## Advection management in suspension-based flow batteries

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Semi-solid flow cells leverage the high energy density of solid-state lithium ion cells with the scalability of redox flow batteries through the co-suspension of solid-state electrochemical storage materials within an electronconducting gel supported by ion-conducting electrolyte [1]. The fluidity of semi-solid suspensions enables the bulk transfer of electrochemically active material to the electroactive region (i.e., the region over which current collector extends) for the storage and delivery of electrical power to and from an external circuit.

Charge transfer in heterogeneous, mixed-conducting semi-solid suspension occurs via mechanisms absent from conventional flow batteries. Specifically, the mechanical energy dissipated by pumping semi-solid suspensions necessitates the operation of semi-solid flow cells in an intermittent mode characterized by transient fluid pulses [1,2]. The efficiency of this process depends strongly on the shape of the non-Newtonian, pressure-driven flow profile. Plug-like flow profiles that preserve the shape of suspension aliquots (i.e., displaced volumes) upon pumping yield greatest electrochemical efficiency [3].

The time scale associated with intermittent flow pulses is assumed to be substantially shorter than those of the intrinsic electrochemical transport mechanisms in the semi-solid (e.g., electronic and ionic conduction). Therefore, the shape of the velocity field is the primary agent for the redistribution of semi-solid during an intermittent flow pulse. A simplified pumping scenario is depicted in Fig. 1 to illustrate this effect. While a pluglike velocity profile displaces the charged semi-solid volume uniformly, the Newtonian-like velocity field does not transfer charged suspension near the wall outside the active charging zone. Also, the Newtonian-like flow distends discharged suspension into the charged tank.



Fig. 1: A simplified pumping scenario in which (a) a unit aliquot of suspension is charged and that aliquot is ejected from the charging region by (b) a high-velocity, Newtonian-like flow and (c) a low-velocity, plug-like flow, each with equal size of the displaced aliquot.

The non-uniform advection (i.e., bulk fluid motion) of charged suspension decreases the efficiency of semi-solid flow cells, firstly, because of the state-of-charge gradients induced in the electroactive region. Secondly, increased interfacial area between charged and uncharged suspension leads to rapid equilibration of charge outside of the electroactive region.

Based on this understanding pumping sequences are explored through the numerical simulation of charge and discharge steps in sequence with intermittent flow pulses of various flow types (e.g., slip flows and Binghamplastic flows). Sequences are identified that simultaneously maximize energetic efficiency and discharge energy of the semi-solid flow cell.

## **References:**

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## Acknowledgements:

This work was funded by the Advanced Research Projects Agency – Energy (ARPA-E), U. S. Department of Energy award number DE-AR0000065.