

In-operando temperature measurement across the interfaces of a lithium-ion battery cell



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ARTICLE INFO

Article history:

Received 1 November 2012

Received in revised form 11 July 2013

Accepted 16 August 2013

Available online 30 August 2013

Keywords:

Lithium-ion battery cells

Temperature distribution

Heat sources

ABSTRACT

In this work an experimental setup for *in-operando* temperature measurements across the interfaces anode – separator/electrolyte – cathode of a lithium-ion battery cell is developed to get a better understanding of the heat generating mechanisms. The results show differences in the heat evolution rates of the anode, the separator and the cathode according to the electrochemical reactions, the state of charge, the overvoltage and the electric current density. The LiCoO₂ cathode was identified as the most decisive component for the heat evolution in the investigated battery stack. Changes of the ohmic resistance and the entropy of LiCoO₂ with the state of charge were reflected by the temperature measurements.

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1. Introduction

Actually, lithium ion batteries are well established as energy storage devices in portable electronics. Due to their high energy density and high voltage, they are predestinated for implementation in high power applications e.g. electric vehicles. Nevertheless, these advantages are faced with a high temperature sensibility and safety hazards caused by the heat evolution during rapid charge and discharge. Current solutions of these problems base on active thermal management. By adding supplementary components to the battery system, its weight increases and overall energy density decreases. Consequently, lithium ion technology becomes less efficient. An alternative approach could be the development of electrode and separator materials which reduce the need of a thermal management. However, such a pointed material development requires a detailed understanding of the heat generating processes as well as the relationship between operation conditions, heat evolution mechanisms, material properties and structure on the micro and macro scale. Regarding the heat evolution in commercial lithium-ion batteries the literature is commonly dealing with experimental investigations and theoretical modeling on the influence of the operating conditions, the geometry, the ambient temperature, the active materials, etc. However, in these publications usually only integral measurement techniques and a homogenization of the material properties and mechanisms are applied [1–3]. In reality, heat generating mechanisms distinctly differ between the

individual battery components. This point has to be taken into consideration for understanding the heat generation in a battery cell.

Therefore, it is necessary to develop an experimental setup for *in-operando* and in situ temperature measurements in a lithium-ion battery cell. Such a setup is realized in the form of a special electrochemical cell. The cell design must meet the following requirements:

- The measurement of the temperature *in-operando* as well as the electric current and the cell voltage.
- A one dimensional spatial resolution of the temperature measurement in the thickness range of the electrodes.
- A cell body that is adiabatic and sealed.

This work presents a recently developed appropriate cell for *in-operando* temperature measurements across the interfaces anode – separator/electrolyte – cathode and first results obtained from measurements at an electrochemical reference system. Thus, the measurements offer experimental data relevant to the thermodynamic properties of the reference cell and include information about transport processes within the electrodes and the electrolyte.

2. Experimental

2.1. Experimental setup

The measurements were carried out in the electrochemical cell schematically shown in Fig. 1a. It is a two electrode cell with parallel arrangement of anode, separator and cathode within a polyetheretherketone (PEEK) housing. The used PEEK is a organic

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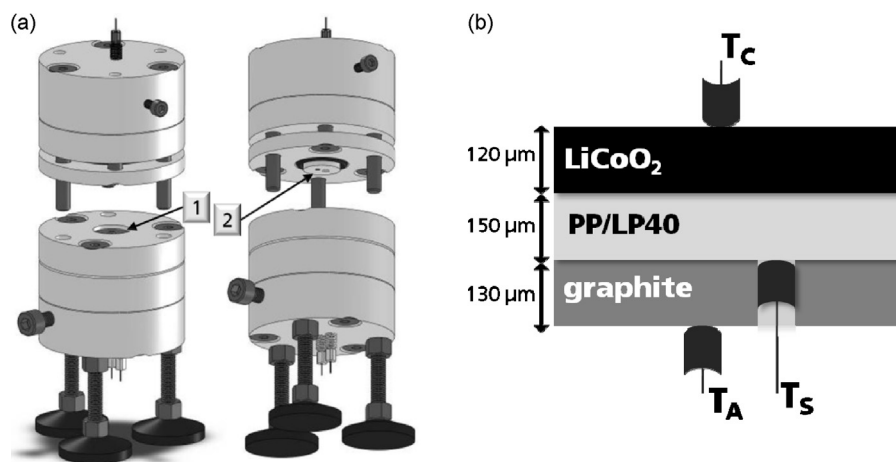


Fig. 1. (a) PEEK housing with sample chamber (1) and spring mounted stamp (2). (b) Positioning of thermocouples T_c (cathode), T_s (separator/electrolyte) and T_a (anode).

polymer thermoplastic made from 4,4'-difluorobenzophenone and the disodium salt of hydroquinone. The specific heat capacity is approximately $0.32 \text{ W m}^{-1} \text{ K}^{-1}$ and the glass temperature is about 143°C . The stack system of anode, separator/electrolyte and cathode is spring mounted to guarantee a reproducible contact pressure between the components. Anode and cathode were connected from the backside by electrical contacts implemented into the cell body. All connections and critical spots were sealed twice with gaskets.

The spatial resolution of temperature measurement was realized by a specific arrangement of three thermocouples as schematically drawn in Fig. 1b. Two thermocouples are pressed against the backside of the electrodes. The third one penetrates an electrode and is pressed against the separator. All thermocouples are spring mounted for a reproducible positioning. The thermocouples used in the cell are of type K, what offers a temperature resolution of 5 mK in combination with the applied measurement device. The cell was placed in a thermally insulating container during the experiments to minimize an influence by fluctuations of the ambient temperature.

2.2. Experimental details

Commercial electrode foils (MTI) were applied in the cell. The lithium cobalt oxide (LiCoO_2) cathode has a capacity of 145 mA h g^{-1} and the graphite anode (C) a nominal capacity of 330 mA h g^{-1} . In both cases the electrodes are composites containing active material, conductive additives and binder on a current collector. Commercially available LP40 (1 M lithium hexafluorophosphate (LiPF_6) in ethylene carbonate: diethyl carbonate (1:1 w/w), Merck) was used as electrolyte. The separator was a polypropylene non-woven fabric (Viledon® Separators, Freudenberg). The assembly of the cell was carried out in an argon filled glove box (Jacomex). The electrochemical experiments included constant current charge–discharge cycles in the cell voltage range of 2.9–4.3 V. Therefore a potentiostat/galvanostat with frequency response analyser Reference 3000™ (Gamry) was used. The temperature measurements were carried out as described above in parallel to the galvanostatic charge–discharge experiments.

3. Results and discussion

Fig. 2 shows a section of charge–discharge experiments that were carried out within the developed cell. The experiments combine constant current charging and discharging of the battery cell with measurements of the open circuit voltage. The applied current density amounts to 1.5 mA cm^{-2} . This current density leads to

a consumed charge of 10.8 C cm^{-2} during a charging cycle of 2 h. In the battery community such a charge rate is usually referred as 0.5 C, whereby C represents the theoretical capacity of the LiCoO_2 cathode. However, the actual capacity of the cell is lower than its nominal value. This becomes obvious in the shortened charging times until the cell voltage reaches its prescribed limitations. This might be due to the effect of an overvoltage evolving during galvanostatic load.

Fig. 3 shows the temperature measurements according to the charge–discharge experiment from Fig. 2. It includes the course of the ambient temperature as well as the temperature response at the

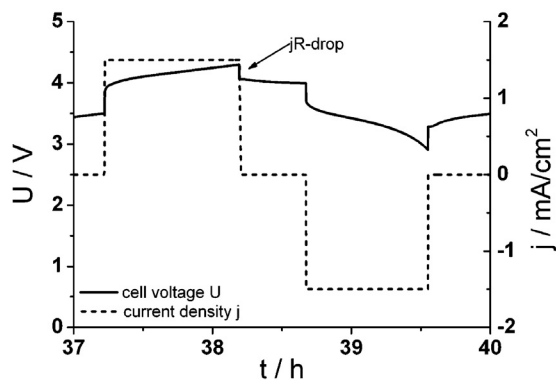


Fig. 2. Extract of the galvanostatic charge–discharge regime of the *in-operando* temperature cell ($j = 1.5 \text{ mA cm}^{-2}$).

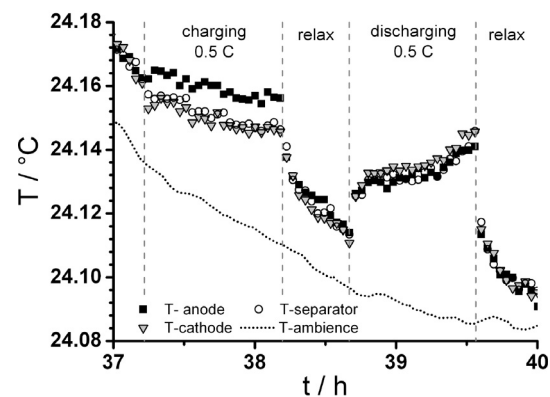


Fig. 3. Course of the ambient temperature and the temperatures at the cathode, separator and anode during battery cycling.

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