Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supplementary Information

The roles of nucleation and growth kinetics in determining Li metal morphology for Li metal batteries: columnar versus spherical growth

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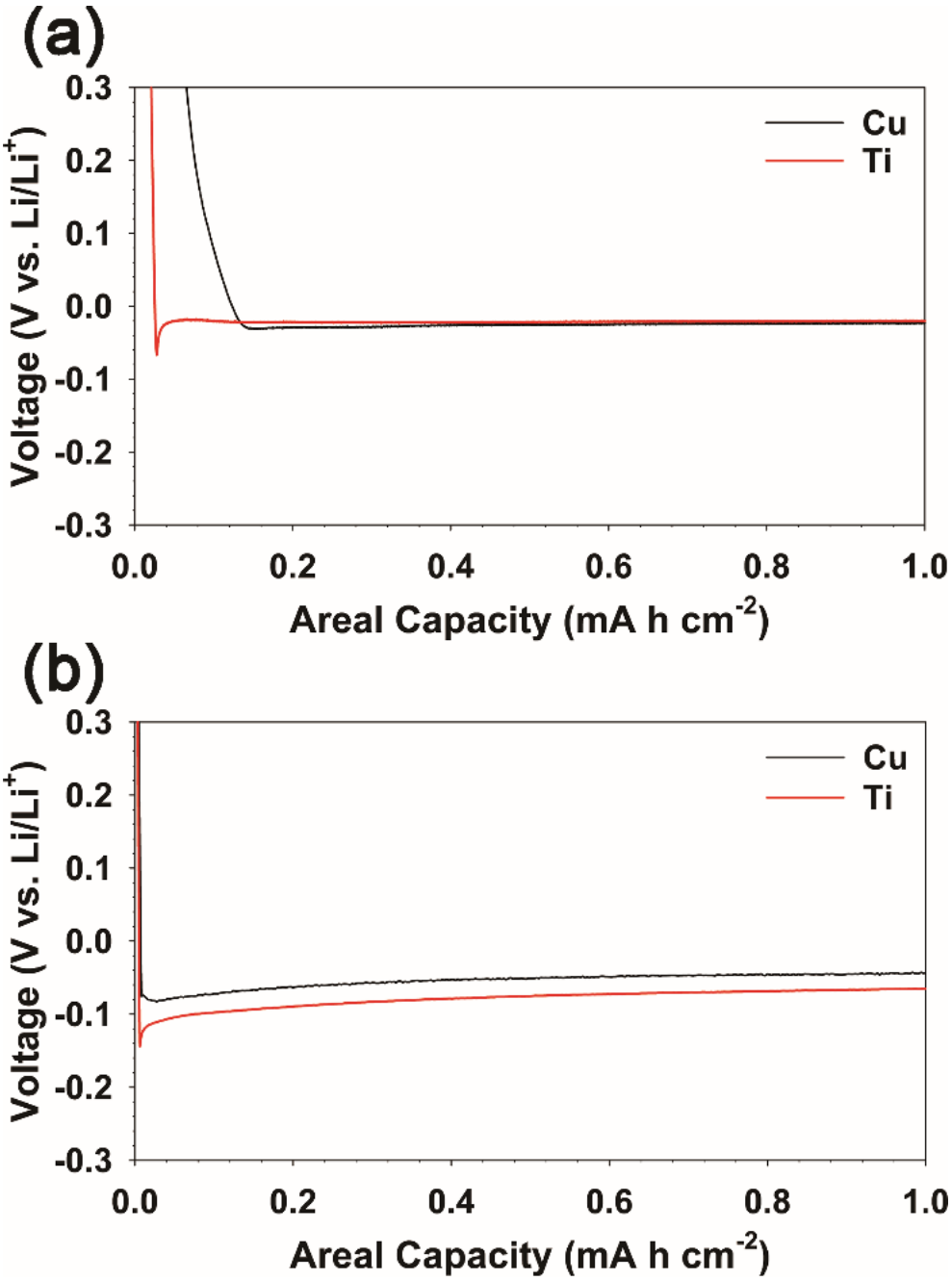

Fig. S1 Voltage profiles of Cu and Ti substrates during the initial nucleation and growth at a current density of (a) 0.1 mA cm^{-2} and (b) 1 mA cm^{-2} using three-electrode convective beaker-type cells.

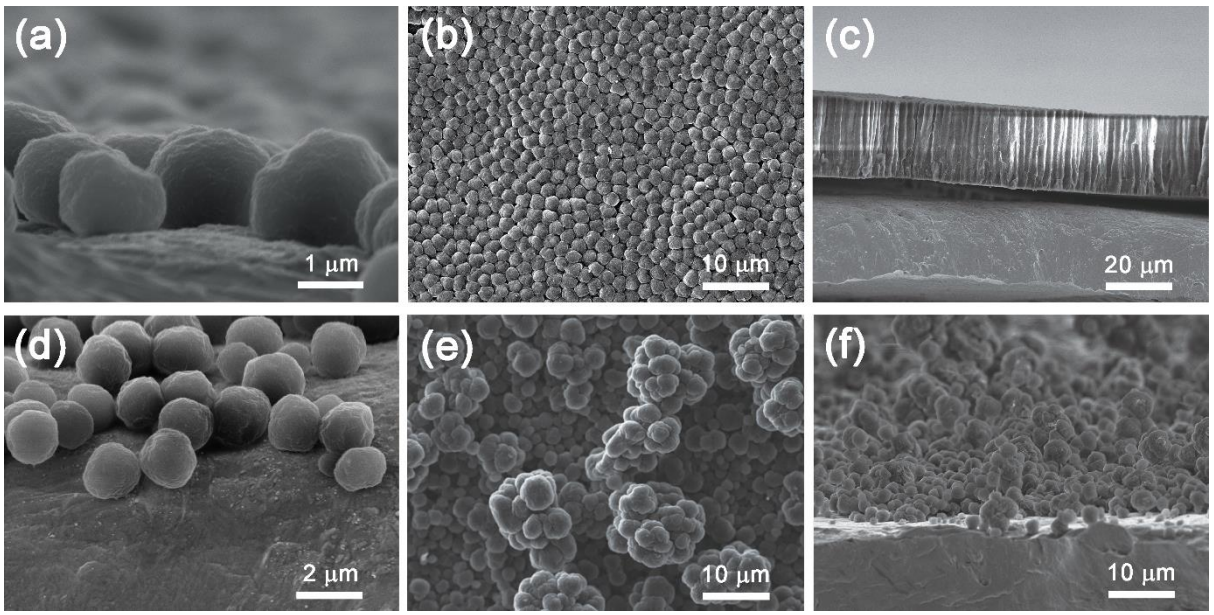

Fig. S2 SEM images of Li metal grown on the brass substrate for various areal capacities of (a) 0.2 and (b), (c) 6 mA h cm^{-2}. SEM images of Li metal grown on the SUS substrate for various areal capacities of (d) 0.2, (e), (f) 4 mA h cm^{-2}. (a, c, d, and f) Cross-sectional and (b and e) top-view SEM images. Electroplating was performed using $LiPF_6$ in EC/DEC at a current density of 0.1 mA cm^{-2}.

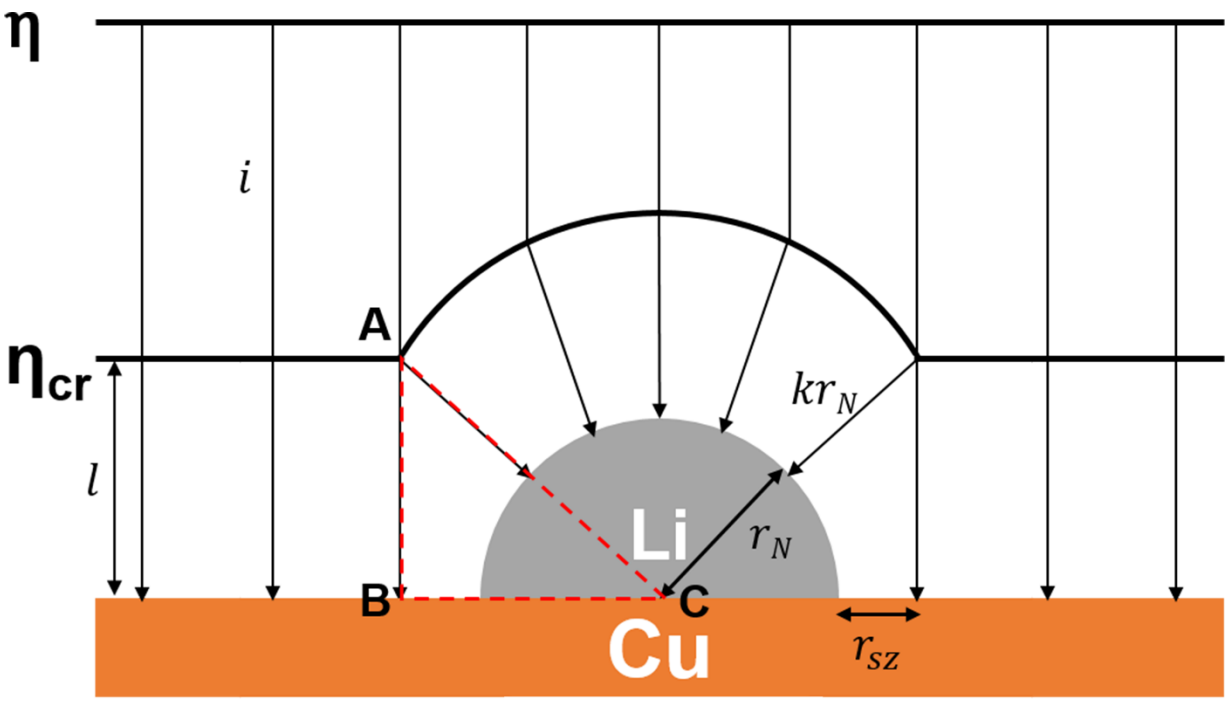

Fig. S3 Schematic illustration of current distribution during nucleation.[1]

At the end of nucleation exclusion zone, $l = kR$.

$$\overline{AB} = l = kr_N$$

$$\overline{AC} = r_N$$

$$\overline{BC} = \sqrt{\overline{AC}^2 - \overline{AB}^2} = r_N\sqrt{2k+1}$$

$$r_{sz} = r_N(\sqrt{2k+1} - 1)$$

$$r_{sz} = r_N(\sqrt{\frac{2l}{r_N} + 1} - 1)$$

$$\rho i l = \eta_{cr}$$

$$r_{sz} = r_N(\sqrt{\frac{2\eta_{cr}}{r_N \rho i} + 1} - 1)$$

References

1 S. Štrbac, Z. Rakočević, K. I. Popov, M.G. Pavlović and R. Petrović, *J. Serb. Chem. Soc.*, 1999, **64**, 483-493.

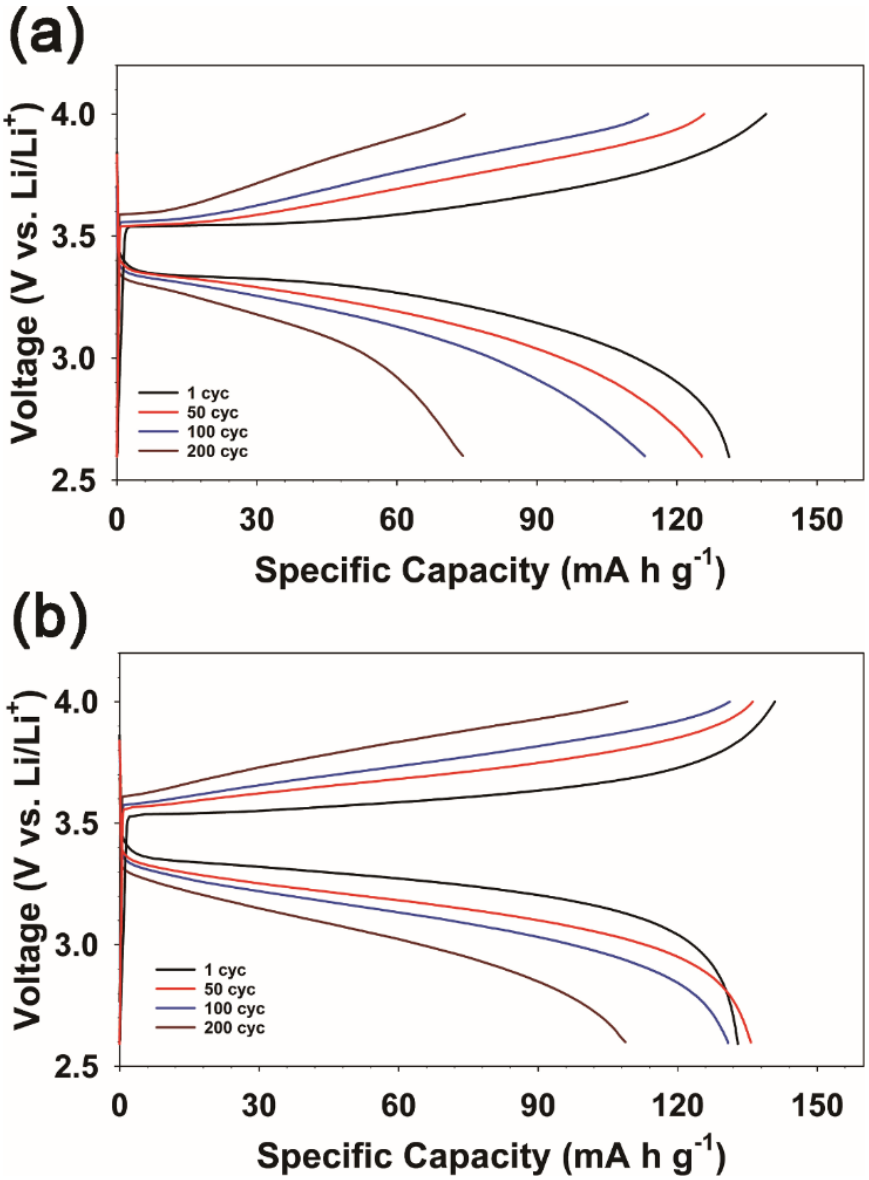

Fig. S4 Voltage profiles of the Li (20 μm) | $LiFePO_4$ full cells using (a) conventional and (b) columnar Li metal electrodes at a 1 C rate for various cycle numbers.

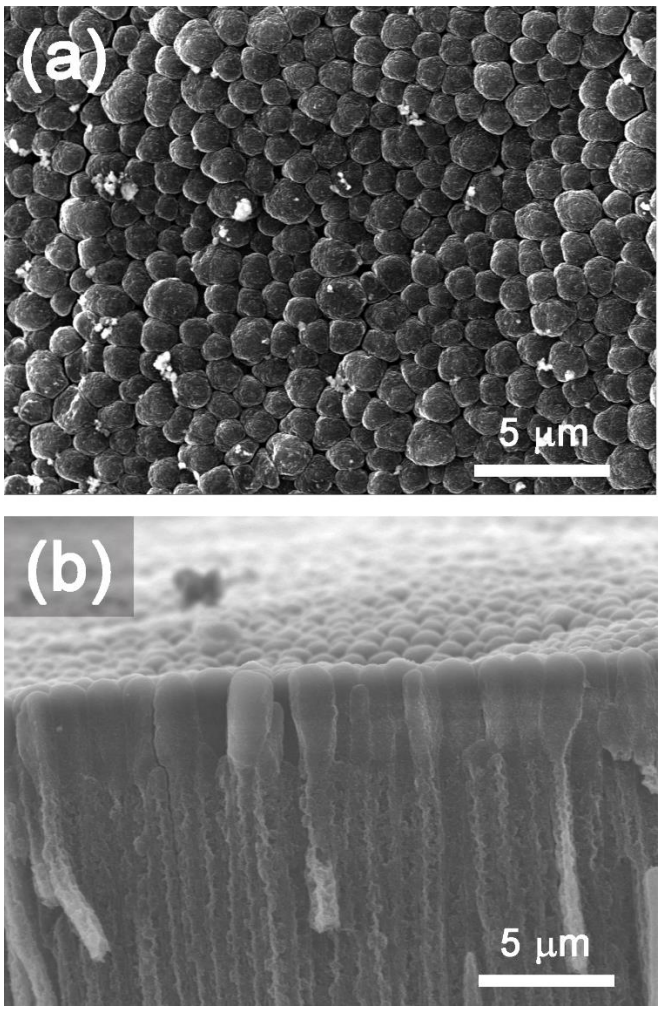

Fig. S5 (a) Top-view and (b) cross-sectional SEM images of the initially stripped columnar Li metal after the subsequent electroplating. Columnar Li metal was initially stripped at a current density of 0.1 mA cm^{-2} for the areal capacity of 2 mA h cm^{-2}. The stripped columnar Li metal was then electroplated at a current density of 0.1 mA cm^{-2} for the areal capacity of 2 mA h cm^{-2}. Stripping and electroplating were performed using convective beaker-type cells.

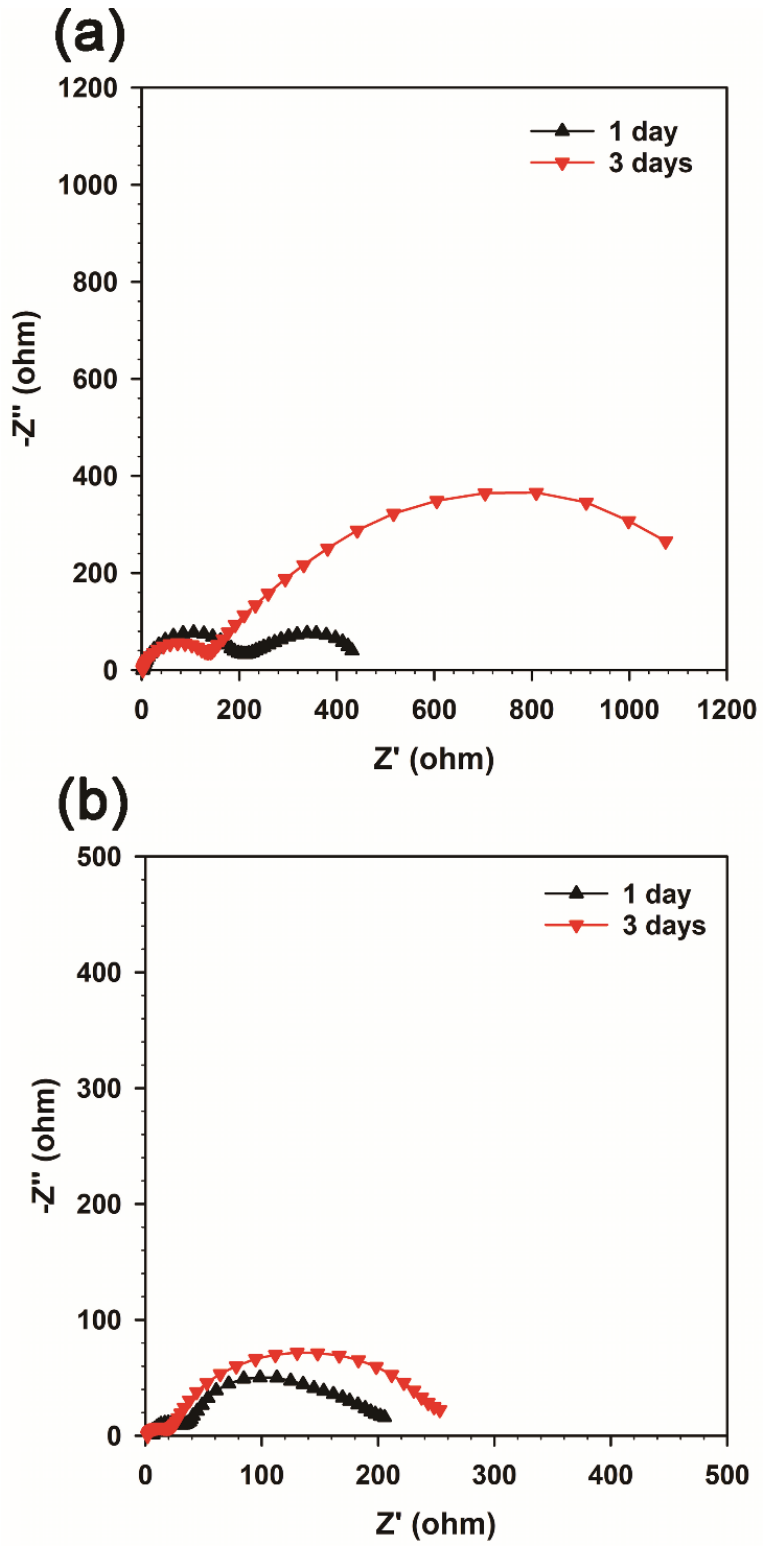

Fig. S6 Changes in the Nyquist plots of the Li/Li symmetric cells containing 1 M LiTFSI in DOL/DME with 1 wt% $LiNO_3$ for (a) conventional Li metal and (b) columnar Li metal during various aging periods. Both Li metal electrodes were 20 μm in thickness.

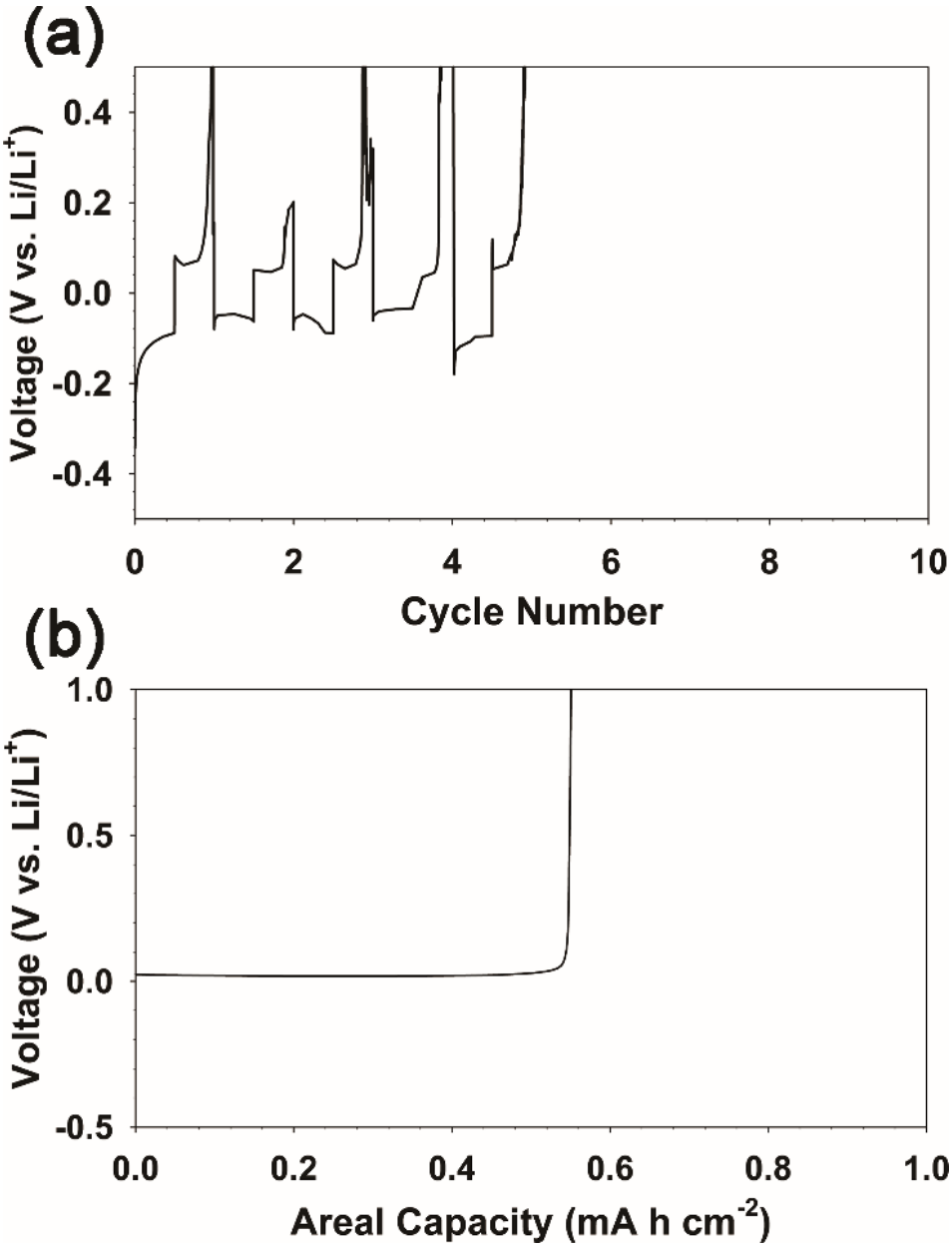

Fig. S7 (a) Voltage profile of the multilayers of Li metal spheres using the asymmetric cell of Li (Li metal spheres) | Li (conventional Li foil, 700 μm) at a current density of 5 mA cm^{-2} with the areal capacity of 1 mA h cm^{-2}. (b) Voltage profile of the multilayers of Li metal spheres for complete stripping at 0.1 mA cm^{-2}. The multilayers of Li metal spheres electrodes were fabricated using the same electroplating condition as Fig. 1f.