

# The structure of the interelectrode layers in low-conductive liquid flows in microchannels with a ion source affected by an applied field

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## Abstract

The interest in electrohydrodynamics of flows in micro- and nano-size channels, which has been observed in recent years in the literature, is caused by the need to manage the motion of ultrasmall portions of fluids in various devices employed in biological research, biotechnology, pharmacy, medicine, and other areas of science. This work is focused on the study of the influence of strong non-uniform electric fields on the ionization processes in partially ionized liquid mixtures as they flow through plane microchannels. The effects caused by the action of a non-uniform field on the bulk electrochemical kinetics in low-conductive liquids at moderate values of the Debye number are analyzed. In weakly conducting chemically reacting environments the applied electric field can affect the ionization process both indirectly, through redistribution of reactive charged components, and directly, by means of increasing the kinetic rate of ionization. In the last case, in the near-wall layers with a strong non-uniformity of the field, bipolar structures of uncompensated space charge are formed, which can be affected by the applied longitudinal field. The presence of the layers with the opposite Coulomb forces brings about inflection points in the velocity profile. This may cause instability of the flow and thereby induce the mixing of the liquid in the microchannel.

## 1 Description of the fluid model

We consider a two-dimensional nonstationary flow of a multicomponent liquid containing charged particles of two kinds: positively and negatively charged. Diffusion and drift of the particles of each kind of charge in the electric field, bulk reactions of ionization and recombination, and surface electrochemical processes that cause generation and absorption of charged particles are taken into account. The motion of the liquid as a whole is described by the conventional equations of conservation of momentum for viscous medium with due regard for the bulk Coulomb force, and the continuity equation for an incompressible fluid. The laws of conservation of mass

for the charged components and Poisson's equations for the field have the form [1]:

$$\begin{aligned} \frac{\partial n_i}{\partial t} + \operatorname{div} n_i \vec{U}_i &= w(E) - an_1 n_2, \quad i = 1, 2, \\ n_i \vec{U}_i &= n_i \vec{u} + \operatorname{sign}(e_i) n_i b_i \vec{E} - D_i \nabla n_i, \quad e_1 > 0, \quad e_2 < 0, \\ \varepsilon \operatorname{div} \vec{E} &= 4\pi q, \quad \vec{E} = -\nabla F. \end{aligned} \quad (1)$$

The boundary conditions for Equations (1) represent the balance between the ion fluxes from the bulk to the surface and the fluxes of the particles generated at the surface [2]:

$$n_i \vec{U}_i \cdot \vec{v} = A_i^k(E, n_m, \dots) - K_i^k n_i. \quad (2)$$

Here  $v$  is the external normal to the interface,  $A_i^k$ ,  $K_i^k$  are the effective parameters of the surface electrochemical processes, which depend on the properties of the  $k$ th surface of the channel walls, liquid content, etc. In formulae (1), (2) the quantities  $\vec{u}$ ,  $n_i$ ,  $U_i$ ,  $D_i$ ,  $b_i$ ,  $e_i$ ,  $q$ ,  $E$ ,  $F$  are, respectively, the velocity of the liquid as a whole, concentration, velocity, coefficients of diffusion and mobility, and the charge of the particles of  $i$ th kind, the volume charge density, the strength and the potential of the electric field,  $\varepsilon$  is the dielectric permeability,  $w$  is the bulk ionization rate (the rate of dissociation of the neutral molecules of the liquid into positive and negative ions), which may depend on the field strength, and  $a$  is the coefficient of the bulk recombination of ions.

## 2 Results

Let us consider the results of the calculation of a steady flow through a plane channel under the conditions that an electrical potential difference is applied to the channel walls from an external source and the channel walls exhibit different surface characteristics. Suppose that at one of the walls the surface reaction for the positive ions  $n_1$  is in equilibrium with the corresponding value of their concentration, and for the negative ions  $n_2$  this wall is chemically neutral (zero flux of this kind of ions to the wall). The potential of this wall (anode) is positive. The situation at the other wall is supposed to be the opposite: the equilibrium for the negative ions (for the sake of simplicity, with the same value of the equilibrium concentration), and zero flux of the positive ions to the wall. The potential of this wall (cathode) is equal to zero. The dependence of the bulk source of ions on the applied field was studied in [3], [4]. In the calculations presented in this work we use Frenkel's relation [3] ( $w_0$  is the rate of dissociation in the absence of the applied field):

$$w(E) = w_0(T, \dots) \exp(2\gamma E^{0.5}), \quad \gamma = e^{1.5}/(\varepsilon^{0.5} kT). \quad (3)$$

Note that one of the first works devoted to the study of electrohydrodynamic flows through channels with regard to the effect caused by the applied field on the rate of the bulk dissociation within the framework of Onsager's model [4] was carried out by professor Antonio Castellanos [5].

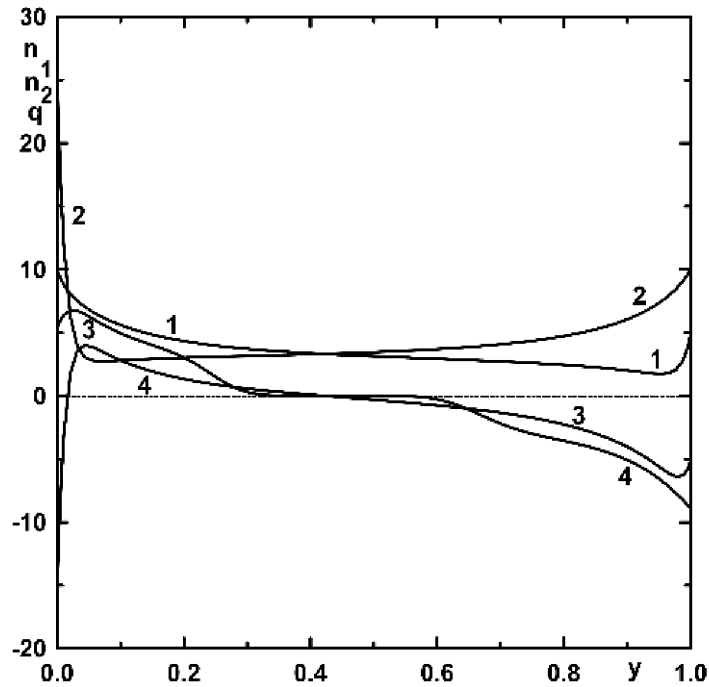


Figure 1: Distribution of the concentrations of ions  $n_1$  (curve 1) and  $n_2$  (curve 2), and density of the volume charge  $q$  with (curve 4) and without regard for the field dependence of the ionization rate ( $\gamma = 0$ , curve 3) in the channel cross-section.

The distributions of the dimensionless concentrations of ions  $n_1$ ,  $n_2$  and the space charge  $q$  in the cross-section of the channel for a given difference of the potentials between the walls are presented in Fig. 1. The concentrations are divided by the initial quasineutral concentration of ions, the coordinate is normalized by the channel width. The values of the equilibrium concentrations of ions specified in the calculations are presented in the figure.

It is seen that even in the case of a plane channel, when the applied uniform field is not distorted by the geometry and is constant across the channel, the presence of a field-dependent source of ions essentially changes the distribution of the space charge near the electrodes. In particular, in the vicinity of the anode ( $y = 0$ ) a bipolar structure arises: the negatively charged near-wall layer is replaced with a layer of positive charge. This is due to the fact that the additional source of ions that arises in a strongly nonuniform field appreciably alters the distribution of their concentrations in the near-wall layers. The distribution of the dimensionless field and potential across the channel is presented in Fig. 2. The field and the potential are divided by the thermal potential and the corresponding field. It is seen that at moderate values of the Debye number, when the Debye length has the order of the characteristic size of the channel, the charged layers occupy a sufficiently large part of the channel cross-section and significantly alter the applied field. In particular, as the volume charge changes its sign in the bipolar layer near the anode, a local

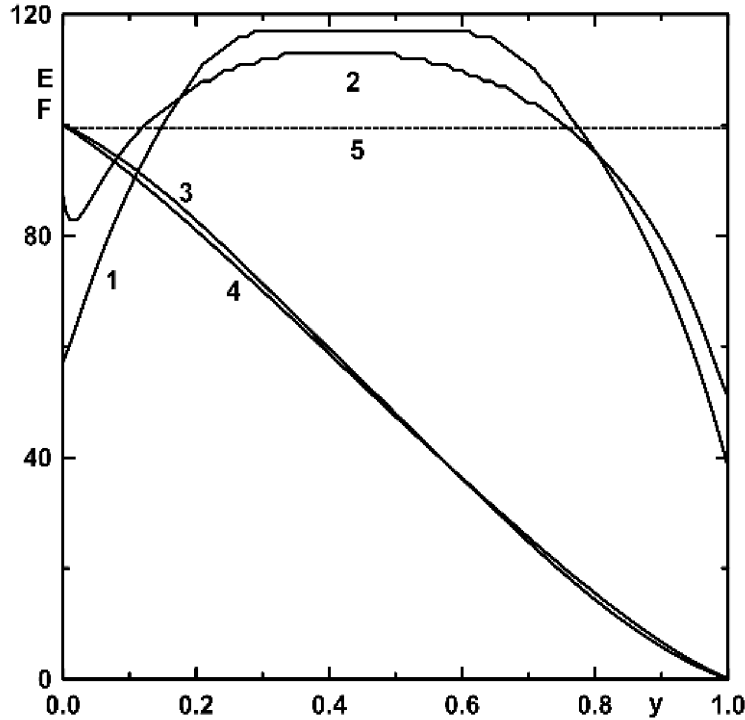


Figure 2: Distribution of the normal components of the field strength  $E$  (curves 1, 2) and potential  $F$  (curves 3, 4) in the channel cross-section. Curves 2, 4 — with regard for the field dependence of the ionization rate, curves 1, 3 — without regard ( $\gamma = 0$ ). Curve 5 shows the applied external field.

extremum of the field strength occurs. Note that a local extremum of the field strength near the surface of the plane electrode caused by the bipolar structure of the near-wall layer was experimentally observed in [6]. In paper [7] in the framework of the tree-ion model a mechanism for the formation of the near-electrode bipolar layer with the field extremum was proposed. The key role was played by the bulk recombination processes which involve the ions injected by the wall. The effect caused by the field on the rate of ionization was disregarded. The results of this work suggest that the formation of the bipolar near-electrode structures may be governed by another mechanism.

Figure 3 shows the distribution of the bulk Coulomb forces  $FK = qE$  affecting the fluid in the applied transverse field. It is seen that in most part of the charged domains the field repels the fluid from the walls. Such a configuration of forces directed towards each other is favourable for the rise of an instability analogous to the Rayleigh–Bénard thermogravitational instability in conventional hydrodynamics [8]. The fact that the bulk source depends on the field aggravates the matter.

As is shown in Fig. 1, under nonsymmetric boundary conditions for the ion concentrations, in the channel cross-section domains with positive and negative space charge are formed, which are adjacent to different walls. If an external electric field is applied along the channel, then the arising longitudinal component of the Coulomb

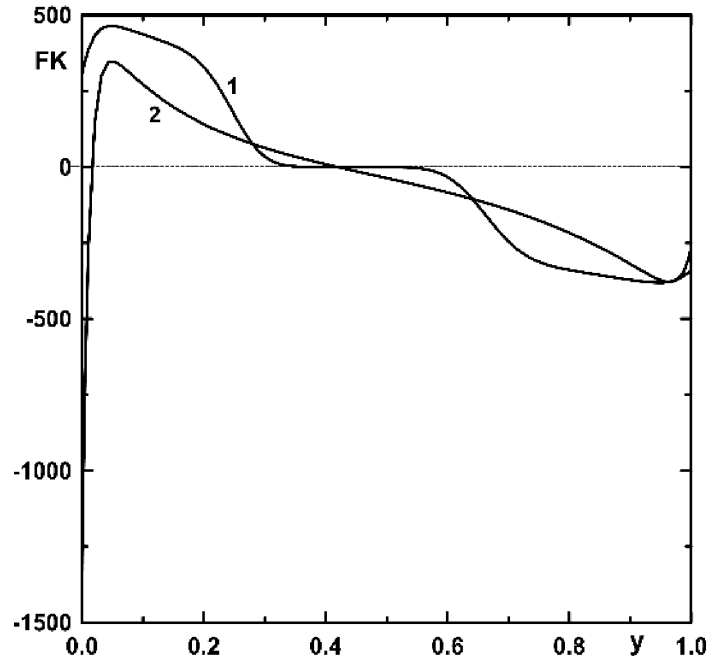


Figure 3: Distribution of the normal component of the Coulomb force  $FK = qE$  affecting the fluid. Curve 2 — with regard for the field dependence of the ionization rate, curve 1 — without regard ( $\gamma = 0$ ).

force is directed in the opposite ways near the opposite channel walls, thereby speeding up and slowing down the flow of the fluid. Figure 4 shows the cross profiles of the fluid velocity calculated with and without regard for these forces.

It is seen that in the domain of the negative space charge the Coulomb force, which slows down the flow, gives rise to a thin layer of the return flow and causes the appearance of inflection points in the velocity profile. This may cause the Kelvin–Helmholtz shear instability of the flow and thereby enhance the mixing of the fluid in the channel [8].

### 3 Conclusions

The study of the structure of the interelectrode space in a plane channel shows that the direct effect caused by the field on the rate of dissociation of the molecules of the fluid into positive and negative ions can significantly redistribute the concentration of these particles in the domains of the strong nonuniform field in the near-wall layers of the plane channel. If the channel walls have different surface electrochemical properties, then the external electric field applied along the channel may cause vigorous mixing of the fluid that flows along the micro-channel.

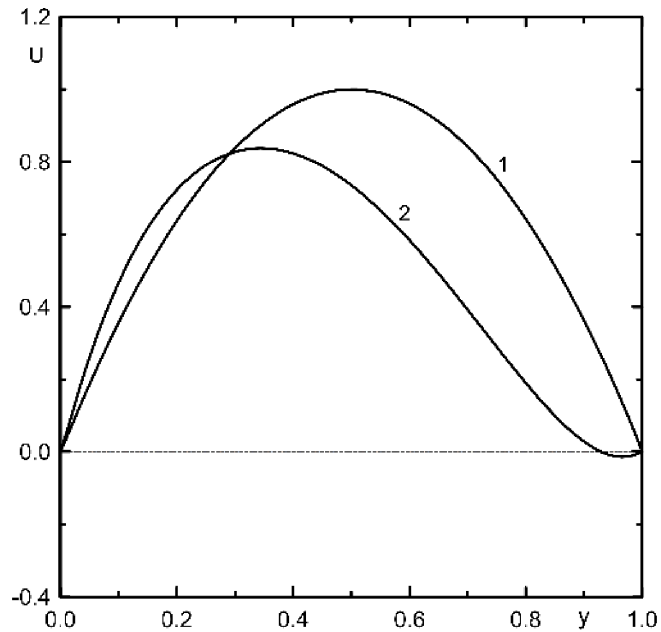


Figure 4: Velocity profiles of the liquid flow in the channel: the Poiseuille profile (curve 1) and the profile that has been transformed under the action of the longitudinal Coulomb forces in the applied longitudinal field (curve 2).

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