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A multiphysics model for the *in situ* stress analysis of the separator in a lithium-ion battery cell

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ABSTRACT

In lithium-ion (Li-ion) batteries, stresses arise as the results of the mechanical loading and constraint, the Li intercalation induced deformation in the active materials, and the thermal expansion mismatch between the battery components. Such stresses in the separator are of great concern for the safety and durability of the batteries. To assess the stress in the separator, a multiphysics model for a basic Li-ion battery cell including all three types of deformation has been developed. The time and temperature dependent constitutive behavior of a PP separator was characterized and modeled. The multiphysics model was used to analyze the stress in the PP separator. The results showed that the effects of Li intercalation, thermal mismatch and temperature on the stress in the separator are not a simple summation and hence must be considered concurrently. The Li intercalation induced dimensional change in the through-thickness direction can be minimized by design optimization.

1. Introduction

A battery separator is a membrane that prevents physical contacts between the positive and negative electrodes while enabling ionic transportation [1–3]. The structural integrity of the separator is vital to the performance and reliability of a battery. The failure of the separator can result in short circuit of the battery and lead to a thermal event. The stress can also result in creep deformation in polymeric separators which causes pore closure and therefore capacity fading of the battery [4,5]. To better understand the performance of battery separators, the stress in the separator must be known. This knowledge will serve as the foundation for further investigations into the separator property evolution (degradation) and its ability to tolerate abusive loadings. It will also provide the guidance in the future development of new separator materials, and in the search of an appropriate charge–discharge strategy for the enhanced battery durability in immediate applications.

Presently, there is no method to evaluate the stresses in a separator *in situ* in a battery. It has been recognized that, under normal operation conditions, the stress in the separator in a Li-ion battery cell is the results of the mechanical loading and constraints, the Li intercalation induced deformation in the active materials in the electrodes, and the thermal expansion mismatch between the battery components [6].

The intercalation stress is also known as the diffusion induced stress [7,8]. The intercalations tress inside a spherical electrode particle under various charge/discharge conditions have been investigated analytically [8–13]. The particle stress model has also been coupled with battery models and solved numerically [14–16]. However, the effect of the intercalation stresses on other battery components and the stress due to thermal expansion mismatch have not been reported. The focus of this work is to develop a model which can provide stress analysis for battery components including the separator with the consideration of ongoing electrochemical reaction and other physical phenomena which may influence the stress and deformation.

In a previous work, a multiphysics model which considers the intercalation induced stress in a basic Li-ion battery cell has been developed [6]. This model was built upon a pseudo-2D (P2D) battery model developed by Newman's group [17,18] and implemented in a finite element (FE) based multiphysics code COMSOL for a $Li_xC_6/LiPF_6/Li_yMn_2O_4$ cell [19]. For stress analysis, an idealized meso-scale representative volume element (RVE) of the battery cell was considered, which was added to the COMSOL battery model in the form of a sub-model. This work is the first that computes the effect of the intercalation induced stresses on battery components within a multiphysics framework with ongoing electrochemical reaction.

Batteries usually do not operate under an isothermal condition. The temperature is an important factor in transport and reaction kinetics [20,21]. To consider the effect of battery heat on electrochemical reaction and species transport, the P2D battery model was extended to a fully coupled thermal–electrochemical

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Nomenclature

E	Young's modulus (GPa)
F	Faraday's constant, 96487 (C mol^{-1})
G	stress relaxation modulus (Pa)
h	lumped heat transfer coefficient (W m^{-2} K^{-1})
I_0	current density (A m^{-2})
j_n	local current density (A m^{-2})
N_0	Li^+ flux (mol m^{-2} s^{-1})
S_t	tensile strength (Pa)
t	time (s)
T	absolute temperature (K)
U	open circuit potential of the electrode (V)

Greek letters

α_{ij}	thermal expansion coefficient (K^{-1})
δ_{ij}	Dirac delta function
ε_{ij}	strain component
ν	Poisson's ratio
σ	stress (Pa)
σ_Y	yield stress (Pa)
τ	relaxation time (s)
V	electrical potential
Ω	Partial molar volume (m^3 mol^{-1})

Subscripts and superscripts

1, 2, 3	the first, second and third principal directions
ei_T	eigen strain due to thermal expansion
ei_c	eigen strain due to intercalation
me	mechanical
VM	Vom Mises
∞	infinity

model [22] with local heat generation [23,24]. The model was validated against available experimental data in terms of cell temperature rise and heat generation rates. It was then used in the investigation of the relationships between the battery design parameters, temperature rise, and cell performance. It is worth noting that the heating in a battery may not be uniform, particularly in the areas near the current collecting tabs. The 1D macrohomogeneous approach cannot capture the 3D temperature field in a real battery. Rather, it serves as a tool in the investigation of coupled thermal multiphysics phenomena in a basic cell.

Currently, polymeric separators are used predominantly due to their low cost and relative thinness. A thin separator will facilitate the ionic transport and provide higher energy and power densities. The mechanical behaviors of the polymeric materials are known to have a strong dependence on time and temperature. A temperature rise of 20 °C is not uncommon for a battery during operation. This temperature variation may have a negligible effect on the mechanical properties of metallic and ceramic materials but it can cause a significant change in the properties of a polymer which in turn will influence the stress in the material.

This paper presents the recent development in the multiphysics battery model. In this work, the thermal stress and the temperature dependence of the separator were considered. The time temperature dependent constitutive behavior of a PP separator was characterized through the time–temperature superposition principle (TTSP) and modeled with a thermo-viscoelastic model. The multiphysics model was used to analyze the stress in the PP separator *in situ* in a basic battery cell. As in the previous work, the analysis was performed for a $Li_xC_6/LiPF_6/Li_yMn_2O_4$ cell.

2. Model description

The multiphysics model considers the charge/species transport through diffusion and migration, electrochemical reaction, heat generation, heat transfer, as well as the stress and deformation due to mechanical, thermal and Li intercalation effects. This model consists of four sub-models: Battery, Electrode, Thermal and Stress. Fig. 1 depicts the physical phenomena considered in each sub-model and the coupling relationship between them. The model was implemented in COMSOL, as illustrated in Fig. 2. To describe different physical phenomena, the model inevitably involves a large number of equations and parameters. Because the Battery, Electrode and Thermal sub-models have been described previously [6,22], the details about these sub-models are omitted here. Only the Stress sub-model and the coupling between the sub-models are discussed.

2.1. Stress sub-model

The Stress sub-model considers the strain due to mechanical load

$$\varepsilon_{ij}^{me} = \frac{1}{E}((1+\nu)\sigma_{ij} - \nu\sigma_{kk}\delta_{ij}) \tag{1}$$

the eigen strain due to thermal expansion

$$\varepsilon_{ij}^{ei\_T} = \alpha\Delta T\delta_{ij} \tag{2}$$

and the eigen strain induced by Li concentration gradient inside the active particles

$$\varepsilon_{ij}^{ei\_c} = \frac{1}{3}\Delta c\Omega\delta_{ij} \tag{3}$$

where σ_{ij} is the stress component, ε_{ij} is the strain component, E is the modulus of elasticity, ν is the Poisson's ratio, δ_{ij} is the Dirac delta function, α is the thermal expansion coefficient, c is the Li concentration, Ω is the partial molar volume, the superscript ^{me} refers to the strain due to mechanical loading, and $^{ei\_T}$ and $^{ei\_c}$ refer to the eigen strain due to thermal expansion and Li intercalation, respectively.

The total strain across the cell is the result of these three types of deformations

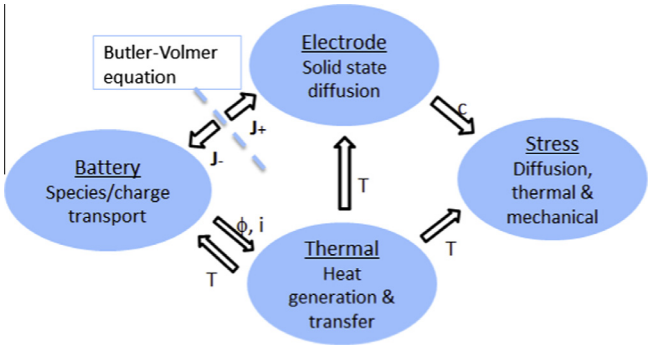

Fig. 1. A multiphysics model for a basic Li-ion battery cell. This fully coupled thermal-electrochemical-stress model consists of four sub-models. The schematic shows the physical phenomena in each sub-model and the coupling relationships among four sub-models.