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Steady-shear rheological properties for suspensions of axisymmetric particles in second-order fluids

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ABSTRACT

Following Leal who gave the motion of a slender axisymmetric rod in a second-order fluid, we derived a complete rheological constitutive equation for dilute and semidilute slender rod suspensions in a viscoelastic solvent based on a cell model. Numerical solutions for the Fokker–Planck equation are obtained for simple shear flows at low and large Peclet numbers using a finite volume method, hence avoiding the need for closure approximations. The second normal stress difference coefficient of the solvent plays a non-negligible role in the particle contribution to the stress as well as on the rod orientation dynamics: a spread of the particle orientation in the flow-vorticity plane and an enhancement of the alignment along the vorticity direction are predicted when increasing the second normal stress difference coefficient. Brunn extended the Leal analysis to dumbbells and tri-dumbbells, for which both normal stress difference coefficients have to be considered. The original Pipkin diagram is finally modified to help guide the choice of relevant constitutive equations for particles in viscoelastic fluids.

1. Introduction

The rheological characterization of rod-filled media is of major concern to many industries, such as printing and papermaking, petroleum, polymer processing, aerospace, bioengineering, pharmaceutical industry, construction, ceramics, food, etc. Indeed, the behavior of the suspension is usually significantly different from that of the suspending fluid. The orientation distribution of rods induced by the flow field strongly influences certain macroscopic physical properties such as the rheological behavior of the suspension, which itself governs the flow pattern. A large body of work in the literature has focused on the study of rod-filled Newtonian liquids, in which the rheological effects and the orientation evolution of the rods are described [1,2]. Despite the fact that almost all solvents used in the industry are viscoelastic by nature, the understanding and especially the modeling of the rheological behavior of rod-filled viscoelastic media remain a formidable challenge. Due to their complexity, only a limited amount of studies has attempted to embark on such an endeavor.

Constitutive equations for rod filled viscoelastic systems may generally be considered as a two-component fluid, in which the total stress of the composite can be assumed as [3]

$$\boldsymbol{\sigma} = -P\boldsymbol{\delta} + \boldsymbol{\tau}^m + \boldsymbol{\tau}^p, \quad (1)$$

where P is the isotropic pressure, $\boldsymbol{\delta}$ is the identity tensor, $\boldsymbol{\tau}^m$ is the matrix contribution and $\boldsymbol{\tau}^p$ is the particle contribution to the extra stress tensor.

1.1. Newtonian suspending fluids

When dealing with a Newtonian solvent of viscosity η_0, the particle contribution to the extra stress tensor ($\boldsymbol{\tau}^p$) at low rod volume fraction ϕ takes the following general form [4]

$$\boldsymbol{\tau}^p = \eta_0\phi\left[\mu_1\mathbf{a_4} : \dot{\boldsymbol{\gamma}} + \mu_2\left(\dot{\boldsymbol{\gamma}}\cdot\mathbf{a_2} + \mathbf{a_2}\cdot\dot{\boldsymbol{\gamma}}\right) + \mu_3\dot{\boldsymbol{\gamma}} + 2\mu_4\mathbf{a_2}D_r\right], \quad (2)$$

where $\dot{\boldsymbol{\gamma}}$ is the deformation rate tensor. $\mathbf{a_2}$ and $\mathbf{a_4}$ are respectively the second- and fourth-order orientation tensors [5] which are commonly used to describe the average rod orientation state in an efficient and concise way, without any significant loss of information. The coefficients $\{\mu_i, i = 1, 2, 3, 4\}$ in Eq. (2) are geometric shape factors (see Table 1 in [2]), and D_r is the rotary diffusivity due to Brownian motion. For slender rods, particle thickness can be ignored and this is achieved by setting μ_2 and μ_3 equal to zero. If the particles are large enough so that Brownian motion can be ignored, the last term containing D_r can be omitted. For instance, Sepehr et al. [6] have theoretically checked this assumption for short glass fiber suspensions, where the particle aspect ratio is close to 20. Once μ_2, μ_3 and μ_4 (or equivalently D_r) are set

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to zero and μ_1 is suitable chosen, Eq. (2) reduces to the expression of Dinh and Armstrong [7], where the particle thickness has been neglected in the derivation. Three regimes of rod concentrations related to characteristic particle dimensions are proposed in the literature [8]: dilute, for which $\phi < D^2/L^2$; semidilute $D^2/L^2 < \phi < D/L$ and concentrated $\phi > D/L$, where L and D are respectively the length and the diameter of the particle.

Particle motion in a Newtonian fluid was investigated theoretically by Jeffery [9], who solved the creeping flow equations for a rigid ellipsoid freely suspended in an infinite Newtonian fluid. In a simple shear flow, Jeffery's solution shows that the particle center translates with the local fluid velocity and rotates in a time-dependent periodic orbit about the vorticity axis of the flow [see Eq. (6)]. Bretherton [10] indicates that the period of rotation for any axisymmetric particle is given by $T_r = 2\pi(a_r + a_r^{-1})/\dot{\gamma}$, where $a_r = L/D$ is the particle aspect ratio and $\dot{\gamma}$ is the applied bulk shear rate. Note that Jeffery's theory is supported by extensive experimental results [11–13].

The orientation dynamics of a population of rods is commonly modeled through a time evolution equation of the second-order orientation tensor. This requires the use of closure approximations to express higher order orientation tensors [14–17]. For non-dilute rod suspensions in Newtonian fluids, most theories make use of the following expression

$$\frac{D\mathbf{a_2}}{Dt} = -\frac{1}{2}(\boldsymbol{\omega}\cdot\mathbf{a_2} - \mathbf{a_2}\cdot\boldsymbol{\omega}) + \frac{\lambda}{2}\left(\dot{\boldsymbol{\gamma}}\cdot\mathbf{a_2} + \mathbf{a_2}\cdot\dot{\boldsymbol{\gamma}} - 2\mathbf{a_4}:\dot{\boldsymbol{\gamma}}\right) + 2D_r\left(\boldsymbol{\delta} - 3\mathbf{a_2}\right), \quad (3)$$

where $\boldsymbol{\omega}$ is the vorticity tensor, $\lambda = (a_r^2 - 1)/(a_r^2 + 1)$ is a shape factor and D/Dt denotes the material derivative. The first two terms on the left-hand side of Eq. (3) represent the hydrodynamic contribution derived from the Jeffery's equation and are valid for dilute suspensions of ellipsoids in a Newtonian fluid at low Reynolds numbers. In order to describe concentrated non-Brownian particle suspensions, Folgar and Tucker [18] suggested modeling particle-particle interactions by means of $D_r = C_I|\dot{\gamma}|$, where C_I is an interaction coefficient [19,20] and $|\dot{\gamma}|$ is the effective deformation rate.

1.2. Viscoelastic suspending fluids

With the prospect of modeling phenomena in composite processing, the general form of the constitutive equation cited above has been extended in various manners to include the effect of the viscoelastic polymer matrix on the suspension behavior. Except for a few studies in which rods are omitted and therefore the whole suspension is treated as a homogeneous viscoelastic fluid [21], the particle contribution to the extra stress tensor, $\boldsymbol{\tau}^p$, is simply obtained by replacing the Newtonian viscosity in Eq. (2) by that of the matrix $\eta_0 \equiv \eta^m(\dot{\gamma}, t)$, which can be shear rate-dependent, time-dependent or both. As for the evolution equation of $\mathbf{a_2}$, the expression in Eq. (3) is used without modification.

Fan [22] derived a constitutive equation in the general framework of phase-space kinetic theory. In this study, the suspending fluid was assumed to behave as an Oldroyd-B fluid. Assuming that polymer chain motion was more strongly hindered in a direction crosswise to the rod axis compared to the lengthwise direction, interactions between fluid and rods were modeled by means of an anisotropic resistance coefficient [23,24]. Azaiez [3] used the kinetic theory of elastic dumbbells and a rod orientation-dependent friction factor to develop constitutive equations for fiber suspensions in polymer solutions based on the FENE-P (Finitely Extensible Non-linear Elastic - Peterlin), FENE-CR (Finitely Extensible Non-linear Elastic - Chilcott and Rallison), and Giesekus models. Ait-Kadi and Grmela [25] assumed that the viscoelastic matrix behavior is governed by a second-order conformation tensor and obtained its time-evolution equation from the generalized Poisson bracket formalism. Their choice for the Helmholtz free energy function yields a FENE-P type viscoelastic matrix. This work was then extended by Ramazani and co-authors [26,27], who introduced fiber-matrix interactions through anisotropic expressions for the mobility tensor. A similar approach was adopted by [28] to establish a rheological model for semi-flexible fiber suspensions in polymeric fluids described by a FENE-P model. Beaulne and Mitsoulis [29] used the K-BKZ integral constitutive equation with multiple relaxation times as proposed by [30] for the polymer matrix. Some authors [31,32] applied the multi-mode Giesekus model [33] to predict the strain rate-dependent viscoelastic behavior of the polymer matrix.

Nevertheless, none of the theories cited above considered the effects of the normal stress differences exhibited by the viscoelastic matrix. In view of the state of current approaches, the questions remaining open as to what should models include are: do rod suspensions behave differently in a viscoelastic matrix as compared to a Newtonian matrix, and does the suspending fluid elasticity contributes additional components to the particle stress tensor? In most of the previous studies, the rod orientation dynamic is based on the Jeffery's equation, which was derived for Newtonian fluids. What would be the effect of elasticity on fiber orientations?

Recently, D'Avino and Maffettone [34] compiled an exhaustive literature review on particle dynamics in viscoelastic liquids. Numerical simulations of the motion of spherical and ellipsoidal particles in viscoelastic liquids are addressed as well as some experimental results. Pioneer experimental work was carried out by Saffman [35], who observed that rods immersed in a non-Newtonian fluid undergoing Couette flow align along the vorticity axis. The same conclusions were reached over several decades by Mason and coworkers [36–39], by Fuller and coworkers [40,41] using linear dichroism measurements, by Iso and co-authors [42,43] for weakly and highly elastic fluids, and by Gunes et al. [44], who coupled rheo-optical methods and flow microscopy to analyze the dynamics of spheroidal particles. However, no complete model was clearly identified as only few theoretical studies deal with the behavior of rods in a viscoelastic fluid.

A leading modeling study was conducted by Leal [45], who derived the motion of a slender rod in a second-order fluid (SOF) undergoing simple shear flow. The velocity field produced by the rod was expressed as a perturbation from the Newtonian flow solution. As compared to the Jeffery's solution, the particle still translates with the local undisturbed velocity of the suspending fluid but its orientation time evolution involves the second normal stress difference coefficient of the fluid [Eq. (7)]. Later, Brunn [46,47] derived analogous equations for rigid tri-dumbbells [Eq. (8)] and 1st-order dumbbells [Eq. (9)], for which the condition of a non-zero second normal stress difference can be relaxed. Harlen and Koch [48] considered an Oldroyd-B fluid and showed that the first normal stress difference is responsible for the particle alignment along the vorticity axis. Hence, all these theories predict a particle drift towards the vorticity axis, instead of following a closed orbit as suggested by Jeffery's analysis in Newtonian fluids. This drift appears to arise from the normal stress differences of the fluid.

Despite good quantitative agreements between Leal's theoretical predictions and experimental results, few models have emerged to describe the rheological behavior of such systems. The orientation distribution of rods in a SOF in simple shear flow has been analyzed at the asymptotic limit of weak [49] and strong [50] Brownian diffusion. In the latter, the Fokker–Planck equation is solved for the near-equilibrium conditions by using spherical harmonics and truncating the resulting infinite series. The non-Newtonian properties of the fluid result in a narrower distribution of particles near the shear plane, but cause a spread in the orientation in the flow direction due to the drift towards the vorticity axis. Using a finite