



Multi-scale investigation of thickness changes in a commercial pouch type lithium-ion battery



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ARTICLE INFO

Article history:

Received 14 November 2015

Received in revised form 18 January 2016

Accepted 19 January 2016

Available online 15 February 2016

Keywords:

Lithium-ion battery

Swelling

Volume change

Strain

3D scanning

ABSTRACT

The thickness change of a commercial pouch type lithium-ion battery with a graphite anode and a LiCoO₂ cathode is investigated at multiple scales. The fraction of anode and cathode expansion in the total thickness change of the battery is derived by dilatometry and 1D battery displacement measurements. The LiCoO₂ cathode shows a thickness change of 1.8% and the graphite anode expands by 5.2% within the battery's operating range. Both anode and cathode expansion sum up in an overall thickness change of 2.4% of the pouch battery including the nonactive material. The changes in the differential displacement curve of the battery are related to phase transitions in the graphite anode, which correlate with differential potential curves. Furthermore, the thickness change of the battery is investigated by structured-light 3D scanning. Using this contactless technique, the spatial distribution of the thickness change and the impact of the pouch packaging is visualized. The pouch foil shows to have only a small influence on the displacement distribution. In sum, the results of the applied techniques at different scales correlate well and provide a better understanding of the intercalation-induced volumetric changes in lithium-ion batteries.

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1. Introduction

Lithium-ion secondary batteries are applied in a wide range of mobile devices such as cell phones, laptops and electric vehicles as a result of their high energy and power density. Intercalation processes of lithium-ions cause volumetric changes of the active material integrated inside these batteries during charging and discharging operation [1,2]. This results either in thickness changes of unconstrained batteries or force exertion when the expansion is restricted [3,4]. For a better understanding of the underlying mechanisms, the volumetric changes during charging and discharging are investigated for a commercial pouch type battery at multiple scales to correlate directly the results of the adjacent scales.

In previous publications, the intercalation-induced volumetric changes of batteries and single electrode materials were investigated at various dimensions [2,5–8]. To study the expansion of a

single electrode layer due to intercalation, there are two reported techniques. The first one is to use a special dilatometer cell. Its structural design suppresses the expansion of the counter electrode, and the expansion of the working electrode can be measured by a displacement sensor [9]. Dilatometric investigations are also reported in Refs. [6,10–12], but the focus was set on anode materials only. Another method to measure the expansion of a single electrode is the assembly of a pouch battery with a Li[Li_{1/3}Ti_{5/3}]O₄ (LTO) anode, which generally shows negligible expansion during lithiation [13]. The thickness change of such batteries was measured by a precision displacement sensor to study the expansion of different cathode materials by Nagayama et al. [14]. To the authors' knowledge, no separate single electrode expansion was measured for electrodes of a commercial battery yet.

At the battery scale, various studies report on the thickness change of commercial batteries during charging and discharging [3,7,15,16]. In the following, those experiments will be named 1D battery displacement measurement, as the displacement is detected in one direction. The displacement of the batteries was measured from one side [7,15,16] or from both sides [3,17]. The derivative of the potential and displacement curves was investigated with respect to capacity to correlate variations in the displacement curve to phase changes in the electrode materials

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[17–19]. It was proposed that the cathode contributes to the thickness change of the battery as well, but the percentage and the direction could not be quantified due to the lack of dilatometric experiments.

Only few publications investigated the distribution of the thickness change in lithium-ion batteries [8,17,20]. Those experiments will be called 3D battery scale, as the displacement of the entire battery geometry is detected. Siegel et al. [20] used neutron imaging to detect the swelling of single electrode layers in a cross section of a battery and reported a larger thickness increase of the active material at the housing center compared to the edges. Oh et al. [17] measured the thickness change of an automotive hardcase battery at different positions with precise displacement sensors. The thickness change was found to be highly dependent on the measurement location with the largest expansion occurring at the center of the battery surface due to the high stiffness of the aluminum housing. As thermal expansion occurs when the battery heats up during operation, temperature gradients are likely to result in variations of the thickness change on the cell surface when a high current is applied [21]. Leung et al. [8] used 3D digital image correlation (DIC) to study the strains in width, length and thickness (x , y and z) direction during charging and discharging of a commercial battery with nickel-manganese-cobalt oxide (NMC) cathode and graphite anode. The pouch foil was removed before starting the experiment to eliminate the influence of the housing on the displacement. The expansion in thickness direction (z -direction) was unevenly distributed over the battery surface with a significantly larger thickness increase at the vicinity of the jelly roll structure compared to the center of the battery, which may be possible because the jelly roll structure was free to expand in those regions. Additionally, the displacement in x - and y -direction was found to be negligible compared to the displacement in z -direction.

However, all aforementioned investigations at different scales have not been performed with the same battery, which makes it difficult to adopt the results on other battery configurations. In the present work, the thickness change of a commercial pouch type battery with a graphite anode and a LiCoO_2 (LCO) cathode was investigated at single electrode scale, 1D battery scale and 3D battery scale to get a better understanding of the underlying mechanisms and their impact on effects at the adjacent scales.

2. Test methods

A commercial pouch type battery (EnerTech SPB655060) with a graphite anode and a LCO cathode is chosen as both LCO and graphite are expected to expand during battery charging, which increases the measurable displacement signal [1,22]. A battery designed for high energy content is chosen because of its larger coating thickness which is expected to result in a higher absolute thickness change compared to a high power configuration. The

battery is disassembled to gain information on the interior structure. The battery consists of 17 double-coated anode layers with a coating thickness of ca. $77 \mu\text{m}$, 16 double-coated and 2 single-coated cathode layers with a coating thickness of ca. $68 \mu\text{m}$. The polymer separator with a thickness of ca. $25 \mu\text{m}$ is wound around the electrode sheets. The obtained information on the interior structure of the battery is summarized in Fig. 1.

Detailed information on the battery is provided in Table 1. The ingredients of the liquid electrolyte are determined by Gas Chromatography–Mass Spectrometry (GC–MS). Porosities of the cathode and the anode are measured by mercury porosimetry. The active material volume fractions of both electrodes are estimated assuming a nonactive material content (binder and conductive filler) of 5% in the cathode and 7% in the anode.

The main cathode active material is LCO, which is doped with Nickel (Ni) and Manganese (Mn). All batteries are cycled for 15 cycles with 0.5C charge and discharge current prior to any experiment in order to reduce the higher irreversible thickness change compared to the steady operating condition in the first cycles caused by delayed formation processes [23]. Fig. 2 illustrates the applied techniques used to study the intercalation-induced thickness change at the different scales. The objective for using the specific techniques is indicated at the lower section in Fig. 2. The expansion of single anode and cathode layers is investigated by dilatometry, which is shown in Fig. 2a. Together with 1D displacement measurements conducted on the pouch battery, the fractions of the anode and cathode expansion contributing to the overall thickness change of the battery are derived (Fig. 2b). The displacement distribution on the pouch surface of the battery is investigated by enhancing the high resolution measurement of the thickness change at the center of the battery gained by 1D displacement measurements with 3D scanning data of the battery geometry (Fig. 2c).

2.1. Dilatometry

Dilatometric experiments provide information on the thickness change of a single electrode during lithiation and delithiation, as the separator is fixed in position (see Fig. 2a). The pouch battery is discharged to 2.8 V and disassembled inside a glovebox in argon atmosphere, where the dilatometer is assembled as well. The working electrode operates under a constant mechanical load of 1 N. To measure the single electrode potential profiles during normal cycling, 2016 type coin cells are built with 16 mm diameter electrodes punched out from the electrode sheets and a lithium metal counter electrode. The dilatometer cell requires 10 mm diameter for the working electrode.

2.1.1. Dilatometry of LCO

As shown in Fig. 1, the battery has two single-coated cathode layers at the top and the bottom, which are used for the

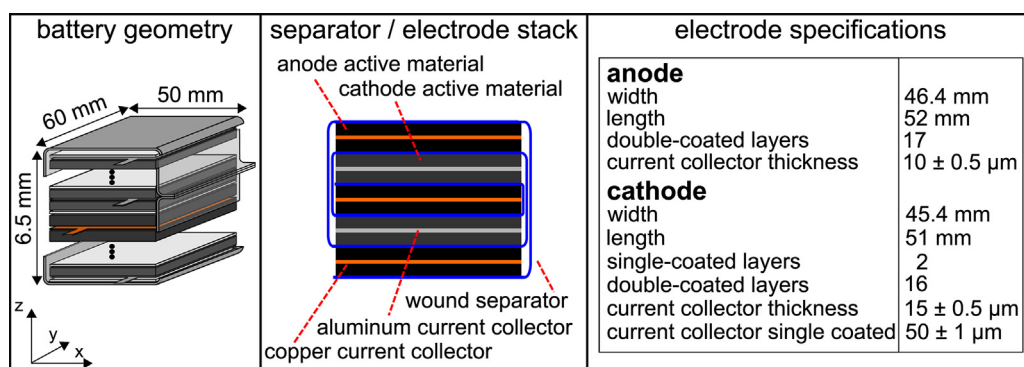


Fig. 1. Mechanical properties and specifications of the investigated battery.

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