

Supplementary Information for:

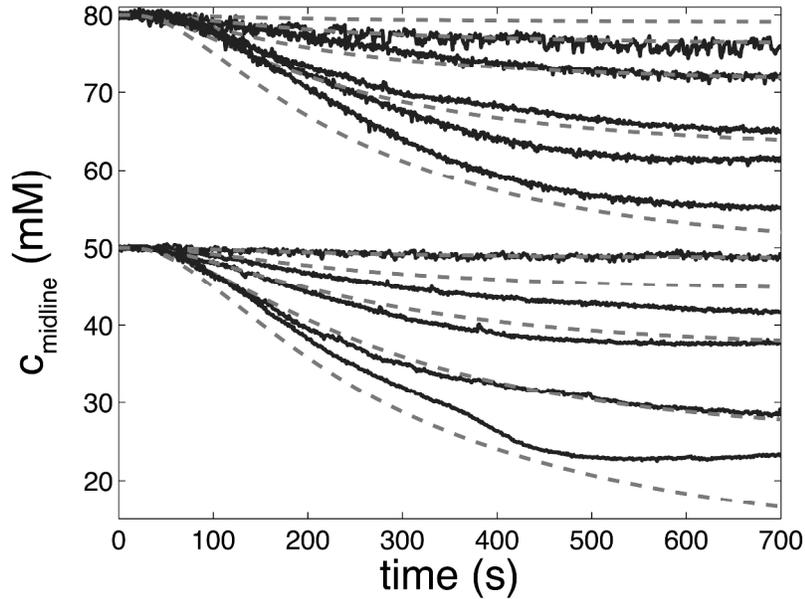
In situ spatially and temporally resolved measurements of salt concentration between charging porous electrodes for desalination by capacitive deionization

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This document includes two pages of additional comparisons between our modified Donnan (mD) model results and experimental results, including two additional figures.

### SI1: Additional comparisons of salt concentration profiles

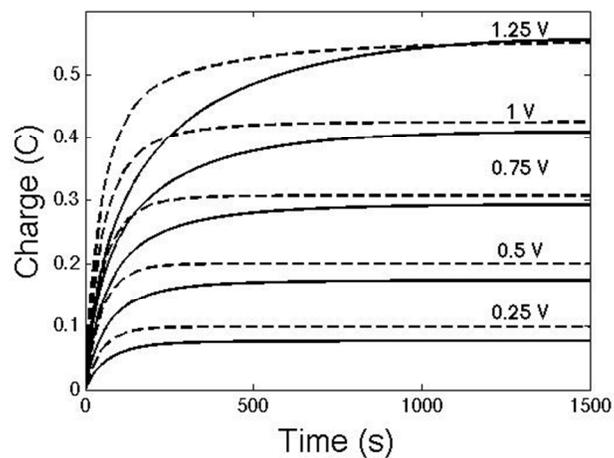
In Figure SI1, we present additional model-to-experiment comparisons for a wider range of experimental conditions, including 50 and 80 mM KCl concentrations and applied cell voltages of 0.25 to 1.25 V.



**Figure SI1:** Comparison of the predicted (dashed lines) and measured (solid lines) cell midline concentration,  $C_{midline}$ , versus time after application of a step potential to the CDI cell. The results are shown for initial concentrations of 50 and 80 mM KCl, and for 0.25, 0.5, 0.75, 1, 1.25 V applied potentials. For both model and experimental results,  $C_{midline}$  at 700 s decreased with higher voltage applied. Also, the model captures the general trend seen in the experimental results in that at early times ( $< 100$  s), very little desalination occurs. At later times, significant desalination occurs and  $C_{midline}$  approaches a steady value.

### SI2: Comparisons of electric charge stored

In Figure SI2, we present model to experiment comparisons for electric charge stored in the cell.



**Figure SI2:** Comparison of the predicted (dashed lines) and measured (solid lines) electric charge stored in the CDI cell versus time after application of a step potential. The results are shown for initial concentrations of 50 mM, and for 0.25, 0.5, 0.75, 1, 1.25 V applied to the cell. For both model and experimental results, electric charge increases rapidly within the first 100 s, and then plateaus after about 500 s. Charge stored increases with cell voltage applied. Small discrepancies between the model and experimental equilibrium charge stored can be seen, as can be also seen in Figure 2b of the main text. At higher voltages such as 1.25 V, differences in the dynamics between the model and experimental results can be seen, namely that at early times, the model underpredicts the experimentally observed charging rate.