Evaluation of Mechanical Properties and Durability Performance of HDPE-Wood Composites

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ABSTRACT: This article evaluates the mechanical properties and biodegradability of wood-plastic composite materials made from sawdust and thermoplastic polymer (HDPE). For the preparation of the composites, sawdust in different proportions with Maleic Anhydride grafted Polyethylene (MAPE) as the coupling agent was used. The mechanical properties and biodegradability of the biocomposites were successively characterized. The results indicate that adding sawdust particles to a polymer matrix improves the mechanical strength and stiffness of composites. The tensile strength of a composite with 3% coupling agent was improved by 13%, 34% and 54% respectively when 20%, 30% and 40% wood fillers were added to the polymer matrix. Furthermore, resistance to fungi attacks decreased. The biodegradability of biocomposites up to 97 days indicates that, by increasing the sawdust content, the amount of weight loss increased as well. Also, it has been found that the microbial effects were more important in the case of composite with high wood-filler content and without MAPE. The maximum weight loss was estimated to be equal to 1.67%, corresponding to the high wood content composite of HDP-40% wood without MAPE. In other words, even though the addition of sawdust to polymer improves the mechanical performance of a composite material, it also accelerates the biodegradation rate. An optimum amount of sawdust content might compromise the effect of the biodegradation and mechanical properties of composite materials.

KEYWORDS: Wood plastic composites (WPCs), mechanical properties, biodegradability

1 INTRODUCTION

The mechanical properties of wood-plastic composite (WPC) materials are highly influenced by the polymeric matrix/sawdust particles interface. However, incompatibility between the sawdust particles (hydrophilic) and hydrophobic polymer matrix (e.g., HDPE) leads to poor adhesion between the two components; resulting in ineffective mechanical performance [1,2]. Many studies have been performed to address this issue, most of them concentrating on the surface modification of sawdust particles or use of coupling agents for bonding improvement (hydrogen bonds with the hydroxyl groups of sawdust particles) [3]. The most important component of wood is cellulose, which is a crystalline structure and rigid part. Its modulus of rigidity is about 136 GPa [4]. Addition of the wood particles leads to a significant increase of the polymer stiffness. Lignin as an amorphous polymer does not greatly contribute to the mechanical properties of wood particles, but plays an important role in binding the cellulose fibrils that allows efficient stress transfer to the cellulose molecules. However, these composites possess a high number of hydroxyl groups available to react with water molecules in a humid environment [5]. It is well known from the literature, that moisture content greatly affects the tensile strength, swelling and porosity of wood particles [6]. Certain authors have reported that the water absorption for biocomposites increases the functionality of the wood particles content, and is typically up to 18–22% after several months [7]. Accordingly, an additional amount of wood particles increases the water uptake, which can worsen the mechanical strength of biocomposites.

In addition, wood plastic composites are highly sensitive to attack by microorganisms attack, especially when used in outdoor applications. As mentioned in the literature, incorporation of sawdust particles to a polymer matrix appears to influence the durability properties as well as the biodegradation rate of composite materials [8]. The degradation rate of wood

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differs from one species to another. Such differences are difficult to explain from the chemical composition of the species, as some fungi decompose cellulose or hemicelluloses more rapidly than lignin, while others decompose lignin more efficiently than polysaccharides [9,10]. Toxic content, density of wood species, size of wood particles, and the degree of adhesion between polymer and wood particles are also positive factors in preventing the rapid rate of biodegradation [11]. Based on the literature, the addition of sawdust particles to the polymer matrix improves the mechanical strength of composite materials but also increases their sensitivity to environmental issues. Natural filler-reinforced composites face adverse effects when exposed to moisture. It causes a decrease in mechanical properties, provides the necessary condition for biodegradation, and changes their dimensions [5]. Decrease in mechanical properties is caused mainly by degradation of the particles–matrix interface, which results in poor stress transfer at the interface. In this study [12], the authors reported that the tensile and flexural strength of polyester hybrid composites based on sisal and roselle tested under wet conditions is a decreasing function of the reinforcement's content.

There is an important need to expand the research on natural fillers-polymer composites with regards to their biodegradability and mechanical performance under different environmental conditions. The originality of this study is the evaluation of the durability of composite properties through mechanical measures and biodegradability tests. The effects of the sawdust content and coupling agent addition on the mechanical characteristics and biodegradability of HDPE-wood composites were studied. Based on the evaluation conducted, it was concluded that an optimum blend ratio of wood particles and HDPE polymer must be employed to achieve a balance between strength and durability requirements for natural fillers composites.

2 MATERIALS AND METHODS

2.1 Materials

The sawdust used in this study was supplied by Tembec sawmill located in Béarn (QC, Canada). It consists of 65% white spruce (*Picea glauca*), 20% black spruce (*Picea marica*) and 15% balsam fir (*Abies balsamea*). It is a softwood type composed essentially of cellulose: 42–44%; hemicellulose: 27–28%; lignin: 24–28%; and some minor constituents (extractives: 3–4%). The sawdust density measured with a helium pycnometer (Accupic model 1330) is equal to 1.48 g/cm^3. The particle size analysis of the sawdust particles was performed on more than 5000 particles using a Fiber Quality Analyzer (FQA). The average geometry and length distribution of sawdust particles were shown respectively in this article [13]. The sawdust was sieved to a diameter of less than 700 microns (Ø< 0.7 millimeters). The polymeric matrix is high-density polyethylene (*Sclair A59*), with a melting temperature of 138°C and density of 980 kg/m^3. The coupling agent is *Fusabond 226DE* from Dupont, which is a polyethylene grafted with maleic anhydride (melting temperature 120°C, melt flow index (MFI) 1.5 g/10 min, at 190°C, 2.16 kg). The amount of coupling agent used is 3% of the total weight of the composite, an amount which is known to improve the homogeneity of the mixture, ensuring better distribution of particles in the polymer matrix [1,14]. The sieved sawdust was previously dried at 105°C for 24 h and stored in plastic bags. The residual moisture content in wood particles used for samples preparation is estimated by a second drying of 120°C for 24 h. The wood humidity amount calculated from the mass of sawdust before and after drying was found to be less than 3 wt%.

2.2 Methods

The composite samples were prepared using a co-rotating twin screw extruder (HAAKE) with an angular speed of 50 rpm at a temperature of 170°C, which is lower in fact than the sawdust particles degradation temperature (200°C). In the first step, similarly sized wood particles were compounded into pellets at 20 wt%, 30 wt%, 40 wt%, 50 wt% and 60 wt% by weight of HDPE (Fig. 1a). In the next step, flat disc samples for the biodegradation test were prepared by compression molding using a hot press set at 150°C (above the melting temperature of the polymer, 138°C) with aluminum mold plates (Fig. 1c). In order to ensure the mixture was softened properly, a preload was first applied for 2 min. This preload is also beneficial for removing any air that has possibly accumulated in the material. Loading on the material was then continued up to 8 MPa for 4 min to give the final shape. The dog-bone shaped samples prepared by injection molding with a HAAKE Minijet from Thermo Scientific are shown in Figure 1b.

2.2.1 Mechanical Properties Characterization

The tensile behavior of WPCs material was analyzed using a Zwick machine (TA Instruments, USA) maximum load 100 kN. Tests were conducted in accordance with ASTM standard D.638. The measurements were performed at room temperature (~23°C) at a strain rate of 5 mm-min^{-1}. The composite samples were dog-bone shaped and their dimensions were analogue to

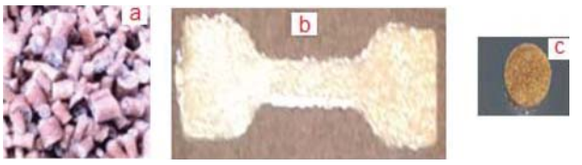

Figure 1 (a) Pellets, (b) dog-bone shaped sample from injection molding, and (c) compression molded samples.

test specimen of the Type I [15]. The results were normalized from six different measurements.

2.2.2 *Biodegradability Tests*

The biodegradability tests were performed according to European Standard EN113. *Gloephyllyllum trabeum* fungal species, which are responsible for the degradation of softwood in nature, were used in this research. Three disk specimens for each composite formulation (25 mm diameter and 4 mm thickness) were prepared. In addition, for a better understanding of wood-polymer composite degradation, three neat HDPE samples were prepared as the control samples. In order to observe the possible biodegradation of the samples, they were placed on a dish containing malt and microorganism for 97 days. To do so, a cultural substrate for fungi species growth was prepared. A Petri extract agar and a fungal species were incubated at ambient temperature for one week (until the malt extract agar was covered by mycelium). The composite samples were sterilized by exposure to UV rays before they were put in contact with the microorganisms. Sterilized test blocks (shown in Figure 2) were then incubated in a controlled environment (humidity 70%, and temperature 30°C) for 97days. Each composite sample was placed in the Petri dish (15 dishes in all) and, in order to avoid any contamination, only malt extract agar was used (Figure 2). After the incubation period, the samples were taken out and cleaned to remove any adhering mycelium. The samples were oven-dried at 105°C for 48 h and then weighed. Changes between the initial and final weight of the samples were interpreted as a measurement of biodegradation by fungal attack.

3. RESULTS AND DISCUSSION

3.1 Tensile Tests

Figure 3 illustrates the effects of the sawdust content and the coupling agent, respectively, on the tensile properties, elastic modulus E(MPa) and strength modulus (MPa) of the composites. In all the samples, the elastic modulus increases with the increase in the

Figure 2 Composite samples exposed to microbial attack (left) and negative samples (right).

wood particles content. However, this increase was more predominant in samples with a coupling agent, which was in fact expected because of the higher modulus of wood particles. For example, Woodhams and coauthors estimated that Young's modulus of wood fibers varies between 10 and 80 GPa [16], and Buttery suggested 4.9 to 14 GPa for wood flour [17]. The moduli of filled composites are higher than that of neat polymer. The additions of a coupling agent improve the compatibility between the wood filler and HDPE through esterification and thus enhance the impregnation of wood particles by polymer, resulting in much more rigid composites.

A similar result was reported in the literature of Godard and coauthors [14] who have demonstrated that the tensile modulus of HDPE polymer reinforced by fine wood particles in presence of polyethylene maleic anhydride as coupling agent, was an increasing function of particles content.

By contrast, as mentioned in several studies, the mechanical strength of biocomposites without a coupling agent will be decreased by adding sawdust because of stress concentration at the edge of the particles [18,14]. Addition of a coupling agent makes for a significant improvement in the mechanical properties of composite materials because of a better adhesion and improved stress transfer between sawdust particles and matrix. As a result, the tensile strength will be increased by the addition of wood particles [19].

In comparison to neat HDPE, the tensile strength of a composite with 3% coupling agent was improved by 13%, 34% and 54% respectively when 20%, 30% and 40% wood fillers were added to the polymer matrix (Figure 3). These results were comparable to those mentioned in the literature. In this study [20], the authors reported that the tensile strength of the HDPE matrix reinforced by treated wood fiber composites increased by 35% showed that the MAPE treatment and the matrix-resin pre-impregnation of the fiber produced a significant increase in tensile strength.

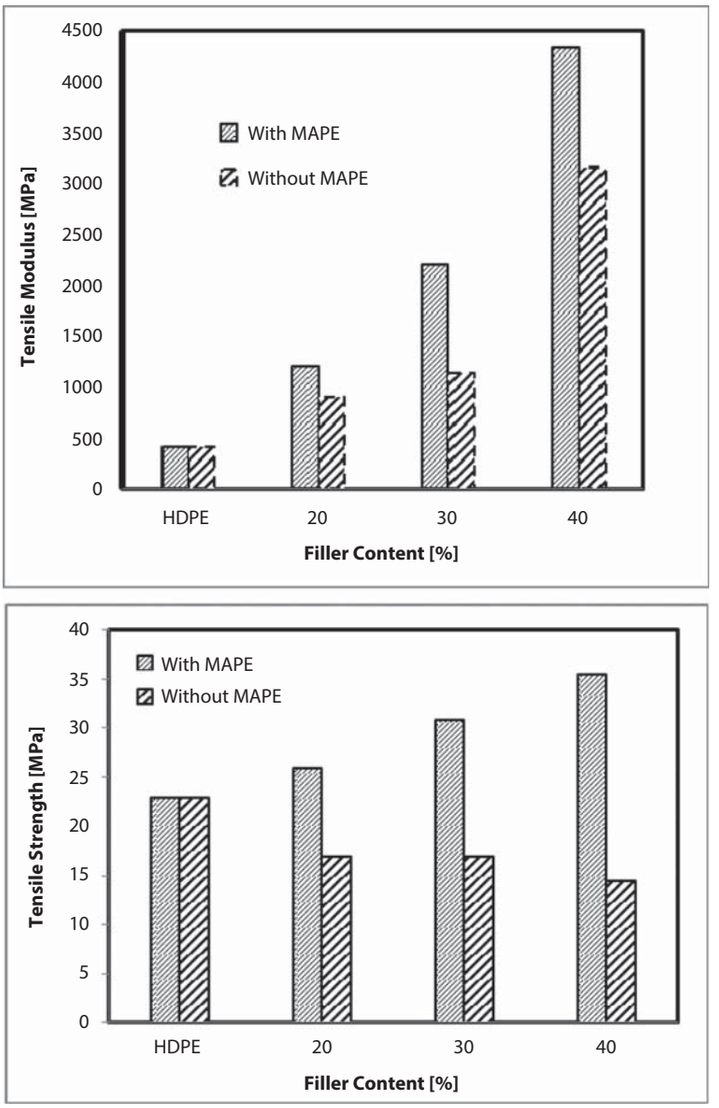

Figure 3 Effect of MAPE in mechanical properties of biocomposites with different filler contents.

Increasing the sawdust content leads to a considerable increase in tensile strength, which can be attributed to the intrinsic adhesion of the particles-matrix interface caused by the chemical modification of the coupling agent (MAPE). As the wood content increases, more particles are available per unit cross-section area of the composite; hence, the stress at break increases [6].

3.2 Biodegradability Tests

The effect of fungi species *G. trabeum* on wood degradation as a function of the wood content and coupling agent was investigated. The fungal attack was more obvious in the case of composite with high wood fillers content and without a coupling agent. The presence of coupling agent greatly reduces the weight loss for the composite material by improving the compatibility between the wood filler and HDPE through esterification. This enhances the impregnation of wood particles by polymer, resulting in much more resistant composites. Indeed, weigh lost was estimated 45 times less for composite made with 40 wt% wood filler when 3 wt% of coupling agent has been added to mixture. Figure 4 shows the weight loss of composites with different concentrations of wood (0 wt%, 20 wt%, 30 wt% and 40 wt%) for up to 97 days. Based on this research, adding MAPE as the coupling agent significantly improves the decay resistance of composites.

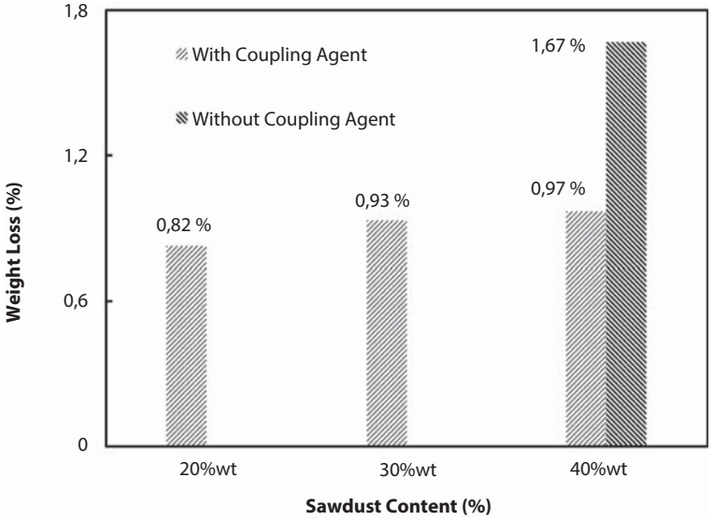

Figure 4 Results of biological degradation of wood plastic composites.

While there was no microorganism effect on the neat matrix, an attack by microorganisms on wood fillers would be the only reason for this weight loss. This could be attributed to the moisture effect, which can accelerate the biodegradation mechanism. The sawdust particles are sensitive to water absorption, so the uptake of more water in the composite material results in more dimension changes in the polymer matrix, leading to cracking and fissuring of the polymer surface. Thus, fungal species can reach the wood fillers through such discontinuities. Some authors have reported that scanning electron microscopy of wood-plastic composite specimens confirms the growth of fungal mycelium, which is concentrated in discontinuities between the wood fillers and thermoplastic component near the specimen surface [21].

Composites with a high content of wood fillers and without a coupling agent are more susceptible to absorb a large amount of water, which makes them more degradable, resulting in a higher weight loss when exposed to fungi decay. It should be noted that this degradation is promoted by a high content of wood filler and, also, by the weakness of the chemical interaction between wood particles and polymer chains. Meanwhile, a high content of HDPE and use of a coupling agent minimize a susceptibility to weight loss during the accelerated decay test. Investigation of the structural alteration and change in the surface chemistry of a composite interface can lead to a better understanding of the degradation mechanism. Therefore, rheology could be an efficient tool to assess the morphology and interfacial status change of filled polymers exposed to divers conditions [13].

4 CONCLUSION

We studied the mechanical properties of biocomposites made from high-density polyethylene reinforced with different levels of sawdust in the presence of a coupling agent. The biodegradability of biocomposites up to 97 days was also investigated. Our results indicate that the addition of sawdust improves the mechanical performance of biocomposites. It allows a considerable increase in both modulus and tensile strength, although the composite becomes more vulnerable to moisture uptake and susceptible to microorganism attacks. An optimized amount of fiber content could alleviate the composite degradation process as a result of the decomposition of wood fillers.

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