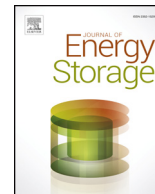




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Journal of Energy Storage

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Inhomogeneous active layer contact loss in a cycled prismatic lithium-ion cell caused by the jelly-roll curvature

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ARTICLE INFO

Keywords:

Curvature
Contact loss
Bending stress
Mechanical ageing
Lithium-ion battery
Diffusion induced stress (DIS)

ABSTRACT

Internal resistance is a key parameter that affects the power, energy, efficiency, lifetime, and safety of a lithium-ion battery. It grows due to chemical and mechanical battery wear during ageing. In this work, the effect of the jelly-roll winding curvature on impedance rise is investigated. NMC electrode samples, harvested from the curved as well as the flat regions of the jelly-roll from cycle-aged and calendar-aged prismatic cells (25 Ah, hard casing) are investigated by electrochemical impedance spectroscopy. After cycling, larger impedance rise is observed at the outer radius (concave) of the curved region compared to the inner radius (convex) or the flat region of the jelly-roll, and the difference increases with a decrease in the jelly-roll radius of curvature, from the cell skin towards the core. To identify the causes behind the observed difference in the impedance rise, investigations at different external compression (0 and 2.5 MPa) and temperature (5 and 25 °C) are performed. The results show that contact loss between the current collector and the active layer is the main source of the difference in impedance rise. Mechanical mechanisms that may cause the contact loss are discussed and design recommendations to mitigate the rise in impedance are given.

1. Introduction

Internal resistance is a key parameter for a lithium-ion battery that influences its power, energy, efficiency, lifetime, and safety. It increases with battery ageing due to various chemical and mechanical degradation mechanisms [1]. The degradation is usually inhomogeneous in a large-format cell [2–4]. As a result, the overall battery performance can be limited by the weaker spots [5]. Hence, the possible reasons that cause the inhomogeneous increase in the internal resistance should be identified and minimized in order to make a durable battery and achieve optimal performance throughout its lifetime. To this end, several mechanisms and effects of mechanical stress that cause inhomogeneous ageing have been studied [3,6,7]. However, the spatially inhomogeneous impedance rise induced by different states of mechanical stress in a prismatic cell geometric structure has not been reported.

Various sources of mechanical stress exist in a prismatic lithium-ion battery at the cell, electrode, and particle level [8–14]. On the cell level, confining the jelly-roll in the prismatic cell casing creates bending stress at the winding curvature which leads to inhomogeneous stress in the

cell [10,12,15]. The hard metal casing also exerts compressive stress on the jelly-roll as it expands during charging [8,9]. Other sources of mechanical stress include solid electrolyte interphase (SEI) layer growth [11], gas evolution [12], and thermal expansion [9]. The composite electrode consisting of the active layer and the current collector is subjected to tensile, compressive, and bending stresses during manufacturing [13]. Compressive stress is applied to the composite electrode during the calendaring process which increases the adhesion between the current collector and the active layer, and cohesion between particles within the active layer as the porosity decreases. The tensile stress that is applied to the composite electrode during manufacturing causes a residual compressive stress along the plane of the active layer. This stress in the active layer is balanced by a tensile stress in the current collector. At the electrode and particle level, diffusion induced stress (DIS) exists as a result of swelling and shrinking caused by varying lithium concentration in the particles and in the active layer [14]. The DIS is influenced by the presence of the substrate current collector which restricts the deformation of the active coating [14]. Additionally, the electrochemical reaction retards the diffusion process and reduces the compressive stress at the interface between the current

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<https://doi.org/10.1016/j.est.2018.09.012>

Received 8 April 2018; Received in revised form 17 September 2018; Accepted 18 September 2018

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collector and the active layer as a result [16]. The magnitude and non-uniformity of the DIS is higher in the transition metal based positive electrode than in the graphite based negative electrode [17]. The bending stress in a curved electrode is influenced by the DIS and continuously varies with the state of charge (SOC). Such combination of stresses may affect various material interfaces, especially the contact between the current collector and the active layer, non-uniformly between the curved and flat regions. Hence investigation of the influence of long-term cell cycling on the inhomogeneous ageing, especially of any impairing of the contact between the current collector and the active electrode layer, is required. No previous study has been reported regarding this issue.

In this work, the effect of jelly-roll curvature on cycling induced impedance rise is investigated by electrochemical impedance spectroscopy (EIS). For this, calendar and cycle aged prismatic cells are opened in an argon-filled glove box. Electrode samples are harvested at curved and flat regions along the jelly-roll winding. EIS measurements are performed using symmetric cells at varying temperatures (5 and 25 °C) and external compression (0 and 2.5 MPa) applied on the cells. Additionally, the electronic conductivity of the composite electrode without the current collector is measured by four-point probe measurements at the different regions of interest.

2. Experimental

A calendar aged and a cycle aged cell used in a previous study [18] were acquired for this investigation. The cells were hard case prismatic $\text{Li}_x\text{Ni}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3}\text{O}_2$ (NMC)/Graphite energy-optimized cells with an initial rated capacity of 25 Ah. The dimensions of the cells were $148 \times 26.5 \times 91$ mm (W x D x H). Cycle-aging of duplicate cells were performed at a constant current 1C charge and 1C discharge rate between 20%–80% state of charge (SOC) until the capacity dropped to 75% of the beginning of life (BOL) capacity. One of the cycled cells was opened in the argon-filled glovebox and analyzed upon opening. The calendar aged cell (3.5 V floating potential, 25 °C) used as a reference for comparison was opened and then stored for 12 months in an argon-filled glovebox (H_2O and $\text{O}_2 < 1$ ppm) before our measurement.

In this investigation, NMC electrode samples (10 mm diameter) were harvested from curved and flat regions of the jelly-roll winding both from the inside concave and the outside convex faces of the double-sided electrode tape, as schematically shown in Fig. 1. Samples were taken from locations at the core as well as from locations closer towards the skin of the cell, i.e. with curvature radii of 9 and 12 mm, respectively. One side of the double-coated electrode tape was removed by gently rubbing it with *N*-Methyl Pyridine (NMP) soaked cotton buds prior to punching out the circular electrode samples. Care was taken to avoid NMP contamination on the opposite side of the electrode. The electrodes samples were analyzed without rinsing with dimethyl carbonate (DMC) [19], in order not to alter the electrode surface or bulk.

The circular electrodes were electrochemically characterized in single electrode layer pouch (polymer coated aluminum foil) cells that were assembled in an argon-filled glovebox. The pouch bag material was dried overnight at 60 °C under vacuum condition in order to remove any residual water before cell assembly. The exposed current collector at the back of the electrode was positioned on top of b-shaped aluminum tabs for external contact, where the circular part was

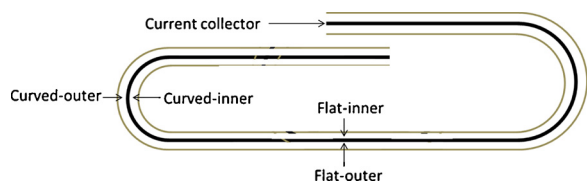


Fig. 1. Schematic of a single composite electrode layer of a prismatic cell with locations where samples are harvested.

supporting the entire electrode sample (Supplementary information, S1). These tabs were used in order to ensure a uniform pressure over the circular electrode. Contact between the b-shaped aluminum tab and the electrode was ensured by compression due to vacuum sealing. In all cells, a tri-layer (PE/PP/PE) layer commercial Celgard separator and commercial electrolyte (Merck LP40) consisting of lithium hexafluorophosphate salt dissolved in ethylene carbonate and diethyl carbonate mixture (1:1 w/w) were used.

Electrode capacity was measured with Li foil as counter electrode between 2.9–4.3 V at a slow cycling rate of C/20 (about 20 h) at 2.5 MPa external compression.

Symmetric NMC/NMC pouch cells were prepared from the electrodes used in the half-cells that had been set to 3.95 V vs. Li/Li^+ , before cell reassembly, and were subsequently used for EIS measurements. EIS was measured in the frequency range of 100 kHz–5 mHz using 5 mV root-mean-square (rms) sinusoidal perturbation at open circuit condition, at 5 °C and 25 °C (± 0.2 °C) in a climate chamber (Friocell MMM, model: MC000921), and with either 0 and 2.5 MPa external compression applied on the cells. The conductivity of the active electrode layer gently delaminated from the current collector was measured using a four-point probe. A Gamry PCI4/750 potentiostat was used for both EIS and conductivity measurements.

3. Results

3.1. Capacity fade of prismatic cells and NMC electrodes

The capacity fade as a function of equivalent cycle and the corresponding discharge curves before and after cycling of the duplicated prismatic cells is shown in Fig. 2a and b, respectively. As shown, the cells cycled at 1C charge/discharge reached capacity retention of around 72%, after about 4000 equivalent cycles in a reproducible manner, which is an acceptable degradation rate for heavy-duty plug-in electric vehicles (PHEVs). The calendar aged cell lost negligible capacity during the storage time. The low-rate capacity measurements of the NMC samples harvested from the cycled cell show approximately

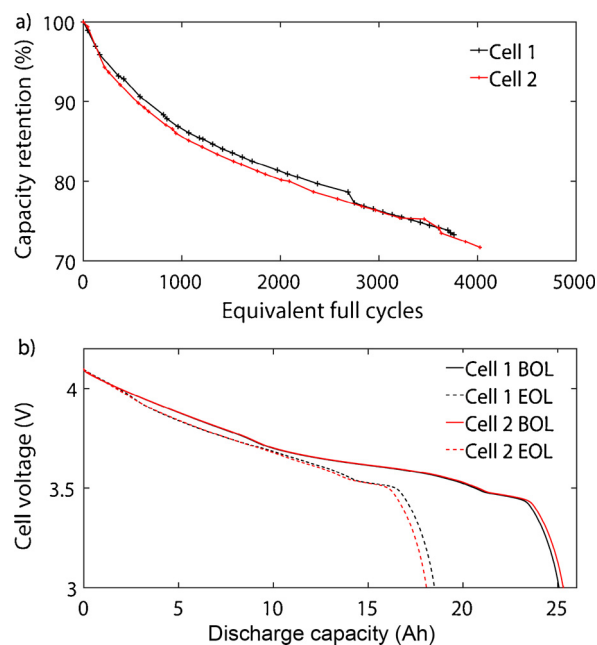


Fig. 2. a) Capacity fade as a function of 1C equivalent cycle for two identical cells cycled with +1C/-1C (where a 1C equivalent cycle corresponds to 50 Ah capacity throughput). b) Discharge curves of the prismatic cells at the beginning of life (BOL) and end of life (EOL). Discharge capacity is measured at C/4 rate in a and b.

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