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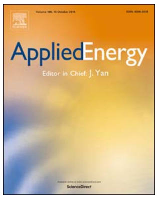

A single particle model with chemical/mechanical degradation physics for lithium ion battery State of Health (SOH) estimation

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HIGHLIGHTS

- Developed a single particle model coupled chemical/mechanical degradation physics.
- Capacity fade and voltage changes are accurately and quickly predicted during cycling.
- The impact of operating parameters on battery capacity and power are revealed.

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Single particle model
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State of health
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ABSTRACT

State of Health (SOH) estimation of lithium ion batteries is critical for Battery Management Systems (BMSs) in Electric Vehicles (EVs). Many estimation techniques utilize a battery model; however, the model must have high accuracy and high computational efficiency. Conventional electrochemical full-order models can accurately capture battery states, but they are too complex and computationally expensive to be used in a BMS. A Single Particle (SP) model is a good alternative that addresses this issue; however, existing SP models do not consider degradation physics. In this work, an SP-based degradation model is developed by including Solid Electrolyte Interface (SEI) layer formation, coupled with crack propagation due to the stress generated by the volume expansion of the particles in the active materials. A model of lithium ion loss from SEI layer formation is integrated with an advanced SP model that includes electrolytic physics. This low-order model quickly predicts capacity fade and voltage profile changes as a function of cycle number and temperature with high accuracy, allowing for the use of online estimation techniques. Lithium ion loss due to SEI layer formation, increase in battery resistance, and changes in the electrodes' open circuit potential operating windows are examined to account for capacity fade and power loss. In addition to the low-order implementation to facilitate on-line estimation, the model proposed in this paper provides quantitative information regarding SEI layer formation and crack propagation, as well as the resulting battery capacity fade and power dissipation, which are essential for SOH estimation in a BMS.

1. Introduction

Lithium Ion Batteries (LIBs) are key energy storage devices for many applications due to their high energy and power densities, and are widely used in Electric Vehicles (EVs) and Hybrid Electric Vehicles (HEVs). A critical challenge, however, is the capacity degradation experienced during repeated charge/discharge cycles. Battery performance declines over time due to irreversible physical and chemical changes that naturally occur until the battery can no longer be used. Therefore, State of Health (SOH) estimation is an essential component of a Battery Management System (BMS) for a variety of energy storage systems in transportation and stationary applications [1,2]. However, there are challenges in performing SOH estimation in a BMS. First, SOH cannot be directly measured. Here, SOH refers to the state of a battery's condition compared to its initial condition, and is expressed as a loss of capacity relative to the initial value. For instance, when the battery capacity in EVs/HEVs reaches 80% of its initial capacity, the battery is no longer considered usable [1].

Secondly, battery performance continuously degrades due to various mechanisms, both mechanical and chemical, which affect LIBs during their lifetime. Therefore, studying these mechanisms require long-term, in situ testing that typically requires the battery to be destroyed. Moreover, it is challenging to decouple the effects of each of these mechanisms on battery health and performance. Modern LIBs,

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Nomenclature

a	crack length (m)
a_0	initial crack length (m)
$brug$	Bruggeman coefficient
c_0	initial concentration (mol m^{-3})
$C_{\max,\ pos}$	positive maximum concentration (mol m^{-3})
$C_{\max,\ neg}$	negative maximum concentration (mol m^{-3})
D_e	diffusion coefficient in electrolyte (m^2 s^{-1})
$D_{s,j}$	solid-phase Li diffusivity (m^2 s^{-1})
E_{a1}	activation energy for crack propagation (kcal mol^{-1})
E_{a2}	activation energy for SEI layer growth (kcal mol^{-1})
E	Young's modulus of electrode material (N m^{-2})
F	Faraday's constant (C mol^{-1})
$i = p/s/n$	positive/separator/negative
I_{app}	applied current density (A m^{-2})
$j = p/n$	positive/negative
J_j^{Li}	molar flux density (mol s m^{-2})
k_i	electrolyte conductivities (S m^{-1})
k_j	reaction rate constant (m$^{2.5}$ mol$^{-0.5}$ s^{-1})
k_0	crack propagation coefficient
K_{SEI0}	SEI layer growth coefficient
k_{SEI}	SEI layer conductivity (S m^{-1})
l_{cr0}	initial crack width (m)
L_{SEI}^0	initial SEI layer thickness (m)
L_i	electrode thickness (m)
M_{SEI}	molecular weight of compounds constituting SEI (g mol^{-1})
n_{Li}	initial total number of lithium ions inside battery
n_{SEI}	consumed lithium ion for 1 mol of SEI layer formation
N	cycle number
Q_0	battery capacity after formation cycle
Q_{in}	battery capacity before SEI layer formation cycle
Q_g	graphite specific capacity (A h g^{-1})
Q_N	capacity after Nth cycle
R	universal gas constant (J mol^{-1} K^{-1})
r_j	particle radius (m)
R_{SEI}^0	initial SEI layer resistance (Ω)
t_+	cationic transport number
$V(0)\vert_N$	initial voltage at each cycle (V)
ε_i	electrode porosity
ρ_{cr}	number of cracks per unit area of particle (m^{-2})
ρ_g	graphite density (g cm^{-3})
ρ_{SEI}	density of SEI films (g m^{-3})
Ω	partial molar volume of solute (m^3 mol^{-1})

which are fabricated from a variety of anode and cathode materials, degrade due to a number of mechanisms that depend on the chemical nature of their constituent materials. In general, battery degradation mechanisms [3–6] include current collector corrosion, morphological changes of active materials, electrolyte decomposition, Solid Electrolyte Interphase (SEI) layer formation, and material dissolution. For example, carbonaceous materials, which are the most common anode materials in modern LIBs, have a significant amount of irreversible capacity loss during initial cycling as the SEI layer is formed on the carbon surface [3–5,7]. Moreover, the SEI layer continues to grow due to the continuous reduction of the electrolyte and the reformation of the SEI layers, which re-consumes lithium ions and results in irreversible battery capacity loss. For instance, about 8–15% irreversible capacity loss due to lithium ion loss is expected for MesoCarbon MicroBeads [5]. Further, mechanical damage to the battery will accelerate chemical degradation. The basic function of an electrochemical material is fulfilled by ions entering the active materials due to an electrochemical potential gradient [8]. During this intercalation process, volume change causes considerable stresses inside the particles, leading to mechanical failures such as pulverization of, or cracks and fractures in, the active materials. These cracks generate new surfaces on the particle, which are then exposed to the electrolyte, leading to additional SEI layer formation and the acceleration of capacity fade and power loss [8–14].

Despite these difficulties, the foundation of many models of different battery aging mechanisms has been presented by the research community to predict battery life. The studies in [15–20] utilized physics-based models that provide very detailed information regarding the battery electrochemical response. Capacity loss due to SEI layer formation was simulated through a continuum-scale mathematical model by considering the flux of the side reaction at the anode particle surface [15,16] or by modifying the solid phase concentration to be a function of the cycle according to lithium ion loss [17–19]. The effect of mechanical degradation on capacity fade has also been simulated using a full order physics-based model [20]. These models solve governing physical equations, which include mass conservation and charge conservation in the solid and electrolyte phases, as well as kinetic reactions at the interface between the solid and electrolyte.

A comprehensive and detailed state-of-the-art review of SOH estimation for LIBs has been conducted in [21]. These methods can be categorized as experimental techniques and adaptive methods. Experimental techniques depend on recorded experimental data and previous knowledge about the effect of the operating conditions such as temperature, cycle number, SOC, current magnitude on the battery cycle life [21]. Although these methods are easy to implement onboard BMSs, their validity is limited to the calibration data used in their development. Furthermore, the wide range of operating conditions encountered in different battery applications, such as EVs, necessitates the use of an adaptive SOH estimation methods. Adaptive methods utilize parameters from battery models to estimate the SOH. Electrochemical models can provide highly accurate predictions of the battery behavior while providing insight into internal battery phenomena. Due to these advantages, these models are gaining popularity in recent years in the development of different BMS functionalities, such as SOH estimation. For BMSs, however, there is still a challenge in incorporating battery physics and degradation during on-line estimation.

Although a high fidelity electrochemical modes are ideal for the detailed analysis of battery phenomena, they are too computationally intensive to be efficiently utilized in a BMS [22]. This has led to efforts to reduce the complexity of electrochemical models, such as the studies in [23,24] that reduced the electrochemical model proposed by Doyle et al. [25] into a form suitable for a BMS. To further simplify battery models, most studies employ an Equivalent Circuit Model (ECM), which describes the battery dynamic behavior as a voltage source and a series of resistors and capacitors [26] and is widely used in BMSs due to the model's low complexity and ease of online implementation. However, ECM models have no physical significance, which leads to low fidelity and limited prediction capability. In addition, the prediction of battery side-reactions is not feasible due to the difficulty of obtaining the battery internal dynamic characteristics. Therefore, higher accuracies can be attained only by considering time-variant model parameters. The Single Particle (SP) model is a common type of reduced-order model. The SP model strikes the necessary balance between full order electrochemical models and ECMs, and is becoming a popular model in recent years for SOC and SOH estimation [27–31]. It assumes both electrodes are composed of multiple uniform sized spherical particles, and that the current distribution is uniform across both electrodes. Thus, each electrode can be approximated by a single spherical particle. The SP model is described by a set of ordinary differential equations, yet it is directly derived from comprehensive electrochemical models and, thus, explicitly retains many important battery properties with