

## Supplemental Information

### **Profiling the Nanoscale Gradient in Stoichiometric Layered Cathode Particles for Lithium-ion Batteries**

Feng Lin,<sup>1,\*</sup> Dennis Nordlund,<sup>2</sup> Isaac M. Markus,<sup>1,3</sup> Tsu-Chien Weng,<sup>2</sup> Huolin L. Xin,<sup>4,\*</sup>  
Marca M. Doeffl,<sup>1,\*</sup>

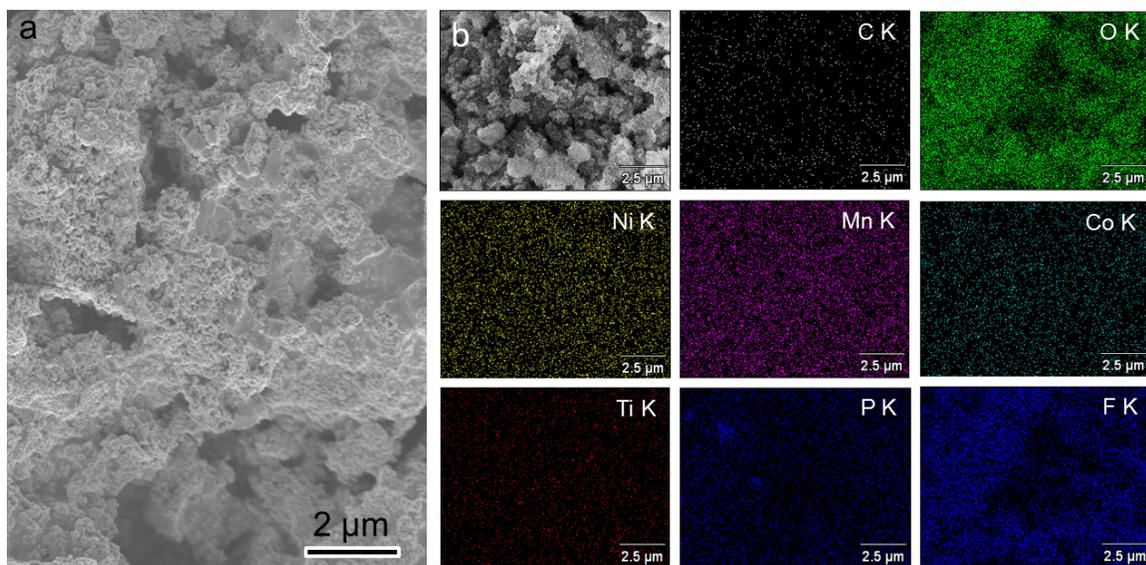
1. Environmental Energy Technologies Division, Lawrence Berkeley National  
Laboratory, Berkeley, CA 94720, USA

2. Stanford Synchrotron Radiation Lightsource, SLAC National Accelerator Laboratory,  
Menlo Park, CA 94025, USA

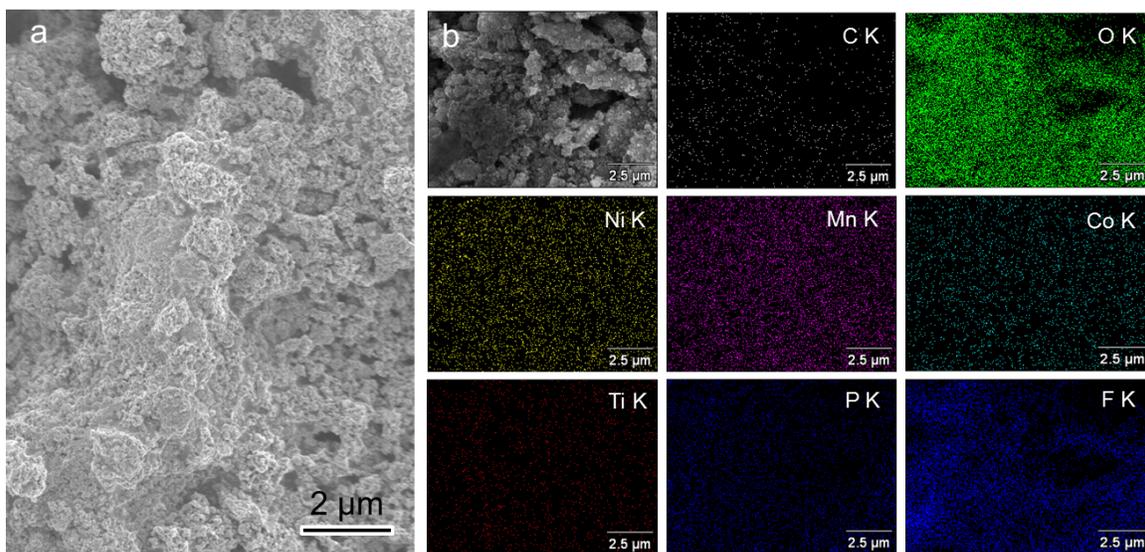
3. Department of Materials Science and Engineering, University of California, Berkeley,  
CA 94720, USA

4. Center for Functional Nanomaterials, Brookhaven National Laboratory, Upton, NY  
11973, USA

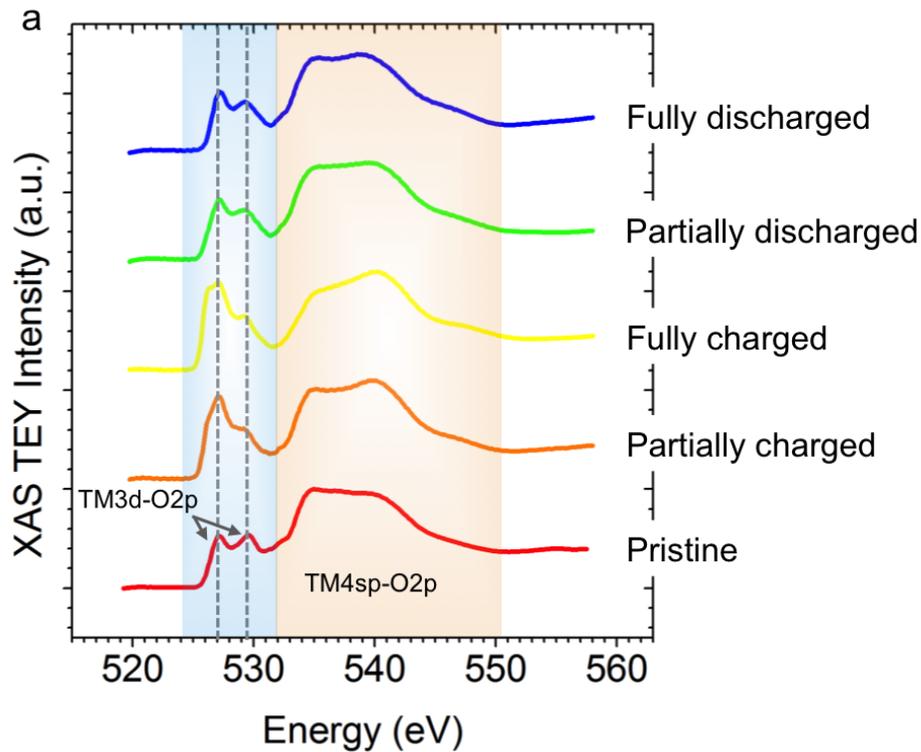
All correspondence should be addressed to F.L. [flin@lbl.gov](mailto:flin@lbl.gov), H.L.X. [hxin@bnl.gov](mailto:hxin@bnl.gov),  
M.M.D. [mmdoeff@lbl.gov](mailto:mmdoeff@lbl.gov)



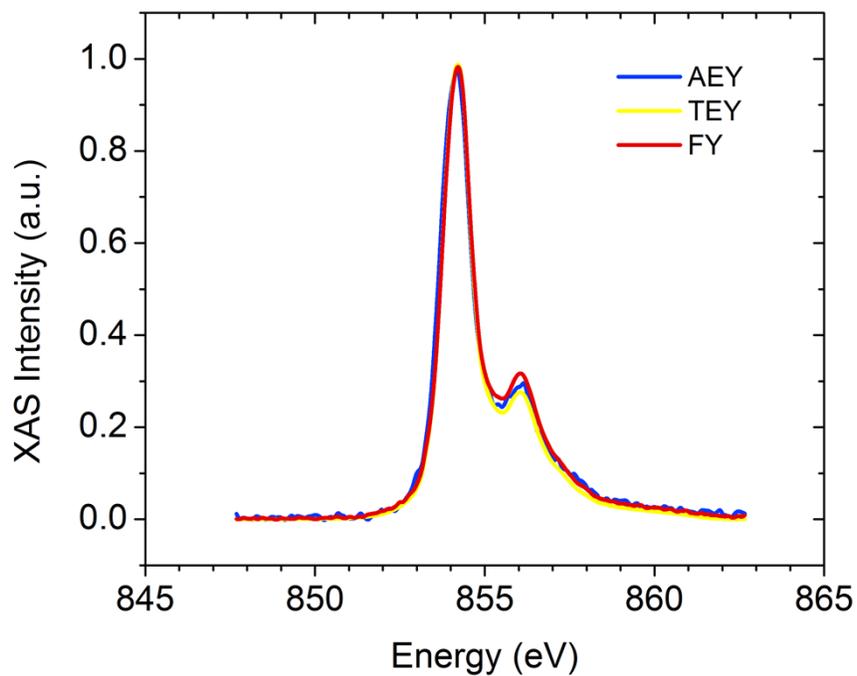
**Figure S1.** SEM and EDS mapping of a carbon- and binder-free NMC electrode after exposure to electrolytic solution (1 M  $\text{LiPF}_6$  dissolved in EC: DMC 1:2) and being dried naturally in a He-filled glovebox. The presence of P and F is due to residual  $\text{LiPF}_6$  at the surface.



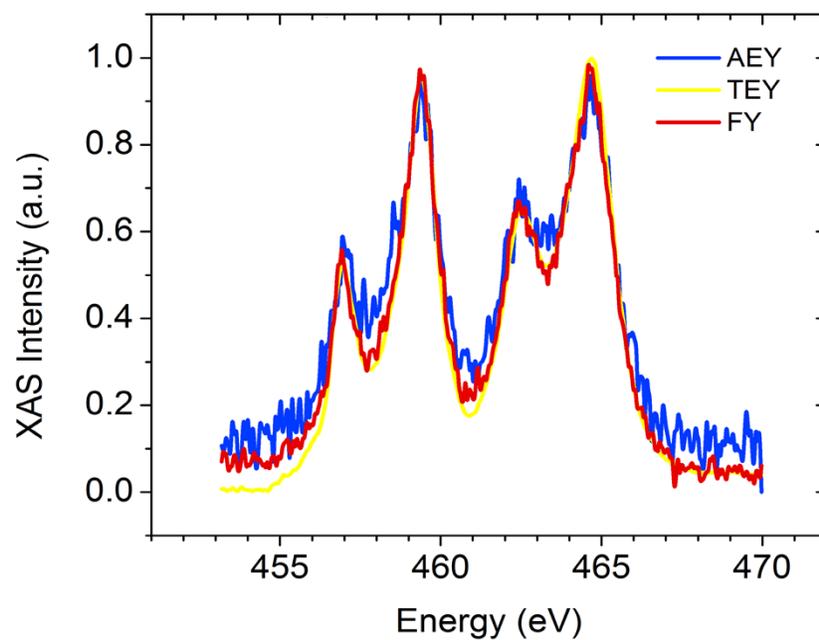
**Figure S2.** SEM and EDS mapping of a carbon- and binder-free NMC electrode in the charged state, rinsed by DMC and dried naturally in a He-filled glovebox. The presence of P and F is due to residual  $\text{LiPF}_6$  and a surface reaction layer.



**Figure S3.** XAS/TEY O K-edge spectra of battery electrodes with various SOC. The pre-edge features associated with the TM3d-O2p hybridization states in the  $\text{TMO}_6$  crystal field are indicated by the two dashed lines in the figure. From the bottom to the top,  $x \approx 1$ , 0.6, 0.2, 0.7 and 0.9 in  $\text{Li}_x\text{Ni}_{0.4}\text{Mn}_{0.4}\text{Co}_{0.18}\text{Ti}_{0.02}\text{O}_2$ .



**Figure S4.** XAS Ni L3-edge spectra of a battery electrode in the fully discharged state ( $x \approx 0.9$  in  $\text{Li}_x\text{Ni}_{0.4}\text{Mn}_{0.4}\text{Co}_{0.18}\text{Ti}_{0.02}\text{O}_2$ ).



**Figure S5.** XAS Ti L-edge spectra of a battery electrode in the fully charged state ( $x \approx 0.2$  in  $\text{Li}_x\text{Ni}_{0.4}\text{Mn}_{0.4}\text{Co}_{0.18}\text{Ti}_{0.02}\text{O}_2$ ), where the most surface sensitive Auger electrons were detected for AEY collection.