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Key differences on the compaction response of natural and glass fiber preforms in liquid composite molding

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Abstract

In the present work, the effects of fiber structure and fluid absorption on the compaction behavior of jute woven fabrics and sisal mats were analyzed and compared with the response of glass fiber mats. It was found that the fiber content that can be achieved with a certain compaction pressure is lower in the case of natural fiber preforms. In addition, due to the hollow structure of these natural fibers, jute and sisal preforms suffered larger permanent deformation than glass fiber preforms after the compressive loading cycle. In addition, it was found that fluid absorption reduced the compaction pressure in natural reinforcements due to fiber softening. These phenomena were not observed in glass fiber mats.

Keywords

Natural fiber reinforcements, glass fiber mats, liquid composite molding (LCM), resin transfer molding (RTM), compaction

Introduction

The low cost and the renewable character of natural fibers have made these fibers an attractive alternative to synthetic fibers as reinforcements in composite materials. In addition, natural fibers present other advantages over synthetic fibers:[1] low density, good specific properties, no health risk and easy availability in some countries. In order to manufacture high quality green composite parts, advanced processing techniques such as liquid composite molding (LCM) could be used. In these techniques fibers do not suffer thermal or thermomechanical degradation as in thermoplastic matrix manufacturing processes. In every LCM process such as resin transfer molding (RTM) or vacuum assisted resin transfer molding (VARTM) the reinforcement is compressed inside a mold and a thermoset resin is injected to fill the empty spaces in the mold. After the resin cures, the composite part is demolded.[2] The study of the compaction response of fibrous preforms is important because it will determine the maximum fiber content that can be achieved with the available clamping forces. It is also very important in single-sided molding processes like VARI or SCRIMP, where the maximum compaction pressure is limited to the atmospheric pressure, 1 bar. It should be taken into account that in general, the higher the fiber volume fraction of a composite is, the higher is its performance. Moreover, in composites made with natural fibers and thermoset resins, higher fiber contents will result in more eco-friendly composites, since the percentage of materials from renewable sources is increased.[3] Besides the maximum compaction pressures needed to achieve a certain fiber volume fraction, the other properties that characterize the compaction behavior of fibrous preforms are the permanent deformation after subsequent loading cycles[4–6] and the stress relaxation that occurs while the desired thickness is kept constant.[7–9] The permanent

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deformation of woven preforms is attributed to the irreversible yarn cross-section deformation, flattening, and nesting of the yarns, while in random mats permanent deformation is caused by the filaments from one layer of fabric that are intertwined or embedded in the adjoining layer. In most LCM processes, a pre-forming step is done prior to placing the fabrics in the mold. This step consists of compressing a stack of fabric layers in a mold to a shape and thickness close to those of the final part. A binder is sometimes used to avoid preform deformation during handling. Preforms that experience high permanent deformation are easier to place in the mold and because their thickness is closer to the thickness of the final part, the clamping forces needed are lower. In the same way, a higher fiber volume fraction can be obtained after a pre-forming step in single-sided molding processes such as VARI. Therefore, during the processing of composite materials, subsequent loading cycles are expected to take place. Stress relaxation means that the load required for maintaining a fixed thickness in a preform decreases with time. In the same way, if a constant load is applied to a preform as happens in the vacuum infusion technique, its thickness will decrease with time. This phenomenon is known as fiber settling.

The presence of a fluid affects the compaction properties of fibrous materials, as was demonstrated by several authors.[10–12] The fluid acts as a lubricant and reduces the clamping forces needed to achieve the desired volume fraction. On the other hand, an internal fluid pressure is generated as the compaction plate descends and the fluid is drained through the preform[6,7] which increases the compaction pressure. In addition, the permeability of the preform decreases as the thickness is reduced during the compaction test, increasing gradually the fluid pressure.

Natural fibers are different than synthetic fibers in structure and chemical composition. They can be classified into several categories, such as wood fibers, vegetable fibers, animal fibers and mineral fibers. Jute and sisal fibers belong to the group of vegetable fibers, and are extracted from the bast and the leaf of the plants respectively.[13] The technical fiber is a bundle (diameter: 50–100 μm) of elementary fibers (diameter: 10–20 μm) called macro fibrils glued together with lignin and hemicelluloses. These macro fibrils are made of several layers of cellulose microfibrils embedded in a matrix of lignin and hemicelluloses. The elementary fibers have an open channel on the centre called lumen. Furthermore, these natural fibers are highly hydrophilic as a consequence of their chemical composition. Many authors studied the effect of the distinctiveness on the mechanical properties of the fibers[14,15] and the composites.[16–20] Li et al.[21] conducted a very interesting review on the processing by RTM of natural fiber based composites and their mechanical properties. In addition, Masoodi and Pillai[22] wrote a review about the modeling of the processing by LCM of composites made with biofibers. It was shown in previous works[23] that during the resin injection stage, the permeability of these natural fiber preforms is reduced by fluid absorption and fiber swelling. Some authors studied the compaction behavior of wood fiber mats,[6] and they found that the impregnation with polar liquids reduced the compaction forces due to the fluid absorption and softening of the wood fibers, while non-polar liquids had no effect on the compaction stress. The compaction behavior of jute woven fabrics was studied in detail in a previous publication[24] where the effect of the final fiber volume fraction and compaction speed on the maximum compaction pressure, permanent deformation and stress relaxation of the preforms was analyzed. The aim of this work is to extend the knowledge on the issues that may arise in the manufacturing process of natural fiber composites. The characterization of the deformation response of natural fiber preforms to compressive forces was carried out. The effect of fluid absorption on the compaction behavior of jute fabrics and sisal fiber mats was analyzed by changing the immersion time of the preforms in the test fluid. The results were compared to those obtained with synthetic preforms (glass fiber mats).

Experimental procedure

Reinforcement materials

Two different natural fiber fabrics were used in this study, bidirectional woven jute fabric (Castanhal Textil, Brasil; surface density = 0.0300 g/cm^2) and sisal random mat (surface density = 0.085 g/cm^2). The fabrics were washed with a 2% v/v distilled water and detergent (15% active matter) solution, to remove contaminants and normalize the conditions of the fabrics for all the tests. Then, the fabrics were dried under vacuum at 60°C for 24 hours. In addition, glass fiber random mat (surface density = 0.047 g/cm^2) was used for comparison purposes. 130 mmdiameter circular samples of the preforms were compressed in the compaction tests. Glass and jute preforms consisted of six layers of fabric while sisal preforms consisted of two layers of fabric. Pictures of the reinforcements used in this study are shown in Figure 1.

Compaction mechanisms observation – micro-structural analysis

Natural fiber structure was analyzed by scanning electron microscopy (SEM, model JEOL JSM 6460 LV). Composite plates of jute woven fabrics and unsaturated polyester resin were manufactured by the compression

Figure 1. Reinforcements used in the compaction tests. a) Jute woven fabric, b) sisal fiber mat, c) glass fiber mat.

molding technique. Fiber volume fractions of 0.24, 0.30, 0.37 and 0.42 were achieved by stacking 4, 5, 6 and 7 layers of reinforcement, respectively, while keeping the mold cavity thickness fixed (0.315 cm). The composite plates were sectioned and polished, and the preform microstructure was observed using an optical microscope (Olympus PMG 3).

Compaction experiments

An Instron Universal Testing Machine with a 30 kN load cell was used to carry out the compaction tests. Preform samples were compressed at a constant compaction rate (5 mm/min), until the desired thickness or fiber volume fraction was achieved. Jute and glass preforms were compacted to a fiber volume fraction of 0.55 while sisal preforms were compressed to a volume fraction of 0.35, due to the excessive clamping forces required to achieve higher fiber contents. The machine was programmed to hold the final thickness for ten minutes while the load was recorded in order to analyze the preform stress relaxation. After the relaxation step, the test was repeated. Thus, two loading cycles were obtained. Figure 2 shows a sketch of the experimental set up.

Experiments were conducted in dry and wet conditions. In the wet condition, in order to evaluate the effect of the immersion time on the compaction response of the preforms, compaction tests of samples that were immersed for different periods of time in the test fluid were also performed. A 20% v/v water/glycerin solution was chosen as the test fluid because of its viscosity (0.130 Pa.s) is similar to commercial infusion resin viscosities. The viscosity was measured by means of a Brookfield DV–II + cone and plate viscometer (precision ± 0.0025 Pa.s).

Permeability tests

Unidirectional injection experiments were performed in a rectangular steel mold (500 mm × 100 mm) with an acrylic lid. The depth of the mold cavity used for each injection was set using steel frames of different

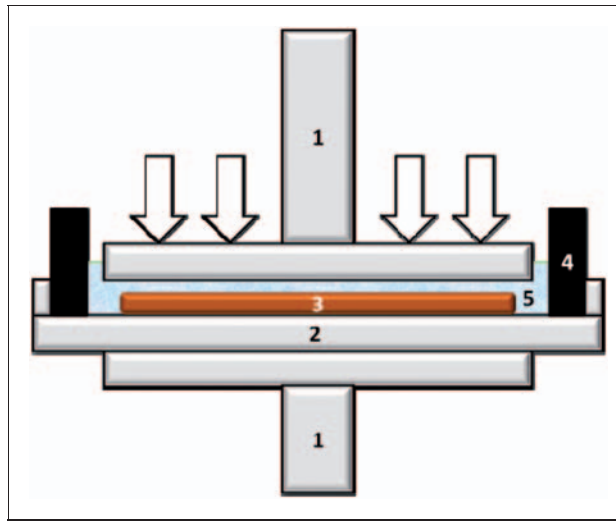

Figure 2. Compaction tests set up. 1) INSTRON compaction plates, 2) round aluminum container, 3) circular preform, 4) rubber seals, 5) test fluid (if used).

thicknesses placed between the mold and the lid in order to obtain the desired values of porosity. In order to avoid mold deflection during the infiltration tests, a 3 cmin thickness lid was used. The viscosity of the fluid used was measured before every infusion in a Brookfield DV–II + cone and plate viscometer (precision ± 0.0025 Pa.s). A vacuum pump was used to force the fluid flow through the mold cavity. The pressure gradient achieved was measured with a vacuum gauge (precision ± 1 kPa), located at the outlet line of the mold. Two injections were conducted for each porosity and type of fabric.

Data analysis

Permeability tests

In this study, Darcy's Law for unidirectional flow was used to estimate the permeability. Saturated permeability (measured after the reinforcement is fully saturated with fluid) was calculated by measuring the fluid volumetric flow rate with a standard flow meter connected at the output line of the mold (equation 1).

$$K_{sat} = \frac{Q \cdot \mu \cdot \Delta L}{A \Delta P} \quad (1)$$

In equation 1, K_{sat} is the saturated permeability (m^2), Q is the volumetric flow rate (m^3/s), ΔL the preform length (m), μ is the fluid viscosity (Pa.s), ΔP is the applied pressure gradient (Pa), and A is the mold cavity transverse area (m^2). The modified Carman–Kozeny model (equation 2) was used to establish a relationship between permeability and porosity. This relationship is a modification of the Carman–Kozeny model that 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.[25] The modification of the original model was done because for many types of preforms the assumptions behind the Carman–Kozeny model were not justified,[26] and the equation was not able to properly fit experimental values of permeability.

$$K = \frac{(1 - V_f)^{n+1}}{c(V_f)^n} \quad (2)$$

In equation 2, V_f is the fiber volume fraction and n and C are empirical parameters. The use of n exponent other than 2, is not based on a flow mechanism and the model can be taken as an empirical model which fitted the experimental data.[27,28] The relationship between the permeability and the fiber content was also fitted with a three parameter exponential model, as proposed by other authors.[29] It was found that both models fitted accurately the curves, but the Carman–Kozeny model results were reported in this work because it is the most convenient as it has only two fitting parameters.

Compaction tests

The compaction pressure (MPa) was calculated using equation 3, where L is the load (N) acquired by the load cell of the testing machine and A is the sample area (mm^2). The fiber volume fraction was obtained with equation 4, where n is the number of reinforcement layers stacked in the preform, ζ is the surface density (g/cm^2), ρ is the fiber density (g/cm^3) and t is the preform thickness (cm) calculated as the initial thickness minus the testing machine cross-head displacement.

$$Compaction\ Pressure = \frac{L}{A} \quad (3)$$

$$Fiber\ volume\ fraction = \frac{n \cdot \zeta}{\rho \cdot t} \quad (4)$$

In order to calculate the permanent deformation suffered by the preform, the unstressed preform thickness was defined as the thickness measured by the testing machine when the compaction pressure reached 7 kPa, which is a very low compaction pressure.[30] Therefore the permanent deformation suffered by the preform was calculated using equation 5,

$$Permanent\ Deformation\ (\%) = \frac{t_0 - t_1}{t_0} \cdot 100 \quad (5)$$

where t_0 is the unstressed preform thickness in the initial loading cycle (cm), and t_i is the unstressed preform thickness in the subsequent loading cycles (cm).

The stress relaxation of the preforms was measured during ten minutes after the final fiber volume fraction was achieved. In order to compare the results for the different testing conditions, a normalized compaction pressure was calculated with equation 6, and plotted against time. The amount of stress relaxation can be seen directly in the plots, or calculated with equation 7.

$$Normalized\ Compaction\ Pressure = \frac{Instantaneous\ Compaction\ Pressure}{Maximum\ Compaction\ Pressure} \quad (6)$$

$$Stress\ Relaxation\ (\%) = (1 - Normalized\ Compaction\ Pressure) \times 100 \quad (7)$$

Results and discussion

Permeability of the reinforcements

Fabric permeability is the key parameter that governs the flow in the fiber bed, together with the fluid viscosity. The pressure gradient needed to fill a mold that contains a dry preform increases with the viscosity of the fluid, and when the permeability of the reinforcement decreases. Preform permeability depends on the fabric architecture, fiber type and the porosity (1 – fiber volume fraction). Rising the fiber content (decreasing the porosity) the amount of open flow paths are reduced. Although the main objective of the present work is to characterize the compaction response of natural fiber fabrics, it was necessary to calculate their permeability.

When the compaction tests are performed in wet condition, the compaction pressure developed depends on the mechanisms that contribute to dry compaction of the preforms (yarn cross-section deformation, yarn flattening, yarn bending deformation, void/gap condensation and nesting) but also on the pressure needed to force the fluid to flow through the reinforcement, as the compression plate of the testing machine moves down. More permeable preforms and lower fluid viscosities lead to lower compaction pressures developed due to

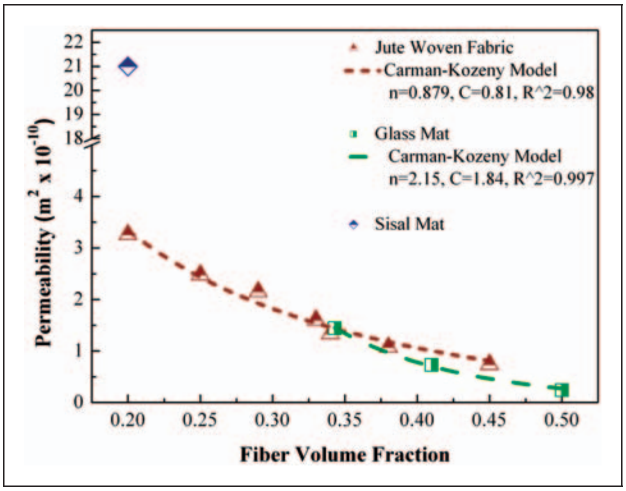

Figure 3. Saturated permeability vs. fiber volume fraction for jute woven fabrics and glass mats. The model fitting parameters are shown.

fluid flow through the reinforcement, as shown in Equation 8,

$$p = \frac{\mu Q}{2\pi h K} \ln \frac{r_0}{r_i} \tag{8}$$

where K is the isotropic permeability, h is the cavity thickness, Q is the constant flow rate, μ is the fluid viscosity, and r_0 and r_i are the outer and inner radii of the sample respectively.[6]

On the other hand, a lubrication effect is expected to take place when preforms are compressed in the presence of a liquid, which facilitates fiber movement and rearrangement during the compaction process. This effect would lead to a decrease in the pressure needed to compress the preforms to a certain thickness, thus it would be easier to achieve the desired fiber volume fraction in wetted preforms than in dry preforms. Therefore a compromise exists between the lubrication effect and the pressure build-up due to the fluid flow through the reinforcement.

Because the wet compaction tests were performed on samples saturated with the test fluid, saturated permeability data should be used to estimate the increase in the pressure due to the fluid flow through the preform. Figure 3 shows the relationship between the saturated permeability (according to equation 8 is inversely proportional to the pressure developed in the mold) and the fiber volume fraction for glass random mats and jute woven fabric.

The permeability of sisal mats was calculated for a single porosity value of 80%, because of the high clamping forces required to achieve larger fiber contents. It was found that permeability of sisal mats was 2.1 $E^{-9}\,m^2$, which is five times higher than jute fabric permeability for the same porosity. The permeability of sisal mats was measured by other authors in previous publications, and different results were found. Rodriguez et al.[28] reported a permeability value of 1.5 $E^{-6}\,m^2$ and a similar value for jute woven fabrics for a porosity of 80%, which is three orders of magnitude higher than the values obtained in this work. Li et al.[31] reported a permeability value of sisal mats of about 6 $E^{-9}\,m^2$ for the same porosity, which is three times higher than the result obtained in the present work. These discrepancies could be attributed to experimental issues that could modify the measured permeability values, such as measurement artifacts errors, textile non-uniformity, preform structure, mold deflection, fiber channeling, air bubbles and fiber treatments.[31,32] In addition, those authors reported the unsaturated permeability value for that reinforcement, and therefore the capillary effects developed during the infiltration of the preform[33,34] could be responsible for the large difference between their results and the ones reported in this work that correspond to the saturated permeability values.

Sisal and glass fiber mats are fairly isotropic, due to the randomness of the reinforcement architecture. Therefore, in these cases the permeability measured with the unidirectional flow experiments corresponds to the isotropic permeability and can be used in equation 8 to predict the hydrodynamic pressure generated during the compression of the preforms. On the other hand, woven fabrics are anisotropic. As a consequence, the shape of the squeeze flow front is not circular but ellipsoidal and the permeability should be reported as a tensor instead of a single value.[35,36] Performing unidirectional flow tests in the 0°, 45° and 90° directions, and assuming that no flow occurs in the transversal direction of the preform, the ratio between principal in-plane permeabilities (K1, K2) could be obtained following the procedure described by Advani et al.[37] In addition, it was found that the principal directions are rotated 13° with respect to the warp and weft directions of the fabric. The ratio K1/K2 is a direct measurement of the fabric anisotropy. In the case of these jute fabrics, $K1/K2 = 1.24$, which means that the ratio between the radius of the ellipse $r1/r2 = (K1/K2)^{0.5} = 1.11$.[35] In order to calculate the hydrodynamic pressure, the effective permeability, $(K1.K2)^{0.5}$,[35] was used as the isotropic permeability in equation 8.

Micro-structural analysis

Scanning electron micrographs of sisal and jute fibers are shown in Figure 4a. It can be seen that sisal fibers are much bigger than jute fibers, and they are composed of a larger number of macro fibrils, while the lumen size and the wall thickness of the macro fibrils are similar in both

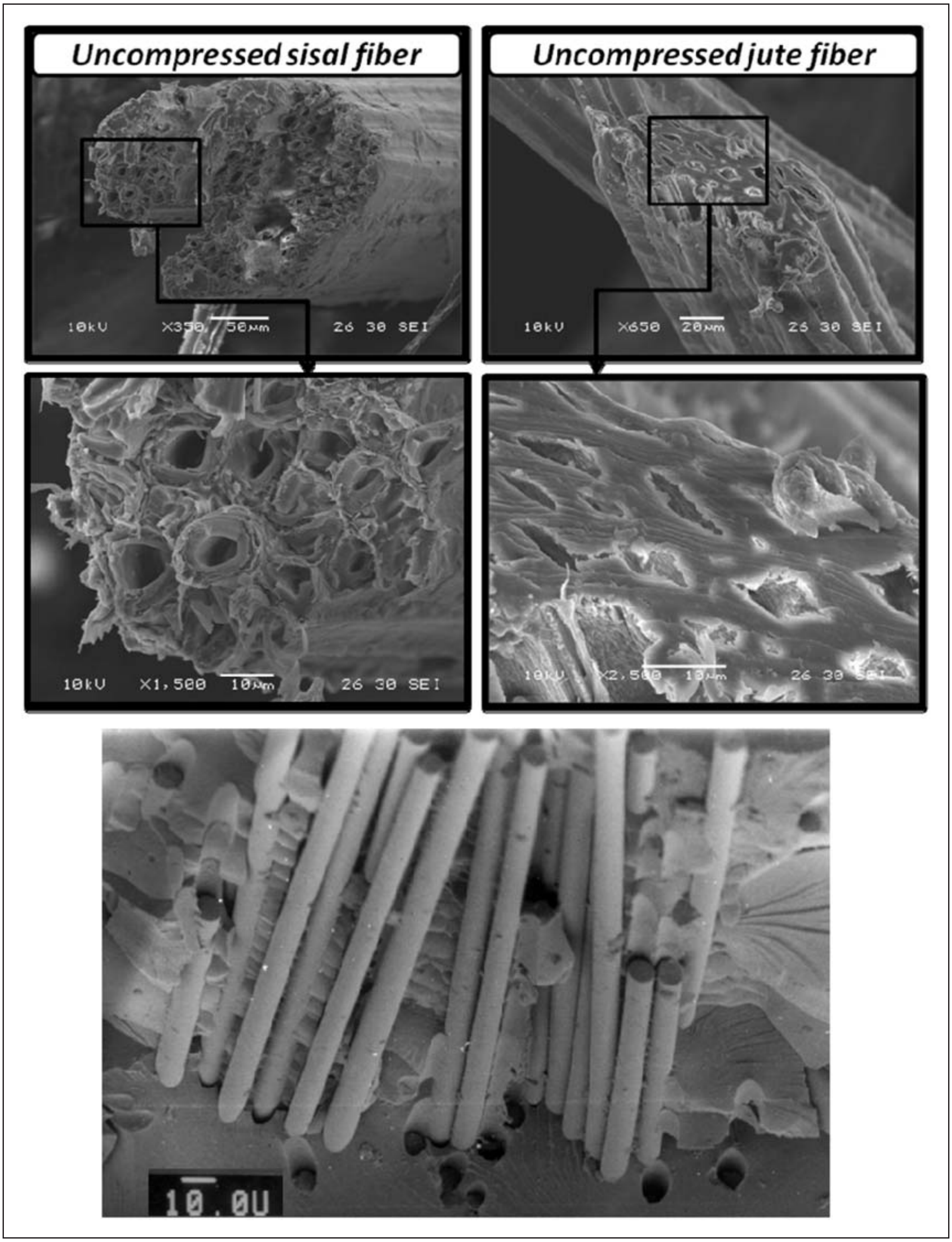

Figure 4. SEM images of sisal and jute fibers (a) and glass fibers (b).

types of fibers. Figure 4b shows SEM images of the fracture surface of a glass reinforced composite material.[38] The surface morphology of glass fibers is smooth, opposite to that found on natural fibers, which are rough and irregular.

Figure 5, shows micrographs obtained by optical microscopy at 300× of the transversal section of jute/unsaturated polyester resin composite specimens manufactured by the compression molding technique. It can be seen that natural fiber cell walls collapsed and the lumen was closed when compressive forces were applied to the preform, and this phenomenon increased as the fiber content (and transverse deformation) increased.

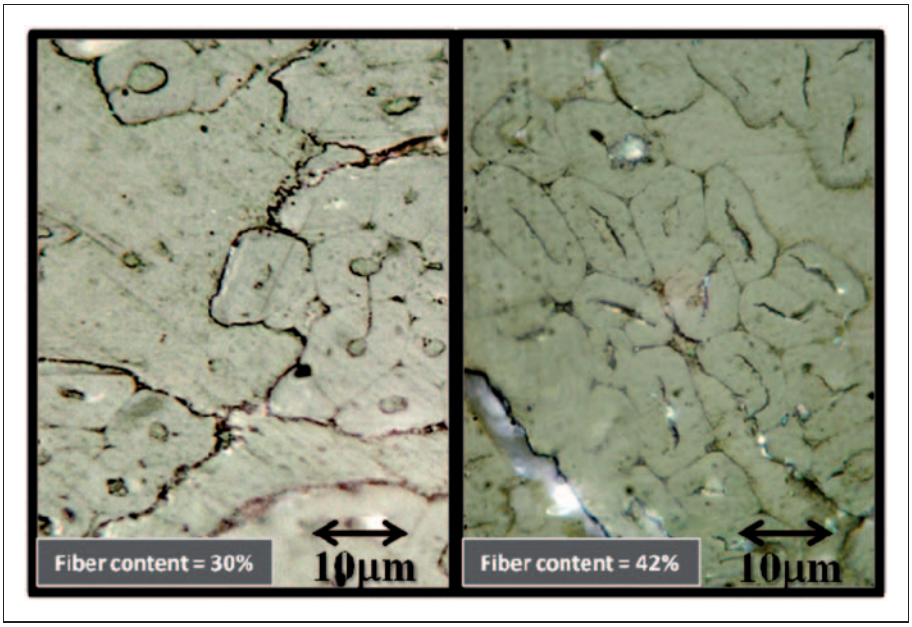

Figure 5. Optical micrographs at 300× of the cross section of jute woven fabric composites. Macro fibrils collapsed and their lumen was closed due to the high clamping forces.

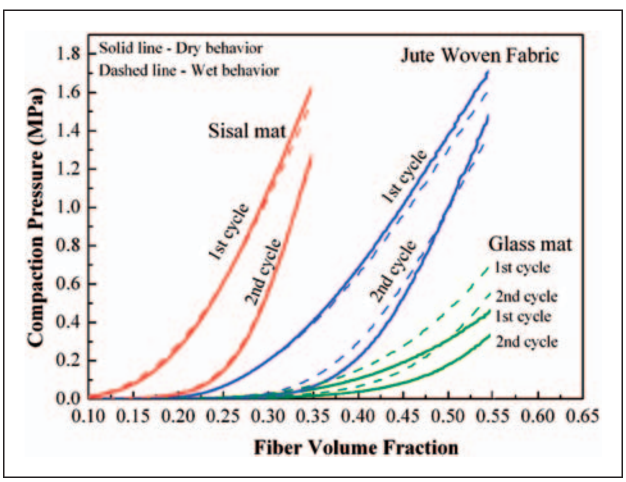

Figure 6. Compaction behavior of sisal mats, glass mats and jute woven fabrics.

Compaction behavior of jute, glass and sisal preforms

The compaction response of dry sisal and glass fiber mats as well as dry jute woven fabrics is shown with solid lines in Figure 6. It is clear that the compaction pressure needed to achieve any fiber volume fraction within the range studied was significantly lower in the case of glass fiber mats, followed by the jute woven fabrics, and then sisal fiber mats which required such a large pressure to be compressed that they could only be compacted to a fiber content of 35%, due to limitations of the mechanical testing machine. Let us compare sisal and glass mats due to their similar fabric architecture. Despite different number of fabric layers being used in the compaction tests of sisal and glass fibers, the effect of the number of layers is much more important when woven fabrics are used, due to the nesting of the 'Hills' and 'Valleys' of the adjacent layers, as demonstrated by Chen et al.[4] The effect of the number of layers in random mats is less significant and only observable at very high compaction levels. Those authors observed a maximum difference in the thickness per layer of 15% when the compaction tests were performed on one layer and 10 layers of strand mat and the pressure reached was 1.5 MPa. Therefore the enormous differences observed in the maximum compaction pressures of sisal and glass fiber mats are enough to suggest that vegetable fiber mats are harder to compress that glass fiber mats. The rough surface of these natural fibers leads to large inter-fiber friction forces, increasing the compaction pressure needed to reach high fiber contents. On the other hand, the surface of glass fibers is smooth and thus the inter-fiber friction force is much lower. The results shown in Figure 6 reveal one important issue of using natural fibers in composite materials, which is the maximum fiber content that can be achieved with the available clamping forces. If the vacuum infusion technique is used to manufacture composite parts, the maximum compaction pressure is limited to the atmospheric pressure (about 0.1 MPa). In this case, the curves show that the volumetric fiber content in the composites would be approximately 16%, 26% and 40% if sisal mats, jute fabrics and glass mats are used respectively. The low fiber content obtained in natural fiber composites leads to low mechanical properties and, what is more important, less eco-friendly composites.

In addition, the difference between successive cycles was larger in the case of natural fiber than in glass fiber preforms, suggesting that natural fiber fabrics experienced larger permanent deformation than glass fiber mats. However, jute fabric architecture is totally different from sisal and glass mat architecture, thus the compaction mechanisms responsible for the amount of permanent deformation are not the same in those cases. Therefore it is more appropriate to compare sisal with glass random mats in terms of permanent deformation, due to the similarity in the architecture of the arrangement of the fibers. Using equation 5, it was found that the permanent deformation was 42% for sisal and 18% for glass random mats. Moreover, by increasing the number of layers, the permanent deformation of preforms should increase, because mechanisms of permanent deformation that result from the interaction between adjoining layers increase. However, opposite results were found in this work, where the permanent deformation experienced by two layers of sisal was more than 100% higher than that experienced by six layers of glass mats. The entire permanent deformation observed on glass mats was probably caused by some filaments from one layer of fabric that got intertwined or embedded in the adjoining layer, as proposed by Somashekar et.al.[8] On the other hand, sisal fibers suffered lumen collapse just as jute fibers did (Figure 5). When natural fiber fabrics are used, breakage of elementary fibers and lumen collapse are two important mechanisms responsible for the permanent deformation of preforms. Furthermore, the strength of natural fibers is lower than that of glass fibers, and the compaction pressures applied to reach certain fiber content is higher when compressing natural fiber fabrics. Therefore, fiber breakage is more likely to happen when natural fibers are compressed. In addition, the same mechanisms present in glass mats occurred with sisal mats. Consequently the permanent deformation experienced by sisal mats was significantly larger than that experienced by glass fiber mats.

The significant permanent deformation observed in natural fiber reinforcements can be used to increase the fiber volume fraction of the composites. Let us consider the vacuum infusion technique, where the maximum fiber content achievable is 16% for sisal mats and 26% for jute fabrics: if a compaction step identical to the first compaction cycle shown in Figure 7 is performed prior to placing the reinforcements in the mold, the fiber content obtained would increase up to 23% and 37% for sisal mats and jute fabrics respectively. Those values correspond to the fiber content obtained at the second loading cycle for a compaction pressure of 0.1 MPa(1 atm).

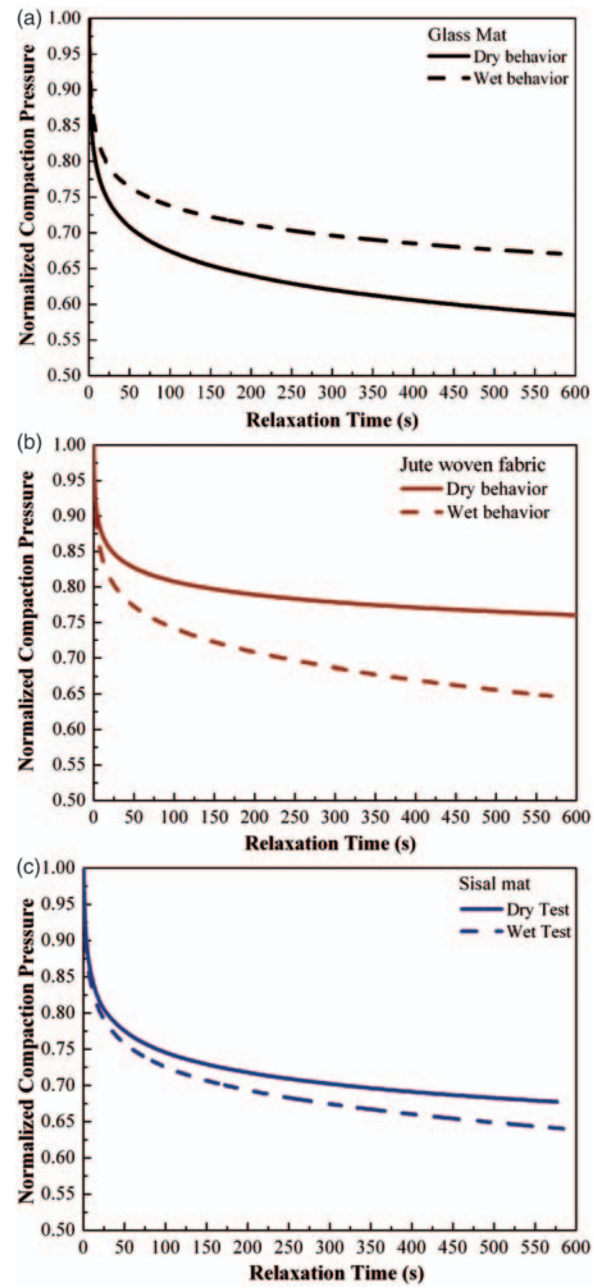

Figure 7. Stress relaxation of glass mats (a), jute woven fabrics (b) and sisal mats (c). Solid lines correspond to dry compaction test while dashed lines correspond to wet tests.

The compaction response of the reinforcements impregnated with the test fluid is shown with dashed lines in Figure 6. Apparently, the pressure caused by the fluid flow through the preform is more significant than the lubrication effect in glass fiber mats, while jute woven fabrics and sisal mats behaved in the opposite manner. However, when the compaction tests are performed on preforms which had been saturated with fluid, the pressure build-up due to the fluid flow through the reinforcement should be taken into account. In this case, the total stress applied to the saturated preform, σ_{total}, can be decomposed according to Terzaghi's law:[7]

$$\sigma_{total} = \sigma_{fiber} + p \tag{9}$$

where σ_{fiber} is the stress taken up by the fibers and p is the fluid pressure induced by squeezing flow. The latter contribution to the overall compaction pressure depends on the preform permeability, fluid viscosity and the compaction speed. An analytical expression suitable to estimate the maximum pressure build-up due to the fluid flow though the preform was developed by Bickerton et al.[39] They considered that during the compaction the resin flow is described by Darcy's Law, all resin velocities remain in-plane, and the entire preform is fully saturated with resin. In addition, they assumed that the resin or test fluid was Newtonian. The mathematical expressions derived by Bickerton et al.[39] to determine the magnitude of the hydrodynamic force developed in the wet tests was modified in a previous publication[24] according to the preform geometry used in our experiments. The resulting expression is shown in equation 10, where μ is the fluid viscosity, $\dot{h}$ is the compaction speed, K is the preform permeability, h is the cavity thickness, and r is the radius of the preform. Therefore, the maximum pressure induced by squeezing flow was obtained by dividing the fluid force by the sample area, as shown in equation 11.

$$F_{fluid} = \frac{\pi \mu \dot{h}}{4kh}\left(-\frac{r^4}{2}\right) \tag{10}$$

$$p = \frac{F_{fluid}}{\pi(r^2)} \tag{11}$$

Table 1 presents the maximum compaction pressure obtained in the dry and wet tests, the fluid pressure calculated using equation 11, and the compaction pressure obtained as the difference between the wet compaction pressure and the fluid pressure. This compaction pressure corresponds to the stress taken up by the fibers (σ_{fiber}) during the wet tests. Results showed that the maximum compaction pressure decreased 3% due to the lubrication effect when the glass fiber preforms were compressed with the test fluid. On the other hand, natural fiber reinforcements showed a decrease in compaction pressure of 10% on jute and 4% on sisal fiber preforms, respectively. In this case, besides the lubrication effect, fluid absorption and fiber softening could have occurred during the compaction tests decreasing the compaction pressure.

Figure 7 shows the stress relaxation of the different reinforcements studied. It is clear that the relaxation process is more significant in glass random mats (42% after 10 minutes), followed by sisal random mats (32%) and then jute woven fabrics (24%). Once again, due to the differences in the fabric architecture, only sisal and glass random mats results could be compared. The results obtained in the wet tests showed an opposite trend for synthetic fiber than for natural fiber preforms. Glass fiber random mats experienced larger stress relaxation when compressed dry than when compressed in the presence of the test fluid, while sisal and jute fiber preforms responded in the opposite manner. The lubrication effect was expected to decrease the amount of stress relaxation, because both fiber and tow realignment and reorientation, as well as the interactions between consecutive layers should have been easier during the compaction stage. However, natural fabrics showed an opposite trend. Therefore the compaction load could have dropped due to fiber softening instead of relaxation processes, decreasing the compaction load during the relaxation step.

Influence of the test fluid on the compaction response – effect of the immersion time

Figure 8 shows the compaction response during the initial loading cycle of glass fiber mat (8a), sisal fiber mat (8b) and jute woven fabric (8c) preforms which were immersed in the test fluid for different time periods before performing the compaction test. Figure 9 shows the ratio between the maximum compaction pressure taken up by the fibers at the wet tests and the dry maximum compaction pressure as a function of the immersion time for all the reinforcements used in this work.

Table 1. Maximum compaction pressures, maximum pressure induced by squeezing flow and corrected wet compaction pressure

	Maximum compaction pressure – dry- (MPa)	Maximum compaction pressure – wet- (MPa)	Fluid pressure caused by Darcy effect 'p' (MPa)	Stress taken up by the fibers, 'σ_{fiber}' (MPa)	Decrease in compaction pressure (%) in the wet compaction tests
Jute	1.73	1.65	0.097	1.55	10
Sisal	1.63	1.57	0.01	1.56	4
Glass	0.47	0.71	0.26	0.455	3

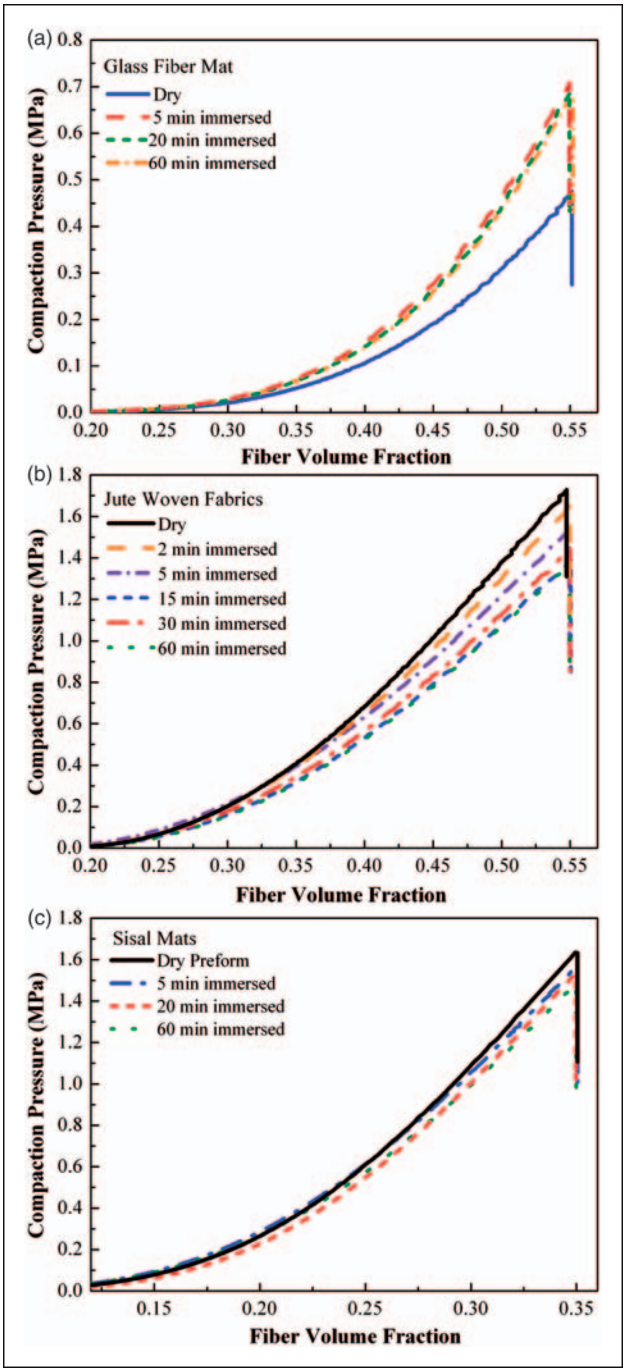

Figure 8. Compaction response and maximum compaction pressures reached in preforms immersed in the test fluid different time periods before performing the compaction test. (a) Glass mats, (b) jute woven fabrics, (c) sisal mats.

Glass fiber mats

The increase of the pressure due to the fluid flow through the preform was more important than the lubrication effect. This can be seen in Figure 8a, where the slope of saturated preform curves was higher than the slope of the dry preform curve among the fiber

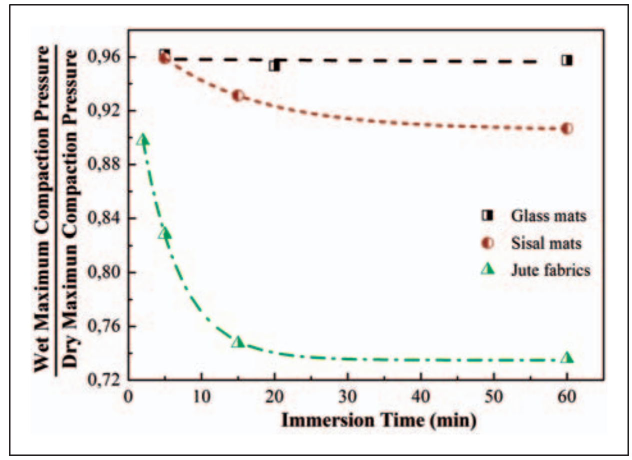

Figure 9. Effect of immersion time on the compaction behavior.

volume fraction range studied. However, if the fluid pressure developed due to the Darcy flow is subtracted from the compaction pressure of the wet tests, the resulting flow-independent compaction pressure was lower than the dry compaction pressure, as expected. It can be seen in Figure 9 that the lubrication effect decreased the compaction pressure by a 3%. In addition, the compaction behavior of glass fiber mat preforms did not change as the immersion time in the test fluid increased, which means that the fibers did not suffer fluid absorption and softening, as expected.

Sisal fiber mat and jute woven fabric

Sisal mats and jute woven fabrics showed a similar behavior, despite of the different fabric architecture. It was found that jute preforms soften as the immersion time increased up to 15 minutes. After 15 minutes of immersion, the compaction behavior reached a steady state, and it was difficult to distinguish any significant differences among the samples. The total pressure drop observed was around 27%, and was caused by the lubrication effect and the fiber softening. The exact contribution of each effect is difficult to calculate because both happen simultaneously. However, if the pressure drop at immersion times of 2 and 60 minutes are compared, it can be seen that the fiber softening was responsible for a pressure drop of at least 17%. This pressure drop is consistent with the results obtained in the relaxation step. Apparently, 17% of the stress relaxation observed previously was caused by fiber softening, which explains why the stress relaxation observed on impregnated jute preforms was larger than that of dry preforms. Sisal preforms also softened as the immersion time increased. Apparently, the fluid absorption was slower, and the steady state was just reached after 60 minutes of immersion, showing a total pressure dropped of 10%. Once again, comparing the results at 2 and 60 minutes of immersion, the fiber softening contribution (6%) could be estimated. Within 15 minutes of immersion, which is the time needed to perform the compaction test and the relaxation step, the pressure decreased by 4%. This could explain why the stress relaxation curves for the dry and wet preforms shown in Figure 7 did not show a difference as large as jute fiber preforms.

Final remarks

Natural fibers soften as they absorb fluid, and therefore the compaction response of the fabrics is dependent on the type of fluid used during the compaction stage. A 20% v/v water/glycerin solution was used in the experiments performed in this work, which is a very polar fluid. Consequently, fluid absorption and fiber softening were considerable. If non-polar fluids are used, fibers will absorb less amount of fluid and the softening effect will not be significant. However, it was found in previous investigations[23] that jute fibers absorbed a small extent (6%) of vinylester resin and swelled as a consequence. Therefore, if natural fiber fabrics are used in LCM techniques, the compaction behavior of the fibers should be studied for each type of resin used. Moreover, it should be taken into account that the fiber compaction response during the processing of the composites will not match the compaction response observed in the experiments performed in the laboratory using a test fluid different from the resin.

Summary and conclusions

In addition to the well known drawbacks of vegetal fibers such as water absorption and low thermal stability, it was found in the present work that they require higher clamping forces to achieve large fiber contents than synthetic fiber preforms. This issue is very important if the composites will be manufactured by vacuum bagging techniques.

Regarding the compaction behavior of preforms made from these natural fibers, the following conclusions could be made:

- Structure of vegetal fibers affected the compaction response of the preform. Lumen closure increased the permanent deformation of natural fiber preforms after loading cycles.
- Fluid absorption of vegetal fibers caused fiber softening decreasing the compaction pressures. The fluid absorption increased as the immersion time was raised, thus some of the pressure drop due to fiber softening happened at the relaxation stage. As a consequence, the stress relaxation of impregnated performs was higher than dry performs. Glass fiber mat performs showed the opposite trend, because the lubrication effect increased fiber and tow realignment, and reorientation, as well as the interactions between consecutive layers during the compaction stage.
- The nature of the fluid affects the compaction behavior of natural fiber preforms significantly. Therefore the clamping forces required during the LCM processing of natural fiber based composites and thermosetting polymer matrix may differ from the compaction forces measured in laboratory experiments using a different fluid.

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