

## Supporting Information

### Submicron Interfacial Layers for Nanoscale Control of Lithium Deposition in Surface-Engineered Current Collectors

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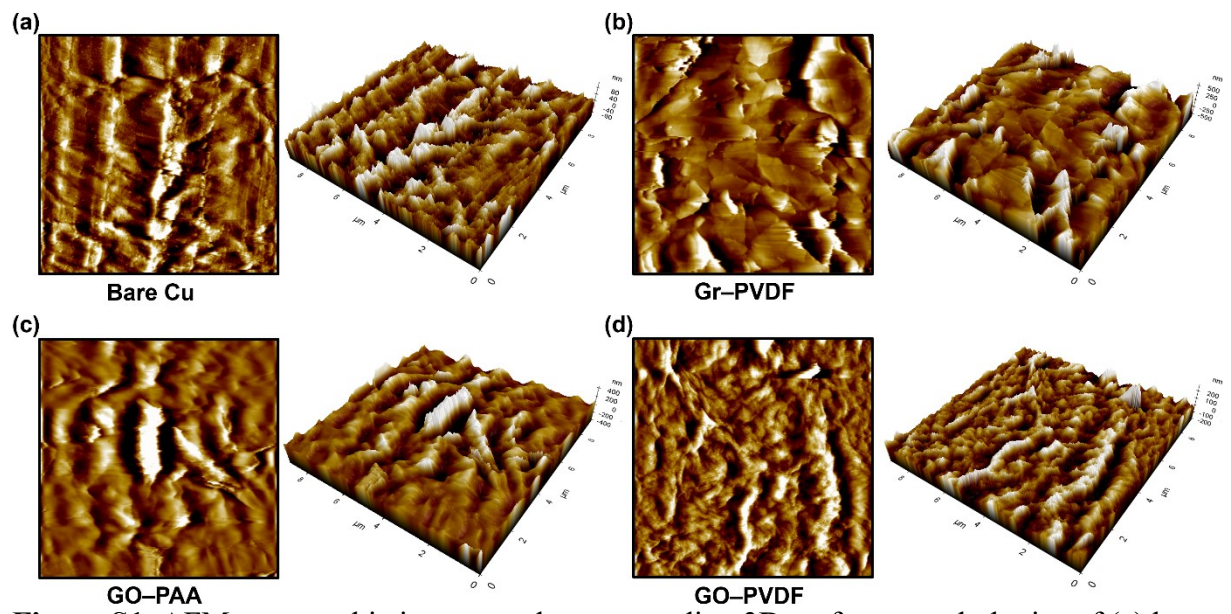
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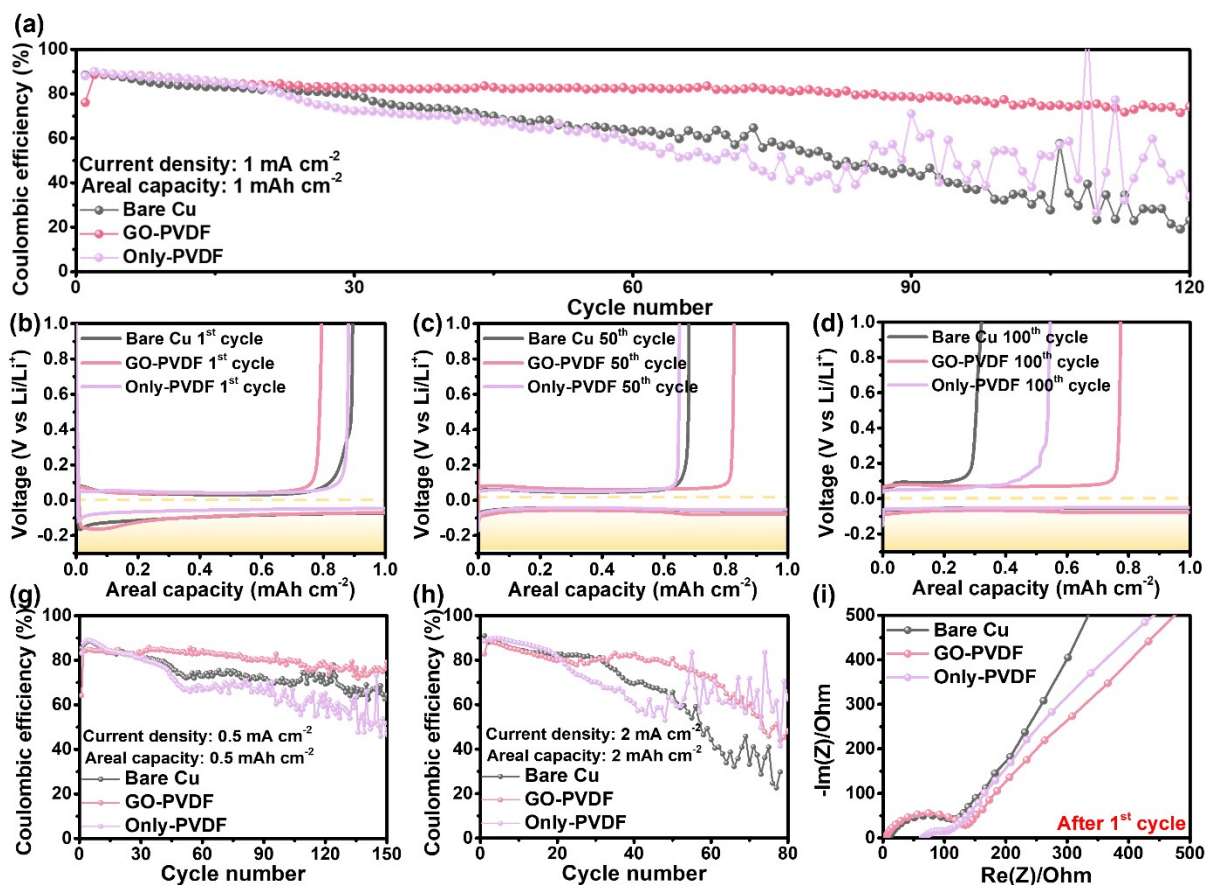
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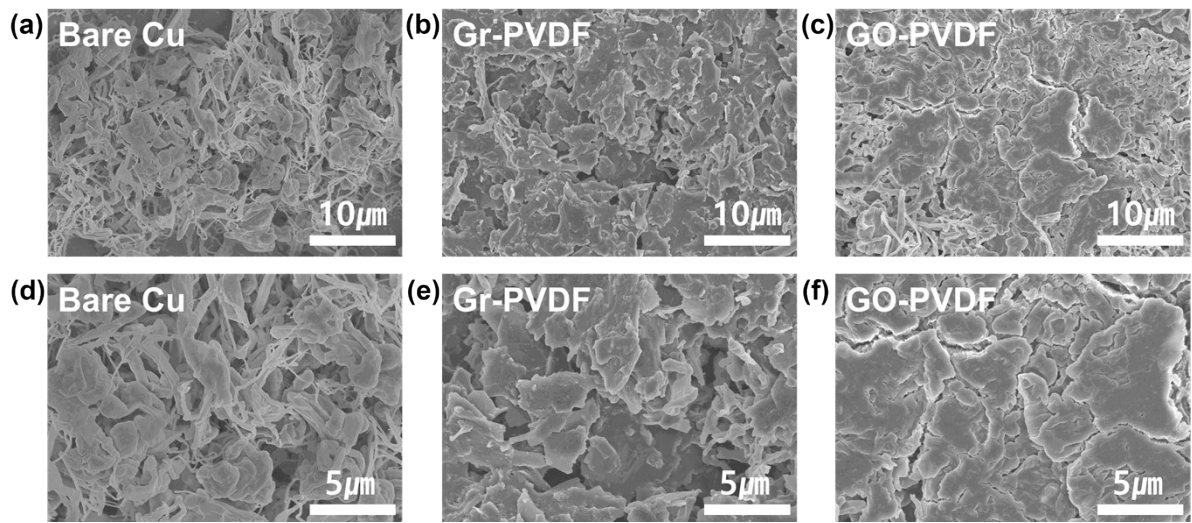
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**Figure S1.** AFM topographic images and corresponding 3D surface morphologies of (a) bare Cu, (b) Gr-PVDF, (c) GO-PAA, and (d) GO-PVDF.



**Figure S2.** (a) Cycling performance of Li||Cu half cells employing bare Cu, GO–PVDF, and Only-PVDF at a current density of  $1.0 \text{ mA cm}^{-2}$  and an areal capacity of  $1.0 \text{ mAh cm}^{-2}$ . Galvanostatic charge–discharge profiles at the (b) 1st, (c) 50th, and (d) 100th cycles. Cycling performance at (g)  $0.5 \text{ mA cm}^{-2}$  with an areal capacity of  $0.5 \text{ mAh cm}^{-2}$  and (h)  $2.0 \text{ mA cm}^{-2}$  with an areal capacity of  $2.0 \text{ mAh cm}^{-2}$ . (i) Electrochemical impedance spectroscopy (EIS) spectra acquired after the 1st cycle.



**Figure S3.** (a–c) Low-magnification SEM images and (d–f) high-magnification SEM images of the initially deposited Li on bare Cu, Gr–PVDF and GO–PVDF current collectors after Li plating at  $0.5 \text{ mA cm}^{-2}$  and  $0.5 \text{ mAh cm}^{-2}$ .

	Li deposit width ( $\mu\text{m}$ )	$R_{\text{ct}}$ after 1 cycle( $\Omega$ )	$R_{\text{ct}}$ after 100 cycles ( $\Omega$ )
Bare Cu	0.61	113	381
Gr-PVDF	1.37	36	175
GO-PAA	1.28	102	171
GO-PVDF	2.27	134	142

**Table S1.** Correlation between Li growth morphology and charge-transfer resistance evolution during cycling.