

A Variational Model for Two-Phase Immiscible Electroosmotic Flow at Solid Surfaces

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Abstract. We develop a continuum hydrodynamic model for two-phase immiscible flows that involve electroosmotic effect in an electrolyte and moving contact line at solid surfaces. The model is derived through a variational approach based on the Onsager principle of minimum energy dissipation. This approach was first presented in the derivation of a continuum hydrodynamic model for moving contact line in neutral two-phase immiscible flows (Qian, Wang, and Sheng, *J. Fluid Mech.* 564, 333–360 (2006)). Physically, the electroosmotic effect can be formulated by the Onsager principle as well in the linear response regime. Therefore, the same variational approach is applied here to the derivation of the continuum hydrodynamic model for charged two-phase immiscible flows where one fluid component is an electrolyte exhibiting electroosmotic effect on a charged surface. A phase field is employed to model the diffuse interface between two immiscible fluid components, one being the electrolyte and the other a nonconductive fluid, both allowed to slip at solid surfaces. Our model consists of the incompressible Navier-Stokes equation for momentum transport, the Nernst-Planck equation for ion transport, the Cahn-Hilliard phase-field equation for interface motion, and the Poisson equation for electric potential, along with all the necessary boundary conditions. In particular, all the dynamic boundary conditions at solid surfaces, including the generalized Navier boundary condition for slip, are derived together with the equations of motion in the bulk region. Numerical examples in two-dimensional space, which involve overlapped electric double layer fields, have been presented to demonstrate the validity and applicability of the model, and a few salient features of the two-phase immiscible electroosmotic flows at solid surface. The wall slip in the vicinity of moving contact line and the Smoluchowski slip in the electric double layer are both investigated.

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1 Introduction

Recently, there have been rapid developments in the design, patterning, and utilization of microfluidic and nanofluidic devices which have found many applications in the devised transport, separation, identification, synthesis, and manipulation of a wide range of chemical and biological species [1]. In this respect, electroosmotic flows have been widely used to transport and mix fluids using electric fields in micro- and nanofluidic systems based on the lab-on-a-chip concept [2,3]. The use of mechanical pumps or valves with moving components is therefore avoided. When a solid wall is in contact with an electrolyte solution, it acquires a certain amount of charge on the surface through an electrochemical adsorption/desorption process, while the counterions are released from the solid surface into the solution. The electric double layer (EDL) is formed through an accumulation of the counterions in the solution adjacent to the wall. These excess counterions move under an external electric field and drag the fluid surrounding them. This electrokinetic effect causes a flow, termed as electroosmotic flow (EOF), in the bulk region via the viscous coupling. If the characteristic thickness of the EDL (i.e., the Debye length λ_D) is much smaller than the channel width (diameter), then the EOF will exhibit the plug-flow velocity profile, which is very different from the parabolic velocity profile in conventional pressure driven flows. Generally speaking, EOF is preferred for biomedical/chemical separation or detection applications since the plug-flow velocity profile reduces the sample dispersion effect, and consequently improve the device performance [4,5].

The fluid dynamics in confined geometries can be affected by the friction with confining walls. Theoretically, wall friction is quantified by setting some boundary conditions at the wall. The no-slip boundary condition, which states that there is no relative motion at the fluid-solid interface, has been extensively used and demonstrated in numerous macroscopic flows. However, over the past two decades, experiments have shown that slip occurs in mechanically driven flows over smooth solvophobic surfaces, with slip lengths typically of the order of nanometer [6]. Slip effects become increasingly important as the confinement space for fluids is reduced to sub-micrometer or nanometer scale. The flow enhancement due to slip could be up to two or three orders of magnitude [7,8]. Excellent reviews on the experimental and theoretical aspects of liquid slip can be found in [6,9]. This constitutes the first reason to explicitly take into account the wall slip in this work.

Research interests in the dynamics of two-phase fluids in narrow channels stem from many applications that microfluidic and nanofluidic devices have found in biological and chemical analyses, drug delivery, and chemical synthesis [10]. In particular, droplets of one fluid in the other immiscible fluid have been found useful in a wide range of applications, especially when the droplet size and the size distribution can be prescribed on a micro- or nanoscale [11]. Droplet generation, manipulation, and delivery within micro and nano systems have been extensively studied experimentally and numerically [12,13]. The flow phenomena in these applications constantly involve a classical problem in con-