INFLUENCE OF PREFORM COMPRESSIVE BEHAVIOR IN LIQUID COMPOSITE MOLDING

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ABSTRACT

In many cases of composite processing by liquid matrix impregnation, the fiber reinforcement is compressed when it comes in contact with the liquid and then relaxes as the matrix flows within its pores. This phenomenon can be analyzed in terms of local fluid flow, mass conservation and mechanical equilibrium. A model is proposed to simulate the kinetics of impregnation, and the evolution of the fiber volume fraction profile as the resin front progresses, as well as after the front has reached the end of the mold. The predictions are compared with experimental results obtained using model systems such as polyethylene glycol and glass fiber mats, as well as industrially relevant systems such as polypropylene and glass fiber mats used in the production of Glass Mat Thermoplastic (GMT) blanks. It is shown that the time for preform relaxation in a viscous fluid may be much larger than that for full impregnation. As a result, an apparently well impregnated part may exhibit an inhomogeneous distribution of the reinforcement, in turn inducing a modification of the mechanical behavior and residual stress distribution.

INTRODUCTION

In most composite processing techniques, the resin in liquid form (molten thermoplastic or still uncured thermoset) must penetrate into a fiber assembly in order to produce a sound material. Matrix and reinforcement sizings are generally designed to enhance the thermodynamic capillary forces, so that spontaneous infiltration could be expected. In practical cases, however, external pressure must be applied on the polymer to speed up the process, in particular with resins of high viscosity. The applied pressure is transmitted to the preform assembly, which may thus deform during impregnation, thereby altering the kinetics of the process and the homogeneity of the final part. Since fiber preforms are rather compliant porous materials [1,2], significant preform deformation is often observed in composite processing [1-6], and should not be neglected in analyzing the process. The phenomenon of fluid flow within deformable porous media being encountered in many other branches of engineering such as soil science or biomechanics, methods derived in these fields have been successfully applied in composite processing studies, and are reviewed in Refs.[7,8]. In particular, Sommer et al. [7] recently proposed a treatment of infiltration of initially dry deformable porous media, based on soil mechanics approach. This treatment neglects inertial forces as well as thermal and chemical transport phenomena, and uses the slug-flow assumption, which takes all infiltration to take place along a two-dimensional front within the preform. This analysis was implemented fully for unidirectional infiltration under constant

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applied pressure using the Boltzmann transformation, and was validated using experiments on the infiltration of a polyurethane sponge by Ethylene Glycol. The method was further extended to the case of non-isothermal infiltration of a porous preform by molten metal, taking into account solid phase formation [9]. An analysis along similar lines by Preziosi and co-workers [10,11] provided a theoretical evaluation of the effects of inertial forces and of various constitutive equations for the stress term for infiltration of a dry porous preform and for relaxation of an initially compressed wet porous medium.

In this paper, we present ongoing work investigating the applicability of this analytical method to practical cases of polymer composite impregnation processes. A model is proposed to predict the kinetics of impregnation, and the evolution of the fiber volume fraction distribution during impregnation, as well as after the front has reached the end of the mold. A model system is used for validation of the relaxation prediction. The analysis is then applied to liquid composite molding of Polypropylene into glass fiber mats.

GENERAL STATEMENT OF THE PROBLEM

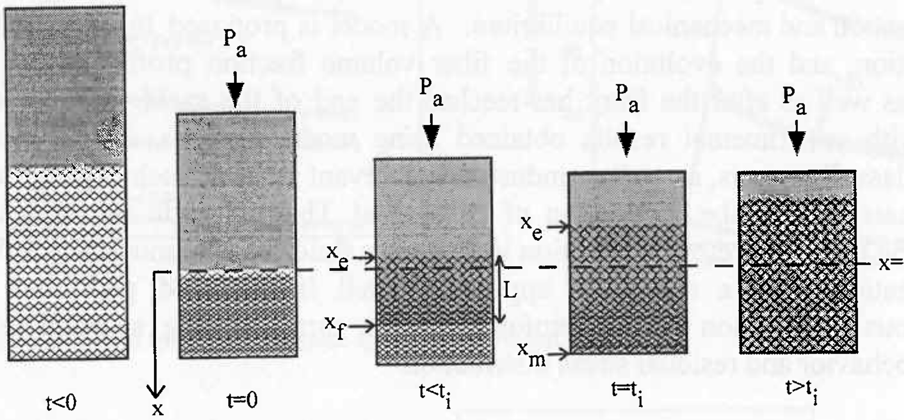

Figure1. Schematic description of the preform impregnation process

We consider the isothermal unidirectional infiltration of an initially dry fiber preform by molten polymer under constant applied pressure, as described schematically in Fig. 1. We assume that infiltration takes place in a closed mold, so that one end of the preform is fixed by the mold wall. Both flow and strain are assumed to take place along the x-direction. Capillary forces are neglected at the infiltration front, as these are generally low compared to the level of applied pressure, and are estimated to about 0.03 MPa for the application considered in this work [12]. The pressure at the infiltration front is thus exactly the gas pressure at that location, P_g. We assimilate the porous medium to a continuum, and define the local fiber volume fraction as V_f. We neglect all body forces and inertial forces, and assume for simplicity the matrix behavior to be Newtonian, so that Darcy's law is valid. We further assume that the porous medium is isotropic in a plane perpendicular to the x-axis, such that the infiltration direction is a principal axis of the permeability tensor. In the first instants of pressure application, the liquid is decelerated by the porous preform during a transient period. Simultaneously, the preform is compressed to the volume fraction V_f^c corresponding to the applied pressure P_a. As in previous studies [7, 9], we do not consider the dynamics of this transient phase. We thus simplify the problem by taking compression of the porous medium to be instantaneous. The position of the compressed preform entrance at t=0 is defined as x=0, and the x axis remains fixed in relation to mold wall, and oriented along the direction of infiltration.

As infiltration proceeds, the local pressure P in the liquid increases from P_g to P_a from behind the infiltration front to the preform entrance. The preform accordingly relaxes behind the infiltration front, along the stress-strain curve of the preform in relaxation. When the polymer reaches the mold wall, at $t=t_i$, infiltration is complete. The preform then relaxes further, with almost no fluid flow, to eventually reach the other end of the mold or a fully relaxed state, $V_f = V_f^r$.

GOVERNING EQUATIONS

The governing equations are written in one dimension over a representative volume element ΔV, following Ref.[7]. Darcy's law dictates:

$$u_l - u_s = -\frac{K}{(1-V_f)\,\mu}\frac{\partial P}{\partial x} \qquad (1)$$

where u_l is the (positive) average local velocity of the liquid within the pores, u_s the (negative) local velocity of the solid, K (a function of V_f) is the permeability of the porous medium in ΔV, μ is the liquid viscosity, and P is the pressure in the liquid.

Mass conservation in the solid and liquid phases, respectively, dictates:

$$\frac{\partial V_f}{\partial t} + \frac{\partial (V_f\, u_s)}{\partial x} = 0 \qquad (2)$$

and:

$$-\frac{\partial V_f}{\partial t} + \frac{\partial ((1-V_f)\, u_l)}{\partial x} = 0. \qquad (3)$$

Finally, having neglected inertial and body forces in both solid and liquid, stress equilibrium dictates:

$$\frac{\partial P}{\partial x} = -\frac{\partial \sigma}{\partial x} \qquad (4)$$

where σ is the effective stress acting in the solid along x, counted as positive in compression and averaged over a surface area comprising both solid and liquid.

The boundary conditions are written for the two steps which determine the process. In a first step (i), an initially dry porous medium is infiltrated by polymer. Thus, at the infiltration front, $x=x_f(t)$, $V_f = V_f^c$ and $u_s=0$, and at the preform entrance, $x=x_e(t)$, $V_f = V_f^r$.

In the second step (ii), the polymer has reached the end of the mold and the preform further relaxes. Therefore, at the front, which is the mold wall position, $x=x_m$, $u_s=u_l=0$. At the preform entrance, $x=x_e(t)$, $V_f = V_f^r$.

SOLUTION METHODOLOGY

Due to the different boundary conditions, the equations governing both steps are solved separately, the results of step (i) being implemented as initial volume fraction distribution in the solution of step (ii).

Step (i) is solved following the methodology proposed in Ref.[7], using the Boltzmann transformation to define a reduced parameter χ as

$$\chi = \frac{(x - x_e)}{\psi \sqrt{t}}$$

where ψ is chosen such that $L = x_f - x_e = \psi\sqrt{t}$.

The set of non-linear first order equations to find V_f, u_s and u_l as a function of χ is solved numerically using Mathematica™ as described in Ref.[9].

In step (ii), as the value of V_f at $x=x_m$ decreases with time, no similarity solution can be used. The set of equations is solved with a finite difference scheme, explicit in time, centered in space, including a front tracking method as described in Ref.[13], written in FORTRAN.

EXPERIMENTS

As a practical application of the present analysis, we consider the unidirectional infiltration under constant applied pressure of polymer into a needled glass fiber mat (manufactured by Symalit) of initial volume fraction 4%. The mats consist of fiber bundles about 250 μm in diameter randomly distributed in a plane, with some fibers in the orthogonal plane resulting from the needling operation performed to confer some structural integrity to the mat.

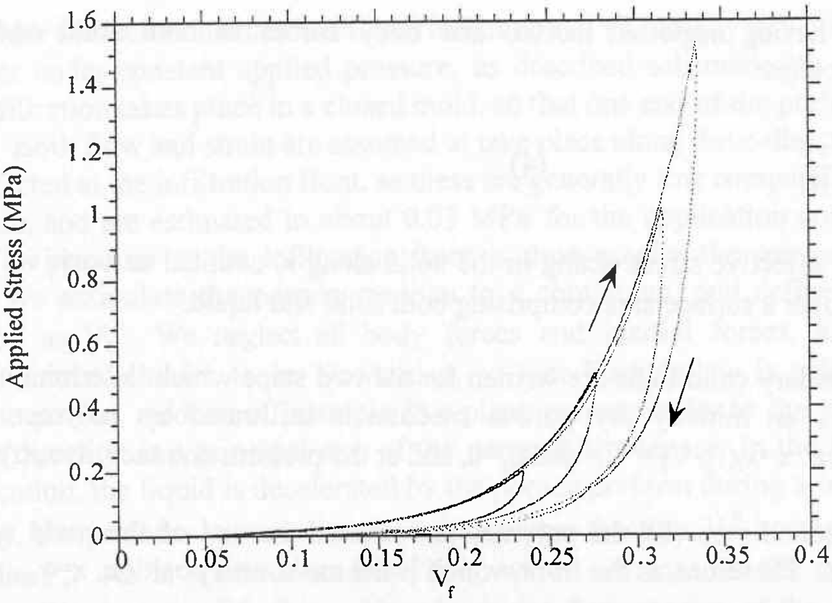

Figure 2. Stress-strain curves for the glass fiber mats at 4 different compression levels.

The compressive behavior of the fiber mat at various pressure levels was measured by compression experiments between parallel platens using a tensile testing machine (UTS 100), and is presented in Fig.2. It was verified that the stress-strain behavior of the mats remains similar in the presence of lubricants such as water, Polyethylene Glycol and WD40, and for various compression velocities. As expected, the fiber mats do not exhibit a linear behavior, and a large hysteresis is observed, most probably caused by fiber bundle rearrangement by sliding and friction during compression and relaxation. The relaxation portion of the curves is fitted using a function similar to that described in Ref.[9]. The stress-strain curve has the general form as found in Ref.[2,14], typical of random fiber assemblies.

The permeability of the fiber mats in the direction orthogonal to the fiber axis was measured in a permeameter using Polyethylene Glycol as the Newtonian flowing fluid, for various values of V_f. The evolution of K with V_f is given in Fig.3, together with the $K(V_f)$ curve used in the calculation, estimated as $K(V_f) = 1.0722\ 10^{-9}\ (1 - V_f)^{10.743}\ m^2$, as proposed in Ref. [15].

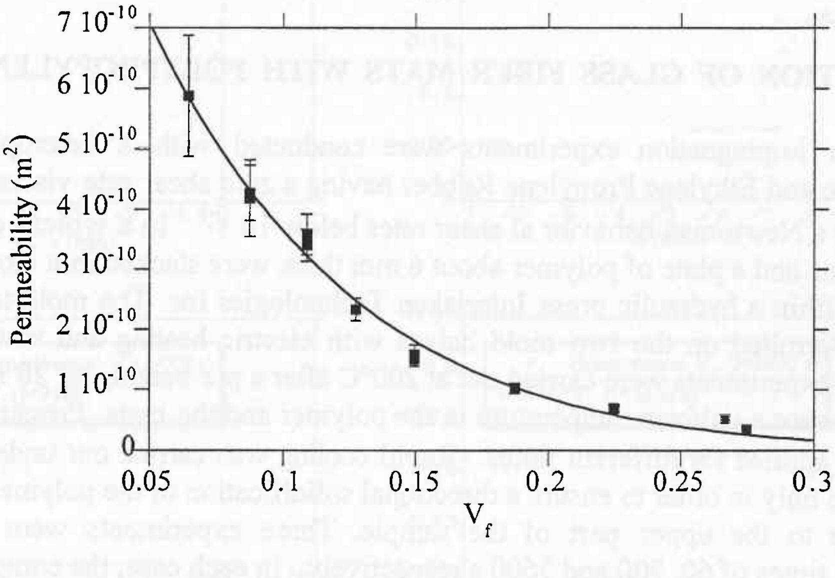

Figure3. Permeability of the fiber mats as a function of fiber volume fraction.

To check the validity of the relaxation model for the porous system considered, a set of experiments was conducted, whereby a stack of fiber mats was placed in a cylindrical tube, fully infiltrated with an aqueous solution of Polyethylene Glycol and compressed by a porous steel plunger to 0.2 MPa using a tensile testing machine (UTS 100) in compression. The plunger was then quickly removed, and the evolution of the thickness of the stack of mats recorded with a video camera. The results of the thickness evolution of the fiber mats versus time are shown for three values of the fluid viscosity, along with the results of the calculation in Fig. 4. Good agreement is observed, although we neglected gravity and friction forces at the cylinder walls, as well as a possible suction effect exerted by the plunger when it is raised in high viscosity liquids. The present analysis for step (ii) of the process thus describes well the relaxation behavior of the glass fiber mats considered in this work.

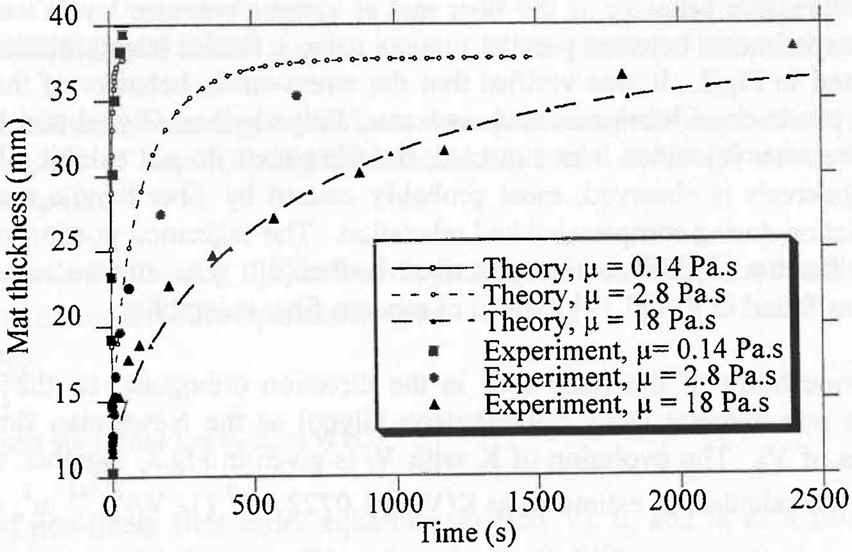

Figure 4. Theoretical and experimental mat relaxation in PEG solutions.

IMPREGNATION OF GLASS FIBER MATS WITH POLYPROPYLENE

Polymer impregnation experiments were conducted with a heterophasic blend of Polypropylene and Ethylene Propylene Rubber having a zero shear rate viscosity of 250 Pa.s at 200°C, and a Newtonian behavior at shear rates below 10 s^{-1}. In a typical experiment, two glass fiber mats and a plate of polymer about 6 mm thick were stacked in a closed mold 5cm x 5cm x 2cm within a hydraulic press Interlaken Technologies Inc. The mold temperature was separately controlled on the two mold halves with electric heating and water cooling. The impregnation experiments were carried out at 200°C after a pre-heating of 20 minutes without pressure to ensure a uniform temperature in the polymer and the mats. Pressure equal to P_a = 0.5 MPa was applied for different times. Rapid cooling was carried out under pressure from the lower side only in order to ensure a directional solidification of the polymer and to confine the shrinkage to the upper part of the sample. Three experiments were performed, for impregnation times of 60, 900 and 3600 s respectively. In each case, the composite was cut in half and polished to observe the position of the preforms, and volume fraction measurements were performed by immersion technique on small slices of composite cut through the thickness.

Simulation was performed in two steps as described above. The initial thickness of the two mats compressed at 0.5MPa was independently estimated as x_m = 1.8 mm. With the experimental conditions, and a matrix viscosity of μ=250 Pa.s, V_f^c=0.275 as seen Fig.2. The kinetic parameter is then calculated in step (i) as ψ=5.47 10^{-4} m^2/s, so that the time for the polymer to reach the end of the mold is calculated as t_i =15.93 s. At this time, L=2.18 mm, x_e=-0.385 mm, and $x_f = x_m$ = 1.8 mm. The fiber volume fraction distribution at this time is given in Fig. 5 (a), compared to that at time t=0.

Calculations for step (ii) are then performed for further relaxation of the fiber mats. The results for t=60, 900, and 3600 s are shown in Fig. 5 (b-d), together with the experimental data. It is seen that in this case, the time for relaxation is about 200 times longer than that for impregnation. Comparison with experimental values of V_f is not quantitative, as the scatter in volume fraction measurement is very large. The sample size for density measurements (about $1cm^2$ x 0.1cm) was much smaller than the representative volume element of the rather inhomogeneous mats (estimated to 25 cm^2 of mat surface). Further measurements with a larger sample size will be performed for better confidence. Also, friction on the mold walls was neglected, whereas this may prevent the mats from compressing fully and from relaxing. The trends, however, show good agreement. Impregnation of the mats is indeed complete before 60 s, with a large gradient in fiber volume fraction remaining in the solidified composite, as seen in Fig.5 (b). This gradient slowly decreases with time, while the mat thickness increases roughly as predicted, to reach an equilibrium relaxed value after about 45 minutes.

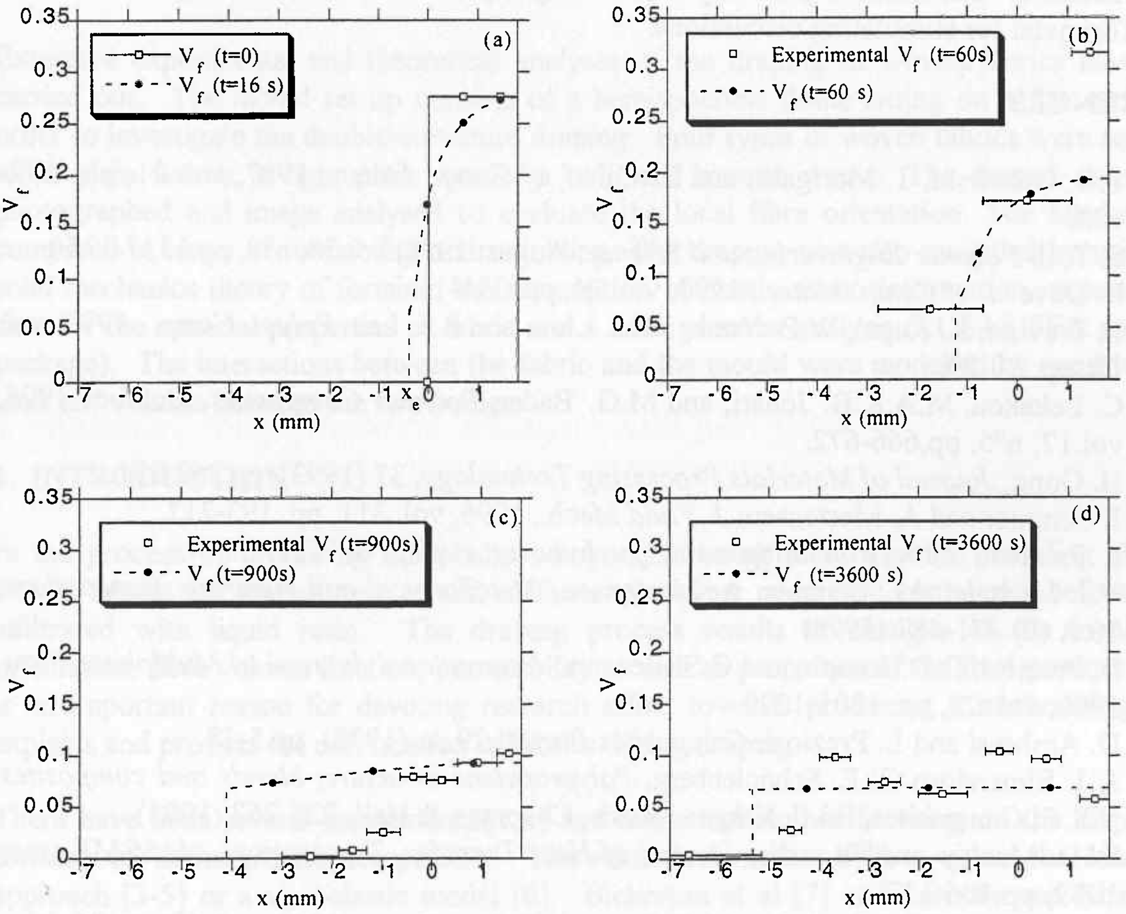

Figure 5. Theoretical and experimental fiber volume fraction distributions at various stages of the impregnation process: (a) initially and at $t=t_i$, (b) after 60s, (c) after 900s, (d) after 3600s.

CONCLUSION

A model is currently being developed to simulate the kinetics of impregnation, and the evolution of the fiber volume fraction profile in liquid composite molding as the resin front progresses, as well as after the front has reached the end of the mold. The predictions for relaxation were validated using a model fluid, polyethylene glycol. The analysis was applied to an industrially relevant system, polypropylene and glass fiber mats used in the production of Glass Mat Thermoplastic blanks. It is shown that the time for preform relaxation in a viscous fluid may be much larger than that for full impregnation. As a result, an apparently well impregnated part may exhibit an inhomogeneous distribution of the reinforcement, in turn inducing a modification of the mechanical behavior and residual stress distribution. The present analysis can therefore be applied to provide guidelines for producing an homogenous composite, or inversely, to produce a graded structure in a controlled manner.

ACKNOWLEDGMENTS

This work is funded by the Fonds National de la Recherche Scientifique under contract n° 20-52625.97. The authors gratefully acknowledge Symalit AG for donating material and M. R. Törnqvist for stimulating discussions.

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