Evanescent-wave particle velocimetry studies of combined electroosmotic and Poiseuille flow

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ABSTRACT

Electric fields, i.e., voltage gradients, are commonly used to drive flows in microfluidic devices because the resulting electroosmotic (EO) flow, which is uniform outside the very thin counterion screening layer next to the charged wall, has less convective dispersion than Poiseuille flow. Recent evanescent-wave particle velocimetry studies suggest, based on measurements of near-wall particle distributions, that colloidal neutrally buoyant particle tracers of radii \( a = O(0.1-1 \, \mu\text{m}) \) experience a repulsive wall-normal force in the presence of an electric field of magnitude \( E \) applied parallel to the wall of \( O(10^{14} \, \text{N}) \). The magnitude of the force appears to scale as \( E^2 \) and \( a^2 \), in agreement with theoretical predictions of a nonlinear “dielectrophoretic-like” force, although the experimental estimates of the actual force magnitude are 10–30 times greater than the theoretical predictions.

Given that the force magnitude is proportional to \( a^2 \), particles of different radii will have different average wall-particle separations, and therefore sample different average velocities, in a nonuniform flow in the presence of an electric field. Evanescent-wave particle velocimetry was therefore used to study the combined EO and Poiseuille flow of dilute aqueous monovalent electrolyte solutions at \( E < 33 \, \text{V/cm} \) and pressure gradients \( \Delta P/L < 1.1 \, \text{Bar/m} \) in 30 \( \mu\text{m} \) fused-silica microchannels using \( a = 125 \, \text{nm} \) and \( a = 245 \, \text{nm} \) fluorescent polystyrene (PS) tracer particles.

For this creeping flow, the velocity of the combined flow will be a simple shear flow due to the superposition of the EO and Poiseuille flow velocities. As expected, the average velocities measured by tracers of different sizes in combined flow under otherwise identical conditions differ, due to the discrepancy in their average wall-particle separations. The velocity profiles measured by evanescent-wave particle velocimetry for both tracers are, however, in good agreement with each other and with theory after accounting for the wall-normal positions of the particles.

The effects of the repulsive wall-normal force observed in EO flow, and the shear-induced electrokinetic lift force first reported twenty years ago in Poiseuille flow, are observed in the near-wall distributions of the larger particles. Interestingly, a repulsive force with an estimated magnitude that appears to exceed the sum of these two forces is observed in combined electroosmotic and Poiseuille flow for the \( a = 245 \, \text{nm} \) particles. The results suggest that there is a nonlinear interaction between the electric field and flow shear, and that combining these two effects may lead to new ways to manipulate microparticles near surfaces.

1. INTRODUCTION

Microfluidic devices, which often involve the flow of incompressible Newtonian liquids through channels with diameters of 500 \( \mu\text{m} \) or less, are of interest in a variety of applications including “point of care” medical diagnostics, and thermal management. Given that diffusion processes, with time scales proportional to the square of the length scale, become much faster at the micron scale, mass and thermal transport are enhanced in microfluidic devices such as “Labs-on-a-Chip” (LoC) and micro-total analysis systems (µTAS). Surface forces, which scale with area, are also enhanced compared with body forces, which scale with volume, at these small length scales. Given that most surface forces are very short-ranged, and hence usually significant only within ~1 \( \mu\text{m} \) of the (wall) surface, understanding how surface forces affect transport requires characterizing flows in this near-wall region.

Particle velocimetry methods that estimate flow velocities from the displacements of particle tracers illuminated by evanescent waves generated by the total internal reflection (TIR) of light at the wall-fluid refractive-index interface have been used by various researchers to study microchannel flows over the first ~0.5 \( \mu\text{m} \) next to the wall [1–4]. Although this type of illumination inherently limits this method to near-wall flows, the spatial resolution of evanescent-wave particle velocimetry is significantly better than that of micro-particle image velocimetry (µPIV), with its typical minimum depth of correlation of 2 \( \mu\text{m} \).
Accurately mapping the displacement of particle tracers over a known time interval to actual flow velocities this close to the wall can, however, be an issue. Most µPIV and evanescent-wave particle velocimetry studies use (nearly) neutrally buoyant fluorescent dielectric polystyrene (PS) particle tracers of radii $a = 100$ nm–2 µm that are suspended in an electrically conducting aqueous solution at low enough volume fractions so that interparticle interactions are negligible. Nevertheless, even particles that are suspended in a quiescent fluid will be nonuniformly distributed along the wall-normal direction because they are subject to both (usually attractive) van der Waals forces and (usually repulsive) electrostatic forces, as described by the classic Derjaguin-Landau-Verwey-Overbeek (DLVO) theory of colloid science [5].

Moreover, suspended particles convected by a flow can, in many cases, be subject to additional forces. Neutrally buoyant $a = 2.5 - 5$ µm particles convected by a simple shear flow, as is the case for near-wall Poiseuille flow driven by a pressure gradient, experience a repulsive force due to electroviscous effects [6]. Although this phenomenon was first observed in the 1980s, we still lack a theoretical understanding of this shear-induced electrokinetic lift force, and very recent work suggests that electroviscous effects due to both spatial nonuniformities in the electric field and viscous stresses due to flow perturbations must be considered to accurately model this force [7].

Recently, it has been shown that smaller neutrally buoyant $a = 0.2 - 0.5$ µm particles convected by a uniform flow in the presence of an electric field parallel to the wall, as is the case for electroosmotic (EO) flow driven by a voltage gradient, also experience a repulsive force [8]. Initial results over a limited range of parameters suggest that the force magnitude is proportional to $E^2$ and $a^2$, in agreement with a model of a “dielectrophoretic-like” repulsive force due to nonuniformities in the local electric field in gap between the particle and wall [9]. However, the experimental estimates of the force magnitudes are one to two orders of magnitude greater than the model predictions, suggesting that we also lack a theoretical understanding of this nonlinear repulsive force.

Given that microchannel flows are often driven by voltage and pressure gradients, these experimental observations suggest that the tracer particles within 1 µm of the wall will have a nonuniform distribution along the $z$-coordinate normal to the wall in many particle velocimetry studies in microfluidic devices. Furthermore, this distribution will depend on the flow parameters, and cannot at present be predicted by theory.

Hence accurate near-wall particle velocimetry requires measuring not only the particle position in the plane parallel to the wall, but also the distance between the tracer and the wall. And since the magnitude of the dielectrophoretic-like repulsive force depends upon $a$, particles of different sizes will have different average velocities in a nonuniform flow if there is also an electric field parallel to the wall.

Evanescent-wave particle velocimetry was therefore used to study near-wall particle dynamics of $a = 125$ nm and 245 nm fluorescent PS tracers in a nonuniform flow with an electric field parallel to the wall, specifically in combined EO and Poiseuille flow, through wet-etched fused-silica microchannels with nominal cross-sectional dimensions of 30 µm $\times$ 300 µm. The particle-wall separation of each tracer $h$ was determined from the particle image intensity, assuming that this measure of the particle brightness, like the illumination intensity, decayed exponentially in $h$, and the ensemble of $h$ values for $O(10^3)$ tracers was used to determine the near-wall particle distribution. The particle images were then “binned” based on their brightness, and flow velocities were estimated using particle-tracking methods in three distinct layers within ~500 nm of the wall. Given that the flow velocity should simply be the superposition of the uniform flow due to EO flow and the simple shear flow due to the Poiseuille flow (since the parabolic velocity profile is well-approximated by a linearly varying profile this close to the wall) for these creeping flows, the velocity data were also used to validate the near-wall particle distribution estimates.

The remainder of this paper is organized as follows. Section 2 describes the experimental studies and the data processing. Section 3 presents and discusses the results, most of which are for the larger $a = 245$ nm particles. Finally, the results are summarized in Section 4.

### 2. EXPERIMENTS AND IMAGE PROCESSING

Both electroosmotic (EO) and Poiseuille flows were studied in wet-etched fused-silica microchannels with trapezoidal cross-sections of depth ~30 µm and width ~300 µm at electric fields of magnitude $E < 33$ V/cm and pressure gradients $\Delta p/L < 1.1$ Bar/m, corresponding to a simple shear flow with a shear rate $\dot{\gamma} < 1600$ s$^{-1}$. The EO flow was driven by an electric field parallel to the wall generated either by a high-voltage dc power supply (Stanford Research Systems PS325) or a low-voltage dc power supply (Instek GPR-3510HD) to platinum electrodes sealed to the four-way connectors at the upstream and downstream reservoirs. The pressure gradient required for the Poiseuille flow was generated hydrostatically using a reservoir of double distilled deionized (DDI) water with an adjustable height that was
attached to the reservoir at the inlet of the microchannel (a ~5 mm segment of ID 4 mm borosilicate glass tubing) with Tygon tubing and a 4-way tubing connector (Fisher Scientific 15-315-32B). An identical tubing connector was attached to the reservoir at the exit of the microchannel; in all cases the water height at the exit remains constant throughout the experimental run (Fig. 1 [top]).

![Diagram of the setup](image)

**Figure 1** Sketch showing the arrangement of the 4-way tubing connectors at the channel inlet and exit [top] and the evanescent-wave illumination of the microchannel cross-section [bottom]. Note that the flow direction is into the page, and $z$ is the wall-normal direction, in the bottom sketch.

The EO and Poiseuille flows were in the same direction in these experiments. The electric field magnitudes were kept relatively low in these experiments to minimize Joule heating; in all cases, the temperature of the fluid, measured by a thermocouple at the exit in some of the experiments, was within 1 °C of the ambient temperature, which varied from 19 °C to 21 °C.

The working fluid was an aqueous sodium tetraborate (Na$_2$B$_4$O$_7$) solution which was diluted from a 20 mM Na$_2$B$_4$O$_7$ stock solution to a molar concentration of 1 mM with DDI for each experiment. The DDI water had an initial resistivity in excess of 16 MΩ·cm. The fluid was then seeded with fluorescent carboxylate-terminated polystyrene (PS) particles of radii $a = 125$ nm with a manufacturer-quoted polydispersity of 4.5 nm (Invitrogen F8811) or $a = 245 \pm 7.5$ nm (Invitrogen F8812). The bulk particle number density of the working fluid $c_\infty = 2.7 \times 10^{16}$ m$^{-3}$, corresponding to volume fractions of $2.2 \times 10^{-4}$ and $1.7 \times 10^{-3}$ for the $a = 125$ nm and $a = 245$ nm particles, respectively. The particle solution was sonicated for 15 min and filtered through syringe filters with 0.45 µm (Millipore SLHV033RS) or 0.8 µm (Millipore SLAA033SS) pore sizes for the $a = 125$ nm and $a = 245$ nm particles, respectively, to remove aggregated particles. After filtration, the solutions were degassed under vacuum at an absolute pressure of 0.1 kPa for another 15 min.

The microchannel was cleaned before and after each experimental run by flowing DDI water, acetone, and methanol, followed again by DDI water, 1 M sodium hydroxide (NaOH) (to create a “fresh” layer of silanol surface groups) and 1 mM Na$_2$B$_4$O$_7$. After cleaning, the degassed working fluid was injected into the tubing until it filled the tubing, the 4-way connectors and the microchannel. After checking to make sure that there were no air bubbles, the microchannel
was mounted on the stage of an inverted epi-fluorescence microscope (Leica DMIRE2) and illuminated with evanescent waves which were generated at the bottom surface of the microchannel (with a manufacturer-quoted rms roughness of 3 nm) from the total internal reflection (TIR) of an argon-ion laser beam coupled into the substrate with a fused-silica isosceles right triangle prism (Fig. 1 [bottom]).

At the beginning of each experimental run for the combined EO and Poiseuille flow experiments, the pressure gradient $\Delta p/\ell$ was set by adjusting the height of the reservoir, and an image sequence was acquired starting at $E = 0$, and then at progressively increasing values of $E$. As will be discussed in the next Section, the particles were strongly repelled from the wall in the combined flow experiments, and data were only obtained at electric field magnitudes $E \leq 8.8$ V/cm because there were too few particles in the near-wall region illuminated by the evanescent wave to obtain meaningful statistics on the particle distributions at higher $E$. No images were acquired for the first 120 s of the combined flow experiments for each different value of $E$ to ensure that the flow velocity reached steady-state. After running experiments over the entire range of electric fields, $E$ was set again to zero, and $\Delta p/\ell$ was adjusted to a new value. It took much longer for the flow to reach steady-state after changing $\Delta p/\ell$, and so no images were acquired for the first 15 min in these cases.

At the end of each sequence of experiments, a magnesium chloride (MgCl$_2$) solution (of a few mM molar concentration) was injected into the microchannel. The divalent cations in the solution cause the particles to become electrostatically attached to the channel surface, and images of these attached particles obtained with the same illumination and imaging setup as that used in the actual experiments were used to determine $I'_p$, the area-averaged intensity of particles touching the wall.

In these studies, separate experiments were performed with particles of different radii. Because the particles of different radii were labeled with different fluorophores, they were illuminated at different wavelengths $\lambda$ and imaged using different filters. The $a = 125$ nm green particles, which have an emission maximum at 513 nm, were illuminated by the 488 nm line from the argon-ion laser, and imaged through a 525 ± 25 nm bandpass filter. The $a = 245$ nm red particles, which have an emission maximum at 606 nm, were illuminated by the 514.5 nm line and imaged through a 615 nm longpass wavelength filter.

An acousto-optic modulator (AOM) was used to “shutter” the laser and generate two images of the particles convected by the flow of exposure 0.5 ms spaced by a time interval $\Delta t = 2$ ms; the image pairs were spaced 200 ms apart. A sequence of 500 image pairs (= 1000 images) were recorded of each experimental case over a total data acquisition time of about 100 s. The fluorescence from the particles near the wall was imaged through a magnification 63, numerical aperture 0.7 microscope objective (Leica PL Fluotar L) onto an electron multiplying charge-coupled device (EMCCD) camera (Hamamatsu C9100-13). The resulting 512×144 pixels images, with physical dimensions of 130 $\mu$m × 37 $\mu$m, were of the flow in the center of the microchannel to minimize any effects from the side walls.

In each of the EMCCD images, the location of the center of each particle was determined using cross-correlation with a 2D Gaussian function. After identifying and removing the images of overlapping or aggregated tracers, the displacements of the remaining particles in the plane parallel to the wall were determined by matching particles between the two images in the pair to their nearest neighbor, since the average interparticle distance was significantly greater than the particle displacements due to convection.

The particle-wall separation $h$, or the distance between the particle edge and the wall, was next determined from the particle image intensity $I_p$, which was defined to be the area-averaged integral of the grayscale values in the image. Assuming that the particle image intensity has the same exponential decay as the evanescent-wave illumination where the length scale of the decay is the intensity-based penetration depth $z_p$,

$$I_p(h) = I'_p \exp \left( -\frac{h}{z_p} \right)$$  \hspace{1cm} (1)

The measured penetration depth $z_p$ varied from 110 nm to 120 nm. The ensemble of particle-wall separations determined over about $3 \times 10^4$ particle images was then used to calculate the particle number density profile $c(h)$ over bins with a width (dimension normal to the wall) of 20 nm.

The displacements of the $a = 125$ nm and $a = 245$ nm particles were then divided into three layers, each containing a similar number of particle samples, based on $h$:

I. $0 \leq h \leq 100$ nm
II. $100$ nm $\leq h \leq 200$ nm
III. 200 nm \leq h \leq 300 nm

The particle velocity components parallel to the wall were then simply the particle displacement divided by \( \Delta t \).

This measured particle velocity \( u_p \) differs from the actual flow velocity \( U \) in the presence of an electric field due to particle electrophoresis, so

\[
   u_p = U + u_{ep} = U + \frac{\varepsilon \zeta_p E}{\mu}
\]

where the electrophoretic velocity \( u_{ep} \) is given by the Helmholtz-Smoluchowski relation since the hindrance of the electrophoretic mobility by particle-wall interactions is negligible [8]. Here, \( \varepsilon \) and \( \mu \) are the permittivity and absolute viscosity, respectively, of the fluid, and \( \zeta_p \) is the particle zeta-potential. The zeta-potentials of the particles suspended in the working fluid were measured using laser-Doppler microelectrophoresis with a Malvern Zetasizer; \( \zeta_p = -67.8 \pm 1.0 \) mV (average \pm standard deviation) for the \( a = 125 \) nm particles and \( \zeta_p = -49.9 \pm 0.6 \) mV for the \( a = 245 \) nm particles.

3. RESULTS AND DISCUSSION

Figure 2 shows the flow velocity profiles \( U(z) \) for steady Poiseuille flow (no electric field) at pressure gradients \( \Delta p/L = 0.43 \) Bar/m (circles), 0.74 Bar/m (triangles), and 1.04 Bar/m (squares) measured by evanescent-wave particle velocimetry with \( a = 125 \) nm (open symbols) and \( a = 245 \) nm (filled symbols) particles for \( z \leq 540 \) nm. The data are consistently higher than the velocity profile predicted by the analytical solution (solid lines), and the discrepancy appears to increase with \( \Delta p/L \). Given, however, that the slope of the data are in good agreement with the predicted shear rate, and that the data for both \( a \) are in good agreement, we hypothesize that the discrepancy may be due to calibration errors, specifically in determining the intensity of a particle touching the wall \( I_p \).

Figure 3 shows the flow velocities for combined EO and Poiseuille flow at an electric field magnitude \( E = 8.8 \) V/cm and \( \Delta p/L = 0.43 \) Bar/m (circles), and 1.04 Bar/m (squares). Note that the flow velocities were estimated by subtracting the electrophoretic velocity (cf. Eq. 2) from the measured velocities of the \( a = 125 \) nm (open symbols) and \( a = 245 \) nm (filled symbols) tracers. The EO flow creates an “offset” in the flow velocity, and the slope of the data are in good agreement with that of the Poiseuille flow (dashed lines). Although results are not shown here, the velocity “offset” due to EO flow for \( E = 2.4 \) V/cm–8.8 V/cm is, as expected, proportional to \( E \).
Figure 3  Flow velocity profiles for combined electroosmotic flow at $E = 8.8$ V/cm and Poiseuille flow at $\Delta p/L = 0.43$ Bar/m (circles) and 1.04 Bar/m (squares) estimated from the particle speeds using $a = 125$ nm (open) and $a = 245$ nm (filled symbols) particles. The dashed lines represent the analytical solution for Poiseuille flow at the same values of $\Delta p/L$. The error bars denote the standard deviations over three independent experiments.

The $z$-positions of these velocity data have already been corrected (albeit with some uncertainty) for the nonuniform distribution of the tracers using the procedure described in the previous Section, and the excluded volume effect where the minimum velocity of the particle (center) is that of the flow at $z = a$. It one were to assume instead that the particles are uniformly distributed over particle-wall separations $0 \leq h \leq 300$ nm, the average velocity sampled by the particles in this simple shear flow should be the flow velocity at the average $z$-position of the particle center of $z = a + 150$ nm. Table I compares the average velocity sampled by the $a = 125$ nm and 245 nm particles with that at $z = 275$ nm and 395 nm, respectively, based on our experimental data for combined EO and Poiseuille flow at $E = 8.8$ V/cm. Particles of different sizes should of course have different velocities in this simple shear flow due to excluded volume effects, as can be seen from the “expected velocity” columns. However, the average velocity sampled by the particles is much larger than the expected value for the larger particles at $\Delta p/L = 1.04$ Bar/m. These results demonstrate that particles of different sizes can measure different velocities in the same flow due to excluded volume effects, as can be seen from the “expected velocity” columns. However, the average velocity sampled by the particles is much larger than the expected value for the larger particles at $\Delta p/L = 1.04$ Bar/m. These results demonstrate that particles of different sizes can measure different velocities in the same flow—here, the velocities measured by the $a = 125$ nm particles are in reasonable agreement with the expected values, while those measured by the larger $a = 245$ nm particles are not because of changes in the near-wall particle distribution due to the (relatively) large forces that repel the particles from the wall, as will be seen subsequently. We suspect that the discrepancy is much less for the lower $\Delta p/L$ case because it has a much lower shear rate, and hence the flow velocity varies more slowly with $z$.

Table I. Comparison of average velocities in combined EO flow and Poiseuille flow at $E = 8.8$ V/cm measured by particles of different radii with the expected velocities at the average distance sampled by the particles (if they were uniformly distributed along the $z$-coordinate normal to the wall).

<table>
<thead>
<tr>
<th>$\Delta p/L$</th>
<th>Average velocity $a = 125$ nm [µm/s]</th>
<th>Expected velocity At $z = 275$ nm [µm/s]</th>
<th>Average velocity $a = 245$ nm [µm/s]</th>
<th>Expected velocity at $z = 395$ nm [µm/s]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.43 Bar/m</td>
<td>250 µm/s</td>
<td>270 µm/s</td>
<td>360 µm/s</td>
<td>350 µm/s</td>
</tr>
<tr>
<td>1.04 Bar/m</td>
<td>530 µm/s</td>
<td>520 µm/s</td>
<td>810 µm/s</td>
<td>690 µm/s</td>
</tr>
</tbody>
</table>
why the average velocity sampled by these particles is significantly larger than the expected velocity for the larger micron particles, with the number density decreasing slightly as particle size decreases at a given value of $\Delta p/L$. The decrease in the number of near-wall particles and the resultant increase in the average $z$-position of the tracers, explains why the average velocity sampled by these particles is significantly larger than the expected velocity for the larger $\Delta p/L$ case (Table I).

Figure 4 shows $c(h)$ for the $a = 245$ nm particles convected by Poiseuille flow (with no electric field) at $\Delta p/L = 0.43$ Bar/m and 1.04 Bar/m. The effects of shear-induced electrokinetic lift are evident, even for these sub-micron particles, with the number density decreasing slightly as $\Delta p/L$ increases at a given $h$. The decrease in the number of near-wall particles with $E$ appears to be larger for the higher pressure gradient, at least at a given value of $E$ and $h$, consistent with the expected effect of shear-induced electrokinetic lift. However, the decrease is surprisingly large for the combined flow cases, with numbers densities an order of magnitude less than $c_a$ for $E \geq 7.1$ V/cm. Indeed, as noted earlier, almost no particles were observed in the region illuminated by the evanescent wave at $E > 8.8$ V/cm. This marked decrease in the number of near-wall particles, and the resultant increase in the average $z$-position of the tracers, explains why the average velocity sampled by these particles is significantly larger than the expected velocity for the larger $\Delta p/L$ case (Table I).
Figure 5  Particle number density \(c\) as a function of the particle-wall separation \(h\) for Poiseuille flow at \(\Delta p/L = 0.43\) Bar/m (○), 0.74 Bar/m (△), and 1.04 Bar/m (■) for the \(a = 245\) nm particles. The average estimated uncertainty in \(c\) is at most 6% for \(h \geq 40\) nm. The bulk fluid particle density \(c_\infty = 2.7\times10^{16}\) m\(^{-3}\) is denoted by the dashed line.

Figure 6  Near-wall particle distributions given in terms of the particle number density \(c\) as a function of the particle-wall separation \(h\) (so a particle touching the wall is at \(h = 0\)) for combined EO flow at \(E = 0\) V/cm (○), 2.4 V/cm (△), 4.7 V/cm (□), 7.1 V/cm (▽), and 8.8 V/cm (◇) and Poiseuille flow at (a) \(\Delta p/L = 0.43\) Bar/m and (b) 1.04 Bar/m for the \(a = 245\) nm particles. Both plots have the same vertical axis. The nominal particle number density in the bulk fluid \(c_\infty = 2.7\times10^{16}\) m\(^3\).

This large decrease also suggests that the force driving these particles away from the wall may exceed the sum of the dielectrophoretic-like repulsive and shear-induced electrokinetic lift forces. The magnitude of this force can be estimated from the slope of the potential energy profile \(\phi(h)\) of the particle-wall interaction, which can be from these number density profiles. To calculate \(\phi\), we assumed that the \(c(h)\) obtained from the evanescent-wave particle velocimetry studies from about \(3\times10^4\) samples obtained over a total image acquisition time of \(~100\) s was the steady-
state particle distribution, and that this distribution would hence be a Boltzmann distribution. The particle-wall interaction potential energy is then:

$$\frac{\phi(h)}{kT} = \ln \left( \frac{c(h)}{c_e} \right)$$

(3)

Here, $k$ is the Boltzmann constant and $T$ is the absolute temperature of the fluid. We also assume that the particle-wall interaction potential energy in the bulk fluid is zero, i.e. $\phi \to 0$ as $h \to \infty$. The potential energy profiles determined using Eq. 3 were smoothed using a lowpass filter over a window with a $z$-dimension of 60 nm.

Figure 7 Normalized particle-wall interaction potential energies $\phi/(kT)$ as a function of particle-wall separation $h$ for EO flow (and negligible pressure gradient) at $E = 0$ V/cm (○), 2.4 V/cm (△), 4.7 V/cm (□), 7.1 V/cm (◇), 8.8 V/cm (◇), 15.9 V/cm (×), 23 V/cm (●) and 32.5 V/cm (▲).

Figure 7 shows the resulting particle potentials obtained from the particle distributions of Figure 4 for EO flow (with a negligible pressure gradient) at $E = 0$ V/cm (○), 2.4 V/cm (△), 4.7 V/cm (□), 7.1 V/cm (◇), 8.8 V/cm (◇), 15.9 V/cm (×), 23 V/cm (●) and 32.5 V/cm (▲). Note that $\phi < kT$ for all values of $h$ shown for EO flow driven by $E \leq 8.8$ V/cm. The increase in the slope of $\phi/(kT)$ with $E$ should then correspond to the dielectrophoretic-like repulsive force. A comparison with the slopes of the particle potential profiles (Fig. 8) estimated for the combined EO flow at $E = 0$ V/cm (○), 2.4 V/cm (△), 4.7 V/cm (□), 7.1 V/cm (◇), 8.8 V/cm (◇) and Poiseuille flow at $\Delta p/L = 0.43$ Bar/m suggests that the effect of shear-induced electrokinetic lift is much less evident, as expected.

As an initial attempt to isolate the effect of the electric field, the normalized particle-wall interaction potential energy for the case of “only” Poiseuille flow (i.e., when $E = 0$) was subtracted from those for combined Poiseuille and EO flow at the same pressure gradient. Figure 9 shows $\Delta\phi_{p_e}/(kT)$ as a function of $h$ where $\Delta\phi_{p_e}$ is the difference between the potential energies of the particle-wall interaction for the case of combined Poiseuille flow at $\Delta p/L = 0.43$ Bar/m (Fig. 8) and $\Delta p/L = 1.04$ Bar/m (estimated from the data shown in Fig. 6b) and EO flow at $E = 0$ V/cm (○), 2.4 V/cm (△), 4.7 V/cm (□), 7.1 V/cm (◇), and 8.8 V/cm (◇) (Fig. 8) and the case of (only) Poiseuille flow at the same $\Delta p/L$ (estimated from the data of Fig. 5). A comparison between these potential energy estimates and those shown in Figure 7 (note that Figs. 7 and 9 use the same symbols for a given value of $E$) for “only” EO flow at the same driving electric field magnitude clearly shows that the values for $\Delta\phi_{p_e}/(kT)$ are as much as an order of magnitude greater than those for $\phi/(kT)$ for only EO flow at a given value of $h$ (and $E$).
Figure 8 Normalized particle-wall interaction potential energies $\phi/(kT)$ as a function of particle-wall separation $h$ for combined Poiseuille flow at $\Delta p/L = 0.43$ Bar/m and EO flow at $E = 0$ V/cm (○), 2.4 V/cm (△), 4.7 V/cm (□), 7.1 V/cm (▽), and 8.8 V/cm (◇).

Figure 9 The difference between normalized particle-wall interaction potential energies for combined Poiseuille flow at $\Delta p/L = 0.43$ Bar/m and EO flow at $E = 0$ V/cm (△), 2.4 V/cm (□), 4.7 V/cm (▽), and 8.8 V/cm (◇) and that for only Poiseuille flow (i.e., at $E = 0$) at $\Delta p/L = 0.43$ Bar/m, $\Delta \phi_e/(kT)$, as a function of particle-wall separation $h$.

In theory, the magnitude of the repulsive force that drives the $a = 490$ nm particles away from the wall should be the slope of the potential profile. Numerical differentiation is, however, very sensitive to experimental uncertainty and noise. To reduce the effects of noise, one can estimate instead the magnitude of the force averaged over a range of particle edge-wall separation distances. The magnitude of the dielectrophoretic-like repulsion force observed in EOF (with negligible pressure gradient) averaged over $h_1 \leq h \leq h_2$ can be estimated as follows:
\[ F_{\text{avg}} = \frac{1}{h_2 - h_1} \int_{h_1}^{h_2} F(h) \, dh = -\frac{\phi(h_2) - \phi(h_1)}{h_2 - h_1} \tag{4} \]

Similarly, the magnitude of the repulsion force due only to the effects of the electric field in combined EO and Poiseuille flow can be estimated as follows:

\[ F_{\text{avg}}^E = \frac{1}{h_2 - h_1} \int_{h_1}^{h_2} F(h) \, dh = -\frac{\Delta \phi_E(h_2) - \Delta \phi_E(h_1)}{h_2 - h_1} \tag{5} \]

where \( h_1 = 80 \text{ nm} \) and \( h_2 = 280 \text{ nm} \) for Eqs. 4 and 5. A comparison of \( F_{\text{avg}} \) and \( F_{\text{avg}}^E \) (Table II), which is an attempt to estimate the “additional” force due to the presence of the electric field in combined EO and Poiseuille flow, clearly shows that this additional force \( F_{\text{avg}}^E \) is much greater than the dielectrophoretic-like repulsion force observed in EOF in the absence of shear. The uncertainty in these force estimates is quite large (the estimates of \( F_{\text{avg}}^E \) for the two lowest values of \( E \) are less than 0.1 fN in magnitude), about 3 fN based on the results of three independent experiments, but the differences between \( F_{\text{avg}}^E \) and \( F_{\text{avg}} \) are greater than the uncertainty for \( E \approx 4.7 \text{ V/cm} \).

**Table II** Estimates of the forces due to the presence of an electric field in “pure” EOF and in combined EO and Poiseuille flow. The uncertainty in these force estimates is about 3 fN.

<table>
<thead>
<tr>
<th>( E ) [V/cm]</th>
<th>( F_{\text{avg}} ) [fN]</th>
<th>( \Delta p/L = 0.43 \text{ Bar/m} ) [fN]</th>
<th>( \Delta p/L = 1.04 \text{ Bar/m} ) [fN]</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.4</td>
<td>0</td>
<td>2.0</td>
<td>3.5</td>
</tr>
<tr>
<td>4.7</td>
<td>0</td>
<td>5.1</td>
<td>5.4</td>
</tr>
<tr>
<td>7.1</td>
<td>1.2</td>
<td>8.3</td>
<td>8.9</td>
</tr>
<tr>
<td>8.8</td>
<td>2.1</td>
<td>9.2</td>
<td>14.2</td>
</tr>
</tbody>
</table>

These results therefore show that the combination of shear and an electric field parallel to the wall creates a strong wall-normal force that repels negatively charged dielectric particles suspended in a conducting medium with mobile electrolytes from the surface of a negatively charged wall. Moreover, initial estimates of the magnitude of this force suggest that the force is significantly greater than the superposition of the dielectrophoretic-like repulsive force observed in EO flow and the shear-induced electrokinetic lift force observed in Poiseuille flow, and that there is a nonlinear interaction between the electric field and shear.

4. SUMMARY

Combined electroosmotic and Poiseuille flow in ~30 \( \mu \text{m} \) deep fused-silica microchannels at electric fields less than 9 V/cm and pressure gradients less than 1.1 Bar/m was studied using evanescent-wave particle velocimetry with 125 nm and 245 nm radii fluorescent tracers to investigate the effects of the dielectrophoretic-like repulsion due to the presence of an electric field parallel to the wall on a shear flow. As expected, the changes in near-wall distribution of the larger particles due to repulsive forces affect the particle velocimetry results, and increase the average velocity sampled by the tracers. Nevertheless, the particle velocimetry results are in reasonable agreement with analytical predictions after accounting for these changes in the near-wall particle distribution, and show that this flow is, as expected, the superposition of a simple shear flow due to the Poiseuille flow and the uniform flow due to the electroosmotic flow.

The near-wall distributions of the larger particles obtained in electroosmotic flow at a negligible pressure gradient clearly show the effects of dielectrophoretic-like repulsion, and those obtained in Poiseuille flow at zero electric field show the (weaker) effects of shear-induced electrokinetic lift. No such effects were observed for the smaller particles, in agreement with previous studies. Very strong repulsion was observed, however, in the combination of the two flows, or an electric field with shear. Moreover, initial estimates of the magnitude of this repulsive force suggest that it is greater than the superposition of dielectrophoretic-like repulsion and shear-induced electrokinetic lift force magnitudes. Future efforts will therefore focus on clarifying this and other nonlinear effects on near-wall microparticle dynamics in combined electroosmotic and Poiseuille flow.

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