
On particle cracking in Li-ion Si-based battery electrodes

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Abstract

Silicon is recognized as a promising material for next-generation lithium-ion batteries, offering a storage capacity approximately 10 times higher than that of conventional carbon-based electrodes. Despite extensive research and industrial development, the practical use of silicon-based electrodes remains constrained by a significant loss of capacity. This issue is primarily attributed to microscopic cracks caused by substantial swelling of Si particles during cycling, which places an extremely high burden on the mechanical stability and structural integrity of the electrode (1). Mechanical stresses, induced by chemical reactions as lithium ions diffuse into silicon particles during cycling, nucleate and drive cracks that hinder further ion transport, ultimately leading to capacity loss. The lithiation of silicon is a complex electro-chemo-mechanical process that is not yet fully understood.

Our high-fidelity computational multiphysics in-house models incorporate key structural features across various length and time scales while capturing some of the dominant physical phenomena that dictate battery efficiency. The simulation results are compared with findings reported in the literature, including observations of deforming silicon particles in real Si-based battery cells under cycling (e.g. in-situ synchrotron X-ray CT experiments (2)). While the striking agreement between simulations and experiments is promising, it also highlights the need for a deeper understanding of the lithium-ion diffusion process in silicon, particularly the multiple material phase changes that occur between lithium and silicon, Li_xSi_y .

(1) He, Huang, Wang, Mizota, Liu, Hou (2021). *Energy Fuels* 35 944-64

(2) Taiwo, Paz-García, Hall, Heenan, Finegan, Mokso, Villanueva-Pérez, Patera, Brett, Shearing (2017). *Journal of Power Sources* 342 904-912

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