Simulating, Controlling, and Understanding Lithium-ion Battery Models

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Lithium-ion batteries are widespread in consumer electronics, electric vehicles, and grid storage. Developing better batteries and more intelligent battery management systems is an active area of research – operating batteries safely, maximizing their lifetimes, and inventing new materials are essential for their continued proliferation. To create the next generation of batteries, researchers must tackle challenging, multi-disciplinary problems with an enormous design space.

Experimentally testing batteries can be expensive and time-consuming. Extensive analyses usually involve dozens of batteries that may be tested continuously over several weeks or months. Efficient physics-based and data-driven modeling can significantly reduce the cost and time requirements of battery development. Still, physically accurate simulations face numerous technical and theoretical barriers stemming from difficulties in measuring and analyzing battery internals during cycling.

This thesis focuses on simulating, controlling, and understanding rigorous physics-based lithium-ion battery models. The first Part of this thesis presents PETLION, an open-source, high-performance implementation of the Porous Electrode Theory (PET) model. PET typically contains several hundred nonlinear differential-algebraic equations (DAEs) after discretization. This package is designed from a systems engineering perspective to be robust and highly efficient, about 100–1000x faster than other available implementations of PET while maintaining the same physical accuracy. PETLION is the cornerstone for the following Parts of this thesis, permitting deep analyses of PET which would otherwise be prohibitively expensive.

The second Part of this thesis investigates a mixed continuous-discrete (hybrid) approach for fast charging of batteries in real-time. Traditional fast charging problem setups perform optimal control with a reduced-order/empirical model to find the optimal current profile, maximizing the capacity subject to safety and degradation constraints. Instead, a hybrid charging procedure is proposed, simultaneously solving the battery system of equations and the embedded solution to the constraint-based control problem. Here, the fast-charging current profile is found via direct simulation, which dynamically switches between active path constraints. Novel operating modes (such as constant temperature and lithium plating overpotential) are efficiently simulated using DAEs in PET and other simpler models without deriving analytical solutions. Next, this methodology is applied to PET coupled with an empirical degradation model to simulate long-term cycling behavior using physics-based operating modes. These approaches move beyond conventional constant current-constant voltage (CC-CV) charging protocols and show that degradation-aware protocols using physics-based operating modes such as constant temperature, constant solid-electrolyte interface (SEI) growth rate, and constant State of Health (SOH) rate of decay can charge batteries more quickly than CC-CV while generating equal levels of degradation at similar computational costs.

The third and final Part of this thesis applies nonlinear identifiability analyses to the PET model, describing what can and cannot be learned from a combination of model, data, and unknown parameters. The first step theoretically investigates the uniqueness of five diffusion and transport coefficients for a synthetic discharge curve generated from a $LiCoO_2$ battery. Markov chain Monte Carlo (MCMC) shows that only a single parameter, the anodic solid diffusion coefficient, can be

uniquely estimated within a 95% confidence interval, signifying that the resistance to lithium flow in the anode is also meaningful and measurable from this model-data combination. Parameter unidentifiabilities can be resolved by incorporating new and different measurements of the system. Next, this methodology is applied to 7776 discharge curves spanning the lifetime of 95 nickel cobalt aluminum oxide (NCA) cells with Si-doped graphite anodes. With PETLION, 1 billion PET simulations for the MCMC analysis can be efficiently computed in about one week. Parameter identifiability trends are observed to change as cells become more degraded. While the anodic solid diffusion coefficient began as the only identifiable parameter for pristine cells, the cathodic solid diffusion coefficient and anodic rate constant (and their resistances) became identifiable as the cells degrade. Simple fitted relationships for the parameters are proposed as a function of discharge capacity which may be implemented in hybridized battery models to simulate capacity fade.

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