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WOOD FIBRE REINFORCED PLA AND PLA/TPS BIOCOMPOSITES: PROCESSING, FORMULATION AND MECHANICAL PROPERTIES

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Introduction

Public awareness about the environment, increasing crude oil prices, and new regulations on the use of nonrenewable resources to generate materials as well as the need for reduced weight components are the main drivers to develop bio-based composites. Different additives, impact modifiers and reinforcing agents have been used in biobased polymers to improve their mechanical performance. Few studies have been reported on the use of wood flour and plant fibres (flax, kenaf and hemp) as reinforcing phase in biopolymers (PLA, PHB). The introduction of natural fibres into PLA improves the tensile and flexural properties of the biocomposites. The tensile strength and stiffness of PLA flax fibre composites was proved to be superior to those of PP flax fibre composites [1]. It was shown that the addition of triacetin, maleic anhydride grafted PLA and cellulose acetate could act as coupling agents to improve the adhesion between the fibres and the polymer [2, 3]. This paper discusses the processing and the properties of biocomposites composed of a biopolymer reinforced with wood fibres obtained from a thermo-mechanical refining process. In this study, polylactide (PLA) and polylactide/thermoplastic starch blends (PLA/TPS) were used as thermoplastic matrices. Two wood fibre types, two grades of PLA, and three TPS concentrations were selected in order to investigate their effects on the final properties of the composites. The effect of different additives on the biocomposite properties was also studied for the purpose of enhancing the biopolymer-wood fibre affinity to reach the best mechanical performance. The biocomposites were obtained by co-rotating twin-screw extrusion and injection moulding. The properties are described in terms of morphology and mechanical properties.

Experimental Part

Two different grades of PLA from NatureWorks were used in this study: PLA8302D is amorphous (aPLA) and PLA2002D is semicrystalline (cPLA). Details concerning the thermoplastic starch blends and the grafting of maleic anhydride onto PLA can be found in previous work [4]. The wood fibres, extracted from yellow birch (HW) and black spruce (SW), were prepared by FPInnovations in Quebec from wood chips via a thermo-mechanical refining process usually used to produce medium density fibreboard (MDF). The triacetin (TA) was provided by MAT Laboratories and the cellulose acetate (CA) in the form of powder was provided by Scientific Polymer Products Inc. The chain extender (CE), CesaExtend OMAN698493, was obtained from Clariant and used to preserve PLA molecular weight processing. A special pelletizing step of the wood fibers was added prior to composite processing in order to facilitate the feeding of the natural fibres upon extrusion. A Buhler 20 mm co-rotating twin-screw extruder was used to process the composites. The polymer and pelletized wood fibres were fed simultaneously in the first zone. The extrusion temperature was between 170 - 180°C. The extruder, equipped with a 2 mm diameter capillary die, was operated at a constant screw speed of 150 rpm for a flow rate of 2 kg/h. The pellets of polymers and fibres were dried for 12 hours at 80°C before extrusion. In all composites, the wood fibre concentration was maintained at 30 % by weight.

Scanning electron microscopy (SEM) was carried out on samples coated with a gold/palladium alloy before observation. The extruded composite pellets were dried at 55°C for 24 hours and then injection moulded using a 30-ton Boy injection press with a screw temperature of about 190°C and a mould temperature around 30°C. The tensile testing was performed according to ASTM D638 on type I specimens. The samples were conditioned at 50°C under vacuum for 40 h before mechanical testing. At least five specimens were tested for each formulation.

Results and Discussion

Wood fibres used in this work were received in a wet state with a moisture content of 50 wt %. This high water content was needed to facilitate the fibre pelletizing operation in which the wet wood fibres were pressed through a die plate by rotating roll-mills. There was no thermal degradation on the fibres after the pelletizing and the fibre integrity was preserved. The screw configuration used for the extrusion experiments was designed to obtain a very good dispersion of the wood fibres into the polymer melt as verified by SEM observation.

The tensile strength at break and elastic modulus of cPLA and aPLA based composites reinforced with SW are presented in Figure 1. The results obtained on the HW biocomposites, not shown here, are very similar to the tensile properties obtained for the SW biocomposites. The materials that contained CE performed very well. The ultimate tensile strength values are increased by 11% compared to the non-compatibilized biocomposites and more than 20% compared to the virgin PLA materials. This improvement is primarily the result of the branching effect of the chain extender on the matrix. The tensile modulus showed an increment of nearly 100% in all cases compared to the pure PLA matrix. When CA and TA were used as compatibilizers, a high decrease in tensile strength was observed, contrarily to the results described in the literature [2, 3].

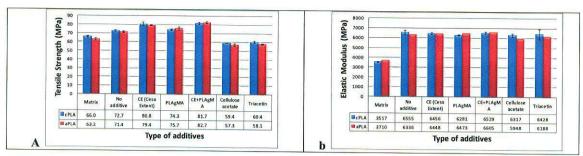


Figure 1. a) Tensile strength and b) elastic modulus of PLA/SW composites for different additive formulations.

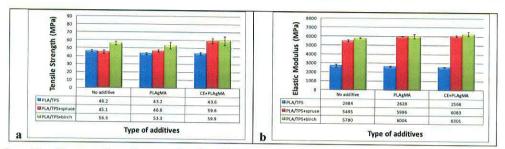


Figure 2. Tensile properties of composites based on aPLA/50%TPS blend with different additive formulations:

a) tensile strength at break and b) elastic modulus.

Regarding the PLA/TPS blends, the elastic modulus as measured on the biocomposites increased more than two times compared to the matrix (i.e. 6300 MPa vs. 2600 MPa), which is similar to the PLA-based biocomposites (Figure 2b). The tensile strength is also increased by 31% in comparison with the corresponding matrix.

Fracture surfaces were also analyzed and the SEM micrographs are presented in Figure 3. The addition of CE or CE/PLA-g-MA into the biocomposites led to a high attenuation of the fibre pull out phenomenon. The related composites displayed almost no sign of fibre pull out while a limited amount of pull out was visible for the PLA-g-MA formulations. The same microstructural observation, i.e. the disappearing of the pull out when CE was used, was taken in the case of aPLA/50 wt % TPS based biocomposites (results no shown here).

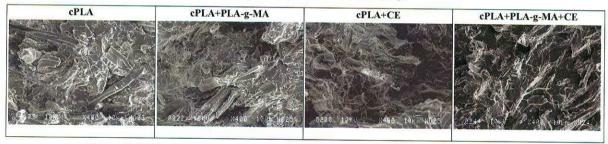


Figure 3. SEM microstructural details of fracture surfaces of the cPLA-HW composites

Conclusions

This work aimed at developing and characterizing novel biocomposites based on PLA or PLA/TPS blend reinforced with wood fibres by varying the matrix properties and wood fibre nature as well as using additives for their coupling capability. The ability of the studied wood fibres to reinforce PLA or PLA/TPS matrices has been assessed. A twofold increase in elastic modulus and an increase in tensile strength of 14% were reached for the non-compatibilized PLA biocomposites. In the presence of reactive additives, the best results were achieved for composites that contained the chain extender, and a combination of coupling agent/chain extender. PLA/wood fibre biocomposites may present higher mechanical properties than PP counterpart. They are very promising candidates for various packaging applications and for interior ones in construction industry.

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