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Study of saturated and unsaturated permeability in natural fiber fabrics

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ABSTRACT

The main goal of this work is to understand how the main processing variables are affected when glass fibers are replaced by natural fibers in reinforced plastics. In this publication, a jute fabric was characterized in terms of its saturated and unsaturated permeability. It was found that fluid absorption and swelling are mechanisms present in natural fibers that reduce both permeabilities. Fluid absorption 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. Also, the saturation of the natural fibers cause swelling, reducing the porosity and increasing flow resistance during saturated flow.
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1. 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 industries. Many studies of sustainability and life cycle assessments have demonstrated 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, it is crucial to understand how the main processing variables are affected when glass fibers are replaced by natural fibers, which have different structure, different fabric architecture and different chemical interactions with the resins. One of those variables is the fabric 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]. 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 studying the injection process itself and the effect of using natural fibers on the processing variables. Richardson et al. 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 for the case of the recycled paper, the obtained values was high enough for infusing 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 when using 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. Recently, Liu and Dai [13] studied the impregnation of a jute fiber mat by a thermoplastic resin. They found for natural fabrics permeabilities an order of magnitude higher than the obtained for glass fiber mat. Due to the high viscosity melt used in their work, they did not observe impregnation inside fiber bundles.

Even though some permeability values have been reported for natural reinforcements, a detailed insight on their flow behavior is still required. In addition, the results obtained for one kind of fiber and fabric architecture are difficult to compare with that obtained for other fibers. Therefore, it is very 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. [14] and Diallo et al. [15] found that the saturated permeability

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was always lower than the unsaturated permeability, while other authors obtained opposite results [16–18]. Also, results have been reported showing that the saturated and unsaturated permeabilities were almost equal [19]. These discrepancies are usually attributed to experimental issues that could modify the saturated and unsaturated permeability ratio, such as mold deflection, capillary effect, microscopic flow, fiber channeling, and air bubbles [18]. Pillai and Advani [20] 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. Micro flow can also occur through the micro pores generated during the stacking and compression of the layers of reinforcement. Unlike 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 [21], and it could reduce the porosity and increase flow resistance during saturated flow.

The aim of this work is to achieve knowledge on mechanisms related to natural fibers impregnation in order to understand the resin flow behavior through natural fiber preforms, and improve the quality and performance of their composites. The relation between permeability and porosity of jute bidirectional fabrics is obtained, by using the Vacuum Assisted Resin Infusion (VARI) process. Both saturated and unsaturated permeability are obtained from the infusion tests. The Carman–Kozeny model is used in order to get an analytical relation between permeability and porosity. Furthermore, a brief analysis on jute fibers water absorption and its effect on permeability values is performed. Jute fabrics are coated with polyhydroxybutyrate (PHB) in order to reduce the fiber fluid absorption. PHB (a type of polyhydroxyalkanoate) is a biodegradable thermoplastic polymer with a high hydrophobic character. Permeability tests results obtained with jute and PHB treated jute fabrics are compared in order to study the fluid absorption effect on the permeability values. Also, the swelling degree of the treated and untreated fibers is determined and related to the saturated–unsaturated permeability ratio.

2. Experimental

2.1. Raw materials

Commercial bidirectional woven jute fabrics (Castanhal Textil, Brasil; surface density = 0.0300 g/cm^2) have been used in this study. The fabrics were washed with a 2% V/V distilled water and detergent solution, to remove contaminants and normalize the fabrics conditions for all the injections.

2.2. Treatment of PHB on natural fibers

In order to study the fluid absorption effect on permeability measurements, two different treatments were performed to make fibers less hydrophilic. The first treatment (treatment A = TA) consisted of wetting out the jute fabric with a 2% polyhydroxybutyrate (PHB, Biocycle-Brasil) in chloroform solution [22]. After wetting, the fabrics were air dried leading to surface density values of 0.033 g/cm^2. The second one (treatment B = TB) consisted of immerging the fabric into a container with 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 g/cm^2. Also, bidirectional glass fabrics were used for comparative purposes (surface density = 0.02 g/cm^2).

2.3. Flow experiments

The test fluid used for the flow experiments was a 22% V/V water/glycerin solution, leading to viscosity values near 0.150 Pa s. Few drops of red colorant were used to improve the flow front visibility. For the swelling tests, besides glycerin, two thermosetting resins were used: a commercial vinylester (Derakane 411-350, from Ashland) and a phenolic resin synthesized in our laboratory (molar ratio: 1.3; solid content: 53.67%) [23].

Unidirectional injection experiments were performed in a rectangular metallic mold (500 mm × 100 mm) with an acrylic lid. The depth of the mold cavity used for each injection was set in order to obtain the desired values of porosity. In order to avoid mold deflection during the infiltration tests, a 3 cm thick lid was used. The uniformity of the cavity depth was checked by putting clay in different parts of the cavity and conducting one injection. The depth of the mold cavity was estimated by measuring the thickness of the clay. Two injections were conducted for each porosity and type of fabric.

The viscosity of the fluid used was measured before every infusion by means of a Brookfield DV–II + cone and plate viscometer. 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.

2.4. Data analysis

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 the use of the quasi-steady-state assumption. Unsaturated permeability can be obtained using the following equation:

$$K_{\text{unsat}} = \frac{(\Phi \cdot m \cdot \mu)}{2\Delta P} \tag{1}$$

where K_{unsat} is the unsaturated permeability (m^2), m (m^2/s) is the slope of the curve x^2 (square of the flow front position) vs. time, μ is the fluid viscosity (Pa s) and ΔP (Pa) is the pressure drop along the fiber bed. The relation between the flow front position and the injection time was obtained by recording the infusion process with a camera mounted on top of the transparent flow cell.

Saturated permeability was calculated by measuring the fluid volumetric flow rate, once the reinforcement was fully saturated (Eq. (2)). A standard flow meter connected at the output line of the mold was used for this purpose and a plot of volume vs. time could be obtained.

$$K_{\text{sat}} = \frac{(Q \cdot \mu \cdot \Delta L)}{(A \cdot \Delta P)} \tag{2}$$

where K_{sat} is saturated permeability (m^2), Q the volumetric flow rate (m^3/s), ΔL the preform length (m) and A the mold cavity transverse area (m^2). The Carman–Kozeny model was used to establish a relationship between permeability and porosity. This relationship was developed to predict the behavior of a flow passing through a porous medium, and it was deduced by taking the medium as an arrangement of parallel tubes of any cross section [24]. The model has the following expression:

$$K = \frac{d_f^2}{k} \frac{\phi^3}{(1-\phi)^2} \tag{3}$$

where ϕ is the porosity, d_f the fiber diameter and k is the so-called Kozeny constant. Unfortunately, for many types of performs the assumptions behind the Carman–Kozeny model are not justified [25], and this equation is not able to properly fit permeability

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