HYBRID COMPOSITES WITH SYNTHETIC AND NATURAL FIBERS: DEGRADATION BY MOISTURE ABSORPTION

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Abstract: *The aim of this work was to develop a hybrid sandwich composite combining synthetic (fiberglass) and natural (jute) fibers. The laminate is composed by orthophthalic polyester resin reinforced by bi-directional fabrics of glass and jute fibers. There is also a central layer of polyethylene fabric (**coremat** firet - xx). Tensile and tree point bend tests, besides the damage mechanism developed in both tests were performed for the composites characterization. Moisture absorption tests were also accomplished having in mind future applications in structural elements in contact with high moisture environments, such as, containers and piping. The mechanical tests were performed for dry and wet conditions. In the wet condition different immersion times in distilled water were utilized: 7, 35 and 90 days, besides the saturated condition. Different immersion times are necessary due to hybridization process, i.e., the synthetic and natural fibers present different behavior in relation to moisture absorption. In the moisture absorption test the hybrid composite presented larger absorption than the commonly observed in fiberglass composites due to the natural fibers, as well as, the **coremat**. It was verified that under wet conditions there was a decrease in the mechanical properties, characterizing the strong influence of the moisture absorption on these properties.*

Key words: *Hybrid composite, glass fibers, jute fibers, moisture absorption.*

1. Introduction

The bio-fibers derived from renewable resources have been contributing positively in the environmental preservation concerning the new materials development. In general, studies developed in this area are limited to reinforced plastics with thermosetting or thermoplastic resins (Joseph *et al*. 2002, Singleton N. *et al*. 2003, Dipa *et al*. 2002, Espert *et al*. 2004), however, there is not always a definition regarding the final product application.

Due to poor mechanical performance of most natural fibers, some limitations were observed concerning the expected final application. In order to prevent these limitations the hybrid composites combining natural and synthetic fibers were developed aimed to associate improvement in the mechanical performance with low production cost (Moe *et al*. 2002, Seena *et al*. 2002).

Hybrid composites have been studied for the last years, however, initially emphasis was given to the ones using synthetic fibers (Mander *et al*. 1981, Fernando *et al*. 1988, Maron *et al*. 1989); only more recently attention has been given to combinations of synthetic and natural fibers (Kalaprasad and Kuruvilla, 1997, Thwe and Liao, 2002).

The aim of this work was to develop a hybrid sandwich composite and to evaluate its mechanical behavior through tensile and the tree point bend tests, besides the damage mechanism developed in both tests. The laminate is composed of orthophtalic polyester resin reinforced with bi-directional fabrics of fiberglass and jute fibers. There is also a central layer of polyethylene fabric (**coremat**). Jute fabric was selected because of its capacity to absorb the same amount of resin as the glassfiber mats and a little more than the glassfiber fabric, during the molding process, thus there is no significant alteration in the production cost. It is worth mentioning that the jute fabric was used "in natura", that means, without any special treatment to improve the interfacial adherence. Having in mind possible applications in structural elements that can have contact with water, such as, piping, reservoirs and boats ballast; the influence of the moisture on the mechanical behaviour of the composite was investigated.

2. Experimental

The composites were manufactured by industrial production using the hand-lay-up technique and the orthophthalic polyester resin as matrix. They are composed of seven layers with the following configuration: $[FJ/FV/FJ/C]_s$. Where ***C*** is the central layer of polyethylene fabric (named **coremat** firet xx), ***FJ*** is the jute fabric (warp: 3617.1 denier, weft: 3245.4 denier) and ***FV*** is the E-glass fabric (450 g/m^2). As final result, 7 mm-thick laminates were obtained. The use of the **coremat** in the industry has the objective to increase the laminate stiffness.

Tensile and the three point bend tests were performed according to ASTM D3039-00 and ASTM D790-96, respectively, in a Shimadzu AG-I testing system. Tests were carried out at room temperature and a minimum of five specimens were tested. Prior to the tests microscopic analysis were carried out to identify any flaw in the specimens due to the cut and molding processes.

Water absorption tests were carried out according to ASTM D570-81. Five specimens were tested. Firstly, the specimens were weighed and immersed in distilled water. At pre-determined intervals of time the specimens were emerged, weighed and placed again into the water. This procedure was repeated up to saturation, until there was no significant variation in the mass of the specimens tested.

After obtaining the saturation curve (Weight increase versus Immersion time) immersion times were selected for the tensile and the three point bend tests, as follows: 7, 35, 90 and 330 days, of which 330 days (approximately 11 months) is the saturated condition. The damage mechanism analysis was carried out with the tested specimens, using an optical microscope.

3. Results and Discussion

3.1. Water absorption test

The plot of water absorption versus immersion time is shown in Fig. 1. Saturation was reached after 330 days (11 months) with 7.64 % weight increase. It was observed that the absorption was faster in the first 30 days, while after the 7th day the absorption content corresponded to 2.86 %, comprising 37.43 % of the total.

For fiberglass composites the saturation was reached after 2 or 3 immersion months with a weight increase of 1.5 % (Aquino and Margaria, 1997). Consequently, it could be observed that the greatest absorption of the hybrid composite under study was mainly due to the hydrophilic nature of the vegetal fibers. The **coremat** had less or no significant influence on this process.

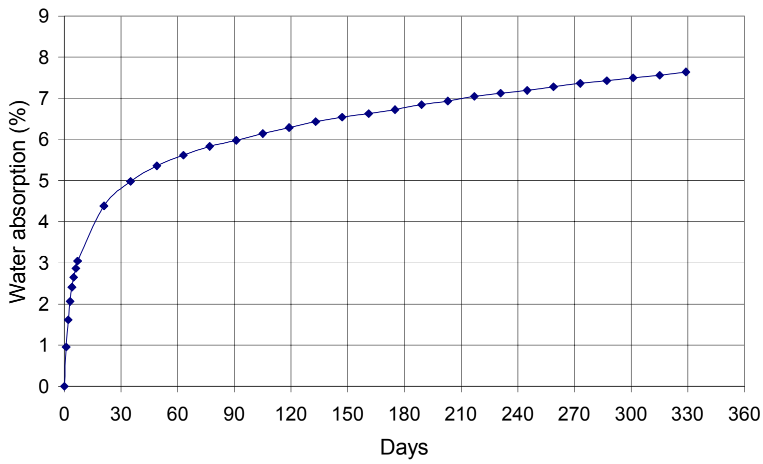


Figure 1. Water absorption (weight %) versus immersion time.

3.2. Tensile test

In hybrid composites the moisture absorption depends on the individual characteristics of each fiber, as different fibers present different absorption contents. Aiming at knowing the effect of the moisture absorption of each fiber in the composite final response, different immersion times for the tensile and the three points bend tests were selected. The immersion times were selected according to the water absorption curve presented in Fig. 1. The immersion times and correspondent absorption contents (% weight) were: 7 days (2.86%), 35 days (4.98%), 90 days (6.02%) and saturated condition (7.64%). The influence of the different moisture absorption contents of the glass and jute fibers could be observed in the tests with 90 days and in the saturated condition, because 90 days is the time limit for the fiberglass saturation (Aquino and Margaria, 1997).

The Stress x Strain curves for dry and wet conditions are shown in Fig. 2. The composites, in dry or wet condition, presented a linear behavior as observed in the curves. This behaviour agrees with the one usually observed in thermosetting resin composites (e. g. polyester and epoxy) and synthetic fibers. It can be noted that the blend of natural and synthetic fibers did not alter this linear behavior. This characteristic facilitates any modeling process, as the one used for the interlaminar stress determination.

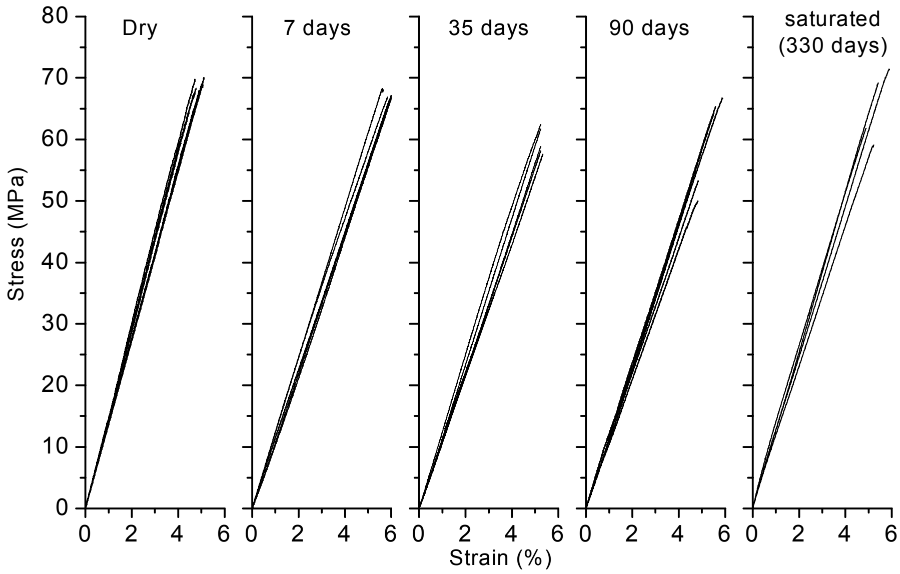


Figure 2 – Stress x Strain curves for composites in dry and wet conditions.

The mechanical properties of tensile strength, Young modulus in the loading direction and elongation are shown in Fig. 3. The dispersion (absolute deviation) is larger for the wet condition than for the dry condition. However, the dispersion values are considered low for hybrid composites with synthetic and natural fibers which present different elastic properties.

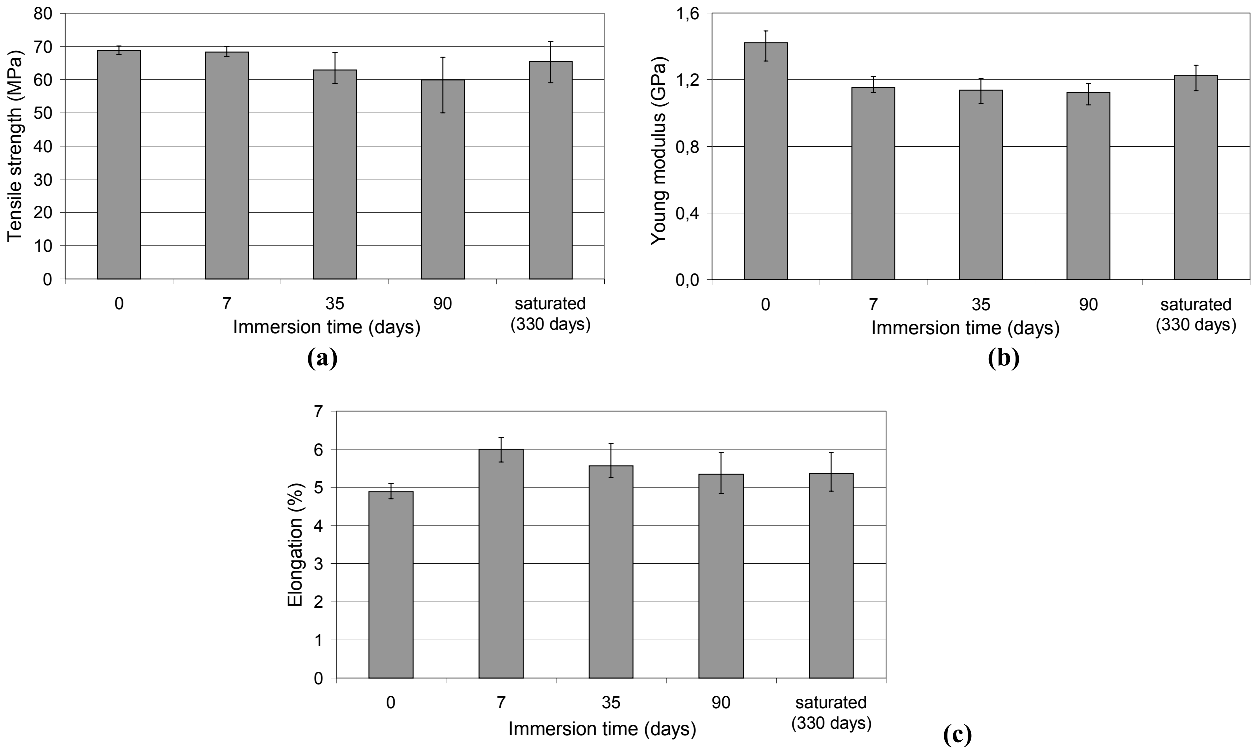


Figure 3. Mechanical properties of the tensile test: (a) tensile strength, (b) Young modulus in the loading direction, (c) elongation.

The composite in the wet condition showed an initial decrease in the tensile strength and Young modulus in relation to dry condition. However, a recovery of these properties was verified for the saturated condition. This behavior can be explained by the different saturation times of fiberglass and jute fiber.

Polymeric composites with polyester resin and fiberglass present saturation time around 75 days with reduction in tensile strength and Young modulus (Aquino and Margaria, 1997). On the other hand for natural fibers composites, some increase in these properties is commonly noted, even though highest saturation time occurs, around 300 days. This phenomenon is related to fiber swelling that promotes an increase in the area of load support and consequently increases the mechanical resistance. This behavior remains until the beginning of the fiber degradation, when progressive resistance loss is observed. This way, for the hybrid composite under study the initial decrease (7, 35 and 90 days) in the tensile strength and Young modulus was due to fiberglass saturation and the subsequent increase (saturated condition) due to jute fiber saturation.

Although the mechanical properties decrease under wet condition in relation to dry condition, the tensile strength and Young modulus presented order of magnitude acceptable for many structural elements, especially when compared to laminated composites with fiberglass mats (low modulus) and same number of layers (Margaria, Nascimento and Aquino, 1996). Besides, the values of the tensile strength and Young modulus in dry condition were about 30% smaller than that of a fiberglass composite with equal configuration (Silva, Freire Júnior and Aquino, 2000).

Regarding the elongation results an increase under wet condition in relation to dry condition was observed. This increase was more significant for 7 days followed by a gradual decrease until stability. The tensile strength and Young modulus increase for longer immersion times could have influenced this behavior.

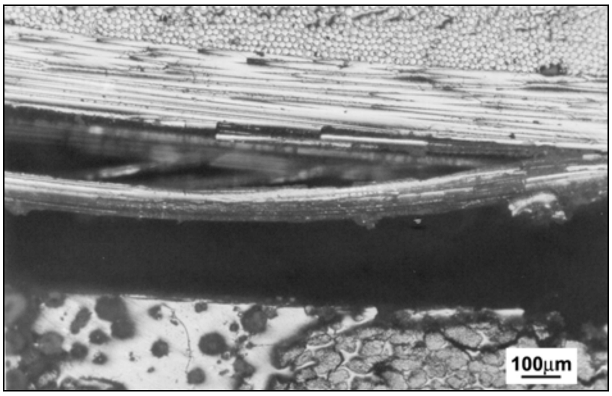
Table 1 presents the percentages of decreases (-) and increases (+) in the tensile properties, in relation to dry condition, for all immersion times. It is observed that the highest percentage was of 23% corresponding to elongation increase for 7 immersion days. The other percentages are lower and do not significantly affect the strength and stiffness properties, so that, it is possible the use of the hybrid composite even in humid environments.

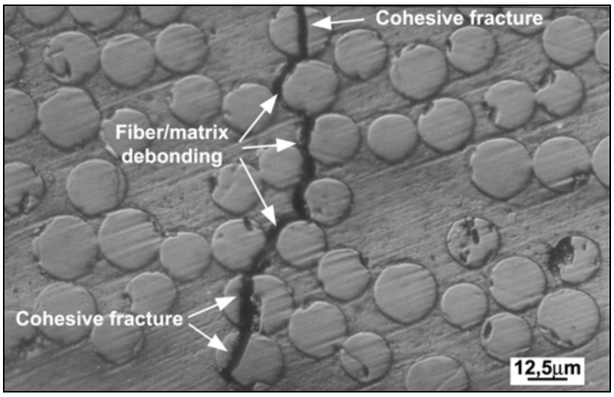
Table 1. Percentages of decrease (-) and increase (+) in the tensile properties in relation to dry conditions.

Condition	Tensile Strength	Young Modulus	Elongation
Dry	68.6 MPa	1.2 GPa	4.9 %
7 days	- 0.5 %	- 7.4 %	+ 23 %
35 days	- 8.23 %	- 9. 01 %	+ 14.14 %
90 days	- 12.62 %	- 13.5 %	+ 9.53 %
Saturated (330 days)	- 4.66%	- 0.35 %	+ 10.0 %

3.2.1. Damage mechanism

In general, the specimens under dry and wet conditions presented the same damage characteristics, which are:
- Delamination between the layers of fiberglass fabric and jute fabric (Fig. 4a).
- Micro-cracking in the matrix, fiberglass and jute fibers, transverse to loading direction. These micro-cracking arise, in some cases, from defects as voids. Figure 4b shows the micro-cracking propagation in the fiberglass layer.





(a) **(b)**

Figure 4. (a) Delamination between the layers of fiberglass fabric and jute fabric (35 immersion days). (b) Micro-cracking in the fiberglass layer with fiber/matrix debonding and cohesive fracture (7 immersion days).

It is worth to mention that for specimens with longer immersion times the delamination extension is reduced, on the other hand, the number of transverse micro-cracking is increased. Another important point was the excellent adherence between the jute fabric and the **coremat**; while delamination was not verified under the dry or wet condition.

It could be concluded that the moisture absorption did not influence the main characteristics of the damage mechanism, on the other hand it influenced the propagation process. For a better analysis of the direct influence of the damage propagation on the mechanical properties, it would be necessary the damage monitoring during the test and not only the fracture specimens analysis.

3.3. Three point bend test

The main characteristic of the flexural tests was the "premature" shear fracture of the **coremat**, in the neutral line of the laminate, predominant for most of the specimens, under dry or wet condition. The shear fracture caused loss of load support capacity of the composite thus the subsequent stress was unable to promote the flexural fracture, that means, the fracture in the outer fibers on the tensile face of the laminate.

The Maximum flexural stress, Modulus of elasticity and Maximum strain were determined according to ASTM D790-90. It is important to point out that the calculations refer the moment of the shear fracture since there was no flexural fracture.

Figure 5a shows load-deflection curves for some specimens under dry and wet conditions. The curves are similar for both conditions and present a linear initial behavior which allows the calculation of the modulus of elasticity.

The curves for the specimens of 7 days immersion are shown in Fig. 5b. These presented a peculiar behavior since for some specimens flexural fracture was observed. From Fig. 5b it was possible to estimate the flexural resistance decrease due to premature shear fracture of the **coremat**.

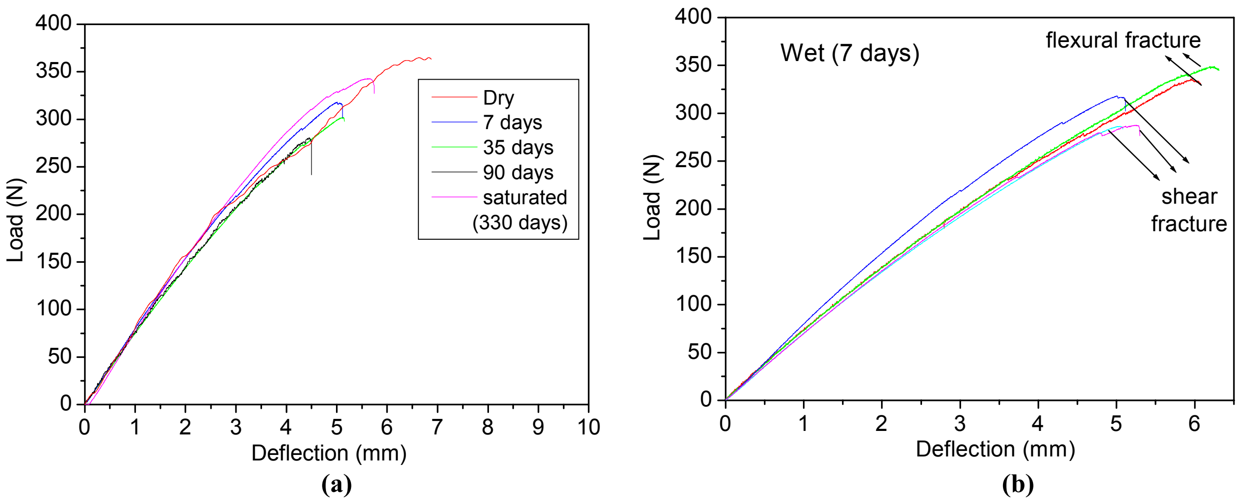


Figure 5. (a) Load-Deflection curves for some specimens in dry and wet conditions.
(b) Load-Deflection curves for specimens with 7 immersion days.

The flexural mechanical properties (calculated at the moment of the shear fracture) are shown in Fig. 6. There was a gradual decrease of the Maximum flexural stress according to immersion time. Differently from the tensile test there was not resistance recovery for the saturated condition. The Maximum flexural stress shows similar values for specimens with 90 immersion days and for the saturated condition. This behavior is probably related to premature shear fracture of the **coremat** that masked a possible recovery for the saturated condition.

Table 2 presents the percentages of decreases (-) and increases (+) in the flexural properties, in relation to dry condition, for all immersion times. It could be observed that, in general, the decreases in the Modulus are lesser than the ones for Maximum flexural stress. This behavior was expected since the objective of the **coremat** addition is to increase the laminate stiffness, as described previously.

Regarding the Maximum strain (measurements at the moment of shear fracture) the decreases are especially related to strong reduction in the Maximum flexural stress for longer immersion times, since the Modulus of elasticity remains practically stable.

Table 2. Percentages of decreases (-) and increases (+) in the flexural properties in relation to dry condition.

Condition	Maximum flexural stress	Modulus of elasticity	Maximum strain
Dry	113.56 MPa	5.5 GPa	4.9 %
7 days	-9.0 %	-8.0 %	+7.3 %
35 days	-15.3 %	-5.3 %	-4.2 %
90 days	-20.8 %	-6.4 %	-9.1 %
Saturated (330 days)	-22.2 %	-10.4 %	-9.3 %

The dispersion (absolute deviation showed in Fig. 6) was quite high compared to non hybrid composites. As already mentioned this behavior is related to the association of synthetic and natural fibers, which promote a mismatch of elastic properties in the interface between the layers. For specimens with 7 immersion days the high dispersion is also related to the difference between the magnitude of the shear and the flexural stresses (see Fig. 5b).

It could be concluded that despite the premature shear fracture in the **coremat** a better performance of the composite in the flexural loading compared to the tensile loading is evident. However, the low shear resistance of the **coremat** can hinder possible structural applications in components submitted to high flexural loading.

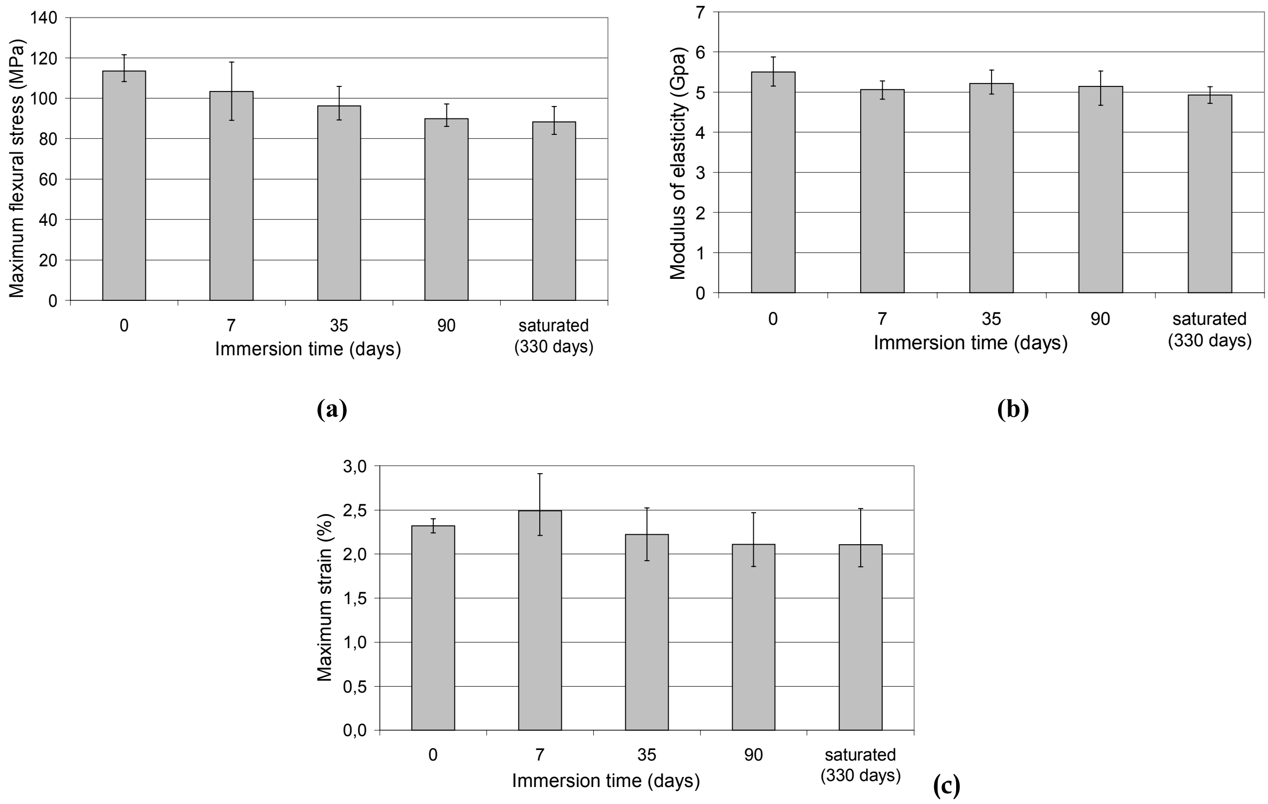


Figure 6. Properties of three point bend test: (a) Maximum flexural stress, (b) Modulus of elasticity, (c) Maximum strain.

3.3.1. Damage Mechanism

In the analysis of damage mechanism of the specimens submitted to flexural test the "premature" shear fracture of the **coremat**, in the neutral line of the laminate, as shown in Fig. 7, was the main characteristic. Shear fracture was predominant for most of the specimens, under dry or wet condition.

Under dry condition, besides the shearing fracture in the neutral line, some transverse micro-cracking as well as longitudinal splitting were observed. These represent isolated events in the jute fibers layers adjacent to **coremat**.

Under wet condition, transverse micro-cracking and longitudinal splitting were observed in the matrix, fiberglass (fiber/matrix debonding and cohesive fracture) and jute fiber (only fiber/matrix debonding). Figure 8 shows examples of these events. The damage is predominant on the tensile-side and in the central area of the specimen. In the compressive-side the number of events is much lower and limited to micro-cracking in the matrix and jute fiber; there is no damage in the fiberglass. Regarding the immersion time no influence in the damage mechanism or its intensity (number of events) was verified.

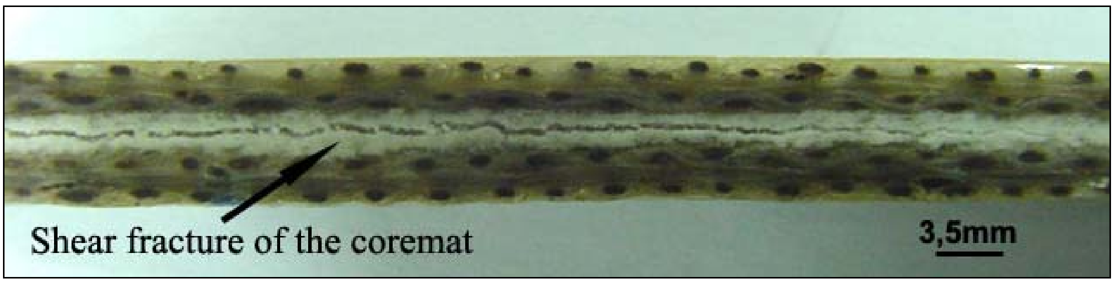


Figure 7. Lateral view (thickness) of a three point bend specimen with shear fracture in the **coremat** (saturated condition).

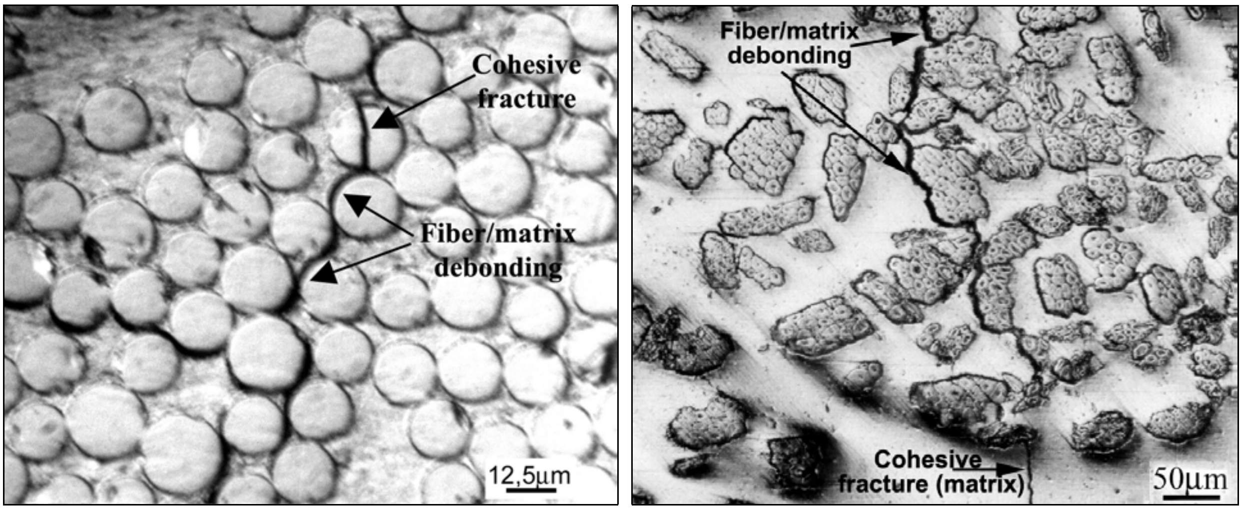


Figure 8. (a) Cohesive fracture and in the fiberglass layer (7 immersion days).
(b) Fiber/matrix debonding and cohesive fracture in the matrix in the jute layer (35 immersion days).

4. Conclusions

The tensile mechanical properties showed that the hybrid composite with synthetic and natural fibers had a good mechanical performance, thus it can be recommended for structural applications particularly for components subjected to light or moderate loading. The values of the tensile strength and Young modulus in dry condition were about 30% lower than those of a fiberglass composite with equal configuration.

The moisture absorption of 7.64 % (saturated condition) in 11 months of immersion is considered high when compared to absorption of most composites with fiberglass (1.5 %).

The moisture absorption caused loss in the mechanical properties of the laminate for both loading types, i. e., tensile and three point bend.

Regarding the damage mechanism the moisture absorption did not have any influence on its main characteristics, on the other hand some influence on its propagation process was observed.

The main characteristic of the flexural test was the "premature" shear fracture in the **coremat** causing loss in the flexural resistance.

Despite the premature shear fracture in the **coremat** the best performance of the composite in the flexural loading compared to that of the tensile loading, for both wet and dry conditions, it was evident. However, the low shear resistance of the **coremat** can hinder possible structural applications in components submitted to high flexural loading.

5. References

Aquino, E. M. F. and Margaria, G., 1997, "Influence of moisture absorption on the mechanical properties of polyester/fiber glass-E composites", Proceedings of Second international congress on metallurgical and materials technology, São Paulo, Brazil.

ASTM D3039-00, 2000, "Standard Test Methods for Tensile Properties of Polymer Matrix Composites".

ASTM D570-81, 1981, "Standard Test Methods for Water absorption of plastics".

ASTM D790-96a, 1997, "Standard Test Methods for Flexural Properties of Unreinforced and Reinforced Plastics and Electrical Insulating Materials".

Dipa, R. et al., 2002, "Dynamic mechanical and thermal analysis of vinylester-resin-matrix composites reinforced with untreated and alcali-treated jute fibres", Composites Science and Technology, vol. 62, pp. 911-917,.

Espert, A., Vilaplana, F., Karlsson, S., 2004, "Comparison of water absorption in natural cellulosic fibres from wood and one-year crops in polypropylene composites and its influence on their mechanical properties", Composites Part A, vol. 35, No. 11, pp. 1267-1276.

Fernando, G. et al., 1988, "Fatigue behavior of hybrid composites: part 1 carbon/kevlar hybrids", Journal of Materials Science, vol. 23, pp. 3732-3743.

Joseph, P. V. et al., 2002, "Environemntal Effects on the Degradation Behavior of Sisal Fibre Reinforced Polypropylene Composites", Composites Science and Tecnology, vol. 68, pp. 1357-1372.

Kalaprasad, G. and Kuruvilla, J., 1997, "Influence of short glass fiber addition on the mechanical properties of sisal reinforced low density polyethylene composites", Journal of composite materials, vol. 31, No. 5, pp. 509-527.

Mander, P.W. et al., 1981, "The strength of hybrid glass/carbon fibre composites: part 1: failure strain enhancement and failure mode", Journal of Materials Science, vol. 16, pp. 2233-2245.

Margaria, G., Nascimento, R. M. and Aquino, E. M. F., 1996, "Estudo da umidade na resistência e fratura de compósitos tubulares", Anais do IV congresso de Eng. Mec N/NE, vol 1, pp. 133-137.

Maron, G. et al., 1989, “Fatigue behavior and rate dependent properties of aramid fibre/carbon fibre hybrid composite”, Composites, vol. 20, No. 6, pp. 537-544.
Moe, M. T. et al., 2002, “Effects of environmental aging on the mechanical properties of bamboo-glass fiber reinforced polymer matrix hybrid composites”, Composites. Part A, vol. 33, pp. 43-52.
Seena, Y. et al., 2002, “A Comparison of the Mechanical Properties of Phenol Formaldehyde Composites Reinforced with Banana Fibres and Glass Fibres”, Composites Science and Technology, Vol. 62, pp. 1857-1868.
Silva, C. D., Freire Júnior, R. C. S. and Aquino, E. M. F., 2000, “Influência da presença de fibras naturais em compósitos híbridos tipo sandwich”, Anais do Congresso Nacional de Eng. Mecânica – CONEM, Belém, Brazil, pp. 1-10.
Singleton, A. C. N. et al., 2003, “On the Mechanical properties, deformation and fracture of a natural fibre/recycled polymer composite”, Composites- Part B, vol. 34, pp. 519-526.
Thwe, M. M. and Liao, K., 2002, Composites: Part A, vol. 33, pp. 43-52.

6. Responsibility notice

The authors: Aquino, E. M. F.; Silva, R. V., Rodrigues, L. P. S. and Oliveira, W. are the only responsible for the printed material included in this paper