Supporting Information

Enhancing the Structural Durability of Ni-Rich Layered Materials by Post-process: Washing and Heat-treatment

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Table S1. The amount of residual lithium compounds for untreated NCA and treated NCA

Sample	Powder [g]	DI Water [g]	Solvent [g]	(1/2)Li_2CO_3 [ppm]	LiOH [ppm]	Free-Li [ppm]
Untreated	10	100	20	**4,132**	**5,159**	**2,271**
Treated	10	100	20	**1,419**	**1,851**	**803**

Table S2a. Rietveld refinement on the XRD pattern of the pristine $Li_{1-x}Ni_{0.88}Co_{0.11}Al_{0.01}O_2$ (untreated NCA)

Space group : $R\bar{3}m$						
a_{hex} : 2.87148(1) Å	c_{hex} : 14.1892(1) Å	Volume : 101.32 Å^3				$S(R_{wp}/R_{exp})$ = 2.09
Atom	**Wyckoff position**	**x**	**y**	**z**	B_{iso}	**Occupancy**
Li	3b	0	0	0.5	1.00	0.915(5)
Ni_{Li}	3b	0	0	0.5	1.00	0.084(5)
Ni	3a	0	0	0	1.3(1)	0.795(5)
Co	3a	0	0	0	1.3(1)	0.11
Al	3a	0	0	0	1.3(1)	0.01
O	6c	0	0	0.2560(6)	1.00	2.00

Table S2b. Rietveld refinement on the XRD pattern of the pristine $Li_{1-x}Ni_{0.88}Co_{0.11}Al_{0.01}O_2$ (treated NCA)

Space group : $R\bar{3}m$						
a_{hex} : 2.87079(2) Å	c_{hex} : 14.1837(2) Å	Volume : 101.23 Å^3				$S(R_{wp}/R_{exp})$ = 2.19
Atom	**Wyckoff position**	**x**	**y**	**z**	B_{iso}	**Occupancy**
Li	3b	0	0	0.5	1.00	0.944(5)
Ni_{Li}	3b	0	0	0.5	1.00	0.055(5)
Ni	3a	0	0	0	1.8(1)	0.824(5)
Co	3a	0	0	0	1.8(1)	0.11
Mn	3a	0	0	0	1.8(1)	0.01
O	6c	0	0	0.2574(6)	1.00	2.00

Table S3a. Rietveld refinement on the XRD pattern of the 2nd cycled $Li_{1-x}Ni_{0.88}Co_{0.11}Al_{0.01}O_2$ (untreated NCA)

Space group : R-3m						
a_{hex} : 2.86535(3) Å	c_{hex} : 14.2248(2) Å	Volume : 101.143 Å^3				$S(R_{wp}/R_{exp})$ = 1.63
Atom	**Wyckoff position**	**x**	**y**	**z**	B_{iso}	**Occupancy**
Li	3b	0	0	0.5	1.00	0.877(6)
Ni_{Li}	3b	0	0	0.5	1.00	0.036(6)
Ni	3a	0	0	0	0.37(8)	0.843(6)
Co	3a	0	0	0	0.37(8)	0.11
Al	3a	0	0	0	0.37(8)	0.01
O	6c	0	0	0.2590(6)	1.00	2.00

Table S3b. Rietveld refinement on the XRD pattern of the 2nd cycled $Li_{1-x}Ni_{0.88}Co_{0.11}Al_{0.01}O_2$ (treated NCA)

Space group : R-3m						
a_{hex} : 2.86465(2) Å	c_{hex} : 14.2190(2) Å	Volume : 101.052 Å^3				$S(R_{wp}/R_{exp})$ = 2.57
Atom	**Wyckoff position**	**x**	**y**	**z**	B_{iso}	**Occupancy**
Li	3b	0	0	0.5	1.00	0.908(5)
Ni_{Li}	3b	0	0	0.5	1.00	0.030(5)
Ni	3a	0	0	0	1.16(8)	0.849(5)
Co	3a	0	0	0	1.16(8)	0.11
Mn	3a	0	0	0	1.16(8)	0.01
O	6c	0	0	0.2593(5)	1.00	2.00

Table S4a. Rietveld refinement on the XRD pattern of the 300th cycled $Li_{1-x}Ni_{0.88}Co_{0.11}Al_{0.01}O_2$ (untreated NCA)

Space group : R-3m						
a_{hex} : 2.85826(7) Å	**c_{hex} : 14.2659(1) Å**	**Volume : 100.934 Å³**				**S(R_{wp}/R_{exp}) = 2.37**
Atom	**Wyckoff position**	**x**	**y**	**z**	**B_{iso}**	**Occupancy**
Li	**3b**	**0**	**0**	**0.5**	**1.00**	**0.585(4)**
Ni_{Li}	**3b**	**0**	**0**	**0.5**	**1.00**	**0.059(4)**
Ni	**3a**	**0**	**0**	**0**	**0.45(6)**	**0.820(4)**
Co	**3a**	**0**	**0**	**0**	**0.45(6)**	**0.11**
Al	**3a**	**0**	**0**	**0**	**0.45(6)**	**0.01**
O	**6c**	**0**	**0**	**0.2595(4)**	**1.00**	**2.00**

Table S4b. Rietveld refinement on the XRD pattern of the 300th cycled $Li_{1-x}Ni_{0.88}Co_{0.11}Al_{0.01}O_2$ (treated NCA)

Space group : R-3m						
a_{hex} : 2.86152(1) Å	**c_{hex} : 14.2362(1) Å**	**Volume : 100.954 Å³**				**S(R_{wp}/R_{exp}) = 2.49**
Atom	**Wyckoff position**	**x**	**y**	**z**	**B_{iso}**	**Occupancy**
Li	**3b**	**0**	**0**	**0.5**	**1.00**	**0.770(5)**
Ni_{Li}	**3b**	**0**	**0**	**0.5**	**1.00**	**0.045(5)**
Ni	**3a**	**0**	**0**	**0**	**0.72(7)**	**0.834(5)**
Co	**3a**	**0**	**0**	**0**	**0.72(7)**	**0.11**
Mn	**3a**	**0**	**0**	**0**	**0.72(7)**	**0.01**
O	**6c**	**0**	**0**	**0.2595(5)**	**1.00**	**2.00**

Table S5a. Rate capability of untreated $Li_{1-x}Ni_{0.88}Co_{0.11}Al_{0.01}O_2$ (untreated NCA)

Discharge capacities	C-rate			
Cell No.	**0.2**	**0.5**	**1.0**	**2.0**
1	**100 %**	**96.6 %**	**93.8 %**	**87.4 %**
2	**100 %**	**96.9 %**	**94.4 %**	**87.3 %**
3	**100 %**	**96.9 %**	**94.3 %**	**86.9 %**
Average	**100 %**	**96.8 %**	**94.2 %**	**87.2 %**

Table S5b. Rate capability of treated $Li_{1-x}Ni_{0.88}Co_{0.11}Al_{0.01}O_2$ (treated NCA)

Discharge capacities	C-rate			
Cell No.	**0.2**	**0.5**	**1.0**	**2.0**
1	**100 %**	**97.2 %**	**95.1 %**	**88.0 %**
2	**100 %**	**97.3 %**	**95.3 %**	**88.7 %**
3	**100 %**	**97.2 %**	**95.1 %**	**89.3 %**
Average	**100 %**	**97.2 %**	**95.1 %**	**88.6 %**

Table S6. Curve fitting results for the Ni K-edge EXAFS of untreated and treated samples at 2nd charge and 2nd discharge state.

$LiNi_{0.88}Co_{0.11}Al_{0.01}O_2$		Shell	CN	R(Å)	σ^2 ($x10^{-4}$ Å^2)	R-factor
Untreated	**2nd Charge**	Ni-O	6	1.883(2)	31(3)	0.00361
	2nd Discharge	Ni-O	6	1.916(1)	83(1)	0.00210
Treated	**2nd Charge**	Ni-O	6	1.879(3)	27(3)	0.00293
	2nd Discharge	Ni-O	6	1.931(3)	78(1)	0.00308

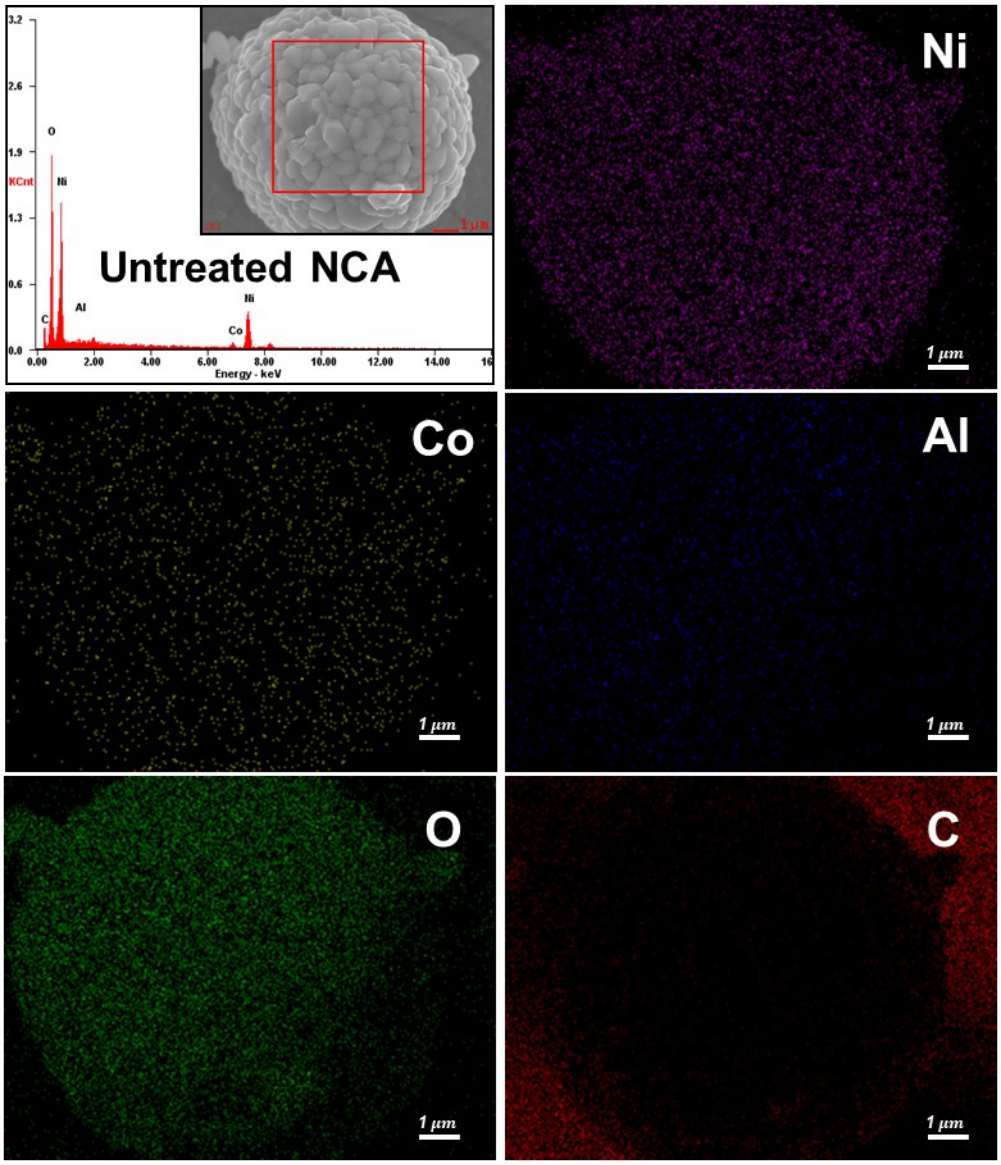

Figure S1. EDX mapping of untreated $Li_{1-x}Ni_{0.88}Co_{0.11}Al_{0.01}O_2$ cathode materials

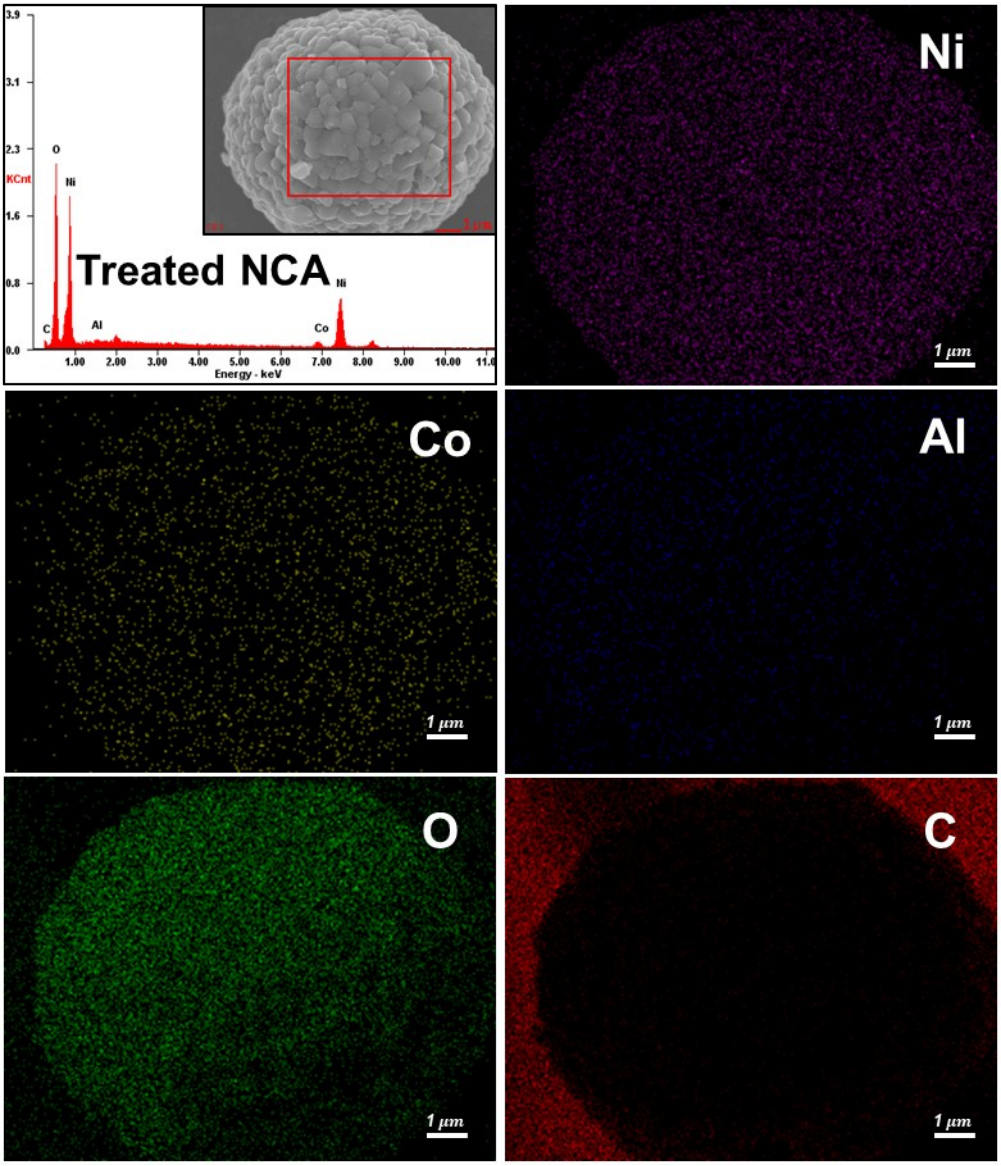

Figure S2. EDX mapping of treated $Li_{1-x}Ni_{0.88}Co_{0.11}Al_{0.01}O_2$ cathode materials

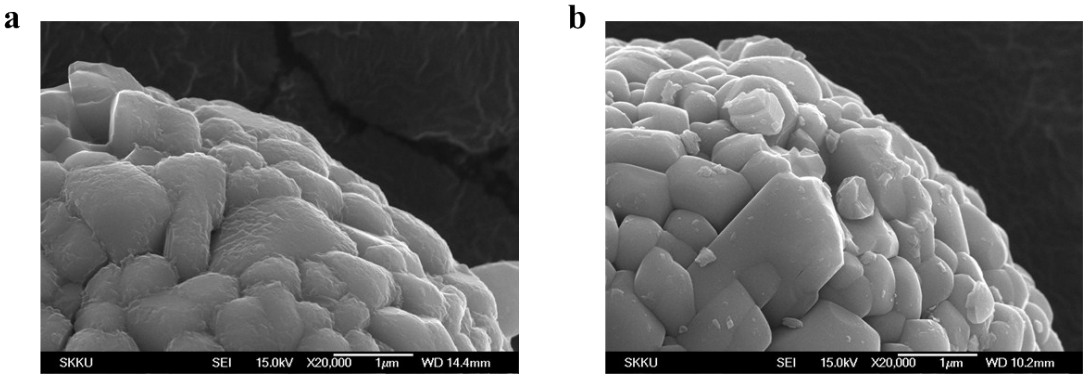

Figure S3. SEM images of as-prepared (a) untreated NCA and (b) treated NCA materials.

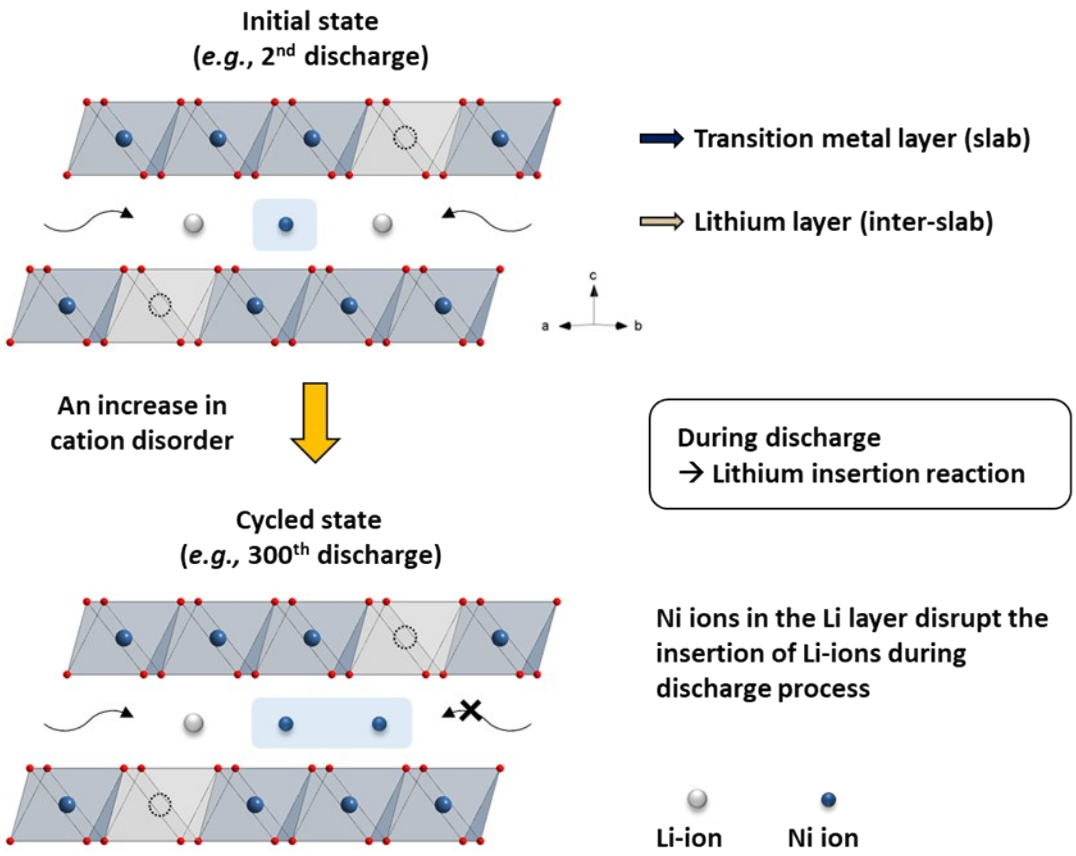

Figure S4. Schematic illustration of the lithium intercalation during discharge process before/after cycle.

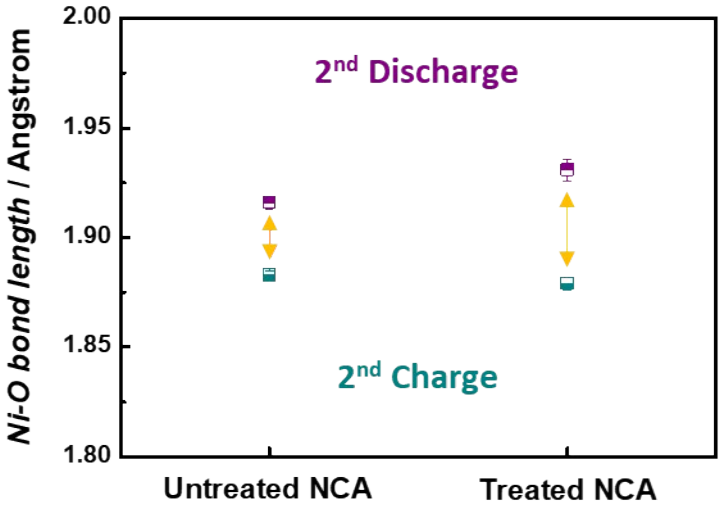

Figure S5. Structural parameters of the first coordination shell Ni-O for untreated and treated, respectively. Cyan color: Ni-O bond length at 2nd charge. Purple color: Ni- bond length at 2nd discharge.

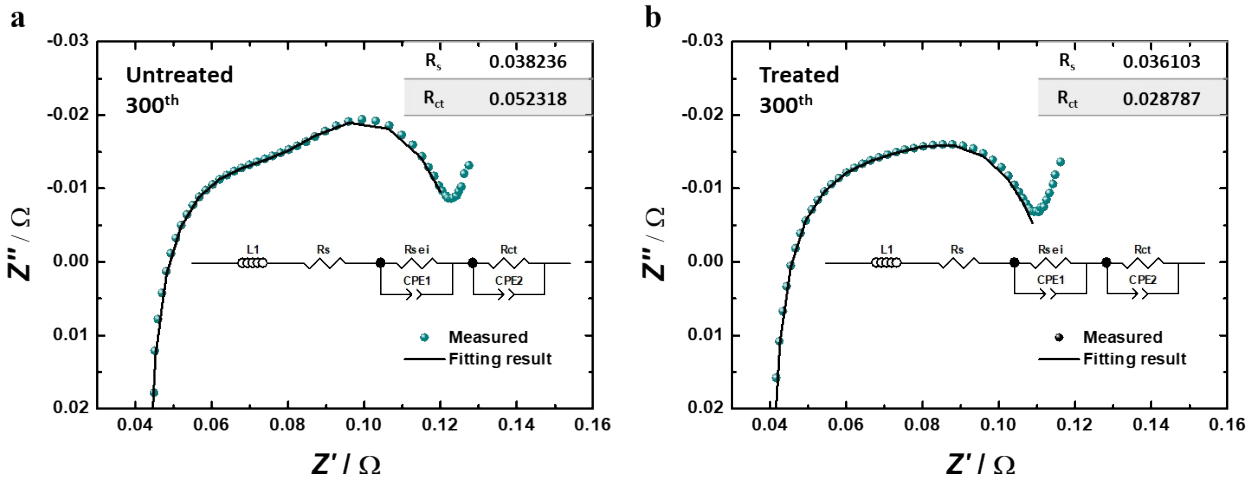

Figure S6. Electrochemical impedance spectra (EIS) of untreated and treated sample at 300th cycle and the equivalent circuits are shown as an inset.

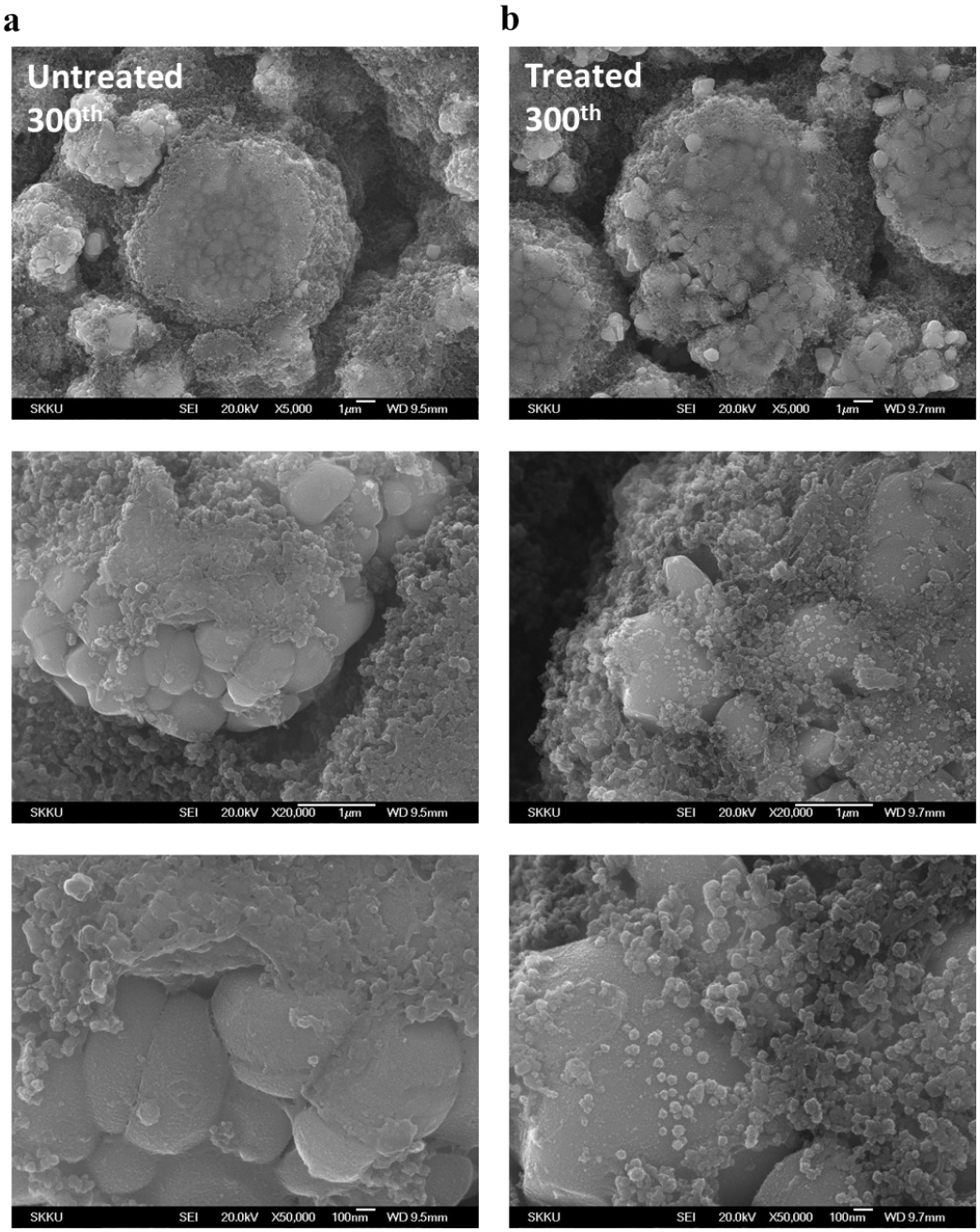

Figure S7. SEM images for (a) untreated and (b) treated sample at 300th cycle.

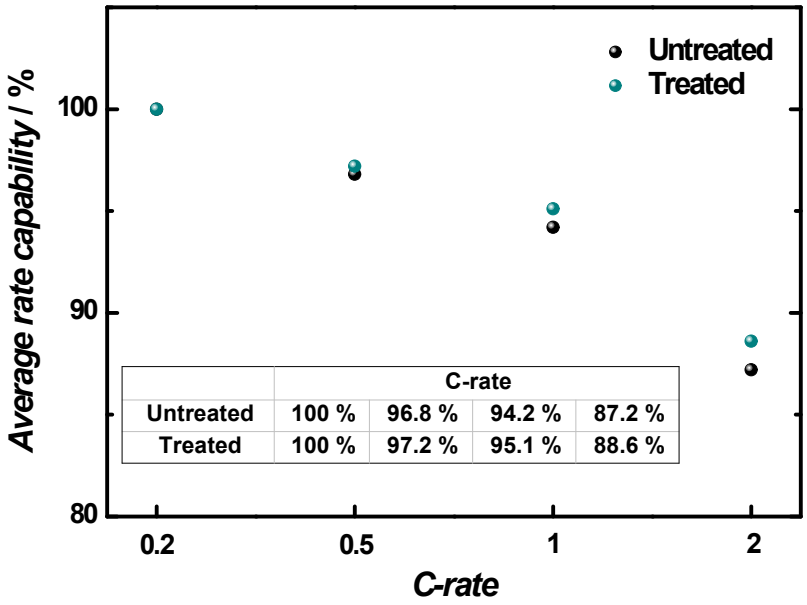

Figure S8. A comparison of the average rate capability between untreated and treated $Li_{1-x}Ni_{0.88}Co_{0.11}Al_{0.01}O_2$ cathode materials

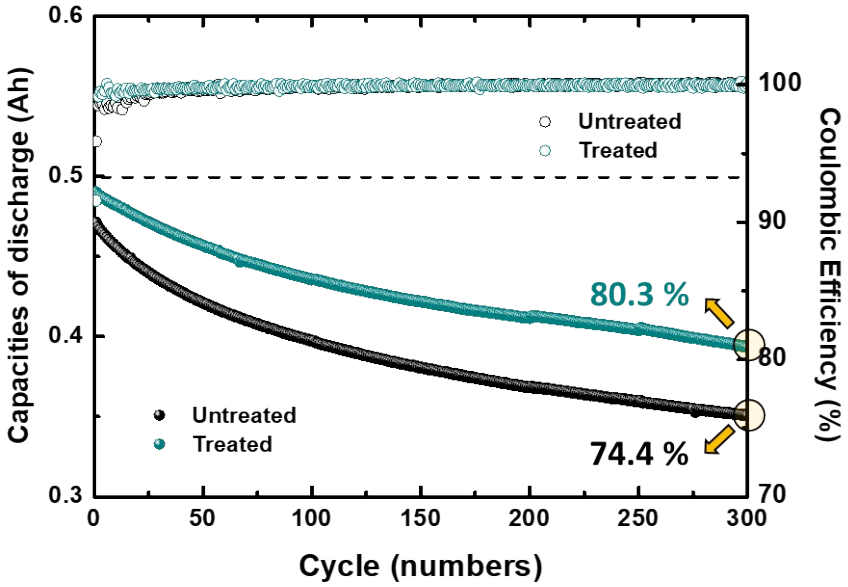

Figure S9. Cycling performance of untreated and treated $Li_{1-x}Ni_{0.88}Co_{0.11}Al_{0.01}O_2$