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STUDY OF SATURATED AND UNSATURATED PERMEABILITY IN NATURAL FIBER FABRICS

Exequiel S. Rodríguez[1], Gastón Francucci[1] and Analía Vázquez[1, 2]

[1] *Research Institute of Material Science and Technology, INTEMA, National University of Mar del Plata, J. B. Justo 4302, B7608FDQl, Mar del Plata, Argentina*
[2] *Corresponding author's Email: anvazque@fi.mdp.edu.ar*

SUMMARY: Natural fibers present some advantages which made them useful in composite parts: reduced dependence on non-renewable resources, less wear on tooling, ease of handling, lower health risk for the producer workers than glass fibers, better thermal and sound insulation properties, low density, cost about one third of glass fibers. RTM process can be use to manufacture natural fiber composites, but in order to perform an optimum process design and maximize the part quality, it is very important to have knowledge of the flow pattern during the injection process. The key parameter that governs the flow in the fiber bed is the permeability, together with the fluid viscosity. In this work, the saturated and unsaturated permeability of woven-bidirectional jute fabrics are studied for different porosities of the fibrous reinforcement. Also the fluid absorption by the natural fibers and its effect on the permeability is analyzed. It was found, as expected, that permeability increases as the porosity does, and that saturated permeability was greater than unsaturated permeability. This difference tends to vanish when porosity reaches values of about 75% or higher. It was found also, that jute fibers absorb large amounts of fluid. The fluid absorption affects flow behavior and reduces both saturated and unsaturated permeability values.

KEYWORDS: **n**atural fibers, fibrous reinforcements, permeability measurement, fluid absorption, Resin Transfer Molding (RTM)

INTRODUCTION

In the last years, governmental regulations about carbon dioxide emissions and recyclability of the materials have produced an increase in the use of natural fiber composites both in the automotive and construction industry. Many studies of sustainability and life cycle assessments have demonstrate the environmental advantages of these materials [1, 5]. But one of the keys of its success is the possibility of using the well-studied glass fiber composites processing techniques, like RTM, VARTM or SCRIMP. Therefore, is crucial to understand what happen with the main processing variables when replacing glass by natural fibers, which have different structure, different fabric architecture and different chemical interactions with the resins. One of those variables is the fabrics permeability, which is the key parameter that governs the flow in the fiber bed, together with the fluid viscosity. Fabrics permeability is especially important in low pressure injection techniques like VARTM or vacuum infusion where void formation and injection time can be increased dramatically when the permeability decreases.

The processing of natural fiber composites by Liquid Composite Molding (LCM) techniques has been studied by several authors in the last years [6, 10] (Sebè et.al., 2000; Oksman et.al., 2001; Rouison et al., 2004; Richardson and Zhang, 2000; Dweib et.al., 2006). Most of the works have focused on the determination of the physical and mechanical properties of the composites obtained but little research have been conducted studding the injection process itself and the effect of using natural fibers on the processing variables. Richardson and Zhang [10] studied the mold filling process in RTM with non-woven hemp and phenolic resin. Fiber washing and edge flow were the main problems founded while injecting with these materials. O'Donnell et al. [11] developed composites panels of soy oil-based resin and different natural fibers (flax, hemp and cellulose mats and recycled paper). They determined the permeability of the reinforcements and, except in the case of recycled paper, the values obtained were high enough for infusion by Vacuum assisted RTM (VARTM). Umer et al. [12] characterized the permeability and compaction behavior of wood fiber mats obtained by different manufacturing techniques. The permeability values that they obtained depended on the test fluid used for the experiments. When using glucose syrup the permeability was lower than the obtained with mineral oil. This behavior is due to the swelling of the fibers exposed to the water-base solution that reduces the size of the open flow paths.

Even though some permeability values have been reported for natural reinforcements, a detailed insight on their flow behavior is still required. What is more, the results obtained for one kind of fiber and fabric architecture are difficult to compare with the obtained for other fibers. So, what is important is to identify the main mechanisms present in natural fibers impregnation.

One key aspect that has been studied by several authors in glass fibers is the difference between saturated and unsaturated permeability. The investigations made on this topic are not consistent, and a wide variety of results have been reported. Kim et al. [13] and Diallo et al. [14] found that the saturated permeability was always lower than unsaturated permeability. On the other hand, Lundstrom et al. [15] and Foley et al. [16] obtained opposite results, founding that the saturated permeability was always higher than the unsaturated permeability. Also, results have been reported showing that the saturated and unsaturated permeabilities were almost equal [17]. These discrepancies are usually attributed to inevitable process mechanisms that could modify the saturated and unsaturated permeability ratio, such as mold deflection, capillary effect, microscopic flow, fiber channeling, and air bubbles [18].

When a fluid travels through a dry reinforcement of natural fibers, different mechanisms occur. First of all, as well as in synthetic woven fibers preforms, a macroscopic flow through the inter-tow regions takes place, followed by a delayed microscopic flow through the intra-tow region. Pillai and Advani [19] have studied in detail the unsaturated flow in woven fibers preforms, taking into account the delayed impregnation of fiber tows through the use of a sink function in the equation of continuity for the macro flow. Microscopic flows can also occur through the micro pores generated during the stacking and compression of the layers of reinforcement. Instead of synthetic fibers, natural fibers absorb fluid, acting as a sink. The fluid absorption consumes fluid from the main stream as it travels through the reinforcement, increasing flow resistance during the unsaturated flow. Furthermore, saturation of natural fibers can cause swelling, thus reducing the porosity and increasing flow resistance during saturated flow.

The aim of this work is to achieve knowledge on mechanisms related to natural's fibers impregnation in order to understand the resin flow behavior through natural fiber preforms, and improve the quality and performance of natural fiber composite products. The relation between permeability and porosity of jute bidirectional fabrics was obtained in the case of the VARI (Vacuum Assisted Resin Infusion) process. Both saturated and unsaturated permeability were obtained from the infusion tests. The Carman – Cozeny model was used in order to get an analytical relation between permeability and porosity. Furthermore, a brief analysis on jute fiber water absorption and its effect on permeability values was performed. Jute fabrics were coated with PHB polymer in order to reduce the fiber fluid absorption. Permeability tests results obtained on jute and PHB treated jute fabrics were compared in order to study the fluid absorption effect on the permeability values.

EXPERIMENTAL DETAILS

Bidirectional jute fabrics (surface density = 0.03 gr/cm^3) have been used in this study. The fabrics were washed with a 2% V/V distillate water and detergent solution, to remove contaminants and normalize the fabric condition for all the injections. In order to study the fluid absorption effect on permeability measurements, two different treatments were performed to make fibers less permeable. The first treatment (treatment A - TA) consisted in wetting out the jute fabric with a Polyhydroxybutyrate (PHB)- chloroform solution (2%). PHB (a type of polyhydroxyalkanoate) is a biodegradable thermoplastic polymer with a high hydrophyle character. After wetting, the fabrics were air dried leading to surface density values of 0.033 gr/cm3. The second one (treatment B - TB) consisted in submerging the fabric in a recipient that contains the same solution, and leaving the fibers immersed until the solvent completely evaporates. In this case PHB content obtained was much higher than with TA, and surface density reached 0.046 gr/cm3. Also, bidirectional glass fabrics were used for comparative purposes (surface density = 0.02 gr/cm^3).

The fluid used in this study was a 22 % V/V water/glycerin solution, leading to viscosity values near 0.150 Pa.s. Red colorant was used to improve the flow front visibility. Unidirectional injection experiments were performed in a rectangular metallic mold (500 mm x 100 mm) with an acrylic lid. The thickness of the mold cavity used for each injection was set in order to obtain the desired values of porosity. The viscosity of the fluid used was measured in a cone and plate viscometer (Brookfield DV – II+). A vacuum pump was used to force the fluid flow though the mold cavity. The pressure gradient achieved was measured with a vacuometer located at the outlet line of the mold.

Humidity absorption test were performed in a humidity chamber, with controlled atmospheric humidity of 65.1 %. The samples were weighted using an analytical scale and the time of each weight measure was controlled with a chronometer. Samples of jute, J-TA, J-TB and bidirectional glass fabric were dried under vacuum at 90ºC for 24 hs, before performing the humidity absorption test. Images of the fibers were obtained by Scanning Electron Microscopy (SEM JEOL JSM-6460 LV) to analyze the PHB treatment effect on fiber morphology and structure. Sample preparation for this technique consisted in coating the fibers with a thin layer of gold.

DATA ANALYSIS

Unidirectional Flow Experiments

In this study, Darcy's Law for unidirectional flow was used to estimate the permeability. To validate the use of Darcy´s Law, it is assumed that the pore volume of the portion of the preform behind the fluid front is fully saturated with fluid, which allows using the quasi – steady – state assumption. Unsaturated permeability can be obtained using equation 1:

$$K_{unsat} = \frac{\Phi.m.\mu}{2\Delta P} \quad \textbf{(1)}$$

where K_{unsat} = unsaturated permeability (m^2), m = slope of the curve x^2 (square of the flow front position) as a function of time. The relation between the front flow position and the injection time was obtained from a camera mounted on top of the transparent flow cell, which was recording the front flow position. Saturated permeability was calculated by measuring the fluid volumetric flow rate, once the reinforcement was fully saturated. A standard caudal meter connected to the output line of the mold was used for this purpose, so a plot of volume in time could be obtained. This yields the saturated permeability as follows:

$$K_{sat} = \frac{Q.\mu.\Delta L}{(A.\Delta P)} \quad \textbf{(2)}$$

where K_{sat} = saturated permeability (m^2), Q = volumetric flow rate (m^3/s), ΔL = preform length (m), A = mold cavity transverse area (m^2). Finally, the Carman – Kozeny model was used to establish a relationship between permeability and porosity. This model has two adjusting parameters, n and C, in the form of Equation 3 below:

$$K = \frac{\phi^{n+1}}{C(1-\phi)^n} \quad \textbf{(3)}$$

Humidity Absorption Test

Humidity absorption tests gave the weight vs. time. In order to become independent of the weight of the samples tested, the relative humidity absorption was calculated by the following equation:

$$A_r(t) = \frac{[W(t) - W_0]}{W_0} \tag{4}$$

where $A_r(t)$ = relative humidity absorption as a function of time, $W(t)$ = sample weight as a function of time, W_0 = initial sample weight.

RESULTS AND DISCUSSION

The result of the humidity absorption tests is shown in Fig. 1. Glass fabric showed almost no humidity absorption. On the other hand, the relative humidity absorption values seen in jute fabrics reached the 25 %, and the saturation humidity value was reached very fast. As seen in the plot, PHB treatments were effective, reducing fiber water absorption. Treatment B has reduced the relative humidity absorption value to 5%, a result more effective than treatment A, which lead to values of 7,5 % of water absorption. This difference was expected, because in treatment B the resulting PHB coating is thicker and more uniform, due to the longer exposure time to the chloroform-PHB solution.

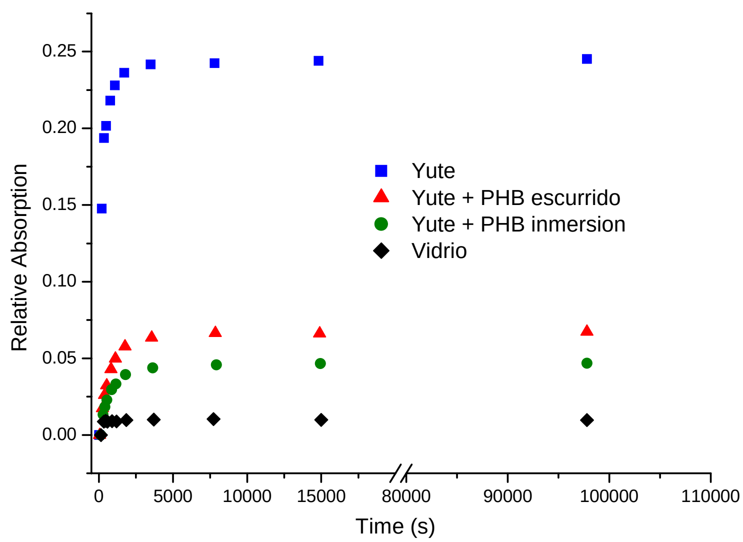

Fig. 1 Humidity absorption test results.

Scanning Electron Microscopy images of the fibers are shown in Fig. 2. It can be seen that PHB treatments reduce fabric tortuosity, by packing together the bundles of fibers. This should have an important effect over permeability values.

Results of the permeability tests are shown in Fig. 3. The error bars resulted from an uncertainty propagation analysis performed, which is not shown in this work. Table 1 shows the values of the parameters of the Carman – Kozeny equation, which fit the experimental data of K_{sat} and K_{unsat}. As expected, both saturated and unsaturated permeabilities increase as porosity does. Also, it was observed, accordingly with other authors, that permeability values are higher when the reinforcement is fully saturated than during the filling process. This observation can be attributed to the delayed impregnation of the dense fiber tows, with respect to the macroscopic flow front, because of the difference in local permeability values of the inter-tow and intra-tow regions. Fiber tows acts as a sink, removing fluid from the main stream as it travels along the mold cavity, leading to lower permeability values. In other words, flow resistance increase when the micro pores are being filled with fluid.

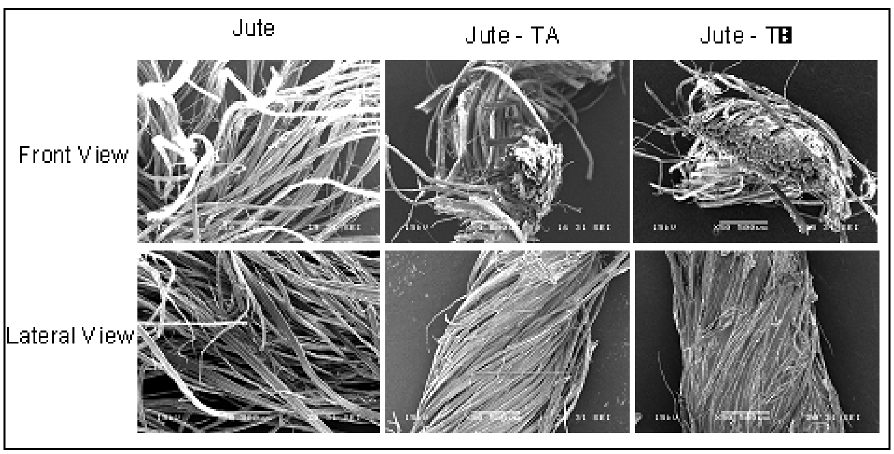

Fig. 2 SEM images of jute fibers with and without PHB treatment.

It was observed that the saturation gradient mentioned above exists in a narrow region next to the macroscopic flow front, so the quasi – steady – state assumption should hold valid. Another possible explanation to the difference observed between saturated and unsaturated permeability values, is because of reinforcement fluid absorption. Jute fabrics absorbs great amount of liquid, as is shown in the absorption test. Absorption acts as another sink component, removing more liquid from the main fluid stream and retarding the fluid flow, thus the flow resistance increase and permeability values drops. When mold filling is complete, and the reinforcement is fully impregnated and saturated with fluid (no more micro pores impregnation or fluid absorption takes place), its sink nature vanishes, and flow resistance decrease. Thus, saturated permeability values are higher than unsaturated ones.

An interesting observation is that at high porosity values the difference mentioned decrease and almost disappears, due to the low fiber volume fraction. As discussed before, in natural fibers, unsaturated permeability is affected by fluid absorption and saturated permeability is affected by fiber swells (a consequence of fluid absorption), both effects leading to decrease the value of the permeability. This statement is confirmed by the results obtained from permeability tests performed on J-TA and J-TB samples shown in Table 2, where untreated jute permeability values obtained from the Carman Cozeny model are shown between brackets. Unfortunately, the treatment also produces some changes in the fabric architecture that could affect the permeability values. Intra-tow region, that is responsible for the microflow, is expected to be more compact due to the presence of PHB (Fig. 2), that decreases the available space for the flow, decreasing the local permeability. But PHB also sticks the fibers of the bundles together reducing the flow resistance in the inter-tow region (macroflow), increasing the permeability value in that region. From the balance of this two opposite effects is expected an increase in the global permeability due to the higher influence of the macroflow on that parameter. Nevertheless, this mechanism cannot explain itself such an increment in fiber permeability (more than 5-9 times fot TA and TB respectively). Reduction in the sink effect by the PHB is then an important mechanism that contributes to the increase in fabrics permeability

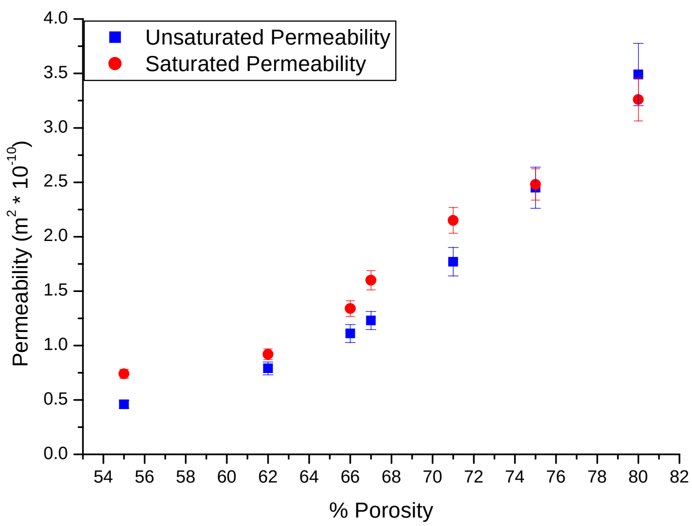

Fig. 3 Relationship between porosity and permeability for jute performs.

Table 1 Carman-Cozeny parameters

	K saturated	K unsaturated
n	0.9146	1.2886
C	8464 10^6	1338 10^7

Table 2 Permeability results of the jute treated preforms. Untreated jute permeability values (obtained with the Carman Kozeny equation to mach those porosities) are shown between brackets

Sample	Porosity (%)	K unsat (m^2 * 10^{-10})	K sat (m^2 * 10^{-10})
Jute - TA	45	1.25 (0,263)	1.74 (0,326)
Jute - TB	40	1.54 (0,177)	1.86 (0,446)

CONCLUSIONS

The relation between the porosity and the permeability was found for bidirectional jute reinforcement fabrics. As expected, both saturated and unsaturated permeabilities increase as porosity does. Also, it was observed that saturated permeability values are higher than unsaturated permeability values, but this difference tends to vanish for porosities above 75%, due to the low fiber volume fraction.

The fluid absorption of jute fibers and its effect on the permeability was also analyzed. It was found that jute reinforcements absorb great amount of fluid during the infusion process, when compared to glass fiber reinforcements. The fluid absorption affects the permeability value of the perform, because it removes fluid from the main stream as it travels through the reinforcement, acting as a sink component and thus decreasing flow velocity during the unsaturated flow. Furthermore, saturation of natural fibers can cause swelling, thus reducing the porosity and increasing flow resistance during saturated flow. So both, saturated and unsaturated permeability values of the jute perform are reduced because of the fluid absorption, and controlling this phenomenon is it possible to control fabrics permeability.

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