THEORETICAL AND EXPERIMENTAL STUDY OF THE IN-PLANE 1D-FLOW OF PARTICLE-FILLED RESIN THROUGH A FIBROUS PREFORM

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Abstract

The use of particle-filled resin to produce composite parts with a Liquid Composite Molding (LCM) process such as Resin Transfer Molding (RTM) raises the issue of the possible filtration of the particles by the fibrous preform during the impregnation stage. This may results in an uneven distribution of the particles throughout the part or even in dry spots.

The first part of this paper is dedicated to an empirical investigation on the occurence of clogging/no clogging of the preform by the particles during the injection of a composite part. The second part proposes a model of the filtration, based on both theoretical and experimental studies.

Introduction

Liquid Composite Molding (LCM) processes such as Resin Transfer Molding (RTM) or Liquid Resin Infusion (LRI), tend to be more and more employed to produce large and/or complex shaped composite parts. In such technologies, a fibrous preform is first placed in a mould, and then liquid resin is forced through it. High fibre content and good surface finish are some of the advantages of LCM.

In some cases, composite manufacturers add particulate fillers to the resin for different purposes, such as cost reduction or flame resistance improvement. The present study is both restricted to micron scale particles for which filler content may vary from a few percent to 40%vol or more and to the in-plane unidirectionnal flow of the resin through the fibrous preform.

Working with particle-filled resin in LCM processes raises two major concerns during the impregnation stage: viscosity increase of the resin and possible filtration of the particles by the fibrous preform. Both issues contribute to slow down the resin flow for constant pressure driven injection and might be responsible for dry spots, poor saturation of the fiber tows, longer production cycle time or even partially filled composite part. Non-homogeneous distribution of the particles is an additional specific drawback of filtration. However, few researches related to the flow of particle-filled resin through a fibrous preform have been published [1-2].

Filtration phenomena have been well studied in several industrial fields such as water treatment [3-4], oil-well recovery [5-6] or paper industry [7]. Hence, both experimental and theoretical studies have been published on these particular matters.

The first part of this paper is dedicated to an empirical investigation on the occurence of clogging/no clogging of the preform by the partisles during the injection of a composite part. The second part proposes a model of the filtration, based on both theoretical and experimental studies.

1 Empirical approach

1.1 Geometry

Working with particle-filled resin (also named suspension in the following) first raises the question of comparing the filler size with the fibrous preform pore size. Three different behaviours may roughly be distinguished (Fig. 1):

Case I: Particles size is larger than a critical size that depends on the preform characteristics: the particles deposit onto the filter and form an accumulation named *cake.* Thus, the suspension, and therefore particles, will not penetrate into the fibrous preform and *cake filtration* takes place;

Case II: Particles are very small compared to the fibrous preform pore size: there is *no or very little retention* inside the fibrous medium; in this case, atttention should be paid to the suspension viscosity. Indeed, decreasing the particles size leads to a significant increase of the suspension viscosity and this increase may become incompatible with LCM processes.

Case III: Intermediate stage: particles are progressively retained inside the filter media. This phenomenon is called *deep (bed) filtration* and is of key interest in the case of this study. As a matter of fact, the rest of this paper is dedicated to the understanding of deep filtration occurence during the flow of a particle-filled resin through a fibrous preform.

1.2 Literature

Deep filtration theory has been well developed and studied during the past decades. As a first approach, several authors tried to define empirical conditions to predict deep filtration effectiveness.

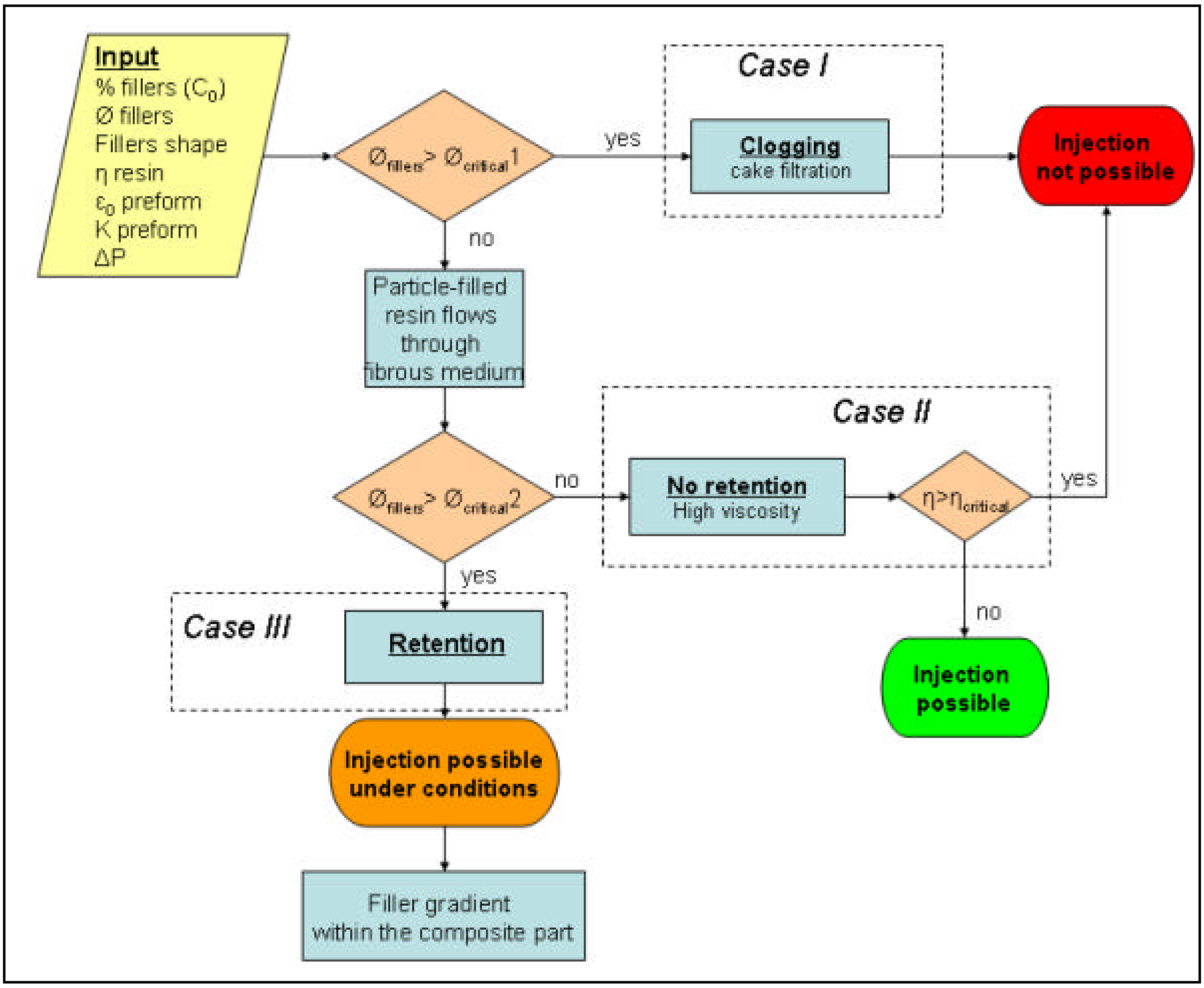

Fig. 1. Flow chart of the particle-filled resin flow through a fibrous preform

Table 1. Empirical criteria for particle retention by the filter from different authors

Authors	**Criteria**	**Observed mechanism**
Sakthivadivel [8]	D/d = 10	Particles do not enter the filter (cake filtration)
	10 = D/d < 20	Regular retention that may lead to clogging (straining in small and large pores)
	D/d > 20	Few retention that quickly reaches an equilibrium (straining in small pores)
Herzig [3]	D/d = 20	Retention in crevice sites (small sites)
	D/d < 12	Straining in constrictions (big pores)
Sherard * [9]	D/d = 8	Retention near the filter surface (cake filtration)
	D/d ˜ 9	Retention
	D/d = 10	No retention

* in Sherard et al. study, the filter grain and suspended particle diameters criteria is D_{15} and d_{85} respectively, where D_{15} means 15% by weight of filter grains are finer than D_{15} and 85% by weight of suspended particles are finer than d_{85}.

These empirical conditions, summarized in table 1, are only based on geometrical considerations, i.e. on the ratio of the filter bed grain size D (in all these previous studies, the filter is composed of grains) to the suspended particle size d, which appears to be the key parameter [8].

Even if the three authors found different values for that ratio, it is clear from table 1 that a boundary between the case where there is no retention and the case of cake filtration can be found in the range of D/d=10-20. These two cases are actually limit cases and deep bed filtration represents an intermediate stage.

The Kozeny-Carman equation (Eq. 1) links the permeability K of a granular bed to the size D of the grains constituting this bed:

$$K = \frac{e_d^{\,3} D^2}{150(1-e_d)^2} \qquad (1)$$

where e_d refers to the bed porosity.

Therefore, it is possible to evaluate the mean diameter of the grains constituting a granular bed, by knowing its permeability and porosity. In this study, the filter bed is made of cylindrical long fibers, but it is easy to measure its permeability and then to calculate the equivalent diameter D_{eq}, i.e. the size of the grains that would constitute a granular bed of the same permeability. From Eq. 1, the equivalent diameter of the fibrous preform is given by:

$$D_{eq} = \sqrt{\frac{150K(1-e_d)^2}{e_d^{\,3}}} \qquad (2)$$

1.3 Experimental investigation

As a very first step to investigate the flow of filled resin through a fibrous preform, it was necessary to determine in which experimental conditions cake or deep filtration occurs, that is to say if the particles can flow through the fibrous preform or not. Several sets of experimental conditions were applied to carry out injection experiments.

1.3.1 Materials

The suspension is constituted of a newtonian fluid (DOP) and spherical microbeads. The fluid has a viscosity of 0.07 Pa.s at room temperature and a density of 0,96g/cm^3. Fillers specifications are described in table 2. When microbeads and DOP are blended, an additive (BYK-W 980, BYK-Chemie, Germany) is used to limit filler sedimentation and aggregation. The additive quantity is 1.5% of the mass of fillers.

Table 2. Particles references and physical properties

Particle average diameter (µm)	**3**	**12**	**48**	**30**
Reference	G200	MP5	MP40	SG2000
Nature	ceramic	glass		
Density (g/cm^3)	2.5	2.46		2.54
Brand name	Zeeospheres®	Microperl®		Spheriglass®
Retailer	3M, France	Sovitec, France		Potters, UK

The preform is made of a polyester fiber mat (PES340, Diatex, France). Its areal weight ranges from 300 to 400 g/m². As it shows discrepancies, synthetic mat weight, and thus fiber volume fraction, is systematically measured before each test.

1.3.2 Experimental setup

The experimental setup is composed of a rigid tooling made of a steel half mold and a thick PMMA top plate. The molding cavity is 90 mm wide, 4 mm thick and 400 mm long. The mold is fed at constant pressure (0.2 MPa) using a pressure pot. The latter is equipped of a motorized mixing device so as to maintain a homogeneous blend of the suspension (Fig. 2).

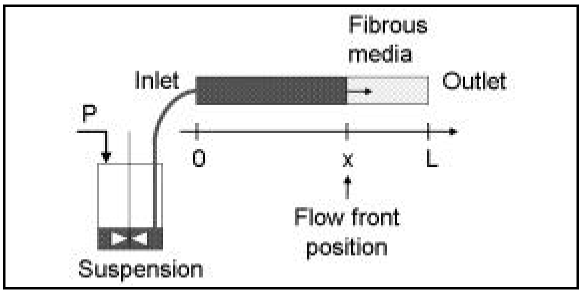

Fig. 2. Injection experiment setup

1.4 Results

1.4.1 Equivalent diameter

Measurements of the clean preform permeability are performed by recording the 1D-flow front position *l* and pressure drop *Dp* of the pure newtonian fluid (without particles) through the fibrous preform as a function of time during mold filling (Fig. 2). Assuming unidirectional flow, Darcy's law is integrated and permeability *K* is given by:

$$K = \frac{(1 - V_f)hl^2}{2t\Delta p} \qquad (3)$$

where V_f is the fiber volume fraction, ? is the fluid viscosity and *t* is the time.

The fibrous preform is supposed to be equivalent to a granular bed of same permeability. The equivalent diameter of the fibrous preform is calculated using Eq. 2. In this equation, the porosity of the equivalent granular bed, assumed to be a random pack of monodisperse spheres, is taken as 0.36. The experimental data are approximated by a power law function. In the case of our study: V_f=18672 $D_{eq}^{-1.0305}$ (Fig.3).

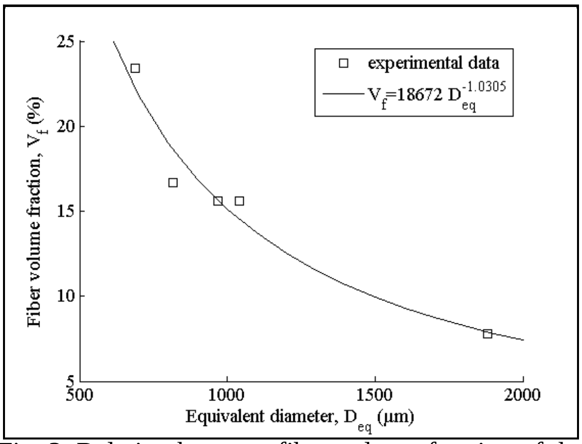

Fig. 3. Relation between fiber volume fraction of the fibrous medium and the equivalent granular bed diameter

1.4.2 Clogging/No clogging criteria

The purpose of this set of experiments is to visually observe whether the fibrous preform is clogged or not by the particles during the flow of the suspension through this preform. For the whole set of experiment, the initial concentration of the suspension is kept constant (C_0=20%vol).

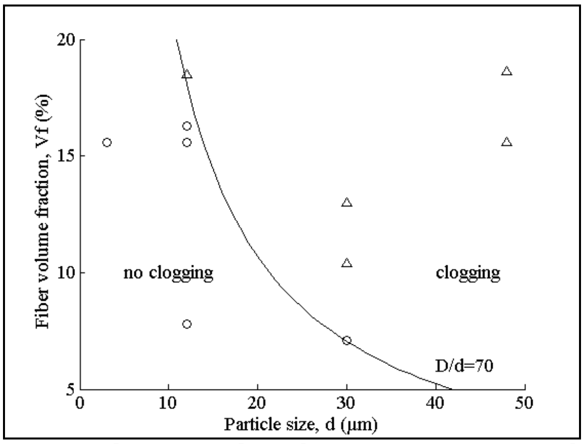

Fig. 4. Clogging and no clogging experiments with respect to fiber volume fraction and particle size (C_0 = 20%vol)

The results are plotted in Fig. 4. For example, the point in the top-right corner means that the experiment was conducted with a suspension of particles of 48µm diameter, the fibrous preform has a fiber volume fraction of 18,6% and the particles accumulate ahead of the preform and clogged it. The clogging/no clogging limit was found to be D/d=70, which appears to be several times greater than ratios reported in the literature for granular beds.

Therefore, this set of experiments shows that further investigations should be led in the no-clogging area, that is D/d<70. In order to maintain the viscosity of the suspension as low as possible, particles with the biggest size in the no-clogging area (Fig.4) are chosen for the rest of the study (12µm).

2 Filtration model

2.1 Introduction

Deep bed filtration is governed by a complex combination of transport and attachment mechanisms that involve physical and chemical properties of the particle, the suspending liquid and the porous medium, on one hand and the operating conditions, on the other hand.

Mathematical models have been formulated in order to predict filter efficiency. Two approaches can roughly be distinguished: macroscopic models, based on a phenomenological approach [1,3], and microscopic models, based on the trajectory analysis [4,10-11].

As the present work belongs to the very first attempts to understand the occurence of filtration in LCM processes, a macroscopic approach was chosen. Thus, experimental validation will be necessary for the determination of the model parameters.

The work of Herzig et al [3] in the field of water treatment served as a basis to this analysis. In the case of composite manufacturing, physical parameters are not in the same order of magnitude (nature of the filter media, filler content, flow velocity, fluid viscosity), so the filtration theory equations need to be derived [1]. A similar approach has been presented by Erdal et al. [1]. They adapted the Resin Transfer Molding (RTM) process used in the organic matrix composites industry to the manufacturing of ceramic-ceramic composites. However, the model they defined predicts monotically decreasing concentration profile, which does not totally agree with what was experienced during the current study.

2.2 Theory

2.2.1 Problem definition

For purpose of simplification, the whole study is reduced to an in-plane 1D problem (Fig. 5). Thus, all parameters are function of time and/or position *x*.

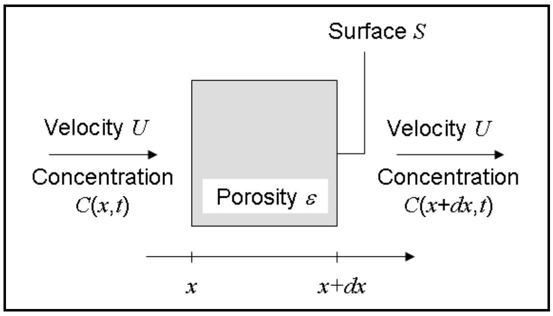

Fig. 5. 1D geometry

As the suspension flows through the element, particles are retained in the fibrous media. This phenomenon is called the *retention* and is expressed by *s*, which is the volume fraction of retained particles in the filter volume. The retention implies that the porosity *e* decreases with time as the filter is clogged by particles, i.e.:

$$e = e_0 - b\,s \qquad (4)$$

where *b* represents the presence of entrapped liquid between the retained particles that does not participate to the suspension flow.

It is also assumed that *b* is not constant during the injection and that it depends on the retention *s* :

$$b = b_0 - r s \qquad (5)$$

where b_0 is the initial value of *b* and *r* is a constant.

2.2.2 Mass balance equation

C(x,t) represents the concentration (in volume) of the suspension at the position *x* and time *t*, i.e. the volume occupied by particles *in the moving suspension*. In addition, it is assumed that diffusion of particles is negligible because of their size (>10µm). The mass balance of particles can be written as:

$$\frac{\partial\left(s + eC\right)}{\partial t} + U\frac{\partial C}{\partial x} = 0 \qquad (6)$$

2.2.3 Kinetic equation of retention

Some authors [3] stated that the particle deposition rate can be written as:

$$\frac{\partial s}{\partial t} = k_0 F(s)UC \qquad (7)$$

Eq. 7 assumes that deposition rate is proportional to the particles present and convected in the suspension, i.e. *UC*. The proportionality coefficient k_0 is called the *initial filtration coefficient*.

Many forms of the retention function $F(s)$ have been proposed so far. See for example [12] for more details. However, for this study, the simple form $F(s)=1$ is chosen.

Therefore, the filtration model is defined by the system of equations 5, 6 and 7. The unknown variables are C, s and b and the parameters of the model are k_0, b_0 and r.

2.3 Experimental

The experimental setup is the same as the one used in the first part of this study (Fig. 2).

The suspension is constituted of unsaturated polyester resin and spherical microbeads. The polyester resin (Crystic 3027 LV, Scott Bader) has a viscosity of 0.2 Pa.s and a density of 1.2 g/cm^3. Spherical glass microbeads (Microperl, Sovitec) are 12 µm mean diameter (Table 2). The same additive as in part 1 is used to limit filler sedimentation and aggregation.

The preform is also the same as in part 1. Such material was chosen because it is completely eliminated after sample burning, so filler content can be easily determined.

During an injection, since the mold top plate is made of PMMA, the flow front velocity can be recorded. That value is required to calculate and input Darcy's velocity during the resolution of the model's equations.

After polymerization, samples are cut out from the composite part at known locations. Samples are weighed, measured and burnt at high temperature (500°C) so that filler content can be evaluated at any distance from the inlet.

Note that only the total quantity of fillers as a function of x can be evaluated by this mean, i.e. $s+eC$, and that it is the values at the final time T of the injection. The experimental conditions are summarized in table 3.

Table 3. Experimental conditions

Experiment	1	2	3
Filler content in the resin (%vol)	21.5	30.3	40.5
Fiber volume fraction (%)	18.1	18	18
Length (cm)	20	20	20
Time (s)	190	320	1065
Filler content targeted in the part (%vol)	17.6	24.9	33.2

2.4 Simulation

2.4.1 Initial and boundary conditions

The model is used to simulate the injections of composite parts made experimentally. At t=0, there are no particles in the domain and thus, for any location within the media, the particle concentration is $C(x,t=0)=0$ and the retention is $s(x,t=0)=0$. The initial porosity is the one of the clean fiber bed i.e. $e(x,t=0)=e_0$.

The boundary conditions are $C(x=0,t)=C_0$ at the inlet and free convective flux at the outlet ($x=L$).

2.4.2 Resolution

Model resolution is performed by implementing the system of equations 5, 6 and 7 in the commercial multiphysics finite element software COMSOL 3.3 (COMSOL AB). The numerical parameters optimization is realized thanks to MATLAB (The Mathworks) minimization routines.

2.5 Results and discussion

All the model's parameters have been identified from experiment 3 (k_0=0.0075 cm^{-1}, b_0=4.54, r=52.29). Then, those parameters have been used to calculate the filler volume fraction of experiments 1 and 2. Experimental and model distributions of particles along the part length are presented in Fig. 6 and show a good agreement.

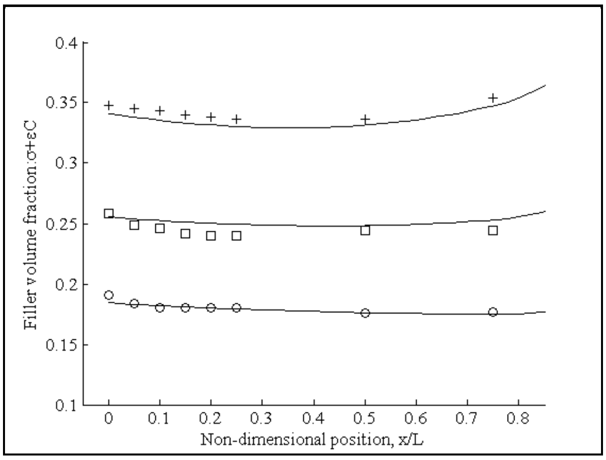

Fig. 6. Filler volume fraction from experiments 1(○),2(□) and 3(+) and model (—)

Near the inlet, the decrease is attributed to filtration. Close to the outlet, the filler concentrations rise, likely due to a suspension depletion mechanism. As the suspension is flowing along the preform, particles are continuously retained by the fibers. A proportional quantity of the suspending liquid is entrapped between the retained particles: this is expressed by *b*. Thus, this quantity of liquid does not belong anymore to the moving suspension so that the suspending liquid fraction decreases, or in other words, the filler fraction increases. Further investigations are on-going to verify this phenomenon of concentration build-up.

Also more experiments will be carried out to measure model parameters (k_0, b_0 and *r*) and relate them to fabric architecture and filler geometries.

Conclusion

The flow of particle-filled resin through a fibrous preform in the case of Liquid Composite Molding processes may be subjected to filtration of the particles by the fibers. As it may affect the composite ultimate properties, this phenomenon is a key issue.

Empirical investigations were performed to identify a simple geometric criterium for the occurence of cake filtration. Hence, the experimental conditions of the study were defined. The experimental setup for measuring particle filtration along the composite part during an injection was presented.

Based on the phenomenological approach first developed for water treatment, filtration equations were derived for the case of the 1-D flow of particle-filled resin through a fibrous preform. A single experiment was necessary to identify all the model parameters. The confrontation between calculation and further experimental data showed a rather good agreement. Investigations are still in progress in order to evaluate the influence of particle size and shape and preform architecture on the model parameters.

Nomenclature

b	Coefficient representing the presence of entrapped liquid between the retained particles that does not participate to the suspension flow
Dp	Pressure drop
e	Porosity of the filter
e_d	Granular bed porosity
e_0	Initial porosity of the filter (clean bed filter)
h	Fluid viscosity
s	Retention i.e. ratio of the volume of deposited particles to the total volume
C	Concentration of the suspension i.e. ratio of the volume of mobile particles to the volume of the suspension
C_0	Inlet concentration of the suspension
d	Particle mean diameter
D	Filter bed grain diamater
D_{eq}	Equivalent diameter
$F(s)$	Retention function
K	Porous medium permeability
k_0	Initial filtration coefficient
l	Flow front position at time t
L	Total length of the fibrous medium
r	Constant
t	Time
T	Filling time
U	Suspension approach velocity (Darcy's velocity)
V_f	Fiber volume fraction of the preform
x	1D coordinate

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