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## Supporting information

# Synergistic aluminum lattice doping and surface coating for high-performance Co-free Ni-rich cathodes

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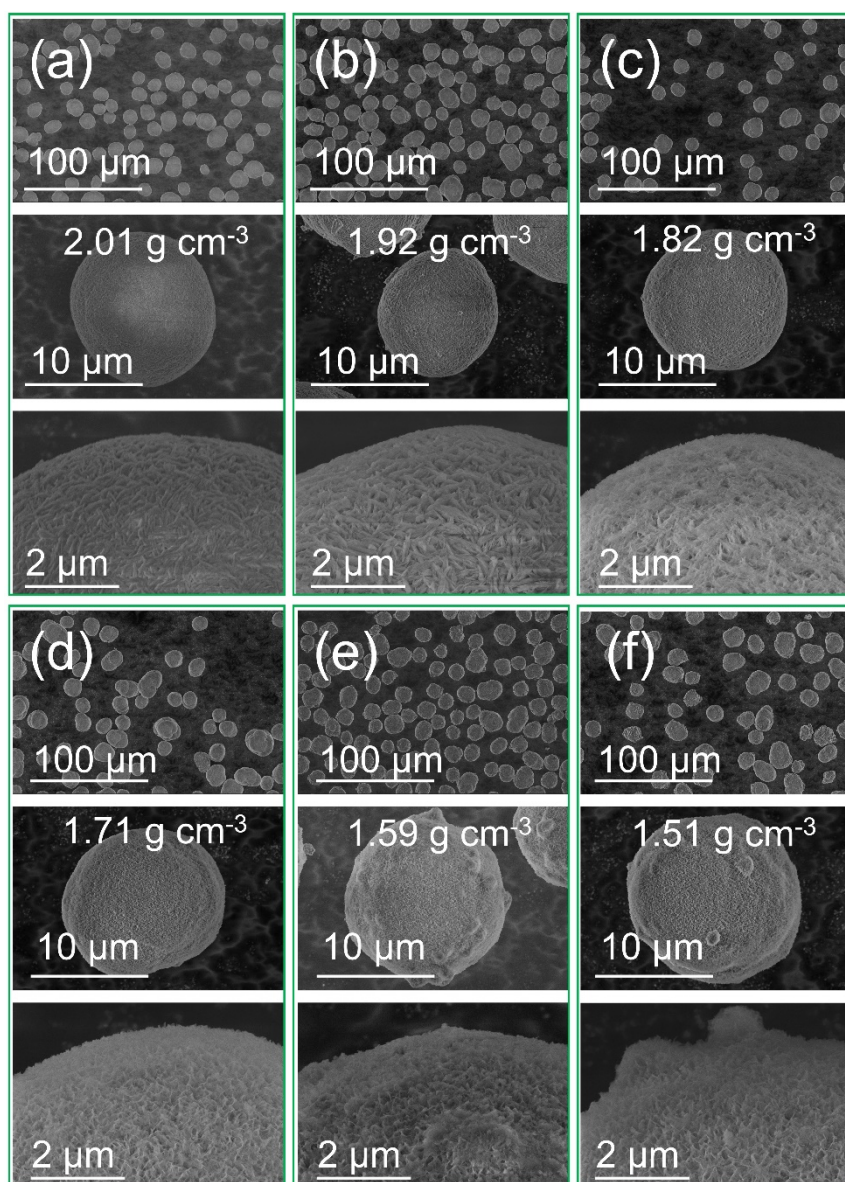
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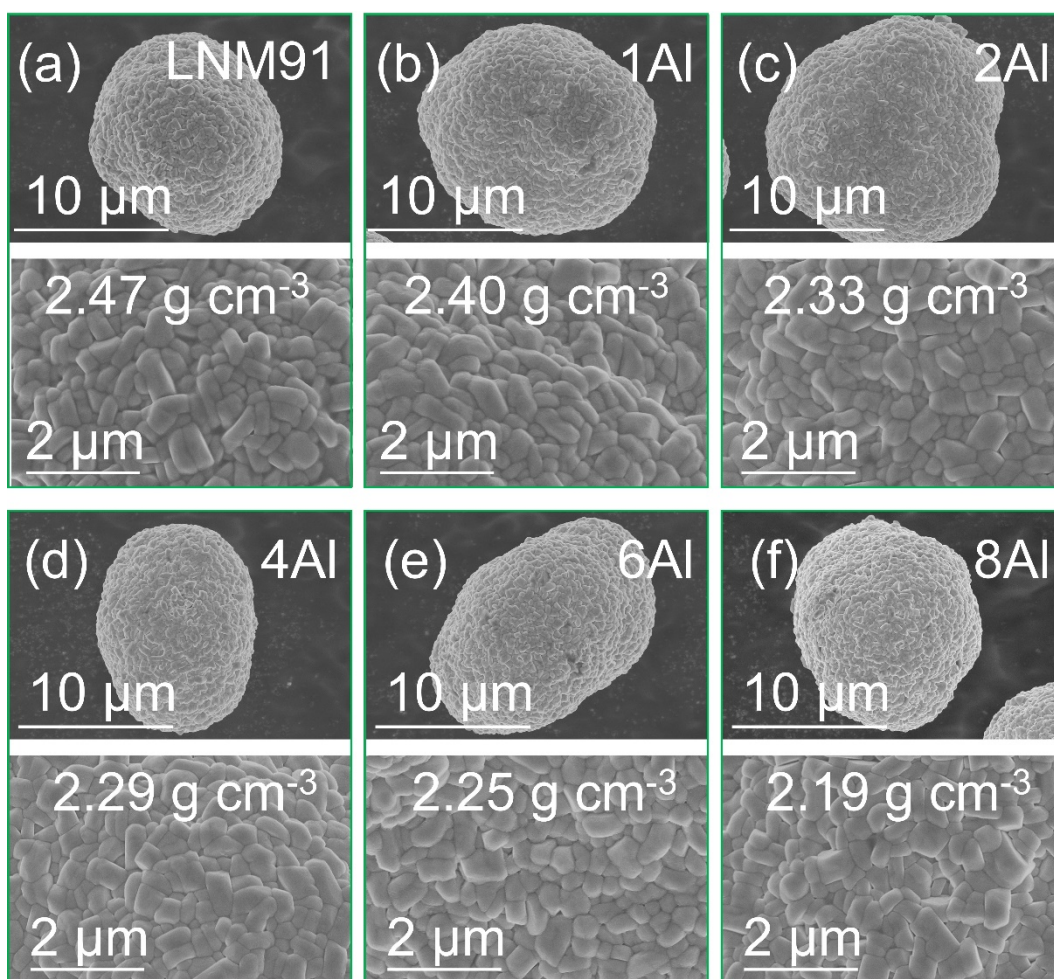
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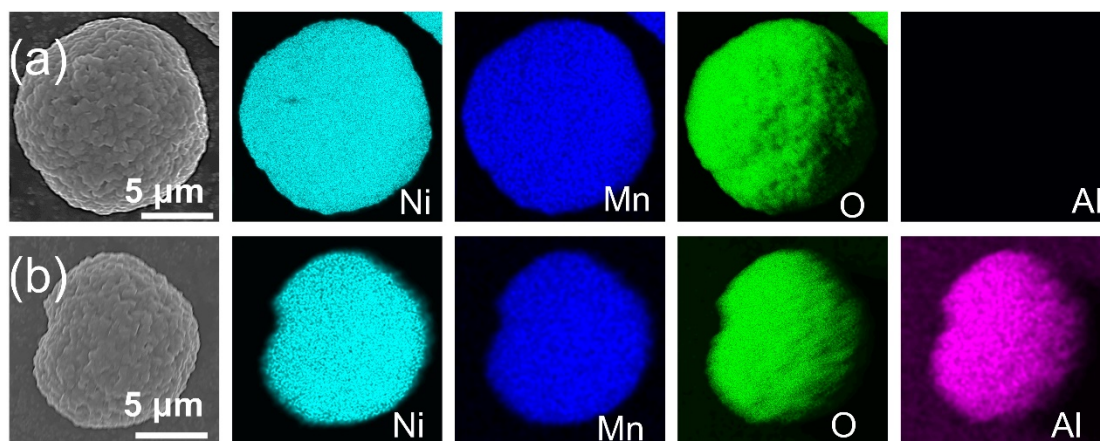
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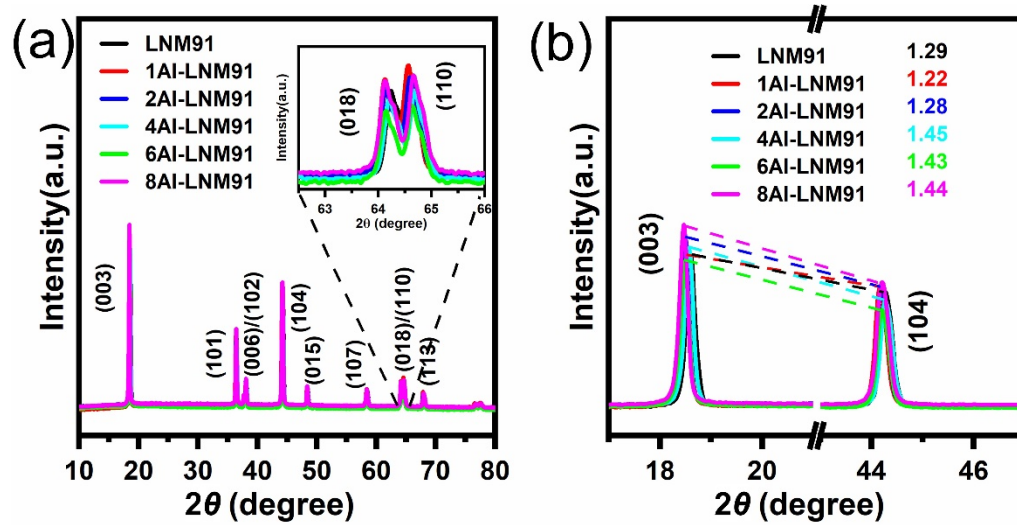
**Fig. S1.** SEM images of the pristine and Al modified Ni<sub>0.9</sub>Mn<sub>0.1</sub>(OH)<sub>2</sub> precursor with varied modification amounts: (a) Ni<sub>0.9</sub>Mn<sub>0.1</sub>(OH)<sub>2</sub>, (b) 1 mol%, (c) 2 mol%, (d) 4 mol%, (e) 6 mol%, (f) 8 mol%.



**Fig. S2.** SEM images of (a) LNM91, (b) 1Al-LNM91, (c) 2Al-LNM91, (d) 4Al-LNM91, (e) 6Al-LNM91 and (f) 8Al-LNM91 cathode samples calcined at 750 °C under O<sub>2</sub> for 15h.



**Fig. S3.** EDS mapping of (a) LNM91 and (b) 6Al-LNM91



**Fig. S4.** (a) XRD patterns of all cathodes, (b) magnified XRD spectra to highlight the intensity ratio of (003)/(104).

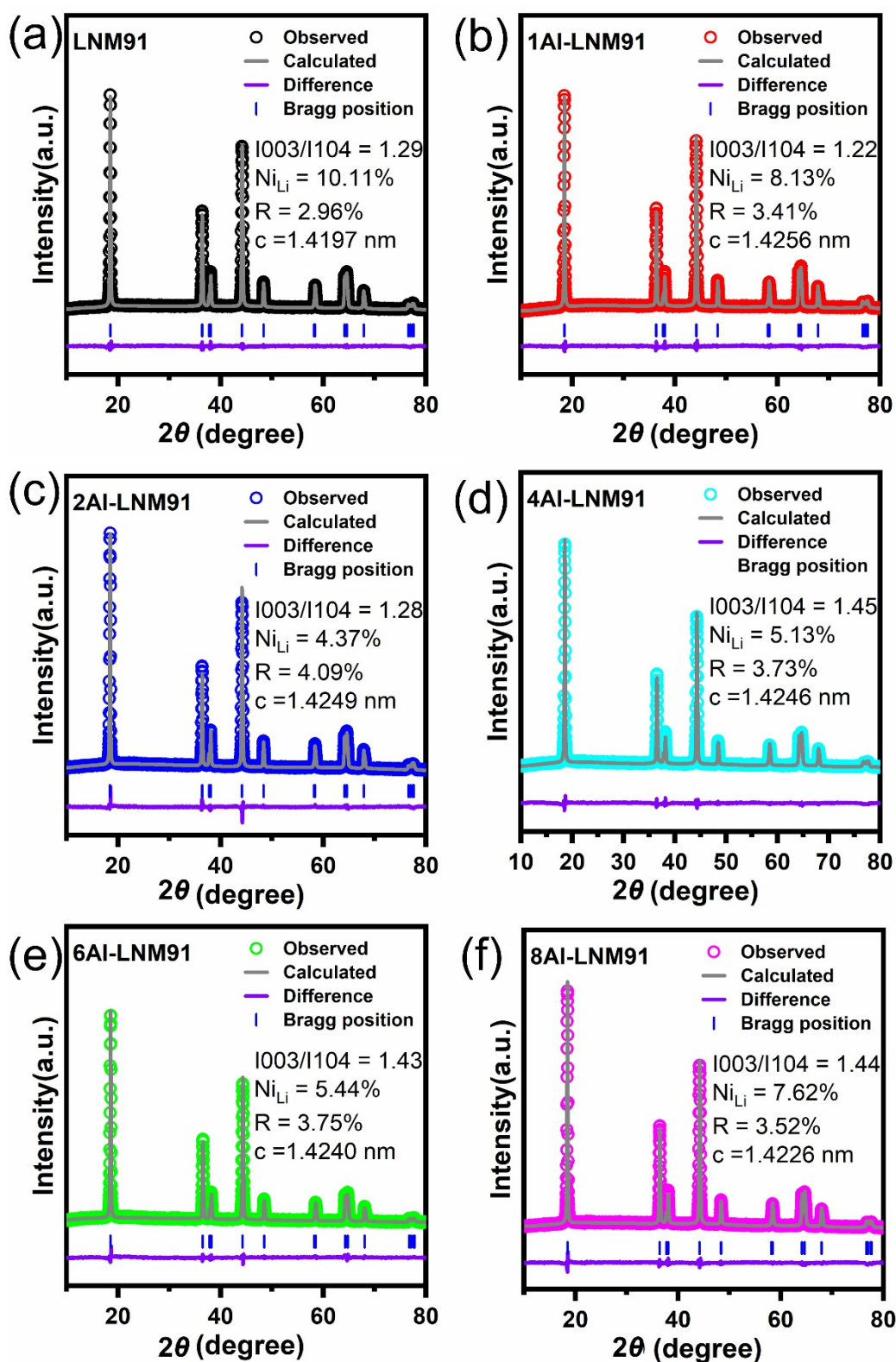
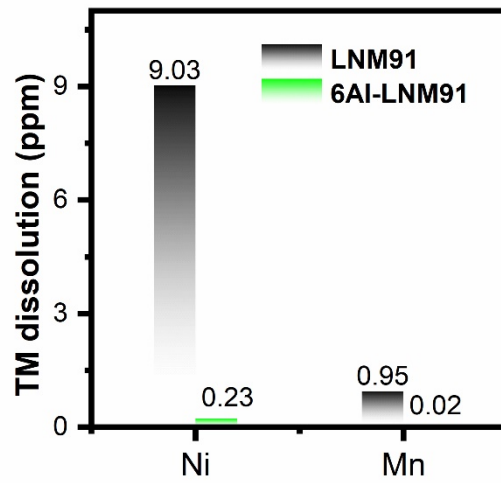
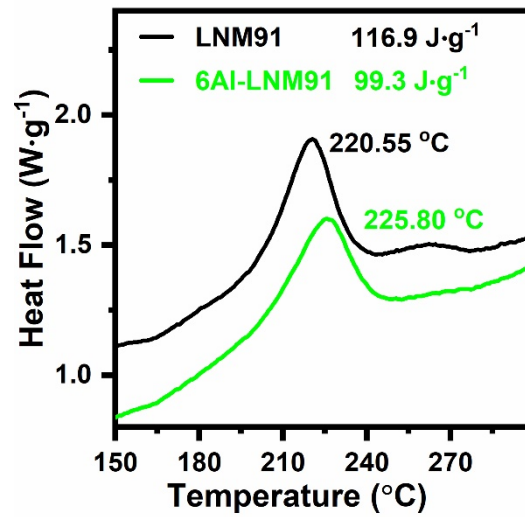


Fig. S5. XRD of (a) LNM91, (b) 1Al-LNM91, (c) 2Al-LNM91, (d) 4Al-LNM91, (e) 6Al-LNM91, (f) 8Al-LNM91.



**Fig. S6.** Transition metal ion dissolution curve LNM91 and 6Al-LNM91 after 120 cycles at 0.5 C.



**Fig. S7.** DSC curves of the LNM91 and 6Al-LNM91 electrodes after charging to 4.4V.

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**Table S1.** Chemical compositions of the precursor, pristine and Al-modified  $\text{LiNi}_{0.9}\text{Mn}_{0.1}\text{O}_2$  cathodes with varied modification amounts using ICP-AES.

Materials	Ni(at%)	Mn(at%)	Al(at%)
NM91	89.46	10.54	--
LN91	89.31	10.69	--
1Al-LNM91	88.20	10.81	0.99
2Al-LNM91	87.31	10.75	1.95
4Al-LNM91	85.72	10.36	3.93
6Al-LNM91	83.97	10.24	5.78
8Al-LNM91	82.67	10.07	7.26

**Table S2.** Comparison of cycling stability of Al-modified LNM91 cathode with previously Ni-rich cathodes.

Materials	Temperature	Voltage range	Current density	Capacity retention in half-cell (%)	References
6Al-LNM91	21±3°C	2.8-4.4V	0.5C	94.03% after 120 cycles	This work
Mo <sup>4+</sup> doping and Li <sub>2</sub> MoO <sub>4</sub> coating LiNi <sub>0.9</sub> Mn <sub>0.1</sub> O <sub>2</sub>	25°C	2.7-4.3V	0.2C	75.6% after 100 cycles	[S1]
Sb <sup>5+</sup> doping and Li <sub>3</sub> SbO <sub>4</sub> coating LiNi <sub>0.9</sub> Mn <sub>0.1</sub> O <sub>2</sub>	25°C	3.0-4.3V	0.33C	90.3% after 100 cycles	[S2]
Al <sup>3+</sup> doping and LiAlO <sub>2</sub> coating LiNi <sub>0.8</sub> Co <sub>0.1</sub> Mn <sub>0.1</sub> O <sub>2</sub>	25°C	2.7-4.3V	1C	92.1% after 100 cycles	[S3]
Ti <sup>4+</sup> doping and Li <sub>3</sub> PO <sub>4</sub> coating LiNi <sub>0.9</sub> Co <sub>0.09</sub> Mo <sub>0.01</sub> O <sub>2</sub>	--	2.7-4.3V	0.5C	93.7% after 100 cycles	[S4]
Al and Ti doping LiNi <sub>0.9</sub> Mn <sub>0.1</sub> O <sub>2</sub>	25°C	2.75-4.35V	0.5C	89.77% after 100 cycles	[S5]
Al and Ti doping LiNi <sub>0.9</sub> Mn <sub>0.1</sub> O <sub>2</sub>	25°C	2.75-4.5V	0.5C	80.32% after 100 cycles	[S5]
Ti and Zr doping LiNi <sub>0.9</sub> Mn <sub>0.1</sub> O <sub>2</sub>	--	3.0-4.3V	0.5C	91.2% after 100 cycles	[S6]
Li <sub>2</sub> MoO <sub>4</sub> coating single crystal LiNi <sub>0.9</sub> Mn <sub>0.1</sub> O <sub>2</sub>	25°C	2.7-4.3V	0.2C	80.8% after 100 cycles	[S7]
B/Al doping and LiBO <sub>2</sub> /LiAlO <sub>2</sub> coating LiNi <sub>0.9</sub> Mn <sub>0.1</sub> O <sub>2</sub>	25°C	2.7-4.3V	1C	86.59% after 100 cycles	[S8]

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**Table S3.** Impedance fitting parameters for LNM91 and 6Al-LNM91.

Sample	Before cycling		After cycling		
	$R_s$	$R_{ct}+R_f$	$R_s$	$R_f$	$R_{ct}$
LNM91	2.87	10.99	6.21	6.59	25.9
1Al+1Mg-LNM91	2.21	6.31	4.95	5.46	8.46

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