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Phenomenological force and swelling models for rechargeable lithium-ion battery cells

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HIGHLIGHTS

- Three phenomenological models are developed to predict Li-ion battery behavior.
- The 1-D force model predicts Li-ion intercalation induced force in pack conditions.
- The 1st order relaxation model predicts Li-ion swelling during rest periods.
- The 3-D swelling model predicts the swelling shape for all SOC regions.
- Using advanced controls, these models can enhance battery pack capabilities.

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ABSTRACT

Three phenomenological force and swelling models are developed to predict mechanical phenomena caused by Li-ion intercalation: a 1-D force model, a 1st order relaxation model, and a 3-D swelling model. The 1-D force model can estimate the Li-ion intercalation induced force for actual pack conditions with preloads. The model incorporates a nonlinear elastic stiffness to capture the mechanical consequences of Li-ion intercalation swelling. The model also separates the entire state of charge range into three regions considering phase transitions. The 1st order relaxation model predicts dynamic swelling during relaxation periods. A coefficient of relaxation is estimated from dynamic and quasi-static swelling at operational conditions. The 3-D swelling model predicts the swelling shape on the battery surface for all states of charge. This model introduces an equivalent modulus of elasticity, which is dependent on the state of charge, to capture material transformations of the electrodes, and the orthotropic expansion of the jellyroll in a direction perpendicular to the electrode surfaces. Considering the simplicity of the measurements and direct physical correlations between stress and strain, the proposed models can enhance battery management systems and power management strategies.

1. Introduction

Volatility of oil prices, diminishing natural resources, and climate change are triggering many countries to investigate ways to reduce energy consumption. These trends motivate automobile industries to concentrate on the development of eco-friendly, high-efficiency vehicles. With the growing market for electrified vehicles, attention for Lithium-ion (Li-ion) batteries has increased because Li-ion batteries are reversible power sources used in electrified vehicles. This trend can be attributed mainly to their ability to combine a high gravimetric/volumetric energy and power density, which leads to compact and low-weight batteries [1–3]. Moreover, low self-discharge rate and long cycle-life make Li-ion batteries widely popular in portable electronics. Indeed, Li-ion batteries are highly versatile energy storage devices for a variety of applications from small-scale portable electronics to large-scale electrified vehicles.

Recent market demands for advanced Li-ion batteries emphasize not only high-energy/power density but also improved reliability and safety for the application of electric vehicles (EVs) and hybrid electric vehicles (HEVs) due to the stringent safety standards for air and ground applications [4]. The safety and reliability of Li-ion batteries, both of which are critical for the development of EVs and HEVs, can be improved with accurate battery models combined with novel battery management strategies. Thus, there have been considerable efforts to develop battery models to depict

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the dynamics of Li-ion batteries. These include electrical equivalent circuit models [5,6], electro-chemical models [7], and the heat transfer models [8–11]. Moreover, the exploitation of various control methods and strategies for battery management system (BMS) is increasing [12–14].

The effects of stress caused by Li-ion intercalation/deintercalation and the effects of mechanical loads on the cell performance depend on physics ranging from micro-scale to macro-scale. The mechanical fatigue associated with cycling may lead to performance degradation, capacity loss, and eventual failure [15–20]. The effects of prestress and stress evolution on the fading of the battery capacity over time/cycling have been studied through experiments [21,22]. This suggests that mechanical damage in electrodes is driven by the stress and strain fields that are induced from repeated cycling. These studies have provided a useful foundation for developing fracture criteria and characterizing the relationship between mechanical forces and Li-ion intercalation during charge and discharge cycles. In contrast to micro-scale [23–25], the macro-scale stress and strain responses are directly observable and measurable with high accuracy. This suggests that the development of phenomenological battery models can improve the safety and reliability of batteries, and eventually enhance the lifespan and capability of battery cells and packs.

This paper proposes three phenomenological force and swelling models for Li-ion batteries. The 1-D force model includes a nonlinear elastic stiffness to capture the inherent mechanical consequences of Li-ion intercalation; the measured force not only varies with the state of charge significantly but also shows significant nonlinear characteristics with respect to the amplitude of Li-ion intercalation swelling. Moreover, this model recognizes three separate SOC regions delimited by phase transitions. The 1st order relaxation model predicts dynamic (transient) swelling during relaxation periods by using a visco-elastic mechanical relaxation approach. The 3-D swelling model uses a high fidelity finite element model of the battery. This 3-D swelling model accounts for the dependence of an equivalent modulus of elasticity on the state of charge (SOC). Also, the model captures the orthotropic expansion in a direction perpendicular to the electrode surfaces. Therefore, this model can predict the swelling shape on the battery surface over SOC. Validation was accomplished by comparing predicted overall free swelling shapes with measurements at a variety of SOCs. This model is useful to predict the overall swelling shape and the magnitude of swelling at a certain location during actual operational conditions, i.e. constrained conditions. The three models proposed herein can be used to improve existing battery management systems by enabling novel power management schemes as suggested in Ref. [51].

2. Experimental set-up and measurements

This study used a flat-wound type prismatic 5 Ah Li-ion battery cell. Detailed information of the Li-ion battery cell and experimental set-up is available in Refs. [26,27].

In a first experiment, dynamic (transient) free swelling was measured over SOC. The net displacement at the center of the battery (location 13 in Fig. 1) was measured with respect to SOC during discharge using a standard constant current discharging profile. Prior to discharge, the battery was fully charged using a CCCV protocol at 2 A (0.4 C) and rested 3 h at a fixed temperature of 25 °C regulated by a thermal chamber. The voltage was clamped after reaching 4.1 V, at which time it was held until the current tapered to C/100 (50 mA). Discharge was performed down to 3.0 V at 1.0 A (0.2 C). Measurements with a thermocouple showed that the applied current did not cause significant heating without thermal swelling. Also, the battery cell surface remained within 0.1 °C from the regulated 25 °C ambient temperature during discharge. The swelling at 0.2 C therefore allows direct correlations to be made between swelling and Li-ion intercalation in a cell sandwich without significant convolution by thermal expansion. Note that discharge at a low C-rate is essential to obtain pure Li-ion intercalation swelling because discharge at a high C-rate is accompanied by thermal swelling [26].

In a second experiment, the quasi-static free swelling was measured over SOC. In this experiment, the relative expansion in the *z* direction (perpendicular to the electrodes), was measured at 5 locations labeled as 7, 9, 13, 17, and 19 in Fig. 1. Prior to discharge, the battery was fully charged using a CCCV protocol. To obtain the desired SOC ranging from 0% to 100% with 5% increments, the battery was incrementally discharged at a 0.4 C rate (0.4 C current of actual capacity) with an appropriate time (7.5 min). Each incremental discharge was followed by 3 h of rest time to ensure that the system reached equilibrium after each incremental discharge.

In a third experiment, swelling at the fully charged state was measured with sensors sequentially placed at locations 1 to 25 in Fig. 1. This experiment used the same protocol as the first experiment except for the C-rate which was 0.4 C. The swelling at the fully charged state was measured after 3 h of rest time to ensure that thermal swelling due to the increased C-rate did not affect the measurements.

In a fourth experiment, the quasi-static force was measured over SOC with another experimental set-up. Details of the experimental set-up can be found in Ref. [28]. Specifically, forces created in a pack of 3 cells were measured during quasi-static charge and discharge with 3 different initial preloads. The battery pack of interest consists of dozens of batteries connected in series, and plastic spacers are placed between cells [28]. To mimic these conditions, the experimental set-up consists of 3 nominally identical cells connected in series and mechanically sandwiched between two 1-inch thick garolite plates assembled with 4 bolts at their corners. Each bolt was instrumented with a load sensor (LC8150-250-100, USA). The entire set-up was placed inside a thermal chamber (Cincinnati Sub-Zero ZPHS16-3.5-SCT/AC, USA) that controls the desired temperature (25 °C). The plastic spacers between cells allow air to flow between batteries while also constraining the batteries from expanding. Hence, the force measured in this set-up can represent the force induced from the volume change of batteries in an actual battery pack.

3. Phenomenological 1-D force model

3.1. Model description

Two different experiments were carried out to characterize the quasi-static response of the battery cell in Section 2, namely the swelling versus SOC and the force versus SOC at steady state thermal equilibrium. The relationship between force and swelling can be determined when such two experiments are used together (and the curve of force versus swelling is parameterized by the SOC). However, experimental conditions were different; the swelling was measured at free conditions without the plastic spacer and with no preload, while the reaction force was measured at constrained conditions with the plastic spacer and with preload. Hence, governing equations have to be obtained to couple these two experiments considering these differences. These equations are derived in this section. Based on governing equations, equivalent stiffness values are obtained for the case, jellyroll, and spacer. Next, the force is predicted at actual operational/pack conditions.

Three different conditions are considered to describe the experiments. These conditions represent the quasi-static behavior of: (a) the battery cell at free conditions during Li-ion intercalation; (b)