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(54) Title: AUTOMOTIVE COMPOUNDS FEATURING LOW SURFACE TACK

(57) Abstract: The present invention relates to a polymer composition comprising a heterophasic propylene copolymer, a mineral filler and a light stabilizers comprising a fatty acid derivative and a benzoate derivative. The polymer composition according to the present invention is applicable for automotive articles whereupon the undesired effect of stickiness is reduced significantly compared to automotive articles comprising conventional light stabilizers.



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Automotive compounds featuring low surface tack

The present invention is related to a polymer composition comprising a heterophasic propylene copolymer (HECO) and a specific combination of UV-absorbers. Furthermore the present invention relates to the use of a composition as well as a combination of UV-absorbers in a polymer composition to improve surface tack of the same.

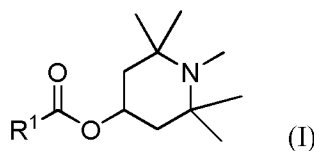
Automotive interior parts like dashboards, door claddings, trims etc. are commonly made from polymers. For such parts it is often tried to mimic a leather- or fabric-like surface and touch in order to give occupants a high-quality impression of the car. As a result, haptic and surface characteristics are important properties regarding automotive interior parts. One factor in this respect is the surface tack of a part.

Regarding the UV-absorber, it was unfortunately found that the application of the frequently used combinations of hindered amine light stabilizers (HALS) “Tinuvin770/Chimasorb119” or “Chimasorb119/Chimasorb944” which are known to the skilled person in fact lead to a tacky feel of the surface.

Therefore, the objection of the present invention is the finding of polymer composition comprising a new UV-absorber system which leads to an improved surface tack behavior.

Accordingly, the present invention is related to a polymer composition (C), comprising

- (a) a heterophasic propylene copolymer (HECO),
- (b) a mineral filler (F),
- (c) a 1,2,2,6,6-pentamethyl-4-piperidylester derivative of formula (I),

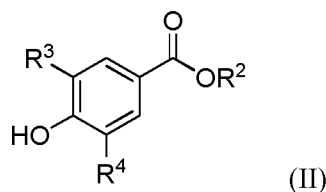


wherein

the molecular weight is below 1000 g/mol and R¹ is a linear or branched C₅ to C₂₅ alkyl residue, and

- (d) a benzoate derivative of formula (II),

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wherein

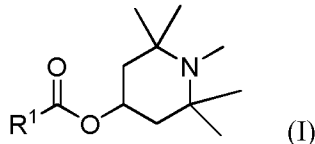
R^2 is a linear or branched C_5 to C_{25} alkyl residue and

R^3 and R^4 are independently from each other methyl, ethyl, linear and/or branched C_3 to C_{12} alkyl residues.

The present invention also relates to an automotive article comprising the above polymer composition (C).

10 The present invention also relates to the use of a combination of

(a) a 1,2,2,6,6-pentamethyl-4-piperidylester derivative of formula (I)

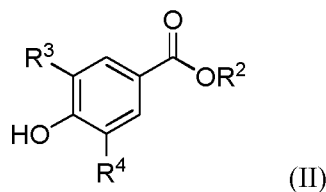


wherein

the molecular weight is below 1000 g/mol and R^1 is a linear or branched C_5 to C_{25} alkyl residue, and

and

(b) a benzoate derivative of formula (II)



wherein

R^2 is a linear or branched C_5 to C_{25} alkyl residue and

R^3 and R^4 are independently from each other methyl, ethyl, linear and/or branched C_3 to C_{12} alkyl residues,

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in a polymer composition (C) comprising further a heterophasic propylene copolymer (HECO) and a mineral filler (F)

to reduce the effect of stickiness of the polymer composition (C), like of the extruded polymer composition (C), and/or of the automotive article comprising said polymer

5 composition (C).

The present invention further relates to a process for the preparation of the above polymer composition (C) by extruding the heterophasic propylene copolymer (HECO), the mineral filler (F), the 1,2,2,6,6-pentamethyl-4-piperidylester derivative of formula (I) and the

10 benzoate derivative of formula (II) in an extruder.

A further aspect of the present invention is the use of the above polymer composition (C) in an automotive application.

15 Further preferred embodiments of the present invention are described in the appended claims.

The polymer composition (C) of the present invention comprises in a preferred embodiment

(a) the heterophasic propylene copolymer (HECO) in an amount of 30 to 80 wt.-%, more preferably 40 to 75 wt.-%, still more preferably 50 to 70 wt.-%,

(b) the mineral filler (F) in an amount no higher than 40 wt.-%, more preferably no higher than 30 wt.-%, still more preferably no higher than 20 wt.-%,

(c) the 1,2,2,6,6-pentamethyl-4-piperidylester derivative of formula (I) and the benzoate derivative of formula (II) combined in an amount of 0.01 to 5.0 wt.-%, more preferably 0.03 to 3.0 wt.-%, still more preferably 0.04 to 0.5 wt.-%, yet more preferably 0.05 to 0.5 wt.-%, like 0.08 to 0.5 wt.-%,

25 based on the total composition.

In a further preferred embodiment, the polymer composition further comprises a polypropylene (PP).

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In another preferred embodiment, the polymer composition further comprises a high density polyethylene (HDPE).

It is apparent from the wording used for the different polymers (HECO, PP and HDPE)
5 according to the present invention that they must (chemically) differ from each other.

Accordingly in one embodiment the polymer composition (C) of the present invention comprises in a preferred embodiment

- 10 (a) the heterophasic propylene copolymer (HECO) in an amount of 30 to 80 wt.-%, more preferably 40 to 75 wt.-%, still more preferably 50 to 70 wt.-%,
- (b) the mineral filler (F) in an amount of 5 to 30 wt.-%, more preferably 10 to 20 wt.-%,
- (c) optionally a polypropylene (PP) in an amount of 5 to 20 wt.-%, more preferably 5 to 15 wt.-%,
- (d) optionally the high density polyethylene (HDPE) in an amount 5 to 20 wt.-%, more
15 preferably 8 to 15 wt.-%, and
- (e) the 1,2,2,6,6-pentamethyl-4-piperidylester derivative of formula (I) and the benzoate derivative of formula (II) combined in an amount of 0.01 to 5.0 wt.-%, more preferably 0.03 to 3.0 wt.-%, still more preferably 0.04 to 0.5 wt.-%, yet more preferably 0.05 to 0.5 wt.-%, like 0.08 to 0.5 wt.-%,
20 based on the total composition.

The expression "heterophasic" indicates that an elastomeric copolymer is (finely) dispersed in a matrix. In other words the elastomeric copolymer forms inclusions in the matrix. Thus the matrix contains (finely) dispersed inclusions being not part of the matrix and said
25 inclusions contain the elastomeric copolymer. The term "inclusion" according to this invention shall preferably indicate that the matrix and the inclusion form different phases within the heterophasic propylene copolymer, said inclusions are for instance visible by high resolution microscopy, like electron microscopy or scanning force microscopy. In case the composition comprises the polypropylene (PP) and/or the high density polyethylene
30 (HDPE), the final composition is probably of a complex structure. Probably the matrix of the heterophasic propylene copolymer (HECO) forms a continuous phase with the

- 5 -

polypropylene (PP), and optionally the high density polyethylene (HDPE) forms individually inclusions dispersed therein.

Additionally the inclusions of the final composition may also contain the mineral filler (F);
5 however preferably the mineral filler (F) forms separate inclusions within the matrix.

The reduction of stickiness is preferably accomplished in case the stickiness index, especially in terms of the surface tack (QT) as determined in the example section, is below a value of 0.75, preferably below a value of 0.72, still more preferably below a value of 0.70,
10 like in the range of 0.50 to 0.72 or in the range of 0.50 to 0.70.

The polymer composition according to the present invention may be prepared by compounding the components within suitable melt mixing devices for preparing polymeric compounds, including in particular extruders, like single screw extruders as well as twin
15 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.

20 In the following the individual components are defined in more detail.

Heterophasic propylene copolymer (HECO)

One essential component of the present invention is the presence of a heterophasic propylene
25 copolymer (HECO). As already defined above, a heterophasic system comprises a polypropylene matrix and an amorphous elastomer, i.e. the elastomeric propylene copolymer (E), dispersed therein. Such polymer compositions are well known in the art and commercially available. This applies especially for the heterophasic propylene copolymer (HECO) as defined in the instant invention.

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The polypropylene matrix (M) of the heterophasic propylene copolymer (HECO) can be a propylene homopolymer or a propylene copolymer with comonomers selected from ethylene and/or C₄ to C₁₂ α -olefins. Preferably, the polypropylene matrix (M) of the heterophasic propylene copolymer (HECO) is a propylene homopolymer.

5

The xylene cold insoluble (XCI) fraction of the heterophasic propylene copolymer (HECO) is dominated by the polypropylene matrix (M), whereas the main component of the xylene cold soluble fraction is the elastomeric propylene copolymer (E). Accordingly on the one hand the properties of the xylene cold insoluble (XCI) fraction and the polypropylene matrix (M) and on the other hand the properties of the xylene cold soluble (XCS) fraction and the elastomeric propylene copolymer (E) are essentially the same.

10

The expression propylene homopolymer used in the instant invention relates to a polypropylene that consists substantially, i.e. of more than 99.7 wt.-%, still more preferably of at least 99.8 wt.-%, of propylene units. In a preferred embodiment only propylene units in the propylene homopolymer are detectable.

15

Accordingly the comonomer content of the polypropylene matrix (M) and/or of the xylene cold insoluble (XCI) fraction is preferably equal or below 1.0 wt.-%, more preferably not more than 0.8 wt.-%, still more preferably not more than 0.5 wt.-%, like not more than 0.2 wt.-%, e.g. not detectable.

20

Preferably the polypropylene matrix (M) and/or of the xylene cold insoluble (XCI) fraction of the heterophasic propylene copolymer (HECO) has a melt flow rate MFR₂ (230 °C) in the range of 30 to 90 g/10 min, more preferably in the range of 40 to 70 g/10 min, still more preferably in the range of 45 to 60 g/10 min.

25

As mentioned above, in addition to the polypropylene matrix (M), the heterophasic propylene copolymer (HECO) comprises an elastomeric propylene copolymer (E) which is dispersed within said polypropylene matrix (M).

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According to one embodiment, the elastomeric propylene copolymer (E) comprises monomers copolymerizable with propylene, for example, comonomers such as ethylene and/or C₄ to C₁₂ α -olefins, e.g. 1-butene and/or 1-hexene. Preferably the elastomeric propylene copolymer (E) comprises, especially consists of, monomers copolymerizable with propylene selected 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) phase comprises units derivable from ethylene and propylene only.

10

In case the polypropylene matrix (M) of the heterophasic propylene copolymer (HECO) is a propylene copolymer, it is preferred that the comonomer(s) of the propylene copolymer and the elastomeric propylene copolymer (E) are the same.

15 In a preferred embodiment, the elastomeric propylene copolymer (E) and/or the xylene cold soluble (XCS) fraction of the heterophasic propylene copolymer (HECO) has a comonomer content in the range of 10 to 50 wt.-%, more preferably 20 to 45 wt.-%, still more preferably 30 to 42 wt.-%.

20 Additionally or alternatively to the comonomer content it is preferred that the elastomeric propylene copolymer (E) and/or the xylene cold soluble (XCS) fraction of the heterophasic propylene copolymer (HECO) has an intrinsic viscosity (IV) in the range of 1.0 to 8.0 dl/g, more preferably in the range of 1.5 to 6.0 dl/g, still more preferably in the range of 2.0 to 3.5 dl/g.

25

According to one embodiment of the present invention, the amount of the elastomeric propylene copolymer (E) and/or of the xylene cold soluble (XCS) fraction of the heterophasic propylene copolymer (HECO) is in the range of 10 to 50 wt.-%, more preferably 15 to 40 wt.-%, still more preferably 20 to 35 wt.-%, based on the total amount of the heterophasic propylene copolymer (HECO).

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The comonomer content of the heterophasic propylene copolymer (HECO) is preferably in the range of 3.0 to 25 wt.-%, more preferably in the range of 5.0 to 20 wt.-%, still more preferably in the range of 10 to 18 wt.-%, based on the total amount of the heterophasic propylene copolymer (HECO).

5

Preferably the heterophasic propylene copolymer (HECO) has a melt flow rate MFR_2 (230 °C) in the range of 1.0 to 50 g/10 min, more preferably 2.0 to 30 g/10 min, still more preferably 5.0 to 20 g/10 min.

10 In a preferred embodiment, the heterophasic propylene copolymer (HECO) has a flexural modulus in the range of 600 to 950 MPa, more preferably 650 to 900 MPa, still more preferably 700 to 850 MPa, even more preferably in the range of 700 to 800 MPa.

Especially preferred heterophasic propylene copolymer (HECO) is the commercial product
15 of the EF-series of Borealis AG.

Polypropylene (PP)

The polymer composition (C) according to the present invention optionally comprises a
20 polypropylene (PP).

The polypropylene (PP) can be a propylene homopolymer or a propylene copolymer with comonomers selected from ethylene and/or C_4 to C_{12} α -olefins. Preferably, the polypropylene (PP) is a propylene homopolymer according to the definition described above.

25

The polypropylene (PP) preferably has a melt flow rate MFR_2 (230 °C) in the range of 3 to 50 g/10 min, preferably in the range of 5 to 35 g/10 min, still more preferably in the range of 10 to 25 g/10 min.

30 Preferably the polypropylene (PP) has a density of at least 890 kg/m³, more preferably at least 900 kg/m³, still more preferably in the range of 900 to 912 kg/m³.

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In one embodiment the polypropylene (PP) is the commercial product HF955MO of Borealis AG.

5 **High density polyethylene (HDPE)**

The polymer composition (C) according to the present invention optionally comprises a high density polyethylene (HDPE).

- 10 The high density polyethylene (HDPE) preferably has a melt flow rate MFR_2 (190 °C) in the range of 2 to 30 g/10 min, more preferably in the range of 3 to 20 g/10 min, still more preferably in the range of 5 to 15 g/10 min.

- 15 Preferably, the high density polyethylene (HDPE) has a density of at least 930 kg/m^3 , more preferably in the range of 930 to 980 kg/m^3 , still more preferably in the range of 940 to 970 kg/m^3 .

In one embodiment the high density polyethylene (HDPE) is the commercial product MG9641 of Borealis AG.

20

Mineral filler (F)

- 25 In addition to the polymer components the polymer composition (C) according to the present invention comprises a mineral filler, preferably in amounts of up to 40 wt.-%, preferably up to 30 wt.-%, more preferably up to 20 wt.-%, yet more preferably in the range of 5.0 to 40 wt.-%, still more preferably in the range of 5.0 to 30 wt.-%, like in the range of 10.0 to 20 wt.-%.

- 30 Preferably the mineral filler (F) is a phyllosilicate, mica or wollastonite. Even more preferably the mineral filler (F) is selected from the group of mica, wollastonite, kaolinite, smectite, montmorillonite and talc. The most preferred mineral filler (F) is talc.

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The mineral filler (F) preferably has a specific surface area (BET) in the range of 5 to 25 m²/g, more preferably in the range of 8 to 20 m²/g, still more preferably in the range of 12 to 16 m²/g.

5

Light stabilizers

Essential finding of the present invention is that the instant polymer composition must comprise two specifically selected light stabilizers,

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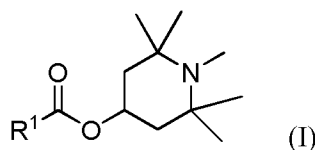
Light stabilizers are chemical compounds capable of interfering with the physical and chemical processes of light-induced degradation.

The specific finding of the present invention is that the polymer composition (C) must comprise two different light stabilizers, one belonging to the class of UV-absorbers, like 4-hydroxybenzoates, and one belonging to the class of free radical scavengers, like hindered amine light stabilizers (HALS).

Hindered amine light stabilizers (HALS) are a class of free radical scavengers known to the skilled person. A hindered amine light stabilizer (HALS) contains one or more sterically hindered amine functionalities. Typically, 2,2,6,6-tetramethyl piperidine derivatives are frequently applied as hindered amine light stabilizers (HALS).

Accordingly the instant polymer composition (C) must comprise

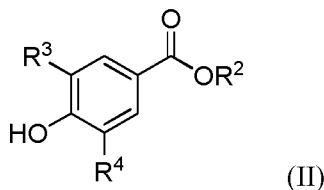
25 (a) a 1,2,2,6,6-pentamethyl-4-piperidylester derivative of formula (I),



wherein the molecular weight is below 1000 g/mol and R¹ is a linear or branched C₅ to C₂₅ alkyl residue, and

(b) a benzoate derivative of formula (II),

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wherein R² is a linear or branched C₅ to C₂₅ alkyl residue and R³ and R⁴ are independently from each other Methyl, Ethyl, linear and/or branched C₃ to C₁₂ alkyl residues.

- 5 Preferably the combination of the 1,2,2,6,6-pentamethyl-4-piperidylester derivative of formula (I) and the benzoate derivative of formula (II) is the only light stabilizer combination within the polymer composition (C).

Thus according to the present invention, one component of the light stabilizer is a 1,2,2,6,6-
 10 pentamethyl-4-piperidylester of a fatty acid according to formula (I), whereupon R¹ is a linear or branched C₅ to C₂₅ alkyl residue, more preferably C₁₁ to C₁₈. In a preferred embodiment, R¹ is a linear C₁₆ to C₁₈ alkyl residue.

The other component of the light stabilizers according to the present invention is the
 15 benzoate derivative of formula (II), whereupon R² is a linear or branched C₅ to C₂₅ alkyl residue, like a linear C₅ to C₂₅ alkyl residue, preferably C₁₀ to C₂₀ alkyl residue, like a linear C₁₀ to C₂₀ alkyl residue, more preferably C₁₂ to C₁₈ alkyl residue, like a linear C₁₂ to C₁₈ alkyl residue. In a preferred embodiment, R² is a linear C₁₆ alkyl residue. R³ and R⁴ of the benzoate derivative of formula (II) are independently from each other methyl, ethyl, linear and/or
 20 branched C₃ to C₁₂ alkyl residues. Preferably, R³ and R⁴ contain no more than 4 carbon atoms, respectively. Thus, in a preferred embodiment R³ and R⁴ are independently selected from the group consisting of methyl, ethyl, *n*-propyl, *iso*-propyl, *n*-butyl, *sec*.-butyl and/or *tert*.-butyl. More preferably, R³ and R⁴ are *tert*.-butyl residues. Thus, in an especially preferred embodiment, the benzoate derivative of formula (II) is *n*-hexadecyl-3,5-di-*tert*.-
 25 butyl-4-hydroxybenzoate.

Preferably the weight ratio between the UV-absorber, like the 4-hydroxybenzoate, e.g. the benzoate derivative of formula (II), and the free radical scavenger, like hindered amine light

stabilizer (HALS), e.g. the 1,2,2,6,6-pentamethyl-4-piperidylester derivative of formula (I), is in the range of 70:30 to 30:70, more preferably in the range of 60:40 to 40:60, still more preferably in the range of 55:45 to 45:55, like 50:50.

- 5 As mentioned above, typically the amount of the two light stabilizers together in the polymer composition (C), e.g. the total amount of the 1,2,2,6,6-pentamethyl-4-piperidylester derivative of formula (I) and the benzoate derivative of formula (II) together in the polymer composition (C), is in the range of 0.01 to 5.0 wt.-%, more preferably 0.05 to 3.0 wt.-%, still more preferably 0.1 to 1.0 wt.-%, based on the total amount of the polymer composition (C).

10

Further components

- The instant polymer composition (C) may comprise additional typical additives different to those mentioned above, i.e. different to the light stabilizers and the mineral filler (F). Such
15 typical additives are antioxidants and slip agents and pigments. Preferably the amount of additives excluding the mineral filler (F) and the light stabilizers shall not exceed 8 wt.-%, more preferably 5 wt.-%, still more preferably 3 wt.-% within on the total composition.

- All components used for the preparation of the instant composition are known. Accordingly
20 also their preparation is well known. For instance the heterophasic propylene copolymer (HECO) according to this invention is preferably produced in a multistage process known in the art, wherein the matrix is produced at least in one slurry reactor and subsequently the elastomeric copolymer is produced at least in one gas phase reactor.

- 25 Thus, the polymerization system can comprise one or more conventional stirred slurry reactors and/or one or more gas phase reactors. Preferably the reactors used are selected from the group of loop and gas phase reactors and, in particular, the process employs at least one loop reactor and at least one gas phase reactor. It is also possible to use several reactors of each type, e.g. one loop and two or three gas phase reactors, or two loops and one or two gas
30 phase reactors, in series.

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Preferably the process comprises also a prepolymerisation with the chosen catalyst system, as described for instance in detail below comprising (i) a Ziegler-Natta procatalyst, (ii) a cocatalyst and (iii) an external donor.

- 5 In a preferred embodiment, the prepolymerisation 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 prepolymerisation reaction is typically conducted at a temperature of 0 to 50 °C,
10 preferably from 10 to 45 °C, and more preferably from 15 to 40 °C.

The pressure in the prepolymerisation 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.

15

The catalyst components are preferably all introduced to the prepolymerisation 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 prepolymerisation stage and the remaining part into subsequent polymerization stages. Also in such cases it is
20 necessary to introduce so much cocatalyst into the prepolymerisation 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
25 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.

30

A slurry reactor designates any reactor, such as a continuous or simple batch stirred tank reactor or loop reactor, operating in bulk or slurry and in which the polymer forms in particulate form. "Bulk" means a polymerization in reaction medium that comprises at least 60 wt.-% monomer. According to a preferred embodiment the slurry reactor comprises a
5 bulk loop reactor.

"Gas phase reactor" means any mechanically mixed or fluid bed reactor. Preferably the gas phase reactor comprises a mechanically agitated fluid bed reactor with gas velocities of at least 0.2 m/sec.

10

The particularly preferred embodiment for the preparation of the heterophasic propylene copolymer (HECO) of the invention comprises carrying out the polymerization in a process comprising either a combination of one loop and one or two gas phase reactors or a combination of two loops and one or two gas phase reactors.

15

A preferred multistage process is a slurry-gas phase process, such as developed by Borealis and known as the Borstar[®] technology. In this respect, reference is made to EP 0 887 379 A1, WO 92/12182, WO 2004/000899, WO 2004/111095, WO 99/24478, WO 99/24479 and WO 00/68315. They are incorporated herein by reference.

20

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

Preferably the heterophasic propylene copolymer (HECO) is produced by using a special Ziegler-Natta procatalyst in combination with a special external donor, as described below in
25 detail, preferably in the Spheripol[®] or in the Borstar[®]-PP process.

One preferred multistage process may therefore comprise the steps of:

- producing a polypropylene matrix in the presence of the chosen catalyst system, as for instance described in detail below, comprising the special Ziegler-Natta
30 procatalyst (i), an external donor (iii) and the cocatalyst (ii) in a first slurry reactor

- 15 -

and optionally in a second slurry reactor, both slurry reactors using the same polymerization conditions,

- transferring the slurry reactor product into at least one first gas phase reactor, like one gas phase reactor or a first and a second gas phase reactor connected in series,
- 5 - producing an elastomeric copolymer in the presence of the polypropylene matrix and in the presence of the catalyst system in said at least first gas phase reactor,
- recovering the polymer product for further processing.

With respect to the above-mentioned preferred slurry-gas phase process, the following
10 general information can be provided with respect to the process conditions.

Temperature is preferably from 40 to 110 °C, preferably between 50 and 100 °C, in particular between 60 and 90 °C, with a pressure in the range of from 20 to 80 bar, preferably 30 to 60 bar, with the option of adding hydrogen in order to control the molecular
15 weight in a manner known per se.

The reaction product of the slurry polymerization, which preferably is carried out in a loop reactor, is then transferred to the subsequent gas phase reactor(s), wherein the temperature preferably is within the range of from 50 to 130 °C, more preferably 60 to 100 °C, at a
20 pressure in the range of from 5 to 50 bar, preferably 8 to 35 bar, again with the option of adding hydrogen in order to control the molecular weight in a manner known per se.

The average residence time can vary in the reactor zones identified above. In one embodiment, the average residence time in the slurry reactor, for example a loop reactor, is
25 in the range of from 0.5 to 5 hours, for example 0.5 to 2 hours, while the average residence time in the gas phase reactor generally will be from 1 to 8 hours.

If desired, the polymerization may be effected in a known manner under supercritical conditions in the slurry, preferably loop reactor, and/or as a condensed mode in the gas phase
30 reactor.

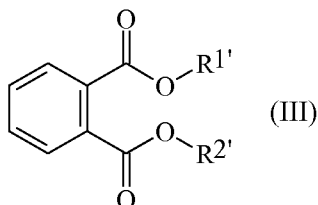
- 16 -

According to the invention the heterophasic propylene copolymer (HECO) is preferably obtained by a multistage polymerization process, as described above, in the presence of a catalyst system comprising as component (i) a Ziegler-Natta procatalyst which contains a trans-esterification product of a lower alcohol and a phthalic ester.

5

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 (III)



10

wherein $\text{R}^{1'}$ and $\text{R}^{2'}$ 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 (III) takes place to form the internal donor

- c) washing the product of stage b) or
- d) optionally reacting the product of step c) with additional TiCl_4 .

15

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.

20

First an adduct of MgCl_2 and a C_1 - C_2 alcohol of the formula $\text{MgCl}_2 \cdot n\text{ROH}$, wherein R is methyl or ethyl and n is 1 to 6, is formed. Ethanol is preferably used as alcohol.

25

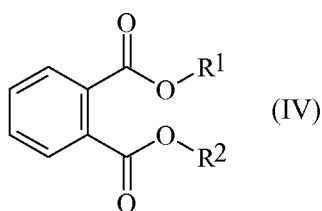
The adduct, which is first melted and then spray crystallized or emulsion solidified, is used as catalyst carrier.

- 17 -

In the next step the spray crystallized or emulsion solidified adduct of the formula $\text{MgCl}_2 \cdot n\text{ROH}$, wherein R is methyl or ethyl, preferably ethyl and n is 1 to 6, is contacting with TiCl_4 to form a titanised carrier, followed by the steps of

- adding to said titanised carrier
- 5 (i) a dialkylphthalate of formula (III) with $\text{R}^{1'}$ and $\text{R}^{2'}$ being independently at least a C_5 -alkyl, like at least a C_8 -alkyl, or preferably
- (ii) a dialkylphthalate of formula (III) with $\text{R}^{1'}$ and $\text{R}^{2'}$ being the same and being at least a C_5 -alkyl, like at least a C_8 -alkyl,
- 10 or more preferably
- (iii) a dialkylphthalate of formula (III) selected from the group consisting of propylhexylphthalate (PrHP), dioctylphthalate (DOP), di-iso-decylphthalate (DIDP), and ditiidecylphthalate (DTDP), yet more preferably the dialkylphthalate of formula (I) is a dioctylphthalate (DOP),
- 15 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

20 between 130 to 150°C , such that said methanol or ethanol is transesterified with said ester groups of said dialkylphthalate of formula (III) to form preferably at least 80 mol-%, more preferably 90 mol-%, most preferably 95 mol.-%, of a dialkylphthalate of formula (IV)



- 25 with R^1 and R^2 being methyl or ethyl, preferably ethyl, the dialkylphthalat of formula (IV) being the internal donor and

- 18 -

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

5 The adduct of the formula $\text{MgCl}_2 \cdot n\text{ROH}$, 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.

10 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.

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

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

20

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.-%.

25

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 (III), yielding diethyl phthalate (DEP) as the internal donor compound.

- 19 -

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 (III).

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



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

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

- 20 For the production of the heterophasic propylene copolymer (HECO), 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
 25 trialkylaluminium, like triethylaluminium (TEA), dialkyl aluminium chloride and alkyl aluminium sesquichloride.

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

- 30 $\text{Si}(\text{OCH}_3)_2\text{R}_2^5$ (Va)

- 20 -

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

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

Formula (Vb) is defined by



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

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

- 25 More preferably the external donor of formula (Vb) is diethylaminotriethoxysilane .

Most preferably the external donor is of formula (Va), like dicyclopentyl dimethoxy silane $[\text{Si}(\text{OCH}_3)_2(\text{cyclo-pentyl})_2]$ or diisopropyl dimethoxy silane $[\text{Si}(\text{OCH}_3)_2(\text{CH}(\text{CH}_3)_2)_2]$.

- 30 For mixing the individual components of the instant composition, a conventional compounding or blending apparatus, e.g. a Banbury mixer, a 2-roll rubber mill, Buss-co-

kneader or a twin screw extruder may be used. The polymer materials recovered from the extruder are usually in the form of pellets. These pellets are then preferably further processed, e.g. by injection moulding to generate articles and products of the inventive composition.

5

Accordingly the present invention is also directed to a process for the preparation of the instant composition (C) comprising the steps of adding the polymer components, i.e. the heterophasic propylene copolymer (HECO) and optionally the high density polyethylene (HDPE) and/or the polypropylene (PP), the mineral filler (F) and the composition of the

10 1,2,2,6,6-pentamethyl-4-piperidylester derivative of formula (I) and the benzoate derivative of formula (II) to an extruder (as mentioned above) and extruding the same obtaining thereby said polypropylene composition (C).

The polypropylene composition according to the invention may be pelletized and

15 compounded using any of the variety of compounding and blending methods well known and commonly used in the resin compounding art.

Articles made from the polymer composition (C)

20 The current invention also provides (automotive) articles, like injection molded articles, comprising at least to 60 wt.-%, more preferably at least 80 wt.-%, yet more preferably at least 95 wt.-%, like consisting, of the inventive polymer composition (C). Accordingly the present invention is especially directed to automotive articles, especially to car interiors and exteriors, like bumpers, side trims, step assists, body panels, spoilers, dashboards, interior

25 trims and the like, comprising at least to 60 wt.-%, more preferably at least 80 wt.-%, yet more preferably at least 95 wt.-%, like consisting, of the inventive polymer composition (C).

Uses according to the invention

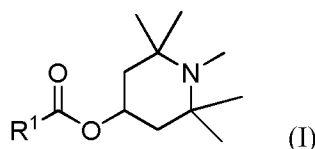
30 The polymer composition (C) of the present invention is preferably used for automotive articles, like moulded automotive articles, preferably automotive injection moulded articles.

- 22 -

Even more preferred is the use of the polymer composition (C) for car interiors and exteriors, like bumpers, side trims, step assists, body panels, spoilers, dashboards, interior trims and the like.

- 5 The present invention is especially directed to the use of a 1,2,2,6,6-pentamethyl-4-piperidylester derivative of formula (I) and a benzoate derivative of formula (II) in a polymer composition (C), like in an extruded polymer composition (C), comprising a heterophasic propylene copolymer (HECO) and a mineral filler (F) to reduce the effect of stickiness.

- 10 The present invention is especially directed to the use of a combination of
(a) a 1,2,2,6,6-pentamethyl-4-piperidylester derivative of formula (I)



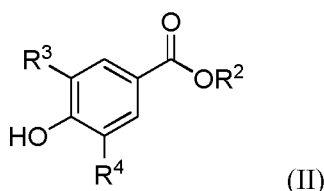
wherein

the molecular weight is below 1000 g/mol and R¹ is a linear or branched C₅ to C₂₅ alkyl residue, and

15

and

- (b) a benzoate derivative of formula (II)



wherein

20

R² is a linear or branched C₅ to C₂₅ alkyl residue and

R³ and R⁴ are independently from each other methyl, ethyl, linear and/or branched C₃ to C₁₂ alkyl residues,

in a polymer composition (C) comprising further a heterophasic propylene copolymer (HECO) and a mineral filler (F)

25

to reduce the effect of stickiness of the polymer composition (C), like the extruded polymer composition (C), and/or of the automotive article comprising said polymer composition (C).

Preferably, the of the reduction of stickiness is accomplished in case the stickiness index, especially in terms of surface tack (QT) as determined in the example section, is below a value of 0.75, preferably below a value of 0.72, still more preferably below a value of 0.70, like in the range of 0.50 to 0.72 or 0.50 to 0.70.

Concerning the preferred embodiments of the polymer composition (C) and its components, especially with regard to 1,2,2,6,6-pentamethyl-4-piperidylester derivative of formula (I) and the benzoate derivative of formula (II), reference is made to the information provided above.

10

Thus for instance the use of a 1,2,2,6,6-pentamethyl-4-piperidylester derivative of formula (I) and a benzoate derivative of formula (II) in a polymer composition (C) comprising

- (a) the heterophasic propylene copolymer (HECO) in an amount of 30 to 80 wt.-%, more preferably 40 to 75 wt.-%, still more preferably 50 to 70 wt.-%,
- 15 (b) the mineral filler (F) in an amount no higher than 40 wt.-%, more preferably no higher than 30 wt.-%, still more preferably no higher than 20 wt.-%,
- (c) the 1,2,2,6,6-pentamethyl-4-piperidylester derivative of formula (I) and the benzoate derivative of formula (II) combined in an amount of 0.01 to 5.0 wt.-%, more preferably 0.03 to 3.0 wt.-%, still more preferably 0.04 to 0.5 wt.-%, yet more
- 20 preferably 0.05 to 0.5 wt.-%, like 0.08 to 0.5 wt.-%,

based on the total composition,

reduces the effect of stickiness of the polymer composition (C), like the extruded polymer composition (C), and/or of the automotive article comprising said polymer composition (C).

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

E X A M P L E S

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.

5 **1. Definitions/Measuring Methods**

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

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

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

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

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.

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

Quantification of comonomer content by FTIR spectroscopy

The comonomer content is determined by quantitative Fourier transform infrared spectroscopy (FTIR) after basic assignment calibrated via quantitative ¹³C nuclear magnetic resonance (NMR) spectroscopy in a manner well known in the art. Thin films are pressed to a thickness of between 100-500 µm and spectra recorded in transmission mode.

Specifically, the ethylene content of a polypropylene-co-ethylene copolymer is determined using the baseline corrected peak area of the quantitative bands found at 720-722 and 730-733 cm⁻¹. Specifically, the butene or hexene content of a polyethylene copolymer is
25 determined using the baseline corrected peak area of the quantitative bands found at 1377-1379 cm⁻¹. Quantitative results are obtained based upon reference to the film thickness.

Flexural Modulus: The flexural modulus was determined in 3-point-bending at 23°C according to ISO 178 on 80x10x4 mm³ test bars injection moulded in line with EN ISO 1873-2

30 **Specific surface area** is determined as the BET surface according to DIN 66131/2 (N₂).

Surface tack (Stickiness index)

The surface tack measurement was performed on injection moulded multigrain plaques with a film gate using the grain K29. The K29 surface is defined as a grain with a grain depth of 0.01 mm, and a minimum draft angle of 1° for ejection from the injection moulding tool. The section with this grain was cut out from the plaque with a bench shear. The sample size was 60x60 mm. Before evaluation of the stickiness index (in term of the tack quotient Q_T) the samples were stored under following conditions:

black-panel-temperature	80 +/- 5 °C
chamber temperature	45 +/-2 °C
rel. Humidity	20 +/-10 %
radiation intensity	40 W/m ²
Filter system	Borosilikat/Sodalime
Period of storage	500h

To perform the experiments an Instron tensile testing machine was used (ElectroPuls E3000, Instron Deutschland GmbH, Germany) with an elastomer die tip having a diameter of 25 mm and a thickness of 5 mm. The compressive force F was -50 N, the holding time t_H was 91 s, and the haul-off speed v was 55 mm/s. The tests were performed at standard laboratory climate conditions (23°C and 50% relative humidity). Each single surface tack measurement was performed with this setup in the following way: After the elastomer (NR/SBR) tip had been cleaned and attached to the die by means of a double-side adhesive tape, the tackiness force F_T of both the aluminium reference and the specimen (sample) were measured. The surface tack (stickiness index) is reported as tack quotient Q_T , which was calculated by the following Equation:

$$Q_T = \frac{F_{T, \text{sample}}}{F_{T, \text{reference}}}$$

and averaged over a minimum of three successive measurements.

A detailed description of the test method for quantifying surface tack can also be found in the paper “A novel test method for quantifying surface tack of polypropylene compound surfaces” by Çakmak et al., which will be published in eXPRESS Polymer Letters in 2011.

2. Examples

Table 1: Properties of the heterophasic propylene copolymer (HECO)

		HECO
MFR	[g/10min]	10.5
MFR of XCI	[g/10min]	50
XCS	[wt%]	29.0
C2 total	[wt%]	15.5
C2 in XCS	[wt%]	39.0
IV of XCS	[dl/g]	3.0
Flexural Modulus	[MPa]	770

Table 2: Inventive and Comparative Examples

Example*		CE 1	CE 2	IE
HECO	[wt%]	62.35	62.35	62.45
Talc	[wt%]	16.0	16.0	16.0
UV1a	[wt%]	-	-	0.05
UV1b	[wt%]	-	-	0.05
UV2	[wt%]	-	0.1	
UV3	[wt%]	0.1	-	-
UV4	[wt%]	0.1	0.1	-
HDPE	[wt%]	10.0	10.0	10.0
PP	[wt%]	9.0	9.0	9.0
Stickiness index	[-]	0.79	0.85	0.62

5 Rest to 100 wt.-% are additives, like antioxidants and pigments

“HECO” is the commercial product EE013AE of Borealis AG.

“Talc” is the commercial talc Jetfine 3CA of Imerys Talc Austria GmbH, having a specific surface area (BET) of 14.5 m²/g.

10 “UV1a” is the commercial product CYASORB UV-2908 of Cytec.

“UV1b” is the commercial product CYASORB UV-3853 of Cytec.

“UV2” is the commercial product TINUVIN770 of BASF.

“UV3” is the commercial product CHIMASORB 944 of BASF.

“UV4” is the commercial product CHIMASORB 119FL of BASF.

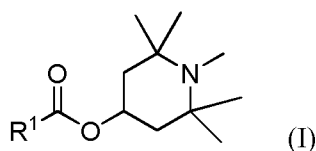
15 “HDPE” is the commercial product MG9641 of Borealis AG having a MFR₂ (190 °C) of 8 g/10 min and a density of 964 kg/m³.

“PP” is the commercial product HF955MO of Borealis AG having a MFR₂ (230 °C) of 20 g/10 min and a density of 908 kg/m³.

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CLAIMS

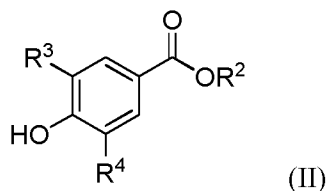
1. Polymer Composition (C), comprising
- 5 (a) a heterophasic propylene copolymer (HECO),
- (b) a mineral filler (F),
- (c) a 1,2,2,6,6-pentamethyl-4-piperidylester derivative of formula (I),



wherein

- 10 the molecular weight is below 1000 g/mol and R¹ is a linear or branched C₅ to C₂₅ alkyl residue, and

- (d) a benzoate derivative of formula (II),



wherein

- 15 R² is a linear or branched C₅ to C₂₅ alkyl residue and
- R³ and R⁴ are independently from each other Methyl, Ethyl, linear and/or branched C₃ to C₁₂ alkyl residues.

2. Polymer composition (C) according to claim 1 or 2, wherein R¹ of formula (I) is C₁₁ to C₁₈ alkyl and the benzoate derivative of formula (II) is *n*-hexadecyl-3,5-di-*tert*-butyl-4-hydroxybenzoate.
- 20
3. Polymer composition (C) according to any one of the preceding claims, wherein said polymer composition (C) comprises
- 25 (a) the heterophasic propylene copolymer (HECO) in an amount of 30 to 80 wt.-%,

- 28 -

- (b) the mineral filler (F) in an amount no higher than 40 wt.-%, and
(c) the 1,2,2,6,6-pentamethyl-4-piperidylester derivative of formula (I) and the benzoate derivative of formula (II) combined in an amount of 0.01 to 5 wt.-%,
5 based on the total composition.
4. Polymer composition (C) according to any one of the preceding claims, wherein the heterophasic propylene copolymer (HECO) comprises
(a) a polypropylene matrix (M) being a propylene homopolymer or a propylene
10 copolymer, and
(b) an elastomeric propylene copolymer (E).
5. Polymer composition (C) according to any one of the preceding claims, wherein the heterophasic propylene copolymer (HECO) has
15 (a) a melt flow rate MFR_2 (230 °C), measured according to ISO 1133, in the range of 1.0 to 50 g/10 min,
(b) a comonomer content in the range of 3.0 to 25 wt.-%, based on the total heterophasic propylene copolymer (HECO), and
(c) a xylene soluble content (XCS) in the range of 10 to 50 wt.-%, based on the
20 total heterophasic propylene copolymer (HECO).
6. Polymer composition (C) according to any one of the preceding claims, wherein the matrix (M) of the heterophasic propylene copolymer (HECO) has a melt flow rate MFR_2 (230 °C), measured according to ISO 1133, in the range of 30 to 90 g/10 min.
25
7. Polymer composition (C) according to any one of the preceding claims, wherein the mineral filler (F) is talc, preferably said talc a specific surface area (BET) in the range of 5 to 25 m²/g.
- 30 8. Polymer composition (C) according to any one of the preceding claims, wherein said polymer composition comprises a polypropylene (PP), e.g. a polypropylene (PP)

- 29 -

being a propylene homopolymer, wherein said propylene (PP), e.g. the polypropylene (PP) being a propylene homopolymer, preferably has a melt flow rate MFR_2 (230 °C), measured according to ISO 1133, in the range of 3 to 50 g/10 min.

- 5 9. Polymer composition (C) according to any one of the preceding claims, wherein said polymer composition comprises a high density polyethylene (HDPE), preferably said high density polyethylene (HDPE) has

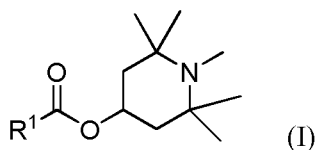
(a) a melt flow rate MFR_2 (190 °C), measured according to ISO 1133, in the range of 2 to 30 g/10 min

10 and/or

(b) a density, measured according to ISO 1183, of at least 930 kg/m³.

10. Use of a combination of

(a) a 1,2,2,6,6-pentamethyl-4-piperidylester derivative of formula (I)

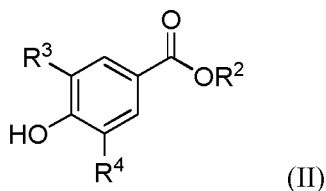


wherein

the molecular weight is below 1000 g/mol and R¹ is a linear or branched C₅ to C₂₅ alkyl residue, and

and

- 20 (b) a benzoate derivative of formula (II)



wherein

R² is a linear or branched C₅ to C₂₅ alkyl residue and

R³ and R⁴ are independently from each other methyl, ethyl, linear and/or branched C₃ to C₁₂ alkyl residues,

25

- 30 -

in a polymer composition (C) comprising further a heterophasic propylene copolymer (HECO) and a mineral filler (F) to reduce the effect of stickiness of the polymer composition (C) and/or of the automotive article comprising said polymer composition (C).

5

11. Use according to claim 10, wherein the reduction of stickiness is accomplished in case the stickiness index is below a value of 0.75.

10 12. Use according to claim 10 or 11, wherein the 1,2,2,6,6-pentamethyl-4-piperidylester derivative of formula (I) and/or the benzoate derivative of formula (II) are further defined as in claim 2.

13. Use according to any one of the claims 10 to 12, wherein the polymer composition (C) is further defined as in any one of the claims 1, and 3 to 9.

15

14. Automotive article comprising a polymer composition (C) according to any one of the preceding claims 1 to 9.

20 15. Process for producing a polymer composition (C) according to any one of the preceding claims 1 to 9 comprising the step of mixing the heterophasic propylene copolymer (HECO), the mineral filler (F), the 1,2,2,6,6-pentamethyl-4-piperidylester derivative of formula (I), the benzoate derivative of formula (II) and optionally the high density polyethylene (HDPE) and/or the polypropylene (PP) in an extruder.

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2013/076879

A. CLASSIFICATION OF SUBJECT MATTER

INV. C08L23/10 C08K5/3435
ADD. C08K3/3495 C08K3/3475 C08K3/00 C08K5/103 C08L23/16

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

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

C08L C08K

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

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

EPO-Internal, PAJ, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2006/105178 A1 (HONDA MOTOR CO LTD [JP]; WALKER ERIC [US]; MERHEB ELIE N [US]; RYAN JA) 5 October 2006 (2006-10-05) the whole document	1, 15
Y	EP 2 386 602 A1 (BOREALIS AG [AT]) 16 November 2011 (2011-11-16) the whole document	1-15
Y	US 2009/170984 A1 (BLUM TILL [DE] ET AL) 2 July 2009 (2009-07-02) the whole document	1-15
A	EP 2 410 007 A1 (BOREALIS AG [AT]) 25 January 2012 (2012-01-25) abstract; claims 1-15	1-15
	----- -/-	



Further documents are listed in the continuation of Box C.



See patent family annex.

* Special categories of cited documents :

"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

7 January 2014

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