Finding a Needle in the Haystack: Identification of Functionally Important Minority Phases in an Operating Battery

The materials and devices used in modern society are often structurally complex and chemically heterogeneous. The complexity in the material is usually caused by the desired functionality that has requirements in many different aspects of the material properties. Taking Li-ion battery as an example, the device is often evaluated by combining several different characteristics, including the energy density, capacity, cyclability, temperature stability, price etc. As a result, material scientists need to look into the realistic systems, in which both the anticipated and the unanticipated material phases/reactions occur.

The importance of locating functionally important minority phases in obtaining a deeper understanding of complex working materials and devices has driven the rapid development of fast, high-resolution, chemically sensitive probes, especially at large scale experimental facilities such as synchrotrons. While the experimental capability is being greatly advanced, more effort is needed for the development of computing methods that could effectively and efficiently extract the scientifically important information from the large datasets.

A research team led by Drs. Yijin Liu and Apurva Mehta, SSRL, Dr. Xiqian Yu, Institute of Physics (Beijing), Dr. Xiao-Qing Yang, Brookhaven National Laboratory, applied the big data mining concept to the *in situ* study of $LiCoO_2$ battery electrodes. In a paper published earlier this year [*ACS Energy Lett.* **2**, 1240 (2017)], this team demonstrated the *in situ* monitoring of a selected $LiCoO_2$ particle over many electrochemical cycles. Their results in that paper suggested that the particle is capable of readjusting itself in response to different local chemical environments (Figure 1).

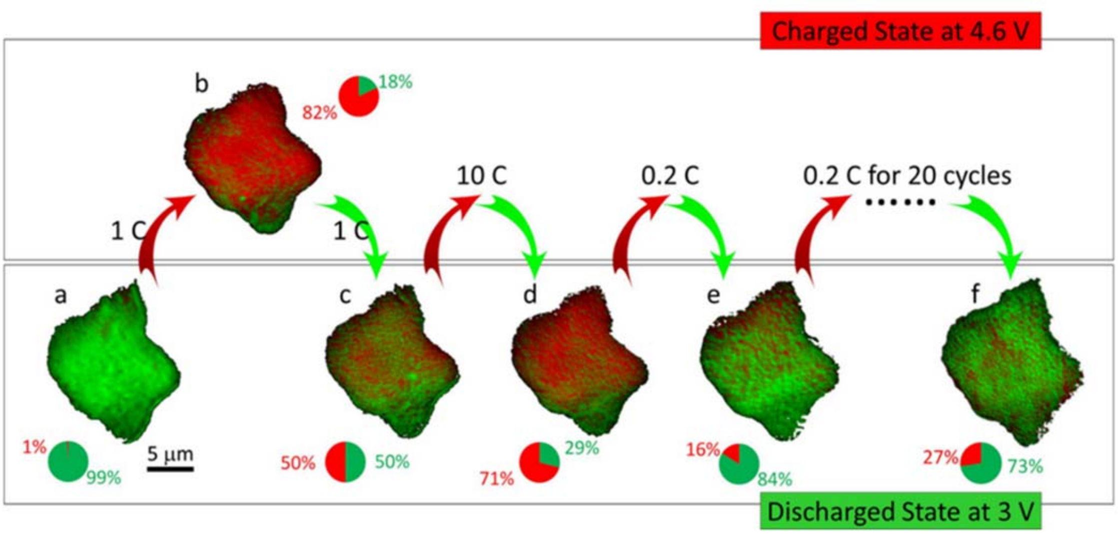

Figure 1. *In situ* monitoring of the chemical heterogeneity in a single particle of $LiCoO_2$ up to 23 cycles at different cycling rates. The red area represents the domains at charged state (Co^{4+}), and the green area represents the domains at discharged state (Co^{3+}).

While it is exciting to visualize the individual particle's behavior over long-term cycling, scientists will naturally question the representativeness of a single particle to the entire battery cell. To address this question, the team surveyed the battery cell at many different locations. After initial data reduction, they effectively retrieved over 10 million x-ray absorption spectrum covering more than 100 active particles. They developed an algorithm for extraction of spectroscopic data attributes, which are then fed into the computing engine for clustering. The developed computing method identified two different particles that are abnormal in terms of their spectroscopic fingerprints. These two particles were further attributed to two different unwanted side reactions that happened during the electrochemical cycling (Figure 2).

While the combination of *in situ* nano imaging capability and the big data mining method has provided very useful insights in the field of battery research as presented in the current paper, it is anticipated that the development in data mining method will play an even more important role as the instrument/facility continues to advance.

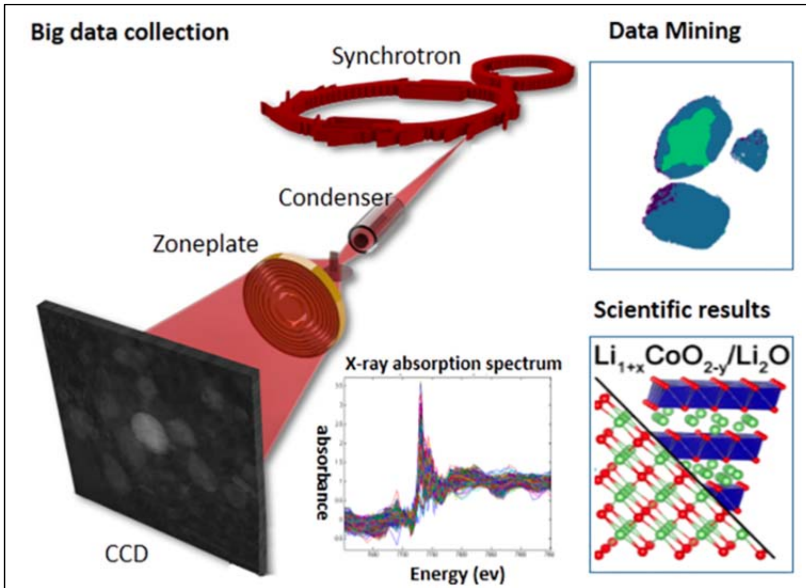

Figure 2. **Big Data collection:** Nanoscale x-ray spectro-microscopy technique (Beam Line 6-2C of SSRL) collects spatially resolved x-ray absorption spectra at very fast data rate. **Data Mining:** Advanced data mining approach developed in this work searches through over 10 million spectra and identified two particles with unanticipated chemical fingerprints. **Scientific results:** These two particles are attributed to two different unwanted side reactions.

References

1. Y. Xu, E. Hu, K. Zhang, X. Wang, V. Borzenets, Z. Sun, P. Pianetta, X. Yu, Y. Liu, X.-Q. Yang and H. Li, "*In situ* Visualization of State-of-Charge Heterogeneity within a $LiCoO_2$ Particle that Evolves upon Cycling at Different Rates", *ACS Energ. Lett.* **2**, 1240 (2017), doi: 10.1021/acsenergylett.7b00263
2. K. Zhang, F. Ren, X. Wang, E. Hu, Y. Xu, X.-Q. Yang, H. Li, L. Chen, P. Pianetta, A. Mehta, X. Yu and Y. Liu, "Finding a Needle in the Haystack: Identification of Functionally Important Minority Phases in Operating Battery", *Nano Lett.* **17**, 7782 (2017) doi: 10.1021/acs.nanolett.7b03985

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