Neutron Imaging of Degraded LFP Pouch Cells Illustrate Dendrite Formation and Internal Shorts

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Neutron imaging can be used to investigate, *in situ*, the transport and accumulation of Lithium between the positive and negative electrodes in an operating battery cell. In the present study, batteries were placed in front of a neutron sensitive detector with the separator perpendicular to the detector surface. In this orientation the positive and negative electrodes can each be resolved, and the local change in neutron intensity can be related to the local change in lithium concentration[1].

Neutron imaging of Lithium Iron-Phosphate batteries (10 layer pouch cells) with electrodes supplied by two different manufacturers were investigated. The pouch cells were 55 mAh with active area 7mm wide x 45mm tall. The electrodes were cut from larger sheets and re-packaged (10 layers stacked and connected in parallel) to minimize the neutron beam path length through the material to maintain high transmission. Both batteries exhibited degraded performance and some capacity loss due to cycling at elevated temperature (45 C) before starting the imaging experiment. The formation of lithium plating and dendrite growth was observed in both cells. Internal shorting of the pouch layers, however, was only observed in one of the batteries where we suspected that the dendrite had grown sufficiently large to short out two of the current collecting plates corresponding to the adjacent positive electrodes as shown in Fig. 1.

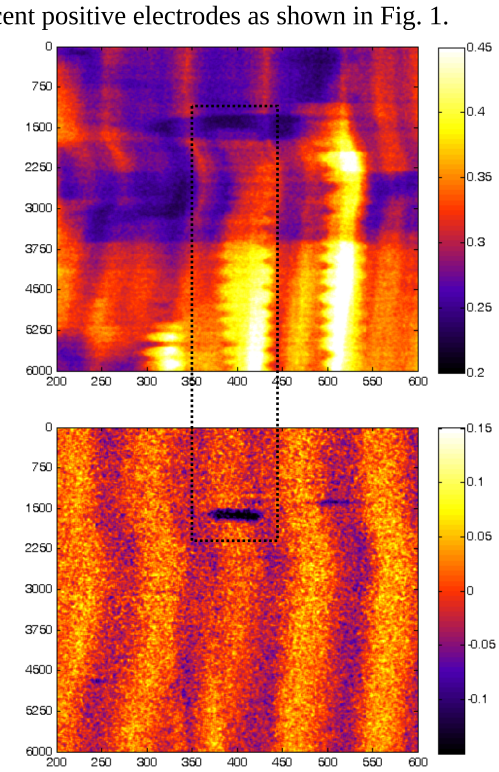

Figure 1: (Top) Neutron transmission image of the battery in the charged state; bright vertical lines correspond to the aluminum current collector. (Bottom) Optical density image of the charged state relative to the discharged state of the battery. The dark region in the center of the negative electrode corresponds to the shrinking of a lithium dendrite during charging.

Specifically, Figure 1 (top image) shows the neutron transmission through four of the cell layers inside the pouch, which are electrically connected in parallel. The dark regions (low intensity) correspond to areas with higher lithium concentration because lithium is a strong neutron absorber. The very bright vertical stripes correspond to the aluminum current collectors of the positive electrodes, which have very low neutron attenuation. One can see that due to the construction there is some irregularity in the folds. In this image it is also possible to observe the dark horizontal area centered around 400 abscissa and 1500 ordinate pixels that correspond to an area with higher than normal concentration of lithium. This formation corresponds to a Li dendrite growth between two of the current collectors.

The lower image in Figure 1 shows the optical density, i.e., the change in transmission intensity between the charged and the discharged states. Bright areas correspond to a relative increase in Li and dark areas correspond to a decrease in Li concentration. This image shows that the Li concentration changes in the four negative electrodes adjacent to the dendrite are very uniform and have same level of increase in lithium concentration during charging. The negative electrode of the shorted cell however does not have the same increase in lithium concentration as the other four.

The internal short can be seen more clearly in Fig. 2 which shows the line profile (averaged along the y-direction) of the neutron transmission across the battery electrode. The region corresponding to the lithium dendrite shown in Fig. 1 between pixels 350-425 does not exhibit any change in neutron transmission between the charged and discharged states of the battery. The remaining 9 layers demonstrate a change in neutron transmission due to the change in lithium concentration; therefore the dendrite must have shorted out the battery.

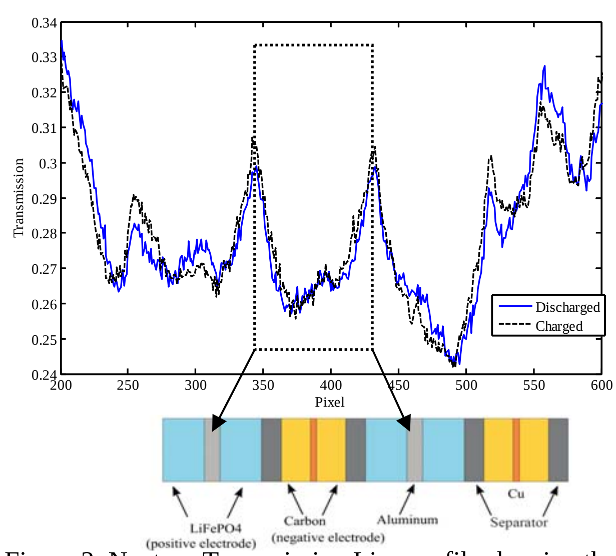

Figure 2: Neutron Transmission Line profile showing the de-activated (shorted) layer of the battery pouch in the boxed region between two positive (aluminum) current collectors.

Ex-situ observation of the electrode structure will be performed and the results can be mapped back to the corresponding location of dendrite formation observed in the neutron imaging data to confirm the hypothesis.

1. J. B. Siegel, X. Lin, A. G. Stefanopoulou, D. S. Hussey, D. L. Jacobson, and D. Gorsich, J. Electrochem. Soc. 158, A523 (2011)