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# Non-destructive fast charging algorithm of lithium-ion batteries based on the control-oriented electrochemical model

Zhengyu Chu, Xuning Feng, Languang Lu, Jianqiu Li, Xuebing Han, Mingguo Ouyang\*

State Key Laboratory of Automotive Safety and Energy, Tsinghua University, Beijing 100084, China

## HIGHLIGHTS

- A novel non-destructive fast charging algorithm of lithium-ion batteries is proposed.
- A close-loop observer of lithium deposition status is constructed based on the SP2D model.
- The charging current is modified online using the feedback of the lithium deposition status.
- The algorithm can shorten the charging time and can be used for charging from different initial SOCs.
- The post-mortem observation and degradation tests show that no lithium deposition occurs during fast charging.

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## ABSTRACT

Fast charging is critical for the application of lithium-ion batteries in electric vehicles. Conventional fast charging algorithms may shorten the cycle life of lithium-ion batteries and induce safety problems, such as internal short circuit caused by lithium deposition at the negative electrode. In this paper, a novel, non-destructive model-based fast charging algorithm is proposed. The fast charging algorithm is composed of two closed loops. The first loop includes an anode over-potential observer that can observe the status of lithium deposition online, whereas the second loop includes a feedback structure that can modify the current based on the observed status of lithium deposition. The charging algorithm enhances the charging current to maintain the observed anode over-potential near the preset threshold potential. Therefore, the fast charging algorithm can decrease the charging time while protecting the health of the battery. The fast charging algorithm is validated on a commercial large-format nickel cobalt manganese/graphite cell. The results showed that 96.8% of the battery capacity can be charged within 52 min. The post-mortem observation of the surface of the negative electrode and degradation tests revealed that the fast charging algorithm proposed here protected the battery from lithium deposition.

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## 1. Introduction

The energy crisis and environmental pollution have aroused interest in clean energy vehicles using electrochemical power sources with lithium-ion batteries worldwide [1]. Fast charging is one of the most critical technologies for application in electrical vehicles [2]. Without fast charging, recharging these batteries takes several hours, and the charging rate is limited to 1 C or less [3] by manufacturers because of concerns relating to safety and durability. Therefore, a non-destructive fast charging algorithm is indispensable for use in electric vehicles.

The fast charging approaches based on empirical strategies have been widely discussed to reduce the charging time. Gener-

ally, empirical equations considering voltage or current cut-off values are used to determine the charging current. The most common charging protocol is constant current- constant voltage (CC-CV) charging, which consists of a CC stage followed by a CV stage to avoid overcharge [4]. The rule-based CC-CV charging protocol can be easily implemented in a battery management system (BMS) because of its simple logic and low computational load. However, the charging current and voltage limits for the CC-CV protocol are too conservative, leading to long charging times [5]. To accelerate charging, some fast charging strategies based on CC-CV have been proposed in recent years. For example, a scheme combining multi-stage CC with CV (MCC-CV), as presented in [6], can reduce the charging time. However, choosing the current amplitude and duration of each stage is essential but difficult. Furthermore, an algorithm called the boost charging algorithm, which includes CV-CC-CV charging stages, has been proposed and was

\* Corresponding author.

E-mail address: [ouymg@tsinghua.edu.cn](mailto:ouymg@tsinghua.edu.cn) (M. Ouyang).

## Nomenclature

$a_s$	specific interfacial surface area of particle ( $\text{m}^{-1}$ )	$U_{\text{ref}}^s$	equilibrium potential of the side reaction (V)
$a_i$	fitting coefficient of approximation expression of electrolyte concentration	<i>Greek</i>	
$A$	electrode plate area ( $\text{m}^2$ )	$\varepsilon_{\text{hj}}$	volume fraction in the phase “h” of electrode “j” or separator
$b_i$	fitting coefficient of approximation expression of equilibrium potential	$\kappa$	electrolyte phase conductivity ( $\text{S m}^{-1}$ )
$C_{s,\text{max}}$	maximum lithium concentration in the solid phase ( $\text{mol m}^{-3}$ )	$\kappa^{\text{eff}}$	effective electrolyte phase conductivity ( $=\kappa\varepsilon_e^{\text{Brugg}}$ ) ( $\text{S m}^{-1}$ )
$C_{s,\text{mean}}$	mean lithium concentration in the solid phase ( $\text{mol m}^{-3}$ )	$\kappa_D^{\text{eff}}$	diffusional effective electrolyte phase conductivity ( $=2RT\kappa^{\text{eff}}(t_+ - 1)/F$ ) ( $\text{S m}^{-1}$ )
$C_{s,\text{surface}}$	surface lithium concentration in the solid phase ( $\text{mol m}^{-3}$ )	$\phi_h$	local potential in the phase “h” (V)
$D_{e,j}^{\text{eff}}$	effective electrolyte ionic diffusivity of electrode “j” or separator ( $\text{m}^2 \text{s}^{-1}$ )	$\alpha$	charge transfer coefficient
$F$	Faraday constant ( $\text{C mol}^{-1}$ )	$\delta_j$	thickness of electrode “j” or separator (m)
$i_0$	exchange current density ( $\text{A m}^{-2}$ )	$\lambda$	weighting coefficient of the first-order process term
$I$	charge current (A)	$\omega$	first-order process term ( $\text{mol m}^{-3}$ )
$j_{f,j}$	volumetric current density of electrode “j” ( $\text{A m}^{-3}$ )	$\eta_{\text{act}}$	activation over-potential (V)
$j_f^s$	local volumetric current density of the side reaction ( $\text{A m}^{-3}$ )	$\eta_{\text{sr}}$	over-potential resulted by side reaction (V)
$j_n$	pore wall flux of the particle surface ( $\text{mol m}^{-2} \text{s}^{-1}$ )	$\eta_{\text{SEI}}$	over-potential across the SEI film (V)
$k$	reaction rate constant ( $\text{m}^{2.5} \text{A mol}^{-1.5} \text{s}^{-1}$ )	$\sigma$	solid phase conductivity ( $\text{S m}^{-1}$ )
$r$	radial coordinate (m)	$\sigma^{\text{eff}}$	effective solid phase conductivity ( $=\sigma\varepsilon_s$ ) ( $\text{S m}^{-1}$ )
$R$	gas constant ( $\text{J mol}^{-1} \text{K}^{-1}$ )	<i>Subscribe</i>	
$R_s$	Particle Radius (m)	j	electrode, =n or p
$R_{\text{SEI},j}$	resistance of the SEI film of electrode “j” ( $\Omega \text{m}^{-2}$ )	h	phase, =s or e
$t$	time (s)	s	solid phase
$t_+$	lithium ion transfer number	e	electrolyte phase
$T$	time constant of the first-order process (s)	n	negative electrode
$U_{\text{ref},j}$	equilibrium potential of electrode “j” or separator (V)	p	positive electrode
		sep	separator

demonstrated to operate with little capacity degradation [7]. However, it does not function when charging from a random state of charge (SOC). Compared with the algorithm containing CC and CV regimes, the linear current decay (LCD) protocol charges the battery with a high rate of current in the initial stage, and subsequently, the current decays linearly to reduce the polarization at the end of charging [8]. Unfortunately, LCD may lead to overcharge, which is detrimental to the battery's safety and life. Similarly, the variant current decay (VCD) protocol starts with a high pulse current, and then, the current decreases nonlinearly according to an empirical equation. VCD can be utilized to charge a battery at a higher rate but also leads to capacity fade and impedance increase at both electrodes [9]. Pulse charging (PC) has the potential to eliminate the polarization effect and increase the speed of charging [10,11], but the current waveform profile is difficult to set properly.

Although fast charging algorithms based on empirical parameters and rules are simple to implement in real vehicle BMSs, they are not based on the battery state and are hence inclined to negatively impact in the battery performance. To improve the reliability and non-destructiveness of fast charging, recent research has begun to address model-based methods. The electrochemical model [5,12–15] and the equivalent circuit model (ECM) [16–19], which are two major varieties of battery models, are reformulated and incorporated in optimal control frameworks to generate optimal charging strategies using various optimal algorithms. The electrochemical model, whose basic framework was first proposed by Newman et al. [20,21], is a mechanistic and comprehensive model based on the first principle describing the electrochemical behavior of the lithium-ion battery. The underlying model consists of four partial differential equations (PDEs) that describe the lithium-ion

concentration and charge conservation in the solid and electrolyte phase coordinated with one electrochemical kinetics equation. The lithium deposition status of the battery can be derived by using proper initial boundary conditions. Although the model can generate the real electrochemical status of the battery, it is not suitable for real-time use because of its high computational cost and non-linearity [22,23]. Compared with mechanistic models, the ECM has intrinsic limits in providing electrochemical information about a battery's internal states. Moreover, most of the aforementioned algorithms are designed for offline use without adaptive functions and, thus, have limited anti-interference capability in a real-time system. To enable monitoring the internal states of a battery online, a computationally inexpensive electrochemical battery model is needed.

The ultimate target of fast charging methods is to minimize both the charging time and the damages caused to the battery. Thermal characteristics [18], mechanical damage to the particles [14] and current and voltage constraints are general concerns during charging. However, lithium deposition might be a more critical problem because it can cause battery degradation and, potentially, critical safety issues. High-rate charging brings irreversible capacity loss, because of lithium deposition induced by high polarization [24]. Lithium metal deposits on the anode surface when the lithium-ion concentration on the particle surface reaches the saturation limit [25,26]. Lithium deposition also occurs under low-temperature or overcharging conditions. The active metallic lithium deposited during charging reacts with the electrolyte, resulting in loss of lithium inventory (LLI) in the battery [27]. The reaction gradually forms new sporadic solid electrolyte interphase (SEI) films, causing the resistance of the battery to increase. The deposited lithium is also likely to become dead lithium once

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