

Neutron powder diffraction

Table S1 Structure parameters for $\text{Li}_x\text{Ni}_{0.77}\text{Co}_{0.14}\text{Al}_{0.09}\text{O}_2$ ($x = 0.87$ and 0.71).

g is the occupancy of the atom at the site. Space group: $R\bar{3}m$ (No.166).

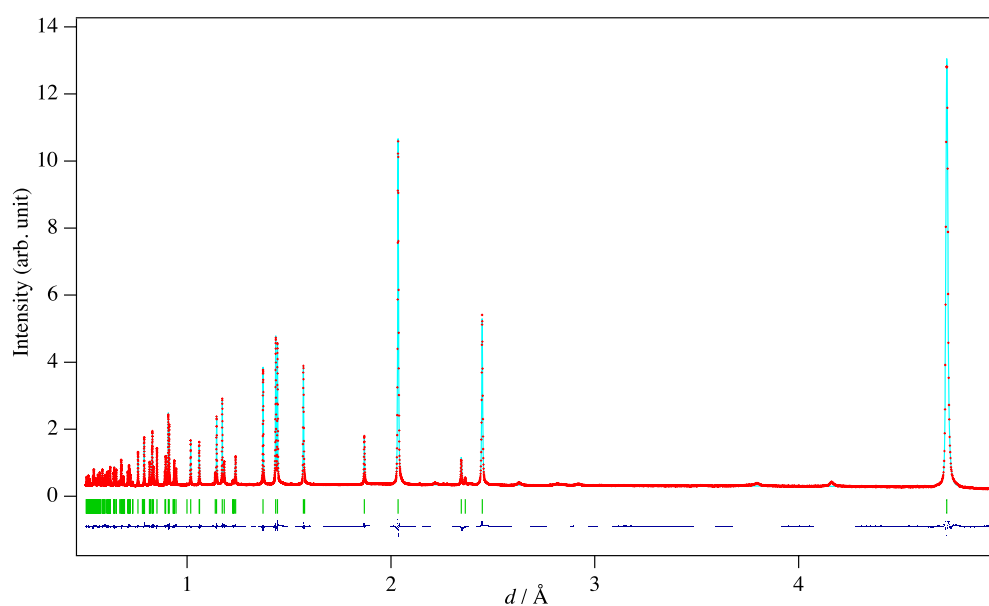
(a) $x = 0.87$. $a = b = 2.8697176(11)$ Å and $c = 14.187776(13)$ Å. $S = 4.72$, $R_{\text{wp}} = 2.66$ %, $R_e = 0.564$ %, $R_B = 2.32$ % and $R_F = 2.58$ %.

Atom	Label	site	g	x	y	z	B (Å)
Li	Li1	$3b$	0.8704(11)	0	0	1/2	1.169(8)
Co	Co2	$3a$	0.14	0	0	0	0.2754(15)
Ni	Ni2	$3a$	0.77	0	0	0	0.2754(15)
Al	Al2	$3a$	0.09	0	0	0	0.2754(15)
O	O1	$6c$	1	0	0	0.259125(5)	0.8472(18)

(b) $x = 0.71$. $a = b = 2.8688887(14)$ Å and $c = 14.182904(15)$ Å. $S = 4.94$, $R_{\text{wp}} = 3.09$ %, $R_e = 0.624$ %, $R_B = 1.70$ % and $R_F = 2.79$ %.

Atom	Label	site	g	x	y	z	B (Å)
Li	Li1	$3b$	0.7080(12)	0	0	1/2	1.391(10)
Co	Co2	$3a$	0.14	0	0	0	0.3457(16)
Ni	Ni2	$3a$	0.77	0	0	0	0.3457(16)
Al	Al2	$3a$	0.09	0	0	0	0.3457(16)
O	O1	$6c$	1	0	0	0.259113(5)	0.9022(19)

(a)



(b)

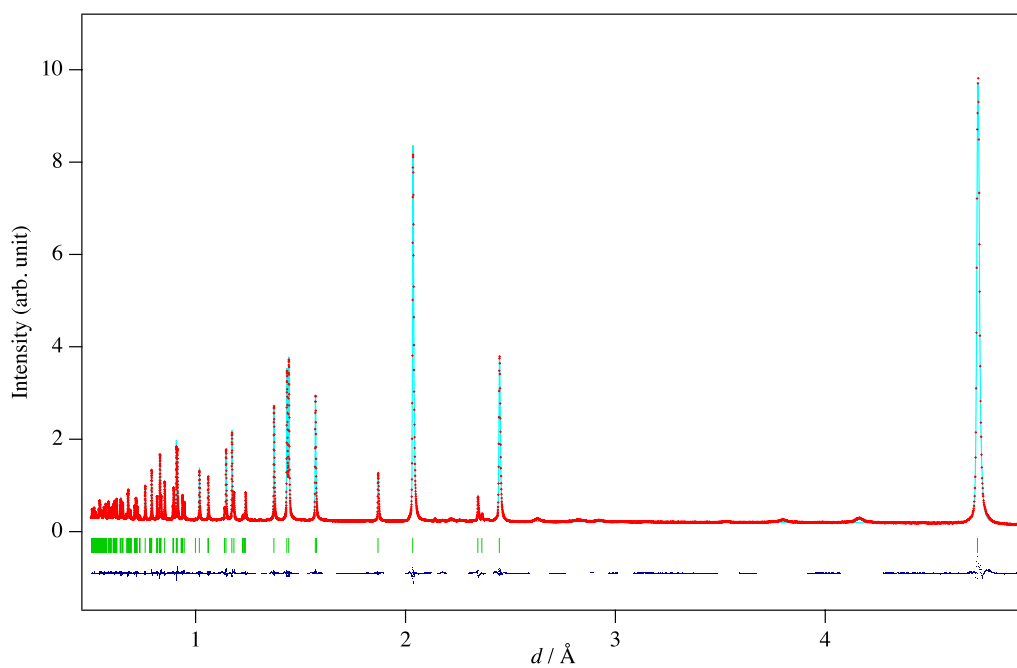


Figure S1: Rietveld refinement patterns of $\text{Li}_x\text{Ni}_{0.77}\text{Co}_{0.14}\text{Al}_{0.09}\text{O}_2$: (a) $x = 0.87$, (b) $x = 0.71$. Observed intensity data are shown by + marks, and the solid line overlying them is the calculated intensity. Vertical markers below the diffraction patterns indicate positions of possible Bragg reflections. Differences between the observed and calculated intensities are plotted at the bottom in the same scale. Some regions with small unknown peaks are removed from refinements.

Electrochemical impedance spectroscopy

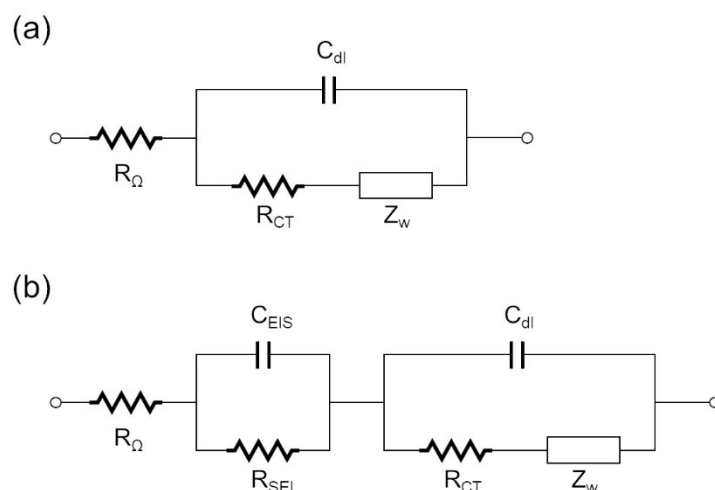


Figure S2: Modified Randles cell models for (a) NCA pre-cycling and (b) NCA post-cycling including contributions from solid electrolyte interphase formation.

Continuous hydrothermal flow synthesis of NCA precursor

Figure S3 shows a simplified flow diagram of the CHFS process used for the synthesis of the nanoparticles. In general, three diaphragm pumps (Primeroyal K, Milton Roy, Pont-Saint-Pierre, France) were used to supply feeds of deionized water (P_{DI}), an aqueous solution 0.4 M $Ni(NO_3)_2 \cdot 6H_2O$, 0.15 M $Co(NO_3)_3 \cdot 6H_2O$ and 0.05 M $Al(NO_3)_3 \cdot 9H_2O$ (Pump 2), and an aqueous solution of 1.5 M KOH (P3). The deionized water feed from Pump 1 was heated in flow to 450 °C using a 7 kW custom-built in-line electrical heater. Precursor feeds from pumps Pump 2 and Pump 3 were separately mixed in a T-piece (at room temperature) prior to mixing with the combined superheated deionized water feed inside the Confined Jet Mixer (CJM) in flow. The design of the CJM is detailed in previous publications of the authors, and in patent GB1008721.³¹ In all cases, the total concentration of metal nitrates was maintained at 0.5 M and KOH at 1.5 M. The particle-laden flow in the CHFS process was then cooled to ~40°C

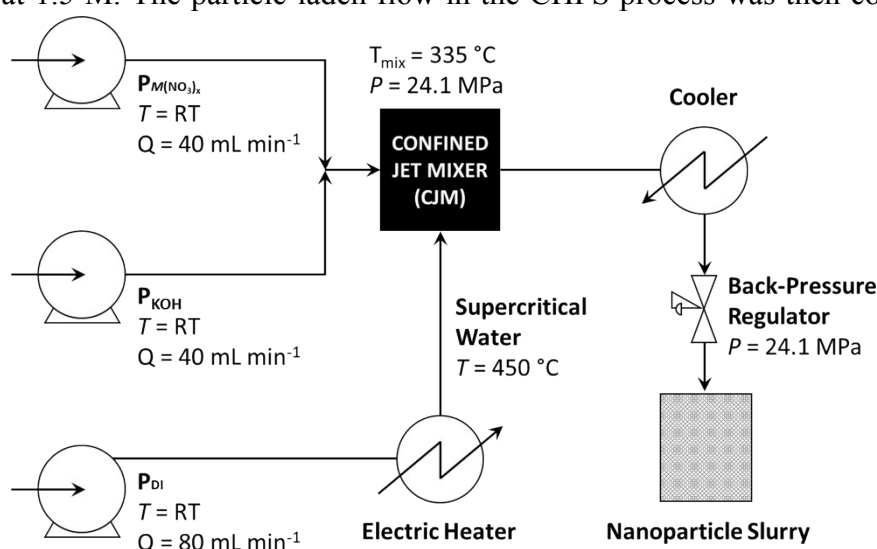


Figure S3: Diagram depicting the continuous hydrothermal flow synthesis (CHFS) employed to synthesise the CHFS-NCA precursor.

using a 1.5 m pipe-in-pipe heat exchanger before the slurry passed through a back-pressure regulator (BPR) valve. The aqueous nanoparticle slurry exiting the BPR was collected in a beaker and then cleaned by repeated centrifugation and washing with deionized water until the conductivity of the supernatant was consistently below 50 μS , as measured by a conductivity probe (Hanna Instruments, model HI98311). The concentrated slurry was then freeze-dried (Virtis Genesis 35XL) by slowly heating a sample from -60 to 25 $^{\circ}\text{C}$ (over 24 h) in vacuo (<100 mTorr), which yielded a free-flowing, brown nanoparticulate powder.