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Supplementary Information

Development of Diverse Aluminium Concentration Gradient Profiles in Ni-Rich Layered Cathodes for Enhanced Electrochemical and Thermal Performances

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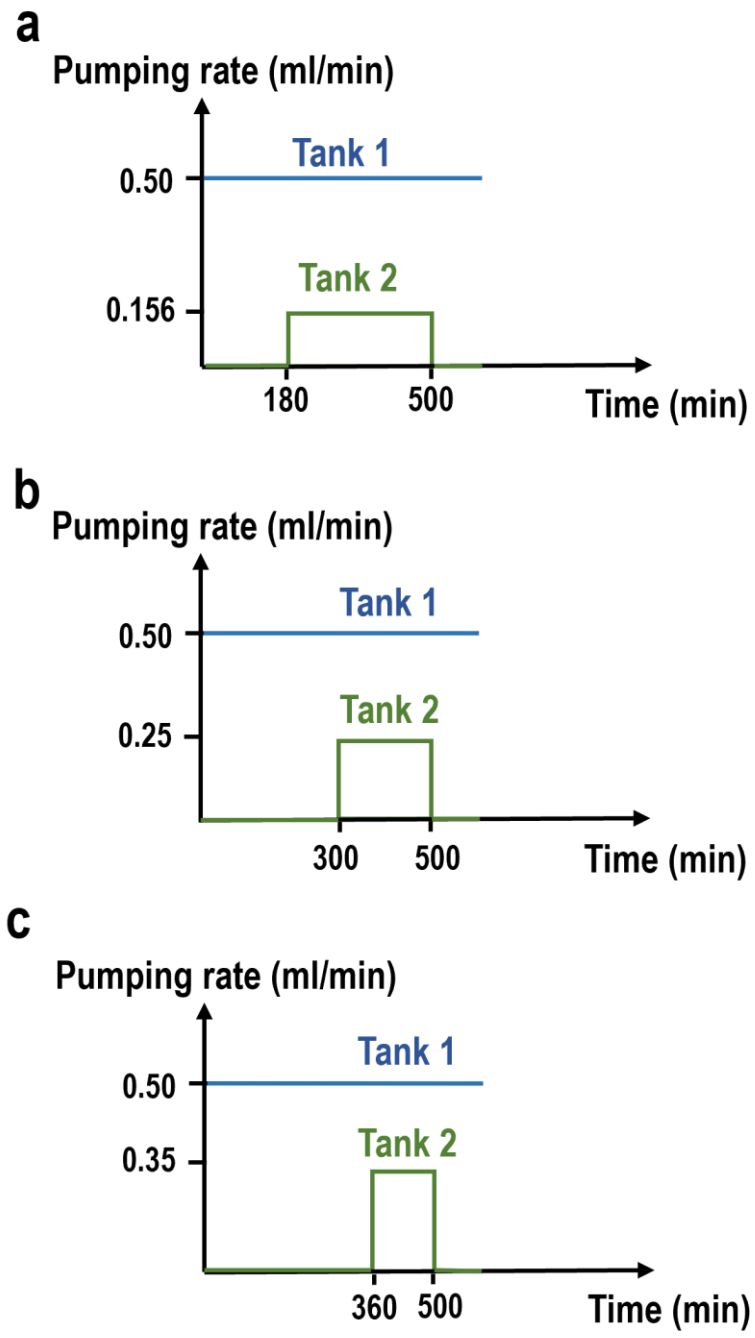

Fig. S1 The illustration of the injection timing of $Al_2(SO_4)_3 \cdot 12H_2O$ solution from Tank 2 (green), into Tank 1 at three different intervals following the initial injection of $NiSO_4 \cdot 6H_2O$ (blue) at the beginning of the synthesis: after (a) 3 h, (b) 5 h, and (c) 6 h.

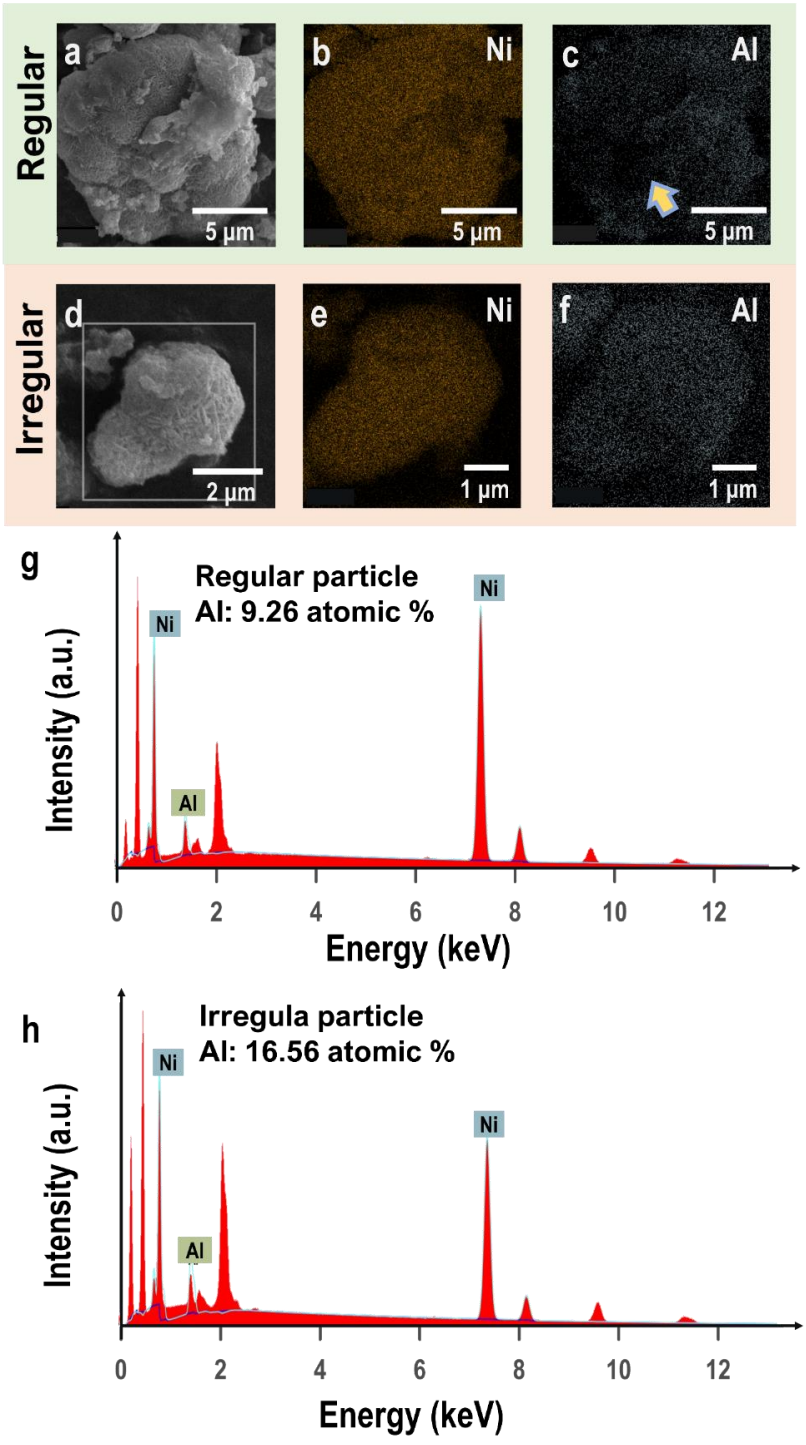

Fig. S2 SEM and elemental mapping images of (a, b, c) large and (d, e, f) small particles from concentration-gradient $Ni_{0.95}(OH)_{1.9} – Al_{0.05}(OH)_{0.15}$ precursors, obtained by delaying Al-injection schedule to 6 h. EDS spectra obtained from the (g) large particle and (h) small particles.

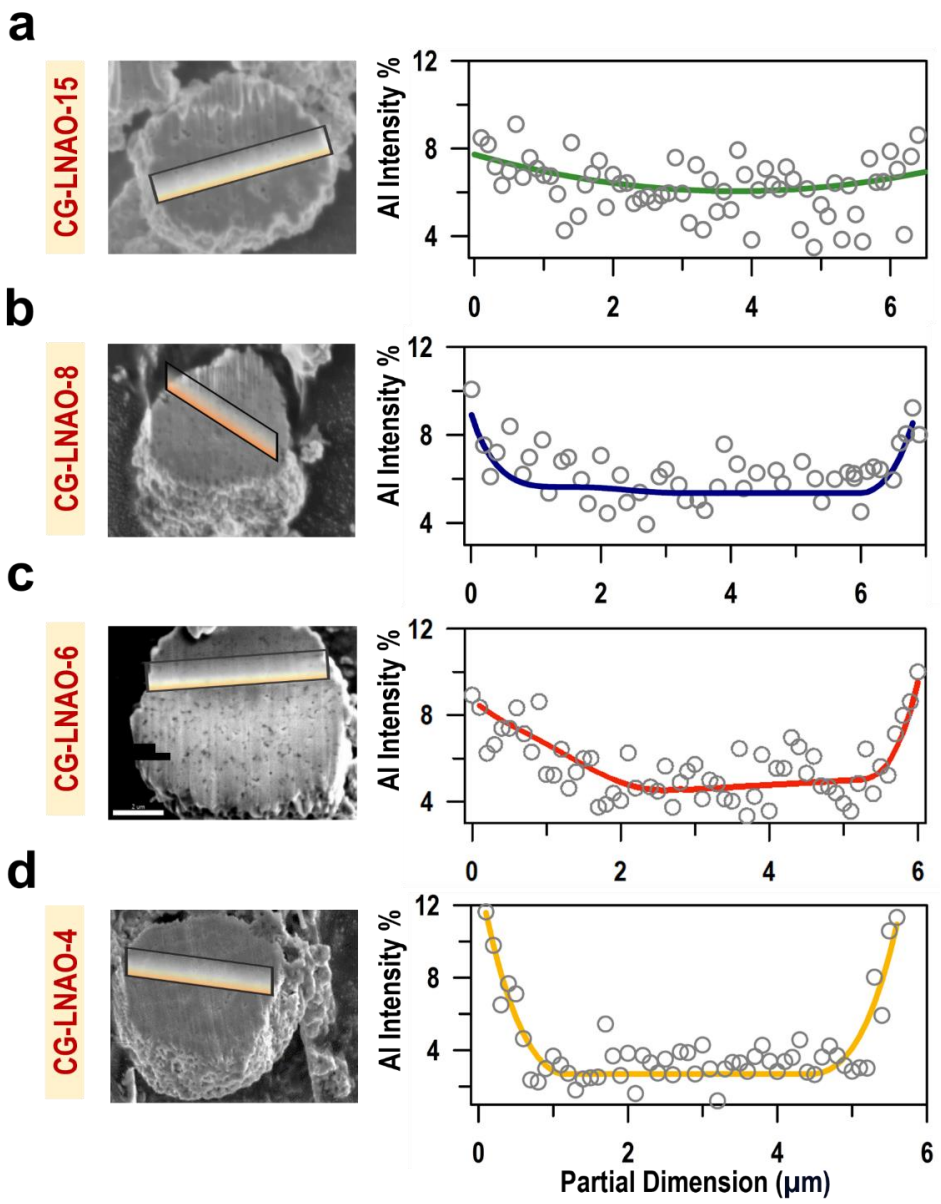

Fig. S3 Cross-sectional EDS line scan analyses of CG-LNAO particles sintered at 710 ^{o}C in O_2 for (a) 15 h, (b) 8 h, (c) 6 h, and (d) 4 h.

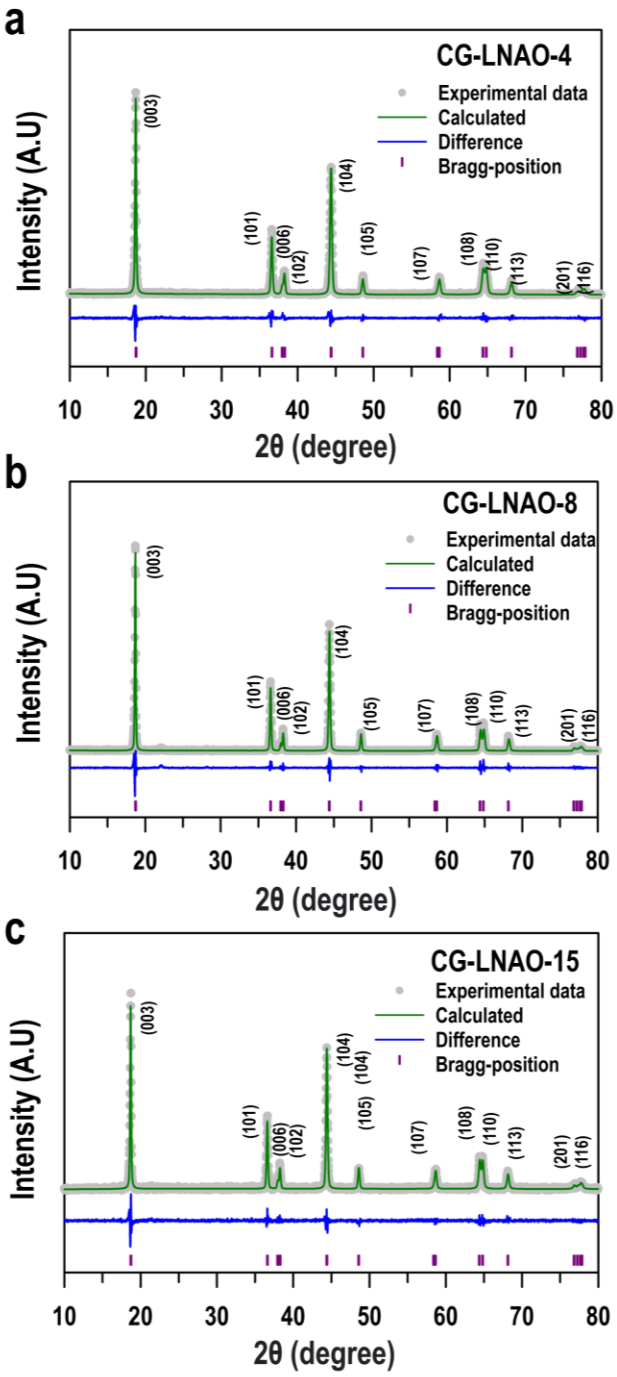

Fig. S4 XRD data and Rietveld refinement profiles of (a) CG-LNAO-4, (b) CG-LNAO-8, and (c) CG-LNAO-15.

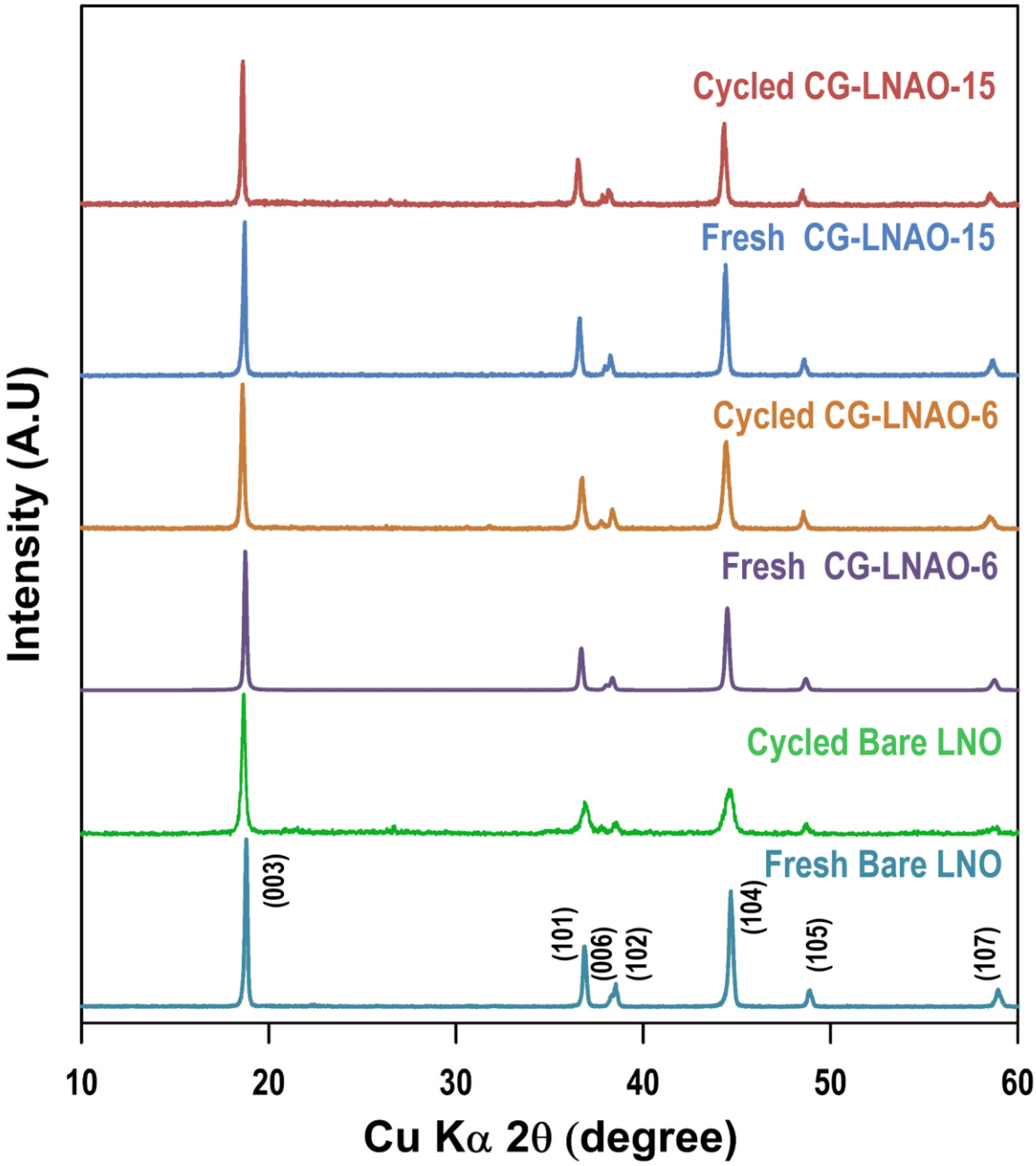

Fig. S5 XRD pattern of fresh and cycled bare $LiNiO_2$, CG-LNAO-6 and CG-LNAO-15 cathodes.

Table S1. Structural parameters of various cathode materials.

Samples	a (Å)	c (Å)	c/a ratio	Unit cell volume ($Å^3$)	Ni^{2+} in Li Layer (%)†	$I_{(003)}/I_{(104)}$ Ratio	$(R_{wp}/R_{exp})^2$
Bare LNO	2.878	14.187	4.931	101.676	4.95	1.37	2.12
CG-LNAO 4h	2.877	14.213	4.940	101.819	2.77	1.56	1.85
CG-LNAO 6h	2.875	14.212	4.944	101.748	2.19	1.63	1.33
CG-LNAO 8h	2.874	14.212	4.945	101.689	2.65	1.61	1.68
CG-LNAO 15h	2.877	14.211	4.939	101.761	3.12	1.41	1.32

†Referred to as cation mixing.

Table S2. Elemental composition of various cathodes measured by ICP-MS.

Samples	Li	Al	Ni
Bare LNO	0.984	0.0	1.0
CG-LNAO 4 h	1.021	0.052	0.948
CG-LNAO 6 h	1.002	0.056	0.944
CG-LNAO 8 h	1.002	0.053	0.947
CG-LNAO 15 h	0.986	0.060	0.940