Role of Strain and Defects in Anion Redox Electrochemistry

Y. Shirley Meng[*1,2], Minghao Zhang[2], Bao Qiu[3] and Andrej Singer[4]

1. Sustainable Power & Energy Center, University of California San Diego
2. Department of NanoEngineering, University of California San Diego
3. Ningbo Institute of Materials Technology and Engineering, Chinese Academy of Sciences
4. Department of Physics, University of California San Diego
***shirleymeng@ucsd.edu**

Oxygen-redox reactions are believed to be responsible for the excess capacity in Li rich layered oxides [1]. Nevertheless, the major issue of anionic redox is the irreversibility correlated with defects generation, which results in serious structure degradation and the voltage decay during extended cycles [2]. Despite their importance, deep understanding of the defects formation and its effect on voltage fade has not been explored. Here, using operando Bragg coherent diffractive imaging, we directly confirm dislocations and strain form more readily in the LRLO particles as compared with classical layered oxide (Fig. 1). Combining the DFT + *U* calculations with experimental observations, we demonstrate the unique metastable structure of the cycled LRLO due to different defects generation, which raises the system energy and leads to voltage fade. Novel treatment methods are applied on the cycled material, which successfully eliminates the defects and recovers the voltage fade .

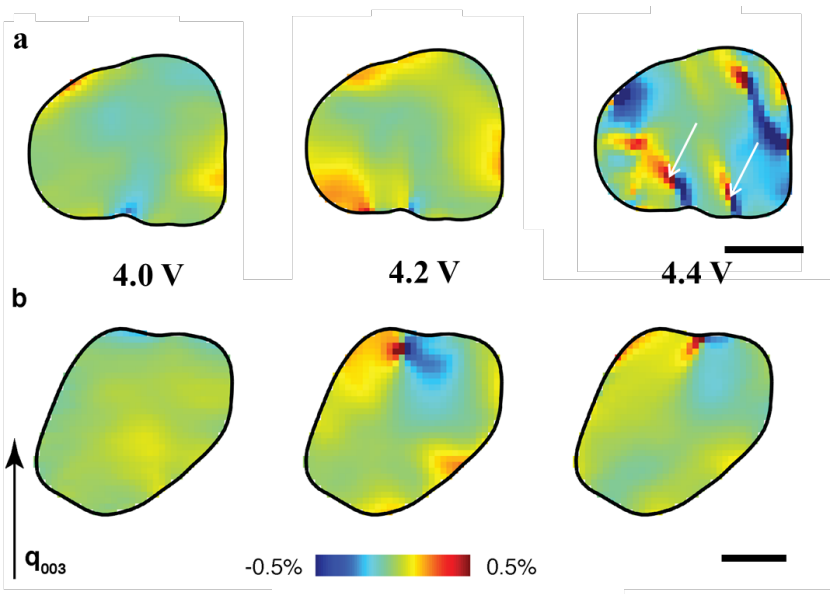

Figure 1: Evolution of strain along the [001] direction in a nanoparticle of (a) LRLO and (b) NCA captured in situ during charge. The state of charge is indicated and the scale bar is 100 nm. White arrows in (a) indicate the positions of the edge dislocations [2].

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References:

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