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(54) **Title:** POLYPROPYLENE COMPOSITE

(57) **Abstract:** Polyvinyl alcohol fiber reinforced polypropylene composition with excellent impact/stiffness balance as well as to its preparation and use.



Polypropylene composite

The present invention is directed to a polyvinyl alcohol fiber reinforced polypropylene composition with excellent impact/stiffness balance as well as to its preparation and use.

5 Polypropylene is a material used in a wide variety of technical fields, and reinforced polypropylenes have in particular gained relevance in fields previously exclusively relying on non-polymeric materials, in particular metals. For example in automotive parts, engineering plastics have been and still are extensively replaced by polypropylene and polypropylene-based composites, mainly driven by the need to provide lightweight-solutions and to reduce
10 costs.

The addition of reinforcing fibres to polypropylene (PP) adds new design-parameters to polypropylene-based composites targeting the use of such materials in structural and semi-structural parts. Especially PP/glass-fibre (GF)-composites have found widespread use in such applications as they provide a unique combination of comparatively low cost and good
15 stiffness and strength.

The conversion (compounding and injection-moulding) of PP/GF-composites is demanding as it needs to respect the rigid glass-fibre's sensitivity to shear-induced fibre breakage reducing the aspect-ratio and thus the reinforcing potential of the fibres – overall, structure-property-processing correlations in PP/GF-composites tend to be complex.

20 One additional drawback of PP/GF-composites is the significant difference in modulus of the fibre and modulus of the matrix being especially critical in the non-linear irreversible deformation regime where PP shows plastic deformation whereas the glass fibre is still fully elastic. This not only limits ultimate tensile elongation but also affects toughness and instrumented puncture-performance.

25 Moreover, rigid fibres tend to break easily during processing, resulting in overall complex structure-property-processing correlations.

There is additionally a need in the art to have fiber reinforced polypropylene (PP) grades combining an excellent impact/stiffness balance with an increased tenacity. A key parameter
30 in this context is the strain at break (or elongation at break, ϵ_B) which normally is at a very low level, i.e. < 3.0% for PP/GF grades.

As alternative to glass fibers it is known to use organic fibers for reinforcing polypropylene. Such organic fibers can be made of polyamide, polyester, polyimide, cellulose, polyvinyl
35 alcohol, etc..

It is further known that polyvinyl alcohol (PVA) fibers have higher strength, elastic modulus, resistances to weather and chemicals, and adhesiveness than e.g. polyamide and

polyester fibers and have developed unique uses mostly in industrial field, e.g. under the commercial name "Vinylon". In recent years these fibers have caught much attention as reinforcement fiber for cement as a substitute for asbestos fibers.

- 5 From US 20100267888 it is further known to use such PVA fibers for incorporation into a polyolefin composition comprising a polyolefin and a modified polyolefin as coupling agent. It is stated that such PVA fiber containing polyolefin compositions have improved tensile strength and flexural strength.

10 US 20100267888 is absolutely silent about the strain at break and about impact performance.

Accordingly, although much development work has been done in the field of fiber reinforced polypropylene compositions, there still remains the need for further improved fiber reinforced grades.

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Thus, the object of the present invention is to provide a fiber reinforced composition with excellent strain at break and at the same time excellent impact performance.

20 The finding of the present invention is that a fiber reinforced material with excellent strain at break and at the same time excellent impact performance can be obtained with fibers embedded in a polypropylene matrix and not necessarily needing a coupling agent.

Thus the present invention is directed to a fiber reinforced polypropylene composition comprising

- 25 (a) 98.0 to 50.0 wt% of a matrix (M) comprising a polypropylene (PP),
(b) 2.0 to 50.0 wt% of polyvinyl alcohol (PVA) fibers and
(c) 0.0 to 5.0 wt% of a polar modified polypropylene as coupling agent (CA),
based on the total weight of the fiber reinforced composition,
wherein the sum of (a), (b) and (c) is 100.0 wt% and wherein the composition
30 (i) has a tensile strain at break measured at 23°C according to ISO 527-2 (cross head speed 50 mm/min) of at least 8% and
(ii) a Charpy notched impact strength at 23 °C ISO 179-1eA:2000 of at least 10.0 kJ/m².

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Ad matrix (M)

The term "matrix" in the meaning of the present invention is to be interpreted in its commonly accepted meaning, i.e. it refers to a continuous phase (in the present invention a continuous polymer phase) in which isolated or discrete particles such as fibers are dispersed. The matrix (M) is present in such an amount so as to form a continuous phase which can act as a matrix.

In one embodiment the polypropylene (PP) of the matrix (M) is a propylene homopolymer (H-PP1) having a melt flow rate MFR₂ (230 °C) measured according to ISO 1133 of from 1 to 500 g/10min, preferably of from 2 to 300 g/10min, still more preferably of from 5 to 100 g/10min and most preferably of from 8 to 80 g/10 min.

According to another embodiment of the present invention, the polypropylene (PP) of the matrix (M) is a heterophasic propylene copolymer (HECO) comprising a polypropylene matrix (M-HECO). Preferably the polypropylene matrix (M-HECO) is a propylene homopolymer (H-PP2), and dispersed therein is an elastomeric propylene copolymer (E) comprising units derived from propylene and ethylene and/or C₄ to C₈ α-olefin.

It is preferred that such a heterophasic propylene copolymer (HECO) has

- a) a xylene cold soluble content (XCS) measured according ISO 6427 (23 °C) in the range of 8.0 to 35 wt%, and/or
- b) a melt flow rate MFR₂ (230 °C) measured according to ISO 1133 of from 1 to 300 g/10min, and/or
- c) a total ethylene and/or C₄ to C₈ α-olefin content of 5.0 to 25 wt%, based on the total weight of the heterophasic propylene copolymer (HECO).

Preferably, the polypropylene (PP), i.e. the propylene homopolymer (H-PP1) and/or the heterophasic propylene copolymer (HECO), has a melting temperature T_m of equal to or below 175 °C, more preferably of below 170 °C, like of equal or below 168 °C. For example, the melting temperature ranges from 130 to 175 °C, more preferably ranges from 140 to 170 °C and most preferably ranges from 150 to 168 °C.

As mentioned above, in one embodiment of the present invention, the polypropylene (PP) is a propylene homopolymer (H-PP1).

The expression propylene homopolymer as used throughout the instant invention relates to a polypropylene that consists substantially, i.e. of more than 99.5 wt%, still more preferably of

at least 99.7 wt%, like of at least 99.8 wt%, of propylene units. In a preferred embodiment only propylene units in the propylene homopolymer are detectable.

5 Preferably, the propylene homopolymer (H-PP1) has a melting temperature T_m in the range of 150 to 175 °C, more preferably in the range of 155 to 170 °C and most preferably in the range of 158 to 168 °C.

10 The propylene homopolymer (H-PP1) is preferably an isotactic propylene homopolymer. Accordingly, it is appreciated that the polypropylene matrix (H-PP1) has a rather high isotactic pentad concentration, i.e. higher than 90 mol%, more preferably higher than 92 mol%, still more preferably higher than 93 mol% and yet more preferably higher than 95 mol%, like higher than 97 mol%.

15 Furthermore, the propylene homopolymer (H-PP1) preferably has a xylene cold soluble content (XCS) of not more than 5 wt%, more preferably in the range of 0.1 to 3.5 wt%, still more preferably in the range of 0.5 to 3.0 wt%.

20 The propylene homopolymer (H-PP1) may be produced in the presence of a single-site catalyst, e.g. a metallocene catalyst, or in the presence of a Ziegler-Natta catalyst. The propylene homopolymer (H-PP1) is commercially available and known to the skilled person.

In the other specific embodiment of the present invention, the polypropylene (PP) is a heterophasic propylene copolymer (HECO).

In the following the heterophasic propylene copolymer (HECO) is defined in more detail.

25 Preferably the heterophasic propylene copolymer (HECO) comprises
a) a polypropylene matrix (M-HECO), and
b) an elastomeric propylene or ethylene copolymer (E).

30 The expression "heterophasic" indicates that the elastomeric copolymer (E) is preferably (finely) dispersed at least in the polypropylene matrix (M-HECO) of the heterophasic propylene copolymer (HECO). In other words the elastomeric copolymer (E) forms inclusions in the polypropylene matrix (M-HECO). Thus, the polypropylene matrix (M-HECO) contains (finely) dispersed inclusions being not part of the matrix and said inclusions contain the elastomeric copolymer (E). The term "inclusion" according to this invention shall preferably
35 indicate that the matrix and the inclusion form different phases within the heterophasic propylene copolymer (HECO), said inclusions are for instance visible by high resolution microscopy, like electron microscopy or scanning force microscopy.

Furthermore, the heterophasic propylene copolymer (HECO) preferably comprises as polymer components only the polypropylene matrix (M-HECO) and the elastomeric copolymer (E). In other words the heterophasic propylene copolymer (HECO) may contain further additives but no other polymer in an amount exceeding 5 wt%, more preferably exceeding 3 wt%, like exceeding 1 wt%, based on the total heterophasic propylene copolymer (HECO), more preferably based on the polymers present in the heterophasic propylene copolymer (HECO). One additional polymer which may be present in such low amounts is a polyethylene which is a reaction product obtained by the preparation of the heterophasic propylene copolymer (HECO). Accordingly, it is in particular appreciated that a heterophasic propylene copolymer (HECO) as defined in the instant invention contains only a polypropylene matrix (M-HECO), an elastomeric copolymer (E) and optionally a polyethylene in amounts as mentioned in this paragraph.

The elastomeric copolymer (E) is preferably an elastomeric ethylene copolymer (E1) and/or an elastomeric propylene copolymer (E2), the latter being preferred.

As explained above a heterophasic propylene copolymer (HECO) comprises a polypropylene matrix (M-HECO) in which the elastomeric copolymer (E) is dispersed.

The polypropylene matrix (M-HECO) can be a propylene homopolymer (H-PP2) or a propylene copolymer (C-PP1).

However, it is preferred that the propylene matrix (M-HECO) is a propylene homopolymer (H-PP2).

The polypropylene matrix (M-HECO) being a propylene homopolymer (H-PP2) is preferably an isotactic propylene homopolymer. Accordingly it is appreciated that the propylene homopolymer (H-PP2) has a rather high pentad concentration, i.e. higher than 90 mol%, more preferably higher than 92 mol%, still more preferably higher than 93 mol% and yet more preferably higher than 95 mol%, like higher than 99 mol%.

The polypropylene matrix (M-HECO) being a propylene homopolymer (H-PP2) has a rather low xylene cold soluble (XCS) content, i.e. of not more than 3.5 wt%, preferably of not more than 3.0 wt%, like not more than 2.6 wt%, based on the total weight of the polypropylene matrix (M-HECO). Thus, a preferred range is 0.5 to 3.0 wt%, more preferred 0.5 to 2.5 wt%, still more preferred 0.7 to 2.2 wt% and most preferred 0.7 to 2.0 wt%, based on the total weight of the propylene homopolymer (H-PP2).

In one embodiment of the present invention, the polypropylene matrix (M-HECO) is a propylene homopolymer (H-PP2) having a melt flow rate MFR2 (230 °C) from 1 to 500

g/10min, more preferably of from 2 to 300 g/10min, still more preferably of from 5 to 100 g/10min and most preferably of from 8 to 80 g/10 min.

Preferably, the propylene homopolymer (H-PP2) has a melting temperature T_m in the range of 150 to 175 °C, more preferably in the range of 155 to 170 °C and most preferably in the range of 158 to 168 °C.

The second component of the heterophasic propylene copolymer (HECO) is the elastomeric copolymer (E). As mentioned above the elastomeric copolymer (E) can be an elastomeric ethylene copolymer (E1) and/or an elastomeric propylene copolymer (E2). In the following both elastomers are defined more precisely.

Preferably the elastomeric ethylene copolymer (E1) comprises units derived from (i) ethylene and (ii) propylene and/or C_4 to C_{20} α -olefins, preferably from (i) ethylene and (ii) selected from the group consisting of propylene, 1-butene, 1-hexene, and 1-octene. Preferably the ethylene content in the elastomeric ethylene copolymer (E1) is at least 50 wt%, more preferably at least 60 wt%. Thus in one preferred embodiment the elastomeric ethylene copolymer (E1) comprises 50.0 to 85.0 wt%, more preferably 60.0 to 78.0 wt%, units derivable from ethylene. The comonomers present in the elastomeric ethylene copolymer (E1) are preferably propylene and/or C_4 to C_{20} α -olefins, like 1-butene, 1-hexene and 1-octene, the latter especially preferred. Accordingly in one specific embodiment elastomeric ethylene copolymer (E1) is an ethylene-1-octene polymer with the amounts given in this paragraph.

In turn the elastomeric propylene copolymer (E2) preferably comprises units derived from (i) propylene and (ii) ethylene and/or C_4 to C_8 α -olefin. Accordingly the elastomeric propylene copolymer (E2) comprises, preferably consists of, units derivable from (i) propylene and (ii) ethylene and/or at least another C_4 to C_6 α -olefin, more preferably units derivable from (i) propylene and (ii) ethylene and at least another α -olefin selected from the group consisting of 1-butene, 1-pentene, 1-hexene, 1-heptene and 1-octene. The elastomeric propylene copolymer (E2) may additionally contain units derived from a non-conjugated diene, however it is preferred that the elastomeric propylene copolymer (E2) consists of units derivable from (i) propylene and (ii) ethylene and/or C_4 to C_8 α -olefins only.

Suitable non-conjugated dienes, if used, include straight-chain and branched-chain acyclic dienes, such as 1,4-hexadiene, 1,5-hexadiene, 1,6-octadiene, 5-methyl-1, 4-hexadiene, 3,7-dimethyl-1,6-octadiene, 3,7-dimethyl-1,7-octadiene, and the mixed isomers of dihydromyrcene and dihydro-ocimene, and single ring alicyclic dienes such as 1,4-cyclohexadiene, 1,5-cyclooctadiene, 1,5-cyclododecadiene, 4-vinyl cyclohexene, 1-allyl-4-

isopropylidene cyclohexane, 3-allyl cyclopentene, 4-cyclohexene and 1-isopropenyl-4-(4-butenyl) cyclohexane. Multi-ring alicyclic fused and bridged ring dienes are also suitable including tetrahydroindene, methyltetrahydroindene, dicyclopentadiene, bicyclo (2,2,1) hepta-2,5-diene, 2-methyl bicycloheptadiene, and alkenyl, alkylidene, cycloalkenyl and cycloalkylidene norbornenes, such as 5-methylene-2-norbornene, 5-isopropylidene norbornene, 5-(4-cyclopentenyl)-2-norbornene; and 5-cyclohexylidene-2-norbornene. Preferred non-conjugated dienes are 5-ethylidene-2-norbornene, 1,4-hexadiene and dicyclopentadiene.

- 10 Accordingly, the elastomeric propylene copolymer (E2) comprises at least units derivable from propylene and ethylene and may comprise other units derivable from a further α -olefin as defined in the previous paragraph. However, it is in particular preferred that the elastomeric propylene copolymer (E2) comprises units only derivable from propylene and ethylene and optionally a non-conjugated diene as defined in the previous paragraph, like
15 1,4-hexadiene. Thus, an ethylene propylene non-conjugated diene monomer polymer (EPDM) and/or an ethylene propylene rubber (EPR) as elastomeric propylene copolymer (E2) are especially preferred, the latter most preferred.

In one embodiment of the present invention, the elastomeric propylene copolymer (E2) is an
20 ethylene propylene rubber (EPR).

Preferably the amount of propylene in the elastomeric propylene copolymer (E2) ranges from 50 to 75 wt%, more preferably 55 to 70 wt%. Thus, in a specific embodiment the elastomeric propylene copolymer (E2) comprises from 25 to 50 wt%, more preferably 30 to 45 wt%, units
25 derivable from ethylene. Preferably, the elastomeric propylene copolymer (E2) is an ethylene propylene non-conjugated diene monomer polymer (EPDM) or an ethylene propylene rubber (EPR), the latter especially preferred, with propylene and/or ethylene content as defined in this paragraph.

30 It is especially preferred that heterophasic propylene copolymer (HECO) comprises a propylene homopolymer (H-PP2) as the polypropylene matrix (M-HECO) and an ethylene propylene rubber (EPR) as the elastomeric propylene copolymer (E2).

Preferably, the heterophasic propylene copolymer (HECO) has a melt flow rate MFR₂ (230
35 °C) of from 1 to 300 g/10 min, more preferably of from 2 to 100 g/10 min, still more preferably of from 3 to 80 g/10min, yet more preferably of from 4 to 40 g/10 min, like in the range of 5 to 30 g/10min.

Such heterophasic propylene copolymers (HECOs) can be produced either by melt mixing of the polypropylene matrix (M-HECO) with the elastomeric copolymer (E) or in-situ by sequential polymerization as is known to an art skilled person, e.g. the matrix (M-HECO) being produced at least in one slurry reactor and subsequently the elastomeric copolymer (E) being produced at least in one gas phase reactor.

Ad polyvinyl alcohol (PVA) fiber

PVA fibers are well known in the art and are preferably produced by a wet spinning process or a dry spinning process.

PVA itself is synthesized from acetylene [74-86-2] or ethylene [74-85-1] by reaction with acetic acid (and oxygen in the case of ethylene), in the presence of a catalyst such as zinc acetate, to form vinyl acetate [108-05-4] which is then polymerized in methanol. The polymer obtained is subjected to methanolysis with sodium hydroxide, whereby PVA precipitates from the methanol solution.

PVA used for the manufacture of fiber generally has a degree of polymerization of not less than 1000, preferably not less than 1200 and more preferably not less than 1500. Most preferably the PVA has a degree of polymerization of around 1700, e.g. 1500 up to 2000.

The degree of hydrolysis of the vinyl acetate is generally at least 99 mol%.

The mechanical properties of PVA fibers vary depending on the conditions of fiber manufacture such as spinning process, drawing process, and acetalization conditions, and the manufacture conditions of raw material PVA.

The PVA fibers can be in the form of (multi)filaments or staple fibers.

PVA fibers are characterized by high strength, low elongation, and high modulus.

Suitable PVA fibers preferably have a tenacity of at least 0.4 N/tex up to 1.7 N/tex, more preferably of at least 0.6 N/tex up to 1.4 N/tex and most preferably of at least 0.7 N/tex up to 1.0 N/tex.

Furthermore such fibers preferably have a Young Modulus in the range of 3.0 up to 35.0 N/tex, preferably in the range of 10.0 to 30.0 N/tex and more preferably in the range of 15.0 to 25.0 N/tex (ISO 5079).

PVA fibers being suitable for the present invention have a fiber length of 2.0 to 20 mm, preferably of 2.5 to 15 mm, more preferably from 3.0 to 10 mm and most preferably from 3.5 to 6.0 mm.

The fiber average diameter of suitable PVA fibers is in the range of 10 to 20 μm , preferably in the range of 12 to 18 μm .

PVA fibers being suitable for the present invention are furthermore surface treated with a so called sizing agent. This can be done with known methods, like for example immersing the fibers in a tank in which a sizing agent is placed, being nipped and then drying in a hot-air oven, or with a hot roller or a hot plate.

Example of sizing agents include polyolefin resin, polyurethane resin, polyester resin, acrylic resin, epoxy resin, starch, vegetable oil, modified polyolefin.

The amount of the sizing agent related to the polyvinyl alcohol fibers is within the common knowledge of an art skilled person and can be, for example in the range of is 0.1 to 10 parts by weight of the sizing agent with respect to 100 parts by weight of the polyvinyl alcohol fibers.

A surface treating agent may be incorporated in the sizing agent to improve the wettability or adhesiveness between the polyvinyl alcohol fibers and the polypropylene composition.

Examples of the surface treating agent include silane coupling agents, titanate coupling agents, aluminum coupling agents, chromium coupling agents, zirconium coupling agents, borane coupling agents, and preferred are silane coupling agents or titanate coupling agents, and more preferably silane coupling agents.

Ad polar modified polypropylene coupling agent (CA)

The fiber reinforced polypropylene composition optionally may comprise a coupling agent (CA).

Suitable coupling agents (CA) are polar modified propylene homopolymers and copolymers, like copolymers of propylene with ethylene and or with other α -olefins, as they are highly compatible with the polymers of the fiber reinforced composition.

Preference is given to modified polypropylenes containing groups deriving from polar compounds, in particular selected from the group consisting of acid anhydrides, carboxylic acids, carboxylic acid derivatives, primary and secondary amines, hydroxyl compounds, oxazoline and epoxides.

Specific examples of the said polar compounds are unsaturated cyclic anhydrides and their aliphatic diesters, and the diacid derivatives. In particular, one can use maleic anhydride and compounds selected from C_1 to C_{10} linear and branched dialkyl maleates, C_1 to C_{10} linear and branched dialkyl fumarates, itaconic anhydride, C_1 to C_{10} linear and branched itaconic acid dialkyl esters, maleic acid, fumaric acid, itaconic acid and mixtures thereof.

Particular preference is given to using a propylene polymer grafted with maleic anhydride as the modified polymer, i.e. as the coupling agent (CA).

The modified polymer, i.e. the coupling agent (CA), can be produced in a simple manner by reactive extrusion of the polymer, for example with maleic anhydride in the presence of free radical generators (like organic peroxides), as disclosed for instance in EP 0 572 028.

The amounts of groups deriving from polar compounds in the modified polymer, i.e. the coupling agent (CA), are from 0.5 to 5.0 wt%, preferably from 0.5 to 4.0 wt%, and more preferably from 0.5 to 3.0 wt%.

Preferred values of the melt flow rate MFR2 (230°C) for the modified polymer, i.e. for the coupling agent (CA), are from 1.0 to 500 g/10 min.

Ad fiber reinforced polypropylene composition

The fiber reinforced polypropylene composition of the present invention therefore comprises (a) 98.0 to 50.0 wt% of a matrix (M) comprising a polypropylene (PP) as described above, (b) 2.0 to 50.0 wt% of polyvinyl alcohol (PVA) fibers, as described above and (c) 0.0 to 5.0 wt% of a polar modified polypropylene as coupling agent (CA), based on the total weight of the fiber reinforced composition, wherein the sum of (a), (b) and (c) is 100.0 wt%.

Preferably the fiber reinforced polypropylene composition comprises (a) 95.0 to 55.0 wt% of a matrix (M) comprising a polypropylene (PP) as described above, (b) 5.0 to 45.0 wt% of polyvinyl alcohol (PVA) fibers, as described above and (c) 0.0 to 2.5 wt% of a polar modified polypropylene as coupling agent (CA), based on the total weight of the fiber reinforced composition, wherein the sum of (a), (b) and (c) is 100.0 wt%.

More preferably the fiber reinforced polypropylene composition comprises (a) 92.0 to 60.0 wt% of a matrix (M) comprising a polypropylene (PP) as described above, (b) 8.0 to 40.0 wt% of polyvinyl alcohol (PVA) fibers, as described above and (c) 0.0 to 2.5 wt% of a polar modified polypropylene as coupling agent (CA), based on the total weight of the fiber reinforced composition, wherein the sum of (a), (b) and (c) is 100.0 wt%.

Most preferably the fiber reinforced polypropylene composition comprises (a) 90.0 to 65.0 wt% of a matrix (M) comprising a polypropylene (PP) as described above,

(b) 10.0 to 35.0 wt% of polyvinyl alcohol (PVA) fibers, as described above and
(c) 0.0 to 2.5 wt% of a polar modified polypropylene as coupling agent (CA),
based on the total weight of the fiber reinforced composition,
wherein the sum of (a), (b) and (c) is 100.0 wt%.

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In addition to the above described components, the instant composition may additionally
contain typical other additives useful for instance in the automobile sector, like carbon black,
other pigments, antioxidants, UV stabilizers, nucleating agents, antistatic agents and slip
agents, in amounts usual in the art, providing that the overall sum of (a), (b), (c) and other
additive is 100.0 wt%.

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The additives as stated above are added to the polypropylene (PP), which is either collected
from the final reactor of the polymer production process or in case of heterophasic propylene
copolymer (HECO) also to the polymer obtained by melt-mixing.

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Preferably, these additives are mixed into the polypropylene (PP) or during the extrusion
process in a one-step compounding process. Alternatively, a master batch may be
formulated, wherein the polypropylene (PP) is first mixed with only some of the additives.

For mixing the individual components of the instant fiber reinforced composition, a
conventional compounding or blending apparatus, e.g. a Banbury mixer, a 2-roll rubber mill,
Buss-co-kneader or a twin screw extruder may be used. Preferably, mixing is accomplished
in a co-rotating twin screw extruder. The polymer materials recovered from the extruder are
usually in the form of pellets. These pellets are then preferably further processed, e.g. by
injection molding to generate articles and products of the inventive fiber reinforced
composition.

25

The fiber reinforced polypropylene composition according to the invention has the following
properties:

- (i) a tensile strain at break measured at 23°C according to ISO 527-2 (cross head speed 50
mm/min) of at least 8%, preferably of at least 10% and more preferably of at least 12%
and
(ii) a Charpy notched impact strength at 23 °C ISO 179-1eA:2000 of at least 10.0 kJ/m²,
preferably of at least 12 kJ/m² and more preferably of at least 15 kJ/m².

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Additionally the fiber reinforced polypropylene composition according to the invention has a
tensile strength measured at 23°C according to ISO 527-2 (cross head speed 50 mm/min) of
at least 35 MPa, preferably of at least 40 MPa and more preferably of at least 50 MPa.

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Thus, the fiber reinforced polypropylene composition show an improved strain at break and tensile strength balance and additionally excellent impact performance.

The polypropylene composition of the present invention is preferably used for the production of articles, especially automotive articles, like moulded automotive articles, preferably automotive injection moulded articles. Even more preferred is the use for the production of car interiors and exteriors, like bumpers, side trims, step assists, body panels, spoilers, dashboards, interior trims and the like.

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

In addition, the present invention also relates to a process for the preparation of the fiber reinforced composition as described above, comprising the steps of adding

- (a) polypropylene (PP),
 - (b) the polyvinyl alcohol (PVA) fibers , and
 - (c) optionally the polar modified polypropylene as coupling agent (CA)
- to an extruder and extruding the same obtaining said fiber reinforced composition.

EXPERIMENTAL PART:**A) METHODS****Tensile tests:**

- 5 The tensile strength and the tensile strain at break were measured at 23 °C according to ISO 527-2 (cross head speed 50 mm/min) using injection moulded specimens moulded at 230 °C according to ISO 527-2(1A), produced according to EN ISO 1873-2 (dog bone shape, 4 mm thickness).
- 10 **Charpy impact test:** The Charpy notched impact strength (NIS) was measured according to ISO 179-1eA:2000 at +23 °C, using injection-molded bar test specimens of 80x10x4 mm³ prepared in accordance with ISO 1873-2:2007.

B) MATERIALS USED**15 For matrix (M):****PP-homopolymer (PP-H):**

HJ325MO: a polypropylene homopolymer containing nucleating and antistatic additives, provided by Borealis. (CAS-No: 9003-07-0)

- 20 This polymer is a CR (controlled rheology) grade with narrow molecular weight distribution, density of 905 kg/m³ (ISO1183) and an MFR₂ of 50 g/10 min (230°C; 2.16 kg; ISO 1133); XS of 2.2 wt% and melting temperature of 164°C and a Charpy Notched Impact Strength at 23°C of 2.0 kJ/m²

Heterophasic copolymer (PP-HECO):

- 25 BG055Al: nucleated high crystallinity PP impact copolymer having an MFR (230°C/2,16kg) of 22 g/10min, an elastomer content of 18 wt% as determined by the content of xylene solubles (XS) and a density of 920 kg/m³. The polymer contains 2 wt% of talc, based on the total weight of the polymer.
- Tensile Strength [MPa] is 35 MPa, Charpy Notched Impact Strength at 23°C is 3.5 kJ/m²;
- 30 Tensile Strain at Break is 32%.

PVA fibers:

- Chopped PVA-fibres Mewlon 2000T-750F HM1 (High Modulus), fibre-length 4mm, tenacity of 1N/tex, Young Modulus of 21.5 N/tex with a specific surface-treatment for PP, supplied by
- 35 Unitika

Coupling agent (CA):

commercial maleic anhydride functionalized polypropylene "Scona TPPP 8112FA" of
5 Kometra GmbH, Germany with a density of 0.9 g/cm³, having an MFR₂ (190°C; 2.16 kg) of
100 g/10min and an MAH content of 1.4 mol%.

Comparative Example 1 (CE1)

GB205U is a commercially glass fibre reinforced composite of Borealis AG containing 20
10 wt% chemically coupled glass fibers embedded in a propylene homopolymer matrix, having
an MFR₂ (230 °C) of 2.2 g/10min and a melting temperature of 166 °C.

EXAMPLES:

PP-compositions were produced using a parallel, co-rotating twin-screw extruder Brabender
15 DSE20 (screw-diameter 20mm, length 40d) with four vertical ports at 0, 10, 20 and 30d
which can be used for dosing and venting. All four downstream-zones were electrically
heated and water-cooled. The extruder has two side-feeders at 11 and 22d. Polymer and
additives were fed through vp1, vp2 and vp4 were used for atmospheric venting (vp3
remained closed). The fibres were fed through sp1 via the side-feeder.

20 The standard compounding-melt-temperature was 190°C.

A water-bath, cooled to about 15°C, and a strand pelletizer Primo 50 by Rieter were used to
granulate the melt extruded through a die with a diameter of 3.0 mm. All components were
fed via Motan-Colortronic gravimetric dosing scales. Single screw scales were employed for
the polymer and the MA-PP. The fibres were fed via a GBS-C twin screw system.

25

From Table 1 the composition of the Inventive Examples 1 to 5 (IE1 to IE5) can be seen:

Table 1:

		IE1	IE2	IE3	IE4	IE5
Matrix (M)	[wt%]					
PP-H		78.0		90.0	80.0	70.0
PP-HECO			78.0			
Fibers	[wt%]					
PVA-fiber		20.0	20.0	10.0	20.0	30.0
Tencel-fiber						
Coupling Agent (CA)	[wt%]					
Scona TPPP		2.0	2.0	0.0	0.0	0.0

- 5 From Table 2 the properties of the Inventive Examples 1 to 5 (IE1 to IE5) and of Comparative Example 1 can be seen:

Table 2:

		IE1	IE2	IE3	IE4	IE5	CE1
Tensile Strength	[MPa]	68	57	45	56	59	89
Tensile Strain at Break	%	13.3	12.4	14.2	13.2	12.7	3.4
Charpy Notched (23°C)	kJ/m ²	29.8	16.9	12.7	30.4	48.9	10.5

10

From this table and from Figures 1 to 5 it can be clearly seen that the compositions of the invention, i.e. the PVA-fiber reinforced PP compositions have an improved strain@break and tensile strength balance in combination with excellent impact performance, even if no coupling agent is used.

- 15 The advantages of the compositions of the invention, i.e. the PVA-fiber reinforced PP compositions over comparable compositions (using glass fibers) can be furthermore seen in the determined notched impact performance which is superior to conventional homo PP based GF composites.

Claims:

1. Fiber reinforced polypropylene composition comprising
 - (a) 98.0 to 50.0 wt% of a matrix (M) comprising a polypropylene (PP),
 - (b) 2.0 to 50.0 wt% of polyvinyl alcohol (PVA) fibers and
 - 5 (c) 0.0 to 5.0 wt% of a polar modified polypropylene as coupling agent (CA),
based on the total weight of the fiber reinforced composition,
wherein the sum of (a), (b) and (c) is 100.0 wt% and wherein the composition
 - (i) has a tensile strain at break measured at 23°C according to ISO 527-2 (cross head
speed 50 mm/min) of at least 8% and
 - 10 (ii) a Charpy notched impact strength at 23 °C ISO 179-1eA:2000 of at least 10.0 kJ/m².
2. Fiber reinforced polypropylene composition according to claim 1, wherein the
polypropylene (PP) of the matrix (M) is a propylene homopolymer (H-PP1) having
 - (i) a melt flow rate MFR₂ (230°C) measured according to ISO 1133 of from 1 to 500
 - 15 g/10min, (ii) a melting temperature T_m in the range of 150 to 175°C,
 - (iii) a isotactic pentad concentration of higher than 90 mol% and
 - (iv) a xylene cold soluble content (XCS) of not more than 5 wt%,
3. Fiber reinforced polypropylene composition according to claim 1, wherein the
polypropylene (PP) of the matrix (M) is a heterophasic propylene copolymer (HECO)
having
 - a) a xylene cold soluble content (XCS) measured according ISO 6427 (23°C) in the
range of 8.0 to 35 wt%, and/or
 - b) a melt flow rate MFR₂ (230 °C) measured according to ISO 1133 of from 1 to 300
 - 25 g/10min, and/or
 - c) a total ethylene and/or C₄ to C₈ α-olefin content of 5.0 to 25 wt%, based on the total
weight of the heterophasic propylene copolymer (HECO).
4. Fiber reinforced polypropylene composition according to claim 3 wherein the heterophasic
propylene copolymer (HECO) comprises
 - a) a polypropylene matrix (M-HECO), being a propylene homopolymer (H-PP2), and
 - b) an elastomeric copolymer (E).
5. Fiber reinforced polypropylene composition according to any of the preceding claims 1 to 4
wherein the polyvinyl alcohol (PVA) fibers have
 - (i) a tenacity of at least 0.4 N/tex up to 1.7 N/tex,
 - (ii) a fiber length of 2.0 to 20 mm and

(ii) a fiber average diameter in the range of 10 to 20 μm .

6. Fiber reinforced polypropylene composition according to any of the preceding claims 1 to 5 wherein the polar modified polypropylene as coupling agent (CA) is selected from the group of modified polypropylenes containing groups deriving from polar compounds selected from the group consisting of acid anhydrides, carboxylic acids, carboxylic acid derivatives, primary and secondary amines, hydroxyl compounds, oxazoline and epoxides.

7. Fiber reinforced polypropylene composition according to claim 6, wherein the polar compounds are selected from maleic anhydride and compounds selected from C_1 to C_{10} linear and branched dialkyl maleates, C_1 to C_{10} linear and branched dialkyl fumarates, itaconic anhydride, C_1 to C_{10} linear and branched itaconic acid dialkyl esters, maleic acid, fumaric acid, itaconic acid and mixtures thereof.

8. Fiber reinforced polypropylene composition according to any of the preceding claims 1 to 7, wherein the composition has
has a tensile strain at break measured at 23°C according to ISO 527-2 (cross head speed 50 mm/min) of at least 10% and

(ii) a Charpy notched impact strength at 23 °C ISO 179-1eA:2000 of at least 12.0 kJ/m².

9. Fiber reinforced polypropylene composition according to any of the preceding claims 1 to 8 wherein the composition has a tensile strength measured at 23°C according to ISO 527-2 (cross head speed 50 mm/min) of at least 35 MPa, preferably of at least 40 MPa and more preferably of at least 50 MPa.

10. Articles, comprising the polypropylene composition according to any of the preceding claims 1 to 9.

11. Articles according to claim 10, being automotive articles selected from car interiors and exteriors, like bumpers, side trims, step assists, body panels, spoilers, dashboards, interior trims and the like, comprising at least to 60 wt%, more preferably at least 80 wt%, yet more preferably at least 95 wt%, like consisting, of the polypropylene composition.

12. Process for the preparation of the fiber reinforced composition according to any of the preceding claims 1 to 9. comprising the steps of adding

(a) polypropylene (PP),

(b) the polyvinyl alcohol (PVA) fibers , and

5 (c) optionally the polar modified polypropylene as coupling agent (CA)

to an extruder and extruding the same obtaining said fiber reinforced composition.

Figures:

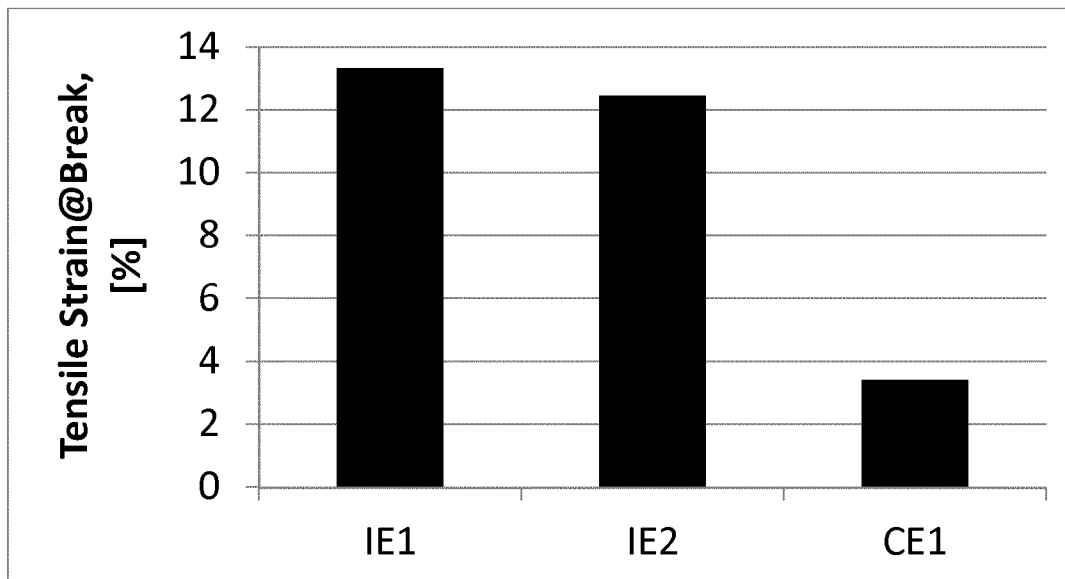


Fig. 1: Comparison of tensile strain@break of IE1, IE2 and CE1.

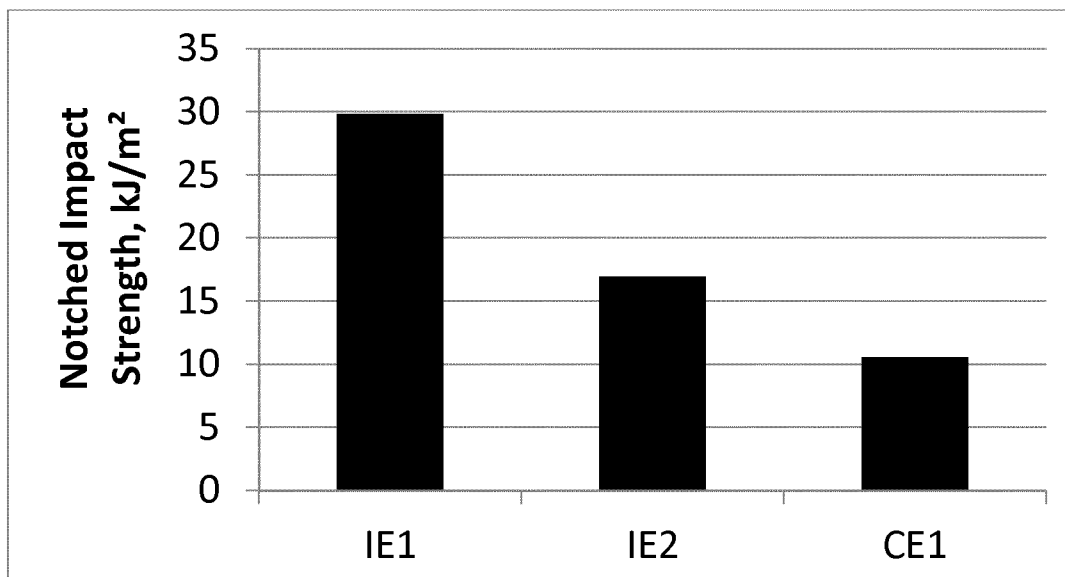


Fig. 2: Comparison of notched impact strength of IE1, IE2 and CE1.

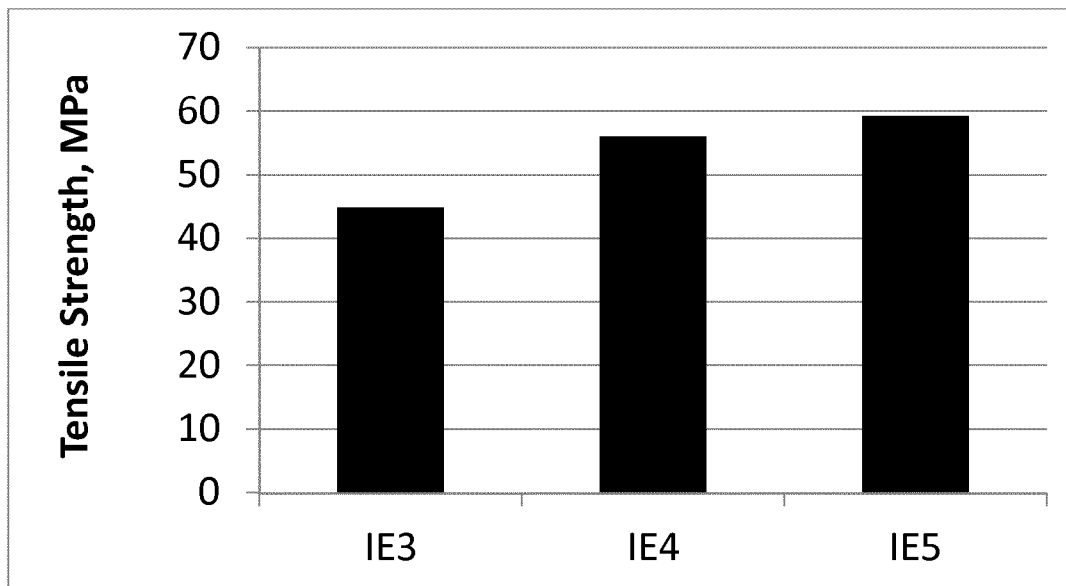


Fig. 3: Tensile strength of IE3, IE4 and IE5.

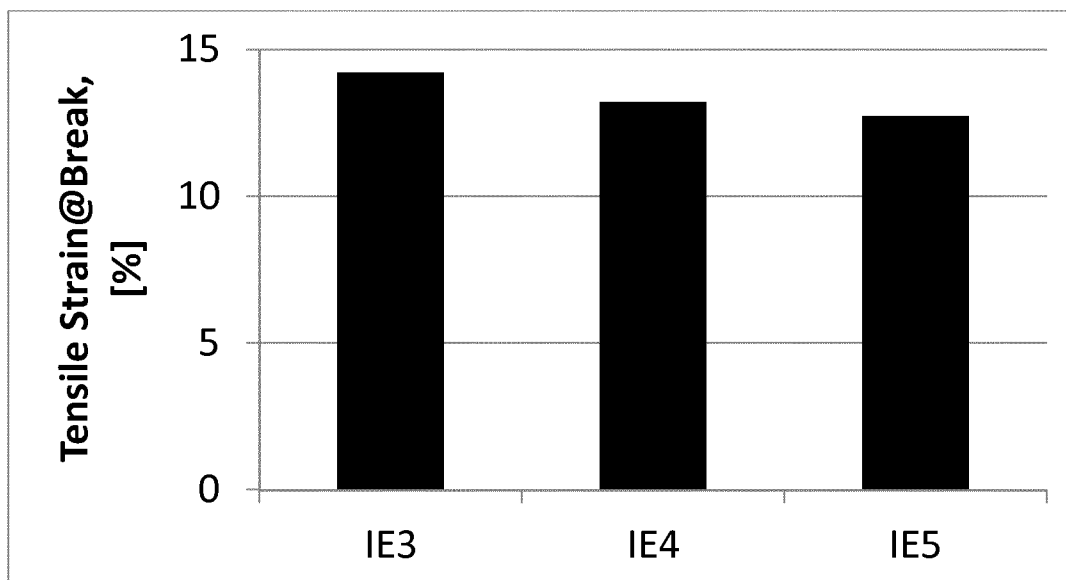


Fig. 4: Tensile strain@break of IE3, IE4 and IE5.

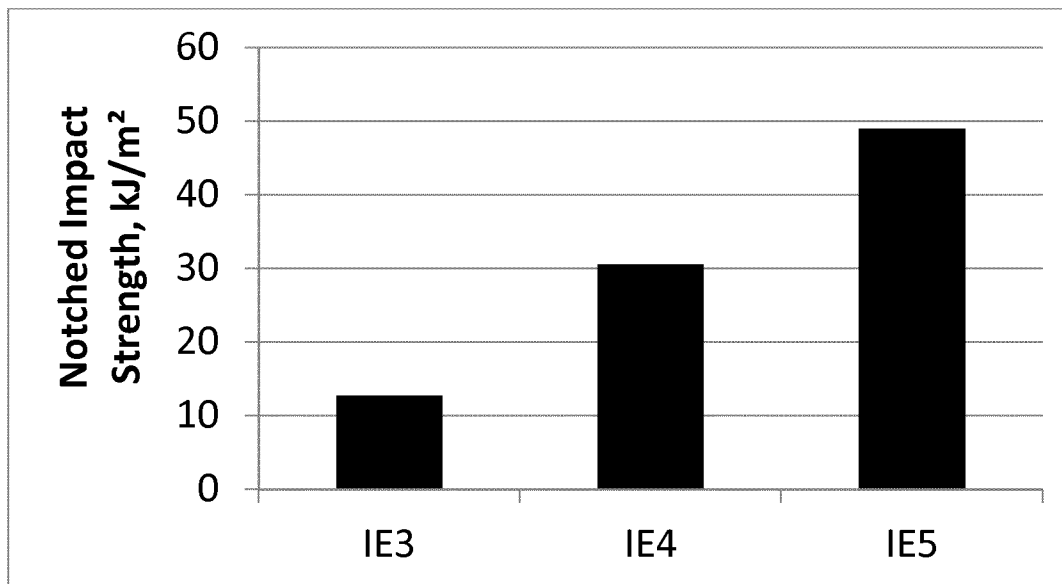


Fig. 5: Notched impact strength of IE3, IE4 and IE5.

PATENT COOPERATION TREATY

PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference AT1427/PCT	FOR FURTHER ACTION see Form PCT/ISA/220 as well as, where applicable, item 5 below.	
International application No. PCT/EP2016/059148	International filing date (<i>day/month/year</i>) 25 April 2016 (25-04-2016)	(Earliest) Priority Date (<i>day/month/year</i>) 27 April 2015 (27-04-2015)
Applicant BOREALIS AG		

This international search report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This international search report consists of a total of 3 sheets.

☒

It is also accompanied by a copy of each prior art document cited in this report.

1. **Basis of the report**

a. With regard to the **language**, the international search was carried out on the basis of:

☒

the international application in the language in which it was filed

☐

a translation of the international application into _____, which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1(b))

b. ☐

This international search report has been established taking into account the **rectification of an obvious mistake** authorized by or notified to this Authority under Rule 91 (Rule 43.6**bis**(a)).

c. ☐

With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, see Box No. I.

2. ☐

Certain claims were found unsearchable (See Box No. II)

3. ☐

Unity of invention is lacking (see Box No III)

4. With regard to the **title**,

☒

the text is approved as submitted by the applicant

☐

the text has been established by this Authority to read as follows:

5. With regard to the **abstract**,

☒

the text is approved as submitted by the applicant

☐

the text has been established, according to Rule 38.2, by this Authority as it appears in Box No. IV. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority

6. With regard to the **drawings**,

a. the figure of the **drawings** to be published with the abstract is Figure No. _____

☐

as suggested by the applicant

☐

as selected by this Authority, because the applicant failed to suggest a figure

☐

as selected by this Authority, because this figure better characterizes the invention

b. ☒

none of the figures is to be published with the abstract

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2016/059148

A. CLASSIFICATION OF SUBJECT MATTER
INV. C08L23/12 C08L23/14
ADD.

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

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
C08L

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

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

EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X A	JP 2008 150414 A (KURARAY CO) 3 July 2008 (2008-07-03) examples 1-3 -----	1,3,4, 8-12 2,5-7
X A	WO 2011/152439 A1 (UNITIKA LTD [JP]; ANDO YASUYUKI; YAMAMURA KIYOHICO; SEJIMA SEIJI; FURU) 8 December 2011 (2011-12-08) examples ce 5-6 -----	1,3,4, 8-12 2,5-7
X A	US 2010/267888 A1 (KATAYAMA TAKASHI [JP] ET AL) 21 October 2010 (2010-10-21) cited in the application examples 1,2, ce 1-3 -----	1,3,4, 8-12 2,5-7
X A	US 2011/263738 A1 (YAMAMOTO YUYA [JP] ET AL) 27 October 2011 (2011-10-27) example 1 -----	1,3,4, 8-12 2,5-7



Further documents are listed in the continuation of Box C.



See patent family annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier application or patent but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search

18 May 2016

Date of mailing of the international search report

27/05/2016

Name and mailing address of the ISA/

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Ritter, Nicola

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/EP2016/059148

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
JP 2008150414	A	03-07-2008	NONE
WO 2011152439	A1	08-12-2011	NONE
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