Supporting Information

Understanding the role of aluminium on determining the surface heterogeneity and electrochemical performance of layered cathodes

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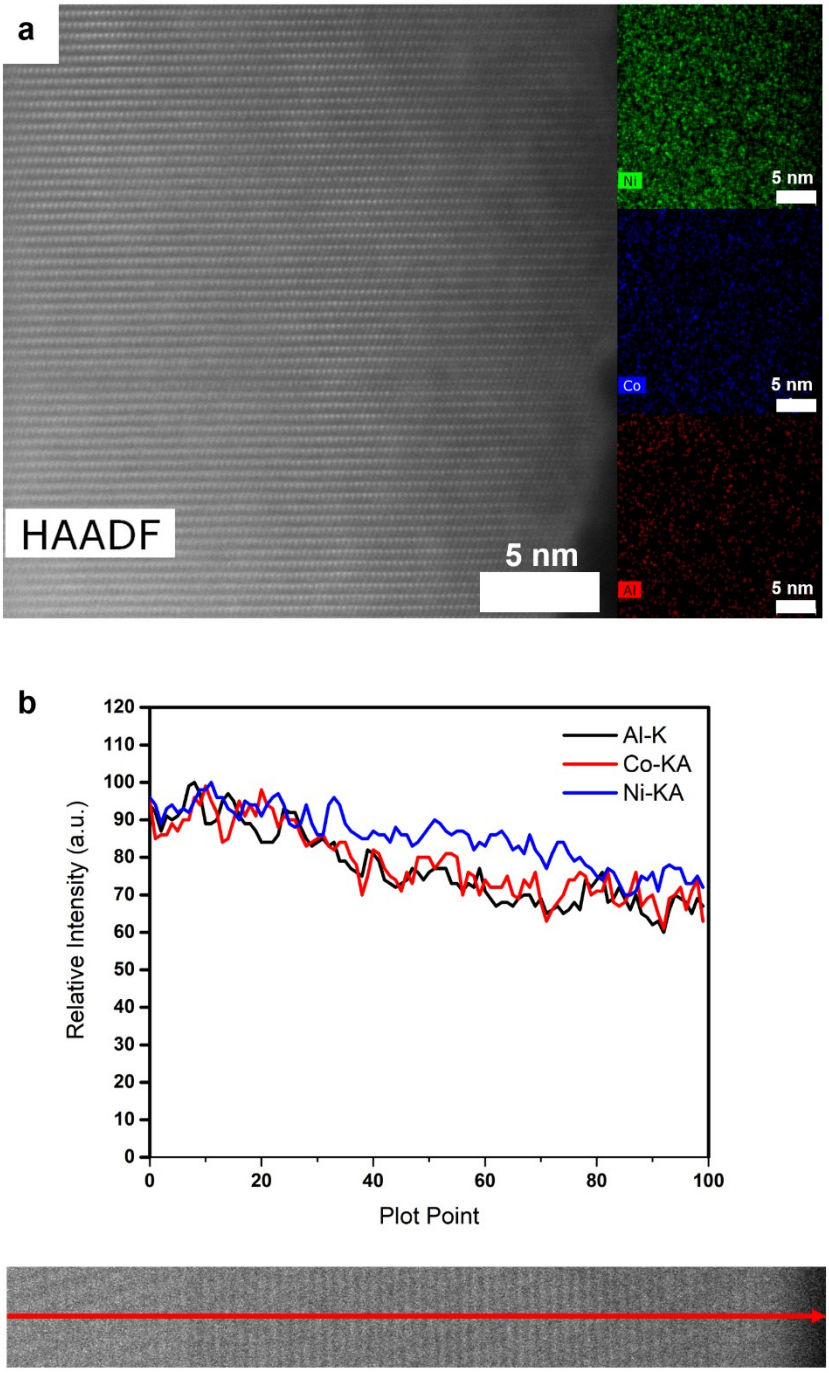

Figure S1. a) HAADF-STEM image of the D30 and corresponding elemental mapping images of Ni, Co, Al. b) EELS line scan spectrum of the Ni, Co, Al.

In the EELS line scan test, the variation trend of the three element signals is similar, which could eliminate the effect of sample thickness on the signal. Furthermore, the curves of the three elements fluctuate slight, which could prove the uniform distribution of three elements in the particles.

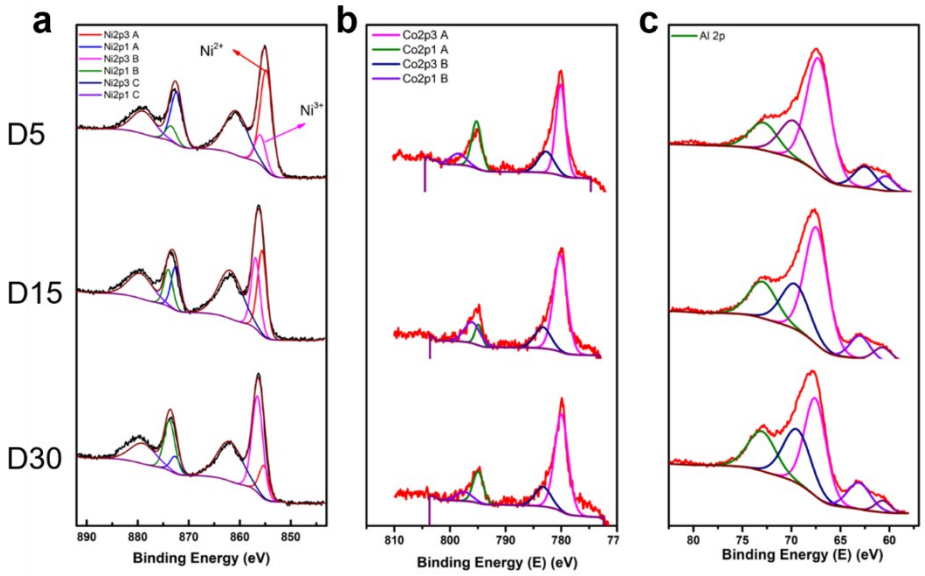

Figure S2. XPS High-resolution a) Ni 2p, b) Co 2p, c) Al 2p spectra of the D5, D15 and D30.

Table S1. Ni^{2+}/Ni^{3+}, Co^{2+}/Co^{3+} ratio of three samples.

Sample	Ni^{2+}/Ni^{3+} ratio	Co^{2+}/Co^{3+} ratio
D5	79.12:20.88	31.06:68.94
D15	55.20:44.80	35.54:64.46
D30	24.90:75.10	31.43:68.57

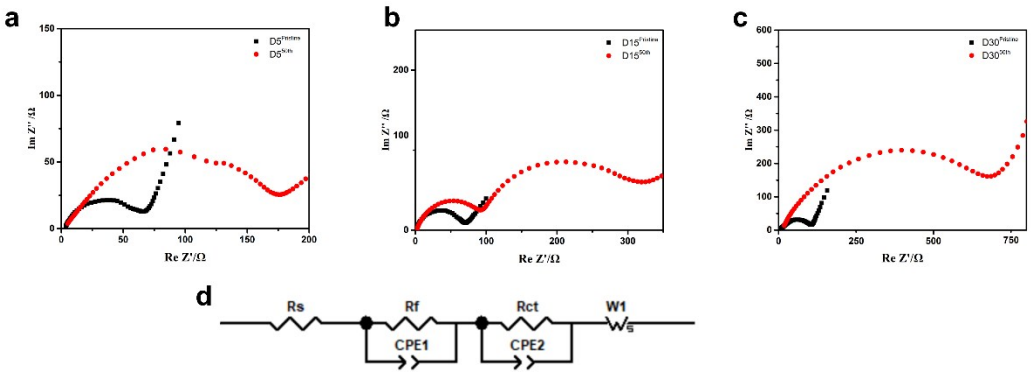

Figure S3. EIS spectra of pristine electrodes and prepared electrodes after 50 cycles. a) D5, b) D15, c) D30. d) The equivalent circuit for the electrochemical impedance of three samples.

Table S2. The impedance value of pristine and cycled LNCA samples

	$D5^{Pristine}$	$D5^{50th}$	$D15^{Pristine}$	$D15^{50th}$	$D30^{Pristine}$	$D30^{50th}$
Rf	45.81	58.9	14.21	69.08	6.962	42.88
Rct	16.91	146.5	48.21	296.3	91.84	586.2

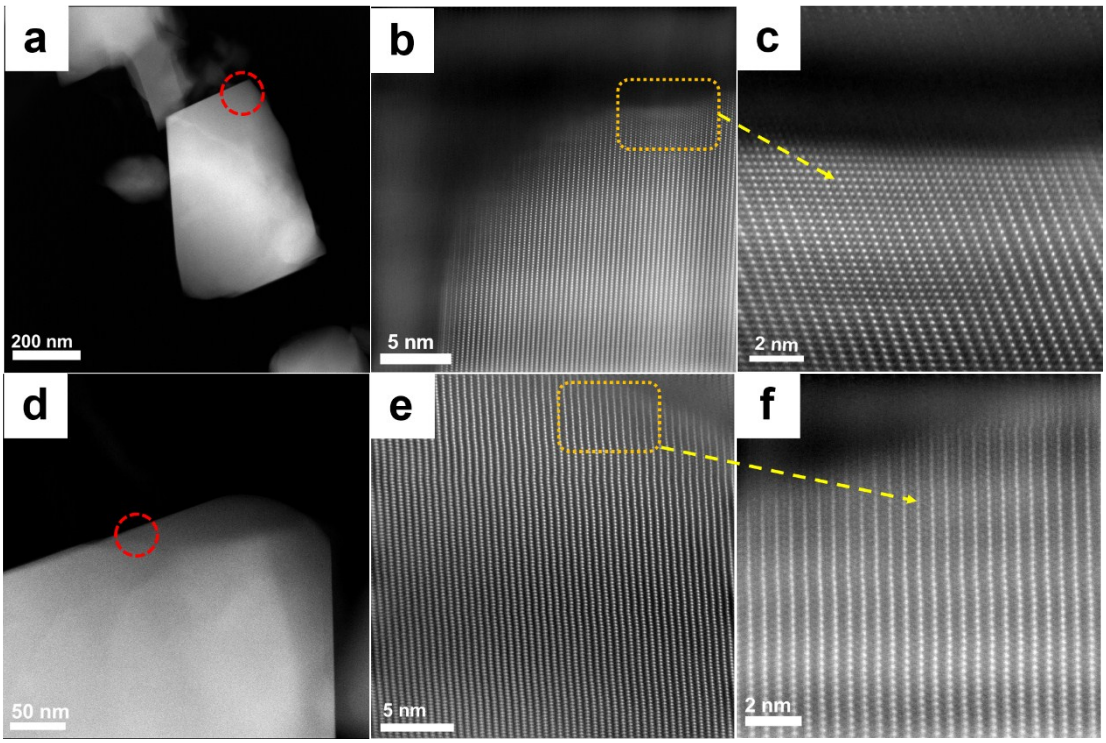

Figure S4. a)–c) HAADF-STEM images of sample D5 along [010] zone axis from low magnification to high magnification, c) exhibits the lattice of D5 margin. d)–f) HAADF images of sample D30 from low magnification to high magnification, f) exhibits the lattice of D30 margin.

From **Figure S4 a, b, d, and e**, both samples have regular layered structures in the interior lattice. However, for the samples' boundary, D30 still maintains relatively layered lattice (**Figure S4f**), D5 instead exhibits bright spots between Li layers which should not appear (**Figure S4c**). The bright spots represent heavy elements permeate in Li layers and it is only possible to be a transition metal here.

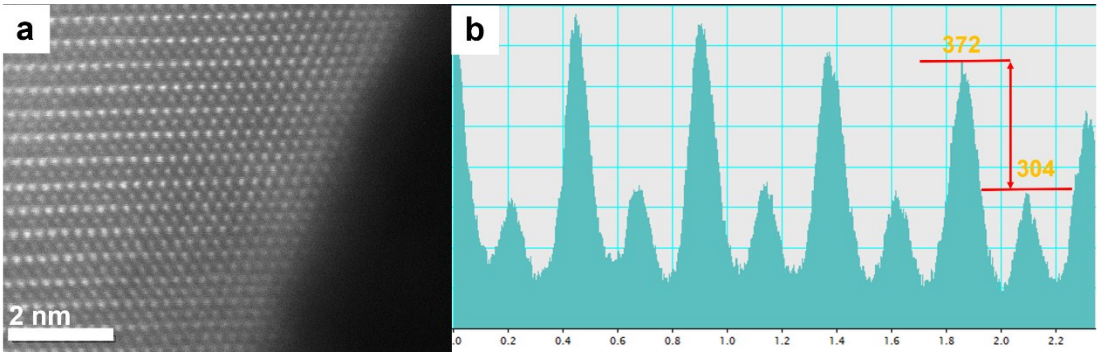

Figure S5. a) HAADF-STEM image of CLP form D5 surface. b) Corresponding line profiles of the Z-contrast information.

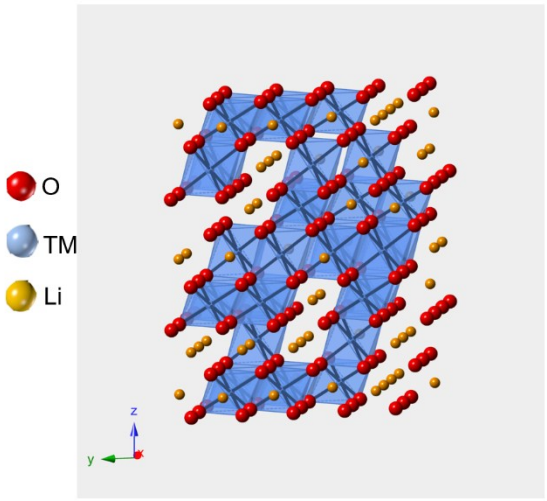

Figure S6. Atom model of Li-rich $Li_xTM_{2-x}O_2$ disordered layered structure (CLP).

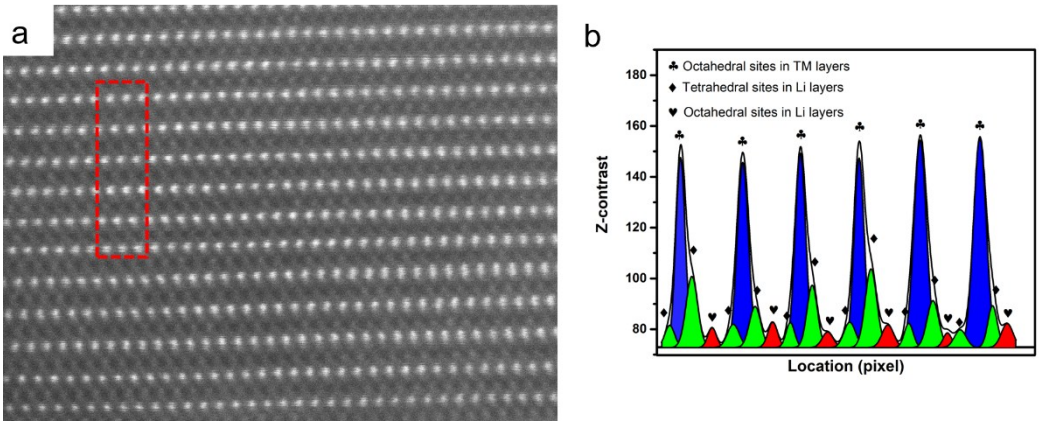

Figure S7. a) HAADF-STEM image of the margin of D30. b) Images of corresponding HAADF-STEM signal profiles (red rectangle) and its fitting peak curve. The blue peaks represent the TM-layers contrast; the green peaks are attributed TM-ions located in tetrahedral sites.