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Natural Fiber Polymer Composites Technology Applied to the Recovery and Protection of Tropical Forests Allied to the Recycling of Industrial and Urban Residues

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1. Introduction

"*Often when pursuing research into green composites we say we are protecting the environment, that we are working for nature. We may as well stop kidding ourselves – nature will be fine; nature will work out OK and adapt to changes. It´s humans that will cease to exist if we continue the way we are at present. Some scientists and engineers have realized that they need to take responsibility for the outcome of their work. Researching ways of creating faster machines and bigger toys, without due consideration of the effects on the environment or on people, is irresponsible… We need to consider the impact that our material choice and design will have on the society and the environment*"*

Generation of residues is inherent to human activities. After the Industrialization Period, with modernization of the society, the amount of industrial and postconsumer residues, together with the associated environmental problems, has been increasing at alarming levels. Residue management has become a major problem in modern society. The search for innovative solutions for the reuse of solid residues increased in the late 20th century and has intensified with growing urgency for environmental preservation. Many residue management solutions aim to add value to residue through the development of new materials and processes.

Natural fiber polymer composites basic technologies are already relatively well established. Excellent textbooks can be found on this theme (Klyosov, 2007; Niska & Sain, 2008; Mohanty et al., 2005). Its concepts can be extended to the reuse of solid industrial and urban residues. In these composites the matrix is a polymer, and wood, vegetable or animal fibers are used as fillers or reinforcements. Both polymer and fiber may have been generated as an industrial or postconsumer residue.

Another environmental problem that must be accounted for is the huge amount of deforested areas worldwide, also a consequence of human activities. Important research has

* Baillie (2004)

been carried out on the possibility of recovering forests using modern concepts of Short Cycle Agrosilviculture, which allow generation of wood, fibers, food, biofuels, oils, pharmaceuticals etc, creating synergy between the recovery and protection of the environment and the development of new materials.
Within this context, the main purpose of this text is to present case studies where wood and natural fiber polymer composites technologies are applied to the recovery and protection of tropical forests allied to the recycling of industrial and urban residues.

1.1 Natural fiber composites

Natural fibers can be classified according to their origin: vegetable, animal or mineral. Vegetable fibers include wood flour (or sawdust) from a huge variety of softwood and hardwood specimens and plant fibers, such as hemp, kenaf, curaua, coir, jute, sisal, bamboo, among others. Animal fibers include silk and leather. The most well-known mineral fibers are glass, boron and asbestos; the latter in disuse nowadays as it has been found to be harmful to humans.
Over the past years sustainable eco-efficient practices and products have gained increasing attention and the use of natural fibers as reinforcement for polymers has been rapidly expanding (Bettini et al., 2010; Bonse et al., 2010; Kalia et al. 2009; Mohanti, 2005; Saheb & Jog, 1999; Sanadi et al, 1994). In certain composite applications biofibers have shown to be competitive in relation to glass fiber (Sudell & Evans, 2005). Advantages of biofibers over synthetic ones include the fact that they are renewable and biodegradable, present low cost, light weight, low energy consumption, carbon dioxide sequestration, low abrasiveness and excellent strength to weight ratio [Sanadi et al, 1994]. Limitations of biofibers as reinforcement in polymers are: difficulties in maintaining homogenization of fiber properties; processing temperature, which should not exceed the degradation temperature of these fibers of around 200°C; and high moisture absorption, which may impair mechanical properties as well as facilitate fungus growth (Kalia et al. 2009). Typical thermoplastic polymers that can be easily processed at temperatures up to 200-220°C, and thus may be used as matrix, include: polypropylene (PP), polyethylene (PE), polystyrene (PS) acrylonitrile-butadiene-styrene (ABS), PVC and more recently polyvinyl butyral (PVB) (Ambrósio et al., 2011).
Vegetable or lignocellulosic fibers consist mainly of cellulose, hemicelluloses and lignin, at major levels and pectin and waxes, at lower levels. In these fibers hollow cellulose fibrils are maintained united by lignin and cellulose. The fibrils have a thin primary wall surrounding a secondary wall, which consists of three layers (S1, S2 and S3). The S2 layer is usually thicker and determines fiber strength. This layer contains helical cellulose microfibers which form a certain angle in relation to fiber orientation. The various types of lignocellulosic fibers (coir, curauá, sisal, sawdust, bamboo etc) present significantly different contents of lignin, cellulose and hemicellulose, rendering very different adhesion and mechanical properties to the composites, as fibers containing higher lignin content tend to present improved compatibility towards non-polar polymers, since lignin is the most non-polar constituent of the fiber. Fiber properties may be altered by this microfibrillar angle, as well as by constituent concentration. Climatic and soil conditions also tend to affect fiber properties.
These fibers are not necessarily cultivated for polymer reinforcement purposes. In many cases the source of natural fibers used as reinforcement is waste or residues from other industries, such as architectural, furniture, wood manufacturing etc. When a commercial product is intended to be developed it is necessary to guarantee a continuous and homogeneous source of wood or fibers.

There are many residue materials generated in great quantities for which no large-scale application has yet been found, such as chromium-treated leather. The treatment of leather with products made of heavy metals such as chromium (chromium oxide) is still allowed in several countries. Leather treated with chromium oxide during the tanning process is used on a large scale in the footwear, automotive, garment and personal protective equipment (PPE) industries, generating a large amount of residue (Ambrósio et al., 2011). Leather fibers are mainly constituted of collagen.

Incorporation of natural fibers into polymers will result in composites with adequate properties only when a set of factors are judiciously controlled in their manufacture, such as moisture and particle size which may adversely affect composite properties. Fiber moisture should not exceed the 1 to 2% range, since excessive moisture can cause internal bubbles in the composites and fiber degradation. On the other hand if the fiber is completely dried, firing explosions can be caused during transport and feeding processing operations. Particle size studies indicate that very small particles may burn during processing, leading to degradation of the thermoplastic composites. Another important factor in the manufacture of these composites is that mixing of the polymer and the fibers should preferably be carried out in twin-screw extruders, which provide high level of mixing and homogeneity resulting in the production of high performance materials. A smooth screw elements profile, a high enough length to diameter ratio (L/D) and degassing zones are factors that must be accounted for. The feed process may also be a problem since the non-homogeneity of fiber surface topography may cause bridging in hoppers. In addition to temperature during extrusion and injection molding, other processing conditions must be carefully chosen to avoid excessive shearing of the fiber: low screw speed, flow rates and back pressure levels. Tools (molds and dies) should also be correctly designed to avoid excessive shear and stagnation regions. Excessive shearing will cause shear heating which consequently can cause fiber degradation.

It is necessary to create an adequate interface between the polymer matrix and the fibers, as in all composites, to generate good stress transfer during loading. Chemical polarity of the matrix will favor interaction with the fibers, since cellulose has a large number of –OH groups on the surface. When the polymer is non polar, such as PP and PE, compatibilizers and coupling agents must be used to improve adhesion between matrix and fibers. They are bifunctional molecules that can interact or even react with both the non polar matrix and with the very polar fiber. Another alternative is to promote chemical attack of the fiber surface, rendering it more irregular to facilitate adhesion to the polymer matrix. Among these the use of compatibilizer has shown to be more efficient in improving interface adhesion.

It should be pointed out that the polymer and the fibers are not the only components in the formulation of these composites. Depending on the final application and the composite, different levels of additives are incorporated: compatibilizers or coupling agents, internal and external lubricants, UV stabilizers, antioxidants, mineral fillers (such as talc, calcium carbonate and mica), flame retardants and smoke suppressants, biocides, acid scavengers, blowing agents, among others (Klyosov, 2007; Niska & Sain, 2008).

1.2 The Amazonian Phoenix Project concept

The Amazonian Phoenix Project is a very clever compilation of an integrated system of sustainable enterprises in Amazon lands, elaborated by Dr. Antonio Donato Nobre, during

his long periods of research in the Rain Forest. Further details can be obtained from the author (Nobre, 2006). The concept of recovery and protection of the Amazon Rainforest of the Phoenix Project can be better viewed in figure 1. The fibers and wood collected from plantations in already deforested areas could be used for biocomposite production in association with polymers from urban and industrial residues. The protection of the Forest would come from the demand for seeds of pioneer, thorny and noble specimens. It should be mentioned that exploitation of the Forest would not be stopped, but it would be done in a very sustainable and well-planned manner.

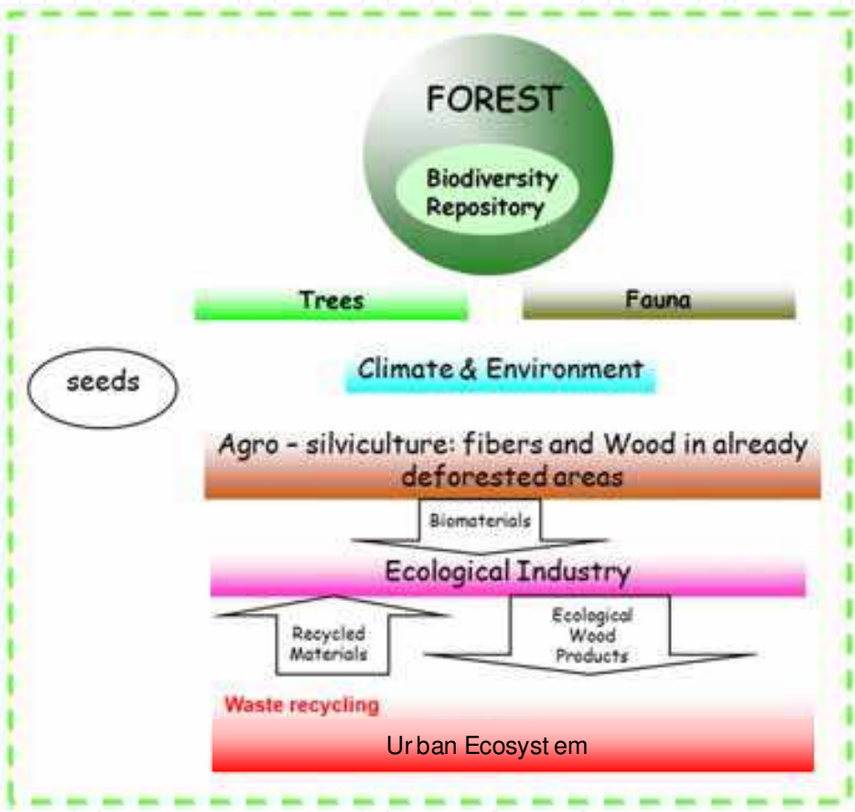


Fig. 1. Principal components of one of the production systems of the Amazonian Phoenix ecosystem (Nobre, 2006)

In the last decades, the Amazon Rain Forest has attracted worldwide attention due to its effect on the water cycles in America, the green house gas emissions caused by burnings, as well as the deforestation caused by the exploitation of timber, grain agriculture and more recently cattle ranching. It is known that deforestation in the Amazon Forest occurs naturally on a very limited scale as a consequence of storms and the Forest has developed very smart self-recovery mechanisms. There are pioneer plant specimens that grow very fast, under very extreme environmental conditions, such as direct sunlight and rain, forming a dense, secondary forest and creating protective conditions for the complex, diverse, massive and long lasting forest that needs this protection for recovery. The pioneer trees have the ability to close the gaps in the forest canopy. Unfortunately, this natural recovery process cannot keep pace with the very high rates of destruction caused by human activities.

Recent studies (Barbosa et al., 2003) have shown that a clever Short Cycle Agrosilviculture System can be used in deforested areas. These pioneer plants can be cultivated with the purpose of collecting fibers and wood of commercial interest, to be used, for example, in wood flour and fiber polymer composites. These pioneer trees include balsawood (*Ochrama Pyramidale*), marupa (*Simarolba amara*) and caroba (*Jacaranda copaia*), surucucumira (Spathelia excelsa). To protect these plantations from young herbivores, they are surrounded by thorny plants such as curaua (*Ananas erectifolius*), pupunha palm (*Bactris gasipae*) and tucum palm (*Astrocaryum vulgare*), just to cite a few among dozens of specimens that can be planted. The planted pioneer trees can be harvested after 2-3 years and be used in several applications including furniture, structural and architectural buildings, biofuels, papers; most of these of very high value added. All these industrial processes will generate residues and byproducts which can be used as fillers and reinforcement in plastics.
As materials engineers and scientists, our role in this project is to evaluate the use of the aforementioned useful specimens in the development of polymer biocomposites. The basic idea is to mimic the regenerative dynamics of the natural ecosystem in Short Cycle Agrosilvicultural Production areas, utilizing a variety of technologies to transform raw wood and fibers from these fast growth native plants into a variety of of high value added materials.

2. Case studies

2.1 Balsa wood polymer composites

As mentioned before, balsa wood (BW) is one of the very fast growing pioneer native species in a plantation on degraded pastureland near Manaus, AM, Brazil (Barbosa et al., 2003). Balsa wood is a very light weight wood and the residues from the processing of sticks, blocks and sheets for model airplanes and building models were used. In this context, balsa wood fiber composites were produced with two polypropylenes as matrices: a virgin (vPP) and a recycled one (rPP), both with MFI = 3.5 g/10min. The BW content was varied from 10 to 30 wt%.
Polypropylene is used as matrix in lignocellulosic fiber composites because of its relatively low melt temperature (165 °C) which allows processing within the limits imposed by the fibers. However, PP is incompatible with these fibers, because of its non polar character against the polar character of the fiber constituents. These differences result in poor adhesion between the phases, with high surface tension, rendering an incompatible system. As compatibilizer Orevac C100, a 1% maleic anhydride grafted polypropylene from Atofina, was used at 2 wt% fixed level.
Mixing was performed in a corotating twin-screw extruder from Imacom, Brazil, (L/D = 40 and a diameter of 30 mm) with a screw profile adequate for natural fiber polymer composites.
The thermal stability of the fiber was evaluated by Thermo Gravimetric Analysis (TGA), where the weight of the sample was monitored as a function of the temperature at a heating rate of 20 °C/min. Figure 2 presents the TG curve and its derivative (DTG). It can be observed that the degradation process of balsa starts at around 200 - 220^{o}C. Therefore, the temperature profile throughout the extruder screw zones was set from 140 °C to 185 °C, from the feed to the metering zone.
The morphology of balsa wood flour can be seen in figure 3, parallel and normal to tree growth. The Scanning Electron Microscopy used was a Zeiss Model Stereoscan 440. A very opened cell structure can be seen, which explains the very low density of balsa wood.

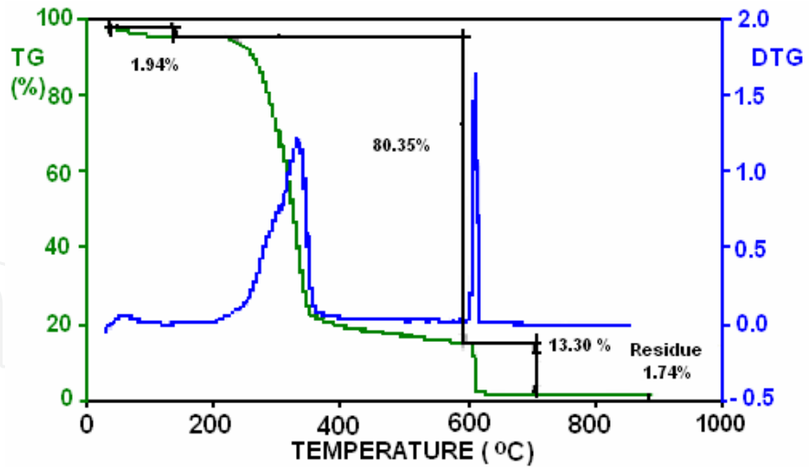


Fig. 2. Thermo gravimetric analysis of balsa wood (Marinelli et al., 2008)

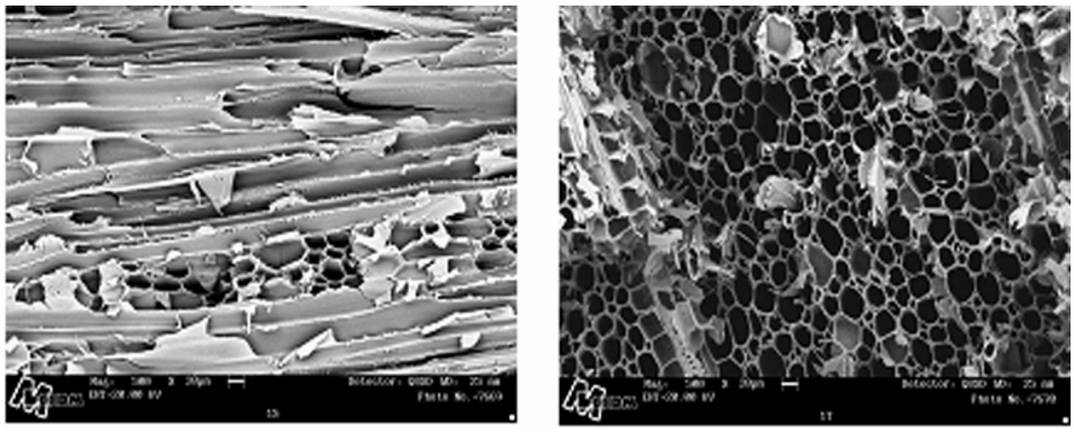


Fig. 3. SEM observation of (b) Longitudinal and (c) Transversal morphology (Marinelli et.al, 2008, Branciforti et al., 2009, Nobre et al., 2009)

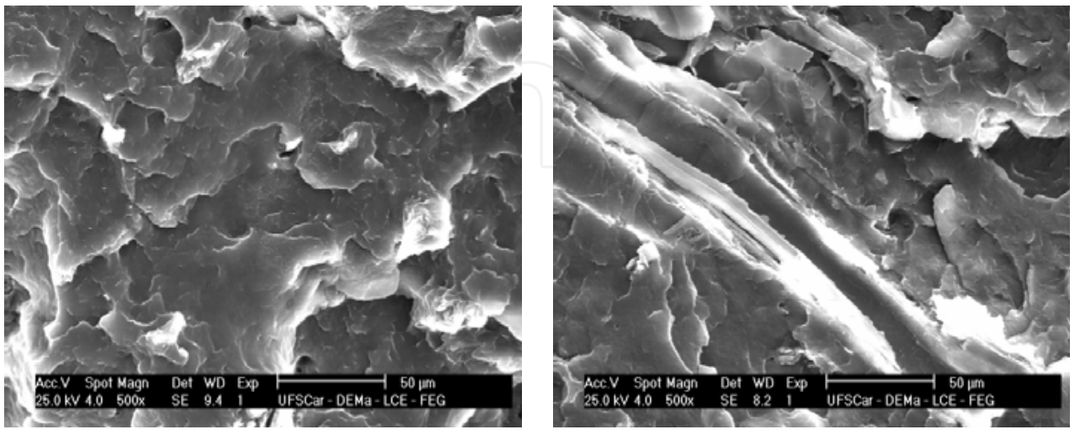


Fig. 4. SEM characterization of the transversal and longitudinal surface of the compatibilized PP/Balsa Wood Composites (80/20) showing the good interface between the fiber and the polymer matrix (Nobre et al., 2009)

Figure 4 presents the SEM morphology of the composite sample vPP/PPgMA/BW (78/2/20), cryogenically fractured and sputtered with gold. The SEM used was FEG from Phillips, model XL30. A very good interface between the fibers and the PP matrix can be observed, since no gaps are seen. This is a very interesting result, especially if we consider that the sample was cryogenically fractured.

Figure 5a and 5b show the mechanical properties of the PP/BW composites under flexural and tensile loading. Both flexural and tensile moduli increase with fiber content. The same trend is observed for flexural strength. The tensile strength of the samples did not change very much. A decrease in this property would be expected if no compatibilizer were used. The amount used was only enough to keep this property similar to that of pure PP. Higher amounts of PPgMa should be used if higher values are desired. Further discussion on compatibilizer level effects is presented in the next case studies.

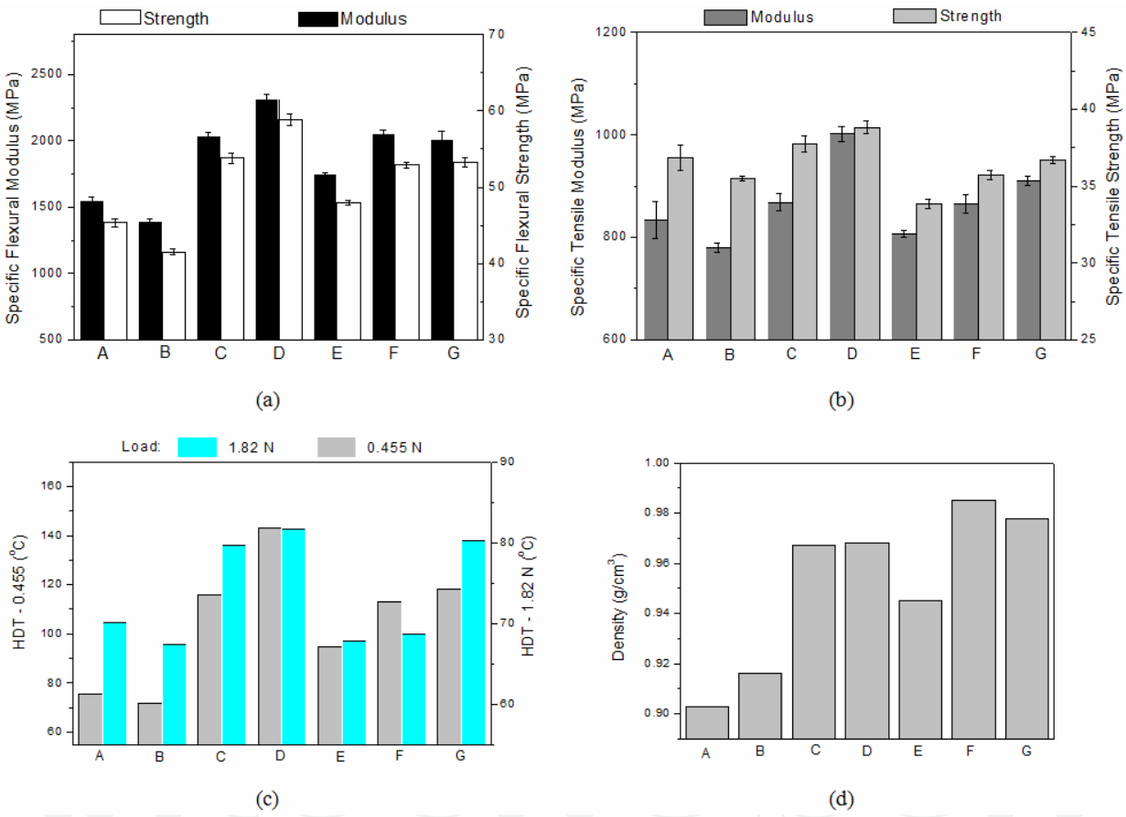


Fig. 5. (a) Flexural Properties; (b) Tensile Properties; (c) Heat Deflection Temperature and (d) Density of the PP/BW composites. Legend: **A**- vPP; **B** – rPP; **C** – vPP/PP-g-AM/BW (78/2/20); **D** - vPP/PP-g-AM/BW (68/2/30); **E** - rPP/PP-g-AM/BW (88/2/10); **F** - rPP/PP-g-AM/BW (78/2/20); G –vPP/rPP/PP-g-AM/BW (39/39/2/20), (Nobre et al., 2009)

Figures 5c and 5d present the Heat Deflection Temperature (HDT) and the density of the composites. HDT is related to the thermal and dimensional stability of the composite under load. At the lower load (0.455 N) HDT of both vPP and rPP increased with increasing BW content. At the upper load (1.82 N) the same behavior was observed for vPP and blend composites, but no changes were observed for the composites containing rPP. All the density values of the samples remained lower than 1g/cm^3, even though a good interface of

the composite phases was observed. Wood flour polymer composites may present density near the density of the cell walls of the wood flour, 1.45 -1.50 g/cm^3, since the wood and fibers are compressible during processing due to the high levels of shear and pressure, causing the hollow cell fibers to collapse or the hollow cells may become filled with polymer or low molecular weight additives. This increase in density was not observed and the low density character of balsa wood fibers was predominant.

Three results must be underlined in this case study: 1) the recycled PP reinforced with 20 wt% balsa wood fibers has presented the same properties of virgin PP reinforced with the same fiber content; 2) all the samples presented a density lower than 1g/cm^3 associated with a very good interface between the fibers and PP matrices and 3) The 30 wt% BW composites have mechanical properties comparable to wood flour PP composites (Niska & Sain, 2008). These results reflect important contributions of recovering recycled PP properties producing light weight composites using balsa wood residues.

2.2 PP/Sawdust composites

Sawdust is a wood processing residue from consumer good manufacture, which is commonly very little used or disposed of. The most common method of disposal is burning, which is economically little feasible as well as environmentally unfriendly. Some countries, mainly European, have therefore created laws that limit burning of cellulose residues.

Incorporation of sawdust in polymers has become a feasible possibility in obtaining products with suitable characteristics for use as a substitute for wood or in applications of important ecological appeal. The sawdust used in this study was waste from a sawmill "indústria Madeireira Uliana Ltda" (Tietê, Brazil) and despite the fact that this waste was ground in a grinder equipment with a 0.8 mm screen to adjust particle size, large dispersion in particle shape and size was observed. Micrographs of the sawdust particles are shown in Figure 6, which shows they are very non-homogeneous in shape and size (Bettini et al., 2008).

Sawdust was collected from a storage silo containing a mix of several Brazilian timber types: pink cedar (*Cedrela* sp), arana cedar (*Cedrelinga cateniformis*), ipê (*Tabebuia* spp), angelim (*Hymenolobiem petroeum*), marupá (*Simarolba amara*), loro vermelho (*Nectanda rubra*), cumarú (*Dipterysa odorata*), and jatobá (*Hymenoea* sp). The relative amounts of the timber types have not been determined.

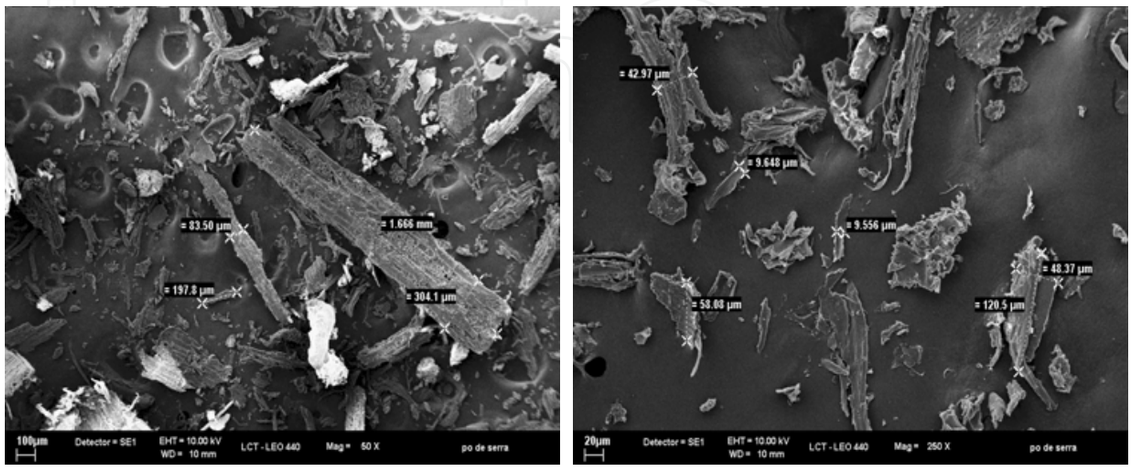


Fig. 6. SEM micrographs of sawdust particles

If we consider that sawmill activities in Amazon lands have a very low productivity level, this study also reflects a very important contribution to the Amazonian Phoenix Project concept, since there are huge sawdust amounts as byproducts of wood exploitation and sawmill activities in the Amazon Forest that may be used for the production of wood polymer composites.

Several studies have been undertaken by our group for better insight into the role of compatibilizers and coupling agents on the adhesion in PP/lignocellulosic composites, as well as the interaction with processing additives, such as lubricants.

Treating the fibers with silane coupling agents has shown little effect on mechanical properties. The main contribution showed to be related to water absorption.

2.2.1 Processing and compatibilizer levels effects

Figure 7 presents micrographs of PP/sawdust composites, with no compatibilizer and containing 3 wt% compatibilizer (PPgMA), extruded in a twin-screw extruder coupled to a Haake torque rheometer in the presence of lubricants.

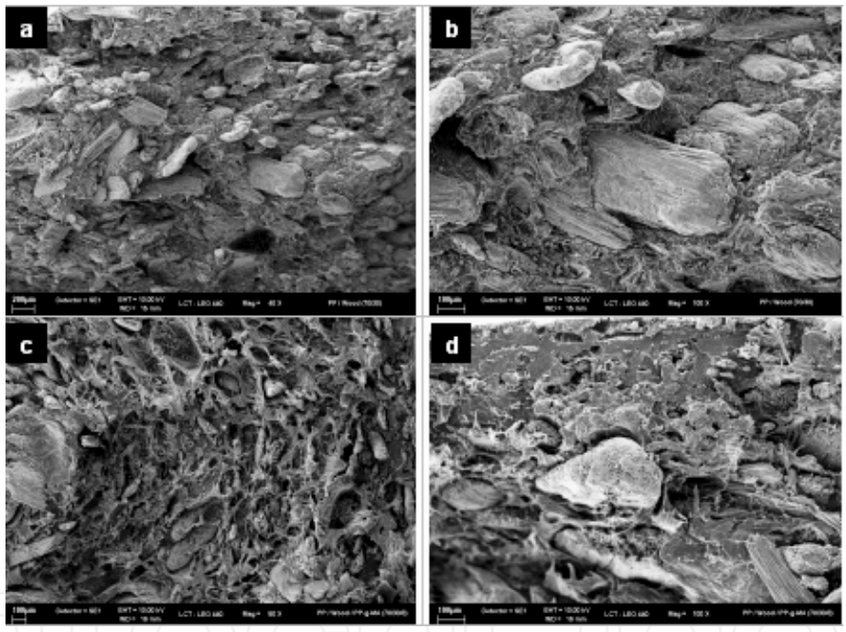


Fig. 7. SEM micrographs of the fractured surfaces of the PP/sawdust/PPgMA compounds: a) and b) 70/30/0, c) and d) 67/30/3 (Bettini et al., 2008)

Figures 7a and 7b show fiber pull-out of the matrix, observed throughout almost the entire sample, indicating poor adhesion between the fibers and the polypropylene. The fractured surfaces in Figures 7c and 7d show pronounced deformation of the polymer matrix and significant reduction in fiber pull-out, indicating the compatibilizer likely affects interface, reducing interfacial tension and improving adhesion.

During Haake extrusion, samples without compatibilizer were difficult to process as they broke easily on exiting the die. Furthermore, these samples presented white spots when injection molded.

Composites were prepared according to a central composite design in which sawdust and PPgMA contents were varied and tensile and flexural properties were assessed. Superior

and inferior concentration levels were 40 and 60 wt% for sawdust and 3 and 5 wt% for PPgMA, respectively.

Multivariate analysis of the properties flexural modulus, tensile strength and % tensile elongation at break of samples, processed in the Haake torque rheometer showed that incorporated sawdust content (independent variable) was the only variable affecting these properties.

The fact that increasing PPgMA content did not affect the dependent variables indicates that PPgMA did not act as compatibilizer, contradicting the preliminary tests, as well as previous investigations (Keener et al., 2004; Oksman & Clemon, 1998; Suarez et al., 2003).

This behavior might arise from two factors: the presence of retained moisture in the pellet, which might have settled at the PP/sawdust interface, impairing compatibilizer effectiveness, and inadequate mixing of the compound. The first factor might be explained by the lack of a degassing zone in the extruder coupled to the Haake rheometer as well as the fact that the extrudate was cooled in a water bath. With regard to inadequate mixing, this might have occurred because of the low L/D ratio (about 28) of the Haake rheometer for composite processing.

It was therefore decided to reprocess the composite pellets in a ZSK 25 Werner & Pfleiderer extruder with two degassing zones. The results were, then, submitted to multivariate analysis and the investigated dependent variables (tensile strength, % elongation at break and flexural modulus) could be fitted in relation to the independent variables (sawdust and compatibilizer content). The results showed that both sawdust and compatibilizer content affected the investigated dependent variables.

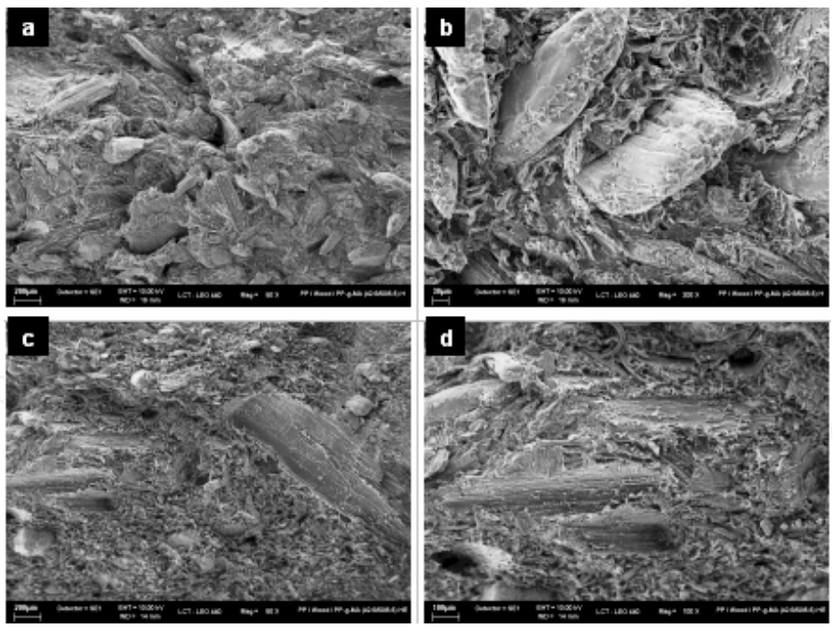


Fig. 8. SEM micrographs of the fractured surfaces of the PP/sawdust/PPgMA compounds (42.5/50/5.5): a) and b) processed in the Haake torque rheometer only (H); c) and d) processed in the Haake torque rheometer and subsequently in the Werner Pfleiderer Extruder (HE) (Bettini et al., 2008)

SEM analysis was performed on the surface of tensile fractured samples PP/Sawdust/PPgMA, 42.5/50/5.5, processed in the twin-screw extruder coupled to the

Haake torque rheometer (H), extruded in the Haake and reextruded in the Werner Pfleiderer Extruder (HE). Figure 8 shows analyses of samples 10H and 10HE.
Comparison between samples H and HE, shown in Figure 8, shows increased deformation of the matrix of the samples reprocessed in the extruder, as well as improved fiber adhesion. The samples processed in the Haake torque rheometer only, showed a less deformed matrix, a greater amount of voids, as well as poor fiber adhesion. This behavior is an indication of the improved adhesion brought about by reprocessing in the extruder due to more adequate mixing and moisture removal.
Analyses of the fractured surfaces (Figure 8) and mechanical properties (Figure 9) show the importance of processing conditions on composite properties. As can be seen both PPgMA and sawdust content influenced mechanical properties. The highest tensile strengths occurred at low sawdust and high compatibilizer contents, indicating that at higher sawdust contents the amount of incorporated compatibilizer should exceed the investigated levels.

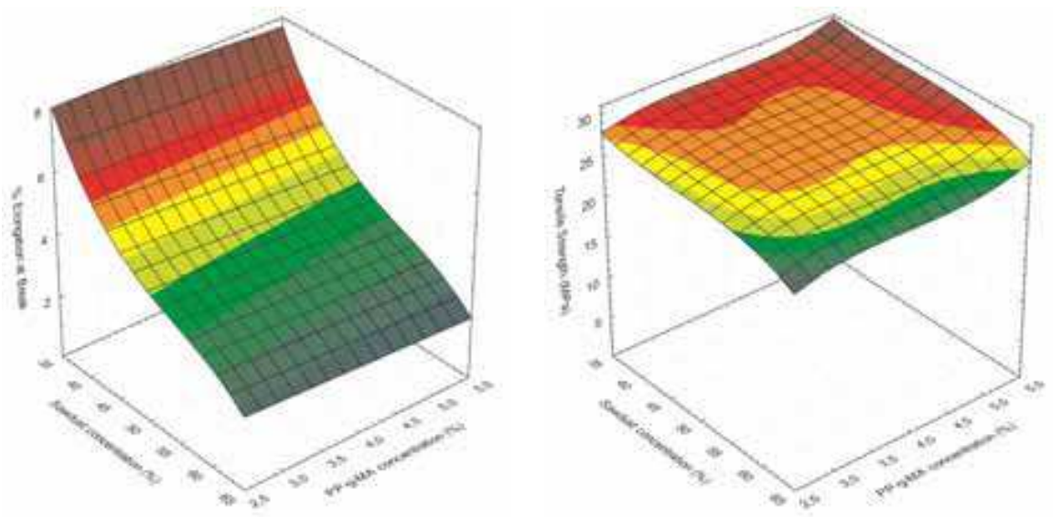


Fig. 9. Effect of sawdust and compatibilizer (PPgMA) concentrations on % elongation at break and tensile strength (Bettini et al., 2008)

2.2.2 Interactions between compatibilizer and lubricants

The wood plastic industry, due to economical reasons, uses expressive loading levels of sawdust, as high as 60 wt%. Hence, a drawback in the processing of these composites is the very high viscosities and, consequently, also very high pressures required, which sometimes can become impracticable.
The addition of lubricants may overcome this difficulty. There are a few works in the literature evaluating lubricants for HDPE based wood composites (Harper and Wolcott, 2003; Li and Wolcott, 2004, 2005, 2006; Santi el al., 2009). In these works, there is some evidence that the compatibilizer, which was maleic anhydride grafted polyethylene (PE-g-MA), acted not only as a compatibilizer, but also as an internal lubricant. Though, depending on the nature of the lubricant, the compatibilization effect may be reduced, Li and Wolcott (2006) did not observe a decrease in compatibilizer efficiency when an ester-based lubricant was used. On the other hand, zinc stearate lubricant reduced PE-g-MA activity.
In a previous study, Bettini et al. (2008) observed that conventional PPgMA concentrations in PP/sawdust composites, when in the presence of ester-based lubricants, were insufficient

to achieve the required mechanical properties. Santi et al. (2009) also noticed that the mechanical properties of HDPE/sawdust composites can be reduced when ester-based lubricant and PE-g-MA compatibilizer are used simultaneously.

The influence of an ester-based lubricant on the compatibilization effect of PP/sawdust composites, both in the presence and absence of PPgMA, were assessed through the analysis of mechanical and rheological properties (Bettini et al., 2010b).

Table 1 presents tensile strength and elongation at break results. Polymer and sawdust contents were maintained at 70 and 30 wt%, respectively, and the sawdust used was ground pinus wood. Increase in sawdust content, without the presence of the compatibilizer, resulted in decrease in both properties, due to the incompatibility between polypropylene and sawdust.

With the introduction of lubricant in the polymer/sawdust system (formulations 10 and 11) tensile strength also decreased. This behavior was expected, since the lubricant does not act as a compatibilizer. Furthermore, the remaining lubricant in the polymer matrix after processing might contribute to the decrease in composite strength due to its low molar mass.

On the other hand, introduction of compatibilizer PPgMA in the composites (formulations 8 and 9) resulted in a noticeable increase in tensile strength in comparison to the non-compatibilized composite. However, at PPgMA contents exceeding 6 wt%, this trend was not observed, indicating that this concentration might be the optimum for the investigated system.

The unexpected decrease in tensile strength when compatibilizer was increased to 8 wt% might be an indication that this additive has reached a limit in the system and is in excess, leading to the formation of an extra PPgMA phase, thus reducing its efficiency.

These results imply that the introduction of compatibilizer in these composites is crucial for the achievement of high tensile strength. However, addition of lubricant to the compatibilized system reduces tensile strength. This suggests that, possibly, the lubricant interacts with the compatibilizer and reduces its efficiency.

The response surface plot of the influence of compatibilizer and lubricant concentrations on tensile strength is presented in Figure 10.

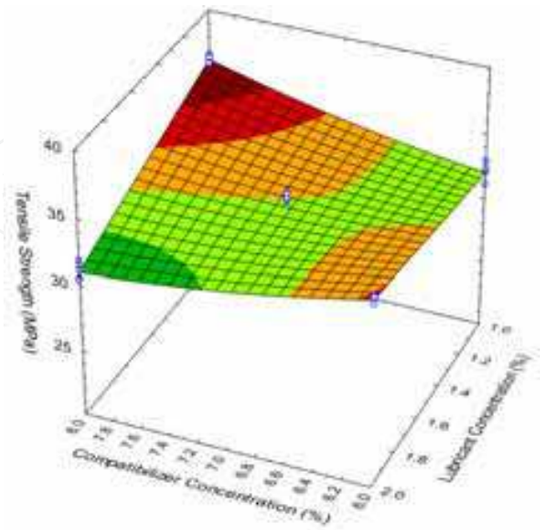


Fig. 10. Effect of compatibilizer (PPgMA) and lubricant (Struktol TPW 113) concentrations on tensile strength (Bettini et al., 2010b)

As can be seen in Figure 10 and Table 1, tensile strength is affected by all variables. For instance, at the lower lubricant level, increasing the amount of the compatibilizer leads to an improvement in tensile strength. However, an opposite trend is observed at higher lubricant level. Despite the assumption that the lubricant could decrease compatibilization efficiency, the higher its amount, the greater are the chances of anhydride or acid groups to interact with sawdust hydroxyl groups. The lowest tensile strength of the composite is achieved when both additives are at their highest level (i.e. 8 wt% compatibilizer and 2 wt% lubricant). As mentioned earlier in this text, there is evidence that when at high concentration levels, the compatibilizer may form a distinct phase in the composite, decreasing compatibilization efficiency.

	C_C (%)	C_{TPW} (%)	Tensile Strength (MPa)	Elongation at break (%)
1	6	1	32.15 ± 0.64	2.76 ± 0.29
2	8	1	36.32 ± 0.26	2.99 ± 0.24
3	6	2	34.33 ± 0.24	3.19 ± 0.20
4	8	2	31.16 ± 0.62	3.24 ± 0.43
5	7	1.5	33.41 ± 0.30	3.11 ± 0.26
6	7	1.5	32.12 ± 0.44	3.22 ± 0.48
7	0	0	30.55 ± 0.71	2.57 ± 0.40
8	6	0	40.35 ± 0.78	4.53 ± 0.48
9	8	0	38.68 ± 0.36	4.28 ± 0.51
10	0	1	28.90 ± 0.42	5.40 ± 1.07
11	0	2	27.91 ± 0.12	4.28 ± 0.78
12	0	0	31.42 ± 0.3	> 500

Table 1. Tensile test results of PP/30 wt% pinus sawdust composite with varying contents of compatibilizer (Cc) and lubricant (C_{TPW}) (Bettini et al., 2010b)

Therefore, if there is less compatibilizer available, due to the formation of a new phase, and if lubricant concentration is at its highest level, which increases the chances of reaction between these additives, then a decrease in compatibilization is expected. This may be a result of the reduced availability of the PPgMA anhydride and/or acid group to interact with the sawdust hydroxyl groups, which might explain the low tensile strength shown by Formulation 4.

Despite the adverse effects of lubricants on the adhesion in PP/sawdust composites, problems encountered at the die exit when high levels of sawdust are incorporated can be overcome by adding external lubricants. Compatibilizer should therefore be used at amounts suited to the required purpose.

2.3 PP/Coir composites

As mentioned, disposal of green coconut husks after consumption of coconut water has great impact in tropical countries like Brazil and has become a sanitation and a public health

problem. Coir is extracted from the green coconut husks and may be used in a wide spectrum of applications such as car seat stuffing, mats, brushes, rope, insulation, as well as reinforcement in polymers.

Coir consists of lignocellulosic fibers, obtained from the mesocarp of the coconut fruit and makes up about 25 % of the nut. Some characteristics of coir are that the fibers are more flexible than glass fiber, nontoxic and can be chemically modified (Tomczak et al., 2007).

The use of coir fiber as reinforcement in polymers has been the object of several works. The most commonly used matrixes include polypropylene (Bettini et al., 2010, 2011; Haque et al., 2009; Ishizaki et al., 2006 Islam et al., 2010; Leblanc et al., 2006; Rozman et al., 2000; Santos et al., 2008; Wambua et al., 2003), natural rubber (Geethamma et al., 1995, 2005) and polyester resins (Monteiro et al., 2006; Prasad et al., 1983; Rout et al., 2001).

Extracted coir usually consists of long fibers and has therefore improved reinforcement potential, as final size may be controlled by grinding. Figure 11 contains micrographs of coir used in our research.

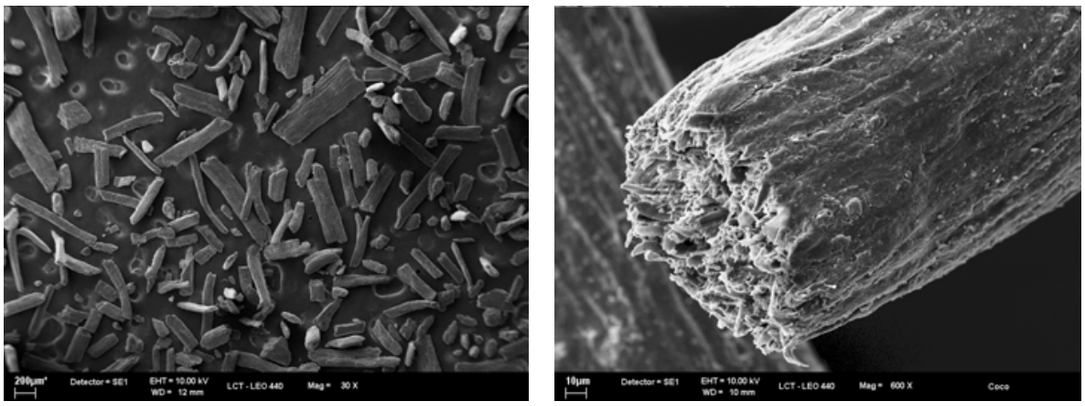


Fig. 11. SEM micrograph of coir fibers (Bettini et al., 2010)

PP/coir fiber composites present the same difficulties regarding adhesion as other lignocellulosic fibers due to the chemical differences between matrix and fiber. Therefore, compatibilizers should be incorporated into these composites. One of the most commonly used compatibilizer in PP/natural fiber composites is maleic anhydride grafted polypropylene (PPgMA).

The effect of lignin as compatibilizer in PP/coir fiber composites has also been investigated by Rozman et al. (2000). This investigation showed that the presence and increase in lignin content increased flexural properties; however no significant increase was seen in tensile properties when compared to the composites without this compatibilizer. For comparison sake PPgMA was also used as compatibilizer in this work and the composites compatibilized with PPgMA showed better mechanical properties than those compatibilized with lignin.

Based on the investigations regarding PP reinforced with sawdust, PP/coir composites were prepared according to a central composite design, employing coir levels of 20 and 40 wt%, and PPgMA levels of 4 and 8 wt%. Adhesion was only achieved in the presence of PPgMA as shown in the SEM micrographs (Figure 12). Figure 13a and Figure 13b present the surface responses of tensile strength and elongation at break, when the independent variables, coir content and compatibilizer content, were varied (Bettini et a., 2010).

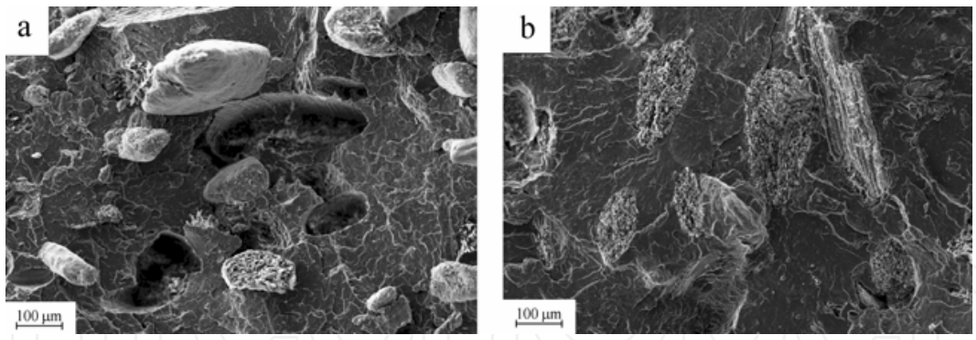
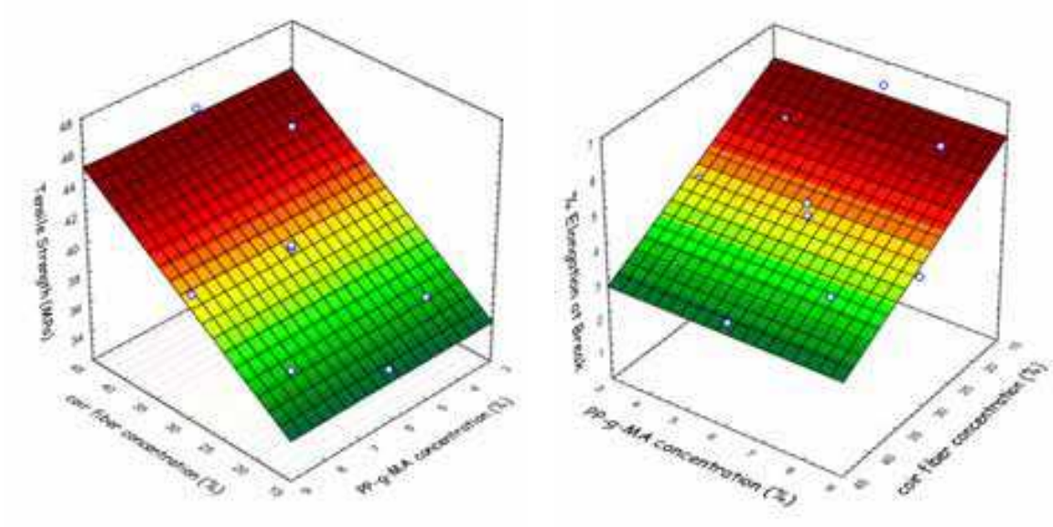


Fig. 12. SEM micrographs of tensile fractured surfaces of 30 wt% CF composites with (a) no PPgMA and (b) 6 wt% PPgMA (Bettini et al., 2010)



(a) (b)

Fig. 13. Effect of fiber and compatibilizer content (PPgMA) on (a) tensile strength and (b)% elongation at break of the composites (Bettini et al., 2010)

It can be seen in these figures that in the presence of compatibilizer increase in coir fiber content results in increase in tensile strength and a reduction in elongation at break and that the increase in amount of compatibilizer (PPgMA) does not alter the investigated properties. This analysis shows the addition of compatibilizer is important for reducing interfacial tension, with consequent increase in adhesion between the phases. When coir fibers were added to PP without the presence of compatibilizer, tensile strength was seen to reduce with increasing fiber content. So, although the effect of compatibilizer content was not observed, its presence is necessary. It is also verified that for the investigated coir fiber contents and processing used the lower level of PPgMA content would be sufficient to achieve good adhesion.

Compatibilizer did not affect the properties in the investigated range, which means that smaller amounts of compatibilizer could have been used in the PP/coir composites. Some factors may affect this analysis in relation to the concentration of the same compatibilizer in PP/sawdust composites. Different fibers have different contents of lignin, cellulose and

hemicellulose, as well as different structures. Moreover, no ester lubricant, which has already shown to reduce compatibilizer efficiency, was added to the PP/coir composites.
Compatibilized and non compatibilized polypropylene/coir fiber composites were also submitted to fatigue testing. Fatigue life of these composites was also compared with that of pure polypropylene.
Fatigue tests were performed in an MTS 810A servo hydraulic universal testing machine at a testing frequency of 6 Hz. S-N curves were obtained with maximum applied force varying from 1400 N to 850 N; considering that compressive loads could not be applied due to occurrence of buckling, minimum force was maintained at 50 N, resulting in cyclic loadings with the ratio between minimum and maximum stress (R) approximately zero. The results allowed calculating the applied maximum stress as a function of number of cycles to failure. When no fracture occurred, failure was considered when specimen displayed plastic deformation (Bettini et al., 2011).
The data obtained from the fatigue tests allowed the construction of S-N curves presented in Figure 14, which shows that the fatigue behavior of pure PP is different from the behavior of the two composites. These composites exhibit similar behavior, but the compatibilized composite presented longer fatigue life at a same loading.
Results from Figure 14 indicate that incorporation of coir fiber reduces fatigue life of the composite when compared to pure PP, and that the compatibilized composite presents a higher number of fatigue cycles for the same applied stress amplitude, evidencing the need to use compatibilizer.

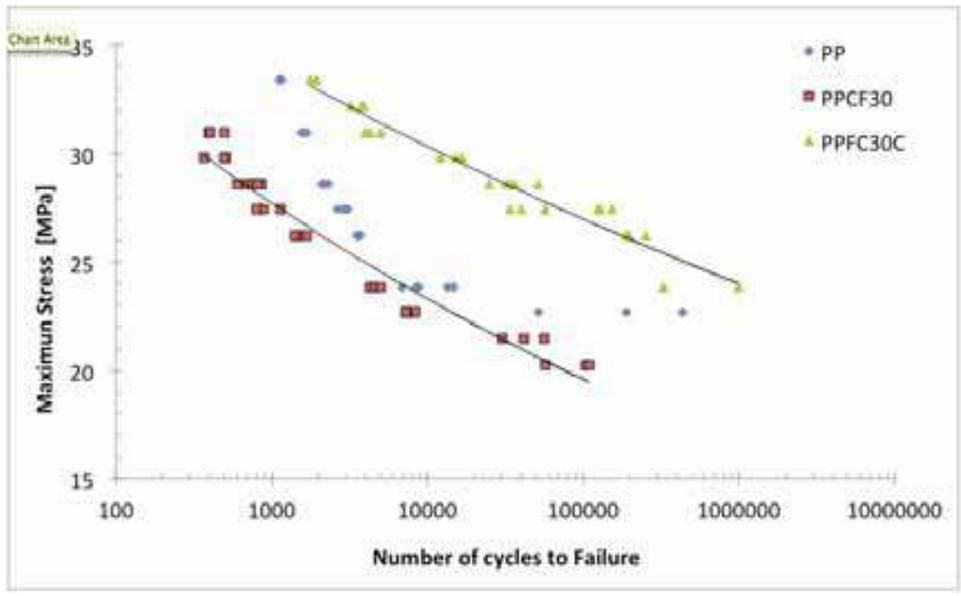


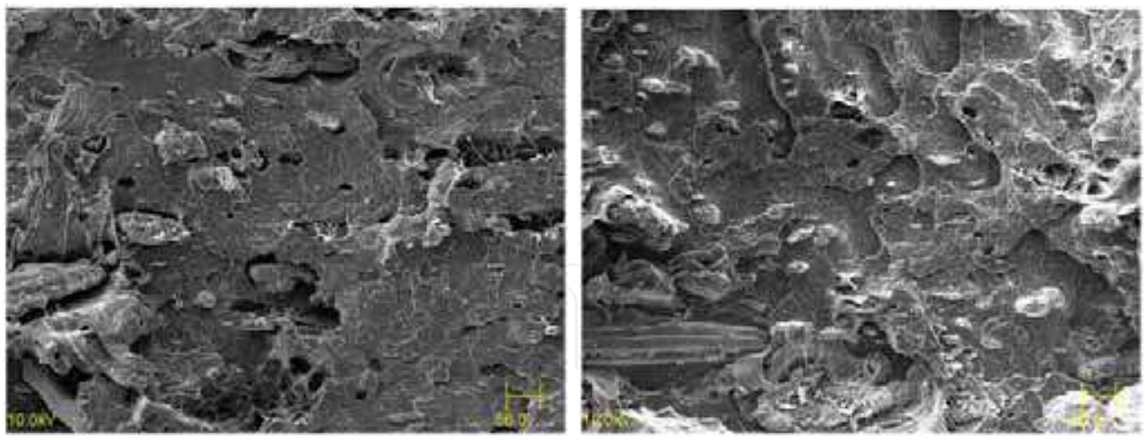
Fig. 14. S-N curves obtained in the fatigue tests, for pure polypropylene (PP), polypropylene /coir fiber composites without compatibilizer (PPFC30) and with compatibilizer (PPFC30C) (Bettini et al., 2011)

An optical pyrometer allowed observing that during the fatigue tests PP heated up to approximately 50°C and failed by yielding instead of rupturing. Initially the center of the specimen whitened, indicating polymeric chain alignment and microfibriling, followed by necking and finally yielding.

This phenomenon was observed in all fatigue tests on PP, however yielding was considerably less at the lowest tested stress (22.67 MPa), where the material, besides yielding, also fractured and the maximum temperature in this test was approximately 30°C.
Specimen heating at higher stresses can be explained by the low thermal conductivity of the polymer. In each cycle the heat generated by molecular friction is not readily dissipated. However, at lower loadings the generated heat is less in each cycle and therefore accumulated heat is less. This indicates that at higher stresses the main failure mechanism in pure polypropylene is thermal fatigue and at low stresses mechanical failure predominates. This change in behavior of the failure mechanisms is also noticed by the inflection of the fatigue curve of pure PP (Figure 14).
The compatibilized and non compatibilized polypropylene/coir fiber composite specimens displayed fracture without macroscopic plastic deformation. The maximum temperature recorded during the fatigue tests of the composites was 30°C, indicating preferential mechanical failure mode. This behavior might be explained by the reduced mobility of the polymer chains due to the presence of fibers, which leads to lower hysteresis and consequently less heat generation compared to pure PP.

2.4 PP/Bamboo fiber composites

Bamboo plantations are another abundant source of natural fibers. Bamboo has a very high strength/weight ratio. Brazil has the largest native giant bamboo plantations in the world, located in Acre, a State in the Southwest region of the Amazon. Approximately every 30 years these plants bloom and die, opening a clearing in the forest, which serves as a source of fires under lightning, contributing to the emission of greenhouse gases. The use of bamboo fibers in composites and laminates, before plant death, not only would avoid the emission of greenhouse gases to the atmosphere but would also serve as an alternative sustainable activity for the local population. There are about 300 different specimens of bamboo in Brazil, around 400 in the whole world. All these populations present the same natural cycle characteristic of flowering followed by death.
In this context an investigation was carried out regarding the effect of fiber size, compatibilizer (maleic anhydride grafted PP) and bamboo fiber content on the physical properties of polypropylene/bamboo fiber composites. Bamboo fibers used were ground waste generated during cutting, milling and finishing operations from a small bamboo laminating mill. Bamboo fiber levels of 20 and 40 wt% were employed and PPgMA levels of 1 and 4 wt%, against the 4 and 8 wt% levels used with coir fibers. As can be inferred from figures 15a and 15b, which show SEM micrographs of 20 wt% bamboo fiber composites with no compatibilizer and with 4 wt% compatibilizer, adequate adhesion could be obtained at 4 wt%. Lack of adhesion in composites without compatibilizer is evidenced in the SEM micrograph of Figure 15a, whereas in Figure 15b the fibers in the composite containing compatibilizer are seen to be well adhered to the matrix.
Except for impact strength, the behavior of all mechanical properties showed similar trends when comparing bamboo and coir fiber composites. Impact strength of coir composites with no compatibilizer increased at increasing coir contents above 20 wt%, whereas that of bamboo fiber composites decreased. It should however be mentioned that depending on composite nature (different fibers with different stiffness) and type of impact test, apparent impact strength may either increase or decrease with fiber content (Nielsen & Landel, 1994).



(a) (b)

Fig. 15. SEM micrographs of tensile fractured surfaces of 20 wt% BF composites with (a) no PPgMA and (b) 4 wt% PPgMA. (Bonse et al., 2010)

Comparing tensile strength of 40 wt% compatibilized with non-compatibilized bamboo fiber composite showed increase in tensile strength of 66% and with pure PP an increase of 34%. These values are similar to those obtained with the same amounts of coir fiber and compatibilizer (63% and 35%, respectively), despite the higher reported tensile strength and modulus of bamboo fiber (BF) in relation to coir (Suddell & Evans, 2005). When the same analysis is applied to flexural modulus the compatibilized BF composite showed an increase of 114% in relation to pure PP, against 78% obtained with coir. Thus fiber stiffness seems to have a stronger effect on composite flexural properties than on tensile properties.

In addition to compatibilizer and bamboo fiber content, the effect of fiber size on the physical properties of the composites was also assessed according to a 2x2x2 factorial design. Physical properties assessed were tensile and flexural modulus, tensile and impact strength, percent elongation at break, fatigue life and heat deflection temperature (HDT).

As-received bamboo waste fibers were first submitted to screening after which size distribution measurements by optical microscopy revealed an average equivalent diameter for the inferior and superior level fibers of 0.94 mm and 2.19 mm, respectively.

To assess the effect of fiber size, bamboo fiber and PPgMA content - and possible interactions - on the mechanical properties of the composites, multiple regression analysis was performed by means of software Statistica® on all data used to generate Table 2. The encountered R^2 values indicate that, except for impact strength, the model fits the data reasonably well. Results of the 2x2x2 experimental design are presented in Table 3.

Analyzing Table 3 and the statistically significant effects obtained, it can be concluded that the effect of bamboo fiber size showed to be significant on tensile strength, however to a lesser extent than compatibilizer and BF content, whose effects have already been discussed. Increasing fiber length is expected to increase tensile strength. Fiber ends support less load than the remaining parts of the fiber, so the longer the fiber, the higher its ability to support load. However, reinforcement effect will be small when fiber length is smaller than a certain critical length. This might have been the case here. Fibers were apparently also randomly distributed in the matrix with no preferential orientation in relation to applied load. Fiber size would be more efficient in distributing and transferring applied tensile stress if fibers were aligned parallel to the applied load.

independent variables			dependent variables					
D_{eq} (mm)	**BF (wt%)**	**PPgMA (wt%)**	**TS (MPa)**	**ε_{break} (%)**	**E (GPa)**	**IS (J/m)**	**FM (GPa) 0.5%**	**σfl (MI**
0.94	20	1	37.5±0.3	5.6±0.5	3.15±0.11	11.2±1.6	1.96±0.11	40.8±
0.94	20	4	40.1±0.2	9.0±0.9	3.33±0.12	11.6±0.3	2.15±0.06	42.6±
0.94	40	1	38.6±0.1	2.4±0.2	4.64±0.06	10.4±0.6	3.45±0.14	52.8±
0.94	40	4	46.6±0.5	5.0±0.2	4.68±0.09	10.6±1.8	3.28±0.19	53.1±
2.19	20	1	38.8±0.3	6.6±0.6	3.25±0.12	9.3±0.2	2.42±0.08	37.1±
2.19	20	4	40.2±0.3	8.7±1.1	3.16±0.08	12.3±0.2	2.51±0.10	40.3±
2.19	40	1	40.9±0.2	2.8±0.3	4.63±0.07	10.0±0.9	3.27±0.05	46.1±
2.19	40	4	45.0±0.2	4.3±0.2	4.52±0.09	13.4±0.6	3.57±0.09	54.0±
neat PP			34.1±0.3	>500	2.22±0.11	12.8±1.3	1.80±0.06	31.4±

D_{eq}: equivalent diameter; TS: tensile strength; ε_{break}: strain at break; E: tensile modulus; FM: flexural modulus at 0.5% strain; σflex: flexural stress at 2% strain; IS: notched Charpy impact strength

Table 2. Results of the different tests: average of five specimens, except for fatigue and HDT (three specimens). (Bonse et al, 2011)

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	a_0	a_1	a_2	a_3	a_4	a_5	a_6	a_7	R^2
TS (MPa)	40.97	0.26	1.81	1.99	--	-0.65	1.02	-0.34	0.99
ε break (%)	5.55	--	-1.91	1.21	--	--	-0.30	--	0.935
E (GPa)	3.92147	-0.03186	0. 69766	--	--	-0.0518	--	--	0.9815
FM (GPa)	2.8256	0.11715	0.056539	0.05339	-0.088.01	0.04659	--	0.07203	0.9653
σflex (MPa)	45.85	-1.46	5.64	1.66	--	1.13	0.41	0.77	0.9692
Fatigue (Log_{10} cycles to failure)	3.18	--	0.24	0.34	--	-0.08	0.23	-0.08	0.9634
HDT(°C)	100.44	-2.05	13.77	--	1.89	-1.15	--	--	0.9803

TS: tensile strength; ε_{break}: strain at break; E: tensile modulus; FM: flexural modulus at 0.5% strain; σflex: flexural stress at 2% strain

Table 3. Coefficients of the regression model $\mathbf{a_0+a_1x_1+a_2x_2+a_3x_3+a_4x_1x_2+a_5x_1x_3+a_6x_2x_3+a_7x_1x_2x_3}$ used for the different properties (x_1: fiber size; x_2: BF content; x_3: PPgMA content). (Bonse et al., 2011)

The effect of BF size on strain at break, i.e. ductility, was not significant. As expected and already discussed increasing BF content reduces ductility, as fibers impose restraint on molecular mobility. Not surprisingly this variable also showed to be the most important for the apparent elastic modulus. On the other hand compatibilizer has no significant effect on elastic modulus and tends to increase ductility. The interactions promoted by PPgMA likely allowed improved molecular mobility and flow.

Properties obtained in bending mode (HDT, flexural modulus and strength) are strongly affected when fibers are perpendicular to the applied load. The bamboo fibers however were apparently randomly distributed and the effect of BF size was hence less pronounced than the effect of BF content on the flexural properties.

With regard to fatigue life fiber size showed no significant effect, in contrast to fiber and compatibilizer content which tend to maximize fatigue life with increasing contents, similar to their effect on tensile strength. It should be mentioned that fatigue tests were performed in tensile mode.

It can thus be concluded that incorporation of compatibilizers is essential for achieving higher tensile and flexural strength than those of the pure polymer; however, increase in stiffness, proportional to elastic modulus, is obtained even for the non compatibilized composites.

2.5 PVB/wood flour composites

The composites developed with the aforementioned thermoplastics matrixes are usually rigid and generally have low flexibility due to the characteristics of the matrix. Nevertheless, some natural fiber composite applications require rubbery behavior or high flexibility, such as curved coatings, soles and insoles for shoes.

A thermoplastic matrix that fulfills this requirement is an amorphous random copolymer from poly(vinyl butyral) (PVB), produced from the condensation reaction of polyvinyl

alcohol with n-butyraldehyde in the presence of an acid catalyst (Dhaliwal & Hay, 2002; Valera & Demarquette, 2008). The properties of this copolymer, such as elasticity, toughness and adhesion to different substrates depend on the ratio between the monomers in the copolymer. The main application of PVB is in laminated safety glass, particularly in the automotive, aerospace and architectural glass sectors. PVB used in sandwiched laminated glass has between 10% and 35% alkyl phthalate, dibutyl sebacate, triethylene glycol bis (2-ethylhexanoate) or dihexyl adipate plasticizer (El-Din & Sabaa, 1995; Jeong et al., 1998; Saflex®, 2010; Valera & Demarquette, 2008). Due to its molecular structure and the presence of plasticizers, PVB used in automotive glass has properties similar to those of an elastomer (Cha et al., 1998). The production of PVB laminated glass generates large volumes of residues that have not yet been widely destined for recycling.

2.5.1 Preparation of the composites

Prior to extrusion, the leather fibers, wood flour and PVB flakes were dried in an oven at 70°C for 4 hours to remove moisture and prevent degradation. The two composites (PVB/leather and PVB/wood flour) were mechanically mixed in the solid state at three mass proportions and processed in an extruder equipped with a 45-mm diameter Maillefer screw, L/D = 25, and a pineapple mixing element in the metering zone. A flat die (150.0 mm wide, 2.0 mm thick) was used to obtain flexible sheets. The temperature profile was 170/170°C/175°C in the extrusion barrel and 180°C in the extrusion head. After extrusion, the composite sheets were compression-molded at a temperature of 180°C and pressure of 20 ton to improve surface finishing. A final thickness of 3.50 mm was obtained for the samples to be used in mechanical tests. Figure 15 shows a schematic drawing of the extruder screw used for preparing the flexible PVB/leather and PVB/wood flour composites.

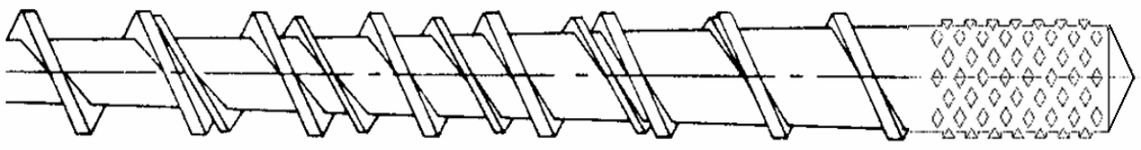


Fig. 15. Maillefer screw with pineapple mixing element in the metering zone (Rauwendaal, 2001)

Results of mechanical properties obtained with flexible composites of PVB/leather and PVB/wood flour, for application as coatings in products such as shoes, cars and architectural design, are presented next.

Figure 16 presents a micrograph of a cryogenically fractured surface of the PVB/wood flour composite sample. Sites exhibiting good interfacial adhesion with continuity of the interface are observed. As previously mentioned it is usually difficult to achieve good adhesion between a nonpolar thermoplastic matrix and natural fibers. One way to improve interfacial adhesion and obtain continuous surfaces in thermoplastic composites containing wood fibers is to use coupling agents or compatibilizers such as maleic anhydride grafted PP (PPgMA) (Oksman & Clemons, 1998). The good adhesion at the interface of the PVB/wood flour composites without the addition of coupling agents or compatibilizers may be due to improved interaction between the PVB matrix and the wood fibers. Just as in the leather fibers, some molecules that compose the chemical structure of the various components of wood contain -OH groups, which may have created hydrogen bonds with PVB molecules.

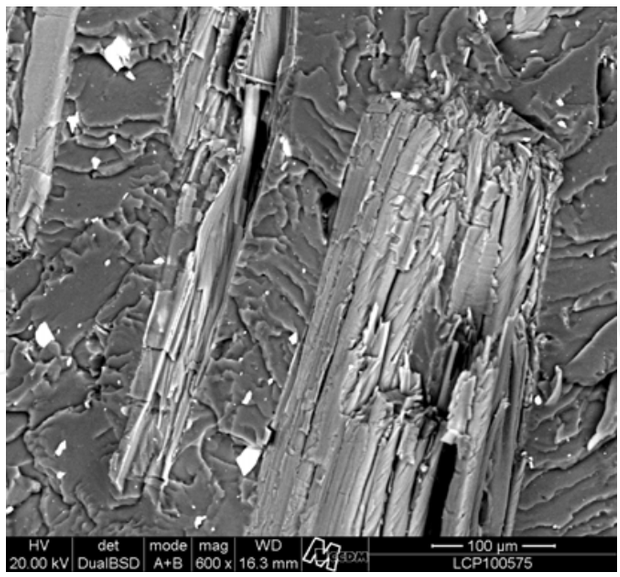


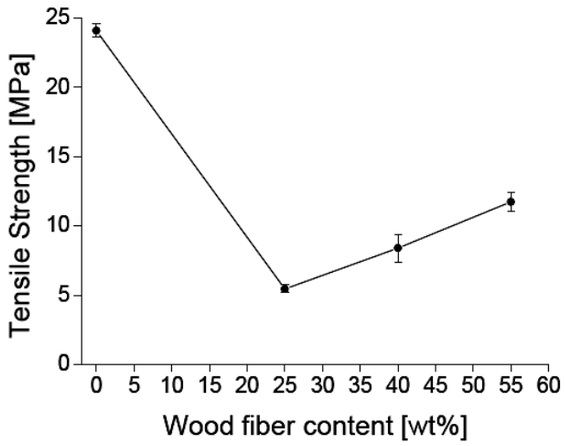
Fig. 16. Cryogenically fractured surface micrographs of the PVB/wood flour (60/40) composite

Figure 17 shows the results of the tensile tests for PVB/wood flour composites. The graph in Figure 17a shows that the tensile strength values of the composites are significantly lower than those found for neat PVB. However, the tensile strength tends to increase when the amount of wood flour in the composites increases from 25% to 55%. The mechanical properties of fiber-reinforced composites depend not only on the characteristics of the polymer matrix and the reinforcement fibers but also on the interface between the matrix and the fiber, which is important for the transfer of interfacial stress (George, 2001; Madera-Santana et al., 2002; Saheb & Jog, 1999). A reduction in tensile strength is a general phenomenon that occurs in the case of thermoplastics filled with natural fillers as the filler loading increases, thereby increasing the interfacial area. The increase in tensile strength with increasing wood flour content observed in the PVB/wood flour composite without the addition of coupling agents or compatibilizers is unusual for thermoplastic composites with natural fibers. This behavior may be related to the good interfacial adhesion observed by the continuity of the wood flour with the PVB matrix shown in Figure 16.

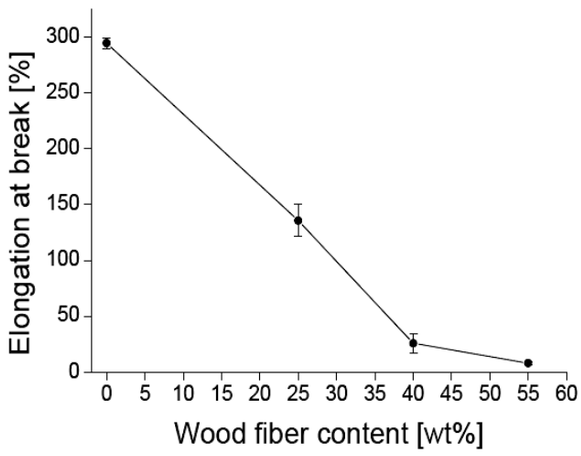
Figure 17b shows the decrease in elongation at break with increasing wood flour content as usually expected in composites where the disperse phase is stiffer than the continuous phase. This behavior is also observed in Figure 17c which presents the tensile modulus as a function of wood fiber content. The increase in Young's modulus show that wood fiber increased composite stiffness.

The low standard deviation and the increase in mechanical properties with leather fiber and wood flour content show that the mixing process in a single screw extruder with Maillefer type screw and pineapple mixing element in the metering zone can produce homogeneous composites.

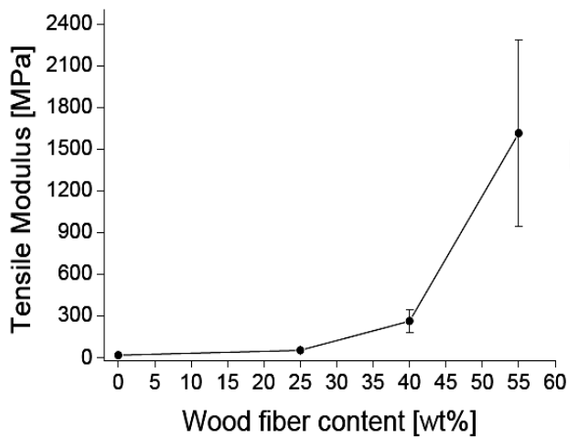
The PVB/wood flour composites were obtained from fully recycled raw materials, resulting in innovative flexible composite sheets, as seen in figure 18, which can be used as coatings on curved surfaces, allowing freedom of creation and design. The composites showed a good interface between fiber and polymer, observed by Electron Microscopy (SEM).



(a)



(b)



(c)

Fig. 17. Tensile tests results of PVB/wood composites fiber. a) Tensile strength, b) Elongation at break and c) Tensile modulus



Fig. 18. Flexible sheet of PVB/wood flour (60/40) composite

2.6 PVB/Leather composites

Due to the presence of chromium in leather fibers, relatively low processing temperatures must be used, which makes PVB a very interesting polymer matrix for the development of PVB/leather composites.

Figure 19 presents SEM micrographs of cryogenically fractured surfaces of the PVB/leather composite samples, where leather fiber agglomerates appear distributed in the PVB matrix. These composites were obtained by the same process used to prepare PVB/wood flour composites. Although there are some voids in the matrix, possibly due to defibrillation and pull-out of the leather fiber agglomerates, good interfacial adhesion can be observed by the continuity of the leather fiber agglomerates with the PVB matrix (Ambrósio et al., 2011). As mentioned before, good adhesion is usually difficult to achieve between nonpolar thermoplastic matrix and natural fibers due to the polarity differences between the hydrophilic fibers (composed of collagen macromolecules) and the hydrophobic thermoplastic matrices. PVB copolymer contains vinyl alcohol hydroxyl groups which may interact with the OH and -C(=O)-OH groups of the collagen macromolecules in the leather fibers . The interaction of these groups may have created an interface which managed to transfer stress from the PVB matrix to the leather fibers upon cryogenic fracture (Ambrósio et al., 2011).

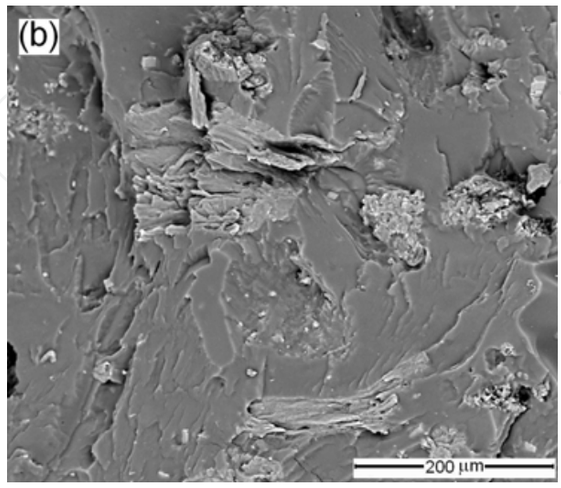


Fig. 19. Cryogenically fractured surface micrographs of the PVB/leather fiber (70/30) composites (Ambrósio et al., 2011)

Figure 20 shows the results of the tensile tests for PVB composites with recycled leather fibers. The elastic modulus increases considerably with increasing amounts of leather fiber in the composite. The elastic modulus increased from 4 MPa for neat PVB to 270 MPa for the composite with 70 wt% leather fiber. Such a rapid and nonlinear increase of the elastic modulus with fiber content is not commonly observed in thermoplastic composites reinforced with natural fibers. As previously discussed, the PVB contains plasticizer, which imparts improved flexibility to the composites (Ambrósio et al., 2011).

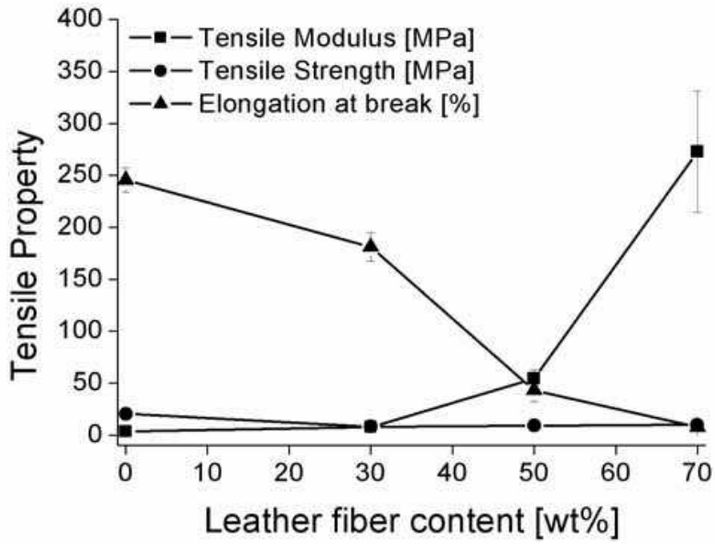


Fig. 20. Variation in tensile mechanical properties of the composites as a function of leather fiber content

There are some studies in the literature on thermoplastic composites containing leather fibers. Matrices include highly plasticized PVC (pPVC) (Madera-Santana et al., 1998), acrylonitrile-butadiene-styrene terpolymer (ABS) (Ramaraj, 2006) and poly(methyl methacrylate) (PMMA) (Madera-Santana & Moreno, 1999), where the elastic modulus has shown different behavior. While ABS/leather and PMMA/leather composites exhibit decrease in elastic modulus or a slight increase as leather fiber content increases, p-PVC/leather composites show similar behavior to PVB/leather composites, i.e., the elastic modulus is strongly influenced by fiber content, especially at compositions exceeding 30 wt% leather fibers. It should be pointed out that both polymer/leather composites where the elastic modulus increases considerably with increasing amounts of leather fiber, had plasticized thermoplastic matrices, i.e., p-PVC and PVB used in this study. The reduction in tensile strain of the PVB/leather composites may be attributed to a reduction in the deformation ability of the matrix due to the concentration of leather fiber agglomerates (Ambrósio et al., 2011).

It should be mentioned that, despite the fact that the mechanical properties of this composite did not show great improvement, this material has presented all the adequate standard properties for its use in shoe soles and insoles: abrasion resistance, hardness, tear resistance, adhesion to commercial glues, flexibility or folding ability, appearance etc. and is currently being tested in shoe factories with success. By means of a simple and cheap processing technique, i.e., a harvest feed extrusion equipped with a Maillefer single screw, composites

were manufactured with suitable properties for use as shoe soles, to be returned as a raw material to the shoe industry chain, facilitating approval of the recycled products by the regulatory agencies.

3. A few words on life cycle assessment (LCA) of natural fiber polymer composites

There are many methodologies in use to determine the LCA of a final product. Whatever the methodology adopted, the following characteristics may impact positively on the environmental impact of natural fiber polymer based products, if adequately handled: resources, production, distribution, use, disposal and re-use.

With regard to the recyclability of these composites, Tajvidi & Takemura (2009) have shown that these composites can be reprocessed many times, even with gain in stiffness, water absorption, and no effect on thermal stability, as a consequence of an observed improvement of the interface quality of the composites.

According to Murphy (2004) as cited in Patel *et al.* (2003) at least in the context of bio-based polymers, reductions of the order of 20% in most environmental impacts (and specifically a saving of 20 MJ per kg of polymer and avoidance of 1 kg CO_2 per kg of polymer) would be a useful guide for a good environmental improvement target.

Pervaiz & Sain (2003) have performed a comparative life cycle analysis focused on nonrenewable energy consumption of natural and glass fiber composites. Their analysis shows that a net saving of 50 000 MJ (3 ton CO_2 emissions) per ton of thermoplastic can be achieved by replacing 30% glass fiber reinforcement with 65% hemp fiber. It is further estimated that 3.07 million ton CO_2 emissions (4.3% of total USA industrial emissions) and 1.19 million m^3 crude oil (1.0% of total Canadian oil consumption) can be saved by substituting 50% fiber glass plastics with natural fiber composites in North American auto applications. However, to compete with glass fiber effectively, further research is needed to improve natural fiber processing, interfacial bonding and control moisture sensitivity in longer run.

Joshi et al. (2004) performed extensive studies comparing life cycle environmental performance of natural fiber composites with glass fiber reinforced composites and found that natural fiber composites are environmentally superior in the specific applications studied, that means, each case or application must be studied separately.

When dealing with leather residues, the benefits go beyond: the leather scraps may be used as shoe soles in the shoe making chain itself, avoiding land fill of tons of scraps. This might generate economy in the correct disposal of this residue (which must be very well controlled and then paid for), and in the energy used for its production, also serving as a cheaper raw material for new recycled shoe soles and insoles. From an environmental point of view, tons of chrome may be kept inside the polymer as Cr^{3+}, without the risk of oxidizing to Cr^{6+} under the action of sun light and heat, reducing the risk of water and soil contamination. Regarding balsa wood, wood flour, saw dust, bamboo and coir polymer composites we also have to consider the CO_2 sequestration capacity of these composites during their useful life.

Of course, the use of industrial and postconsumer residues and the recovery of forests must be stimulated by public policies and programs, in addition to education of the people, who will be the major beneficiaries. Research studies may point out solutions and alternatives. However, adequate waste management and recycling programs are required to make these materials become reality.

4. Concluding remarks

From the few case studies presented in this text, we may conclude that high value added natural fiber reinforced polymer composites can be developed from different industrial and postconsumer urban residues or waste, with positive impact on the environment by reducing contamination and sequestering CO_2 from the atmosphere, contributing to the recovery of not only the Amazon Rainforest, but also any forest worldwide.

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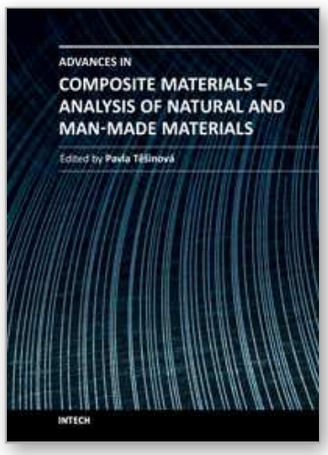
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Composites are made up of constituent materials with high engineering potential. This potential is wide as wide is the variation of materials and structure constructions when new updates are invented every day. Technological advances in composite field are included in the equipment surrounding us daily; our lives are becoming safer, hand in hand with economical and ecological advantages. This book collects original studies concerning composite materials, their properties and testing from various points of view. Chapters are divided into groups according to their main aim. Material properties are described in innovative way either for standard components as glass, epoxy, carbon, etc. or biomaterials and natural sources materials as ramie, bone, wood, etc. Manufacturing processes are represented by moulding methods; lamination process includes monitoring during process. Innovative testing procedures are described in electrochemistry, pulse velocity, fracture toughness in macro-micro mechanical behaviour and more.

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