

Decoupling structural and chemical changes in mixed-anion batteries

Scientific Achievement

We combined operando pair distribution function (PDF) and NMR spectroscopy to resolve the complex reaction of a mixed anion battery electrode. Despite reversible energy storage, the original electrode never regenerated, instead the active electrode partitions into O- and F-rich components.

Significance and Impact

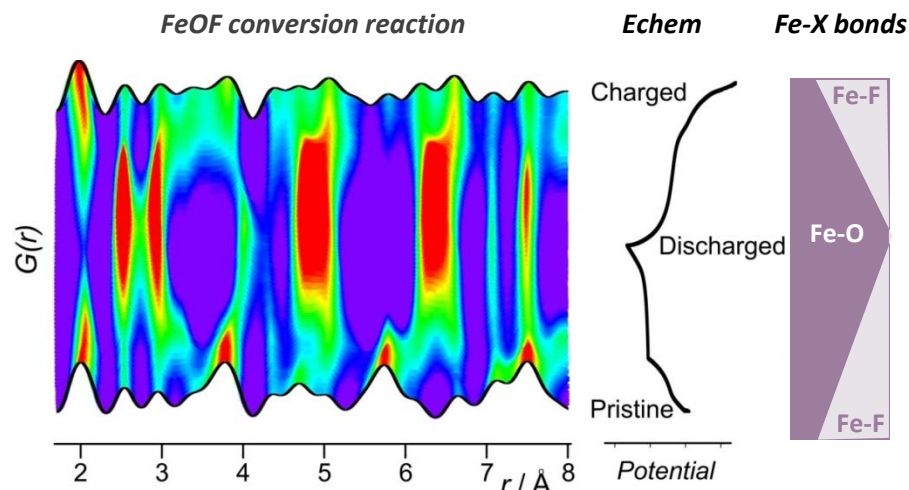
Competition between electrochemical reactions involving the oxide and fluoride components may contribute to the enhanced cyclability of this electrode. The multi-phase nature of the active electrode suggests a new (simpler) approach to developing new electrodes with the enhanced performance of oxyfluorides through nanoscale mixing of pure oxides and fluorides, including systems where oxyfluorides are unknown.

Research Details

High energy X-ray scattering data were collected at 30 minute intervals during cycling at 11-ID-B – the dedicated PDF beamline at the Advanced Photon Source, Argonne National Laboratory. The first-of-a-kind operando PDF study yielded insights of unprecedented detail, allowing the complex multi-phase behavior and changes in chemistry and particle morphology to be resolved. The operando studies were feasible only due to a newly designed electrochemical cell compatible with PDF measurements.

K. M. Wiaderek, O. J. Borkiewicz, E. Castillo-Martínez, R. Robert, N. Pereira, G. G. Amatucci, C. P. Grey, P. J. Chupas, K. W. Chapman – J. Am. Chem. Soc. 2013, 135 (10), 4070. DOI:10.1021/ja400229v

This work was performed at Argonne National Laboratory, Rutgers University, Stony Brook University and University of Cambridge.



Structural changes probed using operando PDF analysis indicate a partitioning of a FeOF-based electrode into fluorine- and oxygen-rich phases with a different reactivity for each component



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