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(57) Abstract: Upgraded recycling polyolefin composition including a styrene-grafted polypropylene compatibilizer.

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Upgraded recycled polyolefin

Field of Invention

The present invention relates to upgraded recycled polyolefin composition, and to a
5 process for obtaining such upgraded recycled polyolefin compositions.

Background

Polyolefins, in particular polyethylene and polypropylene are increasingly consumed
in large amounts in a wide range of applications, including packaging for food and
10 other goods, fibres, automotive components, and a great variety of manufactured
articles. Polyethylene based materials are a particular problem as these materials are
extensively used in packaging. Taking into account the huge amount of waste
collected compared to the amount of waste recycled back into the stream, there is
still a great potential for intelligent reuse of plastic waste streams and for mechanical
15 recycling of plastic wastes. Generally, recycled quantities of polypropylene on the
market are mixtures of both polypropylene (PP) and polyethylene (PE), this is
especially true for post-consumer waste streams. Moreover, commercial recyclates
from post-consumer waste sources are conventionally cross-contaminated with
non-polyolefin materials such as polyethylene terephthalate, polyamide, polystyrene
20 or non-polymeric substances like wood, paper, glass or aluminium. These cross-
contaminations drastically limit final applications of recycling streams such that no
profitable final uses remain.

If PET, in the form of bottles, is properly separated, postconsumer waste contains
mostly various polyolefins (high density, low density and linear low-density
25 polyethylenes and polypropylene including also copolymers thereof) and polystyrene
(neat, high-impact and foamed). Fortelny et al have suggested combined EPDM-SBS
compatibilizers on the basis of model systems including 10 wt.-% PS. In another
study, R. M. C. Santana and S. Manrich have suggested the use of copolymer
poly(styrene-b-ethylene-co-butylene-b-styrene) (SEBS) as compatibilizer for blends
30 of PP and IPS in weight ratios of up to 6:1. They found SEBS reduces the diameter of
HIPS dispersed particles and a concentration of 5 wt.-% of SEBS was found to be

beneficial. S. E. Luzuriaga, J. Kovarova, and I. Fortelny studied the effect of SEBS/EPR compatibilizers at PE/PP/PS/HIPS systems. It is further known from A.A. Dewole that graft copolymers of polypropylene and polystyrene (PP-g-PS) have limited impact resistance and require rubber toughening agents such as EPR or
5 styrene-b-ethylene-alt-butylene-b-styrene (SEBS) triblock copolymer, in a downstream compounding operation. G. Radonjic, V. Musil and I. Smit further found compatibilizing effects of the triblock copolymer poly(styrene-b-butadiene-b-styrene) (SBS) on the morphology and mechanical properties of polypropylene/polystyrene (PP/PS) blends. In yet a further study N. Equiza, W. Yave, R. Quijada, M. Yazdani-
10 Pedram have dealt with compatibilization of PE/PP/PS/HIPS blends by use of SEBS/EPR compatibilizers.

It is known from AKOVALI, Güneri, et al. (Hg.). *Frontiers in the science and technology of polymer recycling*. Springer Science & Business Media, 2013 that PP-g-PS having a content of 4% styrene when used as a compatibilizer for a recycling
15 PP-PS blend result in either maintenance or deterioration of impact strength and tensile strength.

It is further known from KARIAN, Harutun (Hg.). *Handbook of polypropylene and polypropylene composites, revised and expanded*. CRC press, 2003 that grade Hivalloy, a styrenic product, having allegedly 10% grafted polystyrene, was a
20 potential compatibilizer for polyolefins, polystyrene and PET recycling streams. However, due to various problems with Hivalloy, the resin was taken from the market. However, most studies that have dealt with relatively high PS and/or HIPS content in the streams to be recycled, whereby valid conclusions are impossible. Contents of styrene units such as of 10 wt.-% or higher result in completely different properties
25 profile. Moreover, high contents of styrene can be better addressed via mechanical separation when producing the recycle to be upgraded.

There was a long felt need for upgrading blends of polypropylene, polyethylene further containing low amounts of polystyrene, such as amounts of 1 to 5 wt.-% as
30 occurring in high quality polyolefin recycling streams. There was further a long felt need for providing an alternative for polystyrene-b-poly(ethylene/propylene)-b-styrene

(SEPS) compatibilizers. In yet a further aspect there was a need for avoiding rubber toughening by EPR or SEBS. Moreover, previously employed compatibilizers have addressed impact strength of recyclates but had to be used in amounts leading to a reduction of stiffness. The present invention at least in part addresses these and
5 related objects.

Summary of the Invention

The present invention is based on the finding that recycling blend comprising low
10 amounts of polystyrene, such as 0.5 to 5.0 wt.-%, can be upgraded surprisingly well by blending 0.3 to 4.0 wt.-% of a styrene-grafted polypropylene having a melt flow rate (ISO 1133, 2.16 kg, 190°C) of 4.0 to 20 g/10min, and a specific styrene distribution characterized by
three fractions when being subjected to preparative Temperature Rising Elution
15 Fractionation (p-TREF) using TCB as eluent, in amounts of
fraction (i) eluting up to 50°C 5 to 15 wt.-%
fraction (ii) eluting from 50°C to 105°C 5 to 15 wt.-% and
fraction (iii) eluting above 105°C 70 to 90 wt.-%,
whereby
20 fraction (i) has a polystyrene content as determined by ¹H NMR in an amount of 50 to 70 wt.-%;
and whereby
fraction (ii) has a polystyrene content as determined by ¹H NMR in an amount of 1 to 10 wt.-%;
25 and whereby
fraction (iii) has a polystyrene content as determined by ¹H NMR in an amount of less than 5 wt.-%;
and whereby
the total polystyrene content as determined by ¹H NMR of fractions (i) to (iii) is from 6
30 to 14 wt.-%.

The present invention insofar provides

a polyolefin composition obtainable by blending

a) 96.0 to 99.7 wt.-% of a blend (A) comprising polypropylene, polyethylene, polystyrene, and limonene, having

- 5 A-1) a content of isotactic polypropylene of 30 – 70 wt.-%,
A-2) a content of ethylene derived from polyethylene and ethylene containing copolymers of 20 – 50 wt.-%,
A-3) 0.5 to 5.0 wt.-% of polystyrene,
A-4) 0 to 3.0 wt.-% stabilizers,
10 A-5) 0 to 4.0 wt.-% polyamide-6,
A-6) 0 to 3.0 wt.-% talc,
A-7) 0 to 3.0 wt.-% chalk,
A-8) 0 to 1.0 wt.-% paper,
A-9) 0 to 1.0 wt.-% wood,
15 A-10) 0 to 0.5 wt.-% metal,
A-11) 0.1 ppm to 100 ppm of limonene as determined by using solid phase microextraction (HS-SPME-GC-MS),
A-12) 0 to 200 ppm total fatty acid content as determined by using solid phase microextraction (HS-SPME-GC-MS) wherein all amounts are given with respect to
20 the total weight of blend (A),
wherein blend (A) is a recycled material, which is recovered from a waste plastic material derived from post-consumer and/or post-industrial waste; and wherein blend (A) has a melt flow rate (ISO 1133, 2.16 kg, 230°C) of 4.0 to 20 g/10min;
and
25 b) 0.3 to 4.0 wt.-% of a compatibilizer (B)
being a styrene-grafted polypropylene having
a melt flow rate (ISO 1133, 2.16 kg, 190°C) of 4.0 to 20 g/10min, and
three fractions when being subjected to preparative Temperature Rising Elution Fractionation (p-TREF) using TCB as eluent, in amounts of
30 fraction (i) eluting up to 50°C 5 to 15 wt%
fraction (ii) eluting from 50°C to 105°C 5 to 15 wt% and

fraction (iii) eluting above 105°C 70 to 90 wt%,

whereby

fraction (i) has a polystyrene content as determined by ¹H NMR in an amount of 50 to 70 wt%;

5 and whereby

fraction (ii) has a polystyrene content as determined by ¹H NMR in an amount of 1 to 10 wt%%;

and whereby

fraction (iii) has a polystyrene content as determined by ¹H NMR in an amount of less
10 than 5 wt%, and whereby

the total polystyrene content as determined by ¹H NMR of fractions (i) to (iii) is from 6 to 14 wt%,

whereby the polyolefin composition has a tensile modulus of at least 850 MPa (ISO 527-1,2) when measured on an injection molded test specimen.

15

The present invention is further directed to an article comprising the polyolefin composition according to the present invention.

In a further aspect, the present invention is concerned with a process for providing a
20 polyolefin composition according to the present invention, the process comprising the steps of:

a) providing the blend (A) in an amount of 96.0 to 99.7 wt.-%, based on the overall weight of the polyolefin composition

b) providing the compatibilizer (B) in an amount of 0.3 to 4.0 wt.-%, based on the
25 overall weight of the polyolefin composition

c) melting and mixing the blend of blend (A) and the compatibilizer (B) in an extruder, and

d) optionally pelletizing the obtained polyolefin composition.

30 In a yet a further aspect, the present invention is concerned with the use of a compatibilizer (B) being a styrene-grafted polypropylene having

- a) a melt flow rate (ISO 1133, 2.16 kg, 190°C) of 4.0 to 20 g/10min, and
 b) three fractions when being subjected to preparative Temperature Rising Elution Fractionation (p-TREF) with TCB as eluent, in amounts of
- | | | |
|---|--|-------------------|
| | fraction (i) eluting up to 50°C | 5 to 15 wt.-% |
| 5 | fraction (ii) eluting from 50°C to 105°C | 5 to 15 wt.-% and |
| | fraction (iii) eluting above 105°C | 70 to 90 wt.-%, |

whereby

fraction (i) has a polystyrene content as determined by ^1H NMR in an amount of 50 to 70 wt%;

10 and whereby

fraction (ii) has a polystyrene content as determined by ^1H NMR in an amount of 1 to 10 wt%;

and whereby

15 fraction (iii) has a polystyrene content as determined by ^1H NMR in an amount of less than 5 wt%, and whereby

the total polystyrene content as determined by ^1H NMR of fractions (i) to (iii) is from 6 to 14 wt.-%,

for blending with of a blend (A) comprising polypropylene, polyethylene, polystyrene, and limonene, having

20 A-1) a content of isotactic polypropylene of 30 – 70 wt.-%,

A-2) a content of ethylene derived from polyethylene and ethylene containing copolymers of 20 – 50 wt.-%,

A-3) 0.5 to 5.0 wt.-% of polystyrene,

A-4) 0 to 3.0 wt.-% stabilizers,

25 A-5) 0 to 4.0 wt.-% polyamide-6,

A-6) 0 to 3.0 wt.-% talc,

A-7) 0 to 3.0 wt.-% chalk,

A-8) 0 to 1.0 wt.-% paper,

A-9) 0 to 1.0 wt.-% wood,

30 A-10) 0 to 0.5 wt.-% metal,

A-11) 0.1 ppm to 100 ppm of limonene as determined by using solid phase microextraction (HS-SPME-GC-MS), and

A-12) 0 to 200 ppm total fatty acid content using solid phase microextraction (HS-SPME-GC-MS)

- 5 wherein all amounts are given with respect to the total weight of blend (A) wherein blend (A) is a recycled material, which is recovered from a waste plastic material derived from post-consumer and/or post-industrial waste; and wherein blend (A) has a melt flow rate (ISO 1133, 2.16 kg, 230°C) of 4 to 20 g/10min, to obtain a polyolefin composition having a tensile modulus of at least 850 MPa (ISO
10 527-1,2) and a Charpy Impact Strength (ISO 179-1; 1eA, 23°C) of more than 6.0 kJ/m² and up to 10.0 kJ/m².

Unless defined otherwise, all technical and scientific terms used herein have the same meaning as commonly understood by one of ordinary skill in the art to which
15 the invention pertains. Although, any methods and materials similar or equivalent to those described herein can be used in practice for testing of the present invention, the preferred materials and methods are described herein. In describing and claiming the present invention, the following terminology will be used in accordance with the definitions set out below.

20

Unless clearly indicated otherwise, use of the terms “a,” “an,” and the like refers to one or more.

For the purposes of the present description and of the subsequent claims, the term
25 “recycled waste” is used to indicate a material recovered from both post-consumer waste and industrial waste, as opposed to virgin polymers. Post-consumer waste refers to objects having completed at least a first use cycle (or life cycle), i.e. having already served their first purpose; while industrial waste refers to manufacturing scrap, which does not normally reach a consumer.

30

The term “virgin” denotes the newly produced materials and/or objects prior to their first use, which have not already been recycled.

Many different kinds of polyethylene or polypropylene can be present in “recycled waste”. Blend (A) according to the present invention includes at least polypropylene,
5 polyethylene, polystyrene, limonene and fatty acids.

Blend (A) is further characterized by a content of isotactic polypropylene of 30 – 70 wt.-%.

10 Blend (A) is further characterized by a content of ethylene derived from polyethylene and ethylene containing copolymers of 20 – 50 wt.-%. Polyethylene denotes any of the conventional polyethylenes such as LDPE, LLDPE, MDPE, and HDPE.
Ethylene containing copolymers are extremely widespread and may include for example ethylene propylene copolymers such as ethylene propylene rubber,
15 plastomers such as C₂C₈ rubbers, and countless other polymers including ethylene-derived units.

The amount of 0 to 4.0 wt.-% polyamide-6 is further a realistic measurement of the total amount of units derived from amides considering the common and widespread
20 use of polyamide-6 resulting in an acceptable error margin.

The term “recycled material” such as used herein denotes materials reprocessed from “recycled waste”.

25 A polymer blend denotes a mixture of several polymeric components.

In general, blend (A) can be prepared by mixing the two or more polymeric streams as well known in the art.

A “compatibilizer” is a substance in polymer chemistry, which is added to a blend of polymers having limited miscibility in order to improve the mechanical properties thereof.

- 5 If not indicated otherwise “%” refers to weight-%.

Detailed Description

Blend (A)

10

The polyethylene-polypropylene composition according to the present invention comprises from 96 to 99.7 wt.-% of blend (A). It is the essence of the present invention that blend (A) is obtained from a recycled waste stream. Blend (A) can be either recycled post-consumer waste- or industrial waste, such as for example from
15 the automobile industry, or alternatively, a combination of both.

It is particularly preferred that blend (A) consists of recycled post-consumer waste and/or industrial waste.

- 20 Preferably, blend (A) is obtained from recycled waste by means of plastic recycling processes known in the art. Such recyclates are commercially available, e.g. from Corepla (Italian Consortium for the collection, recovery, recycling of packaging plastic wastes), Resource Plastics Corp. (Brampton, ON), Kruschitz GmbH, Plastics and Recycling (AT), Vogt Plastik GmbH (DE), Mtm Plastics GmbH (DE) etc. Non-
25 exhaustive examples of polyethylene rich recycled materials include: DIPOLEN S (Mtm Plastics GmbH), food grade rHDPE (BIFFA PLC) and a range of polyethylene rich materials, such as e.g. HD-LM02041 from PLASgran Ltd.

- In a certain preferred embodiment, the recycled polyethylene rich material is
30 DIPOLEN (Mtm Plastics GmbH), such as DIPOLEN S or DIPOLEN H, preferably DIPOLEN S. DIPOLEN is obtained from domestic waste streams (i.e. it is a product

of domestic recycling) for example the “yellow bag” recycling system, which operates in some parts of Germany.

Blend (A) comprises the following components:

- 5 A-1) a content of isotactic polypropylene of 30 – 70 wt.-%,
- A-2) a content of ethylene derived from polyethylene and ethylene containing copolymers of 20 – 50 wt.-%,
- A-3) 0.5 to 5.0 wt.-% of polystyrene,
- A-4) 0 to 3.0 wt.-% stabilizers,
- 10 A-5) 0 to 4.0 wt.-% polyamide,
- A-6) 0 to 3.0 wt.-% talc,
- A-7) 0 to 3.0 wt.-% chalk,
- A-8) 0 to 1.0 wt.-% paper,
- A-9) 0 to 1.0 wt.-% wood, and
- 15 A-10) 0 to 0.5 wt.-% metal, and
- A-11) 0.1 ppm to 100 ppm of limonene as determined by using solid phase microextraction (HS-SPME-GC-MS)
- A-12) 0 to 200 ppm total fatty acid content as determined by using solid phase microextraction (HS-SPME-GC-MS)
- 20 wherein all amounts are given with respect to the total weight of blend (A),

Blend (A) preferably may have a content of ethylene derived from polyethylene and ethylene containing copolymers of greater than 35 wt.-%, more preferably greater than 40 wt.-% with respect to the total weight of blend (A).

25

In addition, blend (A) usually has a content of isotactic polypropylene of greater than 30 wt.-%, but less than 70 wt.-%, with respect to the total weight of blend (A), preferably may have content of isotactic polypropylene of greater than 40 wt.-%, but less than 65 wt.-%, with respect to the total weight of blend (A).

30

Blend (A) may also have a relative amount of polystyrene of between 0.5 and 5.0 wt.-%, preferably between 1.0 and 4.0 wt.-%, more preferably between 1.0 and 3.0 wt.-%, most preferably between 1.5 and 2.5 wt.-%.

- 5 The polyethylene of the recycled material typically includes recycled high-density polyethylene (rHDPE), recycled medium-density polyethylene (rMDPE), recycled low-density polyethylene (rLDPE) and the mixtures thereof.

In a certain embodiment, the polyethylene is high density PE with an average
10 density of greater than 0.8 g/cm³, preferably greater than 0.9 g/cm³, most preferably greater than 0.91 g/cm³.

According to the present invention, blend (A) has a content of limonene as determined using solid phase microextraction (HS-SPME-GC-MS) of 0.1 ppm to 50
15 ppm, more preferably from 0.1 ppm to 30 ppm, most preferably from 0.1 ppm to 10 ppm.

Limonene is conventionally found in recycled polyolefin materials and originates from packaging applications in the field of cosmetics, detergents, shampoos and similar products. Therefore, blend (A) contains limonene, when blend (A) contains material
20 that originates from such types of domestic waste streams.

Lower amounts of the preferred ranges such as mentioned above, i.e. 0.1 ppm to 50 ppm, more preferred range from 0.1 ppm to 30 ppm, and most preferred range from 0.1 ppm to 10 ppm are easily accessible by aeration and/or by washing, preferable by aeration and repeated washing.

25

The fatty acid content is yet another indication of the recycling origin of blend (A). However, in some cases, the fatty acid content may be below the detection limit due to specific treatments in the recycling process. According to the present invention, blend (A) preferably has a content of fatty acids as determined using solid phase
30 microextraction (HS-SPME-GC-MS) of from 1 ppm to 200 ppm, preferably from 1

ppm to 150 ppm, more preferably from 2 ppm to 100 ppm, most preferably from 3 ppm to 80 ppm.

Due to the recycling origin blend (A) may also contain:

- 5 – organic fillers, and/or
 – inorganic fillers, and/or
 – additives

in amounts of up to 3 wt.-% with respect to the weight of blend (A).

- 10 According to the present invention, blend (A) has a melt flow rate (ISO 1133, 2.16 kg, 230°C) of 4 to 20 g/10min, preferably of 5 to 15 g/10min, more preferably of 6 to 12 g/10min.

As stated above blend (A) may include one or more further components, selected
15 from:

- A-4) up to 3.0 wt.-% stabilizers, preferably up to 2.0 wt.-% stabilizers,
A-5) up to 4.0 wt.-% polyamide-6, preferably up to 2.0 wt.-% polyamide-6,
A-6) up to 3.0 wt.-% talc, preferably up to 1.0 wt.-% talc,
A-7) up to 3.0 wt.-% chalk, preferably up to 1.0 wt.-% chalk,
20 A-8) up to 1.0 wt.-% paper, preferably up to 0.5 wt.-% paper,
A-9) up to 1.0 wt.-% wood, preferably up to 0.5 wt.-% wood, and
A-10) up to 0.5 wt.-% metal, preferably up to 0.1 wt.-% metal,
based on the overall weight of blend (A).

- 25 It is needless to say, during recycling usually any reasonable measure will be taken for lowering polyamides, talc, chalk, paper, wood and metal as far as final application or use suggests such measure.

Compatibilizer (B)

The polyolefin composition of the invention comprises 0.3 to 4.0 wt.-% of a compatibilizer (B), being a styrene-grafted polypropylene with respect to the total of the polyolefin composition. In a preferred embodiment the amount of compatibilizer (B) is 0.4 to 3.5 wt.-% with respect to the total of the polyolefin composition.

The term 'styrene-grafted' has a well-known meaning in the art.

The styrene-grafted polypropylene compatibilizer (B) has a melt flow rate (ISO 1133, 2.16 kg, 190°C) of 4 to 20 g/10 min, preferably of 6 to 18 g/10min, more preferably of 8 to 16 g/10min.

and

three fractions when being subjected to preparative Temperature Rising Elution

Fractionation (p-TREF) TCB as eluent, in amounts of

fraction (i) eluting up to 50°C 5 to 15 wt.-%

fraction (ii) eluting from 50°C to 105°C 5 to 15 wt.-% and

fraction (iii) eluting above 105°C 70 to 90 wt.-%,

whereby

fraction (i) has a polystyrene content as determined by ¹H NMR in an amount of 50 to 70 wt.-%;

and whereby

fraction (ii) has a polystyrene content as determined by ¹H NMR in an amount of 1 to 10 wt.-%;

and whereby

fraction (iii) has a polystyrene content as determined by ¹H NMR in an amount of less than 5 wt.-%, and whereby

the total polystyrene content as determined by ¹H NMR of fractions (i) to (iii) is from 6 to 14 wt.-%,

30

The fractions (i) to (iii) of compatibilizer (B) preferably fulfill the following one or more criteria:

isotacticity of 30 to 60 mmmm% ^{13}C NMR in fraction (i) eluting up to 50°C
and/or

5 isotacticity of 70 to 90 mmmm% ^{13}C NMR in fraction (ii) eluting from 50°C to 105°C
and/or

isotacticity of 90 to 99 mmmm% ^{13}C NMR in fraction (iii) eluting above 105°C.

Even more preferred the fractions (i) to (iii) of compatibilizer (B) fulfill the following
10 one or more criteria:

isotacticity of 40 to 60 mmmm% ^{13}C NMR in fraction (i) eluting up to 50°C
and/or

isotacticity of 70 to 90 mmmm% ^{13}C NMR in fraction (ii) eluting from 50°C to 105°C
and/or

15 isotacticity of 95 to 99 mmmm% ^{13}C NMR in fraction (iii) eluting above 105°C.

Without wishing to be bound by theory it is believed the high amount of the fraction
eluting above 105°C and further, in an preferred embodiment, the high isotacticity of
said fractions is responsible for the high stiffness of the resulting upgraded
20 composition.

Further preferred is an embodiment in which the styrene-grafted polypropylene
compatibilizer (B) has a melt flow rate (ISO 1133, 2.16 kg, 190°C) of 8 to 16 g/10
min, a total polystyrene content as determined by ^1H NMR of fractions (i) to (iii) of 8
25 to 12 wt.-% as determined by ^1H NMR.

The preparation of the compatibilizer (B) can be done as described in SUN, Yi-Jun, et
al. In situ compatibilization of polyolefin and polystyrene using Friedel—Crafts
alkylation through reactive extrusion. *Polymer*, 1998, 39. Jg., Nr. 11, S. 2201-2208.

30

Commercial compatibilizers (B) fulfilling the requirements of the present invention are also available with the most prominent resin being Byk™ Kometra SCONA TPPP 1616 FA. A person skilled in the art will understand that existing commercial grades may be further modified for the present invention.

5

Polyolefin composition

The polyolefin composition of the invention is obtainable by blending

10 a) 96.0 to 99.7 wt.-% of of a blend (A) comprising polypropylene, polyethylene, polystyrene, and limonene, having

A-1) a content of isotactic polypropylene of 30 – 70 wt.-%,

A-2) a content of ethylene derived from polyethylene and ethylene containing copolymers of 20 – 50 wt.-%,

15 A-3) 0.5 to 5.0 wt.-% of polystyrene,

A-4) 0 to 3.0 wt.-% stabilizers,

A-5) 0 to 4.0 wt.-% polyamide-6,

A-6) 0 to 3.0 wt.-% talc,

A-7) 0 to 3.0 wt.-% chalk,

20 A-8) 0 to 1.0 wt.-% paper,

A-9) 0 to 1.0 wt.-% wood,

A-10) 0 to 0.5 wt.-% metal,

A-11) 0.1 ppm to 100 ppm of limonene as determined by using solid phase microextraction (HS-SPME-GC-MS), and

25 A-12) 0 to 200 ppm total fatty acid content by using solid phase microextraction (HS-SPME-GC-MS)

wherein all amounts are given with respect to the total weight of blend (A),

30 wherein blend (A) is a recycled material, which is recovered from a waste plastic material derived from post-consumer and/or post-industrial waste; and

wherein blend (A) has a melt flow rate (ISO 1133, 2.16 kg, 230°C) of 4.0 to 20.0 g/10min;

and

b) 0.3 to 4.0 wt.-% of a compatibilizer (B)

5 wherein blend (A) and compatibilizer (B) are as described in the above sections.

The inventive polyolefin composition may have a melt flow rate (ISO 1133, 2.16 kg, 230°C) of between 3.0 and 10.0 g/10min, preferably between 4.0 and 8.0 g/10min, more preferably between 4.0 and 7.0 g/10min, and still further preferably between 4.5
10 and 6.0 g/10 min.

As outlined above, it is appreciated that the polyolefin composition according to the invention features a good impact strength without compromising the stiffness reflected by the tensile modulus of at least 850 MPa (ISO 527-1,2) when measured
15 on an injection molded test specimen .

Accordingly, it is preferred that the polyolefin composition of the invention has a Charpy Impact Strength (ISO 179-1; 1eA, 23°C) of more than 6.0 kJ/m² and up to 10 kJ/m², preferably of more than 6.2 kJ/m² and up to 10 kJ/m², more preferably of more
20 than 6.4 kJ/m² and up to 10 kJ/m². Usually the Charpy Impact Strength (ISO 179-1; 1eA, 23°C) will not be higher than 10.0 kJ/m² and in some embodiments even not higher than 9 kJ/m².

Additionally, the inventive polyolefin composition has a tensile modulus of at least
25 850 MPa (ISO 527-1,2) when measured on an injection molded test specimen, preferably of at least 880 MPa, more preferably of at least 900 MPa, still more preferably of at least 920 MPa, further preferably 940 MPa and most preferably of at least 950 MPa. Usually the tensile modulus (ISO 527-1,2) of the inventive polyolefin composition when measured on an injection molded test specimen will not be higher
30 than 1200 MPa.

In one embodiment, the inventive polyolefin composition is characterized by at least one, preferably all, of the following features:

- 5 a) a melt flow rate (ISO 1133, 2.16 kg, 230°C) of between 3.0 and 10.0 g/10min, preferably between 4.0 and 8.0 g/10min, more preferably between 4.0 and 7.0 g/10min, and still further preferably between 4.5 and 6.0 g/10 min
- b) a tensile modulus of at least 850 MPa (ISO 527-1,2) when measured on an injection molded test specimen, preferably of at least 920 MPa, more preferably of at least 940 MPa, and most preferably of at least 950 MPa
- 10 c) a Charpy Impact Strength (ISO 179-1; 1eA, 23°C) of more than 6.0 kJ/m² and up to 10 kJ/m², preferably of more than 6.2 kJ/m² and up to 10 kJ/m², more preferably of more than 6.4 kJ/m² and up to 10 kJ/m².

15 In a further embodiment, the ratio of the melt flow rate (ISO 1133, 2.16 kg, 190°C) of the compatibilizer (B) to the melt flow rate of the blend (A) (ISO 1133, 2.16 kg, 230°C) $\text{MFR(B, (ISO 1133, 2.16 kg, 190°C))} / \text{MFR (A, ISO 1133, 2.16 kg, 230°C)}$ is in the range of 0.5 to 2.0.

20 **Stabilizers**

As described above, the polyolefin composition, and more preferably the blend (A), can and usually will comprise stabilizers. These stabilizers are typically compounds such as antioxidants, anti-acids, anti-blocking agents, anti-UVs, nucleating agents
25 and anti-static agents.

Examples of antioxidants which are commonly used in the art are sterically hindered phenols (such as CAS No. 6683-19-8, also sold as Irganox 1010 FF™ by BASF), phosphorous based antioxidants (such as CAS No. 31570-04-4, also sold as
30 Hostanox PAR 24 (FF)™ by Clariant, or Irgafos 168 (FF)™ by BASF), sulphur based antioxidants (such as CAS No. 693- 36-7, sold as Irganox PS-802 FL™ by

BASF), nitrogen-based antioxidants (such as 4,4'-bis(1,1'-dimethylbenzyl)diphenylamine), or antioxidant blends.

Anti-acids are also commonly known in the art. Examples are calcium stearates,
5 sodium stearates, zinc stearates, magnesium and zinc oxides, synthetic hydrotalcite (e.g. SHT, CAS-No. 11097-59-9), lactates and lactylates, as well as calcium stearate (CAS No. 1592-23-0) and zinc stearate (CAS No. 557-05-1);

Common antiblocking agents are natural silica such as diatomaceous earth (such as
10 CAS No. 60676-86-0 (SuperfFloss™), CAS-No. 60676-86-0 (SuperFloss E™), or CAS-No. 60676-86-0 (Celite 499™)), synthetic silica (such as CAS-No. 7631-86-9, CAS-No. 7631-86-9, CAS-No. 7631-86-9, CAS-No. 7631-86-9, CAS-No. 7631-86-9, CAS-No. 112926-00-8, CAS-No. 7631-86-9, or CAS-No. 7631-86-9), silicates (such as aluminium silicate (Kaolin) CAS-no. 1318-74-7, sodium
15 aluminum silicate CAS-No. 1344-00-9, calcined kaolin CAS-No. 92704-41-1, aluminum silicate CAS-No. 1327-36-2, or calcium silicate CAS-No. 1344-95-2), synthetic zeolites (such as sodium calcium aluminosilicate hydrate CAS-No. 1344-01-0, CAS-No. 1344-01-0, or sodium calcium aluminosilicate, hydrate CAS-No. 1344-01-0).

20

Anti-UVs are, for example, Bis-(2,2,6,6-tetramethyl-4-piperidyl)-sebacate (CAS –No. 52829-07-9, Tinuvin™ 770); 2-hydroxy-4-n-°Ctoxy-benzophenone (CAS-No. 1843-05-6, Chimassorb 81).

25 Nucleating agents like sodium benzoate (CAS No. 532-32-1); 1,3:2,4-bis(3,4-dimethylbenzylidene)sorbitol (CAS 135861-56-2, Millad 3988) can be contained.

Suitable antistatic agents are, for example, glycerol esters (CAS No. 97593-29-8) or ethoxylated amines (CAS No. 71786-60-2 or 61791-31-9) or ethoxylated amides
30 (CAS No. 204-393-1).

Article

The present application is further directed to an article comprising the polyolefin composition as described above.

5

All preferred aspects and embodiments as described above shall also hold for the article.

Process

10

The process according to the present invention for preparing the polyolefin composition, comprises the steps of

- a) providing the blend (A) as described herein in an amount of 96.0 to 99.7 wt.-%, based on the overall weight of the polyolefin composition
- 15 b) providing the compatibilizer (B) as described herein in an amount of 0.3 to 4.0 wt.-%, based on the overall weight of the polyolefin composition
- c) melting and mixing the blend of blend (A) and the compatibilizer (B) in an extruder, and
- d) optionally pelletizing the obtained polyolefin composition.

20

All preferred aspects and embodiments as described above shall also hold for the process.

Use of compatibilizer (B)

25

The present invention is further directed to the use of a compatibilizer (B) being a styrene-grafted polypropylene having

- a) a melt flow rate (ISO 1133, 2.16 kg, 190°C) of 4.0 to 20.0 g/10min, and
- b) three fractions when being subjected to preparative Temperature Rising Elution Fractionation (p-TREF) using TCB as eluent, in amounts
- 30 of

fraction (i) eluting up to 50°C 5 to 15 wt.-%, and

fraction (ii) eluting from 50°C to 105°C 5 to 15 wt.-% and

fraction (iii) eluting above 105°C 70 to 90 wt.-%,

whereby

5 fraction (i) has a polystyrene content as determined by ^1H NMR in an amount of 50 to 70 wt.-%;

and whereby

fraction (ii) has a polystyrene content as determined by ^1H NMR in an amount of 1 to 10 wt.-%;

10 and whereby

fraction (iii) has a polystyrene content as determined by ^1H NMR in an amount of less than 5 wt.-%, and whereby

the total polystyrene content as determined by ^1H NMR of fractions (i) to (iii) is from 6 to 14 wt.-%,

15 for blending with of a blend (A) comprising polypropylene, polyethylene, polystyrene, and limonene, having

A-1) a content of isotactic polypropylene of 30 – 70 wt.-%,

A-2) a content of ethylene derived from polyethylene and ethylene containing copolymers of 20 – 50 wt.-%,

20 A-3) 0.5 to 5.0 wt.-% of polystyrene,

A-4) 0 to 3.0 wt.-% stabilizers,

A-5) 0 to 4.0 wt.-% polyamide-6,

A-6) 0 to 3.0 wt.-% talc,

A-7) 0 to 3.0 wt.-% chalk,

25 A-8) 0 to 1.0 wt.-% paper,

A-9) 0 to 1.0 wt.-% wood,

A-10) 0 to 0.5 wt.-% metal,

A-11) 0.1 ppm to 100 ppm of limonene as determined by using solid phase microextraction (HS-SPME-GC-MS), and

A-12) 0 to 200 ppm total fatty acid content as determined using solid phase microextraction (HS-SPME-GC-MS) wherein all amounts are given with respect to the total weight of blend (A)

wherein blend (A) is a recycled material, which is recovered from a waste plastic
5 material derived from post-consumer and/or post-industrial waste; and wherein blend (A) has a melt flow rate (ISO 1133, 2.16 kg, 230°C) of 4.0 to 20.0 g/10min, to obtain an upgraded polyolefin composition, preferably an upgraded polyolefin composition having a tensile modulus of at least 850 MPa (ISO 527-1,2) and a Charpy Impact Strength (ISO 179-1; 1eA, 23°C) of more than 6.0 kJ/m² and up to 10
10 kJ/m².

All preferred aspects, definitions and embodiments as described above shall also hold for the use.

Experimental Section

The following Examples are included to demonstrate certain aspects and
5 embodiments of the invention as described in the claims. It should be appreciated by
those of skill in the art, however, that the following description is illustrative only and
should not be taken in any way as a restriction of the invention.

Test Methods

a) Amount of iPP, Polystyrene, Content of ethylene (and ethylene containing 10 copolymers) and Amount of Polyamide-6

To establish different calibration curves iPP and HDPE and iPP, PS and PA6 were
blended. For the quantification of the content of the foreign polymers, IR spectra were
15 recorded in the solid-state using a Bruker Vertex 70 FTIR spectrometer. Films were
prepared with a compression-moulding device at 190 °C 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
20 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 apodisation.

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
25 content in weight %).

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
30 (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.

b) Amount of Talc and Chalk

5 were measured by Thermogravimetric Analysis (TGA); experiments were performed with a Perkin Elmer TGA 8000. 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 (WCO₂) was assigned to CO₂ evolving from
10 CaCO₃, and therefore the chalk content was evaluated as:

$$\text{Chalk content} = 100/44 \times \text{WCO}_2$$

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 (W_{cb}). Knowing the
15 content of carbon black and chalk, the ash content excluding chalk and carbon black was calculated as:

$$\text{Ash content} = (\text{Ash residue}) - 56/44 \times \text{WCO}_2 - \text{W}_{cb}$$

20 Where Ash residue is the weight% 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.

c) Amount of Paper, Wood

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

d) Amount of Metals

was determined by x ray fluorescence (XRF).

30

e) Amount of Limonene

was determined by solid phase microextraction (HS-SPME-GC-MS).
Additional details are given below with respect to the specific sample.

f) Amount of total fatty acids

- 5 was determined by solid phase microextraction (HS-SPME-GC-MS).
Additional details are given below with respect to the specific sample.

- g) Melt flow rates** were measured with a load of 2.16 kg (MFR₂) at 230 °C or 190°C
as indicated. The melt flow rate is that quantity of polymer in grams which the test
10 apparatus standardized to ISO 1133 extrudes within 10 minutes at a temperature of
230 °C (or 190°C) under a load of 2.16 kg.

**h) Crossfractionation Chromatography (CFC) for compatibilizer (B) (a-TREF
x SEC analysis) for compatibilizer (B)**

- 15 The chemical composition distribution as well as the determination of the molecular
weight distribution and the corresponded molecular weight averages (M_n, M_w and M_v)
at a certain elution temperature (polymer crystallinity in solution) also can be
determined by a fully automated Cross Fractionation Chromatography (CFC) as
described by Ortin A., Monrabal B., Sancho-Tello J., *Macromol. Symp.*, **2007**, 257, 13-
20 28.

A CFC instrument (PolymerChar, Valencia, Spain) was used to perform the cross-
fractionation chromatography (TREF x SEC). A four band IR5 infrared detector
(PolymerChar, Valencia, Spain) was used to monitor the concentration. The polymer
was dissolved at 160°C for 150 minutes at a concentration of around 1mg/ml.

- 25 To avoid injecting possible gels and polymers which do not dissolve in TCB at 160°C,
like PET and PA, the weighed out sample was packed into stainless steel mesh MW
0,077/D 0,05mmm.

- Once the sample was completely dissolved an aliquot of 0.5 ml was loaded into the
TREF column and stabilized for 45 minutes at 110 °C. The polymer was crystallized
30 and precipitate to a temperature of 35°C by applying a constant cooling rate of
0.2 °C/min. A discontinuous elution process was performed using the following

temperature steps: (35, 40, 50, 60, 65, 70, 75, 80, 85, 90, 95, 100, 103, 106, 109, 112, 115, 117, 119, 121, 123, 125, 127, 130, 135 and 140).

In the second dimension, the GPC analysis, 3 PL Olexis columns and 1x Olexis Guard columns from Agilent (Church Stretton, UK) were used as stationary phase. As eluent
 5 1,2,4-trichlorobenzene (TCB, stabilized with 250 mg/L 2,6-Di tert butyl-4-methyl-phenol) at 150 °C and a constant flow rate of 1 mL/min were applied. The column set was calibrated using universal calibration (according to ISO 16014-2:2003) with at least 15 narrow MWD polystyrene (PS) standards in the range of 0,5 kg/mol to 11 500 kg/mol. Following Mark Houwink constants were used to convert PS molecular
 10 weights into the PP molecular weight equivalents.

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

$$K_{PP} = 19 \times 10^{-3} \text{ mL/g}, \quad \alpha_{PP} = 0.725$$

A third order polynomial fit was used to fit the calibration data. Data processing was performed using the software provided from PolymerChar with the CFC instrument.

Calibration of Detector (Calibration of Composition Detector for the Determination of SCB/1000TC)

The IR 5 detector provides different detector signals, which were designated as concentration signal (broad spectral band covering the spectral region from 2800 cm-
 20 1 to 3000 cm-1), methyl (CH3) (narrow band filter centered at 2959 cm-1) and methylene (CH2) (centered at 2928 cm-1) signal. The ratio of the methyl to the methylene detector signals is correlating to the total amount of methylene (CH3) per 1000 carbon atoms (CH3/1000TC) (A. Ortin, B. Monrabal, J. Montesinos, P. del Hierro, *Macromol. Symp.* 2009, 282, 65-70). The determination of the CH3/1000TC
 25 using an IR5 detector can be performed by calibrating the CH3/CH2 ratio versus the nominal CH3/1000TC content. The nominal CH3/1000TC content was obtained by ¹³C-NMR spectroscopy (as described further below). A linear fit was used for this purpose. The calibration set used for this method includes minimum 17 different short chain branched polyethylenes, including polyethylene-co-butene, polyethylene-co-
 30 hexene and polyethylene-co-octene covering an overall branching level up to 80 methyl groups per 1000 carbons (CH3/1000C). The short chain branching was

determined as methyl branching per 1000 total carbons and might be corrected for up to 2 methyl chain end groups per polymer chain.

Preparative Temperature Rising Elution Fractionation (p-TREF) for 5 compatibilizer B

The PP-g-PS polymer of the example (compatibilizer B) was separated according to its chemical composition distribution by preparative Temperature Rising Elution fractionation. The basics of this technique are described by Soares, J.B.P.,
10 Fractionation, In: Encyclopedia Of Polymer Science and Technology, John Wiley & Sons, New York, pp. 75-131, Vol. 10, 2001. The separations were generated using a PREP Mc2 instrument manufactured by PolymerChar S.A. (Valencia, Spain). To avoid injecting possible crosslinked polymers which do not dissolve in TCB at 160°C, the weight out sample was packed into stainless steel mesh (MW 0,077/D
15 0,05mm). Approximately 500 mg of the polymer sample were dissolved in 200 mL 1,2,4-trichlorobenzene (TCB, stabilized with 250 mg/L 2,6-Di tert butyl-4-methyl-phenol) at 160 °C for 60 min. After dissolution the temperature was then rapidly cooled to 122°C and held at 122°C for 10 min for stabilization purpose. Later the temperature was slowly cooled to 40°C under a constant cooling rate (0.1 °C/min).
20 Approximately 200 ml of polymer solution was collected at 50°C, 105°C and 140°C. The polymer was precipitated with around 500 ml cold Methanol (Temperature ~ 8°C). The TCB/Methanol solution was kept overnight in the refrigerator. The filtration step was performed on a vacuum assisted filtration station using a 5.0 µm polytetrafluoroethylene coated filter paper. The filtered fractions were dried over night
25 at 60°C in a vacuum oven and weighted out before further testing.

i) Quantification of microstructure by NMR spectroscopy (for 30 compatibilizer (B); validation experiments)

Quantitative nuclear-magnetic resonance (NMR) spectroscopy was used to quantify the polystyrene content of compatibilizer (B).

Quantitative $^{13}\text{C}\{^1\text{H}\}$ NMR spectra were recorded in the solution-state using a Bruker
5 Avance III 400 NMR spectrometer operating at 400.15 and 100.62 MHz for ^1H and ^{13}C respectively. All spectra were recorded using a ^{13}C optimized 10 mm extended temperature probehead at 125°C using nitrogen gas for all pneumatics. Approximately 200 mg of material was dissolved in 3 ml of 1,2-tetrachloroethane- d_2 (TCE- d_2) using Ditertiarybutylhydroxytoluen (BHT) (CAS 128-37-0) as stabilizer. Standard single-pulse
10 excitation was employed utilizing a 30 degree pulse, a relaxation delay of 1s and 10 Hz sample rotation.

A total of 16 transients were acquired per spectra using 4 dummy scans. A total of 32k data points were collected per FID with a dwell time of 60 micro-seconds, which corresponds to a spectral window of approximately 20 ppm. The FID was then zero
15 filled to 64k data points and an exponential window function applied with 0.3 Hz line-broadening.

Quantitative ^1H NMR were processed, integrated and quantitative properties determined. All chemical shifts were internally referenced to the residual protonated solvent signal at 5.95 ppm.

20 Characteristic signals corresponding to polystyrene and polypropylene were observed and contents calculated. Reference is made to BRANDOLINI, Anita J.; HILLS, Deborah D. *NMR spectra of polymers and polymer additives*. CRC press, 2000.

25 Characteristic signals resulting from the additional use of BHT as stabilizer were also observed. For the BHT compensation, the integral of the signal at 4.83 ppm assigned to the -OH site of BHT was used, accounting for the number of reporting nuclei per molecule.

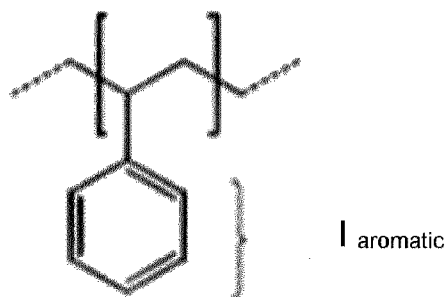
$$\text{BHT} = I_{\text{OH-BHT}}$$

30 Characteristic signals resulting from polystyrene were observed and the content was quantified using the integral of the aromatic signals (I_{aromatic}) between 7.6 ppm and

6.3 ppm assigned to aromatic protons, accounting for the number of reporting nuclei per polystyrene.

Aromatic protons from BHT influencing integral region (I_{aromatic}) must be compensated for

$$5 \quad PS = [I_{\text{aromatic}} - (2 * BHT)] / 5$$



The propylene content was quantified using the integral of the propylene bulk aliphatic (I_{bulk}) signal between 0.00 to 3.00 ppm. This integral included the aliphatic sites from polystyrene (CH and CH₂) and the aliphatic sites from BHT as well. The propylene content was calculated based on the bulk integral and compensated for aliphatic polystyrene signals and BHT, accounting for the number of reporting nuclei per polypropylene.

10

$$PP = [I_{\text{bulk}} - (18 * BHT) - (3 * BHT) - (3 * PS)] / 6$$

The total mole fraction of polystyrene in the polymer was calculated

$$fPS = PS / (PS + PP)$$

15 The total PS content in mole percent was calculated from the mole fraction

$$PS \text{ [mole\%]} = 100 * fPS$$

The total PS content in weight percent was calculated from the mole fraction

$$PS \text{ [wt\%]} = 100 * (fPS * 104.15) / [(fPS * 104.15) + ((1 - fPS) * 42.08)]$$

20 j) Isotacticity

Quantitative nuclear-magnetic resonance (NMR) spectroscopy was used to quantify the isotacticity and tacticity distribution. Quantitative ¹³C{¹H} NMR spectra recorded in the solution-state using a Bruker Avance III 400 NMR spectrometer operating at 400.15 and 100.62 MHz for ¹H and ¹³C respectively. All spectra were recorded using

a ^{13}C optimised 10 mm selective excitation probehead at 125°C using nitrogen gas for all pneumatics. Ideally approximately 200 mg of material was dissolved in 1,2-tetrachloroethane- d_2 (TCE- d_2), if only less amount of material available (e.g. fractions) extended number of transients needed. This setup was chosen primarily for the high resolution needed for tacticity distribution quantification {busico01, busico97}. Standard single-pulse excitation was employed utilising the NOE and bi-level WALTZ16 decoupling scheme {zhou07, busico07}. A total of 6144 (6k) transients respectively 16384 (16k) for extended measurements were acquired per spectra. All chemical shifts are internally referenced to the methyl signal of the isotactic pentad mmmm at 21.85 ppm.

The tacticity distribution was quantified through integration of the methyl region between 23.6 and 19.7 ppm correcting for any sites not related to the stereo sequences of interest {busico01, busico97}.

The pentad tacticity distribution was determined through direct separate integration of each methyl signal from a given steric pentad followed by normalisation to the sum of methyl signals from all steric pentads. The relative content of a specific steric pentad was reported as the mole fraction or percentage of a given steric pentad xxxx with respect to all steric pentads:

$$[\text{xxxx}] = \text{xxxx} / (\text{mmmm} + \text{mmmr} + \text{rmmr} + \text{mmrr} + \text{xmrx} + \text{mrmr} + \text{rrrr} + \text{mrrr} + \text{mrrm})$$

where xmrx represents the combined integral of both mrrm and rmrr as signal from these steric pentads are not commonly resolved. The pentad isotacticity was thus given by:

$$[\text{mmmm}] = \text{mmmm} / (\text{mmmm} + \text{mmmr} + \text{rmmr} + \text{mmrr} + \text{xmrx} + \text{mrmr} + \text{rrrr} + \text{mrrr} + \text{mrrm})$$

The triad tacticity distribution was indirectly determined from the pentad tacticity distribution using the known pentad-triad necessary relationships:

$$[\text{mm}] = [\text{mmmm}] + [\text{mmmr}] + [\text{rmmr}]$$

$$[\text{mr}] = [\text{mmrr}] + [\text{xmrx}] + [\text{mrmr}]$$

$$[\text{rr}] = [\text{rrrr}] + [\text{mrrr}] + [\text{mrrm}]$$

busico01:

Busico, V., Cipullo, R., Prog. Polym. Sci. 26 (2001) 443

busico97:

Busico, V., Cipullo, R., Monaco, G., Vacatello, M., Segre, A.L., Macromolecules 30 (1997) 6251

5 zhou07:

Zhou, Z., Kuemmerle, R., Qiu, X., Redwine, D., Cong, R., Taha, A., Baugh, D. Winniford, B., J. Mag. Reson. 187 (2007) 225

busico07:

10 Busico, V., Carbonniere, P., Cipullo, R., Pellecchia, R., Severn, J., Talarico, G., Macromol. Rapid Commun. 2007, 28, 1128

k) Tensile modulus was measured according to ISO 527-2 (cross head speed = 1 mm/min; test speed 50 mm/min at 23 °C) using compression moulded specimens as
15 described in EN ISO 1873-2 (dog bone shape, 4 mm thickness). The measurement was done after 96 h conditioning time of the specimen.

l) Impact strength was determined as Charpy Notched Impact Strength according to ISO 179-1 eA at +23 °C on injection moulded specimens of 80 x 10 x 4 mm
20 prepared according to EN ISO 1873-2. According to this standard samples are tested after 96 hours.

Experiments

A number of blends was produced with DIPOLEN S as blend (A), a polyethylene-polypropylene blend from Mtm Plastics GmbH, materials according to the August 2018 specifications.

	Dipolen S-sample	
	Polypropylene	59 wt.-%
10	Polyethylene	33 wt.-%
	Polystyrene	2.3 wt.-%
	Polyamide-6	2.9 wt.-%
	Talc content	0.4 wt.-%
	Chalk content	0.2 wt.-%
15	Paper content	< 1.0 wt.-%
	Wood content	< 1.0 wt.-%
	Metal content	< 0.2 wt.-%
	Limonene content	32 mg/kg
	Total fatty acid content	71 mg/kg

20

Compatibilizer SCONA TPPP 1616 FA (a SEBS-g-PS, commercially available from Byk Kometra; Preparative Temperature Rising Elution Fractionation (p-TREF) using TCB as eluent and analysis as described above)

	fraction (i) eluting up to 50°C	10.2 wt.-%
25	fraction (ii) eluting from 50°C to 105°C	9.5 wt.-%
	fraction (iii) eluting above 105°C	80.3 wt.-%

whereby

- fraction (i), polystyrene content as determined by ^1H NMR : 59.6 wt.-%;
fraction (ii), polystyrene content as determined by ^1H NMR: 6.0 wt.-%;
30 fraction (iii), polystyrene content as determined by ^1H NMR: 1.9 wt.-%,
the total polystyrene content by ^1H NMR of fractions (i-iii): 8.3 wt.-%

Comparative example 1:

For CE1, no compatibilizer was used, therefore the composition consists of DIPOLEN
5 S.

Comparative examples 2-4:

For CE2 to CE4, the compatibilizer Queo 8207 (an ethylene based 1-octene
10 plastomer, commercially available from Borealis AG) was used in amounts of 5 wt.-
%, 10 wt.-%, and 20 wt.-% (In CE2, CE3 and CE4 respectively)

Inventive examples:

15 In each of the inventive examples IE1 to IE3, SCONA TPPP 1616 FA (a SEBS-g-PS,
commercially available from Byk Kometra) was used as compatibilizer (B), in
amounts disclosed in Table 3.

Additionally, inventive examples IE1 to IE3 contained Irganox B225F in 0.3 wt.-%,
20 (See Table 3) as stabiliser.

The compositions were prepared via melt blending on a co-rotating twin screw
extruder.

25 The polymer melt mixture was discharged and pelletized. For testing the mechanical
properties, specimens were produced and tested according to ISO 179 with 1eA
notched specimens to measure the Charpy notched impact strength and according to
ISO 527-1/2 with 1A specimens to measure the tensile properties at room
temperature.

30

Limonene content in DIPOLEN - details

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

5

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, 1mg/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 microextraction 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:

20 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
25 Acquisition: SIM scan mode
Scan parameter: 20-300 amu
SIM Parameter: m/Z 93, 100 ms dwell time

Table 1: Limonene content in DIPOLEN (Blend (A))

Sample	Limonen [mg/ kg] HS-SPME-GC-MS ¹
Dipolen S	31.5±2.6

¹Headspace Solidphase Microextraction. Materials available from mtm plastics GmbH, according to 2018 specifications.

5

Total free fatty acid content

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

10

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.

20

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)

25

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

Table 2: Total fatty acid content in Dipolen (Blend (A))

Sample	Total fatty acid concentration [mg/kg] ¹
Dipolen S	70.6

¹The concentration of acetic acid, propionic acid, butyric acid, pentanoic acid, hexanoic acid octanoic acid, nonanoic acid and decanoic acid in each sample was

5 added together to give a totally fatty acid concentration value.

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Table 3: Results

		CE1	CE2	CE3	CE4	IE1	IE2	IE3
DIPOLEN S (blend A)	wt.-%	100	95	90	80	99.2	98.2	96.7
Queo 8207 (comparative compatibilizer)	wt.-%		5	10	20			
SCONA TPPP 1616 FA (compatibilizer B)	wt.-%					0.5	1.5	3.0
Irganox B225F	wt.-%					0.3	0.3	0.3
MFR (2.16 kg, 230 °C)	g/10min	6	5.9	6.2	6.8	5.5	5.6	5.7
Nominal tensile strain at break (ISO 527-1)	%	110.9	230	286	434	70	69	68
Tensile Modulus	MPa	980	798	745	602	953	951	963
Impact strength @ 23°C (ISO 189-1)	kJ/m ²	5.9	6.4	8.2	30.2	7.0	6.9	6.5
Failure mode			C	C	P			
Impact strength @ -30°C	kJ/m ²	2.1				1.8	1.8	1.8

As can be seen from the table, the inventive examples display improved impact properties over DIPOLEN S (CE1). Furthermore, the amount of compatibilizer required is far lower for the inventive examples (using the compatibilizer of the present invention) than for comparative examples CE2 to CE4 (using Queo 8207).
5 Compare for example IE1 and CE2; an impact strength of 7.0 kJ/m² is observed for IE1, with the addition of just 0.5 wt.-% of compatibilizer, in comparison to CE2, which achieves an impact strength of 6.4 kJ/m² whilst using 5.0 wt.-% of compatibilizer. Consequently the tensile properties of the resulting composition are much improved
10 (compare IE1 to IE3 with CE1 to CE3).

CLAIMS:

1. A polyolefin composition obtained by blending
- 5 a) 96.0 to 99.7 wt.-% of a blend (A) comprising polypropylene, polyethylene, polystyrene, and limonene, having
- A-1) a content of isotactic polypropylene of 30 – 70 wt.-%,
- A-2) a content of ethylene derived from polyethylene and ethylene containing copolymers of 20 – 50 wt.-%,
- A-3) 0.5 to 5.0 wt.-% of polystyrene,
- 10 A-4) 0 to 3.0 wt.-% stabilizers,
- A-5) 0 to 4.0 wt.-% polyamide-6,
- A-6) 0 to 3.0 wt.-% talc,
- A-7) 0 to 3.0 wt.-% chalk,
- A-8) 0 to 1.0 wt.-% paper,
- 15 A-9) 0 to 1.0 wt.-% wood,
- A-10) 0 to 0.5 wt.-% metal,
- A-11) 0.1 ppm to 100 ppm of limonene as determined by using solid phase microextraction (HS-SPME-GC-MS), and
- A-12) 0 to 200 ppm total fatty acid content as determined by using
- 20 solid phase microextraction (HS-SPME-GC-MS)
- wherein all amounts are given with respect to the total weight of blend (A),
- wherein blend (A) is a recycled material, which is recovered from a waste plastic material derived from post-consumer and/or post-industrial
- 25 waste; and wherein blend (A) has a melt flow rate (ISO 1133, 2.16 kg, 230°C) of 4.0 to 20.0 g/10min;
- and
- b) 0.3 to 4.0 wt.-% of a compatibilizer (B)
- being a styrene-grafted polypropylene having
- 30 a melt flow rate (ISO 1133, 2.16 kg, 190°C) of 4.0 to 20.0 g/10min, and three fractions when being subjected Preparative Temperature Rising Elution Fractionation (p-TREF) using TCB as eluent, in amounts of

fraction (i) eluting up to 50°C 5 to 15 wt.-%
fraction (ii) eluting from 50°C to 105°C 5 to 15 wt.-% and
fraction (iii) eluting above 105°C 70 to 90 wt.-%,

whereby

5 fraction (i) has a polystyrene content as determined by ¹H NMR in an amount of 50 to 70 wt.-%;

and whereby

fraction (ii) has a polystyrene content as determined by ¹H NMR in an amount of 1 to 10 wt.-%;

10 and whereby

fraction (iii) has a polystyrene content as determined by ¹H NMR in an amount of less than 5 wt.-%, and whereby

the total polystyrene content as determined by ¹H NMR of fractions (i) to (iii) is from 6 to 14 wt.-%, and

15 whereby the polyolefin composition has a tensile modulus of at least 850 MPa (ISO 527-1,2) when measured on an injection molded test specimen.

2. The polyolefin composition according to claim 1, whereby the compatibilizer
20 (B) has a total polystyrene content as determined by ¹H NMR of fractions (i) to (iii) of 8 to 12 wt.-% as determined by ¹H NMR.

3. The polyolefin composition according to claim 1 or 2, whereby the
compatibilizer (B) has a melt flow rate (ISO 1133, 2.16 kg, 190°C) of 8.0 to
25 16.0 g/10min.

4. The polyolefin composition according to any one of claims 1 to 3, whereby the
ratio of the melt flow rate (ISO 1133, 2.16 kg, 190°C) of the compatibilizer (B)
to the melt flow rate of the blend (A) (ISO 1133, 2.16 kg, 230°C)
30 MFR(B, (ISO 1133, 2.16 kg, 190°C) / MFR (A, ISO 1133, 2.16 kg, 230°C) is in the range of 0.5 to 2.0.

5. The polyolefin composition according to any one of claims 1 to 4, whereby blend (A) has a content of limonene as determined by using solid phase microextraction (HS-SPME-GC-MS) of from 0.1 ppm to 50 ppm.
- 5 6. The polyolefin composition according to claim 5, wherein the content of limonene is from 0.1 ppm to 30 ppm.
7. The polyolefin composition according to claim 5, wherein the content of limonene is from 0.1 ppm to 10 ppm.
- 10 8. The polyolefin composition according to any one of claims 1 to 7, whereby blend (A) has a content of fatty acids as determined by using solid phase microextraction (HS-SPME-GC-MS) of from 1 ppm to 150 ppm.
- 15 9. The polyolefin composition according to claim 8, wherein the content of fatty acids is 2 ppm to 100 ppm.
10. The polyolefin composition according to claim 8, wherein the content of fatty acids is 3 ppm to 80 ppm.
- 20 11. The polyolefin composition according to any one of claims 1 to 10 having a melt flow rate (ISO 1133, 2.16 kg, 230°C) of 3.0 to 10.0 g/10min.
12. The polyolefin composition according to any one of claims 1 to 11, whereby the fractions (i) to (iii) of compatibilizer (B) fulfill the following one or more criteria:
25 isotacticity of 30 to 60 mmmm% ¹³C NMR in fraction (i) eluting up to 50°C
and/or
isotacticity of 70 to 90 mmmm% ¹³C NMR in fraction (ii) eluting from 50°C to 105°C
30 and/or
isotacticity of 90 to 99 mmmm% ¹³C NMR in fraction (iii) eluting above 105°C.

13. The polyolefin composition according to any one of claims 1 to 12, having a Charpy Impact Strength (ISO 179-1; 1eA, 23°C) of more than 6.0 kJ/m² and up to 10.0 kJ/m².
- 5 14. An article, comprising the polyolefin composition according to any one of claims 1 to 13.
15. A process for preparing the polyolefin composition according to any one of claims 1 to 13, comprising the steps of
- 10 a) providing the blend (A) in an amount of 96.0 to 99.7 wt.-%, based on the overall weight of the polyolefin composition
- b) providing the compatibilizer (B) in an amount of 0.3 to 4.0 wt.-%, based on the overall weight of the polyolefin composition
- c) melting and mixing the blend of blend (A) and the compatibilizer (B) in an
- 15 extruder, and
- d) optionally pelletizing the obtained polyolefin composition.
16. A use of a compatibilizer (B) being a styrene-grafted polypropylene having
- a) a melt flow rate (ISO 1133, 2.16 kg, 190°C) of 4.0 to 20 g/10min, and
- 20 b) three fractions when being subjected to preparative temperature rising elution fractionation (p-TREF) using TCB as eluent, in amounts of
- | | |
|--|-------------------|
| fraction (i) eluting up to 50°C | 5 to 15 wt.-% |
| fraction (ii) eluting from 50°C to 105°C | 5 to 15 wt.-% and |
| 25 fraction (iii) eluting above 105°C | 70 to 90 wt.-%, |
- whereby
- fraction (i) has a polystyrene content as determined by ¹H NMR in an amount of 50 to 70 wt.-%;
- and whereby
- 30 fraction (ii) has a polystyrene content as determined by ¹H NMR in an amount of 1 to 10 wt.-%;
- and whereby

fraction (iii) has a polystyrene content as determined by ^1H NMR in an amount of less than 5 wt.-%, and whereby

the total polystyrene content as determined by ^1H NMR of fractions (i) to (iii) is from 6 to 14 wt.-%,

5 for blending with of a blend (A) comprising polypropylene, polyethylene, polystyrene, and limonene, having

A-1) a content of isotactic polypropylene of 30 – 70 wt.-%,

A-2) a content of ethylene derived from polyethylene and ethylene containing copolymers of 20 – 50 wt.-%,

10 A-3) 0.5 to 5.0 wt.-% of polystyrene,

A-4) 0 to 3.0 wt.-% stabilizers,

A-5) 0 to 4.0 wt.-% polyamide-6,

A-6) 0 to 3.0 wt.-% talc,

A-7) 0 to 3.0 wt.-% chalk,

15 A-8) 0 to 1.0 wt.-% paper,

A-9) 0 to 1.0 wt.-% wood,

A-10) 0 to 0.5 wt.-% metal,

A-11) 0.1 ppm to 100 ppm of limonene as determined by using solid phase microextraction (HS-SPME-GC-MS), and

20 A-12) 0 to 200 ppm total fatty acid content using solid phase microextraction (HS-SPME-GC-MS) wherein all amounts are given with respect to the total weight of blend (A)

wherein blend (A) is a recycled material, which is recovered from a waste plastic material derived from post-consumer and/or post-industrial waste; and wherein

25 blend (A) has a melt flow rate (ISO 1133, 2.16 kg, 230°C) of 4 to 20 g/10min, to obtain an upgraded polyolefin composition.

17. Use according to claim 16, wherein the upgraded polyolefin composition has a tensile modulus of at least 850 MPa (ISO 527-1,2) and/or a Charpy Impact
30 Strength (ISO 179-1; 1eA, 23°C) of more than 6.0 kJ/m² and up to 10 kJ/m².