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Selection of charge methods for lithium ion batteries by considering diffusion induced stress and charge time

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HIGHLIGHTS

- Charge method is selected by considering both diffusion induced stress and charge time.
- An initial high current stage accelerates the charge process and reduces stress.
- Electrical current and time duration of the high current stage are designed.

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ABSTRACT

This article demonstrates the design of charging strategies for lithium ion batteries with considering the balance between diffusion induced stress and total charge time for two- and three-stage charge methods. For the two-stage galvanostatic-potentiostatic charge method the low mechanical stress can be achieved without increasing total charge time by switching the galvanostatic to the potentiostatic at the time moment when the lithium concentration at the surface of particles reaches the limit $\bar{c}_{\mathrm{surf}} = 0$. A three-stage method, which consists of an initial galvanostatic stage of high current, a galvanostatic stage of low current and a potentiostatic ending stage, is suggested. Employing the initial galvanostatic stage of high current is helpful not only in accelerating the charge process, but also in controlling the mechanical stress once the electrical current and time duration of the initial galvanostatic stage are properly designed.

1. Introduction

Lithium ion (Li-ion) battery is widely used in many applications nowadays due to its advantages such as high energy density, high operating voltage and low self-discharge rate. It is also one of the most promising technologies that meet the demands of electric vehicles (EV) [1]. There are many aging mechanisms, such as lithium metal plating, formation and growth of solid electrolyte interphase (SEI), mechanical failures and structural changes etc., that affect battery life-time [2]. Diffusion induced stress (DIS), which arises during charge and discharge, is considered as the direct cause of mechanical failures which may leads to loss of capacity and degradation of cyclic performance [2,3]. Although the battery aging does not originate from one single cause, it is believed that reducing and controlling DIS would improve the battery lifetime.

Evolution of diffusion induced stress in electrodes depends significantly on charge/discharge operations. There are two kinds of elementary operations, i.e. galvanostatic and potentiostatic, corresponding to constant current and constant potential, respectively. Cheng et al. [4] formulated DISes in a spherical particle under galvanostatic and potentiostatic operations, which were shown quite different. The stress increases initially and then decreases with time in potentiostatic operation, whereas it keeps increasing until a steady-state in galvanostatic operation. Zhang et al. [5] performed similar works for layered electrodes and suggested an optimized charge procedure, i.e. galvanostatic first and

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potentiostatic followed.

In real battery operations, charge/discharge usually follows complex procedures including galvanostatic and potentiostatic operations [6]. However, in literature, a large amount of works simulated the DISes under a single operation. For example, under the assumption of galvanostatic operation, Christensen and Newman calculated the concentration and stress profiles in spherical particles [7,8], Zhang et al. discussed the impacts of aspect ratio on DISes in an ellipsoidal particle [9], Bhandakkar and Gao investigated crack nucleation in thin strip and nanowire electrodes [10,11], as well as the works on electrode critical size by Zhao et al. [12] and Lu et al. [13–15]. These works, although provided important insights on DISes in electrodes, are incapable of revealing the DISes under real combinational galvanostatic and potentiostatic charge/discharge procedure of a battery.

In recent years, the combinational operations have been considered in several works. For example, Song et al. [16] discussed the effects of plasticity of current collector on battery performance in the cycles consisting galvanostatic and potentiostatic operations. Purushothaman et al. [6,17] proposed a pulsed currents charging method that contains multiple galvanostatic stages with different currents so as to accelerate charging process. However, it is still vacant comprehensive discussions on optimization of the combinational operation, including the current flux in galvanostatic stage and the switch time to potentiostatic stage, with consideration of both DIS and charge time.

In this paper, we will discuss the charge strategy which takes into consideration of both total charge time and mechanical safety. Based on the pulsed currents charging method reported by Purushothaman et al. [6,17], three-stage charge methods which includes two connective galvanostatic stages followed by a potentiostatic stage will be evaluated. Analytical models will be developed to simulate the evolutions of Li-ion concentration and DISes, with the aim to achieve a good balance between total charge time and mechanical stress. Our work will be able to provide a general solution for the Li-ion concentration and stress profile under the described combinational operations, and optimize the charge procedure by considering both total charge time and DISes.

2. Analysis

Consider a general charge procedure which combines multiple galvanostatic and potentiostatic operations with arbitrary time sequence. In order to analyze Li-ion concentration distribution in battery at the present time, the present single elementary (galvanostatic or potentiostatic) operation in the charge procedure will be focused and the concentration accumulated during the past charge history is assumed as the initial condition.

2.1. Concentration profile with general initial condition

A spherical active particle of radius R in electrodes is considered, within which the diffusion of Li-ions is assumed to be governed by Fick's law [4,18]:

$$D\frac{1}{r}\frac{\partial^2(rc)}{\partial r^2}=\frac{\partial c}{\partial t}, \tag{1}$$

where c is the molar concentration of lithium, D is the diffusion coefficient of lithium in the particle, t stands for time in a single elementary operation. The boundary conditions are

$$D\frac{\partial c}{\partial r}=\frac{i_n}{F}\ \text{for}\ r=R \tag{2}$$

for galvanostatic operation, where $F=96485.3C/mol$ is Faraday's constant, i_n is the surface current density, $i_n>0$ represents lithiation and $i_n<0$ indicates delithiation; and

$$c=c_{\text{surf}}\ \text{for}\ r=R \tag{3}$$

for potentiostatic operation, where c_{surf} is the surface concentration.

A lot of literature have investigated the concentration profile with a uniform initial solute concentration [4,7,9,19]. However, when operation switches, the initial concentration of the present operation is the final state of the previous one, usually not uniform. In this case, the initial condition of concentration profile is in a general form of

$$c=c_0(r)\ \text{for}\ t=0. \tag{4}$$

If the present operation is galvanostatic, by solving Equations (1), (2) and (4) we have the evolution of concentration follows:

$$\bar{c}=I\cdot f(\bar{r},\tau)+g(\bar{r},\tau), \tag{5a}$$

$$\text{with}\ f(\bar{r},\tau)=3\tau+\frac{1}{2}\bar{r}^2-\frac{3}{10}-\frac{2}{\bar{r}}\sum_{n=1}^{\infty}e^{-\lambda_n^2\tau}\frac{\sin(\lambda_n\bar{r})}{\lambda_n^2\sin(\lambda_n)}, \tag{5b}$$

$$g(\bar{r},\tau)=Q_0+\frac{2}{\bar{r}}\sum_{n=1}^{\infty}e^{-\lambda_n^2\tau}\frac{\sin(\lambda_n\bar{r})}{\sin^2(\lambda_n)}\int_0^1\sin\left(\lambda_n\bar{\zeta}\right)\bar{\zeta}\bar{c}_0\left(\bar{\zeta}\right)d\bar{\zeta}, \tag{5c}$$

where $\bar{c}=c/c_{\max}$, $\bar{c}_0=c_0/c_{\max}$, $\bar{r}=r/R$, $\bar{\zeta}=\zeta/R$, $\tau=Dt/R^2$, λ_n are the solutions of $\lambda_n\cot\lambda_n=1$, $c_{\max}$ is the saturation concentration, $I=i_nR/(FDc_{\max})$ is the dimensionless charge current density, $Q_0=3\int_0^1\bar{\zeta}^2\bar{c}_0(\bar{\zeta})d\bar{\zeta}$ is the normalized amount of lithium has been inserted into the particle in the past operations. The first term of the right-hand side of Equation (5a) represents the contribution of the present operation to the concentration which is proportional to the charge current density. The second term of the right-hand side of Equation (5a) denotes the contribution of past operations to the concentration at the present time. The spatial distribution of this part evolves with time but the total amount of lithium is unchanged during the present operation, i.e., $3\int_0^1\bar{r}^2g(\bar{r},\tau)d\bar{r}=Q_0$.

If the present operation is potentiostatic, the concentration at the present time can be obtained by solving Equations (1), (3) and (4), i.e.

$$\begin{aligned}\bar{c}\left(\bar{c}_{\text{surf}},\tau,\bar{r}\right)&=\bar{c}_{\text{surf}}\left[1+2\sum_{m=1}^{\infty}e^{-(m\pi)^2\tau}\frac{(-1)^m}{m\pi\bar{r}}\sin(m\pi\bar{r})\right]\\&+\frac{2}{\bar{r}}\sum_{m=1}^{\infty}e^{-(m\pi)^2\tau}\sin(m\pi\bar{r})\int_0^1\bar{c}_0(\bar{\xi})\bar{\xi}\sin(m\pi\bar{\xi})d\bar{\xi},\end{aligned} \tag{6}$$

where $\bar{c}_{\text{surf}}=c_{\text{surf}}/c_{\max}$. Again, the first term in the equation represents the contribution of the present operation while the second term gives the contribution of the past operations to the concentration at the present time.

If the initial concentration $\bar{c}_0$ is constant and does not vary spatially, Equations (5a) and (6) will degenerate to