Capillary Effects in Vacuum-Assisted Resin Transfer Molding With Natural Fibers

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The study of the capillary flow developed during the processing of composite materials is key because it acts as an important driving force for the impregnation of the fiber tows. It is also the main mechanism of void formation during infiltration of the fibers. In this work, capillary pressure of jute/vinylester composites was measured and the impact of capillary forces on fabric permeability was analyzed. It was found that the capillary pressure was significantly higher in vegetal than in synthetic fiber fabrics. In addition, the permeability of the fibers was characterized using various fluids. The resulting permeability was influenced by the nature of fluid and its polar property. Finally, the capillary pressure measured by this work was used to correct the experimental permeability in order to obtain a property independent of the test fluid. POLYM. COMPOS., 33:1593–1602, 2012. © 2012 Society of Plastics Engineers

INTRODUCTION

In the last years, the automotive industry has shown a growing interest in the use of natural fiber composites (NFC). Natural reinforcements have the advantage of being renewable, abundant, and cheaper than synthetic fibers. In addition, natural fibers are more environmentally friendly than synthetic and ceramic ones. They also possess good mechanical properties. In the automotive industry, parts that combine low weight and good mechanical properties are especially important considering that the reduction in vehicle weight results in less fossil fuel consumption and therefore in a reduction in the emissions of greenhouse gases (CO_2, CH_4). At the same time, natural fibers consume the same amount of CO_2 during plant growth as that produced during fiber degradation. By replacing synthetic or ceramic fibers by natural fibers in composite parts, more than 50% of the material can be obtained from renewable resources.

Liquid composite molding (LCM) techniques have proven to be suitable for processing NFC [1]. In previous works [2, 3], we have studied the relationship between saturated and unsaturated permeability and the compaction response of natural fiber fabrics. Nevertheless, there are still some practical issues regarding the infusion of natural reinforcements, for example, the difficulty to impregnate the fibers under vacuum at room temperature, which leads to important void formation within the tows. Capillary pressure has an important influence on the impregnation of natural fibers due to their hollow shape. Capillary pressure is defined as the energy per unit volume of a porous medium needed for replacing a gas (or vacuum) by a liquid. Capillary effects have been shown to be determinant in the mechanism of void formation during infiltration of fabrics [4–7]. There is an optimal infiltration velocity that produces composites with minimal void content. Below that velocity, capillary forces dominate the infiltration process, leading to void formation in the inter-tow region, whereas above the optimal value, viscous flow dominates and the voids are created mainly within the fiber tows [8]. Residual porosity has a detrimental effect on the mechanical properties of composite materials. Leclerc and Ruiz [9] studied the impact

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of the injection conditions in resin transfer molding (RTM) manufacturing on the overall quality and mechanical performance of parts, both of which are strongly dependent on the percentage of macro- and micro-voids. Ruiz et al. [10] proposed an optimization methodology to reduce the percentage of macro-/micro-voids formed during RTM manufacturing. Basically, the optimization consists in calculating the capillary number at the fluid flow front and then correcting the injection flow rate at each time step to ensure the optimal capillary number at the flow front.

Capillary pressure is also important at low impregnation rates since it acts as the driving force for the impregnation of the reinforcement [11]. In general, capillary pressure is dependent on flow velocity, and for a certain fiber–fluid system, it can vary from negative values (wetting case, where flow is enhanced) to positive values (non-wetting case, where flow is retarded) when the velocity is increased.

Several studies have been conducted to determine the magnitude of the capillary pressure developed in synthetic fabric infiltration. Batch et al. [6] investigated the capillary impregnation of aligned fibrous beds and found an optimal fluid speed at which the micro- and macro-flow have the same rate and the void formation is minimized. Capillary pressures obtained for liquid dioctyl phthalate (DOP) and glass fiber (40–60 vol%) were in the order of 10 kPa. Patel and Lee [4] analyzed the resin-fiber wettability by conducting wicking tests and capillary pressure measurements with different test fluids, and found, as a general trend, an increase in the capillary pressure with the decrease in matrix surface tension. Also, Bayramli and Powell [12] conducted axial and normal wicking experiments to study the wetting dynamics of carbon fiber bundles and found that the wicking rate is governed by the narrow pores with local porosity lower than the average porosity of the bundles. These authors also demonstrated that high fluid velocities give large dynamic contact angles, which can adversely affect impregnation. Verrey et al. [13] conducted infiltration experiments in noncrimp fabrics with different test fluids and found negative values of capillary pressure for polyethylene glycol and lauryllactam 12 and positive values for epoxy resin. Other authors have used different approaches to model the capillary forces acting on fiber preforms. Lawrence et al. [14] modeled the effect of capillary pressure on fiber saturation in LCM processes and defined a new parameter called capillary ratio (C), which is the ratio between the capillary pressure and the external injection pressure. For higher values of the capillary ratio, the saturation of the fibers along the mold at the end of an injection was complete. As the capillary ratio decreased, full saturation was achieved only along 65% of the length of the mold, demonstrating the impact of capillary pressure at low flow velocities. Also, Dimitrovova and Advani [15] modeled the capillary pressure for the flow across an array of aligned cylindrical fibers, and by means of a free boundary program, they concluded that both surface tension and viscous forces influence relative permeability and capillary pressure.

In the case of natural fibers (like jute), capillary effects can be increased due to the hollow structure, low diameter, and a polar character of the fibers. Nevertheless, scanty work has been conducted to study capillary effects in natural fiber preforms. In this work, the capillary pressure developed during the infusion of jute woven fabrics with vinylester resin and a water/glycerin solution was obtained using the methods proposed by Verrey et al. [13], and its effect on fabric permeability measurements was analyzed. In addition, the effect of the injection method on the capillary pressure and the permeability measurements was studied.

THEORY

The process of fibers infiltration in LCM techniques has been historically described using Darcy's law, which states a proportional relationship between the volumetric flow rate (Q) through a cross-sectional area (A) and the pressure gradient per unit length $\Delta P/L$.

$$Q = -\frac{A.K}{\mu}\frac{\Delta P}{L} \tag{1}$$

Equation 1 shows Darcy's law for the case of unidirectional flow, where μ is the fluid viscosity. The constant of proportionality is the fabric permeability (K). The general expression for 3D flow is shown in *Eq. 2*, where $\bar{u}_x$, $\bar{u}_y$, and $\bar{u}_z$ are the fluid linear velocities in the three coordinate directions and ϕ is the porosity of the medium (defined as 1-fiber volume fraction).

$$\begin{matrix}\bar{u}_x \\ \bar{u}_y \\ \bar{u}_z\end{matrix} = -\frac{1}{\phi\mu}\begin{pmatrix}K_{xx} & K_{xy} & K_{xz} \\ K_{yx} & K_{yy} & K_{yz} \\ K_{zx} & K_{zy} & K_{zz}\end{pmatrix}\begin{pmatrix}\partial P/\partial x \\ \partial P/\partial y \\ \partial P/\partial z\end{pmatrix} \tag{2}$$

This equation is valid for fully saturated flow, but in LCM, the processes imply the infiltration of a dry reinforcement. Several authors have shown that the use of Darcy's law to describe unsaturated flow can lead to inappropriate predictions of the flow front position and pressure profile [11, 16–24], because it does not take into account the contribution of the capillary effects to the total pressure gradient. Thus, a more accurate expression of Darcy's law can be written if the capillary pressure is considered on the pressure gradient (ΔP). A more accurate expression of the pressure differences through the impregnated fibers can be written as follows:

$$\Delta P = P_{\text{int}} + \Delta P_{\gamma} - P_{\text{inj}} \tag{3}$$

where P_{int} is the pressure inside the mold prior to resin injection, P_{inj} is the injection pressure applied, and ΔP_{γ} is the capillary pressure developed at the flow front. The higher the capillary effects developed during fabric impregnation, the worse the prediction of the flow behav-

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ior using the saturated Darcy's law. Thermodynamically, the capillary pressure is defined as [25]:

$$\Delta P_\gamma = -S_f \gamma_{ma} \cos\theta \tag{4}$$

where γ_{ma} is the surface tension between the liquid, the fiber, and the surrounding gas, S_f is the area of liquid–fiber interface per unit volume of liquid, and θ is the dynamic wetting angle. A Whilhelmy micro-scale is generally used to measure γ_{ma} and cos θ while S_f can be determined by BET techniques [13]. Another way to measure the capillary pressure is the wicking test, where the liquid impregnates the fabric spontaneously, and the flow front position is recorded as a function of time. The value of capillary pressure obtained by these techniques is the value at the thermodynamic equilibrium, i.e., with no fluid flow. It is considered that the measured value is close to the equilibrium if the capillary number is below 10^{-5} [13]. This non-dimensional number relates the contributions of the capillary forces to the viscous forces as the fluid flows through capillary channels. The capillary number is defined as follows:

$$C_A = \frac{v_l \mu}{\gamma_{ma}} \tag{5}$$

where v_l is the liquid velocity.

However, in most infiltration processes, the liquid resin flows at a certain velocity, and the fluid behavior is not at the thermodynamic equilibrium. In addition, for viscous polymers, the dynamic contact angle θ is a function of the local fluid velocity [13], which indicates that the dynamic contact angle varies if the capillary number does. Therefore, the magnitude of the capillary pressure developed during the infiltration of the reinforcement should be measured in dynamic conditions. Verrey et al. [13] used a method to measure the capillary pressure in situ while performing infiltration tests identical to those used to calculate the fabric permeability. The same technique had been previously used by Amico and Lekakou [11] to determine capillary pressure in glass woven fabrics. The main assumption of the method is that the flow front is in accordance with the slug-flow assumption, where the fabric is either fully dry or fully impregnated [25] with a sharp flow front in between. Then, considering an incompressible reinforcement and an unidirectional flow, it can be assumed that after a certain time "t" the front flow reaches position $\ell(t)$, as shown in Fig. 1. The pressure acting on the dry region of the preform is the atmospheric pressure, P_{int}, while capillary and Darcy's pressures are applied in the partially and fully saturated regions of the part. Darcy's law is usually solved in one dimension to obtain the expression that describes the flow front advancement:

$$\frac{d\ell}{dt} = -\frac{K}{\mu\phi}\frac{dP}{dx} \tag{6}$$

where K is the permeability of the reinforcement in the x direction.

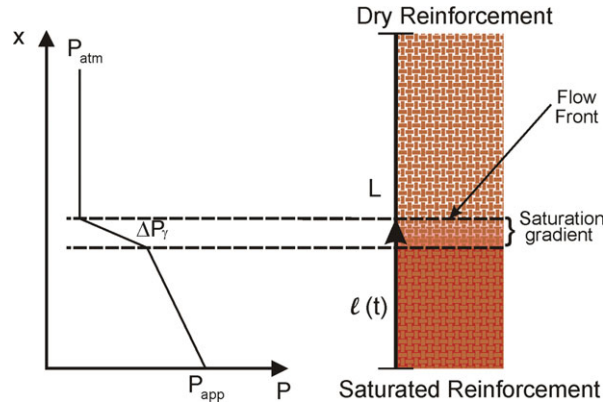

FIG. 1. Pressure distribution along the fiber bed. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

As in the infiltration process of preforms for permeability determination, there are two different ways to measure the capillary pressure: infiltrating the reinforcement with a constant flow rate or at a constant applied pressure. If the experiments are performed at constant applied pressure, and considering the mass conservation law, dP/dx equals $\Delta P/\ell(t)$, where ΔP is defined by *Eq. 3*. Then, by integration of *Eq. 6*:

$$\ell^2 = \Psi^2 t \tag{7}$$

$$\Psi^2 = -\frac{2K}{\mu\phi}(P_{int} - P_{inj} + \Delta P_\gamma) \tag{8}$$

Therefore, the capillary pressure drop can be obtained by plotting the parameter Ψ^2 as a function of the pressure difference $\Delta P = P_{int} - P_{inj}$ for several experiments carried out at different applied pressures. The dependence should be linear (according to *Eq. 8*), and the capillary pressure is obtained by extrapolating the linear regression curve to $\Psi^2 = 0$ and noting the corresponding value of $\Delta P = P_{int} - P_{inj}$.

In experiments performed at constant flow rate, *Eq. 6* is still valid. In this case, $d\ell/dt$ is constant and equal to L/t, where L is the total length of the fiber bed. Then, by substituting the total length by its expression using the flow rate and time, the following expression can be obtained:

$$(P_{inj} - P_{int}) = -\frac{Q^2\mu}{A^2 K\phi}t + \Delta P_\gamma \tag{9}$$

Therefore, the capillary pressure can be determined in a single experiment by plotting the pressure difference at the inlet as a function of time. The value at time zero, when the flow front touches the fiber bed, is a direct measurement of the capillary pressure drop. This procedure is explained with more detail in the Results and Discussion section, where the capillary pressure was obtained by extrapolating the linear region of the curve (Region II in Fig. 6) to the pressure value at time zero.

It should be taken into account that in the constant pressure tests, the flow front velocity decreases while the fluid travels through the preform, which affects the dynamic contact angle and, thus, the capillary pressure developed at the different flow front positions. Therefore, the value obtained in this case is an averaged capillary pressure. On the other hand, in the constant flow rate method, the fluid velocity is constant, thus the results are more representative and correspond to the capillary pressure developed at a given flow velocity.

EXPERIMENTAL

Materials and Fluids

The capillary pressure developed during the infusion was measured on commercial bidirectional woven jute fabrics (Castanhal Textil, Brazil; surface density = 300 g/m^2). The fabrics were washed with a 2% vol/vol distilled water and detergent solution, and then dried under vacuum at 70°C for 24 h to remove contaminants and normalize the fabrics conditions for all the injections.

The test fluids used were a 22% vol/vol water/glycerin solution (surface tension = 63 ± 0.1 mN/m) and a vinylester resin (Derakane 411-350, from Ashland, surface tension = 37 ± 0.1 mN/m). The surface tension was measured using the ring method, and the viscosity of the fluids was measured before every infusion by means of a Brookfield DV-II+ cone and plate viscometer (precision ± 0.0025 Pa s).

Micro-Structural Observation

The structure of the natural fibers was analyzed by scanning electron microscopy (SEM, model JEOL JSM 6460 LV). In addition, the microstructure of the fiber yarns was observed using an optical microscope (Olympus PMG 3).

Permeability Tests

Unidirectional injection experiments were performed in a rectangular steel mold (500 mm long by 100 mm wide). The thickness of the mold cavity was set to obtain the desired porosity on each injection. In order to avoid mold deflection during the infiltration tests, a 3 cm thick acrylic lid was use. The uniformity of the cavity thickness was checked by putting clay in different parts of the mold cavity. Two injections were conducted for each porosity and type of fabric.

A vacuum pump was used to infuse the liquid into the mold cavity. The pressure gradient achieved was measured with a vacuum gauge (precision ± 1 kPa), located at the outlet line of the mold.

Darcy's law for unidirectional flow was used to estimate the unsaturated permeability of the natural fibers.

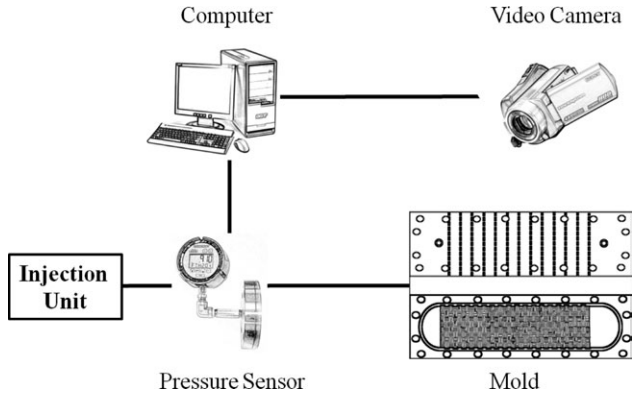

FIG. 2. Experimental setup used for permeability measurements.

Unsaturated permeability was obtained using *Eq. 10* as follows:

$$K_{\text{unsat}} = -\frac{(\phi m \mu)}{2\Delta P} \tag{10}$$

where K_{unsat} is the unsaturated permeability (m^2), ϕ is the porosity of the preform, m is slope of the curve ℓ^2 (square of the flow front position) vs. time, μ is the fluid viscosity (Pa s) and ΔP (Pa) is the total 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.

Figure 2 shows schematically the experimental setup used in this work. The injection unit corresponds to a vacuum pump in the constant pressure experiments or a double action piston machine that pumps the fluid at a constant flow rate.

It should be taken into account that the permeability was calculated using the standard procedure and the capillary pressure was not considered on the pressure gradient. Therefore, the differences observed among the permeability curves were useful to determine the effect of the capillary forces on the permeability of the preform.

Constant Pressure Capillary Experiments

The capillary pressure developed in jute preforms was calculated for a single porosity value of 60%, following the constant pressure procedure described by Verrey et al. The infiltration experiments were conducted in the same way as the permeability test. The vacuum level of the pump was controlled to obtain different applied pressure gradients.

Constant Flow Rate Capillary Experiments

The capillary pressure at the flow front during the infiltration of jute preforms was calculated with the constant flow rate procedure described by Verrey et al. The mold used in the experimental setup was identical to the one

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used in the permeability tests, and it was connected to a machine that pumps the fluid at constant flow rate, which was set at 0.075 ml/s through a computer interface. A pressure transducer was placed at the injection gate of the mold to measure the applied pressure as a function of time. Therefore, capillary pressure could be calculated following the procedure proposed by Verrey et al. for different fiber contents. In order to guarantee a representative value, five measurements obtained at each experimental condition were averaged to obtain the reported values.

RESULTS AND DISCUSSION

Micro-Structural Observations

In plant fibers, the technical fiber is a bundle (50–100 μm) of elementary fibers (10–20 μm). These fiber bundles are called macro-fibrils and are glued together with a matrix of lignin and hemicellulose. Macro-fibrils are composite materials made of several layers of cellulose microfibrils (which are the main responsible ones for the mechanical properties of the fiber) embedded in a matrix of lignin and hemicellulose. The elementary fibers are seen as hollow tubes, where the open channel on the center of the macro-fibril is called lumen. SEM images of jute fibers are shown in Fig. 3a. Furthermore, the surface of the natural fiber is rough, as can be seen in the micrograph, and the yarns are composed of discontinuous short and twisted fibers (see Fig. 3b). This generates numerous micro-channels in which capillary flow can occur.

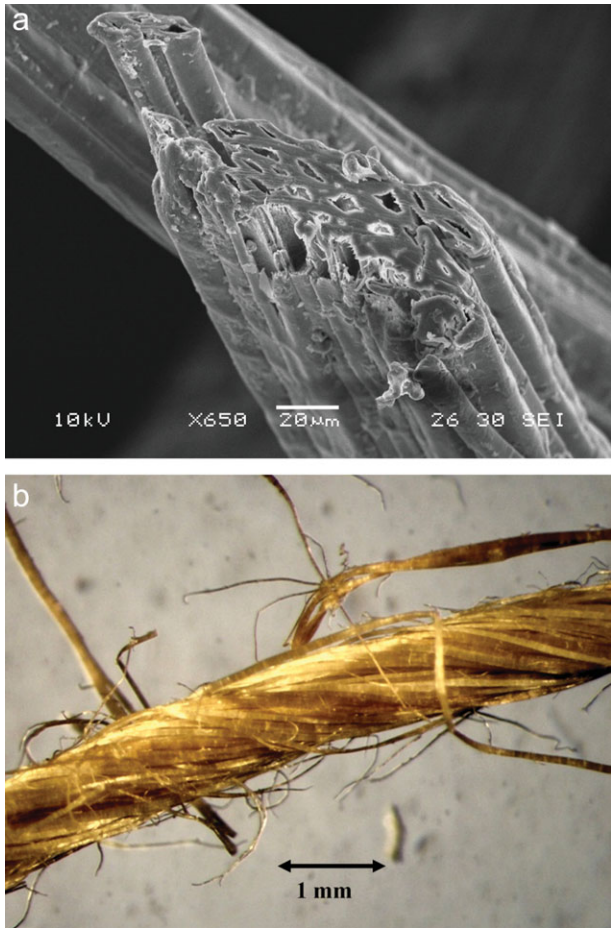

FIG. 3. (a) SEM image of a jute fiber. (b) Optical micrograph of a fabric yarn made of jute fibers. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

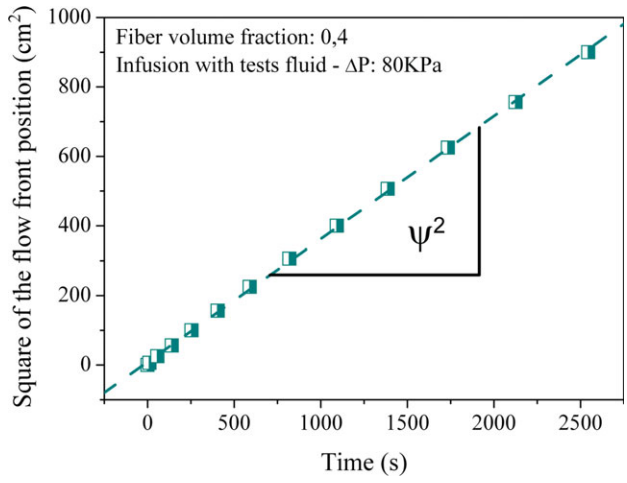

FIG. 4. Constant pressure procedure. Determination of Ψ^2 from flow front position vs. time data. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Therefore, the magnitude of the capillary pressure developed during impregnation of natural fiber reinforcements is expected to be high, due to the hollow structure of the fibers and the fiber disposition in the yarn.

Capillary Pressure Measurements

Constant Pressure Procedure. The relationship between the square of the flow front position and time obtained with a video camera was used to calculate the parameter Ψ^2 from *Eq. 7*. Neither fiber washing nor race tracking was observed during the experiments and, as a consequence, the flow front remained straight during injections. Therefore, the "slug-flow" assumption remained valid and the curve resulted perfectly linear, as shown in Fig. 4. This curve corresponds to preforms compressed to a volume fraction of 0.4, and injected with test fluid with an applied pressure gradient of 80 kPa. Similar plots were obtained in other experimental conditions. This procedure was repeated for different applied pressure gradients, to establish a relationship between pressure and Ψ^2 according to *Eq. 8*.

The viscosity of the fluid changed from test to test due to variations in the room temperature. Therefore, in order to plot all the experimental data in the same graph, the coefficient Ψ^2 was multiplied for the fluid viscosity and this product was plotted against pressure gradient. Figure 5 shows the procedure used to estimate the capillary pres-

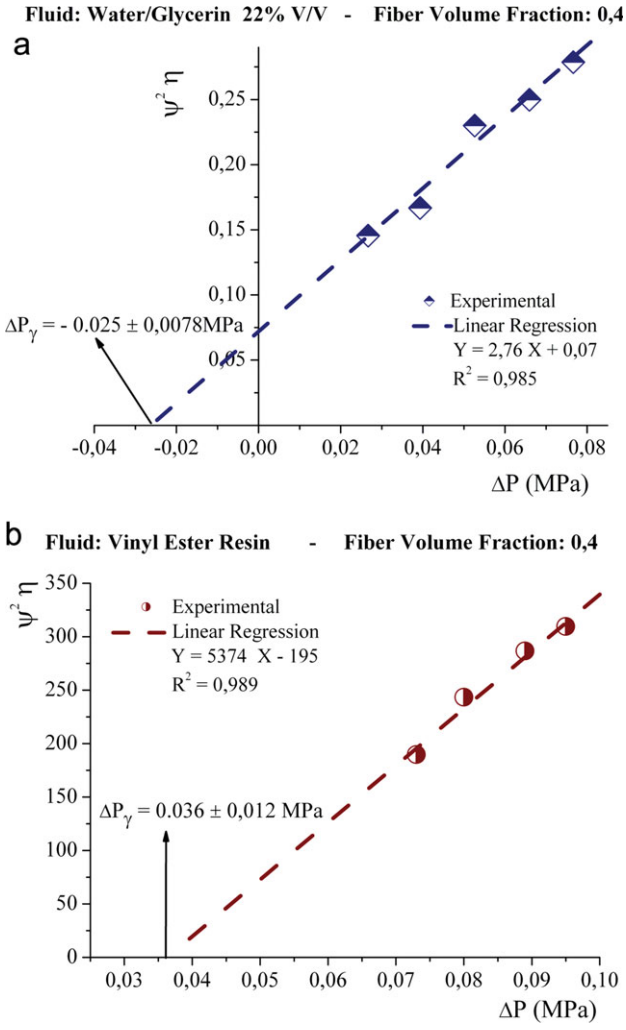

FIG. 5. Experimental determination of the capillary pressure developed during the infusion with (a) water/glycerin solution and (b) vinylester resin. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

sure developed on jute preforms with a fiber volume fraction of 0.4, infused with the test fluid (Fig. 5a) and a vinylester resin (Fig. 5b).

Capillary pressure was negative in the jute–water/glycerin system and positive in the jute–vinylester resin system. These results show that spontaneous infiltration takes place during the infusion of jute fabrics with the test fluid, while the capillary forces act against the flow when vinylester resin is used. This behavior was expected due to the polar character of the water/glycerin solution, which is more compatible with the hydrophilic nature of the jute fibers than the vinylester resin. The magnitude of the capillary pressure was -0.025 ± 0.008 MPa and 0.036 ± 0.012 MPa for the test fluid and the resin, respectively. The errors were estimated with respect to the variation of the linear regression. Although these values are in the order of magnitude of the values found by other authors [6, 24], the capillary forces are higher, probably due to the imperfect and hollow structure of natural fibers (see Fig. 3). It is interesting to note that the capillary pressure can represent more than 20% of the external applied pressure when the water/glycerin fluid was injected at constant pressure and more than 35% when the vinylester resin was used. These results demonstrate the importance of taking into account the capillary effects to estimate permeability and to evaluate the manufacturing of LCM parts. During an infiltration process at constant pressure, the fluid velocity changes and so does the capillary number. As a consequence, the capillary pressure changes during the infiltration process

Constant Flow Rate Procedure. A more accurate way to measure capillary pressure is to use constant flow rate infiltrations. Figure 6 shows an example of the plot obtained from the pressure transducer located at the injection gate of the mold during such experiments. In this case the fiber volume content was 35%. Verrey et al. [13] identified three different regions of the plot, as shown in Fig. 6 (see Regions I–III). These authors attributed the first nonlinear part of the curve (Region I) to a transient state, in which the resin progressively saturates the porous media and the "slug-flow" assumption is not valid. After certain time, a steady regime where the saturation gradient at the front flow is narrow and the "slug-flow" assumption is applicable is reached. In this region of the plot (Region II), the curve is linear, as expected from *Eq. 9*. Once the flow front reaches the end of the preform, the pressure increase at the resin inlet is no longer observed (Region III). The capillary pressure was calculated as the intersection of the linear regression curve of Region II with the pressure axis, as shown in Fig. 6.

In order to obtain statistically relevant values, five measurements were made for each experimental condi-

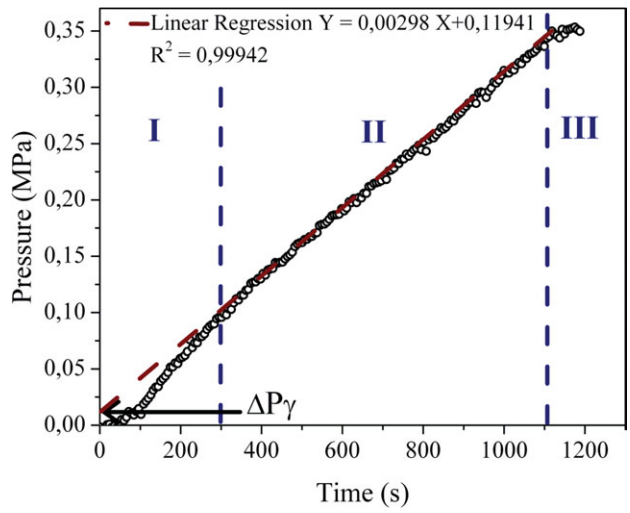

FIG. 6. Experimental determination of the capillary pressure using a constant flow rate injection and water/glycerin fluid. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

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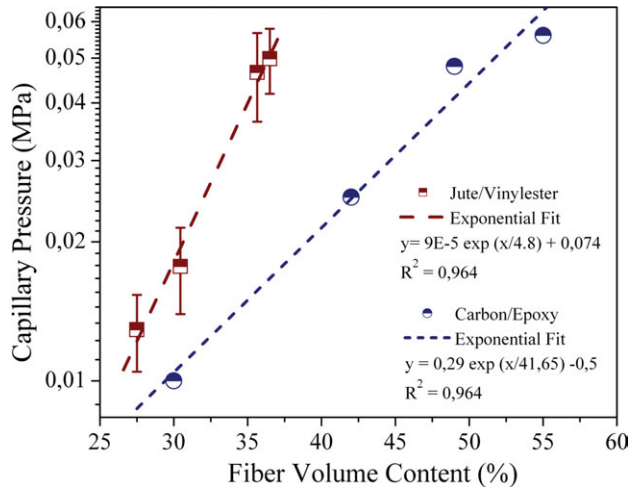

FIG. 7. Capillary pressure drop as a function of the fiber volume content obtained from constant flow rate experiments. Results for carbon fibers and epoxy resin are shown for comparison purposes [13]. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

tion. Figure 7 shows the capillary pressure vs. fiber volume fraction for jute using vinylester resin as injection fluid. Results from the literature for carbon fibers and epoxy resin were added to the plot for comparison purposes [13]. When the fiber volume fraction increases, the capillary forces grow, but unlike the experiments conducted with the glycerin solution, the values are positive. In accordance with the results obtained with the constant pressure procedure, no spontaneous infiltration takes place with the vinylester resin. When comparing with the results obtained in the literature, it can be seen that capillary effects grow at a high rate in the natural fiber fabrics.

Effect of the Type of Fluid on Permeability Measurements

Figure 8 shows the permeability of the jute fabric for different porosities measured with the two different test fluids. As observed by other authors [18, 19], the dependence of unsaturated permeability on the type of fluid evidences the importance of the wetting behavior and capillary forces acting during the infiltration process. Although the saturated permeability is a geometrical property not dependent on the pressure field or the capillary pressure, the measured transient permeability (or unsaturated permeability) is affected by the pressure distribution along the fiber bed. This issue arises by the procedure commonly used to calculate the transient permeability based on Darcy's law, in which the pressure gradient taken into account ignores the capillary pressure contribution to the flow movement. Therefore, if the magnitude of the capillary pressure is important, the transient permeability could be underestimated if the capillary opposes to the flow, and overestimated if it enhances the flow.

Due to its chemical composition, the glycerin solution has a higher polar character than the vinylester resin. Unlike synthetic fibers, natural fibers have hydroxyl groups in their surface, which make them more compatible with polar fluids. Therefore, as shown in the previous section, the capillary pressure was negative and enhances flow when the reinforcement was injected with a water/glycerin solution, while it was positive (against flow) when vinylester resin was used. This difference in the capillary pressure sign leads to an increase in the measured transient permeability when the test fluid is injected and to a lower permeability value when vinylester resin is used.

In addition, Fig. 8 shows that the difference between the measured transient permeabilities with both fluids increases for higher fiber volume contents. This behavior is assumed to be a consequence of the pore size in the reinforcement, which decreases when the fiber volume content increases. At higher V_f, the capillary forces become dominant and the injected fluid has a greater influence on the transient permeability value. At lower V_f (i.e., higher pore size), the capillary forces are less important and viscous forces dominate the fluid flow. As a consequence, the transient permeability curves with the two fluids converge to the same value at lower V_f.

From the previous observations on the impact of capillary forces on transient permeability measures, it can be concluded that unsaturated permeability data can be corrected if the capillary forces acting at the fluid flow front are known. As an example of this statement, a correction was carried out for the transient permeability values obtained for a fiber volume content of 40% (see Fig. 8). To correct these unsaturated permeability values, the capillary pressures measured for the same fiber volume content (see Fig. 7) were added to the measured pressure gradient during the permeability test.

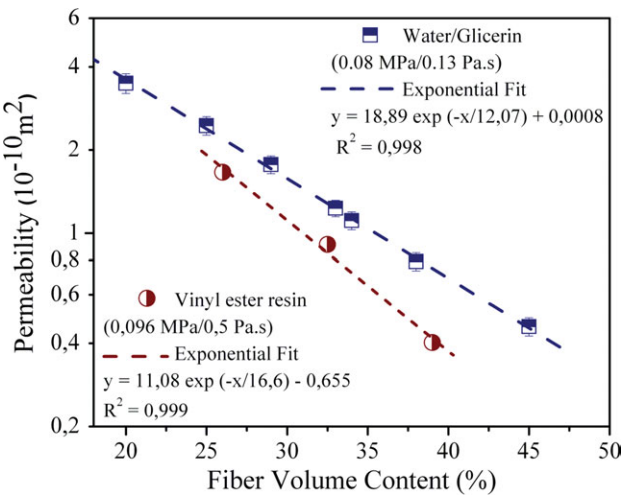

FIG. 8. Transient permeability of jute fibers measured with different test fluids. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

TABLE 1. Corrected permeabilities obtained with *Eq. 11* for a V_f = 40%.

	Water/glycerin	Vinylester resin
Original pressure gradient—ΔP (MPa)	−0.08	−0.096
Corrected pressure gradient considering capillary effects—$\Delta P_{corrected}$ (MPa)	−0.105	−0.06
Original permeability—K_{unsat} (m^2)	6.95 $E-11$	3.48 $E-11$
Corrected permeability considering capillary effects—$K_{corrected}$ (m^2)	5.3 $E-11$	5.57 $E-11$

Taking into account the capillary effects, the pressure gradients measured in the permeability mold can be corrected as follows for the two fluids used in this test:

(a) for the impregnation of jute fibers with water/glycerin solution

$$\begin{aligned}\Delta P_{corrected} &= P_{int} + \Delta P_\gamma - P_{inj} \\ &= -0.08\,\text{MPa} + (-0.025\,\text{MPa}) - 0\,\text{MPa} \\ &= -0.105\,\text{MPa}\end{aligned}$$

(b) for the impregnation of jute fibers with vinylester resin:

$$\begin{aligned}\Delta P_{corrected} &= P_{int} + \Delta P_\gamma - P_{inj} \\ &= -0.096\,\text{MPa} + 0.036\,\text{MPa} - 0\,\text{MPa} \\ &= -0.06\,\text{MPa}\end{aligned}$$

The corrected transient permeability, $K_{corrected}$, can then be calculated from *Eq. 10* by replacing the pressure gradient ΔP by the corrected value $\Delta P_{corrected}$ for each fluid. Therefore, it can be written that:

$$K_{corrected} = \frac{K_{unsat}.\Delta P}{\Delta P_{corrected}} \tag{11}$$

Equation 11 was then used to correct the measured permeabilities using the two reference fluids. The results of the permeability correction for a fiber volume fraction of 40% are presented in Table 1. It results from this calculation that both permeabilities converge to nearly the same value, with a difference below 5%. This transient permeability value is therefore independent of the fluid used for the experiment.

Effect of the Injection Method on Permeability Measurements

Capillary effects depend on the dynamic contact angle, which is affected by the flow front velocity as described by the Hoffman–Voinov–Tanner law [26]:

$$\theta^3 - \theta_0^3 \cong C_T\, C_A \tag{12}$$

where C_T is a coefficient which needs to be experimentally determined, and θ_0 is the contact angle at thermodynamic equilibrium. Therefore, a relationship between the capillary pressure and the capillary number can be obtained combining *Eq. 4* and *Eq. 12*:

$$\Delta P_\gamma = -S_F \gamma_{ma} \cos\left(\sqrt[3]{C_T\, C_A + \theta_0^3}\right) \tag{13}$$

Verrey et al. [13] experimentally verified the validity of the Hoffman–Voinov–Tanner law on epoxy resin, and found that the capillary pressure increased with the capillary number. The equilibrium capillary pressure was negative, thus at a capillary number about 0.014 the capillary pressure was zero and above that value the behavior of the resin changed from wetting to non-wetting.

Table 2 shows the capillary numbers obtained in all the experiments performed in this study. It can be seen that experimental conditions led to capillary numbers higher that 10^{-5}, which means that the thermodynamic equilibrium was not respected. Therefore, static measurements of the capillary pressure would not have been accurate. In addition, the capillary number changed in more than one order of magnitude in the constant pressure procedure.

In accordance with that found by Verrey et al. [13], the capillary pressure calculated with the constant flow rate procedure was higher than that calculated with the constant pressure test. This difference can be explained considering the influence of the capillary number on the capillary pressure. Constant pressure experiments led to a wide range of capillary numbers because the fluid velocity decreased as the front moved further in the preform and also, the experiments were carried out with different applied pressure gradients. On the other hand, the capillary number was constant (and equal to 0.0034) through all the preform length during the constant flow rate experiments. Figure 9 shows the capillary numbers obtained during the constant pressure and constant flow rate experiments, as a function of the percentage of the preform impregnated by the liquid flow. It can be seen that the capillary number was lower in the constant pressure experiments in more than 60% of the flow distance in the tests performed with the maximum pressure gradient (95 kPa), and in more than 75% in the tests performed with the lowest pressure gradient (73 kPa). Consequently, as the vinylester resin behavior was non-wetting in both cases, the constant flow rate experiments led to a higher capillary pressure than the constant pressure tests, because the resin flow reached higher capillary numbers.

The effect of the capillary number on the capillary pressure can also be seen in the results obtained in the transient permeability tests. In both the constant pressure and constant flow rate experiments, vinylester resin was used and, as a result, no differences were expected because permeability should be a property of the reinforcement. However, the transient permeability of the jute woven fabric calculated from the constant pressure experiments was higher than that calculated with the constant flow rate procedure (Fig. 10). These results are consistent

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TABLE 2. Capillary number obtained in all the experimental conditions.

Experimental setup	Porosity (%)	Viscosity—μ—(Pa s)	Fluid velocity range—v_L—(m/s)	$C_A = \frac{\mu_L v}{\gamma_{ma}}$
Surface tension of vinylester resin—γ_{ma}— = 0.037 N/m				
Capillarity pressure tests—Constant flow rate—Resin	63.5–65–70–72.5	0.502	0.00025	0.0034
Capillarity pressure tests—Constant pressure—Resin	60	0.55–0.65	$0.0023–5 \times 10^{-5}$	$0.04–4.8 \times 10^{-4}$
Capillarity pressure tests—Constant pressure—Water/Glycerin	60	0.110–0.140	0.013–0.00035	$0.027–7.2 \times 10^{-4}$
Permeability tests—Constant pressure—Resin	61	0.650	0.0023–0.0005	0.04–0.009
	68	0.490	0.0036–0.0001	0.05–0.0013
	74	0.52	0.0042–0.0001	0.06–0.0014
Permeability tests—Constant pressure—Water/Glycerin	55	0.11	0.0028–0.0002	$0.005–4.1 \times 10^{-4}$
	80	0.13	0.0083–0.0009	0.017–0.0019
Surface tension of water/glycerin solution—γ_{ma}— = 0.063 N/m				

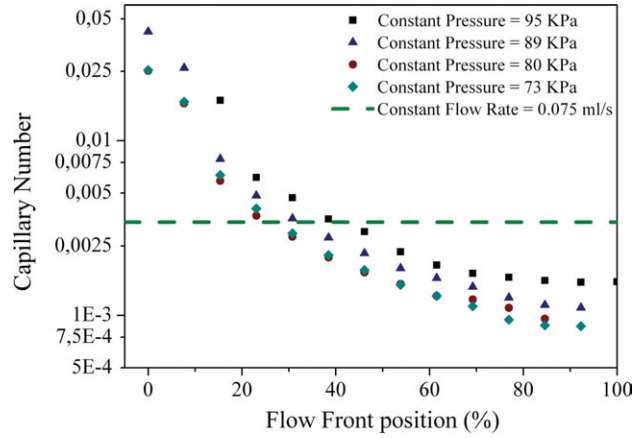

FIG. 9. Capillary numbers obtained in constant pressure and the constant flow rate tests, as a function of the flow front position. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

with the capillary pressure values found in both tests. This pressure drop is positive in both cases, so the lower capillary pressure developed in the constant pressure experiments had less effect on delaying the flow front, and the measured transient permeability resulted higher. These results show, in accordance with that found by other authors [16, 17, 20–22], that the injection method and flow rate affect the unsaturated permeability values, and therefore, that all the experimental conditions should be reported together with permeability results.

CONCLUSIONS

The capillary pressure drop developed at the flow front during infusion of jute woven fabric was calculated using the methods proposed by Verrey et al. [13]. These methods were chosen because they allow the capillary pressure

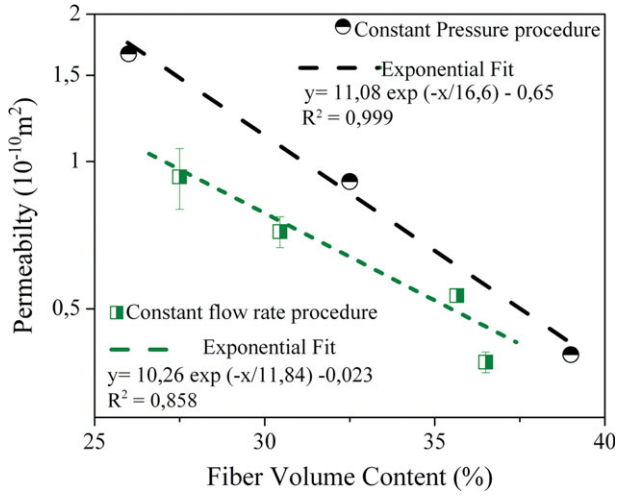

FIG. 10. Transient permeability determined with different injection methods: constant flow rate and constant applied pressure. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

to be estimated under dynamic conditions, which is more representative of the real infusion process. Both constant flow rate and constant applied pressure experiments were performed. However, in accordance with that found by other authors, our results showed that the capillary pressure depends on the capillary number. Thus, the results obtained from the constant pressure experiments represent an average capillary pressure, because this parameter changes strongly during impregnation of the fiber bed. Therefore, the constant flow rate experiments provided more consistent results.

The effect of the type of fluid on the capillary pressure was observed on the constant pressure experiments. The polar nature of the water/glycerin solution makes this fluid very compatible with the hydroxyl groups on the fiber surface, and the capillary pressure resulted negative, i.e., enhancing flow. On the other hand, the lower polar nature of the vinylester resin led to positive values of capillary pressure.

The results obtained with the constant flow rate experiments showed that no spontaneous infiltration occurs when jute preforms are injected with vinylester resin, in accordance with the results obtained with the constant pressure method. These experiments also showed that capillary pressure increases with fiber volume content.

Overall, we found that the magnitude of capillary pressure in natural fiber fabrics was two to three times higher than that reported for synthetic fibers, because the hollow and imperfect structure of natural fibers provides more capillary channels in which micro-flow can occur. Therefore, it is important not to ignore this effect to avoid miscalculation of the unsaturated permeability, especially when using low injection pressures. Although saturated permeability is supposed to be a property of the reinforcement, we found that transient (unsaturated) permeability was affected by the injection fluid and the procedure used to measure this property. If the magnitude of the capillary pressure is significant (as it is in natural fiber fabrics), the permeability measured using fluids more compatible with the fibers will be higher than that measured with less compatible liquids. In this study, the measured capillary pressure was used to correct the pressure gradient applied in the permeability tests. Consequently, a corrected transient permeability independent of the test fluid was obtained. This correction resulted in a transient permeability value as close as 5% between the one measured using a vinylester resin and the one obtained with a water/glycerin solution, demonstrating the importance of taking into account the capillary effects on such characterization.

In addition, we found that the injection method affects the capillary number and therefore, the capillary pressure drops. As a consequence, the measured transient permeability values depend on the injection method. Finally, we found that the lower capillary pressure developed in the constant pressure experiments had less effect on delaying the flow front, and that the measured permeability thus resulted higher than that obtained with the constant flow rate experiments.

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