Cobalt-free Lithium-rich Layered Oxide Cathodes coated with LiPON to improve the Battery cycle performance
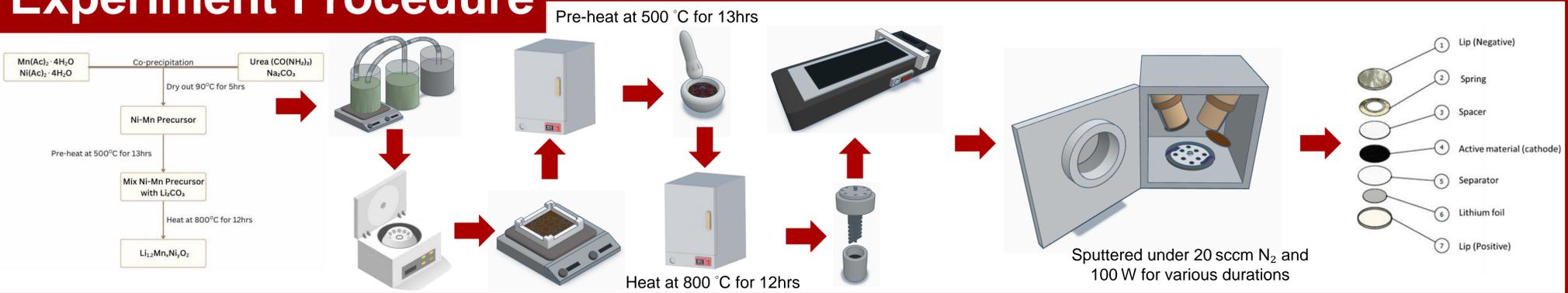
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Abstract

$Li_{1.2}Ni_xMn_{0.8-x}O_2$ (LMNO) is a high-capacity layered cathode material but suffers from surface instability during cycling. In this study, LiPON was deposited as a protective layer, leading to improved initial discharge capacity and enhanced retention after 50 cycles. XPS and Raman confirmed the formation of P–O and P–N bonds, while SEM revealed reduced surface damage. The LiPON layer effectively stabilized the LMNO interface and improved electrochemical performance.

Experiment Procedure

Pre-heat at 500 °C for 13hrs

Heat at 800 °C for 12hrs

Sputtered under 20 sccm N_2 and 100 W for various durations
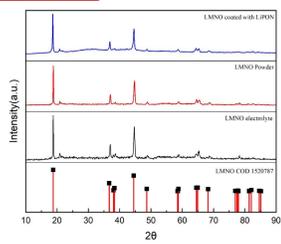
Result & Discussion

XRD

Fig. 1. XRD patterns of pristine LMNO and LMNO with LiPON layer

RAMAN
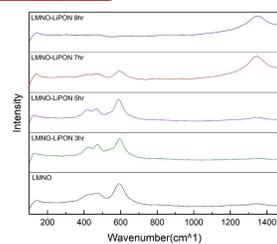
Fig. 2. Raman spectra of pristine LMNO and LMNO with LiPON layers of different deposition durations

SEM

Fig. 3. SEM images of (a) pristine LMNO, (b) LMNO with LiPON layer, (c) cycled pristine LMNO, and (d) cycled LMNO with LiPON layer

Fig. 4. SEM images of (a) pristine LMNO, (b) LMNO with 3 h LiPON layer, (c) LMNO with 7 h LiPON layer, and (d) LMNO with 9 h LiPON layer
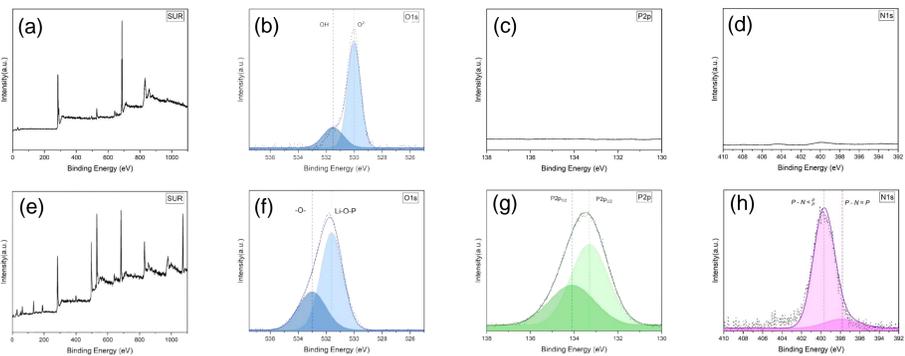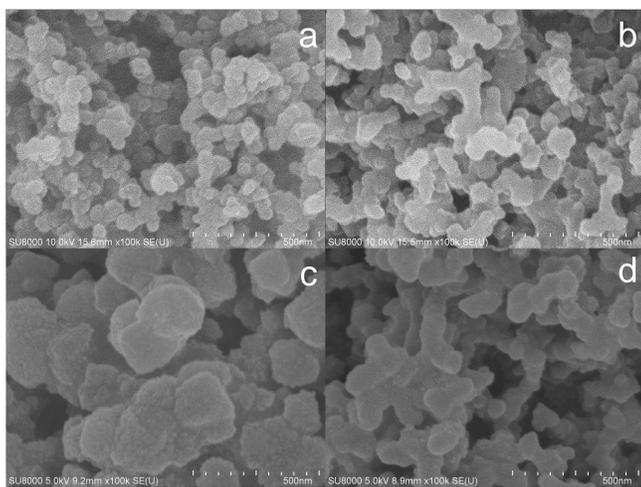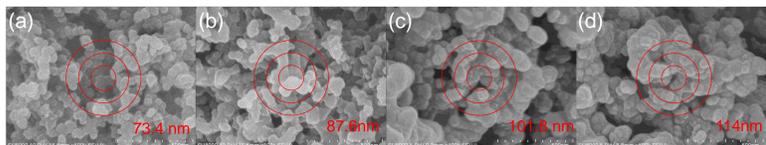
XPS

Fig.5 (a) XPS survey spectrum of the LMNO; (b) O1s XPS patterns of the LMNO; (c) P2p XPS patterns of the LMNO; (d) N1s XPS patterns of the LMNO; (e) XPS survey spectrum of the LMNO with LiPON layer; (f) O1s XPS patterns of the LMNO with LiPON layer; (g) P2p XPS patterns of the LMNO with LiPON layer; (h) N1s XPS patterns of the LMNO with LiPON layer

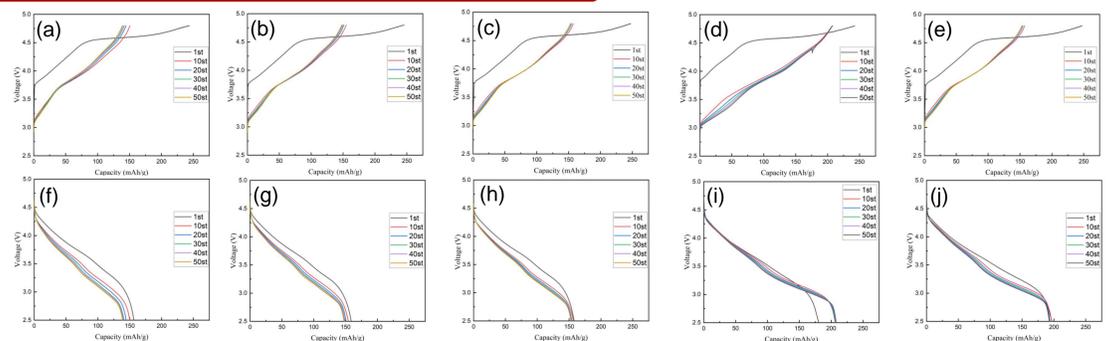### ELECTROCHEMICAL PERFORMANCE

Fig.6 Charge curves of (a) pristine LMNO and (b) LMNO covers with 3hr LiPON layer and (c) LMNO covers with 5hr LiPON layer and (d) LMNO covers with 7hr LiPON layer and (e) LMNO covers with 9hr LiPON layer and discharge curves of (f) pristine LMNO and (g) LMNO covers with 3hr LiPON layer and (h) LMNO covers with 5hr LiPON layer and (i) LMNO covers with 7hr LiPON layer and (j) LMNO covers with 9hr LiPON layer

(a)	Sputter (min)	Delithiation	Lithiation	Efficiency(%)	(b)	10th Delithiation / Lithiation	50th Delithiation / Lithiation	Delithiation Retention(%)
LMNO		243.5	156.4	64.23	P-LMNO	150.7 / 150	138.8 / 138.8	92.10
LMNO with LiPON layer	180	245.6	158.8	64.66	LMNO-LiPON 3hr	155.1 / 154.4	148.1 / 147.6	95.49
	300	247.3	157.7	63.77	LMNO-LiPON 5hr	150.7 / 151	138.8 / 138.9	98.88
	420	242.4	180.6	74.50	LMNO-LiPON 7hr	207.3 / 206.4	207.5 / 207.9	100.10
	540	267.3	197	73.70	LMNO-LiPON 9hr	197.2 / 196.3	193.6 / 193	98.17

Table.1 Electrochemical performance (a) pristine LMNO vs. with LiPON layers of different deposition durations' first lap Delithiation/Lithiation comparison (b) pristine LMNO vs. with LiPON layers of different deposition durations' 10th & 50th lap Delithiation/Lithiation comparison

Conclusion

- Structural and surface analyses (Raman, XPS) confirmed that the layer preserved the layered structure while introducing characteristic P–O and P–N bonding environments.
- Electrochemical tests demonstrated that LMNO electrodes with LiPON layer exhibited improved cycling performance, with capacity retention enhanced from 92% to 100% after 50 cycles.
- LiPON layer acted as an effective artificial CEI layer, suppressing electrolyte side reactions and enhancing the interfacial stability of LMNO cathodes under high-voltage operation.