FILM STACKING IMPREGNATION MODEL FOR THERMOPLASTIC COMPOSITES APPLIED TO A NOVEL NET-SHAPE PREFORMING PROCESS

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ABSTRACT: In order to assess candidate impregnation methods for a novel netshape preforming method that uses low cost feedstock materials and automation, the impregnation phenomena must be understood. The preforming method comprises automated deposition of UD carbon fibre yarns together with a thermoplastic matrix along a preprogrammed path creating preforms of stacked material which are preconsolidated before conventional stamp-forming. In order to evaluate different materials, improve stacking scenarios, and assess alternate preconsolidation routes, the infiltration kinetics must be considered in detail. The transverse infiltration of liquid thermoplastic polymer into a compressible unidirectional fibre bed has therefore been examined under isothermal conditions. An infiltration model, based on local fluid flow in compressible porous media has been extended to simulate infiltration of alternating matrix film and fibre layers, relating pressure, time, and temperature with the local fibre volume fraction, pressure and liquid and solid velocities in the stacked material. The model has been validated by comparison with experimental results. The fibre volume fraction distribution gives the optimum layer thickness values for a given set of conditions. In addition the results show that increased pressure leads to increased fibre bed compaction and hence decreases permeability and limits infiltration.

KEYWORDS: Thermoplastic composites, Impregnation, Net-shape preforming

INTRODUCTION

The high viscosity of most thermoplastics has been one of the main hindrances for introducing high fibre content composites in high volume applications [1]. In order to apply the most appropriate materials for a given process, the infiltration phenomena must be analysed. Methods to decrease impregnation time, such as commingling and powder impregnation, have been reported in the literature and are used in commercial products. In the net shape tailored preforming process examined here, where UD carbon fibres together with a Polyamide matrix are automatically laid along a preprogrammed path, [2,3], film stacking is a candidate route to reduce the infiltration length and lower impregnation time.

Impregnation of dry compressible preforms under constant pressure has been examined by Michaud *et al.* [4] for glass mat thermoplastics (GMT), extending an impregnation model by Sommer *et al.* [6,7], for the infiltration part of the process, previously used for soil theory and metal matrix composites. The preform relaxation and equilibration after infiltration was shown to dominate process time for pressures above 0.05MPa. This was also experimentally verified. Control of these steps is thus important in order to prevent or tailor an inhomogeneous reinforcement distribution [4,5].

To apply and experimentally verify the impregnation model for aligned fibre composites, a fibre bed consisting of aligned UD carbon fibres has been transversally infiltrated with a Polyamide (PA) resin. The impregnation front is anticipated to be well defined, and a slug-flow assumption can be applied [7]. The impregnation time is hence dominated by micro flow behaviour since no macro flow occurs.

In addition, with consideration of the conflicting demands of maximizing final part properties and minimizing production time and hence cost, the optimum stacking scenario must be defined. The uniformity of the final fibre distribution, the effect of resin rich core layers for improved processing [9], and the average fibre content are important parameters to study. To examine these parameters, the effect of processing variables on an industrial film stacking scenario must be understood in order to optimise the entire process. Since no current model was able to examine the film stacking scenario in its complete state, a new model was built to adapt the single interface infiltration model to the problem.

TRANSVERSE INFILTRATION

To mathematically describe the constant pressure driven isothermal saturated infiltration of a compressible porous medium by a Newtonian liquid, a one dimensional infiltration model has been written based on the work of Michaud *et al.* [4,5,8,10] and Sommer *et al.* [6,7]. The model is used as a base for a film stacking impregnation model, where the time and distance space is discretized to examine complex stacking sequences through time.

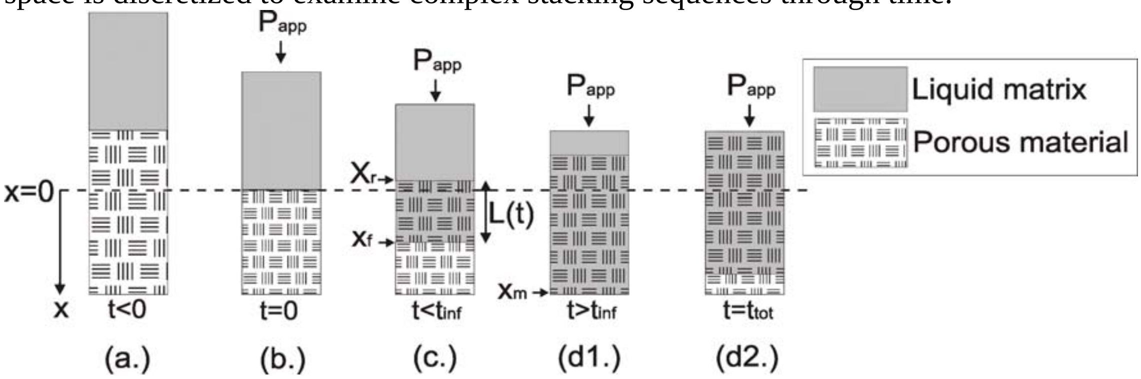

Fig. 1 Stages in the one dimensional infiltration model, after Michaud [4]

The initial condition before infiltration is a relaxed fibre bed adjacent to liquid polymer (Fig. 1a.). Pressure is then applied on the material, instantaneously compacting the fibre bed to the initial fibre volume fraction V_{fi} defined by the fibre network compliance, which has been measured for the UD fibre bed used (Fig. 1b). The impregnation front then progresses into the fibre bed with the velocity u_l (Fig. 1c.). At the same time, the fibre bed behind the impregnation front relaxes back into the matrix with the velocity u_s. At this main impregnation stage, momentum balance prescribes that the applied pressure P_{app} will be balanced between the effective load on the fibre network σ and the liquid matrix pressure P_l. The infiltration model describes the volume fraction evolution between x_r and x_f over time. If the matrix layer has an adequate thickness, the impregnation front may advance to fully impregnate the fibre preform if the right processing parameters are used (Fig. 1 d1.). On the other hand, dry fibres will remain if the matrix layer is too thin (Fig. 1 d2.).

It is assumed that the gas in the dry fibre preform is at constant pressure and is driven out of the preform or absorbed in the matrix with negligible resistance. The liquid and solid phases are taken as incompressible and homogeneous versus time. Euler coordinates are used throughout with x=0 at the interface between liquid and solid after initial compression.

Isothermal steady impregnation of a porous medium with a Newtonian fluid is described by Darcy's law [8]. It is hence required that the flow is governed by viscous forces and has a Reynolds number, $R_e \leq 1$, which is the case here due to the high viscosity and small flow rates. Darcy's law, here without gravity effects states;

$$u_l - u_s = -\frac{K}{\nu\left(1-V_f\right)}\frac{dP}{dx} \tag{1}$$

dP/dx is the pressure gradient, $K(V_f)$ the permeability of the fibre bed calculated using the model from Gebart [11] using the measured fibre radius of 4μm and V_f the volume fraction fibre. The Newtonian matrix viscosity, v, was measured and modelled with an Arrhenius relationship with v=400Pa·s for 235°C and an activation energy of 70KJ/Mol.

Neglecting internal and body forces such as gravitational forces in the solid fibre bed and the liquid matrix, stress equilibrium in one dimension yields;

$$\frac{dP_l}{dx} + \frac{d\sigma}{dx} = 0 \tag{2}$$

Further relationships are found by applying mass conservation for liquid and solid phases respectively;

$$\frac{dV_f}{dt} + \frac{d\left(V_f u_s\right)}{dx} = -\frac{dV_f}{dt} + \frac{d\left(\left(1-V_f\right)u_l\right)}{dx} = 0 \tag{3}$$

A "similarity solution" is used to simplify the problem as described by various authors [6,7]. This implies a Boltzmann transformation in Eulerian coordinates to find a reduced parameter X, which combines distance and time. The similarity solution will, following Mortensen et al [7], be valid for the case of constant applied pressure and a time independent viscosity.

$$X = \frac{x - x_r}{\psi\sqrt{t}} = \frac{x - x_r}{x_f - x_r} \tag{4}$$

where ψ is a time independent measure of the infiltration speed.

Factors for the velocities are then introduced. The infiltration front velocity u_l is governed by the factor *l(X)*.

$$u_l = \frac{\psi l\left(X\right)}{2\sqrt{t}} \tag{5}$$

The relaxation front velocity u_s is governed by the factor *s(X)*.

$$u_s = \frac{\psi s\left(X\right)}{2\sqrt{t}} \tag{6}$$

Boundary conditions
At the infiltration front: $x = x_l(t)$, $u_s = 0$, l=1+s(0), $V_f = V_{fc}$ and $P_l = P_g$, which remains constant as long as the gas is expelled or is absorbed in the matrix, and neglecting the capillary pressure drop at the infiltration front. At the preform relaxation front the conditions are as follows; $x = x_s(t)$, $P_l = P_{app} + P_g$ and $V_f = V_{fr}$.

Combining Eqn. 1 to 6 gives three non-linear first-order differential equations;

$$\frac{dV_f}{dX} = \frac{(l-s)(1-V_f)\nu\psi^2}{2KV_f \, d\sigma / dV_f} \tag{7}$$

$$\frac{ds}{dX} = [s - X - s(0)]\left(\frac{-V_{fi}'}{V_f}\right) \tag{8}$$

$$\frac{dl}{dX} = [l - X - s(0)]\left(\frac{-V_{fi}'}{1-V_f}\right) \tag{9}$$

From the development of Eqn. 8, follows that $d((1\text{-}V_f)l+V_f s)/dX=0$, showing the link between s and l, which simplifies the problem substantially since $l(X)$ can be found explicitly when $V_f(X)$ and $s(X)$ are known. Eqn. 7 and Eqn. 8 were solved for the two functions $V_f(X)$ and $s(X)$, as well as for the parameter ψ, in a commercial code Matlab™. A well converging optimum solution was found using sequential quadratic programming with initial values for ψ^2 and s(X=0) compared with known solutions for V_{fc} and $s(1)$.

The impregnation and relaxation length evolution with time is calculated by integrating the impregnation velocities over time. The corresponding real positions x to the values of X are then found to relate resin and fibre velocities, fibre volume fractions and pressures with x in the material as shown in Fig. 2a) and b).

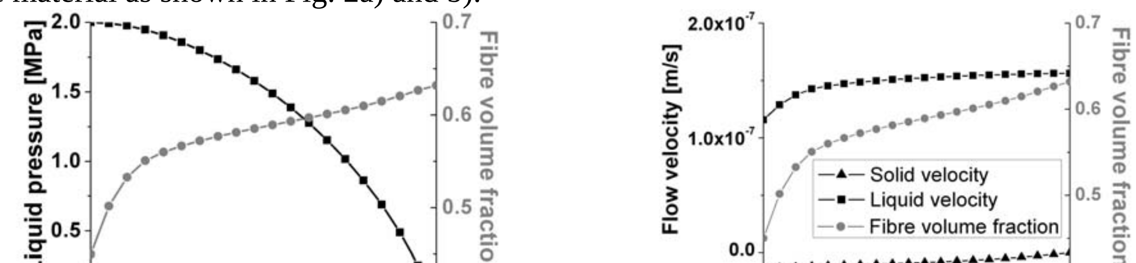

Fig. 2 The results for V_f together with a) polymer pressure and, b) flow velocities for 20bar applied pressure, 250°C and 75s impregnation time

FILM STACKING

The isothermal infiltration model described in the previous section has been extended in order to be able to model a film stacking scenario by discretizing time and position. The model is able to consider any number of layers and layer thicknesses giving freedom to examine, for example, the use of a polymer rich flow core. All data can be input via a graphical user interface (GUI) or be directly written into Excel.

To find the order of impregnation events all stacked layers are examined for each time step (typically 0.5s) from zero to a user specified impregnation time. When a fibre layer has been fully infiltrated or a matrix layer has been 'used up', the exact time for the event is recorded and excessive time is monitored. This ensures that intermediate fibre layers are impregnated from both sides and that impregnation can continue from one side if the other has "run dry" from polymer. The result is a vector called "*Interface*" containing the apparent infiltration time at all interfaces. Using the *Interface* values the amount of relaxation into the matrix for "upper-" $xr_{up}(\mathrm{n})$ and "under-side" $xr_{down}(\mathrm{n})$ for the dry fibre bed can be calculated, where n denotes the fibre layer. The infiltrated lengths $xf_{up}(\mathrm{n})$ and $xf_{down}(\mathrm{n})$ are determined correspondingly.

For each individual fibre/matrix interface, the fibre volume fraction is then found in discrete steps of a user defined step length of typically 1500 per mm. The step lengths are adapted to each individual layer thickness for improved accuracy. The pure matrix and fibre parts are subdivided in the same way. For each of the incremental steps, the fibre volume fraction is found using the previously predicted fibre volume fraction versus depth relation in the composite (x_r and x_f). An example of an arbitrary partially and fully impregnated composite can be seen in Fig. 3a) and b) respectively, from which an important variation in V_f is seen. The obtained simulation results can be used to improve the layer stacking sequence and, for example, the resin layer thickness and appropriate surface matrix layers if, for example, a matrix rich surface is desired.

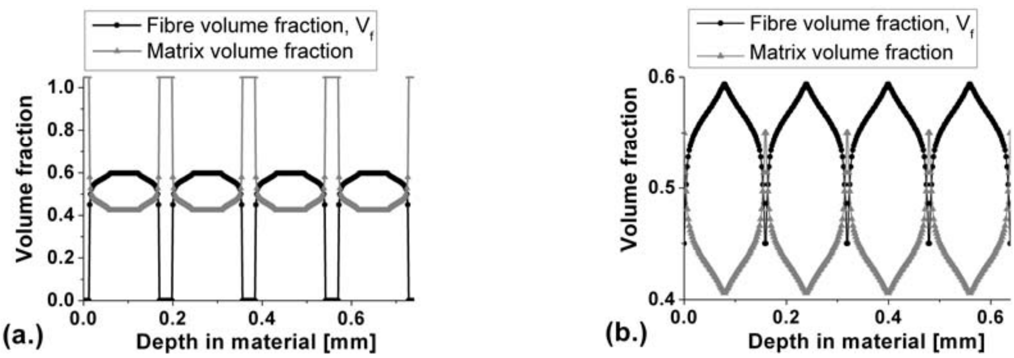

Fig. 3 The results with 4 fibre layers processed at 250°C and 10bar for: a) a partially infiltrated material after 30s and, b) a fully infiltrated material after 100s

The total time to complete impregnation of the composite is obtained by examining all individual layers and noting which has the longest infiltration time. If enough polymer thickness is specified, the optimum i'th layer thickness $T_{film}(\mathrm{i})$ can be found that results in zero excess polymer. For the outer matrix layers the calculation is simpler, but for the i'th layer it takes the general form:

$$T_{film}(i) = xf_{down}(i-1) - xr_{down}(i-1) - xf_{down}(i-1)\cdot V_{fc} + xf_{up}(i) - xr_{up}(i) - xf_{up}(i)\cdot V_{fc} \quad (10)$$

The knowledge of the complete composite fibre volume fraction distribution gives the thickness at any time, the average fibre volume fraction, and the remaining porosity in the composite.

EXPERIMENTAL VERIFICATION

An experimental evaluation of the simulated results was performed by impregnating aligned unidirectional carbon fibres with a Polyamide 6/12 copolymer for varying pressure and time. The impregnation depth was evaluated by analyzing optical micrographs as seen in Fig. 4 illustrating the difference for a 15s and a 60s impregnation time at 250^{o}C and nominally 6 bar. Due to scattering in the measured values, several measurements were taken and average values used for the impregnation length.

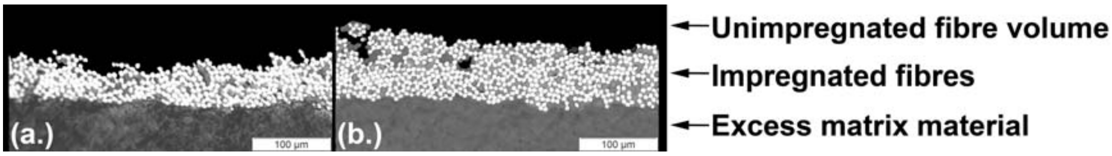

Fig. 4 Impregnation after: a) 15s, and b) 60s by nominally 6bar at 250°C

The first set of verification trials examined the effect of time on impregnation length, as shown in Fig. 5a using 6bar pressure and 250°C. The error bars for the simulation results correspond to the uncertainty in the pressure value for various experiments. The experimental results compare well with the simulation. It should be noted, that due to the machine response time, a non-negligible impregnation has already occurred at zero impregnation time, which is measured as the impregnation occurring when the applied pressure has just been reached and then removed. This will effectively move the experimentally based results towards a larger impregnation distance compared with the simulation results, especially for small impregnation times, which correlated well with the observed results. The second set of verification trials examined the effect of pressure on impregnation length for a fixed impregnation time of 60s. The pressure dependence of impregnation for a constant temperature of 250°C was measured and showed close correlation with simulation results, as seen in Fig. 5b.

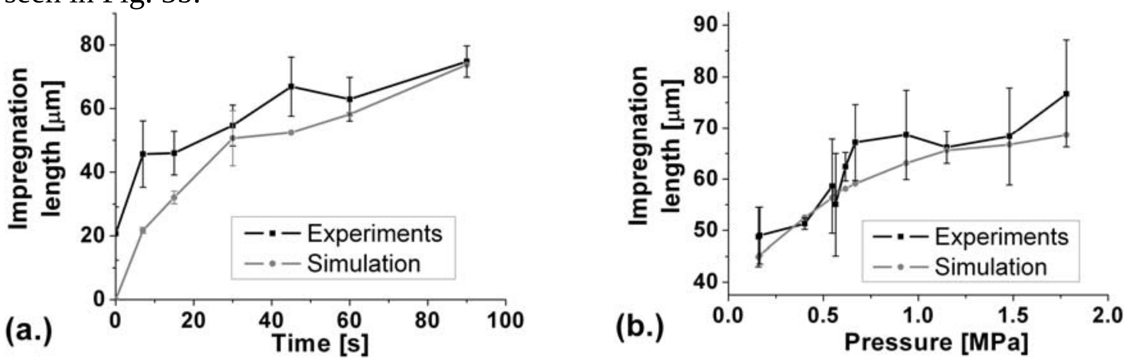

Fig. 5 Simulation results and standard deviation bars for experimental data

SIMULATION RESULTS AND DISCUSSION

Having verified the model against experimental impregnation values for changing time and pressure, the simulation model has been used to predict the effect of different stacking scenarios and processing parameters. Fibre layer thicknesses were measured in the relaxed state. The effect of temperature is shown in Fig. 6a.) for a stacking scenario with constant final thickness. Small variations (0.002mm) are due to discretization step size. When simulating the impregnation behaviour the necessary amount of matrix depends on the applied pressure as shown in Fig. 6b.) and hence fibre bed compression. The optimum thickness, which is twice the size for the inner layers having two fibre interfaces, has been used in all following simulation runs to give the minimum final part thickness.

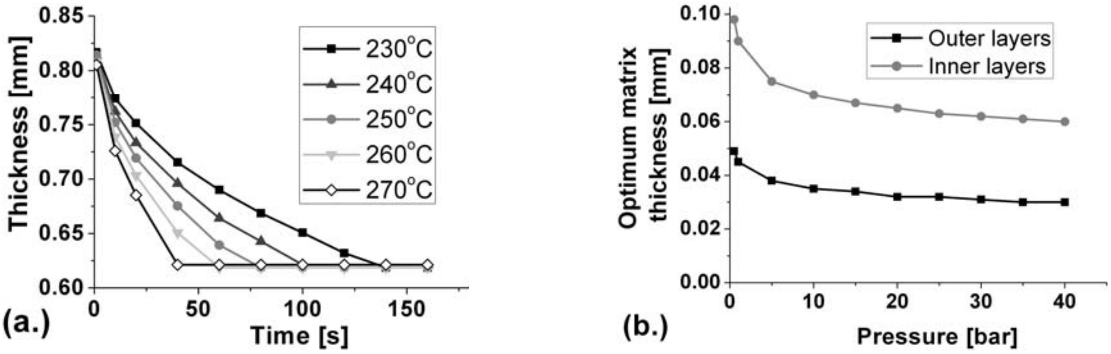

Fig. 6 a) Composite thickness evolution for four 0.2mm fibre layers and constant excess matrix, b) Necessary matrix thickness for outer and inner matrix layers depending on pressure for 0.2mm fibre layer thickness

One of the fundamental results from the simulation is the change in impregnation time with resin viscosity for different fibre layer thicknesses, here seen in Fig. 7 for constant fibre layer thickness scenarios. This can directly be used to find the impregnation cycle time for the given process and material combination.

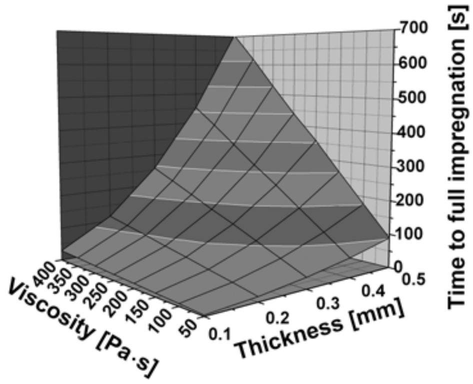

Fig. 7 Time to full impregnation as function of resin viscosity and fibre layer thickness

A pressure increase will increase the impregnation velocity up to a certain threshold value (here 20bar) even though the permeability decreases due to fibre bed compaction, as seen from Fig. 8a. By using this pressure, the impregnation time is optimized, but still depends on temperature, as shown in Fig. 8b, where the porosity content is monitored over time.

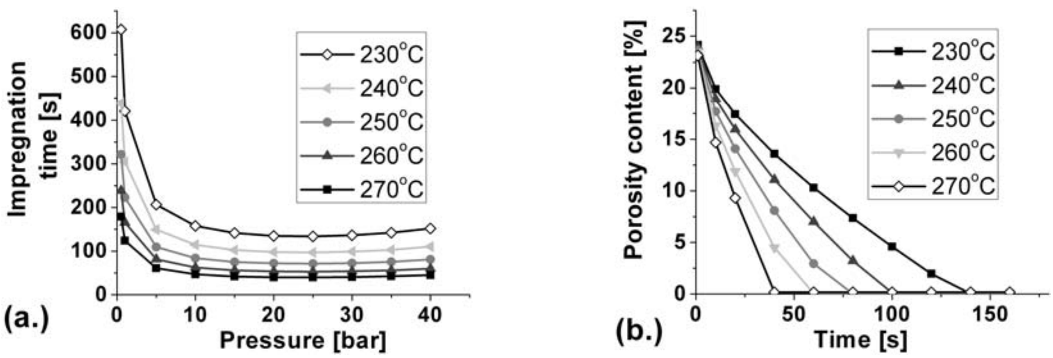

Fig. 8 For 0.2mm fibre layer thickness; a) Time for complete impregnation and b) The porosity content in the composite through process time at 20bar

CONCLUSIONS

An infiltration model has been developed and validated using the current process of film stacking of UD fibres and thermoplastic matrix layers. Experimental measurements show good agreement with simulation results without the need for fitting parameters. Using the model, it has been possible to find optimum layer thickness values for given processing conditions. Furthermore, an optimum pressure with respect to impregnation efficiency was found, which is smaller than the maximum applied pressure. This is explained by the fact that increased pressure compacts the fibre bed and hence decreases permeability and limits infiltration. The model can be used to optimize a given production scenario and the materials used, and thus help reduce the development cost for new fibre impregnation technologies.

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