Inducing AC-electroosmotic flow using electric field manipulation with insulators – Supplementary information

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S1: Derivation of equivalent circuit model;

In this part we derive the equivalent circuit model as presented in figure 1h of the main text:

Total capacitance of two capacitors in series:

$$\frac{1}{C_{total}} = \frac{1}{C_1} + \frac{1}{C_2} \#(1)$$

which can be reorganized as:

$$C_{total} = \frac{C_1 C_2}{C_1 + C_2} \#(2)$$

The specific capacitance of both two-dimensional double layer capacitors is defined as $C_{DL}[F/m]$, so the capacitance of C_1 and C_2 is the product of the C_{DL} and the length in y-direction. C_2 is therefore evaluated as:

$$C_2 = C_{DL} * L \#(3)$$

The length of C_1 is L plus the length of the corner part, which is $\frac{h}{\tan\alpha}$, making C_1:

$$C_1 = C_{DL} * \left(L + \frac{h}{\tan\alpha}\right) \#(4)$$

$$C_{cor}(L) = \frac{C_1 C_2}{C_1 + C_2} = \frac{{C_{DL}}^2 * L * \left(L + \frac{h}{\tan\alpha}\right)}{C_{DL} * \left(L + \frac{h}{\tan\alpha}\right) + C_{DL} * L} \#(5)$$

$$C_{cor}(L) = \frac{C_{DL} * L\left(L + \frac{h}{tan\alpha}\right)}{2L + \frac{h}{\tan\alpha}} \#(6)$$

With C_{cor} the total capacitance of the left branch.

The total electrical resistance, R, of a slab of material is given by Pouillet's law:

$$R = \frac{l}{\sigma A} \#(7)$$

with l[m] the length of the slab and $A[m]$ the cross-section of the slab and σ the conductivity [S]. A series of different slabs can therefore be considered as a series of resistors. Giving;

$$R_{total} = \sum_{i=1}^{n} R_i = \frac{l_i}{\sigma_i A_i} \#(8)$$

In our two dimensional model, the resistance in the left branch, R_{cor} can be described with;

$$R_{cor}(L) = \int_0^h \frac{1}{\sigma A(z)} dz \#(9)$$

With, A [m] (the distance between the left wall and L, at height and σ [S], the conductivity, being constant.

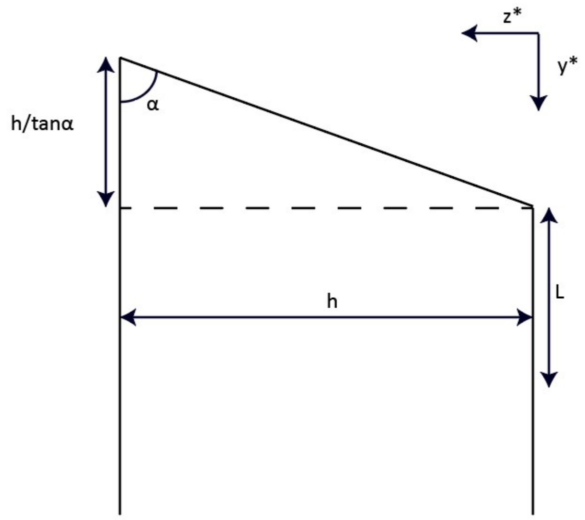

Figure S1 –schematic of the side of the chip, rotated 90 degrees.

For the coordinate system depicted in figure S1, the slope of the triangle is given by $1/\tan\alpha$, making $A(z^*)$;

$$A(z^*) = L + \frac{z^*}{\tan\alpha} \#(10)$$

When substituting equation 10 in equation 9, this gives;

$$R_{cor}(L) = \int_0^h \frac{1}{\sigma A(z)} dz = \left[\tan\alpha * \frac{\ln\left(\sigma\left(L * \tan\alpha + z^*\right)\right)}{\sigma}\right]_0^h \#(11)$$

$$R_{cor}(L) = \frac{\tan\alpha}{\sigma}(\ln\left(\sigma(L * \tan\alpha + h)\right) - \ln\left(\sigma(L * \tan\alpha)\right)) \#(12)$$

$$R_{cor}(L) = \frac{\tan\alpha}{\sigma}\ln\left(1 + \frac{h}{L * \tan\alpha}\right) \#(13)$$

To study the charging behavior of the equivalent circuit we study charging during an applied potential step, since this is the simplest way of charging a circuit. Also, using a step function shows how the system behaves at different times which can give insight in the charging behavior at different AC-frequencies. For a step-function, the voltage over the capacitive part of the left branch, $V_C(t)$, is:

$$V_C(t) = V_0\left(1 - e^{-\frac{t}{R_{cor}(L)C_{cor}(L)}}\right) \#(14)$$

with $t[s]$ the time and $V_0[V]$, the voltage step. The charge is equally distributed over the two capacitors, C_1 and C_2, because $q[C] = \int I dt + q_0$, and the current through both capacitors is equal since they are connected in series and we assume that the initial charge, q_0, at t=0 is zero. The charge density of the capacitors can be evaluated as $\sigma_{C1,L}(t) = C_{cor}(L) * V_c(t)/2L$ and $\sigma_{C2,L}(t) = C_{cor}(L) * V_c(t)/2(L + h/\tan(\alpha))$. Figure 1h (main text) shows the dimensionless charge density by dividing it by the charge in C_3 and C_4 for $t \to \infty$ of the two capacitors for different lengths at different dimensionless times. The time is made dimensionless by dividing it by the RC-time of the right branch of the ECM (figure 1e (main text)). The charge density is highest at the obtuse corner and the lowest at the acute corner, which would lead to lower AC-EOF velocities at the acute corner. From the figure 1e (main text) we get an indication of the direction of the AC-EOF at the sharp and obtuse corners; the voltage drop over the double layer is given by; $V_{d,i} = \frac{\sigma_i}{\epsilon\kappa}$, with $\epsilon[F/m]$, the permittivity of the electrolyte, $\sigma_i[C/m^2]$ the charge density and $\kappa[1/m]$, the reciprocal debye-length, and the tangential component of the electric field is given by $E_t = -\frac{dV_{d,i}}{dy}$. Since $\sigma_{C_1} < \sigma_{C_3}$ and $\sigma_{C_2} > \sigma_{C_4}$, the tangential component of the electric field will be in opposite direction for the two corners with the AC-EOF pointing towards the sharp corner and away from the obtuse corner.

S2: Simulation procedure

The simulation procedure of Green et al(1) has been followed.

Firstly, we use the linear approximation, $q_{dl} = C_{DL}(\phi - V_j)$, with $C_{DL}[F/m]$ the double layer capacitance, $\phi[V]$, the potential at the outer edge of the double layer and $V_j[V]$ the applied voltage.

This approximation is only valid for $V_0 < \frac{kT}{e}$, with $k[JK^{-1}]$ the Boltzmann constant, $T[K]$, the temperature and $e[C]$ the elementary charge. For higher voltages, the relation between the charge in the double layer and the voltage over the double layer is a sine hyperbolic relation. Effects like surface conductivity, an extended double layer and concentration polarization in the bulk are starting to play a role at $V_0 > 125mV$. Because it is our purpose to qualitatively show the principle of how an insulator can be used to shape AC-EOF, following the procedure used in Green's work valid for $V_0 < 25mV$ should be sufficient.

For applied voltages below 25 mV and frequencies far below the charge-relaxation frequency of the bulk electrolyte, the potential in the bulk electrolyte satisfies the Laplace equation:

$$\nabla^2\phi = 0 \tag{15}$$

with the boundary condition outside the double layer on the electrodes as:

$$\sigma\frac{\partial\phi}{\partial n} = \frac{\partial q_{dl}}{\partial t} \tag{16}$$

With, $\sigma[S]$ the conductivity of the electrolyte, $q_{dl}[C]$ the charge in the double layer and n the unitvector normal to the electrode surface, pointing towards the bulk electrolyte . After substituting q_{dl} in equation 16, and writing it in a complex notation, equation 16 becomes:

$$\sigma\frac{\partial\phi}{\partial n} = i\omega C_{DL}(\phi - V_j) \tag{17}$$

where the capacitance in the double layer is a combination of the Stern and diffuse double layer capacitance, C_s and C_d respectively, in series:

$$C_{DL} = \frac{C_s C_d}{C_s + C_d} \tag{18}$$

$$\text{Here } C_{DL} = \epsilon/\lambda_D \tag{19}$$

with the Debye length, $\lambda_D[m]$:

$$\lambda_D = \sqrt{D\epsilon/\sigma} \tag{20}$$

Here $\epsilon[F/m]$ is the permittivity of the solution and $D[m^2/s]$ is the mean diffusion coefficient of the ions in the electrolyte. Since the frequencies are relatively low, and the insulating polymer has a conductivity much lower than the electrolyte, the following relation was used on the insulating-electrolyte interface:

$$\frac{\partial\phi}{\partial n} = 0 \tag{21}$$

Fluid Dynamics

For the fluid dynamic problem, the Navier-Stokes equations were used, wherein because of low Reynolds numbers the inertial term can be neglected:

$$\eta\nabla^2\langle u\rangle - \nabla p = 0 \tag{22}$$

With, $\eta[Pa \cdot s]$, the viscosity, $\langle u\rangle[m/s]$, the time average velocity, and $p[Pa]$, the pressure.

$$\nabla \cdot \langle u\rangle = 0 \tag{23}$$

On the insulator there is a no-slip boundary condition:

$$\langle u\rangle = 0 \tag{24}$$

For the EOF at the electrode-electrolyte boundary (forming the boundary condition for the flow in the bulk) we use a formulation of the Helmholtz-Smoluchowski equation for the time-averaged surface-parallel flow velocity:

$$\langle u\rangle = \frac{\epsilon}{4\eta}\Lambda\frac{\partial}{\partial x}\left|\phi - V_j\right|^2 \tag{25}$$

with $\Lambda = \frac{C_{DL}}{C_d} = \frac{C_s}{C_s + C_d}$. Λ, was set at 0.5 to match the experimental data.

Figure S2 and S3 display the electric field direction at low and high frequencies.

S3 Numerical simulation results:

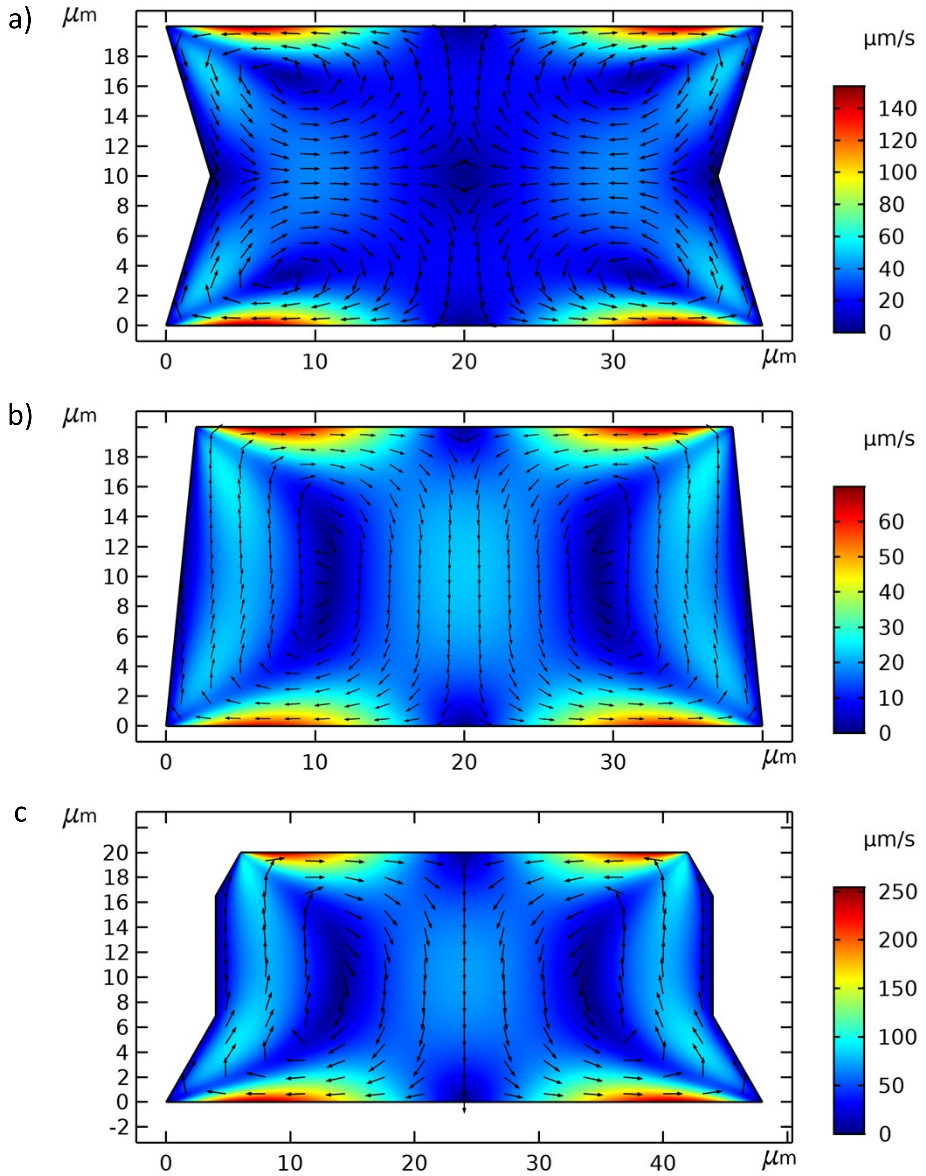

Figure S2- Numerical simulation of different cross-sectional shapes. a) Hourglass shape. b) Trapezium, c) Rectangular with non-orthogonal corners.

S4 Taylor scraping flow:

The flow velocity, in polar coordinates, inside a corner between a plane with a slip velocity and another plane with zero velocity, inclined under an angle can be calculated using:

$$u_r = \frac{U}{\alpha^2 - \sin^2\alpha}\{\theta \sin\alpha[\sin(\alpha - \theta) - \theta\cos(\alpha - \theta)] + \alpha[\sin[\theta - (\alpha - \theta)\cos(\theta)]\} \qquad (26)$$

$$u_\theta = \frac{U}{\alpha^2 - \sin^2\alpha}[\theta \sin\alpha \sin(\alpha - \theta) - \alpha(\alpha - \theta)] \qquad (27)$$

with r and θ, the radial coordinate and the angular coordinate respectively, U, the slip velocity of the lower plate, α, the angle between the two plates. While in the original problem of Taylor's paint scraper, the slip flow was induced by a moving plate, the slip flow in our problem is induced by the electroosmotic flow. Therefore we substitute $U = \langle u_{eof} \rangle$.

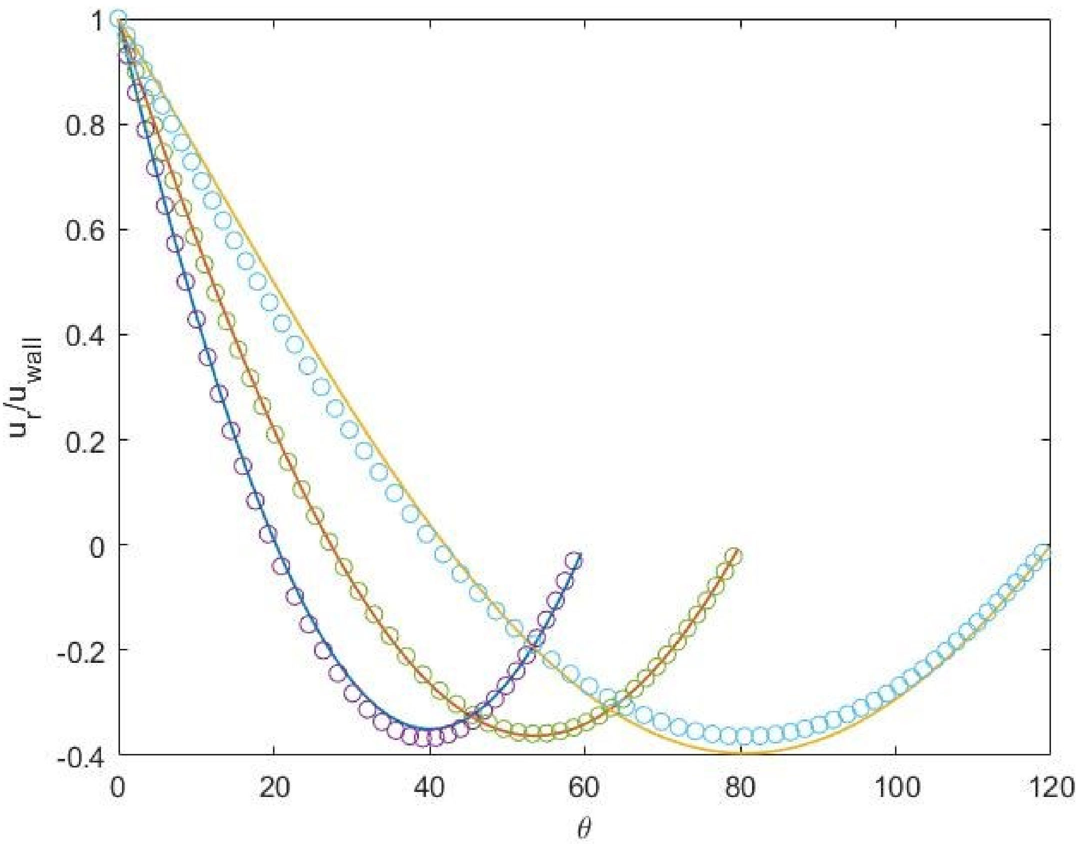

Figure S3- numerical simulation of the radial fluid flow (dots), and the analytic solution of the paint-scraper problem for angles of 60, 80 and 120 degrees

Figure S2 shows the simulated radial fluid velocity at r=5um from the wall relative to the slip velocity at the wall. A positively signed flow indicates a flow in the direction of the electro-osmotic flow, while a negative sign indicates flow in the opposite direction. The analytic paint-scraper solution is in good agreement with the numerical simulations. The flow velocity is only a function of θ and not of r as can be observed in the corners in the results of the numerical simulations (figure 2e (main text)). The pressure driven flows away and towards the corner will connect to each other and form a vortex when in the same direction. Figure S2 shows that the flow changes direction at $\theta = \frac{1}{3}\alpha$ and that the maximum magnitude of the pressure driven flow velocity is around 0.4 times the magnitude of the slip flow. There is only a slight difference in the minimum radial flow velocity, relative to the AC-EOF at the wall, for the different angles. The maximum pressure driven flow velocity, relative to the wall velocity, does not change much with changing α. However, the volume flow rate through a corner, $Q = \int_0^\alpha \frac{|u_r|}{2} d\theta$, does increase with increasing angle α. A high volume flow rate can be interesting for applications such as inducing lateral mixing, making the choice of angle important.

S5 Cross-sectional shape bell-shaped channel (SEM)

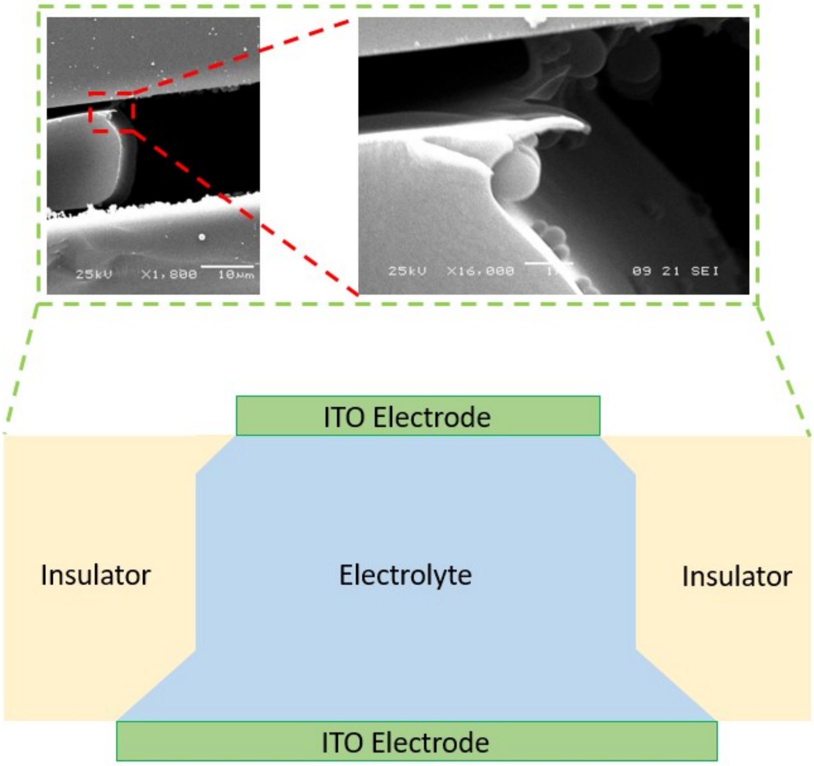

Figure S4: SEM-images of bell-shaped channel (after dicing)

1. Green NG, Ramos A, González A, Morgan H, Castellanos A. Fluid flow induced by nonuniform ac electric fields in electrolytes on microelectrodes. III. Observation of streamlines and numerical simulation. Phys Rev E - Stat Physics, Plasmas, Fluids, Relat Interdiscip Top. 2002;66(2):1–11.