abstract

The electrode-electrolyte interface in electrochemical environments involves the understanding of complex processes relevant for all electrochemical applications. Some of these processes include electronic structure, charge storage, charge transfer, solvent dynamics and structure and surface adsorption. In order to engineer electrochemical systems, no matter the function, requires fundamental intuition of all the processes at the interface. The following work presents different systems in which the electrode-electrolyte interface is highly important. The first is a charge storage electrode utilizing percolation theory to develop an electrode architecture producing high capacities. This is followed by Zn deposition in an ionic liquid in which the deposition morphology is highly dependant on the charge transfer and surface adsorption at the interface.

Electrode Architecture: A three-dimensional manganese oxide supercapacitor electrode architecture is synthesized by leveraging percolation theory to develop a hierarchically designed tri-continuous percolated network. The three percolated phases include a faradaically-active material, electrically conductive material and pore-former templated void space. The micropores create pathways for ionic conductivity, while the nanoscale electrically conducting phase provides both bulk conductivity and local electron transfer with the electrochemically active phase.

Zn Electrodeposition: Zn redox in air and water stable N-ethyl-N-methylmorpholinium bis(trifluoromethanesulfonyl)imide, [C2nmm][NTf2] is presented. Under various conditions, characterization of overpotential, kinetics and diffusion of Zn species and morphological evolution as a function of overpotential and Zn concentration are analyzed. The surface stress evolution during Zn deposition is examined where grain size and texturing play significant roles in compressive stress generation. Morphological repeatability in the IL’s led to a novel study of purity in ionic liquids where it is found that surface adsorption of residual amine and chloride from the organic synthesis affect growth characteristics.

The drivers of this work are to understand the processes occurring at the electrode-electrolyte interface and with that knowledge, engineer systems yielding optimal performance. With this in mind, we were able to design a bulk supercapacitor electrode architecture with excellent composite specific capacitances, as well as develop conditions producing ideal Zn deposition morphologies.