

Exploring Degradation Pathways of Nickel-rich Cathode During High-Temperature Storage in High-Energy Lithium-ion Batteries

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Supporting Figures and Table

| Material selection | | |
|--|---|--|
| | Cathode | Anode |
| Active material | $\text{LiNi}_{0.88}\text{Co}_{0.1}\text{Al}_{0.02}\text{O}_2$ Specific capacity: 210 mAh g ⁻¹ , ICE: 91% | Graphite Specific capacity: 360 mAh g ⁻¹ ICE: 92% |
| Conductive agent | Carbon black | - |
| Binder | Polyvinylidene fluoride (PVDF) | Styrene butadiene rubber (SBR) and carboxymethyl cellulose (CMC) |
| Electrode engineering | | |
| | Cathode | Anode |
| Electrode composition (Active material:Binder:Conductive agent) | 96:02:02 | 97:1.5:1.5 |
| Loading level (mg cm ⁻²) | 22.5 | 14.2 |
| N/P ratio | 1.3 | |
| Electrode density (g cc ⁻¹) | 3.6 | 1.5 |
| Full-cell assembly | | |
| Cell structure | Stacking type pouch cell | |
| | Cathode | Anode |
| Number of stacks | 2 | 3 |
| Cell dimension | 68.5 mm*50 mm | 72.5 mm*54 mm |
| Cell capacity | 0.7 Ah | |

Table S1. Detailed information for the full-cell.

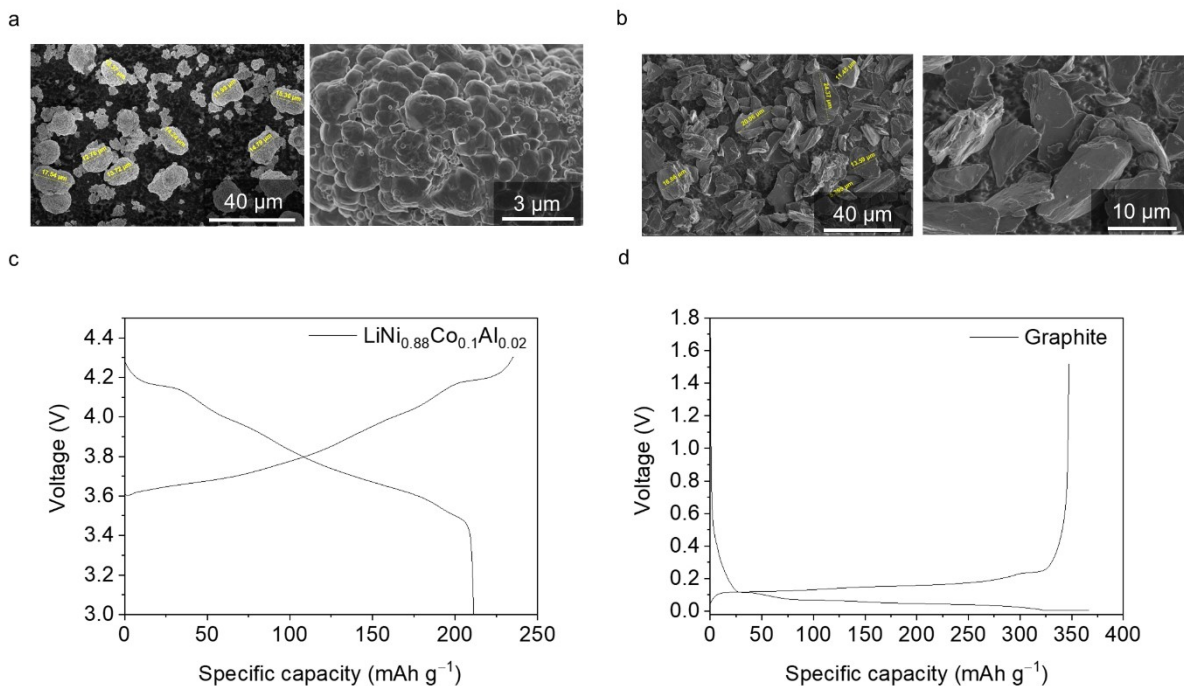
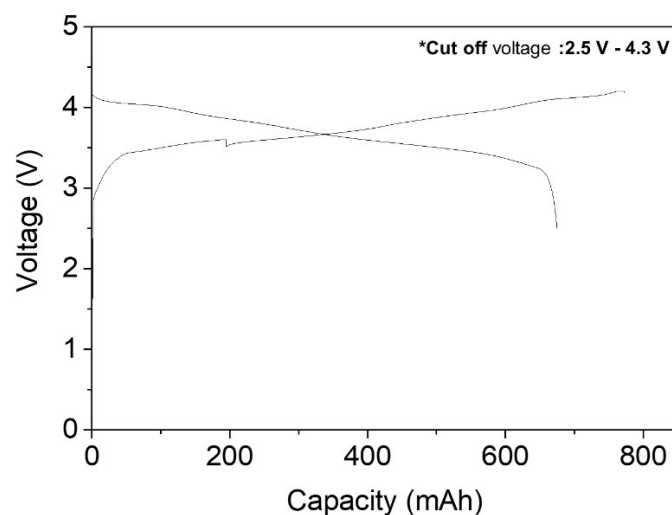


Fig. S1. SEM images of (a) $\text{LiNi}_{0.88}\text{Co}_{0.1}\text{Al}_{0.02}\text{O}_2$ and (b) graphite. Voltage profiles of formation step for (c) $\text{LiNi}_{0.88}\text{Co}_{0.1}\text{Al}_{0.02}\text{O}_2$ cathode at voltage range of 3.0 V - 4.3 V at 0.1C-rate and (d) graphite anode at voltage range of 0.005 V - 1.5 V at 0.1C-rate.



| Charge capacity | Discharge capacity | Initial Coulombic Efficiency |
|-----------------|--------------------|------------------------------|
| 772 mAh | 675 mAh | 87.4 % |

Fig. S2. Voltage profile of $\text{LiNi}_{0.88}\text{Co}_{0.1}\text{Al}_{0.02}\text{O}_2/\text{Graphite}$ full-cell at voltage range of 2.5 V – 4.2 V at 0.1C-rate.

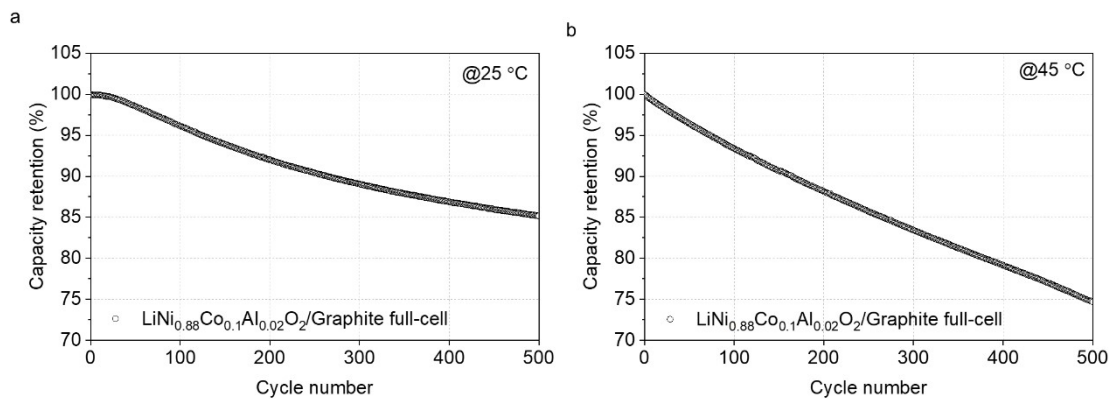


Fig. S3. Cycle performance of $\text{LiNi}_{0.88}\text{Co}_{0.1}\text{Al}_{0.02}\text{O}_2/\text{Graphite}$ full-cell at voltage range of 2.5 V – 4.2 V at 1C-rate at (a) room temperature (25 °C) and (b) 45 °C.

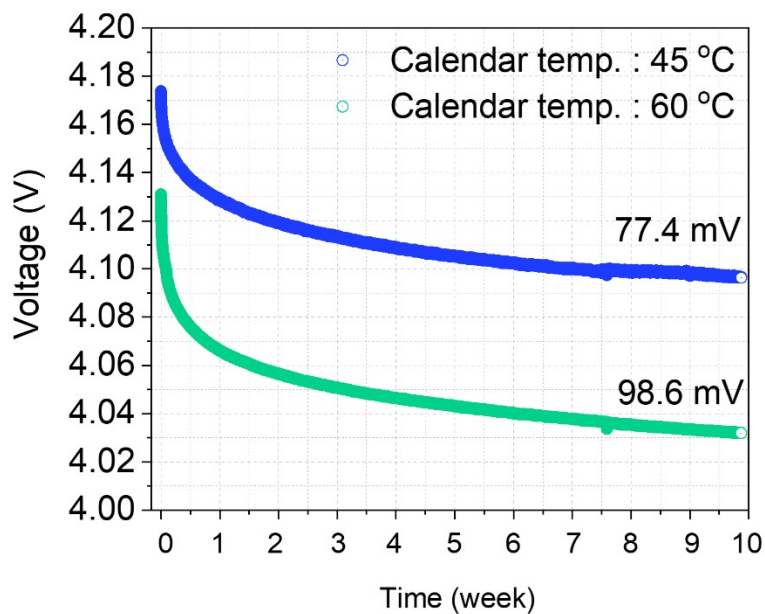


Fig. S4. Self-discharge during calendar life test at 45 °C and 60 °C

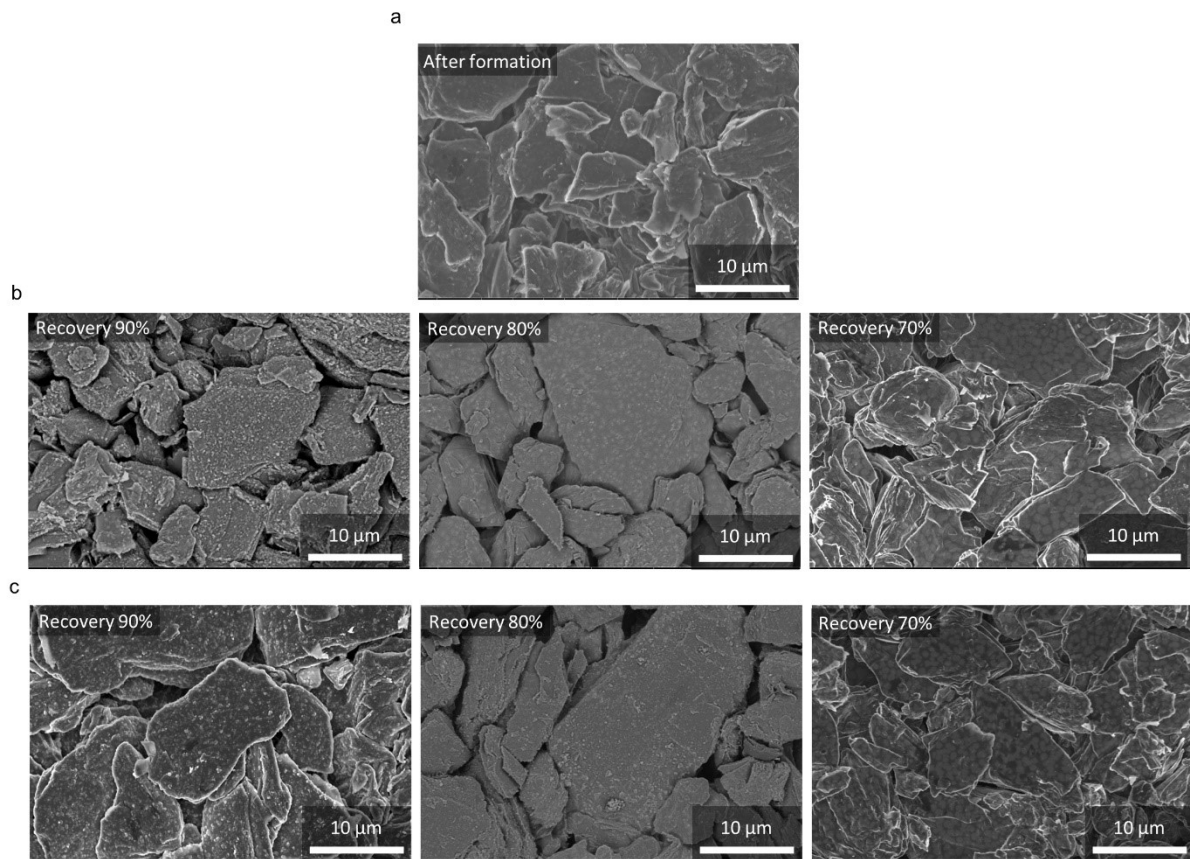


Fig. S5. (a) SEM images graphite anodes after formation. SEM images of graphite anodes of the full-cell with recovery 90%, 80% and 70% stored at (b) 45 °C and (c) 60 °C.

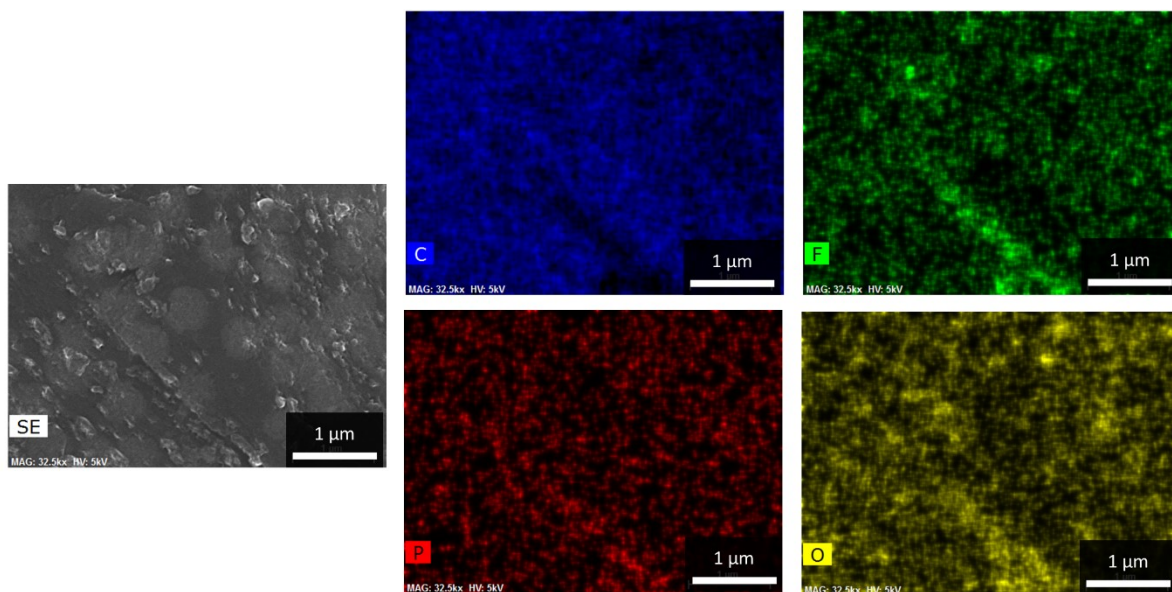


Fig. S6. (a) EDX mapping images of graphite surface, showing capacity retention of 70% after storing at 60 °C.

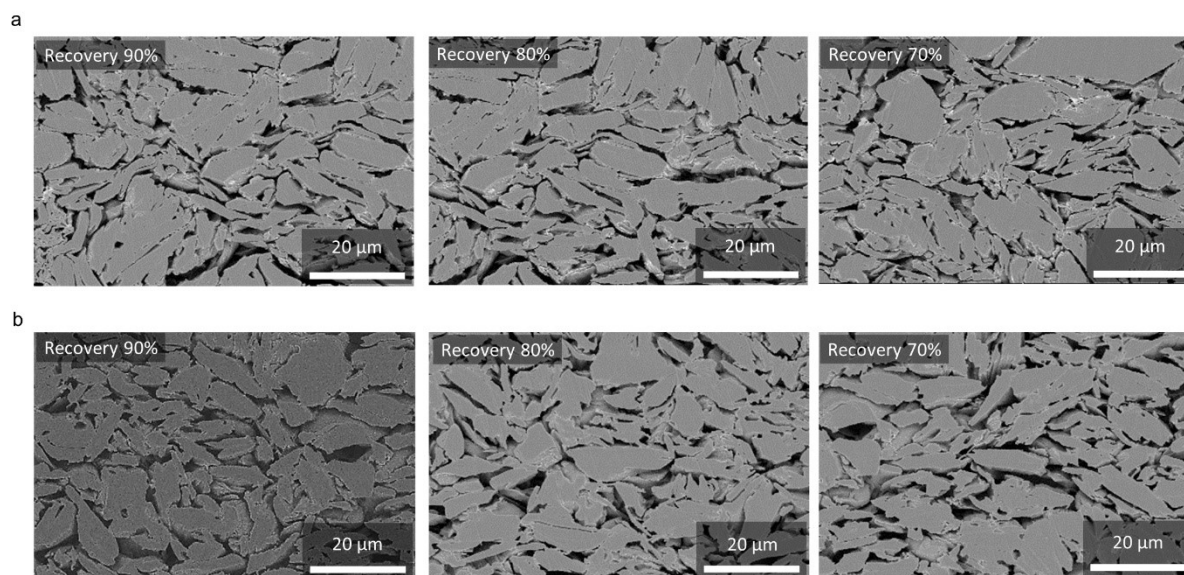


Fig. S7. Cross-section SEM images of graphite anodes of the full-cell with recovery 90%, 80% and 70% stored at (a) 45 °C and (b) 60 °C.

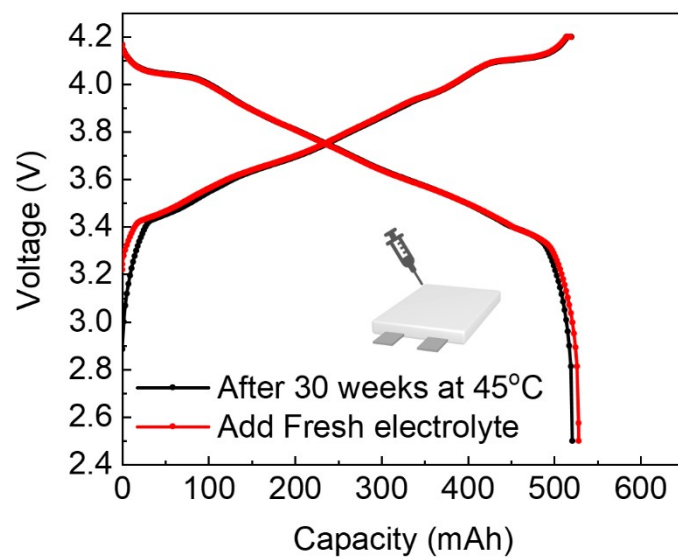


Fig. S8. Voltage profiles of full-cells after 30 weeks stored at 45 °C and after adding 1 mL of electrolyte

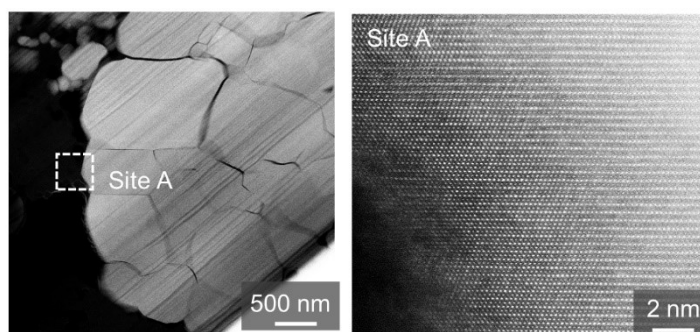


Fig. S9. HR-TEM images of $\text{LiNi}_{0.88}\text{Co}_{0.1}\text{Al}_{0.02}\text{O}_2$ particle after storage at 60 °C for 10 weeks

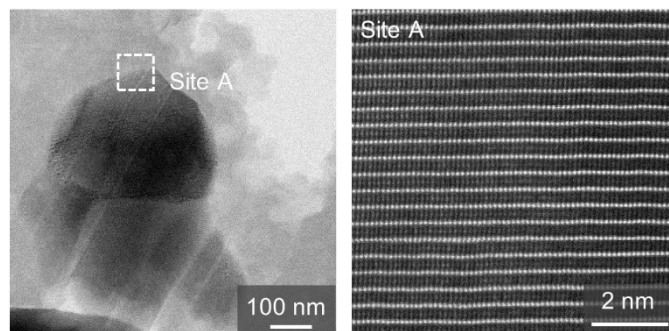


Fig. S10. HR-TEM images of $\text{LiNi}_{0.88}\text{Co}_{0.1}\text{Al}_{0.02}\text{O}_2$ particle before storage