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

Supplementary Information

Resonant dielectrophoresis and electrohydrodynamics for high-sensitive impedance detection of whole-cell bacteria

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1 Generalities

To derive the expression of electrokinetic effects for passivated electrodes, we use similar development as those indicated in [1, 2]. The general situation is represented in Fig. 1 and is assumed two-dimensional. Cylindrical coordinates are used. All parameters are defined in Section 2.5 of the manuscript and the friction factor is defined as $\gamma = 6\pi\eta a$, with η the dynamic viscosity of the electrolyte and a the bacteria radius. Resonance effects that are possibly due to the setup (cables, probes, etc.) are not considered in the following models.

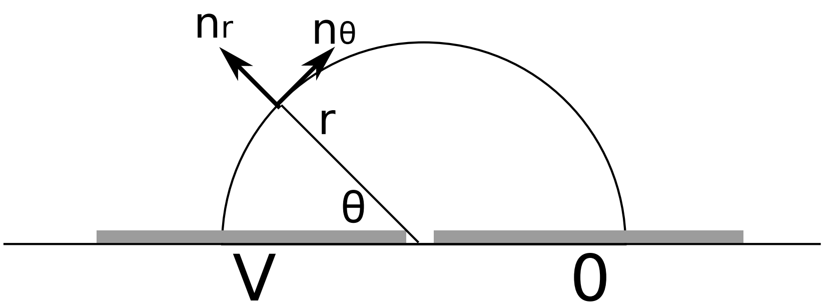

Figure 1 – Schematic view of the situation

2 Expression of the electrical field

Let $\vec{E}_{ins}$ and $\vec{E}_{sol}$ be the electric field contained in the insulating layers of thickness t_{ins} and in the electrolyte, respectively. The current conservation at the insulator-electrolyte interface gives the following relationship: $\varepsilon_{ins}\vec{E}_{ins} = (\varepsilon_{sol} + \frac{\sigma_{sol}}{j\omega})\vec{E}_{sol}$. The applied AC electric potential V is thus expressed as:

$$\begin{aligned} V &= -\int_0^{t_{ins}} E_{ins}dx - \int_{t_{ins}}^{\pi r + t_{ins}} E_{sol}dx - \int_{\pi r + t_{ins}}^{\pi r + 2t_{ins}} E_{ins}dx \\ &= 2t_{ins}E_{ins} + \pi r E_{sol} \\ &= \left[2\frac{t_{ins}}{\varepsilon_{ins}}\left(\varepsilon_{sol} + \frac{\sigma_{sol}}{j\omega}\right) + \pi r\right] \cdot E_{sol} \end{aligned}$$

Consequently, the electric field $\vec{E}_{sol}$ can be expressed as:

$$\vec{E}_{sol} = \frac{V}{\pi r} \cdot \underbrace{\left(\frac{1}{1 + 2 \cdot (\frac{\varepsilon_{sol}}{\varepsilon_{ins}} + \frac{\sigma_{sol}}{j\omega\varepsilon_{ins}}) \cdot \frac{t_{ins}}{\pi r}} \right)}_{G(\omega,r)} \cdot \vec{a}_\theta$$

It can be noticed that the electric field depends on the frequency when $t_{ins} \neq 0$, unlike gold electrodes ($t_{ins} = 0$) immersed in solution where the electric field $\vec{E}_{sol} = \frac{V}{\pi r} \cdot \vec{a}_\theta$ is constant [1, 2]. The modulus of $G(\omega, r)$ and $\vec{E}_{sol}$ are equal to:

$$\begin{aligned} \|G(\omega, r)\|^2 &= \frac{1}{(1 + 2\frac{\varepsilon_{sol} \cdot t_{ins}}{\varepsilon_{ins} \cdot \pi r})^2 + (\frac{2\sigma_{sol} \cdot t_{ins}}{\omega\varepsilon_{ins} \cdot \pi r})^2} \\ \|E_{sol}\|^2 &= \frac{V^2}{\pi^2} \cdot \frac{1}{(r + \frac{2\varepsilon_{sol}t_{ins}}{\varepsilon_{ins}\pi})^2 + (\frac{2\sigma_{sol}t_{ins}}{\omega\varepsilon_{ins}\pi})^2} \end{aligned}$$

3 Expression of the dielectrophoresis

The dielectrophoresis force can generally be expressed as [1, 2]:

$$F_{DEP}(t) = (\vec{m}(t) \cdot \vec{\nabla})\vec{E}_{sol}(t)$$

The time-average dielectrophoresis force $\langle \vec{F}_{DEP} \rangle$ is thus equal to:

$$\begin{aligned}
\langle \vec{F}_{DEP} \rangle &= \frac{1}{2} \cdot \mathbb{R}[(\vec{m}(\omega) \cdot \vec{\nabla})\vec{E}^*_{sol}] \\
&= \frac{1}{2} \cdot \mathbb{R}[(4\pi\varepsilon_{sol} \cdot a^3 f_{CM}(\omega)\vec{E}_{sol} \cdot \vec{\nabla})\vec{E}_{sol}(\omega)^*] \\
&= 2\pi\varepsilon_{sol} \cdot a^3 \cdot \mathbb{R}\left[f_{CM}(\omega) \frac{\vec{\nabla}\{\vec{E}_{sol} \cdot \vec{E}^*_{sol}\}}{2} \right] \\
&= \pi\varepsilon_{sol} \cdot a^3 \cdot \mathbb{R}\left[f_{CM}(\omega)\vec{\nabla}|\vec{E}_{sol}|^2 \right]
\end{aligned}$$

where f_{CM} is the Clausius-Mossoti factor. As $|\vec{E}_{sol}|^2$ is always a real number (even if $\vec{E}_{sol}$ is complex), we get:

$$\begin{aligned}
\langle \vec{F}_{DEP} \rangle &= \pi\varepsilon_{sol} a^3 \cdot \mathbb{R}\{f_{CM}(\omega)\} \cdot \vec{\nabla}|\vec{E}_{sol}|^2 \\
&= \pi\varepsilon_{sol} a^3 \cdot \mathbb{R}\{f_{CM}(\omega)\} \cdot \frac{\partial|\vec{E}_{sol}|^2}{\partial r} \cdot \vec{a}_r \\
&= -\frac{2}{\pi} \cdot a^3 V^2 \varepsilon_{sol} \cdot \mathbb{R}\{f_{CM}(\omega)\} \cdot \left(\frac{r + 2\frac{\varepsilon_{sol} \cdot t_{ins}}{\varepsilon_{ins} \cdot \pi}}{r^4} \right) \cdot \|G(\omega, r)\|^4 \cdot \vec{a}_r
\end{aligned}$$

The resulting bacteria speed is computed as follows:

$$\begin{aligned}
\langle \vec{v}_{DEP} \rangle &= \frac{\langle \vec{F}_{DEP} \rangle}{\gamma} \\
&= -\frac{1}{3\pi^2\eta} \cdot a^2 V^2 \varepsilon_{sol} \cdot \mathbb{R}\{f_{CM}(\omega)\} \cdot \left(\frac{r + 2\frac{\varepsilon_{sol} \cdot t_{ins}}{\varepsilon_{ins} \cdot \pi}}{r^4} \right) \cdot \|G(\omega, r)\|^4 \cdot \vec{a}_r \\
&= -\frac{a^2 V^2 \varepsilon_{sol} \cdot \mathbb{R}\{f_{CM}(\omega)\} \cdot \left(r + 2\frac{\varepsilon_{sol} \cdot t_{ins}}{\varepsilon_{ins} \cdot \pi}\right)}{3\pi^2\eta \cdot \left[(r + 2\frac{\varepsilon_{sol} \cdot t_{ins}}{\varepsilon_{ins}\pi})^2 + (\frac{2\sigma_{sol} t_{ins}}{\omega\varepsilon_{ins}\pi})^2 \right]^2} \cdot \vec{a}_r \\
&= -\frac{a^2 V^2 \varepsilon_{sol} \cdot \mathbb{R}\{f_{CM}(\omega)\}}{3\pi^2\eta \cdot \left(r + 2\frac{\varepsilon_{sol} \cdot t_{ins}}{\varepsilon_{ins} \cdot \pi}\right)^3 \left[1 + (\frac{2\sigma_{sol} t_{ins}}{\omega(r\varepsilon_{ins}\pi + 2\varepsilon_{sol} t_{ins})})^2 \right]^2} \cdot \vec{a}_r
\end{aligned}$$

As $\mathbb{R}\{f_{CM}(\omega)\}$ is positive, the speed is directed towards the sensor centre and positive-dielectrophoresis occurs then.

4 Expression of the AC-electroosmosis

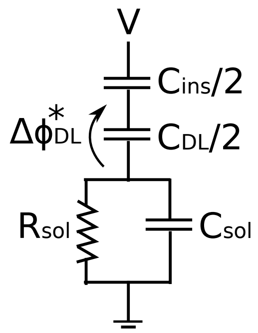

Figure 2 – AC representation of the system complex impedance.

Based on Fig. 2 and the expression of the series capacitance $C_s = [C_{ins}^{-1} + C_{DL}^{-1}]^{-1}$, the voltage drop across one double layer is:

$$
\begin{aligned}
\Delta\phi_{DL} &= \frac{\Delta\phi_{DL}^*}{2} \\
&= \frac{1}{2} \cdot \frac{I_{tot}}{j\omega C_{DL}/2} \\
&= \frac{1}{2} \cdot \frac{\left[(G_{sol} + j\omega C_{sol})^{-1} + (j\omega C_s/2)^{-1}\right]^{-1}}{j\omega C_{DL}/2} \cdot V \\
&= \frac{1}{2} \cdot \frac{1}{C_{DL}/C_s} \cdot \frac{V}{1 + \frac{j\omega C_s/2}{G_{sol}+j\omega C_{sol}}} \\
&= \frac{1}{2} \cdot \frac{1}{C_{DL}/C_s} \cdot \frac{V}{1 + \frac{j\omega C_s/2\cdot(G_{sol}-j\omega C_{sol})}{G_{sol}^2+\omega^2 C_{sol}^2}} \\
&= \frac{1}{2} \cdot \frac{1}{C_{DL}/C_s} \cdot \frac{V}{(1+\omega^2\tau_1\tau_2) + j\omega\tau_1}
\end{aligned}
$$

avec $\tau_1 \triangleq \frac{G_{sol}C_s/2}{G_{sol}^2+\omega^2 C_{sol}^2} = \frac{\pi r\cdot\sigma_{sol}C_s/2}{\sigma_{sol}^2+\omega^2\varepsilon_{sol}^2}$ and $\tau_2 \triangleq \frac{C_{sol}}{G_{sol}} = \frac{\varepsilon_{sol}}{\sigma_{sol}}$, since $C_{DL} = \varepsilon_{sol}/\lambda_D$, $C_{ins} = \varepsilon_{ins}/t_{ins}$, $C_{sol} = \varepsilon_{sol}/(\pi r)$, $G_{sol} = \sigma_{sol}/(\pi r)$, $C_s = \frac{\varepsilon_{sol}\varepsilon_{ins}}{t_{ins}\varepsilon_{sol}+\lambda_D t_{ins}}$. Thus,

$$
|\Delta\phi_{DL}|^2 = \frac{1}{4(C_{DL}/C_s)^2} \cdot \frac{V^2}{\omega^2\tau_1^2 + (1+\omega^2\tau_1\tau_2)^2}
$$

Therefore:

$$
\begin{aligned}
\frac{\partial|\Delta\phi_{DL}|^2}{\partial r} &= \frac{\partial|\Delta\phi_{DL}|^2}{\partial\tau_1} \cdot \frac{\partial\tau_1}{\partial r} \\
&= -\frac{V^2\omega^2}{2(C_{DL}/C_s)^2 r} \cdot \frac{(\tau_1+\tau_2)\tau_1 + \omega^2\tau_1^2\tau_2^2}{(\omega^2\tau_1^2 + (1+\omega^2\tau_1\tau_2)^2)^2}
\end{aligned}
$$

Finally, the slip velocity inside the electrical double layer is approximated by [3]:

$$
\begin{aligned}
\vec{v}_{slip} &\triangleq \frac{\varepsilon_{sol}}{2\eta} \cdot \Lambda \cdot \mathbb{R}\{\Delta\phi_{DL} \cdot \vec{E}_t^*\} \\
&= -\frac{\varepsilon_{sol}}{2\eta} \cdot \Lambda \cdot \mathbb{R}\{\Delta\phi_{DL} \cdot \frac{\partial\Delta\phi_{DL}^*}{\partial r}\} \cdot \vec{a}_r
\end{aligned}
$$

where $\Lambda = \frac{C_{stern}}{C_{stern}+C_{DL}} \simeq 0.25$ is an empirical factor with C_{stern} the Stern capacitance [2]. Because $c \cdot \partial c^*/\partial r = c^* \cdot \partial c/\partial r = 0.5 \cdot \partial(c \cdot c^*)/\partial r = 0.5 \cdot \partial|c|^2/\partial r$ for all complex c, the following formula is finally obtained:

$$\begin{aligned}
\vec{v}_{slip} &= -\frac{\varepsilon_{sol}}{4\eta} \cdot \Lambda \cdot \mathbb{R}\{\frac{\partial|\Delta\phi_{DL}|^2}{\partial r}\} \cdot \vec{a}_r \\
&= -\frac{\varepsilon_{sol}}{4\eta} \cdot \Lambda \cdot \frac{\partial|\Delta\phi_{DL}|^2}{\partial r} \cdot \vec{a}_r \\
&= \frac{V^2\omega^2\varepsilon_{sol}\Lambda}{8\eta(C_{DL}/C_s)^2 r} \cdot \frac{(\tau_1+\tau_2)\tau_1 + \omega^2\tau_1^2\tau_2^2}{\left(\omega^2\tau_1^2 + (1+\omega^2\tau_1\tau_2)^2\right)^2} \cdot \vec{a}_r
\end{aligned}$$

It is important to remember that $\vec{v}_{slip}$ is only a slip velocity which exists only at the electrode surface, so that $\vec{a}_r$ is parallel to $\vec{a}_x$.

5 Expression of the electrothermal flow

To estimate the electrothermal flow, it is required to evaluate the increase of the local temperature induced by the electric field $\vec{E}_{sol}$. The Poisson equation gives: $-\sigma_{sol}E_{sol}(t)^2 = -k\vec{\nabla}^2 T(t)$. Since $E_{sol}(t) = E_{sol}\cos(\omega t)$, we have: $E_{sol}(t)^2 = \frac{E_{sol}^2}{2}\cdot(1+\cos(2\omega t)) = E_{sol,RMS}^2 \cdot (1+\cos(2\omega t))$. The following relationship is obtained by neglecting AC terms:

$$\begin{aligned}
-\sigma_{sol}E_{sol,RMS}^2 &= k\vec{\nabla}^2 T \\
&= \frac{k}{r}\frac{\partial}{\partial r}(r\frac{\partial T}{\partial r}) + \frac{k}{r^2}\frac{\partial^2 T}{\partial \theta^2}
\end{aligned}$$

with $E_{sol,RMS} = \frac{V_{RMS}}{\pi r} \cdot G(\omega, r)$. A particular solution $T(r,\theta)$ to this equation was not found, mainly because of the variation of $G(\omega, r)$ with r. By assuming $G(\omega, r) = G(\omega)$, it is possible to find a particular solution: $T(\theta) = -\frac{\sigma_{sol}}{k} \cdot \frac{V_{RMS}^2 \cdot G(\omega)^2}{2\pi}(\frac{\theta^2}{\pi} - \theta)$. The correctness of the approximation $G(\omega, r) = G(\omega)$ was numerically verified for $f \geq 130$ Hz at $\sigma_{sol} = 1.8$ mS/m, for $f \geq 1.3$ kHz at $\sigma_{sol} = 18$ mS/m, for $f \geq 13$ kHz at $\sigma_{sol} = 180$ mS/m and for $f \geq 130$ kHz at $\sigma_{sol} = 1.8$ mS/m. In other words, it is required that $f/\sigma_{sol} \geq 72$ (kHz.m)/S, otherwise the proposed particular solution cannot be used. When this condition is satisfied, $\frac{k}{r}\frac{\partial}{\partial r}(r\frac{\partial T}{\partial r}) \ll \frac{k}{r^2}\frac{\partial^2 T}{\partial \theta^2}$ and the approximation $G(\omega, r) = G(\omega)$ is correct.

With this assumption, $\vec{\nabla} T = \frac{1}{r} \cdot \frac{\partial T(\theta)}{\partial \theta}\vec{a}_\theta = -\frac{\sigma_{sol}}{kr} \cdot \frac{V_{RMS}^2 \cdot G(\omega,r)^2}{2\pi}(\frac{2\theta}{\pi} - 1)\vec{a}_\theta$. Starting from Coulomb equation like in [1], the force exerted on the fluid can be expressed as:

$$
\begin{aligned}
\langle \vec{F}_E \rangle &= \frac{1}{2}\mathbb{R}\left\{\left[\left(\frac{\sigma_{sol}\vec{\nabla}\varepsilon_{sol} - \varepsilon_{sol}\vec{\nabla}\sigma_{sol}}{\sigma_{sol} + j\omega\varepsilon_{sol}}\right)\vec{E}_0\right]\vec{E}_0^*\right\} - \frac{1}{4}\vec{E}_0 \cdot \vec{E}_0^*\vec{\nabla}\varepsilon_{sol} \\
&= \left[\frac{1}{2}\frac{\sigma_{sol}\cdot(\sigma_{sol}\vec{\nabla}\varepsilon_{sol} - \varepsilon_{sol}\vec{\nabla}\sigma_{sol})}{\sigma_{sol}^2 + \omega^2\varepsilon_{sol}^2} - \frac{1}{4}\vec{\nabla}\varepsilon_{sol}\right]\cdot E_0^2 \\
&= \frac{1}{2}\left[\frac{(\frac{1}{\varepsilon_{sol}}\vec{\nabla}\varepsilon_{sol} - \frac{1}{\sigma_{sol}}\vec{\nabla}\sigma_{sol})}{1 + (\omega\varepsilon_{sol}/\sigma_{sol})^2} - \frac{1}{2\varepsilon_{sol}}\vec{\nabla}\varepsilon_{sol}\right]\cdot \varepsilon_{sol}E_0^2 \\
&= -\underbrace{\left[\frac{-\frac{T}{\varepsilon_{sol}}\frac{\partial\varepsilon_{sol}}{\partial T} + \frac{T}{\sigma_{sol}}\frac{\partial\sigma_{sol}}{\partial T}}{1 + (\omega\varepsilon_{sol}/\sigma_{sol})^2} + \frac{T}{2\varepsilon_{sol}}\frac{\partial\varepsilon_{sol}}{\partial T}\right]}_{M(\omega,T)}\cdot\frac{\varepsilon_{sol}E_{RMS}^2}{T}\cdot\vec{\nabla}T \\
&= -M(\omega,T)\cdot\frac{\varepsilon_{sol}\sigma_{sol}V_{RMS}^4 G(\omega,r)^4}{2k\pi^3 r^3 T}\cdot\left(1 - \frac{2\theta}{\pi}\right)\cdot\vec{a}_\theta \\
&= -M(\omega,T)\cdot\frac{\varepsilon_{sol}\sigma_{sol}V^4 G(\omega,r)^4}{8k\pi^3 r^3 T}\cdot\left(1 - \frac{2\theta}{\pi}\right)\cdot\vec{a}_\theta
\end{aligned}
$$

To compute the fluid speed, we use the Stokes equation: $\vec{v}_E \approx 0.13 \cdot \langle \vec{F}_E \rangle r^2/\eta$, as Eq. 32 in [1]:

$$
\begin{aligned}
\vec{v}_E(\theta) &= -0.13\cdot M(\omega,T)\cdot\frac{\varepsilon_{sol}\sigma_{sol}V^4 G(\omega)^4}{8k\pi^3 r\eta T}\cdot\left(1 - \frac{2\theta}{\pi}\right)\cdot\vec{a}_\theta \\
&\approx -5\cdot 10^{-4}\cdot M(\omega,T)\cdot\frac{\varepsilon_{sol}\sigma_{sol}V^4 G(\omega)^4}{k\eta r T}\cdot\left(1 - \frac{2\theta}{\pi}\right)\cdot\vec{a}_\theta
\end{aligned}
$$

References

[1] A. Ramos, H. Morgan, N. G. Green, and A. Castellanos, "AC electrokinetics: a review of forces in microelectrode structures," *J. Phys. D: Appl. Phys.*, vol. 31, no. 18, p. 2338, 1998.

[2] A. Castellanos, A. Ramos, A. Gonzalez, N. G. Green, and H. Morgan, "Electrohydrodynamics and dielectrophoresis in microsystems: scaling laws," *J. Phys. D: Appl. Phys.*, vol. 36, no. 20, p. 2584, 2003.

[3] A. González, A. Ramos, N. Green, A. Castellanos, and H. Morgan, "Fluid flow induced by nonuniform ac electric fields in electrolytes on microelectrodes. II. A linear double-layer analysis," *Phys Rev E Stat Phys Plasmas Fluids Relat Interdiscip Topics*, vol. 61, pp. 4019–4028, Apr. 2000.