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Synchrotron Holotomography on Silicon-Based Anode Materials for Improved Lithium Ion Batteries

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Lithium ion batteries are an essential part of our society. Certainly, there is a strong demand for higher energy densities with respect to future applications in energy storage and e-mobility. The use of silicon-based anode materials is considered as a promising approach due to its high theoretical capacity. The major drawback that comes along using silicon as an anode material is its huge volumetric expansion of up to 300% during lithiation, which induces mechanical stresses on the material. These stresses result in the formation of cracks and further pulverization of the Si as well as causing the formation of an in-homogeneously growing solid electrolyte interphase (SEI) [1-4]. Capacity fading results.

Since the microstructure of the anode material is decisive for the cell performance, gaining 3D information on the material down to nm length scales is essential. Synchrotron holotomography [5] provides a powerful tool to retrieve 3D information with respect to the microstructure. The correlation of the microstructure information with electrochemical properties may lead to improved design guidelines for future advanced lithium ion cells.

In this paper, we investigate the microstructure for different anode materials and for different cycling states. We analyze different configurations of anode samples mainly defined by the use of different conducting agents and binder materials. Therefore, we define four sample configurations: 1. (Silicon/Graphite/LiPAA/ /Carbon black = 70/20/7/3 wt%); 2. (Silicon/Graphite/LiPAA/ /Carbon nano tubes = 70/22.8/7/0.2 wt%); 3. (Silicon/Graphite/CMC/SBR/Carbon black = 70/20/5/2/3 wt%); 4 (Silicon/Graphite/CMC/SBR/ Carbon nano tubes = 70/22.8/5/2/0.2 wt%). We perform ex- and in-situ synchrotron holotomography with a voxel size of 25 x 25 x 25 nm^3 at the ID16b beamline, ESRF, Grenoble France. The obtained datasets are reconstructed [6] by (i) phase retrieval calculation using an in-house developed octave script based on a Paganin-like approach, and (ii) filtered back-projection reconstruction using the ESRF software PyHST2. Advanced image analysis tools incorporating conventional and machine learning (ML)-based models are applied to retrieve the microstructure information. The analyzed volume of interest (VOI) is about 39 x 33 x 25 μm^3. Complementary field emission scanning electron microscopy (FE-SEM) is utilized for correlated morphology and chemical element analysis.

Figure 1a-c) depicts the segmented pores projected on the 3D grey value data for the pristine, 3 cycled and 300 cycled anode material, e.g. sample 2. The segmented rendered pore volume is presented below the projected image data. Clearly, a strong decrease of the porosity with cycling is depicted. About half of the pore volume degrades from pristine to 300 cycles.

In Figure 2 in-situ holotomography for sample 1 is presented, to show the evolution of the pores over three different states of discharge (SOD) upon silicon and graphite lithiation. Figure 2a) shows the 2D grayscale image for three different SOD while, Figure 2b) depicts the corresponding segmented image data for the same SOD to quantify the evolution of the pore space [7].

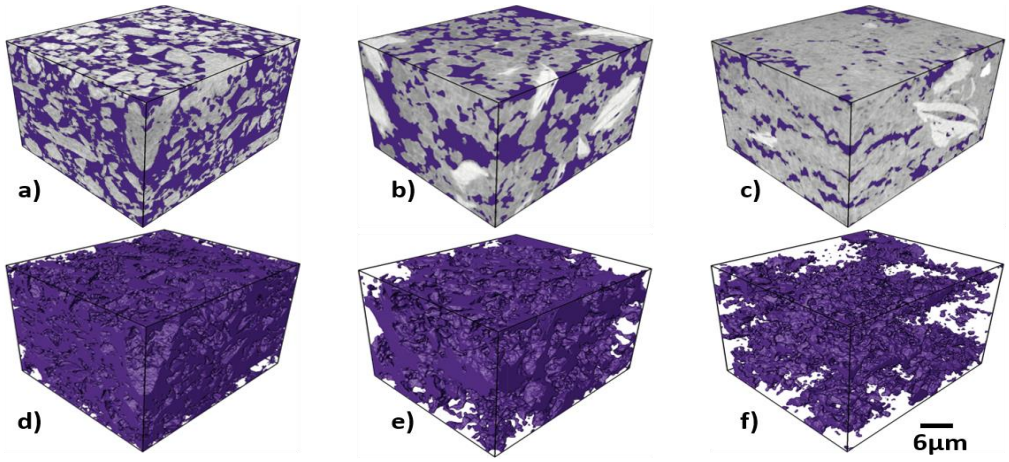

Figure 1. a-c) Synchrotron holotomography images of the silicon-based anode for the pristine state, after 3 and 300 cycles, respectively. The pores are highlighted in purple. The different gray values correspond to different material densities. d-f) Corresponding segmented pore volumes. All images show a representative VOI of 39 x 33 x 25 µm³ with a voxel size of $V_x = V_y = V_z$ = 25 nm. The scale bar with 6 µm is indicated on the bottom (right side) and is valid for all images.

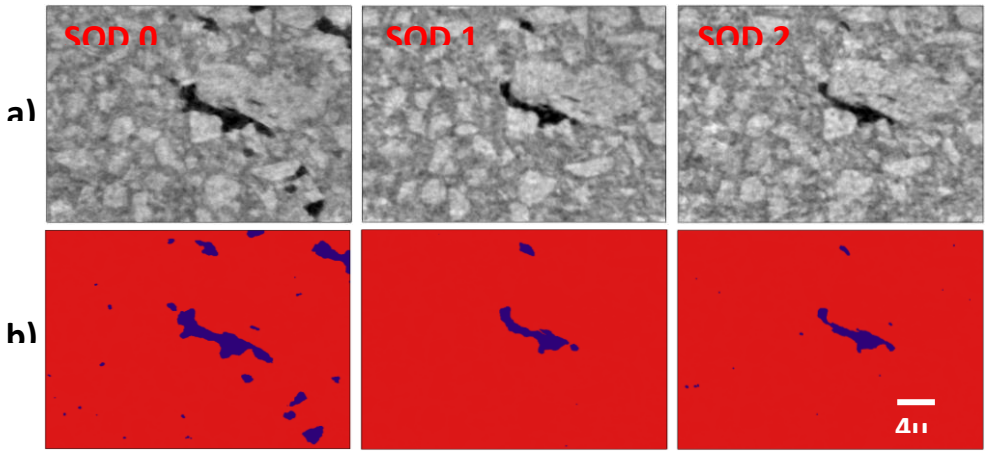

Figure 2. Synchrotron holotomography images of silicon-based anode materials during in-situ experiment. a) Selected 2D slice of the anode at SOD 0 state (Pristine), SOD 1 and SOD 2, respectively. The different gray values correspond to different material densities. b) Corresponding segmented pores (purple). Clearly, a decrease in the pore volume over time is presented. The scale bar with 4 µm is indicated on the bottom (right side) and is valid for all images.

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References:

[1] T Ikonen et al., Sci Rep **10** (2020), p. 5589. https://doi.org/10.1038/s41598-020-62564-0
[2] T Vorauer et al., Microscopy and Microanalysis (2019), p. 356-357. doi:10.1017/S1431927619002514
[3] T Vorauer et al., Commun Chem **3** (2020), p. 141. doi:10.1038/s42004-020-00386-x
[4] P Kumar et al., Small **16** (2020), p. 1906812. doi:10.1002/smll.201906812
[5] P Cloetens, W Ludwig and J Baruchel, Appl. Phys. Lett. **75** (1999), p. 2912. doi: 10.1063/1.125225
[6] C. Hintermüller et al., J. Synchrotron Rad. **17** (2010), p. 550. https://doi.org/10.1107/S0909049510011830

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