Carbohydrate Polymers 77 (2009) 47–53

Contents lists available at ScienceDirect

Carbohydrate Polymers

journal homepage: www.elsevier.com/locate/carbpol

Characterization of lignocellulosic curaua fibres

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ARTICLE INFO

Article history:
Received 29 October 2008
Received in revised form 27 November 2008
Accepted 1 December 2008
Available online 14 December 2008

Keywords:
Curaua fibres
Mechanical properties
Thermal properties

ABSTRACT

Curaua fibres have specific mechanical properties similar to inorganic fibres and are an important renewable raw material. Milled curaua fibres, submitted to different treatments, were characterized by mechanical and thermal properties, moisture content, water absorption, surface morphology, FTIR spectroscopy, density and X-ray diffraction. Except for moisture content and mechanical properties, no other significant changes were observed after the treatments. The treated fibres also show an increase of surface roughness.

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1. Introduction

The use of lignocellulosic fibres as reinforcing agents in polymeric composite materials in substitution of synthetic fibres like glass fibres or carbon fibres is increasing. This is occurring due to the favourable properties of natural fibres, in comparison to fibreglass, such as environmentally friendly character, low cost, low density, non-toxicity, no abrasion during processing and recyclability. Consequently, there is an increased motivation in the use of these fibres by different industrial sectors, like automotive, to replace glass fibres (Avérous & Le Digabel, 2006; Bledzki & Gassan, 1999; Klemm, Heublein, Fink, & Bohn, 2005; Rao & Rao, 2007; Saheb & Jog, 1999). Besides, the use of natural fibres does not consume the energy required to melt and process fibreglass.

Lignocellulosic fibres are constituted by three main components: hemicellulose (20–40 wt%), cellulose (40–60 wt%), and lignin (10–25 wt%), which are known to present very complex structures (Yang, Yan, Chen, Lee, & Zheng, 2007). Cellulose, which is the main fraction of the fibres, is a semicrystalline polysaccharide made up of D-glucosidic bonds. A large amount of hydroxyl groups in cellulose (three in each repeating unit) gives hydrophilic properties to the natural fibres. Hemicellulose is strongly bound to the cellulose fibrils, presumably by hydrogen bonds. Hemicellulose polymers are branched, fully amorphous and have a significantly lower molar mass than cellulose. Because of its open structure containing many hydroxyl and acetyl groups, hemicellulose is partly soluble in water and is hygroscopic. Lignins are amorphous, highly complex, mainly aromatic polymers with phenylpropane units, but have the lowest water sorption of the natural fibre components (Li, Tabil, & Panigrahi, 2007).

Many fibres have been studied and tested for use with polyolefin composites, like: jute, coir, flax, kenaf, hemp, sisal, etc. (Joffe, Andersons, & Wallström, 2003; Bullions, Hoffman, Gillespie, Price-O'Brien, & Loos, 2006; Dhakal, Zhang, & Richardson, 2007; Ouajai & Shanks, 2005; Rao & Rao, 2007). However, like fibreglass, natural fibres present poor compatibility with the non-polar polymeric matrix and relatively high moisture sorption; these factors affect the mechanical properties of the composites. In order to improve polymer-fibre adhesion and to reduce water absorption, the surface of the fibre can be modified by physical or chemical methods (Li et al., 2007).

The fibres studied in this work were extracted from the leaves of the *Ananas erectifolius L. B. Smith* plant (hydrophilous species), usually called *curaua*, which is cultivated in the Amazon region. This plant is cultivated in semi-arid conditions and its chemical composition is 73.6% of cellulose, 9.9% of hemicellulose, 7.5 of lignin and 0.9% of ashes (Caraschi & Leão, 2001). Their 1.5–1.7 m long and 4 cm wide leaves are hard, erect and have flat surfaces, Fig. 1. In 2003, Brazil produced 150 ton of curaua (Leão, Sartor, & Carashi, 2006). Previous studies indicate that curaua fibre is a promising material to reinforce thermosets and thermoplastics (Leão, Rowell, & Tavares, 1998; Santos, Spinacé, Fermoselli, & De Paoli, 2007), however, its surface treatment may improve this effect and was not studied.

Aiming to use natural fibres as reinforcing agents for polymers, this work studied and characterized pristine curaua fibres and curaua fibres washed with water, treated with sodium hypochlorite solution and treated with cold oxygen plasma. The density and the distribution of the diameter of these fibres were measured.

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doi:10.1016/j.carbpol.2008.12.005

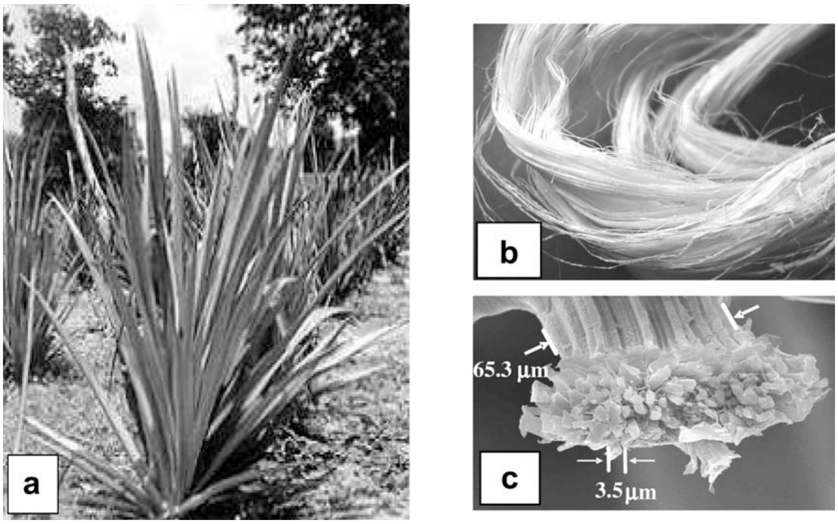

Fig. 1. (a) Curaua plant (*Ananas erectifolius L. B. Smith*), (b) dry curaua fibre and (c) scanning electron micrograph of the tip of a curaua fibre.

The mechanical and thermal properties, surface morphology, water absorption, humidity, FTIR spectroscopy and X-ray diffractions were used to characterize these fibres.

2. Experimental procedure

2.1. Surface treatment of the curaua fibres

The curaua fibres (C) were supplied by POEMA (Nucleus for Sustainable Development Action, Belém, Brazil) in the form of 1.0–1.5 m long fibres bundles. One part of these was washed in a conventional washing machine (Electrolux, Front Load LE05) at 80 °C for 1 h, with cold tap water (CW). Another part (CH) was treated with an aqueous sodium hypochlorite solution (Super Cândida, active chlorine content from 2.0 to 2.5 wt%) for 5 h by immersion with no stirring (using a mass ratio 15:1 sodium hypochlorite solution: fibre). After that, the fibres were washed several times with tap water. In both cases, the fibres (CW and CH) were dried in an open atmosphere (25 °C) for 48 h and in an oven (60 °C) for 12 h. The fibres were not directly exposed to sunlight during these treatments.

A third part of the curaua fibres (CP) was treated for 15 min with cold oxygen plasma in a quartz reactor (13.56 MHz, 30 W, 0.53 Pa).

After these treatments, the fibres were milled in a three knife rotary mill (Rone, model NFA 1533) equipped with a 7 mm mesh sieve and milled again in a four knife rotary mill (Thomas–Wiley Mill, ED-5) equipped with a 0.5 mm mesh sieve.

2.2. Characterization of curaua fibres

The following analyses were made on the original fibre samples:

The diameter of about 120 fibres was determined using a micrometer calliper (Mitutoyo, ±0.001 mm). The diameter was measured at 3 points of 5 cm long samples and the smaller value was considered. Plots of the fibres diameter frequency distribution, using the average frequency, in relation to each diameter interval, are shown in Fig. 2.

The mechanical properties of the single curaua fibres were measured according to ASTM standards for tensile properties (ASTM D3379) using an EMIC DL 2000 equipment. The load was measured with a 500 N load cell at a loading rate of 2.0 mm min^{-1}. Fibres (100) were tested and the average value and standard deviation for each property was calculated for a minimum of 45 fibres.

Micrographies of the fibres were obtained using a scanning electron microscope (JEOL JSM–6360LV) at an acceleration voltage of 25 kV. The samples were coated with gold prior to measurement (Bal-Tec, Multi Coating System MED020).

The following additional analyses were made using the milled fibre samples:

The density of dried fibres (100 °C, 1 h) was determined in duplicate or quintuplicate at 23 °C using a helium Picnometer (Micrometrics 1305).

The moisture content was evaluated in triplicate according to ASTM D 2654, procedure 1, with a room humidity (RH) of 55 (±7)% and a temperature of 25 °C. For this purpose 0.5000 g of fibres were maintained in a conventional oven (FANEM, 315SE) at 100 °C (±5) and the weight of the specimens was measured at time intervals until reaching a constant value.

The water absorption was determined by gravimetry in triplicate using 1 ± 0.0001 g of fibres dried at 100 ± 2 °C for 4 h. The samples were maintained for 572 h at 24 ± 1 °C in a closed system containing a saturated solution of sodium dichromate ($Na_2Cr_2O_7 \cdot 2H_2O$, Merck, P.A.) to maintain the relative humidity, RH, at 54.5%. The weight of the samples was measured at time intervals until reaching a constant value.

The percentage content of moisture and water absorption were calculated using Eq. (1), where W_i and W_f are initial and final weight of specimens.

$$W = \frac{(W_i - W_f) \times 100}{W_i} \tag{1}$$

The thermal analysis of curaua fibre samples (10 mg), with and without treatment, were made by differential scanning calorimetry, DSC (Netzsch STA 409C Thermal Analyzer) under a helium atmosphere in the temperature range from −50 to 600 °C with a heating rate of 10 °C/min. Thermogravimetry, TGA (TA Instruments 2050) was done in the temperature range from 25 to 800 °C with a heating rate of 10 °C/min under an argon atmosphere (50 mL/min). The Fourier Transform Infrared, FTIR, measurements (Bomem, MB-100 Series) were made with KBr pellets in the 450–4000 cm^{-1} region, with 4 cm^{-1} resolution and 64 scans. The mixture of KBr and fibres was dried (100 °C, 1 h) and the samples were prepared immediately before measurement.

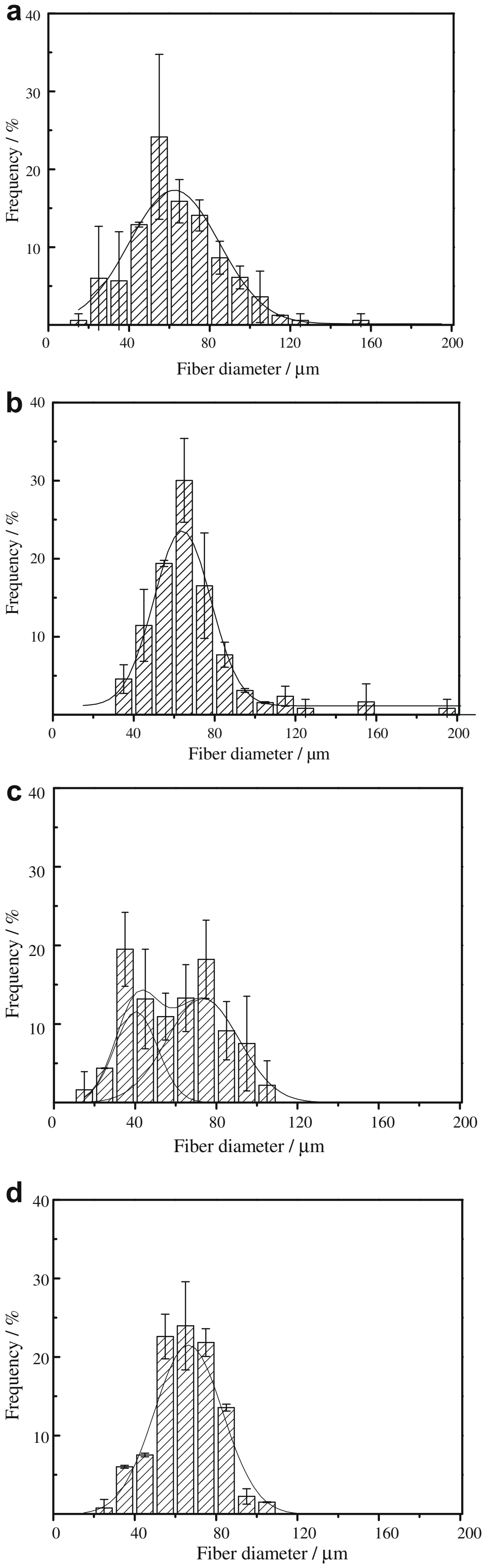

Fig. 2. Distributions of fibre diameters: (a) C, (b) CW, (c) CH and (d) CP.

The degree of crystallinity of the different treated curaua fibres was calculated from X-ray diffractograms, which were recorded on a Shimadzu, XRD6000, with Ni-filtered Cu Kα radiation at 40 kV, 30 mA and (2θ) angle from 5 to 50°.

3. Results and discussion

3.1. Density and diameter distribution of curaua fibres

The densities obtained for C, CW, CH and CP fibres were 1100 (±91), 1200 (±99), 1100 (±91) and 1100 (± 91) kg/m^3, respectively, showing that the surface treatment does not affect the density. These values are comparable to other natural fibres like palm (1030 kg/m^3) and coconut (1150 kg/m^3) and smaller than cotton (1500–1600 kg/m^3), flax, sisal (1500 kg/m^3) and banana fibres (1350 kg/m^3) (Aquino, Sarmento, Oliveira, & Silva, 2007; Bledzki & Gassan, 1999). All vegetal fibres, including curaua, present densities lower than glass fibres (2500 kg/m^3).

The distributions of fibre diameters are shown in Fig. 2. The frequency distribution of diameters, assuming a cylindrical shape, is Gaussian. The diameters change from 20 to ~130 μm, with an average value of 60 μm. The fibres treated with an aqueous sodium hypochlorite solution (CH) present a bimodal distribution of diameter (Fig. 2c), probably because this reagent weakens the interaction between the fibrils in the fibre bundle causing partial fibrillation. The fibres washed with water (CW) and treated by plasma (CP) present a narrower range of distribution (Fig. 2b and d), probably due to removal of surface impurities, but the average values are similar to the non-treated fibre.

The scanning electron micrograph of a curaua fibre (Fig. 1c) shows a fibre with a diameter of ca. 60 μm. It is also possible to see fibrils, which constitute the fibre, with a diameter of ca. 3.5 μm.

3.2. Moisture content and water absorption

The fibres moisture content depend on the atmospheric conditions, type of fibre, age of plant, soil condition in which the plant was grown and the method and duration of preservation. The percentage of moisture in the fibre under normal atmospheric conditions changes from 9.16 to 12.09 wt% (Aquino et al., 2007). The determination of content, temperature and time necessary to eliminate the moisture is important for composite processing, because moisture influences the adhesion between the fibres and the polymeric matrix. For this determination, it was considered that only water evaporation occurs at 100 °C. As seen in Fig. 3a, curaua fibre presents 9.10 wt% of moisture, while 50 min is sufficient to dry the fibre. For treated fibres the moisture content is reduced to 8.30 wt%. This reduction indicates that the treatment changed the hydrophilic character of the curaua fibres. Besides moisture content, it is also important to know how much time is necessary for the fibre to reabsorb moisture. This information can be used to evaluate the properties of the final product. Fig. 3b shows the absorption curves for C, CW, CH and CP fibres in a controlled atmosphere and temperature (RH = 54.5%, T = 25 °C). The time for the fibres to reabsorb the total water content is long, but the absorption rate is high in the first 25 h.

Hemicelluloses, present in the fibres, are mainly responsible for moisture absorption; but accessible cellulose, amorphous cellulose and lignin also contribute to this process (Aquino et al., 2007). Moisture swells the cell wall and the curaua fibre swell until the cell wall is saturated with water. Beyond this point, moisture exists as free water in the void spaces of the fibre and does not swell the fibre further. Although the treated fibres presented lower moisture

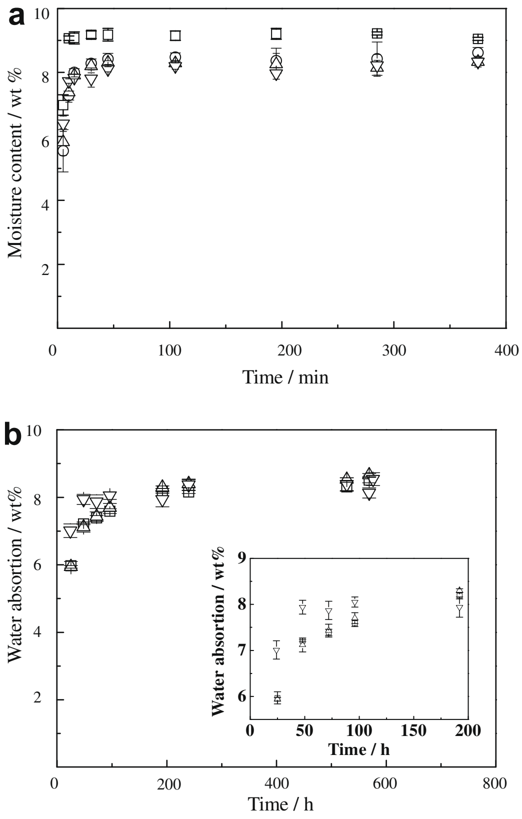

Fig. 3. (a) Moisture content and (b) water absorption of curaua fibre as a function of time for samples: C (■), CW (●), CH (▲) and CP (▼).

content than pristine curaua fibres, the water absorption process is similar for both treated and untreated fibres, Fig. 3b.

3.3. Mechanical properties

The tensile stress–strain behaviour of single cellulose-based fibres is complex and can not be described by a simple linear elasticity model. Previous studies (Sedighi-Gilani & Navi, 2007) of tensile behaviour of wood cells report that, beyond the yield point, permanent deformation occurs and the rigidity increases. Consequently, the fibre shows a load-history dependent behaviour. Non-uniformity of microfibril angle (MFA) and other defects in the wood cell can influence the results of the test. Successive damage of the hemicelluloses and lignin and reduction of MFA is mainly responsible for the visco-elastic behaviour of a wood cell. Curaua fibre presents elastic behaviour with brittle failure, or silk-like curves, being almost constant until the rupture point (Billmeyer, 1984). The literature reports different fibres that present mechanical properties featuring a sharp drop in modulus at low stress followed by a period of elongation at almost constant stress (Prasad, Pavitram, & Rohatgi, 1983; d'Almeida, Aquino, & Monteiro, 2006; Kompella & Lambros, 2002).

The results of tensile strength (σ), Young modulus (E) and elongation at break (ε_b) for C, CW, CH and CP fibres are shown in Fig. 4a and c. The tensile strength (900 ± 200 MPa) and Young modulus (36 ± 10 GPa) values obtained for curaua fibres are higher than for other fibres like bamboo, palm, coconut or sisal. Consequently, curaua fibres are stiffer and stronger than these fibres (Rao & Rao, 2007). The treated fibres, CH and CP, show slightly lower values of σ and E than untreated curaua fibres, however, unchanged within the experimental error. The fibres washed with water, CW, show a different behaviour, with a considerable decrease in these properties and a higher elongation at break. This could be due to mechanical damage during the washing or a plasticization effect.

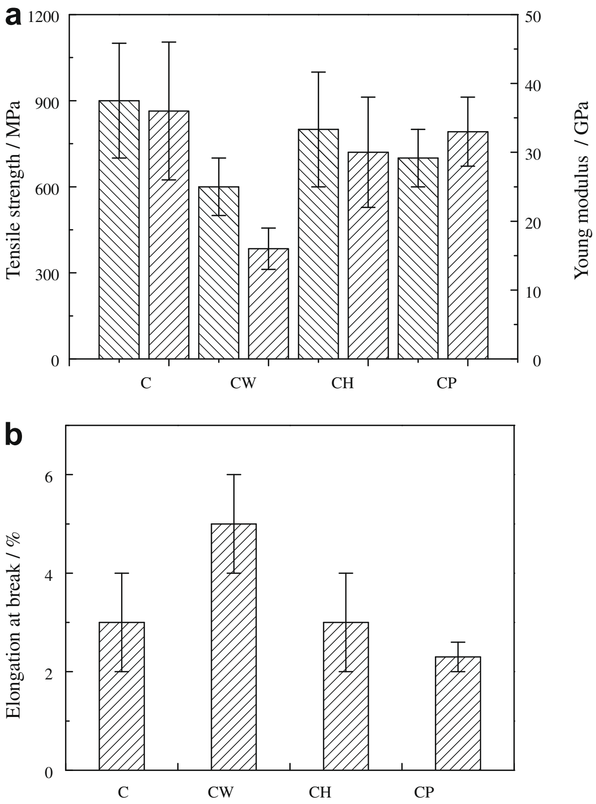

Fig. 4. Values of: (a) yield tensile strength (\\\\\\) and Young modulus (///) and (b) elongation at break for curaua fibres with and without surface treatments.

Fig. 4b and Table 1 shows the specific values (divided by density, ρ) of σ, E and ε_b for C, CW, CH and CP fibres. These values for Curaua fibres with and without surface treatment are two times larger than for sisal fibres (σ = 341–423 MPa cm^3/g and E = 6.3–14.7 GPa cm^3/g) and comparable to the specific mechanical properties of commercial short glass fibres used as reinforcing agent for thermoplastics (Bledzki & Gassan, 1999).

The specific mechanical properties of the curaua fibres are very important because the good mechanical properties are combined with a low density (Bledzki & Gassan, 1999; Li et al., 2007; Munawar, Umemura, & Kawai, 2007). Although the surface treatment of these fibres caused a small decrease in these properties; the results indicate that this fibre is an interesting material as a reinforcing agent for polymers, after adjusting the interfacial adhesion.

3.4. Thermal properties

Studies on the kinetics of lignocellulosic fibre thermal degradation indicated the existence of some interactions between hemicellulose, cellulose and lignin. Thus, the behaviour of this degradation process is not a simple additive function of each contributing frac-

Table 1
Values of specific yield stress (σ), Young's modulus (E) and elongation at break (ε_b) for treated and untreated curaua fibres (ρ = density).

Fibre	σ/ρ (MPa/g cm^{-3})	E/ρ (GPa cm^3/g)	ε_b/ρ (% cm^3/g)
C	636–1000	36 ± 10	3 ± 1
CW	417–583	13 ± 3	4 ± 1
CH	545–909	30 ± 8	3 ± 1
CP	545–727	31 ± 5	2.2 ± 0.3

tion (Garcìa-Pèrez, Chaala, Yang, & Roy, 2001). The TGA curve of curaua fibres show four degradation processes related to: (i) moisture evolution, (ii) hemicellulose, (iii) cellulose and (iv) lignin degradation. The degradation process for each fraction indicates that, although there are interactions between these fractions, their basic identity is maintained.

The TGA curves for C, CW, CH and CP are shown in Fig. 5. After initial weight loss at 75 °C, onset temperature attributed to water loss in the form of absorbed moisture, there are three other processes: the first one from 236 to 297 °C with maximum decomposition rate at 268 °C is attributed to the decomposition of hemicellulose. The second process occurs from 298 to 366 °C, with a maximum decomposition rate at 335 °C, and is attributed to cellulose degradation. Comparing these three components, lignin was the most difficult to decompose, its decomposition occurred slowly, with a maximum decomposition rate at 439 °C. These results agree with values reported in the literature (d'Almeida, Aquino, & Monteiro, 2006; Garcìa-Pèrez, et al., 2001; Tanobe et al., 2005; Tejado et al., 2007; Yang et al., 2007).

The thermodegradation of polysaccharides, like hemicellulose and cellulose, can occur by cleavage of glycosidic, C—H, C—O and C—C bonds, dehydration, decarboxylation and decarbonylation reactions, with formation of C—C, C=C, C—O bonds as well as carbonyl and carboxyl groups (Garcia-Pèrez, et al., 2001). The weight loss of lignin occurs at higher temperatures and its degradation is related to dehydration, yielding derivatives with lateral unsaturated chains and the release of water, CO_2, CO and methane. According toYang et al. (2007) who studied the gas products obtained by pyrolysis of hemicellulose, cellulose and lignin; hemicellulose showed higher CO and CO_2 yield, while lignin displayed higher CH_4 production. Organic compounds (C=O, C—O—C, etc.) were mainly released at low temperatures, in the ranges from 200 to 400 °C and from 300 to 450 °C for hemicellulose and cellulose, respectively. For lignin no complex organic compounds were reported.

The fibre surface treatments do not influence the initial temperature of fibre degradation, which is related to hemicellulose degradation. Analyzing degradation of cellulose fractions in treaded fibres, fibres washed with water (CW) present an increase of 10 °C in the temperature of the maximum rate of the degradation process. The temperature of the degradation process of lignin is shifted to higher values (~476 °C) when the fibres were washed with water (CW) or treated with oxygen cold plasma (CP). The onset temperature of curaua fibre decomposition (220 °C) is comparable to other lignocellulosic fibres (d'Almeida, Aquino, Monteiro, 2006; Garcia-Pèrez, et al., 2001).

Thermal degradation reaction enthalpy was calculated from DSC measurements (Fig. 6). A small endothermic peak was observed at 100 °C, which is attributed to the removal of moisture when the sample was heated, while a large endothermic peak at 355 °C is related to the cellulose fraction. This behaviour is related to the full decomposition of cellulose that might be attributed to quick devolatilization reactions, leading to very little solid residue (as can be seen in the TGA curves). On the other hand, degradation of hemicellulose and lignin generate solid residues (Fig. 5) and the DSC profile (Fig. 6) shows two exothermal peaks at 275 and 365 °C, respectively, attributed to charring (Yang et al., 2007). The DSC profile obtained for the curaua fibres do not show peaks related to hemicellulose or lignin, probably due the smaller content of these fractions, as compared to cellulose. A similar behaviour for C, CW and CP samples was observed. The DSC of the CH sample was not measured to avoid damage to the cell of the equipment.

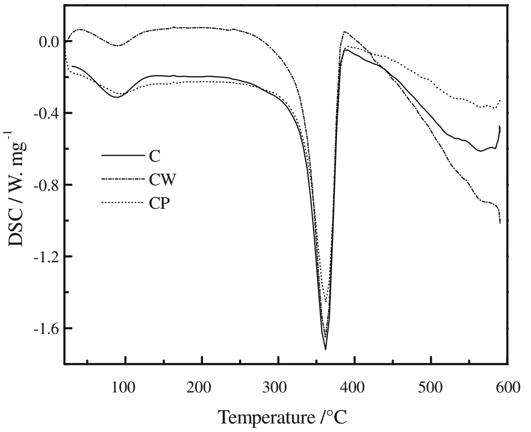

Fig. 6. DSC curves of curaua fibres.

3.5. X-ray diffraction and FTIR spectra

The X-ray diffractograms, XRD, of curaua fibres are shown in Fig. 7. Three peaks were observed for all samples at $2\theta = 16$; 22.6 and 34.7°. These are characteristic of the crystal polymorph I of cellulose. The peak at $2\theta = 16°$ correspond to the $(1\,\bar{1}\,0)$ and $(1\,1\,0)$ crystallographic planes and the peaks at $2\theta = 22.6$ and 34.7° correspond to the (0 0 2), and (0 2 3) or (0 0 4) planes, respectively. For fibres with higher cellulose content, like cotton or flax, two peaks around 16° are observed, but for curaua fibres, only one broad peak was observed due to the presence of amorphous materials like lignin, hemicelluloses and amorphous cellulose, which cover the two peaks Subramanian, Senthil Kumar, Jeyapal, & Venkatesh, 2005;

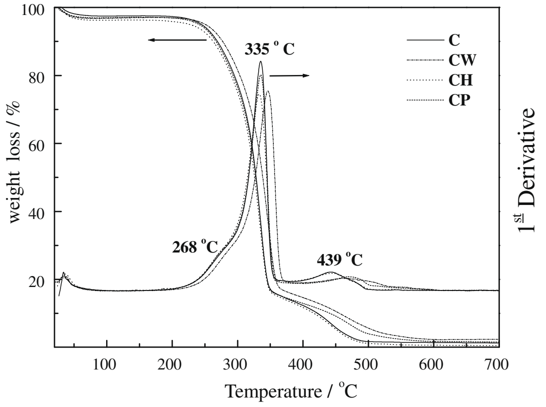

Fig. 5. TGA curves; weight loss and 1st derivative of the weight loss vs. temperature.

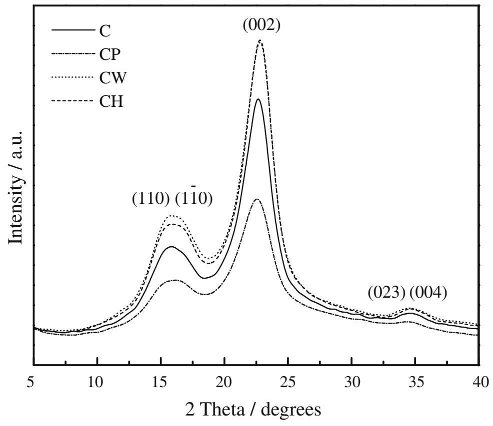

Fig. 7. X-ray diffraction of curaua fibres.

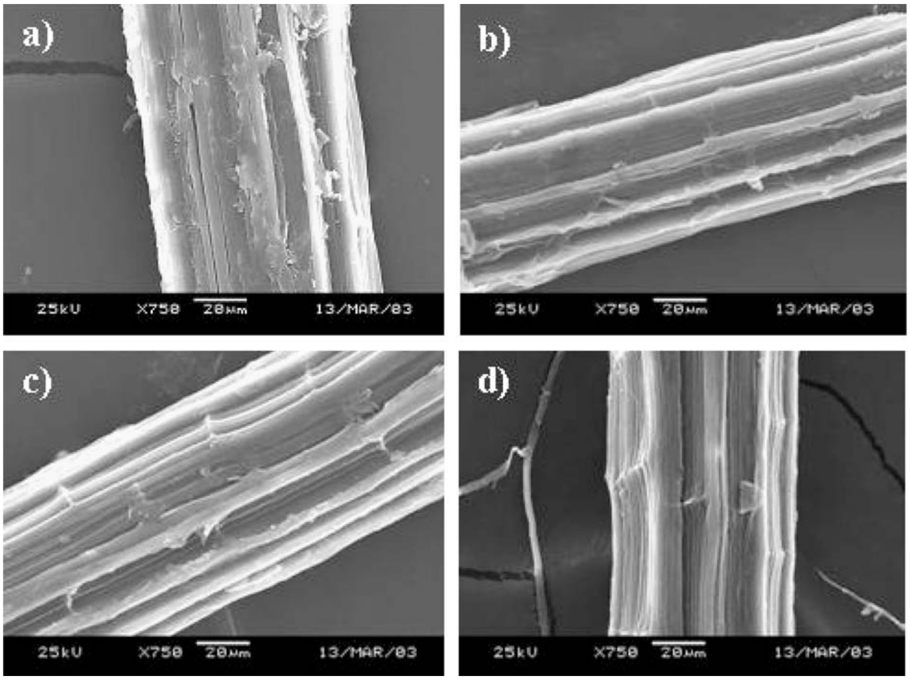

Fig. 8. SEM micrographs of fibres surface: (a) C, (b) CW, (c) CH and (d) CP.

Tserki, Matzinos, Kokkou, & Panayiotou, 2005; Tserki, Zafeiropoulos, Simon, & Panayiotou, 2005.

The crystallinity indices obtained from X-ray diffractograms for C, CW, CH and CP fibres show variations as a function of surface treatment. The CW and CH fibres show an increase of 28% and 29% in the crystallinity index. Zafeiropoulos, Williams, Baillie, and Matthews (2002) observed a similar result when flax fibres were treated by acetylation. Thus was attributed to two effects: (a) removal of some of amorphous constituents of the fibres and (b) there may be a rearrangement of the crystalline regions in such a way that the fibre shows a more crystalline nature. On the other hand, the crystallinity index decreases 33% for CP fibres in relation to C fibres.

FTIR spectra of C, CW, CH and CP fibres show absorption bands of chemical groups characteristic of the curaua fibres components: cellulose, hemicellulose and lignin. These components are most likely composed of alkenes and aromatic groups and different oxygen-containing functional groups, like ester, ketone and alcohol. The absorptions corresponding to these groups are observed at: OH (3400–3200 cm^{-1}), C=O (1776–1715 cm^{-1}), C—O—C (1270 cm^{-1}) and C—O(H) ($\sim$1050 cm^{-1}). The most intense IR band assigned to OH and C—O—C were found for cellulose, while hemicellulose presented stronger C=O bands. A large difference was found in the fingerprint region (1830–730 cm^{-1}) of the IR spectra of lignin compared to the spectra of hemicellulose and cellulose (Yang et al., 2007).

The C—O—C symmetric glycosidic stretching or ring stretching mode at 1100 cm^{-1} and the C—OH stretching vibration of the cellulose backbone at 1060–1050 cm^{-1} (νC—O secondary alcohol) arise from the polysaccharide components (that is, largely cellulose). Others peaks due to the alcohol groups of cellulose (δ OH deformation) appear at 1360 and 1320 cm^{-1} (Bessadok et al., 2007; Ouajai & Shanks, 2005). FTIR spectra of the curaua fibres show intense absorptions in the range from 3500 to 3200 cm^{-1} related to the OH stretching and hydrogen bonds, 2900–2750 cm^{-1} related to the CH stretching in saturated hydrocarbons, 1740 cm^{-1} related to C=O stretching of acetyl or carboxylic ester and 1240 cm^{-1} related to CH angular deformation. Apparently, the surface treatment of curaua fibres did not cause changes in their structure, which could be evidenced by FTIR.

3.6. Morphology

A morphology study of curaua fibres was carried out using scanning electron microscopy (SEM) to evaluate the fibre surfaces, Fig. 8. The changes in morphology are important to predict fibre interaction with the polymer matrix in composites. For pristine curaua fibre (Fig. 8a) a smooth and compact surface was observed, with no fibrillation. For treated curaua fibres (Fig. 8b, c and d), an increase of surface roughness was observed and fibrils could be distinguished on the surface for fibres treated with sodium hypochlorite solution (CH) and cold oxygen Plasma (CP). Consequently, an increase of surface area occurs which could result in an increase of adhesion in a fibre-polymeric matrix interface in the production of composites.

It is important to note that treatment with plasma occurs in only one step and does not generates residues like the treatment with hypochlorite solution does.

4. Conclusion

The type of fibre treatment studied in this work did not produce significant changes in the water absorption, DSC, TGA and FTIR, in comparison to pristine fibres. The treated fibres show smaller values of moisture content, which could indicate a change in their hydrophobic surface. The time for pristine and treated fibres to reabsorb this moisture is very similar. Pristine curaua fibres show specific mechanical properties comparable to inorganic fibres, which is very important for their use in reinforcement of polymer composites. On the other hand, the treatments used in this work, affect the mechanical properties. For fibres treated with water and sodium hypochlorite, XRD measurements indicated an increase of crystallinity, probably due to the extraction of part of the amorphous phase, while for fibres treated with cold oxygen plasma a decrease of crystallinity occurs. Treated fibres show an increase of surface roughness, which is important for mechanical adhesion in composites.

Our results indicate that the fibre treatment should be chosen according to the target application, depending on the hydrophilic character of the polymer with which it will be compounded.

Acknowledgements

We acknowledge financial support from FAPESP (02/09983-0 and 04/15084-6). MAP also thanks CNPq for a research fellowship.

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