On the Impact of Electrode Properties and Their Design for Redox Flow Battery Performance

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Redox flow batteries (RFBs) are a promising technology for grid energy storage. However, cost reductions are required prior to widespread adoption. Advances in the design and engineering of the electrochemical stack may enable cost reductions for multiple redox chemistries. Porous electrodes are a prime target for improvement of system power to lower cost per kilowatt-hour, as they are responsible for multiple critical functions in the flow cell including providing surfaces for electrochemical reactions, distributing liquid electrolytes, and conducting electrons and heat. However, there is limited knowledge on how to systematically design and implement these materials in emerging RFB applications, leading to the repurposing of available materials that are not tailored for this system, i.e. porous carbon papers or felts. For optimal RFB performance, it is necessary to pretreat carbons prior to use to improve electrode wetting and enhance redox kinetics, yet the impact of thermal pretreatment on electrode properties and the correlation between these properties are not well defined, thus the subsequent influence on performance is nebulous. Gaining a deeper understanding of electrode properties and their influence on performance will enable targeted improvements to electrode platforms, allowing system-specific performance gains. Further, identifying essential electrode properties will guide the development of alternative electrocatalytic material that may enable new systems in which carbon is unstable or is not catalytically active.

In this thesis defense, I will discuss the impact of electrode treatments on RFB performance, combining experimental and computational approaches. First, I investigate the interrelated effects of thermal pretreatment on electrode properties and correlate the changes in these properties with performance. Surface functionalization, wetting, and surface area are identified as the key properties that influence electrode performance. Next, I specifically investigate the impact of surface area on electrode performance. I show that, while thermal treatment adds a significant amount of physical surface area to the electrode, electrochemical species are unable to access a large fraction of this surface area. Further, I use a convection-reaction model to show that even when all surface area is accessible, there is a limit to the surface area that will improve electrode performance. This limit to “useful” surface area is dictated by rate of reaction and transport within the electrode. Finally, I investigate the viability of nickel metal electrodeposition on carbon electrodes to enhance the performance of a novel polysulfide-permanganate flow battery. I show that nickel-deposited carbon electrodes outperform commercially available metal materials, including foams and weaves. The overarching goal of this thesis work is to develop a deeper understanding of the influence that electrode properties have on performance. By continuing to characterize the fundamental kinetic and transport properties within complex porous materials under forced convection, the community will be prepared to design novel material sets well-suited for use in RFBs and other challenging electrochemical environments.