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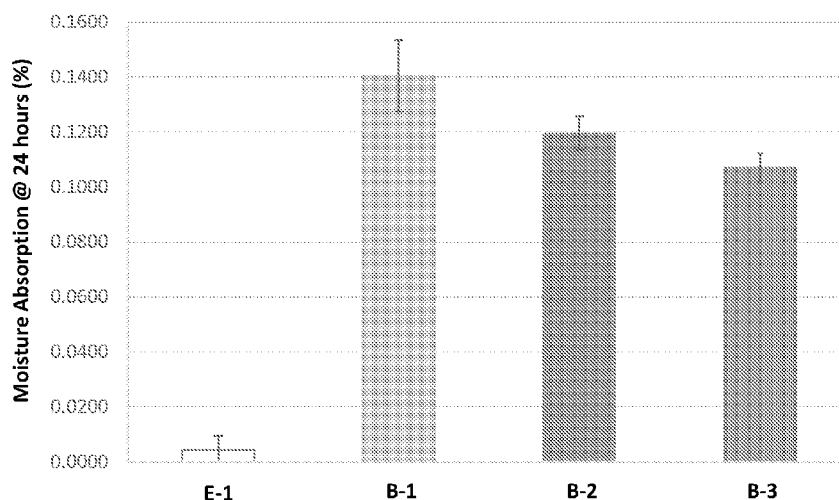


FIGURE 1

(57) **Abstract:** Composite materials are described that comprise a polymer, a lignocellulosic compound, a coupling agent and graphene. The lignocellulosic compound is a shive. When compared with materials formed solely from the neat polymer, the composite materials offer improvements in terms of shrinkage properties, tensile and flexural properties and heat distortion properties. Moreover, the material properties of the composite material can be tailored according to the intended application of the material. Also described is a process for preparing the composite materials, as well as various uses of the composite materials.



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COMPOSITE MATERIAL

INTRODUCTION

[0001] The present invention relates to a composite material comprising a polyolefin and a lignocellulosic compound. The present invention also relates to processes for the preparation of the composite material and uses of the composite material in a variety of structures.

BACKGROUND OF THE INVENTION

[0002] The concept of using naturally derived fibres from plants as reinforcements in composite materials has been implemented since ancient times. However, over the past couple of decades a renewed interest and focus on the advanced utilization of different varieties of naturally occurring fibres as reinforcing agents in polymeric composite materials has been realized worldwide. Emphasis on improving and stimulating rural economies, reducing the global reliance on petroleum-based materials, and promoting sustainability have all been prime factors in revisiting natural fibre reinforced composites. Indeed, governments and think tanks across the globe have begun developing regulatory laws, as well as raising general societal awareness on issues such as pollution, energy consumption, and raw material waste, which has led to increased activity in the field of natural fibre-reinforced composites.

[0003] In the context of natural fibre-reinforced composites, the term *fibre* may refer to either a single elementary fibre or, more commonly, a bundles of these elementary units. Although fibre bundles derived from basts, leaves and grasses have been incorporated into polymeric composites (e.g. Idumah et al (Synthetic Metals 212 (2016) 91-104)), such materials are hampered by the hydrophilic nature of the fibres. In particular, the water uptake tendency of the fibres gives rise to poor compatibility with hydrophobic polymers, which results in the mechanical properties of the composite, e.g. dimensional stability and structural integrity, being compromised. As a consequence, such fibre-reinforced composites are limited in their applications.

[0004] In view of the above, there is a need for new filler-reinforced polymeric composite materials having mechanical properties desirable across a broad range of structural applications.

[0005] The present invention was devised with the foregoing in mind.

SUMMARY OF THE INVENTION

[0006] According to a first aspect of the present invention there is provided a composite material comprising:

25 – 80 wt% of at least one polymer selected from the group consisting of polypropylene, polyethylene, polystyrene, polypropylene-co-ethylene, polypropylene-co-styrene, polyethylene-co-styrene and polypropylene-co-ethylene-co-styrene;
15 – 70 wt% of at least one lignocellulosic compound, wherein the at least one lignocellulosic compound is a shive;
2.0 – 11.0 wt% of at least one coupling agent; and
0.01 – 5.5 wt% of graphene.

[0007] According to a second aspect of the present invention there is provided a process for the preparation of a composite material according to the first aspect, the process comprising the steps of:

A) mixing the at least one polymer, the at least one lignocellulosic compound, the at least one coupling agent and graphene, and
B) extruding the mixture resulting from step A).

[0008] According to a third aspect of the present invention there is provided a composite material obtained, directly obtained or obtainable by a process according to the second aspect.

[0009] According to a fourth aspect of the present invention there is provided a use of a composite material according to the first or third aspect in a component for an automobile, in a container, in a sports article, in an appliance, in an electronic device, or in a consumer good.

DETAILED DESCRIPTION OF THE INVENTION

Composite material

[0010] In a first aspect, the present invention provides a composite material comprising:

25 – 80 wt% of at least one polymer selected from the group consisting of polypropylene, polyethylene, polystyrene, polypropylene-co-ethylene, polypropylene-co-styrene, polyethylene-co-styrene and polypropylene-co-ethylene-co-styrene;
15 – 70 wt% of at least one lignocellulosic compound, wherein the at least one lignocellulosic compound is a shive;
2.0 – 11.0 wt% of at least one coupling agent; and
0.01 – 5.5 wt% of graphene.

[0011] Through detailed studies, the inventor has devised the present composite materials, which surprisingly offer a wealth of advantages relative to materials formed solely from the corresponding neat polymer. In particular, the inventor has demonstrated that the composite materials of the invention offer an improvement in a variety of material properties, including, but not limited to shrinkage properties, tensile and flexural properties, moisture uptake and heat distortion properties. The composite materials also exhibit improved recyclability and fire retardancy relative to the corresponding neat polymer. The reduced amount of petroleum-derived material in the composite materials, relative to materials formed solely from the corresponding neat polymer, gives rise to a significantly reduced environmental impact.

[0012] Moreover, through rigorous investigations, the inventor has demonstrated that the properties of the composite material can be tailored according to the intended application of the material, meaning that the composite materials can be incorporated into a variety of components across a range of different industrial sectors.

Polymer

[0013] The composite material comprises 25 – 80 wt% of at least one polymer selected from the group consisting of polypropylene, polyethylene, polystyrene, polypropylene-co-ethylene, polypropylene-co-styrene, polyethylene-co-styrene and polypropylene-co-ethylene-co-styrene. The composite material is provided as a mixture of all of the components contained therein, meaning that the at least one polymer is dispersed throughout the composite material. The at least one polymer may be present as a polymeric matrix, throughout which the other components of the composite material are dispersed.

[0014] Where the at least one polymer is a copolymer, it will be understood that all known types of copolymer are encompassed, including, but not limited to, random, alternating, block, statistical and graft copolymers.

[0015] In an embodiment, the at least one polymer is selected from the group consisting of polypropylene, polypropylene-co-ethylene, polypropylene-co-styrene and polypropylene-co-ethylene-co-styrene. Suitably, propylene is the dominant repeating unit by mass in polypropylene-co-ethylene, polypropylene-co-styrene and polypropylene-co-ethylene-co-styrene.

[0016] In an embodiment, the at least one polymer is selected from the group consisting of polypropylene and polypropylene-co-ethylene. Suitably, propylene is the dominant repeating unit by mass in polypropylene-co-ethylene. Suitably, the at least one polymer selected from the

group consisting of polypropylene and polypropylene-co-ethylene has a melt flow index of 10 – 75 g/10 min at 230°C/2.16 kg.

[0017] In a particularly suitable embodiment, the at least one polymer is polypropylene. Suitably, the polypropylene has a melt flow index of 25 – 75 g/10 min at 230°C/2.16 kg.

[0018] In another particularly suitable embodiment, the at least one polymer is polypropylene-co-ethylene. Suitably, propylene is the dominant repeating unit by mass in polypropylene-co-ethylene. The polypropylene-co-ethylene copolymer is suitably a random copolymer. Suitably, the polypropylene-co-ethylene copolymer has a melt flow index of 10 – 50 g/10 min at 230°C/2.16 kg.

[0019] The at least one polymer, having any of the definitions outlined above, may be present in the composite material in various amounts.

[0020] In an embodiment, the material comprises 30 – 77.5 wt% of the at least one polymer. Suitably, the material comprises 40 – 77.5 wt% of the at least one polymer. More suitably, the material comprises 40 – 72.5 wt% of the at least one polymer. Even more suitably, the material comprises 40 – 67.5 wt% of the at least one polymer. Yet more suitably, the material comprises 45 – 67.5 wt% of the at least one polymer. Yet even more suitably, the material comprises 45 – 62.5 wt% of the at least one polymer. In a particularly suitable embodiment, the material comprises 45 – 57.5 wt% of the at least one polymer.

[0021] In another embodiment, the material comprises 50 – 80 wt% of the at least one polymer. Suitably, the material comprises 60 – 80 wt% of the at least one polymer. In a particularly suitable embodiment, the material comprises 70 – 78 wt% of the at least one polymer.

Lignocellulosic compound

[0022] The composite material comprises 15 – 70 wt% of at least one lignocellulosic compound, wherein the at least one lignocellulosic compound is a shive. The composite material is provided as a mixture of all of the components contained therein, meaning that the at least one lignocellulosic compound is dispersed throughout the composite material.

[0023] The term shive will be readily understood by one of ordinary skill in the art. In particular, the skilled person will appreciate that shives (also known as hurds) are woody particles derived from the inner stalk that are produced as a waste product during the extraction of valuable bast fibres from fibrous plants such as flax, hemp and kenaf. During breaking and scutching, the woody, inner stalk of the plant is broken into pieces and separated from short fibres and long fibres. Thus, shives are traditionally viewed as a waste product of fibre production.

[0024] The results provided herein, in particular in relation to the tensile properties of the composite materials, illustrate the unexpected advantages associated with using cheaper shive lignocellulosic compounds rather than more expensive bast fibres, such as kenaf fibre.

[0025] In an embodiment, the at least one lignocellulosic compound is selected from the group consisting of hemp shive and flax shive.

[0026] The at least one lignocellulosic compound may have an average particle size of 75 – 250 μm . When used herein in relation to the at least one lignocellulosic compound, the term average particle size means D_{50} , which can be determined, for example, by using an optical microscope and particle size analysis software such as ImageJ. Suitably, the at least one lignocellulosic compound has an average particle size of 100 – 225 μm . More suitably, the at least one lignocellulosic compound has an average particle size of 100 – 200 μm . In a particularly suitable embodiment, the at least one lignocellulosic compound has an average particle size of 125 – 175 μm .

[0027] The at least one lignocellulosic compound may have a particle density of 0.7 – 1.5 g cm^{-3} . Suitably, the at least one lignocellulosic compound has a particle density of 0.9 – 1.3 g cm^{-3} .

[0028] In an embodiment, the at least one lignocellulosic compound comprises 20 – 55 wt% of cellulose, 10 – 35 wt% of hemicellulose and 12 – 50 wt% of lignin. Suitably, the at least one lignocellulosic compound comprises 24 – 51 wt% of cellulose, 10 – 35 wt% of hemicellulose and 14 – 44 wt% of lignin.

[0029] In an embodiment, the at least one lignocellulosic compound comprises 35 – 50 wt% of cellulose, 10 – 35 wt% of hemicellulose and 15 – 30 wt% of lignin. Suitably, the at least one lignocellulosic compound is hemp shives.

[0030] In an embodiment, the at least one lignocellulosic compound comprises 25 – 39 wt% of cellulose, 21 – 32 wt% of hemicellulose and 40 – 43 wt% of lignin. Suitably, the at least one lignocellulosic compound is flax shives.

[0031] In a particularly suitable embodiment, the at least one lignocellulosic compound is hemp shive. Suitably, the hemp shive has an average particle size of 125 – 175 μm and/or a particle density of 0.9 – 1.3 g cm^{-3} .

[0032] In another particularly suitable embodiment, the at least one lignocellulosic compound is flax shive.

[0033] In another particularly suitable embodiment, the at least one lignocellulosic compound is a mixture of hemp shive and flax shive, each having any of the definitions outlined above.

[0034] The at least one lignocellulosic compound, having any of the definitions outlined above, may be present in the composite material in various amounts.

[0035] In an embodiment, the material comprises 17.5 – 65 wt% of the at least one lignocellulosic compound. Suitably, the material comprises 17.5 – 55 wt% of the at least one lignocellulosic compound. More suitably, the material comprises 22.5 – 55 wt% of the at least one lignocellulosic compound. Even more suitably, the material comprises 27.5 – 55 wt% of the at least one lignocellulosic compound. Yet more suitably, the material comprises 27.5 – 50 wt% of the at least one lignocellulosic compound. Yet even more suitably, the material comprises 32.5 – 50 wt% of the at least one lignocellulosic compound. In a particularly suitable embodiment, the material comprises 37.5 – 50 wt% of the at least one lignocellulosic compound.

[0036] In another embodiment, the material comprises 15 – 45 wt% of the at least one lignocellulosic compound. Suitably, the material comprises 15 – 35 wt% of the at least one lignocellulosic compound. In a particularly suitable embodiment, the material comprises 17 – 25 wt% of the at least one lignocellulosic compound.

Graphene

[0037] The composite material comprises 0.01 – 5.5 wt% of graphene. The composite material is provided as a mixture of all of the components contained therein, meaning that the graphene is dispersed throughout the composite material.

[0038] Graphene is the name given to a particular crystalline allotrope of carbon in which each carbon atom is bound to three adjacent carbon atoms (in a sp^2 hybridised manner) so as to define a one atom thick planar sheet of carbon. The carbon atoms in graphene are arranged in the planar sheet in a honeycomb-like network of tessellated hexagons. Graphene can be formed by exfoliation of graphite.

[0039] The term graphene used herein does not encompass graphene oxide or reduced graphene oxide.

[0040] The graphene may comprise 5 – 50 layers. Suitably, the graphene comprises 12 – 30 layers.

[0041] The graphene may have an average thickness of 0.9 – 20 nm. Suitably, the graphene has an average thickness of 2 – 15 nm. More suitably, the graphene has an average thickness of 4 – 10 nm.

[0042] The graphene may have an average particle size of 1 – 35 μm . When used herein in relation to graphene, the term average particle refers to the average lateral dimension of the graphene flakes, which can be determined, for example, by scanning electron microscopy or atomic force microscopy. Suitably, the graphene has an average particle size of 2.5 – 30 μm .

[0043] In one particular embodiment, the graphene has an average particle size of 1 – 10 μm . Suitably, the graphene has an average particle size of 2.5 – 7.5 μm . Graphene of this average particle size may be particularly useful where it is desirable to reduce shrinkage as much as possible during compounding and extrusion. In addition, graphene of this average particle size may be particularly useful where it is desirable to increase tensile and flexural properties of the composite as much as possible, and/or where it is necessary for the composite to have a low heat distortion temperature. Graphene of this average particle size may be referred to herein as “G-S”.

[0044] In another particular embodiment, the graphene has an average particle size of 10 – 20 μm . Suitably, the graphene has an average particle size of 12.5 – 17.5 μm . Graphene of this average particle size may be particularly useful where it is desirable to increase tensile and flexural properties of the composite. Graphene of this average particle size may be referred to herein as “G-M”.

[0045] In another particular embodiment, the graphene has an average particle size of 20 – 30 μm . Suitably, the graphene has an average particle size of 22.5 – 27.5 μm . Graphene of this average particle size may be particularly useful where it is desirable to increase tensile and flexural properties of the composite. Graphene of this average particle size may be referred to herein as “G-L”.

[0046] The composite may comprise a mixture of two or more particle size grades of graphene selected from the aforementioned “G-S”, “G-M” and “G-L” particle size grades.

[0047] The graphene, having any of the definitions outlined above, may be present in the composite material in various amounts.

[0048] In an embodiment, the material comprises 0.05 – 4.0 wt% of graphene. Suitably, the material comprises 0.1 – 3.0 wt% of graphene. More suitably, the material comprises 0.5 – 2 wt% of graphene. In a particular embodiment, the material comprises 0.65 – 1.5 wt% of graphene.

[0049] In another embodiment, the material comprises 0.05 – 1.5 wt% of graphene.

Coupling agent

[0050] The composite material comprises 2.0 – 11.0 wt% of at least one coupling agent. The composite material is provided as a mixture of all of the components contained therein, meaning that the at least one coupling agent is dispersed throughout the composite material.

[0051] As will be familiar to one of ordinary skill in the art, the at least one coupling agent improves the interface between the hydrophobic polymer and the hydrophilic lignocellulosic compound. In this sense, the at least one coupling agent is any agent that is able to reduce the interfacial tension between the at least one polymer and the at least one lignocellulosic compound. The at least one coupling agent may also help to increase mechanical entanglement within the composite.

[0052] The at least one coupling agent may be an amphiphilic compound. For example, the at least one coupling agent may comprise a hydrophobic portion miscible with the at least one polymer and a hydrophilic portion miscible with the at least one lignocellulosic compound.

[0053] In an embodiment, the at least one coupling agent is selected from the group consisting of an amphiphilic copolymer, a (5-35C)fatty acid, a (5-35C) fatty acid anhydride, a (5-35C)fatty acid ester and an organosilane.

[0054] In an embodiment, the at least one coupling agent is an amphiphilic graft copolymer. Amphiphilic graft copolymers (sometimes termed amphiphilic comb copolymers) will be familiar to one of ordinary skill in the art as comprising a backbone portion with a multiplicity of pendant group grafted thereto, wherein the backbone portion and pendant groups have opposing hydrophobicity (i.e. a hydrophobic backbone and hydrophilic pendant groups, or *vice versa*).

[0055] In an embodiment, the at least one coupling agent is an amphiphilic graft copolymer comprising a hydrophobic backbone having pendant hydrophilic groups.

[0056] Where the at least one coupling agent is an amphiphilic graft copolymer comprising a hydrophobic backbone having pendant hydrophilic groups, the hydrophobic backbone is suitably a polyolefin. More suitably, the hydrophobic backbone is selected from the group consisting of polypropylene, polyethylene, polystyrene, polypropylene-co-ethylene, polypropylene-co-styrene, polyethylene-co-styrene and polypropylene-co-ethylene-co-styrene. Even more suitably, the hydrophobic backbone is the same as the least one polymer used in the composite material. In a particular embodiment, the hydrophobic backbone is polypropylene.

[0057] Where the at least one coupling agent is an amphiphilic graft copolymer comprising a hydrophobic backbone having pendant hydrophilic groups, the pendant hydrophilic groups are suitably maleic anhydride. It will be understood that some or all of the maleic anhydride groups

may be present as maleic acid ester groups and/or maleic acid groups within the composite material due to interaction with the at least one lignocellulosic compound.

[0058] In a particularly suitable embodiment, the at least one coupling agent is polypropylene-graft-maleic anhydride comprising 0.5 – 4 wt% of maleic anhydride. Suitably, the at least one coupling agent is polypropylene-graft-maleic anhydride comprising 0.75 – 2.5 wt% of maleic anhydride. Most suitably, the at least one coupling agent is polypropylene-graft-maleic anhydride comprising 1.5 – 2.25 wt% of maleic anhydride. It will be understood that some or all of the maleic anhydride groups may be present as maleic acid groups within the composite material due to interaction with the at least one lignocellulosic compound.

[0059] The polypropylene-graft-maleic anhydride coupling agent outlined above may have a melt volume rate of 30 – 110 cm³ / 10 minutes at 170°C with a load of 1.2 kg.

[0060] The polypropylene-graft-maleic anhydride coupling agent outlined above may have a melt flow index of 10 – 1500 g / 10 minutes at 170°C with a load of 1.2 kg, or 10 – 2000 g / 10 minutes at 190°C with a load of 2.16 kg. Suitably, the polypropylene-graft-maleic anhydride coupling agent has a melt flow index of 20 – 1000 g / 10 minutes at 170°C with a load of 1.2 kg, or 50 – 200 g / 10 minutes at 190°C with a load of 2.16 kg.

[0061] The at least one coupling agent, having any of the definitions outlined above, may be present in the composite material in various amounts.

[0062] In an embodiment, the material comprises 2.5 – 9.5 wt% of the at least one coupling agent. Suitably, the material comprises 2.75 – 8.5 wt% of the at least one coupling agent. More suitably, the material comprises 3.5 – 6.5 wt% of the at least one coupling agent.

[0063] In another embodiment, the material comprises 2.0 – 4.5 wt% of the at least one coupling agent. Suitably, the material comprises 2.5 – 4.0 wt% of the at least one coupling agent.

Preparation of composite material

[0064] In a second aspect, the present invention provides a process for the preparation of a composite material according to the first aspect, the process comprising the steps of:

- A) mixing the at least one polymer, the at least one lignocellulosic compound, the at least one coupling agent and graphene, and
- B) extruding the mixture resulting from step A).

[0065] The composite materials of the invention can be straightforwardly prepared using conventional extrusion apparatus. For example, the composite materials can be prepared using a single screw extrusion apparatus or a twin screw extrusion apparatus.

[0066] It will be understood that the at least one polymer, the at least one lignocellulosic compound, the at least one coupling agent and graphene described in relation to the second aspect of the invention may have any one of those definitions outlined hereinbefore in relation to the first aspect of the invention. In particular, the quantities of the at least one polymer, the at least one lignocellulosic compound, the at least one coupling agent and graphene described hereinbefore in relation to the first aspect of the invention are equally applicable to the second aspect of the invention, due to the fact that substantially no losses occur during the preparation process.

[0067] The at least one polymer, the at least one lignocellulosic compound, the at least one coupling agent and graphene are mixed in step A) at a temperature sufficient to form an intimate mixture. In an embodiment, the at least one polymer is molten during step A).

[0068] The order in which each of the at least one polymer, the at least one lignocellulosic compound, the at least one coupling agent and graphene are fed to the extrusion apparatus and mixed with the other components is not critical. Suitably, the at least one polymer, the at least one lignocellulosic compound, the at least one coupling agent and graphene are fed to the extrusion apparatus and mixed with the other components in such a manner that the graphene is not exposed to a temperature greater than 250°C and the at least one lignocellulosic compound is not exposed to a temperature greater than 185°C.

[0069] In an embodiment, step A) comprises the sub steps:

- A1) mixing the at least one polymer, the at least one coupling agent and graphene, and
- A2) mixing the mixture resulting from step A1) with the at least one lignocellulosic compound.

[0070] Step A1) can be conducted in two different ways.

[0071] In a first way of conducting step A1), all of the at least one polymer is added and mixed with the at least one coupling agent and graphene in a single step. Thus, after addition of the at least one lignocellulosic compound in step A2), all components are present in the quantities required to achieve a composite material having a target composition.

[0072] In a second way of conducting step A1), not all of the at least one polymer is added and mixed with the at least one coupling agent and graphene in an initial step in order to form a masterbatch. Then, in a subsequent step, the masterbatch is mixed with the remaining quantity of the at least one polymer, such that, after addition of the at least one lignocellulosic

compound in step A2), all components are present in the quantities required to achieve a composite material having a target composition.

[0073] Step A1) may be conducted at a temperature of 150 – 250°C.

[0074] In step A1), the screw(s) of the extrusion apparatus may be rotated at a speed of 175 – 650 rpm. Suitably, the screw(s) of the extrusion apparatus may be rotated at a speed of 175 – 425 rpm. The screw speed may depend on the size of the extrusion apparatus.

[0075] Step A2) may be conducted at a temperature of 150 – 185°C.

[0076] In step A2), the screw(s) of the extrusion apparatus may be rotated at a speed of 75 – 250 rpm. Suitably, the screw(s) of the extrusion apparatus may be rotated at a speed of 75 – 225 rpm. More suitably, the screw(s) of the extrusion apparatus may be rotated at a speed of 75 – 150 rpm. The screw speed may depend on the size of the extrusion apparatus.

[0077] In addition to steps A) and B), the process for preparing the composite material may additionally comprise a step C) of drying the extrudate resulting from step B).

[0078] In a third aspect, the present invention provides a composite material obtained, directly obtained or obtainable by a process according to the second aspect of the invention.

Applications of the composite material

[0079] In a fourth aspect, the present invention provides a use of a composite material according to the first or third aspect in a component for an automobile, in a container, in a sports article, in an appliance, in an electronic device, or in a consumer good.

[0080] The properties of the composite material make the latter particularly well suited for use in a variety of internal and external components within cars, bicycles, motorcycles and aircraft. Also, the properties of the composite material make the latter particularly well suited for use in a variety of containers, such as bottles, trays, tubs and boxes.

[0081] The following numbered statements 1 to 117 are not claims, but instead serve to define particular aspects and embodiments of the claimed invention:

1. A composite material comprising:

25 – 80 wt% of at least one polymer selected from the group consisting of polypropylene, polyethylene, polystyrene, polypropylene-co-ethylene, polypropylene-co-styrene, polyethylene-co-styrene and polypropylene-co-ethylene-co-styrene;

15 – 70 wt% of at least one lignocellulosic compound, wherein the at least one lignocellulosic compound is a shive;

2.0 – 11.0 wt% of at least one coupling agent; and

0.01 – 5.5 wt% of graphene.

2. The composite material of statement 1, wherein the material comprises 17.5 – 65 wt% of the at least one lignocellulosic compound.
3. The composite material of statement 1, wherein the material comprises 17.5 – 55 wt% of the at least one lignocellulosic compound.
4. The composite material of statement 1, wherein the material comprises 22.5 – 55 wt% of the at least one lignocellulosic compound.
5. The composite material of statement 1, wherein the material comprises 27.5 – 55 wt% of the at least one lignocellulosic compound.
6. The composite material of statement 1, wherein the material comprises 27.5 – 50 wt% of the at least one lignocellulosic compound.
7. The composite material of statement 1, wherein the material comprises 32.5 – 50 wt% of the at least one lignocellulosic compound.
8. The composite material of statement 1, wherein the material comprises 37.5 – 50 wt% of the at least one lignocellulosic compound.
9. The composite material of statement 1, wherein the material comprises 15 – 45 wt% of the at least one lignocellulosic compound.
10. The composite material of statement 1, wherein the material comprises 15 – 35 wt% of the at least one lignocellulosic compound.
11. The composite material of statement 1, wherein the material comprises 17 – 25 wt% of the at least one lignocellulosic compound.
12. The composite material of any preceding statement, wherein the material comprises 30 – 77.5 wt% of the at least one polymer.
13. The composite material of statement 12, wherein the material comprises 40 – 77.5 wt% of the at least one polymer.
14. The composite material of statement 12, wherein the material comprises 40 – 72.5 wt% of the at least one polymer.
15. The composite material of statement 12, wherein the material comprises 40 – 67.5 wt% of the at least one polymer.
16. The composite material of statement 12, wherein the material comprises 45 – 67.5 wt% of the at least one polymer.
17. The composite material of statement 12, wherein the material comprises 45 – 62.5 wt% of the at least one polymer.

18. The composite material of statement 12, wherein the material comprises 45 – 57.5 wt% of the at least one polymer.
19. The composite material of any one of statements 1 to 11, wherein the material comprises 50 – 80 wt% of the at least one polymer.
20. The composite material of statement 19, wherein the material comprises 60 – 80 wt% of the at least one polymer.
21. The composite material of statement 19, wherein the material comprises 70 – 78 wt% of the at least one polymer.
22. The composite material of statement 1, wherein the material comprises:
30 – 77.5 wt% of the at least one polymer; and
17.5 – 65 wt% of the at least one lignocellulosic compound.
23. The composite material of statement 1, wherein the material comprises:
40 – 77.5 wt% of the at least one polymer; and
17.5 – 55 wt% of the at least one lignocellulosic compound.
24. The composite material of statement 1, wherein the material comprises:
40 – 72.5 wt% of the at least one polymer; and
22.5 – 55 wt% of the at least one lignocellulosic compound.
25. The composite material of statement 1, wherein the material comprises:
40 – 67.5 wt% of the at least one polymer; and
27.5 – 55 wt% of the at least one lignocellulosic compound.
26. The composite material of statement 1, wherein the material comprises:
45 – 67.5 wt% of the at least one polymer; and
27.5 – 50 wt% of the at least one lignocellulosic compound.
27. The composite material of statement 1, wherein the material comprises:
45 – 62.5 wt% of the at least one polymer; and
32.5 – 50 wt% of the at least one lignocellulosic compound.
28. The composite material of statement 1, wherein the material comprises:
45 – 57.5 wt% of the at least one polymer; and
37.5 – 50 wt% of the at least one lignocellulosic compound.
29. The composite material of statement 1, wherein the material comprises:
50 – 80 wt% of the at least one polymer; and
15 – 45 wt% of the at least one lignocellulosic compound.
30. The composite material of statement 1, wherein the material comprises:
60 – 80 wt% of the at least one polymer; and
15 – 35 wt% of the at least one lignocellulosic compound.
31. The composite material of statement 1, wherein the material comprises:
70 – 78 wt% of the at least one polymer; and

- 17 – 25 wt% of the at least one lignocellulosic compound.
32. The composite material of any preceding statement, wherein the at least one lignocellulosic compound is selected from the group consisting of hemp shive and flax shive.
33. The composite material of any preceding statement, wherein the at least one lignocellulosic compound comprises 20 – 55 wt% of cellulose, 10 – 35 wt% of hemicellulose and 12 – 50 wt% of lignin.
34. The composite material of any preceding statement, wherein the at least one lignocellulosic compound comprises 24 – 51 wt% of cellulose, 10 – 35 wt% of hemicellulose and 14 – 44 wt% of lignin.
35. The composite material of any one of claims 1 to 32, wherein the at least one lignocellulosic compound comprises 35 – 50 wt% of cellulose, 10 – 35 wt% of hemicellulose and 15 – 30 wt% of lignin.
36. The composite material of any one of claims 1 to 32, wherein the at least one lignocellulosic compound comprises 25 – 39 wt% of cellulose, 21 – 32 wt% of hemicellulose and 40 – 43 wt% of lignin.
37. The composite material of any preceding statement, wherein the at least one lignocellulosic compound has an average particle size of 75 – 250 μm .
38. The composite material of any preceding statement, wherein the at least one lignocellulosic compound has an average particle size of 75 – 225 μm .
39. The composite material of any preceding statement, wherein the at least one lignocellulosic compound has an average particle size of 100 – 200 μm .
40. The composite material of any preceding statement, wherein the at least one lignocellulosic compound has an average particle size of 125 – 175 μm .
41. The composite material of any preceding statement, wherein the at least one lignocellulosic compound has a particle density of 0.7 – 1.5 g cm^{-3} .
42. The composite material of any preceding statement, wherein the at least one lignocellulosic compound has a particle density of 0.9 – 1.3 g cm^{-3} .
43. The composite material of any one of statements 1 to 31, wherein
the at least one lignocellulosic compound is selected from the group consisting of hemp shive and flax shive; and
the at least one lignocellulosic compound comprises 20 – 55 wt% of cellulose, 10 – 35 wt% of hemicellulose and 12 – 50 wt% of lignin.
44. The composite material of any one of statements 1 to 31, wherein
the at least one lignocellulosic compound is selected from the group consisting of hemp shive and flax shive; and

- the at least one lignocellulosic compound comprises 20 – 55 wt% of cellulose, 10 – 35 wt% of hemicellulose and 12 – 50 wt% of lignin; and
- the at least one lignocellulosic compound has an average particle size of 75 – 250 μm .
45. The composite material of any one of statements 1 to 31, wherein
- the at least one lignocellulosic compound is selected from the group consisting of hemp shive and flax shive; and
- the at least one lignocellulosic compound comprises 20 – 55 wt% of cellulose, 10 – 35 wt% of hemicellulose and 12 – 50 wt% of lignin; and
- the at least one lignocellulosic compound has an average particle size of 75 – 250 μm ; and
- the at least one lignocellulosic compound has a particle density of 0.7 – 1.5 g cm^{-3} .
46. The composite material of any preceding statement, wherein the at least one polymer is selected from the group consisting of polypropylene, polypropylene-co-ethylene, polypropylene-co-styrene and polypropylene-co-ethylene-co-styrene.
47. The composite material of statement 46, wherein propylene is the dominant repeating unit by mass in polypropylene-co-ethylene, polypropylene-co-styrene and polypropylene-co-ethylene-co-styrene.
48. The composite material of any preceding statement, wherein the at least one polymer is polypropylene or polypropylene-co-ethylene.
49. The composite material of statement 48, wherein the at least one polymer has a melt flow index of 10 – 75 g/10 min at 230°C/2.16 kg.
50. The composite material of any preceding statement, wherein the at least one polymer is polypropylene
51. The composite material of statement 50, wherein the polypropylene has a melt flow index of 25 – 75 g/10 min at 230°C/2.16 kg.
52. The composite material of any one of statements 1 to 51, wherein the at least one polymer is polypropylene-co-ethylene.
53. The composite material of statement 52, wherein the polypropylene-co-ethylene has a melt flow index of 10 – 50 g/10 min at 230°C/2.16 kg.
54. The composite material of any one of statements 1 to 45, wherein
- the at least one polymer is polypropylene having a melt flow index of 25 – 75 g/10 min at 230°C/2.16 kg; or
- the at least one polymer is polypropylene-co-ethylene having a melt flow index of 10 – 50 g/10 min at 230°C/2.16 kg
55. The composite material of any preceding statement, wherein the material comprises 0.05 – 4.0 wt% of graphene.

56. The composite material of any preceding statement, wherein the material comprises 0.1 – 3.0 wt% of graphene.
57. The composite material of any preceding statement, wherein the material comprises 0.5 – 2 wt% of graphene.
58. The composite material of any preceding statement, wherein the material comprises 0.65 – 1.5 wt% of graphene.
59. The composite material of any one of statements 1 to 55, wherein the material comprises 0.05 – 1.5 wt% of graphene.
60. The composite material of any preceding statement, wherein the graphene comprises 5 – 50 layers.
61. The composite material of any preceding statement, wherein the graphene has an average thickness of 0.9 – 20 nm.
62. The composite material of any preceding statement, wherein the graphene has an average thickness of 2 – 15 nm.
63. The composite material of any preceding statement, wherein the graphene has an average thickness of 4 – 10 nm.
64. The composite material of any preceding statement, wherein the graphene has an average particle size of 1 – 35 μm .
65. The composite material of any preceding statement, wherein the graphene has an average particle size of 1 – 10 μm .
66. The composite material of any preceding statement, wherein the graphene has an average particle size of 2.5 – 7.5 μm .
67. The composite material of any one of statements 1 to 64, wherein the graphene has an average particle size of 10 – 20 μm .
68. The composite material of any one of statements 1 to 64, wherein the graphene has an average particle size of 12.5 – 17.5 μm .
69. The composite material of any one of statements 1 to 64, wherein the graphene has an average particle size of 20 – 30 μm .
70. The composite material of any one of statements 1 to 64, wherein the graphene has an average particle size of 22.5 – 27.5 μm .
71. The composite material of any one of statements 1 to 64, wherein the graphene has an average thickness of 4 – 10 nm; and the graphene has an average particle size of 1 – 35 μm .
72. The composite material of any one of statements 1 to 64, wherein the graphene has an average thickness of 4 – 10 nm; and the graphene has an average particle size of 2.5 – 7.5 μm .
73. The composite material of any one of statements 1 to 64, wherein

- the graphene has an average thickness of 4 – 10 nm; and
the graphene has an average particle size of 12.5 – 17.5 μm .
74. The composite material of any one of statements 1 to 64, wherein
the graphene has an average thickness of 4 – 10 nm; and
the graphene has an average particle size of 22.5 – 27.5 μm .
75. The composite material of any preceding statement, wherein the material comprises 2.5 – 9.5 wt% of the at least one coupling agent.
76. The composite material of any preceding statement, wherein the material comprises 2.75 – 8.5 wt% of the at least one coupling agent.
77. The composite material of any preceding statement, wherein the material comprises 3.5 – 6.5 wt% of the at least one coupling agent
78. The composite material of any one of statements 1 to 74, wherein the material comprises 2.0 – 4.5 wt% of the at least one coupling agent.
79. The composite material of any one of statements 1 to 74, wherein the material comprises 2.5 – 4.0 wt% of the at least one coupling agent.
80. The composite material of any preceding statement, wherein the at least one coupling agent is an amphiphilic compound.
81. The composite material of any preceding statement, wherein the at least one coupling agent is selected from the group consisting of an amphiphilic copolymer, a (5-35C)fatty acid, a (5-35C) fatty acid anhydride, a (5-35C)fatty acid ester and an organosilane.
82. The composite material of statement 80 or 81, wherein the at least one coupling agent is an amphiphilic graft copolymer.
83. The composite material of statement 81 or 82, wherein the amphiphilic copolymer comprises a hydrophobic backbone having pendant hydrophilic groups.
84. The composite material of statement 81 or 82 wherein the amphiphilic copolymer comprises a polyolefinic backbone having pendant hydrophilic groups.
85. The composite material of statement 84, wherein the polyolefinic backbone is selected from the group consisting of polypropylene, polyethylene, polystyrene, polypropylene-co-ethylene, polypropylene-co-styrene, polyethylene-co-styrene and polypropylene-co-ethylene-co-styrene.
86. The composite material of statement 84 or 85, wherein the polyolefinic backbone is the same as the at least one polymer.
87. The composite material of any one of statements 84, 85 and 86, wherein the polyolefinic backbone is polypropylene.
88. The composite material of any one of statements 83 to 86, wherein the pendant hydrophilic groups are maleic anhydride.

89. The composite material of any preceding statement, wherein the at least one coupling agent is polypropylene-graft-maleic anhydride comprising 0.5 – 4 wt% of maleic anhydride.
90. The composite material of any preceding statement, wherein the at least one coupling agent is polypropylene-graft-maleic anhydride comprising 0.75 – 2.5 wt% of maleic anhydride.
91. The composite material of any preceding statement, wherein the at least one coupling agent is polypropylene-graft-maleic anhydride comprising 1.5 – 2.25 wt% of maleic anhydride.
92. The composite material of any one of statements 89, 90 and 91, wherein the polypropylene-graft-maleic anhydride has a melt volume rate of $30 - 110 \text{ cm}^3 / 10$ minutes at 170°C with a load of 1.2 kg.
93. The composite material of any one of statements 89 to 92, wherein the polypropylene-graft-maleic anhydride has a melt flow index of $10 - 1500 \text{ g} / 10$ minutes at 170°C with a load of 1.2 kg, or $10 - 2000 \text{ g} / 10$ minutes at 190°C with a load of 2.16 kg.
94. The composite material of statement 93, wherein the polypropylene-graft-maleic anhydride has a melt flow index of $20 - 1000 \text{ g} / 10$ minutes at 170°C with a load of 1.2 kg, or $50 - 200 \text{ g} / 10$ minutes at 190°C with a load of 2.16 kg.
95. The composite material of any one of statements 1 to 74, wherein the at least one coupling agent is an amphiphilic graft copolymer; and the amphiphilic copolymer comprises a hydrophobic polyolefinic backbone having pendant hydrophilic groups; and the polyolefinic backbone is the same as the at least one polymer.
96. The composite material of any one of statements 1 to 74, wherein the at least one coupling agent is an amphiphilic graft copolymer; and the amphiphilic copolymer comprises a polypropylene backbone having pendant maleic anhydride groups.
97. The composite material of statement 1, wherein the material comprises:
40 – 77.5 wt% of the at least one polymer;
17.5 – 55 wt% of the at least one lignocellulosic compound;
2.5 – 9.5 wt% of the at least one coupling agent; and
0.05 – 4.0 wt% of graphene;
and wherein
the at least one polymer is as defined in any one of statements 46 to 54, preferably statement 54;
the at least one lignocellulosic compound is as defined in any one of statements 32 to 45, preferably 43 to 45;

the at least one coupling agent is as defined in any one of statements 78 to 94, preferably 93 or 94; and

the graphene is as defined in any one of statements 60 to 74, preferably 71 to 74.

98. The composite material of statement 1, wherein the material comprises:

40 – 67.5 wt% of the at least one polymer;

27.5 – 55 wt% of the at least one lignocellulosic compound;

2.75 – 8.5 wt% of the at least one coupling agent; and

0.1 – 3.0 wt% of graphene;

and wherein

the at least one polymer is as defined in any one of statements 46 to 54, preferably statement 54;

the at least one lignocellulosic compound is as defined in any one of statements 32 to 45, preferably 43 to 45;

the at least one coupling agent is as defined in any one of statements 80 to 96, preferably 95 or 96; and

the graphene is as defined in any one of statements 60 to 74, preferably 71 to 74.

99. The composite material of statement 1, wherein the material comprises:

45 – 62.5 wt% of the at least one polymer; and

32.5 – 50 wt% of the at least one lignocellulosic compound;

3.5 – 6.5 wt% of the at least one coupling agent; and

0.5 – 2 wt% of graphene;

and wherein

the at least one polymer is as defined in any one of statements 46 to 54, preferably statement 54;

the at least one lignocellulosic compound is as defined in any one of statements 32 to 45, preferably 43 to 45;

the at least one coupling agent is as defined in any one of statements 80 to 96, preferably 95 or 96; and

the graphene is as defined in any one of statements 60 to 74, preferably 71 to 74.

100. The composite material of statement 1, wherein the material comprises:

50 – 80 wt% of the at least one polymer;

15 – 45 wt% of the at least one lignocellulosic compound;

2.0 – 4.5 wt% of the at least one coupling agent; and

0.05 – 1.5 wt% of graphene;

and wherein

the at least one polymer is as defined in any one of statements 46 to 54, preferably statement 54;

- the at least one lignocellulosic compound is as defined in any one of statements 32 to 45, preferably 43 to 45;
- the at least one coupling agent is as defined in any one of statements 80 to 96, preferably 95 or 96; and
- the graphene is as defined in any one of statements 60 to 74, preferably 71 to 74.
101. The composite material of statement 1, wherein the material comprises:
- 70 – 78 wt% of the at least one polymer;
 - 17 – 25 wt% of the at least one lignocellulosic compound;
 - 2.5 – 4.0 wt% of the at least one coupling agent; and
 - 0.05 – 1.5 wt% of graphene;
- and wherein
- the at least one polymer is as defined in any one of statements 46 to 54, preferably statement 54;
- the at least one lignocellulosic compound is as defined in any one of statements 32 to 45, preferably 43 to 46;
- the at least one coupling agent is as defined in any one of statements 80 to 96, preferably 95 or 96; and
- the graphene is as defined in any one of statements 60 to 74, preferably 71 to 74.
102. A process for the preparation of a composite material as defined in any preceding statement, the process comprising the steps of:
- A) mixing the at least one polymer, the at least one lignocellulosic compound, the at least one coupling agent and graphene, and
 - B) extruding the mixture resulting from step A).
103. The process of statement 102, wherein the at least one polymer, and optionally also the at least one coupling agent are molten during step A).
104. The process of statement 102 or 103, wherein step A) comprises the sub steps:
- A1) mixing the at least one polymer, the at least one coupling agent and graphene, and
 - A2) mixing the mixture resulting from step A1) with the at least one lignocellulosic compound.
105. The process of statement 104, wherein step A1) comprises mixing the at least one polymer, the at least one coupling agent and graphene to produce a masterbatch, and then mixing the masterbatch with an additional quantity of the at least one polymer
106. The process of statement 104 or 105, wherein step A1) is conducted at a temperature of 150 – 250°C.
107. The process of statement 104, 105 or 106, wherein the mixing in step A1) is conducted at a speed of 175 – 650 rpm.

108. The process of statement 104, 105 or 106, wherein the mixing in step A1) is conducted at a speed of 175 – 425 rpm.
109. The process of any one of statements 104 to 108, wherein step A2) is conducted at a temperature of 150 – 185°C.
110. The process of any one of statements 104 to 109, wherein the mixing in step A2) is conducted at a speed of 75 – 250 rpm.
111. The process of any one of statements 104 to 109, wherein the mixing in step A2) is conducted at a speed of 75 – 225 rpm.
112. The process of any one of statements 104 to 109, wherein the mixing in step A2) is conducted at a speed of 75 – 125 rpm.
113. The process of any one of statements 102 to 112, further comprising the step C) of drying the extrudate resulting from step B).
114. A composite material obtained, directly obtained or obtainable by a process according to any one of statements 102 to 113.
115. Use of a composite material as defined in any one of statements 1 to 101 and 114 in a component for an automobile, in a container, in a sports article, in an appliance, in an electronic device, or in a consumer good.
116. Use of a composite material of statement 115, wherein the composite material is used in a component for an automobile.
117. Use of a composite material of statement 115, wherein the composite material is used in a container.

EXAMPLES

[0082] One or more examples of the invention will now be described, for the purpose of illustration only, with reference to the accompanying figures, in which:

Fig. 1 shows a comparison of the moisture absorption properties of various composite materials of Table 1 after immersion in distilled water at 22°C for 24 hours.

Fig. 2 shows a comparison of the moisture absorption properties of various composite materials of Table 1 after immersion in distilled water at 22°C as a function of time.

Materials

[0083] Polypropylene homopolymer (PP-H, Ineos 100-CA50) is a high flow rate homopolymer (MFR 50 g/10min @ 230 °C/2.16 kg) designed for high speed injection moulding.

[0084] Polypropylene copolymer (PP-C, Ineos 200-CA25) is a clarified random copolymer with a medium ethylene content and good flow (MFR 25 g/10min @ 230 °C/2.16 kg) primarily intended for injection moulding.

[0085] Biomass from hemp (BM-H, Sunstrand HH999) is fine grinded hemp shive with the particle density of 1.178 g/cm³ (bulk density 0.110 g/cm³). The average particle size is around 150 µm (min. 25 µm, max 250 µm).

[0086] Maleic anhydride grafted polypropylene (MAPP, Scona TSPP 10213 GB) is a coupling agent. The maleic anhydride (MAH) content is 2.0 wt.%.

[0087] Graphene nanoplatelets and nanographite (GNPs) are Grade M with an average thickness of approximately 6-8 nm (17~24 layers) from XG Sciences. Three grades were investigated, which cover particle size ranges from small to large. G-S is the Grade M5 with an average particle diameter of 5 µm; G-M is the Grade M15 with an average particle diameter of 15 µm; G-L is the Grade M25 with an average particle diameter of 25 µm.

Example 1 – Preparation of composite materials

[0088] All the materials except GNPs were pre-dried before compounding. Compounding of the composite materials was carried out in a Collin ZK25 co-rotating twin-screw extruder (8 heating zones and the side feeding system is located at zone 3). Owing to their different physical and chemical properties, the screw configuration and temperature requirements for achieving good dispersion of the GNPs and BM in the polymer matrix are different. The ideal compounding conditions for these two components are as follows:

- GNPs – The screw configuration should provide high shear stress and high shear rate to disperse of GNPs and reduce their agglomeration. Ideally, the processing temperature for GNPs should not exceed 240°C, which in any event is significantly higher than the standard processing temperature for PP-H and PP-C.
- BM – The screw configuration should provide mild shear stress and mild shear rate to achieve a homogenous dispersion of BM. As BM requires low processing temperature, the temperature profile is ideally maintained below 185 °C after BM is fed through the side feeder.

[0089] The composite materials were prepared by 3-step process or a 2-step process, details of which are as follows:

3-step process

[0090] The 3-step process for preparing the composite materials is conducted as follows:

- Step 1 – Preparation of masterbatch

PP and MAPP are fed through the main hopper at zone 0 and GNPs are fed through the side feeder at zone 3. The loadings of MAPP were determined by the BM loadings in the final composite (every addition 10 wt.% BM requires 1.5 wt.% MAPP, which means 0.03 wt.% MAH). The screw configuration provides sufficient intensive shear stresses at a rotation speed between 200 rpm and 380 rpm. The temperature profile is between 170 °C and 230 °C. Masterbatches containing 2 wt.% and 5 wt.% GNPs were produced.

- Step 2 – Dilution

The GNPs/PP/MAPP masterbatch from Step 1 is mixed with neat PP to the desired GNPs loading levels (0.1, 0.77, 1, 3 and 5 wt.% in the final composite). The screw configuration, screw speed and temperature profiles are the same as in Step 1.

- Step 3 – Final compounding

The diluted GNPs/PP/MAPP samples from Step 2 were fed through the main hopper and BM is fed through the side feeder. The screw configuration provides mild shear stresses at a rotation speed of 100 rpm. The temperature profile is between 170 °C and 185 °C, except in zones 0, 1 and 2.

2-step process

[0091] To optimise the processing procedures and evaluate the effects of dilution step, 2-step compounding was also carried out using the same extruder as follows:

- Step 1 – Preparation of GNPs/PP/MAPP samples

PP and MAPP are fed through the main hopper and GNPs are fed through the side feeder. The loadings of GNPs and MAPP are calculated based on the loadings in the final composites. The processing conditions are the same as in Step 1 of the 3-step process.

- Step 2 – Final compounding

The GNPs/PP/MAPP samples from Step 1 were fed through the main hopper and BM is fed through the side feeder. The screw configuration provides mild shear stresses at a rotation speed of 100 rpm. The temperature profile is between 170 °C and 185 °C, except in zones 0, 1 and 2.

[0092] Irrespective of whether they are prepared by the 3-step or 2-step process, the resulting composite materials were dried before any further processing (injection or/and compression) was performed. All the test specimens (dumbbell-shaped tensile specimens and rectangle shaped flexural specimens), with a thickness of 4 mm were manufactured by industrial scale injection mould.

[0093] Table 1 below shows the composition of various composite materials.

Table 1: Composition and processing conditions of various composite materials

Sample	PP (wt%)		GNPs (wt%)			BM (wt%)	MAPP (wt%)	Compounding		Test Bars	
	PP-H	PP-C	G-S	G-M	G-L			3-Step	2-Step	Compression	Injection
A-0	100	-	-	-	-	-	-	X		X	X
A-1	87.9	-	-	1.1	-	10	1	X		X	
A-2	81.95	-	-	1.05	-	15	2	X		X	
A-3	77	-	-	-	-	20	3	X		X	X
A-4	75	-	-	-	-	20	5	X		X	
A-5	76.23	-	-	0.77	-	20	3	X		X	X
A-6	76.23	-	0.77	-	-	20	3	X		X	X
A-7	76.23	-	-	0.77	-	20	3		X	X	X
B-0	100	-	-	-	-	-	-	X			X
B-1	77	-	-	-	-	20	3	X			X
B-2	76.23	-	-	0.77	-	20	3	X			X
B-3	76.23	-	0.77	-	-	20	3	X			X
C-0	-	100	-	-	-	-	-	X			X
C-1	-	77	-	-	-	20	3	X			X
C-2	-	76.23	0.77			20	3	X			X
C-3	-	74.76	2.24	-	-	20	3	X			X
D-0	100	-	-	-	-	-	-	X			X
D-1	77	-	-	-	-	20	3	X			X
D-2	76.9	-	0.1	-	-	20	3	X			X
D-3	76.9	-	-	-	0.1	20	3	X			X
D-4	72	-	5	-	-	20	3		X		X
D-5	72	-	-	-	5	20	3		X		X
D-6	65	-	1	-	-	30	4	X			X
D-7	65	-	-	-	1	30	4	X			X
D-8	54	-	1	-	-	40	5	X			X
D-9	54	-	-	-	1	40	5	X			X
E-0	100	-	-	-	-	-	-	X			X
E-1	99	-	-	1	-	-	-	X			X
E-2	97	-	-	3	-	-	-	X			X
E-3	95	-	-	5	-	-	-	X			X

[0094] In Table 1, Sample A series (A-0 to A-8), C series (C-0 to C-3) and D series (D-0 to D-9) were compounded using the same extruder with the same production rate 1~2 kg/hour (masterbatch and dilution steps are 2 kg/hr; final compounding step is 1 kg/hr). Sample B

series (B-0 to B-3) were compounded using an industrial pilot-scale extruder (11 heating zones) with the production rate 13~20 kg/hour (masterbatch and dilution steps are 20 kg/hr; final compounding step is 13 kg/hr). Sample E series were compounded through master batch method (3-step process, without addition of biomass in step 3) and then diluted down to the target loading level, using an industrial pilot-scale extruder (11 heating zones) with the production rate 20 kg/hour.

Example 2 – Characterisation and test results

Density

[0095] All the samples were fully dried before the density test. A common immersing density measurement method was used and minimum 6 specimens were tested for every sample set.

[0096] The densities of various composite materials as well as neat PP-H are listed in Table 2.

Table 2: Density of various composite materials and neat PP-H

Sample	Density (g/cm ³)	Increase (100%)
B-0/A-0	0.9076 ± 0.0005	–
A-3	0.9943 ± 0.0012	9.56
B-1	0.9642 ± 0.0005	6.24
B-2	0.9873 ± 0.0005	8.78
B-3	0.9812 ± 0.0013	8.11

Shrinkage

[0097] The sample shrinkage after injection molding was measured. The sample dimensions were measured twice: before and after the injected test bars were stored a day in a condition chamber.

[0098] As is shown in Table 3, the shrinkage of the injected test bars is significantly reduced in all the composite materials relative to neat PP-H. The use of BM as a filler significantly improves the dimensional stability. The dimensional stability is further improved by adding 0.77 wt.% GNPs. The effect of G-S is more significant than G-M. Table 3 illustrates that PP has a high degree of shrinkage after having been molded into a component, which requires the design engineer to take this into account when producing components, especially those having tight dimensional tolerances. In contrast, the composite materials exhibit significantly reduced shrinkage, making it easier for design engineers to design complex components.

Table 3: Shrinkage results for various composite materials and neat PP-H

Sample	Shrinkage (100%)	Reduction (100%)
B-0	1.20	—
B-1	0.51	57.50
B-2	0.48	60.00
B-3	0.34	71.67

Tensile properties

[0099] Tensile tests were performed according to ASTM D3039 and minimum 6 specimens were tested for every sample. All the test results are listed in Table 4.

[00100] The tensile modulus of a composite is determined by the elastic moduli of both fillers and polymer matrix and the contents of the filler. Since both GNPs and BM have higher elastic moduli than PP, all the tensile moduli of all composites are increased relative to neat PP, as shown in Table 4. The tensile modulus of a composite can be affected by the crystallinity of the material, the alignment of the reinforced fillers and the crystal structure, as well as the dispersion of the fillers. The test bars made by injection may have better filler alignment and better crystal structure alignment than those made by compression due to the flow mechanism of the injection processing. The tensile modulus of A-0 (injection) is higher than A-0 (compression), which may be due to good alignment of crystal regions in PP-H during the injection processing. All the samples of B, C, D and E series were made by injection molding.

[00101] As the BM loading increases, the tensile properties of the composites increase. BM behaves as a reinforcement within the polymer matrix (A-3 vs A-0; B-1 vs B-0; C-1 vs C-0; D-1 vs D-0). With the addition of GNPs, the tensile properties of the composites are further improved (A-5 to A-7; B-2 and B-3; C-2 and C-3; D-2 to D-5), except G-L at low loading (D-3: 0.1 wt.% G-L). The synergistic effect of GNPs and BM further improved the tensile properties of the composites (A-5, A-7 and B-2 vs. A-3, B-1 and E-1).

[00102] Comparing B-2, B-3, D-2, D-3, D-4, and D-5, the results indicate that medium size GNPs (G-M) improve the tensile properties of the composites significantly. Small particle GNPs (G-S) improve the tensile performance slightly, and the large particles show an improvement at high loading level (D-5).

[00103] D-8 shows the best tensile performance: 128.60% improvement on tensile modulus and 56.57 improvement on tensile strength. The results obtained using 20 wt.% BM and 0.77

wt.% GNPs suggest that this improvement of tensile properties could be increased even further by using G-M instead of G-S.

[00104] The C series repeats the work of Idumah et al in Synthetic Metals 212 (2016) 91-104, albeit using hemp shive BM instead of kenaf fibre. The results in Table 4 illustrate show better performance on both tensile modulus and tensile strength than Idumah's work.

Table 4: Tensile properties of various composite materials and neat PP

Sample	Tensile Modulus (GPa)	Improvement (%)	Tensile Strength (MPa)	Improvement (%)
A-0 (Compression)	0.375 ± 0.059	—	29.2 ± 0.56	—
A-0 (Injection)	0.656 ± 0.036	—	31.46 ± 0.24	—
A-1 (Compression)	0.536 ± 0.093	43	29.3 ± 0.98	0
A-2 (Compression)	0.624 ± 0.045	66	26.0 ± 0.77	-11
A-3 (Compression)	0.789 ± 0.047	110	33.3 ± 3.1	14
A-3 (Injection)	1.070 ± 0.049	63	38.76 ± 0.44	23
A-4 (Compression)	0.691 ± 0.039	84	30.5 ± 2.4	4
A-5 (Compression)	0.749 ± 0.046	100	30.6 ± 2.6	5
A-5 (Injection)	1.306 ± 0.028	99	41.46 ± 0.36	32
A-6 (Compression)	0.704 ± 0.034	88	23.6 ± 0.7	-19
A-6 (Injection)	1.220 ± 0.032	86	39.07 ± 1.48	24
A-7 (Compression)	0.727 ± 0.011	94	20.9 ± 3.1	-28
A-7 (Injection)	1.262 ± 0.063	92	41.5 ± 1.21	32
B-0	1.25 ± 0.01	—	34.28 ± 0.42	—
B-1	1.88 ± 0.01	49.71	41.22 ± 0.22	20.23
B-2	2.21 ± 0.02	76.51	44.46 ± 0.29	29.70
B-3	2.12 ± 0.02	69.01	42.37 ± 0.50	23.60
C-0	0.82 ± 0.02	—	26.46 ± 0.10	—
C-1	1.35 ± 0.01	63.82	33.21 ± 0.33	25.54
C-2	1.59 ± 0.07	93.31	36.84 ± 0.36	39.24
C-3	1.50 ± 0.01	81.59	34.39 ± 0.06	29.98
D-0	1.25 ± 0.01	—	34.28 ± 0.42	—
D-1	1.88 ± 0.01	49.72	41.22 ± 0.22	20.23
D-2	1.99 ± 0.06	58.62	42.29 ± 0.31	23.36
D-3	1.73 ± 0.05	38.17	39.75 ± 0.34	15.96
D-4	2.33 ± 0.02	85.86	42.67 ± 0.05	24.48
D-5	2.20 ± 0.02	75.38	42.41 ± 0.15	23.71
D-6	2.57 ± 0.02	105.36	48.54 ± 0.13	41.60
D-7	2.26 ± 0.05	80.38	45.19 ± 0.54	31.82
D-8	2.86 ± 0.04	128.60	53.67 ± 0.24	56.57
D-9	2.58 ± 0.06	106.26	50.09 ± 0.78	46.11
E-0	0.585 ± 0.011	—	31.36 ± 0.46	—
E-1	0.728 ± 0.005	24.40	32.81 ± 0.63	4.64
E-2	0.792 ± 0.014	35.41	32.26 ± 0.18	2.86
E-3	0.851 ± 0.009	45.53	30.59 ± 0.24	-2.45

Flexural properties

[00105] All the flexural tests were performed according to ASTM D790 and minimum 6 specimens were tested for every sample. The results are listed in Table 5.

[00106] Table 5 shows that the addition of BM can improve the flexural properties of PP composites. Following the trends observed for tensile performance, Table 5 shows that adding a small amount of GNPs into the formulation improves the flexural performance of the composites even more. The results suggest that smaller GNPs particle sizes (e.g. G-S) give rise to improved flexural performance. The effect of GNPs particle size on the tensile and flexural properties of the composite allows the properties of the material to be tailored according to the intended application.

Table 5: Flexural properties of various composite materials and neat PP

Sample	Flexural Modulus (GPa)	Improvement (%)	Flexural Strength (MPa)	Improvement (%)
B-0	1.51 ± 0.02	—	41.42 ± 0.36	—
B-1	3.08 ± 0.04	103.78	68.45 ± 0.35	65.27
B-2	3.33 ± 0.03	120.01	74.49 ± 0.12	79.85
B-3	3.45 ± 0.06	128.05	76.29 ± 0.62	84.20
C-0	1.09 ± 0.01	—	30.58 ± 0.17	—
C-1	2.07 ± 0.04	90.15	48.54 ± 0.73	58.72
C-2	2.12 ± 0.01	94.47	49.68 ± 0.32	62.43
C-3	2.24 ± 0.04	105.78	49.90 ± 0.33	63.16
D-0	1.51 ± 0.02	—	41.42 ± 0.36	—
D-1	3.08 ± 0.04	103.78	68.45 ± 0.35	65.27
D-2	2.78 ± 0.03	84.05	61.73 ± 0.19	49.05
D-3	2.78 ± 0.07	83.82	62.73 ± 0.59	51.47
D-4	3.09 ± 0.03	104.53	62.39 ± 0.07	50.63
D-5	2.95 ± 0.04	95.43	61.67 ± 0.34	48.89
D-6	3.91 ± 0.11	158.82	74.45 ± 0.93	79.75
D-7	3.58 ± 0.10	136.96	70.50 ± 0.61	70.22
D-8	4.70 ± 0.10	210.83	79.78 ± 0.76	92.62
D-9	4.25 ± 0.07	181.43	76.85 ± 0.41	85.55

Izod impact strength test (notched)

[00107] All the impact tests were performed according to ISO 180 at room temperature and minimum 10 specimens were tested for every sample. All the test specimens were notched. Table 6 lists the results.

Table 6: Izod impact strength of various composite materials and neat PP

Sample	Impact Strength (KJ/m ²)	Increase (%)
B-0	4.30	
B-1	3.98	-7.44
B-2	3.90	-9.30
B-3	3.70	-13.95
C-0	5.38 ± 0.69	
C-2	2.33 ± 0.28	-56.66
D-0	3.00	
D-6	1.75 ± 0.15	-41.63
D-7	1.75 ± 0.09	-41.80
D-8	1.58 ± 0.08	-47.46
D-9	1.73 ± 0.14	-42.48

Heat distortion temperature (HDT)

[00108] The heat distortion temperature determines the maximum service temperature of the material before the material deforms. HDT of B series was evaluated according to ISO 75 and the testing condition was 1.8 MPa in Flatwise. The results are listed in Table 7.

[00109] Table 7 shows that as BM was added into the PP, there is significant increase in the HDT (34 °C increase), meaning that the material can still maintain its structure at much higher temperature under a constant bending force without any deformation. As a small amount of GNPs (0.77 wt.%) was added into the formulation, HDT of both composites are further increased. The results suggest that the physical properties of GNPs can affect the HDT. The smaller the lateral dimension of GNPs is, the higher the HDT of the composites. The effect of lateral dimension of GNPs on HDT is the similar to its effect on shrinkage.

Table 7: HDT of various composite materials and neat PP

Sample	HDT (°C)	Increase (°C)
B-0	58	—
B-1	92	34
B-2	95	37
B-3	101	43

Coefficient of linear thermal expansion (CLTE)

[00110] The coefficient of thermal expansion (CTE) shows how the material expands or shrinks as the temperature increases. CLTE of the samples were measured through both the thickness and width of the test bars using thermomechanical analysis with a temperature ramp

of 5 °C/min from -30 °C to 160 °C under nitrogen at 50 ml/min. The CLTE results calculated from 20 °C to 70 °C are listed in Table 8.

[00111] The results indicate that the addition of BM can decrease the thermal expansion of the composite significantly, meaning that BM/PP-H composites expand less than neat PP-H. The addition of GNPs having different lateral size can affect the thermal expansion.

Table 8: CLTE of various composite materials and neat PP

Sample	CLTE-Thickness ($\mu\text{m}/(\text{m}\cdot^{\circ}\text{C})$)	Reduction (%)	CLTE-Width ($\mu\text{m}/(\text{m}\cdot^{\circ}\text{C})$)	Reduction (%)
B-0	131.41	—	122.17	—
B-1	123.13	6.30	105.03	14.03
B-2	128.26	2.40	107.24	12.22
B-3	138.41	-5.33	109.49	10.38

Moisture absorption

[00112] The moisture absorption of B series and E-1 samples were tested. A minimum of 5 specimens per sample were tested. All specimens were immersed in distilled water at 22 °C.

[00113] As shown in Fig. 2, the presence of GNPs does not affect the moisture absorption of neat PP, which absorbs a negligible quantity (less than 0.05 wt%) of water. Irrespective of particle size, the presence of GNPs in the composites (B-2 and B-3) reduce the water absorption when compared with BM/PP (B-1). As the testing time increases, the gap between B-1 and B-2/B-3 is enlarged gradually. In the initial 24 hours, G-S shows better barrier performance than G-M (shown in Fig.1), meaning that the properties of the composite can be tailored according to the intended application.

[00114] While specific embodiments of the invention have been described herein for the purpose of reference and illustration, various modifications will be apparent to a person skilled in the art without departing from the scope of the invention as defined by the appended claims.

CLAIMS

1. A composite material comprising:
25 – 80 wt% of at least one polymer selected from the group consisting of polypropylene, polyethylene, polystyrene, polypropylene-co-ethylene, polypropylene-co-styrene, polyethylene-co-styrene and polypropylene-co-ethylene-co-styrene;
15 – 70 wt% of at least one lignocellulosic compound, wherein the at least one lignocellulosic compound is a shive;
2.0 – 11.0 wt% of at least one coupling agent; and
0.01 – 5.5 wt% of graphene.
2. The composite material of claim 1, wherein the material comprises 17.5 – 65 wt% of the at least one lignocellulosic compound.
3. The composite material of claim 1, wherein the material comprises 37.5 – 50 wt% of the at least one lignocellulosic compound.
4. The composite material of claim 1, wherein the material comprises 17 – 25 wt% of the at least one lignocellulosic compound.
5. The composite material of any preceding claim, wherein the material comprises 30 – 77.5 wt% of the at least one polymer.
6. The composite material of any preceding claim, wherein the material comprises 45 – 57.5 wt% of the at least one polymer.
7. The composite material of any one of claims 1 to 4, wherein the material comprises 70 – 78 wt% of the at least one polymer.
8. The composite material of any preceding claim, wherein the at least one lignocellulosic compound is selected from the group consisting of hemp shive and flax shive.
9. The composite material of any preceding claim, wherein the at least one lignocellulosic compound comprises 20 – 55 wt% of cellulose, 10 – 35 wt% of hemicellulose and 12 – 50 wt% of lignin.

10. The composite material of any preceding claim, wherein the at least one polymer is selected from the group consisting of polypropylene, polypropylene-co-ethylene, polypropylene-co-styrene and polypropylene-co-ethylene-co-styrene.
11. The composite material of claim 10, wherein propylene is the dominant repeating unit by mass in polypropylene-co-ethylene, polypropylene-co-styrene and polypropylene-co-ethylene-co-styrene.
12. The composite material of any preceding claim, wherein the at least one polymer is polypropylene or polypropylene-co-ethylene.
13. The composite material of any preceding claim, wherein the material comprises 0.5 – 2 wt% of graphene.
14. The composite material of any preceding claim, wherein the graphene has an average thickness of 4 – 10 nm.
15. The composite material of any preceding claim, wherein the graphene has an average particle size of 1 – 35 μm .
16. The composite material of any preceding claim, wherein the graphene has an average particle size of 2.5 – 7.5 μm .
17. The composite material of any preceding claim, wherein the material comprises 2.5 – 9.5 wt% of the at least one coupling agent.
18. The composite material of any preceding claim, wherein the at least one coupling agent is selected from the group consisting of an amphiphilic copolymer, a (5-35C)fatty acid, a (5-35C) fatty acid anhydride, a (5-35C)fatty acid ester and an organosilane.
19. The composite material of any preceding claim, wherein the at least one coupling agent is polypropylene-graft-maleic anhydride comprising 0.5 – 4 wt% of maleic anhydride.
20. A process for the preparation of a composite material as defined in any preceding claim, the process comprising the steps of:
 - A) mixing the at least one polymer, the at least one lignocellulosic compound, the at least one coupling agent and graphene, and

B) extruding the mixture resulting from step A).

21. The process of claim 20, wherein the at least one polymer, and optionally also the at least one coupling agent are molten during step A).
22. The process of claim 20 or 21, wherein step A) comprises the sub steps:
 - A1) mixing the at least one polymer, the at least one coupling agent and graphene, and
 - A2) mixing the mixture resulting from step A1) with the at least one lignocellulosic compound.
23. The process of claim 22, wherein step A1) comprises mixing the at least one polymer, the at least one coupling agent and graphene to produce a masterbatch, and then mixing the masterbatch with an additional quantity of the at least one polymer
24. The process of claim 22 or 23, wherein
 - step A1) is conducted at a temperature of 150 – 250°C, and/or
 - step A2) is conducted at a temperature of 150 – 185°C.
25. Use of a composite material as defined in any one of claim 1 to 19 in a container or in a component for an automobile.

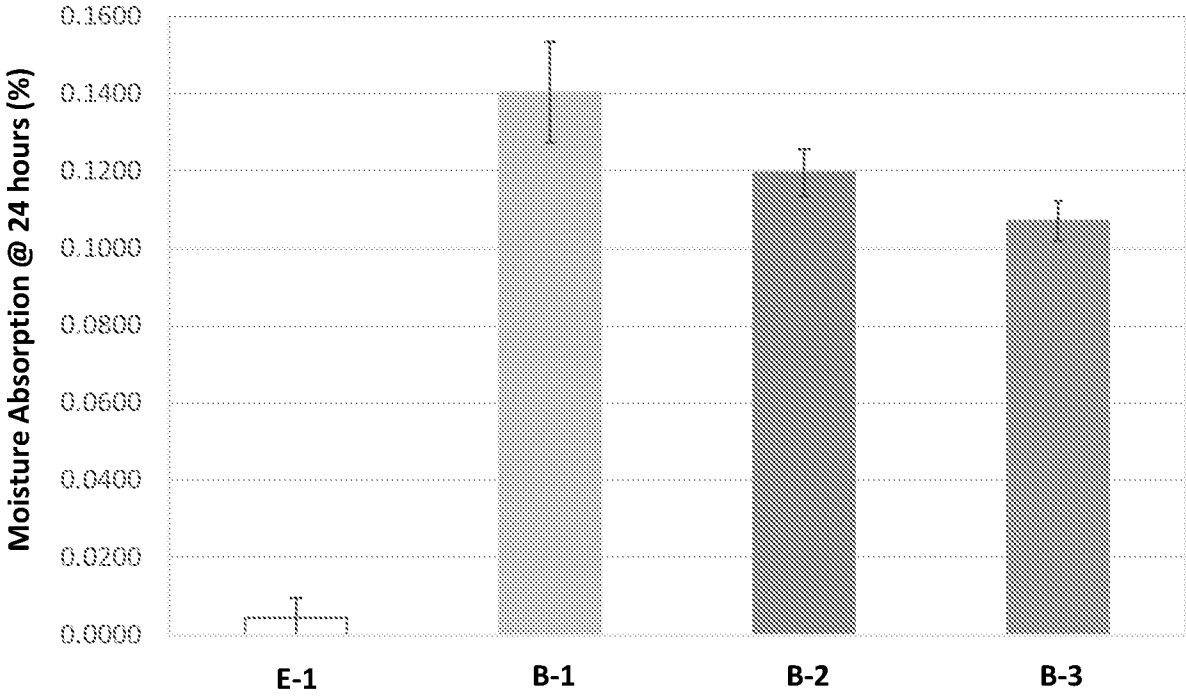


FIGURE 1

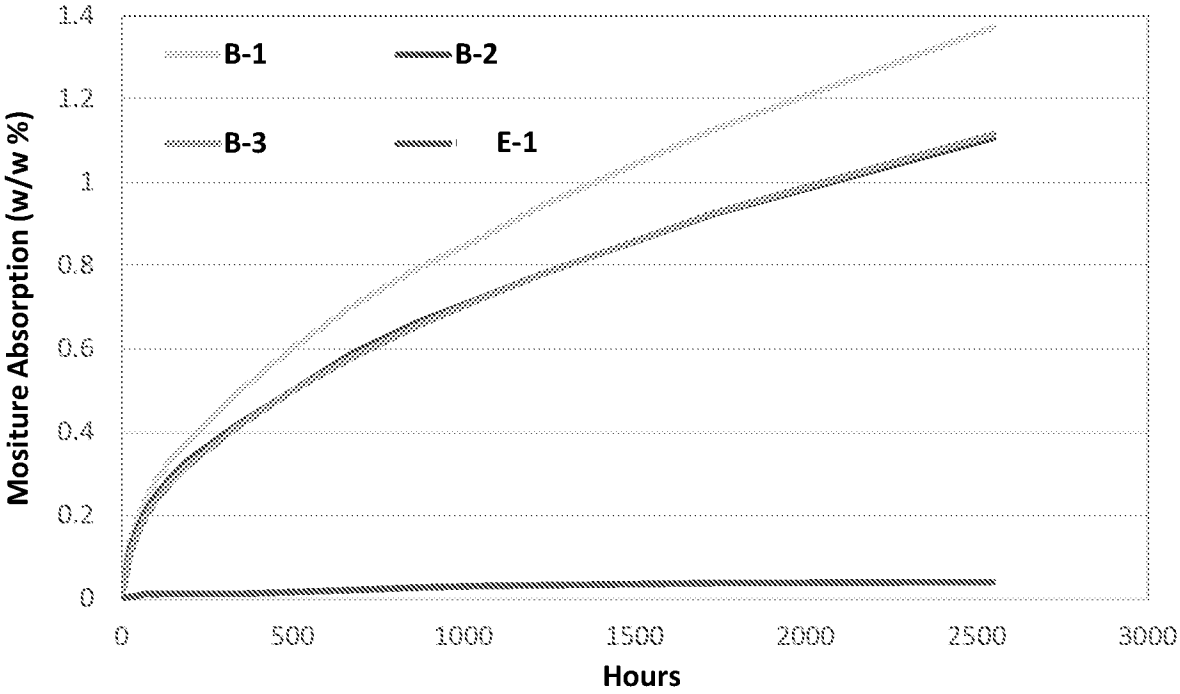


FIGURE 2

INTERNATIONAL SEARCH REPORT

International application No

PCT/GB2021/051727

A. CLASSIFICATION OF SUBJECT MATTER

INV. C08L23/12 C08L97/02
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)

C09J 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

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	WANG JIE ET AL: "Effect of a coupling agent on the properties of hemp-hurd-powder-filled styrene-butadiene rubber", JOURNAL OF APPLIED POLYMER SCIENCE, vol. 121, no. 2, 15 July 2011 (2011-07-15) , pages 681-689, XP055847889, US ISSN: 0021-8995, DOI: 10.1002/app.33744 the whole document ----- -/--	1-25



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

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"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

5 October 2021

Date of mailing of the international search report

20/10/2021

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INTERNATIONAL SEARCH REPORT

International application No

PCT/GB2021/051727

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	KAUSAR AYESHA: "Technical viewpoint on polystyrene/graphene nanocomposite", JOURNAL OF THERMOPLASTIC COMPOSITE MATERIALS, 24 February 2020 (2020-02-24), XP055847968, US ISSN: 0892-7057, DOI: 10.1177/0892705720907653 the whole document -----	1-25
Y	WANG W-P ET AL: "Preparation and characterization of polystyrene/graphite composite prepared by cationic grafting polymerization", POLYMER, ELSEVIER SCIENCE PUBLISHERS B.V, GB, vol. 45, no. 12, 1 May 2004 (2004-05-01), pages 3987-3995, XP004508683, ISSN: 0032-3861, DOI: 10.1016/J.POLYMER.2004.04.023 the whole document -----	1-25
T	RAZA YASIR ET AL: "Production and investigation of mechanical properties of graphene/polystyrene nano composites", JOURNAL OF POLYMER RESEARCH, SPRINGER NETHERLANDS, NL, vol. 28, no. 6, 19 May 2021 (2021-05-19), XP037458770, ISSN: 1022-9760, DOI: 10.1007/S10965-021-02560-8 [retrieved on 2021-05-19] the whole document -----	
A	CRINI GRÉGORIO ET AL: "Applications of hemp in textiles, paper industry, insulation and building materials, horticulture, animal nutrition, food and beverages, nutraceuticals, cosmetics and hygiene, medicine, agrochemistry, energy production and environment: a review", ENVIRONMENTAL CHEMISTRY LETTERS, SPRINGER-VERLAG, HEIDELBERG, DE, vol. 18, no. 5, 8 June 2020 (2020-06-08), pages 1451-1476, XP037229203, ISSN: 1610-3653, DOI: 10.1007/S10311-020-01029-2 [retrieved on 2020-06-08] the whole document -----	1-25