Supplementary Information

Molybdate flux growth of idiomorphic $Li(Ni_{1/3}Co_{1/3}Mn_{1/3})O_2$ single crystals and characterization of their capabilities as cathode materials for lithium-ion batteries.

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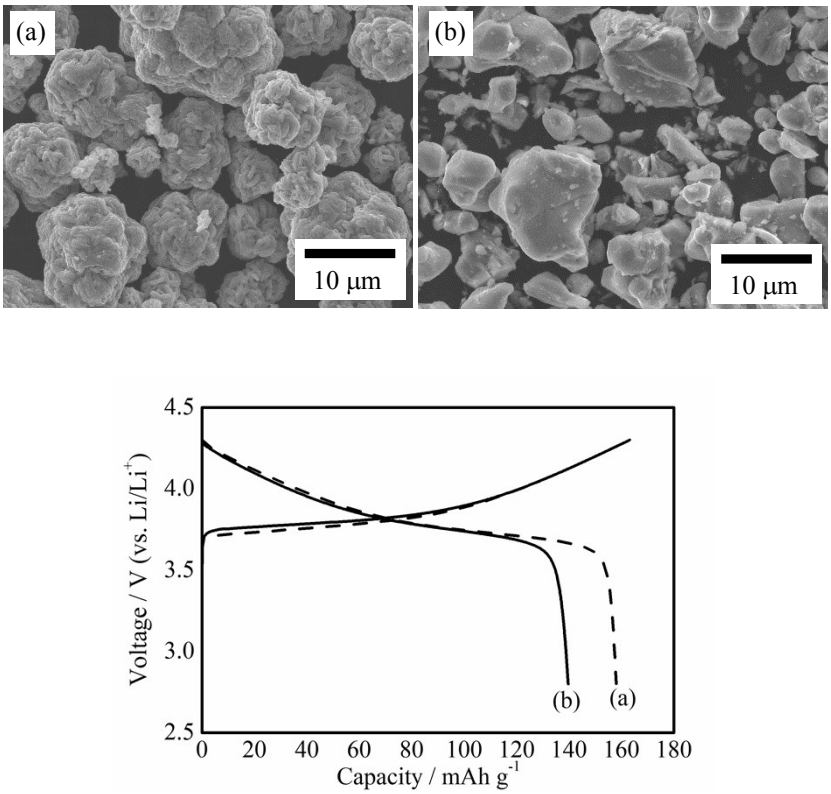


Figure S1. SEM images and corresponding charge-discharge profiles of the NCM crystals purchased from (a) Nippon chemical and (b) Toshima.

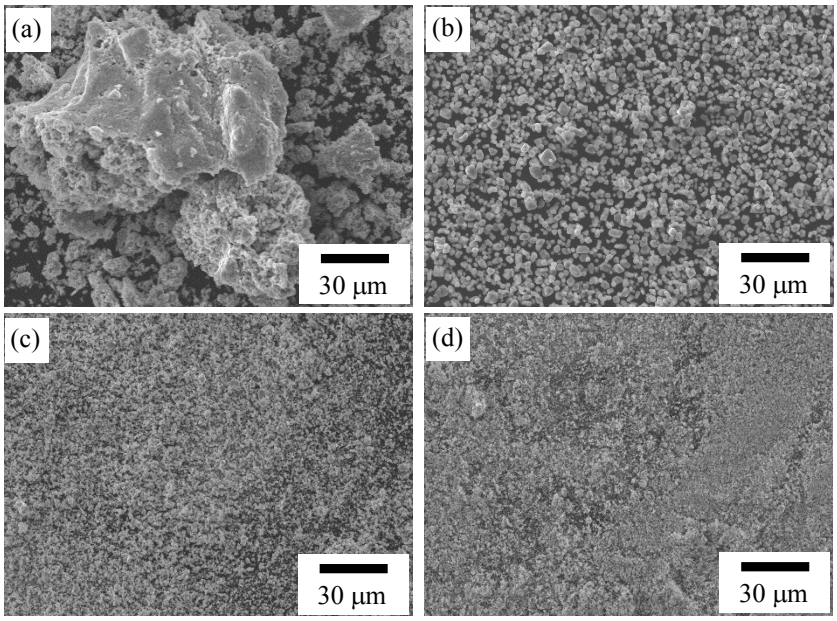


Figure S2. Low-magnification images of the crystas grown at different conditions. Solute concentrations of (a) is 100 mol% and (b-d) are 40 mol%. Holding temperatures of (a, b) are 1000 °C, (c) is 900 °C, and (d) is 800 °C.

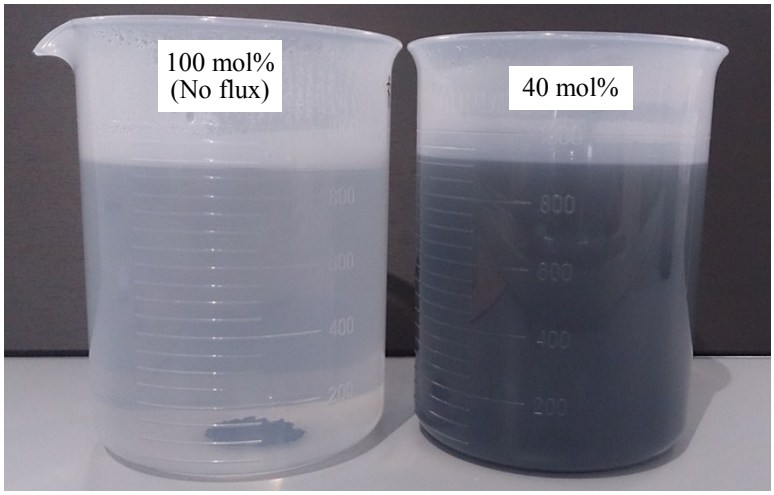


Figure S3. Optical image of the powders dispersed in water soon after stirring.

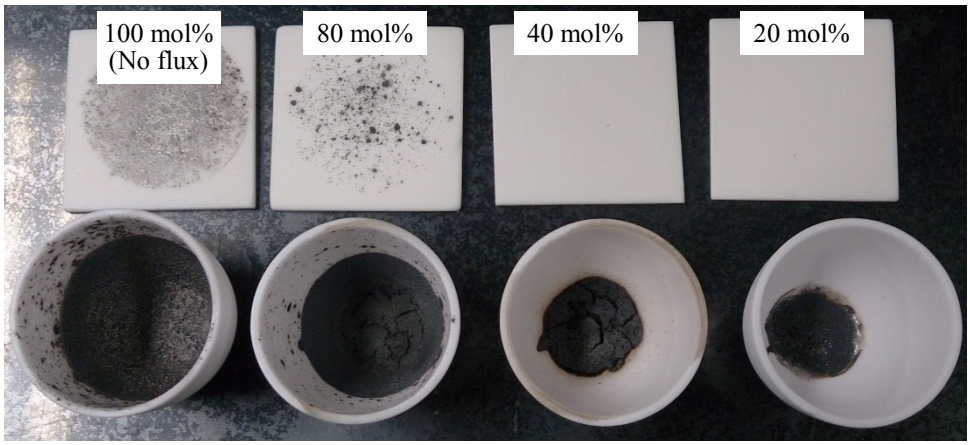


Figure S4. Optical image of the crucible after reaction at different solute concentrations.

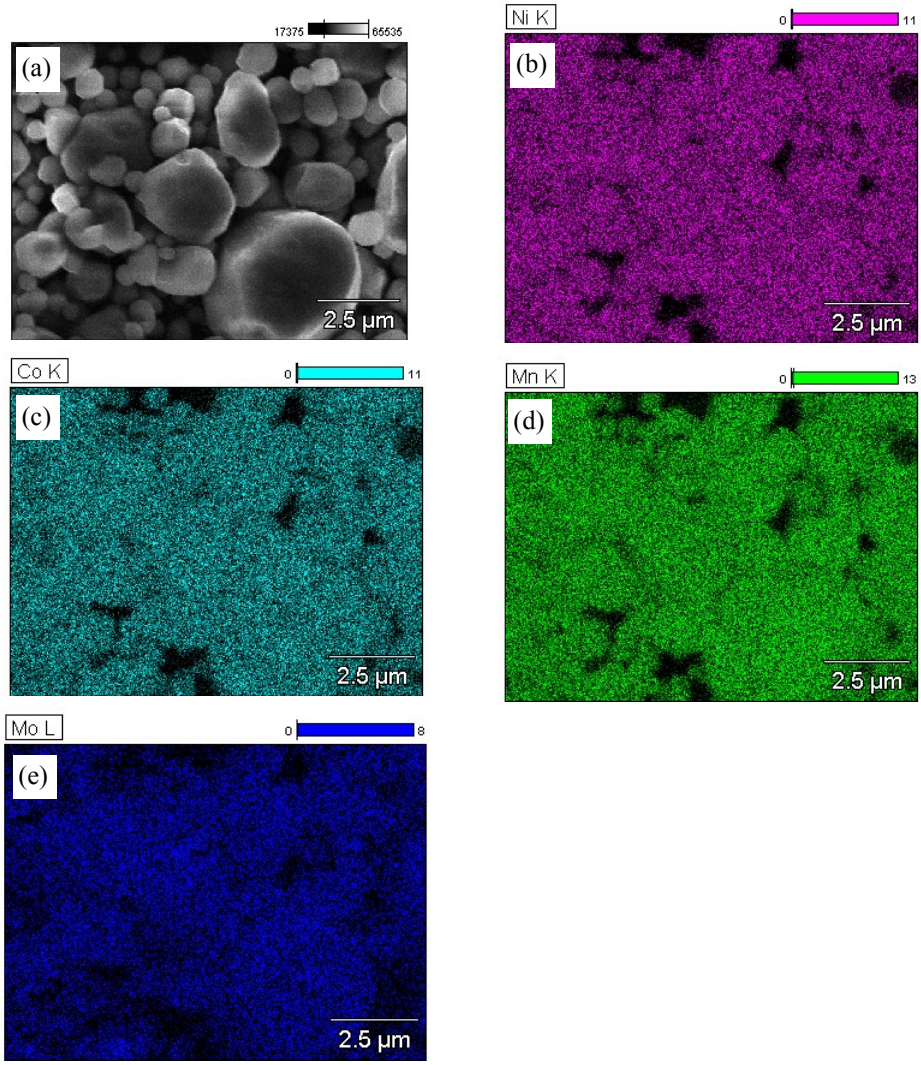


Figure S5. SEM-EDS mapping images the crystals grown at 900 °C. (a) SEM images. The element of (b) is Ni, (c) is Co, (d) is Mn, and (e) is Mo. (ICP Ni:Co:Mn:Mo=0.332:0.328:0.334:0.006; EDS Ni:Co:Mn:Mo=0.344:0.328:0.320:0.008)

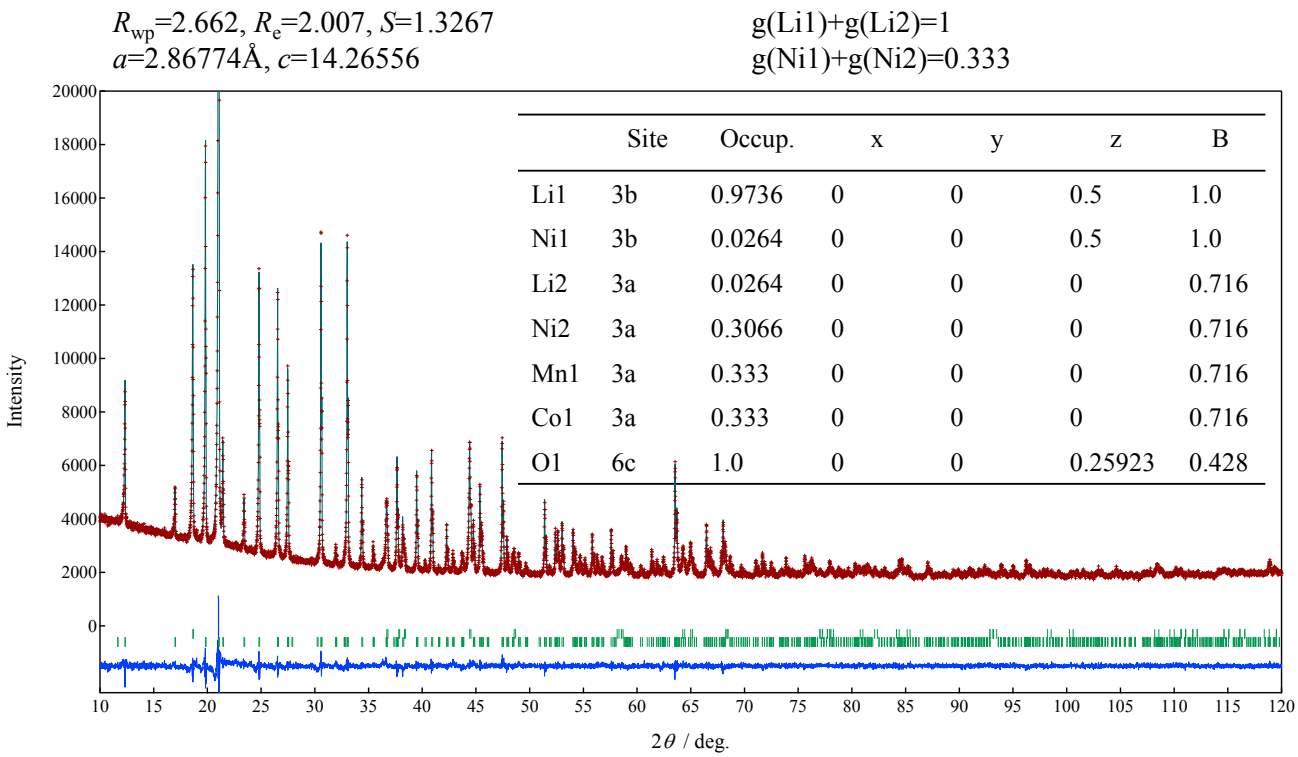


Figure S6. XRD profiles and Rietveld refinement results of the NCM powders of NCM-900 before washing.

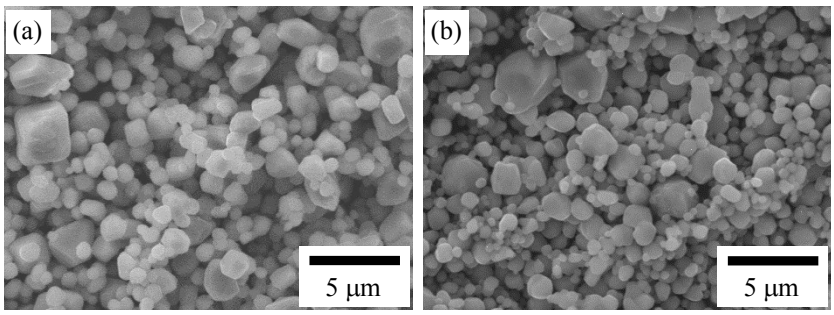
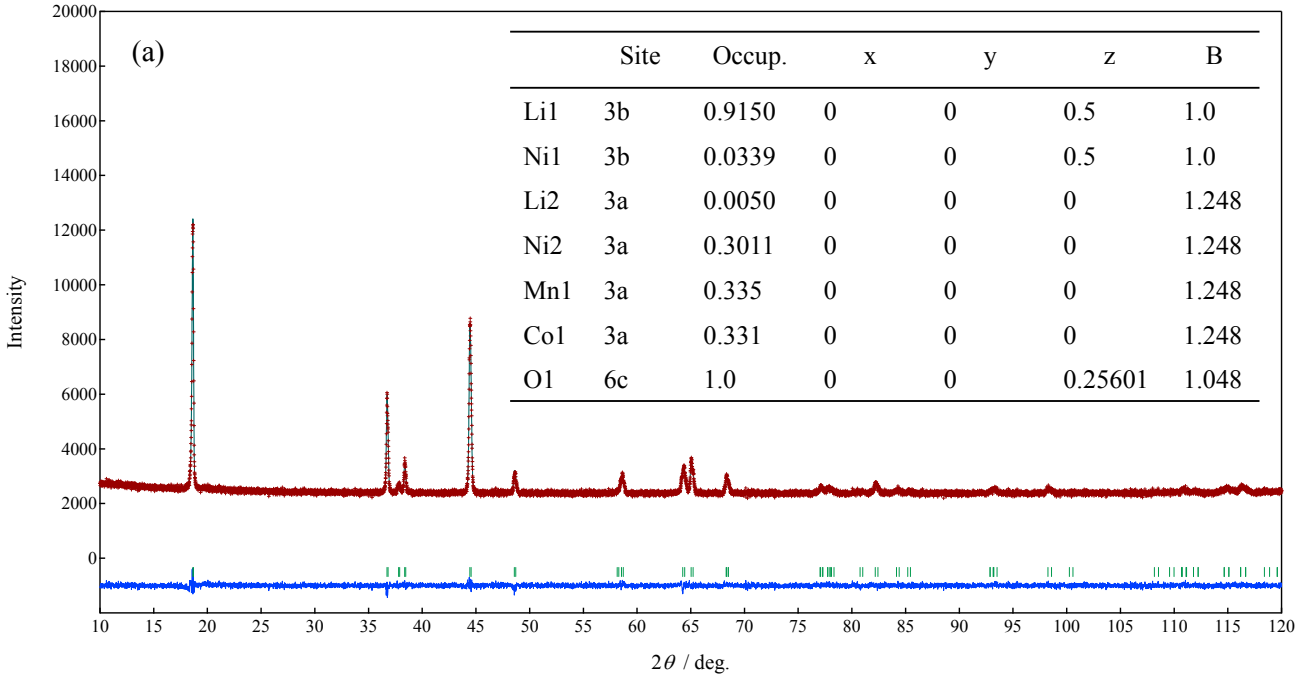


Figure S7. SEM images of (a) NCM-900 and (b) NCM-900HT.

R_{wp}=2.118, R_e=2.006, S=1.0559
a=2.86624Å, c=14.26839

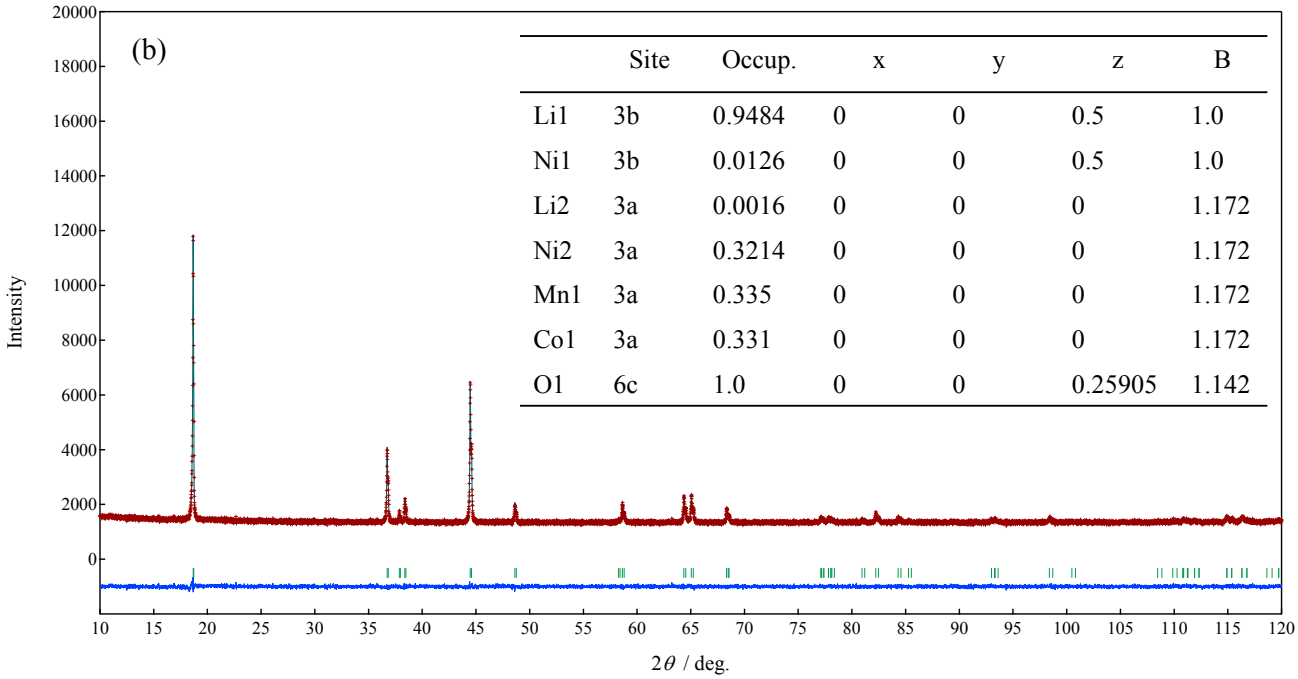
g(Li1)+g(Li2)=0.92
g(Ni1)+g(Ni2)=0.335



	Site	Occup.	x	y	z	B
Li1	3b	0.9150	0	0	0.5	1.0
Ni1	3b	0.0339	0	0	0.5	1.0
Li2	3a	0.0050	0	0	0	1.248
Ni2	3a	0.3011	0	0	0	1.248
Mn1	3a	0.335	0	0	0	1.248
Co1	3a	0.331	0	0	0	1.248
O1	6c	1.0	0	0	0.25601	1.048

R_{wp}=2.589, R_e=2.665, S=0.9717
a=2.86505Å, c=14.24149

g(Li1)+g(Li2)=0.95
g(Ni1)+g(Ni2)=0.334



	Site	Occup.	x	y	z	B
Li1	3b	0.9484	0	0	0.5	1.0
Ni1	3b	0.0126	0	0	0.5	1.0
Li2	3a	0.0016	0	0	0	1.172
Ni2	3a	0.3214	0	0	0	1.172
Mn1	3a	0.335	0	0	0	1.172
Co1	3a	0.331	0	0	0	1.172
O1	6c	1.0	0	0	0.25905	1.142

Figure S8. XRD profiles and Rietveld refinement results of the NCM powders (a) NCM-900 and (b) NCM-900HT.

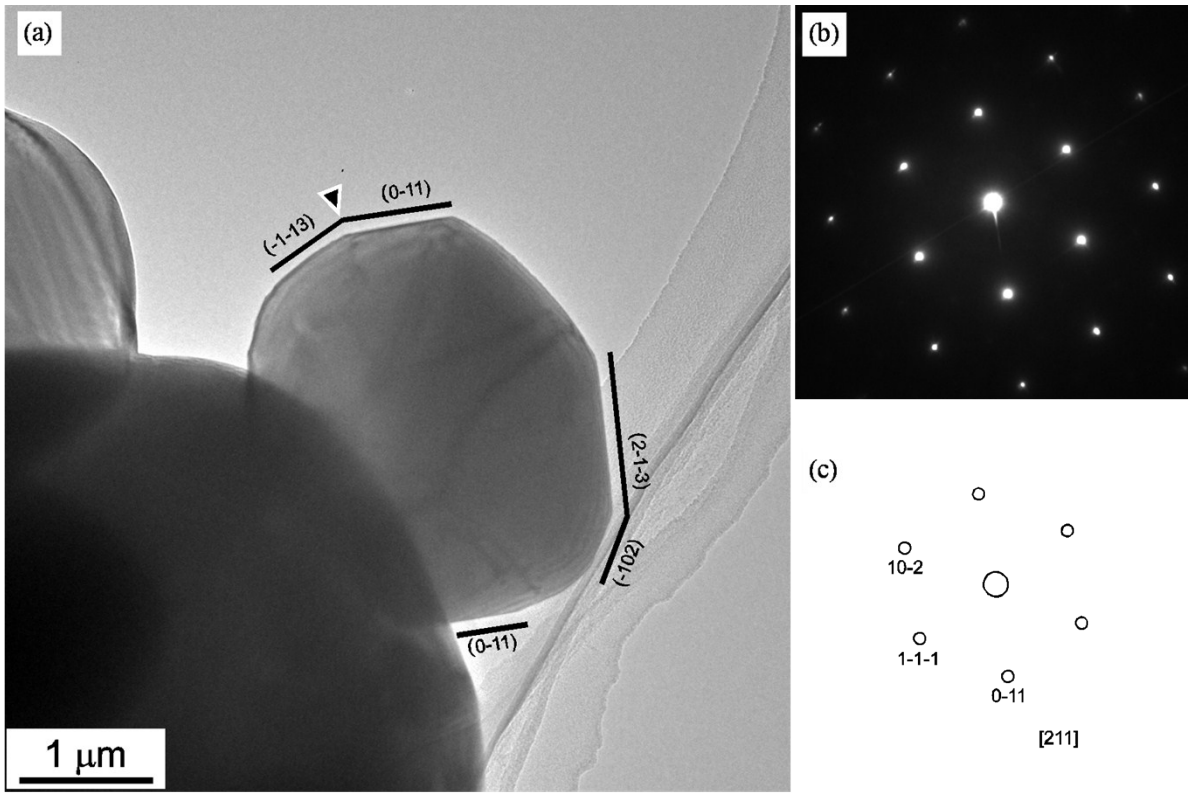


Figure S9. (a) TEM image and (b) corresponding SAED image of NCM-900HT. (c) Illustration of SAED spot.

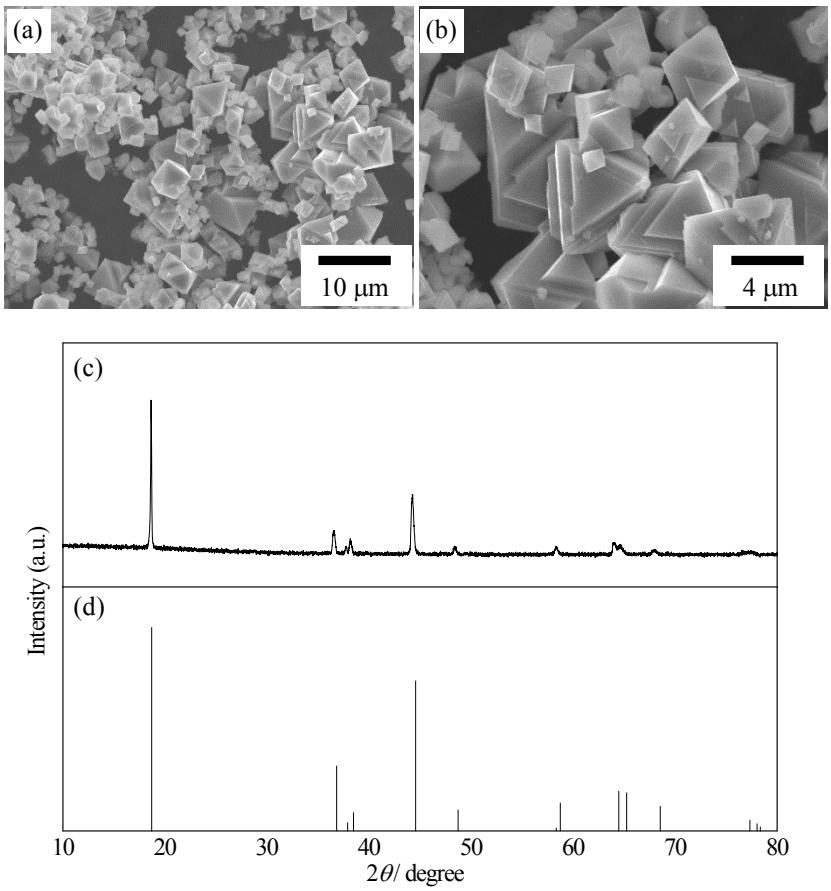


Figure S10. (a) Low- and (b) high-magnification SEM images and (c) XRD profile of the crystals grown by use of Na_2MoO_4 flux. (d) $Li(Ni_{0.333}Co_{0.333}Mn_{0.333})O_2$ (ICDD PDF 56-0147).

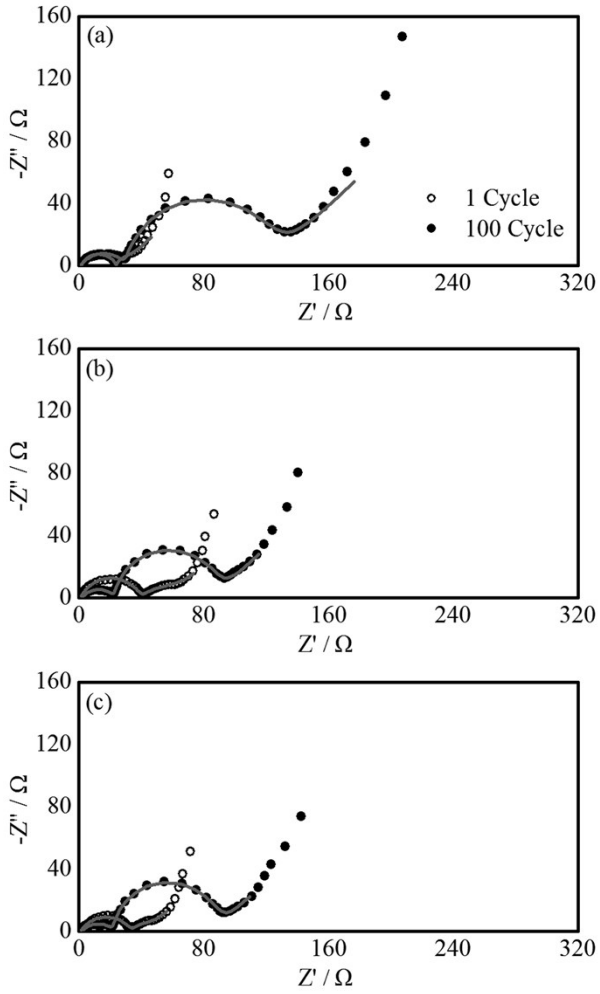


Figure S11. Impedance spectra of the electrode using (a) NCM-800HT, (b) NCM-900HT, and (c) NCM-1000HT at 1st and 100th cycle.

	800HT C_{sf}	800HT C_{ct}	900HT C_{sf}	900HT C_{ct}	1000HT C_{sf}	1000HT C_{ct}
1 cycle	23 Ω	8 Ω	39 Ω	31 Ω	33 Ω	16 Ω
100 cycle	30 Ω	94 Ω	21 Ω	65 Ω	21 Ω	66 Ω

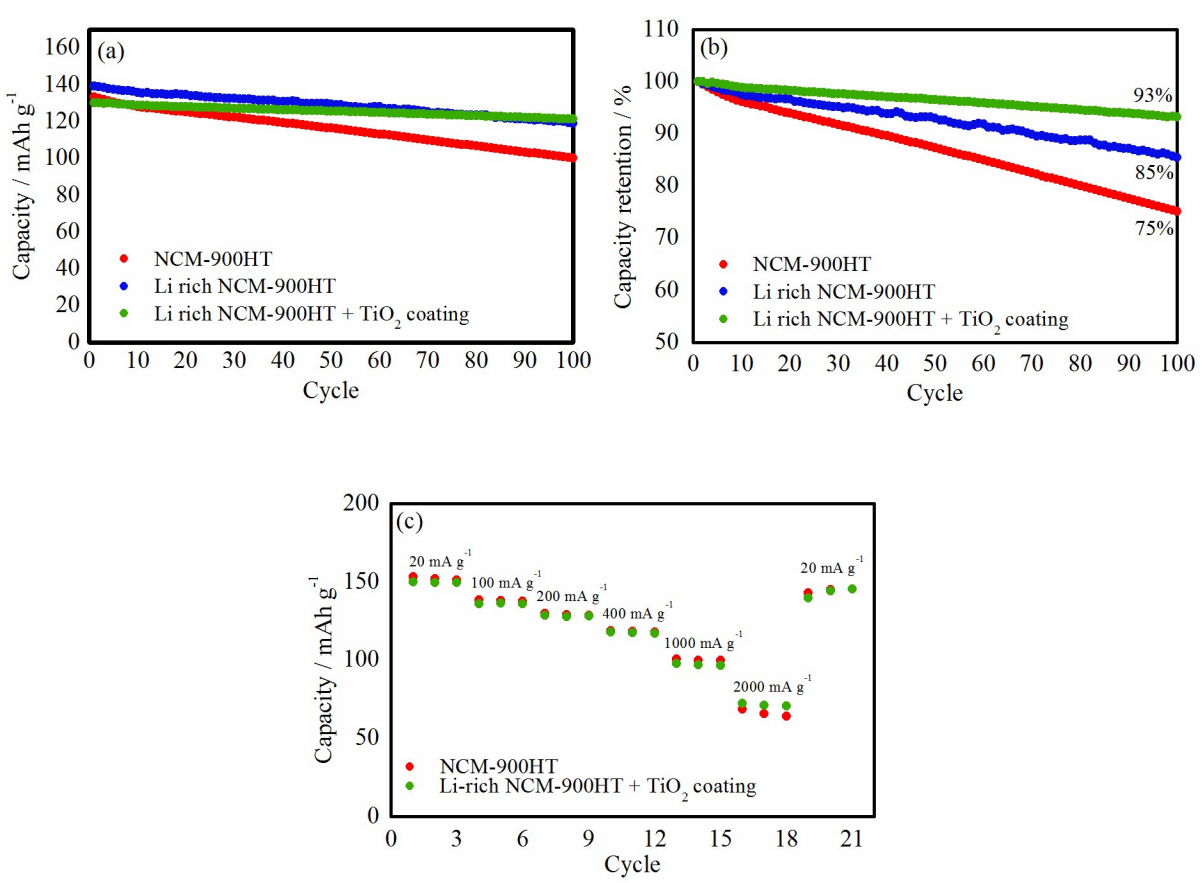


Figure S12. (a) The changes in the discharge capacity and (b) corresponding discharge capacity retention as a function of cycle numbers: (1) NCM-900HT; (2) Li-rich NCM-900HT; (3) Li-rich NCM-900HT + TiO_2 coating. (c) Discharge capacities of NCM-900HT and Li-rich NCM-900HT + TiO_2 coating at different rates. The TiO_2 coating was demonstrated based on the literature (H. Liu et al., *Solid State Ionics*, **2004**, *166*, 317-325). Briefly, Li-rich NCM-900HT powders were dispersed in an ethanolic solution, and then titanium (IV) tetraisopropoxide was added under vigorous stirring. After evaporation of all solution at 70 °C, the obtained powders were heated at 400 °C for 5 h. The molar ratio of Ti/(Ni + Co + Mn) was set to 0.03.