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Current density distribution in cylindrical Li-Ion cells during impedance measurements

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HIGHLIGHTS

- Modified commercial cylindrical cell with 4 current tabs for anode and cathode.
- Effect of different tab pattern and cell design on the impedance spectra of the cell.
- AC current density distribution depending on applied current frequency.
- AC current density distribution depending on cell temperature.

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ABSTRACT

In this work, modified commercial cylindrical lithium-ion cells with multiple separate current tabs are used to analyze the influence of tab pattern, frequency and temperature on electrochemical impedance spectroscopy. In a first step, the effect of different current tab arrangements on the impedance spectra is analyzed and possible electrochemical causes are discussed. In a second step, one terminal is used to apply a sinusoidal current while the other terminals are used to monitor the local potential distribution at different positions along the electrodes of the cell. It is observed that the characteristic decay of the voltage amplitude along the electrode changes non-linearly with frequency, where high-frequent currents experience a stronger attenuation along the current collector than low-frequent currents.

In further experiments, the decay characteristic is controlled by the cell temperature, driven by the increasing resistance of the current collector and the enhanced kinetic and transport properties of the active material and electrolyte. Measurements indicate that the ac current distribution depends strongly on the frequency and the temperature. In this context, the challenges for electrochemical impedance spectroscopy as cell diagnostic technique for commercial cells are discussed.

1. Introduction

For more than a decade, batteries have become one of the main research objects to tackle the problem of air pollution caused by fossil fuels for transportation and energy production. Intense research and development effort has been devoted to lithium ion batteries (LIB), as they were identified as one of the most promising solution. Nowadays, due to the steady increase in power density, safety and reliability, LIB are used not only for consumer electronics anymore, but also stationary electrical energy storage as well as electric and hybrid vehicles. As diverse these applications are, as diverse are the different types of cells used for them. Most common designs are cylindrical cells, prismatic hard-case cells and pouch cells [1–3]. All of those cell types have in common that they can be monitored by electrochemical

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impedance spectroscopy (EIS) to determine their inner states [4]. The sensitivities of EIS measurements to the state of charge (SOC) [5,6], state of health (SOH) [7–9], cell temperature [10,11] and short-term previous charge and discharge history [12] are discussed in literature. Since commercial cells have one single electrical terminal, EIS measurements for diagnostic issues face the drawback that different impedance contributions of different locations at the current collectors are superimposed at the terminal. On the contrary, studies of current density distributions at typical cycling conditions show an inhomogeneous operation along the electrodes [13–15]. The distribution of a sinusoidal current through the electrodes is not discussed in present literature. Thus, it can be assumed that an alternating current (ac) operation also leads to inhomogeneous current distribution through the electrode. As a consequence, EIS measurements would provide impedance data from undefined fractions of the electrodes. Especially in the context of monitoring aging in commercial cells, the impact of local inhomogeneities on the measured spectra have to be considered. This paper discusses experimental investigations of the current path evoked by an ac excitation current in dependence of different tab patterns, frequencies and temperature. Therefore, a commercial 26650 type lithium-ion cell with a lithium iron phosphate (LFP) cathode and a graphite anode was modified to allow for local measurements of the electrode potentials [15,16]. The experimental details and testing scenarios are described, followed by the discussion of the experimental data involving both, the time and frequency domain.

2. Experimental work

In this work, measurements were performed on modified commercial 26650 cylindrical cells. The original cell has a nominal capacity of 2.5 Ah with LFP as cathode and graphite as anode material. Prior to modification, the cell has four positive and four negative current tabs, distributed equidistantly along the electrodes and welded together to the respective part of the cell casing. By modification, we have disconnected these welding joints and separated the single current tabs, allowing an individual access to each of the eight current tabs, as shown in Fig. 1 a). For more details on the modification process and cell parameters, please refer to our previous work [15]. Based on the tab arrangement A_m and C_n, with $m, n \in \{1;4\}$ used for the measurement, we define a terminal T_{mn} with its respective terminal voltage U_{Tmn} and impedance Z_{Tmn}, respectively. A cylindrical cell is used with T_{11} (A_1–C_1) as the outermost and T_{44} (A_4–C_4) as the innermost terminal. To help visualizing the cell, Fig. 1 depicts a sketch of the modified cell (b) together with pictures of the current tabs of an opened cell at anode (c) and cathode side (d). After resealing, the modified cells can be operated outside of the glovebox. All tests were performed within the first 20 cycles. As shown in our previous work, modified cells age during cycling for 200 cycles similar to unmodified cells and show a capacity fade of 10% after 1000 cycles [15]. Therefore, we concluded that no electrochemical change will occur during our measurements.

To charge and discharge the cells, a BaSyTec CTS was used. To adjust the SOC of the cells, a current of 0.1C (250 mA) was applied using two positive current tabs (C_1 and C_3) and two negative current tabs (A_2 and A_4). This asymmetrical operation mode is assumed to reduce local SOC inhomogeneities and, therefore, to reduce the time until the cell reaches an equilibrated state, a prerequisite for accurate impedance measurements [17,18]. For impedance measurements, a Bio-Logic SAS VMP-3 has been used and different excitation currents between 30 mA and 200 mA were applied. The

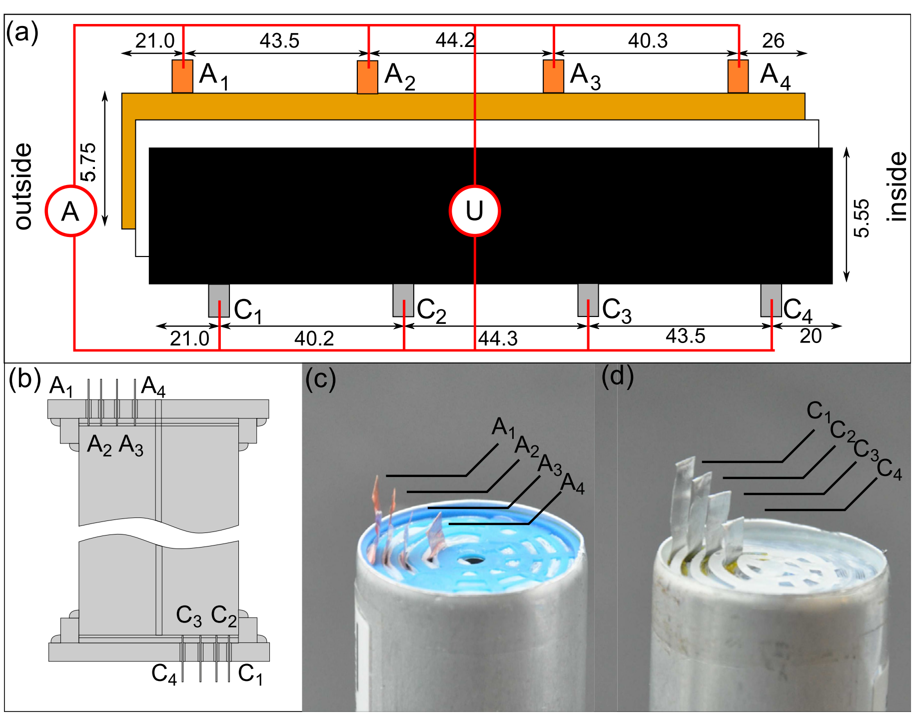

Fig. 1. Drawing of the cell with dimensions in cm (a) and schematic cross-section after the modification process (b) [15]. The pictures show the opened cell with four current tabs on anode (c) and cathode (d).