

MAR15-2014-020303

Abstract for an Invited Paper
for the MAR15 Meeting of
the American Physical Society

In-situ Studies of Structures and Processes at Model Battery Electrode/Electrolyte Interfaces¹

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The ability to understand and control materials properties within electrochemical energy storage systems is a significant scientific and technical challenge. This is due, at least in part, to the extreme conditions present within these systems, and the significant structural and chemical changes that take place as lithium ions are incorporated in the active electrode material. In particular, the behavior of interfaces in such systems is poorly understood, notably the solid-liquid interface that separates the electrode and the liquid electrolyte. I will review our recent work in which we seek to isolate and understand the role of interfacial reactivity in these systems through in-situ, real-time, observations of electrochemically driven lithiation/delithiation reactions. This is achieved by observing well-defined model electrode-electrolyte interfaces using X-ray reflectivity. These results reveal novel understandings of interfacial reactivity in conversion reactions (e.g., Si, Si_xCr, Ge, NiO) that can be used to control the complex reaction lithiation pathway through the use of thin-film and multilayer electrode structures.

¹This work was supported by the Center for Electrochemical Energy Science, an Energy Frontier Research Center funded by the U.S. Department of Energy, Office of Science, in collaboration with T. Fister, A. Gewirth, M.J. Bedzyk and others.