

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property

Organization

International Bureau

(43) International Publication Date

23 September 2021 (23.09.2021)



(10) International Publication Number

WO 2021/185924 A1

(51) International Patent Classification:

C08F 210/06 (2006.01) C08L 23/06 (2006.01)

C08F 4/6592 (2006.01) C08F 210/16 (2006.01)

Published:

- with international search report (Art. 21(3))
- before the expiration of the time limit for amending the claims and to be republished in the event of receipt of amendments (Rule 48.2(h))

(21) International Application Number:

PCT/EP2021/056854

(22) International Filing Date:

17 March 2021 (17.03.2021)

(25) Filing Language:

English

(26) Publication Language:

English

(30) Priority Data:

20164256.8 19 March 2020 (19.03.2020) EP

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(81) Designated States (unless otherwise indicated, for every kind of national protection available): AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CZ, DE, DJ, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IR, IS, IT, JO, JP, KE, KG, KH, KN, KP, KR, KW, KZ, LA, LC, LK, LR, LS, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SA, SC, SD, SE, SG, SK, SL, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, WS, ZA, ZM, ZW.

(84) Designated States (unless otherwise indicated, for every kind of regional protection available): ARIPO (BW, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SD, SL, ST, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, KM, ML, MR, NE, SN, TD, TG).

(54) Title: HETEROPHASIC POLYPROPYLENE COMPOSITION

(57) Abstract: The present invention provides a heterophasic polypropylene composition with good response to nucleating agents, increased crystallisation temperature, good mechanical properties The present invention further relates to articles made thereof, and the use of the heterophasic polypropylene composition for specific applications and producing these.



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Heterophasic polypropylene composition

The present invention provides a heterophasic polypropylene composition with good response to nucleating agents, increased crystallisation temperature, and good mechanical properties

- 5 The present invention further relates to articles made thereof, and the use of the heterophasic polypropylene composition for specific applications and producing these.

Background Information

10 Heterophasic polypropylene compositions are known in the art. They are composed of a continuous propylene homo- or copolymer phase acting as a matrix, in which an elastomeric phase, e.g. and ethylene propylene rubber (EPR) is dispersed.

Heterophasic polypropylene copolymers are widely used in injection moulding applications, especially for producing packaging articles, as they are well known and appreciated for their mechanical properties as well as their very convenient processing. Typical applications are
15 e.g. thin wall packaging, houseware applications, containers for food and other packaging issues, etc.

Said heterophasic polypropylene copolymers are often further modified with various nucleating agents to increase the crystallization temperature, or improve mechanical properties like stiffness.

20 A high crystallization temperature is also desired to allow faster conversion and consequently a reduction of cycle time, e.g. in injection moulding.

Said increase of stiffness however can negatively influence the impact behaviour, making polymers more brittle.

25 There is accordingly in general a conflict of aims if one wants at the same time provide a heterophasic polypropylene composition with high stiffness and high impact behaviour at the same time.

Single site catalysts (SSC) are well known in the field of producing polypropylenes. They are
30 increasingly used for copolymerisation of propylene with other comonomers, due to their highly random insertion of the comonomer units along the chain.

Propylene polymers produced with single site catalysts are known to provide better stiffness/impact balance than comparable polymers produced with Ziegler-Natta catalysts. However, due to the differences in the molecular structures, SSC-based polymers are widely

reported to have weaker response to nucleation. This is mainly expressed in lower crystallization temperatures than observed in polymers produced via Ziegler-Natta catalysis while having similar polymer characteristics as e.g. comonomer content and type, amount of soluble fractions, etc.

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Description of the prior art:

M. Varma-Nair et al. report in "Quiescent crystallization kinetics of nucleated metallocene and ZN isotactic polypropylenes", (*Journal of Thermal Analysis and Calorimetry*, **59** (2000) 483-495), that both α - and β -nucleation response is worse in polypropylene copolymers produced via single-site catalysis, although the respective homopolymers have almost the same crystallization behaviour as those based on heterogeneous Ti-halogenide (Ziegler-Natta) catalysts. The crystallisation temperatures of metallocene polypropylenes are reported to be at most 123 °C, and only homopolymers of limited toughness are presented.

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WO/2020/011825 discloses a heterophasic polypropylene composition, which comprises 60.0 to 95.0 wt.-% of a crystalline propylene homopolymer, 5.0 to 20.0 wt.-% of an elastomeric ethylene-propylene rubber, and at least one polymeric nucleating agent, wherein the crystalline propylene homopolymer (H-PP) and the elastomeric ethylene-propylene rubber (EPR) have been produced in the presence of a single-site catalyst. It discloses polymers having lower amounts of comonomer in the soluble fraction, which have good transparency but very limited toughness, especially at -20 °C.

20

EP 2075284 A1 discloses a heterophasic propylene copolymer having a high flowability with a fraction insoluble in cold xylene (XCI) having intrinsic viscosity of below 1.5 dl/g, a fraction soluble in cold xylene (XCS) having an intrinsic viscosity of 1.5 to 3.0 dl/g and a comonomer content in the fraction soluble in cold xylene (XCS) of 25 to 50 mol %. Its example have an intrinsic viscosity of the in fraction insoluble in cold xylene, IV of XCI of 0.7 – 0.9 dl/g. It aims for improved impact behaviour at low temperatures combined with good flowability, but is however poor in toughness, especially at -20 °C.

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WO/2019/134951 discloses a heterophasic propylene copolymer comprising a) 65 to 90 wt.-% of a matrix component (M) selected from a propylene homo- or random copolymer (PP); and b) 10 to 35 wt.-% of an ethylene-propylene rubber (EPR), dispersed in the propylene homo- or random copolymer (PP), whereby the xylene cold soluble fraction (XCS) of the heterophasic propylene polymer (HECO) has an ethylene content of 12 to 20 wt.%; an

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intrinsic viscosity (IV), determined according to ISO 1628-1:2009, of at least 1.6 dl/g; and the xylene cold insoluble fraction (XCI) of the heterophasic propylene polymer (HECO) has 2,1-erythro regiodefects in an amount of at least 0.4 mol%. It discusses the addition of nucleating agents.

- 5 The examples disclosed have a comonomer content of the soluble fraction, C2 (XCS) in between 14.1 and 14.8 wt.-%, having good transparency but very limited toughness, especially at -20 °C.

Object of the invention

- 10 The present invention overcomes the drawbacks of the prior art. It discloses a heterophasic polypropylene composition with a specific combination of the amount of the soluble fraction in the heterophasic polypropylene composition, its comonomer content and the nucleating agent. This leads to a heterophasic polypropylene composition having good response to nucleating agents, increased crystallisation temperature and good mechanical properties.

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In particular, it discloses a heterophasic propylene composition comprising 99.0 – 99.99 wt.-% of a heterophasic propylene copolymer and 0.01 – 1.0 wt.-% of a nucleating agent,

wherein the heterophasic propylene copolymer is characterised by comprising

- 20 i) 65.0 to 90.0 wt.-% of a crystalline matrix being a propylene homo- or copolymer, said crystalline matrix corresponding to the crystalline fraction (CF) determined according to CRYSTEX QC method, ISO 6427-B, having an intrinsic viscosity, IV(CF) in the range of 1.0 – 4.0 dl/g and containing 1.5 to 3.5 wt.-% comonomer and
- ii) 10.0 to 35.0 wt.-% of an amorphous propylene-ethylene elastomer, optionally comprising
- 25 of C4-C12 alpha -olefin(s) as further comonomers, dispersed in said crystalline matrix (i), said amorphous propylene ethylene elastomer (ii) corresponds to the soluble fraction (SF) determined according to CRYSTEX QC method, ISO 6427-B, and has comonomer content of soluble fraction, (C2 of SF) in the range of 20.0 - 35.0 wt.-%, wherein i) and ii) add up to 100 wt.-%,
- 30 characterised in that the heterophasic polypropylene composition has a crystallisation temperature (Tc) of at least 124 °C.

The present invention also discloses articles, like injection moulded articles comprising said heterophasic polypropylene composition as well as the use of the heterophasic

- 35 polypropylene composition for producing such articles.

The present polymers provide an optimized correlation between crystallisation temperature, T_c and melting Temperature T_m . The increase in the crystallisation temperature ensures a fast crystallisation onset, which is more favourable during processing than a small range of under cooling, i.e. a small ΔT_{mc} .

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Detailed description:

The heterophasic polypropylene composition of the present invention comprises

- a) 99.0 – 99.99 wt.-% of a heterophasic propylene copolymer and
- b) 0.01 – 1.0 wt.-% of a nucleating agent.

10

Preferably, the heterophasic polypropylene composition comprises 99.2 to 99.95 wt.-%, preferably 99.5 to 99.9 wt.-% of heterophasic propylene copolymer and 0.05 to 0.8 wt.-%, preferably 0.1 to 0.5 wt.-% of a nucleating agent.

Preferably, the heterophasic polypropylene composition of the present invention consists of 99.0 – 99.99 wt.-% of a heterophasic propylene copolymer and 0.01 – 1.0 wt.-% of a nucleating agent.

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The MFR_{230/2.16} of the heterophasic polypropylene composition may be in the range of 0.1 to 100 g/10 min, preferably in the range of 0.5 to 50 g/10 min, more preferably in the range of 1.0 to 20 g/10 min.

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The crystallisation temperature (T_c) of the heterophasic polypropylene composition may be in the range of at least 124 to 135 °C, preferably in the range of 125 to 132 °C, more preferably in the range of 126 to 130 °C.

25

The melting temperature of the heterophasic polypropylene composition (T_m) may be in the range of 150 to 165 °C, preferably in the range of 155 to 164 °C, more preferably in the range of 157 to 163 °C.

The melting enthalpy of the heterophasic polypropylene composition may be in the range of 70 to 100 J/g, preferably in the range of 75 to 98 J/g, more preferably in the range of 78 to 96 J/g.

30

The heterophasic polypropylene composition may have a fraction soluble in cold xylene (XCS) in the range of 10.0 to 35.0 wt.-%, preferably in the range of 12.0 to 31.0 wt.-%, more preferably in the range of 14.0 to 28.0 wt.-%.

Referring also to the Crystex method description, it is herewith noted, that the values for XCS
5 roughly, but not precisely, correspond to the soluble fraction (SF) as determined in Crystex.

The heterophasic propylene composition is formed to a major part by the heterophasic polypropylene copolymer.

Accordingly, properties relating to the polymer (like comonomer content or quality, amount
10 and compositions of the crystalline or soluble fractions, viscosities of the crystalline and soluble fractions, etc.) are applicable to both the heterophasic polypropylene composition as well as the heterophasic propylene copolymer likewise.

Heterophasic propylene copolymer

15 The heterophasic propylene copolymer forms the major part of the heterophasic polypropylene composition and accordingly the first influences the properties of the latter.

The heterophasic propylene copolymer comprises 65.0 to 90.0 wt.-%, preferably 69.0 to 88.0 wt.-%, more preferably in the range of 72.0 to 86.0 wt.-% of a propylene homo- or copolymer,
20 forming the crystalline matrix (i), which corresponds to the crystalline fraction CF determined by CRYSTEX QC method ISO6427-B.

Accordingly, the heterophasic polypropylene copolymer comprises an (ii) amorphous propylene ethylene elastomer dispersed in said crystalline matrix in the range of 10.0 to 35.0 wt.-%, preferably in the range of 12.0 to 31.0 wt.-%, more preferably in the range of 14.0 to
25 28.0 wt.-%. Said amorphous elastomer corresponds to the soluble fraction (SF) determined by CRYSTEX QC method ISO6427-B.

In a preferred embodiment, the heterophasic propylene copolymer consists of

30 i) 65.0 to 90.0 wt.-%, preferably 69.0 to 88.0 wt.-%, more preferably in the range of 72.0 to 86.0 wt.-% of a propylene homo- or copolymer, forming the crystalline matrix (i), and
ii) 10.0 to 35.0 wt.-%, preferably in the range of 12.0 to 31.0 wt.-%, more preferably in the range of 14.0 to 28.0 wt.-% of an amorphous propylene ethylene elastomer dispersed in said crystalline matrix,

The heterophasic polypropylene copolymer contains comonomers, preferably alpha-olefins selected from ethylene and C4 to C8 alpha olefins, preferably from ethylene, 1-butene or 1-hexene.

5 In a preferred embodiment, the heterophasic polypropylene copolymer comprises ethylene and 1-butene as comonomer.

In an especially preferred embodiment, the heterophasic polypropylene copolymer comprises, only ethylene as the sole comonomer.

10 The comonomer content of the heterophasic propylene copolymer, C2 (total), may be in the range of may be in the range of 1.0 to 10.0 wt.-%, preferably in the range of 2.5 to 8.5 wt.-%, more preferably in the range of 4.0 to 7.5 wt.-%

15 The amount of the crystalline fraction (CF) of the heterophasic polypropylene copolymer is determined via determined by CRYSTEX QC method ISO6427-B and may be in the range of 65.0 – 90.0 wt.-%, preferably 69.0 – 88.0 wt.-%, more preferably in the range of 72.0 to 86.0 wt.-%.

20 The crystalline fraction may comprise comonomer as lined out above. The comonomer content of the crystalline fraction, C2(CF), may be in the range of 1.5 to 3.5 wt.-%, like 1.7 to 3.2 wt.-% or 1.7 to 2.8 wt.-%.

25 The soluble fraction (SF) of heterophasic propylene copolymer is also determined via determined by CRYSTEX QC method ISO6427-B and may be in the range of 10.0 to 35.0 wt.-%, preferably in the range of 12.0 to 31.0 wt.-%, more preferably in the range of 14.0 to 28.0 wt.-%.

The soluble fraction may comprise comonomer(s) as lined out above. The comonomer content of the soluble fraction, C2(SF), may be in the range of 20.0 to 35.0 wt.-%, preferably in the range of 21.0 to 33.0 wt.-%, more preferably in the range of 21.5 to 31.0 wt.-%. or 22.5 to 30.0 wt.-%

30 It is understood that the quality and quantity of the comonomers in the heterophasic polypropylene copolymer, the crystalline fraction as well as the soluble fraction may be selected and varied independently from each other.

35 The intrinsic viscosity of the soluble fraction of the heterophasic propylene copolymer, IV(SF), may be in the range of 2.0 to 5.0 dl/g, preferably in the range of 2.2 to 4.5 dl/g, more preferably in the range of 2.3 to 4.0 dl/g.

The intrinsic viscosity of the crystalline fraction of the heterophasic propylene copolymer, IV(CF), may be in the range of 1.0 to 4.0 dl/g, preferably in the range of 1.3 to 3.7 dl/g, more preferably in the range of 1.5 to 3.4 dl/g like 1.7 to 3.2 dl/g

The ratio between the IV(SF) to the IV(CF) may be in the range of at least 0.83 to 2.5,
5 preferably in the range of 0.85 to 2.2, more preferably in the range of 0.85 to 2.0.

The crystalline fraction (CF) of the heterophasic propylene polymer (HECO) may have 2,1-erythro region defects in an amount of at least 0.3 wt.-%. Usually the amount of 2,1-erythro region defects in the xylene cold insoluble fraction (XCI) of the heterophasic propylene
10 polymer (HECO) may be not higher than 2.0 wt.-%. It may be preferably in the range of 0.5 to 1.5 wt.-% or 0.6 to 1.2 wt.-%.

As well-known in the art, the amount of 2,1- erythro regiodeflects in propylene homo- or random copolymers produced with a Ziegler Natta catalyst is 0.0 wt.-%. Thus, the heterophasic propylene polymer (HECO) is usually produced using a single-site catalyst
15 such as a metallocene catalyst. Suitable catalysts are provided below.

All the properties described for the heterophasic propylene copolymer above are accordingly also applicable for the heterophasic polypropylene composition of the present invention.

20 The heterophasic propylene copolymer does not contain any nucleating agents. Thus, the mechanical and/or crystallisation behaviour of the neat polymer will be different to the according properties of the (final) heterophasic polypropylene composition of the present invention:

25 The crystallisation temperature (T_c) of the neat heterophasic propylene copolymer may be in the range of 110 to 125 °C, preferably in the range of 111 to 122 °C, more preferably in the range of 113 to 120 °C.

The melting temperature (T_m) neat heterophasic propylene copolymer may be in the range of 150 to 165 °C, preferably in the range of 153 to 163 °C, more preferably in the range of 155
30 to 162 °C.

The flexural modulus of the neat heterophasic propylene copolymer may be in the range of 500 to 2000 MPa, preferably in the range of 600 to 1600 MPa, more preferably in the range of 650 to 1300 MPa.

35 Melting enthalpy of the neat heterophasic propylene copolymer may be in the range of 60 to 95 J/g, preferably in the range of 65 to 93 J/g, more preferably in the range of 68 to 90 J/g.

Charpy NIS/23°C of the neat heterophasic propylene copolymer may be in the range of 5.0 to 100.0 kJ/m², preferably in the range of 15.0 to 90.0 kJ/m², more preferably in the range of 20.0 to 80.0 kJ/m².

Charpy NIS/-20 °C of the neat heterophasic propylene copolymer may be in the range of 1.0 to 20 kJ/m², preferably in the range of 2.0 to 15 kJ/m², more preferably in the range of 2.5 to 12.0 kJ/m².

Nucleating agent:

The heterophasic polypropylene composition of the present invention comprises 0.01 – 1.0 wt.-% (based on the total weight of the heterophasic polypropylene composition) of a nucleating agent.

The nucleating agent is selected from the following groups of nucleating agents consisting of:

- (i) monocarboxylic acids and polycarboxylic acids,
- (ii) salts of diesters of phosphoric acid, and
- 15 (iii) polymeric nucleating agents.
- (iv) sorbitol derivatives and
- (v) trisamide derivatives.

Accordingly, the nucleating agent may be selected from the groups comprising

- 20 (i) salts of monocarboxylic acids and polycarboxylic acids, e.g. sodium benzoate or aluminum tert-butylbenzoate; or 1,2-cyclohexane dicarboxylic acid Ca-salt, of which the 1,2-cyclohexane dicarboxylic acid Ca-salt is especially preferred;
- (ii) salts of diesters of phosphoric acid, e.g. sodium 2,2'-methylenebis (4, 6,-di-tertbutylphenyl) phosphate or aluminium-hydroxy-bis[2,2'-methylene-bis(4,6-di-
- 25 tbutylphenyl)phosphate], and hydroxybis (2,4,8,10-tetra-tert- butyl-6-hydroxy-12H-dibenzo(d,g)(1,3,2) dioxaphosphocin 6-oxidato) aluminium, wherein hydroxybis (2,4,8,10-tetra-tert- butyl-6-hydroxy-12H-dibenzo(d,g)(1,3,2) dioxaphosphocin 6-oxidato) aluminium or sodium 2,2'-methylenebis (4, 6,-di-tertbutylphenyl) phosphate are preferred;
- (iii) crystalline polymers, like poly(vinyl cyclohexane) (PVCH), poly(4-methyl pentene-1)
- 30 (P4MP) and poly(tetrafluoro ethylene), where PVCH is preferred;
- (iv) sorbitol derivatives, e.g. di(alkylbenzylidene)sorbitols as 1,3:2,4-dibenzylidene sorbitol, 1,3:2,4-di(4-methylbenzylidene) sorbitol, 1,3:2,4-di(4-ethylbenzylidene) sorbitol and 1,3:2,4-Bis(3,4-dimethylbenzylidene) sorbitol, as well as nonitol derivatives, e.g. 1,2,3- trideoxy-4,6;5,7-bis-O-[(4-propylphenyl)methylene] nonitol; and
- 35 (v) benzene-trisamides like substituted 1,3,5-benzenetrisamides as N,N',N''-tris-tert-butyl-1,3,5- benzenetricarboxamide, N,N',N''-tris-cyclohexyl-1,3,5-benzene-tricarboxamide

and N-[3,5-bis-(2,2-dimethyl-propionylamino)-phenyl]-2,2-dimethyl-propionamide. It is especially preferred, that the at least one α -nucleating agent (c) in the heterophasic polypropylene composition of the present invention is selected from 1,3:2,4-dibenzylidene sorbitol, 1,3:2,4-Bis(3,4-dimethylbenzylidene) sorbitol, 1,2,3-trideoxy-4,6;5,7-bis-O-[(4-propylphenyl)methylene] nonitol and N-[3,5-bis-(2,2-dimethyl propionylamino)-phenyl]-2,2-dimethyl-propionamide.

In a preferred embodiment, the heterophasic polypropylene composition does not comprise polymeric nucleating agents.

10 The following nucleating agents are known as polymeric nucleating agents and accordingly not envisaged in the preferred embodiment of the invention:
crystalline polymers, like poly(vinyl cyclohexane) (PVCH), poly(4-methyl pentene-1) (P4MP) and poly(tetrafluoro ethylene), where PVCH is preferred;

15 In a particular embodiment, particular, i.e. non-soluble nucleating agents as listed in groups (i) or (ii) are preferred.

Especially preferred are non-soluble and non-polymeric nucleating agents as listed in groups (i), or (ii), especially preferred are hydroxybis (2,4,8,10-tetra-tert-butyl-6-hydroxy-12H-dibenzo(d,g)(1,3,2) dioxaphosphocin 6-oxidato) aluminium or sodium 2,2'-methylenebis (4,6,-di-tertbutylphenyl) phosphate.

Additives:

The heterophasic polypropylene composition according to the invention may further comprise conventional additives in an amount of up to 5.0 wt.-%, preferably in an amount of 0.1 to 2.0 wt.-%, more preferably in an amount of 0.3 to 1.0 wt.-%. Examples of additives include, but are not limited to, stabilizers such as antioxidants (for example sterically hindered phenols, phosphites/phosponites, sulphur containing antioxidants, alkyl radical scavengers, aromatic amines, hindered amine stabilizers, or blends thereof), metal deactivators (for example Irganox® MD 1024), or UV stabilizers (for example hindered amine light stabilizers). Other typical additives are modifiers, like antistatic or antifogging agents (for example ethoxylated amines and amides or glycerol esters), acid scavengers (for example Ca-stearate) and blowing agents for foaming. Further modifiers are lubricants and resins (for example ionomer waxes, polyethylene- and ethylene copolymer waxes, Fischer Tropsch waxes, montan-based waxes, fluoro-based compounds, or paraffin waxes), as well as slip and antiblocking agents (for example erucamide, oleamide, talc, natural silica and synthetic silica or zeolites) and mixtures thereof.

It is further understood, that polymers introduced as carriers of masterbatches or the like, may be present in the heterophasic polypropylene composition in the range of 0.1 to 5.0 wt.-%, even in case the claim is formulated in the closed wording, (“heterophasic polypropylene composition consisting of...”).

Mechanical Characterisation:

The heterophasic polypropylene composition of the present invention may be characterised by its physical, i.e. mechanical properties:

The flexural modulus according to ISO178 may be in the range of 600 to 2500 MPa, preferably in the range of 700 to 2000 MPa, more preferably in the range of 850 to 1700 MPa.

The notched impact strength, ISO179, determined at +23 °C, Charpy NIS/23°C, may be in the range of 5.0 to 100.0 kJ/m², preferably in the range of 15.0 to 90.0 kJ/m², more preferably in the range of 20.0 to 80.0 kJ/m².

In a particular embodiment, the Charpy NIS/23°C, may be in the range of 35 – 90 kJ/m², like 60 – 85 kJ/m².

The notched impact strength, ISO179, determined at -20 °C, Charpy NIS/-20 °C, may be in the range of 1.0 to 20 kJ/m², preferably in the range of 2.0 to 15 kJ/m², more preferably in the range of 2.5 to 12.0 kJ/m².

Ideally, the heterophasic polypropylene composition may have a flexural modulus according to ISO178 in the range of 600 to 2500 MPa, preferably in the range of 700 to 2000 MPa, or 850 to 1700 MPa and optionally either or both of:

a Charpy notched impact strength, ISO179, determined at +23 °C in the range of 5.0 to 100.0 kJ/m², preferably in the range of 15.0 to 90.0 kJ/m², or 20.0 to 80.0 kJ/m² and/or

a Charpy notched impact strength, ISO179, determined at -20 °C, in the range of 1.0 to 20 kJ/m², preferably in the range of 2.0 to 15 kJ/m², or 2.5 to 12.0 kJ/m².

Polymerisation process

The heterophasic propylene copolymer encompassed by the heterophasic polypropylene composition of the present invention is preferably produced by sequential polymerisation, wherein in one step the crystalline matrix being a propylene homo- or copolymer, is produced and in a subsequent step the amorphous propylene-ethylene elastomer is produced in the presence of above mentioned propylene homo- or copolymer.

Accordingly, the amorphous propylene ethylene elastomer is not present as a separate fraction.

- 5 The process for polymerising the heterophasic propylene copolymer encompassed by the heterophasic polypropylene composition comprises polymerising propylene in at least two, preferably two polymerisation steps in the presence of a single-site catalyst, wherein
- a) in a first polymerisation reactor propylene and optionally comonomers is polymerised to obtain a crystalline matrix being a propylene homo- or copolymer, said matrix is the
- 10 transferred to a second polymerisation reactor, preferably being a gas phase reactor (GPR),
- b) in the second polymerisation reactor, the amorphous propylene-ethylene elastomer is produced in the presence of above mentioned propylene homo- or copolymer.
- Consecutively, a nucleating agent may be introduced into the heterophasic propylene copolymer either by a compounding step or as a Master Batch, to form the heterophasic
- 15 polypropylene composition of the present invention.
- The process may be preceded by one or more prepolymerisation steps.

Catalyst

20 Catalyst system:

The heterophasic polypropylene composition according to the invention is preferably obtainable by a catalyst system comprising by a single-site catalyst, more preferably being obtainable by a metallocene catalyst complex and cocatalysts.

25 Particularly preferred catalyst are:

rac-anti-dimethylsilanediyl [2-methyl-4,7-bis-(3',5'-dimethyl phenyl)-1,5,6,7-tetrahydro-s-indacen-1-yl] [2-methyl-4-(3',5'-ditert.-butyl-phenyl)-5-methoxy-6-tert-butylinden-1-yl] zirconium dichloride or

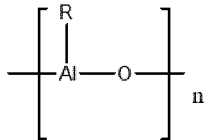
- 30 rac-anti-dimethylsilanediyl [2-methyl-4-(3',5'-dimethyl phenyl)-1,5,6,7-tetrahydro-s-indacen-1-yl][2-methyl-4-(3',5'-dimethylphenyl)-5-methoxy-6-tert-butyl inden-1-yl] zirconium dichloride dichloride).

Cocatalyst

- To form an active catalytic species it is normally necessary to employ a cocatalyst as is well
- 35 known in the art.

According to the present invention a cocatalyst system comprising a boron containing cocatalyst and an aluminoxane cocatalyst is used in combination with the above defined metallocene catalyst complex.

5 The aluminoxane cocatalyst can be one of formula (I):



(I)

where n is from 6 to 20 and R has the meaning below.

Aluminoxanes are formed on partial hydrolysis of organoaluminum compounds, for example those of the formula AlR_3 , AlR_2Y and $\text{Al}_2\text{R}_3\text{Y}_3$ where R can be, for example, C_1 - C_{10} -alkyl, preferably C_1 - C_5 -alkyl, or C_3 - C_{10} -cycloalkyl, C_7 - C_{12} -arylalkyl or -alkylaryl and/or phenyl or naphthyl, and where Y can be hydrogen, halogen, preferably chlorine or bromine, or C_1 - C_{10} -alkoxy, preferably methoxy or ethoxy. The resulting oxygen-containing aluminoxanes are not in general pure compounds but mixtures of oligomers of the formula (II).

The preferred aluminoxane is methylaluminoxane (MAO). Since the aluminoxanes used according to the invention as cocatalysts are not, owing to their mode of preparation, pure compounds, the molarity of aluminoxane solutions hereinafter is based on their aluminium content.

Also, a boron containing cocatalyst is used in combination with the aluminoxane cocatalyst. The catalyst complex ideally comprises a co-catalyst, certain boron containing cocatalysts are preferred. Especially preferred borates of use in the invention therefore comprise the trityl, i.e. triphenylcarbenium, ion. Thus the use of $\text{Ph}_3\text{CB}(\text{PhF}_5)_4$ and analogues therefore are especially favoured.

The catalyst system of the invention is used in supported form. The particulate support material used is silica or a mixed oxide such as silica-alumina, in particular silica.

The use of a silica support is preferred. The skilled man is aware of the procedures required to support a metallocene catalyst.

In a preferred embodiment, the catalyst system corresponds to the ICS3 of EP19177308.4.

30 Articles

The present invention also covers final articles, especially moulded articles comprising the heterophasic polypropylene composition of the present invention.

The articles may be injection moulded and may be used for packaging purposes or for application in the automotive industry.

Preferably, said articles have a wall thickness of 0.1 to 3.0 mm, such as 0.5 to 2.5 mm, like 1.0 to 2.0 mm.

5

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

Examples:

Measuring methods

5 Melt Flow Rate

The melt flow rate (MFR₂) is determined according to ISO 1133 and is indicated in g/10 min. The MFR is an indication of the flowability, and hence the processability, of the polymer. The higher the melt flow rate, the lower the viscosity of the polymer. The MFR₂ of polypropylene is determined at a temperature of 230 °C and a load of 2.16 kg.

10

Differential scanning calorimetry (DSC)

Differential scanning calorimetry (DSC) analysis, melting temperature (T_m) and melt enthalpy (H_m), crystallization temperature (T_c), and heat of crystallization (H_c, H_{CR}) are measured with a TA Instrument Q200 differential scanning calorimetry (DSC) on 5 to 7 mg samples. DSC is run according to ISO 11357 / part 3 / method C2 in a heat / cool / heat cycle with a scan rate of 10 °C/min in the temperature range of -30 to +225 °C. Crystallization temperature (T_c) and heat of crystallization (H_c) are determined from the cooling step, while melting temperature (T_m) and melt enthalpy (H_m) are determined from the second heating step.

20 Xylene Cold Soluble (XCS)

Xylene Cold Soluble fraction at room temperature (XCS, wt.-%) is determined at 25°C according to ISO 16152; 5th edition; 2005-07-01.

Flexural Modulus

25 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.

The ΔFlexural modulus defines the difference of the Flexural Modulus between a nucleated and a non-nucleated polymer.

30

Notched impact strength (NIS)

The Charpy notched impact strength (NIS) was measured according to ISO 179 1eA at +23°C or -20 °C, using injection moulded bar test specimens of 80x10x4 mm³ prepared in accordance with EN ISO 1873-2.

35

Crystex analysis

Crystalline and soluble fractions method

The crystalline (CF) and soluble fractions (SF) of the polypropylene (PP) compositions as well as the comonomer content and intrinsic viscosities of the respective fractions as well as
5 the comonomer content of the final polymers were analyzed by the CRYSTEX QC, Polymer Char (Valencia, Spain).

A schematic representation of the CRYSTEX QC instrument is shown in Figure 1a. The crystalline and amorphous fractions are separated through temperature cycles of dissolution at 160 °C, crystallization at 40 °C and re-dissolution in a 1,2,4-trichlorobenzene (1,2,4-TCB)
10 at 160 °C as shown in Figure 1b. Quantification of SF and CF and determination of ethylene content (C2) are achieved by means of an infrared detector (IR4) and an online 2-capillary viscometer which is used for the determination of the intrinsic viscosity (IV).

The IR4 detector is a multiple wavelength detector detecting IR absorbance at two different bands (CH3 and CH2) for the determination of the concentration and the Ethylene content in
15 Ethylene-Propylene copolymers. IR4 detector is calibrated with series of 8 EP copolymers with known Ethylene content in the range of 2 wt.-% to 69 wt.-% (determined by 13C-NMR) and various concentration between 2 and 13mg/ml for each used EP copolymer used for calibration.

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

The intrinsic viscosity (IV) of the parent EP copolymer and its soluble and crystalline fractions
25 are determined with a use of an online 2-capillary viscometer and are correlated to corresponding IV's determined by standard method in decalin according to ISO 1628.

Calibration is achieved with various EP PP copolymers with IV = 2-4 dL/g.

A sample of the PP composition to be analyzed is weighed out in concentrations of 10mg/ml to 20mg/ml. After automated filling of the vial with 1,2,4-TCB containing 250 mg/l 2,6-tert-
30 butyl-4-methylphenol (BHT) as antioxidant, the sample is dissolved at 160 °C until complete dissolution is achieved, usually for 60 min, with constant stirring of 800rpm.

As shown in a Figure 1a and b, a defined volume of the sample solution is injected into the column filled with inert support where the crystallization of the sample and separation of the soluble fraction from the crystalline part is taking place. This process is repeated two times.
35 During the first injection the whole sample is measured at high temperature, determining the IV[dL/g] and the C2[wt.-%] of the PP composition. During the second injection the soluble

fraction (at low temperature) and the crystalline fraction (at high temperature) with the crystallization cycle are measured (wt.-% SF, wt.-% C2, IV).

EP means ethylene propylene copolymer.

PP means polypropylene.

5

Quantification of microstructure by NMR spectroscopy

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

Quantitative $^{13}\text{C}\{^1\text{H}\}$ NMR spectra were recorded in the solution-state using a Bruker
10 Advance III 400 NMR spectrometer operating at 400.15 and 100.62 MHz for ^1H and ^{13}C respectively. All spectra were recorded using a ^{13}C optimised 10 mm extended temperature probehead at 125°C using nitrogen gas for all pneumatics.

For propylene homopolymers approximately 200 mg of material was dissolved in 1,2-tetrachloroethane- d_2 (TCE- d_2). To ensure a homogenous solution, after initial sample
15 preparation in a heat block, the NMR tube was further heated in a rotary oven for at least 1 hour. Upon insertion into the magnet the tube was spun at 10 Hz. This setup was chosen primarily for the high resolution needed for tacticity distribution quantification (Busico, V., Cipullo, R., Prog. Polym. Sci. 26 (2001) 443; Busico, V.; Cipullo, R., Monaco, G., Vacatello, M., Segre, A.L., Macromolecules 30 (1997) 6251). Standard single-pulse excitation was
20 employed utilising the NOE and bi-level WALTZ16 decoupling scheme (Zhou, Z., Kuemmerle, R., Qiu, X., Redwine, D., Cong, R., Taha, A., Baugh, D. Winniford, B., J. Mag. Reson. 187 (2007) 225; Busico, V., Carbonniere, P., Cipullo, R., Pellecchia, R., Severn, J., Talarico, G., Macromol. Rapid Commun. 2007, 28, 11289). A total of 8192 (8k) transients were acquired per spectra.

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

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

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

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

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

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

$$[\text{mmmm}] \% = 100 * (\text{mmmm} / \text{sum of all pentads})$$

The presence of **2,1 erythro regio-defects** was indicated by the presence of the two methyl sites at 17.7 and 17.2 ppm and confirmed by other characteristic sites. Characteristic signals corresponding to other types of regio-defects were not observed (Resconi, L., Cavallo, L., Fait, A., Piemontesi, F., Chem. Rev. 2000, 100, 1253).

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

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

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

$$P_{12} = I_{\text{CH}_3} + P_{12e}$$

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

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

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

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

25 **Material description:**

Catalyst description:

MC1

30 Catalyst used in PP-CE1 was **MC1**, (rac-anti-dimethylsilylindyl(2-methyl-4-phenyl-5-methoxy-6-tertbutyl-indenyl)(2-methyl-4-(4-tert-butylphenyl)indenyl)zirconium dichloride) has been synthesized as described in WO 2013/007650, E2..

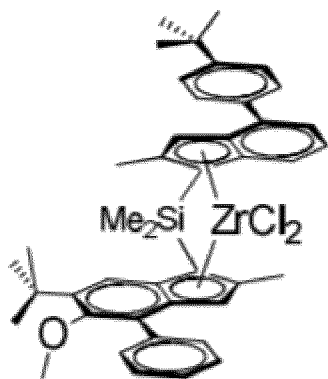


Figure 1: Metallocene MC1 for PP-CE1

The catalyst system was prepared using metallocene MC1 and a cocatalyst system of MAO.

5 The catalyst was supported onto silica

MC2

The catalyst used for PP1 and PP2 was MC2, rac-anti-dimethylsilanediyl [2-methyl-4,8-bis-(3',5'-dimethyl phenyl)-1,5,6,7-tetrahydro-s-indacen-1-yl] [2-methyl-4-(3',5'-ditert.-butyl-phenyl)-5-methoxy-6-tert-butylinden-1-yl] zirconium dichloride.

The catalyst was prepared as disclosed in WO2019/179959, for MC3, on page 40 and page 55.,

The catalyst system was prepared using metallocene MC2 and a cocatalyst system of MAO
15 and boron The catalyst was supported onto silica.

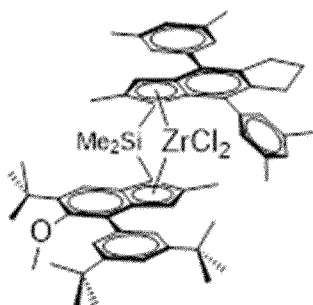


Figure 2: Metallocene MC2 for PP1 and PP2

20 Preparation of MAO-silica support.

A glass reactor equipped with a mechanical stirrer anchor-type stirrer was charged with silica (10.0 g), grade DM-L-303 from AGC Si-Tech Co, pre-calcined at 600 °C. Then toluene (50 mL) was added. The mixture was stirred under N₂, as MAO solution(25 mL, 30 wt% in

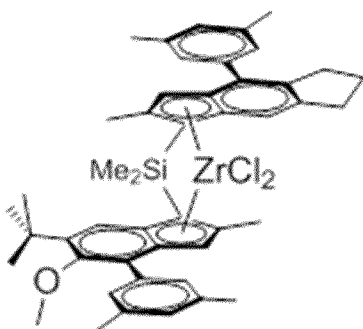
toluene) from Lanxess was added via cannula while maintaining the temperature around 35 °C. Upon completion of the addition, the solids were allowed to settle and the supernatant was removed and discarded. Fresh toluene (50 mL) was added and the mixture was then heated up to 80°C and stirred at 80 °C for additional hour. The slurry was allowed to settle and the mother liquor was filtered off. The catalyst was washed twice with toluene (50 mL) at 80°C, following by settling and filtration. The reactor was cooled down to 60°C and the solid was washed with heptane (50 mL). Finally MAO treated SiO₂ was dried at 60°C for 2h under vacuum. MAO treated support was collected as a free-flowing white powder (13 g).

10 Catalyst preparation

In a nitrogen filled glovebox, a solution of MAO 0.25 mL (30% wt in toluene, AXION 1330 CA Lanxess) in dry toluene (1 mL) was added to an aliquot of metallocene MC-D85 (39.0 mg, 39 μmol). The mixture was stirred for 30 minutes at room temperature. Next, trityl tetrakis(perfluorophenyl)borate (Boulder) (36.0 mg, 39 μmol) was added and the solution was allowed to stir for an additional 30 minutes at room temperature. Next, 1.0 g of MAO treated silica prepared as described above, was placed in a glass vial. A solution of metallocene, MAO and trityl borate in toluene was then slowly added to the support over the course of 5 minutes with gentle mixing. The resulting mixture was shaken well and allowed to stay overnight. Then dry toluene (5 mL) was added, and the slurry was mixed well with the inert gas sparging over 10 minutes. The solid was allowed to settle, and liquid was filtered off and discarded. The resulting cake was dried in vacuum for 1 hour to yield 1.0 g of the catalyst as pink free flowing powder.

MC3

The catalyst used for PP3 was MC3, rac-anti-dimethylsilanediyl [2-methyl-4-(3',5'-dimethylphenyl)-1,5,6,7-tetrahydro-s-indacen-1-yl][2-methyl-4-(3',5'-dimethylphenyl)-5-methoxy-6-tert-butyl inden-1-yl] zirconium dichloride dichloride) was synthesized according to the procedure as described in WO2019/007655, pp 49ff.



30 Figure 3: Metallocene MC3 for PP3

Preparation of MAO-silica support

A steel reactor equipped with a mechanical stirrer and a filter net was flushed with nitrogen and the reactor temperature was set to 20 °C. Next silica grade DM-L-303 from AGC Si-Tech
5 Co, pre-calcined at 600 °C (5.0 kg) was added from a feeding drum followed by careful
pressuring and depressurising with nitrogen using manual valves. Then toluene (22 kg) was
added. The mixture was stirred for 15 min. Next 30 wt% solution of MAO in toluene (9.0 kg)
from Lanxess was added via feed line on the top of the reactor within 70 min. The reaction
10 mixture was then heated up to 90 °C and stirred at 90 °C for additional two hours. The slurry
was allowed to settle and the mother liquor was filtered off. The catalyst was washed twice
with toluene (22 kg) at 90 °C, following by settling and filtration. The reactor was cooled off to
60 °C and the solid was washed with heptane (22.2 kg). Finally MAO treated SiO₂ was dried
at 60 ° under nitrogen flow for 2 hours and then for 5 hours under vacuum (-0.5 barg) with
15 stirring. MAO treated support was collected as a free-flowing white powder found to contain
12.2% Al by weight.

Catalyst preparation

30 wt% MAO in toluene (0.7 kg) was added into a steel nitrogen blanked reactor via a burette
at 20 °C. Toluene (5.4 kg) was then added under stirring. The MC3 as cited above (93 g)
20 was added from a metal cylinder followed by flushing with 1 kg toluene. The mixture was
stirred for 60 minutes at 20 °C. Trityl tetrakis(pentafluorophenyl) borate (91 g) was then
added from a metal cylinder followed by a flush with 1 kg of toluene. The mixture was stirred
for 1 h at room temperature. The resulting solution was added to a stirred cake of MAO-
silica support prepared as described above over 1 hour. The cake was allowed to stay for 12
25 hours, followed by drying under N₂ flow at 60 °C for 2h and additionally for 5 h under vacuum (-
0.5 barg) under stirring. Dried catalyst was sampled in the form of pink free flowing powder
containing 13.9% Al and 0.11% Zr.

The polymerisation was done in a 25L bench scale reactor. The details of the polymerisation
30 are shown in Table 1.

Table 1: Polymerisation Data

		PP-CE1	PP1	PP2	PP3
Catalyst		MC1	MC2	MC2	MC3
Prepolymerisation					
Residence time	min	10	10	10	10
H2	nl	0	0	0	0
Bulk step = Loop					
Temperature	°C	75	75	75	75
Total H2	nl	2	2	2	2
Residence time	min	30	30	30	30
Split	wt%	43	82	80	78
First Gas phase polymerisation					
Temperature	°C	80	70	70	70
Total H2	nl	2,0	0	0,1	0
Residence time	min	90	90	90	90
C2/C3	wt%/wt%	0,05	0,83	0,82	0,69
Pressure	barg	21	25	25	16
Split	wt%	41	18	20	22
Second Gas phase polymerisation					
Temperature	°C	75	not used	not used	not used
Total H2	nl	0			
Residence time	min	90			
C2/C3	wt%/wt%	0,67			
Pressure	barg	16			
Split	wt%	16			

All of the base polymer were made in a bench scale reactor.

The polymer powder were stabilized with 1500 ppm of Irganox B225 (supplied by BASF) and
5 500 ppm of synthetic hydrotalcit (MAHC).

Nucleated polymers contained 2000 ppm of NA 21, commercially available from Adeka.

CE2 was disclosed in WO2020/011825 as CE1.

PP-CE3 corresponds to Sample ID "M-iPP (cycled)" as disclosed in by M. Varma-Nair et
10 al.: "Quiescent crystallization kinetics of nucleated metallocene and ZN isotactic
polypropylenes", (*Journal of Thermal Analysis and Calorimetry*, **59** (2000) 483-495) in table 1
on p 486.

CE3 corresponds to Sample ID "M-iPP +0.2% α -nucl." as disclosed in by M. Varma-Nair et
al.: "Quiescent crystallization kinetics of nucleated metallocene and ZN isotactic

polypropylenes”, (*Journal of Thermal Analysis and Calorimetry*, **59** (2000) 483-495) in table 1 on p 486.

5

Table 2 Polymer properties of the heterophasic propylene copolymers used in the Inventive and Comparative examples

		CE1	CE2	CE3	IE1	IE2	IE3
Polymer:		PP-CE1		PP-CE3	PP1	PP2	PP3
MFR230/2.16	g/10min	4.0	2.5	2.8	5.0	6.0	1.0
Crystex:							
Soluble fraction (SF)	wt.-%	18	12	< 1.0	17	19	23
C2 (total)	wt.-%	5.9	2.3	0	6	.	6.8
C2(SF)	wt.-%	19	17.1	0	27	28.2	23.9
C2(CF)	wt.-%	3.4	1.3	0	2.6	2.5	1.8
IV(SF)	dl/g	1.8	1.9	n.d.	3.6	3.6	2.5
IV(CF)	dl/g	2.2	2.3	n.d.	2.0	1.9	2.9
Ratio IV(SF) / IV(CF)	-	0.82	0.83	-	1.80	1.89	0.86
NMR:							
<mmmm> matrix	%	n.d.	99.4	n.d.	99.7	99.7	99.4
<2,1> regio-defects	mol%	n.d.	0.7	n.d.	0.9	0.9	0.7

Table 3: Mechanical properties of the heterophasic propylene copolymer (not nucleated)

		PP-CE1	PP-CE3	PP1	PP2	PP3
Used in example:		CE1	CE3	IE1	IE2	IE3
Flexural modulus	MPa	493		1056	998	786
Tc	°C	108	114,8	118	118	115
Tm1	°C	145	154,8	160	160	157
Hm1	J/g	63		86	84	77
NIS/23°C	kJ/m ²	61		25.1	45.2	77.2
NIS/-20 °C	kJ/m ²	1.3		5.7	6.4	2.9

Table 4: Mechanical properties of the heterophasic propylene compositions of the comparative and inventive examples

		CE1	CE2	CE3	IE1	IE2	IE3
Nucleation agent:		2000 ppm NA21	15 ppm pVCH	2000 ppm DMDBS	2000 ppm NA21	2000 ppm NA21	2000 ppm NA21
Flexural modulus	MPa	580	1421		1355	1264	998
T _c	°C	119	126	123,0	128	128	126
T _{m1}	°C	149	162	154,4	162	162	159
H _{m1}	J/g	65			94	89	80
NIS/23°C	kJ/m ²	60.0			26.5	49.0	74.6
NIS/-20 °C	kJ/m ²	1.2			5.7	6.6	3.2
ΔFlexural modulus	MPa	87			299	266	212
T _m -T _c	°C	30	36	31,4	34	34	33
ΔT _c	°C	11		8,2	10	10	11

Table 4 clearly shows, that the inventive examples IE1, IE2 and IE3 have a specific
5 combination of the polymer features and fulfil the requirements in view of crystallisation
temperature. The object of the invention is fulfilled.

Claims

- 1) Heterophasic propylene composition comprising
- a) 99.0 – 99.99 wt.-% of a heterophasic propylene copolymer and
 - b) 0.01 – 1.0 wt.-% of a nucleating agent,
- 5 wherein the heterophasic propylene copolymer is characterised by comprising
- i) 65.0 to 90.0 wt.-% of a crystalline matrix being a propylene homo- or copolymer, said crystalline matrix corresponding to the crystalline fraction (CF) determined according to CRYSTEX QC method, ISO 6427-B, having an intrinsic viscosity, IV(CF) in the range of. 1.0 – 4.0 dl/g and
 - 10 containing 1.5 to 3.5 wt.-% comonomer and
 - ii) 10.0 to 35.0 wt.-% of an amorphous propylene-ethylene elastomer, optionally comprising of C4-C12 alpha -olefin(s) as further comonomers, dispersed in said crystalline matrix (i), said amorphous propylene ethylene elastomer (ii) corresponds to the soluble fraction (SF) determined according to CRYSTEX QC
 - 15 method, ISO 6427-B, and has comonomer content of soluble fraction, (C2 of SF) in the range of 20.0 - 35.0 wt.-%,
- wherein i) and ii) add up to 100 wt.-%,
- characterised in that the heterophasic polypropylene composition has a crystallisation temperature (Tc) of at least 124 °C and an Intrinsic Viscosity of the soluble fraction,
- 20 IV(SF), in the range of 2.0 - 5.0 g/dl.
- 2) The heterophasic polypropylene composition according to claim 1, comprising
- i) 69.0 to 88.0 wt.-%, preferably 72.0 to 86.0 wt.-% of a crystalline matrix being a propylene homo- or copolymer, said crystalline matrix corresponding to the
 - 25 crystalline fraction (CF) determined according to CRYSTEX QC method, ISO 6427-B and containing 1.5 to 3.5 wt.-% comonomer and
 - ii) 12.0 to 31.0 wt.-%, preferably 14.0 to 28.0 wt.-% of an amorphous propylene-ethylene elastomer, optionally comprising of C4-C12 alpha -olefin(s) as further comonomers, dispersed in said crystalline matrix (i), said amorphous propylene
 - 30 ethylene elastomer (ii) corresponds to the soluble fraction (SF) determined according to CRYSTEX QC method, ISO 6427-B.
- 3) The heterophasic polypropylene composition according to any of the preceding claims having a melting temperature Tm of at least 155 °C.

- 4) The heterophasic polypropylene composition according to any of the preceding claims, wherein the heterophasic polypropylene composition does not contain any polymeric nucleating agent..
- 5) The heterophasic polypropylene composition according to any of the preceding claims having an MFR_{230/2} according to ISO1133 of in the range 0.1 to 100 g/10 min, preferably in the range of 0.5 to 50 g/10 min, more preferably in the range of 1.0 to 20 g/10 min.
- 6) The heterophasic polypropylene composition according to any of the preceding claims, wherein the heterophasic propylene copolymer has a total comonomer content in the range of 1.0 to 10.0 wt.-%, preferably in the range of 2.5 to 8.5 wt.-%, more preferably in the range of 4.0 to 7.5 wt.-%.
- 7) The heterophasic polypropylene composition according to any of the preceding claims, wherein the heterophasic propylene copolymer has a comonomer content of soluble fraction, (C₂ of SF) in the range 21.5 - 33.0 wt.-%, or in the range of 22.5 - 31.0 wt.-%.
- 8) The heterophasic polypropylene composition according to any of the preceding claims wherein the heterophasic propylene copolymer has an Intrinsic Viscosity of the soluble fraction, IV(SF) 2.2 - 4.5 g/dl, preferably in the range of 2.3 - 4.0 g/dl and/or an Intrinsic Viscosity of the crystalline fraction IV(CF), in the range of 1.3 to 3.7 dl/g, preferably in the range of 1.5 to 3.4 dl/g, like 1.7 to 3.2 dl/g.
- 9) The heterophasic polypropylene composition according to any of the preceding claims having a ratio of the IV(SF) to the IV(CF) in the range of at least 0.83 to 2.5, preferably in the range of 0.85 to 2.2, more preferably in the range of 0.85 to 2.0.
- 10) The heterophasic polypropylene composition according to any of the preceding claims having 2,1-erythro region defects in an amount of at least 0.3 to 2.0 wt.-%, preferably in the range of 0.5 to 1.5 wt.-% or 0.6 to 1.2 wt.-%.
- 11) The heterophasic polypropylene composition according to any of the preceding claims having a crystallisation temperature of in the range of at least 124 to 135 °C, preferably in the range of 125 to 132 °C, more preferably in the range of 126 to 130 °C.

- 12) The heterophasic polypropylene composition according to any of the preceding claims comprising a non-soluble nucleating agent, preferably selected from hydroxybis (2,4,8,10-tetra-tert-butyl-6-hydroxy-12H-dibenzo(d,g)(1,3,2) dioxaphosphocin 6-oxidato) aluminium or sodium 2,2'-methylenebis (4, 6,-di-tertbutylphenyl) phosphate.
- 5
- 13) The heterophasic polypropylene composition according to any of the preceding claims having
- 10 a flexural modulus according to ISO178 in the range of 600 to 2500 MPa, preferably in the range of 700 to 2000 MPa, or 850 to 1700 MPa and optionally either or both of:
- a Charpy notched impact strength, ISO179, determined at +23 °C in the range of 5.0 to 100.0 kJ/m, preferably in the range of 15.0 to 90.0 kJ/m², or 20.0 to 80.0 kJ/m² and/or
- a Charpy notched impact strength, ISO179, determined at -20 °C, in the range of 1.0 to 20 kJ/m², preferably in the range of 2.0 to 15 kJ/m², or 2.5 to 12.0 kJ/m².
- 15
- 14) Moulded article, preferably injection moulded article comprising the heterophasic polypropylene composition according to any of the preceding claims.
- 15) Use of the heterophasic polypropylene composition for producing moulded articles,
- 20 preferably packaging articles.

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2021/056854

A. CLASSIFICATION OF SUBJECT MATTER
INV. C08F210/06 C08F4/6592 C08L23/06 C08F210/16
ADD.
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED
Minimum documentation searched (classification system followed by classification symbols)
C08F C08L

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

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2019/179959 A1 (BOREALIS AG [AT]) 26 September 2019 (2019-09-26) cited in the application Polymers, Polymerization; page 20 - page 21 examples	1-15
X	M. AARNIO-WINTERHOF ET AL: "Structure-property relations of heterophasic ethylene-propylene copolymers based on a single-site catalyst", EXPRESS POLYMER LETTERS, vol. 11, no. 2, 1 January 2017 (2017-01-01), pages 152-161, XP055618058, ISSN: 1788-618X, DOI: 10.3144/expresspolymlett.2017.16 table 1	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
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Date of the actual completion of the international search 16 July 2021	Date of mailing of the international search report 26/07/2021
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Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Authorized officer Thomas, Dominik
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INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2021/056854

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	<p>MANIKA VARMA-NAIR ET AL: "Quiescent Crystallization Kinetics of Nucleated Metallocene and ZN Isotactic Polypropylenes", JOURNAL OF THERMAL ANALYSIS AND CALORIMETRY, KLUWER ACADEMIC PUBLISHERS, DORDRECHT, NL, vol. 59, no. 1-2, 1 January 2000 (2000-01-01), pages 483-495, XP019253185, ISSN: 1572-8943 the whole document -----</p>	1-15

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/EP2021/056854

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO 2019179959	A1	26-09-2019	
		CN 112292406 A	29-01-2021
		EP 3768735 A1	27-01-2021
		KR 20200133264 A	26-11-2020
		US 2021017307 A1	21-01-2021
		WO 2019179959 A1	26-09-2019
