Steady streaming flows in viscoelastic liquids

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We discuss experimental investigations on steady streaming flows of dilute and semi-dilute polymer solutions in microfluidic devices. The effect of non-Newtonian behavior on steady streaming for different model fluids is determined by characterizing the evolution of the inner streaming layer with particle tracking velocimetry, as a function of oscillation frequency. We find that steady streaming velocity profiles in constant-viscosity elastic liquids are qualitatively similar to those in Newtonian liquids. Steady streaming velocity profiles in shear thinning liquids, however, display two unique features: (i) a non-monotonic behavior of the inner streaming layer with frequency, first increasing then decreasing in width, and (ii) a clear asymmetry in the flow profile at high frequencies.

I. INTRODUCTION

Steady streaming or acoustic streaming flows broadly refer to the steady inertial rectified flows that occur in the presence of a primary oscillatory flow in fluids [1–3]. The phenomenon of steady streaming is a viable tool for microfluidic applications ranging from particle sorting [4–6], mixing [7–9], and trapping [10–12]. There is therefore considerable interest in applying these flows to biological systems as a purely hydrodynamic method of manipulation [13, 14]. While non-Newtonian fluids are frequently encountered in biological systems, steady streaming in non-Newtonian fluids remains poorly understood, particularly in the context of small scale flows where there is maximum potential for application.

Comprehensive experimental investigations of steady streaming in non-Newtonian liquids are limited and predominantly consider the streaming flow of dilute polymer solutions around a cylinder oscillating at low frequencies in the range of $10 \leq f \leq 100$ Hz [15–17]. The classical problem of steady inertial flows around a cylinder of size $a$, oscillating with an angular frequency $\omega$ and a far-field amplitude $s$ in a Newtonian liquid is theoretically well-understood [3, 16–20]. Therefore, it provides a convenient baseline for studying steady streaming flows in non-Newtonian liquids.

Compared to a Newtonian liquid, dramatic changes in the steady streaming flow field were observed in dilute polymeric solutions [16]. The flow field was found to reverse in direction at large distances from the cylinder. This effect was attributed to the growth of the inner streaming layer which usually drives the much larger outer vortex of the opposite sense in Newtonian liquids. The growing inner streaming layer completely displaces the driven outer vortex at sufficiently large frequencies giving the appearance of a reversed flow [21].

The growth of the inner streaming layer with elasticity was theoretically predicted using both Oldroyd B [22] and Walters B [10] constitutive models through asymptotic analysis. The analysis for the Oldroyd B model was extended using the method of matched asymptotic expansions to capture the complete displacement of the outer vortex by the growing inner layer [23]. A complementary analysis, using a second order integral constitutive model, for oscillating spheres yielded similar results [24].

Subsequent comparison with experimental results established that the Upper-convected Maxwell model was sufficient for quantitative agreement [21]. The time dependent nature of the oscillatory flow and the high sensitivity to dilute polymeric solutions motivated the use of steady streaming flows as a rheological tool to characterize dilute drag reducing polymer solutions [17]. It was found that polymers of large molecular weight, which display Carreau shear thinning in the $0.1 - 10$ s$^{-1}$ range of shear rates, exhibit deviation from Newtonian behavior in the frequency range of $10 \leq f \leq 100$ Hz. The deviation was characterized by the relaxation time associated with the Upper-convected Maxwell model used for fitting. The resultant relaxation times were far less than those obtained from the Carreau shear thinning and this was attributed to differences in polymer relaxation times with the nature of the flow. Deviations from Newtonian behavior were found to become more pronounced with increasing frequency and it was postulated that other dilute polymeric solutions would display qualitative differences in streaming at sufficiently high frequencies. The obtained relaxation times were found to correlate well with the drag reduction performance of the respective polymeric solution.

The effectiveness of steady streaming as a microrheological tool has been reconsidered recently and demonstrated for low-viscosity Newtonian liquids in microfluidic devices [25]. In this work, we experimentally investigate steady streaming in non-Newtonian fluids using microfluidic devices. In addition to the common advantages in microfluidic techniques such as small sample volumes and high throughput, lower inertia due to small length scales $O(100 \mu m)$ allows us to access higher frequency regimes which test existing theories on the relationship between steady streaming flow fields and polymer relaxation time. The motivation for our study is three-fold: First, to make quantitative observations on...
the inner streaming layer of non-Newtonian liquids in the context of microfluidics where they stand to be most likely encountered in contemporary applications. Second, to explore the relationship between the bulk rheology and molecular properties of model dilute and semi-dilute polymeric solutions with the observed streaming flows, and third, to illustrate the possible mechanism by which the non-Newtonian behavior manifests in streaming.

II. EXPERIMENTAL METHODS

A. Microfluidic steady streaming

Experiments were performed in microfluidic devices molded in PDMS, consisting of a straight channel 20 mm long, 5 mm wide, and 200 μm tall. A fixed cylindrical post with radius of \( a = 100 \) μm was manufactured at the center of the straight channel. An oscillatory flow field, \( U(t) = U_\infty \cos(\omega t) \), was setup in the channel through an external oscillating pressure signal generated using an electro-acoustic transducer over a range of frequencies, \( 40 \leq f \leq 1200 \) Hz, and \( \omega = 2\pi f \) is the angular frequency. The oscillation amplitude was independently controlled over a range, \( 5 \leq s \leq 70 \) μm, such that the non-dimensional amplitude \( (\epsilon = s/a \ll 1) \) is small. For a given data set, the value of \( \epsilon^2 \omega \) was maintained approximately constant. The rate of strain, which scales as \( O(\epsilon \omega) \), increases with frequency by a factor of 4 from lowest to highest values. The Reynolds number, \( Re = \omega a^2/\nu \), and the streaming Reynolds number, \( Re_s = \omega s^2/\nu \), correspond to a range of \( 2 \leq Re \leq 50 \) and \( 0.1 \leq Re_s \leq 1 \), respectively, for deionized water (\( \nu = 0.949 \times 10^{-6} \) m²/s) over the entire frequency range investigated here.

Polystyrene tracer particles, 0.93 μm in diameter, were observed at the mid-height of the straight channel using bright field microscopy at 20× magnification. Images were acquired using a scientific CMOS camera where the sampling frequencies are greater than (high-speed) or perfect divisors (stroboscopic) of the oscillatory flow frequency. High-speed imaging provided high fidelity observation of the oscillatory flow component and measurement of the oscillation amplitude \( s \), while stroboscopic imaging was used to measure the streaming velocity fields. Experiments were performed at room temperature, maintained at 20 °C.

B. Polymer solutions

Solutions were prepared by step-wise dissolution of different polymers into deionized (DI) water with gentle agitation of 60 rpm for 4-6 hours. After initial dispersion, the required final concentration was achieved by successive dilution with DI water or saline solution (to 0.5 M NaCl). The polymers used in this study were xanthan gum (XG, \( 2.7 \times 10^6 \) MW, Sigma Aldrich G1253), non-ionic polyacrylamide (PAA, \( 6 \times 10^6 \) MW, Polysciences 02806), and polyacrylamide-acrylate co-polymer with a 30% degree of hydrolysis (hPAA, \( 18 \times 10^6 \) MW, Polysciences 18522). The properties of the polymer molecules and the polymer solutions are listed in Table I. Estimates of the overlap concentration \( c^* \) for these model solutions have been reported in literature [26–28]. Here, the concentration refers to parts-per-million (ppm) by weight, that is, 1 ppm = \( 10^{-6} \) gram of solute per gram of solvent.

C. Bulk rheology

We characterize all fluids, Newtonian and non-Newtonian, using a temperature-controlled cone-and-plate geometry rheometer (strain-controlled Ares G2, TA Instruments) over a range of constant shear rates at 25

![FIG. 1. Steady state shear viscosity measurements of dilute and semi-dilute polymer solutions used in this study. The characteristic shear rate in steady streaming experiments, depicted by the shaded area, range from \( 30 < \dot{\gamma} < 200 \) s⁻¹.](image-url)
FIG. 2. Steady streaming profiles in Newtonian and non-Newtonian liquids at an oscillation frequency of 600 Hz around a cylinder with radius of 100 µm. (Top row) Pathlines of tracer particles for (a) DI water, (b) 4000 ppm non-ionic polyacrylamide, (c) 400 ppm xanthan gum, and (d) 50 ppm hydrolyzed polyacrylamide. (Bottom row) Corresponding steady streaming velocity field magnitude, normalized by the maximum velocity, obtained from particle tracking velocimetry. High-velocity regions, light (or yellow) regions of the colormap, are located near the cylinder boundary. The red arrow (bottom row, a) demonstrates the location of the normalized tangential velocity, used to measure the full width at half maximum in Figure 3 and Figure 4.

°C. The steady state shear viscosity as a function of shear rate for a few of the polymer solutions is shown in Figure 1. We find strong shear thinning behavior for the most concentrated xanthum gum (XG 1000) and polyacrylamide-acrylate (hPAA 50) solutions. We also find a decreased shear thinning behavior at lower concentration solutions of XG 400 ppm and hPAA 20 ppm (not shown). Minimal shear thinning was observed for the PAA 4000 ppm and hPAA in 0.5 M NaCl solutions which can be considered Boger fluids, or constant viscosity elastic liquids [29, 30]. The characteristic shear rate in steady streaming experiments is estimated by 0.45 ω and has been depicted by the shaded region in Figure 1 for the parameters used here.

The relaxation time for the shear thinning polymer solutions were estimated by fitting a Carreau model to the shear rheology data [31]:

\[
\eta'(\dot{\gamma}) = \eta_\infty + (\eta_0 - \eta_\infty) \left[1 + (\tau_C \dot{\gamma})^2 \right]^{(n-1)/2},
\]

where \(\eta\) is the viscosity, \(\eta_0\) is the zero-shear viscosity, \(\eta_\infty\) is the infinite-shear viscosity, \(n\) is the power-law index, \(\tau\) is the estimated relaxation time. The relaxation times for the constant viscosity polymer solutions were estimated by fitting a Carreau model to the shear rheology data [31]:

\[
\eta(\dot{\gamma}) = \eta_\infty + (\eta_0 - \eta_\infty) \left[1 + (\tau \dot{\gamma})^2 \right]^{(n-1)/2},
\]

where \(\eta_s\) is the solvent viscosity, \(R\) is the universal gas constant, and \(\eta_0\) is the intrinsic shear viscosity at zero shear rate. Here, we take \(\eta_0 = \eta_\infty = 1 \times 10^{-3}\) Pa s (DI water) and \(\eta_0\) was approximated for the given semi-dilute solutions based on the zero-shear viscosity and solvent viscosity. The resulting values of the relaxation times are listed in Table I. Note that the Rouse theory cannot be applied to polyelectrolyte solutions such as XG and hPAA in the absence of salts.

III. RESULTS

The comparison between steady streaming flow profiles in Newtonian and non-Newtonian liquids at an oscillation frequency of 600 Hz is shown in Figure 2. The particle pathlines (top row) are generated from minimum projections of stroboscopic images and the velocity field magnitudes (bottom row) are obtained from particle tracking velocimetry. In all cases, the rectified flow moves toward the cylinder along the axis of oscillation and away from the cylinder normal to the axis of oscillation. Similarly, the quadrupolar topology of the streaming flow field is preserved, consisting of four vortices with distinct centers symmetric with respect to the cylinder.

The pathlines and velocity field magnitude for a Newtonian liquid, deionized water are shown in Figure 2(a, top and bottom). High flow velocities are located in the regions between the cylinder surface and the eddy center, and near the cylinder surface, perpendicular to the axis...
of oscillation. This latter difference, compared to the velocity along the axis, is because the velocity fields are Lagrangian. It can be captured by applying the Stokes drift correction to Eulerian velocity fields [20].

The steady streaming pathlines and velocity field magnitude of a Boger fluid, PAA 4000 ppm, are qualitatively similar to that of a Newtonian liquid, shown in Figure 2 (b, top and bottom). The main difference is the eddy center distance from the cylinder, which is greater for PAA 4000 ppm than for DI. As recently shown by the application of steady streaming for viscometry, a greater eddy center distance would indicate a liquid of higher viscosity [25], which agrees with steady state shear viscosity measurements in Figure 4.

In shear thinning liquids, a qualitative and quantitative difference is observed in particle pathlines and velocity field magnitudes. For XG 400 ppm solutions, the angular position of the eddy centers move closer to the axis of oscillation, shown in Figure 2 (c, top). The velocity field has also observably changed with the velocity maximum normal to the axis of oscillation separated into two regions by a local minimum instead, shown in Figure 2 (c, bottom). For shear thinning hPAA 50 ppm solutions, a more prominent change is seen where the angular position of the eddy center as well as the velocity maximum are moved considerably towards the axis of oscillation, Figure 2 (d, top and bottom).

To quantify differences in the steady streaming profile for various fluids over a range of frequencies, we will examine the normalized tangential velocity profile as a function of the dimensionless radial position $r/a$ along a transect from the cylinder surface through the eddy center. The transect is depicted by the red arrow passing through the eddy center in Figure 2 (a, bottom). A representative experimentally measured velocity profile for a Newtonian liquid is shown in Figure 2 (a, bottom). The particular parameter of interest is the full width at half maximum (FWHM) of the second peak, beyond the eddy center. The FWHM in this region quantifies the size of the inner streaming layer and is the preferred measure on account of sensitivity and reliability.

The variation of the FWHM with frequency for Boger fluids, PAA 4000 ppm and hPAA 500 ppm, and Newtonian liquids, DI water and aqueous 60% glycerol (w/w) with a dynamic viscosity of 5.9 cP, is shown in Figure 4 (a). The Newtonian liquids exhibit slow monotonic decrease with increasing oscillation frequency. The Boger fluids show a nominally faster rate of decrease with increasing frequency, without any discernible features.

This is in sharp contrast with the observations for shear thinning liquids. All four shear thinning polymer solutions exhibit distinct peaks in their FWHM with increasing oscillation frequency, shown in Figure 4 (b). For hPAA 50 ppm solutions, the FWHM grows rapidly from 75 Hz until 150 Hz, which is in agreement with previous observations [17]. However, the FWHM begins to decrease with further increasing frequency until its variation is similar to that of the solvent. Similar trends are observed for hPAA 20 ppm solutions, with a more rapid increase and subsequent rapid decrease to Newtonian-like behavior with the maximum FWHM observed at 75 Hz. The XG 1000 ppm and XG 400 ppm solutions also behave in a manner consistent with our observations for the hPAA solutions, exhibiting a distinct peak in the FWHM followed by a decrease to Newtonian-like behavior with increasing frequency. The key difference between the two polymers is the rate at which they decrease at large frequencies, which is slower in XG solutions. This is in agreement with the qualitative observations where the pathlines and the corresponding velocity field magnitudes are not as distorted in XG as they are in hPAA, even at 600 Hz, shown in Figure 2 (c) and (d). Further increase in frequency is not found to significantly alter the velocity profile of hPAA 20 ppm and hPAA 50 ppm solutions. However, observations at 800 – 1000 Hz for XG 400 ppm show a gradual approach towards streaming and velocity fields similar to hPAA 50 ppm, and this approach is slower still for XG 1000 ppm, therefore reflected completely by the evolution of the FWHM shown in Figure 4 at relatively high frequencies.

IV. DISCUSSION

While the growth of the inner streaming layer for frequencies $f < 100$ Hz has been experimentally observed [17] and predicted by existing models for a cylinder oscillating in a viscoelastic liquid [18, 22, 23], the latter shrinking of the layer has not been experimentally re-
FIG. 4. Evolution of inner streaming layer characterized by the full width at half maximum for Newtonian and non-Newtonian liquids as a function of oscillation frequency in microfluidic devices. (a) The FWHM of Newtonian liquids and Boger fluids exhibit monotonic decrease with increasing oscillation frequency. (b) The FWHM of shear thinning liquids, however, increases to a maximum value and then decreases with increasing oscillation frequency.

FIG. 5. Pathlines of tracer particles illustrating experimental evidence of the predicted cascading inertio-elastic vortices. A set of secondary, same-sense vortices is shown for hPAA 50 ppm solutions at 800 Hz.

FIG. 6. Pathlines of tracer particles illustrating the primary oscillatory flow, captured using high-speed imaging. A half period of oscillation, as the liquid moves from left to right, illustrates regions of (i) extensional flow along the axis of oscillation and (ii) shear flow perpendicular to the axis of oscillation.

ported before. The most likely explanation for the decrease of the inner streaming layer thickness after the initial growth is due to a competition between the effects of inertia and elasticity. The inertia is characterized by the streaming Reynolds number Re_s, and the elasticity by the Deborah number, De = τω, where τ is the characteristic relaxation time of the liquid.

The competition between inertia and elasticity was suggested in the theoretical analysis for an oscillating sphere [24], where increasing the De for a given Re_s was found to increase the thickness of the inner streaming layer. Increasing Re_s for a given finite De and shear thinning, however, led to two theoretical predictions that have yet to be experimentally verified; (i) the decreasing of the thickness of the inner-most streaming layer (studied here) with increasing Re, and (ii) a set of cascad-

The relationship between shear thinning and the relaxation time remains ambiguous. The relaxation time associated with the growth of the inner streaming layer was found to be far less than that associated with the
Carreau model for hPAA \[17\]. This was attributed to the longest relaxation time not being the characteristic relaxation time of the oscillatory flow field. However, we see that for both the XG 1000 ppm and hPAA 50 ppm solutions, the Carreau times are nearly identical, and there is very good agreement in Figure 1(b). Likewise, for the Boger fluids studied, the Carreau times are too small to be measurable coincident with the lack of non-monotonic behavior. For the PAA 4000 ppm and hPAA 500 ppm solutions at 400 Hz, \( \text{De} = 6.87 \) and \( \text{De} = 28.2 \), respectively, based on established values of molecular relaxation time, despite which no change in the streaming profile and velocity field is observed.

One possible reason we do not observe a difference in the streaming profiles of Boger fluids is that the resolution of our data points is insufficient to record the transition, as suggested by the increasing sharpness with decreasing viscosity seen from Figure 1(b). However, no qualitative change is seen in the velocity field, even at high frequencies. Another possibility may be that coil-stretch transitions are not activated \[32, 33\]. Flexible molecules experience large deformation at strain rates comparable to \( \tau_R^{-1} \), the reciprocal of the Rouse time, which was never achieved in our experiments.

The rates of strain scale as \( \mathcal{O}(\epsilon \omega) \) and can be determined from the primary underlying oscillatory flow, shown in Figure 4 for DI water at 100 Hz. Regions along the axis of oscillation, in front and behind the cylinder, are completely extensional, whereas regions perpendicular to the axis of oscillation, above and below the cylinder, are completely shearing. The spatio-temporal average rate of shear and extension (magnitude), obtained from theory \[19\] for conditions shown in Figure 6, are approximately 0.45\( \epsilon \omega \) and 0.15\( \epsilon \omega \), respectively. In contrast, the maximum instantaneous shear and extensional rates for the same conditions are 3.73\( \epsilon \omega \) and 0.3\( \epsilon \omega \), respectively. This is because the shear is localized close to the cylinder surface as compared to the extension. We are therefore led to believe that shear thinning is responsible at least for the qualitative change seen in the inner vortex region where it is maximum in the oscillatory flow field.

V. CONCLUSIONS

In this work, we have studied the inner steady streaming layer of dilute and semi-dilute polymer solutions of both Boger and Carreau steady shear behavior in the vicinity of a cylindrical obstacle using microfluidics. Qualitative differences in the velocity fields are observed for Carreau shear thinning liquids. The width of the inner streaming layer is characterized by the full width at half maximum of the velocity profile along the eddy center. We find that shear thinning liquids exhibit an increase in width followed by a decrease with increasing frequency. This behavior is attributed to a competition between elasticity and inertia, with inertia dominating at high frequencies. Boger fluids show a monotonic decrease in the width with increasing frequency, without any qualitative change in the flow profile even for oscillation periods considerably smaller than the molecular relaxation time. Further experiments in the 10 – 100 kHz frequency range are necessary to elucidate the role of shear thinning on the growth of the inner streaming layer.

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