Electronic Supplementary Information (ESI)

Kinetic stabilization of topotactically transformed texture morphology via doping in Ni-rich lithium layered oxide

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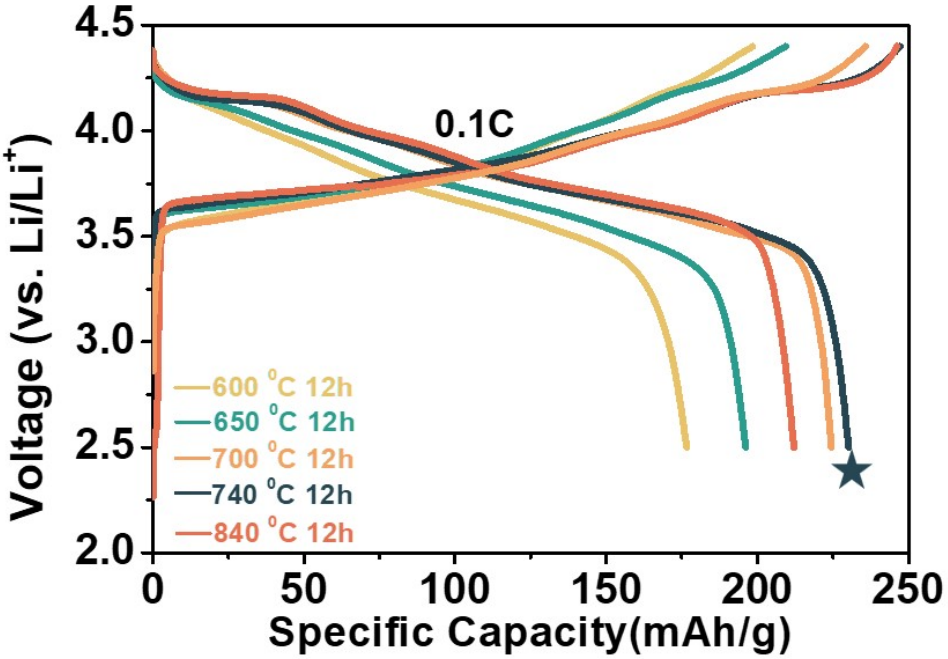

Fig. S1 Initial charge-discharge voltage profiles of U-LNCM electrodes as a function of calcination temperature. The electrodes were cycled within the voltage range of 2.5 ~ 4.4 V (vs. Li/Li^+). 1 C represents 200 mA g^{-1}. The electrode prepared with the powder calcinde at 740 ºC for 12 h showed the highest discharge capacity (~230 mAh g^{-1}) and thus, 740 ºC was selected as an optimum syntheis temperature.

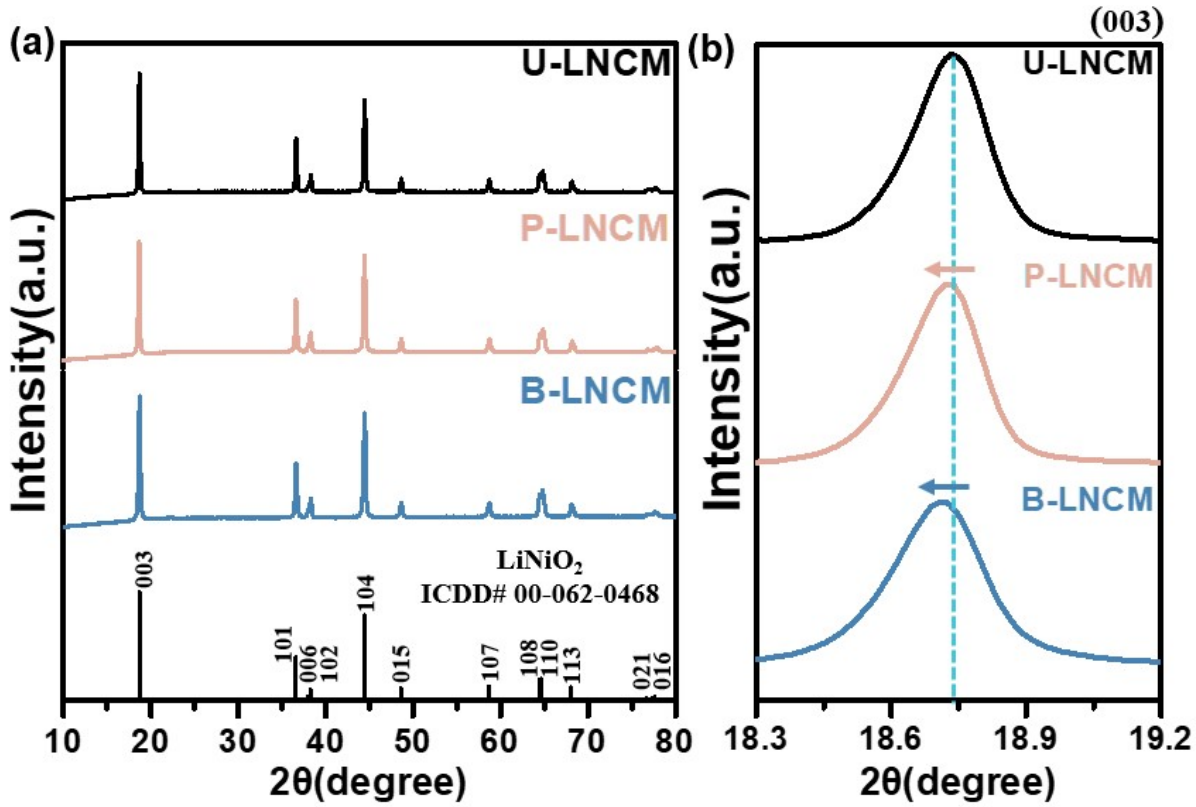

Fig. S2 (a) X-ray diffraction patterns of as-synthesized U-LNCM, P-LNCM, and B-LNCM powders calcined at 740 ºC for 12 h and (b) magnified view of (003) peaks of (a).

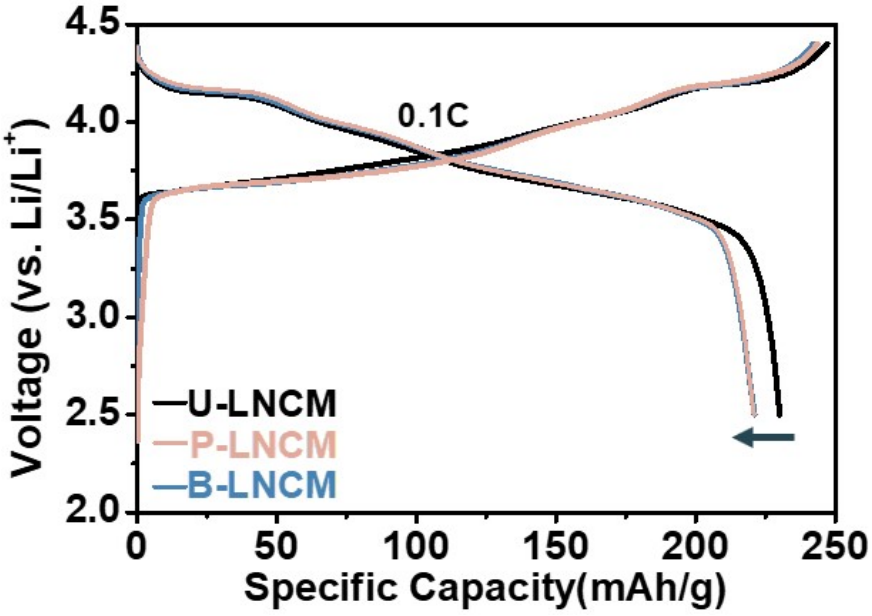

Fig. S3 Initial charge-discharge voltage profiles of three electrodes prepared with the LNCM powders calcined at 740 ºC for 12 h.

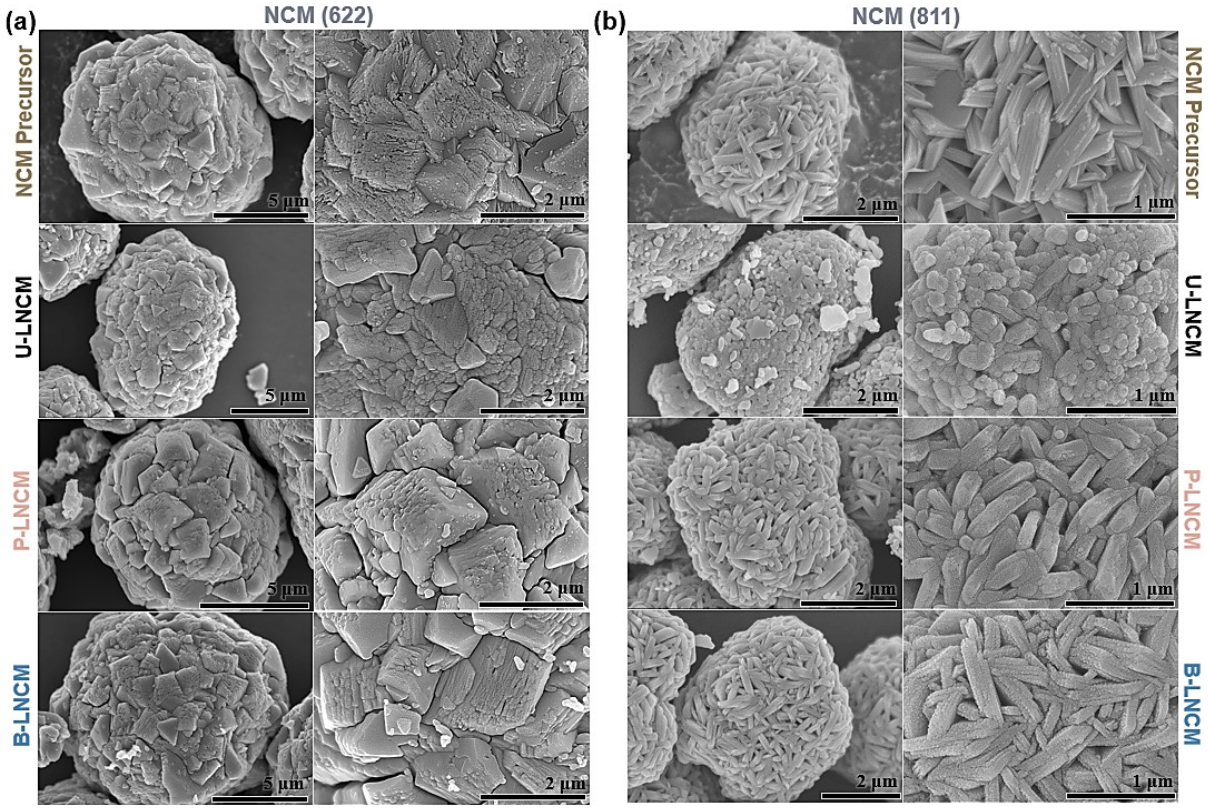

Fig. S4 Microstructure comparison of NCM precursor and as-synthesized U-LNCM, P-LNCM, and B-LNCM made from (a) NCM 622 and (b) NCM 811 precursors.

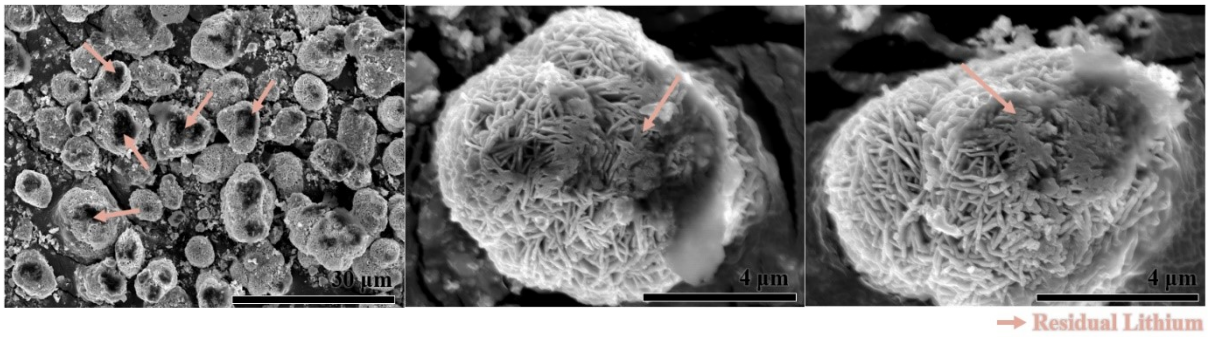

Fig. S5 SEM images of ETOH unwashed U-LNCM calcined at 740 ºC for 0 min. The arrows indicate the residual lithium. When synthesized at a low temperature or for a short calcination time, the residual lithium remained on the particle surface. To clearly view the morphology evolution, the residual lithium was washed by ETOH before the SEM analysis.

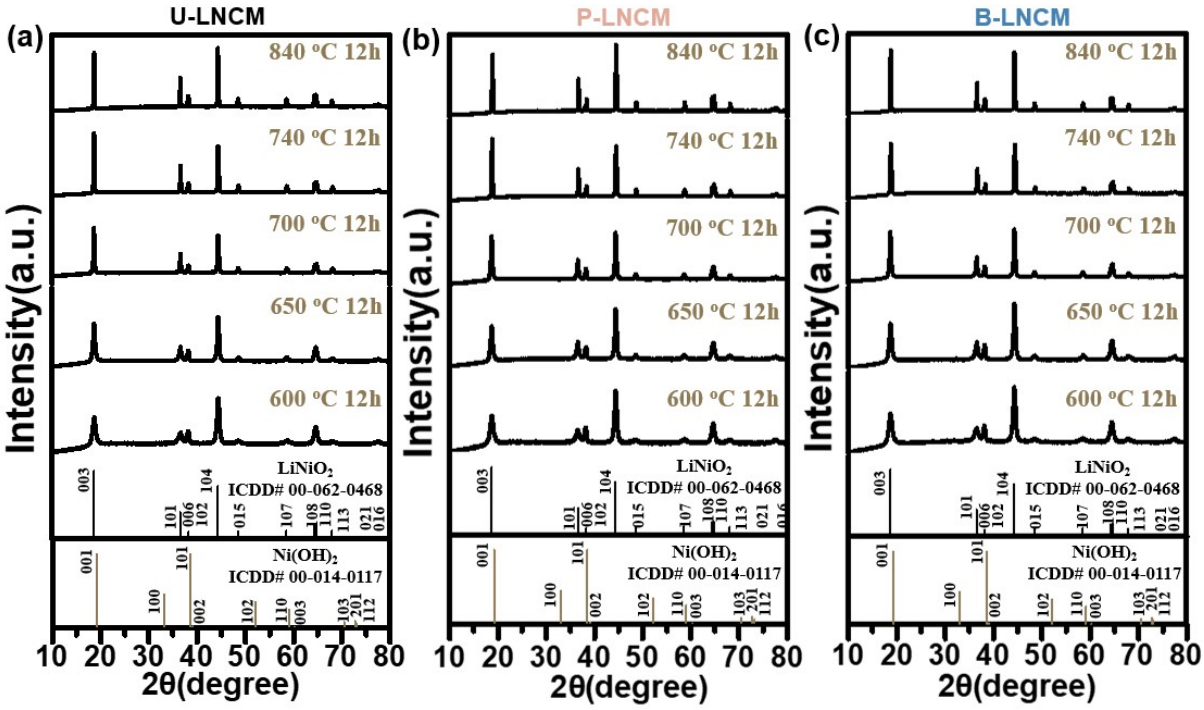

Fig. S6 *Ex-situ* XRD patterns of as-synthesized (a) U-LNCM, (b) P-LNCM, and (c) B-LNCM powders as a function of calcination temperature. Calcination temperature was varied from 600 to 840 ºC, while the calcination time was fixed to 12 h.

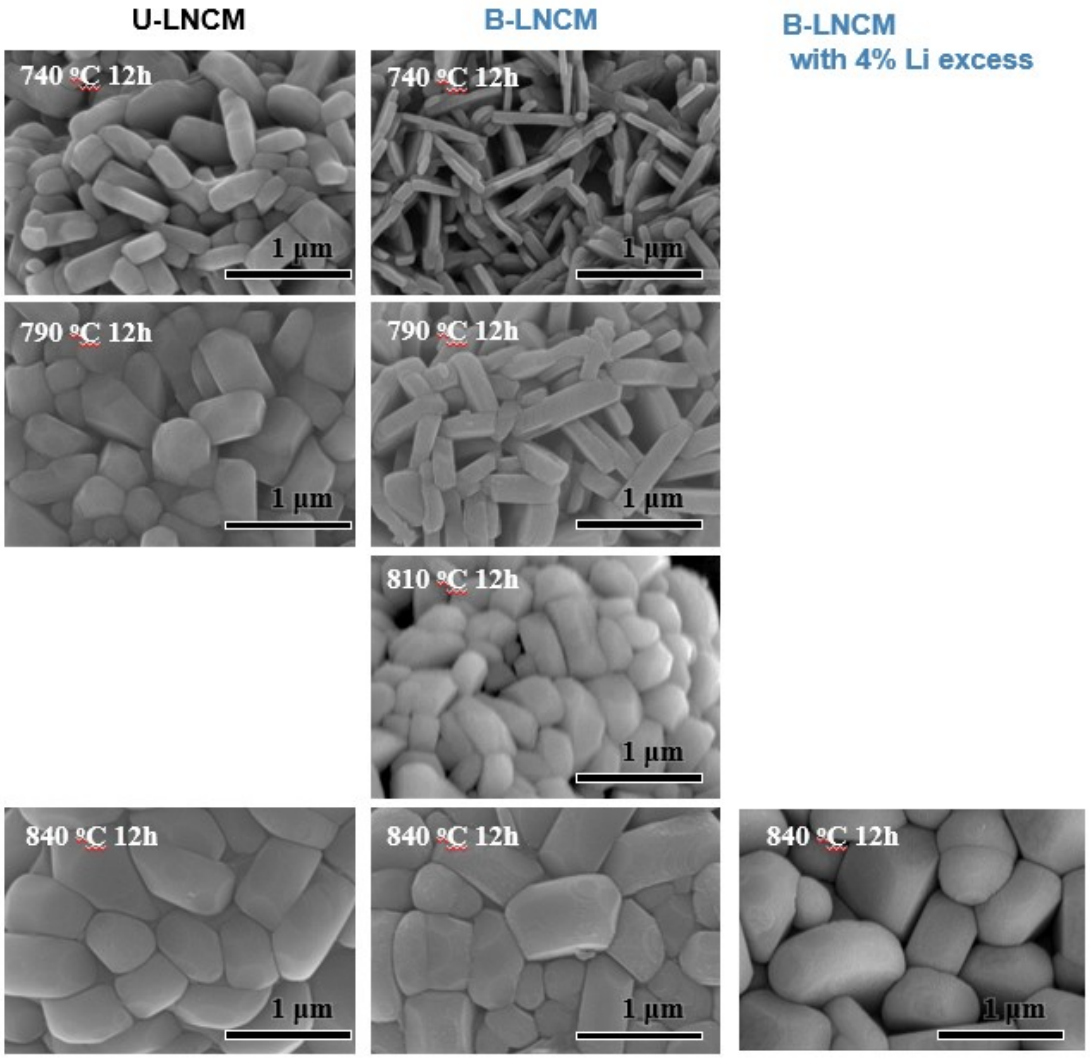

Fig. S7 SEM micrographs of undoped and B-doped LNCMs calcined at 740, 790, 810, and 840 °C for 12 h. 4 wt% excess of lithium was added to compensate the volatilization loss, but the morphology was unaffected.

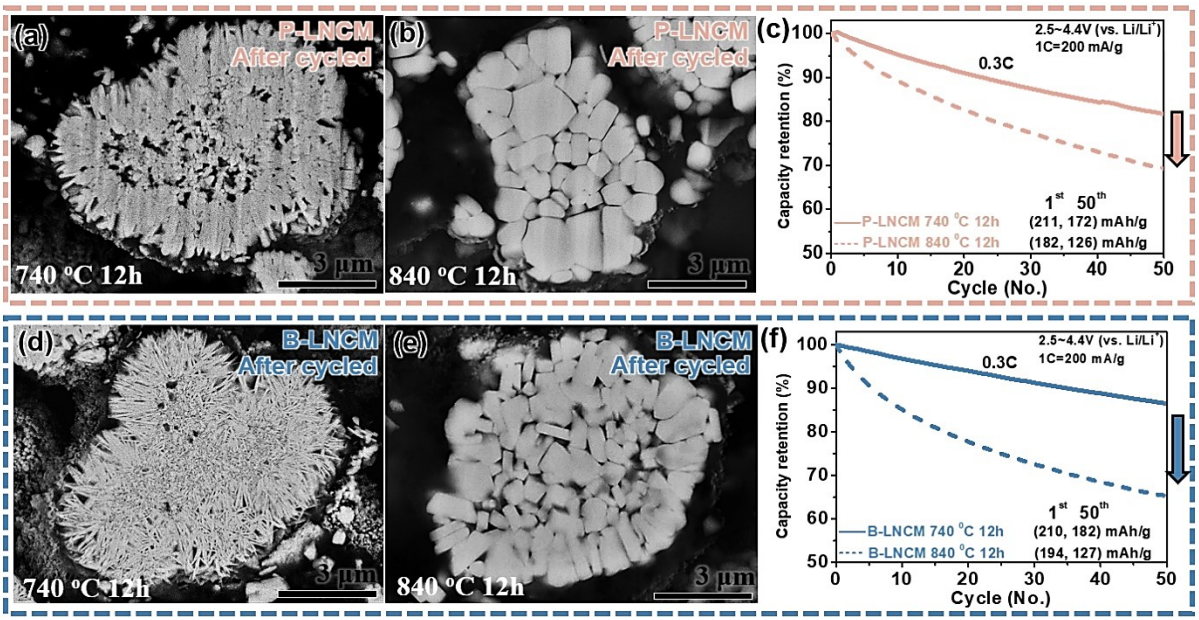

Fig. S8 CP-SEM images of after 50th cycled (a) P-LNCM (740 ºC, 12 h), (b) P-LNCM (840 ºC, 12 h), (d) B-LNCM (740 ºC, 12 h), and (e) B-LNCM (840 ºC, 12 h) electrodes. Cycle performance of (c) P-LNCM and (f) B-LNCM electrodes prepared with the powders calcined at 740 and 840 ºC for 12 h.

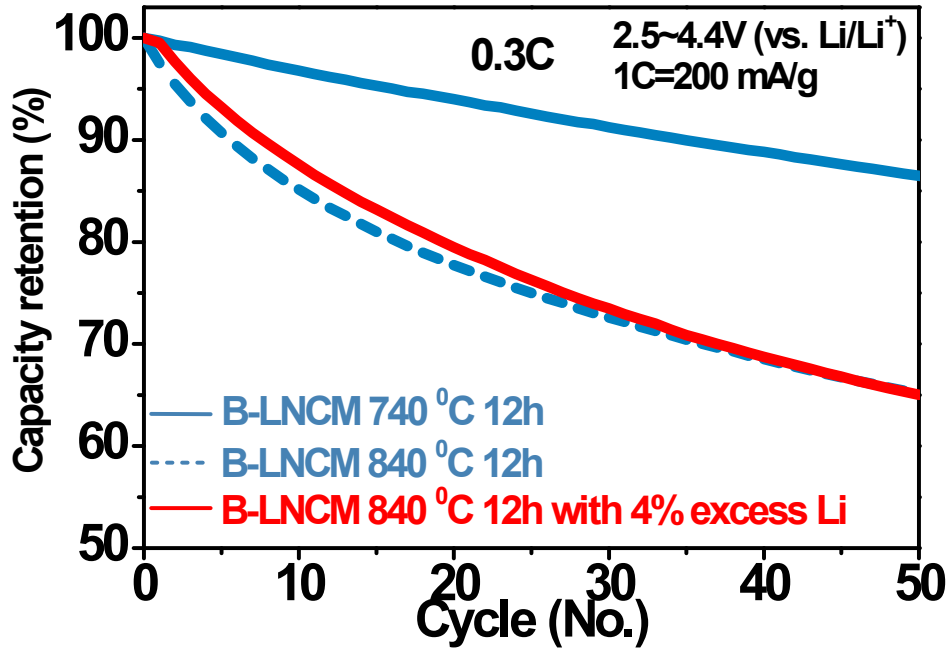

Fig. S9 Cycle performance of B-LNCM electrodes prepared with the powders calcined at 840 ºC for 12 h with/without 4 % Li excess.

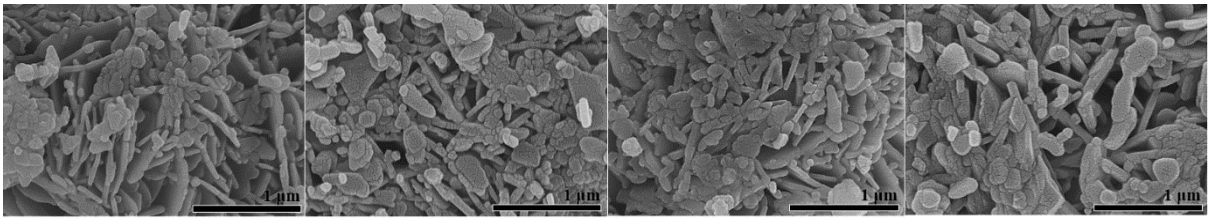

Fig. S10 Another evidence of recrystallization in U-LNCM annealed at 740 ºC for 0 min, which shows the combination of initial plate-like particles and newly formed ellipse-like particles. All SEM micrographs were taken from same U-LNCM clacined at 740 ºC for 0 min.

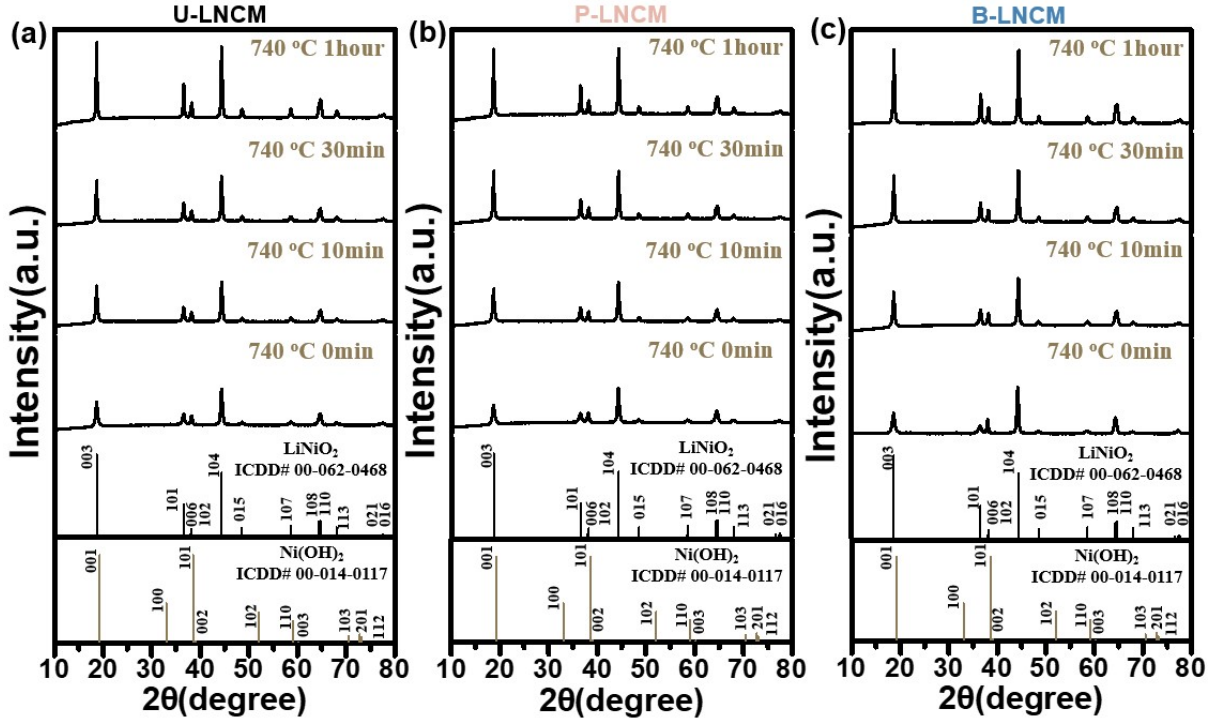

Fig. S11 *Ex-situ* XRD patterns of as-synthesized (a) U-LNCM, (b) P-LNCM, and (c) B-LNCM powders as a function of calcination time. Calcination time was varied from 0 min to 1 h, while the calcination temperature was fixed to 740 ºC.

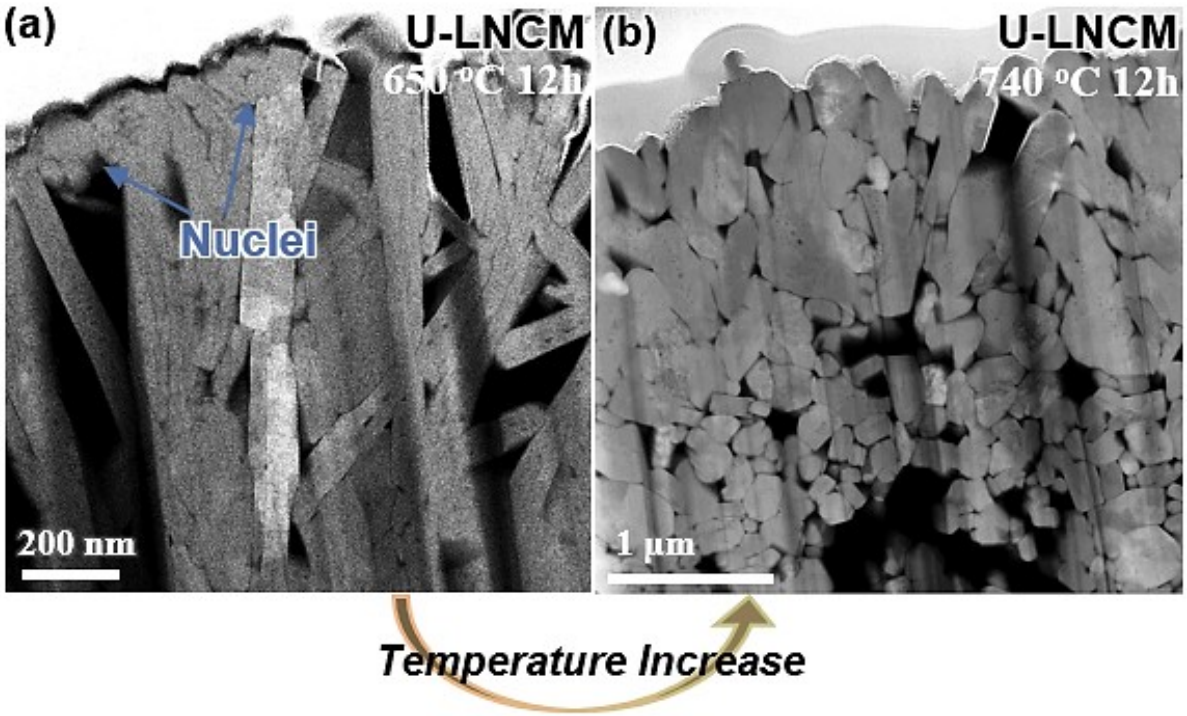

Figure S12 HRTEM images of FIB cross-sectioned U-LNCM annealed (a) at 650 ºC for 12h and (b) at 740 ºC for 12h.

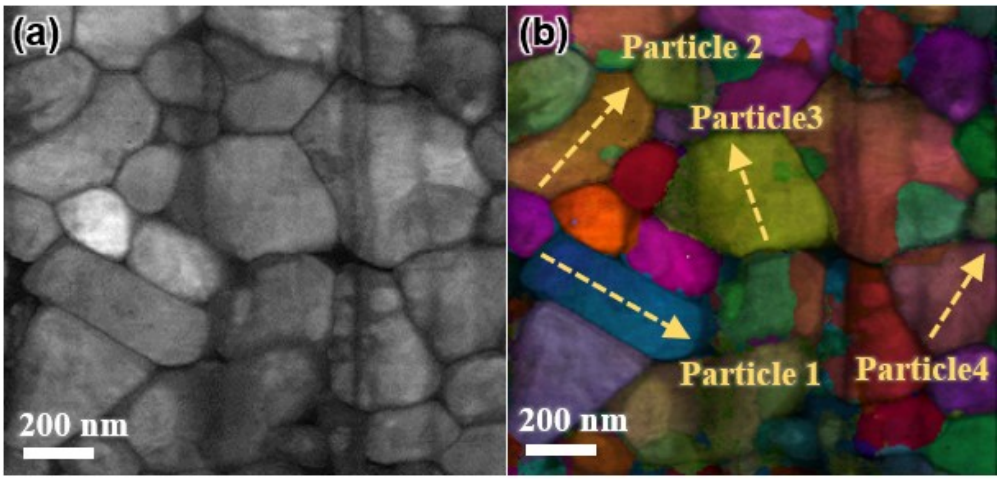

Fig. S13 (a) Index map of U-LNCM (calcined at 740 ºC for 12 h) aquired by ASTAR device and (b) its corresponding color-coded crystallographic orientation map.

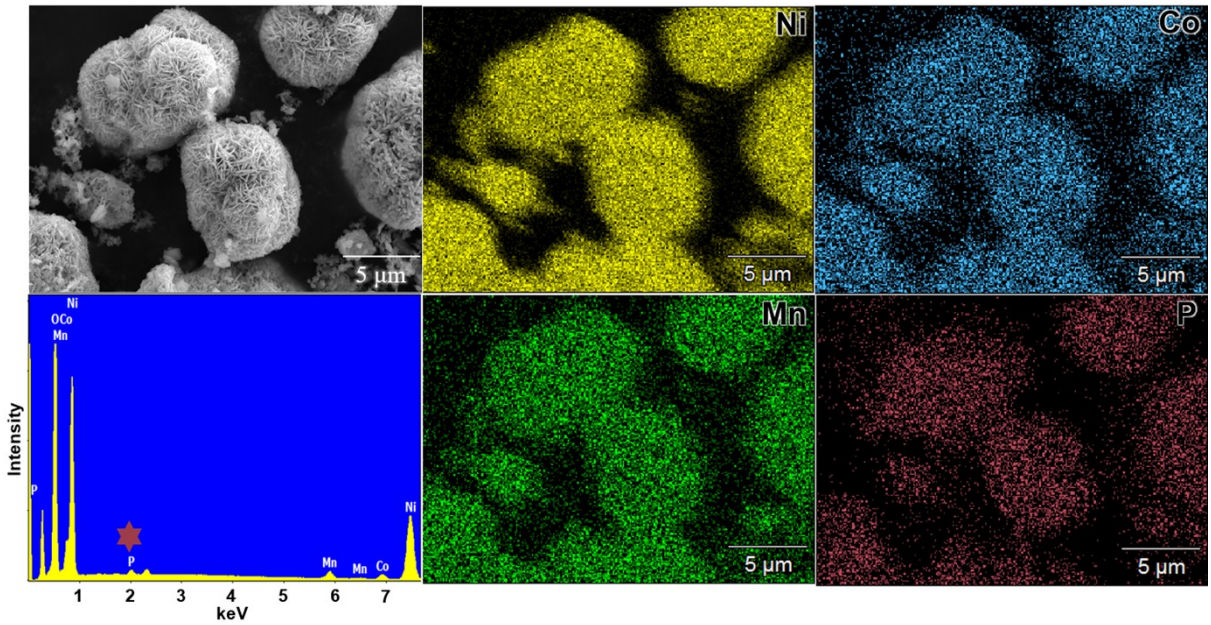

Fig. S14 Energy-dispersive X-ray spectroscopy (EDS) mapping of as-synthesized P-LNCM calcined at 740 ºC for 12h.

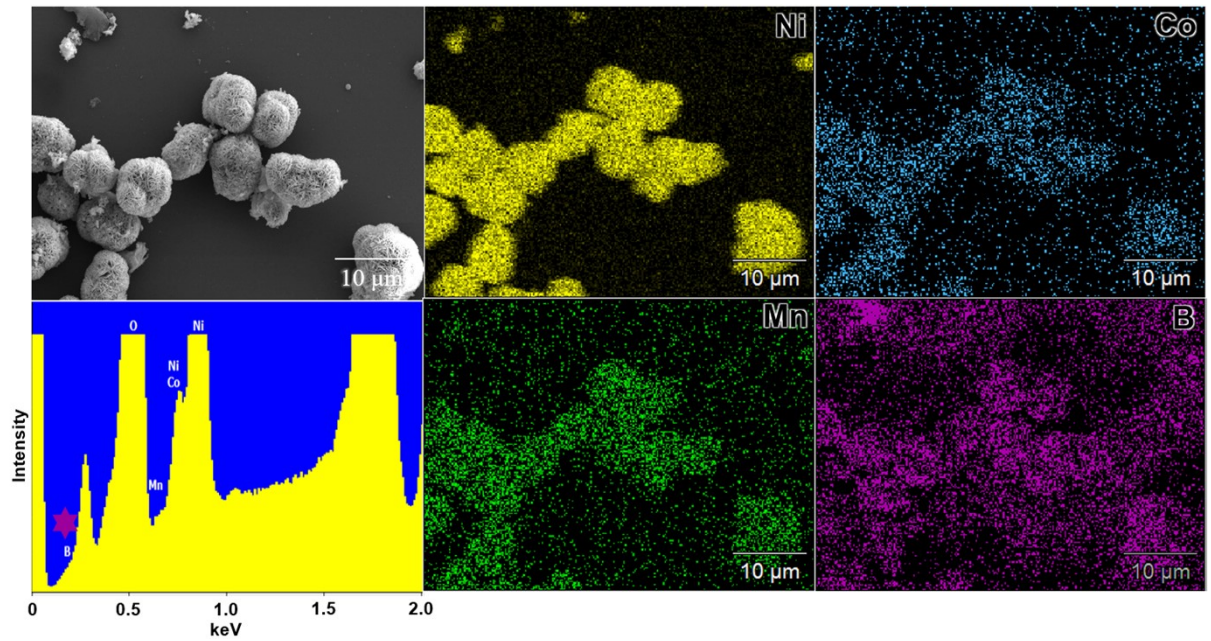

Fig. S15 Energy-dispersive X-ray spectroscopy (EDS) mapping of as-synthesized B-LNCM calcined at 740 ºC for 12 h.

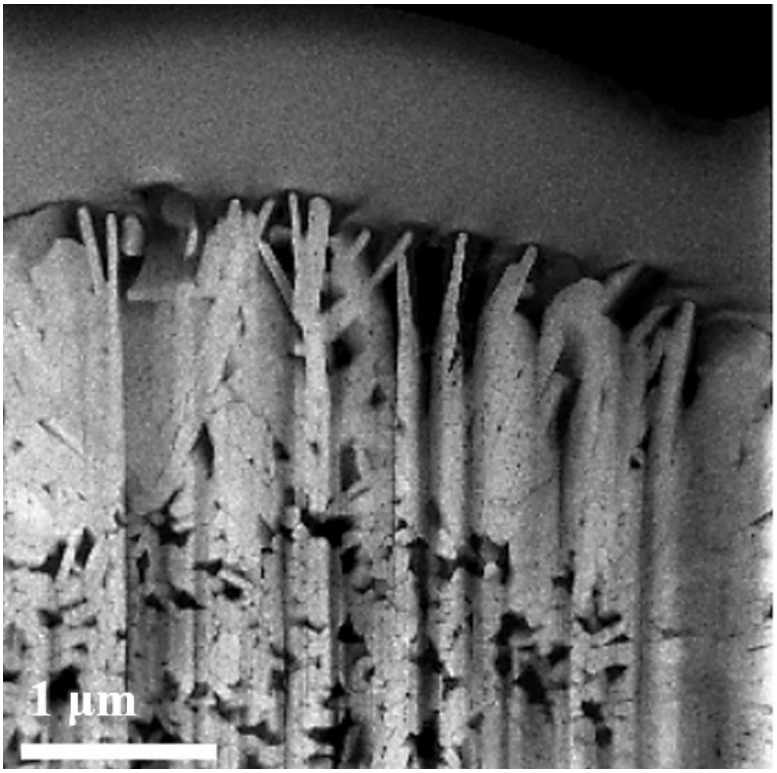

Fig. S16 Low magnification STEM image of P-LNCM FIB specimen calcined at 740 ºC for 12 h.

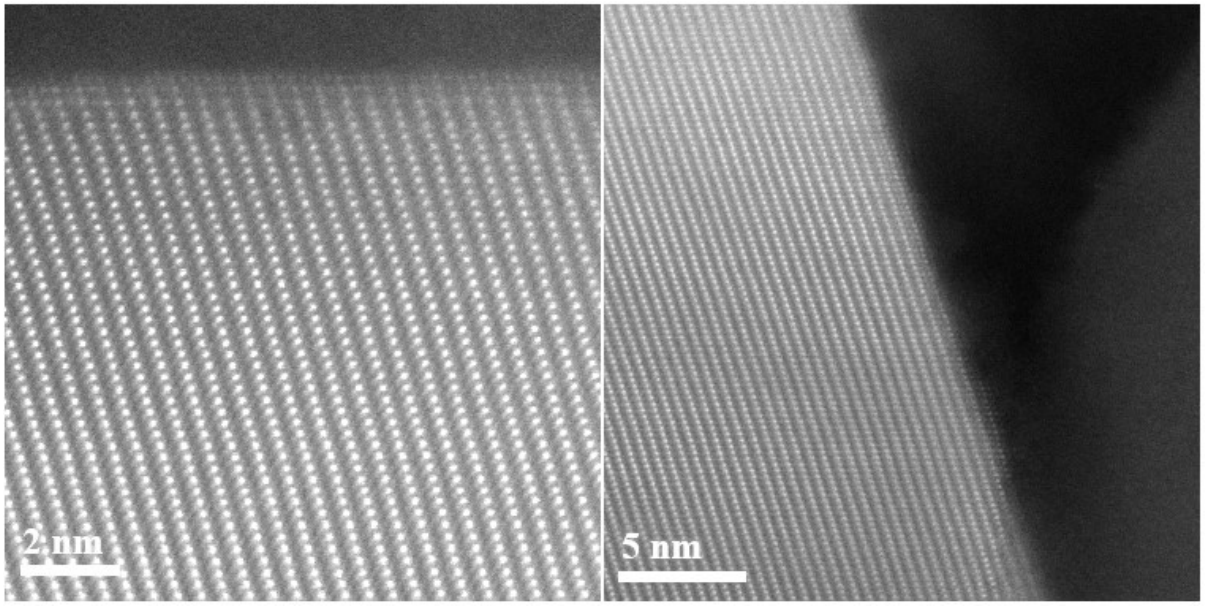

Fig. S17 STEM image of U-LNCM calcined at 740 ºC for 12 h.

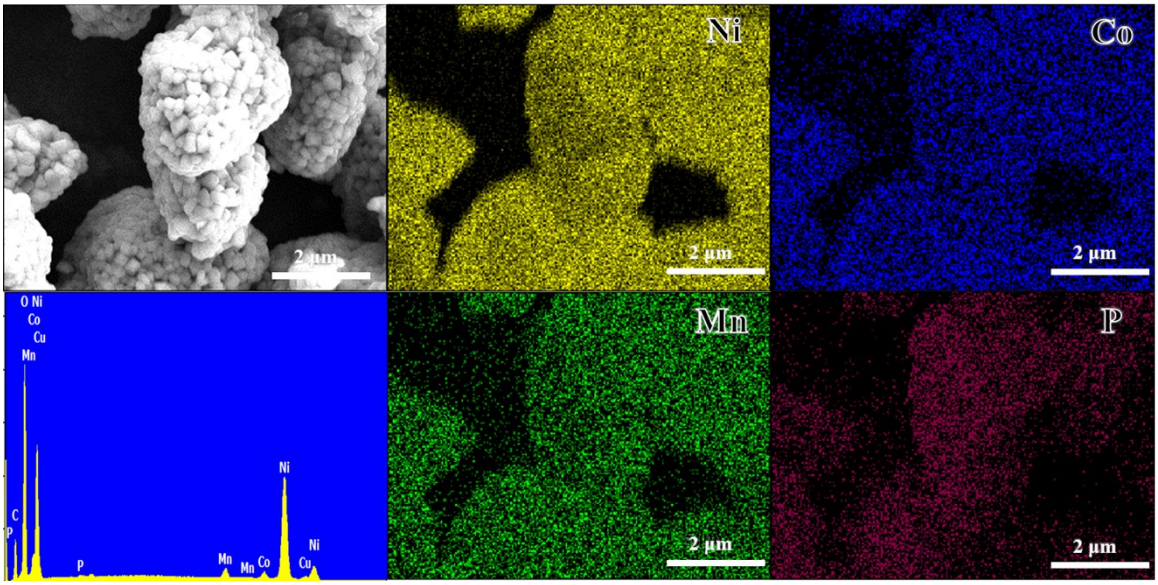

Fig. S18 Energy-dispersive X-ray spectroscopy (EDS) mapping of as-synthesized P-LNCM calcined at 840 ºC for 12h.

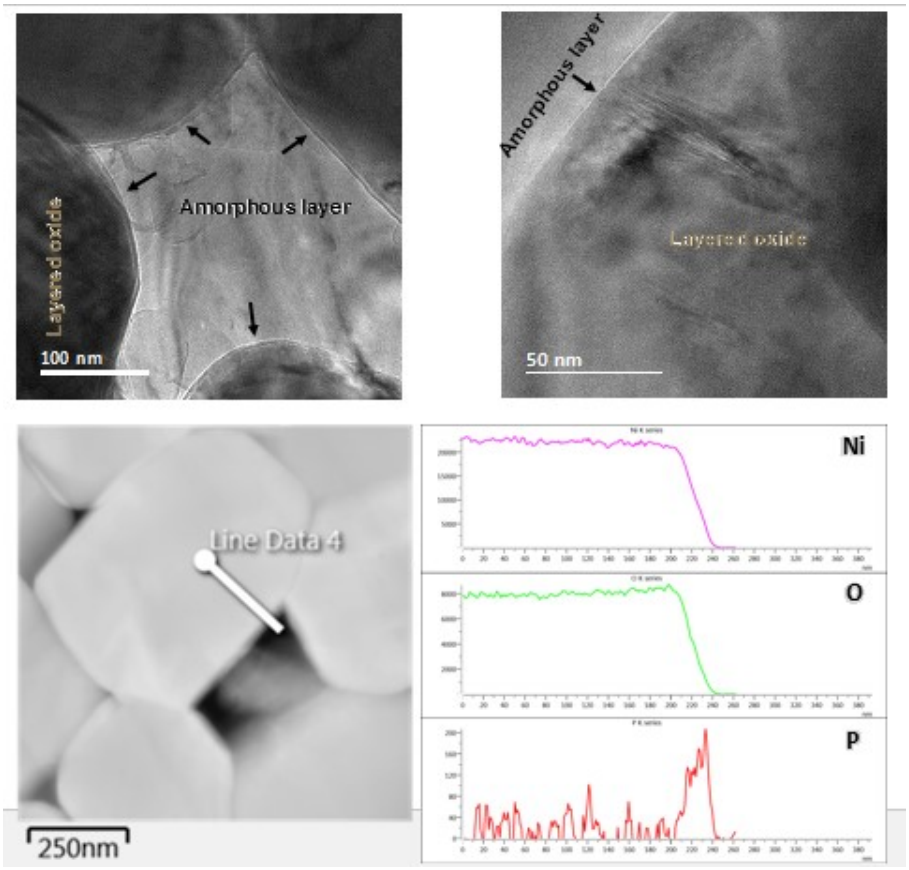

Fig. S19 TEM image and EDS line profiles of P-LNCM calcined at 840 ºC for 12 h.

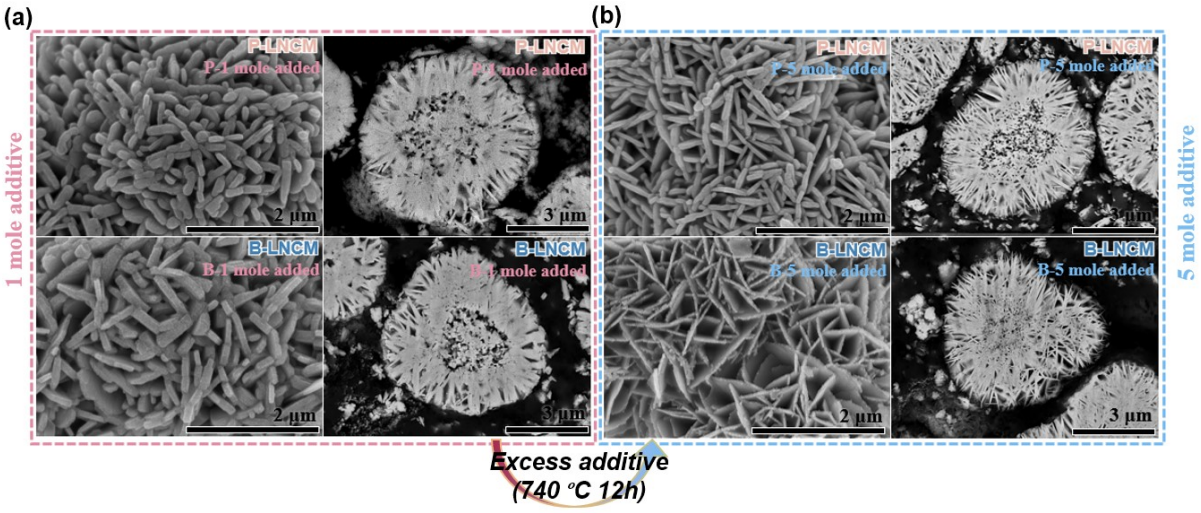

Fig. S20 Microstructure comparison between (a) 1 mole % doped P- and B-LNCMs and (b) 5 mole % doped P- and B-LNCMs. Alll doped LNCMs were calcined at 740 ºC for 12 h.

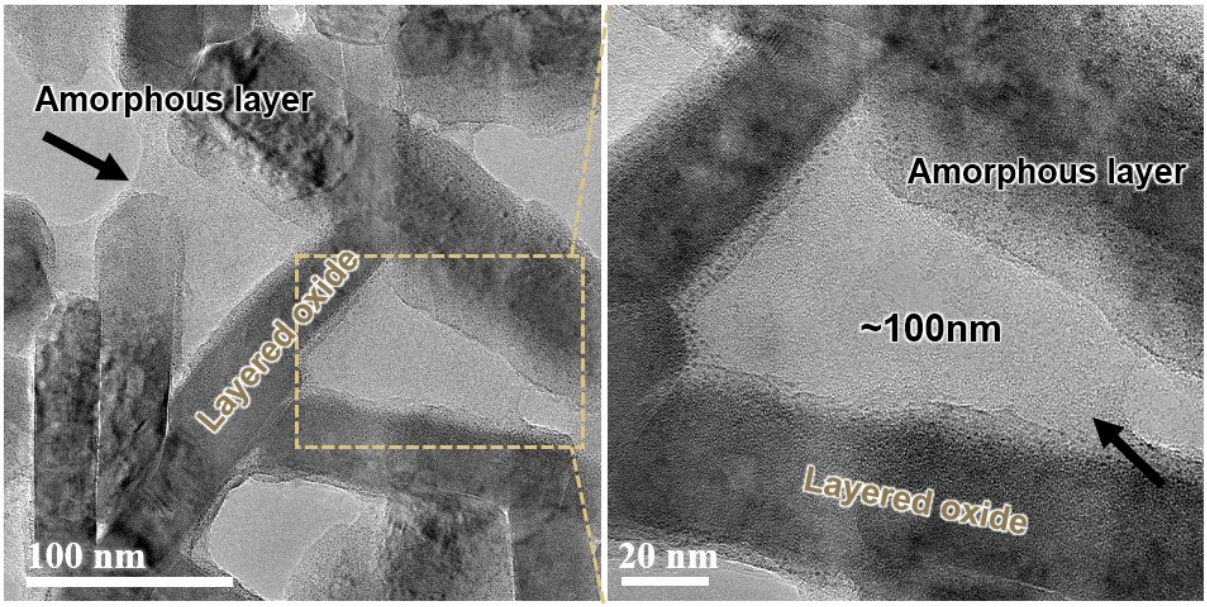

Fig. S21 HRTEM image of FIB cross-sectioned 5 mole % doped P-LNCM, which showed ~100 nm thick amorphous layer covering the primary particles.

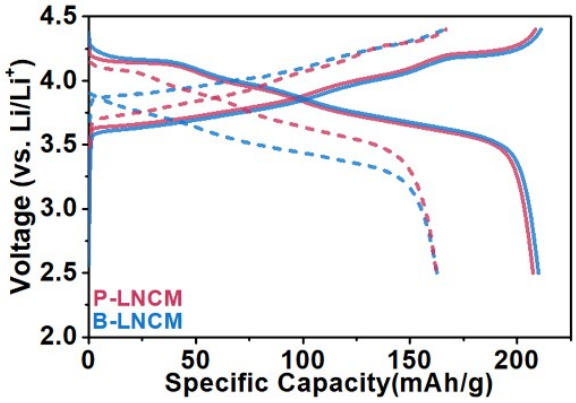

Fig.S22 Initial charge-discharge voltage profiles of P-LNCM and B-LNCM electrodes prepared with the powders calcined at 740 ºC for 12 h. Solid lines-1.0 mole % doping and dot lines-1.5 mole % doping.

Table S1 ICP-AES results of P-LNCM and B-LNCM calcined at 740 ºC for 12 h.

Sample	Li	Ni	Co	Mn	P	B
P-LNCM (counts)	10544.6	9863.3	418.4	388.4	123.9	
mole ratio	0.977	0.913	0.038	0.036	0.0114	
B-LNCM (counts)	10606.3	9871.5	417.3	386.6		115.2
mole ratio	0.982	0.915	0.038	0.035		0.0106

Table S2 ICP-AES results of P-LNCM and B-LNCM calcined at 840 ºC for 12 h.

Sample	Li	Ni	Co	Mn	P	B
P-LNCM (counts)	10269.2	9939.1	415.9	386.5	116.1	
mole ratio	0.946	0.915	0.038	0.035	0.0107	
B-LNCM (counts)	10381.5	10118.9	426.6	395.6		108.5
mole ratio	0.940	0.915	0.038	0.035		0.0098