DERIVATION OF CONSTITUTIVE EQUATIONS FOR ROD SUSPENSIONS IN GENERALISED NEWTONIAN FLUIDS BASED ON A CELL MODEL

J. Férec[1*], E. Bertevas[2], B.C. Khoo[2], G. Ausias[1], N. Phan-Thien[2]

[1] *Institut de Recherche Dupuy de Lôme (IRDL), Univ. Bretagne Sud, FRE CNRS 3744, F-56100 Lorient, France.*
[2] *Department of Mechanical Engineering, National University of Singapore, Singapore 119260, Singapore.*
*Corresponding author (julien.ferec@univ-ubs.fr)

Keywords: *Rod suspension; Cell model; Power-law fluid, Ellis Fluid, Bingham fluid; Constitutive law; Rod orientation.*

Introduction

Suspensions of rod particles in liquids are encountered in several natural and engineered products, with particle size ranging from microscopic (e.g., glass and flax fibers), down to nanoscopic scale (e.g., cellulose nanofibers and carbon nanotubes). Usually, these particles with a length *L* and a diameter *D* exhibit large aspect ratio, that is $a_r = L/D \gg 1$. Such systems represent an important class of non-Newtonian fluids but most of the work found in the literature has focused on the study of rod-filled Newtonian liquids when many unfilled matrices exhibit non-Newtonian behavior. Hence, the current understanding of the effect of the presence of rods on the composite rheological properties remains incomplete.

Constitutive equations for fiber-filled systems may generally be written by considering them as two-component fluids, in which the total stress in the composite is assumed to be the sum of the matrix contribution and the particle contribution, $\boldsymbol{\tau}_p$. When dealing with a Newtonian medium of viscosity η_0 and up to moderate rod volume concentration ϕ, the particle contribution to the extra stress tensor, $\boldsymbol{\tau}_p$, takes the following general form [1]:

$$\boldsymbol{\tau}_p = \eta_0 \phi [\mu_1 \boldsymbol{a_4} : \dot{\boldsymbol{\gamma}} + \mu_2 (\boldsymbol{a_2} \cdot \dot{\boldsymbol{\gamma}} + \dot{\boldsymbol{\gamma}} \cdot \boldsymbol{a_2}) + \mu_3 \dot{\boldsymbol{\gamma}} + 2\mu_4 \boldsymbol{a_2} D_r] \tag{1}$$

where $\dot{\boldsymbol{\gamma}}$ is the deformation rate tensor. $\boldsymbol{a_2}$ and $\boldsymbol{a_4}$, the second- and fourth-order orientation tensors [2], describe the rod orientation state in an efficient and concise way without significant loss of information. The coefficients $\{\mu_i, i = 1, 2, 3, 4\}$ in Eq. (1) are geometric shape factors, and D_r is the rotary diffusivity due to Brownian motion. For slender rods, particle thickness can be ignored (i.e., $a_r \gg 1$) and this is achieved by setting μ_2 and μ_3 equal to zero. If the particle is large enough so that Brownian motion can be ignored, the last term containing D_r can also be omitted. The rod dynamics are based on the Jeffery result [3], who solved the creeping flow equations for a freely suspended, rigid ellipsoid in an infinite Newtonian fluid and can be extended to slender particles.

In order to consider the effect of non-Newtonian matrices, current models consider the particle contribution to the extra stress tensor, $\boldsymbol{\tau}_p$, to simply be obtained by replacing the Newtonian viscosity in Eq. (1) by that of the matrix polymer $\eta_0 = \eta^m(\dot{\gamma}, t)$, which can be shear rate-dependent, time-dependent or both.

In this regard, the open questions are: do rod suspensions behave differently in a non-Newtonian matrix, as compared to a Newtonian one, and does the non-Newtonian character of the suspending fluid changes components to the particle stress tensor? These are some of the questions we wish to address in this work.

Model developments

The generalized Newtonian fluid (GNF), which results from a minor modification of the Newtonian fluid law, incorporates the idea of a shear-rate-dependant viscosity and therefore can describe some non-Newtonian viscosity curves. However, it cannot exhibit normal stress effects or time-dependant elastic effects. Amongst GNF, the power-law model describes a viscosity with a function that is proportional to some power of $\dot{\gamma}$. Hence, to tackle the issue of this suspending fluid describing a shear-thinning behaviour (defined by a consistency K and a power-law index m), a cell model is used to derive

constitutive equations for rod suspensions in this kind of fluid. The extra stress tensor, $\boldsymbol{\tau}_p$, is found to be [4]:

$$\boldsymbol{\tau}_p = K\phi\mu_1^{(m)}\dot{\boldsymbol{\gamma}}:\int_p \boldsymbol{pppp}\,|\dot{\boldsymbol{\gamma}}:\boldsymbol{pp}|^{m-1}\psi_p\,d\boldsymbol{p} \tag{2}$$

where $\mu_1^{(m)}$ is the coupling coefficient, which is a function of a_r and m. Note that if m tends to 1, Eq. (2) recovers the form of Eq. (1) and reduces to the initial expression proposed by Dinh and Armstrong [5]. It is found that the shear-thinning character of the matrix influences considerably the rod contribution to the stress tensor, but has no impact on the rod orientation dynamics: the same microstructure evolution as the one encountered in Newtonian fluids is obtained. The rod suspension behaves differently from the unfilled matrix in the sense that depending on rod orientation, the onset of shear-thinning of the composite occurs at lower or higher shear rates. This approach has been extended to investigate non-Newtonian viscous matrices such as Ellis and Carreau fluids [4]. It provides a semi-analytical model for rod suspensions in Ellis fluid, which appears to be suitable in predicting a Newtonian plateau at low shear rates and a shear-thinning behavior at high shear rates. In addition, the model predictions are in good agreement with shear viscosity measurements of glass fiber-filled polystyrene melts [6], demonstrating its ability to describe the rheological behavior of such polymer composites.

To address the case of a Bingham fluid (with a yield stress τ_0 and a constant plastic viscosity η_B), again the cell model allows expressing the shear stress on the particle surface, and therefore the following particle stress contribution is derived for a Bingham numbers less than unity:

$$\boldsymbol{\tau}_p = \eta_B\phi\mu_1\boldsymbol{a_4}:\dot{\boldsymbol{\gamma}} + \tau_0\phi\mu_B\boldsymbol{a_2} \tag{3}$$

where μ_B is a coefficient exhibiting a dependence on a_r and the cell size. It is found that the presence of rods leads to an increase in the yield stress values, i.e., the last term in Eq. (3) and more significantly, it is related to the average rod orientation. Hence, the effect of rod orientation on the yield stress surface is investigated by considering the response under a shear/elongational flow. It appears that the shear yield stress for randomly oriented rods is lower than the one for the case where all particles are aligned with the flow direction. The model predictions are also tested against shear stress experiments for kaolin pastes filled with steel fibers having two different aspect ratios. Good agreement is observed since the model predictions give qualitative and quantitative bounds for yield stresses of both suspensions.

References
[1]   Hinch, E.J., & Leal, L.G. Constitutive equations in suspension mechanics. Part 1. General formulation. *Journal of Fluid Mechanics*, 71: 481–495, 1975
[2]   Advani, S.G., & Tucker, C.L. The use of tensors to describe and predict fiber orientation in short fiber composites. *Journal of Rheology*, 31: 751–784, 1987
[3]   Jeffery, G.B. The motion of ellipsoidal particles immersed in a viscous fluid. *Proceeding of the Royal Society A*, 102: 161–179, 1922
[4]   Férec, J., Bertevas, E., Khoo, B.C., Ausias, G. & Phan-Thien, N. The effect of shear-thinning behaviour on rod orientation in filled fluids. *Journal of Fluid Mechanics*, 798: 350–370, 2016
[5]   Dinh, S.M. & Armstrong, R.C. A rheological equation of state for semiconcentrated fiber suspensions. *Journal of Rheology*, 28: 207–227, 1984
[6]   Chan, Y., White, J.L. & Oyanagi, Y. Fundamental study of the rheological properties of glass-fiber-reinforced polyethylene and polystyrene melts. *Journal of Rheology*, 22: 507–524,1978
[7]   Férec, J., Bertevas, E., Khoo, B.C., Ausias, G. & Phan-Thien, N. A rheological constitutive model for semiconcentrated rod suspensions in Bingham fluids. *Physics of Fluids*, 29: 073103, 2017
[8]   Férec, J., Perrot, A., & Ausias, G. Toward modeling anisotropic yield stress and consistency induced by fiber in fiber-reinforced viscoplastic fluids. Journal of Non-Newtonian Fluid Mechanics. 220: 69–76, 2015