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(54) Title: BLEND OF POST-CONSUMER RECYCLED (PCR) POLYMER AND MODIFIER

(57) Abstract: The present invention relates to a polymer blend with excellent mechanical properties, comprising a post-consumer recycled polyolefin composition and a heterophasic polypropylene composition. Further, the present invention also relates to a preparation process for the polymer blend comprising the preparation of the heterophasic polypropylene composition in the presence of a Ziegler-Natta catalyst. Moreover, the present invention relates to articles comprising the polymer blend as well as to the use of the polymer blend for the manufacture of articles.

Blend of post-consumer recycled (PCR) polymer and modifier

The present invention relates to a polymer blend with excellent mechanical properties, comprising a post-consumer recycled polyolefin composition and a heterophasic polypropylene composition. Further, the present invention also relates to a preparation process for the polymer blend comprising the preparation of the heterophasic polypropylene composition in the presence of a Ziegler-Natta catalyst. Moreover, the present invention relates to articles comprising the polymer blend as well as to the use of the polymer blend for the manufacture of articles.

Background

Plastic packaging is widely used in daily life due to its good cost/performance balance. Polyolefin polymers based on polypropylene and/or polyethylene are often used in these applications.

However, accumulation of plastic waste has become an environmental problem that must be addressed by the polymer producer and users. Therefore, recycling of plastic material is on-going, where plastic waste can be turned into resources for new plastic products. Hence, environmental and economic aspects can be combined in recycling and reusing plastic material.

Although recycling of plastic material has already begun in the mid-1990s by implementing collection systems, which allow more target oriented collection and separation of plastic materials from other household waste materials, the reuse of plastic material originating from plastic waste is still limited. The so-called post-consumer recycled (PCR) plastic material generally contains mixtures of different plastics and several contaminant materials. Often the post-consumer recycled (PCR) polymers do not have the necessary properties to be used in a wide range of applications.

Accordingly, efforts have been undertaken to modify the properties of post-consumer recycled (PCR) polymers by addition of modifier polymers. For example, international patent application WO-A-2020/070175 suggests blending post-consumer and/or post-industrial recyclates with a heterophasic random copolymer.

However, there is still high need for further modifier polymers for post-consumer recycled (PCR) polymers in order to provide recycled polymers with selectively improved properties, and to allow a broader application of used plastic material in a second life cycle.

Object of the invention

It is thus an object of the present invention to provide new modifiers for improving the properties of post-consumer recycled (PCR) polymers, and respectively to provide new polymer blends of post-consumer recycled (PCR) polymers with these modifiers.

In particular, it is desired that the polymer blends have sufficiently good mechanical properties to be used in a variety of applications.

Summary of the invention

It has been surprisingly found that the above-described object can be achieved by a polymer blend comprising:

a) 60 to 95 wt.-%, based on the total weight of the polymer blend, of a post-consumer recycled polyolefin composition having a melt flow rate MFR_2 (230 °C), determined according to ISO 1133, in the range of from 1.0 to 100.0 g/10 min, and comprising propylene (co)polymer component(s) and ethylene (co)polymer component(s) in a ratio (weight/weight) of from 1:99 to 99:1, determined by FTIR spectroscopy, and

b) 5 to 40 wt.-%, based on the total weight of the polymer blend, of a heterophasic polypropylene composition comprising a propylene copolymer (A) matrix of propylene with ethylene and 1-butene comonomer units and a propylene ethylene elastomer (B) dispersed within the matrix, the heterophasic polypropylene composition comprising

b1) 55 to 85 wt.-%, based on the total weight of the heterophasic polypropylene composition and determined at 25 °C according to ISO 16152, of a xylene insoluble fraction (XI) comprising propylene comonomer units and, based on the total weight of the xylene insoluble fraction (XI), ethylene comonomer units in a content in the range of from 0.5 to 3.0 wt.-% and 1-butene comonomer units in a content in the range of from 5.0 to 10.0 wt.-%, and

b2) 15 to 45 wt.-%, based on the total weight of the heterophasic polypropylene composition and determined at 25 °C according to ISO 16152, of a xylene soluble fraction (XS) comprising propylene comonomer units and, based on the total weight of the xylene soluble fraction (XS), ethylene comonomer units in a content in the range of from 18.0 to 50.0 wt.-% and 1-butene comonomer units in a content in the range of from 1.0 to 10.0 wt.-%, and the xylene soluble fraction

(XS) having an intrinsic viscosity IV, determined according to ISO 1628-1 & 3, in the range of from 1.2 to 6.0 dl/g.

Further, it has been found that the above-described object is achieved by the use of the respective heterophasic polypropylene composition as a modifier for the post-consumer recycled polyolefin composition. Thus, mechanical properties of a post-consumer recycled polyolefin composition can be improved by the use of the heterophasic polypropylene composition as described above.

Accordingly, based on the particular selection of the comonomers and their contents in the xylene insoluble fraction (XI) and the xylene soluble fraction (XS), the heterophasic polypropylene composition can improve the mechanical properties of the post-consumer recycled polyolefin composition (in particular, the impact strength, elongation properties and processability). Thus, the polymer blends according to the present invention are characterized by respectively improved mechanical properties. Other mechanical properties (such as tensile modulus and tensile strength) are maintained in a satisfying range, such that application of the polymer blends in different technical fields is possible.

Still further, it has been found that the above-described object is achieved by a process for the preparation of the polymer blend, wherein the process comprises

A) providing the post-consumer recycled polyolefin composition in any one of the embodiments described herein,

B) providing the heterophasic polypropylene composition in any one of the embodiments described herein by preparing the heterophasic polypropylene composition in a multi-stage polymerization process in the presence of a Ziegler-Natta catalyst, and

C) blending the post-consumer recycled polyolefin composition and the heterophasic polypropylene composition, preferably by melting and mixing the components.

Articles, preferably injection molded or blow molded articles and films, comprising the polymer blend, as well as use of the polymer blend for the manufacture of the respective articles allow to provide articles with improved mechanical properties.

Description of the invention

1. Polymer blend

The present invention relates to a polymer blend comprising a post-consumer recycled polyolefin composition and a heterophasic polypropylene composition.

The heterophasic polypropylene composition is used as a modifier for the post-consumer recycled polyolefin composition to improve its properties and to make it better processable and usable in a broader range of applications.

The term '*polymer blend*' as used herein refers to a mixture of two or more polymeric components. In general, the blend can be prepared by mixing the two or more polymeric components. Suitable mixing procedures known in the art are post-polymerization blending, as e.g. exemplified herein below.

The polymer blend comprises from 60 to 95 wt.-%, preferably from 70 to 93 wt.-% and more preferably from 80 to 92 wt.-%, based on the total weight of the polymer blend, of the post-consumer recycled polyolefin composition.

The polymer blend comprises from 5 to 40 wt.-%, preferably from 7 to 30 wt.-% and more preferably from 8 to 20 wt.-%, based on the total weight of the polymer blend, of the heterophasic polypropylene composition.

Preferably, the post-consumer recycled polyolefin composition and the heterophasic polypropylene composition are the only polymeric components added to prepare the polymer blend. However, it is to be understood herein that additives (e.g. pigments or dyes, stabilizers, anti-acids and/or anti-UVs, antistatic agents, nucleating agents and utilization agents), organic fillers, and/or inorganic fillers (e.g. ash, talc, glass fibers or wood fibers) and/or other components may be further contained in the polymer blend. Generally, the content of these components amounts to less than 10 wt.-%, based on the total weight of the polymer blend. These components may be present in the polymer blend due to their presence in the post-consumer recycled polyolefin composition and/or the heterophasic polypropylene composition and/or they may be added to the blend (before and/or during and/or after the blending process). Additives are generally present in an amount of up to 5 wt.-%, based on the total weight of the polymer blend. In particular, other polyolefins and non-polyolefins may be present in the polymer blend due to their presence in the post-consumer recycled polyolefin composition.

The polymer blend is characterized by excellent mechanical properties in terms of impact strength, tensile modulus and tensile strength and elongation. It may be used in a variety of applications where these properties are important.

Preferably, the polymer blend has an impact strength at +23 °C, determined according to ISO 179/1eA on an injection molded specimen, in the range of from 7.0 to 35.0 kJ/m^2, more preferably from 7.5 to 33.0 kJ/m^2 and most preferably from 8.0 to 30.0 kJ/m^2.

Preferably, the polymer blend has an impact strength at -20 °C, determined according to ISO 179/1eA on an injection molded specimen, in the range of from 2.0 to 12.0 kJ/m^2, more preferably from 2.1 to 11.0 kJ/m^2 and most preferably from 2.2 to 10.0 kJ/m^2.

Preferably, the polymer blend has a tensile modulus, determined according to ISO 527-2 on an injection molded specimen, in the range of from 500 to 1200 MPa, more preferably from 550 to 1100 MPa and most preferably from 600 to 1000 MPa.

Preferably, the polymer blend has a tensile strength, determined according to ISO 527-2 on an injection molded specimen, in the range of from 15.0 to 25.0 MPa, more preferably from 16.0 to 23.0 MPa and most preferably from 16.5 to 21.0 MPa.

Preferably, the polymer blend has an elongation at break, determined according to ISO 527-2 on an injection molded specimen, in the range of from 250 to 600 %, more preferably from 260 to 570 % and most preferably from 270 to 550 %.

In one preferred embodiment, the polymer blend is characterized by all of the above-described mechanical properties.

The polymer blend can be easily processed based on its advantageous flowability.

Preferably, the polymer blend has a melt flow rate MFR_2 (230 °C), determined according to ISO 1133, in the range of from 1.5 to 20.0 g/10 min, more preferably from 2.0 to 15.0 g/10 min and most preferably from 2.5 to 12.0 g/10 min.

The polymer blend comprises a post-consumer recycled polyolefin composition that contains propylene (co)polymer component(s) and ethylene (co)polymer component(s). Accordingly, the polymer blend will have melting temperature(s) T_m and crystallization temperature(s) T_c, which arrive from these components of the post-consumer recycled polyolefin composition.

In one embodiment, the polymer blend has at least one melting temperature T_{m1}, determined according to differential scanning calorimetry (DSC) described herein and related to the polyethylene fraction in the polymer blend, wherein the melting temperatures T_{m1} is in the range of from 115 to 137 °C, more preferably from 120 to 133 °C and most preferably from 123 to 130 °C.

In another embodiment, the polymer blend has at least one melting temperature T_{m2}, determined according to differential scanning calorimetry (DSC) described herein and related to the polypropylene fraction in the polymer blend, wherein

the melting temperature T_{m2} is in the range of from 152 to 163 °C, more preferably from 155 to 161 °C and most preferably from 157 to 160 °C.

In a preferred embodiment, the polymer blend has at least two melting temperatures T_{m1} and T_{m2} as described above.

In one further embodiment, the polymer blend has at least one crystallization temperature T_{c1}, determined according to differential scanning calorimetry (DSC) described herein and related to the polypropylene fraction in the polymer blend, wherein the crystallization temperature T_{c1} is in the range of from 110 to 130 °C, more preferably from 115 to 126 °C and most preferably from 118 to 123 °C.

In one further embodiment, the polymer blend has at least one crystallization temperature T_{c2}, determined according to differential scanning calorimetry (DSC) described herein and related to the polyethylene fraction in the polymer blend, wherein the crystallization temperature T_{c2} is in the range of from 105 to 122 °C, more preferably from 111 to 119 °C and most preferably from 113 to 117 °C.

In a further preferred embodiment, the polymer blend has at least two crystallization temperatures T_{c1} and T_{c2} as described above.

1.1 Post-consumer recycled polyolefin composition

The term '*post-consumer recycled (PCR) polyolefin composition*' as used herein refers to a composition comprising polyolefins obtained from consumer waste. Thus, post-consumer recycled polyolefins have already completed at least a first use cycle (or life cycle), i.e. having already served their first purpose. Post-consumer recycled polyolefin is different from virgin polyolefin, i.e. a newly produced material, which has not already been recycled. Post-consumer recycled polyolefin is also different from industrial waste, i.e. manufacturing scrap, which does normally not reach a consumer.

Virgin materials and recycled materials can easily be differentiated based on the absence or presence of contaminants such as limonene, fatty acids, paper and/or wood and other contaminants, or generally on their ash content. Polyolefins (e.g. polypropylene-polyethylene blends) can further be differentiated with respect to the materials' origin by the possible presence of non-polyolefin polymers such as polystyrene and/or polyamide.

The post-consumer recycled polyolefin composition as contained in the polymer blend of the present invention may be prepared from pre-compositions based on post-consumer recyclates with other contents or forms. For example, post-consumer polymeric waste may be treated by different mechanical (e.g. sorting) processes, washing or chemical (e.g. dissolving) processes, in order to provide

the post-consumer recycled polyolefin composition to be used in the polymer blend of the present invention.

In particular, the post-consumer recycled polyolefin composition may be provided in the desired size and form by extrusion at 180 to 300 °C, preferably from 200 to 280 °C, in a single- or twin-screw extruder, preferably a co-rotating twin-screw extruder, followed by a suitable pelletization process, in which the pellet thickness and diameter are defined. Suitable pelletization processes include underwater pelletization, water-ring pelletization and strand pelletization, the latter comprising solidification of one or more melt strands in a water bath followed by cutting the strand into pellets.

Post-consumer recycled polyolefin compositions as used for the polymer blend according to the present invention are already commercially available, for example under the tradename Dipolen™, such as Dipolen™-S71, available from Borealis.

The post-consumer recycled polyolefin composition has a melt flow rate MFR_2 (230 °C), determined according to ISO 1133, in the range of from 1.0 to 100.0 g/10 min, preferably from 2.0 to 80.0 g/10 min and more preferably from 3.0 to 60.0 g/10 min. In one embodiment, the post-consumer recycled polyolefin composition has a melt flow rate MFR_2 (230 °C) in the range of from 2.0 to 20.0 g/10 min.

The post-consumer recycled polyolefin composition comprises propylene (co)polymer component(s) and ethylene (co)polymer component(s) in a ratio (weight/weight) of from 1:99 to 99:1, preferably from 10:90 to 90:10 and more preferably from 25:75 to 75:25, determined by Fourier transform infrared (FTIR) spectroscopy.

The post-consumer recycled polyolefin composition preferably comprises a mixture (such as a polymer blend) of one or more propylene (co)polymer component(s) and one or more ethylene (co)polymer component(s).

As a direct determination of the propylene (co)polymer and ethylene (co)polymer content or ratio in the post-consumer recycled polyolefin composition is not possible, the weight contents are determined from the equivalent ratio from calibration by isotactic polypropylene (iPP) homopolymer and high density polyethylene (HDPE).

1.1.1 Propylene (co)polymer component

The term '*propylene (co)polymer component*' as used herein refers to a propylene homopolymer component and a propylene copolymer component as well as combinations thereof, e.g. copolymers or blends.

The term '*propylene homopolymer*' denotes a polymer that consists of at least 99.0 wt.-%, preferably at least 99.5 wt.-% and more preferably at least 99.8 wt.-% of propylene monomer units, based on the total weight of the propylene homopolymer.

The term '*propylene copolymer*' as used herein denotes a polymer comprising, based on the total weight of the propylene copolymer, at least 50 wt.-% of propylene monomer units and further comprising alpha-olefin comonomer units different from propylene. Preferably, the content of the propylene monomer units in the propylene copolymer is at least 70 wt.-%, based on the total weight of the propylene copolymer.

The term '*alpha-olefin'* units (e.g., propylene, ethylene, 1-butene units etc.) used herein for contents of the polymer is to be understood to teach these units for preparing the polymer. The resultant polymer contains the respective units derived from these alpha-olefin monomer units. Usually, the term '*alpha-olefin'* refers to C_2 to C_{10} alpha-olefins.

The contents of comonomer units in the homo- and copolymers are generally determined by ^{13}C-NMR spectroscopy.

Unless otherwise indicated, percentages throughout this disclosure are percentages by weight (wt.-%) based on the total weight of the respectively described enclosing entity.

1.1.2 Ethylene (co)polymer component

The term '*ethylene (co)polymer component*' as used herein refers to an ethylene homopolymer component and an ethylene copolymer component as well as combinations thereof, e.g. copolymers or blends.

The term '*ethylene homopolymer*' denotes an ethylene polymer that consists of at least 99.0 wt.-%, preferably at least 99.5 wt.-% and more preferably at least 99.8 wt.-% of ethylene monomer units, based on the total weight of the ethylene homopolymer.

The term '*ethylene copolymer*' as used herein denotes a polymer comprising, based on to the total weight of the ethylene copolymer, at least 50 wt.-% of ethylene monomer units and further comprising alpha-olefin comonomer units different from ethylene. Preferably, the content of the ethylene monomer units in

the ethylene copolymer is at least 70 wt.-%, based on the total weight of the ethylene copolymer.

1.1.3 Further components

Further, in some embodiments, the post-consumer recycled polyolefin composition comprises, based on the total weight of the post-consumer recycled polyolefin composition, one or more of

a) from 0.1 to 100.0 ppm, preferably from 0.1 to 50.0 ppm, more preferably from 0.1 to 40.0 ppm, of limonene, determined by solid phase microextraction (HS-SPME-GC-MS);

b) from 0.1 to 100.0 ppm, preferably from 0.1 to 90.0 ppm, more preferably from 0.1 to 80.0 ppm, of fatty acids, determined by solid phase microextraction (HS-SPME-GC-MS);

c) from 0.1 to 10.0 wt.-%, preferably from 0.1 to 6.0 wt.-%, more preferably from 0.1 to 5.0 wt.-%, of non-polyolefin polymers; in particular, from 0.1 to 3.0 wt.-% of polystyrene and/or from 0.1 to 3.0 wt.-% of polyamide, determined by FTIR spectroscopy;

d) from 0.1 to 10.0 wt.-%, preferably from 0.1 to 5.0 wt.-%, more preferably from 0.1 to 3.0 wt.-%, of other components, selected from talc, chalk, carbon, calcium stearate, titanium dioxide, pigments, metals, glass, paper, wood, and combinations thereof, determined as described below.

In a preferred embodiment, the post-consumer recycled polyolefin composition comprises at least limonene in a content of at least 0.1 ppm. In another preferred embodiment the post-consumer recycled polyolefin composition comprises at least limonene in a content of from 0.1 to 5.0 ppm.

The post-consumer recycled polyolefin composition may also comprise a residual ash content, as determined according to the ISO 3451-1 (1997) standard, of below 3.0 wt.-%, preferably in the range of from 0.5 to 2.7 wt.-% and more preferably from 0.7 to 2.5 wt.-%.

Based on the content of the post-consumer recycled polyolefin composition in the polymer blend, the polymer blend will generally comprise the above identified further components in a content of 60 to 95 % of the above specified contents. For example, the polymer blend preferably comprises at least 0.060 to 0.095 ppm of limonene, based on the total weight of the polymer blend.

1.2 Heterophasic polypropylene composition

The heterophasic polypropylene composition comprises or consists of a propylene copolymer (A) matrix of propylene with ethylene and 1-butene comonomer units and a propylene ethylene elastomer (B) dispersed within the matrix.

The term '*heterophasic polypropylene composition*' as used herein means that at least two distinct phases are present in the composition, i.e., a propylene copolymer (A) and a propylene ethylene elastomer (B). The propylene copolymer (A) forms a crystalline matrix of the heterophasic polypropylene composition and the propylene ethylene elastomer (B) is dispersed in the crystalline matrix. Presence of the distinct phases is easily detectable via DSC analysis: the crystalline matrix propylene copolymer (A) will show a melting point higher than that of the propylene ethylene elastomer (B).

The terms '*propylene copolymer*' and '*alpha-olefin'* units are to be understood as defined above.

The heterophasic polypropylene composition comprises from 55 to 85 wt.-%, preferably from 60 to 83 wt.-% and more preferably from 65 to 80 wt.-%, based on the total weight of the heterophasic polypropylene composition and determined at 25 °C according to ISO 16152, of a xylene insoluble fraction (XI). In one embodiment, the heterophasic polypropylene composition comprises from 65 to 85 wt.-% of a xylene insoluble fraction (XI).

The xylene insoluble fraction (XI) comprises propylene comonomer units and, based on the total weight of the xylene insoluble fraction (XI), ethylene comonomer units in a content in the range of from 0.5 to 3.0 wt.-%, preferably from 0.5 to 2.5 wt.-% and more preferably from 0.5 to 2.0 wt.-%, and 1-butene comonomer units in a content in the range of from 5.0 to 10.0 wt.-%, preferably from 5.5 to 9.0 wt.-% and more preferably from 6.0 to 8.5 wt.-%.

Generally, the xylene insoluble fraction (XI) comprises a higher content of the propylene copolymer (A). Preferably, the xylene insoluble fraction (XI) comprises the propylene copolymer (A) in a content of at least 70 wt.-%, more preferably at least 80 wt.-% and up to 100 wt.-% of, based on the total weight of the xylene insoluble fraction (XI).

The heterophasic polypropylene composition comprises from 15 to 45 wt.-%, preferably from 17 to 40 wt.-% and more preferably from 20 to 35 wt.-%, based on the total weight of the heterophasic polypropylene composition and determined at 25 °C according to ISO 16152, of a xylene soluble fraction (XS).

In one embodiment, the heterophasic polypropylene composition comprises from 15 to 35 wt.-% of a xylene soluble fraction (XS).

Preferably, the xylene insoluble fraction (XI) and the xylene soluble fraction (XS) add up to 100 wt.-% of the propylene polymer content of the heterophasic polypropylene composition. More preferably, they add up to 100 wt.-% of the entire polymer content of the heterophasic polypropylene composition. However, it is to be understood that additives may additionally be present in the heterophasic polypropylene composition as defined below.

Thus, preferably, the heterophasic polypropylene composition consists of the xylene insoluble fraction (XI) and the xylene soluble fraction (XS) and optional additives.

The xylene soluble fraction (XS) comprises propylene comonomer units and, based on the total weight of the xylene soluble fraction (XS), ethylene comonomer units in a content in the range of from 18.0 to 50.0 wt.-%, preferably from 20.0 to 40.0 wt.-% and more preferably from 22.0 to 30.0 wt.-%, and 1-butene comonomer units in a content in the range of from 1.0 to 10.0 wt.-%, preferably from 1.5 to 8.0 wt.-% and more preferably from 2.0 to 6.0 wt.-%. In one embodiment, the xylene soluble fraction (XS) comprises propylene comonomer units, ethylene comonomer units in a content in the range of from 20.0 to 50.0 wt.-% and 1-butene comonomer units in a content in the range of from 1.0 to 10.0 wt.-%.

Preferably, the xylene soluble fraction (XS) comprises, based on the total weight of the xylene soluble fraction (XS), a total content of comonomer units of 1-butene and ethylene in the range of from 19.0 to 60.0 wt.-%, more preferably from 21.5 to 48.0 wt.-% and most preferably from 24.0 to 36.0 wt.-%.

Generally, the xylene soluble fraction (XS) comprises the propylene ethylene elastomer (B). Preferably, the xylene soluble fraction (XS) comprises the propylene ethylene elastomer (B) in a content of at least 50 wt.-%, more preferably at least 60 wt.-% and up to 100 wt.-% of, based on the total weight of the xylene soluble fraction (XS).

The xylene soluble fraction (XS) has an intrinsic viscosity IV, determined according to ISO 1628-1 & 3, in the range of from 1.2 to 6.0 dl/g, preferably from 1.2 to 5.0 dl/g, more preferably from 1.3 to 4.0 dl/g and most preferably from 1.3 to 3.0 dl/g.

Preferably, the heterophasic polypropylene composition has a melt flow rate MFR_2 (230 °C), determined according to ISO 1133, in the range of from 0.5 to

5.0 g/10 min, preferably 0.7 to 4.0 g/10 min and more preferably from 1.0 to 3.5 g/10 min.

Preferably, the heterophasic polypropylene composition has a melting temperature T_m, determined according to differential scanning calorimetry (DSC) described herein, in the range of from 125 to 145 °C, more preferably from 128 to 143 °C and most preferably from 130 to 140 °C.

Preferably, the heterophasic polypropylene composition has a crystallization temperature T_c, determined according to differential scanning calorimetry (DSC) described herein, in the range of from 90 to 100 °C, more preferably from 91 to 99 °C and most preferably from 92 to 98 °C.

Preferably, the heterophasic polypropylene composition comprises, based on the total weight of the heterophasic polypropylene composition and determined by ^{13}C-NMR spectroscopy, a content of 1-butene comonomer units in the range of from 3.0 to 14.0 wt.-%, more preferably from 4.0 to 12.0 wt.-% and most preferably from 5.0 to 10.0 wt.-%.

Preferably, the heterophasic polypropylene composition comprises, based on the total weight of the heterophasic polypropylene composition and determined by ^{13}C-NMR spectroscopy, a content of ethylene comonomer units in the range of from 3.0 to 15.0 wt.-%, more preferably from 4.0 to 13.0 wt.-% and most preferably from 5.0 to 11.5 wt.-%.

Preferably, propylene, 1-butene and ethylene are the only comonomers present in the heterophasic polypropylene composition. It is preferred that the heterophasic polypropylene composition is substantially free of units derived from other alpha-olefin monomers (i.e., C_5 to C_8 alpha-olefin monomers), e.g., it comprises less than 0.10 wt.-%, preferably less than 0.05 wt.-%, and more preferably less than 0.01 wt.-% of other alpha-olefin monomers such as C_5 to C_8 alpha-olefin monomer units.

The comonomer contents in the heterophasic polypropylene composition, the xylene soluble fraction (XS) and xylene insoluble fraction (XI) and in the propylene copolymers are determined by ^{13}C-NMR spectroscopy, as described in the method section.

The heterophasic polypropylene composition is preferably obtained or obtainable by a polymerization process using a Ziegler-Natta catalyst. Preferably, the heterophasic polypropylene composition is produced using a Ziegler-Natta catalyst according to the preferred embodiments as described with respect to the preparation of the heterophasic polypropylene composition herein.

Preferably, the heterophasic polypropylene composition is free of phthalic acid esters and their respective decomposition products.

The heterophasic polypropylene composition is characterized by good mechanical properties.

Preferably, the heterophasic polypropylene composition has a flexural modulus, determined according to ISO 178, in the range of from 250 to 500 MPa, more preferably from 260 to 470 MPa and most preferably from 270 to 450 MPa.

Preferably, the heterophasic polypropylene composition has an impact strength at +23 °C, determined according to ISO 179/1eA, in the range of from 30.0 to 100.0 kJ/m^2, more preferably from 35.0 to 97.0 kJ/m^2 and most preferably from 40.0 to 95.0 kJ/m^2.

Further preferably the heterophasic polypropylene composition has an impact strength at -20 °C, determined according to ISO 179/1eA, in the range of from 1.5 to 7.0 kJ/m^2, more preferably from 1.6 to 6.5 kJ/m^2 and most preferably from 1.7 to 6.0 kJ/m^2.

Preferably, the propylene copolymer (A) and the propylene ethylene elastomer (B) are the only propylene polymer components in the heterophasic polypropylene composition.

The weight ratio of the propylene ethylene elastomer (B) to the propylene copolymer (A) in the heterophasic polypropylene composition is preferably in the range of from 5:95 to 40:60, more preferably from 7:93 to 35:65 and most preferably from 10:90 to 30:70.

It is to be understood herein that the heterophasic polypropylene composition may comprise further components such as additives, which may optionally be added in a mixture with or without a carrier polymer (e.g., in a master batch).

Suitable additives include fillers, lubricants, processing aids, antioxidants, UV absorbers, light stabilizers, nucleating agents, foaming or blowing agents, clarifiers and pigments.

The additives may be present in a content in the range of from 0.1 to 10.0 wt.-%, preferably from 0.3 to 5.0 wt.-% and more preferably from 0.5 to 3.0 wt.-%, based on the total weight of the heterophasic polypropylene composition.

1.2.1 Propylene copolymer (A)

The propylene copolymer (A) forms the crystalline matrix of the heterophasic polypropylene composition. The propylene copolymer (A) is a copolymer of propylene with 1-butene comonomer units and ethylene comonomer units.

Preferably, the propylene copolymer (A) has, based on the total weight of the propylene copolymer (A), a content of 1-butene comonomer units in the range of from 4.0 to 15.0 wt.-%, more preferably from 4.5 to 12.0 wt.-% and most preferably from 5.0 to 10.0 wt.-%, and a content of ethylene comonomer units in the range of from 0.5 to 2.5 wt.-%, more preferably from 0.7 to 2.4 wt.-% and most preferably from 1.0 to 2.3 wt.-%.

The propylene copolymer (A) is preferably a multimodal polymer. The term '*multimodal polymer*' as used herein denotes a polymer composition comprising at least two polymer fractions, which have been produced under different polymerization conditions resulting in different (weight average) molecular weights and/or molecular weight distributions for the fractions and/or different comonomer contents. The prefix '*multi*' relates to the number of different polymer fractions the polymer consists of. The term '*multimodal polymer*' comprises bimodal, trimodal, tetramodal etc. polymers.

More preferably, the propylene copolymer (A) is a bimodal polymer, i.e., it consists of two different polymer fractions, i.e., a first propylene polymer fraction (A1) and a second propylene polymer fraction (A2).

Preferably, the first propylene polymer fraction (A1) and the second propylene polymer fraction (A2) are prepared in subsequent polymerization stages.

Preferably, the propylene copolymer (A) comprises, or consists of, from 30 to 70 wt.-%, based on the total weight of the propylene copolymer (A), of a first propylene polymer fraction (A1) being a propylene copolymer of propylene with ethylene comonomer units and 1-butene comonomer units, and from 30 to 70 wt.-%, based on the total weight of the propylene copolymer (A), of a second propylene polymer fraction (A2), being a propylene copolymer of propylene with ethylene comonomer units and 1-butene comonomer units.

In other words, the weight ratio of the first propylene polymer fraction (A1) to the second propylene polymer fraction (A2) in the propylene copolymer (A) is preferably in the range of from 30:70 to 70:30, more preferably from 35:65 to 65:35 and most preferably from 40:60 to 60:40.

The second propylene polymer fraction (A2) is different from the first propylene polymer fraction (A1). Preferably, the second propylene polymer fraction (A2) has a higher total content of comonomers than the first propylene polymer fraction (A1). More preferably, the second propylene polymer fraction (A2) has a higher content of ethylene comonomer units and 1-butene comonomer units than the first propylene polymer fraction (A1).

Preferably, the propylene copolymer (A) has a molecular weight distribution M_w/M_n, being the ratio of the weight average molecular weight M_w and the number average molecular weight M_n, in the range of from 3.0 to 10.0, more preferably from 4.0 to 8.0 and most preferably 5.5 to 7.0.

The propylene copolymer (A) preferably has a melt flow rate MFR_2 (230 °C), determined according to ISO 1133, in the range of from 0.3 to 6.0 g/10 min, more preferably from 0.5 to 4.5 g/10 min and most preferably from 0.7 to 3.0 g/10 min.

1.2.1.1 First propylene polymer fraction (A1)

The first propylene polymer fraction (A1) of the propylene copolymer (A) preferably consists of a single propylene polymer.

The propylene polymer of the first propylene polymer fraction (A1) preferably is a propylene copolymer of propylene with 1-butene comonomer units and ethylene comonomer units.

Preferably, the first propylene polymer fraction (A1), based on the total weight of the first propylene polymer fraction (A1), has a content of 1-butene comonomer units in the range of from 2.0 to 10.0 wt.-%, more preferably from 3.0 to 8.0 wt.-% and most preferably from 3.5 to 7.5 wt.-%, and a content of ethylene comonomer units in the range of from 0.1 to 2.0 wt.-%, more preferably from 0.5 to 1.8 wt.-% and most preferably from 0.7 to 1.5 wt.-%.

The first propylene polymer fraction (A1) preferably has a melt flow rate MFR_2 (230 °C), determined according to ISO 1133, in the range of from 0.2 to 5.0 g/10 min, more preferably from 0.4 to 3.5 g/10 min and most preferably from 0.5 to 3.0 g/10 min.

1.2.1.2 Second propylene polymer fraction (A2)

The second propylene polymer fraction (A2) of the propylene copolymer (A) preferably consists of a single propylene polymer.

The propylene polymer of the second propylene polymer fraction (A2) preferably is a propylene copolymer of propylene with 1-butene comonomer units and ethylene comonomer units.

The second propylene polymer fraction (A2) preferably has a melt flow rate MFR_2 (230 °C), determined according to ISO 1133, in the range of from 0.1 to 10.0 g/10 min, more preferably from 0.3 to 5.0 g/10 min and most preferably from 0.5 to 3.0 g/10 min.

The MFR_2 of the second propylene polymer fraction (A2), produced in the second polymerization stage is determined according to equation (1):

$$\log(MFR\ (A2)) = \frac{\log(MFR\ (A)) - w(A1) * \log(MFR(A1))}{w(A2)}$$ equation (1)

wherein

MFR(A) is the MFR_2 of the propylene copolymer (A)

w(A1) and *w(A2)* are the weight fractions of the propylene polymer fractions (A1) and (A2) in the propylene copolymer (A)

MFR(A1) is the MFR_2 of the propylene polymer fraction (A1) produced in the first step.

Respectively adapted equations may be used to determine the parameters of the second propylene polymer fraction (A2) and of polymers prepared in further polymerization stages of a preparation process.

1.2.2 Propylene ethylene elastomer (B)

The heterophasic polypropylene composition further comprises a propylene ethylene elastomer (B). The propylene ethylene elastomer (B) forms the dispersed phase that is dispersed in the crystalline matrix.

The propylene ethylene elastomer (B) is a copolymer of propylene and ethylene. Preferably, the ethylene content in the propylene ethylene elastomer (B), based on the total weight of the propylene ethylene elastomer (B), is in the range of from 20 to 60 wt.-%, more preferably from 22 to 50 wt.-% and most preferably from 25 to 45 wt.-%. The ethylene content is determined by ^{13}C-NMR spectroscopy, as described in the method section.

The propylene ethylene elastomer (B) may comprise 1-butene comonomer units. The content of 1-butene comonomer units in the propylene ethylene elastomer (B) is preferably in the range of from 0 to 5.0 wt.-%. Preferably, the propylene ethylene elastomer (B) does not contain 1-butene comonomer units.

Preferably, propylene and ethylene and optionally 1-butene are the only monomers present in the propylene ethylene elastomer (B). It is preferred that propylene ethylene elastomer (B) is substantially free of units derived from other alpha-olefin monomers (i.e., C_5 to C_8 alpha-olefin monomers), e.g., it comprises less than 0.10 wt.-%, preferably less than 0.05 wt.-% and more preferably less than 0.01 wt.-% of other alpha-olefin monomers such as C_5 to C_8 alpha-olefin monomer units.

Preferably, the propylene ethylene elastomer (B) has a molecular weight distribution M_w/M_n, being the ratio of the weight average molecular weight M_w and the number average molecular weight M_n, in the range of from 3.0 to 10.0, more preferably from 4.0 to 8.0 and most preferably 5.5 to 7.0.

2. Preparation of the polymer blend

Generally, the polymer blend according to the present invention may be prepared by any known process in the art for mixing polymeric components.

The present invention also relates to a process for the preparation of the polymer blend according to any one of the embodiments described herein, wherein the process comprises

A) providing the post-consumer recycled polyolefin composition in any of the embodiments described above,

B) providing the heterophasic polypropylene composition in any of the embodiments described above by preparing the heterophasic polypropylene composition in a multi-stage polymerization process in the presence of a Ziegler-Natta catalyst, and

C) blending the post-consumer recycled polyolefin composition and the heterophasic polypropylene composition, preferably by melting and mixing the components.

Suitable mixing procedures known in the art are post-polymerization blending.

Post-polymerization blending can be dry blending of polymeric components such as polymer powders and/or compounded polymer pellets or melt blending by melt mixing the polymeric components.

The polymer blend according to the present invention can be prepared by mechanically blending the components using techniques known in the art for the preparation of polyolefin blends. For example, one can use Banbury, Buss, or Brabender mixers, single-screw or twin-screw extruders.

In step A) of the process, the post-consumer recycled polyolefin composition may be prepared from consumer waste as described above or may be commercially obtained.

In step B) of the process, the heterophasic polypropylene composition may be prepared by any known multi-stage polymerization process in the presence of a Ziegler-Natta catalyst in the art. Ideally, a process is used which produces a homogenous mixture of the various components. Typically, compounding is employed. Compounding usually involves mixing or/and blending the various components in a molten state, often by extrusion. Preferred embodiments of the preparation of the heterophasic polypropylene composition are described below.

2.1 Preparation of the heterophasic polypropylene composition

In a preferred embodiment, the heterophasic polypropylene composition in any of the above-described embodiments is prepared by a multi-stage polymerization process, comprising

I.) preparing a first propylene polymer fraction (A1) being a propylene copolymer of propylene with ethylene comonomer units and 1-butene comonomer units in a bulk phase reactor, preferably a loop reactor, in a first polymerization stage in the presence of a Ziegler-Natta catalyst;

II.) transferring the first propylene polymer fraction (A1) to a second polymerization stage and preparing a second propylene polymer fraction (A2) being a propylene copolymer of propylene with ethylene comonomer units and 1-butene comonomer units in the presence of a Ziegler-Natta catalyst in a first gas phase reactor (GPR1); and

wherein the first propylene polymer fraction (A1) and the second propylene polymer fraction (A2) together form a propylene copolymer (A); and

III.) transferring the propylene copolymer (A) to a third polymerization stage and preparing a propylene ethylene elastomer (B) as a third propylene polymer fraction in the presence of a Ziegler-Natta catalyst in a second gas phase reactor (GPR2), to obtain a heterophasic polypropylene composition.

Accordingly, in step I.) of the process, propylene, 1-butene and ethylene are fed into the bulk phase reactor to prepare the first propylene polymer fraction (A1).

In step II.) of the process, propylene, 1-butene and ethylene are fed into the first gas phase reactor (GPR1), where the first propylene polymer fraction (A1) has been previously placed, to prepare the second propylene polymer fraction (A2). The two propylene polymer fractions (A1) and (A2) form the propylene copolymer (A).

The propylene copolymer (A) is placed in a second gas phase reactor (GPR2), and propylene, ethylene and optionally 1-butene are added in step III.) of the process, to prepare the heterophasic polypropylene composition. The process may further comprise blending the heterophasic composition with additives, extruding and/or pelletizing the heterophasic composition.

Generally, polymers produced in a multi-stage process are also designated as '*in-situ*' blends. The resulting end product consists of an intimate mixture of the polymers from the three or more reactors. These three or more polymers may have different molecular weight distribution curves, and/or they may differ in terms of comonomer content or type. The heterophasic polypropylene

composition thus contains a mixture of three or more polymers with differing properties, i.e., it is a multimodal polymer mixture.

The first polymerization stage for producing the first propylene polymer fraction (A1) is preferably a slurry polymerization step. The slurry polymerization usually takes place in a liquid mixture of the employed monomers, preferably without the presence of an inert diluent, typically like a hydrocarbon diluent.

The temperature in the first polymerization stage is typically from 60 to 100 °C, preferably from 60 to 80 °C. An excessively high temperature should be avoided to prevent partial dissolution of the polymer into the liquid phase, the fouling of the reactor and early deactivation of the catalyst. The pressure is typically from 1 to 150 barg, preferably from 40 to 80 barg.

The slurry polymerization may be conducted in any known reactor used for slurry polymerization, such as any bulk phase reactor. Such reactors include a continuous stirred tank reactor and a loop reactor. It is especially preferred to conduct the polymerization in a loop reactor. In such reactors the slurry is circulated with a high velocity along a closed pipe by using a circulation pump. Loop reactors are generally known in the art and examples are given, for instance, in the patent applications US-A-4582816, US-A-3405109, US-A-3324093, EP-A-479186 and US-A-5391654. It is thus preferred to conduct the first polymerization stage as a slurry polymerization in a loop reactor.

The slurry may be withdrawn from the reactor either continuously or intermittently. A preferred way of intermittent withdrawal is the use of settling legs where the slurry is allowed to concentrate before withdrawing a batch of the concentrated slurry from the reactor. The use of settling legs is disclosed, among others, in US-A-3374211, US-A-3242150 and EP-A-1310295. Continuous withdrawal is disclosed, among others, in EP-A-891990, EP-A-1415999, EP-A-1591460 and WO-A-2007/025640. The continuous withdrawal is advantageously combined with a suitable concentration method, as disclosed in EP-A-1310295 and EP-A-1591460. It is preferred to withdraw the slurry from the first polymerization stage continuously.

Hydrogen is typically introduced into the first polymerization stage for controlling the MFR_2 of the polymer. The amount of hydrogen needed to reach the desired MFR_2 depends on the catalyst used and the polymerization conditions, as will be appreciated by the skilled worker.

The average residence time in the first polymerization stage is typically from 20 to 120 minutes, preferably from 30 to 80 minutes. As it is well known in the art the average residence time τ can be calculated from equation (2) below:

$$\tau = \frac{V_R}{Q_o} \qquad \text{equation (2)}$$

wherein

V_R is the volume of the reaction space (in case of a loop reactor, the volume of the reactor, in case of the fluidized bed reactor, the volume of the fluidized bed)

Q_o is the volumetric flow rate of the product stream (including the polymer product and the fluid reaction mixture).

The production rate is suitably controlled with the catalyst feed rate. It is also possible to influence the production rate by suitable selection of the monomer concentration. The desired monomer concentration can then be achieved by suitably adjusting the propylene feed rate.

The first propylene polymer fraction (A1) is transferred to the second polymerization stage for producing the second propylene polymer fraction (A2) in a gas phase polymerization step, i.e., carried out in a first gas-phase reactor (GPR1). Any suitable gas phase reactor known in the art may be used, such as a fluidized bed gas phase reactor.

For gas phase reactors, the reaction temperature used will generally be in the range 60 to 115 °C (e.g. 70 to 110 °C), the reactor pressure will generally be in the range 10 to 25 barg, and the residence time will generally be 1 to 8 hours. The gas used will commonly be a non-reactive gas such as nitrogen or low boiling point hydrocarbons such as propane together with monomer.

A chain transfer agent (e.g., hydrogen) is typically added to the second polymerization stage.

In the first gas-phase reactor (GPR1), a mixture of the first propylene polymer fraction (A1) and the second propylene polymer fraction (A2) is formed being the propylene copolymer (A).

The propylene copolymer (A) is transferred to the third polymerization stage for producing the propylene ethylene elastomer (B) in a gas phase polymerization step, i.e., carried out in a second gas-phase reactor (GPR2). Any suitable gas phase reactor known in the art may be used, such as a fluidized bed gas phase reactor. The condition ranges described above for the first gas phase reactor are similarly used for the second gas phase reactor.

A preferred multistage process is the above-identified slurry-gas phase process, such as developed by Borealis and known as the Borstar® technology. In this respect, reference is made to the European patent applications EP-A-0887379 and EP-A-0517868.

The process may comprise one or more further polymerization stage(s)/step(s). Particularly preferred is a prepolymerization step, which precedes the polymerization step I.).

The purpose of the prepolymerization is to polymerize a small amount of polymer onto the catalyst at a low temperature and/or a low monomer concentration. By prepolymerization, it is possible to improve the performance of the catalyst in slurry and/or modify the properties of the final polymer. The prepolymerization step is typically conducted in slurry.

Thus, the prepolymerization step may be conducted in a loop reactor. The prepolymerization is again preferably conducted in liquid propylene, without the presence of an inert diluent, typically like a hydrocarbon diluent.

The temperature in the prepolymerization step is typically from 0 to 60 °C, preferably from 10 to 50 °C and more preferably from 15 to 40 °C.

The pressure is not critical and is typically from 1 to 150 barg, preferably from 40 to 80 barg.

The amount of monomer is typically such that from 0.1 to 1000 grams of monomer per one gram of solid catalyst component is polymerized in the prepolymerization step. As the person skilled in the art knows, the catalyst particles recovered from a continuous prepolymerization reactor do not all contain the same amount of prepolymer. Instead, each particle has its own characteristic amount which depends on the residence time of that particle in the prepolymerization reactor. As some particles remain in the reactor for a relatively long time and some for a relatively short time, then also the amount of prepolymer on different particles is different and some individual particles may contain an amount of prepolymer which is outside the above limits. However, the average amount of prepolymer on the catalyst typically is within the limits specified above.

The molecular weight of the prepolymer may be controlled by hydrogen as it is known in the art. Further, antistatic additives may be used to prevent the particles from adhering to each other or the walls of the reactor, as disclosed in WO-A-96/19503 and WO-A-96/32420.

The catalyst components are preferably all introduced to the prepolymerization step when a prepolymerization step is present. However, where the solid catalyst component and the co-catalyst can be fed separately it is possible that only a part of the co-catalyst is introduced into the prepolymerization stage and the remaining part into subsequent polymerization stages. In such cases it is

necessary to introduce as much co-catalyst into the prepolymerization stage that a sufficient polymerization reaction is obtained therein.

It is understood within the scope of the invention, that the amount of polymer produced in the prepolymerization typically lies within 1.0 to 5.0 wt.-%, based on the total weight of the heterophasic propylene composition.

The heterophasic polymer composition is obtained by a polymerization process using a Ziegler-Natta catalyst. In a preferred embodiment, a Ziegler-Natta catalyst is used in all stages of the polymerization process.

Preferably, the Ziegler-Natta catalyst comprises:

a) compound(s) of a transition metal of Group 4 to 6 of IUPAC;

b) a Group 2 metal compound;

c) an internal donor, wherein said internal donor is a non-phthalic compound, preferably is a non-phthalic acid ester;

d) a co-catalyst; and

e) optionally an external donor.

The internal donor is preferably selected from (di)esters of non-phthalic carboxylic (di)acids, 1,3-diethers, derivatives and mixtures thereof. Most preferred internal donors are (di)esters of citraconic acid.

Preferably, the Ziegler-Natta catalyst does not comprise phthalic acid esters, and the heterophasic polypropylene composition is free of phthalic acid esters and their respective decomposition products.

Suitable Ziegler-Natta catalysts for use in the above-described polymerization reactions and preparation thereof are described in EP-A-3562850.

It will be appreciated that the propylene polymers may contain standard polymer additives as described above.

After preparation, the obtained polymer is typically extruded and pelletized. The extrusion may be conducted in the manner generally known in the art, preferably in a twin screw extruder. One example of suitable twin screw extruders is a co-rotating twin screw extruder. Those are manufactured, among others, by Coperion or Japan Steel Works. Another example is a counter-rotating twin screw extruder. Such extruders are manufactured, among others, by Kobe Steel and Japan Steel Works. Before the extrusion at least part of the desired additives, as mentioned above, are preferably mixed with the polymer. The extruders typically include a melting section where the polymer is melted and a mixing section where the polymer melt is homogenized. Melting and

homogenization are achieved by introducing energy into the polymer. Suitable level of specific energy input (SEI) is from about 150 to about 450 kWh/ton polymer, preferably from 175 to 350 kWh/ton.

3. Article and use

The present invention also relates to an article comprising the polymer blend according to the present invention in any of the above-described embodiments, and to the use of the polymer blend for the manufacture of an article. Preferable articles include molded articles, in particular, injection molded articles, blow molded articles and films.

The articles according to the present invention may be employed in a number of end applications where good mechanical properties are required. In particular, these applications may include non-food packaging, pipes, profiles, containers, crates, automotive components or household articles. The present invention also relates to the use of the blend according to the present invention or the article according to the present invention in these applications, particularly in packaging applications.

As used herein the term '*molded article*' denotes an article that is produced by any conventional molding technique, e.g., injection molding, blow molding, stretch molding, compression molding, rotomolding or injection stretch blow molding. Articles produced by injection molding and blow molding are preferred. The molded articles preferably are thin-walled articles having a wall thickness of 300 µm to 2 mm. More preferably, the thin-walled articles have a wall thickness of 300 µm to 1400 µm, and even more preferably the thin-walled articles have a wall thickness of 500 µm to 900 µm. The molded articles of the present invention can be non-food packaging, containers, crates, automotive components or household articles. Preferably, the articles consist of the polymer blend as the sole polymer component.

The films may be prepared by any method known in the art, such as casting or extrusion. The films may be multilayer or monolayer films, but are preferably monolayer films. In one embodiment, the films consist of the polymer blend as the sole polymer component.

4. Use of the heterophasic polypropylene composition

The present invention further relates to the use of a heterophasic polypropylene composition comprising

b1) 55 to 85 wt.-%, based on the total weight of the heterophasic polypropylene composition and determined at 25 °C according to ISO 16152, of a xylene

insoluble fraction (XI) comprising propylene comonomer units and, based on the total weight of the xylene insoluble fraction (XI), ethylene comonomer units in a content in the range of from 0.5 to 3.0 wt.-% and 1-butene comonomer units in a content in the range of from 5.0 to 10.0 wt.-%, and

b2) 15 to 45 wt.-%, based on the total weight of the heterophasic polypropylene composition and determined at 25 °C according to ISO 16152, of a xylene soluble fraction (XS) comprising propylene comonomer units and, based on the total weight of the xylene soluble fraction (XS), ethylene comonomer units in a content in the range of from 18.0 to 50.0 wt.-% and 1-butene comonomer units in a content in the range of from 1.0 to 10.0 wt.-%, and the xylene soluble fraction (XS) having an intrinsic viscosity IV, determined according to ISO 1628-1 & 3, in the range of from 1.2 to 6.0 dl/g,

for improving mechanical properties of a post-consumer recycled polyolefin composition.

The heterophasic polypropylene composition in any of the above-described embodiments may be used.

In particular, the heterophasic polypropylene composition is used for improving the impact strength (i.e. Charpy Notched Impact Strength, as determined according to ISO 179/1eA on an injection molded specimen at +23 °C and at -20 °C) and/or elongation at break (as determined according to ISO 527-2 on an injection molded specimen).

Other mechanical properties (e.g. tensile modulus and tensile strength) are maintained in a range that makes the application of the polymer blend possible in a wide range of technical fields.

In particular, these properties are reached when using the heterophasic polypropylene composition in any of the above-described embodiments (i.e. with any of the preferred properties) to prepare blends with a post-consumer recycled polyolefin composition, wherein, based on the total weight of the polymer blend, from 5 to 40 wt.-%, preferably from 7 to 30 wt.-% and more preferably from 8 to 20 wt.-%, of the heterophasic polypropylene composition are comprised in the polymer blend.

It is particularly preferred that the impact strength as determined by ISO 179/1eA on an injection molded specimen at +23 °C is improved by enhancing the impact strength value of the post-consumer recycled polyolefin composition by at least 20 %, more preferably at least 30 % and even more preferably at least 35 % or more, in the polymer blend.

It is also preferred that the elongation at break as determined according to ISO 527-2 on an injection molded specimen is improved by enhancing the elongation at break value of the post-consumer recycled polyolefin composition by at least 30 %, more preferably at least 50 % and even more preferably at least 70 % or more, in the polymer blend.

Further preferred is that the heterophasic polypropylene composition does not increase the melt flow rate MFR_2 (230 °C) as determined according to ISO 1133, and preferably decreases the melt flow rate MFR_2 (230 °C), such that the processability of the polymer blend is facilitated.

Measurement methods

All parameters mentioned in the description of the invention or the examples were measured according to the methods described below.

Melt Flow Rate

The melt flow rate (MFR) was determined according to ISO 1133 and is indicated in g/10 min. The MFR is an indication of the melt viscosity of the polymer. The MFR is determined at 190 °C for PE and 230 °C for PP. The load under which the melt flow rate is determined is usually indicated as a subscript, for instance MFR_2 is measured under 2.16 kg load (condition D).

Description of microstructure quantification by NMR spectroscopy

Quantitative nuclear-magnetic resonance (NMR) spectroscopy was used to quantify the comonomer content of the polymers (i.e. the heterophasic polypropylene composition and its components).

Quantitative $^{13}C\{^1H\}$ NMR spectra were recorded in the molten-state using a Bruker Avance III 500 NMR spectrometer operating at 500.13 and 125.76 MHz for ^1H and ^{13}C respectively. All spectra were recorded using a ^{13}C optimised 7 mm magic-angle spinning (MAS) probe head at 180° C using nitrogen gas for all pneumatics. Approximately 200 mg of material was packed into a 7 mm outer diameter zirconia MAS rotor and spun at 4 kHz. This setup was chosen primarily for the high sensitivity needed for rapid identification and accurate quantification {klimke06, parkinson07, castignolles09}. Standard single-pulse excitation was employed utilising the NOE at short recycle delays of 3s {pollard04, klimke06} and the RS-HEPT decoupling scheme {fillip05,griffin07}. A total of 1024 (1k) transients were acquired per spectra.

Quantitative $^{13}C\{^1H\}$ NMR spectra were processed, integrated and relevant quantitative properties determined from the integrals. All chemical shifts are

internally referenced to the methyl isotactic pentad (*mmmm*) at 21.85 ppm and assignments done according {brandolini01, randall89, resconi00}.

Characteristic signals corresponding to the incorporation of 1-butene were observed and the comonomer content quantified in the following way. The amount isolated 1-butene incorporated in PBP sequences was quantified using the integral of the αB2 sites at 43.6 ppm accounting for the number of reporting sites per comonomer:

$B = I_{\alpha B2} / 2$

If observed the amount consecutively incorporated 1-butene in PBBP sequences was quantified using the integral of the $\alpha\alpha$B2 site at 40.6 ppm accounting for the number of reporting sites per comonomer:

$BB = 2 * I_{\alpha\alpha B2}$

In case of presence of consecutively incorporated 1-butene (BB) the isolated incorporated butene (B) needs to be corrected due to influencing signals by subtracting BB/2.

Characteristic signals corresponding to different incorporations of ethylene were observed and the comonomer content quantified utilising following assignments and equations for the respective sequences:

chemical shift [ppm]	assignment
47.9 - 45.1	$S\alpha\alpha$
30.1	$S\gamma\delta$
29.7	$S\delta\delta$
27.7 - 26.5	$S\beta\delta$
25.1 - 23.9	$S\beta\beta$

sequence	signal / equation
E	$I_{S\beta\beta}$
EE	$I_{S\beta\delta}$
EEE	$I_{(S\delta\delta/2) + (S\gamma\delta/4)}$

The amount of P was quantified based on the $S\alpha\alpha$ methylene site including additional propene units not covered by $S\alpha\alpha$:

$P = I_{S\alpha\alpha} + B + (0.5*BB) + E + (0.5*EE)$

The total amount of comonomers was then calculated as follows:

$B_{total} = B + BB$ $E_{total} = E + EE + EEE$ $P_{total} = P$

$B[mol\%] = 100 * B_{total} / (B_{total} + E_{total} + P_{total})$

$E[mol\%] = 100 * E_{total} / (B_{total} + E_{total} + P_{total})$

$P[mol\%] = 100 * P_{total} / (B_{total} + E_{total} + P_{total})$

The weight percent comonomer incorporation was calculated from the mol%:

$B\ [wt\%] = 100*(B[mol\%]*56.11) / ((E[mol\%] *28.05) + (B[mol\%] *56.11) + (P[mol\%]*42.08))$

$E\ [wt\%] = 100*(E[mol\%]*28.05) / ((E[mol\%] *28.05) + (B[mol\%] *56.11) + (P[mol\%]*42.08))$

For e.g. xylene soluble fraction where butene was not observed the related equations collapse to the following:

$P= I_{S\alpha\alpha} + E + (0.5*EE)$

The total amount of comonomer was then calculated as follows:

$E_{total} = E + EE + EEE$ $P_{total} = P$

$E[mol\%] = 100 * E_{total} / (E_{total} + P_{total})$

$P[mol\%] = 100 * P_{total} / (E_{total} + P_{total})$

The weight percent comonomer incorporation was calculated from the mol%:

$E\ [wt\%] = 100*(E[mol\%]*28.05) / ((E[mol\%] *28.05) + (P[mol\%]*42.08))$

References

klimke06: Klimke, K., Parkinson, M., Piel, C., Kaminsky, W., Spiess, H.W., Wilhelm, M., Macromol. Chem. Phys. 2006;207:382.

parkinson07: Parkinson, M., Klimke, K., Spiess, H.W., Wilhelm, M., Macromol. Chem. Phys. 2007;208:2128.

pollard04: Pollard, M., Klimke, K., Graf, R., Spiess, H.W., Wilhelm, M., Sperber, O., Piel, C., Kaminsky, W., Macromolecules 2004;37:813.

filip05: Filip, X., Tripon, C., Filip, C., J. Mag. Resn. 2005, 176, 239

griffin07: Griffin, J.M., Tripon, C., Samoson, A., Filip, C., and Brown, S.P., Mag. Res. in Chem. 2007 45, S1, S198

castignolles09: Castignolles, P., Graf, R., Parkinson, M., Wilhelm, M., Gaborieau, M., Polymer 50 (2009) 2373

resconi00: Resconi, L., Cavallo, L., Fait, A., Piemontesi, F., Chem. Rev. 2000, 100, 1253

randall89: J. Randall, Macromol. Sci., Rev. Macromol. Chem. Phys. 1989, C29, 201.

brandolini01: A.J. Brandolini, D.D. Hills, "NMR spectra of polymers and polymer additives", Marcel Deker Inc., 2000

abis86: L. Abis, Mackromol. Chem. 187, 1877-1886 (1986)

PCR polyolefin composition: Amount of propylene (co)polymer, ethylene (co)polymer, polystyrene, and polyamide-6

To establish different calibration curves different standards, first iPP and HDPE and second iPP, PS and PA6 were blended in relevant fractions. For the quantification of the content of the foreign polymers, IR spectra were recorded in the solid-state using a Bruker Vertex 70 FTIR spectrometer. Films were prepared with a compression-molding device at 190 ºC with 4 - 6 MPa clamping force. The thickness of the films for the calibration standards for iPP and HDPE was 300 µm and for the quantification of the iPP, PS and PA 6 50-100 µm film thickness was used. Standard transmission FTIR spectroscopy is employed using a spectral range of 4000-400 cm^{-1}, an aperture of 6 mm, a spectral resolution of 2 cm^{-1}, 16 background scans, 16 spectrum scans, an interferogram zero filling factor of 32 and Norton Beer strong apodization.

The absorption of the band at 1167 cm^{-1} in iPP is measured and the iPP content is quantified according to a calibration curve (absorption/thickness in cm versus iPP content in wt.-%).

The absorption of the band at 1601 cm^{-1} (PS) and 3300 cm^{-1} (PA6) are measured and the PS and PA6 content quantified according to the calibration curve (absorption/thickness in cm versus PS and PA content in wt.-%). The content of polyethylene and ethylene containing copolymers is obtained by subtracting (iPP+PS+PA6) from 100, taking into account the content of non-polymeric impurities as determined in the methods below. The analysis is performed as a double determination.

PCR polyolefin composition: Talc and chalk content

Thermogravimetric Analysis (TGA) experiments were performed with a Perkin Elmer TGA 8000 in line with ISO 3451-1 (1997). Approximately 10-20 mg of material was placed in a platinum pan. The temperature was equilibrated at 50 °C for 10 minutes, and afterwards raised to 950 °C under nitrogen at a heating rate of 20 °C/min. The weight loss between ca. 550 °C and 700 °C (WCO_2) was

assigned to CO_2 evolving from $CaCO_3$, and therefore the chalk content was evaluated as:

$$\text{Chalk content} = 100/44 \times WCO_2$$

Afterwards the temperature was lowered to 300°C at a cooling rate of 20 °C/min. Then the gas was switched to oxygen, and the temperature was raised again to 900 °C. The weight loss in this step was assigned to carbon black (Wcb). Knowing the content of carbon black and chalk, the ash content excluding chalk and carbon black was calculated as:

$$\text{Ash content} = (\text{Ash residue}) - 56/44 \times WCO_2 - Wcb$$

where Ash residue is the wt.-% measured at 900 °C in the first step conducted under nitrogen. The ash content is estimated to be the same as the talc content for the investigated recyclates.

PCR polyolefin composition: Amount of Metals

The metal amount was determined by X-ray fluorescence (XRF).

PCR polyolefin composition: Amount of Paper and Wood

Paper and wood amounts were determined by conventional laboratory methods including milling, floatation, microscopy and Thermogravimetric Analysis (TGA).

PCR polyolefin composition: Limonene measurement

Limonene quantification was carried out using solid phase micro-extraction (HS-SPME-GC-MS) by standard addition.

50 mg ground samples were weighed into 20 mL headspace vials and after the addition of limonene in different concentrations and a glass-coated magnetic stir bar. The vial was closed with a magnetic cap lined with silicone/PTFE. Micro capillaries (10 pL) were used to add diluted limonene standards of known concentrations to the sample. Addition of 0, 2, 20 and 100 ng equals 0 mg/kg, 0.1 mg/kg, 1 mg/kg and 5 mg/kg limonene, in addition standard amounts of 6.6 mg/kg, 11 mg/kg and 16.5 mg/kg limonene were used in combination with some of the samples tested in this application. For quantification, ion 93 acquired in SIM mode was used. Enrichment of the volatile fraction was carried out by headspace solid phase micro-extraction with a 2 cm stable flex 50/30 pm

DVB/Carboxen/PDMS fibre at 60 °C for 20 minutes. Desorption was carried out directly in the heated injection port of a GCMS system at 270 °C.

GCMS Parameters:
Column: 30 m HP 5 MS 0.25*0.25
Injector: Splitless with 0.75 mm SPME Liner, 270 °C
Temperature program: -10°C (1 min)
Carrier gas: Helium 5.0, 31 cm/s linear velocity, constant flow
MS: Single quadrupole, direct interface, 280 °C interface temperature
Acquisition: SIM scan mode
Scan parameter: 20-300 amu
SIM Parameter: m/Z 93, 100 ms dwell time

PCR polyolefin composition: Total free fatty acid content

Fatty acid quantification was carried out using headspace solid phase micro extraction (HS-SPME-GC-MS) by standard addition.

50 mg ground samples were weighed in 20 mL headspace vial and after the addition of limonene in different concentrations and a glass-coated magnetic stir bar the vial was closed with a magnetic cap lined with silicone/PTFE. 10 µL Micro-capillaries were used to add diluted free fatty acid mix (acetic acid, propionic acid, butyric acid, pentanoic acid, hexanoic acid and octanoic acid) standards of known concentrations to the sample at three different levels. Addition of 0, 50, 100 and 500 ng equals 0 mg/kg, 1 mg/kg, 2 mg/kg and 10 mg/kg of each individual acid. For quantification ion 60 acquired in SIM mode was used for all acids except propanoic acid, here ion 74 was used.

GCMS Parameter:
Column: 20 m ZB Wax plus 0.25*0.25
Injector: Split 5:1 with glass lined split liner, 250 °C
Temperature program: 40°C (1 min) @6 °C/min to 120 °C, @15 °C to 245 °C (5 min)
Carrier: Helium 5.0, 40 cm/s linear velocity, constant flow
MS: Single quadrupole, direct interface, 220°C inter face temperature
Acquisition: SIM scan mode
Scan parameter: 46-250 amu 6.6 scans/s
SIM Parameter: m/z 60,74, 6.6 scans/s

Molecular weight and molecular weight distribution

Molecular weight averages (M_z, M_w and M_n), Molecular weight distribution (MWD) and its broadness, described by polydispersity index, PDI= M_w/M_n (wherein M_n is the number average molecular weight and M_w is the weight average molecular weight) were determined by Gel Permeation Chromatography (GPC) according to ISO 16014-1:2003, ISO 16014-2:2003, ISO 16014-4:2003 and ASTM D 6474-12 using the following formulas:

$$M_n = \frac{\sum_{i=1}^{N} A_i}{\sum_{i=1}^{N} (A_i/M_i)} \quad (1)$$

$$M_w = \frac{\sum_{i=1}^{N} (A_i x\, M_i)}{\sum_{i=1}^{N} A_i} \quad (2)$$

$$M_z = \frac{\sum_{i=1}^{N} (A_i\, x\, M_i^2)}{\sum_{i=1}^{N} (A_i/M_i)} \quad (3)$$

For a constant elution volume interval ΔV_i, where A_i, and M_i are the chromatographic peak slice area and polyolefin molecular weight (MW), respectively associated with the elution volume, V_i, where N is equal to the number of data points obtained from the chromatogram between the integration limits.

A high temperature GPC instrument, equipped with either infrared (IR) detector (IR4 or IR5) from PolymerChar (Valencia, Spain) or differential refractometer (RI) from Agilent Technologies, equipped with 3x Agilent-PLgel Olexis and 1x Agilent-PLgel Olexis Guard columns was used. As the solvent and mobile phase 1,2,4-trichlorobenzene (TCB) stabilized with 250 mg/L 2,6-Di-tert-butyl-4-methyl-phenol) was used. The chromatographic system was operated at 160 °C and at a constant flow rate of 1 mL/min. 200 µL of sample solution was injected per analysis. Data collection was performed using either Agilent Cirrus software version 3.3 or PolymerChar GPC-IR control software.

The column set was calibrated using universal calibration (according to ISO 16014-2:2003) with 19 narrow MWD polystyrene (PS) standards in the range of 0.5 kg/mol to 11500 kg/mol. The PS standards were dissolved at room temperature over several hours. The conversion of the polystyrene peak molecular weight to polyolefin molecular weights is accomplished by using the Mark Houwink equation and the following Mark Houwink constants:

$K_{PS} = 19 \times 10^{-3}$ mL/g, $\alpha_{PS} = 0.655$

K_{PE} = 39 x 10^{-3} mL/g, α_{PE} = 0.725

K_{PP} = 19 x 10^{-3} mL/g, α_{PP} = 0.725

A third order polynomial fit was used to fit the calibration data.

All samples were prepared in the concentration range of 0.5 to 1 mg/ml and dissolved at 160 °C for 2.5 hours (PP) or 3 hours (PE) under continuous gentle shaking.

Flexural Modulus (FM)

The flexural modulus was determined in a 3-point-bending test at 23 °C according to ISO 178 on 80x10x4 mm^3 test bars injection molded in line with ISO 1873-2.

Differential scanning calorimetry (DSC)

Differential scanning calorimetry (DSC) analysis, melting temperature (T_m) and melt enthalpy (H_m) as well as crystallization temperature (T_c) were measured with a TA Instrument Q200 differential scanning calorimetry (DSC) on 5 to 7 mg samples. DSC was run according to ISO 11357 / part 3 /method C2 in a heat/cool/heat cycle with a scan rate of 10 °C/min in the temperature range of -30 to +225 °C. Crystallization temperature (T_c) was determined from the cooling step, while melting temperature (T_m) and melt enthalpy (H_m) were determined from the second heating step.

Throughout the present description, the term T_m is understood as peak temperature of melting as determined by DSC at a heating rate of 10 K/min.

Throughout the present description, the term T_c is understood as peak temperature of crystallization as determined by DSC at a cooling rate of 10 K/min.

Notched Impact Strength (NIS)

The Charpy notched impact strength (NIS) was measured according to ISO 179 1eA at +23 °C or at -20 °C, respectively, using injection molded bar test specimens of 80x10x4 mm^3 prepared in accordance with ISO 1873-2.

Xylene soluble fraction (XS) and xylene insoluble fraction (XI)

The xylene soluble fraction (XS) and xylene insoluble fraction (XI) were determined according to ISO 16152 at 25 °C.

Intrinsic viscosity (IV)

The IV of the xylene solubles was measured at 135 °C in decalin according to ISO 1628-1 & 3.

Tensile test

The tensile modulus (TM), tensile strength (TS) and elongation at break (EB) of the polymers were determined according to ISO 527-2 (cross head speed for linear range = 1 mm/min; test speed to break 50 mm/min at 23 °C) using injection molded specimens as described in EN ISO 1873-2 (dog bone 25 shape, 4 mm thickness).

Examples

Preparation of the heterophasic polypropylene composition

A heterophasic polypropylene composition was prepared (IE-PP) - which was later used in the polymer blends of the Inventive Examples IE1, IE2 and IE3 - in a multi-stage polymerization process as described above, under the conditions outlined in Table 1. The catalyst was a Ziegler-Natta catalyst prepared as described in EP-A-3562850. The solid catalyst component was used along with triethylaluminium (TEAL) as co-catalyst and dicyclo pentyl dimethoxy silane (D-donor) as external donor.

Additionally, three heterophasic polypropylene compositions (CE2-PP, CE3-PP and CE4,5-PP) were prepared in a similar way, i.e using the same catalyst and external donor, as also indicated in Table 1. These were further used in the polymer blends of the Comparative Examples CE2 to CE5. Among those polypropylene compositions, CE2-PP contains ethylene and 1-butene comonomers, while CE3-PP and CE4,5-PP are propylene-ethylene polymers. The difference between CE3-PP and CE4,5-PP is mainly the composition of the crystalline matrix that is a propylene homopolymer in case of CE4,5-PP, and a propylene-ethylene copolymer in CE3-PP.

The C2 content in the xylene soluble fraction of CE2-PP is lower than in IE-PP.

Table 1: Process conditions

Prepoly reactor	**IE-PP**	**CE2-PP**	**CE3-PP**	**CE4,5-PP**
Temperature [°C]	20	20	25	25
TEAL/C3 [g/t]	172	172	150	150
Donor/C3 [g/t]	60	60	30	30
Pressure reactor [barg]	52	53	55	55
H_2/C_3 ratio [mol/kmol]	0.68	0.68	0.0	0.0

Loop reactor (A1)				
Temperature [°C]	68	68	75	75
Pressure reactor [barg]	52	52	56	56
H_2/C_3 ratio [mol/kmol]	0.62	0.60	3.4	6.4
C_2/C_3 ratio [mol/kmol]	4.16	4.07	20	0.0
C_4/C_3 ratio [mol/kmol]	42.5	42.4	0.0	0.0
C_4 content [wt.-%]	6.9	6.9	0.0	0.0
C_2 content [wt.-%]	1.2	1.2	2.0	0.0
Split [wt.-%]	39	50	35	51
MFR_2 (230°C) [g/10 min]	1.5	2.3	5.7	42
XS [wt.-%]	4.9	5.2	4.5	1.6
Gas phase reactor 1 (A2, A)				
Temperature [°C]	78	78	75	80
Pressure reactor [barg]	18	18	20	22
H_2/C_3 ratio [mol/kmol]	2.49	2.51	18	69
C_4/C_3 ratio [mol/kmol]	186.9	157.1	0.0	0.0
C_2/C_3 ratio [mol/kmol]	17.2	17.2	55	0.0
Split [wt.-%]	41	44	51	30
MFR_2 (230 °C) [g/10 min]	1.5	1.7	1.6	42
C_4 content [wt.-%]	8.1	8.1	0.0	0.0
C_2 content [wt.-%]	1.8	1.8	6.0	0.0
C_4 content in GPR1 [wt.-%]	9.8	9.6	0.0	0.0
C_2 content in GPR1 [wt.-%]	2.4	2.5	8.5	0.0
XS [wt.-%]	9.3	9.8	11.2	2.0
Gas phase reactor 2 (B)				
Temperature [°C]	75	75	75	80
Pressure reactor [barg]	18	18	18	20
H_2/C_2 ratio [mol/kmol]	201.9	2.1	76	84
C_4/C_3 ratio [mol/kmol]	0.0	0.0	0.0	0.0
C_2/C_3 ratio [mol/kmol]	387	102	155	399
C_4 content [wt.-%]	6.5	7.6	0.0	0.0
C_2 content [wt.-%]	10.0	2.9	8.0	7.5
C_4 content in GPR2 [wt.-%]	0.0	0.0	0.0	0.0
C_2 content in GPR2 [wt.-%]	43.0	27.0	20.5	39.5
Split [wt.-%]	20	6	14	19
MFR_2 (230 °C) [g/10 min]	1.5	0.7	1.5	20
XS [wt.-%]	28.2	19.7	21.0	17.5

The polymers prepared above were mixed with 0.25 wt.-% of Irganox B215 (a synergistic 2:1 blend of antioxidants Irgafos 168 (tris(2,4-ditertbutylphenyl)phosphite, CAS No: 31570-04-4) and Irganox 1010 (pentaerythritol tetrakis[3-[3,5-di-tert-butyl-4-hydroxyphenyl]propionate], CAS No: 6683-19-8, commercially available from BASF SE, Germany) and 0.05 wt.-

% of CEASIT FI (Ca-stearate, CAS No: 1592-23-0, commercially available from Baerlocher GmbH, Germany), and then compounded in a twin screw extruder ZSK 18, with melt temperature of 200-220 °C and throughput of 7 kg/h.

Properties of the heterophasic polypropylene composition

Properties of the polymer compositions were measured and are summarized in Table 2 below.

Table 2: Properties of the polypropylene compositions

	Unit	IE-PP	CE2-PP	CE3-PP	CE4,5-PP
MFR_2 (230°C)	g/10 min	1.5	0.7	1.5	20
C_4 content	wt.-%	6.5	7.6	0.0	0.0
C_2 content	wt.-%	10.0	2.9	8.0	7.5
XS	wt.-%	28.8	19.7	21.0	17.5
IV (XS)	dl/g	2.14	3.58	2.3	2.6
C_3 content (XS)	wt.-%	70.1	81.1	84.3	66.0
C_2 content (XS)	wt.-%	27.2	10.6	15.7	34.0
C_4 content (XS)	wt.-%	2.7	8.3	0.0	0.0
C_2 content (XI)	wt.-%	1.5	1.4	5.9	1.9
C_4 content (XI)	wt.-%	7.5	7.2	0.0	0.0
T_m	°C	138	138	148	165
T_c	°C	95	94	115	130
Flexural Modulus	MPa	427	473	450	1450
Charpy NIS (+23°C)	kJ/m^2	86.3	84.5	75.0	8.5
Charpy NIS (-20°C)	kJ/m^2	2.1	2.1	3.6	4.5

Preparation of the blends

For the preparation of the blends, commercially available PCR product (Dipolen-S71) was compounded with the polypropylene compositions prepared as indicated above. While being subject to the variations expected for PCR products, Dipolen-S71 is typically characterized by the following composition:

Polypropylene content	59 wt.-%
Polyethylene content	33 wt.-%
Polystyrene content	≤ 2.0 wt.-%
Polyamide-6 content	≤ 2.5 wt.-%
Talc content	≤ 0.4 wt.-%
$CaCO_3$ content	≤ 0.2 wt.-%
Paper content	< 0.5 wt.-%

Wood content	< 0.5 wt.-%
Metal content	< 0.2 wt.-%
Limonene content	32 mg/kg
Total fatty acid content	71 mg/kg

The polymers were used in the contents as indicated in Table 3 below for the Inventive Examples and Table 4 below for the Comparative Examples, and compounded in a ZSK 18 twin screw extruder.

The properties of the so-obtained blends are also depicted in Tables 3 and 4.

Table 3: Polymer blends – Inventive Examples

	Unit	IE1	IE2	IE3
DIPOLEN S-71	wt.-%	92	85	75
IE-PP	wt.-%	8	15	25
MFR_2 (230°C)	g/10min	5.0	5.1	3.9
T_{m1} (PE)	°C	126	126	126
T_{m2} (PP)	°C	160	159	159
H_{m1} (PE)	J/g	40	38	35
H_{m2} (PP)	J/g	68	69	62
T_{c1} (PP)	°C	121	122	121
T_{c2} (PE)	°C	115	115	115
Tensile modulus	MPa	802	745	708
Tensile strength	MPa	19.5	18.4	18.1
Elongation at Break	%	415	424	390
Charpy NIS (+23°C)	kJ/m²	8.5	9.5	12.0
Charpy NIS (-20°C)	kJ/m²	2.4	2.6	3.2

Table 4: Polymer blends – Comparative Examples

	Unit	CE1	CE2	CE3	CE4	CE5
DIPOLEN S-71	wt.-%	100	85	85	85	70
CE2-PP	wt.-%		15			
CE3-PP	wt.-%			15		
CE4,5-PP	wt.-%				15	30

MFR_2 (230°C)	g/10min	6.6	4.5	5.2	8.2	14.4
T_{m1} (PE)	°C	126	126	126	126	126
T_{m2} (PP)	°C	161	159	159	165	165
H_{m1} (PE)	J/g	39	39	40	42	45
H_{m2} (PP)	J/g	69	71	69	67	60
T_{c1} (PP)	°C	122	121	121	124	126
T_{c2} (PE)	°C	115	115	115	115	115
Tensile modulus	MPa	831	800	779	952	1240
Tensile strength	MPa	19.8	20.0	19.5	22.5	24.4
Elongation at Break	%	223	411	422	35	28
Charpy NIS (+23°C)	kJ/m²	6.1	7.5	7.6	5.8	7.6
Charpy NIS (-20°C)	kJ/m²	2.1	2.1	2.1	1.9	2.2

Thus, the Inventive Examples were prepared with three different ratios of the PCR polymer to the heterophasic polymer composition IE-PP, namely 92:8, 85:15 and 75:25.

Comparative Example CE1 contained the PCR polymer only.

Comparative Examples CE2 to CE4 contained the PCR polymer and the respective polymer composition CE2-PP to CE4,5-PP in a ratio of 85:15. Comparative Example CE5 contained the PCR polymer and the polymer composition CE4,5-PP in a ratio of 70:30.

It is notable that the Charpy notched impact strength (NIS) is improved in all Inventive Examples – even at the very low concentration of 8 wt.-% of the polypropylene composition IE-PP. An improvement was reached over the pure PCR polymer (CE1) as well as in relation to all comparative polymer blends. In particular, at room temperature, the increase of NIS was more than 25 % compared to the PCR polymer (CE1).

Further, elongation at break was enhanced by more than 70 % in the Inventive Examples when compared to the PCR polymer (CE1).

Tensile modulus and strength were slightly decreased, however, they maintained in a range well suitable for a variety of technical applications.

Melt flow rate in a range of 3.9 to 5.1 g/10 min in the Inventive Examples facilitates the processability of the polymer blends.

Claims

1. A polymer blend comprising:

   a) 60 to 95 wt.-%, based on the total weight of the polymer blend, of a post-consumer recycled polyolefin composition having a melt flow rate MFR_2, determined according to ISO 1133 at 230 °C, in the range of from 1.0 to 100.0 g/10 min, and comprising propylene (co)polymer component(s) and ethylene (co)polymer component(s) in a ratio (weight/weight) of from 1:99 to 99:1, determined by FTIR spectroscopy as described in the specification, and

   b) 5 to 40 wt.-%, based on the total weight of the polymer blend, of a heterophasic polypropylene composition comprising a propylene copolymer (A) matrix of propylene with ethylene and 1-butene comonomer units and a propylene ethylene elastomer (B) dispersed within the matrix, the heterophasic polypropylene composition comprising

      b1) 55 to 85 wt.-%, based on the total weight of the heterophasic polypropylene composition and determined at 25 °C according to ISO 16152, of a xylene insoluble fraction (XI) comprising propylene comonomer units and, based on the total weight of the xylene insoluble fraction (XI) and determined by ^{13}C-NMR spectroscopy as described in the specification, ethylene comonomer units in a content in the range of from 0.5 to 3.0 wt.-% and 1-butene comonomer units in a content in the range of from 5.0 to 10.0 wt.-%, and

      b2) 15 to 45 wt.-%, based on the total weight of the heterophasic polypropylene composition and determined at 25 °C according to ISO 16152, of a xylene soluble fraction (XS) comprising propylene comonomer units and, based on the total weight of the xylene soluble fraction (XS) and determined by ^{13}C-NMR spectroscopy as described in the specification, ethylene comonomer units in a content in the range of from 18.0 to 50.0 wt.-% and 1-butene comonomer units in a content in the range of from 1.0 to 10.0 wt.-%, and the xylene soluble fraction (XS) having an intrinsic viscosity IV, determined according to ISO 1628-1 & 3, in the range of from 1.2 to 6.0 dl/g.

2. The polymer blend according to claim 1, wherein the heterophasic polypropylene composition comprises, based on the total weight of the

heterophasic polypropylene composition and determined by ^{13}C-NMR spectroscopy as described in the specification, a content of 1-butene comonomer units in the range of from 3.0 to 14.0 wt.-%, and/or a content of ethylene comonomer units in the range of from 3.0 to 15.0 wt.-%.

3. The polymer blend according to any one of the preceding claims, wherein the xylene soluble fraction (XS) comprises propylene comonomer units and, based on the total weight of the xylene soluble fraction (XS) and determined by ^{13}C-NMR spectroscopy as described in the specification, ethylene comonomer units in a content in the range of from 20.0 to 50.0 wt.-% and 1-butene comonomer units in a content in the range of from 1.0 to 10.0 wt.-%.

4. The polymer blend according to any one of the preceding claims, wherein the heterophasic polypropylene composition comprises

a) 65 to 85 wt.-%, based on the total weight of the heterophasic polypropylene composition and determined at 25 °C according to ISO 16152, of the xylene insoluble fraction (XI), and

b) 15 to 35 wt.-%, based on the total weight of the heterophasic polypropylene composition and determined at 25 °C according to ISO 16152, of the xylene soluble fraction (XS).

5. The polymer blend according to any one of the preceding claims, wherein the propylene ethylene elastomer (B) is a unimodal propylene copolymer and/or the propylene copolymer (A) is a multi-modal propylene copolymer, the multi-modal propylene copolymer (A) preferably comprising two different propylene polymer fractions of:

a) 30 to 70 wt.-%, based on the total weight of the propylene copolymer (A), of a first propylene polymer fraction (A1) being a propylene copolymer of propylene with ethylene comonomer units and 1-butene comonomer units, and

b) 30 to 70 wt.-%, based on the total weight of the propylene copolymer (A), of a second propylene polymer fraction (A2), being a propylene copolymer of propylene with ethylene comonomer units and 1-butene comonomer units.

6. The polymer blend according to any one of the preceding claims, wherein the heterophasic polypropylene composition has at least one of the following properties:

a) a flexural modulus, determined according to ISO 178, in the range of from 250 to 500 MPa, and/or

b) an impact strength at +23 °C, determined according to ISO 179/1eA, in the range of from 30.0 to 100.0 kJ/m^2, and/or

c) an impact strength at -20 °C, determined according to ISO 179/1eA, in the range of from 1.5 to 7.0 kJ/m^2, and/or

d) a melt flow rate MFR_2, determined according to ISO 1133 at 230 °C, in the range of from 0.5 to 5.0 g/10 min, and/or

e) a melting temperature T_m, determined according to differential scanning calorimetry (DSC) described in the specification, in the range of from 125 to 145 °C; and/or

f) a crystallization temperature T_c, determined according to differential scanning calorimetry (DSC) described in the specification, in the range of from 90 to 100 °C.

7. The polymer blend according to any one of the preceding claims, wherein the post-consumer recycled polyolefin composition further comprises, based on the total weight of the post-consumer recycled polyolefin composition, one or more of

a) from 0.1 to 100.0 ppm, preferably from 0.1 to 50.0 ppm, more preferably from 0.1 to 40.0 ppm, of limonene, determined by solid phase microextraction (HS-SPME-GC-MS);

b) from 0.1 to 100.0 ppm of fatty acids, determined by solid phase microextraction (HS-SPME-GC-MS);

c) from 0.1 to 10.0 wt.-% of non-polyolefin polymers, determined by FTIR spectroscopy as described in the specification;

d) from 0.1 to 10.0 wt.-% of other components, selected from talc, chalk, carbon, calcium stearate, titanium dioxide, pigments, metals, glass, paper, wood and combinations thereof, determined as described in the specification.

8. The polymer blend according to any one of the preceding claims, wherein the post-consumer recycled polyolefin composition has a melt flow rate MFR_2, determined according to ISO 1133 at 230 °C, in the range of from 2.0 to 20.0 g/10 min, and/or the post-consumer recycled polyolefin composition comprises propylene (co)polymer component(s) and ethylene (co)polymer component(s) in a ratio (weight/weight) of from 25:75 to 75:25, determined by FTIR spectroscopy as described in the specification.

9. The polymer blend according to any one of the preceding claims, wherein the polymer blend has at least one of the following properties:

a) an impact strength at +23 °C, determined according to ISO 179/1eA on an injection molded specimen, in the range of from 7.0 to 35.0 kJ/m^2, and/or

b) an impact strength at -20 °C, determined according to ISO 179/1eA on an injection molded specimen, in the range of from 2.0 to 12.0 kJ/m^2, and/or

c) a tensile modulus, determined according to ISO 527-2 on an injection molded specimen, in the range of from 500 to 1200 MPa, and/or

d) a tensile strength, determined according to ISO 527-2 on an injection molded specimen, in the range of from 15.0 to 25.0 MPa, and/or

e) an elongation at break, determined according to ISO 527-2 on an injection molded specimen, in the range of from 250 to 600 %, and/or

f) a melt flow rate MFR_2, determined according to ISO 1133 at 230 °C, in the range of from 1.5 to 20.0 g/10 min.

10. A process for the preparation of the polymer blend according to any one of the preceding claims, wherein the process comprises

A) providing the post-consumer recycled polyolefin composition as defined in any one of claims 1, 7 and 8,

B) providing the heterophasic polypropylene composition as defined in any one of claims 1 to 6 by preparing the heterophasic polypropylene composition in a multi-stage polymerization process in the presence of a Ziegler-Natta catalyst, and

C) blending the post-consumer recycled polyolefin composition and the heterophasic polypropylene composition, preferably by melting and mixing the components.

11. The process according to claim 10, wherein the Ziegler-Natta catalyst comprises:

a) compound(s) of a transition metal of Group 4 to 6 of IUPAC;

b) a Group 2 metal compound;

c) an internal donor, wherein said internal donor is a non-phthalic compound, preferably is a non-phthalic acid ester;

d) a co-catalyst; and

e) optionally an external donor.

12. The process according to claim 11, wherein the internal donor is preferably selected from (di)esters of non-phthalic carboxylic (di)acids, 1,3-diethers, derivatives and mixtures thereof.

13. An article comprising the polymer blend according to any one of claims 1 to 9, preferably being an injection molded article, a blow molded article or a film.

14. Use of the polymer blend according to any one of claims 1 to 9 for the manufacture of an article, preferably being an injection molded article, a blow molded article or a film.

15. Use of a heterophasic polypropylene composition comprising

b1) 55 to 85 wt.-%, based on the total weight of the heterophasic polypropylene composition and determined at 25 °C according to ISO 16152, of a xylene insoluble fraction (XI) comprising propylene comonomer units and, based on the total weight of the xylene insoluble fraction (XI) and determined by ^{13}C-NMR spectroscopy as described in the specification, ethylene comonomer units in a content in the range of from 0.5 to 3.0 wt.-% and 1-butene comonomer units in a content in the range of from 5.0 to 10.0 wt.-%, and

b2) 15 to 45 wt.-%, based on the total weight of the heterophasic polypropylene composition and determined at 25 °C according to ISO 16152, of a xylene soluble fraction (XS) comprising propylene comonomer units and, based on the total weight of the xylene soluble fraction (XS) and determined by ^{13}C-NMR spectroscopy as described in the specification, ethylene comonomer units in a content in the range of from 18.0 to 50.0 wt.-% and 1-butene comonomer units in a content in the range of from 1.0 to 10.0 wt.-%, and the xylene soluble fraction (XS) having an intrinsic viscosity IV, determined according to ISO 1628-1 & 3, in the range of from 1.2 to 6.0 dl/g,

for improving mechanical properties of a post-consumer recycled polyolefin composition.

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2024/067975

A. CLASSIFICATION OF SUBJECT MATTER
INV. C08L23/14 C08F210/06
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)
C08F 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 practicable, 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 2021/209326 A1 (BOREALIS AG [AT]) 21 October 2021 (2021-10-21) TERHECO-1 claims 1-14	1-15

Y	EP 2 143 760 B1 (BOREALIS AG [AT]) 9 March 2011 (2011-03-09) paragraphs [0004], [0014] example 2 claims 1-17	1-15

Y	EP 3 947 554 B1 (BOREALIS AG [AT]) 3 May 2023 (2023-05-03) paragraphs [0004], [112ff.] examples 1-4 claims 1-15	1-15

☐ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

* Special categories of cited documents :

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"E" earlier application or patent but published on or after the international filing date

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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

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"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
13 August 2024	17/09/2024

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/EP2024/067975

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO 2021209326 A1	21-10-2021	BR 112022020713 A2	29-11-2022
		CN 115397872 A	25-11-2022
		EP 4136129 A1	22-02-2023
		US 2023212338 A1	06-07-2023
		WO 2021209326 A1	21-10-2021
EP 2143760 B1	09-03-2011	AT E501216 T1	15-03-2011
		CN 102089378 A	08-06-2011
		EP 2143760 A1	13-01-2010
		WO 2010003953 A1	14-01-2010
EP 3947554 B1	03-05-2023	CN 113646378 A	12-11-2021
		EP 3947554 A1	09-02-2022
		KR 20210135614 A	15-11-2021
		WO 2020201020 A1	08-10-2020