Supporting Information

On the Direct Correlation between the Copper Current Collector Surface Area and 'Dead Li' Formation in Zero Excess Li Metal Batteries

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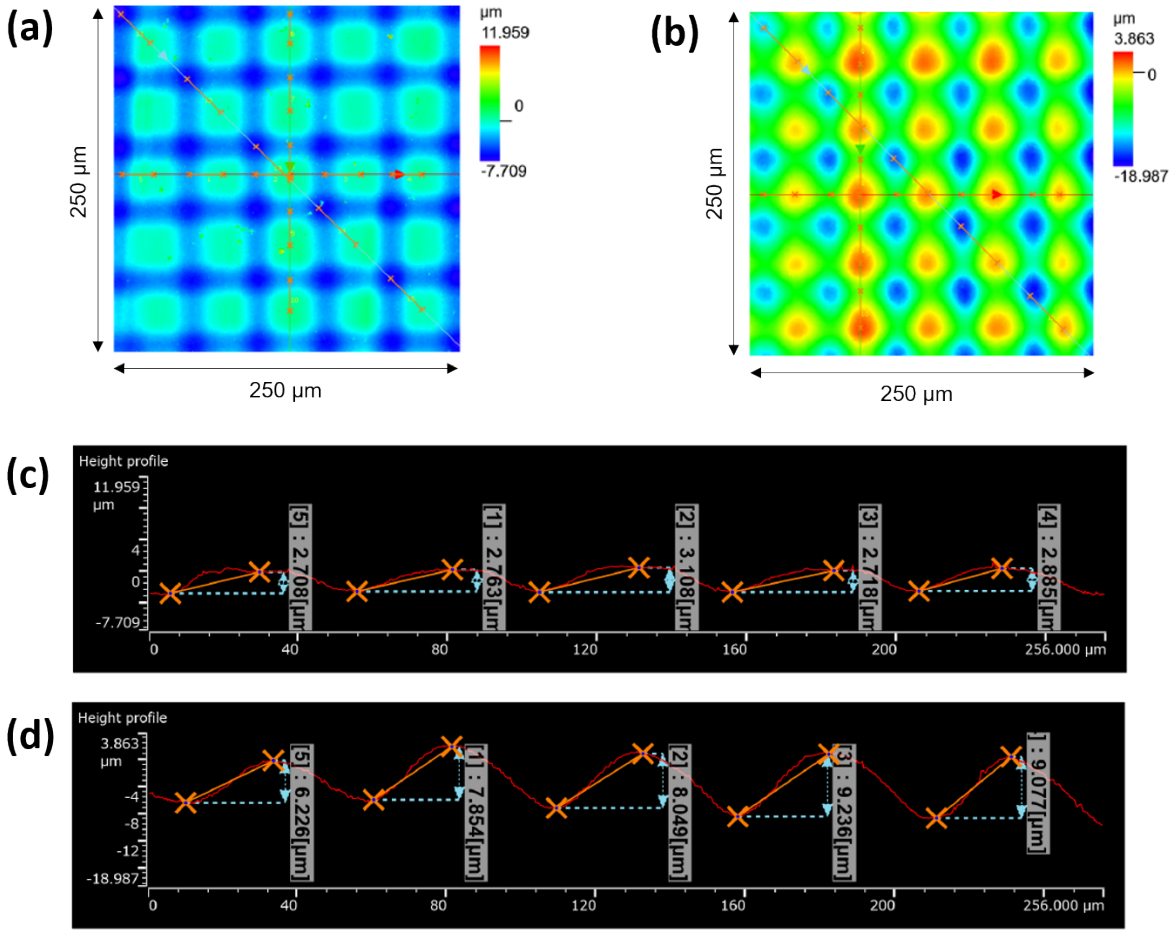


Figure S1. Confocal laser scanning microscope (CLSM) images showing the top-view colored contour profile of (a) Cu/B and (b) Cu/C. Depth profiles of (c) Cu/B and (d) Cu/C display different depths resulting from different laser powers and numbers of repetitions.

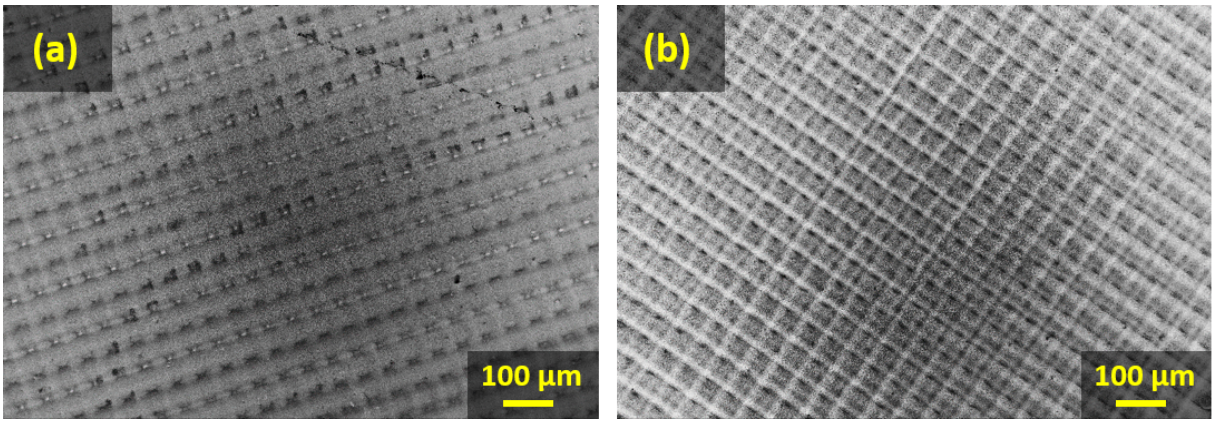


Figure S2. Low-magnification top-view SEM images showing the ordered tortuosity of (a) Cu/B and (b) Cu/C.

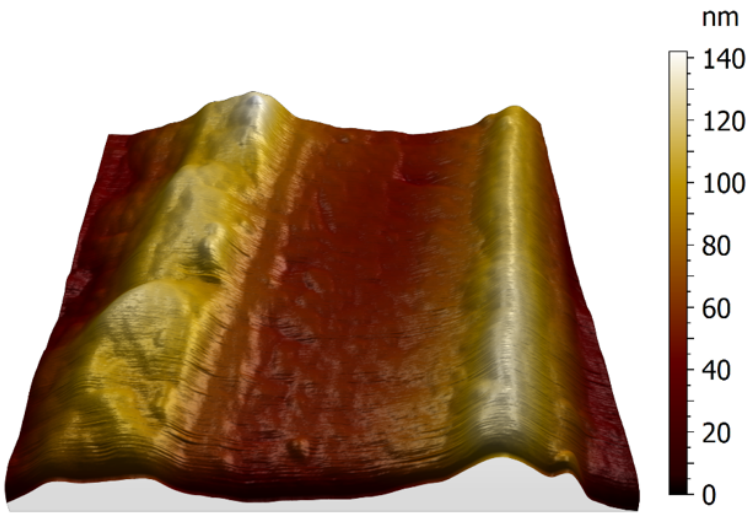
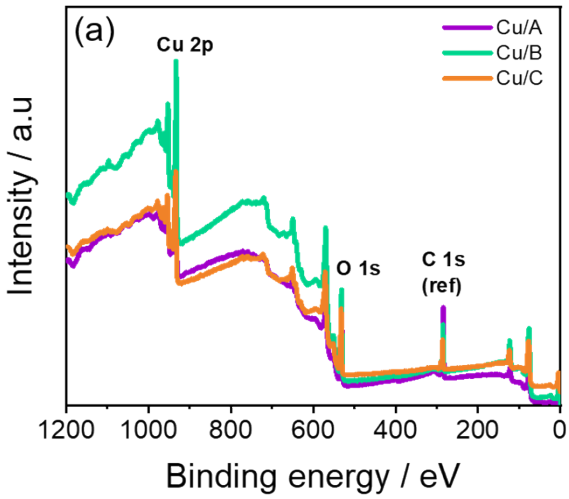


Figure S3. 3D topography images (2 µm × 2 µm) taken with AFM technique of Cu/A



(b)

Samples	Cu 2p / at%	O 1s / at%
Cu/A	41.42 ± 6.76	58.58 ± 6.76
Cu/B	45.40 ± 2.04	54.60 ± 2.04
Cu/C	46.86 ± 0.54	53.14 ± 0.54

Figure S4. (a) Full XPS core spectrum for all Cu CC samples. (b) Associated atomic concentration ratios of all Cu CC derived from the survey scan for Cu 2p and O 1s.

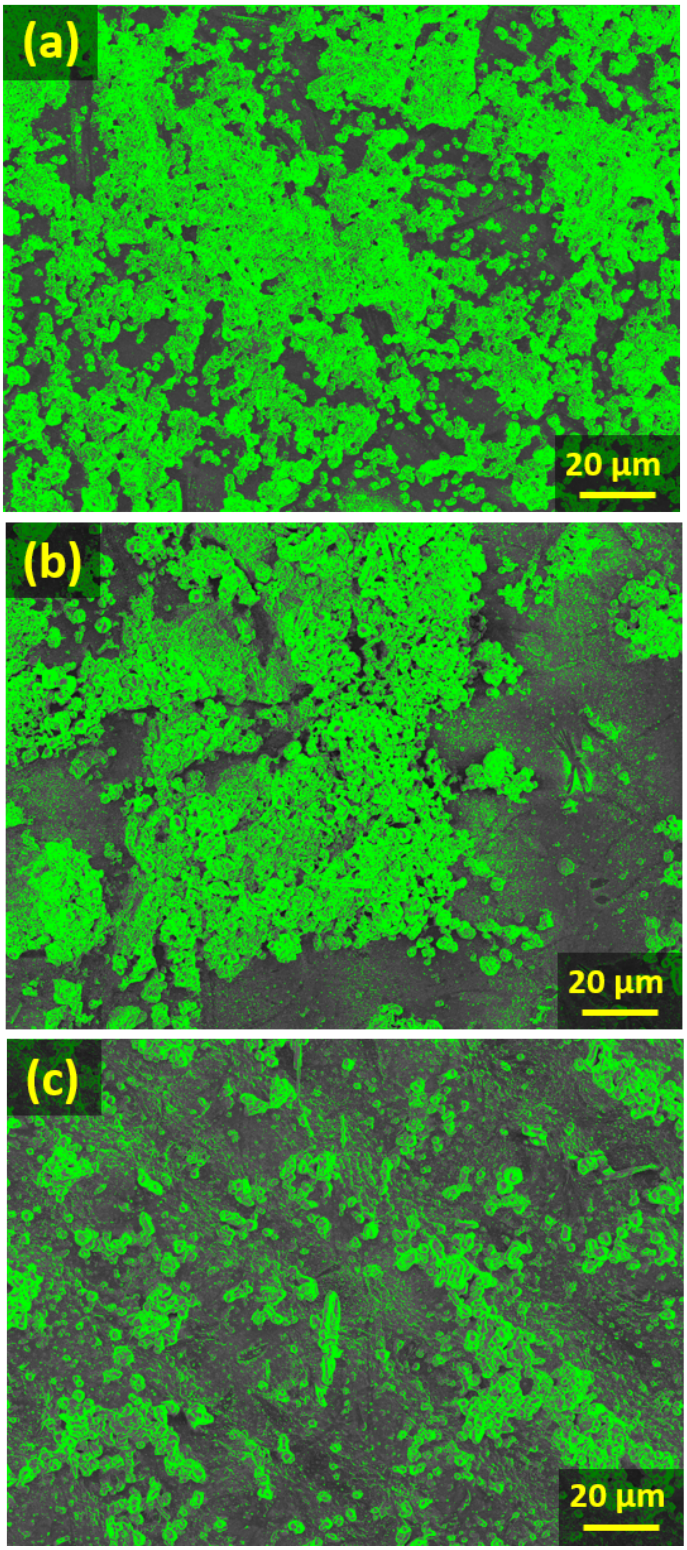


Figure S5. Low magnification top-view SEM images of the 'dead Li' on (a) Cu/A, (b) Cu/B, and (c) Cu/C. Green color indicates the accumulated 'dead Li'. All samples were cycled for 50 cycles at 0.5 mA cm^{-2} for 0.5 mAh cm^{-2}. The measurements were taken in a stripped state from Cu || Li metal cells.

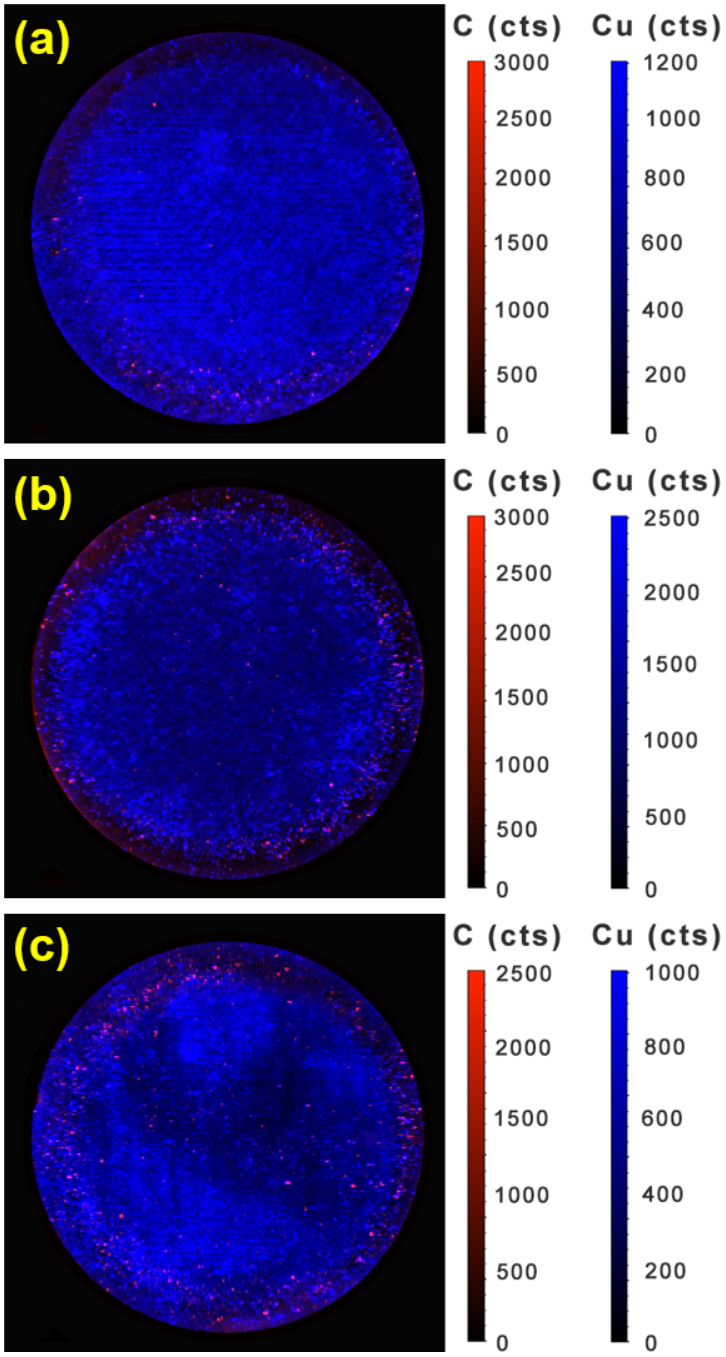


Figure S6. C and Cu element observation at the entire Cu CC electrode surface after 50 cycles in Cu || Li metal cells visualized and analyzed by LA-ICP-MS of (a) Cu/A, (b) Cu/B, and (c) Cu/C.

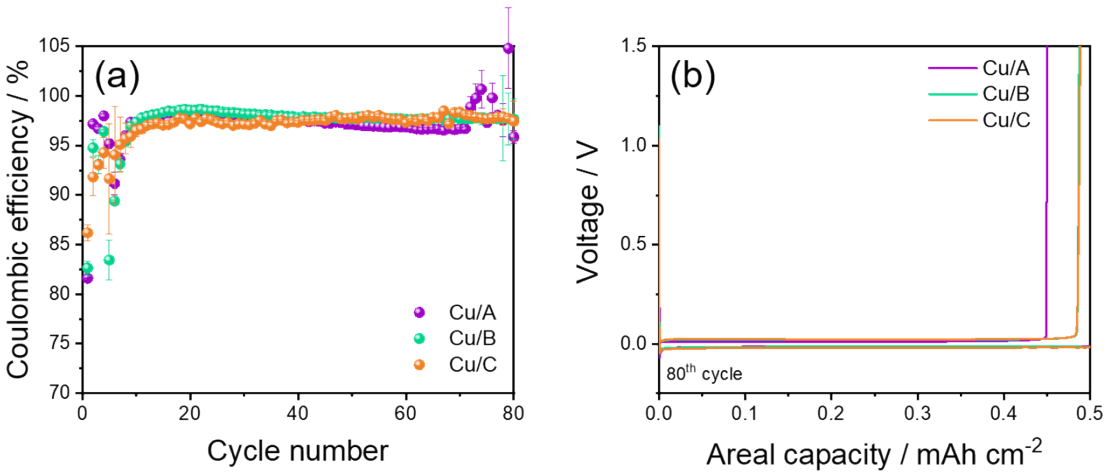


Figure S7. (a) Cycling performance of the three Cu || Li metal cells at 0.025 mA cm^{-2} for the first three cycles followed by 0.5 mA cm^{-2} for subsequent cycles. (b) Electrochemical voltage profile of the 80th cycle for lithium plating and stripping of all three samples. The areal capacity was 0.5 mAh cm^{-2}.

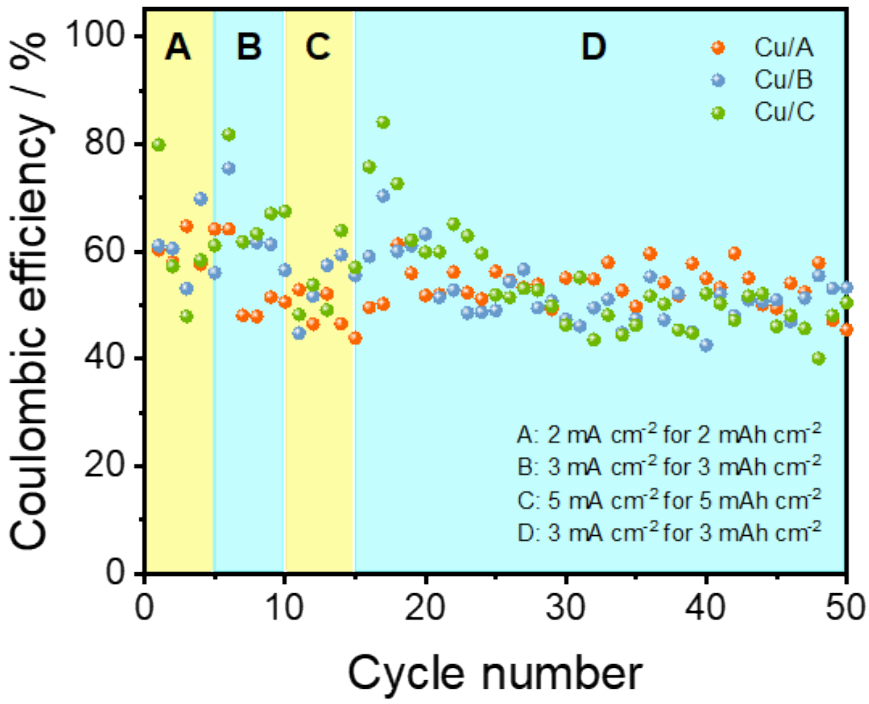


Figure S8. Electrochemical stability profile of the three Cu || Li metal cells at various high current densities and high areal capacities.

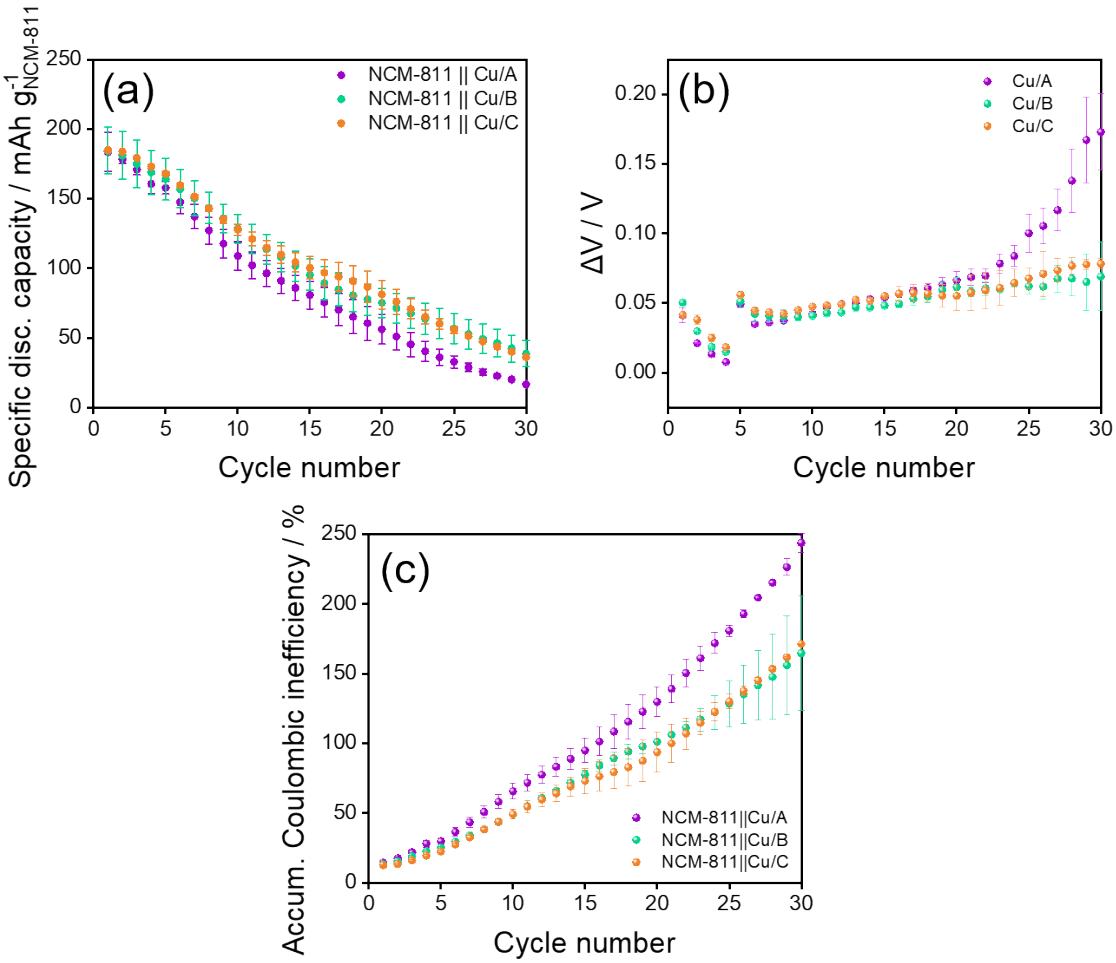


Figure S9. (a) Specific capacity *vs.* cycle number plots, (b) ΔV *vs.* cycle number plots, and (c) Accumulated Coulombic inefficiency *vs.* cycle number plots of all three Cu electrode samples against NCM-811 as positive electrodes within a cell voltage range of 2.8−4.2 V (at 0.33C).

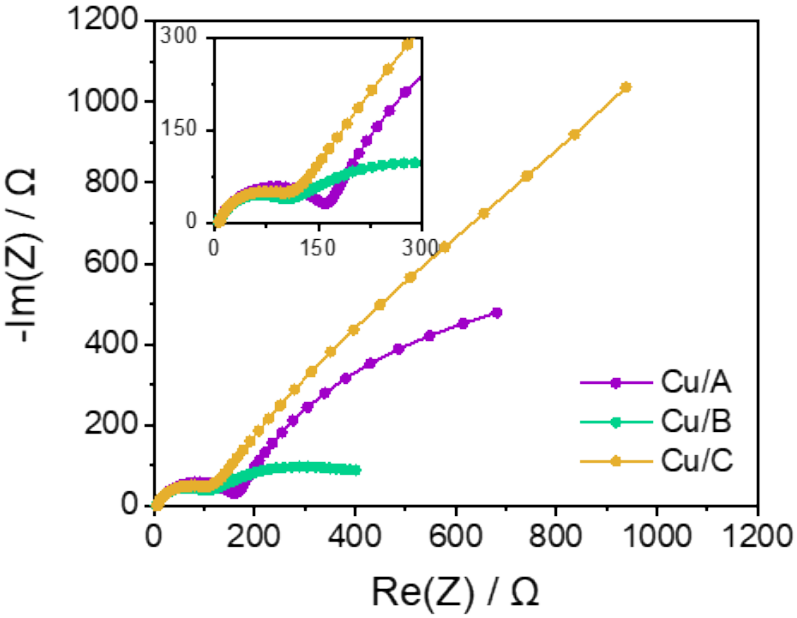


Figure S10. EIS spectra of all samples in Cu || Cu symmetric cells. An amount of 0.5 mAh cm^{-2} of Li was deposited on one of the electrodes and then kept for rest for 24 h.