Interpretable Approaches for Optimizing the Pulse Diagnostics and Formation for Lithium-ion Batteries

by

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Submitted to the Department of Chemical Engineering
on December 4, 2025 in partial fulfillment of the requirements for the degree of

DOCTOR OF PHILOSOPHY IN CHEMICAL ENGINEERING

Lithium-ion Batteries (LiBs) are widely used in electronic devices and energy storage systems owing to their high energy density, long lifespan, and low cost. To further improve their performance and safety, we focus on optimizing two necessary yet time-consuming LiB applications: pulse diagnostics that enable degradation mechanism-level diagnosis, and the formation step that can greatly impact the battery performance.

Interpretability plays a key role in the optimization scheme. Interpretability allows us to break down result-oriented, time-consuming objective functions into tractable problems, enhances the extrapolation capability of the optimization solver, and determines the depth of physical insights we gain from the optimization scheme. Thus, we aim for interpretable approaches for optimizing the pulse diagnostics and formation step.

The first part of this thesis demonstrates the optimization of voltage pulse diagnostics using the fitness model as the source of interpretability. Fitness model is a first-principles model that describes the behavior of degraded LiBs under voltage pulses. Five degradation parameters ($R_{\mathrm{f,c}}$, $R_{\mathrm{f,a}}$, $\tilde{c}_{\mathrm{c}}$, $\tilde{c}_{\mathrm{a}}$, and c_{+}) are identified from pairs of voltage pulse settings and current response measurements. Two objective functions for this task are the practical identifiability of the degradation parameters and the total diagnostic time. Since these objectives can be obtained after the diagnostic protocol is performed, the optimization problem is less tractable. Model-based Design of Experiments (DoE) is used to convert the inverse problem into a forward problem, enabling estimation of the practical identifiability of five degradation parameters beforehand. Scaling analysis is used to estimate the total diagnostic time of the given voltage pulse protocol. We construct a Pareto front plot using the two objectives to find a set of optimal diagnostic protocols. The improved performance of optimally designed protocols is validated via Markov chain Monte Carlo simulations under various degradation scenarios, highlighting the extrapolation capability of the model-based DoE. From this optimization task, we attribute the poor identifiability of $R_{\mathrm{f,a}}$ to its small sensitivity that is comparable to the measurement noise. We propose applying redundant pulses to further improve the estimation accuracy and practical identifiability of $R_{\mathrm{f,a}}$.

The second part of this thesis describes the optimization of formation protocols using a systematic feature engineering framework as the source of interpretability. The framework automatically determines which portion of the measurements to use, which type of data to use as the input data, and which specific values to use for partitioning the input data. The main bottleneck for optimizing the formation protocol is the evaluation process, which could take several months to years for LiBs to reach the end-of-life. Here, we break down the

evaluation process into two parts. We first map the formation protocol to formation features designed by our framework. Then, we map from formation features to the cycle life using data-driven modeling. This allows to complete the evaluation of new formation protocols during the formation step, converting it into a tractable task. The extrapolation capability of the designed formation features is ensured by adopting a nested cross-validation within the feature engineering framework. By combining domain knowledge with observations from the data-driven feature work, we find a missing link connecting the formation condition and the derivatives of the $Q(V)$ curve from the last discharging of the formation step. Inspired by this missing link, we develop a mechanistic model that explains it based on the ion-coupled electron transfer theory.

The last part of this thesis explores how to utilize the information captured in the Acoustic Emission (AE) data to supplement well-established methods that use macroscopic electrochemical measurements, such as cell-level current and voltage. AEs are sensitive to microscopic mechanical changes, such as gas generation or particle fracture. In order to effectively use this information, it is essential to carefully identify the acoustic source of each AE. Given the complexity of this identification, the transferability of knowledge mapping AEs to their acoustic sources from half-cell AEs to full-cell AEs is greatly valuable. We process over wavelet transform, dimension reduction, the simplest structure of convolutional neural network, and supervised clustering to achieve interpretable classification of AEs. Our method shows possible transferability from half-cell AEs to full-cell AEs, as well as potential transferability across different cell chemistries.

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