

WATER SORPTION IN UNSATURATED POLYESTER COMPOSITES REINFORCED WITH JUTE AND JUTE/GLASS FIBER FABRICS: MODELLING AND SIMULATION

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Abstract. *Studies on composites reinforced with natural fibers show that the difficulties associated to the use of the vegetable fibers as reinforcement in polymer composites are frequently related to moisture absorption. The presence of moisture leads to imperfect impregnation and weak fiber-matrix interfacial adhesion, leading to the appearance of internal tensions, porosity, and premature failure of the system. This problem can be reduced by employing hybrid fiber weaved or knitted fabrics. In the present work compression molded unsaturated polyester composites reinforced with jute fabric and a hybrid jute-glass fabric were manufactured. The composites had the following weight compositions: a) 26% jute/74% unsaturated polyester and b) 26% jute/32% glass roving/42% unsaturated polyester with dimensions of 20.00x20.00x2.30 mm³ and 20.00x20.00x2.82 mm³, respectively. A three-dimensional transient diffusion mathematical model was developed to predict mass transfer during water absorption. The mathematical formulation was numerically solved using a finite-volume method and an implicit interpolation scheme. The results of moisture content distribution inside these composites are shown and analyzed. The knowledge of this moisture distribution allows verifying more favorable areas to presents delamination problems due to the weakness of the fiber-matrix interface and consequently reduction in the mechanical properties.*

Keywords: *hybrid fabric, composite, vegetable fiber, water sorption, finite*

1. Introduction

In recent years there has been a fast growth in using renewable vegetable fibers as reinforcements in composite materials. Reinforced plastics made with cellulosic materials, as fillers are low cost, light-weighted, have enhanced mechanical properties, and are free from health hazards.

Despite the attractiveness of vegetable fiber reinforced polymer matrix composites, they display lower mechanical properties and relatively poor moisture resistance compared to synthetic fiber reinforced composites.

Natural fiber reinforced polymer matrix composites are very sensitive to influences from environmental agents such as water (Thwe and Liao, 2002). The presence of moisture at the fiber-matrix interface induces formation of voids at the interface. The interfacial moisture also contributes to initiation of the process of debonding between and the cured resin (Mohanty and Misra, 1995). The weaknesses of vegetable fiber composites can be improved through the use of hybrid reinforcements, either in the laminate combinations or through the mixture of fibers (Cavalcanti, Carvalho and Lima, 2005). Hybridization of vegetable with stronger fibers such as glass fiber, can also improve the moisture resistant behavior of the composite. Using a hybrid composite that contains two or more types of different fibers, the advantages of one type of fiber could complement what are lacking in the other. As a consequence, a balance in performance and cost could be achieved through proper material design (Thwe and Liao, 2002).

Several works on the kinetics of absorption of water and/or moisture in composites systems presenting analytical and/or numerical solutions are reported in the literature. However, all these works consider the diffusion through the solid to be uni-dimensional (Apicella *et al.*, 1982; Ellis and Found, 1983; Han and Koutsky, 1983; Camino *et al.*, 1997; Marcovich *et al.*, 1999; Choi *et al.*, 2001). In some cases, in order to take in consideration three-dimensional problems, some researchers use analytical solutions applicable for long times; that greatly simplifies the problem (Chateauinois *et al.*, 1994; Pavan *et al.*, 2001; Bao and Yee, 2002; Srihari *et al.*, 2002;).

The aim of this work is to use a three-dimensional transient diffusion mathematical model to predict mass transfer during water absorption in a polymer composite using a mathematical formulation numerically solved by the finite-volume method and an implicit interpolation scheme. The composites under investigation were based on unsaturated polyester reinforced with jute and jute/glass fiber fabrics.

2. Experimental

Unsaturated polyester was supplied by Resana S.A. and cured with 1% MEK supplied by VI Fiberglass S.A. Jute (10/2) and glass fiber roving (111A-408) were supplied by Cia. Têxtil Castanhal S.A. / PA and Owens Corning do Brasil S.A., respectively.

Plain weave hybrid jute/glass fabrics having 41 jute strings and 42 glass roving per $10 \times 10 \text{cm}^2$ were hand weaved in a plain weave pattern. The resulting fabric had 44% (wt.) vegetable fibers (jute) in the warp and 56% (wt.) glass fibers in the weft directions.

Composites with varying fiber contents were compression molded. The fabrics were cut up according to the mold dimensions (170 x 140mm), pre-impregnated with the resin and placed in the mold. Excess resin was poured over the fabrics, the mold was closed under 8-ton force applied for 4h before demolding. Composite plates with to 2 layers of jute fabric (26%jute/74%polyester) (dimensions 140.00x170.00x2.30mm) and with 4 layers jute/glass hybrid (26% jute; 32% glass and 42% unsaturated polyester) (dimensions 140.00x170.00x2.82mm), were manufactured. Samples (20.00 x 20.00mm) were sawed-off from these plates and their edges sealed with resin prior to water absorption testing and dried to constant weight in an air-circulating oven operating at 105°C.

Water sorption of pre-dried composites samples was performed by immersion in water at room temperature and the water uptake was measured gravimetrically.

3. Mathematical modelling

□ Mass transfer model

To describe the mass transfer in the solid with parallelepiped shape (Figure 1) the following considerations are adopted, in the mathematical model:

- The solid is heterogeneous and anisotropic;
- The effect of capillarity is negligible;
- symmetry exists in the center of the solid;
- the boundary condition is of equilibrium with the surrounding, in the surface of the solid;
- the density of the solid is considered constant;
- any mass generation inside the solid happens;
- the solid is totally considered dry in beginning of the process.

The Figure 1 illustrates a solid parallelepiped of dimensions $2R_1 \times 2R_2 \times 2R_3$. For this case, the general differential equation that describes the diffusion phenomenon it is the way:

$$\frac{\partial M}{\partial t} = \nabla \cdot (D \nabla M) \quad (1)$$

where D is the diffusion coefficient and M is the moisture content.

Due to the symmetry of the problem we consider 1/8 of the volume of the solid. The initial, symmetry and boundary conditions are as follows:

□ Initial condition:

$$M(x, y, z, t=0) = M_o, \quad 0 < x < R_1, \quad 0 < y < R_2, \quad 0 < z < R_3 \quad (2)$$

□ Symmetry conditions:

$$\frac{\partial M(x=0, y, z, t)}{\partial x} = \frac{\partial M(x, y=0, z, t)}{\partial y} = \frac{\partial M(x, y, z=0, t)}{\partial z} = 0, t > 0 \quad (3)$$

□ Equilibrium boundary conditions:

$$M(x = R_1, y, z, t) = M(x, y = R_2, z, t) = M(x, y, z = R_3, t) = M_e, t > 0 \quad (4)$$

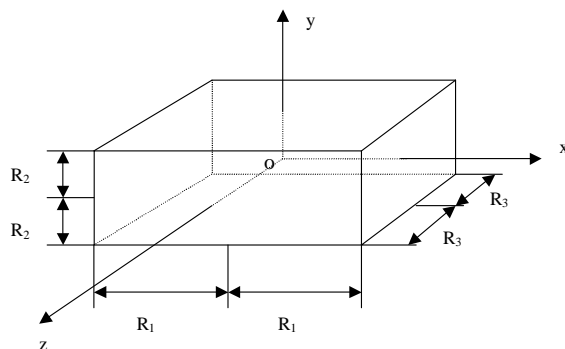


Figure 1. Geometrical configuration of the physical problem

The average moisture content was computed as:

$$\bar{M} = \frac{1}{V} \int_V M dV \tag{5}$$

In this work, the diffusion coefficient was considered dependent of the moisture content of the solid in agreement with the following equation.

$$D = D_o \text{Exp}(a\bar{M}) \tag{6}$$

where D_o and a are constant and \bar{M} is the average moisture content.

3.2 Numerical Method

In this work a finite-volume method was used to discretize the governing equations. Figure 2 represents the differential volume of the physical domain (Figure 1), where the nodal points (W, E, N, S, F and T), the dimensions and length of the control volume are presented.

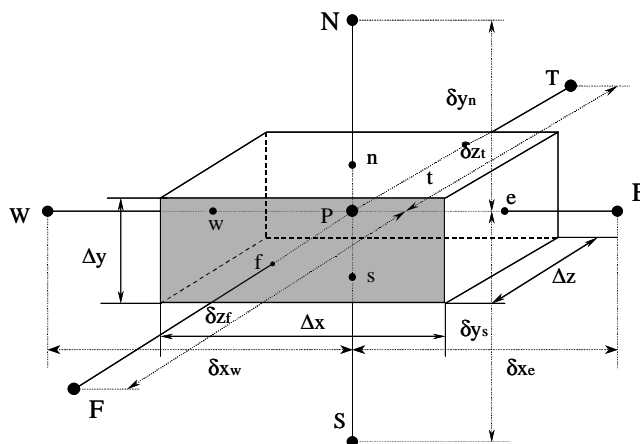


Figure 2. Control volume used in this work

Assuming fully implicit formulation, where all terms are estimated in $t+\Delta t$, equation (1) was integrated in the control volume of the Figure 2, that correspond to the internal points of the domain, and also in time. As a results equation (1) was obtained in the discretized by the finite-volume method utilizing the practice B (nodal points in the center of control-volume) (Patankar, 1980; Maliska, 2004) and it can be written in the linear form as:

$$A_P M_P^* = A_E M_E^* + A_W M_W^* + A_N M_N^* + A_S M_S^* + A_T M_T^* + A_F M_F^* + B \tag{7}$$

where:

$$\begin{aligned}
 A_E &= \frac{D_e \Delta y \Delta z}{\delta x_e} & A_W &= \frac{D_w \Delta y \Delta z}{\delta x_w} & A_N &= \frac{D_n \Delta z \Delta x}{\delta y_n} & A_S &= \frac{D_s \Delta z \Delta x}{\delta y_s} \\
 A_T &= \frac{D_t \Delta x \Delta y}{\delta z_t} & A_F &= \frac{D_f \Delta x \Delta y}{\delta z_f} & A_P^o &= \frac{\Delta x \Delta y \Delta z}{\Delta t} & B &= A_P^o M_p^o \\
 A_P &= A_E + A_W + A_N + A_S + A_T + A_F + A_P^o & M^* &= \frac{M - M_e}{M_o - M_e}
 \end{aligned}$$

where D in the interface of the control-volume was calculated according to literature (Patankar, 1980).

The set equations are solved interactively using the Gauss-Seidel method. The following convergence criterion was used:

$$|M^{*n-1} - M^{*n}| \leq 10^{-8} \quad (8)$$

where n represents the n-th iteration in each time.

The diffusion and mass transfer coefficients were found by varying the D to minimize the sum of squared deviations between the actual and predicted data. The relative deviation between experimental and calculated values (relative residual, ERMQ) and the variance (S^2) are defined as follows (Figliola and Beasley, 1995):

$$ERMQ = \sum_{i=1}^n (\bar{M}_{i,Num}^* - \bar{M}_{i,Exp}^*)^2 \quad (9a)$$

$$S^2 = \frac{E}{(m-1)} \quad (9b)$$

where m is the number of experimental points. The smallest values of ERMQ and S^2 were used as criteria to obtain the best value of the diffusion and mass transfer coefficients.

4. Results and discussion

The numerical results of the average moisture content of the composites under investigation were compared with the experimental data. In order to obtain the numerical results, a computational code using a grid of 20 x 20 x 20 point and $\Delta t=1s$ was used. These parameters were determined after mesh and time refining study described by Nascimento (2002) was carried through. The numerical results obtained from a given mathematical model are strongly dependent to boundary conditions, thermo-physical properties and geometry.

A comparison between the experimental and numerically predicted average moisture content for the composites investigated, shown in Figure 3, indicates that the model employed here is appropriate. Modeling led to errors of 0.0234468 kg/kg² with 0.000239 kg/kg² variance at the end of the sorption process (600h, i.e., 25 days) for the polyester/jute (74/26 w/w) composite and of 0.2151 kg/kg² with 0.00129 kg/kg² variance at the end of the process (1248h, i.e., 52 days) for the hybrid polyester/jute-glass (42/26/32 w/w) composite. Figure 3 also evinces that an equivalent amount of water was absorbed by composites with similar jute content which is taken an indication that water sorption in these composites is dominated by the vegetable fiber (jute).

The moisture content distribution of the polyester/jute and polyester/ jute-glass composites in the plane $x^* = x/R=0.5$ and $z^* = z/R = 0.5$ for the material 26% jute/74% unsaturated polyester (21 Jute) and $x^* = x/R=0.5$ for the material 26% jute/32% glass roving/42% unsaturated polyester (41 Jute/Glass) at times 3, 20, 44, 133 and 208 h is presented in Figures 4 (a and b) and Figure 5, respectively. The existence of high moisture gradients in the region next to the vertex of the solid is illustrated. Figures 4a and 4b and Figure 5 show the moisture content distribution to be uniform and to increase with immersion time. When compared the Figure 4a with a Figure 4b is observed that the moisture content distribution is similar in the two sides of the solid. In the area near the points ($x=R_1, y=R_2, z=R_3$) the solid humidifies faster as this area is in direct contact with water.

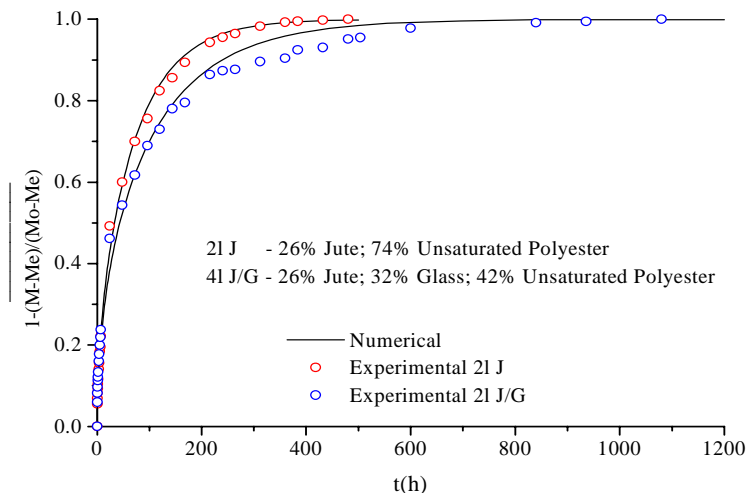


Figure 3. Comparison between predicted and experimental dimensionless mean moisture content during the water sorption in jute and jute/glass reinforced unsaturated polyester composites

A comparison between Figures 4a and 5 indicates that, despite their differences in thickness, the humidifying process of the jute reinforced composite was faster than that of the hybrid fabric reinforced composite. This behavior is attributed to the increase in solid area/volume ratio as well as to the higher water affinity of the cellulose material. This indicates that hybridization of vegetable fibers with synthetic ones such as, for instance, glass fibers, is an efficient way to increase the water uptake resistance of polymer composites.

It is important to know the profiles and the moisture content inside the material in order to predict which areas are more sensitive to mechanical stresses, which may cause cracks and deformations, and consequently decrease the quality of the product (Nascimento, 2002).

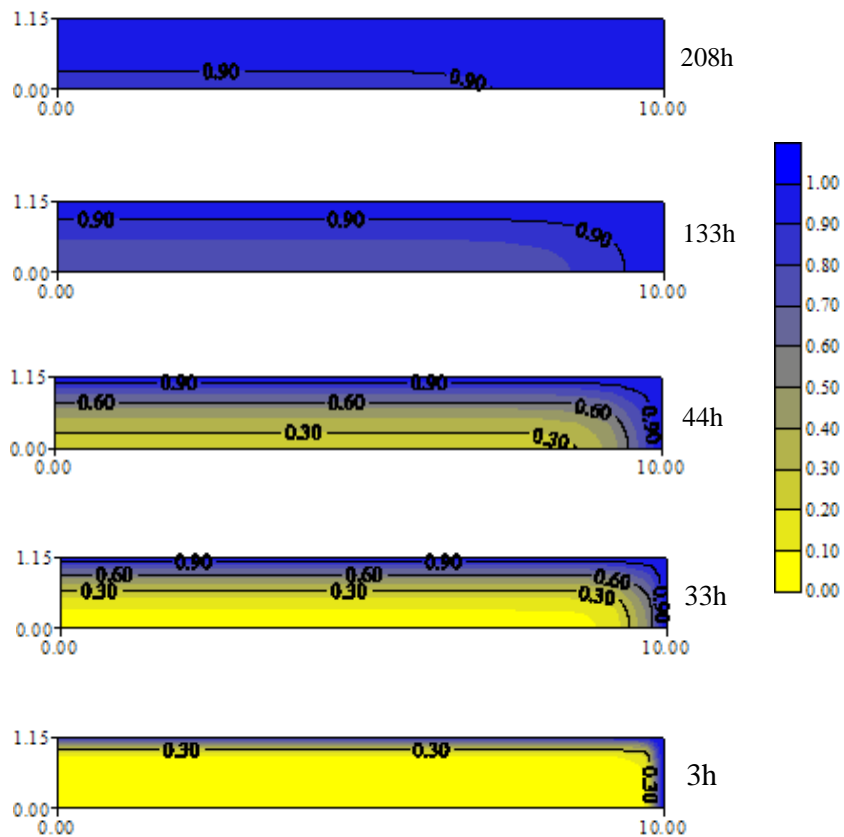


Figure 4a. Moisture content profiles inside the material 26% jute/74% unsaturated polyester in the plane $x^* = x/R = 0.5$ at several times.

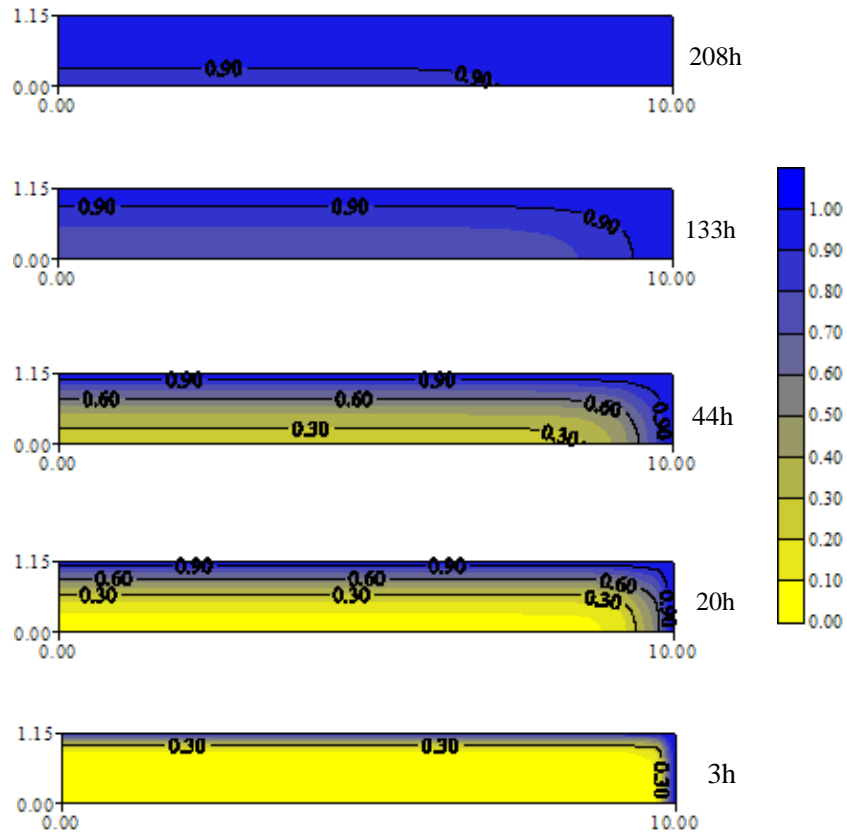


Figure 4b. Moisture content profiles inside the material 26% jute/74% unsaturated polyester in the plane $z^* = z/R = 0.5$ at several times.

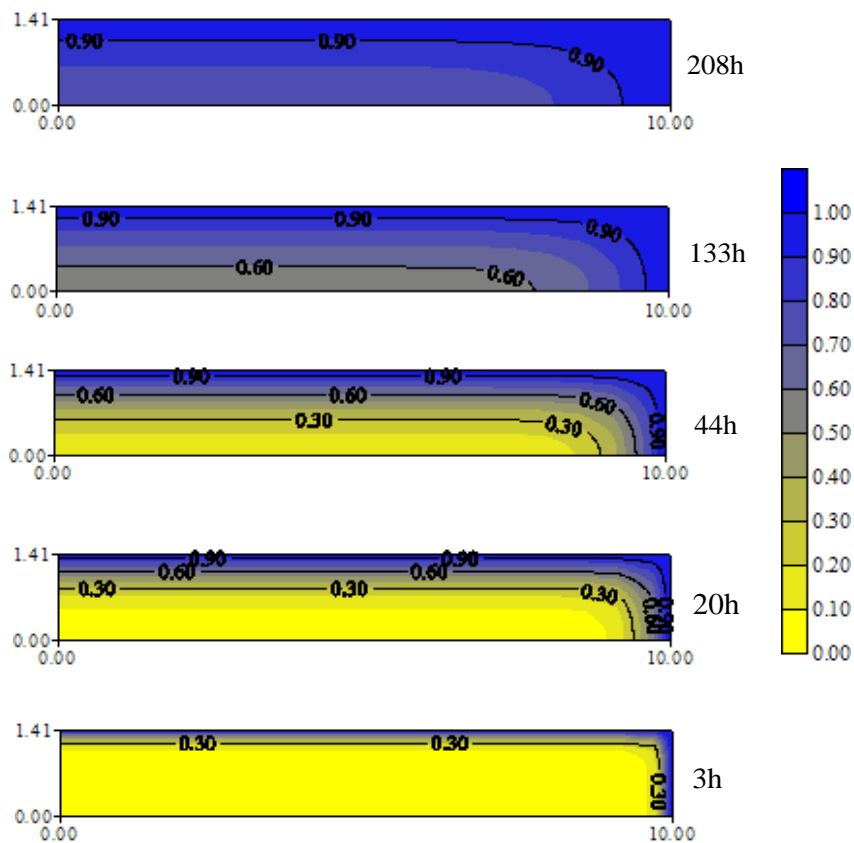


Figure 5. Moisture content profiles inside the material 26% jute/32% glass roving/42% unsaturated polyester in the plane $x^* = x/R = 0.5$ at several times.

5. Conclusions

The mathematical model used adequately describes the process of water diffusion inside the composites. High water diffusion was obtained for the first 50 h of sorption, and the rate of sorption decreased at longer water immersion times. The regions in the neighborhood of the vertices of the solid present the largest rates of mass transfer.

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