PREDICTION OF RHEOLOGICAL PROPERTIES OF STRUCTURED FLUIDS IN HOMOGENEOUS SHEAR BASED ON A REALIZABLE MODEL FOR THE ORIENTATION DYAD

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EXTENDED ABSTRACT

Non-spherical particles dispersed in a Newtonian fluid have a tendency to align in shear flows because of viscous drag. This phenomenon is opposed by rotary diffusion induced by *particle-particle interactions.* At high concentrations and in the absence of hydrodynamic couples, *self-alignment* can also occur because *excluded volume interactions* prevent the return to isotropy of anisotropic states by rotary Brownian motion. The consequences of the foregoing balance between hydrodynamic and diffusive alignment processes on the microstructure directly impact the rheology and, thereby, the processing of suspensions. The objective of this research is to predict the microstructure and rheological properties of axisymmetric "ellipsoidal" particle suspensions in homogeneous flows.

Low-order moments of the orientation distribution function governed by Smoluchowski's equation are commonly used to characterize the microstructure of "rigid-rod" fluids. This classical approach has also been employed to study the flow-induced alignment of other fluids, such as thermotropic and lyotropic liquid crystalline polymers. The resulting theory for the instantaneous orientation dyad $<\underline{p}\,\underline{p}>$ depends on four dimensionless groups: the particle aspect ratio, L/d; the Péclet number, $\mathrm{Pe} \equiv \left\| \underline{\underline{S}} \right\| / (6 D_R)$; a phenomenological coefficient associated with the Maier-Saupe excluded volume potential, U; and, a dimensionless time $t \equiv 6 D_R \hat{t}$, where D_R is a rotary diffusion coefficient. A critical step in the practical application of the theory is the development of a closure hypothesis that relates the orientation tetrad $<\underline{p}\,\underline{p}\,\underline{p}\,\underline{p}>$ to the orientation dyad $<\underline{p}\,\underline{p}>$ by an algebraic equation:

$$<\underline{p}\,\underline{p}\,\underline{p}\,\underline{p}> = \Im\,(<\underline{p}\,\underline{p}>)\,.$$

The Cayley-Hamilton theorem of linear algebra is used to develop the following irreducible representation for the orientation tetrad in terms of two tetradic operators, $\Im_1$ and $\Im_2$, that satisfy all six-fold symmetry and six-fold contraction properties associated with the exact orientation tetrad:

$$< \underline{p}\,\underline{p}\,\underline{p}\,\underline{p} > \; = (1 - C_2(II_b, III_b))\, \Im_1(< \underline{p}\,\underline{p} >) + C_2(II_b, III_b)\, \Im_2(< \underline{p}\,\underline{p} >).$$

In the above equation, $\Im_1$ is linear in $< \underline{p}\,\underline{p} >$ and $\Im_2$ is quadratic in $< \underline{p}\,\underline{p} >$. C_2 is a scalar-valued function of the invariants of the anisotropic component of the orientation dyad: $\underline{\underline{b}} \equiv < \underline{p}\,\underline{p} > - \underline{\underline{I}}/3$, $II_b \equiv \mathrm{tr}(\underline{\underline{b}} \cdot \underline{\underline{b}})$ and $III_b \equiv \mathrm{tr}(\underline{\underline{b}} \cdot \underline{\underline{b}} \cdot \underline{\underline{b}})$.

In this research, the coefficient C_2 is determined by assuming that all planar anisotropic states, defined by $II_b = 2/9 + 2\,III_b$, $-1/36 \le III_b \le +8/36$, are marginally realizable in the absence of an external alignment field. This strategy implies that

$$C_2(II_b, III_b) = C_2(\frac{2}{9} + 2\,III_b, III_b) = \frac{8 + 45\,III_b}{18(1 + 9\,III_b)}.$$

With the above closure, all two-dimensional and three-dimensional *realizable* microstructures subjected to a homogeneous shear flow will relax to either an anisotropic realizable steady state or an anisotropic realizable periodic state, depending on the values of L/d, U, and Pe. Moreover, in the absence of an external hydrodynamic force field (i.e., $Pe = 0$), the theory predicts the existence of a biphasic region for $4.72 \le U \le 5.00$ (i.e., coexistence of stable isotropic and anisotropic states).

For L/d < ∞ and U > 25, *tumbling* and *wagging* of the director occurs at low to moderate values of the Péclet number. If the initial state has a director with a component in the vorticity direction, then director *kayaking* and director *log-rolling* may occur.

The influence of the Péclet number on the shear viscosity agrees qualitatively with previous theories and with experimental observations reported for particulate suspensions and for liquid crystalline polymers. For L/d = ∞, the shear viscosity shows a Newtonian plateau at low shear rates and a shear thinning region at high shear rates. However, for L/d < ∞ , a low and high shear rate Newtonian plateau obtains in the absence of *tube dilation* phenomena. If the microstructure affects the rotary diffusion coefficient through *tube dilation*, then the shear viscosity shows a low and high Newtonian plateau as well as a shear thickening and a shear thinning region for U > 25 and L/d < ∞. This unanticipated result is related to director tumbling and wagging. A modified elastic stress model must be introduced in future studies in order to recover the anomalous shear thinning phenomena observed for some liquid crystalline polymers at very low shear rates.

The behavior of the primary normal stress difference N_1 and the secondary normal stress difference N_2 also agree qualitatively with previous theories and with experimental observations reported for particulate suspensions and for liquid crystalline polymers. For example, for steady-state alignment of the orientation director (U < 35), the theory predicts that $N_1 > 0$ and $N_2 < 0$ with $|N_2| \leq |N_1| / 200$. On the other hand, if director tumbling occurs, then the time average primary and secondary normal stress differences flip signs, i.e., $N_1 < 0$ and $N_2 > 0$.