

Self-Organizing Electrochemical Devices via Particle Phase Separation

Steven M. Tobias,^a Jennifer Giocondi,^a Dong-Wan Kim,^a
W. Douglas Moorehead,^b Sung-Yoon Chung^c and
Yet-Ming Chiang^a

^a Department of Materials Science and Engineering
Massachusetts Institute of Technology
Cambridge, MA 02139

^b U.S. Navy, Virginia Beach, VA

^c Inha University, Incheon, Korea

In this work we are exploring alternative battery designs and fabrication methods that have the potential for significant improvements in performance over conventional laminated cell technologies. A variety of separator-less, three-dimensional designs have recently been described¹, which due to better volumetric utilization by the active components, and reduced ion transport distances, could significantly improve energy and power density. One self-assembling method employing heterogeneous colloids will be discussed in this talk. This approach to fabricating batteries and other electrochemical devices utilizes repulsive surface forces to form electrochemical junctions between dissimilar electrochemically active materials, and employs attractive forces between like particles to form electrically conductive particle networks. The goal of this approach is the self-organization of an entire device rather than a single component, and the creation of integrated three-dimensional architecture, as illustrated in Figure 1. This technique is potentially applicable to devices of a wide range of size scales.

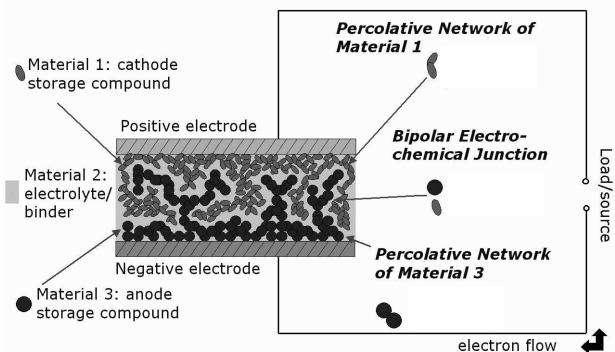


Figure 1 – Schematic diagram of a self-organizing electrochemical device based on interparticle surface forces.

Colloidal suspensions in general, including those of interest in the present work, typically have a variety of surface forces at play. Amongst these, one surface force that can be controlled to form the electrochemical junction between electronically conductive ion storage materials is the repulsive van der Waals (vdW) force. Our approach is to use measured thermodynamic and spectroscopic data to select relevant material systems for such devices. The materials selection process includes calculations using frequency-dependent optical properties to determine the sign and magnitude of the vdW interaction between a cathode and anode material separated by a polymer electrolyte solution. Atomic force microscopy (AFM) is then used to measure the interaction between single particles. Figure 2 shows characteristic repulsive behavior which we observe between anode and cathode particles measured via AFM. This is the first

instance in which repulsive vdW forces have been demonstrated between electronically conductive materials, and is expected to enable this new class of devices. In contrast, Figure 3 shows the typical attractive vdW interaction seen between like particles.

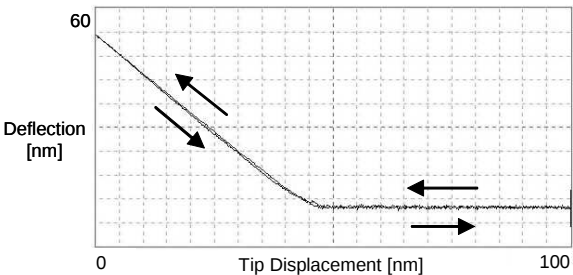


Figure 2 – Example of the AFM measured repulsive behavior between cathode and anode particles which is then used to self-organize a separator-less electrochemical junction.

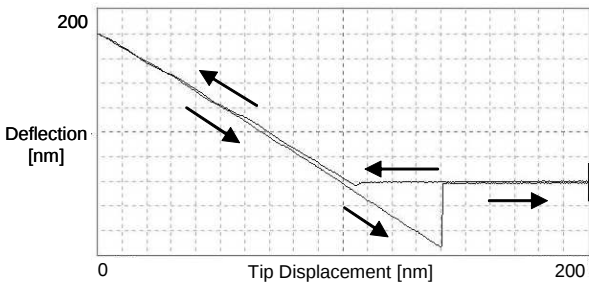


Figure 3 – Example of the characteristic snap-to and snap-off behavior of an attractive interaction between like particles measured by AFM.

Following the direct verification of surface forces tuned for these purposes, we prepare suspensions of anode and cathode particles mixed with polymer electrolyte solutions, and electrically characterize their behavior in the wet and dry states by impedance spectroscopy. The electrochemical stability of junctions formed using such surface forces has been characterized using cyclic voltammetry. Stability over the range of redox potentials relevant to rechargeable lithium ion battery systems has been demonstrated. These and other electrochemical properties of self-organized colloidal devices will be presented in the talk.

¹ Chiang *et al.*, Int. Patent App. WO 03/012908

This work is supported by U.S. AFOSR Grant No. F49620-02-1-0406