Supporting information

Optimized Al Doping Improves both Interphase Stability and Bulk Structural Integrity of Ni-Rich NMC Cathode Materials

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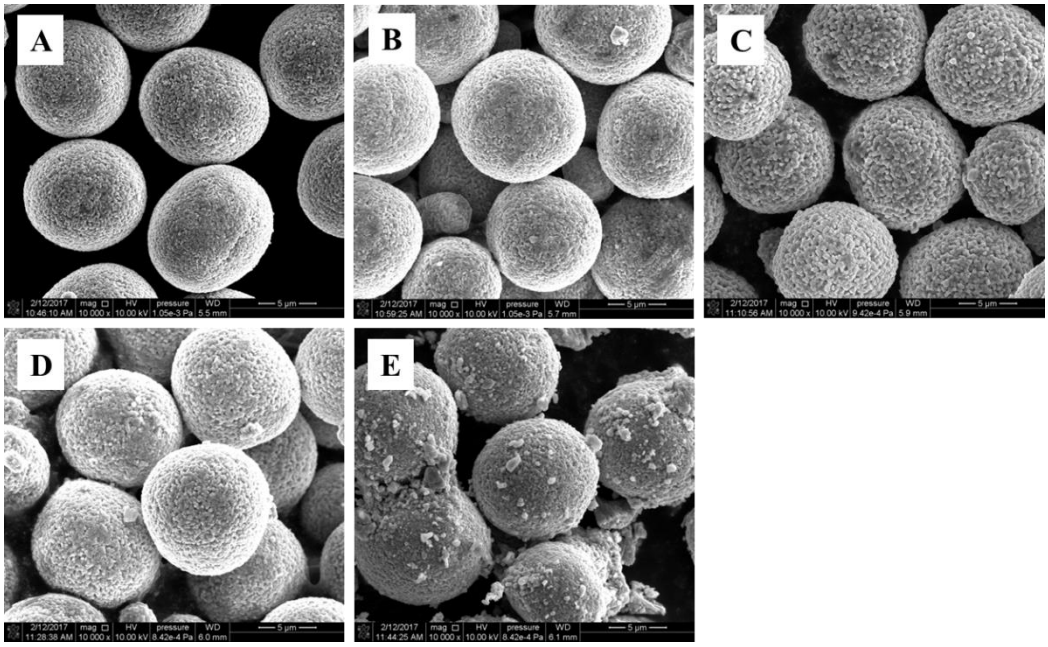

Figure S1. SEM images of pristine NMC76 and Al-doped NMC76 with different Al contents: (A) pristine; (B) 0.5%Al-NMC76; (C) 1%Al-NMC76; (D) 2%Al-NMC76; (E) 5%Al-NMC76.

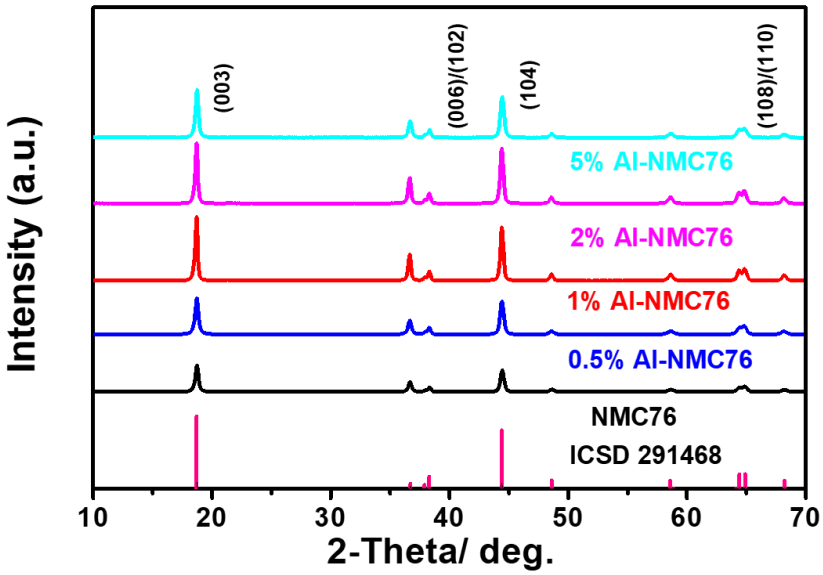

Figure S2. XRD patterns of pristine NMC76 and NMC76 doped with various Al contents.

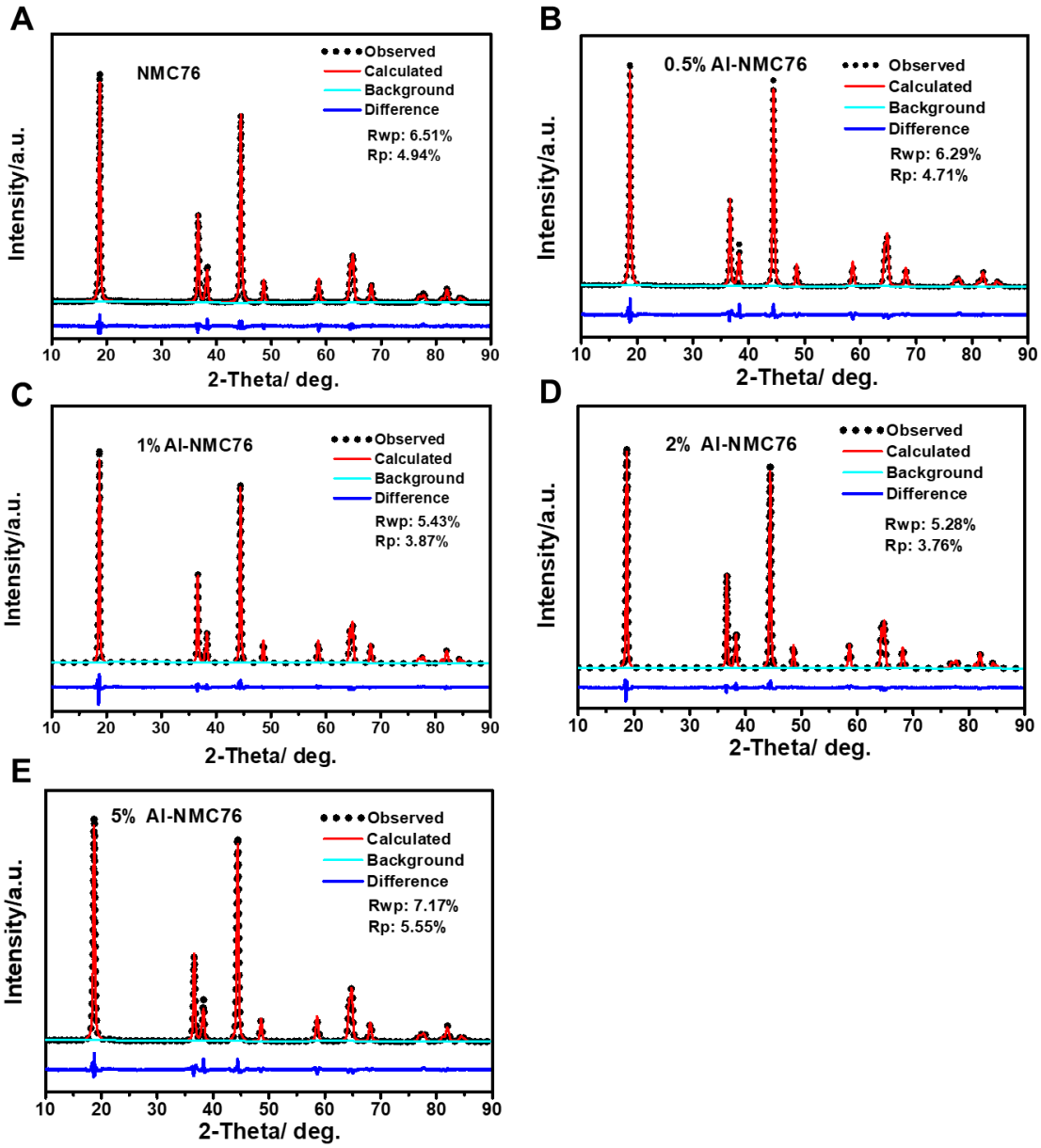

Figure S3. Rietveld refinement results of XRD patterns for NMC76 (A) and Al-doped NMC76 with different Al content, (B) 0.5%Al-NMC76, (C) 1%Al-NMC76, (D)2%Al-NMC76, and (E) 5%Al-NMC76.

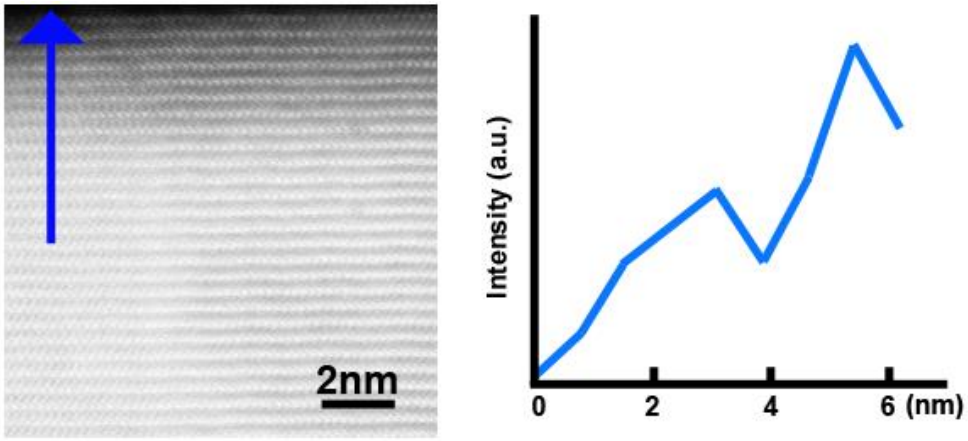

Figure S4. STEM line scanning of the 1%Al-NMC76.

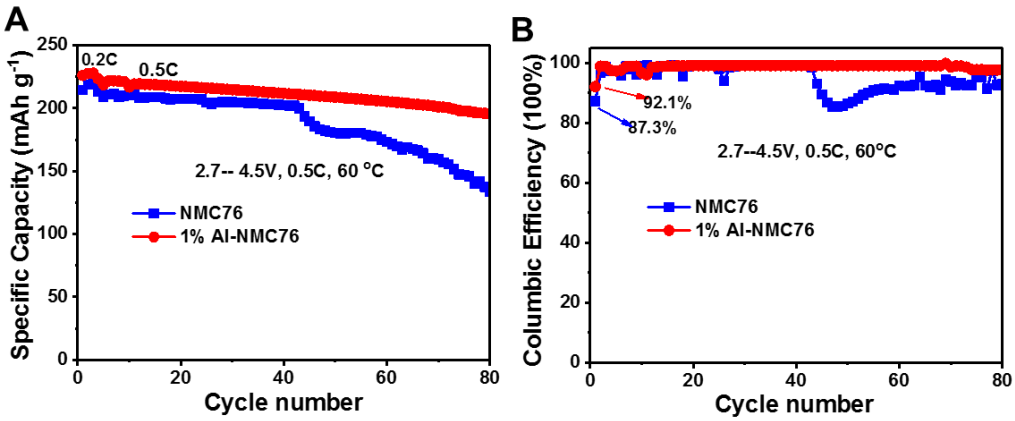

Figure S5. High temperature (60°C) cycling performance (A) and CE (B) of NMC76 and 1%Al-NMC in Li half-cells at a current density of 0.5C.

Table S1. Electrochemical performance comparison between our sample and other Al-doped Ni-rich NMC cathodes.

NMC cathode	Electrolyte	Voltage (V)	Capacity retention	References
$LiNi_{0.8}Co_{0.1}Mn_{0.09}Al_{0.01}O_2$	1.0M $LiPF_6$ in EC: DMC = 1:1 v%	2.7-4.3V	78.9% (1C, 200 cycles)	34
$LiNi_{0.48}Co_{0.2}Mn_{0.3}Al_{0.02}O_2$	1.0M $LiPF_6$ in LEC: DMC: EMC (1:1:1 v%)	3.0-4.6V	82.1% (0.5C, 200 cycles)	35
$Li[Ni_{0.865}Co_{0.120}Al_{0.015}]O_2$	1.2M $LiPF_6$ in EC: EMC (3: 7 v%) + 2 wt% VC	2.7-4.3	91.6% (0.5C, 100 cycles)	25
$LiNi_{0.88}Co_{0.095}Mn_{0.025}Al_{0.02}O_2$	1.2M $LiPF_6$ in EC: EMC (3:7v%) + 2 wt% VC	2.8-4.3V	91.5% (1C, 150 cycles)	36
$LiNi_{0.92}Co_{0.06}Al_{0.0}2O_2$	1.2M $LiPF_6$ in EC: EMC (3: 7 v%) + 2 vol% VC	2.8-4.4V	86% (1C, 300 cycles)	11
1%Al-NMC76	1.0M $LiPF_6$ in EC: EMC = 4: 6 v%	2.7-4.5V	87.7% (0.33C, 250) 79.2% (0.2C/5C, 500)	This work

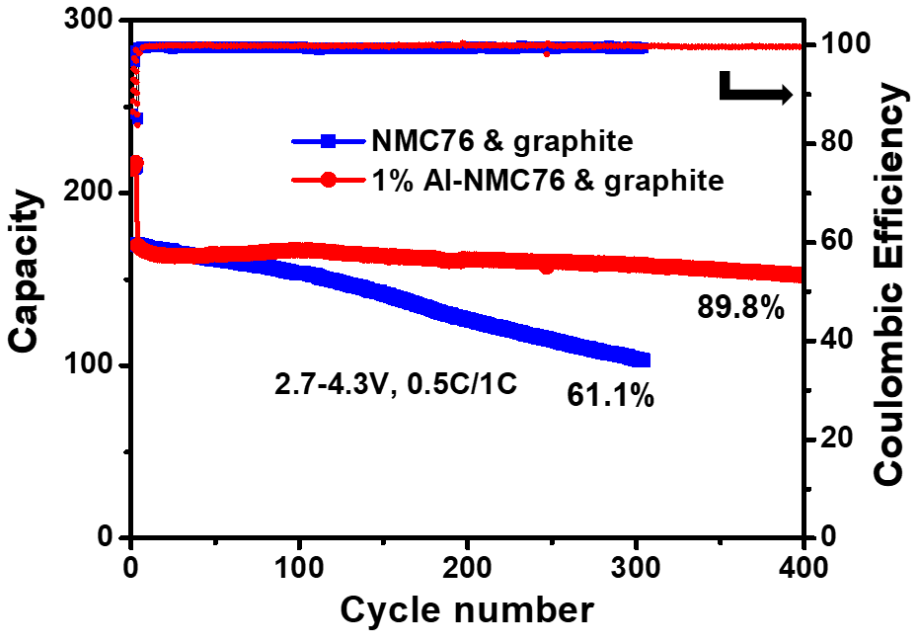

Figure S6. Comparison of long-term cycling performance of NMC76 and 1%Al-NMC76 cathodes with graphite anodes in coin-type full cells between 2.7 and 4.3 V at 0.5C/1C.

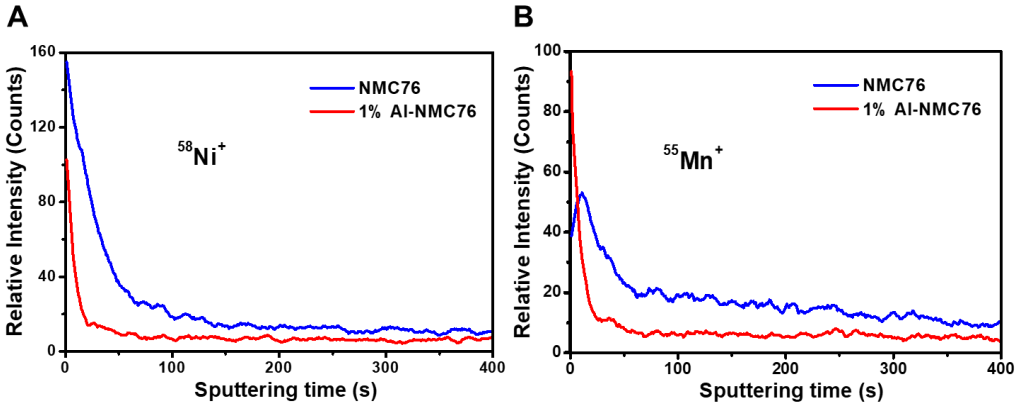

Figure S7. TOF-SIMS depth profiles (normalized with cycled graphite in 1%Al-NMC76 based cell) of secondary ions fragments on the cycled graphite: (A) $^{58}Ni^{+}$; (B) $^{55}Mn^{+}$. The cycled graphite anodes were harvested from coin-type full cells composed NMC76 and 1%Al-NMC76 with graphite anode after 400 cycles at 0.5C/1C charge/discharge in the voltage of 2.5-4.3V.

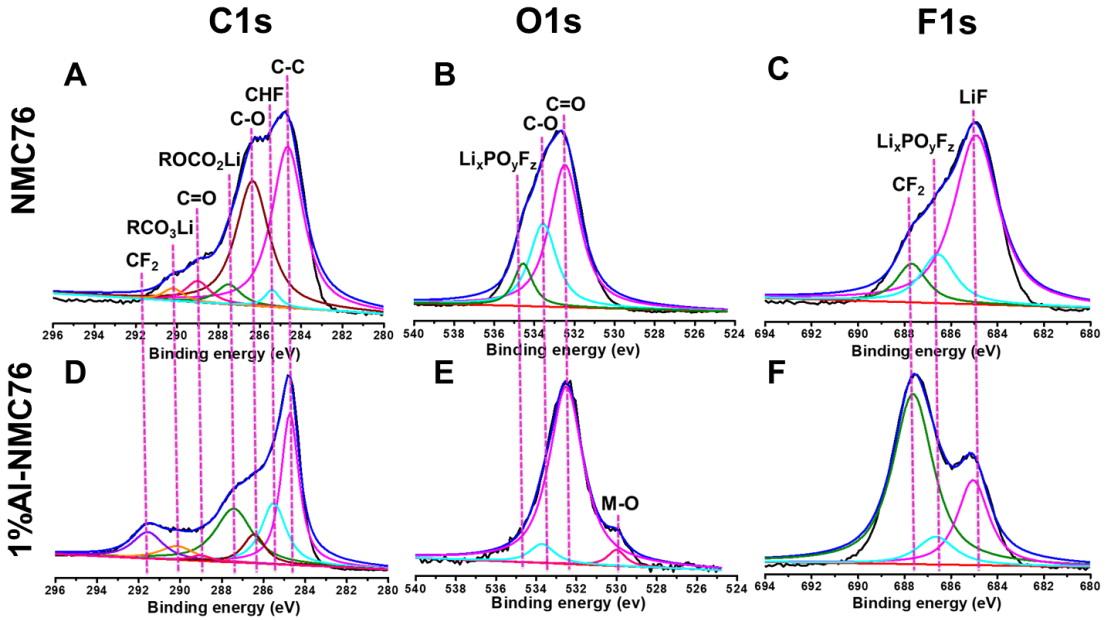

Figure S8. XPS characterization of cycled 1%Al-NMC after 500 cycles. Top row: (A) C1s, (B) O1s, and (C) F 1s of cycled NMC76; Bottom row: (D) C 1s, (E) O1s, and (D) F 1s of cycled 1%Al-NMC76.

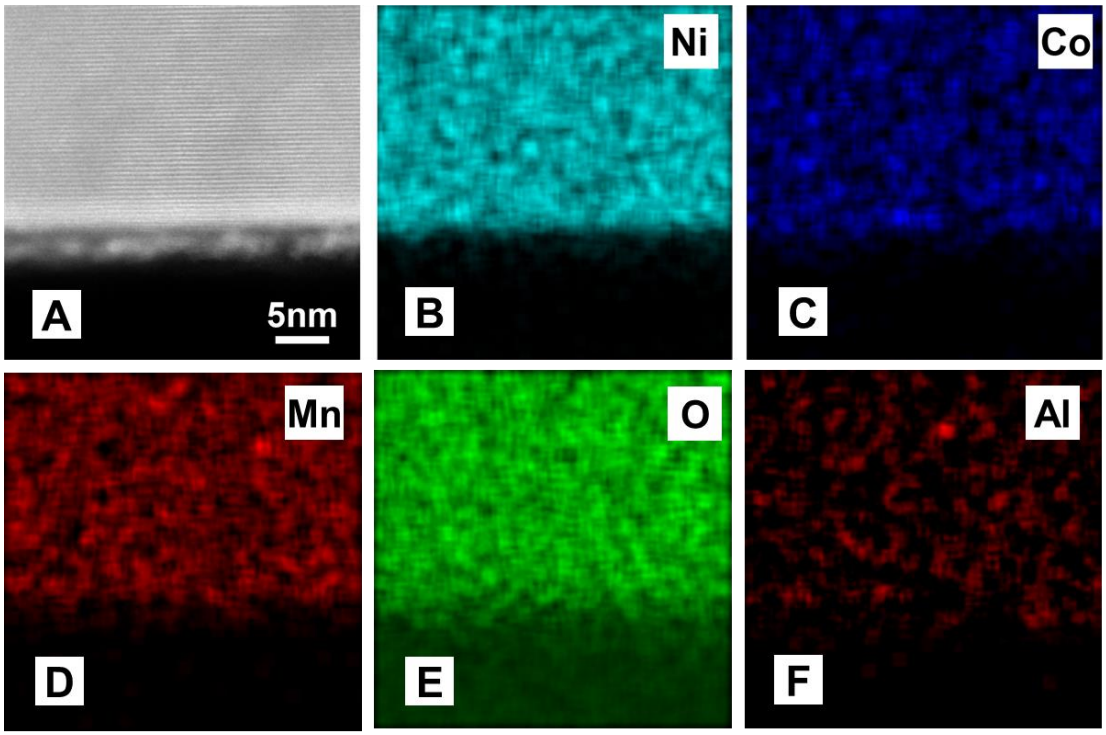

Figure S9. HAADF-EDS mapping of 1%Al-NMC76 after 500 cycles. (A) HAADF-STEM image. Corresponding EDS maps of Ni (B), Co (C), Mn (D), O (E), and Al (F).