Chemical States of Overcharged $LiCoO_2$ Particle Surfaces and Interiors Observed Using Electron Energy-Loss Spectroscopy

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1. Surface chemical states of the 40% charged particle

Figure S1b shows EELS spectra from the areas at edge, β and inside of the 40% charged particle, α as presented in Figure S1a. The EELS spectrum from α reveals CoO-like phase at the narrow area of the particle surface.

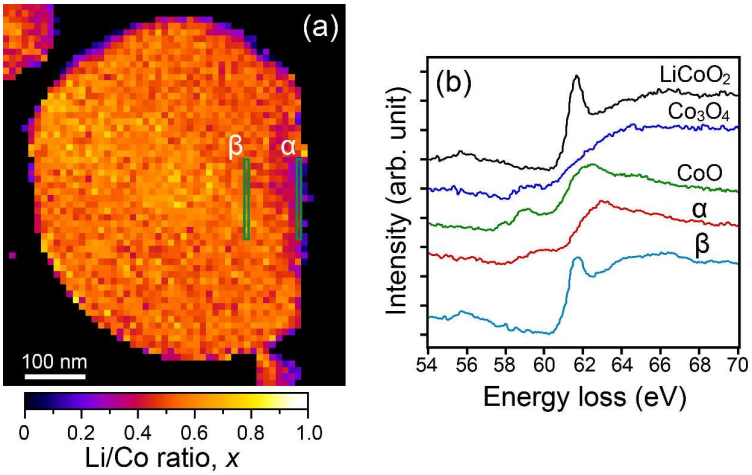

Figure S1: EELS spectra collected from the rectangle areas of α and β in (a), with the reference EELS spectra of $LiCoO_2$, Co_3O_4, and CoO.

2. Charged particle with nano-cracks in Figure 5

Selected-area electron diffraction (SAED) patterns were recorded using an electron microscope (HF-3000S; Hitachi Ltd.) operated at 300 kV with an imaging plate system (FDL-5000; Fujifilm). Figure S2 displays the SAED pattern acquired from a 400 nmφ area of the particle center in Figure 5a (including the area of Figure 5d), where the indices are based on the original hexagonal unit-cell of the $LiCoO_2$. Solid and dotted

circles in Figure S2b respectively show fundamental and extra spots for the original $LiCoO_2$. The nano-cracks are extended to the [*h*,*k*,0] (*h*, *k*: integers) directions.

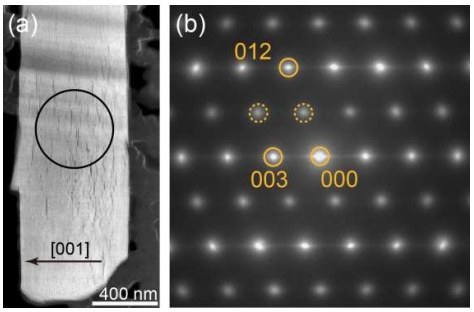

Figure S2: SAED pattern in (b) acquired from the circled area in the 60% charged particle with nano-cracks in the ADF-STEM image (a).

Figure S3 shows a histogram of Li/Co ratio, *x* for Figure 5b. Gaussian fits show the presence of the main component at *x*=0.28 and minor component at *x*=0.08.

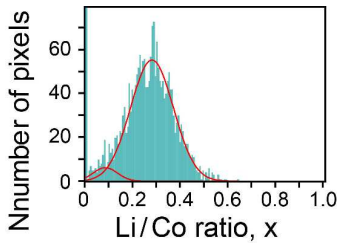

Figure S3: Histogram of *x* for Figure 5b with Gaussian fits.