



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

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Abstract: Alloy-type metal foils are attractive anodes for high-energy lithium-ion batteries because they eliminate inactive electrode components while providing high volumetric capacity. However, their practical implementation is hindered by a fundamental transport–mechanics conflict. Uniform alloying reactions require open ion pathways, whereas suppressing large lithiation-induced volume strain demands strong mechanical confinement. Here, we demonstrate that this conflict can be mitigated by regulating the spatial continuity of the Al phase in multilayer Sn foil anodes. In contrast to pure Sn multilayers that undergo severe pulverization and continuous Al/Sn laminates that impose a Li⁺ transport barrier, a mesh-derived discontinuous Al skeleton embedded in a continuous Sn network simultaneously preserves Li⁺ accessibility and redistributes alloying-induced stress. The optimized Al_mSn-ML anode sustains 430 cycles at 2 mAh cm^{−2} and 1 mA cm^{−2}, while full cells paired with high-loading LiFePO₄ (LFP) cathodes retain 91.51% capacity after 160 cycles. Moreover, Al_mSn-ML/Li_{0.8}Co_{0.1}Mn_{0.1}O₂ (NCM811) full cells deliver a high volumetric energy density. These findings highlight topology-regulated mechanical reinforcement as an effective strategy for developing low-strain, high-capacity foil anodes.

Keywords: alloy-type foil anodes; Sn/Al multilayers; Al-phase continuity; Li⁺ transport; mechanical confinement; structural stability

1. Introduction

Developing high-capacity anodes is essential for advancing lithium-ion batteries toward higher energy density [1–5]. Alloy-type anodes are particularly attractive because they can store Li through multi-electron alloying reactions and deliver capacities far beyond those of conventional graphite [6–8]. Among various alloying materials, tin (Sn) has been widely investigated owing to its high theoretical capacity, high volumetric energy density, and moderate lithiation potential [9–13]. Moreover, Sn foil anodes provide a binder-free and current-collector-free electrode configuration, offering an opportunity to reduce inactive components and improve electrode-level energy density [14–17]. However, the practical application of Sn foil anodes remains severely limited by the large volume variation associated with repeated lithiation and delithiation [18–20]. The drastic volume fluctuation induces severe stress accumulation, crack propagation, structural pulverization, and electrical contact loss, eventually causing rapid capacity decay [21–23].

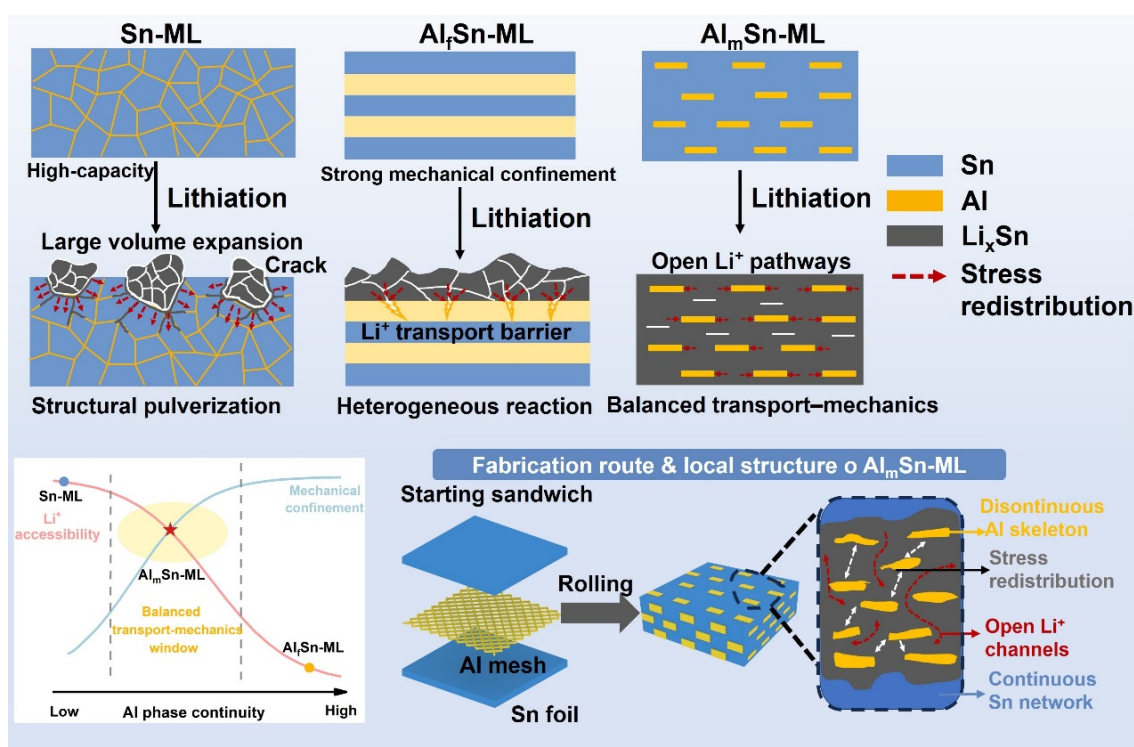
To address the chemo-mechanical instability of Sn foils, structural engineering has been widely explored to regulate lithiation uniformity and strain accommodation. Tu et al. designed a three-dimensional interconnected porous Sn foil with refined grains to alleviate nonuniform alloying reactions and localized stress accumulation during lithiation/delithiation [24]. The resulting porous Sn foil showed greatly improved cycling stability,



confirming that regulating the lithiation pathway and stress distribution is critical for suppressing chemo-mechanical failure in Sn foil anodes [25]. Nevertheless, such pore-dominated buffering strategies mainly rely on internal free volume to accommodate expansion, which may weaken the intrinsic density advantage of foil-type electrodes [26]. Therefore, for dense Sn foil anodes, it is desirable to develop a compact structural design that can regulate stress without relying solely on large internal voids. Dense laminated Sn foils provide such a potential route because internal interfaces can deflect cracks and redistribute local deformation while preserving the compact foil configuration. However, since all Sn layers remain electrochemically active and mechanically compliant, multilayering Sn alone cannot fundamentally suppress lithiation-induced swelling [23]. An additional mechanical supporting phase is therefore required to restrain Sn expansion while maintaining efficient Li^+ transport.

Introducing a mechanically robust secondary phase into Sn laminates represents a logical strategy for enhancing swelling resistance. Aluminum (Al) is a particularly attractive reinforcing phase because of its low density, metallic conductivity, mechanical rigidity, and compatibility with scalable foil processing [27,28]. Ichitsubo et al. indicated that alloying-induced volume strain can be directionally regulated by an appropriate metallic matrix, emphasizing the importance of mechanical confinement in alloy-type foil electrodes [29]. Therefore, introducing Al into Sn laminates is expected to provide internal mechanical support for restraining Sn expansion. Nevertheless, when Al exists as a continuous interlayer, it may also act as a through-plane transport barrier, interrupting Li^+ penetration and lengthening ion-transport pathways across the laminate [30].

Herein, we propose an Al-phase-continuity regulation strategy for Sn-based laminated foil anodes. Three representative foil architectures were designed to clarify the role of Al-phase topology. These include pure multilayer Sn foil (Sn-ML), continuous Al foil reinforced Sn multilayer foil ($\text{Al}_f\text{Sn-ML}$), and discontinuous Al mesh skeleton reinforced Sn multilayer foil ($\text{Al}_m\text{Sn-ML}$). A synergistic design balancing lithiation kinetics and mechanical confinement is adopted in $\text{Al}_m\text{Sn-ML}$. Specifically, the continuous Al layer is reconstructed into a discontinuous Al-mesh skeleton, allowing Sn to maintain a continuous electrochemically active network while Al serves as local mechanical support. This fundamentally overcomes the Li^+ transport bottleneck caused by a continuous Al phase, thereby achieving uniform stress distribution, crack deflection, and effective suppression of volume expansion, and ultimately ensuring homogeneous lithiation and structural integrity. Concretely, the introduction of the discontinuous Al mesh tailors the mechanical constraint and ion-transport pathways at the Sn/Al interface, providing sufficient swelling resistance while preserving fast Li^+ diffusion. This approach effectively avoids local stress accumulation and enables rational strain partitioning during repeated lithiation/delithiation. Moreover, the inherent discontinuous topology of the Al mesh forms a three-dimensionally embedded supporting framework within the Sn matrix and induces transverse crack deflection, thereby minimizing macroscopic pulverization and deformation of the electrode (Scheme 1).



Scheme 1. Design principle and fabrication route of multilayer Sn/Al foil anodes with Al phase continuity regulation.

A strain-regulation system integrating crack deflection, stress dissipation, and volume buffering is thus established. Under practically relevant conditions (2 mAh cm^{-2} , >400 cycles), the $\text{Al}_m\text{Sn-ML}$ anode exhibits substantially suppressed volume expansion while maintaining excellent dimensional stability, and its Coulombic efficiency (CE) approaches 100%. In full cells paired with a LiFePO_4 (LFP) cathode at a high areal loading of 17.2 mg cm^{-2} , outstanding long-term cycling stability is achieved. This work offers a mechanism-guided and experimentally validated design strategy for next-generation alloy-type foil anodes with intrinsically low strain and superior durability.

2. Experimental Section

2.1. Materials and Methods

Preparation of Sn-ML: Sn foil with a thickness of $200 \mu\text{m}$ was cut into 60 square pieces of $3 \text{ cm} \times 3 \text{ cm}$. These pieces were stacked in sequence and then compressed on a cold press at a loading rate of 2.5 kN s^{-1} until the force reached 500 kN, and the pressure was maintained for 300 s. A compact Sn block with a thickness of approximately 3.5 mm was obtained. Subsequently, the block was rolled at room temperature. By precisely controlling the rolling parameters, a cumulative thickness reduction of 98.6% was achieved, and a Multi-layer Sn foil (Sn-ML) with a final thickness of $50 \mu\text{m}$ was obtained.

Preparation of $\text{Al}_f\text{Sn-ML}/\text{Al}_m\text{Sn-ML}$: Sn foil ($200 \mu\text{m}$ thick) and Al foil ($50 \mu\text{m}$ thick)/Al mesh were each cut into 30 square pieces of $3 \text{ cm} \times 3 \text{ cm}$. They were then stacked in the order of Sn foil/Al foil (or Al mesh) / Sn foil. The same cold-pressing parameters as described above were applied, yielding a compact Sn/Al composite block with a thickness of approximately 3 mm. Finally, the block was rolled at room temperature. By controlling the rolling parameters, a cumulative thickness reduction of 98.4% was achieved, and Multi-layer Sn–Al foil ($\text{Al}_f\text{Sn-ML}$) or Multi-layer Sn–Al foil ($\text{Al}_m\text{Sn-ML}$) with a final thickness of $50 \mu\text{m}$ was obtained.

2.2. Material Characterization

Phase composition was analyzed using X-ray diffraction (XRD, Bruker D8 Discover, Bruker, Karlsruhe, Germany). Surface and cross-sectional morphologies were examined by scanning electron microscopy (SEM, JEOL JSM-7100F, JEOL Ltd., Tokyo, Japan) operated at 15 kV with a working distance of 10 mm. Hardness measurements were performed using a HVS-1000Z micro Vickers hardness tester (Shanghai Yizong Instrument Co., Ltd., Shanghai, China). A load of 98.08 mN was applied with a dwell time of 15 s, using the HV0.01 hardness scale. Tensile properties were evaluated using a USD-001 universal dual-column tensile testing machine (HuiShiDa Instrument Co., Ltd., Dongguan, China).

2.3. Electrochemical Measurement

Standard CR2025 coin cells were assembled to evaluate the electrochemical performance. Celgard 2500 membrane was used as the separator, and the electrolyte consisted of 1 M LiPF_6 in a mixture of ethylene carbonate (EC) and diethyl carbonate (DEC) (1:1 by volume), supplemented with 10 vol% fluoroethylene carbonate (FEC) and 1 vol% vinylene carbonate (VC). In half-cells, alloy-type foils were punched into 10 mm discs and used as the working electrode, while lithium metal foil served as both counter and reference electrodes. For full cells, the alloy foils were prelithiated prior to assembly. The LiFePO_4 (LFP) $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$ (NCM811) cathode slurry was prepared by mixing LFP, Super P, and PVDF in N-methyl-2-pyrrolidone (NMP) at a mass ratio of 8:1:1. All electrochemical tests were conducted at 25°C using a BTS4000 NEWARE battery testing system (NEWARE, Shenzhen, China). Electrochemical impedance spectroscopy (EIS) was carried out on an AMETEK Princeton VersaSTAT workstation (USA) in the frequency range of 0.1 Hz–100 kHz.

3. Results and Discussion

3.1. Structural Topology and Mechanical Properties of Multi-Layer Sn/Al Foils

To clarify the role of Al-phase spatial continuity in Sn-based laminated foil anodes, three representative multi-layer architectures were constructed: Sn-ML, $\text{Al}_f\text{Sn-ML}$, and $\text{Al}_m\text{Sn-ML}$. Cross-sectional Scanning Electron Microscope (SEM) images first confirm the successful construction of the three laminated structures. As shown in Figure 1a–c, all samples exhibit a compact foil-like morphology after rolling, but their internal architectures are distinctly different. Sn-ML presents a relatively homogeneous Sn-based laminated structure without an additional reinforcing phase. In contrast, $\text{Al}_f\text{Sn-ML}$ shows a clear continuous interlayer embedded between Sn layers, indicating that the original Al foil largely retains its laterally continuous configuration after rolling. For $\text{Al}_m\text{Sn-ML}$, the Al mesh-

derived phase does not form a fully continuous barrier layer. Instead, the initial Al mesh is transformed into distributed strip-like domains within the Sn matrix, suggesting a discontinuous Al-reinforced topology.

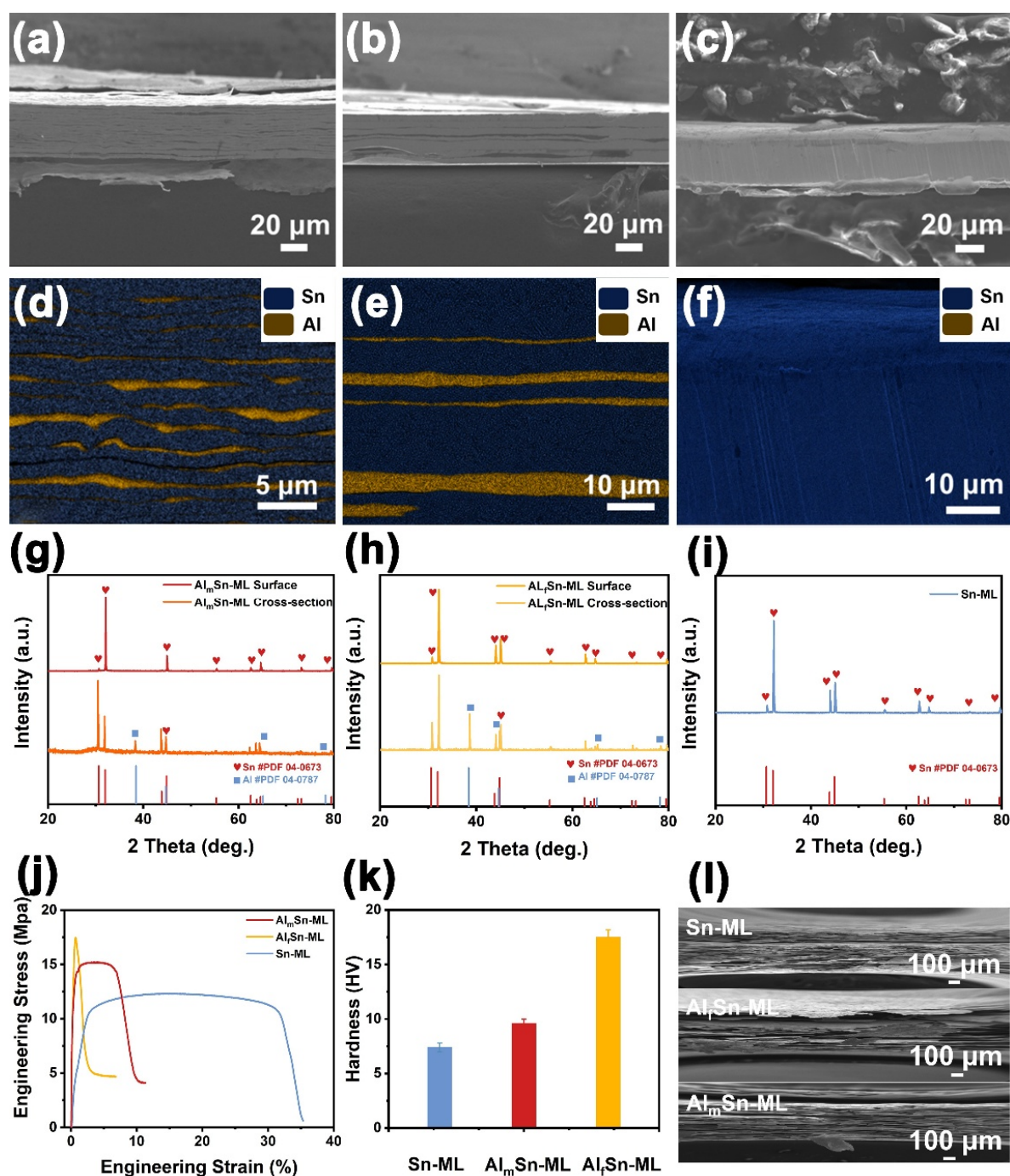


Figure 1. Cross-sectional SEM images of (a) $\text{Al}_m\text{Sn-ML}$, (b) $\text{Al}_l\text{Sn-ML}$, and (c) Sn-ML foils. Corresponding elemental mapping images of (d) $\text{Al}_m\text{Sn-ML}$, (e) $\text{Al}_l\text{Sn-ML}$, and (f) Sn-ML , where blue and yellow represent Sn and Al, respectively. (g–i) XRD patterns of $\text{Al}_m\text{Sn-ML}$, $\text{Al}_l\text{Sn-ML}$, and Sn-ML , respectively. (j) Engineering stress–strain curves of the three foils. (k) Hardness comparison of $\text{Al}_m\text{Sn-ML}$, $\text{Al}_l\text{Sn-ML}$, and Sn-ML . (l) Fracture morphologies of $\text{Al}_m\text{Sn-ML}$, $\text{Al}_l\text{Sn-ML}$, and Sn-ML after tensile testing.

Elemental mapping further verifies this difference in Al-phase spatial distribution. In Figure 1d, $\text{Al}_m\text{Sn-ML}$ shows discrete and staggered Al-rich regions dispersed within the Sn-rich matrix. These Al domains are elongated along the rolling direction but remain locally discontinuous, which is favorable for preserving through-thickness and in-plane Sn continuity. By comparison, the mapping image of $\text{Al}_l\text{Sn-ML}$ in Figure 1e displays several continuous Al-rich bands extending across the cross-section. This configuration can provide stronger mechanical support. For Sn-ML , only Sn is detected throughout the cross-section, confirming the absence of an Al reinforcing phase (Figure 1f). X-ray diffraction (XRD) patterns were collected from the surface and cross-section to confirm

the phase constitution of the laminated foils. As shown in Figure 1g–i, all samples mainly exhibit characteristic diffraction peaks of Sn, while Al-related peaks are observed in the Al-containing samples. No obvious impurity phase is detected, indicating that the rolling process primarily induces physical lamination and topological reconstruction rather than the formation of new intermetallic compounds.

The mechanical response of the three foils was then evaluated by tensile testing and hardness measurements. The engineering stress–strain curves in Figure 1j reveal that Sn-ML possesses the highest fracture strain, reflecting the intrinsic ductility of Sn-based laminated foils. However, its relatively low tensile strength suggests insufficient resistance to mechanical deformation, which may be unfavorable for suppressing lithiation-induced swelling. After introducing Al, both $\text{Al}_f\text{Sn-ML}$ and $\text{Al}_m\text{Sn-ML}$ exhibit enhanced strength but reduced ductility. In particular, $\text{Al}_f\text{Sn-ML}$ shows a sharp stress drop at a small strain, indicating that the continuous Al layer increases stiffness but also makes the laminate more prone to early fracture or interfacial failure under tensile loading. $\text{Al}_m\text{Sn-ML}$ maintains a higher strength than Sn-ML while avoiding an excessively continuous Al barrier, suggesting a more balanced mechanical response between reinforcement and deformability. The hardness results in Figure 1k further support this conclusion. Sn-ML exhibits the lowest hardness, while $\text{Al}_m\text{Sn-ML}$ shows an increased hardness after introducing the mesh-derived Al phase. $\text{Al}_f\text{Sn-ML}$ displays the highest hardness due to the presence of continuous Al foil layers. This trend confirms that Al effectively improves the local mechanical resistance of Sn-based laminated foils. The fracture morphologies after tensile testing provide further insight into the deformation behavior of the laminated foils (Figure 1l). Sn-ML shows a layered tearing morphology, consistent with its high ductility and lack of rigid reinforcement. $\text{Al}_f\text{Sn-ML}$ displays a more abrupt and planar fracture feature, which can be attributed to the strong constraint and stress concentration induced by the continuous Al interlayer. In contrast, $\text{Al}_m\text{Sn-ML}$ exhibits a more tortuous fracture morphology, indicating that the discontinuous Al domains can locally reinforce the Sn matrix and deflect fracture propagation without forming a fully continuous brittle pathway. Higher-magnification SEM images of the fracture morphologies are provided in Figure S1. This fracture behavior is consistent with the SEM/mapping results and supports the role of mesh-derived Al as a distributed mechanical skeleton.

3.2. Electrochemical Superiority of Discontinuous Al-Skeleton Reinforced Sn Foils

To evaluate whether the regulation of Al-phase continuity can effectively improve the electrochemical behavior of Sn-based laminated foil anodes, the cycling performance of different Al-mesh-derived laminates was first compared. The Al mesh sizes used in the screening are shown in Figure S2. As shown in Figure 2a, the sandwich-structured Sn/ Al_m /Sn electrode exhibits the best cycling performance and the highest CE. It achieves 193 cycles at 1 mA cm^{-2} and 2 mAh cm^{-2} , outperforming Sn/ Al_f /Sn and Sn/ Al_3 /Sn. This suggests that an appropriate Al-mesh topology is essential for balancing ion accessibility and mechanical support. The optimized Al_m -type Al mesh was selected to fabricate the $\text{Al}_m\text{Sn-ML}$ foil anode. Long-term cycling tests were then conducted to demonstrate its durability and electrochemical stability. At a current density of 1 mA cm^{-2} and an areal capacity of 2 mAh cm^{-2} , $\text{Al}_m\text{Sn-ML}$ maintains a CE exceeding 96.47% for 430 cycles, whereas Sn-ML and $\text{Al}_f\text{Sn-ML}$ suffer from rapid capacity decay and fail after only 173 and 123 cycles, respectively (Figure 2b). The average CE of $\text{Al}_m\text{Sn-ML}$ is 98.35%, significantly higher than that of $\text{Al}_f\text{Sn-ML}$ (96.49%). $\text{Al}_m\text{Sn-ML}$ shows the longest cycling life and the most stable CE, demonstrating that the discontinuous Al skeleton can effectively improve cycling reversibility while avoiding the severe transport penalty associated with continuous Al layers. To further examine the high-current cycling stability, the three electrodes were tested at 2 mA cm^{-2} with a fixed areal capacity of 2 mAh cm^{-2} . As shown in Figure S3, $\text{Al}_m\text{Sn-ML}$ maintains stable CE over 100 cycles under this increased current density. The corresponding GCD profiles show no obvious distortion of the charge/discharge plateaus or severe polarization growth during cycling, indicating that the discontinuous Al skeleton does not impose serious kinetic limitation under accelerated lithiation/delithiation conditions. This result further supports the balanced transport–mechanics response of $\text{Al}_m\text{Sn-ML}$.

The representative galvanostatic charge/discharge (GCD) profiles further reveal the difference in reaction reversibility among the three electrodes. As shown in Figure 2c, Sn-ML exhibits obvious profile evolution upon cycling, reflecting unstable alloying/dealloying reactions caused by continuous structural deformation. $\text{Al}_f\text{Sn-ML}$ shows more pronounced polarization and lower reversible capacity, consistent with the Li^+ transport limitation induced by the continuous Al interlayer. By contrast, $\text{Al}_m\text{Sn-ML}$ maintains more stable voltage profiles from the 10th to the 100th cycle, indicating improved reaction reversibility and reduced polarization during repeated lithiation and delithiation. This suggests that the mesh-derived discontinuous Al skeleton does not block the main Sn-based electrochemical reaction but instead helps stabilize the electrode structure during cycling. To further clarify the kinetic origin of the improved cycling behavior, electrochemical impedance spectroscopy was performed after different cycles (Figure S4). As shown in Figure 2d, Sn-ML exhibits relatively low charge-transfer

resistance (R_{ct}), with R_{ct} values of 13.26, 12.54, 11.92 Ω and 6.56 Ω after the 1st, 10th, 20th and 50th cycles, respectively. In contrast, Al_fSn -ML shows the highest R_{ct} among the three electrodes, decreasing from 67.83 Ω at the 1st cycle to 58.34 Ω , 45.03 Ω and 12.94 Ω after the 10th, 20th and 50th cycles. Although the continuous Al foil layer can provide strong mechanical confinement, its laterally continuous configuration substantially increases the interfacial/transport resistance, suggesting that the continuous Al layer acts as a kinetic barrier for Li^+ penetration. By comparison, Al_mSn -ML displays an intermediate R_{ct} , decreasing from 27.39 Ω to 20.52 Ω and 16.85 Ω from the 1st to the 20th cycle, and further to 8.97 Ω after the 50th cycle. Although the absolute R_{ct} of Al_mSn -ML remains slightly higher than that of Sn-ML during the measured cycles, the gap gradually narrows with cycling, indicating improved interfacial activation and stabilized charge-transfer kinetics. More importantly, Al_mSn -ML exhibits a much lower R_{ct} than Al_fSn -ML throughout cycling, confirming that the mesh-derived discontinuous Al skeleton avoids the severe transport penalty imposed by the continuous Al layer. Therefore, the electrochemical advantage of Al_mSn -ML does not arise from having the lowest initial charge-transfer resistance, but from achieving a balanced kinetic–mechanical response: sufficient Li^+ accessibility is maintained while the discontinuous Al skeleton provides mechanical reinforcement and suppresses structural degradation. The Li^+ diffusion coefficients extracted at selected cycles further support this conclusion (Figure S5). As shown in Figure 2e, Sn-ML shows apparent Li^+ diffusion coefficients of 6.55×10^{-15} and 6.71×10^{-14} $cm^2 s^{-1}$ at the 10th and 50th cycles, respectively. The increased apparent diffusion coefficient after cycling is likely associated with progressive structural loosening and crack formation, which increases electrolyte accessibility but simultaneously accelerates mechanical failure. Al_fSn -ML exhibits much lower diffusion coefficients of 0.31×10^{-16} and 0.48×10^{-14} $cm^2 s^{-1}$ at the 10th and 50th cycles, respectively, confirming the sluggish Li^+ transport caused by the continuous Al interlayer. In contrast, Al_mSn -ML maintains diffusion coefficients of 5.24×10^{-15} and 3.34×10^{-14} $cm^2 s^{-1}$ at the 10th and 50th cycles, respectively, which are much higher than those of Al_fSn -ML. Therefore, the discontinuous Al skeleton in Al_mSn -ML preserves effective Li^+ transport pathways through the continuous Sn network while preventing the excessive structural collapse observed in Sn-ML. The advantage of Al_mSn -ML becomes more evident under increased areal-capacity conditions. As summarized in Figure 2f, Al_mSn -ML delivers stable cycling for 430, 135, and 80 cycles at areal capacities of 2, 4, and 6 $mAh cm^{-2}$, respectively. Under the same conditions, Sn-ML only maintains 160, 80, and 60 cycles. The cycle life of Al_mSn -ML is therefore enhanced by approximately 169%, 69%, and 33% at 2, 4, and 6 $mAh cm^{-2}$, respectively. Higher areal capacity usually intensifies lithiation-induced swelling and stress accumulation in alloy-type foil anodes. Therefore, the improved high-loading durability of Al_mSn -ML demonstrates that the mesh-derived Al skeleton can effectively buffer the volume change of Sn without blocking Li^+ access. The performance of Al_mSn -ML was further benchmarked against previously reported Sn-based foil or alloy-type anodes. As shown in Figure 2g, the present Al_mSn -ML electrode occupies a competitive position in terms of cycle number and current density, especially considering its high areal-capacity operation.

The practical applicability of the optimized Al_mSn -ML electrode was further evaluated in full cells. When paired with LFP cathodes, the Al_mSn -ML full cell delivers an initial areal capacity of 2.5 $mAh cm^{-2}$ in the voltage range of 2–3.65 V and retains 91.51% of its capacity after 160 cycles at 0.5 C (1C=150 $mA g^{-1}$), corresponding to a retained areal capacity of approximately 2.29 $mAh cm^{-2}$ (Figure 2h). The corresponding GCD profiles are shown in Figure S6. The high initial capacity, long cycle life, and high retention of the Al_mSn -ML full cell confirm that the structural advantage of the discontinuous Al skeleton can be translated from Li half-cells to practical full-cell configurations. The superiority of Al_mSn -ML is also maintained when paired with high-loading NCM811 cathodes. As shown in Figure S7, the Al_mSn -ML/NCM811 full cell operates for 120 cycles at 0.5 C in the voltage range of 2.8–4.2 V with a capacity retention of 75.21%. Moreover, based on the detailed cell-stack parameters listed in Table S2, the Al_mSn -ML/NCM811 full cell delivers volumetric energy densities of 765.7 and 1005.3 $Wh L^{-1}$ at areal capacities of 2 and 4 $mAh cm^{-2}$, respectively. Compared with representative Sn-based and alloy-foil-anode full cells, Al_mSn -ML/NCM811 delivers competitive volumetric energy densities at both 2 and 4 $mAh cm^{-2}$, indicating that the discontinuous Al-skeleton design remains effective across different areal-capacity regimes (Figure S7). Overall, these electrochemical results reveal that the performance improvement of Al_mSn -ML does not arise from simply lowering the interfacial resistance or maximizing the apparent Li^+ diffusion coefficient. Instead, Al_mSn -ML achieves a more balanced electrochemical response. Its discontinuous Al skeleton provides mechanical support while maintaining sufficiently open Li^+ transport pathways through the continuous Sn network. This balance enables lower R_{ct} than the continuous Al-foil laminate, higher structural stability than Sn-ML, improved high-areal-capacity cyclability, and superior full-cell performance. Thus, the spatial continuity of the Al phase is a decisive factor governing the electrochemical performance of Sn/Al laminated foil anodes.

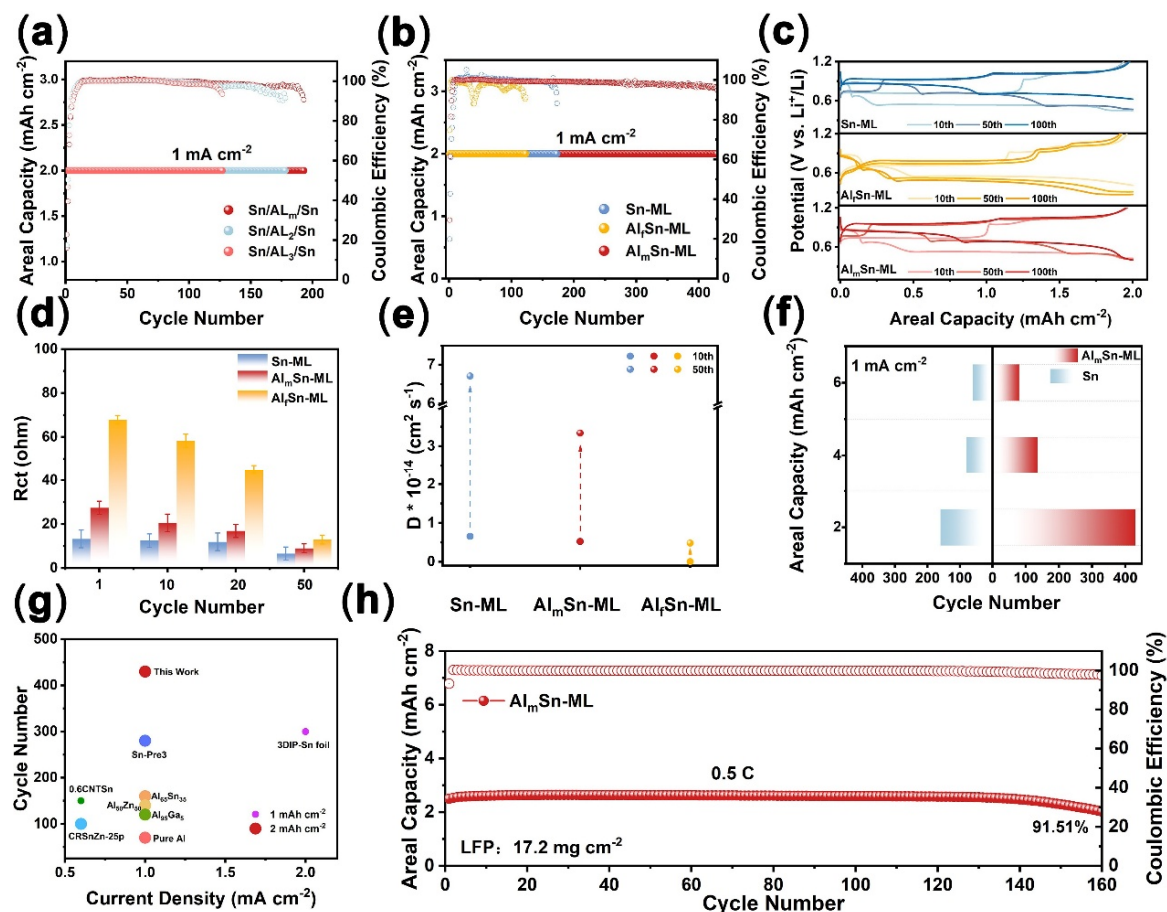


Figure 2. (a) Cycling performance of Sn/Al_m/Sn foils prepared with Al meshes of different pore sizes at 1 mA cm⁻². (b) Cycling performance of Sn-ML, Al_fSn-ML, and Al_mSn-ML electrodes at 1 mA cm⁻² with areal capacity of 2 mAh cm⁻². (c) Representative GCD profiles of Sn-ML, Al_fSn-ML, and Al_mSn-ML electrodes at selected cycles. (d) Rct evolution of Sn-ML, Al_mSn-ML, and Al_fSn-ML after the 1st, 10th, 20th, and 50th cycles. (e) Li⁺ diffusion coefficients of Sn-ML, Al_fSn-ML, and Al_mSn-ML electrodes at the 10th and 50th cycles. (f) Cycling performance comparison of Sn-ML and Al_mSn-ML electrodes under different areal capacities. (g) Performance comparison of Al_mSn-ML with previously reported Sn-based foil/alloy anodes (Table S1 for detailed performance parameters). (h) Full-cell cycling performance of Al_mSn-ML paired with high-loading LFP cathodes at 0.5 C.

3.3. Topology-Governed Lithiation Pathway and Phase Evolution

To clarify the topology-governed lithiation behavior of the laminated foils, Density Functional Theory (DFT) calculations, electrochemical analysis, ex-situ XRD, and post-cycling SEM were combined to reveal the reaction pathway and structural evolution. The topology-dependent lithiation scenarios of Sn-ML, Al_fSn-ML, and Al_mSn-ML are schematically summarized in Figure S8. Briefly, Sn-ML allows direct Li⁺ access but suffers from severe alloying-induced deformation, whereas Al_fSn-ML provides strong mechanical confinement at the expense of through-thickness Li⁺ transport. In contrast, Al_mSn-ML preserves a continuous Sn reaction network while introducing a discontinuous Al skeleton for local mechanical support.

DFT calculations were first performed to evaluate Li affinity on different sites (Figure 3a). The Li adsorption energies on Al surface, Sn/Al interface, and Sn surface are -0.73, -1.42, and -2.01 eV, respectively, indicating that Li preferentially interacts with Sn and the Sn/Al interface rather than pure Al. This result suggests a Sn-dominated lithiation pathway, where the Al phase mainly acts as a mechanical skeleton while the Sn/Al interface assists Li accommodation. Therefore, transforming Al from a continuous layer into a discontinuous skeleton is beneficial for maintaining Sn-based lithiation pathways and avoiding a transport-blocking Al barrier. The GCD profiles in Figure 3b show the typical alloying/dealloying features of Sn-based anodes. During discharge, Sn undergoes multi-step alloying with Li to form Li_xSn phases. Sn can sequentially transform into Li₂Sn₅, LiSn, and more Li-rich Li_xSn phases such as Li₇Sn₃/Li₇Sn₂ during lithiation, while these phases partially reverse during delithiation [31–33]. Compared with Sn-ML, Al_fSn-ML delivers a much lower capacity and exhibits larger polarization, indicating that the continuous Al layer restricts the utilization of the active Sn phase. However,

Al_mSn -ML preserves clear charge/discharge plateaus and a relatively high reversible capacity, suggesting that the discontinuous Al skeleton does not block the main Sn alloying reaction. The lower apparent capacity and slightly larger polarization of Al_mSn -ML compared with Sn-ML can be attributed to the partial replacement of active Sn by the Al skeleton and the increased local transport tortuosity introduced by the composite architecture. However, unlike the continuous Al layer in Al_fSn -ML, the discontinuous Al skeleton does not form a severe through-thickness barrier. This is supported by the lower R_{ct} , higher Li^+ diffusion coefficient, and more pronounced redox peaks of Al_mSn -ML compared with Al_fSn -ML. Therefore, the polarization of Al_mSn -ML reflects a necessary trade-off between mechanical reinforcement and ion transport, while its long-term cycling stability benefits from the resulting transport–mechanics balance.

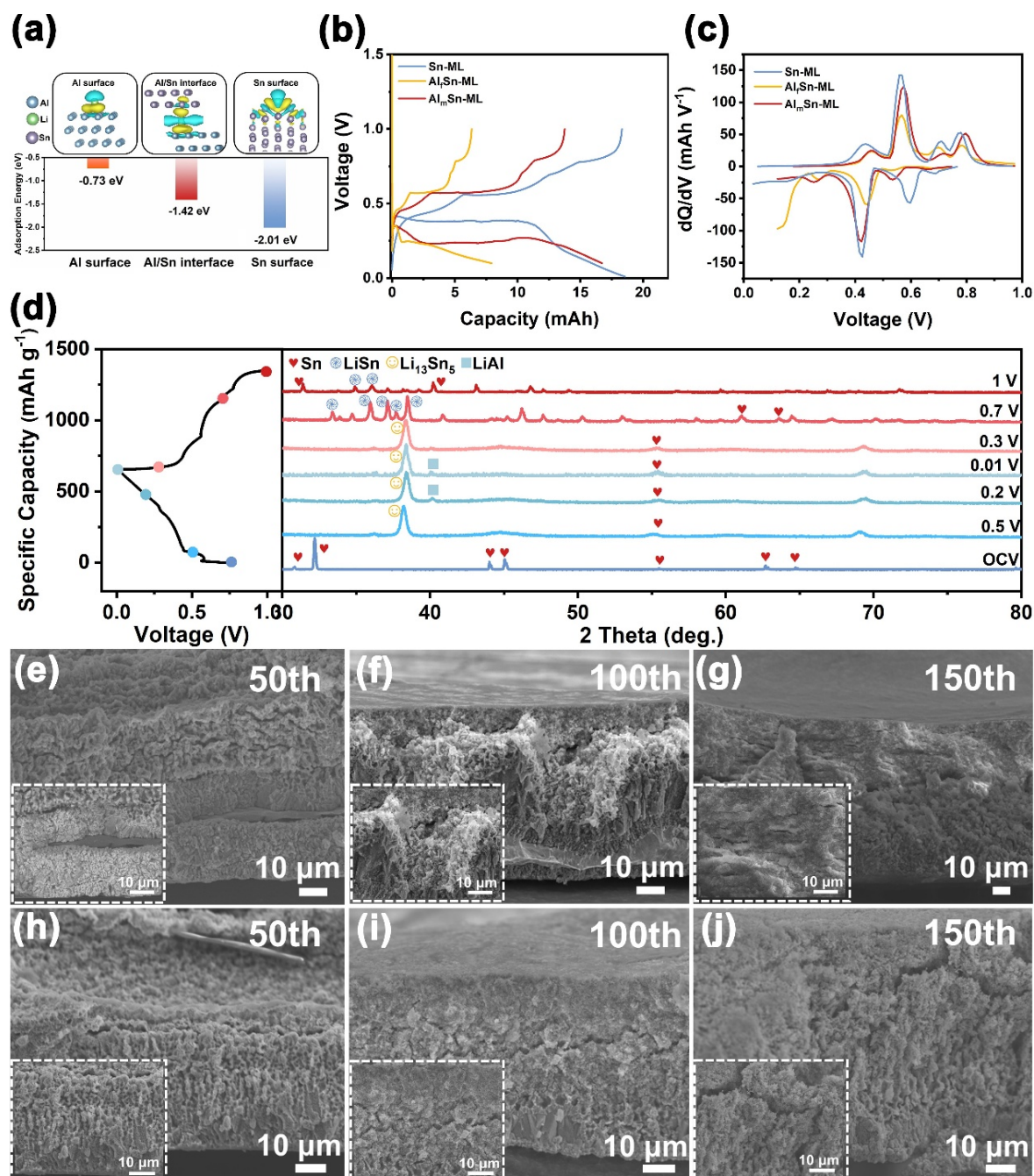


Figure 3. (a) DFT-calculated Li adsorption energies and charge-density-difference maps for Li adsorbed on the Al surface, Sn/Al interface, and Sn surface. (b) Initial GCD profiles of the three electrodes. (c) Corresponding dQ/dV profiles showing the alloying/dealloying reactions of Sn-based phases. (d) Ex-situ XRD patterns of Al_mSn -ML collected at selected discharge/charge states. Cross-sectional SEM images of Al_mSn -ML (e–g) and Sn-ML (h–j) after 50, 100, and 150 cycles, respectively.

The dQ/dV profiles in Figure 3c further reveal the reaction sequence. For Sn-based electrodes, reduction peaks around approximately 0.65, 0.40, and near 0.01 V are commonly assigned to stepwise Li–Sn alloying reactions, while oxidation peaks around 0.50, 0.59, 0.71, and 0.79 V correspond to the dealloying of Li_xSn phases [34–36]. In the present samples, the cathodic feature in the intermediate-voltage region can therefore be attributed to the conversion of Sn into Sn-rich Li_xSn phases [37]. The lower-voltage peak/shoulder corresponds to deeper lithiation toward Li-rich Li_xSn phases [38]. For Al-containing samples, the electrochemical signal of Al lithiation may overlap with the low-voltage Sn alloying reaction. β -LiAl formation generally occurs at a low potential, with a reduction feature near 0.2–0.3 V [39–42]. Therefore, the low-voltage response of Al_mSn -ML should be interpreted as a coupled process involving deep lithiation of Sn and partial activation of the Al phase. Importantly, the dQ/dV peaks of Al_mSn -ML are more pronounced than those of Al_fSn -ML, confirming that the discontinuous Al skeleton allows the Sn alloying reaction to proceed more effectively than the continuous Al foil laminate. Ex-situ XRD was then used to directly track the crystalline phase evolution during discharge and charge. As shown in Figure 3d, the pristine electrode mainly exhibits Sn diffraction peaks at the open-circuit state. When discharged to 0.5 V, the intensity of Sn peaks decreases, indicating the consumption of crystalline Sn and the onset of Li–Sn alloy formation. Upon further discharge to 0.2 and 0.01 V, the Sn peaks are further weakened, while diffraction signals assigned to Li–Sn alloy phases become evident. In particular, the appearance of $\text{Li}_{13}\text{Sn}_5$ -related reflections indicates that the Sn phase undergoes deeper lithiation into Li-rich Li_xSn phases [43,44]. Meanwhile, LiAl-related peaks appear at low potentials, demonstrating that the Al phase is not completely electrochemically inert under deep lithiation. However, because the LiAl signal appears mainly after substantial Sn lithiation, Al is more likely activated after or together with the deep lithiation stage rather than serving as the primary initial Li-storage phase.

During the subsequent charge process, the diffraction signals of Li–Sn alloy phases gradually weaken as the voltage increases to 0.3, 0.7, and 1.0 V, while Sn-related peaks recover. This reversible change verifies the dealloying reaction from Li_xSn back toward Sn. The partial recovery of Sn diffraction peaks at high voltage confirms that the main capacity contribution originates from the reversible $\text{Sn} \leftrightarrow \text{Li}_x\text{Sn}$ alloying/dealloying process. At the same time, the evolution of LiAl-related peaks indicates that Al participates locally in the lithiation/delithiation process, especially in regions close to the Sn/Al interface. Specifically, the appearance of weak LiAl-related reflections at deep lithiation states indicates that Al is not completely electrochemically inert. However, because these LiAl signals emerge mainly after substantial Sn lithiation and weaken during subsequent charging, Al is considered to contribute partially and reversibly at low potentials rather than serving as the dominant Li-storage phase. Therefore, the primary capacity of Al_mSn -ML originates from the reversible $\text{Sn} \leftrightarrow \text{Li}_x\text{Sn}$ alloying/dealloying process, while the Al skeleton provides mechanical support with limited interfacial electrochemical participation. Therefore, the ex-situ XRD results provide direct phase evidence for a sequential and coupled lithiation mechanism. Sn is preferentially lithiated to form Li_xSn phases, while the Al phase is activated mainly at lower potentials and contributes to interfacial stabilization rather than acting as the dominant capacity source.

The cross-sectional SEM images after cycling further connect the phase evolution with structural stability. For Al_mSn -ML, the cross-sections after 50, 100, and 150 cycles still maintain a relatively integrated layered morphology (Figure 3e–g). Although a porous and roughened lithiated region is formed due to repeated Sn alloying/dealloying, no catastrophic through-thickness cracking or complete structural pulverization is observed. The residual layered framework suggests that the discontinuous Al skeleton redistribute local deformation and suppress the continuous propagation of lithiation-induced cracks. By comparison, Sn-ML displays more severe structural roughening and loosening after cycling (Figure 3h–j), indicating that the pure multilayer Sn structure lacks sufficient internal mechanical support to withstand repeated volume expansion. The post-cycling morphology of Al_fSn -ML further confirms the adverse effect of excessive Al-phase continuity (Figures S9 and S10). After 10–100 cycles, Al_fSn -ML develops a porous and pulverized reaction layer near the electrolyte-facing side, while the underlying laminate remains relatively compact, revealing an asymmetric lithiation process across the thickness direction. This structural heterogeneity indicates that the continuous Al interlayer restricts Li^+ penetration into deeper Sn layers, causing localized Sn pulverization and interfacial separation. Therefore, Sn-ML suffers from insufficient mechanical confinement, whereas Al_fSn -ML suffers from transport obstruction induced by the continuous Al layer. In contrast, Al_mSn -ML avoids both failure modes by combining a continuous Sn reaction network with a discontinuous Al skeleton, thereby maintaining more homogeneous lithiation and improved structural integrity during cycling. These results clarify the role of Al-phase topology in regulating lithiation. In Al_fSn -ML, the continuous Al foil layer provides mechanical confinement but also delays Li^+ penetration, leading to sluggish reaction kinetics and incomplete utilization of Sn. In Sn-ML, Li^+ transport is not strongly blocked, but the absence of a reinforcing phase causes severe crack development and structural instability. Al_mSn -ML resolves this contradiction by transforming the Al phase into a discontinuous skeleton. This structure maintains a continuous

Sn reaction network, enables stepwise $\text{Sn} \leftrightarrow \text{Li}_x\text{Sn}$ phase transformation, allows partial low-voltage LiAl formation at the Sn/Al interface, and mitigates cycling-induced structural collapse. The post-cycling cross-sectional elemental mapping further confirms the structural persistence of the Al mesh-derived framework. As shown in Figure S11, the Sn signal remains broadly distributed throughout the cycled electrode, while the Al signal is still present as discontinuous strip-/island-like regions embedded in the Sn-rich matrix. The overlay image clearly shows that the Al phase does not evolve into a continuous through-thickness layer after repeated lithiation/delithiation, but instead retains its discontinuous skeletal character. This result provides direct evidence that the Al mesh-derived skeleton remains effective during cycling, thereby supporting its dual role in maintaining mechanical support while preserving open Li^+ transport pathways. Thus, the lithiation mechanism is governed not only by the intrinsic alloying chemistry of Sn and Al, but also by the spatial continuity of the Al phase within the laminated foil.

3.4. Swelling Regulation and Stress Redistribution Enabled by Discontinuous Al Skeletons

The dimensional evolution of the cycled foils was further examined to reveal how the Al-phase topology regulates lithiation-induced swelling and stress accumulation. As shown in Figure 4a, Sn-ML exhibits severe surface wrinkling and morphological distortion after cycling, indicating substantial mechanical instability caused by repeated Sn alloying/dealloying. $\text{Al}_f\text{Sn-ML}$ shows a more constrained macroscopic morphology, but its continuous Al interlayers also lead to heterogeneous deformation. In contrast, $\text{Al}_m\text{Sn-ML}$ maintains a more integrated and compact foil morphology after cycling, suggesting that the mesh-derived Al skeleton can provide effective mechanical buffering while avoiding catastrophic structural collapse.

The thickness evolution provides direct evidence for the different swelling behaviors (Figure 4b). Starting from an initial foil thickness of 50 μm , Sn-ML rapidly expands to 205.5, 216.5, 221.5, and 259.5 μm after 10, 20, 50, and 100 cycles, respectively. This corresponds to a thickness increase of 311.0%, 333.0%, 343.0%, and 419.0%, confirming the severe through-thickness expansion of pure multilayer Sn. After introducing the mesh-derived Al skeleton, the thickness of $\text{Al}_m\text{Sn-ML}$ is significantly reduced to 155.2, 159.25, 164.0, and 174.8 μm at the same cycle numbers, corresponding to thickness increases of 210.4%, 218.5%, 228.0%, and 249.6%. Compared with Sn-ML, $\text{Al}_m\text{Sn-ML}$ reduces the 100-cycle thickness by approximately 32.6%, demonstrating that the discontinuous Al skeleton can effectively suppress the accumulation of lithiation-induced expansion. It should be noted that although $\text{Al}_m\text{Sn-ML}$ significantly suppresses swelling compared with Sn-ML, its thickness still increases from 50 to 174.8 μm after 100 cycles. Such expansion may influence stack-pressure distribution and interfacial contact in practical pouch-cell configurations. Therefore, the present Al-phase-topology strategy should be viewed as an effective approach to mitigate, rather than completely eliminate, alloying-induced swelling. Further optimization of electrode thickness, areal loading, stack pressure, and cell-level swelling tolerance will be necessary for practical cell implementation.

$\text{Al}_f\text{Sn-ML}$ exhibits the smallest apparent thickness, increasing from 50 μm to 96.85, 99.3, 104.5, and 108.7 μm after 10, 20, 50, and 100 cycles, respectively (The swelling ratio statistics are shown in Figure S12). However, this low thickness growth should not be simply regarded as superior swelling regulation. Combined with its high R_{ct} and sluggish Li^+ diffusion, the suppressed expansion of $\text{Al}_f\text{Sn-ML}$ is more likely associated with the transport-blocking effect of continuous Al layers and incomplete utilization of the Sn active phase. Therefore, $\text{Al}_f\text{Sn-ML}$ represents an over-confined structure, where mechanical constraint is achieved at the expense of lithiation accessibility.

The diameter evolution further clarifies the deformation mode of the three foils (Figure 4c). For Sn-ML, the diameter increases from the initial 10 mm to 10.57, 10.75, 10.81, and 11.12 mm after 10, 20, 50, and 100 cycles, corresponding to an in-plane expansion of 5.79%, 7.54%, 8.14%, and 11.85%, respectively. $\text{Al}_m\text{Sn-ML}$ shows much smaller diameter growth, increasing only to 10.26, 10.28, 10.29, and 10.37 mm, corresponding to 2.58%, 2.84%, 2.97%, and 3.66%. $\text{Al}_f\text{Sn-ML}$ exhibits intermediate diameter expansion of 4.32%, 5.15%, 6.00%, and 6.70% (The swelling ratio statistics are shown in Figure S13). These results show that the dimensional instability of Sn-ML is not limited to thickness swelling, but also involves obvious in-plane deformation. The much smaller diameter increase of $\text{Al}_m\text{Sn-ML}$ indicates that the discontinuous Al skeleton suppresses both through-thickness swelling and lateral distortion, thereby maintaining better dimensional integrity during cycling.

To probe the lithiation-induced internal strain state, partial lithiation–relaxation tests were performed (Figure 4d,e). The theoretical equilibrium potential of $\text{Li}_{13}\text{Sn}_5$ is marked at approximately 0.485 V [31,45,46]. After relaxation, $\text{Al}_m\text{Sn-ML}$ shows the highest potential and approaches this equilibrium value most closely, indicating a lower residual lithiation driving force and more effective stress relaxation [47,48]. In contrast, Sn-ML exhibits a lower relaxed potential, which is associated with severe expansion and strain accumulation in the unsupported Sn layers. $\text{Al}_f\text{Sn-ML}$ shows the largest deviation from the equilibrium potential, suggesting that the continuous Al layer

hinders Li^+ redistribution and leads to a more pronounced non-equilibrium lithiation state [48]. These results indicate that the discontinuous Al skeleton in $\text{Al}_m\text{Sn-ML}$ promotes more homogeneous lithiation and mitigates stress accumulation during the alloying reaction. The controlled lithiation curves in Figure 4f further reveal the effect of the discontinuous Al skeleton on Li uptake behavior. To reach the same target lithiation depth of 5 mAh, Sn-ML completes lithiation within 14.0 h and exhibits a relatively larger current response. Such rapid Li uptake is consistent with the absence of a transport barrier, but it also produces abrupt local volume expansion and promotes stress concentration in the Sn layers. In comparison, $\text{Al}_m\text{Sn-ML}$ requires 16.9 h to reach the same lithiation depth, and its current remains nearly constant during the lithiation process. This more stable current response indicates a more regulated Li insertion process, in which the discontinuous Al skeleton moderates local deformation and suppresses sudden structural activation caused by cracking. Importantly, this moderated lithiation behavior is different from the sluggish transport observed in $\text{Al}_f\text{Sn-ML}$; $\text{Al}_m\text{Sn-ML}$ still maintains much lower R_{ct} and higher Li^+ diffusion than $\text{Al}_f\text{Sn-ML}$, indicating that its slower lithiation reflects mechanical buffering rather than severe kinetic blockage.

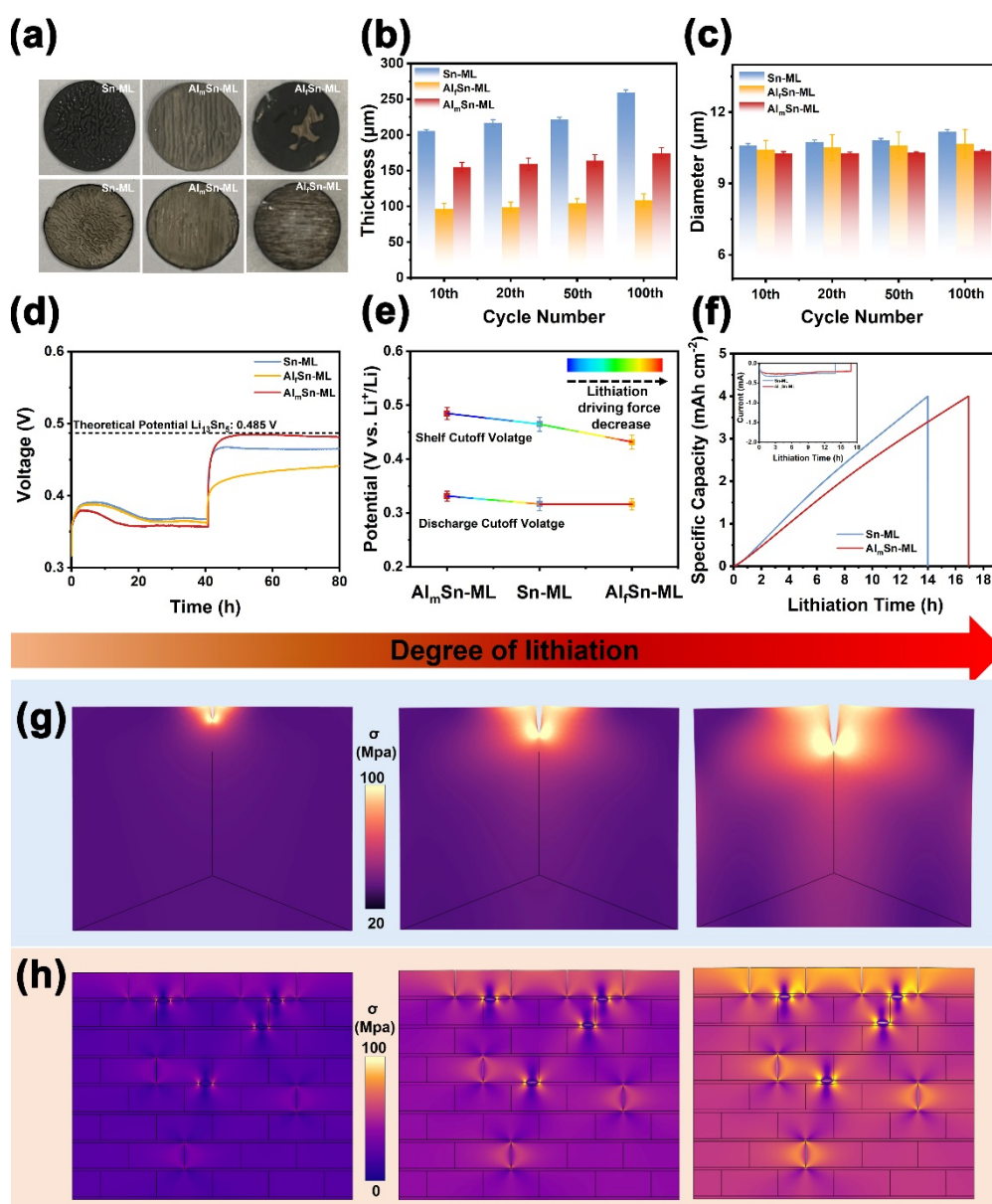


Figure 4. (a) Optical images of Sn-ML, $\text{Al}_m\text{Sn-ML}$, and $\text{Al}_f\text{Sn-ML}$ electrodes after 100 cycles. (b,c) Thickness and diameter evolution of Sn-ML, $\text{Al}_m\text{Sn-ML}$, and $\text{Al}_f\text{Sn-ML}$ after 10, 20, 50, and 100 cycles, respectively. (d) Voltage relaxation profiles of the three electrodes after controlled lithiation. (e) Comparison of discharge cutoff voltage and shelf cutoff voltage. (f) Lithiation capacity and corresponding current evolution of Sn-ML and $\text{Al}_m\text{Sn-ML}$ during controlled lithiation to a fixed capacity. (g,h) COMSOL-simulated lithiation-induced stress-field evolution of Sn-ML and $\text{Al}_m\text{Sn-ML}$ at different degrees of lithiation, respectively.

Finite-element simulation was further employed to visualize the lithiation-induced stress evolution in Sn-ML and Al_mSn-ML. In Sn-ML, the stress field is strongly localized near the lithiated surface region at the initial stage and progressively extends into the bulk with increasing degree of lithiation (Figure 4g). The formation of a continuous high-stress region indicates that the pure Sn multilayer lacks an effective internal framework to dissipate lithiation-induced strain. Such localized stress accumulation is expected to promote crack initiation and structural pulverization, consistent with the severe swelling and morphological distortion observed experimentally. By contrast, Al_mSn-ML exhibits a more dispersed stress distribution (Figure 4h). The high-stress regions are spatially partitioned around the discontinuous Al segments instead of propagating as a continuous stress band. This suggests that the mesh-derived Al skeleton functions as a local stress-transfer framework, redistributing deformation and interrupting long-range stress propagation in the Sn matrix. Therefore, the discontinuous Al skeleton simultaneously preserves open Li⁺ pathways and improves mechanical strain accommodation, enabling Al_mSn-ML to suppress swelling without sacrificing lithiation accessibility.

Overall, these results demonstrate that the swelling behavior of Sn/Al laminated foils is governed by the spatial continuity of the Al phase. Sn-ML allows relatively facile Li⁺ access but lacks mechanical confinement, leading to severe thickness growth, lateral expansion, and structural distortion. Al_fSn-ML provides strong apparent confinement, but the continuous Al layers restrict Li⁺ transport and cause incomplete or heterogeneous lithiation. Al_mSn-ML achieves a more favorable balance: the discontinuous Al skeleton redistributes lithiation-induced deformation, suppresses stress concentration, and limits both thickness and diameter expansion while preserving effective Sn-based lithiation pathways. Therefore, the mesh-derived discontinuous Al skeleton enables swelling regulation and stress redistribution without imposing the severe transport penalty of continuous Al layers.

4. Conclusions

This study demonstrates that the failure of Sn-based foil anodes can be effectively regulated by tailoring the spatial continuity of the reinforcing Al phase. Rather than simply increasing the amount of mechanical support, the critical design principle is to balance Li⁺ accessibility with alloying-induced strain confinement. Pure Sn multilayers suffer from insufficient mechanical support, whereas continuous Al/Sn laminates over-constrain the structure and restrict through-thickness Li⁺ transport. The mesh-derived Al_mSn-ML architecture resolves this contradiction by maintaining a continuous Sn lithiation network and introducing a discontinuous Al skeleton for local stress redistribution. This topology-guided structure enables reversible Sn-dominated alloying, mitigates crack propagation and swelling, and preserves electrode integrity during cycling. Benefiting from this transport–mechanics synergy, Al_mSn-ML delivers stable high-areal-capacity cycling over 430 cycles at 2 mAh cm^{−2} and 1 mA cm^{−2}. Its structural advantage is retained in full cells, with Al_mSn-ML/LFP cells showing 91.51% capacity retention after 160 cycles. These findings establish Al-phase topology regulation as an effective route to transform mechanically fragile alloy foils into ion-accessible, dimensionally stable, and full-cell-compatible anodes for high-energy lithium-ion batteries.

Supplementary Materials

The additional data and information can be downloaded at: <https://media.scilit.com/articles/others/2607221345320726/eChem-26050193-SM.pdf>. References [49–63] are cited in supplementary materials.

Author Contributions

G.W., H.G. and X.Z.: conceived and supervised the research; Y.J. and J.L.: performed the experiments; C.W.: contributed to data interpretation and collectively wrote the manuscript with assistance from all other authors. All authors have read and agreed to the published version of the manuscript.

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The data that support the findings of this study are available from the corresponding author upon reasonable request.

Conflicts of Interest

The authors declare no conflict of interest.

Use of AI and AI-Assisted Technologies

No AI tools were utilized for this paper.

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