X-ray computer tomography investigations on solid-state lithium-ion battery system

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Abstract

Forschungszentrum Jülich (FZJ) is one of Europe's largest interdisciplinary research centers, working in many scientific areas. The Institute of Energy and Climate – Fundamental Electrochemistry (IEK-9) focuses on the development of materials for electrochemical storage and energy conversion. The Central Institute for Engineering, Electronics and Analytics – Engineering and Technology (ZEA-1) has capabilities in mechanical and electronics development, as well as analytical services. The development of measurement and testing methods is one of the ZEA-1 focus areas.

Solid-state lithium-ion batteries are an emerging topic in the field of battery research because they promise to solve safety issues in current lithium-ion batteries [1,2]. One of the critical problems for this system is volume expansion/contraction of active materials in anode and cathode, which may impede their direct incorporation into solid-state batteries. Recently, we have developed the phosphate solid-state lithium-ion battery prototypes, in which the volume changes during charge and discharge in the materials are minimal according to theory [3,4]. In order to understand and to optimize the solid-state battery systems, we study the organic-inorganic composite electrolytes and sintered ceramic electrolyte to gain information on the mechanical stability limits.

The scientific problem under this study is experimental investigation of cracks formation and evolution in a bulk-type solid-state battery during different loads and at different electrochemical stages. It appears that one suitable approach is non-destructive method, the X-ray computed tomography (CT). With the help of CT, we are able to detect the issue of possible cracks propagation and their characterization in the solid electrolyte material. The morphology of the interfaces of battery components are also the topic of current studies. This could help to find the optimal correlation between mechanical and electrochemical properties for the practical application of solid-state battery.

To carry out the measurement noted above, the newly upgraded industrial X-ray CT equipment at the Institute ZEA-1 was applied [5,6]. The X-ray CT system shown in Figure 1 has been specifically designed in cooperation with the Development Center X-ray Technology (EZRT Saarbrücken) to meet the requirements of various fields of
research at FZJ (especially in the field of high-performance materials for energy conversion, storage, metallurgy). The system includes a high-precision (granite base with air bearings) set-up with enhanced geometric configurability within 3D space, both micro- and nano-focus tubes, giving the resolution down to 2-3 μm. Various detectors with up to 4096² pixels and 40 cm² are available for different applications. As result, the X-ray CT configuration allows to obtain 3D images of the sample-objects of around 50 cm diameter with μm-resolution [7,8].

![Fig. 1. X-ray CT set up at ZEA-1 of Forschungszentrum Jülich GmbH. Left grey part is the mounting system for the micro-focus X-ray Tube (exchanhable with nano-focus X-ray tube), table in the middle is the mounting and motion system for the sample-holder, right blue-back part is detector system.](image)

In the present work, a monolithic bulk-type all solid state lithium ion battery based on a phosphate backbone with LiTi2(PO4)3 (LTP) anode, Li1.3Al0.3Ti1.7(PO4)3 (LATP) as solid electrolyte (SE) and Li3V2(PO4)3 (LVP) as cathode is demonstrated. All samples studied are analogous and with an overall thickness larger than 500 μm.

As it was checked after synthesis, the phosphate backbone electrodes and SE ensure the compatibility on both structural and electrochemical aspects. The crucial interfacial issues are circumvented without high temperature treatment of the battery. High mechanical and (electro-) chemical stabilities are achieved by tailored composite electrodes and a carefully designed architecture of the battery. The ASLiB shows excellent properties in terms of cycleability and power density.

Two CT images of particular battery prototypes before and after electrochemical charge/discharge test (0.1 C, 50 cycles in the voltage range of 0.5-2.2 V) are presented in Fig. 2 as example. It is clearly seen that all three layers of the prototype before cycling are homogeneous with different densities (Fig. 2a). After cycling the cracks, which are perpendicular to the surface, appear over the sample of prototype (Fig. 2b).
Fig. 2. CT images of battery prototypes before (a) and after cycling (b)

The preliminary ex-situ studies of first prototypes of cycled batteries by X-ray CT have indeed found clearly the appearance of cracks in the volume. In-situ measurements during cycling will be performed. The visualized correlations of electrochemical properties, microstructure and mechanical characteristics of bulk-type solid-state batteries are presented and discussed.