Full-field Chemical Mapping with 2D in situ X-ray Nano-imaging and 3D Nano-tomography

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Chemical, electrochemical and morphological evolutions of battery electrode materials during the cycling are critical information for they are strictly tied to the mechanism and performance of a battery. Two aspects in this need are crucial. Firstly, it is important to characterize these changes in situ – for the continuous evolution from the same sample volume can bring more insights to study the battery system. Secondly, it is also critical to spatially resolve the evolution in three dimensions. Compared to obtaining averaged bulk information or studying in a 2D geometry where only the sample surfaces or projected overlapped features were observed, the details can be more clearly revealed in 3D, especially when concerning critical parameters such as volumetric strains, specific areas, tortuosity and 3D chemical distributions.

Here we present the advance in x-ray full-field spectroscopic imaging technique and computational analysis methods from the newly developed transmission x-ray microscopy (TXM) at the beamline X8C of National Synchrotron Light Source. Through least-squares linear combination fitting using computational models on the in situ spectroscopic images, a time-dependent and spatially resolved chemical composition mapping can be obtained. Therefore, the oxidation states evolution can be quantitatively analyzed when a battery electrode is undergone chemical/electrochemical reactions. We successfully applied this method to study the electrochemical and morphological evolution of metal oxides as anode materials of Li-ion battery, during the lithiation-delitiation cycling (Fig. 1). The results provide critical insights regarding the reversibility, reaction mechanism and morphological stability of the material.

Furthermore, three-dimensional (3D) spectroscopic imaging has been developed and used to characterize lithium iron phosphate (LiFePO₄, Fig. 2) and lithium manganese phosphate (LiMnPO₄, Fig. 3). Although these two materials were predicted by theory to exhibit similar capacity, it is not well-understood why it has been demonstrated experimentally that LiFePO₄ has a much higher capacity than LiMnPO₄. Through computational analysis of 3D spectroscopic imaging, the correlation of morphology, oxidation states distribution changes and electrochemical reaction can be revealed in these systems.


Fig. 1. Full-field x-ray spectroscopic imaging and computational mapping method, illustrated using an metallic oxide anode of the lithium ion battery undergo cycling. The relative volumetric fractions of can be quantified [1].