Supporting Information: Understanding improved capacity retention at 4.3 V in modified single crystal Ni-rich NMC//graphite pouch cells

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1. ToF-SIMS point-to-point data of pristine SC Al-surface-doped-Ni-rich NMC

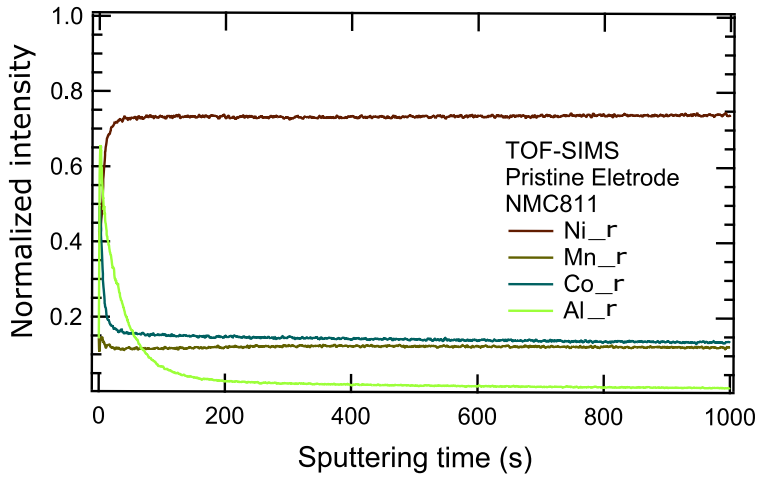

Figure S1: Rescaling normalization of the Point-to-point intensities for qualitative comparison.

The Ni_r, Mn_r, Co_r, and Al_r relative values in Figure S2 are obtained following $(XO^- + XO_2^-)/(NiO^- + NiO_2^- + MnO^- + MnO_2^- + CoO^- + CoO_2^-)$, where X is Ni, Mn, Co, and Al. It only presents the relative behaviour of each element from surface to bulk. Intensities show an opposite behavior at long sputtering time signifying deeper SC NMC-811 particle depth. At small sputtering times, Al signal is greater in intensity and decays in deeper particle depth in a depth profiling experiment. The trends for the transition metals oppose the Al's trend with higher intensities at deeper depth into the SC NMC-811 particle. Moreover, the relative values for the transition metal at long sputtering time shows a steady composition in the bulk of SC NMC-811. To be noted the intensities are not only proportional to the concentrations but significantly affected by the known matrix effects. Lack of the relative sensitive factor (RSF) for each specie makes it is very difficult to perform a quantitative analysis in SIMS. The values do not reflect the real bulk composition values here, and the bulk composition is determined by ICP.

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2. XAS TM experiments on graphite to confirm TM oxidation states

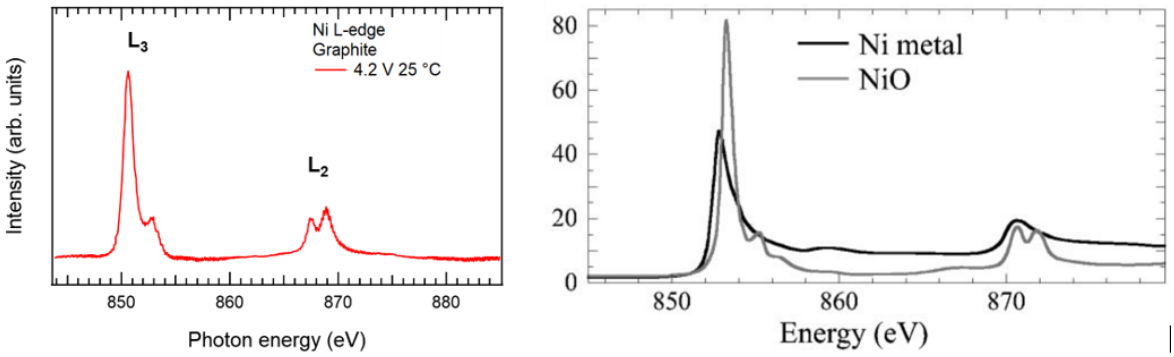

Figure S2: L-edge XAS experiments on graphite after 500 cycles and comparison with literature

The left figure is our Ni L-edge measurements on graphite after 500 cycles at 4.2 V 25 °C. The right figure is literature reference from [Phys. Rev. B (2001) 64, 21442, https://doi.org/10.1103/PhysRevB.64.214422]. A metallic environment shows a single broad L_3 and L_2 peak while Ni ions with ligands show splitting on these peaks indicating that oxidation states other than the metallic is expected for the ions deposited on the graphite anode.