

Polyolefin composition

The present invention relates to a polyolefin composition, specifically to a polyolefin composition comprising a polyethylene polymer, which is a copolymer of ethylene and one or more C₃-C₁₂ α -olefins and a high density polyethylene wax. The compositions find utility, *inter alia*, as films and hot melt adhesives. The invention further relates to use of a high density polyethylene wax to improve film processability of a polyethylene polymer, use of a high density polyethylene wax to improve pelletisation of a polyethylene polymer, the use of said polyolefin composition as a hot melt adhesive, a film comprising said polyolefin composition and to a substrate which is at least partially coated with a hot melt adhesive, said adhesive comprising said polyolefin composition.

Background of the Invention

Challenges exist in developing polyethylene polymers, for example as films or hot melt adhesives, which have a desirable balance of properties. Linear low density polyethylenes (LLDPEs), and especially polyethylene plastomers, may cause issues during production and processing due to their low crystallinity and high softness.

Increasing the crystallisation rate of polyethylene (PE) plastomers has been previously considered. Nucleation is one possibility to improve the situation, but its value is limited as the problem lies not in the limited nucleation density (N) but in the crystal growth rate (G), both of which are required for fast crystallisation. Blends of plastomers with high density polyethylene (HDPE) are known to crystallise faster, but a rather high concentration of HDPE (≥ 20 wt%) is needed which will necessarily affect other properties of the base polymer as well. Such blends are, for example, described by Rana *et al.*, in Journal of Applied Polymer Science, 1998, 69, 2441-2450, which reports results demonstrating the loss of softness and flexibility with increasing HDPE content.

Polymer film manufacturers seek films which have excellent mechanical properties, e.g. high impact strength, puncture resistance, toughness and stiffness. The polymers used to make the film must also have good processability, i.e. during the extrusion procedure the bubble formed must be stable and the extruded film should have an even film thickness distribution.

Unfortunately, a challenge faced by the person skilled in the art is that when improving one property it seems inevitable that another property is adversely affected.

5 For example, low density polyethylene (LDPE) gives rise to films having good optical properties and can be processed at low temperatures and pressures whilst maintaining melt strength and excellent processability however films made from LDPE contribute little to mechanical properties.

Conventional unimodal Ziegler-Natta produced linear low density polyethylenes (znLLDPEs) have moderate mechanical properties but not so
10 remarkable processability, meaning both poor bubble stability and extrusion pressure.

Optical properties and puncture resistance have been improved by using metallocene linear low density polyethylenes but at the expense of processability. These polymers are sensitive to film processing conditions and lack melt strength.

15 Thus, a problem faced by the film manufacturer is that by trying to improve one property, another equally important property tends to be detrimentally affected.

Various blends of these materials have been proposed in the art to try to maximise film performance by combining the advantageous properties of certain polymers.

20 Polyolefin films are often produced by extruding the polymer melt through a suitable apparatus and, in the form of a film, into a waterbath, where the film is cooled and solidified. The film is then passed out of the waterbath and subjected to further processing steps. However, on emerging from the waterbath, the film can entrain water, which interferes with subsequent processing steps. EP 1506253
25 relates to a process for reducing this "water carry-over" by using a polyolefin composition which comprises at least one polyolefin wax.

Hot melt adhesives (HMAs) are used in many areas of commerce such as bookbinding, consumer and industrial packaging, diapers, furniture, footwear and the like. A HMA is a thermoplastic polymer composition which is placed between
30 two substrates in a molten state. Upon cooling, the HMA solidifies and crystallises, forming a bond between the substrates. Ideally, the HMA should provide good adhesion over a wide temperature range. It is also desirable for the HMA to have a short set time. The set time is defined as the time taken for the adhesive to solidify to an extent which is sufficient to provide a bond strength between the substrates
35 large enough to prevent them separating upon removal of the compression force

usually applied in the first stages of packaging production lines. A key factor which influences the set time is the crystallisation rate of the adhesive, or of one or more crystallisable components of said adhesive.

Typically, a reduction in set time must be weighed up against adhesion at
5 low temperature. There is thus a continual need to find new compositions which offer an improved balancing of these attributes. In general, PE tends to crystallise faster than any other polymer, thus making it an attractive option. However, PE plastomers combine low crystallinity with slow solidification, meaning that their use can lead to problems in production and processes. Nevertheless, employing PE
10 plastomers in a hot melt adhesive application offers some attractive prospects, namely the good transparency and softness of the base polymer in a wide temperature range, the latter resulting from the low glass transition temperature of the PE plastomers.

US 8242198 provides an adhesive composition which has advantageous
15 processing properties. The composition comprises a polyolefin base polymer, a functionalised polyolefin and at least one wax.

US 9469791 aims to provide a hot melt adhesive which has a higher rate of crystalline content formation and therefore faster cohesive strength formation. US 9469791 describes a hot melt adhesive comprising a propylene copolymer, a
20 polyethylene wax, a functionalised polyolefin, a hindered phenol compound and a propylene based elastomer.

EP 3498799 aims to develop a hot melt adhesive which has a desirable balance of properties. EP 3498799 describes mixtures of polyethylene plastomers with polypropylene copolymer wax. Although solidification is enhanced, the
25 transparency and softness of the base polymer is not fully preserved.

There thus remains the need to develop new compositions based on PE plastomers which possess suitable properties for application as HMAs and films.

It is thus an object of the present invention to provide a new composition for use as a hot melt adhesive (HMA) or for use in film applications which comprises a
30 PE polymer as the base polymer resin and which offers accelerated solidification. In particular, the polymer composition of the invention comprises a wax modifier in relatively small amounts so its presence does not manifestly alter many of the valuable properties of the base resin. Ideally, the composition will retain the transparency and softness of the base polymer. It would be advantageous if
35 increases in crystallisation rate of PE plastomers could be achieved without the

addition of large amounts of HDPE, polypropylene waxes or of soluble nucleating agents (such as 1,3:2,4-bis(3,4-dimethylbenzylidene)sorbitol, DMDBS), while preserving transparency and softness of the base polymer.

Moreover, the compositions of the invention which comprises the PE
5 polymer (A) as defined herein as the base polymer possess improved crystallisation. The present inventors surprisingly found that the addition of high density polyethylene wax is successful in enhancing solidification of a linear low density polyethylene, like a polyethylene plastomer. The effect is expressed by an increase in the rheological solidification temperature. The enhanced solidification
10 and mechanical performance is beneficial for hot melt-adhesive applications and for speeding up or stabilising pelletisation of polyethylene plastomers after production.

Summary of the Invention

15 Thus viewed from one aspect the invention provides a polyolefin composition comprising

a) 70 to 99 wt% of a polyethylene polymer (A), wherein said polyethylene polymer is a copolymer of ethylene and one or more C₃-C₁₂ α-olefins, having an MFR (190 °C/2.16 kg, ISO 1133) of 0.1 to 5.0 g/10 min, a melting point
20 T_m (DSC, ISO 11357/3) of less than 100 °C and a density (ASTM D792) of 865 to 905 kg/m³; and

b) 1 to 30 wt% of a high density polyethylene wax (B) having an MFR (190°C/2.16 kg) of more than 300 g/10 min and a T_m (DSC, ISO 11357/3) in the range of 120 to 135 °C.

25 Alternatively viewed, the invention provides a polyolefin composition comprising

a) 70 to 99 wt% of a polyethylene polymer (A), wherein said polyethylene polymer is a copolymer of ethylene and one or more C₃-C₁₂ α-olefins, having an MFR (190 °C/2.16 kg, ISO 1133) of 0.1 to 5.0 g/10 min, a melting point
30 T_m (DSC, ISO 11357/3) of less than 100 °C and a density (ASTM D792) of 865 to 905 kg/m³; and

b) 1 to 30 wt% of a high density polyethylene wax (B) having a weight average molecular weight (M_w) of 1,000 to 35,000 (GPC) and a T_m (DSC, ISO 11357/3) in the range of 120 to 135 °C.

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The invention also provides use of a high density polyethylene wax (B) having an MFR (190 °C/2.16 kg) of more than 300 g/10 min and/or having a weight average molecular weight (Mw) of 1,000 to 35,000 and a Tm in the range of 120 to 135 °C to improve film processability of a polyethylene polymer (A), wherein said
5 polyethylene polymer is a copolymer of ethylene and one or more C₃-C₁₂ α-olefins, having an MFR (190 °C/2.16 kg) of 0.1 to 5.0 g/10 min, a melting point Tm of less than 100 °C and a density of 865 to 905 kg/m³.

In another aspect the invention provides use of a high density polyethylene wax (B) having an MFR (190 °C/2.16 kg) of more than 300 g/10min having a weight
10 average molecular weight (Mw) of 1,000 to 35,000 and a Tm in the range of 120 to 135 °C to improve pelletisation of a polyethylene polymer (A), wherein said polyethylene polymer is a copolymer of ethylene and one or more C₃-C₁₂ α-olefins, having an MFR (190 °C/2.16 kg) of 0.1 to 5.0 g/10 min, a melting point Tm of less than 100 °C and a density of 865 to 905 kg/m³.

15 In a further aspect the invention provides use of a polyolefin composition as defined herein as a hot melt adhesive.

In a further aspect the invention provides a film comprising a polyolefin composition as defined herein.

20 In a further aspect the invention provides a substrate which is at least partially covered with a hot melt adhesive, said adhesive comprising a polyolefin composition as defined herein.

Brief Description of the Figures

25 Figure 1 shows storage modulus measured (a) in dynamic temperature sweep tests and (b) via dynamic mechanical thermal analysis (DMTA) tests for neat polyethylene polymers and for blends with the polyethylene wax.

Definitions

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The term molecular weight is used herein to refer to weight average molecular weight Mw unless otherwise specified.

All MFR values are MFR₂ values as determined in accordance with ISO 1133 at a load of 2.16 kg unless otherwise stated.

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Detailed Description

5 This invention relates to a new polyolefin composition which comprises a polyethylene polymer and a high density polyethylene wax. Whilst it is within the ambit of the invention for the polyolefin composition to comprise other polymer components, it is preferred if the polyethylene polymer and the high density polyethylene wax are the only polymer components in the composition.

Polyethylene polymer (A)

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The polyethylene (PE) polymer (A) in the compositions of the invention is preferably a plastomer.

The PE polymer (A) has a density (ASTM D792) of 865 to 905 kg/m³. Typically, the PE polymer will have a density of 865 to 904 kg/m³. Preferably the PE polymer (A) will have a density in the range of 865 to 903 kg/m³ and more preferably in the range of 870 to 903 kg/m³, such as 870 to 902 kg/m³.

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The MFR₂ (ISO 1133) of the PE polymer (A) at 190 °C is 0.1 to 5.0 g/10 min, preferably 0.1 to 3.0 g/10 min, more preferably 0.5 to 3.0 g/10 min, such as 0.5 to 2.0 g/10 min.

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The melting point T_m (DSC, ISO 11357/3) of the PE polymer (A) is less than 100 °C, preferably less than 95 °C. A reasonable lower limit for the melting point of suitable PE polymers may be 35 °C. A range of 45 to 95 °C is ideal.

The Mw/Mn value of the polyethylene polymer (A), representing the broadness of the molecular weight distribution (MWD), is preferably in the range of 1.5 to 4.5, more preferably 2.0 to 4.0, even more preferably 2.5 to 3.8.

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The PE polymer (A) can be unimodal or multimodal, preferably unimodal. The polyethylene polymer is a copolymer of ethylene with one or more C₃-C₁₂ alpha olefins. It will be understood that the majority by weight of the PE polymer (A) derives from ethylene monomer units. Suitable PE polymers (A) have an ethylene content from 60 to 95 wt%, preferably from 65 to 90 wt% and more preferably from 70 to 88 wt%. The comonomer contribution preferably is up to 40 mol%, more preferably up to 25 mol%. The comonomer contents of conventional ethylene plastomers are familiar to the person skilled in the art.

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The comonomers are preferably C₃-C₁₀-alpha olefins such as propene, 1-butene, 1-hexene, 1-octene, and 4-methyl-1-pentene. The use of 1-hexene, 1-

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octene and 1-butene are particularly preferred. Ideally there is only one comonomer present. That comonomer is especially 1-octene.

Preferably, the polyethylene polymer (A) is a metallocene catalysed polymer although Ziegler-Natta based polyethylene polymer (A) are also possible.

5 The PE polymer (A) may be prepared by known processes, in a one stage or two stage polymerization process, comprising solution polymerization, slurry polymerization, gas phase polymerization or combinations therefrom, in the presence of suitable metallocene catalysts, known to the art skilled persons.

10 Preferably these PE polymers (A) are prepared by a one stage or two stage solution polymerization process, especially by high temperature solution polymerization process at temperatures higher than 100 °C.

15 Such process is essentially based on polymerizing the monomer and a suitable comonomer in a liquid hydrocarbon solvent in which the resulting polymer is soluble. The polymerization is carried out at a temperature above the melting point of the polymer, as a result of which a polymer solution is obtained. This solution is flashed in order to separate the polymer from the unreacted monomer and the solvent. The solvent is then recovered and recycled in the process.

20 Preferably the solution polymerization process is a high temperature solution polymerization process, using a polymerization temperature of higher than 100 °C. Preferably the polymerization temperature is at least 110 °C, more preferably at least 150 °C. The polymerization temperature can be up to 250 °C.

 The pressure in such a solution polymerization process is preferably in a range of 10 to 100 bar, preferably 15 to 100 bar and more preferably 20 to 100 bar.

25 The liquid hydrocarbon solvent used is preferably a C5-12-hydrocarbon which may be unsubstituted or substituted by C1-4 alkyl groups such as pentane, methyl pentane, hexane, heptane, octane, cyclohexane, methylcyclohexane and hydrogenated naphtha. More preferably unsubstituted C6-10-hydrocarbon solvents are used. A known solution technology suitable for the process according to the invention is the COMPACT technology.

30 It will be appreciated that the PE polymer (A) may contain standard polymer additives.

 Suitable PE polymers (A) are available commercially and are sold under the trade name QUEO by Borealis AG. Suitable grades include Queo 7001, Queo 8201 and Queo 0201.

The polyethylene polymer (A) forms 70 to 99 wt% of the polymer composition, wherein said wt% values are relative to the total weight of the composition as a whole. In a preferable embodiment, the polyethylene (A) forms 75 to 98 wt%, such as 85 to 97 wt% of the polymer composition. The use of 92 to 97 wt% of polyethylene polymer (A) is most preferred.

High density polyethylene wax

The polyethylene wax employed in the compositions of the invention typically has a density in the range of 950 to 990 kg/m³, preferably 960 to 980 kg/m³. Preferably the polyethylene wax will have a density of at least 960 kg/m³, such as at least 970 kg/m³.

The melt flow rate (MFR) at 190 °C/2.16 kg is more than 300 g/10 min. Preferably the MFR₂ is at least 400 g/10 min, more preferably at least 450 g/10 min, such as at least 490 g/10 min. A reasonable upper limit for the MFR₂ of the polyethylene wax may be 10,000 g/10 min. The MFR₂ may be too high to measure.

The polyethylene wax employed in the compositions of the invention preferably has a weight average molecular weight (Mw) of 1,000 to 35,000, preferably is 3,000 to 25,000, such as 4,000 to 20,000.

The Mw/Mn value of the polyethylene wax, representing the broadness of the molecular weight distribution (MWD), is preferably in the range of 2 to 15 such as 2.0 to 4.0, more preferably in the range of 2.0 to 3.0, even more preferably in the range of 2.1 to 3.0.

The wax may be a homopolymer or copolymer. Preferably the wax is a homopolymer.

The polyethylene wax may be a Ziegler-Natta catalysed polymer or a metallocene catalysed polymer. Preferably the polyethylene wax is prepared using a metallocene catalyst.

The melting point (measured with DSC according to ISO 11357/3) of the polyethylene wax is in the range of 120 to 135 °C, preferably in the range of 123 to 135 °C, such as in the range of 125 to 133 °C. In one embodiment, it is preferred if the melting point of the polyethylene wax is at least 30 °C, more preferably at least 40 °C, higher than the melting point of the polyethylene polymer.

Furthermore, the polyethylene wax preferably has a melt viscosity η (140 °C) of from 100 mPa.s to 100 000 mPa.s, preferably from 100 mPa.s to 80 000

mPa.s and more preferably from 300 mPa.s to 75 000 mPa.s measured using a Brookfield Viscosimeter.

5 The polyethylene wax may be prepared by known processes, in a one stage or two stage polymerization process, comprising solution polymerization, slurry polymerization, gas phase polymerization or combinations therefrom, in the presence of suitable Ziegler-Natta or metallocene catalysts, known to the art skilled persons, optionally followed by visbreaking to adjust the Mw and MFR.

It will be appreciated that the polyethylene wax may contain standard polymer additives.

10 Suitable waxes are available commercially and are sold under trade names such as Excerox, Licowax and Licocene. Suitable waxes are mentioned in the examples such as EXCEREX 40800T and Licocene PE4201.

The polyethylene wax forms 1 to 30 wt% of the polymer composition, wherein said wt% values are relative to the total weight of the composition as a whole. In a preferable embodiment, the polyethylene wax forms 2 to 25 wt%, such as 3 to 15 wt% (relative to the total weight of the composition as a whole) of the polymer composition. It is most preferred to use 3 to 8 wt% of the polyethylene wax.

15 In a preferred embodiment the polyethylene polymer and the wax are the only polymer components in the composition other than any polymers that are part of a standard polymer additive (e.g. as a carrier in an additive masterbatch). The polymer composition of the invention therefore preferably consists essentially of the polyethylene polymer (A) and the polyethylene wax. It is preferred that no other component (other than additives) is present.

20 In one embodiment the invention provides a polyolefin composition comprising

- 25 a) 70 to 99 wt% of a polyethylene polymer (A), wherein said polyethylene polymer is a copolymer of ethylene and one or more C₃-C₁₂ α -olefins, having an MFR (190 °C/2.16 kg, ISO 1133) of 0.1 to 5.0 g/10 min, a melting point T_m (DSC, ISO 11357/3) of less than 100 °C, a density (ASTM D792) of 865 to 905 kg/m³ and having an ethylene content of 60 to 95 wt%; and
- 30 b) 1 to 30 wt% of a high density polyethylene wax (B) having an MFR (ISO1133, 190 °C/2.16 kg) of more than 300 g/10 min and/or a weight average molecular weight of 1,000 to 35,000 (GPC), a

density of 950 to 990 kg/m³, and a T_m (DSC, ISO 11357/3) in the range of 120 to 135 °C.

Polymer Composition properties

- 5 Preferably the storage modulus E'(23 °C) of the polyolefin composition is increased by 2.0 to 4.0 times for the polyolefin composition relative to the neat polyethylene polymer (ISO 6721-1, 6721-4, 6721-11). E'(23 °C) of the polyolefin composition may range from 35 to 300 MPa measured as described in the test methods section below.
- 10 Preferably the solidification temperature of the polyolefin composition as determined by dynamic rheology (T_c(rheo)) is at least 12 °C higher for the polyolefin composition than that of the neat polyethylene polymer. T_c (rheo) values may range from 50 to 120 °C measured as described in the test methods section below.
- 15 Preferably the softening temperature increases for the polyolefin composition by 10 to 30 °C relative to the neat polyethylene polymer (ISO 6721-1, 6721-4, 6721-11). T_{soft} values may range from 55 to 120 °C measured as described in the test methods section below.

20 Applications

- It is envisaged that one benefit of the composition of the invention is that it aids the successful pelletisation of the polyethylene polymer (A). The use of the polyethylene wax may ensure that pelletisation of the polyethylene polymer (A) is
- 25 accelerated and stabilised. In one embodiment the invention provides use of a high density polyethylene wax (B) having an MFR (190 °C/2.16 kg) of more than 300 g/ 10 min and/or a Mw of 1000 to 35,000 and a T_m in the range of 120 to 135 °C to improve pelletisation of a polyethylene polymer (A), wherein said polyethylene polymer is a copolymer of ethylene and one or more C₃-C₁₂ α-olefins, having an
- 30 MFR (190 °C/2.16 kg) of 0.1 to 5.0 g/ 10 min, a melting point T_m of less than 100 °C and a density of 865 to 905 kg/m³.

 It is envisaged that another benefit of the composition of the invention is that it aids film processability, e.g. in terms of melt fracture and/or bubble stability. The addition of the wax therefore makes films easier to process.

It is particularly preferred if the polymer composition of the invention is employed in hot melt adhesive compositions. Viewed from another aspect therefore the invention provides a hot melt adhesive comprising a polyolefin composition comprising

- 5 a) 70 to 99 wt% of a polyethylene polymer (A), wherein said polyethylene polymer is a copolymer of ethylene and one or more C3-C12 α -olefins, having an MFR (190 °C/2.16 kg, ISO 1133) of 0.1 to 5.0 g/10 min, a melting point T_m (DSC, ISO 11357/3) of less than 100 °C and a density (ASTM D792) of 865 to 905 kg/m³; and
- 10 b) 1 to 30 wt% of a high density polyethylene wax (B) having an MFR (190 °C/2.16 kg) of more than 300 g/10 min and/or a M_w of 1000 to 35,000 and a T_m (DSC, ISO 11357/3) in the range of 120 to 135 °C.

The invention also provides the use of a polyolefin composition as hereinbefore defined as a hot melt adhesive.

- 15 The polyolefin composition of the invention may be the only component of the hot melt adhesive or the adhesive may further comprise one or more of tackifiers, plasticizers and other waxes. The polyolefin composition can therefore form 10 to 100 wt% of the HMA.

- 20 If present, tackifiers may preferably be selected from the group consisting of aliphatic, alicyclic and aromatic resin, and modified material and hydrogenated derivatives thereof, rosin and modified materials and derivatives of rosin, terpenes and modified materials and derivatives of terpenes. The tackifier can be one of the above resins, or can be a combination of more than one of the above resins.

Thus, as used herein, the term "tackifier" includes:

- 25 (a) aliphatic and cycloaliphatic petroleum hydrocarbon resins having Ring and Ball softening points of from 10 °C to 160 °C, as determined by ASTM method E28-58T, the latter resins resulting from the polymerization of monomers consisting primarily of aliphatic and/or cycloaliphatic olefins and diolefins; also included are the hydrogenated aliphatic and cycloaliphatic petroleum hydrocarbon resins; examples
- 30 of such commercially available resins based on a C5 olefin fraction of this type are Piccotac 95 tackifying resin sold by Hercules Corp. and Escoreze 1310LC solid by ExxonMobil Chemical Company;

(b) Aromatic petroleum hydrocarbon resins and the hydrogenated derivatives thereof;

(c) Aliphatic/aromatic petroleum derived hydrocarbon resins and the hydrogenated derivatives thereof;

(d) Aromatic modified cycloaliphatic resins and the hydrogenated derivatives thereof;

5 (e) Polyterpene resins having a softening point of from about 10 °C to about 140 °C, the latter polyterpene resins generally resulting from the polymerization of terpene hydrocarbons, such as the mono-terpene known as pinene, in the presence of Friedel-Crafts catalysts at moderately low temperatures; also included are the hydrogenated polyterpene resins;

10 (f) Copolymers and terpolymers of natural terpenes, e.g. styrene/terpene, α -methyl styrene/terpene and vinyl toluene/terpene;

(g) natural and modified rosin such as, for example, gun rosin, wood rosin, tall-oil rosin, distilled rosin, hydrogenated rosin, dimerized rosin and polymerized rosin;

15 (h) glycerol and pentaerythritol esters of natural and modified rosin, such as, for example, the glycerol ester of pale wood rosin, the glycerol ester of hydrogenated rosin, the glycerol ester of polymerized rosin, the pentaerythritol ester of pale wood rosin, the pentaerythritol ester of hydrogenated rosin, the pentaerythritol ester of tall-oil rosin, and the phenolic modified pentaerythritol ester of rosin;

20 (i) phenolic-modified terpene resins such as, for example, the resin product resulting from the condensation in an acidic medium of a terpene and a phenol.

Mixtures of two or more of the above described tackifiers may be required for some formulations.

25 Preferably, the tackifiers can be selected from any of the nonpolar types, which are commercially available. e.g. Eastotac from Eastman Chemical Co., Escorez from Exxon Chemical Co., Wingtack from Goodyear Chemical Co., Herculite from Hercules Inc., Zonatac from Arizona Chemical Co. Preferred resins are aliphatic petroleum hydrocarbon resins examples of which are based on C5
30 olefins such as Hercotac 1148 available from Hercules Corp. Most preferred are nonpolar products which are hydrogenated di-cyclo-penta-diene (DCPD) based or aromatically modified derivatives thereof with softening points above 70 °C.

Examples of such resins are Escoreze 5400 and Escoreze 5600 sold by ExxonMobil Chemical company.

If one or more tackifiers are present in the hot melt adhesive they may typically form between 10 and 70 wt% (based on the total weight of the HMA), preferably in the range of 15 to 60 wt%.

5 The hot melt adhesive of the present invention can optionally further comprise plasticizer(s) and/or other wax component(s).

The plasticizer(s) which can be utilized in the hot melt adhesive of the present invention provide viscosity control and wetting and specific adhesion to bonded substrates. These plasticizers may be selected from a group comprising paraffin oils, chlorinated paraffines, phthalates and adipate esters, oligomers of
10 polypropylene, polybutenes, polyisoprene, hydrogenated polyisoprene and polybutadiene, benzoate esters (e.g. 1,4-cyclohexane dimethanol dibenzoate, glyceryl tribenzoate or pentaerythritol tetrabenzoate), and vegetable and animal oils and derivatives thereof and mixtures of two or more of any of the foregoing.

The preferred primary plasticizer is a benzoate or paraffinic oil, a
15 commercial grade of paraffinic oil widely used is available from Witco under the tradename Kaydol oil.

Plasticizers can broadly represent from about 0 to about 60 wt% of the hot melt adhesive of this invention and preferably represent from about 15 to about 25 wt% of the hot melt adhesive of this invention.

20 Wax components, other than the polyethylene wax, can also be optionally utilized in the HMA of the present invention. Wax components are useful for multi-purpose hot melt adhesives, in that these components offer additional wetting and added creep resistance for foamed elastic attachments. These wax additives can be selected from a group comprised of paraffin wax, microcrystalline wax, Fischer-Tropsch wax, fatty amide waxes, polyethylene wax, ethylene vinyl acetate wax,
25 oxidized polyethylene wax, hydrogenated castor oil and derivatives thereof, polyethylene wax and mixtures of two or more of any of the foregoing. The preferred waxes for the present invention are selected from a group of oxidized polyethylene waxes, of which a commercial grade is available from Allied Signal
30 under the tradename AC-395. Additional wax component(s) can represent from 0 to about 10 wt% of the hot melt adhesive of this invention and preferably represent from about 3 to about 7 wt% of the hot melt adhesive of this invention.

If necessary, the hot melt adhesive according to the present invention may further contain various additives. Examples of the various additives include a
35 stabilizer and fine particle filler.

The "stabilizer" is blended so as to improve stability of the hot melt adhesive by preventing decrease in molecular weight, gelation, coloration, and generation of odour of the hot melt adhesive due to heat, and there is no particular limitation on the stabilizer as long as the objective hot melt adhesive of the present invention can be obtained. Examples of the "stabilizer" include an antioxidant and an ultraviolet absorber.

The "ultraviolet absorber" is used so as to improve light resistance of the hot melt adhesive.

The "antioxidant" is used so as to prevent oxidation degradation of the hot melt adhesive.

The antioxidant and ultraviolet absorber are commonly used in disposable products and can be used without particular limitation as long as the below-mentioned objective disposable products can be obtained.

Examples of the antioxidant include a phenol-based antioxidant, a sulphur-based antioxidant and a phosphorus-based antioxidant. Examples of the ultraviolet absorber include a benzotriazole-based ultraviolet absorber and a benzophenone-based ultraviolet absorber. It is also possible to add a lactone-based stabilizer. These additives can be used alone, or in combination.

It is possible to use, as the stabilizer, commercially available products. Examples thereof include SUMILIZER GM (trade name), SUMILIZER TPD (trade name) and SUMILIZER TPS(trade name) manufactured by Sumitomo Chemical Co. Ltd.; IRGANOX 1010 (trade name), IRGANOX HP2225FF (trade name), IRGAFOS 168 (trade name) and IRGANOX 1520 (trade name) manufactured by Ciba Specialty Chemicals Inc.; and JF77 (trade name) manufactured by Johoku Chemical Co., Ltd. These stabilizers can be used alone, or in combination.

The hot melt adhesive for disposable products of the present invention can further include fine particle fillers. Commonly used fine particle fillers may be used, and there is no particular limitation as long as the objective hot melt adhesive of the present invention can be obtained.

Examples of the "fine particle filler" include mica, calcium carbonate, kaolin, talc, titanium oxide, diatomaceous earth, urea-based resin, styrene beads, calcined clay, starch and the like. These particles preferably have a spherical shape, and there is no particular limitation on the size (diameter in case of a spherical shape).

Where components other than the polyolefin composition of the invention are present in the hot melt adhesive, the adhesive may be produced by blending

the polyolefin composition of the invention with such components, melting the mixture with heating, followed by mixing. Specifically, the hot melt adhesive can be produced by charging the above components in a melt-mixing vessel equipped with a stirrer, followed by heating and mixing.

5 In a further embodiment, the invention provides a substrate which is at least partially coated with a hot melt adhesive, said adhesive comprising a polyolefin composition as hereinbefore defined.

 The hot melt adhesive of the present invention may be applied to a desired substrate by any method known in the art, and include, without limitation roll
10 coating, painting, dry-brushing, dip coating, spraying, slot-coating, swirl spraying, printing (e.g., ink jet printing), flexographic, extrusion, atomized spraying, gravure (pattern wheel transfer), electrostatic, vapor deposition, fiberization and/or screen printing.

 The hot melt adhesive compositions of the invention are useful as
15 construction adhesives, core adhesives or elastic adhesives, and are particularly suitable for being used for packaging applications and disposables.

 Examples of disposables are diapers, adult incontinent products, bed pads, a sanitary napkin, a pet sheet, a hospital gown, surgical capes, drapes or white garment and the like.

20 Packaging applications are for example like liquid packaging for milk, juice, wine or other liquids, flexible packaging for food like meat and cheese and medical products, rigid packaging like detergent cartons, cup and plate boards for oven or microwave use or sterilizable food packaging, but also for photographic paper or industrial applications like paper reel and ream wraps.

25 In addition the hot melt adhesive composition of the invention can be used in the field of bookbinding, furniture, footwear and transportation.

 It is particularly preferred if the polymer composition of the invention is employed in film applications.

 Films comprising the polymer composition according to the present
30 invention can be produced with several known conversion techniques, such as extrusion via blown or cast film technology, wherein blown films are preferred.

 Films according to the present invention may be subjected to post-treatment processes, e.g. surface modifications, lamination or orientation processes or the like. Such orientation can be mono-axially (MDO) or bi-axially orientation wherein
35 mono-axial orientation is preferred.

The films according to the present invention typically have a thickness of 100 μm or below, such as 70 μm or below or 60 μm or below. Preferred are films having a thickness of 10 μm or higher, such as 15 μm or higher, such as 18 μm or 20 μm or 22 μm or 25 μm or higher. More preferred are films having a thickness of 10-70 μm , such as 15-60 μm or 18-55 μm .

Films according to the present invention may be mono- or multilayer films, comprising one or more layers, like two, three or five layers, even up to seven, up to 9 or up to 12 layers.

It is within the scope of the present invention, that a monolayer film may comprise 1-100 wt% of the polyolefin composition according to the present invention. It is also within the scope of the invention that such monolayer film can comprise 1-90 wt%, such as 30-70 wt%, or like 40-60 wt% or 45-55 wt% of the polyolefin composition according to the present invention.

However, a monolayer film comprising 100 % of the polyolefin composition of the present invention is preferred.

It is within the scope of the invention, that the films are characterised by a high DDI. i.e. a DDI of at least 400 g or higher, such as 470 or 550 or higher such as 650; 750 g or above. An upper limit of 2500 g is possible.

It is within the scope of the invention that the films are characterised by a high relative DDI (i.e. DDI/film-thickness), i.e. a relative DDI of 15 g/ μm or above, such as 18 or 25 g/ μm or above. It is especially preferred that films of the present invention have a relative DDI of 30 g/ μm or above.

Films of the invention may be of low haze, e.g. 10 % or less.

Films of the invention may have a low seal initiation temperature, such as within the range of 75 to 90 $^{\circ}\text{C}$.

The films according to the present invention are highly useful to being used in various packaging applications, wherein applications related to food packaging are preferred.

Films comprising the polyolefin composition of the current invention comprise shrink films, collation shrink films, wrap films, lamination films, etc.

Packaging articles comprising the polyolefin composition of the current invention comprise bags, pouches, wrapping or collation films, and the like.

It is within the scope of the present invention that the polyolefin composition may comprise additives as used in the art, such as phenolic stabilizers,

antioxidants, slip and antistatic agents, antiblock agents processing aids, colorants and the like.

The invention will now be described with reference to the following non-limiting examples and figures.

5

Test methods

MFR₂ (190 °C) is measured according to ISO 1133 (190 °C, 2.16 kg load).

10

Density

Density of the polymer was measured according to ASTM; D792, Method B (density by balance at 23°C) on compression moulded specimen prepared according to EN ISO 1872-2 and is given in kg/m³.

15

Melting temperature T_m was measured with a TA instruments Q2000 differential scanning calorimetry device (DSC) according to ISO 11357/3 on 5 to 10 mg samples. Melting temperatures were obtained in a heat/cool/heat cycle with a scan rate of 10 °C/min between 30 °C and 180 °C. Melting and crystallisation temperatures were taken as the peaks of the endotherms and exotherms in the cooling cycle and the second heating cycle respectively. (DSC tests were carried out at 10 °C/min from -40 to 180 °C (ISO 11357-3, BTM 00121).

20

Determination of the “softening temperature” using dynamic mechanical thermal analysis – tensile stressed

25

In a dynamic mechanical thermal analysis in tensile mode the sample is subjected to a constant load together with an applied sinusoidal tensile strain. Under low enough deformation material response is kept within the linear viscoelastic region which is independent of strain amplitude.

30

The tensile storage modulus E' (1) and the tensile loss modulus E'' (2) are determined from the following equations (“Dynamic mechanical analysis : a practical introduction” Kevin P. Menard © 2008 by Taylor & Francis Group, LLC, Dynamic Testing and Instrumentation, 71-76, 2008)

$$E' = \frac{\Delta F_A}{s_A} * \frac{L_a}{bd} * \cos \delta \text{ [Pa]} \quad (1)$$

$$E'' = \frac{\Delta F_A}{s_A} * \frac{L_a}{bd} * \sin \delta \text{ [Pa]} \quad (2)$$

5

where

ΔF_A is the measured amplitude of dynamic force, in newton

s_A is the measured amplitude of the dynamic displacement, in metres

10 L_a is the distance between the clamps, in metres

b is the width of the specimen, in metres

d is the thickness of the specimen, in metres

δ is the measured phase angle, in degrees.

15 The determination of the so-called damping factor is done as described in following equation.

$$\tan \delta = \frac{E''}{E'} \text{ [1]}$$

20 The characterization of dynamic-mechanic properties complies with ISO standards 6721-1, 6721-4, 6721-11. The measurements is performed on a "Netzsch DMA 242E Artemis" strain/stress-controlled dynamic mechanical Analyzer, equipped with a tensional-sample holder for rectangular specimen geometry. Measurements are undertaken on rectangular sample cut from compression moulded plates produced

25 with a "Collin 400P/M" thermo-press, using 200 °C and an annealing time of 300 seconds for melting at a pressure of 5 bar, then a compression pressure of 25 bar is used for 300 seconds and a pressure of 50 bar for cooling down to room-temperature using a cooling rate of 15 K/min. The compression moulded plate with a suitable geometry of is prepared and stored for a minimal rest time of 96 hours

30 after compression moulding. The rectangular sample prepared using a laboratory cutter for clamping the specimen. The free tensile-length is about 10 mm (+0.5mm) measured with a calliper at room temperature with an accuracy of 0.05 mm. The

width and the thickness are measured using a suitable length gauge with an accuracy of 0.001 mm. The dynamic mechanic thermal analysis is performed under inert atmosphere using liquid nitrogen for cooling within the temperature range from -170 °C to +130 °C using a heating rate of 2 K/min, a frequency of 1 Hz, in strain-stress controlled mode with a maximum dynamic applied stress of 10 MPa, a static load of 0.20 MPa and a maximum strain amplitude of 0.20 %. The clamping of the specimens are performed using a torque of 7.5 cNm on screws. Clamping of soft-materials are performed after cooling down to -100°C to avoid deformation within the clamps. The annealing at the start-temperature of -170 °C is carried out with an isothermal section of 20 minutes. The evaluation is performed using instrument software "Proteus Thermal Analysis – Version 6.1.0" to point out softening temperature $T(E'=10\text{MPa})$ [°C] of the E' /temperature-plot.

Determination of solidification temperature using dynamic shear temperature sweep

The measurements were performed on a modular compact rheometer 501 series - instrument of Anton Paar equipped with convection oven and parallel plates with a diameter of 25 mm. The sample discs with a diameter of 25mm were compression moulded at 200 °C to a thickness of about 1.5 mm and cooled to room temperature within 5 minutes. The temperature-sweep experiment was separated into 2 segments using from start temperature of 180 °C to 120 °C a cooling rate of 2 K/min & a datapoint acquisition of 1 pts/K. In the second segment from 120 °C to 30 °C a cooling rate of 1 K/min and a datapoint acquisition of 2 pts/K was used. After introduction of the sample a gap of 1.3 mm was set after trimming and an annealing time of 5 min at the start temperature was carried out. A strain within the linear viscoelastic regime of the material and a constant frequency of 6.28 rad/s was used. To ensure proper contact of the sample within the parallel plates a normal force of 0.2 N was applied the whole temperature sweep test. The experiment was stopped automatically after achieving a torque value greater than 100mNm. The achieved storage modulus was plotted over temperature to interpolate to the temperature when G' equals 106 Pa. If the storage modulus did not reach the value of 106 Pa during the experiment a second plot of the first derivative of the loss factor ($\tan \Delta$) over temperature was done and an onset temperature of first derivative function was determined from the liquid like region ($G' < G''$) to the transition region ($G' \approx G''$). A determination of the solidification temperature

(Tc(Rheo)) was then carried out fitting T/G'-function using following equation (eq. 1) with a datapoint interval setting starting from the previously achieved onset temperature to the last experimental datapoint reached during the temperature sweep test.

5 Eq.1)

$$T_{G'} = \frac{C_1}{(1 + (C_2 * G')^{(1-C_3)})}$$

Where c_1 is the starting temperature of solidification expressed in [°C], c_2 is the solidificational compliance expressed in [1/Pa] and c_3 is the solidificational exponent without unit.

10

Tensile test

The tensile modulus was determined in accordance with ISO 527-3 on the film described below.

15

GPC

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

20

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

25

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

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

30 For a constant elution volume interval ΔV_i , where A_i , and M_i are the chromatographic peak slice area and polyolefin molecular weight (MW),

respectively associated with the elution volume, V_i , where N is equal to the number of data points obtained from the chromatogram between the integration limits.

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

The column set was calibrated using universal calibration (according to ISO 16014-2:2003) with 19 narrow MWD polystyrene (PS) standards in the range of 0,5 kg/mol
15 to 11 500 kg/mol. The PS standards were dissolved at room temperature over several hours. The conversion of the polystyrene peak molecular weight to polyolefin molecular weights is accomplished by using the Mark Houwink equation and the following Mark Houwink constants:

20 $K_{PS} = 19 \times 10^{-3} \text{ mL/g}$, $\alpha_{PS} = 0.655$
 $K_{PE} = 39 \times 10^{-3} \text{ mL/g}$, $\alpha_{PE} = 0.725$
 $K_{PP} = 19 \times 10^{-3} \text{ mL/g}$, $\alpha_{PP} = 0.725$

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

25 All samples were prepared in the concentration range of 0,5 -1 mg/ml and dissolved at 160 °C for 2.5 hours for PP or 3 hours for PE under continuous gentle shaking.

Impact resistance on film (DDI) is determined by Dart-drop (g/50%). Dart-drop is
30 measured using ISO 7765-1, method "A". A dart with a 38 mm diameter hemispherical head is dropped from a height of 0.66 m onto a film clamped over a hole. If the specimen fails, the weight of the dart is reduced and if it does not fail the weight is increased. At least 20 specimens are tested. The weight resulting in failure of 50% of the specimens is calculated.

Haze The haze was determined according to ASTM D1003-00 on the film described below.

Sealing initiation temperature (SIT); sealing end temperature (SET), sealing range:

- 5 The method determines the sealing temperature range (sealing range) of polyethylene films, in particular blown films or cast films. The sealing temperature range is the temperature range, in which the films can be sealed according to conditions given below.
- The lower limit (heat sealing initiation temperature (SIT)) is the sealing temperature
- 10 at which a sealing strength of 5 N is achieved. The upper limit (sealing end temperature (SET)) is reached, when the films stick to the sealing device.
- The sealing range is determined on a J&B Hot Tack Tester with a film of 25 µm thickness with the following further parameters:
- 15 Specimen width: 25 mm
Seal Pressure: 0.4 N/mm²
Seal Time: 1 sec
Cool time: 30 sec
Grip separation rate: 42 mm/sec
- 20 Start temperature: 80 °C
End temperature: 150 °C
Increments: 5 °C

Materials used

- 25 Polyethylene polymers (A)
Queo 7001, Queo 8201 and Queo 0201 are commercially available from Borealis and details are provided in Table 1.
- 30 LLDPE A is described in WO2022/268963 as IE1
- LLDPE B is produced in Borstar pilot plant, with same catalyst and plant set up as for LLDPE A. The details are in Table 2. The polymers were mixed with 2400 ppm of Irganox B561. 270 ppm of Dynamar FX 5922 compounded and extruded under

nitrogen atmosphere to pellets by using a JSW extruder so that the SEI was 230 kWh/kg and the melt temperature 250°C. The properties of pellets are in Table 1.

High density polyethylene Waxes

- 5
- 10
- 15
- 20
- Licowax PE190, a ZN-based HDPE from Clariant (Germany), with melt viscosity η (140 °C) 25 Pa·s, molecular weight (Mw) 19,200 g/mol, MWD 3.1, density 960 kg/m³, softening temperature (T_{soft}) 132-138 °C. T_m 126°C
 - Excerox 40800, is a single-site catalyst (SSC)-based polyethylene from Mitsui chemicals, with melt viscosity η (140 °C) 0.43 Pa·s (Brookfield), Mw 6955 g/mol, MWD 2.6.
 - HDPE Wax C, is produced in Borstar pilot plant, with Prepoly reactor -loop set up. The catalyst was Lynx 200, which is a commercially available Ziegler-Natta catalyst manufactured and supplied by Grace Catalysts Technologies. Typical conditions are in Table 2. The polymer was mixed with 2400 ppm of Irganox B561 and 500ppm of Calcium Stearate PLC Vegetable MB (Faci S.p.A.). Melt viscosity η (140 °C) 68 Pa·s, Mw 31150 g/mol, MWD 9.5, further properties are shown in Table 1.

The properties of the polyethylene polymers and waxes used in the examples are shown in Table 1.

Table 1. Components for inventive and comparative compositions.

Base polymers		Queo 7001	Queo 8201	Queo 0201	LLDPE A	LLDPE B (+ additives)
MFR 190 °C/2.16 kg	g/10 min	1.0	1.1	1.1	1.3	0.6
Density	kg/m ³	870	883	902	913	926
Tm (DSC)	°C	59	73	94	123	128
Tc (Rheo)	°C	44	62	85	115	119
E' (23 °C)	MPa	16	36	96	790	712
T soft	°C	41.5	62.8	89.8	>125	>125

Base polymers		Queo 7001	Queo 8201	Queo 0201	LLDPE A	LLDPE B (+ additives)
Modifiers (PE waxes)		Licowax PE190	Wax C (+ additives)	Excerec 40800		
MFR 190 °C/2.16 kg	g/10 min	>500	387	>500		
Mw		19200	31150	6955		
Density	kg/m ³	960	970	980		
Tm (DSC)	°C	126	131	128		
Tc (Rheo)	°C	-	119.9	-		

Table 2. Polymerization parameters

	LLDPE B	Wax C
Prepoly reactor		
Temp. (°C)	50	60
Catalyst feed (g/h)	53,4	2,0
Press. (kPa)	5699	5493
C2 (kg/h)	4,0	2,0
H2 (g/h)	0,04	5,00
C4 (g/h)	88,7	-
Split (%)	3,6	3,2
First loop		
Temp. (°C)	85	not use
Press. (kPa)	5446	
C2 conc. (mol-%)	3,7	
H2/C2 ratio (mol/kmol)	0,45	
C4/C2 ratio (mol/kmol)	4,8	
Split %	21,3	
Density (kg/m3)	954,0	
MFR2 (g/10 min)	11,0	
Second loop		
Temp. (°C)	85	95
Press. (kPa)	5265	5199

	LLDPE B	Wax C
C2 conc. (mol-%)	5,0	3,2
H2/C2 ratio (mol/kmol)	1,5	977,9
C4/C2 ratio (mol/kmol)	1	0
Split (%)	22,6	96,8
Density (kg/m ³)	966,0	970,0
MFR2 (g/10 min)	63,4	387,0
GPR		
Temp. (°C)	75	not used
Press. (kPa)	2000	
H2/C2 ratio (mol/kmol)	0,61	
C4/C2 ratio (mol/kmol)	0,00	
C6/C2 ratio (mol/kmol)	12,69	
Split %	52,5	
Powder		
Density (kg/m ³)	926,0	970,0
MFR2 (g/10 min)	0,67	387,00

Examples

95 wt% of base polymer and 5 wt% of wax were compounded together. All of the compounding was done on a ZSK 18 twin screw extruder, with a melt temperature of 210 °C, production rate of 7 kg/h. Comparative examples 1-5 do not comprise wax. Comparative examples 6-9 comprise wax but LLDPEs A and B have a density outside the range required in the present invention. The properties of the various compositions are set out in Table 3.

10

The effect of the polyethylene wax modifier on crystallisation of the base polymers is shown in Figure 1. Figure 1a shows an increase in rheological crystallisation temperature and Figure 1b shows an increase in terminal softening temperature. This combined effect is achieved by the crystal network structure developed by the polyethylene wax.

15

Table 3. Characterisation results of inventive and comparative compound examples (all modifiers added at 5 wt% of the composition)

Examples	IE1	IE2	IE3	CE1	CE2	CE3	CE4	CE5	CE6	CE7	CE8	CE9
Base polymer	Queo 7001	Queo 8201	Queo 0201	Queo 7001	Queo 8201	Queo 0201	LLDPE A	LLDPE B	LLDPE A	LLDPE B	LLDPE B	LLDPE B
Modifier	Excerec 40800	Excerec 40800	Excerec 40800	-	-	-	-	-	Excerec 40800	Excerec 40800	Licowax	Wax C
Tc (Rheo)	59	78	100	44	62	85	115	119	116	119	119	119
ΔTc	15	16	15	-	-	-	-	-	1	0	-	-
E' (23 °C)	40	98	244	16	36	96	790	712	510	854	751	405
ΔE'	24	62	148	-	-	-	-	-	-280	142	39	-307
E' ratio	2.5x	2.7x	2.6x	-	-	-	-	-	-	1.2x	1.1x	-
T soft	67.0	86.5	103.0	41.5	62.8	89.8	>125	>125	114.7	>125	>125	109.0
ΔT soft	25.5	23.7	13.2	-	-	-	-	-	<0	-	-	<0

The above results demonstrate that the IE1-IE3 have enhanced crystallisation rate and increase of the storage modulus E' . The increase in storage modulus is 2.7 x. Rheological measurements show that the addition of wax modifier has a significant effect on the solidification behaviour of the Queo polymers.

Figure 1a shows E' measured in dynamic temperature sweep for neat Queos and the blends. The transition occurs at higher temperature and becomes faster (steeper variation of E' with temperature) in the blends with respect to the neat Queo material.

Figure 1b shows the storage modulus E' reported as a function of the temperature for the Queo grades and their blends with the Excerec modifier. The addition of the modifier causes E' to increase by 2.5 to 2.8 times for blends with respect to the neat Queos (Table 2). This is due to the formed interpenetrating network structure, which remains stable up to the melting point of the lamellae.

T_{soft} also increases for the Queo blends (Table 3).

For blends in which the base polymer had density above 913 kg/m^3 (LLDPE A and LLDPE B), no variation of E' (23°C) was found for the LLDPE B/Licowax PE190 (CE8) and the LLDPE B/Excerec (CE7) blends.

For the LLDPE A/Excerec (CE6) and the LLDPE B/Wax C (CE9) blends, a lower E' (23°C) and T_{soft} was found with respect to the neat base polymers.

For blends with high density polyethylene, the temperature at which solidification occurs (T_c (Rheo)) is almost unaffected.

Film Formation

The blends were also employed as a film. The film was produced on a Collin 40 lab scale blown film line, with melt temperature of 195°C , BUR 1:2.5, thickness of $40\mu\text{m}$, and uptake speed of 7m/min , frostline distance of 120 mm .

The inventive example produces films with higher toughness, lower haze and lower seal initiation temperature. The compositions of the present invention therefore have a good balance of properties. See Table 4 below.

Table 4.

		IE3	CE7
LLDPE B	wt%		95
Queo 0201	wt%	95	
Excerex 40800	wt%	5	5
Tensile Modulus /MD	MPa	99	380
Tensile Modulus /TD	MPa	93	465
Dart Drop Index	g	1370	1275
Haze	%	8.97	59.22
Seal Initiation Temp	°C	82	118

Summary

- 5 • Queo materials with density up to 902 kg/m³ blended with Excerex show enhanced crystallisation rate, and an increase of at most 2.7 times was found for E'(@23 °C).
- In the Queo/Excerex blend, gelation or network formation could not be distinguished from the overall crystallisation process, due the obtained high
- 10 rate of transition from melt to solid state.
- For blends in which the base polymer had density above 913 kg/m³, no variation of E'(@23 °C) was found.

15 In the Queo blends, a lamellar network is formed during solidification, while for the blends with the high density base polymers a more homogeneous morphology with tightly packed lamellae was observed.

Claims

1. A polyolefin composition comprising
 - a) 70 to 99 wt% of a polyethylene polymer (A), wherein said
5 polyethylene polymer is a copolymer of ethylene and one or more
C₃-C₁₂ α -olefins, having an MFR (190 °C/2.16 kg, ISO 1133) of 0.1 to
5.0 g/10 min, a melting point T_m (DSC, ISO 11357/3) of less than
100 °C and a density (ASTM D792) of 865 to 905 kg/m³; and
 - b) 1 to 30 wt% of a high density polyethylene wax (B) having an MFR
10 (ISO1133, 190 °C/2.16 kg) of more than 300 g/10 min and/or a
weight average molecular weight of 1,000 to 35,000 (GPC) and a T_m
(DSC, ISO 11357/3) in the range of 120 to 135 °C.
2. A polyolefin composition as claimed in claim 1, wherein the polyethylene
15 polymer (A) is present in an amount of 75 to 98 wt%, preferably 85 to 97
wt% relative to the total weight of the composition as a whole; and/or
wherein wax is present in an amount of 2 to 25 wt%, preferably 3 to 15 wt%
relative to the total weight of the composition as a whole.
- 20 3. A polyolefin composition as claimed in any preceding claim, wherein the
polyethylene polymer (A) has a density in the range 865 to 904 kg/m³,
preferably 865 to 903 kg/m³.
- 25 4. A polyolefin composition as claimed in any preceding claim, wherein the
polyethylene polymer (A) has a melt flow rate (MFR) at 190 °C/2.16 kg of
0.5 to 2.0 g/10 min.
5. A polyolefin composition as claimed in any preceding claim, wherein the
polyethylene polymer (A) is a polyethylene plastomer.
30
6. A polyolefin composition as claimed in any preceding claim, wherein the
wax has a melt flow rate (MFR) at 190 °C/2.16 kg of at least 400 g/10 min,
preferably at least 450 g/10 min, more preferably at least 490 g/10 min
and/or a weight average molecular weight of 3000 to 25000.
35

7. A polyolefin composition as claimed in any preceding claim, wherein the wax is a homopolymer.
8. A polyolefin composition as claimed in any preceding claim, wherein the wax is prepared using a metallocene catalyst.
9. A polyolefin composition as claimed in any preceding claim, wherein the wax has a density of 950 to 990 kg/m³, such as at least 960 kg/m³.
10. A polyolefin composition as claimed in any preceding claim, wherein the polyethylene polymer (A) and the wax are the only polymer components in the composition.
11. A polyolefin composition as claimed in any preceding claim, wherein the storage modulus E'(23°C) is increased by 2.0 to 4.0 times for the polyolefin composition relative to the neat polymer (A); and/or
- wherein the solidification temperature (T_c (Rheo)) as determined by dynamic rheology is at least 12 °C higher for the polyolefin composition than that of the neat polymer (A) and/or
- wherein the softening temperature T_{soft} increases for the polyolefin composition by 10 to 30 °C relative to the neat polymer (A).
12. A polyolefin composition as claimed in any preceding claim, wherein the polyethylene polymer (A) has an ethylene content from 60 to 95 wt%, preferably from 65 to 90 wt% and more preferably from 70 to 88 wt%.
13. Use of a high density polyethylene wax (B) having an MFR (190 °C/2.16 kg) of more than 300 g/10 min and/or a weight average molecular weight of 1,000 to 35,000 and a T_m in the range of 120 to 135 °C to improve film processability of a polyethylene polymer (A), wherein said polyethylene polymer is a copolymer of ethylene and one or more C₃-C₁₂ α-olefins, having an MFR (190 °C/2.16 kg) of 0.1 to 5.0 g/10 min, a melting point T_m of less than 100 °C and a density of 865 to 905 kg/m³.

14. Use of a high density polyethylene wax (B) having an MFR (190 °C/2.16 kg) of more than 300 g/10 min and/or a weight average molecular weight of 1,000 to 35,000 and a T_m in the range of 120 to 135 °C to improve pelletisation of a polyethylene polymer (A), wherein said polyethylene
5 polymer is a copolymer of ethylene and one or more C₃-C₁₂ α-olefins, having an MFR (190 °C/2.16 kg) of 0.1 to 5.0 g/10 min, a melting point T_m of less than 100 °C and a density of 865 to 905 kg/m³.
15. Use of a polyolefin composition as defined in any of claims 1 to 12 as a hot
10 melt adhesive.
16. A film comprising a polyolefin composition as defined in any of claims 1 to 12.
- 15 17. A substrate which is at least partially coated with a hot melt adhesive, said adhesive comprising a polyolefin composition as defined in any of claims 1 to 12.