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Electrochemical impedance spectroscopy modeling for numerically reconstructed Li-Ion battery graphite anodes

Bereket Tsegai Habte
Guangzhou Institute of Energy Conversion, China

Carbon graphite has received much attention over the last decades as negative Li-ion battery electrode due to its thermal stability and optimal cycling capability. This paper aims to give a comprehensive account of the effect of microstructure morphology such as porosity, tortuosity, solid-electrolyte interfacial area and active material particle size and shape on the underlying species transport and interfacial reaction kinetics during battery cycling. Simulated annealing method (SAM) was used to generate the graphite anisotropic microstructures based on physical and statistical information gained by the scanning electron microscope (SEM) imaging technique of a real graphite anode. Equivalent circuit model composed of resistance, capacitance and Warburg impedance were applied to model the electrochemical impedance spectroscopy (EIS) of the underlying electrochemical activities during charge/discharge cycling over a wide range of frequency spectra (*1mHz – 20kHz*). Results clearly indicate that the active material particle shape and size dictate the anisotropic behavior of the microstructure and the total surface area of the electrode subjected to the electrolyte is the main factor that influences the charge transfer rate. Moreover, the porosity and tortuosity parameters of the microstructures affect directly the ionic diffusion inside the electrolyte. Nyquist plot reveals the effect of microstructure morphology on the impedance of the ionic and electronic migration in the electrolyte and diffusion in solid electrode. The semi-circle in the high frequency region is associated with the charge transfer resistance and the dielectric behavior of the solid-electrolyte interface (SEI) while the 45° slope at the low frequency region is a result of solid-state diffusion of lithium into electrode

berekett6@gmail.com