Characterizing and Controlling Matter away from Equilibrium Using the Model Compound Electrode $LiFePO_4$

Scientific Achievement

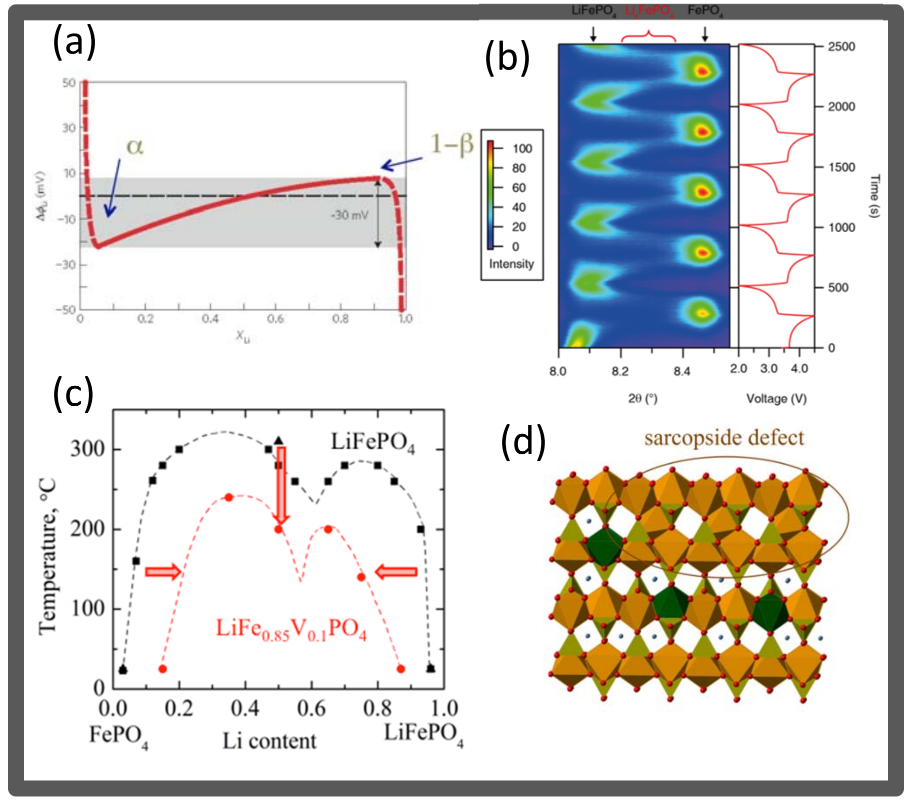
Theoretical model combined with advanced operando characterization and designed synthesis explain why some nanostructured materials react via a fast non-equilibrium route

Significance and Impact

Mechanism shows both that a non-equilibrium single phase reaction route can lead to higher rate battery materials, and how to control formation of this single phase

Research Details

- Theoretical model predicts that lithium disorder and a single phase in Li_xFePO_4 can be formed and controlled by applying a small applied overpotential
- Operando x-ray diffraction on lithium cells show a single phase reaction pathway, that is controlled by particle size and applied potential (reaction rate)
- Doping of the material increases the disorder to the point that no overpotential is required, and an equilibrium phase is formed



Findings: (a) Model showing 30 mV overpotential creating non-equilibrium single phase reaction region from α to 1-β; (b) x-ray diffraction showing single phase between $LiFePO_4$ and $FePO_4$; (c) Change of equilibrium phase diagram on doping; and (d) the sarcopside defect formed at high levels of doping.

Work was performed at Argonne, Binghamton, Cambridge, MIT, San Diego and Stony Brook.

M. Stanley Whittingham, *Chemical Reviews* 114, 11414 (2014).

U.S. DEPARTMENT OF ENERGY | Office of Science | NECCES | BINGHAMTON UNIVERSITY STATE UNIVERSITY OF NEW YORK | RUTGERS | UNIVERSITY OF CAMBRIDGE | UC San Diego | UCSB | Argonne | UIC | UNIVERSITY OF MICHIGAN | MIT Massachusetts Institute of Technology