

Society of Engineering Science 51st Annual Technical Meeting

1–3 October 2014

Purdue University, West Lafayette, Indiana, USA

Modulus variation of composite graphite electrodes in lithium-ion batteries during electrochemical cycling

Jones, Elizabeth M. C., ecler2@illinois.edu; Tavassol, Hadi; Gewirth, Andrew A.; White, Scott R.; Sottos, Nancy R., University of Illinois at Urbana-Champaign, United States

ABSTRACT

Graphite is currently the most common anode material used in commercial lithium-ion batteries. During battery charging and discharging processes, lithium ions intercalate into and deintercalate from graphite, forming several distinct stages of graphite-lithium intercalation compounds (G-LICs). Each stage of G-LIC has a unique spacing between graphene layers, with the spacing increasing for increasing lithium content. In graphite-based composite electrodes (graphite particles in a porous polymer matrix), the changing layer spacing leads to stress and strain evolution on the composite length scale. In two separate experiments, we use substrate-curvature measurements to monitor stress changes in a thin electrode constrained on an inert, rigid substrate, and we use digital image correlation to track strain changes in a free-standing, unconstrained electrode. Combining the in-situ stress and strain analyses enables us to extract the change in the apparent modulus of the composite graphite electrode as a function of electrode potential and lithium content. As expected, we found that constrained electrodes develop compressive stress during lithiation (~10 MPa) and that unconstrained electrodes undergo free expansion (~1.5% linear strain). Interestingly, the apparent modulus of the electrode increases the most significantly during the formation of the dilute stage I compound, increases slightly with the formation of the stage IV, dilute stage II, and stage II compounds, and then decreases with the formation of the stage I compound (LiC₆). During delithiation, unconstrained electrodes contract, recovering nearly their original size. In constrained electrodes during delithiation, however, the compressive stress is first relaxed, and then a tensile stress develops and is subsequently relaxed. The tensile stress leads to an apparent softening of the composite electrode over a broad range of electrode potential and capacity. At the end of one complete lithiation/delithiation cycle, the apparent modulus returns to approximately its original value. The evolution of stress, strain, and modulus data provides quantitative information on the coupled electro-chemo-mechanical response of battery electrodes and insight on material strategies to increase battery reliability.