Composites Science and Technology 69 (2009) 612–619

Contents lists available at ScienceDirect

Composites Science and Technology

journal homepage: www.elsevier.com/locate/compscitech

Flow of non-Newtonian liquid polymers through deformed composites reinforcements

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ARTICLE INFO

Article history:
Received 19 August 2008
Received in revised form 8 December 2008
Accepted 10 December 2008
Available online 24 December 2008

Keywords:
E. Liquid composites moulding (LCM)
B. Non-Newtonian liquid polymers
B. Permeability
C. Multiscale modeling
C. Computational analysis

ABSTRACT

The flow of non-Newtonian liquid polymers through fibrous reinforcements is a phenomenon which is often encountered during polymer composites manufacturing. In a previous work, we have proposed from a multiscale theoretical approach a method to model this phenomenon when the polymer can be regarded as a generalised Newtonian fluid [Orgéas et al. J. Non-Newtonian Fluid Mech. 2007; 145]. In this paper, the capability of the method is tested with power-law fluids flowing through deformed plain weave fabrics. For that purpose, the flow problem is firstly analysed at the mesoscale from numerical simulations performed on representative elementary volumes of the fabrics. The influences of both the current deformation of the fabrics and the fluid rheology on the macroscopic flow law are emphasised. Secondly, it is shown that the proposed method allows a nice fit of numerical results.

1. Introduction

Understanding, gauging and controlling physical phenomena occurring during the processing of fibre-reinforced polymer composites is crucial to produce optimised structural or functional components with such materials. Among these phenomena, (i) the evolution of fibrous microstructures as well as (ii) the flow of the liquid polymer through the fibrous reinforcements are still not very well understood and require a deeper analysis.

(i) Whatever the considered fibrous reinforcements, *i.e.* impregnated/dry short/long woven/non-woven networks of fibres/fibre bundles, the evolutions of their microstructures induced during the various stages of forming strongly affect their rheology [1–3]. This also significantly changes the possible flow of the liquid polymers through the fibrous networks [4–9].
(ii) The considered polymer matrixes, *i.e.* polymer blends, charged polymers or curing polymers, may exhibit complex rheologies, far from that of the idealised Newtonian fluid model. When non-Newtonian effects become pronounced, significant deviations from the flow of a standard Newtonian fluid through porous media are observed and cannot be neglected [10–15]. In such situations, the well-known Darcy's law is no longer relevant. Unfortunately, if the literature dealing with the flow of Newtonian fluid through fibrous media is abundant, much less is published concerning the flow of non-Newtonian fluids through anisotropic fibrous media, even in the very simple case where liquid polymers or polymer suspensions are assumed to behave, as a first rough but reasonable approximation, as purely non-linear viscous fluids. Consequently, resulting macroscopic models able to describe such macroscale flows are scarce [13,15–18].

In this work, we propose a method to model the second phenomenon at the macroscale, by investigating the impact of the evolution of the fibrous microstructures, *i.e.* the first phenomenon, on the macroscopic flow law. The method is based on a multiscale approach of which the theoretical background has been published in a previous work [18]. For that purpose, the flow problem at the fibre scale is first presented (Section 2). Theoretical results obtained from the upscaling process are briefly recalled (Section 3). A method to build continuous macroscopic flow laws is presented (Section 4). It is based on mesoscale simulations which are carried out on representative elementary volumes of fibrous microstructures. Its capability is here tested with generalised Newtonian fluids, *i.e.* power-law fluids, flowing through pre-sheared plain weaves (Section 5). In particular, the influences of both the current deformation of the plain weaves and the fluid rheology on the macroscopic flow law are emphasized.

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doi:10.1016/j.compscitech.2008.12.007

2. Fluid flow problem at the mesoscale

We consider the slow and isothermal flow of an incompressible generalised Newtonian fluid through a rigid fibrous medium, by assuming a no slip condition at fluid–solid interfaces Γ. The fibrous medium, *e.g.* a textile reinforcement made up of continuous fibre bundles, is seen as an assembly of a large number of identical cells, called Representative Elementary Volume (REV), whose typical size $l_{\rm REV}$ is of the same order of magnitude as l_c, the characteristic thickness of sheared fluid at the heterogeneity scale (here the fibre bundle scale), *i.e.* $l_{\rm REV} = \mathcal{O}(l_c)$. l_c is supposed to be very small compared to the size L_c upon which macroscopic pressure gradients occur. Hence, the scale separation parameter $\varepsilon = l_c/L_c$ is very small, *i.e.* $\varepsilon \ll 1$. Within the REV of volume $\Omega_{\rm REV}$, the flowing fluid occupies a volume Ω_f, whereas fibre bundles occupy a volume Ω_s. For the sake of simplicity, the flow within them is ignored in this study. The stress tensor $\boldsymbol{\sigma}$ of the considered flowing fluids is:

$$\boldsymbol{\sigma} = -p\boldsymbol{\delta} + \boldsymbol{\tau} \quad \text{with} \quad \boldsymbol{\tau} = 2\eta\mathbf{D}, \tag{1}$$

where p is the incompressibility pressure, $\boldsymbol{\delta}$ the identity tensor and where the extra stress $\boldsymbol{\tau}$ depends on both the shear viscosity η and the strain rate tensor $\mathbf{D} = (\boldsymbol{\nabla} \otimes \mathbf{v} + \mathbf{v} \otimes \boldsymbol{\nabla})/2$, $\mathbf{v}$ being the local fluid velocity field. The shear viscosity η is assumed to be a function of the microscopic equivalent shear strain rate $\dot{\gamma}_{eq} = \sqrt{2\mathbf{D}:\mathbf{D}}$, *i.e.* $\eta(\dot{\gamma}_{eq})$. Many well-known rheological models belong to this fluid category: Newtonian fluids, power-law fluids, Cross fluids, Carreau–Yasuda fluids, regularised versions of Bingham or Herschel–Bulkley fluids... From the very simple Newtonian approximation, the other cited models constitute a first step to better account for the complex and non-Newtonian rheology of liquid polymers or polymer suspensions during the processing of polymer composites: they can capture the non-linear influence of strain rates on stress levels required to induce the flow of these complex fluids at finite strain or in steady state situations. Let us note that $\boldsymbol{\tau}$ can also be defined as the gradient of a volumetric viscous dissipation potential Φ:

$$\boldsymbol{\tau} = \frac{\partial \Phi}{\partial \mathbf{D}} = \frac{\partial \Phi}{\partial \dot{\gamma}_{eq}} \frac{\partial \dot{\gamma}_{eq}}{\partial \mathbf{D}} = \tau_{eq} \frac{\partial \dot{\gamma}_{eq}}{\partial \mathbf{D}} = 2\eta\mathbf{D}, \tag{2}$$

where the equivalent shear stress τ_{eq} is defined as:

$$\tau_{eq} = \frac{\partial \Phi}{\partial \dot{\gamma}_{eq}} = \eta \dot{\gamma}_{eq}, \tag{3}$$

so that the volumetric mechanical local dissipation $\mathscr{P}_{dis}$ is expressed as:

$$\mathscr{P}_{dis} = \boldsymbol{\tau} : \mathbf{D} = \tau_{eq}\dot{\gamma}_{eq}. \tag{4}$$

This study is restricted to fluids which verify $d\tau_{eq}/d\dot{\gamma}_{eq} > 0$. This assumption endows the so-defined dissipation potential Φ with the convexity property. It is required to ensure the solution unicity of the localisation problem (7) (see Section 3, [18]). For example, in case of power-law fluids, for which $\eta = \eta_0 \dot{\gamma}_{eq}^{n-1}$ (η_0 being the positive consistency and n the power-law exponent), the last restriction imposes $n > 0$.

Finally, let us point out that when a homogeneous permeation experiment performed with a fibrous sample of length L_c is considered, the present fluid flow problem is driven by a balance between a macroscopic pressure gradient of characteristic value $\Delta p_c/L_c$ and viscous drag forces of characteristic value $f_c = \tau_c/l_c$, τ_c being the characteristic shear stresses induced by the local shearing of the fluid at a charateristic shear rate v_c/l_c [18]:

$$\frac{\Delta p_c}{L_c} = \mathcal{O}(f_c) = \mathcal{O}\left(\frac{1}{l_c}\tau_c\right) = \mathcal{O}\left(\frac{1}{l_c}\eta_c \frac{v_c}{l_c}\right) \quad \text{with } \eta_c = \eta\left(\frac{v_c}{l_c}\right). \tag{5}$$

3. Upscaling process: main results

The above local fluid flow problem was theoretically upscaled in previous studies for Newtonian fluids [19], power-law fluids [20,21], and more recently for generalised Newtonian fluids [18] by using the homogenisation method with multiple scale asymptotic expansions [22]. We briefly recall here the main results deduced from these studies.

3.1. Macroscopic balance equations

The homogenisation process shows that for generalised Newtonian fluids, the mass and momentum balance equations of the macroscopic equivalent continuum associated with the above local physics are, respectively [18]:

$$\begin{cases} \boldsymbol{\nabla} \cdot \langle \mathbf{v} \rangle = 0, \\ \boldsymbol{\nabla}\bar{p} = \mathbf{f}(\langle \mathbf{v} \rangle, \eta, microstructure), \end{cases} \tag{6}$$

where $\boldsymbol{\nabla}\bar{p}$ stands for the macroscopic pressure gradient, $\langle \mathbf{v} \rangle$ is the macroscopic velocity defined as the volume average of the first order component $\bar{\mathbf{v}}$ of the velocity field $\mathbf{v}$, and $\mathbf{f}$ is a macroscopic volumetric viscous drag force depending on $\langle \mathbf{v} \rangle$, the shear viscosity η and the fibrous microstructure.

3.2. First order localisation problem

In order to estimate the macroscopic flow law, *i.e.* the form of $\mathbf{f}$, the macroscopic velocity field $\langle \mathbf{v} \rangle$ must be determined. $\langle \mathbf{v} \rangle$ can be obtained by determining the first order periodic velocity field $\bar{\mathbf{v}}$ in a given REV, by solving the following localisation problem resulting from the homogenisation process [18]:

$$\begin{cases} \boldsymbol{\nabla} \cdot \bar{\mathbf{v}} = 0 & \text{in } \Omega_f, \\ 2\boldsymbol{\nabla} \cdot \left(\eta(\dot{\bar{\gamma}}_{eq})\bar{\mathbf{D}}\right) = \boldsymbol{\nabla}\bar{p} + \boldsymbol{\nabla}\delta p & \text{in } \Omega_f, \\ \bar{\mathbf{v}} = \underline{0} & \text{on} \Gamma, \end{cases} \tag{7}$$

where the macroscopic pressure gradient $\boldsymbol{\nabla}\bar{p}$ acts as a constant and given volume force in the whole REV, and δp is the first order periodic fluctuations of the pressure field around $\bar{p}$.

3.3. Properties and forms of the macroscopic flow law

When the flowing fluid is Newtonian, *i.e.* when $\eta = \eta_0$, it can be shown that the macroscopic flow law reduces to the well-known Darcy's law [23,19]:

$$\mathbf{f} = -\eta_0 \mathbf{K}^{-1} \cdot \langle \mathbf{v} \rangle, \tag{8}$$

where $\mathbf{K}$ is the definite, positive and symmetric permeability tensor. When considering a power-law fluid, the linear Darcy's law (8) is not valid any more. However, it can further be proved that $\mathbf{f}$ is a homogeneous function of degree n of the average velocity $\langle \mathbf{v} \rangle$ [21]:

$$\mathbf{f}(\xi\langle \mathbf{v} \rangle) = \xi^n \mathbf{f}(\langle \mathbf{v} \rangle), \ \forall \xi \in \mathbb{R}^+. \tag{9}$$

For other generalised Newtonian fluids, relations (8) and (9) are no more satisfied. Nonetheless, it can be shown that the macroscopical drag force $\mathbf{f}$ is the gradient, with respect to $\langle \mathbf{v} \rangle$, of the volume averaged viscous dissipation $\langle \Phi \rangle$ [18]:

$$\mathbf{f} = -\frac{\partial \langle \Phi \rangle}{\partial \langle \mathbf{v} \rangle}. \tag{10}$$

As shown by (2), the viscous dissipation potential Φ can be expressed at microscopic scale as a function of $\dot{\gamma}_{eq}$. Similarly, the macroscopic dissipation potential $\langle \Phi \rangle$ can be expressed as a function of a macroscopic equivalent velocity v_{eq}, defined as a norm in the velocity space and depending on $\langle \mathbf{v} \rangle$: