Supplementary Information

Formation sequence of solid-electrolyte-interphases and

impacts on lithium deposition and dissolution on copper: an

in-situ atomic force microscopic study

Wei-Wei Wang,^a Yu Gu,^a Hao Yan,^a Kai-Xuan Li,^a Zhao-Bin Chen,^a Qi-Hui Wu,^b Christine Kranz,^c Jia-Wei Yan,^{*a} and Bing-Wei Mao^{*a}



Fig. S1 The experimental current-time transient at -100 mV and the corresponding theoretical current-time transient j_{3D-DC} and j_{SEI} by nonlinear curve fitting.



Fig. S2 Linear sweep voltammetry from 2 V down to 0.1 V of Cu electrode in 0.5 M LiTFSI/Py₁₄TFSI-Py₁₄FSI (1:1, v/v) electrolyte containing 1300 ppm H₂O showing the formation of SEI-Cu-Isv. Scan rate: 10 mV s⁻¹. The SEI formation is almost completed in the first negative potential sweeping prior to Li OPD, and the reduction of electrolyte at the 1.33 V becomes almost absent and the background of the UPD peak significantly reduced in the subsequent two sweepings.



Fig. S3 XPS depth profiles of S 2p, N 1s, Cu 2p and Li 1s elements recorded after different time of argon ion sputtering at (a) SEI-Cu-lsv, (b) SEI-Cu-step and (c) SEI-Li.



Fig. S4 (a and b) AFM images of pristine Cu(100) electrode at 2 V in 0.5 M LiTFSI/Py₁₄TFSI-Py₁₄FSI. Scale bars are 6 μ m in (a) and 2 μ m in (b).



Fig. S5 In-situ AFM images of different Li deposition capacity (a, b) after SEI-Cu-Isv formation and (e, f) potential stepping from 2 V to -100 mV. (c, g) Cross section profiles of particles in (a, b) and (e, f), respectively. (d, h) Development of height and width of a growing Li nucleus marked by circle in (a) and (f), respectively. Scale bars are 1 μ m in (a) and 2 μ m in (b, e and f).