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(Invited) Advanced Insitu Electron Microscopy for Batteries: Insights for Lithium-Ion and Beyond

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Abstract

Rechargeable batteries are crucial for energy storage across consumer electronics and automobile propulsion applications, facilitating the transition towards carbon neutrality and advancing clean energy technologies. Despite great success of Li-ion batteries (LIBs) in the commercial market, alternative technologies based on beyond-Li chemistry are highly demanded for large-scale and power-intensive applications necessitating enhanced energy density, lifetime, and safety, where fundamental understanding of the structure-property relationship of novel battery materials is critically needed. Transmission electron microscopy (TEM) is an indispensable method to characterize materials structures and compositions at the atomic scale, which is of particular importance for battery research to investigate crystal lattices, defects, as well as microstructural and chemical heterogeneities within materials used in electrodes, electrolytes, and their interfaces. Further, with rapid technical development, in-situ TEM has enabled real-time observations of various dynamical phenomena and chemical processes during battery cycling and phase transformations. Leveraging advanced in-situ TEM techniques, our collaborative endeavors with Dr. Marca Doeff have enabled us to conduct comparative analyses of Li and Na reactions within battery electrodes, offering unique insights into in early-stage beyond-Li chemistry. Herein, we present a systematic exploration of in-situ TEM studies for LIBs and beyond, focusing on electrode materials through intercalation, alloying, and conversion reaction mechanisms. By direct comparison between electrochemical reactions with Li and Na, we found substantial differences in reaction mechanisms, pathways, and kinetics between lithiation and sodiation processes, which are fundamentally related to various factors, such as ionic diffusion barrier, electrochemically induced stress, and geometric constraints. This concept has been demonstrated in multiple case studies that allows us to enhance the sodiation kinetics by tuning the overall reaction energetics through nanostructure optimization and interfacial engineering. We envision that the knowledge learned from in-situ TEM will provide valuable insights into understanding the alkali-ion electrochemistry and kinetics, thereby serving as foundational principles guiding the advancement of beyond Li-ion battery technologies.

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