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Quantitative interpretation of reaction distribution in thin-film cathode of all-solid-state lithium-ion batteries by using *operando* depth-resolved soft x-ray absorption spectroscopy

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Abstract

All-solid-state lithium-ion batteries (ASSLIBs) were put in the spotlight due to the superior power and energy densities, and higher thermal robustness, which pushing ASSLIBs to be a promising choice for the next generation rechargeable batteries. However, the utilization of electrode significantly decreased by the rapid charging rate. One of the reasons belongs to the sluggish Li-ion/electron transport within cathode during charging and discharging that could form the reaction distribution. In order to trace the formation of reaction distribution, *operando* depth-resolved soft x-ray absorption spectroscopy (DR-sXAS) has been developed to dynamically observe the chemical state of cathode in depth at specific charging state. Thin-film $LiCoO_2$/LICGC/PEO/Li has been used as model battery. The *operando* DR-sXAS measurement has been carried out every two hours during the cycling test at 0.1 C and cut-off voltage between 2.0-4.5 V. DR-sXAS spectra were quantified by using the angle-resolved weight subtraction analysis. The results showed the different chemical state of $LiCoO_2$ emerged after 1st charging cycle when the $LiCoO_2$ film is thicker than 100 nm. It is considered that the Li-ion transport in depth of $LiCoO_2$ was significantly slow because the Li-ion has to transport across the CoO_6 layered structure in order to de/intercalate with the $LiCoO_2$.

Keywords: All-solid-state lithium-ion batteries, X-ray absorption spectroscopy, Depth-resolved measurement, Operando measurement, Reaction distribution