Supplementary information to the article:

Electroactuation with Single Charge Carrier Ionomers: The Roles of Electrostatic Pressure and Steric Strain

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A Derivation of the transcendental equation for curvature

Following from the free energy functional, the curvature κ can be written as Eq.13. Deriving our simple analytical result from Eq.13 is not straightforward and we outline that progression here for the interested reader.

$$\begin{aligned} \kappa \approx & -\frac{3}{2}\frac{\mathcal{S}}{\mathcal{S}_0}\frac{l_D}{Eh^2}\left(\int_0^{\mathrm{H}} \mathrm{d}X \left\{ kTc_0 \left[\frac{1}{2}\left(\frac{\mathrm{d}u}{\mathrm{d}X}\right)^2 - \frac{\mathrm{d}}{\mathrm{d}X}\left(u\frac{\mathrm{d}u}{\mathrm{d}X}\right)\right] - Kv\rho \right\}\right. \\ & \left. -\int_{-\mathrm{H}}^0 \mathrm{d}X \left\{ k_{\mathrm{B}}Tc_0 \left[\frac{1}{2}\left(\frac{\mathrm{d}u}{\mathrm{d}X}\right)^2 - \frac{\mathrm{d}}{\mathrm{d}X}\left(u\frac{\mathrm{d}u}{\mathrm{d}X}\right)\right] - Kv\rho \right\}\right). \qquad \text{(S1)} \end{aligned}$$

The integral (S 1) can be considerably simplified. In fact, the first term in the integral can be rewritten as

$$\begin{aligned} \int \mathrm{d}X \left(\frac{\mathrm{d}u}{\mathrm{d}X}\right)^2 &= \int \mathrm{d}X \left(\frac{\mathrm{d}y}{\mathrm{d}X} + p\kappa l_D\right)^2 \\ &= \int dy \frac{\mathrm{d}X}{dy}\left[\left(\frac{\mathrm{d}y}{\mathrm{d}X}\right)^2 + 2p\kappa l_D \frac{\mathrm{d}y}{\mathrm{d}X} + (p\kappa l_D)^2\right] \qquad \text{(S2)} \\ &= \int dy \frac{\mathrm{d}y}{\mathrm{d}X} + \int dX\,(p\kappa l_D)^2 + \int dy\, 2p\kappa l_D \qquad \text{(S3)} \end{aligned}$$

Combining the integral for positive X and negative X,

$$\begin{aligned}
\int_0^{\mathrm{H}} \mathrm{d}X \left(\frac{\mathrm{d}u}{\mathrm{d}X}\right)^2 - \int_{-\mathrm{H}}^0 \mathrm{d}X \left(\frac{\mathrm{d}u}{\mathrm{d}X}\right)^2 &= \int_0^{V_1 - p\kappa H} dy \frac{\mathrm{d}y}{\mathrm{d}X} - \int_{-V_2 + p\kappa H}^0 dy \frac{\mathrm{d}y}{\mathrm{d}X} \\
&\quad + 2p\kappa l_D (V_1 - V_2) \\
&= \int_0^{V_1 - p\kappa H} dy \sqrt{2\left[y + \frac{1}{\gamma}\ln\left(1 - \gamma + \gamma e^{-y}\right)\right]} \\
&\quad - \int_{-V_2 + p\kappa H}^0 dy \sqrt{2\left[y + \frac{1}{\gamma}\ln\left(1 - \gamma + \gamma e^{-y}\right)\right]} \\
&\quad + 2p\kappa l_D (V_1 - V_2) \qquad \text{(S4)}
\end{aligned}$$

The second terms in the integrals of Eq. 38, with derivatives, can be easily integrated

$$\begin{aligned}
\int_0^{\mathrm{H}} \mathrm{d}X \frac{\mathrm{d}}{\mathrm{d}X}\left(u\frac{\mathrm{d}u}{\mathrm{d}X}\right) - \int_{-\mathrm{H}}^0 \mathrm{d}X \frac{\mathrm{d}}{\mathrm{d}X}\left(u\frac{\mathrm{d}u}{\mathrm{d}X}\right) &= V_1 E(X = H) - V_2 E(X = -H) \\
&= (V_1 - V_2)\, E(X = H) \qquad \text{(S5)}
\end{aligned}$$

Where $E(X = H) \equiv du/dX$ is the electric field. The third term in the integral can be evaluated using Gauss law, equalising the surface charge densities of the electrodes with the induction as in ref [1]

$$\begin{aligned}
\int_0^{\mathrm{H}} \mathrm{d}X\, \rho - \int_{-\mathrm{H}}^0 \mathrm{d}X\, \rho &= c_0 \left\{ \sqrt{2\left[V_1 - p\kappa H + \frac{1}{\gamma}\ln\left(1 - \gamma + \gamma e^{-V_1 + p\kappa H}\right)\right]} \right. \\
&\quad \left. + \sqrt{2\left[-V_2 + p\kappa H + \frac{1}{\gamma}\ln\left(1 - \gamma + \gamma e^{V_2 - p\kappa H}\right)\right]} \right\}
\end{aligned} \qquad \text{(S6)}$$

B Large Voltage Asymptotics

To consider the integration of Eq. 15 $\int \mathrm{d}y\sqrt{2\left[y+\frac{1}{\gamma}\ln\left(1-\gamma+\gamma e^{-y}\right)\right]}$ we expand the integrand in the limit of large positive and negative voltages,

$$\sqrt{2\left[y+\frac{1}{\gamma}\ln\left(1-\gamma+\gamma e^{-y}\right)\right]} \approx \begin{cases} \sqrt{2\left[y+\frac{1}{\gamma}\ln\left(1-\gamma\right)\right]} & , y \gg 1 \\ \sqrt{2\left[\left(1-\frac{1}{\gamma}\right)y+\frac{1}{\gamma}\ln\gamma\right]} & , y \ll -1 \end{cases} \tag{S7}$$

However, as $\frac{1}{\gamma}\ln(1-\gamma)$ and $\frac{1}{\gamma}\ln\gamma$ are negative quantities, the radicals are undefined when $|y| \ll 1$ and the large voltage approximations in Eq (S 7) do not hold at low voltages. Hence, the integral must be approximated by combining the linear-response and non-linear approximants with $y = -\frac{1}{\gamma}\ln(1-\gamma)$ and $y = -\frac{1}{1-\gamma}\ln\gamma$ as cutoff for the case of positive y and negative y respectively.

For the integral extending to large positive values of y

$$\begin{aligned} &\int_0^{V_1-p\kappa h} dy\sqrt{2\left[y+\frac{1}{\gamma}\ln\left(1-\gamma+\gamma e^{-y}\right)\right]} \\ &\approx \sqrt{1-\gamma}\int_0^{-\frac{1}{\gamma}\ln(1-\gamma)} y \quad \mathrm{d}y + \int_{-\frac{1}{\gamma}\ln(1-\gamma)}^{V_1-p\kappa h} dy\sqrt{2\left[y+\frac{1}{\gamma}\ln\left(1-\gamma\right)\right]} \\ &= \frac{\sqrt{1-\gamma}}{2}\left(\frac{1}{\gamma}\ln\left(1-\gamma\right)\right)^2 + \frac{2\sqrt{2}}{3}\left(V_1-p\kappa h+\frac{1}{\gamma}\ln\left(1-\gamma\right)\right)^{3/2} \end{aligned} \tag{S8}$$

Similarly, for the integral extending to large negative y

$$\begin{aligned} &\int_{-V_2+p\kappa h}^{0} dy\sqrt{2\left[y+\frac{1}{\gamma}\ln\left(1-\gamma+\gamma e^{-y}\right)\right]} \\ &\approx \sqrt{1-\gamma}\int_{\frac{1}{\gamma-1}\ln\gamma}^{0} y \quad dy + \int_{-V_2+p\kappa h}^{\frac{1}{\gamma-1}\ln\gamma} dy\sqrt{2\left[\left(1-\frac{1}{\gamma}\right)y+\frac{1}{\gamma}\ln\gamma\right]} \\ &= -\frac{\sqrt{1-\gamma}}{2}\left(\frac{1}{\gamma-1}\ln\gamma\right)^2 - \frac{2\sqrt{2}}{3}\frac{1}{1-\frac{1}{\gamma}}\left(\left(1-\frac{1}{\gamma}\right)\left(-V_2+p\kappa h\right)+\frac{1}{\gamma}\ln\gamma\right)^{3/2} \end{aligned} \tag{S9}$$

All in all,

$$\begin{aligned}
&\int_0^{V_1-p\kappa h} dy\sqrt{2\left[y+\frac{1}{\gamma}\ln\left(1-\gamma+\gamma e^{-y}\right)\right]}-\int_{-V_2+p\kappa h}^{0} dy\sqrt{2\left[y+\frac{1}{\gamma}\ln\left(1-\gamma+\gamma e^{-y}\right)\right]}\\
&\approx \frac{2\sqrt{2}}{3}\left(V_1-p\kappa h+\frac{1}{\gamma}\ln\left(1-\gamma\right)\right)^{3/2}+\frac{2\sqrt{2}}{3}\frac{1}{1-\frac{1}{\gamma}}\left[\left(1-\frac{1}{\gamma}\right)\left(-V_2+p\kappa h\right)+\frac{1}{\gamma}\ln\gamma\right]^{3/2}\\
&\quad+\frac{\sqrt{1-\gamma}}{2}\left(\frac{1}{\gamma}\ln\left(1-\gamma\right)\right)^2+\frac{\sqrt{1-\gamma}}{2}\left(\frac{1}{\gamma-1}\ln\gamma\right)^2\\
&\sim\frac{\sqrt{8}}{3}\left[\left[\left(1-\gamma\right)V\right]^{3/2}+\frac{1}{1-\frac{1}{\gamma}}\left[-\left(1-\frac{1}{\gamma}\right)\gamma V\right]^{3/2}\right]\\
&=\frac{\sqrt{8}}{3}\left(1-2\gamma\right)\sqrt{1-\gamma}V^{3/2}
\end{aligned} \tag{S10}$$

Thus the integral $\int \mathrm{d}y\sqrt{2\left[y+\frac{1}{\gamma}\ln\left(1-\gamma+\gamma e^{-y}\right)\right]}$ scales as $\sim V^{3/2}$.

References

[1] A.A. Lee, R.H. Colby, and A.A. Kornyshev. *Electroactuation with Single Charge Carrier Ionomers*. arXiv:1212.2148, 2012.