

Integration of a Kinetic Monte Carlo Algorithm with Continuum Models to Examine Capacity Fade

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Rechargeable lithium-ion batteries have been extensively used in mobile communication and portable instruments due to its high volumetric and gravimetric energy density and low self-discharge rate. Any scientists and engineers to work towards developing lithium-ion batteries with improved performance and longer life.

In normal operation an active solid-electrolyte-interface (SEI) layer forms mainly in the first cycle of charging. The active SEI layer provides a barrier between the electrolyte and electrode while allowing lithium ions to permeate through the SEI layer in order to intercalate into and out of the bulk solid to store and release energy. Repeated cycling can lead to the formation of unwanted byproducts on the SEI layer, forming the passive SEI layer. This prevents diffusion of the lithium ions to the bulk active material, increasing resistance and reducing the efficiency of the battery. This phenomenon produces high temperature and can result in thermal runaway [1-3] within the battery. The properties and chemical composition of the SEI layer (active and passive) has been a subject of intense research due to its importance in the safety, capacity fade, and cycle life of Li-ion secondary batteries [4-10].

Examination of the growth of the SEI layer as a mechanism for capacity fade has been performed using Kinetic Monte Carlo (KMC) simulations [11]. This has provided valuable insight into the growth of the SEI layer and has shown the effects of high rates of charging on SEI layer passivation. These KMC simulations can be used to study the surface heterogeneity of the electrode surface as the passive layer is formed.

KMC simulations will be closely coupled with the pseudo 2D model in a way not done previously. The P2D model will be converted from a system of partial differential equations to a system of ordinary differential equations by using reformulation techniques developed for the continuum model [12].

The KMC model studies the events that occur on the particle surface. For example, lithium-ion adsorption and passivation are determined using KMC approach by considering the relative rate constants of each event and using a random number generator to determine the event which occurs. In contrast, the continuum P2D model explains what happens in the bulk phases, and does not use a random factor.

This coupled model will perform continuum calculations simultaneously with the KMC calculations. At each time step, the KMC simulation will calculate the surface coverage of lithium (passive and active) as well as the lithium ion flux, while the continuum equations will account for diffusion in the bulk phases.

As passivation is considered in the KMC model, the capacity of the battery will fade with time. This is due to the increased resistance preventing lithium-ion intercalation. Furthermore, since the formation of the passive layer involves the reaction of lithium in the electrolyte, the concentration of available lithium will

decrease, further reducing available capacity. This will lead to better understanding of capacity fade and better operation of lithium-ion batteries.

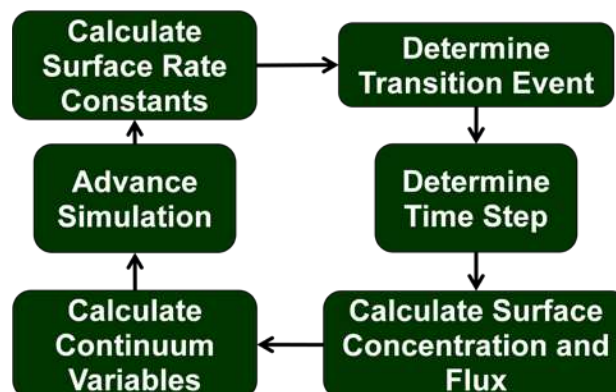


Figure 1: Algorithm for the coupled model for charging

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