

# Neutron Depth Profiling (NDP) – a versatile tool for studying battery components

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## Motivation & Introduction

The energy density of lithium-metal batteries (LMBs) largely relies on the thickness of the lithium-metal anode. However, a bare, thin lithium foil is vulnerable to fragmentation due to the commonly inhomogeneous lithium stripping/plating, thereby disrupting the electron conduction pathway across the electrode. For this reason – and as it is very hard to produce free-standing thin lithium foils – they are commonly placed on a copper foil serving as current collector and mechanical support; nevertheless, at the expense of a greatly increased mass that limits the achievable specific energy. Herein, we report a novel polymer/carbon composite current collector that allows for a substantial reduction in mass, while simultaneously extending the cycle life of the lithium-metal anode. The specific mass of the ultra-light, 10 μm thick polymer-based current collector is only 1.03 mg cm<sup>-2</sup> which is less than 1/8 of copper (8.96 mg cm<sup>-2</sup>). As a result, the LMB cells employing this novel current collector provide a specific energy of 447 Wh kg<sup>-1</sup>, which is almost 20% higher than the specific energy when using a copper current collector (378 Wh kg<sup>-1</sup>). Neutron depth profiling (NDP) is a versatile tool to quantify the small Li amount distribution in the polymer/carbon composite versus copper.

## Neutron depth profiling

### 1.) Principle of neutron depth profiling

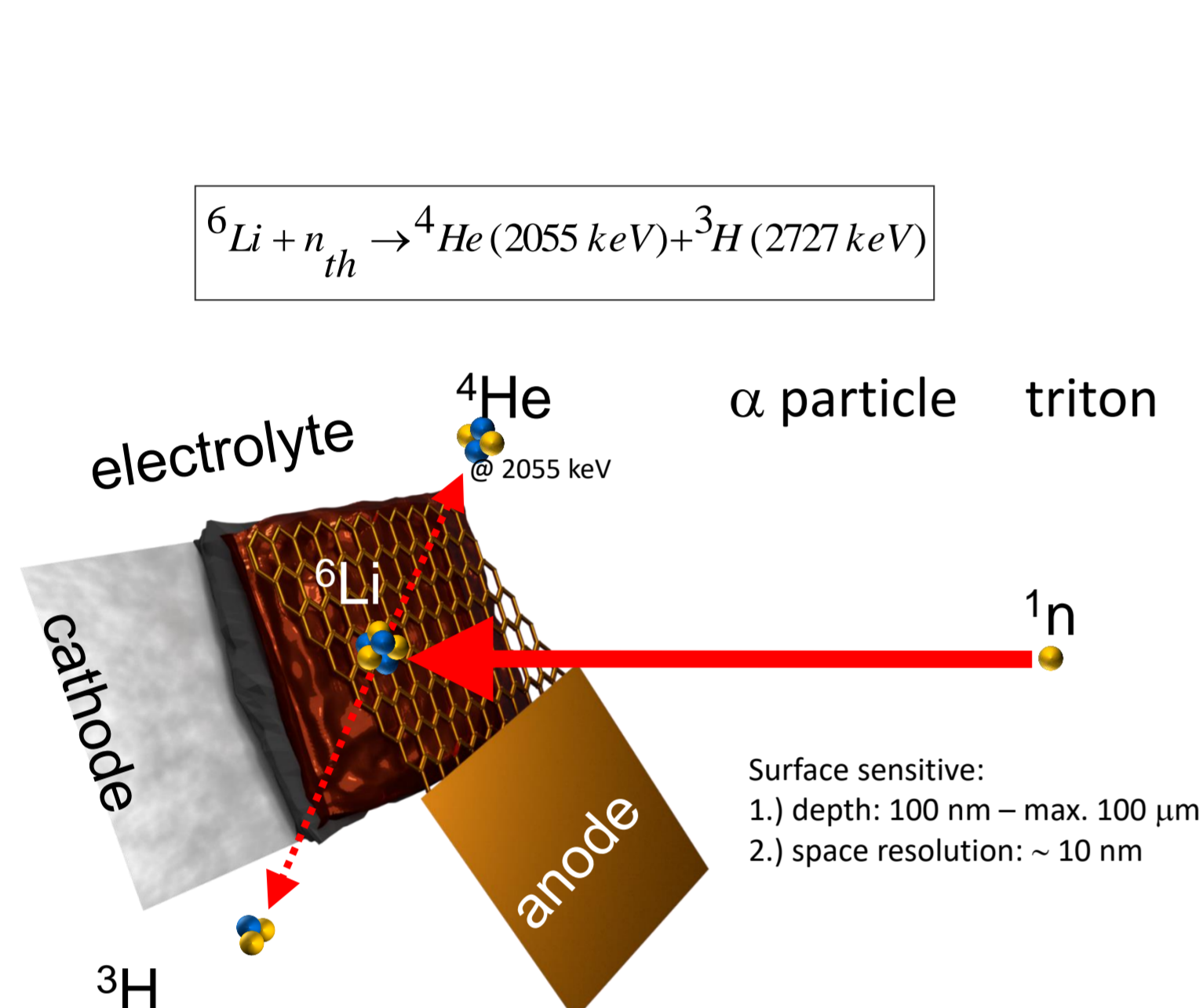


Fig. 1: Li detection via n + <sup>6</sup>Li reaction to a particle and triton particle

### 2.) Neutron depth profiling: Li detection from surface layers to bulk distributions

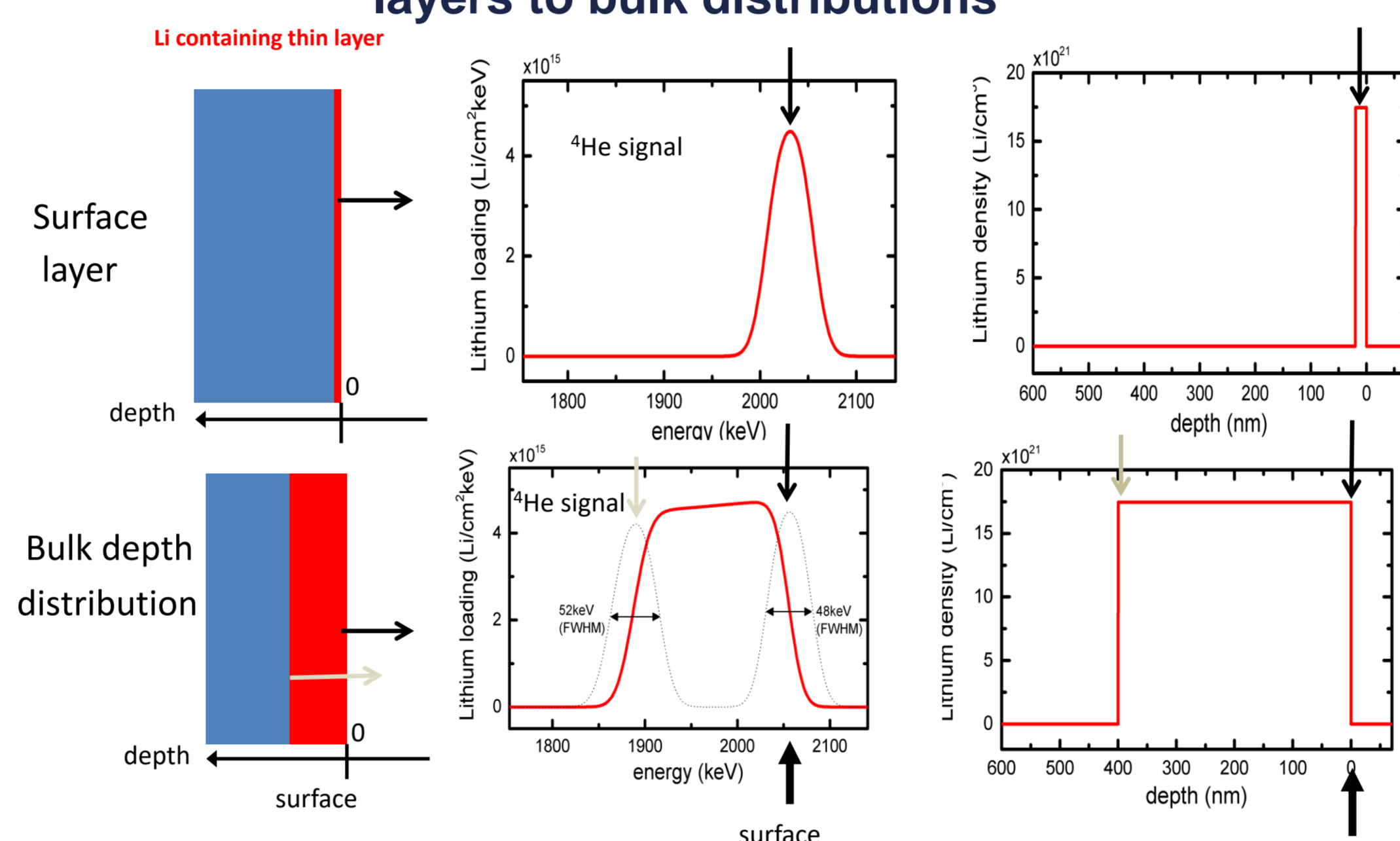


Fig. 2: Stopping power (energy distribution) is dependent on mass and charge of nuclear particle

### 3.) Neutrons applied on Li Ion Batteries

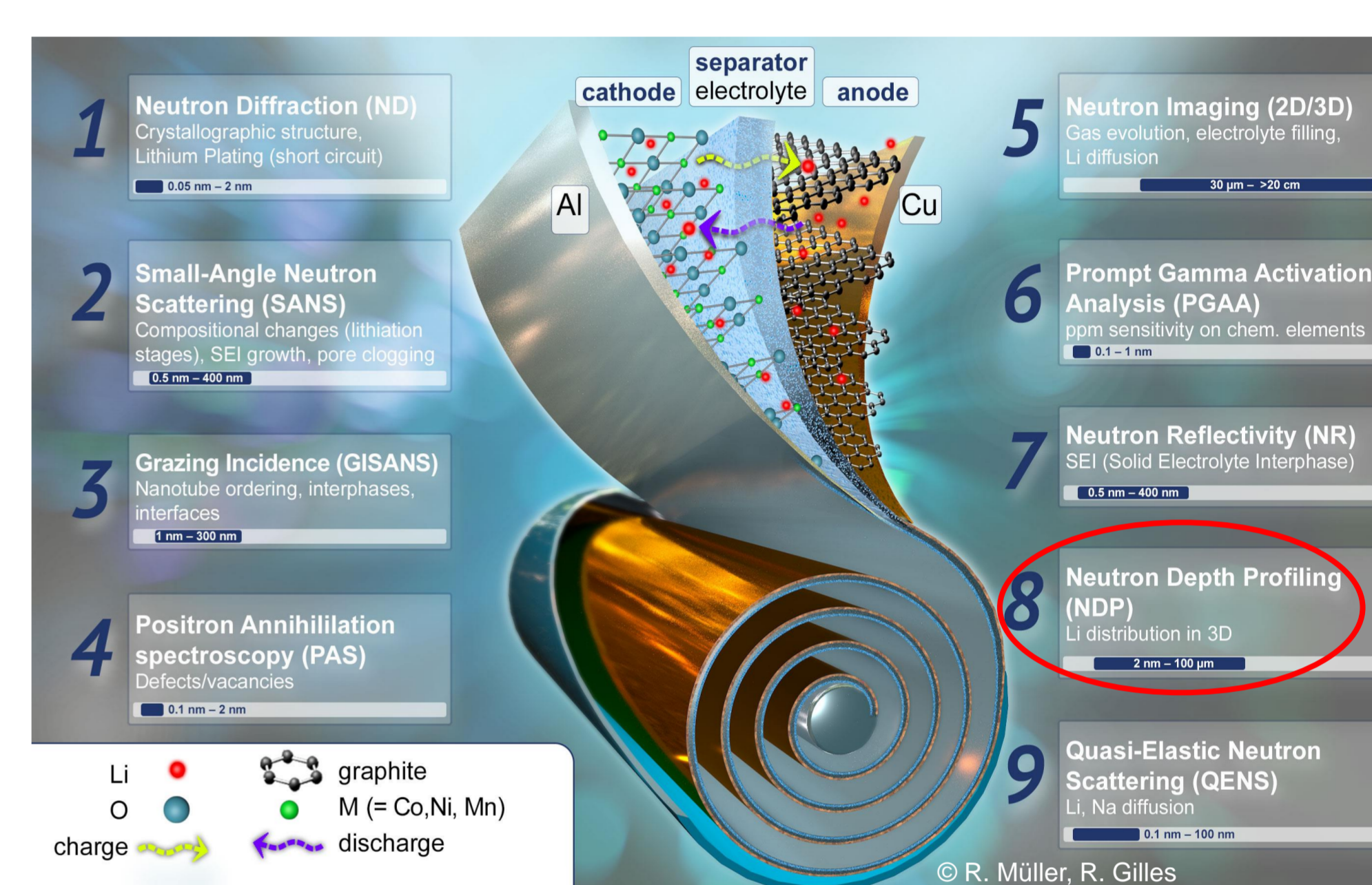


Fig. 3: Neutron methods to study Li-ion batteries

## Metal-free polymer-based current collector for high energy density Lithium-metal batteries

### Introduction

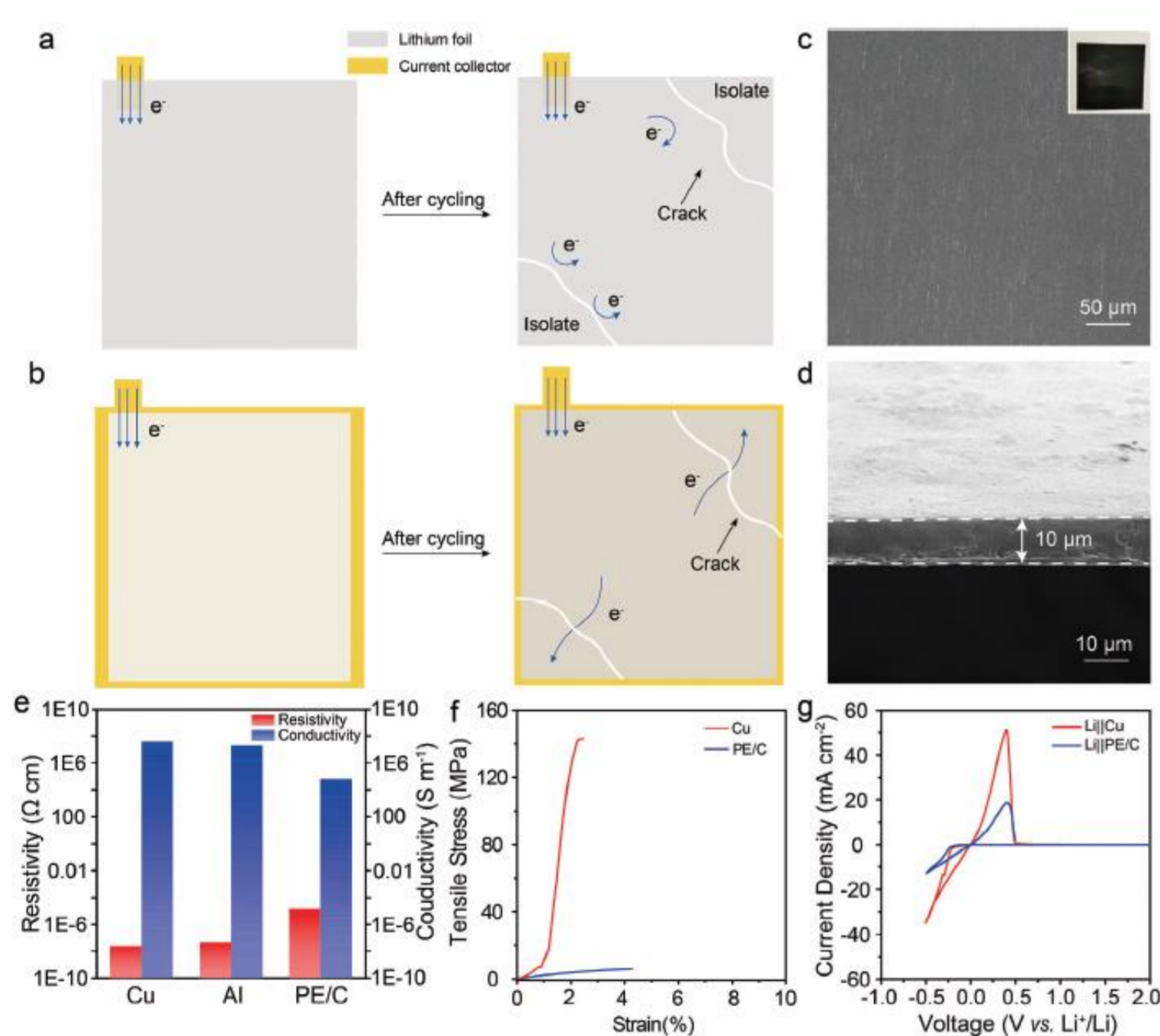


Fig. 4: Characterization of the PE/C current collector. a,b) Schematic illustration of the impact of cracks in the lithium foil formed upon continuous cycling in the a) absence and b) presence of a current collector on the electron transport. c,d) SEM micrographs of c) the surface (the inset shows the corresponding photo) and d) the cross-section of the PE/C current collector. e) Electronic conductivity of the Cu, Al, and PE/C current collector. f) Tensile strength of the Cu and the PE/C current collector. g) Cyclic voltammograms conducted on Li||Cu and the Li||PE/C cells (sweep rate: 10 mV s<sup>-1</sup>; reversing potentials: -0.5 and 1.0 V vs Li+/Li).

### Electrochemical performance of the PE/C current collector

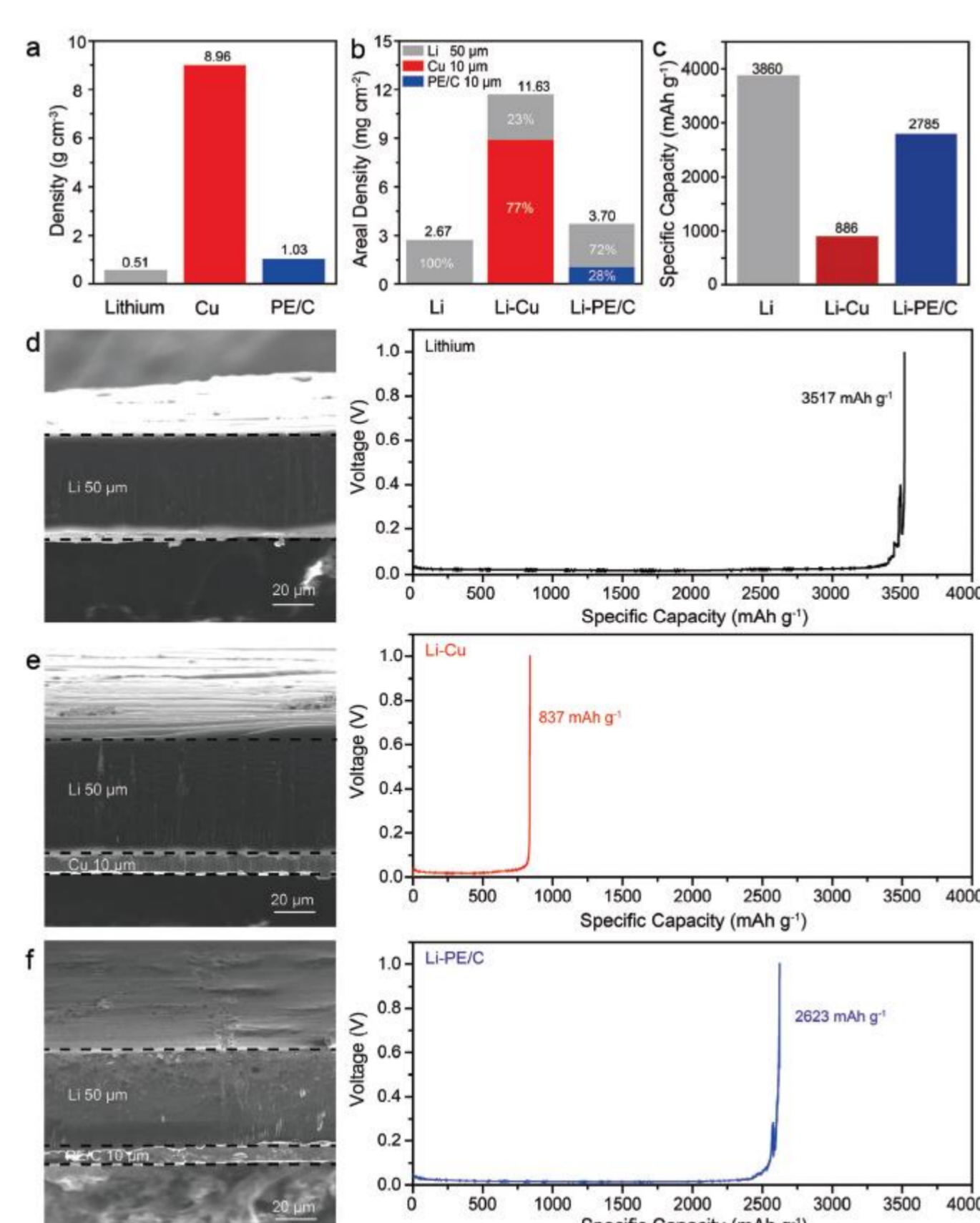


Fig. 5: Impact of the mass of the PE/C current collector on the overall specific capacity of the negative electrode. a) Comparison of the density of neat lithium foil, copper foil, and the PE/C foil, b,c) Comparison of the b) areal density and c) theoretical specific capacity of the neat lithium-metal anode, the Li-Cu anode, and the Li-PE/C anode (in all cases, the lithium foil/layer has a thickness of 50 μm). d-f) Cross-sectional SEM micrographs (left) and the experimentally determined specific capacity (right) of d) the neat lithium-metal anode, e) the Li-Cu anode, and f) the Li-PE/C anode; the specific capacity values refer to the mass of the complete negative electrode, i.e., including the mass of the current collector in the case of Li-Cu and Li-PE/C.

Reference:  
M. Wan, R. Gilles, J. Vacik, H. Liu, N.L. Wu, S. Passerini, D. Bresser  
UltraLight Polymer Current Collector for High Energy Density Lithium-Metal Batteries, Small (2024), 2404437

### Neutron depth profiling measurements

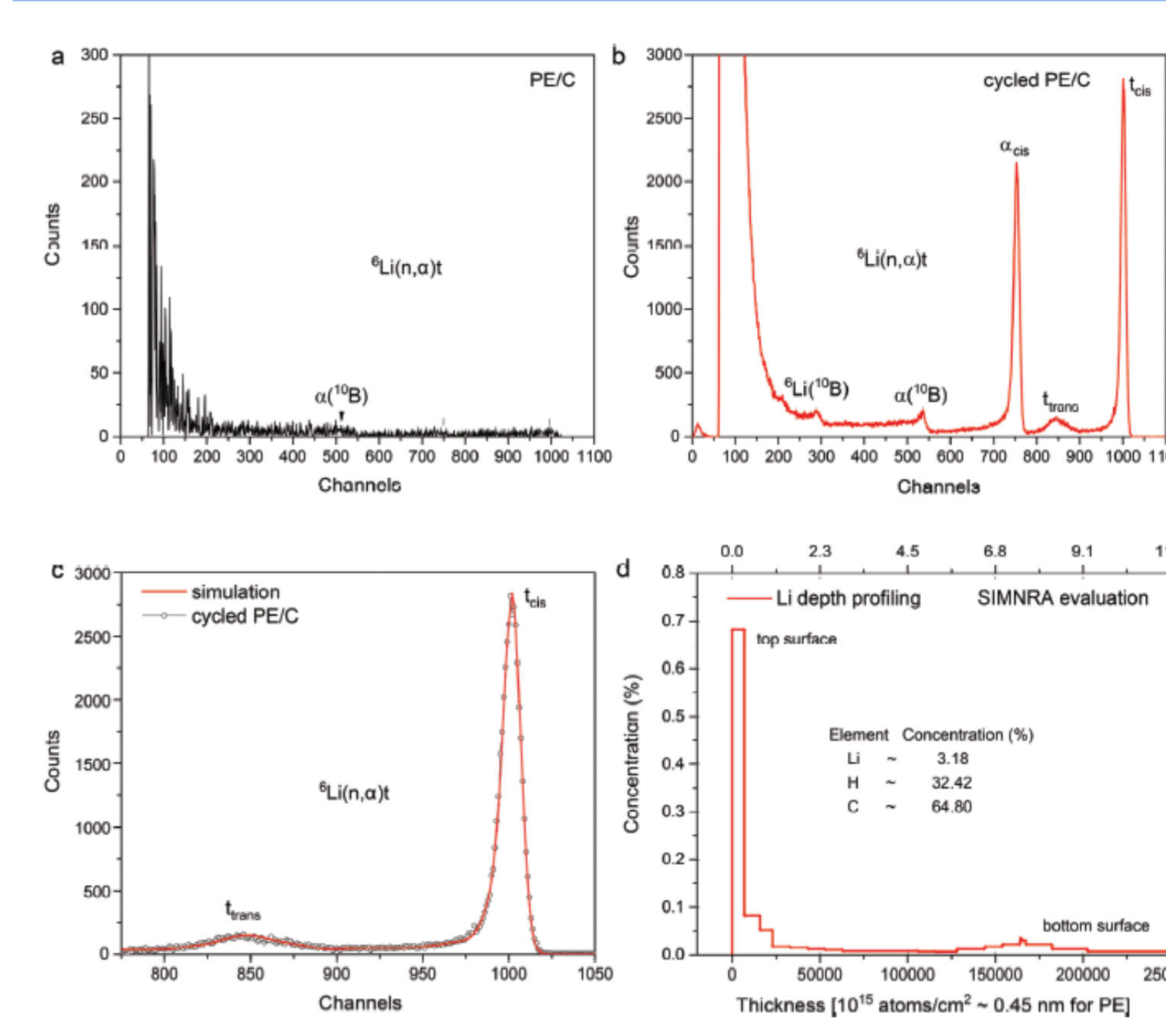


Fig. 6: NDP analysis of pristine and cycled PE/C current collectors. a,b) NDP spectrum of a) the pristine PE/C current collector and b) the PE/C current collector after one lithium plating/stripping cycle with a fixed areal capacity of 1.0 mAh cm<sup>-2</sup> at a current density of 1.0 mA cm<sup>-2</sup>. c,d) NDP spectrum of <sup>6</sup>Li for c) the cycled PE/C current collector, and d) its simulated depth distribution across the PE/C current collector (the sample was studied from the front side).

### Fading mechanism

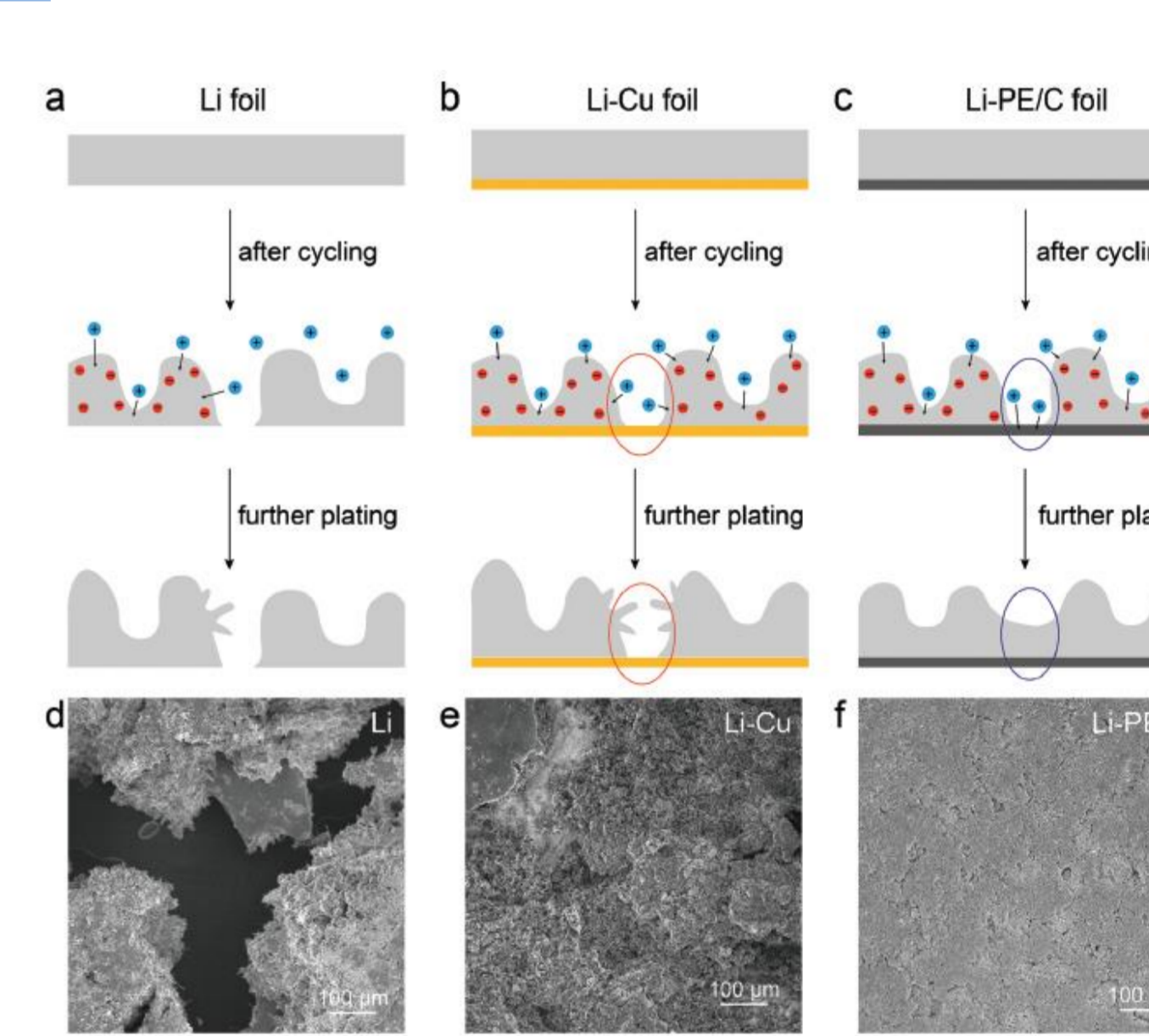


Fig. 7: Fading mechanism of the Li-metal negative electrodes. a-c) Schematic illustration of the fading mechanism for the a) Li, b) Li-Cu, and c) Li-PE/C negative electrodes. d-f) Ex situ top-view SEM micrographs of the d) Li, e) Li-Cu, and f) Li-PE/C negative electrodes after 20 cycles in LMB pouch cells employing NMC622-based positive electrodes (mass loading: 24 mg cm<sup>-2</sup>, N/P ratio: 2.4).

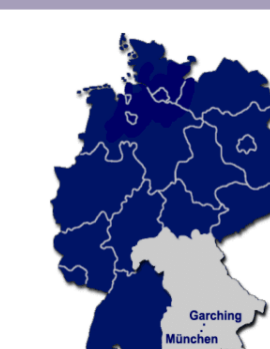
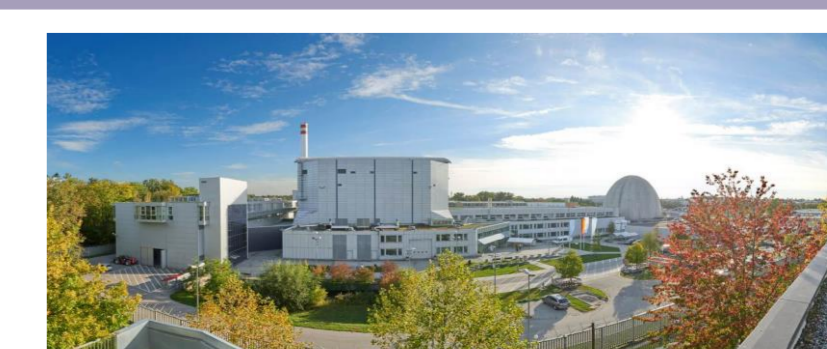
## Conclusion

We prepared an electronically conductive polymer film comprising two cheap components, polyethylene (PE) and carbon black (C), via a simple hot pressing method. This ultra-light PE/C polymer film was used as alternative current collector for lithium-metal batteries, allowing for a substantial reduction of the overall mass of the cell and, thus, a substantial increase in specific energy. Simultaneously, the use of this PE/C current collector yields a more homogeneous and denser lithium deposition compared to commonly employed copper current collectors. As a result, Li-PE/C||NMC<sub>622</sub> cells provide a greater cycle life than Li||NMC<sub>622</sub> or Li-Cu||NMC<sub>622</sub> cells and a substantially improved specific energy of 447 Wh kg<sup>-1</sup>, which is about 18% higher than the specific energy provided by the Li-Cu||NMC<sub>622</sub> cells with 378 Wh kg<sup>-1</sup> and close to the value of the cells comprising the neat lithium foil. Besides, the use of the PE/C current collector promises a significant cost reduction compared to metallic copper. Hence, we may anticipate that the results reported herein, which are shedding light on a commonly overlooked, but central challenge, provide a new avenue for the development of cost-competitive, high-performance LMBs.

## Acknowledgements

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