

## Supporting Information

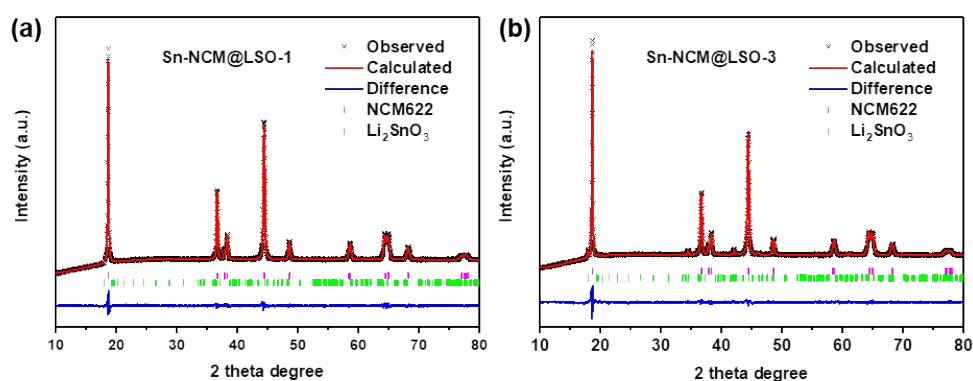
### An Effective Dual-Modification Strategy to Enhance the Performance of the $\text{LiNi}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2}\text{O}_2$ Cathode for Li-ion Battery

Liang Wang, Jiashun Liang, Xiaoyu Zhang, Shenzhou Li, Tanyuan Wang, Feng Ma,

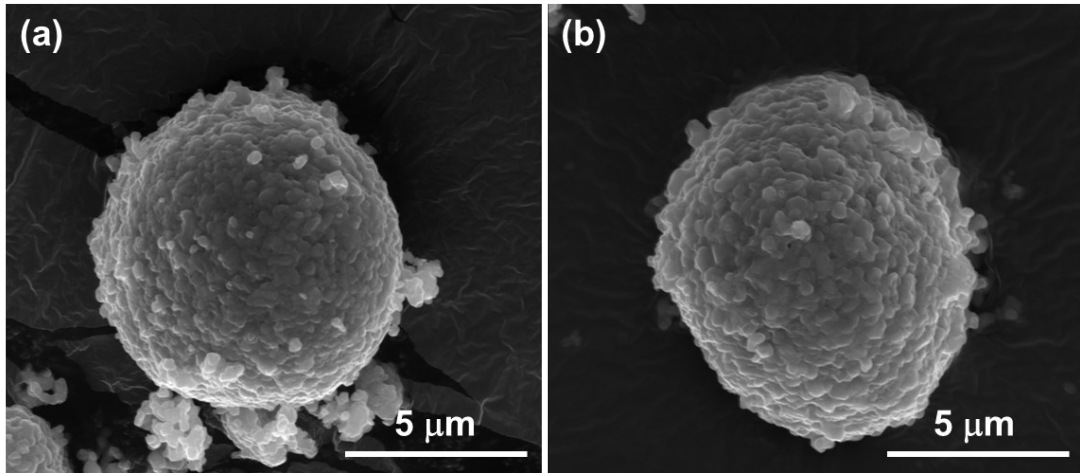
Jiantao Han, Yunhui Huang, Qing Li\*

State Key Laboratory of Material Processing and Die & Mould Technology, School of Materials Science and Engineering, Huazhong University of Science and Technology, Wuhan 430074, China.

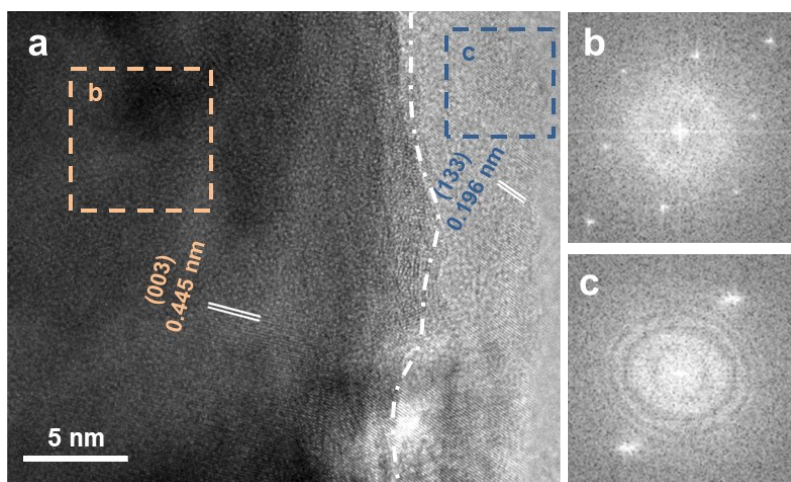
\*Corresponding Author. E-mail: [qing\\_li@hust.edu.cn](mailto:qing_li@hust.edu.cn)



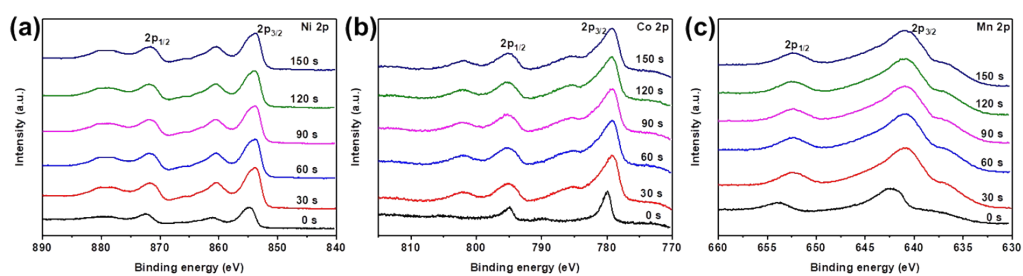
**Figure S1.** Rietveld refinements from XRD pattern of (a) Sn-NCM@LSO-1 and (b) Sn-NCM@LSO-3.



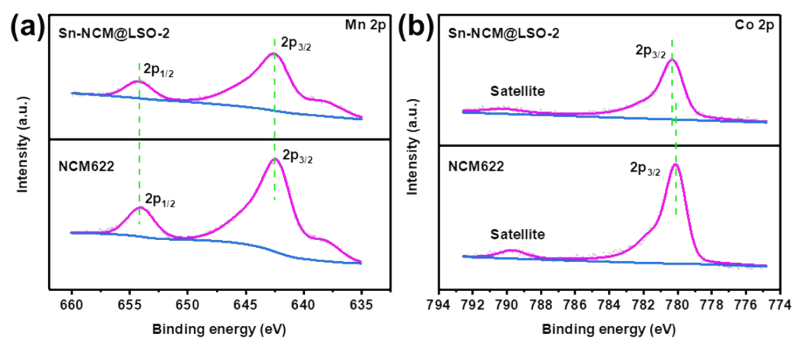
**Figure S2.** SEM images of (a) NCM622 and (b) Sn-NCM@LSO-2.



**Figure S3.** The HRTEM image recorded from different primary particle of Sn-NCM@LSO-2.



**Figure S4.** XPS depth profiles of the (a) Ni 2p, (b) Co 2p and (c) Mn 2p for Sn-NCM@LSO-2.



**Figure S5.** (a) Ni 2p and (b) Mn 2p XPS spectra for the surface of NCM622 and Sn-NCM@LSO-2.

### **Titration of residual lithium**

Since the excessive amount of lithium is necessary to produce well-defined structure for Ni-rich layer compounds, unreacted lithium ingredient can be remained on the surface of Ni-rich materials, presumably as an oxide form,  $\text{Li}_2\text{O}$ . Furthermore, the outer part of the  $\text{Li}_2\text{O}$  is contaminated with moisture and  $\text{CO}_2$  in air, forming residual Li compounds ( $\text{Li}_2\text{CO}_3$  and  $\text{LiOH}$ ). To better evaluate the residual lithium of Ni-rich ternary materials, Aishui Yu et al. carried out chemical titration method. Use hydrochloric acid to titrate  $\text{OH}^-$  and  $\text{CO}_3^{2-}$ . (J. Power Sources 482 (2021) 228940) The indicators are phenolphthalein and methyl red. The color of phenolphthalein corresponds to  $\text{H}^+ + \text{OH}^- \rightarrow \text{H}_2\text{O}$ ,  $\text{CO}_3^{2-} + \text{H}^+ \rightarrow \text{HCO}_3^-$ , and the color of methyl red corresponds to  $\text{HCO}_3^- + \text{H}^+ \rightarrow \text{H}_2\text{O} + \text{CO}_2$ .

Here we referred to the work reported by Aishui Yu et al. and the residual lithium compounds of the samples were collected by chemical titration. The detailed experimental steps were followed: A total of 5 g of sample was dispersed into 50 mL of deionized water with magnetic stirring for 15 min. The suitable amount of solution was titrated with 0.1 mol/L hydrochloric acid (HCl) with methyl red and phenolphthalein as indicators. Before titration, recorded the burette value  $V_1$ . When phenolphthalein (2 drops) changed color, the reaction was suspended, and the burette value was recorded  $V_2$ . As the methyl red (5 drops) changed color again, recorded the burette value  $V_3$ .

The contents of the residual lithium compounds were calculated on the basis of the volume of HCl and the content of the residual lithium compounds can be calculated as follows:

$$w = \frac{\left[ \frac{C(V_3 - V_1)}{1000 * 2} * 5 * 73.8909 \right]}{M} * 1000000 \quad (1)$$

w—residual lithium content, ppm

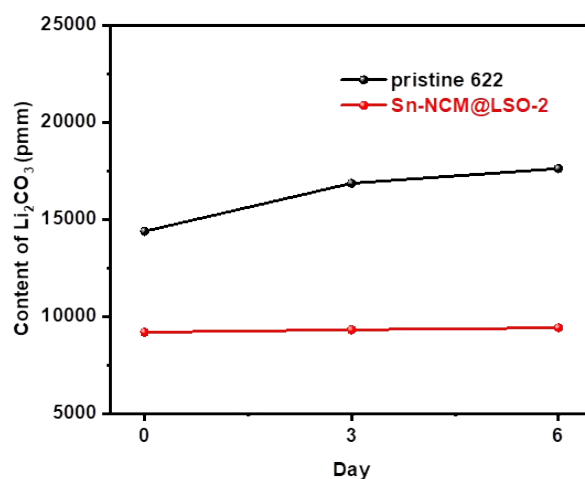
C—the concentration of hydrochloric acid, 0.1 mol/L

M—sample mass, 5g

V<sub>3</sub>—value after the end of titration, mL

V<sub>1</sub>—value before titration, mL

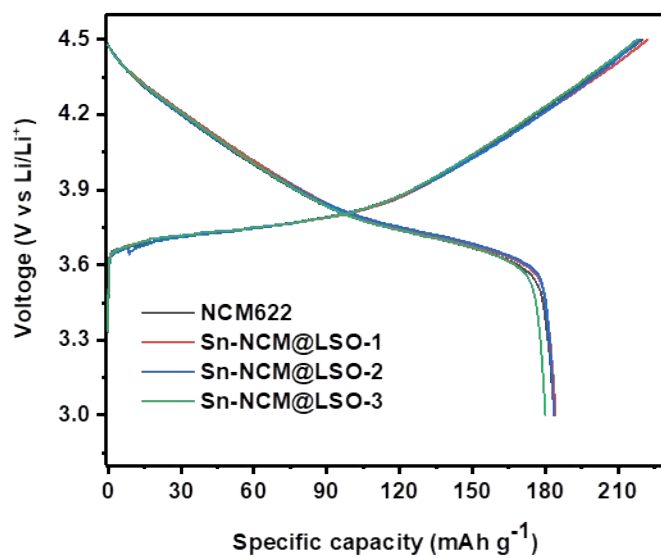
(V<sub>2</sub> can be eliminated in the simplification of the residual lithium calculation formula, so there is no V<sub>2</sub>)



**Figure S6.** The contents of residual lithium for the pristine 622 and Sn-NCM@LSO-2 samples.

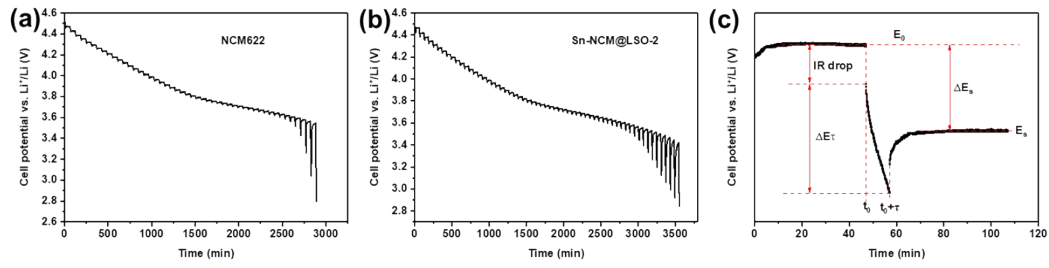
As shown in **Figure S6**, it can be seen that the amount of remaining residual lithium for the newly synthesized NCM622 was 14381 ppm, and it increases to 17615 ppm after 6 days of exposure. In contrast, the content of residual lithium for Sn-NCM@LSO-2 increases from 9181 to 9411 ppm. The lower content of residual

lithium indicates that the formation of  $\text{Li}_2\text{SnO}_3$  coating layer can reduce residual Li and protect pristine from side reactions with air.



**Figure S7.** The initial charge/discharge curves of the NCM622 and Sn-NCM@LSO samples at 0.2 C.





**Figure S8.** GITT curves for the discharge of bare (a) NCM622 and (b) Sn-NCM@LSO-2 in the voltage range of 3.0–4.5 V; (c) scheme for a single titration step of GITT curves.

The lithium diffusion coefficient ( $D_{Li^+}$ ) is calculated using following formula (2):

$$D_{Li^+} = \frac{4}{\pi\tau} \left( \frac{n_M V_M}{S} \right)^2 \left( \frac{\Delta E_s}{\Delta E_\tau} \right)^2 \quad \left( \tau \ll \frac{L^2}{D_{Li^+}} \right) \quad (2)$$

Where  $\tau$  is the duration of the current pulse (s);  $n_M$  is the number of moles (mol);  $V_M$  is the molar volume of the electrode ( $\text{cm}^3/\text{mol}$ );  $S$  is the electrode/electrolyte contact area ( $\text{cm}^2$ );  $\Delta E_s$  is the steady-state voltage change, due to the current pulse and  $\Delta E_\tau$  is the voltage change during the constant current pulse, eliminating the IR drop)