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Modelling the flow of particle-filled resin through a fibrous preform in liquid composite molding technologies

Delphine Lefevre, Sébastien Comas-Cardona^{*}, Christophe Binétruy, Patricia Krawczak

Ecole des Mines de Douai, Polymers and Composites Technology and Mechanical Engineering Department, 941 rue Charles Bourseul, BP 10838, 59508 Douai Cedex, France

During the manufacturing of particle-filled resin composite parts with a liquid composite molding (LCM) process, undesirable issues arise like resin viscosity increase or particles filtration. As the filled resin flow is taking place, the fibrous preform may act as a filter and hinder the even repartition of the fillers throughout the part or even stop the mold filling. The present paper proposes an experimental investigation of the particle filtration during the injection of a composite part. The model proposed by Erdal et al. is analysed and improved in order to take liquid retention phenomenon into account. Finally, simulated and experimental data are compared.

Keywords: Particle reinforcement; Computational modeling; Resin transfer molding; Resin flow; Filtration

1. Introduction

Liquid composite molding processes, such as resin transfer molding (RTM) or liquid resin infusion (LRI) tend to be more and more employed to produce large and complex shaped composite parts. In such technologies, a dry fibrous preform is first placed in a mold and the liquid resin is forced through it. Versatility and good surface finish are some of the advantages of LCM.

In some cases, composites manufacturers use particle-filled resins for different purposes, such as cost reduction or flame resistance improvement. Two classes of particles must be distinguished:

- Nanoscale particles, for which filler content is usually kept low (a few percent up to 20%vol.).
- Micron-scale particles, for example calcium carbonate, for which filler content may vary from a few percent to 40%vol or more.

Working with particle-filled resin (also named suspension in the following) in LCM processes raises two major issues during the impregnation stage, namely *viscosity increase* of the resin and possible *filtration* of the particles by the fibrous preform. Both issues contribute to slow the resin flow and might be responsible for dry spots, poor saturation of the fiber tows or longer production cycle time.

As the filled resin is flowing through the fibrous medium, particles may be retained by the fiber array and filtration takes place. Filler content and, thus, the subsequent properties (e.g., fire resistance) are not homogeneous throughout the composite part, which may become defective. In the most severe conditions when use of high fiber content fabrics is necessary, mold filling may be difficult or impossible. However, few researches related to the flow of particle-filled resin through a fibrous preform has been published [7,10].

Filtration phenomena have been well studied in several industrial fields:

- In water treatment where polluted water flows through a bed of sand or gravel under the action of gravity [16,6].
- In oil well recovery where water is injected in one point of the oil well and carries oil droplets and solid particles to an exit point [2,4].

^{*} Corresponding author. Tel.: +33 (0)3 27 71 21 87; fax: +33 (0)3 27 71 29 81.

E-mail address: comas-cardona@ensm-douai.fr (S. Comas-Cardona).

Nomenclature

β	coefficient representing the presence of entrapped liquid between the retained particles that does not participate to the suspension flow	L	total length of the fibrous medium
β_0	initial value of β	L_{tube}	length of the tube
ε	porosity of the filter	n	constant
ε_0	initial porosity of the filter (clean bed filter)	P	applied pressure
σ	retention, i.e. ratio of the volume of deposited particles to the total volume	q	constant
σ_{max}	maximum value of σ	r	constant
a	constant	R	radius of the tube
b	constant	S	cross section of the filter element
C	concentration of the suspension, i.e. ratio of the volume of mobile particles to the volume of the suspension	S_m	cross section of the mold cavity
C_0	inlet concentration of the suspension	t	time
d	particle mean diameter	t^*	non-dimensional time
$F(\sigma)$	retention function	T	filling time
k	filtration coefficient	u^*	non-dimensional velocity
k_0	initial filtration coefficient	U	suspension approach velocity (Darcy's velocity)
k_r	re-suspension coefficient	U_0	initial suspension approach velocity
		V	interstitial velocity of suspension
		V_f	interstitial velocity at the flow front
		x	1D coordinate
		x^*	non-dimensional coordinate

– In paper industry where fillers are added to improve the printing and mechanical properties of paper [23].

Hence, both experimental and theoretical studies have been published on these particular matters. In the case of composites manufacturing, filler content, flow velocity, fluid viscosity do not belong to the same range, so the filtration theory equations need to be derived [10]. A macroscopic model of filtration during the flow of a particle-filled resin through a fibrous preform has already been proposed by Erdal et al. [10]. However, they were not able to validate a reliable way of characterizing the filtration during impregnation [11]. Hence, their model was not validated by experimental results.

The present paper proposes an experimental investigation of the particle filtration during the injection of a composite part. The model proposed by Erdal et al. is analysed and improved in order to take liquid retention phenomenon into account. Finally, simulated and experimental data are compared. It is emphasized that this study is limited to single scale media. Extension to dual scale media, as usually encountered in composites manufacturing, raises issues that will be presented in the conclusion.

2. Empirical investigations

2.1. Particle-filled resin viscosity

Numerous authors have studied the behaviour of suspension viscosity and in particular the influence of filler content. Suspension viscosity strongly depends on the filler content and increases when the quantity of particles

increases. Thus in some cases, it will not be possible to use LCM process because of a dramatic increase of viscosity.

Actually, the suspension viscosity will increase when:

- The filler content increases ([21,22,13,5]).
- Small particles rather than coarse ones are used [24].
- Irregular shaped, rough surfaced particles rather than smooth spherical ones are employed [18].

In addition, even when the suspending liquid is Newtonian, the suspension often shows a shear-thinning and/or thixotropic behaviour, meaning that viscosity decreases when shear-rate and time increase, respectively [8]. This is particularly true when the filler content is high enough so that particles may interact and form clusters. It must be noticed that the presence of fillers may influence the cure kinetics and viscosity of the suspension to some extent. As given filler content is sometimes required, it may be useful to perform an optimization of the viscosity. A quite simple solution would be to prefer large particles (in the 100–200 μm range), but this may favor filtration since the space between the fiber tows is of the order of 100 μm [1]. Thus, a compromise of particle size will be necessary: on one hand, one should increase the particle size to decrease the suspension viscosity, but on the other hand, one should avoid too large particles otherwise their size may exceed the fibrous preform pore size. Thus, intermediate size particles are required.

Another way to decrease suspension viscosity is to optimize particle size distribution. Indeed, some authors have reported that mixing 25% of “fines” with 75% of “coarse

particles” where the diameter ratio of coarse particles to fines is at least seven provides a rather good optimization [12,20,14]. However, attention is focused on the fact that improvements are only visible for quite high filler content (i.e. 40%vol.).

The use of proper additives is an additional solution but attention should be paid to their influence on resin polymerization kinetics and/or mechanical performances, for example.

Depending on the particle size and the fibrous preform characteristics, different behaviours may be observed. They are presented in the next section.

2.2. Different types of behaviour

Working with particle-filled resin first raises the question of comparing the filler size with the fibrous preform pore size. Three different behaviours may roughly be distinguished:

- **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 *surface 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.
- **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. Indeed, it has been seen in the previous section that ‘intermediate’ size particles are required for purpose of viscosity tailoring. As a matter of fact, the rest of this paper is dedicated to the understanding of deep filtration occurrence during the flow of a particle-filled resin through a fibrous preform.

Actually, the experimental investigations performed in this study revealed two different behaviours in the case of deep filtration, i.e. two types of concentration profile were found. They are described in the next section that also details experimental investigations.

3. Experimental investigations

3.1. Materials

The suspension is constituted of an unsaturated polyester (UP) resin and spherical microbeads. The UP resin (Crystic 3027 LV, Scott Bader) has a viscosity of 0.2 Pa.s at room temperature and a density of 1.2 g/cm³. For fillers, spherical glass microbeads (Microperl, Sovitec) were preferred to irregular shaped particles. Their physical properties are presented in Table 1. When microbeads and resin are blended, an additive (BYK-W 980, BYK-Chemie) is

Table 1
Microbeads physical properties

Reference	MP5	MP40
Volume average diameter (μm)	12	48
Density (g/cm ³)		2.46

used to limit filler sedimentation and aggregation. The additive quantity is 1.5% of the mass of fillers.

The fiber reinforcement used during injection experiments is a synthetic (PET) fiber mat. Such material was chosen for two reasons: it is a single scale porous media and also it is completely eliminated after sample burning, so filler content can be easily determined. 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. The fiber density is 1.39 g/cm³.

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 bucket, equipped with a motorized mixing device so as to maintain a homogeneous blend of the suspension (Fig. 1).

3.3. Injection experiment details

The injection experiment consists in impregnating a preform of length L with the suspension (resin and particles) during a time T . Since the mold top plate is made of PMMA, the flow front velocity can be easily monitored (Fig. 1). That value is required to calculate and input Darcy’s velocity during the resolution of the model’s equations because the injections are realized under constant pressure.

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.

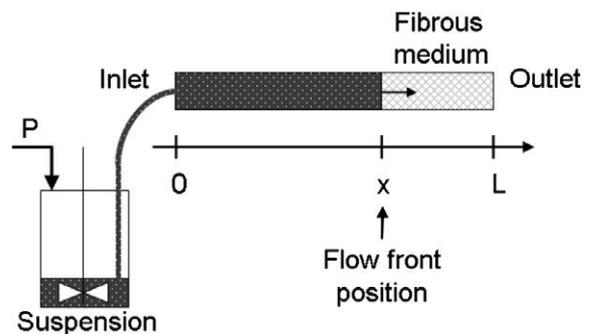


Fig. 1. Injection experiment setup.

The reason for choosing synthetic fiber mat rather than glass fiber mat is that it is virtually impossible to separate properly the glass microbeads from the glass fibers, after sample calcination. On the contrary, synthetic fibers are completely eliminated during samples calcination.

The filler content is calculated as the volume percentage of filler in the sample, that is:

$$\%_{\text{vol}} = \frac{V_{\text{particles}}}{V_{\text{particles}} + V_{\text{resin}} + V_{\text{fibers}}} \quad (1)$$

where $V_{\text{particles}}$, V_{resin} and V_{fibers} are the volume of particles, resin and fibers, respectively, present in the sample. Each volume is linked to the mass by the density, so:

$$V_{\text{particles}} = \frac{m_{\text{particles}}}{\rho_{\text{particles}}}; \quad V_{\text{resin}} = \frac{m_{\text{resin}}}{\rho_{\text{resin}}}; \quad V_{\text{fibers}} = \frac{m_{\text{fibers}}}{\rho_{\text{fibers}}} \quad (2)$$

where $m_{\text{particles}}$, m_{resin} and m_{fibers} are the mass of particles, resin and fibers, respectively, and $\rho_{\text{particles}}$, ρ_{resin} and ρ_{fibers} are the density of particles, resin and fibers, respectively. Densities are known characteristics of the materials. The mass of particles is directly measured after sample burn off, whereas the mass of fibers is evaluated from the sample dimensions and the fiber mat areal weight, which has been measured prior to the injection. The mass of resin is deduced from the difference between the sample total mass and the particles and fibers masses. Thus, the volume percentage of particles in the sample does not account for a possible porosity within the sample. From a practical point of view, filler fraction at a given position x is the mean value of three samples and measure precision is 3% of the nominal value at most (except for experiment 1 where the error is 17% at most).

It should be noted that this test configuration implies that (i) an approximate value of the fibers weight is calculated as it cannot be directly measured, (ii) the porosity is neglected, which might lead to small errors on the filler percentage, and (iii) only the total quantity of fillers as a function of location (x) can be evaluated by this mean (i.e. the particles retained in the medium and the particles suspended in the liquid), and that it is the values at the final time T of the injection.

The experimental conditions are summarized in Table 2. Since injection experiments are conducted with constant pressure, time for complete filling of the mold depends on the length of the preform and the suspension viscosity,

which is governed by the filler content. This explains the different injection times reported in Table 2.

Also, the initial suspension velocity, U_0 , is calculated for each experiment, while assuming that the suspension velocity at the mold inlet equals the one at the exit of the tube connecting the pressure pot and the mold (Eq. (3)):

$$U_0 = \frac{\pi R^4 P}{8 \mu S_m L_{\text{tube}}} \quad (3)$$

where R and L_{tube} are respectively the radius and length of the tube, S_m the mold cavity cross-section, μ the suspension viscosity and P the applied pressure.

3.4. Experimental results

As mentioned previously, two types of concentration profile have been observed. They are illustrated in Fig. 2 where experiments 1 and 2 differ on the particle size and the fiber volume fraction (Table 2).

In the case of experiment 1, concentration profile is strongly and monotonically decreasing. The retention inside the medium is important. This behaviour is conform to what may be expected.

On the contrary, experiment 2 presents small concentration changes. It is also noticeable that concentration is

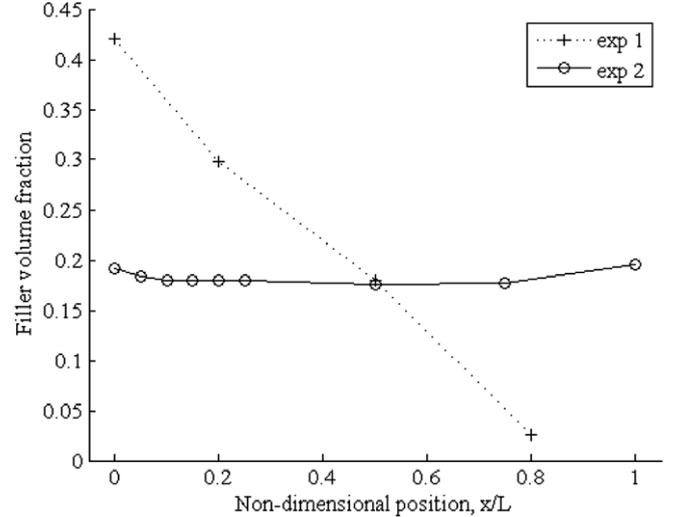


Fig. 2. Effect of particles size and fiber volume fraction on experimental concentration profile in composite parts ($L = 20$ cm).

Table 2
Experimental conditions

Experiment	1	2	3	4	5	6	7	8
Initial filler content in the resin C_0 (%vol)	21.6	21.5	21.8	22.1	30.3	40.2	40.5	40.9
Filler diameter (μm)	48	12	12	12	12	12	12	12
Fiber volume fraction (%)	12.5	18.1	18	18	18	17.9	18	18
Length L (cm)	20	20	30	37	20	15	20	24
Injection time T (s)	320	190	450	645	320	470	1065	1340
Initial suspension velocity, U_0 (cm/s)		1.85	1.93	3.75	0.94	0.36	0.265	1.34
Filler content targeted in the part (%vol)	18.9	17.6	17.9	18.1	24.9	33.0	33.2	33.5

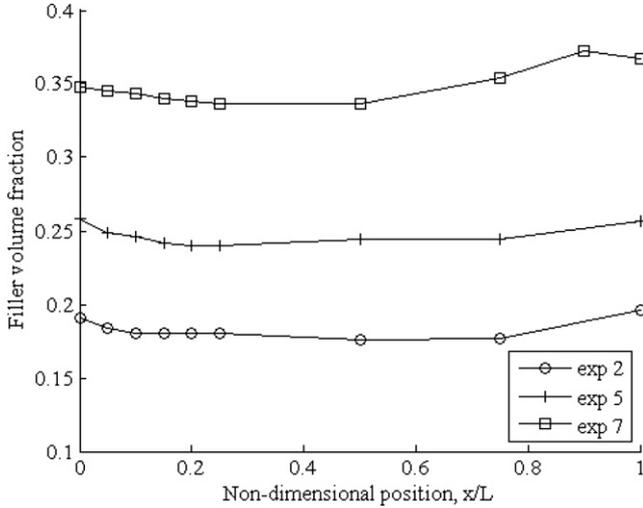


Fig. 3. Effect of the initial suspension concentration on experimental concentration profile in composite parts ($L = 20$ cm).

decreasing at the beginning of the composite part (i.e. near the inlet), and increasing at the end (near the outlet), which was not expected.

Further experiments were performed and confirmed this behaviour. Indeed, Fig. 3 features the experimental results obtained with three different initial suspension concentrations when fiber volume fraction, preform length and particle size are kept identical. They all show a similar U-profile.

It is believed that this phenomenon of concentration build-up is noticeable in the case of experiments 2-5-7 because retention inside the medium is low. On the contrary, in the case of experiment 1, retention is so important that any other phenomenon is negligible.

Since the particles retention inside the preform will change the porosity and, thus, the permeability of the preform, the resin flow will be affected. Therefore, the resin flow simulations based on Darcy's law should be realized including those variations at each time and location. This first experimental investigation is completed by a theoretical study in order to define a filtration model.

4. Theoretical study of deep filtration

4.1. 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 media, on one hand, and the operating conditions, on the other hand. Whatever the particle size, mechanical capture will occur if the particle is bigger than the filter pore size. When the particle is smaller than the pore size, surface interactions become predominant in the case of small particles ($<3 \mu\text{m}$) and physico-chemical capture may then take place. When particles are large ($>30 \mu\text{m}$), mechanical capture is the main mecha-

nism and particles will be entrapped in the filter constrictions [16]. When particle diameter is ranging between 3 and $30 \mu\text{m}$, both mechanisms are expected.

For more details on these mechanisms, see for example [16,19,17].

Mathematical models have been formulated in order to predict filter efficiency. Two approaches can roughly be distinguished:

- Macroscopic models, based on a phenomenological approach were developed until the 70s [16].
- Microscopic models, based on the trajectory analysis have been preferred more recently. This latter approach studies the flow of a particle near a filter grain and calculates its trajectory, by identifying the different forces that affect transport and interception. This work relies on more fundamental predictions but is extensively more complex and difficult to handle (see for example [6]).

As a consequence, the trajectory analysis is more likely to predict the behaviour during the initial stage when the filter bed is clean whereas the phenomenological approach focuses on the transient stage where particles can be removed from the suspension by both filter grain or already retained particles. Though, several studies attempted to take partial deposition or particle size distribution into account and thus to describe the transient stage. For more details on the microscopic approach, readers may refer to [19,17]. It is worth noticing that some authors applied the trajectory analysis to the study of the filtration process by fibrous media instead of granular media as usually encountered in the literature. See [9,15] for more details.

As the present work belongs to the first attempts to understand the occurrence of filtration in liquid composite molding processes, a macroscopic and phenomenological analysis is chosen. Thus, experimental validation will be necessary for the determination of the model parameters.

The work of Herzig et al. [16] serves as a basis for this study but since the equations and the usual simplifications are not directly applicable to our case, governing equations need to be derived. A similar approach has been presented by Erdal et al. [10] in the case of ceramic composites manufacturing. 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 monotonically decreasing concentration profile, which does not totally agree with what was experienced during the current study (Fig. 3). In addition, they were not able to conduct reliable experiments to validate their model [11].

4.2. Problem definition

For simplification, the whole study is reduced to a 1D problem (Fig. 4). Thus, all parameters are function of time and/or axial position.

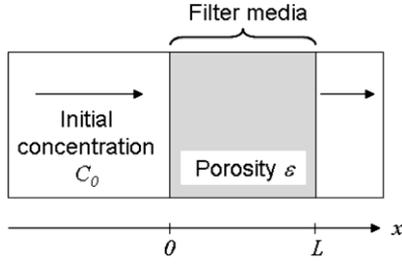


Fig. 4. 1D geometry.

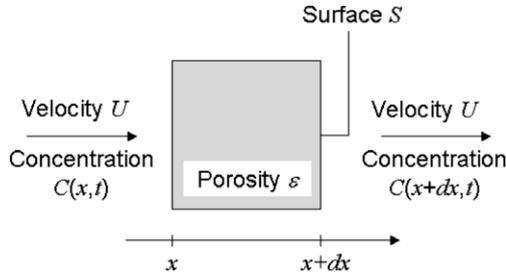


Fig. 5. Filter element.

Let us consider an element of the filter, of length dx and cross-section S , placed at the position x , in which a suspension flows through during an interval of time dt (Fig. 5).

Suspension interstitial velocity is $V(t, x)$ and it varies with the porosity of the filter. It is linked to the Darcy's velocity U , which does not depend on porosity, by Eq. (4):

$$U = \varepsilon V \quad (4)$$

where ε is the porosity of the filter.

Furthermore, when considering the flow front of the suspension, the conservation of the fluid implies for each location x and time t :

$$\varepsilon(x, t)V(x, t) = \varepsilon_0 V_f(t) \quad (5)$$

where ε_0 is the initial (clean fiber bed) porosity and V_f is the flow front velocity, which depends only on the time t . This equation comes from the assumption that there is no air bubbles in the system, so the porous volume available for the suspension flow is exactly ε .

Thus, it is clear from Eqs. (4) and (5) that U only depends on t , as illustrated in Fig. 5.

As the suspension flows through the filter element, particles are retained in the fibrous media. This phenomenon is called the *retention* and is expressed by σ , which is the volume fraction of retained particles in the filter volume. Thus, σ cannot exceed ε_0 . From the *moving* suspension point of view, it may be considered that some of the particles it is constituted of are lost i.e. the particles that are retained, so its local concentration and, thus, viscosity are varying.

It is assumed that the porosity of the filter ε available to the suspension decreases with time as it is clogged by particles, i.e.:

$$\varepsilon = \varepsilon_0 - \beta\sigma \quad (6)$$

where β represents the presence of entrapped liquid between the retained particles or between the retained particles and the fibrous medium. Thus, this quantity of liquid does not participate to the suspension flow. Here again, from the *moving* suspension point of view, since a quantity of liquid is lost, the local concentration and, thus, viscosity of the suspension are varying.

Furthermore, it is also assumed that β is not constant during the injection and that it directly depends on the retention σ :

$$\beta = \beta_0 - r\sigma \quad (7)$$

where β_0 is the initial value of β and r is a constant.

4.3. Continuity 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*. $C(x, t)$ is in the $[0, 1]$ range.

The conservation of particles in the volume $S \cdot dx$ during the time interval dt states (Fig. 5):

incoming particles - exiting particles	=	particles present in the filter	=	retained + suspended particles
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- incoming particles = $C(x, t)USdt$
- exiting particles = $C(x + dx, t)USdt$
- retained particles = $(\sigma(x, t + dt) - \sigma(x, t))Sdx$
- suspended particles = $(\varepsilon(x, t + dt)C(x, t + dt) - \varepsilon(x, t)C(x, t))Sdx$

It is assumed that diffusion of particles is negligible because of their size ($>10 \mu\text{m}$) and sedimentation is also neglected in the mass balance equation.

Thus, the continuity equation can be written as:

$$\frac{\partial(\sigma + \varepsilon C)}{\partial t} + U \frac{\partial C}{\partial x} = 0 \quad (8)$$

Eq. (8) combined with Eqs. (6) and (7) leads to:

$$\varepsilon \frac{\partial C}{\partial t} + U \frac{\partial C}{\partial x} = (\beta_0 C - 2rC\sigma - 1) \frac{\partial \sigma}{\partial t} \quad (9)$$

4.4. Kinetic equation of retention

Some authors [16,10] stated that the particle deposition rate could be written as:

$$\frac{\partial \sigma}{\partial t} = kUC - k_r\sigma \quad (10)$$

Eq. (10) assumes that deposition rate is proportional to the particles present and convected in the suspension, i.e. UC . The proportionality coefficient k is called the *filtration coefficient*. It is assumed that this probability is independent of the suspended particles, i.e. C , but depends on the quantity of retained particles, that is σ . So:

$$k = k_0 F(\sigma) \quad (11)$$

where k_0 is the initial filtration coefficient and $F(\sigma)$ is the *retention function*. The term $k_r \sigma$ in Eq. (10) represents the possibility that retained particles are re-suspended (de-clogging) under the suspension flow.

Then, when combining Eqs. (10) and (11), the kinetic equation of retention is:

$$\frac{\partial \sigma}{\partial t} = k_0 F(\sigma) UC - k_r \sigma \quad (12)$$

4.5. Retention function

Many forms of the retention function $F(\sigma)$ have been proposed. See for example [3] for more details. However, Herzig et al. [16] proposed the following generalized form:

$$F(\sigma) = (1 + a\sigma) \left(1 - \frac{\sigma}{\sigma_{\max}}\right)^q \quad (13)$$

where a and q are constants and σ_{\max} is the maximum value of σ .

Most of the retention functions that have previously been proposed can be described by Eq. (13) with appropriate coefficients a , q and σ_{\max} .

5. Simulation

The model is used to simulate the filtration during the 1D injections of composite parts made experimentally.

5.1. Basic assumptions

In addition to the simplifications presented previously (1D geometry, diffusion and sedimentation neglected), more assumptions are introduced here:

- Since the applied pressure and the flow direction are kept constant throughout the injection, re-suspension of the particles, i.e. the re-suspended term ($k_r \sigma$) in Eq. (12), is neglected.
- A simple form of the retention function has been chosen, as a first approach, that is:

$$F(\sigma) = 1 \quad (14)$$

This form of the retention function implies that the filtration coefficient is constant and equals its initial value. Despite its simplicity, this form of $F(\sigma)$ will prove to be sufficient to obtain good results in the simulation. In addition, this form limits the number of numerical parameters to be identified in the model.

5.2. Non-dimensional model definition

The unknowns of the problem are: C , σ , β and ε . So far, taking into account the previous simplifications, two governing equations (Eqs. (6) and (9)) and two constitutive

equations (Eqs. (7) and (12)) have been expressed and combined in a system of Eq. (15):

$$\begin{cases} \varepsilon = \varepsilon_0 - \beta\sigma \\ \beta = \beta_0 - r\sigma \\ \varepsilon \frac{\partial C}{\partial t} + U \frac{\partial C}{\partial x} = (\beta_0 C - 2rC\sigma - 1) \frac{\partial \sigma}{\partial t} \\ \frac{\partial \sigma}{\partial t} = k_0 UC \end{cases} \quad (15)$$

C , ε and σ are non-dimensional unknown variables, meaning they are in the $[0,1]$ range. To facilitate the solving of the system of Eq. (15), time t , velocity U and x -coordinate are non-dimensionalized. So, the following notations are introduced:

$$u^* = \frac{U}{U_0}; \quad x^* = \frac{x}{L}; \quad t^* = \frac{t}{T} \quad (16)$$

where U_0 is the initial suspension approach velocity, L is the total length of the preform and T is the filling time of the mold. Hence, these new variables also belong to the $[0,1]$ range.

The non-dimensional form of the model becomes:

$$\begin{cases} \varepsilon = \varepsilon_0 - \beta\sigma \\ \beta = \beta_0 - r\sigma \\ \frac{\varepsilon L}{U_0 T} \frac{\partial C}{\partial t^*} + u^* \frac{\partial C}{\partial x^*} = (\beta_0 C - 2rC\sigma - 1) \frac{L}{U_0 T} \frac{\partial \sigma}{\partial t^*} \\ \frac{\partial \sigma}{\partial t^*} = k_0 T U_0 u^* C \end{cases} \quad (17)$$

5.3. 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 $\sigma(x, t = 0) = 0$. The initial porosity is the one of the clean fiber bed i.e. $\varepsilon(x, t = 0) = \varepsilon_0$. However, because of the concentration singularity at $x = 0$ and $t = 0$, the concentration will be gradually raised from 0 towards C_0 , within the first 10^{-2} s of the injection, using a Heaviside function.

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

5.4. Resolution

Model resolution is performed by implementing the following elements in the commercial multiphysics finite element software COMSOL 3.3 (COMSOL AB):

- The system of Eq. (17).
- The input data:
 - initial concentration C_0 ,
 - initial porosity of the fibrous preform ε_0 ,
 - length of the fibrous preform L ,
 - Darcy's velocity U .

- Model's parameters: k_0 , β_0 , r . Those parameters will be determined using numerical optimization from a single experiment.

5.5. Darcy's velocity

Since experiments are realized with constant pressure, it is necessary to determine the Darcy's velocity U during the injection. It follows from Eqs. (4) and (5) that U is determined when the flow front velocity V_f is known. The flow front velocity is experimentally identified by measuring the flow front position at given intermediate times of the experiment. It is assumed that the flow front position x is a power function of time (Eq. (18)):

$$x = bt^n \quad (18)$$

where b and n are numerical parameters.

Thus, the flow front velocity is derived:

$$V_f = nbt^{n-1} \quad (19)$$

Combining Eqs. (4), (5) and (19) leads to:

$$U = \varepsilon_0 nbt^{n-1} \quad (20)$$

Thus, n and b are numerical parameters that do not directly belong to the filtration model presented in this paper. They are only required to get additional modeling information on the flow velocity, which is an input data of the filtration model. Since the target of this work is to be able to simulate an injection of a composite part from the input parameters C_0 , ε_0 and L , a relationship for n and b has to be identified. This will be performed in the next section.

6. Results and discussion

6.1. Empirical determination of n and b

The set of experiments 2–8 has been used to obtain an empirical relationship between the initial concentration of the suspension C_0 and the numerical parameters n and b .

For each experiment, the flow front velocity has been identified to Eq. (19), leading to a set of experimental values (n, b). It appears that n does not depend on C_0 . Its value has been calculated as the average of the value obtained for the experiments: $n = 0.528$. On the contrary, b is a linear function of C_0 : $b = -2.8826C_0 + 1.8071$.

6.2. Determination of k_0 , β_0 and r

The three model's parameters to be identified are k_0 , β_0 and r . It was assumed in Section 5 that they do not depend on the initial suspension concentration C_0 . Hence, a single injection experiment is sufficient to evaluate those parameters. It has been performed through an optimization MATLAB routine on experiment 5. The result is: $k_0 = 0.0075 \text{ cm}^{-1}$, $\beta_0 = 4.725$ and $r = 50$. Since these three

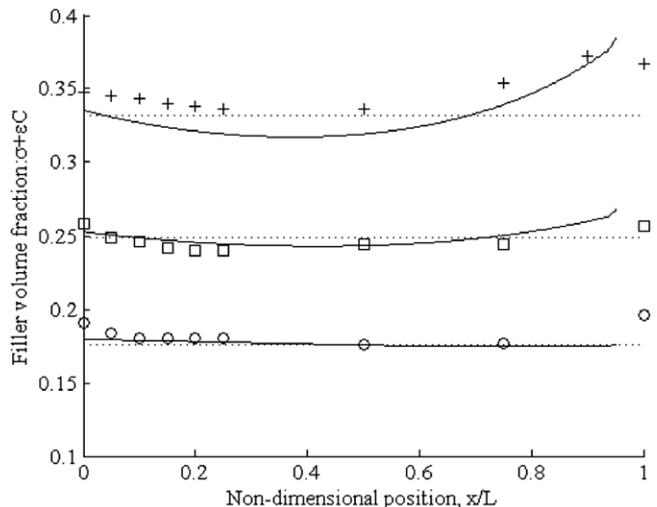


Fig. 6. Experiments 2 (○), 5(□) and 7 (+), target value (- -) and model (—).

parameters are essential for the filtration model, determining them owing to a single experiment is of prime interest.

6.3. Simulation

The latter parameters have been used to simulate composite part injections corresponding to conditions of experiments 2 and 7. The simulated and experimental data of particle distribution along the part length are presented together in Fig. 6.

Concerning experiment 7, the apparent offset between experimental data and simulation may be attributed to the possible error in the filler content calculation mentioned earlier. However, it shows a rather good agreement. In particular the experimental non-linear trends are well described by the simulations.

Near the inlet, the decrease is attributed to filtration. Close to the outlet, the filler concentration rises, likely due to a liquid depletion mechanism in the suspension. 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 β . 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. Researches are still on-going to verify this phenomenon of concentration build-up.

7. Conclusions

When a liquid resin with particulate-fillers is injected into a fibrous preform for the manufacturing of a composite part, two major issues may arise. The first one is the increase of the suspension viscosity, which may result in difficulties to fill the mold properly. The viscosity increase is emphasized by increasing the quantity of filler and/or

by the use of smaller or irregular shaped particles. The second issue is the possible filtration of the particles by the fiber bed. In particular, larger particles than the fibrous preform pore size lead to early clogging of the preform. Therefore, it is necessary to select 'intermediate size' particles: small enough to enter the medium but as large as possible to hinder viscosity increase. Hence, deep filtration may still occur inside the preform. As it may affect the composite ultimate properties, this phenomenon is a key issue.

This study is limited to the case of single scale media. A simple experimental setup was designed, based on a 1D composite part injection. Practical filler profile measurement difficulties were overcome and the final concentration profile along the part length was obtained.

The experimental investigations revealed two distinguished behaviours. In the first case, filler concentration along the composite part strongly decreases showing an important retention of the particles by the fibrous preform. This behaviour was predicted by the model proposed by Erdal et al. [10]. In the second case, filler concentration along the composite part was merely varying compared to the first case mentioned but with a characteristic U-shape curve. The decreasing at the inlet vicinity was attributed to retention whereas the accretion near the outlet was assumed to depend on a liquid depletion mechanism in the suspension in the flow front area. The filtration model was thus improved to take into account these phenomena.

Based on the phenomenological approach first developed for water treatment, filtration equations were derived for the case of the 1D flow of particle-filled resin through a fibrous preform. This system of equations and input data have been implemented in a software using the Finite Elements Method for solving. The model parameters were all identified from a single experiment. The so-determined model was then applied to other experimental conditions and showed a quite good agreement.

In composites parts manufacturing, fibrous reinforcements are usually composed of fiber tows, which are comparable to fiber bundles. Because of textile architecture the distance (few microns) between fibers within the tow and the distance (hundred of microns) between tows create a dual-length scale of porosity. As a matter of fact, the fibrous preform pore size is best represented by a bimodal distribution.

Thus, a set of related questions arises. For example, for specific fire resistance properties, is it more interesting to have fillers dispersed in both inter- and intra-tow spaces or only in the inter-tow spaces? The answer may not be obvious for a given reinforcement and part, but whatever the answer, the particle size should always be compared to the two preform pore scales.

If a monomodal particle distribution is chosen, deep filtration may occur in the inter-tow region whereas surface filtration may take place at the fiber bundle surface. In such a case, the available porosity is reduced to the inter-tow one. However, if deep filtration also occurs in the intra-tows region, meaning that the particle size is quite small,

particles are submitted to sieving. The particle size distribution of the moving suspension is then modified and so is the viscosity. Suspension viscosity model should take into account this variation.

If a bimodal particle size distribution is used, it would impact the suspension viscosity and the filtration mechanisms in both inter- and intra-tow regions.

On a modelling standpoint, the macroscopic model that has been proposed would give insights on a monomodal distribution of fillers in dual-scale reinforcements. However, in the case of multimodal distribution of fillers, the model would not be sufficient to predict the distribution of the multi-size particles in the dual-scale reinforcement. The model should be improved so as to track the fillers in terms of size and location within the porous media (inter- or intra-tow regions).

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