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Stability of the electroosmotic flow of a two-layer electrolyte-dielectric system with external pressure gradient[⋆]

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Abstract. The stability of the electroosmotic flow of electrolyte-dielectric viscous liquids under the influence of the DC and AC electric fields along with the external pressure gradient is studied theoretically. Liquids are bounded by two infinite parallel plates. The lower wall bordering the electrolyte is assumed to be a charged surface, and the upper wall is electrically isolated. The charge at the lower boundary is assumed to be immobile, while the surface charge at the free surface is assumed to be mobile. In this paper, we study the micro- and nanosized liquid layers. The mathematical model is described by a nonlinear system of the Nernst-Planck-Poisson-Stokes partial differential equations with the appropriate boundary conditions on the solid surface, the electrolyte/dielectric interface, and on the upper wall. The pressure gradient is highly important for the stability of the flow. For the DC case, the external pressure could either stabilize and destabilize the flow depending on the relative directions of the electroosmotic flow and the pressure-driven flow. For the AC case, the dependence on the value of the external pressure is not monotonous for different wave numbers of perturbations, but, as a rule, the external pressure destabilizes the flow. As the frequency of the electric field increases, the one-dimensional solution of the problem becomes stable.

1 Introduction

In connection with the rapid development of biotechnology, the problems of electrokinetics attracted much attention. One of its applications is medical diagnostics in laboratories on a chip. Electroosmotic flows are an important element in the laboratories on a chip for transport and mixing of liquids. Nevertheless, when the liquid is exposed to a constant high power electric field, various undesirable effects occur, such as sample degradation, electrolysis, bubble formation on electrodes. These effects can be prevented by using an alternating electric field. The two-layer system is used to drive non-conducting liquids at microscale scales so that the electroosmotic and pressure-driven flow of the electrolyte entrains a layer of dielectric fluid. However, even with the use of such techniques, the theoretical aspects of transport and mixing of non-conducting liquids remain insufficiently studied.

Previous works on the AC electroosmotic flows in microchannels are based on the one-phase flow [1] or on the microfilm flow with an unperturbed flat interface [2]. The main limitation of the use of the electroosmotic flows is that the liquid must have a sufficiently high conductivity. Despite the fact that the biological fluids, such as blood, serum, etc., are conductors, their conductivity is not high enough to be moved by the electric field. To solve this practical problem, a two-layer system of immiscible liquids is taken, where one liquid is a non-conducting one while the other is an electrolyte. The non-conducting liquid is entrained by the shear of the neighboring conducting liquid [3]. Studies of the electrostatic effects at the interface were presented in [4,5]. The stability of such flows with the application of a constant field was investigated for a special case, where air was used as a dielectric, in work [6], and for a flow with an additional external pressure gradient in operation [7].

Investigations of the problem with AC electric field are absent in the literature, so the main goal of the present article is to fill in the theoretical gap and to help other researchers with experimental investigations. Critical values of the parameters can serve as a guide for the experiments.

2 Formulation of the problem

A flow of two thin liquid immiscible layers with a constant viscosity and permittivity is considered. The flow is

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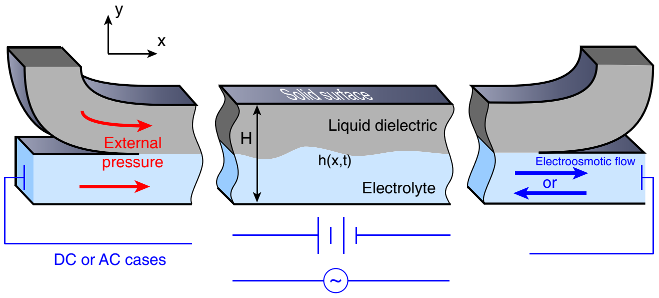

Fig. 1. Electroosmotic two-phase liquid-liquid flow subject to an external tangential electric field.

enclosed between two infinite parallel plates, $y = 0$ and $y = H$, see (fig. 1). The region $0 < y < h(x,t)$ contains a symmetric, binary electrolyte with equal diffusivities of cations and anions, while the region $h(x,t) < y < H$ is filled with a dielectric.

Tilded symbols $(\tilde{f})$ will be used for the dimensional variables, in contrast to their untilded dimensionless counterparts (f). The following characteristic quantities are used as the basic values:

- $\tilde{h}_0$ is the characteristic length, a thickness of the electrolyte layer;
- $\tilde{h}_0^2/\tilde{D}$ is the characteristic time, where $\tilde{D}$ is the ion diffusivity in the electrolyte;
- $\tilde{\mu}_e$ is dynamical viscosity of the electrolyte is taken as a characteristic dynamical value;
- $\tilde{\Phi}_0 = \tilde{R}\tilde{T}/\tilde{F}$ is the thermic potential is taken as the characteristic one; here $\tilde{F}$ is Faraday's constant; $\tilde{R}$ is the universal gas constant; $\tilde{T}$ is the absolute temperature;
- $\tilde{C}_0$ is the characteristic ion concentration.

Then the problem is described by eleven dimensionless parameters, namely:

- $E_0 = \tilde{E}_0\,\tilde{h}_0/\tilde{\Phi}_0$ is the external electric field;
- $\Pi = \tilde{\Pi}\,\tilde{h}_0^2/\tilde{\mu}_e\tilde{D}$ is the pressure gradient;
- $\omega = \tilde{\omega}\tilde{h}_0^2/\tilde{D}$ is the frequency of the external electric field;
- $\nu = \tilde{\lambda}_D/\tilde{h}_0$ is the Debye number;
- $\delta = \tilde{\varepsilon}_d/\tilde{\varepsilon}_e$ is the ratio of the electric permittivities of the dielectric and electrolyte;
- $\mu = \tilde{\mu}_d/\tilde{\mu}_e$ is the ratio of the viscosities of the dielectric and electrolyte;
- $\varkappa = \tilde{\varepsilon}_e\tilde{\Phi}_0^2/\tilde{\mu}_e\tilde{D}$ is the coupling coefficient between the hydrodynamics and the electrostatics;
- $We = \tilde{\gamma}\tilde{h}_0/\tilde{\varepsilon}_e\tilde{\Phi}_0^2$ is the Weber number, where $\tilde{\gamma}$ is the surface tension coefficient;
- $q = \tilde{q}/\tilde{\sigma}_s$ is the dimensionless immobile wall surface charge;
- $\sigma_0 = \tilde{\sigma}_0/\tilde{\sigma}_s$ is the dimensionless mobile surface charge on the interface for the one-dimensional solution;
- $H = \tilde{H}/\tilde{h}_0$ is the ratio of the layers thickness of the dielectric and the electrolyte.

Here $\tilde{\lambda}_D$ is the Debye length,

$$\tilde{\lambda}_D = \left(\frac{\tilde{\varepsilon}_e\tilde{\Phi}_0}{\tilde{F}\tilde{C}_0}\right)^{1/2} = \left(\frac{\tilde{\varepsilon}_e\tilde{R}\tilde{T}}{\tilde{F}^2\tilde{C}_0}\right)^{1/2},$$

and $\tilde{\sigma}_s$ is the characteristic surface charge,

$$\tilde{\sigma}_s = \frac{\tilde{\varepsilon}_e\tilde{\Phi}_0}{\tilde{\lambda}_D}.$$

Note that, if for the one-dimensional steady-state case σ is some constant, $\sigma = \sigma_0$, for the two-dimensional case the surface charge at the interface is a function of x and t, $\sigma = \sigma(x,t)$.

The typical parameter values are taken in all our calculations: $q = -3$, $\sigma = 1$, $\nu = 0.1$, $\varkappa = 0.2$, $We = 2\times 10^4$, $\delta = 1.25\times 10^{-2}$, $\mu = 2$, $H = 4$. The control parameters E_0, Π, and ω vary in the intervals, respectively, -200; 200, 0; 100, and 0; 10.

The electrolyte is described by the Nernst-Planck-Poisson-Stokes equations, taken in a dimensionless form, as follows:

$$\frac{\partial C^{\pm}}{\partial t} + \mathbf{U}\cdot\nabla C^{\pm} = \pm\nabla\cdot\left(C^{\pm}\nabla\Phi\right) + \nabla^2 C^{\pm}, \tag{1}$$

$$\nu^2\,\nabla^2\Phi = C^- - C^+, \tag{2}$$

$$-\nabla P + \nabla^2\mathbf{U} = \frac{\varkappa}{\nu^2}(C^+ - C^-)\nabla\Phi, \tag{3}$$

$$\nabla\cdot\mathbf{U} = 0, \tag{4}$$

where (1) is the ion transport equations, (2) is the Poisson equation for the electric potential, and (3), (4) are the Stokes equations with the Coulomb force in the right-hand side. Here $C^{\pm}$ are the concentration of the positive and negative ions, $\mathbf{U} = (U,V)$ is electroosmotic velocity, Φ is the electrical potential, and P is pressure. All the unknowns are taken in the electrolyte.

The dielectric liquid is electrically neutral, the potential φ is described by the Laplace equation, and hydrodynamics is described by the Stokes equation without the Coulomb force,

$$\nabla^2\varphi = 0, \tag{5}$$

$$-\nabla p + \mu\nabla^2\mathbf{u} = 0, \tag{6}$$

$$\nabla\cdot\mathbf{u} = 0. \tag{7}$$

The boundary conditions (BCs) on the solid surfaces, $y = 0$ and $y = H$, are

$$y = 0: \qquad \nu\frac{\partial\Phi}{\partial y} = -q, \tag{8}$$

$$C^+\frac{\partial\Phi}{\partial y} + \frac{\partial C^+}{\partial y} = 0, \qquad -C^-\frac{\partial\Phi}{\partial y} + \frac{\partial C^-}{\partial y} = 0, \tag{9}$$

$$V = 0, \qquad U = 0, \tag{10}$$

$$y = H: \qquad \frac{\partial\varphi}{\partial y} = 0, \quad u = v = 0. \tag{11}$$

At the interface the unknowns obey the following BCs:

$$y = h(x,t): \quad C^+ \frac{\partial \Phi}{\partial n} + \frac{\partial C^+}{\partial n} = 0, \quad -C^- \frac{\partial \Phi}{\partial n} + \frac{\partial C^-}{\partial n} = 0, \tag{12}$$

$$\Phi = \varphi, \qquad \nu \frac{\partial \Phi}{\partial n} = \delta \nu \frac{\partial \varphi}{\partial n} + \sigma, \tag{13}$$

$$\frac{\partial \sigma}{\partial t} + \frac{\partial (U_s \sigma)}{\partial x} = 0, \tag{14}$$

$$\begin{aligned} &P + \frac{We}{r} + 2\frac{\partial U}{\partial x}\frac{1-h_x^2}{1+h_x^2} + 2\left(\frac{\partial U}{\partial y} + \frac{\partial V}{\partial x}\right)\frac{h_x}{1+h_x^2} \\ &-\frac{1}{2}\varkappa\left\{\left(\frac{\partial \Phi}{\partial n}\right)^2 - \left(\frac{\partial \Phi}{\partial s}\right)^2\right\} = \\ &p + 2\mu\frac{\partial u}{\partial x}\frac{1-h_x^2}{1+h_x^2} + 2\mu\left(\frac{\partial u}{\partial y} + \frac{\partial v}{\partial x}\right)\frac{h_x}{1+h_x^2} \\ &-\frac{\delta}{2}\varkappa\left\{\left(\frac{\partial \varphi}{\partial n}\right)^2 - \left(\frac{\partial \varphi}{\partial s}\right)^2\right\}, \end{aligned} \tag{15}$$

$$\begin{aligned} &-4\frac{\partial U}{\partial x}\frac{h_x}{1+h_x^2} + \left(\frac{\partial U}{\partial y} + \frac{\partial V}{\partial x}\right)\frac{1-h_x^2}{1+h_x^2} + \varkappa\frac{\partial \Phi}{\partial n}\frac{\partial \Phi}{\partial s} = \\ &\mu\left(-4\frac{\partial u}{\partial x}\frac{h_x}{1+h_x^2} + \left(\frac{\partial u}{\partial y} + \frac{\partial v}{\partial x}\right)\frac{1-h_x^2}{1+h_x^2}\right) + \delta\varkappa\frac{\partial \varphi}{\partial n}\frac{\partial \varphi}{\partial s}, \end{aligned} \tag{16}$$

$$V = \frac{\partial h}{\partial t} + U\frac{\partial h}{\partial x}, \quad U = u, \quad V = v. \tag{17}$$

The BCs (9), (12) means that the surface is impermeable to the cations and anions, the condition (13) means that the electric potential is continuous on the surface, while its normal to the surface derivative has a jump associated with the mobile surface charge, and eq. (14) is the surface charge conservation equation. The dynamic BCs for the normal and tangential stresses, (15) and (16), and the kinematic conditions (17) complement the statement.

3 One-dimensional solution

The electrostatic problem (1), (2) can be solved independently of the hydrodynamic problem. The electric potential can be represented as the following superposition:

$$\Phi = \Phi_t(t,x) + \Phi_n(y), \qquad \Phi_t(t,x) = E_\infty x = E_0 x \cos \omega t. \tag{18}$$

The tangential field $E \equiv \mathrm{d}\Phi_n/\mathrm{d}y$ and the ion concentrations $C^\pm$ obey the following system:

$$C^+ E + \frac{\mathrm{d}C^+}{\mathrm{d}y} = 0, \tag{19}$$

$$-C^- E + \frac{\mathrm{d}C^-}{\mathrm{d}y} = 0, \tag{20}$$

$$\nu^2 \frac{\mathrm{d}E}{\mathrm{d}y} + C^+ - C^- = 0 \tag{21}$$

with the BCs

$$y = 0: \qquad E = -\frac{1}{\nu}\, q, \tag{22}$$

$$y = 1: \qquad E = \frac{1}{\nu}\, \sigma. \tag{23}$$

After solution of the electrostatic part of the problem, the hydrodynamic movement can be easily found. The equations for the upper, dielectric liquid (6),(7) with the pressure gradient $\mathrm{d}p/\mathrm{d}x = -\Pi$ turn into

$$\mu \frac{\mathrm{d}^2 u}{\mathrm{d}y^2} = -\Pi, \tag{24}$$

$$y = H: \qquad u = 0, \tag{25}$$

$$y = 1: \qquad \mu \frac{\mathrm{d}u}{\mathrm{d}y} = \frac{\mathrm{d}U}{\mathrm{d}y} - \varkappa E E_\infty, \qquad u = U. \tag{26}$$

Upon integration of (24) and using the corresponding BCs, we get

$$\mu \frac{\mathrm{d}u}{\mathrm{d}y} = A_1 - \Pi(y-H), \qquad \mu\, u = A_1(y-H) - \frac{\Pi}{2}(y-H)^2,$$

where A_1 is the constant of integration,

$$A_1 = \frac{\mu\, u(1)}{(1-H)} + \frac{\Pi}{2}(1-H).$$

Eventually, we get the effective BC for the electrolyte:

$$y = 1: \qquad \frac{\mathrm{d}U}{\mathrm{d}y} - \frac{\mu}{1-H} U = \varkappa E E_\infty - \frac{\Pi}{2}(1-H). \tag{27}$$

Equations (3), (4) with the BCs $U(0) = 0$ and (27) become

$$\frac{\mathrm{d}P}{\mathrm{d}y} = \varkappa\, E \frac{\mathrm{d}E}{\mathrm{d}y} = \frac{\varkappa}{2}\frac{\mathrm{d}E^2}{\mathrm{d}y}, \tag{28}$$

$$\frac{\mathrm{d}^2 U}{\mathrm{d}y^2} - \varkappa \frac{\mathrm{d}E}{\mathrm{d}y} E_\infty = -\Pi. \tag{29}$$

Upon first integration of (29),

$$\frac{\mathrm{d}U}{\mathrm{d}y} = \varkappa E_\infty\, E - \Pi y + \mathrm{const.} \tag{30}$$

For the second BC at $y = 1$, const $= 0$. Upon the second integration, we get

$$U = \varkappa E_\infty \int_0^y E\, \mathrm{d}y + C_1 y - \Pi \frac{y}{2}, \tag{31}$$

where

$$C_1 = \frac{\varkappa \mu \int_0^1 E\, \mathrm{d}y E_\infty - \frac{\Pi}{2}(\mu - 1 - H^2)}{1 - H - \mu}. \tag{32}$$

The pressure is found from eq. (28), *i.e.*,

$$P = \mathrm{const}(y) + \frac{\varkappa}{2} E^2 - \Pi x.$$

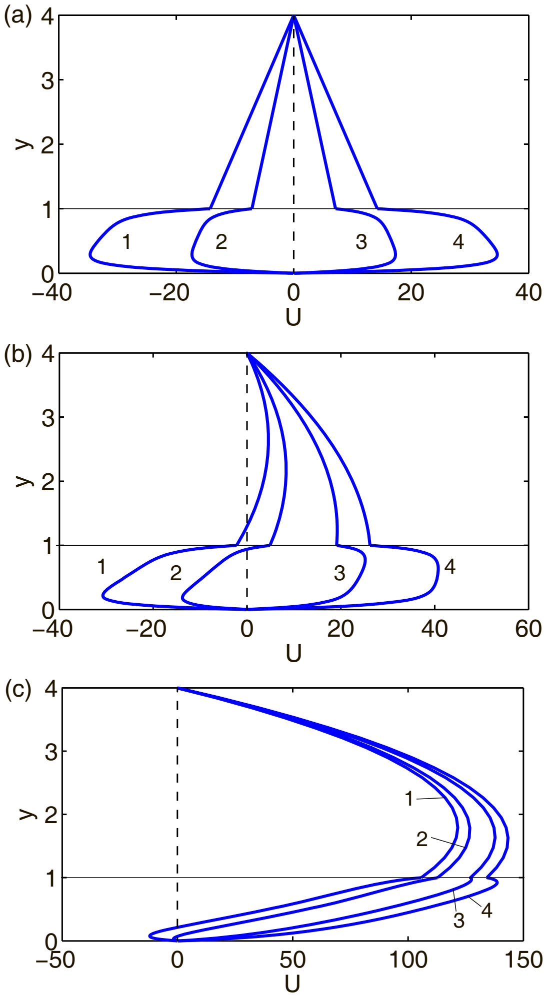

Fig. 2. Velocity profiles for $E_0 = 100$ for different values of Π: (a) $\Pi = 0$, (b) $\Pi = 10$, (c) $\Pi = 100$ and different times: $t = 0$ (1), $t = \frac{\pi}{3\omega}$ (2), $t = \frac{2\pi}{3\omega}$ (3), $t = \frac{\pi}{\omega}$ (4).

The constant of integration can be found from the BC (15) for the normal stresses. For the one-dimensional case it is

$$y = 1: \qquad P - \frac{\varkappa}{2}E^2 + \frac{\varkappa}{2}(1-\delta)E_\infty^2 = p, \tag{33}$$

or

$$\text{const} = -\frac{\varkappa}{2}(1-\delta)E_\infty^2.$$

Thus,

$$P = \frac{\varkappa}{2}E^2 - \frac{\varkappa}{2}E_\infty^2(1-\delta) - \Pi x. \tag{34}$$

In fig. 2 the velocity profile $U(y)$ is presented for the AC case for the different time moments, t; the electric field, $E_0 = 100$, and several values of the pressure gradient Π are taken. If $\Pi = 0$, the mean flow is absent, so the profiles are symmetrical about zero. We emphasize that all the relations in sect. 3, by virtue of the parametric dependence on time for the one-dimensional case, are valid either for the DC or AC cases.

For the positive E_∞, the pressure-driven and the electroosmotic flows are counterdirected, while for E_∞ negative they are codirected.

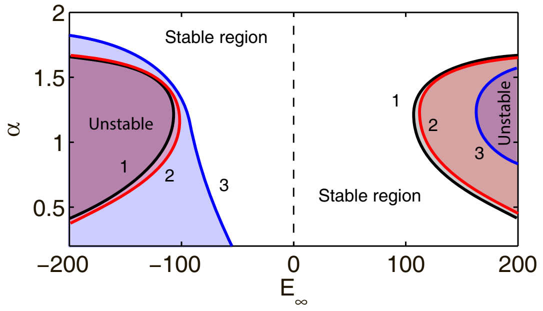

Fig. 3. Marginal stability curves for the DC case and different values of Π: $\Pi = 0$ (1), $\Pi = 10$ (2), $\Pi = 100$ (3).

4 Investigation of instability

In contrast to the one-dimensional flow, the DC and AC cases for the stability problem are drastically different. Let us superimpose small, sinusoidal perturbations with a wave number α along the x-axis to the one-dimensional solution,

$$f(t,x,y) = f_0(t,y) + \hat{f}(t,y)\exp(\mathrm{i}\alpha x), \tag{35}$$

where f represents all the unknowns in the electrolyte and the dielectric. The substitution of the expansion (35) into eqs. (1)–(17) leads to the system of equations with respect to perturbations $\hat{f}$. The resulting cumbersome system of ordinary differential equations is not given in this short paper. For the DC electric field, the dependences of perturbations on time could be found in the form $\hat{f}(t,y) = \hat{f}_1(y)\exp(\lambda t)$. The one-dimensional flow is stable, if for all wave numbers α $Re(\lambda) < 0$; if there are wave numbers α, for which $Re(\lambda) > 0$, the flow is unstable. The neutral marginal curve separates the stable and unstable cases.

For the AC electric field according to the Floquet theorem the perturbations can be sought in the form

$$\hat{f}(t,y) = \sum_{k=-\infty}^{\infty} \hat{f}_k(y)\exp[(\lambda + \mathrm{i}k\omega)t], \tag{36}$$

where now λ is the Floquet coefficient. As for the DC case positive $Re(\lambda)$ corresponds to the instability, while negative $Re(\lambda)$ to the stability of the system.

Two types of wave instability in the two-phase electroosmotic flows were found in the papers [6,7]. Here we concentrate on the most dangerous, either for the DC or the AC fields, short wave instability.

In fig. 3 the marginal stability curves for the DC case for several values of Π are presented. One can see that the pressure gradient has a different influence for the cases $E_\infty < 0$ and $E_\infty > 0$. In the case when the pressure-driven flow and the electroosmotic flow are codirected ($E_\infty < 0$) the increasing of the pressure gradient leads to the system destabilization and, on the contrary, when the pressure-driven flow and the electroosmotic flow are the counterdirected ($E_\infty > 0$), the increasing of the pressure gradient leads to the system stabilization.

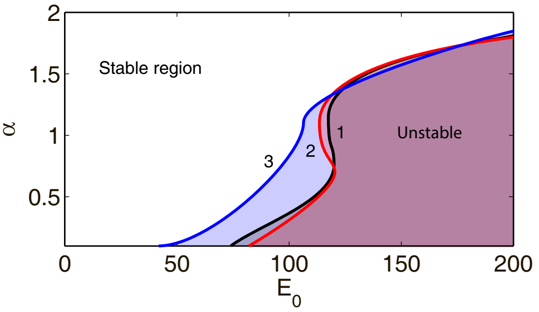

Fig. 4. Marginal stability curves for the AC field with frequency $\omega = 1$ and different values of Π: $\Pi = 0$ (1), $\Pi = 10$ (2), $\Pi = 100$ (3).

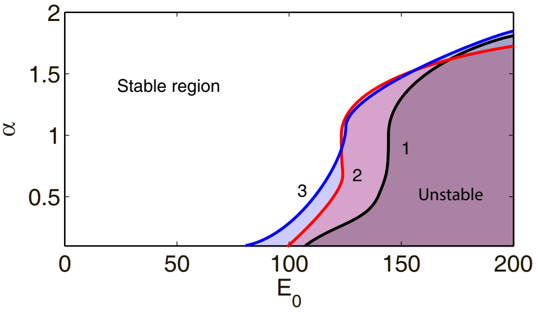

Fig. 5. Marginal stability curves for the AC field with frequency $\omega = 10$ and different values of Π: $\Pi = 0$ (1), $\Pi = 10$ (2), $\Pi = 100$ (3).

For the case of the AC electric field the influence of the external pressure gradient is not monotonous and for different wave numbers α it can either stabilize or destabilize the system, see fig. 4; usually the persistence of the external pressure gradient mostly destabilizes the one-dimensional flow and the instability arises at smaller values of E_0. The increase of the electric field frequency (fig. 5) leads to the stabilizing of the one-dimensional flow for all values of the external pressure gradient.

5 Conclusion

The stability of the electroosmotic flow of conductive (electrolyte) and non-conductive (dielectric) viscous liquids under the influence of constant and alternating electric fields was investigated. The fluids are bounded by two infinite parallel plates. The lower solid wall is a charged surface, and the upper wall is isolated. The charge at the lower boundary is assumed to be stationary, and the surface charge at the free interface of the fluids is assumed to be mobile. The problem was investigated using asymptotic and numerical methods. For a sufficiently strong external field, the one-dimensional solution becomes unstable.

The dependences of the critical values of the external electric field intensity on the different values of external pressure and electric field frequency were obtained theoretically. It was found out that for the case of AC electric field the increase of the external pressure mostly leads to the destabilization of the one-dimensional flow. For the DC electric field the influence depends on the direction of the electric field.

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Author contribution statement

The statement of the problem belongs to E.A. Demekhin, G.S. Ganchenko worked on the computer realization of the numerical solution and on the analysis of the obtained results, E.V. Gorbacheva conducted the calculations and worked on the paper along with G.S. Ganchenko.

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