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Unoriented film based on soft polypropylene

The present invention is directed to a new unoriented film as well as to its manufacture.

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There is an increasing trend in the food packaging industry to use plastic containers, notably pouches containing sterilized or pre-cooked foods. Retort pouches offer many advantages over rigid metal packages such as faster cooking/sterilizing time, less shelf storage space, easier disposal, improved food taste, etc. Typical pouches have a multilayer structure with polyolefins such as polyethylene or polypropylene, adhesives, barrier and external layers. It is desired that the polyolefin material imparts stiffness as well as high impact strength to the final packaging material.

The same trend, i.e. increased use of polyolefinic materials, is seen in the medical packaging industry as well. Again, the polymer should impart sufficient stiffness as well as high impact strength to the final packaging material. In the case of medical applications, softness rather than stiffness is a key-requirement. Of course also these medical products must be sterilizable.

- It is known that impact strength of polypropylene can be improved by dispersing a rubber phase within the polymer matrix, thereby obtaining a heterophasic polypropylene composition. Such a material is also called "impact-modified polypropylene".
- As discussed above, it is known that heterophasic propylene polymers (impact-modified propylene polymers) provide high impact strength if the amount of rubber dispersed within the matrix is sufficiently high, e.g. in stand-up pouches typically at least 10 wt% or even at least 15 wt%.
- As mentioned above, for some food packaging applications such as retort pouches or some medical packaging applications, a sterilization treatment is needed. The most common sterilization procedures are the use of heat (steam), radiation (beta radiation, electrons, or gamma radiation) or chemicals (usually ethylene oxide). Steam sterilisation is usually carried out in a temperature range of about 120 to 130°C. Of course, treatment of a polymer under the sterilization conditions outlined above can impair its final properties.

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However, it has turned out that standard heterophasic systems change significantly their properties after sterilization. Typically the optical properties as well as the mechanical properties are undesirable impaired.

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Considering the drawbacks outlined above, it is an object of the present invention to provide a soft polymer composition with good impact behavior and optical properties which can be used as material for unoriented films.

The finding of the present invention is to provide an unoriented film of at least one layer, said layer is mainly based on a composition comprising a heterophasic propylene copolymer and a styrenic based elastomer having rather low styrene content. Further the heterophasic propylene copolymer used must have a rather high comonomer content and xylene cold soluble (XCS) content, wherein the xylene cold soluble fraction is featured by high propylene content.

Accordingly, the present invention is directed an unoriented film comprising at least one layer (L1), preferably comprising at least three layers (L1), (L2) and (L3) preferably with the stacking order (L2)/(L1)/(L3), wherein said layer (L1) comprises at least 70 wt.-% of a polymer composition (Co), said composition (Co) comprises

- (a) a propylene copolymer (A) having
 - (a1) a melt flow rate MFR₂ (230 °C) measured according to ISO 1133 in the range of more than 2.0 to 15.0 g/10min,
 - (a2) a comonomer content in the range of more than 7.5 to 16.5 wt.-%, and
 - (a3) a xylene cold soluble content (XCS) determined according ISO 16152 (25 °C) in the range of 20.0 to 55.0 wt.-%,

wherein

- (a4) the comonomer content of xylene cold soluble (XCS) fraction of the propylene copolymer is in the range of 14.0 to 30.0 wt.-%,
- 30 and

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(b) (a) styrenic based elastomer(s) (B) having a styrene content in the range of 5 to equal or below 15 wt.-%.

Preferably the styrenic based elastomer(s) (B) is (are) preferably a styrenethylene/butylene-styrene (SEBS) block copolymer (B-1) and/or a hydrogenated styrenevinyl isoprene (SIS) block rubber (B-2).

Accordingly the present invention is preferably directed to an unoriented film comprising at least one layer (L1), preferably comprising at least three layers (L1), (L2) and (L3)

- preferably with the stacking order (L2)/(L1)/(L3), wherein said layer (L1) comprises at least 70 wt.-% of a polymer composition (Co), said composition (Co) comprises
 - (a) a propylene copolymer (A) having
 - (a1) a melt flow rate MFR₂ (230 °C) measured according to ISO 1133 in the range of more than 2.0 to 15.0 g/10min,
 - (a2) a comonomer content in the range of more than 7.5 to 16.5 wt.-%, and
 - (a3) a xylene cold soluble content (XCS) determined according ISO 16152 (25 °C) in the range of 20.0 to 55.0 wt.-%, wherein
 - the comonomer content of xylene cold soluble (XCS) fraction of the propylene copolymer is in the range of 14.0 to 30.0 wt.-%,

and

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(b) (a) styrenic based elastomer(s) (B) being a styrene-ethylene/butylene-styrene (SEBS) block copolymer (B-1) and/or a hydrogenated styrene-vinyl isoprene (SIS) block rubber (B-2), wherein the styrene content in each styrenic based elastomer (B) is in the range of 5 to equal or below 15 wt.-%.

More preferably the polymer composition (Co) comprises as styrenic based elastomer(s) (B) only a styrene-ethylene/butylene-styrene (SEBS) block copolymer (B-1) and/or a hydrogenated styrene-vinyl isoprene (SIS) block rubber (B-2). Thus it is in particular appreciated that the polymer composition (Co) comprises as styrenic based elastomers (B)

either a hydrogenated styrene-vinyl isoprene (SIS) block rubber (B-2) or a styrene-ethylene/butylene-styrene (SEBS) block copolymer (B-1), the latter especially preferred.

Further it is appreciated that the propylene copolymer (A) and the styrenic based elastomer(s) (B) are the only polymer components in the polymer composition (Co) and/or in the layer (L1).

Accordingly it is in particular preferred that the polymer composition (Co) and/or the layer (L1) according to this invention comprise(s) as the only polymer components

- 10 (i) the propylene copolymer (A) and
 - (ii) the styrene-ethylene/butylene-styrene (SEBS) block copolymer (B-1) and/or the hydrogenated styrene-vinyl isoprene (SIS) block rubber (B-2), preferably the styrene-ethylene/butylene-styrene (SEBS) block copolymer (B-1).
- In further preferred embodiment of the instant invention the weight ratio of propylene copolymer (A) and the styrenic based elastomer(s) (B) [(A)/(B)] is in the range of 9/1 to 3/2.

Preferably the propylene copolymer (A) is a heterophasic propylene copolymer (RAHECO) as defined in more detail below.

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It has surprisingly been found that such unoriented films have a low tensile modulus, high penetration energy and good optical properties before and after sterilization.

In the following the present invention is described in more detail.

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The unoriented film / layer (L1)

One distinguishes between unoriented and oriented films (see for instance polypropylene handbook, Nello Pasquini, 2nd edition, Hanser). Oriented films are typically monoaxially or biaxially oriented films, whereas unoriented films are cast or blown films. Accordingly an unoriented film is not drawn intensively in machine and/or transverse direction as done by

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oriented films. Thus the unoriented film according to this invention is not a monoaxially or biaxially oriented film. Preferably the unoriented film according to the instant invention is a blown film or cast film.

5 Further the unoriented film according to this invention can have just one layer (L1) and is thus a mono-layer film, i.e. a mono-layer (L1) film, or comprises two or more layers, like three or more layers, and is thus a multi-layer film.

In case the unoriented film is a mono-layer film it is preferred that the layer (L1) has a thickness of 10 to 1,000 μ m, preferably of 40 to 700 μ m, more preferably of 50 to 500 μ m, like of 80 to 300 μ m. The process thereof is described in more detail below.

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In case the unoriented film is a multi-layer film, said film may comprise at least two layers, preferably may consist of two layers, or may comprise at least three layers, preferably may consist of three layers. In any case at least one layer, more preferably one layer, of the multi-layer film, i.e. of the at least two layer film or of the at least three layer film, is the layer (L1) as defined in the instant invention. Thus the unoriented film may be a two layer film wherein one layer is the layer (L1) whereas the other layer is preferably a sealing layer as known in the art. In another preferred embodiment the unoriented film comprises at least three layers (L1), (L2), and (L3), preferably consists of three layers (L1), (L2), and (L3), wherein preferably the stacking order is (L2)/(L1)/(L3), i.e. the layer (L1) forms the core layer in the multi-layer film. Accordingly in case the unoriented film is an at least three layer film, preferably is a three layer film, layer (L1) is preferably the core layer, whereas the layer (L2) is an outer layer and the layer (L3) is a sealing layer. In another preferred embodiment the unoriented film is an at least three layer film, preferably is a three layer film, wherein the layer (L1) constitutes the core layer, whereas the layer (L3) is a sealing layer and (L2) is a heat resistant outer layer.

The terms sealing layer and outer layer are understood as commonly known in the art.

30 Accordingly the total multi-layer film has a thickness of 10 to 1,000 µm, preferably of 40 to 700 µm, more preferably of 50 to 500 µm, even more preferably 80 to 300 µm, like of 100 to

 $250 \,\mu\text{m}$. Preferably the sealing layer(s) has/have a thickness that is substantially less than the thickness of the core layer and substantially less than the thickness of the total multi-layer film. In one embodiment the thickness of the sealing layer(s) is/are substantially less, usually 5 to 40 %, preferably 10 to 30 %, more preferably 15 to 25 % of the thickness of the total multi-layer film.

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The outer layer (OL) - if present - may have a thickness in the range of 3 to 30 %, preferably 5 to 20 %, even more preferably 5 to 15 % of the thickness of the total multi-layer film.

Accordingly in one embodiment the multi-layer film comprises at least three layers, namely at least one core layer being layer (L1), and two sealing layers, namely the layer (L2) and the layer (L3), wherein the multi-layer film has the stacking order (L2)-(L1)-(L3). Accordingly in one preferred embodiment the sealing layers (L2) and (L3) are directly co-extruded with layer (L1) which constitutes core layer of the multi-layer film. Thus in one specific preferred embodiment the multi-layer film consists of two sealing layer, namely layer (L2) and layer (L3), and layer (L1) being the core layer, wherein the stacking order is (L2)-(L1)-(L3). The sealing layers can be chemically different or identical.

In another preferred embodiment the multi-layer film comprises at least three layers, namely a core layer being layer (L1), a sealing layer being layer (L2) and a outer layer being layer (L3), wherein the sealing layer is located, i.e. joined, on the one side (surface) of the core layer and the outer layer is located, i.e. joined, on the other side (surface) of the core layer. Accordingly the multi-layer film has the stacking order (L2)-(L1)-(L3). Preferably the sealing layer and the outer layer are co-extruded with the core layer on a blown film line or cast film line.

As mentioned above the mono-layer film as well as the multi-layer film is preferably obtained by extrusion and coextrusion, respectively. The cooling of the layer(s) when the blown film line is used can be effected by air cooling or water cooling, the latter being preferred. In case of use of a cast film line, cooling is accomplished by chill rolls. An

especially preferred process for the preparation of a mono-layer film and multi-layer film according to this invention is described in more detail below.

The material applied for the outer layer and sealing layer, respectively, is known in the art.

For instance for the outer layer a material selected from the group consisting of polyvinyl alcohol, polyacrylate, polyamide, polyester, like poly(ethylene terephthalate), polyolefin (PO) and mixtures thereof is used. In one embodiment the outer layer is a polyamide, a polyester, a polyvinyl alcohol, a polyethylene or a polypropylene, like a propylene homopolymer or a propylene copolymer. Tie layers can be used between outer and core layer in order to improve adhesion.

As material for sealing layers preferably random co- and terpolymers of propylene with ethylene and/or C4-C10 alpha-olefins having a Tm < 150 °C can be used. Ethylene copolymers with C4-C10 alpha-olefins having a Tm > 80 °C can also be used. The sealing layer can also be a blend of 2 or more olefin copolymers and further modifiers like elastomers or hydrocarbon resins, alternatively also a cold-seal adhesive layer can be used.

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A further requirement of the present invention is that layer (L1) of the instant unoriented film must comprise the composition (Co). Accordingly it is preferred that layer (L1) comprises at least 70 wt.-%, preferably at least 80 wt.-%, more preferably at least 90 wt.-%, still more preferably at least 95 wt.-%, yet more preferably at least 99 wt.-%, of the composition (Co). In one preferred embodiment layer (L1) consists of the composition (Co).

As mentioned above the unoriented film can be produced by a blown film process or cast

film process. Accordingly the unoriented film is not subjected to a stretching step as used for
the manufacture of monoaxially or biaxially oriented polymer films. Technologies with rapid
cooling are preferred like cast film, water quenched blown film or sleeve touch technology.

In case the unoriented film is produced by cast film technology the molten polymer composition (Co) is extruded through a slot extrusion die onto a chill roll to cool the polymer to a solid film. Typically the polymer composition (Co) is firstly compressed and liquefied in

an extruder. It is possible for any additives to be already added to the polymer composition (Co) or introduced at this stage via a masterbatch. The melt is then forced through a flat-film die (slot die), and the extruded film is taken off on one or more take-off rolls, during which it cools and solidifies. It has proven particularly favorable to keep the take-off roll or rolls, by means of which the extruded film is cooled and solidified, at a temperature from 10 to 40°C, preferably from 12 to 25°C. In case a multi-layer film is produced the polymer of each respective individual layer is firstly compressed and liquefied in an extruder. A co-extrusion adapter (feed block) joins the different polymers of the respective layers to the final film structure. The melts of the different polymers are then forced simultaneously through a flat-film die (slot die), and the extruded multi-layer film is taken off on one or more take-off rolls, during which it cools and solidifies.

In the blown film process the polymer composition (Co) melt is extruded through an annular die and blown into a tubular film by forming a bubble which is collapsed between nip rollers after solidification. The blown extrusion can be preferably effected at a temperature in the range 160 to 240 °C, and cooled by water or by blowing gas (generally air) at a temperature of 0 to 50 °C to provide a frost line height of 0.5 to 8 times the diameter of the die. Tubular water quenching is preferred in order to achieve high cooling rates. The cooling water temperature is in the range of 5 to 20 °C. The blow up ratio should generally be in the range of from 1.5 to 4, such as from 2 to 4, preferably 2.5 to 3.5. In case of coextrusion the polymer melts of the polymer for the individual layers are extruded through an annular die and blown into a tubular film by forming a bubble which is collapsed between nip rollers after solidification.

Both techniques, i.e. the blown film and the cast film technique, are well known in the art and thus in the skilled knowledge (see for instance polypropylene handbook, Nello Pasquini, 2nd edition, Hanser).

The composition (Co)

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In the following the instant composition (Co) is described in more detail.

As stated above the polymer composition (Co) is defined by the mixture of a propylene copolymer (A), e.g. a heterophasic propylene copolymer (RAHECO) as defined in more detail below, and styrenic based elastomer(s) (B), i.e. preferably a styrene-ethylene/butylene-styrene (SEBS) block copolymer (B-1) and/or a hydrogenated styrene-vinyl isoprene (SIS) block rubber (B-2).

Especially good results are achievable in case the propylene copolymer (A), i.e. the heterophasic propylene copolymer (RAHECO), and the styrenic based elastomer(s) (B) are present in a specific amount. Accordingly it is appreciated that the weight ratio between the propylene copolymer (A), i.e. the heterophasic propylene copolymer (RAHECO), and the styrenic based elastomer(s) (B) [(A)/(B)] is in the range of 9/1 to 3/2, preferably in the range of 22/3 to 13/7, more preferably in the range of 4/1 to 14/6.

- Thus it is preferred that the polymer composition (Co) comprises from 10 to 40 wt.-%, more preferably from 12 to 35 wt.-%, still more preferably from 20 to 30 wt.-%, of the styrenic based elastomer(s) (B). The weight percentages given in this paragraph are based on the total amount of the polymer composition (Co), preferably based on the total amount of polymers present in the polymer composition (Co), more preferably based on the amount of the propylene copolymer (A), i.e. the heterophasic propylene copolymer (RAHECO), and the styrenic based elastomer(s) (B) together. Accordingly in one specific embodiment the polymer composition (Co) comprises
 - (a) 60 to 90 wt.-%, preferably 65 to 88 wt.-%, more preferably 70 to 80 wt.-%, of the propylene copolymer (A), i.e. the heterophasic propylene copolymer (RAHECO), and
- 25 (b) 10 to 40 wt.-%, preferably from 12 to 35 wt.-%, more preferably from 20 to 30 wt.-%, of the styrenic based elastomer(s) (B), based on the total amount of polymers present in the polymer composition (Co), more preferably based on the amount of the propylene copolymer (A), i.e. the heterophasic propylene copolymer (RAHECO), and the styrenic based elastomer(s) (B) together.

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Therefore in one specific embodiment the polymer composition (Co) comprises as polymer components only the propylene copolymer (A), i.e. the heterophasic propylene copolymer (RAHECO), and the styrenic based elastomer(s) (B), preferably the styrene-ethylene/butylene-styrene (SEBS) block copolymer (B-1). In other words the polymer composition (Co) may comprise additives but no further polymers other than propylene copolymer (A) and the styrenic based elastomer(s) (B).

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Typical additives are those mentioned below in the section propylene copolymer (A). The total amount of additives shall be not more than 10.0 wt.-%, preferably not more than 8.0 wt.-%, more preferably not more than 5.0 wt.-%, yet more preferably not more than 4.0 wt.-%, still more preferably not more than 3.0 wt.-%, in the total polymer composition (Co) and/or layer (L1).

Thus it is appreciated that the propylene copolymer (A), i.e. the heterophasic propylene copolymer (RAHECO), and styrenic based elastomer(s) (B), preferably the styrene-ethylene/butylene-styrene (SEBS) block copolymer (B-1) and/or the hydrogenated styrene-vinyl isoprene (SIS) block rubber (B-2), together constitute at least 60.0 wt.-%, more preferably at least 70.0 wt-%, yet more preferably at least 80.0 wt-%, still more preferably 90.0 wt-%, e. g. 92.0 wt.-%, yet still more preferably 95.0 wt.-%, like 97.0 wt.-%, of the polymer composition and/or layer (L1).

It has been further discovered that the properties can be further enhanced in case both main components, i.e. the propylene copolymer (A), preferably the heterophasic propylene copolymer (RAHECO), and styrenic based elastomer(s) (B), preferably the styrene-ethylene/butylene-styrene (SEBS) block copolymer (B-1) and/or the hydrogenated styrene-vinyl isoprene (SIS) block rubber (B-2), have concerted melt flow rates. Accordingly it is appreciated that the MFR₂ (230 °C) ratio of the heterophasic propylene copolymer (A) and styrenic based elastomer (B), preferably the styrene-ethylene/butylene-styrene (SEBS) block copolymer (B-1) and/or the hydrogenated styrene-vinyl isoprene (SIS) block rubber (B-2), (MFR_(A) / MFR_(B)) is equal or above 1.0. On the other hand the differences in the viscosity should be not too high. Thus a preferred the MFR_(A) / MFR_(B) ratio is of 1.0 to 3.0.

It is further appreciated that the instant polymer composition has a melt flow rate MFR₂ (230 °C) in the range of 2.5 to 15.0 g/10min, more preferably in the range of 2.5 to 12.0 g/10min, more preferably in the range of 2.5 to 8.5 g/10min, still more preferably in the range of 2.5 to 5.0 g/10min. In case the polymer composition (Co) is used as blown film material it has preferably a melt flow rate MFR₂ (230°C) in the rage of 2.5 to 5.0 g/10min, more preferably in the range of 3.0 to 4.5 g/10min. In turn in case the polymer composition (Co) is used for cast film it is preferred that the melt flow rate MFR₂ (230°C) is in the range of 3.0 to 15.0 g/10min, like in the range of 3.5 to 10.0 g/10min.

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Preferably the individual components, i.e. the propylene copolymer (A), the styrenic based elastomer(s) (B), and optionally additives, like α -nucleating agents, if not already present in the propylene copolymer (A) and/or in the styrenic based elastomer(s) (B), are mixed in an extruder.

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More precisely the polymer composition (Co) may be prepared by compounding the components within suitable melt mixing devices for preparing polymeric compounds, including in particular extruders single screw extruders as well as twin screw extruders. Other suitable melt mixing devices include planet extruders and single screw co-kneaders. Especially preferred are twin screw extruders including high intensity mixing and kneading sections. Suitable melt temperatures for preparing the compositions are in the range from 170 to 300 °C, preferably in the range from 200 to 260 °C.

The two main polymer components of the polymer composition (Co), i.e. the propylene copolymer (A), preferably the heterophasic propylene copolymer (RAHECO), and the styrenic based elastomer(s) (B), will be described now in more detail separately.

The propylene copolymer (A)

The propylene copolymer (A) comprises apart from propylene also comonomers. Preferably the propylene copolymer (A) comprises apart from propylene ethylene and/or C_4 to C_{12} α -

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olefins. Accordingly the term "propylene copolymer" according to this invention is understood as a polypropylene comprising, preferably consisting of, units derivable from

(a) propylene

and

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5 (b) ethylene and/or C_4 to C_{12} α -olefins.

Thus the propylene copolymer (A) according to this invention comprises monomers copolymerizable with propylene, for example comonomers such as ethylene and/or C₄ to C₁₂ α-olefins, in particular ethylene and/or C₄ to C₈ α-olefins, e.g. 1-butene and/or 1-hexene.

Preferably the propylene copolymer (A) according to this invention comprises, especially consists of, monomers copolymerizable with propylene from the group consisting of ethylene, 1-butene and 1-hexene. More specifically the propylene copolymer (A) of this invention comprises - apart from propylene - units derivable from ethylene and/or 1-butene. In a preferred embodiment the propylene copolymer (A) according to this invention comprises units derivable from ethylene and propylene only.

Additionally it is appreciated that the propylene copolymer (A) preferably has a rather high total comonomer content which contributes to the softness of the overall polymer composition (Co) and/or layer (L1). Thus it is required that the comonomer content of the propylene copolymer (A) is at least 7.5 wt.-%, preferably in the range of 7.5 to 16.5 wt.-%, more preferably in the range of equal or above 8.0 to 15.0 wt.-%, yet more preferably in the range of 8.0 to 13.0 wt.-%.

The propylene copolymer (A) of the instant invention can be further defined by the amount of comonomers within the xylene cold soluble (XCS) fraction. Accordingly it is preferred that the comonomer content in the xylene cold soluble fraction (XCS) of the propylene copolymer (A) is rather moderate. Thus it is appreciated that the comonomer content of the xylene cold soluble fraction (XCS) of the propylene copolymer (A) is in the range of 14.0 to 30.0 wt.-%, yet more preferably in the range of 15.0 to 28.0 wt.-%, still more preferably in the range of 16.0 to 24.0 wt.-%, like in the range of 20.0 to 24.0 wt.-%.

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Concerning the comonomers present in the xylene cold soluble fraction (XCS) it is referred to the information provided for the propylene copolymer (A). Accordingly in a specific embodiment the xylene cold soluble fraction (XCS) comprises, especially consists of, monomers copolymerizable with propylene from the group consisting of ethylene, 1-butene and 1-hexene. More specifically the xylene cold soluble fraction (XCS) comprises - apart from propylene - units derivable from ethylene and/or 1-butene. In a preferred embodiment the xylene cold soluble fraction (XCS) comprises units derivable from ethylene and propylene only.

10 Considering the information provided above, it is preferred that the propylene copolymer (A) fulfills inequation (I), more preferably inequation (Ia), yet more preferably inequation (Ib), still more preferably inequation (Ic),

$$0.30 \le \frac{Co (total)}{Co (XCS)} \le 0.75$$
 (I),

$$0.30 \le \frac{Co\ (total)}{Co\ (XCS)} \le 0.70$$
 (Ia),

$$0.35 \le \frac{Co(total)}{Co(XCS)} \le 0.65$$
 (Ib),

$$0.40 \le \frac{Co (total)}{Co (XCS)} \le 0.62$$
 (Ic),

wherein

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Co (total) is the comonomer content [wt.-%] of the propylene copolymer (A)

Co (XCS) is the comonomer content [wt.-%] of the xylene cold soluble fraction (XCS)

of the propylene copolymer (A).

In a further preferred embodiment the propylene copolymer (A) is additionally or alternatively defined by the rather overall comonomer content to its xylene cold soluble (XCS) fraction. Accordingly it is preferred that the propylene copolymer (A) fulfills in a preferred to (III) where preferred that the propylene copolymer (A) fulfills

25 inequation (II), more preferably inequation (IIa), yet more preferably inequation (IIb),

$$\frac{Co\ (total)}{XCS} \le 0.30$$
 (II)

$$0.20 \le \frac{Co\ (total)}{XCS} \le 0.30,$$
 (IIa)

$$0.25 \le \frac{Co\ (total)}{XCS} \le 0.29,$$
 (IIb)

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wherein

Co (total) is the comonomer content [wt.-%] of the propylene copolymer (A),

XCS is the content of the xylene cold soluble fraction (XCS) [wt.-%] of the

propylene copolymer (A).

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Further it is appreciated that the xylene cold soluble (XCS) fraction the propylene copolymer (A) is specified by its intrinsic viscosity. A low intrinsic viscosity (IV) value reflects a low weight average molecular weight. For the present invention it is preferably required that the xylene cold soluble fraction (XCS) of propylene copolymer (A) has an intrinsic viscosity (IV) measured according to ISO 1628/1 (at 135 °C in decalin) of below 3.0 dl/g, more preferably below 2.1 dl/g, still more preferably of in the range of 1.5 to below 3.0 dl/g, yet more preferably in the range of 1.6 to equal or below 2.1 dl/g, still yet more preferably in the range of 1.6 to equal or below 2.0 dl/g.

Additionally it is appreciated that the propylene copolymer (A) has a xylene cold soluble fraction in the range of 20.0 to 55.0 wt.-%, more preferably in the range of 25.0 to 53.0 wt.-%, still yet more preferably in the range of 25.0 to below 48.0 wt.-%, like in the range of 30.0 to 47.0 wt.-%. The remaining part of the propylene copolymer (A) being not soluble in cold xylene is the cold insoluble fraction (XCI) which is further defined below.

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Accordingly it is preferred that the comonomer content in the cold insoluble fraction (XCI) of the propylene copolymer (A) is in the range of 1.5 to 6.0 wt.-%, yet more preferably in the range of 2.0 to 5.5 wt.-%, still more preferably in the range of 2.5 to 5.5 wt.-%.

Further it is preferred that the comonomer content in both fractions is in a specific ratio to each other. Accordingly it is preferred that the propylene copolymer (A) fulfills inequation (III) more preferably inequation (IIIa), yet more preferably inequation (IIIb),

$$3.0 \le \frac{\text{Co}(XCS)}{\text{Co}(XCI)} \le 6.5 \quad \text{(III)}$$

$$3.3 \le \frac{\text{Co}(XCS)}{\text{Co}(XCI)} \le 6.0$$
 (IIIa)

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$$3.6 \le \frac{\text{Co}(XCS)}{\text{Co}(XCI)} \le 5.5$$
 (IIIb)

wherein

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Co (XCS) is the comonomer content [wt.-%] of the xylene cold soluble (XCS) of the propylene copolymer (A),

5 Co (XCI) is the comonomer content [wt.-%] of the xylene cold insoluble (XCI) of the propylene copolymer (A).

Preferably it is desired that the propylene copolymer (A) is thermo mechanically stable, so that for instance a thermal sterilization process can be accomplished. Accordingly it is appreciated that the propylene copolymer (A) has a melting temperature of at least 145 °C, more preferably in the range of 145 to 160 °C, still more preferably in the range of 145 to 159 °C, like in the range of 146 to below 151 °C.

The propylene copolymer (A) according to this invention can have a melt flow rate MFR₂ (230 °C) in the range of 2.0 to 15.0 g/10min, more preferably in the range of 2.5 to 12.0 g/10min, more preferably in the range of 2.5 to 8.5 g/10min, still more preferably in the range of 2.5 to 5.0 g/10min. In case the propylene copolymer (A) is used as blown film material it has preferably a melt flow rate MFR₂ (230°C) in the rage of 2.5 to 5.0 g/10min, more preferably in the range of 3.0 to 4.5 g/10min. In turn in case the propylene copolymer (A) is used for cast film it is preferred that the melt flow rate MFR₂ (230°C) is in the range of 3.0 to 15.0 g/10min, like in the range of 3.5 to 10.0 g/10min.

As indicated above, the propylene copolymer (A) is featured by a rather high amount of a xylene cold soluble (XCS) fraction. On the other hand the propylene copolymer (A) is also preferably featured by a rather high amount of a crystalline fraction melting at high temperature. Accordingly the instant propylene copolymer (A) is a mixture of a crystalline polymer and amorphous material. Such type of polymer is classified as heterophasic propylene copolymer. A heterophasic propylene copolymer comprises a polymer matrix, like a (semi)crystalline polypropylene, in which the amorphous material, like an elastomeric propylene copolymer, is dispersed. Thus in a preferred embodiment the instant propylene

copolymer (A) is heterophasic propylene copolymer (RAHECO). More precisely the instant propylene copolymer is heterophasic propylene copolymer (RAHECO) comprising a matrix (M) being a random propylene copolymer (R-PP) and dispersed therein an elastomeric propylene copolymer (E). Thus the matrix (M) contains (finely) dispersed inclusions being not part of the matrix (M) and said inclusions contain the elastomeric propylene copolymer (E). The term "inclusion" according to this invention shall preferably indicate that the matrix and the inclusion form different phases within the heterophasic propylene copolymer (RAHECO), said inclusions are for instance visible by high resolution microscopy, like electron microscopy or atomic force microscopy, or by dynamic mechanical thermal analysis (DMTA). Specifically in DMTA the presence of a multiphase structure can be identified by the presence of at least two distinct glass transition temperatures.

Preferably the heterophasic propylene copolymer (RAHECO) according to this invention comprises as polymer components only the random propylene copolymer (R-PP) and the elastomeric propylene copolymer (E). In other words the heterophasic propylene copolymer (RAHECO) 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 heterophasic propylene copolymer (RAHECO). One additional polymer which may be present in such low amounts is a polyethylene which is a by-reaction product obtained by the preparation of heterophasic propylene copolymer (RAHECO) (see in detail below). Accordingly it is in particular appreciated that the heterophasic propylene copolymer (RAHECO) contains only the random propylene copolymer (R-PP), the elastomeric propylene copolymer (E) and optionally polyethylene in amounts as mentioned in this paragraph.

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Preferably the weight ratio between the matrix (M), i.e. the random propylene copolymer (R-PP), and the elastomeric propylene copolymer (E) is 50/50 to 90/10, more preferably 60/40 to 85/15, yet more preferably 70/30 to 85/15.

In the following the random propylene copolymer (R-PP) and the elastomeric propylene copolymer (E) are defined more precisely.

The random propylene copolymer (R-PP) comprises monomers copolymerizable with propylene, for example comonomers such as ethylene and/or C₄ to C₁₂ α-olefins, in particular ethylene and/or C₄ to C₈ α-olefins, e.g. 1-butene and/or 1-hexene. Preferably the random propylene copolymer (R-PP) according to this invention comprises, especially consists of, monomers copolymerizable with propylene from the group consisting of ethylene, 1-butene and 1-hexene. More specifically the random propylene copolymer (R-PP) of this invention comprises - apart from propylene - units derivable from ethylene and/or 1-butene. In a preferred embodiment the random propylene copolymer (R-PP) comprises units derivable from ethylene and propylene only.

The comonomer content of the random propylene copolymer (R-PP) is preferably in the range of 3.0 to 12.5 wt.-%, yet more preferably in the range of 3.5 to 11.0 wt.-%, still more preferably in the range of 4.0 to 10.0 wt.-%, like in the range of 4.0 to 9.0 wt.-%.

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Further it is appreciated that the propylene copolymer (A) fulfills inequation (IV), more preferably inequation (IVa), yet more preferably inequation (IVb), still more preferably inequation (IVc),

$$\frac{Co\ (total)}{Co\ (RPP)} \ge 0.9,$$
 (IV)

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$$4.0 \ge \frac{Co (total)}{Co (RPP)} \ge 0.9, (IVa)$$

$$3.0 \ge \frac{Co\ (total)}{Co\ (RPP)} \ge 1.2, (IVb)$$

$$2.0 \ge \frac{Co\ (total)}{Co\ (RPP)} \ge 1.3, (IVc)$$

wherein

Co (total) is the comonomer content [wt.-%] of the propylene copolymer (A),

25 Co (RPP)

is the comonomer content [wt.-%] of the random propylene copolymer (R-PP).

The term "random" indicates that the comonomers of the random propylene copolymer (R-PP), as well as of the first propylene copolymer fraction (R-PP1) and the second propylene

copolymer fraction (R-PP2) are randomly distributed within the propylene copolymers. The term random is understood according to IUPAC (Glossary of basic terms in polymer science; IUPAC recommendations 1996).

The comonomer content of the matrix (M), i.e. of the random propylene copolymer (R-PP), has also impact on the amount of xylene cold solubles in the matrix (M). Thus it is preferred that the amount of the xylene cold soluble (XCS) fraction of the matrix (M), i.e. of the random propylene copolymer (R-PP), is in the range of 5.0 to 50.0 wt.-%, more preferably in the range of 7.0 to 45.0 wt.-%, still more preferably is in the range of 7.0 to 35.0 wt.-%.

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The random propylene copolymer (R-PP) preferably comprises at least two polymer fractions, like two or three polymer fraction, all of them being propylene copolymers. Even more preferred the random propylene copolymer (R-PP) comprises, preferably consists of, a first propylene copolymer fraction (R-PP1) and a second propylene copolymer fraction (R-PP2). It is preferred that the first propylene copolymer fraction (R-PP1) is the comonomer lean fraction whereas the second propylene copolymer fraction (R-PP2) is the comonomer rich fraction.

Thus it is preferred that the first random propylene copolymer fraction (R-PP1) has a comonomer content of equal or below 5.0 wt.-%, more preferably of equal or below 4.0 wt.-%, yet more preferably in the range 0.5 to 5.0 wt.-%, still more preferably in the range 0.5 to 4.0 wt.-%, yet still more preferably in the range 0.5 to 3.5 wt.-%.

As the comonomer of the first random propylene copolymer fraction (R-PP1) preferably is rather low, also its xylene cold soluble (XCS) is comparably low. Thus it is preferred that the amount of the xylene cold soluble (XCS) fraction of the first random propylene copolymer fraction (R-PP1) is equal or below than 12.0 wt.-%, more preferably is in the range of 1.0 to 10.0 wt.-%, still more preferably is in the range of 2.0 to 9.0 wt.-%, yet more preferably is in the range of 2.5 to 8.0 wt.-%, like in the range of 3.0 to 8.0 wt.-%. The xylene cold soluble (XCS) values provided in this paragraph are especially applicable before vis-breaking as mentioned in detail below.

On the other hand the second random propylene copolymer fraction (R-PP2) preferably has a comonomer content of at least 3.0 wt.-%, more preferably in the range 3.0 to 20.0 wt.-%, still more preferably in the range 4.0 to 19.0 wt.-%, yet more preferably in the range 5.0 to 18.0 wt.-%.

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The comonomers of the first random propylene copolymer fraction (R-PP1) and second random propylene copolymer fraction (R-PP2), respectively, copolymerizable with propylene are ethylene and/or C₄ to C₁₂ α-olefins, in particular ethylene and/or C₄ to C₈ α-olefins, e.g. 1-butene and/or 1-hexene. Preferably the first random propylene copolymer fraction (R-PP1) and second random propylene copolymer fraction (R-PP2), respectively, comprise, especially consist of, monomers copolymerizable with propylene from the group consisting of ethylene, 1-butene and 1-hexene. More specifically the first random propylene copolymer fraction (R-PP1) and second random propylene copolymer fraction (R-PP2), respectively, comprise - apart from propylene - units derivable from ethylene and/or 1-butene. In a preferred embodiment the first random propylene copolymer fraction (R-PP1) and the second random propylene copolymer fraction (R-PP1) and the second random propylene copolymer fraction (R-PP2) comprise the same comonomers, i.e. ethylene only.

Preferably the weight ratio between the first random propylene copolymer fraction (R-PP1) and the second random propylene copolymer fraction (R-PP2) is 20/80 to 80/20, more preferably 30/70 to 70/30, still more preferably 40/60 to 60/40.

As mentioned above a further component of the heterophasic propylene copolymer

(RAHECO) is the elastomeric propylene copolymer (E) dispersed in the matrix (M), i.e. in the random propylene copolymer (R-PP). Concerning the comonomers used in the elastomeric propylene copolymer (E) it is referred to the information provided for the heterophasic propylene copolymer (RAHECO) and the random propylene copolymer (R-PP), respectively. Accordingly the elastomeric propylene copolymer (E) comprises monomers copolymerizable with propylene, for example comonomers such as ethylene and/or C₄ to C₁₂ α-olefins, in particular ethylene and/or C₄ to C₈ α-olefins, e.g. 1-butene

and/or 1-hexene. Preferably the elastomeric propylene copolymer (E) 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) comprises - apart from propylene - units derivable from ethylene and/or 1-butene. Thus in an especially preferred embodiment the elastomeric propylene copolymer (E) comprises units derivable from ethylene and propylene only. It is especially preferred that the random propylene copolymer (R-PP) and the elastomeric propylene copolymer (E) comprises the same comonomers. Accordingly in one specific embodiment the random propylene copolymer (R-PP) and the elastomeric propylene copolymer (E) comprise propylene and ethylene only.

The comonomer content of the elastomeric propylene copolymer (E) preferably is not more than 35.0 wt.-%, more preferably not more than 30.0 wt.-%, still more preferably in the range of 14.0 to 35.0 wt.-%, yet more preferably in the range of 15.0 to 30.0 wt.-%.

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The instant propylene copolymer (A), i.e. the heterophasic propylene copolymer (RAHECO), is preferably obtained by a specific process. Accordingly the instant propylene copolymer (A), i.e. the heterophasic propylene copolymer (RAHECO), is preferably obtained by a sequential polymerization process comprising the steps of

- 20 (a) polymerizing in a first reactor (R1) propylene and ethylene and/or a C4 to C12 α-olefin, preferably ethylene, obtaining a first polymer fraction, i.e. a first random propylene copolymer fraction (R-PP1),
- 25 (b) transferring the first polymer fraction, i.e. the first random propylene copolymer fraction (R-PP1), into a second reactor (R2),
 - (c) polymerizing in said second reactor (R2) in the presence of the first polymer fraction, i.e. of the first random propylene copolymer fraction (R-PP1), propylene and
- ethylene and/or a C4 to C12 α -olefin, preferably ethylene,

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obtaining a second polymer fraction, i.e. a second random propylene copolymer fraction (R-PP2), the first and second polymer fraction form a first mixture, i.e. the random propylene copolymer (R-PP),

- (d) transferring said first mixture, the random propylene copolymer (R-PP), into a third reactor (R3),
 - (e) polymerizing in said third reactor (R3) in the presence of the first mixture, i.e. the random propylene copolymer (R-PP),
 propylene and
 ethylene and/or a C4 to C12 α-olefin, preferably ethylene,
 obtaining a third polymer fraction, said third polymer fraction is preferably the first
- obtaining a third polymer fraction, said third polymer fraction is preferably the first fraction of the elastomeric propylene copolymer (E); the third polymer fraction and the first mixture, i.e. the random propylene copolymer (R-PP), form a second mixture,
 - (f) transferring said second mixture into a fourth reactor (R4),

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- polymerizing in said forth reactor (R4) in the presence of the second mixture propylene and ethylene and/or a C4 to C12 α-olefin, preferably ethylene, obtaining a forth polymer fraction, said forth polymer fraction is preferably the second fraction of the elastomeric propylene copolymer (E); the forth polymer fraction and the second mixture form the propylene copolymer (A), i.e. the heterophasic propylene copolymer (RAHECO),
 - (h) removing the propylene copolymer from the fourth reactor (R4), and
 - (i) optionally visbreaking said propylene copolymer (A), i.e. said heterophasic propylene copolymer (RAHECO).

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

The term "sequential polymerization process" indicates that the propylene copolymer (A),

i.e. the heterophasic propylene copolymer (RAHECO), is produced in at least four reactors,
preferably in four reactors, connected in series. Accordingly the present process comprises at

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least 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.

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As stated above in the first two reactors the matrix (M), i.e. the random propylene copolymer (R-PP) is produced. More precisely, in the first reactor (R1) the first random propylene copolymer fraction (R-PP1) is produced whereas in the second reactor (R2) the second random propylene copolymer fraction (R-PP2).

The preferred comonomers used in the first reactor (R1) are the same as indicated above, for the first random propylene copolymer fraction (R-PP1). Accordingly especially preferred comonomers are ethylene, 1-butene and 1-hexene. In one specific embodiment the comonomer is ethylene.

Preferably the weight ratio between the first random propylene copolymer fraction (R-PP1) and the second random propylene copolymer fraction (R-PP2) is 20/80 to 80/20, more preferably 30/70 to 70/30, yet more preferably 40/60 to 60/40.

Accordingly in the first reactor (R1) a first random propylene copolymer fraction (R-PP1) is produced having a comonomer content of equal or below 5.0 wt.-%, more preferably of equal or below 4.0 wt.-%, yet more preferably in the range 0.5 to 5.0 wt.-%, still more preferably in the range 0.5 to 4.0 wt.-%.

In the second rector (R2) the second random propylene copolymer fraction (R-PP2) is produced obtaining thereby the random propylene copolymer (R-PP).

The preferred comonomers used in the second reactor (R2) are the same as indicated above, for the first reactor (R1). Accordingly especially preferred comonomers are ethylene, 1-butene and 1-hexene. In one specific embodiment the comonomer is ethylene.

The second random propylene copolymer fraction (R-PP2) preferably has commonmer content of at least 3.0 wt.-%, more preferably in the range 3.0 to 20.0 wt.-%, still more preferably in the range 4.0 to 19.0 wt.-%, yet more preferably in the range 5.0 to 18.0 wt.-%.

Thus the overall comonomer content in the second reactor (R2), i.e. the comonomer content of the random propylene copolymer (R-PP), is preferably in the range of 3.0 to 12.5 wt.-%, yet more preferably in the range of 3.5 to 11.0 wt.-%, still more preferably in the range of 4.0 to 10.0 wt.-%, like in the range of 4.0 to 9.0 wt.-%.

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The comonomers of the random propylene copolymer (R-PP), of the first random propylene copolymer fraction (R-PP1), and of the second random propylene copolymer fraction (R-PP2) copolymerizable with propylene are ethylene and/or C_4 to C_{12} α -olefins, in particular ethylene and/or C_4 to C_8 α -olefins, e.g. 1-butene and/or 1-hexene. Preferably the random propylene copolymer (R-PP), the first random propylene copolymer fraction (R-PP1), and the second random propylene copolymer fraction (R-PP2) comprise, especially consist of, monomers copolymerizable with propylene from the group consisting of ethylene, 1-butene and 1-hexene. More specifically the random propylene copolymer (R-PP), the first random propylene copolymer fraction (R-PP1) and the second random propylene copolymer fraction (R-PP2) comprise - apart from propylene - units derivable from ethylene and/or 1-butene. In a preferred embodiment the random propylene copolymer (R-PP), the first random propylene copolymer fraction (R-PP1) and the second random propylene copolymer fraction (R-PP2) comprise the same comonomers, i.e. ethylene only.

Further the first random propylene copolymer fraction (R-PP1), i.e. the polymer of the first reactor (R1), has preferably a xylene cold soluble (XCS) fraction of equal or below than 12.0 wt.-%, more preferably in the range of 1.0 to 10.0 wt.-%, still more preferably in the range of

2.0 to 9.0 wt.-%, yet more preferably in the range of 2.5 to 8.0 wt.-%, still yet more preferably in the range of 3.0 to 8.0 wt.-%.

On the other hand the second random propylene copolymer fraction (R-PP2), i.e. the polymer produced in the second reactor (R2), preferably has a xylene cold soluble (XCS) fraction of below 80 wt.-%, more preferably in the range of 10 to 80 wt.-%, yet more preferably in the range of 15 to 70 wt.-%.

Accordingly the overall xylene cold soluble (XCS) content in the second reactor, i.e. the xylene cold soluble (XCS) fraction of the random propylene copolymer (R-PP), is preferably in the range of 5.0 to 50.0 wt.-%, more preferably is in the range of 7.0 to 45.0 wt.-%, still more preferably is in the range of 7.0 to 35.0 wt.-%.

Preferably the first random propylene copolymer fraction (R-PP1) preferably has a melt flow rate MFR₂ (230 °C) in the range of in the range of 0.5 to 3.0 g/10min, more preferably in the range 1.0 to 3.0 g/10min.

On the other hand the second random propylene copolymer fraction (R-PP2), i.e. the polymer produced in the second reactor (R2), preferably has a melt flow rate MFR₂ (230 °C) in the range of 0.1 to 3.0 g/10min, more preferably in the range of 0.1 to 2.5 g/10min.

Accordingly the overall melt flow rate MFR₂ (230 °C) in the second reactor, i.e. the melt flow rate MFR₂ (230 °C) of the random propylene copolymer (R-PP), preferably is in the range of 0.2 to 3.0 g/10min, more preferably in the range 0.5 to 3.0 g/10min.

Thus after the second reactor (R2) the matrix (M), i.e. the random propylene copolymer (R-PP), of the propylene copolymer, i.e. the heterophasic propylene copolymer (RAHECO), is obtained. This matrix (M) is subsequently transferred into the third reactor (R3) in which the first fraction of the elastomeric propylene copolymer (E) is produced (step (e)).

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The preferred comonomers used in the third reactor (R3) are the same as indicated above, for the first reactor (R1). Accordingly especially preferred comonomers are ethylene, 1-butene and 1-hexene. In one specific embodiment the comonomer is ethylene.

The obtained second mixture comprises the matrix (M) in which the first fraction of the elastomeric propylene copolymer (E) is dispersed. Said second mixture has preferably a comonomer content which is higher than the comonomer content of the second reactor (R2). On the other hand the comonomer content should not be too high. Thus it is preferred that the comonomer content of the second mixture, i.e. after step (e)), is not more than 30.0 wt.-10 %, preferably in the range of 5.0 to 28.0 wt.-%, more preferably in the range of 6.0 to 20.0 wt.-%, like in the range of 6.0 to 15.0 wt.-%.

Another characteristic feature of the second mixture is its xylene cold soluble (XCS) content. Accordingly it is appreciated that the second mixture has xylene cold soluble (XCS) fraction of not more than 55.0 wt.-%, more preferably of not more than 50.0 wt.-%, still more preferably in the range of 20.0 to 55.0 wt.-%, yet more preferably in the range of 25.0 to 48.0 wt.-%, like in the range of 25.0 to 43.0 wt.-%.

The comonomer content in the xylene cold soluble fraction (XCS) of the second mixture preferably is at least 14.0 wt.-%, more preferably in the range of 14.0 to 28.0 wt.-%, yet more preferably in the range of 15.0 to 26.0 wt.-%, still more preferably in the range of 16.0 to 25.0 wt.-%.

In step (f) the second mixture is transferred into the fourth reactor (R4). In the fourth reactor (R4) the second fraction of the elastomeric propylene copolymer (E) is produced (step (g)).

The preferred comonomers used in the fourth reactor (R4) are the same as indicated above for the first reactor (R1). Accordingly especially preferred comonomers are ethylene, 1-butene and 1-hexene. In one specific embodiment the comonomer is ethylene.

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The so obtained polymer is the propylene copolymer (A), i.e. the heterophasic propylene copolymer (RAHECO), of the instant invention.

Preferably the propylene copolymer (A), i.e. the heterophasic propylene copolymer (RAHECO), after step (g) has a melt flow rate MFR₂ (230 °C) in the range of 0.5 to 2.0 g/10min, like 0.5 to 1.9 g/10min.

It is preferred that the comonomer content of the xylene cold soluble fraction (XCS) after step (g), i.e. the comonomer content of the xylene cold soluble fraction (XCS) of the propylene copolymer (A), i.e. of the heterophasic propylene copolymer (RAHECO), is in the range of 14.0 to 30.0 wt.-%, yet more preferably in the range of 15.0 to 28.0 wt.-%, still more preferably in the range of 16.0 to 24.0 wt.-%, like in the range of 20.0 to 24.0 wt.-%.

Further the propylene copolymer (A), i.e. the heterophasic propylene copolymer

(RAHECO), preferably has xylene cold soluble (XCS) fraction of at least 20.0 wt.-%, more preferably in the range of 20.0 to 55.0 wt.-%, yet more preferably in the range of 25.0 to 53.0 wt.-%, still yet more preferably in the range 25.0 to 48.0 wt.-%, like in the range of 30.0 to 47.0.

In another preferred embodiment the intrinsic viscosity (IV) measured according to ISO 1628/1 (at 135 °C in decalin) of the xylene cold soluble fraction (XCS) of propylene copolymer after step (g) preferably is below 3.0 dl/g, more preferably below 2.2 dl/g, yet more preferably in the range of 1.5 to below 3.0 dl/g, still more preferably in the range of 1.6 to equal or below 2.2 dl/g, like in the range of 1.6 to 2.0 dl/g.

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Concerning the melting temperature of the propylene copolymer (A), i.e. of the heterophasic propylene copolymer (RAHECO), after step (g) it is referred to the information provide above.

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Preferably the weight ratio between the matrix (M), i.e. the random propylene copolymer (R-PP), after step (c) and the elastomeric propylene copolymer (E) produced in the steps (e) to (g) is 50/50 to 90/10, more preferably 60/40 to 85/15, yet more preferably 70/30 to 85/15.

5 After step (g) the propylene copolymer (A), i.e. the heterophasic propylene copolymer (RAHECO), can be optionally subjected a visbreaking step (step (i)) obtaining thereby a propylene copolymer (A), i.e. a heterophasic propylene copolymer (RAHECO), with enhanced melt flow rate. The visbreaking may be carried out in any known manner, but typically the present invention envisages chemical visbreaking using a peroxide visbreaking 10 agent. Typical visbreaking agents are 2,5-dimethyl-2,5-bis(tert.butyl-peroxy)hexane (DHBP) (for instance sold under the tradenames Luperox 101 and Trigonox 101), 2,5-dimethyl-2,5bis(tert.butyl-peroxy)hexyne-3 (DYBP) (for instance sold under the tradenames Luperox 130 and Trigonox 145), dicumyl-peroxide (DCUP) (for instance sold under the tradenames Luperox DC and Perkadox BC), di-tert.butyl-peroxide (DTBP) (for instance sold under the 15 tradenames Trigonox B and Luperox Di), tert.butyl-cumyl-peroxide (BCUP) (for instance sold under the tradenames Trigonox T and Luperox 801) and bis (tert.butylperoxyisopropyl)benzene (DIPP) (for instance sold under the tradenames Perkadox 14S and Lupperox DC). Suitable amounts of peroxide to be employed in accordance with the present invention are in principle known to the skilled person and can easily be calculated on the 20 basis of the amount of propylene copolymer (A) of step (g) to be subjected to visbreaking, the MFR₂ (230 °C) value of the propylene copolymer (A) of step (g) to be subjected to visbreaking and the desired target MFR₂ (230 °C) of the product to be obtained. Accordingly, typical amounts of peroxide visbreaking agent are from 0.005 to 0.5 wt.-%, more preferably from 0.005 to 0.2 wt.-%, based on the amount of propylene copolymer 25 employed.

Typically, visbreaking in accordance with the present invention is carried out in an extruder, so that under the suitable conditions, an increase of melt flow rate is obtained. During visbreaking, higher molar mass chains of the starting product are broken statistically more frequently than lower molar mass molecules, resulting in an overall decrease of the average molecular weight and an increase in melt flow rate.

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Due to the visbreaking the melt flow rate, the amount of xylene cold solubles (XCS) and the intrinsic viscosity (IV) of the xylene cold soluble (XCS) fraction of the propylene copolymer (A), i.e. of the heterophasic propylene copolymer (RAHECO), might be affected. On the other hand the melting temperature, the total comonomer content and the comonomer content of the xylene cold soluble (XCS) fraction of the propylene copolymer (A), i.e. of the heterophasic propylene copolymer (RAHECO), are not affected. Thus the non-visbroken propylene copolymer (A) (after step (g)) and the visbroken propylene copolymer (A) (after step (ii)) have the same melting temperature, the same total comonomer content and the same comonomer content of the xylene cold soluble (XCS) fraction. Thus with regard to these embodiments reference is made to the information provided above.

On the other hand, the visbroken propylene copolymer (A) after step (i) may differ in the melt flow rate, the xylene cold soluble fraction (XCS) and the intrinsic viscosity (IV) from the non visbroken propylene copolymer (A) after step (g). Accordingly the propylene copolymer (A) after step (i) preferably has a melt flow rate MFR₂ (230 °C) in the range of 2.0 to 15.0 g/10 min, more preferably in the range of 2.5 to 12.0 g/10min, yet more preferably in the range of 2.5 to 8.5 g/10min, like in the range of 2.5 to 5.0 g/10min.

- Further it is preferred that the xylene cold soluble fraction (XCS) of propylene copolymer (A) after step (i) is in the range of 20.0 to 55.0 wt.-%, yet more preferably in the range of 25.0 to 53.0 wt.-%, still more preferably in the range of 25.0 to 48.0 wt.-%, like in the range of 30.0 to 47.0 wt.-%.
- Preferably the xylene cold soluble fraction (XCS) of propylene copolymer (A) after step (i) has an intrinsic viscosity (IV) measured according to ISO 1628/1 (at 135 °C in decalin) of below 3.0 dl/g, more preferably below 2.1 dl/g, still more preferably of in the range of 1.5 to below 3.0 dl/g, yet more preferably in the range of 1.6 to equal or below 2.1 dl/g, yet more preferably in the range of 1.6 to equal or below 2.0 dl/g.

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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 loop reactor (LR), whereas the second reactor (R2), 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 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 prepolymerization reactor is placed.

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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.

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

Preferably, in the instant process for producing the propylene copolymer (A), i.e. the heterophasic propylene copolymer (RAHECO), 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 40 °C to 110 °C, preferably between 60 °C and 100 °C, like 68 to 90 °C,
- the pressure is within the range of 20 bar to 80 bar, preferably between 40 bar to 70 bar,
- 5 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:

- 10 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), preferably in the second gas phase reactor (GPR-2), and the fourth reactor (R4), preferably in the third gas phase reactor (GPR-3), are similar to the second reactor (R2).

The residence time can vary in the three reactor zones.

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In one embodiment of the process for producing the propylene copolymer (A), i.e. the heterophasic propylene copolymer (RAHECO), the residence time the first reactor (R1), 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

25 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 (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).

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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 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.

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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.

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According to the invention the propylene copolymer (A), i.e. the heterophasic propylene copolymer (RAHECO), 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 (Iamb), preferably represented by formula (IIIa).

The process runs especially efficient by using a Ziegler-Natta catalyst system, preferably by using a Ziegler-Natta catalyst system as defined herein detail below, and a specific comonomer/propylene ratio in the second reactor (R2) and/or in the third (R3) and forth reactor (R4), respectively. Accordingly it is preferred that

(a) the comonomer/propylene ratio [Co/C3], like the ethylene/propylene ratio [C2/C3], in the second reactor (R2), i.e. in step (c), is in the range of 30 to 220 mol/kmol, more preferably in the range of 40 to 210 mol/kmol, still more preferably in the range of 40 to 190 mol/kmol,

and/or

- (b) the comonomer/propylene ratio [Co/C3], like the ethylene/propylene ratio [C2/C3], in the third reactor (R3), i.e. in step (e), is in the range of 120 to 320 mol/kmol, more preferably in the range of 130 to 310 mol/kmol, like in the range of 160 to 310 mol/kmol, and/or
 - (c) the comonomer/propylene ratio [Co/C3], like the ethylene/propylene ratio [C2/C3], in the fourth reactor (R4), i.e. in step (g), is in the range of 120 to 320 mol/kmol, more preferably in the range of 130 to 310 mol/kmol, like in the range of 160 to 310 mol/kmol

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In the following the used catalyst is defined in more detail.

The procatalyst used according to the invention 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)

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$$\bigcap_{Q \in \mathbb{R}^{2'}}^{Q \cap \mathbb{R}^{1'}} (1)$$

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

5 c) washing the product of stage b) or

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d) optionally reacting the product of step c) with additional TiCl₄.

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 MgCl₂ and a C₁-C₂ alcohol of the formula MgCl₂*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 MgCl₂*nROH, wherein R is methyl or ethyl, preferably ethyl, and n is 1 to 6, is contacting with TiCl₄ to form a titanised 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

25 (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

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(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)

$$\bigcap_{Q \in \mathbb{R}^2} \mathbb{C}^{R^1}$$
 (II)

- with R¹ and R² 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 MgCl₂*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.

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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 propylene copolymer (A), i.e. the heterophasic propylene copolymer (RAHECO), 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.

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

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

wherein R⁵ represents a branched-alkyl group having 3 to 12 carbon atoms, preferably a 5 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 R⁵ is selected from the group consisting of iso-propyl, isobutyl, iso-pentyl, tert.-butyl, tert.-amyl, neopentyl, cyclopentyl, cyclohexyl,

10 methylcyclopentyl and cycloheptyl.

Formula (IIIb) is defined by

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Si(OCH₂CH₃)₃(NR^xR^y) (IIIb)

wherein R^x and R^y can be the same or different a represent 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, isopentyl, tert.-butyl, tert.-amyl, neopentyl, cyclopentyl, cyclohexyl, methylcyclopentyl and cycloheptyl.

25 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 of formula (IIIb) is diethylaminotriethoxysilane.

30 More preferably the external donor is selected from the group consisting of diethylaminotriethoxysilane [Si(OCH₂CH₃)₃(N(CH₂CH₃)₂)], dicyclopentyl dimethoxy silane

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 $[Si(OCH_3)_2(cyclo-pentyl)_2]$, diisopropyl dimethoxy silane $[Si(OCH_3)_2(CH(CH_3)_2)_2]$ and mixtures thereof. Most preferably the external donor is dicyclopentyl dimethoxy silane $[Si(OCH_3)_2(cyclo-pentyl)_2]$.

If desired the Ziegler-Natta procatalyst is modified by polymerizing a vinyl compound in the presence of the catalyst system, comprising the special Ziegler-Natta procatalyst (component (i)), the external donor (component (iii)) and optionally the cocatalyst (component (ii)), wherein the vinyl compound has the formula:

wherein R³ and R⁴ together form a 5- or 6-membered saturated, unsaturated or aromatic ring or independently represent an alkyl group comprising 1 to 4 carbon atoms. The so modified catalyst is used for the preparation of the propylene copolymer (A), i.e. of the heterophasic propylene copolymer (RAHECO), to accomplish α-nucleation of the propylene copolymer (A) and thus of the total composition (Co) and/or layer (L1) (BNT-technology).

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The propylene copolymer (A), i.e. the heterophasic propylene copolymer (RAHECO), can be additivated prior to mixing it with the styrenic based elastomers(s) (B). In such as case the additives are added prior or after visbreaking to the propylene copolymer (A), i.e. to the heterophasic propylene copolymer (RAHECO). Typical additives are nucleating agents, antioxidants, as well as slip agents and antiblocking agents. Preferably the additive content is below 5.0 wt.-%, preferably below 3.0 wt.-%, like below 1.0 wt.-%.

In view of the use of α -nucleating agents the following should be mentioned. In principle any α -nucleating agent can be used. Examples of especially suitable α -nucleating agents are 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.

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However it is preferred that the α -nucleating agent is in particular selected from the group consisting of

- (i) salts of monocarboxylic acids and polycarboxylic acids, e.g. sodium benzoate or aluminum tert-butylbenzoate,
- (ii) dibenzylidenesorbitol (e.g. 1,3 : 2,4 dibenzylidenesorbitol) and C₁-C₈-alkyl-substituted dibenzylidenesorbitol derivatives, such as methyldibenzylidenesorbitol, ethyldibenzylidenesorbitol or dimethyldibenzylidenesorbitol (e.g. 1,3 : 2,4 bis(dimethylbenzylidene) sorbitol),
- (iii) substituted nonitol-derivatives, such as 1,2,3,-trideoxy-4,6:5,7-bis-O-[(4-propylphenyl)methylene]-nonitol,
- (iv) 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-tert-butylphenyl)phosphate], like aluminium-hydroxy-bis[2,2'-methylene-bis(4,6-di-tert-butylphenyl)phosphate] and Li-myristate (NA21),
 - (v) trisamide-benze derivatives, i.e. N-[3,5-bis-(2,2-dimethyl-propionylamino)-phenyl]-2,2-dimethyl-propionamide,
- 25 (vi) vinylcycloalkane polymer and vinylalkane polymer, and
 - (vii) mixtures thereof.

In a specific embodiment α -nucleating agents as listed under (ii), (iii), or (iv) of the previous paragraph are used.

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

The α -nucleating agent content of the propylene copolymer (A), i.e. the heterophasic 5 propylene copolymer (RAHECO), and thus of the polymer composition (Co) and/or layer (L1) is preferably up to 5.0 wt.-%. In a preferred embodiment, the propylene copolymer (A), i.e. the heterophasic propylene copolymer (RAHECO), and thus the polymer composition (Co) and/or layer (L1) contain(s) from equal or below 0.0001 to equal or below 1.0 wt.-%, more preferably from 0.0005 to 1.0 wt.-%, yet more preferably from 0.01 to 1.0 wt.-%, of a 10 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 15 especially preferred that the polypropylene composition contains a vinyleycloalkane, like vinylcyclohexane (VCH), polymer and/or vinylalkane polymer.

The styrenic based elastomers(s) (B)

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- The additional required component in the polymer composition (Co) is a styrenic thermoplastic elastomer. This additional component may be dispersed in the propylene copolymer (A), i.e. in the heterophasic propylene copolymer (RAHECO). It has been discovered that not any styrenic thermoplastic elastomer is suitable for the present invention but only a specific class of such elastomers. Accordingly for the present invention (a) styrenic based elastomer(s) (B) with a rather low styrene content, i.e. being in the range of 5 to equal or below 15 wt.-%, must be used.
 - Preferably the styrenic based elastomer(s) (B) can be a styrene-ethylene/butylene-styrene (SEBS) block copolymer (B-1) and/or a hydrogenated styrene-vinyl isoprene (SIS) block rubber (B-2).

Accordingly it is appreciated that the styrenic based elastomer(s) (B), preferably the styrene-ethylene/butylene-styrene (SEBS) block copolymer (B-1) and/or the hydrogenated styrene-vinyl isoprene (SIS) block rubber (B-2), has (have) a styrene content of equal or below 15 wt.-%, more preferably of equal or below 14 wt-%, yet more preferably of equal or below 13 wt.-%. On the other hand the styrene content in the styrenic based elastomer(s) (B), preferably in the styrene-ethylene/butylene-styrene (SEBS) block copolymer (B-1) and/or in the hydrogenated styrene-vinyl isoprene (SIS) block rubber (B-2), shall not fall below 5 wt.-%. Thus a preferred range is of 5 to 15 wt.-%, more preferred of 10 to 15 wt.-% and yet more preferred of 11 to 14 wt.-%.

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Further it is appreciated that the styrenic based elastomer(s) (B), preferably the styrene-ethylene/butylene-styrene (SEBS) block copolymer (B-1) and/or the hydrogenated styrene-vinyl isoprene (SIS) block rubber (B-2), has (have) a moderate melt flow rate MFR₂ (230 °C), i.e. not more than 20.0 g/10min, more preferably not more than 10.0 g/10min, still more preferably not more than 6.0 g/10min. On the other hand the melt flow rate of the styrenic based elastomer(s) (B), preferably the styrene-ethylene/butylene-styrene (SEBS) block copolymer (B-1) and/or the hydrogenated styrene-vinyl isoprene (SIS) block rubber (B-2), shall not fall below 1.0 g/10min. Accordingly, a preferred range is of 1.0 to 20.0 g/10min, more preferred of 2.0 to 10.0 g/10min.

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Further the styrenic based elastomer(s) (B), preferably the styrene-ethylene/butylene-styrene (SEBS) block copolymer (B-1) and/or the hydrogenated styrene-vinyl isoprene (SIS) block rubber (B-2), may be defined by its density. Thus it is appreciated that the styrenic based elastomer (B), preferably the styrene-ethylene/butylene-styrene (SEBS) block copolymer (B-1) and/or the hydrogenated styrene-vinyl isoprene (SIS) block rubber (B-2), has (have) a density of equal or below 0.905 g/cm³, more preferred in the range of 0,850 to 0.905 g/cm³.

Additionally or alternatively the styrenic based elastomer (B), preferably the styreneethylene/butylene-styrene (SEBS) block copolymer (B-1) and/or the hydrogenated styrenevinyl isoprene (SIS) block rubber (B-2), can be defined by the Shore A hardness. Thus it is appreciated that the styrenic based elastomer (B), preferably the styrene-ethylene/butylene- 41 -

styrene (SEBS) block copolymer (B-1) and/or the hydrogenated styrene-vinyl isoprene (SIS) block rubber (B-2), has/have a Shore A hardness measured according to ASTM D 2240 from 25 to 70, preferably from 30 to 60.

5 **Applications**

Further the present invention is directed to a sterilisable or sterilised unoriented film. More preferably the invention is directed to containers, i.e. pouches, especially to sterilisable or sterilised containers, i.e. pouches, comprising, preferably consisting of, the unoriented film 10 as defined in herein. The container is in particular a pouch. Further said container, i.e. pouch, has been preferably subjected to a sterilisation treatment.

In the following the present invention is further illustrated by means of examples.

15 EXAMPLES

1. 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.

20 **Calculation** of comonomer content of the second propylene copolymer fraction (R-PP2):

$$\frac{C(PP) - w(PP1)x C(PP1)}{w(PP2)} = C(PP2) \quad (I)$$

wherein

- w(PP1) is the weight fraction [in wt.-%] of the first propylene copolymer fraction (R-PP1),
- w(PP2) is the weight fraction [in wt.-%] of second propylene copolymer fraction (R-25
 - C(PP1) is the comonomer content [in wt.-%] of the first propylene copolymer fraction (R-PP1),
 - C(PP) is the comonomer content [in wt.-%] of the random propylene copolymer (R-PP),

C(PP2) is the calculated comonomer content [in wt.-%] of the second propylene copolymer fraction (R-PP2).

Calculation of the xylene cold soluble (XCS) content of the second propylene copolymer fraction (R-PP2):

$$\frac{XS(PP) - w(PP1)x XS(PP1)}{w(PP2)} = XS(PP2) \quad (II)$$

- 5 wherein
 - w(PP1) is the weight fraction [in wt.-%] of the first propylene copolymer fraction (R-PP1),
 - w(PP2) is the weight fraction [in wt.-%] of second propylene copolymer fraction (R-PP2),
- 10 XS(PP1) is the xylene cold soluble (XCS) content [in wt.-%] of the first propylene copolymer fraction (R-PP1),
 - XS(PP) is the xylene cold soluble (XCS) content [in wt.-%] of the random propylene copolymer (R-PP),
- is the calculated xylene cold soluble (XCS) content [in wt.-%] of the second propylene copolymer fraction (R-PP2), respectively.

Calculation of melt flow rate MFR₂ (230 $^{\circ}$ C) of the second propylene copolymer fraction (R-PP2):

$$MFR(PP2) = 10^{\left[\frac{\log(MFR(PP)) - w(PP1) \times \log(MFR(PP1))}{w(PP2)}\right]} (III)$$

wherein

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- w(PP1) is the weight fraction [in wt.-%] of the first propylene copolymer fraction (R-PP1),
 - w(PP2) is the weight fraction [in wt.-%] of second propylene copolymer fraction (R-PP2).
 - MFR(PP1) is the melt flow rate MFR₂ (230 °C) [in g/10min] of the first propylene copolymer fraction (R-PP1),
- 25 MFR(PP) is the melt flow rate MFR₂ (230 °C) [in g/10min] of the random propylene copolymer (R-PP),
 - MFR(PP2) is the calculated melt flow rate MFR₂ (230 °C) [in g/10min] of the second propylene copolymer fraction (R-PP2).

Calculation of comonomer content of the elastomeric propylene copolymer (E), respectively:

$$\frac{C(RAHECO) - w(PP)x C(PP)}{w(E)} = C(E) \quad (IV)$$

wherein

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w(PP) is the weight fraction [in wt.-%] of the random propylene copolymer (R-PP), i.e. polymer produced in the first and second reactor (R1 + R2),

w(E) is the weight fraction [in wt.-%] of the elastomeric propylene copolymer (E),

i.e. polymer produced in the third and fourth reactor (R3 + R4)

C(PP) is the comonomer content [in wt.-%] of the random propylene copolymer (R-PP), i.e. comonomer content [in wt.-%] of the polymer produced in the

first and second reactor (R1 + R2),

C(RAHECO) is the comonomer content [in wt.-%] of the propylene copolymer, i.e. is the comonomer content [in wt.-%] of the polymer obtained after polymerization in the fourth reactor (R4),

C(E) is the calculated comonomer content [in wt.-%] of elastomeric propylene copolymer (E), i.e. of the polymer produced in the third and fourth reactor (R3 + R4).

MFR₂ (230 °C) is measured according to ISO 1133 (230 °C, 2.16 kg load).

Comonomer content, especially ethylene content is measured with Fourier transform infrared spectroscopy (FTIR) calibrated with ¹³C-NMR. When measuring the ethylene content in polypropylene, a thin film of the sample (thickness about 250 μm) was prepared by hot-pressing. The area of absorption peaks 720 and 733 cm⁻¹ for propylene-ethylene-copolymers was measured with Perkin Elmer FTIR 1600 spectrometer. Propylene-1-butene-copolymers were evaluated at 767 cm⁻¹. The method was calibrated by ethylene content data measured by ¹³C-NMR. See also "IR-Spektroskopie für Anwender"; WILEY-VCH, 1997 and "Validierung in der Analytik", WILEY-VCH, 1997.

Styrene content

The styrene content is measured by Fourier transform infrared spectroscopy (FTIR). A thin film of 300 µm thickness is prepared from pelletized material by hot-pressing (190 °C, 100 bar, 1 minute). Per sample, two films are prepared. The so prepared film-samples are

measured by a Perkin Elmer IR-Spectrophotometer System 2000FTIR. The peak at 1602 cm⁻¹ (Phenyl-Absorption) is integrated and evaluated by using an internally established calibration curve. The arithmetic mean of two measurements is given as result.

Calibration: Various polypropylene-compounds consisting of PP and a styrene-containing elastomer (of known styrene-content) are prepared and measured according to the method described above.

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

The xylene solubles (XCS, wt.-%): Content of xylene cold solubles (XCS) is determined at 25 °C according ISO 16152; first edition; 2005-07-01. The part which remains insoluble is the xylene cold insoluble (XCI) fraction.

Melting temperature (T_m) and heat of fusion (H_f), crystallization temperature (T_c) and heat of crystallization (H_c): measured with Mettler TA820 differential scanning calorimetry (DSC) on 5 to 10 mg samples. DSC is run according to ISO 11357-3:1999 in a heat / cool /

heat cycle with a scan rate of 10 °C/min in the temperature range of +23 to +210°C. Crystallization temperature and **heat of crystallization** (H_c) are determined from the cooling step, while melting temperature and **heat of fusion** (H_f) are determined from the second heating step

Tensile modulus in machine and transverse direction was determined according to ISO 527-20 3 at 23°C on film as described in the examples below. Testing was performed at a cross head speed of 1 mm/min.

Total Penetration Energy:

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The impact strength of films is determined by the "Dynatest" method according to ISO 7725-2 on films as described in the examples below. The value "Wbreak" [J/mm] represents the total penetration energy per mm thickness that a film can absorb before it breaks. The higher this value, the tougher the material is.

Haze is determined according to ASTM D1003-00 on films as described in the examples below

Steam sterilization was performed in a Systec D series machine (Systec Inc., USA). The samples were heated up at a heating rate of 5 °C/min starting from 23°C. After having been

kept for 30 min at 121°C, they were removed immediately from the steam sterilizer and stored at room temperature till processed further.

2. Examples

polymer in the same step.

The catalyst used in the polymerization process for examples RAHECOS 1 and 2 has been 5 produced as follows: First, 0.1 mol of MgCl₂ 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 TiCl₄ 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 10 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 TiCl₄ 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 in 15 general e.g. in patent publications EP491566, EP591224 and EP586390. As co-catalyst triethyl-aluminium (TEAL) and as donor dicyclo pentyl dimethoxy silane (D-donor) was used. The aluminium to donor ratio is indicated in table 1 For RAHECO 2 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. 20 The resulting polymer was visbroken in a co-rotating twin-screw extruder (type: Coperion ZSK 57) with suitable amounts of 2,5-dimethyl-2,5-di-(tert. butylperoxy)hexane (Trigonox® 101 supplied by AKZO Nobel, Netherlands) added to the extruder as concentrate of 1 wt.-% on polypropylene powder. As additives 0.04 wt.% synthetic hydrotalcite (DHT-4A supplied by Kisuma Chemicals, Netherlands) and 0.15 wt% Irganox B 215 (1:2-blend of Irganox 25 1010 (Pentaerythrityl-tetrakis(3-(3',5'-di-tert.butyl-4-hydroxytoluyl)-propionate and tris

(2,4-di-t-butylphenyl) phosphate) phosphite) of BASF AG, Germany were added to the

Table 1: Polymerization conditions and properties for the RAHECOs 1 and 2

		RAHECO 1	RAHECO 2
TEAL/D	[mol/mol]	10	15
Loop	[
MFR ₂	[g/10min]	1.4	2.5
C2 content	[wt%]	2.0	4.2
XCS	[wt%]	4.0	3.6
C2/C3 ratio	[mol/kmol]	4.8	5.1
1 GPR			
MFR_2	[g/10min]	1.0	0.7
C2 content	[wt%]	7.3	11.2
XCS	[wt%]	29.5	38.9
C2/C3 ratio	[mol/kmol]	117	200
2 GPR			
MFR_2	[g/10min]	1.2	1.5
C2 content	[wt%]	10.1	
XCS	[wt%]	33.7	48.7
C2 of XCS	[wt%]	20.2	19.3
C2/C3 ratio	[mol/kmol]	304	154
3 GPR			
MFR_2	[g/10min]	1.1	1.8
C2 content	[wt%]	11.8	12.1
XCS	[wt%]	42.7	50.9
C2 of XCS	[wt%]	22.5	19.2
IV of XCS	[dl/g]	1.9	2.2
C2/C3 ratio	[mol/kmol]	304	159
Split			
Loop	[wt%]	41.4	36.1
1GPR	[wt%]	39.9	44.3
(2GPR+3GPR)	[wt%]	18.7	19.6
Visbreaking			
POX	[wt%]	0.01	0.015
MFR_2	[g/10min]	3.4	3.5
Tm	[°C]	147	151
XCS	[wt%]	41.0	48.0
IV of XCS	[dl/g]	1.9	2.1

C2 ethylene

IV intrinsic viscosity

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H₂/C3 ratio hydrogen / propylene ratio

C2/C3 ratio ethylene / propylene ratio

POX 2,5-dimethyl-2,5-di-(tert. butylperoxy)hexane

1/2/3 GPR 1/2/3 gas phase reactor

5 Loop Loop reactor

Table 2: Properties of the styrene-ethylene/butylene-styrene (SEBS) block copolymer

		SEBS
type		Kraton G 1645M
styrene	[wt%]	12.5
Shore A	[-]	35
MFR ₂ (230 °C)	[g/10min]	2.0
density	[g/cm ³]	0.900

Kraton G 1645M is supplied by Kraton Polymers LLC

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Table 3: Properties of PP1

		PP1
MFR total	[g/10min]	3.9
MFR matrix	[g/10min]	3.9
XCS	[wt%]	23
C2 of XCS	[wt%]	25
IV of XCS	[dl/g]	1.3
C2 total	[wt%]	8.0
Tm	[°C]	141

PP1 is the commercial product Bormed SC820CF of Borealis AG.

15 Preparation of mono-layer water-quenched blown film

The film examples according to table 5 are produced on a water quenched blown film line.

The film is blown downwards and water is used to quench the bubble in order to decrease the cooling time (by a factor 30 compared to air cooled). The detailed film processing conditions are shown in table 4.

 Table 4:
 Processing conditions for the water quenched blown film examples

			Tubular water quenching mono layer
			blown film
	T		150μm
IE1BF	Material	Unit	75% RAHECO 1 + 25% SEBS
	Melting temperature	[°C]	221
	Melt pressure	[bar]	195
	Output	[kg/h]	70
	T		
CE1BF	Material		100% RAHECO 2
	Melting temperature	[°C]	223
	Melt pressure	[bar]	205
	Output	[kg/h]	70
		T	
IE2 BF	Material		75% RAHECO 2 + 25% SEBS
	Melting temperature	[°C]	222
	Melt pressure	[bar]	198
	Output	[kg/h]	70
CEA DE	N. 6 - 4 - 1 - 1		1000/ DD1
CE2 BF	Material	50.03	100% PP1
	Melting temperature	[°C]	223
	Melt pressure	[bar]	207
	Output	[kg/h]	70
CE3 BF	Material		75% PP1 + 25% SEBS
	Melting temperature	[°C]	221
	Melt pressure	[bar]	200
	Output	[kg/h]	70
	1		
	Die temperature	[°C]	225
	Colling water temp.	[°C]	10°C
valid	BUR	[-]	1:1.3
for all films	Take off speed winder	[m/min]	9.6
1111113	Die diameter:	[mm]	220
	Die gap	[mm]	2.4

Table 5: Properties of mono-layer water-quenched blown films (150 μm)

		IE 1 BF	CE 1 BF	IE 2 BF	CE 2 BF	CE 3 BF
RAHECO 1	[wt%]	75	-	-	-	-
RAHECO 2	[wt%]	-	100	75	-	-
PP 1	[wt%]	-	-	-	100	75
SEBS	[wt%]	25	-	25	-	25
Haze, b.s.	[%]	1.8	9.3	4.6	4.3	2.4
Haze, a.s.	[%]	8.9	32.6	14.3	14.3	7.2
delta Haze	[%]	7.1	23.3	10.3	10.0	5.2
Wbreak 0°C, b.s.	[J/mm]	17	11	15	11	15
Wbreak 0°C, a.s.	[J/mm]	15	13	16	14	16
Tensile modulus MD, b.s.	[MPa]	56	200	57	302	107
Tensile modulus MD, a.s.	[MPa]	74	234	58	556	187

Preparation of three-layer cast film

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The films according to table 7 are produced on a multi layer cast film line equipped with 3 extruders. All three extruders are equipped with a notched feeding zone and a 3 zone screw with mixing and shear parts. The diameter of the cylinder of extruder A is 40 mm and the screw length 25D. Extruder B has a cylinder diameter of 60 mm and a screw length of 30D and extruder C a cylinder diameter of 45 mm and a screw length of 25D. Each extruder is fed by a gravimetric dosing system. A feed block with lamellas and following distribution was used as co-extrusion adapter: Extruder A 10% (outer layer air knife side), extruder C 80% (core layer) and extruder B 10% (inner layer chill roll side = sealing layer of the film). A coat hanger die with automatic die gap regulation was used, die width 800 mm and die gap 0.5mm. The chill roll unit has a diameter of 450 mm and the 2nd cooling roll 250 mm. The detailed processing parameters are shown in tables 6.

Table 6a: Processing conditions for the 3 layer cast film of Example CE1-CF

		Extruder A	Extruder C	Extruder B
Layer thickness	[µm]	20μm	140µm	30µm
Layer function	[-]	outer layer	core layer	sealing layer
Material	[-]	HD800CF	RAHECO 1	RB801CF
Melting temperature	[°C]	255	247	264
Melt pressure	[bar]	50	50	59
Screw speed	[U/min]	8	41	11
Output	[kg/h]	6.3	44.4	9.3

Table 6b: Processing conditions for the 3 layer cast film of Example IE1-CF

		Extruder A Extruder C		Extruder B
Layer thickness	[µm]	20μm	140µm	30µm
Layer function	[-]	outer layer	core layer	sealing layer
Material	[-]	HD800CF	75% RAHECO 1+ 25% SEBS	RB801CF
Melting temperature	[°C]	226	244	255
Melt pressure	[bar]	69	67	96
Screw speed	[U/min]	9	63	11
Output	[kg/h]	6.3	44.4	9.3

5 **Table 6c:** Processing conditions for the 3 layer cast film of Example CE2-CF

		Extruder A	Extruder C	Extruder B
Layer thickness	[µm]	20μm	140μm	30µm
Layer function	[-]	outer layer	core layer	sealing layer
Material	[-]	HD800CF	RAHECO 2	RB801CF
Melting temperature	[°C]	258	245	265
Melt pressure	[bar]	63	78	93
Screw speed	[U/min]	9	34	9
Output	[kg/h]	6.3	44.4	9.3

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Table 6d: Processing conditions for the 3 layer cast film of Example IE2-CF

		Extruder C Extruder C		Extruder B
Layer thickness	[µm]	20μm	140µm	30μm
Layer function	[-]	outer layer	core layer	sealing layer
Material	[-]	HD800CF	75% RAHECO 2+ 25% SEBS	RB801CF
Melting temperature	[°C]	244	251	273
Melt pressure	[bar]	51	43	60
Screw speed	[U/min]	10	61	11
Output	[kg/h]	6.3	44.4	9.3

Table 6e: Processing conditions for the 3 layer cast film of Example CE3-CF

		Extruder A Extruder C		Extruder B
Layer thickness	[µm]	20μm	160µm	20μm
Layer function	[-]	outer layer	core layer	sealing layer
Material	[-]	HD800CF	PP1	RD801CF
Melting temperature	[°C]	248	242	241
Melt pressure	[bar]	64	78	51
Screw speed	[U/min]	10	44	9
Output	[kg/h]	6	48	6

5 **Table 6f:** Further processing conditions for the 3 layer cast films as produced in tables 6a to 6e

Coex adapter temperature	[°C]	260
Die temperature	[°C]	250
Chill roll temperature	[°C]	12
2nd cooling roll temperature	[°C]	21
Take off speed winder	[m/min]	8,4

Table 7: Properties of three layer cast films $(20\mu m/140\mu m/30\mu m)$

		CE 1	IE 1	CE 2	IE 2	CE 3
		CF	CF	CF	CF	CF
Core layer (140µm)						
RAHECO 1	[wt%]	100	75	-	-	-
RAHECO 2	[wt%]	-	i	100	75	-
PP 1	[wt%]	-	i	-	-	100
SEBS	[wt%]	-	25	-	25	-
Properties 3 layer film						
Haze, b.s.	[%]	17	17	21	18	7
Haze, a.s.	[%]	18	16	23	21	15
delta Haze	[%]	1	1	2	3	8
Wbreak 23°C, b.s.	[J/mm]	16	28	12	35	-
Wbreak 0°C, b.s.	[J/mm]	11	15	11	15	-
Tensile modulus MD, b.s.	[MPa]	242	142	251	153	343
Tensile modulus TD, b.s.	[MPa]	217	125	232	140	293

The polymer used for the outer layer is the polypropylene homopolymer Bormed HD800CF (HD800CF), commercially available from Borealis AG, with a MFR₂ of 8 g/10min and a melting temperature Tm of 164 °C. The polymer used as sealing layer is the polypropylene random co-polymer Bormed RB801CF (RB801CF), commercially available from Borealis AG, with a MFR₂ of 1.9 g/10min and a melting temperature Tm of 140 °C.

CLAIMS

- 1. Unoriented film comprising at least one layer (L1), said layer (L1) comprises at least 70 wt.-% of a polymer composition (Co), said composition (Co) comprises
- (a) a propylene copolymer (A) having
 - (a1) a melt flow rate MFR₂ (230 °C) measured according to ISO 1133 in the range of more than 2.0 to 15.0 g/10min,
 - (a2) a comonomer content in the range of more than 7.5 to 16.5 wt.-%, and
 - (a3) a xylene cold soluble content (XCS) determined according ISO 16152 (25 °C) in the range of 20.0 to 55.0 wt.-%, wherein
 - (a4) the comonomer content of xylene cold soluble (XCS) fraction of the propylene copolymer is in the range of 14.0 to 30.0 wt.-%,
- 15 and

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- (b) a styrenic based elastomer(s) (B) having a styrene content in the range of 5 to equal or below 15 wt.-%.
- 2. Unoriented film according to claim 1, wherein the unoriented film is a mono-layer or at least three layer film comprising the layers (L1), (L2) and (L3) with the stacking order (L2)/(L1)/(L3).
 - 3. Unoriented film according to claim 1 or 2, wherein the weight ratio of propylene copolymer (A) and the styrenic based elastomer(s) (B) [(A)/(B)] in the composition (Co) is in the range of 9/1 to 3/2.
 - 4. Unoriented film according to one of the preceding claims, wherein the propylene copolymer (A)
 - (a) has a melt flow rate MFR $_2$ (230 °C) measured according to ISO 1133 in the range of more than 2.5 to below 5.0 g/10min, and/or
 - (b) fulfills inequation (I)

$$0.3 \le \frac{Co(total)}{Co(XCS)} \le 0.7$$

wherein

Co (total) is the comonomer content [wt.-%] of the propylene copolymer (A),

Co (XCS) is the comonomer content [wt.-%] of the xylene cold soluble fraction (XCS) of the propylene copolymer (A),

and/or

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fulfills inequation (II) (c)

$$\frac{Co\ (total)}{XCS} \le 0.30$$

wherein

Co (total) is the comonomer content [wt.-%] of the propylene copolymer (A), **XCS** is the content of the xylene cold soluble fraction (XCS) [wt.-%] of

the propylene copolymer (A).

- 5. Unoriented film according to one of the preceding claims, wherein the xylene cold insoluble (XCI) fraction of the propylene copolymer (A) has a comonomer content in the range of 1.5 to 6.0 wt.-%.
- 6. Unoriented film according to one of the preceding claims, wherein the propylene copolymer (A) has an intrinsic viscosity (IV) of the xylene cold soluble (XCS) fraction in the range of equal or more than 1.5 to equal or below 3.0 dl/g, wherein the intrinsic viscosity (IV) is determined according to DIN ISO 1628/1, (in Decalin at 135 °C).
- 7. Unoriented film according to one of the preceding claims, wherein the propylene copolymer (A) is a heterophasic propylene copolymer (RAHECO) comprising a matrix (M) and an elastomeric propylene copolymer (E) dispersed in said matrix (M), wherein said matrix (M) is a random propylene copolymer (R-PP).
- 8. Unoriented film according to claim 7, wherein

> (a) the weight ratio between the matrix (M) and the elastomeric propylene copolymer (E) is 50/50 to 90/10,

and/or

the comonomer content of the random propylene copolymer (R-PP) is in the (b) range of 3.0 to 12.5 wt.-%,

and/or

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- the random propylene copolymer (R-PP) has a xylene cold soluble (XCS) (c) fraction in the range of 5.0 to 50.0 wt.-%.
- 10 9. Unoriented film according to one of the preceding claims, wherein the styrenic based elastomer(s) (B) is(are) (a) styrene-ethylene/butylene-styrene (SEBS) block copolymer(s) (B-1) and/or (a) hydrogenated styrene-vinyl isoprene (SIS) block rubber(s) (B-2).
- 15 10. Unoriented film according to one of the preceding claims, wherein the polymer composition (Co) comprises as styrenic based elastomer(s) (B) only (a) styreneethylene/butylene-styrene (SEBS) block copolymer(s) (B-1) and/or (a) hydrogenated styrene-vinyl isoprene (SIS) block rubber(s) (B-2).
- 20 11. Unoriented film according to one of the preceding claims, wherein the styrenic based elastomer (B) has
 - a melt flow rate MFR₂ (230 °C) of below 10.0 g/10min, (a) and/or
 - a density of below 0.905 g/m³. (b)

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- 12. Unoriented film according to one of the preceding claims, wherein the unoriented film is a cast film or a blown film.
- 13. Unoriented film according to claim 12, wherein the cast film or blown film is 30 rapidly cooled by water or sleeve touch technology.

- 14. A container, comprising the unoriented film according to one of the preceding claims.
- 15. The container according to claim 14 or the unoriented film according to one of the preceding claims 1 to 13, wherein the container or the film has been subjected to a sterilisation treatment.

INTERNATIONAL SEARCH REPORT

International application No PCT/EP2013/056209

A. CLASSIFICATION OF SUBJECT MATTER INV. C08L23/14 B32B27/32 C08J5/18 ADD.

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

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

C08L B32B C08J

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

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

EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages		Relevant to claim No.
Υ	WO 2010/015539 A1 (BOREALIS AG SCHEDENIG TONJA [AT]; GREIN CHR [AT]; GLOGER) 11 February 2010 (2010-02-11) the whole document	1-15	
Υ	EP 1 428 854 A1 (BOREALIS TECH OY [FI]) 16 June 2004 (2004-06-16) paragraphs [0001] - [0008], [0031] claim 1		1-15
Α	EP 0 738 589 A2 (GRACE W R & CO CRYOVAC INC [US]) 23 October 1996 (1996-10-23) the whole document	[US] -/	1-15
X Furt	I her documents are listed in the continuation of Box C.	X See patent family annex.	
"A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier application or patent but published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed		"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art "&" document member of the same patent family	
Date of the actual completion of the international search		Date of mailing of the international search report	
2 May 2013		13/05/2013	
Name and r	mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040,	Authorized officer Lippert, Sigrid	

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2013/056209

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT					
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.			
		Relevant to claim No. 1-15			

INTERNATIONAL SEARCH REPORT

Information on patent family members

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