EUROPEAN SYNCHROTRON RADIATION FACILITY

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ESRF

Experiment Report Form

The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF.

Once completed, the report should be submitted electronically to the User Office via the User Portal:

https://wwws.esrf.fr/misapps/SMISWebClient/protected/welcome.do

Reports supporting requests for additional beam time

Reports can be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

Reports on experiments relating to long term projects

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

Published papers

All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

Deadlines for submission of Experimental Reports

- 1st March for experiments carried out up until June of the previous year;
- 1st September for experiments carried out up until January of the same year.

Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

ESRF	**Experiment title:** Understanding microscopic chemical and microstructural changes during Li-ion battery degradation and failure	**Experiment number**: ME1422
Beamline: ID15	**Date of experiment**: from: 10 Nov 17 to: 14 Nov 17	**Date of report**: 8 Mar 18
Shifts: 12	**Local contact(s)**: Marco Di Michiel	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

*Donal Finegan (US National Renewable Energy Lab)

Paul Shearing (PI) (University College London)

Report:

$LiMnO_2$ is a non-toxic, abundant and cost-effective material for use as an electrode in lithium ion batteries. However, it exhibits rapid degradation due to residual strains inside it's crystal structure known as Jahn Teller distortions. Consequently, the Mn atoms inside it's lattice migrate and ultimately dissolve from the surface of the electrode particles into commonly used electrolytes.

The purpose of this experiment was to capture the migration of Mn in the $LiMnO_2$ structure as it occurs during operation in a bespoke lithium ion cell. We manufactured suitable cells based on the Swagelok union fitting with a custom-made PEEK central piece that accomodates a cell with a diameter of around 1 mm (Figure 1 a). This narrow geometry was required to achieve a high signal to noise ratio during the XRD experiments.

This cell was then cycled whilst taking 2D high-speed XRD measurements (Figure 1 b), hence simultaneous electrochemical measurements and XRD were carried out. Before and after each cycle, 3D XRDCT scans captured a slice of the entire electrode (Figure 1 c) to monitor the spatial and temporal degradation of the $LiMnO_2$ electrodes. In total, we successfully imaged 3 x $LiMnO_2$ cells for between 2 and 4 cycles each. Furthermore, pre-cycled cells (50, 100 and 150 cycles) were brought to the beamline to image using high resolution XRDCT ex-situ, the results of which will contribute to building a map of the short-term as well as long-term degradation effects observed in Li-ion batteries.

In addition to the LiMnO2 cells, we imaged one cell with a Si-graphite composite electrode. Utilising the high speed point XRD capability of ID15, we constructed an experiment where during charging a different electrochemical potential would be achieved between the Si and graphite particles. Hence, when the cell is switched to open circuit (OCV), the Si and graphite would continue to communicate via transfer of Li ions. Imaging at 100 Hz, we hope to capture this communication at open circuit and show that equilibration periods need to be considered in composite electrodes.

At ID15 we achieved a 1 µm resolution point XRD scans as well as 3D XRDCT recontrustions. Using the Pilatus Dectris CdTe 2M detector, we were also able to achieve measurements at up to 100 Hz at 75 keV monochromatic beam. This capability cannot be achieved anywhere else, making this experiment stand out as a technological precipice.

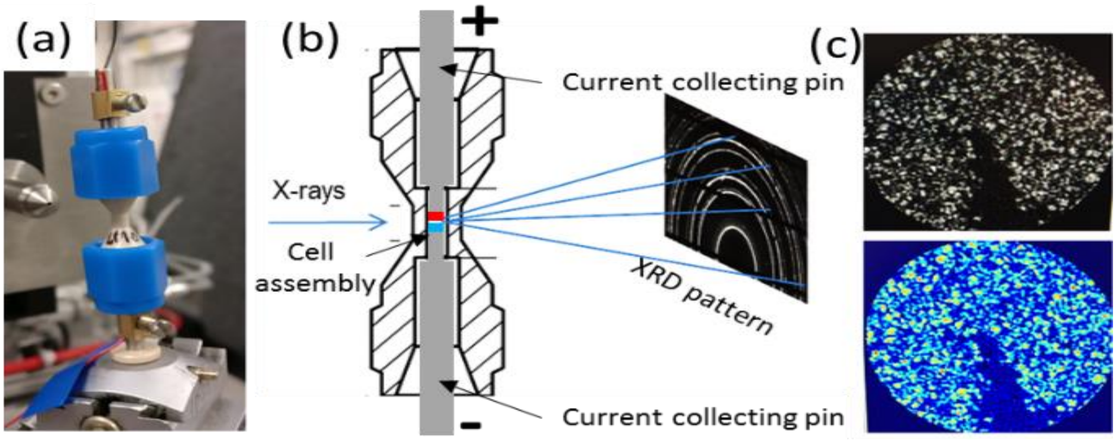

Figure 1: (a) Swagelok union-inspired cell design with a PEEK core. (b) Cross-section of the Swagelok cell showing the shape of the current collecting pins, the position of the cell, and the incident beam. (c) Reconstructions of a XRDCT scan of a LiMnO2 electrode.

The preliminary images shown in Figure 1c are demonstrative of the success of this experiment. The crytalline particle features are clearly observed and, for the first time, sub-particle spatial XRD information can be extracted. The unique high flux and high energy capability of ID15, as well as the skills of Marco Di Michiel and Antony Vamvakeros, made this proposed experiment possible. Based on the resultes observed so far, we expect one or more high impact publications.