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Reducing diffusion induced stress in planar electrodes by plastic shakedown and cyclic plasticity of current collector

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HIGHLIGHTS

- A strategy to reduce stress and enhance capacity by plastic yield of current collector.
- Identify 3 elastoplastic types: pure elastic, plastic shakedown, cyclic plasticity.
- Plastic shakedown reduces stress and enhances capacity with good safety.
- Cyclic plasticity further reduces stress and enhances capacity.
- Design schemes are provided for plastic shakedown and cyclic plasticity.

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ABSTRACT

This paper proposes a strategy to reduce the diffusion induced stress and enhance the capacity of a layered electrode by allowing the plastic deformation of current collector. Based on analytical formulations of the stress in whole electrode, three types of elastoplastic behaviors of current collector, i.e. pure elastic deformation, plastic shakedown and cyclic plasticity, are identified. Criterions separating the three cases are proposed. It is found applying a thin current collector and allowing it to plastically yield in the charge/discharge cycles is beneficial not only to capacity as more space can be provided for active materials but also to electrochemical stability because the stress in active layer is significantly reduced. Structural design corresponding to plastic shakedown shows good balance between the said improvements and structural safety, whereas the case of cyclic plasticity further enhances the improvements. Therefore, structural designing scheme is provided for the former case according to the criterion of plastic shakedown but for the latter one based on the Coffin–Manson relation with expected cycle life.

1. Introduction

Lithium ion batteries have been widely used in the applications from portable electronics to electric vehicles. The electrode of lithium ion battery usually employs a multilayer structure which is composed of active materials such as graphite, silicon or $LiCoO_2$, and current collector made of copper or aluminum. In the cycles of charge and discharge, stress field is developed in the whole electrode partially because the lithiation induced deformation of active layer is restricted by the current collector, leading to mechanical and further electrochemical degradation of the electrodes [1].

Among the intensive studies devoted to the diffusion induced stress (DIS), the current collector was customarily considered as an elastic material [2–5]. Mechanical plastic yield is usually avoided perhaps because of the traditional believe in mechanical design that plastic deformation is harmful to structural safety. However, the main function of current collector in batteries is to provide a passageway for electron transportation, not for load bearing. In addition, if the current collector plastically yields in charge and discharge, the relaxation brought by the plastic deformation may reduce the stress in active layer from which the electrochemical performance may benefit. Therefore, this article attempts to explore the possibility of allowing the plastic deformation of current collector in designing and taking advantage of the elastoplastic behavior of current collector to improve the performance of a battery. In literature, although the plastic yield of active material

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has been widely discussed [6–14], the impacts of plastic yield of current collector have been rarely investigated.

In this manuscript, we will demonstrate a strategy to reduce the DIS and enhance the capacity of a layered electrode by applying a thin current collector whose plastic deformation is utilized. There are three types of possible elastoplastic behaviors for the current collector, i.e. pure elastic deformation (PED), plastic shakedown (PS) and cyclic plasticity (CP). See Fig. 1, taking a cyclic uniaxial tension/compression test as example, pure elastic deformation is one in which plastic yield does not occur. Plastic shakedown is one in which plastic yield takes place only during the first loading, while the subsequent unloading and reloading are perfectly elastic. Cyclic plasticity is one in which alternating plastic yield of the material takes place in each loading–unloading cycle, leading to a closed elastic–plastic loop.

In order to characterize the impacts of the elastoplastic behavior of current collector on battery performance and designing insights, an analytical model will be established for a planar layered electrode. Yield criterion of current collector will be proposed, taking into account the impacts of material properties, electrode configuration and charging states. Evolutions of the stresses in both current collector and active layer in the cycles of charge and discharge will be simulated. Finally, designing insights will be provided based on the discussions.

2. Methodology

Consider a layered electrode in which two active layers of equal thickness h_1 are symmetrically bonded to a current collector of thickness h_c, Fig. 2. Let the thickness direction be aligned with the z-axis and the in-plane of plate with x- and y-axes. Lithium ions are inserted into and extracted out of the electrode from both side surfaces.

The Li-ion diffusion in the active layer is assumed to be governed by Fick's law. Other factors affecting the diffusion such as saturation cap [15], stress coupling [16], and concentration dependent elastic modulus [2,17], are neglected because the focus here is to investigate the impacts of plastic yield of current collector. Therefore, the diffusion is described by

$$\frac{\partial c}{\partial t} = D\frac{\partial^2 c}{\partial z^2} \tag{1}$$

where c is the molar concentration of Li-ions in the plate, D is the diffusivity of lithium ions.

Consider a full cycle, see Fig. 3. Both charge and discharge start with galvanostatic operation followed by potentiostatic operation.

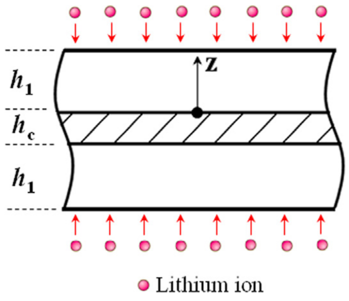

Fig. 2. The symmetric layered electrode investigated in this work.

In the charge phase of the cycle the lithiation of anode starts from a lithium free state. Hence, the initial condition and the boundary conditions for galvanostatic charging are

$$c = 0 \quad \text{for } t = 0 \tag{2a}$$

$$D\frac{\partial c}{\partial z} = \frac{i_n}{F} \quad \text{for } z = h_1 \tag{2b}$$

$$D\frac{\partial c}{\partial z} = 0 \quad \text{for } z = 0 \tag{2c}$$

where $F = 96485.3$ C mol^{-1} is Faraday's constant and i_n is the surface current density which is positive for lithiation but negative for delithiation. Due to the symmetry, only the equations of the upper active layer are provided. The distribution of Li-ion concentration is provided by Crank [18]:

$$c(z,t) = \frac{i_n h_1}{FD}\left\{\frac{Dt}{h_1^2} + \frac{3z^2 - h_1^2}{6h_1^2} - \frac{2}{\pi^2}\sum_{n=1}^{\infty}\frac{(-1)^n}{n^2}\cos\frac{n\pi z}{h_1}\exp\left(-n^2\pi^2\frac{Dt}{h_1^2}\right)\right\} \tag{3}$$

It is assumed that the charge operation switches from the galvanostatic to the potentiostatic at the time moment t_0 with Li-ion concentration $c(z,t_0)$. Hence, the initial and boundary conditions for potentiostatic charging are

$$c = c(z,t_0) \quad \text{for } t = t_0 \tag{4a}$$

$$c = c_0 \quad \text{for } z = h_1 \tag{4b}$$

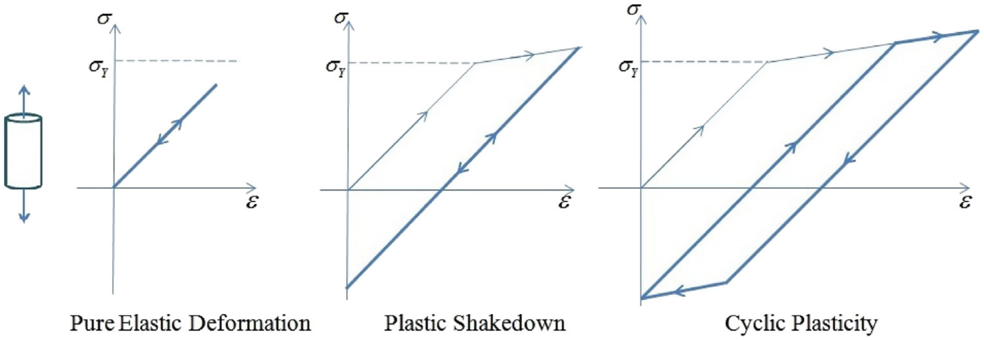

Fig. 1. Explanation of the terms "pure elastic deformation", "plastic shakedown" and "cyclic plasticity" with the stress–strain curve of a strain controlled uniaxial tension/compression test. σ_Y is the yield strength.