An ellipsoidal droplet model for dilute emulsions: application to complex mixing flows

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Abstract

A compact droplet model and numerical simulation are used to examine the behavior of multiphase fluids in complex mixing flows. The two-dimensional, time-periodic fluid between eccentric cylinders is used as a prototypical chaotic mixing flow. The multiphase fluid is a dilute dispersion of Newtonian droplets in a Newtonian matrix. The droplets are assumed to be much smaller than the mixer dimensions, and to have no interfacial tension, a situation for which there is an exact, yet compact, model for droplet deformation. We examine the spatial distribution of droplet stretching, as well as global stretching statistics, for a variety of droplet-to-matrix viscosity ratios. Chaotic flows are able to stretch high-viscosity droplets (which cannot be stretched in simple shear flow), and many features of the stretching statistics follow the "universal" laws for chaotic mixing of passive fluids.

1 Introduction

Most studies of complex multiphase fluids focus on simple flows, such as simple shear or planar elongation. While understanding fluid behavior in these situations is an essential step, flows of practical interest may be much more complex. A case in point is laminar mixing flows, which are used in the processing of polymer blends and other viscous multiphase systems. The most effective laminar mixing flows are chaotic, and so involve exponential stretching, folding, and the development of fractal structures. Thus, it is of considerable interest to understand the effect of these complex flows on multiphase fluids.
From another viewpoint, the overwhelming majority of chaotic mixing studies are limited to passive mixing, where the two fluids have equal viscosity and no interfacial tension. A great deal is known about the nature of chaotic mixing for this special case (e.g., [1–3]). We would like to know which parts of that understanding carry over to more complicated fluid systems.

In this paper we use an analytical model for the deformation of ellipsoidal droplets with no interfacial tension, examining its behavior in the two-dimensional, time-periodic flow between eccentric cylinders. Section 2 reviews the droplet deformation model and explains the numerical methods used in this study. Section 3 presents results for spatial distributions of stretching and global stretching statistics. Some summary comments and discussion appear in Section 4.

2 Theory and numerical methods

2.1 Droplet deformation model

We are interested in the mixing of highly viscous fluids, so we henceforth neglect inertia and body forces. To treat a complex flow with a spatial distribution of microstructure one must follow many thousands of droplets, so direct numerical simulation on the scale of a single droplet is not feasible. Instead, one must resort to some simpler model, which is easier to compute but which still represents the important physics.

Recently, droplet models based on an ellipsoidal droplet shape have proved quite successful in modeling the structure and rheology of immiscible polymer blends (see [4] for a review). Here we use the model of Wetzel and Tucker [5]. Based on the ellipsoidal inclusion model of Eshelby [6], they developed the governing equations for a three-dimensional ellipsoidal droplet with no interfacial tension in a linear far-field flow. Their analysis shows that if the droplet begins with an ellipsoidal shape (which could be spherical) then it will be an ellipsoid for all time. This particular model is an exact solution to the Stokes equations for a single droplet in an infinite matrix.

The instantaneous droplet shape is described by a symmetric second-order tensor $\mathbf{G}$, defined such that points on the droplet surface satisfy

$$\mathbf{x} \cdot \mathbf{G} \cdot \mathbf{x} = 1,$$

where $\mathbf{x}$ is the position vector from the droplet centroid. $\mathbf{G}$ is called the droplet shape tensor. The eigenvectors of $\mathbf{G}$ correspond to the principal axes of the ellipsoid. In this principal axis system the components of $\mathbf{G}$ are

$$\mathbf{G}_{\text{prin}} = \begin{bmatrix} 1/a^2 & 0 & 0 \\ 0 & 1/b^2 & 0 \\ 0 & 0 & 1/c^2 \end{bmatrix}$$

where $a$, $b$, and $c$ are the principal semi-axis lengths of the ellipsoid. Thus, the lengths and orientation of the droplet axes are easily deduced from $\mathbf{G}$. Figure 1
Figure 1: Ellipsoidal droplet with in-plane semi-axes $a$ and $b$ and out-of-plane semi-axis $c$.

shows an ellipsoidal droplet and the semi-axis lengths. Since the present study is restricted to two-dimensional flows, we choose $a$ and $b$ to be the semi-axis lengths in the plane of the flow, with $a \geq b$, and let $c$ denote the out-of-plane semi-axis.

The complete velocity solution gives the remarkable result that the velocity gradient $\mathbf{L}^*$ within the droplet is uniform. Kinematic arguments then show that the rate of change of the droplet shape tensor is

$$\dot{\mathbf{G}} + \mathbf{L}_*^T \cdot \mathbf{G} + \mathbf{G} \cdot \mathbf{L}_*^T = 0. \tag{3}$$

The dot denotes a material derivative, and the centroid of the droplet advects with the unperturbed flow.

The internal droplet velocity gradient is a linear function of the far-field velocity gradient $\mathbf{L}$, where we use the convention $L_{ij} = \partial v_i / \partial x_j$. Separating $\mathbf{L}$ into its symmetric part $\mathbf{D}$ and its anti-symmetric part $\mathbf{W}$, the internal droplet velocity gradient is given by

$$\mathbf{L}_*^T = \mathbf{W} + (\mathbb{B} + \mathbb{C}) : \mathbf{D} \tag{4}$$

The fourth-order tensors $\mathbb{B}$ and $\mathbb{C}$ are analytical functions of the droplet shape $\mathbf{G}$ and the viscosity ratio $p = \mu_{\text{droplet}} / \mu_{\text{matrix}}$; see [5] for details. For the special case of $p = 1$, the values of $\mathbb{B}$ and $\mathbb{C}$ give $\mathbf{L}_* = \mathbf{L}$, which is the affine deformation of a passive droplet. Combining Eqns. (3)–(4) gives a closed droplet evolution equation that can be integrated at any material point.

One striking prediction of this model is that an initially spherical droplet with $p$ greater than approximately four cannot be stretched in simple shear flow. Instead, the droplet executes a tumbling motion, where the length and orientation of its major axis vary periodically in time. This is connected to the well-known phenomenon that a droplet with interfacial tension and $p > 4$ will not break up in a simple shear flow, regardless of the capillary number [7, 8]. However, droplets with $p > 4$ will stretch in an elongational flow, and they can also be stretched in a simple shear flow if they have a non-spherical initial shape. Practitioners of mixing know that dispersions with $p > 4$ are difficult to mix. Thus, we are quite interested in the mixing capabilities of chaotic flows under these conditions.
2.2 Mixing flow

We analyze a time-periodic, two-dimensional mixing flow, which has received considerable study for passive mixing. This is the flow between eccentric cylinders, also called the journal bearing flow [9–11]. A cylinder of radius $R_{\text{in}}$ sits within a cylinder of radius $R_{\text{out}}$, with its center displaced a distance $d$ from the center of the outer cylinder. We use $R_{\text{out}}/R_{\text{in}} = 3$ and $d/R_{\text{out}} = 0.3$.

We consider mixing protocols where each period involves rotating one cylinder though a specified angle, then stopping that cylinder and rotating the other cylinder through a specified angle. Only creeping flow is considered, so the two angles completely define the protocol. This paper reports results for a motion we call protocol A, in which the outer cylinder first rotates through $\theta_{\text{out}} = 2\pi$, then the inner cylinder rotates by $\theta_{\text{in}} = 6\pi$.

The Poincaré map for this protocol is shown in Fig. 2(a). Protocol A, which has been studied by a number of authors [12–15], is almost globally chaotic. However, points close to the boundary move slowly, remaining close to the boundary for many periods, before they finally move away from the boundary near a stagnation point on the right outer wall.

2.3 Numerical methods

Similar to many computational studies of passive mixing, we use a Lagrangian particle method to follow the temporal and spatial development of the microstructure. We seed the flow with a large number of particles, typically 110,000, randomly distributed. In all cases the initial droplet shape is spherical, $a_0 = b_0 = c_0$. The position $\mathbf{x}(t)$ for each particle is tracked by integrating $\dot{\mathbf{x}} = \mathbf{v}(\mathbf{x}, t)$, while the microstructural variable $\mathbf{G}$ is updated by integrating the droplet shape evolution...
equation (3). Velocities are calculated using an analytical solution for the eccentric cylinder flow [16]. Velocity gradients, which drive the evolution of microstructure, are found by analytically differentiating the velocity solution.

The differential equations for position and microstructure are integrated numerically, using a fourth-order Runge-Kutta algorithm with fifth-order error estimation and adaptive step sizing [17]. When plotting the spatial distribution of any microstructural variable, we use a quadrilateral mesh covering the flow domain, and color each cell based on an average value for all particles within the cell. For the eccentric cylinder flow the plotting mesh has 40 divisions in the radial direction and 196 divisions in the angular direction. The average number of particles in each element is therefore approximately 14.

3 Results

3.1 Spatial distribution of stretch

The stretch ratio of the long droplet axis, $a/a_0$, is a convenient measure of the local extent of mixing. Also, analysis of our droplet model by Cristini [18] shows that $b/b_0$ and $c/c_0$ are completely determined by $a/a_0$ and $p$ in any two-dimensional flow. Figure 2(b) shows the spatial distribution of this variable for ten periods of protocol A for $p = 3$. Each cell is shaded using the geometric average of $a/a_0$, so that a few material points with very large stretch ratios do not dominate the cell average.

Figure 2(b) shows that stretching is highly non-uniform, even in the chaotic flow of protocol A. In fact, it is necessary to color the figure using the (base 10) logarithm of $a/a_0$, to visually display the wide range of stretching values. This is typical of calculations that model the complete spatial distribution of stretching or interfacial area [14, 19].

Protocol A exhibits high stretching over most of the flow, with only a small poorly-mixed (dark) region near the right-hand side of the mixer. This region surrounds a stagnation point on the outer cylinder, which returns to the same position at the end of each period. Material points close to this stagnation point are actually part of the chaotic region, but they move away from the stagnation point slowly, and thus experience small amounts of stretch for many periods. This is an undesirable, but perhaps tolerable, feature in a mixing flow.

The complexity of the spatial distribution of stretching is highlighted in Fig. 3. While there are significant variations on the scale of the mixer (Fig. 3(a)), a close-up look (Fig. 3(b)) shows similarly large variations on a small scale. To further examine the fine-scale details we placed 100,000 points in the same box as Fig. 3(b), translated them backward for five periods, initialized their microstructure to a spherical shape, and then integrated forward for five periods. Stretching values for these points, averaged on a much finer grid, were used to draw Fig. 3(c). This figure shows even more fine-scale structure than Fig. 3(b), with narrow bands of low stretch interspersed with bands of high stretch. As mixing continues, we expect the amount of fine-scale detail to increase, generating a fractal spatial structure in the
Figure 3: Spatial distribution of droplet slenderness, \( \log a/b \), for five periods of protocol A with \( p = 3 \). (a) Entire mixer. (b) Closeup of the indicated region from (a). (c) Same region as (b), but with much higher spatial resolution. All figures use the same color scale.

stretching distribution.

### 3.2 Global stretching statistics

A more quantitative way to characterize mixing is to examine the global statistics for stretching. Studies for passive mixing \((p = 1)\) [12, 20, 21] show that the statistics for stretching follow certain universal laws. We wish to see how the viscosity ratio affects this behavior.

Following Alvarez et al. [20], we focus on the probability density function for the logarithm of the stretch ratio. Here we use \( s \equiv \log(a/a_0) \). At period \( n \) the probability density function \( H_n \) is computed as

\[
H_n(s) = \frac{1}{N} \frac{dN(s)}{ds}
\]  

(5)

where \( N \) is the total number of particles in the simulation result and \( dN(s) \) is the number of particles with values between \( s \) and \( s + ds \). With this definition the total area under the curve equals unity, and visual assessment of the distribution is easy.

Figure 4(a) shows that, as the number of periods increases, the mean of the stretching distribution \( H_n(s) \) increases, and the distribution becomes broader. This figure is for protocol A and \( p = 3 \); stretching distributions for other values of \( p \) show the same trends.

For a given value of \( p \) the stretching distributions for different periods collapse to a universal distribution when plotted in terms of a normalized stretch variable \( z \),

\[
z = \frac{s - \langle s(n) \rangle}{\sigma_s(n)}
\]  

(6)
Figure 4: Probability density functions of log of droplet stretch for protocol A with $p = 3$. (a) Unscaled distributions for $n = \{1, 5, 9, \ldots, 21, 25\}$ periods. (b) Same distributions scaled using Eqn. (6).

Here $\langle s(n) \rangle$ is the mean of $s$ and $\sigma_s^2(n)$ is the variance of $s$, both at period $n$. This collapse is shown in Figs. 4(b), where $G_n(z)$ is defined like $H_n(s)$, but with $z$ as the variable. The overall shape of the stretching distribution is Gaussian, with a narrow peak of low-stretch material. As the number of periods increases the size of this low-stretch peak decreases, and the peak moves farther to the left of the scaled mean. Muzzio et al. [12] analyzed the same flow for $p = 1$ and attributed this peak to the material near the stagnation point and along the outer wall (the dark areas on the right in Fig. 2(b)). We find the same phenomena, but for $p > 1$ the low-stretch peak also includes some droplets in the main part of the flow that are still tumbling.

Given this universal scaling, we can characterize the performance of a mixing flow by the time dependence the mean and variance of the stretching distribution. These too follow a relatively simple pattern, as shown in Fig. 5(a). This figure plots the geometric mean stretch ratio $\lambda_g$ versus the number of periods, where $\log \lambda_g = \langle s \rangle$.

First, the curve for $p = 1$ rises linearly (after a short initial transient of about one period) on this semi-log plot, indicating that $\lambda_g$ is rising exponentially. This is the exponential stretching typical of chaotic flows, and the slope of this curve is governed by the Lyapunov exponent of the flow, which equals $d(\log \lambda_g)/dn$.

For other viscosity ratios the initial growth rate of $\lambda_g$ decreases as $p$ increases. However, eventually $\lambda_g$ achieves the same exponential growth rate as for $p = 1$. The physical mechanism underlying this phenomenon is hinted at by Fig. 5(b), which shows the stretch history of initially spherical droplets subjected to a planar elongational flow. A passive droplet ($p = 1$) extends exponentially in this flow, giving a straight line on this semi-log plot. Spherical droplets with $p > 1$ extend more slowly. However, as the droplets stretch their surface area increases, and
the matrix fluid can more easily exert traction along the droplet axis via shear stresses on the droplet surface. Eventually the droplet stretches in the direction parallel to the droplet axis at the same rate as the matrix. A similar phenomenon occurs in chaotic mixing flows, where the overall deformation experienced by fluid elements is extensional in nature. The initial transient period is longer in the mixing flow, because all droplets do not experience the same stretching rate during the first few periods. However, the droplets gradually extend, and are oriented along the unstable manifolds of the flow, so they eventually experience the underlying exponential stretching.

To finish characterizing the stretching history of a chaotic flow one must find how the variance $\sigma_s^2$ changes with time. For large numbers of periods this should also grow linearly with $n$, however some chaotic flows exhibit a power-law growth for “intermediate” numbers of periods [22], which might include the entire range of practical mixing times.

4 Discussion

Globally chaotic flows exhibit a universal asymptotic behavior for stretching distributions, with exponential growth of the mean and variance of the distribution. Strikingly, these features are preserved when mixing dispersions of droplets with no interfacial tension. However, as the viscosity ratio increases the number of periods required to reach the asymptotic behavior increases.

The behavior of flows that contain both large regular regions as well as chaotic regions is quite different. Points in the chaotic region will follow the same asymptotic behavior, while points in the regular region will stretch linearly at best. In the worst case, high-viscosity droplets in the regular region may tumble indefinitely,
never achieving large stretch values.

While the present model does not include interfacial tension, extensions of the model and other closely related theories do include retraction of the droplet due to interfacial tension [23–27]. In contrast to the exact solution underlying the present model, all ellipsoidal droplet models that include interfacial tension are necessarily approximate. The application of these models to mixing analysis is an obvious extension to this work.

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References


