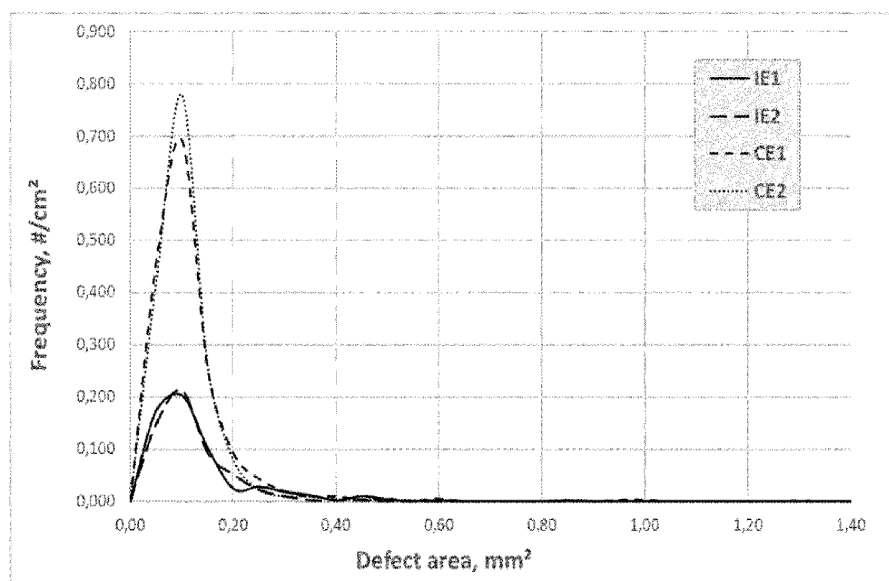




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(54) Title: MIXED-PLASTICS-POLYPROPYLENE BLEND

Figure 1



(57) Abstract: Mixed-plastics polypropylene blend including mainly polypropylene being benzene free with defined CIELAB color.

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Mixed-Plastics-Polypropylene Blend

Field of the Invention

The present invention relates to mixed-plastics polypropylene blends as typically
5 originating from recyclates.

Background

Many attempts have been made for purifying recycling streams as originating
from post-consumer trash. Among those measures washing, sieving, aeration,
10 distillation and the like may be mentioned. For example, WO2018046578
discloses a process for the production of polyolefin recyclates from mixed color
polyolefin waste including packaging waste comprising cold washing the waste
with water followed by washing with an alkali medium at 60 °C, followed by flake
color sorting to receive color sorted (white, transparent, other colors) mono
15 polyolefin rich fractions. Those fractions are then treated at 50 – 155 °C.
US5767230A describes a process comprising contacting PCR polyolefin chips
containing volatile impurities with a heated gas at a superficial velocity sufficient
to substantially reduce the volatile impurities such as odor active substances.
However, up to now contamination by residual amounts of benzene turned out to
20 be a problem. The origin of residual amounts of benzene in post-consumer
recyclates is still dubious but constitutes a hurdle for end-uses in fields such as
medical packaging, food packaging and the like. Residual amounts, i.e. traces of
benzene constitutes a particularly problem as odor tests by sniffing experiments
become impossible. Thus, end-uses having certain demands as to odor are
25 blocked. Color is still a remaining problem not completely addressed. Many re-
use applications require material being close to what is usually denoted white
color. As yet a further problem known recyclates suffer from moderate
homogeneity as reflected by surface contamination occurring in injection molded
products. A specific demand exists for recyclates suitable for injection molding
30 for dosing caps, toileteries, screw caps, caps and closures.

Thus, the problem of providing a more valuable polypropylene blend remains.

Summary of the Invention

The present invention provides a mixed-plastics polypropylene blend having

- (i) a crystalline fraction (CF) content determined according to CRYSTEX
 5 QC analysis in the range from 86.0 to 94.0 wt.-%, and
- (ii) a soluble fraction (SF) content determined according to CRYSTEX
 QC analysis in the range from 6.0 to 14.0 wt.-%, whereby
- (iii) said crystalline fraction (CF) has a propylene content (C3(CF)) as
 10 determined by FT-IR spectroscopy calibrated by quantitative ¹³C-
 NMR spectroscopy, in the range from 95.0 to 99.0 wt.-%; preferably
 96.0 to 98.0 wt.-% and whereby
- (iv) said crystalline fraction (CF) has an ethylene content (C2(CF)), as
 15 determined by FT-IR spectroscopy calibrated by quantitative ¹³C-
 NMR spectroscopy, in the range from 1.0 to 5.0 wt.-% preferably 2.0
 to 4.0 wt.-% and more preferably 2.5 to 3.5 wt.-%; and
- (v) said soluble fraction (SF) has an intrinsic viscosity (iV(SF)) in the
 range from 1.10 to below 1.50 dl/g, preferably 1.25 to 1.45 dl/g; and

whereby

- (vi) the mixed-plastics polypropylene blend has inorganic residues as
 20 measured by calcination analysis (TGA) according to DIN ISO
 1172:1996 of 0.05 to 3.0 wt.-%, preferably 0.05 to 2.5 wt.-%,
 optionally 1.0 to 2.5 wt.-% with respect to the mixed-plastics
 polypropylene blend; and

whereby

- 25 (vii) the mixed-plastics polypropylene blend does not contain benzene
 above the detection limit of HS GC-MS 80°C/2h; and

whereby

- (viii) the mixed-plastics polypropylene blend has a CIELAB color space
 (L*a*b*) of
- 30 - L* from 72.0 to 97.0, preferably from 80.0 to 97.0;
- a* from -5.0 to 0.0;

- b^* from 0.0 to below 22.0; and

whereby

- (ix) the mixed-plastics polypropylene blend has a Large Amplitude Oscillatory Shear – Non-Linear Factor [LAOS –NLF] (190°C, 1000%)
5 higher than 2.3, whereby

$$\text{LAOS – NLF} = \left| \frac{G'_1}{G'_3} \right|$$

whereby

G'_1 is the first order Fourier Coefficient

G'_3 is the third order Fourier Coefficient.

10

The present invention is also concerned with the mixed-plastics polypropylene blend in pellet form and mixed-plastics polypropylene blend being visbroken by peroxides. The present invention further provides articles made from the mixed-plastics polypropylene blend and a use for packaging and/or in the medical field.

- 15 In yet a further aspect, the present invention concerns blends of the mixed-plastics polypropylene blend with at least one virgin polyolefin.

Mixed plastics is defined as the presence of low amounts of compounds usually not found in virgin polypropylene blends such as polystyrenes, polyamides,
20 polyesters, wood, paper, limonene, aldehydes, ketones, fatty acids, metals, and/or long term decomposition products of stabilizers. Virgin polypropylene blends denote blends as directly originating from the production process without intermediate use.

25 As a matter of definition, “mixed plastics” can be equated with detectable amounts of polystyrene and/or polyamide-6 and/or limonene and/or fatty acids.

The mixed-plastics polypropylene blend further has a broadened molecular weight distribution because it is a mechanical blend of countless polypropylenes and some very minor amount of low density polyethylene as well as linear low density polyethylenes. It will be understood by those skilled in the art that
30 polypropylenes from various manufactures end up in plastic trash streams particularly when presorted in polyolefin-rich plastic trash streams.

It further will be understood by those skilled in the art that a soluble fraction (SF) as obtained by CRYSTEX QC analysis having an intrinsic viscosity (iV(SF)) in the range from 1.10 to below 1.50 dl/g is typically found in material from recycling streams. In a preferred aspect of the invention the soluble fraction (SF) as
 5 obtained by CRYSTEX QC analysis has an intrinsic viscosity (iV(SF)) in the range from 1.25 to below 1.45 dl/g.

It has been surprisingly found that the mixed-plastics polypropylene blend
 10 according to the present invention provides better surface properties enabling numerous demanding end-use applications.

In a first embodiment, it is preferred that the amounts of the crystalline fraction (CF) and soluble fraction (SF) in CRYSTEX QC analysis are:

15 87.0 to 90.0 wt.-% crystalline fraction (CF) content and
 10.0 to 13.0 wt.-%, soluble fraction (SF) content.

In a second embodiment the amounts of the crystalline fraction (CF) and soluble fraction (SF) in CRYSTEX QC analysis are:

20 91.0 to 94.0 wt.-% crystalline fraction (CF) content and
 6.0 to 9.0 wt.-%, soluble fraction (SF) content.

In the first embodiment the mixed-plastics polypropylene blend has a CIELAB color space ($L^*a^*b^*$) of

- L^* from 85.0 to 97.0;
- 25 - a^* from -5.0 to 0.0;
- b^* from 0.0 to below 8.0.

In the second embodiment the mixed-plastics polypropylene blend has a CIELAB color space ($L^*a^*b^*$) of

- 30 - L^* from 72.0 to 97.0, preferably from 80.0 to 97.0;
- a^* from -5.0 to 0.0;
- b^* from 0.0 to below 22.0, usually from above 8.0 to below 22.0

The CIELAB color can be influenced by the sorting process. The more yellowish material is accepted, the higher b^* .

5 The mixed-plastics polypropylene blend according to the present invention typically has a melt flow rate (ISO1133, 2.16kg; 230°C) of 2.0 to 100 g/10min. The melt flow rate can be influenced by splitting post-consumer plastic waste streams, for example, but not limited to: originating from extended producer's responsibility schemes, like from the German DSD, or sorted out of municipal solid waste into a high number of pre-sorted fractions and recombine them in an
10 adequate way. As a further way of modifying melt flow rate of the final mixed-plastics polypropylene blend peroxides can be introduced in the final pelletization step. Usually MFR ranges from 2.0 to 100 g/10min, preferably from 5.0 to 80 g/10min, more preferably from 10 to 60 g/10min. and most preferably from 12 to 55 g/10min.

15 The MFR of the second embodiment preferably ranges from 2.0 to 12 g/10 min (ISO1133, 2.16kg; 230°C). This MFR range particularly holds for the non-visbroken mixed-plastics polypropylene blend. Visbreaking allows increase of MFR to 30 g/10 min also for the second embodiment.

20 Usually the mixed-plastics polypropylene blend according to the present invention will be a recycled material.

Typically, the recycling nature can be assessed by the presence of one or more of the following substances:

- a) polystyrene
- b) polyamide-6
- 25 c) limonene as determined by using solid phase microextraction (HS-SPME-GC-MS)
- d) fatty acids as determined by using solid phase microextraction (HS-SPME-GC-MS).

30 Presence means detectable limits. The detection limit for limonene and fatty acids in solid phase microextraction (HS-SPME-GC-MS) is below 0.1 ppm, i.e. traces of these substances easily allow figuring out recycling nature.

The following amounts are preferred

- a) polystyrene: 0 to 2.0 wt.-%; more preferred 0 to 0.5 wt.-%
- b) polyamide-6: 0 to 1.5 wt.-%; more preferred 0 to 0.5 wt.-%
- c) limonene as determined by using solid phase microextraction (HS-
5 SPME-GC-MS): 0.1 ppm to 50 ppm
- d) fatty acids as determined by using solid phase microextraction (HS-
SPME-GC-MS): 0.1 ppm to 200 ppm, more preferably 50 ppm.

It goes without saying that the amounts of a), b), c) and d) should be as low as possible. In a specifically preferred embodiment, the mixed-plastics
10 polypropylene blend is free of polystyrene and is free of polyamide meaning both polymers are below the detection limit.

The mixed-plastics polypropylene blend according to the present invention preferably has a soluble fraction (SF) obtained by CRYSTEX QC analysis with a content of ethylene (C2(SF)), as determined by FT-IR spectroscopy calibrated
15 by quantitative ¹³C-NMR spectroscopy, in the range from 12.0 to 32.0 wt.-%.

In the first embodiment, the mixed-plastics polypropylene blend according to the present invention preferably has a soluble fraction (SF) obtained by CRYSTEX QC analysis with a content of ethylene (C2(SF)), as determined by FT-IR spectroscopy calibrated by quantitative ¹³C-NMR spectroscopy, in the range
20 from 25.0 to 32.0 wt.-%.

In the second embodiment, the mixed-plastics polypropylene blend according to the present invention preferably has a soluble fraction (SF) obtained by CRYSTEX QC analysis with a content of ethylene (C2(SF)), as determined by FT-IR spectroscopy calibrated by quantitative ¹³C-NMR spectroscopy, in the
25 range from 10.0 to 25.0 wt.-%, more preferably 12.0 to 25.0 wt.-%, even more preferably 12.0 to 20.0 wt.-% and most preferably 14.0 to 19.0 wt.-%.

The mixed-plastics polypropylene blend according to the present invention is preferably characterized by an odor (VDA270-B3) of 4.0 or lower, preferably 3.0 or lower. It should be understood that many commercial recycling grades which
30 do not report odor are in fact even worse as an odor test under VDA270 is forbidden due to the presence of problematic substances.

In a further aspect the mixed-plastics polypropylene blend according to present invention, particularly of the first embodiment, has a Large Amplitude Oscillatory Shear – Non-Linear Factor (LAOS –NLF) (190°C; 1000%) preferably higher than 2.7, more preferably higher than 3.3, whereby

$$5 \quad \text{LAOS – NLF} = \left| \frac{G'_1}{G'_3} \right|$$

whereby

G'_1 is the first order Fourier Coefficient

G'_3 is the third order Fourier Coefficient

In the second embodiment, the Large Amplitude Oscillatory Shear – Non-Linear Factor (LAOS –NLF) (190°C; 1000%) is merely higher than 2.3.

Without wishing to be bound by theory, it is believed that the processing of the polymer contributes to branching triggered by enclosed contaminants. The LAOS-NLF may be influenced by selecting feedstock such that about 10 wt.-% of the material is soft polypropylene. In this respect, “soft polypropylene” means a
 15 tensile modulus (measured as described in the experimental part) of below 900 MPa. The incorporation of low density polyethylenes and linear low density polyethylenes, more precisely the presence of low crystalline PE Fraction (LCF-PE) as observed in CFC analysis in an amount of 2.0 to 4.0 wt.-% also contributes to the Large Amplitude Oscillatory Shear – Non-Linear Factor (LAOS
 20 –NLF) (190°C; 1000%) of higher than 2.3. It should be understood that several regions operate collection stations collecting highly consumer pre-sorted plastics. Such highly valuable plastics streams are commercially available and allow upgrading of other low quality streams (such as by a softer polypropylene mixture) from other waste disposal resources. The second embodiment having
 25 higher amount of crystalline fraction (CF), i.e. 91.0 to 94.0 wt.-% crystalline fraction (CF) content and 6.0 to 9.0 wt.-%, soluble fraction (SF) content, is more limited, whereby the Large Amplitude Oscillatory Shear – Non-Linear Factor (LAOS –NLF) (190°C; 1000%) is somewhat lower.

In yet a further aspect, the mixed-plastics polypropylene blend according to the
 30 first embodiment of the present invention has a tensile modulus (ISO 527-2 at a cross head speed of 1 mm/min; 23°C) using injection molded specimens as

described in EN ISO 1873-2 (dog bone shape, 4 mm thickness) of at least 1300 MPa, preferably at least 1350 MPa, most preferably at least 1390 MPa. Such relatively high tensile modulus results from the relatively low amounts of rubber like and plastomer like materials. Usually the tensile modulus (ISO 527-2 at a cross head speed of 1 mm/min; 23°C) of the first embodiment will not be higher than 1500 MPa.

The mixed-plastics polypropylene blend according to the second embodiment of the present invention has a tensile modulus (ISO 527-2 at a cross head speed of 1 mm/min; 23°C) using injection molded specimens as described in EN ISO 1873-2 (dog bone shape, 4 mm thickness) of at least 1200 MPa, preferably at least 1250 MPa. Usually the tensile modulus (ISO 527-2 at a cross head speed of 1 mm/min; 23°C) of the second embodiment will not be higher than 1400 MPa.

The mixed-plastics polypropylene blend according to the first embodiment of the present invention surprisingly results in exceptional time stability as to melt flow rate, η_a (2.7kPa) and η_a (300 rad/s) As yet a further and surprising advantage an exceptionally high melt strength was observed. Further surprisingly it turned out there is exceptionally good homogeneity of the mixed-plastics polypropylene blend according to the present invention. For example, different (selected) pellets of one batch showed essentially the same values of melt flow rate, η_a (2.7kPa) and η_a (300 rad/s) when analyzed separately. This is an exceptional finding since substantial variation is expected with recycled materials.

The mixed-plastics polypropylene blend according to the second embodiment turned out to have excellent processability reflected by a shear thinning factor (STF) being the ratio of η_a 0.05 and η_a 300 of above 13.0.

Charpy notched impact strength (non-instrumented, ISO 179-1 at +23 °C) of the mixed-plastics polypropylene blend according to the present invention is preferably higher than 4.0 kJ/m² , more preferably higher than 4.5 kJ/m². The Charpy notched impact strength (non-instrumented, ISO 179-1 at +23 °C) of the mixed-plastics polypropylene blend according to the second embodiment is

preferably higher than 6.0 kJ/m², more preferably higher than 8.0 kJ/m², most preferably higher than 8.3 kJ/m².

In specifically preferred embodiment, the mixed-plastics polypropylene blend according to present invention has a notched Charpy impact strength (NIS) (1eA) (non-instrumented, ISO 179-1 at +23 °C) according to ISO 179-1 eA at +23 °C on injection moulded specimens of 80 x 10 x 4 mm prepared according to EN ISO 1873-2 of at least 8.0 kJ/m², preferably 8.3 kJ/m², whereby further said soluble fraction (SF) obtained by CRYSTEX QC analysis has an ethylene content (C2(SF)), as determined by FT-IR spectroscopy calibrated by quantitative ¹³C-NMR spectroscopy, in the range from 12.0 to 20.0 wt.-% and further preferably the mixed-plastics polypropylene blend has a CIELAB color space (L*a*b) of

- L* from 72.0 to 97.0, preferably from 80.0 to 97.0;
- a* from -5.0 to 0.0;
- b* from 0.0 to below 22.0

In this specifically preferred aspect of the second embodiment, the crystalline fraction (CF) content determined according to CRYSTEX QC analysis is preferably in the range from 91.0 to 94.0 wt.-% and the soluble fraction (SF) content determined according to CRYSTEX QC analysis is preferably in the range of 6.0 to 9.0 wt.-%.

The mixed-plastics polypropylene blend according to the present invention, i.e. first and second embodiment, is preferably present in the form of pellets. Pelletization contributes to the low amounts of volatile substances.

In an embodiment, the mixed-plastics polypropylene blend according to the present invention is visbroken by one or more peroxides. The mixed-plastics polypropylene blend can be subjected to visbreaking as any other virgin polypropylene blend. If the mixed-plastics polypropylene blend according to the present invention has been subjected to visbreaking, the decomposition products of the visbreaking process can be found in the resulting blend. It should be understood that decomposition products of visbreaking process (as

commonly used in the art for virgin materials) are not considered as impurities. Visbreaking can be done with the first and the second embodiment.

In a further aspect, the present invention concerns a molded article made from the mixed-plastics polypropylene blend as described herein. This applies to the
5 first and the second embodiment.

In yet a further aspect, the present invention concerns blends containing the mixed-plastics polypropylene blend as described herein and at least one virgin polyolefin. Again, this applies for the first and second embodiment. For example, virgin polypropylene homopolymer as contained in heterophasic
10 polypropylenes can be substituted by the mixed-plastics polypropylene blend as described herein.

The present invention also pertains to the use of the mixed-plastics polypropylene blend according to present invention, i.e. the first and the second embodiment, for packaging and/or in the medical field.

15

Detailed description

The process for providing the mixed-plastics polypropylene blend according to the present invention is pretty demanding. The process comprises the following steps:

- 20 a) providing post-consumer plastic trash;
- b) sorting out goods made from polystyrene, polyamide, polyethylene, metals, paper, wood and other non-polypropylene materials thereby providing a post-consumer plastic material;
- 25 c) sorting out colored goods thereby providing a post-consumer plastic material containing mainly white bottles, mainly white yoghurt cups, mainly white cans, mainly colorless panels, mainly colorless component parts and the like;
- d) subjecting the selected post-consumer plastic material having mainly white color or being colorless to milling, washing in an aqueous solution
30 with various detergents and subsequent drying, windsifting and screening;

- e) subjecting the pretreated post-consumer plastic material to a further sorting for eliminating non-polyolefin and colored parts yielding an intermediate;
- f) quality control step, wherein the intermediate from step e) is subjected to Cross Fractionation Chromatography and whereby determining the low-crystalline-polyethylene fraction (LCF-PE) in CFC and double-checking whether said amount falls into the range of 2.0 to 4.0 wt.-%;
- g) discarding intermediate not having a low-crystalline-polyethylene fraction in the range of 2.8 to 4.2 wt.-%;
- h) extruding the material and yielding the polypropylene blend according to the present invention in the form of pellets;
- i) optional aeration which is preferably carried out at a temperature in a range of 100-130°C by preheating the post-consumer plastic material to such temperature using an air stream having a temperature of at least 100°C.

Several possible feedstocks from both separate collection systems (e.g. from extended producer responsibility schemes) as well as particularly municipal trash collection systems are commercially available and allow providing post-consumer plastic trash. Depending on the participation of the consumer and the quality of the sorting plants involved, the purity of those feedstocks will differ which is usually indicated by the collecting systems. It is further possible to screen the intermediate after step b) for the presence of apparently very old ('ancient') mainly colorless / natural plastic articles. Discoloration (e.g. pronounced yellowing) and/or pronounced scratches of the mainly colorless / natural plastic articles allow the sorting. Such step makes it possible to get rid of so-called substances of very high concern. Those substances such as Pb, Hg, polybrominated diphenyl ethers, and the like have been banned for quite some time but are still present in the real world as consumers tend to stockpile plastic articles e.g. in the form of plastic toys for many years and eventually throw them away into collection systems. The additional screen step can be assisted by analysis controls for said substances of very high concern.

Odor control and assessment is possible by a number of methods. An overview is provided inter alia by Demets, Ruben, et al. "Development and application of an analytical method to quantify odour removal in plastic waste recycling processes." *Resources, Conservation and Recycling* 161 (2020): 104907 being
5 incorporated by reference herewith.

Experimental

The following Examples are included to demonstrate certain aspects and embodiments of the invention as described in the claims. It should be appreciated
10 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) CRYSTEX

15 Determination of Crystalline and soluble fractions and their respective properties (IV and Ethylene content)

The crystalline (CF) and soluble fractions (SF) of the polypropylene (PP) compositions as well as the comonomer content and intrinsic viscosities of the respective fractions were analyzed by use of the CRYSTEX instrument, Polymer
20 Char (Valencia, Spain). Details of the technique and the method can be found in literature (Ljiljana Jeremic, Andreas Albrecht, Martina Sandholzer & Markus Gahleitner (2020) Rapid characterization of high-impact ethylene-propylene copolymer composition by crystallization extraction separation: comparability to standard separation methods, *International Journal of Polymer Analysis and*
25 *Characterization*, 25:8, 581-596)

The crystalline and amorphous fractions are separated through temperature cycles of dissolution at 160°C, crystallization at 40°C and re-dissolution in 1,2,4-trichlorobenzene at 160°C. Quantification of SF and CF and determination of ethylene content (C2) are achieved by means of an integrated infrared detector
30 (IR4) and for the determination of the intrinsic viscosity (IV) an online 2-capillary viscometer is used.

The IR4 detector is a multiple wavelength detector measuring IR absorbance at two different bands (CH₃ stretching vibration (centred at app. 2960 cm⁻¹) and the CH stretching vibration (2700-3000 cm⁻¹) that are serving for the determination of the concentration and the Ethylene content in Ethylene-Propylene copolymers. The IR4 detector is calibrated with series of 8 EP copolymers with known Ethylene content in the range of 2 wt.-% to 69 wt.-% (determined by ¹³C-NMR) and each at various concentrations, in the range of 2 and 13mg/ml. To encounter for both features, concentration and ethylene content at the same time for various polymer concentrations expected during Crystex analyses the following calibration equations were applied:

$$\text{Conc} = a + b \cdot \text{Abs}(\text{CH}) + c \cdot (\text{Abs}(\text{CH}))^2 + d \cdot \text{Abs}(\text{CH}_3) + e \cdot (\text{Abs}(\text{CH}_3))^2 + f \cdot \text{Abs}(\text{CH}) \cdot \text{Abs}(\text{CH}_3) \quad (\text{Equation 1})$$

$$\text{CH}_3/1000\text{C} = a + b \cdot \text{Abs}(\text{CH}) + c \cdot \text{Abs}(\text{CH}_3) + d \cdot (\text{Abs}(\text{CH}_3)/\text{Abs}(\text{CH})) + e \cdot (\text{Abs}(\text{CH}_3)/\text{Abs}(\text{CH}))^2 \quad (\text{Equation 2})$$

The constants a to e for equation 1 and a to f for equation 2 were determined by using least square regression analysis.

The CH₃/1000C is converted to the ethylene content in wt.-% using following relationship:

$$\text{Wt.-% (Ethylene in EP Copolymers)} = 100 - \text{CH}_3/1000\text{TC} \cdot 0.3 \quad (\text{Equation 3})$$

Amounts of Soluble Fraction (SF) and Crystalline Fraction (CF) are correlated through the XS calibration to the "Xylene Cold Soluble" (XCS) quantity and respectively Xylene Cold Insoluble (XCI) fractions, determined according to standard gravimetric method as per ISO16152. XS calibration is achieved by testing various EP copolymers with XS content in the range 2-31 Wt%. The determined XS calibration is linear:

$$\text{Wt.-% XS} = 1,01 \cdot \text{Wt.-% SF} \quad (\text{Equation 4})$$

Intrinsic viscosity (IV) of the parent EP copolymer and its soluble and crystalline fractions are determined with a use of an online 2-capillary viscometer and are correlated to corresponding IV's determined by standard method in decalin according to ISO 1628-3. Calibration is achieved with various EP PP copolymers
5 with IV = 2-4 dL/g. The determined calibration curve is linear:

$$IV \text{ (dL/g)} = a \cdot V_{sp}/c \text{ (equation 5)}$$

The samples to be analyzed are weighed out in concentrations of 10mg/ml to
10 20mg/ml. To avoid injecting possible gels and/or polymers, which do not dissolve in TCB at 160°C, like PET and PA, the weighed out sample was packed into a stainless steel mesh MW 0,077/D 0,05mm.

After automated filling of the vial with 1,2,4-TCB containing 250 mg/l 2,6-tert-butyl-4-methylphenol (BHT) as antioxidant, the sample is dissolved at 160°C until
15 complete dissolution is achieved, usually for 60 min, with constant stirring of 400rpm. To avoid sample degradation, the polymer solution is blanketed with the N₂ atmosphere during dissolution.

A defined volume of the sample solution is injected into the column filled with inert support where the crystallization of the sample and separation of the soluble
20 fraction from the crystalline part is taking place. This process is repeated two times. During the first injection the whole sample is measured at high temperature, determining the IV[dL/g] and the C₂[wt%] of the PP composition. During the second injection, the soluble fraction (at low temperature) and the crystalline fraction (at high temperature) with the crystallization cycle are
25 measured (Wt% SF, Wt% C₂, IV).

b) Quantification of microstructure by NMR spectroscopy (calibration only)

Quantitative nuclear-magnetic resonance (NMR) spectroscopy was used for
30 calibration.

Quantitative ¹³C{¹H} NMR spectra were recorded in the solution-state using a Bruker Avance Neo 400 NMR spectrometer operating at 400.15 and 100.62

MHz for ¹H and ¹³C respectively. All spectra were recorded using a ¹³C optimized 10 mm extended temperature probe head at 125°C using nitrogen gas for all pneumatics. Approximately 200 mg of material was dissolved in approximately 3 ml of 1,2-tetrachloroethane-d₂ (TCE-d₂) along with approximately 3 mg BHT (2,6-di-tert-butyl-4-methylphenol CAS 128-37-0) and chromium-(III)-acetylacetonate (Cr(acac)₃) resulting in a 60 mM solution of relaxation agent in solvent as described in G. Singh, A. Kothari, V. Gupta, *Polymer Testing* 2009, 28(5), 475.

To ensure a homogenous solution, after initial sample preparation in a heat block, the NMR tube was further heated in a rotatory oven for at least 1 hour. Upon insertion into the magnet the tube was spun at 10 Hz. This setup was chosen primarily for the high resolution and quantitatively needed for accurate ethylene content quantification. Standard single-pulse excitation was employed without NOE, using an optimised tip angle, 1 s recycle delay and a bi-level WALTZ16 decoupling scheme as described in Z. Zhou, R. Kuemmerle, X. Qiu, D. Redwine, R. Cong, A. Taha, D. Baugh, B. Winniford, *J. Mag. Reson.* 187 (2007) 225 and V. Busico, P. Carbonniere, R. Cipullo, C. Pellecchia, J. Severn, G. Talarico, *Macromol. Rapid Commun.* 2007, 28, 1128. A total of 6144 (6k) transients were acquired per spectra.

Quantitative ¹³C{¹H} NMR spectra were processed, integrated and relevant quantitative properties determined from the integrals. All chemical shifts were indirectly referenced to the central methylene group of the ethylene block (EEE) at 30.00 ppm using the chemical shift of the solvent. This approach allowed comparable referencing even when this structural unit was not present.

Characteristic signals corresponding to the incorporation of ethylene were observed (as described in Cheng, H. N., *Macromolecules* 1984, 17, 1950) and the comonomer fraction calculated as the fraction of ethylene in the polymer with respect to all monomer in the polymer:

$$f_E = (E / (P + E))$$

The comonomer fraction was quantified using the method of W-J. Wang and S. Zhu, *Macromolecules* 2000, 33 1157, through integration of multiple signals across the whole spectral region in the $^{13}\text{C}\{^1\text{H}\}$ spectra. Integral regions were slightly adjusted to increase applicability across the whole range of encountered comonomer contents.

The mole percent comonomer incorporation was calculated from the mole fraction:

$$E [\text{mol}\%] = 100 * fE$$

The weight percent comonomer incorporation was calculated from the mole fraction:

$$E [\text{wt}\%] = 100 * (fE * 28.06) / ((fE * 28.06) + ((1-fE) * 42.08)).$$

c) Tensile modulus and tensile strain at break were measured according to ISO 527-2 (cross head speed = 1 mm/min; test speed 50 mm/min at 23 °C) using injection molded specimens as described in EN ISO 1873-2 (dog bone shape, 4 mm thickness). The measurement is done after 96 h conditioning time of the specimen.

d) Impact strength was determined as notched Charpy impact strength (1eA) (non-instrumented, ISO 179-1 at +23 °C) according to ISO 179-1 eA at +23 °C on injection moulded specimens of 80 x 10 x 4 mm prepared according to EN ISO 1873-2.

e) Inorganic residues: TGA according to DIN ISO 1172:1996 using 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 ash content was evaluated as the weight % at 850°C.

f) MFR: melt flow rates were measured with a load of 2.16 kg (MFR_2) at 230 °C. The melt flow rate is that quantity of polymer in grams, which the test

apparatus standardized to ISO 1133 extrudes within 10 minutes at a temperature of 230 °C under a load of 2.16 kg.

g) Amount of Metals

5 determined by x ray fluorescence (XRF).

h) Amount of Paper, Wood (for control purposes only)

10 paper and wood can be determined by conventional laboratory methods including milling, floatation, microscopy and Thermogravimetric Analysis (TGA).

i) Benzene content

by **HS GC-MS 80°C/2h**, which is described as the following

Static headspace analysis

15 The parameters of the applied static headspace gas chromatography mass spectrometry (HS/GC/MS) method are described here.

4.000 ± 0.100 g sample were weighed in a 20 ml HS vial and tightly sealed with a PTFE cap.

20

The mass spectrometer was operated in scan mode and a total ion chromatogram (TIC) was recorded for each analysis. More detailed information on applicable method parameters and data evaluation are given below:

25

- HS parameter (Agilent G1888 Headspace Sampler)

Vial equilibration time: 120 min

Oven temperature: 80 °C

Loop temperature: 205 °C

30 Transfer line temperature: 210 °C

Low shaking

- GC parameter (Agilent 7890A GC System)
- Column: ZB-WAX 7HG-G007-22 (30 m x 250 μ m x 1 μ m)
- Carrier gas: Helium 5.0
- Flow: 2 ml/min
- 5 Split: 5:1
- GC oven program: 35 °C for 0.1 min
10 °C/min until 250 °C
250 °C for 1 min
- MS parameter (Agilent 5975C inert XL MSD)
- 10 Acquisition mode: Scan
- Scan parameters:
- Low mass: 20
- High mass: 200
- Threshold: 10
- 15 - Software/Data evaluation
- MSD ChemStation E.02.02.1431
- MassHunter GC/MS Acquisition B.07.05.2479
- AMDIS GC/MS Analysis Version 2.71
- NIST Mass Spectral Library Version 2.0 g
- 20 - AMDIS deconvolution parameters
- Minimum match factor: 80
- Threshold: Low
- Scan direction: High to Low
- Data file format: Agilent files
- 25 Instrument type: Quadrupole
- Component width: 20
- Adjacent peak subtraction: Two
- Resolution: High
- Sensitivity: Very high
- 30 Shape requirements: Medium
- Solvent tailing: 44 m/z
- Column bleed: 207 m/z
- Min. model peaks: 2

Min. S/N: 10
Min. certain peaks: 0.5

Data evaluation

5 The TIC data were further deconvoluted with the aid of AMDIS software (see parameters stated above) and compared to a custom target library which was based on the mass spectral library (NIST). In the custom target library, the respective mass spectra of selected substances (e.g. benzene) were included. Only when the recognised peak showed a minimum match factor of 80 and an experienced mass spectroscopist confirmed the match, a substance was accepted as "tentatively identified".
10 In this study, the statement "below the limit of detection (< LOD)" referred to a condition where either the match factor was below 80 (AMDIS) or the peak as such was not even recognised. The results refer solely to the measured samples, time of measurement and the applied parameters.
15

j) **CIELAB color space (L*a*b*)**

In the CIE L*a*b* uniform color space, measured according to DIN EN ISO 11664-4, the color coordinates are: L*—the lightness coordinate; a*—the red/green coordinate, with +a* indicating red, and -a* indicating green; and b*—the yellow/blue coordinate, with +b* indicating yellow, and -b* indicating blue. The L*, a*, and b* coordinate axis define the three dimensional CIE color space. Standard Konica/Minolta Colorimeter CM-3700A.
20
25

k) **Odor VDA270-B3**

VDA 270 is a determination of the odor characteristics of trim-materials in motor vehicles. In this study, the odor is determined following VDA 270 (2018) variant B3.. The odor of the respective sample is evaluated by each assessor according to the VDA 270 scale after lifting the jar's lid as little as possible. The hexamorous scale consists of the following grades: Grade 1: not perceptible, Grade 2: perceptible, not disturbing, Grade 3: clearly
30

perceptible, but not disturbing, Grade 4: disturbing, Grade 5: strongly disturbing, Grade 6: not acceptable. Assessors stay calm during the assessment and are not allowed to bias each other by discussing individual results during the test. They are not allowed to adjust their assessment after testing another sample, either. For statistical reasons (and as accepted by the VDA 270) assessors are forced to use whole steps in their evaluation. Consequently, the odor grade is based on the average mean of all individual assessments, and rounded to whole numbers.

10 **I) Limonene detection**

Limonene quantification can be carried out using solid phase microextraction (HS-SPME-GC-MS) by standard addition. 50 mg ground samples are weighed into 20 mL headspace vials and after the addition of limonene in different concentrations and a glass-coated magnetic stir bar, the vial is closed with a magnetic cap lined with silicone/PTFE. Micro capillaries (10 pL) are 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 is used in combination with some of the samples tested in this application. For quantification, ion-93 acquired in SIM mode is used. Enrichment of the volatile fraction is 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 is carried out directly in the heated injection port of a GCMS system at 270°C.

25 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)

30 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

m) Fatty acid detection

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

50 mg ground samples are weighed in 20 mL headspace vial and after the addition of limonene in different concentrations and a glass coated magnetic stir bar the vial is closed with a magnetic cap lined with silicone/PTFE. 10 μ L Micro-capillaries are 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 is used for all acids except propanoic acid, here ion 74 is used.

GCMS Parameter:

Column: 20 m ZB Wax plus 0.25*0.25

Injector: Split 5:1 with glass lined split liner, 250°C

20 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

25 SIM Parameter: m/z 60, 74, 6.6 scans/s

n) Presence of polyamide-6 and polystyrene

30 By FTIR spectroscopy using the absorption of the band at 1601 cm^{-1} (PS) and 3300 cm^{-1} (PA6).

o) Determination of contaminations on the plaques

The plaques are injection-moulded, 150x80x2mm dimension. Then, a high-resolution image (photograph) is taken on the 5 plaques (putting them close

to each other). The image is then analyzed by a software allowing an automatic counting of the number of visual defects (by naked-eyes) due to contaminations.

5

p) Dynamic Shear Measurements (Eta(2.7kPa) and Eta(300rad/s))

The characterisation of polymer melts by dynamic shear measurements complies with ISO standards 6721-1 and 6721-10. The measurements were performed on an Anton Paar MCR501 stress controlled rotational rheometer, equipped with a 25 mm parallel plate geometry. Measurements were undertaken on compression-moulded plates, using nitrogen atmosphere and setting a strain within the linear viscoelastic regime. The oscillatory shear tests were done at 230°C applying a frequency range between 0.01 and 600 rad/s and setting a gap of 1.3 mm.

In a dynamic shear experiment the probe is subjected to a homogeneous deformation at a sinusoidal varying shear strain or shear stress (strain and stress controlled mode, respectively). On a controlled strain experiment, the probe is subjected to a sinusoidal strain that can be expressed by

$$\gamma(t) = \gamma_0 \sin(\omega t) \quad (1)$$

If the applied strain is within the linear viscoelastic regime, the resulting sinusoidal stress response can be given by

$$\sigma(t) = \sigma_0 \sin(\omega t + \delta) \quad (2)$$

where

σ_0 and γ_0 are the stress and strain amplitudes, respectively

ω frequency is the angular

δ is the phase shift (loss angle between applied strain and stress response)

t is the time

Dynamic test results are typically expressed by means of several different rheological functions, namely the shear storage modulus G' , the shear loss modulus, G'' , the complex shear modulus, G^* , the complex shear viscosity, η^* , the dynamic shear viscosity, η' , the out-of-phase component of the complex shear viscosity η'' and the loss tangent, $\tan \delta$ which can be expressed as follows:

$$G' = \frac{\sigma_0}{\gamma_0} \cos \delta \quad [\text{Pa}] \quad (3)$$

$$G'' = \frac{\sigma_0}{\gamma_0} \sin \delta \quad [\text{Pa}] \quad (4)$$

5

$$G^* = G' + iG'' \quad [\text{Pa}] \quad (5)$$

$$\eta^* = \eta' - i\eta'' \quad [\text{Pa.s}] \quad (6)$$

10

$$\eta' = \frac{G''}{\omega} \quad [\text{Pa.s}] \quad (7)$$

$$\eta'' = \frac{G'}{\omega} \quad [\text{Pa.s}] \quad (8)$$

ETA(x kPa) is determined according with equation 9.

15

$$ETA(x \text{ kPa}) = \text{Eta}^* \text{ for } (G^* = x \text{ kPa}) \quad [\text{Pa.s}] \quad (9)$$

For example, the ETA(2.7 kPa) is the defined by the value of the complex viscosity, determined for a value of complex modulus equal to 2.7 kPa.

Eta (x rad/s) is determined according with equation 10.

20

$$ETA(x \text{ rad/s}) = \text{Eta}^* \text{ for } (\omega = x \text{ rad/s}) \quad [\text{Pa.s}] \quad (10)$$

For example, the ETA(300 rad/s) is defined by the value of the complex viscosity, determined at a frequency sweep of 300 rad/s.

q) Shear Thinning Factor (STF) is defined as

25

$$\text{STF} = \frac{\text{Eta}^* \text{ for } (\omega = 0.05 \text{ rad/s})}{\text{Eta}^* \text{ for } (\omega = 300 \text{ rad/s})} \quad (11)$$

The values are determined by means of a single point interpolation procedure, as defined by Rheoplus software. In situations for which a given G^* value is not experimentally reached, the value is determined by means of an extrapolation, using the same procedure as before. In both cases (interpolation or extrapolation), the option from Rheoplus "Interpolate y-values to x-values from parameter" and the "logarithmic interpolation type" were applied.

References:

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r) **LARGE AMPLITUDE OSCILLATORY SHEAR (LAOS)**

The investigation of the non-linear viscoelastic behavior under shear flow was done resorting to Large Amplitude Oscillatory Shear. The method requires the application of a sinusoidal strain amplitude, γ_0 , imposed at a given angular frequency, ω , for a given time, t . Provided that the applied sinusoidal strain is high enough, a non-linear response is generated. The stress, σ is in this case a function of the applied strain amplitude, time and the angular frequency. Under these conditions, the non-linear stress response is still a periodic function; however, it can no longer be expressed by a single harmonic sinusoid. The stress resulting from a non-linear viscoelastic response [0-0] can be expressed by a Fourier series, which includes the higher harmonics contributions:

$$\sigma(t, \omega, \gamma_0) = \gamma_0 \cdot \sum_n [G'_n(\omega, \gamma_0) \cdot \sin(n\omega t) + G''_n(\omega, \gamma_0) \cdot \cos(n\omega t)] \quad (1)$$

- with, σ - stress response
- 5 t - time
- ω - frequency
- γ_0 - strain amplitude
- n - harmonic number
- G'_n - n order elastic Fourier coefficient
- 10 G''_n - n order viscous Fourier coefficient

The non-linear viscoelastic response was analysed applying Large Amplitude Oscillatory Shear (LAOS). Time sweep measurements were undertaken on an RPA 2000 rheometer from Alpha Technologies coupled with a standard biconical die. During the course of the measurement the test chamber is sealed and a pressure of about 6 MPa is applied. The LAOS test is done applying a temperature of 190 °C, an angular frequency of 0.628 rad/s and a strain of 1000 %. In order to ensure that steady state conditions are reached, the non-linear response is only determined after at least 20 cycles per measurement are completed. The Large Amplitude Oscillatory Shear Non-Linear Factor (LAOS_NLF) is defined by:

$$LAOS_{NLF}(190^\circ C, 1000\%) = \left| \frac{G'_1}{G'_3} \right| \quad (2)$$

- where G'_1 - first order Fourier Coefficient
- 25 G'_3 - third order Fourier Coefficient

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s) Cross Fractionation Chromatography

The chemical composition distribution as well as the determination of the molecular weight distribution and the corresponded molecular weight averages (Mn, Mw and Mv) at a certain elution temperature (polymer crystallinity in solution) were determined by a full automated Cross Fractionation Chromatography (CFC) as described by Ortin A., Monrabal B., Sancho-Tello J., Macromol. Symp., **2007**, 257, 13-28.

15

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.

20

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.

25

Once the sample was completely dissolved an aliquot of 0.5 ml was loaded into the TREF column and stabilized for a while at 110 °C. The polymer was crystallized and precipitate to a temperature of 30°C by applying a constant cooling rate of 0.1 °C/min. A discontinuous elution process is performed using the following temperature steps: (35, 40, 50, 55, 60, 65, 70, 75, 80, 85, 90, 95, 100, 103, 106, 109, 112, 115, 117, 119, 121, 123, 125, 127, 130, 135 and 140).

30

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 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 weights into the PP molecular weight equivalents.

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

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

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.

Calculation of the relative fraction at certain molecular weight and elution temperature areas of iso-PP in wt.-%.

20 To calculate the relative fraction at certain molecular weight and elution temperature areas of iso-PP in wt.-% in the first step the amount of iso-PP in wt.-% from the CFC contour plot needs to be calculated:

$$\text{Iso PP in wt.-%} = 100 - \text{EPR fraction} - \text{PE fraction} \quad \text{equation (1)}$$

25

Where the EPR is the fraction with a molar mass higher than $\log M$ of 3.5 of the soluble fraction (SF) in TCB at 35°C obtained by CFC analysis

$$EPR \text{ fraction in wt. } - \% = \frac{\sum_{j=3.5}^8 H_j}{\sum_{j=2}^8 H_j} * SF \quad \text{equation (2)}$$

Due to the slightly dependence of the TREF profile on the low MW part, the molecular weight limit of the low MW limit is elution temperature (T_{el}) dependent.

- 5 The low MW limit was determined using the following formula:

$$\text{Low MW limit (for PE Fraction)} = 0.0185 * T_{el} + 3.1538$$

Taking this into account the PE fraction is calculated using the following approach.

$$PE_{Fraction} = \frac{\sum_{i=35}^{99} \sum_{j=0.0185*i+3.1538}^8 H_{ij}}{\sum_{i=30}^{140} \sum_{j=2}^8 H_{ij}} * 100 \quad \text{(equation 3)}$$

- 10 Where H_{ij} is the 2D differential distribution at the corresponded elution temperature (T_{el}) i and the logM value j , obtained with the corresponded data processing software.

The High crystalline PE fraction (HCF-PE) is defined as the part of the PE fraction eluting from 90°C to 100°C of PE fraction.

$$15 \quad HCF - PE = \frac{\sum_{i=90}^{99} \sum_{j=0.0185*i+3.1538}^8 H_{ij}}{\sum_{i=30}^{140} \sum_{j=2}^8 H_{ij}} * 100 \quad \text{(equation 4)}$$

This fraction contains mainly homo PE and PE copolymers with very low amount of comonomer, below app. 3 SCB/1000TC (L. Wild, T.R. Ryle, D.C. Knoblauch, I.R. Peat, J. Polym. Sci, Polym. Phys. 20, (1982), 441-455).

- 20 Where the low crystalline PE Fraction (LCF-PE) is defined as the part of the PE fraction eluting between 35°C and 89°C of the PE fraction.

$$HCF - PE = \frac{\sum_{i=35}^{89} \sum_{j=0.0185*i+3.1538}^8 H_{ij}}{\sum_{i=30}^{140} \sum_{j=2}^8 H_{ij}} * 100 \quad \text{(equation 5)}$$

This fraction contains mainly the copolymer fraction from HDPE and LLDPE obtained by ZN catalysts or the LLDPE from SS catalysts but also LDPE, as this kind of polymer are co-eluting due to their comparable amount of SCB/1000TC.

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Examples

A post-consumer plastic trash was coarsely sorted as to polymer nature and as to color. In a further step, white and colorless parts were selected. The selected parts were subjected to milling, washing in an aqueous solution with various
15 detergents and subsequent drying and screening. The pretreated post-consumer plastic material was further sorted thereby reducing colored parts. In a further quality control step the intermediate was subjected to Cross Fractionation Chromatography (automated instrument combining TREF and CPC; available
20 from Polymer Char; details cf. above). All intermediates having a low crystalline PE Fraction (LCF-PE) outside the range of 2.8 to 4.2 wt.-% were discarded and subjected back to the incoming post-consumer trash. After extrusion into pellets, the pellets were subjected to aeration (aeration only done with IE3; aeration conditions: at 120°C air, pre-heating substrate).

25

For examples IE4 the same process was followed. However, after coarsely sorting as to polymer nature and as to color, the post-consumer plastic trash was screened as to apparently very old ('ancient') mainly colorless / natural plastic articles recognizable by discoloration (e.g. pronounced yellowing) and/or
30 pronounced scratches of the mainly colorless / natural plastic articles. Such very

- old ('ancient') mainly colorless / natural plastic articles were sorted out, whereby a strict standard was followed, i.e. in case of doubt, the article in question was sorted out. This was done for excluding incorporation of polybrominated diphenyl ethers which have been banned in many countries for more than about 10 years.
- 5 Intermediates were also screened as to substances of very high concern. Partial warping turned out to be not necessary.

All examples were subjected to CRYSTEX QC analysis.

Table 1: Characteristics of the inventive examples

	IE1	IE2	IE3	IE4	CE3 virgin random heterophasic polypropylene copolymer (RAHECO)	CE4 virgin random visbroken	claimed features	
target								
		1 st embodiment (CIELAB color) L* from 85.0 to 97.0; a* from -5.0 to 0.0; b* from 0.0 to below 8.0		2 nd embodiment (CIELAB color) L* from 72.0 to 97.0, preferably from 80.0 to 97.0; a* from -5.0 to 0.0; b* from 0.0 to below 22.0				
	87.0 to 90.0 wt.-% crystalline fraction (CF) 10.0 to 13.0 wt.-%, soluble fraction (SF) content	+ CRYSTEX QC analysis content and		+ CRYSTEX QC analysis 91.0 to 94.0 wt.-% crystalline fraction (CF) content and				

	6.0 to 9.0 wt.- %, soluble fraction (SF) content					
CF wt.-%	88.9	89.4	89.2	84.9	91.2	86.0 to 94.0
SF wt.-%	11.1	10.6	10.8	15.1	8.8	6.0 to 14.0
C3 (CF) wt.-%	97.2	96.9	96.9	96.5	97.3	95.0 to 99.0 pref. 96.0 to 98.0
C2 (CF) wt.-%	2.8	3.1	3.1	3.5	2.7	1.0 to 5.0 pref. 2.0 to 4.0 more pref. 2.5 to 3.5
IV(SF) dl/g	1.41	1.39	1.36	2.40	0.72	1.10 to below 1.50 pref. 1.25 to 1.45
Inorganic residues (TGA), wt.-% DIN ISO 1172	2.0	2.0	2.0	< 0.05	< 0.05	0.05 to 3.0 pref. 0.05 to 2.5 optionally. 1.0 to 2.5
Benzene presence (HS GC-MS 80°C/2h)	< LOD (Limit of Detection)	< LOD (Limit of Detection)	< LOD (Limit of Detection)	< LOD (Limit of Detection)	< LOD (Limit of Detection)	< LOD (Limit of Detection)
Color L* a* b*	92.0 -1.4 6.4	92.5 -1.5 6.4	92.5 -1.5 6.4	84 -0.8 -2.0		80.0 to 97.0 -5.0 to 0.0 0.0 to below 22.0

MFR(230°C/2.16 kg), g/10min	19	51	51	7.4	7	8	2.0 to 100 (claim 2)
Density, kg/m ³ ISO-1183	919	919	919	909	n.d.	n.d.	-
Recycled material	yes	yes	yes	Yes	no, virgin	no, virgin	claim 3
mixture of countless polypropylenes	yes	yes	yes	yes	no	no	<>
Polystyrene	not detectable	not detectable	not detectable	not detectable	no	no	claim 4
Polyamide-6	not detectable	not detectable	not detectable	not detectable	no	no	claim 4
Limonene (HS-SPME-GC-MS)	> 1 ppm	> 1 ppm	> 1 ppm	n.d.	no	no	claim 4
Fatty acids (HS-SPME-GC-MS)	n.d.	n.d.	n.d.	n.d.	no	no	claim 4
C2(SF)	28.3	27.1	27.0	17.7	36.3	20.4	15.0 to 32.0 pref. 15.0 to 20.0 (claim 5)
Odor grade VDA270-B3	4	n.d.	3	3	n.m.	n.m.	4 or lower (claim 6) pref. 3 or lower (claim 6)
low crystalline PE Fraction (LCF-PE) in CFC (wt.-%)	3.1	n.m.	3.7	3.9	n.m.	n.m.	

LAOS – NLF 1000% 190°C	2.76	3.67	3.50	2.4	2.0	3.0	> 2.3 (claim 1) pref. > 2.7 pref. > 3.3 (claim 7)
Tensile Modulus, MPa	1444	1390	1397	1267	1224	709	at least 1200 pref. 1250 / 1300 / 1350 /1390 (claim 8)
Melting point; °C				160	164	140	
Charpy NIS +23°C, kJ/m ² (>96h)	4.9 ± 0.6	4.7 ± 0.6	4.6 ± 0.2	8.5 ± 0.4	7.1 ± 0.1	7.6 ± 0.1	-
eta (2.7 kPa)	1271	481	354	2910	3108	2377	
stability of eta (2.7 kPa)	1306 ++	483 ++	379 +		n.m.	n.m.	
eta (0.05 rad/s) Pa.s.	1833		451	4080	4246	2759	
eta (300 rad/s), Pa.s.	199	141	123	275	306	316	
stability of eta (300 rad/s), Pa.s.	203 ++	142 ++	129 ++		n.m.	n.m.	
Shear thinning factor STF eta 0.05 / eta 300	9.0	n.d.	3.7	14.8	13.9	8.7	
VOC (VDA278) pellets, µg/g	n.m.	n.m.	6	6	159	98	
FOG (VDA278) pellets, µg/g	n.m.	n.m.	244	253	169	112	

VDA277 (total carbon emission) pellet, 5h/120°C	10	n.m.	n.m.	3	n.m.	33
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It can be seen that the prior art is enriched by the inventive recycling composition. The inventive recycling composition showed only minor drawbacks as to impact when compared with virgin compositions and virgin visbroken compositions. Moreover, the processability as reflected by high LAOS-NLF was really good for IE2 and IE3. The VOC (VDA) was surprisingly good for IE3 and IE4.

Comparative Example CE3 was a virgin random heterophasic polypropylene copolymer. Accordingly the tensile modulus was relatively high for the total amount of ethylene (reflected by C2(CF) and C2(SF) and the amounts of CF and SF). However, this is not a recydate, processability as reflected by LAOS – NLF is relatively poor at a Charpy NIS of 7.1 kJ/m².

Comparative Example CE4 was a virgin random copolymer, which was subjected to visbreaking (for adaptation of the melt flow rate to about the same value as IE4). CE4 was significantly less stiff compared to CE3 and had a Charpy NIS of 7.6 kJ/m².

Inventive example IE4 had a marginally higher stiffness than CE3 and simultaneously the best overall Charpy NIS of 8.5 kJ/m². Inventive Examples IE1 to 3 all had significantly higher stiffness and simultaneously only moderate Charpy NIS drawbacks.

The examples were subjected to evaluation of surface defects at injection molded test articles. The best commercial grades (CE1 and CE2) as available on the market were compared. Results are shown in Fig. 1.

5 CE1: is PP off-white product from Van Werven having a density of 920 kg/m³ and MFR(230°C/2.16kg) of 24 g/10min.

CE2: Morssinkhof - Rymoplast supplies regrinds and regranulates, with the name MOPRYLENE®, having a density of 921 kg/m³ and MFR (230°C/2.16kg)
10 of 27 g/10min.

Fig. 1 shows the results of the defect evaluation. It can be seen that the inventive mixed-plastics polypropylene blends resulted in the lowest number of defects and also an even distribution thereof.

15

CE3 is a random heterophasic copolymer without slip and antiblock additives. It has a random copolymer PP matrix and a C3C2 rubber.

CE3 was produced in a Borstar polypropylene plant with a prepolymerization
20 reactor, one slurry loop reactor, a first gas phase reactor and a second gas phase reactor configuration. The loop and first gas phase reactor were used to produce the matrix and second gas phase reactor for rubber phase.

The chemical composition of the reactants in each reactor were adjusted to reach the desired polymer design.

25

CE4 is a random copolymer, which was produced in a Borstar polypropylene plant with a prepolymerization reactor, one slurry loop reactor and one gas
phase reactor configuration.

30

	unit	CE3	CE4
Prepolymerizer			
Catalyst		as decribed below	as decribed below
Teal	g/tC3	150	170
Donor	g/tC3	40	40
Donor type	n.a.	D	D
Temperature	°C	20	30
Residence time	h	0,33	0,35
Loop			
Temperature	°C	80	70
Feeding H2/C3	mol/kmol	0	0,6
Feeding C2/C3	mol/kmol	3,3	7,5
Split	wt%	51	41
MFR	g/10min	12	1,8
C2	wt%	0	3,5
XCS	wt%	3	5,2
First gas phase reactor			
Temperature	°C	80	80
H2/C3	mol/kmol	12	5,8
C2/C3	mol/kmol	0	27
Split	wt%	34	59
MFR	g/10min	12	1,9
C2	wt%	0	4,1
MFR ₂	g/10 min	12	1.9
Second gas phase reactor			
Temperature	°C	70	
H2/C2	mol/kmol	200	
C2/C3	mol/kmol	50	

Split	wt%	15	
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After polymerization the melt flow rate of the random copolymer CE4 was modified by vis-breaking during a compounding step in a twin screw extruder at 200-230°C and using an appropriate amount of Luperox 101 (2,5-dimethyl-2,5-di(tert-butylperoxy)hexane) to achieve the target MFR2 of 8.0 g/10 min.

During said compounding step the following additives 1000 ppm Irganox B215 (a 1:2-mixture of Pentaerythrityl-tetrakis(3-(3',5'-di-tert. butyl-4-hydroxyphenyl)propionate, CAS-no. 6683-19-8, and Tris (2,4-di-t-butylphenyl) phosphite, CAS-no. 31570-04-4, commercially available from BASF SE, Germany) and 150 ppm magnesium oxide (CAS-no. 1309-48-4) as acid scavenger were added during the compounding step.

The catalyst used in the polymerization process for the CE3 and CE4 was prepared as follows:

Used chemicals:

20 % solution in toluene of butyl ethyl magnesium (Mg(Bu)(Et), BEM), provided by Chemtura

2-ethylhexanol, provided by Amphochem

3-Butoxy-2-propanol - (DOWANOL™ PnB), provided by Dow

bis(2-ethylhexyl)citraconate, provided by SynphaBase

TiCl₄, provided by Millenium Chemicals

Toluene, provided by Aspokem

Viscoplex® 1-254, provided by Evonik

Heptane, provided by Chevron

25

Preparation of a Mg alkoxy compound

Mg alkoxide solution was prepared by adding, with stirring (70 rpm), into 11 kg of a 20 wt-% solution in toluene of butyl ethyl magnesium (Mg(Bu)(Et)), a mixture of 4.7 kg of 2-ethylhexanol and 1.2 kg of butoxypropanol in a 20 l stainless steel reactor. During the addition the reactor contents were maintained below 45 °C. After addition was completed, mixing (70 rpm) of the reaction mixture was continued at 60 °C for 30 minutes. After cooling to room temperature 2.3 kg of

the donor bis(2-ethylhexyl)citraconate was added to the Mg-alkoxide solution keeping temperature below 25 °C. Mixing was continued for 15 minutes under stirring (70 rpm).

Preparation of solid catalyst component

- 5 20.3 kg of TiCl₄ and 1.1 kg of toluene were added into a 20 l stainless steel reactor. Under 350 rpm mixing and keeping the temperature at 0 °C, 14.5 kg of the prepared Mg alkoxy compound was added during 1.5 hours. 1.7 l of Viscoplex® 1-254 and 7.5 kg of heptane were added and after 1 hour mixing at 0 °C the temperature of the formed emulsion was raised to 90 °C within 1 hour.
- 10 After 30 minutes mixing was stopped, the catalyst droplets were solidified and the formed catalyst particles were allowed to settle. After settling (1 hour), the supernatant liquid was siphoned away. Then the catalyst particles were washed with 45 kg of toluene at 90°C for 20 minutes followed by two heptane washes (30 kg, 15 min). During the first heptane wash the temperature was decreased to 50
- 15 °C and during the second wash to room temperature.

Alternatively for the polymerization of the random copolymer (comparative CE3, CE4) a phthalate-free Ziegler Natta catalyst prepared as described in the example section of WO 2020/064673 A1 as "Reference Catalyst" is used.

20

The thus obtained catalyst was used along with triethyl-aluminium (TEAL) as co-catalyst and dicyclopentyl dimethoxy silane (D-Donor) as donor.

IE4 was further evaluated as to substances of very high concern (SVHC).

Results are shown in the Table 2 below.

25

Table 2: Evaluation substances of very high concern for IE4:

Test item	Reporting Limit (RL), mg/kg	RoHS limit, mg/kg	IE4 mg/kg
Cd	1	100	<1
Pb	10	1000	<10
Hg	0.5	1000	<0.5
Cr (VI)	1	1000	<1
Polybrominated diphenyl ethers (PBDE)		sum 1000	not detected
Polybrominated biphenyls (PBB)		sum 1000	not detected
Bis(2-ethylhexyl)phthalate (DEHP)	100	1000	not detected
Benzyl Butyl phthalate (BBP)	100	1000	not detected
Dibutyl phthalate (DBP)	100	1000	not detected
Diisobutyl phthalate DiBP	100	1000	not detected
Polycyclic aromatic hydrocarbons PAH		Sum of 18 <10	0.2
Short chain chlorinated paraffins (SCCP)		<1500	<50
Phthalates		<0.1% in article	<100
Chlorinated Phenols		<5	<0.1
Other flame retardants		X	<100
Cl		50	<50
Br		50	<50
F		50	<50
I		50	<50
SCORE			Pass
SVHC			<0.1%
Total SCORE (for RoHS and SVHC)			pass

RoHS - Restriction of Hazardous Substances in Electrical and Electronic

5 Equipment

RL – Reporting Limit (test data will be shown if it is ≥RL. RL is not regulatory limit)

Method for SVHCs (according to REACH regulation 1907/2006/EU)

10 The whole analysis was done at SGS.

In-house method at SGS are CTS-HL-114-1, CTS-HL-234-5 analysed by ICP-OES, UV-VIS, GC-MS, HPLC-DAD/MS and colorimetric method

Determination of Cadmium by ICP-OES, acc. to IEC 62321-5:2013-6

15 Determination of Lead by ICP-OES, acc. to IEC 62321-5:2013-6

Determination of Mercury by CV-AAS, acc. to IEC 62321-4:2013-6

Determination of Chromium by ICP-OES, acc. to IEC 62321-5:2013-6

Determination of Chromium(VI) acc. to IEC 62321-> Non-metallic samples:

5 Determination by ion chromatography, acc. to IEC 62321-7-2:2017-03;

Remark: The concentration of Cr(VI) in a corrosion-protection can change depending on storage time and conditions.

10 Determination of PBB/PBDE (flame retardants) by GC/MS, acc. to IEC 62321-6:2015-6

Remark: Acc. to IEC, the testing for PBB/PBDE is only intended for polymers.

Softeners DEHP, DBP; BBP, DIBP and an extended list according to REACH (IEC 62321-8:2017, GC-MS

15

Determination of Phthalates by GC/MS after extraction with THF, acc. to IEC 62321-8:2017-3; Method not under accreditation

Remark: Acc. to IEC, the testing for phthalates is only intended for polymers.

20

CE3 is a bimodal virgin random polypropylene which was evaluated for comparative purposes. It can be seen that virgin random polypropylenes can be substituted by the inventive blends with surprising benefits as to impact.

25 As a further application example, injection molded pails were produced with standard process setting. The surface quality turned out to be as good as the surface quality of comparable virgin material. Thickness distribution and mechanical properties were also excellent.

30

Claims

1. Mixed-plastics polypropylene blend having
- 5 (i) a crystalline fraction (CF) content determined according to CRYSTEX QC analysis in the range from 86.0 to 94.0 wt.-%, and
- (ii) a soluble fraction (SF) content determined according to CRYSTEX QC analysis in the range from 6.0 to 14.0 wt.-%, whereby
- 10 (iii) said crystalline fraction (CF) has a propylene content (C3(CF)) as determined by FT-IR spectroscopy calibrated by quantitative ¹³C-NMR spectroscopy, in the range from 95.0 to 99.0 wt.-%; preferably 96.0 to 98.0 wt.-% and whereby
- (iv) said crystalline fraction (CF) has an ethylene content (C2(CF)), as determined by FT-IR spectroscopy calibrated by quantitative ¹³C-NMR spectroscopy, in the range from 1.0 to 5.0 wt.-% preferably 2.0 to 4.0 wt.-%, more preferably 2.5 to 3.5 wt.-%; and
- 15 (v) said soluble fraction (SF) has an intrinsic viscosity (iV(SF)) in the range from 1.10 to below 1.50 dl/g, preferably 1.25 to 1.45 dl/g; and
- 20
- whereby
- (vi) the mixed-plastics polypropylene blend has inorganic residues as measured by calcination analysis (TGA) according to DIN
- 25 ISO 1172:1996 of 0.05 to 3.0 wt.-%, preferably 0.05 to 2.5 wt.-%, optionally 1.0 to 2.5 wt.-% with respect to the mixed-plastics polypropylene blend; and
- whereby
- (vii) the mixed-plastics polypropylene blend does not contain
- 30 benzene above the detection limit of HS GC-MS 80°C/2h; and
- whereby

- 5 (viii) the mixed-plastics polypropylene blend has a CIELAB color space ($L^*a^*b^*$) of
- L^* from 72.0 to 97.0 preferably from 80.0 to 97.0;
 - a^* from -5.0 to 0.0;
 - b^* from 0.0 to below 22.0; and
- (ix) the mixed-plastics polypropylene blend has a Large Amplitude Oscillatory Shear – Non-Linear Factor [LAOS –NLF] (190°C, 1000%) higher than 2.3, whereby

$$\text{LAOS – NLF} = \left| \frac{G'_1}{G'_3} \right|$$

10

whereby

 G'_1 is the first order Fourier Coefficient G'_3 is the third order Fourier Coefficient.

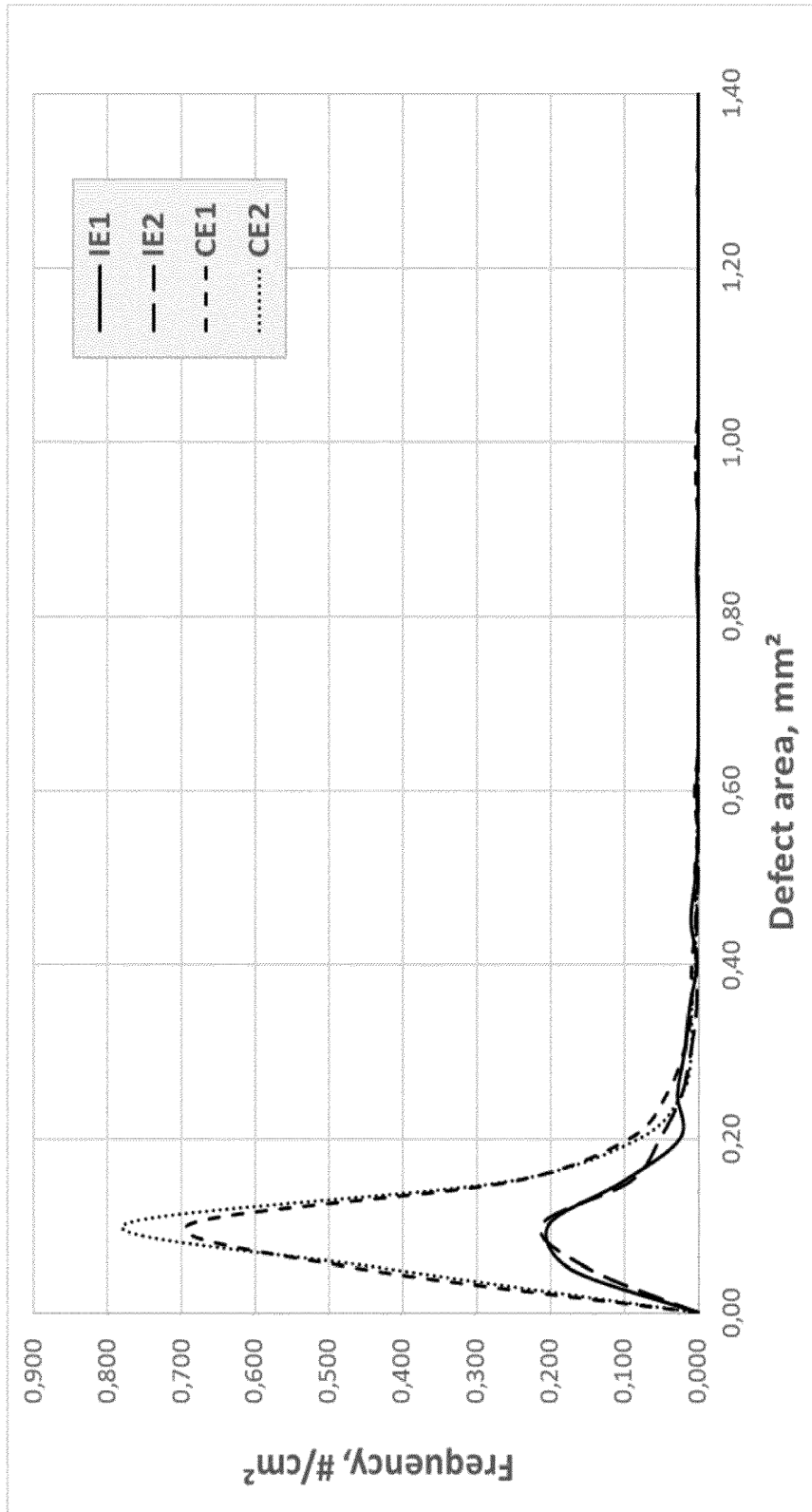
- 15 2. Mixed-plastics polypropylene blend according to claim 1 having a melt flow rate (ISO1133, 2.16kg; 230°C) of 2.0 to 100 g/10min, preferably 5.0 to 80 g/10min, more preferably 10 to 60 g/10min and most preferably 12 to 55 g/10min.
- 20 3. Mixed-plastics polypropylene blend according to claim 1 or 2 being a recycled material.
4. Mixed-plastics polypropylene blend according to any one of the preceding claims containing one or more of the following substances:
- 25 e) polystyrene
- f) polyamide-6
- g) limonene as determined by using solid phase microextraction (HS-SPME-GC-MS)
- h) fatty acids as determined by using solid phase microextraction (HS-SPME-GC-MS).

30

5. Mixed-plastics polypropylene blend according to any one of the preceding claims whereby said soluble fraction (SF) obtained by CRYSTEX QC analysis has an ethylene content (C2(SF)), as determined by FT-IR spectroscopy calibrated by quantitative ¹³C-NMR spectroscopy, in the range from 12.0 to 32.0 wt.-%, preferably 12.0 to 20.0 wt.-%.
6. Mixed-plastics polypropylene blend according to any one of the preceding claims having an odor (VDA270-B3) of 4 or lower, preferably 3 or lower.
7. Mixed-plastics polypropylene blend according to any one of the preceding claims having a Large Amplitude Oscillatory Shear – Non-Linear Factor [LAOS –NLF] (190°C, 1000%) higher than 2.7, whereby
- $$\text{LAOS – NLF} = \left| \frac{G'_1}{G'_3} \right|$$
- whereby
 G'_1 is the first order Fourier Coefficient
 G'_3 is the third order Fourier Coefficient
8. Mixed-plastics polypropylene blend according to any one of the preceding claims having a tensile modulus (ISO 527-2 at a cross head speed of 1 mm/min; 23°C) using injection molded specimens as described in EN ISO 1873-2 (dog bone shape, 4 mm thickness) of at least 1200 MPa, preferably of at least 1250 MPa, more preferably at least 1300 MPa, even more preferably at least 1350 MPa, most preferably at least 1390 MPa.
9. Mixed-plastics polypropylene blend according to any one of the preceding claims having a notched Charpy impact strength (NIS) (1eA) (non-instrumented, ISO 179-1 at +23 °C) according to ISO 179-1 eA at +23 °C on injection moulded specimens of 80 x 10 x 4 mm prepared according to EN ISO 1873-2 of at least 8.0 kJ/m², preferably 8.3 kJ/m²,

- whereby further preferably said soluble fraction (SF) obtained by CRYSTEX QC analysis has an ethylene content (C2(SF)), as determined by FT-IR spectroscopy calibrated by quantitative ¹³C-NMR spectroscopy, in the range from 12.0 to 20.0 wt.-% and further
- 5 preferably the mixed-plastics polypropylene blend has a CIELAB color space (L*a*b) of
- L* from 72.0 to 97.0, preferably from 80.0 to 97.0;
 - a* from -5.0 to 0.0;
 - b* from 0.0 to below 22.0
- 10 and further a crystalline fraction (CF) content determined according to CRYSTEX QC analysis in the range from 91.0 to 94.0 wt.-% and a soluble fraction (SF) content determined according to CRYSTEX QC analysis of 6.0 to 9.0 wt.-%.
- 15 10. Mixed-plastics polypropylene blend according to any one of the preceding claims being in the form of pellets.
11. Mixed-plastics polypropylene blend according to any one of the preceding claims being visbroken by peroxides.
- 20 12. Molded article made from the mixed-plastics polypropylene blend according to any one of the preceding claims.
13. Blend containing the mixed-plastics polypropylene blend according to
- 25 any one of the preceding claims 1 to 11 and at least one virgin polyolefin.
14. Blend according to claim 13 being a heterophasic polypropylene.
- 30 15. Use of the mixed-plastics polypropylene blend according to any one of claims 1 to 10 for packaging and/or in the medical field.

Figure 1



INTERNATIONAL SEARCH REPORT

International application No PCT/EP2022/057953
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A. CLASSIFICATION OF SUBJECT MATTER INV. C08L23/10 ADD.				
According to International Patent Classification (IPC) or to both national classification and IPC				
B. FIELDS SEARCHED				
Minimum documentation searched (classification system followed by classification symbols) C08L B29B				
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, WPI Data				
C. DOCUMENTS CONSIDERED TO BE RELEVANT				
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.		
X	EP 3 502 177 A1 (BOREALIS AG [AT]) 26 June 2019 (2019-06-26) page 20; example HECO; table 1 page 21; example IE1; tables 3,4 claim 5 paragraphs [0003], [0035] - [0045], [0110], [0111], [0120] - [0120] -----	1-15		
X	WO 2018/046578 A1 (DER GRUENE PUNKT DUALES SYSTEM DEUTSCHLAND GMBH [DE]) 15 March 2018 (2018-03-15) cited in the application the whole document -----	1-15		
<input type="checkbox"/> Further documents are listed in the continuation of Box C. <input checked="" type="checkbox"/> See patent family annex.				
* Special categories of cited documents : <table style="width: 100%; border: none;"> <tr> <td style="width: 50%; border: none; vertical-align: top;"> "A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier application or patent but published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed </td> <td style="width: 50%; border: none; vertical-align: top;"> "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art "&" document member of the same patent family </td> </tr> </table>			"A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier application or patent but published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art "&" document member of the same patent family
"A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier application or patent but published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art "&" document member of the same patent family			
Date of the actual completion of the international search 6 July 2022		Date of mailing of the international search report 18/07/2022		
Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016		Authorized officer Lippert, Sigrid		

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/EP2022/057953

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