2D extinction (including Bragg edge) and 2D polarimetric imaging to monitor development of phase, microstructure and electrical currents in Li-ion batteries and electrode materials during operation

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1. Introduction

During charging and discharging of Li-ion batteries, it is well-known that the electrode materials transform as Li-ions are inserted or extracted from the crystal structure. Kinetic behaviour of Li and phase transformation mechanisms are poorly understood due to difficulties in isolating these factors/variables experimentally. To tackle this challenge, we proposed to apply novel methodologies, Bragg edge imaging and Polarimetric Magnetic Field Neutron Imaging. In this experiment we aimed to monitor phase transitions and distributions together with microstructure changes in custom-built Li-ion cells during charge-discharge, and to monitor the flow of electrical current in the material, respectively.

2. Experiment

Two Li-ion batteries were assembled as half-cell (Reference Electrode: metallic Li and Working Electrode: graphite:carbon black:polyvinylidene fluoride(8:1:1)). They were charged and discharged at different applied currents, see Figure 1 top. Between changes in the current values there were short periods (approx. 1h) of open circuit voltage measurements to allow the system to relax. Throughout the electrochemical cycling, neutron radiographies were acquired for 17,5 min. The time-of-flight images are analyzed in Matlab. The edges are smoothed and fitted, in order to compare their height, width and position.

Neutron polarimetric radiographies were furthermore recorded of a battery cell with graphite pellet. For this an external current flow of 1A was sent through the sample, so that the polarimetric signal can be used to determine current flow paths. Measurements with 0A current were made for comparison.

3. Results

So far, we have looked at the most prominent edge of graphite (at 4.07 Å) in the spectra, and we can see a change in the height, meaning that Li is intercalated in the graphite, thus a crystallographic phase change takes place. Some of the preliminary results can be seen in Figure 1 bottom.

We have not looked at Na-ion batteries, because form the preliminary tests at our home-institute, we determined that we needed more time for cycling the Li-ion batteries.

The polarimetric experiments show a clear change in signal with current running. Models for simple vertical current flows along the sample had been made in advance and it could be seen that they were insufficient for describing the recorded signal, as indication for more complicated current flow was observed. As expected further modelling will be needed to explain the recorded data.

Figured 2 and 3 show the polarimetric signals for two regions of interest (ROIs): outside and inside the graphite pellet, respectively

4. Conclusion

The radiographies of the batteries show changes in the absorption of the Lithium and in the Bragg-edges of the graphite. Further analysis is needed for a proper interpretation of the data. As follow up we consider that 3D scans are required for in-depth understanding of the phases distributions.

The polarimetric experiments revealed a strong signal, that can assist in for the first time a description of the 3D current flow inside a battery cell. Further data analysis and modelling will take place in order to interpret the measured data.

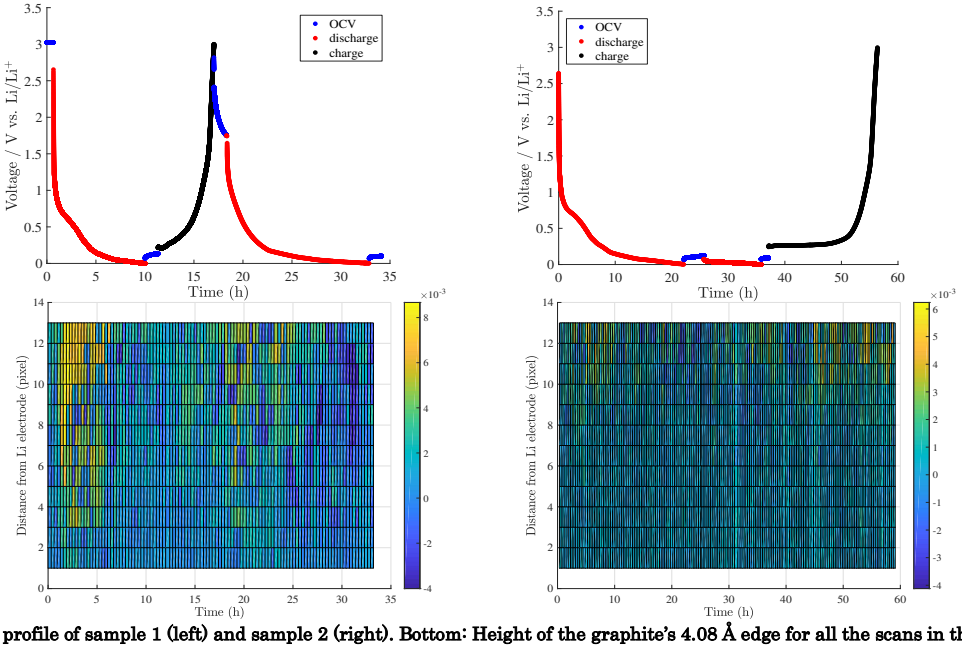

Figure 1: Top: Electrical profile of sample 1 (left) and sample 2 (right). Bottom: Height of the graphite's 4.08 Å edge for all the scans in the pellet (1 pixel = 55µm)

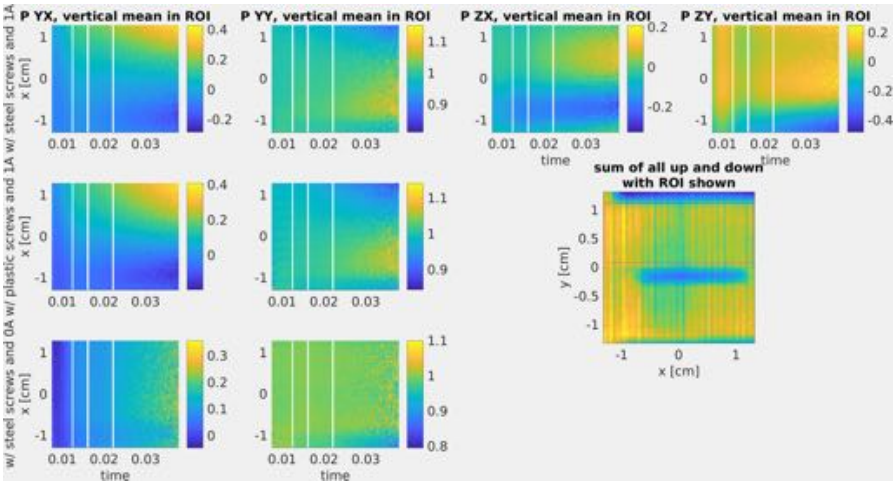

Figure 2: Polarimetric signal for ROI outside graphite pellet, comparing 0A, 1A and a sample where steel screws were exchanged for plastic ones to ensure they weren't interfering with the signal

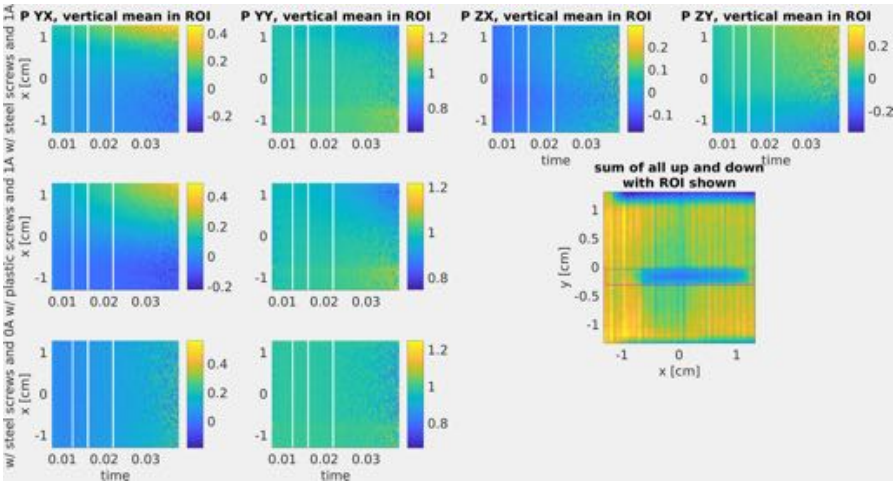

Figure 3: Polarimetric signal for ROI inside graphite pellet, comparing 0A, 1A and a sample where steel screws were exchanged for plastic ones to ensure they weren't interfering with the signal