ALTERNATIVE FIBER TO REINFORCED PLASTICS

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Abstract*. This research is based, in principle, in search of reinforcement natural alternative to polymeric composites, also known as reinforced plastics. This sense, the research present work begins with a complete characterization study of the* ***licuri fibers****, such as alternative proposal for reinforcement the polymeric composites. Then, a study was conducted to develop a composite laminate only the basis of licuri fiber, in order to know the behavior when the fiber impregnated with thermosetting resins (orthophthalic unsaturated polyester). The composite was developed as a laminar structure (plate with a single layer of reinforcement) and industrially manufactured. The study of characterization of the fiber was based on the determination of physical, chemical and mechanical properties. The mechanical properties of the sheets were determined through longitudinal and transversal tensile tests. A study with objective to understand the influence of interface adherence of fiber/matrix, in the final behavior of the composite, was necessary.*

Keywords: Polymer composite laminates, licuri fibers, mechanical properties.

1. INTRODUCTION

Composites resulting from the combination of different materials, which are better or more adequate together than their original structures, are increasingly relevant. They often have properties that traditional materials do not.

The structural application of composite material has increased in recent decades due to the perfecting of manufacturing processes, new reinforcement "fabrics" and laminate structures (Mander, 1981; Fernando et al., 1998; Maron et al., 1989). The increase in the use of "natural" fiber-based composites should also be pointed out, especially in structures subjected to minor stress. This is because natural fibers usually have lower mechanical property values than those of synthetics, preventing their use in medium or high performance structures.

The use of naturally reinforced polymer resin composites is still a challenge for researchers of these materials. The search for better physical, mechanical and chemical properties in natural fibers has led researchers to study new species that may meet the increasingly high demand (Moe and Liao, 2002; Aquino et al., 2007).

The pursuit of new composites, using natural fibers or a mix of these with synthetic fibers as reinforcement, aims at reducing environmental impact. Natural fibers also have other advantages over synthetics fibers, such as cost, lightness and strength. However, some of these fibers have deficiencies in mechanical performance and moisture absorption, among others. This demands a search for new materials that correspond to desired structural expectations.

The present study aims at investigating new natural reinforcements sufficiently compatible with polymer resins to form new composite materials. The fiber of the Licuri Palm *Syagrus coronata* (Martius) Becari, native to Brazil, was investigated for this purpose. The **licurí fiber** is belongs to the subfamily Arecoideae, tribe Cocoeae, subtribe Butineae (Noblick, 1991). This subfamily is the largest of the Arecaceae, currently comprising 115 genera and 1500 species (Uhl, 1995). Licuri fiber is obtained from the palm leaf and is flexible enough to be woven. It is therefore presented in this study as a unidirectional fabric obtained on a manual loom.

Of the many names by which the species is known, **licuri** is the name most used in Brazil (Bahia). The palm tree varies in height from 6 – 10m. Although it flowers and bears fruit all year with few variations, the harvest period in March, June and July has the highest fruit production (Noblick, 1991).

Many studies have been carried out in the food sector on its high-calorie fruit as a means of fighting malnutrition in the area or for use as alternative energy source and in the production of biodiesel. The Centro Federal de Educação Tecnológica da Bahia CEFET-BA has been conducting a study of this kind since 1993, as well as developing and improving machines and equipment for the breakdown and improvement of licuri. However, little is known about the properties of its fibers, especially in relation to its characteristics at reinforcement.

The plant is resistant to parasites and disease and is estimated to produce for 100 years or more. It does not choose where to germinate in this dry terrain – wherever a seed falls another palm tree will sprout (Cadernos Temático, 2004).

Special attention will be given in this study to microstructural characterization, strength, rigidity and the physical and chemical properties of the fiber. A polymer resin composite (orthophthalic polyester) will then be developed and reinforced solely with licuri fibers.

The composite is made in the form of a sheet. The micrographic study of the sheet focuses on the interfacial adhesion between the fibers and the matrix. This is fundamentally important in the mechanical response of the final product. The mechanical performance of the composite (strength, elastic modulus and fracture) is determined by uniaxial tensile and three-point bending tests. The composite was made in the form of a sheet at Tecniplast C&A Ltda by hand lamination (hand lay-up). Figure 1 shows the licuri palm (*syagrus coronata*).

Figure 1. Licuri Palm Tree (*Syagrus coronata*) (DESER, 2008).

2. MATERIALS AND METHODS

The composite material used in this study was in the form of a lamina, in other words only one layer of reinforced fabric was impregnated. Since natural licuri fiber (***Syagrus coronata***) was chosen as reinforcement, the fabric was made using this fiber. The fibers used came from the city of **Caldeirão Grande** in the semi-arid region of Bahia and were taken from licuri palms in the same soil. The unidirectional fabric was manufactured on a manual loom (418.20 g/m^2). Figure 2 (a) and (b) show the fabric made with licuri fibers.

(a)

(b)

Figure 2. (a) and (b) – Licuri fiber fabric.

The transverse fibers seen in the fabric keep the fabric strands parallel. The resin used to produce the composite was unsaturated orthophthalic polyester with a mean tensile strength and longitudinal modulus of 30 MPa and 2.3 GPa, respectively.

The composites were obtained industrially (Tecniplast ind. Ltda) in the form of lamina, using manual lamination (hand lay-up). The licuri fiber fabric was impregnated in orthophthalic polyester resin, common in the reinforced plastic industry. The lamina produced was 45cm wide and 65cm long. The fibers were used “in natura” without previous treatment, so as not to increase production costs.

The microstructure of the licuri fibers was analyzed by scanning electron microscopy (SEM) and the characteristics of the longitudinal and transverse sections were determined. The fiber surface was previously coated with gold and the equipment used was a scanning electron microscope (Shimadzu SS-550). Were used fibers “in natura”, fibers washed in running water and others treated with ethanol/toluene to remove the wax.

X-ray diffraction (XRD) was used to analyze crystallographic structure. The powder method was used with grains below 50 μ and scanning was from 0^o to 80^o with copper tubing and a voltage and current of 40.0 kV and 30.0 mA, respectively. A Shimadzu XRD- 6000 was used for the analyses.

In thermogravimetric (TGA) and differential thermal (DTA) analyses, specimens of dry and green licuri fibers were used to determine if there are behavioral differences these specimens.

Analyses were done to determine the contents of the main components of the licuri fiber studied; that is, extractives, lignin, cellulose, hemicellulose and ashes. Complementary analyses such as cold-water solubility and wax content were performed to determine the interference of soluble on sorption data (agitated and static system) and explain the water impermeability of the fibers. The study followed standers established by the Associação Brasileira Técnica de Celulose e Papel – ABTCP and the Technical Association of the Pulp and Paper Industry – TAPPI, as well as the method described by BLEDZKI and GASSAN (Bledzki and Gassan, 1999).

Calculations were made at 2h and 24h to determine specific mass (volumetric density) and weight was measured at immersion times of 5.0 minutes, 30 minutes, 1 hour and 2 hours to establish the water absorption percentage.

The tensile strength and elastic modulus of the fibers was determined according to **ASTM D3822-96**. Fifteen specimen's fibers of 200 mm long were tested at an assay speed of 10 mm/min with an Emic DL 2000 universal testing machine.

The mechanical behavior of the composite lamina was determined by uniaxial tensile tests. These were performed at ambient temperature (25°C) with a Shimadzu AG-I. The test specimens were cut with a diamond wire and their sides smoothed and polished according to metallography techniques.

Test specimen dimensions and test specifications for the uniaxial tensile tests were in accordance with ASTM D3039-00. The test specimens were cut from the lamina and tabs were used in the uniaxial tensile tests to better accommodate them to the clamps on the testing machine.

Mechanical properties of strength, stiffness (elastic modulus) and fracture strain were determined for the testing. In the uniaxial tensile test, maximum stress (strength) and elastic modulus were determined both longitudinally (where the direction of the fiber corresponds to load application) and transversally. In other words, the direction of load application is perpendicular to the direction of the fiber. Elastic modulus was measured before initial damage.

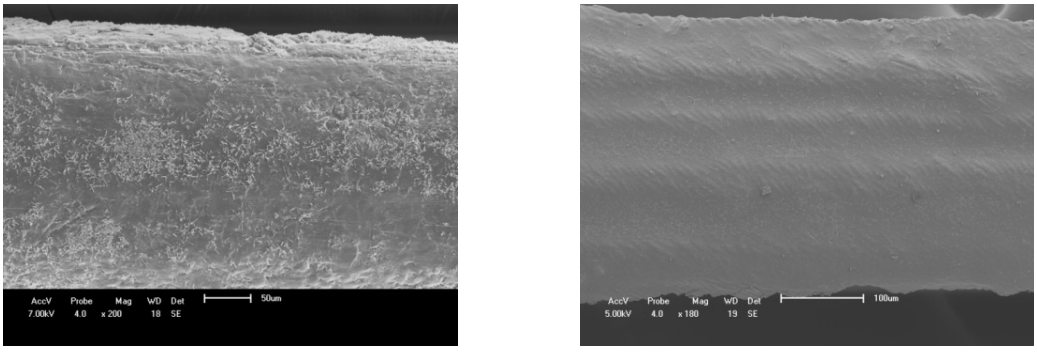
Special care was taken during the tests to adjust the test specimens in the machine clamps to minimize "dragging" and ensure exact dimensions of length effective of the specimens. Eight specimens were tested under the same conditions. The test specimens that fractured within their effective length, in other words behaving according to ASTM D3039/D-00, were selected as valid testing.

All tests were conducted at a temperature of 25 ºC ±2. Following mechanical testing a macroscopic analysis of the fracture characteristics was done on all the specimens.

3. RESULTS AND DISCUSSION

3.1. Morphology

Figures 3 (a) and (b) show the morphology of the licuri fiber in its natural condition tested longitudinally using an electron microscope. The fiber in figure 3 (a) was not cleaned and leaf residue can be seen adhered to the surface. The fiber in figure 3 (b) was washed in running water and dried in an oven at 60 ^{o}C.



(a) (b)

Figure 3. Morphology of licuri fiber: (a) fiber with leaf residue adhered to the surface; (b) fiber washed in running water and oven-dried at 60 ^{o}C.

The fiber is covered by a thick wax cuticle. This explains the high percentage of wax found in the chemical analysis shown in table 5, obstructing the view of its internal structure.

The presence of wax on some plant leaves, as occurs with licuri, may be due to its adaptation to dry areas, since this waxy layer hinders water loss through transpiration and protects the plant from fungi.

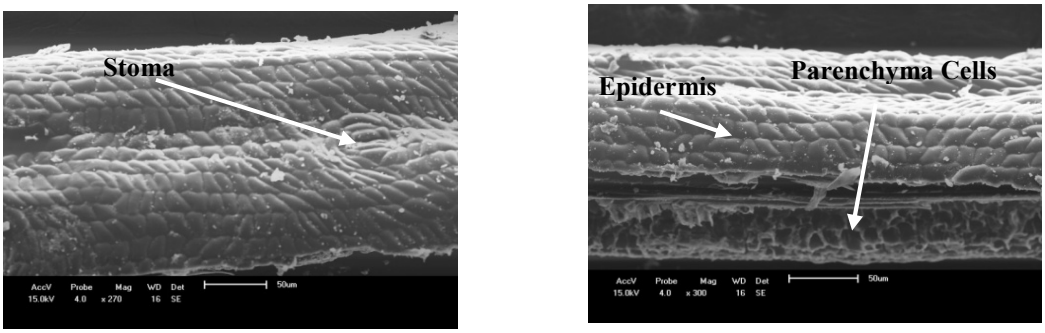
The wax was removed according to method (Bledzki and Gassan, 1999) and another scanning electron microscopy (SEM) was performed on the wax-free fiber for a better examination of its internal structure.

Figures 4 (a) and (b) show the epidermis of the fiber after wax removal (longitudinal view). In figure 4 (a), the stoma or stomata complex can be seen. This is an opening in the epidermis used for gas exchange between the plant and the environment.

Figure 4 (b) shows the fiber with part of the epidermis removed, with a view of the lower parenchyma cells.

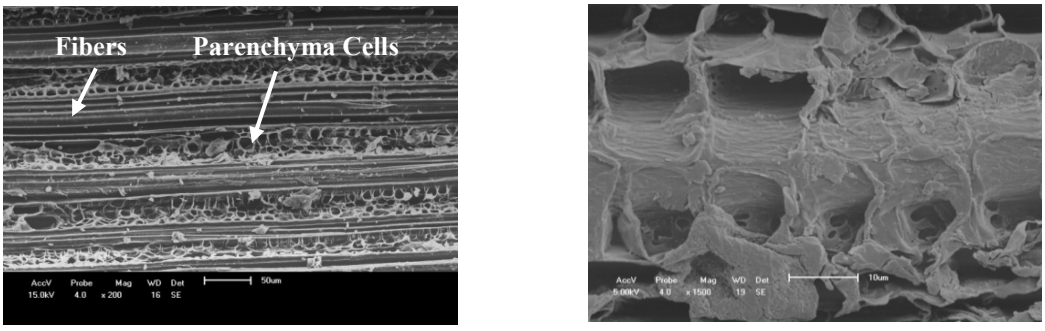
The epidermis is the main outer covering, usually a single layer with no intercellular spaces. In plant parts above ground, it is covered by a wax cuticle that reduces drying.

Figures 5 (a) and (b) show the fiber with the wax and epidermis removed, seen longitudinally and transversally, respectively. The connective tissue (fibers) and parenchyma cells are shown in 5a and the parenchyma cells seen transversally in 5b.



(a) (b)

Figure 4. Wax-free fiber seen longitudinally: (a) epidermis with stoma; (b) Fiber with part of the epidermis removed.



(a) (b)

Figure 5. Wax-free fiber seen longitudinally and transversally: (a) connective tissue and parenchyma cells; (b) parenchyma cells seen transversally.

The parenchymas are tissues located between the epidermis and the connective tissue. Their many roles include filling, assimilation, storage and secretion. They are living cells with large vacuoles. Connective tissue is made up of long, tapering fibers and is responsible for carrying nutrients throughout the plant and for plant resistance.

3.2. Physical, chemical and mechanical properties

Fifteen fibers were tested according to **ASTM D3822-96** to determine the strength and stiffness (elastic modulus) of the licuri fiber. Figure 6 shows the results obtained in uniaxial tensile tests of the fifteen fibers.

A significant variation is apparent in the tensile strength and elastic modulus results of a single fiber. This finding is due to the considerable difference between the diameters measured along the length of the plant fiber.

From the results obtained, despite the wide scattering common to all natural fibers, the licuri fiber strength is shown to be within the range of plant fibers with the highest mechanical strength, such as curaua and jute fibers. The same is observed with stiffness.

According to the literature, fiber compositions vary even among plants of the same species and consequently, so do their properties. The wax content of licuri fibers is higher than that of other fibers discussed in the literature. This elevated content may influence fiber/matrix interface adherence when obtaining polymer composites, a situation that can be solved by prior treatment of the fiber surface.

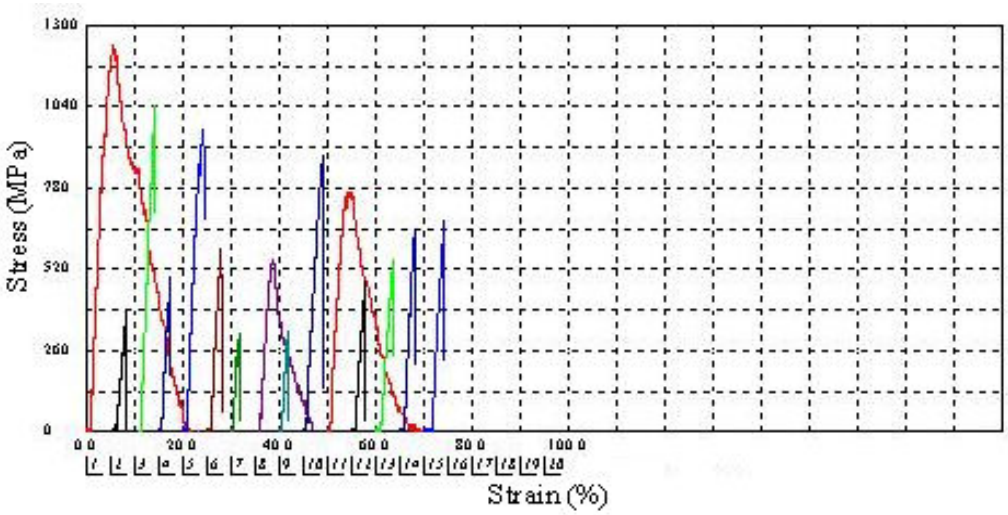


Figure 6. Stress versus strain diagram – Uniaxial tensile strength of the licuri fiber.

Table 1. Physical, chemical and mechanical properties of licuri fiber.

Chemical Components		**Physical and Mechanical Properties**	
Component	***Percentage***	***Properties***	***Values***
Cellulose	53.21± 4.08	Specific mass (g/cm^3)	0.53 – 0.56
Hemicelulose	11.61± 1.07	Maximum absorption (%)	109.98
Lignin	20.69± 0.14	Diameter (μm)	132 – 165
Extractives	20.86± 1.30	Tensile strength (MPa)	369.8– 902.0
Soluble in water	17.60± 0.66	Elastic modulus (GPa)	4.5 – 43.25
Wax	12.72 ± 0.38	-	-
Moisture	8.08 ± 0.00	-	-
Ashes	2.86 ± 0.02	-	-

3.3. Thermal Analyses

As previously mentioned, this test used dry (low drying times) and green licuri fibers to compare the behavior of both specimens. Figures 7 and 8 show the thermogravimetric (TG) curves for the green licuri fiber and the dry fiber, respectively. Table 2 shows a comparative study of the thermogravimetric curves of both specimens.

Table 2. Comparative study of the thermogravimetric curves of green and dry licuri fibers.

Green licuri fiber			***Dry licuri fiber***		
WEIGHT LOSS		TEMPERATURE RANGE (ºC)	WEIGHT LOSS		TEMPERATURE RANGE (ºC)
(mg)	(%)		(mg)	(%)	
0.358	8.041	0 – 38.60	0.481	7.072	0 – 40.83
0.730	16.397	38.60 – 276.86	1.291	18.983	40.83 – 282.07
1.748	39.263	276.86 – 334.63	2.613	38.421	282.07 – 328.18
0.859	19.295	334.63 – 483.14	1.911	28.099	328.18 – 456.36

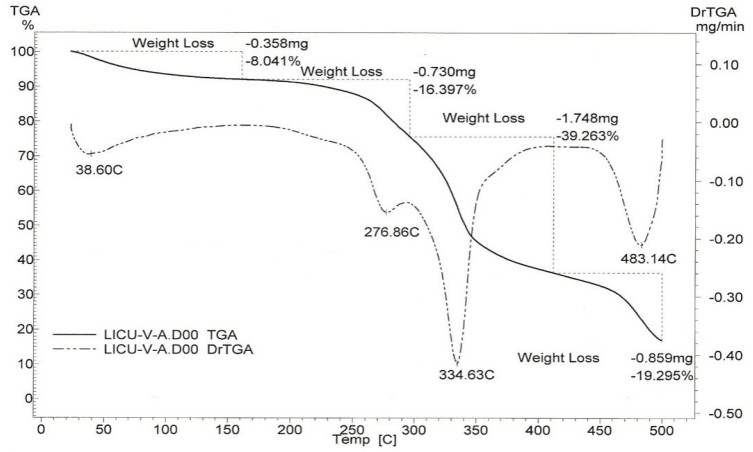


Figure 7. Thermogravimetric curve of green licuri fiber.

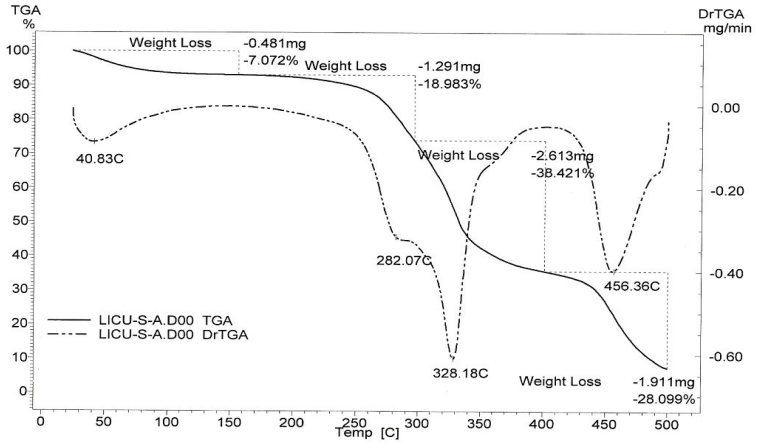


Figure 8. Thermogravimetric curve of dry licuri fiber.

Different thermal analyses of the green and dry licuri fibers were performed; however, the thermal events found were very small and showed calorie points of little significance.

3.4. X-Ray Diffraction (XRD) Test

An evaluation of the crystallographic structure of the material shows two distinct phases: one amorphous, owing to the possible presence of lignin and hemicellulose and one crystalline, due to the presence of cellulose. Figure 9 shows the diffractogram profile of the licuri fiber.

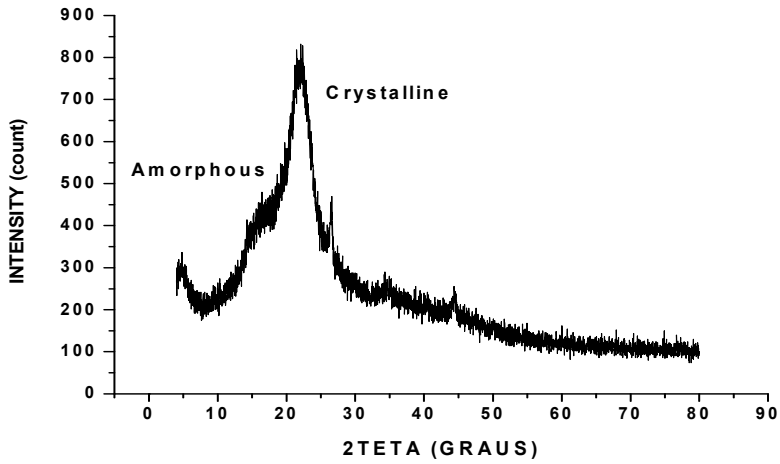


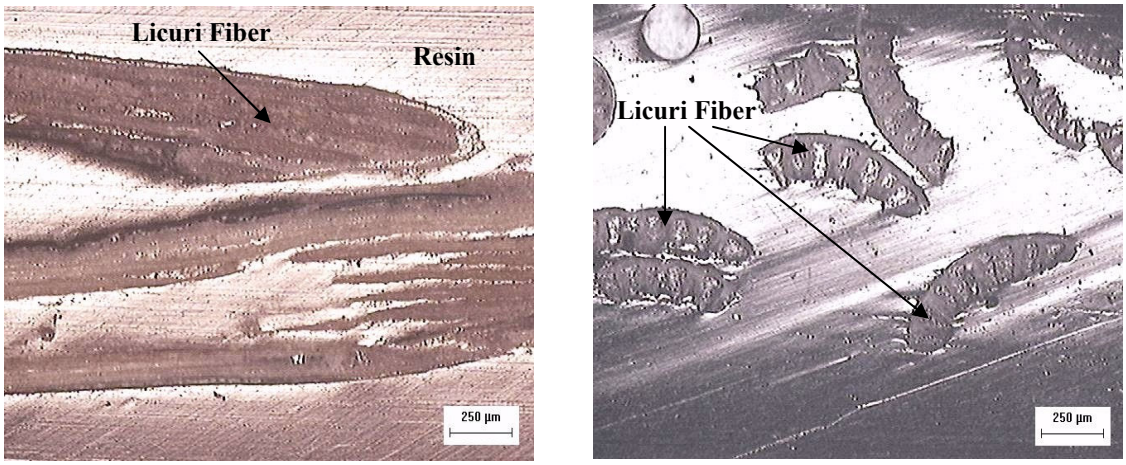
Figure 9. Diffractogram profile of the licuri fiber.

4. LICURI FIBER REINFORCED COMPOSITE

4.1. Microstructure of Licuri Fiber Reinforced Composite .

The microstructural analysis of the composite was performed with an optical microscope to determine the influence of the manufacturing process (***hand lay- up***) on the quality of the fiber/matrix interface and the consequent distribution of fabric strands after impregnating. Figure 10 (a) and (b) show microstructural aspects of the composite, in both longitudinal and transverse directions, where a high percentage of resin compared to the fibers can be observed.

This high resin percentage is due to both the manufacturing process and the type of reinforcement material. The material was obtained in its natural state, with no control over the linear density of the strands in its manufacture. These facts compromise the impregnating process and hinder the elimination of excess resin, especially in the area in contact with the mold. Figure 10 (b) shows frontal sections of some licuri fiber strands separated in a disorderly way due to the type of fabric and impregnating.



(a) (b)

Figure 10. Composite Microstructure: (a) Longitudinal section of the lamina; (b) Transverse section of the lamina.

4.2. Longitudinal Uniaxial Tensile Test of the Composite

The uniaxial tensile tests where stress was applied parallel to the fibers (longitudinal) show that the scattering of stress and strain (standard deviation) values in the results were higher for maximum strain than longitudinal elastic modulus, especially after the initial load of damage (matrix fracture), as shown in figure 11. The scattering for maximum stress (fracture stress) and elastic modulus was 2.26 and 0.23, respectively. Longitudinal elastic modulus (measured in the direction of the load) was calculated at a stress and strain interval before damage to the composite.

The maximum stress in longitudinal traction was an average of **36 MPa** while the average longitudinal elastic modulus was **2.48 GPa**. The composite showed a slightly higher tensile strength and longitudinal elastic modulus than that of polyester resin. According to industrial Tecniplast ind. Ltda, the tensile resistance values and longitudinal modulus of polyester resin are an average of **30 MPa** and **2.3 GPa**, respectively.

When these data are analyzed with fracture characteristics, it can be concluded that important parameters such as prior cleaning of the fibers to remove the stalks, eliminating high wax content, control of strand linear density in fabric production and the manufacturing process used may cause higher mechanical response values in the composite. Controlling these parameters ensures better impregnating and improved quality in the fiber/matrix interface, which is responsible for the transfer of internal stress to the composite.

An impregnating reinforcement by the matrix, a situation that results in an interface without manufacturing defects such as bubbles or deviations (waviness) in fiber direction, may result in more uniformly distributed mechanical damage. This uniformity makes the final fracture characteristic consistent with that of materials considered “brittle” under normal stress. This “brittle” behavior is characterized by the section fracture at 90° of load application, where normal stress occurs, as with reinforced plastics.

All the mentioned comments are relevant for the fracture observed in the longitudinal tensile tests of the composite. Deviations initially occurred in fiber placement due to the handcrafted manufacture of the fabric and were later compounded by the manual manufacturing process. This resulted in the irregular behavior of the final fracture, as shown in figures 12 and 13.

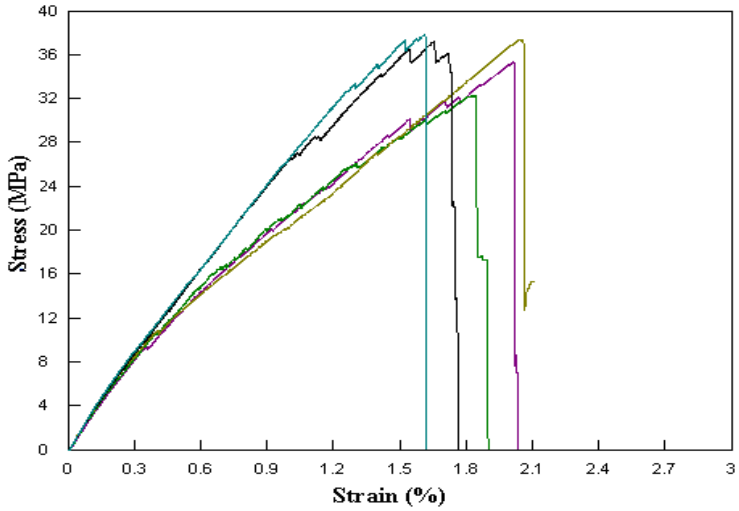


Figure 11. Stress x Strain Diagram – Longitudinal uniaxial tensile test.

Figure 12. Longitudinal uniaxial tensile fracture in the test specimen.

Figure 13. Different characteristics of the final fracture – Longitudinal uniaxial tensile test.

Figures 12 and 13 show that the different final fractures were responsible for the scattering in the data of stress x strain curves after intense load application (see figure 11). The fracture also began in a flat section (consistent with the fracture of brittle material), but followed a different course when it encountered fibers deviating from the load direction, that is, it preferred resin-rich areas that offered less resistance to its propagation. These facts directly influenced the final mechanical response of the composite.

All the specimens showed intense transverse microcracking along their entire length before the final fracture. The fracture process is shown in figure 14.

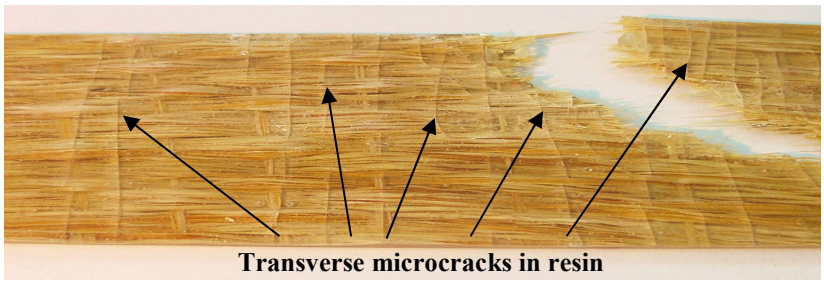


Figure14. Transverse microcracks in the matrix –Longitudinal uniaxial tension.

4.3. Transverse Uniaxial Tensile Test of the Composite

Since this study was conducted comparatively between several fiber/matrix systems (Cahn, 1997), the uniaxial tensile test with load applied perpendicular to the fiber of the lamina can be used to qualitatively measure the strength of the fiber/matrix interface. Although the results of the tests performed with the composite are not part of a comparative study, they show weak interfacial adherence with respect to both mechanical properties and fracture characteristics.

Figure 15 shows the Stress x Strain diagram. Mean maximum transverse stress values of **2.79 MPa** and transverse elastic modulus of **1.9 GPa** were obtained.

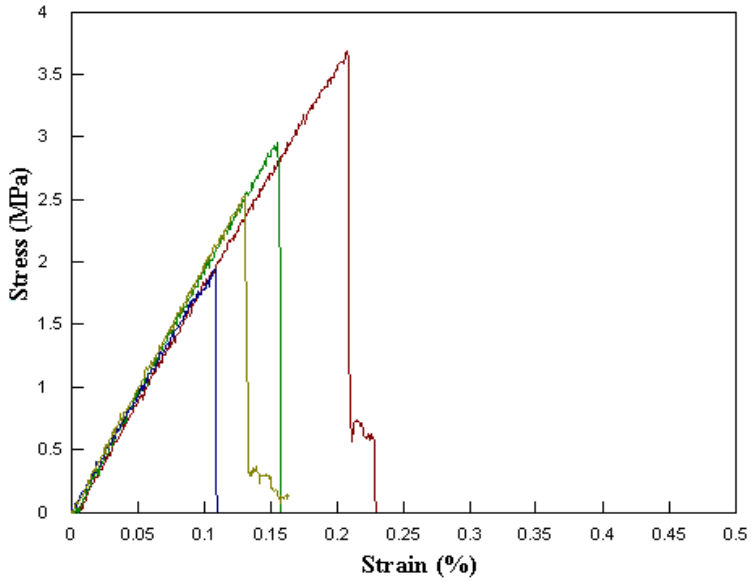


Figure 15. Stress x Strain Diagram – Transverse uniaxial tensile test.

The deviation patterns found (equal to the longitudinal tensile test but higher for strength than for the elastic modulus) were 0.73 and 0.078 for maximum stress and elastic modulus, respectively.

Since maximum transverse ultimate stress is approximately (mean value) 7.73% of maximum longitudinal tensile ultimate stress, the presence of the fibers has a negative effect on the composite (Hull, 1988), given that stress cannot be transferred between the matrix and the fiber (weak interface).

This fact is confirmed by the characteristic fracture observed in the test, since it was caused by a single crack in the resin, as shown in figure 16.

Microscopic analysis of the fracture showed no cracks in the matrix along the length of the test specimen, in contrast to the longitudinal tensile test. The final fracture was completely uniform and flat, corresponding to the characteristic of brittle materials under normal stress.

Figure 16. Flat fracture obtained in transverse uniaxial tensile tests.

5. CONCLUSIONS

Licuri fibers show a similar microstructure to other fibers from plant leaves. The high percentage of cellulose microfibers leads to good results in mechanical properties; however the high wax content may hinder the impregnating process in obtaining polymer composites.

The lignin content was significantly higher than that of other fibers with the exception of coconut fiber. The high lignin content gives durability to the fiber, as well as firmness and structural stiffness. Studies show that lignin acts as a compatibilizing agent in the mechanical properties of plant fiber composites, improving their flexibility.

The mechanical properties of the composite under longitudinal tensile testing were higher at maximum stress and elastic modulus when compared to pure polyester resin properties. The manufacture of the reinforcement fabric, the state of the fiber surface and the type of impregnating (manufacturing process) directly influenced the results. This is confirmed by the mechanical fracture in the composite. The influence of these factors on the mechanical properties is more marked in the transverse tensile test. In this type of test the weakness of the fiber/matrix interface is shown by the presence of a single crack in the matrix and consequently responsible for the final fracture of the composite.

In the overall mechanical behavior of the composite, the use of licuri fiber to obtain natural fiber-based plastic is completely viable if special care is taken in prior surface treatment of the fiber and especially in improving the weaving quality of reinforcement.

6. ACKNOWLEDGEMENTS

The authors acknowledge the financial support provided by CNPq and CAPES.

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8. RESPONSIBILITY NOTICE

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