ESRF	**Experiment title:** Observing Charge Compensation Mechanisms in Li-ion Battery Cathodes with X-ray Raman Spectroscopy	**Experiment number**: MA-5753
Beamline: ID20	**Date of experiment**: from:10/04/2023 to:17/04/2023	**Date of report**:
Shifts: 18	**Local contact(s)**: Christoph Sahle	*Received at ESRF:*

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Report:

In this experiment we investigated the charge compensation mechanisms of Li-ion battery cathodes in a bulk-sesntive manner, using hard X-ray Raman Scattering (XRS) to probe the Ni L edge and O K edge transitions and Ni Kβ valence to core (V2C). Being the first time we have performed these experiments, we planned a fairly ambitious set of measurements involving both ex-situ and operando approches for three materials systems ($LiCoO_2$, $LiNiO_2$ and $LiNiMnCoO_2$), which gave several options depending on which seemed most promising with the measurement setup. We quickly narrowed this to the two material systems that were of most interest ($LiNiO_2$ and $LiNiMnCoO_2$), and after the first day decided to focus on ex-situ samples only (although we received excellent support on setting up the electrochemistry equipment by Blanka Detlefs and Valentin Vinci which was valuable to understand what may be possible in future).

Christoph Sahle provided outstanding support and discussion throughout the beamtime (amongst the very best we have had at any synchrotron facility), and has since been willing to assist in further data analysis post-beamtime. Part of the outcome of the experiment for us was to apply a lesser known (in the field of batteries) spectroscopic technique to systems where bulk information is difficult to obtain. We obtained high quality XRS and V2C data from all samples (22 electrodes at different states of charge) which show consistent trends between the $LiNiO_2$ and $LiNiMnCoO_2$. We have since performed more surface sensitive soft X-ray absorption spectroscopy measurements of the same samples. Comaprison of these results shows some extremely interesting differences that have allowed us to seperate near surface degradation processes from bulk redox. We are now writing up a manuscript on this which we hope to submit in the coming months, and some of the data is likely to go into a second manuscript although this may require some further measurements.

Since the result turned out so well we intend to extend this project and apply for further time in the next call for proposals, as we expect XRS to provide further insight to clarify bulk redox process in several battery cathode materials that we are interested in.