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Conditions for Similitude between the Fluid Velocity and Electric Field in Electroosmotic Flow

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Electroosmotic flow is fluid motion driven by an electric field acting on the net fluid charge produced by charge separation at a fluid−solid interface. Under many conditions of practical interest, the resulting fluid velocity is proportional to the local electric field, and the constant of proportionality is everywhere the same. Here we show that the main conditions necessary for this similitude are a steady electric field, uniform fluid and electric properties, an electric Debye layer that is thin compared to any physical dimension, and fluid velocities on all inlet and outlet boundaries that satisfy the Helmholtz−Smoluchowski relation normally applicable to fluid−solid boundaries. Under these conditions, the velocity field can be determined directly from the Laplace equation governing the electric potential, without solving either the continuity or momentum equations. Three important consequences of these conditions are that the fluid motion is everywhere irrotational, that fluid velocities in two-dimensional channels bounded by parallel planes are independent of the channel depth, and that such flows exhibit no dependence on the Reynolds number. Similitude is demonstrated by comparing measured and computed fluid streamlines with computed electric flux lines.

Microfluidic systems are finding increasing use in the separation, identification, and synthesis of a wide range of chemical and biological species.[1−3] Employing transverse channel dimensions in the range from several to a few hundred micrometers, such systems may permit the large-scale integration of wet analytical methods in a manner analogous to that already achieved in microelectronics. Applications for microfluidics now under development include such diverse processes as DNA sequencing, immunochromatography, and the identification of explosives and chemical and biological warfare agents. The analytical methods used in these processes include traditional chromatography, electrochromatography, and electrophoresis.

One promising method for driving fluid motion in microfluidic systems is electroosmosis.[4,5] Here, motion is induced by applying an electric field to a fluid that is bounded by an insulating solid. Since charge separation generally occurs at a fluid−solid boundary, a layer of fluid near the interface carries a net electric charge. The applied field acts on this net fluid charge to produce a body force that drives fluid motion. The region of net charge is usually confined to a thin Debye layer immediately adjacent to the surface, so the body force is nearly coincident with the bounding surface. The resulting boundary or sheath velocities impart a pluglike motion to the remaining neutral fluid inside a channel.

Electroosmotic flows offer several important advantages over pressure-driven flows for the small physical dimensions characteristic of microfluidic systems. First, electroosmosis provides a direct means of transporting fluids in microchannel networks using only applied electric fields. Further, fluid speeds in electroosmotic flows are independent of the transverse tube or channel dimension over a wide range of conditions, making this technique extensible to extremely small physical scales. In contrast, pressure-driven flows require that the pressure gradient increase inversely with the square of the transverse channel dimension to maintain a given fluid speed. Finally, the profile of the fluid velocity across a channel is essentially flat whenever the Debye layer thickness is small compared to the channel width. Because of this, analyte samples can be transported over long ranges with little hydrodynamic dispersion.[6−9]

Direct numerical simulation of electroosmotic flow is a challenging task.[10,11] In addition to the usual Navier−Stokes and species transport equations, the electric field equation must also be solved, and these equations are generally coupled through the unknown charge density. Further, these solutions must resolve length scales ranging from a Debye layer thickness of perhaps 10 nm, to channel widths on the order of 100 μm, and to device lengths of nearly 10 cm. These widely disparate length scales, spanning roughly 7 orders of magnitude, make traditional numerical meshes impractical for all but the very simplest geometries. Patankar and Hu[10] previously discussed this problem and found a practical remedy in artifically increasing the thickness of the Debye layer by 3 orders of magnitude.

These disparate length scales persist in the problem even when the fluid transport and electric potential are not coupled, necessitating either a highly nonuniform computational mesh or the

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use of multiple meshes to address separately the Navier−Stokes and Poisson equations. One means for avoiding this problem is to assume that the Debye layer thickness is very small compared to any channel dimension, as done by Ermakov et al.[11] In this case, the electric potential is governed by the much simpler Laplace equation, but the Navier−Stokes equations describing fluid motion must still be solved.[11,12] This complexity in solving the most general equations governing electroosmotic flow motivates our present interest in one unique aspect of these flows: under fairly broad conditions of practical interest, the steady velocity field of an electroosmotic flow is uniformly proportional to the electric field.

Such similitude between the fluid velocity and electric field was previously revealed by Morrison[12] for the motion of a single particle in an unbounded fluid. In that work, he showed that the electrophoretic particle speed was independent of the particle shape and that fluid motion outside the Debye layer was irrotational provided that the particle was free of external forces and moments other than those induced by the applied electric field. Here we examine internal flows bounded by the walls of channel or capillary networks and derive the conditions necessary and sufficient for similitude. When these conditions are met, both the electric potential and fluid velocity fields can be computed by solving only the Laplace equation.

PRELIMINARY OBSERVATIONS

Many of the conditions necessary for similitude between the electroosmotic fluid velocity and applied electric field can be discerned by counterexample. For instance, body forces acting on the fluid appear only near fluid−solid boundaries when the Debye layer is thin, so an electric field suddenly applied will produce a fluid transient as the neutral fluid accelerates. Such a transient cannot exhibit similitude with the steady electric field; thus, we see that the velocity and electric fields must be at least quasi-steady. Applied electric fields ramped at sufficiently low rates can, of course, satisfy this condition.

Similarly, any applied pressure difference between the ends of a channel will produce a velocity profile that is at least in part parabolic. This nonuniform fluid velocity is clearly not similar to a uniform electric field, so a uniform pressure on all inlet and outlet boundaries is generally required for similitude. Flows induced by gravity likewise cannot resemble an applied uniform electric field. When the thickness of a channel is smaller than the Debye length, a net charge is present everywhere in the fluid. In this case, an applied uniform electric field produces a uniform body force resulting in a parabolic velocity profile like that of flows driven by pressure or gravity. Only when Debye layers are thin can the velocity field in the neutral fluid be uniformly proportional to the applied electric field. Thus, thin Debye layers are necessary for similitude.

Similitude of the two fields also requires several less stringent conditions. These include uniform density, viscosity, and conductivity of the neutral fluid, a uniform surface potential at the fluid−solid interface, and a Debye layer conductance in the direction of fluid motion that is very small compared to that of the neutral fluid. In all but very specialized cases, the channel walls must also be impermeable, and the conductivity of the solid must be negligible compared to that of the neutral fluid. Counterexamples supporting these requirements are apparent from the results that follow.

Finally, the fluid velocity on all inlet and outlet boundaries must also be proportional to the electric field. That this condition is necessary follows directly from the definition of similitude: the flow is uniformly proportional to the electric field. Here we show that this condition, along with the necessary conditions above, is sufficient for global similitude between the fluid velocity and electric field.

EQUATIONS DESCRIBING ELECTROOSMOTIC FLOW

To derive the conditions sufficient for similitude, we consider a simply or multiply connected fluid volume, bounded in its entirety by a mix of two surface types. The surfaces S_1 describe the interface between the fluid and an impermeable insulating solid, while the surfaces S_2 describe inlet or outlet boundaries.

The electric potential, ϕ, within this volume is governed by the Poisson equation relating the divergence of the electric field to the local charge density,

$$\nabla\cdot(\epsilon\nabla\phi) = -\rho_e \tag{1}$$

where ϵ is the dielectric constant of the fluid and ρ_e is the local charge density. The local charge density may be related to the electric potential through the Boltzmann distribution or similar relations.

Boundary conditions for the electric potential on the charged surface may be specified either through a prescribed density of the surface charge in conjunction with the governing eq 1 or, alternatively, as a prescribed wall potential with respect to the potential of the adjacent neutral fluid. Finally, the applied electric field is generally specified by prescribed potentials on inlet and outlet boundaries.

Restricting our attention to liquid flows, the working fluid may be considered incompressible and the continuity equation reduces to

$$\nabla\cdot\mathbf{u} = 0 \tag{2}$$

Under the additional assumptions that the fluid velocity, $\mathbf{u}$, is steady and that the viscosity, μ, is constant, the momentum equation becomes

$$\rho(\mathbf{u}\cdot\nabla)\mathbf{u} = -\nabla p + \rho_e\nabla\phi + \mu\nabla^2\mathbf{u} \tag{3}$$

where ρ is the uniform fluid density. Using well-known vector identities,[14] the momentum equation can also be written as

$$-\rho\mathbf{u}\times(\nabla\times\mathbf{u}) = -\nabla\left(p + \frac{\rho}{2}\mathbf{u}\cdot\mathbf{u}\right) + \rho_e\nabla\phi - \mu\nabla\times(\nabla\times\mathbf{u}) \tag{4}$$

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This form of the equation is again generally applicable only to an incompressible fluid having constant viscosity.

Boundary conditions on the fluid velocity follow directly from the nature of the fluid−solid interface. Since no flow crosses the impermeable solid boundaries,

$$\mathbf{u}\cdot\hat{\mathbf{n}} = 0 \quad \text{on} \quad S_1 \tag{5}$$

where $\hat{\mathbf{n}}$ is a unit vector locally normal to the interface. The velocity tangential to these impermeable boundaries must also obey the no-slip condition at the fluid−solid interface,

$$\mathbf{u}\cdot\hat{\boldsymbol{\tau}} = 0 \quad \text{on} \quad S_1 \tag{6}$$

where $\hat{\boldsymbol{\tau}}$ is any unit vector lying in the plane of the boundary. We will next consider an alternate form of eq 6 describing the fluid velocity at the outer edge of the Debye layer.

THIN DEBYE LAYER LIMIT

Assuming that the Debye layer thickness, λ, is much smaller than the channel width, a, a boundary-layer approximation may be used to sequentially solve for the velocity fields in the regions within and outside the Debye layer. Under all foreseeable conditions, the Reynolds number based on the Debye layer thickness is extremely small, so the inner Debye layer solution can be constructed by considering only the balance between electric and viscous forces. The maximum velocity at the outer edge of the layer is then used as a boundary condition in calculating the larger scale outer flow field. Sequential solution procedures of this type are best justified using the formalism of matched asymptotic expansions.[13] Here we present only an outline of the matching procedure for first-order terms.

To identify nonessential terms, the governing equations are first rewritten in terms of normal and tangential coordinates rescaled by the Debye layer thickness and the geometric length scale, respectively. The electric potential is then split into an applied field $\phi_a(\tau)$ having no normal gradients and an intrinsic component $\phi_i(\eta)$ having no tangential gradients or at most a tangential gradient induced by linear polarization that is proportional to the applied field. In the latter case, polarization of the intrinsic field can be accounted for by a suitable choice of the dielectric constant. After dropping those terms of order λ/a and smaller, the resulting Poisson and momentum equations are combined to obtain a balance between viscous and body forces,

$$\mu\frac{d^2\mathbf{u}}{d\eta^2} = -\epsilon\frac{d^2\phi_i}{d\eta^2}\nabla\phi_a \tag{7}$$

where η is the scaled coordinate normal to the surface. Since the intrinsic field exists only within the Debye layer and gradients within this layer are much steeper than those outside, ϕ_i, $d\mathbf{u}/d\eta$, and $d\phi_i/d\eta$ all must vanish as $\eta \to \infty$. Subject to these boundary conditions and the assumption of constant fluid properties, eq 7 can be integrated twice to obtain the Helmholtz−Smoluchowski equation relating the velocity at the outer edge of the Debye layer, $\mathbf{u}_\infty$, to the applied field gradient, $\nabla\phi_a$, and the zeta potential, ζ, describing the difference in potential across the Debye layer.[4]

$$\mathbf{u}_\infty = -\frac{\epsilon\zeta}{\mu}\nabla\phi_a = \alpha\nabla\phi_a \tag{8}$$

This relationship is best viewed as a defining equation for the electroosmotic mobility, $\alpha = \epsilon\zeta/\mu$.[4,11] Since the mobility is relatively constant for a given fluid and surface, eq 8 serves as a convenient boundary condition relating the local fluid velocity to the electric field along all S_1 surfaces.

The electroosmotic mobility, α, can be obtained directly by measuring the flow rate through a simple channel for a known applied field, without specific knowledge of the viscosity, dielectric constant or zeta potential appearing in eq 8. This practical approach avoids an otherwise complex problem since the viscosity and dielectric constant may vary substantially in crossing the Debye layer, and the zeta potential cannot be measured or computed readily. Moreover, these details are not important to the flow outside the Debye layers, provided that these layers remain thin compared to lateral channel dimensions. Under this restriction, and our earlier assumption of uniform fluid and surface properties, the mobility will be the same on all surfaces, regardless of channel geometry.

Outside the Debye layer, the charge density and intrinsic field ϕ_i both vanish. The applied electric potential in this region is thus governed by the Laplace equation

$$\nabla^2\phi = \nabla^2\phi_a = 0 \tag{9}$$

subject to prescribed potentials on S_2 boundaries and a zero-flux condition on all S_1 boundaries. For any nonzero fluid conductivity, the latter is equivalent to

$$\nabla\phi\cdot\hat{\mathbf{n}} = 0 \quad \text{on} \quad S_1 \tag{10}$$

where again $\hat{\mathbf{n}}$ is a unit vector locally normal to the interface. Fluid velocities on S_1 are given by the Helmholtz−Smoluchowski relation, so it remains only to specify velocity conditions on the inlet and outlet boundaries, S_2.

In splitting the inner and outer regions we have neglected terms of order λ/a and smaller. One such term involves convective transport normal to the surface. As a result, no account is made of charge redistribution resulting from convective charge transport. However, these polarization effects are unimportant provided that the Peclet number based on the Debye layer thickness, electroosmotic speed, and ion diffusivity remains small. This is expected in most practical applications. Convective charge transport tangential to the surface cannot influence charge distribution within the Debye layer so long as this distribution does not vary along the surface.

CONDITIONS FOR SIMILITUDE

From continuity and the Laplace equation governing the electric potential in the neutral fluid outside the Debye layer we can write

$$\nabla\cdot(\mathbf{u} + \alpha\nabla\phi) = 0 \tag{11}$$

where the electroosmotic mobility, α, is a constant. This expression applies without any loss of generality since $\nabla^2\phi = 0$ describes the electric field in this region and $\nabla \cdot \mathbf{u} = 0$ describes continuity. The term $\mathbf{u} = \nabla\phi$ is thus solenoidal,[14] and hence there exists some vector function $\boldsymbol{\psi}$ such that

$$\mathbf{u} = -\alpha\nabla\phi + \nabla \times \boldsymbol{\psi} \tag{12}$$

The condition for similitude between the local electric field and the local fluid velocity is therefore equivalent to the condition that $\nabla \times \boldsymbol{\psi} = \mathbf{0}$. When this latter condition is satisfied at all points in the flow, the steady fluid velocity is proportional to the electric field and the constant of proportionality is everywhere the same.

Since the fluid outside the Debye layer is electrically neutral, the body force $\rho_e\nabla\phi$ in this region vanishes. Dropping this term, the momentum eq 4 may be rewritten in terms of the vorticity,

$$-\rho\mathbf{u} \times \boldsymbol{\omega} = -\nabla\left(p + \frac{\rho}{2}\mathbf{u}\cdot\mathbf{u}\right) - \mu\nabla \times \boldsymbol{\omega} \tag{13}$$

where the vorticity $\boldsymbol{\omega}$ is

$$\boldsymbol{\omega} \equiv \nabla \times \mathbf{u} = \nabla \times (\nabla \times \boldsymbol{\psi}) \tag{14}$$

Note that the far right side of this result is obtained by dropping terms of the form $\nabla \times \nabla\phi$ since they are identically zero.[14] Taking the curl of eq 13 likewise eliminates the gradient of the total pressure on the right-hand side, yielding

$$\rho\nabla \times (\boldsymbol{\omega} \times \mathbf{u}) = \mu\nabla^2\boldsymbol{\omega} \tag{15}$$

The corresponding elliptic equation governing the vector function $\boldsymbol{\psi}$ is given directly by the definition of the vorticity in eq 14. The coupled eqs 12, 14, and 15 thus generally describe incompressible flow within any three-dimensional volume. There are no restrictions on the shape of this volume, and the domain of the volume may be either simply or multiply connected.

Boundary conditions for the function $\boldsymbol{\psi}$ are derived from the primitive conditions on the normal and tangential components of the fluid velocity. Because the boundary condition on the tangential fluid velocity is automatically satisfied by the first term on the right of eq 12, the remaining portion of the velocity must satisfy

$$\nabla \times \boldsymbol{\psi} = \mathbf{0} \quad \text{on} \quad S_1 \tag{16}$$

Note that this condition also satisfies the requirement that the normal component of the fluid velocity vanish everywhere on the impermeable fluid−solid boundary. Also note the eq 16 makes no statement about the fluid vorticity on the S_1 surface. Despite the fact that $\mathbf{u} = -\alpha\nabla\phi$ everywhere on this boundary, it is not necessarily the case that $\boldsymbol{\omega} = -\alpha\nabla \times (\nabla\phi)$ vanishes. Although this appears to be the curl of a gradient, which would ordinarily be identically zero, such is not the case since $\mathbf{u} = -\alpha\nabla\phi$ applies only on the boundary and therefore conveys nothing of how the velocity varies moving into the fluid. Thus it is impossible to make any a priori claim that vorticity vanishes at the S_1 slip boundaries of an electroosmotic flow.

To show the conditions under which $\nabla \times \boldsymbol{\psi}$ is identically zero, we now substitute eq 14 into eq 15 yielding

$$\rho\nabla \times [(\nabla \times \xi) \times \mathbf{u})] = \mu\nabla^2(\nabla \times \xi) \tag{17}$$

where

$$\xi \equiv \nabla \times \boldsymbol{\psi} \tag{18}$$

From eq 16, the boundary condition for ξ is

$$\xi = \mathbf{0} \quad \text{on} \quad S_1 \tag{19}$$

Given this homogeneous governing equation and boundary condition on S_1, we see that a necessary and sufficient condition for $\xi = \mathbf{0}$ everywhere is that $\xi = \mathbf{0}$ on the inlet and outlet boundaries S_2. By eq 12, this is equivalent to requiring

$$\mathbf{u} = -\alpha\nabla\phi \quad \text{on} \quad S_2 \tag{20}$$

Thus, the fluid velocities on all inlet and outlet boundaries must satisfy the Helmholtz−Smoluchowski relation normally applicable to fluid−solid boundaries. This result is the most general condition for similitude between the fluid velocity and electric field. When this condition is satisfied, along with the other restrictions previously described, the fluid velocity is uniformly proportional to the gradient of the electric potential.

CONSEQUENCES

Similitude between the fluid velocity and electric field has several important consequences. First, the flow is everywhere irrotational outside the Debye layer, regardless of the geometry of bounding surfaces. All fluid motion within the domain is simply potential flow, and the vorticity defined by eq 14 is exactly zero. Such flows cannot manifest closed cells of fluid recirculation. Thus, electroosmotic flows satisfying the conditions for similitude cannot contain any vortex, regardless of how complex the geometry may be. The absence of vorticity further implies, according to eq 13, that the total pressure is everywhere uniform and is therefore the same on all inlet and outlet boundaries. This automatically holds when the condition $\mathbf{u} = -\alpha\nabla\phi$ is satisfied on these boundaries.

Second, the velocity field under the conditions for similitude exhibits no dependence on the Reynolds number. This is because any potential flow satisfies the full Navier−Stokes equations for all Reynolds numbers. These solutions thus remain valid even when inertial forces are not negligible.

Finally, under the conditions for similitude, the velocity field in any two-dimensional channel is independent of the channel depth, provided that the depth is uniform and the electric field is vertically uniform on all inlet and outlet boundaries. For similitude to hold, the top and bottom boundaries of these two-dimensional channels must be electrically insulated planes. Given these restrictions, the electric field can have no vertical component and all horizontal planes through the channel are equivalent. The electric field, and hence the fluid velocity, are then truly two-dimensional and will show no dependence on the channel depth. This was demonstrated for a special case by the three-dimensional computational results of Patankar and Hu.[10] In that case, they report that the velocity in the third direction is very small and that the fluid moves on planar layers.

PRACTICAL REALIZATION OF SIMILITUDE

The condition $\mathbf{u} = -\alpha\nabla\phi$ on the surfaces S_2 is effectively satisfied in many practical devices employing electroosmotic flow. This condition naturally arises for flow in any long channel between fluid reservoirs maintained at fixed potentials by embedded electrodes. Such arrangements are common in electrochromatographic systems. Although this S_2 boundary condition may not be satisfied very near an electrode and, in fact, cannot be satisfied on an electrode surface, the flow field in a long narrow channel should relax to satisfy this condition within a few channel widths of its ends. This condition may thus be satisfied on the interior of a complex channel network, including turns and junctions, even if it is not satisfied in the immediate vicinity of electrodes.

In considering conditions on the boundaries S_2, we must keep in mind that these inlet and outlet surfaces need not be the physical ends of a channel or channel network. The surfaces S_2 are simply defined as open boundaries that may be crossed by the fluid, so their positions can be specified rather arbitrarily. Thus, any portion of a device bounded in its entirety by S_1 and S_2 surfaces will exhibit similitude between the velocity and electric field when the necessary conditions are satisfied and $\mathbf{u} = -\alpha\nabla\phi$ on all specified S_2 surfaces. It follows immediately that any subset of such a domain will also exhibit similitude since the velocities on all S_2 boundaries of the subset must satisfy $\mathbf{u} = -\alpha\nabla\phi$. As such, the flow in components of a channel network, such as expansions, contractions, and junctions, can be analyzed separately using solutions to the Laplace equation without simulating the network in its entirety.

One of the necessary conditions for similitude is that all S_1 surfaces are electrically nonconductive relative to the working fluid. While this condition is readily satisfied by many materials of practical importance, it contains a hidden and somewhat subtle constraint. No electrodes or conductive elements may appear within the domain bounded by S_1 and S_2 if similitude is to hold in a rigorous sense. Thus, even if there exists a set of S_2 boundaries satisfying the sufficient condition for similitude, an imbedded electrode within the domain will violate the necessary condition that no electric currents cross the S_1 boundaries. From a practical view, this means that some devices, such as electrokinetic pumps, may never exhibit similitude.

As described above, one consequence of similitude is that the total pressure is the same on all inlet and outlet boundaries. This uniformity may thus also be viewed as a necessary condition for similitude to hold. At low fluid speeds, this new condition is readily satisfied by holding the static pressures fixed and uniform on the inlet and outlet boundaries. However, the required equality of the total pressures becomes more difficult to ensure as the dynamic pressure increases. For example, a difference in cross-sectional area between a channel inlet and outlet yields differing fluid speeds at the two ends. If the dynamic portion of the total pressure is not negligible, this produces a difference in the total pressures. In such a case, the static pressures must be adjusted to ensure that the total pressures are once again equal. This, of course, would not be very practical. Fortunately, the fluid speeds in most real applications are sufficiently small that dynamic contributions to the total pressure are negligible from a practical point of view.

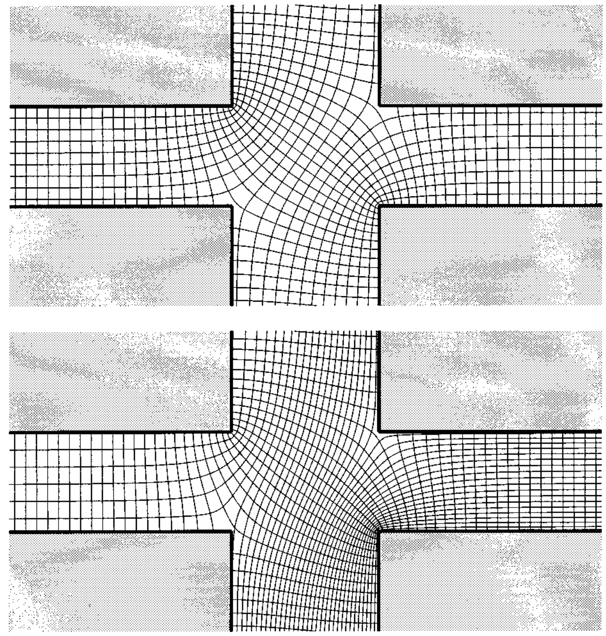

Figure 1. Computed fluid streamlines and electric isopotentials for electroosmotic flow in an intersection. Conditions for the top frame yield a potential flow; those for the bottom frame do not owing to a variable zeta potential.

DEMONSTRATION OF SIMILITUDE

To demonstrate similitude in complex geometries, we now consider two sample problems. The first of these involves only computational results, while the second employs experimental observations as well.

The first sample problem is illustrated in the two frames of Figure 1. In both frames, the geometry is the same and consists of the intersection of two deep channels having differing widths. Only a portion of the computational domain is shown in these images. The channels actually extend about four channel widths from the intersection. Boundary conditions for both cases are a unit potential at the ends of the left and bottom channel segments; the ends of the right and top segments are grounded.

Two sets of curves are shown in each of the frames of Figure 1. Those curves generally orthogonal to the channel walls are electric isopotentials, uniformly spaced in potential. Those curves generally following the channel walls are fluid streamlines, again spaced at uniform increments of the stream function. The potential field in both cases was computed using a two-dimensional finite-difference form of eq 9. Also, in both cases, the velocity field was computed using a finite difference form of eqs 14 and 15, along with boundary conditions given by eq 8 using the local electric field. The streamlines shown in both frames were thus computed by solving the stream function and vorticity form of the Navier–Stokes equations without any assumptions regarding similitude between the local electric field and fluid velocity in the interior of the intersection.

The results shown in the top frame of Figure 1 are for the case in which the properties of the fluid entering the left and lower channel segments are the same and the zeta potential is uniform

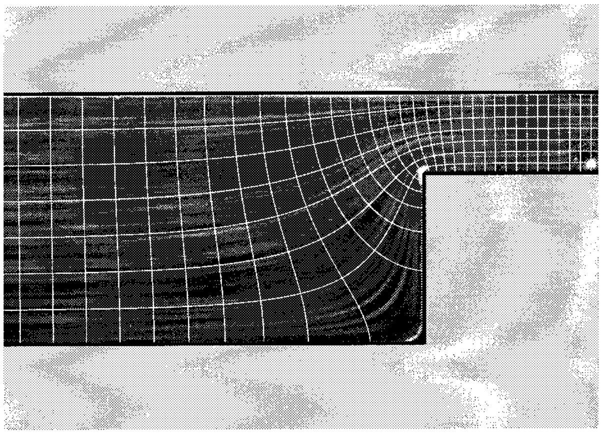

Figure 2. Streamline image overlaid with computed isopotentials and electric flux lines for electroosmotic flow into a restriction. Orthogonal intersection of streamlines and isopotentials indicates a potential flow.

along all channels boundaries. In this case, the necessary conditions for similitude are satisfied. As a result, we see that the streamlines are everywhere orthogonal to the isopotentials, indicating that the fluid velocity is proportional to the local electric field. Further, we see that the aspect ratios of the cells bounded by the streamlines and isopotentials are everywhere the same, indicating that the constant of proportionality is spatially uniform. This is therefore a potential flow, and the same flow field could have been obtained by solving the Laplace equation alone. These results further demonstrate that similitude can be realized in complex geometries of practical importance.

Results in the lower frame of Figure 1 illustrate a case in which the fluid entering from the bottom channel segment yields a zeta potential 4 times that of the fluid entering from the left. All other fluid properties of the two inlet streams were assumed to be the same. In this case, we see that the streamlines and isopotentials are reasonably orthogonal in regions removed from the intersection but are decidedly not perpendicular where the two channels meet. Moreover, it is clear that the aspect ratio of the enclosed cells in this case is not uniform. Both of these indicate that this is not a potential flow, and the fluid velocities must be computed from the Navier−Stokes equations.

Our second sample problem is illustrated in Figure 2. The geometry shown consists of a step constriction at the junction of two channel segments of equal depth but differing width. The mean width of the left-hand segment is ~200 μm and that of the right-hand segment is ~60 μm. The mean channel depth is ~10 μm, and the total channel length is 20 mm. Only a portion of the channel near the junction is shown.

The channel was formed by isotropically etching a glass substrate and then thermally bonding a cover of the same material onto the top. The solution in the channel is millimolar phosphate-buffered saline solution with a low concentration of micrometer-sized latex particles used as fluorescent flow tracers. Electroosmotic flow was induced in the channel by applying 50 V across gold electrodes submerged in open reservoirs at the channel ends.

The light and somewhat diffuse lines in Figure 2 are streak lines formed by the fluorescent tracer particles in a 0.27-s photographic exposure. Because the flow is steady, these streak lines are equivalent to fluid streamlines. The distinct lines overlaying this image are the isopotentials and electric flux lines computed for this geometry by solving only the Laplace equation. The Navier−Stokes equations were not used in producing these curves. We see that the measured streamlines are everywhere perpendicular to the computed isopotentials and that the streamlines and flux lines are nearly parallel at all points in the flow. Thus, the observed velocity field is simply proportional to the electric field.

Note that a small aggregate of particles is attached to the corner of the step. This was not included in the numerical simulation, leading to slight deviations between the streamlines and flux lines in the region close to the corner. Except for these local deviations, the agreement between the observed flow field and computed electric flux lines is excellent, and the streamlines and isopotentials form orthogonal intersections even in the nearly stagnant region on the face of the step. Also note that this flow yields no recirculation cell just upstream of the step. Such a cell would normally be present in other flows, including pressure-driven flows at all Reynolds numbers and, perhaps, electroosmotic flows not satisfying the conditions for similitude. Such a cell is not possible when the conditions for similitude are satisfied since potential flows exhibit no vorticity.

SUMMARY

In many cases of practical interest, the fluid velocity of an electroosmotic flow is proportional to the applied electric field and the constant of proportionality is everywhere the same. Here we have shown that necessary conditions for such similitude include a quasi-steady electric field, uniform fluid density, and uniform viscosity of the neutral fluid outside the Debye layer. The condition of uniform viscosity may be relaxed in the special case in which gradients of the viscosity are everywhere orthogonal to gradients of the fluid velocity. Further, the Debye layer thickness must be small compared to any channel dimension, and the conductance of the layer in the direction of fluid motion must be small compared to that of the neutral fluid. In addition, all solid surfaces bounding the fluid must have a uniform surface charge or surface potential, must be impermeable to flow, and must be electrically nonconducting relative to the fluid. The last of these implies that conductors and electrodes may not appear anywhere within the domain of similitude.

To derive the remaining less-obvious conditions required for similitude, the velocity field was expressed as the sum of a component uniformly proportional to the electric field and a residual component of unknown value. Using the full three-dimensional stream function and vorticity formulation of the steady Navier−Stokes equation, we showed that the unknown residual component vanishes everywhere if and only if the velocity on all inlet and outlet boundaries satisfies the Helmholtz−Smoluchowski relation normally applicable to the fluid−solid interface. This condition, along with the necessary conditions above, is both necessary and sufficient for similitude between the velocity and electric field. The resulting electroosmotic flows are irrotational and therefore have a uniform total pressure. This proof employs no assumptions regarding the geometry of the channel, so the results are equally applicable to simple channels, channel networks, and complex three-dimensional geometries such as those in the interior of packed beds.

When the conditions for similitude are fully satisfied, the resulting fluid motion is simply a potential flow. As a result, the flow is irrotational, the total pressure is uniform, and the fluid velocity field exhibits no dependence on the Reynolds number. However, if dynamic pressures are large, care must be taken to ensure that the total pressures on all inlet and outlet boundaries remain equal. Fortunately, dynamic pressures are rarely significant in systems of practical interest.

The principal benefit of similitude is that modeling these flows is greatly simplified. Both the electric field and the velocity field are obtained from a single solution of the Laplace equation. The Navier–Stokes and Poisson equations need not be solved, and thin boundary layers on the channel walls need not be resolved. Moreover, under the conditions for similitude, flow in two-dimensional channel networks bounded by parallel planes is independent of the channel depth provided that the depth is uniform and the electric potential is vertically uniform on the inlet and outlet boundaries. In this case, the flow is strictly two-dimensional and two-dimensional models may be employed. Thus, an understanding of similitude is useful in modeling for the design and evaluation of microfluidic systems.

Beyond these mathematical benefits, similitude provides useful physical insight into the design and operation of electroosmotic systems. For example, the absence of vorticity generally impedes mixing processes. This was previously recognized as an important benefit of electroosmotic flow in long narrow channels. Here we show that the same benefit can be realized in channel junctions, nozzles, injectors, flow dividers, control devices, and other such components of microfluidic systems. In contrast, irrotational flows may not be desirable when mixing is the goal. The conditions for similitude in this case also provide guidance in the means to produce vorticity by violating one or more of the requirements.

NOMENCLATURE

U	characteristic speed
a	characteristic transverse dimension
l	distance traveled along a streamline
L	length of streamline within domain
S	boundary surface
T	temperature
$\mathbf{u}$	local fluid velocity
α	electroosmotic mobility: $\alpha = \epsilon\zeta/\mu$
ϵ	dielectric constant
ζ	effective surface electric potential
λ	Debye length
μ	kinematic viscosity
$\boldsymbol{\omega}$	vorticity vector
ρ_e	charge density
ϕ	electric potential
$\boldsymbol{\psi}$	stream function vector

Subscripts and Superscripts

1	on fluid–solid boundary
2	on inlet and outlet boundaries
i	intrinsic
a	applied

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