



# Electroosmotic flow of a two-layer fluid in a slit channel with gradually varying wall shape and zeta potential

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## ABSTRACT

This study aims to investigate electroosmotic (EO) flow of a two-layer fluid through a slit microchannel where the wall shape as well as zeta potential may vary slowly and periodically with axial position. The two-layer EO flow is a model for the flow of two immiscible fluids: a non-conducting working fluid being dragged into motion by a conducting sheath fluid. Electric double layers may develop in the conducting fluid near the interface between the two fluids, and near a wall that is assumed to be non-uniform in both shape and zeta potential distribution. Because of these geometrical and electrical non-uniformities, pressure is internally induced. The two-fluid flow is therefore driven by electrokinetic and hydrodynamic forcings, while subjected to the combined effects of the axial variations of the wall shape and potential distribution. The present problem is formulated by invoking the lubrication approximation, for a nearly parallel flow of low Reynolds number, and the governing equations are solved as analytically as possible. The induced pressure gradient and the deformed shape of the interface, which are functions of axial position, as well as the flow rates of the two fluids, are determined by an iterative trial-and-error numerical scheme. Results are generated to show the effects due to various factors, including the applied pressure difference, interfacial potential, viscosity ratio, wall undulation, and phase shift between the wall shape and potential distribution. Some of the effects on flow in a non-uniform channel can be qualitatively different from that in a uniform channel.

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## 1. Introduction

Electrokinetic pumping has now been widely used in the transport of fluid in microfluidic devices. Under an applied electric field, fluid is driven into motion through Lorentz force acting on unbalanced free ions in an electric double layer (EDL) that develops in the vicinity of a solid-liquid interface. The resulting flow, known as electroosmotic (EO) flow, offers some outstanding features. First, the velocity profile in an EO flow is nearly plug-like, ensuring a low sample dispersion [1] and avoiding mixing and rheological changes produced by lateral velocity variations [2]. Second, the section-mean velocity is almost independent of the channel size, which is desirable for fluid transport in an ultrafine channel. Other advantages include a more precise control of flow magnitude and direction, few mechanical moving parts (thus less mechanical failures), easy fabrication, high reliability, little noise, and so on. These advantages have made EO a promising pumping mechanism for a

wide range of applications, ranging from drug delivery [3] to chip cooling [4].

EO pumping can be applied not only to single fluids, but also to a two-layer fluid [5,6]. Some non-polar fluids with very low ionic conductivity, such as oil, ethanol and organic solvents, do not develop EDLs [7], and hence cannot be pumped by EO forcing directly. Nevertheless, EO effect can serve as an indirect actuation mechanism to pull non-polar fluids into motion via some sheath liquid with significant ionic conductivity. This idea was practically realized by Brask et al. [8] and Watanabe et al. [9]. Pioneering theoretical studies were conducted by Gao et al. [5] and Ngoma and Erchiqui [7], who looked into steady two-layer EO flows in a rectangular microchannel and between two parallel plates. These authors, however, ignored electrostatic contribution at the liquid-liquid interface. The effect of Maxwell stress, which is caused by the interaction between free charges on a liquid-liquid interface and the externally applied electric field, is found to be significant and has to be taken into consideration [10,11]. This has motivated Gao et al. [12] to incorporate the Maxwell stress into the interfacial condition in their study on transient two-layer EO flow. Li et al. [13,14] further proposed a three-layer-fluid model for EO flow of

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a layer of non-conducting fluid bounded by two layers of conducting fluid in a rectangular microchannel. Liu et al. [15] developed analytical models for two-layer EO flow in a circular microchannel, where Newtonian–Newtonian and Newtonian–Casson fluids were examined. The studies by Choi et al. [16] and Gaikwad et al. [17] were focused on the zeta-potential jump and the interfacial hydrodynamic slip on the liquid–liquid interface for two-layer EO flow in a parallel-plate channel.

The above-mentioned theoretical studies have only considered uniform zeta potential at flat walls, or a plane interface between two immiscible fluids. Mandal et al. [18] analyzed two-layer EO flow between two parallel plates in the presence of axially varying zeta potentials. They obtained analytical solutions for small deformation of the interface through an asymptotic analysis, and applied the phase field formalism to the case of large interfacial deformation. In addition to surface charge modulation, which itself can offer a wealth of interesting flow patterns, the problem will become more interesting when wall undulation is considered as well. Ajdari [19,20] found that the combined effect of these two wall patterns can generate a net flow even when the walls are on average electro-neutral. Axially modulated zeta potentials with a zero average alone can only give rise to recirculation rolls, but not a net flow since the flux/reflux caused by equal positive/negative surface charges will exactly balance each other. A net flow may happen only when this symmetry is broken by the superposition of undulated walls. Chen and Cho [21] further investigated steady-state mixing characteristics for EO flow with heterogeneous charge patterns on wavy surfaces. Their numerical results suggest that the mixing performance can be remarkably improved by increasing either the wave amplitude or the length of the wavy surface or the magnitude of the heterogeneous zeta potential. Other authors who also looked into the combined effect of wall shape and charge patterns on EO flow include, among others, Xuan and Li [22], Bhattacharyya and Nayak [23], Ng and Qi [24], Bhattacharyya and Bera [25], and Yoshida et al. [26]. All these studies, however, only considered single-fluid EO flow.

Motivated by previous studies on the interaction between wall undulation and charge modulation, this paper aims to look into such interaction in a two-fluid configuration of EO flow. In order to enable the present problem to be solved as analytically as possible, we shall adopt the lubrication approximation [27,28] to simplify the problem. The lubrication technique has been utilized previously by various authors to deal with EO flow in non-uniform channels; examples include Long et al. [29] and Ajdari [30]. For EO flow in microchannels with arbitrary cross-sectional geometry and arbitrary surface charge distribution, Ghosal [31] obtained asymptotic solutions with the aid of the lubrication approximation. Essentially this approximation is to reduce two-dimensional flow to quasi-one-dimensional flow. Its validity is based on two conditions. One condition is a sharp contrast in length scales for much slower variations in the axial than the transverse directions. Another condition requires the Reynolds number to be sufficiently small for negligible inertia.

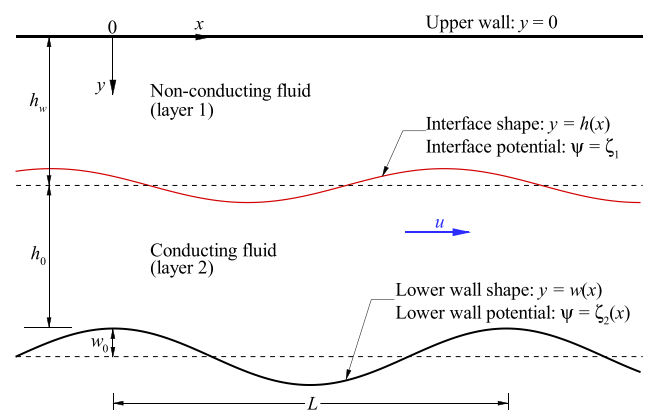
For EO flow of two immiscible fluids in a uniform channel, the liquid–liquid interface is a flat surface [5–7,12–17]. If axial variations in wall shape and/or wall charge are allowed, the interface between the two immiscible fluids will change in shape as a function of axial position. The deformed shape of the interface is part of the flow structure, and hence has to be found as part of the solution. In the present study, the lower wall of a slit channel is assumed to be patterned with periodic variations in both wall shape and wall potential, and these two periodic functions, one geometrical and one electrical, have the same wavelength but with a possible phase shift between them. The displacement of the interface, under the combined effects due to wall undulation and charge modulation, may give rise to a variety of interesting fea-

tures. As shown by Mandal et al. [18], a strongly deformed interface is usually associated with a complex flow structure, where the deformation can be affected by fluid properties, such as viscosities and permittivities of the two fluids. These authors also found that an increase in the wavelength of wall charge modulation may result in significant augmentation of the interface deformation. These previous findings have guided us to define the scope of the present problem, which is formulated such that it can be solved as analytically as possible, and yet it can reveal the effects of various geometrical, electrical, and hydrodynamic factors on a two-layer EO flow.

Our problem is described in further detail in Section 2. On the basis of the lubrication approximation (i.e., a nearly parallel flow with negligible inertia), the Poisson–Boltzmann and momentum equations are formally solved for the potential distribution and axial velocity for the two fluids. The special case for two-layer EO flow through a uniform channel is presented in Section 3. An iterative trial-and-error numerical scheme, which is used to determine the pressure gradient and interface shape as functions of axial position, is then introduced in Section 4. Numerical results are presented in Section 5, where we examine how different parameters (including the ratio of viscosities, applied pressure drop, interfacial potential, and parameters pertaining to the wall patterns) may have various effects on the two-fluid EO flow.

## 2. Problem formulation

Our problem is to investigate EO flow of a two-layer fluid in a non-uniform slit microchannel, where the two fluids are of distinct chemical and physical properties and separated by a sharp interface. The channel is non-uniform in both height and wall potential distribution, which may vary slowly and periodically with axial position. Fig. 1 depicts a definition sketch of our problem, where the axial  $x$ -axis is positioned along the upper planar wall, and the transverse  $y$ -axis points vertically downward. The lower wall has a wavy shape given by  $y = w(x)$ , and a wall potential given by  $\zeta_2 = \zeta_2(x)$ . These two functions are periodic functions of  $x$  with the same wavelength, denoted by  $L$ . The amplitude of the wavy wall is  $w_0$  (or the bottom groove depth is  $2w_0$ ). The zeta potential at the liquid–liquid interface is a known constant, denoted by  $\zeta_1$ . The flow is driven under the combined action of a pressure gradient,  $\Delta P/L$ , and an electric field,  $E_x$ , which are applied externally along the  $x$ -direction. It is also assumed that the lower fluid is conducting while the upper fluid is non-conducting. In other words,



**Fig. 1.** Definition sketch of the present problem: two-fluid electroosmotic flow in a slit channel with gradually varying shape and potential on the lower wall, where the upper fluid is non-conducting and the lower fluid is conducting. The upper dashed line denotes the mean position of the interface between the two fluids, and the lower dashed line denotes the mean position of the lower wall.

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