Contribution ID : **163** Type : **Poster Presentation**

Atomistic Mechanisms of Phase Stability in Mn-Based Layered Oxide Cathodes for Sodium-Ion Batteries

Manganese-based layered oxides (NaxMnO2) are promising cathode materials for next-generation sodium-ion batteries (NIBs) due to their high energy density and cost-effectiveness. [1] Among the key structural phases, P2 and O3 differ in oxygen stacking, with P2 exhibiting superior sodium-ion diffusion. [1] However, its electrochemical performance is hindered by solid-state phase transitions during Na+ insertion/extraction, leading to structural degradation and capacity fade. [2] A fundamental understanding of these transitions at the atomic level is crucial for improving long-term stability. In this work, we investigate the thermodynamic and kinetic factors governing phase evolution in P2-NaxMnO2. [3] Density functional theory (DFT) calculations reveal that phase stability and transformation pathways are dictated by cooperative Jahn–Teller effects (CJTE) from Mn3+ centres, Na+ ordering, and electrostatic/covalent interactions. To probe the kinetics of these transitions, we employ variable-cell nudged elastic band (VC-NEB) calculations, [4] identifying shear-driven gliding and tetrahedral transition states as dominant energy barriers in P2-to-P2' and P2-to-OP4/O2 transformations. Furthermore, we introduce structural distortion metrics as predictive descriptors of phase stability, offering a new approach to designing more resilient cathodes. [5] Our findings establish a comprehensive framework for optimizing Mn-based layered oxides by balancing thermodynamic stability and kinetic accessibility. This work provides critical design guidelines for enhancing electrochemical durability in sodium-ion batteries, advancing their viability for large-scale energy storage.

Primary author(s) : LANGELLA, Aniello

Co-author(s) : Dr. MASSARO, Arianna; MUNOZ GARCIA, ANA BELEN (Dipartimento di Fisica "Ettore Pancini"); Prof. PAVONE, Michele (University of Naples Federico II)

Presenter(s) : LANGELLA, Aniello