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Effect of hybridization on the physical and mechanical properties of high density polyethylene–(pine/agave) composites

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ABSTRACT

This work reports on the properties of high density polyethylene based hybrid composites made with two natural fibers: agave and pine. The composites were produced by a combination of extrusion and injection molding. The effect of hybridization was analyzed via morphological, mechanical and water immersion tests for two total fiber contents, 20 and 30 wt.%, and different pine-agave fiber ratios (100–0, 80–20, 60–40, 40–60 and 0–100). Moreover, the effect of coupling agent (maleated polyethylene) in the hybrid composite formulation was evaluated. The results showed that addition of agave fibers improves tensile, flexural and impact strength, while pine fibers decreases water uptake. As expected, the addition of a coupling agent improves substantially the quality of the polymer–fiber interface as well as the mechanical properties, but this effect was more important for composites produced with higher agave fibers content due to the their chemical composition.

1. Introduction

In recent years ecological awareness and other environmental issues led to the development of composite materials based on renewable resources such as natural fibers. These materials are environmentally friendly and low-cost alternatives to replace synthetic fibers like glass and carbon fibers [1]. Natural fibers are used as reinforcements for composite materials due to various advantages compared to conventional fibers. The main advantages are lower densities, non-toxicity and lower cost. In addition several fiber varieties are locally available [2]. However, due to hydrophobic–hydrophilic interactions the combination of natural fibers with most polymer matrices results in poor adhesion and therefore limited stress transfer from the matrix to the reinforcing fibers [3]. For that reason, several investigations explored the possibility of using coupling agents to improve adhesion and consequently properties such as tensile and flexural strength and modulus [4,5]. Lu et al. [6] investigated the effect of maleated polyethylene (MAPE) in wood fiber/high density polyethylene composites and observed that for untreated composites the wood fibers were mainly linked to the thermoplastic matrix through mechanical connections, while evidence of chemical bridges via esterification were present in maleated wood fiber/HDPE composites.

Hybrid composites are materials made by combining two or more different types of reinforcements in a common matrix. Hybridization may offset the disadvantages of one component by the addition of another. A requisite for the occurrence of a hybrid effect is that both reinforcements differ in mechanical properties and their interaction with the matrix. The strength of hybrid composites is dependent on the properties of each reinforcement such as aspect ratio, content, geometry, orientation, intermingling extent and interfacial bonding [7]. For example, Haq et al. [8] mentioned that hybridization enables to exploit the synergy between natural fibers and inorganic compounds, leading to properties improvement while maintaining environmental appeal.

One advantage of hybridization is cost reduction; some reinforcements are very expensive and could be combined with less expensive materials maintaining good properties. For example, with the addition of natural fibers, good mechanical properties can be obtained at a lower cost. Ramesh et al. [9] prepared hybrid composites of sisal–jute–glass fibers and found that the incorporation of sisal–jute fibers can improve tensile, flexural and impact strength and used as an alternate material for glass fiber reinforced

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polymer composites. Arrakhiz et al. [10] elaborated PP/pine cone fiber/clay composites and found that the addition of clay to PP/pine composites improved the tensile properties.

Hybrid composites made from waste materials are also a great option to develop new materials with low costs and specific properties. Ashori [11] prepared hybrid composites made with newspaper fibers and poplar wood flour. They observed that the addition of both fibers enhanced the tensile and flexural modulus compare with neat polypropylene, but increasing wood flour reduced flexural and tensile moduli. Fernandes et al. [12] prepared composites from high density polyethylene (HDPE) filled with cork powder and coconut short fibers. The addition of coconut fiber to cork–HDPE composites increased the elastic modulus and tensile strength by 27% and 47% respectively, compared to cork–HDPE composites. Boopalan et al. [13] studied the effect of banana fiber addition to jute–epoxy composites and obtained increased mechanical properties with lower moisture absorption.

In this study, the effect of agave fiber addition to HDPE/pine composites was investigated in terms of mechanical properties and water absorption. Both fibers used here are actually local industrial wastes as agave fibers are residues of tequila processing and pine fibers are obtained from timber industry. Overall, two total fiber contents and six different fiber combinations were used to analyze the effects of these parameters.

2. Materials and methods

High density polyethylene 60120U (PADMEX, Mexico) with a melt flow index of 19 g/10 min (190 °C/2.16 kg) and a density of 0.96 g/cm^3 was used as the matrix. As a coupling agent, maleated polyethylene Fusabond M603 (MAPE) was provided by Dupont Packaging and Industrial Polymers (USA). Pine sawdust was supplied by Aserraderos Gomez Farias (Mexico) and agave fibers (*Agave tequilana* Weber, var. Azul) was obtained from a local tequila company in Jalisco (Mexico). The chemical composition of the fibers presented in Table 1 was determined according to TAPPI standards T-204cm-97 (extractives), T-222-om-98 (lignin) and the Jayme–Wise method (holocellulose).

2.1. Composites preparation

The agave fibers received a previous treatment before being used. The fibers were soaked in water for 24 h and then passed through a Sprout-Waldron refiner (D2A509NH) with two 30 cm diameter discs, one fixed and the other rotating at 1770 rpm to separate the pith from the fibers. The fibers were then placed in a centrifuge to remove excess water and finally dried outdoors. Pine fibers did not receive any previous treatment and were used as received from the sawmill. Both agave fibers and pine sawdust were milled and sieved to keep particles between 50 and 70 mesh. Two total fiber contents (20% and 30%) were used in the formulations with different pine–agave fiber ratios (100–0, 80–20, 60–40, 40–60 and 0–100). All the compositions were prepared with and without 3% of MAPE with respect to the total fiber content. The composites were processed in a twin-screw extruder Leistritz Micro 27 GL/GG 32D with a temperature profile set to 140/150/150/160/160/160/170/170/160 °C. The composites were cooled in a water bath and then pelletized. The composite pellets were oven-dried for 24 h at 65 °C and then injection molded on a NISSEI ES 1000 with a mold temperature of 40 °C and a screw temperature profile of 130/170/185/195 °C. All the samples were prepared in a rectangular mold cavity with dimensions of 80 × 40 × 2.5 mm^3.

Table 1
Chemical composition of pine and agave fibers.

Fiber	Extractives (wt.%)	Lignins	Cellulose	Hemicellulose
Agave	6–7	21–24	58–65	10–13
Pine	18–20	27–30	40–45	12–15

2.2. Morphology

Samples were submerged in liquid nitrogen and then fractured. Micrographs of the exposed surfaces were obtained by a scanning electron microscope JOEL JSM 840A to characterize the morphology, mainly the state of adhesion/dispersion of the fibers in the matrix.

2.3. Water absorption test

Water absorption tests were conducted in accordance to ASTM: D570 at room temperature. Before testing, the weight of each specimen was measured. Then, five specimens were submerged in distilled water at 30 ± 0.5 °C for 160 days. The samples were removed after specific time periods to get water absorption kinetics curves. Each time, the surface water was wiped off with a dry cloth and the sample was immediately weighed before being returned in the water bath. The amount of water absorption (M_t) was calculated as follows:

$$M_t = \frac{w_i - w_0}{w_0} \times 100 \quad (1)$$

where w_i is the weight of the sample after immersion and w_0 is the initial weight before water immersion.

In this case, water absorption in the samples could be considered as Fickian behavior (i.e. following Fick's law) and the next equation can be used [14]:

$$\frac{M_t}{M_\infty} = 4\left(\frac{D_x t}{\pi h^2}\right)^{1/2} \quad (2)$$

where M_t is the moisture content at a specific time (t), M_∞ is the equilibrium moisture content, D_x is the water diffusion coefficient and h is the thickness of the sample.

2.4. Density

Density was obtained by a gas pycnometer ULTRAPYC 1200e from Quantachrome Instruments, using nitrogen as the gas phase. The data reported are the average of three measurements.

2.5. Mechanical properties

Impact strength of the composites was determined by an impact tester Tinius Olsen model 104. The specimens were prepared according to ASTM: D6110 and each value obtained represents the average of 10 notched samples. Flexural and tensile properties were evaluated using a universal machine Instron model 4411 before and after water absorption to determine the effect of water ageing. Flexural tests were carried out according to ASTM: D790 and the sample dimensions were 80 × 13 × 2.5 mm^3 with a span length of 41 mm. Six samples per composition were tested at a cross-head speed of 10 mm/min. Tensile tests were carried out using a load cell capacity of 1 kN. The specimens were tested according to ASTM: D638 using type V specimens. Testing was performed at a cross-head speed of 5 mm/min with seven samples per composition. All the tests were performed at room temperature (23 °C).