Integrating State of Charge (SOC) Dependent Material Properties into Li-ion Battery Failure Modeling

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ABSTRACT

During battery operation, Li flows into and out of electrode particles, causing microstructural changes and deformation-induced degradation. A variety of models have been proposed to interpret these mechanical and microstructural changes, but they lack of proper input materials' properties and direct experimental supports. To address this challenge, we developed "atomically informed" battery mechanics models by integrating density functional theory (DFT) predicted mechanical properties into continuum models to predict diffusion induced deformation and stress evolution and make direct comparisons with experiments.

Based on DFT calculations, we found many electrode materials change their elastic properties, fracture energies, interfacial properties, and bonding natures upon lithiation. For example, in graphite electrodes, we predicted the modulus was tripled due to lithiation. The *in-situ* lithiation and strain maps displayed both dilation and contraction during lithiation in a commercial graphite electrode. Based on our model, the unexpected contraction during lithiation is explained by the stiffening of graphite upon lithiation. In contrast, lithiation induced softening in Si was predicted. In order to design mechanically robust Si-C electrode, interfacial properties became critical. Both unlithiated and lithiated Si/C and Si/Cu interfaces were studied. The native adhesion between Si and sp2 carbon layers was predicted to be inherently weak; however stronger Si/C interface can be achieved by functioning carbon nanotube (CNT) surface with carboxylic functional groups. A novel beaded-string structure comprising amorphous Si beads threaded on CNT with controlled Si/C interfacial chemistry was synthesized. The enhanced interface played key roles in changing the lithiation morphology and mitigating stress generation, thus enabling the observed non-cracking of the beaded-string structure during lithiation/delithiation cycles.

All these results underscore the importance of integrating the SOC dependent material properties into the structural design of mechanically robust electrodes for Li-ion batteries.