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# COMPACTION BEHAVIOUR AND PERMEABILITY OF CELLULOSIC FIBRE FOR RTM APPLICATIONS

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## Abstract

With the current driving force to use more sustainable and/or recyclable materials, the automotive market is considering cellulosic fibres and biocomposites with a growing interest. With the recent developments in injection equipment and resin formulation, liquid composite moulding processes, such as Resin Transfer Moulding (RTM), appear promising to manufacture cellulosic fibre composites with a high throughput suitable for automotive applications. However, for RTM components to exhibit the required characteristics, reinforcement compaction response and permeability must be well-understood as they govern the resin flow, the injection time and the void formation, and therefore are key to success. In this paper, the compaction response and permeability of flax and hemp mats were investigated. A custom setup was used to determine the effect of the compaction and fibre volume fraction on the cellulosic fibre bed compaction response. Maximum compaction pressure and change in fibre volume fraction were then related to the ease of resin flow via permeability measurements. Finally, the results were compared to the compaction response and the permeability of traditional glass fibre mats to highlight the difference in reinforcement behaviour while using cellulosic fibre reinforcements instead of traditional glass fibres.

## Introduction

In the past decade, the use of cellulosic fibres as a reinforcement for composite material has grown interest in various industrial fields due to environmental regulations pushing to use more sustainable and/or recyclable materials. Cellulosic fibres present some advantages over traditional synthetic reinforcements (glass, carbon, aramid). They have high specific properties due to their low densities and good insulation properties, they are biodegradables and have less impact on human health. However, some drawbacks should be also mentioned such as material heterogeneity, weaker mechanical properties, water absorption, low thermal stability and weak matrix-fibre interface.

Despite their weaker mechanical properties compared to glass reinforcement (generally lower strength in tension, flexion and impact), the low density of cellulosic fibres (typically 1.45 $g/cm^3$ for hemp and flax fibres) makes them very interesting for automotive application to reduce the vehicle weight. Attempts to use cellulosic fibres as a replacement to glass fibres in vehicle are not new and they can nowadays be found in interior components or non-structural components, such as door panels, instrumental panels, seat backs, boot liner and spare tire linings [1-4]. Moreover, there is also a potential to use biocomposites for more semi-structural applications giving the right manufacturing process and right targeted properties and applications. From the different methods available to make cellulosic fibre reinforced thermoset composites, the Resin Transfer Moulding (RTM) process appears to be a promising option to manufacture industrial biofibre composite parts. This close-mould system is used in automotive

and aerospace field with conventional composites. It generally allows the production of large and complex composite structures with high consistency, tight geometrical tolerances, good surface finish, high fibre volume fraction and high mechanical properties. This process can be also easily automated, and have a great potential to manufacture cellulosic fibre composites at the industrial level.

In the past decade, various studies investigated the manufacturing of cellulosic fibre based composites by RTM with injection pressure in the range of 0.05-0.2 MPa (7 – 30 psi) [5-17]. Most of these studies focused on the quality and mechanical properties of the composite part [5-8, 10, 13-16] and only few studies examined the processing aspect of resin transfer moulded biocomposites, such as the resin flow, mould filling, and resin curing behaviour [6, 8, 9, 11-13, 17]. However, for RTM components to exhibit the required characteristics, material properties such as reinforcement compaction response and permeability must be well-understood as they govern the resin flow, the injection time and the void formation, and therefore are key to success. The permeability of hemp and flax chopped strands mats was measured in the order of $10^{-10}$ $m^2$ for a fibre volume fraction of 20%, which is 10 times smaller than the permeability of a 20% vol. glass fibre chopped strands mat [6, 11, 18, 19]. This means that, at equivalent fibre content, the filling time will be longer for hemp and flax fibres than glass fibre. Some studies demonstrated that the permeability was dependent of the fibre surface morphology, the fibre polarity as well and the fibre diameter and length [8, 17]. Francucci et al. examined the compaction behaviour of jute and sisal cellulosic fibre [19, 20] and showed that it was quite different than glass fibre mat: higher compaction forces are required and higher permanent deformation were measured due to the intrinsic characteristic of the cellulosic fibres. In wet conditions, the compaction can be decreased due to fibre fluid absorption and fibre softening.

However, no study has investigated the interaction between the compaction behaviour and the preform permeability for cellulosic fibres in liquid moulding processing. This paper presents the compaction and permeability responses of flax and hemp mats. A custom setup was used to determine the effect of the compaction and fibre volume fraction on the cellulosic fibre bed compaction response. Maximum compaction pressure and change in fibre volume fraction were then related to the ease of resin flow via permeability measurements. The results were compared to the compaction response and the permeability of traditional glass fibre mats to highlight the difference in material behaviour while using cellulosic fibre reinforcements instead of traditional glass fibres.

## Experimentation

### Materials

Hemp and flax chopped strands mats with an areal density of 500 $g/m^2$ were used in this study. Both cellulosic fibres were retted and transformed into randomly oriented mats by the TTS company using an air laid deposition process. This process consists in dispersing the long fibres in an air stream and laying them into a screen to obtain a layer of randomly oriented fibres or random mats. The thickness of the mats is adjusted by passing the layer between two blades spaced apart with the required distance and cutting the exceeding fibres in the z-direction.

Discontinuous E-glass chopped strands fibre mat with an areal density of 450 g/m2 was chosen for comparison purpose.

For the compaction and permeability tests, the different selected fibre volume fractions were calculated as follows:

$$V_f = \frac{\rho_s . nb\ plis}{\rho_f . h} \quad (1)$$

where $\rho_s$ is the reinforcement areal density (500 g/m$^2$ for the hemp and flax mats and 450 g/m$^2$ for the glass fibre mat), $\rho_f$ is the fibre density (1.45 g/m$^3$ for hemp and flax fibre, 2.54 g/m$^3$ for glass fibre) and *h* is the part thickness.

### Compaction tests

Square mat samples (10 cm x 10 cm) were cut and placed in a room with controlled atmosphere (temperature: 23 ºC, humidity: 50 %) for at least 24h prior testing. The compaction tests were performed with an Instron 5582 testing machine mounted with a 100 kN load cell and a compression fixture. Three plies of cellulosic fibre mat were stacked for each compression test. An initial pressure of 10 N, equivalent to a compression stress of 1 kPa, was applied to the cellulosic fibre mat stack before each test. The following testing procedure was then applied:

1) sample compression at a rate of 50 mm/min up to a specified thickness corresponding to a desired fibre volume fraction ($V_f$),

2) 10 minutes hold at the desired thickness to record the preform stress relaxation,

3) release of the compaction pressure and

4) repeat step 1 to 3.

Cellulosic fibre compression tests were performed up to four fibre volume fractions: 35%, 50%, 65% and 80% (calculated from Equation 1). Three samples were tested for each condition. For comparison purpose, the same procedure was carried out for a stack of 4 glass fibre mat plies.

From the compaction tests, the compression pressure ($P$), the normalized compression pressure ($P_N$), the permanent deformation ($\varepsilon$) and the stress relaxation ($\Delta P$) were calculated as follows:

$$P = \frac{F}{A} \quad (2)$$

$$P_N = \frac{P_i}{P_{\max}} \quad (3)$$

$$\varepsilon = \frac{t_1 - t_2}{t_1}.100 \quad (4)$$

$$\Delta P = 1 - P_N \quad (5)$$

where $F$ is the applied load, $A$ is the sample area, $P_i$ and $P_{max}$ are the instantaneous and maximum compression pressure, and $t_1$ and $t_2$ are the preform initial thickness at the beginning of cycle 1 and cycle2 .

## Permeability tests

A unidirectional in-plane RTM permeability setup was used to measure the cellulosic fibre mat unsaturated and saturated permeability. The permeability corresponds to the rate of flow of a fluid through a porous media. In the case of the transient state, when the porous media is being wetted by the fluid, the permeability is called unsaturated permeability, while when the porous media is fully impregnated by the fluid (steady state), the permeability is called saturated permeability.

Plies of cellulosic fibres (40 cm x 10 cm) were laid up in the mould cavity. A glass top was laid on the reinforcement and the cavity thickness was adjusted using shims of different thicknesses, between the steel frame and the glass top, in order to have a required fibre volume fraction. Four fibre volume fractions were tested: 18%, 30%, 43% and 46% (calculated from Equation 1). Due to design setup restriction, the maximum fibre volume fraction tested for cellulosic fibre is 50%. Silicone oil with a viscosity of 0.1 Pa.s, similar to the viscosity of typical RTM reactive resin was used as impregnation fluid. A pressure pot was used to inject the fluid into the preform at 1.5 bar (20 psi). The pressure gradient between the inlet and the outlet of the permeability setup was measured with a pressure sensor located at the inlet line. Two to three injections were conducted for each material and each fibre volume fraction. For comparison purposes, permeability of glass fibre mat was also measured using the same procedure.

Darcy's law was used to determine the permeability of porous media [21]. In the case of a one dimensional filling analysis at constant injection pressure, Darcy's law can be expressed as follows:

$$q_x = \phi v_x = \phi \frac{dx}{dt} = -\frac{K_{unsat}\, dP}{\mu\, dx} = \frac{K_{unsat}\, \Delta P}{\mu\, x} \tag{6}$$

where $q_x$ is the superficial fluid velocity, $v_x$ is the interstitial fluid velocity, $\phi$ is the preform porosity, $\mu$ is the fluid viscosity, $x$ is the wet length, $\Delta P$ is the pressure gradient and $K_{unsat}$ is the preform unsaturated permeability. By integrating equation (5), the unsaturated permeability can be calculated as follows:

$$K_{unsat} = \frac{x^2 \mu\, \phi}{2\, t_f\, \Delta P} \tag{7}$$

where $t_f$ is the filling time for the length $x$.

Once the preform is fully impregnated, the saturated permeability $K_{sat}$ can be calculated using equation (7):

$$K_{sat} = \frac{QL\mu}{AP} \tag{8}$$

where $Q$ is the fluid volumetric flow rate, $L$ is the total length of the preform, $A$ is the preform cross-section area and $P$ is the fluid pressure. The measurements of the saturated permeability were done 10 minutes after full impregnation of the preform.

In order to see the influence of the preform compaction on the in-plane permeability, tests with non-compacted and pre-compacted cellulosic fibre mats were carried out. A pressure of 1 MPa applied for 10 minutes was used to compress the mats before testing their permeability.

# Results

## Compaction results

Figure 1 and Figure 2 show the compaction behaviour of the different fibre mats while Figure 3 and Figure 4 present their relaxation behaviour. The compaction and relaxation behaviours are very comparable for the two types of cellulosic fibre mats.

The compaction behaviour of the cellulosic fibres is different from the one of the glass fibre. From Figure 1, it can be noticed that a higher compression pressure is needed to compact cellulosic fibre mats at a similar fibre volume fraction to glass fibre mat. For example, to achieve a fibre volume fraction of 50%, a 4 time higher compression pressure is needed for flax and hemp mats compared to glass fibre mat. The required compression pressure significantly decreases when the preform is compressed a second time, but still remains higher than the compression pressure needed for glass reinforcement. From a processing point of view, this indicates that subsequent compactions might be required to reach a higher fibre volume fraction and to close a mould under limited clamping forces. For a given composite part and moulding press, this will also engender some limitation in term of maximum achievable fibre volume fraction.

Figure 2 presents the evolution of the preform permanent deformation with an increase in fibre volume fraction. The cellulosic fibres and the glass fibre behave differently with an increase in the permanent deformation for the first ones and a decrease for the second. The permanent deformation is also significantly larger for the cellulosic fibres. This is due to the hollow structure of the cellulosic fibres and the collapse of the lumen during the compression.

On the other hand, the relaxation behaviour of cellulosic fibre mats and glass fibre mat is similar, as shown in Figure 3 and Figure 4, especially at high fibre volume content. Overall, the relaxation occurred during the first 5 minutes and the stress relaxation reaches a plateau after 10 minutes. The maximum stress relaxation decreases with an increase in fibre volume fraction (Figure 4). At lower fibre volume fraction ($V_f < 50\%$), the maximum stress relaxation of the glass fibre mat is twice as big as the one of cellulosic fibres. It then decreases to reach a similar level for Vf > 50%. Similar behaviour was observed by Francucci et al. with jute and sisal fibres [19, 20].

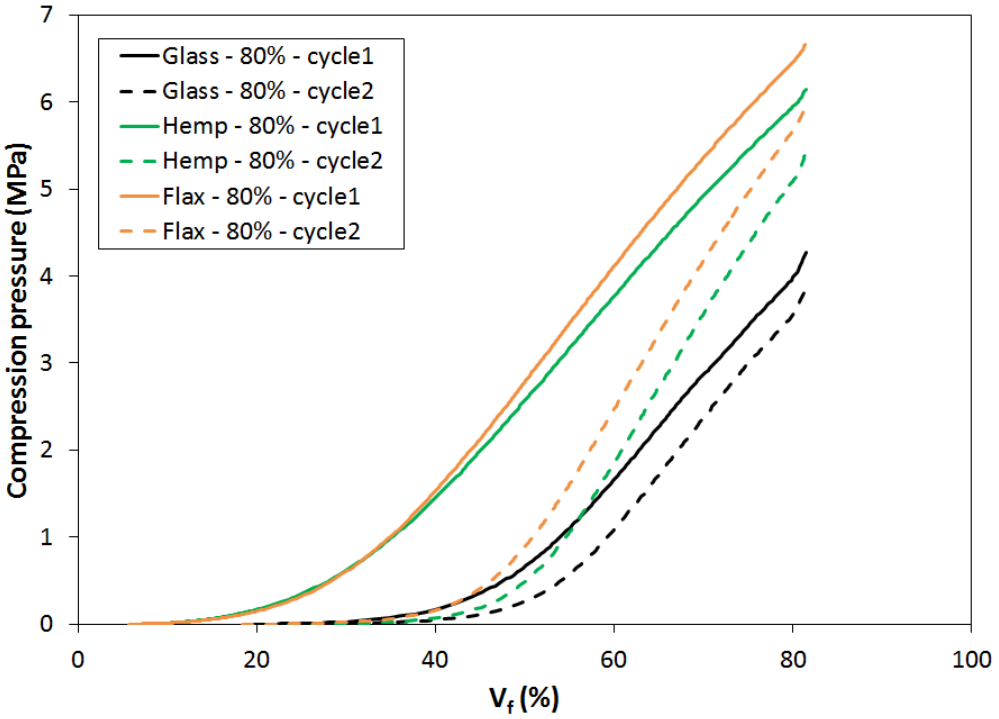


*Figure 1: Compaction behaviour of glass, hemp and flax mats at 50mm/min*

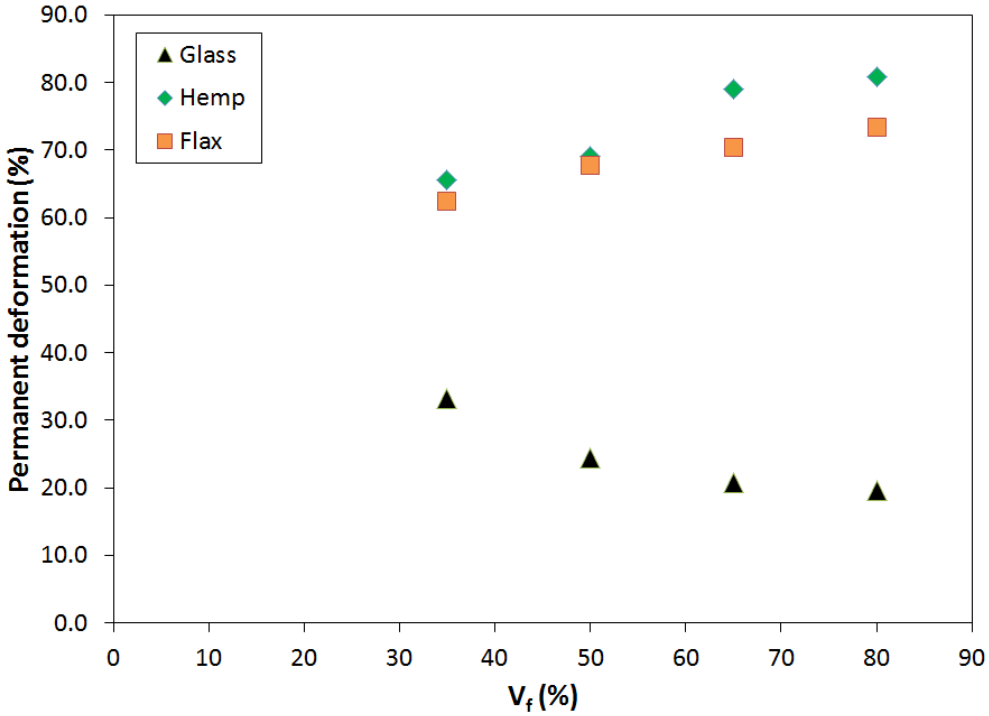


*Figure 2: Evolution of the permanent deformation with fibre volume fraction. The results were very reproducible with less than 3% of standard deviation measured for each reinforcement*

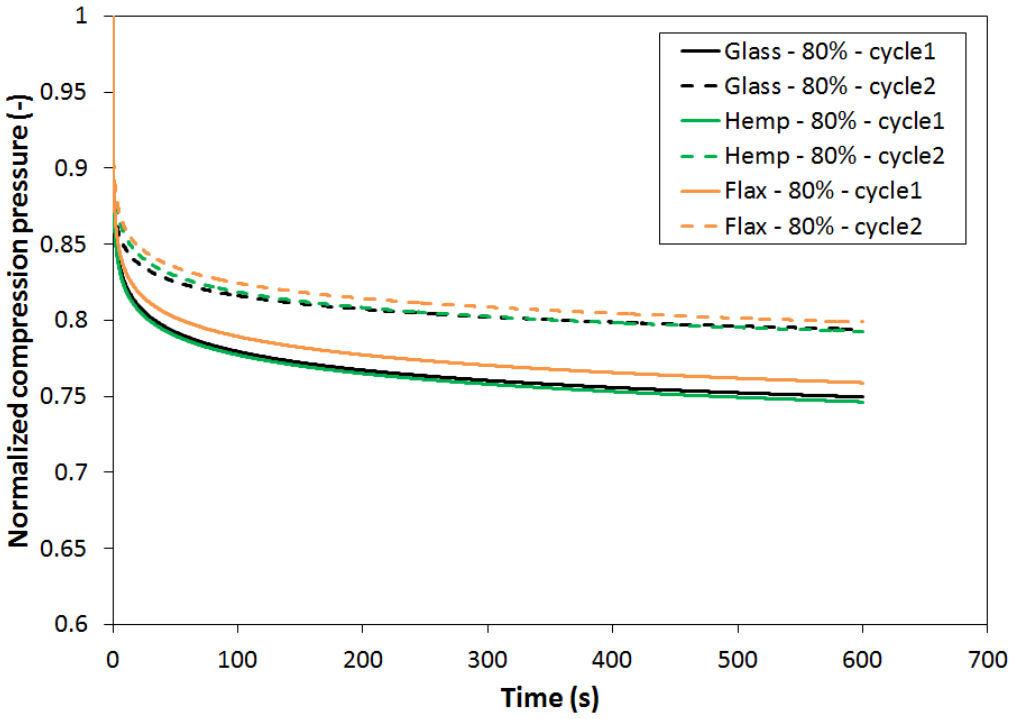


*Figure 3: Relaxation behaviour of glass, hemp and flax mats*

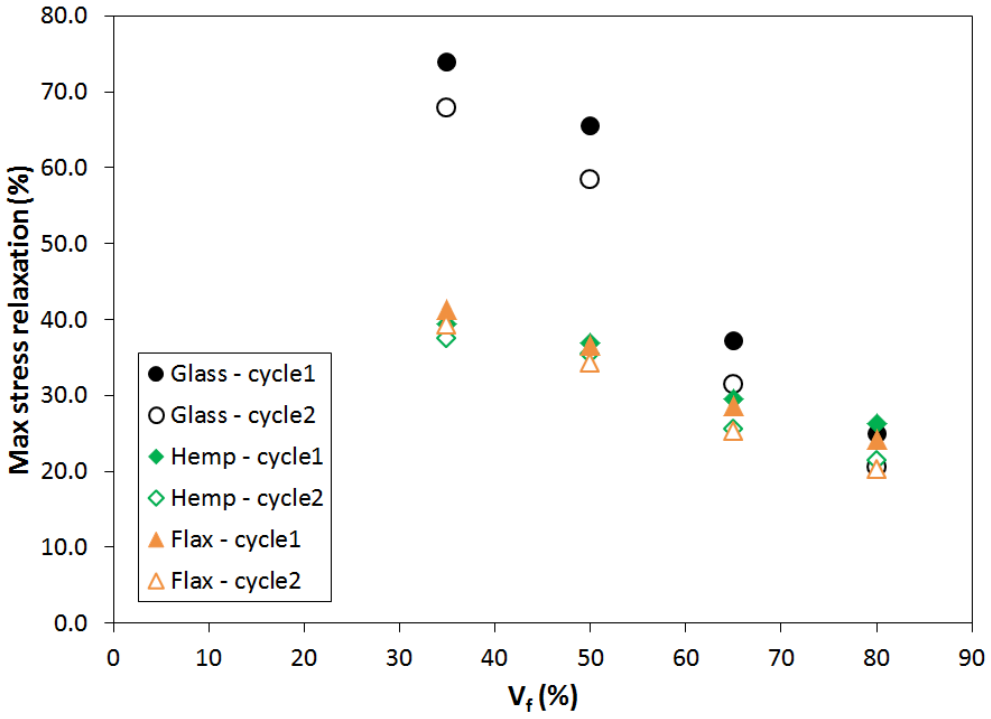


*Figure 4: Evolution of the maximum stress relaxation with fibre volume fraction. The results were very reproducible with less than 3% of standard deviation measured for each reinforcement*

## Permeability results

Figure 5 and Figure 6 show the variation of the flax, hemp and glass mat permeability with the fibre volume fraction, in unsaturated and saturated conditions, respectively. Despite the cellulosic mat variability (ply-to-ply homogeneity and areal density variation, presence of wood residues and shives), the permeability measurements are reproducible and consistent. As expected, as the fibre volume fraction increases, the preform porosity decreases, leading to a decrease in permeability. Flax and hemp mats have very similar behaviour, with a permeability ranging from $1.10^{-12}$ $m^2$ to $5.10^{-10}$ $m^2$ between 20% and 50% fibre volume fraction. Glass fibre mat has a higher permeability (up to 10 times higher) ranging between $5.10^{-11}$ $m^2$ and $2.10^{-9}$ $m^2$ for the same fibre volume fraction variation. For the three types of reinforcement, the permeability appears to be slightly higher in saturated conditions.

From a processing point of view, a lower permeability will induce a longer filling time or a shorter filling length, as defined in equation 6. Thus for a given glass fibre reinforced composite part, in order to substitute glass fibre by cellulosic fibres using the same process parameters, lower fibre volume fraction and resin with a longer gel time would have to be considered.

It is also interesting to notice that the compaction of the mats prior testing does not affect the permeability behaviour of the reinforcement: no significant permeability reduction was observed after compaction of the mats, despite the remaining permanent deformation observed during the compaction tests.

The variation of the fibre mat permeability with the fibre volume fraction can be expressed by a power law as follows:

$$K = A\left(V_f\right)^n \tag{9}$$

where $A$ and $n$ are empirical parameters determined from the experimental measurements. In unsaturated conditions, the value of $A$ and $n$ are $4.10^{-12}$ $m^2$ and -2.442 respectively for the hemp and flax fibre mats used in this project, and $4.10^{-12}$ and -3.665 for the glass fibre mat (see Figure 7).

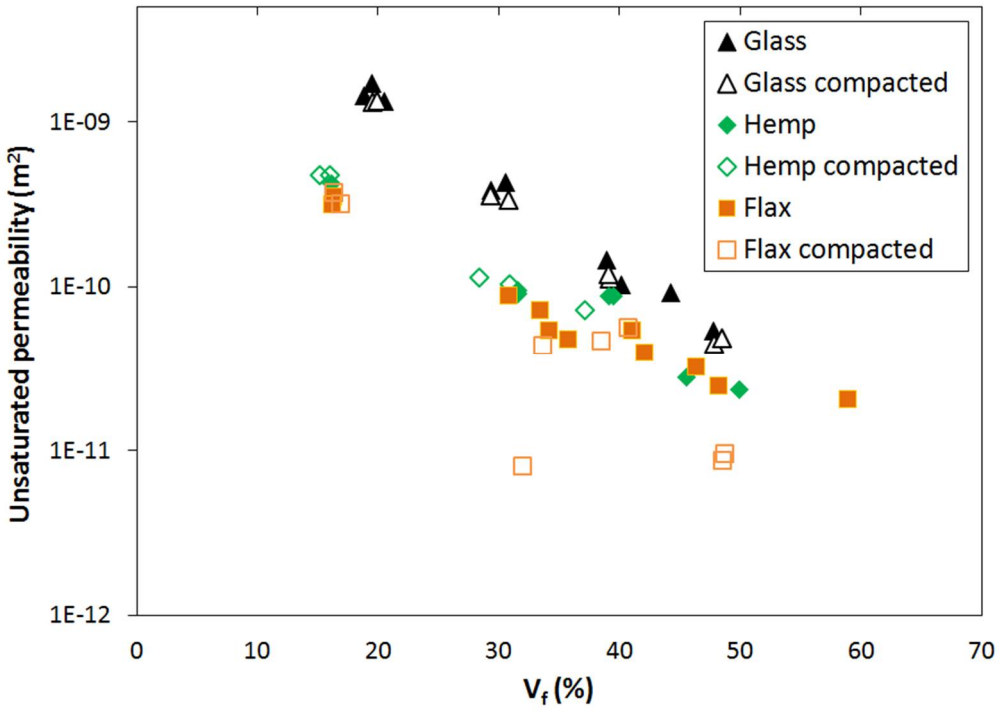


*Figure 5: Experimental variation of unsaturated permeability ($K_{unsat}$) with fibre volume fraction*

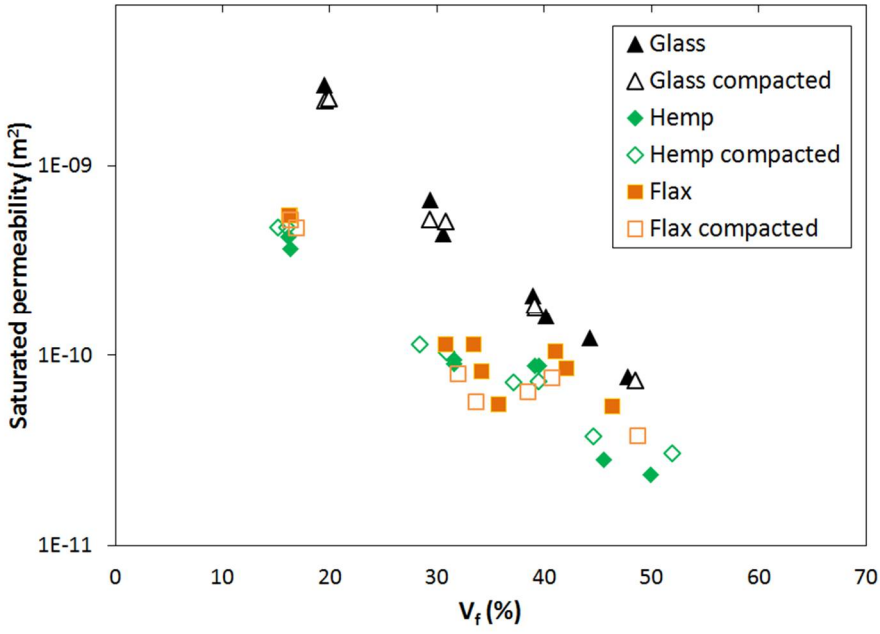


*Figure 6: Experimental variation of the saturated permeability ($K_{sat}$) with fibre volume fraction*

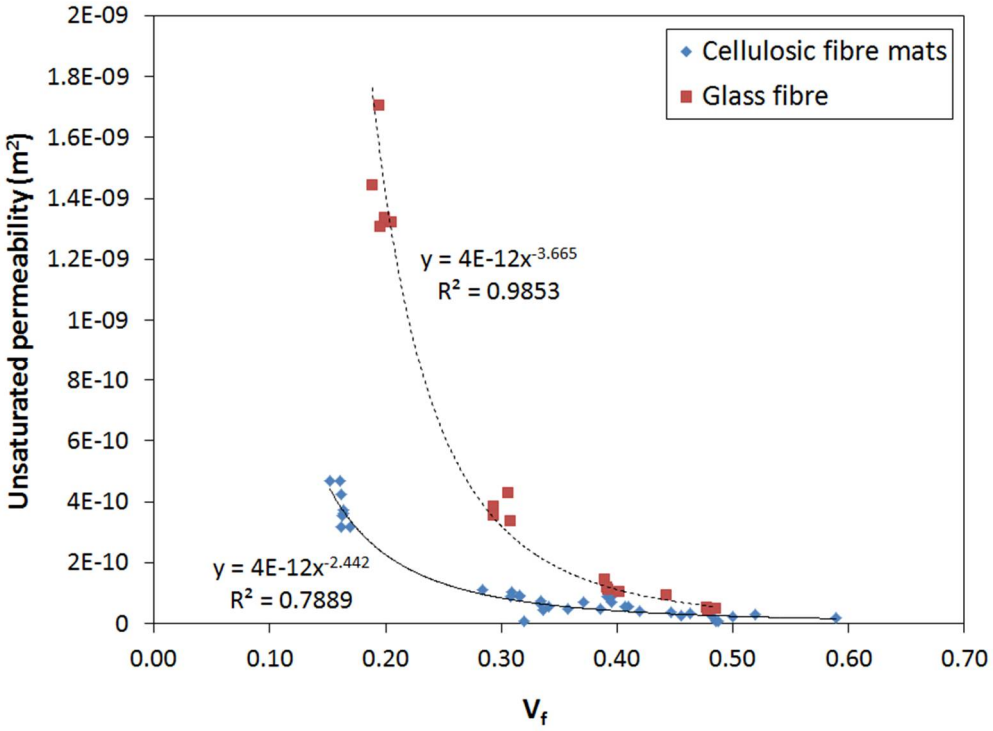


*Figure 7: Cellulosic and glass fibre mats empirical models for the evolution of the unsaturated permeability with the fibre volume fraction*

## Discussion: implications in term of processing

From the characterization of the compaction and permeability response of the cellulosic fibre mats (hemp and flax), a quite different behaviour compared to glass fibre mat was observed. The effect of these variations has to be understood and taken into account for RTM processing of such reinforcements.

Figure 8 presents a RTM processability diagram combining the compaction and permeability response of cellulosic reinforcement in function of the fibre volume fraction. The permeability curve was plotted using the empirical model described in equation 9, while the compaction response corresponds to the experimental measurement. This diagram can be used in two ways:

- to determine the maximum fibre volume fraction achievable for cellulosic fibre reinforced composite part, given a permeability limit and a compaction limit;
- or inversely, to determine the maximum pressure needed to compress the cellulosic fibres up to the required fibre volume fraction and the viscosity or gel time needed.

The permeability and compaction limits can be defined based on processing parameters. Using equation 7, the permeability limit can be defined based on the resin characteristics (viscosity and gel time) and the length of the part to fill, while the compaction limit can be determined based on the maximum tonnage of the moulding press (equation 2).

For example, assuming a permeability limit of $5.10^{-11}$ $m^2$ and a compaction limit of 1 MPa, as shown in Figure 8, the diagram can be used as follows:

- *Step A-B-C*: Considering fist only the permeability influence, the maximum predicted fibre volume fraction $V_{f1}$ is obtained at point A, where the permeability curve crosses the permeability limit. This fibre volume fraction $V_{f1}$ corresponds to a compression pressure B, needed to compact the reinforcement. If the compression pressure is higher than the compaction limit, the compression pressure has to be reduced following the compression curve decay until crossing the compaction limit at point C. The maximum fibre volume fraction achievable considering the compaction ability is then $V_{f2}$.
- *Step A*-B': On the other hand, if the compression pressure B' is lower than the compaction limit (using a pre-compaction of the preform for example), the molding press will have sufficient tonnage to compress the reinforcement and the maximum fibre volume fraction achievable will be then $V_{f1}$.

Therefore, this diagram can be then used to design the RTM process and determine the best compromise between fibre volume fraction, preform permeability and allowable compression pressure to successfully mould cellulosic fibre reinforced composite parts.

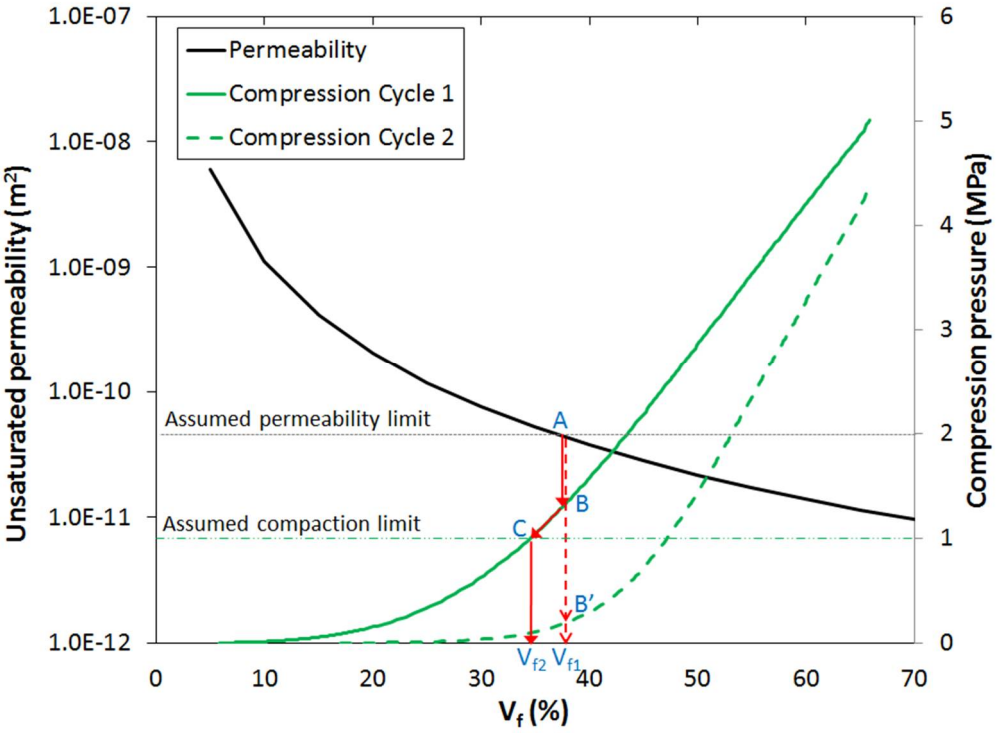


*Figure 8: RTM processability diagram*

## Conclusion

In this paper, the compaction response and permeability of cellulosic fibre mats (hemp and flax) were examined and compared to discontinuous glass fibre mat. Hemp and flax reinforcements have a very similar behaviour in term of compaction response and permeability despite their difference in fibre type. However, the cellulosic reinforcements behave quite

differently from the glass one. Overall, significantly higher compression pressure is required with cellulosic fibres to reach a specific fibre volume fraction, and lower permeability was measured compared to the glass fibre mat. The interaction between the compaction and permeability responses was then analyzed and used to understand the implications of these behaviours in term of processing. A RTM processability diagram was finally proposed by the authors to help cellulosic fibres users with the manufacturing cellulosic fibre reinforced composites by RTM process.

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