



Model based identification of aging parameters in lithium ion batteries

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HIGHLIGHTS

- ▶ Developed a control oriented single particle model for a Li-ion battery.
- ▶ Developed least squares and recursive parameter estimators for SOH estimation.
- ▶ Observed a monotonic increase in the estimates of resistance and diffusion time.
- ▶ Correlated the parameter variations to aging mechanisms such as SEI layer growth.

ARTICLE INFO

Article history:

Received 9 October 2012
 Received in revised form
 5 December 2012
 Accepted 3 January 2013
 Available online 12 January 2013

Keywords:

Battery management systems
 Degradation mechanisms
 State of health estimation
 Control oriented model

ABSTRACT

As lithium ion cells age, they experience power and energy fade associated with impedance rise and capacity loss, respectively. Identification of key aging parameters in lithium ion battery models can validate degradation hypotheses and provide a foundation for State of Health (SOH) estimation. This paper develops and simplifies an electrochemical model that depends on two key aging parameters, cell resistance and the solid phase diffusion time of Li^+ species in the positive electrode. Off-line linear least squares and on-line adaptive gradient update processing of voltage and current data from fresh and aged lithium ion cells produce estimates of these aging parameters. These estimated parameters vary monotonically with age, consistent with accepted degradation mechanisms such as solid electrolyte interface (SEI) layer growth and contact loss.

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1. Introduction

Hybrid and electric vehicles can contribute to a greener and cleaner environment by substantially reducing the dependence on non-renewable fossil fuels such as gasoline and diesel. Lithium ion batteries are widely used for energy storage in these vehicles mainly due to their high power and energy density. Hybrid electric vehicles have repeated charge and discharge cycles which deteriorate the battery due to irreversible physical and chemical degradation. State of health (SOH) in a lithium ion battery is typically defined as the ratio of the current capacity over the nominal capacity of a fresh battery and monotonically decreases as the battery ages. Impedance also rises as a battery ages, decreasing the maximum power output and efficiency.

Researchers have extensively studied capacity and power fade in lithium ion batteries [1–3]. Power fade is primarily due to an increase in internal resistance or impedance. Internal resistance

causes ohmic losses that waste energy, produce heat, and accelerate aging. Lithium ion batteries lose capacity over time due to degradation of the positive and negative electrodes and the electrolyte. The degradation mechanisms are complex, coupled, and dependant on cell chemistry, design, and manufacturer [4].

Considerable effort has been put into the development of high fidelity battery models that accurately predict voltage given the input current and model parameters [5,6]. The model parameters that provide the best match between the model predicted and experimentally measured voltage change with age. The change in system parameters due to aging depends on the degradation mechanism in a given cell. If the predominant degradation mechanism can be determined then the parameters that are most closely associated with that mechanism would be most likely to change. If the degradation mechanism involves unmodeled dynamics in the cell, however, then the correlation between the mechanism and system parameters becomes unclear.

Ramadass et al. [3] link cell aging to the change of only a few parameters in an electrochemical battery model. For a lithium ion cell, they find that the solid electrolyte film resistance and the solid state

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diffusion coefficient of the anodic active material are linked to cell degradation. Schmidt et al. [7] find that electrolyte conductivity and cathodic porosity are key parameters to estimate the rate capability fade and the capacity loss of a Li-ion cell. Zhang et al. [8] characterize the cycle life of lithium ion batteries with LiNiO₂ cathode and conclude that the impedance rise and capacity fade during cycling are primarily caused by the positive electrode. A solid electrolyte interface (SEI) layer forms on the positive electrode and it thickens and changes properties during cycling, causing a cell impedance rise and power fade.

Parameter estimation techniques based on equivalent circuit models have been developed to quantify the degradation in lithium ion batteries. Remmlinger et al. [9] monitor the state of health of lithium ion batteries in electric vehicles using an onboard internal resistance estimation technique. Kalman filters [10–14] have been developed to estimate the SOH using the equivalent circuit models. Troltzsch et al. [15] characterize aging effects in lithium ion batteries using impedance spectroscopy. Kim et al. [16] implement a dual sliding mode observer to estimate the capacity fade in lithium ion batteries.

In this paper, we estimate the SOH of a lithium ion battery based on a control oriented single particle model that explicitly relates the coefficients of the Padé approximated transfer function to the fundamental physical parameters of the battery. Two key composite parameters, the cell resistance and the diffusion time of the Li⁺ species in the positive electrode are estimated based on voltage and current data from batteries that have been aged using a 5C rate cycle. The parameter variation with age is correlated to capacity loss and specific degradation mechanisms. The identified parameters act as SOH indicators if they can be shown to vary monotonically as the battery degrades.

2. Control oriented single particle model

A lithium ion cell consists of three domains: A negative composite electrode, a separator, and a positive composite electrode as shown in Fig. 1. Lithium metal oxide (LiMO₂) and lithiated carbon (Li_xC) are the active materials in the positive and negative electrodes, respectively. The metal in the positive electrode is a transition metal, typically Co. The active materials are bonded to metal foil current collectors at both ends of the cell and electrically insulated by a microporous polymer separator film or gel-polymer. Liquid or gel-polymer electrolytes enable lithium ions (Li⁺) to diffuse between the positive and negative electrodes. The lithium ions insert into or deinsert from the active materials via an intercalation process. The insulating separator forces electrons to follow an opposite path through an external circuit or load.

The single particle (SP) model used in this paper assumes a single electrode particle in each electrode and negligible electrolyte diffusion and is detailed in [17]. The governing equations, transcendental transfer functions, and Padé approximation are summarized here for completeness.

Conservation of Li⁺ species in a single spherical active material particle is described by Fick's law of diffusion,

$$\frac{\partial c_s}{\partial t} = \frac{D_s}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial c_s}{\partial r} \right) \quad \text{for } r \in (0, R_s), \quad (1)$$

where $r \in (0, R_s)$ is the radial coordinate, R_s is the particle radius, $c_s(r, t)$ is the concentration of Li⁺ ions in the particle as a function of radial position r and time t , and D_s is the solid phase diffusion coefficient. We use the subscripts s and e to indicate solid phase and solid/electrolyte interface, respectively. The boundary conditions are

$$\left. \frac{\partial c_s}{\partial r} \right|_{r=0} = 0, \quad (2)$$

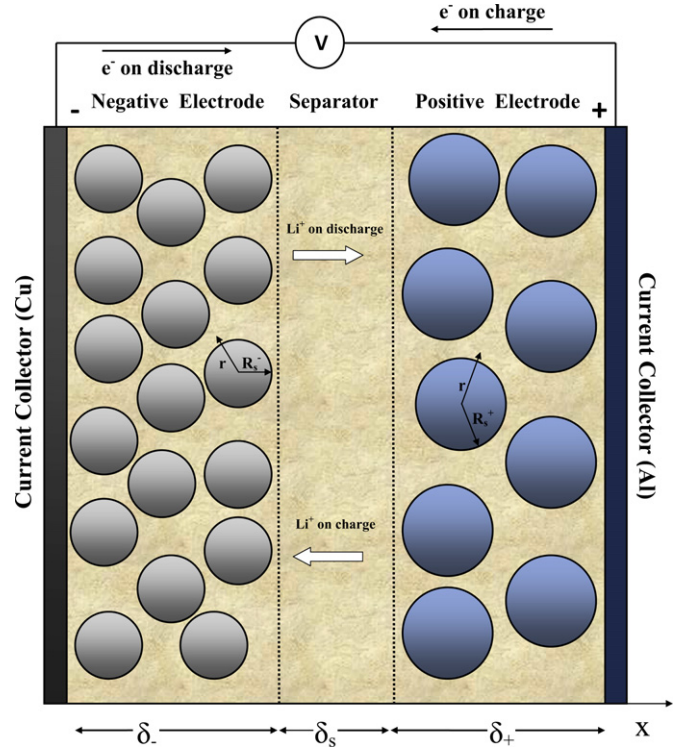


Fig. 1. 1D schematic of the lithium ion battery model.

$$D_s \left. \frac{\partial c_s}{\partial r} \right|_{r=R_s} = -\frac{j}{a_s F}, \quad (3)$$

where $j(x, t)$ is the rate of electrochemical reaction at the particle surface (with $j > 0$ indicating ion discharge), F is Faraday's constant (96487 C mol⁻¹), and a_s is the specific interfacial surface area. For the spherical active material particles occupying electrode volume fraction ε_s , $a_s = 3\varepsilon_s/R_s$. The linearized Butler–Volmer electrochemical kinetics is given by

$$\eta = \frac{R_{ct} j}{a_s} \quad (4)$$

where the overpotential η drives the current flow across the electrode/electrolyte interface, $R_{ct} = RT/i_0 F(\alpha_a + \alpha_c)$ is the charge transfer resistance, i_0 is the exchange current density, R is the universal gas constant, T is the temperature, and α_a and α_c are the anodic and cathodic transfer coefficients, respectively.

The final transcendental impedance transfer function is given by

$$\begin{aligned} \frac{V(s)}{I(s)} = & \frac{R_{ct+}}{a_s} \frac{1}{A\delta_+} - \frac{R_{ct-}}{a_s} \frac{1}{A\delta_-} \\ & - \frac{\partial U^+}{\partial c_{s,e+}} \frac{1}{A\delta_+} \frac{R_s}{a_s + FD_{s+}} \left[\frac{\tanh(\beta_+)}{\tanh(\beta_+) - \beta_+} \right] \\ & - \frac{\partial U^-}{\partial c_{s,e-}} \frac{1}{A\delta_-} \frac{R_s}{a_s - FD_{s-}} \left[\frac{\tanh(\beta_-)}{\tanh(\beta_-) - \beta_-} \right] - \frac{R_f}{A} \end{aligned} \quad (5)$$

where the electrode open circuit potentials U are linearized with the assumed constant slope $\partial U/\partial c_{s,e}$, $\beta = R_s \sqrt{s/D_s}$ with s the Laplace variable, δ is the thickness of the electrode, R_f is the contact resistance associated with physical connections to the cell and A is the cell surface area.

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