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Modelling flow and filtration in liquid composite moulding of nanoparticle loaded thermosets

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ABSTRACT

This paper presents analytical and numerical models of liquid moulding of hybrid composites. An 1-D analytical solution of Darcy's problem, accompanied by nanoparticle filtration kinetics and conservation, has been developed. A non-linear finite difference model incorporating variations in permeability, porosity and viscosity as a function of local nanoparticle loading was formulated. Comparison of the two models allowed verification of their validity, whilst a mesh sensitivity study demonstrated the convergence of the numerical scheme. The limits of validity of the analytical solution were established over a range of infiltration lengths and filtration rates for different nanoparticle loadings. The analytical model provides an accurate and efficient approximation of through thickness infusion of hybrid composites, whereas use of the numerical scheme is necessary for accurate simulation of in-plane filling processes. The models developed here can serve as the basis of process design/optimisation for the production of hybrid composites with controlled distribution of nano-reinforcement.

1. Introduction

The incorporation of carbon nanoparticles in fibre reinforced composites has become a matter of great interest in the aerospace industry in recent years. The extraordinary electrical, thermal and mechanical properties of carbon nanoparticles combined with the structural and properties of lightweight fibrous composites makes hybrid composites an attractive class of materials. Liquid moulding techniques such as resin transfer moulding (RTM) and vacuum assisted resin transfer moulding (VARTM) are common techniques for the manufacture of fibre reinforced composites. When carbon nanoparticle filled resins are utilised in liquid moulding processes, a stable and homogenous dispersion of the nanoparticles in suspension is paramount for an efficient transfer of their unique properties to the final composite [1,2]. Increasingly higher nanoparticle contents lead to unacceptably high viscosity suspensions for infusion. In addition to the viscosity issues related to high particle content, filtration of particles by the reinforcement can also slow down the resin flow front and lead to long infusion cycles or ultimately to incomplete filling. Particle filtration is a complex phenomenon, which depends on a combination of processing conditions, such as the injection pressure or flow rate and flow direction; as well as the material properties, namely chemical and physical characteristics of the particles, the resin and the porous media. Cake filtration and deep bed filtration are the two main mechanisms occurring during liquid moulding of carbon nanoparticle filled resins. Cake filtration is manifested as volume capture taking place when the particle size is larger than the pore size. Deep bed filtration is characterised by the gradual capture of particles smaller than the pore channels. Continuous capture of particles leads to narrowing of the available pore channels which may ultimately result in cake filtration. Particle size governs the distinct volume and/or surface phenomena taking place during deep bed filtration [3]. Generally, for suspensions containing large particles ($d \geqslant 30\ \mu m$) volume phenomena prevail over surface phenomena; whilst for small particles ($d \sim 1\ \mu m$) surface interactions are predominant; for particle dimensions between 3 μm and 30 μm, both volume and surface phenomena can occur. Other classifications of the filtration mechanisms are based on the ratio between the particle mean diameter and the grain mean diameter of a grain bed [4,5].

In composites processing the objective may be to entrap all the particles in one layer, or achieve an uniform distribution of particles in the composite, or even create a particle concentration gradient characterised by high carbon nanoparticle content in some regions critical to the design/functionality of the component and low content regions in the rest of the part. In all of this potential scenaria a good understanding of the infusion process and the main effects filtration has on process parameters such as viscosity, porosity and permeability is paramount for process design and control.

Deep bed filtration phenomena through porous media have been widely studied in several natural and industrial fields/areas,

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such as oil extraction, wastewater treatment and contaminated ground water flow [6–8]. The modelling of flow of particle filled resins in fibrous media in the manufacture of hybrid composites by liquid moulding has received limited attention up to date. A 2-D Eulerian multiphase approach combined with a control volume finite element model has been used in order to predict the trajectories of spherical carbon nanoparticles in a resin suspension during liquid moulding [9,10]. A 2-D numerical model coupling Stokes–Brinkman laws, accounting for hydrodynamic interactions between the particles and the fibre walls, was utilised to describe the flow in dual-scale porous media during liquid composite moulding [11]. Particle filtration mechanisms were investigated by Nordlund et al. [12] in a resin infusion scenario by velocimetry and microscopy. A stochastic approach, based on the Monte Carlo method has been proposed to simulate liquid filtration of spherical particles through non-woven fibrous media [13]. Macroscopic models of filtration such as the ones developed by Erdal et al. [14] and subsequently enhanced by Lefevre et al. [15,16] couple Darcy's flow with a conservation of mass and filtration kinetics. This type of approach was used in combination with probabilistic methods to determine the particle concentration distribution in the thickness direction of a composite manufactured during VARTM infusion [17]. Recently, a constitutive model approach for filtration was developed in [18], while the filtration constant was determined experimentally as a function of the suspension concentration and the shear rate. Despite the contribution of this experimental methodology, the experimental results did not validate the model.

In the present work an analytical solution for the linear flow of carbon nanoparticle filled resins during liquid moulding of composites is derived. The solution for the concentration of suspended and filtered particles is obtained by coupling Darcy's law with mass conservation and filtration kinetics. In addition a finite difference filling simulation methodology accounting for porosity, permeability and viscosity variations in time and position is implemented for the non-linear case. The two models are compared and the convergence of the numerical model is investigated. The limits of validity of the linear approximation associated with the analytical solution are explored over a wide range of processing conditions.

2. Model development

2.1. Boundary value problem

The physics of the one-dimensional flow and filtration problem are represented by conventional Darcy's law (Eq. (2)) associated with a continuity condition (Eq. (1)) and a particle mass conservation (Eq. (3)) combined with filtration kinetics (Eq. (4)) based on work by Lefevre et al. [15,16]. The suspension Darcy velocity U is driven by the pressure P gradient in the cavity, and is proportional to the permeability over viscosity ratio K/η. The mass balance represented by Eq. (3) accounts for the amount of particles entering and exiting the domain, which corresponds to the total flux of retained and suspended particles, where C and σ represent the concentration of suspended and retained particles, respectively. The concentration time derivative of retained $\partial\sigma/\partial t$ and suspended particles $\partial\varepsilon C/\partial t$, together with the flux of suspended particles along the reinforcement length $U\partial C/\partial x$ contribute to the total balance of particles in the composite at each time step and position. The latter equation neglects both particle diffusion and sedimentation. A constitutive law (Eq. (4)) describes the kinetics of retention and possible re-suspension of particles. The first term of Eq. (4) corresponds to the retention of particles which is proportional to the flux of suspended particles UC. The proportionality constant k_o is called the filtration constant. Any dependence of the filtration constant on the concentration of retained particles was assumed negligible. The second term in the RHS of Eq. (4) represents the rate of particle re-suspension. The latter is considered to be proportional to the product of the concentration of retained particles by the flux of suspended particles and k_r represents the re-suspension constant. The problem described by Eqs. (1)–(4) has four unknowns: the velocity (U), the pressure (P) and the concentration of suspended (C) and retained (σ) particles.

$$\frac{\partial U}{\partial x} = 0 \tag{1}$$

$$U = -\frac{K}{\eta}\frac{\partial P}{\partial x} \tag{2}$$

$$\frac{\partial(\sigma + \varepsilon C)}{\partial t} + U\frac{\partial C}{\partial x} = 0 \tag{3}$$

$$\frac{\partial \sigma}{\partial t} = k_0 UC - k_r \sigma UC \tag{4}$$

A schematic of the 1-D flow and filtration problem is shown in Fig. 1. The resin flow front position (h) is considered equal to zero at the beginning of the filling process. Throughout the injection period, the concentration of suspended particles at the inlet equals the initial concentration of particles in the resin C_0, whilst the retention of particles at the resin flow front position is considered to be equal to zero. The pressure at the flow front position is equal to the vacuum pressure P_∞. Particle re-suspension is considered negligible ($k_r = 0$) since the flow direction is constant during injection.

The boundary condition of the flow problem at the inlet of the flow can be of the first type (Dirichlet), the second type (Neumann) or a combination of the two depending on the control strategy implemented in production. When considering a pressure controlled injection, the pressure at the inlet position corresponds to the injection pressure P_o. In the case of flow control the resin flow at the inlet V_o is kept constant throughout the process. In the case of flow control with a maximum pressure constraint, which is the most realistic condition for an industrial setup, the resin flow is constant at V_o up to the time t_o at which the pressure required to sustain the constant flow exceeds a certain pressure limit P_o. This type of boundary condition is implemented as a complementarity problem. Eqs. (5) and (6) summarise this set of boundary conditions.

$$h(0) = 0, \quad C(0,t) = C_0, \quad \sigma(h(t),t) = 0, \quad P(h(t),t) = P_\infty \tag{5}$$

$$P(0,t) = P_o \tag{6a}$$

$$U(0,t) = \varepsilon V_o \tag{6b}$$

$$(P(0,t) - P_o)\ (U(0,t) - \varepsilon V_o) = 0, \quad P(0,t) - P_o < 0, U(0,t) - \varepsilon V_o < 0 \tag{6c}$$

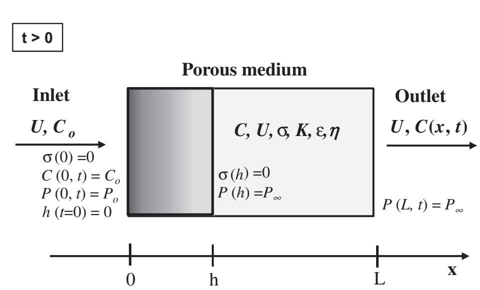

Fig. 1. Schematic of the flow and filtration problem.