

## Supporting Information

### **A Lattice-matched Interface between In-situ/Artificial SEIs Inhibiting SEI Decomposition for Enhanced Lithium Storage**

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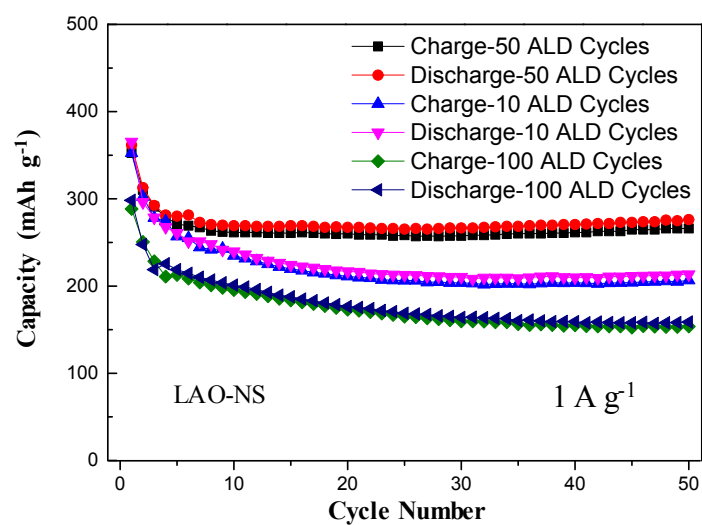
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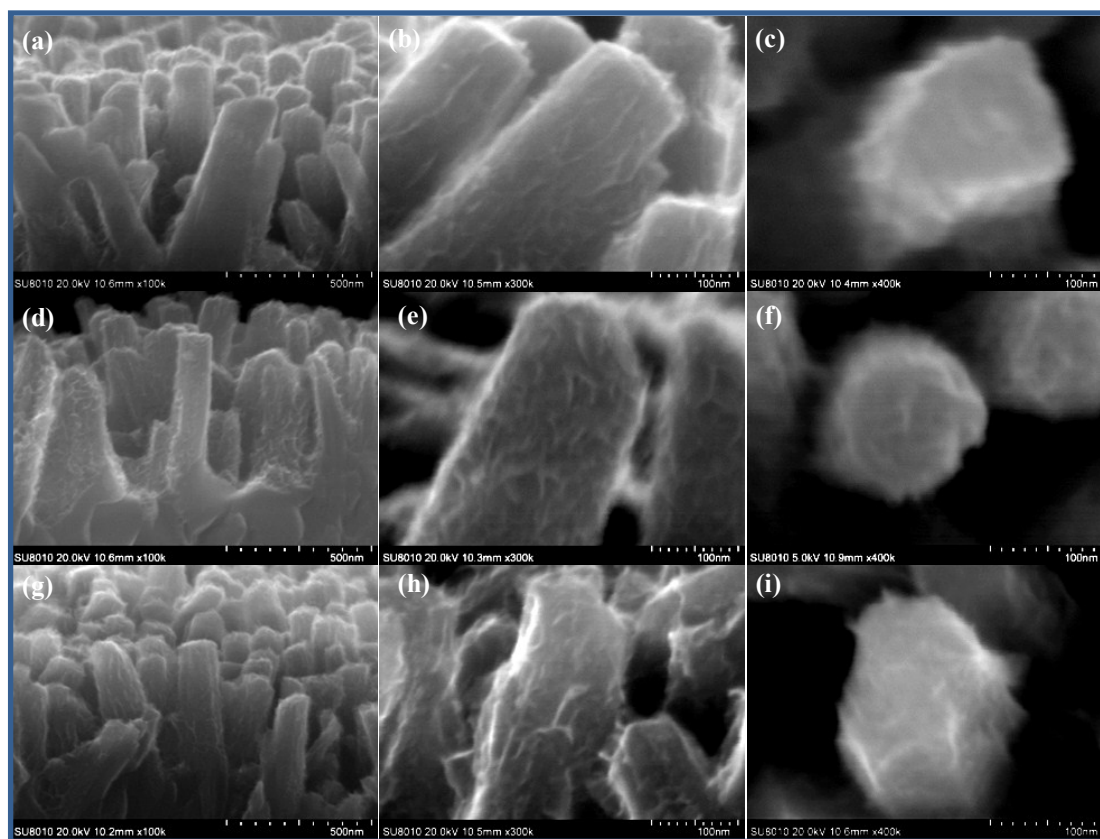
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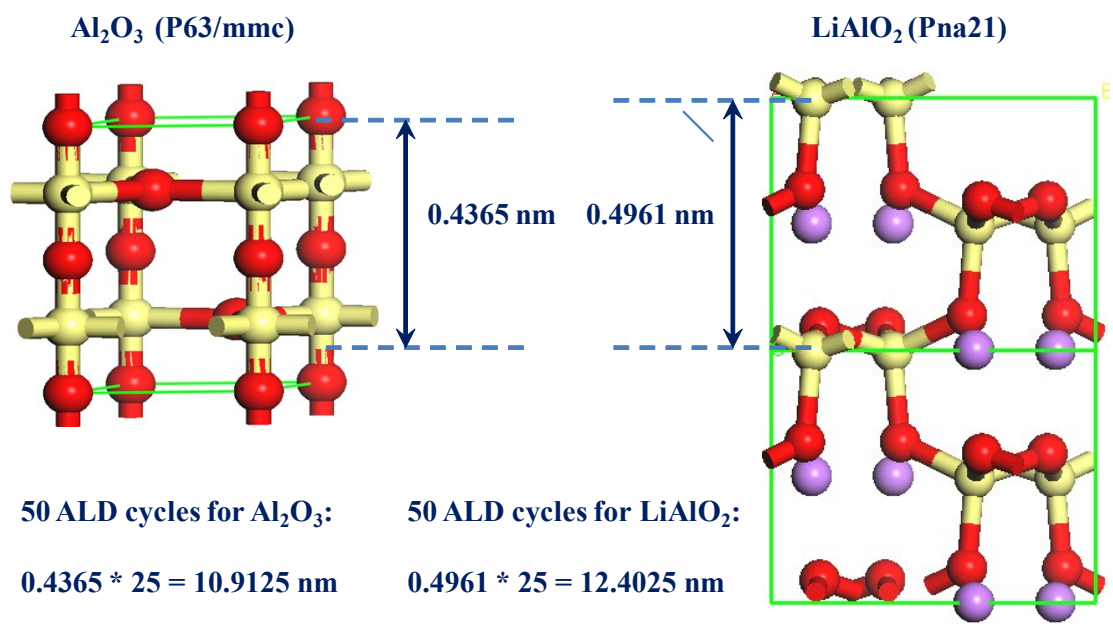
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**Figure S1** Cycling performance at 1 A g<sup>-1</sup> of LAO-NS for 10, 50, 100 ALD cycles, respectively.



**Figure S2** Morphologies of (a, b, and c) NS, (d, e, and f) AO-NS, (g, h, and i) LAO-NS. (a, d, and g): side view, (b, e, and h): magnified side view, and (c, f, and i): top view of (NS, AO-NS, and LAO-NS), respectively.



**Figure S3** Theoretical thickness calculation of Al<sub>2</sub>O<sub>3</sub> with P63/mmc and LiAlO<sub>2</sub> with Pna21 for 50 ALD cycles.

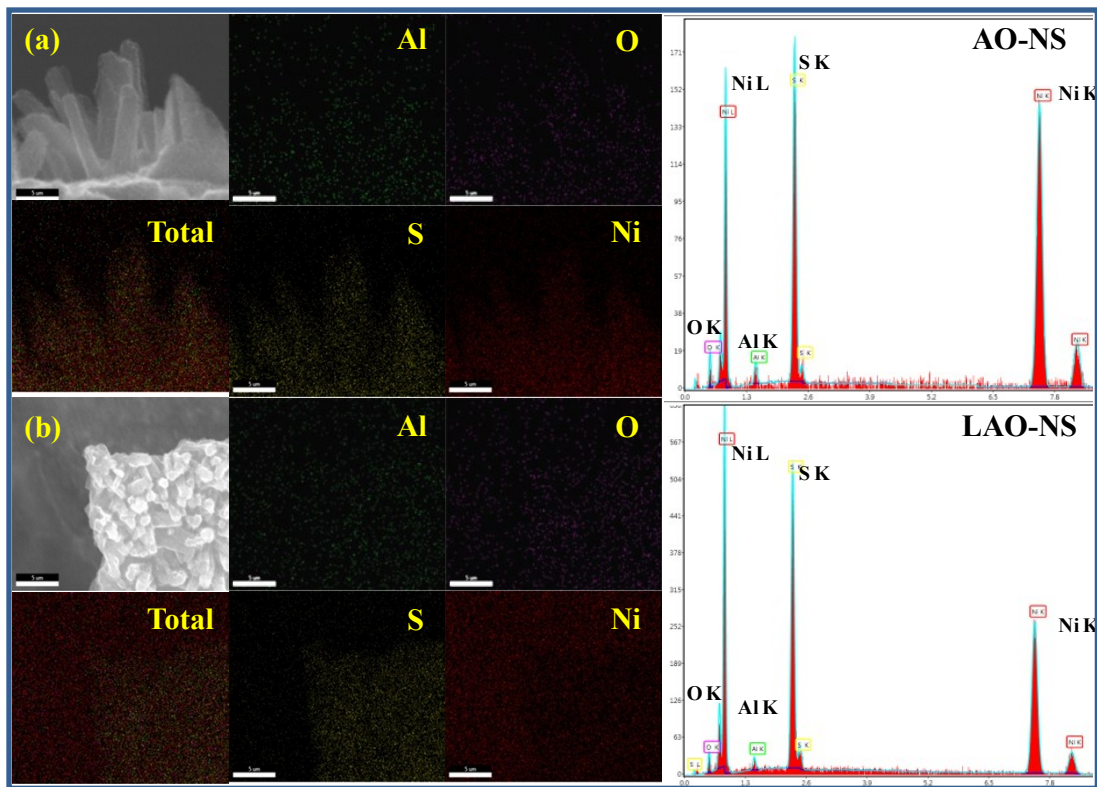
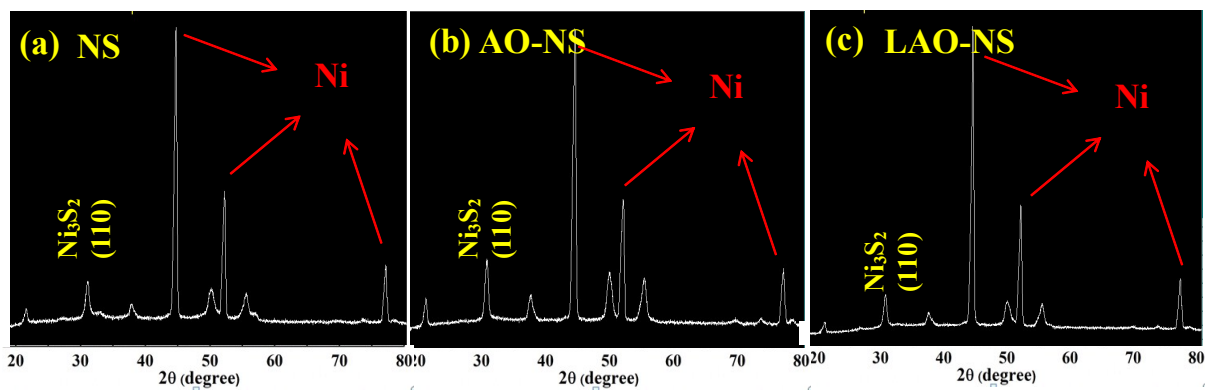


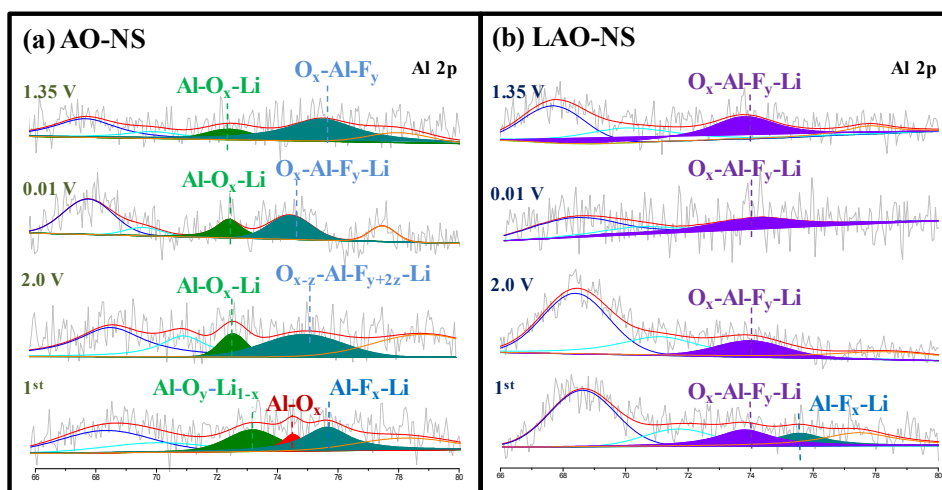
Figure S4 EDS analysis of (a) AO-NS and (b) LAO-NS.



**Figure S5** XRD patterns of (a) NS, (b) AO-NS, and (c) LAO-NS, referring to heazlewoodite  $\text{Ni}_3\text{S}_2$ .

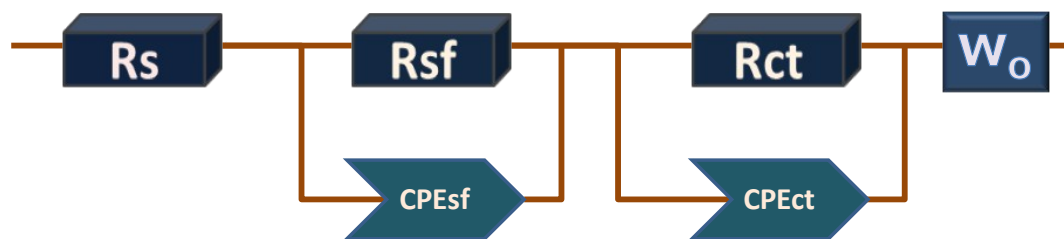
**Table S1.** Comparison of the capacity retention (refer to the capacity of the 2<sup>nd</sup> cycle) of ours LAO-NS to other reported nickel sulfide based electrodes for LIBs.

<b>Capacity retention /%</b>	<b>Ours</b>								
	<b>LAO-NS</b>	<b>Ref. 28</b>	<b>Ref. 30a</b>	<b>Ref. 30b</b>	<b>Ref. 30c</b>	<b>Ref. 30d</b>	<b>Ref. 30e</b>	<b>Ref. 30f</b>	
<b>Current density (mA g<sup>-1</sup>)</b>	50	100	890	45	50	170	50	500	
<b>After 20 cycles</b>	98.4		80.8				82.7		
<b>After 60 cycles</b>	98.5	89.0			90.3				
<b>After 100 cycles</b>	98.8			74.4		95.3		96.0	

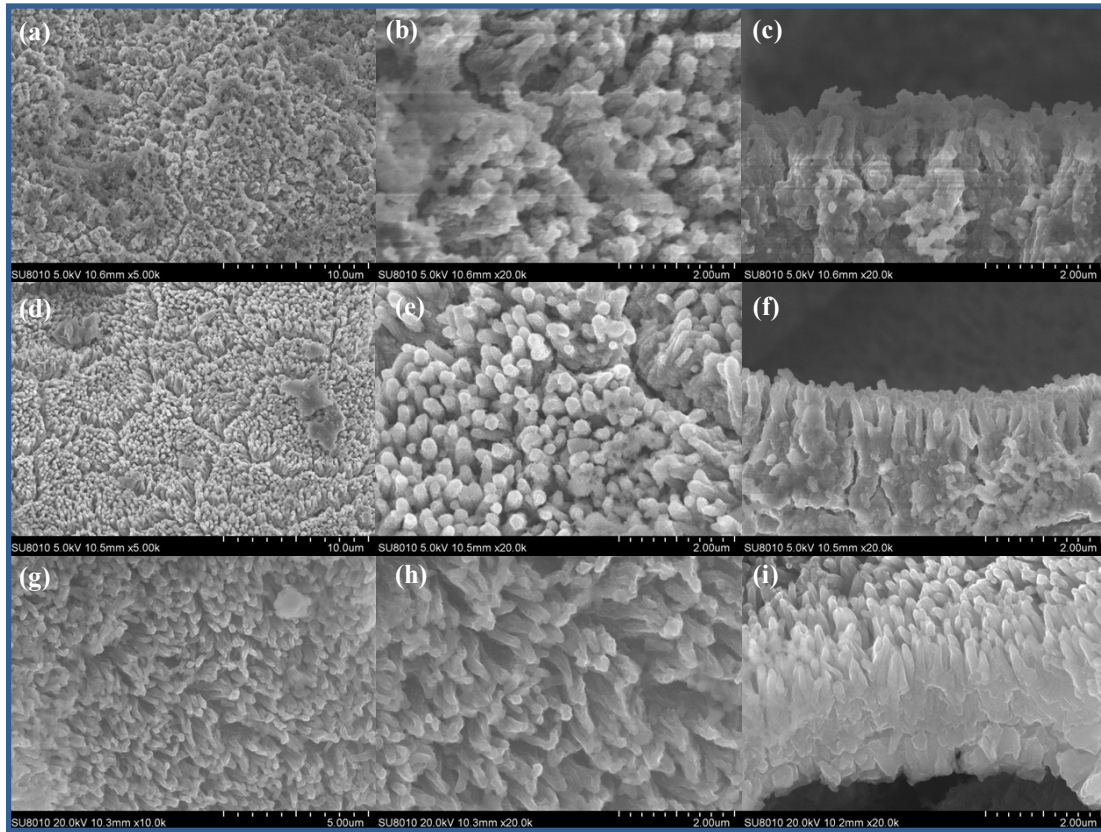


**Figure S6** Al 2p High-resolution XPS spectra of (a) AO-NS and (b) LAO-NS after discharging to 1.35 V, 0.01 V, and recharging to 2.0 V and 3.0 V, respectively.





**Figure S7** Equivalent circuit used to simulate resultant Nyquist plots.



**Figure S8** Morphologies of (a, b, and c) NS, (d, e, and f) AO-NS, (g, h, and i) LAO-NS after 100 cycles. (a, d, and g): top view, (b, e, and h): magnified top view, and (c, f, and i): side view of (NS, AO-NS, and LAO-NS), respectively.