Supporting Information

Study of degradation mechanisms in aqueous-processed Ni-rich cathodes for Enhanced Sustainability of Batteries

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Supplementary Figures

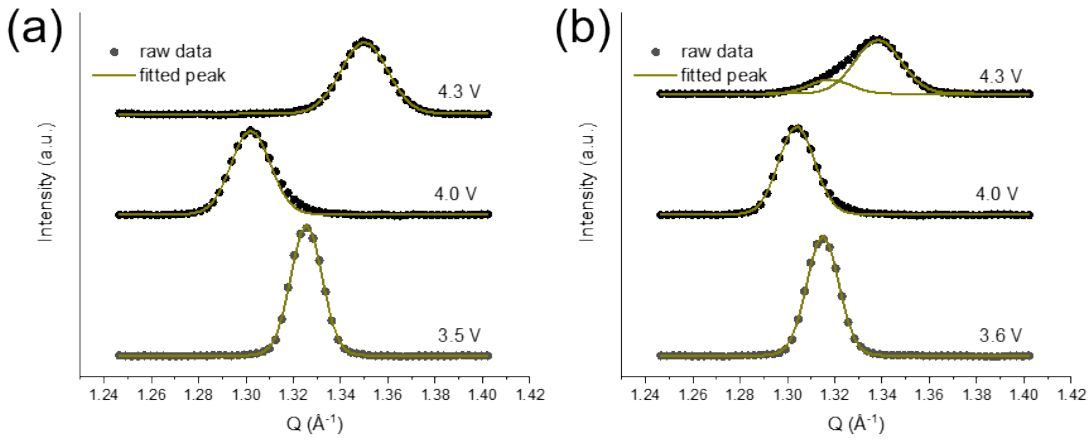

Figure S1. Single peak fitting for the 003 reflection of fresh NMP_{NMC} electrodes (a), single and double peaks fitting for the 003 reflection of aged H_2O_{NMC} electrodes (b) under various states of charge.

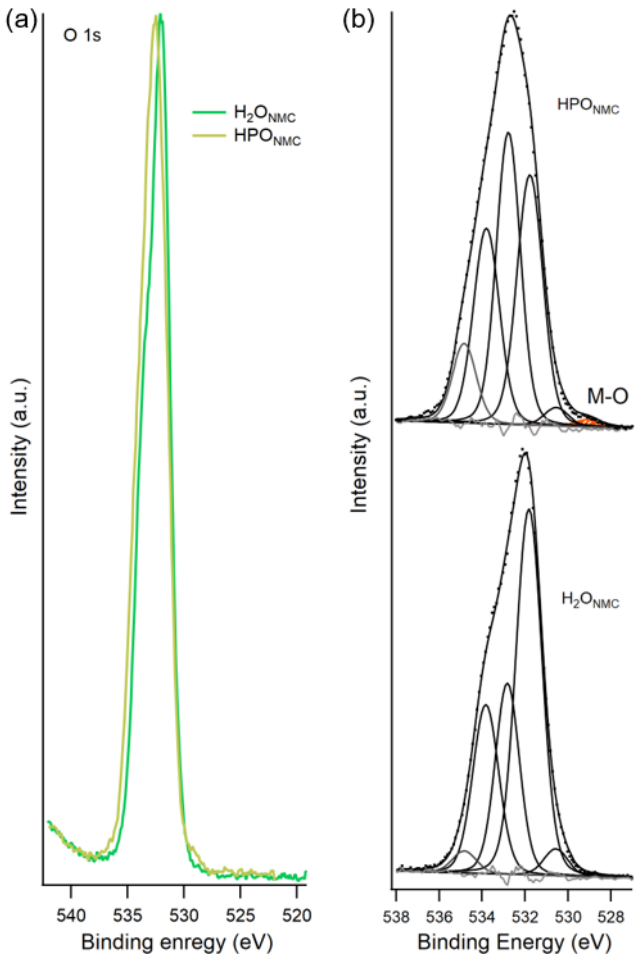

Figure S 2. The overlaid (a) and the curve fitted (b) O 1s spectra collected from H_2O_{NMC} and HPO_{NMC} electrodes after 105 cycles with photon energy 1486.7 eV, normalized to the highest peak intensity.

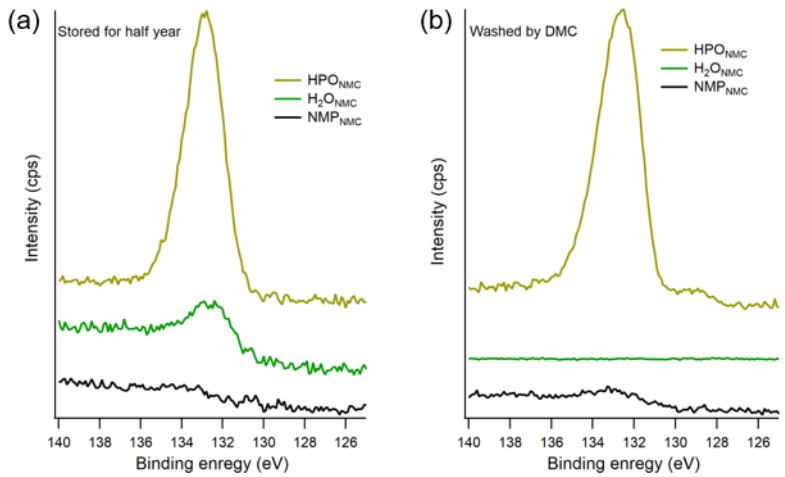

Figure S3. The P2p spectra were collected from pristine $NMP_{NMC,}$ $H_2O_{NMC,}$ and HPO_{NMC} electrodes after half a year of storage inside the Ar-filled glovebox (a) and after being washed by DMC (b) using the Al Kα source

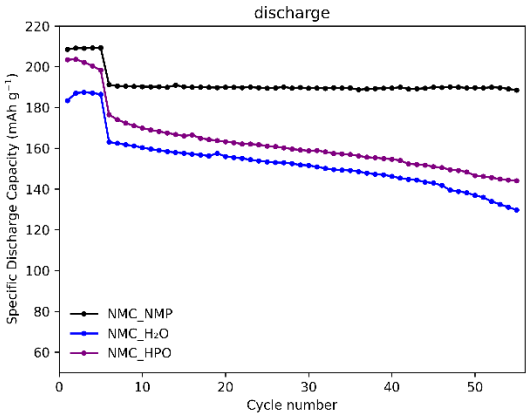

Figure S4. The specific discharge capacity of three electrodes with galvanostatic cycling at 0.1C for 5 cycles and 0.5C for 55 cycles within the voltage range of 3.0-4.3V vs. Li+/Li before Operando XRD measurements.

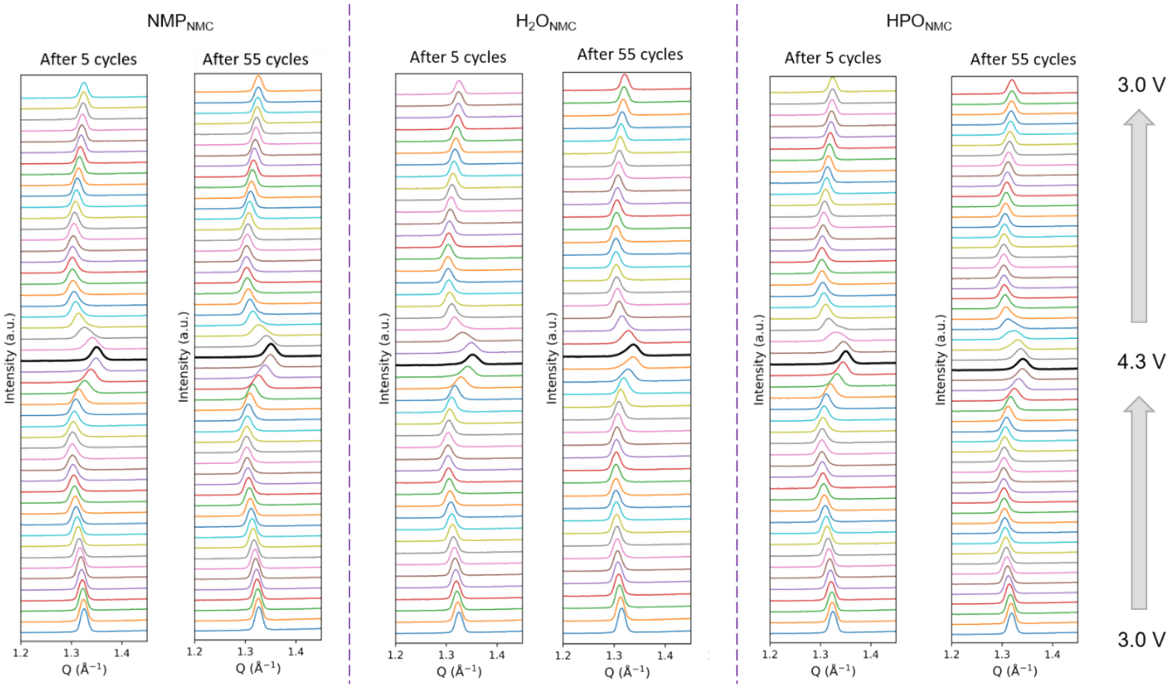

Figure S5. Waterfall plot of 003 reflection of NMP_{NMC} (a), H_2O_{NMC} (b), and HPO_{NMC} (c) electrodes after 5 formation and 55 cycles during cycling between 3.0 V and 4.3 V.

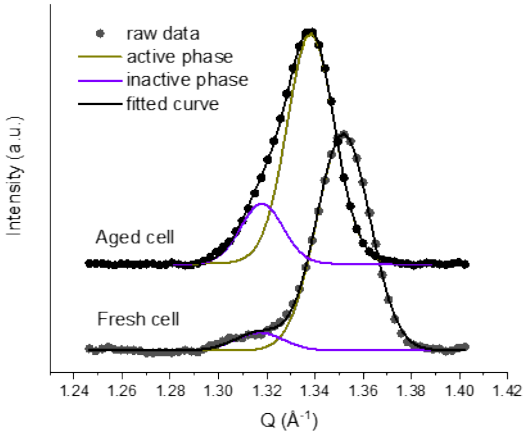

Figure S6. 003 reflection curves of fresh and aged H_2O_{NMC} at 4.3V fitted with two distinct peaks representing to electrochemically active and inactive phase, respectively.

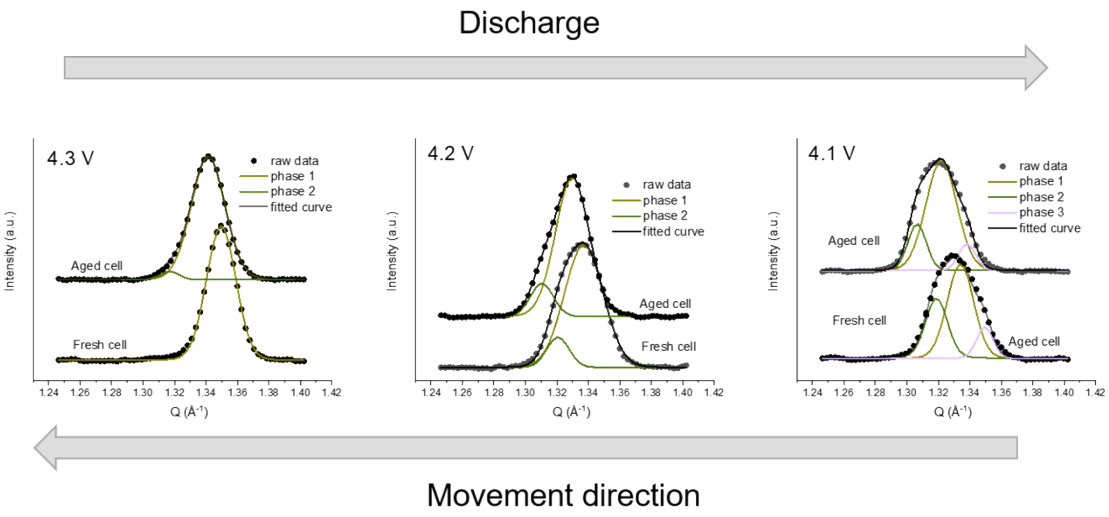

Figure S7. 003 reflection curves of the fresh and aged HPO_{NMC} electrode during discharge at ~4.3 V, ~4.2 V, and ~4.1 V, fitted with two or three peaks corresponding to different distinct phases.

Supplementary Table

Table S1. Ni concentrations in the electrolytes collected from three aged batteries.

Cathodes	NMP_{NMC}	H_2O_{NMC}	HPO_{NMC}
Concentration of Ni (ppm) in electrolyte	6.0	2.4	4.6