Supplementary Information

Understanding the characteristics of conducting polymer-redox biopolymer supercapacitors

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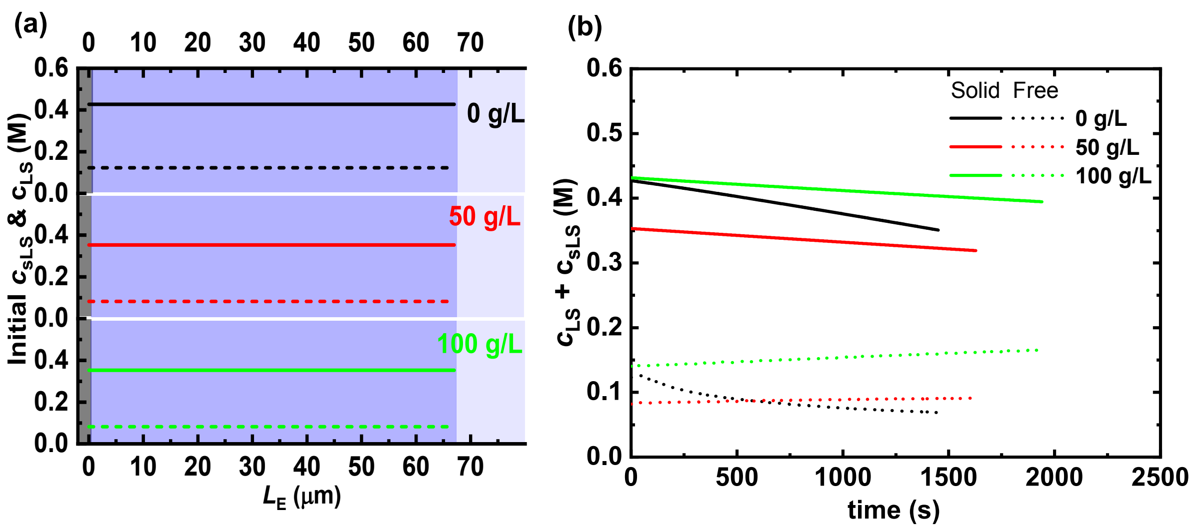

Fig. S1. Concentration of total solid LS species ($sLS,Red + sLS,Ox$) and total dissolved LS ($LS,Red + LS,Ox$) for the three 1_2 electrode systems. (a) Uniform initial profile in electrode thickness L_E used for the model. (b) Simulated transition during 20 cycles (mid-point of electrode).

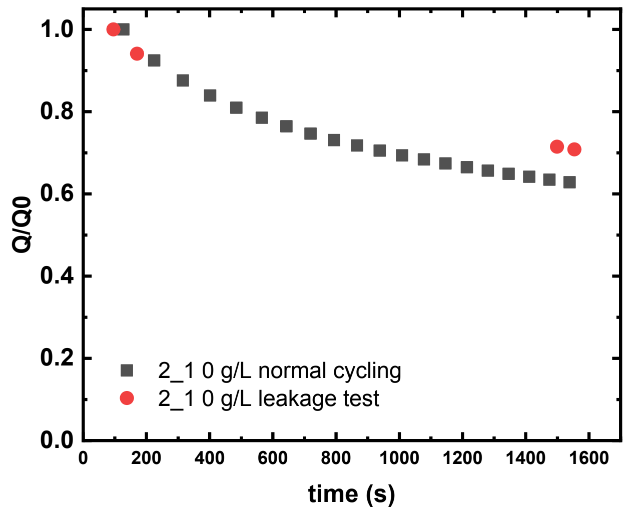

Fig.S2. Experimental results for charge-discharge at 1689 μA with 1_2 0g/L. Comparison of relative discharge capacity for normal cycling and a leakage test (2 cycles, then 20 mins wait before continuation). The test shows that capacity decay is mostly dictated by passive release of preloaded LS from the electrode with time.

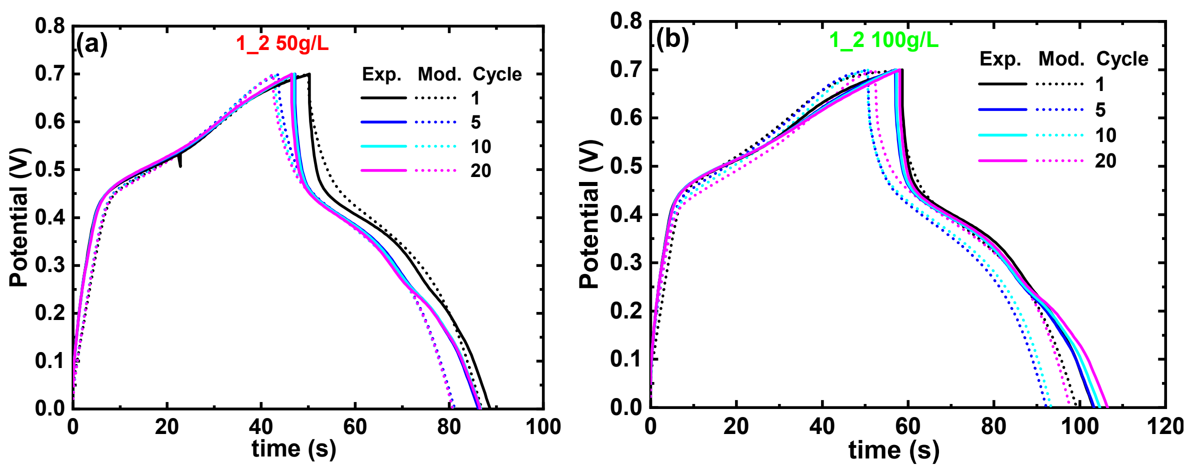

Fig. S3. Experimental (exp.) and model (mod.) results showing transient response during cycling at 1689 μA for (a) 1_2 50g/L, and (b) 1_2 100g/L.

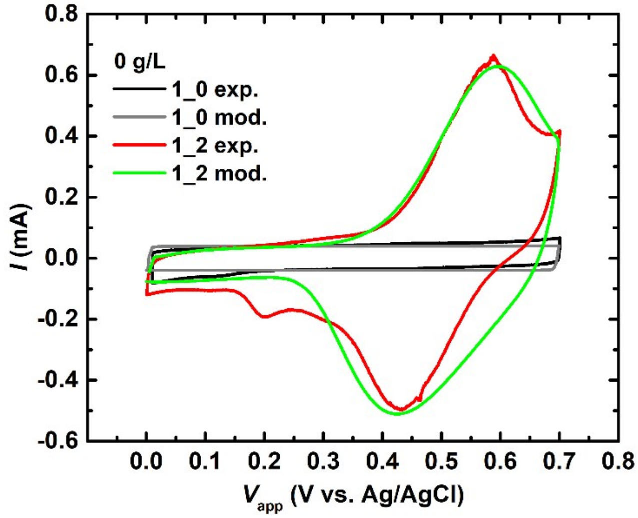

Fig.S4. Simulated and experimental (from J. Edberg *et al*., Journal of Materials Chemistry A, 2018, 6, 145-152.) cyclic voltammograms at 2 mV/s for 1_0 and 1_2 electrodes.

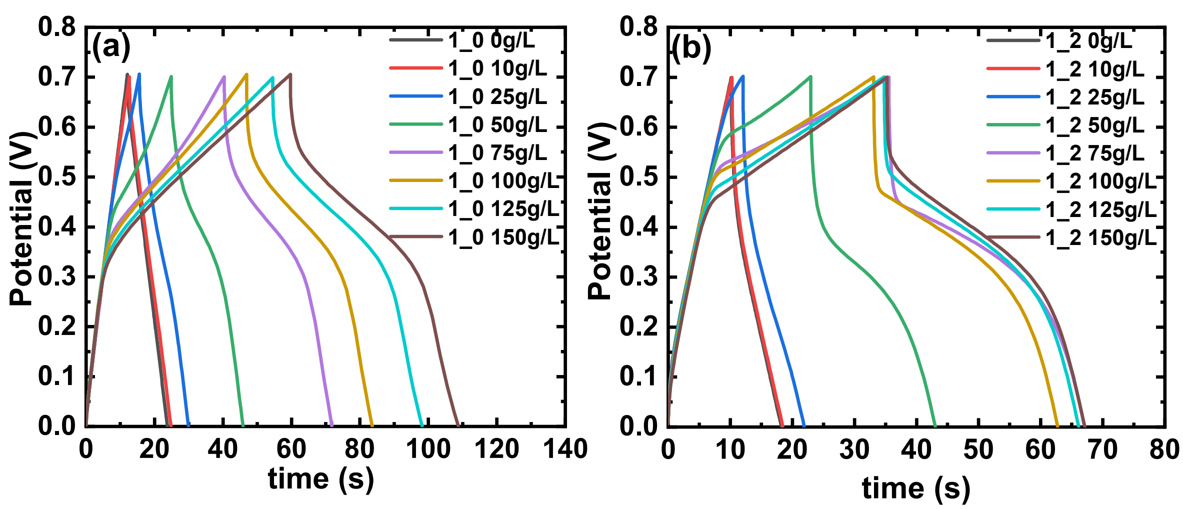

Fig.S5. Simulated charge-discharge responses under at 1689 μA at steady operation (after long cycling) for a half-cell used to construct Ragone plot: (b) 1_0 electrode, and (b) 1_2 electrode.

Supplementary Text

Para-benzoquinone (Q) reduction to hydroquinone (QH_2) mechanism in aqueous solution and overall reaction in high acidity medium (P. S. Guin, S. Das and P. C. Mandal, International Journal of Electrochemistry, 2011, 2011, 22.).

$$Q + e^- \leftrightarrow Q^{*-} \quad \text{(Step 1)}$$

$$Q^{*-} + H^+ \leftrightarrow QH^* \quad \text{(Step 2)}$$

$$QH^* + e^- \leftrightarrow QH^- \quad \text{(Step 3)}$$

$$QH^- + H^+ \leftrightarrow QH_2 \quad \text{(Step 4)}$$

$$Q + 2H^+ + 2e^- \leftrightarrow QH_2 \quad \text{(Overall)}$$

Supplementary Table 1: Nominal operating parameters and variables for the model.

Description	**Model**				
	1_0 50 g/L	**1_0 50 g/L**	**1_2 0 g/L**	**1_2 50 g/L**	**1_2 100 g/L**
Hole diffusivity in PEDOT (cm^2s^{-1})	2.05e-1				
Relative Permittivity PEDOT	40				
Relative Permittivity bulk electrolyte	75				
Cation/anion diffusivity in bulk electrolyte (cm^2s^{-1})	2.2e-5/1.7e-5				
Dissolved LS diffusivity in electrolyte (cm^2s^{-1})	1.1e-6				
Cation/anion diffusivity in electrode (cm^2s^{-1})	1.3e-7/1.0e-7		3.3e-8/2.6e-8		6.6e-8/5.1e-8
***PSS*$^-$ concentration (M)**	0.96		0.74		
Electrode thickness (µm)	52		67		
Dissolved LS diffusivity in electrode (cm^2s^{-1})	-	6.6e-9	1.7e-8		4.6e-10
$k_{sLS,Red}/k_{sLS,Ox}$ **(s^{-1})**	-	-	2.5e-7/7.5e-6	2.4e-7/7.2e-6	4.2e-7/8.4e-6
$k_{LS,Red}/k_{LS,Ox}$ **(s^{-1})**	-	9.2e-3/9.2e-4	4.5e-4/6.3e-5	1.3e-4/5.2e-6	2.0e-3/4.0e-6
$\alpha_{sLS,Red}/\alpha_{sLS,Ox}$	-	-	0.5/0.2		
$\alpha_{LS,Red}/\alpha_{LS,Ox}$	-	0.5/0.2			