

Material properties

Materials prepared from biopolyethylene and curaua fibers: Composites from biomass

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ABSTRACT

Composites of high-density biopolyethylene (HDBPE) obtained from ethylene derived from sugarcane ethanol and curaua fibers were formed by first mixing in an internal mixer followed by thermopressing. Additionally, hydroxyl-terminated polybutadiene (LHPB), which is usually used as an impact modifier, was mainly used in this study as a compatibilizer agent. The fibers, HDBPE and LHPB were also compounded using an inter-meshing twin-screw extruder and, subsequently, injection molded. The presence of the curaua fibers enhanced some of the properties of the HDBPE, such as its flexural strength and storage modulus. SEM images showed that the addition of LHPB improved the adhesion of the fiber/matrix at the interface, which increased the impact strength of the composite. The higher shear experienced during processing probably led to a more homogeneous distribution of fibers, making the composite that was prepared through extruder/injection molding more resistant to impact than the composite processed by the internal mixer/thermopressing.

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1. Introduction

The development and marketing of composite materials produced from natural sources (e.g., biocomposites) are considered extremely important, as these materials will reduce dependence on materials produced using non-renewable resources [1–4] and are preferable for environmental and economic reasons [5–8]. In recent years, many studies have been carried out with a goal of developing relatively low-cost composite materials with good performance and low environmental impact [9–12].

Synthetic fibers are widely used as reinforcement material in plastics [13]; for instance, glass fibers [14,15]. The use of lignocellulosic fibers is particularly attractive

because their use is environmentally safe and is a solution to the petroleum supply problem [8,16–19].

In recent years, composites that are reinforced with various natural fibers, such as jute [20,21], coconut, sugarcane [22–24], cotton [25], bamboo, sisal [26] and curaua [24,27], have been developed.

Natural fibers are excellent reinforcing agents for plastics as they are resistant, lightweight and non-abrasive. Because of their low cost and the diversity of the sources, the applications for natural fibers are economically attractive [28–31]. Curaua fibers, as well as other lignocellulosic fibers, are composed of cellulose, hemicellulose (or polyoses) and lignin [23,32,33]. Cellulose is comprised of a linear chain of linked cellobiose (Fig. 1a), and hemicelluloses are copolymers of different monosaccharides (Fig. 1b). The structure of lignin is made up of three basic units in different proportions (Fig. 1c).

The hydrophilic characteristics of lignocellulosic fibers, which are primarily a consequence of hydroxyl groups

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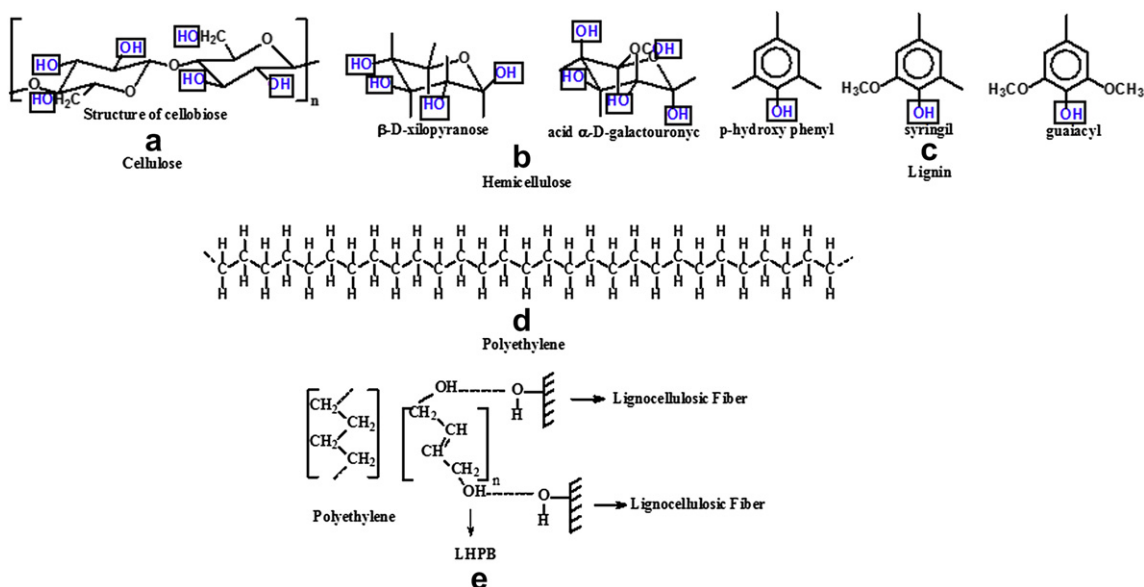


Fig. 1. Structure of cellobiose (a), units present in hemicellulose (b), lignin (c) (Fengel, 1984), and chain of polyethylene (d); schematic representation of the hydrophilic (LHPE-fiber) and hydrophobic (LHPE-polyethylene) interactions between the components of the composites (e).

(Fig. 1a–c), can pose a problem in the preparation of composites that they reinforce if the matrix has hydrophobic features. Examples of this are polyolefins, such as polyethylene (Fig. 1d). The treatment of fibers, or the use of additives during the preparation of composites, can improve the mechanical properties of the materials [34–37]. Several studies have reported the use of polyethylene that has been grafted with maleic anhydride (PEMA) in an attempt to increase the affinity of the polymer/fiber, thereby improving the performance of polymer composites [38–42].

The introduction of a small amount of rubber, such as liquid hydroxylated polybutadiene (LHPB), which is used in this study, can increase the fracture resistance of polymeric composites, although it may cause a decrease in the flexural modulus [43,44]. In the present work, the intention was to introduce a new application for LHPB because, in this study, it can act not only as an impact modifier but also as a compatibilizer. LHPB with hydrocarbon chains having affinity for polyethylene can be used as a matrix, and has hydroxyl groups with an affinity for the polar groups of the lignocellulosic fibers used for reinforcement, as shown in Fig. 1e.

The polymer that was used as the matrix in this study, hereafter called biopolyethylene or HDBPE (high density biopolyethylene), was prepared industrially from ethylene generated from ethanol, which comes from the process of fermentation and distillation of sugarcane juice. Using materials produced in this manner reduces the dependence on raw materials of fossil origin for the manufacture of plastic products¹* (“greener” alternatives) [45].

Curaua, which is obtained from a plant that grows in the Amazon region [16], was used in the preparation of composites based on HDBPE. This fiber was chosen due to its high cellulose content (as described later) because the

fiber strength usually increases with increasing cellulose content [46].

A variety of mixing equipment, such as intensive mixers and single or double screw extruders [47–51], can be used for preparing cellulosic thermoplastic composites. To compare the influence of different processing techniques on the properties of the composites in the present study, composites were prepared using two different processes: compounding the mixture using an internal mixer followed by thermopress molding, and compounding the mixture using an intermeshing twin-screw extruder followed by injection molding.

2. Experimental

2.1. Materials

The HDBPE, melt flow index of 0.10 ± 0.01 g (10 min⁻¹), as determined by a Plastometer MI-3 following ASTM D1238 (temperature of 190 °C, load of 2.16 kg), was kindly provided by Braskem (Triunfo, Rio Grande do Sul, Brazil), and the curaua fibers were purchased from Pematec-Triangel of Brazil Ltda (Sao Paulo, SP, Brazil). The LHPB, trade name Liquiflex H, was kindly donated by Petroflex Ind. Com (Rio de Janeiro, RJ, Brazil).

2.2. Fiber characterization

The fibers were characterized as described elsewhere [16,34], and the respective procedures are provided here. For Klason lignin, holocellulose and α -cellulose contents an average for each was calculated from repeat measurements on three different samples.

The Klason lignin content was determined by following the TAPPI T13M-54 method, which is based on the isolation of lignin following polysaccharide hydrolysis using concentrated sulfuric acid (72%).

¹ *(available at <<http://www.braskem.com.br>>).

The holocellulose content was determined according to TAPPI T19m-54, which consists of selective degradation of lignin by sodium hypochlorite.

To determine the α -cellulose content, a sodium hydroxide solution (17.5%) was added to the cellulose (1.0 g) at room temperature. The mixture was then further ground with sodium hydroxide (17.5%) for 8 min, and subsequently allowed to rest for 20 min. After resting, water (40 mL) was added to the mixture, and the solid residue was filtered and washed with aqueous acetic acid and water. The remaining solid, namely α -cellulose, was dried at 105 °C until a constant weight was obtained.

The hemicellulose content was obtained by subtracting the percentage of α -cellulose from the holocellulose content.

2.3. Preparation of HDBPE/LHPB/curaua fiber composites

- a) *Internal mixer/thermopress molding*: For this procedure, mixtures of differing amounts of curaua fibers with an average length of 1 cm and differing amounts of LHPB were added to HDBPE and mixed in a Haake torque rheometer equipped with a RHEOMIX 3000 P mixing chamber, capable of 45 g, at 180 °C and 60 rpm for a period of 6 min. Then, 40 g of the HDBPE/LHPB/curaua fiber mix was placed between two sheets of Teflon, which were then placed between two sheets of stainless steel, on which an aluminum frame was placed to ensure the desired end thickness. The mixture was then thermopressed at 180 °C for 3 min. The assembly was kept under pressure (force of 2 ton) and then cooled to room temperature by the circulation of water through the system. The same procedure was employed to prepare control samples (HDBPE/fiber and HDBPE/LHPB) [8] and to prepare a composite with 3 mm length fibers for comparison with the samples prepared as described in (b).
- b) *Inter-meshing twin-screw extrude/injection molding*: The materials were prepared from the molten polymer using a MT19TC co-rotating twin screw extruder from B&P Process Equipment and Systems (diameter 19.0 mm; L/D 25, screw speed 80 rpm, profile temperature 160 °C/170 °C/180 °C/180 °C/190 °C). The screw that was used has two kneading zones, as shown in Fig. 2. Then, the HDBPE/5%LHPB/10%Fiber (the fibers had an average length of 3 mm) composite was prepared via injection molding using an Arburg Allrounder model 270V injection molding machine. The specimens were injected and molded according to the ASTM D790 for flexural testing and ASTM D256 for Izod impact. During this process, the material first passes through a heating zone (205 °C/210 °C/215 °C/215 °C/215 °C) at 100 bar and is then

injected (injection speed of 20 cm³ s⁻¹ and a back pressure of 25 bar) into a mold that was pre-conditioned at 50 °C. The whole process took 48 s.

2.4. Characterization of materials

2.4.1. Scanning electron microscopy (SEM)

SEM was performed using a Zeiss DSM 940. In the case of the composites, it was necessary to use fractured samples to analyze the interfacial adhesion. The samples were covered with a thin layer of gold using a sputter-coat system prior to analysis.

2.4.2. Izod impact strength

The impact strength measurements were performed according to ASTM Standard D256 using Izod-type notched samples (Izod Impact Tester, CEAST Resil 25, room temperature, impact speed of 4 m s⁻¹).

2.4.3. Flexural tests

Flexural tests were performed using a universal testing machine (EMIC DL2000) with a cross head speed of 5 mm min⁻¹, according to ASTM Standard D790-03.

2.4.4. Dynamic mechanical thermoanalysis (DMTA)

Dynamic mechanical thermoanalysis (DMTA) was performed using a TA Instruments Model 2980 thermal analyzer operating in flexural mode with an oscillation amplitude of 20 μ m, frequency of 1 Hz and heating rate of 2 °C min⁻¹ over a temperature range of -130 to 105 °C.

2.4.5. Thermogravimetry (TG)

TG was performed using a ShimadzuTG 50, according to NBR 14692. Approximately 5 mg of the samples were placed in appropriate pans and heated from 20 to 800 °C at a rate of 10 °C min⁻¹ under an atmosphere of N₂ at a flow rate of 10 mL min⁻¹.

2.4.6. Differential scanning calorimetry (DSC)

DSC measurements were performed using a ShimadzuTA-50WSI at a heating rate of 10 °C min⁻¹ under an atmosphere of N₂ at a flow rate of 20 mL min⁻¹ over a temperature range of 25 to 200 °C.

3. Results and discussion

The α -cellulose (63.4%) and hemicellulose (29.6%) contents found in this study are higher, whereas the lignin (5.2%) content was lower, than those of previous studies. Trindade et al. obtained 83.5% holocellulose (73.6% α -

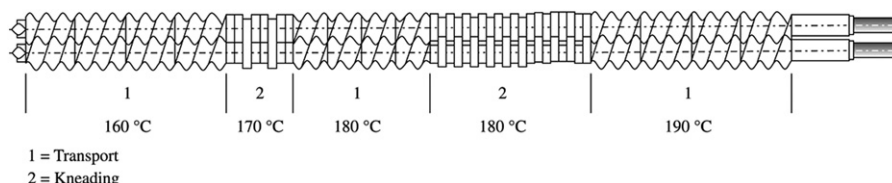


Fig. 2. Profile of the screw used.

cellulose and 9.9% hemicellulose). The composition of fibers may have different proportions of their constituents due to different conditions in which the plants grow [2].

The process of mixing the HDBPE, LHPB and curaua fibers was first carried out utilizing torque rheometry with an internal mixer. Fig. 3 shows the torque curves for the different compositions of the HDBPE/LHPB/fiber composites. There are two peaks: the first is related to the loading of the fiber in the mixing chamber [52] followed by the melting of polyethylene, which decreases the torque. The next increase in torque is related to the increase of the mixture viscosity, which is significant at approximately 1 min of mixing (Fig. 3).

3.1. Thermal analysis

The TG of the curaua fiber (figure not shown) exhibited a mass loss of 5% at approximately 100 °C, which can be attributed to the loss of water attached to the fibers. Curaua fibers exhibit thermal stability up to 260 °C, at which temperature the decomposition of hemicellulose begins, followed by the decomposition of cellulose, as indicated by the peaks related to the maximum weight loss at 305 and 372 °C, respectively, in the dTG curves (Fig. 4a, Fiber). At approximately 400 °C another decomposition process begins that involves the breaking of the lignin bonds [24,35], as indicated by the peak at 523 °C.

The decomposition began at lower temperatures in the composites (HDBPE/10 Fiber, HDBPE/5 LHPB/10 Fiber) than in the thermoplastic (HDBPE processed) due to the presence of lignocellulosic fibers in the former (Fig. 4a). The DSC curve of the curaua fiber (Fig. 4b) shows an endothermic peak (85 °C) resulting from the volatilization of the residual moisture. The narrow endothermic peak that is observed at approximately 130 °C in the DSC curves of the composites (Fig. 4b) corresponds to the melting of HDBPE, which is characteristic of semi-crystalline materials [53].

3.1.1. Crystallinity from DSC

From the DSC curves, the crystallinity index for the composites and the HDBPE can be calculated using the following relationship [54]:

$$X_c = \frac{\Delta H_m}{\Delta H_{m\phi_m}^0} \times 100 \quad (1)$$

where X_c is the crystallinity index, ΔH_m is the melting enthalpy of the sample, ΔH_m^0 is the melting enthalpy of hypothetical 100% crystalline HDPE and ϕ_m is the mass fraction of HDBPE in the composites. The index of crystallinity can be estimated for the different samples of bio-polyethylene (HDBPE) by considering that the melting enthalpy for 100% crystalline HDPE is 293 J g⁻¹ [55]. In this calculation, ΔH_m^0 is considered to be the same for both HDPE and HDBPE because the difference between the two polymers lies mainly in the route used to prepare the ethylene monomer from which the polymers were obtained.

Table 1 presents the crystallinity index (X_c) for the composites and the HDBPE, as well as the ratio between ΔH_m and ϕ_m .

The crystallinity indices of HDBPE in the composites are lower than that of the HDBPE, and they increase with fiber content. For samples containing 15 and 20 wt% fiber, with or without 5 wt% LHPB, significant differences are observed, which suggests an increase in the crystallinity of the matrix relative to other composites.

3.2. DMTA

The incorporation of fibers in the polymer matrix can change the mechanical properties of the composites. High storage modulus values were obtained throughout the temperature range examined by increasing the fraction of the curaua fiber in the composites from 5 wt% to 10–15 wt% (Fig. 5).

Typically, HDBPE exhibits two relaxations. The first occurs at approximately –120 °C, which is called γ by many authors and is related to the non-crystalline region; some researchers associate this relaxation with the glass transition [56]. The second relaxation appears between 50 and 120 °C, and can be associated with an interlamellar shear process; this relaxation can be split into two peaks due to the non-homogeneity of the crystalline regions [56]. In the samples analyzed here, these transitions appear between 30 and 50 °C, as observed in Fig. 5b.

For the HDBPE/LHPE/fiber composite, the peak that appears at lower temperatures occurs at approximately –125 °C. A peak between –75 and –50 °C, which is characteristic of the LHPB glass transition, is also observed [57]. The peak at approximately 35 °C is characteristic of

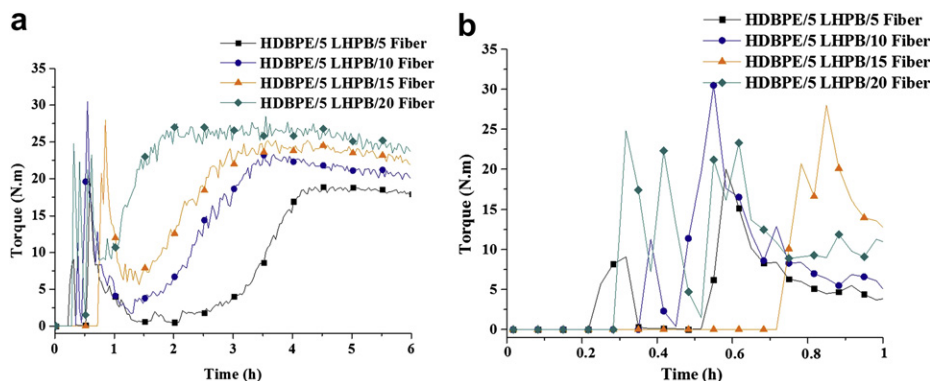


Fig. 3. Processing curves of HDBPE and related curaua composites: (a) Torque as a function of time (0–6 min), (b) 0–1 min.

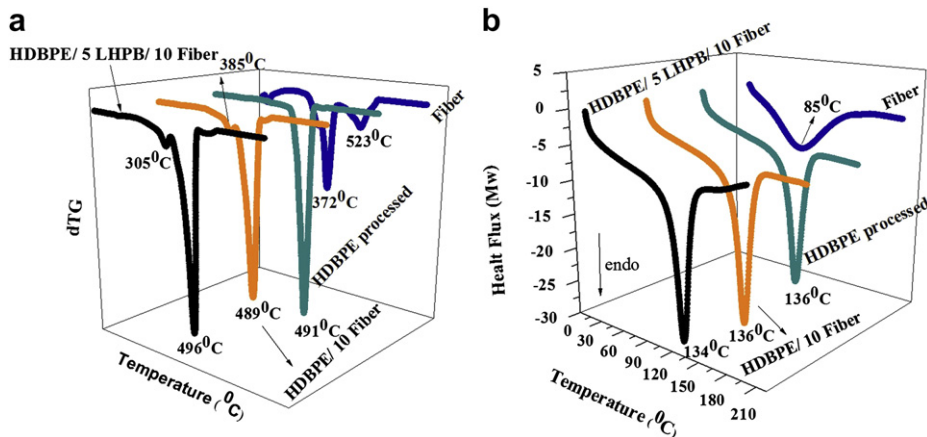


Fig. 4. Curaua fiber, processed HDBPE and curaua composites (HDBPE/10 wt% of fiber, HDBPE/5 wt% of LHPB/10 wt% of fiber): (a) dTG curves, (b) DSC curves.

a relaxation that involves the shear interlayer region [56], and a shift of the peak towards higher temperatures is observed for the composites (Table 2).

For the HDBPE/fiber composite, the peak that is observed at approximately -120 °C for HDBPE shifts to approximately -125 °C. The peak that occurs around -70 °C is not observed in the composites, and a third peak is observed in the range 34.8 – 36.4 °C for the composites with HDBPE/fiber (Table 2).

3.3. Impact strength

In comparing the composites processed by internal mixer/thermopressing molding with the neat polymer, HDBPE presented impact resistance of 234 J m $^{-1}$, and the addition of 5 wt% LHPB to HDBPE resulted in a composite with impact strength of 209 J m $^{-1}$. An increase in the impact strength of the HDBPE/LHPB blend, at first expected as the rubber can act as an impact modifier, was not observed. The rubber used in this study (LHPB) is hydroxylated, meaning that it has polar groups that have no affinity for the nonpolar polymer. This structural characteristic probably leads to preferential interactions between the rubber chains themselves, which in turn leads to aggregates. Thus, the presence of the elastomeric phase decreased the impact strength, rather than increasing it,

because the formation of clusters may have introduced flaws in the material. However, it must be emphasized that the rubber used in this study was chosen due to the presence of polar groups and nonpolar chains in its structure, which promotes favorable interactions in the composites prepared from a nonpolar matrix and fibers that contain polar groups.

The introduction of LHPB increases the impact strength of the composites (Fig. 6a). In this case, the presence of a rubber may have contributed a toughening effect because the interactions between both the polar groups of the LHPB/fibers and the nonpolar chains of the LHPB/HDBPE (Fig. 1e) might lead to a better dispersion of the rubber into the material when compared to the LHPB/HDBPE blend, in which no groups are available to interact with the polar sites of LHPB. It must be noted, however, that the previously mentioned interactions between the LHPB/fibers and the LHPB/HDBPE also cause LHPB to act as a compatibilizing agent. Therefore, the presence of LHPB also improved the impact strength due to the higher adhesion at the interface; that is, the load was transferred better from the matrix to the fiber through this interface with stronger interactions.

The impact strength decreased slightly as the percentage of fibers for the 15 and 20 wt% composites (80/5/15 and 75/5/20, respectively, Fig. 6a) increased, probably because the dispersion of a larger amount of fiber into the matrix was more difficult. In addition, the percentage of the compatibilizer, LHPB, was the same (5 wt%) for all composites; therefore, the relative ratio of %LHPB to %fiber decreases as the fiber content increases from 5 to 20 wt%.

The composite HDBPE/5 wt% LHPB/10 wt% fiber composite was easily prepared using an internal mixer (Haake) followed by thermopress molding, and exhibited good impact (Fig. 6a) and flexural properties (Fig. 8a, which is discussed later). The impact resistance and flexural properties were similar to those of a composite reinforced with a smaller amount of fiber, HDBPE/5 wt%LHPB/5 wt% fiber (90/5/5, Figs. 6 and 8a), and better than that of the other composites reinforced with larger amounts of fiber (15 and 20 wt%, 80/5/15 and 75/5/20, Figs. 6 and 8a).

Table 1

The melting enthalpy, the ratio between ΔH_m and ϕ_m and the crystallinity index.

Composite	ΔH_m (J g $^{-1}$)	$\Delta H_m/\phi_m$	Xc (%)
HDBPE processed	196.0	196.0	66.9
HDBPE/5%LHPB/5%Fiber	145.0	161.1	55.0
HDBPE/5%LHPB/10%Fiber	142.0	167.0	57.0
HDBPE/5%LHPB/15%Fiber	144.8	181.0	61.8
HDBPE/5%LHPB/20%Fiber	143.9	191.8	65.5
HDBPE/5%Fiber	151.2	159.0	54.3
HDBPE/10%Fiber	149.0	166.0	56.7
HDBPE/15%Fiber	148.6	175.0	59.7
HDBPE/20%Fiber	143.1	179.0	61.1

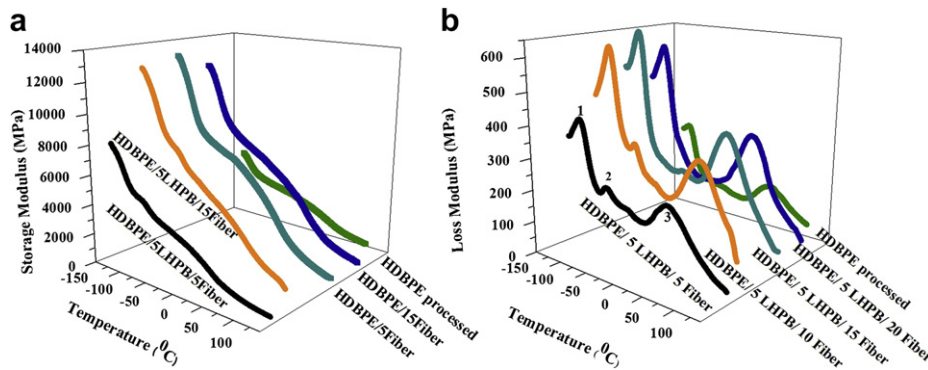


Fig. 5. HDBPE and related curaua composites: (a) Storage modulus and (b) loss modulus as a function of temperature.

Table 2

HDBPE and related curaua composites: DMTA peaks.

	Temperature (°C)		
	Peak 1	Peak 2	Peak 3
HDBPE processed	-120.0	-	36.0
HDBPE/5% LHPB/5% fiber	-122.0	-73.2	36.5
HDBPE/5% LHPB/10% fiber	-121.1	-73.4	39.8
HDBPE/5% LHPB/15% fiber	-121.1	-73.6	40.8
HDBPE/5% LHPB/20% fiber	-121.3	-72.0	40.9
HDBPE/5% fiber	-124.0	-	34.8
HDBPE/10% fiber	-123.6	-	36.4
HDBPE/15% fiber	-125.2	-	36.2
HDBPE/20% fiber	-126.0	-	35.6

Therefore, this composition was selected for preparation by extrusion and was molded by injection to characterize the mechanical properties, specifically the impact and flexural strengths.

It was observed that the impact strength of the composite that was prepared from extrusion and injection molding was considerably higher than that of the composite prepared from internal mixing (Haake) and thermopressing (Fig. 6b). The shearing that occurs during processing via extrusion is more intense than that observed during processing via internal mixing (Haake); the increase

in shear intensity is likely to lead to better dispersion of the fiber in the material, thereby favoring the load transfer from the matrix to the fiber during impact.

Araújo et al. [52] evaluated the effects of adding curaua fibers to high-density polyethylene (HDPE) using PEMA (polyethylene grafted with maleic anhydride) and EVA (copolymer poly (ethylene-co-vinylacetate)) as coupling agents; the mixture was then processed by extrusion and injection molded with a ratio of 20% fiber. Izod impact tests, using notched samples, showed that the coupling agent PEMA was efficient and promoted an increase of 15% in the impact resistance of the composite (65 J m^{-1}) compared to the composite that was reinforced with 20% curaua fiber without a coupling agent (58 J m^{-1}). In the present work, the notched impact strength of the composites with HDBPE/10 wt% curaua fiber and HDBPE/5 wt% LHPB/10 wt% curaua fiber (which were prepared using an internal mixer (Haake) followed by thermopress molding) were 97 and 152 J m^{-1} (Fig. 6a), respectively. This is an increase of approximately 55% when the compatibilizer (LHPB) was added to the composite. In addition, when the mixture of HDBPE/5 wt% LHPB/10 wt% curaua fiber was processed using an inter-meshing twin-screw extruder followed by injection molding, which results in both the presence of the

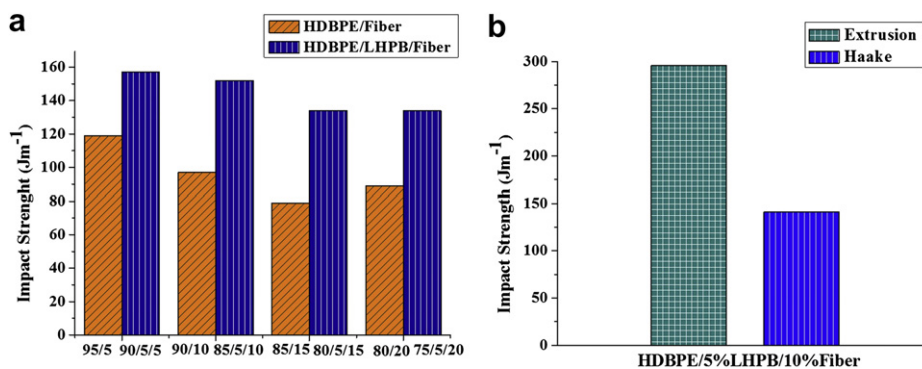


Fig. 6. Izod impact strengths for HDBPE curaua composites (notched samples, standard deviation between 5 and 10%). (a) Composites processed by internal mixer (Haake)/thermopress molding; (b) Comparison between the composite processed by internal mixer (Haake)/thermopress molding and Inter-meshing twin-screw extruder/injection molding.

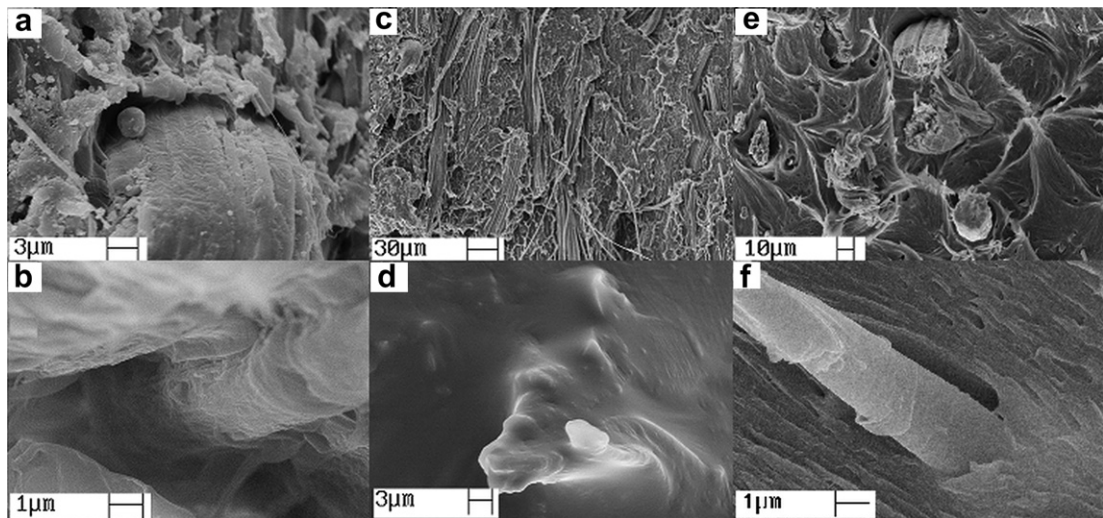


Fig. 7. SEM images of fractured surfaces of HDBPE curaua composites: (a) HDBPE/10%fiber; (b) HDBPE/5%LHPB/5%fiber; (c) HDBPE/5%LHPB/5%fiber; (d) HDBPE/5%LHPB/10%fiber; (e) HDBPE/5%LHPB/10%fiber extruded; (f) HDBPE/5%LHPB/10%fiber extruded.

compatibilizer (LHPB) and the influence of a different process to act on the impact strength of the resulting composite, the notched impact strength increased to approximately 300 J m^{-1} (Fig. 6b).

The composites prepared in the present study exhibited higher impact strength than a commercial HDPE composite reinforced with 30% glass fibers (notched Izod impact strength of 70 J m^{-1}) [52]. These results suggest a positive contribution of the LHPB to the performance of the composite.

3.4. SEM analysis

Following the impact test, the fractured surfaces of the samples were analyzed using scanning electron microscopy. In the micrograph of the HDBPE/10%fiber composite (Fig. 7a), detachment of the fiber from the matrix is observed. However, in some images, such as the micrographs of the HDBPE/5%LHPB/5%fiber composite (Fig. 7b) and the HDBPE/5%LHPB/10%fiber composite that was prepared from extrusion and injection molding (Fig. 7f), it is observed that there is a strong adhesion of fiber to the

matrix, which confirms the compatibilizing action of LHPB (Fig. 1e), and results in a better interface.

In the SEM image of the HDBPE/5%LHPB/10%fiber composite (Fig. 7d), we observe a layer of hydroxy-terminated polybutadiene (LHPB) that coats the surface of the fiber and matrix, whereas in the image of the HDBPE/10%fiber composite (Fig. 7c), the fiber and matrix can be viewed individually, which suggests a higher adherence of the fiber to the matrix when the rubber is present (Fig. 7d).

SEM analyses were also performed for the other composites, HDBPE/fiber and HDBPE/LHPB/fiber, which have different compositions (figures not shown), and the observed images were similar to those shown in Fig. 7.

3.5. Flexural strength

The flexural strengths of the neat polymer (HDBPE) and the blended HDBPE/5 wt% LHPB polymer at 5% deformation were 16.5 and 12.5 MPa, respectively; both polymers were prepared using an internal mixer (Haake) and thermopress molding. Neither the neat polymer nor the blended polymer breaks up at a deformation of up to 5%. Thus, the

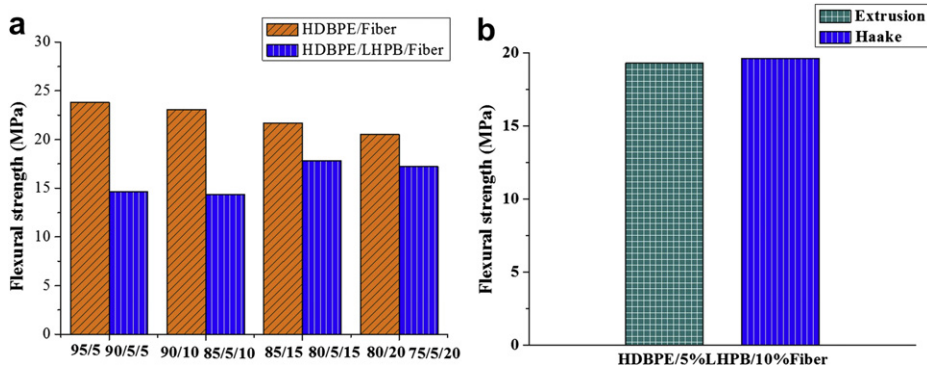


Fig. 8. Flexural strength at 5% of deformation for HDBPE curaua composites (standard deviation between 5 and 10%). (a) Composites processed by internal mixer (Haake)/thermopress molding; (b) Comparison between the composite processed by internal mixer (Haake)/thermopress molding and Inter-meshing twin-screw extruder/injection molding.

flexural strength values can be viewed as an indication that lower force was required to deform the material by 5% when rubber (LHPB) is present.

As shown in Fig. 8, the flexural strengths of the composites with different percentages of curaua fiber and LHPB, as well as for the neat polymer, HDBPE, were obtained. The stress versus strain curves showed the same profile for all samples (composites of HDBPE/LHPB/fiber and HDBPE/fiber, and HDBPE, figures not shown). These materials also do not break up when deformed up to 5%.

When comparing the set of composites that do or do not contain LHPB, the flexural strengths were similar to those loaded with 5, 10, 15 and 20 wt% fiber (Fig. 8a). For the HDPE/fiber and HDPE/LHPB/fiber composites, increasing the fiber content from 10 to 15 wt% and 10 to 20 wt% resulted in materials that had slightly lower and higher flexural strengths, respectively. This difference can be viewed as an indication that the mechanism that acts on the transference of the load from the matrix to the fiber changed due to the presence of LHPB, probably because the presence of LHPB resulted in stronger adhesion at the interface.

The presence of LHPB resulted in composites that had lower flexural strength (Fig. 8a). It should be highlighted again that the composites did not break during the test; therefore, the data plotted in Fig. 8 corresponds to a flexural strength at 5% strain. Thus, when the composites were submitted to bending stress, even though the presence of the compatibilizer LHPB led to stronger adhesion at the interface, its rubber behavior resulted in a material that required lower flexural force to bend by 5% deformation when compared to the composites in which there was no LHPB.

No significant differences were observed between the flexural strength of the extruded composite and the composite processed via Haake (Fig. 8b). As previously noted, the fibers were probably better dispersed in the composite that was subject to more intense shearing when processed via extrusion when compared to the composites prepared from internal mixing (Haake). Thus, the composite obtained via extrusion exhibited higher impact strength (Fig. 6b). The flexural strength did not appear to be influenced by the properties of the bulk of the composite; however, due to the characteristics of the flexural test, the properties of the outer surfaces of the samples may have had more influence on the flexural strength, and the outer surfaces were not influenced by the differences in processing.

4. Conclusions

In this study, it was possible to prepare materials using raw materials that are primarily based on renewable resources, e.g., biocomposite-type materials from biopolyethylene and curaua fibers, which possessed good properties.

DMTA analyses showed that the presence of fibers in the composites led to a more rigid material when compared to the pristine polymer. This result indicates that these composites can be used in applications where a higher storage modulus is needed.

An analysis of the morphology of the composites showed good adhesion between the fibers and the matrices, mainly when LHPB was present, because LHPB acted as

a compatibilizing agent, which allowed efficient load transfer from the matrix to the fiber through the interface. Thus, the impact strength, which is a very important property of composites, was improved in the presence of LHPB.

The type of processing that was used to prepare the composites proved to be very influential on the impact strength, as for a single composition (HDBPE/5%LHPB/10% Fiber), the impact strength increased from approximately 150 to 300 J m⁻¹ when the processing method was changed from internal mixing (Haake) and thermopress molding to inter-meshing twin-screw extrusion and injection-molding.

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