Supplementary Information for

Enhancing the durability of aluminium-foil anodes in rechargeable lithium batteries via uniformly distributed alloy addition in the matrix phase

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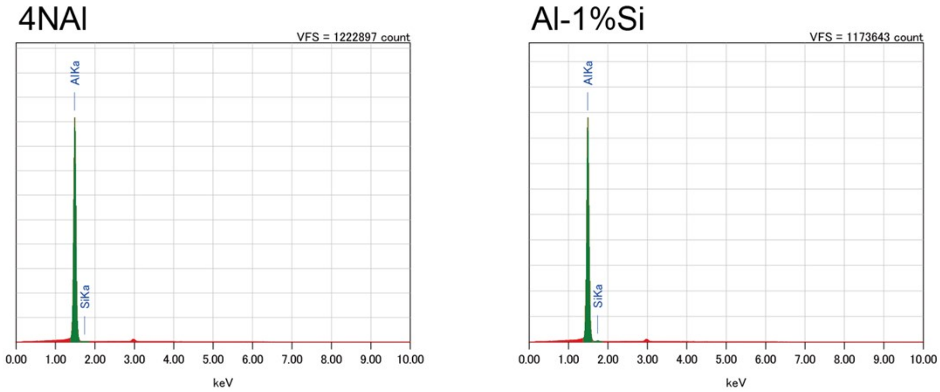

Figure S1. EDX spectra of 4NAl and Al-1%Si corresponding to Figure 1. EDX peak of Si was not observed in 4NAl.

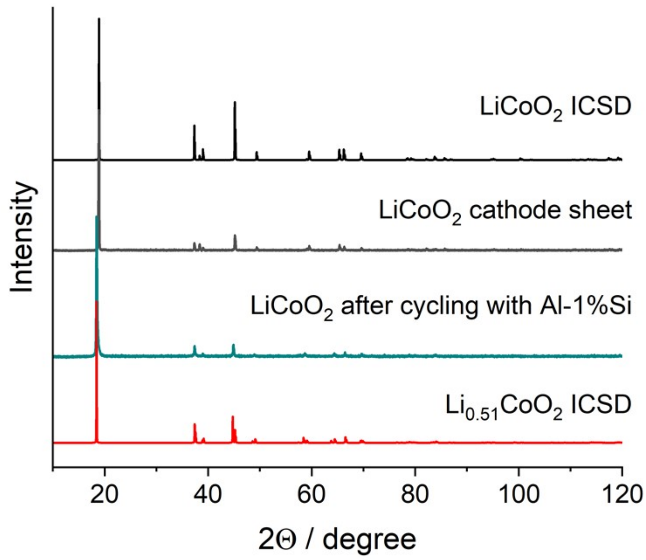

Figure S2. XRD of $LiCoO_2$ cathode after cycling with Al-1%Si corresponding to Figure 2. The $LiCoO_2$ after 120 cycles showed a similar XRD pattern with $Li_{0.51}CoO_2$ without significant structure degradation.

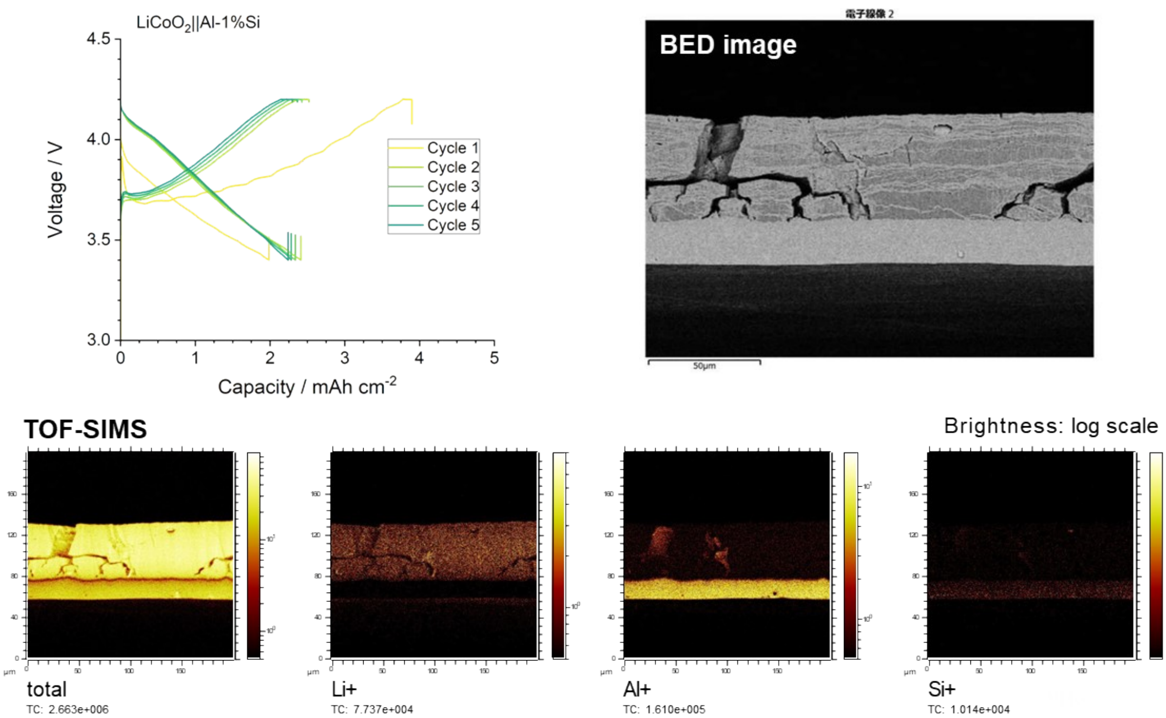

Figure S3. Element mapping by time-of-flight secondary ion mass spectrometry (TOF-SIMS) on an Al-1%Si foil anode after 5 cycles in a $LiCoO_2$||Al-1%Si cell. Part of the inserted Li atoms were kinetically trapped in the surface layer.

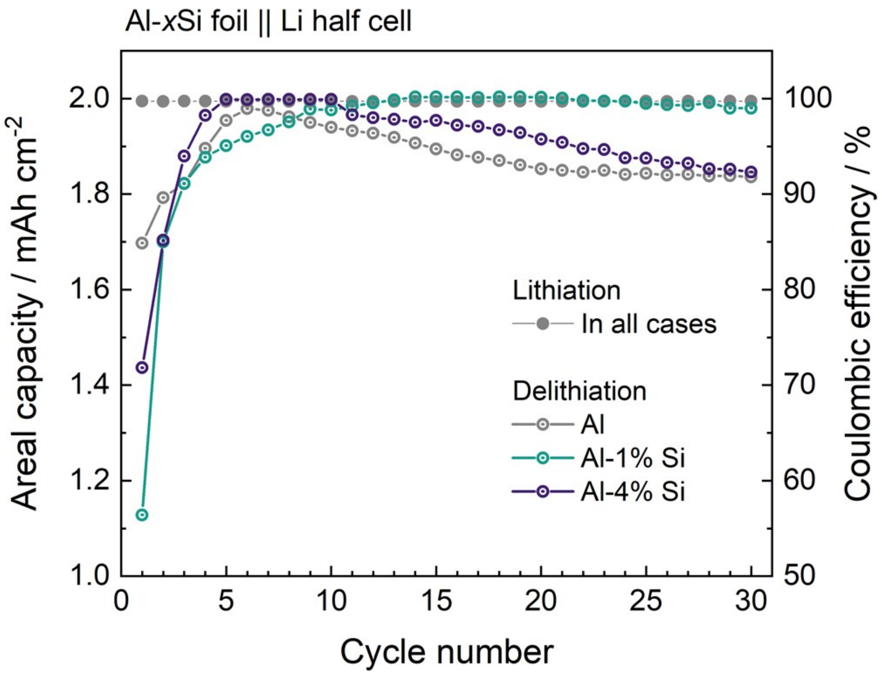

Figure S4. Capacity retention of Al-*x*Si||Li half cells corresponding to Figure 3.

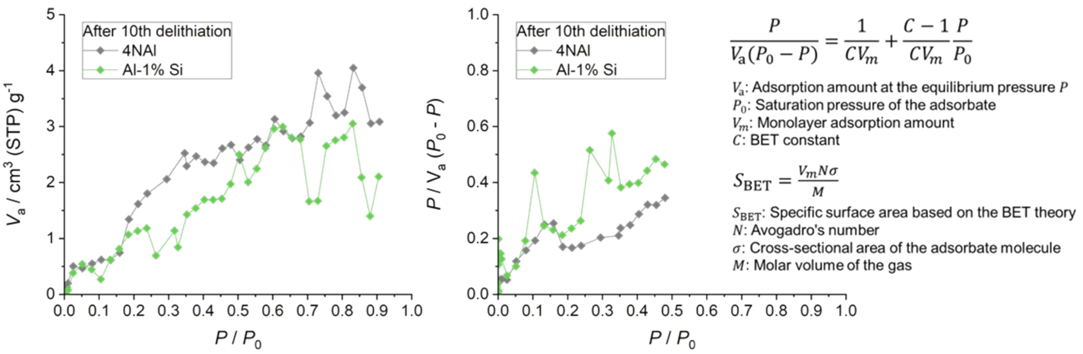

Figure S5. Nitrogen absorption measurements on 4NAl and Al-1%Si foil after the 10th delithiation in half cells with a Li counter electrode. The results correspond to the samples shown in Figure S6 at the current density of 1 mA cm^{-2}. The absorption profiles qualitatively show that the Al-1%Si foil had a relatively low surface area compared to 4NAl, which is consistent with the morphology changes observed in the SEM images. However, due to the fluctuation of the data, it was technically difficult to obtain valid curve fitting results to quantitatively estimate the surface area based on the BET theory. The fluctuation of the measured data would be attributed to the limited amount of sample resulting in a low amount of gas absorption.

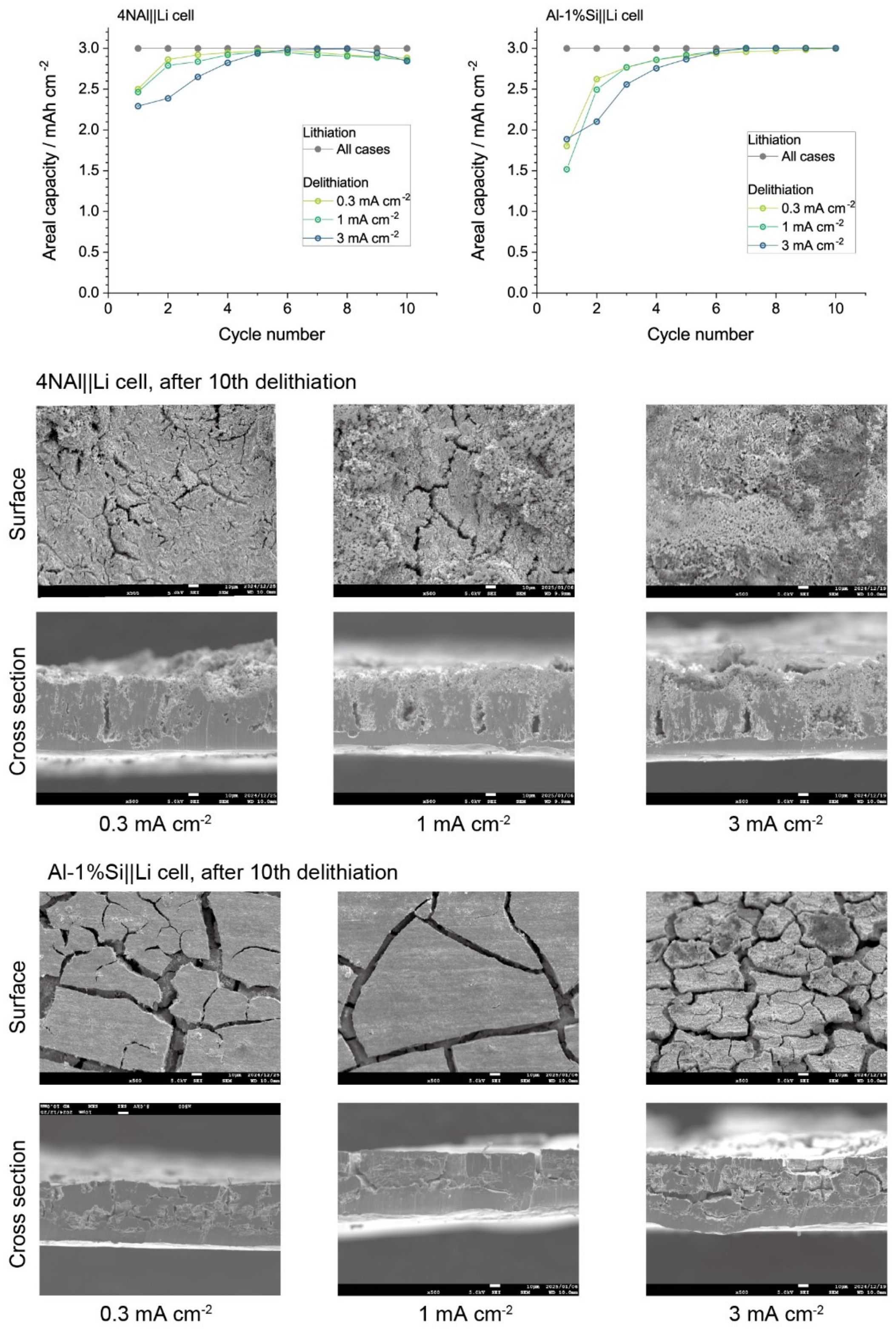

Figure S6. Effects of C-rate on morphology and capacity retention. The irreversible capacity loss in the early cycles increased as increasing the current density, which would be attributed to insufficient Li extraction at higher current densities. In addition, the Al grains in the surface layer become finer as the current density increases. The rapid Li extraction may cause larger stress between the Al grains, accelerating the pulverization during cycling.

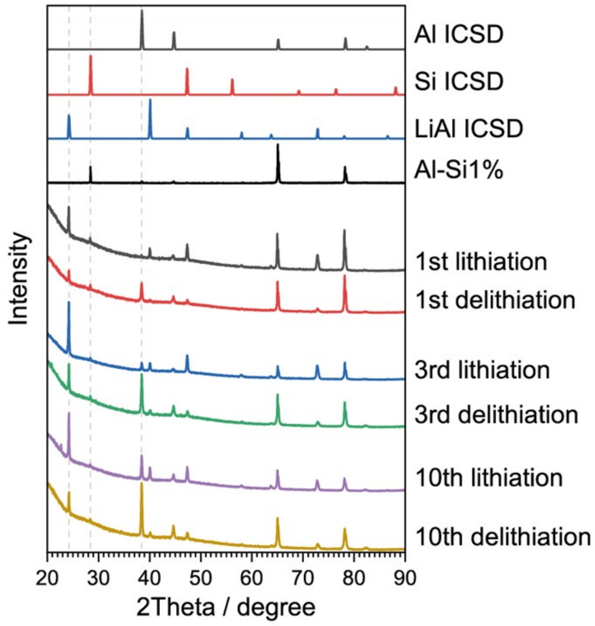

Figure S7. XRD of Al-1Si% after cycling in Al-1%Si||Li half cells corresponding to Figure 3.

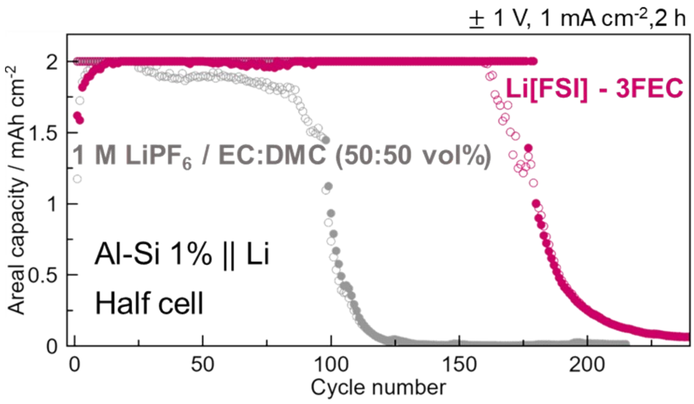

Figure S8. Comparison of cycle tests Al-1%Si||Li half cells with 1 M $LiPF_6$/EC-DMC and Li[FSI]-3FEC electrolytes. The cycle life of Al-1%Si has doubled just by changing the electrolyte.

Table S1. Theoretical capacity of the Al-*x*Si foil anodes.

	Theoretical specific capacity / mAh g^{-1}		
	Total	Al	Si
Al	993	993	0
Al-Si0.6%	1008.516	987.042	21.474
Al-Si1%	1018.86	983.07	35.79
Al-Si2%	1044.72	973.14	71.58
Al-Si4%	1096.44	953.28	143.16

The specific capacities were calculated with respect to the following lithiation reactions: $Li^+ + e^- + Al = LiAl$ (~0.38 V vs Li) and $15Li^+ + 15e^- + 4Si = Li_{15}Si_4$ (~0.4 V vs Li).[1]

[1] M. N. Obrovac, V. L. Chevrier, *Chem Rev* 2014, 114, 11444.

Table S2. Electromotive force of compounds in Li-Al-Si ternary system.

	Phase	Formation energy / eV $atom^{-1}$	Material ID in the Materials Project	Electromotive force / V
Li-Al	LiAl	-3.00	mp-1067	0.34
	Li_3Al_2	-2.83	mp-16506	0.31
	Li_9Al_4	-2.62	mp-568404	0.21
Li-Si	Li_7Si_3	-3.21	mp-1201871	0.35
	$Li_{13}Si_4$	-2.97	mp-672287	0.30
	$Li_{15}Si_4$	-2.87	mp-569849	0.28
	$Li_{21}Si_5$	-2.79	mp-29720	0.25
Li-Al-Si	LiAlSi	-3.88	mp-3161	0.55