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## Supporting Information

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A High Capacity Calcium Primary Cell Based on the Ca–S System

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**Figure S1.** Characterization of hexagonal mesoporous carbon (CMK-3) and cubic mesoporous carbon (OMC-8) sulfur supports. Characterization of these materials includes (a) small angle X-ray diffraction showing the long range ordering of the pore structure, (b) X-ray diffraction showing two broad peaks indicative of amorphous carbon, (c) pore size distributions as determined by the BJH algorithm on the N<sub>2</sub> adsorption isotherm and (d) N<sub>2</sub> adsorption isotherms showing mesoporosity. The Brunauer-Emmett-Teller surface areas are indicated in (d).

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**Figure S2.** Raman spectra (bottom traces) and Fourier-Transform Infrared Spectroscopy (top traces) of electrolyte before cycling (dashed black) and after discharging to 0V (solid red) at -15 °C. There are no additional Raman transitions or FTIR stretches visible indicating that the electrolyte does not contain any contaminants FTIR or Raman active.



**Figure S3.** Electrospray Mass Spectrometry of 0.5 M Ca(ClO<sub>4</sub>)<sub>2</sub> in ACN before and after discharge. The strong signals (99 m/z, 339 m/z, 577 m/z, etc.) are due to ClO<sub>4</sub><sup>-</sup>, Ca(ClO<sub>4</sub>)<sub>3</sub><sup>-</sup>, etc.; however, the weaker signals at 752 m/z, 1262 m/z, etc. in the discharged electrolyte are due to clustering with ClO<sub>3</sub><sup>-</sup>.