Mixing in Microchannels Using Induced Charge Electroosmosis

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1 Introduction

In the design of microfluidic lab-on-a-chip systems it’s often needed to mix the contents of a channel, and for fast analysis it is important to have effective mixing. Since it is almost impossible to mix by turbulent flow in microfluidic systems, most of the mixing will be by diffusion, which is a rather slow process. Therefore it’s proposed to use a combination of electroosmotic flow and induced-charge electroosmosis (ICEO) to generate a flow with vortices for enhanced mixing. This is done by inserting metal objects in the channel, so an applied electric field in the longitudinal direction of the channel will generate an ICEO flow and an electroosmotic flow.

The electroosmotic flow is a phenomenon related to the surface chemistry between fluid and solid. By rather complicated chemistry there will build up a net charge on the wall which will attract and repel the different ions in the fluid. This will result in a fluid with a nonzero space charge density that can be affected by an electric field. It is therefore possible to sustain a flow in a microfluidic channel just by applying an electric field in the longitudinal direction. Much related is the ICEO which isn’t based on chemistry related surface charge but originate from the induced charge on a metal object placed in an electric field.

In this report we will treat this problem numerically, and attep to reproduce some of the results of the article “Efficient Micromixing Using Induced-Charge Electroosmosis”, [1] and compare them. We will study the effect of having different numbers of semicylinders, $n$, as well as the effect of the spacing between - and the mutual displacement of the metal cylinders. To evaluate the system we use the parameters $\eta_{mix}$ and $\eta'_{mix}$ defined as in the article.

$$\eta_{mix} = \left(1 - \frac{\sqrt{\frac{1}{N} \sum_{I=1}^{N} (\tilde{c}_s - \tilde{c}^*_s)^2}}{\tilde{c}_s - \tilde{c}^0_s}\right) \cdot \frac{L}{L_{mix}}$$

$$\eta'_{mix} = \left(1 - \frac{\sqrt{\frac{1}{N} \sum_{I=1}^{N} (\tilde{c}_s - \tilde{c}^*_s)^2}}{\tilde{c}^*_s - \tilde{c}^0_s}\right) \cdot \frac{L}{L_{mix}}$$

(1)

where $\tilde{c}_s$ is the scaled concentration in the local point while $\tilde{c}^*_s$ and $\tilde{c}^0_s$ are the scaled concentrations if the solution was mixed perfectly and unmixed, respectively. $N$ is the number of points used to estimate the mixing index, $L$ is the length of the channel and $L_{mix}$ is the length where $\eta_{mix} > 0.99$ is obtained. If $\eta_{mix} > 0.99$ isn’t obtained at the outlet of the channel $L_{mix} = L$.

As this expression will not take the flowrate into account, this is not the best way to evaluate how effective the mixing performance is, since obstructing the flow will result in greater mixing by diffusion and larger mixing index, even though it isn’t more efficient.

We will still evaluate our system by this mixing index so we are able to compare our results with the article [1].
2 Theory

This section will treat the fundamental theory for the phenomena which is the subject of the remainder of the report.

In macroscopic fluid systems the extent of these areas will usually be negligible compared to the dimensions of the systems, and thus electrohydrodynamic effects will seldom be significant. In microfluidic systems however the charges in these areas can be significant, and an electrohydrodynamic description is therefore often useful when dealing with microfluidic systems.

The fundamental equation of fluid dynamics is the Navier-Stokes equation which relates the pressure, \( p \), the velocity, \( u \) and the body forces, \( \mathbf{F}_{\text{body}} \), which is a collective name for all external forces effective throughout the bulk of the fluid. The gravitational force, \( \rho g \), is a familiar example of a body force and the electrical force, \( \rho_{el} \mathbf{E} \), is another such force. As we are dealing with horizontal systems in this report the gravitational force will be balanced by a normal force, and we will therefore omit it from our version of the Navier-Stokes equation. The Navier-Stokes equation thus read

\[
\rho \left( \partial_t \mathbf{u} + (\mathbf{u} \cdot \nabla) \mathbf{u} \right) = -\nabla p + \eta \nabla^2 \mathbf{u} + \rho_{el} \mathbf{E} \quad \text{Navier-Stokes equation (2)}
\]

where \( \rho \) is the density, \( \eta \) is the viscosity, \( \rho_{el} \) is the charge density and \( \mathbf{E} \) is the electric field. In this report we will only be dealing with incompressible Newtonian fluids, so the simple continuity equation stating that the velocity field is divergence free applies

\[
\nabla \cdot \mathbf{u} = 0 \quad \text{The continuity equation (3)}
\]

As we will only be dealing with electrostatic effects in this report it is sufficient to include Ohm’s law and two of Maxwell’s equations, namely Gauss’s law and the Maxwell-Faraday equation, in order to make an electromagnetic description of the system. The Maxwell-Faraday equation is in fact only used to show that the electric field is rotation free, which ensures that there exists a potential for the electric field. As we are assuming that the fluids we are working with are all linear media, the possibility of a potential formulation leads from Gauss’s law to the Poisson equation. The basic electromagnetic equations we shall be applying are therefore

\[
\begin{align*}
\nabla \cdot \mathbf{D} &= \rho_{el} & \text{Gauss’s law (4)} \\
\mathbf{J}_{el} &= \sigma_{el} \mathbf{E} & \text{Ohm’s law (5)} \\
\nabla^2 \phi(r) &= -\frac{1}{\epsilon} \rho_{el}(r) & \text{The Poisson equation (6)}
\end{align*}
\]

Any charge density that might be present in a fluid will always be due to soluted ions. The distribution of these ions will be governed by three factors namely the diffusion, the convection and the electrical force affecting them. The equation describing these effects is most easily arrived at by considering the ion current density, \( \mathbf{J} \), at a given point. As we are dealing with both cations and anions we will adopt the subscript \( \pm \) to allow
for convenient description of both ion species. $+$ will denote properties relating to the
cations and $-$ denotes properties relating to the anions. In the following $J$ denotes a
particle current density, $c$ is the particle concentration, $Z$ is the valence of the particle
and $e$ is the elementary charge.
The current density due to diffusion is given by Fick’s law as $J_{\text{diff}, \pm} = -D_{\pm} \nabla c_{\pm}$. The
convection will obviously sweep the ions soluted in a given fluid particle along and so
it will give rise to a convection current density of $J_{\text{conv}, \pm} = c_{\pm} u$. Lastly we have the
current density due to the electric field. If we divide the electrical current density given
by Ohm’s law by the charge of an ion, $Z_{\pm}e$, we will get the particle current density. By
utilizing the relationship between electrical conductivity and mobility, $\sigma_{\pm} = c_{\pm} Z_{\pm} e \mu_{\pm}$,
together with the Einstein relation, $\mu_{\pm} = \frac{Z_{\pm} e}{k_B T} D_{\pm}$, we arrive at an expression for the
ionic current density: $J_{E, \pm} = D_{\pm} \frac{c_{\pm} Z_{\pm} e}{k_B T} E$. The sum of these current density terms is
known as the Nernst-Planck equation and reads

$$J_{\pm} = -D_{\pm} \nabla c_{\pm} + c_{\pm} v - D_{\pm} \frac{c_{\pm} Z_{\pm} e}{k_B T} \nabla \phi$$  \hspace{1cm} \text{Nernst-Planck equation (7)}

where we have used the potential formulation of the electric field. To obtain the sought
equation governing the particle distribution we invoke particle conservation. Remem-
bering that $\nabla \cdot u = 0$ we get

$$\partial_t c_{\pm} + \nabla \cdot J_{\pm} = 0 \Rightarrow \partial_t c_{\pm} - D_{\pm} \nabla^2 c_{\pm} + v \nabla c_{\pm} - D_{\pm} \frac{c_{\pm} Z_{\pm} e}{k_B T} \nabla^2 \phi = 0$$  \hspace{1cm} \text{(8)}

These are the fundamental equations applied in this report.

2.1 The Debye layer

We will now look at an electrohydrodynamic phenomenon of particular interest which
is known as the Debye layer. The Debye layer is a layer of typically $1 - 100$ nm with
a nonzero charge density that forms where a fluid with soluted ions meets a charged
surface. However insignificant this layer might seem it will be shown that a number of
interesting properties can be attributed to it.
From electrochemistry we know that different elements have different electron affini-
ties, which gives rise to an electron exchange between an electrolytic fluid and a channel
wall of appropriate composition. Such an exchange will obviously result in a shifting of
the charge in the system, so that while global charge neutrality is conserved the walls
in the system will have a net charge and the fluid will be oppositely charged. From
basic electromagnetism we know that a body of positive charge will attract negatively
charged particles and repel positively charged ones. It is therefore no surprise that a
charge separation in an electrolytic fluid will occur. More to the point there will be a
build up of counterions and a reduction in coions near the walls in the system. In this
report coions will refer to ions of the same charge as the walls and counterions to ions
of opposite charge.
In order to describe the Debye layer quantitatively we will use the concept of the chemical potential, $\mu_\pm$, which for this system is given by

$$\mu_\pm(r) = \mu_0 + k_BT \ln \left( \frac{c_\pm(r)}{c_0} \right) + Z_\pm e\phi(r)$$  \hspace{1cm} (9)$$

where $\mu_0$ and $c_0$ is the chemical potential and ionic density which would be observed without the electrical potential originating from the charged wall. The chemical potential is defined as the energy of the last added particle for fixed entropy. Thus if the chemical potential in the system varied with position, there could be gained energy by rearranging the particles. In equilibrium the chemical potential must therefore be constant throughout the system, which is to say that the gradient of the chemical potential vanishes everywhere. This yields

$$\nabla \mu_\pm(r) = 0 \Rightarrow k_BT \nabla \ln \left( \frac{c_\pm(r)}{c_0} \right) = -Z_\pm e\phi(r)$$  \hspace{1cm} (10)$$

$$\nabla c_\pm(r) = -c_\pm(r) \frac{Z_\pm e}{k_BT} \nabla \phi(r) \Leftrightarrow c_\pm(r) = c_0 \exp \left( -\frac{Z_\pm e}{k_BT} \phi(r) \right)$$  \hspace{1cm} (11)$$

where in obtaining the final result we have made the assumption that the potential is zero at infinity which implies that the concentration is $c_0$ at infinity. To simplify the treatment we will now assume that the valence of the cations and anions is equal in magnitude. The charge density is easily gained from the concentrations as

$$\rho_{el}(r) = Z e(c_+(r) - c_-(r)) = -2Ze c_0 \sinh \left( \frac{Ze}{k_BT}\phi(r) \right)$$  \hspace{1cm} (12)$$

Inserting this in the Poisson equation leads to the Poisson-Boltzmann equation

$$\nabla^2 \phi(r) = 2\frac{Ze c_0}{\epsilon} \sinh \left( \frac{Ze}{k_BT}\phi(r) \right)$$ \hspace{1cm} Poisson-Boltzmann equation (13)$$

This equation does obviously not lend itself to an easy solution. Indeed there exists only one exact solution known as the Gouy-Chapman solution, which can be obtained under the rather restrictive conditions of an infinite plane surface. Placing the surface in the $(xz)$-plane at $y = 0$ we get.

$$\phi(y) = 4V_t \arctanh \left[ \tanh \left( \frac{\zeta}{V_t} \right) \exp \left( -\frac{y}{\lambda_D} \right) \right]$$ \hspace{1cm} Gouy-Chapman solution (14)$$

$$V_t = \frac{k_BT}{Ze} \hspace{1cm} \lambda_D = \sqrt{\frac{ek_BT}{2Ze^2c_0}}$$  \hspace{1cm} (15)$$

Where $V_t$ is the thermal voltage, $\lambda_D$ is the so-called Debye length and $\zeta$ is the potential of the surface, i.e. the potential which can be attributed to the surface charge density on the wall. In this case where the surface charge is negative, the $\zeta$-potential will also be negative. This solution is however an inconveniently cumbersome expression to work with, and it’s complexity precludes the simple derivation of some of the properties of
the Debye layer. Therefore we shall employ the so-called Debye-Hückel assumption, which states that the \( \zeta \)-potential is appreciably smaller than the thermal voltage \( V_t \). As the \( \zeta \)-potential has the highest absolute value of the potential, this assumption ensures that the argument of the hyperbolic sine in (13) is always much smaller than 1, which allows for a Taylor expansion to the first order of the hyperbolic sine. In this way we get

\[
\nabla^2 \phi(r) \approx 2Z^2e^2c_0 \frac{\phi(r)}{ek_BT} = \frac{1}{\lambda_D^2} \phi(r) \tag{16}
\]

In the simple translation symmetric case of a planar surface in the \( xz \)-plane at \( y = 0 \) this equation is easily seen to yield the solution

\[
\phi(y) = \zeta \exp \left( -\frac{y}{\lambda_d} \right) \tag{17}
\]

Two trivial results can readily be derived from this expression for the potential. By inserting it in the Poisson equation the charge density is obtained

\[
\rho_{el} = -\frac{\epsilon \zeta}{\lambda_D^2} \exp \left[ -\frac{y}{\lambda_D} \right] \tag{18}
\]

By virtue of the Debye-Hückel assumption the concentration from (11) can be Taylor expanded and upon insertion of the potential from (17) in this expression we get

\[
c_\pm = c_0 \left[ 1 \mp \frac{\zeta}{V_t} \exp \left( -\frac{y}{\lambda_d} \right) \right] \tag{19}
\]

A final useful result is the potential in an infinite parallel plate channel, where the surfaces are situated at \( y = \pm h/2 \). We get the same basic solution of the Poisson-Boltzmann equation as in (17), but by applying the boundary conditions we get the full solution

\[
\phi(y) = \zeta \frac{\cosh \left( \frac{y}{\lambda_D} \right)}{\cosh \left( \frac{h}{2\lambda_D} \right)} \tag{20}
\]

As it turns out the Debye-Hückel approximation is actually valid over a wider range of values than should be expected. In fact by Taylor expanding the Gouy-Chapman solution in \( \zeta^4 V_t \) around zero it is seen that it is identical to the Debye-Hückel solution up to and including the third term. By investigating the two solutions further it is seen that the Debye-Hückel approximation is valid for \( \zeta \leq 4V_t \).

In the numerical implementation of the model it is of interest to know the relation between surface charge and \( \zeta \)-potential. We observe a plane surface with surface charge \( \tilde{q}_s \) and use Gauss’s law to find the enclosed charge in a thin box of upper surface area, \( A \), and height, \( \delta h \). Integrating the divergence of the \( \mathbf{D} \)-field over the volume of the box and applying the divergence theorem, we find that the charge enclosed in the box,
Q = Aq_s equals the flux out through the upper surface of the box, εE. Using the Debye-Hückel expression for the potential this gives

\[ q_s = εE = -ε\frac{∂φ(y)}{∂y}\bigg|_{y=0} = \frac{ε}{λD}ζ \]  

(21)

So far we have only described the Debye layer’s influence on the electrical part of the system. What is of real interest though is to find it’s influence on the fluid-dynamic part of the system. In the following sections we will therefore attempt to make a full electrohydrodynamic description of the Debye layer and the system.

2.2 Electroosmosis

A concept that is of particular interest in relation to the Debye layer is the electroosmotic effect. We remember that the formation of the Debye layer left the fluid with a net charge, and that there was a charge build up along the surface of the walls. If we now were to bias the channel along the longitudinal axis the resulting electric field, \( E = E_x e_x \) would affect the ions in the liquid. Assuming that we have negatively charged walls and the highest potential on the left terminal of the channel, the counterions in the Debye layer would be cations and they would be swept right by the field. The liquid in the Debye layer would thus move right and through viscous forces drag the rest of the liquid along. The electric field would of course also affect the anions, but as the concentration of those would be slightly lower than that of the cations, the net movement of the liquid would be to the right. In the simplified case of an infinite parallel plate channel with a ζ-potential not violating the Debye-Hückel assumption, we can quantitatively describe the electroosmotic effect by using the Navier-Stokes equation, (2), in conjunction with the equation describing the charge density, (20). In order to make the description possible we will make some simplifying assumptions.

(a) It is assumed that we can use the charge density expression derived for a Debye layer in equilibrium, ie. that the applied electric field does not influence the charge density. As the electric field only affects the ions in the Debye layer horizontally this should be a valid assumption. (b) The ζ-potential is assumed to be constant. (c) The electric field is assumed to be homogenous, ie. the fields from ions and surface charge on the walls are neglected. (d) The flow is in steady-state. (e) The Debye length is assumed to be negligible compared to the height of the channel. (f) There is no pressure difference between the terminals of the channel.

Under these conditions the velocity field only has an \( x \)-component and the Navier-Stokes equation reduces to

\[ η\frac{∂^2}{∂y^2}u_x(y) = -ρel(y)E_x = εE_x\frac{∂^2}{∂y^2}φ(y) \]  

(22)

where we have used the Poisson equation. Rearranging we get

\[ η\left(\frac{∂^2}{∂y^2}u_x(y) - \frac{εE_x}{η}φ(y)\right) = 0 \quad ⇔ \quad u_x(y) - \frac{εE_x}{η}φ(y) = αy + β \]  

(23)
At the boundaries \( y = \pm \frac{h}{2} \) the potential is \( \zeta \) and the velocity is zero. These two boundary conditions gives us that \( \beta = -\frac{\epsilon E}{\eta} \zeta \), and since the velocity field and potential needs to be symmetric it is required that \( \alpha = 0 \). By inserting the potential expression we get

\[
 u_x(y) = \frac{\epsilon E}{\eta} (\phi(y) - \zeta) = \left( 1 - \frac{\cosh \left( \frac{y}{\lambda D} \right)}{\cosh \left( \frac{h}{\lambda D} \right)} \right) v_{eo} \tag{24}
\]

where the Helmholtz-Smoluchowski velocity is given by

\[
 v_{eo} = -\frac{\epsilon \zeta}{\eta} E_x \tag{25}
\]

which is a positive value for negative surface charges.

### 2.3 Induced Charge Electroosmosis

Basic electromagnetism tells us that any metal placed in an external field will develop a surface charge in order to shield its interior from the field. That is the electric field will induce a surface charge in the metal. In this section we will look into the electrohydrodynamic significance of this effect.

If we place a piece of metal in an electrolyte and apply a bias across the system, there will be induced a surface charge on the metal surface which will in turn create a Debye layer around the metal. As we have shown in the previous section this will give rise to an electroosmotic effect, which will move the liquid around in the system. We will now treat this problem analytically in the simple case of an immersed metal cylinder of radius \( a \) with a homogenous field applied orthogonal to the cylinder axis.

We assume that the Debye layer around the cylinder has already formed, and thus we will only deal with the steady-state description of the system. The external field is \( \mathbf{E} = E_0\mathbf{e}_x \) and the corresponding potential is \( \phi_0 = -E_0 r \cos(\theta) \) in cylindrical coordinates. As the Debye layer constitutes a charge density the electric potential is described by the Poisson equation. It is however not trivial to solve the Poisson equation in this case, and we will therefore assume an infinitesimal thin Debye layer and solve the Laplace equation instead. The boundary condition on the cylinder surface must thus be taken to be the condition just outside the Debye layer. As the Debye layer forms an effective shielding of the field this condition is that the normal component of the field is zero at the boundary, \( \mathbf{E} \cdot \mathbf{e}_r \big|_{r=a} = 0 \). At infinity the potential is undisturbed by the metal cylinder, ie. \( \lim_{r=\infty} \phi(r) = \lim_{r=\infty} \phi_0(r) \). In order to solve the Laplace equation we make a guess at a solution. The solution is assumed to have the same angular dependence as at infinity, and the remainder of the solution can be found using the Frobenius method. As the Laplace equation is a fairly simple equation we can however have a justified hope that it will be sufficient to just use a single term of the infinite power series. Our guess at a solution thus becomes \( \phi(r, \theta) = r^p \cos(\theta) \). Inserting this in
the Laplace equation gives
\[ \nabla^2 \{ r^p \cos (\theta) \} = \frac{1}{r} \partial_r \left[ r \partial_r \{ r^p \cos (\theta) \} \right] + \frac{1}{r^2} \partial_\theta \{ r^p \cos (\theta) \} = 0 \] (26)
\[ \Rightarrow (p^2 - 1) \cos (\theta) = 0 \quad \Leftrightarrow \quad p = \pm 1 \] (27)

As the Laplace equation is a second order homogeneous differential equation, there are only these two solutions and our guess at a solution has thus been succesful. The full solution is a linear combination of the found solutions, and we will now use our two boundary conditions to find the two arbitrary constants. The boundary condition for the far field yields
\[ \lim_{r \to \infty} \phi(r, \theta) = \lim_{r \to \infty} \phi_0(r, \theta) \Rightarrow \lim_{r \to \infty} \left[ A \frac{1}{r} \cos (\theta) + Br \cos (\theta) \right] = \lim_{r \to \infty} (-E_0 r \cos (\theta)) \] (28)
\[ \Leftrightarrow \lim_{r \to \infty} Br = \lim_{r \to \infty} (-E_0 r) \quad \Leftrightarrow \quad B = -E_0 \] (29)

To find \( A \) we write the electrical field in terms of the potential and employ the boundary condition at the cylinder surface
\[ E = -\nabla \phi(r, \theta) = \left( E_0 + \frac{A}{r^2} \right) \cos (\theta)e_r + \left( -E_0 + \frac{A}{r^2} \right) \sin (\theta)e_\theta \] (30)
\[ E \cdot e_r |_{r=a} = 0 \quad \Leftrightarrow \quad \left( E_0 + \frac{A}{a^2} \right) \cos (\theta) = 0 \quad \Leftrightarrow \quad A = -E_0 a^2 \] (31)

In this way we have found that the full solution is
\[ \phi(r, \theta) = -E_0 r \cos (\theta) \left( 1 + \frac{a^2}{r^2} \right) \] (32)

The \( \zeta \)-potential is the potential drop across the Debye layer, and because the surface potential of the cylinder is zero we get
\[ \zeta(\theta) = 0 - \phi(a + \lambda_D, \theta) \approx -\phi(a, \theta) = 2E_0 a \cos (\theta) \] (33)

From (32) we get the tangential component of the field near the surface
\[ E_{\parallel}(a, \theta) = -e_\theta \cdot \nabla \phi(r, \theta) |_{r=a} = -\frac{1}{r} \partial_\theta \phi(r, \theta) |_{r=a} = -E_0 \left( 1 + \frac{a^2}{r^2} \right) \sin (\theta) |_{r=a} = -2E_0 \sin (\theta) \] (34)

The Helmholtz-Sholuchowski velocity thus is
\[ \mathbf{v}_{eo}(\theta) = -\frac{e \zeta(\theta)}{\eta} E_{\parallel}e_\theta = 4 \frac{e E_0^2 a}{\eta} \cos (\theta) \sin (\theta) e_\theta = 2 \frac{e E_0^2 a}{\eta} \sin (2\theta) e_\theta \] (35)

where we have used an addition formula to obtain the final result. It is seen that this gives rise to a symmetric flow around the cylinder as the one seen in figure 1.
3 Dimensionless equations

To use COMSOL with dimensionless variables, we have to find the nondimensional forms of the equations, and then compare them with the equations, that are used in COMSOL. The nondimensional variables will be marked with a ~

We introduce the following reference parameters, so that \( x = x_{\text{ref}} \tilde{x} \):

<table>
<thead>
<tr>
<th>Variable</th>
<th>Dimensionless Form</th>
</tr>
</thead>
<tbody>
<tr>
<td>( L_{\text{ref}} )</td>
<td>( W )</td>
</tr>
<tr>
<td>( c_{\text{ref}} )</td>
<td>( c_0 )</td>
</tr>
<tr>
<td>( u_{\text{ref}} )</td>
<td>( \frac{e_0 c_{\text{ref}}}{\eta} E_{\text{ref}} )</td>
</tr>
<tr>
<td>( \phi_{\text{ref}} )</td>
<td>( \frac{k_B T}{\epsilon_0} )</td>
</tr>
<tr>
<td>( E_{\text{ref}} )</td>
<td>( \frac{\phi_{\text{ref}}}{L_{\text{ref}}} )</td>
</tr>
<tr>
<td>( t_{\text{ref}} )</td>
<td>( \frac{L_{\text{ref}}}{u_{\text{ref}}} )</td>
</tr>
<tr>
<td>( p_{\text{ref}} )</td>
<td>( \frac{u_{\text{ref}} \eta}{L_{\text{ref}}} )</td>
</tr>
<tr>
<td>( q_{\text{ref}} )</td>
<td>( \frac{e_0 c_{\text{ref}}}{L_{\text{ref}}} )</td>
</tr>
</tbody>
</table>

3.1 The Navier-Stokes equation

We will start with the Navier-Stokes equation (2).

\[
\rho (\partial_t u + (u \cdot \nabla)u) = -\nabla p + \eta \nabla^2 u + \rho_{el} E
\]

\[
\rho_{el} = Ze (c_+ - c_-), \quad E = -\nabla \phi
\]
which in a nondimensional form can be written as

\[ \rho \left( \frac{u_{\text{ref}}}{L_{\text{ref}}} \partial_t \tilde{u} + \frac{u_{\text{ref}}}{L_{\text{ref}}} (\tilde{u} \cdot \nabla) \tilde{u} \right) = -\frac{1}{L_{\text{ref}}} \nabla p_{\text{ref}} + \frac{\eta}{L_{\text{ref}}} \nabla^2 u_{\text{ref}} \tilde{u} - \frac{N_A c_{\text{ref}} Z e (c_+ - c_-) \phi_{\text{ref}}}{L_{\text{ref}}} \nabla \tilde{\phi} \]

By dividing the equation with the coefficient on the second term, \( \frac{\eta}{L_{\text{ref}}} \), it becomes

\[ \frac{\rho u_{\text{ref}} L_{\text{ref}}}{\eta} \left( \partial_t \tilde{u} + (\tilde{u} \cdot \nabla) \tilde{u} \right) = -\nabla \tilde{p} + \nabla^2 \tilde{u} - \frac{N_A c_{\text{ref}} Z e (c_+ - c_-) L_{\text{ref}} \phi_{\text{ref}}}{u_{\text{ref}} \eta} \nabla \tilde{\phi} \]

By defining the reference Reynolds number for this system, \( \text{Re}_{\text{ref}} = \frac{\rho L_{\text{ref}} u_{\text{ref}}}{\eta} \), and introducing the reference Debye length, \( \lambda_{D,\text{ref}} = \sqrt{\frac{\epsilon_0 \epsilon_r k_B T}{2Z^2 e^2 N_A c_{\text{ref}}}} \), with \( \phi_{\text{ref}} \) and \( u_{\text{ref}} \) inserted, we get

\[ \text{Re}_{\text{ref}} \left( \partial_t \tilde{u} + (\tilde{u} \cdot \nabla) \tilde{u} \right) = -\nabla \tilde{p} + \nabla^2 \tilde{u} - \frac{1}{2} \frac{L_{\text{ref}}^2}{\lambda_{D,\text{ref}}^2} (c_+ - c_-) \nabla \tilde{\phi} \quad (36) \]

The continuity equation

As the continuity equation

\[ \nabla^2 \phi = 0 \]

is homogeneous, we have the same equation in the nondimensional form;

\[ \nabla^2 \tilde{\phi} = 0 \quad (37) \]

The Poisson equation

The Poisson equation is given by equation (6) and the nondimensional form is found

\[
\begin{align*}
\nabla^2 \phi &= -\frac{\rho_{\text{el}}}{\epsilon} \quad \text{(6)} \\
\phi_{\text{ref}} \nabla^2 \tilde{\phi} &= -\frac{c_{\text{ref}} N_A Z e (c_+ - c_-)}{\epsilon_0 \epsilon_r} \Rightarrow \\
\nabla^2 \tilde{\phi} &= -\frac{1}{2} \frac{L_{\text{ref}}^2}{\lambda_{D,\text{ref}}^2} \frac{2c_{\text{ref}} N_A Z^2 e^2 (c_+ - c_-)}{k_B T \epsilon_0 \epsilon_r} = -\frac{1}{2} \frac{L_{\text{ref}}^2}{\lambda_{D,\text{ref}}^2} \frac{(c_+ - c_-)}{k_B T \epsilon_0 \epsilon_r} \quad (38)
\end{align*}
\]

Where we have used the reference Debye length, \( \lambda_{D,\text{ref}} = \sqrt{\frac{\epsilon_0 \epsilon_r k_B T}{2Z^2 e^2 N_A c_{\text{ref}}}} \).
3.2 Convection-diffusion equation

The convection-diffusion equation is given by equation (8). Because we look at systems in steady state, the time-dependent term vanishes. The nondimensional form of this equation becomes

\[- \frac{D \pm}{L^2_{ref}} \nabla^2 c_{\pm} + \frac{u_{ref}}{L_{ref}} \nabla c_{\pm} = \frac{D}{L^2_{ref}} \left( \frac{\phi_{ref}}{L^2_{ref}} \nabla c_{\pm} \cdot \nabla \phi_{\pm} + \frac{\phi_{ref}}{L^2_{ref}} c_{\pm} \nabla^2 \phi_{\pm} \right) = 0\]

Where we have divided with $c_{ref} N_A$. Now we divide with $\frac{D}{L^2_{ref}}$ and by introducing the reference Peclet number, $Pe_{ref} = \frac{u_{ref} L_{ref}}{D_{\pm}}$, we get

\[- \nabla^2 c_{\pm} + Pe_{\pm(\text{ref})} \nabla c_{\pm} = 0\]  \hspace{1cm} (39)

The Helmholtz-Smoluchowski velocity

To find the nondimensional form of the Helmholtz-Smoluchowski velocity we insert the nondimensional form of the velocity and the electric field in equation (25);

\[\tilde{u} u_{ref} = -\frac{\varepsilon_r \varepsilon_0 \phi_{ref} \xi}{\eta} E_{ref} \tilde{E} \iff \tilde{u} = -\xi \tilde{E}\]  \hspace{1cm} (40)

4 Numerical simulations

To simulate how the flow and the concentrations develop in the channel, we use the program COMSOL, which uses the finite element method to find the solution. With this tool we can solve this coupled problem with six differential equations. We draw the channel in the GUI and implement all the boundary and initial conditions as well as the constants and expressions which are used in the equations. In figure 2 is shown a top view of the structure of a dummy model with length, $L = 3$, width, $W = 1$ and one pair of semicylinders. The mesh can also be seen in.

In the convection-diffusion equation the diffusion term is dependent on the second derivative of the concentration, so in the interpolation between the mesh points the concentration needs to be expressed in polynomials of at least second order to take the diffusion into account. Therefore COMSOL is programmed to fit the concentration between the mesh points to a second order polynomial. This gives rise to a problem when there is discontinuities in the concentration between two mesh points, since this cannot be fitted without getting concentrations in the nearby mesh points which are not physical acceptable, i.e. below zero or larger than inlet concentration. When modelling the full system with resolved Debye layer there will naturally be concentrations above inlet concentrations, making it hard to check if the model is suffering from these mesh problems, but negative concentrations is never physical acceptable.
4.1 The full model

The problem can be modelled in two ways. The most correct way is to have charged walls and simulate the cylinders as metal, which can be done by giving them a huge permittivity. This creates a Debye layer on the walls and get induced charge on the metal cylinders which also creates a Debye layer on these. As described in the theory, such a layer is around $1 - 100 \text{ nm}$. We define a parameter, $\frac{W}{\lambda_D}$, to compare the Debye length with the width of the channel, $W$. If we have a Debye layer of 100 nm, we need a fine mesh, and if we have a structure with dimensions of $1 \mu \text{m} \times 10 \mu \text{m}$, we need a very large computer memory to resolve the Debye layer. To see if our program works we starts with a concentration which gives a Debye layer of minimum 300 nm.

In COMSOL we implement three modules in the model; Electrokinetic Flow, Electrostatics and Incompressible Navier-Stokes.

Electrokinetic flow

At first we define the electrokenetic flow as inactive in the metal cylinders because there can not be any concentration in these domains. On the boundary where the fluid should flow into the channel we make the boundary condition for the charged ions, to be a constant concentration, $c_{+,0}$ and $c_{-,0}$. For the uncharged solute, that we wants to mix, we have a concentration of $c_{s,0}$ in one half of the channel and zero in the other half. Because of this we have a discontinuity, which means that we need a fine mesh in this area.

On all the walls including the metal semicylinders, except the outlet, the boundary condition is chosen to be “Insulation/Symmetry”.

The end of the channel is the outlet for the fluid, and therefore also for the concentration, so the boundary conditions for the solute concentration should be “Convective flux”. For the ions it is important to remember that the potential drop across the channel
gives rise to a current. This means that for an electric field in the \( x \)-direction, \( \mathbf{E} = E_0 \hat{e}_x \), positive ions will be going in the positive \( x \)-direction and negative ions in the negative \( x \)-direction. To sustain a current like this, we need a constant concentration of positive ions at the inlet and negative ions at the outlet.

In this multiphysics module, we consider the concentration distribution, which means that the equations that are used in COMSOL is the convection-diffusion equation. To use the nondimensional equation, we compare it with the one used in COMSOL. In COMSOL the equation is written as

\[
\nabla(-Dc_{\pm,s} - Zu_m Fc_{\pm,s} \nabla V) = R - \mathbf{u} \cdot \nabla c_{\pm,s}
\]

If we compare this with equation (39) we have that

\[
D_{\pm,s} = \frac{1}{Pe_{\pm,s(ref)}}, \quad s \text{ refers to the solute} \quad (42)
\]

\[
u_m = \frac{1}{e_N A Pe_{\pm,s(ref)}} \quad (43)
\]

\[
Z = [Z_p, Z_m, Z_s], \quad u = u, \quad v = v, \quad V = V \quad (44)
\]

**Incompressible Navier-Stokes**

As in the “electrokinetic flow” module, the equations are inactive in the semicylinders, because there is no flow in these domains. The boundary conditions on the walls including the semicylinders are “no slip”, and for the inlet and the outlet, they are “Pressure, no viscous stress” with zero pressure. With these boundary conditions the fluid would not flow without a voltage drop. The equations are Navier-Stokes and the continuity equation. In COMSOL these equations are written as

\[
\rho (\mathbf{u} \cdot \nabla) \mathbf{u} = \nabla \cdot \left[-p \mathbf{I} + \eta \left( \nabla \mathbf{u} + (\nabla \mathbf{u})^T \right) \right] + \mathbf{F}
\]

and

\[
\nabla \mathbf{u} = 0
\]

A comparison with equation (36) and (37) respectively, gives us that

\[
\rho = R e_{ref}, \quad \eta = 1, \quad F_{x,y} = -\frac{1}{2} \frac{L_{ref}^2}{\lambda_{D,ref}^2} \partial_{x,y} V (c_+ - c_-)
\]

**Electrostatics**

At the inlet we have an electric potential, \( V \), while the outlet is grounded. The walls have a negative surface charge, \( -\tilde{q}_s \), while the semicylinders just have continuity at the boundaries. The walls behind the semicylinders, which are not really a part of the
channel, have “Zero charge/Symmetry” so that they will not affect the system. The equation acting in this multiphysics module is the Poisson equation, (38). In COMSOL it’s written as

\[-\nabla (d \epsilon_r \nabla V) = d \rho (48)\]

Comparing this equation with equation (38), we have

\[d = 1, \quad \rho = (Z_m \epsilon_m + Z_p \epsilon_p) e N_A\]

and \(\epsilon_r\) is the relative permittivity, which is very large in the semicylinders, so that it acts like metal. We use a relative permittivity in the fluid \(\epsilon_{r, fluid} = 80\) and in the metal \(\epsilon_{r, metal} = 10^{18}\)

### Problems with the modelling

Due to memory limitations, we run into problems when modelling the problem as described in the previous. It is possible to simulate a channel which in nondimensional parameters has the dimensions \(W = 1\) and \(L = 3\) and one set of semicylinders. By using the parametric solver in COMSOL, we succeeded in simulating a channel with \(L = 10\) and one set of semicylinders, with a surface charges varying from \(\tilde{q}_s = 20\) to \(\tilde{q}_s = 160\). With larger charges we get a larger flow rate, and the equations become more nonlinear, see figure 3.

Even with the parametric solver we run out of memory, and with more than one set of semicylinders we can’t find a solution for any values of \(\tilde{q}_s\). Because we want to find out how to make the best mixer, i.e. how many semicylinders and what the distance between them, should be, we have to use another model, which does not crave as much memory.

#### 4.2 Slip model

When using the slip model we model the problem with sliding walls which moves the fluid, instead of having the Debye layer moving it. This means that we do not need as fine a mesh as in the previous model, because we don’t need to resolve the Debye layer. The slip model uses the Helmholtz-Smoluchowski velocity as the velocity on the walls, described in the theory, equation (25).

In this modelling we still use the “Incompressible Navier-Stokes” module and the “Electrostatics” module but we have no charged ions, which means that there is no electroosmotic flow. Instead we use the “convection-diffusion” module to implement the concentration distribution of the solute we want to mix in the channel.

In the “Incompressible Navier-Stokes” the same values and expressions are implemented in the subdomain, except that the forces are zero, because the concentrations of
the charged particles are zero. The boundaries are now walls with electroosmotic velocity, which simulates the Debye layer. As seen in the theory, the velocity depends on the electric field and the $\zeta$-potential. The electric field is just the gradient of the potential, so $E_x = -\partial_x V$ and likewise for $E_y$.

Comparing equation (25) with the equation in COMSOL, we have that

$$\zeta_C = \frac{\tilde{\zeta}}{\varepsilon_r \varepsilon_0}$$  \hspace{1cm} (49)

Where $\zeta_C$ is the $\zeta$-potential used in COMSOL, and $\tilde{\zeta}$ is the real nondimensional $\zeta$-potential which for the cylinder boundaries is calculated as the difference between the potential inside a cylinder ($\phi_0$) and the potential just outside ($V$)

$$\zeta = \phi_0 - V$$

and for the channel edges as[2]

$$\zeta_0 = 2\arcsinh\left(\frac{-q_s \lambda_{D,ref}}{2 \frac{L_{ref}}{L}}\right)$$  \hspace{1cm} (50)

In the “Electrostatics” module we have the same subdomain settings as before, except that there is no field in the cylinders, because now we don’t use that it is metal, but just

Figure 3: The concentration of the charged ions for a high and a low surface charge value. As can be seen, the concentration becomes nonlinear for high values of the surface charge.
use the slip-velocity on the surface. The boundary condition is therefore changed to "Zero charge/Symmetry" on the cylinder edges, while the rest of the boundary conditions are the same.

Convection and diffusion

In the "convection and diffusion" module, COMSOL uses the convection-diffusion equation, equation (8) and in the nondimensional form equation (39). In COMSOL this equation is

\[ \nabla(-D \nabla c) = R - u \cdot \nabla c \]  

(51)

When comparing these equations we arrive at

\[ D = \frac{1}{Pe_{ref}}, \quad R = 0, \quad u = u, \quad \text{and} \quad v = v \]

At the inlet we have a concentration, which is zero on half of it and \( c_{s,0} \) in the other half. At the outlet there is convective flux.

5 Validation of the slip model COMSOL script

Before we start using the COMSOL script based on the slip model, we want to make sure that it produces physically acceptable results. We can do this by comparing the results obtained in COMSOL with the analytical results in some simple cases. As we have decoupled the flow-, convection-diffusion-, and electrostatic problems in the slip model, we can check the modules one at a time. The electrostatic module isn’t that hard to validate as the potential isn’t affected by anything in the slip model, and as the potential is only used to read off a value at the top of each semicylinder. Theoretically the potential should vary linearly through the channel, and by simple inspection in the GUI we see that it is so.

Secondly we look at the velocity field in a channel without any semicylinders. The velocity field should in this case be a simple plug-flow with the same velocity everywhere, namely the Helmholtz-Smoluchowski velocity. Again this is easy to check by inspection, and as we have defined the velocity at the walls to be the Helmholtz-Smoluchowski velocity in the model, it isn’t really surprising that the desired result is obtained.

Lastly we need to validate the convection-diffusion module. Because the convection-diffusion problem can’t be reduced to a 1D problem and because of the discontinous boundary condition at the channel inlet, this isn’t as easily accomplished as the validation of the other modules. First and foremost we will need an analytical solution to compare our results with. Because of the concentration discontinuity at the boundary, separation of variables is bound to fail, and indeed there exists no easy solution. In a
system such as the one we are looking at, the solution can be found in the form of an infinite series \[3\]

\[
\tilde{c}(\tilde{x}, \tilde{y}) = r + \frac{2}{\pi} \sum_{n=1}^{\infty} \frac{\sin (n\pi \tilde{y})}{n} \cos (n\pi \tilde{y}) \exp \left( -\frac{2n^2\pi^2}{Pe + \sqrt{Pe^2 + 4n^2\pi^2}} \tilde{x} \right)
\]  \hspace{1cm} (52)

where \(\tilde{x}\) and \(\tilde{y}\) are the nondimensionalized spatial coordinates given by \(\tilde{x} = \frac{x}{L_{ref}}\) and \(\tilde{y} = \frac{y}{L_{ref}}\). \(Pe\) is the Peclet number given by \(Pe = \frac{UL_{ref}}{D}\), where in contrast to the definition used elsewhere in this report the velocity is not the reference velocity. \(r\) is the mixing ratio, i.e. a ratio determining how much of the channel is initially filled with solute. In our case the mixing ratio is \(r = 0.5\).

By inserting (52) in (1) we get an analytical expression for \(\eta'\) which can be compared to the \(\eta'\) found in the COMSOL model. The plot of both \(\eta'\)'s versus \(\tilde{q}_s\) can be seen in figure 4 and they are seen to agree excellently.

The COMSOL model has now been validated and we can proceed with the simulations.

![Mixing index for modelled and analytical concentrations](image)

**Figure 4:** \(\eta'_{mix}\) is plotted as a function of the scaled surface charge, both for the analytical and the slip model concentrations for a channel with no semicylinders. As can be seen the mixing indexes are very similar.

### 6 The dummy models

Since we are unable to model the system with more than one semicylinder pair and resolved Debye layer, and therefore are forced to simplify our model to the slip model, we need to compare the two types of models. To do that we use two dummy models.
with the two types of model setup. These dummy models are created as $1\,\mu m \times 3\,\mu m$ channels with a single pair of semicylinders positioned $1\,\mu m$ from the inlet. We implement the $\eta_{mix}$ function in both scripts together with the appropriate multiphysics modules and compare the development of $\eta_{mix}$ as a function of the dimensionless position and different values of $\tilde{q}_s$. These results are shown in figure 5 and 6. Its clear that

![Figure 5: Mixing index throughout the channel for the full dummy model and different values of $\tilde{q}_s$. Its easy to see the effect of the boundary condition on the mixing index, so to determine a valid mixing index for the system its important not to read out the outlet value.](image)

the boundary condition at the outlet has an effect on the mixing index and its therefore important not to check the value of the mixing index at the outlet. So for the modelling of the long channel we decided to take the maximum value of $\eta_{mix}$ as the output value. As seen in figure 6 this method should give the right output value of the mixing index. A plot of the velocity profiles for both models at different values of $\tilde{q}_s$ shows that the slip model takes advantage of the velocity independent mixing index, because the velocity profile is parabolic (see figure 7) with maximum velocity at the maximum concentration. Whereas the plot of the velocity profile for the full model is much more like a plug-flow (see figure 8), so it won’t change much with a velocity dependent definition of the mixing index. This means that for a more accurate description one should take the flow rate at the given point into account when determining the mixing index. This could easily be done by multiplying with the velocity at each point in the sum in equation (1). Then the mixing index would be an indication of the amount of mixed
liquid leaving the system, which seems to be a much better way of comparing the mixing performance of different systems. In this report we will however use the original definition of the mixing index, equation (1), to allow for a comparison with the results of the article [1].

7 Optimization of the model

Before making the final configuration of the model, we want to investigate which geometry to use in order to get the best mixing performance. The dimensions of the channel are fixed, and so are the dimensions of the semicylinders. The reason why we will not change the dimensions of the semicylinders is that we want the ICEO to be the dominant effect, and increasing the semicylinder size would create significant hydraulic resistance which alone would allow for better mixing.

The parameters we can vary are the cylinder spacing and the mutual displacement, i.e. the relative positions of the upper and lower semicylinders.
7.1 Mixing performance dependence on cylinder spacing

In order to investigate the cylinder spacing’s effect on the mixing performance, we have made a geometry with four semicylinders at either side of the channel. We have then made a script which varies the scaled distance between the semicylinder centers from 0.5 to 1.8, while at the same time varying the surface charge $\tilde{q}_s$ from 20 to 200. In figure 9 we have plotted the unmodified mixing index $\eta_{mix}$ versus semicylinder spacing for each value of $\tilde{q}_s$. It is seen that the mixing index increases monotonically with semicylinder spacing for all values of $\tilde{q}_s$, but that the performance increase is negligible when the distance is more than 1. As it is desirable to have as short mixing distance as possible, it can be concluded that the best overall mixing performance will be achieved for a semicylinder spacing of around 1.

7.2 Mixing performance dependence on mutual displacement

We now want to investigate any effect of a mutual displacement of the semicylinders. To do this we use the same basic geometry as above, and then displace all of the semicylinders, in the lower half, to the right. By plotting the unmodified mixing index versus the nondimensionalized $\tilde{x}$-coordinate it is seen, that the mixing index is slightly lower when the upper and lower semicylinders are displaced. In figure 10 the unmodified mixing index is plotted versus the nondimensionalized $\tilde{x}$-coordinate for the case where the semicylinders are aligned, and the case where they are displaced by half the cylinder spacing. In figure 11 the concentration distribution, in the channel with mutual displaced semicylinders, is plotted.

In order to get the best mixing performance we should thus make a symmetric ge-
Figure 9: Unmodified mixing index $\eta_{mix}$ plotted versus semicylinder spacing for $\tilde{q}_s$ varying from 20 to 200. It is seen that the mixing index increases monotonically with semicylinder spacing for all values of $\tilde{q}_s$, but that the performance increase is negligible when the spacing becomes larger than the height of the channel.

Figure 10: The mixing index for a channel with four semicylinder pairs, with a mutual displacement of a half spacing, $\tilde{q}_s = 80$. The mixing index is slightly lower than for the symmetric channel with $\tilde{q}_s = 80$. 
ometry. However, we can see that the difference in mixing index is pretty small, so a misalignment in a fabrication process would not affect the mixing performance particularly.

8 Dependence on the number of cylinder pairs

To determine the dependence of the number of semicylinderpairs we have simulated 5 different microfluidic systems, with 7, 5, 3, 1, and 0 semicylinderpairs, and seen how their mixing performance changes with the surface charge $\tilde{q}_s$.

We have replicated the results from the article [1], shown in figure 12, by defining the 5 different geometries in COMSOL and used the slip model described earlier. These geometries were implemented in a MATLAB script with corresponding multiphysics modules, which then calculated $\eta'_{mix}$ for different values of the surface charge $\tilde{q}_s$. For the simplest model (with no semicylinders) we have used the validation methods described in the validation section to ensure correct results, as seen in figure 4 our modelling is valid. By comparing figure 13 with figure 12 it is shown that we are able to generate almost the same results using the slip model as the full model with resolved Debye layer. Quantitatively the results varies a little where we get a slightly better performance for large number of semicylinderpairs and a slightly worse performance for a few number of cylinder pairs. So the slip model is fairly accurate in modelling these kind of systems, and it is possible to obtain qualitatively the same results and reasonable quantitative results. Even though it needs a lot fewer computations to solve.
Figure 11: The concentration distribution in the channel with mutual displaced semicylinders.

Figure 12: The mixing index dependence of the number of semicylinder pairs and the scaled surface charge using the full model. This graph is made by Mrnal Jain, Anthony Yeund, and Krishnaswamy Nandakumar and is taken from the article [1](Fig. 11).

Figure 13: The mixing index, $\eta_{\text{mix}}'$, plotted as a function of the scaled surface charge for different numbers of semicylinder pairs; 7, 5, 3, 1, and 0. Slip model.
9 Conclusion

In this report we have simulated an ICEO micromixer and optimized it with regards to the spacing between the semicylinders and their displacement. To evaluate the micromixer we used a mixing index, $\eta_{mix}$. Due to computational limitations we made the simulations using a simpler model, the slip model, instead of solving the full problem with a resolved Debye layer. The effect of making this simplification was investigated using a dummy model, and we found that we should expect to get a slightly better mixing performance when using the slip model. We discovered that the mixing index increases monotonically with semicylinder spacing, but that it was nearly constant for semicylinder spacings of more than one channel height. We found that a displacement of one of the semicylinder rows only affected the mixing index slightly, and that the optimal configuration is with the semicylinders placed directly across from each other. Finally we compared our results to the ones obtained with a resolved Debye layer by Mranal Jain, Anthony Yeung, and Krishnaswamy Nandakumar in their paper *Efficient Micromixing Using Induced-Charge Electroosmosis* [1]. We found our results to be qualitatively similar, but our results generally had higher mixing indexes for the channels with many semicylinders and lower mixing indexes for the channels with few semicylinders. The higher mixing indexes were expected on the basis of our dummy model investigation, but we are unable to account for the lower mixing indexes we get with few semicylinders.
References

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