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# Analysis of the film stacking processing parameters for PLLA/flax fibre biocomposites

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**ABSTRACT:**

Nowadays, the market demand for environmentally friendly materials is rapidly increasing. Biodegradable fibres and biodegradable polymers mainly extracted from renewable resources are expected to be a major contribution to the production of new industrial high performance biodegradable composites, partially solving the problem of waste management. At the end of its lifetime, a structural biodegradable composite can be crushed and recycled through a controlled industrial composting process. Bodros *et al.* [1] showed that biodegradable PLLA (L-polylactide acid)/flax fibres mat composites exhibiting specific tensile properties equivalent to glass fibre polyester composites can be manufactured by an un-optimised film stacking process. In our study, the process has been investigated more extensively. Indeed, the compaction of flax mats requires a higher load than for glass mats of similar areal weight. The transverse permeability of flax mats has also been shown to be lower than for glass mats. In both cases, this is due to a higher degree of entanglement of the flax fibres within the mat. However, the range of permeability and compressibility values of the flax mats are well within the values that allow a good through-the-thickness impregnation. Flax fibres cannot sustain long exposures at the impregnation temperature of the mats by PLLA resin. Through-the-thickness impregnation of flax mats processes such as film stacking are more suitable than in-plane impregnation processes such as Resin Transfer Molding because the flow of resin is limited on short distances and allows short times of impregnation.

**KEY WORDS:** flax mats, PLLA bio-composites, transverse permeability, compressibility, film stacking process.

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# INTRODUCTION

## Biocomposites

A wide range of thermoplastic composites combining the benefits and properties of thermoplastics with reinforcing components such as mineral or natural fibres are currently used in industries. The first advantage of thermoplastic composites is their ability of being formed quickly in production lines, as it is already the case in the automotive industry. The second main advantage of thermoplastic composites is their ability to be recycled by remelting and remolding. However, after several recycling cycles, the mechanical performances of the matrix are affected and the composites need to be sent for incineration or disposal. In most cases, inorganic reinforcements such as glass fibres remain. In order to fully recycle the composite materials without creating any extra pollution, the use of natural biodegradable reinforcements such as flax, nettle or hemp fibres combined to a biodegradable matrix is suggested. These composites can then be composted [2]. The carbon dioxide formed during the composting step is balanced by the carbon dioxide fixed during the growing step of the flax plant [3].

Many studies have been carried out to investigate the suitability of natural fibres such as flax, hemp, ramie and sisal as reinforcing components [4-6]. Flax and nettles fibres have proved to exhibit very interesting mechanical properties. Baley [7], Baley and Bodros [8] and Charlet *et al.* [9, 10] showed that flax and nettles fibres (depending on their origins and variety) have specific tensile properties greater than those of *E* glass fibres.

Biodegradable composites or biocomposites are a combination of biodegradable thermoplastics with natural fibres. Their properties depend on the type of fibres and matrix used. The final properties of the biocomposites also depend on the manufacturing process. Any process cannot be used to manufacture composites made from natural fibres as they cannot resist exposures to

temperatures exceeding 200°C for more than few minutes. This would lead to severe fibre degradation and a loss of integrity of the reinforcement [11].

**Interest of Film Stacking (for Biocomposites Manufacturing)**

Several processes such as injection molding and film stacking used for thermoplastic composite manufacturing may be suitable to manufacture biocomposites. Film stacking process consists in heating and compressing a stack of alternated layers of polymer films and dry fibres as shown on Figure 1. Injection molding consists in injecting a mixture of liquid resin and short fibres in a closed mould. Garkhail *et al.* [12] have shown that the mechanical properties of flax/PHB (Poly-b-hydroxybutyrate) biocomposites within a range of fibre volume fraction from 10% to 40% are greater when processed using film stacking in comparison to injection molding. Indeed, fibres suffer from high shear strain during the injection, hence reducing the size and the aspect ratio length to diameter. Injection molding also has the tendency of creating non-homogeneous materials with fibre rich zones in contrast with other poorly charged zones.

To illustrate the interest of the film stacking process, Table 1 compares mechanical properties of pure PLLA biopolymer and PLLA/flax biocomposite transformed either by injection molding (Le Duigou *et al.* [13]) or by film stacking process (Bodros *et al.* [1]).

Table 1: Tensile properties of PLLA biopolymer and Flax/PLLA biocomposite.

| Material | Process | Fibre volume fraction (%) | Tensile modulus (MPa) | Tenile strength at yield (MPa) | Strain at break (%) | Ref. |
|---|---|---|---|---|---|---|
| PLLA | injection | / | 3620± 67 | 60.1 ± 1.7 | 2.4 ± 0.4 | [13] |
| Flax/PLLA | injection | 26 | 7320 ± 380 | 53.1 ± 2.8 | 1.2 ± 0.1 | |
| | film-stacking | 25 | 8856±297 | 81.3±7.7 | 1.2± 0.1 | [1] |

The biopolymer, the flax fibres and the fibre volume fraction are similar for both transformation processes. The mechanical properties determined on parts made by using the film stacking process

are better than the ones measured on parts elaborated by using the injection molding process. These results can be explained as followed:

- The length of the fibres: 10 mm for the film stacking compared to 4 mm at the beginning up to 0.4 mm at the end of the injection process. The decrease in the fibre length is due to shear forces applied to the fibres during both the compounding phase and the injection phase.
- The differences between the two transformation cycles: during the film stacking process, the components are submitted to one pressure-temperature cycle whereas for the injection process the flax fibres/PLLA mixture is extruded, transformed into pellets and injected. The compound is therefore submitted to two pressure-temperature cycles with high thermo-mechanical effects on the fibres.
- A better spreading of the fibres in the matrix for the film stacking process: the injection process may lead to accumulation of fibres at some points and also leaves other areas fibreless.

**Control Parameters of the Film Stacking Process**

Even if the parts made by Bodros *et al.* [1] by film stacking process exhibit good mechanical performances, the process cycle as shown on Figure 2 is un-optimised. Time, mechanical load and temperature are the key parameters to control the process. The values of these parameters depend on the nature of the polymer, more particularly on its melting temperature and viscosity, but also on the load and temperature the fibres can sustain. Hou *et al.* [14] showed that the thermoforming of Carbon/PEI quality parts, under a pressure of 2 MPa, takes 5 minutes at a temperature of 310°C whereas it takes 25 minutes at 280°C to complete impregnation. At 310°C, the viscosity of the resin is lower and therefore the impregnation of the dry porous reinforcement takes less time.

During the film stacking process, the matrix is heated and liquefied. Then it flows in the fibrous reinforcement under the influence of the applied load in the transverse direction. The velocity of the

flow depends on the transverse permeability as expressed by Darcy's law [15]. The transverse permeability needs to be considered because during the film stacking process, the impregnation of each reinforcing ply is carried out by infiltration of the molten thermoplastic between the fibres, mostly in the transverse direction of the laminate.

The transverse permeability can be experimentally measured using a specific experimental set up such as the one used by Saouab *et al.* [16]. It consists in applying a flow rate through the fibrous network. Then, the pressure induced at the entry of the reinforcements is measured.

During the film stacking process, the fibrous preforms are submitted to a compaction load. The compressibility of fibrous reinforcements of any form has been widely studied [17-19]. Compressibility curves can be modelled by power laws as mentioned by Toll and Manson [20].

**Aim of the paper**

The aim of this paper is to emphasize the points which need to be investigated in order to optimise the manufacturing of PLLA/flax fibre biocomposites by film stacking process. The studied parameters are temperature, time, compressibility and permeability. The viscosity of the molten polymer is also studied in order to adjust the process temperature. The degradation of the flax fibres as a function of the applied compaction load or the applied temperature is also discussed. The transverse permeability $K_z$ and transverse compressibility $C_z$ of the flax fibre mats are measured as a function of the fibre volume fraction.

## MATERIALS AND METHODS

### Materials and Sample Preparation

The used Hermes flax species is grown in Normandy (France). The flax stems are dew-retted before being scutched and finally combed. The flax fibres are cut to a 10 mm length and the mats are

prepared using a paper mill technique. The flax fibres have to be randomly scattered in order to be isotropic in a 2D plane. The orientation of the flax fibres is visually checked. Measurements on flax mats samples give an areal weight of 116 $g.m^{-2}$. The mechanical behaviour of flax fibre composites made from these mats was analysed in a previous study [1].

A glass mat coming from the nautical industry with an areal weight ($100g.m^{-}2$) comparable to the flax mat one ($116g.m^{-2}$) is tested to compare the behaviour of both reinforcements.

PLLA L9000 thermoplastic is purchased from Biomer in Germany.

**Viscosity Tests**

Viscosity tests are carried out using a Bolhin Gemini strain controlled rheometer equipped with a parallel plate fixture of 20 mm diameter. The viscosity of PLLA at the newtonian plateau is measured at four temperatures: $T_m$, $T_m$+10°C, $T_m$+20°C, and $T_m$+30°C where $T_m$ is the melting temperature of the PLLA polymer. The melting temperature was measured by DSC, using a Perkin Elmer Pyros 1 apparatus, with a heating rate of 20°C/min.

**Continuous Saturated Permeability Tests**

The continuous saturated permeability was measured on both flax and glass mats with a continuous permeability measurement device developed at the University of Le Havre (Figure 3). This apparatus consists of a stainless steel cylindrical pot within which a guided piston induces the exact amount of fibres compaction in the transverse direction. The device is mounted on a universal testing machine (Instron 8802) to control both the displacement of the piston and the force applied to the fibrous medium. The fluid is guided in the transverse direction by perforated bronze grids. A pressure transducer is placed below the lower grid. Test samples of fibrous preforms are placed between the two grids. A 6 litre syringe is placed on an Instron 5867 universal testing machine in

order to apply a controlled flow rate of silicon oil (47V100 purchased from Rhodorsil, France) to the fibrous reinforcement. The magnitude of the flow rate is controlled by the Instron 5867 crosshead speed. When a newtonian test fluid such as silicon oil is injected at a constant flow rate through the fibrous reinforcement, a pressure rise at the reinforcement entry is measured by the pressure sensor. The transverse permeability $K_z$ is calculated using Darcy's law (Equation 1). Since the piston compresses the fibrous medium at a constant speed, $K_z$ is measured in a continuous manner as a function of the increasing fibre volume fraction.

$$K_Z = \frac{\mu h}{A \Delta P} Q \tag{1}$$

where $\mu$ is the dynamic viscosity of the test fluid, $h$ and $A$ are respectively the thickness and the area of the preform, $Q$ is the imposed flow rate and $\Delta P$ is the difference of pressure induced by the constant flow rate of test fluid through the reinforcement.

The tested silicon oil has a viscosity of 0.1 Pa.s. The flax and glass mats are cut into 100 mm diameter discs by a specially made cutter. Stacks of twenty layers of flax or glass mats are compressed at 1 mm/min and submitted to a flow rate of 1.26 $cm^3/s$.

**Compressibility Tests**

Compressibility tests consist in compressing twenty layers of flax or glass mat discs. The mats are first saturated with the silicon oil and then are compressed. In any case, the following text refers to saturated compressibility.

The static compressibility tests ($\sigma=C_z(V_f)$) are performed at ambient temperature using the continuous permeability measurement device placed on the universal testing machine Instron 8802.

The flax and glass mats are cut into 80 mm diameter discs. The capacity of the load cell is 100 kN. The mats are compacted at low velocity (1 mm/min). The compressive force is measured as a function of the relative displacement of the bronze grids imposed by the machine. The distance between the grids which is inversely proportional to the fibre volume fraction is also measured.

## RESULTS

### Viscosity of PLLA

To define the processing temperature of the PLLA/flax fibre biocomposite, the variation of the viscosity as a function of temperature is measured between the melting temperature (169°C) and 199°C which is considered to be the highest sustainable temperature for flax fibres. The viscosity values are plotted as a function of the increasing temperature on Figure 4 [21].

Figure 4 shows that the viscosity of PLLA decreases quickly between 170 and 190°C whereas it decreases relatively more slowly between 190°C and 200°C. The viscosity of PLLA at 190°C is high (~1000 Pa s). However this temperature has to be chosen as the processing temperature of PLLA/flax fibre biocomposites because at higher temperature the fibre would be too damaged.

### Saturated compressibility

Figure 5 shows the evolution of the stress applied to compress a stack of twenty layers of flax or glass mats at a constant displacement velocity (1 mm/min) for saturated-by-the-fluid mats. In both cases, a stress rise is observed with increasing fibre volume fraction. A higher stress is necessary to compact the flax mats than the glass mats to an equivalent fibre volume fraction.

The compressibility curves of flax and glass mats can be fitted by power laws as explained by Toll and Manson [20] according to Equation (2):

$$\sigma_z = aV_f^{\ b} \tag{2}$$

where $V_f$ is the fibre volume fraction, $a$ and $b$ are the power law coefficients.

For flax and glass mats, the values of the coefficient $b$ are respectively equal to 4.93 and 8.04. The value of $b$ indicates the degree of disorder in the fibrous arrangement [22]. The difference of $b$ values indicates a higher disorder in the arrangement of the flax mat than in the glass mat. Figure 6 shows that the higher disorder in the flax mat could be due to:

- the lower apparent diameters of the flax bundles,
- the important spreading in the flax bundle diameters,
- the curvature of the flax bundles and fibres (mainly due to the existence of kinks) against the apparent straightness of the glass bundles,
- the lower length of flax bundles (~10mm) in comparison to the glass bundles (~40mm).

Moreover the roughness of flax bundles and fibres may prevent a good rearrangement inside the mats during compression.

**Saturated permeability**

Figure 7 shows that the pressure measured at the flax preform entry raises as a function of the fibre volume fraction. As the flow rate is constant, the transverse permeability is deduced from the pressure data (Figure 7) and Equation (1). Figure 8 shows the evolution of the transverse permeability of flax and glass mats as a function of the fibre volume fraction. The increase of pressure and decrease of permeability is related to the reduction of the pores volume between the bundles and between the fibres. At low fibre volume fraction (12-17%), a fast decrease in permeability for both materials is observed. For higher fibre volume fraction, the decrease in permeability slows down progressively.

For any fibre volume fraction, the glass mats permeability values are higher than the flax mats ones. The ratio between the transverse permeability values of glass and flax mats (Figure 8) is situated in the range 1.6-4.2. The ratio seems to increase linearly as a function of the increasing fibre volume fraction between 17% and 40%. Bréard *et al.* [23] investigated longitudinal permeabilities of flax and glass mats with areal weights of 220 $g.m^{-2}$. They found a ratio of 4 at a fibre volume fraction of 24%. The present result confirms the interest of impregnating transversely flax fibres at volume fractions lower than 0.22 where impregnation of flax mats is at the worst only twice harder than for glass fibres. At higher volume fractions, the ratio between flax and glass fibres transverse permeabilities can reach a value of 4.2.

As observed in Figure 6, the lower apparent diameters of the flax bundles induce a high degree of entanglement in the flax mat. The flow paths across the flax mats take place in smaller and more intricate channels explaining the permeability ratio of glass to flax mats.

The permeability values of flax mats being lower than the glass ones, long resin displacement within flax mats are not recommended. The film stacking process may be more suitable than longitudinal injection processes such as RTM to manufacture flax fibre biocomposites.

## DISCUSSION/ ANALYSIS OF THE PROCESS

### Effect of transverse compaction on flax fibres

During the un-optimised film stacking process cycle, the molten thermoplastic flows through the fibrous mats. The high viscosity of the thermoplastic (Figure 4) requires a sufficient compaction load to impregnate the fibres. However, the compaction load should not be high since natural fibres have much lower compression modulus than tension modulus. The transverse compaction of flax mats is highly influenced by the strong anisotropy of the natural fibres. The mean longitudinal

Young's modulus of flax fibres is 59 GPa [6] whereas the transverse modulus is estimated to be 8 GPa [24].

**Effect of temperature increase on flax fibres**

During a rise of temperature, the followings can be observed within the fibres:

- vaporisation of water. Typically, at ambient temperature, the flax fibres contain about seven percent of water on average [2]. Water desorption starts at 60°C and an exposure at higher temperatures may lead to changes of load transfer within the fibres.
- development of internal stresses that may lead to irreversible damage [25],
- physico-chemical changes of the components constituting the fibres. Transitions are observed at different temperatures. For example, pectin degradation starts around 180°C. At 230°C damages on cellulose and hemicelluloses are observed [26].

As a consequence, exposing flax fibres above 200°C leads to severe damages and a loss of their integrity according to Lilholt *et al.* [11]. At temperatures used during the process cycle (below 200°C), water evaporation and physico-chemical changes are expected to take place. A Thermal Gravimetric Analysis (TGA) was carried out on a sample of Hermes flax fibres to confirm this statement. The TGA was realised with an increasing temperature rate of 25 °C/min between 20 °C and 190°C. This is the maximum temperature rate that the apparatus used (Setaram TG-DTA 92-10) can perform. Figure 9 shows the evolution of weight loss as a function of the increasing temperature. Between 35 and 190 °C, a non linear increase in weight loss is observed. In this study, the maximum value of the weight loss is of about 6 %. Usually the flax fibres contain about 7 % of water [2] and therefore it is expected that the weight loss observed is mainly due to water evaporation. The un-optimised process cycle uses an increasing temperature rate of 100 °C/min. Water evaporation is also expected to take place in this case. Baley *et al.* [27] showed that the slow drying in air of flax fibres influences significantly the tensile strength. However, the amount of

damages depends on the total time the fibres are in contact with air at high temperatures as it is expected that water evaporation cannot take place if the fibres are in intimate contact with the resin.

**Influence of PLLA viscosity on the processing temperature**

At 170°C, the PLLA resin has a viscosity of 3500 Pa.s . At 190°C, the viscosity is reduced to 1000 Pa.s . Riedel *et al.* [28] suggested that the viscosity of a polymer used in a film stacking process has to be close to 100 Pa.s. This enables a better impregnation of the fibres. Considering the above section on temperature effects on flax fibres, the best compromise for the maximal temperature process is about 190°C. At this temperature, the viscosity during the impregnation is high (1000 Pa.s) but visual examination of the samples shows that a good impregnation is achieved. Moreover the parts manufactured by Bodros *et al.* exhibit good mechanical properties [1].

**Time of impregnation**

Due to the relative sensitivity to high temperatures of the flax fibres, the time of impregnation is a key parameter and should be minimised. The thickness of uncompressed flax mats is about 0.6 mm. This is low enough to suppose a very fast impregnation during the film stacking process even at high volume fraction. However for mats with higher areal weights, time of impregnation should be longer. A simplified approach is proposed here to calculate the time of impregnation.

The impregnation time of the whole stack of flax mats and PLLA films can be estimated by the time required for the PLLA resin to flow completely through one flax mat. Equation (3) is derived from Darcy's law under unsaturated conditions [29]:

$$t_{1\,mat} = \frac{\mu\, e_{1\,mat}^{\;2}\, \Phi}{2\, KP} \qquad (3)$$

where $e_{1mat}$ is the thickness of one mat, $\Phi$, the porosity volume fraction, $\mu$, the viscosity of the PLLA, $K$, the unsaturated permeability and $P$, the pressure applied on the resin. Three assumptions have to be made to calculate $t_{1\ mat}$, the time of impregnation through one mat:

- $P = C_z$. It is assumed that the pressure applied to the PLLA resin is given by the compression stress. In the film stacking process, the preform compaction is limited by the blocks inserted between the hot plates (Figure 1). The compression stress applied to the stack is greater than the stress required to compact the stack up to the blocks. In the following calculations two compression stress values are considered: the compression stress used in the un-optimised process, 2 MPa, and a higher one, 7 MPa.
- $K = 0.8 \times K_z$. When the resin flows through the mat, the unsaturated permeability has to be considered. For mats, the transverse unsaturated permeability value is considered to be about 80% of the saturated permeability one.
- $e_{1\,mat} = \dfrac{Ms}{\rho_V(F)V_F}$ where $Ms$ = 116 g.m$^{-2}$ is the areal weight of the flax mat and $\rho_V(F)$ = $1.5 \times 10^3$ kg.m$^{-3}$ is the density of flax fibres. $V_F$ is chosen as the final volume fraction.

<u>Equation (3) can be expressed according to Equation (4):</u>

$$t_{1\,mat} = \frac{\mu e_{1\,mat}^{\ 2}(1 - V_F)}{1.6\ K_z C_z} \tag{4}$$

Equation (4) gives an approximate time to impregnate the entire preform. On one side, the distance the resin should flow is probably lower than the total thickness of one mat as impregnation probably takes place on both sizes of each mat. On the other side, the resin may flow laterally to fill all pores and this is not taken into account. It is expected that the second effect is not as significant as the

first one. As a consequence, the time required to fill one mat $t_{1\ mat}$ is probably an overestimate of the impregnation time of the stack.

Figure 10 shows times of impregnation of one mat, $t_{1\ mat}$ , evaluated using Equation (4) for two compaction stresses (2 and 7 MPa) and two areal weights (116 g.m$^{-2}$ and 200 g.m$^{-2}$). A theoretical thicker mat (200 g.m$^{-2}$) has been tested as an input in Equation (4) to evaluate the influence of the areal weight. Results are commented in the following lines:

- The compaction stress used in the un-optimised process (2 MPa) for the real mats (116 g.m$^{-2}$) leads to a maximum volume fraction of 25% whereas a volume fraction of fibres of 35% can be reached with a higher compaction stress (7 MPa). Increasing compaction stress leads to raise the possible volume fraction of fibres.
- Time of impregnation of the 116 g.m$^{-2}$ mat is below 0.7 s when a compaction stress of 7 MPa is applied. This time is raised to 1.8 s if a compaction stress of 2 MPa is applied. For one mat at 200 g.m$^{-2}$, the time of impregnation is below 6 s with a compaction stress of 2 MPa. Therefore, the sixty seconds plateau at 190°C used in the un-optimised process (Figure 2) may be reduced to about 10 s in our context study to prevent long exposure of the mats at such a temperature.
- The time of impregnation of one mat is increased by 33% if one increases the fibre volume fraction by a factor 3 (between 12%-35%). In the same fibre volume fraction range, the permeability decreases by a factor 10 (Figure 8). As a consequence, the permeability does not seem to be the major parameter controlling the time of impregnation.
- The impregnation time of the 200 g.m$^{-2}$ mat is about three times higher than for the 116 g.m$^{-2}$ mat. Considering equation (4), the impregnation time of a mat with an areal weight $Ms_i$ is given by $t_{1mat}(Ms_i) = \frac{Ms_i^{\ 2}}{116^2} t_{1mat}(116)$. Increasing areal weight induces a higher duration of impregnation. It is possible to overcome this effect by increasing the compaction stress.

During the impregnation stage of the film stacking process, the compression stress is the key factor. The impact of permeability is limited because the flow of resin takes place on a very short distance. Increasing the compression stress allows higher fibre volume fractions and lower impregnation times. Mats with higher areal weights can be produced. However, the time of impregnation increases as a function of the square of the areal weight. As a consequence, an upper limit in term of areal weight should not be exceeded to avoid a too long exposure of the flax fibres at 190°C.

Moreover a maximal volume fraction of fibres exists for PLLA/flax mats biocomposites elaborated by film stacking process. Indeed the stress applied to the stack of flax mats and thermoplastic films should not exceed a critical value to prevent mechanical damages of the natural fibres. Therefore a maximal fibre volume fraction cannot be exceeded.

## CONCLUSIONS AND FUTURE WORK

The goal of this work is to investigate parameters with the view to improve the manufacture of PLLA/flax fibres mat biocomposites using a film stacking process. The viscosity of the PLLA biopolymer is first characterized by showing a decreasing trend as a function of the increasing temperature. A minimal temperature of 190°C is required to liquefy the PLLA resin and to impregnate the flax mats. At temperatures above 190°C fibres cannot sustain long exposures without being damaged. A TGA measurement also shows that water evaporation takes place during the process. As a consequence, the impregnation step has to be as fast as possible since it takes place at 190°C. Both the evolution of permeability and compressibility in transverse and saturated conditions for stacks of twenty flax mats are studied as a function of the increasing fibre volume fraction. Results are compared with stacks of twenty glass mats results. Due to the higher degree of entanglement in the flax mat, a higher stress is required to compact flax mats than glass mats. The permeability values of flax mats are lower than the glass mats. The flow paths across the flax mats take place in smaller and more intricate channels explaining the lower permeability of the flax mats.

Long resin displacement within flax mats is not recommended. As a consequence, the film stacking process may be more suitable than longitudinal injection processes such as RTM to manufacture flax fibre mat biocomposites. Our work shows that impregnation takes place in less than ten seconds. The un-optimised process cycle time can be reduced. Only a short exposition time of fibres at 190°C is then required.

During the impregnation stage of the film stacking process, the compression stress is the key factor. The impact of permeability is limited because the flow of resin takes place on a very short distance. Raising the compression stress allows higher fibre volume fractions. However, the time of impregnation increases as a function of the square of the areal weight. As a consequence, an upper limit in term of areal weight should not be exceeded to avoid a too long exposure of the flax fibres at 190°C.

In order to minimize the time of the process especially at high temperature, it may be possible to increase the rates of temperature before and after the impregnation step. The influence of the speed of temperature increase and decrease should be considered in the processing cycle. The mechanical properties may be significantly impacted by high temperatures rates contributing to internal stress build-up within the flax fibres. Moreover, the kinetic of weight loss during the heating phase for different temperature rates may be investigated and put into relation with the final mechanical properties.

A simplified approach to calculate the time of impregnation of the flax mats by PLLA resin is shown in this work. During the film stacking process, the flow of resin takes place in the same time as compression of reinforcements. There is therefore a coupled loading between impregnation and compressibility. In this case, a modelling such as the one proposed by Ouahbi *et al.* [29] should be

considered and specific behaviour laws established to characterise the response of flax mats to hydro-mechanical loads.

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## Captions to Figures

Figure 1: Schematic of the film stacking process

Figure 2: Process characteristics of the un-optimised film stacking cycle

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Figure 9: Influence of the increasing temperature upon the weight loss of flax fibres

Figure 10: Impregnation time asa a function of the areal weight and the compaction stress

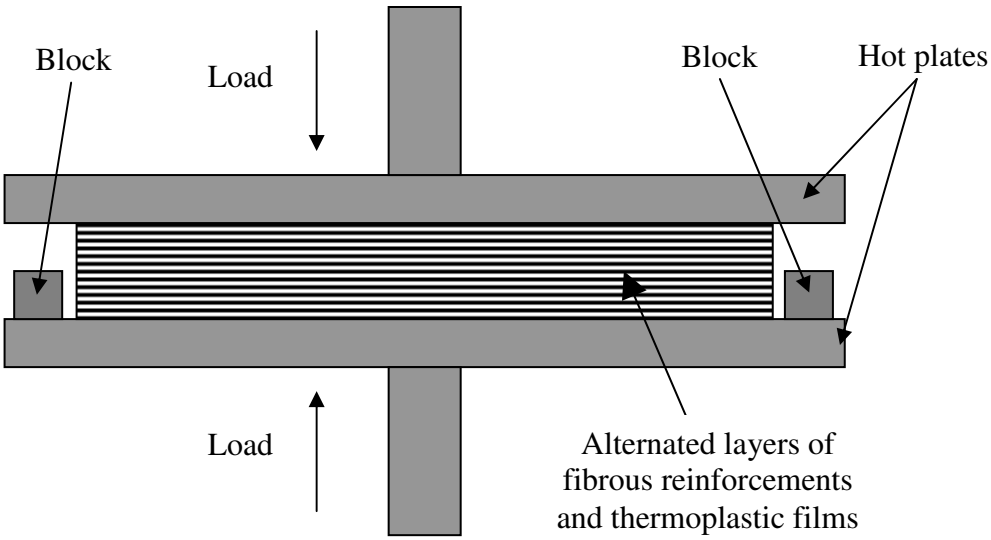



Figure 1: Schematic of the film stacking process

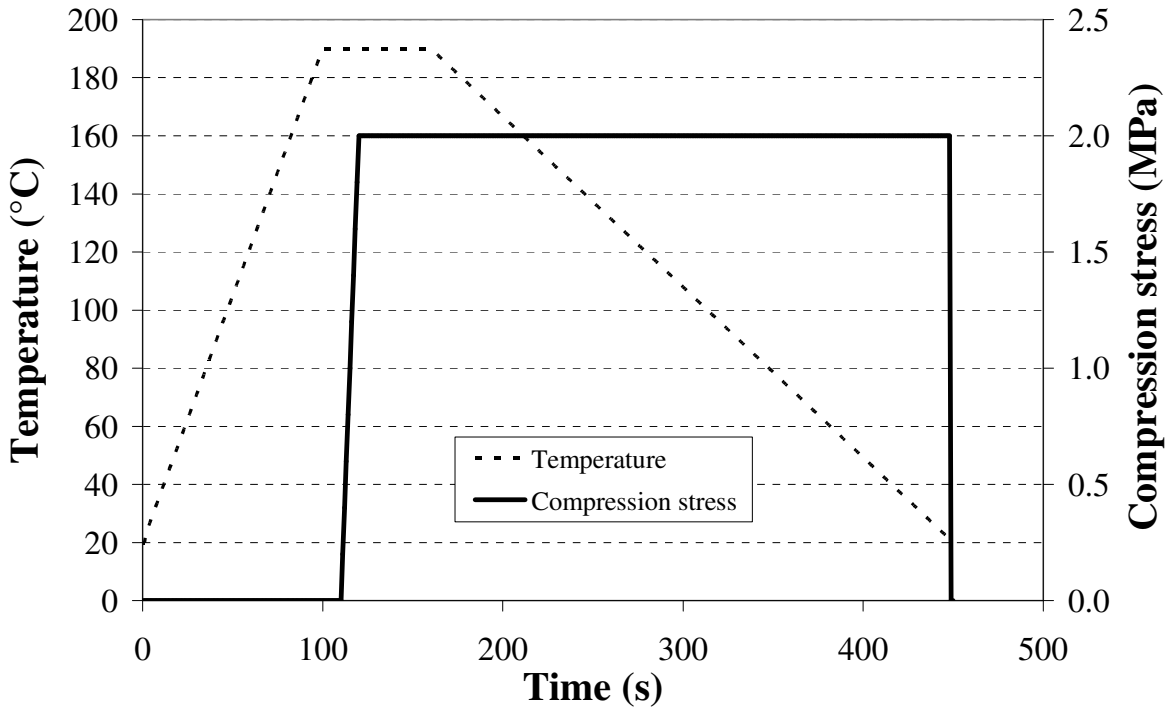



Figure 2: Process characteristics of the un-optimised film stacking cycle

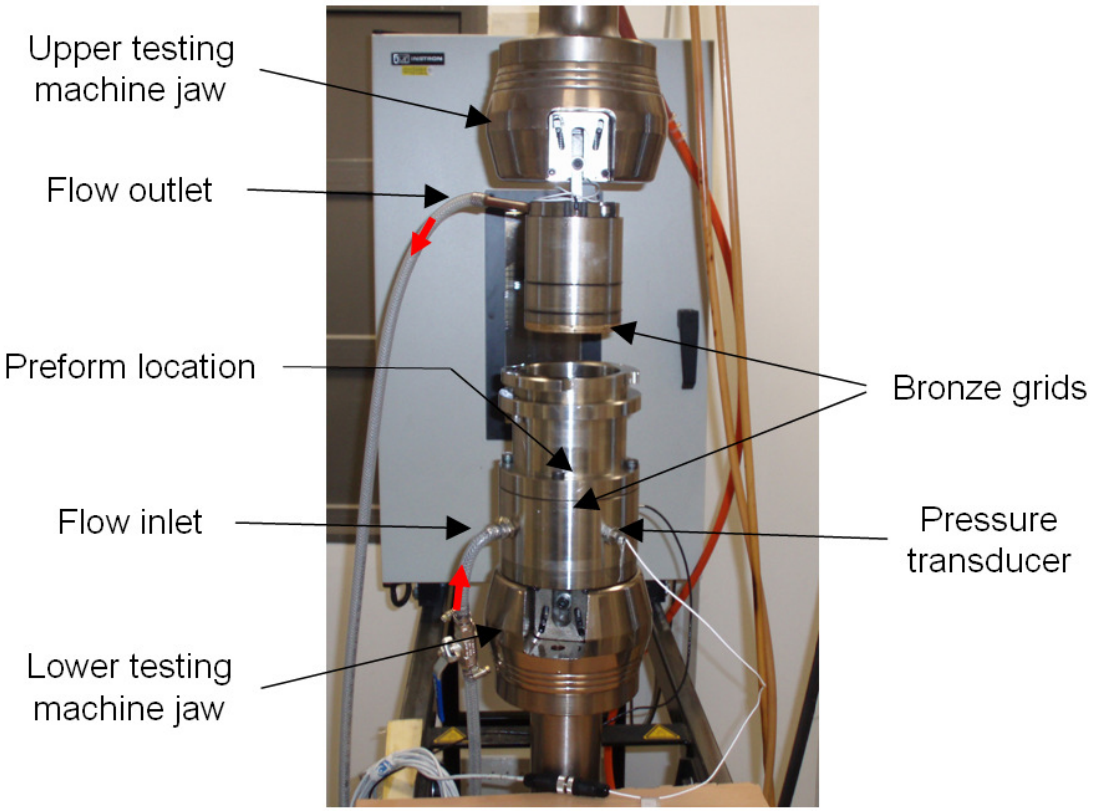
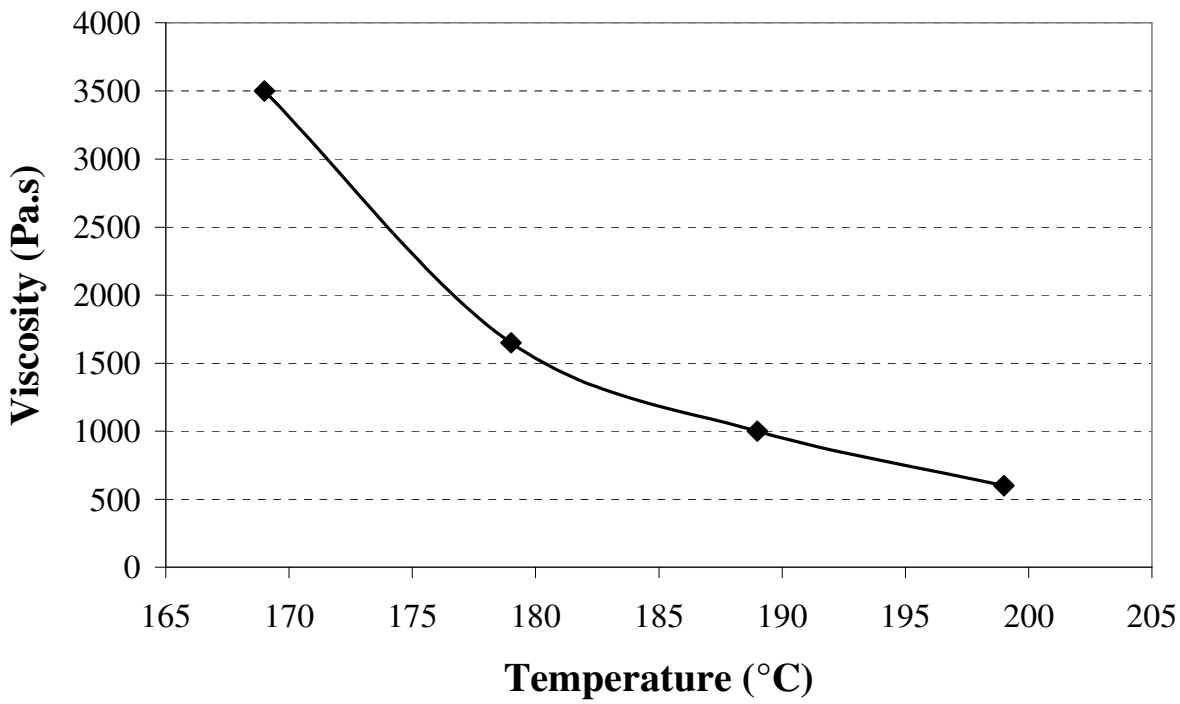



Figure 3: Continuous permeability measurement device

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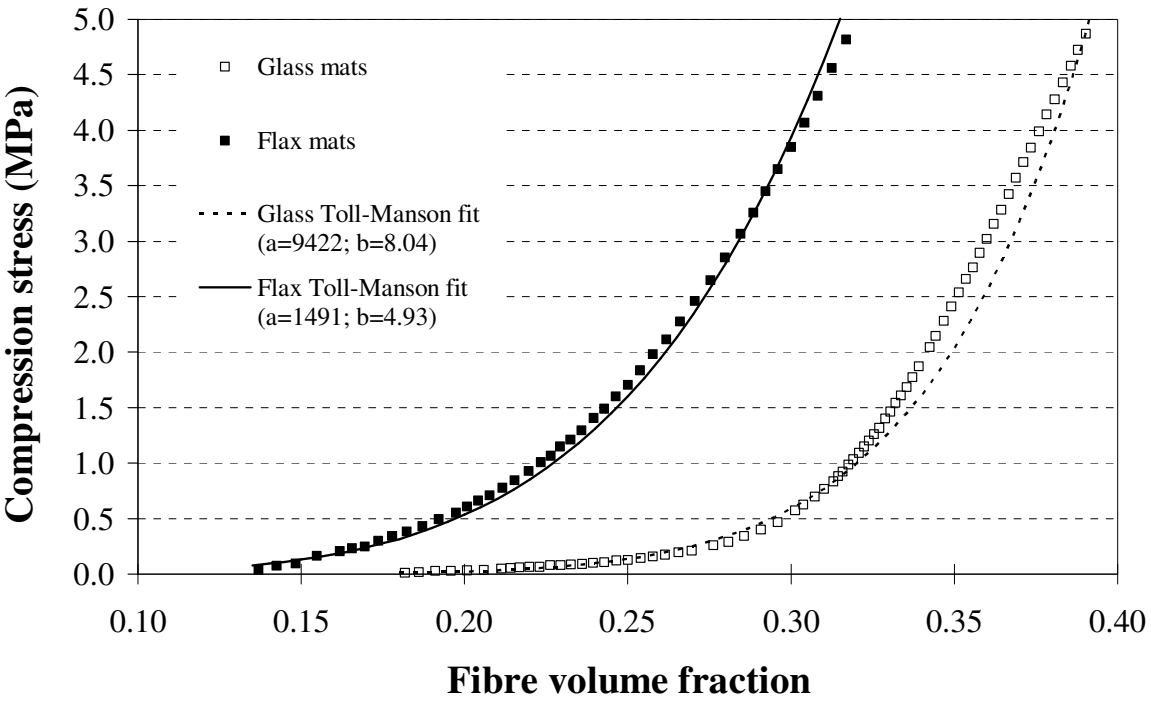
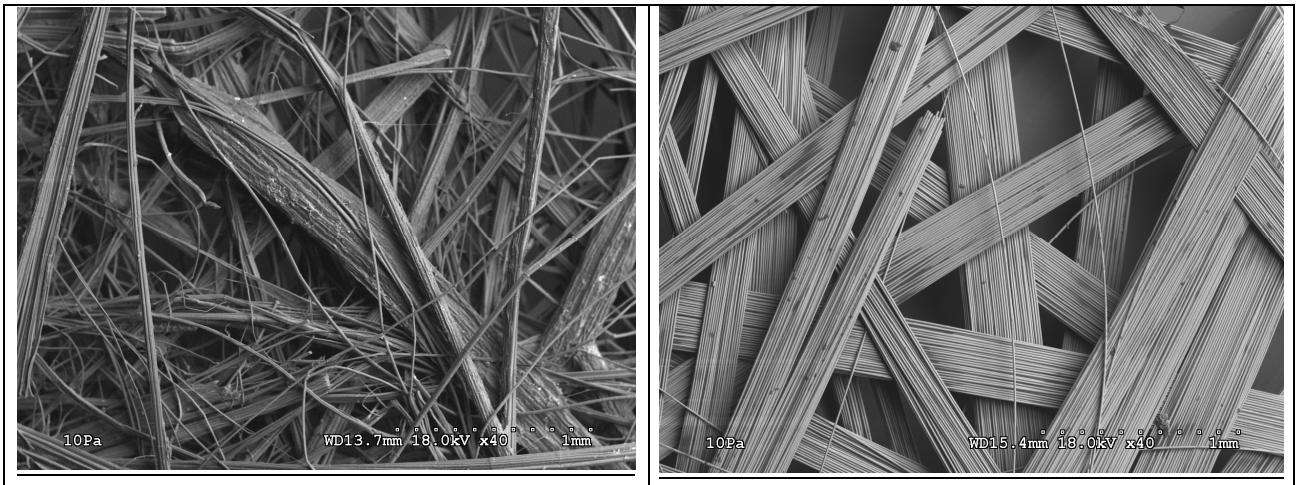



Figure 5: Saturated compressions of flax and glass mat preforms




(a) (b)

Figure 6: Comparative top view microstructures of flax (a) and glass (b) mats

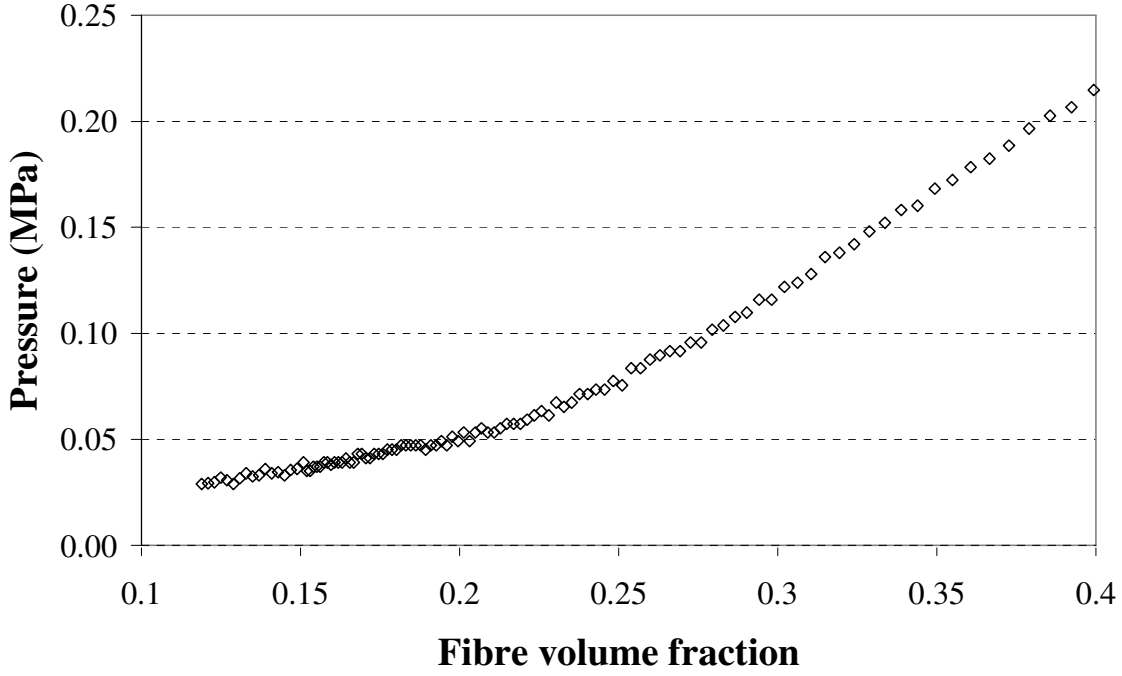



Figure 7: Pressure evolution during compression of flax mat preform

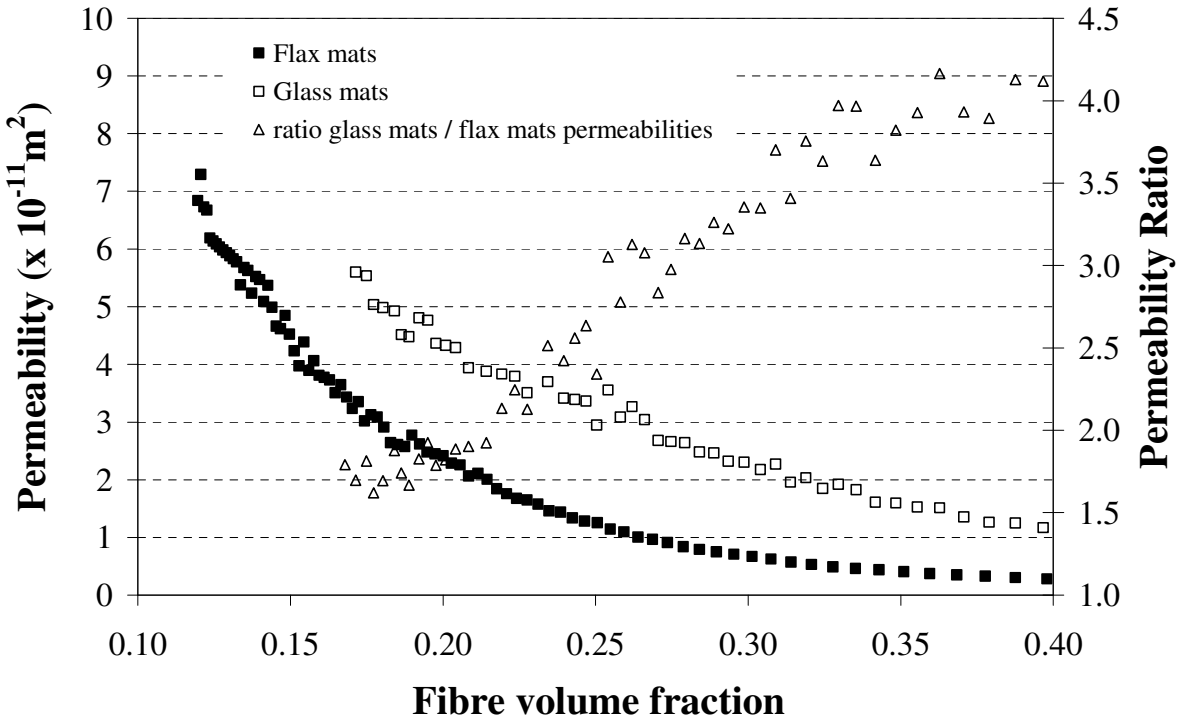
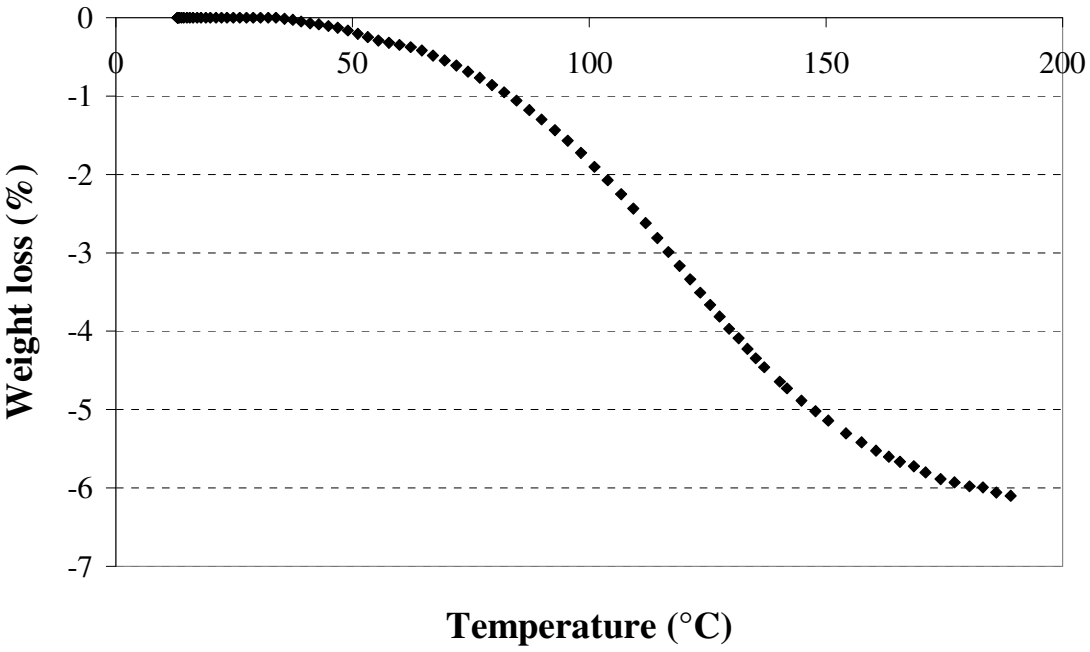



Figure 8: Transverse continuous saturated permeabilities of flax and glass mat preforms

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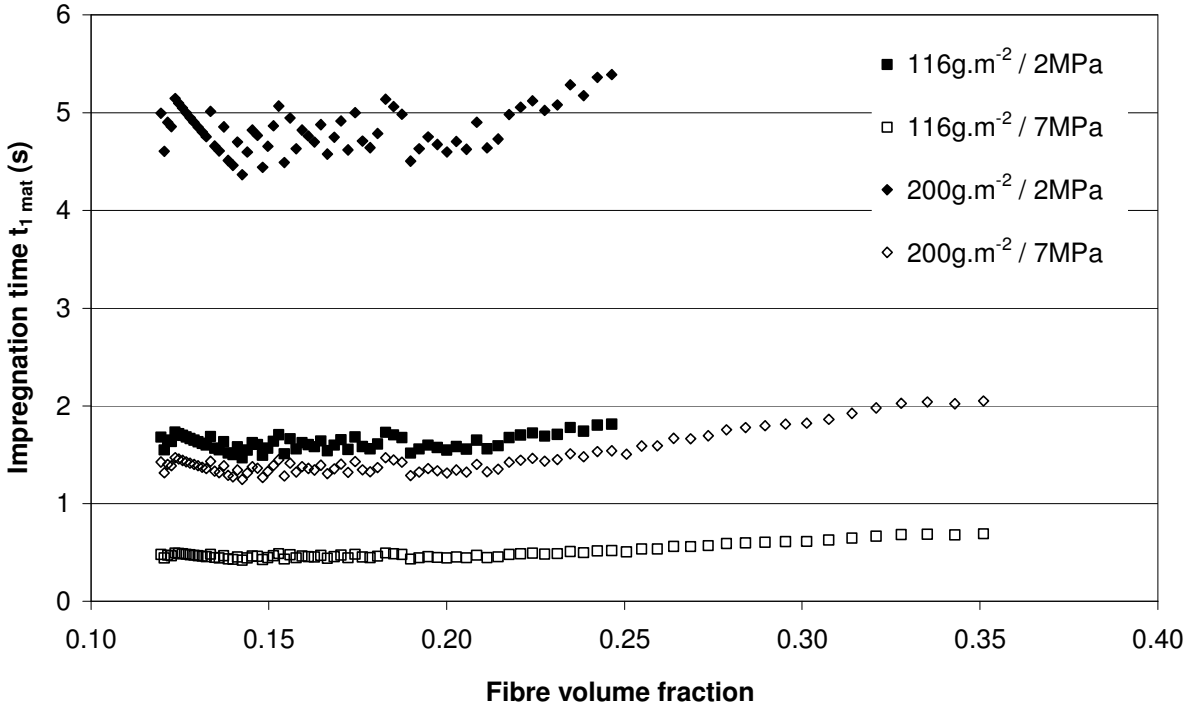


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