Electronic Supplementary Material (ESI) for Lab on a Chip.

Pulsed Electric Field-Assisted Overlimiting Current Enhancement through a Perm-selective Membrane

Soonhyun Kwon[1], Hyomin Lee[2*], and Sung Jae Kim[1,3,4*]

SI Note 1. Characteristic scale and parameters

SI Table 1. Characteristic scales used in this work

Physical quantity	Characteristic scale	Description
Time	$\tilde{\tau}_D = \tilde{L}^2 / D$	Diffusion time scale
Frequency	$1/\tilde{\tau}_D$	Diffusion-scaled frequency
Length	$\tilde{L}$	y-directional length of numerical domain
Electric potential	$R\tilde{T}/F$	Thermal voltage scale
Concentration	c_0	Bulk concentration
Pressure	$\mu D/\tilde{L}^2$	Diffusion-scaled pressure
Flow velocity	$D/\tilde{L}$	Diffusion-scaled velocity
Current density	$FDc_0/\tilde{L}$	Diffusion-limited current density

SI Table 2. Numerical parameters and used values

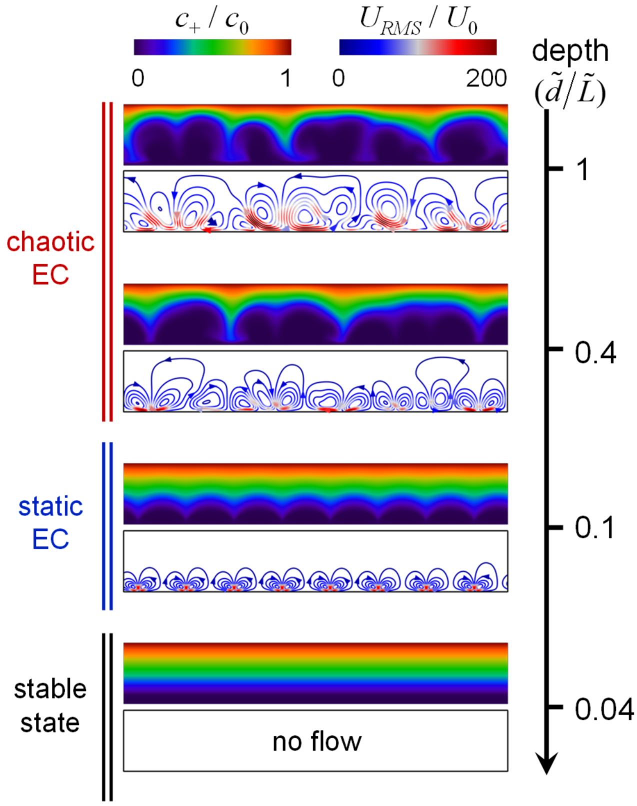
Parameter	Value	Description
Sc	500	Schmidt number
κ	0.5	Electrohydrodynamic coupling constant
N	2	Membrane charge density
λ_D	0.001	Debye length
ϕ_{mean}	80 – 140	Averaged electrical potential
f	5 – 50000	Frequency for applied potential

SI Note 2. Validation of the relation between electroconvection and channel depth

According to literatures about the relation between electroconvection and channel depth[1-4], they proposed that the geometric confinement would critically affect the development of electroconvection, onset voltage and vortex dynamics, *etc*. Additionally, the experimental evidences of such complex interactions between channel depth and electroconvection dynamics were directly visualized in a microchannel platform[4-6]. In order to investigate the confinement effect in this work, further numerical analysis was conducted by utilizing the Navier-Stokes equations with Hele-Shaw approximation rather than equation (4) in main text.

$$\frac{1}{Sc}\frac{D\mathbf{u}}{Dt} = -\nabla p + \nabla^2\mathbf{u} - \frac{\kappa}{2\lambda_D^2}(c^+ - c^-)\nabla\phi - \frac{12}{d^2}\mathbf{u}$$

The last term in RHS is Hele-Shaw approximation representing additional drag by confined geometry with dimensionless depth d. Above modified equations can be found in other literatures as well[2,3]. As a result, SI Figure 1 was obtained.

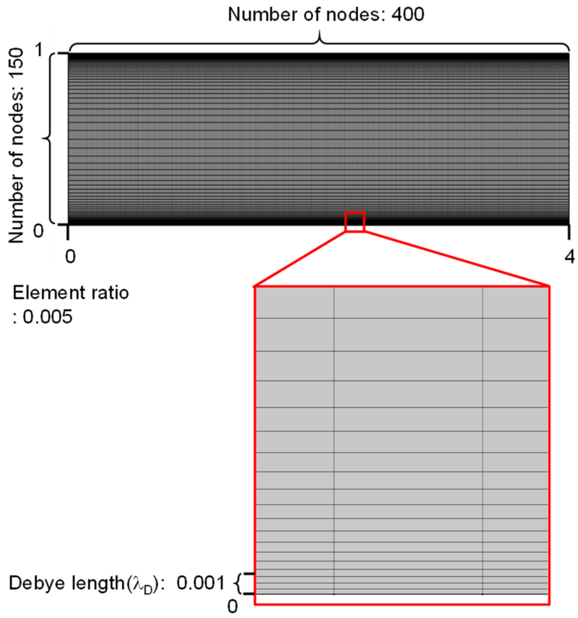


SI Figure 1. The concentration fluctuation and the flow patterns of electroconvective instability as a function of domain depth (perpendicular to the drawing plane) where $\tilde{d}$ denoted dimensional depth. The dimensionless applied voltage was 60.

Based on the numerical results, $\tilde{d}/\tilde{L}$ of our actual experimental platform was approximately 0.34 (= 170 μm / 500 μm) so that the qualitative characteristics of electroconvection was laid in "chaotic EC." This fact leads to the conclusion that the geometric confinement effect was negligible for qualitative analysis under our experimental conditions. However, the quantitative analysis would be limited because the magnitude of electroconvection was directly related with channel confinement. This was why there were small deviations between experimental and numerical results of Figure 2 in main text. Indeed, if full 3D numerical analysis is available which requires significant time and cost, quantitative comparison relating the confinement effect would be possible. Nevertheless, the numerical cost of 3D simulations based on finite element method is so expensive that the qualitative analysis based on 2D simulations was conducted in this work.

SI Note 3. Non-uniform mesh for the electrical double layer

In our work, distance to the bulk : λ_D is 200,000 : 1 in experiment and only 1,000 : 1 in numerical simulation. However, we have solved the fields only near the membrane up to $\tilde{L}$ of 10 μm, not 2 mm. Furthermore, as shown in SI Figure 2, we used non-uniform mesh along y-axis, denser near the membrane than the center. The elements have "element ratio" of 0.005 with symmetric distribution (*i.e.* denser mesh points near bulk and membrane). For example, there are 5 mesh points inside the electrical double layer (red box), while coarse meshes in the middle of bulk for reducing computational cost. Note that "element ratio" is a common terminology in COMSOL, referred the ratio of the first- to the last- element in the direction.



SI Figure 2. Mesh used in the simulation.

References

1. E. V. Dydek, B. Zaltzman, I. Rubinstein, D. S. Deng, A. Mani and M. Z. Bazant, *Phys. Rev. Lett.*, 2011, **107**, 118301.
2. M. B. Andersen, K. M. Wang, J. Schiffbauer and A. Mani, *Electrophoresis*, 2017, **38**, 702-711.
3. J. Schiffbauer, E. A. Demekhin and G. Ganchenko, *Phys. Rev. E*, 2012, **85**, 055302.
4. S. M. Rubinstein, G. Manukyan, A. Staicu, I. Rubinstein, B. Zaltzman, R. G. H. Lammertink, F. Mugele and M. Wessling, *Phys. Rev. Lett.*, 2008, **101**, 236101.
5. S. Nam, I. Cho, J. Heo, G. Lim, M. Z. Bazant, D. J. Moon, G. Y. Sung and S. J. Kim, *Phys. Rev. Lett.*, 2015, **114**, 114501.
6. K. Huh, S.-Y. Yang, J. S. Park, J. A. Lee, H. Lee and S. J. Kim, *Lab Chip*, 2020, **20**, 675-686.