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Short communication

X-ray micro tomography of three-dimensional embroidered current collectors for lithium-ion batteries

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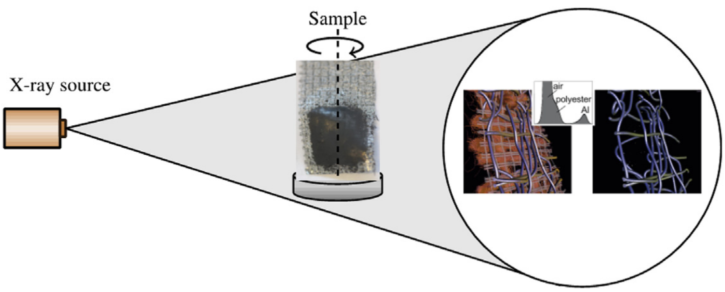
HIGHLIGHTS

- Technical embroidery allows the manufacturing of custom 3D current collectors.
- Embroidered 3D porous current collectors are promising for use in Li-ion batteries.
- µCT permits the three-dimensional characterisation of embroidered electrodes.
- µCT reports elements, distribution and morphology of embroidered electrodes.
- µCT analyses complement the electrochemical measurements of embroidered electrodes.

GRAPHICAL ABSTRACT



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ABSTRACT

X-ray micro computed tomography (µCT) is used for the morphological characterization of three-dimensional (3D) embroidered current collectors for $LiFePO_4$ batteries. A conventional 2D planar configuration (aluminium foil as current collector) and two types of 3D embroidered configurations were compared. The µCT images were used to identify the different components in each configuration as well as to evaluate the corresponding volume fractions, porosities, mass fractions and apparent densities. Microtomographic scans complement analysis of typical galvanostatic charge/discharge cycles and confirm the suitability of 3D embroidered current collectors for their use in lithium-ion batteries.

1. Introduction

Lithium ion (Li-ion) batteries are used in a wide range of applications, from electric vehicles to small electrical devices and in medical tools. Significant effort has been devoted to improving the performance of Li-ion batteries, especially on meeting the energy

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and power densities, requirements in the mentioned applications [1]. Three-dimensional (3D) porous structures are found to be a solution to achieve both high energy and power densities for a given footprint area [2–4]. The loading of high amounts of active material (required for high energy densities) on conventional 2D planar configurations present problems during the coating process (leading to delamination), inhibit electrolyte penetration, and thus increase cell impedance and result in power limitation [5,6].

3D embroidered porous current collectors constructed using technical embroidery offer the opportunity to customize and control the design of high electrical conductive 3D structures. A large variety of metal yarns and threads may be used in diverse layouts, resulting in more flexible and lighter structures [7–9]. In addition, the production technology is easily scalable. For a better understanding of the role of new 3D architectures on the battery performance, a proper morphological characterisation is required. The determination of geometrical features, for instance the spatial distribution of the electrode material, its contact with the current collector, the porosity and mass fractions of the different elements, is essential to optimize 3D porous structures and achieve better electrode kinetics and mass transport of lithium ions in the electrolyte [10,11].

X-ray micro computed tomography (μCT) allows for non-destructive identification of 3D structures [12]. High-resolution systems can achieve voxel sizes of 1 μm^3 and lower [13]. This allows determining fibre positions, fibre orientation, porosity, electrode anisotropy and homogeneity. The application to textiles has already shown to be highly successful [14,15].

To date the 3D structure of electrode materials have been investigated by several groups: Shearing et al. [16] applied μCT for graphite negative electrode characterization, Wilson et al. [17] and Ender et al. [18] applied focused ion beam in combination with electron microscopy (FIB/SEM) to $LiCoO_2$ and $LiFePO_4$ systems, respectively. Ebner et al. [19,20], Tariq et al. [21] and Chen-Wiegart et al. [22], and Yufit et al. [23] have investigated Li-ion systems using X-Ray tomography.

In this paper, we present the μCT characterisation of three configurations of $LiFePO_4$ half-cells: 2D planar Al foils (standard configuration), and two different 3D embroidered Al current collectors, with non-metallic or metallic back yarns, made of polyester (3D-Emb1) and stainless steel (3D-Emb2). The electrochemical behaviour (galvanostatic charge and discharge cycles) for the different configurations is also presented.

2. Experimental

The μCT images were obtained using a high-resolution tomography setup (phoenix nanotom m, GE) [24]. The X-Ray source type was xs 180 nf-C, of voltage range 20–180 kV, with a maximum target power 15 W. Detector was a GE's DXR500L, of 3072 × 2400 pixel, has a 100 μm pixel pitch size and a dynamic range of 10,000:1.

Approximately 2000 projections were captured over a full rotation of the samples. Scan details (X-ray energy, voxel size, number of projections and scan duration) are specified in figure captions. Offset and gain corrections were applied to adjust differences in pixel sensitivity. Image reconstruction was done using filtered back projection methods. Samples were fixed on a borosilicate glass-rod and mounted on a precision positioning stage used for sample centering and rotation.

The 3D embroidered current collectors (3D-Emb1, 3D-Emb2) were prepared by technical embroidery (Tegra 71, Grabher GmbH, Lustenau, Austria) [9]. All construction parameters for the embroidered current collectors were identical except for the back yarn. Photomicrographs of the front and back are shown in Fig. 1a and a schematic drawing of the layout is represented in Fig. 1b. Four layers of Al wires were embroidered on a background of polyester fabric (PES). Polyester Nm 135/2 (PES-BY) or stainless steel Nm 30/3 (SST-BY) was used as the back yarn. It should be noted that the SST-BY is not a pure stainless steel yarn; it is mixed with polyester (2 wires made of stainless steel with 1 filament of polyester). In Table 1 are listed diameters (D_{Al}, D_{PES}, $D_{SST\text{-}BY}$, $D_{PES\text{-}BY}$) and distances between Al wires shown in Fig. 1a,b from the embroidered current collectors as received. The thicknesses of the embroidered current collectors as received (L_0), listed in the Table 1, were measured with a thickness gauge (Karl Schröder KG, Weinheim, Germany). The thickness of 3D-Emb2 is larger than 3D-Emb1 as a consequence of the greater diameter of the back yarn ($D_{SST\text{-}BY} > D_{PES\text{-}BY}$).

The electrode material (EM) was prepared by first mixing 87 wt.% of $LiFePO_4$ (LFP) powders (MTI corporation) with 10 wt.% of carbon black (Super P–Li, Timcal), and afterwards, adding a mixture of 3 wt.% latex binder (Belenos) with 60 wt.% of deionised water [25]. The suspension was placed in an ultrasonic bath for 30 min, cast on the current collectors (4 cm^2), dried in an oven at 80 °C for 24 h, cooled down in vacuum, and compressed at 10.2 MPa for 10 min. Photomicrographs of the different current collectors with the EM (C1, C2, C3) are shown in Fig. 1c.

The LFP cells were assembled in an argon glove box using Li foil as anode and reference electrode. Separator membrane, Celgard® 2400 (samples courtesy of Celgard®) was used as the separator. The electrolyte was 1 M $LiPF_6$ in 1:1 v/v ethylene carbonate and diethyl carbonate (EC/DEC) (reagents provided by Sigma–Aldrich Chemie, Steinheim, Germany). The cells were galvanostatically charged and discharged (EG&G 274A Princeton Applied Research), at a current density of 0.25 mA/cm^2 and a voltage from 2.5 to 3.8 V (versus Li/Li^+).

3. Results and discussion

Galvanostatic charge and discharge cycle measurements on the samples yielded specific capacities of about 130 mAh/g (Fig. 2), close to the theoretical value of 170 mAh/g, which is indicative of a proper electrochemical behaviour of the 3D embroidered current collectors. Further electrochemical investigations will be presented in a future paper.

Fig. 3 shows μCT images for the different configurations C1, C2 and C3. The image contrast depends on the X-ray absorption of the material, which is related to the atomic number and material density.

Fig. 3a shows a representative transverse μCT image of the C1 configuration, current collector (Al foil) and EM. The dark areas are evidence of cracks and holes (voids) in the formed cathodes, which are attributed to water evaporation during the drying process.

Fig. 3b,c show representative transverse (right) and longitudinal (left) μCT images of the C2 and C3 configurations. At the bottom of the transverse images, the back yarns are observed. As expected, stainless steel yarns (SST-BY) are brighter than the polyester yarns (PES-BY). Some Al wires can also be observed at the bottom, due to the knot formation inherent in the embroidery process, where Al wires passed through the back yarn and are tied off. The polyester fabric (PES) is noticed between the Al wires layout and the back yarn. Above the PES, four layers of Al wires are arranged. Two videos are reported in the supplementary material showing different longitudinal μCT sections for the C2 and C3 configurations.

Supplementary video related to this article can be found at http://dx.doi.org/10.1016/j.jpowsour.2015.10.039.

The distances between the Al wires in the embroidered current collectors, after casting the electrode material and compressing at 10.2 MPa for 10 min, were measured from longitudinal and transverse μCT images. The values are shown in Table 2. Due to the