

LETTER TO THE EDITOR

AC Electric-Field-Induced Fluid Flow in Microelectrodes

During the AC electrokinetic manipulation of particles in suspension on microelectrode structures, strong frequency-dependent fluid flow is observed. The fluid movement is predominant at frequencies below the reciprocal charge relaxation time, with a reproducible pattern occurring close to and across the electrode surface. This paper reports measurements of the fluid velocity as a function of frequency and position across the electrode. Evidence is presented indicating that the flow occurs due to electroosmotic stress arising from the interaction of the electric field and the electrical double layer on the electrodes. The electrode polarization plays a significant role in controlling the frequency dependence of the flow.

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Recent work in the field of AC electrokinetics has shown that submicrometer particles can be characterized and manipulated in microelectrode arrays using dielectrophoresis (1–4). Advanced fabrication methods have been used to manufacture complicated electrode structures that can generate precise electric fields up to 10^7 V m⁻¹ over a wide range of frequencies (2–4). However, the high electric fields give rise to fluid movement, particularly close to the electrode surface. The observation of the dielectrophoretic behavior of submicrometer particles in electrolytes has revealed a reproducible pattern of fluid flow at frequencies below the charge relaxation frequency of the liquid. Measurements show that the velocity of the fluid is frequency dependent, tending to zero at upper and lower frequency limits, and with magnitudes up to $500 \mu\text{m s}^{-1}$. This field-induced fluid flow is likely to be the cause of previously unexplained phenomena in the dielectrophoretic manipulation of particles on microelectrodes (3).

We postulate that the driving force for this flow arises from the interaction of the nonuniform electric field with the charge in the diffuse double layer. Other authors have also observed particle motion in microelectrode structures and have related these effects to electrical stresses on the double layer (5, 6). In these references the motion of the particles was attributed to gradients in the conduction current arising due to the collective interaction of the particles or inhomogeneities in the surface conductivity of the microstructures. In this paper we present measurements of fluid motion in AC fields generated in microelectrodes and show the frequency dependent nature of the fluid velocity. A novel mechanism is postulated for the underlying physical origin of the flow, which takes into account the polarization of the electrodes. This could be the origin of previously observed phenomena resulting from the action of AC fields on fluids, in particular, the cessation of fluid motion above 1 MHz (5). We term this mechanism *AC electroosmosis*.

Experimental observations were made on microelectrodes, consisting of two parallel coplanar plates fabricated on planar glass substrates. The electrodes were made from a series of metal layers: 10 nm Ti, 10 nm Pd, 100 nm Au, 10 nm Ti, and each plate was 0.1×2 mm in size separated by a 25- μm gap. Electric fields were generated in the electrodes using signals with frequencies in the range 20 Hz to 1 MHz and potentials up to 5 V peak to peak. Fluorescent

latex particles of 557 and 216 nm diameter (Molecular Probes, Oregon) were used as markers to observe the fluid flow and were suspended in a KCl solution of conductivity 8.6 mS m^{-1} at 2×10^{-3} % by volume. Particle trajectories were observed with a microscope and CCD camera and recorded on S-VHS video. Only particles in focus in the digitized images, and therefore in the focal plane of the objective ($\sim 1 \mu\text{m}$ above the electrode), were measured.

At frequencies below 1 MHz the pattern of fluid flow was as shown in Fig. 1. The particles moved from the interelectrode gap out across the electrode surface. The maximum velocity was observed near the electrode edge and determined to be over $500 \mu\text{m/s}$ for an applied potential of 5 V peak to peak. This velocity is significant when compared with the dielectrophoretic velocity for these particles, which is in the range 1–10 $\mu\text{m/s}$ for the same potential and electrode gap (7). Particle velocities were recorded for an applied potential of 5 V peak to peak and the velocity, plotted against ωz ($\text{m} \times \text{rad/s}$), is shown in Fig. 2 for a series of positions on the electrodes. The variable z is the distance from the center of the gap between the electrodes, i.e., the distance from the edge (marked on the Fig. 1) plus $12.5 \mu\text{m}$ in this case and ω is the angular frequency of the electric field. It can be seen that the velocity varies with frequency, reaching a maximum at a characteristic frequency and tending to zero at low and high limits. When plotted against ωz , the maximum velocity occurs at approximately the same point along the horizontal axis. It also can be seen that the velocity decreases as z increases.

One possible mechanism for the source of the flow is heating of the fluid by the electric field. Localized heating creates gradients in permittivity and conductivity of the fluid, which in turn can give rise to electrical volume forces (electrothermal). However, the fluid flow due to this mechanism has a relatively low velocity and cannot explain the observed frequency-dependent fluid motion (7).

The mechanism that we are proposing as the explanation for the observed flow is electroosmosis, a well-known phenomena in DC fields (8). The fluid velocity is proportional to the tangential electric field, E_t , and the zeta potential of the surface, ζ , according to $v = \epsilon E_t \zeta / \eta$, where η is the viscosity and ϵ is the permittivity of the medium. In the linear approximation, the zeta potential is proportional to the surface charge density in the diffuse double layer σ_q , so that

$$v = \frac{E_t \sigma_q}{\kappa \eta}, \quad [1]$$

where κ is the reciprocal Debye length. This equation can be used to estimate the velocity of the fluid given a value for the tangential field and surface charge density. In classical electroosmosis the driving field is constant throughout the medium and tangential to the surface. In the case of a nonuniform electric field, a component of the field lies tangential to the electrode surface, E_t , as shown schematically in Fig. 1. The charge in the diffuse double layer can be represented as a constant term and a time dependent excess charge, $\Delta\sigma_q(t)$ which varies with the electric field. For AC voltages, double layer polarization effects mean that the magnitude of $E_t(t)$ and $\Delta\sigma_q(t)$ will depend on frequency (9). In the linear approximation and neglecting the compact layer, the excess charge can be estimated to be $\Delta\sigma_q = \epsilon \kappa V_d$ where V_d is the induced potential across the double layer (10). We have measured the complex impedance of the electrode array and analyzed the results by treating the electrolyte/double layer as impedances in series. These measurements show that at high frequencies the

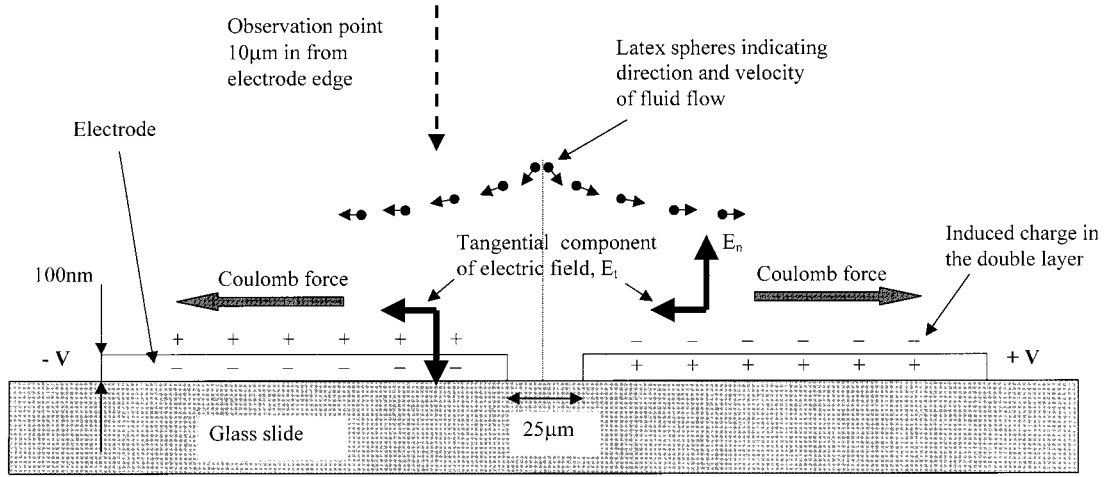


FIG. 1. Diagram illustrating a cross-section of the microelectrode used to measure particle velocities. The figure also illustrates how the tangential component of the electric field can exert a force on the double layer and cause fluid motion. The direction of movement of the latex spheres on the electrode surface is also shown.

potential is dropped entirely across the electrolyte, while at low frequencies the applied potential is dropped almost entirely across the double layer, showing that our electrodes are close to being perfectly polarizable at these frequencies. As a result, since V_d and $\Delta\sigma_q(t)$ tend to zero at high frequencies, Eq. [1] indicates that AC electroosmotic flow will stop. At low frequencies, the electric field in the medium tends to zero implying that, since the tangential field must be continuous, $E_t(t)$ in the double layer is also zero and again, AC electroosmotic flow will cease. Therefore, the velocity profile for AC electroosmosis in perfectly polarizable electrodes is zero at high and low frequencies and maximum at an intermediate characteristic frequency (11), which is similar to the measured frequency dependent fluid velocity (Fig. 2).

Further analysis can be performed using the circuit model shown in Fig. 3, which can be analyzed to give the frequency dependence of V_d and the velocity profile across the electrode. For a small electrode gap, the system can be modeled with the electrolyte represented as discrete current flux tubes in parallel, with the electric field lines assumed to lie mainly along the circumferences of the flux tubes. The double layer is represented as a distributed capacitor of constant value $\Delta C = \epsilon\kappa d\Delta z$, where d is the electrode length (2 mm). At low frequencies ($\omega \ll \sigma/\epsilon$, where σ is the conductivity) the imped-

ance of the electrolyte is purely resistive and the resistance of each tube is $R(z) = \pi z/\sigma d\Delta z$. Since each incremental flux tube is different in length, each incremental capacitor will charge in a different time. The time constant of the incremental circuit elements is given by the product of the capacitance and half the resistance of the tube. The voltage across the double layer as a function of distance z can then be obtained,

$$V_d(z) = \frac{V_o}{2 + j\omega\pi z \frac{\epsilon}{\sigma} \kappa}, \quad [2]$$

where V_o is the amplitude of the AC potential. The charge distribution and tangential electric field can be evaluated from this model as $\Delta\sigma_q(z) = \epsilon\kappa V_d$ and $E_t = -\partial V_d/\partial z$. Estimating the time averaged electroosmotic velocity from Eq. [1] gives

$$\langle v \rangle = \frac{1}{2} \text{Re} \left\{ \frac{\Delta\sigma_q E_t^*}{\eta\kappa} \right\} = \frac{1}{8} \frac{\epsilon V_o^2 \Omega^2}{\eta z (1 + \Omega^2)^2}, \quad [3]$$

where

$$\Omega = \omega \frac{\epsilon}{\sigma} \frac{\pi}{2} z \kappa. \quad [4]$$

Equation [3] gives a velocity profile which tends to zero at low and high frequency limits with a maximum velocity at $\Omega = 1$. As Fig. 2 shows, the velocity curves for the different z appear to have their maxima at the same position along the ωz -axis. Using Eq. [4], the value of ωz for which $\Omega = 1$ can be calculated to be $0.094 \text{ m} \times \text{rad/s}$ for an electrolyte conductivity of 8.6 mS m^{-1} . The calculated ωz matches the value determined by inspection of the experimental graph. Although the predicted velocity magnitudes are higher than the experimental data, the variation in magnitude with z is approximately $1/z$ as indicated by Eq. [4]. Equation [4] also indicates that the frequency for the maximum velocity should increase with conductivity as has been observed previously (11).

The experimental measurements presented in this paper demonstrate the existence of a previously unexamined type of fluid flow in an aqueous system. Qualitative evidence has been presented to indicate that the mechanism gov-

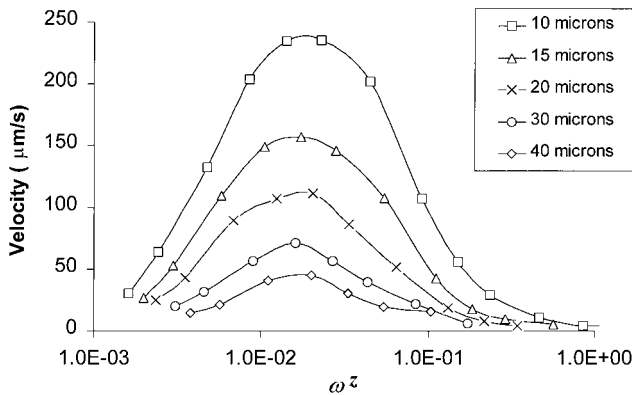


FIG. 2. A graph showing the fluid velocity measured using latex spheres at a number of positions z on the electrode surface. The velocity is plotted against the product of the frequency and the distance of measurement from the center of the two plates, i.e., $22.5 \mu\text{m}$ for a measurement position of $10 \mu\text{m}$.

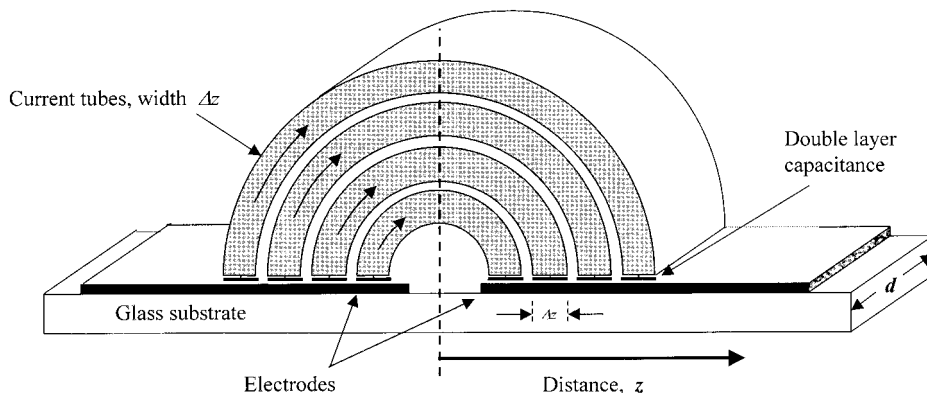


FIG. 3. Diagram illustrating the concept of current flux tubes used in the simple model for calculating the velocity of the fluid. In this model the electrode gap is considered to be very small and the double layer entirely capacitive with a constant value.

erning the fluid flow is related to electroosmosis and electrode polarization. The phenomenon has therefore been termed AC electroosmosis. A simple model based on this idea has been presented which describes the frequency variation of the fluid velocity accurately but not the correct order of magnitude. In order to account fully for the observed flow, a rigorous theoretical approach, possibly considering nonlinear effects in the double layer capacitance, electrode geometry, and the charge, velocity, and electric field distributions, is required. A research study, with more detailed measurements in a range of medium conductivities and experimental conditions, is being carried out in order to better characterize AC electroosmotic flow.

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Antonio Ramos,*
Hywel Morgan,^{†1}
Nicolas G. Green,*
Antonio Castellanos*

*Departamento de Electronica y Electromagnetismo
Universidad de Sevilla
Avda. Reina Mercedes, s/n
41012-Sevilla, Spain
[†]Bioelectronics Research Centre
Department of Electronics and Electrical Engineering
University of Glasgow
Glasgow G12 8LT
United Kingdom

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¹ To whom correspondence should be addressed.