Simulation of hierarchical ordered porous microstructure electrode for high energy density Li-ion batteries

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High specific energy is always desirable in battery systems, especially for electric vehicles. A key towards improving specific energy for a particular combination of anode and cathode active material is to maximize the mass fraction of active material, which means minimizing the fraction of inactive material such as the separator, the binder, electrode additives for improving electronic conductivity, and the current collector. A seemingly simple strategy for maximizing the mass fraction of active materials is to make the electrodes as thick as possible. However, ion transport in thick electrodes can be particularly problematic since salt diffusion into and through the electrode pore space is slow and can easily limit the battery charge / discharge rate. An ideal microstructure is one that allows for direct line of sight or path for the electrode active material access to the electrolyte, for rapid ion-transport throughout the entire electrode thickness. Therefore, a need exists for electrode fabrication methods that create microstructures that maximize electrolyte transport, as is needed to achieve high-rate charging/discharging in electrodes having high mass loading. Optimal microstructures in electrodes help to achieve the ideal balance of active material and electrolyte mass to give high gravimetric and volumetric energy density.

To design a microstructure with the goal of Li ion salt transport through thick electrodes, we synthesized ordered porous microstructure electrodes using combination of freeze casting and tape casting techniques. This method has the advantage of the combination of conventional tape casting, one of the main technique in manufacturing Li ion electrodes in industrial scale, and freeze casting to make anisotropic porous microstructures. This study simulates, using COMSOL Multiphysics® software, two dimensional ordered porous electrode cross-sections and presents the electrochemical performance results. The cell consists of Mo-doped LTO as the cathode, lithium metal anode, polymer separator, and liquid ethylene carbonate and ethyl methyl carbonate electrolyte. The electrochemical performance of ordered porous microstructure is investigated. We show that these engineered porous electrodes considerably reduces the electrolytes mass transport limitation and electrochemical response of electrode is independent of tortuosity, making it possible to synthesize thick electrode for high energy and power density applications. Simulation results show that for freeze cast Mo-doped LTO cathode (electronic conductivity of 1 S/m) with 10 micron wall and pore diameter, tortuosity in the wall as high as 10, electrode thickness of 458 micron and mass loading of 30 mg/cm^2 can deliver full capacity even at 2 C-rate.