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(54) Title: POLYPROPYLENE COMPOSITION WITH LOW SHRINKAGE AND BALANCED MECHANICAL PROPERTIES

(57) Abstract: The present invention relates to a polypropylene composition comprising a propylene homopolyrner, two heterophasic propylene copolymers, an elastomeric ethylene copolymer and inorganic filler having reduced shrinkage and balanced mechanical properties.

Polypropylene composition with low shrinkage and balanced mechanical properties

The present invention is directed to a polyolefin composition having low shrinkage and balanced mechanical properties including high impact, strength, and stiffness.

High impact polystyrenes have been widely applied in the fields of household appliances, medical appliances, automotives, pipes and toys due to a favorable combination of mechanical properties including comprehensive high impact, high strength and high stiffness together with oil resistance, water resistance and electrical insulation.

According to a recent trend, high impact polystyrenes are at least gradually replaced by polypropylenes in order to lower production cost. One reason is that high impact polystyrenes are made from crude oil while polypropylenes can be made from cheaper sources including natural gas or shale gas.

While polypropylenes typically offer an attractive combination of mechanical properties including high impact strength and stiffness, polypropylenes usually exhibit greater shrinkage during molding processes than high impact polystyrenes.

Accordingly, there is the objective of developing improved polypropylene compositions which have reduced shrinkage while the balanced set of other important mechanical properties is maintained.

The foregoing and other objectives are solved by the subject-matter of the present invention.

The specific finding of the present invention is to provide a polypropylene composition (PP) comprising,

(a) a propylene homopolymer (homo-PP) having a melt flow rate MFR_2 (230°C, 2,16 kg) measured according to ISO 1133 of from 20.0 to 70.0 g/10 min,

(b) a first heterophasic propylene copolymer (HECO-1) having a melt flow rate MFR_2 (230 °C, 2.16 kg) measured according to ISO 1133 in the range of from 0.1 to 2.5 g/10min,

(c) a second heterophasic propylene copolymer (HECO-2) having a melt flow rate MFR_2 (230°C, 2.16 kg) measured according to ISO 1133 in the range of from 8.0 to 15.0 g/10min,

(d) an elastomeric ethylene copolymer (EEC) having a melt flow rate MFR_2 (190°C, 2,16 kg) measured according to ISO 1133 in the range of from 0.5 to 50.0 g/10min, and

(e) an inorganic filler (F).

In a preferred embodiment of the present invention,

(a) the first heterophasic propylene copolymer (HECO-1) has

(i) a xylene cold soluble (XCS) fraction measured according to ISO 16152 (25 °C) of from 8.0 to 20.0 wt.-%, based on the total weight of the heterophasic propylene copolymer (HECO-1), and/or

(ii) a comonomer content, preferably an ethylene content, of equal or below 7.0 wt.-%, based on the total weight of the heterophasic propylene copolymer (HECO-1), and/or

(b) the second heterophasic propylene copolymer (HECO-2) has

(i) a xylene cold soluble (XCS) fraction measured according to ISO 16152 (25 °C) of from 15.0 to 45.0 wt.-%, based on the total weight of the heterophasic propylene copolymer (HECO-2), and/or

(ii) a comonomer content, preferably an ethylene content, of 10.0 to 22.0 wt.-%, based on the total weight of the heterophasic propylene copolymer (HECO-2).

In another preferred embodiment of the present invention,

(a) the xylene cold soluble (XCS) fraction of the first heterophasic propylene copolymer (HECO-1) has

(i) an intrinsic viscosity (IV) of from 2.5 to 4.5 dl/g, and/or

(ii) a comonomer content, preferably an ethylene content, of 25.0 to 41.0 wt.-% , and/or

(b) the xylene cold soluble (XCS) fraction of the second heterophasic propylene copolymer (HECO-2) has

(i) an intrinsic viscosity (IV) of from 2.0 to 4.0 dl/g, and/or

(ii) a comonomer content, preferably an ethylene content, of 30.0 to 45.0 wt.-%.

In another preferred embodiment of the present invention, the polypropylene composition (PP) comprises

(a) 10.0 to 30.0 wt.-%, preferably15.0 to 25.0 wt.-%, of the propylene homopolymer (homo-PP), and/or

(b) 5.0 to 25.0 wt.-%, preferably10.0 to 20.0 wt.-%, of the first heterophasic propylene copolymer (HECO-1), and/or

(c) 10.0 to 45.0 wt.-%, preferably15.0 to 20.0 wt.-%, of the second heterophasic propylene copolymer (HECO-2), and/or

(d) 3.0 to 10.0 wt.-%, preferably 4.0 to 9.0 wt.-%, of the elastomeric ethylene copolymer (EEC), and/or

(e) 25.0 to 45.0 wt.-%, preferabl 30.0 to 40.0 wt.-%, of the inorganic filler (F)

based on the total amount of the polypropylene composition (PP).

In another preferred embodiment of the present invention, the comonomer content, preferably the ethylene content, of the xylene cold soluble (XCS) fraction of the second heterophasic propylene copolymer (HECO-2) is at least 3.0 wt-%, higher than the comonomer content, preferably the ethylene content, of the xylene cold soluble (XCS) fraction of the first heterophasic propylene copolymer (HECO-1).

In another preferred embodiment of the present invention, the intrinsic viscosity of the xylene cold soluble (XCS) fraction of the second heterophasic propylene copolymer (HECO-2) is at least 0.3 dl/g, preferably at

least 0.5dl/g, lower than the intrinsic viscosity (IV) of the xylene cold soluble (XCS) fraction of the first heterophasic propylene copolymer (HECO-1).

In another preferred embodiment of the present invention, the propylene homopolymer (homo-PP) has a melt flow rate MFR_2 (230°C) which is at least 3.0 g/10min lower than the melt flow rate MFR_2 (230°C) of the polypropylene matrix (M), i.e. the propylene homopolymer (H-PP2), of the second heterophasic propylene copolymer (HECO-2).

In another preferred embodiment of the present invention, the weight ratio of the combined heterophasic propylene copolymers (HECO-1) and (HECO-2) to the inorganic filler (F) [HECO1+HECO-2 / F] is from 2.0 to 0.3.

In another preferred embodiment of the present invention, the weight ratio of the propylene homopolymer (homo-PP) to the inorganic filler (F) [homo-PP/F] is from 1.2 to 0.2.

In another preferred embodiment of the present invention, the polypropylene composition (PP) is used for the production of household articles, medical articles, automotive articles, pipes and toys.

It has been surprisingly found out that the polypropylene composition (PP) according to the present invention exhibits reduced shrinkage while having balanced mechanical properties including high impact strength and stiffness as well as excellent tensile strength and flexural modulus.

In the following, the invention and all of its components are described in more detail.

When in the following reference is made to preferred embodiments or technical details of the inventive polypropylene composition (PP), it is to be understood that these preferred embodiments or technical details also refer to the inventive article comprising the polypropylene composition (PP).

Propylene homopolymer (homo-PP)

One essential component of the polypropylene composition (PP) according to the present invention is a propylene homopolymer (homo-PP).

It is appreciated that the polypropylene composition (PP) comprises the propylene homopolymer (homo-PP) in an amount of from 10.0 to 30.0 wt.-%, preferably of from 15.0 to 25.0 wt.-%, and even more preferably of from 18.0 to 23.0 wt.-% based on the total weight of the polypropylene composition (PP).

It comes apparent from the wording that the propylene homopolymer (homo-PP) is not a heterophasic polymer, i.e. a system comprising a crystalline matrix phase in which an elastomeric phase is dispersed. Accordingly, it is preferred that the propylene homopolymer (homo-PP) is monophasic, i.e. in DMTA no multiphase structure can be identified as there exists just one glass transition temperature.

Further, the propylene homopolymer (homo-PP) preferably has a melting temperature of more than 155°C, i.e. of more than 155 to 169 °C, more preferably of at least 158°C, i.e. in the range of from 158 to 168 °C, still more preferably in the range of from 162 to 168°C.

Preferably, a further characteristic of the propylene homopolymer (homo-PP) is the low amount of misinsertions of propylene within the polymer chain, which indicates that the propylene homopolymer (homo-PP) is produced in the presence of a Ziegler-Natta catalyst. Accordingly the propylene homopolymer (homo-PP) is preferably featured by low amount of 2,1 erythro regio-defects, i.e. of equal or below 0.4 mol.-%, more preferably of equal or below than 0.2 mol.-%, like of not more than 0.1 mol.-%, determined by ^{13}C-NMR spectroscopy. In an especially preferred embodiment no 2,1 erythro regio-defects are detectable.

The propylene homopolymer (homo-PP) preferably has a melt flow rate MFR_2 (230°C) measured according to ISO 1133 in the range of from 20.0 to 70.0 g/10min, preferably in the range of from 30.0 to 60.0 g/10 min, preferably in the range of from 45.0 to 55.0 g/10min, like in the range of 48.0 to 52.0 g/10min.

The propylene homopolymer (homo-PP) can be chemically identical to the matrices (M) of the two essential heterophasic propylene copolymers (HECO-1) and/or (HECO-2) which are described in more detail below. In another preferred embodiment the propylene homopolymer (homo-PP) is chemical different, prefereably different in the melt flow rate, to the matrix, i.e. to the propylene homopolymer (H-PP-1), of first

heterophasic propylene copolymer (HECO-1), however can be chemically identical or different to the matrix, i.e. to the propylene homopolymer (H-PP-2), of second heterophasic propylene copolymer (HECO-2).

Thus in one preferred embodiment the propylene homopolymer (homo-PP) has a melt flow rate MFR_2 (230°C) which is higher, preferably at least 10 g/10min higher, more preferably at least 20 g/10min higher, still more preferably 20 to 65 g/10min higher, yet more preferably at least 30 to 55 g/10min higher, than the matrix, i.e. the propylene homopolymer (H-PP-1), of first heterophasic propylene copolymer (HECO-1), and optionally said propylene homopolymer (homo-PP) has a melt flow rate MFR_2 (230°C) which is similar, e.g. +/- 5 g/10min, like +/- 2 g/10min, to the melt flow rate MFR_2 (230°C) of the matix (M), i.e. of the propylene homopolymer (H-PP-2), of the second heterophasic propylene copolymer (HECO-2).

Preferably, the propylene homopolymer (homo-PP) has a tensile modulus measured according to ISO 527-2 of at least 1,400 MPa, more preferably of at least 1,500 MPa, like in the range of 1,500 to 1,800 MPa.

The propylene homopolymer (homo-PP) is known in the art and is preferably made with a Ziegler-Natta catalyst.

As already mentioned above, the polypropylene composition (PP) according to the present invention further comprises two specific heterophasic propylene copolymers.

The expression "heterophasic" indicates that an elastomeric copolymer is (finely) dispersed in a matrix. In other words, the elastomeric propylene copolymer forms inclusions in the matrix. Thus, the matrix contains (finely) dispersed inclusions being not part of the matrix and said inclusions contain the elastomeric propylene copolymer. The term "inclusion" according to this invention shall preferably indicate that the matrix and the inclusion form different phases within the heterophasic propylene copolymer, said inclusions are for instance visible by high resolution microscopy, like electron microscopy or scanning force microscopy.

The final composition is probably of a complex structure. For example, the matrix of the heterophasic propylene copolymer may form a continuous phase being the matrix of the composition wherein the elastomeric copolymers and optional additives form together or individually inclusions dispersed therein.

The heterophasic propylene copolymers ensure high impact and other basic mechanical property, such as tensile strength and flexural modulus.

First heterophasic propylene copolymer (HECO-1)

As stated above, the polyolefin composition according to the present invention comprises a first heterophasic propylene copolymer (HECO-1) as an essential component. It is preferred that the first heterophasic propylene copolymer (HECO-1) before mixed with the other components mentioned herein comprises as polymer components only the matrix polypropylene (PP-1) and dispersed therein the elastomeric propylene copolymer (E-1). In other words the first heterophasic propylene copolymer (HECO-1) may contain further additives but no other polymer in an amount exceeding 5 wt-%, more preferably exceeding 3 wt.-%, like exceeding 1 wt.-%, based on the total amount of the first heterophasic propylene copolymer (HECO-1), more preferably based on the polymers present in the first heterophasic propylene copolymer (HECO-1). One additional polymer which may be present in such low amounts is a polyethylene which is a reaction product obtained by the preparation of the first heterophasic propylene copolymer (HECO-1). Accordingly, it is in particular appreciated that a first heterophasic propylene copolymer (HECO-1) as defined in the instant invention contains only a polypropylene (PP-1), an elastomeric propylene copolymer (E-1) and optionally a polyethylene in amounts as mentioned in this paragraph.

One important aspect of the instant invention is that the first heterophasic propylene copolymer (HECO-1) has a rather low melt flow rate, i.e. has a melt flow rate MFR_2 (230 °C) in the range of 0.05 to 4.0 g/10min, more preferably in the range of 0.10 to 3.0 g/10min, yet more preferably in the range of 0.15 to 2.0 g/10 min, still even more preferably in the range of 0.15 to 1.5 g/10min.

Preferably the propylene content in the first heterophasic propylene copolymer (HECO-1) is at least 93.0 wt.-%, more preferably 93.0 to 97.5 wt-%, more preferably 93.0 to 97.0 wt-%, and even more preferably 95.0 to 96.5 wt-% based on the total amount of the first heterophasic propylene copolymer (HECO-1), more preferably based on the amount of the polymer components of the first heterophasic propylene copolymer (HECO-1), yet more preferably based on the amount of the polypropylene (PP-1) and the elastomeric propylene copolymer (E-1) together.

The remaining part constitutes the comonomers, like ethylene, as defined for the polypropylene (PP-1) being a propylene copolymer (R-PP-1) and the elastomeric propylene copolymer (E-1), respectively. Accordingly, the comonomer content, preferably ethylene content, is preferably equal or lower than 7.0 wt.-%, more preferably in the range of from 2.5 to 7.0 wt-%, yet more preferably in the range of from 3.0 to 7.0 wt-% and even more preferably in the range of from 3.5 to 5.0 wt-% based on the total weight of the heterophasic

propylene copolymer (HECO-1), more preferably based on the amount of the polymer components of the first heterophasic propylene copolymer (HECO-1), yet more preferably based on the amount of the polypropylene (PP-1) and the elastomeric propylene copolymer (E-1) together.

As stated above the matrix of the first heterophasic propylene copolymer (HECO-1) is the polypropylene (PP-1).

The polypropylene (PP-1) according to this invention constituting the matrix of the first heterophasic copolymer (HECO-1) shall have a melt flow rate MFR_2 (230 °C) in the range of from 0.05 to 4.0 g/10min, more preferably in the range of 0.10 to 3.0 g/10min, yet more preferably in the range of 0.15 to 2.0 g/10 min, still even more preferably in the range of 0.15 to 1.5 g/10min.

The polypropylene (PP-1) can be a propylene copolymer (R-PP-1) or a propylene homopolymer (H-PP-1), the latter is preferred.

Accordingly, it is appreciated that the polypropylene (PP-1) has a comonomer content equal or below 7.0 wt-%, still more preferably equal or below 5.0 wt-%.

The expression propylene homopolymer, e.g. the expressions propylene homopolymer (H-PP-1),propylene homopolymer (H-PP-2) and propylene homopolymer (homo-PP) used in the instant invention relates to a polypropylene that consists substantially, i.e. of more than 99.5 wt-%, such as at least 99.6 wt-%, still more preferably of at least 99.7 wt-%, like of at least 99.8 wt-%, of propylene units. In case other monomeric units are present in minor amounts, the units are selected from ethylene and/or a C_4 to C_{12} α-olefin as described below. In a preferred embodiment only propylene units in the propylene homopolymer are detectable.

In case the polypropylene (PP-1) is a propylene copolymer (R-PP-1) it comprises monomers copolymerizable with propylene, for example comonomers such as ethylene and/or C_4 to C_{12} α-olefins, in particular ethylene and/or C_4 to C_{10} α-olefins, e.g. 1-butene and/or 1-hexene. Preferably the propylene copolymer (R-PP-1) comprises, especially consists of, monomers copolymerizable with propylene from the group consisting of ethylene, 1-butene and 1-hexene. More specifically the propylene copolymer (R-PP-1) comprises - apart from propylene - units derivable from ethylene and/or 1-butene. In a preferred embodiment the propylene copolymer (R-PP-1) comprises units derivable from ethylene and propylene only. The comonomer content in the propylene copolymer (R-PP-1) is preferably in the range of more than 1.0 to 9.0 wt-%, still more preferably in the range of more than 1.0 to 7.0 wt-%.

The polypropylene (PP-1) can have a xylene cold soluble content (XCS) in a broad range, i.e. up to 5.0 wt.-%. Accordingly the polypropylene (PP-1) may have a xylene cold soluble content (XCS) in the range of 0.3 to 5.0 wt.-%, preferably in the range of 0.5 to 4.5 wt.-%, like in the range of 1.0 to 4.0 wt.-%.

However, in preferred embodiments, the polypropylene (PP-1), in particular in case the polypropylene (PP-1) is a propylene homopolymer (H-PP-1), has a xylene cold soluble (XCS) content in the range of 0.5 to 5.0 wt.-%, more preferably in the range of 1.0 to 4.0 wt.-%, still more preferably of 1.0 to 3.0 wt.-%.

One further essential component of the first heterophasic propylene copolymer (HECO-1) is its elastomeric propylene copolymer (E-1).

The elastomeric propylene copolymer (E-1) preferably comprises monomers copolymerizable with propylene, for example comonomers such as ethylene and/or C_4 to C_{12} α-olefins, in particular ethylene and/or C_4 to C_{10} α-olefins, e.g. 1-butene and/or 1-hexene. Preferably the elastomeric propylene copolymer (E-1) comprises, especially consists of, monomers copolymerizable with propylene from the group consisting of ethylene, 1-butene and 1-hexene. More specifically the elastomeric propylene copolymer (E-1) comprises – apart from propylene – units derivable from ethylene and/or 1-butene. Thus in an especially preferred embodiment the elastomeric propylene copolymer phase (E-1) comprises units derivable from ethylene and propylene only.

In case the polypropylene (PP-1) is a propylene copolymer (R-PP-1) it is preferred that the comonomer(s) of the propylene copolymer (R-PP-1) and the elastomeric propylene copolymer (E-1) are the same.

The properties of the elastomeric propylene copolymer phase (E-1) are mainly influenced by the xylene cold soluble (XCS) content of the first heterophasic propylene copolymer (HECO-1). Thus, according to the present invention the xylene cold soluble (XCS) fraction of the first heterophasic propylene copolymer (HECO-1) is regarded as the elastomeric propylene copolymer (E-1) of the first heterophasic propylene copolymer (HECO-1).

Accordingly, the amount of the elastomeric propylene copolymer (E-1), i.e. of the xylene cold soluble (XCS) fraction, of the first heterophasic propylene copolymer (HECO-1) preferably is in the range of from 8.0 to 20.0 wt-%, more preferably in the range of from 10.0 to 18.0 wt-% and even more preferably in the range of

from 11.0 to 15.0 wt-%.These values are based on the first heterophasic propylene copolymer (HECO-1) and not on the total polypropylene composition (PP).

One important requirement of the present invention is that the elastomeric propylene copolymer (E-1) has a balanced weight average molecular weight. Accordingly the intrinsic viscosity must be carefully chosen.

Low intrinsic viscosity (IV) values reflect a low weight average molecular weight. Thus it is appreciated that the elastomeric propylene copolymer phase (E-1), i.e. the xylene cold soluble fraction (XCS) of the first heterophasic propylene copolymer (HECO-1), has an intrinsic viscosity (IV) determined according to DIN ISO 1628/1 (in decaline at 135 °C) in the range of from 2.5 to 5.0 dl/g, more preferably in the range of from equal or more than 2.8 to 4.5 dl/g, and still even more preferably in the range of equal or more than from 3.0 to 4.0 dl/g, like in the range of 3.3 to 3.8 dl/g.

The comonomer content, preferably the ethylene content, within the elastomeric propylene copolymer phase (E-1) shall be preferably also in a specific range. Accordingly in a preferred embodiment the comonomer content, more preferably ethylene content, of the elastomeric propylene copolymer (E-1), i.e. of the xylene cold soluble fraction (XCS) of the first heterophasic propylene copolymer (HECO-1), is in the range of from 25.0 to 41.0 wt-%, still more preferably in the range of from 28.0 to 38.0 wt-%, yet more preferably in the range of from 30.0 to 38.0 wt-%.

Accordingly, it is appreciated that the propylene content of the elastomeric propylene copolymer (E-1), i.e. of the xylene cold soluble fraction (XCS) of the first heterophasic propylene copolymer (HECO-1), is preferably in the range of from 59.0 to 75.0 wt -%, still more preferably in the range of 62.0 to 72.0 wt-%, yet more preferably in the range of from 62.0 to 70.0 wt-%.

As will be explained below, the first heterophasic polypropylene (HECO-1) as well its individual components (matrix and elastomeric copolymer) can be produced by blending different polymer types, i.e. of different molecular weight and/or comonomer content. However it is preferred that the first heterophasic polypropylene (HECO-1) as well its individual components (matrix and elastomeric copolymer) are produced in a sequential step process, using reactors in serial configuration and operating at different reaction conditions. As a consequence, each fraction prepared in a specific reactor will have its own molecular weight distribution and/or comonomer content distribution.

The first heterophasic propylene copolymer (HECO-1) according to this invention is commercially available. Accordingly a skilled person in the art is in a position to produce a first heterophasic propylene copolymer (HECO-1) as defined herein.

The first heterophasic propylene copolymer (HECO-1) according to this invention is preferably produced in a sequential polymerization process, i.e. in a multistage process, known in the art, wherein the polypropylene (PP-1) is produced at least in one slurry reactor, preferably in a slurry reactor and optionally in a subsequent gas phase reactor, and subsequently the elastomeric propylene copolymer (E-1) is produced at least in one, i.e. one or two, gas phase reactor(s).

Accordingly it is preferred that the first heterophasic propylene copolymer (HECO-1) is produced in a sequential polymerization process comprising the steps of

(a) polymerizing propylene and optionally at least one of ethylene and/or C_4 to C_{12} α-olefin in a first reactor (R1) obtaining the first polypropylene fraction of the polypropylene (PP-1), preferably said first polypropylene fraction is a first propylene homopolymer,

(b) transferring the first polypropylene fraction into a second reactor (R2),

(c) polymerizing in the second reactor (R2) and in the presence of said first polypropylene fraction propylene and optionally at least one of ethylene and/or C_4 to C_{12} α-olefin obtaining thereby the second polypropylene fraction, preferably said second polypropylene fraction is a second propylene homopolymer, said first polypropylene fraction and said second polypropylene fraction form the polypropylene (PP-1), i.e. the matrix of the heterophasic propylene copolymer (HECO-1),

(d) transferring the polypropylene (PP-1) of step (c) into a third reactor (R3),

(e) polymerizing in the third reactor (R3) and in the presence of the polypropylene (PP-1) obtained in step (c) propylene and at least one of ethylene and/or C_4 to C_{12} α-olefin obtaining thereby a first elastomeric propylene copolymer fraction, the first elastomeric propylene copolymer fraction is dispersed in the polypropylene (PP-1),

(f) transferring the polypropylene (PP-1) in which the first elastomeric propylene copolymer fraction is dispersed in a fourth reactor (R4), and

(g) polymerizing in the fourth reactor (R4) and in the presence of the mixture obtained in step (e) propylene and at least one of ethylene and/or C_4 to C_{12} α-olefin obtaining thereby the second elastomeric propylene copolymer fraction,

the polypropylene (PP-1), the first elastomeric propylene copolymer fraction, and the second elastomeric propylene copolymer fraction form the heterophasic propylene copolymer (HECO-1).

Alternatively the elastomeric propylene copolymer (E) can be also produced in one gas phase reactor, i.e. the fourth reactor (R4) is optional.

Of course, in the first reactor (R1) the second polypropylene fraction can be produced and in the second reactor (R2) the first polypropylene fraction can be obtained. The same holds true for the elastomeric propylene copolymer phase. Accordingly in the third reactor (R3) the second elastomeric propylene copolymer fraction can be produced whereas in the fourth reactor (R4) the first elastomeric propylene copolymer fraction is made.

Preferably between the second reactor (R2) and the third reactor (R3) and optionally between the third reactor (R3) and fourth reactor (R4) the monomers are flashed out.

The term "sequential polymerization process" indicates that the first heterophasic propylene copolymer (HECO-1) is produced in at least two, like three or four reactors connected in series. Accordingly the present process comprises at least a first reactor (R1) and a second reactor (R2), more preferably a first reactor (R1), a second reactor (R2), a third reactor (R3) and a fourth reactor (R4). The term "polymerization reactor" shall indicate that the main polymerization takes place. Thus in case the process consists of four polymerization reactors, this definition does not exclude the option that the overall process comprises for instance a pre-polymerization step in a pre-polymerization reactor. The term "consist of" is only a closing formulation in view of the main polymerization reactors.

The first reactor (R1) is preferably a slurry reactor (SR) and can be any continuous or simple stirred batch tank reactor or loop reactor operating in bulk or slurry. Bulk means a polymerization in a reaction medium that comprises of at least 60 % (w/w) monomer. According to the present invention the slurry reactor (SR) is preferably a (bulk) loop reactor (LR).

The second reactor (R2), the third reactor (R3) and the fourth reactor (R4) are preferably gas phase reactors (GPR). Such gas phase reactors (GPR) can be any mechanically mixed or fluid bed reactors. Preferably the gas phase reactors (GPR) comprise a mechanically agitated fluid bed reactor with gas velocities of at least 0.2 m/sec. Thus it is appreciated that the gas phase reactor is a fluidized bed type reactor preferably with a mechanical stirrer.

Thus in a preferred embodiment the first reactor (R1) is a slurry reactor (SR), like a loop reactor (LR), whereas the second reactor (R2), the third reactor (R3) and the fourth reactor (R4) are gas phase reactors

(GPR). Accordingly for the instant process at least four, preferably four polymerization reactors, namely a slurry reactor (SR), like a loop reactor (LR), a first gas phase reactor (GPR-1), a second gas phase reactor (GPR-2) and a third gas phase reactor (GPR-3) connected in series are used. If needed prior to the slurry reactor (SR) a pre-polymerization reactor is placed. Alternatively, the third gas phase reactor (GPR-3) is optional, i.e. three reactors (LR, GPR-1, GPR-2) are used.

A preferred multistage process is a "loop-gas phase"-process, such as developed by Borealis A/S, Denmark (known as BORSTAR® technology) described e.g. in patent literature, such as in EP 0 887 379, WO 92/12182 WO 2004/000899, WO 2004/111095, WO 99/24478, WO 99/24479 or in WO 00/68315.

A further suitable slurry-gas phase process is the Spheripol® process of Basell.

Preferably, in the instant process for producing the first heterophasic propylene copolymer (HECO-1) as defined above the conditions for the first reactor (R1), i.e. the slurry reactor (SR), like a loop reactor (LR), of step (a) may be as follows:

- the temperature is within the range of 50 °C to 110 °C, preferably between 60 °C and 100 °C, more preferably between 68 and 95 °C,
- the pressure is within the range of 20 bar to 80 bar, preferably between 40 bar to 70 bar,
- hydrogen can be added for controlling the molar mass in a manner known per se.

Subsequently, the reaction mixture from step (a) is transferred to the second reactor (R2), i.e. gas phase reactor (GPR-1), i.e. to step (c), whereby the conditions in step (c) are preferably as follows:

- the temperature is within the range of 50 °C to 130 °C, preferably between 60 °C and 100 °C,
- the pressure is within the range of 5 bar to 50 bar, preferably between 15 bar to 35 bar,
- hydrogen can be added for controlling the molar mass in a manner known per se.

The condition in the third reactor (R3) and the fourth reactor (R4), preferably in the second gas phase reactor (GPR-2) and third gas phase reactor (GPR-3), is similar to the second reactor (R2).

The residence time can vary in the three reactor zones.

In one embodiment of the process for producing the polypropylene the residence time in bulk reactor, e.g. loop is in the range 0.1 to 2.5 hours, e.g. 0.15 to 1.5 hours and the residence time in gas phase reactor will generally be 0.2 to 6.0 hours, like 0.5 to 4.0 hours.

If desired, the polymerization may be effected in a known manner under supercritical conditions in the first reactor (R1), i.e. in the slurry reactor (SR), like in the loop reactor (LR), and/or as a condensed mode in the gas phase reactors (GPR).

Preferably the process comprises also a prepolymerization with the catalyst system, as described in detail below, comprising a Ziegler-Natta procatalyst, an external donor and optionally a cocatalyst.

In a preferred embodiment, the prepolymerization is conducted as bulk slurry polymerization in liquid propylene, i.e. the liquid phase mainly comprises propylene, with minor amount of other reactants and optionally inert components dissolved therein.

The prepolymerization reaction is typically conducted at a temperature of 10 to 60 °C, preferably from 15 to 50 °C, and more preferably from 20 to 45 °C.

The pressure in the prepolymerization reactor is not critical but must be sufficiently high to maintain the reaction mixture in liquid phase. Thus, the pressure may be from 20 to 100 bar, for example 30 to 70 bar.

The catalyst components are preferably all introduced to the prepolymerization step. However, where the solid catalyst component (i) and the cocatalyst (ii) can be fed separately it is possible that only a part of the cocatalyst is introduced into the prepolymerization stage and the remaining part into subsequent polymerization stages. Also in such cases it is necessary to introduce so much cocatalyst into the prepolymerization stage that a sufficient polymerization reaction is obtained therein.

It is possible to add other components also to the prepolymerization stage. Thus, hydrogen may be added into the prepolymerization stage to control the molecular weight of the prepolymer as is known in the art. Further, antistatic additive may be used to prevent the particles from adhering to each other or to the walls of the reactor.

The precise control of the prepolymerization conditions and reaction parameters is within the skill of the art.

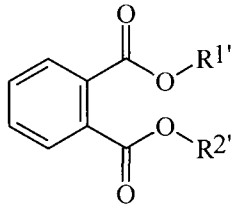
According to the invention the first heterophasic propylene copolymer (HECO-1) is obtained by a multistage polymerization process, as described above, in the presence of a catalyst system comprising as component (i)

a Ziegler-Natta procatalyst which contains a trans-esterification product of a lower alcohol and a phthalic ester.

The procatalyst used according to the invention for preparing the first heterophasic propylene copolymer (HECO-1) is prepared by

a) reacting a spray crystallized or emulsion solidified adduct of $MgCl_2$ and a C_1-C_2 alcohol with $TiCl_4$

b) reacting the product of stage a) with a dialkylphthalate of formula (I)



(I)

wherein $R^{1'}$ and $R^{2'}$ are independently at least a C_5 alkyl

under conditions where a transesterification between said C_1 to C_2 alcohol and said dialkylphthalate of formula (I) takes place to form the internal donor

c) washing the product of stage b) or

d) optionally reacting the product of step c) with additional $TiCl_4$.

The procatalyst is produced as defined for example in the patent applications WO 87/07620, WO 92/19653, WO 92/19658 and EP 0 491 566. The content of these documents is herein included by reference.

First an adduct of $MgCl_2$ and a C_1-C_2 alcohol of the formula $MgCl_2$*nROH, wherein R is methyl or ethyl and n is 1 to 6, is formed. Ethanol is preferably used as alcohol.

The adduct, which is first melted and then spray crystallized or emulsion solidified, is used as catalyst carrier.

In the next step the spray crystallized or emulsion solidified adduct of the formula $MgCl_2$*nROH, wherein R is methyl or ethyl, preferably ethyl and n is 1 to 6, is contacting with $TiCl_4$ to form a titanized carrier, followed by the steps of

- adding to said titanised carrier
  - (i) a dialkylphthalate of formula (I) with $R^{1'}$ and $R^{2'}$ being independently at least a C_5-alkyl, like at least a C_8-alkyl,

  or preferably

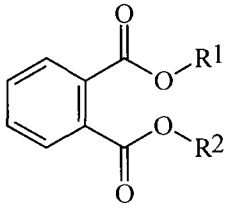
(ii) a dialkylphthalate of formula (I) with $R^{1'}$ and $R^{2'}$ being the same and being at least a C_5-alkyl, like at least a C_8-alkyl,

or more preferably

(iii) a dialkylphthalate of formula (I) selected from the group consisting of propylhexylphthalate (PrHP), dioctylphthalate (DOP), di-iso-decylphthalate (DIDP), and ditridecylphthalate (DTDP), yet more preferably the dialkylphthalate of formula (I) is a dioctylphthalate (DOP), like di-iso-octylphthalate or diethylhexylphthalate, in particular diethylhexylphthalate,

to form a first product,

- subjecting said first product to suitable transesterification conditions, i.e. to a temperature above 100 °C, preferably between 100 to 150 °C, more preferably between 130 to 150 °C, such that said methanol or ethanol is transesterified with said ester groups of said dialkylphthalate of formula (I) to form preferably at least 80 mol-%, more preferably 90 mol-%, most preferably 95 mol.-%, of a dialkylphthalate of formula (II)



(II)

with R^1 and R^2 being methyl or ethyl, preferably ethyl,

the dialkylphthalat of formula (II) being the internal donor and

- recovering said transesterification product as the procatalyst composition (component (i)).

The adduct of the formula $MgCl_2$*nROH, wherein R is methyl or ethyl and n is 1 to 6, is in a preferred embodiment melted and then the melt is preferably injected by a gas into a cooled solvent or a cooled gas, whereby the adduct is crystallized into a morphologically advantageous form, as for example described in WO 87/07620.

This crystallized adduct is preferably used as the catalyst carrier and reacted to the procatalyst useful in the present invention as described in WO 92/19658 and WO 92/19653.

As the catalyst residue is removed by extracting, an adduct of the titanised carrier and the internal donor is obtained, in which the group deriving from the ester alcohol has changed.

In case sufficient titanium remains on the carrier, it will act as an active element of the procatalyst.

Otherwise the titanization is repeated after the above treatment in order to ensure a sufficient titanium concentration and thus activity.

Preferably the procatalyst used according to the invention contains 2.5 wt.-% of titanium at the most, preferably 2.2% wt.-% at the most and more preferably 2.0 wt.-% at the most. Its donor content is preferably between 4 to 12 wt.-% and more preferably between 6 and 10 wt.-%.

More preferably the procatalyst used according to the invention has been produced by using ethanol as the alcohol and dioctylphthalate (DOP) as dialkylphthalate of formula (I), yielding diethyl phthalate (DEP) as the internal donor compound.

Still more preferably the catalyst used according to the invention is the catalyst as described in the example section; especially with the use of dioctylphthalate as dialkylphthalate of formula (I).

For the production of the heterophasic propylene copolymer (HECO-1) according to the invention the catalyst system used preferably comprises in addition to the special Ziegler-Natta procatalyst an organometallic cocatalyst as component (ii).

Accordingly it is preferred to select the cocatalyst from the group consisting of trialkylaluminium, like triethylaluminium (TEA), dialkyl aluminium chloride and alkyl aluminium sesquichloride.

Component (iii) of the catalysts system used is an external donor represented by formula (IIIa) or (IIIb). Formula (IIIa) is defined by

$$Si(OCH_3)_2R_2^5 \quad (IIIa)$$

wherein R^5 represents a branched-alkyl group having 3 to 12 carbon atoms, preferably a branched-alkyl group having 3 to 6 carbon atoms, or a cyclo-alkyl having 4 to 12 carbon atoms, preferably a cyclo-alkyl having 5 to 8 carbon atoms.

It is in particular preferred that R^5 is selected from the group consisting of iso-propyl, iso-butyl, iso-pentyl, tert.-butyl, tert.-amyl, neopentyl, cyclopentyl, cyclohexyl, methylcyclopentyl and cycloheptyl.

Formula (IIIb) is defined by

$Si(OCH_2CH_3)_3(NR^xR^y)$ (IIIb)

wherein R^x and R^y can be the same or different, representing a hydrocarbon group having 1 to 12 carbon atoms.

R^x and R^y are independently selected from the group consisting of linear aliphatic hydrocarbon group having 1 to 12 carbon atoms, branched aliphatic hydrocarbon group having 1 to 12 carbon atoms and cyclic aliphatic hydrocarbon group having 1 to 12 carbon atoms. It is in particular preferred that R^x and R^y are independently selected from the group consisting of methyl, ethyl, n-propyl, n-butyl, octyl, decanyl, iso-propyl, iso-butyl, iso-pentyl, tert.-butyl, tert.-amyl, neopentyl, cyclopentyl, cyclohexyl, methylcyclopentyl and cycloheptyl.

More preferably both R^x and R^y are the same, yet more preferably both R^x and R^y are an ethyl group.

More preferably the external donor is of formula (IIIa), like dicyclopentyl dimethoxy silane [$Si(OCH_3)_2$(cyclo-pentyl)$_2$] or diisopropyl dimethoxy silane [$Si(OCH_3)_2(CH(CH_3)_2)_2$].

Most preferably the external donor of formula (IIIb) is diethylaminotriethoxysilane.

In a further embodiment, the Ziegler-Natta procatalyst can be modified by polymerising a vinyl compound in the presence of the catalyst system, comprising the special Ziegler-Natta procatalyst (component (i)), an external donor (component (iii) and optionally a cocatalyst (component (iii)), which vinyl compound has the formula:

CH_2=CH-CHR^3R^4

wherein R^3 and R^4 together form a 5- or 6-membered saturated, unsaturated or aromatic ring or independently represent an alkyl group comprising 1 to 4 carbon atoms, and the modified catalyst is used for the preparation of the heterophasic propylene copolymer according to this invention. The polymerized vinyl compound can act as an α-nucleating agent.

Concerning the modification of catalyst reference is made to the international applications WO 99/24478, WO 99/24479 and particularly WO 00/68315, incorporated herein by reference with respect to the reaction conditions concerning the modification of the catalyst as well as with respect to the polymerization reaction.

Accordingly it is appreciated that the first heterophasic propylene copolymer (HECO-1) is α-nucleated. In case the α-nucleation is not effected by a vinylcycloalkane polymer or a vinylalkane polymer as indicated above, the following α-nucleating agents may be present

(i) salts of monocarboxylic acids and polycarboxylic acids, e.g. sodium benzoate or aluminum tert-butylbenzoate, and

(ii) dibenzylidenesorbitol (e.g. 1,3 : 2,4 dibenzylidenesorbitol) and C_1-C_8-alkyl-substituted dibenzylidenesorbitol derivatives, such as methyldibenzylidenesorbitol, ethyldibenzylidenesorbitol or dimethyldibenzylidenesorbitol (e.g. 1,3 : 2,4 di(methylbenzylidene) sorbitol), or substituted nonitol-derivatives, such as 1,2,3,-trideoxy-4,6:5,7-bis-O-[(4-propylphenyl)methylene]-nonitol, and

(iii) salts of diesters of phosphoric acid, e.g. sodium 2,2'-methylenebis (4, 6,-di-tert-butylphenyl) phosphate or aluminium-hydroxy-bis[2,2'-methylene-bis(4,6-di-t-butylphenyl)phosphate], and

(iv) mixtures thereof.

Second heterophasic propylene copolymer (HECO-2)

As mentioned above, the polyolefin composition according to the present invention further comprises a second heterophasic propylene copolymer (HECO-2) as an essential component.

The second heterophasic propylene copolymer (HECO-2) typically has a higher melt flow rate MFR_2 than the first heterophasic propylene copolymer (HECO-1). Furthermore, the intrinsic viscosity (IV) of the xylene cold soluble (XCS) fraction of the second heterophasic propylene copolymer (HECO-2) is preferably lower than in the xylene cold soluble (XCS) fraction of the first heterophasic propylene copolymer (HECO-1).

Hence it is preferred that

(a) the melt flow rate MFR_2 (230°C) of the first heterophasic propylene copolymer (HECO-1) is at least 5 g/10min, more preferably at least 8 g/10min yet more preferably at least 10 g/10min, lower than the melt flow rate MFR_2 (230°C) of the second heterophasic propylene copolymer (HECO-2);

and/or

(b) the intrinsic viscosity (IV) of the xylene cold soluble (XCS) fraction of the first heterophasic propylene copolymer (HECO-1) is at least 0.3 dl/g, more preferably at least 0.5 dl/g, still more preferably 0.5 to 1.8 dl/, like 0.8 to 1.5 dl/g, higher than the melt flow rate MFR_2 (230°C) of the second heterophasic propylene copolymer (HECO-2)

Further it is preferred that the comonomer content, like ethylene content, of the first heterophasic propylene copolymer (HECO-1) is at least 2.0 wt.-%, more preferably at least 5.0 wt.-%, yet more preferably in the range of 2.0 to 18.0 wt.-%, like 5.0 to 15.0 wt.-%, like 5.0 to 10.0wt%, lower than the comonomer content, like ethylene content, of the second heterophasic propylene copolymer (HECO-2).

It is preferred that the second heterophasic propylene copolymer (HECO-2) before mixed with the other components mentioned herein comprises as polymer components only the matrix polypropylene (PP-2) and dispersed therein the elastomeric propylene copolymer (E-2). In other words the second heterophasic propylene copolymer (HECO-2) may contain further additives but no other polymer in an amount exceeding 5 wt-%, more preferably exceeding 3 wt.-%, like exceeding 1 wt.-%, based on the total amount of the second heterophasic propylene copolymer (HECO-2), more preferably based on the polymers present in the second heterophasic propylene copolymer (HECO-2). One additional polymer which may be present in such low amounts is a polyethylene which is a reaction product obtained by the preparation of the second heterophasic propylene copolymer (HECO-2). Accordingly, it is in particular appreciated that a second heterophasic propylene copolymer (HECO-2) as defined in the instant invention contains only a polypropylene (PP-2), an elastomeric propylene copolymer (E-2) and optionally a polyethylene in amounts as mentioned in this paragraph.

Preferably the second heterophasic propylene copolymer (HECO-2) has a melt flow rate MFR_2 (230°C) measured according to ISO 1133 in the range of from 7.0 to 19 g/10min, more preferably in the range of 8.0 to 15.0 g/10min, still more preferably in the range of from 9.0 to 14.0 g/10min, and even more preferably in the range of from 10.0 to 12.0 g/10min.

Preferably the propylene content in the second heterophasic propylene copolymer (HECO-2) is preferably in the range 78.0 to 92.0 wt-%, more preferably in the range of 80.0 to 91.0 wt-%, yet more preferably in the range of 82.0 to 90.0 wt-% and even more preferably 85.0 to 89.0 wt-% based on the total weight of the second heterophasic propylene copolymer (HECO-2), more preferably based on the amount of the polymer components of the second heterophasic propylene copolymer (HECO-2), yet more preferably based on the amount of the polypropylene (PP-2) and the elastomeric propylene copolymer (E-2) together.

The remaining part constitutes the comonomers, like ethylene, as defined for the polypropylene (PP-2) being a propylene copolymer (R-PP-2) and the elastomeric propylene copolymer (E-2), respectively. Accordingly, the comonomer content, preferably ethylene content, is preferably in the range of 8.0 to 22.0 wt.-%, more preferably in the range of 9.0 to 20.0 wt-%, yet more preferably in the range of 10.0 to 18.0 wt-% and even more preferably in the range of 11.0 to 15.0 wt-% based on the total weight of the second heterophasic propylene copolymer (HECO-2), more preferably based on the amount of the polymer components of the second heterophasic propylene copolymer (HECO-2), yet more preferably based on the amount of the polypropylene (PP-2) and the elastomeric propylene copolymer (E-2) together.

As stated above the matrix of the second heterophasic propylene copolymer (HECO-2) is the polypropylene (PP-2).

The polypropylene (PP-2) according to this invention constituting the matrix of the second heterophasic copolymer (HECO-2) shall have a melt flow rate MFR_2 (230 °C) in the range of from 40.0 to 80.0 g/10min, more preferably in the range of 45.0 to 70.0 g/10min, yet more preferably in the range of 50.0 to 65.0 g/10 min, still even more preferably in the range of 50.0 to 60.0 g/10min.

The polypropylene (PP-2) can be a propylene copolymer (R-PP-2) or a propylene homopolymer (H-PP-2), the latter is preferred.

Accordingly, it is appreciated that the polypropylene (PP-2) has a comonomer content equal or below 7.0 wt-%, more preferably equal or below 4.0 wt-%, still more preferably equal or below 1.0 wt-%.

In case the polypropylene (PP-2) is a propylene copolymer (R-PP-2) it comprises monomers copolymerizable with propylene, for example comonomers such as ethylene and/or C_4 to C_{12} α-olefins, in particular ethylene and/or C_4 to C_{10} α-olefins, e.g. 1-butene and/or 1-hexene. Preferably the propylene copolymer (R-PP-2) comprises, especially consists of, monomers copolymerizable with propylene from the group consisting of ethylene, 1-butene and 1-hexene. More specifically the propylene copolymer (R-PP-2) comprises - apart from propylene - units derivable from ethylene and/or 1-butene. In a preferred embodiment the propylene copolymer (R-PP-2) comprises units derivable from ethylene and propylene only. The comonomer content in the propylene copolymer (R-PP-2) is preferably in the range of more than 1.0 to 7.0 wt-%, still more preferably in the range of more than 1.0 to 4.0 wt-%.

The polypropylene (PP-2) can have a xylene cold soluble content (XCS) in a broad range, i.e. up to 5.0 wt.-%. Accordingly the polypropylene (PP-2) may have a xylene cold soluble content (XCS) in the range of 0.3 to 5.0 wt.-%, preferably in the range of 0.5 to 4.5 wt.-%, like in the range of 1.0 to 4.0 wt.-%.

However, in preferred embodiments, the polypropylene (PP-2), in particular in case the polypropylene (PP-2) is a propylene homopolymer (H-PP-2), has a xylene cold soluble (XCS) content in the range of 0.5 to 4.0 wt.-%, more preferably in the range of 1.0 to 3.5 wt.-%, still more preferably of 1.0 to 3.0 wt.-%.

One further essential component of the second heterophasic propylene copolymer (HECO-2) is its elastomeric propylene copolymer (E-2).

The elastomeric propylene copolymer (E-2) preferably comprises monomers copolymerizable with propylene, for example comonomers such as ethylene and/or C_4 to C_{12} α-olefins, in particular ethylene and/or C_4 to C_{10} α-olefins, e.g. 1-butene and/or 1-hexene. Preferably the elastomeric propylene copolymer (E-2) comprises, especially consists of, monomers copolymerizable with propylene from the group consisting of ethylene, 1-butene and 1-hexene. More specifically the elastomeric propylene copolymer (E-2) comprises – apart from propylene – units derivable from ethylene and/or 1-butene. Thus in an especially preferred embodiment the elastomeric propylene copolymer phase (E-2) comprises units derivable from ethylene and propylene only.

In case the polypropylene (PP-2) is a propylene copolymer (R-PP-2) it is preferred that the comonomer(s) of the propylene copolymer (R-PP-2) and the elastomeric propylene copolymer (E-2) are the same.

The properties of the elastomeric propylene copolymer phase (E-2) are mainly influenced by the xylene cold soluble (XCS) content of the second heterophasic propylene copolymer (HECO-2). Thus, according to the present invention the xylene cold soluble (XCS) fraction of the second heterophasic propylene copolymer (HECO-2) is regarded as the elastomeric propylene copolymer (E-2) of the second heterophasic propylene copolymer (HECO-2).

Accordingly, the amount of the elastomeric propylene copolymer (E-2), i.e. of the xylene cold soluble (XCS) fraction, of the second heterophasic propylene copolymer (HECO-2) preferably is in the range of from 20.0 to 45.0 wt-%, more preferably in the range of from 25.0 to 40.0 wt-% and even more preferably in the range of from 28.0 to 35.0 wt-%.These values are based on the second heterophasic propylene copolymer (HECO-2) and not on the total polypropylene composition (PP).

One important requirement of the present invention is that the elastomeric propylene copolymer (E-2) has a balanced weight average molecular weight. Accordingly the intrinsic viscosity must be carefully chosen.

It is appreciated that the elastomeric propylene copolymer phase (E-2), i.e. the xylene cold soluble fraction (XCS) of the second heterophasic propylene copolymer (HECO-2), has an intrinsic viscosity (IV) determined according to DIN ISO 1628/1 (in decaline at 135 °C) in the range of from 2.0 to 4.0 dl/g, more preferably in the range of from equal or more than 2.0 to 3.5 dl/g, and still even more preferably in the range of equal or more than from 2.0 to 3.0 dl/g, like in the range of 2.1 to 2.8 dl/g.

The comonomer content, preferably the ethylene content, within the elastomeric propylene copolymer phase (E-2) shall be preferably also in a specific range. Accordingly in a preferred embodiment the comonomer content, more preferably ethylene content, of the elastomeric propylene copolymer (E-2), i.e. of the xylene cold soluble fraction (XCS) of the second heterophasic propylene copolymer (HECO-2), is in the range of from 28.0 to 50.0 wt-%, still more preferably in the range of from 32.0 to 45.0 wt-%, yet more preferably in the range of from 35.0 to 42.0 wt-%.

Accordingly, it is appreciated that the propylene content of the elastomeric propylene copolymer (E-2), i.e. of the xylene cold soluble fraction (XCS) of the second heterophasic propylene copolymer (HECO-2), is preferably in the range of from 50.0 to 72.0 wt -%, still more preferably in the range of 55.0 to 68.0 wt-%, yet more preferably in the range of from 58.0 to 65.0 wt-%.

As will be explained below, the second heterophasic polypropylene (HECO-2) as well its individual components (matrix and elastomeric copolymer) can be produced by blending different polymer types, i.e. of different molecular weight and/or comonomer content. However it is preferred that the second heterophasic polypropylene (HECO-2) as well its individual components (matrix and elastomeric copolymer) are produced in a sequential step process, using reactors in serial configuration and operating at different reaction conditions. As a consequence, each fraction prepared in a specific reactor will have its own molecular weight distribution and/or comonomer content distribution.

Preferably the second heterophasic propylene copolymer (HECO-2) comprises an α-nucleating agent. Even more preferred the present invention is free of β-nucleating agents. Accordingly, the α-nucleating agent is preferably selected from the group consisting of

(i) salts of monocarboxylic acids and polycarboxylic acids, e.g. sodium benzoate or aluminum tert-butylbenzoate, and

(ii) dibenzylidenesorbitol (e.g. 1,3 : 2,4 dibenzylidenesorbitol) and C_1-C_8-alkyl-substituted dibenzylidenesorbitol derivatives, such as methyldibenzylidenesorbitol, ethyldibenzylidenesorbitol or dimethyldibenzylidenesorbitol (e.g. 1,3 : 2,4 di(methylbenzylidene) sorbitol), or substituted nonitol-derivatives, such as 1,2,3,-trideoxy-4,6:5,7-bis-O-[(4-propylphenyl)methylene]-nonitol, and

(iii) salts of diesters of phosphoric acid, e.g. sodium 2,2'-methylenebis (4, 6,-di-tert-butylphenyl) phosphate or aluminium-hydroxy-bis[2,2'-methylene-bis(4,6-di-t-butylphenyl)phosphate], and

(iv) vinylcycloalkane polymer and vinylalkane polymer (as discussed in more detail below), and

(v) mixtures thereof.

Such additives are generally commercially available and are described, for example, in "Plastic Additives Handbook", 5th edition, 2001 of Hans Zweifel, pages 871 to 873.

Preferably the second heterophasic propylene copolymer (HECO-2) contains up to 5 wt.-% of the α-nucleating agent. In a preferred embodiment, the second heterophasic propylene copolymer (HECO-2) contains not more than 200 ppm, more preferably of 1 to 200 ppm, more preferably of 5 to 100 ppm of a α-nucleating agent, in particular selected from the group consisting of dibenzylidenesorbitol (e.g. 1,3 : 2,4 dibenzylidene sorbitol), dibenzylidenesorbitol derivative, preferably dimethyldibenzylidenesorbitol (e.g. 1,3 : 2,4 di(methylbenzylidene) sorbitol), or substituted nonitol-derivatives, such as 1,2,3,-trideoxy-4,6:5,7-bis-O-[(4-propylphenyl)methylene]-nonitol, vinylcycloalkane polymer, vinylalkane polymer, and mixtures thereof.

It is especially preferred the second heterophasic propylene copolymer (HECO-2) contains a vinylcycloalkane, like vinylcyclohexane (VCH), polymer and/or vinylalkane polymer. In one specific embodiment the second heterophasic propylene copolymer (HECO-2) contains a vinylcycloalkane, like vinylcyclohexane (VCH), polymer and/or vinylalkane polymer. Preferably the vinylcycloalkane is vinylcyclohexane (VCH) polymer introduced into the second heterophasic propylene copolymer (HECO-2) by the BNT technology.

The second heterophasic propylene copolymer (HECO-2) according to this invention is commercially available. Accordingly a skilled person in the art is in a position to produce a second heterophasic propylene copolymer (HECO-2) as defined herein.

The second heterophasic propylene copolymer (HECO-2) is preferably obtained by a specific process. Accordingly the second heterophasic propylene copolymer (HECO-2) is preferably obtained by a sequential polymerization process in the first reactor (1st R') and optionally in a second reactor (2nd R') the propylene homopolymer (H-PP2) is produced, whereas in the third reactor (3rd R') and optionally in a fourth reactor (4th R') the elastomeric propylene copolymer (E2) of the second heterophasic propylene copolymer (HECO-2) is obtained.

The term "sequential polymerization process" indicates that the second heterophasic propylene copolymer (HECO-2) is produced in at least two reactors, preferably in three or four reactors, connected in series. Accordingly the present process comprises at least a first reactor (1st R'), an optional second reactor (2nd R'), a third reactor (3rd R') and optional a fourth reactor (4th R') The term "polymerization reactor" shall indicate that the main polymerization takes place. Thus in case the process consists of three or four polymerization

reactors, this definition does not exclude the option that the overall process comprises for instance a pre-polymerization step in a pre-polymerization reactor. The term "consist of" is only a closing formulation in view of the main polymerization reactors.

As stated above in the first (1st R') or in the first two reactors (1st and 2nd R') the matrix (M2), i.e. the propylene homopolymer (H-PP2) is produced. In case two reactors are used for the preparation of the propylene homopolymer (H-PP2), in each reactor a propylene homopolymer fraction (H-PP2a) and (H-PP2b) is produced which may differ or be the same in the melt flow rate as indicated above. Preferably the first propylene homopolymer fraction (H-PP2a) is produced in the first reactor (1st R') whereas the second propylene homopolymer fraction (H-PP2b) is produced in the second reactor (2nd R').

Preferably the weight ratio between the first propylene homopolymer fraction (H-PP2a) and second propylene homopolymer fraction (H-PP2b) is 20/80 to 80/20, more preferably 30/70 to 70/30, yet more preferably 40/60 to 65/35.

After the first reactor (1st R') or optional second reactor (2nd R') the matrix (M2), i.e. the propylene homopolymer (H-PP2), of the second heterophasic propylene copolymer (HECO-2), is obtained. This matrix (M2) is subsequently transferred into the third reactor (3rd R') and optional fourth reactor (4th R') in which the elastomeric propylene copolymer (E2) is produced and thus the second heterophasic propylene copolymer (HECO-2) of the instant invention is obtained.

The first reactor (1st R') is preferably a slurry reactor (SR) and can be any continuous or simple stirred batch tank reactor or loop reactor operating in bulk or slurry. Bulk means a polymerization in a reaction medium that comprises of at least 60 % (w/w) monomer. According to the present invention the slurry reactor (SR) is preferably a (bulk) loop reactor (LR).

The second reactor (2nd R'), the third reactor (3rd R') and fourth reactor (4th R') are preferably gas phase reactors (GPR). Such gas phase reactors (GPR) can be any mechanically mixed or fluid bed reactors. Preferably the gas phase reactors (GPR) comprise a mechanically agitated fluid bed reactor with gas velocities of at least 0.2 m/sec. Thus it is appreciated that the gas phase reactor is a fluidized bed type reactor preferably with a mechanical stirrer.

Thus in a preferred embodiment the first reactor (1st R') is a slurry reactor (SR), like loop reactor (LR), whereas the second reactor (2nd R'), the third reactor (3rd R') and the optional fourth reactor (4th R') are gas

phase reactors (GPR). Accordingly for the instant process at least two, preferably at least two or three polymerization reactors, namely a slurry reactor (SR), like loop reactor (LR), a first gas phase reactor (GPR-1), a second gas phase reactor (GPR-2) and optionally a third gas phase reactor (GPR-3) connected in series are used. If needed prior to the slurry reactor (SR) a pre-polymerization reactor is placed.

A preferred multistage process is a "loop-gas phase"-process, such as developed by Borealis A/S, Denmark (known as BORSTAR® technology) described e.g. in patent literature, such as in EP 0 887 379, WO 92/12182 WO 2004/000899, WO 2004/111095, WO 99/24478, WO 99/24479 or in WO 00/68315.

A further suitable slurry-gas phase process is the Spheripol® process of Basell.

Preferably, in the instant process for producing the second heterophasic propylene copolymer (HECO-2), as defined above the conditions for the first reactor (1st R'), i.e. the slurry reactor (SR), like a loop reactor (LR), may be as follows:

- the temperature is within the range of 40 °C to 110 °C, preferably between 60 °C and 100 °C, like 68 to 95 °C,
- the pressure is within the range of 20 bar to 80 bar, preferably between 40 bar to 70 bar,
- hydrogen can be added for controlling the molar mass in a manner known per se.

Subsequently, the reaction mixture from the first reactor (1st R') is transferred to the second reactor (2nd R'), i.e. gas phase reactor (GPR-1), whereby the conditions are preferably as follows:

- the temperature is within the range of 50 °C to 130 °C, preferably between 60 °C and 100 °C,
- the pressure is within the range of 5 bar to 50 bar, preferably between 15 bar to 35 bar,
- hydrogen can be added for controlling the molar mass in a manner known per se.

The condition in the third reactor (3rd R') and the fourth reactor (4th R'), preferably in the second gas phase reactor (GPR-2) and third gas phase reactor (GPR-3), are similar to the second reactor (2nd R').

The residence time can vary in the three or four reactor zones.

In one embodiment of the process for producing the second heterophasic propylene copolymer (HECO-2), the residence time the first reactor (1st R'), i.e. the slurry reactor (SR), like a loop reactor (LR), is in the range 0.2 to 4 hours, e.g. 0.3 to 1.5 hours and the residence time in the gas phase reactors will generally be 0.2 to 6.0 hours, like 0.5 to 4.0 hours.

If desired, the polymerization may be effected in a known manner under supercritical conditions in the first reactor (1st R'), i.e. in the slurry reactor (SR), like in the loop reactor (LR), and/or as a condensed mode in the gas phase reactors (GPR).

Preferably the process comprises also a prepolymerization with the catalyst system, as mentioned below, comprising a Ziegler-Natta procatalyst, an external donor and optionally a cocatalyst.

In a preferred embodiment, the prepolymerization is conducted as bulk slurry polymerization in liquid propylene, i.e. the liquid phase mainly comprises propylene, with minor amount of other reactants and optionally inert components dissolved therein.

The prepolymerization reaction is typically conducted at a temperature of 0 to 50 °C, preferably from 10 to 45 °C, and more preferably from 15 to 40 °C.

The pressure in the prepolymerization reactor is not critical but must be sufficiently high to maintain the reaction mixture in liquid phase. Thus, the pressure may be from 20 to 100 bar, for example 30 to 70 bar.

The catalyst components are preferably all introduced to the prepolymerization step. However, where the solid catalyst component (i) and the cocatalyst (ii) can be fed separately it is possible that only a part of the cocatalyst is introduced into the prepolymerization stage and the remaining part into subsequent polymerization stages. Also in such cases it is necessary to introduce so much cocatalyst into the prepolymerization stage that a sufficient polymerization reaction is obtained therein.

It is possible to add other components also to the prepolymerization stage. Thus, hydrogen may be added into the prepolymerization stage to control the molecular weight of the prepolymer as is known in the art. Further, antistatic additive may be used to prevent the particles from adhering to each other or to the walls of the reactor.

The precise control of the prepolymerization conditions and reaction parameters is within the skill of the art.

According to the invention the second heterophasic propylene copolymer (HECO-2) is obtained by a sequential polymerization process, as described above, in the presence of a catalyst system comprising a Ziegler-Natta catalyst and optionally an external donor, preferably a catalyst system comprising three

components, namely as component (i) a Ziegler-Natta procatalyst, and optionally as component (ii) an organometallic cocatalyst and as component (iii) an external donor represented by formula (IIIa) or (IIIb), preferably represented by formula (IIIa), as described above in accordance with the preparation of the first heterophasic propylene copolymer (HECO-1).

More preferably the external donor is of formula (IIIa), like dicyclopentyl dimethoxy silane [$Si(OCH_3)_2$(cyclo-pentyl)$_2$] or diisopropyl dimethoxy silane [$Si(OCH_3)_2(CH(CH_3)_2)_2$].

Elastomeric ethylene copolymer (EEC)

A further essential component of the present polypropylene composition (PP) is an elastomeric ethylene copolymer (EEC).

The elastomeric ethylene copolymer (EEC) is added to the polypropylene composition (PP) according to the present invention for improving toughness and reducing shrinkage.

The elastomeric ethylene copolymer (EEC) is an ethylene copolymer comprising ethylene monomer units and comonomer units selected from C_3 to C_{20} α-olefins, preferably propene, 1-butene, 1-hexene and 1-octene, or C_5 to C_{20} α,ω-alkadienes, preferably 1,7-octadiene. In a more preferred embodiment, the comonomer is selected from 1-butene, 1-hexene, and 1- octene, wherein 1-octene is most preferred as the comonomer.

The elastomeric ethylene copolymer (EEC) may contain between 55.0 to 85.0 wt.-% ethylene, preferably 60.0 to 80.0 wt.-% ethylene, and more preferably 65.0 to 75.0 wt.-% ethylene, based on the total amount of the elastomeric polyolefin copolymer. The remaining part to 100 wt.-% constitute the comonomer units.

In a preferred embodiment, the elastomeric ethylene copolymer (EEC) has a melt flow rate MFR_2 (190 °C) measured according to ISO 1133 in the range of from 0.5 to 50.0 g/10min. More preferably, the elastomeric ethylene copolymer (POC) has a melt flow rate MFR_2 (190 °C) in the range of from 20.0 to 40.0 g/10min, more preferably in the range of from 25.0 to 35.0 g/10min, and most preferably in the range of from 28.0 to 32.0 g/10min.

Furthermore, the elastomeric ethylene copolymer (EEC) has preferably a density of below 920 kg/m^3, more preferably of below 900 kg/m^3, still more preferably in the range of from 800 to 920 kg/m^3, yet more preferably in the range of from 850 to 900 kg/m^3 and most preferably in the range of from 860 to 890 kg/m^3.

The elastomeric ethylene copolymer (EEC) is known in the art and belongs in a preferred embodiment to the Exact or Engage series, respectively.

One important aspect of the present invention is that the amount of elastomeric ethylene copolymer (EEC) in the polypropylene composition (PP) is rather low. Accordingly, it is preferred that the elastomeric ethylene copolymer is present in the polypropylene composition (PP) according to the present invention in an amount of between 1.0 and 10.0 wt.-%, preferably in an amount between 3.0 and 10.0 wt.-%, more preferably in an amount between 5.0 and 9.0 wt.-% based on the total weight of the polypropylene composition (PP).

Inorganic filler (F)

As another essential component, the polypropylene composition (PP) according to the present invention comprises inorganic filler (F).

The inorganic filler (F) is present in the polypropylene composition (PP) in amounts of up to 50 wt.-%. It is preferred that the amount of inorganic filler (F) is in the range of from 25.0 to 48.0 wt.-%, more preferably in the range of from 30.0 to 45.0 wt.-%, and even more preferably in the range of from 35.0 to 40.0 wt.-% based on the total weight of the polypropylene composition (PP).

Preferably the inorganic filler (F) is mica, wollastonite, kaolinite, smectite, calcium carbonate, montmorillonite, talc, phyllosilicate or a mixture thereof. The most preferred inorganic filler (F) is talc.

The inorganic filler (F) preferably has a median particle size d_{50} calculated from the particle size distribution in mass percent and measured by laser diffraction in the range of 0.2 to 20.0 μm, more preferably in the range of 0.3 to 15.0 μm, still more preferably in the range of 0.4 to 10.0 μm. The most preferred median particle size d_{50} is in the range of 0.60 to 7.0 μm, including the most appropriate median particle size d_{50} of 2.40 μm.

The mineral filler (F) preferably has a cutoff particle size d_{95} calculated from the particle size distribution in mass percent and measured by laser diffraction of equal or below 20 μm, more preferably of equal or below 15 μm, and even more preferably of equal or below 10 μm.

Additionally or alternatively, the inorganic filler (F) has a specific surface area BET in the range from 1.0 to 50.0 m^2/g, more preferably in the range from 5.0 to 40.0 m^2/g, still more preferably in the range from 10.0 to 30.0 m^2/g and even more preferably in the range of 10.0 to 20.0 m^2/g.

It is preferred that the inorganic filler (F) is present in a specific weight ratio relative to the combined first and second heterophasic propylene copolymer (HECO-1) and (HECO-2) in the polypropylene composition (PP).

For example, the weight ratio of the combined heterophasic propylene copolymers (HECO-1) and (HECO-2) to the inorganic filler (F) [HECO1+HECO-2 / F] is from 2.8 to 0.3. Preferably, the weight ratio of the combined heterophasic propylene copolymers (HECO-1) and (HECO-2) to the inorganic filler (F) [HECO1+HECO-2 / F] is from 1.2 to 0.9, and even more preferably from 1.8 to 0.6.

Additionally or alternatively, the weight ratio of propylene homopolymer (homo-PP) to the inorganic filler (F) [homo-PP/F] is from 1.2 to 0.2. Preferably, the weight ratio of polypropylene homopolymer (homo-PP) to the inorganic filler (F) [homo-PP/F] is from 0.9 to 0.3, and more preferably from 0.7 to 0.4.

Typically the inorganic filler (F) has a surface area measured according to the commonly known BET method with N_2 gas as analysis adsorptive of less than 22 m^2/g, more preferably of less than 20 m^2/g, yet more preferably of less than 18 m^2/g. Inorganic fillers (F) fulfilling these requirements are preferably anisotropic mineral fillers (F), like talc, mica and wollastonite.

It is appreciated that the polypropylene composition (PP) according to the present invention may further (optionally) comprise at least one typical additive selected from the group consisting of acid scavengers, antioxidants, colorants, pigments, light stabilizers, UV-stabilizers, slip agents, anti-scratch agents, dispersing agents, carriers and colorants. Preferably the amount of these additives (excluding the inorganic filler (F) and alpha-nucleating agents) shall not exceed 10.0 wt.-%, preferably not more than 7.0 wt.-%, and most preferably not more than 5.0 wt.-% based on the total weight of the polypropylene composition (PP), within the instant polypropylene composition (PP).

The instant polypropylene composition (PP) contains preferably an α-nucleating agent. Even more preferred the present invention is free of β-nucleating agents. According to the present invention the nucleating agent is understood as a nucleating agent different to the inorganic filler (F). Accordingly, the nucleating agent is preferably selected from the group consisting of

(i) salts of monocarboxylic acids and polycarboxylic acids, e.g. sodium benzoate or aluminum tert-butylbenzoate, and

(ii) dibenzylidenesorbitol (e.g. 1,3 : 2,4 dibenzylidenesorbitol) and C_1-C_8-alkyl-substituted dibenzylidenesorbitol derivatives, such as methyldibenzylidenesorbitol, ethyldibenzylidenesorbitol or dimethyldibenzylidenesorbitol (e.g. 1,3 : 2,4 di(methylbenzylidene) sorbitol), or substituted nonitol-derivatives, such as 1,2,3,-trideoxy-4,6:5,7-bis-O-[(4-propylphenyl)methylene]-nonitol, and

(iii) salts of diesters of phosphoric acid, e.g. sodium 2,2'-methylenebis (4, 6,-di-tert-butylphenyl) phosphate or aluminium-hydroxy-bis[2,2'-methylene-bis(4,6-di-t-butylphenyl)phosphate], and

(iv) vinylcycloalkane polymer and vinylalkane polymer (as discussed above), and

(v) mixtures thereof.

Such additives are generally commercially available and are described, for example, in "Plastic Additives Handbook", 5th edition, 2001 of Hans Zweifel.

Most preferably the α-nucleating agent is part of the first and/or second heterophasic propylene copolymer (HECO-1) and (HECO-2) thus of the polypropylene composition (PP). Accordingly the α-nucleating agent content of the two essential heterophasic propylene copolymers (HECO-1) and (HECO-2) and thus of the polypropylene composition (PP) is preferably up to 5.0 wt.-%. In a preferred embodiment, the two essential heterophasic propylene copolymers (HECO-1) and (HECO-2) and thus the polypropylene composition (PP) contain(s) not more than 3,000 ppm, more preferably of 1 to 2,000 ppm of a α-nucleating agent, in particular selected from the group consisting of dibenzylidenesorbitol (e.g. 1,3 : 2,4 dibenzylidene sorbitol), dibenzylidenesorbitol derivative, preferably dimethyldibenzylidenesorbitol (e.g. 1,3 : 2,4 di(methylbenzylidene) sorbitol), or substituted nonitol-derivatives, such as 1,2,3,-trideoxy-4,6:5,7-bis-O-[(4-propylphenyl)methylene]-nonitol, vinylcycloalkane polymer, vinylalkane polymer, and mixtures thereof.

In a preferred embodiment the two essential heterophasic propylene copolymers (HECO-1) and (HECO-2) and thus the polypropylene composition (PP) contains a vinylcycloalkane, like vinylcyclohexane (VCH), polymer and/or vinylalkane polymer, as the α-nucleating agent. Preferably in this embodiment, the two essential heterophasic propylene copolymers (HECO-1) and (HECO-2) contain a vinylcycloalkane, like vinylcyclohexane (VCH), polymer and/or vinylalkane polymer, preferably vinylcyclohexane (VCH). Preferably the vinylcycloalkane is vinylcyclohexane (VCH) polymer which is optionally introduced into at least one of the two essential heterophasic propylene copolymers (HECO-1) and (HECO-2) and thus into the polypropylene composition (PP) by the BNT technology. More preferably in this preferred embodiment, the amount of vinylcycloalkane, like vinylcyclohexane (VCH), polymer and/or vinylalkane polymer, more

preferably of vinylcyclohexane (VCH) polymer, in the two essential heterophasic propylene copolymers (HECO-1) and (HECO-2) is not more than 500 ppm, more preferably of 0.5 to 200 ppm, most preferably 1 to 100 ppm,. Accordingly it is thus preferred that the polypropylene composition (PP) contains not more than 500 ppm, more preferably of 0.1 to 200 ppm, most preferably 0.2 to 100 ppm, of vinylcyclohexane (VCH) polymer.

With regard to the BNT-technology, reference is made to the international applications WO 99/24478, WO 99/24479 and particularly WO 00/68315. According to this technology a catalyst system, preferably a Ziegler-Natta procatalyst, can be modified by polymerising a vinyl compound in the presence of the catalyst system, comprising in particular the special Ziegler-Natta procatalyst, an external donor and a cocatalyst, which vinyl compound has the formula:

$$CH_2=CH-CHR^3R^4$$

wherein R^3 and R^4 together form a 5- or 6-membered saturated, unsaturated or aromatic ring or independently represent an alkyl group comprising 1 to 4 carbon atoms, and the modified catalyst is used for the preparation of the heterophasic polypropylene according to this invention, i.e. of the heterophasic propylene copolymers (HECO-1) and (HECO-2). The polymerized vinyl compound acts as an α-nucleating agent. The weight ratio of vinyl compound to solid catalyst component in the modification step of the catalyst is preferably of up to 5 (5:1), preferably up to 3 (3:1) most preferably from 0.5 (1:2) to 2 (2:1). The most preferred vinyl compound is vinylcyclohexane (VCH).

The polypropylene composition (PP) of the present invention is preferably used for the production of articles in the field of household appliances, medical appliances, automotive articles, particularly moulded automotive articles or automotive injection moulded articles, pipes and toys. Even more preferred is the use for the production of household appliances, like base or housing of air conditioner, refrigerator, etc.

The present invention will now be described in further detail by the examples provided below.

EXAMPLES

1. Definitions/Measuring Methods

The following definitions of terms and determination methods apply for the above general description of the invention as well as to the below examples unless otherwise defined.

Quantification of microstructure by NMR spectroscopy

Quantitative nuclear-magnetic resonance (NMR) spectroscopy was used to quantify the isotacticity and regio-regularity of the polypropylene homopolymers.

Quantitative $^{13}C\{^1H\}$ NMR spectra were recorded in the solution-state using a Bruker Advance 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 optimised 10 mm extended temperature probehead at 125°C using nitrogen gas for all pneumatics. For polypropylene homopolymers approximately 200 mg of material was dissolved in *1,2*-tetrachloroethane-d_2 (TCE-d_2). To ensure a homogenous solution, after initial sample preparation in a heat block, the NMR tube was further heated in a rotatary 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 needed for tacticity distribution quantification (Busico, V., Cipullo, R., Prog. Polym. Sci. 26 (2001) 443; Busico, V.; Cipullo, R., Monaco, G., Vacatello, M., Segre, A.L., Macromolecules 30 (1997) 6251). Standard single-pulse excitation was employed utilising the NOE and bi-level WALTZ16 decoupling scheme (Zhou, Z., Kuemmerle, R., Qiu, X., Redwine, D., Cong, R., Taha, A., Baugh, D. Winniford, B., J. Mag. Reson. 187 (2007) 225; Busico, V., Carbonniere, P., Cipullo, R., Pellecchia, R., Severn, J., Talarico, G., Macromol. Rapid Commun. 2007, 28, 11289). A total of 8192 (8k) transients were acquired per spectra.

Quantitative $^{13}C\{^1H\}$ NMR spectra were processed, integrated and relevant quantitative properties determined from the integrals using proprietary computer programs.

For polypropylene homopolymers all chemical shifts are internally referenced to the methyl isotactic pentad (mmmm) at 21.85 ppm.

Characteristic signals corresponding to regio defects (Resconi, L., Cavallo, L., Fait, A., Piemontesi, F., Chem. Rev. 2000, 100, 1253;; Wang, W-J., Zhu, S., Macromolecules 33 (2000), 1157; Cheng, H. N., Macromolecules 17 (1984), 1950) or comonomer were observed.

The tacticity distribution was quantified through integration of the methyl region between 23.6-19.7 ppm correcting for any sites not related to the stereo sequences of interest (Busico, V., Cipullo, R., Prog. Polym. Sci. 26 (2001) 443; Busico, V., Cipullo, R., Monaco, G., Vacatello, M., Segre, A.L., Macromolecules 30 (1997) 6251).

Specifically the influence of regio-defects and comonomer on the quantification of the tacticity distribution was corrected for by subtraction of representative regio-defect and comonomer integrals from the specific integral regions of the stereo sequences.

The isotacticity was determined at the pentad level and reported as the percentage of isotactic pentad (mmmm) sequences with respect to all pentad sequences:

[mmmm] % = 100 * (mmmm / sum of all pentads)

The presence of 2,1 erythro regio-defects was indicated by the presence of the two methyl sites at 17.7 and 17.2 ppm and confirmed by other characteristic sites. Characteristic signals corresponding to other types of

regio-defects were not observed (Resconi, L., Cavallo, L., Fait, A., Piemontesi, F., Chem. Rev. 2000, 100, 1253).

The amount of 2,1 erythro regio-defects was quantified using the average integral of the two characteristic methyl sites at 17.7 and 17.2 ppm:

$P_{21e} = (I_{e6} + I_{e8}) / 2$

The amount of 1,2 primary inserted propene was quantified based on the methyl region with correction undertaken for sites included in this region not related to primary insertion and for primary insertion sites excluded from this region:

$P_{12} = I_{CH3} + P_{12e}$

The total amount of propene was quantified as the sum of primary inserted propene and all other present regio-defects:

$P_{total} = P_{12} + P_{21e}$

The mole percent of 2,1 erythro regio-defects was quantified with respect to all propene:

[21e] mol.-% = 100 * (P_{21e} / P_{total})

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.

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 $^{13}C\{^{1}H\}$ spectra.

This method was chosen for its robust nature and ability to account for the presence of regio-defects when needed. 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.

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

Melting temperature (T_m): measured with a TA Instrument Q2000 differential scanning calorimetry (DSC) on 5 to 7 mg samples. DSC is run according to ISO 11357 / part 3 /method C2 in a heat / cool / heat cycle with a scan rate of 10 °C/min in the temperature range of -30 to +225°C. Melting temperature is determined from the second heating step.

Density is measured according to ISO 1183-1 - method A (2004). Sample preparation is done by compression moulding in accordance with ISO 1872-2:2007.

MFR_2 (230°C) is measured according to ISO 1133 (230°C, 2.16 kg load).

MFR_2 (190°C) is measured according to ISO 1133 (190°C, 2.16 kg load).

Xylene cold solubles (XCS, wt.-%): Content of xylene cold solubles (XCS) is determined at 25 °C according ISO 16152; first edition; 2005-07-01

Intrinsic viscosity is measured according to DIN ISO 1628/1, October 1999 (in Decalin at 135 °C).

Flexural Modulus and flexural strength were determined in 3-point-bending according to ISO 178 on injection molded specimens of 80 x 10 x 4 mm prepared in accordance with ISO 294-1:1996.

Tensile strength and Elongation at break are measured according to ISO 527-2 (cross head speed = 50 mm/min; 23 °C) using injection molded specimens as described in EN ISO 1873-2 (dog bone shape, 4 mm thickness).

Izod impact test:

The izod notched impact strength is measured according to ISO 180 / 1A at 23 °C by using injection moulded test specimens as described in EN ISO 1873-2 (80 x 10 x 4 mm).

Median particle size d_{50} and Cutoff particle size d_{95} (Laser diffraction) is calculated from the particle size distribution [mass percent] as determined by laser diffraction (Mastersizer) according to ISO 13320-1.

Specific surface area is determined as the BET surface according to DIN 66131/2.

Shrinkage:

The following test method was used for testing moulding shrinkage (EN ISO294.4: 2001E).

Preparation of the test sheet:

An oblong sheet for test is prepared by injection-moulding in an injection machine, model "Engel 120" of Engel Austria GmbH, Austria. A mold is attached to the injection machine, having an inner oblong cavity with a size of 150mm*90mm*3mm.The injection machine has four heating zones, wherein temperature of the four zones is respectively 195°C, 200°C, 190°C and 180°C. Temperature of the mold is 40°C.

Raw materials for producing the sheet are fed into extruder of the injection machine, melt in the four heating zones, and are injected at a speed of 10-15mm/s into the mold. A pressure of 25 bar is applied and held for 20 seconds to the melt flowing into the mold. A sheet is formed and cooled in the mold for 35 seconds, and to open the mold and to remove the oblong sheet from the inner cavity of the mold. Then, the obtained sheet is conditioned at 23±2°C and a humidity of 50% for 48 hours before the test.

The test follows the procedure of ISO294.4: 2001E

Moulding shrinkage is the difference in dimensions between a dry test sheet specimen and the mould cavity in which it was moulded, both the mould and the test specimen being at room temperature when measured. It is expressed as a percentage (%) of the mold cavity dimension concerned.

S_L: *is the moulding shrinkage parallel to the melt flow direction, which is determined at the mid-point of the* length of the test specimen.

$S_L = 100\% \times (L_0 - L) / L_0$

L_0 is the length, in millimetres, across the centre of the cavity;

L is the corresponding length, in millimetres, of the test specimen.

S_w: is the moulding shrinkage normal to the flow direction, which is determined at the mid-point of the width of the test specimen.

$S_w = 100\% \times (W_0 - W) / W_0$

W_0 is the width, in millimetres, across the centre of the cavity;

W is the corresponding width, in millimetres, of the test specimen.

2. Examples

The present invention is illustrated by the following examples. The hetrophasic propylene copolymers HECO1 and HECO 2-1, and HECO 2-2 were used for the inventive examples, which were prepared with one slurry loop reactor and two/three gas phase reactors by the known Borstar® technology, as disclosed in EP 0,887,379 A1.

The catalyst used in the polymerization processes for HECO-1, HECO-2-1 and HECO-2-2 has been produced by the publically known method as follows: First, 0.1 mol of $MgCl_2$ x 3 EtOH was suspended under inert conditions in 250 ml of decane in a reactor at atmospheric pressure. The solution was cooled to the temperature of −15°C and 300 ml of cold $TiCl_4$ was added while maintaining the temperature at said level. Then, the temperature of the slurry was increased slowly to 20 °C. At this temperature, 0.02 mol of dioctylphthalate (DOP) was added to the slurry. After the addition of the phthalate, the temperature was raised to 135 °C during 90 minutes and the slurry was allowed to stand for 60 minutes. Then, another 300 ml of $TiCl_4$ was added and the temperature was kept at 135 °C for 120 minutes. After this, the catalyst was filtered from the liquid and washed six times with 300 ml heptane at 80 °C. Then, the solid catalyst component was filtered and dried. Catalyst and its preparation concept is described e.g. in patent publications EP491566, EP591224 or EP586390. The catalyst was prepolymerized with vinyl cyclohexane in an amount to achieve a concentration of 200 ppm poly(vinyl cyclohexane) (PVCH) in the final polymer (see EP 1183307 A1). As co-catalyst triethyl-aluminium (TEAL) was used.

As donor for the preparation of HECO-1, HECO-2-1, and HECO-2-2, dicyclo pentyl dimethoxy silane (D-donor) was used. The aluminium to donor ratio is indicated in table 1.

Table 1a: Preparation and Properties of HECO 1 and HECO 2 (Loop/GPR1)

Loop	**Unit**	**HECO-1**	**HECO-2-1**	**HECO-2-2**
TEAL/Ti	[mol/mol]	125	220	262
TEAL/ D donor	[mol/mol]	5	8.1	13.3
Temperature	[°C]	85	72	71
Pressure	[kPa]	55	55	55
H_2/C_3 ratio	[mol/kmol]	0.08	14.5	16.4
MFR_2	[g/10 min]	0.05	55	85
XCS	[wt.-%]	1.5	1.5	1.5
Split	[wt.-%]	46	35	30
GPR 1				
Temperature	[°C]	90	80	77
Pressure	[kPa]	25	21	22
H_2/C_3 ratio	[mol/kmol]	214	155	123
MFR_2	[g/10 min]	0.30	55	85
XCS	[wt.-%]	1.5	1.5	1.5
Split	[wt.-%]	40	30	35

Table 1b: Preparation and Properties of HECO 1 and HECO 2 (GPR2/GPR3)

	Unit	**HECO1**	**HECO-2-1**	**HECO-2-2**
GPR 2				
Temperature	[°C]	80	70	78
Pressure	[kPa]	20	22	21
H_2/C_2 ratio	[mol/kmol]	20	108	166
C_2/C_3 ratio	[mol/kmol]	550	564	485
MFR_2	[g/10 min]	0.25	20	30
XCS	[wt.-%]	13	20	22
C2 of XCS	[wt.-%]	33	38	30
IV of XCS	[dl/g]	3.5	2.5	2.4
C2 total	[wt.-%]	4.6	8.5	7.5
Split	[wt.-%]	14	19	23
GPR 3				
Temperature	[°C]	-	80	80
Pressure	[kPa]	-	15	14
H_2/C_2 ratio	[mol/kmol]	-	87	284
C_2/C_3 ratio	[mol/kmol]	-	600	1254
MFR_2	[g/10 min]	-	11	18
MFR of XCI	[g/10min]	-	55	85
C2 total	[wt%]	-	13	15.5
XCS	[wt.-%]	-	32	33
C2 of XCS	[wt%]	-	38	40
IV of XCS	[dl/g]	-	2.5	2.1
Split	[wt.-%]	-	16	12

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Table 2a: Recipe of Inventive compositions of Inventive Examples 1-6

	Unit	IE1*)	IE2*)	IE3*)	IE4*)	IE5*)	IE6*)
homo-PP							
HJ311MO	w-%				20.0		
HJ325MO	w-%	10.0	20.0	30.0			
HH450FB	w-%					20.0	
HG385MO	w-%						20.0
HECO-1	w-%						
HECO-1	w-%	13.0	13.0	13.0	13.0	13.0	13.0
HECO-2	w-%						
HECO-2-1	w-%	30.0	20.0	10.0	20.0	20.0	20.0
HECO-2-2	w-%						
EEC	w-%	6.0	6.0	6.0	6.0	6.0	6.0
Filler	w-%	39.0	39.0	39.0	39.0	39.0	39.0

Table 2b: Recipe of Inventive compositions of IE7 to IE10

	Unit	IE7*)	IE8*)	IE9*)	IE10*)
homo-PP	w-%				
HJ311MO	w-%				
HJ325MO	w-%	20.0	24.0	20.0	22.0
HH450FB	w-%				
HG385MO	w-%				
HECO-1	w-%				
HECO-1	w-%	13.0	13.0	19.0	20.0
HECO-2	w-%				
HECO-2-1	w-%		20.0	20.0	13.0
HECO-2-2	w-%	20.0			
EEC	w-%	6.0	6.0	7.0	5.0
Filler	w-%	39.0	35.0	32.0	38.0

*) rest to 100 wt.-% were PP powder additive (1.1 wt.-% of PP-140-2), calcium carbonate (0.1 wt.-% OMYA 2), heat stabilizer (0.2 wt.-% AT-225/Irganox B225 FF and 0.2 wt% Irganox PS 802 FL), lubricant (0.3 wt.-% Acrawax C) and MgO (0.1 wt.-%).

"**Homo-PP**" are the following commercially available propylene homopolymers:

- "HJ311MO" of Borouge (Adu Dhabi Polymers Company Ltd., Abu Dhabi, UAE), having a MFR_2 of 60 g/10min (230°C/2.16 kg);
- "HJ325MO" of Borouge having a MFR_2 of 50 g/10min (230°C/2.16 kg) and a melting temperature of 164 °C;
- "HH450FB" of Borealis AG having a MFR_2 of 37 g/10min (230°C/2.16 kg);
- "HG385MO" of Borouge having a MFR_2 of 25 g/10min (230°C/2.16 kg);

"**EEC**" is the commercial ethylene/octylene copolymer "Engage 8407" of Dow Elastomes having MFR_2 of 30 g/10min (190°C/2.16 kg) and a density of 0.870 g/cm^3.

"**Filler**" is the commercial talc-based mineral filler "HTP 2" of IMI Fabi Corporation (Italy) having d_{50} of 2.40 μm, d_{95} of 8.4 μm and a surface area "BET" of 8 g/m^2.

The inventive compositions of IE1to IE10 based on the recipes as summarized in Table 2 are prepared by using a Coperion STS-35 twin-screw extruder (available from Coperion (Nanjing) Corporation, China) with a diameter of 35 mm. The twin-screw extruder runs at an average screw speed of 500 rpm with a temperature profile of zones from 180-230 °C. It has a L/D of 44. The temperature of each zone, throughput and the screw speed of the extruder for preparing the inventive compositions of IE1 to IE10 are listed in Table 3. The temperature of each zone (including zones 1 to 11, zone 1 is feed port of the extruder with Room Temperature), throughput and screw speed of the extruder are initiative parameters, and are set on control panel of the extruder. Melt temperature (temperature of the melt in the die) and torque of the extruder are passive parameters shown on control panel of the extruder. A vacuum bump is located in zone 9 and generates a vacuum of -0.06 MPa inside the extruder.

Table 3a: Extruder conditions of the inventive compositions of IE1 to IE6

Process condition	Unit	IE1	IE2	IE3	IE4	IE5	IE6
zone 1	[°C]	RT	RT	RT	RT	RT	RT
zone 2	[°C]	187	181	181	184	188	185
zone 3	[°C]	191	195	192	199	190	198
zone 4	[°C]	201	198	196	198	197	195
zone 5	[°C]	218	211	217	218	213	214
zone 6	[°C]	218	215	215	210	219	217
zone 7	[°C]	217	212	214	215	216	211
zone 8	[°C]	212	214	211	216	218	217
zone 9	[°C]	212	212	214	216	217	218
zone 10	[°C]	216	213	216	215	215	216
zone 11	[°C]	219	211	210	217	215	213
die	[°C]	220	223	225	224	224	220
melt temp.	[°C]	219	216	214	215	212	217
throughput	[kg/hour]	40	40	40	40	40	40
screw speed	[rpm]	500	500	500	500	500	500
torque	%	68.0	74.0	68.0	73.0	73.0	70.0
vacuum	[MPa]	-0.06	-0.06	-0.06	-0.06	-0.06	-0.06

Table 3b: Extruder conditions of the inventive compositions of IE7 to IE10

Process condition	Unit	IE7	IE8	IE9	IE10
zone 1	[°C]	RT	RT	RT	RT
zone2	[°C]	180	188	180	180
zone 3	[°C]	194	197	196	195
zone 4	[°C]	201	197	196	200
zone 5	[°C]	218	211	210	213

zone 6	[°C]	211	216	210	210
zone 7	[°C]	213	212	215	213
zone 8	[°C]	214	216	214	210
zone 9	[°C]	217	217	214	217
zone 10	[°C]	213	216	211	219
zone 11	[°C]	218	214	211	218
die	[°C]	225	227	221	227
melt temp.	[°C]	213	213	216	217
throughput	[kg/hour]	40	40	40	40
screw speed	[rpm]	500	500	500	500
torque	%	70.0	66.0	70.0	71.0
vacuum	[MPa]	-0.06	-0.06	-0.06	-0.06

Results

The mechanical properties of the inventive compositions of IE1 to IE10 are shown below in Table 4 and compared with the mechanical properties of commercial polymer material "TAIRIREX HP 8250" of Formosa Chemicals & Fibre Corporation, Taiwan.

The comparative polymer is a high impact polystyrene (HIPS) grafted with polybutyl rubber, prepared by polymerizing styrene and grafting with polybutyl rubber simultaneously.

Table 4a: Properties of inventive compositions of IE1 to IE5

Property	Unit	HIPS "HP 8250"	IE1	IE2	IE3	IE4	IE5
MFR_2 (230℃)	[g/10min]	6	4.7	6.3	8.1	7.1	5.6
Flexural Modulus	[MPa]	2200	2480	3300	3700	3180	3210
Tensile Strength	[MPa]	22.7	20.2	25.2	26.9	24.6	24.9
Elongation @ break	[%]	33	89	20	17	21	19
Izod Notched Impact (RT)	[kJ/m^2]	9.5	19.2	8.7	6.2	8.2	8.6
S_L	[%]	0.58	0.68	0.72	0.74	0.72	0.72
S_W	[%]	0.66	0.79	0.83	0.87	0.82	0.81

Table 4b: Properties of inventive compositions of IE6 to IE10

Property	Unit	IE6	IE7	IE8	IE9	IE10
MFR_2 (230℃)	[g/10min]	5.1	7.2	6.7	5.1	5.2
Flexural Modulus	[MPa]	3290	3270	2790	2660	3730
Tensile Strength	[MPa]	25.2	25.1	24.7	24.6	27
Elongation @ break	[%]	22	20	36	51	20
Izod Notched RT	[kJ/m^2]	8.3	8.2	9.2	12.8	7.2
S_L	[%]	0.73	0.72	0.74	0.76	0.74
S_W	[%]	0.81	0.82	0.85	0.86	0.84

S_L: shrinkage parallel to the melt flow direction

S_W: shrinkage normal to the flowing direction

Conclusions

When compared with the comparative HIPS, the inventive composition IE1 to IE10 shows comparative shrinkage and impact, better stiffness (modulus and strength), meanwhile having much lower cost in the material and preparation.

CLAIMS

1. Polypropylene composition (PP) comprising,
   (a) a propylene homopolymer (homo-PP) having a melt flow rate MFR2 (230°C, 2,16 kg) measured according to ISO 1133 of from 20.0 to 70.0 g/10 min,
   (b) a first heterophasic propylene copolymer (HECO-1) having a melt flow rate MFR_2 (230 °C, 2.16 kg) measured according to ISO 1133 in the range of from 0.1 to 2.5 g/10min,
   (c) a second heterophasic propylene copolymer (HECO-2) having a melt flow rate MFR_2 (230°C, 2.16 kg) measured according to ISO 1133 in the range of from 8.0 to 15.0 g/10min,
   (d) an elastomeric ethylene copolymer(EEC) having a melt flow rate MFR_2 (190°C, 2,16 kg) measured according to ISO 1133 in the range of from 0.5 to 50.0 g/10min, and
   (e) an inorganic filler (F).

2. Polypropylene composition (PP) according to claim 1, wherein the first heterophasic propylene copolymer (HECO-1) has
   (a) a xylene cold soluble (XCS) fraction measured according to ISO 16152 (25 °C) of from 8.0 to 20.0 wt.-%, based on the total weight of the heterophasic propylene copolymer (HECO-1), and/or
   (b) an comonomer content of lower than 7.0 wt.-%, based on the total weight of the heterophasic propylene copolymer (HECO-1), wherein the comonomer is ethylene and/or C_4 to C_{10} α-olefin.

3. Polypropylene composition (PP) according to claim 1 or 2, wherein the second heterophasic propylene copolymer (HECO-2) has
   (a) a xylene cold soluble (XCS) fraction measured according to ISO 16152 (25 °C) of from 15.0 to 45.0 wt.-%, based on the total weight of the heterophasic propylene copolymer (HECO-2), and/or
   (b) a comonomer content of 10.0 to 22.0 wt.-%, based on the total weight of the heterophasic propylene copolymer (HECO-2), wherein the comonomer is ethylene and/or C_4 to C_{10} α-olefin..

4. Polypropylene composition (PP) according to any one of the preceding claims, wherein the xylene cold soluble (XCS) fraction of the first heterophasic propylene copolymer (HECO-1) has
   (a) an intrinsic viscosity (IV) of from 2.5 to 4.5 dl/g,
   and/or
   (b) a comonomer content of 25.0 to 41.0 wt.-%, wherein the comonomer is ethylene and/or C_4 to C_{10} α-olefin.

5. Polypropylene composition (PP) according to any one of the preceding claims, wherein the xylene cold soluble (XCS) fraction of the second heterophasic propylene copolymer (HECO-2) has

(a) an intrinsic viscosity (IV) of from 2.0 to 4.0 dl/g,

and/or

(b) a comonomer content of 32.0 to 45.0 wt.-%, wherein the comonomer is ethylene and/or C_4 to C_{10} α-olefin.

6. Polypropylene composition (PP) according to any one of the preceding claims, wherein the polypropylene composition (PP) comprises,

(a) 10 to 30 wt% of the propylene homopolymer (homo-PP)

(b) 5 to 25 wt% of the first heterophasic propylene copolymer (HECO-1)

(c) 10 to 45 wt% of the second heterophasic propylene copolymer (HECO-2)

(d) 3 to 10 wt% of the elastomeric ethylene copolymer (EEC), and

(e) 25 to 45 wt% of the inorganic filler (F),

based on the total weight of the composition.

7. Polypropylene composition (PP) according to any one of the preceding claims, wherein the melt flow rate MFR_2 (230°C) of the first heterophasic propylene copolymer (HECO-1) is at least 5 g/10min lower than the melt flow rate MFR_2 (230°C) of the second heterophasic propylene copolymer (HECO-2).

8. Polypropylene composition(PP) according to any one of the preceding claims, wherein the intrinsic viscosity (IV) of the xylene cold soluble (XCS) fraction of the first heterophasic propylene copolymer (HECO-1) is at least 0.3 dl/g higher than the intrinsic viscosity of the xylene cold soluble fraction of the second heterophasic propylene copolymer (HECO-2).

9. Polypropylene composition (PP) according to any one of the preceding claims, wherein the comonomer content, preferably the ethylene content, of the first heterophasic propylene copolymer (HECO-1) is at least 3.0 wt-% lower than the comonomer content, preferably the ethylene content, of the second heterophasic propylene copolymer (HECO-2).

10. Polypropylene composition (PP) according to any one of the preceding claims, wherein

(a) the first heterophasic propylene copolymer (HECO-1) comprises a polypropylene matrix (PP-1), preferably a propylene homopolymer (H-PP-1), and an elastomeric propylene copolymer (E1), wherein the comonomer is ethylene and/or C_4 to C_{10} α-olefin;

and/or

(b) the second heterophasic propylene copolymer (HECO-2) comprises a polypropylene matrix (PP-2), preferably a propylene homopolymer (H-PP-2), and an elastomeric propylene copolymer (E2), wherein the comonomer is ethylene and/or C_4 to C_{10} α-olefin.

11. Polypropylene composition (PP) according to claim 10, wherein the propylene homopolymer (homo-PP) has a melt flow rate MFR_2 (230°C) which is at least 10 g/10min higher than the melt flow rate MFR_2 (230°C) of the polypropylene matrix (PP-1), preferably of the propylene homopolymer (H-PP-1), of first heterophasic propylene copolymer (HECO-1).

12. Polypropylene composition (PP) according to claim 10 or 11, wherein the propylene homopolymer (homo-PP) has a melt flow rate MFR_2 (230°C) which is +/- 5 g/10min to the melt flow rate MFR_2 (230°C) of the polypropylene matrix (PP-2), preferably of the propylene homopolymer (H-PP-2), of second heterophasic propylene copolymer (HECO-2).

13. Polypropylene composition (PP) according to any one of the preceding claims, wherein the weight ratio of the combined heterophasic propylene copolymers (HECO-1) and (HECO-2) to the inorganic filler (F) [HECO1+HECO-2 / F] is from 2.8 to 0.3.

14. Polypropylene composition (PP) according to any one of the preceding claims, wherein the weight ratio of propylene homopolymer (homo-PP) to the inorganic filler (F) [homo-PP/F] is from 1.2 to 0.2.

15. Article comprising a polypropylene composition (PP) according to any one of the preceding claims.

16. Use of the polypropylene composition (PP) according to any one of the preceding claims 1 to 14 for the production of household articles, medical articles, automotive articles, pipes and toys.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/CN2013/001462

A. CLASSIFICATION OF SUBJECT MATTER

C08L 23/10(2006.01)i; C08L 23/12(2006.01)i; C08L 23/14(2006.01)i; C08K 3/00(2006.01)i

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、C08K

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)

DWPI,SIPOABS,CNABS,CNKI: polypropylene, pp, heterophasic, HECO, +ethylene, filler

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	CN 103080212 A (BOREALIS AG) 01 May 2013 (2013-05-01) description, paragraphs [0060]-[0064], [0090]-[0093], [0097], [0100], [0117], [0122]-[0129], [0138], [0144]-[0149], [0168]-[0174], [0379]-[0382], [309] and [310]	1-16
A	CN 102449047 A (BOREALIS AG) 09 May 2012 (2012-05-09) the whole document	1-16
A	WO 2013149915 A1 (BOREALIS AG) 10 October 2013 (2013-10-10) the whole document	1-16
A	WO 2013064364 A1 (BASELL POLIOLEFINE ITAL SRL) 10 May 2013 (2013-05-10) the whole document	1-16
A	EP 2589623 A1 (BASELL POLIOLEFINE ITAL SRL) 08 May 2013 (2013-05-08) the whole document	1-16
A	EP 2650329 A1 (BOREALIS AG) 16 October 2013 (2013-10-16) the whole document	1-16
A	EP 2615136 A1 (BOREALIS AG) 17 July 2013 (2013-07-17) the whole document	1-16

☐ Further documents are listed in the continuation of Box C. ☑ See patent family annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
26 August 2014	**11 September 2014**
Name and mailing address of the ISA/ **STATE INTELLECTUAL PROPERTY OFFICE OF THE P.R.CHINA(ISA/CN)** **6,Xitucheng Rd., Jimen Bridge, Haidian District, Beijing 100088 China** Facsimile No. **(86-10)62019451**	Authorized officer **ZHAI,Xiaoxiao** Telephone No. **(86-10)62084451**

INTERNATIONAL SEARCH REPORT
Information on patent family members

International application No.
PCT/CN2013/001462

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