



Lithium-ion battery capacity fading dynamics modelling for formulation optimization: A stochastic approach to accelerate the design process



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HIGHLIGHTS

- The model is linked to known physicochemical degradation processes and material properties.
- Aging dynamics of various battery formulations can be understood by the proposed model.
- Large number of experiments will be reduced to accelerate the battery design process.
- This approach can describe batteries under various operating conditions.
- The proposed model is simple and easily implemented.

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ABSTRACT

A five-state nonhomogeneous Markov chain model, which is an effective and promising way to accelerate the Li-ion battery design process by investigating the capacity fading dynamics of different formulations during the battery design phase, is reported. The parameters of this model are linked to known physicochemical degradation dynamics and material properties. Herein, the states and behaviors of the active materials in Li-ion batteries are modelled. To verify the efficiency of the proposed model, a dataset from approximately 3 years of cycling capacity fading experiments of various formulations using several different materials provided by Contemporary Amperex Technology Limited (CATL), as well as a NASA dataset, are employed. The capabilities of the proposed model for different amounts (50%, 70%, and 90%) of available experimental capacity data are tested and analyzed to assist with the final design determination for manufacturers. The average relative errors of life cycling prediction acquired from these tests are less than 2.4%, 0.8%, and 0.3%, even when only 50%, 70%, and 90% of the data, respectively, is available for different anode materials, electrolyte materials, and individual batteries. Furthermore, the variance is 0.518% when only 50% of the data are available; i.e., one can save at least 50% of the total experimental time and cost with an accuracy greater than 97% in the design phase, which demonstrates an effective and promising way to accelerate the Li-ion battery design process. The qualitative and quantitative analyses conducted in this study suggest that the proposed model provides an accurate, robust, and simple way to accelerate the Li-ion battery design process for battery manufacturers, thereby enabling rapid market capture.

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1. Introduction

Lightweight Li-ion batteries with high energy densities [1] have been gaining attention from both academia and industry, becoming

the most promising candidate for various applications [2–6]. To fulfill customer demand or to adjust to rapid changes in the battery market, battery manufacturers must continuously design and produce new batteries with high or specified performance. Beyond all performance constraints, some objectives for battery design are clearly defined for the service life of the battery (10–15 years or 20,000–30,000 charge-discharge cycles [1]).

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However, it is inevitable that the performance of a Li-ion battery gradually degrades over time due to fading, environmental impacts and dynamic cycling [7]. Such degradation can lead to increasing cost and even catastrophic failure [8]; as a result, a series of issues must be addressed before the final design of each formulation is confirmed in the battery design phase:

- (1) Several battery formulations must be proposed in order to find a candidate which meets the specified customer or market requirements.
- (2) The battery cycling life tests for each formulation must be performed for various temperatures and discharging rates, requiring a significant time investment.
- (3) The required experiments are costly in terms of money, person-hours, and equipment.
- (4) Longer times are spent determining a specific formulation, resulting in fewer opportunities to capture the battery market.

All of these issues stress the importance of finding an effective approach which will minimize the number of cycling experiments, saving time and money and accelerating the Li-ion battery design process.

To address battery design phase issues and to accelerate the battery design process, a few publications have addressed the battery design process, all with the purpose of saving money and time. Firstly, a series of experimental studies have been conducted in investigating the impact of different current rates, working conditions, previous history, and other factors on battery performance of Li-ion batteries [9–12]. These efforts were helpful for battery modelling and battery health management especially in cases of considering the factors affects on performances and real applications for battery cells or packs with the same formulation. Besides, Ref. [13] comprehensively summarized the multiscale modelling and numerical simulation for li-ion batteries which could be useful for saving time when establishing physical-chemistry based battery models. Ref. [14] used first principles to calculate voltage curves for electrode materials in Li-ion cells and to predict average intercalation voltages of unknown materials. Ref. [15] derived sensitivity equations to obtain sensitivity coefficients, which are used to guide the thermal design of a battery. Ref. [16] developed an electro-thermal finite element model to predict the thermal performance of nickel-metal-hydride (NiMH) modules with realistic geometry, which are unsuitable for Li-ion batteries with the same power and capacity [17].

These existing methods do save time and money prior to design confirmation and attempt to minimize formulations by predicting and transferring features and performance from one formulation to another. However, considering the stochastic features of both battery operating conditions and battery behaviors, as well as the variety between cells in similar formulations, the calculated results varied widely, even for the same formulations and methods. The variation in results is most likely because (1) some models and parameters were selected unsystematically, often with unfounded assumptions; (2) the coefficients vary widely and are sensitive to external factors.

In Ref. [13], the author pointed out that a good Lithium-ion battery (LIB) model should be flexible, which allows the designers to quickly test the new LIB designs and figure out the optimal LIB with special formulation that satisfies the technical requirements. In addition to those design phase methods, Ref. [18] postulated that a lifetime prediction method based on the battery model is necessary for selecting the most technologically suitable and cost effective battery, which requires a greater understanding of battery capacity cycling fading dynamics. For batteries currently in use, many researchers have been trying to explore and predict battery

cycling fading dynamics [1,19]; a number of models have been presented [6,20–23]. These existing models can be sorted into the following categories: electrochemical models, electrical equivalent circuit models, and mathematical models [24]. Electrochemical models and electrical equivalent circuit models can establish accurate models for complex physical and chemical processes in consideration of the dynamic behavior of batteries; however, they are complicated and always difficult to establish [25]. Some of these models rely on impedance measurements, which require expensive equipment, significant amounts of idle time, and stringent measurement techniques [8]. Importantly, during the design phase, there are large numbers of proposed cells, all with variations in formulation. It may not be possible to generate an electrochemical model for each formulation. Mathematical models such as the kinetic battery model [26], which is too large to handle as a whole (the available charge well is discretized into 27×107 and 45×107 charge units), introduce high computational complexity. Furthermore, all of these models are employed to understand and predict a specific battery's performance for use in battery management.

Recently, Risse et al. [27] introduced a Markov model for analyzing capacity density (mAh/g-S) fading curves of Li/S cells, demonstrating a simple, effective way to investigate the fading dynamics of Li/S cells with different cathode materials. Stochastic-based approaches can therefore provide promising ways to understand the battery dynamics of a cell using various materials while still in the design phase [27,28]. In contrast with the aging processes of Li/S batteries, the solid electrolyte interphase (SEI) layer in Li-ion batteries mainly grows on the anode [29], and capacity loss due to SEI growth is usually attributed to the negative electrode [30–33]. Additionally, Risse [27] treated the transition probabilities as constants, which does not make sense for the continuous aging process of a battery in use. Furthermore, the authors of Ref. [27] did not give any further analysis on how to apply their model for predictions, for reducing the number of experiments needed, or for a way to accelerate the design process.

In summary, few publications have demonstrated an ability to understand and predict cycling fading dynamics for various battery formulations with different materials and working conditions during battery design phase. Those existing methods [14–17] which attempt to evaluate the performance of a battery design by transferring and predicting features from one formulation to another obtain results with wide variations, even for the same formulations and methods. In addition, considering the fact that there are large number of formulations under consideration in the design phase, those complicated and high computational approaches, like electrochemical models [8,25] and kinetic battery model [26], might be not suitable in modelling for each formulation for battery formulation determination. In this study, to overcome the aforementioned issues and to begin research on how to accelerate the design process, a five-state nonhomogeneous Markov chain model whose parameters are linked to known physicochemical degradation dynamics and material properties of Li-ion batteries is designed. The proposed model enables the investigation of and predictions for Li-ion battery capacity cycling fading dynamics. The changes in the transition probability matrix among different cycles are also introduced to model, which represents the differences in various health states properly in the continuous aging process. To verify the efficiency of the proposed model, a multitude of cycling fading experiments for various formulations with different anode and electrolyte materials have been carried out, which has been conducted by the Contemporary Amperex Technology Limited (CATL) for approximately 3 years. In addition, the public dataset from NASA was implemented as one possible Li-ion formulation with which to test the proposed model. The qualitative and quantitative

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