Electrohydrodynamic pumping in microsystems

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Abstract. The physical principles behind the electrohydrodynamic (EHD) actuation in microsystems is presented by reviewing five different EHD micropumps. These are classified into two groups: micropumps that exert electric forces in the liquid bulk and micropumps that exert forces in the diffuse double layer. This review of five EHD micropumps allows us to analyse the EHD actuation ranging from very insulating liquids to electrolytic solutions.

1. Introduction

Among the most promising techniques to handle small liquid volumes in microdevices are those that employ electrical forces, which have the advantages of voltage-based control and dominance over other forces. Microfluidic transport is sometimes achieved by using macroscale pumps such as syringe pumps or pressure/vacuum chambers and valves. However, many applications would benefit from an on-chip active micropump: dispensing therapeutic agents into the body, cooling of microelectronic devices, dispensing liquids in miniature systems for chemical and biological analysis, and more. Here, we are going to deal with dynamic micropumps that exert electric forces on liquids, called electrohydrodynamic (EHD) micropumps. EHD micropumps can work as on-chip active micropumps with no moving parts.

EHD dynamic micropumps may be classified into: a) pumps that exert electric forces in the liquid bulk: the EHD injection, conduction and induction pumps; and b) those that exert electric forces in the diffuse double layer: the electroosmotic and AC induced-charge electroosmotic pumps. The main goal of the present work is to describe the physical principles behind the EHD actuation in microsystems.

2. Common features of EHD Micropumps

The electrical current density in the liquid bulk can be written as the sum of electromigration, diffusion and convection:

\[ j = \sum_i q_i (n_i \mu_i E - D_i \nabla n_i + n_i u) \] (1)

where \( D_i \) and \( \mu_i \) are the diffusion and mobility coefficients of an ion, \( q_i \) its charge, \( n_i \) the ion density, and \( u \) the velocity of the fluid. Species conservation equations for ions and molecules must be included to complete the description of the system [1]:

\[ \frac{\partial n_i}{\partial t} + \nabla \cdot (n_i \mu_i E - D_i \nabla n_i + n_i u) = r_i. \] (2)
Here $r_i$ is the rate of production due to chemical reactions ($\mu_i = 0$ for neutral species).

Working with fluids with conductivities smaller than 10 S/m, with applied ac signals of frequencies up to 1 GHz, the electric fields in microsystems hold the quasi-electrostatic limit of Maxwell equations [1; 2]:

$$\nabla \cdot (\varepsilon \mathbf{E}) = \rho; \quad \nabla \times \mathbf{E} = 0; \quad \nabla \cdot \mathbf{j} + \frac{\partial \rho}{\partial t} = 0. \quad (3)$$

The liquid motion is governed by the Navier-Stokes equations which, under steady-state conditions in microfluidics and in the limit of low Reynolds number, can be written:

$$\nabla \cdot \mathbf{u} = 0; \quad 0 = -\nabla p + \eta \nabla^2 \mathbf{u} + \mathbf{f}. \quad (4)$$

Joule heating is an important consideration in EHD systems. In many situations in microsystems, heat convection is small compared to heat diffusion and the temperature equation in the steady-state reduces to

$$0 = k \nabla^2 T + \mathbf{E} \cdot \mathbf{j}' \quad (5)$$

where $\mathbf{j}'$ is the current density measured in the rest frame of the fluid element.

We are interested in cases where the main body force density is of electrical origin. The electric volume force density is [1]

$$\mathbf{f}_E = \rho \mathbf{E} - \frac{1}{2} \mathbf{E}^2 \varepsilon + \frac{1}{2} \nabla \left[ \mathbf{E}^2 \rho_m \left( \frac{\partial \varepsilon}{\partial \rho_m} \right)_T \right] \quad (6)$$

where the first term is the Coulomb force, the second term is the dielectric force, and the third is the electrostriction, which can be incorporated into a redefined pressure for an incompressible fluid. The Coulomb force is the term primarily responsible for generating pumping in Electrohydrodynamics.

3. Micropumps that exert forces in the liquid bulk

For the following EHD pumps we will analyse the geometry depicted in figure 1: Two perforated electrodes separated a distance $L$, subjected to a potential difference $V$, are inside a pipe of diameter $D$ filled with the working liquid. Using a one-dimensional analysis, the total electric force per unit area exerted on the liquid (the generated pressure) is

$$\Delta p_g = \int_0^L \left( \rho \mathbf{E} - \frac{\mathbf{E}^2}{2} \frac{\partial \varepsilon}{\partial x} \right) dx = \int_0^L \frac{1}{2} \frac{\partial \varepsilon}{\partial x} \mathbf{E}^2 dx = \frac{\varepsilon \mathbf{E}^2}{2} \bigg|_{x=L} - \frac{\varepsilon \mathbf{E}^2}{2} \bigg|_{x=0} \quad (7)$$

where we have used that $\rho = \partial(\varepsilon \mathbf{E})/\partial x$. The expression says that the generated pressure is equal to the jump in electric energy density from $x = 0$ to $x = L$.

3.1. Injection pump

The injection pump is also known as ion-drag pump. Ions emitted from one electrode (the emitter) are collected in the other electrode (the collector). Since ions do not accelerate but move at the terminal velocity $v = \mu E$ (ion inertia is negligible) the electric force acting on them is transmitted directly to the liquid by viscous friction. Therefore, in the bulk we have $f = \rho \mathbf{E}$. The typical working liquid is a very insulating liquid. If the liquid has some residual conductivity $\sigma$, we expect that EHD operation is satisfactory if the time required from an ion to go from emitter to collector is shorter than the charge relaxation time, i.e. $L/\mu E < \varepsilon/\sigma$. To ensure that the ions are injected from the emitter rather than from the collector, the local electric field at the emitter is increased due to presence of sharp edges or points.
For dielectric liquids the ratio of diffusion to electromigration current is negligible to a very good approximation. In microsystems, fluid velocities are commonly on the order of 1 mm/s or less and this implies that convection of charge is also negligible compared to electromigration. Under these approximations and considering unipolar injection, the generated pressure in the regime of space-charge-limited emission (SCLE) is \( \Delta p_g = (9/8)\varepsilon V^2/L^2 \) [2].

In microsystems, Richter and Sandmier [3] fabricated ion-drag micropumps made of two opposite metallic grids through which the pumped fluid moves. Ion-drag micropumps made of co-planar microelectrodes have been studied by Ahn and Kim [4] and Darabi and Wang [5]. Kazemi et al [6] studied an injection micropump made of 3D microelectrodes (metallic pillars) and HFE-7100 as working fluid. A maximum pressure of 2240 Pa was generated at an applied voltage of 900 V.

### 3.2. Conduction pump

This kind of EHD actuation is based upon the Coulomb force on non-equilibrium charged layers adjacent to electrodes that appear in the conduction regime. The representative working fluid is a semi-insulating liquid. For electric fields less than \( 10^7 \) V/m, ions generated in the bulk from dissociation of neutral species move by electromigration towards the electrodes where they discharge. Near the electrodes, charged layers appear of opposite sign to that of the adjacent electrode and are called heterocharge layers [7].

A good picture is to consider a bipolar conduction model where a neutral species \( n_0 \) dissociates reversibly (at a rate \( k_{d,n_0} \)) into univalent positive \( n_+ \) and negative \( n_- \) ions (that recombine at a rate \( k_{r,n_+n_-} \)) [7; 8]. Counter-ions arrive at the electrode and discharge on it but co-ions cannot come from the electrode below the electric field threshold for charge emission. This implies that near the positive (negative) electrode the majority of ions are negative (positive), and that there is an electric force directed towards each electrode. The thickness of this charge layer, \( \lambda \), can be estimated for the positive electrode as follows. At \( x = 0 \) the flux of positive ions is zero (\( \mu_+ n_+ E = 0 \)) and at \( x = \lambda \) it reaches the value in the bulk (\( \mu_+ n_+^{eq} E_{bulk} \)). This jump in positive ion flux should be on the order of the ions produced per unit of time in that distance (\( k_{d,n_0} \lambda \)).

Each electrode is subjected to an electric pressure from the bulk side. The total generated pressure for a geometry like the one depicted in figure 1 is \( \Delta p_g = (\varepsilon E_{2}^{2}/2)((\mu_-/\mu_+) - (\mu_+/\mu_-)) \) [2]. In order to have a pressure difference even in the case of equal mobilities, the electrodes are made nonsymmetric [7; 8] (see an example in figure 2).

EHD conduction pumping has been mainly reported in the millimeter scale [7; 9]. Pearson and Seyed-Yagoobi [10] reported recently a micro-scale EHD conduction pump. Working with refrigerant HCFC-123 at 4 kV they obtained \( \Delta p_g \sim 500 \) Pa.

In order to increase the generated pressure, we can increase the voltage or reduce the dimensions, however, we can then reach either the saturation current regimen or the charge injection regime. In some configurations, the appearance of charge injection is manifested by the liquid changing direction.
3.3. Induction pump

Induction pumping is based on the forces upon charges induced in a liquid by the electric field due to a gradient or discontinuity of the electric conductivity. Gradients in conductivity are usually obtained by imposing a gradient of temperature. The common induction pump consists of an array of electrodes that generate a traveling-wave potential together with a perpendicular gradient of temperature [11]. The travelling-wave electric field attracts or repels the induced charges, leading to fluid flow. The temperature gradient can be imposed externally or it can be obtained by Joule heating generated by the imposed electric field. In microsystems, the EHD induction micropumps fabricated by Fuhr and co-workers [12; 13] employed Joule heating to establish the temperature gradient. Water saline solutions were used as working fluids, which are very difficult (if not impossible) to be pumped using EHD induction at the macroscale. Unidirectional motion can also be obtained with electrodes subjected to a single phase ac signal with an imposed longitudinal temperature gradient [14]. An induction pump can be fabricated with two parallel perforated electrodes subjected to both a potential difference and a temperature difference (geometry like in figure 1 where we impose a temperature \( T_0 \) at \( x = 0 \) and \( T_L \) at \( x = L \)). The charge induced is obtained from \( \rho = \nabla \cdot (\varepsilon \mathbf{E}) = \nabla \cdot (\varepsilon \mathbf{J}/\sigma) = \sigma \mathbf{E} \cdot \nabla (\varepsilon/\sigma) \). In an induction pump like this, the Coulomb force is directed from high to low conductivity. It can be shown that the generated pressure for small increments of temperature is \( \Delta p_g = (\varepsilon V_0^2/2L^2)(2\alpha - \beta)(T_0 - T_L) \) [2], where \( \alpha = (1/\sigma)(d\sigma/dT) \) and \( \beta = (1/\varepsilon)(d\varepsilon/dT) \). For water saline solutions, \( \alpha = 0.02 \text{ K}^{-1} \) and \( \beta = -0.004 \text{ K}^{-1} \) at room temperature, and the force goes from high to low temperature.

Stubbe et al. [14] fabricated an induction micropump using a single phase ac signal in the frequency range 0.3-50 MHz and voltages in the range 0-30 V. They employed aqueous solutions of conductivity in the range \( \sigma = 0.1-1.5 \text{ S/m} \) and observed velocities up to 120 \( \mu \text{m/s} \).

4. Micropumps that exert forces in the double layer

Electrical double layers are formed at solid/liquid interfaces spontaneously because of differences in electrochemical potentials between the solid and liquid phases [15]. The solid surface becomes charged and ions in solution of opposite charge (counterions) are attracted until they screen the electric field created by the surface charge. At equilibrium, the liquid is electroneutral except for a charged layer adjacent to the solid surface, with characteristic thickness given by the Debye length, \( \lambda_D \) [15]. We can distinguish to ways of double layer generation at the solid/liquid interface: (a) a chemical mechanism such as ionization of bound surface groups or ion adsorption; (b) an electrostatic mechanism in which a solid metal surface becomes charged because is subjected to a potential difference with respect to the liquid. We can identify two kinds of pumps based on these charging mechanisms.

4.1. Electroosmotic pump

An example of a spontaneous double layer is the case of silica-based ceramics, like glass, with Si–OH groups at the surface. In contact with water a fraction of the Si–OH bonds change into Si–O– releasing H\(^+\). Therefore, we have a negative surface charge balanced by positive counterions. When an electric field is applied along the surface, the Coulomb force causes the motion of the mobile ions of the double layer dragging the fluid with them. This is called electroosmosis (EO) and has been known for 200 years (F.F. Reuss discovered electroosmosis in 1809). The EO velocity generated in the double layer is seen as a superficial or slip velocity since the thickness of the Debye layer is very small (1-100 nm in water). It is given by the Helmholtz-Smoluchowski expression [15]: \( \nu_{eo} = \varepsilon\zeta E/\eta \), where \( \zeta \) is the potential difference between the plane of non-slip and the bulk. The pressure difference that can be generated in a capillary of diameter \( D \) and length \( L \) for \( \lambda_D \ll D \) is \( \Delta p_g = 3\varepsilon\zeta E L/2D^2 \) [2]. In miniaturized systems for chemistry and life
4.2. AC induced charge electroosmotic pump

The alternating current electroosmotic micropump, also known as induced charge electroosmotic micropump, actuates upon the diffuse charge in the double layer that is induced between an electrolyte in contact with an electrode that is subjected to a potential [21]. Typically, unidirectional flow is obtained by arrays of asymmetric pairs of electrodes subjected to a single phase ac signal, known as ACEO pump [22], or by arrays of symmetric electrodes subjected to a travelling-wave signal, known as TWEO pump [23].

The mechanism responsible for the fluid pumping at low voltages generated in a travelling-wave electrode array is as follows (see figure 4). When a travelling-wave potential is applied to the electrodes, counter-ions accumulate in the electrical double layer in response to the applied signal. The typical time for charging the double layer is given by the RC time of the equivalent circuit, where \( C \) is the capacitance of the double layer and \( R \) is the typical bulk resistance. Due to this double-layer RC charging time, a delay exists between the time of maximum induced charge and maximum applied signal. The electric field acts on the charge pulling the fluid in the direction of the travelling wave. The fluid velocity is maximum for angular frequencies of the order of \( \omega = 1/(RC) \sim (\sigma \lambda_D)/(\varepsilon \ell) \), with \( \ell \) a typical distance. The maximum generated pressure in a microchannel of rectangular cross-section is \( \Delta p_g = \Lambda 3 k V_0^2 L/2 h^2 \) [2], where \( k \) is the wavenumber of the travelling wave, \( L \) is the length of the array of electrodes, \( h \) is the height of the channel, and \( \Lambda \) is a factor less than one that accounts for compact layer effects.

Yang et al [24] performed experiments on TWEO and ACEO pumping using the same microelectrode array by changing the electrical connections of gold microelectrodes. The ACEO velocity was around five times smaller than the TWEO velocity for the same applied voltage. The TWEO slip velocity was around 480 \( \mu \)m/s for a TW signal of amplitude 1.5 volts peak to peak and frequency 1 kHz, using a saline solution of 100 \( \mu \)M KCl in water. Huang et al [25] fabricated an ACEO micropump made of long serpentine array of 3D asymmetric stepped microelectrodes. Using an ac signal of amplitude 1 Volt rms at a frequency of 1 kHz and distilled water as working fluid, the pump was capable of generating a maximum pressure of 1.4 kPa (much higher than previous ACEO pumps) and an effective slip velocity of 1.3 mm/s.

To increase the generated pressure, we could increase the applied voltage, however, this cannot be increased beyond some volts because it eventually leads to Faradaic reactions, which limit the use of the polarisation mechanism. Even before this threshold voltage is reached, fundamental
limitations such as ion crowding play an important role [26]. Liquids that have been pumped using AC induced charge electroosmosis had conductivities below 0.1 S/m.

5. Conclusions
In this work we have reviewed the basic principles of five different EHD micropumps: from those that exert forces in the liquid bulk to those that exert forces in the double layer. Very insulating liquids can be actuated by the injection pump. Semi-insulating liquids can be pumped by the injection, conduction and induction pumps. Aqueous solutions can be pumped in microsystems by the induction, electroosmotic and ac electroosmotic pumps. Electrolytes with conductivities much greater than 1 S/m are very difficult, if not impossible, to pump by using electrical forces in the bulk because of Joule heating leading to boiling. For $\sigma \geq 1$ S/m, the temperature rise can be on the order of 100 K for a voltage amplitude of 10 V [27].

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