

Title: Mixed-Conductor Cascade Electrodes for High Density Energy Storage in Li_2O_2

PI Name: Adam Holewinski

CoPI Name: Se-Hee Lee

Research Objective(s): The goal of this project was development of a novel battery technology, for which we had a small amount of preliminary data in need of validation and expansion to write a competitive proposal. Batteries are inherently critical to the water-energy nexus as they represent a mechanism to store renewable energy and curtail pollution and consumption of finite resources. The battery chemistry involves converting lithium (Li) to lithium peroxide (Li_2O_2), which is extremely energy dense, but is also an electrical insulator. The project concept was to circumvent the conductivity problem by utilizing an all-solid-state battery architecture in order to permit operation under elevated temperatures, at which Li_2O_2 becomes more conductive. A major objective was to characterize whether interfacial reaction products form between Li_2O_2 and other constituent materials, as these could limit the theorized improvements.

Research Activities/Methodology: Research activities included fabrication of solid state cells with different cathode compositions, followed by characterization of the interfaces between the cathode and Li_2O_2 , as well as the between the cathode and the solid electrolyte $\text{Li}_2\text{La}_2\text{Zr}_2\text{O}_{12}$ (LLZO), which has also not been well characterized. A combination of electrochemical impedance spectroscopy (EIS), cross-section electron microscopy, and x-ray photoelectron spectroscopy (XPS) with ion milling were applied to characterize charge/discharge characteristics and to access the composition of the 'buried' solid-solid interface.

Results: We have acquired depth-profiled XPS data through entire solid state cells, before and after extended cycling, as well as after cycling at elevated temperature. We have identified a new interfacial reaction product, but interestingly this reaction does not substantially hinder performance, indicating the formation of a new 'secondary electrode interphase' (SEI) as is common in cells with liquid electrolytes. We have also collected extensive impedance spectroscopy datasets for cells throughout cycle life under numerous conditions, and fit the data to equivalent-circuit models to understand the nature of longer term degradation. This data forms the basis of ongoing proposal submissions.

Accomplishments: (proposals submitted, awarded, papers published, recognition received etc.)

- 1) Submitted pre-proposal to ARPA-E OPEN call (\$2,000,000). Declined
- 2) Submitted pre-proposal to ONR Electrochemistry program (\$450,000), Declined
- 3) Full proposal to DOE BES Materials Chemistry program (\$750,000), in progress
- 4) Manuscript in preparation
- 5) Two new collaborations (Aaron Holder CHBE, Glenn Teeter NREL).

Conclusions/Next Steps:

There still remains a great deal to learn regarding the nature of solid state interface reactions in batteries. The seed grant provided much needed resources to elevate this project to be competitive in proposals. We have gained interest from both a theoretical collaborator (Holder) and an expert in XPS spectroscopy (Teeter), who will be participating in a BES proposal in progress. This proposal will be submitted spring 2019, and we will continue to pursue DOD and DOE opportunities as they become available. The first manuscript on this work will also be submitted shortly.