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Applicability of available Li-ion battery degradation models for system and control algorithm design

Xing Jin [a], Ashish Vora [a], Vaidehi Hoshing [a], Tridib Saha [b], Gregory Shaver [a,*], Oleg Wasynczuk [b], Subbarao Varigonda [c]

[a] School of Mechanical Engineering, Purdue University, 585 Purdue Mall, West Lafayette, IN 47907, USA
[b] School of Electrical and Computer Engineering, Purdue University, 465 Northwestern Ave., West Lafayette, IN 47907, USA
[c] Cummins Inc., Columbus, IN 47202, USA

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ABSTRACT

Within electrified vehicle powertrains, lithium-ion battery performance degrades with aging and usage, resulting in a loss in both energy and power capacity. As a result, models used for system design and control algorithm development would ideally capture the impact of those efforts on battery capacity degradation, be computationally efficient, and simple enough to be used for algorithm development. This paper provides an assessment of the state-of-the-art in lithium-ion battery degradation models, including accuracy, computational complexity, and amenability to control algorithm development. Various aging and degradation models have been studied in the literature, including physics-based electrochemical models, semi-empirical models, and empirical models. Some of these models have been validated with experimental data; however, comparisons of pre-existing degradation models across multiple experimental data sets have not been previously published. Three representative models, a 1-d electrochemical model (a combination of performance model and degradation model), a semi-empirical degradation model (the performance is predicted by an equivalent circuit model) and an empirical degradation model (the performance is predicted by an equivalent circuit model), are compared against four published experimental data sets for a 2.3-Ah commercial graphite/$LiFePO_4$ cell. Based on simulation results and comparisons to experimental data, the key differences in the aging factors captured by each of the models are summarized. The results show that the physics-based model is best able to capture results across all four representative data sets with an error less than 10%, but is $20x$ slower than the empirical model, and $134x$ slower than the semi-empirical model, making it unsuitable for powertrain system design and model-based algorithm development. Despite being computationally efficient, the semi-empirical and empirical models, when used under conditions that lie outside the calibration data set, exhibit up to 71% error in capacity loss prediction. Such models require expensive experimental data collection to recalibrate for every new application. Thus, in the author's opinion, there exists a need for a physically-based model that generalizes well across operating conditions, is computationally efficient for model-based design, and simple enough for control algorithm development.

1. Introduction

Vehicle electrification offers the potential to decrease fuel use, greenhouse gas emissions, and tailpipe emissions of criteria pollutants. Batteries play an important role in electrified propulsion systems. Lithium ion batteries have the advantage of high energy density, high power density, and low self-discharge rate (Oswal, Paul, & Zhao, 2010). These favorable properties, as well as decreasing costs, have made them the most popular technology for automotive applications (Safari & Delacourt, 2011b; Wang et al., 2011).

In spite of these favorable properties of lithium ion batteries, aging and degradation during usage remain an impediment to widespread adaption. Loss of cyclable lithium via side reactions, loss of electrode active material, and resistance increase through interfacial layer growth or contact issues are among the most common aging phenomena encountered in lithium ion batteries (Delacourt & Safari, 2012). The

* Corresponding author.
E-mail address: gshaver@purdue.edu (G. Shaver).

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rate of capacity loss for each mechanism is a complex nonlinear process dependent on various factors including temperature, state of charge (SOC), depth of discharge (DOD), C-rate, and storage condition of the battery.

Several types of models for predicting battery degradation have been widely used in hybrid electric vehicle system-level study, including physics-based electrochemical models (Jin et al., 2017c; Lawder, Northrop, & Subramanian, 2014; Prada et al., 2013; Vora et al., 2015), semi-empirical models (Smith, Markel, & Pesaran, 0000), and empirical models (Peterson, Apt, & Whitacre, 2010; Vora et al., 2017; Wang et al., 2011). These models describe the dependence of battery resistance and capacity fade on various aging factors (Jin, 2017; Smith, Earleywine, Wood, Neubauer, & Pesaran, 2012; Smith et al., 0000). Due to their simplicity and sufficient accuracy for many applications, semi-empirical and empirical models have been used for on-line state-of-health (SOH) estimation. As will be shown, the usability of these models, however, is generally limited to operating conditions characteristic of the data used for calibration. To cover a relatively wide range of operating conditions, expensive and time consuming experiments need to be conducted. In contrast, electrochemical models, which describe aging phenomena with some physical basis, have the potential to allow the analysis to be extended to a larger range of operation conditions once the model is calibrated, but are computationally more expensive than the aforementioned empirical and semi-empirical models.

A semi-empirical degradation model developed by National Renewable Energy Laboratory (NREL) (Smith et al., 0000, 2013) was generated by compiling experimental data under different aging conditions from multiple sources. Reference (Wang et al., 2011) proposed an empirical model that was fitted to experimental data with variable temperatures, C-rates and DODs. Reference (Onori, Spagnol, Marano, Guezennec, & Rizzoni, 0000) presented a weighted ampere-hour throughput model based on a severity factor map that captures the battery degradation due to the different operating factors including temperature and C-rate. Several physics-based aging models for lithium-ion batteries have also been described in the literature. In Safari, Morcrette, Teyssot, and Delacourt (2009) and Delacourt and Safari (2012), an aging model based on the single-particle approach was developed to take into account the solid-electrolyte-interface (SEI) layer thickness growth and possible active material loss at both anode and cathode. A correlation of graphite active material loss as a function of C-rate and temperature was implemented to get better agreement with experimental data. Reference (Prada et al., 2013) showed a simplified isothermal physics-based aging model, in which the SEI layer growth was related to both capacity loss and internal resistance increase. However, to the best of the authors' knowledge, none of these models have been compared with each other and with multiple experimental datasets spanning various operating conditions.

The objective of this paper is to help model users understand the strengths and weaknesses of these different types of degradation models and thus select the most appropriate model, if available, for their specific application. As will be shown, the higher fidelity degradation model described by the electrochemical model relies on the performance information, i.e. solid/electrolyte phase potential, solid/electrolyte concentration, and current density. The two degradation mechanisms described by ordinary differential equations (ODEs) are easy to solve, but the inputs to those ODEs require solving the electrochemical performance model. It should be highlighted that several authors proposed different ways to simplify the performance model, including Padé approximation (Forman, Bashash, Stein, & Fathy, 2011), residue grouping (Smith, Rahn, & Wang, 2008), polynomial approximation (Subramanian, Diwakar, & Tapriyal, 2005), Galerkin projection (Fan, Pan, Canova, Marcicki, & Yang, 2016), etc., however, none of them include degradation model simplification, computational complexity, or accuracy considerations. The semi-empirical and the empirical degradation models, on the other hand, do not need detailed knowledge of the electrochemical system, which is an advantage of these simpler models. The inputs for these models, i.e. current, SOC, voltage, can be obtained by the equivalent circuit model, the electrochemical model, or experimental data.

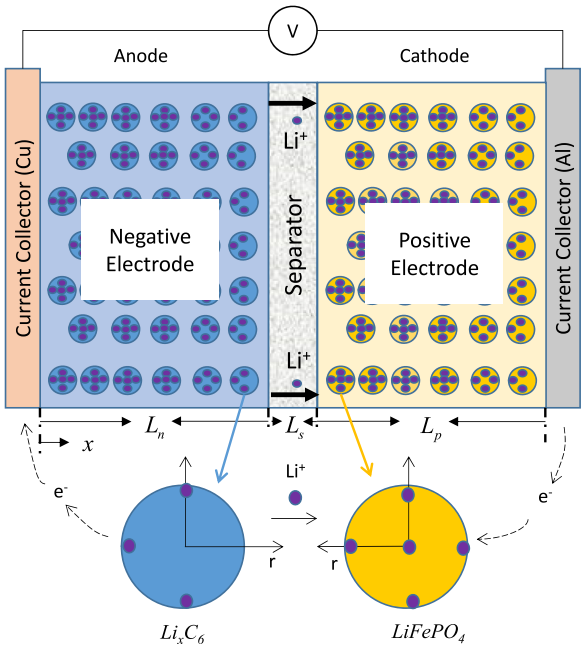

Fig. 1. Schematic of a Li-ion battery (graphite/$LiFePO_4$), with a separator between two electrodes (anode and cathode) (Smith & Wang, 2006).

A commercial graphite/$LiFePO_4$ (LFP) cell, a promising technology for automotive applications due to its excellent chemical and thermal stability and low cost (Wang et al., 2011), is taken as an example to illustrate the differences. This paper is organized as follows: Section 2 describes three battery degradation models, including a 1-D electrochemical model, a semi-empirical model, and an empirical model. Section 3 presents the model-to-model and model-to-data comparisons, and some discussion of these results. It also describes the tradeoff between accuracy and computational time for the 1-D electrochemical model. Finally, Section 4 concludes the paper with some key takeaways.

2. Battery degradation models

A schematic diagram of a typical graphite/$LiFePO_4$ (LFP) cell with a single pair of porous electrodes is shown in Fig. 1 (Smith & Wang, 2006). As shown, the model includes three domains: negative electrode (anode), separator, and positive electrode (cathode). During discharge, Li ions (represented as purple dots) diffuse to the surface of the LiC_6 active material particles within the anode, de-insert from the surface, and then transfer into the electrolyte solution. These lithium ions travel through the electrolyte solution and separator to the cathode, and diffuse towards the inner regions of the $LiFePO_4$ active material particles. The separator is an ionic conductor and electronic insulator, which forces the electrons to flow through an external circuit. The reverse occurs during the charging process.

Along with the charge and discharge operations, aging or degradation phenomena also occur inside a battery, including capacity fade, power fade, stress–strain effects, and mechanical degradation. For each type of degradation model i.e. physics-based, semi-empirical and empirical, there are many models available. AutoLion ST is a commercially available 1-D electrochemical model, which is implemented as a software-in-the-loop model block. It has been validated across different chemistries under different operating conditions (Kalupson, Luo, & Shaffer, 2013; Vora et al., 2015, 2017). Different semi-empirical and empirical models published in the literature are calibrated for different