Humidity Diffusion on Jute/Polyester Laminated Composites and Its Effects on Mechanical Properties

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Abstract: Jute is a natural fiber widely used as reinforcement in composites due to its high tensile strength and stiffness, but they can easily absorb water and have their physical properties compromised. The water absorption properties of jute/polyester composites are evaluated according to ASTM D 570 and the effect of humidity in the composite mechanical behavior is also analyzed. The composite showed a pseudo-Fickian behavior and gained 13.37% in weight after the test. It also lost tensile strength and elasticity modulus, and increased its specific deformation. Scanning electron microscope images showed that wet specimens were more subject to cracks, voids and fiber pullout than dry specimens. Failures produced by water diffusion in composite and polymer plasticization, added to breakdown in the fibers’ cellulosic structures, justify the change in mechanical properties due to water absorption.

Key words: Natural fiber reinforced composites (NFRCs), jute fiber, water absorption, polyester composites, diffusion in composites.

1. Introduction

The growing concern with negative environmental impacts created by disposal of non-biodegradable materials leads the scientific community to research materials with more environmental affinity. Vegetal fibers are alternatives with low environmental impact normally used in manufacture of clothing, fabrics, and crafts and as reinforcement for natural fiber reinforced composites (NFRCs) [1].

NFRCs form a class of materials that have low environmental impact, as they are renewable, biodegradable with low energy consumption for their production. They are also low-cost materials with desirable properties as low density and high specific strength, which contributes to increasing the mechanical properties and reducing the composite weight [2].

Jute is a plant belonging to genus Corchorus, whose fiber is mostly used to make ropes and fabrics. Jute fiber has high tensile strength, high stiffness and low thermal conductivity. Its physical and mechanical properties are shown in Table 1.

A limiting factor for natural fibers application as reinforcement in polymers is their intrinsic water absorption property, due to the capacity of free hydroxyls (-OH) present in cellulosic groups to form hydrogen bonds with water. This factor results in loss of fiber/matrix adhesion interface and formation of cracks in the matrix. Therefore, water absorption directly influences the mechanical performance of composites [3]. Other factors affect absorption of water by composites, such as fiber fraction volume, voids content, matrix viscosity and water temperature [4].

Polymeric matrices also suffer from the effects of water absorption. Unsaturated polyester resins are modified and degraded when in prolonged contact with atmospheric moisture or water. The loss of
adhesion generates cracks in the polymer, which creates spaces for entry of water and diffusion of water molecules in the spaces in polymeric chains. This leads to local deformation and rupture of the chain [5]. The polymeric matrix shapes the composite material, protects the reinforcement and transfers the mechanical loads to the fibers, distributing the stress among them [6]. Therefore, contact with water reduces the useful life of the composite, limiting its application.

This work aims to study the effects of water absorption in laminated composites made with polyester matrix reinforced by bidirectional jute fabrics molded by hot compression. The composite laminate specimens were immersed in water to analyze their diffusion properties and their mechanical behavior after being saturated with water. Their mechanical properties were assessed by tensile, flexural and interlaminar shear tests; finally, they were subject to microstructure analysis by scanning electron microscopy (SEM).

2. Experimental Setup

2.1 Production of Laminate Composites

The object of this study is a laminate composite made of two layers of bidirectional jute fabric with a weight of 118 g/m², shown in Fig. 1a, in an orthophthalic unsaturated polyester resin matrix, cured with methyl ethyl ketone peroxide (MEKP) in the proportion of 1.5% of resin weight. All materials were purchased commercially. The jute/polyester laminate, shown in Fig. 1b, was manufactured by hot compression, by application of 80 °C temperature and 7.8 MPa pressure for 1 h, achieving a thickness of 1.34 mm.

2.2 Water Absorption Test

The absorption test was performed following ASTM D 570, Standard Test Method for Water Absorption of Plastics, to determine the relative rate of absorption of water by composite after immersion. The composite plates were cut down to 76 × 25 mm specimens with an average thickness of 1.34 mm and dried in a laboratory oven at 110 °C for 24 h.

The medium used in this procedure is distilled water that filled a container large enough to completely submerge the absorption test specimens. The test measurements were taken in two-phases: the two-immersion phase, where the specimens are weighted every 2 h, and the twenty-four hour immersion phase, where the specimens were weighted every 24 h. The first phase was necessary because the thin specimens can show a significant weight increase after only two hours.

In both phases, the water was maintained at 23 ±1 °C and it was dried out of the specimens before weighting with dried cloths. This guarantees that only the water present within the specimens counts. The specimens were weighted with a 0.001 g precision.

To complete the absorption analysis, ASTM D 5992, Standard Test Method for Moisture Absorption Properties and Equilibrium Conditioning of Polymer Matrix Composite Materials, was used to calculate the composite diffusivity coefficient $D$ and assess whether the diffusion followed Fick’s law or not, by

<table>
<thead>
<tr>
<th>Density (g·m⁻³)</th>
<th>Elongation (%)</th>
<th>Tensile strength (MPa)</th>
<th>Tensile modulus (GPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.3-1.5</td>
<td>1.5-1.8</td>
<td>393-800</td>
<td>10-55</td>
</tr>
</tbody>
</table>

(a) (b)

Fig. 1 Composite fabrication process: (a) bidirectional jute fabric; (b) jute/polyester laminate composite.
calculating the parameter $n$. According to this standard, the diffusion coefficient is calculated using Eq. (1):

$$D = \pi \left( \frac{h}{4M_m} \right)^2 \left( \frac{M_2 - M_1}{\sqrt{t_2} - \sqrt{t_1}} \right)^2$$  \hspace{1cm} (1)

where:

- $h$ = average specimen thickness (mm);
- $M_m$ = effective moisture equilibrium content (%);
- $M_1$ and $M_2$ = moisture content at points 1 and 2 (%);
- $t_1$ and $t_2$ = time at points 1 and 2 (s).

It is important to register that point 1 and 2 must be chosen in the linear part of the moisture content vs. time curve. All moisture content values $M$ shown previously are given by Eq. (2):

$$M = \left( \frac{W_i - W_o}{W_o} \right) \times 100\%$$  \hspace{1cm} (2)

where:

- $W_i$ = specimen mass at the point measured (g);
- $W_o$ = initial oven-dry specimens mass (g).

The diffusion behavior is given by the Fickian parameter $n$, calculated using Eq. (3):

$$\log \left( \frac{M_t}{M_s} \right) = \log \left( \frac{M_2 - M_1}{\sqrt{t_2} - \sqrt{t_1}} \right) + n \log(t)$$  \hspace{1cm} (3)

where:

- $M_t$ = moisture content at the chosen point (%);
- $M_s$ = moisture content at saturation (%);
- $t$ = time at the chosen point (s).

The diffusion is pseudo-Fickian when $n < 0.5$, Fickian when $n = 0.5$, anomalous when $0.5 < n < 1$, and Case II diffusion when $n > 1$.

### 2.3 Mechanical Tests

Ten specimens were manufactured for each mechanical test, totaling thirty specimens. The mechanical behavior of the composites was assessed under two conditions: the dry state and the wet state, after distilled water absorption. The mechanical tests performed were uniaxial tensile, three point bending and interlaminar shear strength (ILSS) tests, according to standards ASTM D3039-17, ASTM D790-17 and ASTM D2344-16, respectively. The specimen dimensions were $250 \times 25$ mm for tensile test; $100 \times 13$ mm for three points bending test; and $16 \times 6$ mm for interlaminar shear test.

### 2.4 Fracture Analysis

The tested specimens were subjected to SEM of the fracture site, to evaluate the failure mechanisms of composites both in the dry and wet states. The samples were metallized with gold.

### 3. Experimental Results

#### 3.1 Water Absorption Test

The result of water absorption test is shown in Fig. 2. The parameters calculated are shown in Table 2.

The Fickian parameter $n$ indicates that the diffusion follows a pseudo-Fickian behavior. Fick’s law of diffusion predicts that the amount of absorbed water increases linearly with the square root of time and eventually stabilizes, reaching equilibrium [7]. Fick’s law is commonly used to predict the evolution of water absorption over time due to its simplicity and ability to describe the diffusion of water molecules in the composite [8].

It is observed that the jute/polyester laminate suffered an average weight gain of 13.36% after nine days. In the first 4 h, the composite mass increased linearly at a rate of 0.0548 g/h. From the 4th day (96 h) on, the curve slope is close to zero. From this point on, the water absorption rate dropped to about $4.45 \times 10^{-5}$ g/h, indicating that the amount of absorbed water was close to equilibrium.

#### 3.2 Mechanical Tests

The results of the tensile tests are shown in Fig. 3, in the dry (Fig. 3a) and wet (Fig. 3b) states. The jute/polyester composite in the dry state reached an average maximum tensile strength of 4.873 MPa and an average elastic modulus of 0.215 GPa. After water absorption, the tensile strength and elastic modulus dropped to 4.59 MPa and 0.123 GPa, respectively. Regarding specific deformation, there was an increase from 3.477% to 3.973%.
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Fig. 2 Evolution of weight gain during absorption test.

Table 2 Water diffusion parameters for jute/polyester laminated composite.

<table>
<thead>
<tr>
<th>Diffusion coefficient $D$ (mm$^2$/s)</th>
<th>Fickian parameter $n$ (non-dimensional)</th>
<th>Moisture contents $M_m, M_\infty$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$1.1689 \times 10^{-5}$</td>
<td>0.229</td>
<td>13.36</td>
</tr>
</tbody>
</table>

Fig. 3 Stress vs. strain curve resulted from tensile test for dry jute/polyester composite and jute composite after water absorption.

Fig. 4 Stress vs. strain curve resulted from flexural test for dry jute/polyester composite and jute composite after water absorption.

The results of the mechanical tests have shown that water absorption has led to loss in mechanical strength and elasticity modulus. Deformation, on the other hand, increased after prolonged contact with water. This is a consequence of diffusion of water molecules due to capillary action in the spaces in the polymer chains, the fibers and at the fiber/matrix interface. The natural

Fig. 4 shows the curve obtained from the flexural test. The maximum flexural strength in dry state was 11.350 MPa, the flexural modulus was 0.250 GPa and the specific strain was 0.454%. After absorption test, flexural strength, flexural modulus and deflection were 8.505 MPa, 0.133 GPa and 0.068%, respectively.

The results of the interlaminar shear test are shown in Fig. 5. An average maximum shear strength of 6.57 MPa and a maximum displacement of 0.58 mm were found for the dry state. The absorption of water led to a decrease in shear strength, which dropped to an average of 5.47 MPa, and to an increase in maximum displacement, which reached 0.87 mm.
fibers swell with absorption of water which exerts a stress on the matrix, eventually breaking the polymer. This mechanism weakens the fiber’s adhesion in the matrix, decreasing the composite’s mechanical strength [4].

Diffusion of water molecules among polymer macromolecules leads to hydrolysis and plasticization, which decreases the polymer’s stiffness at room temperature. These two mechanisms affect the polymer chains and their connections, decreasing the elasticity modules for tensile and bending loads. Plasticization makes the polymer softer and, as consequence, its deformation is increased, as demonstrated in the tensile and shear tests [9].

### 3.2 Morphological Properties

Fig. 6 shows the micrographs obtained by SEM of the fracture region of dry state composites subjected to the bending test. The image shows only cohesive fractures in matrix and fiber, indicating a good adhesion between jute and polyester.

Fig. 7 shows the micrographs obtained in the fracture region of the wet state composites submitted to the flexural test. Matrix residues in the jute fiber indicate good adhesion between fiber and matrix. However, there is a reduction of adhesion strength in some regions, resulting in adhesive failures in the composite, such as the detachment of the fiber from the matrix.
The degradation of the fiber/matrix interface, observed in the fracture analysis by SEM, is one of the factors responsible for the loss of composite’s mechanical strength. This ductility gain occurs due to breakdown of the fibers’ cellulosic structures due to the water absorption. These structures are responsible for the natural fiber stiffness [10].

4. Conclusions

This work examined the effects of water absorption in jute/polyester composites. The water absorption test showed that water diffusion in the composite occurs in accordance with Fick’s law, with a marked increase in weight gain followed by an almost unchanging weight variation after 96 h. The mechanical tests showed a decrease in composite’s mechanical strength and elasticity modulus, as well as an increase in the material ductility due to effects of water diffusion through fiber and matrix, causing a reduction in adhesion between jute and polyester. The fracture analysis corroborated what was assessed in the mechanical tests, as it was possible to observe the appearance of adhesive fractures in the composite after water absorption.

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References


