

ADVANCED FUNCTIONAL MATERIALS

Supporting Information

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Atomic Insights into the Enhanced Surface Stability in High Voltage Cathode Materials by Ultrathin Coating

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Fine-tuning of $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ synthesis method

The amount of lithium acetate added in the second round of 500°C heating will affect the quality of the final product. Since some of the nickel acetate and manganese acetate sticks to the wall of the crucible after heating, when adding Li source to the mixture, the amount is carefully calculated based on the actual amount of Ni and Mn sources taken out of the crucible, where anything stuck on the wall was excluded. In addition, since the top portion of the mixture experiences more Li source volatilization, the total amount of precursors used in each round was adjusted until the influence from Li source volatilization was minimized.

Figure S1

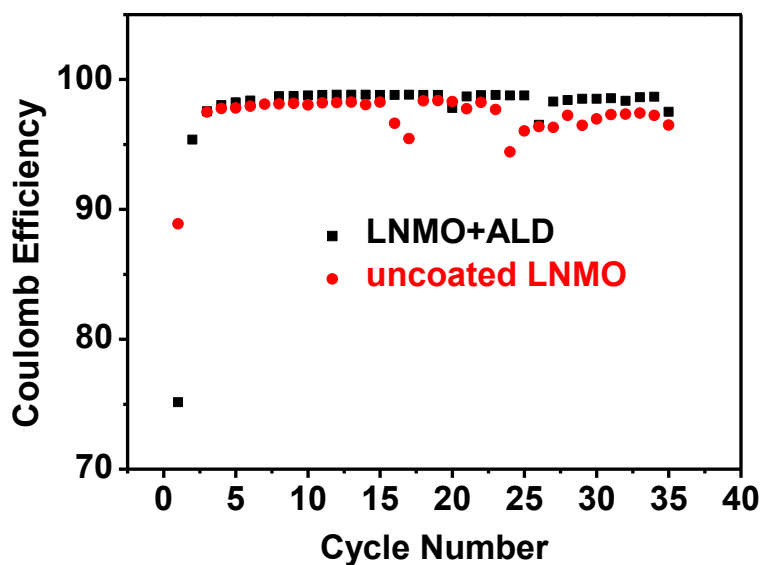


Figure S1 Coulombic efficiency of coated and uncoated samples cycled at C/5.

We note here we have tested more than 10 cells and the ones shown here are the ones

used for XAS measurement. The mean values of first cycle coulombic efficiency between coated and uncoated samples are similar, 86.80% (coated) vs. 86.05% (uncoated). The slight difference may be due to the side reaction between electrolyte and $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$, where in the uncoated samples the reaction is more severe and leads to a slightly lower first cycle coulombic efficiency.

EIS measured before cycling

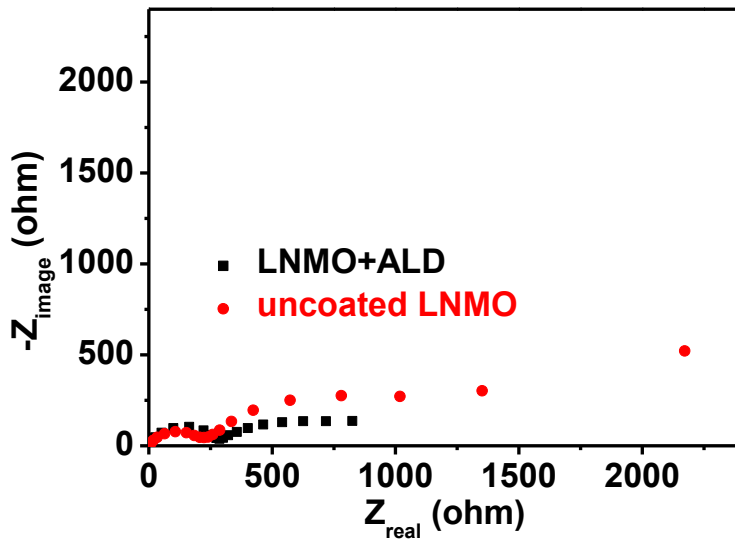


Figure S2 Comparison of EIS before cycling

EIS data on cells measured before cycling is shown in Figure S2. The cell containing the ALD coated sample showed a slightly larger charge transfer resistance which might be caused by the added Al_2O_3 layer.

EELS profiles at each depth of scanning

Figure S3

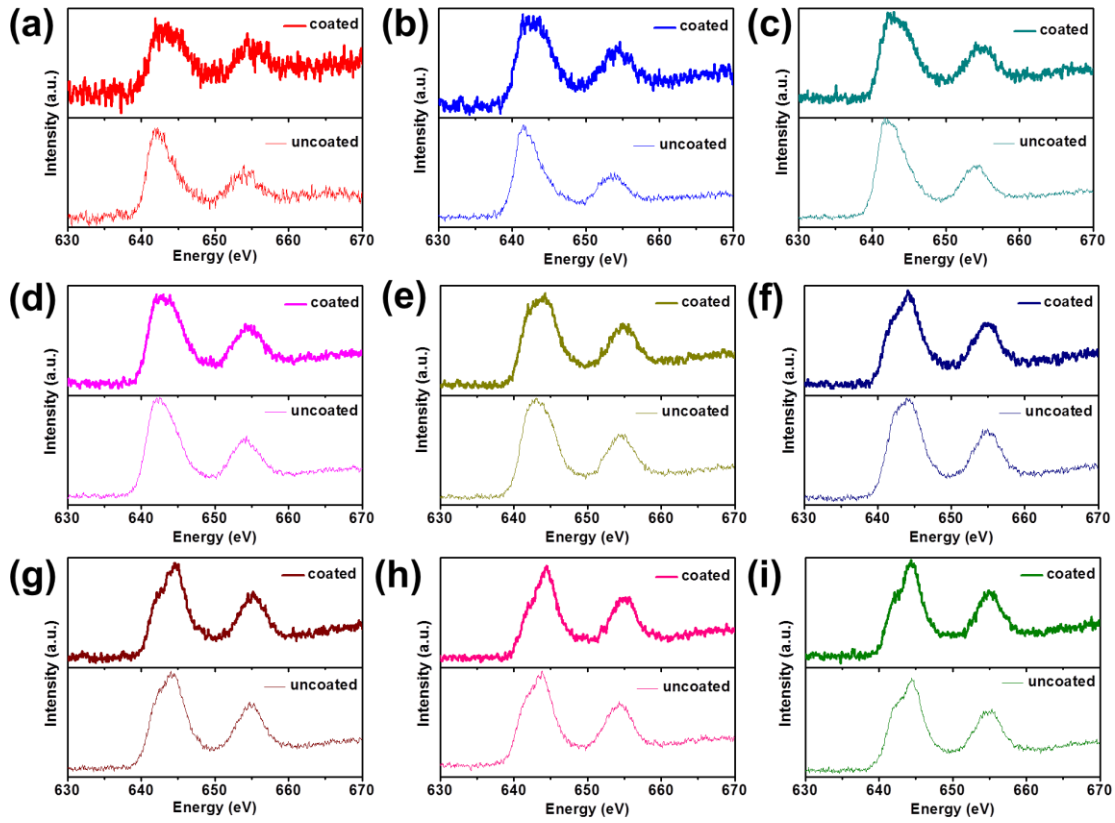


Figure S3 Comparison of EELS profiles for coated and uncoated samples shown in Figure 4a and 4b at each depth from surface (a) along the scanning path to bulk (i). Each color corresponds to the respective color in Figure 4a and 4b. Step size is 1.5 nm.