Developing methods for mathematical modeling of a two-phase dielectric-electrolyte microsystem

E V Gorbacheva and E N Kalaidin
Kuban State University, st. Stavropolskaya, 149, 350040, Krasnodar, Russia
E-mail: katya1911@list.ru

Abstract. In this paper, we propose a numerical solution to the problem of stability of a two-phase dielectric/electrolyte system under direct and alternating electric fields. The lower wall adjacent to the electrolyte is assumed to be a charged surface, while the upper one is electrically insulated. The charge on the lower surface is supposed to be stationary, and the surface charge on the free interface between liquids is assumed to be mobile. The model is described by a system of Nernst-Planck-Poisson-Stokes equations. The mathematical model is closed by the corresponding boundary conditions. The linear stability of the one-dimensional flow is investigated. At a constant electric field, and the presence of two types of instabilities is found: short-wave and long-wave.

1. Introduction
Due to the rapid development of micro-/nano-fluid systems, a variety of electroosmosis phenomena, especially off-scale, have attracted a lot of attention in the academic sector. Electrical double layer and electroosmotic flows play an important role in functional analysis and optimal design of micro-/nano-fluidic chips.

However, the behavior of liquids in microsystems cannot be considered as fully studied; it can differ significantly from macroscopic flows, since it is characterized by a different ratio of viscous, surface and inertial forces. In microchannels, the complexity of description increases greatly due to the significant role of the interaction of the liquid with the wall. In this regard, the development of an effective and reliable numerical technique for modeling two-phase flows in microchannels is considered an urgent task.

Until now, the research of stability and bifurcations of flows has been a complicated problem. An algorithm of direct numerical simulation of flows frequently gives results close to experimental ones. Even though the calculations are quite complex, the results are not always correct. In most cases, the method of direct numerical modeling complements the analysis of the flow stability, which is often useful for studying transient processes in flows.

Prolonged research [1, 2] has shown that the best solution to problems of stability of viscous fluid flows is obtained numerically. When it is necessary to solve stability problems, numerical methods of a high order of accuracy are frequently used. Low-order accuracy methods can show good results only in the one-dimensional case, while a refined computational grid is applied to achieve good accuracy.

2. Problem formulation
A two-phase micro-/nano-flow of conductive and non-conductive viscous liquids is considered. The fluid flow is confined to two solid walls in an external electric field. The charged boundaries are
supposed to be the electrolyte/dielectric, solid-electrolyte interfaces, while the solid-electric interface is an electrically isolated surface. The charge in the vicinity of a solid is stationary, while the surface charge at the interface between the two liquids is mobile.

The movement of an electrolyte under the action of an electric field is described by a coupled system of Nernst-Planck-Poisson-Stokes equations.

\[
\begin{align*}
\frac{\partial C^\pm}{\partial t} + U \cdot \nabla C^\pm &= \pm \nabla \cdot (C^\pm \nabla \Phi) + \nabla^2 C^\pm \\
\nu^2 \nabla^2 \Phi &= C^- - C^+ \\
\text{Re} \frac{\partial U}{\partial t} + \nabla P &= \nabla^2 U - \frac{\kappa}{\nu^2} (C^+ - C^-) \nabla \Phi, \\
\nabla \cdot U &= 0,
\end{align*}
\]

where:
- \( C^\pm \) is the concentration of cations and anions;
- \( U \) is the vector of fluid velocity;
- \( \Phi \) is the electric potential;
- \( P \) is the pressure in the electrolyte;
- \( \kappa \) is the coupling coefficient between the hydrodynamics and the electrostatics;
- \( \nu \) is the Debye number;
- \( \text{Re} = \rho_e D / \mu_e \) is the Reynolds number, here \( D \) is diffusion coefficient of cations and anions, \( \rho_e \) is the density of the electrolyte, \( \mu_e \) is the viscosity of the electrolyte.

The dielectric fluid is electrically neutral, and is described by the Laplace equation, and the hydrodynamics of the problem is described by the Stokes equation.

\[
\nabla^2 \varphi = 0,
\]

\[
r \text{Re} \frac{\partial u}{\partial t} + \nabla p = \mu \nabla^2 u, \\
\nabla \cdot u = 0,
\]

where:
- \( \varphi \) is the electrical potential (in dielectric phase);
- \( u = \{u, v\} \) is the dielectric velocity vector;
- \( p \) is the pressure in the dielectric;
- \( \rho_d \) is the density of the dielectric;
- \( r = \rho_d / \rho_e \) is the ratio of the densities;
- \( \mu_d \) is the viscosity of the dielectric;
- \( \mu = \mu_d / \mu_e \) is the ratio of the viscosities.

The boundary conditions are set as follows:
1) the surface is impermeable to positive and negative ions;
2) the electric potential is continuous on the surface, while its normal to the derivative of the surface has a jump associated with the mobile charge of the surface;
3) the surface charge is constant;
4) dynamic conditions for normal and shear stresses;
5) kinematic condition.

For the convenience of the solution, a non-orthogonal coordinate system is introduced:

\[
\tau = t, \quad \xi = x, \quad \eta = y/h(x, t).
\]

3. Basic simulation

Analyzing the resulting system of equations, we can see that it is homogeneous with respect to the space coordinate \( \xi \), and therefore, the electrostatic part of the problem may be solved separately from the hydrodynamic one. Consequently, the concentration of cations and anions just as the normal electric field, is independent of time \( \tau \) and external electric field \( E_{\infty} \).

The electric potential in the system can be represented as the sum of the potentials of the space charge fields \( \Phi_n (\eta) \) and the tangential electric field \( \Phi_t (\xi) \) applied from the outside:
\[ \Phi = \Phi_1(\xi) + \Phi_2(\eta). \]

Equation (1) is converted into curvilinear coordinates and integrated over \( \eta \); and the integration constants are found from the boundary condition. Equation (2) supplements the system of ordinary differential equations for concentrations and normal electric field:

\[
\begin{align*}
C^+ E + \frac{dC^+}{d\eta} &= 0, \\
-\mathcal{C}_d - E + \frac{d\mathcal{C}_d}{d\eta} &= 0
\end{align*}
\]

(3)

Boundary conditions are:

\[ \eta = 0: E = -q/v, \quad \eta = 1: E = \sigma_0/v \]

(5)

where:

- \( E \equiv \frac{\partial \Phi}{\partial \eta} \) is the normal electric field strength;
- \( q \) is the dimensionless surface charge on a solid surface;
- \( \sigma_0 \) is the dimensionless surface charge.

The one-dimensional basic stationary problem (3)(5) was solved using the Galerkin quasi-spectral method of \( \tau \)-modification. When solving, Chebyshev polynomials were used, which are convenient for approximating any of the unknown functions.

After solving the electrostatic part of the problem, our system of equations (the hydrodynamic part of the problem) will be as follows:

Solution for

\[
\text{Re} \frac{\partial U}{\partial \tau} = \frac{\partial^2 U}{\partial \eta^2} - \kappa \frac{dE}{d\eta} E_{\infty}, \quad r \text{Re} \frac{\partial u}{\partial \tau} = \mu \frac{\partial^2 u}{\partial \eta^2}.
\]

Boundary conditions are:

\[ \eta = H: u = 0, \quad \eta = 1: \frac{\partial u}{\partial \eta} = \frac{\partial U}{\partial \eta} - \kappa E_{\infty}, \quad u = U, \quad \eta = 0: U = 0. \]

The solution for \( 0 < \omega < 2 \) (Re = 0) has the following form:

\[ U = \kappa E_{\infty} \left( \int_0^\eta E d\eta + \mu \int_0^\eta E d\eta \right). \]

This equation is valid either for the case of direct current, when \( E_{\infty} = \text{const} \), or for the case of alternating current, when \( E_{\infty} = E_0 \cos \omega t \) (\( \omega \) is the frequency of the external field).

4. **Numerical simulation**

To study the linear stability, small periodic disturbances along the \( \xi \) axis are imposed on the one-dimensional stationary solution.

\[
\begin{align*}
C(\tau, \xi, \eta) &= C^0(\tau, \eta) + \hat{C}^0(\tau, \eta)e^{i\omega \xi}, \\
\Phi(\tau, \xi, \eta) &= \Phi_0(\tau, \eta) + \hat{\Phi}(\tau, \eta)e^{i\omega \xi}, \\
U(\tau, \xi, \eta) &= U_0(\tau, \eta) + \hat{U}(\tau, \eta)e^{i\omega \xi}, \\
V(\tau, \xi, \eta) &= V_0(\tau, \eta) + \hat{V}(\tau, \eta)e^{i\omega \xi}, \\
\psi(\tau, \xi, \eta) &= \psi_0(\tau, \eta) + \hat{\psi}(\tau, \eta)e^{i\omega \xi}, \\
v(\tau, \xi, \eta) &= v_0(\tau, \eta) + \hat{v}(\tau, \eta)e^{i\omega \xi}.
\end{align*}
\]

Disturbances superimposed on \( \sigma \) and \( h \):

\[
\sigma(\tau, \xi) = \sigma_0 + \hat{\sigma}(\tau)e^{i\omega \xi}, \quad h(\tau, \xi) = 1 + \hat{h}(\tau)e^{i\omega \xi}.
\]

Studying the resulting system, we can see that it is time periodic \( \tau \) (the period is \( 2\pi / \omega \)), so the system can be represented by an operator:

\[
\frac{\partial f}{\partial \tau} + A(f) = 0 \quad \text{or} \quad \frac{\partial f}{\partial \tau} + A_0(f) + iE_{\infty}A_1(f) = 0.
\]

According to Floquet's theorem, disturbances are found in the following form:

\[ \hat{f}(\tau, \eta) = \sum_{k=-\infty}^{\infty} \hat{f}_k(\eta)e^{(\lambda + ik\omega)\tau}, \]

where \( \lambda \) is the unknown Floquet factor.
The resulting expression is substituted into the system and takes \( E_\infty = E_0 \cos \omega t \) (where \( E_0 \) is the external field). The discretization of the problem in space was carried out by the Chebyshev polynomials:

\[
f_k = \sum_{n=0}^{\infty} f_k^{(n)} T_n(z), \quad z = 2\eta - 1.
\]

To find a nontrivial solution, the problem was reduced to the algebraic eigenvalue problem of dimension \((2M + 1)N\). To solve the generalized matrix eigenvalue problem, the QR algorithm was used.

In the case of a large matrix, the algorithm program may fail. There are two ways to solve this problem:

1) Generalization of the eigenvalue problem.
2) Inversion of one of the equations and substituting it into other equation.

In our case, both methods were used.

5. Results and conclusions

The typical curves of ultimate stability at constant and alternating electric fields are shown in figure 1 for the same amplitude \( E_0 \) and applied frequency \( \omega = 1 \). These are curves for which \( \lambda_R = \text{Re}(\lambda) = 0 \).

Instability zones, in which at least one of their values has a positive real part \( \lambda_R > 0 \), are shaded on the graph.

![Figure 1. Ultimate stability curves for \( \mu = 1 \).](image)

Solid line – case of alternating current (AC) with \( \omega = 1 \), dashed line – case of direct current (DC).

Based on the calculations obtained, we can conclude that:

1) with a growth in the frequency of the electric field \( \omega \), and the one-dimensional solution is stabilized, and the limiting curve shifts in the direction of an increase in the number \( E_0 \) (in the case of a variable electric field);

2) with a growth in viscosity \( \mu \), the stationary flow becomes less stable due to long-wave disturbances and more stable due to short-wave disturbances;

3) a growth in the height of the channel \( H \) causes the flow stabilization for the two types of disturbances (in the case of a variable electric field);

4) for the case of a constant electric field, only 2 eigenvalues have positive real numbers, one corresponds to long-wave instability, and the other to the short-wave one. It was emphasized in [4] that the long-wavelength instability is due to disturbances of the surface charge at the liquid interface, whereas the short-wave instability is due to disturbances of the conductivity of the electrolyte. For an alternating electric field, these 2 regions of instability also exist;

5) the speed of a one-dimensional electrolyte is directly proportional to \( \kappa \). As \( \kappa \) increases, the flow becomes faster and less stable.
6) the dependence of the resulting solution on the Weber number is essentially the same as for the air/electrolyte case; therefore, all the details of this behavior can be found in [3, 4]. In particular, it has been found that at infinitely large $\text{We}$, the short-wavelength instability is preserved, while the long-wavelength one disappears (see figure 2). This kind of behavior indicates that short-wave instability is due to volume disturbance, since the interfacial effects are the least important when the $\text{We}$ value is large enough;

7) in the case of alternating current, the curves of the limiting stability for other parameters show qualitatively similar results as for the electric field of direct current [3].

![Figure 2. Marginal stability curves for $\mu = 1, w = 1, H = 4$ and different values of $\text{We}$. 1: $\text{We} = 200$; 2: $\text{We} = 2 \times 10^4$; 3: $\text{We} = 2 \times 10^6$.](image)

Analyzing the results obtained, we can conclude that in practice it is more convenient to use short-wave instability, since it can occur in rather short channels, and it is also more intense than long-wavelength. These data can form the foundation for a new type of non-mechanical micromixers for emulsions.

References

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