In Situ Synthesis of Nickel Concentration Gradient Structure Promising Superior Electrochemical Properties of Ni-Rich $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ at High Cut-Off Voltage

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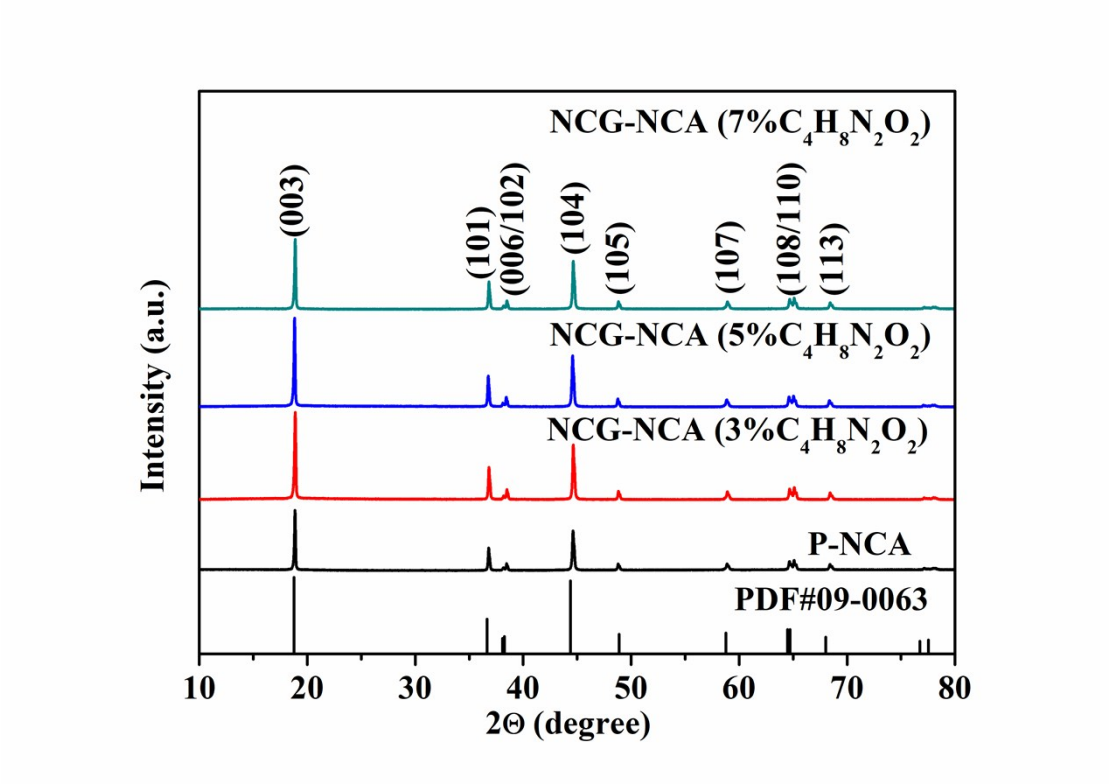

Fig. S1 XRD patterns of P-NCA and NCG-NCA (x% $C_4H_8N_2O_2$) materials and XRD ICDD card for P-NCA.

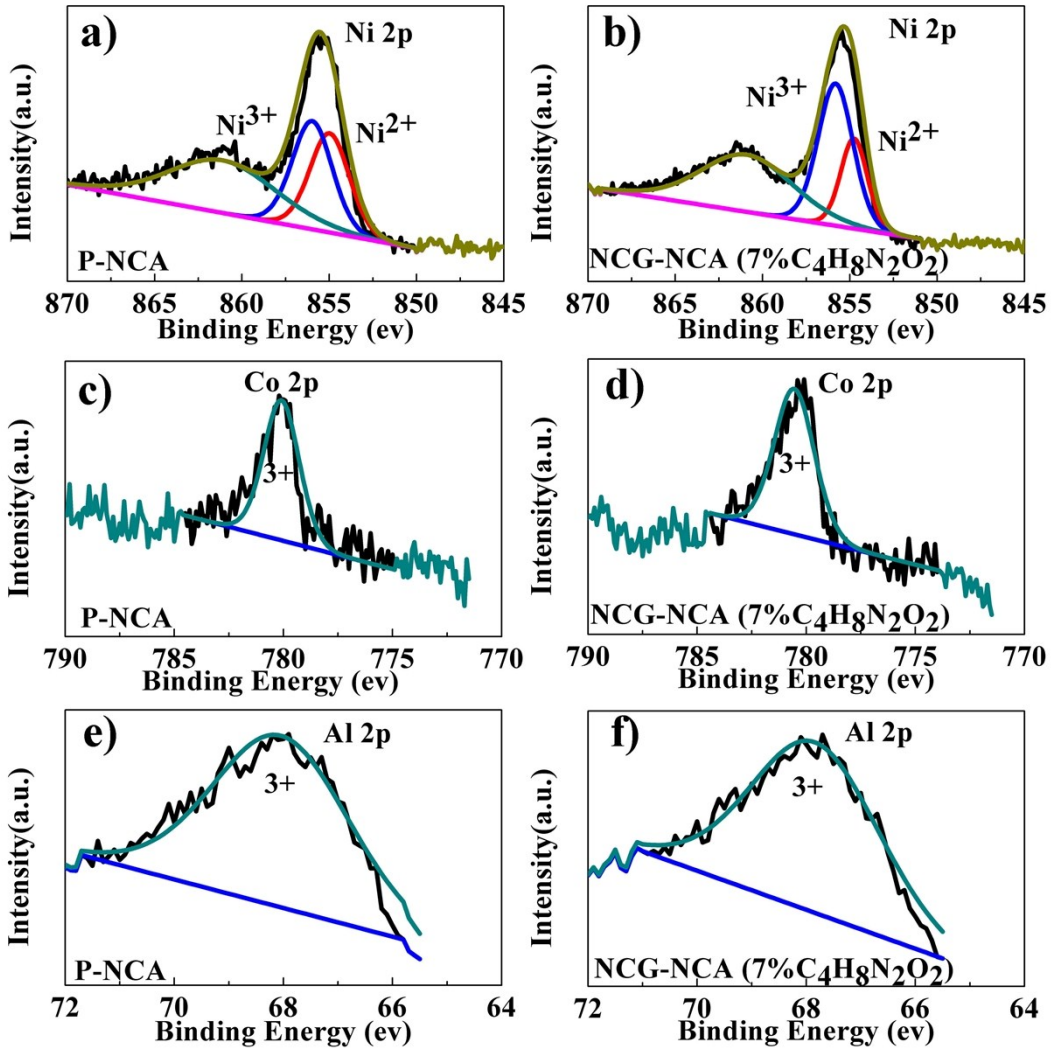

Fig. S2 X-ray photoelectron spectroscopy (XPS) spectra for the surface elements of the P-NCA and NCG-NCA (7% $C_4H_8N_2O_2$) materials.

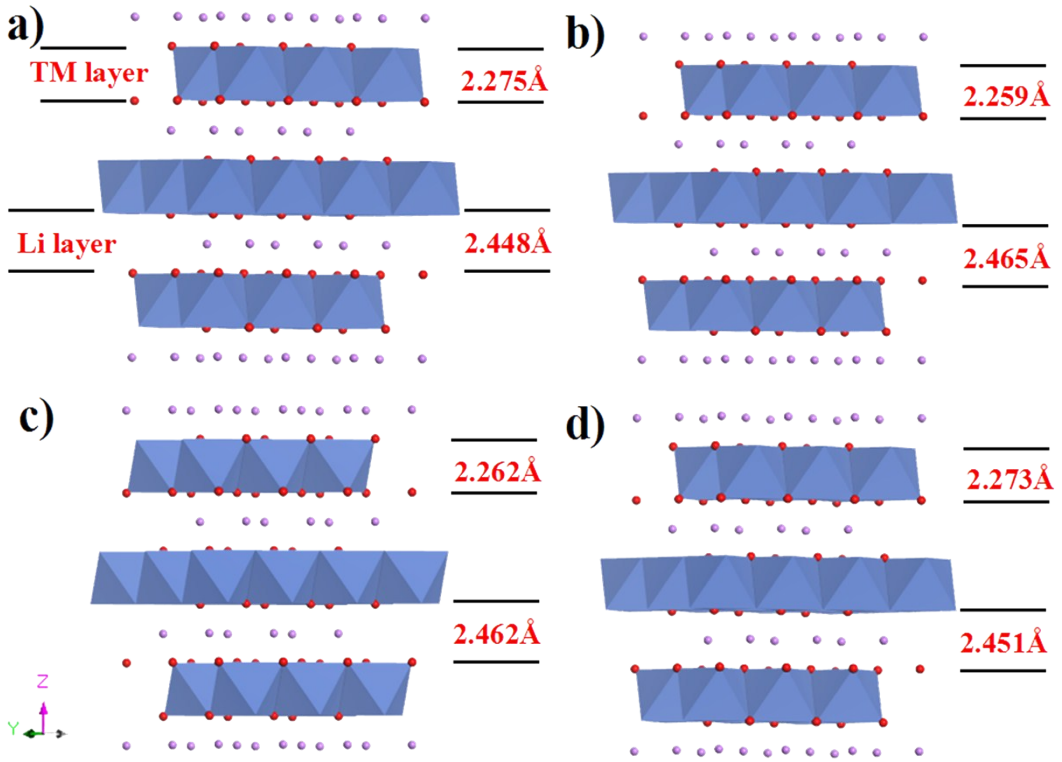

Fig. S3 The refined crystal ball and stick model structure of each material. The purple sphere is Li, the red sphere is O, the blue sphere is Ni/Co/Al. a) P-NCA, b) NCG-NCA (3% $C_4H_8N_2O_2$), c) NCG-NCA (5% $C_4H_8N_2O_2$), d) NCG-NCA (7% $C_4H_8N_2O_2$).

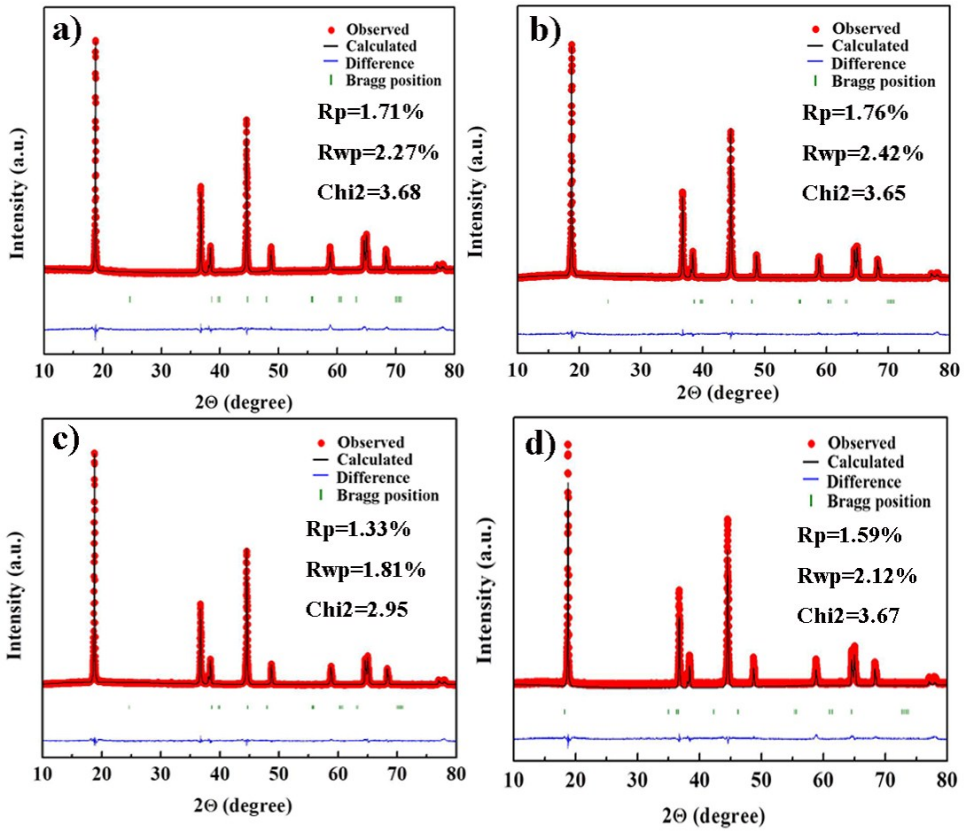

Fig. S4 The Rietveld refinement results of the X-ray diffraction patterns of a) P-NCA, b) NCG-NCA (3% $C_4H_8N_2O_2$), c) NCG-NCA (5% $C_4H_8N_2O_2$), d) NCG-NCA (7% $C_4H_8N_2O_2$).

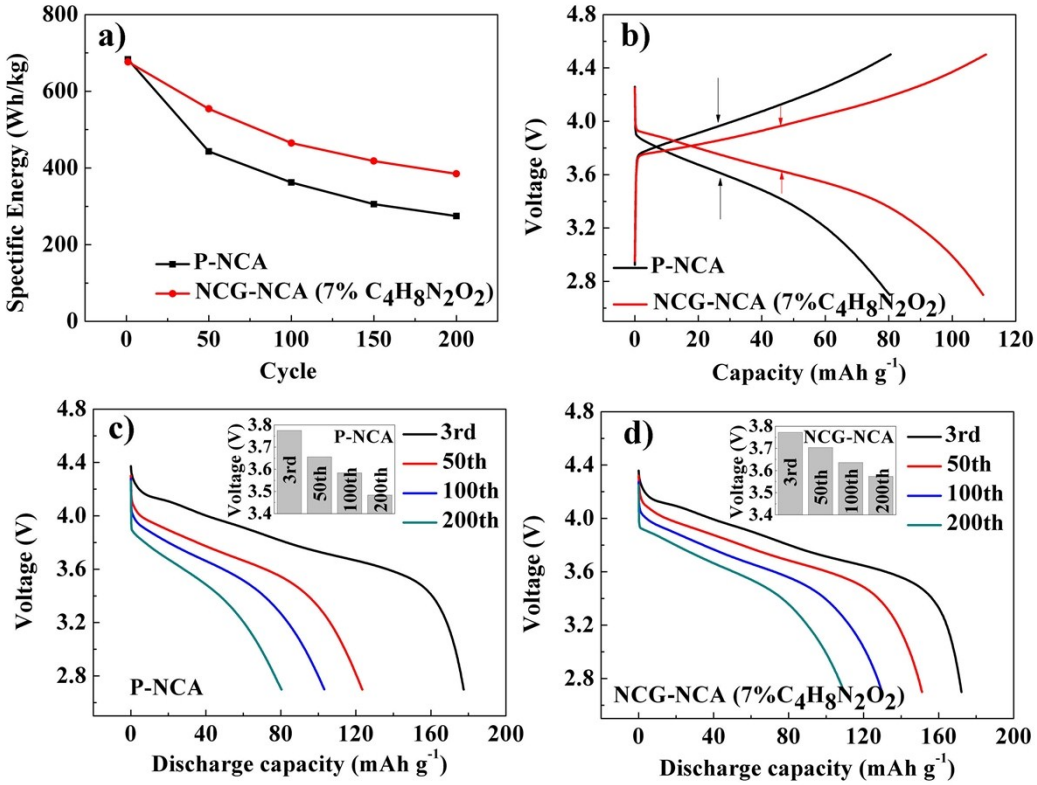

Fig. S5 a) Plots of specific energy vs. cycle number for the P-NCA and NCG-NCA (7% $C_4H_8N_2O_2$) electrodes at 1C. b) Capacity vs. voltage curves for the pristine P-NCA and NCG-NCA (7% $C_4H_8N_2O_2$) electrodes at the 100th cycle. Discharge capacity vs. voltage curve at the 3rd, 50th, 100th and 200th cycle at 1C for P-NCA c) and NCG-NCA (7% $C_4H_8N_2O_2$) d). The inset histograms suggest variation of the voltage after different cycles.

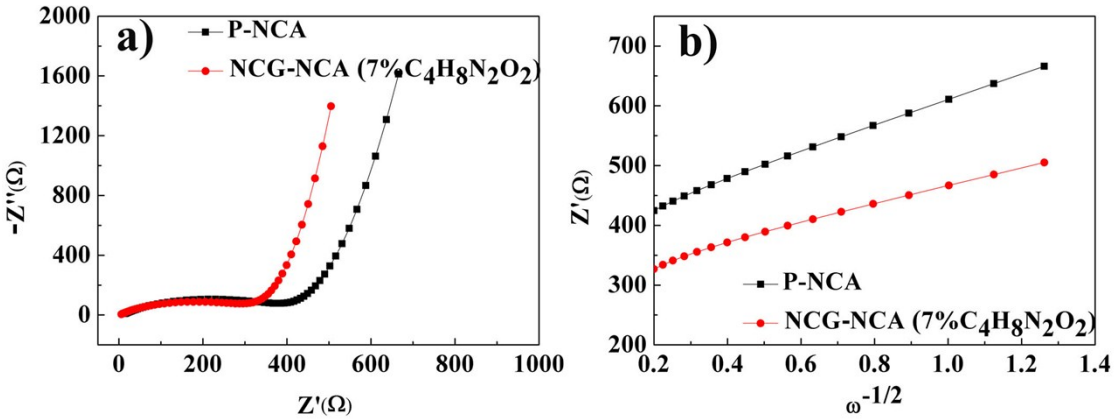

Fig. S6 a) Nyquist plots for the pristine P-NCA and NCG-NCA (7% $C_4H_8N_2O_2$) electrodes before cycling. b) The mathematical relationship between Z' and $\omega^{-1/2}$.

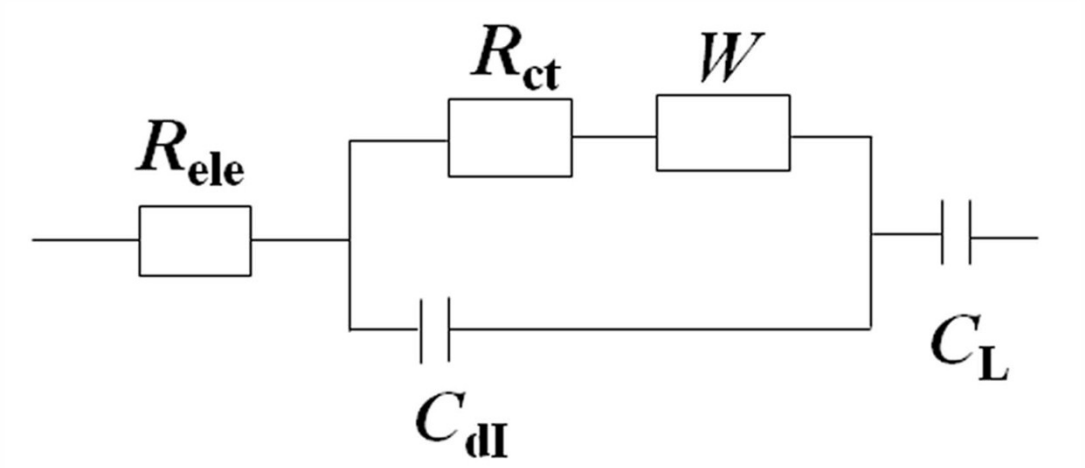

Fig. S7 Related equivalent circuit.

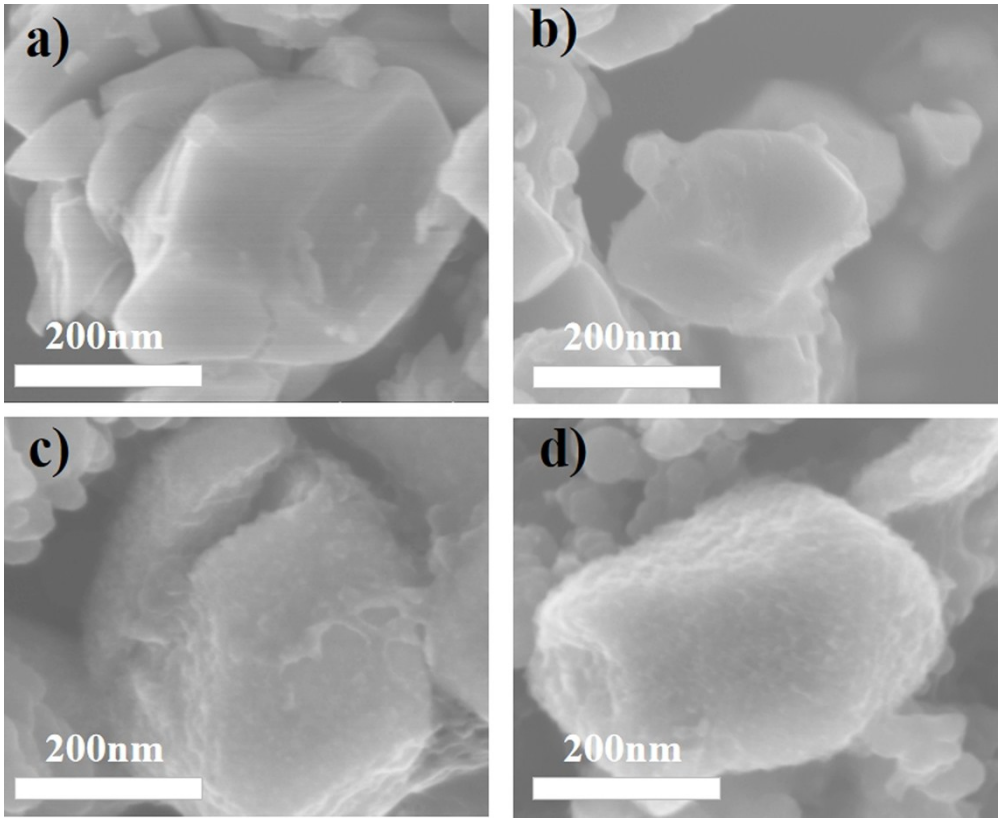

Fig. S8 The Scanning electron microscopy images of a,c) P-NCA and b,d) NCG-NCA (7% $C_4H_8N_2O_2$) materials collected at different stages: a,b) as-synthesized, c,d) after 100 cycles at 1C.

Table S1 The electron binding energy and peak area of the elements of P-NCA and NCG-NCA (7% $C_4H_8N_2O_2$) materials.

Sample	P-NCA	NCG-NCA
Ni^{2+} $2p_{3/2}$ (eV)	854.982	854.762
Ni^{3+} $2p_{3/2}$ (eV)	855.98	855.819
Al^{3+} $2p_{3/2}$ (eV)	68.0474	67.8449
Co^{3+} $2p_{3/2}$(eV)	780.09	780.549
Peak area/Ni^{2+} $2p_{3/2}$ (eV)	5206	5113.9
Peak area/Al^{3+} $2p_{3/2}$ (eV)	1807.8	2619.8
Peak area/Co^{3+} $2p_{3/2}$(eV)	1794.8	2601.9

Table S2 The cell parameters derived from Rietveld refinement of P-NCA, 3% $C_4H_8N_2O_2$-NCA, 5% $C_4H_8N_2O_2$-NCA and 7% $C_4H_8N_2O_2$-NCA.

Samples	a(Å)	c(Å)	V(Å^3)	Chi2	d-spacing(Å)
P-NCA	2.86650	14.16889	100.825	3.68	2.448
3% $C_4H_8N_2O_2$-NCA	2.86695	14.17180	100.878	3.65	2.465
5% $C_4H_8N_2O_2$-NCA	2.86691	14.17038	100.865	2.95	2.462
7% $C_4H_8N_2O_2$-NCA	2.86692	14.17114	100.871	3.67	2.451

Table S3 Pawley refinement results of P-NCA and NCG-NCA (7% $C_4H_8N_2O_2$) materials.

Sample	a (Å)	c (Å)	Rwp (%)	GOF
P-NCA	2.868892	14.178110	1.95	1.66
P-NCA-cycled	2.858419	14.230938	2.06	1.31
NCG-NCA	2.869698	14.178716	1.949	1.62
NCG-NCA-cycled	2.848906	14.176133	1.85	1.13

Table S4 The simulation results of EIS of the P-NCA and NCG-NCA (7% $C_4H_8N_2O_2$) materials.

Samples	P-NCA	NCG-NCA
Rele (Ω)	6.78	2.38
Rct (Ω)	406.04	333
Estimated error/Rele (%)	4.55	8.293
Estimated error/Rct (%)	1.01	1.783

Table S5 The linear fit for Z' (Y-axis) and $\omega^{-1/2}$ (X-axis) in low frequency simulating two lines of the P-NCA and NCG-NCA (7% $C_4H_8N_2O_2$) materials.

Equation	Y=a+b*x	
Plot	P-NCA	NCG-NCA
Intercept	232.49	174.60
Slope	378	291.82