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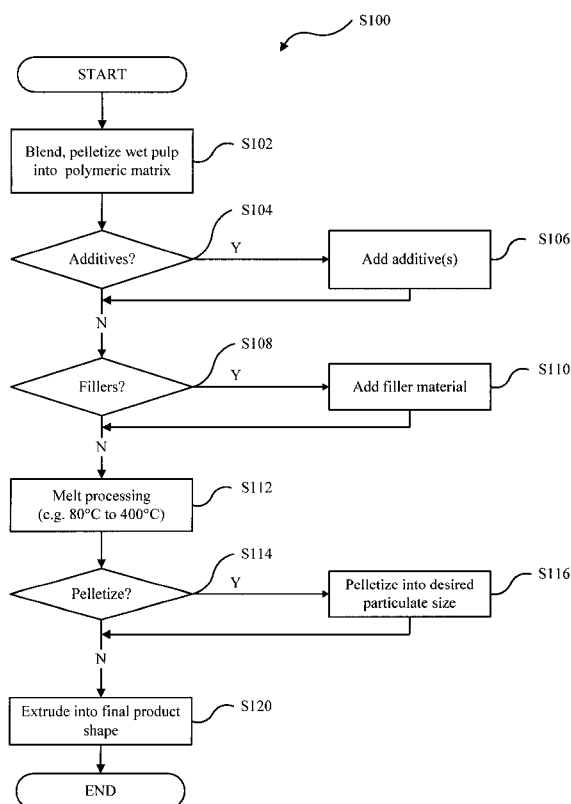
(54) **Title:** CELLULOSIC COMPOSITES COMPRISING WOOD PULP

FIG. 1

(57) **Abstract:** The present invention discloses cellulosic composites that include mechanical or chemical pulp, and methods for producing such cellulosic composites. Embodiments of such composites may exhibit improved mechanical properties and moisture resistance when compared to composites derived from conventional cellulosic feedstock.



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## CELLULOSIC COMPOSITES COMPRISING WOOD PULP

[0001] This application claims the benefit of U.S. Provisional Application No. 62/315,731 filed on March 31, 2016, and U.S. Provisional Application No. 62/315,722 filed on March 31, 2016, and U.S. Provisional Application No. 62/315,737 filed on March 31, 2016, and  
5 U.S. Provisional Application No. 62/315,744 filed on March 31, 2016, the contents of each of which are incorporated herein by reference.

## TECHNICAL FIELD

[0002] The present invention relates to cellulosic composites that include mechanical or chemical wood pulp, and methods for producing such cellulosic composites. Embodiments  
10 of such composites may exhibit improved mechanical properties and moisture resistance when compared to composites derived from conventional cellulosic feedstock.

## BACKGROUND ART

[0003] There is increasing demand for cost effective composite materials derived from renewable feedstock. Cellulosic materials have been previously used as fillers in  
15 thermoplastics. For example, wood plastic composites (WPCs) have found application in a multitude of commercial products in recent years, and the overall market for WPCs is estimated to be billions of dollars annually. WPCs are widely used in the construction and automotive industries. WPCs are often more cost effective than conventional mineral or glass filled composites. WPCs also generally have the look of natural wood, which can be  
20 desirable. However, conventional WPCs typically have poorer mechanical properties and lower moisture resistance than to mineral and glass filled composites.

[0004] Cellulosic composites based on chemically processed pulp have been recently developed and commercialized. Chemically processed pulp is cellulosic material produced using chemical pulping processes, such as kraft or sulfite processes, that involve chemical  
25 treatment and high temperatures. These chemical pulping processes remove much of the

hemicelluloses and lignin from the pulp, so that the resulting chemical pulp contains little or no residual lignin. For example, the lignin content of chemical pulp is usually less than 5% and often less than 1% by weight.

5 [0005] Composites based on chemically processed pulp generally have improved mechanical properties, lower odor than WPCs. Further, chemically processed pulp composites can be pigmented or colored like conventional thermoplastics. However, composites based on chemically processed pulp are typically more expensive than WPCs and mineral filled composites. They also have poorer mechanical properties and moisture resistance when compared to mineral or glass filled composites. For these reasons, to date,  
10 commercial adoption of composites based on chemically processed pulp has usually occurred in niche applications.

[0006] It is therefore desirable to find cellulosic composites that have improved moisture resistance or improved mechanical properties than conventional known cellulosic composites.

## 15 SUMMARY OF INVENTION

[0007] According to an aspect of the invention, there is provided a cellulosic composite that includes a mechanical pulp having lignin and a polymeric matrix. The lignin in the mechanical pulp is about 15% to 35% of the dry weight of the mechanical pulp.

20 [0008] According to another aspect of the invention, there is provided a process for making a cellulosic composite including the steps of melt processing a mixture comprising mechanical pulp and a polymeric matrix to form a masterbatch; and further melt processing the masterbatch to form the composite.

[0009] According to yet another aspect of the invention, there is provided a process for making an article of manufacture including the steps of melt processing a mixture  
25 comprising mechanical pulp and a polymeric matrix; and extruding the melt processed mixture into the article.

## BRIEF DESCRIPTION OF DRAWINGS

[0010] In the figures which illustrate by way of example only, embodiments of the present invention,

[0011] FIG. 1 is a simplified flowchart depicting steps in an exemplary process for making cellulosic composites and articles manufactured therefrom; and

[0012] FIG. 2 is a flowchart diagram depicting steps involved in a process, exemplary of an embodiment of the present invention, for preparing cellulosic composites comprising mechanical pulp.

## DESCRIPTION OF EMBODIMENTS

[0013] Aspects of the present disclosure relate to cellulosic composites based on wood pulps. Some embodiments of these composites are more cost effective, have improved moisture resistance and/or improved mechanical properties when compared to cellulosic composites known in the art, such as composites based on chemically processed pulp.

[0014] Cellulosic composites that contain wood pulp and a polymeric matrix are described. When such pulp is incorporated into a polymeric matrix, it is capable of improving certain attributes of the resulting polymeric composite. In certain embodiments, such composites have exhibited improved mechanical properties and improved moisture resistance.

[0015] Exemplary processes for the preparation of cellulosic composites that have wood pulp incorporated in a polymeric matrix are also described. Exemplary embodiments of these processes involve melt processing pulp with a relatively high moisture content. The moisture content of the pulp prior to melt processing may be greater than 10 wt% in some embodiments; while in other embodiments, the moisture content is greater than 20 wt%; and in yet other embodiments, the moisture content may be greater than 30 wt%.

[0016] These processes for creating cellulosic composites can enable the preparation of composites with pulp substantially uniformly dispersed in a polymeric matrix. In some embodiments they are used to produce a masterbatch, comprising a high concentration of pulp in a polymeric matrix. The masterbatch can be let down or diluted with further polymeric matrix to a desired loading level.

[0017] The use of wet pulp or pulp with high moisture content in these exemplary embodiments of the processes described herein has the beneficial effect of enabling the preparation of composites with pulp that is substantially uniformly dispersed in the polymeric matrix. The pulp is better separated and randomized within the matrix, with fewer hydrophilic clumps.

[0018] Exemplary cellulosic composites as described herein may also include one or more additives that further improve one or both of the mechanical or chemical properties of the composites. For example, in some embodiments, the cellulosic composites may include a coupling agent or an antioxidant.

[0019] Other exemplary cellulosic composites may also include one or more additional fillers that further improve the mechanical or chemical properties of the composites.

[0020] Yet other embodiments of the cellulosic composites described herein can be converted into articles using conventional extrusion and molding techniques. These articles have utility in a variety of markets including automotive, building and construction, consumer and appliance applications, and 3D printing among others.

[0021] The following terms found in this disclosure are used as follows:

[0022] “Cellulosic Composite” is used to refer to a composite material that comprises a polymeric matrix and a cellulosic filler.

[0023] “Chemically processed pulp” or “chemical pulp” is used to refer to cellulosic material produced using a chemical pulping process, such as kraft or sulfite pulping processes.

5 [0024] “Composite” is used to refer to a material comprising a polymeric matrix and a filler.

[0025] “Coupling Agent” is used to refer to an additive that improves the interfacial adhesion between a polymeric matrix and a cellulosic filler.

10 [0026] “Mechanical Pulp” is used to refer to a cellulosic material produced using a mechanical pulping process or a chemi-mechanical pulping process. For example, “mechanical pulp” includes, thermo-mechanical pulp (TMP), refiner mechanical pulp (RMP), chemi-thermomechanical pulp (CTMP), medium density fiber (MDF), ground wood pulp (GWP) and bleached chemi-thermomechanical pulp (BCTMP).

15 [0027] “Melt Processable Composition” is used to refer to a formulation that is capable of being melt processed, typically at elevated temperatures, by means of conventional polymer melt processing techniques such as extrusion or injection molding, for example.

[0028] “Melt Processing Techniques” is used to refer to various melt processing techniques that may include, for example, extrusion, injection molding, blow molding, rotomolding, thermokinetic mixing or batch mixing.

[0029] “Polymeric Matrix” is used to refer to a melt processable polymeric material.

20 [0030] The description is not intended to describe all embodiments or every possible implementation of the present technology. The detailed description is intended to provide some illustrative embodiments.

25 [0031] The present disclosure relates to composites based on wood pulp that, in at least some embodiments, are more cost-effective, and/or have improved moisture resistance and/or have improved mechanical properties compared to known cellulosic composites.

The cellulosic composites described herein include wood pulp incorporated into a polymeric matrix.

[0032] The wood pulp that is incorporated into the cellulosic composites may be manufactured using a mechanical or a chemi-thermomechanical process. In producing  
5 mechanical pulp, wood is generally ground against a water-lubricated abrasive surface such as a rotating stone. The heat generated by grinding softens the lignin binding the fibers, and the mechanized forces separate the fibers referred to as ground wood (GWD).

[0033] In embodiments of the present invention, refiners may be used. Refiners were developed during the second half of the 20<sup>th</sup> century. In a refiner, woodchips are subjected to  
10 intensive shearing forces between a rotating steel disc and a fixed plate to produce refiner mechanical pulp (RMP). In subsequent modifications to this process, the woodchips are pre-softened by heat make the fibrillation more effective, producing thermo-mechanical pulp (TMP) that is generally light-colored and has longer fibers.

[0034] A further development of thermo-mechanical pulp is CTMP, in which the wood  
15 chips are impregnated with chemicals such as sodium sulphite before the grinding step. The resulting CTMP is generally even lighter in color and has better strength characteristics. After grinding, the pulp is sorted by screening to remove unrefined fiber bundles. It can then be further bleached with peroxide to produce BCTMP for use in higher value-added products.

20 [0035] The mechanical pulp used in the composites described is characterized by having a high content of residual lignin. In particular the lignin content in mechanical pulp is higher than in chemical pulps. In some embodiments, the lignin content in the mechanical pulp is greater than 10 wt%; while in other embodiments the lignin content in the mechanical pulp is greater than 15 wt%; and in yet other embodiments the lignin content in the mechanical  
25 pulp may be greater than 20 wt%. In some cases the mechanical pulp is a chemi-thermomechanical pulp with a lignin content in the range of 15 wt% to 35 wt%. These are dry weight percentages of lignin in the mechanical pulp.

[0036] The wood pulp can be incorporated into any suitable polymeric matrix to form cellulosic composites with enhanced mechanical and chemical properties. In some embodiments, the pulp loading in the polymeric matrix is in the range of about 5-99 wt%; while in other embodiments, the pulp loading is in the range of about 10-95 wt%; and in yet other embodiments the pulp loading is between 10-90 wt%. In some embodiments the pulp loading in a masterbatch is in the range of 50-99 wt%. These are dry weight percentages of pulp in the final composite or masterbatch.

[0037] The polymeric matrix may comprise one or more polymers. Non-limiting examples of polymers that can be used in embodiments of the present cellulosic composites include: high density polyethylene (HDPE), low density polyethylene (LDPE), linear low density polyethylene (LLDPE), functional polyolefin copolymers including polyolefin-based ionomers, polypropylene (PP), polyolefin copolymers (e.g., ethylene-butene, ethylene-octene, ethylene vinyl alcohol), polystyrene, polystyrene copolymers (e.g., high impact polystyrene, acrylonitrile butadiene styrene copolymer), polyacrylates, polymethacrylates, polyesters, polyvinylchloride (PVC), fluoropolymers, polyamides, polyether imides, polyphenylene sulfides, polysulfones, polylactic acid (PLA), polyacetals, polycarbonates, polyphenylene oxides, polyurethanes, thermoplastic elastomers (e.g., SIS, SEBS, SBS), or combinations thereof. For some end-use applications, polyolefins are well-suited to serve as polymeric matrices, for example, in articles useful as automotive components.

[0038] Bioplastics polymers useful in this invention include, biobased, biodegradable or compostable polyesters, polyamides, polyurethanes, polyacrylates, polyolefins, thermoplastic starches and cellulotics. Bioplastics of particular interest include biobased, biodegradable or compostable polyesters. Non limiting examples of biobased or biodegradable or compostable polyesters include: PLA (Polylactic acid), PHA (Polyhydroxyalkanoates), PBAT (polybutyrate adipate terephthalate), PBS (polybutylene succinate), PCL (polycaprolactones), PGA (Polyglycolic acid).

[0039] Polylactic acid is increasingly proving to be a viable alternative to petrochemical-based plastics in many applications. PLA is produced from renewable resources and is biodegradable. This makes it well suited for green or environmentally sensitive applications. In addition, PLA has unique physical properties that make it useful in several industrial applications including paper coating, fibers, films, packaging materials and the like.

[0040] The polymeric matrix may optionally contain one or more additives. Non-limiting examples of conventional additives include antioxidants, light stabilizers, fibers, blowing agents, foaming additives, antiblocking agents, heat stabilizers, impact modifiers, biocides, antimicrobial additives, compatibilizers, plasticizers, tackifiers, processing aids, lubricants, coupling agents, flame retardants, pigments and colorants. The additives may be incorporated into the melt processable composition in the form of powders, pellets, granules, or in other extrudable forms. The amount and type of additives incorporated in the melt processable composition can be suitably chosen, depending upon the polymeric matrix and the desired physical properties of the finished composition. Those skilled in the art of melt processing are capable of selecting appropriate amounts and types of additives for a specific polymeric matrix and mechanical pulp filler in order to achieve desired physical properties of the finished composite material.

[0041] Some embodiments of the present cellulosic compositions include coupling agents and/or antioxidants as additives. Non-limiting examples of coupling agents include silanes, zirconates, titanates and functionalized polymers. Preferred coupling agents include silane and maleic anhydride grafted polymers. Non-limiting examples of maleic anhydride grafted polymers include those sold under the tradenames Polybond (Addivant), Extinity (NWP), Integrate (Lyondell Basell), and Fusabond (DuPont). Preferred antioxidants include monomeric, polymeric and oligomeric phenols. Non-limiting examples of antioxidants include those sold under the tradenames Irganox, Irgaphos (BASF) and Hostanox (Clariant). Typical loading levels of coupling agents and antioxidants are approximately 0.1 to 10 wt% of the composite formulation.

[0042] Some embodiments of the present cellulosic compositions comprise one or more additional fillers. These can be incorporated in the melt processable composition, and can be used to adjust the mechanical properties of the final cellulosic composite material or articles made therefrom. For example, fillers can function to improve mechanical and thermal properties of the cellulosic composite. Fillers can also be utilized to adjust the coefficient of thermal expansion (CTE) of the cellulosic composite, to make it more compatible with other materials with which it is to be used, for example. Non-limiting examples of fillers include mineral and organic fillers (e.g., talc, mica, clay, silica, alumina, carbon fiber, carbon black, glass fiber) and conventional cellulosic materials (e.g., wood flour, wood fibers, non-wood plant fibers, sawdust, wood shavings, newsprint, paper, flax, hemp, wheat straw, rice hulls, kenaf, jute, sisal, peanut shells, soy hulls, or other cellulose containing materials), and optionally additional lignin. The amount and type of filler in the melt processable composition can be suitably chosen depending upon the polymeric matrix and the desired physical properties of the finished composition. Fillers such as calcium carbonate, talc, clay and cellulosic fiber are well-suited for many applications. In some embodiments, the additional filler makes up 1 wt% to 90 wt% of the composite; in some embodiments, 5 wt% to 75 wt % of the composite; and in some embodiments 1 wt% to 60 wt % of the composite.

[0043] Cellulosic composites based on pulp, and incorporating optional additives and/or additional fillers, can be prepared by blending the components into the polymeric matrix. Depending on the type and nature of polymeric matrix, this can be done using a variety of conventional mixing processes. For melt processable thermoplastic compositions, the polymeric matrix and additives can be combined by any suitable blending technique commonly employed in the plastics industry, such as with a compounding mill, a Banbury mixer, or a mixing extruder. The mixing operation may be conveniently carried out at a temperature above the melting point or softening point of the polymeric matrix. In some cases melt processing of the mixture is performed at a temperature from 80 °C to 400 °C, although suitable operating temperatures are selected depending upon the melting point, melt viscosity, and thermal stability of the composite formulation. Different types of melt

processing equipment, such as extruders, may be used to process the melt processable compositions described herein.

[0044] The resulting melt-blended mixture can be either extruded directly into the form of the final product shape or can be pelletized or otherwise comminuted into a desired particulate size or size distribution, and then fed to an extruder, such as a twin-screw extruder, that melt-processes the blended mixture to form the final product shape.

[0045] A flowchart depicted in FIG. 1 illustrates the above exemplary process. An exemplary process S100 starts with mixing wet pulp into a polymer matrix in step S102. If the optional use of additives is desired in step S104 then additives are added to the mixture (step S106), but otherwise the step S106 is bypassed. Similarly, if the optional use of fillers is desired (step S108) then fillers are added to the mixture (step S110), but otherwise step S110 is bypassed. As noted above, for melt processable thermoplastic compositions, the polymeric matrix and additives can be combined by any suitable blending technique such as with a compounding mill, a Banbury mixer, or a mixing extruder. In Step S112 melt processing is used, which in this embodiment may be at a temperature from 80°C to 400°C. Depending on the decision on whether to pelletize/comminute at step S114, the output of step S112 is either extruded directly into the form of the final product shape (step S120) or can be pelletized or otherwise comminuted (S116) into a desired particulate size or size distribution, and then fed to an extruder, such as a twin-screw extruder, that melt-processes the blended mixture to form the final product shape (step S120).

[0046] In known processes for the preparation of cellulosic composites, the pulp is typically dry or has a low moisture content. It can be difficult to pull apart or break up the pulp and achieve adequately uniform dispersion of the pulp in the polymeric matrix.

[0047] Preferred embodiments of processes for the preparation of cellulosic composites comprising mechanical or chemical pulp involve melt processing the pulp with a relatively high moisture content. In some embodiments, the moisture content of the pulp prior to melt processing is greater than 10 wt%; in preferred embodiments, the moisture content is greater

than 20 wt%; and in particularly preferred embodiments, the moisture content is greater than 30 wt%. In some embodiments, the moisture content of the pulp prior to melt processing is in the range of 40 wt% to 60 wt%.

5 [0048] It has been found that using mechanical or chemical pulp with a relatively high moisture content can facilitate the dispersion of the pulp in the polymeric matrix during melt processing of the mixture. Addition of a solvent can further improve dispersion. The moisture tends to aid separation of the pulp fibers, and moist pulp tends to break apart more easily than dry pulp, which has a tendency to become matted and consolidated. The water is gradually removed, for example by evaporation, during the melt processing steps. This  
10 innovative wet processing approach can enable the preparation of composites with pulp substantially uniformly dispersed in a polymeric matrix, and can enable the preparation of composites with high loadings of pulp in a polymeric matrix. For example, loadings of greater than 95 wt% in the composites can be achieved.

15 [0049] In some embodiments of processes for the preparation of cellulosic composites comprising pulp, the pulp is provided dry or with a low moisture content, and the water content of the pulp is increased prior to melt processing the pulp with the polymeric matrix. For example, dry cakes of pulp can be rehydrated by adding water, and then the pulp and can be melt processed with the polymeric matrix and any other desired components of the composite as described herein.

20 [0050] In some embodiments of processes for preparing the present cellulosic composites and articles made therefrom, the cellulosic composites are produced in a process involving two melt processing steps. First, a masterbatch of the pulp composite is produced by melt processing pulp with a high moisture content as described above, with a thermoplastic polymeric matrix, and optionally other additives or fillers. The resulting masterbatch has a  
25 high concentration of pulp, and can be subsequently letdown (or diluted) to a more suitable loading level for the final application using a second melt processing step (e.g., compounding, injection molding or extrusion). In preferred embodiments, the masterbatch

has a pulp content in the range of about 50 wt% to 99 wt%, and the letdown has a pulp content in the range of about 5 wt% to 50 wt%.

[0051] The above exemplary process is illustrated in **FIG. 2**. As shown, an exemplary process **S200** starts with obtaining pulp **S201**. As noted above, the pulp at step **S201** may be provided dry or with a low moisture content. In step **S202** moisture is introduced into the pulp, for example, by adding water.

[0052] In step **S203** the pulp is mixed or combined with the polymeric matrix. If the optional use of additives is desired in step **S204** then additives are added to the mixture (step **S206**), but otherwise the step **S206** is bypassed. Similarly, if the optional use of fillers is desired (step **S208**) then fillers are added to the mixture (step **S210**), but otherwise step **S210** is bypassed.

[0053] In Step **S212** a first of two melt processing steps is used to form a masterbatch. The resulting masterbatch may have high concentration of pulp. In some embodiments, the resulting masterbatch from step **S212** may contain pulp that is in the range of about 50wt% to 99wt%.

[0054] This masterbatch from step **S212** is subsequently let down or diluted in step **S214**. The diluted masterbatch from step **S214** may contain pulp in the range of about 5wt% to 50wt%.

[0055] A second melt processing step **S216** is subsequently employed to obtain the desired cellulosic composite comprising pulp. The melt processing in step **S216** may include compounding, injection or extrusion.

[0056] Embodiments of the cellulosic composites described herein have broad utility in the automotive, building and construction, consumer and appliance markets. Non-limiting examples of potential uses of cellulosic composites of this disclosure include automotive components, decking, fencing, railing, roofing, siding, consumer utensils, containers and 3D printed components.

**[0057]** Articles produced by melt processing the cellulosic composites described herein can exhibit superior characteristics. For example, they may have improved mechanical properties and/or moisture resistance.

5

TABLE 1: MATERIALS

| Material                         | Supplier                                                                                                   |
|----------------------------------|------------------------------------------------------------------------------------------------------------|
| High density polyethylene (HDPE) | Ineos T5-440 119 HDPE, commercially available from Bamberger Polymers, Inc, Jericho, NY                    |
| Polypropylene (PP)               | Ineos H35G-00, Polypropylene Homopolymer, commercially available from Bamberger Polymers, Inc, Jericho, NY |
| CTMP                             | Chemi-Thermomechanical Pulp, commercially available from West Fraser Mills, Quesnel, BC, Canada            |
| Glass                            | StarStran 738, commercially available from Johns Manville Inc., Denver, CO                                 |
| Talc                             | Silverline 303, commercially available from Imerys, Inc, San Jose, CA                                      |
| Anti-oxidant (AO)                | Hostanox PEPQ, commercially available from Clariant Inc., Muttenz, Switzerland                             |
| Coupling Agent (CA)              | Integrate NP507030, commercially available from Lyondell Basell Inc., Houston, TX                          |
| Thrive™                          | 30% Cellulose filled PP, commercially available from Weyerhaeuser Inc., Federal Way, WA.                   |

TABLE 2: EXPERIMENTAL MASTERBATCH FORMULATIONS

| Sample | HDPE<br>wt% | PP<br>wt% | CTMP<br>wt% | AO<br>wt% |
|--------|-------------|-----------|-------------|-----------|
| MB1    | 5           | -         | 95          | -         |
| MB2    | 4           | -         | 95          | 1         |
| MB3    | -           | 5         | 95          | -         |
| MB4    | -           | 4         | 95          | 1         |

TABLE 3: EXPERIMENTAL COMPOSITE SAMPLE FORMULATIONS

| Sample | HDPE<br>wt% | PP<br>wt% | MB1<br>wt% | MB2<br>wt% | MB3<br>wt% | MB4<br>wt% | Talc<br>wt% | Glass<br>wt% | CA<br>wt% | Thrive™<br>wt% |
|--------|-------------|-----------|------------|------------|------------|------------|-------------|--------------|-----------|----------------|
| 1      | 66.7        | -         | 33.3       | -          | -          | -          | -           | -            | -         | -              |
| 2      | 66.7        | -         | -          | 33.3       | -          | -          | -           | -            | -         | -              |
| 3      | -           | 66.7      | -          | -          | 33.3       | -          | -           | -            | -         | -              |
| 4      | -           | 66.7      | -          | -          | -          | 33.3       | -           | -            | -         | -              |
| 5      | -           | 65.7      | -          | -          | 33.3       | -          | -           | -            | 1         | -              |
| 6      | -           | 65.7      | -          | -          | -          | 33.3       | -           | -            | 1         | -              |
| CE1    | 70          | -         | -          | -          | -          | -          | 30          | -            | -         | -              |
| CE2    | 70          | -         | -          | -          | -          | -          | -           | 30           | -         | -              |
| CE3    | -           | 70        | -          | -          | -          | -          | 30          | -            | -         | -              |
| CE4    | -           | 70        | -          | -          | -          | -          | -           | 30           | -         | -              |
| CE5    | -           | -         | -          | -          | -          | -          | -           | -            | -         | 100            |

- 5 [0058] Mechanical pulp, in this case CTMP, having a moisture content of approximately 50% and a lignin content of approximately 25% wt% based on dry weight, was first run through an electric 5" pellet mill such as are commercially available from Pellet Masters, Chippewa Falls, WI) to densify and pelletize the material. Four different masterbatch samples, MB1-4, were prepared with different compositions as shown in TABLE 2, each
- 10 having a high concentration of CTMP (95%). The masterbatch samples were prepared by

dry blending the pelletized moist CTMP with HDPE or PP, and in two cases an anti-oxidant additive, in a plastic bag, and then gravimetrically feeding the mixture into a 27 mm twin screw extruder (52:1 L:D, commercially available from Entek Extruders, Lebanon, OR). The compounding was performed using the following temperature profile in zones 1-13  
5 (°F): 100, 350, 400, 400, 400, 400, 400, 400, 400, 400, 400, 400, 400. The material was run though the extruder with the die removed and collected as a dry particulate.

[0059] Masterbatch samples, MB1-4, were subsequently letdown (diluted), by mixing with an additional quantity of the thermoplastic polymeric matrix (HDPE or PP), and in two cases a coupling agent, as shown in TABLE 3, to form six composite Samples 1-6. The  
10 components were dry blended in a plastic bag and gravimetrically fed into a 27 mm twin screw extruder (52:1 L:D, commercially available from Entek Extruders, Lebanon, OR). The compounding was performed using the following temperature profile in zones 1-13 (°F): 100, 350, 400, 400, 400, 400, 400, 400, 400, 400, 400, 400, 400 and a die temperature of 380 °F. The composites were extruded into strands and pelletized into pellets  
15 approximately 1-2 mm in length.

[0060] Samples CE1-4, having compositions as indicated in TABLE 3 were also similarly prepared as comparative examples. The talc and glass fiber were side fed downstream in zone 6. Sample CE5 was obtained from Weyerhaeuser Inc.

[0061] The resulting composite samples were injection molded into test specimens and  
20 their properties tested following ASTM D790 (flexural properties) and ASTM D638 (tensile properties). Specific Gravity was determined using Archimedes Method. Impact testing (Izod impact) was performed following ASTM D256. Moisture uptake was determined by gravimetric analysis after 24 and 96 hour submersion in water. The results of this testing are given in TABLE 4 below.

25

TABLE 4: EXPERIMENTAL RESULTS

| Sample | Flexural Modulus (kpsi) | Flexural Strength (kpsi) | Specific Gravity (g/cm <sup>3</sup> ) | Izod Impact Unnotched (ft-lbs/in) | Moisture Uptake 96 hr (%) |
|--------|-------------------------|--------------------------|---------------------------------------|-----------------------------------|---------------------------|
| 1      | 285                     | 4.9                      | 1.05                                  | 2.01                              | 2.4                       |
| 2      | 342                     | 5.4                      | 1.05                                  | 2.06                              | 1.3                       |
| 3      | 410                     | 8.0                      | 1.02                                  | 2.72                              | 1.2                       |
| 4      | 429                     | 8.1                      | 1.02                                  | 2.95                              | 0.8                       |
| 5      | 417                     | 10.5                     | 1.02                                  | 2.95                              | 0.8                       |
| 6      | 444                     | 11.0                     | 1.02                                  | 4.14                              | 0.8                       |
| CE1    | 222                     | 3.9                      | 1.16                                  | 4.13                              | 0.02                      |
| CE2    | 509                     | 4.7                      | 1.16                                  | 2.17                              | 0.01                      |
| CE3    | 389                     | 7.5                      | 1.12                                  | 3.56                              | 0.01                      |
| CE4    | 688                     | 8.8                      | 1.12                                  | 2.78                              | 0.01                      |
| CE5    | 422                     | 9.5                      | 1.02                                  | 4.9                               | 1.3                       |

[0062] Results for Samples CE1–CE5 are provided as comparative examples, and demonstrate properties for conventional glass- and talc-filled PP and HDPE composites.

5 CE5 demonstrates properties reported for Thrive™, which is a commercially available cellulose-filled polypropylene composite comprising chemical pulp from a kraft pulping process. Results for Samples 1 to 6 demonstrate properties of cellulosic mechanical pulp based composites according to certain embodiments of the present invention.

[0063] Some of the composite samples comprising mechanical pulp (CTMP) exhibited

10 comparable or superior properties relative to CE5 (Thrive™), with respect to flexural modulus and moisture uptake. Samples 5 and 6, comprising a polypropylene matrix and a coupling agent, performed particularly well. The coupling agent appeared to enhance the properties as may be observed by comparing results for Sample 3 with Sample 5, and Sample 4 with Sample 6.

[0064] Other lignin-containing materials, besides the mechanical pulps described herein, may be incorporated into a polymeric matrix to produce composites in a similar manner. For example, lignin-containing fibers from woody and non-woody sources, including grasses and recycled materials, for example, may be processed and incorporated into composites.

5 Lignin contents of non-woody materials and pulps can range upwards from 5% by weight.

[0065] Although specific embodiments have been illustrated and described herein, it will be appreciated by those of ordinary skill in the art that a wide variety of alternate embodiments or equivalent implementations may be substituted for the specific embodiments shown and described without departing from the scope of the present invention. This application is  
10 intended to cover adaptations or variations of the embodiments discussed herein. Therefore, it is manifestly intended that this invention be limited only by the claims and the equivalents thereof.

What is claimed is:

1. A cellulosic composite comprising wood pulp and polymeric matrix, wherein the pulp is substantially uniformly dispersed within the polymeric matrix.
- 5 2. The cellulosic composite of claim 1, wherein the wood pulp is one of mechanical pulp and chemical pulp.
3. The cellulosic composite of claim 2, wherein the wood pulp is said chemical pulp.
- 10 4. The cellulosic composite of claim 3, wherein said chemical pulp is kraft pulp obtained via a kraft process.
5. The cellulosic composite of claim 2, wherein the pulp is said mechanical pulp.
- 15 6. The cellulosic composite of claim 5, wherein the mechanical pulp comprises lignin, the lignin making up 5% to 35% of the dry weight of the pulp.
7. The cellulosic composite of claim 1, wherein the proportion of the pulp is in the range of 5% to 99% by weight of the composite.
- 20 8. The cellulosic composite of claim 7, wherein the proportion of the pulp is in the range of 10% to 95% by weight of the composite.
- 25 9. The cellulosic composite of claim 8, wherein the proportion of the pulp is in the range of 10% to 90% by weight of the composite.
10. The cellulosic composite of claim 7, wherein the proportion of the pulp is in the range of 80% to 99% by weight of the composite.

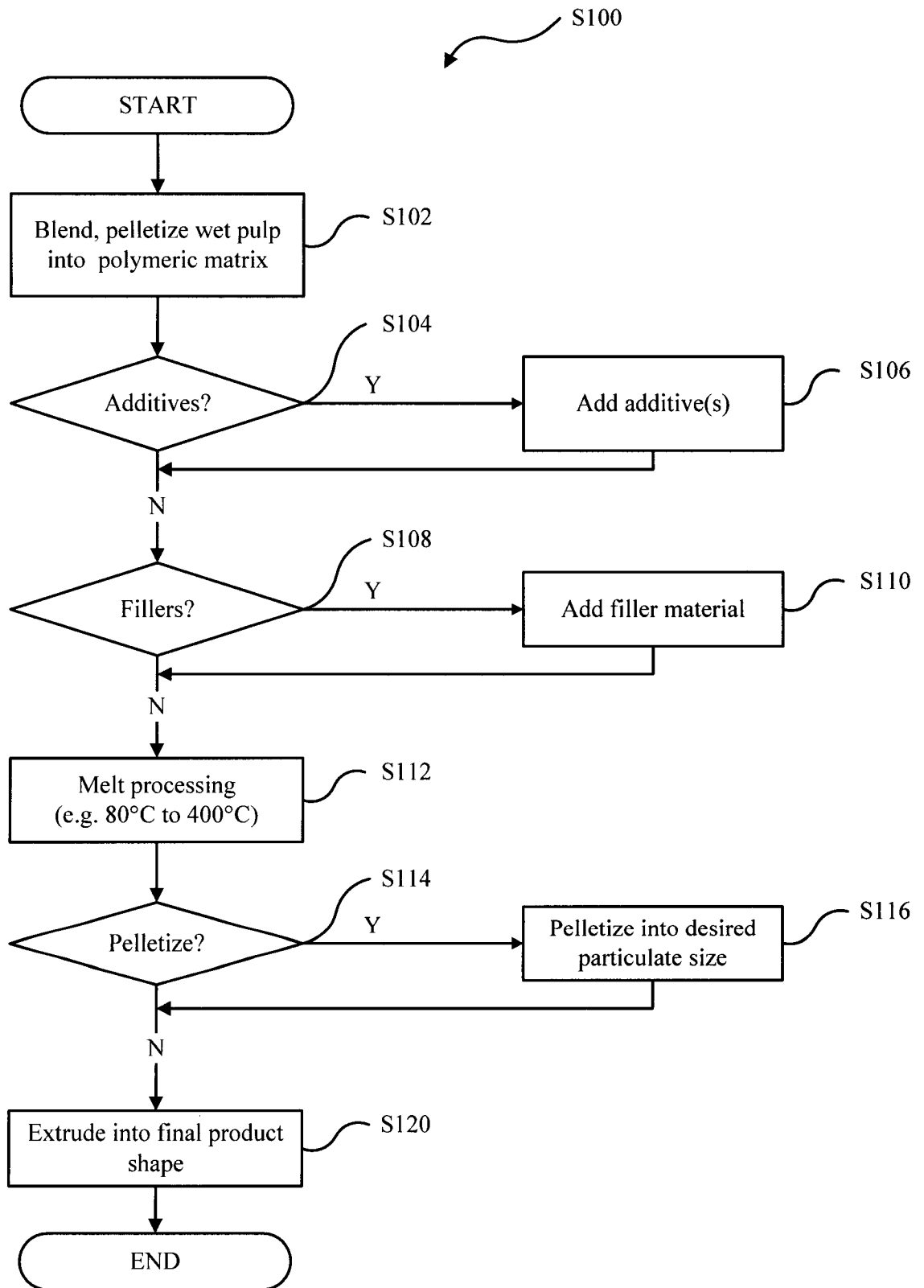
11. The cellulosic composite of claim 1, wherein the polymeric matrix comprises at least one of: high density polyethylene, low density polyethylene, linear low density polyethylene, functional polyolefin copolymers including polyolefin based ionomers, polypropylene, polyolefin copolymers, polystyrene, polystyrene copolymers, polyacrylates, polymethacrylates, polyesters, polyvinylchloride (PVC), fluoropolymers, polyamides, polyether imides, polyphenylene sulfides, polysulfones, polylactic acid (PLA), polyacetals, polycarbonates, polyphenylene oxides, polyurethanes, and thermoplastic elastomers.
12. The cellulosic composite of claim 1, wherein the polymeric matrix comprises bioplastics polymers comprising at least one of: biobased polyesters, biodegradable polyesters, compostable polyesters, polyamides, polyurethanes, polyacrylates, polyolefins, thermoplastic starches, cellulotics, PLA (Polylactic acid), PHA (Polyhydroxyalkanoates), PBAT (polybutyrate adipate terephthalate), PBS (polybutylene succinate), PCL (polycaprolactones), and PGA (Polyglycolic acid).
13. The cellulosic composite of claim 11, wherein the polyolefin copolymers comprise one of: ethylene-butene, ethylene-octene and ethylene vinyl alcohol; and wherein said polystyrene copolymers comprise one of: high impact polystyrene, and acrylonitrile butadiene styrene copolymer.
14. The cellulosic composite of claim 1, comprising an additive.
15. The cellulosic composite of claim 14, wherein the additive is at least one of: antioxidant, light stabilizer, fiber, blowing agent, foaming additive, antiblocking agent, heat stabilizer, impact modifier, biocide, antimicrobial additive,

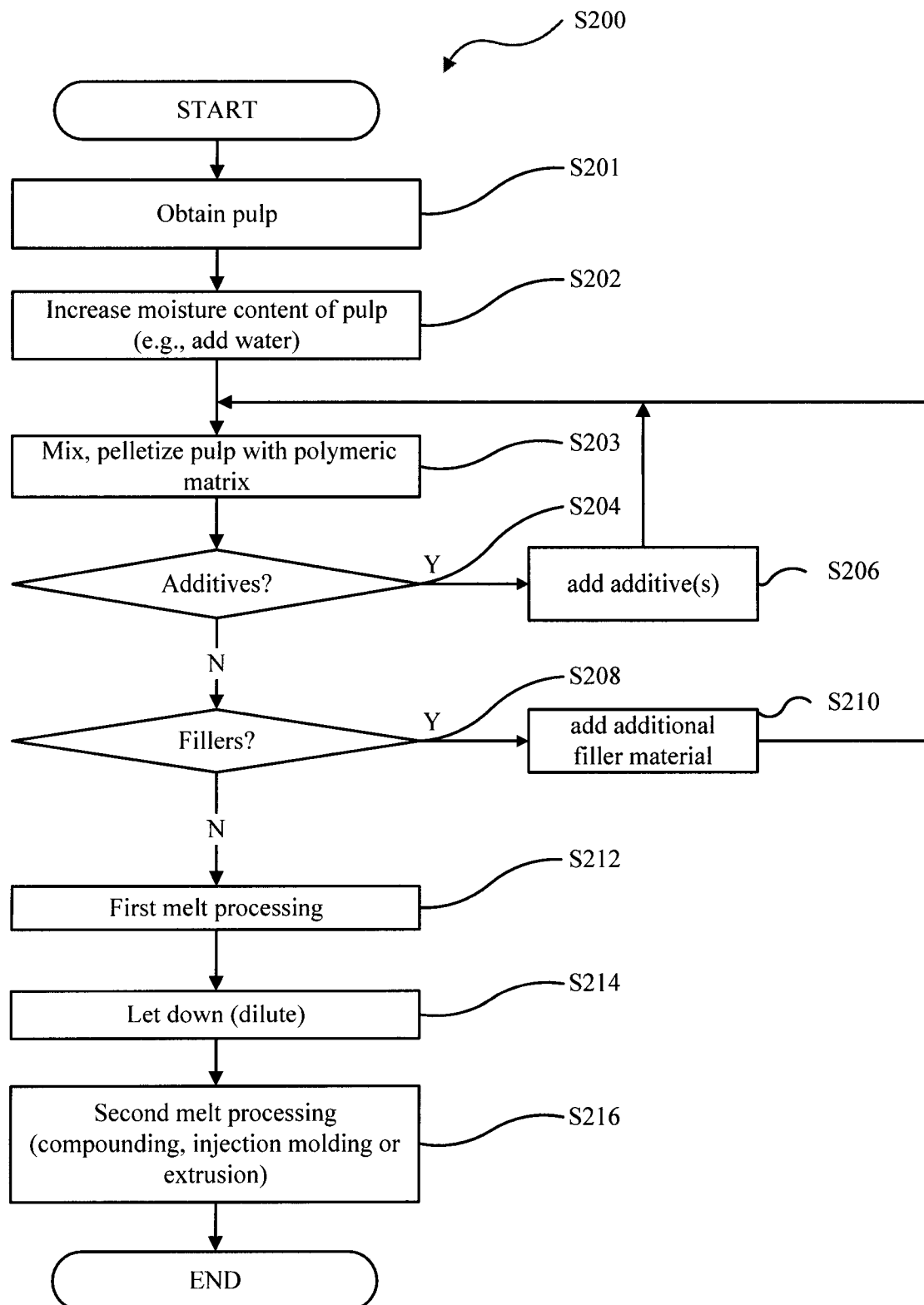
compatibilizer, plasticizer, tackifier, processing aid, lubricant, coupling agent, flame retardant, pigment and colorant.

- 5
16. The cellulosic composite of claim 14, wherein the additive is a coupling agent.
17. The cellulosic composite of claim 16, wherein the coupling agent is at least one of: silane, zirconate, titanate, functionalized polymer and maleic anhydride grafted polymer.
- 10 18. The cellulosic composite of claim 16, wherein the coupling agent is in the range of 0.1% to 10% by weight of the composite.
19. The cellulosic composite of claim 1, further comprising additional filler.
- 15 20. The cellulosic composite of claim 19, wherein said additional filler comprises at least one of: talc, mica, clay, silica, alumina, carbon fiber, carbon black, glass fiber, wood flour, wood fibers, non-wood plant fibers, sawdust, wood shavings, newsprint, paper, flax, hemp, wheat straw, rice hulls, kenaf, jute, sisal, peanut shells, soy hulls, lignin, calcium carbonate and cellulosic fiber.
- 20
21. The cellulosic composite of claim 19, wherein the additional filler is in the range of 1% to 90% by weight of the composite.
22. The cellulosic composite of claim 21, wherein the additional filler is in the range
- 25 23. The cellulosic composite according to any one of claim 1 to 22, wherein the composite has a moisture uptake of less than 1 wt % after 96 hours of immersion in water.

24. A cellulosic composite according to claim 23, wherein the polymeric composite forms at least a portion of an article selected from the group consisting of: an automotive component, decking, fencing, railing, roofing, siding, a consumer article, an appliance component and a 3D printed component.
25. A process for making a cellulosic composite, the process comprising: a first step of melt processing a mixture comprising wet pulp and a polymeric matrix.
26. The process of claim 25, wherein the moisture content said pulp is at least 10% by weight of the pulp.
27. The process of claim 26, wherein the moisture content said pulp is at least 20% by weight of the pulp.
28. The process of claim 27, wherein the moisture content said pulp is at least 30% by weight of the pulp.
29. The process of claim 25, further comprising a second step of melt processing a masterbatch formed from said first step, to form the composite.
30. The process of claim 29, further comprising diluting the masterbatch prior to said second step of melt processing.
31. The process of claim 29, wherein the mixture further comprises at least one of: an additive and a filler.
32. The process of claim 29, wherein each of the first and second steps of melt processing is one of: compounding, injection molding and extrusion.

33. The process of claim 29, wherein after the first step of melt processing the masterbatch contains pulp the range of 50% to 99% by weight and, after the second step of melt processing the composite contains pulp the range of 5% to 50% by weight.
- 5
34. The process of claim 29, wherein the polymeric matrix is a thermoplastic polymeric matrix.
35. The process of claim 29, further comprising increasing the moisture content of the pulp prior to the first step of melt processing.
- 10
36. A cellulosic composite produced according to the process of any one of claims 25 to 35.
- 15
37. A process for making an article of manufacture comprising:  
a) melt processing a mixture comprising wet pulp and a polymeric matrix; and  
b) extruding the melt processed mixture into the article.
- 20
38. The process of claim 37, further comprising pelletizing the melt processed mixture prior to said extruding.

**FIG. 1**

**FIG. 2**

## INTERNATIONAL SEARCH REPORT

International application No.

**PCT/CA2017/000071**

A. CLASSIFICATION OF SUBJECT MATTER  
 IPC: **C08L 97/02** (2006.01), **B29C 70/28** (2006.01), **C08L 101/00** (2006.01)

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)  
 IPC: **C08L 97/02** (2006.01), **B29C 70/28** (2006.01), **C08L 101/00** (2006.01)

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

Electronic database(s) consulted during the international search (name of database(s) and, where practicable, search terms used)  
 Questel (Fampat); CIPO Library Discovery Tool (sample keywords: pulp, wood pulp, chemical pulp, mechanical pulp, polymer matrix, wet pulp, bioplastic)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

| Category* | Citation of document, with indication, where appropriate, of the relevant passages                                                                                                                                                                                               | Relevant to claim No.               |
|-----------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-------------------------------------|
| X         | US 2012/0208933 A1 [HAMILTON, R. T. et al.] 16 August 2012 (16-08-2012)<br>(see paragraphs [0008], [0017], [0021], [0024] and [0042])                                                                                                                                            | 1-4 and 7-22                        |
| X         | López, J. P. et al. "Stone-ground wood pulp-reinforced polypropylene composites: water uptake and thermal properties" <i>Bioresources</i> <b>2012</b> , 7(4), 5478-5487. Published 01 November 2012 (01-11-2012).<br>(see entire document)                                       | 1, 2, 5-9, 11, 12, 14, 16-18 and 23 |
| X         | Du, Y. et al. "A simplified fabrication process for biofiber-reinforced polymer composites for automotive interior trim applications" <i>Journal of Materials Science</i> <b>2014</b> , 49, 2630-2639. Published online 19 December 2013 (19-12-2013).<br>(see entire document). | 1-4 and 24-38                       |

☒ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

|                                      |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                  |                          |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                          |
|--------------------------------------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|--------------------------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| *<br>"A"<br>"E"<br>"L"<br>"O"<br>"P" | Special categories of cited documents:<br>document defining the general state of the art which is not considered to be of particular relevance<br>earlier application or patent but published on or after the international filing date<br>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)<br>document referring to an oral disclosure, use, exhibition or other means<br>document published prior to the international filing date but later than the priority date claimed | "T"<br>"X"<br>"Y"<br>"&" | 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<br>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<br>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<br>document member of the same patent family |
|--------------------------------------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|--------------------------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|

Date of the actual completion of the international search  
 07 June 2017 (07-06-2017)

Date of mailing of the international search report  
 27 June 2017 (27-06-2017)

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## INTERNATIONAL SEARCH REPORT

International application No.

**PCT/CA2017/000071**

| C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT |                                                                                                   |                              |
|-------------------------------------------------------|---------------------------------------------------------------------------------------------------|------------------------------|
| Category*                                             | Citation of document, with indication, where appropriate, of the relevant passages                | Relevant to claim No.        |
| X<br>Y                                                | US 2013/0207302 A1 [CERNOHOUS, J. J. et al.] 15 August 2013 (15-08-2013)<br>(see entire document) | 1-4 and 7-22<br>29-33 and 38 |

**INTERNATIONAL SEARCH REPORT**

Information on patent family members

International application No.

**PCT/CA2017/000071**

| Patent Document<br>Cited in Search Report | Publication<br>Date         | Patent Family<br>Member(s)                                                                                                           | Publication<br>Date                                                                                                                                                                                                                                             |
|-------------------------------------------|-----------------------------|--------------------------------------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| US2012208933A1                            | 16 August 2012 (16-08-2012) | BR112013018613A2<br>CA2824137A1<br>CN103502533A<br>EP2675952A1<br>JP2014505158A<br>KR20130124358A<br>MX2013009371A<br>WO2012112398A1 | 18 October 2016 (18-10-2016)<br>23 August 2012 (23-08-2012)<br>08 January 2014 (08-01-2014)<br>25 December 2013 (25-12-2013)<br>27 February 2014 (27-02-2014)<br>13 November 2013 (13-11-2013)<br>26 September 2013 (26-09-2013)<br>23 August 2012 (23-08-2012) |
| US2013207302A1                            | 15 August 2013 (15-08-2013) | CN104334794A<br>EP2815020A1<br>JP2015511187A<br>MX2014009787A<br>WO2013122658A1                                                      | 04 February 2015 (04-02-2015)<br>24 December 2014 (24-12-2014)<br>16 April 2015 (16-04-2015)<br>08 September 2014 (08-09-2014)<br>22 August 2013 (22-08-2013)                                                                                                   |