Aqueous Semi-Solid Flow Cell
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Low-cost and scalable energy storage is vitally important for an electric grid with increasing integration of renewable energy sources such as wind and solar. Various novel energy storage technologies have been proposed to improve grid reliability and utilization, including high energy density flow batteries, aqueous Li-ion, Na-ion,10,12 and K-ion13,14 batteries. Flow batteries, which store active electrochemical “fuel” in external reservoirs and pump them into an ion-exchange-electron extraction power stack to deliver electricity, are considered promising candidates. The inherent flexibility in the architecture of such devices decouples energy and power, which can be exploited to design inexpensive large-scale systems. To meet long-term performance and cost requirements for grid storage, a system-level cost of about $100/kWh is desired.1 Amongst flow battery systems, one of the most well-developed is the vanadium redox battery (VRB), which has 40 Wh/L-reactants and 20-35 Wh/L at the system level.13 Projected system costs currently range between $300-$800/kWh for VRBs.11,16

Accordingly, alternative strategies are sought to increase the energy density of flow batteries and decrease system-level cost.17,18 Dututa et al.1 demonstrated a novel approach, the semi-solid flow cell (SSFC), in which the nominal molar concentration of the flow electrodes (also known as catholyte and anolyte) is markedly increased by integrating solid-state intercalation compounds into a flowing, mixed-conducting suspension. This approach is differentiated from all previous flow batteries by imparting electronic conductivity to the flow electrodes themselves, through the use of a conductive nanoparticle suspension, in contrast to the conventional case where the electronically insulating solutions undergo redox reactions only upon contact with current collectors (typically a porous carbon). The SSFC approach has been explored to date only for non-aqueous Li-ion chemistry,17,18 where the combination of higher effective molarity and higher cell voltage in principle allows energy density to exceed that of aqueous flow batteries by over ten-fold. An SSFC based on aqueous chemistry is of interest for flow batteries by over ten-fold.

In this paper, the LiTi4PO4/LiFePO4 (LTP/LFP) couple is used to demonstrate for the first time an aqueous semi-solid flow cell (A-SSFC). In order to interpret the coupled electrochemical and adveective response inherent to SSFCs, rheological and transport properties of the suspensions are measured, and used to guide computational modeling of charge/discharge behavior with concurrent non-Newtonian flow. Sources of inefficiency resulting from flow-induced equilibration and chemical side reactions are isolated. High coulombic efficiency is demonstrated, and operating conditions (including the amount and rate of pumping) for optimal round-trip coulombic efficiency are determined. A main remaining challenge for practical application is shown to be the side reactions prevalent in aqueous Li-ion chemistry, including cathode stability and anode-mediated hydrolysis.

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