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Supporting Information

Essential Effect of the Electrolyte on the Mechanical and Chemical Degradation of $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ Cathodes upon Long-Term Cycling

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Supplementary Figures:

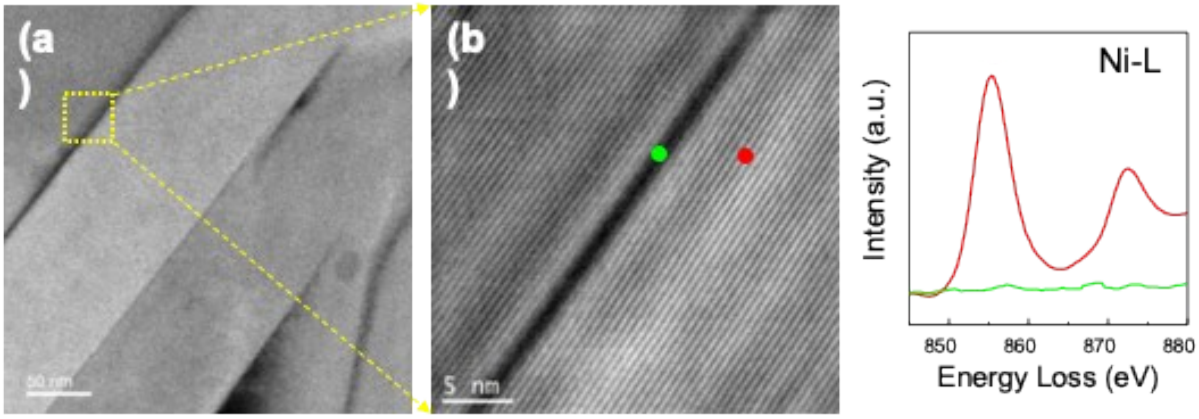

Figure S1. (a-b) HAADF-STEM images acquired at some loosely contacted grains in the pristine NCA secondary particle shown in Figure 1a. The inset of panel b shows Ni L-edge EELS spectra obtained at the two points indicated.

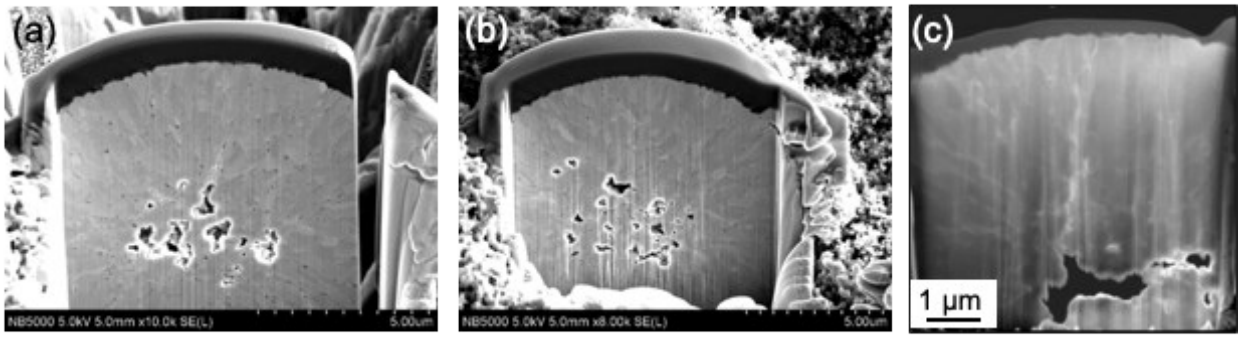

Figure S2. (a-c) SEM images of the cross-section in a secondary NCA particle after long-term cycling.

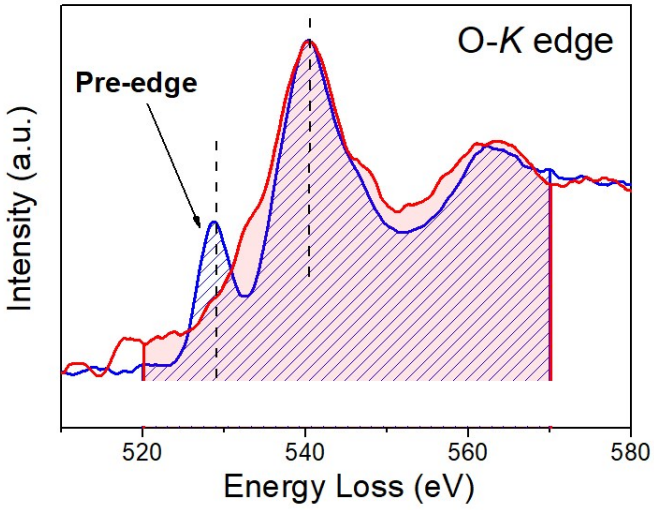

Figure S3. Integration of the O-*K* edge between 520 and 570 eV at surface (red shadow) and bulk (blue shadow) after the spectra were normalized to the total intensity of signal between 510-950eV.

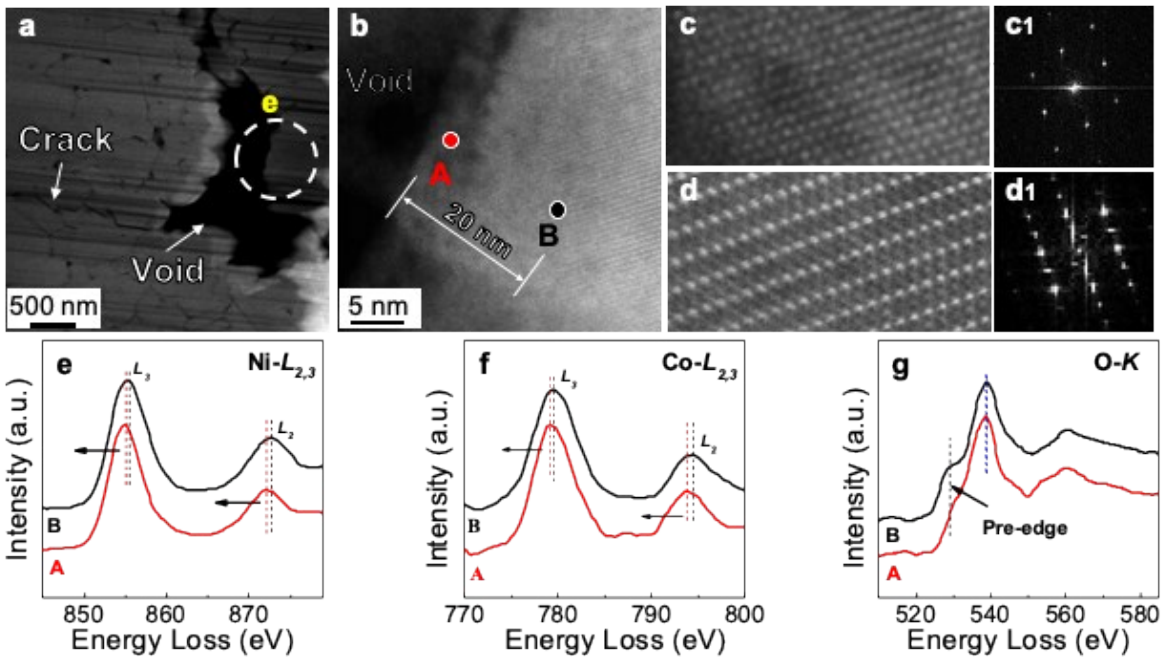

Figure S4. HAADF-STEM images and EELS spectra of cycled NCA. (a) A void in the center of the secondary particle. (b) A close examination of the circled area in panel a. (e-d) High-resolution STEM images and FFT patterns of the highlighted points in panel b. EELS spectra of Ni *L*-edge (e), Co *L*-edge (f), and O *K*-edge (g) at the bulk (B) and reconstruction layer (A) of the particle near the void.