Theory of Polymer-Electrolyte-Composite Electroactuator Sensors with Flat or Volume-Filling Electrodes
Supplementary Information

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1 Short-Circuit: Slit Pore

Here we outline the equations for reverse actuation with slit pores, as depicted in Fig. 2 of the main text. All assumptions from the main text are employed and it is further assumed that there is no variation in the z coordinate. Hence, the relevant dimensionless Helmholtz equation is as follows

$$\frac{\partial^2 \tilde{\psi}_s(\tilde{x},\tilde{y})}{\partial \tilde{y}^2} + \frac{\partial^2 \tilde{\psi}_s(\tilde{x},\tilde{y})}{\partial \tilde{x}^2} = (1-\gamma)\tilde{\psi}_s(\tilde{x},\tilde{y}). \tag{S1}$$

Here $\tilde{y} = \kappa_D y$ is the dimensionless coordinate that describes the distribution normal to the electrodes surface in the slit pore; and $\tilde{\psi}_s(\tilde{x},\tilde{y}) = u_s(\tilde{x},\tilde{y}) - \zeta\mathcal{K}\tilde{x}/\tilde{h}$.

Again, it is assumed that the partial differential equation is separable ($\tilde{\psi}_s(\tilde{x},\tilde{y}) = X_s(\tilde{x})Y_s(\tilde{y})$). Since edge effects are ignored, the solution in $\tilde{x}$ does not differ from the case of porous cylindrical electrodes. Hence, the solution shall not be stated agin for brevity (see main text). The differential equation in the $\tilde{y}$ coordinate is

$$\frac{d^2 Y_s(y)}{d\tilde{y}^2} = (1-\gamma)Y_s(\tilde{y}). \tag{S2}$$

The boundary conditions are as follows $u_s^{\tilde{y}}(\tilde{x},0) = 0$ (superscript $\tilde{y}$ denotes the derivative with respect to $\tilde{y}$) and $u_s(\tilde{x},\tilde{l}/2) = 0$,[1] where $\tilde{l} = l\kappa_D$ with l corresponding to the width of the split pore, as seen in Fig. 2 of the main text. Thus, the distribution of electrostatic potential, as shown in Fig. S1a) is given by

$$u_s(\tilde{x},\tilde{y}) = \frac{\zeta\mathcal{K}\tilde{x}}{\tilde{h}}\left[1 - \frac{\cosh\{\sqrt{1-\gamma}\tilde{y}\}}{\cosh\{\sqrt{1-\gamma}\tilde{l}/2\}}\right]. \tag{S3}$$

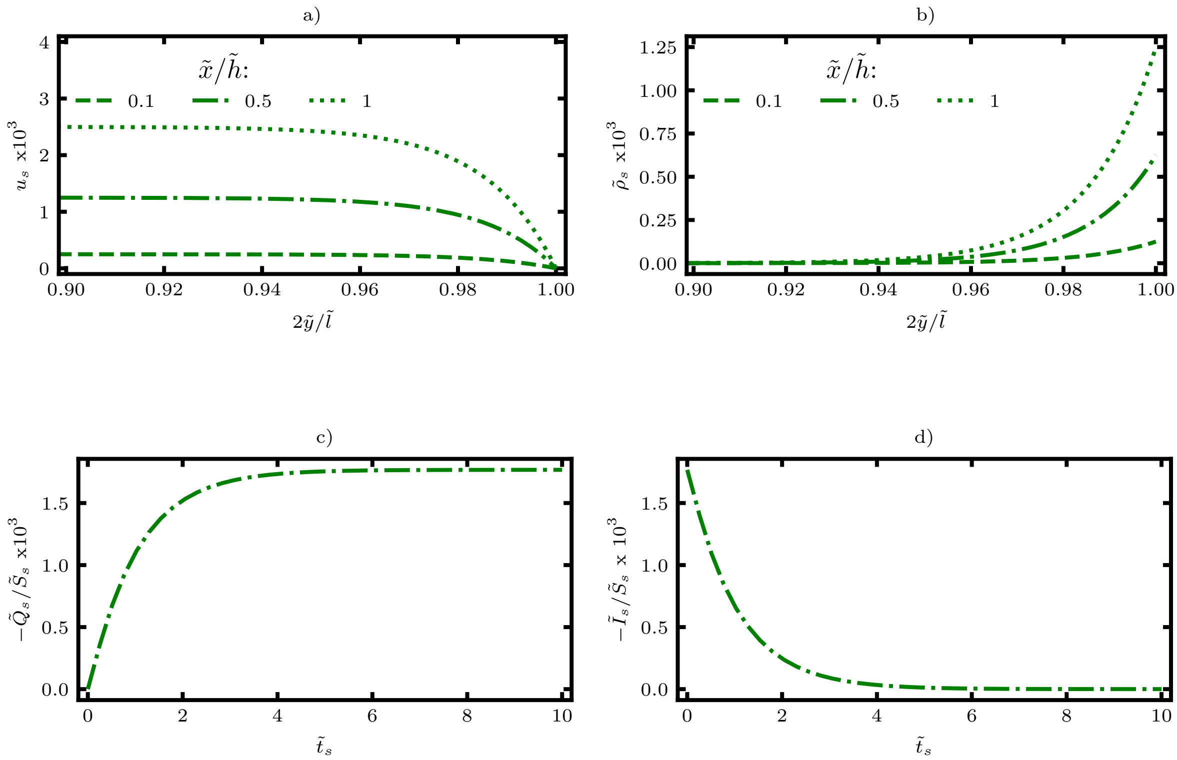

Figure S1: **Short-circuit characteristics for an electroactuator with micro-structured electrodes with slit pores.** The parameters used were $\mathcal{K} = 5$ x10^{-3}, $\tilde{l} = 200$, $\zeta = 0.5$ and $\gamma = 0.5$. a) - Dimensionless Electrostatic potential as a function of the normal coordinate to the electrode in the slit pore for the indicated pore depths. b) - Dimensionless excess charge concentration as a function of normal coordinate to the electrode in the pore for the indicated pore depths. c) - Dimensionless charge scaled to the surface area factor as a function of of time in the units of τ_c. d) - Dimensionless current scaled to the surface area factor as a function dimensionless time.

From which, at linear response, it is trivial to obtain the corresponding excess charge concentration

$$\tilde{\rho}_s(\tilde{x}, \tilde{y}) = (1-\gamma)\frac{\zeta\mathcal{K}\tilde{x}}{\tilde{h}}\frac{\cosh\{\sqrt{1-\gamma}\tilde{y}\}}{\cosh\{\sqrt{1-\gamma}\tilde{l}/2\}}. \tag{S4}$$

See Fig. S1b) for example curve.

Evaluating the dimensionless surface charge density yields

$$\tilde{\sigma}_s(\tilde{x}) = \sqrt{1-\gamma}\frac{\zeta\mathcal{K}\tilde{x}}{\tilde{h}}\tanh\{\sqrt{1-\gamma}\tilde{l}/2\}. \tag{S5}$$

Since the half-thickness is much larger than the Debye length ($\tilde{l}/2 \gg 1$), the dimensionless surface charge density can be simplified to

$$\tilde{\sigma}_s(\tilde{x}) \approx \sqrt{1-\gamma}\frac{\zeta\mathcal{K}\tilde{x}}{\tilde{h}}, \tag{S6}$$

which does not differ in form from micro-structured electrodes with cylindrical pores.

The total charge accumulated,

$$\tilde{Q}_s = -\sqrt{1-\gamma}\zeta\mathcal{K}\tilde{S}_s\Bigg\{1 - \frac{32}{\pi^3}\sum_{n=0}^{\infty}\frac{(-)^n}{\big(2n+1\big)^3}\exp\Big\{-\big(2n+1\big)^2\tilde{t}_s\Big\}\Bigg\}, \tag{S7}$$

once again, scales with the surface area of the electrodes, where $\tilde{S}_s = W_0hL_0/4_\pi l_B\lambda_D l_u$ and $\tau_s = 8h^2C/\pi^2 l\Sigma$. In Fig. S1c) the equation is plotted for dimensionless charge as a function of time, in units of τ_s.

The dimensionless current is given by

$$\tilde{I}_s = -\sqrt{1-\gamma}\zeta\mathcal{K}\tilde{S}_s\Bigg\{\frac{32}{\pi^3}\sum_{n=0}^{\infty}\frac{(-)^n}{2n+1}\exp\Big\{-\big(2n+1\big)^2\tilde{t}_s\Big\}\Bigg\}, \tag{S8}$$

Note that the dimensionless form of the dynamics of charging are identical to that of electrodes with cylindrical pores. In Fig. S1d) the current has been displayed again for completeness.

References

[1] K. Farinholt, D. J. Leo, Modeling of electromechanical charge sensing in ionic polymer transducers, Mechanics of Materials 36 (2004) 421–433.