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Comments on the conditions for similitude in electroosmotic flows

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Abstract

This note provides a few comments on the conditions required for similitude between velocity and electric field in electroosmotic flows. The velocity fields of certain electroosmotic flows with relatively thin electric double layers (EDLs) are known to be irrotational in regions outside of the EDL. Under restricted conditions, the velocity field, $\bar{V}$, can be expressed in terms of the electric field, $\bar{E}$, as $\bar{V} = c\bar{E}$, where c is a scalar constant. The irrotationality solution is certainly unique and exact for Stokes flow, but may not be stable (or unique) for flows with Reynolds numbers significantly greater than unity.

Keywords: Electrokinetics; Electroosmosis; Similitude; Microfluidics; Irrotational; Vorticity

1. Background

The main subject of the papers of Cummings et al. [1], Santiago [2], and Oh and Kang [3] are the specification of the sufficient and necessary conditions for similitude, and descriptions of sources of vorticity. The Navier–Stokes formulation for electroosmotic flow (EOF) of a liquid outside of the EDL in an arbitrary geometry can be expressed in non-dimensional form as [2]

$$\nabla \cdot \bar{V}' = 0, \tag{1}$$

$$\mathrm{St}\,\mathrm{Re}\,\partial \bar{V}'/\partial t' + \mathrm{Re}\bar{V}' \cdot \nabla' \bar{V}' = -\nabla' p' + \nabla'^2 \bar{V}'. \tag{2}$$

Here Re and St are the characteristic Reynolds and Strouhal numbers and pressure, p, has been scaled by a viscous stress [4]. For relatively thin EDL, the mobile charge regions at the interface between walls and the liquid can be modeled as a slip layer with the following local property [2]:

$$\bar{V}' = \mu_0' \bar{E}', \tag{3}$$

where μ_0' is a non-dimensional EOF mobility. This boundary condition applies to the slip plane (bounding flow regions that exclude the EDL), and to inlets and outlets. This approximate condition supports both a slip and a shear stress that can balance pressure and inertial forces (i.e., there is no imposed restriction on the derivative of $\bar{V}$).

2. Mathematical aspects of irrotational electroosmotic flows

Conditions required [2] for irrotationality of the "outer flow" (outside EDL) velocity field, $\bar{V}'$, include uniform liquid properties (including uniform permittivity, viscosity, and conductivity); uniform electroosmotic mobility; an EDL thickness relatively thin compared to the geometry (so Eq. (3) may be used); electrically insulating and impermeable channels walls, parallel flow at inlets and outlets; and equal total (stagnation) pressure at all inlets and outlets. The three papers discussed above agree on these as necessary conditions. The following conclusions may be drawn:

- If we add the restrictions that the flow has both negligible Re and St Re, then $\bar{V}' = \mu_0' \bar{E}'$ is an easily verifiable and unique solution to outer flow Eqs. (1)–(3) (solutions to the Laplace equation are unique). The (very useful) irrotational solution has been validated with flow visualization experiments in various studies including [5–8].
- Despite the arguments of Oh and Kang, a proven and accepted method of exploring solutions of differential equa-

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tions is to propose the form of a solution and apply this to the governing equation [9] (e.g., this is indeed the strategy behind similarity solutions in fluid mechanics). We can thereby explore the validity and ramifications of our assumption. If we propose a solution of the form, $\bar{V}' = c\bar{E}'$ for Eqs. (1)–(3), we come up with a condition [2] that $\mathrm{St\,Re}\,\partial\bar{V}'/\partial t' + \mathrm{Re}\bar{V}' \cdot \nabla'\bar{V}' = -\nabla' p'$, the Euler formulation, which can here be interpreted as a condition required for $\bar{V}' = \mu_0'\bar{E}'$ (and not a governing equation). Evaluating Eq. (3), we obtain $c = \mu_0$. A key question: Is $\bar{V}' = \mu_0'\bar{E}'$ a solution to the full Eqs. (1)–(3) for all values of Re and St Re? This question is discussed below.

- For $\mathrm{Re} \to 0$ but finite St Re, $\bar{V}' = \mu_0'\bar{E}'$ is certainly not a general solution to Eqs. (1)–(3). This has been shown by various experimental and numerical studies of EOF with suddenly applied [10–12] and oscillating electric fields [10,13]. Indeed, it is difficult to imagine an "unsteady Stokes," startup problem that is irrotational.
- For $\mathrm{St\,Re} \to 0$ (quasi-steady) but finite Re, Cummings et al. [1] argue that $\bar{V}' = \mu_0'\bar{E}'$ is a general solution to Eqs. (1)–(3) for all Re. In my paper [2], I argue that $\bar{V}' = \mu_0'\bar{E}'$ is in fact a solution to Eqs. (1)–(3), but there is no assurance that this solution is unique. That is, $\bar{V}' = \mu_0'\bar{E}'$ may not be a stable solution observable at finite Re. (One analogy here is pressure-driven Poisseuille flow in a cylindrical channel [4]; where the parabolic velocity profile is a mathematical solution at all Re but is not stable or observable at high Re.) Is $\bar{V}' = \mu_0'\bar{E}'$ a unique solution for EOF at all Re? I submit that this is an open problem.
- Oh and Kang's [3] proposition that irrotationality is first broken when vorticity advects out of the EDL and into the outer flow is very interesting. The argument is consistent with their simulations of flows near channel corners and should be explored further. The effect they show on EDL thickness is also interesting and new. One caveat is that their simulations show significant deviation at $\mathrm{Re} > 100$, which may be difficult to achieve experimentally.
- One subtle but interesting issue is that, if the EDL has a finite thickness, the conductivity of the liquid must be uniform throughout the entire liquid domain (inside and outside the EDL) for irrotationality. At zeta potentials on the order of (or larger) than the thermal voltage [14], the conductivity in the EDL can be significantly higher than that of the outer flow. In such cases, area-averaged conductivity of a channel is a function of its cross-sectional area; so electric field lines can pass from regions of relatively high conductivity to regions of low conductivity (e.g., into and out of the EDL). This situation can clearly generate internal pressure gradients [15] and net charge regions that couple with applied fields to inject vorticity outside of the EDL [16,17].

3. Observable physics

- A fair question: Is the Re dependence of EOF observable in a laboratory? It will be very difficult. Typical EOFs have Re of order unity or less. The electric fields and channel dimensions needed for $\mathrm{Re} > 10$ quickly develop strong temperature gradients due to Joule heating [18,19]. For example, consider a channel with a depth h that is smaller than its width w. Conduction heat flux, Joule heating, and Re all scale as h, so temperature raise in the liquid scales as $\Delta T \approx \kappa \mathrm{Re}^2$. Here $\kappa = (1/2)\sigma\nu^2/(\mu^2 k)$, where σ is ionic conductivity, ν is kinematic viscosity, k is thermal conductivity, and μ is electroosmotic mobility (velocity per field). κ varies from 3 °C for 1 mM aqueous electrolytes to order 4×10^{-3} °C (best case) for acetone and acetonitrile [20–22]. This suggests a maximum Re of roughly 40 can be achieved with only a few degrees temperature rise. Limiting the maximum field to 10^6 V/m, this requires an $h = \nu\,\mathrm{Re}/(\mu E) \sim 600$ µm or greater. Such length and electric field scales are highly susceptible to flow instabilities (see below).
- Further complication is that flow seeding (e.g., with fluorescent microspheres for micron-resolution particle image velocimetry [23]) of EOFs of organic liquids may be very difficult as these liquids can dissolve and swell polystyrene, latex, etc. Colloidal stability of these suspensions may also be a challenge.
- Even a few degrees temperature rise is probably enough of a conductivity gradient to invalidate irrotational flows, and may generate electrokinetic flow instabilities. The latter are electrohydrodynamic instabilities in the EOF regime due to conductivity gradients [16,24], and which can couple with EOF to generate advective and absolute instabilities [17,25]. We observe these instabilities in channels of 100 µm depth with conductivity gradients caused by just a few degrees temperature rise (due to Joule heating) and electric fields of order 10^5 V/m [26]. The critical field for unstable flow scales as h^{-1} and the inverse of the square root of conductivity gradient [25], making electrohydrodynamically stable flow with higher electric fields and larger channels very difficult.
- One last and fair question is "Are finite Reynolds number EOF flows (e.g., order 10 and larger) interesting to the design of real devices?" I know of no such application, but will make no predictions here.

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