Mediated Lithium-Sulfur Flow Batteries

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Overview:
Lithium-sulfur is a next-generation battery technology which leverages an inexpensive sulfur cathode to significantly increase specific capacity. We are working to translate this lithium-sulfur technology to a mediated redox flow battery (RFB), where soluble redox-active molecules are circulated, reducing sulfur particles stored in a reservoir. This design also physically separates the anode and cathode, minimizing safety risks in case of failure. While such systems are attractive for cost-competitive long duration energy storage due to their potential for ultra-high sulfur concentrations, the current density of these RFBs need to be increased to reach competitive power outputs.

Operating Principle
Soluble redox mediators (RM) oxidize and reduce solid, energy-storing sulfur particles kept in the catholyte reservoir tank. Catholyte is flowed into an electrochemical cell where electrons are extracted from mediators at a porous carbon electrode.

Flow Cell Performs Well at Low Current
Redox mediated Li-S flow batteries can be cycled stably over 50 cycles with high capacity and voltage efficiency demonstrated at 2.4 mg, cm⁻²: 1142 mAh g⁻¹ and 86.9% VE. Sulfur loadings of up to 50 mg, cm⁻² were achieved enabling a discharge time of over 60 hours.

Future Work
- Scale up Li-S flow cells to ultra-high S loadings in larger (100 cm²) cells.
- Improve capacity utilization of cells with ZnO-Ni foam anode at higher rates.
- Improve “anode-less” Li-S RFB capacity utilization.

Higher Currents Enabled by a 3D Anode
Replacing planar Li foil with high surface area Ni foam decreases the local current. To increase Li wettability and decrease nucleation overpotentials, the Ni foam was decorated with ZnO nanorods.
Li-Zn alloy was formed on initial charge, and Li metal was plated on top. (CryoFIB cross section)
Symmetric Li cells were cycled at 10 mA cm⁻² with 96.7% Coulombic efficiency over 48 cycles.
10x increase in current density!

Higher Current RFB Cycling with or without Li Metal
The ZnO-Ni foam was prelithiated with molten Li, and cycled in an RFB at high current. Higher RM concentration or faster pumping can increase capacity at 10 mA cm⁻².
RFBs can also be run without any Li metal initially present. Li metal is plated at the anode on initial charge. Highest S loading achieved 20.3 Wh L⁻¹ (8.41 Ah L⁻¹).

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