

Physics-Based Modeling Of Lithium Plating On Graphite Anode Of Commercial Lithium-Ion Batteries

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Abstract

Lithium plating on the graphite anode is a significant factor contributing to the degradation of cell capacity, initiation of internal short circuits, and escalation of thermal runaway in lithium-ion batteries. Non-intrusive detection methods for lithium plating are critical for the safe and reliable operation of lithium-ion batteries. This study presents a physics-based pseudo-two-dimensional (P2D) model that incorporates lithium plating and stripping reactions to describe the electrochemical behavior of commercial 18650 cylindrical cells with graphite and LiFePO₄ (LFP) electrodes at high current rates and low temperatures. Simulations were performed using COMSOL MULTIPHYSICS 6.1, and the results were compared with experimental measurements obtained from a Neware CT-4000 series battery testing system. The voltage response and surface temperature of 48 commercial 18650 LFP cells at varying states of health (100% to 75% at an approximately 5% increment) and charge (100% to 0% at an approximately 5% increment) were collected. These results can inform the establishment of operational and design limits to mitigate capacity degradation and safety hazards inherent in these cells.

Figures used in the abstract

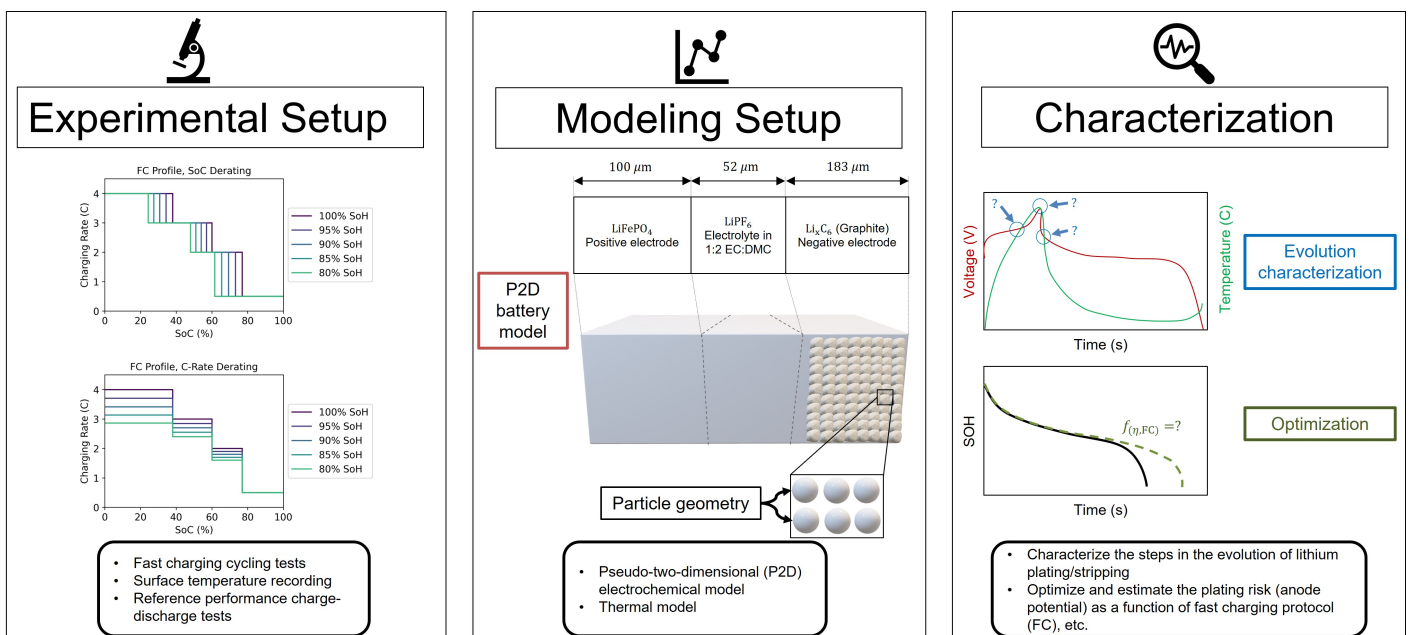


Figure 1 : Graphical abstract

Process	Reaction	Current (A/m ²)	Kinetics Equations
Intercalation	$\text{Li}_x\text{C}_6 + \Delta x \text{Li}^+ + \text{e}^- \rightarrow \text{Li}_{x+\Delta x}\text{C}_6$	j_1	$j_1 = i_{0,1} \left[\exp\left(\frac{\alpha_{a,1} F \eta_1}{RT}\right) - \exp\left(\frac{\alpha_{c,1} F \eta_1}{RT}\right) \right]$ <ul style="list-style-type: none"> $i_{0,1} = k_1 \cdot c_e^{\alpha_{a,1}} (c_{s,\max} - c_{s,\text{surf}})^{\alpha_{a,1}} c_{s,\text{surf}}^{\alpha_{c,1}}$ $\eta_1 = \phi_s - \phi_e - U - FjR_{\text{SEI}}$ $U = U_{\text{ref}}(x) - (T - T_{\text{ref}}) \left(\frac{dU}{dT}\right)$ $R_{\text{SEI}} = (\delta_{\text{SEI},0} + \Delta\delta_{\text{SEI}}) / \sigma_{\text{SEI}}$
Plating	$\text{Li}^+ + \text{e}^- \rightarrow z_1 \text{Li}_{\text{rev}}^{(s)} + z_2 \text{Li}_{\text{dead}}^{(s)} + z_3 \text{SEI}$	j_2	$j_2 = i_{0,2} \left[\exp\left(\frac{\alpha_{a,2} F \eta_{\text{Li}}}{RT}\right) - \exp\left(\frac{\alpha_{c,2} F \eta_{\text{Li}}}{RT}\right) \right], \eta_{\text{Li}} < 0$ <ul style="list-style-type: none"> $i_{0,2} = k_2 \cdot c_e^{\alpha_{a,2}}$ $\eta_{\text{Li}} = \phi_s - \phi_e - U_{\text{Li}} - FjR_{\text{SEI}}$
Stripping	$\text{Li}_{\text{rev}}^{(s)} \rightarrow \text{Li}^+ + \text{e}^-$	j_3	$j_3 = i_{0,2} \left[\exp\left(\frac{\alpha_{a,2} F \eta_{\text{Li}}}{RT}\right) - \exp\left(\frac{\alpha_{c,2} F \eta_{\text{Li}}}{RT}\right) \right] \cdot \left(\frac{\beta \cdot n_{\text{Li,rev}}}{1 + \beta \cdot n_{\text{Li,rev}}} \right),$ $\eta_{\text{Li}} > 0, n_{\text{Li,rev}} > 0$

Figure 2 : Modeling overview for the kinetics of the main reactions

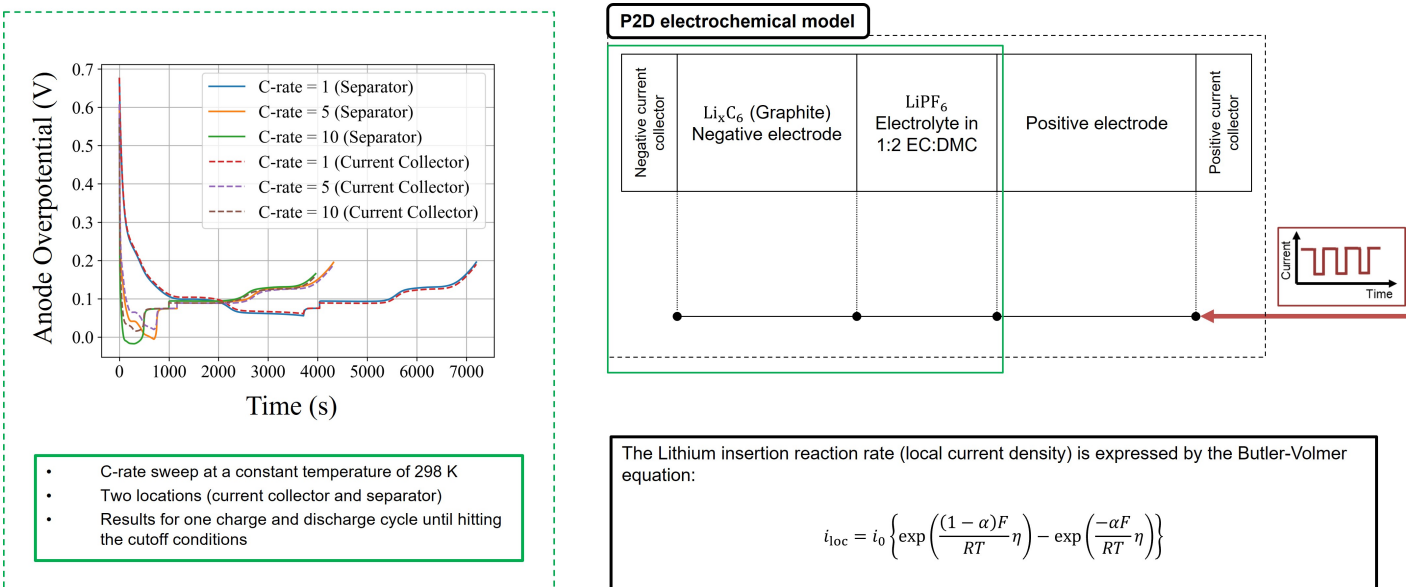


Figure 3 : Preliminary results - graphite anode overpotential

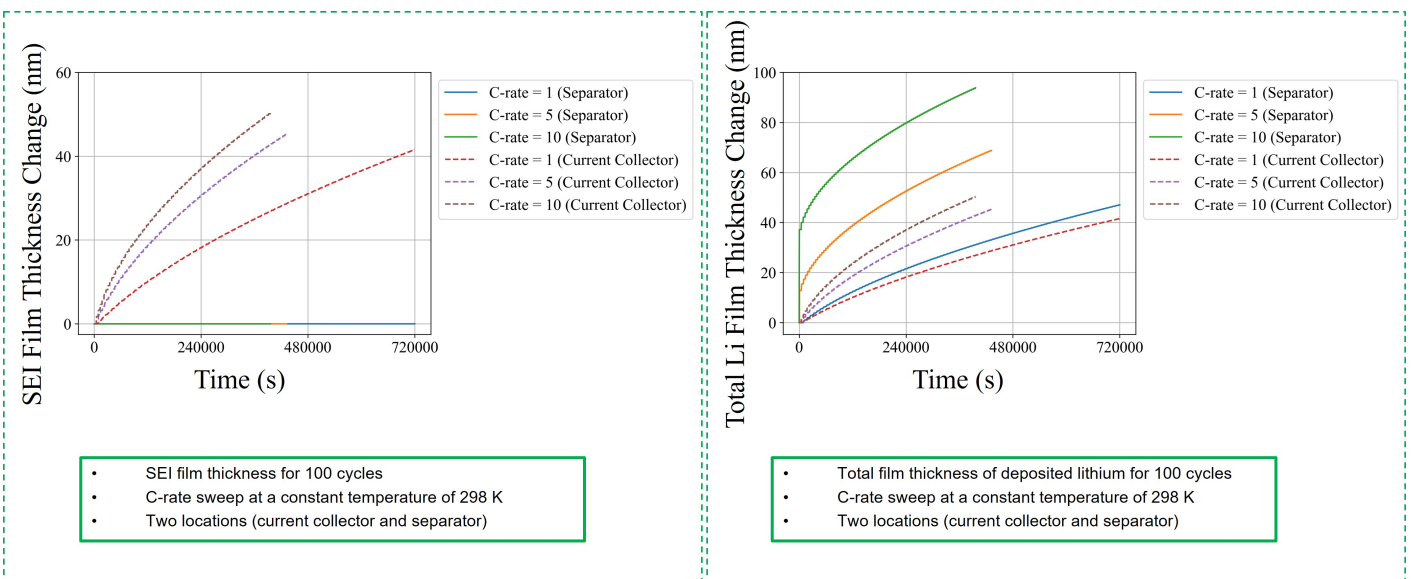


Figure 4 : Preliminary results - deposited lithium film thickness

