1 Introduction

Liquid Composite Molding (LCM) and especially Resin Transfer Molding (RTM) are increasingly used manufacturing processes for producing high quality and complex composite structural parts. Moreover, RTM process has recently gained great interest due to its ability of producing thermoplastic structural parts, which could open up new opportunities to many domains requiring high performance and recyclable materials, such as automotive, aerospace or aeronautics for example.

A typical LCM process can be divided into several steps. The dry fiber reinforcement called preform is cut and placed into the mold cavity. The mold halves are then clamped. The liquid resin is subsequently injected and flows through the fibrous network. During injection, the resin evacuates air out of the mold cavity and impregnates the preform before the consolidation phase occurs. In that stage, dry spot as well as voids might be created. Voids can also form due to partial volatilization of gases during curing. However, the leading cause of void formation in LCM is mechanical entrapment of air during the resin injection step. Hence, we will focus only on flow-induced void formation in this work.

The quality of LCM products and the efficiency of the process depend strongly on the impregnation of fiber preform during the mold-filling stage. It has been indeed shown [1] that porosities drastically affect mechanical performances, such as interlaminar shear strength, flexural strength and compressive strength. They also have a detrimental effect on crack initiation, fatigue life and on moisture absorption. In this context, reliability and security criteria have led aeronautics and automotive industries to require a strict determination of the residual void fraction after manufacture, and to reject final parts containing more than 2% of void [2].

Several investigators [2–5] have conducted theoretical analysis of void formation and void transport during impregnation through a fibrous media. Researchers have been found that the dual-scale architecture of woven reinforcement is mainly responsible for air entrapment [2-3]. The fabrics that are commonly used consist of tows which are woven together. These tows themselves are made of several thousands of fibers. Therefore, they generally present two kinds of porosities with significantly different pore sizes: micro-porosity, which is located within the tows, and macro-porosity, which is located between the tows. These double-scale porous media leads to a competition between the viscous flow in the macropore and the capillary wicking in the micropore (figure 1). It was found that for low fluid velocities, capillary forces become dominant inducing the fluid to flow through the fabric tows, where the porosity is smaller and the capillary pressure is higher. As a result, air is entrapped in the inter-tow area. However, when the resin velocity increases, the viscous forces prevail over the capillary ones, making the resin surround fiber tows. As a consequence, bubbles are created within tows.

Once air has been entrapped nearby the resin front, the bubbles can vary in size and position with time, according many scenarios, as explained in [2], leading to the formation of a partially saturated zone behind the flow front.
Numerous experimental studies from several authors have been carried out in order to have an understanding of the mechanisms of void formation. Bascom and Romans [1] were the first to confirm the presence of micro-voids by microscopic observation of composite samples, and to show their effect on material strength. Many researchers [3], [4], [6-7] quantified a posteriori the void content in a fibrous preform, and showed that it might be correlated with a dimensionless parameter called capillary number $Ca$, which reflects the relative effect of viscous forces and surface tension.

$$Ca = \frac{\mu \bar{u}}{\gamma}$$  \hspace{1cm} (1) 

where $\mu$ is the dynamic resin viscosity, $\bar{u}$ is the global resin velocity, and $\gamma$ its surface tension. By plotting the void content versus $Ca$, results draw a characteristic V-shaped curve, so that one can determine an optimal $Ca$ (or impregnation velocity) for which the amount of created voids is minimal. Lebel et al. [8] recently proposed a model based on the fibrous reinforcement properties to predict this optimal value.

On the other hand, research oriented towards quantifying saturation during the filling is quite limited. Even if several authors have contributed to a better understanding and modeling of the mechanisms of formation and transport of voids during injection, very few experimental approaches allowed a direct measurement of the dynamic saturation curve. Ruiz et al. [9] obtained quite interesting results using a non-intrusive monitoring technique based on a 2D optical tomography reconstruction (VLT method). This setup allows the determination of the partial saturation of the preform by measuring the filtered visible light transmitted. Nordlund et al. [10] used a similar methodology also based on optical observations. They conducted an injection at constant flow rate in a transparent mould. Video acquisitions are thereafter transformed into saturation level by image analysis. This approach was validated by a microscopy analysis and coupled to a multiphase flow model. We may also mention Seto [11] who performed a similar technique based on image analysis for an anisotropic woven fabric. Labat [12] developed a homemade conductivity sensor made of two brass electrodes in order to measure the electrical conductance of the fibrous media. Several injections of a conductive model fluid in an insulating preform made of glass fiber were carried out at constant pressure and flow rate, allowing the determination of the evolution of saturation as a function of time and position. Later, Guéroult [13-14] continued this work by using permittivity sensors whose response depends on dielectric properties of the fibrous media and of the liquid. Theses sensors were able to perform for a wide range of liquids and preforms. This study presents an original approach in which heat transfers are used to detect the saturation profile. Heat conduction is indeed sensitive to porosities, therefore by the degree of filling of the preform. Therefore, the idea is to develop an experimental bench allowing the dissipation of a heat flux within the fibrous preform. Thereafter, heat transfer will be exploited in order to identify the saturation curve during the impregnation of the studied dual-scale fibrous media.

2 Experimental bench

2.1 General description of the bench

An experimental bench [13] initially devoted to permeability measurements was modified (a picture of the bench can be seen in figure 2 and a cross-section in figure 3). This apparatus allows the unidirectional injection of a model fluid into a textile preform. The position of the lower part of the mold (made of steel) can be adjusted vertically very precisely thanks to several dial indicators, in order to obtain the desired thickness of the molding cavity, and therefore, the desired fiber volume fraction. The upper part of the mold, made of glass, allows a direct visual control during the impregnation of the reinforcement. The preform has a size of 400 mm x 100 mm, and a thickness of 2.74 mm. A compressible silicone tape is surrounding the preform in order to ensure the sealing and to prevent racetrackings. A dosing syringe containing the liquid mounted on a tensile testing system allows an accurate control of the fluid flow rate. The maximum allowable pressure in the mold is $3.10^5$ Pa.
Thin transparent heaters made of optical grade polyester are glued to the underside of the glass plate, that keep an excellent view of the injection in progress. They are able to dissipate a uniform heat flux density which is quantified accurately. A heat exchanger fixed under the steel plate absorbs the heat released. Three heat flux sensors, mentioned as HFS1 to HFS3, are integrated in the bottom steel plate at positions 85, 210, and 335 mm from the beginning of the preform. They record the thermal response of the porous material through the transverse wall heat flux (z-axis). Furthermore, several K-type thermocouples are placed at various locations of the bench. This setup is also equipped for the saturation measurement by a conductometric method, which is detailed in [13]. The sensor is made of two rectangular brass electrodes (100x6x0.05 mm), placed facing each other on both sides of the preform. By applying a periodic voltage between them, the conductance of the medium is derived from the measurement of the voltage at the terminals of a shunt resistor. Temperatures and voltages are recorded using Yokogawa™ DL750 and DAS™ 1400 acquisition systems with an acquisition frequency of 20 Hz.

2.2 Experimental protocol
All experiments were performed by injecting a liquid through the length of glass fabrics at constant flow rate. The fluid has been selected so that its properties (viscosity, surface tension and contact angle) are close to those of a classical resin used in RTM process. It is a mixture between glycerol (87 %,), water (12.6 %,), surfactant (0.3 %,), and a small amount of potassium chloride (0.1 %,), in order to enhance the electrical conductivity. The reinforcement is a stitched unidirectional fiberglass cloth (2K filaments/tow), and the injection is made along the fibers (x-axis). A particular care is taken for cutting and placement of reinforcements in the mold. To ensure that the stitches which are oriented in the direction perpendicular to the flow do not overlap, their positions are alternated between each ply. All properties are summed up in Table 1.

The injection of fluid is performed at constant flow rate (~2.1 mL/min) and is isothermal. The heaters are turned on as soon as the liquid penetrates into the preform, and power is kept constant during the whole duration of the injection. The intensity of this heat source is chosen so as to avoid a temperature rise of more than 10 °C in the preform.

3 Modeling
3.1 Heat transfer modeling
The imbibition of the preform by the liquid changes significantly the thermal properties of the porous medium, going from a completely unsaturated state (S=0), to a state of maximum saturation (S=1). Heat transfer in the porous medium is modeled in a simplified manner by means of the convection-diffusion equation (Eq. 2) and solved with FEM using Comsol Multiphysics™ software. Three-dimensional modeling is performed to account of the lateral heat losses. More specifically, saturation is taken into account in this model through the evolution of thermophysical properties $\rho, C_p$ and $\lambda$ of the porous media, as described in the next subsection.

$$
\left(\frac{\rho C_p(S(x,t))}{\partial t} + \left(\frac{\rho C_p(S(x,t))}{\partial t}\right)\vec{u} \cdot \nabla T \right) = \vec{v} \cdot \left(\lambda(S(x,t)) \nabla^2 T\right) \tag{2}
$$

The interface is assumed to be planar, and the front velocity (noted $\vec{u}$ in the above equation) to be constant and equal to the average interstitial velocity, or Darcy velocity.

$$
\vec{u} = \frac{Q}{A \left(1 - \phi_f^\gamma\right)} \tag{3}
$$

where $Q$ is the volumetric flow rate, $A$ the section of the molding cavity, and $\phi_f^\gamma$ the fiber volume fraction.

The thermophysical properties of the porous media depend on the saturation value at the abscissa considered. Many researchers showed, either by experimental measurements [9,10,12,14] or modeling [2,10,15], that the saturation of a fibrous preform by a liquid was described by a characteristic curve such as the one depicted in figure 4. This latter is modeled using several constant geometric parameters: the length of the unsaturated front ($l$), the value of saturation at the inflection point ($y$), and the maximal saturation at the beginning of the
preform \((S_{\text{mm}})\). Guéroult et al. [13-14] observed that this curve followed a simple translation in the case of an injection at constant flow rate. The average interstitial velocity being constant, the ratio between viscous and capillary forces remains indeed constant, which is not the case in a constant-pressure injection. \(x_j\) is the only time-dependent parameter which allows to translate this curve to complete the filling.

A sensitivity study has been carried out firstly to confirm the sensitivity of each parameter of the saturation curve on the transverse heat flux, as shown in figure 5. The reduced sensitivity \(X_w\) of the variable \(Z\) to a parameter \(w\) is expressed by

\[
X_w = w \frac{\partial Z}{\partial w} \quad (4)
\]

The graphs 5a, 5b, and 5c show that the transverse heat flux is sensitive to all of these parameters for the three sensors. These results highlight the ability of using heat fluxes to detect the saturation, provided that material properties are accurately characterized.

3.2 Characterization of thermo-physical properties

Effective thermophysical properties of the dry and fully-saturated porous medium in transverse and longitudinal directions have been measured by several methods (see Table 2), and their results have been then cross-checked and compared with good accuracy.

In order to describe these properties between the lower and upper values (namely when the saturation evolves between 0 and 1), their evolutions have been modeled. The sensitivity analysis shows that the transverse heat flux is almost not affected by the longitudinal conductivity \(\lambda_x\) (fig. 5d). Therefore, a straightforward law of mixtures for the longitudinal conductivity as well as for the volumetric heat capacity \((\rho C_p)\) has been used (eq. 5 & 6). In the following, the notation \(S_T\) for the saturation (or total saturation) is introduced.

\[
\lambda_x(S_T) = (\lambda_x)_{S_T=0} + S_T \left[ (\lambda_x)_{S_T=1} - (\lambda_x)_{S_T=0} \right] \quad (5)
\]

\[
(\rho C_p)(S_T) = (\rho C_p)_{S_T=0} + S_T \left[ (\rho C_p)_{S_T=1} - (\rho C_p)_{S_T=0} \right] \quad (6)
\]

However, the conductivity being an intensive property, a particular attention has been paid for the modeling of the transverse thermal conductivity.

3.3 Transverse thermal conductivity modeling using a homogenization methodology

In order to describe the thermal behavior of the porous medium during the injection, one can distinguish two types of voids:

- the micro-voids (~10-50 \(\mu m\)), which appear inside tows, and
- the macro-voids (~0.5-1.0 \(mm\)), which appear between each tow.

Hereinafter, two different saturations are considered:

- the micro-saturation \(S_\mu\) defined as the tows degree of filling (figure 6).
- the macro-saturation \(S_M\) defined as the inter-tows degree of filling (figure 7).

These multi-scale saturations are linked to the total saturation by a linear combination involving geometric parameters related to the reinforcement, according

\[
S_T = \left( \frac{\phi_\mu}{1 - \phi_\mu} \right) S_\mu + \left( \frac{1 - \phi_\mu}{1 - \phi_\mu} \right) S_M \quad (9)
\]

where \(\phi_\mu\) represents the fiber volume fraction, and \(\phi_T\) the tow volume fraction in the composite.

Assuming a certain pore morphology and by means of microscopic observations of the preform, two representative elementary volumes have been built (see figures 6 and 7) with the appropriate dimensions (see fibrous preform data in Table 1). The saturation at each scale has been modeled by growing air bubbles within tows, or in the inter-tow area. Afterwards, assuming periodic boundary conditions over the edges of the unit cell, a homogenization method based on asymptotic expansion [18] has been performed sequentially at the two scales. The effective thermal transverse conductivity has been computed for all possible scenarios, that is to say for all pairs \{micro-saturation/macro-saturation\}. The input parameters
used in the homogenization procedure are listed in Table 3. Thermal conductivities of the fluids and of the glass-fiber are assumed to be isotropic. Results are plotted in figure 8.

This abacus firstly reveals a large conductivity contrast between completely-dry and saturated preform, the value going from 0.08 to 0.55 W·m⁻¹·K⁻¹, which we will benefit in the identification. Secondly, it clearly appears that for a single saturation value \( S_T \), there are multitude of potential micro / macro distributions, representing a wide scattering of thermal conductivities. If we could determine both the conductivity and saturation \( S_T \), it implies we would be able to discriminate the proportions of macro and micro voids in the composite. By defining the parameter \( \alpha \) as the ratio between micro- and macro-saturation, we can draw ‘iso-\( \alpha \)’ curves (represented as black lines in figure 8).

\[
\alpha = \frac{S_M}{S_T}
\]

(10)

In fact, the capillary number is slightly different from the optimum capillary number for this injection, which means that it remains few voids once the preform is filled. Studies that were conducted on the same reinforcement and the same fluid [13] showed that the residual void content never exceeded 3% at this flow rate, which provides us directly limit values for alpha \((0.9<\alpha<1.1)\). We observe that the conductivities are relatively insensitive to \( \alpha \), so that the extreme values represent a reasonable envelope of thermal conductivity, which will be considered in the following.

4 Identification of the saturation

4.1 Thermal identification

Preliminary experiments without any flow were performed in order to estimate the thermal boundary conditions, and to ensure the accuracy of the calibration of the sensors that was carried out beforehand. These experiments were conducted with fluid only (figure 9), or with dry preform only. Three thin heat flux sensors and three thermocouples are placed on top of the glass plate (fig. 3), and allow the determination of the heat transfer coefficient between the glass plate and the surroundings.

Besides, numerical temperatures and heat fluxes are computed by solving a 1D heat conduction problem, so as to estimate the thermal contact resistance (TCR) between the heat exchanger and the steel plate. The estimation of these parameters leads to similar results for both experiments. Table 4 sums up some of the properties and boundary conditions that are used in the following.

Experimental wall heat fluxes have been plotted in figure 10. The heaters are turned on at \( t=0 \), when the liquid impregnates the very beginning of the preform, and dissipate a heat flux density of 1600 W/m². As long as the preform remains dry, the problem reduces to a simple 1D-conduction problem, and the three heat flux curves are roughly the same and correspond to the calibration (fig. 9). However, more complex heat transfer occurs in the vicinity of the liquid front. In fact, there is non-negligible planar conductive transfer in the glass plate in the upstream direction due to heat that has been stored above the preform remained unsaturated (the dry preform being more insulating than the glass plate). This heat supply contributes to enhance the transverse heat flux, which emphasizes the passage of fluid. As a consequence, the arrival of the partially saturated zone in front of each sensor corresponds to the sharp rise of the measured heat fluxes. After the front, the advection term being negligible, heat transfer becomes quasi-stationary and transverse heat fluxes tend to constant values. The saturation curve parameters are then determined by minimizing the cost function defined as the square difference between the measured and computed wall heat flux (Eq. 11).

\[
J(S_{\text{max}}, y_i, l) = \sum_{n} \sum_{k=1}^{3} (\phi_k^n - \phi_k^l)^2
\]

(11)

where \( n \) is the number of time step, \( k \) the sensor number, \( \phi \) the experimental transverse heat flux, and \( \phi \) the computed transverse heat flux.

Numerical wall heat fluxes have been plotted in dashed lines. Heat losses between the mold and its metallic structure have been modeled using a boundary condition of the third kind on the lateral
faces of the mold. We observe a good agreement between computed and experimental heat fluxes, even if we can notice some discrepancies for time steps greater than 1300 s.

4.2 Identification by conductometry

A periodic voltage of 1V and 100Hz is applied between the electrodes, and their overlapping area is sized to avoid electrical edge effect and is placed in the mid-section of the mould to avoid flow edge-effect. The voltage at the terminals of a shunt resistor is monitored during the injection. On each electrode is fixed an 80µm diameter K-type thermocouple (its small size is supposed not to interfere with the flow). Particular care has been paid to the electrical insulation of these thermocouples as well as that of the electrodes. The relation between conductance and void fraction has been established by adding small glass beads of known diameter in a conductometric test cell, their negligible electrical conductivity acting as air bubbles. The electrical conductivity of the liquid being highly temperature dependent, the voltage signal is corrected afterwards, by interpolating the temperature profile between the two electrodes. The signal is also adjusted, taking into account the width of the sensor, which tends to average the measurement.

The saturation curve obtained through thermal analysis is finally compared with the one measured by the conductometric sensor. The comparison between these two estimated saturations is depicted in figure 11. One can note a very good agreement between both methods. The experimental results confirm that there is still residual voids near the front, defining thus an unsaturated zone (the length of this bubbly zone ‘l’ is estimated to 1.0cm). The degree of saturation at the inflection point is found to be about \( y_i = 0.90 \) for both methods. Nevertheless, slightly different results are observed for the value of the maximum saturation, this parameter clearly having the lowest sensitivity.

5 Conclusion

An innovative method was proposed to determine the saturation curve of a fibrous media in typical conditions of a RTM-process. An experimental bench has been developed in which injections of a liquid in a unidirectional glass preform have been performed at constant flow rate. Heat transfer has been quantified using a simplified model and restrictive assumptions. A special care has been paid to the modeling of the transverse thermal conductivity, which has been realized using a homogenization methodology. This estimation has been coupled with a well-tried method based on conductometric measurements. Comparison of the results reveals a good agreement between both estimated saturation profiles, which tends to prove the ability of detecting the saturation using heat transfer.

References


Experimental study on the identification of the saturation of a porous media through thermal analysis.


induced void formation in a dual-scale porous media. (a) Formation of macro-voids due to capillary wicking: low injection velocity (b) Formation of micro-voids due to viscous forces: high injection velocity.

![Image](image1)

**Fig. 1** Flow-induced void formation in a dual-scale porous media.

![Image](image2)

**Fig. 2** View of the experimental device

![Image](image3)

**Fig. 3** Cross-section of the mold

![Image](image4)

**Fig. 4** Modeling of the saturation curve using geometric parameters $l$, $y$, and $S_{\text{max}}$.

![Image](image5)

**Figure 5**: Sensitivity of the transverse heat flux to several parameters of the saturation curve: to $y_1$ (a), to $S_{\text{max}}$ (b), to $l$ (c), and to the longitudinal thermal conductivity of the fibrous medium $\lambda_L$ (d).

![Image](image6)

**Figure 6**: Representative elementary volume at micro-scale. The micro-saturation is defined as the complementary of the micro-void fraction, i.e. the total amount of micro-voids divided by the maximum volume they may occupy.

![Image](image7)

**Figure 7**: Representative elementary volume at macro-scale. The macro-saturation is defined as the complementary of the macro-void fraction, i.e. the total amount of macro-voids divided by the maximum volume they may occupy.
Figure 8 Abacus giving the effective transverse thermal conductivity depending on the degree of filling of micro and macro-porosities. Each colored line represents a constant value of macro-saturation.

Figure 9 Responses of the three heat flux sensors during calibration with liquid only.

Figure 10 Numerical and experimental wall heat fluxes for sensors 1, 2 and 3 during an injection at average interstitial velocity of \( u = 0.25 \text{ mm.s}^{-1} \).

Figure 11 Comparison of the estimated saturations by conductometric method and by thermal identification. The ideal front represents the hypothetical case where there would be no unsaturated zone.

<table>
<thead>
<tr>
<th>Fibrous preform</th>
<th>Areal Weight (g.m(^{-2}))</th>
<th>646</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Density (kg.m(^{-3}))</td>
<td>2600</td>
</tr>
<tr>
<td></td>
<td>Fiber volume fraction (%v)</td>
<td>49</td>
</tr>
<tr>
<td></td>
<td>Number of plies</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td>Number of filament per tow</td>
<td>~2000</td>
</tr>
<tr>
<td></td>
<td>Average fiber diameter (µm)</td>
<td>7.8</td>
</tr>
<tr>
<td></td>
<td>Major axis of a tow (mm)</td>
<td>1.86</td>
</tr>
<tr>
<td></td>
<td>Minor axis of a tow (mm)</td>
<td>0.29</td>
</tr>
<tr>
<td></td>
<td>Viscosity (mPa.s)</td>
<td>132</td>
</tr>
<tr>
<td>Model liquid</td>
<td>Surface tension (mN.m(^{-1}))</td>
<td>46.96</td>
</tr>
<tr>
<td></td>
<td>Static contact angle (°)</td>
<td>45</td>
</tr>
</tbody>
</table>

Table 1 Physical properties of the glass fabric and of the model liquid.

<table>
<thead>
<tr>
<th></th>
<th>( \lambda_x )</th>
<th>( \lambda_z )</th>
<th>( C_p )</th>
</tr>
</thead>
<tbody>
<tr>
<td>S=0</td>
<td>Two-step inverse analysis method</td>
<td>Hot Disk Method</td>
<td>DSC</td>
</tr>
<tr>
<td>S=1</td>
<td>Mini RTM mold [16]</td>
<td>Hot Disk Method</td>
<td>/</td>
</tr>
</tbody>
</table>

Table 2 Set of experimental setups used to characterize thermal properties of unsaturated and saturated preform.
\[ \lambda \text{ (W.m}^{-1}.\text{K}^{-1}) \quad \Phi_i \quad \Phi_f \quad \Phi_f' \]

<table>
<thead>
<tr>
<th>Material</th>
<th>( \rho ) (kg.m(^{-3}))</th>
<th>( C_p ) (J.kg(^{-1}).K(^{-1}))</th>
<th>( \lambda ) (W.m(^{-1}).K(^{-1}))</th>
<th>( h ) (W/(m(^2).K))</th>
<th>TCR (m(^2).K.W(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glass fiber</td>
<td>2550</td>
<td>803</td>
<td>1.1</td>
<td>7</td>
<td>3.3.10(^{-4})</td>
</tr>
<tr>
<td>Glass fiber</td>
<td>2550</td>
<td>803</td>
<td>1.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Air</td>
<td>1.2</td>
<td>1006</td>
<td>( =f(T)^* )</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Model liquid</td>
<td>1226</td>
<td>2434</td>
<td>( =f(T)^{**} )</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Metal</td>
<td>7850</td>
<td>460</td>
<td>35.0</td>
<td>3.3.10(^{-4})</td>
<td></td>
</tr>
</tbody>
</table>

\*\( \lambda_{air} = 1.5207.10^{-11}T^3 - 4.8574.10^{-8}T^2 + 1.0184.10^{-4}T - 3.9333.10^{-4} \)

\**\( \lambda_{liquid} = 0.344 - 1.335.10^{-4}T + 8.082.10^{-6}T^2 \) (measured by Hot Disk method)

Table 3 Properties of the three phases solid, liquid and gas used as input parameters in the homogenization procedure.

Table 4 Thermophysical properties and estimated boundary conditions of the experimental bench. Temperature dependence of the conductivities of air and liquid has been taken into account.