Rational Design Strategies for Redox Flow Batteries

by

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Submitted to the Department of Chemical Engineering on 05 May 2023, in partial fulfillment of the requirements for the degree of
Doctor of Philosophy in Chemical Engineering

Abstract:

Global decarbonization of the energy sector necessitates development of storage technologies to mediate the inherent intermittency of renewable resources. Electrochemical systems are well-positioned to support this transition with redox flow batteries (RFBs) emerging as a promising grid-scale platform, as their unique architecture offers decoupled energy / power scaling, simplified manufacturing, and long service life. Despite these favorable characteristics, current embodiments remain prohibitively expensive for broad adoption, motivating the development of new electrolyte formulations (e.g., redox molecules, supporting salts, solvents) and reactor materials (e.g., electrodes, flow fields, membranes) to meet performance and cost targets for emerging applications. While many next-generation materials offer performance improvements, they must carefully balance complex tradeoffs between power / energy density, cycling stability, energy efficiency, and capital costs. This multifaceted parameter space frustrates the articulation of unambiguous design criteria, as the relationships between constituent material properties and cell performance metrics are not yet well-understood.

In this thesis, I address these knowledge gaps by advancing theoretical and experimental methods that support the rational design of RFBs. First, I develop an in-line electrochemical sensor to measure electrolyte concentrations during RFB operation, providing insight into the dynamics of flow cell cycling. Second, I introduce an analytical zero-dimensional modeling framework for describing RFB cycling behavior, enabling facile simulation of charge / discharge behavior and device performance metrics. I then further simplify this approach by deriving closed-form expressions, facilitating the use of spreadsheet models for cycling simulations. Third, I apply these models to assess design tradeoffs for two-electron materials, demonstrating marked performance limitations. Finally, I leverage the analytical zero-dimensional framework to develop a new technique—compositionally unbalanced symmetric cell cycling—for characterizing crossover rates in redox flow cells. Broadly, the methods developed in this work have the potential to advance foundational understanding in RFB design and operation, leading to more rigorous selection criteria for candidate materials and ultimately supporting more robust, cost-competitive, and durable grid-scale energy storage.

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