Supplementary Information (SI) for Journal of Materials Chemistry A.

Supporting information

Achieving Dynamic Stability of Single-Crystal Low-Co Ni-Rich Cathode Material for High performance Lithium Batteries

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Table S1: Rietveld XRD refinement results of the pristine cathode materials.

Electrode material	***a* (Å)**	***c* (Å)**	**Rp (%)**	**Rwp (%)**	**Rexp (%)**	**Li/Ni cation mixing (%)**
Fresh-NMC	2.878	14.241	5.03	8.56	3.08	3.28
Fresh-NMFAC	2.865	14.289	4.99	7.89	3.36	2.96

Table S2: Metal-oxygen bond lengths for NMC and NMFAC obtained from the DFT calculation of this study.

NMC		**NMFAC**	
Bonds	a (Å)	b (Å)	c (Å)
Ni-O	-	Ni-O	1.980
Li-O	-	Li-O	2.016
Mn-O	1.986	Fe-O	2.012
Co-O	1.967	Al-O	1.95

Table S3. Chemical compositions of Ni, Mn, Fe, Al, and Co for NMC and NMFAC measured via the ICP-AES test.

Sample	**Chemical composition (at. %)**				
NMC	Ni	Mn	Co	-	-
	90.02	4.97	5.01	-	-
NMFAC	Ni	Mn	Fe	Al	Co
	90.01	5.02	2.04	1.95	0.98

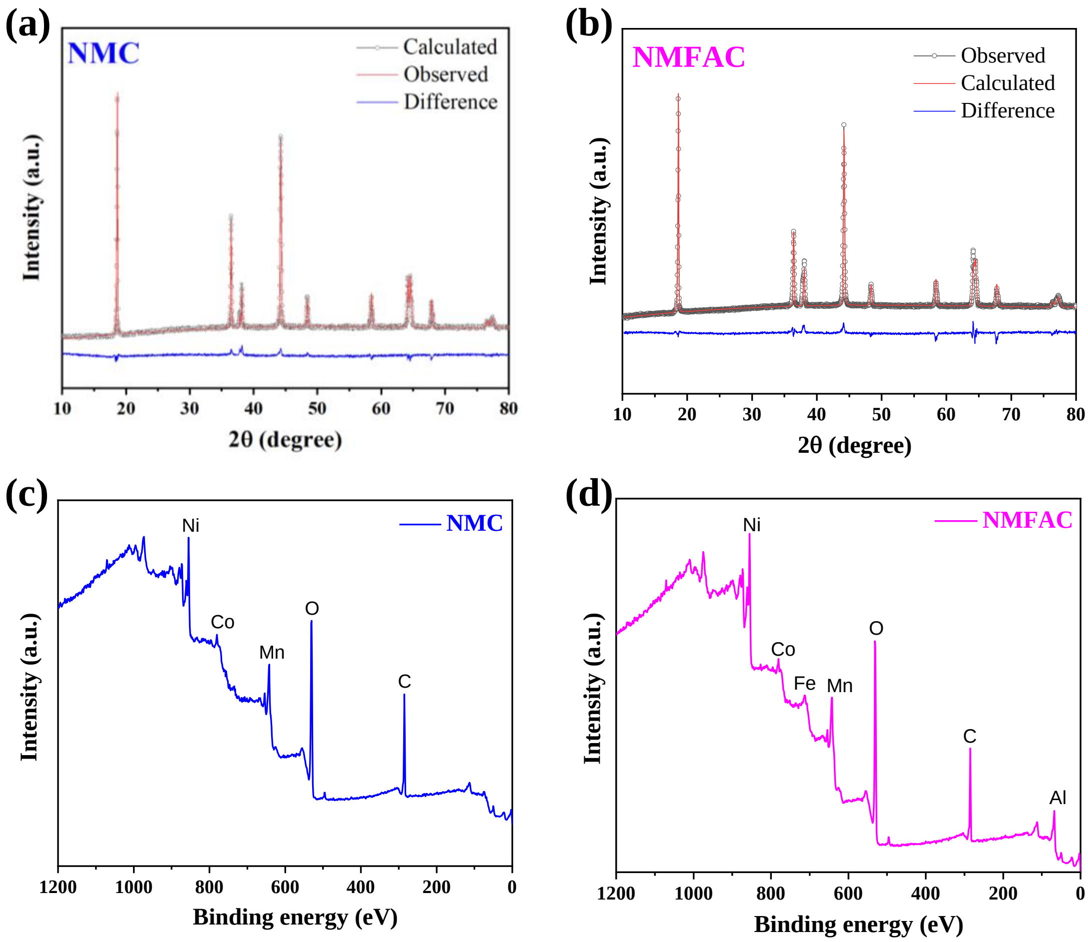

Figure S1: (a, b) Rietveld refinement results of the XRD patterns, and **(c, d)** XPS survey spectrum for NMC and NMFAC, respectively.

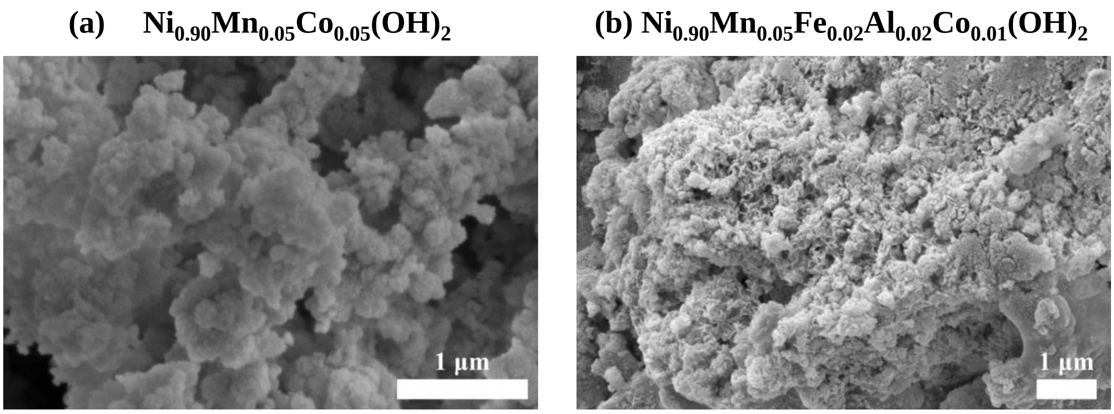

Figure S2: SEM images of (a) $Ni_{0.90}Mn_{0.05}Co_{0.05}(OH)_2$ and (b) $Ni_{0.90}Mn_{0.05}Fe_{0.02}Al_{0.02}Co_{0.01}(OH)_2$ precursors.

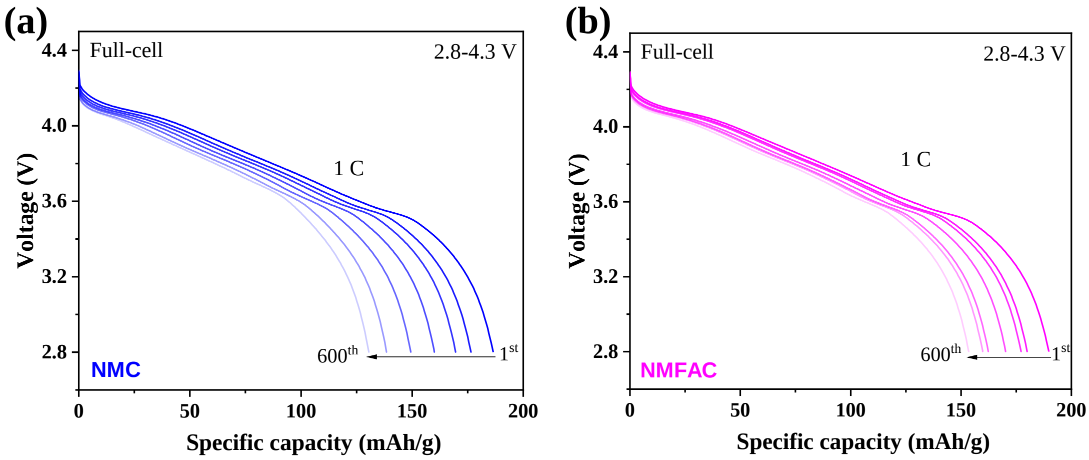

Figure S3: Discharge voltage profiles of **(a)** NMC and **(b)** NMFAC full cells with a graphite anode at 2.8 - 4.3 V.

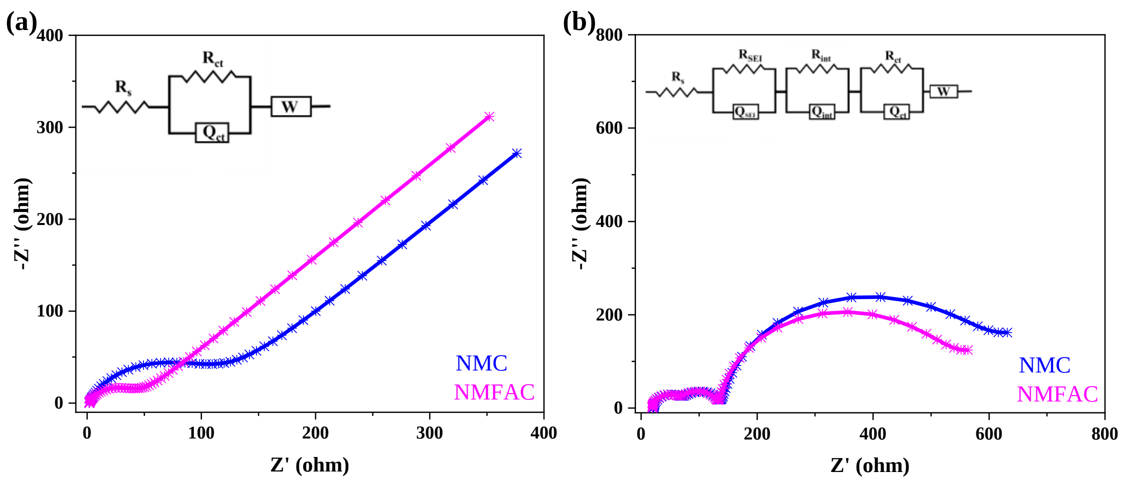

Figure S4: Nyquist plots of (a) before and (b) after 100 cycles for NMC and NMFAC cathode materials.

Table S4. EIS data before and after 100 cycles for NMC and NMFAC cathode materials.

Cathode		R_s(Ω)	R_{SEI}(Ω)	R_{int}(Ω)	R_{ct} (Ω)
NMC	Before cycle	1.69	-	-	109
	After 100 cycles	2.15	5.05	129	495
NMFAC	Before cycle	1.75	-	-	42
	After 100 cycles	1.93	4.80	126	431

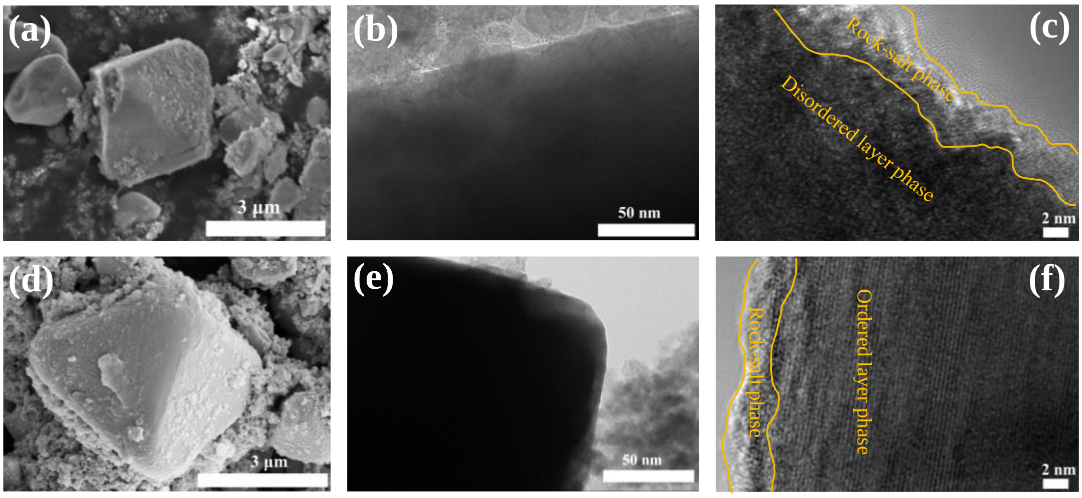

Figure S5: SEM and TEM images of **(a-c)** NMC and **(d-f)** NMFAC after 600 cycles.

Table S5: Electrochemical performance comparison of NMC and NMFAC with half-cell and full cell (our work).

Cathode	**Cycling Stability**					
	Initial Discharge capacity [mAh g^{-1}]	Voltage	No. of Cycles	Loading (mg/cm^2)	Rate (C)	Retention (%)
NMC	196	4.3 V vs. Li	100	~2.25	1C	90
NMFAC	198	4.3 V vs. Li	100	~2.25	1C	93
NMC	186	4.3 V vs. Gr	600	~2.22	1C	70
NMFAC	189	4.3 V vs. Gr	600	~2.22	1C	81

Table S6: Comparison of the specific capacity and cycling performance of SC NMC and NMFAC cathodes with the previously reported Ni-rich cathodes.

Cathode	Cycling Stability						Ref.
	Initial Discharge capacity [mAh g^{-1}]	Voltage	No. of Cycles	Loading (mg/cm^2)	Rate (C)	Retention (%)	
SCNMC811	185	4.3 V vs. Li	25	3	0.1C	~50%	[1]
SCNMC83	184	4.2 V vs. Gr/SiO	600	47	1C	84%	[2]
$LiNi_{0.89}Mn_{0.055}$-$Co_{0.055}O_2$	226	4.4 V vs. Li	100	-	0.1C	91%	[3]
$LiNi_{0.883}Mn_{0.056}$-$Al_{0.061}O_2$	216	4.4 V vs. Li	100	2.5	1/3C	90%	[3]
$LiNi_{0.89}Mn_{0.044}Co_{0.042}$-$Al_{0.013}Mg_{0.011}O_2$	213	4.4 V vs. Li	100	2.5	1/3C	93%	[3]
$LiNi_{0.883}Co_{0.053}$-$Al_{0.064}O_2$	220	4.4 V vs. Li	100	2.5	1/3C	88%	[3]
$LiNi_xFe_yAl_zO_2$	180	4.5 V vs. Li	100	5	0.3C	~70%	[4]
$LiNi_{0.95}Mg_{0.05}O_2$	200	4.3 V vs. Li	100	10-12	0.05/ 0.2C	~90%	[5]
$LiNi_{0.95}Al_{0.05}O_2$	220	4.3 V vs. Li	100	10-12	0.05/ 0.2C	~86%	[5]
$LiNi_{0.93}Al_{0.05}Ti_{0.01}$-$Mg_{0.01}O_2$	221	4.25 V vs. Gr	800	2.0	0.5C	52%	[6]
$LiNi_{0.96}Mg_{0.02}Ti_{0.02}O_2$	180	4.4 V vs. Gr	300	1.4	1C	85%	[7]
$LiNi_{0.8}Co_{0.1}Mn_{0.09}$-$Cu_{0.01}O_2$	175	4.3 V vs. Li	100	-	0.5C	94%	[8]
NMC	196	4.3 V vs. Li	100	~2.25	1C	90	**Our Work**
NMFAC	198	4.3 V vs. Li	100	~2.25	1C	93	

Table S7: Comparison of the previously reported co-dopped Ni-rich cathodes with our work.

Cathode	Doping elements	Synthesis technique	Initial charge capacity [mAh g^{-1}]	Rate (C)	Ref.
$LiNi_{0.905}Co_{0.04}Mn_{0.04}$-$Al_{0.005}Nb_{0.01}O_2$	Al/Nb	Co-precipitation	230	0.1	[9]
$LiNi_{0.890}Mn_{0.044}Co_{0.042}Al_{0.013}Mg_{0.011}O_2$	Al/Mg	Co-precipitation	213	0.1	[3]
$LiNi_{0.598}Co_{0.08}Mn_{0.3}Zr_{0.002}$-$Ti_{0.002}O_2$	Zr/Ti	Co-precipitation	~180	0.3	[10]
SC-NMC	Ce/Gd	Solid State	211	0.1	[11]
$LiNiO_2$	Mg/Al	Interdiffusion strategy	252	0.1	[12]
NMFAC	Fe/Al	Co-precipitation	248	0.2	**Our Work**

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