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Revealing Solid Electrolyte Interphase Formation Through Interface-Sensitive Operando X-ray Absorption Spectroscopy

Content

The solid electrolyte interphase (SEI) that forms on Li-ion battery anodes is critical to their long-term performance, however observing SEI formation processes at the buried electrode-electrolyte interface is a significant challenge. Here we show that operando soft X-ray absorption spectroscopy in total electron yield mode can resolve the chemical evolution of the SEI during electrochemical formation in a Li-ion cell, with nm-scale interface sensitivity [1]. O, F, and Si K-edge spectra, acquired as a function of potential, reveal when key reactions occur on high-capacity amorphous Si anodes cycled with and without fluoroethylene carbonate (FEC). The sequential formation of inorganic (LiF) and organic (-(C=O)O-) components is thereby revealed, and results in layering of the SEI. The addition of FEC leads to SEI formation at higher potentials which is implicated in the rapid healing of SEI defects and the improved cycling performance observed. Operando TEY-XAS offers new insights into the formation mechanisms of electrode-electrolyte interphases and their stability for a wide variety of electrode materials and electrolyte formulations.

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