APPLICATIONS OF NATURAL FIBERS IN AUTOMOTIVE INDUSTRY IN BRAZIL - THERMOFORMING PROCESS

Alcides L. Leao[1]; Roger Rowell[2]; and Nilton Tavares[3];

[1]Professor - UNESP, FCA, Campus Botucatu, SP, Brazil - 18.603-970
[2]Project Leader - USDA Forest Products Laboratory, Madison, WI, USA
[3]R&D Manager - Toro Ind. e Com. Ltd., Diadema, SP, Brazil

INTRODUCTION

Environmental and economical concerns are stimulating research in the development of new materials for construction, furniture, packaging and automotive industries. Particularly attractive are the new materials in which a good part is based on natural renewable resources, preventing further stresses on the environment by depleting dwindling wood resources from forests. Examples of such raw material sources are annual growth native crops/plants/fibers, which are abundantly available in tropical regions. These plants/fibers (like jute and sisal) have been used for hundreds of years for many applications such as ropes, beds, bags, etc. If new uses of fast growing, native plants can be developed for high value, non-timber based materials, there is a tremendous potential of creating jobs in the rural sector. These renewable, non-timber based materials could reduce the use of traditional materials such as wood, minerals and plastics for some applications. There is a tremendous interest by the pharmaceutical industry in exploring the rain forest for new drugs, but so far there has been little interest in exploring the rain forest for fast growing native plants as a fiber source. In applications such as ropes, new materials, such as nylon, have replaced locally grown fibers like sisal and jute. Therefore, increasing interest in saving the forest *and* at the same time creating rural employment means that new materials have to be developed to use locally available non-wood renewable resources. The advantages of these plants are that they are fast-growing and renewable, and sometimes are also a source of food supply for animals and even humans.

The materials made through the combination of native fibers with different plastics, such as Polyvinylcloride (PVC), Polypropylene (PP), and Polyethylene (PE) are composites that have wide application possibilities, having a potential of developing new industries in the near future using local crops, wastes and labor, and helping to reduce the demand for tropical hardwoods (housing, furniture, pellets, etc.), and plastics.

The technology developed in this project could easily be applied in other tropical countries, with similar ecosystems.

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LITERATURE REVIEW

Lignocellulosic fibers are a kind of biopolymer composite, with the following components, in different proportions, depending on the species considered: cellulose, hemicellulose, lignin, and other components in small proportions. These polymers are the basic constituents of the cell wall and are responsible for most of the physical and chemical properties, such as dimensional instability to moisture, biodegradability, flammability, thermoplasticity, and degradability by ultraviolet light, acids, and bases. All of these characteristics will result in specific end-use of lignocellulosics in composite formulation. Lignocellulosics, can also be called *phytomass*, biobased fibers, or biofibers, including wood, agricultural residues, water plants, grass, agricultural fibers, and any other plant substance (ROWELL, et al. 1993; SCHNEIDER et al, 1994).

Lignocellulosics can be put together with one or more substances, using some kind of matrix, that would hold the components together, resulting in a composite material. Several options are available, with different processes, compositions and bonding agents. Thermoplastics can be used to make composites for nonstructural parts, and thermosetting resins can be used for structural composites. These composites can be designed based on end-use requirements, under the most important aspects nowadays; cost, availability, renewability, recyclability, energy use, and environmental safety. Fiber technology, high performance adhesives, and chemical modification for the fibers can result in a composite with uniform properties, durable in adverse environments, and mainly with high performance, suitable to compete with other man-made materials. This new technology can be applied to either virgin or recycled fibers from any plant. Recycled lignocellulosic fibers can be used as a reinforcement filler in composites with polyolefins. Recycled plastics can be used as binders producing a high performance - low cost composite material. (ROWELL, et. al. 1993).

Lignocellulosics can be used as reinforcing fillers in thermoplastics composites, which would lead to increasing strength, stiffness, and heat distortion temperature, although a decrease in impact resistance is expected. Fibers with high aspect ratio, such as cellulose, are more convenient than flours. Other properties can be enhanced using small amounts of additives or coupling agents, to aid dispersion of the polar fillers in the non-polar matrix polymer, therefore increasing the bondability between the filler and the polymer (SCHNEIDER, et al. 1994).

Thermoformable polypropylene and lignocellulosic fibers can be extruded in sheet form for use in automotive interior components, such as door panels, trunk liners, sealbacks, package and speaker trays, reducing the consumption of petro-dependent materials in the thermo-moldable plastics by less than 50% (BALATINECZ & WOODHAMS, 1993). Several alternatives have already been reported with very promising results: bagasse, cereal straw, kenaf, rice husks, rice straw, sunflower hull and stalks, jute, sisal, banana, etc. The USDA Forest Service, Forest Products Laboratory surveyed worldwide for agricultural fibers and residues utilization and selected a total of 1,039 citations from a vast number available, showing a strong interest all around the world for these kind of materials, depending only on its local availability (YOUNGQUIST et al. 1993, 1994). . Most of the research has been conducted using polypropylene, with good results (CLEMONS & MYERS, 1993).

There are, however, problems regarding the compatibility of surface energies between hydrophilic (polar) cellulose and hydrophobic (non-polar) polyolefins. Consequently this characteristic, poor match of surface polarities results in poor bonding between polyolefins and cellulose. Therefore, cellulose must be treated on its surface, or totally, by various methods to increase its bondability (determined by e.g. contact angle -wettability determination). Differences in adhesion between modified cellulose and polypropylene can be measured by internal bond and peel tests. The use of sizing agents reduces the acid/base (hydrogen bonding) character of the cellulose surface. There is a corresponding relation between hydrogen bonding

and adhesion between cellulose and polypropylene. It is recommended to modify the hydrophobic polymer in a way to introduce chemical modification in its surface, that could result in strong acid/base (hydrogen bonding) interactions (QUILLIN, et al. 1992). The evaluation of the quality of the fiber-matrix bond is done by a macroscopic pull-out technique that determines the interphase properties in lignocellulosics/low-molecular-weight-thermoplastic systems. It is also useful to evaluate the effect of water exposure over the dimensional stability of lignocellulosics fiber based composites (SANADI, ROWELL & YOUNG, 1993).

From the over 1,500 citations found, related to agricultural fibers, 101 are for coir, 14 for areca, 12 for banana, 5 for cassava, 23 for jute, and 15 for palm, showing the potential for fibers that are commonly found in the rain forest region. Any of these fibers, alone or in combination, blended with man-made fibers, could replace wood as a major component in fiberboards, and even replace plastics in several industrial and commercial applications (BENATTI & LEÃO, 1994; SCHNEIDER 1994). Many plants with fibers yet unknown scientifically can be found in the tropical regions that would replace wood and man-made fibers in several different applications (YOUNGQUIST et al. 1994). Fibers from the Amazon and other tropical rain-forests show high potential due to their mechanical and physical characteristics such as: curaua, malva, pupunha, jute, banana, etc. (LEÃO & TAVARES, 1995). Among these tropical fibers, one is of particular interest and will be studied in this project: the curaua, whose properties are described below.

-CURAUA - *Ananas erectifolius* - This is a hydrophilous species from the Amazon region. Its leaves are hard, erect and have flat surfaces. The leaves are about one meter long, or more, and 4 cm wide. The plant requires 2,000 mm or more of annual precipitation, preferring silil-humus soils, but also grows in clay-sillic soils. It is commonly used by the Indians as a favorite plant for fibers utilization for hammocks and fishing lines. Eight months old leaves can reach up 1.5m in length, and 50-60 leaves per year. The dry fiber content in leaves is about 5-8%. The fiber is commonly extracted by a primitive process called “forca” (hanger), washed and beaten with a circular rod and left in water in order to mercerize for 36 hours. They are again washed and allowed to dry. Natural Curaua fiber (wet) shows an average elongation of 4.5%, MOE of 10.5 GPa, and MOR 439 MPa. Dry fiber (OD) values are respectively 3.2%, 27.1 GPa, and 117 MPa. Cutted and defibred the values are respectively 3.7%, 9.7 GPa, and 428 MPa. Cutted, defibred and dried (OD), the values are respectively 4.3%, 11.8 GPa, and 502 MPa. Curaua is very competitive among the traditional fibers, always ranking in the top three for economical analysis and top four for stiffness (TORO, 1994). It has been reported in a study using several agricultural fibers, such as: jute, kenaf, abaca, sisal and henequem, that bast fibers composites have higher properties than leaf fiber composites. On the other hand, impact toughness values are higher for leaf fibers compared to bast fiber composites (JACOBSON, ROWELL & CAULFIELD, 1995).

METHODOLOGY

Several natural fibers, such as curaua, coir, jute, ramie and sisal were studied in this project. Several combinations of plastics and fibers were studied with and without coupling agent (Maleated Polypropylene - MAPP - G3002, from Eastman Kodak). Combinations of three components were also studied, such as viscose-jute-polypropylene, in different ratios. The density varies from 0.59 to 0.95. The thickness was between 1.75 up to 2.95 mm. The statistical design used was an incomplete factorial. Three samples of each treatment were tested and analyzed through the statistical package SAS. The forming process was the same for all treatments, using a commercial industrial size machine located at the Toro Ind. & Com. Ltd. plant, located in Diadema, SP, Brazil.

The fiber components were mixed on a dry weight basis and mixed by hand prior to the picker, which cuts the fibers to 5 cm in length, also giving more mixing. After the picker, the

mixture goes to the cards, where more mixing can be obtained, and finally it goes to the needle machine. The mats were cut and pressed at different temperatures, densities, pressure and time of heating and cooling. The heating time was 30 to 180 seconds. The pressure varies from 3.25 MPa up to 9.8 MPa. The pressure time in the cold press was 20-30 seconds. The samples were taken from the boards using a pressing knife. The parameters evaluated were EMC (Equilibrium Moisture Content), bending (ASTM D790M-93) tension (ASTM D638M-89) and dimensional stability.

RESULTS AND DISCUSSION

The moisture content (MC) in the air at uncontrolled conditions, after 10 days of pressing was very low for all the treatments, and therefore it will not be considered as a variable. The results obtained varied from 1.6 to 2.8%, showing that there was no significant variation among the treatments, and according to the objectives of obtaining a stable composite.

It was used in some trials recycled materials that are resultant from parts with non conformity, that are scrapped and returned for the line. The use of recycled materials in the tecnomix mixture was very deleterious for all properties tested, the maximum of recycled material usable in the tecnomix mixture being 10%. The mixture of viscose, jute and polypropylene (25/30/45) gave poor results for tensile strength (20.26 MPa), but average results for flexural strength (23.09 MPa), and flexural (2.07 GPa) and tensile modulus (1.83 GPa), showing that it can be used in some applications, where tension strength in not so important.

Short fibers, as coir and sugar cane bagasse, are limited in their strength properties due to the non-woven technologies, but can compete in prices with fibers such as jute, ramie and sisal, Curaua has no competition in performance, since the prices of all natural fibers are about the same in Brazil (US$0.60/kg FOB).

The limit for natural fibers content, at the processing conditions, were 60% for all the treatments, including curaua. For curaua, at 20% polypropylene, a strong reduction in tensile and flexural strength was found, much more than for tensile and flexural modules (Table 1).

For jute, the differences among the treatments were not so high as for curaua (Table 2). Ratios of 30/70 and 40/60 for jute/polypropylene showed very little differences, with low values, compared to standard ratio considered of 50/50, mainly for flexural and tension strength.

For sisal, high natural fiber content resulted in lower values compared to the standard 50/50 natural fibers/polypropylene. At 70/30, the values were similar to curaua (Table 3). The modules showed very little differences.

At standard ratio, 50/50, curaua was much higher than ramie and jute, in this sequence (Table 4). Again, values of modulus showed no significant differences.

Table 1. Effect of Natural Fibers Content on Curaua/Polypropylene Composites.

RATIO CURAUA/PP (%)	TENSION STRENGTH MPa	TENSION MODULUS GPa	FLEXURAL STRENGTH MPa	FLEXURAL MODULUS GPa
80/20	19.98	1.56	14.67	1.44
70/30	21.83	1.63	24.13	3.03
60/40	50.75	3.34	24.97	2.53
50/50	46.38	3.78	33.10	2.51

Table 2. Effect of Natural Fibers Content on Jute/Polypropylene Composites.

RATIO JUTE/PP (%)	TENSION STRENGTH MPa	TENSION MODULUS GPa	FLEXURAL STRENGTH MPa	FLEXURAL MODULUS GPa
30/70	23.21	2.17	24.47	2.24
40/60	24	2.72	26.74	2.38
50/50	16.52	2.63	27.33	2.9

Table 3. Effect of Natural Fibers Content on Sisal/Polypropylene Composites.

RATIO SISAL/PP (%)	TENSION STRENGTH MPa	TENSION MODULUS GPa	FLEXURAL STRENGTH MPa	FLEXURAL MODULUS GPa
80/20	11.12	1.25	16.36	1.36
70/30	25.41	2.35	23.76	1.83
60/40	23.25	1.97	32.3	2.78

Table 4. Effect of Species on Standard Ratio 50/50 Natural Fibers/Polypropylene Composites.

RATIO NAT.FIB./PP 50/50%	TENSION STRENGTH MPa	TENSION MODULUS GPa	FLEXURAL STRENGTH MPa	FLEXURAL MODULUS GPa
RAMIE	34.67	3.43	29.33	3.02
JUTE	15.66	2.4	23.97	2.5
CURAUA	46.58	3.78	33.1	2.51

Heating time was a significant variable. It must be enough to heat and melt the polypropylene inside the mats. The minimum observed was 60 seconds. The temperature cannot go above 220°C, to prevent burning of the natural fibers. The low results for jute 50/50, compared to the other fibers is explained by the short time of heating in this treatment.

The basic weight was a significant variable on the strength properties, although, at differences of 200 g/m^2, the results showed no differences. Therefore, if high values are needed in modules, the basic weight must go above 2000 g/m^2. In this case the basic weight will make a difference, more than the species of the fibers.

The thickness was not important for the levels studied (1.8-3.0 mm), showing, that it is closely related to basic weight, but it is not so sensible to small variations. Above 3.0 and below 1.5 mm, the properties have a significant reduction.

The pressure at the values tested, varying from 3.25 MPa up to 9.8 MPa, at the forming Press (cool) were not significantly different. The heating pressure was used only to compress the mats and carry the heat throughout, with values varying from 0.82 to 3.67 Kg/cm^2. The

Table 5. Effect of Species on Standard Ratio 50/50 Natural Fibers/Polypropylene Composites.

RATIO NAT. FIB./PP 80/20%	TENSION STRENGTH MPa	TENSION MODULUS GPa	FLEXURAL STRENGTH MPa	FLEXURAL MODULUS GPa
CURAUA	19.98	1.56	14.67	1.44
SISAL	11.12	1.25	16.36	1.36
COIR	9.25	0.45	9.23	0.43

cooling time at the forming press also was not significant at the values tested, showing that it was above the minimum limit.

Values of 80% of natural fibers were too high to get a good mat, resulting in composites with poor properties for all the fibers tested, more for coir, due to be the shortest fiber tested, as observed in Table 5. Even in this case, curaua showed much better values than coir. Sisal showed values at middle range.

Further studies must be developed in accelerated aging, and IZOD impact resistance for these composites. By the preliminary results observed with curaua, this will be the only fiber that could compete with glass fibers.

CONCLUSIONS

Curaua is by far the best natural fiber for composites applications at the conditions tested. The others, showed a similar behavior, indicating that price and availability will be the main aspect. The natural fibers tested showed a very high competitiveness with traditional materials, mainly wood and fiber-glass based composites. The applications of the composites tested are ideal for sound insulation and structural parts for interiors in the automotive industry.

The temperature of processing, must be high enough to melt the polypropylene, but below the technical limit of 220°C. The basic weight was important, showing better properties for denser materials. However, since it represents a technical and economical trade-off, values of 1550-1800 g/cm^2 showed very similar behavior. Other variables such as pressing, and cooling time were not significant.

The use of recycled material in the mixture must be limited to 10%, because the fibers are shortened and damaged in each processing pass.

Further analysis has been developed with several natural fibers using injection molding technique, which will be described in another paper.

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Edited by

Paras N. Prasad
State University of New York at Buffalo
Buffalo, New York

James E. Mark
University of Cincinnati
Cincinnati, Ohio

Sherif H. Kandil
Alexandria University
Alexandria, Egypt

and

Zakya H. Kafafi
Naval Research Laboratory
Washington, D. C.

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