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A theory and a simulation capability for the growth of a solid electrolyte interphase layer at an anode particle in a Li-ion battery



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ABSTRACT

A major mechanism for electrochemical aging of Li-ion batteries is the growth of a solid electrolyte interphase (SEI) layer on the surface of anode particles, which leads to capacity fade and also results in a rise in cell resistance. We have formulated a continuum theory for the growth of an SEI layer—a theory which accounts for the generation of the attendant growth stresses. The theory has been numerically implemented in a finite-element program. This simulation capability for SEI growth is coupled with our previously published chemo-mechanical simulation capability for intercalation of Li-ions in electrode particles. Using this new combined capability we have simulated the formation and growth of an SEI layer during cyclic lithiation and delithiation of an anode particle, and predicted the evolution of the growth stresses in the SEI layer. The evolution of the stress state within the SEI layer and at the SEI/anode-particle interface for spherical- and spheroidal-shaped graphite particles is studied. This knowledge of the local interfacial stresses provides a good estimate for the propensity of potential delamination of an SEI layer from an anode particle.

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1. Introduction

Li-ion batteries are slated to play an increasingly prominent role in large-scale stationary energy storage systems in renewable energy plants, as well as in power systems for sustainable electric transportation vehicles. In these applications a battery must tolerate large numbers of cycles at high rates of charge and discharge, and continue to serve its function without any degradation in performance. Much remains to be done to improve upon the performance of the current generation of Li-ion batteries to meet the desired performance levels. One of the major concerns is the mitigation of “aging” of batteries—which refers to electro-chemo-mechanical degradation, capacity fade, and power loss of the batteries.

Amongst the many mechanisms which cause aging in Li-ion batteries, a major mechanism is the formation, due to decomposition of the electrolyte, of a thin *solid electrolyte interphase* (SEI) layer on the surface of anode particles. The SEI layer is electronically insulating which prevents further decomposition of the electrolyte, but it is ionically conductive and allows the diffusion of Li ions through it. However, the formation of SEI consumes Li-ions, which competes with the desired amount of Li for intercalation in the active anode material, and this causes capacity fade. In addition, since Li-ions must

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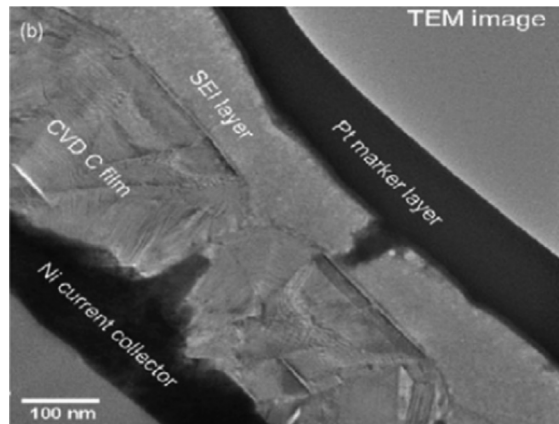


Fig. 1. TEM micrograph of a SEI layer on a graphite anode. From Mukhopadhyay et al. (2012).

diffuse through the SEI layer, the formation of a SEI layer is accompanied by an increase of cell resistance. Often, the cyclic volume changes of an anode particle during Li-ion intercalation and deintercalation can cause the SEI layer to crack, delaminate, and spall from the surface of the anode particle. New SEI is then formed on the freshly exposed particle surface, which consumes more Li-ions and causes additional capacity fade. Both the chemical and mechanical integrity of the SEI are critical to the safety and performance of Li-ion batteries (cf., e.g., Lee et al., 2007; Verma et al., 2010; Barré et al., 2013).

The purpose of this paper is to report on a new theory and a finite-element-based capability for the simulation of growth of a solid electrolyte interphase layer at an anode particle in a Li-ion battery. The theory attempts to account for the generation of stress due to the growth of a SEI layer, as well as the stress that arises due to the lithiation and delithiation of the anode particle. In the literature, the stress generated due to the growth of the SEI layer is often called “irreversible” while the stress related to the swelling/de-swelling of the anode particles during lithiation/delithiation is called “reversible.”

In a recent novel paper, Mukhopadhyay et al. (2012) presented experimental measurements of the reversible and irreversible stresses during SEI formation on a thin-film graphite anode. In their experiments they used a 250 μm thick, 1 in diameter quartz substrate on which, using chemical vapor deposition (CVD) techniques, they deposited (i) a 15 nm thick Ti layer; (ii) a 200 nm thick Ni layer; and finally (iii) a 200 nm layer of c-axis oriented graphite. The Ti and Ni were deposited to act as a catalyst for graphitization and as a current collector. This multilayered plate was then assembled into an electrochemical cell which was charged and discharged under galvanostatic conditions against a Li-metal cathode to develop a SEI layer on the graphite anode. The TEM micrograph in Fig. 1, taken from Mukhopadhyay et al. (2012), shows a ≈ 100 nm – thick SEI layer which has formed on the graphite (CVD C) anode after 50 cycles of charging and discharging. After the initial deposition of the graphite layer, as well as during electrochemical cycling during which the SEI layer was formed, Mukhopadhyay et al. measured the curvature of the plate using an array of parallel laser beams focused on the back side of the quartz substrate. The results from their experiments are schematically shown in Fig. 2:

- Fig. 2(a) shows the undeformed quartz substrate.
- After CVD deposition of the graphite layer at 1000 $^{\circ}\text{C}$ and cool-down to room temperature, the graphite contracts more than the substrate and this results in a positive curvature, Fig. 2(b).
- At the end of the first half-cycle of charging the authors observed a reduction in the positive curvature of the plate; cf. Fig. 2(c) relative to Fig. 2(b). This reduction in curvature occurs due to the expansion of the graphite upon lithiation, and

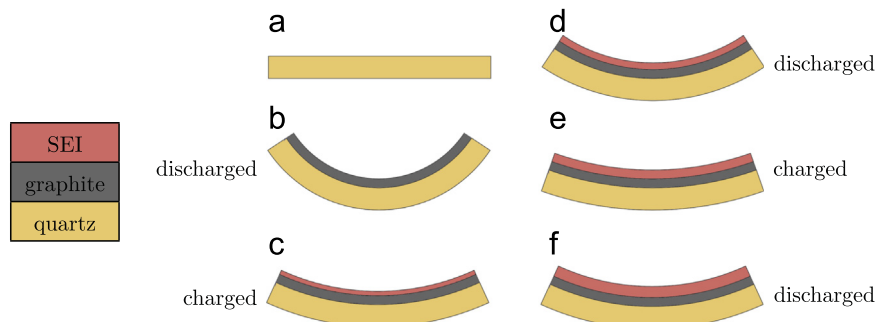


Fig. 2. A schematic of the observed plate-curvature changes in the experiments of Mukhopadhyay et al. (2012): (a) the initially straight plate; (b) the curved plate after carbon deposition; (c) the curved plate, with the charged graphite at the end of the first half-cycle; (d) the curved plate at the end of the first charging/discharging cycle; (e) the curved plate, charged graphite after one and a half cycles; and (f) the curved plate, discharged at the end of two cycles.

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