Data-driven Modeling of Lithium Intercalation Materials

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Technical Summary

For multi-scale electrochemical systems such as batteries, a number of tools including microscopy, diffraction, spectroscopy, and impedance exist to probe and measure from single active particles to the whole cell, but traditional modeling approaches fail to capture all the available information. With the arrival of high-throughput computation and experimentation, there is an unprecedented oppor-tunity to solve key challenges in energy storage via data-driven methods. In this thesis, I developed the framework of learning the physics and extracting quantitative models of lithium intercalation materials from experimental data. Built upon the theoretical foundation of the reaction kinetics, transport phenomenon, thermodynamics, and electrochemistry of lithium intercalation materials, the theory describes the single-particle behavior and the population dynamics in a porous electrode. PDE-constrained optimization and Bayesian inference is used to infer and quantify the uncertainty of the constitutive laws from multiple data streams. Applications include learning the constitutive laws from images of pattern formation, inverting the unknown free energy, reaction kinetics, spatial heterogeneity, and chemo-mechanical coupling from images of lithium iron phosphate particles, and quantifying the autocatalytic reaction kinetics from X-ray diffraction of Li layered oxides. The results show the possibility to achieve full utilization of the datasets with quantitative agreement, enable model selection and validation, and advance the modeling of batteries.

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