Supplementary Materials

Elucidating Spatial Distribution of Electrochemical Reaction in a Porous Electrode by Electrochemical Impedance Spectra for Flow Batteries

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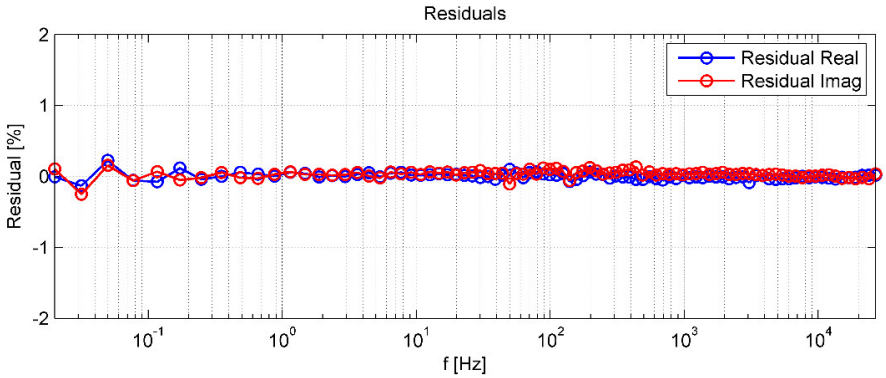

Figure S1. Relative residuals of the linear Kramers-Kronig transform.
The cell was assembled using 5 mm-thick electrodes. The electrolyte concentration was 0.050 M $K_3[Fe(CN)_6]$ at 50% SOC. The impedance spectrum was collected using the two-electrode configuration at OCV at a flow rate of 80 mL min^{-1}.

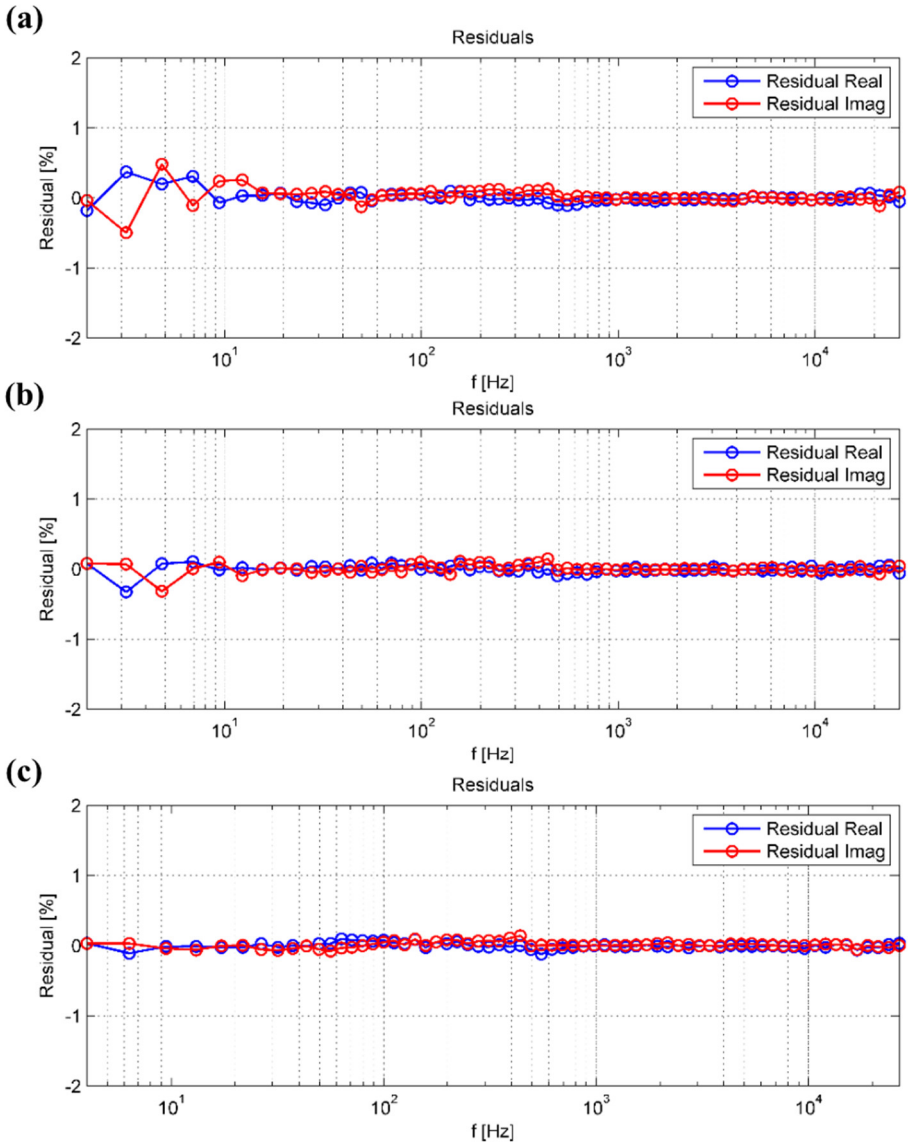

Figure S2. Relative residuals of the linear Kramers-Kronig transform. (**a**) 3 mm-thick electrodes with an anodic polarization of 200 mV; (**b**) 5 mm-thick electrodes with an anodic polarization of 275 mV; (**c**) 8 mm-thick electrodes with an anodic polarization of 375 mV. The electrolyte concentration was 0.050 M $K_3[Fe(CN)_6]$ at 50% SOC. The electrolyte flow rate was 20 mL min^{-1}. The impedance spectra were collected using the three-electrode configuration.

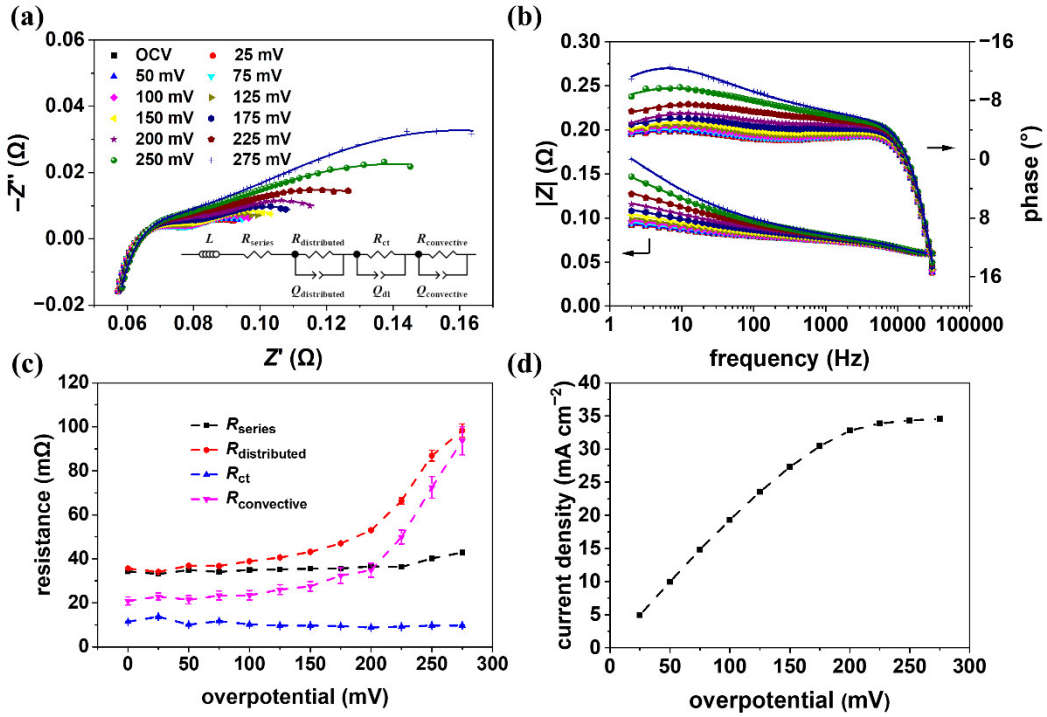

Figure S3. Effect of anodic polarization on the electrochemical impedance and the current density. (**a**) Nyquist plots; (**b**) Bode plots; (**c**) resistance value derived from the data fitting; (**d**) polarization curve. The cell was assembled using 5 mm-thick electrodes. The electrolyte concentration was 0.050 M $K_3[Fe(CN)_6]$ at 50% SOC. The measurements were performed at a flow rate 20 mL min^{-1}. The symbols and the solid lines in Nyquist and Bode plots show the experimental and the fitted data, respectively.

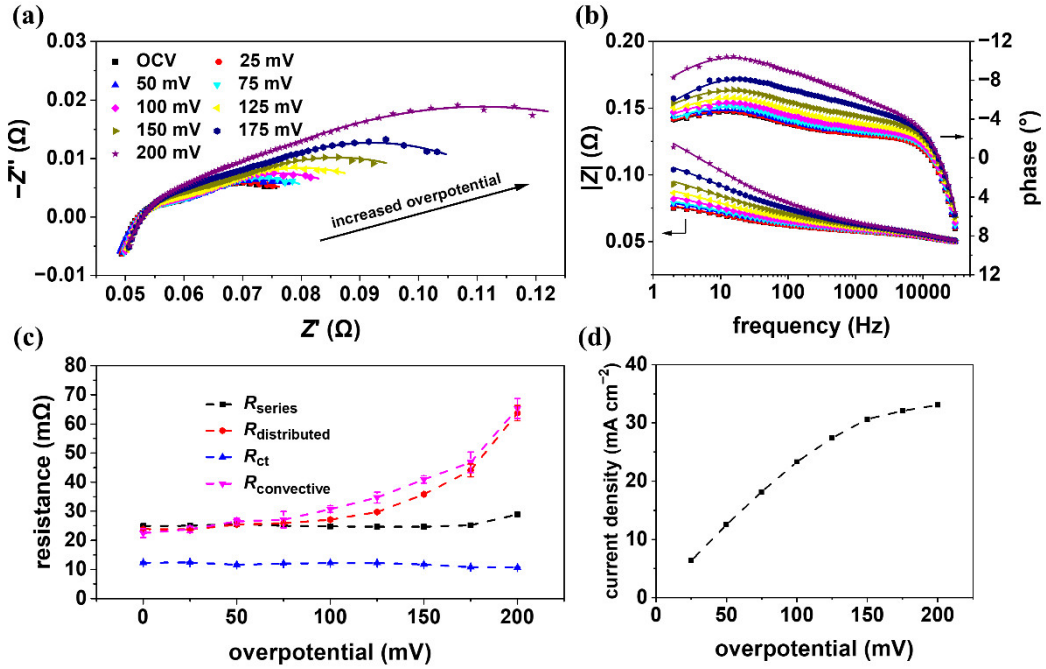

Figure S4. Effect of anodic polarization on the electrochemical impedance and the current density. (a) Nyquist plots; (b) Bode plots; (c) resistance value derived from the data fitting; (d) polarization curve. The cell was assembled using 3 mm-thick electrodes. The electrolyte concentration was 0.050 M $K_3[Fe(CN)_6]$ at 50% SOC. The measurements were performed at a flow rate 20 mL min^{-1}. The symbols and the solid lines in Nyquist and Bode plots show the experimental and the fitted data, respectively.

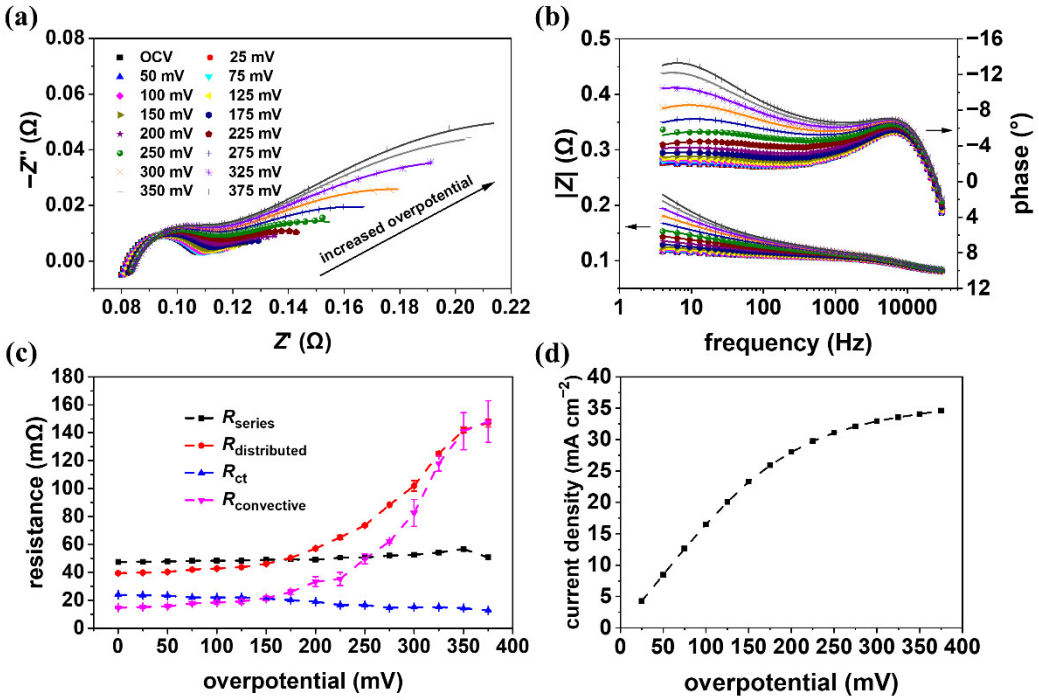

Figure S5. Effect of anodic polarization on the electrochemical impedance and the current density. (a) Nyquist plots; (b) Bode plots; (c) resistance value derived from the data fitting; (d) polarization curve. The cell was assembled using 8 mm-thick electrodes. The electrolyte concentration was 0.050 M $K_3[Fe(CN)_6]$ at 50% SOC. The measurements were performed at a flow rate 20 mL min^{-1}. The symbols and the solid lines in Nyquist and Bode plots show the experimental and the fitted data, respectively.

Table S1. Fitting results of flow battery impedance using the two-electrode configuration at 80 mL min^{-1} with 0.050 M $K_3[Fe(CN)_6]$ at 50% SOC.

Parameter	Value	Significance	Error
L (nH)	198	0.514	1.00%
R_{series} (mΩ)	49.0	0.677	0.46%
$R_{distributed}$ (mΩ)	46.3	0.346	0.14%
$Q_{distributed}$ (mS·s^{α})	80.2	0.179	0.37%
$\alpha_{distributed}$	0.497	0.318	1.61%
R_{ct} (mΩ)	27.2	0.227	0.24%
Q_{dl} (mS·s^{α})	1.59	0.139	0.15%
α_{dl}	0.927	0.395	0.26%
$R_{convection}$ (mΩ)	11.8	0.081	2.51%
$Q_{convection}$ (S·s^{α})	2.66	0.034	1.09%
$\alpha_{convection}$	0.765	0.120	0.73%
$R_{diffusion}$ (mΩ)	76.6	0.197	3.74%
$Q_{diffusion}$ (S·s^{α})	39.8	0.191	1.16%
$\alpha_{diffusion}$	0.904	1.70	0.13%