High-Pressure Multi-Probe Studies of Lithium-Ion Battery Materials

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Abstract

Recent advances in high-pressure and multi-probe characterization techniques offer new opportunities to investigate and engineer materials for next-generation lithium-based batteries. Pressure is a particularly powerful parameter because it can directly modify atomic arrangements, defect populations, microstructure, and transport properties, providing insight into structure–property relationships that are difficult to access under ambient conditions.

Our research employs a multiscale approach combining advanced synthesis, in situ high-pressure experiments, and complementary characterization methods to explore the response of battery materials under compression. Particular attention is given to understanding how pressure influences both structural order and functional properties in electrode and solid-electrolyte materials.

For the cathode material $LiNiO_2$, a promising cobalt-free positive electrode, in situ synchrotron X-ray diffraction and Raman spectroscopy measurements were performed in a diamond anvil cell up to 50 GPa. Compression induces significant modifications of the layered structure, including variations in Li/Ni cation mixing and local structural disorder. The results reveal a competition between pressure-induced ordering and disordering mechanisms: moderate compression stabilizes the layered framework, whereas higher pressures promote cation mixing and local distortions. These observations provide new insight into the structural stability of Ni-rich cathodes and the influence of lattice disorder on electrochemical performance.

Pressure also plays a key role in the processing and optimization of solid electrolytes for all-solid-state batteries. Using the UToPEc setup at the Psiché beamline of Synchrotron SOLEIL, we conducted in situ high-pressure sintering experiments on amorphous Li_3PS_4 and crystalline Li_6PS_5Cl under compressive stresses up to 3700 MPa (Thompson et al., Adv. Funct. Mater., 2026, DOI: 10.1002/adfm.75195). This unique platform enables the simultaneous acquisition of synchrotron X-ray diffraction, X-ray absorption tomography, density measurements, and electrochemical impedance spectroscopy. By correlating structural evolution, densification, and porosity changes with ionic conductivity, we identify the mechanisms governing pressure-assisted consolidation and ion transport.

Together, these studies demonstrate how pressure can be used both as a probe of fundamental structural behavior and as a processing tool for optimizing battery materials.

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