

POLYMERIC BIOCOMPOSITE MECHANICAL PROPERTIES UNDER INFLUENCE OF MOISTURE

Ana Claudia de Melo Caldas Batista

Federal University of Rio Grande do Norte/PPGEM Natal-RN, Brazil
aninha_olem@hotmail.com

Kilder César de Araújo Ribeiro

Federal University of Rio Grande do Norte/PPGEM/DEART/Design Natal-RN, Brazil
kildercesar@gmail.com

Eve Maria Freire de Aquino

Federal University of Rio Grande do Norte/PPGEM Natal-RN, Brazil
eve@dem.ufrn.br

***Abstract.** With the growing demand for technology appeared to need to develop materials with more specific properties and meet the new market demands. In this context, polymer composites have been presented as a good alternative to the conventional use of materials, especially the biocomposites, a combination of different components where at least one has a natural origin, usually vegetable fibers. Follows the development of these materials the concept of reducing the harmful effects to the environment caused by its production and disposal. Therefore, this article proposes to study the mechanical performance in terms of strength and stiffness of a laminated polymer biocomposite, as sandwich panel under severe conditions of moisture absorption. The ability of moisture absorption was observed under two conditions of service: distilled water and salt water, and then comparing the mechanical performance under uniaxial tensile and flexure at three points loading between samples in the dry state (natural) and saturated humid. The study intends to apply this biocomposite as a coating or partitions for environments under the action of weather. The panel, made industrially, comprised nautical resin reinforced with layers of woven jute bidirectional and a core of wood waste collected in sawmills in the region.*

***Keywords:** Polymeric Composites, Sandwich Panel, Humidity, Mechanical Properties.*

1. INTRODUCTION

Sustainable development is a recurrent theme nowadays and, because of this issue arises the need for searching new solutions that aim at minimizing environmental degradation. The use of green wood has been causing damage to nature due to deforestation with consequent biodiversity degradation.

Sawmills only use about 1/3 (one third) of the trees, and the waste is usually discharged inappropriately, generally being thrown away in the environment. Despite the reduced toxicity of wood waste, its reuse for new applications adds up to the constant search for meeting the society's demands. In the search for perfecting wood reuse, the number of researches has grown in several areas, such as obtaining chipboards, MDF, MHF and even materials such as reinforced plastics and biocomposites.

Thus, a material that would harm the environment in a lower level and which displays a higher compatibility degree with wood would be a great solution. The biocomposite proposed in this study, even though not biodegradable (due to its resin's nature), may be recycled once woven jute and wood waste are used, and being the woven jute characterized as renewable. The choice of nautical resin is due to a possible application of the laminated biocomposite for coating panels and partition walls in internal and/or external environments subjected to severe conditions of the humidity absorption.

The laminated biocomposite is displayed in the form of a sandwich panel, having as its matrix the isoftalic polyester resin with **NPG** - Neo Pentil Glicol, enhanced with 04 (four) layers of bidirectional woven jute and a core of processed wood waste of different types (also known as wood-floor), constituting a total of 05 (five) layers. The laminated biocomposite, manufactured industrially, was obtained through the hand lay-up process.

The mechanical performance of such biocomposite, characterized by the determination of its strength and stiffness properties, was determined with uniaxial tensile and three-point flexural tests. The specimen followed the ASTM D 3039-08 norm for the uniaxial tensile tests, ASTM D5229/D 570 – 98 for the moisture absorption test and ASTM D 790-07 for the three-point flexural test.

For a better understanding on how moisture absorption can affect the mechanical behavior of the laminated biocomposite, it was necessary to carry out a comparative study of these properties in the conditions of dry state (Natural Laminate Biocomposite – **NLB**) and the saturated humid state, in this case in distilled water (Water Laminate Biocomposite - **WLB**). Also, we carried out the study of moisture absorption capacity of the biocomposite in relation to seawater (Seawater Laminate Biocomposite - **SLB**).

2. MATERIALS AND METHODS

2.1 Materials used in the laminated composite

The following materials were used for manufacturing the laminated biocomposite: the reinforcement used was the bidirectional woven jute “in natura”, and stored away from sunlight and moisture for two months before going under the same impregnation process.

The biocomposite had as its non-reinforcing load (constitution of the panel’s core) the wood wastes from sawmills and other industries in the field. The wood waste, as well as the woven jute, was protected from the sun and humidity, in this case, being stored in an 80°C greenhouse for 24h, afterwards, processed and classified in terms of granulometry in compliance with the ASTM E11-95 norm. It is worth highlighting the waste’s hybrid characteristic, since it is derived from different types of wood.

The matrix was composed of non-saturated isoftalic polyester resin with an additive (**NPG**), known as nautical resin – RESAPOL LP 10134, provided by Reichhold Technology Brazil.

The resin was stored and prepared for its application in the *Tecniplas Tubos e Conexões Ltda* plant. The gel time was adjusted to 15 minutes, which is appropriate for the process. Since it is a direct function with room temperature, concentrations of accelerator at 1% of Cobalt Octoate at 6% of Metal and 1% of Methyl-Ethyl-Ketone Peroxide as a catalyzer were used, according to the limits informed on the technical bulletin provided by the manufacturer.

2.2 Manufacturing the sandwich panel and specimen

In the manufacturing of the laminated biocomposite through hand-lay-up, a 1,0m² panel with 10,5mm of thickness was obtained. In figure 1, it is possible to visualize, through a 3D simulation, the panel’s final configuration with all of its components. It is highlighted that the transparent resin layers do not separate during the confection process, since they are directly impregnated in the panel’s reinforcement layers. In the case of the woven jute, it was positioned with its fibers in a 0/90° direction, in relation to the load application direction.

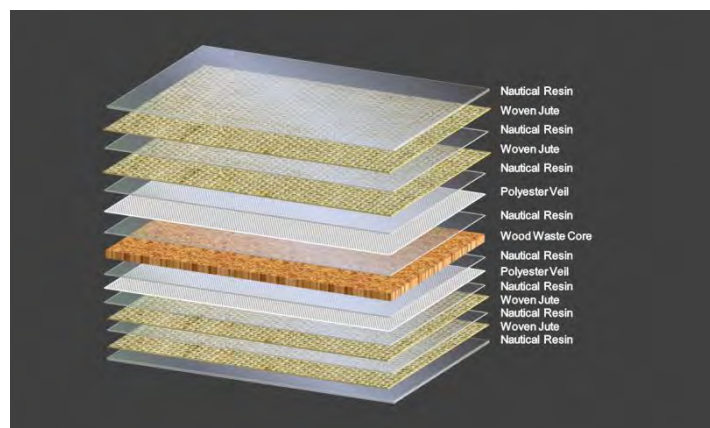


Figure 1. Sandwich panel configuration (biocomposite).

In order to perform the moisture absorption tests, 06 (six) specimen were manufactured for immersion in distilled water (**WLB**) and 06 (six) specimen for seawater (**SLB**). In the case of uniaxial tensile tests, 08 (eight) specimen were manufactured without moisture effect (natural state) (**NLB-T**) and 08 (eight) specimen under moisture effect (**WLB-T**). For the three-point flexural test, 08 (eight) specimen were prepared without moisture effect (natural state) (**NLB-F**) and 08 (eight) specimen under moisture effect (**WLB-F**).

The specimen used in the uniaxial tensile and three-point flexural tests under the effect of moisture were submerged along with the specimen in the moisture absorption test, for the specific case of distilled water, until they are saturated and, afterwards, the tests were performed.

For cutting the specimen, a diamond cut-off wheel was used (DIFER D252). It was dry in order to avoid tearing fibers or other types of damages. For obtaining better cuts, irregular “fins” of 5.0cm were removed from the sides of the panel, aiming at eliminating possible discontinuities and bad finishing, such as variations in thickness and a bigger number of internal defects derived from the manufacturing process.

After obtaining the appropriate dimensions of the specimen, the procedures of metallographic sanding and polishing were performed in the sides cut with the wheel. In order to do so, water sandpaper numbers 150, 180, 240, 320, 400, 600 and 1200 were used, and the polishing was done in a motorized polishing machine, using the alumina of 0,01 μm as the abrasive agent.

3. RESULTS AND DISCUSSIONS

Regarding the moisture absorption test, we will showcase the results of the tests performed in distilled water and seawater, followed by a comparative study between them.

Regarding the uniaxial tensile and three-point flexural tests, the results will be displayed for natural state (without the effect of moisture), for the saturated humid state, and a comparative study between them.

3.1 Moisture absorption

Natural fibers, when in humid environments, have a big capacity of absorbing water, due to the presence of cellulose in their constitution. On the other hand, the nautical resin is characterized by having polymers with high molecular weights which result in a material with low sensitivity to water.

Literature related to the studies that involve moisture absorption in polymeric composites with a synthetic fiber base points out that the average saturation time of moisture in these materials occur between 02 and 03 months (Oliveira, 2007, 1997; Aquino, 1996). However, for composites with natural fibers this amount of time is considerably longer. With this composite, the necessary amount of time for saturation to be achieved in both situations was approximately 08 months.

The natural fibers are composed by several layers with different characteristics and thicknesses, placed around the lumen. This justifies the fact that the absorption test takes longer when compared to synthetic fibers, because it requires more time for the water to penetrate in all layers. Additionally, the layers have the ability of absorbing water also through the lumen by capillarity, which is an effect that, even though it is small when compared to total absorption, it may interfere in the process.

This study's challenge consists of analyzing, initially, how the nautical resin behaves in relation to the impregnation with natural fibers. That is, as a barrier in the absorption process, derived from a good quality in the fiber/matrix's interface. Subsequently, this result will be quantified regarding the possible losses in the composite's mechanical properties with reference to its registered absorption degree.

Besides the presence of natural fibers, the biocomposite has one more constituent with lignocellulosic properties, which is the case of wood (Silva, 2008). In this sense, the expected result displays a relatively high level of absorption when compared to the level of absorption involving composites, whether they are reinforced only with natural fibers or hybrid composites that involve natural and synthetic fibers (Oliveira, 2007). It is also expected that, somehow, the nautical resin ends up influencing the results.

3.1.1 Moisture absorption using distilled water (WLB)

Regarding the behavior of moisture absorption of **WLB** during 08 months of immersion (saturated state), we were able to notice that, after this amount of time, and saturation state was achieved with an increment in total weight of 4.14%. It is also noticeable that the bigger absorption percentage happened in the first two months of immersion, once the absorption percentage in the first week was 1.55%, which corresponds to 37.44% of the total absorption. The mean determined values in all absorption tests correspond to measurements of 03 specimens, according to the technical norm's requirements.

Some papers on this subject show that composites which are reinforced only with natural fibers absorb, generally, a superior percentage. Dantas (2011) studied a composite with five layers of bidirectional woven jute as a reinforcement and terephthalic polyester resin as the matrix, which achieved the absorption of 5.53% of the total weight. Whereas, the same study carried out in hybrid composites with bidirectional woven jute and glass fibers (three layers of jute with two bidirectional layers of glass fibers) with the same terephthalic polyester resin as the matrix, demonstrated the absorption of 2.48%. In this case, the glass fiber layers were external and had the function of diminishing the moisture absorption capacity in the laminated composite.

Oliveira (2007) studied a hybrid laminated composite with bidirectional woven jute and glass fiber layers. However, with only two layers of jute and four layers of glass fibers, it absorbed 4.04%, using the orthophthalic polyester resin as matrix. The absorption of 4.14% can be noticed. Despite the fact that the absorption of hybrid composites (that use synthetic fibers) is superior, it displayed an inferior absorption when compared to the ones that contained only natural fibers, showing that the nautical resin influences directly in moisture absorption.

3.1.2 Moisture absorption using seawater (SLB)

Regarding the behavior of moisture absorption of **SLB** in the 08 months of immersion, we noticed that, after this period, the saturation state was achieved and the increment in total weight was 3.44%. It is also noticeable that the bigger absorption percentage happened in the first two months of immersion, whereas in the first week the absorption percentage was 1.31%, corresponding to 38.08% of the total absorption.

3.1.3 Comparative study (WLB X SLB)

Figure 2 displays the behavior of moisture absorption of the **WLB** panel in both of the immersion means that were tested. We noticed that, after the saturation state was achieved, distilled water displayed a 16.91% bigger absorption than seawater.

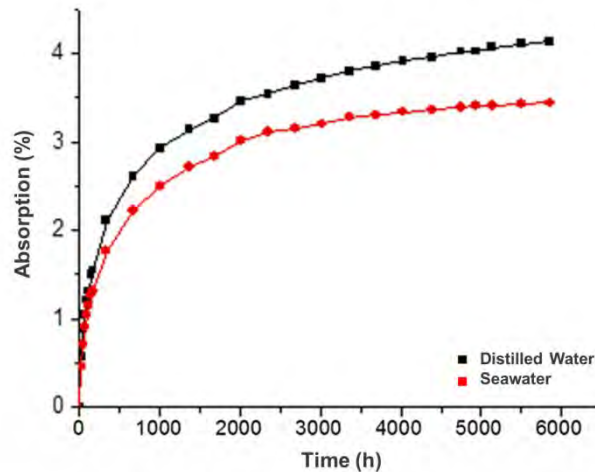


Figure 2. Comparative curves of moisture absorption - **WLB X SLB**.

It can also be noticed that the higher percentage in the difference of absorption was in saturation, that is, in the first week, the distilled water had a superior absorption of 15.48%. This fact had already been observed in a paper by Rodrigues (2012), who studied two composites reinforced with glass fibers, one of them was a hybrid composite reinforced with fibers of curauá (*Ananás erectifolius*). It was verified that, in both cases, immersion in distilled water provided higher moisture absorption and, consequently, higher losses in the mechanical properties of the hybrid laminated composite.

In the comparative analysis, we have also observed that absorption, in the case of seawater, is smaller, and that is justified by the accumulation of NaCl ions in the fiber's surface, which increases with time and prevents water diffusion in the specimen. It is interesting to highlight that, even though the absorption percentage has been smaller for seawater, the saturation time was practically the same (Silva *et al*, 2008 *apud* Davies *et al* 2001).

The big difference of absorption between the means tested justifies the sequence of mechanical uniaxial tensile and three-point flexural tests for the panel which was saturated only through immersion in distilled water, since the study's objective is to verify means and conditions that degrade the material the most, in order to possibly determine the best conditions for the industrial use of the biocomposite.

3.2 Uniaxial tensile in natural state (NLB-T)

Regarding the uniaxial tensile test for the **NLB-T** specimen, figure 3 displays the Stress x Strain curves for this laminated composite.

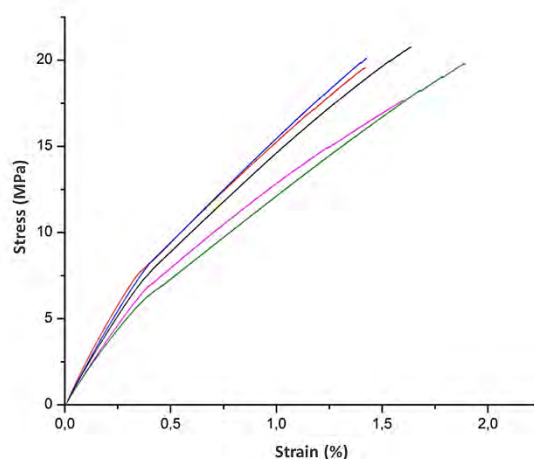


Figure 3. **NLB-T** stress x strain curves.

The material shows a “fragile” behavior until fracture. That is, the surface runoff phenomenon, characteristic of materials with ductile behavior, is not displayed. The inclination observed in the initial region of the test corresponds to the damage's initial load, which is characterized by brittleness in the matrix.

The average values, as well as its respective dispersion obtained for ultimate tensile strength, deformation and bursting and for the longitudinal elasticity modulus (measured in the direction of load application) are displayed in table 1. The dispersions refer to the absolute difference in percentage between the maximum and minimum values obtained in each test, for each parameter analyzed. It must be noted that, for all tests, the elasticity modulus values are determined before the damage's initial load, in order to avoid influence of this damage in the properties of the laminated composite.

Table 1. Mechanical properties - **NLB-T** - Uniaxial tensile.

MECHANICAL PROPERTY	MEAN VALUE	DISPERSION (%)
Tensile Strength (MPa)	19.60	5.91
Longitudinal Elasticity Modulus (GPa)	1.69	15.94
Strain (%)	1.59	12.06

3.3 Uniaxial tensile in the saturated humid state **WLB-T**

Regarding the uniaxial test for the **WLB-T** specimen, figure 4 displays the Tensile x Deformation curves for the laminated composite.

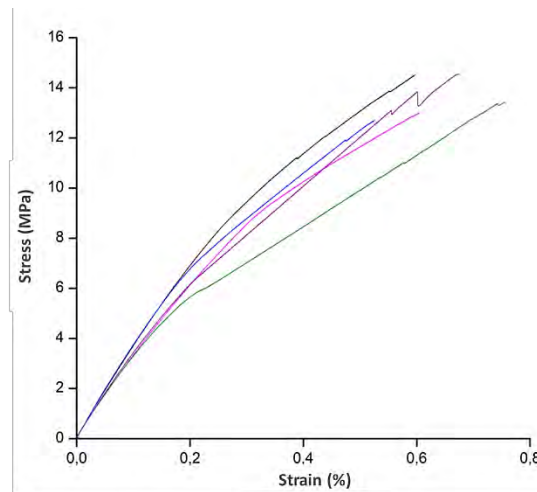


Figure 4. **WLB-T** stress x strain curves

The material displays a “fragile” behavior until fracture; such behavior was also observed in **NLB-T**. Thus, it can be stated that the moisture saturation state did not alter the material's tensile x deformation profile.

The mean values, as well as its respective dispersions, obtained for the ultimate tensile strength, deformation and bursting and for the longitudinal elasticity modulus (measured in the load application direction) are shown in table 2. Once again, the dispersions also refer to the absolute difference in percentage between the maximum and minimum values obtained in each test, for each parameter analyzed.

Table 2. Mechanical properties - **WLB-T** – Uniaxial tensile.

MECHANICAL PROPERTY	MEAN VALUE	DISPERSION (%)
Tensile Strength (MPa)	13.63	6.28
Longitudinal Elasticity Modulus (GPa)	3.17	7.28
Strain (%)	0.62	12.44

3.4 Comparative study between **NLB-T** X **WLB-T**

The comparative study related to the mechanical properties of ultimate tensile strength and elasticity modulus, for the **NLB-T** and **WLB-T** specimen, is displayed in the curves in figure 5. The curves shows the mean value line obtained in the respective tests.

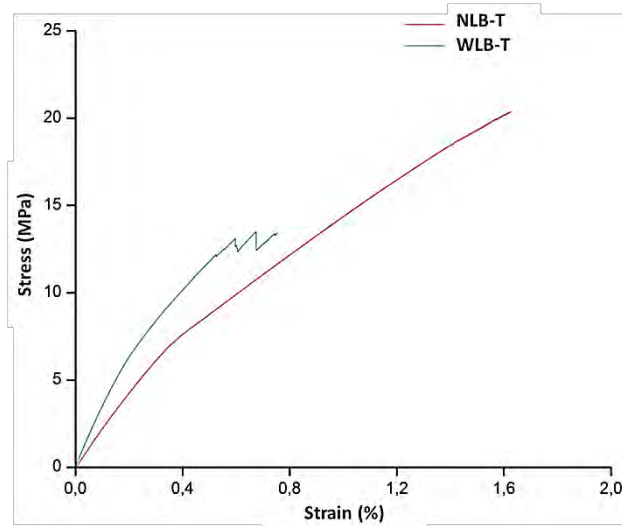


Figure 5. Comparative curves between the dry and humid saturated states for the biocomposite.

In figure 5, we can notice that **NLB-T** displays superiority in relation to the ultimate tensile strength and deformation and bursting when compared to **WLB-T**. Whereas, analyzing the elasticity modulus, it is noticed that **WLB-T** was fairly superior (noticeable by the more accentuated inclination). Analyzing the data in tables 1 and 2, we can quantify that:

- NLB-T** displayed a superiority of 30.45% in relation to **WLB-T** in Tensile Strength;
- WLB-T** displayed a superiority of 46.69% in Elasticity Modulus;

3.5 Three-point flexural test in natural state (NLB-F)

Regarding the three-point flexural test for the **NLB-F** specimen, figure 6 displays the stress x deflection curves for this laminated composite. The inflections shown in the curves characterize an initial fracture of the tensioned woven jute layer, characterized mainly by cracking in the matrix.

The mean values, as well as its respective dispersions, obtained for flexural strength, maximum deflection and flexural elasticity modulus are displayed in table 3.

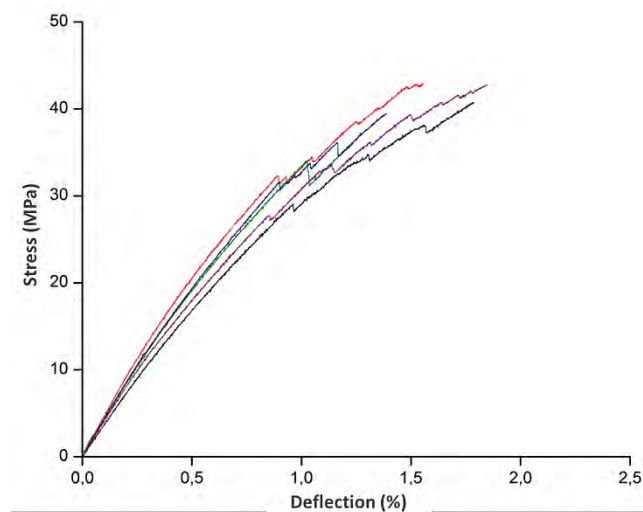


Figure 6. **NLB-F** stress x deflection curves.

Table 3. Mechanical properties - **NLB-F** – three-point flexural test.

MECHANICAL PROPERTY	MEAN VALUE	DISPERSION (%)
Flexural Strength (MPa)	40.37	7.16
Flexural Elasticity Modulus (GPa)	3.71	8.51
Maximum Deflection (%)	1.56	16.67

3.6 Three-point flexural in saturated humid state (WLB-F)

Regarding the three-point flexural test for the **WLB-F** specimen, figure 7 displays the Stress x Deflection curves for this laminated composite. The behavior observed is similar to the one obtained in the natural state, but it is important to highlight that the effect of fractures in the external layer of woven jute is more intense for the case of the laminated composite in saturated humid state.

The mean values, as well as its respective dispersions, obtained for resistance to the flexural strength, maximum deflection and for the flexural elasticity modulus are displayed in table 4.

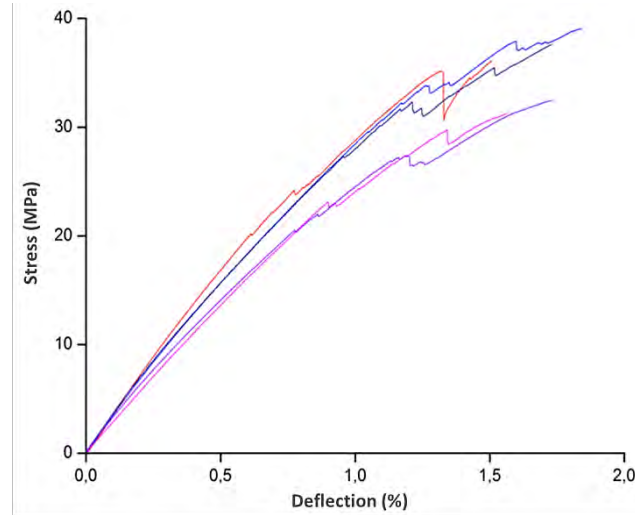


Figure 7. **WLB-F** stress x deflection curves.

Table 4. Mechanical properties - **WLB-F** – Three-point flexural test.

MECHANICAL PROPERTY	MEAN VALUE	DISPERSION (%)
Flexural Strength (MPa)	35.30	9.41
Flexural Elasticity Modulus (GPa)	2.92	9.96
Maximum Deflection (%)	1.67	8.30

3.7 Comparative study between NLB-F X WLB-F

The comparative study that related the mechanical properties of flexural strength and elasticity modulus, for the **NLB-F** and **WLB-F** specimen is displayed in curves in Figure 8. The curves show the mean value line obtained in the respective tests.

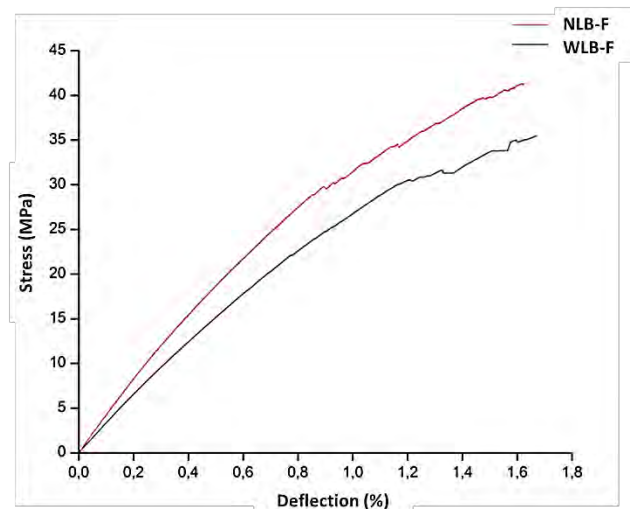


Figure 8. Comparative curves of flexural strength in dry and saturated humid states for the biocomposite.

In Figure 8, we noticed that the **NLB-F** displayed superiority in flexural resistance and elasticity modulus (noticeable by the curve's more accentuated inclination). There was not a significant difference in deflection.

Analyzing the data in Tables 3 and 4, we noticed the negative influence of the effect of moisture absorption in the properties studied. That is, leading to superiority in the **NLB-F** in relation to the **WLB-F** in the following percentage: 12.55% in ultimate flexural strength and 21.29% in flexural elasticity modulus.

4. CONCLUSIONS

In this study, some conclusive points may be highlighted:

Regarding the moisture absorption, we can affirm that the biocomposite submerged in seawater (**SLB**), absorbed less than the biocomposite submerged in distilled water (**WLB**), due to the accumulation of NaCl ions in the **SLB**'s surface, which increases with immersion time and prevents water diffusion in the specimen.

The biocomposite submerged in distilled water (**WLB**) had a 16.91% superior absorption in relation to the **SLB**.

In the uniaxial tensile test, the behavior of the biocomposite in all conditions studied was displayed as "fragile" until fracture. That is, the surface runoff phenomenon, characteristic of materials with ductile behavior, is not displayed.

Regarding the property of ultimate tensile strength, the biocomposite in dry state (natural **NLB-T**) displayed superiority when compared to the biocomposite in humid saturated state (**WLB-T**). For longitudinal elasticity modulus, the biocomposite in the saturated humid state (**WLB-T**) was superior. Quantitatively, moisture absorption caused a loss in resistance to the ultimate tensile strength of approximately 30.45%. For the longitudinal elasticity modulus, there was the gain of 46.69%.

Regarding the property of flexural resistance and flexural elasticity modulus, the biocomposite in dry state (natural **NLB-F**) displayed superiority in relation to the biocomposite in saturated humid state (**WLB-F**). Moisture absorption caused a loss of approximately 12.55% in resistance and 21.29% in elasticity modulus.

The justification for the losses displayed in flexural strength is due, partly, to the characteristic of each test. The necessary strain for the tensile test requires a lot from the specimen as a whole, overall the quality of its interface. In the case of flexural, the strain happens in every layer and, in this case, the most external layer displays special protection factors for humid environments, avoiding that the mechanical performance be harmed by the natural absorption characteristic of the structure's internal reinforcements.

However, the moderate losses in flexural strength are aspects that have already been reported in other paper with biocomposites. Le Duigou *et al.* (2009) had already observed a similar result in seawater immersion.

5. REFERENCES

- Aquino, E. M. F. "Estudo da umidade na resistência e fratura de compósitos tubulares". IV Congresso de engenharia mecânica N/NE. Recife, Brazil, 1996.
- ASTM D 3039-08. "Standard Test Methods for Tensile Properties of Polymer Matrix Composites", 2008.
- ASTM D 3878-98, "Standard Terminology for Composite Materials", Philadelphia.
- ASTM D5229/D 229M – 04, "Standard Test Methods for Moisture Absorption Properties and Equilibrium Conditioning of Polymer Matrix Composite Materials", 2004
- ASTM D570-98. "Standard test method for water absorption of plastics", Reapproved 2010.
- ASTM D 790 – 10. "Standard test method for flexural properties of unreinforced and reinforced and electrical insulating", 2010.
- Dantas, C. M. "Influência da quantidade de absorção de água nas propriedades mecânicas de compósitos poliméricos de manta de fibra de vidro e tecido juta" 2011. 96f. Master Thesis in Mechanical Engineering - Post-Graduate Programme in Mechanical Engineering, Federal University of Rio Grande do Norte, Rio Grande do Norte, Brazil, 2011.
- Le Duigou, A.; Davies, P. e Baley, C.: "Seawater ageing of flax/poly(lactic acid) biocomposites". Polymer Degradation and Stability, Vol. 94, 2009.
- Oliveira, J. F. S. "Estudos da configuração e condições ambientais adversas em compósitos poliméricos híbridos" 2007. 113f. Master Thesis in Mechanical Engineering - Post-Graduate Programme in Mechanical Engineering, Federal University of Rio Grande do Norte, Rio Grande do Norte, Brazil, 2007.
- Rodrigues, L. P. S.; Silva, R. V. & Aquino, E. M. F.: "Effect of accelerated environmental aging on mechanical behavior of curaua/glass hybrid composite". Journal of Composite Materials, Vol. 46. n. 17, 2012.
- Silva, R. V., Aquino, E.M.F., Rodrigues, L.P.S., Barros, A.R.F., "Desenvolvimento de um compósito laminado híbrido com fibras natural e sintética". vol.13 no.1 Rio de Janeiro Jan./Mar. 2008. *On-line* version ISSN 1517-7076. Available in: <http://www.scielo.br/scielo>
- Silva, L. C. F. "Utilização de Resíduo Lignocelulósico na obtenção de Chapa de Madeira Aglomerada Homogênea e Compósito Reforçado com Fibra de Vidro-E" 2008. 183f. Master Thesis in Mechanical Engineering - Post-Graduate Programme in Mechanical Engineering, Federal University of Rio Grande do Norte, Rio Grande do Norte, Brazil, 2008