Zero Dimensional Models for SEI Formation in Lithium-Ion Battery

Parametric Study to Compare Formation Cycle with Subsequent Cycles

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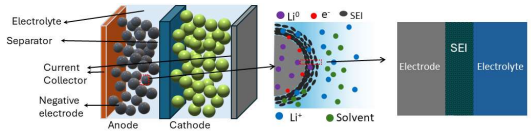
Abstract

The solid electrolyte interphase (SEI) is a critical yet poorly understood component in Li-ion batteries (LIBs). Zero-dimensional models (ZDMs) are a common physics-based approach to modelling SEI growth while remaining computationally simple, and various models have been proposed in the literature[1-4]. However, to the best of our knowledge, none of them has been successfully applied and validated to describe SEI growth during the formation cycle, the very first cycle after the battery is assembled, where the capacity fade is much larger than in the subsequent cycles.

To reduce the complexity of modeling the SEI film, a simplified approach is adopted by considering a thin segment of the SEI along the direction perpendicular to the active material surface of the electrode particle, as illustrated in the schematic below.



Motivation

Motivationz

- Understanding the initial formation of the **Solid Electrolyte Interphase (SEI)** is crucial as it significantly impacts **cell performance** and **degradation** processes.

Research Question

- How do **ZDMs** of the **SEI** film behaviour differ between the formation cycle and subsequent cycles in LIBs?.

Research Aims

- Identifying the critical parameters that influence the formation cycle according to common ZDMs.

Theory

Assuming the SEI layer is the only source of capacity loss:

$$J_{\text{total}} = J_{\text{int}} + J_{\text{SEI}},$$

where J_{int} is the intercalation current density, usually described by Butler-Volmer equation and J_{SEI} represents the SEI current density, which can take different forms base on the rate-limiting mechanism. **Common rate-limiting mechanisms** for J_{SEI} include[1-4]:

1. Electron-Migration Limited (E-ML)
2. Interstitial-Diffusion limited (I-DL)
3. Solvent-Diffusion Limited (S-DL)
4. EC Reaction Limited (EC-RL)
5. Reaction Limited (RL)

Numerical Results

The Doyle-Fuller-Newman (DFN) model was implemented in conjunction with all five ZDMs using four C-rates for charging and discharging in PyBaMM[5]. More details about used parameter sets and model setup can be found at(https://github.com/brosaplanella/formation).

1. Impact of C-rate on SEI growth at formation cycle and subsequent cycles:

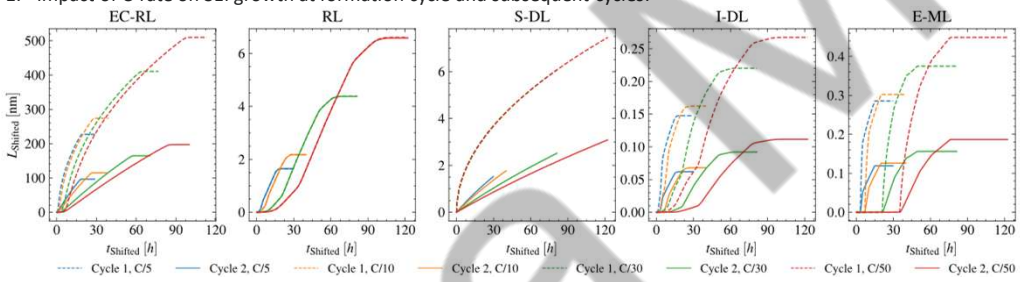


Figure 1: Growth of SEI thickness.

2. How capacity fade charge with State of Charge (SOC) at formation cycle and subsequent cycles:

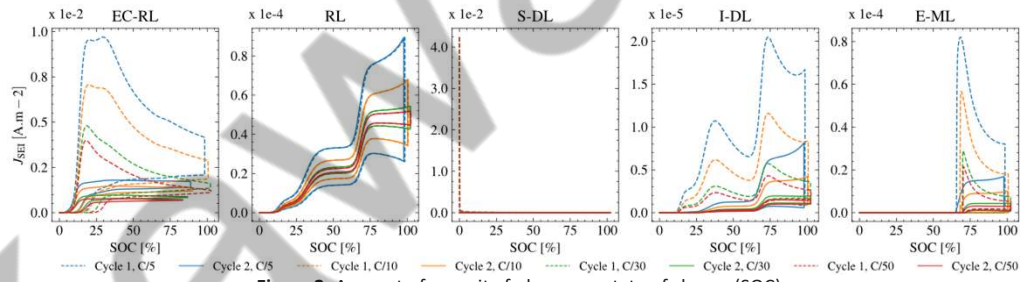


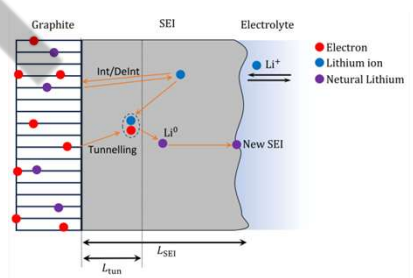
Figure 2: Amount of capacity fade versus state of charge (SOC)

Discussion and Conclusions

- In this work, we compare the behaviour of the most common SEI growth models in the literature for the formation cycle and the subsequent ones .
- SEI growth increases with C-rate at any instant[6] in all ZDMs except S-DL, which surprisingly shows SEI growth is C-rate independent in the formation cycle. However, total SEI formed per cycle trends inversely to C-rate, with higher SEI growth in cycle 1 than later cycles across all ZDMs (Figure 1).
- Maximum capacity fade varies by SOC, but not C-rate. Fade fluctuation along SOC depends on the model (Figure 2).
- This preliminary work provides direction to gain deeper insight into complex SEI formation dynamics.

Impact / Next steps

- Developing a Continuum Model to Capture SEI Growth Including Electron Tunnelling Phenomena



References

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Researcher bio

Dr. Kawa is a Faraday Institution Research Fellow at the University of Warwick, UK since June 2023. He completed his PhD in Applied Mathematics in 2015 at the University of Birmingham, UK, and has several years of research experience in numerical fluid dynamics in academia. His current research focuses on developing physics-based mathematical models of batteries, specifically the SEI layer, to investigate how it initially forms and grows over time. He utilizes the open-source PyBaMM software for battery modeling in his research

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