

Supporting Information

Mitigating Hydrogen Gas Evolution in High Nickel Cathodes Using Single-Crystalline NCM Particles

Nyung Joo Kong[‡], Jee Ho Ha[‡], Yeon Jeong Hwang, Yujin Kim, Byung Un Hwang, Kyeong-

Min Jeong, Jaephil Cho and Seok Ju Kang**

Department of Energy Engineering, School of Energy and Chemical Engineering, Ulsan National Institute of Science and Technology (UNIST), Ulsan 44919, Republic of Korea; orcid.org/0000-0002-9921-6674; Email: jpcho@unist.ac.kr, sjkang@unist.ac.kr [‡]These authors contributed equally to this work.

SUPPORTING TABLE AND FIGURES

Table S1. Rietveld refinement results for poly- and single-crystal-NCM particles, representing the cation mixing ratio based on the lattice constants of a and c .

	a	c	Cation mixing
PC-NCM	2.8739	14.2041	1.404%
SC-NCM	2.8790	14.1968	2.868%

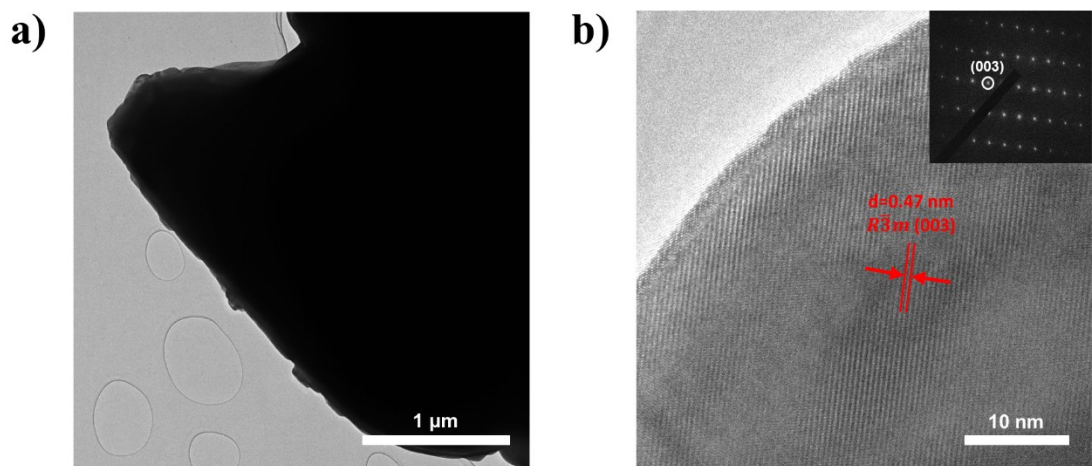


Figure S1. a) Bright field TEM image of single-crystal NCM on a holey carbon-supported Cu grid. b) High-resolution TEM image and corresponding SAED pattern of single-crystal NCM.

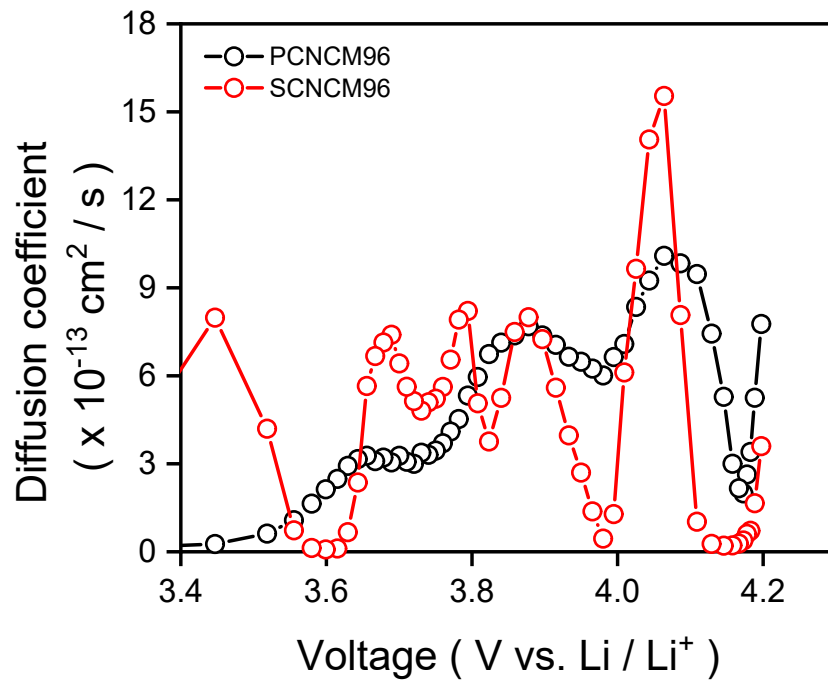


Figure S2. Li diffusivity coefficients for poly- and single-crystal NCM cathodes.

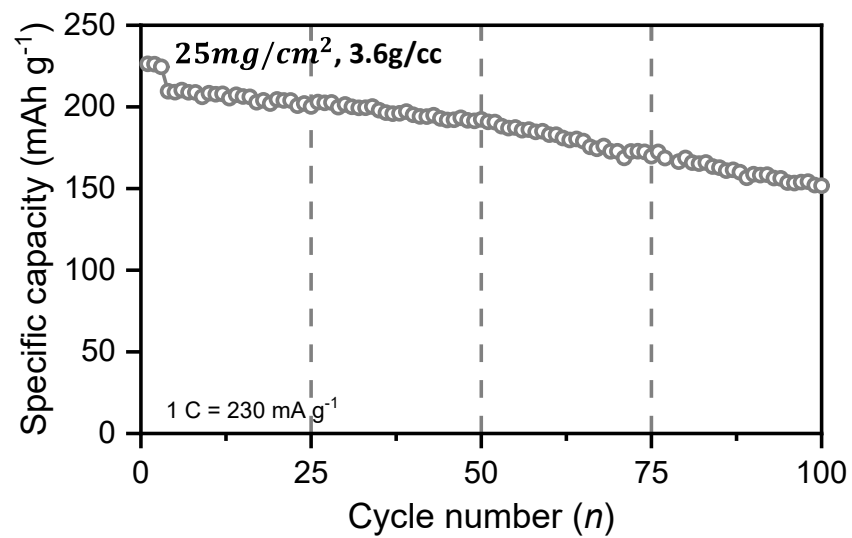


Figure S3. Cycling performance of high tap density single-crystal NCM cathodes at a current density of 0.2C.

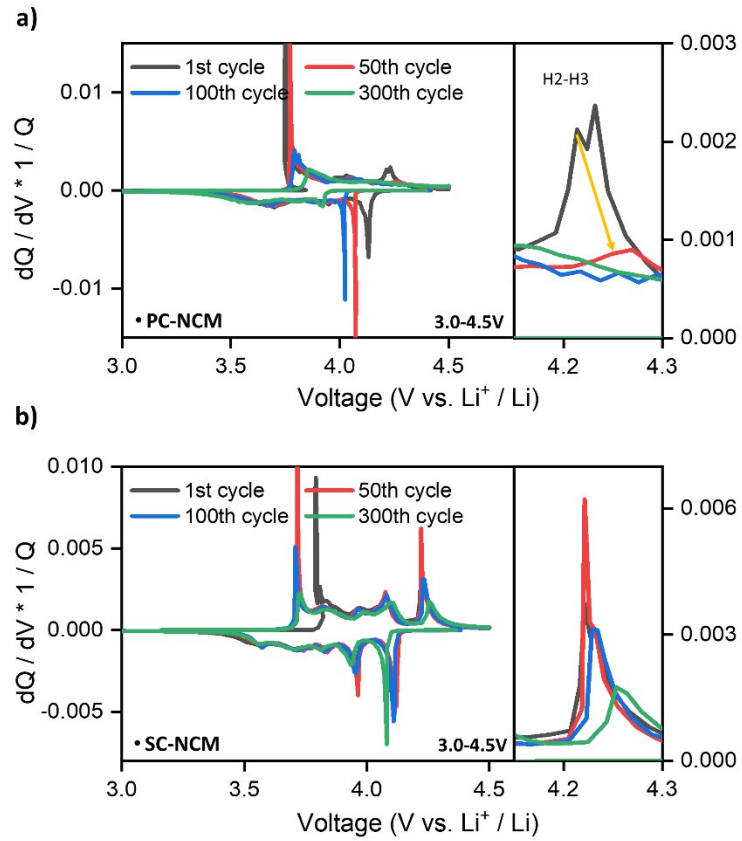


Figure S4. Plots of dQ/dV versus voltage for a) poly- and b) single-crystal NCM cathodes during the cycling process. Right panels in a) and b) show magnified views of the H2-H3 phase transition redox peaks from the main panels.

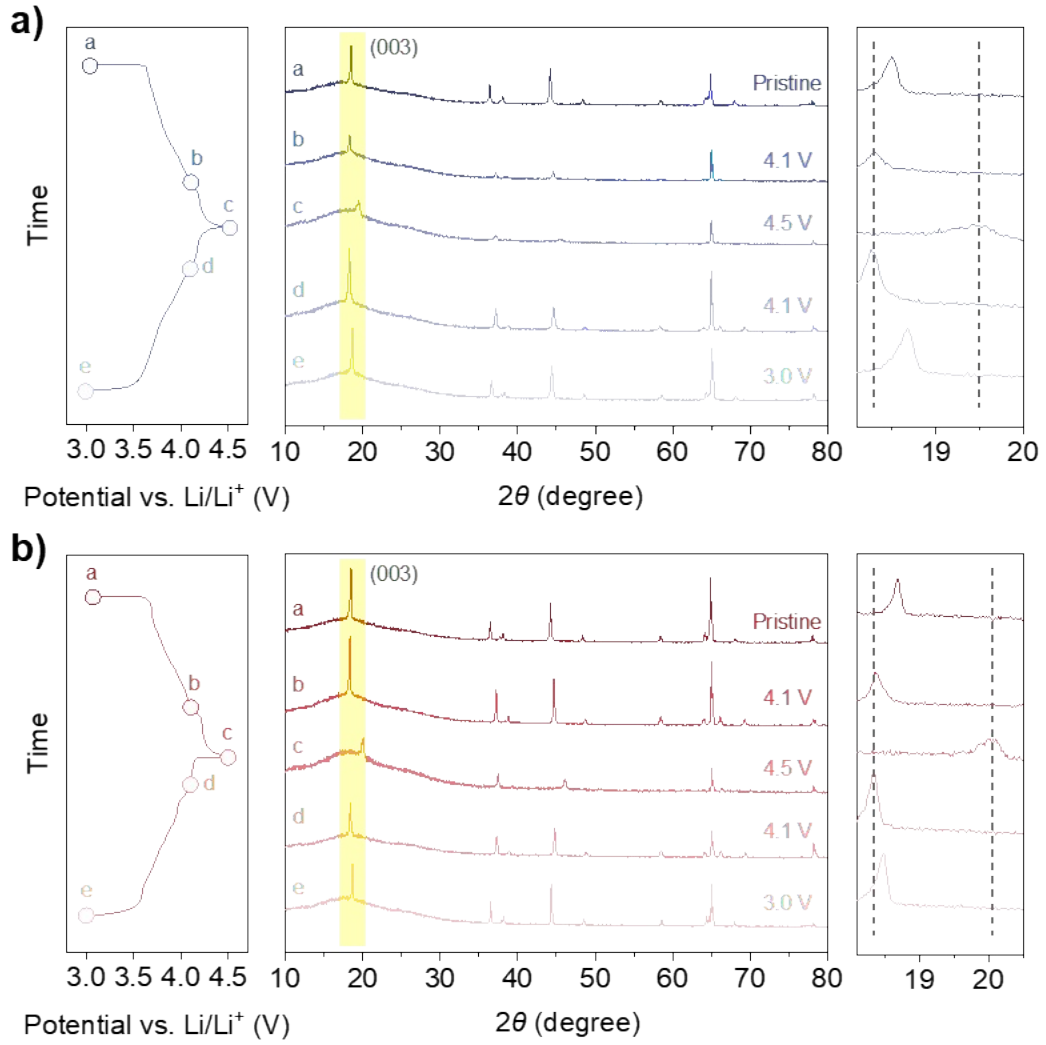


Figure S5. Ex-situ XRD patterns of a) poly- and b) single-crystal NCM cathodes at various charge and discharge states.

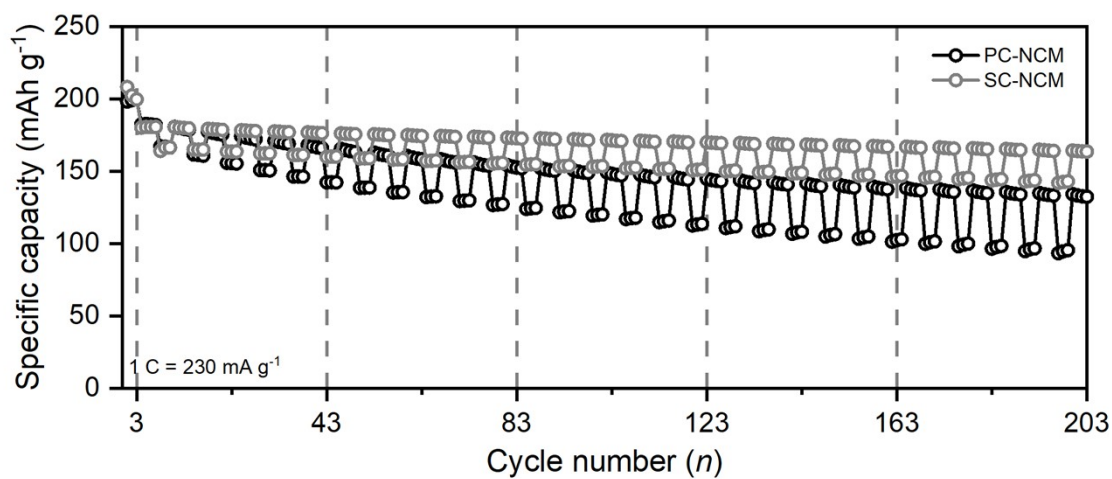


Figure S6. Cycling performance of full cells containing poly-(black) and single-crystal (grey) NCM cathodes under the charge conditions: four normal (0.5 C) and three fast (3 C) charge protocols.

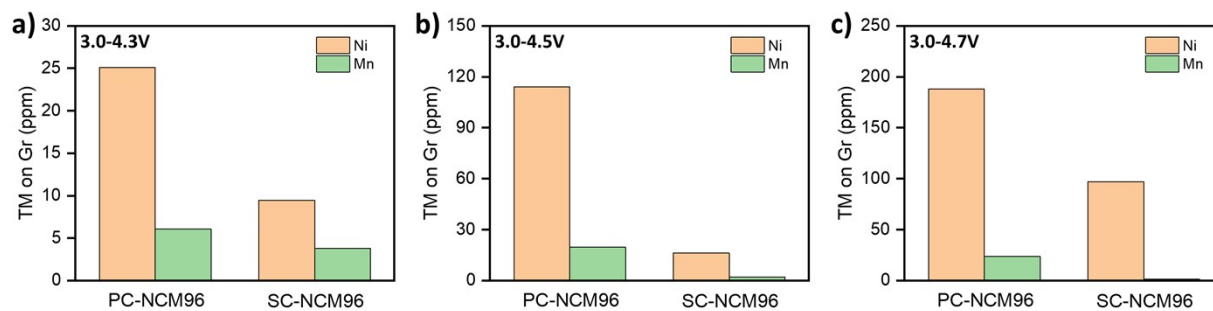
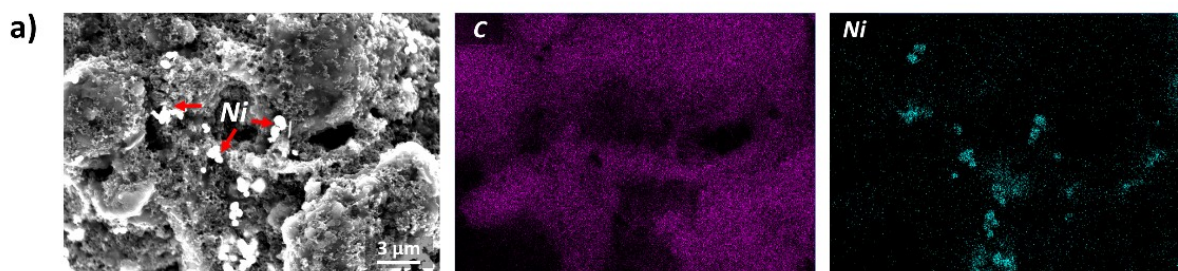
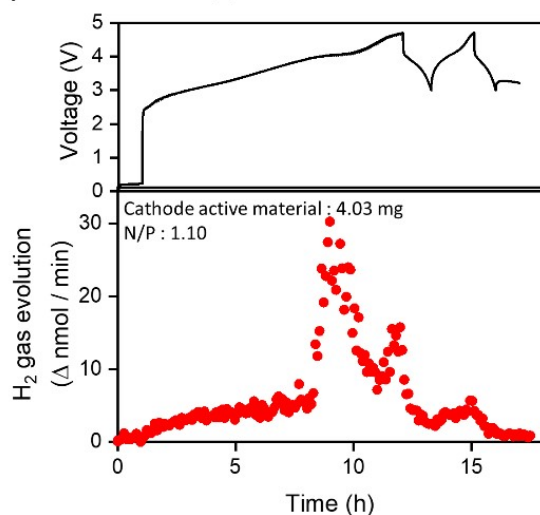


Figure S7. ICP-OES results for 1000 cycled graphite anodes in full cells containing poly- and single-crystal cathodes, under various cutoff conditions: a) 3.0–4.3 V, b) 3.0–4.5 V, and c) 3.0–4.7 V.



b) •PC-NCM96 || Gr + Ni



•SC-NCM96 || Gr + Ni

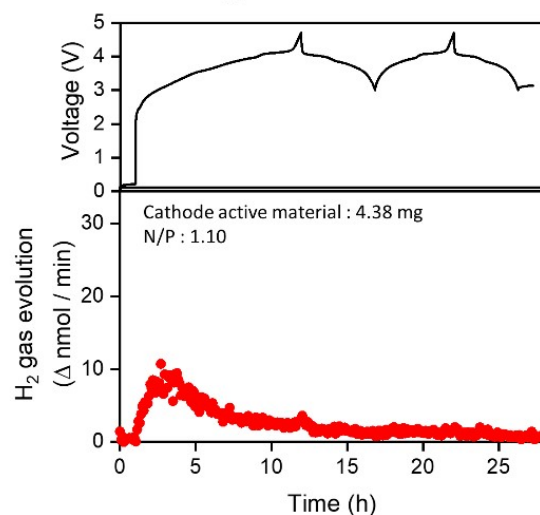


Figure S8. a) SEM image and corresponding EDS elemental mapping of graphite electrode with 5 wt% Ni powder composite. b) Initial galvanostatic charge and discharge profiles with corresponding H_2 evolution for full cells containing (b) poly- and (c) single-crystal NCM cathodes paired with graphite anodes with 5 wt% Ni powder.

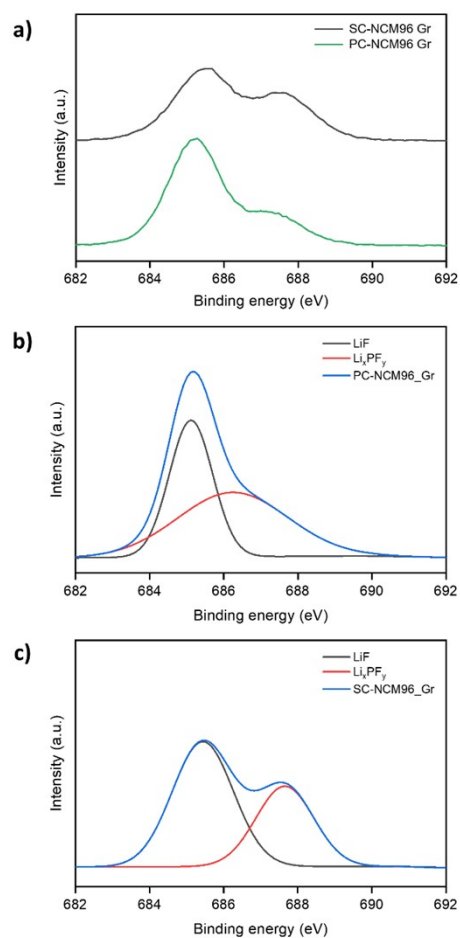


Figure S9. High resolution F 1s XPS spectra of a) pristine poly- and single-crystal-NCM cathodes and cycled b) poly- and c) single-crystal-NCM cathodes. The deconvolution in b) and c) shows the relative amount of LiF (black line) in cycled graphite anodes.