Supporting Information

Influences of Al-doping on the structure and electrochemical performance of Co-free LiNi$_{0.8}$Mn$_{0.2}$O$_2$ cathode material

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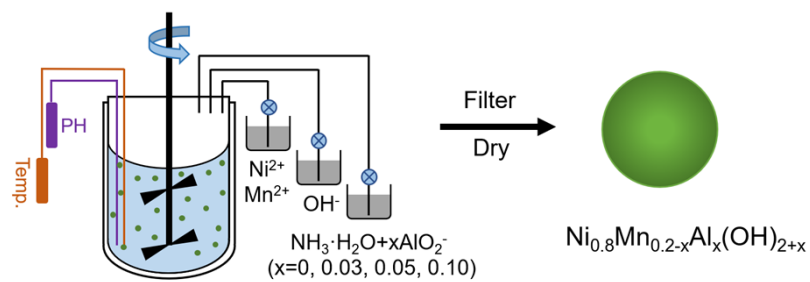
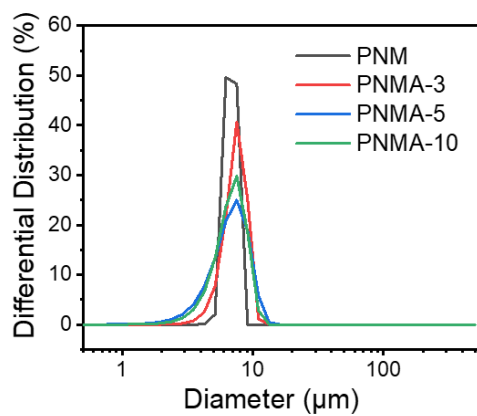**Fig. S1** Schematic diagram of $Ni_{0.8}Mn_{0.2-x}Al_x(OH)_{2+x}$ precursor preparation.

Fig. S2 Particle size distribution curves of PNM, PNMA-3, PNMA-5 and PNMA-10.
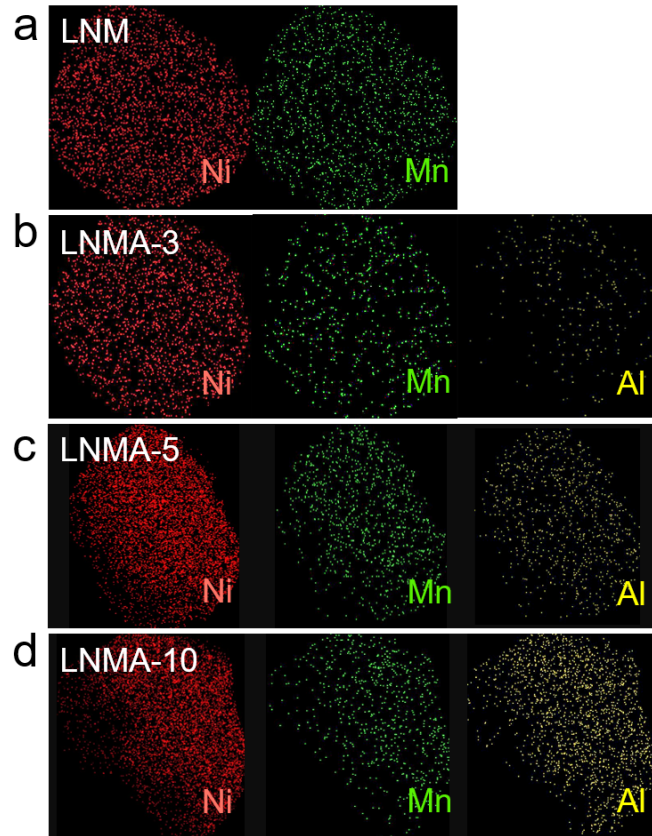
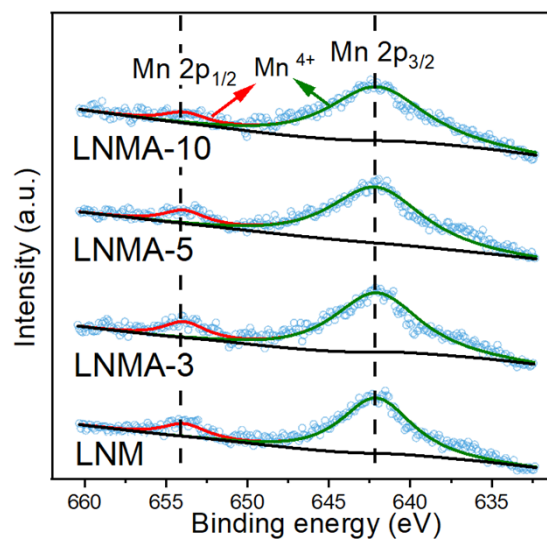**Fig.S3** EDS Mapping of (a) LNM, (b) LNMA-3, (c) LNMA-5 and (d) LNMA-10.

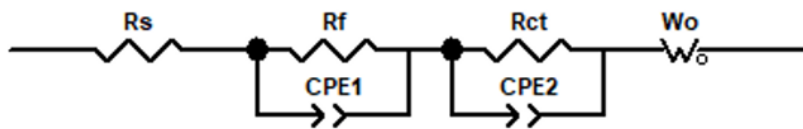**Fig. S4** XPS spectra of manganese.

Fig. S5 The equivalent circuit used to simulate the EIS data.

Fig. S6 XRD patterns comparation of four electrodes before and after 300 cycles.

Table S1 ICP chemical elemental compositions of LNM, LNMA-3, LNMA-5 and LNMA-10 materials.

Sample	Ni	Mn	Al
LNM	0.799	0.202	-
LNMA-3	0.798	0.173	0.029
LNMA-5	0.798	0.147	0.052
LNMA-10	0.799	0.100	0.099

Table S2 The refined lattice parameters of LNM, LNMA-3, LNMA-5 and LNMA-10 samples.

Samples	a=b (Å)	c (Å)	c/a	Li^+/Ni^{2+} (%)	(003)/(104) Observed	(003)/(104) Fitted
LNM	2.8787	14.2341	4.9446	7.19	1.072	1.002
LNMA-3	2.8752	14.2368	4.9516	6.08	1.169	1.184
LNMA-5	2.8737	14.2390	4.9549	5.84	1.180	1.233
LNMA-10	2.8695	14.2419	4.9632	5.32	1.276	1.354

Table S3 Refined lattice parameters of LNM.

Name	Fractional coordinates			Occupancy	Uiso
	x	y	z		
Li	0	0	0	0.9281	0.03970
Li (in Ni layer)	0	0	0.5	0.0719	0.03274
Ni	0	0	0.5	0.7281	0.01018
Ni (in Li layer)	0	0	0	0.0719	0.03403
Mn	0	0	0.5	0.2000	0.03911
O	0	0	0.2440	1.0000	0.03316

Table S4 Refined lattice parameters of LNMA-3.

Name	Fractional coordinates			Occupancy	Uiso
	x	y	z		
Li	0	0	0	0.9392	0.05211
Li (in Ni layer)	0	0	0.5	0.0608	0.04734
Ni	0	0	0.5	0.7392	0.01690
Ni (in Li layer)	0	0	0	0.0608	0.04734
Mn	0	0	0.5	0.1700	0.02804
Al	0	0	0.5	0.0300	0.03490
O	0	0	0.2441	1.0000	0.03645

Table S5 Refined lattice parameters of LNMA-5.

Name	Fractional coordinates			Occupancy	Uiso
	x	y	z		
Li	0	0	0	0.9416	0.04363
Li (in Ni layer)	0	0	0.5	0.0584	0.03734
Ni	0	0	0.5	0.7416	0.01211
Ni (in Li layer)	0	0	0	0.0584	0.02450
Mn	0	0	0.5	0.1500	0.02453
Al	0	0	0.5	0.0500	0.03096
O	0	0	0.2438	1.0000	0.03221

Table S6 Refined lattice parameters of LNMA-10.

Name	Fractional coordinates			Occupancy	Uiso
	x	y	z		
Li	0	0	0	0.9468	0.04788
Li (in Ni layer)	0	0	0.5	0.0532	0.03479
Ni	0	0	0.5	0.7468	0.00753
Ni (in Li layer)	0	0	0	0.0532	0.04479
Mn	0	0	0.5	0.1000	0.03264
Al	0	0	0.5	0.1000	0.04264
O	0	0	0.2438	1.0000	0.02803

Table S7 Refined lattice parameters of LNM, LNMA-3, LNMA-5 and LNMA-10.

Average bond length	LNM	LNMA-3	LNMA-5	LNMA-10
Li-O	2.0851 Å	2.0921 Å	2.0958 Å	2.0964 Å
(Ni/Co/Mn/Al)-O	1.9914 Å	1.9939 Å	1.9858 Å	1.9869 Å

Table S8 Half-cell performance of Ni-rich Co-free layered oxides cathodes.

Cathode material	Capacity (mAh g^{-1} at 0.1 C)	Cycling performance (capacity retention at 100 cycles)	Cycling voltage (V)	Ref.
$LiNi_{0.8}Mn_{0.2}O_2$	4.3 V, 201.7	88.3%	4.3 V	[1]
$LiNi_{0.85}Fe_{0.052}Al_{0.091}O_2$	4.3V, 160.0	88.0%	4.3 V	[2]
$LiNi_{0.8}Mn_{0.2}O_2$	4.5 V, 210.0	86.2%	4.5 V	[3]
$LiNi_{0.9}Mn_{0.05}Al_{0.05}O_2$	4.6 V, 223.7	79.6%	4.6 V	[4]
$LiNi_{0.8}Mn_{0.13}Ti_{0.02}Mg_{0.02}Nb_{0.01}Mo_{0.02}O_2$	4.5 V, 210.1	98.9%	4.3 V	[5]
$LiNi_{0.9}Mn_{0.1}O_2$	4.4 V, 232.4	91.5%	4.4 V	[6]
$LiNi_{0.8}Mn_{0.15}Al_{0.05}O_2$	4.3 V, 183.1	96.2%	4.3 V	This work

Table S9 Electrochemical impedance spectrum values of the LNM, LNMA-3, LNMA-5 and LNMA-10 cathodes after one cycle and 30 cycles.

	1st			30th		
	R_s (Ω)	R_f (Ω)	R_{ct} (Ω)	R_s (Ω)	R_f (Ω)	R_{ct} (Ω)
LNM	2.082	4.100	10.630	4.526	6.947	21.760
LNMA-3	1.964	4.651	6.876	3.174	8.877	15.464
LNMA-5	1.686	3.680	6.068	4.467	5.086	11.553
LNMA-10	1.637	4.421	9.266	4.712	4.888	14.122

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