Interactions of electrical fields with fluids: laboratory-on-a-chip applications

J. Wu

Department of Electrical and Computer Engineering, The University of Tennessee, Knoxville, TN 37996, USA
E-mail: jaynewu@utk.edu

Abstract: The area of ‘laboratory-on-a-chip’, miniaturised or microfluidic analysis systems, is a rapidly developing field. At the microscale, electrokinetic processes become enhanced, and the advent of AC electrokinetics (EK) in recent years further promotes the development of electrokinetic devices for microfluidics. ACEK has demonstrated to manipulate fluids and polarisable particles at low voltages without some of the disadvantages from DCEK, such as electrochemical reactions and the limitation of low ionic strength fluids. The three major mechanisms of ACEK, that is, dielectrophoresis, AC electro-osmosis and AC electrothermal effect, provide versatility and flexibility to interface with many current methods and technologies in multiple biological, chemical and physical disciplines. This paper gives an overview of ACEK and its applications, with an emphasis on fluid manipulation by electric fields.

1 Introduction

The last 20 years have witnessed significant advances in the development of laboratory-on-a-chip technology, which aims to perform bio/chemical analysis at microscale. Research in lab-on-a-chip is primarily motivated by reducing consumption of chemical reagents, improving efficiency, decreasing process time due to faster mixing and reactions, and realising automation by integration with sensors and control mechanism that provides safer operation and so on.

For lab-on-a-chip to perform bio/chemical analysis effectively, the following fluidic functions are expected: mixing, pumping, concentration steps to assist detection, for example. At the early stage of research, much of the effort is aimed at developing microscale counterparts of larger-scale components for handling fluids in microchannels. However, as device dimension scales down, dominant physical mechanisms change. Pressure-driven flow becomes increasingly inefficient due to the friction from high surface-volume ratio. Fluids are highly laminar and viscous, so pumping and mixing become problematic in microsystems. Besides, mechanically actuated microfluidic devices are difficult to fabricate and maintain.

As an alternative, electrokinetics (EK) is gaining popularity as a microfluidic actuation mechanism, and in many ways EK devices can offer advantages over mechanical pumps and actuators with gears and valves. Basically, EK induces micro fluid or particle motions within a microchannel by an impressed electric field. By scaling down the devices, sufficiently high electric field strength can be achieved with relatively low voltage for EK operation. EK devices involve no moving parts and therefore they are simple and reliable.

Numerous applications have demonstrated the use of electric fields to achieve separation, transport or analysis in microfluidic systems. This review summarises the main electrokinetic phenomena in terms of the theory and properties, with relevance to lab-on-a-chip devices, and hopefully in a style helpful to a cross-disciplinary readership. Representative applications are given in the text to illustrate various concepts of EK; however, the examples are by no means exhaustive.

2 Microfluidic EK

Traditional EK, or DC electrokinetics (DCEK), including electro-osmosis (EO) and electrophoresis,
has almost a 200 years of history and has been rather thoroughly investigated. They are widely used in microfluidic devices and form the mainstay of commercial applications. Electrophoresis is a non-contact method of moving and separating particles by the effects of an electric field on the net charge of a particle. EO in a channel arises from the interaction of the electric field along the channel with the electrical double layer at the interface of electrolytes and solids. DC fields applied along the channel can therefore produce particle separation, bulk flow or both. In DCEK, there is less sample diffusion than in pressure-driven flow in the direction of flow – an advantage for sample separations.

Typically, electric field strengths in the order of $10^5 \text{V/m}$ are needed to produce electrokinetic flow, so DCEK requires a hazardous high voltage. Because of the difficulty of making high-voltage insulation on chip, it hinders the monolithic integration with control circuits. Secondly, high voltage causes pH gradients from electrochemical reactions and bubble generation, which prevents a gas-tight system. One other issue is the difficulty in predicting voltage for local flow control, especially in a large system with many branches and reservoirs. When there are conductivity differences within the liquid samples, the voltage drop over the fluid is no longer uniform. As the fluid is pumped through the system by voltage differences, it is difficult to calculate at each node a proper electric potential to achieve a desired flow rate. Moreover, in a fluid network, the voltage difference between the inlet and outlet will be the sum of the voltage difference of the whole fluidic path, which could be prohibitively high for a large network.

To minimise these adverse effects, AC electrokinetics (ACEK) has emerged recently for on-chip pumping and particle manipulation at low voltage. Mobile charges can be induced at an interface (e.g. EO) or in the bulk of a fluid when there is a gradient in the fluid attributes (e.g. electrothermal effect). Because the induced charges in a fluid change their polarity with external electric fields, steady (not oscillatory) fluid motion can be generated with AC signals.

ACEK has many attractive features. The nonlinear nature of ACEK produces higher transport efficiency than DCEK. ACEK also minimises undesirable by-products of electrochemical reactions that are unavoidable with DC excitation. Fluid or particle motion generated by ACEK is local, so complex flow patterns can be generated by addressing electrodes individually. This property also makes it possible to manipulate and characterise a single cell or particle [1]. AC electrokinetic devices usually require microscale electrodes and structures, which can now be easily fabricated with good precision by micro technologies such as photolithography and laser micromachining. All the above factors have greatly motivated the development of ACEK.

There are mainly three types of ACEK phenomena, dielectrophoresis (DEP) [1–3], AC electro-osmosis (ACEO) [4–6] and AC electrothermal effect (ACET) [7, 8]. DEP is the particle motion arising from the force acting on the particles due to the difference in polarisability between the particles and the fluid. The study of DEP dates back to 1958 by Pohl's work [9], and it has been revived in the last decade because of its potential in the manipulation and characterisation of particles. Since then, DEP has evolved into a sophisticated and powerful tool for separation and characterisation of bioparticles, with many applications in medical diagnostics, cell therapy and so on. Interested readers are referred to [10] for an overview of cell DEP and experimental protocols. ACEO is the fluid motion induced by moving charges in the double layer. Pioneering work on ACEO can be found in [4, 5, 11]. ACET refers to the fluid motion caused by the interaction of electric fields and gradients of conductivity and permittivity of a fluid through Joule heating. ACEO is mainly effective for a low-conductivity fluid [e.g. deionised (DI) water], while ACET is more pronounced with higher conductivity, as energy dissipation in a fluid increases with conductivity at a fixed voltage. In the following, this review will outline the underpinning theories and attributes of different ACEK mechanisms, with an emphasis on fluid manipulation, i.e. ACEO and ACET.

## 3 Electrolyte–electrode microsystem

At the interface of an electrode and electrolyte, a transition occurs from electronic conduction in the electrode material to ionic conduction in the fluid. Ionic conduction involves mass transport, and as a result, fluid motion is induced. However, a fundamental problem with DCEK is the formation of bubbles at the electrodes when DC voltages larger than a few volts are used. The bubbles will disrupt the flow field, making microfluidic control difficult.

When an electric potential is applied between two electrodes in a solution, reactions can take place at the electrode–electrolyte interface when the applied voltage is more than a couple of volts, either DC or AC. Once the applied voltage exceeds the threshold for reactions to happen, co-ions will be produced at the electrodes. Only then can an electric current flow through the fluid to establish electric fields within the medium. Current conduction by reactions exhibits a resistive characteristic. So, this process can be
modelled as a resistor in series with a voltage source that equals the over potential of the reaction. In most electrokinetic devices, gold and platinum are used as the electrode materials. For such metals, the reaction with the lowest half-cell potential is the electrolysis of water, \( \sim 1.2 \) V.

It is well recognised that if electrokinetic transport could be generated without forming bubbles, electrokinetic microfluidics will enjoy a much higher degree of success. In the following, it is explained that electrode reactions can be avoided, however, only by using AC signals. For an energised electrode, ions of polarity opposite to the excitation voltage, that is, counter-ions, will be electrostatically attracted to the electrodes. This polarisation process is termed ‘induced charging’ \[12\] or ‘capacitive charging’ \[13\] by various groups. Together, the ion layer and the electrode are known as the double layer, modelled as a capacitor electrically. This ion layer has a thickness \( 1–10 \) nm in thickness.

At small DC voltage, without charge-transfer reactions, the electrode–electrolyte system acts like a charged capacitor. There is no electric field in the solution, resulting in no electrokinetic flow. So for DC electrokinetics (DCEK), electrode reactions are essential to establish an electric field in the fluid. The fluid bulk is treated as a resistor in series with a voltage source that equals the over potential of the reaction. As the frequency increases, the impedance from \( C_{\text{cell}} \) is much larger than that from \( R_{\text{solute}} \), and most of the voltage drop occurs across the interfacial double layer, which is desirable for ACEO techniques.

As the frequency increases, the impedance from \( C_{\text{dbl}} \) decreases. More voltage drops across the fluid bulk, and electric current dissipates energy into the fluid according to Ohm’s law, so that electrothermal effects begin to dominate. As for the DEP effect, it is also the electric fields in the fluid bulk that induce the particle movement, so a higher frequency is also favoured because the electric field strength will be higher, given that the frequency is far away from the so-called DEP cross-over frequency \[1, 14\].

### 4 AC electrokinetics

ACEK investigates the behaviour of electrolytes and polarisable particles within the fluids when they are subjected to AC electrical fields. Mobile charges are induced near the electrodes by the applied potential, and they move under the influence of local electrical fields to produce particle or fluid motion. ACEK mainly includes ACEO, ACET and DEP. An overview of each technique is given as follows.

#### 4.1 Dielectrophoresis

The research of DEP studies the interactions between a field-induced dipole moment on a particle and a non-uniform field. A particle with a polarisability (conductivity, permittivity) different from that of the suspending medium will influence the electric field distribution around it. As shown by the numerical simulations in Fig. 2, the electric field lines will bend towards the particle if it is more polarisable (Fig. 2\( a \)) and bend away from it if it is less polarisable (Fig. 2\( b \)). If the particle is placed in a non-uniform electric field, the density of the electric field lines will be greater on one side of the particle than the other side. This imbalance leads to a net force on the particle, and this effect is known as DEP. When the particle is more polarisable than its surroundings, it will move towards the high field region, known as negative DEP. The opposite situation leads to negative DEP, and the particle moves away from the high field region. DEP
can occur at DC as well, however, it will often be masked by other effects such as electrophoresis, EO and electrolysis.

The most common way to realise non-uniform electric fields is by adopting asymmetric microelectrodes, and there are numerous examples for that. Within the fluids, non-uniform fields can also be induced by placing conductive or insulating objects (non-electrodes) in the paths of electric fields. This principle is the same as that of electric fields disturbed around particles (shown in Fig. 2), also based on the difference in polarisability of the objects and the fluid medium. Two examples of such applications are Cummings and Singh [15], who introduced a so-called iDEP device consisting of an array of insulating posts in a microchannel, and Banerjee et al. [16] who used metal posts between electrodes to align carbon nanotubes.

A spherical particle experiencing DEP will exhibit a velocity as

$$
\langle u_{DEP} \rangle = \frac{a^3 \varepsilon_m}{6 \eta} \text{Re} \left[ \frac{\varepsilon_p - \varepsilon_m}{\varepsilon_p + 2 \varepsilon_m} \right] \nabla |E|^2
$$

where \( \varepsilon_p, \varepsilon_m \) are the complex permittivities of the particle and the medium, and \( \varepsilon_{p,m} = \varepsilon_p - i(\sigma_{p,m}/\omega) \); \( \sigma_{p,m} \) are the conductivities and \( \omega \) is the frequency. \( f_{CM} \), known as the Clausius–Mossotti factor, is determined by the difference between the dielectric properties of the particles and surrounding medium, indicating whether the particle exhibits positive or negative DEP. \( f_{CM} \) is frequency dependent with a value between \(-0.5\) and \(1\), and \( f_{CM} \) changes its sign at a cross-over frequency.

DEP provides an effective means to manipulate particles, such as DNA, protein molecules, virus, bacteria, plant and animal cells, and inorganic particles. This technique has been studied in great details for controlled manipulation of particles, binary separation and characterisation of particles [1–3]. Additionally, the study of \( f_{CM} \) can derive information on particle structure. In cell electrophoresis, an important and consistent finding is that small changes in the physico-chemical properties of a cell’s membrane can produce measurable changes of the cross-over frequency.

In microfluidic study, particles are used as tracers in fluid experiments and they also experience the DEP effect. From (1), it can be seen that DEP velocity is size dependent, and decreases rapidly with the distance to the electrode. Order of magnitude estimation shows that 1 \( \mu \)m particles will exhibit no more than 0.9 \( \mu \)m/s DEP velocity when they are 10 \( \mu \)m from the electrode edge at 5 Vrms. Since the peak velocity of EK flows often exceed 100 \( \mu \)m/s, DEP velocity can be neglected in the analysis. Also smaller tracer particles will significantly reduce the DEP effect.

Another application of DEP is to drive fluid, more specifically, to move droplets on the surfaces by DC (a.k.a. electrowetting) or AC signals [17]. This

![Figure 2: Numerically simulated electric field distribution around a sphere in a non-uniform electric field](image_url)

- **Figure 2** Numerically simulated electric field distribution around a sphere in a non-uniform electric field
  - **a** Sphere more polarisable than the suspending medium
  - **b** Sphere less polarisable than the suspending medium
technique is applicable for ‘open channel’ systems, where the liquid in the form of a droplet moves along microelectrodes on the surface of planar substrates. The application of a DC or AC electric field from the microelectrodes underneath the droplet results in electrical stress (DEP force) on the droplet, and attracts fluids to regions of strong electric field. Removing the electric field will pinch off small amounts of fluid to form/dispense droplets. Liquid fronts can be driven at speeds up to $\sim 25$ cm/s, and multiple liquid droplets of volumes down to $\sim 1$ nl can be dispensed from microlitre volumes. Chemical analysis and reactions can then be performed on the sub-picolitre scale in a controlled manner by using this method to bring droplets together. Even though such a system does require hermetic sealing due to evaporation, it provides a new tool for analytical and separation science.

4.2 AC electro-osmosis

4.2.1 Mechanism: ACEO refers to the microfluidic motions generated at electrode surfaces when AC signals are applied. It typically happens at low frequency when the interfacial impedance dominates.

When a small AC signal is applied over an electrode pair, the electrode surfaces become capacitively charged, that is, form counter-ion accumulation. The counter-ions migrate with or against an electric field that is tangential to the electrode surface, which in turn produces fluid motion due to fluid viscosity. Similar to DCEO, the fluid velocity is approximately given as

$$u_{\text{ACEO}} = -\left(\frac{\varepsilon_m}{\eta}\right) \cdot \Delta \xi \cdot E_t$$

where $\varepsilon_m$ and $\eta$ are the permittivity and viscosity of the medium, $E_t$ is the electric field parallel to the solid surface and $\Delta \xi$ is the voltage drop over the charged double layer, which is proportional to the induced charge density $Q \sim \varepsilon / \varepsilon_0 \Delta \xi$. With the electro-osmotic velocity as the boundary conditions, the fluid motion in the bulk can be solved using Navier–Stokes equation.

With AC signals, the double layer changes its polarity periodically. Since the tangential electric field changes its direction with the double layer charges, steady fluid motion instead of oscillatory flows is produced. Synchronised tangential and normal electric fields can be obtained from a single voltage source with proper electrode design. Two major configurations of ACEO electrodes have been reported so far. One uses ‘side-by-side’ electrode pairs [4, 5], and the other uses ‘face-to-face’ electrode plates with asymmetric patterns/sizes [18, 19]. Both designs create non-uniform field distribution, which results in the coexistence of tangential and normal electric fields at the electrode surface. In the following, the characteristics of ACEO will be explained using the ‘side-by-side’ electrode design.

Fig. 3a shows the side view of a pair of ‘side-by-side’ electrodes and the simulated electric field distribution around them. It can be seen that the tangential component of the electric fields changes direction over the span of one electrode. As a result, microflows in opposite directions are produced, leading to four counter-rotating vortices over an isolated pair of electrodes, as shown in Fig. 3b. The change of direction happens at a location of $\sqrt{(L + a^2 + a^2)/2}$ away from the electrode’s inner edge ($L$ is the electrode width and $a$ the half electrode gap) [20].

The fluid motion over the electrode surface can be tracked by seeding the fluid with particles. Fig. 4 shows that 3 $\mu$m latex particles are being swept into two lines by the four vortices [21]. Particles are seen
to move in directions consistent with capacitive charging of the electrodes and to assemble into lines. The microflows move vertically upwards in the planes of the stagnation line due to mass conservation. The particles become trapped at the stagnation lines because the vertical flow is weak at the stagnation point and an opposite vertical force such as gravity can prevent this vertical motion away from the electrodes. The line locations will deviate from the predicted \( \sqrt{(l + \alpha)^2 + \alpha^2}/2 \) at high frequency (>1 kHz), and move closer to the inner edge. This occurs because the fluid velocity becomes lower as it reaches further into the electrode, and this decrease is more pronounced at higher frequencies [22].

Following the analysis presented in [11], the time-averaged expression for ACEO slip velocity can be given as \( \langle u_{\text{ACEO}} \rangle \sim (eV^2/\eta)\Omega^2/(1 + \Omega^2)^2 \), where \( r \) is the distance to the electrode edge, \( \Omega \) is the normalised frequency \( \Omega = \omega/\omega_0 \) and \( \omega_0 \approx 2\sigma\lambda_d/\varepsilon \pi \). \( \lambda_d/(\varepsilon \pi) \) represents the process of charging the double layer through the resistive fluid bulk. It can be seen that ACEO velocity has a bell-shaped dependency on frequency. Around \( \omega_0 \), the velocity reaches its maximum. At lower frequencies, the electric field in the fluid is reduced and exerts less force on the ions to migrate. At higher frequencies, there is not enough time to charge the double layer and ACEO also becomes insignificant. Experimental data on ACEO velocity are presented in [4] as a function of frequency and electrolyte conductivity, and there is good agreement with a theoretical prediction of \( \omega_0 \). At its peak frequency, ACEO has demonstrated to generate surface velocity of 50–600 \( \mu \)m/s by applying AC signals in the order of 1–5 V\( \text{p-p} \) with an electrode spacing of 25 \( \mu \)m [4].

Although ACEO is a technique for fluid manipulation, it can exert a drag force on particles through fluid motion. Therefore ACEO can be used to transport particles as well as fluid, and there is no size dependency. The applications of ACEO will be discussed in more detail in a following section.

4.2.2 ACEO due to faradaic charging: In the ACEO discussed above, the charges at the electrode surfaces are counter-ions to the electrode potentials, attracted from the electrolyte to screen the electrode potential. Electrically, the process is manifested as charging the double layer capacitors. Until recently, capacitive charging was the only ACEO mechanism reported in the literature. However, as mentioned in Section 3, there exists another AC electrode process, - electrochemical reactions, populating the electrode surfaces with co-ions instead of counter-ions. Consequently, it is expected to produce surface-flows in the opposite directions to those by capacitive charging.

It has been observed by several groups [23–26] that ACEO flows could reverse their directions at a higher applied voltage, for example, 2 V\( \text{rms} \). Faradaic charging of electrodes has been suggested as a possible mechanism of flow reversal at low frequencies [25, 26]. At an appropriate electrode potential, co-ions instead of counter-ions are generated from electrochemical reactions at the electrodes following Faraday’s law therefore dubbed as ‘Faradaic charging’. The threshold voltage for electrochemical reaction or electrolysis is higher for AC signals than for DC (\( \sim 1.2 \) V) [25].

Capacitive and Faradaic charging differ in the following aspects. (1) There exist two charging mechanisms, capacitive charging and Faradaic charging, producing ions of opposite signs in an electrical field, which in turn result in EO flows in opposite directions. (2) The EO velocity produced by the two charging processes vary differently as a function of the applied voltage. The difference arises from the charge density \( \rho \) in the double layer. \( \rho \sim \Delta \xi \) for capacitive charging and \( \rho \sim \exp(\Delta \xi) \) for Faradaic charging, respectively [27]. It also means that the microflows from Faradaic charging can be stronger than those from capacitive charging. (3) Because there is a threshold voltage for Faradaic reaction to occur, the electrode charging at low voltages will be capacitive charging because Faradaic charging

![Figure 4](image-url)
dominates at higher voltages with its exponential dependence on potential.

Capacitive charging and Faradaic charging coexist and compete for dominance, which has been experimentally observed [25]. Fig. 5 shows the erasure of particle assembly by Faradaic charging. The left frame shows the particle line assembly at 1.0 \( V_{\text{rms}} \). When the applied voltage is at 2.2 \( V_{\text{rms}} \), particle lines are erased by Faradaic charging (right image), since Faradaic flows at the stagnation locations are diverging. Impedance analysis at different voltage levels also suggests ion generation. As shown in Fig. 6, with increasing testing signals, the impedance magnitude decreases and the phase angle approaches 0, which is consistent with ion generation [28].

4.2.3 Applications: Fluid and particle manipulation using ACEO have been reported in various forms. The foregoing discussion has shown that reflective-symmetric, counter-rotating vortices are generated over a pair of identical electrodes. This characteristic can be implemented for mixing in microchannels. Other applications include fluid transport and particle collection.

To use ACEO for pumping, it is essential to break the symmetry of electric fields within an electrode pair to produce a unidirectional flow. This can be achieved by spatial asymmetry in electrode design, or by polarisation asymmetry. Designs with spatial asymmetry to induce a net flow include asymmetric ‘side-by-side’ electrodes [24, 29, 30], orthogonal electrodes [25] and a 3D ACEO pump [23, 31]. Fig. 7a shows a concept schematic of generating net flow by unequal width of the electrodes in a pair, that is, asymmetric electrodes. The surface flow velocity of asymmetric electrode pump is shown to increase with decreased channel height, due to higher surface/volume ratio, and it also exhibits voltage dependence as \( u \sim V^2 \) [30].

Another strategy to implement pumping is to induce polarisation asymmetry with biased ACEO [6]. Fig. 7b illustrates the principle of asymmetric polarisation in two symmetric electrodes, with a biased AC signal impressed over the electrodes. With the applied signal as shown, the left electrode is always at a positive potential and more prone to Faradaic charging, while the other is always at a negative potential and subject to capacitive charging. When the voltage exceeds the threshold for reaction, positive charges are induced on both electrodes, and unidirectional flows are produced for pumping action. Since Faradaic charging is involved in a biased ACEO pump, the surface flow velocity has an exponential dependence on voltage, as can be seen in Fig. 8 [28]. Two channel heights are used and the lower channel height produces significantly higher surface flow velocity. The flow velocity plots are curve-fitted both quadratically and exponentially. The flow velocity grows much faster with voltage than quadratic relationship, and good agreement is found between the measurement and exponential curve-fit. ACEO pump can also be implemented with a travelling wave of four signals [32].

Besides fluid manipulation, ACEO (and ACET in a later section) can also be used to collect and concentrate particles for lab-on-a-chip applications [33–35]. A concentration step is critical when detecting low concentrations of bioparticles, as it can increase particle count at the detection sites to a detectable level. ACEO particle concentrator generates microflows that convey particles from the bulk of the fluid onto the electrode surface. Since non-uniform flow fields are produced in ACEO, at locations of low flow velocity, particles can become adhered to substrates from a variety of forces such as gravity, electrostatic attraction and DEP. In order to effectively convect particles to the electrode surface and deposit them there, it is important there are converging flows towards the stagnation location and the flows are not too strong that the upward force

---

**Figure 5** Particle line dispersion due to Faradaic charging of the double layer

AC signals at 100 Hz were used
Left: 1.0 \( V_{\text{rms}} \), right: 2.2 \( V_{\text{rms}} \)
Polystyrene spheres at a diameter of 5 \( \mu \)m were used at a particle concentration of \(~5.0 \times 10^6\) particles/ml [13]
outbalances the gravity, DEP and other surface forces. By taking advantage of microfabrication technology, various particle assemblies can be formed using different electrode designs [36], and onto a designated part of microstructures, such as a microcantilever surface [19].

4.3 AC electrothermal effect

ACEO is typically limited to fluids with low ionic strength, for example, DI water. High conductivity compresses the thickness of the double layer, rendering EO ineffective. For the same applied voltage, more conductive fluids have a lower peak velocity, and the highest fluid conductivity with which ACEO has been observed is 85 mS/m. Most biological applications involve conductive fluids, up to 1–2 S/m. To actuate fluid at such high conductivities, a viable option is to use the ACET. Recent studies have demonstrated that ACET is a promising technique to manipulate conductive fluids.

4.3.1 Mechanism: At higher frequencies, for example, 500 kHz, the impedance at the electrode surface is much lower than that of the resistive bulk for a typical electrokinetic microdevice. In this case, the overall applied voltage can be rather high without causing reactions at the electrode because the actual voltage drop across the interfaces will be low. Most of the AC signal drops across the fluid, which will induce temperature rise and other effects, such as the electrothermal effect.

The electrothermal effect was first utilised in macro applications. Development of miniaturised ACET devices are hindered by the fact that ACET could become suppressed at the microscale due to a no-slip boundary condition. Too small a hydraulic diameter will inhibit the formation of ACET flow.

The ACET refers to fluid motion resulting from the interactions of AC electric fields and temperature gradients in the fluid. When an electric field $E$ is

![Figure 6](image)

**Figure 6** Impedances of an electrode pair immersed in de-ionized water at various potential levels for measurement [28]

$a$ Impedance magnitude

$b$ Phase

![Figure 7](image)

**Figure 7** Concept of generating net fluid transport

$a$ By asymmetric electrodes [29]

$b$ By asymmetric polarisation
applied over a fluid of electrical conductivity \( \sigma \), Joule heating of the fluid will take place according to the energy balance equation

\[
k \nabla^2 T + \frac{1}{2} (\sigma E)^2 = 0
\]

(3)

where \( T \) is the temperature and \( k \) is the thermal conductivity. If the electric field is non-uniform, the heat generation will be non-uniform as well, which leads to temperature gradients \( \nabla T \) in the fluid. In turn, the temperature gradient \( \nabla T \) will produce spatial gradient in fluid conductivity and permittivity by \( \nabla \varepsilon = (\partial \varepsilon / \partial T) \nabla T \) and \( \nabla \sigma = (\partial \sigma / \partial T) \nabla T \), which induces mobile space charges, \( \rho_s \), in the fluid bulk according to \( \partial \rho_s / \partial t + \nabla \cdot (\sigma \varepsilon) = 0 \) and \( \rho_e = \nabla \cdot (\varepsilon \varepsilon) \).

The electric field will impose on the induced space charges a force

\[
F_{ct} = \rho_e E - \frac{1}{2} |E|^2 \nabla \varepsilon
\]

(4)

The first term in (2) is the Coulomb force and the second term is the dielectric force. The time-averaged force is

\[
\langle F_{ct} \rangle = -\frac{1}{2} \left( \frac{\nabla \sigma}{\sigma} - \frac{\nabla \varepsilon}{\varepsilon} \right) \cdot \frac{|E|^2}{1 + (\omega \tau)^2} - \frac{1}{4} \nabla |E|^2
\]

(5)

where \( \sigma \) and \( \varepsilon \) are the electrical conductivity and permittivity of the medium, \( \tau = \varepsilon / \sigma \) is its charge relaxation time, and \( \omega = 2\pi f \) is the radian frequency. Equation (5) can be rewritten as

\[
\langle F_{ct} \rangle = -\frac{1}{2} \left( \frac{1}{\sigma} \frac{\partial \sigma}{\partial T} - \frac{\partial \varepsilon}{\partial T} \right) \cdot \frac{1}{1 + (\omega \tau)^2} \frac{\nabla |E|^2}{\varepsilon}
\]

(6)

It can be seen that the electrothermal force is proportional to \( \nabla T \), which can be imposed externally, such as by strong illumination or generated by electric currents in the fluid system.

For aqueous media at 293 K, \( (1/\varepsilon)(\partial \varepsilon / \partial T) = -0.004 \) and \( (1/\sigma)(\partial \sigma / \partial T) = 0.02 \), so (6) can be calculated to be

\[
\langle F_{ct} \rangle = -0.012 \cdot \nabla T \cdot \frac{|E|^2}{1 + (\omega \tau)^2} + 0.001 \cdot \nabla T \cdot |E|^2
\]

(7)

Interesting to note, according to (6) and (7), the ACET force has no obvious dependency on fluid conductivity. It is because that the conductivity dependency is manifested through \( \nabla T \). For internally induced \( \nabla T \), that is from electric currents passing through the fluid, the ACET force depends on fluid conductivity according to (3). If \( \nabla T \) is generated predominately by external sources, then the ACET has little dependence on fluid conductivity.

At low frequencies, \( 1/(1 + (\omega \tau)^2) \simeq 1 \), and \( \langle F_{ct} \rangle \simeq -0.011 \cdot \nabla T \cdot |E|^2 \), and its direction is that of the Coulomb force. As the frequency approaches \( 1/2 \pi \tau \), the Coulomb force decreases and becomes less than the dielectric force at a crossover frequency \( f_c = \sqrt{1/2 \pi \tau} \). The space charges will thus experience a weaker \( \langle F_{ct} \rangle \) in the opposite direction.

As a result of forces on the space charges, fluid motion will be induced as

\[
\frac{\partial u}{\partial t} + \rho (\nabla \cdot u) u - \eta \nabla^2 u + \nabla P = F_{ct} \text{(Navier-Stokes equation)}
\]

(8)

where \( \rho \) is the fluid density, \( \eta \) is the dynamic viscosity of the fluid, \( P \) is the external pressure and \( u \) is the velocity of the fluid. Together with \( \nabla \cdot u = 0 \) for an incompressible fluid, fluid velocity can be found for an electrokinetic microdevice.
In the case of a pair of ‘side-by-side’ electrodes, without an external thermal gradient, the ACET velocity can be approximately expressed as [7],

$$u_{ACET} = 5 \times 10^{-4} \rho \frac{\sigma V^2}{C_2} \frac{\partial T}{\partial y} \frac{1}{k} \frac{\rho}{\eta} \frac{1}{r} \sigma \frac{\partial T}{\partial y}$$

(9)

where $k$ is the thermal conductivity of the fluid. The fluid velocity increases linearly with conductivity, due to increased heat generation and temperature gradients. If the temperature gradient $\nabla T$ is purely external, then there is no dependence on fluid conductivity. (Of course, this is a hypothetical situation since electric currents going through conductive fluids will always dissipate heat within.) When there exists an external thermal gradient, ACET velocity should be augmented by an amount of [7]

$$u_{ACET, ext} \approx 3 \times 10^{-3} \frac{\rho}{\eta} \frac{\partial T}{\partial y} \left| \frac{1}{k} \frac{\rho}{\eta} \frac{1}{r} \sigma \frac{\partial T}{\partial y} \right|$$

(10)

where $|\partial T/\partial y|$ is the external thermal gradient.

It is obvious from (10) that ACET flow could also exhibit flow reversal from a change in heat transfer conditions, which has also been experimentally demonstrated [37, 38]. At low voltages with low-conductivity fluids, metal electrodes can quickly transfer the generated heat to the ambient to remain at room temperature. The fluid moves along the same direction as the ACEO flows. By illuminating the electrodes (heating up the electrodes), thermal gradients at the electrodes reverse and the fluid moves in the opposite direction. Without external heat, it is still possible that electrodes will reach an elevated temperature at high voltages and result in flow reversal, which might be responsible for the flow reversal at high frequencies observed by some groups.

4.3.2 Applications: ACET devices are in many ways similar to ACEO devices. Almost all the ACEO devices can be converted to ACET devices by operating at a higher frequency, and they can also be implemented as mixer, pump and concentrator in a lab-on-a-chip. The designs of ACET devices can also be roughly classified into ‘side-by-side’ [37, 39] or ‘face-to-face’ [40] configurations.

One of the earliest microscale prototypes of ACET pump was reported by Fuhr et al. in early 1990s, using a travelling wave of four-phase signals to drive fluids [39]. With an electrode array of 10 $\mu$m width and 10 $\mu$m gap, a peak velocity of $\approx 180 \mu$m/s is achieved at 35 $V_{p-p}$, 100 kHz. The conductivity of fluid they use is up to 9.3 mS/m, which is much higher than $\approx 10^{-6}$ S/m reported for electrohydrodynamic micropumps at that time, but still much lower than bio-buffers.

In 2000, Green et al. [37] reported the swirling motion of fluid ($\sigma = 2.1$ mS/m) above a pair of microelectrodes, and these vortices were related to both external illumination and electric field. The illumination is determined as a source of temperature gradients in the fluids. A recent paper by Gonzalez et al. [38] has further presented a detailed study of the ACET effect in the presence of an external temperature gradient (strong illumination). The experiments in the paper verified many theoretical predictions for ACET, including that of fluid velocity changes with illumination intensity and exhibits a quadratic relationship with voltage. With a solution of $\sigma = 2.5$ mS/m and external illumination, a peak velocity of 80–110 $\mu$m/s at 10 $V_{rms}$ is observed. Using four electrodes arranged in a circle and a four-phase travelling wave, electro-rotation of the fluid has also been demonstrated in the paper. This effect can be used as a mixer for lab-on-a-chip devices.

Recent study has shown that ACET can be generated by electric fields alone in conductive fluids (20–700 mS/m), provided that proper boundary and operating conditions are applied. Sigurdson et al. [41] reported use of the ACET effect to induce vortices within a pressure-driven flow-through system ($\approx 0.6$ S/m), improving the binding rate of antigen-antibody. Lian et al. [40] also developed two ACET devices without external thermal sources, a parallel plate particle trap and an asymmetric electrode micropump. Both particle trapping and micropumping are demonstrated at low voltages. For fluids with conductivity of 224 mS/m and a peak fluid velocity of $\approx 100$ $\mu$m/s a modest field of $1.6 \times 10^4$ $V_{rms}$/m at 200 kHz is observed.

Using a pair of ‘face-to-face’ electrodes, the experiment verified the theoretical prediction that fluid velocity indeed depends on the applied voltage $V^2$, as shown in Fig 9, which also agrees with the simulation results. Both in the experiments and simulation, the fluid velocity maxima are $\approx 20$ $\mu$m above the electrode edge, which is consistent with the non-slip boundary conditions, and also a feature different from EO. The capability of operating on an electric field alone will facilitate the application of ACET devices.

4.3.3 Competition with Buoyancy: Accompanying the electrothermal effect is the temperature rise $\Delta T$ in the device, which will lead to a change in fluid density and thus natural convection. The buoyancy force is...
4.4 Comparison of ACEO with ACET

Both ACEO and ACET induce fluid flows by applying a non-uniform electric field. In some cases, ACEO and ACET produce very similar flow patterns, but they are of different origin. ACEO originates from electric stress at the electrode surface, while ACET exerts volume forces on fluids. This results in differences between ACEO and ACET in many ways, in terms of the device design, operating conditions and flow patterns.

With respect to the design of electrokinetic devices, ACEO velocity is observed to increase with reduced hydraulic diameters, while the ACET force becomes suppressed when the hydraulic diameter becomes less than 50 μm. For ACEO flow, the highest fluid velocity is at the electrode surface, while ACET flow adopts a no-slip boundary condition and its velocity maxima are observed to be appreciably above the electrodes.

With respect to the operating conditions, referring back to the equivalent circuit in Fig. 1, low operating frequencies (typically lower than 100 kHz) will favour ACEO as the interface impedance dominates. ACEO velocity has a bell-shaped dependence on frequency with an optimal frequency around \((\sigma/\varepsilon)(\omega/L)\). At higher frequencies, the ions at the surface do not have sufficient time to charge and discharge the double layer, and the ACEO velocity goes to zero. From the prospective of an equivalent circuit, there is not sufficient voltage drop across the double layer capacitance. ACET velocity has little dependency on frequency when it is well below the cross-over frequency \(3\sigma/\varepsilon\). However, because the actual voltage drop across the resistive bulk changes with frequency, ACET flow may exhibit frequency dependence, that is, slower at low frequency. The ACET flow will reverse its direction at a frequency higher than \(3\sigma/\varepsilon\), as the fluid volume forces the change from electric force to dielectric force.

Additionally, ACEO is limited to fluids with low ionic strength. Peak ACEO velocity (at its optimal frequency) decreases with increasing conductivity, and ACEO flows become weak for fluids with conductivity above 100 mS/m [23]. As a contrast, ACET velocity increases with conductivity, which is expected as energy dissipation increases as \(\langle P\rangle = \sigma E_{v_{\text{rms}}}^2\). ACET flow has been observed for fluid conductivity up to 700 mS/m. However, the heat generation should not be excessive; otherwise, the buoyancy force will predominate. Hence, good thermal conduction is important for ACET devices in order to produce a thermal gradient instead of temperature rise.

Another difference is the flow pattern generated by ACEO or ACET. For the electrode configurations presented so far, ACEO and ACET produce very similar flow fields. However, they are fundamentally different in that ACEO is an interfacial phenomenon following a direction tangential to E-fields, whereas ACET is a volume force that follows the directions of E-fields (at low frequencies, AC fields exert volume force on fluids as \(f_E = \rho E\), that is \(f_E = [(\sigma\varepsilon\partial\varepsilon/\partial T) - \varepsilon(\partial\sigma/\partial T)]((\sigma + i\omega\varepsilon))\nabla T \cdot E\)).
Therefore different flow fields will be generated for certain electrode designs.

This point can be illustrated using a pin-line electrode pair, that is, a pair of platinum electrodes in T configuration as shown in Fig. 10 [42]. The wires are 250 μm in diameter and separated by 400 μm at the tip. At frequencies below 5 kHz, two major vortices are formed beside the ‘pin’ electrode. As shown in Fig. 10a, the fluid goes up along the sides of the pin, then extends into the fluid bulk and comes down to form vortices. There are also two minor vortices between the pin tip and the ‘line’ electrodes. Their directions mirror those of the major vortices. At frequencies above 5 kHz, the microflows that go up along the pin start to diminish and transform into the flow pattern at high frequencies (≈ 100 kHz) as shown in Fig. 10b. The fluid emanates from the pin tip towards the line electrode and continues to go over the line, and no frequency dependency on fluid velocity is observed until 20 MHz, beyond which the dielectric coupling between the electrodes dominates.

The change of flow patterns from that of Fig. 10a and b can be explained as the transition from ACEO flows to ACET flows. At low frequencies, the flow pattern is predominately induced by ACEO, which can be deduced from the simulated electric field distribution in Fig. 11a. There are both tangential and normal components of electric field around the pin tip. ACEO flows will move upwards along the pin and move outwards from the centre along the line electrode, which is consistent with experimental observation. The ACET effect is responsible for the flow pattern at high frequencies. The electrodes are set at a fixed temperature at the boundaries, but assume continuity for the sections in the fluid chamber. So the highest thermal gradient (also the highest charge density) happens at the space between the two electrodes, where the electric field is also the strongest, and this
produces a different flow pattern from ACEO. The simulated fluid field agrees well with the experimental results.

5 Summary

The last several years have witnessed a steady expansion in the new methods and devices associated with ACEK. The versatility of ACEK devices allows interfacing with many current methods and technologies. It is evident that the inherent flexibility of ACEK technologies will allow them to permeate and advance the development of the lab-on-a-chip.

The purpose of this review is to assist newcomers to the field by giving a broad overview of the novel achievements. As a result, for every distinct method or device, only some relevant examples have been given in the review. Interested readers are recommended to follow the bibliography for more comprehensive research.

6 Acknowledgment

The author acknowledges the support of National Science Foundation, USA, under the grant No. 0448896.

7 References


