021

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

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A	EP 2 031 018 A1 (SUMITOMO CHEMICAL CO [JP]; TOYOTA MOTOR CO LTD [JP]) 4 March 2009 (2009-03-04) the whole document -----	1-10
A	US 2002/035191 A1 (HIRAKAWA MANABU [JP] ET AL) 21 March 2002 (2002-03-21) the whole document -----	1-10

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Information on patent family members

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