

Supplementary Materials

Decoupling Mechanical Confinement from Ion-Transport Penalty in Sn Foil Anodes via Al-Phase Topology Regulation

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How To Cite: Jia, Y.; Wu, C.; Li, J.; et al. Decoupling Mechanical Confinement from Ion-Transport Penalty in Sn Foil Anodes via Al-Phase Topology Regulation. *eChem* 2026, 2(2), 8. <https://doi.org/10.53941/echem.2026.100008>

Calculation Method

The density functional theory (DFT) calculation was performed using the Vienna ab initio simulation package (VASP) based on the projector augmented wave (PAW) method [1]. The calculations were carried out using the Perdew–Burke–Ernzerhof (PBE) exchange and correlation functional within the generalized gradient approximation (GGA). For calculating the adsorption characteristics of Li on Al (Sn) surface, a (111) surface of bulk Al was modeled by the $3 \times 3 \times 1$ supercells and a (101) surface of bulk Sn was modeled by the $2 \times 2 \times 1$ supercells. For Al/Sn interface, a (111) surface of bulk Al was cleaved to make a slab with enough vacuum region, while the Sn (101) was placed on it [2]. To avoid spurious interactions between neighboring slabs, we introduce a vacuum of ~ 15 Å, which ensures dispersion forces are sufficiently small on the scale of key energetics. The adsorption energy (E_{ads}) of Li adsorption on the Al (111), Sn (101) surface and Al/Sn interface are defined as $E_{\text{ads}} = E_{\text{total}} - E_{\text{Li}} - E_{\text{sub}}$, where E_{total} and E_{sub} represent the total energy of Al (111), Sn (101) and Al/Sn systems with and without Li, respectively, and E_{Li} represents the energy of an isolated Li. A $2 \times 2 \times 1$ K-point mesh was used for structural optimization, with the energy and force convergence criteria set to 1×10^{-6} eV and 0.015 eV Å⁻¹, respectively [3]. The energy cut-off of the plane-wave basis set was 450 eV.

Finite element simulations were performed using COMSOL Multiphysics, coupling the Solid Mechanics and transport of diluted species physics fields to model the concurrent processes of stress evolution and crack development during lithiation in Sn-ML and Al_mSn-ML. A 2D polycrystalline geometry was constructed based on experimental observations, with a rectangular simulation domain measuring 20.0×16.0 μm². Material properties were sourced from experimental data, built-in databases, and published literature wherever applicable [4,5]. For boundary conditions, the bottom surface of the foil was fixed, while lateral edges were left unconstrained. A Dirichlet boundary condition for Li concentration was applied to the top surface to simulate lithium insertion.

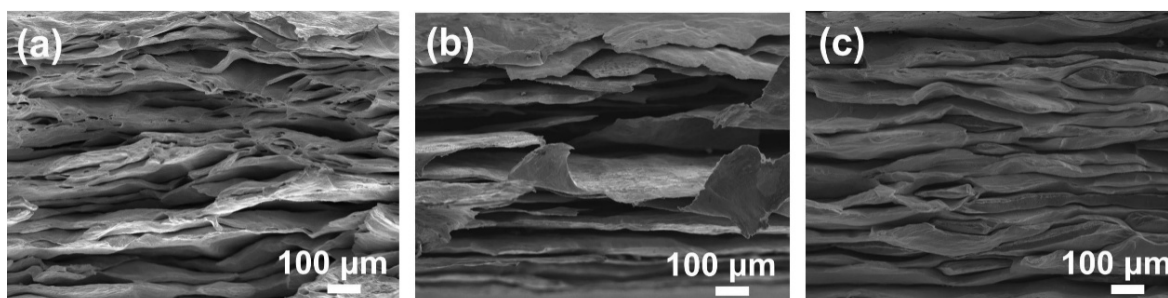


Figure S1. Higher-magnification SEM images of the fracture morphologies of Al_mSn-ML, Al_sSn-ML, and Sn-ML after tensile testing.



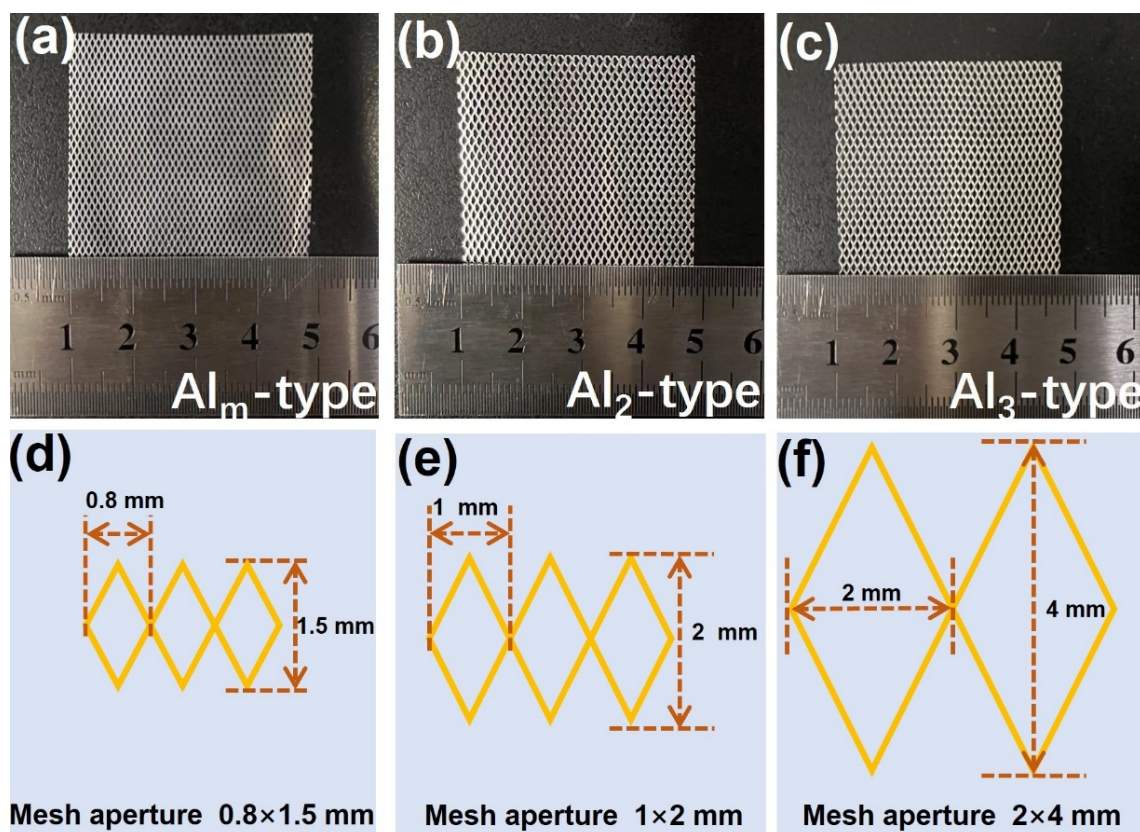


Figure S2. Optical images and corresponding dimensions of (a,d) Al_m , (b,e) Al_2 , (c,f) Al_3 .

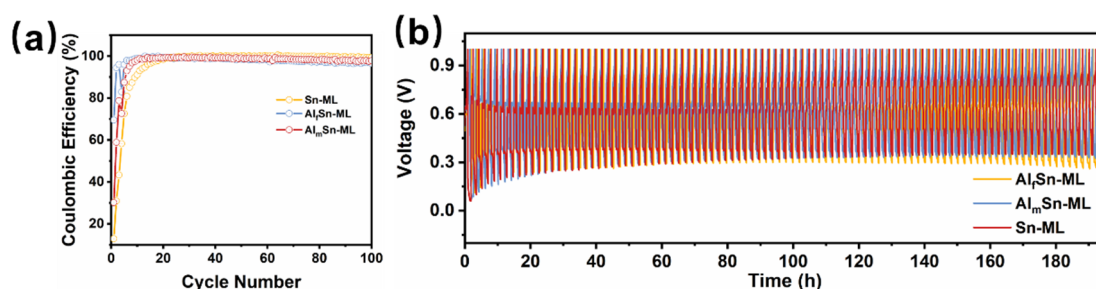


Figure S3. High-current cycling performance of Sn-ML, Al_I Sn-ML, and Al_m Sn-ML electrodes at 2 mA cm^{-2} with a fixed areal capacity of 2 mAh cm^{-2} . (a) Coulombic efficiency during 100 cycles. (b) Representative GCD profiles of Sn-ML, Al_I Sn-ML, and Al_m Sn-ML at selected cycles.

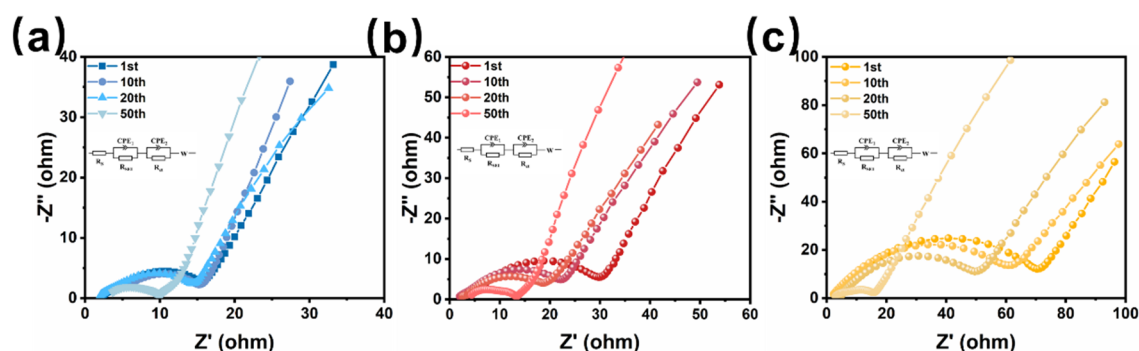


Figure S4. Nyquist plots of (a) Sn-ML, (b) Al_m Sn-ML, and (c) Al_I Sn-ML after the 1st, 10th, 20th and 50th cycles, respectively.

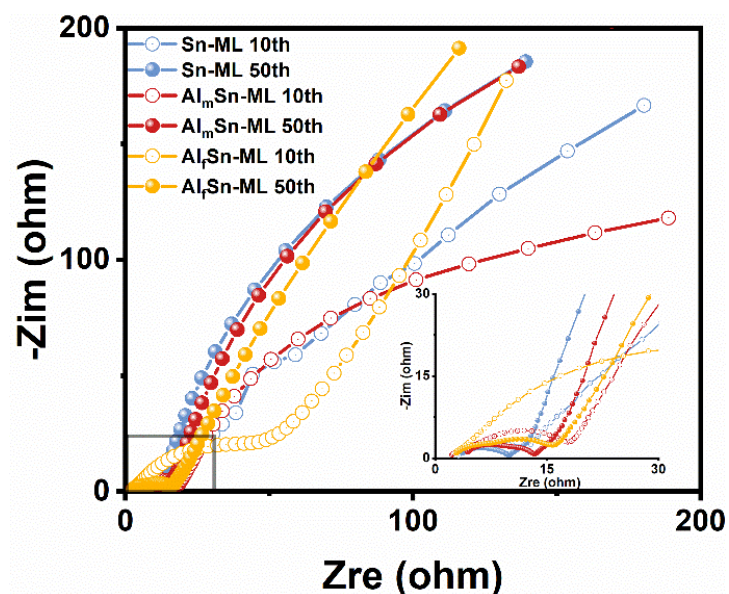


Figure S5. Nyquist plots of Sn-ML, Al_mSn-ML, and Al_lSn-ML after the 10th and 50th cycles, respectively.

Table S1. Detailed performance parameters of the previously reported Sn-based anodes used for comparison in Figure 2g.

Materials	Current Density	Areal Capacity	Electrode Thickness	Cycle Number	Ref.
0.6 CNTSn	0.6 mA cm ⁻²	1 mAh cm ⁻²	20 μm	150	[6]
CRSnZn-25p	0.6 mA cm ⁻²	2 mAh cm ⁻²	20 μm	100	[7]
Pure Al	1 mA cm ⁻²	2 mAh cm ⁻²	30 μm	70	[8]
Al ₉₅ Ga ₅	1 mA cm ⁻²	2 mAh cm ⁻²	30 μm	120	[8]
Al ₅₀ Zn ₅₀	1 mA cm ⁻²	2 mAh cm ⁻²	30 μm	140	[8]
Al ₆₅ Sn ₃₅	1 mA cm ⁻²	2 mAh cm ⁻²	30 μm	160	[8]
Sn-Pre3	1 mA cm ⁻²	2 mAh cm ⁻²	50 μm	280	[9]
3DIP-Sn foil	2 mA cm ⁻²	1 mAh cm ⁻²	25 μm	300	[4]
This Work	1 mA cm ⁻²	2 mAh cm ⁻²	50 μm	430	

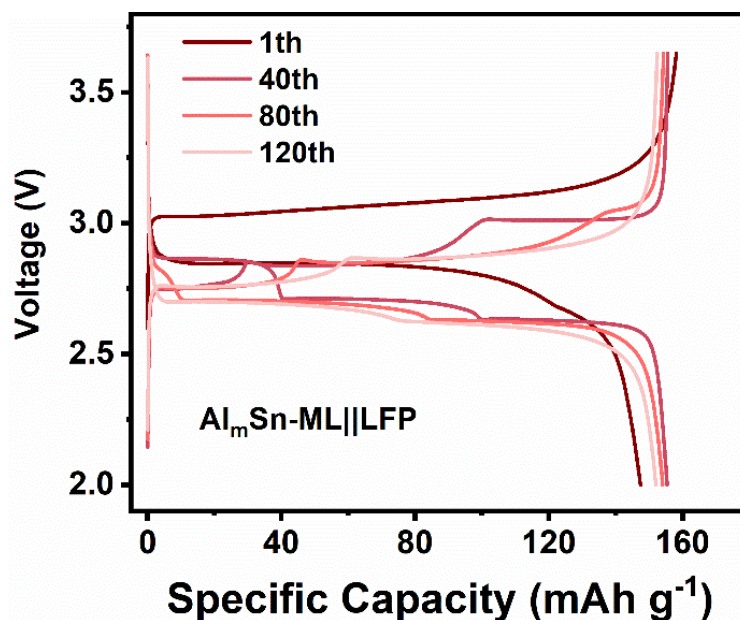


Figure S6. GCD curves of the full cells based on the Al_mSn-ML anode paired with LFP cathodes.

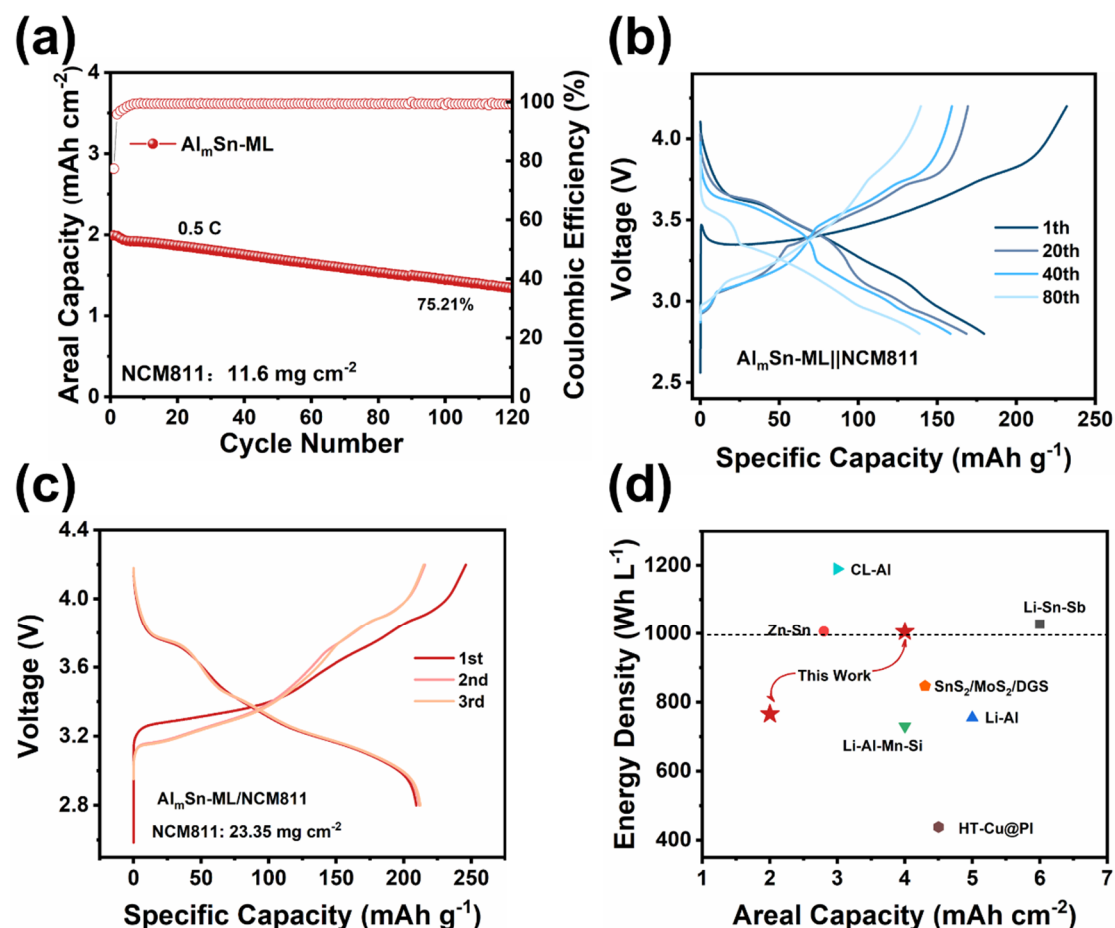


Figure S7. (a) Cycling performance of the Al_mSn-ML anode paired with NCM811 cathodes at 2 mAh cm⁻²; corresponding GCD curves at (b) 2 mAh cm⁻² and (c) 4 mAh cm⁻²; (d) comparison of the volumetric energy density of the Al_mSn-ML/NCM811 full cell with previously reported cells [10–16].

Table S2. Cell-stack parameters used for calculating the volumetric energy density of Al_mSn-ML/NCM811 full cells.

Cell Component	Specification	Parameters	
Cathode	Mass loading (mg cm ⁻²)	11.6 mg cm ⁻²	23.35 mg cm ⁻²
	Weight (mg)	9.11 mg	18.33 mg
	Thickness (μm)	39.7 μm	75.3 μm
	Compaction density (g cm ⁻³)	3.2 g cm ⁻³	3.2 g cm ⁻³
	Al foil (μm)	14 μm	14 μm
Anode	Prelithiated anode foil thickness (μm)	17.5 μm	27 μm
	Δd (μm)	9.5 μm	19 μm
	Average cell voltage (V)	3.3 V	3.3 V
Separator	Thickness (μm)	15 μm	15 μm
	Areal capacity (mAh cm ⁻²)	2 mAh cm ⁻²	4 mAh cm ⁻²
Full cell	Energy density (Wh L ⁻¹)	765.7 Wh L ⁻¹	1005.3 Wh L ⁻¹

In practical LIB applications, volumetric energy density is a critical metric for evaluating the performance of energy-storage systems. The volumetric energy density of the cell configuration can be quantitatively estimated using Equations (S1) and (S2) [11,17,]:

$$\Delta d = \frac{k \times x \times m_1}{s} \quad (\text{S1})$$

$$W_v = \frac{U \times m_2 \times Q_s}{d_{\text{cathode}} + d_{\text{anode}} + d_{\text{current collectors}} + d_{\text{separator}} + \Delta d} \quad (\text{S2})$$

where Δd represents the thickness increase caused by volume expansion during electrode lithiation (μm), k is the molar volume coefficient associated with the lithiation/alloying reaction ($8.9\text{ cm}^3\text{ mol}^{-1}$), x is the molar coefficient of lithiation/alloying per unit mass (mol g^{-1}), m_1 is the mass of active material in the electrode (mg), and s is the effective reaction area of the electrode (0.785 cm^2). In Equation (S2), W_v is the volumetric energy density (Wh L^{-1}), U is the working voltage (V), m_2 is the areal loading of active material (mg cm^{-2}), Q_s is the discharge specific capacity (mAh g^{-1}), and d represents the thickness of each cell component (μm).

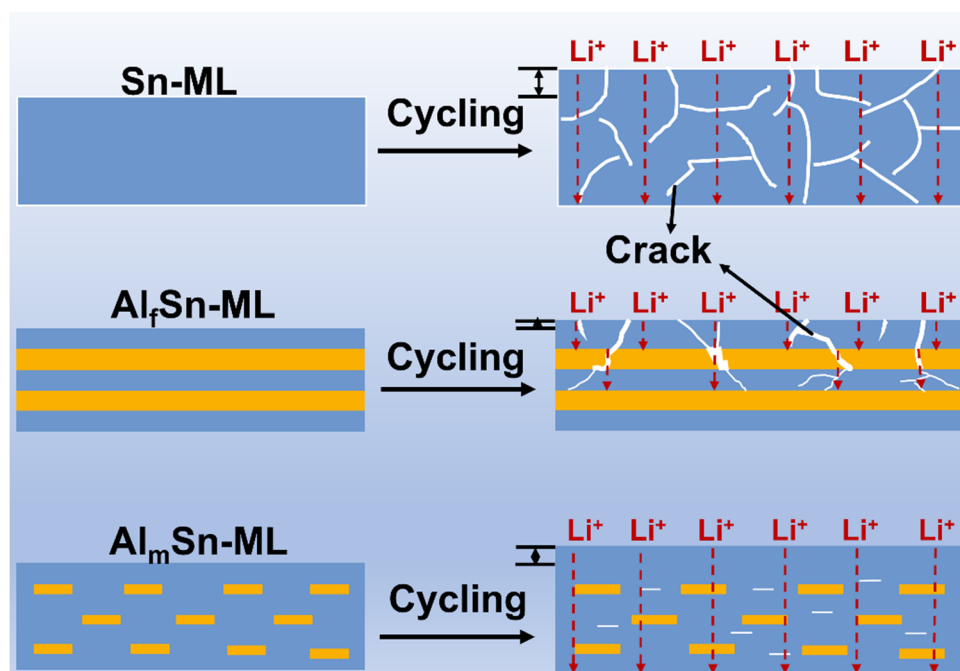


Figure S8. Schematic illustration of the lithiation behavior of Sn-ML, Al_fSn-ML, and Al_mSn-ML during cycling.

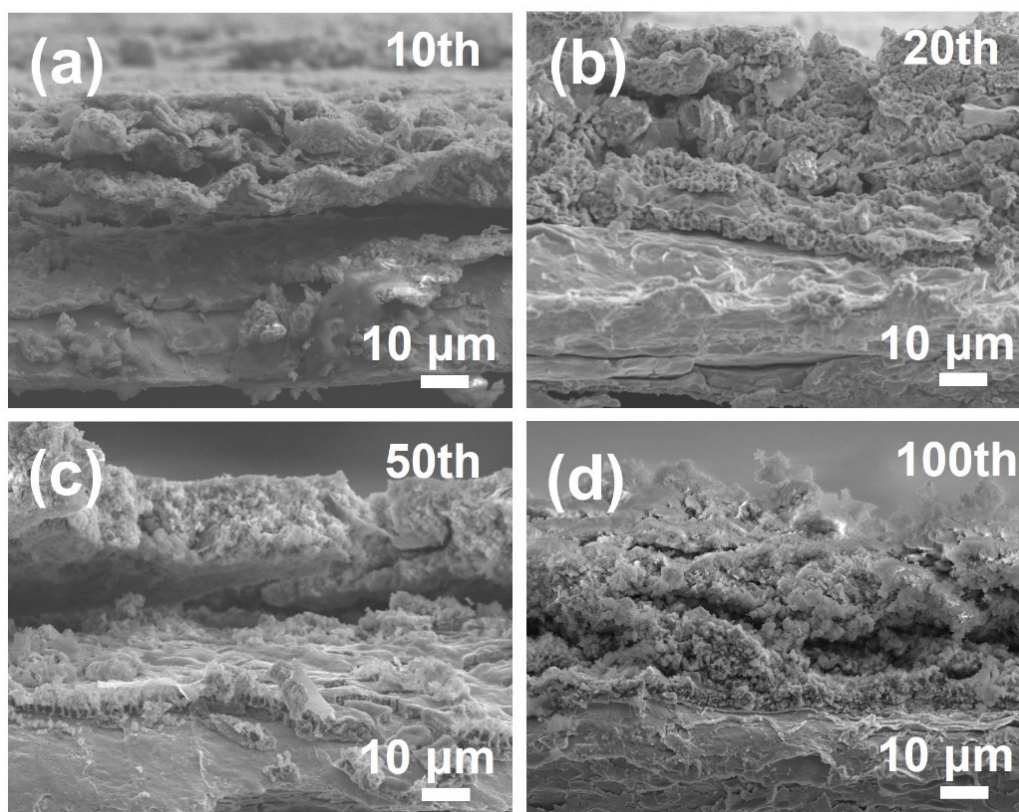


Figure S9. (a–d) Cross-sectional SEM images of the Al_fSn-ML electrode after 10, 20, 50, and 100 cycles, respectively.

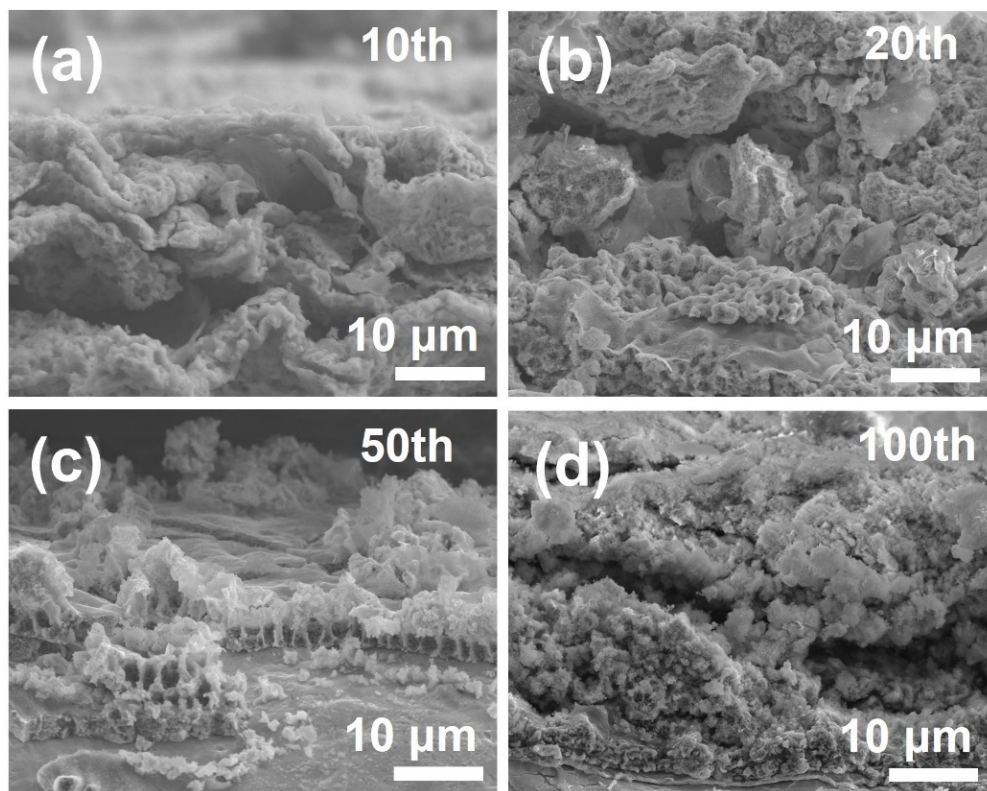


Figure S10. (a–d) Higher-magnification cross-sectional SEM images of the Al_fSn-ML electrode after 10, 20, 50, and 100 cycles, respectively (corresponding to the images Figure S9).

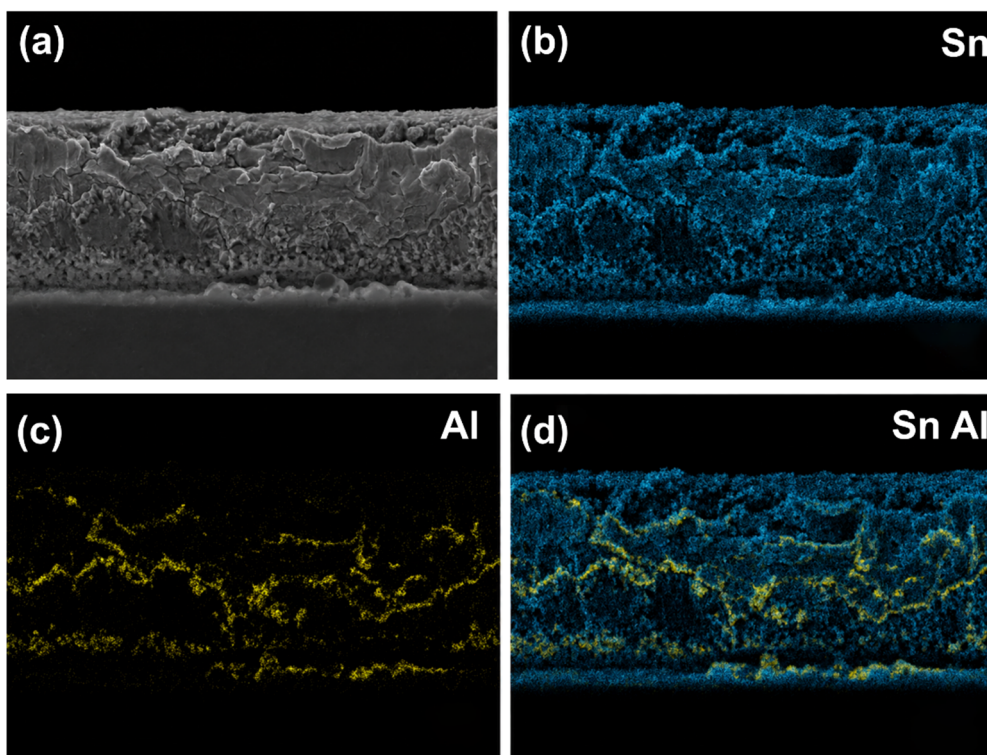


Figure S11. Cross-sectional SEM image and corresponding EDS elemental mapping of the cycled Al_mSn-ML electrode: (a) SEM image; (b) Sn mapping; (c) Al mapping, and overlay image; and (d) overlay image.

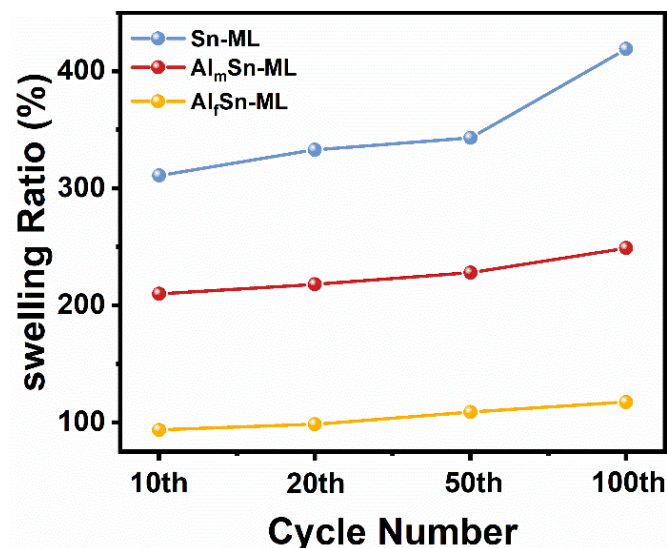


Figure S12. Thickness expansion ratio statistics of Al_mSn-ML, Al_lSn-ML, and Sn-ML measured after 10, 20, 50, and 100 cycles, respectively.

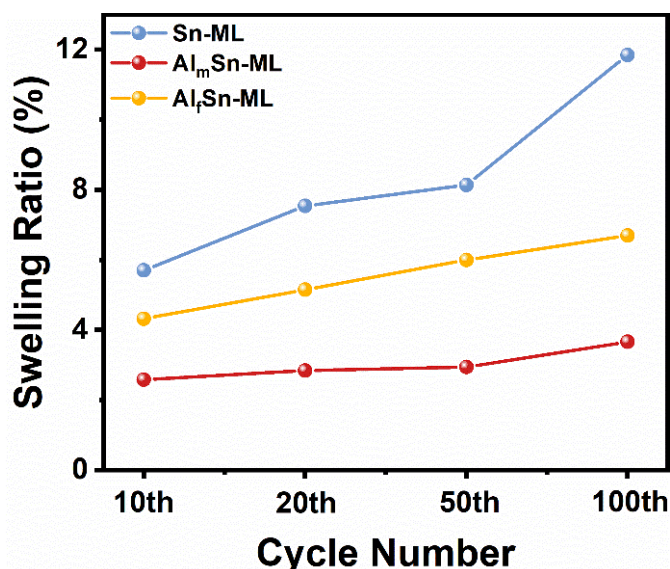


Figure S13. Diameter expansion ratio statistics of Al_mSn-ML, Al_lSn-ML, and Sn-ML measured after 10, 20, 50, and 100 cycles, respectively.

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