

## Supplementary Information

### Stepwise Ammonium Enrichment Using Selective Battery Electrodes

Moon Son <sup>a</sup>, Eric Kolvek <sup>b</sup>, Taeyoung Kim <sup>c</sup>, Wulin Yang <sup>a</sup>, Johannes S. Vrouwenvelder <sup>d</sup>, Christopher A. Gorski <sup>a</sup>, Bruce E. Logan <sup>a,\*</sup>

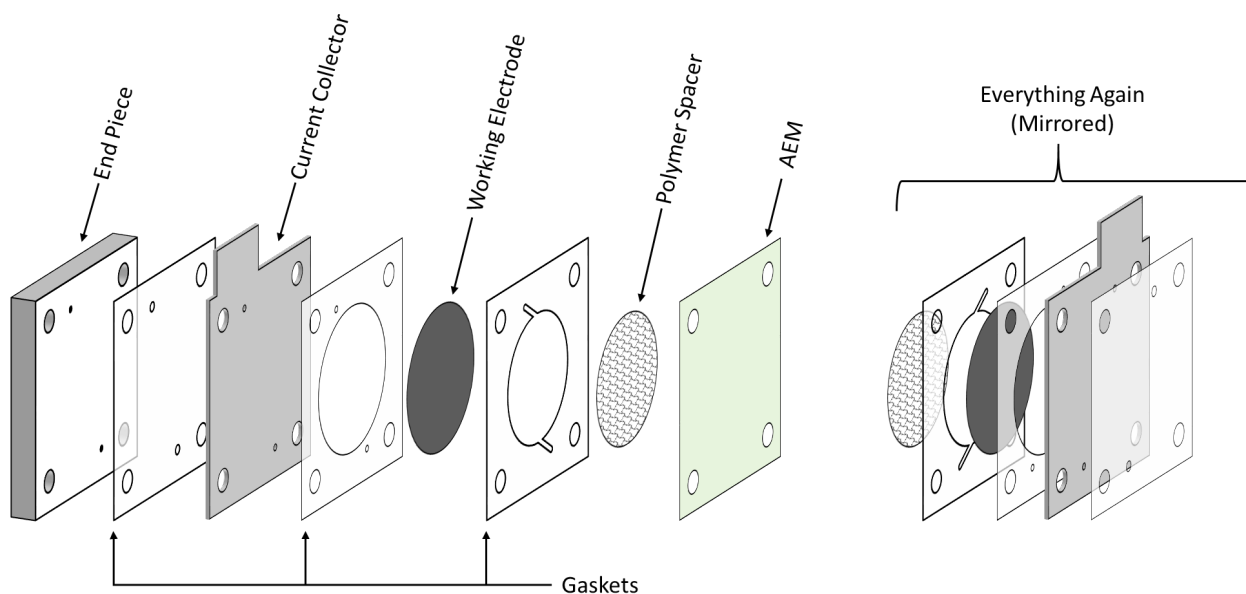
<sup>a</sup> Department of Civil and Environmental Engineering, The Pennsylvania State University, University Park, PA 16802, USA

<sup>b</sup> Department of Chemical Engineering, The Pennsylvania State University, University Park, PA 16802, USA

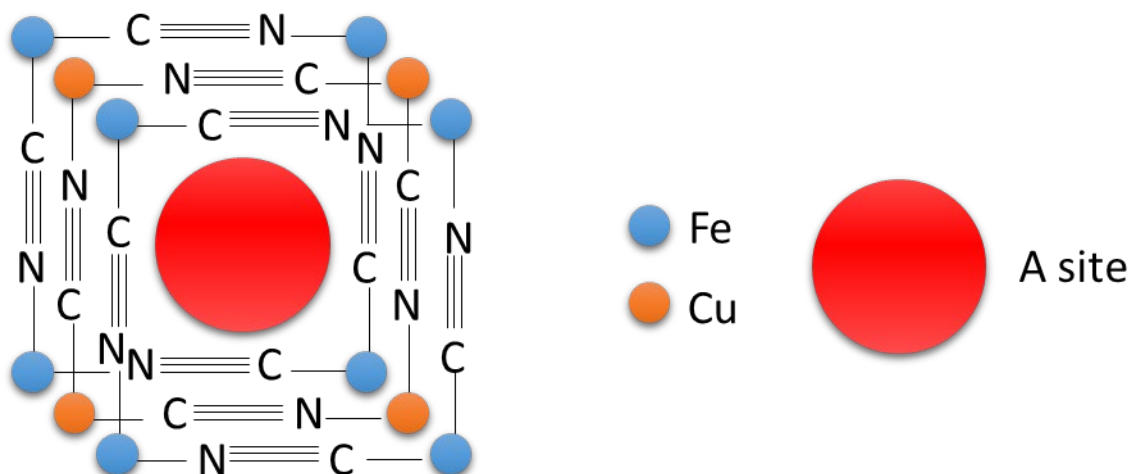
<sup>c</sup> Department of Chemical and Biomolecular Engineering, and Institute for a Sustainable Environment, Clarkson University, Potsdam, NY 13699, USA

<sup>d</sup> Water Desalination and Reuse Center (WDRC), Division of Biological and Environmental Science and Engineering (BESE), King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Saudi Arabia

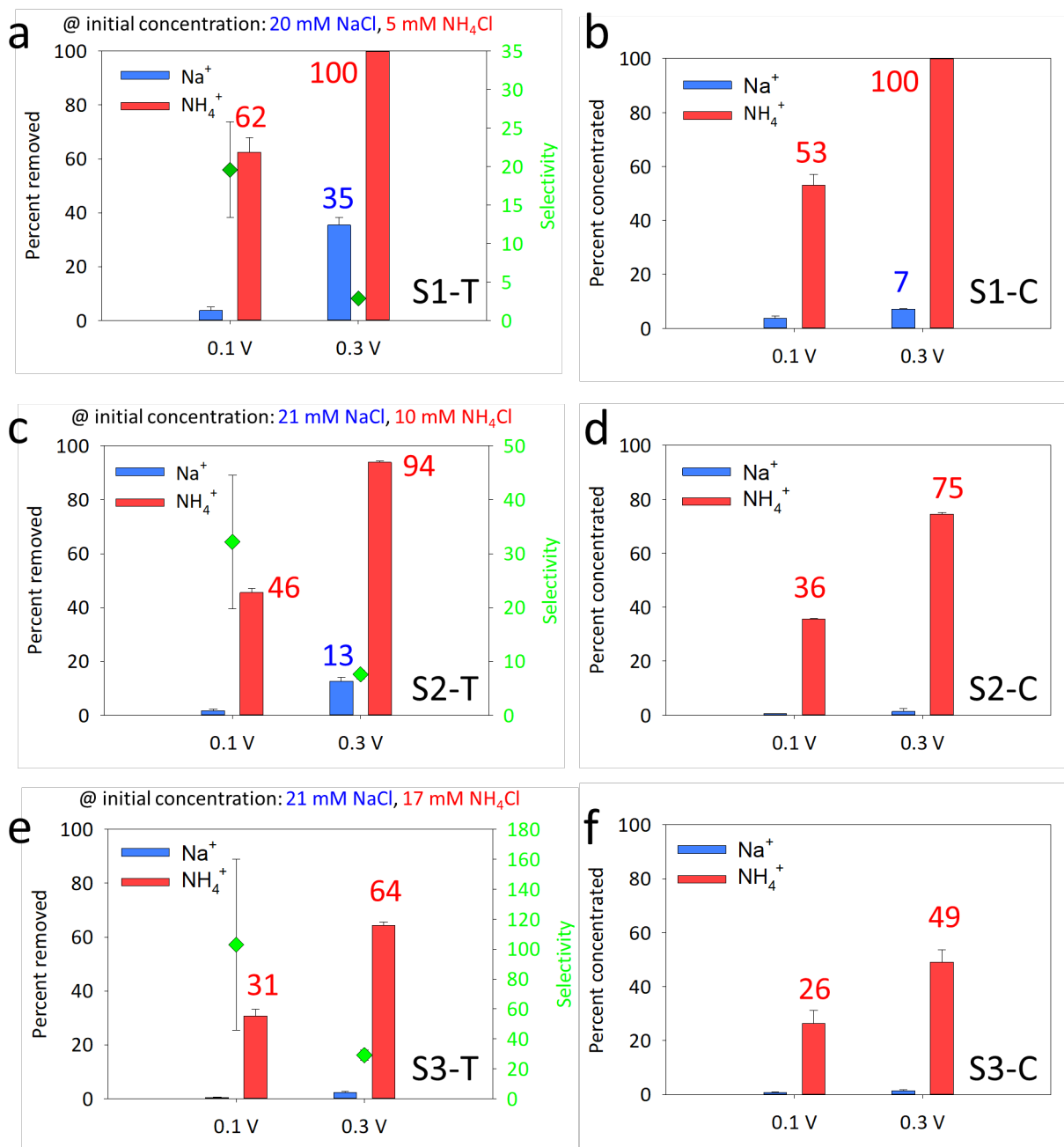
\* Corresponding author. Email: [blogan@psu.edu](mailto:blogan@psu.edu); Tel.: +1-814-863-7908



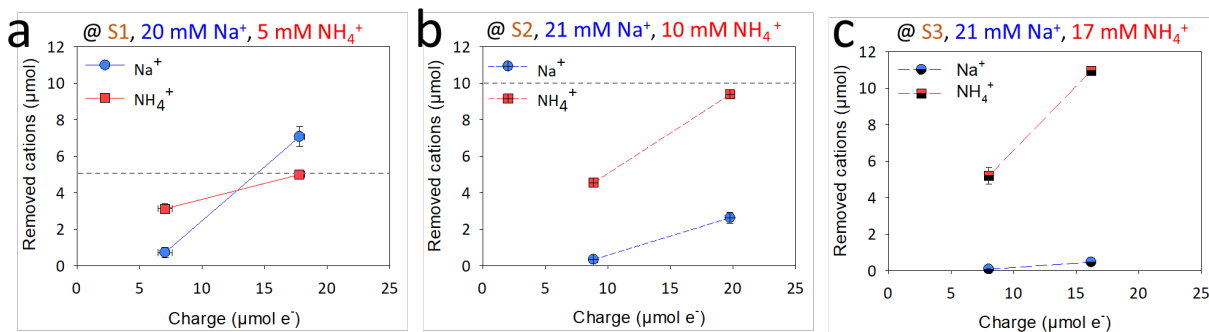
**Fig. S1** Schematic of the flow cell assembly. Redrawn image from previous work.<sup>S1</sup> The effective membrane area was 7 cm<sup>2</sup>.



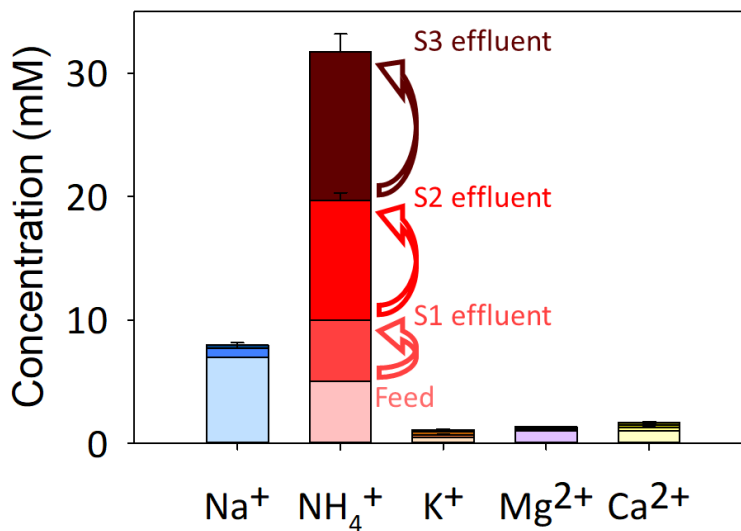
**Fig. S2** (a) Structure of copper hexacyanoferrate (CuHCF). The size of the interstitial sites (A site) is 3.2~4.6 Å. Redrawn image from previous work.<sup>S2</sup>



**Fig. S3** Percent removed or concentrated of  $\text{Na}^+$  and  $\text{NH}_4^+$  of (a–b) S1, (c–d) S2, and (e–f) S3 as a function of the cell voltage from 0.1 to 0.3 V. Selectivity of ions removal ( $\text{NH}_4^+/\text{Na}^+$ ) is also shown for the treated stream.



**Fig. S4** Removed cations of (a) S1, (b) S2, and (c) S3 as a function of the amount of charge. Concentration of the feed waters was varied depending on the effluent concentration of the previous stage (S1: 20 mM NaCl and 5 mM NH<sub>4</sub>Cl, S2: 21 mM NaCl and 10 mM NH<sub>4</sub>Cl, and S3: 21 mM NaCl and 17 mM NH<sub>4</sub>Cl). Dash lines indicate initial NH<sub>4</sub><sup>+</sup> concentration present in the solution.



**Fig. S5** Concentration of the feed and concentrated effluents of each stage of cations over three stages at an applied voltage of 0.3 V using synthetic wastewater.

## References

- S1. T. Kim, C. A. Gorski and B. E. Logan, Ammonium removal from domestic wastewater using selective battery electrodes, *Environ. Sci. Technol. Lett.*, 2018, **5**, 578-583.
- S2. C. D. Wessells, S. V. Peddada, M. T. McDowell, R. A. Huggins and Y. Cui, The effect of insertion species on nanostructured open framework hexacyanoferrate battery electrodes, *J. Electrochem. Soc.*, 2011, **159**, A98-A103.