

## Supporting information for

### Chemically Sodiated Iron Disulfide as a High-Capacity Charge-Storage Host with Cation- and Anion-sited Redox

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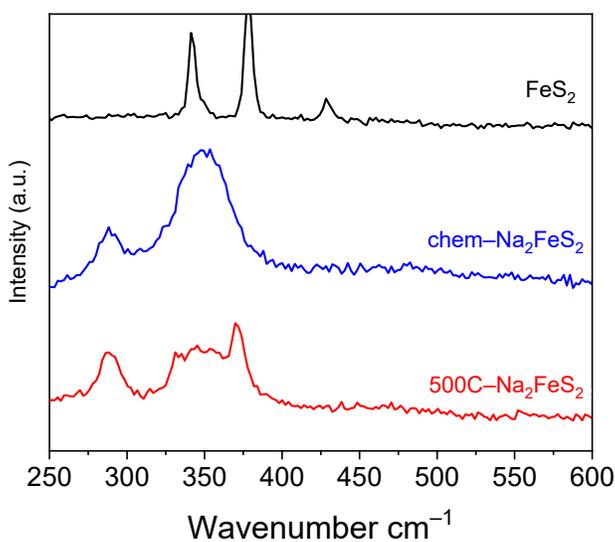


Figure S1. Raman spectra of FeS<sub>2</sub>, chem-Na<sub>2</sub>FeS<sub>2</sub>, and 500C-Na<sub>2</sub>FeS<sub>2</sub>.

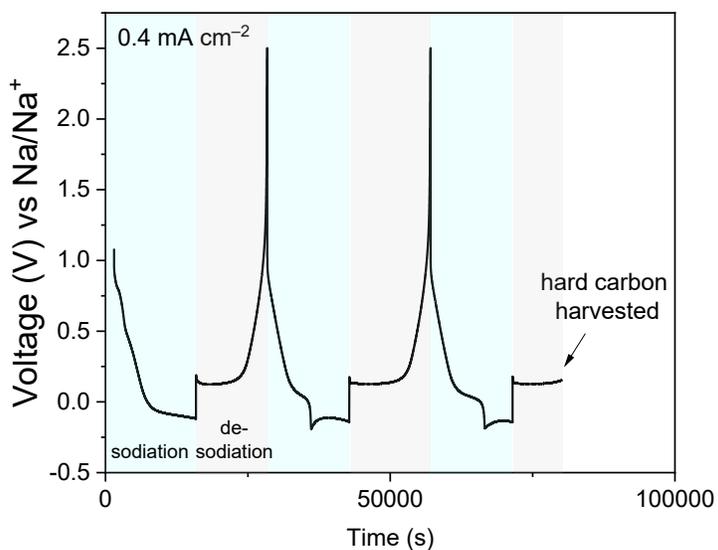


Figure S2. Conditioning profile for hard-carbon || Na metal cells, where both Na metal and HC electrodes are harvested and used as anodes in  $\text{Na}_2\text{FeS}_2$  electrochemical experiments. For the condition profile, the areal current density =  $0.4 \text{ mA cm}^{-2}$ . The desodiation half-cycle is potential controlled, where the cutoff potential is  $2.5 \text{ V vs Na/Na}^+$  for the first two cycles, and  $0.15 \text{ V}$  for the final cycle. The sodiation half-cycle is time controlled (4 h).

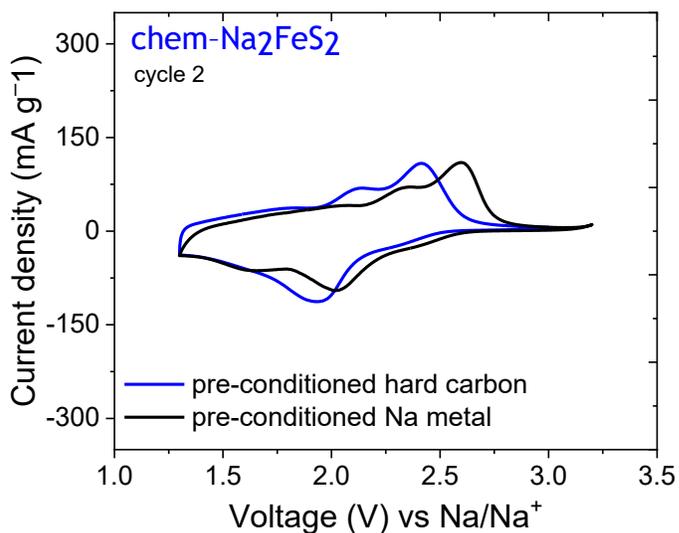


Figure S3. Comparison of chem- $\text{Na}_2\text{FeS}_2$  || pre-conditioned hard carbon anode and chem- $\text{Na}_2\text{FeS}_2$  || pre-conditioned Na metal anode.

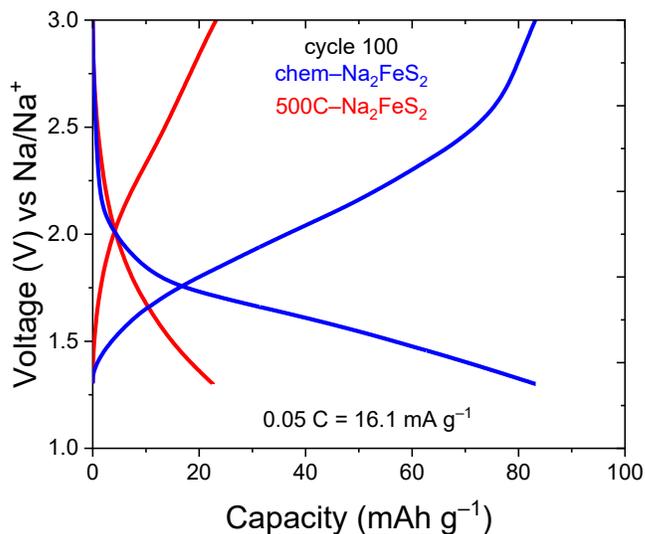


Figure S4. Cycle 100 galvanostatic charge/discharge curves.



Na metal electrodes

Celgard + glass fiber  
separators

NaFeS<sub>2</sub> cathode  
@ Ni foam

Figure S5. Optical image of recovered electrodes and separators from extended cycling chem-Na<sub>2</sub>FeS<sub>2</sub> cells.

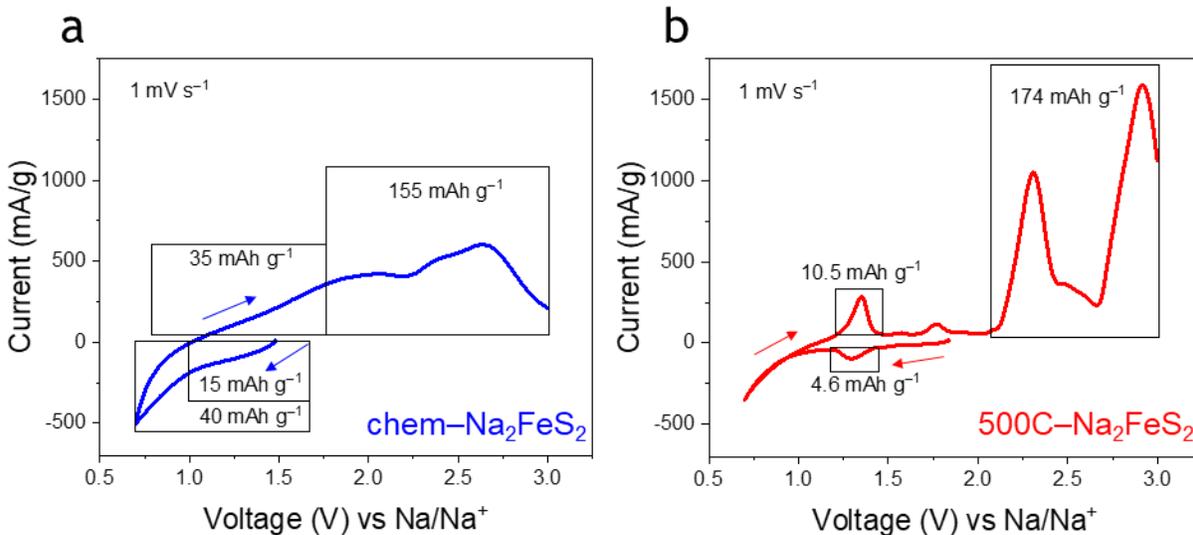


Figure S6. Cyclic voltammograms at  $1 \text{ mV s}^{-1}$  for electrodes containing (a) chem-Na<sub>2</sub>FeS<sub>2</sub> and (b) 500C-Na<sub>2</sub>FeS<sub>2</sub>. Cells start at open-circuit, are swept in the negative direction to 0.7 V (sodiated), then in the positive direction to 3.0 V (desodiated). Boxed regions show the area of curve integrated to calculate capacity. Two-electrode configuration: counter/quasi-reference is a Na metal electrode.

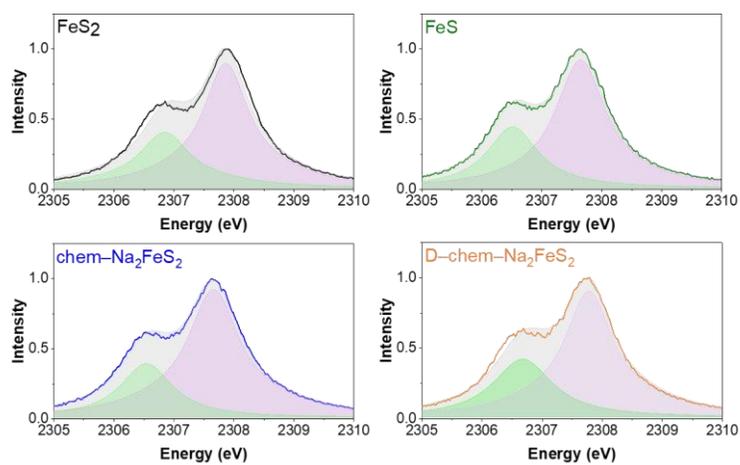


Figure S7. Peak-fits of X-ray emission spectra for FeS<sub>2</sub>, FeS, chem-Na<sub>2</sub>FeS<sub>2</sub>, and D-chem-Na<sub>2</sub>FeS<sub>2</sub>.

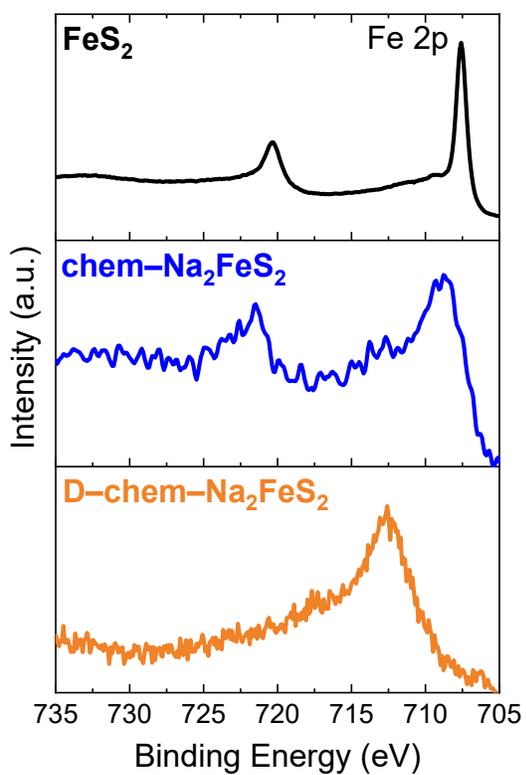


Figure S8. Fe 2p X-ray photoelectron spectroscopy of chem-Na<sub>2</sub>FeS<sub>2</sub>, D-chem-Na<sub>2</sub>FeS<sub>2</sub> and FeS<sub>2</sub>.