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(54) **LOW MELTING PP HOMOPOLYMER WITH HIGH CONTENT OF REGIOERRORS AND HIGH MOLECULAR WEIGHT**

PROPYLENHOMOPOLYMER MIT GERINGER SCHMELZTEMPERATUR MIT HOHEM GEHALT AN REGIODEFEKTN UND HOHEM MOLEKULARGEWICHT

HOMOPOLYMÈRE DE PROPYLÈNE À FAIBLE POINT DE FUSION À TENEUR ÉLEVÉE EN RÉGIO-DÉFAUTS ET MASSE MOLÉCULAIRE ÉLEVÉE

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(73) Proprietor: **Borealis AG
1220 Vienna (AT)**

(72) Inventors:

- RESCONI, Luigi
4501 Neuhofen an der Krems (AT)**
- REICHELT, Kristin
A-4501 Neuhofen/Krems (AT)**

(74) Representative: **Maiwald Patentanwalts GmbH**

**Elisenhof
Elisenstrasse 3
80335 München (DE)**

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Description

[0001] The present invention relates to a polypropylene homopolymer, a process for the preparation of such polypropylene homopolymer and a polypropylene blend comprising such polypropylene homopolymer as well as a pipe, film, sheet, fiber, moulded article or extrusion coating comprising such polypropylene homopolymer or polypropylene blend.

[0002] In practice, metallocene catalysts are used in the manufacture of polyolefins such as polyethylenes and polypropylenes. Countless academic and patent publications describe the use of these catalysts in olefin polymerisation. Metallocenes are now used industrially and polyethylenes and polypropylenes in particular are often produced using cyclopentadienyl based catalyst systems with different substitution patterns.

[0003] In particular, polypropylenes prepared by using an isospecific, C_2 -symmetric metallocene provide a different microstructure compared to polypropylenes prepared by using Ziegler-Natta (ZN) catalysts. The most significant difference is the presence of regio-defects in metallocene-made polypropylenes. These regio-defects can be of three different types, namely 2,1-erythro (2,1e), 2,1-threo (2,1t) and 3,1 defects. A detailed description of the structure and mechanism of formation of regio-defects in polypropylene can be found in Chemical Reviews 2000, 100(4), pages 1316-1327.

[0004] The presence of such regio-defects is a sufficient (although not necessary) feature to unambiguously identify a polypropylene as produced with a metallocene catalyst. The most common regio-defect, present in almost all isotactic polypropylenes produced with highly isospecific metallocene catalysts, is the 2,1e defect.

[0005] There are several documents describing polypropylenes with a high content of regio-defects. For example, in Organometallics 1996, 15, 5046 and JACS 1998, 120, 2308, it is referred to metallocene rac - $C_2H_4(4,7\text{-Me}_2\text{Ind})_2ZrCl_2$ producing polypropylene with 1.8 % regio-defects, a melting temperature of 131 °C, but a very low molecular weight of only 6,700. US 5,504,172 refers to an isotactic polypropylene having a content of 2,1e-regio-defects of 0.7 to 1.1 %. WO 2011/135004 A2 and WO 2011/135005 A2 describe a propylene homopolymer with a melting temperature of less than 147 °C, a percentage of 2,1 defects of at least 1 % and a xylene soluble fraction of less than 0.5 wt.-%. In particular, it is disclosed that the propylene homopolymer has a percentage of 2,1 defects of below 1.8 %.

[0006] It is known that, by introducing defects into the polymer chain, such as comonomers, stereoerrors or regio-defects, the physical properties of polypropylene can be modified. In particular, by increasing the amount of chain defects, crystallinity and melting point of polypropylene can be reduced whereupon a soft material can be obtained. These properties are advantageous for certain applications. Such chain defects can be introduced by copolymerization (introducing comonomer units in the chain) or by modifying the chemical structure of the catalyst (that is, modifying its selectivity) in order to allow more stereo- or regio-defects to be formed during polymerization.

[0007] In the case of regio-defects, the described propylene homopolymers have the disadvantage that an amount of 2,1 regio-defects of above 1.8 % in the polypropylene chain is not achieved and, furthermore, that already an increase in 2,1 regio-defects usually causes a significant decrease of the molecular weight of the propylene homopolymer.

[0008] Therefore, there is a continuous need for alternative propylene homopolymers having different polymer properties than existing propylene homopolymers, and especially having an increased amount of 2,1 regio-defects which in turn reduce the melting temperature of the polymer (T_m), while at the same time keeping the molecular weight of the propylene homopolymers on a high level.

[0009] Accordingly, it is an objective of the present invention to provide a new propylene homopolymer. Another objective is to provide a propylene homopolymer being prepared with a specific tailored catalyst system. A further objective of the present invention is to provide a propylene homopolymer having a high amount of 2,1 regio-defects and, especially, an amount of 2,1 regio-defects of above 2.0 %. Another objective of the present invention is to provide a propylene homopolymer having a low melting temperature T_m . A further objective of the present invention is to provide a propylene homopolymer having a low tensile modulus and a high tensile strain at break. A still further objective is to provide a propylene homopolymer which has a high molecular weight, as measured by its melt flow rate (MFR), intrinsic viscosity (IV) or gel permeation chromatography (GPC), and at the same time a low xylene soluble fraction at room temperature.

[0010] The foregoing and other objectives are solved by propylene homopolymer (H-PP1), having

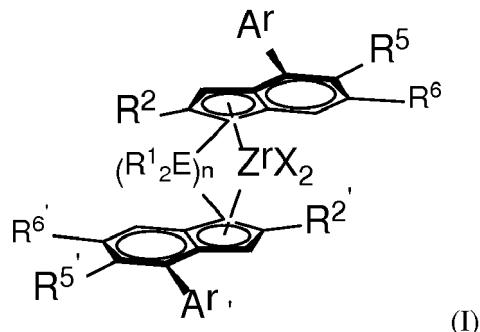
- a) a melting temperature T_m of less than 150 °C,
- b) 2,1 regio-defects of above 2.0 % determined by ^{13}C -NMR spectroscopy, and
- c) a weight average molecular weight M_w of above 100 kg/mol.

[0011] The inventors surprisingly found out that the foregoing propylene homopolymer (H-PP1) according to the present invention is obtained by preparing the propylene homopolymer (H-PP1) with a specific tailored catalyst system at a specific polymerization temperature. This specific tailored catalyst system enables a skilled person to produce a propylene homopolymer (H-PP1) featured by a high amount of 2,1 regio-defects and a low melting temperature T_m , while keeping the molecular weight of the propylene homopolymer (H-PP1) still at a high level.

[0012] According to another aspect of the present invention, a process for the preparation of such a propylene homopol-

5 ymer (H-PP1) is provided, the process comprising the step of polymerizing propylene at a polymerization temperature of less than 60°C, preferably not more than 55 °C, in the presence of a solid catalyst system, said solid catalyst system comprises

10 (i) a complex of formula (I)



20 wherein

25 each X is a sigma ligand;

30 $R^{12}E$ is a divalent group selected from $-R^{12}C-$, $-R^{12}Si-$, $-R^{12}Ge-$, wherein each R^1 is independently a hydrogen atom, C_1-C_{20} hydrocarbyl, tri(C_{1-20} -alkyl)silyl, C_6-C_{20} aryl, C_7-C_{20} arylalkyl or C_7-C_{20} alkylaryl, and n has a value from 1 to 2;

35 R^2 and $R^{2'}$, which can be the same or different, are a linear C_1-C_{20} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC);

40 Ar and Ar' which can be the same or different, are an aryl or heteroaryl group having up to 20 carbon atoms;

45 R^5 is hydrogen or a C_1-C_{40} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC);

50 $R^{5'}$ is a C_{1-40} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC); R^6 and $R^{6'}$, which can be the same or different, are a tertiary C_{4-20} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC);

55 (ii) a cocatalyst comprising a first organometallic compound of a Group 13 metal (IUPAC); and

60 (iii) optionally a second organometallic compound of a Group 13 metal (IUPAC) being different to the first organometallic compound.

[0013] It is preferred that R^2 and $R^{2'}$, which can be the same or different, are a linear C_{1-10} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16; Ar and Ar' which can be the same or different, are an aryl or heteroaryl group having up to 20 carbon atoms; R^5 is hydrogen or a C_1-C_{20} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC); $R^{5'}$ is a C_1-C_{20} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC);

65 R^6 and $R^{6'}$, which can be the same or different, are a tertiary C_4-C_{20} hydrocarbyl radical. It is also preferred that $R^{12}E$ is $-R^{12}Si-$, wherein each R^1 is independently a C_1-C_{50} hydrocarbyl, and n has a value of 1; each R^2 and $R^{2'}$ is a methyl radical; R^5 is hydrogen or a C_1-C_{10} hydrocarbyl radical containing one or more heteroatoms from group 16 (IUPAC); $R^{5'}$ is a C_1-C_{10} hydrocarbyl group containing one or more heteroatoms from group 16; each R^6 and $R^{6'}$ is a tertiary butyl radical.

[0014] It is also preferred that the polymerization temperature in the process for producing the propylene homopolymer (H-PP1) is from 10 to less than 60 °C, e.g. from 10 to 55 °C, more preferably from 15 to 50 °C, still more preferably from 15 to 45 °C, yet more preferably from 20 to 40°C and most preferably from 25 to 35 °C.

[0015] According to a further aspect of the present invention, a polypropylene blend (PB) is provided, the polypropylene blend (PB) comprises a propylene homopolymer (H-PP1) as defined in the instant invention and a second propylene homopolymer (H-PP2) having a melting temperature T_m at least 5 °C higher, preferably 8 °C higher, even more preferably at least 10°C higher than the propylene homopolymer (H-PP1).

[0016] According to a still further aspect of the present invention, a pipe, film, sheet, fiber, moulded article or extrusion coating is provided, comprising the propylene homopolymer (H-PP1) or the polypropylene blend (PB). It is preferred that the film is a cast, blown or biaxially oriented polypropylene (BOPP) film.

[0017] Advantageous embodiments of the present invention are defined in the corresponding subclaims.

The propylene homopolymer (H-PP1)

[0018] According to one embodiment of the present invention, the propylene homopolymer (H-PP1) has

5 a)

a1) a melting temperature T_m lower than 145 °C, preferably lower than 142 °C, more preferably lower than 140 °C,
 a2) 2,1 regio-defects, more preferably, 2,1 erythro regio-defects, of above 2.0 %, preferably above 2.1 %, more
 10 preferably above 2.2 %, determined by ^{13}C -NMR spectroscopy, and
 a3) a weight average molecular weight M_w of above 100 kg/m, preferably of above 200 kg/mol, more preferably
 above 300 kg/mol;
 or

15 b)

b1) a melting temperature T_m lower than 145 °C, preferably lower than 142 °C, more preferably lower than 140 °C,
 b2) 2,1 regio-defects, more preferably, 2,1 erythro regio-defects, of above 2.0 %, preferably above 2.1 %, more
 20 preferably above 2.2 %, determined by ^{13}C -NMR spectroscopy,
 b3) a weight average molecular weight M_w of above 100 kg/m, preferably of above 200 kg/mol, more preferably
 above 300 kg/mol, and
 b4) a xylene soluble (XS) fraction content of below 2.0 wt.-%, preferably below 1.0 wt.-%, more preferably below
 25 0.5 wt.-%;
 or

c)

c1) a melting temperature T_m lower than 145 °C, preferably lower than 142 °C, more preferably lower than 140 °C,
 c2) 2,1 regio-defects, more preferably, 2,1 erythro regio-defects, of above 2.0 %, preferably above 2.1 %, more
 30 preferably above 2.2 %, determined by ^{13}C -NMR spectroscopy,
 c3) stereo-defects (measured as mrrm pentad content) determined by ^{13}C -NMR spectroscopy lower than 1.0
 %, preferably lower than 0.8 %, more preferably lower than 0.6 %, and
 c4) a weight average molecular weight M_w of above 100 kg/m, preferably of above 200 kg/mol, more preferably
 above 300 kg/mol;
 or

35 d)

d1) a melting temperature T_m lower than 145 °C, preferably lower than 142 °C, more preferably lower than 140 °C,
 d2) 2,1 regio-defects, more preferably, 2,1 erythro regio-defects, of above 2.0 %, preferably above 2.1 %, more
 40 preferably above 2.2 %, determined by ^{13}C -NMR spectroscopy,
 d3) stereo-defects (measured as mrrm pentad content) determined by ^{13}C -NMR spectroscopy spectroscopy
 lower than 1.0 %, preferably lower than 0.8 %, more preferably lower than 0.6 %,
 d4) a weight average molecular weight M_w of above 100 kg/m, preferably of above 200 kg/mol, more preferably
 above 300 kg/mol, and
 45 d5) a xylene soluble (XS) fraction content of below 2.0 wt.-%, preferably below 1.0 wt.-%, more preferably below
 0.5 wt.-%.

[0019] It should be understood that for the purposes of the present invention, the following terms have the following meaning:

50 The term "hydrocarbyl" radical in the meaning of the present invention refers preferably to alkyl, alkenyl, alkynyl, cycloalkyl,
 cycloalkenyl, aryl groups, alkylaryl groups or arylalkyl groups or of course mixtures of these groups such as cycloalkyl
 substituted by alkyl. Preferred hydrocarbyl radicals are methyl, ethyl, propyl, isopropyl, tert.-butyl, isobutyl, $C_5\text{-}C_6$ cy-
 cloalkyl or cyclohexylmethyl radicals.

[0020] The term " $C_1\text{-}C_{40}$ hydrocarbyl" radical therefore includes $C_1\text{-}C_{40}$ alkyl, $C_2\text{-}C_{40}$ alkenyl, $C_2\text{-}C_{40}$ alkynyl, $C_3\text{-}C_{40}$
 cycloalkyl, $C_3\text{-}C_{40}$ cycloalkenyl, $C_6\text{-}C_{40}$ aryl groups, $C_7\text{-}C_{40}$ alkylaryl groups or $C_7\text{-}C_{40}$ arylalkyl groups or of course
 mixtures of these groups such as cycloalkyl substituted by alkyl.

[0021] The term " $C_1\text{-}C_{20}$ hydrocarbyl" radical therefore includes $C_1\text{-}C_{20}$ alkyl, $C_2\text{-}C_{20}$ alkenyl, $C_2\text{-}C_{20}$ alkynyl, $C_3\text{-}C_{20}$
 cycloalkyl, $C_3\text{-}C_{20}$ cycloalkenyl, $C_6\text{-}C_{20}$ aryl groups, $C_7\text{-}C_{20}$ alkylaryl groups or $C_7\text{-}C_{20}$ arylalkyl groups or of course

mixtures of these groups such as cycloalkyl substituted by alkyl.

[0022] The term "C₁-C₁₀ hydrocarbyl" radical therefore includes C₁-C₁₀ alkyl, C₂-C₁₀ alkenyl, C₂-C₁₀ alkynyl, C₃-C₁₀ cycloalkyl, C₃-C₁₀ cycloalkenyl, C₆-C₁₀ aryl groups, C₇-C₁₀ alkylaryl groups or C₇-C₁₀ arylalkyl groups or of course mixtures of these groups such as cycloalkyl substituted by alkyl.

[0023] The term "C₁-C₅ hydrocarbyl" radical therefore includes C₁-C₅ alkyl, C₂-C₅ alkenyl, C₂-C₅ alkynyl, C₃-C₅ cycloalkyl, C₃-C₅ cycloalkenyl or of course mixtures of these groups such as cycloalkyl substituted by alkyl.

[0024] The term "aryl" means a preferably monocyclic aromatic ring structure. Preferably, the aryl group is a phenyl group.

[0025] The term "heteroaryl" means a preferably monocyclic aromatic ring structure comprising at least one heteroatom.

10 Preferred heteroaryl groups have 1 to 4 heteroatoms selected from O, S and N. Preferred heteroaryl groups include furanyl, thiophenyl, oxazole, thiazole, isothiazole, isooxazole, triazole and pyridyl.

[0026] Any group including "one or more heteroatoms from groups 14 to 16 (IUPAC)" preferably means Si, O, S or N. N groups may present as -NH- or -NR"- where R" is C₁-C₁₀ alkyl. In one embodiment of the present invention, the one or more heteroatoms from groups 14 to 16 (IUPAC) is a heteroatom from group 16, preferably O. There may, for example, be 1 to 4 heteroatoms.

[0027] The term "2,1 regio defects" defines the sum of 2,1 erythro regio-defects and 2,1 threo regio-defects.

[0028] The term "regio defects" defines the sum of 2,1 erythro regio-defects, 2,1 threo regio-defects and 3,1 regio-defects.

[0029] Wherever in the instant invention the amount of defects, i.e. regio defects, like 2,1 regio defects, i.e. 2,1 erythro regio-defects, 2,1 threo regio-defects and 3,1 regio-defects, and stereo-defects, are indicated by "%" the average percentage of propylene units in the polymer chain is meant.

[0030] Where the term "comprising" is used in the present description and claims, it does not exclude other non-specified elements of major or minor functional importance. For the purposes of the present invention, the term "consisting of" is considered to be a preferred embodiment of the term "comprising of". If hereinafter a group is defined to comprise at least a certain number of embodiments, this is also to be understood to disclose a group, which preferably consists only of these embodiments.

[0031] Whenever the terms "including" or "having" are used, these terms are meant to be equivalent to "comprising" as defined above.

[0032] Where an indefinite or definite article is used when referring to a singular noun, e.g. "a", "an" or "the", this includes a plural of that noun unless something else is specifically stated.

[0033] In the following, it is referred to further details of the present invention.

[0034] It is important to note that there exists a crucial difference in the chain-microstructure between polypropylenes produced by a metallocene catalyst and a Ziegler-Natta catalyst. The polymer chains of metallocene-made polypropylene may contain either stereo- or regio-defects, or both, whereas the chains of Ziegler-Natta based polypropylenes has only stereo defects. As the propylene homopolymer according to this invention is produced by a metallocene catalyst it has preferably a high amount of 2,1 regio-defects, for example a high amount of 2,1-erythro regio-defects.

[0035] Thus, the present invention relates to novel propylene homopolymers (H-PP1) which have high amounts of 2,1-regio-defects, especially high 2,1-erythro regio-defects, low melting temperatures and high molecular weight. These polymers can be obtained by polymerisation of propylene at a temperature and in the presence of certain catalysts as described in detail herein.

[0036] In particular, the invention relates to a polypropylene homopolymer (H-PP1) having

- a) a melting temperature T_m of less than 145 °C,
- b) 2,1-regio-defects, preferably 2,1 erythro regio-defects, of above 2.0 % determined by ¹³C-NMR spectroscopy, and
- c) a weight average molecular weight M_w of above 100 kg/mol.

[0037] Accordingly, the propylene homopolymer (H-PP1) can be defined by the melting temperature (T_m). Thus, it is appreciated that the propylene homopolymer (H-PP1) of the instant invention has a melting temperature (T_m) measured by differential scanning calorimetry (DSC) of below 145 C. In one embodiment of the present invention, the propylene homopolymer (H-PP1) of the instant invention has a melting temperature (T_m) measured by differential scanning calorimetry (DSC) of less than 143°C, preferably of less than 140°C, more preferably in the range of from 120 to 143 °C, still more preferably in the range of 120 to 142.5 °C, still yet more preferably in the range of 120 to 140 °C, like in the range of 120 to 138 °C. For example, the propylene homopolymer (H-PP1) of the instant invention has a melting temperature (T_m) measured by differential scanning calorimetry (DSC) in the range of from 130 to 142.5°C or 130 to 140 °C.

[0038] Additionally, the inventive propylene homopolymer (H-PP1) is featured by a rather high concentration of 2,1 regio-defects, like 2,1-erythro regio-defects, compared to known metallocene based polypropylenes. In one embodiment of the present invention, it is preferred that the propylene homopolymer (H-PP1) of the instant invention has 2,1 regio-defects, like 2,1 erythro regio-defects, of above 2.0 %, preferably of above 2.1 %, more preferably of above 2.2 %, yet

more preferably from 2.0 to 5.0 %, still more preferably from 2.1 to 4.5 %, still yet more preferably from 2.2 to 4.0 %, like 2.5 to 3.8 %, determined by ^{13}C -NMR spectroscopy. For example, the propylene homopolymer (H-PP1) of the instant invention has 2,1 regio-defects, like 2,1 erythro regio-defects, of from 2.0 to 3.5 %, like 2.2 to 3.5 %, determined by ^{13}C -NMR spectroscopy. In one embodiment the amount of 2,1 regio-defects and 2,1 erythro regio-defects are the same and specified as indicated in this paragraph.

[0039] It is further appreciated that the propylene homopolymer (H-PP1) of the instant invention has a rather low amount of 3,1 regio-defects, i.e. equal or less than 0.6 %, more preferably equal or less than 0.5 %, still more preferably equal or less than 0.4 %, as determined by ^{13}C -NMR spectroscopy.

[0040] Accordingly, the propylene homopolymer (H-PP1) of the instant invention can be additionally or alternatively defined by its total amount of regio defects, i.e. by its sum of 2,1 erythro regio-defects, 2,1 threo regio-defects and 3,1 regio-defects. In one embodiment of the present invention, the propylene homopolymer (H-PP1) has a total amount of regio defects of above 2.0 %, preferably of above 2.1 %, more preferably of above 2.2 %, yet more preferably from 2.0 to 5.0 %, still more preferably from 2.1 to 4.5 %, still yet more preferably from 2.2 to 4.0 %, like 2.5 to 3.8 %, determined by ^{13}C -NMR spectroscopy.

[0041] The propylene homopolymer (H-PP1) of the instant invention can, additionally to the regio-defects, like the 2,1 regio-defects, also be defined by its stereoregularity, i.e. by its isotacticity. Thus, it is preferred that the propylene homopolymer (H-PP1) has a rather low mrrm pentad content (stereo-defects), i.e. equal or lower than 1.0 %, more preferably equal or lower than 0.8 %, still more preferably equal or lower than 0.6 %, determined by ^{13}C -NMR spectroscopy.

[0042] Thus, additionally or alternatively, the propylene homopolymer (H-PP1) of the instant invention can be defined by its total amount of defects in the polymer chain, i.e. by its sum of 2,1 erythro regio-defects, 2,1 threo regio-defects, 3,1 region-defects and mrrm pentad content. In one embodiment of the present invention, the propylene homopolymer (H-PP1) has a total amount of defects of above 2.0 %, preferably of above 2.1 %, more preferably of above 2.2 %, yet more preferably from 2.0 to 5.0 %, still more preferably from 2.1 to 4.5 %, still yet more preferably from 2.2 to 4.0 %, like 2.5 to 3.8 %, determined by ^{13}C -NMR spectroscopy.

[0043] A further requirement of the propylene homopolymer (H-PP1) of the instant invention is that the propylene homopolymer (H-PP1) has a high weight average molecular weight M_w . In particular, the propylene homopolymer (H-PP1) of the instant invention has a weight average molecular weight M_w of above 100 kg/mol. In one embodiment of the present invention, the propylene homopolymer (H-PP1) has weight average molecular weight M_w of above 200 kg/mol, preferably of above 300 kg/mol, like of above 1,000 kg/mol, and most preferably of from 100 to 2,000 kg/mol. For example, the propylene homopolymer (H-PP1) of the instant invention has weight average molecular weight M_w of from 100 to 2,000 kg/mol, preferably from 200 to 1,500 kg/mol, more preferably from 300 to 1,500 kg/mol.

[0044] The number average molecular weight (M_n) of the propylene homopolymer (H-PP1) is preferably in the range of 50 to 1,000 kg/mol, more preferably from 60 to 700 kg/mol.

[0045] A broad molecular weight distribution (MWD) improves the processability of the polypropylene. Accordingly, it is appreciated that the polydispersity index (M_w/M_n) of the propylene homopolymer (H-PP1) is at least 2.0, preferably at least 2.2 and most preferably at least 2.4. On the other hand the molecular weight distribution (MWD) should be not too broad. Therefore, it is preferred that the molecular weight distribution (MWD) of the propylene homopolymer (H-PP1) is less than 30.0, preferably less than 25.0 and most preferably less than 20.0. For example, the molecular weight distribution (MWD) of the propylene homopolymer (H-PP1) is from 2.0 to 30.0, preferably from 2.2 to 25.0 and most preferably from 2.4 to 20.0.

[0046] Additionally or alternatively, the propylene homopolymer (H-PP1) of the instant invention is featured by rather low content of the xylene soluble fraction (XS), i.e. by a xylene soluble fraction (XS) content of equal or below 2.0 wt.-%, more preferably of equal or below 1.5 wt.-%, yet more preferably equal or below 1.0 wt.-%, like equal or below 0.5 wt.-% based on the total weight of the propylene homopolymer (H-PP1). For example, the propylene homopolymer (H-PP1) of the instant invention has a xylene soluble fraction (XS) content in the range of from 0.05 to 2.0 wt.-%, more preferably in the range of from 0.1 to 1.5 wt.-%, yet more preferably in the range of from 0.1 to 1.0 wt.-%, like in the range of from 0.1 to 0.5 wt.-%, based on the total weight of the propylene homopolymer (H-PP1).

[0047] Additionally or alternatively, the propylene homopolymer (H-PP1) of the instant invention is featured by a rather low tensile modulus and a rather high tensile strain at break. The tensile modulus is preferably of equal or below 1500 MPa, more preferable of equal or below 1300 MPa, still more preferably of equal or below 1200 MPa. For example the propylene homopolymer (H-PP1) of the instant invention has a tensile modulus in the range from 800 to 1500 MPa, more preferably in the range from 900 to 1300 MPa, yet more preferably in the range from 1000 to 1200 MPa, like in the range from 1100 to 1180 MPa. The tensile strain at break is preferably equal or higher than 150 %, more preferably higher than 250 %, yet more preferable higher than 400 %. For example the propylene homopolymer (H-PP1) of the instant invention has a tensile strain at break in the range from 150 to 800 %, more preferably in the range of 250 to 750 %, yet more preferably in the range of 350 to 700 %, like in the range from 400 to 650 %. A propylene homopolymer featured by a rather low tensile modulus and a rather high tensile strain at break, e.g. the propylene homopolymer (H-

PP1) of the instant invention, represents a rather soft material which is advantageous for various applications.

[0048] The propylene homopolymer (H-PP1) of the instant invention does not define a composition of different polymers. Accordingly, the propylene homopolymer may comprise further additives but no other polymer components than the propylene homopolymer.

[0049] It is appreciated that the propylene homopolymer (H-PP1) of the instant invention may comprise standard polymer additives well known in the art. For example, the propylene homopolymer (H-PP1) comprises additives selected from the group consisting of UV stabilisers, antioxidants, pigments, fillers and the like.

[0050] In one embodiment of the present invention, the propylene homopolymer (H-PP1) of the instant invention is nucleated, i.e. α -nucleated.

[0051] Nucleation increases the crystallization temperature significantly, e.g. by up to 10 °C. Accordingly, a propylene homopolymer (H-PP1) having low melting temperature T_m but high crystallisation temperature T_c can be formed. The increase in melting temperature T_m is much less, around 2 °C. Thus, melting temperature T_m of nucleated propylene homopolymer (H-PP1) may be a bit higher than corresponding non-nucleated propylene homopolymer (H-PP1). The melting temperature T_m of the propylene homopolymer (H-PP1) of the instant invention is thus preferably determined in the absence of a nucleating agent, like an α -nucleating agent, if not otherwise indicated.

[0052] In one embodiment of the present invention, the propylene homopolymer (H-PP1) being nucleated, like α -nucleated, has a melting temperature T_m of less than 150 °C, preferably less than 145 °C.

[0053] Preferably, the mrrm pentad content of such a nucleated, like α -nucleated, propylene homopolymer (H-PP1) is less than 0.5 %, more preferably of less than 0.3 % and most preferably of less than 0.2 %, determined by ^{13}C -NMR spectroscopy.

[0054] In one embodiment of the present invention, a nucleation, i.e. α -nucleation, of the propylene homopolymer (H-PP1) is preferred. This may also improve gloss and transparency. Suitable α -nucleating agents are those commonly used in the art and listed for example in chapter 2 of the review by Gahleitner et al. in International Polymer Processing 26 (2011) p. 2-20.

[0055] Especially suitable are inorganic α -nucleating agents such as talc. Talc can be added in an amount of 0.05 to 30.00 wt.-%, based on the total weight of the propylene homopolymer (H-PP1). Other nucleating agents are organic particulate α -nucleating agents such as carboxylic or phosphoric acid salts with a substituted or un- substituted aromatic ring. These can be added in an amount of 0.01 to 1.0 wt.-%, based on the total weight of the propylene homopolymer (H-PP1). Other α -nucleating agents are polymeric nucleating agents such as poly(tetrafluoro ethylene) or isotactic poly(vinyl cyclohexane). These can be added in an amount of 0.001 to 0.20 wt.-%, based on the total weight of the propylene homopolymer (H-PP1). Other α -nucleating agents are soluble organic nucleating agents such as sorbitol derivatives, nonitol derivatives or aliphatic substituted trisamides. These can be added in an amount of 0.05 to 1.0 wt.-%, based on the total weight of the propylene homopolymer (H-PP1).

[0056] Typical examples for suitable organic particulate α -nucleating agents are sodium benzoate, sodium 2,2'-methylen-bis-(4,6-di-tert-butylphenyl) phosphate (CAS No. 85209-91-2, trade name Adekastab NA-11, commercially available from Adeka Palmarole, France), a mixture consisting of 60 wt.-% Hydroxybis (2,4,8,10- tetra-tert. butyl-6-hydroxy-12H-dibenzo(d,g)(1,3,2) dioxaphosphocin 6-oxidato) aluminium (CAS No. 151841-65-5) and 40 wt.-% Li-myristate (CAS No. 20336-96- 3) (trade name Adekastab NA-21, commercially available from Adeka Palmarole, France), the disodium salt of Cis-endobicyclo[2.2.1]heptane-2,3-dicarboxylic acid (CAS No. 351870-33- 2; trade name Hyperform HPN-68, commercially available from Milliken Inc., USA). A typical examples for suitable polymeric nucleating agents is isotactic poly(vinyl cyclohexane) (CAS No. 25498-06-0).

[0057] Typical examples for suitable soluble organic α -nucleating agents are 1,3:2,4-Bis(3,4-dimethylbenzylidene)sorbitol (CAS No. 135861-56-2; trade name Millad 3988, commercially available from Milliken Inc., USA), 1,2,3-trideoxy-4,6:5,7-bis-0[(4-propylphenyl)methylene] -nonitol (CAS No. 882073-43-0; trade name Millad NX8000, commercially available from Milliken Inc., USA) and N,N',N"-tris-tert- Butyl-1,3,5-benzenetricarboxamide (CAS No. 745070-61-5; trade name Irgaclear XT386, commercially available from BASF AG, Germany).

[0058] In one embodiment of the present invention, the propylene homopolymer (H-PP1) is free of nucleating agents, in particular free of β -nucleating agents and/or α -nucleating agents. In another embodiment the propylene homopolymer (H-PP1) is free of β -nucleating agents but contains α -nucleating agents.

[0059] The propylene homopolymer (H-PP1) of the instant invention is obtainable, preferably obtained, by a process for the preparation of a propylene homopolymer (H-PP1) in the presence of specific catalysts as described in detail below.

The second propylene homopolymer (H-PP2)

[0060] As mentioned above the present invention is additionally directed to a polypropylene blend (PB) which contains in addition to the propylene homopolymer (H-PP1) a second propylene homopolymer (H-PP2). The second propylene homopolymer (H-PP2) can be any propylene homopolymer as long as it has a higher melting temperature compared to the propylene homopolymer (H-PP1). Accordingly the second propylene homopolymer (H-PP2) has a melting temper-

ature T_m at least 5 °C higher, preferably 8 °C higher, even more preferably at least 10°C higher than the propylene homopolymer (H-PP1) used in the instant polypropylene blend (PB).

[0061] Accordingly, the second propylene homopolymer (H-PP2) has a melting temperature (T_m) measured by differential scanning calorimetry (DSC) of at least 140 C. In one embodiment of the present invention, the second propylene homopolymer (H-PP2) has a melting temperature (T_m) of at least 143 °C, preferably of at least 145 °C, more preferably in the range of from 140 to 168 °C, yet more preferably in the range of from 143 to 165 °C, still more preferably in the range of from 145 to 163 °C, like in the range of 150 to 163 °C.

[0062] Additionally, the second propylene homopolymer (H-PP2) is featured by a rather low concentration of 2,1 regio-defects, like 2,1-erythro regio-defects, compared to the propylene homopolymer (H-PP1). Thus it is appreciated that the second propylene homopolymer (H-PP2) has 2,1 regio-defects, like 2,1 erythro regio-defects, being at least 0.8 % lower, more preferably 0.9 %, yet more preferably 1.0 % lower, than the 2,1 regio-defects, like 2,1-erythro regio-defects, of the propylene homopolymer (H-PP1). In one preferred embodiment the second propylene homopolymer (H-PP2) has 2,1 regio-defects, like 2,1-erythro regio-defects, equal or below 1.8 %, more preferably of equal or below 1.6 %, still more preferably equal or below 1.4 %. For example, the second propylene homopolymer (H-PP2) has 2,1 regio-defects, like 2,1 erythro regio-defects, of from 1.1 to equal or below 1.8 %, like of from 1.1 to equal or below 1.6 %.

[0063] The second propylene homopolymer (H-PP2) can be additionally or alternatively defined by its total amount of regio defects, i.e. by its sum of 2,1 erythro regio-defects, 2,1 threo regio-defects and 3,1 regio-defects. In one embodiment of the present invention, the second propylene homopolymer (H-PP2) has a total amount of regio defects of equal or below 1.8 %, more preferably of equal or below 1.6 %, still more preferably equal or below 1.4 %, yet more preferably of from 1.1 to equal or below 1.8 %, like of from 1.1 to equal or below 1.6 %.

[0064] Further it is appreciated that the polydispersity index (Mw/Mn) of the second propylene homopolymer (H-PP2) is at least 2.0, more preferably at least 2.2, still more preferably from 2.0 to 30.0, yet more preferably from 2.2 to 25.0.

[0065] The preparation of the second propylene homopolymer (H-PP2) is in the skilled knowledge. For instance the second propylene homopolymer (H-PP2) can be produced in the presence of a Ziegler-Natta catalyst or a metallocene catalyst.

The polypropylene blend (PB)

[0066] A further aspect of the instant invention is a polypropylene blend (PB) comprising the propylene homopolymer (H-PP1) and the second propylene homopolymer (H-PP2). Preferably the polypropylene blend (PB) contains as polymer component only the propylene homopolymer (H-PP1) and the second propylene homopolymer (H-PP2). Accordingly the polypropylene blend (PB) may contain additives in an amount up to 10 wt.-%, more preferably up to 5 wt.-%, based on the total amount of the polypropylene blend (PB). Preferably the weight ratio between the propylene homopolymer (H-PP1) and the second propylene homopolymer (H-PP2) is 90/10 to 10/90, more preferably 80/20 to 20/80, yet more preferably from 70/30 to 30/70.

[0067] Preferably, the polypropylene blend (PB) has a melting temperature (T_m) in the range of from 135 to 160 °C, more preferably in the range of from 140 to 155 °C.

[0068] The polypropylene comprising the two propylene homopolymers (H-PP1) and (H-PP2) shows a broader melting range compared to its individual fractions, and thus broadens the processability window in e.g. film manufacturing.

[0069] Additionally, the polypropylene blend (PB) preferably is featured by 2,1 regio-defects, like 2,1 erythro regio-defects, from 0.1 to 4.7 %, more preferably from 0.5 to 4.0.5 %, like in the range of 1.0 to 3.0 %.

[0070] The polypropylene blend (PB) can be additionally or alternatively defined by its total amount of regio defects, i.e. by its sum of 2,1 erythro regio-defects, 2,1 threo regio-defects and 3,1 regio-defects. In one embodiment the polypropylene blend (PB) has a total amount of regio defects from 0.1 to 4.7 %, more preferably from 0.5 to 4.0.5 %, like in the range of 1.0 to 3.0 %.

[0071] Preferably the polypropylene blend (PB) has a weight average molecular weight M_w of above 50 kg/mol, more preferably of above 100 kg/mol, still more preferably of from 50 to 2,000 kg/mol, still yet more preferably of from 100 to 1,500 kg/mol, like from 300 to 1,500 kg/mol.

[0072] Further it is appreciated that the polydispersity index (Mw/Mn) of the polypropylene blend (PB) is at least 2.0, more preferably at least 2.2, still more preferably from 2.0 to 30.0, yet more preferably from 2.2 to 25.0.

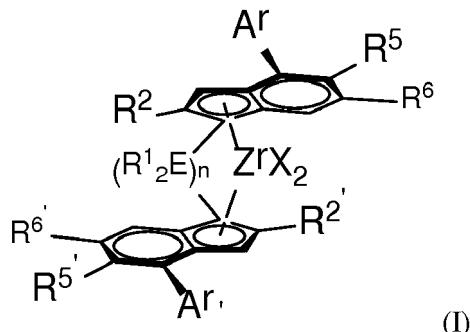
[0073] The preparation of the polypropylene blend (PB) is in the skilled knowledge. For instance the polypropylene blend (PB) can be obtained by mixing the propylene homopolymer (H-PP1) with the second propylene homopolymer (H-PP2) in an extruder or by producing the polypropylene blend (PB) in a sequential polymerization process as known by the skilled person.

Preparation the propylene homopolymer (H-PP1)

The solid catalyst system

5 [0074] The propylene homopolymer (H-PP1) of this invention has been preferably prepared in the presence of a metallocene catalyst, like a metallocene catalyst of formula (I). Accordingly, the process for the preparation of the instant propylene homopolymer (H-PP1) comprises the step of polymerizing propylene in the presence of a solid catalyst system, said solid catalyst system comprises

10 (i) a complex of formula (I)



25 wherein

each X is a sigma ligand;

R_1^2E is a divalent group selected from $-R_1^2C-$, $-R_1^2Si-$, $-R_1^2Ge-$, wherein each R^1 is independently a hydrogen atom, C_{1-20} -hydrocarbyl, tri(C_{1-20} -alkyl)silyl, C_{6-20} -aryl, C_{7-20} -arylalkyl or C_{7-20} -alkylaryl, and n has a value from 1 to 2;

30 R^2 and $R^{2'}$, which can be the same or different, are a linear C_{1-20} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC);

Ar and Ar' which can be the same or different, are an aryl or heteroaryl group having up to 20 carbon atoms;

R^5 is hydrogen or a C_{1-40} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC);

35 $R^{5'}$ is a C_{1-40} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC); R^6 and $R^{6'}$, which can be the same or different, are a branched C_{3-20} hydrocarbyl radical;

40 (ii) a cocatalyst comprising a first organometallic compound of a Group 13 metal (IUPAC); and

(iii) optionally a second organometallic compound of a Group 13 metal (IUPAC) being different to the first organometallic compound.

[0075] In one embodiment of the present invention, the substituents of the two multicyclic ligands making up the complex of formula (I) are identical.

[0076] Alternatively, it is preferred that the substituents of the two multicyclic ligands making up the complex of formula (I) are different. For example, it is preferred that at least one substituent on one multicyclic ligand differs from the corresponding substituent on the other multicyclic ligand. For example, one substituent on one multicyclic ligand differs from the corresponding substituent on the other multicyclic ligand. Alternatively, two substituents on one multicyclic ligand differ from the corresponding substituents on the other multicyclic ligand.

[0077] In one embodiment of the present invention, it is appreciated that substituent R^5 on one multicyclic ligand differs from the corresponding $R^{5'}$ substituent on the other multicyclic ligand. Additionally or alternatively, it is appreciated that substituent Ar on one multicyclic ligand differs from the corresponding Ar' substituent on the other multicyclic ligand.

[0078] In one embodiment of the present invention, substituents R^5 and Ar on one multicyclic ligand differ from the corresponding $R^{5'}$ and Ar' substituents on the other multicyclic ligand.

[0079] Preferably, the metallocene catalysts of the present invention are in their racemic (rac) or racemic-anti- form.

[0080] For the purpose of the present invention the term "racemic(rac) form" means that the like substituents of the two multicyclic ligands are on opposite sides with respect to the plane containing the zirconium and the centre of the cyclopentadienyl ring of the ligands, "anti-racemic form" means that the bulkier substituents of the two multicyclic ligands on the metallocene compound are on opposite sides with respect to the plane containing the zirconium and the centre

of the cyclopentadienyl ring of said multicyclic ligands, as described for example in EP 1 117 334.4.

[0081] In one embodiment of the present invention, each X, which may be the same or different, is a hydrogen atom, a halogen atom, a R, OR, OSO₂CF₃, OCOR, SR, NR₂ or PR₂ radical wherein R is a linear or branched, cyclic or acyclic, C₁-C₄₀ alkyl, C₂-C₄₀ alkenyl, C₂-C₄₀ alkynyl, C₆-C₄₀ aryl, C₇-C₄₀ alkylaryl or C₇-C₄₀ arylalkyl radical; optionally containing one or more heteroatoms from groups 14 to 16. For example, each X, which may be the same or different, is a hydrogen atom, a halogen atom, a R, OR, OSO₂CF₃, OCOR, SR, NR₂ or PR₂ group wherein R is a linear or branched, cyclic or acyclic, C₁-C₂₀ alkyl, C₂-C₂₀ alkenyl, C₂-C₂₀ alkynyl, C₆-C₂₀ aryl, C₇-C₂₀ alkylaryl or C₇-C₂₀ arylalkyl radical; optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC). For example, R is preferably a C₁-C₆ alkyl, phenyl or benzyl group.

[0082] In one embodiment of the present invention, each X, which are the same, is a hydrogen atom, a halogen atom, a R, OR, OSO₂CF₃, OCOR, SR, NR₂ or PR₂ radical wherein R is a linear or branched, cyclic or acyclic, C₁-C₄₀ alkyl, C₂-C₄₀ alkenyl, C₂-C₄₀ alkynyl, C₆-C₄₀ aryl, C₇-C₄₀ alkylaryl or C₇-C₄₀ arylalkyl radical; optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC). For example, R is preferably a C₁-C₆ alkyl, phenyl or benzyl radical.

[0083] In one embodiment of the present invention, each X is independently a hydrogen atom, a halogen atom, C₁-C₆ alkoxy group or an R group, e.g. preferably a C₁-C₆ alkyl, phenyl or benzyl group. Most preferably X is chlorine or a methyl radical. Preferably each X radical is the same. For example, both X radicals, which are the same, are methyl.

[0084] R₁₂E is a divalent bridge selected from -R₁₂C-, -R₁₂Si-, -R₁₂Ge-, wherein each R¹ is independently a hydrogen atom, C₁-C₂₀ hydrocarbyl, tri(C₁-C₂₀-alkyl)silyl, C₆-C₂₀ aryl, C₇-C₂₀ arylalkyl or C₇-C₂₀ alkylaryl, and n has a value from 1 to 2.

[0085] In one embodiment of the present invention, R₁₂E is a divalent group selected from -R₁₂C-, -R₁₂Si-, wherein each R¹ is independently a hydrogen atom, C₁-C₂₀-hydrocarbyl, tri(C₁-C₂₀-alkyl)silyl, C₆-C₂₀ aryl, C₇-C₂₀ arylalkyl or C₇-C₂₀ alkylaryl, and n has a value from 1 to 2. For example, R₁₂E is -R₁₂Si-, wherein each R¹ is independently a hydrogen atom, C₁-C₂₀ hydrocarbyl, tri(C₁-C₂₀-alkyl)silyl, C₆-C₂₀ aryl, C₇-C₂₀ arylalkyl or C₇-C₂₀ alkylaryl, and n has a value from 1 to 2.

[0086] In one embodiment of the present invention, R₁₂E is -R₁₂Si-, wherein each R¹ is independently a hydrogen atom, C₁-C₂₀ hydrocarbyl, tri(C₁-C₂₀-alkyl)silyl, C₆-C₂₀ aryl, C₇-C₂₀ arylalkyl or C₇-C₂₀ alkylaryl, and n has a value of 1. For example, R₁₂E is -R₁₂Si-, wherein each R¹ is independently a C₁-C₂₀-hydrocarbyl and n has a value of 1 or R₁₂E is -R₁₂Si-, wherein each R¹ is independently a C₁-C₁₀ hydrocarbyl and n has a value of 1. In one embodiment of the present invention, R₁₂E is -R₁₂Si-, wherein each R¹ is independently a C₁-C₅ hydrocarbyl and n has a value of 1. Preferably, each R¹ is a C₁-hydrocarbyl and n has a value of 1.

[0087] R² and R²', which can be the same or different, are a linear C₁-C₂₀ hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC).

[0088] In one embodiment of the present invention, R² and R²', which can be the same or different, are a linear C₁-C₁₀ hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC). Alternatively, R² and R²', which can be the same or different, are a linear C₁-C₅ hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC).

[0089] In one embodiment of the present invention, R² and R²', which can be the same or different, are a linear C₁-C₅ hydrocarbyl radical. For example, each R² and R²', which are the same, is a linear C₁-C₅ hydrocarbyl radical. Preferably, each R² and R²', which are the same, is a methyl or ethyl radical.

[0090] In one embodiment of the present invention, each R² and R²' is a methyl radical.

[0091] Ar and Ar', which can be the same or different, are an aryl or heteroaryl group having up to 20 carbon atoms.

[0092] In one embodiment of the present invention, the Ar and Ar' groups are different.

[0093] The Ar group is preferably a C₆-C₂₀ aryl group such as a phenyl group. In one embodiment of the present invention, the Ar' group is an unsubstituted C₆-C₂₀ aryl group, preferably an unsubstituted phenyl group. Alternatively, the Ar' group is a substituted C₆-C₂₀ aryl group, preferably a phenyl based group substituted by groups R¹, preferably by one R¹ group in position 4, i.e. located para to the bond to the indenyl group, i.e. the multicyclic ligand.

[0094] The Ar group is preferably a C₆-C₂₀ aryl group such as a phenyl group or naphthyl group. Whilst the Ar group can be a heteroaryl group, such as carbazolyl, it is preferable that Ar is not a heteroaryl group. The Ar group can be unsubstituted or substituted by one or more groups R¹, more preferably by one or two R¹ groups, especially in position 4 of the aryl ring bound to the indenyl ligand, i.e. located para to the bond to the indenyl group, i.e. the multicyclic ligand.

[0095] In one embodiment of the present invention, both Ar and Ar' are unsubstituted. In another embodiment of the present invention, Ar' is unsubstituted and Ar is substituted by one or two groups R¹, preferably by one R¹ group in position 4, i.e. located para to the bond to the indenyl group. Preferably, R¹ is a C₁-C₂₀ hydrocarbyl and preferably a C₁-C₁₀ hydrocarbyl, such as a C₁-C₁₀ alkyl radical. For example, R¹ is a C₂-C₁₀ alkyl group such as C₃-C₈ alkyl group. In one embodiment of the present invention, R¹ is a bulky group, i.e. is branched. The branching may be alpha or beta to the aryl or heteroaryl group. Preferably, R¹ is a branched C₃-C₈ alkyl group. For example, R¹ is a tert.-butyl or isopropyl group.

[0096] R⁵ is hydrogen or a C₁-C₄₀ hydrocarbyl radical optionally containing one or more heteroatoms from groups 14

to 16 (IUPAC).

[0097] In one embodiment of the present invention, R⁵ is hydrogen or a C₁₋₂₀ hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC) and preferably R⁵ is hydrogen or a C_{1-C₁₀} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC). For example, R⁵ is hydrogen or a C_{1-C₁₀} hydrocarbyl radical containing one or more heteroatoms from groups 14 to 16 (IUPAC).

[0098] In one embodiment of the present invention, R⁵ is hydrogen or a C_{1-C₁₀} hydrocarbyl radical optionally containing one or more heteroatoms from group 16 (IUPAC). For example, R⁵ is hydrogen or a C_{1-C₁₀} hydrocarbyl radical containing one or more heteroatoms from group 16 (IUPAC).

[0099] It is preferred that R⁵ is hydrogen or a C_{1-C₅} hydrocarbyl radical optionally containing one or more oxygen, preferably one oxygen. For example, R⁵ is hydrogen or a C_{1-C₅} hydrocarbyl radical containing one or more oxygen, preferably one oxygen. It is preferred that R⁵ is hydrogen, methoxy, ethoxy or isopropoxy. In one embodiment of the present invention, R⁵ is hydrogen or methoxy.

[0100] In particular, R⁵ is a C_{1-C₂₀} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 and preferably R⁵ is a C_{1-C₁₀} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC). For example, R⁵ is a C_{1-C₁₀} hydrocarbyl radical containing one or more heteroatoms from groups 14 to 16 (IUPAC).

[0101] In one embodiment of the present invention, R⁵ is a C_{1-C₁₀} hydrocarbyl radical optionally containing one or more heteroatoms from group 16 (IUPAC). For example, R⁵ is a C_{1-C₁₀} hydrocarbyl radical containing one or more heteroatoms from group 16 (IUPAC).

[0102] It is preferred that R⁵ is a C_{61-C₅} hydrocarbyl radical optionally containing one or more oxygen, preferably one oxygen. For example, R⁵ is a C_{1-C₅} hydrocarbyl radical containing one or more oxygen, preferably one oxygen. It is preferred that R⁵ is methoxy, ethoxy or isopropoxy. In one embodiment of the present invention, R⁵ is methoxy.

[0103] It is appreciated that R⁵ and R^{5'} may be the same or different.

[0104] In one embodiment of the present invention, R⁵ and R^{5'} are the same. In case R⁵ and R^{5'} are the same, it is appreciated that each of R⁵ and R^{5'} is a C_{1-C₄₀} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC), preferably a C_{1-C₂₀} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC) and most preferably a C_{1-C₁₀} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC).

[0105] It is preferred that each of R⁵ and R^{5'} is a C_{1-C₁₀} hydrocarbyl radical containing one or more heteroatoms from groups 14 to 16 (IUPAC), preferably a C_{1-C₁₀} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC) and most preferably a C_{1-C₁₀} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC). For example, each of R⁵ and R^{5'} is a C_{1-C₁₀} hydrocarbyl radical containing one or more heteroatoms from groups 14 to 16 (IUPAC).

[0106] In one embodiment of the present invention, each of R⁵ and R^{5'} is a C_{1-C₁₀} hydrocarbyl radical optionally containing one or more heteroatoms from group 16. For example, each of R⁵ and R^{5'} is a C_{1-C₁₀} hydrocarbyl radical containing one or more heteroatoms from group 16 (IUPAC).

[0107] It is preferred that each of R⁵ and R^{5'} is a C_{1-C₅} hydrocarbyl radical optionally containing one or more oxygen, preferably one oxygen. For example, each of R⁵ and R^{5'} is a C_{1-C₅} hydrocarbyl radical containing one or more oxygen, preferably one oxygen. It is preferred that each of R⁵ and R^{5'} is methoxy, ethoxy or isopropoxy. In one embodiment of the present invention, each of R⁵ and R^{5'} is methoxy.

[0108] Alternatively, R⁵ and R^{5'} are different. In case R⁵ and R^{5'} are different, it is appreciated that R⁵ is hydrogen and R^{5'} is a C_{1-C₄₀} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC), preferably a C_{1-C₂₀} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC) and most preferably a C_{1-C₁₀} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC).

[0109] It is preferred that R⁵ is hydrogen and R^{5'} is a C_{1-C₄₀} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC), preferably a C_{1-C₂₀} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC) and most preferably a C_{1-C₁₀} hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC). For example, R⁵ is hydrogen and R^{5'} is a C_{1-C₁₀} hydrocarbyl radical containing one or more heteroatoms from groups 14 to 16 (IUPAC).

[0110] In one embodiment of the present invention, R⁵ is hydrogen and R^{5'} is a C_{1-C₁₀} hydrocarbyl radical optionally containing one or more heteroatoms from group 16 (IUPAC). For example, R⁵ is hydrogen and R^{5'} is a C_{1-C₁₀} hydrocarbyl radical containing one or more heteroatoms from group 16 (IUPAC).

[0111] It is preferred that R⁵ is hydrogen and R^{5'} is a C_{1-C₅} hydrocarbyl radical optionally containing one or more oxygen, preferably one oxygen. For example, R⁵ is hydrogen and R^{5'} is a C_{1-C₅} hydrocarbyl radical containing one or more oxygen, preferably one oxygen. It is preferred that each of R⁵ and R^{5'} is methoxy, ethoxy or isopropoxy. In one embodiment of the present invention, R⁵ is hydrogen and R^{5'} is methoxy.

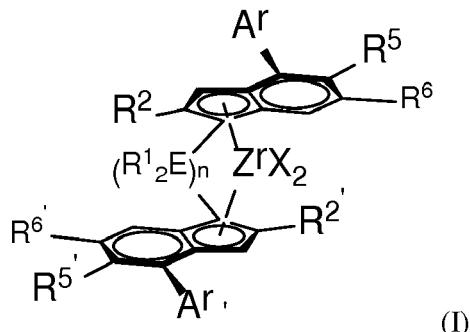
[0112] R⁶ and R^{6'}, which can be the same or different, are a branched C₃₋₂₀ hydrocarbyl radical.

[0113] In one embodiment of the present invention, R⁶ and R^{6'}, which can be the same or different, are a tertiary C₃-C₂₀ hydrocarbyl radical. For example, R⁶ and R^{6'}, which can be the same or different, are a tertiary C₄-C₂₀ hydrocarbyl radical.

5 [0114] In one embodiment of the present invention, R⁶ and R^{6'}, which can be the same or different, are a tertiary C₄-C₁₀ hydrocarbyl radical. For example, R⁶ and R^{6'}, which can be the same or different, are a tertiary C₄-C₈ hydrocarbyl radical.

[0115] In one embodiment of the present invention, R⁶ and R^{6'} are the same. In case R⁶ and R^{6'} are the same, it is appreciated that each R⁶ and R^{6'} is a tertiary C₄-C₈ hydrocarbyl radical. Preferably, each R⁶ and R^{6'} is a tertiary butyl radical or pentyl radical. For example, each R⁶ and R^{6'} is a tertiary butyl radical.

10 [0116] In one embodiment of the present invention, the solid catalyst preferably comprises a complex of formula (I)



wherein

25 each X is a sigma ligand;
R¹₂E is a divalent bridge selected from -R¹₂C-, -R¹₂Si-, -R¹₂Ge-, wherein each R¹ is independently a hydrogen atom, C₁-C₂₀ hydrocarbyl, tri(C₁-C₂₀-alkyl)silyl, C₆-C₂₀ aryl, C₇-C₂₀ arylalkyl or C₇-C₂₀ alkylaryl, and n has a value from 1 to 2;

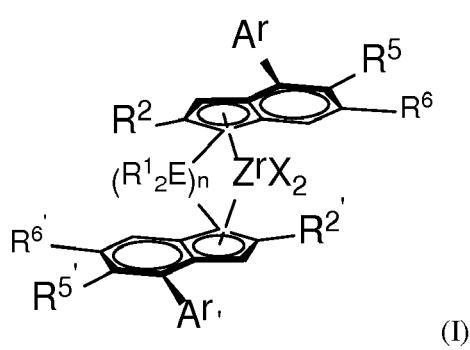
30 R² and R^{2'}, which can be the same or different, are a linear C₁-C₁₀ hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC);

Ar and Ar', which can be the same or different, are an aryl or heteroaryl group having up to 20 carbon atoms and is substituted in position 4 by R¹;

35 R⁵ is hydrogen or a C₁-C₂₀ hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC);

R^{5'} is a C₁-C₂₀ hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC);
R⁶ and R^{6'}, which can be the same or different, are a tertiary C₄-C₂₀ hydrocarbyl radical.

40 [0117] In one embodiment of the present invention, the solid catalyst preferably comprises a complex of formula (I)



wherein

55 each X is a sigma ligand;
R¹₂E is -R¹₂Si-, wherein each R¹ is independently a C₁-C₅-hydrocarbyl, and n has a value of 1;
each R² and R^{2'} is a methyl radical;
Ar and Ar', which can be the same or different, are an aryl or heteroaryl group having up to 20 carbon atoms and

no substituents in positions 3 and 5;

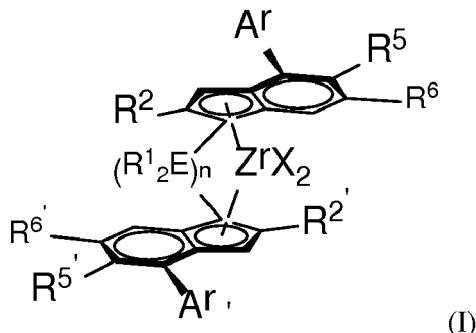
R⁵ is hydrogen or a C₁-C₁₀ hydrocarbyl radical containing one or more heteroatoms from group 16 (IUPAC);

R^{5'} is a C₁-C₁₀ hydrocarbyl group containing one or more heteroatoms from group 16 (IUPAC);

each R⁶ and R^{6'} is a tertiary butyl radical.

5

[0118] Alternatively, the solid catalyst preferably comprises a complex of formula (I)



10

15

20 wherein

each X is a sigma ligand;

R¹₂E is -R¹₂Si-, wherein each R¹ is independently a C₁-₅-hydrocarbyl, and n has a value of 1;

each R² and R^{2'} is a methyl radical;

25

Ar and Ar', which can be the same or different, are an aryl or heteroaryl group having up to 20 carbon atoms and is substituted in position 4 by R¹;

R⁵ is hydrogen or a C₁-C₁₀ hydrocarbyl radical containing one or more heteroatoms from group 16 (IUPAC);

R^{5'} is a C₁-C₁₀ hydrocarbyl group containing one or more heteroatoms from group 16 (IUPAC);

each R⁶ and R^{6'} is a tertiary butyl radical.

30

[0119] Representatives of catalysts of the above formulas include e.g. *rac*-1,1'-dimethylsilylene-bis[2-methyl-4-phenyl-5-methoxy-6-*tert*-butyl-indenyl]zirconium dichloride, *anti*-1,1'-dimethylsilylene-[2-methyl-4-phenyl-5-methoxy-6-*tert*-butyl-indenyl][2-methyl-4-phenyl-6-*tert*-butyl-indenyl]zirconium dichloride, and *anti*-1,1'-dimethylsilylene-[2-methyl-4-phenyl-5-methoxy-6-*tert*-butyl-indenyl][2-methyl-4-(4-*tert*-butylphenyl)-6-*tert*-butyl-indenyl] zirconium dichloride

[0120] Throughout the disclosure above, where a narrower or preferred definition of a substituent is presented, that narrower or preferred definition is deemed disclosed in conjunction with all broader and narrower and preferred definitions of other substituents in the application.

[0121] The ligands required to form the complexes and hence catalysts of the invention can be synthesised by any process and the skilled organic chemist would be able to devise various synthetic protocols for the manufacture of the necessary ligand materials.

WO 2007/107448 A1, WO 2007/116034 A1 and WO2011/135004 A2 disclose the necessary chemistry and are herein incorporated by reference.

Cocatalyst

45

[0122] To form an active catalytic species it is normally necessary to employ a cocatalyst as is well known in the art. Cocatalysts comprising an organometallic compound of Group 13 (IUPAC) metal, like organoaluminium compounds used to activate metallocene catalysts are suitable for use in this invention.

[0123] The instant solid catalyst system comprises (i) a complex of Formula (I) as defined herein; and normally (ii) an aluminium alkyl compound (or other appropriate cocatalyst), or the reaction product thereof. Thus, the cocatalyst is preferably an alumoxane, like MAO or an alumoxane other than MAO.

[0124] Borate cocatalysts can also be employed. It will be appreciated by the skilled man that where boron based cocatalysts are employed, it is normal to preactivate the complex by reaction thereof with an aluminium alkyl compound, such as TIBA. This procedure is well known and any suitable aluminium alkyl, e.g. Al(C₁-C₆-alkyl)₃, can be used.

55

Boron based cocatalysts of interest include those of formula

BY₃

wherein Y is the same or different and is an alkyl group of from 1 to about 20 carbon atoms, an aryl group of from 6 to about 15 carbon atoms, alkylaryl, arylalkyl, haloalkyl or haloaryl each having from 1 to 10 carbon atoms in the alkyl radical and from 6 to 20 carbon atoms in the aryl radical or fluorine, chlorine, bromine or iodine. Preferred examples for Y are fluorine, trifluoromethyl, p-fluorophenyl, 3,5-difluorophenyl, pentafluorophenyl, 3,4,5-trifluorophenyl and 3,5-di(trifluoromethyl)phenyl. Preferred options are trifluoroborane, tris(4-fluorophenyl)borane, tris(3,5-difluorophenyl)borane, tris(4-fluoromethylphenyl)borane, tris(2,4,6-trifluorophenyl)borane, tris(pentafluorophenyl)borane, tris(3,5-difluorophenyl)borane and/or tris (3,4,5-trifluorophenyl) borane.

[0125] Particular preference is given to tris(pentafluorophenyl)borane.

[0126] It is preferred however that borates are used, i.e. compounds containing a borate 4- ion. Such ionic cocatalysts preferably contain a non-coordinating anion such as tetrakis(pentafluorophenyl)borate. Suitable counterions are triarylcationium or protonated amine or aniline derivatives such as methylammonium, anilinium, dimethylammonium, diethylammonium, N- methylanilinium, diphenylammonium, N,N-dimethylanilinium, trimethylammonium, triethylammonium, tri-n-butylammonium, methyl diphenylammonium, pyridinium, p-bromo-N,N-dimethylanilinium or p-nitro-N,N-dimethylanilinium.

[0127] Preferred ionic compounds which can be used according to the present invention include: triphenylcarbenium tetrakis(pentafluorophenyl)borate, tributylammonium tetrakis(pentafluorophenyl)borate, tributylammonium tetrakis(trifluoromethylphenyl)borate, tributylammonium tetrakis(4-fluorophenyl)borate, N,N-dimethylcyclohexylammonium tetrakis(pentafluorophenyl)borate, N,N-dimethylbenzylammonium tetrakis(pentafluorophenyl)borate, N,N-dimethylanilinium tetrakis(pentafluorophenyl)borate, N,N- di(propyl)ammonium tetrakis(pentafluorophenyl)borate, di(cyclohexyl)ammonium tetrakis(pentafluorophenyl)borate, triphenylcarbenium tetrakis(pentafluorophenyl)borate, or ferrocenium tetrakis(pentafluorophenyl)borate.

[0128] Preference is given to triphenylcarbenium tetrakis(pentafluorophenyl) borate, N,N- dimethylcyclohexylammoniumtetrakis(pentafluorophenyl)borate or N,N- dimethylbenzylammoniumtetrakis(pentafluorophenyl)borate.

[0129] The use of $B(C_6F_5)_3$, $C_6H_5N(CH_3)_2H:B(C_6F_5)_4$, $(C_6H_5)_3C:B(C_6F_5)_4$ or $Ni(CN)_4[B(C_6F_5)_3]_4^{2-}$ is especially preferred.

[0130] Suitable amounts of cocatalyst will be well known to the skilled man.

Second organometallic compound

[0131] In some cases the catalyst system comprises optionally a second organometallic compound of a Group 13 metal (IUPAC) being different to the first organometallic compound. This second Group 13 metal organometallic compound has the specific functions of alkylating the metallocene complex, or acting as a scavenger for monomer impurities, or both. It can be added to the metallocene complex before it is reacted with the cocatalyst, or after the reaction with the cocatalyst, or it can be directly fed into the polymerization medium before the catalyst/cocatalyst mixture is added to it.

[0132] Preferably the second organometallic compound is an organometallic compound of B, Al, or Ga, more preferably an organometallic compound of B or Al, yet more preferably an organometallic compound of Al. Specific preferred organometallic compounds are selected from the group consisting of aluminium trialkyls, such as aluminium triethyl, aluminium triisobutyl, aluminium trioctyl and the like.

[0133] The catalyst system of the invention is preferably used in solid form and thus is used as a heterogeneous catalyst system.

[0134] The catalyst system of the invention can be either supported on an external carrier material, like silica or alumina, or, in a particularly preferred embodiment, is free from an external carrier, however still being in solid form. For example, the solid catalyst is obtainable by a process in which

- 45 (a) a liquid/liquid emulsion system is formed, said liquid/liquid emulsion system comprising a solution of the catalyst components dispersed in a solvent so as to form dispersed droplets; and
- (b) solid particles are formed by solidifying said dispersed droplets.

[0135] Reference is made to WO2006/069733 and EP 1 117 334.4 describing principles of such a continuous or semicontinuous preparation method of the solid catalyst types, prepared via emulsion/solidification method.

Polymerisation

[0136] The polymerization of propylene by using the above defined solid catalyst system may be effected in one or more, e.g. 1, 2 or 3, polymerization reactors, using conventional polymerization techniques, e.g. gas phase, solution phase, slurry or bulk polymerization.

[0137] In general, a combination of slurry (or bulk) and at least one gas phase reactor is often preferred, particularly with the reactor order being slurry (or bulk) then one or more gas phase reactors.

[0138] In one embodiment of the present invention the polymerization of propylene is effected in liquid monomer slurry phase.

[0139] In one embodiment of the present invention, the polymerization temperature is of less than 60 °C, like not more than 55 °C. For example, the polymerization temperature is from 10 to less than 60 °C, e.g. from 10 to 55 °C, more preferably from 15 to 50 °C, still more preferably from 15 to 45 °C, yet more preferably from 20 to 40 °C and most preferably from 25 to 35 °C.

[0140] For polymerizations in liquid monomer slurry phase, the reaction temperature will generally be in the range from 10 to 55 °C, preferably in the range from 15 to 50 °C, like in the range of 10 to 50 °C, e.g. 20 to 40 °C. The reactor pressure will generally be in the range from 5 to 80 bar (e.g. 20 to 60 bar), and the residence time will generally be in the range from 0.1 to 5 hours (e.g. 0.3 to 2 hours).

[0141] Generally the quantity of solid catalyst system used will depend upon the nature of the catalyst, the reactor types and conditions and the properties desired for the propylene homopolymer (H-PP1). As well known in the art, hydrogen can be used for controlling the molecular weight of the propylene homopolymer.

[0142] It is a feature of the invention that the claimed propylene homopolymer (H-PP1) obtainable, preferably obtained, by the process of the present invention has specific features as defined above.

[0143] The polypropylene homopolymer (H-PP1) or the polypropylene blend (PB) of the present invention, preferably made by the catalysts of the invention, are useful in all kinds of end articles such as pipes, sheets, films (cast, blown or BOPP films, such as for example BOPP for capacitor film), fibres, moulded articles (e.g. injection moulded, blow moulded, rotomoulded articles), extrusion coatings and so on.

[0144] The invention will now be illustrated by reference to the following non-limiting examples.

EXAMPLES

A. Measuring methods

[0145] The following definitions of terms and determination methods apply for the above general description of the invention as well as to the below examples unless otherwise defined.

Quantification of microstructure by NMR spectroscopy

[0146] Quantitative nuclear-magnetic resonance (NMR) spectroscopy was used to quantify the isotacticity and regioregularity of the polypropylene homopolymers.

[0147] Quantitative $^{13}\text{C}\{^1\text{H}\}$ NMR spectra were recorded in the solution-state using a Bruker Advance III 400 NMR spectrometer operating at 400.15 and 100.62 MHz for ^1H and ^{13}C respectively. All spectra were recorded using a ^{13}C optimised 10 mm extended temperature probehead at 125 °C using nitrogen gas for all pneumatics.

[0148] For polypropylene homopolymers approximately 200 mg of material was dissolved in 1,2-tetrachloroethane- d_2 (TCE- d_2). To ensure a homogenous solution, after initial sample preparation in a heat block, the NMR tube was further heated in a rotatory oven for at least 1 hour. Upon insertion into the magnet the tube was spun at 10 Hz. This setup was chosen primarily for the high resolution needed for tacticity distribution quantification (Busico, V., Cipullo, R., Prog. Polym. Sci. 26 (2001) 443; Busico, V.; Cipullo, R., Monaco, G., Vacatello, M., Segre, A.L., Macromolecules 30 (1997) 6251). Standard single-pulse excitation was employed utilising the NOE and bi-level WALTZ16 decoupling scheme (Zhou, Z., Kuemmerle, R., Qiu, X., Redwine, D., Cong, R., Taha, A., Baugh, D., Winniford, B., J. Mag. Reson. 187 (2007) 225; Busico, V., Carbonniere, P., Cipullo, R., Pellecchia, R., Severn, J., Talarico, G., Macromol. Rapid Commun. 2007, 28, 11289). A total of 8192 (8k) transients were acquired per spectra.

[0149] Quantitative $^{13}\text{C}\{^1\text{H}\}$ NMR spectra were processed, integrated and relevant quantitative properties determined from the integrals using proprietary computer programs.

[0150] For polypropylene homopolymers all chemical shifts are internally referenced to the methyl isotactic pentad (mmmm) at 21.85 ppm.

[0151] The tacticity distribution was quantified through integration of the methyl region between 23.6-19.7 ppm correcting for any sites not related to the stereo sequences of interest (Busico, V., Cipullo, R., Prog. Polym. Sci. 26 (2001) 443; Busico, V., Cipullo, R., Monaco, G., Vacatello, M., Segre, A.L., Macromolecules 30 (1997) 6251).

[0152] Specifically the influence of regio-defects on the quantification of the tacticity distribution was corrected for by subtraction of representative regio-defect integrals from the specific integral regions of the stereo sequences.

[0153] The isotacticity was determined at the pentad level and reported as the percentage of isotactic pentad (mmmm) sequences with respect to all pentad sequences:

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

[0154] 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).

5 [0155] 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:

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

10 [0156] 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:

15 [0157] The total amount of propene was quantified as the sum of primary inserted propene and all other present regio-defects:

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

25 [0158] The mole percent of 2,1 erythro regio-defects was quantified with respect to all propene:

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

30 **Number average molecular weight (M_n), weight average molecular weight (M_w) and molecular weight distribution (MWD)**

35 [0159] Molecular weight averages (M_w , M_n), Molecular weight distribution (MWD) and its broadness, described by polydispersity index, $PDI = M_w/M_n$ (wherein M_n is the number average molecular weight and M_w is the weight average molecular weight) were determined by Gel Permeation Chromatography (GPC) according to ISO 16014-4:2003 and ASTM D 6474-99. A PolymerChar GPC instrument, equipped with infrared (IR) detector was used with 3 x Olexis and 1x Olexis Guard columns from Polymer Laboratories and 1,2,4-trichlorobenzene (TCB, stabilized with 250 mg/L 2,6-Di tert butyl-4-methyl-phenol) as solvent at 160 °C and at a constant flow rate of 1 mL/min. 200 μ L of sample solution were injected per analysis. The column set was calibrated using universal calibration (according to ISO 16014-2:2003) with 40 at least 15 narrow MWD polystyrene (PS) standards in the range of 0,5 kg/mol to 11 500 kg/mol. Mark Houwink constants for PS, PE and PP used are as described per ASTM D 6474-99. All samples were prepared by dissolving 5.0 - 9.0 mg of polymer in 8 mL (at 160 °C) of stabilized TCB (same as mobile phase) for 2.5 hours for PP or 3 hours for PE at max. 160°C under continuous gentle shaking in the autosampler of the GPC instrument.

45 [0160] **MFR₂ (230 °C)** is measured according to ISO 1133 (230 °C, 2.16 kg load)

[0161] **The xylene soluble fraction at room temperature (XS, wt.-%):** The amount of the polymer soluble in xylene is determined at 25 °C according to ISO 16152; first edition; 2005-07-01.

50 [0162] **DSC analysis, melting temperature (T_m) and crystallization temperature (T_c):** measured with a TA Instrument Q200 differential scanning calorimetry (DSC) on 5 to 7 mg samples. DSC is run according to ISO 3146 / 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 is determined from the cooling step, while melting temperature is determined from the second heating step.

55 [0163] **Tensile measurements:** Stress-strain curves have been generated with a Zwick Z010 instrument according to ISO 527-1,2, on specimens cut out from compression moulded 100x100x2 mm plates, using test speed of 1 mm/min for tensile modulus (up to 0.25 % deformation) and 50 mm/min for the rest of the test. The tensile modulus (Et) was determined from the slope of the stress strain curve $\sigma(\varepsilon)$ in the strain interval $0.05 \% \leq \varepsilon \leq 0.25 \%$, calculated as secant slope in this interval:

$$E_t = \frac{\sigma_2 - \sigma_1}{\varepsilon_2 - \varepsilon_1}$$

5 where E_t is the tensile modulus, expressed in megapascals; σ_1 is the stress, in megapascals, measured at the strain value $\varepsilon_1 = 0.0005$ (0.05 %); σ_2 is the stress, in megapascals, measured at the strain value $\varepsilon_2 = 0.0025$ (0.25 %).

B. Examples

10 [0164] MAO Chemtura (30 wt% in toluene) or Albemarle (30 wt% in toluene) were used as received.

[0165] Surfactant: The mixture of perfluoroalkylethyl acrylate esters (CAS 65605-70-1) used as the surfactant was purchased from the Cytonix corporation or Wilshire Technologies, dried over activated molecular sieves (2 times) and degassed by argon bubbling prior to use. Perfluoro-1,3-dimethylcyclohexane (PFC, CAS 335-27-3) was dried over activated molecular sieves (2 times) and degassed by argon bubbling prior to use.

15

B.1 Preparation of the used solid catalyst systems

Solid catalyst system C1 (Comparative):

20 [0166] The metallocene was prepared according to the procedure described in Spaleck et al. Organometallics 1994, 13, 954 with *rac*-1,1'-dimethylsilylene-*bis*[2-methyl-4-phenyl-indenyl] zirconium dichlorides (metallocene 1).

[0167] The detailed catalyst preparation was performed as follows:

25 Inside the glovebox, 80 μ L of dry and degassed surfactant were mixed with 2 mL of 30 wt% MAO solution in toluene (Albemarle) in a septum bottle and left to react overnight under stirring. The following day, 47.8 mg of metallocene 1 (0.076 mmol, 1 equivalent) were dissolved with 4 mL of MAO solution in another septum bottle and left to stir inside the glovebox.

30 [0168] After 60 minutes, 1 mL of the surfactant solution and the 4 mL of the MAO-metallocene solution were successively added into a 50mL emulsification glass reactor containing 40 mL of perfluoro-1,3-dimethylcyclohexane at -10 °C and equipped with an overhead stirrer (stirring speed = 600 rpm). Total amount of MAO is 5 mL (300 equivalents). A red-orange emulsion formed immediately and stirred during 15 minutes at -10 °C / 600rpm. Then the emulsion was transferred via a 2/4 teflon tube to 100mL of hot perfluoro-1,3-dimethylcyclohexane at 90 °C, and stirred at 600rpm until the transfer is completed, then the speed was reduced to 300 rpm. After 15 minutes stirring, the oil bath was removed and the stirrer turned off. The catalyst was left to settle up on top of the perfluoro-1,3-dimethylcyclohexane and after 35 minutes the solvent was siphoned off. The remaining orange catalyst was dried during 2 hours at 50 °C under argon flow. 0.51g of a red free flowing powder (C1) was obtained.

35

Solid catalyst system C2 (Inventive)

40 [0169] The metallocene *rac*-1,1'-dimethylsilylene-*bis*[2-methyl-4-phenyl-5-methoxy-6-*tert*-butylindenyl] zirconium dichloride was prepared according to the procedure described in WO 2007/116034 (metallocene 2).

[0170] The detailed catalyst preparation was performed as follows:

45 Inside the glovebox, 80 μ L of dry and degassed surfactant was mixed with 2 mL of 30 wt% MAO solution in toluene (Albemarle) in a septum bottle and left to react overnight under stirring. The following day, inside the glovebox, 60.9 mg of metallocene 2 was mixed with 4 ml of MAO solution in a septum bottle and the solution was stirred for 60 minutes. After, 4 mL of the MAO-metallocene solution and 1 mL of the surfactant-MAO solution were successively added into a 50mL emulsification glass reactor containing 40mL of perfluoro-1,3-dimethylcyclohexane at -10 °C and equipped with an overhead stirrer (stirring speed = 600 rpm). Total amount of MAO is 5 mL (300 equivalents). A red emulsion formed immediately and stirred during 15 minutes at -10 °C / 600 rpm. Then the emulsion was transferred via a 2/4 teflon tube to 100mL of hot perfluoro-1,3-dimethylcyclohexane at 90 °C and stirred at 600 rpm until the transfer is completed. Then the speed was reduced to 300 rpm. After 15 minutes stirring, the oil bath was removed and the stirrer turned off. The catalyst was left to settle up on top of the perfluoro-1,3-dimethylcyclohexane and after 35 minutes the solvent was siphoned off. The remaining red catalyst was dried during 2 hours at 50 °C under argon flow and 0.60 g of a red powder (C2) was obtained.

55

Solid catalyst system C3 (Inventive)

[0171] The metallocene *anti*-1,1'-dimethylsilylene-[2-methyl-4-phenyl-6-*tert*-butyl-indenyl][2-methyl-4-phenyl-5-methoxy-6-*tert*-butyl-indenyl]zirconium dichloride was prepared according to the procedure described in WO2013007650

(Metallocene E1) (metallocene 3). The detailed catalyst preparation was performed as follows:

Inside the glovebox, 80 μ L of dry and degassed surfactant were mixed with 2 mL of 30 wt% MAO solution in toluene (Albemarle) in a septum bottle and left to react overnight under stirring. The following day, 44.2 mg of metallocene 3 (0.057 mmol, 1 equivalent) were dissolved with 4 mL of the MAO solution in another septum bottle and left to stir inside the glovebox.

[0172] After 60 minutes, 1 mL of the surfactant solution and the 4 mL of the MAO-metallocene solution were successively added into a 50 mL emulsification glass reactor containing 40 mL of perfluoro-1,3-dimethylcyclohexane at -10 °C and equipped with an overhead stirrer (stirring speed = 600 rpm). Total amount of MAO is 5 mL (400 equivalents). A red emulsion formed immediately and stirred during 15 minutes at -10 °C / 600rpm. Then the emulsion was transferred via a 2/4 teflon tube to 100mL of hot of perfluoro-1,3-dimethylcyclohexane at 90 °C, and stirred at 600rpm until the transfer is completed, then the speed was reduced to 300 rpm. After 15 minutes stirring, the oil bath was removed and the stirrer turned off. The catalyst was left to settle up on top of the perfluoro-1,3-dimethylcyclohexane and after 35 minutes the solvent was siphoned off. The remaining red catalyst was dried during 2 hours at 50 °C under argon flow. 0.63g of a red free flowing powder (C3) was obtained.

Solid catalyst system C4 (Inventive)

[0173] Inside the glovebox, 80 μ L of dry and degassed surfactant were mixed with 2 mL of 30 wt % MAO solution in toluene (Chemtura) in a septum bottle and left to react overnight under stirring. The following day, 60.9 mg of metallocene 2 (0.076 mmol, 1 equivalent) were dissolved with 4 mL of the MAO solution in another septum bottle and left to stir inside the glove box. After 60 minutes, 1 mL of the surfactant solution and 4 mL of the MAO-metallocene solution were successively added into a 50mL emulsification glass reactor containing 40mL of PFC at -10 °C and equipped with an overhead stirrer (stirring speed = 600 rpm). Total amount of MAO is 5 mL (300 equivalents). A red emulsion formed immediately and stirred during 15 minutes at -10 °C / 600rpm. Then the emulsion was transferred via a 2/4 Teflon tube to 100mL of hot PFC at 90 °C, and stirred at 600rpm until the transfer is completed, then the speed was reduced to 300 rpm. After 15 minutes stirring, the oil bath was removed and the stirrer turned off. The catalyst was left to settle on top of the PFC and after 35 minutes the solvent was siphoned off. The remaining catalyst was dried during 2 hours at 50 °C under argon flow. 0.64 g of a red solid catalyst was obtained.

B.2 Off-line pre-polymerization of the used solid catalyst systems

[0174] The catalysts C1, C2, C3 and C4 were pre-polymerised according to the following procedure:

Off-line pre-polymerisation experiments were done in a 125 mL pressure reactor equipped with gas-feeding lines and an overhead stirrer. Dry and degassed perfluoro-1,3-dimethylcyclohexane (15 cm³) and the desired amount of catalyst to be pre-polymerised were loaded into the reactor inside a glovebox and the reactor was sealed. The reactor was then taken out from the glovebox and placed inside a water cooled bath. The overhead stirrer and the feeding lines were then connected. For the prepolymerization of the catalysts C1, C3 and the catalyst C2 as applied for the preparation of propylene homopolymers in a 480-mL reactor, the feeding line was pressurized with hydrogen, and the experiment was started by opening the valve between the H₂ feed line and the reactor. At the same time propylene feed was started through the same H₂ feeding line in order to ensure that all the hydrogen would be fed into the reactor. For the prepolymerization of the catalyst C2 as applied for the preparation of propylene homopolymers in a 20-L reactor and the catalyst C4, no H₂ was fed into the reactor. The propylene feed was left open, and the monomer consumption was compensated by keeping the total pressure in the reactor constant (about 5 barg). The experiment was continued until a polymerisation time sufficient to provide the desired degree of polymerisation. The reactor was then taken back inside the glovebox before opening and the content was poured into a glass vessel. The perfluoro-1,3-dimethylcyclohexane was evaporated until a constant weight was obtained to yield a pre-polymerised pink catalyst.

[0175] The details regarding the off-line pre-polymerization experiments are listed in Table 1 below:

Table 1:

Catalyst synthesis	Catalyst	prepolymerization time	Yield*	DP**
lot	[mg]	[min]	[mg]	[g/g]
C1	314.5	13	1659	4.3
C2	544.3	11	2297	3.2
C3	591.0	17	2683	3.5

(continued)

Catalyst synthesis	Catalyst	prepolymerization time	Yield*	DP**
lot	[mg]	[min]	[mg]	[g/g]
C4	497.4	11	1384	1.8

* total mass of prepped catalyst
** The pre-polymerisation degree (DP) was calculated using the following formula:

$$\text{Pre-polymerisation degree (DP)} = \frac{\text{total yield (mg)} - \text{catalyst (mg)}}{\text{catalyst (mg)}}$$

B.3 Preparation of propylene homopolymers in a 480 mL reactor

[0176] Propylene was provided by Borealis and adequately purified before use. Triethylaluminum was purchased from Chemtura and diluted to a concentration of 0.5 molar with heptane. Hydrogen is provided by Air Liquide.

[0177] The dosing of the catalyst was carried out in that the respective amount of catalyst C1, C2 or C3 was weighted into a metal vial. 6 ml of hexane was added to each vial to create a catalyst suspension. From each vial the volume required to reach an amount of 10 mg of catalyst was taken up (ca. 2 ml) and injected into the reactor.

[0178] The polymerizations were performed in a 480 mL reactor. 100 μ L of triethylaluminum was fed as a 0.5 M solution of dry and degassed heptane. 100 g of liquid propylene was fed into the reactor and the desired amount of hydrogen was then pressurized into the reactor. The contacting time of triethylaluminum and propylene was 20 min. The temperature was set to 20 or 30 °C. The catalyst slurry having a catalyst concentration of 3.3 mg/ml in hexane was then injected into the reactor and temperature raised to 20 to 30 °C. At this temperature, a pre-polymerization was conducted for 5 minutes. Then, the reactor was heated up to the polymerization temperature for 30 or 60 minutes. The polymerization was stopped by venting the reactor and flushing with nitrogen before the polymer was collected.

[0179] The details regarding the procedure are also outlined in Table 2 below:

Table 2:

Catalyst		C1 and C2	C3
reactor volume	[mL]	480	
Propylene	[g]	100	
TEAL 0.5M in heptane	[mL]	0.1	
Cat injection solvent	type	hexane	
Catalyst slurry concentration	[mg/ml]	3.3	
Contact time hexane-catalyst	[min]	<30min	
Cat injection Temperature	[°C]	20	30
Prepoly Temperature	[°C]	20	30
Prepoly time	[min]	5	0
Polymerization time	[min]	60	30
H ₂	[mL]	0	60-105

[0180] The detailed polymerization conditions for experiments conducted with C1 (comparative) are outlined in Table 3 below:

Table 3:

Sample ID	catalyst amount*	Polymerization Temperature	yield	Polymerization activity
	[mg]	[°C]	[g]	[kg/g _{cat} /h]
1	20.00	30	1.9	0.5

(continued)

Sample ID	catalyst amount*	Polymerization Temperature	yield	Polymerization activity
	[mg]	[°C]	[g]	[kg/g _{cat} /h]
2	10.00	40	3.1	1.7
3	6.00	50	3.9	3.4
4	6.00	60	6.6	5.8
5	6.00	70	4.5	4.0
6	6.00	80	2.8	2.5
7	6.00	85	1.6	1.4
8	6.00	75	4.4	3.9

* off-line pre-polymerized

[0181] The detailed polymerization conditions for experiments conducted with C2 (inventive) are outlined in Table 4 below:

Sample ID	catalyst amount*	Polymerization Temperature	yield	Polymerization activity
	[mg]	[°C]	[g]	[kg/g _{cat} /h]
1	9.96	30	3.4	1.4
2	5.08	40	4.2	3.5
3	2.99	50	5.5	7.7

* off-line pre-polymerized

[0182] The detailed polymerization conditions for experiments conducted with C3 (inventive) are outlined in Table 5 below:

Sample ID	catalyst amount*	Polymerization Temperature	H ₂	yield	Polymerization activity
	[mg]	[°C]	[ml]	[g]	[kg/g _{cat} /h]
1	3.33	50	100	6.0	16.2

* off-line pre-polymerized

[0183] The sample characterization results for experiments conducted with C1 (comparative), C2 (inventive) and C3 (inventive) are outlined in tables 6, 7 and 8 below.

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Table 6:

Sample ID	catalyst	T _m	T _c	mmmm	mmrm	2,1e	3,1	total regio errors	total errors	Mn	Mw	M _v	Mw/Mn
		[°C]	[°C]	[%]	[%]	[%]	[%]	[%]	[%]	[kg/mol]	[kg/mol]	[kg/mol]	[‐]
1	C1	142.7	106.2	98.75	0.25	1.72	0	1.72	1.97	304	773	707	2.54
2	C1	145.0	105.5	98.95						410	977	894	2.38
3	C1	146.9	107.1	99.04	0.19	1.21	0	1.21	1.4	444	1046	957	2.36
4	C1	148.7	106.9	99.24	0.15	1.04	0	1.04	1.19	416	987	903	2.37
5	C1	150.5	106.9	98.5						303	794	720	2.62
6	C1	151.5	107.3	98.65	0.27	0.84	0.01	0.85	1.12	243	687	620	2.83
7	C1	151.0	109.2	98.6	0.28	0.86	0.01	0.87	1.15	197	566	509	2.87
8	C1	150.7	106.2	98.95	0.21	0.92	0	0.92	1.13	308	802	728	2.60

Table 7:

Sample ID	catalyst	T _m	T _c	mmmm	mrrm	2,1e	3,1	total regio errors	total errors	Mn	Mw	M _v	Mw/Mn
		[°C]	[°C]	[%]	[%]	[%]	[%]	[%]	[%]	[kg/mol]	[kg/mol]	[kg/mol]	[‐]
1	C2	132.2	95.9	99	0.2	3.06	0	3.06	3.26	399	1048	943	2.63
2	C2	134.6	97.1	99.39	0.12	2.59	0	2.59	2.71	502	1247	1127	2.48
3	C2	138.0	99.4	99.49	0.1	2.21	0	2.21	2.31	566	1394	1256	2.46

Table 8:

Sample ID	catalyst	T _m	T _c	mmmm	r	2,1e	3,1	total regio errors	total errors	Mn	Mw	M _v	Mw/Mn
		[°C]	[°C]	[%]	[%]	[%]	[%]	[%]	[%]	[kg/mol]	[kg/mol]	[kg/mol]	[–]
1	c3	138.0	101.3	99.2	0.15	2.31	0.01	2.32	2.47	280	753	682	2.7

[0184] It is evident that the C2 and the C3 catalysts produce polypropylene homopolymers with a new property combination, i.e. that is low melting point (below 150°C) and a high molecular weight (above 100 kg/mol). The melting point of PP is connected to the amount of chain defects (regio- and stereoerrors). ^{13}C NMR analysis of the samples of tables 1-3 has shown that most of these chain defects are regio-defects of the 2,1-erythro type (see tables 6-8). By increasing the polymerization temperature T_m , the number of regioerrors decreases, which is in turn reflected in an increase of the melting temperature (see Figure 1). It is further to be noted that the amount of 2,1-erythro regio-defects for the inventive catalyst C2, at any given polymerization temperature T_m , is higher compared to the comparative catalyst C1.

B.4 Preparation of propylene homopolymers in a 20-L reactor

[0185] Propylene was provided by Borealis and adequately purified before use. Triethylaluminum was purchased from Aldrich as a 1 M solution in hexane. Hydrogen is provided by Air Liquide.

[0186] The polymerizations were performed in a 20 L reactor.

15 Polymerization of propylene with catalyst C2 at 30°C (inventive):

[0187] A stirred autoclave (double helix stirrer) with a volume of 21.2 dm³ containing 0.2 barg propylene is filled with additional 3.97 kg propylene. After adding of 0.73 mmol triethylaluminium (Aldrich, 1 molar solution in n-hexane) using a stream of 250 g propylene, the solution is stirred at 30 °C and 250 rpm for 20 min. Then the catalyst is injected as described in the following. The solid, pre-polymerized catalyst (type, amount and degree of polymerisation as listed in the tables) is loaded into a 5-mL stainless steel vial inside the glovebox, the vial is attached to the autoclave, then a second 5-mL vial containing 4 ml n-hexane and pressurized with 10 bars of N₂ is added on top, the valve between the two vials is opened and the solid catalyst is contacted with hexane under N₂ pressure for 2 s, then flushed into the reactor with 250 g propylene. Stirring speed is increased to 250 rpm. Afterwards the desired H₂ amount is added with a defined flow via thermal mass flow controller. The reactor temperature is held constant throughout the polymerization. The polymerization time is measured starting when the hydrogen is added. Then the reaction is stopped by adding 5 ml methanol, cooling the reactor and flashing the volatile components. After flushing the reactor twice with N₂ and one vacuum/ N₂ cycle, the product is taken out and dried overnight in a hood. 100g of the polymer is additivated with 0.2 wt% Ionol and 0.1 wt.-% PEPQ (dissolved in acetone) and dried overnight in a hood followed by 2 hours in a vacuum drying oven at 60°C.

Polymerization of propylene with catalyst C4 at 55°C:

[0188] A stirred autoclave (double helix stirrer) with a volume of 21.2 dm³ containing 0.2 barg propylene is filled with additional 3.97 kg propylene. After adding of 0.73 mmol triethylaluminium (Aldrich, 1 molar solution in n-hexane) using a stream of 250 g propylene, the solution is stirred at 20 °C and 250 rpm for 20 min, then the reactor is brought up to the set prepolymerization temperature (HB-Therm) and the catalyst is injected as described in the following. The solid, pre-polymerized catalyst (type, amount and degree of polymerisation as listed in the tables) is loaded into a 5-mL stainless steel vial inside the glovebox, the vial is attached to the autoclave, then a second 5-mL vial containing 4 ml n-hexane and pressurized with 10 bars of N₂ is added on top, the valve between the two vials is opened and the solid catalyst is contacted with hexane under N₂ pressure for 2 s, then flushed into the reactor with 250 g propylene. Stirring speed is increased to 250 rpm and pre-polymerisation is run for the set time. At the end of the prepolymerization step the stirring speed is increased to 350 rpm. Now the target polymerisation temperature of 55°C, respectively, is adjusted. When the internally reactor temperature value is 51°C, respectively, the desired H₂ amount is added with a defined flow via thermal mass flow controller. The reactor temperature is held constant throughout the polymerization. The polymerization time is measured starting when the temperature is 2 °C below the set polymerization temperature. Then the reaction is stopped by adding 5 ml methanol, cooling the reactor and flashing the volatile components.

[0189] After flushing the reactor twice with N₂ and one vacuum/ N₂ cycle, the product is taken out and dried overnight in a hood. 100g of the polymer is additivated with 0.2 wt% Ionol and 0.1 wt.-% PEPQ(dissolved in acetone) and dried overnight in a hood followed by 2 hours in a vacuum drying oven at 60°C.

[0190] The relevant polymerisation conditions and polymerisation results of the three PP homopolymers prepared by liquid propylene polymerization at with catalyst C2 at 30°C (inventive) and with catalyst C4 at 55°C are described in Table 10 and Table 11.

[0191] All homopolymers have been additivated with 1500 ppm of B225 and 500 ppm of CaSt, and pelletized on a twin-screw extruder Prism TSE-16TC.

Table 10:

Sample ID	catalyst	catalyst amount	propylene	T	H ₂	time	yield
		[mg]	[g]	[°C]	[NL]	[min]	[g]
9	C2	188.5	4436	30	0.40	360	716
2	C4	102.0	4421	55	2.72	134	1459

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Table 11:

Sample ID	catalyst	T _c	T _m	[2,1e]	[2,1t]	[3,1]	[mrrm]	total defects	Tensile modulus	Tensile strain at break	Work at break
		[°C]	[°C]	[%]	[%]	[%]	[%]	[%]	[%]	[%]	[J]
9	C2	93.3	132.0	3.3	0	0	0.2	3.5	1113	611	21.0
2	C4	103.2	142.5	2.0	0	0	0.1	2.1	1639	415	9.7

Mechanical characterization: Tensile

[0192] Mechanical characterisation was performed as defined above. The results of the tensile measurements show that the polypropylene homopolymer of the invention has a much higher toughness as measured by the work at break value (work necessary to break the polymer specimen), higher ductility as measured by the tensile strain at break and lower tensile modulus, compared to the propylene homopolymers produced with the same catalyst but produced at higher polymerisation temperature.

10 **Claims**

1. Polypropylene homopolymer (H-PP1), having

15 a) a melting temperature T_m of less than 150 °C,
 b) 2,1 regio-defects of above 2.0 % determined by ^{13}C -NMR spectroscopy, and
 c) a weight average molecular weight M_w of above 100 kg/mol.

2. Polypropylene homopolymer (H-PP1) according to claim 1, having

20 a) a melting temperature T_m of less than 145 °C;
 and/or
 b) 2,1 erythro regio-defects of above 2.0 % determined by ^{13}C -NMR spectroscopy,
 and/or
 c) a weight average molecular weight M_w of above 200 kg/mol.

25 3. Polypropylene homopolymer (H-PP1) according to claim 1 or 2, having

30 a) a melting temperature T_m of less than 140 °C;
 and/or
 b) 2,1 regio-defects and/or 2,1 erythro regio-defects of above 2.0 % determined by ^{13}C -NMR spectroscopy,
 and/or
 c) a weight average molecular weight M_w of above 300 kg/mol.

35 4. Polypropylene homopolymer (H-PP1) according to any one of claims 1 to 3, having

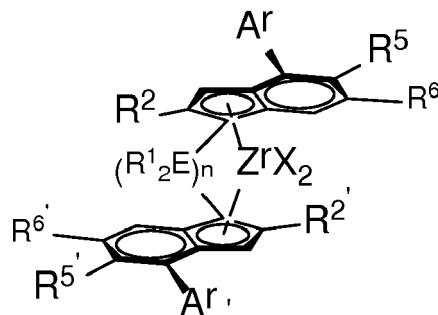
35 a) a melting temperature T_m of from 120 to 142 °C;
 and/or
 b) 2,1 regio-defects and/or 2,1 erythro regio-defects are above 2.0 to 3.5 % determined by ^{13}C -NMR spectroscopy,
 and/or
 c) a weight average molecular weight M_w of from 300 to 2,000 kg/mol.

40 5. Polypropylene homopolymer (H-PP1) according to any one of claims 1 to 4, having

45 a) stereo-defects, i.e. a mrrm pentad content, of equal or lower than 1.0 %, determined by ^{13}C -NMR spectroscopy;
 and/or
 b) 3,1 regio-defects of less than 0.6 % determined by ^{13}C -NMR spectroscopy; and/or
 c) a total amount of regio-defects of more than 2.0 % determined by ^{13}C -NMR spectroscopy;
 and/or
 d) a total amount of defects of more than 2.0 % determined by ^{13}C -NMR spectroscopy
 and/or
 e) xylene soluble (XS) fraction content of below 1.0 wt.-%, preferably below 0.5 wt.-%.

50 6. Process for the preparation of a polypropylene homopolymer (H-PP1) according to any one of the preceding claims 1 to 6, comprising the step of polymerizing propylene at a polymerization temperature of equal or below 55 °C in the presence of a solid catalyst system, said solid catalyst system comprises

(i) a complex of formula (I)



5 wherein

15 each X is a sigma ligand;

R¹₂E is a divalent bridge selected from -R¹₂C-, -R¹₂Si-, -R¹₂Ge-, wherein each R¹ is independently a hydrogen atom, C₁-C₂₀ hydrocarbyl, tri(C₁-C₂₀-alkyl)silyl, C₆-C₂₀ aryl, C₇-C₂₀ arylalkyl or C₇-C₂₀ alkylaryl, and n has a value from 1 to 2;

20 R² and R²', which can be the same or different, are a linear C₁-C₂₀ hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC);

Ar and Ar', which can be the same or different, are an aryl or heteroaryl group having up to 20 carbon atoms; R⁵ is hydrogen or a C₁-C₄₀ hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC);

25 R⁵' is a C₁-C₄₀ hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC);

R⁶ and R⁶', which can be the same or different, are a tertiary C₄-C₂₀ hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC); and

30 (ii) a cocatalyst comprising a first organometallic compound of a Group 13 metal (IUPAC).

35 7. The process according to claim 6, wherein

each X is a sigma ligand;

R¹₂E is a divalent bridge selected from -R¹₂C-, -R¹₂Si-, -R¹₂Ge-, wherein each R¹ is independently a hydrogen atom, C₁-C₂₀ hydrocarbyl, tri(C₁-C₂₀-alkyl)silyl, C₆-C₂₀ aryl, C₇-C₂₀ arylalkyl or C₇-C₂₀ alkylaryl, and n has a value from 1 to 2;

40 R² and R²', which can be the same or different, are a linear C₁-C₁₀ hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC);

Ar and Ar', which can be the same or different, are an aryl or heteroaryl group having up to 20 carbon atoms and is substituted in position 4 by R¹;

R⁵ is hydrogen or a C₁-C₂₀ hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC);

R⁵' is a C₁-C₂₀ hydrocarbyl radical optionally containing one or more heteroatoms from groups 14 to 16 (IUPAC); R⁶ and R⁶', which can be the same or different, are a tertiary C₄-C₂₀ hydrocarbyl radical.

45 8. The process according to claim 6 or 7, wherein

each X is a sigma ligand;

R¹₂E is -R¹₂Si-, wherein each R¹ is independently a C₁-C₅-hydrocarbyl, and n has a value of 1; each R² and R²' is a methyl radical;

50 Ar and Ar', which can be the same or different, are an aryl or heteroaryl group having up to 20 carbon atoms and no substituents in positions 3 and 5;

R⁵ is hydrogen or a C₁-C₁₀ hydrocarbyl radical containing one or more heteroatoms from group 16 (IUPAC);

R⁵' is a C₁-C₁₀ hydrocarbyl group containing one or more heteroatoms from group 16 (IUPAC); each R⁶ and R⁶' is a tertiary butyl radical.

55 9. The process according to any one of the preceding claims 6 to 8, wherein the polymerization temperature is from 10 to less than 55°C.

10. A polypropylene blend (PB) comprising a polypropylene homopolymer (H-PP1) as defined in anyone of the preceding

claims 1 to 5 and a second propylene homopolymer (H-PP2), said second propylene homopolymer (H-PP2) has a melting temperature T_m at least 5 °C higher, preferably 8 °C higher, even more preferably at least 10°C higher than the propylene homopolymer (H-PP1).

5 **11.** A polypropylene blend (PB) according to claim 10, wherein the propylene homopolymer (H-PP2) has

- (a) a melting temperature (T_m) of at least 140 C;
and/or
- (b) 2,1 regio-defects and/or 2,1-erythro regio-defects equal or below 1.8 %, more preferably of equal or below 1.6 %;
and/or
- (c) a weight average molecular weight M_w of above 100 kg/mol, preferably of from 100 to 2,000 kg/mol;
and/or
- (d) polydispersity index (M_w/M_n) of at least 2.0, preferably at least 2.2, more preferably from 2.0 to 30.0.

10 **12.** A polypropylene blend (PB) according to claim 10 or 11, wherein

- (a) the propylene homopolymer (H-PP1) and the second propylene homopolymer (H-PP2) are the only polymer components in the polypropylene blend (PB),
and/or
- (b) the weight ratio between the propylene homopolymer (H-PP1) and the second propylene homopolymer (H-PP2) is in the range of 10/90 to 90/10, preferably in the range of 20/80 to 80/20.

15 **13.** A pipe, sheet, film, fiber, moulded article or extrusion coating comprising a polypropylene homopolymer (H-PP1) according to any one of claims 1 to 5 or a polypropylene blend (PB) according to any one of the claims 10 to 12.

20 **14.** The film according to claim 13, wherein the film is a cast, blown or biaxially oriented polypropylene (BOPP) film.

30 Patentansprüche

1. Polypropylen-Homopolymer (H-PP1) mit
 - a) einer Schmelztemperatur T_m von weniger als 150°C;
 - b) 2,1 Regiodefekten von mehr als 2,0 %, bestimmt durch ^{13}C -NMR Spektroskopie, und
 - c) einem Gewichtsmittel des Molekulargewicht M_w von über 100 kg/mol.
2. Polypropylen-Homopolymer (H-PP1) gemäß Anspruch 1, mit
 - a) einer Schmelztemperatur T_m von weniger als 145°C;
und/oder
 - b) 2,1 Erythro-Regiodefekten von über 2,0 %, bestimmt durch ^{13}C -NMR Spektroskopie,
und/oder
 - c) einem Gewichtsmittel des Molekulargewichts M_w von über 200 kg/mol.
3. Polypropylen-Homopolymer (H-PP1) gemäß Anspruch 1 oder 2, mit
 - a) einer Schmelztemperatur T_m von weniger als 140°C;
und/oder
 - b) 2,1 Regiodefekten und/oder 2,1 Erythro-Regiodefekten von über 2,0 %, bestimmt durch ^{13}C -NMR Sepktroskopie,
und/oder
 - c) einem Gewichtsmittel des Molekulargewichts M_w von über 300 kg/mol.
4. Polypropylen-Homopolymer (H-PP1) gemäß einem der Ansprüche 1 bis 3, mit
 - a) einer Schmelztemperatur T_m von 120 bis 142°C;
und/oder

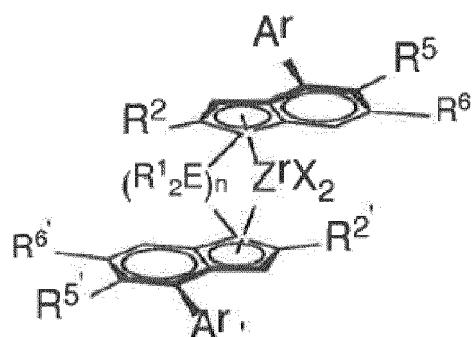
5 b) 2,1 Regiodefekten und/oder 2,1 Erythro-Regiodefekten von über 2,0 bis 3,5 %, bestimmt durch ^{13}C -NMR Spektroskopie,
und/oder
c) einem Gewichtsmittel des Molekulargewichts M_w von 300 bis 2.000 kg/mol.

10 5. Polypropylen-Homopolymer (H-PP1) gemäß einem der Ansprüche 1 bis 4, mit

15 a) Stereodefekten, i.e..mmmm Pentaden, von gleich oder weniger als 1,0 %, bestimmt durch ^{13}C -NMR Spektroskopie;
und/oder
b) 3,1 Regiodefekten von weniger als 0,6 %, bestimmt durch ^{13}C -NMR Spektroskopie;
und/oder
c) einer Gesamtmenge von Regiodefekten von mehr als 2,0 %, bestimmt durch ^{13}C -NMR Spektroskopie;
und/oder
d) einer Gesamtmenge an Defekten von mehr als 2,0 %, bestimmt durch ^{13}C -NMR Spektroskopie;
und/oder
e) einem in Xylol löslichen (XS) Anteil von weniger als 1,0 Gew.-%, vorzugsweise weniger als 0,5 Gew.-%.

20 6. Verfahren zur Herstellung eines Polypropylen-Homopolymers (H-PP1) gemäß einem der vorhergehenden Ansprüche 1 bis 6, umfassend den Schritt des Polymerisierens von Propylen bei einer Polymerisationstemperatur von gleich oder weniger als 55°C, in Gegenwart eines Feststoffkatalysatorsystems, das Feststoffkatalysatorsystem umfassend

25 (i) einen Komplex gemäß Formel (I)



30 wobei

35 jedes X ein Sigmaligand ist;
 R^1E eine divalente Brücke ausgewählt aus $-\text{R}^1_2\text{C}-$, $-\text{R}^1_2\text{Si}-$, $-\text{R}^1_2\text{Geist}$, wobei jedes R^1 unabhängig von-
einander ein Wasserstoffatom, $\text{C}_1\text{-C}_{20}$ Hydrocarbyl, $\text{Tri}(\text{C}_{1\text{-}20}\text{-alkyl})\text{silyl}$, $\text{C}_6\text{-C}_{20}$ Aryl,
 $\text{C}_7\text{-C}_{20}$ Arylalkyl oder $\text{C}_7\text{-C}_{20}$ Alkylaryl ist, und n einen Wert von 1 bis 2 hat;

40 R^2 und R^2' , welche gleich oder unterschiedlich sein können, lineare $\text{C}_1\text{-C}_{20}$ Hydrocarbylradikale sind, optional
enthaltend ein oder mehrere Heteroatome aus den Gruppen 14 bis 16 (IUPAC);

45 Ar und Ar' , welche gleich oder unterschiedlich sein können, Aryl oder Heteroarylgruppen mit bis zu 20
Kohlenstoffatomen sind;

50 R^5 Wasserstoff oder ein $\text{C}_1\text{-C}_{40}$ Hydrocarbylradikal ist, optional umfassend ein oder mehrere Heteroatome aus den Gruppen 14 bis 16 (IUPAC);

55 R^5' ein $\text{C}_{1\text{-}40}$ Hydrocarbylradikal ist, optional umfassend ein oder mehrere Heteroatome aus den Gruppen 14 bis 16 (IUPAC);

R^6 und R^6' , welche gleich oder unterschiedlich sein können, tertiäre $\text{C}_{4\text{-}20}$ Hydrocarbylradikale sind, optional
umfassend ein oder mehrere Heteroatome aus den Gruppen 14 bis 16 (IUPAC); und

(ii) einen Cokatalysator umfassend eine erste metallorganische Verbindung aus einem Gruppe 13 Metall (IUPAC).

7. Verfahren gemäß Anspruch 6, wobei

jedes X ein Sigmaligand ist;

R¹₂E eine divalente Brücke ausgewählt aus -R¹₂C-, -R¹₂Si-, -R¹₂Ge- ist, wobei jedes R¹ unabhängig voneinander ein Wasserstoffatom, C₁-C₂₀ Hydrocarbyl, Tri(C₁₋₂₀-alkyl)silyl, C₆-C₂₀ Aryl, C₇-C₂₀ Arylalkyl oder C₇-C₂₀ Alkylaryl ist, und n einen Wert von 1 bis 2 hat;

5 R² und R²', welche gleich oder unterschiedlich sein können, lineare C₁-C₁₀ Hydrocarbylradikale sind, optional umfassend ein oder mehrere Heteroatome aus den Gruppen 14 bis 16 (IUPAC);

Ar und Ar', welche gleich oder unterschiedlich sein können, Aryl- oder Heteroarylgruppen mit bis zu 20 Kohlenstoffatomen sind und in Position 4 mit R¹ substituiert sind;

10 R⁵ Wasserstoff oder ein C₁-C₂₀ Hydrocarbylradikal ist, optional umfassend ein oder mehrere Heteroatome aus Gruppen 14 bis 16 (IUPAC);

R⁵' ein C₁-C₂₀ Hydrocarbylradikal ist, optional umfassend ein oder mehrere Heteroatome aus den Gruppen 14 bis 16 (IUPAC);

R⁶ und R⁶', welche gleich oder unterschiedlich sein können, tertiäre C₄-C₂₀ Hydrocarbylradikale sind.

15 8. Das Verfahren gemäß nach Anspruch 6 oder 7, wobei

jedes X ein Sigmaligand ist;

R¹₂E -R¹₂Si- ist, wobei jedes R¹ unabhängig voneinander ein C₁₋₅-Hydrocarbyl ist und n einen Wert von 1 hat;

jedes R² und R²' ein Methylradikal ist;

20 Ar und Ar', welche gleich oder unterschiedlich sein können, eine Aryl- oder Heteroarylgruppe mit bis zu 20 Kohlenstoffatomen sind und keinen Substituenten in den Positionen 3 und 5 aufweisen;

R⁵ Wasserstoff oder ein C₁-C₁₀ Hydrocarbylradikal ist, umfassend ein oder mehrere Heteroatome aus der Gruppe 16 (IUPAC);

R⁵' eine C₁-C₁₀ Hydrocarbylgruppe ist, umfassend ein oder mehrere Heteroatome aus der Gruppe 16 (IUPAC);

jedes R⁶ und R⁶' ein tertiäres Butylradikal ist.

25 9. Verfahren gemäß einem der vorhergehenden Ansprüche 6 bis 8, wobei die Polymerisationstemperatur von 10 bis weniger als 55°C liegt.

30 10. Polypropylenmischung (PB) umfassend ein Polypropylen-Homopolymer (H-PP1) gemäß einem der vorhergehenden Ansprüche 1 bis 5 und ein zweites Propylen-Homopolymer (H-PP2), wobei das zweite Propylene-Homopolymer (H-PP2) eine Schmelztemperatur T_m aufweist, die mindestens 5 °C höher ist, vorzugsweise 8 °C höher ist, weiter bevorzugt mindestens 10 °C höher ist, als die des Propylen-Homopolymers (H-PP1).

35 11. Polypropylenmischung (PB) gemäß Anspruch 10, wobei das Propylen-Homopolymer (H-PP2)

(a) eine Schmelztemperatur T_m von mindestens 140 °C;

und/oder

(b) 2,1 Regiodefekte und/oder 2,1 Erythro-Regiodefekte gleich oder weniger 1,8 %, vorzugsweise gleich oder weniger 1,6 %;

und/oder

(c) ein Gewichtsmittel des Molekulargewichts M_w von über 100 kg/mol, vorzugsweise von 100 bis 2.000 kg/mol;

und/oder

(d) einen Polydispersitätsindex (M_w/M_n) von mindestens 2,0, vorzugsweise mindestens 2,2, weiter bevorzugt von 2,0 bis 30,0 hat.

45 12. Polypropylenmischung (PB) gemäß Anspruch 10 oder 11, wobei

(a) das Propylen-Homopolymer (H-PP1) und das zweite Propylen-Homopolymer (H-PP2) die einzigen Polymerbestandteile der Polypropylenmischung (PB) sind,

und/oder

(b) das Gewichtsverhältnis zwischen dem Propylen-Homopolymer (H-PP1) und dem zweiten Propylen-Homopolymer (H-PP2) in dem Bereich von 10/90 bis 90/10 liegt, vorzugsweise in dem Bereich von 20/80 bis 80/20.

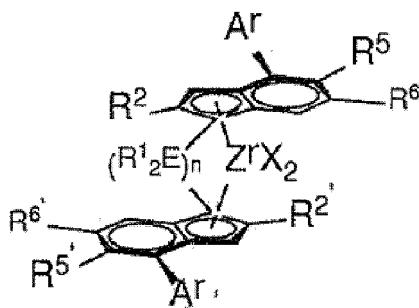
50 13. Rohr, Schicht, Folie, Faser, Pressteil oder Extrusionsbeschichtung umfassend ein Polypropylen-Homopolymer (H-PP1), gemäß einem der Ansprüche 1 bis 5 oder eine Polypropylenmischung (PB) gemäß einem der Ansprüche 10 bis 12.

55 14. Folie gemäß Anspruch 13, wobei die Folie eine Gießfolie, eine Blasfolie oder eine biaxial orientierte Folie (BOPP)

aus Polypropylen ist.

Revendications

- 5 1. Homopolymère de polypropylène (H-PP1)
 - a) présentant un point de fusion T_m inférieur à 150°C,
 - b) comportant des régio-défauts 2,1, déterminés par spectroscopie RMN- ^{13}C , à raison de plus de 2,0 %, et
 - 10 c) présentant une masse moléculaire moyenne en masse M_w supérieure à 100 kg/mol.
- 15 2. Homopolymère de polypropylène (H-PP1) selon la revendication 1,
 - a) présentant un point de fusion T_m inférieur à 145°C, et/ou
 - 15 b) comportant des régio-défauts 2,1 érythro, déterminés par spectroscopie RMN- ^{13}C , à raison de plus de 2,0 %, et/ou
 - c) présentant une masse moléculaire moyenne en masse M_w supérieure à 200 kg/mol.
- 20 3. Homopolymère de polypropylène (H-PP1) selon la revendication 1 ou 2,
 - a) présentant un point de fusion T_m inférieur à 140°C, et/ou
 - b) comportant des régio-défauts 2,1 et/ou des régio-défauts 2,1 érythro, déterminés par spectroscopie RMN- ^{13}C , à raison de plus de 2,0 %,
 - 25 et/ou
 - c) présentant une masse moléculaire moyenne en masse M_w supérieure à 300 kg/mol.
4. Homopolymère de polypropylène (H-PP1) selon l'une quelconque des revendications 1 à 3,
 - a) présentant un point de fusion T_m de 120 à 142°C, et/ou
 - 30 b) comportant des régio-défauts 2,1 et/ou des régio-défauts 2,1 érythro, déterminés par spectroscopie RMN- ^{13}C , à raison de plus de 2,0 % à 3,5 %,
 - et/ou
 - c) présentant une masse moléculaire moyenne en masse M_w de 300 à 2000 kg/mol.
- 35 5. Homopolymère de polypropylène (H-PP1) selon l'une quelconque des revendications 1 à 4, comportant
 - a) des stéréo-défauts, c'est-à-dire une teneur en pentade mrrm, déterminés par spectroscopie RMN- ^{13}C , à raison de 1,0 % ou moins,
 - 40 et/ou
 - b) des régio-défauts 3,1, déterminés par spectroscopie RMN- ^{13}C , à raison de moins de 0,6 %, et/ou
 - c) une quantité totale de régio-défauts, déterminée par spectroscopie RMN- ^{13}C , supérieure à 2,0 %,
 - et/ou
 - d) une quantité totale de défauts, déterminée par spectroscopie RMN- ^{13}C , supérieure à 2,0 %,
 - et/ou
 - 45 e) une teneur en fraction soluble dans le xylène (XS) inférieure à 1,0 % en poids, de préférence inférieure à 0,5 % en poids.
6. Procédé pour la préparation d'un homopolymère de polypropylène (H-PP1) selon l'une quelconque des revendications 1 à 6, comprenant l'étape de polymérisation de propylène à une température de polymérisation égale ou inférieure à 55°C en présence d'un système catalyseur solide, ledit système catalyseur solide comprenant
 - (i) un complexe de formule (I)



dans laquelle

chaque X est un ligand sigma ;

15 $R^{12}E$ est un pont divalent choisi parmi $-R^{12}C-$, $-R^{12}Si-$, $-R^{12}Ge-$, où chaque R^1 est indépendamment un atome d'hydrogène ou un radical hydrocarbyle en C_1 à C_{20} , tri (alkyle en C_1 à C_{20})silyle, aryle en C_6 à C_{20} , arylalkyle en C_7 à C_{20} ou alkylaryl en C_7 à C_{20} , et n vaut de 1 à 2 ;

20 R^2 et $R^{2'}$, qui peuvent être identiques ou différents, sont des radicaux hydrocarbyle linéaires en C_1 à C_{20} contenant éventuellement un ou plusieurs hétéroatome(s) des Groupes 14 à 16 (IUPAC) ;

25 Ar et Ar' , qui peuvent être identiques ou différents, sont des groupes aryle ou hétéroaryle comportant jusqu'à 20 atomes de carbone ;

R^5 est l'hydrogène ou un radical hydrocarbyle en C_1 à C_{40} contenant éventuellement un ou plusieurs hétéroatome(s) des Groupes 14 à 16 (IUPAC) ;

30 $R^{5'}$ est un radical hydrocarbyle en C_1 à C_{40} contenant éventuellement un ou plusieurs hétéroatomes des Groupes 14 à 16 (IUPAC) ;

35 R^6 et $R^{6'}$, qui peuvent être identiques ou différents, sont des radicaux hydrocarbyle tertiaires en C_4 à C_{20} contenant éventuellement un ou plusieurs hétéroatome(s) des Groupes 14 à 16 (IUPAC) ; et

(ii) un cocatalyseur comprenant un premier composé organométallique d'un métal du Groupe 13 (IUPAC).

30 7. Procédé selon la revendication 6, dans lequel chaque X est un ligand sigma ;

$R^{12}E$ est un pont divalent choisi parmi $-R^{12}C-$, $-R^{12}Si-$, $-R^{12}Ge-$, où chaque R^1 est indépendamment un atome d'hydrogène ou un radical hydrocarbyle en C_1 à C_{20} , tri (alkyle en C_1 à C_{20})silyle, aryle en C_6 à C_{20} , arylalkyle en C_7 à C_{20} ou alkylaryl en C_7 à C_{20} , et n vaut de 1 à 2 ;

35 R^2 et $R^{2'}$, qui peuvent être identiques ou différents, sont des radicaux hydrocarbyle linéaires en C_1 à C_{10} contenant éventuellement un ou plusieurs hétéroatome(s) des Groupes 14 à 16 (IUPAC) ;

40 Ar et Ar' , qui peuvent être identiques ou différents, sont des groupes aryle ou hétéroaryle comportant jusqu'à 20 atomes de carbone et substitués en position 4 par R^1 ;

R^5 est l'hydrogène ou un radical hydrocarbyle en C_1 à C_{20} contenant éventuellement un ou plusieurs hétéroatome(s) des Groupes 14 à 16 (IUPAC) ;

45 $R^{5'}$ est un radical hydrocarbyle en C_1 à C_{20} contenant éventuellement un ou plusieurs hétéroatome(s) des Groupes 14 à 16 (IUPAC) ;

R^6 et $R^{6'}$, qui peuvent être identiques ou différents, sont des radicaux hydrocarbyle tertiaires en C_4 à C_{20} .

45 8. Procédé selon la revendication 6 ou 7, dans lequel

chaque X est un ligand sigma ;

$R^{12}E$ est $-R^{12}Si-$ où chaque R^1 est indépendamment un radical hydrocarbyle en C_1 à C_5 , et n vaut 1 ;

chaque R^2 et $R^{2'}$ est un radical méthyle ;

50 Ar et Ar' , qui peuvent être identiques ou différents, sont des groupes aryle ou hétéroaryle comportant jusqu'à 20 atomes de carbone et ne portant pas de substituants aux positions 3 et 5 ;

R^5 est l'hydrogène ou un radical hydrocarbyle en C_1 à C_{10} contenant un ou plusieurs hétéroatome(s) du Groupe 16 (IUPAC) ;

55 $R^{5'}$ est un radical hydrocarbyle en C_1 à C_{10} contenant un ou plusieurs hétéroatome(s) du Groupe 16 (IUPAC) ; chaque R^6 et $R^{6'}$ est un radical tert-butyle.

9. Procédé selon l'une quelconque des revendications 6 à 8, dans lequel la température de polymérisation va de 10 à moins de 55°C.

10. Mélange de polypropylène (PB) comprenant un homopolymère de polypropylène (H-PP1) tel que défini dans l'une quelconque des revendications 1 à 5 et un deuxième homopolymère de propylène (H-PP2), ledit deuxième homopolymère de propylène (H-PP2) présentant un point de fusion T_m supérieur d'au moins 5°C, de préférence supérieur de 8°C, plus préféablement supérieur d'au moins 10°C à celui de l'homopolymère de propylène (H-PP1).

5

11. Mélange de polypropylène (PB) selon la revendication 10, dans lequel l'homopolymère de propylène (H-PP2)

- a) présente un point de fusion T_m d'au moins 140°C, et/ou
- b) comporte des régio-défauts 2,1 et/ou des régio-défauts 2,1 érythro à raison de 1,8 % ou moins, plus préféablement de 1,6 % ou moins, et/ou
- c) présente une masse moléculaire moyenne en masse M_w supérieure à 100 kg/mol, de préférence de 100 à 2000 kg/mol, et/ou
- d) présente un indice de polydispersité (M_w/M_n) d'au moins 2,0, de préférence d'au moins 2,2, plus préféablement de 2,0 à 30,0.

10

12. Mélange de polypropylène (PB) selon la revendication 10 ou 11, dans lequel

- 20 (a) l'homopolymère de propylène (H-PP1) et le deuxième homopolymère de propylène (H-PP2) sont les seuls composants polymères dans le mélange de polypropylène (PB), et/ou
- (b) le rapport en poids entre l'homopolymère de propylène (H-PP1) et le deuxième homopolymère de propylène (H-PP2) est situé dans la plage allant de 10/90 à 90/10, de préférence dans la plage allant de 20/80 à 80/20.

25

13. Tuyau, feuille, film, fibre, article moulé ou revêtement extrudé comprenant un homopolymère de polypropylène (H-PP1) selon l'une quelconque des revendications 1 à 5 ou un mélange de polypropylène (PB) selon l'une quelconque des revendications 10 à 12.

30

14. Film selon la revendication 13, lequel film est un film de polypropylène coulé, soufflé, ou à orientation biaxiale (BOPP).

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REFERENCES CITED IN THE DESCRIPTION

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