Charles Monroe

Assistant Professor,
Chemical Engineering
University of Michigan

Host: Elizabeth Podlaha-Murphy
e.podlaha-murphy@neu.edu

"Resolving the Mechanisms that Control Lithium/Oxygen Battery Performance"

Tuesday, July 15
325 Shillman Hall
11:45 a.m. – 1:00 p.m.

Refreshments will be served

ABSTRACT A viable rechargeable lithium/oxygen battery would impact transportation markets dramatically, by expediting the development of highway-capable full-electric vehicles with ranges comparable to gasoline-powered automobiles. Even conservative estimates show that Li/O_2 electrochemistry could support batteries with energy densities 4x greater than present-day lithium-ion technology. Cell compositions have been reported that select for a Li_2O_2 discharge product, which can be returned to its constituent elements during recharge. At present, however, the reaction is found to be energetically inefficient, and experimental cells operated at practical rates achieve only a small fraction of the theoretical charge capacity. This seminar will describe experimental and theoretical research our group has undertaken to elucidate design factors that control the rate capabilities and energy capacities of Li/O_2 cells. The positive electrode in a metal/oxygen battery cell comprises a liquid-permeated, electronically conductive porous solid, which poses many of the main barriers that limit its performance. During discharge, lithium ions and dissolved O_2 diffuse through the liquid phase to meet at the solid surface, where they receive an electron and precipitate the Li_2O_2 discharge product. We have performed systematic experiments to illustrate how Li/O_2 cell capacities, voltage responses, and discharge-product morphologies vary with respect to discharge current. The data suggest several possible mechanisms that could limit capacity and affect the rate capability of the system. For instance, discharge-product formation may shrink – or even block – liquid-filled pores, decrease the surface area available for charge transfer, or introduce interfacial resistances to charge (electronic) or material (ionic/molecular) exchange. The relative importance of each hypothetical performance-limiting mechanism can be illustrated by multicomponent, multiphase transport modeling. We developed a PDE-based cell model that accounts for the three distinct phases comprising the positive electrode, as well as the individual components that constitute each phase. Simulations yield discharge-voltage curves that agree both qualitatively and quantitatively with experiments, displaying a 'voltage plateau' before 'sudden death' of the cell. Examination of the oxygen, porosity and reaction distributions in the positive electrode at various discharge levels suggests that oxygen depletion due to pore clogging by the discharge product causes sudden death, limiting the accessible capacity. Based on these observations, several key characteristics of electrodes and electrolytes are revealed that can be tailored to improve the capacities and power capabilities of lithium/oxygen cells.

BIOGRAPHY Dr. Monroe has a BS in Chemical Eng from Princeton Univ He earned his PhD in Chemical Eng under John Newman at the Univ of Cal, Berkeley, studying failure mechanisms of lithium-ion batteries with the support of a research fellowship from the Shell foundation. Dr. Monroe spent 2004-07 as a Research Associate of Alexei Kornyshev in the Chemistry Dept. at Imperial College London, researching the mechanics and electrochemistry of electrified liquid/liquid interfaces. He returned to North America in 2007 as a Postdoc with Michael Eikerling at Simon Fraser University, the National Research Center Canada Institute for Fuel-Cell Innovation, and Ballard Fuel Cell Corp. He has published more than 30 peer-reviewed articles on a diverse range of topics, including fundamental work on irreversible thermodynamics, multicomponent transport analysis, electromechanics of liquid/liquid interfaces, gas kinetic theory, and electrolyte characterization, as well as more applied work on semiconductor processing, fuel-cell membranes and battery separators, electrode materials synthesis, thermal modeling of Li-ion batteries, and redox-flow-battery design. Dr. Monroe has written pedagogical papers for *Chemical Engineering Education*; he has won a teaching award from the Univ of California, as well as the Dow Teaching Prize. Since 2013, his research on redox flow batteries has been supported by the NSF through a CAREER award. Additional federal grants from the DoE, Army, and NSF, as well as industrial awards from Robert Bosch LLC, Ford Motor Co., DENSO, and Procter & Gamble currently support his group's research.