Steady flow of ionic liquid through a cylindrical microfluidic contraction–expansion pipe: Electroviscous effects and pressure drop

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** Abstract**

Electroviscous effects in steady, pressure-driven flow of a symmetric 1:1 electrolyte in a cylindrical microfluidic 4:1:4 contraction–expansion at low Reynolds number are investigated numerically by solving the field equations using a finite volume method. Predicted profiles of electrical potential, charge distribution, pressure drop and apparent viscosity are qualitatively similar to those for the slit-like contraction–expansion studied previously by the authors. However, the changes in electrical potential and pressure drop along the channel are greater than those for a corresponding slit-like geometry. The apparent viscosity is lower in the cylindrical contraction–expansion than it is in the equivalent slit-like geometry, whereas the converse is found for uniform channels except when the electrical double layers (EDLs) overlap. As for the slit-like case, a simple model is developed to calculate the pressure drop, and hence the apparent viscosity, by adding the pressure drops over the inlet, outlet and contracted sections of the channel (based on the fully developed flow in a uniform pipe), and an extra pressure drop due to contraction–expansion using the low Reynolds number analytical solution for a circular orifice. For the parameter range considered, the predictions of the simple model overestimate the apparent viscosity by up to 5–12% compared with that obtained by a finite volume numerical solution. The differences become smaller when the thickness of the EDL or the surface charge density are reduced.

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1. Introduction

Over recent decades, enormous progress has been made in the development of technologies that pertain to miniaturization of conventional complex chemical analytical procedures into extremely small microchips. These are commonly referred to as Lab-On-Chip (LOC), Micro Total Analysis Systems (μTAS) or microfluidic chips and are a subset of Micro-Electro-Mechanical Systems (MEMS). The key advantages of microfluidic devices include the significantly reduced quantity of samples and increased rate of heat/mass transfer and chemical reactions. The rapid growth of novel and complex microfluidic devices has mainly been motivated by the fast development in MEMS-related fluidic devices ranging from pH and temperature sensors to fluid actuators, such as micro-nozzles, pumps, mixers and valves (Ho and Tai, 1998), biotechnology areas such as the analysis of DNA and proteins, and biodefence (Whitesides and Stroock, 2001), as well as LOC systems for drug delivery, chemical analysis and biomedical diagnosis (Bousse et al., 2000).

The above-mentioned applications require the ability to transport, manipulate and process fluids (in general aqueous-based solutions) through micro-channels. The microfluidic flows can be driven by pressure or electrical potential difference. For the optimal development of new microfluidic systems, it is important to understand the microchannel flow characteristics, such as pressure distribution, heat transfer and transport behaviour of the flow. However, the relative importance of the forces that can influence fluid flow is different at the micrometer length scales of these devices. For example, as the size of the device is reduced, the importance of surface-based phenomena, such as surface tension and electrokinetic effects, increases (Ho and Tai, 1998; Stone and Kim, 2001; Stone et al., 2004).

Most solid surfaces bear electrostatic charges. When an ionic liquid is brought into contact with a charged surface, electrokinetic phenomena develop as the surface charges attract counter-ions in the liquid. The rearrangement and balancing of the charges gives rise to a region called the electrical double layer (EDL; Hunter, 1981; Li, 2001). The EDL is the region of variation in electric potential away from the surface. The presence of an EDL in pressure-driven flow of ionic liquids results in an extra hydrodynamic resistance (electroviscous effect). This occurs because of a flow-induced electric field resulting from the transport of excess ions along the channel by the
flow (streaming current: Li, 2001, 2004; Stone et al., 2004; Masliyah and Bhattacharjee, 2006).

Substantial information associated with the flow through microfluidic geometries is available in the literature (Hunter, 1981, 1989; Ho and Tai, 1998; Bousse et al., 2000; Li, 2004; Stone et al., 2004; Squires and Quake, 2005; Gad-El-Hak, 2006; Masliyah and Bhattacharjee, 2006). Many experimental studies (Mala et al., 1997a,b; Ren et al., 2001a,b; Li, 2001, 2004; Brutin and Tadrist, 2005) have reported that pressure-driven flows through microfluidic channels are influenced by electrokinetic effects. The electroviscous effect has been investigated for slit-like microchannels (Mala et al., 1997a,b; Chun and Kwak, 2003; Davidson and Harvie, 2007; Davidson et al., 2007), for rectangular microchannels (Yang et al., 1997; Yang et al., 1998; Ren et al., 2001a,b; Li, 2001, 2004), for cylinders (Bowen and Jenner, 1995; Bhattacharyya et al., 2003; Brutin and Tadrist, 2005), for divergent (cone shaped) (Chun et al., 2003) and for elliptical (Hsu et al., 2002, 2006) microchannels. Electroviscous effects in microfluidic channels have also been studied analytically (e.g., see Yang and Li, 1998; Bhattacharyya et al., 2003; Chun et al., 2003; Soong and Wang, 2003; Yang and Kwok, 2003a,b, and references therein).

Most of the available studies are concerned with microchannels of uniform cross-section. However, microfluidic elements commonly feature abrupt non-uniform geometries such as contraction/expansions, T-junctions or other branchings. Electroviscous effects in non-uniform slit-like 4:1:4 contraction–expansion microchannels have been studied by Davidson and Harvie (2007) for low Reynolds number flow using a finite volume method. They developed a simple theoretical model to calculate the pressure drop, and thus the apparent viscosity, by adding the pressure drop in different sections of the geometry, based on the classical solution of fully developed flow in a uniform slit, and an extra pressure drop due to contraction–expansion using a low Reynolds number analytical solution for a slit orifice. At present, there is no corresponding study of electroviscous flow through a cylindrical microfluidic contraction–expansion. The aim of present work is to extend the study of Davidson and Harvie (2007) to investigate electroviscous effects in a cylindrical microfluidic 4:1:4 contraction–expansion for the same parameter ranges considered therein.

2. Problem statement and mathematical formulation

Consider the axisymmetric, steady, fully developed pressure-driven flow (with an average inlet velocity \( \bar{V} \)) of an incompressible, Newtonian electrolyte solution through a cylindrical 4:1:4 contraction–expansion, as shown in Fig. 1. The cylindrical contraction (length, \( L_c \) and radius, \( R_c \)) is situated at a distance \( L_i \) (length of inlet pipe) from the inlet and at a distance \( L_o \) (length of outlet pipe) from the outflow boundary (Fig. 1). Thus the total length \( L = L_i + L_c + L_o \). The inlet and outlet pipes are assumed to be of equal radius \( R_i = R_o = R \) with \( R = 4R_c \) (i.e., contraction ratio, \( d = R_c / R = 0.25 \)).

Assume that the pipe wall carries a net immobile electrostatic charge of surface density \( \sigma \). Furthermore, we assume that the liquid contains symmetric anions and cations (specified by \( + \) and \( - \), respectively) with valencies \( z_+ = z_- = z \), equal diffusivities (\( \mathcal{D}_+ = \mathcal{D}_- = \mathcal{D} \)), and that the bulk ion concentration of each ionic species is \( n_0 \). The dielectric constant of the wall material is considered to be much less than that of liquid (\( \varepsilon \approx \varepsilon_0 \)).

2.1. Governing equations

Electrokinetic flows of liquid containing ionic species are described by the equation of continuity and the Navier–Stokes equations with an electrical body force term. These flow field equations are coupled to the Poisson equation relating the electrical potential to the charge distribution, and Nernst–Planck equation for conservation of each ion species (Hunter, 1989; Ghosal, 2006).
electrokinetic flow equations have been rendered dimensionless using $R$, $\overline{V}$, $R/\overline{V}$, $n_0$ and $k_b T/ze$ as scaling variables for lengths, velocities, time, ion number densities and electrical potential, respectively. The non-dimensionalization using these scaling parameters results in the following dimensionless groups:

$$Re = \frac{\rho \overline{V} R}{\mu}, \quad Sc = \frac{\mu}{\rho \overline{V}}, \quad B = \frac{k_b T R}{2 \pi \rho \overline{V} e^2}, \quad K = \frac{2 \pi n_0 R^2}{e_0 k_b T}$$

where $Re$ and $Sc$ are the Reynolds and Schmidt numbers, respectively. $K$ is the dimensionless inverse Debye length (proportional to the ratio of the pipe radius to the EDL thickness) which depends on the bulk ion concentration ($n_0$) and $B$ is a dimensionless parameter which depends on the liquid properties at a specified temperature. Here, $e$, $k_b$, $T$, $r$ and $e_0$ are the elementary charge, the Boltzmann constant, temperature, dielectric constant of the solution and the permittivity of the free space, respectively. The dimensionless governing equations with the above-noted simplifying assumptions are now presented.

According to the theory of electrostatics, for a simple symmetric $(z: z)$ electrolyte solution, the relationship between the total electrical potential ($U$) and the net charge density at any point in the liquid is described by the Poisson equation:

$$\nabla^2 U = -\frac{1}{\varepsilon} K^2 (n_+ - n_-)$$

where $n_+$ and $n_-$ are the numbers per unit volume of anions and cations, respectively.

Most electrokinetic flow models are concerned with channels of uniform cross-section and typically split the total electrical potential ($U$) into an EDL potential ($\psi$) and a streaming potential which varies linearly along the channel. In channels of uniform cross-section, the decoupling of the two potentials is possible as the streaming potential field is everywhere parallel to the wall. However, for a non-uniform channel such as a contraction–expansion, there is no simple way of splitting the potentials and so we consider the total potential.

For incompressible Newtonian fluids, the continuity equation is

$$\nabla \cdot \mathbf{V} = 0$$

where $\mathbf{V}$ is the velocity (with components $V_r$ and $V_x$ in cylindrical polar coordinates $(r, x)$).

The momentum equation for flow of incompressible Newtonian liquids incorporating electrokinetic effects is

$$\frac{\partial \mathbf{V}}{\partial t} + \nabla \cdot (\mathbf{V} \mathbf{V}) = -\nabla P + \frac{1}{Re} \nabla \cdot \left[ \nabla \mathbf{V} + (\nabla \mathbf{V})^T \right] - \frac{B K^2}{Re^2} (n_+ - n_-) \nabla U$$

where $P$ is the pressure. The last term in the right side of Eq. (4) represents an electrical force due to the free charge. The nonlinear term in the above equation is retained for completeness, even though it will be negligible for the low $Re$ flows.

The Nernst–Planck equation describing conservation of each ion species ($n_+$ and $n_-$) is

$$\frac{\partial n_{\pm}}{\partial t} + \nabla \cdot (n_{\pm} \mathbf{V}) = \frac{1}{Pe} \left[ \nabla^2 n_{\pm} \pm \nabla \cdot (n_{\pm} \nabla U) \right]$$

where $Pe = Re Sc$

2.2. Boundary conditions

Boundary conditions must be specified at the inlet, exit and walls of the contraction–expansion pipe. Conditions at the inlet and exit are chosen to be consistent with fully developed electrokinetic flow conditions and ignore any reservoir or end effects.

The boundary conditions for this flow problem are:

- **Inlet**: The velocity and ion concentration at the inlet are imposed from the solution for steady, fully developed electroviscous flow in a uniform pipe where

$$V_r = 0, \quad V_x = V_0 (r), \quad n_+ = e^{-\psi(r)}, \quad n_- = e^{\psi(r)}$$

Here $V_0$ and $\psi$ are the velocity and the EDL potential fields, respectively, for uniform pipe flow. They are obtained by solving the field equations numerically over the pipe radius using a finite-difference method (FDM). The FDM solution procedure for the fully developed electrokinetic flow in a uniform pipe is summarized in Appendix A.

- Although $\psi(r)$ is used to calculate the inlet boundary conditions for velocity and ion concentrations in Eq. (6), it is not used explicitly as an inlet condition for the potential $U$. Instead, a gradient inlet condition for $U$ is used where $\partial U/\partial x$ is calculated by imposing zero net axial current ($I_{net} = I_b + I_d + I_c = 0$) at the inlet. It is given by

$$2 \pi \int_0^1 \left[ \frac{(n_+ - n_-) V_0 (r)}{l_s} - \frac{1}{Pe} \left( \frac{\partial n_+}{\partial x} - \frac{\partial n_-}{\partial x} \right) \right] r \, dr = 0$$
All quantities in Eq. (7) are evaluated at the inlet $x = 0$. The $I_s$, $I_d$ and $I_c$ are the streaming, diffusion and conduction currents, respectively. At steady state, $I_d = 0$ at the inlet. Since $U(r, x)$ only appears as a gradient in the governing equations and the boundary conditions (potential gradients are also set at exit and wall boundaries as is discussed below), $U$ is only determined to within a constant. We therefore set $U(0, 0) = 0$ as a reference value.

- Exit: The axial potential gradient at the pipe outlet is taken to be uniform and is chosen to satisfy Gauss’s law over the flow domain (volume integral of Eq. (2) followed by application of the Divergence theorem). At steady state, the total current passing through the outlet cross-section becomes zero, i.e., Eq. (7) is satisfied. The axial gradients of the axial velocity and ion concentrations are taken to be zero, and the axial pressure gradient (taken to be uniform over the outlet cross-section) is chosen to ensure global mass conservation. Since the pressure $P(r, x)$ only appears in the governing equations as a gradient and it is not specified anywhere, we can set a reference value for it. We choose $P(0, 0) = 0$.

- On the pipe walls: The electrical potential boundary condition at the wall is based on a uniform surface charge density. It can be written as

$$\nabla U \cdot n_w = S \quad \text{where} \quad S = \frac{\text{zero}}{e_0 \varepsilon_b \varepsilon_f}$$

and $n_w$ denotes the outward unit vector normal to the pipe wall and $S$ is the dimensionless surface charge density. The other wall boundary conditions are zero flux of ions normal to the wall and the no-slip velocity condition.

- Both the EDL, and possibly the stagnant layer behind EDL, contribute to the surface conduction (Delgado et al., 2005). Usually the stagnant layer conductivity is ignored, as is the case here. As in Davidson and Harvie (2007), EDL conductivity is accounted for in this work as we are solving within the EDL region.

3. Numerical solution methodology

Owing to the axial symmetry of the flow, the solution is obtained only in the one symmetric half ($r > 0$) of the domain (Fig. 1). Eqs. (2)–(5) along with the above-noted boundary conditions are solved numerically. This yields the velocity ($V_r$ and $V_t$), pressure ($P$), electrical potential ($U$) and ion concentration ($n_+$ and $n_-$) fields. The simulations are performed using an adapted single phase version of the unsteady two-fluid finite volume method due to Rudman (1998). Steady-state solutions are obtained by using a false transient method and multi-grid Poisson solver (Rudman, 1998). A semi-implicit time stepping procedure (Li and Renardy, 2000) is used to relax the viscous time step limitation of a fully explicit scheme at low Reynolds number. This allows larger time steps and thus increases the rate of approach to steady state. To reduce the computational time, the time steps are controlled in two ways. First, the time steps are limited to 10 times the explicit viscous time step to ensure that the spatial discretization errors resulting from the semi-implicit formulation are minimized. Secondly, after obtaining the fully converged semi-implicit solution, additional time steps limited to the explicit viscous time step are performed to achieve higher accuracy. The time step is also subjected to modified Courant and other conditions to ensure numerical stability, as detailed by Davidson and Harvie (2007).

The calculations are taken to be converged to a steady state when the net current (normalized with respect to the streaming current, $I_{\text{net}}/I_s$) at the exit of the pipe is less than $10^{-8}$. The numerical calculations have been performed on a uniform ($\Delta x = \Delta r = \frac{1}{32}$) staggered grid. Grid resolution test calculations with a refined grid ($\Delta x = \Delta r = \frac{1}{64}$) yielded differences of less than $2\%$ in the prediction of overall pressure drop, and almost coincident predictions for each of the electrical potential, charge and velocity profiles.

4. Results and discussions

Calculations for a cylindrical contraction—expansion are performed with the same parameter ranges considered by Davidson and Harvie (2007) for a slit-like contraction. The parameters are based on a symmetric 1:1 aqueous electrolyte solution at a temperature of 298 K ($B = 2.34 \times 10^{-4}$ and $S_c = 10^3$) for two values of the contraction length ($L_c/R = 1, 5$). The lengths of the inlet and outlet sections are taken to be $L_i = L_o = 5R$. These lengths ($L_i$ and $L_o$) were found to be sufficient to ensure that the flow near the entry and exit to the channel is unaffected by the flow in the contraction, and that the flow at the outlet is fully developed. Results for the pressure drop and electroviscous effects are presented for Reynolds number $Re = 0.01$, at five values of the scaled inverse Debye length ($K = 2, 4, 6, 8, 20$) and three values of dimensionless surface charge density ($S = 4, 8, 16$). The EDLs are overlapping when $K = 2$ and are very thin at $K = 20$. Only positive values of $S$ are considered since the results for negative $S$ can be obtained by setting new values of $U = -U$ and $n_+ = n_-$, respectively. The studied parameter range corresponds to zeta potentials approximately in the range 50–100 and 12–50 mV when $K = 2$ and 8, respectively, for a uniform microchannel.

4.1. Total electrical potential ($U$) profiles

Contours of the dimensionless total electrical potential ($U$) are shown in Fig. 2 for dimensionless surface charge density of $S = 8$ and contraction length of $L_c/R = 5$. Fig. 3 shows the variation of the total electrical potential at the centreline along the length of the microfluidic pipe and compares it with the variation on the centreline of a corresponding slit-like contraction–expansion microchannel (Davidson and Harvie, 2007). For a fixed value of the scaled inverse Debye length ($K$), the total electrical potential decreases in the direction of flow because of the advection of negative ions (for $S = 0$) along the length of the pipe. The lateral curving of the contours in Fig. 2 occurs due to the nonzero potential gradient normal to the wall (Eq. (8)). The magnitude of the electrical potential gradient increases with the decreasing value of $K$. This increase is due to the thickening of the EDL, which occupies the greatest cross-section of the pipe at the smallest value of $K$. This exposes more counter-ions to the flow, requiring a larger potential gradient to give zero net current. For the same reason, the potential gradient is much greater in the contraction section where the pipe radius is smallest. While the qualitative behaviour of total electrical potential profiles (Figs. 2 and 3) is similar to that for slit-like microfluidic contraction–expansions (Davidson and Harvie, 2007; Davidson et al., 2007), quantitatively these predictions are different. For instance, the total electrical potential drop on the centreline varies from 350 to 763 as $K$ is decreased from 8 to 2 for $S = 8$ and $L_c/R = 5$ in the cylindrical case, whereas the corresponding values for a slit-like geometry vary from 66 to 251, under otherwise identical conditions. This occurs because, for a depth of the slit chosen so that the cross-sectional areas of the slit and cylinder are equal, the charged wall surface area for the slit is less than that for the cylinder. In that case the surface charge, and hence the charge in the EDL, is of greater magnitude in the cylinder. Consequently, the streaming current, and the induced potential gradient, is greater in the cylinder.

4.2. Dimensionless excess electrical charge ($n^*$) distribution

The difference between the counter-ion and co-ion number concentration is referred to as the excess ionic number concentration ($n^* = n_+ - n_-$). For a symmetric electrolyte, the dimensionless net
Charge density equals $n^*$. Dimensionless charge density ($n^*$) distribution profiles through the microfluidic cylindrical pipe are shown in Fig. 4 for a dimensionless surface charge density of $S = 8$ and contraction length of $L_c/R = 5$. The charge is negative, reflecting the dominance of the negative ion concentration ($n^-$) along the length of pipe.

For fixed value of $K$, the contours of the dimensionless charge density show the clustering of negative values near the pipe wall because of the positive surface charge density at the wall. Fig. 4 shows that the magnitude of the dimensionless charge density is smaller at high values of $K$. It increases with decreasing values of $K$. For instance, the minimum value of $n^*$ changes from $-6.26$ to $-83.03$ as the $K$ decreased from 8 to 2 at $S = 8$. At high values of $K$, the contours of $n^*$ cluster very close to the pipe wall, which is consistent with a very thin EDL. The region of negative charge, induced by the positive surface charge, expands towards the centre of the pipe from the wall as $K$ decreases. The charge contours from the inlet region penetrate into the contraction region because of the converging flow field.

Fig. 5 shows the distribution of the dimensionless charge on the centreline along the length of the microfluidic cylindrical pipe for three values of dimensionless surface charge density ($S = 4, 8, 16$) and five values of inverse Debye length ($K = 2, 4, 6, 8, 20$). All the centreline curves are coincident at near zero charge in the inlet and outlet sections except for $K = 2$. For $K = 2$ the EDLs overlap, and the centreline charge is negative for all surface charge densities ($S$) considered. Also, the dimensionless charge in the inlet section of the pipe is almost uniform regardless of the Debye parameter ($K$), except in the region very close to the contraction. In the contraction region, the charge is more negative than it is in the entry or exit sections off the channel. This is because EDL occupies more of the cross-section in the contraction.

Qualitatively, the dimensionless charge distributions for cylinder-like (Figs. 4 and 5) and slit-like (see Figs. 2 and 4 in Davidson and Harvie, 2007) microfluidic contractions are similar. In each case, both decreasing $K$ (thickening of EDL) and an increasing $S > 0$ enhances the negative charge in a similar manner. However, the charge is of greater magnitude in the cylinder because the wall surface area is greater in that case compared with a corresponding slit of finite depth, as discussed in the previous section in relation to the total potential gradient.
Fig. 3. Variation of the dimensionless total electrical potential ($U$) at the centreline along the length of the contraction-expansion channels.

Fig. 4. Dimensionless charge ($n^*$) distribution at $S = 8$ for $L_c/R = 5$ and for different values of $K$. 

Re = 0.01, Sc = 1000, $B = 2.34 \times 10^{-4}$, $S = 8$
The length of pipe.

4.3. Variation of pressure drop

Fig. 5. Dimensionless charge \((n^* - n_+ - n_-)\) profile on the centre line \((r = 0)\) along the length of pipe.

4.4. Apparent viscosity

In pressure-driven electrokinetic flow, the streaming potential field produces an additional hydrodynamic resistance represented by the additional body force in the momentum equation \((\text{i.e.}, \text{electrical force in Eq. (4)}). For a fixed volumetric flow rate \((Q)\), this additional resistance manifests as a pressure drop \((\Delta P)\) along the pipe that is higher than the pressure drop in absence of the electrical forces \((\Delta P_0)\). This electroviscous effect is often quantified in terms of an apparent (or effective) viscosity. The apparent viscosity \((\mu_{\text{eff}})\) is the viscosity of the fluid (without electrical forcing) required to achieve the pressure drop \((\Delta P)\). For low \(Re\) steady flow, the nonlinear convection term in the momentum equation is small, and the apparent viscosity \((\text{represented in terms of the electroviscous correction factor} Y = \mu_{\text{eff}}/\mu)\) is related to the pressure drop as

\[
Y = \frac{\mu_{\text{eff}}}{\mu} = \frac{\Delta P}{\Delta P_0} \tag{9}
\]

where \(\mu\) is the physical viscosity of the liquid. For the special case of a uniform channel, \(Y\) can also be expressed as

\[
Y = 1 + \frac{\sigma E^*}{\mu (-\nabla V / \nabla r)^{\ast \text{w}}} = 1 + \frac{2 R S E}{R e (\nabla V / \nabla r)^{\ast \text{w}}} \tag{10}
\]

where * denotes a dimensional variable. Eq. (10) shows that the apparent viscosity ratio exceeds 1 by the ratio of electrical to viscous stresses at the channel wall. The equation can be derived by integrating the axial momentum equation over the volume of the channel.

Fig. 8 shows the variation of the electroviscous correction factor \((Y, \text{Eq. (9)})\) with the parameters \(K\) and \(S\), evaluated from the numerical finite volume simulations. As expected, the value of \(Y\) exceeds 1 over the range of conditions. It decreases with both an increasing \(K\) (i.e., decreasing EDL thickness) and a decreasing \(S\). The value \(Y = 1\) represents liquid flow without electrokinetic effects. Qualitatively similar trends are reported in the literature for a slit-like channels \((\text{Chun and Kwak, 2003; Davidson and Harvie, 2007).}\)

As discussed above, the local flow characteristics can be obtained numerically, however, for easy use in process design calculations, it would be more convenient to predict them via a simpler model, as was done by \(\text{Davidson and Harvie (2007)}\) for a slit-like microfluidic contraction–expansion. A corresponding theoretical analysis for pressure drop in cylindrical microfluidic contraction–expansion is presented below.

\(\text{Sisavath et al. (2002)}\) show that the pressure drop in non-electroviscous creeping flow through a contraction–expansion can be reasonably well predicted by adding the pressure losses due to fully developed flow in the pipe segments and an extra pressure drop due to the contraction–expansion (excess pressure drop) as follows:

\[
\Delta P_{0,m} = \Delta P_{r,0} + \Delta P_{c,0} + \Delta P_{0,0} + \Delta P_{e,0} \tag{11}
\]
where $\Delta P_{l,0}$, $\Delta P_{c,0}$ and $\Delta P_{o,0}$ are the pressure drops in a uniform pipe of length $L_i$ and radius $R$ (inlet section), in a pipe of length $L_c$ and radius $R_c (= R/4)$ (contraction section), and in a uniform pipe of length $L_o$ and radius $R$ (outlet section), respectively. The $\Delta P_{c,0}$ is the excess pressure drop due to the contraction–expansion.

In this work, $\Delta P_{l,0}$ and $\Delta P_{o,0}$ for non-electrokinetic flow are calculated from the dimensionless pressure drop in fully developed creeping flow through a uniform uncharged pipe (of radius $R$) as

$$\Delta P_{l,0} = \frac{8}{Re} \left( \frac{L_i}{R} \right)$$

and

$$\Delta P_{o,0} = \frac{8}{Re} \left( \frac{L_o}{R} \right)$$

The $\Delta P_{c,0}$ is obtained by rescaling of Eq. (12) in terms of the contraction parameters to give

$$\Delta P_{c,0} = \frac{8}{d^4Re} \left( \frac{L_c}{R} \right)$$

By using the Sampson analytical solution (Happel and Brenner, 1965) for creeping non-electrokinetic flow through a circular orifice (of zero thickness) in an infinite wall, Sisavath et al. (2002) obtained an approximate expression for the excess pressure drop $\Delta P_{c,0}$ due to axisymmetric sudden contraction–expansion with finite expansion.
are shown. Through contraction-expansion microchannels. Results for two contraction lengths of the contraction-expansion pipe is given by

\[
\Delta P_{e,0} = \frac{3\pi}{4d^2Re} (1 - d^2)
\]  

(14)

where \((1 - d^2)\) is an area correction factor. Thus, the total pressure drop \(\Delta P_{0,m}\) in non-electroviscous flow through contraction-expansion pipe is given by

\[
\Delta P_{0,m} = \frac{8}{Re} \left[ \frac{1}{R} \left( L_i + \frac{L_e}{d^4} + L_o \right) + \frac{3\pi}{8d^3} (1 - d^2) \right]
\]  

(15)

Similar to Eq. (11), the pressure drop in electroviscous flow can be calculated as

\[
\Delta P_m = \Delta P_c + \Delta P_e + \Delta P_0 + \Delta P_{e,0}
\]  

(16)

Apart from approximate solutions valid for small EDL potential, analytical solutions for fully developed electroviscous flow in a uniform pipe are not available to determine the pressure drop terms in Eq. (16); therefore, a numerical solution [see Appendix A] has been used. The excess pressure drop \(\Delta P_e\) in electrokinetic flow is approximated by Eq. (14), i.e., \(\Delta P_e = \Delta P_{e,0}\). Note that Eq. (14) does not account for the electrokinetic effects.

Fig. 8 also shows the numerical values of the correction factor with that estimated using the simple model discussed above. The simple model overestimates the correction factor, and hence the apparent viscosity, by a maximum of 11.42% and 5.30% for \(L_e/R=1\) and 5, respectively, over the ranges of conditions. For fixed value of surface charge density (S), the difference in the two values diminishes with an increasing \(K\) (i.e., thinning of EDL). Similarly, for fixed value of \(K\), the difference becomes smaller with the decreasing values of \(S\). Fig. 8 shows that the two predictions are almost same at the highest value of \(K\) where there are weak EDL effects, irrespective of the value of the surface charge density (S). As noted earlier the excess pressure drop (\(\Delta P_e\), Eq. (14)) in the simple model does not include electroviscous effects and this causes the overestimate of \(Y\). In particular, Eq. (14) ignores the effect of surface charge on the front and backward facing steps of the contraction-expansion. At the front facing step of the contraction \((x=L_i)\), the flow is directed towards the centreline \((r=0)\) and a local transverse potential gradient \((\partial U/\partial r > 0)\) is promoted for \(S > 0\). This is reinforced by the radial potential gradient at the wall \((r=R)\) of the inlet pipe. The combined transverse potential gradient produces a force that reduces the inward radial flow parallel to the front facing step. Consequently, the wall friction on the front step is lower than that which occurs when surface charge on the step is ignored. The corresponding effect at the backward facing step of the pipe expansion \((x=L_i+L_e)\) is small.

Fig. 8 also compares the electroviscous correction factor \((Y = \mu_{eff}/\mu)\) for corresponding cylindrical and slit-like contraction-expansion geometries. The variations in each case are qualitatively similar but \(Y\) is larger in the slit-like compared to the cylindrical microchannel over the range of conditions. The difference increases for decreasing \(K\) and increasing \(S\). The weaker electroviscous effect in the cylindrical geometry is unexpected since the surface area, and hence the total wall charge, is greater in a cylinder than it is in a slit with a depth chosen to give the same cross-sectional area. The explanation can be found by comparing the \(Y\) variation for uniform cylindrical and slit-like channels in Fig. 9. This figure shows that the apparent viscosity is indeed larger for a uniform cylindrical channel provided \(K\) is not too small. In contrast, when \(K\) is less than 3 and 2 (i.e., the EDLs overlap) for \(S = 16\) and 4, respectively, the apparent viscosity is lower for the cylinder. (This was verified by also evaluating the apparent viscosity in each case using Eq. (10).) Low \(K\) values such as these are achieved by the effective \(K\) in the contracted part of the contraction-expansion, which is a factor of 4 smaller than those in Fig. 8 because of the 1:4 contraction ratio. Since the contracted section dominates the flow resistance, this explains the lower electroviscous resistance in the cylindrical contraction-expansion, when \(K = 2\) in Fig. 8, compared with that in the corresponding slit-like geometry. Note that when \(K = 20\) the effective value in the
contraction is 5 and the apparent viscosity is greater in the cylindrical contraction–expansion (Fig. 8), consistent with the behaviour in a uniform channel with \( K = 5 \).

Fig. 8 shows that for \( S = 4 \) and 16, the electroviscous correction factor \( Y \) is smaller for contraction length \( L_c/R > 1 \) than it is for \( L_c/R = 5 \). This occurs because the electrical force resisting the flow reduces as the contraction length decreases. Note that \( L_c/R = 0 \) does not give a unique channel, but a channel with an orifice plate, and we expect \( Y \) to be even smaller in that case. In contrast, Figs. 8 and 9 show that \( Y \) values for a uniform channel (no obstruction) are greater than their counterparts for a pipe with a contraction. This occurs for the same reason that the simple pressure-drop model overestimates \( Y \); viz., neither circumstance includes the electroviscous effect of the front step of a contraction (or orifice) that acts to reduce \( Y \). However, it seems likely that if \( Y \) for the contraction flow continues to increase with increasing contraction length \( L_c/R > 5 \), then it may exceed the corresponding \( Y \) value for a uniform channel.

When the contraction ratio \( d \to 1 \) (i.e. the contraction vanishes radially) the solution approaches that of a uniform channel, but we do not explore this limit in the paper (we consider only \( d = 0.25 \) and 1). As \( d \) increases, the reducing length of the front step of the contraction will tend to increase \( Y \) while the increasing diameter of the contraction will promote a reduced \( Y \). Figs. 8 and 9 show that the net effect of increasing \( d \) from 0.25 to 1 is an increase in \( Y \) for the cases considered here.

Fig. 9 predicts that the apparent viscosity decreases monotonically towards the physical viscosity as \( K \) increases. This contrasts with predictions for a uniform duct having constant wall EDL potential that show a local maximum occurs as \( K \) varies (Bowen and Jenner, 1995; Masliyah and Bhattacharjee, 2006). The occurrence of the maximum was explained by Li (2001). Additional calculations for lower values of \( K \) verify the absence of a local maximum in the present work. The different behaviour occurs because we assume constant charge density at walls, rather than constant EDL potential there. The explanation can be seen by inspecting Eq. (A.7) in Davidson and Harvie (2007) (re-labelled Eq. (17) here) that relates the dimensionless surface charge density \( S \) to the EDL potential \( \psi_s \) at the wall for a uniform slit-like channel:

\[
2^{1/2}K(cosh\psi_s - cosh\psi_0)^{1/2} = |S|
\]

where \( \psi_s \) is the EDL potential at the centreline.

Eq. (17) shows that the surface charge density (and hence the electroviscous effect) tends to zero as \( K \) approaches zero for fixed wall potential \( \psi_s \). For example, if \( K \) is reduced by decreasing the bulk ion concentration \( n_0 \), the charge in the liquid decreases and so does the surface charge density. Since the electroviscous effect is also zero for large values of \( K \), there must be a maximum at some intermediate \( K \) value. However, the behaviour is different if \( S \) is held fixed and \( \psi_s \) is allowed to vary (the case in this paper) as \( K \) is reduced to zero. Now Eq. (17) shows that \( \psi_s \) (and indeed \( \psi \) throughout the EDL) becomes increasingly large as \( K \) decreases to zero, since \( S \) is now fixed. For the uniform slit-like channel, one can show (by approximating, for small \( K \), the solution in the Appendix of Davidson and Harvie, 2007) that the apparent viscosity remains greater than the physical value as \( K \) approaches zero for fixed \( S \), consistent with Fig. 9.

5. Conclusions

Electroviscous effects in axisymmetric, steady, pressure-driven flow of a symmetric 1:1 electrolyte in a cylindrical microfluidic 4:1:4 contraction–expansion pipe at low Reynolds number (\( Re = 0.01 \)) are investigated numerically. The Poisson equation for the electrical potential, the Nernst–Planck equations for the ion concentrations and the Navier–Stokes equations with electrical forcing are solved using a finite volume method. The pipe walls are assumed to have a uniform surface charge density. The fully developed electroviscous flow solution in a uniform pipe is imposed as an inlet condition for the contraction–expansion geometry. Numerical results for the total electrical potential, ion concentration, charge distribution, pressure drop and apparent viscosity are presented and discussed over the following parameter ranges: scaled Debye length, \( 2 < K < 20 \) and dimensionless surface charge density, \( 4 < S < 16 \) at Schmidt number \( Sc = 1000 \) and a liquid specified parameter \( B = 2.34 \times 10^{-4} \).

The predictions are qualitatively similar to those for the slit-like contraction–expansion studied by Davidson and Harvie (2007). Quantitatively, the total electrical potential drop, dimensionless charge and pressure drop along the centreline are larger than for a corresponding slit-like contraction. As for the slit-like geometry, the apparent viscosity increases with decreasing values of \( K \) and increasing values of \( S \), as expected. However, the apparent viscosity is lower than for a corresponding slit-like expansion–contraction. The converse is true for uniform channels, except when \( K \) is small and the EDLs overlap. The reason for the contrasting behaviour in the expansion–contraction is that the contracted section, which dominates the flow resistance, has an effective \( K \) value that is small (because of the contraction ratio). In that case, the apparent viscosity in the cylindrical contraction–expansion is less than for the corresponding slit geometry, consistent with the behaviour in a uniform channel with small \( K \).

Following the approach of Davidson and Harvie (2007) for a slit-like contraction–expansion, a simple model is developed to calculate the pressure drop along the channel by adding the pressure losses in the inlet, contraction and outlet sections (based on a numerical solution for fully developed flow in a uniform pipe) to an extra pressure drop due to the contraction–expansion (based on a low \( Re \) analytical solution for circular orifice). For the parameter range considered, this simple model overestimates the apparent viscosity in the cylindrical expansion–contraction by up to 5–12% compared to that obtained by the finite volume solution. However, the differences are smaller when the surface charge density or EDL thickness is small, or when the overall pressure drop is very large.

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Appendix A. Finite difference solution of steady, fully developed electrokinetic flow through a uniform pipe

The steady-state fully developed solution for uniform pipe flow is given in dimensionless form with the total electrical potential expressed as \( U = \psi - Ex \), where \( E \) is the constant induced electric field along the uniform pipe, and \( \psi \) (the EDL potential) is related to the ion concentration in a symmetric electrolyte solution

\[
n_+ = e^{-\psi} \quad \text{and} \quad n_- = e^{\psi}
\]

\[\text{(A.1)}\]

A.1. EDL potential field in a cylindrical microchannel

\[
\frac{1}{r} \frac{d}{dr} \left( r \frac{d\psi(r)}{dr} \right) = K^2 \sinh\psi(r)
\]

subject to the following boundary conditions:

\[
\frac{d\psi}{dr} \bigg|_{r=0} = 0 \quad \text{and} \quad \frac{d\psi}{dr} \bigg|_{r=1} = S
\]

\[\text{(A.3)}\]
where $S$ is the dimensionless surface charge density, given by Eq. (8).

The iterative finite difference solution of Eqs. (A.2) and (A.3) yields the EDL potential, $\psi(r)$. The iterative finite difference solution of Eqs. (A.8), (A.10), (A.12) and (A.13) yields the electric field strength ($E$), fully developed velocity profile ($V_0(r)$), ion concentrations ($n_\pm$) and pressure gradient ($dP/dr$) for the flow through uniform pipe. These profiles ($V_0, n_\pm$) are used as the inlet boundary condition (Eq. (6)).

A.2. Flow field coupled with electrokinetic interaction

The equation of motion for steady, fully developed flow of incompressible fluids through a cylindrical pipe at low Reynolds number, in dimensionless form, is given by

$$0 = - \left( \frac{dP}{dx} \right) + \frac{1}{Re} \int_0^1 \left( r \frac{dV_x}{dr} \right) dr + F_x$$  \hspace{1cm} (A.4)

where $dP/dx$ is the applied pressure gradient which is constant and uniform, and the body force per unit volume ($F_x$) caused by the influence of an axial electric field strength ($E$) on the net charge density in the liquid is given by

$$F_x = \frac{B E_k^2}{Re^2} (n_+ - n_-)E$$ \hspace{1cm} (A.5)

By substituting Eqs. (A.1) and (A.5) into Eq. (A.4), we get

$$0 = -A_1 \int_0^1 r \frac{dV_x}{dr} dr + \left( \frac{2BE_k^2}{Re} \right) \sinh \psi(r)$$  \hspace{1cm} (A.6)

where the constant

$$A_1 = \frac{Re}{\int_0^1 \frac{dV_x}{dx}}$$ \hspace{1cm} (A.7)

Let $V_x^* = V_x/A_1$ and $E^* = E/A_1$. Eq. (A.6) can then be written as

$$0 = -1 + \int_0^1 r \frac{dV_x^*}{dr} dr - \left( \frac{2BE_k^2}{Re} \right) \sinh \psi(r)$$ \hspace{1cm} (A.8)

In dimensionless form, the average velocity equals unity so that

$$2 \int_0^1 r V_x dr = 1$$ \hspace{1cm} (A.9)

and hence

$$A_1 = \left( 2 \int_0^1 r V_x^* dr \right)^{-1}$$ \hspace{1cm} (A.10)

Now, the electrical field strength ($E$) appearing in the above equations is calculated from the zero net axial current condition, given by Eq. (7). Under steady, fully developed flow condition and the diffusion current $I_d = 0$ (Eq. (7)), and $E$ is derived as follows:

$$2\pi \int_0^1 \left( (n_+ - n_-) V_x + \frac{1}{Pe} (n_+ + n_-) E \right) r dr = 0$$ \hspace{1cm} (A.11)

After substituting Eq. (A.1) in the above equation, the electric field strength ($E$) is given by the following expression:

$$E = Pe \left( \frac{1}{r} \int_0^1 V_x \sinh \psi(r) r dr \right)$$ \hspace{1cm} or

$$E^* = Pe \left( \frac{1}{r} \int_0^1 V_x^* \sinh \psi(r) r dr \right)$$ \hspace{1cm} (A.12)

Velocity boundary conditions are

$$\frac{dV_x}{dr} |_{r=0} = 0 \text{ and } V_x |_{r=1} = 0$$ \hspace{1cm} (A.13)

References


