Materials Science and Engineering

PhD Thesis Defense

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CHARGE STORAGE MECHANISM OF REDOX POLYMERS FOR ENERGY STORAGE

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Abstract

Organic redox polymers have recently attracted significant attention for energy storage applications due to their high capacity, high abundance, and environmental friendliness. While most studies focused on electrochemical performance, their charge storage mechanisms, especially *n*-type polymers, are not well-studied. This dissertation aims to investigate the charge storage mechanism of *n*-type polymers by electrochemical quartz crystal microbalance with dissipation monitoring (EQCM-D). The swelling behaviors of polymers and solvation structures for charge carriers are investigated, and their implications for battery performance are discussed.

First, we investigate the charge storage mechanism for a π-conjugated *n*-type polymer P(NDI2OD-T2). We find that the solvated ion clusters serve as charge carriers for polymer redox reactions, whose solvation number varies with solvents. The rate performance of the polymer electrode is determined by its degree of swelling in electrolytes. Secondly, we study the charge storage mechanism of a non-conjugated polyimide-based polymer in different electrolytes. The solvation number for charge carrier varies with electrolyte concentration and solvent types. The reversible capacity of the polymer electrode is determined by the degree of polymer swelling, while its cycle stability is affected by the ion cluster size. Thirdly, the charge storage mechanism of poly(benzoquinonyl sulfide) is investigated for zinc-ion battery applications. Non-hydrated zinc ions are found to be the cation species associated with the quinone-related redox reaction, while the counter anions also participate in the reaction probably due to the unique *p*-dopable linker present in the polymer. The dimension change of polymer electrodes during cycling can be related to the solvated anions, which is the root cause of the capacity decay. Finally, zinc electrodeposition

process is studied in the presence of separators. Porous separators such as commercial glass fiber and polypropylene membranes promote directional pore-filling by deposited zinc. The thus-formed porous zinc forms ‘dead zinc’ upon stripping. In contrast, a nonporous poly(dimethylsiloxane-ethylene glycol polymeric) network separator confines zinc deposition beneath the separator and prevents the formation of dead zinc. This work paves the way for the further application of EQCM-D methodology for charge storage mechanism study and promotes the development of polymeric electrodes for energy storage applications.