

Supporting Information

Running out of Lithium?

A Route to Differentiate between Capacity Losses and Active Lithium

Losses in Lithium-Ion Batteries

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Table S1. Lithium, iron and phosphorus stoichiometry of pristine LFP electrodes, measured by ICP-OES and normalized to the iron content. SD: standard deviation; RSD: relative standard deviation.

	Li	Fe	P
LFP electrodes (fresh)	1.112	1	1.110
	1.121	1	1.116
	1.121	1	1.107
	1.140	1	1.103
	1.155	1	1.117
Arithmetic mean	1.130	1	1.111
SD	0.015	-	0.005
RSD / %	1.4	-	0.5

Table S2. Lithium, iron and phosphorus stoichiometry of LFP electrodes which were cycled for 103 cycles against graphite, measured by ICP-OES and normalized to iron and percentage of active lithium loss.

Cycled Cells	Li	Fe	P	Relative lithium loss (by ICP-OES)	Relative lithium loss (by AIC)	<i>ICP-OES</i> <i>AIC</i>
A	1.053	1	1.155	6.8%	14.5%	47%
B	0.999	1	1.154	11.6%	20.4%	57%
C	1.029	1	1.153	8.9%	16.9%	53%
D	1.032	1	1.153	8.7%	16.2%	54%
E	1.024	1	1.151	9.3%	16.8%	56%
Range:						53% ± 3%

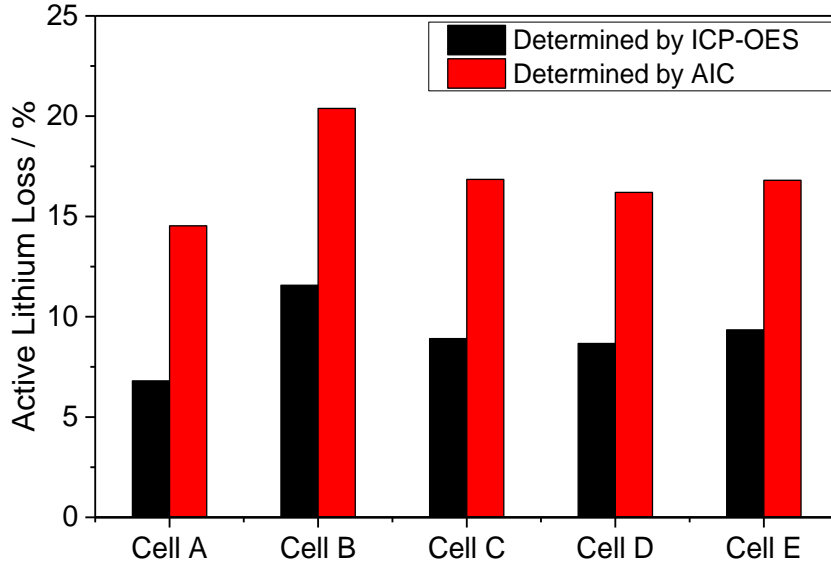


Figure S1. Comparison of active lithium loss after 103 cycles, determined by ICP-OES and accumulated irreversible capacity (AIC).

Silicon/Graphite Composite

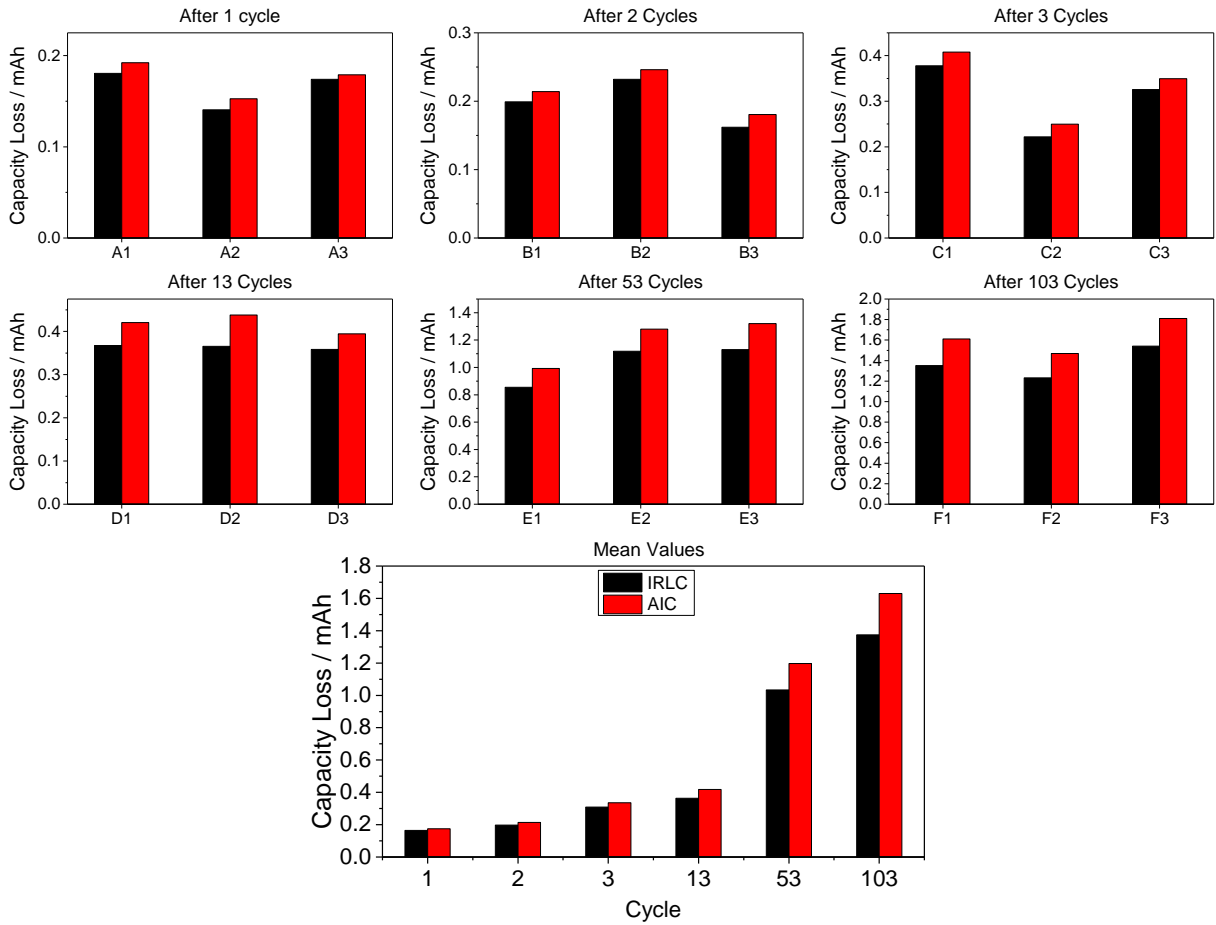


Figure S2. Capacity loss (Q_{ALL}) determined by IRLC (red) and accumulated irreversible capacity (Q_{AIC} , black) after 1, 2, 3, 13, 53 and 103 cycles for three different cells. Standard cycling: Silicon-graphite composite as WE; LFP as CE; metallic lithium as RE; C-Rate: 1C; cut-off potentials: 0.02 V vs. Li/Li⁺ and 1.5 vs. Li/Li⁺. Discharge against RE: C-Rate: C/30; cut-off voltage: 4.1 V.

Carbon Spheres

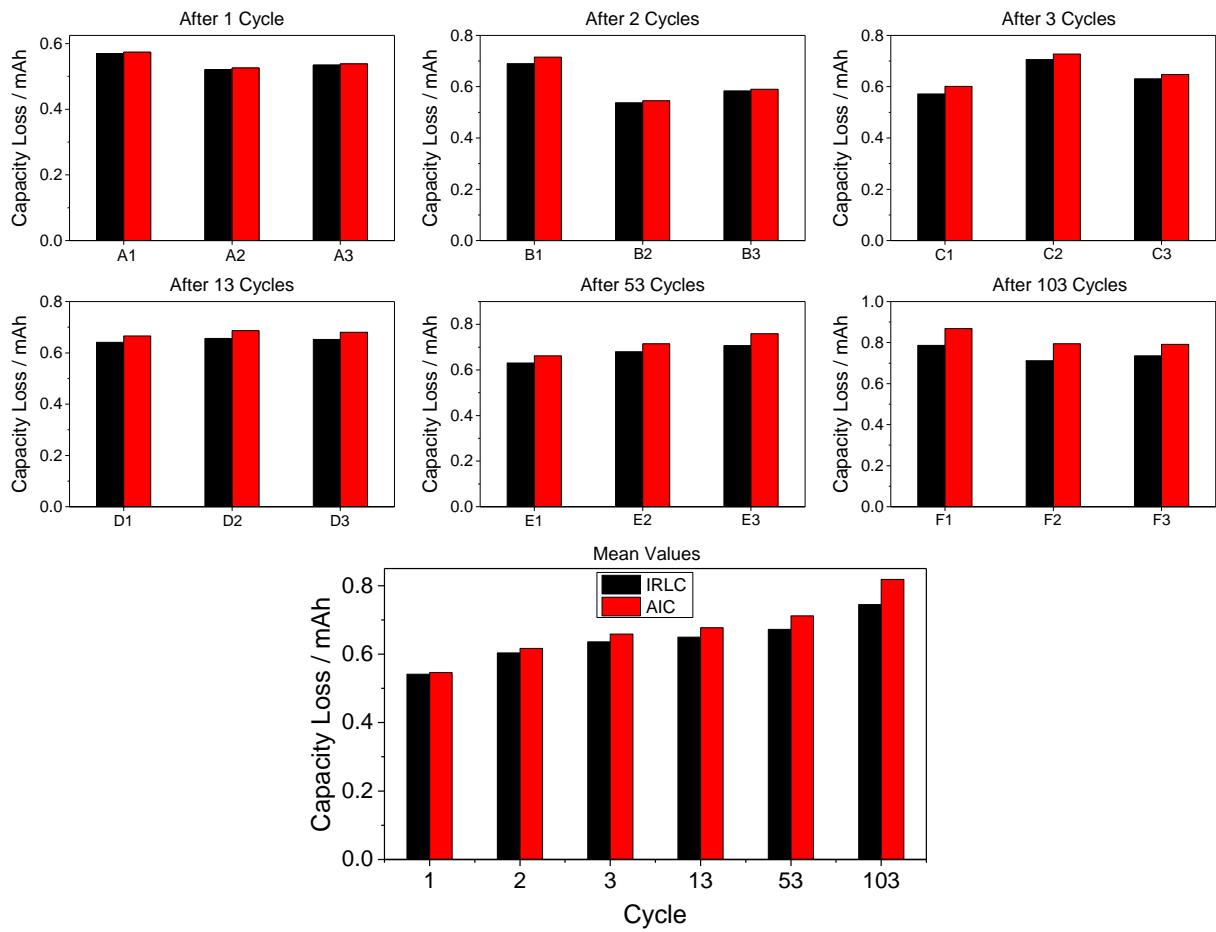


Figure S3. Capacity loss (Q_{ALL}) determined by IRLC (red) and accumulated irreversible capacity (Q_{AIC} , black) after 1, 2, 3, 13, 53 and 103 cycles for three different cells named A, B and C. Standard cycling: Carbon sphere as WE; LFP as CE; metallic lithium as RE; C-Rate: 1C; cut-off potentials: 0.02 V vs. Li/Li⁺ and 1.5 vs. Li/Li⁺. Discharge against RE: C-Rate: C/30; cut-off voltage: 4.1 V.

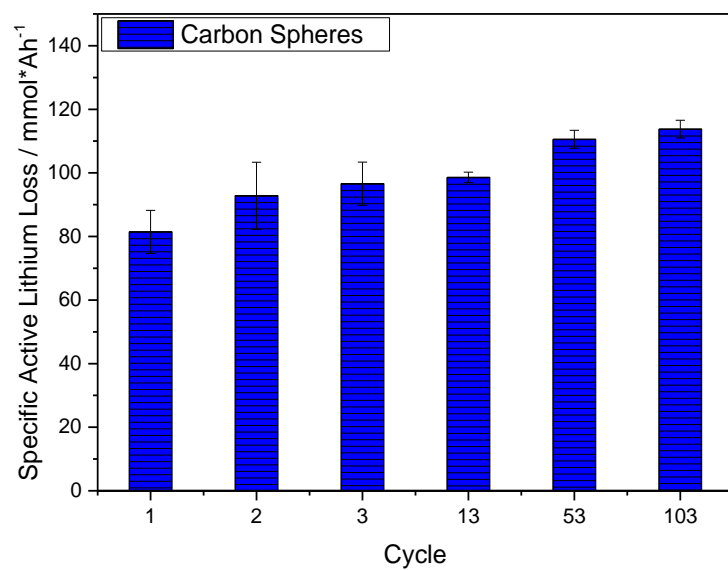


Figure S4. Average active lithium loss (n_{ALL}) of carbon spheres with regard to the electrode capacity.

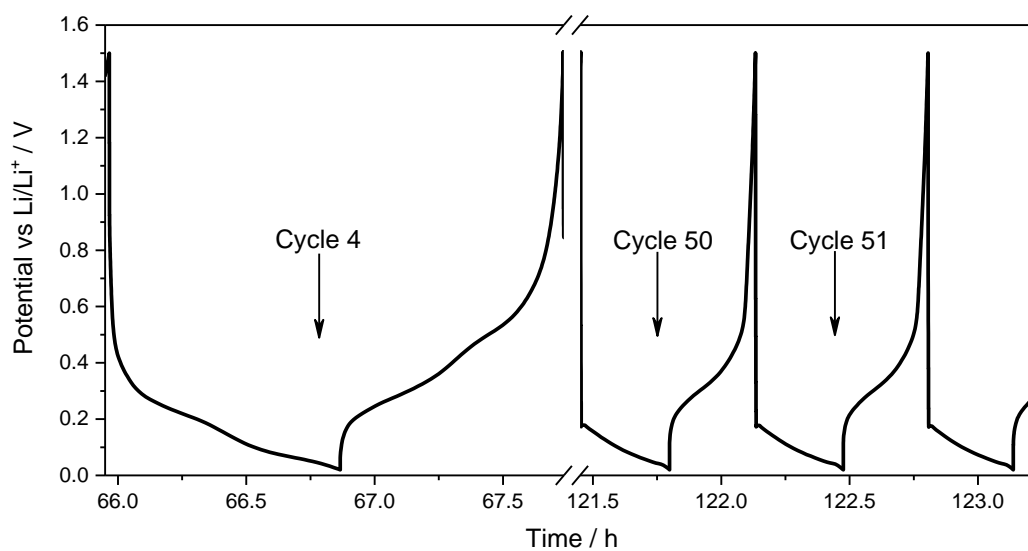


Figure S5. Potential profile of the silicon/graphite composite electrode showing cycle 5, 50 and 51. Silicon/graphite composite as WE; LFP as CE; metallic lithium as RE; C-Rate: 1C; cut-off potentials: 0.02 V and 1.5 vs. Li/Li⁺.