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Technical Report

Development of novel green and biocomposite materials: Tensile and flexural properties and damage analysis using acoustic emission

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ABSTRACT

A new green composite made of natural polyethylene (NPE) has never been produced using short birch fibers and compared with others biocomposites with matrices of linear low-density polyethylene (LLDPE) and high-density polyethylene (HDPE). Versions with and without a coupling agent (CA) in fiber ratios of 10, 20, 30 and 40 wt% were produced. Tensile and 3-point flexural tests were conducted to measure the mechanical properties of the composites, and acoustic-emission testing was used to measure the evolution of damage caused by irreversible changes in the materials in correlation with an analysis of the damage modes. It was concluded that the extent of the damage and the contribution of each damage mode depend on the material, the test performed and, especially the presence of a CA. The results prove that the choice of composite for a particular application must be a judicious one and should consider not only the mechanical properties but also the damage processes of the composite, which may be crucial for long-term applications.

1. Introduction

It is often a difficult task, therefore, to reconcile the long-term utilization of a composite with minimal environmental impact at the end of the product's life cycle. Nevertheless, in recent years, we have seen an increase in government awareness and public pressure that has resulted in the use of more eco-friendly and sustainable materials.

The term 'biocomposite' is employed when natural fibers are used as an environmentally friendly alternative to composites with traditional fibers. The advantages of natural fibers include certain notable properties, such as low density, high specific strength, enhanced energy recovery, CO_2 neutrality after burning, easy processing, bio-degradability and low cost [1,2]. However, these biocomposites still depend on oil to some extent and can cause waste problems because of the use of non-degradable polymer matrices.

Among other ecological solutions, a noteworthy class of composites with structurally sound properties has been developed: polyethylene (PE) with birch fiber. PE is the most affordable thermoplastic with the highest share of production of any polymer type, representing 29.1% of the world's plastic production [3]. PE is obtained through the polymerization of ethylene (C_2H_4), which produces macromolecules consisting of a repeating monomer unit (CH_2-CH_2). In addition, the most widely available natural filler in province of Québec is birch fiber. Because this hardwood tree grows in cool areas with abundant precipitation, the region contains approximately 50% of the growing stock volume of yellow birch in North America [4]. The combination of these two materials results in an eco-solution in the form of a biocomposite that is simple to manufacture and, consequently, should be very affordable.

The most affordable variant of PE is linear low-density polyethylene (LLDPE), which has the highest ductility and the poorest Young modulus and mechanical strength at room temperature among all semi-crystalline polymers [5]. Studies of the mechanical properties of LLDPE biocomposites with aspen fibers have been conducted by Gu and Raj et al. [6,7]. In a recent work, Mijiyawa et al. [8,9] studied the thermal degradation of LLDPE/birch but did not analyze the resulting mechanical damage. More recently, Lafia-Araga et al. [10] has tested red balau fibers using various thermal treatments at the fiber level and has observed an increase in the matrix/fiber adhesion that improved the mechanical properties of the material. It is now acknowledged that the use of a coupling agent (CA) is essential to improve the overall quality of biocomposites with such constituents [11–13].

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The properties of PE are determined largely by the arrangement of the polymer chain. Properties such as the crystalline melting point, the density, the hardness and the permeability are determined by the type, number and distribution of short-chain branches, whereas the viscosity is affected primarily by long-chain branches [14,15]. High-density polyethylene (HDPE) is a linear polymer with no short branching but and a small fraction of long branches [16]. HDPE thus possesses improved mechanical properties for a small price increase. Some biocomposites have been studied using HDPE as the matrix. Migneault et al. [17] have studied the properties of the polymeric matrix impregnated with white birch fibers under various loads using a rheometer. They concluded that not only were the mechanical properties affected by the fiber weight, but the melting properties were affected as well. Raj and Kokta [18] have arrived at similar conclusions using aspen fibers. The use of a CA is also strongly indicated. Lu et al. [19] have provided evidence that of the various CAs they tested, maleated polyethylene (MAPE) was the best option. In addition, Colom et al. [20] have demonstrated the 'bridging' effect of MAPE using scanning electron microscopy (SEM) images. Adhikarya et al. [21] have investigated the differences in stability, mechanical properties and microstructure between recycled and virgin HDPE using fibers of Pinus radiata. The results were very encouraging from an ecological perspective because they demonstrated that regardless of whether virgin or recycled HDPE was used, the mechanical properties of the biocomposite were the same.

Nevertheless, there are two aspects in which the literature lacks data regarding these composites: (1) the improvement of the green nature of composites with natural fiber content below 50 wt% (i.e., with one or more thermoplastics as the major constituent of the matrix) and (2) a thorough analysis of the progression of mechanical damage mechanisms within the composites caused by the application of external stresses in various scenarios.

To address point 1, green composites can be created using a biosourced matrix and natural fibers [22]. Among the green matrix materials that are available, polylactides (PLAs), polyhydroxyalkanoates (PHAs) and bio polyvinyl chlorides (PVCs) are of particular interest [23]. These materials bear no similarity to PE; however, in 2010, Braskem (America's top producer of thermoplastic [24]) developed a "natural (green) polyethylene" (NPE) sourced entirely from sugarcane, which was a technological breakthrough. We were therefore able to fabricate a green family of composites using this material for comparison with composites based on PE. To produce the biopolymer, Braskem converts sugarcane-derived ethanol into ethylene using a process known as ethanol dehydration. This process converts 99% of the ethanol carbon into polyethylene [25]. Contrary to the petroleum-based PE process, the sugarcane CO_2 remains fixed during the complete life cycle.

According to Braskem, green PE presents ecological advantages over the life cycle assessment (LCA) when compared to the conventional PE: this polymer captures 2.5 tonnes of CO_2 per ton of product [26]. Today, several end users are using NPE (e.g.: Johnson & Johnson, Nestle, Toyota, Danone, P&G) [26]. To our knowledge, a green composite based on NPE has never before been produced using short birch fibers.

The advantages of increasing the fiber ratio in a NPE composite are twofold: (1) Price benefit for using a widely available material (in addition, these fiber can obtained from waste) and (2) The enlargement of application range due to the increase in the mechanical properties. Furthermore, there is no record of the effect of the use of a CA on the material behavior of such green composites.

To address point 2, tensile and 3-point flexural tests can be conducted to measure the mechanical properties and the mechanisms and evolution of the damage caused by irreversible changes in the material under study. The acoustic-emission (AE) technique is generally defined as elastic energy spontaneously released during local, dynamic and irreversible changes of the (micro)structure of the materials [27]. AE is also observed during phase transformations and plastic deformations [27]. With this definition is important to note that damage (irreversible change) due to stress can occur without plasticity. In this case, AE becomes most useful when not only the progression but the nature of the mechanism that originated each sound wave can be tracked, specially dealing with materials where the damage process is not well understood. AE testing can be used to characterize the evolution and competition of the various damage modes in a material.

The damage model associated with the mechanical processes proposed in this study is based on the model introduced by Kachanov [28], which assumes that the damage to a material can be interpreted in terms of the density of defects in that material. Mehan and Mullin [29] was the first researcher to correlate a damage mechanism with an acoustic signature (AS). Damage-mechanism analysis has traditionally been performed using simple investigative histograms of cumulative hits versus amplitude [30,31]. However, this methodology can be inaccurate for complex materials [32], especially biological materials (i.e., cellular structures assembled through a hierarchical process in nature) using thermoplastics (with strong damping properties) [33,34]. In this case, a more complex analysis is recommended, specifically, the use of fuzzy logic systems [35–37], which have the ability to detect clusters among data even when the boundaries between groups overlap [38]. The use of three known parameters for damage-mode identification in the PE/birch composite family is proposed [34]: the burst amplitude, the counts and the duration. Three families of composites using natural birch fibers and matrices of LLDPE, HDPE and NPE were developed. For each matrix, we produced versions with and without a CA in fiber ratios of 10, 20, 30 and 40 wt%. Thus, 54 varieties of composites totaling 270 specimens were produced in this comprehensive study.

This paper is organized as follows. First, the materials, methodology and procedures of experimental testing are described. Second, the mechanical behavior and properties of the various composites as determined from the tensile and flexural testing are discussed and compared. Third, an AE damage analysis is presented based on the results of tensile and flexural testing and the results are discussed. Finally, the previous results are correlated with SEM images before presenting the final conclusions.

2. Materials and experimental testing

2.1. Materials

Industrial short fibers (thermomechanical pulping, 35 mesh size) of yellow birch (Betula alleghaniensis) were used in this experiment. The fibers were produced by the Lignocellulosic Materials Research Centre, Trois-Rivières (Canada), and dried at 60 °C in an air-circulating oven for 24 h before use.

The three thermoplastic matrices that were used were LLDPE (Novacor® HI-0753-H), donated by NOVA Chemicals; HDPE (Sclair® 2909), donated by NOVA Chemicals; and NPE (version HDPE-SHA7260), donated by Braskem. MAPE (maleated polyethylene, G2010), supplied by Eastman Chemical Company (Kingsport, TN, USA), was used as the CA. The content of maleic acid grafts was 1.5% with a molecular weight of 15,000. The CA chemical composition leads to the formation of chemical bridges between the natural fibers and the PE matrix. The use of CA in quantities beyond 4 wt% can lead to self-entanglement among CA chains rather than with the polymer matrix [39]. For this reason, CA was used at 3 wt%.

All specimens were prepared using a two-roll mill (Thermon C.W. Brabender, Model T-303) with a 0.6 gear ratio. The grains of

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