

TRANSPORT IN BATTERY ELECTRODES: SCALING BEHAVIOR ASSESSED BASED ON REAL AND SIMULATED MICROSTRUCTURES

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ABSTRACT Electrochemical energy conversion and storage devices, including batteries and fuel cells, are multiscale functional material systems. Within these systems mesoscale electrode structures play a key role in the delivery of the charged and neutral species that support reactions at electrochemically active interfaces. The further development of batteries for next generation energy storage systems relies upon the deterministic design of such mesoscale electrode structures. Strengthening the understanding of interactions between complex electrode structure and material performance is instrumental in more deterministic electrode design. This goal may be achieved by development of scaling relations that describe the diffusion response within electrode active materials subject to surface electrochemical reactions. The analysis of diffusive transport in lithium ion battery electrode active materials is presented using electrode data at multiple length scales. Electrode microstructures imaged using x-ray nanotomography and microtomography serve as the primary computational domains for 3D and 2D finite element models. Lithium diffusion within these real electrode structures is compared to the behavior of simulated structures. First, the 3D analysis of Li diffusion in cathode active material particles is presented based on x-ray nanotomography and microtomography data. Samples extracted from pristine cathodes were imaged using x-ray nanotomography and microtomography, yielding a multiscale dataset of electrode geometry with clear contrast between the active material and the carbon/binder/pore regions within the samples. Particle geometry for these samples is characterized based on the particle size distribution, surface area, volume, and sphericity. Tomographic data is then applied as a computational domain in a numerical transport model to assess the effect of particle geometry on intercalation behavior. This assessment provides a means of determining a particle size for defining dimensionless parameters describing concentration gradients and intercalation time. Mass transfer Biot and Fourier numbers are calculated for active material particles. The former provides a basis for understanding the severity of concentration gradients that induce stress during intercalation while the latter provides a means of estimating intercalation time. The deviation from spherical particle approximations is addressed through comparison to spherical particles of comparable Biot number. Following the discussion of diffusion scaling behavior, the role of surface complexity is addressed based on 2D diffusion models. Diffusion behavior in real particle cross-sections is compared to diffusion in microstructure simulated using prefractal curves. Real particle structures exhibit surface structure and scaling behavior comparable to these prefractal geometries. This behavior suggests that for certain active material and electrolyte combinations complex surface structures may limit active surface access and attendant electrode performance. Together these efforts demonstrate the development of scaling relations that can enable deterministic design of battery electrodes across multiple scales.