

Supporting Information

**Morphological Control of Electrodeposited Lithium Metal *via* Seeded
Growth: Stepwise Spherical to Fibrous Lithium Growth**

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Fig. S1

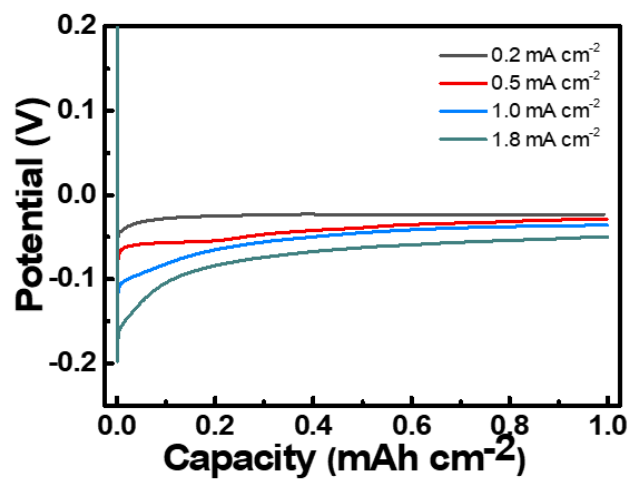


Fig. S1. Voltage profiles of Li plating on Cu at current densities of 0.2, 0.5, 1.0, and 1.8 mA cm⁻² deposited to 1 mAh cm⁻².

Fig. S2

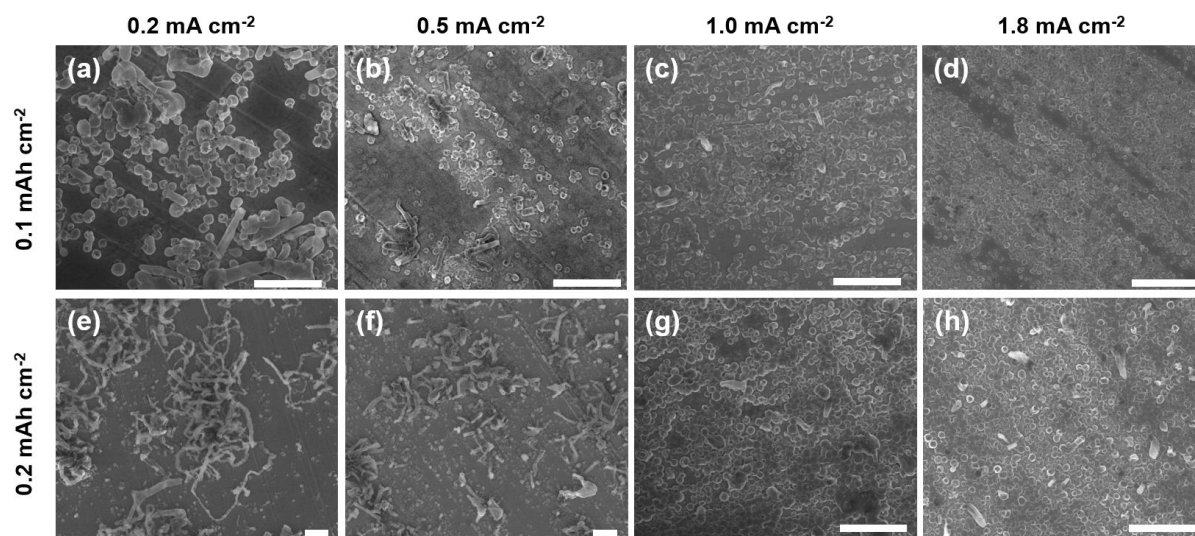


Fig. S2. SEM images of Li-deposited Cu foils plated to the capacity of 0.1 and 0.2 mAh cm⁻² at different current densities. (a–d) for 0.1 and (e–f) for 0.2 mAh cm⁻² at 0.2 (a,e), 0.5 (b,f), 1.0 (c,g), and 1.8 mA cm⁻² (d,h), respectively. All scale bars are 10 μm.

Fig. S3

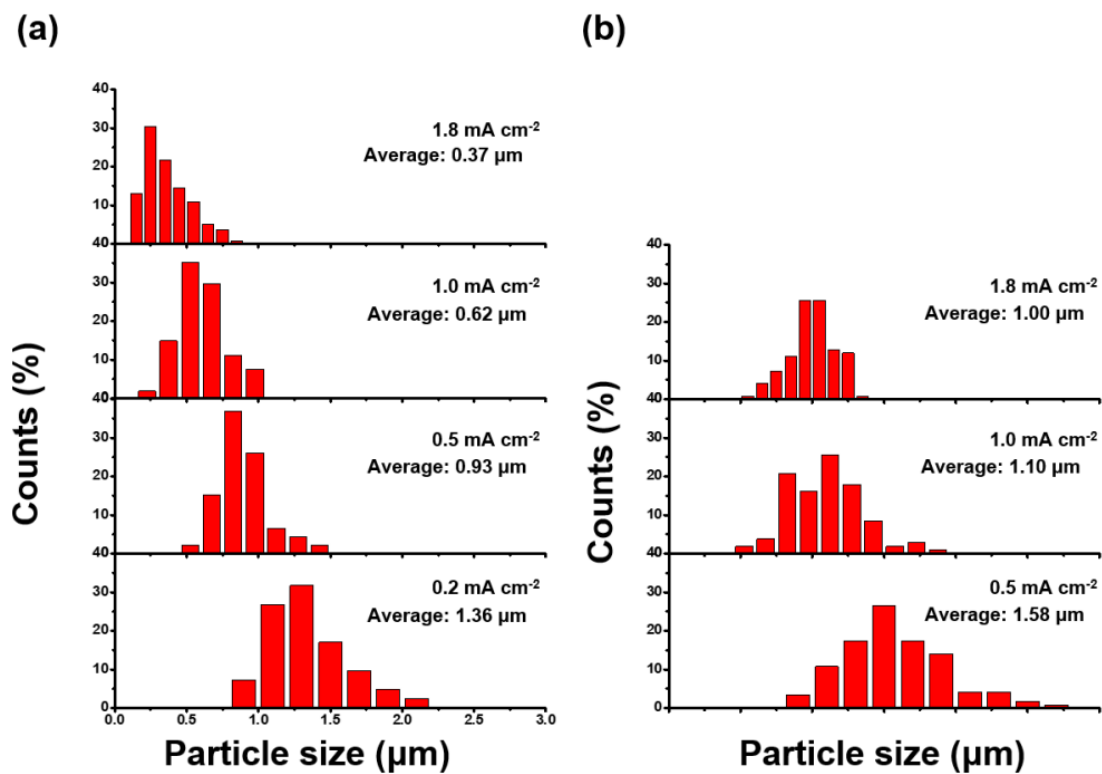


Fig. S3. Size distribution of spherical Li particles deposited on Cu foil for (a) 0.1 and (b) 0.2 mAh cm⁻². Because the measurement of diameter for particles underneath the top surface in multilayer Li deposit is unclear, only the Li particles on the top layer were measured at 1.0 and 1.8 mA cm⁻² current density for capacity of 0.2 mAh cm⁻².

Fig. S4

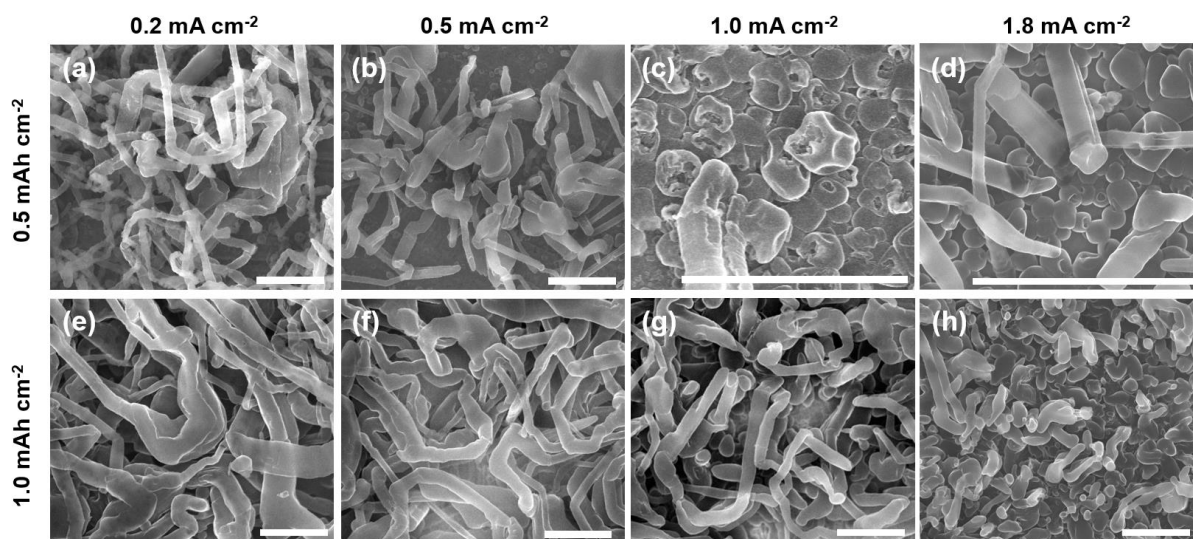


Fig. S4. SEM images of Li-deposited Cu foils plated to the capacity of 0.5 and 1 mAh cm⁻² at different current densities. (a–d) for 0.5 and (e–f) for 1.0 mAh cm⁻² at 0.2 (a,e), 0.5 (b,f), 1.0 (c,g) and 1.8 mA cm⁻² (d,h), respectively. All scale bars are 10 μm.

Fig. S5

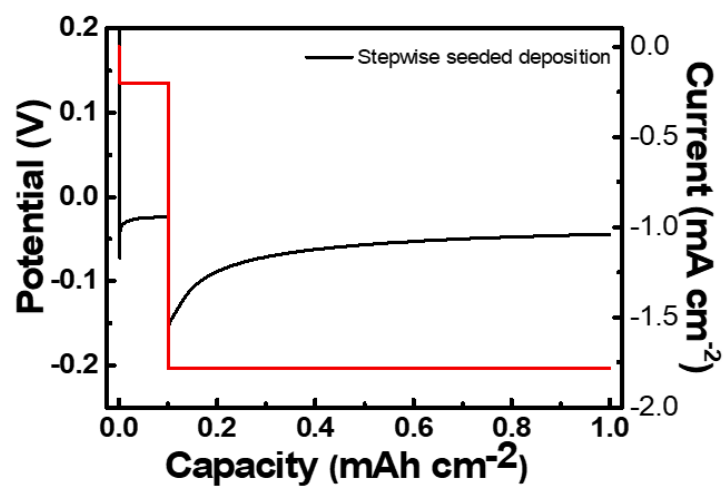


Fig. S5. Voltage profile of stepwise seeded Li deposition on Cu to a capacity of 1 mAh cm^{-2} .

First, 0.2 mA cm^{-2} current was applied to a capacity of 0.1 mAh cm^{-2} and then 1.8 mA cm^{-2} current for the remaining 0.9 mAh cm^{-2} .

Fig. S6

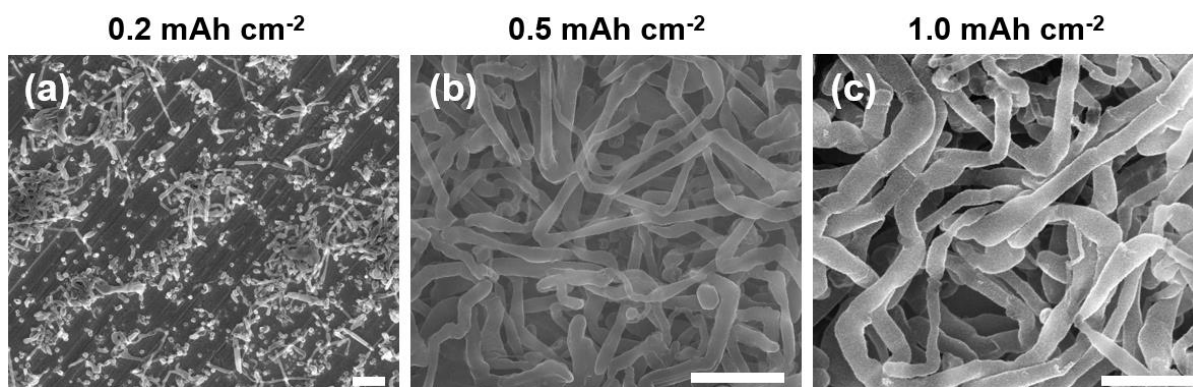


Fig. S6. SEM images of Li deposits on Cu foil at different capacity of (a) 0.2, (b) 0.5, and (c) 1.0 mAh cm⁻² deposited through the two-step seeded deposition. First, 0.2 mA cm⁻² current was applied to a capacity of 0.1 mAh cm⁻² and then 1.8 mA cm⁻² current for the remaining 0.1, 0.4, and 0.9 mAh cm⁻² for (a), (b), and (c), respectively

Fig. S7

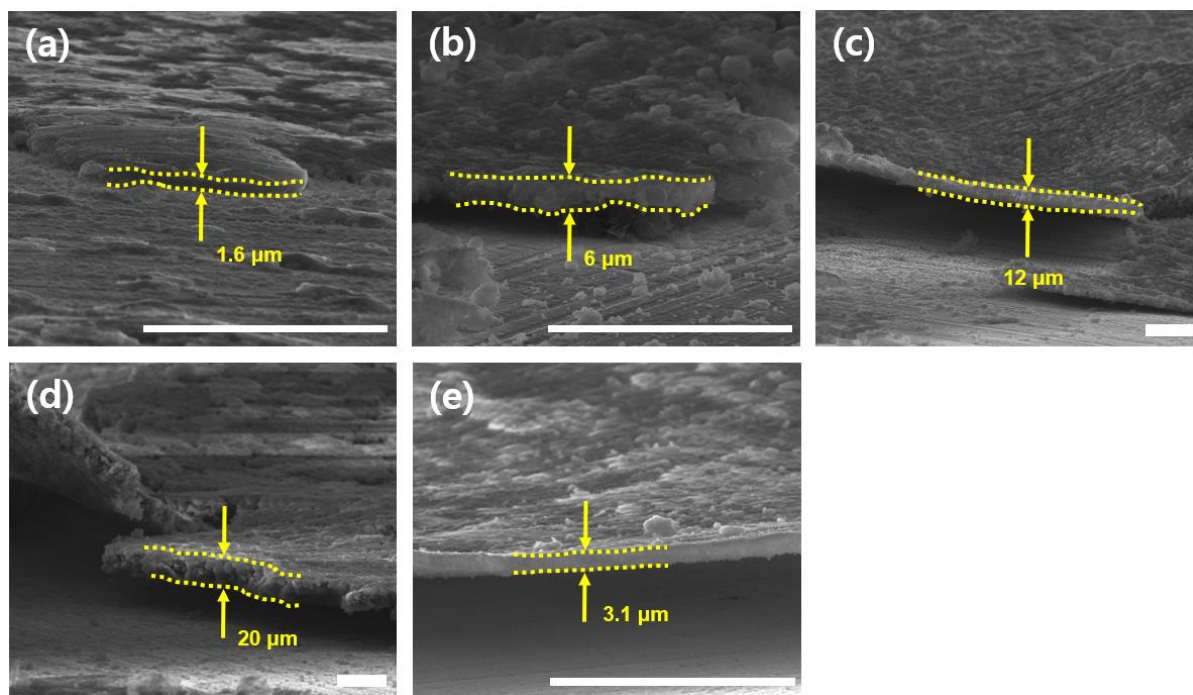


Fig. S7. SEM images of Cu electrode after 5 cycles of Li plating and stripping. Li was deposited at varied single current densities (0.2 (a), 0.5 (b), 1.0 (c) and 1.8 mA cm⁻² (d)) and by the stepwise seeded growth (e) and stripped at a fixed single current density of 1 mA cm⁻² with the curtailing capacity of 1 mAh cm⁻². The thickness of residue layer increases as the current density increase at single current depositions. All the scale bar is 40 μm.

Fig. S8

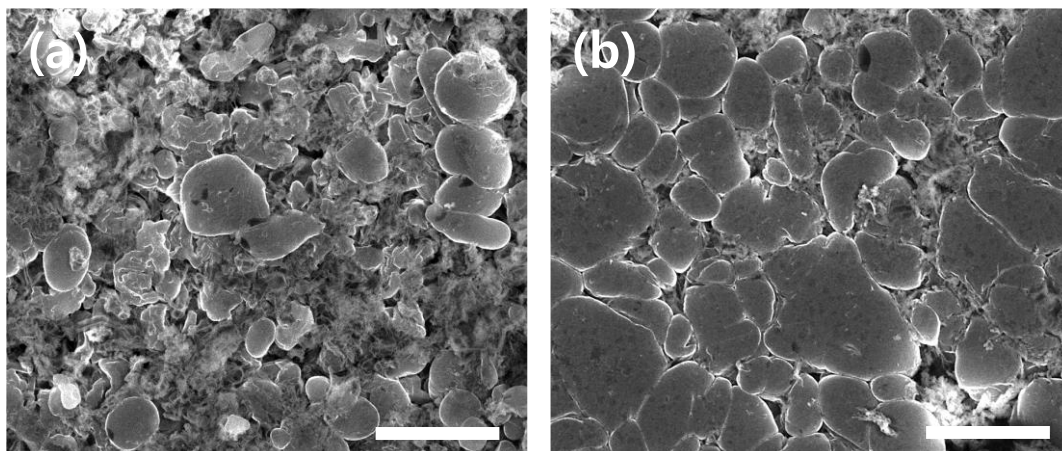


Fig. S8. SEM images of Li deposits after 10 cycles of Li deposition/stripping and deposition. (a) 1 mA cm^{-2} single current and (b) 0.2 to 1.8 mA cm^{-2} stepwise seeded deposition. All the scale bar is $10 \text{ }\mu\text{m}$.

Fig. S9

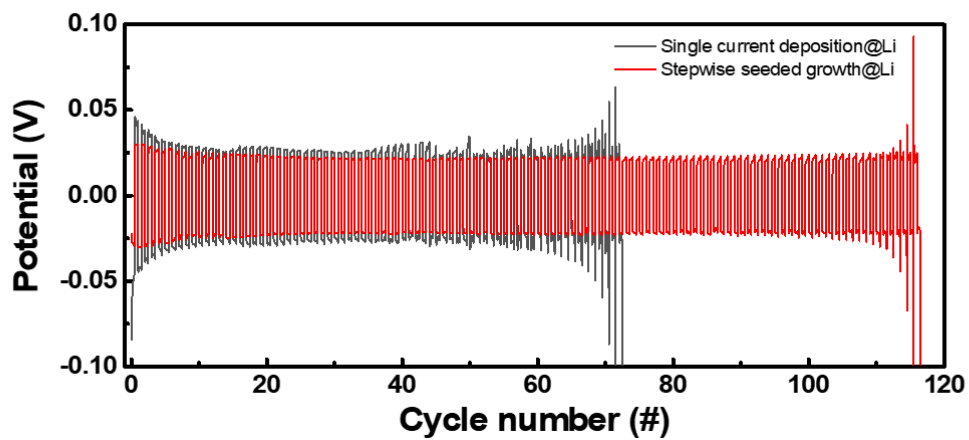


Fig. S9. Cycling performance of symmetric cells using a pair of single current deposited Li and stepwise seeded deposited Li electrodes. The cycling was at 1 mA cm^{-2} current to the capacity of 0.5 mAh cm^{-2} .

Fig. S10

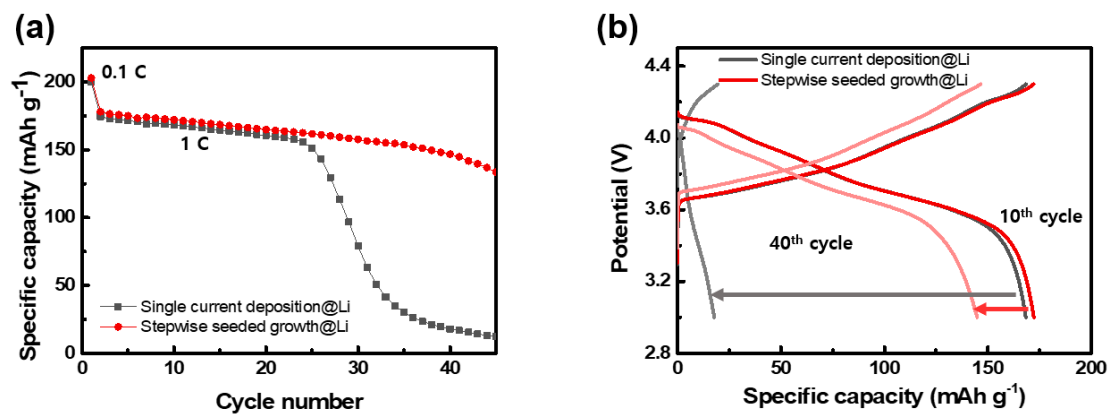


Fig. S10. (a) Variation of the specific capacity of NCM cathodes with cycle number paired with single current deposited Li and stepwise seeded deposited Li electrode. (b) Charge-discharge curves of NCM/deposited Li full cell at 10th and 40th cycle.

Fig. S11

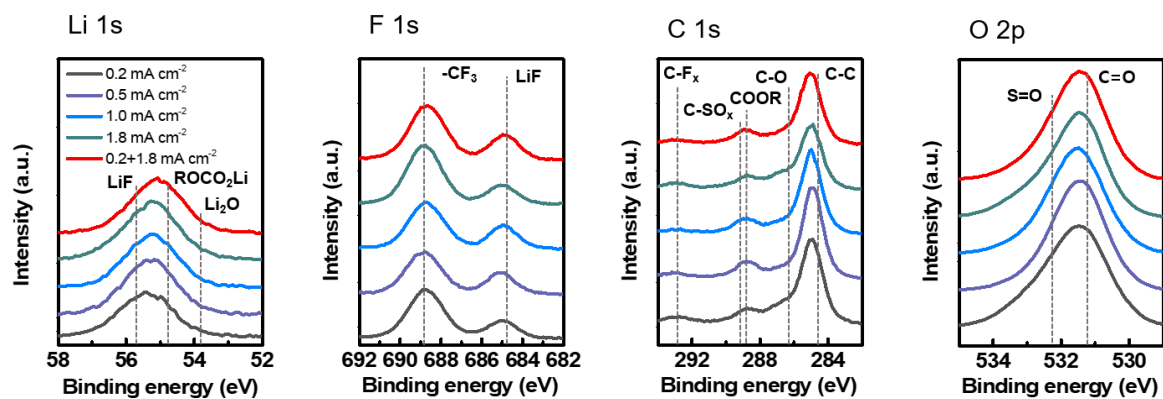


Fig. S11. XPS spectra of Cu electrode after 5 cycles of Li plating and stripping. Li metal was deposited at various single current densities (0.2 to 1.8 mA cm⁻²) and a stepwise (0.2 to 1.8 mA cm⁻²) current density and stripped at a fixed single current density of 1 mA cm⁻² with a curtailing capacity of 1 mAh cm⁻².

Fig. S12

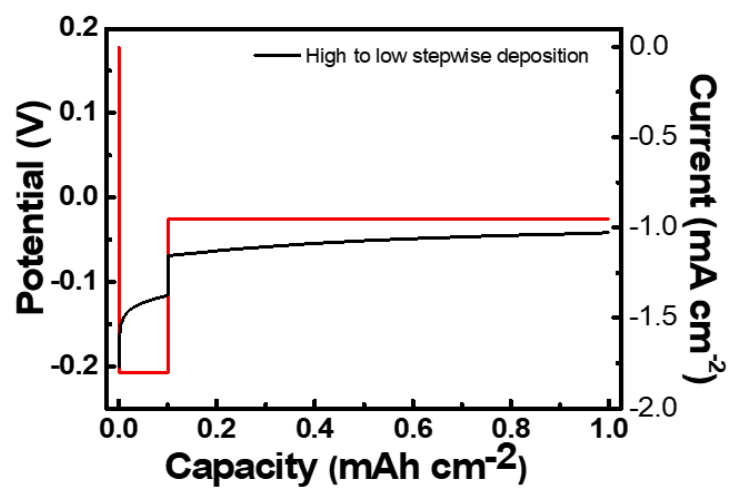


Fig. S12. Voltage profile of high to low stepwise Li deposition on Cu to a capacity of 1 mAh cm^{-2} . First, 1.8 mA cm^{-2} current was applied to a capacity of 0.1 mAh cm^{-2} and then 0.96 mA cm^{-2} current for the remaining 0.9 mAh cm^{-2} .

Fig. S13

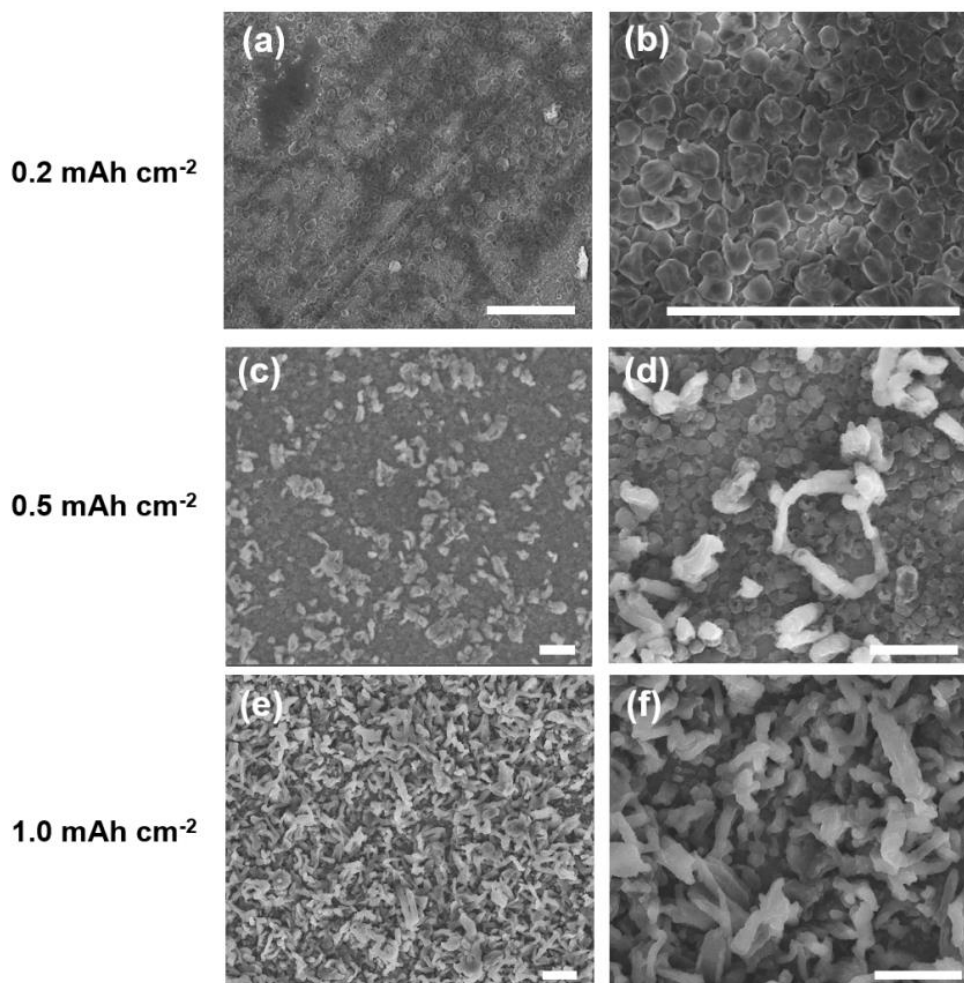


Fig. S13. SEM images of Li deposits on Cu foil at different capacities of (a, b) 0.2, (c, d), 0.5, and (e, f) 1.0 mAh cm⁻² deposited through the high to low stepwise seeded growth strategy. First, 1.8 mA cm⁻² current was applied to a capacity of 0.1 mAh cm⁻² and then 0.96 mA cm⁻² current was applied for the remaining 0.1, 0.4, and 0.9 mAh cm⁻² for (a,b), (c,d), and (e,f), respectively. All scale bars are 10 μm.

Fig. S14

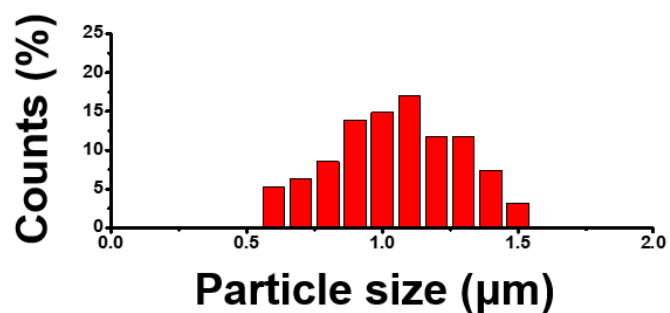


Fig. S14. Size distribution of spherical Li particles deposited on Cu foil for initial 0.1 mAh cm^{-2} at 1.8 mA cm^{-2} and subsequent 0.1 mAh cm^{-2} at 0.96 mA cm^{-2} (final total capacity is 0.2 mAh cm^{-2}). Because the measurement of diameter for particles underneath the top surface in multilayer Li deposit is unclear, only the Li particles on the top layer was measured.

Fig. S15

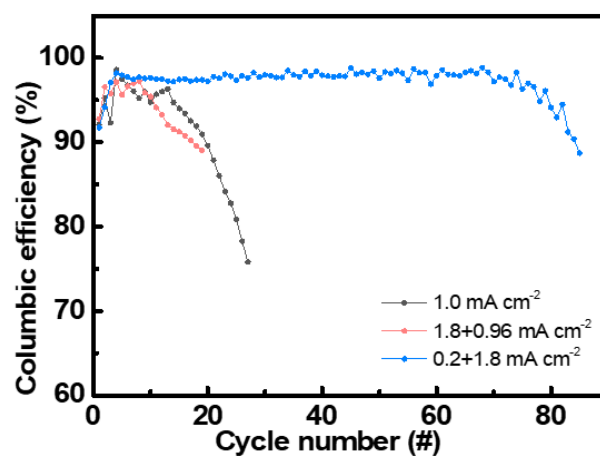


Fig. 15. Variation in coulombic efficiency (CE) during repeated Li plating and stripping through the single current and stepwise seeded deposition methods of high to low and low to high. Li metal was plated at a single current density of 1.0 (single current deposition) ,0.2–1.8 (low to high stepwise seeded deposition), and 1.8-0.96 mA cm⁻² (high to low stepwise deposition) mA cm⁻² and stripped at 1 mA cm⁻² current density. The final capacity was 1 mAh cm⁻² and average current density was 1 mA cm⁻² for all cases.

Fig. S16

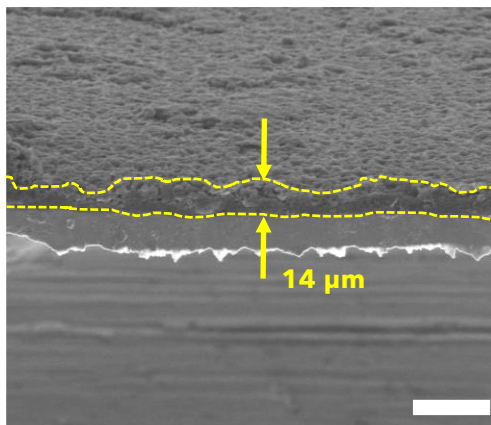


Fig. S16. SEM images of Cu electrode after 5 cycles of Li plating and stripping. Li was deposited by the high to low stepwise deposition (1.8 to 0.96 mA cm^{-2}) and stripped at a fixed single current density of 1 mA cm^{-2} with the curtailing capacity of 1 mAh cm^{-2} . Scale bar is 40 μm .

Fig. S17

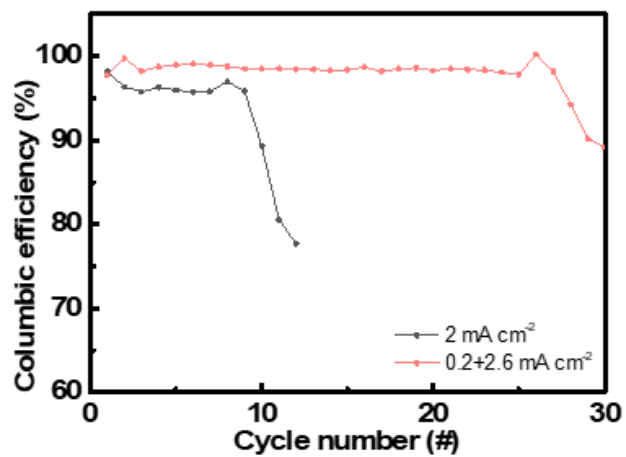


Fig. S17. Variation of coulombic efficiency during the repeated Li plating and stripping for Li electrodes deposited by constant single and two-step deposition. Li metal was plated at single current density of 2.0 mA cm⁻² and 0.2 to 2.6 mA cm⁻² (stepwise seeded deposition) to the capacity of 4 mAh cm⁻² and stripped at a 2 mA cm⁻² current density.