

Mechanical Engineering Doctoral Defense

Understanding the Mechanical Behaviors of Lithium-Based Battery Anodes—Silicon and Lithium Metal

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abstract

Despite many existing studies on silicon (Si) anodes for lithium ion batteries (LIBs), many essential questions still exist on compound formation, composition, and properties. Here it is shown that some previously accepted findings do not truthfully reflect the actual lithiation mechanisms in realistic battery configurations. Furthermore the correlation between structure and mechanical properties in these materials has not been properly established. Here a rigorous and thorough study is reported to comprehensively understand the electrochemical reaction mechanisms of amorphous-Si (α -Si) in a realistic LIB configuration. In-depth microstructural characterization was performed and correlations were established between Li-Si composition, volumetric expansion, and modulus/hardness. It is found that the lithiation process of α -Si in a real battery setup is a single-phase reaction rather than the accepted two-phase reaction obtained from in-situ TEM experiments. The findings in this paper establish a reference to quantitatively explain many key metrics for lithiated α -Si as anodes in real LIBs, and can be used to rationally design α -Si based high-performance LIBs guided by high-fidelity modeling and simulations. On the other hand, problems related to dendrite growth on lithium metal anodes such as capacity loss and short circuit present major barriers to the next-generation high-energy-density batteries. The development of successful mitigation strategies is impeded by the incomplete understanding of the Li dendrite growth mechanisms. Here the enabling role of plating residual stress in dendrite initiation through novel experiments of Li electrodeposition on soft substrates is confirmed, and the observations is explained with a stress-driven dendrite growth model. Dendrite growth is mitigated on such soft substrates through surface-wrinkling-induced stress relaxation in deposited Li film. It is demonstrated that this new dendrite mitigation mechanism can be utilized synergistically with other existing approaches in the form of three-dimensional (3D) soft scaffolds for Li plating, which achieves superior coulombic efficiency over conventional hard copper current collectors under large current density.

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