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Influence of cell design on impedance characteristics of cylindrical lithium-ion cells: A model-based assessment from electrode to cell level

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ABSTRACT

Electrochemical impedance spectroscopy (EIS) is a fast, inexpensive and non-destructive tool for identification of electrical and electrochemical loss processes of battery cells in a wide range of frequencies. Among those, particularly interface processes like the migration of Li species through surface films and the charge transfer reaction are easy accessible by EIS. One of the main drawbacks of its application to commercial cells is the limitation to the full cell spectrum due to the absence of an appropriate reference potential to separate cathodic and anodic contributions. Further, the impedance response of cell geometries such as cylindrical cells is characterized by a complex superposition of contributions arising from electrochemistry and cell design. In this study, we address these issues by a planar transmission line model allowing to reconstruct the impedance response of cylindrical cells by impedance data obtained from three electrode measurements on extracted electrodes and geometrical information obtained from microscopical cross sections. The reconstruction results indicate a substantial contribution from the spiral geometry of the current collectors and the position of the current collector tabs along the jelly roll. Furthermore, the impedance response is significantly determined by the power/energy design of the cell, for instance by the ratio of electrode areas and thicknesses. Overall, the proposed methodology presents a sound modeling basis for a deeper understanding of the impedance response of cylindrical cells and a tool for cell design optimization from electrode to cell level.
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1. Introduction

In recent years, electrochemical impedance spectroscopy (EIS) has become an inevitable tool for characterization and modeling of lithium-ion cells [1,2]. It has been widely applied to the characterization of new materials [3,4] and systems [5–7] as well as to the study of degradation modes in state of the art systems [8–13]. Furthermore, EIS has been frequently used to derive model parameters for either equivalent circuit or more sophisticated models based on porous electrode theory [14–17]. The EIS response of lithium-ion cells usually exhibits a great variety of loss processes spread over a wide frequency range. In this manuscript, the expression *loss process* is used to describe processes leading to considerable overpotentials during operation of the cell, thus to a deviation between open circuit and terminal voltage. Among those are the transport of charged species through the electrode and electrolyte domain, e.g. Li migration/diffusion as well as charge transfer reactions at the electrode/electrolyte interface. For commercial cells, temporal and spatial separation of these loss processes is a challenging task. First, due to the fact that commercial cells are two electrode systems, cathodic and anodic processes are superimposed in the full cell spectrum. Second, processes associated to either half-cell might take place at comparable timescales impeding unambiguous identification. Finally, the loss processes might additionally be distributed among different positions inside the cell implying a complex dependency from cell geometry and design. In [18], the impedance response of a commercial 18650 cell has been addressed by Distribution of Relaxation Times (DRT) analysis applied on impedance spectra obtained in three electrode measurements of extracted electrodes from the original cell. The most important outcome of this study was that the spectra recorded for the original and the reconstructed full cell came close when both were scaled by the equivalent active electrode areas. Especially in the low frequency

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region, both spectra nearly coincided indicating that mapping of the low frequency behavior might be feasible by a simple scaling operation. For the high frequency region, significant deviations between both cell formats were observed. The authors ascribed these deviations to inductive contributions of the current collectors and electrical contacts in the spirally wound cell. However, they were not able to prove their assumptions due to the absence of an appropriate modeling concept. Generally, only few studies dealing with the inductive behavior of lithium-ion cells are reported in literature so far [3,19,20]. In context of equivalent circuit based approaches, the influence of inductive contributions is usually treated very simplified by either an inductance or parallel resistor-inductance element in series to the equivalent circuit representing the electrochemical loss processes [21–23]. Among the very few studies examining the physical nature of the inductive contributions, Laman et al. recognized the significance of the spirally wound geometry of the current collectors for the inductive characteristics of cylindrical cells [19]. By modifying such a cell by a third current collector tab connected to the innermost end of the anodic current collector, they were able to elaborate the impact of current flow direction on the impedance response. In case of parallel current flow in cathodic and anodic current collector, they found an amplification of the high frequency tail in the fourth quadrant of the Nyquist plane, whereas in case of anti-parallel current flow the inductive contributions of the collectors were observed to cancel each other out. The authors explained this observation by the effective current amplitude in two adjacent conductor pairs which becomes zero in case of anti-parallel current flow. Since the inductive loss is directly correlated to the derivation of the current, the cancellation is projected by the mutual interaction between the current collectors. The influence of the tab pattern and current direction on the inductive characteristics of cylindrical cells has recently been confirmed by Osswald et al. [20]. In that study, the authors tried to quantify the inductive contributions of the current collectors by systematic impedance measurements at different tab positions along the current collectors accessed in a modified 26650 cell. They found a close correlation between the amount of traversed windings and the maximal imaginary value $Z''(\omega)$ measured in the fourth quadrant depending on the addressed tab.

Summing up, the works introduced above highlight the significance of the cell design in context of the global impedance behavior of cylindrical cells. In this study, we aim to extend the understanding of the influence of cell design by a combined experimental and model-based approach to reconstruct the impedance response of a typical commercial $LiFePO_4$/graphite 18650 cell. Therefore, an equivalent circuit model comprising domains for the electrical and electrochemical loss processes occurring in the cell is developed. The electrochemical domain is parametrized from three electrode impedance data acquired from extracted electrodes of the original cell. The electrical domain is parametrized from information on the spiral geometry of the 18650 cell (e.g. winding numbers) obtained from microscopical cross sections and material parameters of the current collector materials. In detail, the reconstruction implies the following steps:

(1) Scaling of three electrode impedance data according to the ratio of active electrode areas of experimental and commercial cell
(2) Modulation of the scaled impedance by the resistive-inductive contributions of the current collectors
(3) Correction of the ohmic offset traced back to different electrolyte volumes

In Section 2, the model structure used for reconstruction is introduced based on the loss processes expected to be rate-limiting for cylindrical cell formats. Afterwards, Section 3 deals with the model parametrization, i.e. how the crucial model parameters are extracted from the experimental data set for the studied cell. Subsequently, the so found model structure is validated against the impedance response of the original 18650 cell and applied to study the influence of tab pattern and power design on the impedance characteristics of the cylindrical cell (Section 4). It will be shown that the impedance response of the original cell can be reproduced with high fidelity and that the influence of tab pattern predicted by the model is in accordance with experimental results found in [20]. Additionally, the impedance characteristics turns out to be significantly determined by the energy/power design of the cell, namely by cell design parameters such as electrode thicknesses and active electrode areas.

2. Model structure

The model used for reconstruction consists of an electrochemical domain comprising the loss processes associated to Li transport, charge transfer and diffusion of Li species and an electrical domain representing losses correlated to the conduction of electrons through the cell contacts and current collectors. The model structure is developed to cover the loss processes of both domains with respect to a typical cylindrical cell geometry shown in Fig. 1(a). For such a geometry, cathode and anode sheets are double-sided coated and the current collectors are nested to ensure a face-to-face alignment of the active materials of both electrodes. In our model, the loss processes of the electrochemical domain are covered by δZ_{ec}, whereas the loss processes of the electrical domain are represented by δZ_{el}. Since both contributions

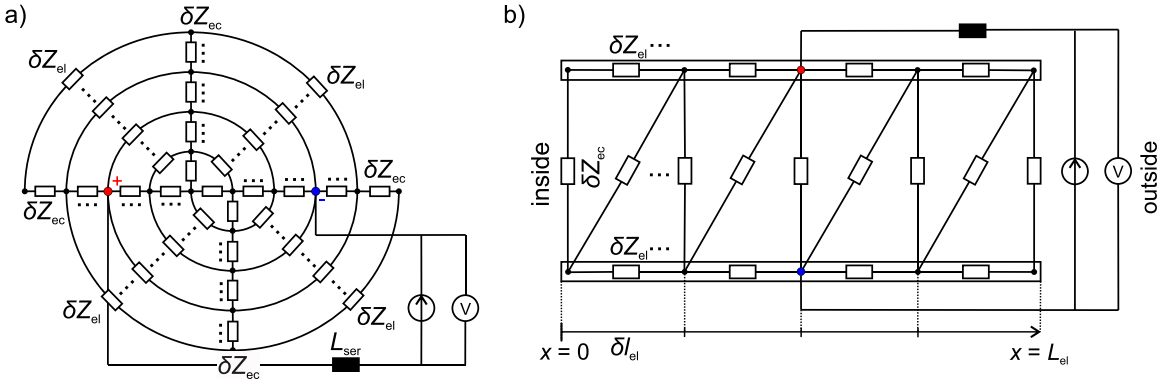

Fig. 1. Equivalent circuit structures to simulate the impedance spectrum of the studied cylindrical cell format: (a) spiral geometry (b) planar transmission line. The impedance behavior results from a concatenation of an electrochemical domain (depicted by the generalized impedance elements δZ_{ec}), an electrical domain (depicted by δZ_{el}) and a serial inductance to map the influence of cabling/contacts.

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