

CHALMERS



TITLE OF PROJECT

Master's Thesis in ...

ANDREAS BÜLLING

Department of Mathematical Sciences
CHALMERS UNIVERSITY OF TECHNOLOGY
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Abstract

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Acknowledgements

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The Authors, Location 11/9/11

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1

Introduction

Short intro for this section

1.1 Background

Historically - LBM and modelling of electrohydrodynamics. Development til today.

1.2 General problem description

Description of problem + what we want to achieve in this work.

1.3 Outline

How is this report structured?

2

Electrohydrodynamics in microchannels

In this chapter, the fundamental physics behind electrokinetic flow, important for later discussions, will be presented. Particularly, a modelling approach based on the coupling of Navier-Stokes, Nernst-Planck and Poisson's equations is given.

2.1 Basic concepts of electrokinetic flow

Electrohydrodynamics involves the study of electric phenomena on fluid flow. How fluids carrying electrical charges (electrolytes) react upon external electrical fields or interact with charged objects are examples of problems that arise in this field.

2.1.1 Electrical double layers

As a charged object is brought into contact with an electrolyte it is, qualitatively, easily deduced that ions with a sign of charge opposite to that of the object will be attracted to the object and ions with the same sign of charge will be repelled. These two distinct categories of ions will from hereon be referred to as counter- and co-ions respectively. In this case, for a neutral electrolyte, a surplus of counter-ions will be present in the direct vicinity of the object and a surplus of co-ions will be present at some other location further from the object.

The area with a surplus of counter-ions in an electrolyte in contact with a charged object is often referred to as an electrical double layer. Two distinct regions will be formed in this area, thus the name double layer. The two layers are often referred to as the Stern layer (adsorbed ions) and the diffusive layer (mobile ions). The Stern layer is usually several orders of magnitude thinner than the diffusive layer and is therefore seldom considered when it comes to modelling [?].

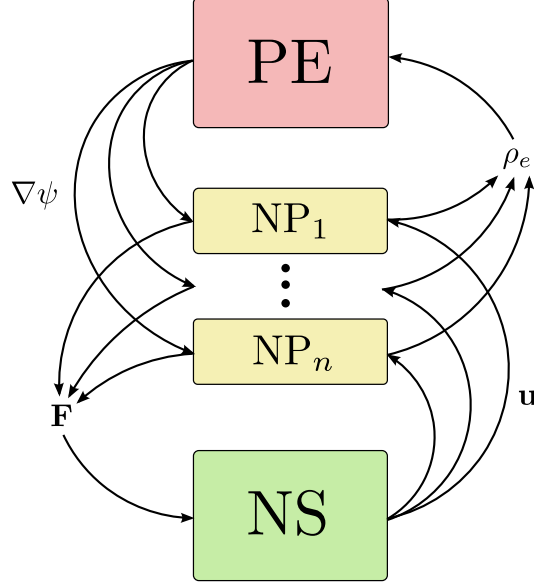


Figure 2.1: Visualisation of the coupling between the three equations present in the model. Poisson's equation (PE), The set of Nernst-Planck equations ($\text{NP}_1 \dots \text{NP}_n$) and the Navier-Stokes equations (NS). The dependencies have also be marked with arrows indicating what quantities for a certain equation that are needed from an other.

2.1.2 Electroosmosis

As fluid carrying a net charge, e.g. in the diffusive layer of an EDL, is under influence of an electric field, the charged particles will move due to the electric forces. As the charge particles move, they will affect the surrounding liquid, causing it move as well. This liquid motion is often referred to as electroosmotic flow. [?]

2.2 Complete physical model

To model the fluid motion of a charged fluid under influences of electrostatic forces, a coupling between different models is considered.

The electric field and potential in the system are obtained from solving Poisson's equation for electrostatics (section 2.3) with a given charge density. This Charge density is obtained from the Nernst-Planck equation (section 2.4) by including effects on the charge distribution from the electric field previously mentioned, diffusion and advection. The advective charge flux is given from the velocity field in the fluid that is obtained by solving the Navier-Stokes equations (section 2.5). Forces due to present electric fields on net charged areas of the fluid also couples the NS equations to the NP equation. More about the force coupling is discussed in sections. 2.6 and 2.7. The coupling between the different equations are visualised in fig. 2.1.

2.3 The potential - Poisson's equation

To be able to model the flow dynamics of liquids in a channel with present EDLs, the potential and charge distribution in the channel must be determined. These quantities are mutually related through Poisson's equation for electrostatics:

$$\nabla^2 \psi = -\frac{\rho_e}{\epsilon_r \epsilon_0} \quad (2.1)$$

where ψ is the electrical potential, ρ_e the electrical charge density, ϵ_r is the relative permittivity and ϵ_0 the vacuum permittivity. Under certain assumptions, the charge density may be explicitly determined as a function of the potential distribution, one such result is the so called Poisson-Boltzmann equation, further discussed in section 2.4.2.

2.3.1 Boundary conditions

At the charged boundaries, most physical situations may be covered by either specifying the potential or the surface charge density. The former would be a boundary condition of Dirichlet type:

$$\psi(\mathbf{x}) = \zeta(\mathbf{x}), \quad \mathbf{x} \in \Gamma \quad (2.2)$$

and the latter a boundary condition of Neumann type:

$$\nabla \psi(\mathbf{x}) \cdot \mathbf{n} = -\frac{\sigma(\mathbf{x})}{\epsilon_0 \epsilon_r}, \quad \mathbf{x} \in \Gamma \quad (2.3)$$

where Γ denotes the boundary of the domain and \mathbf{n} is the normal to the boundary surface. [?]

2.4 The transport of charges - Nernst-Planck equation

The charge concentration in an electrolyte is indeed affected by its environment. In the model proposed here, influences from: advection of the electrolyte, diffusion due to concentration gradients and effects from the electric field originating from charged objects placed at the border or in the flow is considered. Charge conservation without any external sources of the ion density, $C(\mathbf{x}, t)$, gives:

$$\frac{\partial C}{\partial t} + \nabla \cdot \mathbf{J} = 0 \quad (2.4)$$

where $\mathbf{J}(\mathbf{x}, t)$ is the net flux induced by the effects described above. Explicit expressions for the fluxes due to advection and diffusion respectively are

$$\mathbf{J}_{adv} = C\mathbf{u} \quad (2.5)$$

and

$$\mathbf{J}_{dif} = -D\nabla C \quad (2.6)$$

where \mathbf{u} is the advective velocity and D is a diffusion coefficient. The ionic flux due to the presence of an electric potential, $\psi(\mathbf{x}, t)$, is given by the Nernst equation [?]:

$$\mathbf{J}_{ele} = -\frac{zq_e D}{k_B T} C \nabla \psi \quad (2.7)$$

where z is the relative charge of an ion, q_e is the fundamental charge, k_B is the Boltzmann constant and T is the temperature of the fluid.

Summing up the fluxes and putting them into eq. (2.4) gives

$$\frac{\partial C}{\partial t} = \nabla \cdot \left[D\nabla C - C\mathbf{u} + \frac{zq_e D}{k_B T} C \nabla \psi \right] \quad (2.8)$$

which is a known result often referred to as the Nernst-Planck equation. The advective velocity, \mathbf{u} , and the potential gradient, $\nabla \psi$, are obtained from couplings to the Navier-Stokes and Poisson's equation respectively. More about the coupling between the equations is discussed in section 2.2.

2.4.1 Boundary conditions

Depending on the physical situation that is being modelled, different conditions may be imposed at the boundaries of the domain. Throughout this work, at hard boundaries (walls), the charge flux through the boundary is set to zero, i.e.:

$$\mathbf{J} \cdot \mathbf{n} = 0, \quad \mathbf{x} \in \Gamma \quad (2.9)$$

where \mathbf{n} denotes the normal to the surface and Γ is the boundary of the domain.

2.4.2 Poisson-Boltzmann equation

Consider a system consisting of an electrolyte in contact with a (flat) charged wall. Under certain assumptions, it is possible to explicitly determine the charge density in eq. (2.8) as a function of the electric potential. E.g. if there is no advection present and if the system has reached a steady state, i.e. $\partial C / \partial t = 0$ and $\mathbf{u} = \mathbf{0}$ we have:

$$D\nabla C + \frac{zq_e D}{k_B T} C \nabla \psi = \alpha \quad (2.10)$$

where α is some arbitrary constant. Due to the steady state assumption, what the equation above actually says is that the net flux of charge in the system is constant. Since no flux of charge is wanted to flow through the wall boundary, the flux is set to zero on the wall and since the flux is constant it will therefore be zero everywhere in the liquid, i.e. $\alpha = 0$.

Considering only a one-dimensional situation with a position variable y varying in a direction out from the wall into the liquid, eq. (2.10) reads

$$\frac{1}{C} \frac{dC}{dy} + \frac{zq_e}{k_B T} \frac{d\psi}{dy} = 0. \quad (2.11)$$

The charge density is determined by solving eq. (2.11) for C , i.e. integrating the equation. In order to avoid introducing additional unknown quantities, the equation is integrated to far away from the wall where the potential from the EDL is assumed to have decreased to zero and where the concentrations, C^∞ , of the electrolyte is known.

$$\int_y^\infty d \ln(C(y')) = -\frac{zq_e}{k_B T} \int_y^\infty d\psi(y') \quad (2.12)$$

This gives an expression for $C(y)$:

$$C(y) = C^\infty \exp \left(-\frac{zq_e \psi(y)}{k_B T} \right). \quad (2.13)$$

In a general case, there may be several species of ions in the electrolyte, the net charge density, ρ_e , is then given by simply summing up the contributions from the different species:

$$\rho_e = q_e \sum_i z_i C_i. \quad (2.14)$$

Summarising eqs. (2.1), (2.13) and (2.14) gives the Poisson-Boltzmann equation in one dimension

$$\frac{d^2 \psi(y)}{dy^2} = -\frac{q_e}{\epsilon_r \epsilon_0} \sum_i z_i C_i^\infty \exp \left(-\frac{z_i q_e \psi(y)}{k_B T} \right). \quad (2.15)$$

The Debye–Hückel approximation

Historically, the non-linear nature of eq. (2.15) complicated when it came to solving it. This was a major difficulty in the past when the computational power at hands were rather limited. A linearisation is therefore sometimes done, this linear version of the PB equation is often referred to as the Debye–Hückel approximation. The solution of the linearisation gives however, something to compare with and will be used when defining a characteristic length scale of the EDL.

For a 1:1 electrolyte solution, eq. (2.15) reduces to

$$\frac{d^2 \psi(x)}{dx^2} = \frac{2n^\infty q_e z}{\epsilon_r \epsilon_0} \sinh \left(\frac{zq_e \psi(x)}{k_B T} \right). \quad (2.16)$$

and the linearised equation is

$$\frac{d^2 \psi(x)}{dx^2} = \frac{2n^\infty q_e^2 z^2}{\epsilon_r \epsilon_0 k_B T} \psi(x) = \kappa^2 \psi(x) \quad (2.17)$$

where κ^{-1} is the Debye length and gives a measure for the characteristic size of the EDL.

Limitations of the Poisson-Boltzmann model

As the Poisson-Boltzmann equation is derived several assumptions are made. First, the net flux of ions are assumed to be zero and that the advective contribution the flux is negligible. Thus, the PB equation is only applicable when the system is at (or very close) thermodynamical equilibrium.

TODO: Continue this discussion...

2.5 The velocity field - Navier-Stokes equations

In hydrodynamics, the Navier-Stokes equations are one of the most fundamental corner stones. They describe the motion of a fluid under the influence of various internal and external forces.

For later convenience and for reference when it comes to deriving the Lattice-Boltzmann formulation of the NS equation, a brief sketch of a derivation will here be presented. A most general form of the Navier-Stokes equation follows from momentum conservation

$$\frac{\partial(\rho \mathbf{u})}{\partial t} + \nabla \cdot (\rho \mathbf{u} \otimes \mathbf{u}) + \mathbf{Q} = 0 \quad (2.18)$$

where, ρ is fluid density, \mathbf{u} is velocity and \mathbf{Q} is a momentum source term (force per volume). Expanding the time derivative and the divergence terms respectively gives

$$\mathbf{u} \left(\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{u}) \right) + \rho \left(\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} \right) + \mathbf{Q} = 0. \quad (2.19)$$

By assuring mass conservation (without sources) we have that

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{u}) = 0 \quad (2.20)$$

and eq. (2.19) reduces to

$$\rho \left(\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} \right) + \mathbf{Q} = 0 \quad (2.21)$$

which together with eq. (2.20) is a general formulation of the Navier stokes equations.

The force term \mathbf{Q} , is determined by the physical properties of the fluid and from its environment. In this work, only incompressible Newtonian fluids will be studied. The force contribution to \mathbf{Q} involved in that case is limited to viscous forces, pressure gradients in the fluid and to external force fields. Putting that into eqs. (2.20) and (2.21) gives

$$\nabla \cdot \mathbf{u} = 0 \quad (2.22)$$

and

$$\rho \left(\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} \right) = -\nabla P + \mu \nabla^2 \mathbf{u} + \mathbf{F} \quad (2.23)$$

where P is the pressure, μ the kinematic viscosity and \mathbf{F} is the contributions from external forces.

2.5.1 Boundary conditions

At hard boundaries (walls), the boundary conditions to eqs. (2.22) and (2.23) are set on the velocity of either a Dirichlet or Neumann type. In most physical situations the Dirichlet condition is used which corresponds to that there is a friction between the fluid and the wall, usually full friction, i.e. when no relative movement between fluid and wall is present and the velocity at the wall boundary is set to zero, i.e.

$$\mathbf{u} = 0, \quad \mathbf{x} \in \Gamma \quad (2.24)$$

where Γ denotes the boundary. The Neumann type conditions are used for no-friction walls where the normal component of the derivative of the velocity is specified, usually to zero.

At wet boundaries, inlets and outlets, of the domain various boundary conditions may be set. For instance the pressure or the velocity could be fixed. In the case of a fixed pressure boundary, a flow direction must also be specified for completeness. [?]]

2.6 Pressure-driven electrokinetic flow

As a charged fluid is driven by a pressure gradient, a movement of charges, i.e. an electrical current will be induced. Due to the charge flux, a potential gradient will build up along the flow direction. This potential is usually referred to as the streaming potential, $\phi(\mathbf{x})$, and its magnitude is determined from the induced current through Ohm's law

$$\mathbf{J} = -\sigma \nabla \phi \quad (2.25)$$

where σ is the conductivity of the fluid. For a perfectly conducting fluid, no potential difference will be built up. Also a complete neutral solution will carry no net current and therefore will no potential difference build up in that case either.

Charges under the influence of an electric field will be affected by a force. Charges moving due to this force will, in a liquid, also pull liquid (uncharged) molecules with them. In a macroscopic limit, the force density affecting the charges in the liquid is assumed to affect the liquid as whole. The volumetric force affecting the fluid from the presence of the streaming potential is then given by:

$$\mathbf{F} = \rho_e \nabla \phi \quad (2.26)$$

where ρ_e is the charge density. This is an example of how the charge density from the Nernst-Planck equation may couple to the force term in Navier-Stokes equations.

This force will always be affecting the fluid in an direction opposite of that of the net flux of charge, i.e. the force will slow the fluid down, this is illustrated in fig. ???. This effect that a moving net charged fluid is slowed down is called the *electroviscous effect*. The name originates from that a similar effect might be achieved by increasing the viscosity of the fluid.

TODO: bild ion channel!!

2.7 Electroosmotic flow

Instead of driving the fluid flow through a pressure drop, a net charged fluid may be driven by an external electric field. This may be seen as the opposite case to that in section 2.6 where a current is induced by a pressure drop.

The volumetric force on the fluid from the external field, \mathbf{E}_{ext} , is given by

$$\mathbf{F} = -\rho_e \mathbf{E}_{ext} \quad (2.27)$$

where ρ_e is the charge density. If the electric field is constant (or at least has the same direction) everywhere, the sign of the force is not in the same direction for a net charged positive area of the fluid as for a net charged negative. Thus the fluid may be either slowed down or sped up. This is a qualitative difference to pressure driven situation and is illustrated in fig. ???.

The electroviscous effect is in the case of electroosmotic flow usually neglected as the field due to the streaming potential is, in most physical cases, small to the applied external field. [?]

TODO: bild ion channel!

3

The lattice-Boltzmann method

short intro of this section

3.1 Historical overview

A few words on the history of the method. Lattice automata etc.

3.2 Maybe something on asymptotic analysis

Some theory that could be useful in the chapman-enskog derivations.

3.3 Basic idea

The principle behind the method, what it does and does not.

3.4 Collision operator

discussions on different col. operators, focus on BGK since that is the one used.

3.5 Streaming

maybe not so much to say here

3.6 Boundary conditions

discussion and description of the boundary conditions

3.6.1 bounce back

accuracy, e.g. second order accurate if placed between node planes...

3.6.2 slip

3.6.3 he-zou, constant density/velocity

3.6.4 Maybe something on non-local boundary conditions

3.7 Forcing schemes

how to add a "forcing" term in the method.

3.8 LBM for Navier-Stokes

3.8.1 Chapman-Enskog

3.9 LBM for Poisson's equation

3.9.1 Chapman-Enskog

3.10 LBM for Nernst-Planck

3.10.1 Chapman-Enskog

3.11 Algorithm/Scheme for solving the coupled equations

the iterative scheme used.

4

A few notes on high performance computing

intro what is said below refer to computers of a certain architecture...

4.1 The pipeline

keep it full.

4.2 Locality

using the caches in a good way. some examples of its importance.

4.2.1 Locality and LBM

4.3 Parallelisation

shared memory, distributed memory...

data dependence, LBM good!

OpenMP/MPI

4.4 Maybe something about profiling

men kanske inte tillför något vettigt.

4.5 Choice of programming language

4.6 Some stats on the performance of the code...

Lattice updates/s

5

Model benchmarks

intro 2D

5.1 Poiseuille flow

Navier-Stokes density + velocity profiles.

5.2 Taylor-Green vortex

5.3 Helmholtz equation

Poisson's eq.

5.4 Advection-Diffusion

Before the implementation of the Nernst-Planck part of the model is tested, a special case is considered, i.e. when the electrical potential in the domain is constant. This makes the source term including the electrical potential in eq. (??) vanish and we have to solve only for advection and diffusion.

Introducing characteristic scales for the concentration (C_0), advective velocity (u_0) and length (l_0) respectively, gives the non-dimensional advection-diffusion equation for incompressible flow:

$$\frac{\partial C}{\partial t} + \mathbf{u} \cdot \nabla C = \frac{D}{u_0 l_0} \nabla^2 C. \quad (5.1)$$

All variables in (5.1) are non-dimensional. The quantity $Pe = u_0 l_0 / D$ is often referred to as the Péclet number. It determines the relation between contributions to

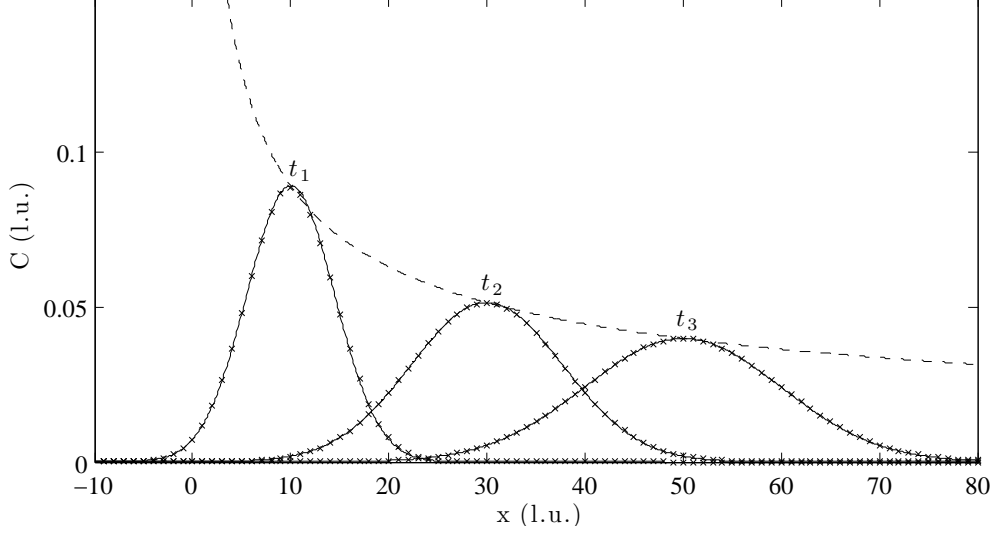


Figure 5.1: Obtained solutions (\times) of the advection-diffusion equation for a point mass evolving in time and space. Three different times ($t_n = 100n$) are compared to analytical solutions (solid). The Amplitude of the solutions as function of time has also been plotted (dashed). The advecting velocity, $u_0 = 0.1$ and the Peclet number, $Pe = 10$. All units are in lattice units.

the dynamics from advection and diffusion respectively. For $Pe \gg 1$ the dynamics is dominated by advection and for $Pe \ll 1$ by diffusion.

The LB model described in section ?? was tested by studying the evolution in time and space of a point mass in one dimension. The analytical solution of eq. (5.1) in one dimension with initial conditions $C(x, t = 0) = \delta(x)$ on an infinite domain is:

$$C(x, t) = \sqrt{\frac{Pe}{4\pi t}} \exp\left(-\frac{(x - ut)^2 Pe}{4t}\right). \quad (5.2)$$

In the numerical computations the parameters $Pe = 10$ and $|\mathbf{u}| = 0.1$ were used. The domain consisted of 200 lattice nodes and three snapshots in time at $t = 100, 200, 300$ were compared to the analytical solution. The result is presented in fig. 5.4.

5.5 Nernst-Planck, a special case

6

Modelling of electrokinetic flow

6.1 Electric potential in 2D channel - PB

section in the channel, + debye-huckel comparison

6.1.1 It might be interesting to compare the two models Chai and Wang

6.2 Compare with Nernst-Planck

6.2.1 Potential

6.2.2 Charge distribution

6.3 Electroviscous effect

NP + PB differences?

6.4 Flow in array of charged squares

maybe other geometries as well?

6.5 3D?

only god knows...

7

Conclusions

And what do we conclude of this?