Abstract

Electrochemical and Rheological Analysis of Flowable Conducting Suspension Electrodes for Scalable Energy Storage

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Following the advancements in materials development for electrochemical systems, flowable suspension-type (i.e. semi-solid) electrodes have gained much attraction in the last decade, offering scalability and flexible design for many conventional energy storage technologies. Flowable suspension electrodes are multi-phase mixtures of liquid electrolytes and electrochemically active solid particles that can offer fluidization of traditional electrode chemistries and allow adaptation of highly scalable flow-assisted system architectures for large-scale energy storage applications. In suspension electrodes, volume spanning networks of conducting particle clusters facilitate the transport of charges under dynamically changing conditions and the liquid electrolyte phase is responsible for the physical transport of ions and the solid particles. Due to their complex nature, experimental and theoretical approaches are required from a host of disciplines to fully investigate the governing properties of suspension electrodes. Despite it's significance, optimizing the electrochemical and rheological parameters not only for efficient electrochemical performance but also for low-dissipation flow, remains to be one of the least explored areas.

This thesis aims to address the existing gap in rheological understanding of capacitive suspensions in relation to their electrochemical performance and focuses on establishing key structure-property relationships that can ultimately help us achieve low-dissipation flow, high electrochemical performance capacitive flowable systems. This is accomplished through systematic experimentation to establish a link between key rheological properties and the electrochemical performance. Multi-component carbon suspensions consisting of high-surface area porous carbons and conducting carbon nanoparticles are used as model systems to investigate the effect of particle morphology on charge percolation and rheological behavior of flowable suspension electrodes. A systematic study focusing on different types of slurry preparation protocols is carried out to determine the impact of mixing on electrical conductivity and electrochemical performance of capacitive suspensions. Alternative electroactive materials with hydrophilic surface groups such as MXenes are explored, investigating the potential of clay-like conducting particles for low-dissipation flow FE applications. Lastly, focusing on minimizing rheology dependent losses (e.g. pumping) and improving the power output of low-viscosity suspensions, emphasis is given to alternative approaches to flow cell design and an overview of porous electrode integration to slurry electrode systems is provided.