

## ***Electronic Supplementary Information***

### **Towards Understanding the Nucleation and Growth Mechanism of Li Dendrites on Zinc oxide-coated Nickel Electrodes**

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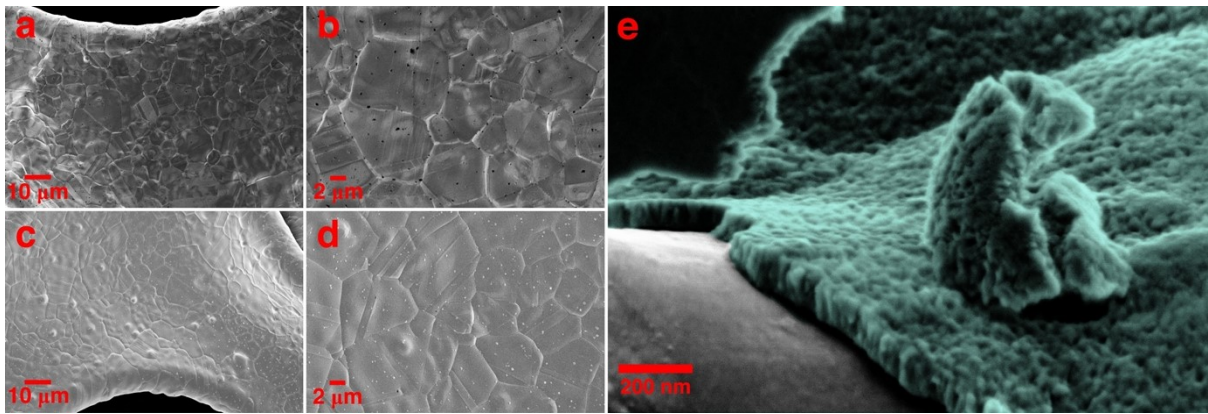
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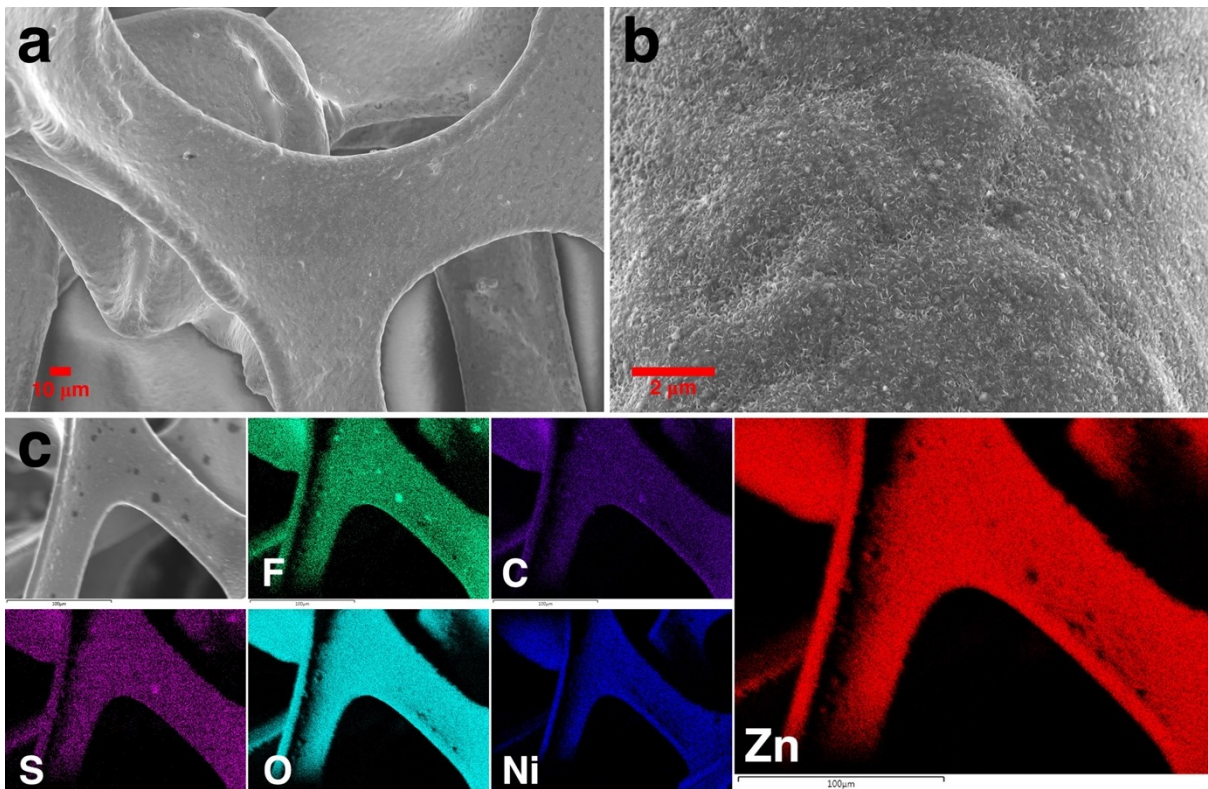
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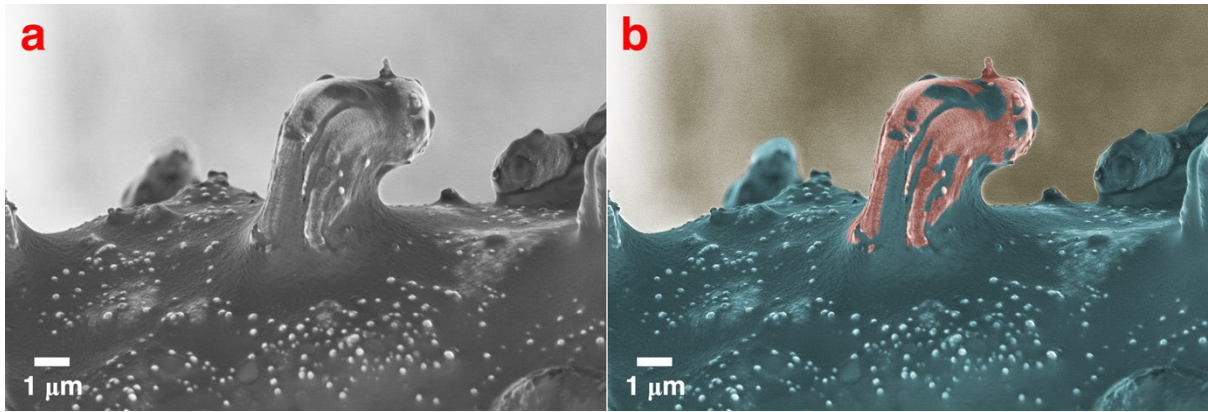
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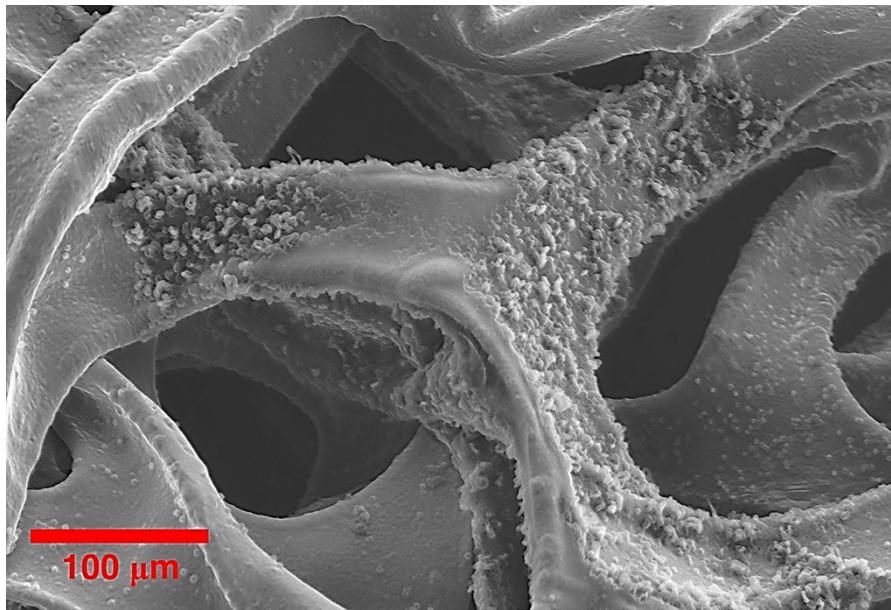
*Fig. S1: Zinc oxide modified electrode morphology: SEM image of pure Ni (a, b) and ZnO@Ni (c, d) captured at increasing magnification, and e) spongy morphology of ZnO at higher magnification (the colorized green part of the photo shows the morphology of zinc oxide. This image was taken in the part of the sample which was scratched).*



*Fig. S2: Conversion reaction and SEI formation: a, b) Low and high magnification SEM image of ZnO@Ni electrode after discharge to 0.0 V, and c) SEM image and corresponding elemental mapping (F, C, S, O, Ni, Zn) of the electrode.*



*Fig. S3: Early stage of Li dendrite nucleation: SEM image of Li dendrite on the surface of ZnO modified nickel foam in a 1 kV voltage electron (blue: LiZn and Li<sub>2</sub>O layers, red: part of the surface layers that lift off by Li deposition).*



*Fig. S4: SEM images of initial Li deposited on the surface of pure nickel foam with the capacity of 0.25 mAh cm<sup>-2</sup> at 0.5 mA cm<sup>-2</sup>.*

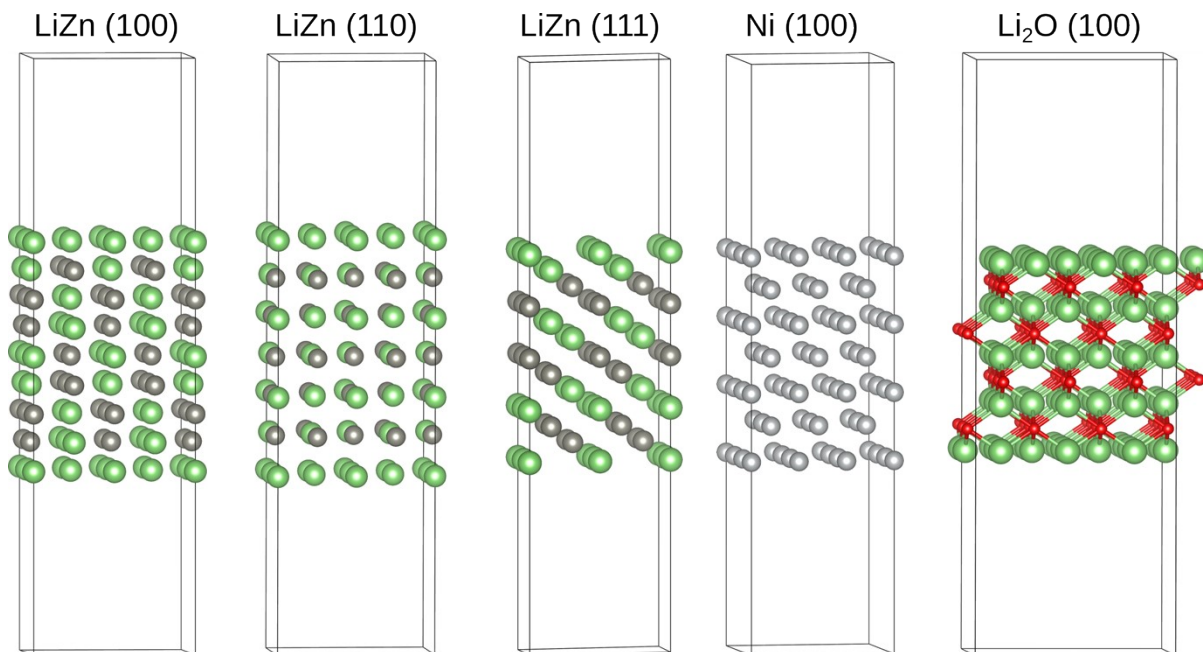


Fig. S5: Cells used for DFT calculations. From left to right: LiZn (100), LiZn (110), LiZn (111), Ni (100), Li<sub>2</sub>O (100). Cells are built symmetric and a vacuum layer of 20 Å has been set in the z-direction. 2 × 2 supercells were considered for all LiZn surfaces and 3 × 3 supercells were considered for Ni and Li<sub>2</sub>O surfaces. Green, dark grey, light grey, and red atoms correspond to lithium, zinc, nickel, and oxygen, respectively.

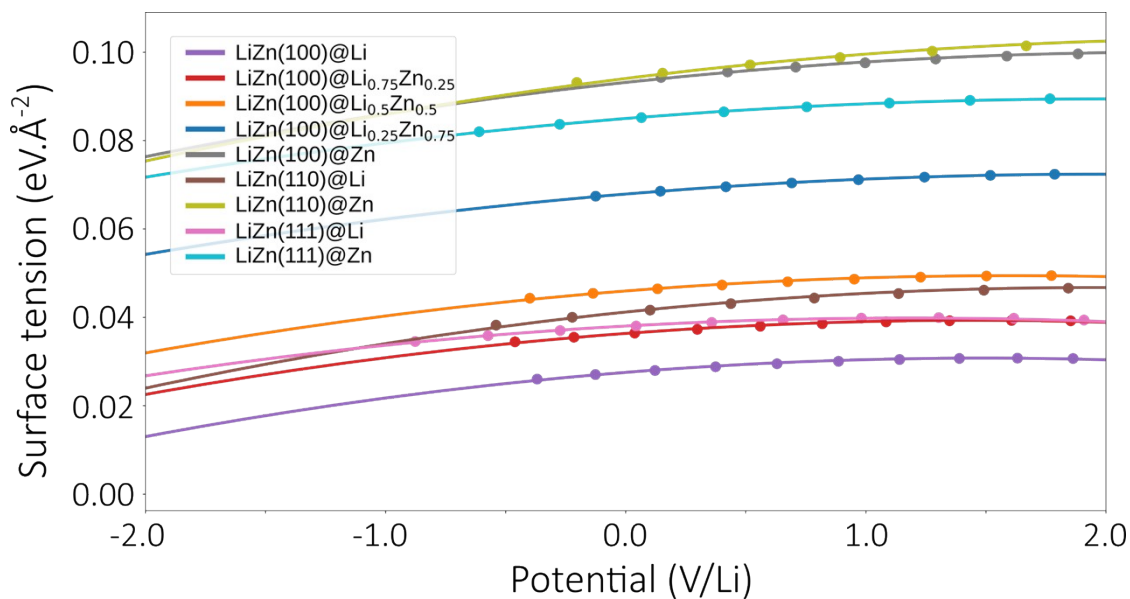


Fig. S6: Electro-capillary curves of the different LiZn surfaces, computed within the GC-DFT framework. Fits correspond to 2<sup>nd</sup> order polynomial fits.



Fig. S7: Simulation cell a) without and b) with a lithium tip. The electrostatic potential profiles provided in the main manuscript are projected in the  $[200]$  plane, as represented by the blue planes.

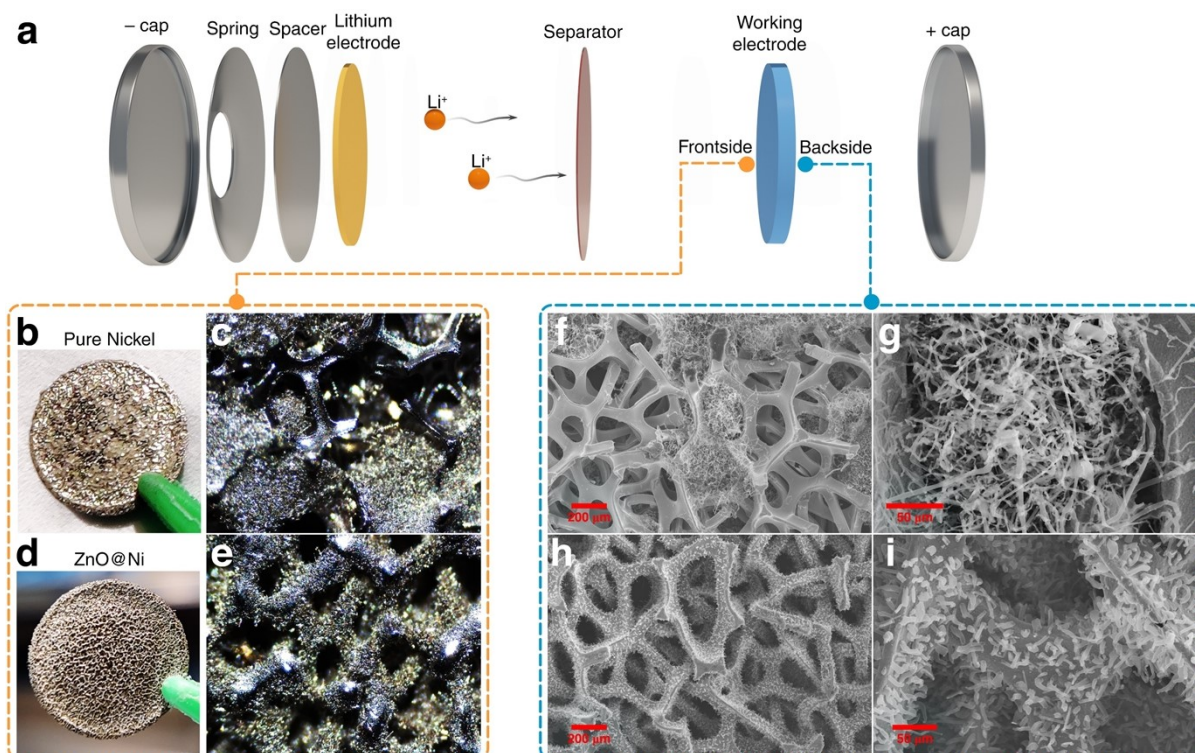
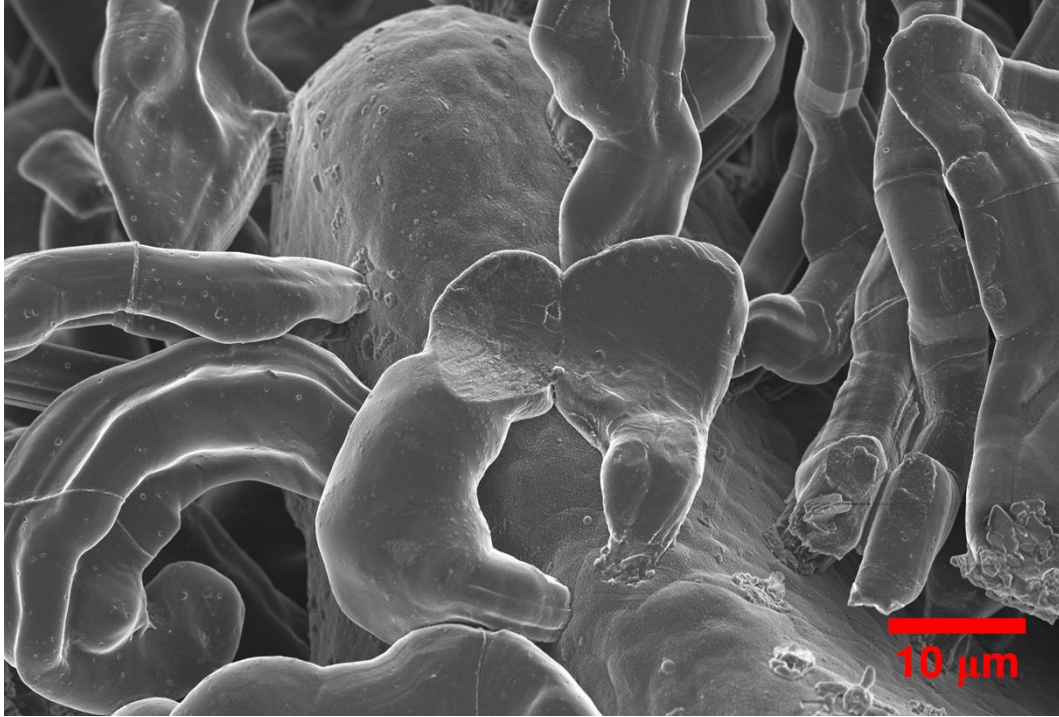


Fig. S8: Morphology of Li deposition on the top and bottom side of electrode: a) Schematic illustration of the coin cell, b, d) photographs of the pure Ni foam and ZnO@Ni after Li deposition, c, e) optical microscope image of the pure Ni foam and ZnO@Ni after Li deposition on the front side of the electrode, and the SEM images of Li deposited on the backside of electrodes for pure Ni (f, g) and ZnO@Ni (h, i) in two different magnification.



*Fig. S9: SEM images of Li deposited on the surface of ZnO modified nickel foam with the capacity of  $4.0 \text{ mAh cm}^{-2}$  at  $0.5 \text{ mA cm}^{-2}$ .*