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System theoretic analysis of battery charging optimization

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ABSTRACT

Innovative optimal charging sequences are sought to minimize battery degradation and reduce charging loses. One charging sequence under investigation incorporates high frequency pulses or sinusoidal perturbations. However, there is disagreement in the literature on the benefits/disadvantages of pulsed charging. This work analytically determines optimal battery charging sequences for two cases: minimizing energy losses and maximizing charge supplied to the battery while respecting lithium plating constraints. Assuming relevant battery physics have linear, time-invariant dynamic behavior, optimal charging strategies are derived analytically. The analysis exposes specific features of the battery dynamic response (frequency response and impulse response) that must be present for optimal charging sequences to have periodic components. In the case of energy minimization, the battery's electrochemical impedance must approach the imaginary axis. In the case of limiting lithium plating, a criterion is developed involving impulse responses of relevant battery variables.

1. Introduction

Lithium-ion batteries have attractive features for many applications, and are widely used as high energy density storage devices. The internal electrochemical and chemical reactions occurring during charge/discharge make Li-ion batteries sensitive to demand sequences and operating conditions [1]. Because cells are sensitive to system demands and operating conditions, battery management systems are developed to keep cells within acceptable operating ranges to prolong life, reduce charging losses, and mitigate failure [2,3]. This paper uses analytical tools to provide analysis and insight into optimal charging profiles proposed by the research community to better inform battery management systems.

A particular area of interest in the research community are charge/discharge protocols that incorporate pulses or sinusoidal perturbations. Experimental and simulation evidence has been published arguing that pulsed charging optimally charges/discharges cells [4–9], while others have argued that pulsed charging has no effect or hinders performance [10–12]. It can be argued that experimental results evaluating pulsed charging are subtle enough that experimental conditions and uncertainty can result in both positive and negative interpretation. A physical, mechanistic explanation for the positive effects of perturbations is a key element to guiding further work.

There are several ways to evaluate 'optimal' charging. This paper will focus on two particular criteria for optimal charging: efficiency of charging and reduction of lithium plating. The goal of this paper is to use basic system theoretic analysis to determine optimal charging sequences and evaluate mechanisms that contribute to the 'shape' of optimal pulsed charging sequences.

1.1. Increasing charging efficiency

It has been argued that sinusoidal/pulse sequences increase the charging efficiency, meaning less energy is required to charge a battery to a desired state-of-charge. Some explanations provided in the literature include:

- If a sinusoidal perturbation is chosen at the frequency where the electrochemical impedance is minimized, "... the energy loss in electrical energy transfer to chemical energy is minimized" [7].
- If a sinusoidal perturbation is chosen in the correct frequency range, "... concentration polarization in the electrodes is minimized because charge accumulation is reduced" [9]. That is, a phase shift occurs between peak current and peak concentration polarization.
- Pulsed charging with a rest period allows "ions to diffuse and distribute electrolyte ions more evenly to change the

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concentration at the surface of the electrode" increasing the exchange current density [5].

1.2. Reduction of lithium plating

Sinusoidal/pulse sequences are also argued to minimize aging due to lithium plating. Two (related) explanations for this are:

- Pulsing reduces lithium concentration at the electrode interface. "The proposed pulse sequence enables higher charging rates, without ever reaching lithium saturation" [4].
- Pulsing reduces the polarization voltage. "Large polarization not only renders the battery undercharged but also results in active material loss" [8].

1.3. Scope of this paper

This paper provides an answer to the following question: What are the features of a battery, as a linear dynamic system, that must be present for optimal charging strategies to include oscillatory components such as pulses or sinusoids? The results will, in fact, provide evidence against several of these mechanistic explanations given above regarding the benefits of pulsed charging. Of course, the restriction to linear dynamics limits the scope of our conclusions. However, within this limited scope, we believe this analysis provides useful insight into how battery dynamic behavior impacts optimal charging sequences, and can provide a starting point for interpreting the results of experimental and numerical results of higher fidelity.

1.4. Related work

A standard charging protocol for Li-ion batteries is to charge the battery at a constant current until a voltage limit is reached. Once the voltage limit is reached, the current is reduced to maintain this maximum voltage. This protocol is called constant-current/constant-voltage (CC–CV) charging [13]. While simple to implement and widely used, it is primarily derived from basic principles of battery operation. There are efforts seeking to optimize these charging sequences to reduce charging time, increase charging efficiency, and other objectives. The most systematic analysis involves numerically searching for an optimal charging sequence using a battery model [14–21]. These methods investigate optimization of a variety of cost functions, including charging time and charging energy. Constraints include internal temperature, terminal voltage, and overpotential. Since these methods do not result in an optimal charging sequences that contain an oscillatory component, it can call into question whether pulsed or sinusoidal charging is truly useful. On the other hand, these numerical methods only provide the optimal sequence for a specific battery model, limiting their ability to provide general insight. In addition, it can also be argued that an optimal charging current containing a periodic component can be lost in numerical error, especially if the periodic component is relatively high frequency.

The current paper provides a different type of analysis, that while not capturing all possible battery behavior, does address *dynamic* effects that are captured by linear models. This paper does not purport to offer an alternative method for numerically finding an optimal charging sequence, but instead uses analytic results to provide insight between battery dynamics and the qualitative behavior of the optimal charging sequence.

1.5. Notation

$j \equiv \sqrt{-1}$. L_2 is the set of square integrable functions. L_1 is the set of absolutely integrable functions. $L_2[0, T]$ is the set of functions square integrable over the domain $[0, T]$. For a complex number x, $\mathrm{Re}\{x\}$ is the real part and $\mathrm{Im}\{x\}$ is the imaginary part. For a function $x(t)$, $\mathcal{L}\{x(t)\}$ is the one-sided Laplace Transform of $x(t)$,

$$\mathcal{L}\{x(t)\} = \int_0^\infty x(t)e^{-st}dt. \tag{1}$$

$\mathcal{L}^{-1}$ is the inverse Laplace transform operator.

2. Analysis of energy optimal charging

To evaluate charging sequences that minimize losses, consider a battery model consisting of a linear circuit element in series with a non-linear capacitive element. The non-linear capacitive element models the open circuit voltage as a function of state of charge, while the linear circuit models transient effects, such as diffusion and charge transfer. To charge the battery, a current $i(t)$ is supplied, which in turn causes a response in the terminal voltage $v(t)$. Based on the modeling assumptions, the battery's electrical response has the following relationships between applied current, voltage, and state of charge

$$v(t) = \int_0^T g(t-\tau)i(\tau)d\tau + v_{\mathrm{oc}}(s_{\mathrm{c}}(t)), \tag{2}$$

$$s_{\mathrm{c}}(t) = s_{\mathrm{c}}(0) + \frac{1}{Q_0}\int_0^t i(\tau)d\tau, \tag{3}$$

where $g(t)$ is the impulse response of the linear element (that is, the voltage trajectory across the element due to a short pulse of current through the element), T is the total amount of charging time, and v_{oc} is the open circuit voltage, which is a function of the state of charge $s_{\mathrm{c}}(t)$. The integral involving $g(t)$ and $i(t)$ is a convolution, and is a time-domain solution for the response of a linear system with impulse response $g(t)$ to the input signal $i(t)$ [22]. The state of charge, in turn, is the integral of the applied current, normalized by the charge at full capacity, Q_0, plus the initial state of charge $s_{\mathrm{c}}(0)$.

There are many different physical behaviors that contribute to battery dynamics, including diffusion of lithium and lithium ions in anode and cathode, charge transfer across double layers that develop at electrode/electrolyte interfaces, and electrochemical reactions associated with lithium intercalation, among others. In this model, the internal processes (i.e., polarization and chemical kinetics) are exposed only through their influence on the terminal voltage. However, when trying to minimize charging losses (i.e. maximize charging *efficiency*) the terminal voltage completely specifies the energy required to supply a given current, and thus reflects all internal losses or chemical efficiency inside a cell. In the literature, battery models that predict the terminal voltage are often modeled using lumped equivalent circuit models. Fig. 1 shows a relatively simple equivalent circuit model interpreted from the literature [12], with parameters shown in Table 1. In this case, the open circuit voltage is modeled by a voltage V_0 representing the voltage at low state of charge, and a capacitor providing a linear increase in open circuit voltage with respect to state of charge. While this simple equivalent circuit model will be used as an example, our results apply to any general open circuit voltage relationship.

As will be shown, the existence or non-existence of an optimal charging sequence containing a sinusoidal component is determined by the battery's frequency response. For batteries, the