

Preview

Machine learning accelerates identification of lithiated phases in X-ray images of battery hosts

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Santos et al. (2022) propose a machine learning-based approach to identify various lithiated phases across lengthscales in X-ray images of battery particles, thus enabling automatic interpretation of such information in much bigger datasets and creating opportunities to unravel previously inaccessible scientific understanding.

Have you ever wondered how physicians and radiologists read medical X-rays? Their interpretation of X-ray images largely relies on having looked at numerous images of healthy and ailed bodies and consequently recognizing unusual features. Now wonder how exhausting it would be if someone had to interpret thousands of images simultaneously. Would they be able to maintain the same level of accuracy while interpreting each of those thousand images? In lithium-ion batteries, there are thousands of particles,^{1,2} and our interest is understanding the lithium distribution in each of these particles. Scanning transmission X-ray microscopy (STXM) is one of the X-ray imaging techniques with a spatial resolution of tens of nanometers and a large enough field of view to simultaneously capture lithium distribution in multiple such particles.³ However, such information is difficult to accurately interpret, especially for materials exhibiting multiple lithiated phases (e.g., V_2O_5), and consequently, the challenge is to convert the STXM images to reliable lithiation maps. In the latest study, Santos et al. (2022)⁴ propose a machine learning approach to sift through such large datasets of V_2O_5 electrodes and accurately identify the underlying lithiated phases.

As shown in Figure 1A, the same particle is imaged at multiple X-ray energies. These energies are chosen such that the X-ray transmission changes as the particle lithiates. This dataset is further analyzed using reference spectra of pure uniformly lithiated phases (Figure 1B) to obtain the

lithiation map in Figure 1C. The lithiation map is composed of the underlying distribution of these phases. While such an analysis may seem straightforward, our ability to implement it in practice relies on (1) having accurate spectra of pure lithiated phases and (2) a deconvolution technique that can convert the STXM data to lithiation maps.

Herein, the authors meticulously address both these aspects. First, they obtain pure lithiated phases by chemically lithiating V_2O_5 particles in stoichiometric quantities, so their composition is precisely known. These particles are subsequently used to generate the accurate reference spectra in Figure 1B. The authors use machine learning algorithms like singular value decomposition (SVD) and k-means clustering to create a robust analysis scheme that deconvolves the STXM data (images at multiple X-ray energies) to generate the corresponding lithiation map. The authors further demonstrate the usefulness of this approach by analyzing different STXM datasets of V_2O_5 particles at multiple lengthscales.

The cleverness of this approach lies in using the reference spectra to create the deconvolution scheme. In addition to the reference spectra being true references (e.g., not based on theoretical assumptions of the underlying atomistic picture), they implicitly account for instrumentation nonlinearities with X-ray energies. Unlike convolutional neural networks (CNNs) and other machine learning techniques that would have just correlated the STXM dataset directly to the lithium distribution, the

approach proposed here deconvolves the X-ray images using reference spectra and creates the corresponding lithiation maps. Hence, it is equally applicable to any STXM dataset of the same materials with a different resolution or field of view. As a result, the authors can apply the same approach to obtain lithiation maps across lengthscales ranging from within a single particle to multiple particles. Additionally, as the authors showed, the same analysis is also valid for interpreting X-ray ptychography data for lithiation maps at lengthscales smaller than STXM. The authors also make these datasets and the developed tools open access for the broader scientific community.

While the results shown here by the authors refer to *ex situ* imaging of the battery particles, one can extend the usefulness of the proposed approach by selectively imaging only a few representative energies, thus effectively enabling *operando* measurements.² Yet another extension can be imaging these particles from different angles to create a 3D tomographic reconstruction of the lithiation fields in these particles.⁵ With parallel advances in X-ray imaging of electrolyte concentration field,⁶ we may now be able use a single setup to map concentration fields in both the energy-storing particles and the adjoining ion transporting electrolyte.

Besides such instrumentation advances allowing us to visualize fields that were inaccessible just a few years ago, such schemes enable us to pose important scientific questions to build better batteries. The operation of any battery is



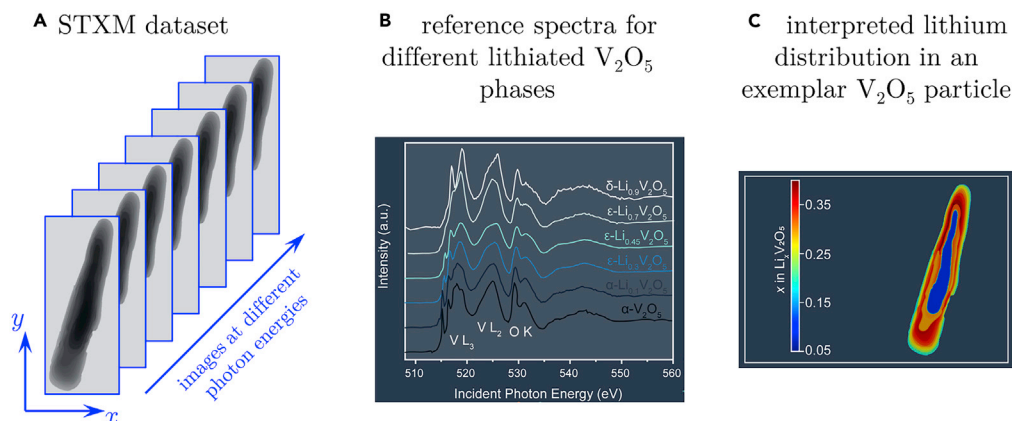


Figure 1. Different steps for machine learning-enabled identification of lithiated phases

(A) Representative STXM dataset of a V_2O_5 particle imaged at different X-ray photon energies.

(B) Reference X-ray spectra of different lithiated V_2O_5 phases.

(C) The corresponding lithium distribution interpreted from the STXM dataset using reference spectra of pure phases.

The images are reproduced from Santos et al. (2022).⁴

microscopically a combination of interactions between multiple fields, such as species concentrations, mechanical stresses, and electric potentials. Consequently, understanding the mechanisms causing spatiotemporal inhomogeneities and asymmetries is quite important. The approach of Santos et al.⁴ and its aforementioned extensions can help understand many such mechanisms. For example, the coupled effects of lithiation and mechanical stress have been described in many different ways in the literature.^{4,5,7} Each of these descriptions for lithiation–mechanics coupling predicts a distinct lithiation dynamics for the same operation. We can combine such predictions with the observations of lithiation fields to characterize the lithiation–mechanics behavior in the intercalation material being examined. This will not only be useful for cathode materials⁸ like V_2O_5 but also relevant in next-generation anodes like silicon and other alloying materials. Apart from electrode manufacturing defects, carbon-binder domains are an intrinsic source of geometrical inhomogeneity in battery electrodes. Imaging lithiation fields with different carbon-binder distributions would help to identify the relevance of their hypothesized contributions.¹ Equivalently, one can formulate an *operando* study to underpin the role of grain structure in battery particles⁹ and how it can alter lithiation preferences at the scale of an individual particle. In a similar spirit, such spatiotemporal information about lithiation fields can help elucidate the potential field distribution, specif-

ically in low conductivity materials.^{5,10} The electric potential is expected to vary even within such particles and in turn skew the intercalation reaction at the particle–electrolyte interface, which in turn causes a distinct evolution in the particle-scale lithiation field.

Thus, being able to accurately observe the concentration field in battery particles at relevant lengthscales opens up a range of opportunities to explore previously inaccessible aspects of the battery operation and accordingly make systematic improvements to build better batteries.

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DECLARATION OF INTERESTS

The authors declare no competing interests.

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