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Modeling Orientational Diffusion in Short Fiber Composite Processing Simulations

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Abstract: Numerical simulations of fiber orientation in short-fiber composites have relied on the Folgar and Tucker (1984) model for diffusion for over twenty years. Unfortunately, it has recently been shown that this fiber collision model tends to over-predict the rate of alignment; exposing the need for a new fundamental approach to more accurately capture fiber interactions within the melt flow. Here we present our initial work in the development of an objective directional diffusion model and a variable lambda model for fiber collisions where we modify Jeffery's model (1922) to incorporate local directionally dependent effects assumed proportional to the probability of fiber-fiber collisions. We show that our directional diffusion model performs well in extensional flows, whereas its usefulness appears limited in shearing flows. Conversely, preliminary results from the variable lambda model in both elongational and shearing flows are quite promising and will be the focus of future investigations.

1. Introduction: Among published short fiber orientation distribution simulation approaches, few enjoy the widespread acceptance of the Folgar and Tucker model [1] which forms the basis for most industrial fiber orientation simulations (see, for example, [2–5] for applications). Recent experiments (see e.g. [6–14]), however, have exposed previously unknown limitations of the Folgar-Tucker model, raising questions on the appropriate form for representing diffusion in fiber orientation simulations [7, 10, 11, 14–17].

Most fiber collision models assume fiber interactions are due to volume averaged effects similar to the theory of rotary Brownian motion [18]. In traditional Brownian motion, particles not subjected to an outside force will return to an isotropic orientation, whereas fibers in a polymer matrix will not reorient unless subjected to a fluid deformation. In the present study we accept Bird's model [18] for diffusion whereby the equation of motion of a fiber orientation probability distribution $\psi(\mathbf{p})$ is given as

$$\frac{D\psi}{Dt} = \nabla \cdot \left(-\dot{\mathbf{p}}^h \psi + \nabla \left(D_r \psi \right) \right) \tag{1}$$

where $\mathbf{p}$ is the unit vector along a fiber axis, D_r is the rotary diffusivity, and the symbol ∇ denotes the gradient projected onto the tangent space of the sphere $\mathbb{S}$, that is, $\nabla = (\mathbf{I} - \mathbf{pp}) \cdot \frac{\partial}{\partial \mathbf{p}}$. Γ is the rate of deformation tensor such that $\Gamma_{ij} = \frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i}$ where $\mathbf{v}$ is the velocity vector of the surrounding fluid. Observe that the time derivative is taken as the material derivative $\frac{D}{Dt}$ to recognize that orientation may be a function of spatial location and will convect with the bulk motion of the fluid. $\dot{\mathbf{p}}^h$ is the time derivative of the hydraulic component of rotational motion of the ellipsoid which is often assumed to be well represented by Jeffery's [19] equation expressed as,

$$\dot{\mathbf{p}}^h = \frac{D\mathbf{p}}{Dt} = -\frac{1}{2}\Omega \cdot \mathbf{p} + \lambda \frac{1}{2} \left(\Gamma \cdot \mathbf{p} - \Gamma : \mathbf{ppp} \right) \tag{2}$$

where Ω is the vorticity tensor, $\Omega_{ij} = \frac{\partial v_j}{\partial x_i} - \frac{\partial v_i}{\partial x_j}$.

Folgar and Tucker [1] proposed a diffusivity model based on the rate of deformation tensor and an empirical parameter C_I as $D_r = \gamma C_I$, where $\gamma = \sqrt{\frac{1}{2}\Gamma : \Gamma}$. Their model neglects the directional dependance of the collisions notwithstanding the suggestion by Folgar and Tucker that "...while it is possible, and even likely, that these orientation changes have a directional bias, or are different in nearly random and nearly aligned suspensions, we have chosen to ignore these features" [1].

Kamel and Mutel [20] proposed a diffusivity model that is a function of volume fraction, but for large volume fractions of fibers is independent of shear rate, violating the physical system being modeled. Koch [15] presents a model for diffusion resulting from hydrodynamic fiber-fiber interactions, but provides no experimental results nor a derivation to validate the model and claims validity only for low suspension concentrations in shearing flows. Phan-Thien *et al.* [7] assume rotary diffusion is an anisotropic second-order tensor that follows a "white noise" random force behavior obtained from experimental samples for the steady state solution.

The Reduced Strain Closure (RSC) model of Wang *et al.* [11] shows great promise as an objective diffusion model and provides a scaling parameter κ for the equation of motion that subtracts an objective diffusion term from Equation (1). The change in orientation is scaled via the eigenvalues and eigenvectors of the second-order moment tensor of the orientation distribution. The ARD model of Phelps and Tucker [14] is a more generalized form of the Koch [15] model and expands the rotary diffusion function in terms of the invariants of the second-order moment tensor of the distribution and the rate of deformation tensor. Both the RSC and the ARD models, although a significant step forward, do not lend themselves to a general derivation scheme and it is thus difficult to quantify their effectiveness for similar systems that share similarities with a short fiber system (i.e. agglomerating nanotube suspensions [10, 21, 22]). There still remains a considerable amount of work to be accomplished if a general form for the equation of motion of a semi-concentrated or a concentrated suspension of ellipsoidal inclusions is to be realized.

2. Spherical Harmonic Expansion: The solution of Equation (1) is not a trivial undertaking. Direct solutions using the control volume approach of Bay and Tucker [23] is particularly popular in determining high accuracy solutions, but solutions are computationally prohibitive (see e.g. [5]) for even simple flows. For example, solutions of Equation (1) with a control volume approach for Folgar and Tucker diffusion require several hours to days to solve for even the simplest of academic flows (i.e. pure shear and pure elongation). This is particularly relevant considering that recent diffusion models and those presented in this work require a greater computational demand due to the relative complexity of their assumed form for the rotary diffusion. For example, solutions using the Koch model [15] with control volumes require more than an order of magnitude increase in computational effort relative to the Folgar and Tucker model.

To alleviate this computational burden, Advani and Tucker introduced orientation tensors which are defined by taking moments of the fiber orientation probability distribution as

$$A_{ij\cdots} = \oint_{\mathbb{S}} p_i p_j \cdots \psi(\mathbf{p})\, d\mathbb{S} \tag{3}$$

where $\mathbb{S}$ is the surface of the unit sphere. Orientation tensors capture the stochastic nature of the fiber orientation distribution in a compact form via the dyadic products of the unit vector $\mathbf{p}$ and the fiber distribution function ψ. Computations that evaluate orientation tensors are on the order of seconds for simple flows. Unfortunately, their use introduces the issue of a closure whereby the resulting equation of motion for a given orientation tensor requires knowledge of a higher-order orientation tensor. For example with the second-order orientation tensor equation of motion the Folgar and Tucker diffusion model requires knowledge of the fourth-order orientation tensor. Similarly, the second-order tensor equation of motion with the Koch diffusion mode requires knowledge of both the fourth- and the sixth-order orientation tensor.

The method of spherical harmonic expansions developed by the authors (see e.g. Montgomery-Smith *et al.* [24, 25]) has been developed to solve partial differential equations on a sphere. In particular, solutions of Equation (1) have been solved in an efficient manner. The order of the expansion can be selected based on the desired order of accuracy and is only limited by the desired level of computational effort. The method is equivalent to using the higher-order moment tensors with a linear closure (see e.g. Hand [26]), and solutions have been performed by the authors for expansions up to order 400 with computational efforts equivalent to solutions using the second-order orientation tensors with a fourth-order orthotropic closure (see e.g. [4, 27–29] for several of the orthotropic closures) with accuracy approaching machine precision.

The spherical harmonic approach can be summed up by recognizing that a distribution defined on the unit sphere can be reconstructed as a system of ordinary differential equations as (see e.g. [25])

$$\frac{\partial}{\partial t}\hat{\psi}_l^m(\mathbf{p}) = \sum_{l'=0}^{\infty}\sum_{m'=-l'}^{l'} c_{l,l'}^{m,m'}\,\hat{\psi}_{l'}^{m'}(\mathbf{p}) \tag{4}$$

where each of the coefficients of expansion for the distribution function can be expressed as

$$\hat{\psi}_{l'}^{m'}(\mathbf{p}) = \int_{\mathbb{S}} \psi(\mathbf{p})\, \bar{Y}_l^m(\mathbf{p})\, d\mathbb{S} \tag{5}$$

where $\bar{Y}_l^m$ is the set of complex conjugates of spherical harmonics as described in [25]. The complex coefficients $c_{l,l'}^{m,m'}$ can be computed explicitly, and their resulting system of equations is extremely sparse thus lending itself to efficient and rapid computations. The choice of expansion is arbitrarily selected sufficiently large enough such that the error in selecting the linear closing of the expansion is of a similar order of magnitude as machine precision, thus bypassing the closure issue that plagues orientation tensor equations of motion. In addition, the form of Equation (4) is sufficiently general such that the equation of motion for each of the previously mentioned diffusion models have been solved (see e.g. Montgomery-Smith *et al.* [25]) using the spherical harmonic approach for many of the classical flows (i.e. shearing, elongational, etc.)

in an efficient manner which could have previously only been solved with the orientation tensor approach with a closure approximation.

3. Directional Diffusion: As part of this research, we investigate an anisotropic diffusion model that incorporates two effects, (1) local directionally dependant effects proportional to the probability of collision between two fibers represented by the unit vectors $\mathbf{p}$ and $\boldsymbol{\rho}$ shown in Figure 1, and (2) large scale volume averaged diffusion behavior analogous to shear rate dependant Brownian motion. To retain coordinate frame invariance when looking at the relative motion between two fibers $\boldsymbol{\rho}$ and $\mathbf{p}$, we assume both fibers experience the same vorticity, we consider Jeffery's model for the motion of the fiber $\boldsymbol{\rho}$ without rigid body effects expressed as

$$\dot{\underline{\boldsymbol{\rho}}}^h = \dot{\boldsymbol{\rho}}^h + \frac{1}{2}\Omega \cdot \boldsymbol{\rho} = \frac{1}{2}\lambda\left(\Gamma \cdot \boldsymbol{\rho} - \Gamma : \boldsymbol{\rho\rho\rho}\right) \tag{6}$$

During an infinitesimal period of time, the fiber $\boldsymbol{\rho}$ rotates through an angle relative to $\mathbf{p}$ proportional to $\dot{\underline{\boldsymbol{\rho}}}^h$, and the area through which $\boldsymbol{\rho}$ rotates is proportional to the magnitude of $\boldsymbol{\rho} \times \dot{\underline{\boldsymbol{\rho}}}^h$, with a direction normal to the plane containing $\boldsymbol{\rho}$. The probability of a hit between $\mathbf{p}$ and $\boldsymbol{\rho}$ will be zero when $\mathbf{p}$ is parallel to the plane containing $\boldsymbol{\rho}$ and $\dot{\underline{\boldsymbol{\rho}}}^h$, and greatest when $\mathbf{p}$ is parallel to $\boldsymbol{\rho} \times \dot{\underline{\boldsymbol{\rho}}}^h$. Therefore, we assume the probability of a collision $P_{\boldsymbol{\rho}\,\text{hit}\,\mathbf{p}}$ proportional to the scalar triple product of $\mathbf{p}$ with the vector $\boldsymbol{\rho} \times \dot{\underline{\boldsymbol{\rho}}}^h$ as

$$\begin{aligned} P_{\boldsymbol{\rho}\,\text{hit}\,\mathbf{p}} &= C_1 \left|\mathbf{p} \cdot \left(\boldsymbol{\rho} \times \dot{\underline{\boldsymbol{\rho}}}^h\right)\right| \\ P_{\mathbf{p}\,\text{hit}\,\boldsymbol{\rho}} &= C_1 \left|\boldsymbol{\rho} \cdot \left(\mathbf{p} \times \dot{\underline{\mathbf{p}}}^h\right)\right| \end{aligned} \tag{7}$$

where the second expression in Equation (7) expresses the probability of fiber $\mathbf{p}$ rotating into fiber $\boldsymbol{\rho}$, and the constant C_1 is allowed to be a function of fiber aspect ratio and volume fraction. The fibers $\boldsymbol{\rho}$ are sampled from $\psi(\boldsymbol{\rho})$, ergo the expectation of a collision with a given fiber $\mathbf{p}$ may be represented by the integral over $\boldsymbol{\rho} \in \mathbb{S}$. In addition to local collisions, we incorporate volume averaged effects assumed to satisfy the Brownian-type behavior observed by Folgar and Tucker [1]. Thus the combination of local collision effects and the volume averaged effects yields

$$\begin{aligned} D_r(\mathbf{p}) = {} & C_1 \oint_{\boldsymbol{\rho}\in\mathbb{S}} \psi(\boldsymbol{\rho}) \left(\left|\boldsymbol{\rho} \cdot \left(\mathbf{p} \times \dot{\underline{\boldsymbol{\rho}}}^h\right)\right| + \left|\mathbf{p} \cdot \left(\boldsymbol{\rho} \times \dot{\underline{\mathbf{p}}}^h\right)\right|\right) d\mathbb{S} \\ & + C_2 \oint_{\boldsymbol{\rho}\in\mathbb{S}} \dot{\gamma}\psi(\boldsymbol{\rho})\, d\mathbb{S} \end{aligned} \tag{8}$$

It is worthwhile to note that the Folgar-Tucker model is obtained by setting $C_1 = 0$ and $C_2 = C_I$. Note that in Equation (8) the rotary diffusivity remains a function of the velocity gradients through the rate of deformation tensor Γ and the fiber orientation through the unit vector $\mathbf{p}$, which is independent of the integration for $\boldsymbol{\rho} \in \mathbb{S}$.

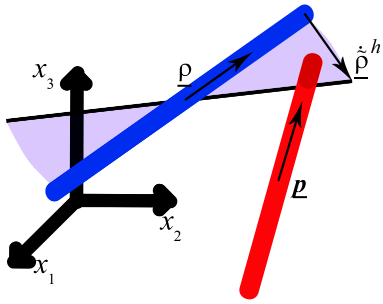

Figure 1: Coordinate system depicting the path through which the fiber $\boldsymbol{\rho}$ passes into $\mathbf{p}$.

It is important to note that like our previously published solutions of Equation (1) (see e.g. Jack [30] and Jack *et al.* [17]) with the directional diffusion model of Equation (8), this new approach also requires that each of the absolute value terms in Equation (8) be replaced by the expression squared, i.e. $|x| \simeq x^2$. This was included in the earlier work to cast the equation of motion for ψ in terms of its moments (see e.g. Jack [30]), thus avoiding the prohibitive computational expense when solving for ψ via the usual control volume approach (estimated to be nearly $\sim 2.4 \times 10^6$ seconds for simple shear flow). Unfortunately, the prior formulation that directly computed the second-order orientation tensor required the tenth-order orientation tensor. Several higher-order tensor closures approximations were introduced which significantly increased the error and decreased the confidence in the results. The new approach employed here solves Equation (1) with directional diffusion using spherical harmonic expansions. As a result, these solutions can now be obtained without higher-order closures, thus allowing a fair and objective criticism of the directional diffusion model.

Two flows are studied, both beginning from an initially isotropic orientation state, i.e. $\psi = \frac{1}{4\pi}$. Recall that an effective diffusion model should have the tendency to slow down the rate of alignment away from an isotropic state without drastically altering the final orientation state. For graphical purposes, all results are presented in the orientation tensor format where the rate of alignment is easily observed. Notice in Figure 2 the Folgar and Tucker solution with $C_I = 10^{-3}$, designated by the solid line, begins from an initial isotropic orientation state (i.e. $A_{11} = A_{22} = A_{33} = 1/3$), but as time progresses, the A_{11} parameter goes to 1 and the $A_{22} = A_{33}$ parameters go to zero which would be indicative of a perfectly aligned orientation state with the alignment occurring in the x_1 direction. For each of the directional diffusion simulations it was found that there was little to no change in the final orientation state between the Folgar and Tucker solutions and the directional diffusion solutions if the coefficient C_2 (corresponding to the volume averaged diffusion) was set to 10^{-3} regardless of C_1. Notice that as the coefficient C_1 increases (thus implying that the contribution from local collision effects is increasing) the rate of alignment appears to diminish and the steady state solution occurs at a much later moment in time.

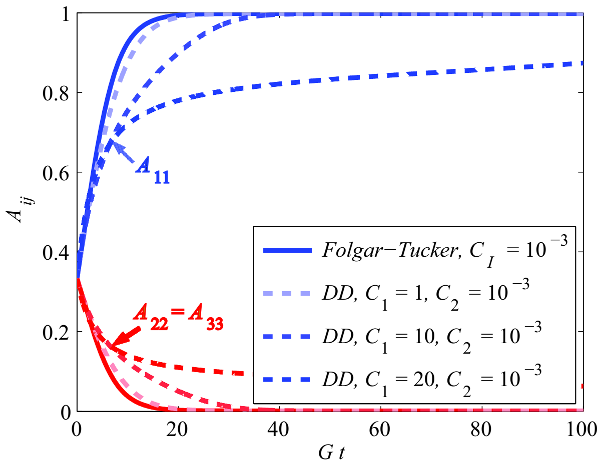

Figure 2: Transient solution for A_{ij} from uniaxial elongation, directional diffusion model, $C_2 = 10^{-3}$.

The second flow studied is a pure shearing flow with the bulk fluid motion being constrained to the x_1 direction with shearing occurring solely in the x_3 direction (i.e. $v_1 = Gx_3$, $v_2 = v_3 = 0$). Unlike uniaxial elongation flow, the coefficient C_2 is strongly dependant on the choice of C_1. The range for viable C_1 values is $C_1 \in \left(0, 5 \times 10^{-2}\right)$ is greatly diminished without drastically altering the final orientation state as observed in Figure 3. Notice the directional diffusion model has only nominal effects on orientation states near isotropic.

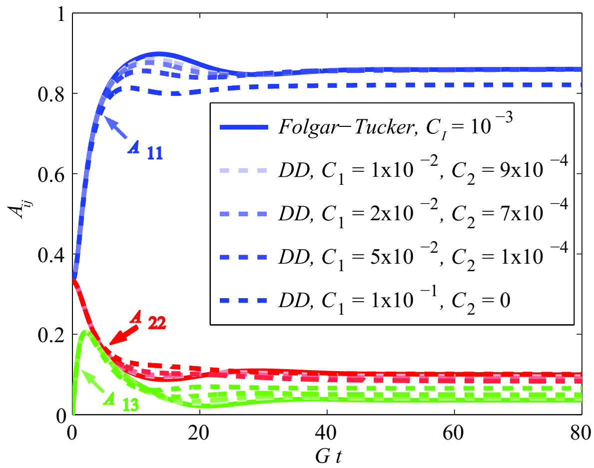

Figure 3: Transient solution for A_{ij} from Simple Shearing flow, directional diffusion model.

It is worthwhile to take a closer look at the directional diffusion model characteristics nearing the isotropic orientation state. It is desired that an appropriate diffusion model would slow down the rate of alignment, but in particular it is desired to diminish the rate of alignment for an orientation state near isotropic. A close-up of the uniaxial elongation simulation from Figure 2 is presented in Figure 4. Observe how the A_{11} component of the orientation tensor initially increases quickly from the isotropic orientation state, and then the rate of orientation diminishes once the orientation state achieves some level of alignment. This characteristic is quite undesirable and further work is required to construct a viable rotary diffusion form with the desired characteristics.

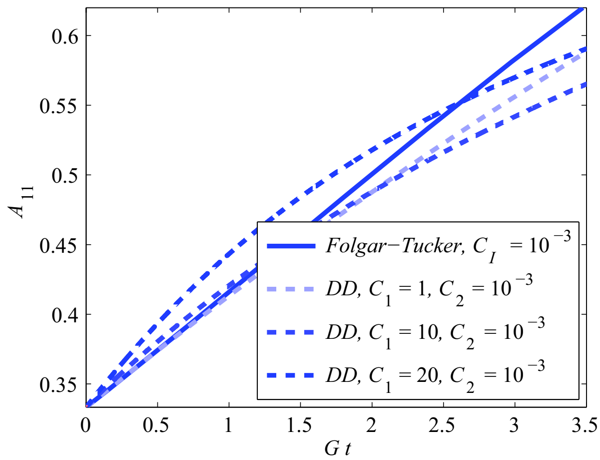

Figure 4: Transient solution for A_{ij} from uniaxial elongation, directional diffusion model, $C_2 = 10^{-3}$ for $Gt < 1$.

4. Variable Lambda Model: An alternative approach to investigating collision models in the form of Equation (1) is to focus on the λ term within $\dot{\boldsymbol{p}}^h$ which represents the 'equivalent aspect ratio' [19]. Jeffery's derivation assumes that the fiber is surrounded by a fluid that obeys the Newtonian Stokes equation [19]. He assumes that the fiber is by itself in an infinite fluid. This is a reasonable approximation when fraction of fibers within the fluid is very close to zero. But as the volume fraction becomes large, the fibers interact with each other via Stokesian effects upon one another. Each fiber moving within the fluid affects the fluid that is pushing it, and the effect upon the fluid is transferred via Stokes' equation to effect the fluid surrounding other fibers. An exhaustive treatment would mean having to solve the complete Stokes equation in the presence of many fibers, and attempts by the authors and their students to do this using finite element methods have so far proved computationally prohibitive.

We assume the motion of the fiber itself alters the motion of the surrounding fluid. This is described by observing the two ends of the fiber of length ϵ, that are separated by the vector $\epsilon \mathbf{p}$. The fluid attempts to pull each end slightly differently, such that the difference between the velocity of each respective end is $\mathbf{v} = \frac{1}{2}\epsilon(-\Omega\cdot\mathbf{p}+\Gamma\cdot\mathbf{p})$. If the two ends move freely, then the fluid motion would be independent of the existence of the fiber. But the two ends are constrained to stay the same distance from each other by the rigid rod assumption. Thus one needs to subtract the projection of $\mathbf{v}$ onto $\mathbf{p}$ from this relative velocity. This projection computes as $\mathbf{v}\cdot\mathbf{pp} = \frac{1}{2}\epsilon\Gamma : \mathbf{ppp}$. This exerts a counter reaction upon the fluid, whose size is proportional to $\frac{1}{2}\epsilon\Gamma : \mathbf{pp}$. In the case that the fiber is oriented in a direction $\mathbf{p}$ such that $\Gamma : \mathbf{pp} = 0$, the fiber has no effect on the fluid that is pushing it around. Average this quantity, the following expression can be readily obtained

$$s := \gamma^{-1}\int_{\mathbb{S}} \psi\Gamma : \mathbf{pp}\, d\mathbb{S} = \gamma^{-1}A : \Gamma, \tag{9}$$

where A is the second-order orientation tensor defined in Equation (3) and the quantity γ^{-1} is present to make s dimensionless.

Thus we propose a model where, if $s = 0$, the pure Jeffery's model applies with $\lambda = 1$ and $D_r = 0$. Alternatively, if $s \neq 0$, then we propose that interaction provides two effects. First, the Stokesian lubrication between the fibers will cause them to stick together. Then the calculation of Jeffery's parameter λ should consider the aggregate of the fibers as a single, albeit flexible object, which would be a single object whose aspect ratio is significantly less than infinity, that is, with λ quite a bit less than one. Second, the fibers will push each other via Stokesian interactions, thus suggesting the use of a non-zero diffusion term.

The 'variable lambda' (Vλ) model sets the λ and D_r parameters in Equation (1) as

$$\begin{aligned} \lambda &= f(s) \\ D_r &= \gamma g(s) \end{aligned} \tag{10}$$

As a preliminary try, we propose the functions $f(s)$ and $g(s)$ defined as

$$\begin{aligned} f(s) &= \max\{1-\kappa^{-1}|s|, \kappa\} \\ g(s) &= \mu \min\{\kappa, |s|\} \end{aligned} \tag{11}$$

Note that

$$|s| \le \sqrt{2} \tag{12}$$

from the Cauchy-Schwartz inequality which yields

$$|A:\Gamma| \le (A:A)^{1/2}(\Gamma:\Gamma)^{1/2} \le \sqrt{2}\gamma(A:A)^{1/2} \tag{13}$$

where $(A : A)^{1/2}$ is the Frobenius norm of A, which is bounded by the sum of its singular values. Since A is positive semi-definite, the singular values are the same as the eigenvalues, and hence the sum of the singular values is equal to the trace of A, which is one.

Other possibilities are to replace the quantity s by

$$\gamma^{-1}\int_{\mathbb{S}} \psi|\Gamma : \mathbf{pp}| d\mathbb{S} \quad \text{or} \quad \gamma^{-1}\left(\int_{\mathbb{S}} \psi|\Gamma : \mathbf{pp}|^2 d\mathbb{S}\right)^{1/2} \tag{14}$$

The former quantity is very difficult to work with in the framework of spherical harmonics. The latter quantity can be easily computed using the fourth order moment tensor. However the authors found that using it did not create reasonable numerical results.

As with the directional diffusion two flows are studied, a pure shear flow and an elongational flow, both beginning at an initially isotropic orientation state, i.e. $\psi = \frac{1}{4\pi}$. Results are compared with the Folgar-Tucker model for an interaction coefficient of $C_I = 10^{-3}$. The results for the pure elongational flow are shown in Figure (5) for values of $\mu = 10^{-3}$ and $\kappa = 1, \frac{1}{2}, \frac{1}{4}, \frac{1}{10}$. Observe that the variable lambda model does an excellent job of elongating the rate of alignment. The Folgar-Tucker model attains an alignment state nearing steady state before $Gt = 25$, but depending on the choice of the parameter κ the same steady state solution is not attained until a much later point in time. It is also worthwhile that the steady state solution between the Vλ model and the Folgar-Tucker model are graphically indistinguishable.

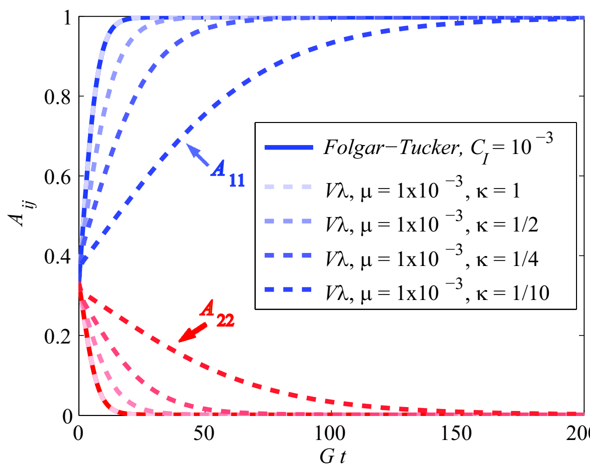

Figure 5: Transient solution for A_{ij} from uniaxial elongation, variable lambda model.

In the pure shearing flow, the variable lambda model performs quite well as observed in Figure 6. The rate of alignment is considerably reduced as the parameter κ is reduced as compared to the Folgar-Tucker results for $C_I = 10^{-3}$. It is worthwhile to mention that the A_{11} and the A_{22} components between the two diffusion models are the same at steady state, which in the case of $\kappa = \frac{1}{10}$ is observed numerically for $Gt > 5{,}000$.

It is also worthwhile to observe that the A_{13} component from the variable lambda model decays at a much quicker rate than in the Folgar-Tucker model. Although both models experience the same steady state, this may

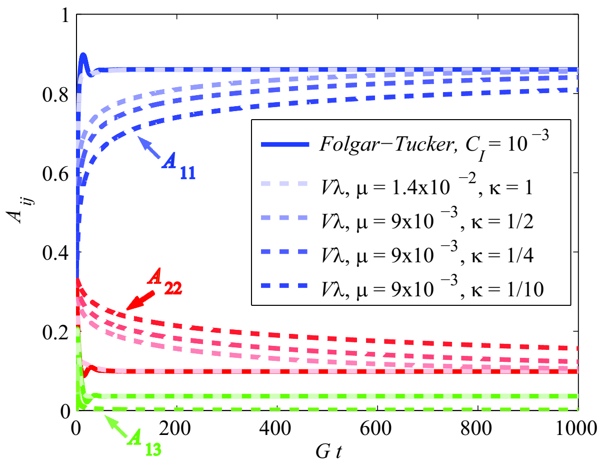

Figure 6: Transient solution for A_{ij} from Simple Shearing flow, variable lambda model.

not be a desirable characteristic when forming predictions for the viscosity (see e.g. [11,31,32])

$$\boldsymbol{\tau} = 2\eta\Gamma + 2\eta N_p \mathbb{A} : \Gamma \qquad (15)$$

where the extra stress $\boldsymbol{\tau}$ from the Navier-Stokes equations is a function of the matrix viscosity η, the dimensionless parameter N_p that relates to the relative importance of the fiber effects, and $\mathbb{A}$ is the fourth-order orientation tensor. A component of particular difficulty to predict in an industrial setting is the τ_{13} term (see e.g. Wang *et al.* [11]), which is related to the A_{13} component of the second-order orientation tensor. It will be the focus of future work to quantify the relevance of this effect in coupled industrial flows.

5. Conclusions: Numerical representations for short-fiber kinematic motion has relied on the Folgar and Tucker [1] model of diffusion for over twenty years, but recent research has demonstrated the propensity of this model to over-predict the rate of alignment. We presented two models to represent fiber orientation kinematics, with the preferred model being the variable lambda (Vλ) construction. We believe that the Vλ model, or a related modification, has the potential to provide the necessary corrections to the Folgar-Tucker-Jeffery's equation that will represent the real data more accurately. As experimental evidence becomes available, it may be possible to find appropriate adjustments to the various parameters such that the constitutive model results correspond with experimental observations.

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