

Developing active microfluidic pump and mixer driven by AC field-effect-mediated induced-charge electroosmosis of metal-dielectric Janus micropillars: physical perspective and simulation analysis – Supplementary Information

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Contents included in the Supplementary Information (SI):

Section A. Numerical simulation methodology

Section B. Model validation against standard benchmarks

Section A. Numerical simulation methodology

A commercial software package, Comsol Multiphysics (version 6.0 update 1), is used for analyzing the biased ICEO fluid motion under AC field-effect flow control and its application in simultaneous electro-convective transport and mixing in microchannels. The simulation procedure is as follows:

(1) Firstly, we compute the three Laplace equations (Eq.1) to acquire the AC electric potential within the liquid solution, as well as the solid metal-dielectric Janus pillar. RC charging condition Eq.(9) is imposed to the DE/electrolyte interface on channel sidewalls, where the right conducting DE plate is grounded with $V_{DE}=0$ and the left counterpart is supplied by a AC voltage input of $V_{DE}=V_D$ in amplitude, to mathematically describe electrochemical ion relaxation on DE surfaces. Conjugating conditions (Eq.(3)(4), Eq.(5)(6), and Eq.(7)(8)) are used to treat double-layer dispersion as a thin capacitor skin on dielectric surface, metal surface and their contact interface of the Janus cylinder. The normal current flux vanishes at both the channel inlet and outlet.

(2) Secondly, the time-independent Stokes equation (Eq.(11) (12)) is solved to acquire the AC field-effect-reconfigurable ICEO flow field around Janus post of uneven interfacial polarizability. The inlet and outlet of the main-channel are regarded as open boundaries for fluid motion with a zero gauge pressure $p=0$, namely, there is no externally applied pressure difference to the fluidic device. Any net axial pump behavior, and lateral rotating perturbation, can only originate from ICEO streaming, whose effect is incorporated by inserting the nonlinear electrokinetic slip expression (Eq.(13)(14)(15)) as a leaking-wall condition at the DE/electrolyte, dielectric/electrolyte, metal/electrolyte interfaces, respectively.

(3) Thirdly, we compute the mass delivery equation Eq.(16) in the liquid bulk, any total flux is inhibited in the normal direction of all the solid/electrolyte interfaces. Fluorescein nanospheres of 40 nm in diameter with thermal diffusivity $D=10^{-11} \text{ m}^2\cdot\text{s}^{-1}$ are chosen as the test analyte, the concentration of which equals $C=1 \text{ nM}$ at the left entrance (T_{AB}) and $C=0 \text{ nM}$ at the right one (T_{AC} in Fig.1(F)), agreeing with the actual situation of fully-automated ICEO mixing under active electrokinetic transport. Besides, the normal diffusive motion reaches zero at the device outlet.

Stationary solvers are implemented for all the control equations subjected to realistic boundary conditions. The AC voltage phasor, fluid mechanics driven by ICEO and sample transfer are solved separately in sequence with the PARDISO algorithm, and grid-independence is carefully examined before doing any formal simulation.

Section B. Model validation against standard benchmarks

Before studying the asymmetric ICEO fluid motion and analyte mass transfer behavior near metal-dielectric Janus micropillars under AC field-effect flow control, it is necessary to validate the accuracy and effectiveness of our proposed simulation model of ICEO applicable for solid surfaces of any electrical polarizability, against recognized benchmarks. The symmetric ICEO vortex flow field next to an ideally polarizable metal cylinder submerged in dilute electrolyte serves as a paradigmatic computation benchmark for nonlinear electroosmotic flow actuation at the interface between two lossy dielectric media of different phases, since it is well known that once the inner cylindrical solid phase consisting of uniform metal material is much more conducting than its surrounding liquid phase, a bipolar IDL forms near their contact interface under external AC voltage excitation, and a time-averaged ICEO flow field of four counter-rotating whirlpools appears with respect to the central metal cylinder due to the action of the imposed electric field on its own induced diffuse charges. Due to such a second-order voltage dependence, as can be also reflected quantitatively by Eq.(25)-(28), ICEO survives well under AC force, despite that its velocity magnitude may decrease with signal frequency by electrochemical ion relaxation.

The time-averaged ICEO flow field depends sensitively on the physical properties of both the solid object and liquid medium, the cylinder size, as well as the parameters of applied AC voltage signal. For the sake of model validation, we focus on the effect of latter on the resulted ICEO fluid flows. Under the approximation of thin double layer and small zeta potential,

namely, the Debye-Hückel limit, the standard model has been already proven to agree well with experimental observation at the phenomenal level. In the present investigation, we verify the improved physical model of ICEO in dilute electrolyte limit proposed in Section 2.2 against the analytical treatment from Squires and Bazant[1].

The linear RC circuit theory of ICEO predicts the tangential velocity of aqueous electrolyte on the surface of a metal cylinder is given:

$$u_{\theta} = \frac{1}{2} \frac{1}{1+\delta} \frac{1}{\left(1 + \left(2\pi f t_{RC}^m\right)^2\right)} \frac{\epsilon_r E^2 R}{\eta} (-\sin 2\theta) \quad \text{at } r=R \text{ for } 0^\circ \leq \theta \leq 360^\circ \quad (S1)$$

Where θ denotes the polar angle, and r the polar radius, and u_{θ} the ICEO flow component in the azimuth direction, as defined explicitly in Fig.1(B). The AC voltage is applied transversally across the pair of conducting DE plates, and the induced Debye screening effect is omitted on their surfaces, to meet the application scenario of the analytical solution where the background electric field is uniform and constant.

Because the phasor amplitude of AC voltage signal is used in our simulation, a prefactor 1/2 appears in Eq.(S1), to generate the time-averaged ICEO flow speed as expected. The direction of u_{θ} is defined in a way that it has a positive value with an in-situ counterclockwise liquid rotation while being negative when the local fluid streams clockwise. As displayed in Fig.2(A), a qualitative picture of the canonical quadrupolar ICEO vortex flow field is well reproduced by our improved numerical model, as induced near a single floating metal cylinder of radius $R=5 \mu\text{m}$ by a transversal AC voltage $V_D=2\text{V}$ at $f=200 \text{ Hz}$. Driven by ICEO, the liquid medium is sucked towards the two poles along the field axis, and ejected from the two equatorial points perpendicular to the field axis, resulting in the formation of four close-looped flow circulations in steady state after a short inertial relaxation (Fig.2(A)).

The numerical results of u_{θ} agree quantitatively with its analytical solution from Squires and Bazant[1] (Fig.2(B)). The regularity of sinusoidal variation of tangential ICEO slipping along the surface of micropillar from theory is accurately captured by current numerical modeling at four different signal frequencies. As shown in Fig.2(B), the liquid motion is clockwise in the 1st and 3rd quadrant with negative velocities, and counterclockwise in the 2nd and 4th quadrant with positive velocities, irrespective of field frequency. As a result, four ICEO velocity peaks appear at the polar angle of 45° , 135° , 225° , and 315° , respectively (Fig.2(B)), as also witnessed by the 2D flow profile in Fig.2(A).

In Fig.2(C), the space-averaged ICEO slipping velocity on cylinder surface is evaluated within a broad range of field frequencies spanning from $f=0 \text{ Hz}$ to $f=10^7 \text{ Hz}$. Both the numerical simulation and analytical prediction can resolve the effect of electrochemical ion relaxation on ICEO fluid motion. The strength of ICEO slipping decays monotonously with signal frequency, and drops dramatically once the field frequency exceeds the reciprocal charge relaxation time $f_{RC}^m = (1+\delta)\lambda_D \sigma_f / 2\pi\epsilon_f R \approx 1000 \text{ Hz}$ for double-layer polarization at the metal/electrolyte interface (Fig.2(C)). In Fig.2(D), the ICEO flow velocity from simulation grows quadratically with the AC voltage amplitude, which accords well with the analytical treatment, consolidating further the effectiveness of our improved ICEO model.

Although they fit so well, nevertheless, there is still a minor difference (no more than 3%) between numerical and analytical results, as clearly observable from a direct comparison of the solid and dashed data curves in Fig.2(B)-(D). Such a discrepancy in flow velocity arises due to a geometric confinement effect, since the square calculation domain has a finite edge length $L_c=W_c=100\mu\text{m}$, which is only ten times the diameter of the central metal post. In fact, the no slipping condition imposed on the upper and lower DE boundaries has to suppress the numerical solution of ICEO speed by an enhanced flow resistance to some degree, as long as the size of calculation domain is not infinitely large compared to the radius of metal cylinder. Based on above analysis and calculation results in Fig.2, it is confirmed the improved simulation model proposed herein can exactly reconstruct the distinct ICEO flow behaviors around solid objects of arbitrary polarizability surrounded by ionic liquids.

References:

1. Squires, T.M.; Bazant, M.Z. Induced-charge electro-osmosis. *Journal of Fluid Mechanics* **2004**, *509*, 217-252.