

Electronic Supplementary Information

Assessing Coulombic Efficiency in Lithium Metal Anodes

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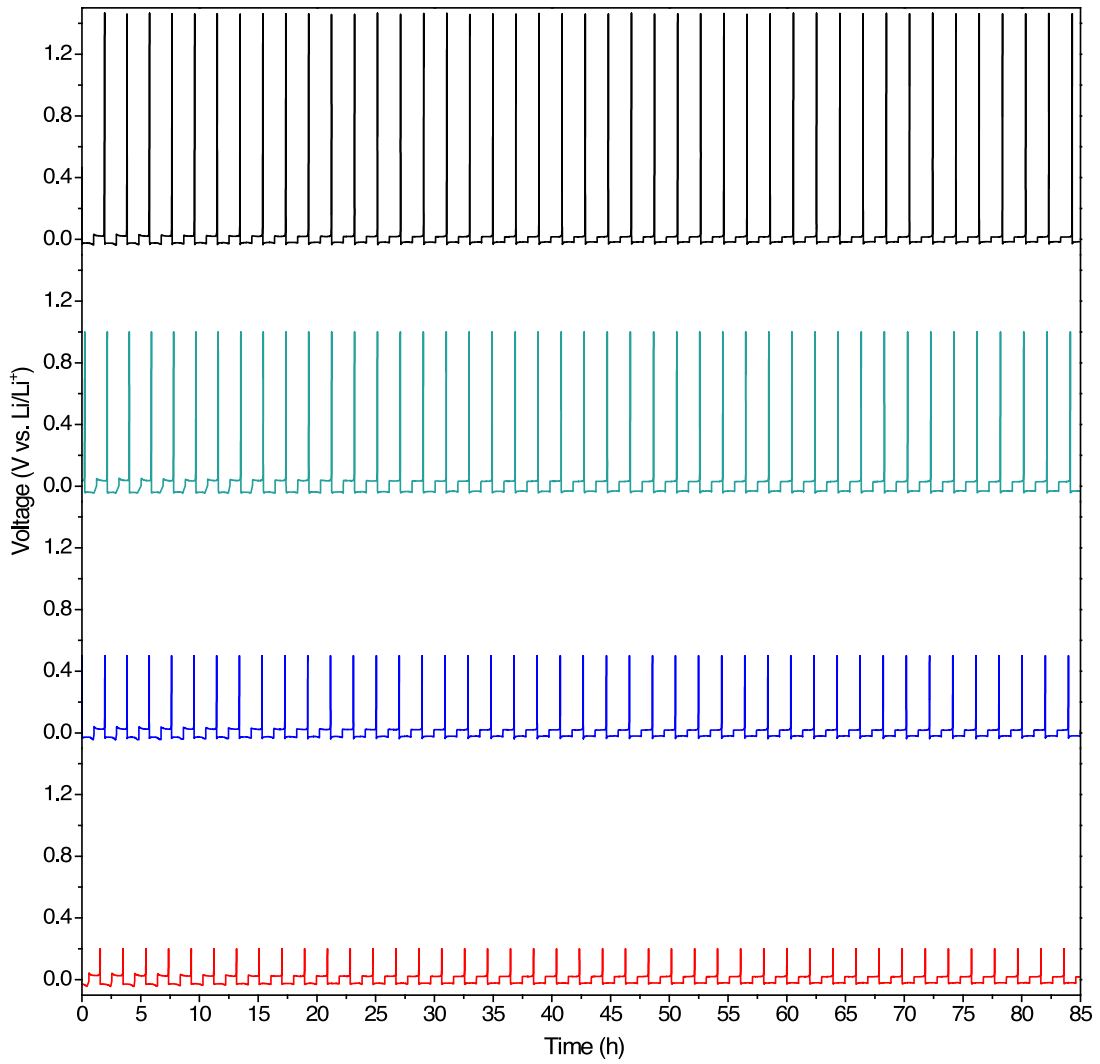


Fig. S1: Conventional plating/stripping curve at the current density of 1 mA cm^{-2} with the different cut-off voltage of 1.5, 1.0, 0.5, and 0.2 V.

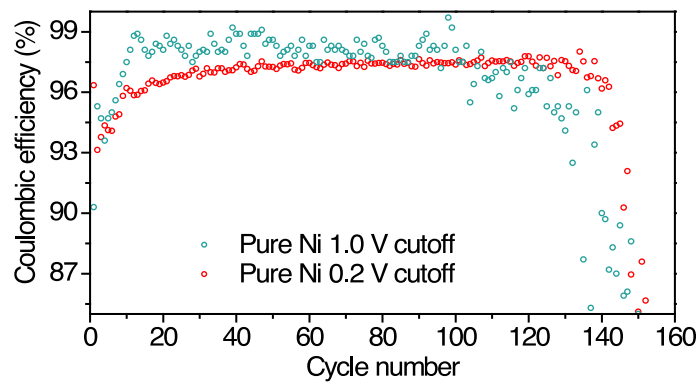


Fig. S2: Cyclic performance and CE dependence of the 0.2 and 1.0 V cutoff voltages at the current density of 1 mA cm^{-2} and capacity of 1 mAh cm^{-2} for pure Ni sample.

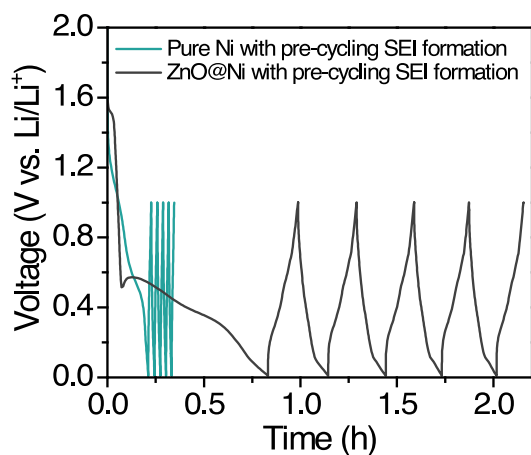


Fig. S3: SEI formation cycles at a current density of 0.2 mA cm^{-2} for the ZnO-coated and pure Ni foam samples.

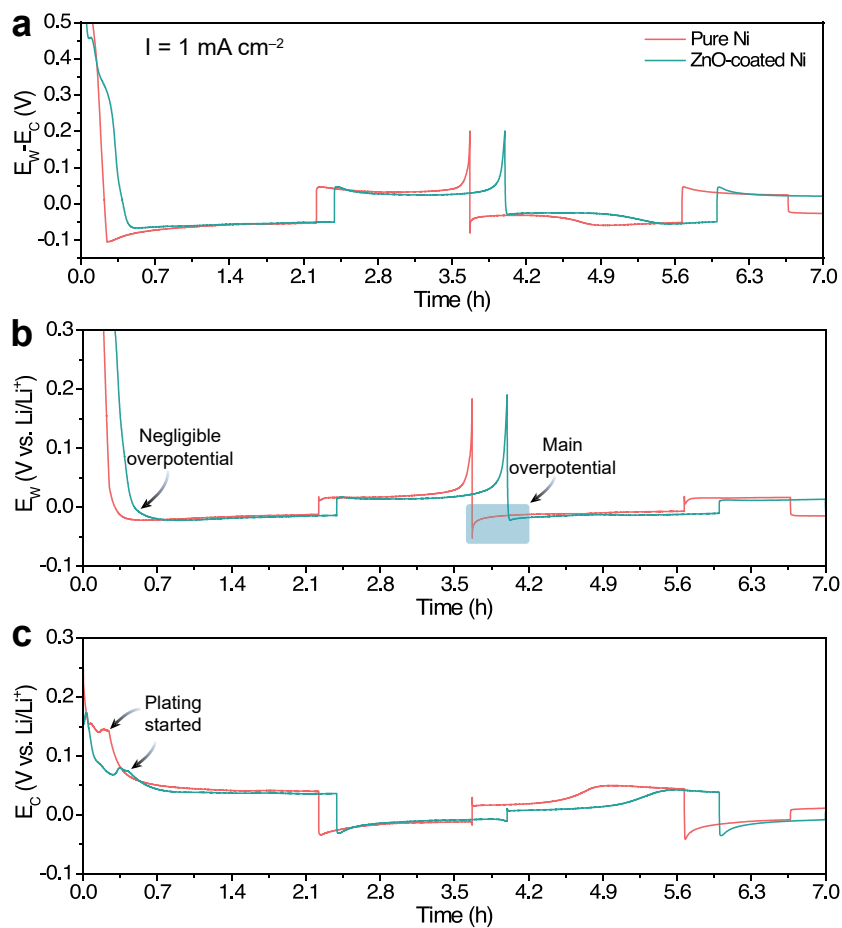


Fig. S4: Swagelok three-electrode measurement with a current density of 1 mA cm^{-2} to decouple the voltage contributions for the pure Ni foam and ZnO-coated Ni electrodes, a) $E_{\text{working}} - E_{\text{counter}}$, b) working electrode, and c) counter electrode contribution.

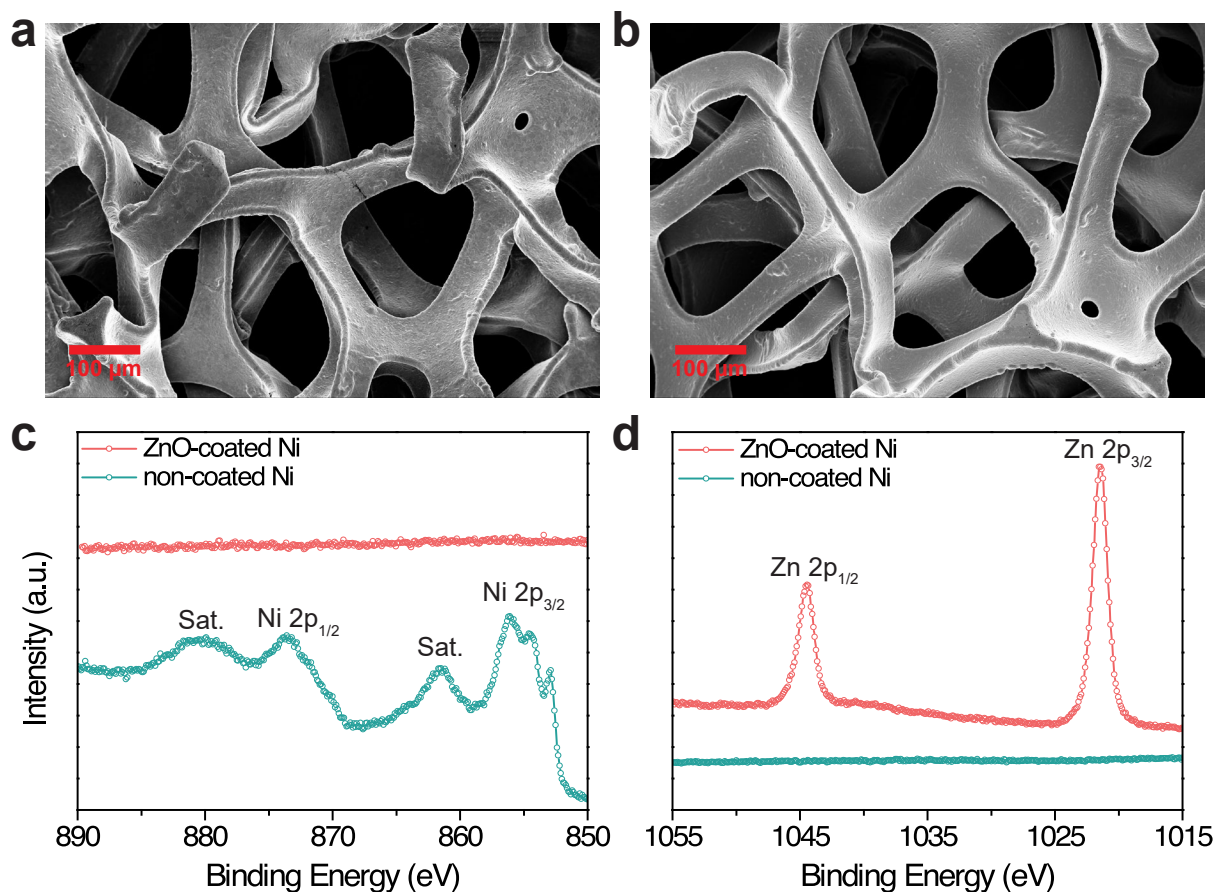


Fig. S5: SEM images of a) non-coated Ni, b) 100 nm ZnO-coated Ni foam, high-resolution XPS spectra of c) Ni 2p, and d) Zn 2p of the non- and 100 nm ZnO-coated samples.

Note: XPS and SEM were used to evaluate the morphology and chemical composition of non- and ZnO-coated Ni foam samples. The bare Ni foam forms a three-dimensional microporous cross-linked skeleton structure, as shown in Fig. S4a. After ALD ZnO modification, there was no significant change to the surface morphology of Ni foam (Fig. S4b). The high-resolution XPS spectra of Ni 2p and Zn 2p for the non- and ZnO-coated samples are shown in Fig. S4c and d, respectively. The high-resolution spectra of Ni 2p of the non-coated sample included the Ni 2p_{3/2} and Ni 2p_{1/2} regions along with their satellite peaks. Comparatively, the relative characteristic peak of Ni 2p was not observed for the ZnO-coated sample (Fig. S4c), indicating that the metallic Ni foam substrate was totally covered by ZnO. Furthermore, the ZnO deposited sample shows the clear characteristic Zn 2p doublet peaks at 1044.48 eV and 1021.46 eV corresponding to Zn 2p_{1/2} and Zn 2p_{3/2}, respectively. The spin-orbit splitting between Zn 2p_{1/2} and Zn 2p_{3/2} is approximately 23 eV, indicating that zinc is in the Zn²⁺ binding state in the ZnO.

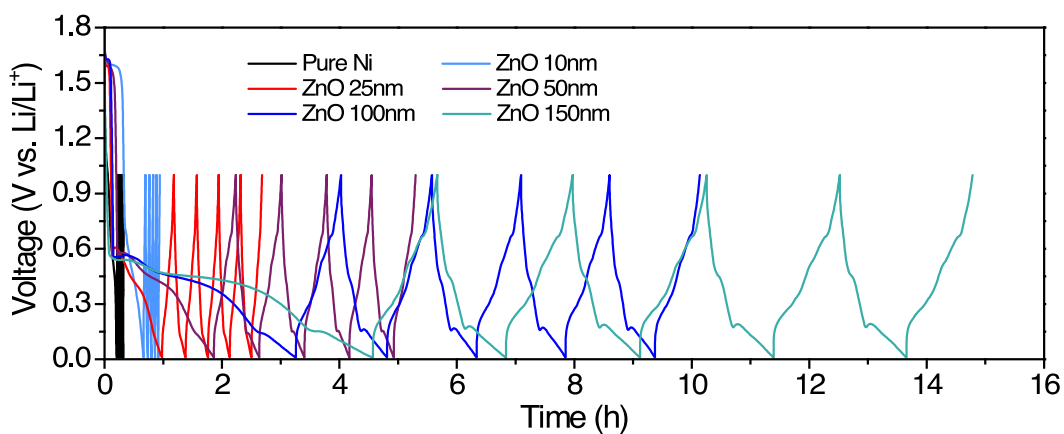


Fig. S6: SEI formation cycles with a current density of 0.2 mA cm^{-2} before the CE measurement in different thicknesses of ZnO-coated Ni foam samples.

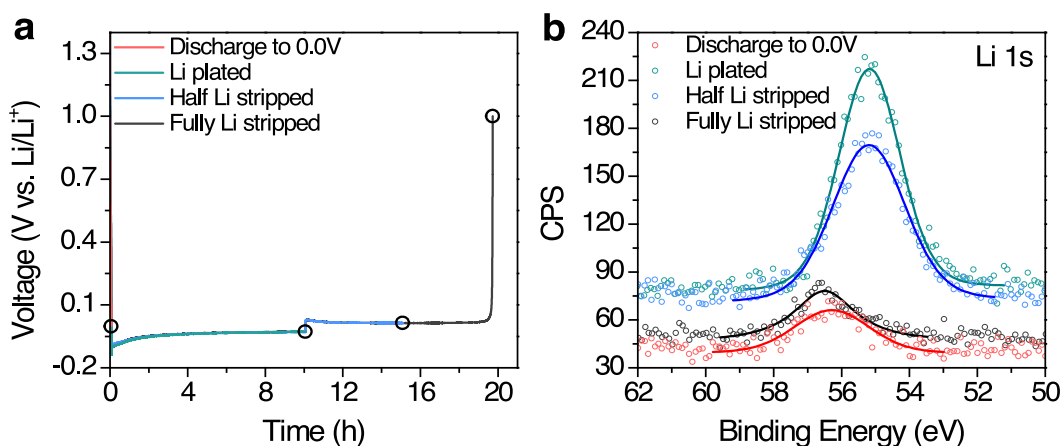


Fig. S7: a) Li plating/stripping curve obtained for XPS analysis in a different point of plating/stripping, SEI formation (discharge the battery to 0.0 V), Li plated on the Ni foam, stripping half of plated Li (define the reservoir method), and stripping all the Li on the surface (define the conventional method) and b) Li 1s spectra of XPS in a various point of plating/stripping. According to the Li 1s spectra, the changes in this spectrum for the Li-plated samples indicate that metallic lithium is deposited on Ni foam.

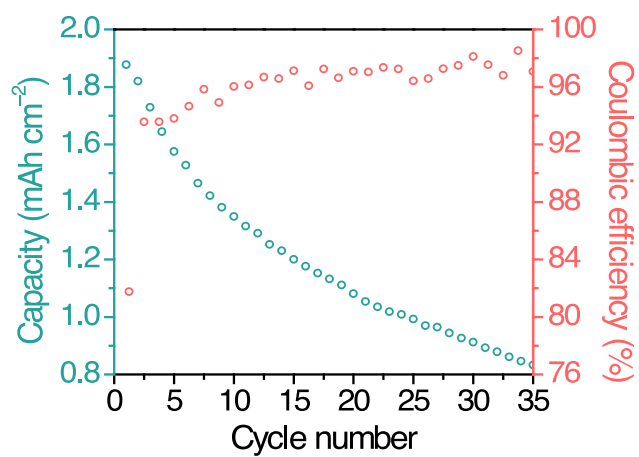


Fig. S8: Capacity retention (green) and CE (red) of pure Ni || LFP cell in the conventional electrochemical method as a function of cycle number when charged/discharged at the first cycle was 0.1 mA cm^{-2} and for the following cycles were 0.5 mA cm^{-2} .