



Aging formula for lithium ion batteries with solid electrolyte interphase layer growth



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HIGHLIGHTS

- Developed a nonlinear, electrolyte enhanced, single particle cell model (NESPM).
- The NESPM includes aging due to solid electrolyte interphase (SEI) layer growth.
- Aging model is simplified to explicit capacity fade and impedance rise formulas.
- Key aging parameters and operating conditions are identified to minimize aging.
- Presents model-based hypothesis on battery management with experimental validation.

ARTICLE INFO

Article history:

Received 31 January 2015
 Received in revised form
 22 May 2015
 Accepted 2 June 2015
 Available online xxx

Keywords:

Lithium ion battery modeling
 Single particle model
 Hybrid electric vehicle
 Battery management system

ABSTRACT

Hybrid Electric Vehicle (HEV) current profiles are dynamic, consisting of repeated charge and discharge pulses. Accurate prediction of the battery response to these inputs requires models with open circuit voltage and Butler–Volmer kinetic nonlinearities. This paper derives a nonlinear, electrolyte-enhanced, single particle model (NESPM) that includes aging due to solid electrolyte interphase layer growth. The model is validated with experimental full charge, discharge, HEV cycle, and aging data from 4.5 Ah graphite/LiFePO₄ cells. The NESPM is capable of operating up to 3C constant charge–discharge cycles and up to 25C and 10 s charge–discharge pulses within 35–65% state of charge (SOC) with less than 2% error. The NESPM aging model is then simplified to obtain explicit formulas for capacity fade and impedance rise that depend on the battery parameters and current input history. The formulas show that aging increases with SOC, operating temperature, time, and root mean square (RMS) current. The formula predicts that HEV current profiles with the (i) same average SOC, (ii) small SOC swing, (iii) same operating temperature, (iv) same cycle length, and (v) same RMS current, will have the same cell capacity fade.

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1. Introduction

Two major goals of the automotive industry are increasing fuel efficiency and reducing green house gas emissions in order to attract customers and comply increasingly stringent emission regulations, e.g. Corporate Average Fuel Economy standard in the U. S. of 54.5 miles per gallon by 2025 [1]. Partial and full electrification of automobiles, trucks, and buses is a viable solution to achieve these goals [2]. Research also suggest that HEVs and Plug-in HEVs (PHEVs) will dominate the market by 2034 and 2045, respectively [3,4].

Lithium ion (Li-ion) batteries are well suited for electric vehicles (EVs), PHEVs, and HEVs [5]. The challenge is to choose the best cells for a specific application and optimal pack size to maximize fuel economy at minimum cost, taking into consideration the fact that batteries age and their performance degrades. It is very important to understand usage associated with a specific application influences cell degradation. Accurate modeling and prediction of the battery State of Health (SOH) is crucially important for the battery management system (BMS) to perform efficiently and economically control, manage, and estimate the pack to prevent unexpected performance deterioration and premature battery end of life (EOL).

A typical HEV current profile is complex, consisting of repeated charge and discharge pulses, operating around a narrow SOC window [5,6]. Sharp edged, sloped, and smooth current pulses are possible. HEV pulse trains have different timings, shapes, and

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polarities. A clear understanding of the controlling parameters for cell aging associated with different HEV current profiles would enable system engineers to optimally design and operate HEV battery packs to enhance life within a desired performance window.

Lithium Iron Phosphate (LFP) cathode-based Li-ion batteries possess high temperature abuse tolerance, low cost, and an environmentally benign nature [7,8]. LFP cathodes are extremely stable due to their olivine crystalline structure. Continuous small scale side reactions at the SEI layer of the negative carbonaceous graphite (gr) anode, however, is considered to be the main cause of aging [9–11]. The SEI aging process of an LFP cell couples with the cell electrochemistry, so model-based aging analysis requires an accurate electrochemical model.

The intercalation/deintercalation processes in LFP is thought to occur through a two phase process between a Li-poor Li_xFePO_4 phase and a Li-rich $\text{Li}_{1-x}\text{FePO}_4$ phase [12–14]. Srinivasan and Newman [12] develop a core–shell model initially proposed by Padhi et al. [7] with a growing shell of LiFePO_4 (Li-rich phase) on a shrinking FePO_4 (Li-poor phase) core during discharge. This core–shell model is improved by other researchers [15–17] through tracking of multiple two-phase boundaries for repeated charge–discharge cycles but the simulation is complicated and computationally demanding. A relatively simplified phase-change diffusivity model is proposed by Thorat [18]. A phase-field model, based on the Cahn–Hilliard theory is also reported for the LFP electrode [19]. Interestingly, some recent studies also claim that the two distinct phase processes during lithiation/delithiation in an LFP cathode occur only at low currents. Large currents suppress the phase transformation [20–22] in the LFP cathode. It is also reported that LFP active material shows resistive reactant property which attributes in increasing cell impedance as charge or discharge proceeds [23,24].

Complex full-order physics-based models run off-line and require higher computational resources [12,25,26]. These models are useful for battery design and performance optimization. An advanced on-board BMS, however, performs various control and management tasks in real-time and warrants a faster model which captures the most important dynamics of the cell within an acceptable error bound. Equivalent circuit models [27–30] are one way to reduce the computational burden but require extensive empirical parametrization, and lack the insight into the underlying physio-chemical processes of the cell. Physics-based reduced order models that include the battery internal dynamics provide another way to achieve the desired real-time computational efficiency. Smith et al. [31,32] first use residue grouping to derive a 7th order isothermal model for Li-ion cells, Klein et al. [33,34] assume volume averaging and constant electrolyte concentration. Single particle models without electrolyte dynamics (SPM) [35–38] are a relatively simple physics based modeling approach but may not be applicable for high C-rate applications [35] typical in HEVs. Researchers have also included the electrolyte dynamics into the SPM. Rahimian et al. [39] added polynomially approximated electrolyte dynamics with the SPM to reduce the PDE system into 13 differential algebraic equations. Marcicki et al. [40] included 1st order truncated electrolyte dynamics from cell specific transcendental transfer functions. Prada et al. [41] modeled a gr/LFP cell in a single particle framework and varied the active material particle sizes. Recently, an analytic, linearized, and control-oriented 7th order SPM with electrolyte diffusion dynamics and temperature effect broadened the conventional SPM's operating range up to 20C and 10 s charge–discharge pulses within a 35–65% SOC window, suitable for HEV applications [42,43].

Mathematical models of the cyclable lithium consumption in the negative electrode that contributes to SEI growth are reported

by many researchers [44–46]. Ramadass et al. [47] first propose a simplified control-oriented incremental aging model of the negative electrode in conjunction with a 1D full order electrochemical model. Randall et al. [48] further reduce this aging model and propose an iterative approach to calculate the SEI side reaction current density to quantify cell aging.

In this study, the control-oriented anode SEI aging model in Ref. [47] is coupled with a nonlinear model for gr/LFP cells. More specifically, this study (i) develops a physics-based, reduced order, nonlinear, and electrolyte enhanced single particle model (NESPM), (ii) couples the SEI aging model of the negative electrode with the NESPM, (iii) further simplifies the NESPM aging model to an analytic aging formula to identify the controlling parameters of cell aging for HEV applications, (iv) compares the NESPM aging and aging formula for two different HEV current profiles, and (v) experimentally validates the aging predictions. Commercially available 4.5 Ah gr/LFP cylindrical cells are used in the experimental studies.

2. Mathematical model

Assuming LFP takes part in conventional intercalation/deintercalation reaction, Fig. 1 shows a schematic view of a gr/LFP Li-ion cell which consists of three domains: a porous anode with spherical graphite particles, porous separator, and porous cathode with spherical active material particles. The most widely used liquid electrolyte typically contains 1.2 M LiPF_6 in ethylene carbonate (EC)/dimethyl carbonate (DMC) and saturates the three cell domains. The separator is a micro porous polymer which allows Li-ion flow but restricts the electron flow. Very thin foils of aluminum (Al) and copper (Cu) are attached to the positive and negative electrodes, respectively, as current collectors.

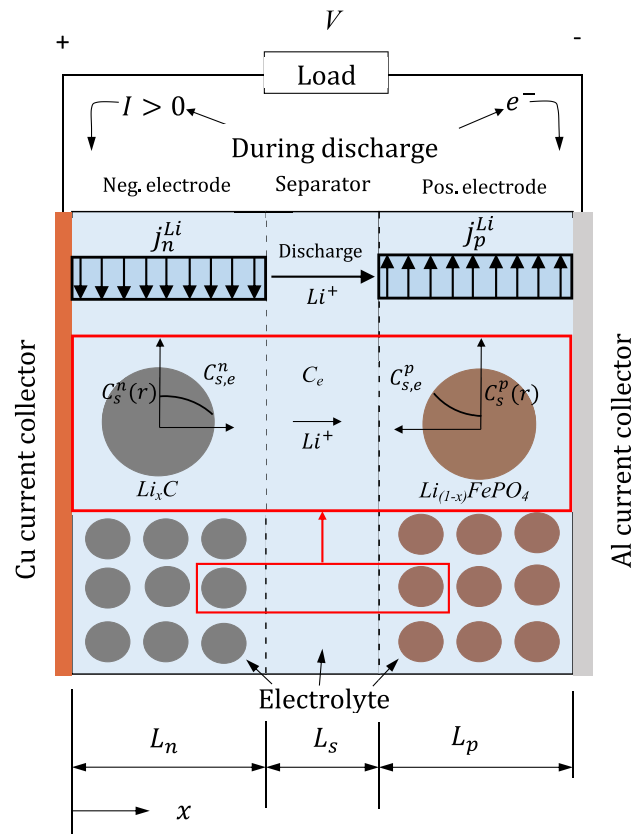


Fig. 1. Schematic diagram of an SPM Li-ion cell model.

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