

# DIRECT THERMOPLASTIC MELT IMPREGNATION OF CARBON FIBRE FABRICS BY INJECTION MOULDING

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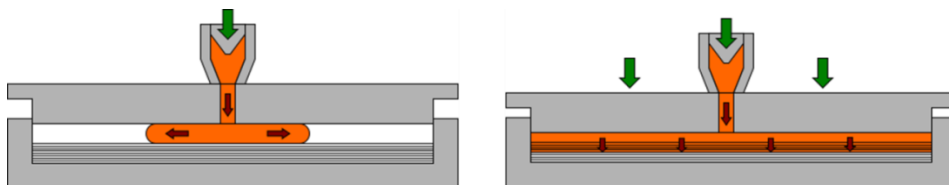
**Keywords:** Flow modelling, through thickness impregnation, thermoplastic composites, fabric compaction, fabric relaxation

## Abstract

In this paper the influence of the fabric architecture on the processing window for a pressure controlled through-thickness impregnation of dry fabrics with thermoplastic melt is discussed by measuring and implementing the compaction and permeability behaviour of two reference fabrics in a numerical analysis of the impregnation process. For the two reference fabric optimum pressure and target fibre volume fraction are defined, and the necessary adaptation of the model for a more accurate description of the direct thermoplastic injection impregnation is discussed.

## 1. Introduction

Compared to thermosets, thermoplastic matrix materials have advantages of recyclability, high fracture toughness, and offer alternative joining processes like welding. On the other hand the thermoplastic matrix materials have a higher viscosity than thermoset resins, which makes the impregnation of fabrics more difficult, especially if a high fibre volume fraction is needed. Direct melt impregnation of dry fabrics leading to high fibre volume composites was to our knowledge only demonstrated for pultrusion [1]. This paper addresses the identification of processing conditions and requirements on machine and mould design to carry out direct thermoplastic injection impregnation of carbon fibre fabrics on an injection moulding machine. The impregnation of fabrics is intended as an through thickness impregnation, combining the thermoplastic injection moulding with the compression resin transfer moulding process used for thermoset resins. The matrix is injected from the top in a gap above the dry preform, and then the fabric is impregnated trough the thickness (Fig. 1).



**Figure 1.** Processing concept for direct thermoplastic injection impregnation where the matrix is injected in a gap above a dry preform and then a uniform pressure is applied via embossing stroke.

The main difference to the through thickness impregnation with thermoset resins is the increased viscosity of the thermoplastic melt. It is intended to use very low viscosity but non-reactive thermoplastic melts with viscosities between 5 and 50 Pas.

The time  $t$  of a polymer melt to impregnate a fabric in one dimension can be estimated by Darcy's law:

$$t = \phi \frac{\eta L^2}{2K\Delta P} \quad (1)$$

where  $\phi$  is the porosity,  $\eta$  is the viscosity,  $L$  is the impregnation length,  $K$  is the permeability and  $\Delta P$  is the pressure gradient.

The porosity and permeability are determined by the fibre volume fraction,  $V_f$ , and the impregnation length by the mould geometry, here the thickness of the part. The impregnation time can be reduced either by reducing the viscosity, which is dependent on temperature and shear rate, or by increasing the pressure. However, the considerable higher injection pressure of this process leads to additional compaction of the fabric and hence to a reduced permeability [2]. In through thickness impregnation the fabric is deformable, so that  $V_f$  and thus  $K$  are dependent on the applied pressure,  $p_{ap}$ .

In this paper the influence of the fabric architecture on the processing window for a through-thickness impregnation of dry fabrics with thermoplastic melt is discussed by implementing the experimental compaction and permeability behaviour of two reference fabrics in a numerical analysis of through thickness impregnation.

## 2. Governing equations of through-thickness impregnation

### 2.1 Pressure and Flow

The through thickness impregnation of a deformable porous media with a matrix under constant pressure is described with the pressure distribution as primary solution variable. During the impregnation  $V_f$  and thus  $K$  are not constant, but a function of the preform stress,  $\sigma_{pref}$ .  $V_f$  of the preform is coupled to the flow and the pressure gradient via coupling of viscous forces in the liquid and the resulting  $\sigma_{pref}$ . This is accounted for by using Terzaghi's law to relate  $\sigma_{pref}$  to the applied pressure,  $p_{ap}$ .

$$p_{ap} = \sigma_{pref} + p \quad (2)$$

where  $p_{ap}$  is the applied pressure,  $\sigma_{pref}$  is the preform stress and  $p$  is the fluid pressure. Figure 2 illustrates Terzaghi's law and resulting variation in fibre volume fraction.

Together with conservation of momentum (Eq. 3) and the conservation of mass (Eq. 4) the process is defined.

The conservation of mass is described as the volume averaged Darcy's law (Eq. 3),

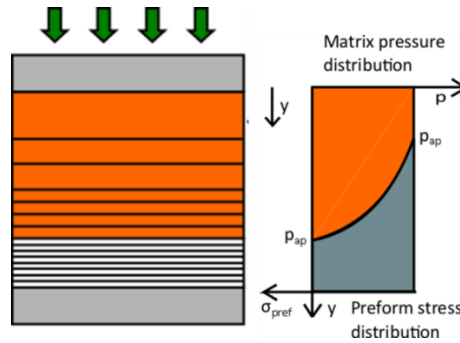
$$w_y = \frac{K_{yy}}{\eta} \cdot \frac{\partial p}{\partial y} \quad (3)$$

where  $w_y$  is the fluid velocity in the thickness direction,  $K_{yy}$  is the fabric permeability in the thickness direction,  $\eta$  is the fluid viscosity,  $p$  is the fluid pressure and  $y$  is the position inside the domain.

The conservation of mass is described for an infinitesimal control volume that is deformable in the  $y$ -direction only, where the deformation is monitored by a volumetric strain rate (Eq. 4)

$$\frac{\partial \varepsilon}{\partial t} = -\frac{\partial w_y}{\partial y} \quad (4)$$

where  $\varepsilon$  is the strain of the control volume in the thickness direction and  $t$  is the time.



**Figure 2.** Illustration of Terzaghi's law and resulting gradient in fibre volume fraction

## 2.2 Compaction

The compaction of a fabric ( $Vf$  as a function of  $\sigma_{pref}$ ) can be described with a hyperbolic tangent fit [3].

$$Vf(\sigma_{pref}) = Vf_0 + (Vf_{max} - Vf_0) \cdot \tanh^n\left(\frac{\sigma_{pref}}{P_{max}}\right) \quad (5)$$

where  $Vf$  is the fibre volume fraction,  $\sigma_{pref}$  is the preform stress,  $Vf_0$  is the minimum fibre volume fraction,  $Vf_{max}$  is the maximum fibre volume fraction,  $P_{max}$  is the maximum pressure and  $n$  is a constant.

## 2.3 Permeability

The permeability of a fabric ( $K$  as a function of  $Vf$ ) can be described with a power law [4].

$$K(Vf) = A \cdot Vf^B \quad (6)$$

where  $K$  is the saturated permeability and  $A$  and  $B$  are constants.

## 3. Numerical implementation of through thickness impregnation

In this study the fabric compaction and permeability models are adapted for a numerical implementation of the through thickness impregnation developed by B. Bachmann and K. Masania based on a work by J. Merotte et al. [5]. A detailed description on their numerical implementation is given in [6]. The following additional assumptions are made:

- Constant viscosity of the matrix: In isothermal conditions the viscosity of the thermoplastic melt is assumed to be constant at the low shear rates occurring in impregnation process.
- Capillary effects can be neglected due to the high pressure and viscosity.
- The fabric viscoelasticity is neglected (the fibre volume fraction is only dependent on preform stress)
- The lubricant effect of the matrix on the fabric compaction is neglected (dry compaction is the same as matrix wetted compaction).

## 4. Materials and Methods

In order to adapt the through thickness impregnation model, two reference reinforcements have been characterised in their compaction behaviour and through thickness permeability at much higher compaction pressure than those usually seen in liquid composite moulding.

As reference reinforcement two carbon fibre fabrics (P200C and P400C, Global Tool Trading AG, Switzerland) with the following specifications (Tab. 1) were used:

**Table 1.** Specifications of the carbon fibre fabrics.

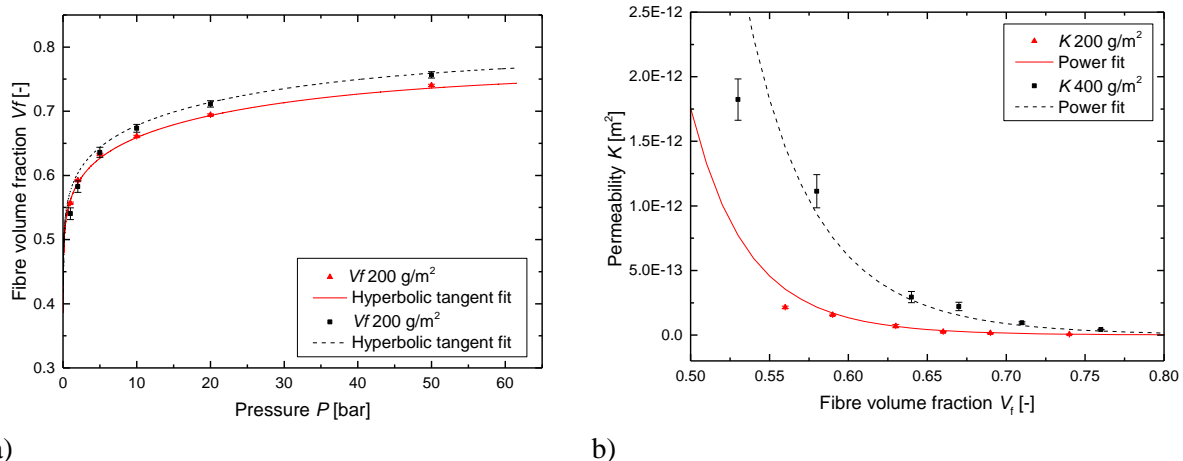
Type	Aerial weight [g/m <sup>2</sup> ]	Weave style	Tow size
P200C	200	plain	3K
P400C	400	plain	12K

#### 4.1. Compaction

10 layers of dry fabric were compacted between two parallel plates with constant velocity (0.5 mm/min) on a mechanical testing machine (Walter & Bai, Switzerland). From the force and the position of the machine  $\sigma_{\text{pref}}$  and  $V_f$  are calculated. The compaction curves are described by a hyperbolic tangent fit (Eq. 5). The mean  $V_f$  of three measurements at pressures of 1, 2, 5, 10, 20 and 50 bar were then used for a least square fit, shown in Fig. 3 a).

#### 4.2. Permeability

Measurements of the saturated through thickness permeability  $K$  at different  $V_f$  were made. Silicon oil with a viscosity of 0.1 Pas (Bluesil V100, Silitech AG, Switzerland) was used as reference fluid. The impregnation length was kept constant and the amount of layers was adapted to the different  $V_f$ .  $K$  as a function of  $V_f$  is described by a power law (Eq. 6). The mean permeability of three measurements at the  $V_f$  corresponding to a compaction at 1, 2, 5, 10, 20 and 50 bar were used for a least square fit, shown in Fig. 3 b).



**Figure 3.** a) Compaction behaviour and b) Saturated through thickness permeability of the 200 g/m<sup>2</sup> (3 K) and 400 g/m<sup>2</sup> (12 K) plain carbon fabric.

The parameters for the compaction and permeability material models are shown in Tab. 2

**Table 2.** Parameters of the hyperbolic tangent fit.

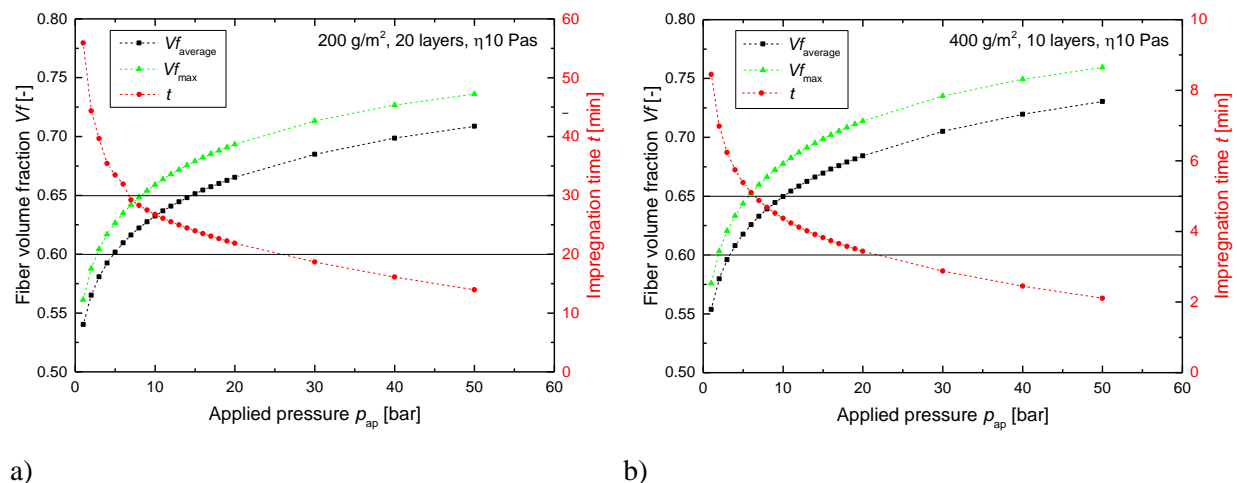
Compaction: hyperbolic tangent fit				
Fabric	$Vf_0$	$Vf_{max}$	$P_{max}$ [bar]	$n$
200 g/m <sup>2</sup>	0.28	0.76	61	0.13
400 g/m <sup>2</sup>	0.307	0.785	61	0.14
Permeability: Power law fit				
Fabric	$A$	$B$		
200 g/m <sup>2</sup>	1E-16	-13.1		
400 g/m <sup>2</sup>	1E-15	-12.56		

## 5. Results and discussion

The simulation ends if the lower boundary of the preform is reached by the flow front or if there is no more change in the positions of the flow front and preform top because the pressure is not high enough.

### 5.1. Impregnation time and average fibre volume fraction

In a first step the model was used to simulate the impregnation of 20 (200 g/m<sup>2</sup>) or 10 (400 g/m<sup>2</sup>) layers of carbon fabric (resulting in the same amount of fibres impregnated) with a thermoplastic matrix ( $\eta = 10$  Pas) from an infinite reservoir. From the results the maximum fibre volume fraction,  $Vf_{max}$ , and the average fibre volume fraction,  $Vf_{av}$ , at the end of the impregnation (when the flow front reaches the bottom of the preform) and the impregnation time,  $t$ , is evaluated, in Fig. 4 a) for the 200 g/m<sup>2</sup> fabric and in Fig. 4 b) for the 400 g/m<sup>2</sup> fabric.



**Figure 4.** Impregnation time, maximum and average fibre volume fraction at the end of impregnation with a matrix with  $\eta 10$  Pas of a) 20 layers of a 200 g/m<sup>2</sup> fabric, b) 10 layers of 400 g/m<sup>2</sup> fabric.

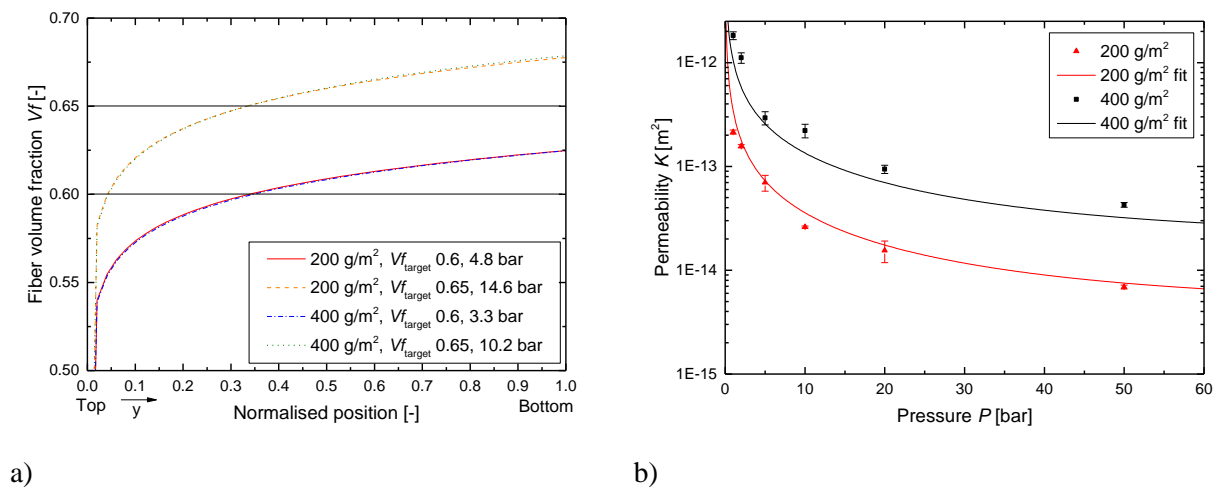
With these results the processing window can be limited. The impregnation time is much lower for the 400 g/m<sup>2</sup> fabric which is due to its higher permeability. It shows that for these fabrics a suitable target  $Vf$  would be between 0.6 and 0.65. Lower target  $Vf$  can only be reached with pressures below 5 bar and long impregnation times.

### 5.2. Optimising processing time

From Fig. 5 the optimum pressure to reach a certain target  $V_f$  while avoiding a pure matrix layer at the end of impregnation can be estimated. The process can be optimised by adapting the pressure to the desired  $V_f$ , so that the relaxation takes place at the same time as the impregnation and no additional relaxation time is needed after the impregnation step. Tab. 3 shows the results for these optimum pressure impregnation simulations and the corresponding  $V_f$  gradients at the end of impregnation are shown in Fig. 5 a).

**Table 3.** Results of the optimum pressure impregnation simulation ( $\eta$  10 Pas).

Fabric	$V_{f,target}$	$P_{ap}$ [bar]	$t_i$ [min]
200 g/m <sup>2</sup>	0.6	4.8	35.0
(20 layers)	0.65	14.6	25.3
400 g/m <sup>2</sup>	0.6	3.3	6.4
(10 layers)	0.65	10.2	4.5



**Figure 5.** a)  $V_f$  gradients resulting from the of the optimum pressure impregnation simulation ( $\eta$  10 Pas); b) Permeability as a function of pressure, explaining the big difference in impregnation time.

The distribution of  $V_f$  for both fabrics and target  $V_f$  is similar (about 0.075), with the 400 g/m<sup>2</sup> having a slightly higher gradient. When the impregnation times are compared, it shows that  $t$  is about 6 times shorter for the 400 g/m<sup>2</sup> fabric, while a lower pressure is used. This is due to the much higher permeability of the 400 g/m<sup>2</sup> fabric even at high pressure, as shown in Fig. 5 b).

An impregnation at a higher than optimum pressure will further decrease the impregnation time, but result in a pure matrix layer at the end of the impregnation, and an additional holding step is needed to allow the fabric to relax. Because of the shorter impregnation time it could still be that the impregnation at high pressure with subsequent relaxation is faster than the impregnation at optimum pressure. With the current model it is not possible to describe the relaxation after the impregnation is finished, but the next step would be to include the relaxation after impregnation in the model.

## 6. Conclusion

### Processing window with model fabrics

For the fabrics investigated a through thickness impregnation with a thermoplastic melt makes only sense for a relatively high target fibre volume fractions between 0.6-0.65.

To reach those values the applied pressure should be between 3.3 and 10.2 bar for the 400 g/m<sup>2</sup> fabric and between 4.8 and 14.6 bar for the 200 g/m<sup>2</sup> fabric, respectively.

With a detailed model for the relaxation of the fabric it should be possible to figure out if a higher pressure further reduces the process time.

### Fabric architecture

Optimised fabric architectures should be as stiff as possible, so that the pressure can be increased while the fibre volume content and the permeability increase only a little. This could be achieved by using 3D woven fabrics or 2D fabrics where the fibre bundles stay clearly separated under high pressure.

### Model optimisation

The main issue for optimisation is a more accurate description of the compaction and relaxation behaviour of the fabric and to allow for subsequent relaxation after impregnation. A combination of dry compaction at the beginning of the process and relaxation of the pre-compacted fabric in a fluid would be needed. By the use of thermoplastic matrix the fabric relaxation is only dependent on the pre-compaction, friction and a constant viscosity. It is also important to further improve the measurements methods of the compaction and relaxation of the fabrics, since the deformation at high pressures is very small.

### Model validation

For the validation of the model it is important to use a high number of fabric layers with a reference fluid to amplify the effects so that the compaction and relaxation are easily visible. These results can then be used to further optimize the relaxation model.

In conclusion of all the above mentioned aspects give a good idea where the processing window of a direct thermoplastic injection impregnation would be, and highlight the relevance to understand the saturated relaxation behaviour of the preform.

## References

- [1] A. Babeau, et al., Modeling of heat transfer and unsaturated flow in woven fiber reinforcements during direct injection-pultrusion process of thermoplastic composites. *Composites Part A: Applied Science and Manufacturing*, 77: 310-318, 2015.
- [2] R. Gennaro, et al., Experimental measurement of transversal micro- and macro permeability during compression molding of PP/Glass composites. *Polymer Composites*, 35(1): 105-112, 2014.
- [3] J. Merotte, P. Simacek and S.G. Advani, Flow analysis during compression of partially impregnated fiber preform under controlled force. *Composites Science and Technology*, 70(5): 725-733, 2010.
- [4] Ouahbi, T., et al., Modelling of hydro-mechanical coupling in infusion processes. *Composites Part A: Applied Science and Manufacturing*, 38(7): 1646-1654, 2007.

- [5] J. Merotte, P. Simacek and S.G. Advani, Resin flow analysis with fiber preform deformation in through thickness direction during Compression Resin Transfer Molding. *Composites Part A: Applied Science and Manufacturing*, 41(7): p. 881-887, 2010.
- [6] K. Masania, B. Bachmann and C. Dransfeld, The compression resin transfer moulding process for efficient composite manufacture. in *Proceedings of the 19th International Conference on Composite Materials ICCM-19*, Montreal, Canada, July 28-August 2 2013.