

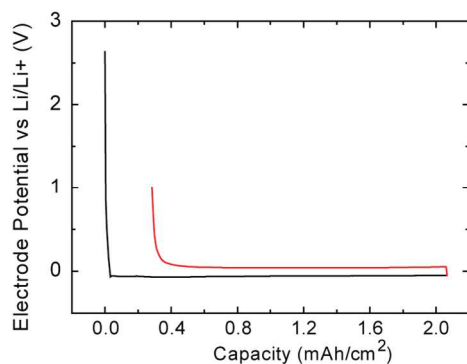
# An Ambient-air Stable Lithiated Anode for Rechargeable Li-ion Batteries with High Energy Density

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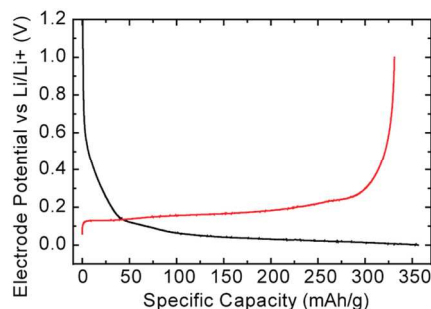
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## Initial cycle of lithium deposition and stripping



**Figure S1.** Electrodeposition and stripping of lithium film on copper foil. The electrolyte is 1 M LiTFSI in 1,3-dioxolane with 1wt% LiNO<sub>3</sub> and the current density is 1 mA/cm<sup>2</sup>. The corresponding coulombic efficiency is 85%.

## Initial cycle of artificial graphite electrode.



**Figure S2.** Voltage profile of the first cycle of an artificial graphite/Li cell. The composition of the artificial graphite electrode is 90% graphite and 10% PAA binder. The electrolyte is 1 M LiPF<sub>6</sub> in EC/DEC. The corresponding coulombic efficiency is 92%.

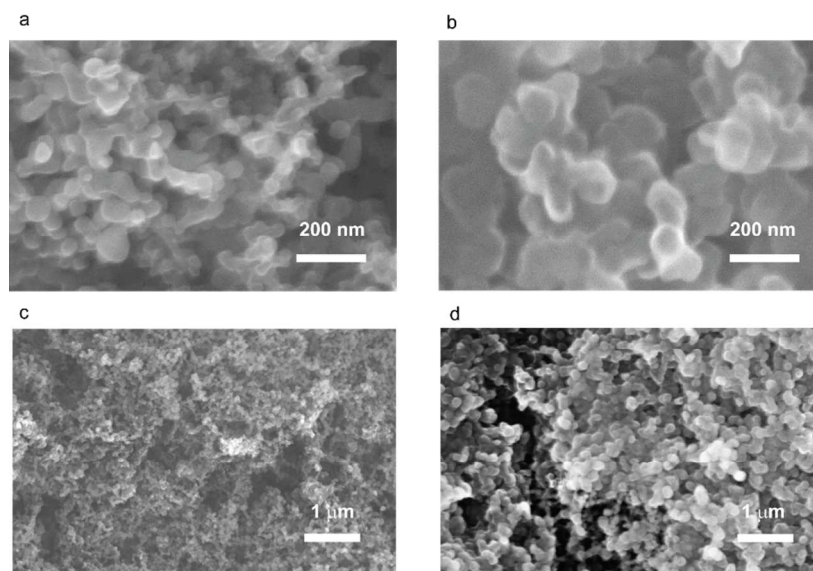
### Effect of Lithium loss on battery energy density.

Figure 4 shows that 40-60% of lithium electrodeposited on Cu is lost due to SEI formation and side reaction with solvent. To understand the effect of such loss on the energy density of a full cell, an NMC/Si full cell is used as an example, where Li protected by PMMA is used to compensate for the loss of lithium during SEI formation in the initial cycle. The capacity per area is assumed to be 3 mAh/cm<sup>2</sup>. If the initial coulombic efficiency is 80%, the initial Li loss due to SEI formation is 0.6 mAh/cm<sup>2</sup>. If 50% of the Li deposited is recovered in the anode/PMMA/lithium electrode, the total amount of Li needed is 1.2 mAh/cm<sup>2</sup>. The amount of materials needed is summarized as follows.

	NMC	Si	Lithium
Capacity (mAh/g)	150	1000	3860
Mass (mg)	20	3	0.31

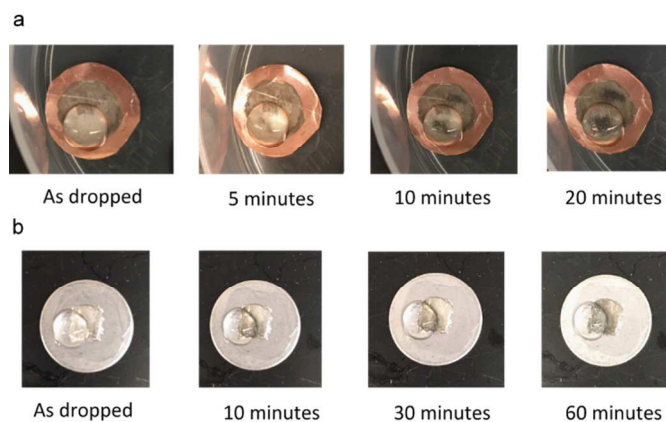
Therefore, the amount of lithium needed is only 1.5 wt% of the electrode, even if only 50% of Li is used. If Si is replaced by graphite, the percentage of lithium compared to all of the active materials will be even lower. Hence, 50% lithium loss does not affect the energy density noticeably.

### Prelithiated Si electrode



**Figure S3.** SEM images: (a) and (c) bare silicon nanoparticle electrodes; and (b) and (d) Si/PMMA/lithium electrodes after sealing in cell, PMMA dissolution and prelithiation. After prelithiation, the particle size is larger due to expansion and SEI formation.

### Exposure of PMMA-protected lithium to water droplet



**Figure S4.** The stability of PMMA-protected lithium to a water droplet. (a) 20  $\mu\text{m}$  PMMA-coated lithium with a nominal capacity of 2  $\text{mAh}/\text{cm}^2$ . (b) 100  $\mu\text{m}$  PMMA-coated commercial lithium film (750  $\mu\text{m}$ ). These results show that PMMA protects lithium from water for times up to 60 min.