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(72) **Inventeur/Inventor:**
SAIN, MOHINI M., CA

(73) **Propriétaire/Owner:**
SAIN, MOHINI M., CA

(74) **Agent:** NA

(54) **Titre : DISPERSION EN PHASE SOLIDE ET TRAITEMENT DE MICRO ET NANO-FIBRES CELLULOSIQUES EN PHASE PLASTIQUE POUR LA FABRICATION DE PRODUITS BIO-NANOCOMPOSITES D'INTERET COMMERCIAL**

(54) **Title: SOLID PHASE DISPERSION AND PROCESSING OF MICRO-AND NANO-CELLULOSIC FIBRES IN PLASTIC PHASE TO MANUFACTURE BIO-NANOCOMPOSITE PRODUCTS OF COMMERCIAL INTEREST**

(57) **Abrégé/Abstract:**

Plant stem are bundles of cellulose nanofibers with a diameter ranging between 10 to 70 nm and lengths of thousands of nanometers. The mechanical performance of the cellulose nanofibers is comparable to other engineering materials such as glass fibers, carbon fibers etc. In this invention a novel process has been developed to disperse cellulosic microfibrils and nanofibers in solid phase into a molten plastic matrix or in a low viscosity water-based resin system in aqueous phase.

Invention relates to manufacturing Bio-nanocomposites of high performance by dispersing micro-and nano-cellulosic fibres into plastic matrix using conventional plastic processing equipment. This invention also overcomes one unique challenge of bio-nanocomposite processing, which is to make them commercially viable by surface modifying cellulosic micro-and nano-fibres derived from renewable resources.



Patent Application: 2,549,844**“Solid Phase Dispersion and Processing of Micro and Nano-Cellulose Fibres in Plastic Phase to Manufacture Bio-Nanocomposite Products of Commercial Interest”**

APPLICANT: Mohni M. Sain

ABSTRACT

Plant stem are bundles of cellulose nanofibers with a diameter ranging between 10 to 70 nm and lengths of thousands of nanometers. The mechanical performance of the cellulose nanofibers is comparable to other engineering materials such as glass fibers, carbon fibers etc. In this invention a novel process has been developed to disperse cellulosic microfibrils and nanofibrils in solid phase into a molten plastic matrix or in a low viscosity water-based resin system in aqueous phase.

Invention relates to manufacturing Bio-nanocomposites of high performance by dispersing micro-and nano-cellulosic fibres into plastic matrix using conventional plastic processing equipment. This invention also overcomes one unique challenge of bio-nanocomposite processing, which is to make them commercially viable by surface modifying cellulosic micro-and nano-fibrils derived from renewable resources.

DESCRIPTION

Background of the Invention

Cellulose micro- and nano-fibres are entangled in solid phase and are hard to obtain in individualized form due to strong hydrogen bonding. In aqueous phase and in dilute solution they can be obtained in highly dispersed form. Therefore, one possible method could be dispersing cellulose micro-and nano-fibres in aqueous or in a phase containing right combination of hydrophilic-hydrophobic character, typically a water-alcohol mixture phase and then solubilizing a polymer that has film-forming property as well as are soluble, either in an aqueous phase or in a mixture of water and other solvent in which, those micro-and nano-fibres can be dispersed.

Summary of the Invention

In this invention cellulose microfibrils or nanofibrils isolated from agro-residues^{1,2} such as wheat straw, soy straw, bagasse, wood pulp, recycled fibre, root fibres are dispersed in water or in a mixture of water and alcohol, in a concentration range 0.5 to 5% and a soluble polymer in the given mixture has been added. Mixture were stirred with or without temperature to dissolve the polymer completely and then casted into film by an efficient drying process. Films, thus produced are cut into small chips and added in a twin screw extruder to further disperse them in plastics in molten phase by introducing solid thermoplastic polymer in the extruder along with the dispersed film particles.

The uniqueness of the invention is as follows:

This process avoid handling of liquid or water during conventional nanocomposite processing by extrusion, compression, injection and other molding processes by introducing a masterbatch of pre-dispersed nanofibre in polymer of high or low molecular weight subsequently taking out excessive liquid by filtration, drying or casting.

The second unique aspect of the invention is to pre-disperse cellulosic micro- and nano-fibres into a polymer in solution phase that are having surface energies compatible

to base polymers in which they are further dispersed during their solid-phase dispersion. For example, acrylic and maleic derivatives of ethylene or vinyl backbone are good for their dispersion in polyethylene.

Extruded products are obtained in different shapes and sizes including profiles, sheets, rods, solid or hollow. Extrusion also includes pellet form of the mixed composition and then further using them for compression molding, blow molding, injection molding, thermoforming, rotomolding. This method of liquid phase casting followed by solid phase mixing provided a unique method to manufacture bio-nanocomposites.

This process of manufacturing bio-nanocomposite in solid phase is unique because it solves the micro-and/or nanofibre dispersion problem in solid or molten phase compounding of plastics with those fibres.

Typical examples of plastic phase could be any thermoplastic materials including those obtained from forest, petroleum and agro-derived sources. Typically, polyvinyl alcohol, polyolefins, vinyl polymers and copolymers, engineered plastics such as styrene-acrylonitrile copolymer (SAN), nylon, polyester, ABS/PC, bio-based plastics such as thermoplastic starch, polylactic acid (PLA), poly-hydroxy-alkonates (PHAs).

Typical examples are cited below:

Example 1:

Polyethylene glycol-maleic anhydride treatments increased cellulose content of soybean stock nanofibers from 40% to 60% when they are first filter deposited or casted into films and then these dispersed nanofibre films were introduced in molten PVA during extrusion, injection or high shear mixing. Nanofibers reinforced PVA films demonstrated at least a 4-5-fold increase in tensile strength, as compared to the untreated fiber/PVA film. In solid phase nanocomposites, improved mechanical properties were achieved with coated nanofibers. Typically, polylactic acid (PLA) having initial tensile strength 52 MPa was increased to about 65 MPa by introducing 3-4% of soybean stalk derived nanofibres.

Example 2:

In yet another example, hemp nanofibre dispersed in ethylene acrylate oligomer by casting process has been dried, chopped into flake and granules for easy introduction in high shear mixer, injection machine and extrusion, were melt-mixed with polyethylene and maleated polyethylene as an interface modifier improved tensile strength of the resulting nanocomposite by 50% and improved modulus by three folds.

Typical examples of plastic phase could be any thermoplastic materials including those obtained from forest, petroleum and agro derived. Typically, polyvinyl alcohol, polyolefins, vinyl polymers and copolymers, engineered plastics such as styrene-acrylonitrile copolymer (SAN), nylon, polyester, ABS/PC, bio-based plastics such as thermoplastic starch, polylactic acid (PLA), poly-hydroxy-alkonates (PHAs).

Example 3:

In yet another process these micro-and/or nanofibres from natural origin are dispersed in surface active oligomers or polymers (styrene copolymers and their derivatives, polyethylene glycol –maleic anhydride , styrene-maleimide , ethylene-acrylic copolymers) in an aqueous or in a solvent phase in which these surface active agents polymers or oligomers are soluble. The liquid phase dispersed micro-and/or nano fibrils are then incorporated in a liquid thermoplastic or thermosetting resins including bio-resins (such as polyesters, vinyl esters, epoxy or polyol-isocyanate resins or in which one or all components of these resins are made from crop sources such as soy bean, canola, corn, sunflower, palm, peanut or any other oil or their derived chemical precursors), thereby partly or totally eliminating issues related to agglomeration of those micro-and/or nanofibres in the liquid resins during their solidification process.

CLAIMS

Claim 1: A process to deposit films from a liquid phase of the cellulosic micro and nanofibre obtained from wood, root and nonwood sources, wherein said cellulosic micro and/or nanofiber is dispersed uniformly in a plastic which is soluble in the above liquid phase, drying the micro-and /or nanofibre film followed by cutting or shaping the micro-and/or nanofibre dispersed plastic film into small sized flakes, granules or pellets and then further incorporating this micro-and/or nanofibre dispersed plastic in a molten thermoplastic by using extrusion, high shear mixing or injection compression process, thereby giving a novel process to obtain a bio-nanocomposite product without any risk of agglomeration of micro-and /or nanofibers in the plastic matrix during their melt mixing process.

Claim 2. A process as cited in claim 1, whereby the liquid phase is selected from materials obtained from forest, petroleum and agro-based resources representing group of thermoplastics of polyvinyl alcohol, polyolefins, vinyl polymers and copolymers, engineered plastics selected from styrene-acrylonitrile copolymer (SAN), nylon, polyester, ABS/PC, and bio-based plastics comprising thermoplastic starch, polylactic acid (PLA), and poly-hydroxy-alkonates (PHAs).

Claim 3. A process as cited in claim 1, wherein the resulting PVA bio-nanocomposite reinforced with soybean stock nanofibers has demonstrated at least 4-5 fold increase in tensile strength compared to untreated fibres, whereas PLA nanocomposites experienced 25% enhancement in tensile strength.

Claim 4. A process as cited in Claim 1 wherein the micro and nano-fibres are dispersed in surface active oligomers or polymers.

Claim 5. A process according to Claim 4, characterized in that the surface active oligomers or polymers are from the group of styrene copolymers and their derivatives, polyethylene glycol –maleic anhydride, styrene-maleimide, and ethylene-acrylic copolymers in soluble liquid phase.

Claim 6. A process as cited in Claim 1 wherein the liquid phase dispersed micro and/or nano fibrils are incorporated in liquid thermoplastic or thermosetting polymers.

Claim 7. A process according to Claim 6, characterized in that thermoplastic or thermosetting polymers belong to unsaturated polyesters, vinyl esters, epoxy or polyol-isocyanate resins and in which at least one of the components of these resins are obtained from renewable crop sources of soy bean, canola, corn, sunflower, palm, peanut or any other oil and their derived chemical precursors.