Supplementary Information

Revealing Degradation Mechanism of Ni-Rich Cathode Materials after Ambient Storage and Related Regeneration Method

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Table S1. Lattice parameters and Ni/Li antisites of NCA, NCA-S, NCA-SO and NCA-SLO.

Samples	*a* (Å)	*c* (Å)	Ni/Li antisities (%)	R_p (%)	R_{wp} (%)
NCA	2.865047	14.181972	1.50	1.32%	2.11%
NCA-S	2.864171	14.195577	0.75	1.33%	2.10%
NCA-SO	2.867153	14.185193	2.27	1.24%	1.98%
NCA-SLO	2.867255	14.183615	1.59	1.38%	2.18%

Table S2. Crystal structure distances of NCA, NCA-S, NCA-SO and NCA-SLO.

Vector	NCA (Å)	NCA-S (Å)	NCA-SO (Å)	NCA-SLO (Å)
Li1-Li1	2.86505	2.86417	2.86715	2.86725
Li1-Ni1	2.86505	2.86417	2.86715	2.86725
Li1-Li2	2.88497	2.88654	2.88611	2.88593
Li1-Ni2	2.88497	2.88654	2.88611	2.88593
Li1-Co1	2.88497	2.88654	2.88611	2.88593
Li1-Al1	2.88497	2.88654	2.88611	2.88593
Li1-O1	2.08094	2.09662	2.08611	2.07830
Ni1-Ni1	2.86505	2.86417	2.86715	2.86725
Ni1-Li2	2.88497	2.88654	2.88611	2.88593
Ni1-Ni2	2.88497	2.88654	2.88611	2.88593
Ni1-Co1	2.88497	2.88654	2.88611	2.88593
Ni1-Al1	2.88497	2.88654	2.88611	2.88593
Ni1-O1	2.08094	2.09662	2.08611	2.07830
Li2-Li2	2.86505	2.86417	2.86715	2.86725
Li2-Ni2	2.86505	2.86417	2.86715	2.86725
Li2-Co1	2.86505	2.86417	2.86715	2.86725
Li2-Al1	2.86505	2.86417	2.86715	2.86725
Li2-O1	1.98709	1.97343	1.98457	1.99170
Ni2-Ni2	2.86505	2.86417	2.86715	2.86725
Ni2-Co1	2.86505	2.86417	2.86715	2.86725
Ni2-Al1	2.86505	2.86417	2.86715	2.86725
Ni2-O1	1.98709	1.97343	1.98457	1.99170
Co1-Co1	2.86505	2.86417	2.86715	2.86725
Co1-Al1	2.86505	2.86417	2.86715	2.86725
Co1-O1	1.98709	1.97343	1.98457	1.9917
Al1-Al1	2.86505	2.86417	2.86715	2.86725
Al1-O1	1.98709	1.97343	1.98457	1.99170

Table S3. ICP-MS results for the chemical composition of the samples.

Samples	Li	Ni	Co	Al
NCA	1.0095	0.8025	0.1477	0.0498

NCA-S	1.0137	0.7808	0.1424	0.0768
NCA-SO	0.9725	0.7903	0.1474	0.0623
NCA-SLO	1.0011	0.7899	0.1469	0.0632

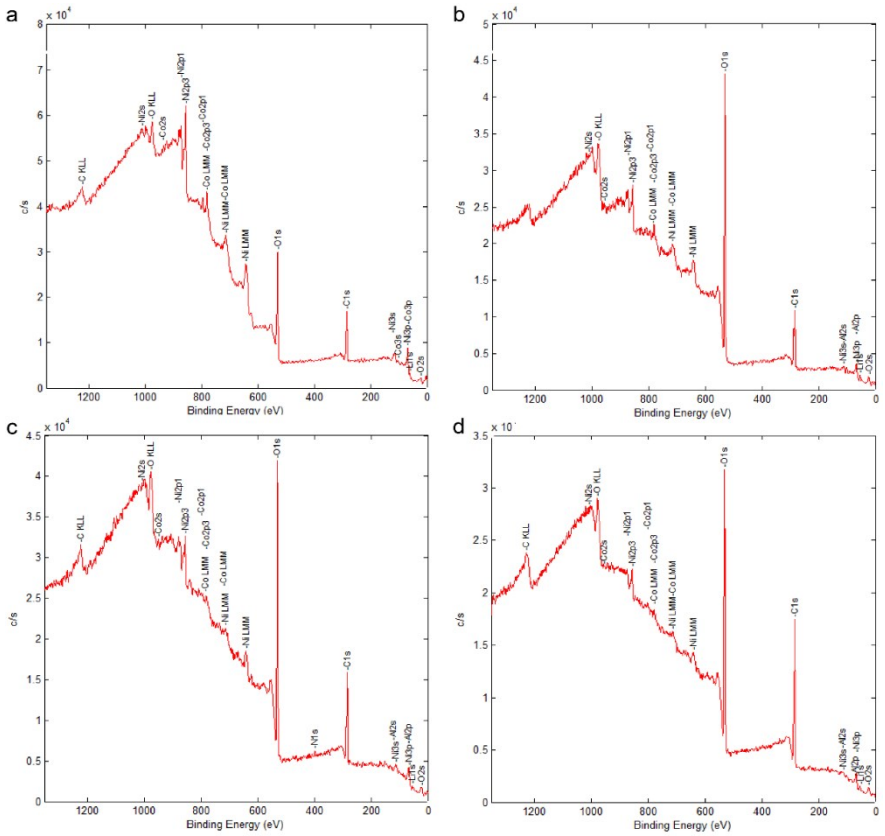

Figure S1. The wide XPS spectra of the (a) NCA, (b) NCA-S, (c) NCA-SO and (d) NCA-SLO.

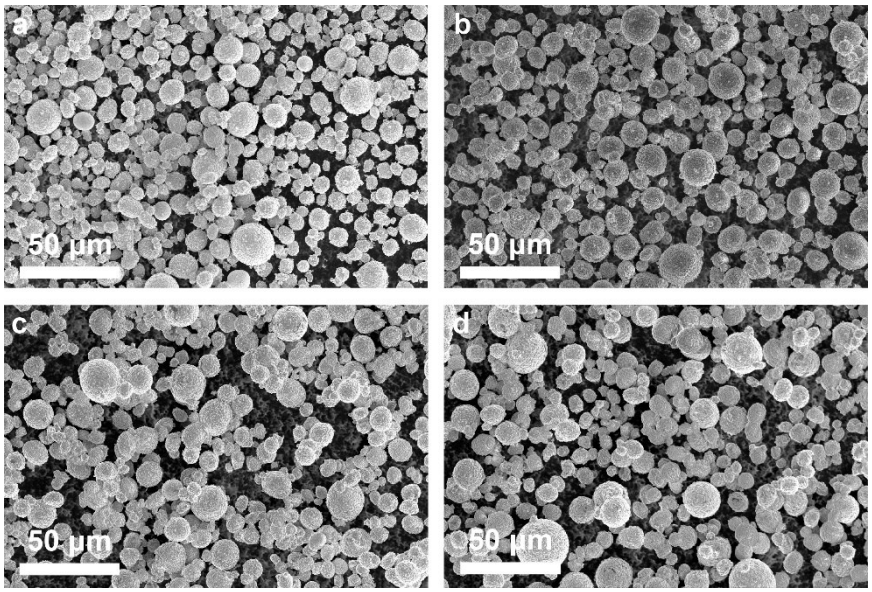

Figure S2. The SEM images of four samples: (a) NCA, (b)NCA-S, (c)NCA-SO and (d) NCA-SLO.

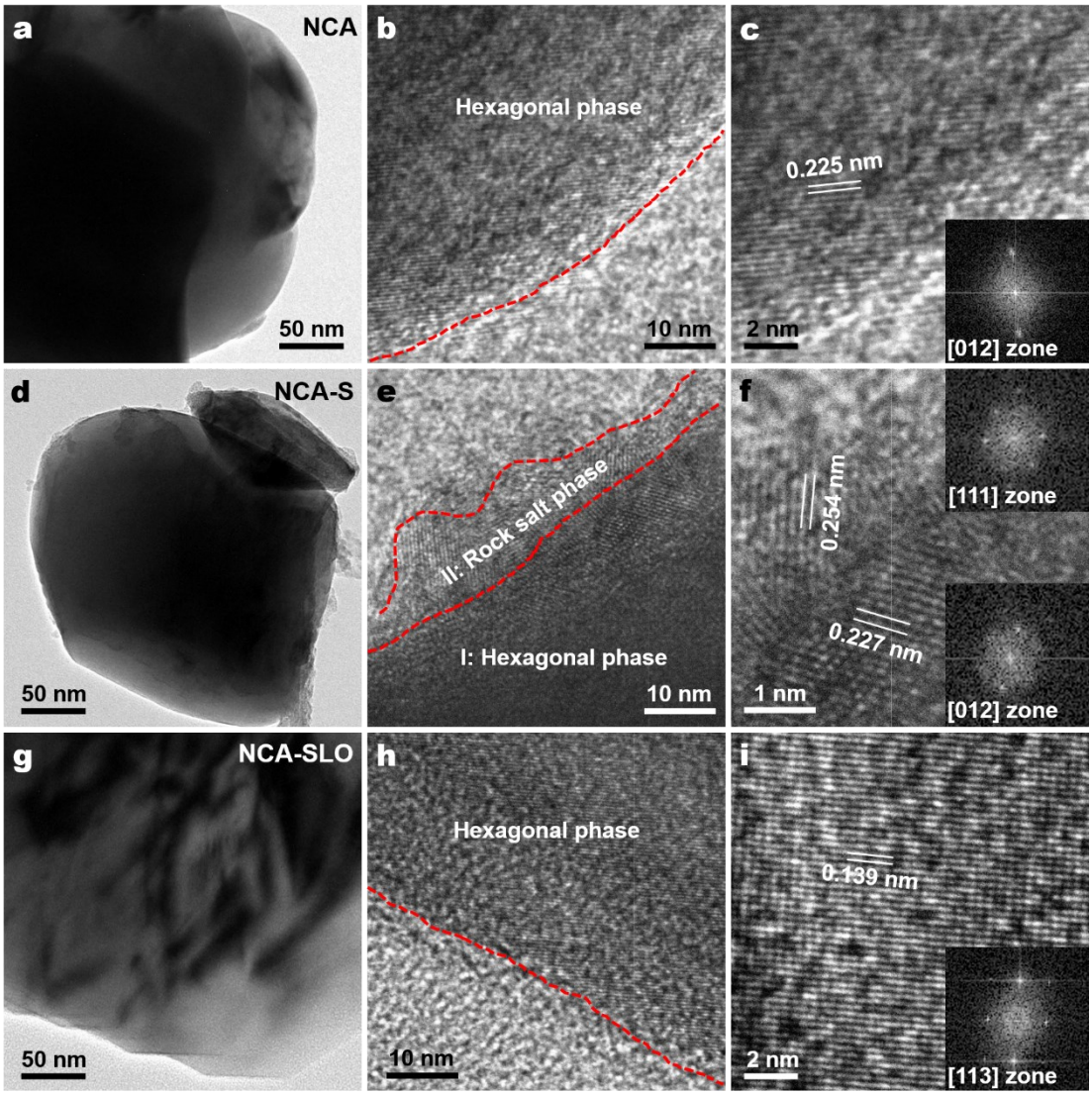

Figure S3. TEM images and corresponding HRTEM images for the microstructure evolution of NCA samples at different stages: (a-c) pristine NCA, (d-f) NCA-S, and (g-i) NCA-SLO.

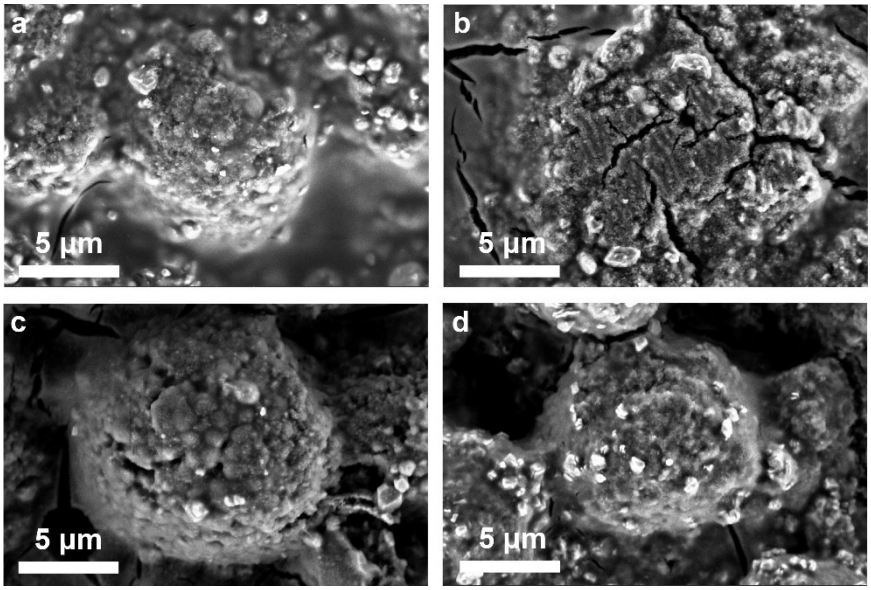

Figure S4. The SEM images of cycled electrodes: (a) NCA, (b)NCA-S, (c)NCA-SO and (d) NCA-SLO.

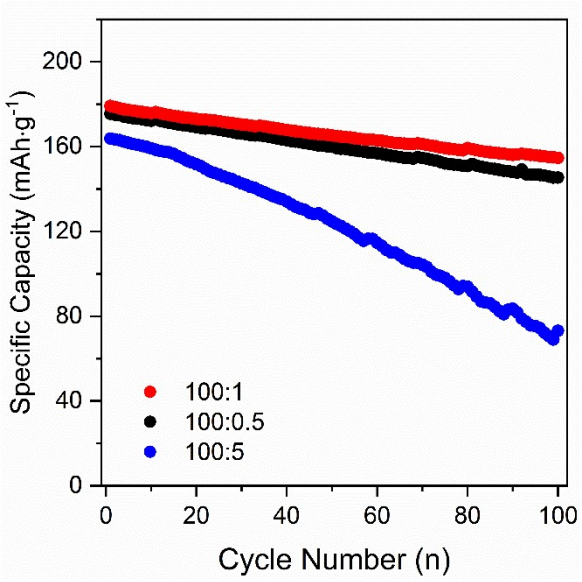

Figure S5. Cycling performances of samples with different mass ratios of NCA-S to $LiOH \cdot H_2O$ at 1 C.

The samples with different mass ratios of NCA-S to $LiOH \cdot H_2O$ manifested various discharge capacities and capacity retention after 100 cycles at 1 C (145.4 $mAh \cdot g^{-1}$ and 82.8% for "100:0.5", 154.7 $mAh \cdot g^{-1}$ and 86.4% for "100:1", and 73.1 $mAh \cdot g^{-1}$ and 44.6% for "100:5"). In the case of a small amount of lithium source, the lack of lithium cannot be completely replenished, resulting in incomplete recovery of the electrochemical performance of the materials. When the amount of lithium source is excessive, the residual alkali will be generated on the surface of the material again, which will damage the electrochemical performance of the material. After systematic comparison, we have determined that the optimal mass ratio of NCA-S to $LiOH \cdot H_2O$ is 100:2.

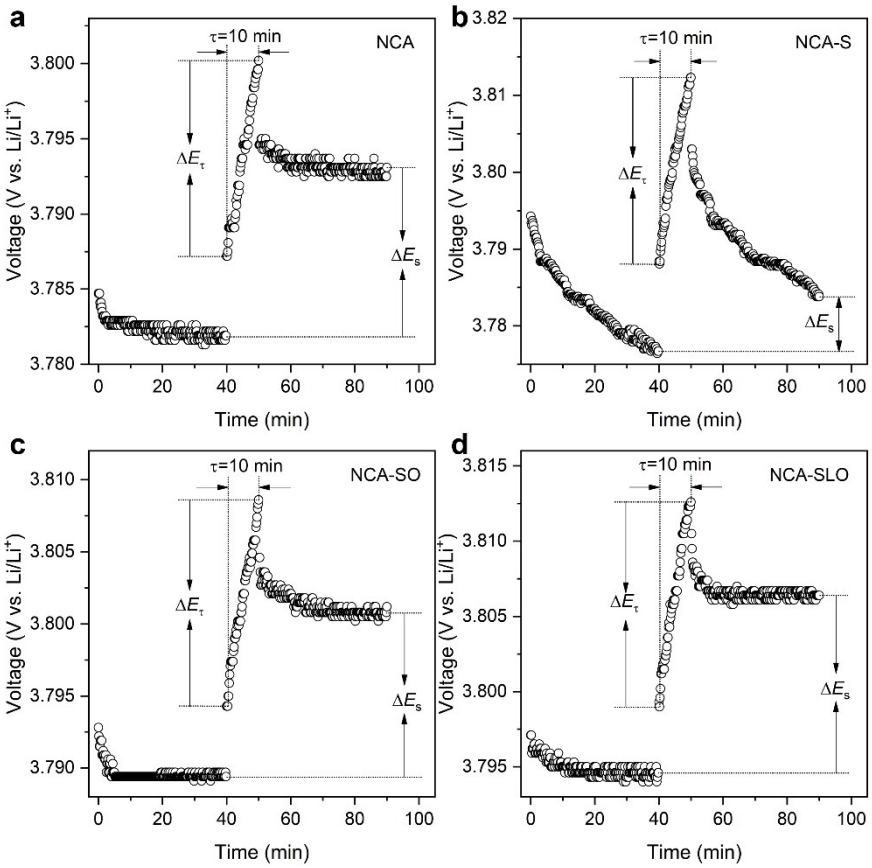

Figure S6. *t* vs. *E* for a single GITT titration of the (a) NCA, (b) NCA-S, (c) NCA-SO and (d) NCA-SLO.

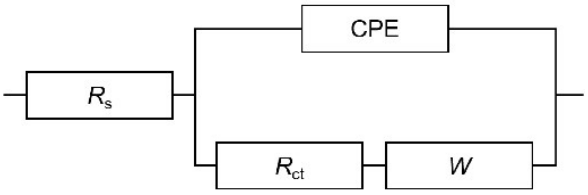

Figure S7. The simplified equivalent circuit of the samples.