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Instability of pressure driven viscous fluid streams in a microchannel under a normal electric field

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Abstract
This paper investigates analytically and experimentally electrohydrodynamic instability of the interface between two viscous fluids with different electrical properties under constant flow rates in a microchannel. In the three-dimensional analytical model, the two-layer system is subjected to an electric field normal to the interface between the two fluids. There is no assumption on the magnitude of the ratio of fluid to electric time scales, and thus the linear Poisson-Boltzmann equation are solved using separation of variable method for densities of bulk charge and surface charge. The electric field and fluid dynamics are coupled only at the interface through the tangential and normal interfacial stress balance equations. In the experiments, two immiscible fluids, aqueous NaHCO₃ (the high electrical mobility fluid) and silicone oil (polydimethylsiloxane, the low electrical mobility fluid) are pumped into a microchannel made in polymethyl methacrylate) (PMMA) substrate. The normal electric field is added using a high voltage power supply. The results showed that the external electric field and increasing width of microchannel destabilize the interface between the immiscible fluids. At the same time, the viscosity of the high electrical mobility fluid and flow rates of fluids has a stabilizing effect. The experimental results and the analytical results show a reasonable agreement.

Keywords: Electrohydrodynamic; Constant flow rate; Linear instability; Microchannel

1. Introduction
In microfluidic devices, interfacial instability can produce a variety of flow patterns or even develop into turbulence. In a microchannel, viscous force dominates and mixing by inertia driven turbulence is impossible. On the one hand, the possibility of using instability to achieve efficient mixing using different methods was explored by various groups [1-3]. On the other hand, stable flow is crucial for pumping immiscible liquids [4, 5]. The identification of parameters for interfacial instability is necessary for controlling the flow pattern. In many conditions, electro-osmotic flow is the primary method of fluid
handing [6]. Electrohydrodynamics is regarded as a branch of fluid mechanics concerned with the effect of electric forces. The field is also considered as a part of electrodynamics involving the interaction between moving media and electric fields [7].

The electrohydrodynamic instability theory was pioneered by Taylor and Melcher using the leaky dielectric model [7]. In general, there are two modeling approaches in the presence of interface instability.

The first approach is the bulk coupled model that assumes a conductivity gradient in a thin diffusion layer between the two fluids, resulting in an electrical body force on the fluid. The interaction between the conductivity gradient and high electric field plays a critical role in the generation of interface instability. A number of researchers studied the effect of conductivity gradient and high electric field [8-12]. Comparing with the work of Oddy et al. [1], Chen and Santigo [8] provided a more systematic investigation to establish the critical importance of conductivity gradient and high electric field strength for inducing the instability. The results showed that the conductivity gradient and their associated bulk charge accumulation are crucial for instability. Lin et al. [9] studied temporal instability in a T-junction merging two fluid streams of different conductivities via linear stability analysis, experiments, and numerical simulations. All the methods show that there are thresholds where the electric body force is too great for diffusion to quench. In this flow, the electric field was normal to the conductivity gradient. Chen et al. [10] used experiments and linear stability analysis to study convective instability occurring in an electroosmotic flow [9]. Using the method of numerical simulations, Storey [11] showed the generation of turbulence with a relatively low Reynolds number, and evaluated the effects of different assumptions in boundary conditions of nonlinear behavior and mixing. Kang et al. [12] studied the initial growth of electrohydrodynamic instability via numerical simulation. The results showed that the molecular diffusion has a dual role in the onset and the development of instability and the system is unstable to high electric fields even without any external disturbances.

The second approach is the surface coupled model that considers a jump in electrical conductivity at the interface of the two fluids. In this method, the bulk electrical force is vanished. The discontinuity of the electrical properties of the fluids across the interface affects the force balance at the fluid-fluid interface, which may either stabilize or destabilize the interface. Mohamed et al. [13] analyzed the stability of the Couette-Poiseuille flow. The results showed that the velocity stratification affects the stability of the fluids. The normal electric field is greatly reduced by the increase of the thickness of the non-conducting fluid layer. Abdella and Rasmussen [14] analyzed Couette flow in an unbounded domain subjected to a normal electric field. Two special cases, the electrohydrodynamic free-charge case and the electrohydrodynamic polarization charge configuration, were analyzed using Ariy functions and Ariy integrals [14]. Thaokar and Kumaran [15] analyzed the stability of the interface between two dielectric fluids confined between parallel plates subjected to a normal electric field in the limit of zero
Reynolds number. The results indicated that the interface becomes unstable when the electric field exceeds a critical value, and the critical value are influenced by the ratio of dielectric constant, ratio of viscosities, ratio of thickness, and surface tension. The results also showed that for a small wave number ($k$), the critical potential increases proportional to $k$. For a large wave number, the critical potential increases proportional to the square root of $k$. Ozen et al. [16] analyzed the linear stability of the interface between two immiscible fluids and found the effects of electric fields and mechanical properties to the instability of the interface. Moatimid and Obied Allah [17] investigated the linear surface wave instability between two finite fluid layers. The fluid layers have different electric properties and are subjected to an electric field normal to their interface. The surface tension showed to have a stabilizing influence, while the streaming velocity was strictly destabilizing.

Using the surface coupled model, Goranović [18] analytically investigated the instability of two immiscible inviscid fluids in microchannels under static state; the effect of interfacial free charge was ignored. The aim of the present paper is to study the effect of normal electric fields on two immiscible viscid fluids under the combined effect of hydrodynamic and electroosmosis in microchannels. The effects of viscosity and surface charge are included in the analysis. In the analytical model, electric field and fluid dynamics are coupled at the interface through the tangential and normal interfacial stress balance equations. The instability of the interface between conducting fluid and non-conducting fluid is both analytically and experimentally investigated.

2 The physical and mathematical model

2.1 Methodology and approach

The usual procedure in linear stability analysis can follow six basic steps. In the first step, general governing equations and general boundary conditions are introduced. The governing equations control the initial and stationary flow conditions in the velocity field $V_0$. The governing equations include the continuity equation, the momentum equations, the Maxwell equations and the Poisson-Boltzmann equation.

In the second step, the perturbation of $v'$ is introduced and superposed to the stationary velocity $V_0$. The velocity field changes into $v = v_0 + v'$. Substituting the equation into governing equations and boundary conditions, the equations governing the perturbation are obtained.

In the third step, the equations governing the perturbation are linearized, the terms with quadratic and higher terms in $v'$ are neglected.
In the forth step, the perturbation quantities \( \mathbf{v}' \) are further expressed in terms of normal modes. A spatially and temporally periodic perturbation can be assumed in the form of
\[
\mathbf{v}_k'(\mathbf{r},t) = v_k(z) \exp(-i\omega_k t) \exp(i(\mathbf{k} \cdot \mathbf{r})).
\]

In the fifth step, the perturbation \( \mathbf{v}_k'(\mathbf{r},t) \) is substituted into the governing equations, the temporal stability can be solved. The complex solution for \( \omega_k \) can be described as \( \omega_k = \text{Real}(\omega_k) + i \text{Im}(\omega_k) \).

In the sixth step, the freely evolving waves are spatially periodic disturbances of infinite spatial extent which travel with a phase velocity, \( c_r = \omega_r/k \) and grow or decrease in amplitude with a temporal growth rate, \( \text{Im} \omega_k \). The system is considered to be linearly unstable to infinitesimal disturbances for \( \text{Im} \omega_k > 0 \). The stability regimes can be distinguished as unstable \( (\text{Im} \omega_k > 0) \), neutral \( (\text{Im} \omega_k = 0) \) and stable \( (\text{Im} \omega_k < 0) \).

### 2.2 General equations of motion

Figure 1(a) shows the stability of the interface between two immiscible fluids under the combined effect of hydrodynamic and electroosmosis. Fluid 1 is conducting with high electroosmotic mobility and fluid 2 is non-conducting with low electroosmotic mobility. The flows are induced by the pressure source and the electric field. The two fluids have different properties. The applied electric field is normal to the unperturbed interface. Fig. 1(c) shows the cross sectional view of the two fluids in the rectangular microchannel, \( h_1 \) and \( h_2 \) are denoted as the fraction of fluid 1 and fluid 2, respectively.

The following notation will be used: superscripts (1) and (2) denote quantities pertaining to fluid 1 and fluid 2. The prime symbol (‘) indicates a perturbed variable; the subscript 0 denotes the unperturbed variables; other subscripts are used for vector component.

The momentum equation for an isotropic incompressible Newtonian liquid is given by
\[
\rho \frac{D\mathbf{v}^{(j)}}{Dt} = -\nabla P^{(j)} + \mu \nabla^2 \mathbf{v}^{(j)} + F_{e}^{(j)} \quad (j = 1, 2)
\]
where \( \rho \) is the density, \( P \) the pressure, \( \mu \) the dynamical viscosity, \( \mathbf{v} \) the velocity, \( F_{e} \) the electric body force. In microsystems, the effect of gravity can be ignored [18]. In the case of a homogeneous dielectric, the permittivity is constant, and the electric force \( F_{e} \) reduces to [18]
\[
F_{e} = \rho_e \mathbf{E}
\]
where \( \rho_e \) is the density of electric charge, \( \mathbf{E} \) is the electric field. The continuity equation for incompressible liquids is
\[
\nabla \cdot \mathbf{v}^{(j)} = 0 \quad (j = 1, 2)
\]
Assuming that the electric charge density is not affected by the external electric field due to thin EDLs and the small fluid velocity, the charge convection can be ignored and
the electric field equation and the fluid flow equation are decoupled [19, 20]. According
the previous work [5, 21, 22], the governing equations for the free charge can be
described as

\[ \nabla^2 \psi = \frac{2z_0 e n_0}{\epsilon} \sinh \left( \frac{z_0 e \psi}{k_b T} \right) \] (4)

and

\[ \rho_e = -2z_0 e n_0 \sinh(z_0 e \psi/k_b T) \] (5)

where \( \psi \) is the electric potential, \( z_0 \) is the valence of the ions, \( e \) is elementary charge, \( n_0 \) is the reference value of the ion concentration, \( \epsilon \) is the permittivity of the fluid, \( k_b \) is Boltzmann constant, \( T \) is the absolute temperature.

The governing equations for the electric fields within the dielectrics are

\[ \nabla \cdot \mathbf{E} = 0 \] (6)

Since large currents are not present in this flow, the effect of magnetic induction is
negligible. Hence, the electric field is assumed to be irrorational

\[ \nabla \times \mathbf{E} = 0 \] (7)

2.3 General boundary condition

On the wall of the microchannel, the non-slip condition is assumed for the velocities

\[ \mathbf{v}^{(1)} = 0; \quad (z = -h_1) \] (8)

\[ \mathbf{v}^{(2)} = 0; \quad (z = h_2) \] (9)

When the electric fields are added to the fluids, the boundary conditions of the
electric field at the wall can be described as

\[ \begin{aligned}
\{ \mathbf{n} \times \mathbf{E}^{(1)} = 0 \} & \text{ at } z = -h_1 \\
\mathbf{v}^{(1)} = 0 & 
\end{aligned} \] (10)

and

\[ \begin{aligned}
\{ \mathbf{n} \times \mathbf{E}^{(2)} = 0 \} & \text{ at } z = h_2 \\
\mathbf{v}^{(2)} = \text{ voltage of power} & 
\end{aligned} \] (11)

where \( \mathbf{n} \) is the normal direction of the wall.

Consider the effect of a small wave disturbance on the interface \( z = 0 \). The surface
disturbance is assumed as

\[ z = \zeta(x, y, t) \] (12)

where \( \zeta \) is the vertical displacement for the equilibrium position \( z = 0 \). We consider only a
small value of \( \zeta \). Any deformed surface is characterized by a unit normal vector \( \mathbf{n} \):
The linear form of Eq. (13) is

\[ n = \frac{1}{\sqrt{(\frac{\partial \zeta}{\partial x})^2 + (\frac{\partial \zeta}{\partial y})^2 + 1}} \left( -\frac{\partial \zeta}{\partial x}, -\frac{\partial \zeta}{\partial y}, 1 \right) \]  

(13)

\( f \) is defined as the surface function of the two immiscible fluids. The notation \([f]\) is the jump quantity between the two values of \( f \) taken respectively on the boundaries of the interfacial layer, \([f] = f_2 - f_1\). The notation is adopted from the previous works [16, 18]. The change in any function \( f(x, y, z, t) \) across the interface in this paper is

\[ [f] = f(x, y, \zeta^-, t) - f(x, y, \zeta^+, t) = (f_1 - f_2)_{z=\zeta} \]  

where the superscripts of – and + means the side of interface facing fluid 1 an fluid 2 respectively. At the interface \( z = \zeta \), the continuity of velocities are

\[ \begin{bmatrix} u \\ v \\ w \end{bmatrix} \bigg|_{t=1} = \begin{bmatrix} u \\ v \\ w \end{bmatrix} \bigg|_{t=2} = \frac{d\zeta}{dt} \]  

(14)

where \( u, v, w \) are the velocities along \( x, y, z \) directions respectively.

The continuity of normal electrical fields at the interface can be described as

\[ \mathbf{n} \cdot \left[ \mathbf{\zeta E} \right] = 0 \]  

(15)

\[ \mathbf{n} \times \left[ \mathbf{E} \right] = 0 \]  

(16)

The stress balance at any part on the interface can be described using the following equation

\[ \left[ -p \right] n_i + \left[ \tau_{ik} + T^M_{ik} \right] n_k + q_s \cdot E_i = \sigma \nabla^2 \zeta n_i \]  

(17)

where \( p \) is the pressure, \( \tau_{ik} \) is the viscous stress, \( T^M \) is the Maxwell stress tensor, \( q_s \) is the surface charge which can be calculated in the previous works [5, 22], \( \sigma \) is the surface tension, \( \nabla^2 \zeta \) describes the surface curvature for small deformations, the subscripts of \( i \) and \( k \) mean the vector component \( i = x, y, z \) and \( k = x, y, z \).

2.4 Perturbation equations

The analysis involves the assumption that the perturbations to the base state are infinitesimally small. To carry out a linear stability analysis, all high-order terms in the perturbation quantities are neglected. The expanding solutions are

\[ \mathbf{v} = \mathbf{v}_0 + \mathbf{v}' + \cdots \]  

(18)

\[ p = p_0 + p' + \cdots \]  

(19)

2.4.1 The base state solution
In the base state, the velocity field and the electric field can be described by the unperturbed equations. Under unperturbed condition, Eqs. (20) and (21) transform into
\[
\mathbf{v} = \mathbf{v}_0 \\
p = p_0
\] (22)

The governing equations of Eqs. (1) and (3) can be transformed into the base state equations:
\[
\frac{\partial (\rho \mathbf{v}_0)}{\partial t} = -\nabla p_0 + \mu \nabla^2 \mathbf{v}_0 \\
\nabla \cdot \mathbf{v}_0 = 0
\] (24)

The details of velocity profile and pressure gradient have been described previously [5].

The vector \( \mathbf{n} \) normal to the unperturbed interface is given by
\[
\mathbf{n}_0 = (0, 0, 1)
\] (26)

The stress balance at the interface can be described as following equations [18]:
\[
[P_0]_{nx} = [P_0]_{ny} = 0
\] (27)
\[
[P_0]_{nz} = \|P\| + \frac{1}{2} \| \varepsilon E_{z0}^2 \| + q_s \cdot E_{z0}
\] (28)

where the subscripts of \( nx, ny \) and \( nz \) means three components of normal vector in \( x, y \) and \( z \) directions respectively.

### 2.4.2 Linearization of the perturbed equations

Under perturbed condition, Eqs (1) and (3) transform into
\[
\frac{\partial (\rho (\mathbf{v}_0 + \mathbf{v}'))}{\partial t} = -\nabla (p_0 + p') + \mu \nabla^2 (\mathbf{v}_0 + \mathbf{v}')
\] (29)
\[
\nabla \cdot (\mathbf{v}_0 + \mathbf{v}') = 0
\] (30)

In this section, the changes in the density, viscosity, and charge density are not considered since they are assumed to be constant in each region. When subtracting the base state of Eqs. (24) and (25) from Eqs. (29) and (30), the equations reduce to
\[
\rho \frac{\partial \mathbf{v}'}{\partial t} = -\nabla p' + \frac{1}{Re} \mu \nabla^2 \mathbf{v}'
\] (31)
\[
\nabla \cdot \mathbf{u}' = 0
\] (32)

Combining Eq. (31) and (32) leads to
\[
\nabla^2 p' = 0
\] (33)

For a constant charge density, the electric field can be described as
\[
\nabla^2 E' = 0
\] (34)

As the electric field is irrotational, the governing equation for \( \mathbf{E}' \) is
\[
\nabla \times \mathbf{E}' = 0
\] (35)
2.4.3 Linearization of the perturbed boundary conditions of rigid boundaries

The boundary of the wall under perturbed conditions can be linearized as following

\[ w^{(1)'} = u^{(1)'} = v^{(1)'} = 0 \quad \text{at} \quad z = -h_1 \]  
\[ w^{(2)'} = u^{(2)'} = v^{(2)'} = 0 \quad \text{at} \quad z = h_2 \]  
\[ n_0 \times E^{(1)'} = 0 \quad \text{at} \quad z = -h_1 \]  
\[ n_0 \times E^{(2)'} = 0 \quad \text{at} \quad z = h_2 \]

2.4.4 Linearization of the perturbed boundary conditions of the interface

Since \( \zeta \) is assumed to be small, the first order approximation

\[ E_i(0 + \zeta) = E_i(0) + \frac{\partial E_i(0)}{\partial z} \cdot \zeta + \cdots \approx E_i(0) \]  

is valid if both \( E_i' \) and \( \zeta \) remains small. In Eq. (40), \( E_i(0) \) is the perturbation of electric field at \( z = 0 \) and \( E_i(0 + \zeta) \) is the perturbation of electric field at \( z = \zeta \). The normal vector is now

\[ \mathbf{n} = n_0 + \mathbf{n}' \]  

where \( \mathbf{n}' \) is

\[ \mathbf{n}' = \left( -\frac{\partial \zeta}{\partial x}, -\frac{\partial \zeta}{\partial y}, 0 \right) \]  

The boundary conditions for perturbation at \( z = \zeta \) are

\[ \begin{bmatrix} u' \\ v' \end{bmatrix} = \begin{bmatrix} u' \\ v' \end{bmatrix} = \begin{bmatrix} w' \end{bmatrix} = 0 \]  
\[ w'(0) = \frac{d\zeta}{dt} \]  

The electric fields is described as

\[ \mathbf{E} = \left( E'_x, E'_y, E_{z0} + E'_z \right). \]  

The boundary condition of Eq. (17) and (18) at the interface is

\[ \begin{bmatrix} E'_x + E_{z0} \frac{\partial \zeta}{\partial x} \end{bmatrix} = 0 \]  
\[ \mathbb{K} \mathbb{E}_z = 0. \]

2.4.5 Linearized stresses

In a microsystem, the effect of gravity is ignored. Substituting Eq. (21) into Eq. (19), the stress conditions of the interface can be described as

\[ -[P'] n_i - [P'] n_i + \left[ \tau_{ik} + \mathbb{T}^M_{ik} \right] n_k + q_s \cdot E_i = \sigma \nabla^2 \zeta n_i \]
Eq. (48) can be transformed into following equations in $x$, $y$, $z$ directions

\[
\left( [\varepsilon E_0^2] + \left[ \rho \right] \right) \cdot \frac{\partial \zeta}{\partial x} + \mu \left( \frac{\partial u'}{\partial z} + \frac{\partial w'}{\partial x} \right) = q_x \cdot E_x + [\varepsilon E_0 E_z] = 0 \tag{49}
\]

\[
\left( [\varepsilon E_0^2] + \left[ \rho \right] \right) \cdot \frac{\partial \zeta}{\partial y} + \mu \left( \frac{\partial u'}{\partial z} + \frac{\partial w'}{\partial y} \right) = q_y \cdot E_y + [\varepsilon E_0 E_z] = 0 \tag{50}
\]

\[
- [p'] + 2\mu \frac{\partial w'}{\partial z} + [\varepsilon E_0 E_z] = q_z \cdot E_z = \sigma \nabla^2 \zeta \tag{51}
\]

\[2.4.6 \text{ Expansion in normal modes}\]

The perturbation can be expanded into the normal model using the following equations

\[
\zeta = \hat{\zeta} \exp[i(k_x x + k_y y) + nt] \tag{52}
\]

\[
\mathbf{v}' = \hat{v}(z) \exp[i(k_x x + k_y y) + nt] \tag{53}
\]

\[
p' = \hat{p}(z) \exp[i(k_x x + k_y y) + nt] \tag{54}
\]

\[
\mathbf{E}'_i = \hat{E}_i(z) \exp[i(k_x x + k_y y) + nt] \tag{55}
\]

where $n = -i\omega$, $\omega$ is the complex phase velocity of the disturbance, $k^2 = k_x^2 + k_y^2$, $k$ is the magnitude of the total wave number, $i$ is the standard imaginary unit. Using Eqs. (52)-(55), the stress balance at the interface (Eq. (49)) is transformed as

\[
\left( [\varepsilon E_0^2] + \left[ \rho \right] \right) k^2 \zeta + \mu \left( \frac{d^2}{dz^2} + k^2 \right) w' + q_z \cdot \left[ \frac{dE_z}{dz} + [\varepsilon E_0 E_z] \right] = 0 \tag{56}
\]

\[2.4.7 \text{ Solutions and linearized boundary conditions}\]

According the boundary conditions and the properties of the perturbation, the boundary conditions for velocity (Eqs. (8) and (9)) and electric field (Eqs. (10) and (11)) can be described as follow

\[
\hat{w}^{(1)} = A \sinh k(z + \hat{h}_1) \tag{57}
\]

\[
\hat{w}^{(2)} = B \sinh k(z - \hat{h}_2) \tag{58}
\]

\[
\hat{E}_x^{(1)} = C_1 \exp(kz) + C_2 \exp(-kz) \tag{59}
\]

\[
\hat{E}_x^{(2)} = D_1 \exp(kz) + D_2 \exp(-kz) \tag{60}
\]

\[
\hat{E}_y = \frac{k_y}{k_x} E_x \tag{61}
\]

\[
\hat{E}_z = \frac{1}{ik_x} \frac{\partial E_x}{\partial z} \tag{62}
\]

Substituting Eqs. (57)-(62) into the boundary conditions of Eqs. (36)-(39), the boundary conditions transform into
At the interface, the continuity of velocities of perturbation for both fluids has the same values; this condition transforms into

\[
\hat{w}^{(1)}(-\bar{h}_1) = 0
\]  
\[
\hat{w}^{(2)}(\bar{h}_2) = 0
\]

\[
\hat{E}_x^{(1)}(-\bar{h}_1) = \hat{E}_y^{(1)}(-\bar{h}_1) = \hat{E}_z^{(1)}(-\bar{h}_1) = 0
\]
\[
\hat{E}_x^{(2)}(\bar{h}_2) = \hat{E}_y^{(2)}(\bar{h}_2) = \hat{E}_z^{(2)}(\bar{h}_2) = 0
\]

Combining Eqs. 44, 52 and 57, we can obtain that

\[
\frac{\hat{w}^{(1)}(0)}{n + u \cdot i k_x} = \frac{\hat{w}^{(1)}(0)}{m}
\]

where

\[
m = n + u \cdot i k_x
\]

Eqs. (46)-(48) describing the stress balance at the interface is transformed into the following form:

\[
\left[ \frac{d(\hat{E}_z(0))}{dz} + k^2 \hat{E}_0 \cdot \hat{w}(0) \right] = 0
\]

\[
\left[ \epsilon \hat{E}_z(0) \right] = 0
\]

\[
\left[ -\rho n + \mu \left( \frac{d^2}{dz} - 3k^2 \right) \right] \frac{d\hat{w}(0)}{dz} = 0
\]

\[
\left[ \epsilon E_{z0} + q_s \cdot [\hat{E}_z(0)] + \alpha k^2 \frac{\hat{w}^{(1)}(0)}{m} \right] = 0
\]

\[
\left[ \epsilon E_{z0}^2 + [P] + q_s \cdot [E_{z0}] \right] k^2 \frac{\hat{w}^{(1)}(0)}{m} + \left[ \mu \left( \frac{d^2}{dz} + k^2 \right) \hat{w}(0) \right]
\]

\[
+ q_s \cdot \left[ \frac{d\hat{E}_z(0)}{dz} \right] + \left[ \epsilon E_{z0} \frac{d\hat{E}_z(0)}{dz} \right] = 0
\]

2.5 *The principle of exchange of stabilities*

In Eq. (52), if \( n < 0 \), the disturbances decay exponentially to zero as time lapses. The disturbance cannot induce the instability of the interface and the two-fluid electroosmotic flow is stable. If \( n > 0 \), the disturbances advance exponentially to a large value as time lapses. The disturbance can be enlarged to induce the instability of the interface, and the two-fluid electroosmotic flow is unstable. According the boundary conditions, Eqs. (57)-(60) reduce to

\[
\hat{w}^{(1)}(z) = A \sinh k(z + \bar{h}_1)
\]
\[
\hat{w}^{(2)} = B \sinh k(z - \bar{h}_2) \\
\hat{E}_{x}^{(1)} = C \sinh k(z + \bar{h}_1) \\
\hat{E}_{x}^{(2)} = D \sinh k(z - \bar{h}_2)
\]

Substituting Eqs. (74)-(77) into Eqs. 70,72,73, the expression of \( n \) is shown using the following equation

\[
n = \frac{k^2}{m} (T_1 - T_2 + T_3 + T_4)
\]

where

\[
T_1 = \frac{2(\mu^{(1)} \coth kh_1 + \mu^{(2)} \coth kh_2)}{\rho^{(1)} \coth kh_1 + \rho^{(2)} \coth kh_2}
\]

\[
T_2 = \frac{\sigma k}{\rho^{(1)} \coth kh_1 + \rho^{(2)} \coth kh_2}
\]

\[
T_3 = \frac{(\varepsilon^{(1)} - \varepsilon^{(2)})^2 \cdot E_{20}^{(1)} E_{20}^{(2)}}{(\rho^{(1)} \coth kh_1 + \rho^{(2)} \coth kh_2) \cdot (\tanh kh_1 \cdot \varepsilon^{(2)} + \tanh kh_2 \cdot \varepsilon^{(1)})}
\]

\[
T_4 = \frac{(\rho^{(1)} \coth kh_1 + \rho^{(2)} \coth kh_2) \cdot (\tanh kh_2 \cdot \varepsilon^{(1)} + \tanh kh_1 \cdot \varepsilon^{(2)})}{E_{20}^{(1)} - E_{20}^{(2)}}
\]

\[
1 = \frac{2(\mu^{(1)} - \mu^{(2)})}{\rho^{(1)} \coth kh_1 + \rho^{(2)} \coth kh_2} \cdot \frac{k^2_{x} + k^2_{y}}{h_1/h_2}
\]

(84)

Term \( T_1 \) represents the effect of viscosity mismatch, term \( T_2 \) is the effect of surface tension force and term \( T_3 \) is the effect of the electric field. For a given set of stability parameters, the temporal evolution of each disturbance mode is governed by the sign of \( n \). \( n = 0 \), is the critical point of exchange of instability (marginal stable).

### 3 Materials and Methods

Fig. 2 shows the schematic of the fabricated microchannel used in the experiments. The adhesive lamination technique. [23] was used to fabricate the microchannel. The microchannel includes two PMMA plates (40 mm × 50 mm) and multiple layers of double-sided adhesive tape. The channel height is adjusted by the number of the adhesive layers. First, the inlets, outlets, holes for the electrodes and the alignment holes are cut through into the top PMMA layer and the bottom PMMA layer using a CO₂ laser, respectively. Next, the grooves for fixing the electrodes are cut in the top and bottom PMMA plates. Subsequently, the channel structures are cut into the adhesive tape (Arclad
8102 transfer adhesive, Adhesives Research Inc.). The electrodes platinum wires (Sigma-Aldrich, 0.1 mm) are then placed into the grooves, and lead out from the electrode opening in the top PMMA plate. The three layers are then bonded together. Finally, the openings for the electrodes are sealed with epoxy glue. The connectors for the inlets and outlets are also glued using epoxy. Using this method, a microchannel with a cross section of 1 mm × 50 μm was fabricated. In this channel, the electrodes are located at the two sides of the fluids and parallel to each other. The flows at the inlets I₁ and I₂ are driven by syringe pumps. The outlets O₁ and O₂ are connected to a waste reservoir.

The experimental setup includes three parts: recording subsystem, pressure source, and electroosmotic source. The recording subsystem of the experimental setup is similar to that described by Li et al. [24]. The pressure source includes two identical syringes (5 ml gastight, Hamilton) and a single syringe pump (Cole-Parmer, 74900-05, 0.2 μl/h to 500 l/h, accuracy of 0.5%). The syringes were driven by the pump to provide the constant flow rates.

A high voltage power supply (Model PS350, Stanford Research System, Inc) was used to provide the controlling electric field. The power supply is capable of producing up to 5000 V and changing the polarity of the voltage. The controlling electric field is applied to the platinum wires.

Aqueous NaHCO₃ (10⁻⁷ M) was used as the conducting fluid. Rhodamine B (C₂₈H₃₁N₂O₃Cl) was added into the aqueous NaHCO₃ (0.1 g/ml) as the fluorescent dye to achieve a distinct interface with fluorescence microscopy. Surfactant of Span 20 was mixed with the aqueous NaHCO₃ (w/w is 2%) to reduce the surface tension. The viscosity and conductivity of NaHCO₃ are 0.85 × 10⁻³ Ns/m² and 86.6 μS/cm, respectively. The viscosity of aqueous NaHCO₃ can be modified by mixing with glycerol (Sigma-Aldrich).

Different types of silicone oil (polydimethylsiloxane) were used as the non-conducting fluid. The surfactant of Span 80 was added to the fluid (w/w is 0.2%) to reduce the surface tension. The conductivity of silicone oil is 0.064 μS/cm; the viscosities of used silicone oil are 1.0 cSt, 5 cSt, and 10 cSt, respectively.

4 Validity of analytical model

Eq. (78) includes the effects of viscosity, surface tension, electric fields and surface charges on flow instability. Under the assumption of inviscid flows and zero interfacial surface charge [18], the effects of viscosity and surface charge are neglected, Eq. (78) can be simplified into

\[
n = \frac{k^2}{m} \left( -\frac{\sigma k}{\rho^{(1)} \coth kh_1 + \rho^{(2)} \coth kh_2} \right) \left( \epsilon^{(1)} - \epsilon^{(2)} \right) \frac{E_{10}^{(1)} E_{20}^{(2)}}{(\rho^{(1)} \coth kh_1 + \rho^{(2)} \coth kh_2) \left( \tanh kh_1 \epsilon^{(2)} + \tanh kh_2 \epsilon^{(1)} \right)} \right)
\]
When the fluids are static, Eq. (68) is simplified into

$$\zeta = \frac{\bar{w}(1)(0)}{n}$$

and

$$m = n$$

Combining Eqs. (85) and (87), Eq. (85) becomes

$$n^2 = k^2 \left( -\frac{\sigma k}{\rho(1) \coth k h_1 + \rho(2) \coth k h_2} + \left( \frac{(\rho(1) - \rho(2))^2}{(\rho(1) \coth k h_1 + \rho(2) \coth k h_2) \left( \tanh k h_1 \rho(2) + \tanh k h_2 \rho(1) \right)} \right) \right)$$

Eq. (88) consider only the effects of surface tension and electric field, which is identical to that presented by Goranović [18].

A second validation was performed by running the case using the same parameters as in the analysis of Khomami and Su [25]. For pressure driven two immiscible fluid flows in microchannel without the application of electric field and surface tension, Eq. (78) reduces into

$$n = \frac{2k^2}{m} \left( \frac{\mu(1) \coth k h_1 + \mu(2) \coth k h_2}{\rho(1) \coth k h_1 + \rho(2) \coth k h_2} \right)$$

where

$$\frac{1}{m} = \frac{2 \left( \mu(2) - \mu(1) \right)}{\left( P_1 - P_2 \right) + \left( k^2 - \mu(1)^2 / \mu(2)^2 \right) \frac{h_1}{h_2}}$$

with the flow parameters $\mu(1)/\mu(2) = 0.203$, $h_1/h_2 = 4.875$ is obtained from Khomami and Su [25]. Fig 3 compares the theoretically calculated growth rate $n$ in Eq. (89) and experimentally measured by Khomami and Su [25], and the updated results by Theofanous et al. [26]. The results show that the proposed model can predict the flow instability under the influence of viscosity reasonably well.

5 Results and discussion

5.1 Instability phenomenon

Figure 4 shows the observed flow pattern at various values of electric field. Two immiscible fluids, aqueous NaHCO$_3$ (high electrical mobility, $\mu(1) = 0.85 \times 10^{-3}$ Ns/m$^2$) and silicone oil, low electrical mobility, $\mu(2) = 4.25 \times 10^{-3}$ Ns/m$^2$) are introduced into the microchannel at $q_1 = q_2 = 0.05$ ml/h. Fig. 4(a) shows a stable flow field using relative low
voltage of 10 kV/m. When a higher normal electric field is added, we noted a slightly unstable flow at a normal electric field of $E_{\text{critical}} = 110$ kV/m (Fig. 4b). The threshold condition ($E_{\text{critical}}$) was determined by the lowest voltage at which the flow behavior was visibly different from the base static condition. Unstable flow was observed at a larger magnitude of electric field (Fig. 4(c)).

Figure 5 shows that the growth rate $n$ depends on the relative magnitude of the relation in Eq. (78). The flow conditions are $q_1 = q_2 = 0.05$ ml/h, $\varepsilon_{r1} = 80$, $w = 0.05$ mm, $\zeta_1 = \zeta_2 = 24.7$ mV, $\zeta_3 = 40.4$ mV, $\mu^{(1)} = 0.85 \times 10^{-3}$ Ns/m², $\mu^{(2)} = 4.25 \times 10^{-3}$ Ns/m², $\sigma = 1.8 \times 10^{-4}$ N/m, and $E_z$ varying from -200kV/m to 200kV/m.

The term of the viscosity mismatch $\frac{\mu^{(1)} \coth k h_1 + \mu^{(2)} \coth k h_2}{\rho^{(1)} \coth k h_1 + \rho^{(2)} \coth k h_2}$ has a destabilizing effect as $T_1 > 0$ (Fig. 5). The viscosity mismatch leads to a velocity mismatch at the perturbed interface which causes viscous instability [27].

The term of the surface tension, $\frac{\sigma k}{\rho^{(1)} \coth k h_1 + \rho^{(2)} \coth k h_2}$, has a stabilizing effect as shown by a negative growth rate ($n < 0$) in Fig. 5. This conclusion is consistent with the findings reported by Moatimid et al. [17] and Hoper et al. [28].

The influence of the electric field and surface charge, which may either stabilize or destabilize the interface, depends on the sign of the term of $\left(\varepsilon^{(1)} - \varepsilon^{(2)}\right)^2 \cdot E_{z0}^{(1)} E_{z0}^{(2)}$ and $q_b \left(\varepsilon^{(2)} - \varepsilon^{(1)}\right) \cdot \left(E_{z0}^{(1)} - E_{z0}^{(2)}\right)$, respectively.

The results clearly show that the orders of magnitude of the four terms are comparable. When the electric field reaches $E_{\text{critical}}$, the perturb ripple wave arises at the interface. As the electric field increases, the destabilizing factor of electric field and surface charge increases, and the flow becomes unstable.

5.2. Electrohydrodynamic instability of interface under the combined effect of electroosmotic flow and pressure gradient

5.2.1 Effect of the viscosity

Fig 6(a) compares the theoretically critical electric field and experimentally measured value. The analytical results agree well with the experiments.

In order to understand the effect of viscosity of the high electrical mobility fluid 1, $\mu^{(1)}$ on the instability of the interface, we fixed the channel size, flow rates, electrical properties and viscosity of fluid 2, and vary the electric field until it reached the critical electric field ($E_{\text{critical}}$) when the marginal stable flow pattern was observed. Fig. 6(b) shows the variations of growth rate and the critical electric field, respectively, with viscosity of the conducting liquid 1. The flow conditions are $q_1 = q_2 = 0.05$ ml/h, $\varepsilon_{r1} = 80$,  

w = 0.05 mm, \( \zeta_1 = \zeta_2 = 24.7 \text{ mV} \), \( \zeta_3 = 40.4 \text{ mV} \), \( \mu^{(2)} = 4.25 \times 10^{-3} \text{ Ns/m}^2 \), \( \sigma = 1.8 \times 10^{-4} \text{ N/m} \), and \( \mu^{(1)} \) varying from \( 0.85 \times 10^{-3} \text{ Ns/m}^2 \) to \( 2.8 \times 10^{-3} \text{ Ns/m}^2 \). The results indicate that for a given channel geometry and flow conditions, the critical electric field increases with increasing viscosity of the high electrical mobility fluid 1. The viscosity of fluid 1 has a stabilizing effect on the flow as the growth rate decreases with increasing \( \mu^{(1)} \).

5.2.2 Effect of the flow rates

Fig 7 (a) compares the theoretically critical electric field and experimentally measured value. The analytical results agree well with the experiments.

Fig 7 (b) shows the effect of different flow rates on the critical electric field and growth rate, respectively, over a range of \( \mu^{(1)} \). We now fix the width of the microchannel \( (w = 0.05 \text{ mm}) \), and the fluid properties \( (\varepsilon_r = 80, \zeta_1 = \zeta_2 = 24.7 \text{ mV}, \zeta_3 = 0 \text{ mV}, \sigma = 1.8 \times 10^{-4} \text{ N/m}, \mu^{(2)} = 4.25 \times 10^{-3} \text{ Ns/m}^2, \mu^{(1)} = 0.85 \times 10^{-3} \text{ Ns/m}^2) \), and vary the flow rates from 0.05 ml/h to 0.2 ml/h. The results show that for a given microchannel geometry and flow properties, \( E_{\text{critical}} \) increases with the increasing of the inlet flow rates, thus the increase of flow rates has a stabilizing effect on the interface between the immiscible fluids.

5.2.3 The effect of the width (h) of the microchannel

Fig 8(a) compares the theoretically critical electric field and experimentally measured value. The analytical results agree well with the experiments.

Fig 8 (b) shows the effect of the width of the microchannel on the critical electric field and growth rates. We now fix flow rates \( (q_1 = q_2 = 0.05 \text{ ml/h}) \), and the fluid properties \( (\varepsilon_r = 80, \zeta_1 = \zeta_2 = 24.7 \text{ mV}, \zeta_3 = 0 \text{ mV}, \sigma = 1.8 \times 10^{-4} \text{ N/m}, \mu^{(2)} = 4.25 \times 10^{-3} \text{ Ns/m}^2, \mu^{(1)} = 0.85 \times 10^{-3} \text{ Ns/m}^2) \), and vary the width of the microchannel from 50 \( \mu \text{m} \) to 200 \( \mu \text{m} \). The results show that for a given inlet flow rates, the growth rate \( n \) increases with increasing width, thus the increase of the width of the microchannel has a destabilizing effect to the interface between the immiscible fluids.

Under fixed inlet flow rates, the velocity of fluids decreases with increasing width. An electric field \( E_{\text{critical}} \) with lower magnitude is needed.

6 Conclusions

This paper reports the electrohydrodynamic instability of the interface between immiscible fluids under the combined effect of hydrodynamics and electroosmosis in a microchannel. The effect of different parameters such as electric field, viscosity, flow rate, and dimension of the channel were studied using an analytical model and validated by experiments.

In the analytical analysis, the electric field and fluid dynamics are coupled only at the interface through the balance equations of the tangential and normal interfacial stress...
under the coupled effect of hydrodynamic and electroosmosis. In the experiments, two immiscible fluids, Aqueous NaHCO₃ (high electrical mobility fluid) and silicone oil (low electrical mobility fluid) are introduced into the PMMA microchannel using a syringe pump. The normal electric field is added to the aqueous NaHCO₃ using a high voltage power supply. The results are recorded using a CCD camera.

The results showed that the external electric field and decreasing width of the microchannel have destabilizing effect to the interface between immiscible fluids. At the same time, the viscosity of the high electrical mobility fluid and flow rates of fluids have stabilized effect.

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References


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Fig. 1
Fig. 2
Fig. 3
Fig. 4

(a) Silicone oil ($q_2$)

Interface

Aqueous NaHCO$_3$($q_1$)

Flow direction ($q_1=q_2$)

$E_z=10$kV/m

(b) Ripple wave

Flow direction ($q_1=q_2$)

$E_z=110$kV/m

(c) 

Flow direction ($q_1=q_2$)

$E_z=150$kV/m
Fig. 6
Fig. 7

(a) Electric field (kV/m) vs. Flow rate (ml/h)

(b) Growth rate $n$ vs. Viscosity of fluid 1 $\mu$ (Pa.s)

- $q_1 = q_2 = 0.05$ ml/h
- $q_1 = q_2 = 0.1$ ml/h
- $q_1 = q_2 = 0.15$ ml/h
- $q_1 = q_2 = 0.2$ ml/h

$n > 0$ (unstable)
$n < 0$ (stable)
Fig. 8