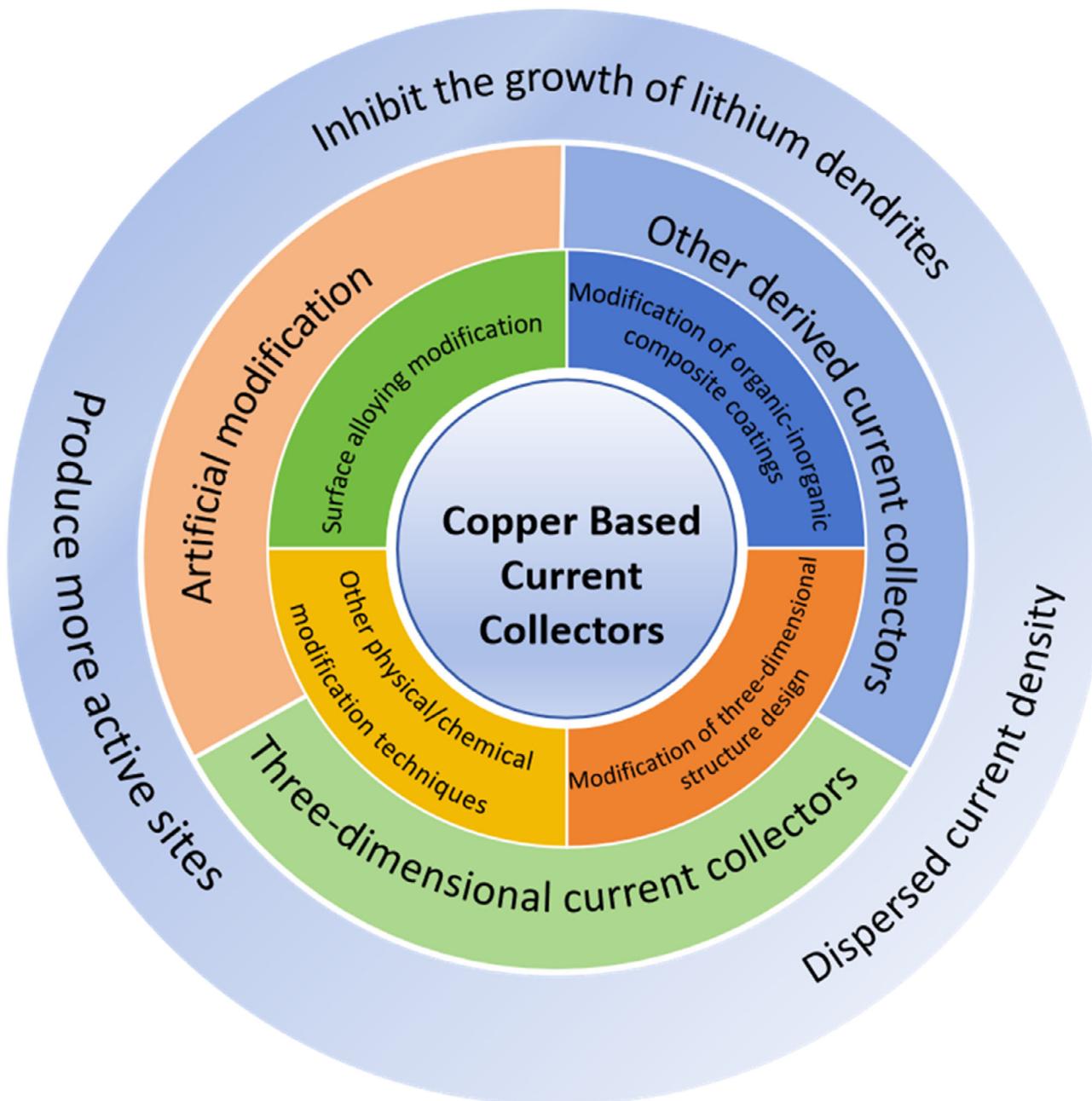


Recent Advances in Current Collectors for Anode-Free Lithium Metal Batteries

Yang Liu, Lina Pan, and Peng Huang*



Lithium metal anodes are promising for next-generation batteries due to their ultrahigh theoretical capacity (3860 mAh g^{-1}) and low redox potential. Anode-free lithium metal batteries offer enhanced energy density by eliminating pre lithiated anodes. However, lithium's high reactivity causes dendrite growth, risking short circuits, while the unstable solid electrolyte interphase consumes active lithium and electrolyte, reducing cycle life and Coulombic efficiency

(CE). Commercial copper foil current collectors, though conductive, promote uneven lithium deposition due to their smooth surfaces. Therefore, the modification of the Cu-based current collectors to reduce local current density, inhibit dendrite growth, and improve CE has been widely studied and reported. Herein, methods for modifying Cu-based current collectors are systematically summarized to guide the rational design of these modifications.

1. Introduction

The global energy structure is undergoing an accelerated transition toward low-carbon solutions. Lithium-ion batteries (LIBs), with their exceptional energy density and long cycle life, have been widely employed in electric vehicles and grid-scale energy storage systems. In 2024, global LIB shipments reached 1545.1 GWh, reflecting explosive industry growth.^[1–5] However, current LIB technology faces dual bottlenecks in material systems. On one hand, the theoretical capacity of conventional graphite anodes is limited to 372 mAh g^{-1} , nearing its energy density ceiling amid the demand for electric vehicles to achieve a driving range exceeding 1000 km.^[6,7] On the other hand, lithium metal batteries (LMBs), regarded as next-generation battery technology, offer a theoretical capacity of 3860 mAh g^{-1} but encounter severe challenges. During charging, uncontrolled lithium dendrite growth—resembling tree branches—can pierce separators and trigger short-circuit risks. Concurrently, the repeated fracture and reconstruction of the solid solid electrolyte interphase (SEI) on electrode surfaces degrade capacity rapidly and lower cycle life considerably below commercial requirements, hindering practical deployment.^[8,9]

Dead lithium formation primarily arises from the nonuniform deposition and stripping of lithium metal and the instability of the SEI.^[10] During battery cycling, uneven current distribution promotes the growth of lithium dendrites or moss-like structures on the current collector, which fracture due to volume expansion and expose fresh lithium to the electrolyte. This triggers continuous SEI breakdown and reconstruction, consuming active lithium and encasing it in insulating layers, thus disconnecting it from the electrical circuit. Moreover, localized high current densities during lithium stripping lead to incomplete dissolution, leaving behind electrochemically inactive lithium particles. The accumulation of dead lithium deteriorates Coulombic efficiency (CE), capacity retention, and interfacial stability, underscoring the need for strategies like 3D current collector engineering or SEI modification to mitigate its formation and enhance battery durability.

In situ X-ray diffraction (XRD) technology plays a crucial role in characterizing lithium deposition on copper current collectors, as it enables real-time, in situ structural insights into lithium

plating/stripping behavior in anode-free batteries by tracking the evolution of diffraction signals from specific crystallographic planes of lithium metal. Zhu et al. employed in situ XRD to monitor the dynamic changes of lithium in Cu//NCM811 batteries. They observed that during charging, lithium deposition on the copper surface generated a Li (110) diffraction peak, whose intensity exhibited an inverse correlation with the lithium content in the NCM811 cathode during charge/discharge cycles, directly demonstrating lithium migration between the electrodes.^[11] However, even after discharge at a low rate of C/24, residual Li (110) peaks persisted, indicating the formation of "dead lithium"—electrochemically inactive lithium due to loss of electrical contact or insulation by the SEI layer, a key factor contributing to low CE.^[12] Further analysis incorporating electrolyte types revealed that carbonate-based electrolytes exhibited higher residual Li (110) peak intensities compared to ether-based systems,^[13] demonstrating that in situ XRD can quantify the impact of different electrolytes on dead lithium formation, providing critical data for interface optimization. Moreover, the attenuation of Li (110) peak intensity during cycling correlated with capacity fade, confirming that dead lithium accumulation is a core degradation mechanism. The tracking logic of the Li (110) peak via in situ XRD can be extended—by analyzing changes in diffraction peak position, intensity, and width, key parameters such as lithium deposition orientation, grain size, and structural stability can be resolved. This approach provides theoretical guidance for designing lithophilic current collectors (e.g., by controlling preferred crystallographic growth orientations), ultimately suppressing dendrite formation and minimizing dead lithium accumulation.

In this context, anode-free lithium metal batteries (AFLMBs) have emerged. This innovative technology deviates from the conventional dual-active-electrode design, employing current collectors directly as the anode. During initial charging, lithium ions migrate from the cathode and are deposited onto the current collector, forming a lithium metal anode.^[14] This design simplifies the cell architecture, reduces manufacturing costs, and fundamentally mitigates lithium dendrite growth caused by anode materials. Furthermore, optimizing electrolyte formulations and modifying current collector surfaces can enhance the uniformity and stability of lithium deposition. Notably, AFLMBs offer a theoretical specific energy of $>500 \text{ Wh kg}^{-1}$ —far surpassing those of existing LIBs—positioning them as promising core technology for future energy-dense battery systems.

In anodized batteries, lithium differs significantly from other elements (sodium, zinc, magnesium, and aluminum) in the application

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of CC. Lithium usually uses copper current collector, but the lithium affinity needs to be improved by 3D structure design (such as gradient porous copper HLT-L-CF), surface alloying (such as Sn, Zn), or artificial SEI layer (such as GO@PVDF-HFP) to suppress dendrite growth.^[15] The theoretical energy density can reach 423 Wh kg⁻¹. However, the CE is limited by the stability of the SEI, usually ranging from 90% to 97.6%. Sodium is mostly used with aluminum or copper current collectors. The deposition uniformity needs to be improved through sodium affinity modification, such as antimony/antimony-zinc alloy heterostructures and 3D carbon-based materials (such as O-CCF). The CE can reach 99.7%, the energy density is 150–270 Wh kg⁻¹, and the cost is 15.1% lower than that of lithium batteries. Zinc commonly uses copper or aluminum current collectors. By regulating the electric field distribution through the antimony/antimony-zinc alloy heterogeneous interface, an ultra-high area capacity of 200 mAh cm⁻² is achieved, with a CE of 98.5%, an energy density of 274 Wh kg⁻¹, and a cycle life of over 400 times.^[16] Magnesium tends to use copper current collectors coated with gold or titanium, and the epitaxial growth is guided by lattice matching (for example, the lattice mismatch between gold and magnesium is less than 15%), while aluminum, due to its easy reaction with the electrolyte, requires the use of corrosion-resistant current collectors such as molybdenum or graphite paper, but the development of its anode-free system is still not mature. Overall, the modification of lithium current collectors focuses on lithium affinity and SEI stability, with significant advantages in energy density but high costs. Elements such as sodium and zinc are modified with low-cost materials to balance cost and performance, making them more suitable for large-scale energy storage scenarios.^[17]

Despite this potential, AFLMBs still face critical challenges. Repeated lithium stripping and deposition cycles induce sharp dendrite growth, continuous SEI degradation, and an abrupt interfacial impedance increase. These effects result in reduced CE, shortened cycle life, and localized overheating during fast charging.^[18–21] To address these issues, research efforts have focused on electrolyte composition optimization, separator modification, and current

collector surface engineering. Traditional copper foil CCs, while widely used, face challenges in lithium dendrite suppression and material costs. Recent innovations focus on lightweight and scalable alternatives. For instance, Oak Ridge National Laboratory (ORNL) developed a metallized polymer CC that integrates ultra-thin copper/aluminum layers with a polymer core, reducing metal usage by 85% and weight by 75% while enhancing energy density by 27%. This design, compatible with roll-to-roll production, achieves 10-minute fast charging and incorporates a built-in safety mechanism that reduces fire risks by 90% through polymer melting during short circuits. Material costs for such hybrid CCs are projected to be 80% lower than conventional copper foils due to minimal metal consumption.^[22] In lithium-metal batteries, MXene-based CCs have shown promise in suppressing dendrite growth through homogeneous lithium nucleation and stable solid–electrolyte interphase formation. While MXene production remains costly ($\approx \$200 \text{ kg}^{-1}$ for $\text{Ti}_3\text{C}_2\text{T}_{x}$), its high conductivity and mechanical robustness make it a viable candidate for next-generation high-energy-density cells. For sodium-ion batteries, aluminum CCs—costing 20%–30% less than copper—are gaining traction, with CATL's "Naxtra" chemistry demonstrating 175 Wh kg⁻¹ energy density and >10,000 cycles in 2025 prototypes. This shift aligns with sodium's abundance and reduced reliance on critical metals like copper.^[23] Cost analysis reveals that material costs dominate traditional CCs (60% for electrolytic copper), while composite designs allocate significant expenses to advanced coatings and scalable processes.^[24] For example, atomic layer deposition (ALD) of protective layers on polymer substrates enhances stability but increases equipment depreciation costs. Conversely, metallized polymer CCs achieve cost parity with conventional foils at high volumes due to simplified roll-to-roll fabrication.

Among these, current collector modification, in particular, offers precise control over the lithium metal–current collector interface, enabling lithium deposition regulation, dendrite suppression, and SEI stabilization. Compared with indirect electrolyte modifications and physical barrier-based separator strategies, tailored current collector designs provide targeted solutions for



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interfacial defects, working synergistically with electrode structures to establish stable electrochemical systems. Their modified solid-state layers exhibit superior durability than electrolyte additives or modified separators, demonstrating unique advantages in specificity, design flexibility, and interfacial stability.

In the field of lithium batteries, the traditional current collector manufacturing process has its own characteristics and costs. There are two main types of traditional copper foil manufacturing processes: rolling and electrolysis. The calendering process uses high-precision copper strip as raw material, after multiple rolls, annealing, relying on high-precision rolling mills, followed by degreasing, antioxidation, and other treatments, the process is extremely complex, resulting in high costs, and is only used in specific scenarios. The electrolysis process is to make copper into electrolyte, which is electrodeposited to form the original foil, and then pickled, surface treated and slotted, the equipment requirements are relatively simple, the cost is low, and it has become the mainstream choice, and with the thickness thinning, the cost can be further reduced, such as the related process of NORD shares has achieved a 35% cost reduction. Aluminum foil manufacturing is to cut the blank after rough rolling, finishing rolling, heat treatment, and surface treatment, which is divided into 1 series, 3 series, and 8 series according to impurities, and the overall process is mature and the cost is reasonable.

The emerging composite current collector adopts a “metal layer-polymer layer-metal layer” structure, and the coating process is the key, and different processes vary significantly in cost and performance. Magnetron sputtering has strong adhesion to the substrate, but the growth is slow, and the equipment investment is large. The evaporation plating efficiency is high and the coating layer is uniform, but the weak adhesion affects the application cost. The growth rate of water plating is fast, but the treatment cost is increased due to problems such as uneven current; Electroless plating coatings are of good quality, but deposition is slow, and catalysts are expensive. In the preparation process of composite copper foil, the one-step process (wet process) requires the use of precious metal catalysts, and the equipment cost is high; One-step (dry-process) equipment is expensive and inefficient; The efficiency of the two-step method has been improved, but the yield rate is low. The three-step method is the most efficient and can be complex. At present, the cost of 6.5 μm composite copper foil is basically the same as that of traditional ones, but with technological advancement and large-scale production, the cost is expected to decrease, such as 3D current collectors, the cost is expected to decrease significantly between 2024 and 2027.

Current collector development for anode-free batteries is advancing through surface functionalization, three-dimensional architectures, and composite coatings, collectively engineered to regulate interfacial lithiophilicity and suppress lithium dendrites. Simultaneously, emerging metallic materials—particularly metallic glasses, gradient lithiophilic alloys, and carbon-based composites—are gaining prominence as key research frontiers focused on enhancing stability and implementing lightweight corrosion-resistant systems. Despite these material innovations, economic viability presents a nuanced challenge: while anode

elimination reduces raw material costs, sophisticated fabrication processes for functionalized current collectors substantially increase production expenses, with 3D structures incurring 30%–50% higher costs than conventional copper foils. Nevertheless, synergistic optimization of current collectors and electrolytes has enabled significant full-cell performance breakthroughs, achieving over 1,000 cycles at high current densities in prototype systems. These coordinated advancements now propel the technology beyond laboratory validation toward practical engineering-scale implementation.

The artificial modification of a copper foil via surface alloying, composite coating, and related methods enables precise interfacial control to inhibit dendrite growth and stabilize SEI. While some methods have achieved scalable fabrication, the long-term durability of modified layers remains suboptimal. Meanwhile, three-dimensional (3D) copper-based current collectors, with their porous structures, provide abundant lithium deposition sites and buffer volumetric expansion, substantially enhancing cycling stability. However, their complex fabrication processes, high costs, and challenges in achieving uniform lithium deposition across 3D frameworks hinder widespread adoption. Additionally, alternative current collectors (such as carbon- or polymer-based materials) have a lightweight nature and are used in flexible applications but face persistent limitations in interfacial conductivity, electrolyte compatibility, and mechanical robustness. Although these three approaches prioritize uniform lithium deposition and interfacial stability through distinct mechanisms, the future integration of their complementary advantages may accelerate the commercialization of AFLMBs (Figure 1).

2. Artificial Modification of Copper-Foil Current Collectors

The artificial modification of copper-foil current collectors allows the precise regulation of lithiophilicity at the interface between lithium metal and current collectors, nucleation overpotential, and the structure of the SEI membrane. This is achieved through strategies such as surface alloying (e.g., with Sn, Zn, Ag), crystal plane engineering, and organic-inorganic composite coating. These approaches inhibit the growth of lithium dendrite at the source and improve the uniformity of lithium deposition.

Dong et al. rapidly constructed a Sn layer on the copper foil surface via a chemical *in situ* electroless Sn modification method (Figure 2a).^[25] During battery activation, Sn reacted with Li to form a dense Li_xSn_2 alloy layer atop the modified current collector surface. By lowering lithium nucleation overpotential and providing uniform adsorption sites, this layer suppressed dendrite growth and mitigated disordered SEI thickening. Calculations showed that energy required for Li atom adsorption on this alloyed surface was substantially lower than that for adsorption on bare copper, thereby guiding the directional deposition of lithium ions along specific crystal planes. This alloyed interface enabled symmetric cells to cycle stably for 800 h at 1 mA cm^{-2} , with CE increasing to 98.1%. When paired with cathodes such as LiFePO_4 (LFP), $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$ (NCM811), and LiCoO_2 , the full-cell lifespan was extended by 1.8 times. This approach enabled

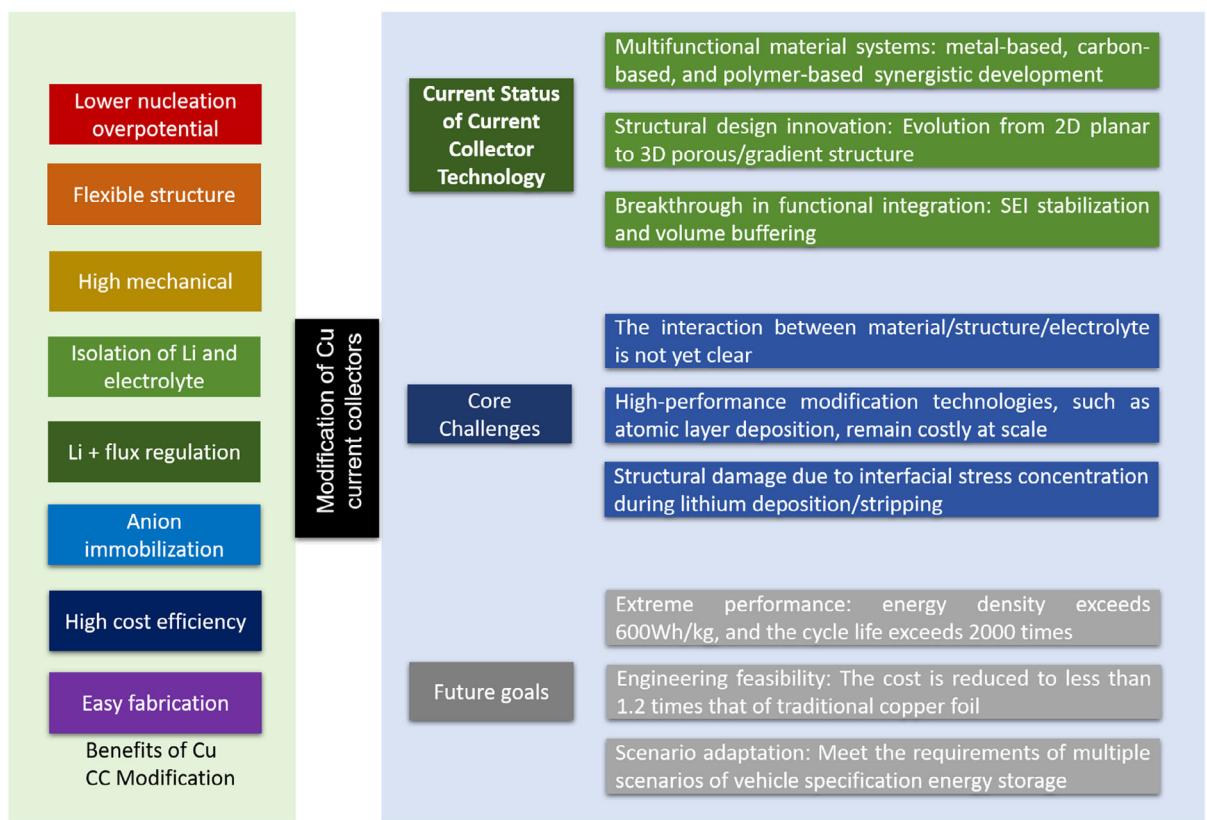


Figure 1. Breakdown of Cu current collector development: from modification advantages to current technical status, key challenges, and future performance and application goals.

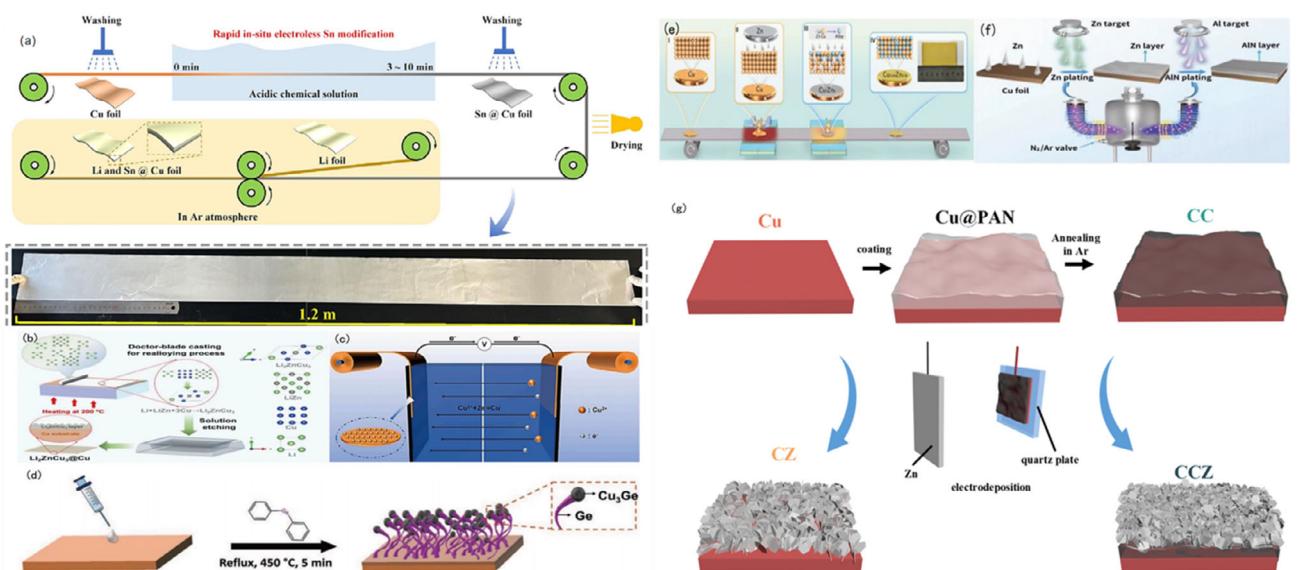


Figure 2. a) Schematic of the rapid in situ electroless Sn modification process and its integration into the continuous production of lithium metal anodes; Reproduced with permission.^[25] Copyright 2023, Elsevier. b) Schematic of the preparation routine for the $\text{Li}_2\text{ZnCu}_3@\text{Cu}$ electrode; Reproduced with permission.^[26] Copyright 2024, Wiley. c) Schematic of the fabrication process for MCP@Cu current collectors; Reproduced with permission.^[27] Copyright 2025, Elsevier. d) Synthesis of Ge nanowires (NWs) on copper foil; Reproduced with permission.^[28] Copyright 2023, Wiley. e) Schematic of the large-scale preparation of $\text{Cu}@\text{Cu}_{0.64}\text{Zn}_{0.36}$ using a two-step UHT method; Reproduced with permission.^[29] Copyright 2025, Royal Society of Chemistry. f) Schematic of the synthesis process for Zn-AlN@Cu; Reproduced with permission.^[30] Copyright 2024, Royal Society of Chemistry. g) Overview of preparation processes for various modified current collectors; Reproduced with permission.^[33] Copyright 2025, Elsevier.

the large-scale fabrication of ultra-thin films over 1200 cm^2 via a simple immersion process, offering a practical pathway for implementing AFLMBs. Similarly, Cao et al. fabricated a Li_2ZnCu_3 ternary alloy nanoarray on a copper foil using an *in situ* surface alloying technique (Figure 2b).^[26] By leveraging high lithiophilicity and increasing the specific surface area, they reduced lithium nucleation overpotential (106 mV) and inhibited dendrite growth. The resulting $\text{Li}||\text{Li}_2\text{ZnCu}_3@\text{Cu}$ asymmetric cells exhibited a CE of 99.2% at $3\text{ mA cm}^{-2}/6\text{ mAh cm}^{-2}$ and maintained stable cycling over 200 times. In addition to enhancing lithiophilicity by forming alloys with lithium or other metals, another strategy is to construct secondary structures on the surface of copper foil to address lithium deposition issues. Li et al. developed a novel electrochemical deposition strategy to construct a uniform micron-sized copper particle layer (MCP@Cu) on the copper foil surface (Figure 2c),^[27] successfully achieving homogeneous lithium metal deposition and dendrite growth suppression. This strategy used the copper foil as the Cu^{2+} source and enabled the *in situ* growth of a dense 3D structure at a current density of 3 mA cm^{-2} . Theoretical calculations and experimental results indicated that, due to the combined effects of a high specific surface area (roughness of 876 nm) and low contact angle (20°) (Figure 2d),^[28] This approach reduced lithium nucleation overpotential, enhanced CE, and exhibited good cycling stability and rate performance in full-cell tests. Alloying with lithium or other metals to enhance lithiophilicity remains a common strategy. Wang et al. constructed a uniform $\text{Cu}_{0.64}\text{Zn}_{0.36}$ alloy "skin" layer on the copper foil surface via ultra-high-temperature (UHT) Joule heating technology (Figure 2e),^[29] achieving uniform lithium metal deposition and dendritic growth suppression. This alloy layer was fabricated by initially preparing a Cu_5Zn_8 precursor at 750 K, followed by rapid alloying at a higher temperature to generate a stable $\text{Cu}_{0.64}\text{Zn}_{0.36}$ phase. Experimental and theoretical analyses showed that this alloy layer had low surface energy (0.109 eV) and high Li adsorption energy (-2.7 eV), reducing the polarization voltage of symmetric cells to 14.6 mV under $0.5\text{ mA cm}^{-2}/1\text{ mAh cm}^{-2}$ and enabling stable cycling for 1200 h. When matched with the LFP and NCM90 cathodes, the full cell sustained 320 cycles at 1C with a capacity retention of 95%. This method enabled the large-scale preparation of ultra-thin films over 1200 cm^2 by precisely controlling the alloy composition and structure, offering a new strategy for the practical deployment of high-energy-density LMBs. Zhu et al. constructed a Zn-AlN bifunctional layer on the surface of a commercial copper current collector using a Co-FCVA device (Figure 2f),^[30] further, they characterized the current collector and evaluated electrochemical performance. They found that this current collector effectively regulated lithium deposition and stripping behavior, enhancing battery performance and offering a new solution for the practical application of AFLMBs.

Zheng et al. developed a lithium graphene current collector (LC-HrGO) to form a LiC_6 phase in highly reduced graphene (HrGO) through metallurgical techniques, providing a uniform nucleation site and good conductivity for lithium metal deposition, reducing the nucleation barrier.^[31] Anode-free batteries with this current collector have a capacity retention rate of 60% after 100 cycles; The lithium-filled anode battery has a stable capacity

of 134.5 mAh g^{-1} after 250 cycles, and still has a capacity of 130.1 mAh g^{-1} at 5 C rates. Zhan et al. developed an autoselective copper current collector that catalyzes the formation of inorganic interfaces (CuCC-SSIC),^[32] which enables the self-selective growth of copper during electrodeposition by adding LiFSI to the copper sulfate electrolyte, which can promote the adsorption and decomposition of lithium salts to form inorganic-rich SEI. The CE of lithium plating/stripping with this current collector was increased from 99.25% to 99.50% (stable for 400 cycles), the capacity attenuation rate of the anode less battery was reduced by 42.4% for 100 cycles, and the pouch cell preparation of 350 Wh kg⁻¹ was realized.

In the research of LMBs, introducing additional modified structures or functional coatings on the surface of copper foil to provide rich active sites for lithium deposition and regulate the deposition microenvironment has become a key strategy for optimizing the behavior of lithium deposition. Kenan Zhong et al. coated cyclised polyacrylonitrile (CPAN) on a commercial copper foil for the first time, preparing a $\text{Cu}@\text{CPAN}@\text{Zn}$ (CCZ) composite current collector for lithium metal anodes (Figure 2g).^[33] They investigated phase transformations occurring during fabrication and their influence on zinc and lithium deposition, revealing that CCZ suppressed lithium dendrite growth and improved battery electrochemical performance. Similarly, Liu et al. fabricated a metal-organic framework (MOF)-CuO@carbon-modified copper foil via a two-step calcination method.^[34] Characterization and electrochemical performance testing demonstrated that this material decreased lithium nucleation overpotential and inhibited dendrite growth. In addition to providing rich active sites for lithium deposition by adding additional modification structures or coatings on copper foil, methods for modifying current collectors have also been reported. Ashok Kushwaha et al. uniformly coated a graphene layer on an aluminum current collector (AlCC) using inkjet printing technology.^[35] This coating effectively suppressed the corrosion of the AlCC under high-voltage conditions, thereby enhancing cycling stability and rate performance in LIBs with an Li-NMC cathode. This work presented a new approach to advance high-voltage alkali metal ion batteries. Hao et al. designed a (100)-textured copper-foil current collector through a crystal plane engineering strategy and achieved directional lithium deposition along the (110) crystal plane by leveraging a low lattice mismatch ($\delta = 0.8\%$).^[36] This study enabled the controllable preparation of a centimeter-scale single-crystal copper foil via electrochemical etching, with a surface roughness of $\text{Ra} < 5\text{ nm}$, offering a new paradigm for interfacial regulation in high-energy-density LMBs. Jin et al. prepared fluorinated, gold-doped titanosilicate to modify copper current collectors for stabilizing lithium metal anodes.^[37] Benefiting from the lithiophilicity of gold nanoparticles, microporous nature of the titanosilicate framework, and promotion of LiF-rich SEI formation owing to fluorine doping, this modification enhanced battery performance. The $\text{Li}||\text{Cu}$ cell achieved a CE of 96.8% after 600 cycles. Zhaofeng Ouyang et al. developed an ultra-light composite current collector (PASC-Cu) for AFLMBs.^[38] Through rational molecular engineering, they strengthened interfacial adhesion between the polymer substrate and copper layer, improving energy density,

rate capability, flexibility, and safety. This work provided a new benchmark for developing high-performance anode-free batteries. The role of fluorine elements is crucial in the evolution process of the SEI, as they promote the formation of high-resistance LiF. Meanwhile, the properties of current collectors can also affect the SEI. Tan et al. pointed out that the surface chemical properties and morphological structure of current collectors influence the distribution and content of fluorides in the SEI, thereby altering the ionic transport performance and stability of the SEI. Ma et al.'s research showed that the interaction between fluorine sources (such as fluorinated additives in electrolytes) and current collectors regulates the nucleation and growth process of LiF. If lithiumophilic sites exist on the surface of current collectors, they can promote the uniform distribution of LiF and reduce the interfacial resistance.^[39] Menkin et al., through in situ characterization, found that in anode-free batteries, the oxidation state of copper current collectors affects the crystallinity of LiF in the SEI. The interface of high-valent copper oxides is more likely to induce LiF to form a dense layer, inhibiting the growth of lithium dendrites.^[40] However, an excessively high LiF content may also lead to battery performance degradation due to increased resistance. Overall, there is a synergistic effect between current collectors and fluorine elements in SEI evolution. Optimizing the surface characteristics of current collectors to regulate the generation behavior of LiF is a key direction for improving battery interface stability.

Bo Rui et al. proposed a pre-strain strategy for current collectors to mitigate electrode delamination in LIBs.^[41] They developed an analytical model specifically designed for symmetric electrodes incorporating deformable and limited-thickness current collectors. This model facilitated a comprehensive analysis of the delamination behavior, accounting for pre-strain in the current collector and intrinsic strain within the active layer. This idea complements the thermal oxidation composite coating method, Darwaish et al. fabricated an in situ oxide layer and artificial protective coating on the copper foil surface.^[42] Initially, CuO/Cu₂O nanoparticles were generated via thermal oxidation at 320 °C, forming an Li₂O-rich SEI layer (ISEI) that reduced lithium nucleation overpotential from 137 to 33 mV. Subsequently, a GO@PVDF-HFP@Li-Nf composite layer (ASEI) was applied. The two-dimensional conductive network of graphene oxide (GO), combined with the ionic conductivity of Li-Nf, suppressed volume expansion. The resulting GO@Cu-30 current collector enabled dense lithium deposition in anode-free full cells, with an interfacial impedance of only 26.7 Ω—93% lower than that of an unmodified copper foil. After 250 cycles at 0.5C, the battery exhibited a capacity retention rate of 93.12% and stable CE of 99.92%, notably better than the 9.54% capacity retention rate of the bare copper foil. X-ray photoelectron spectroscopy analysis revealed an Li₂O content of 57.5% in the SEI layer, and scanning tunneling microscopy images confirmed a uniformly deposited lithium layer. These findings indicated that the composite structure effectively suppressed interfacial side reactions and dendrite formation. This method realized the functional modification of current collectors through simple thermal oxidation and a coating process, offering a new strategy for high-energy-density LMBs. To prevent coating peeling, Wu et al. directly fabricated

an adhesive-free HKUST-1 polycrystalline MOF^[43] membrane on the copper foil surface through in situ epitaxial growth. This study marked the first introduction of MOF polycrystalline membrane technology to the domain of AFLMBs. Notably, it resolved the issue of active site shielding caused by the adhesive dependence of conventional MOF-based modifications. The membrane effectively inhibited dendrite growth and represented a significant technological breakthrough in enhancing battery safety and cycle life, thereby promoting the practical application of high-energy-density LMBs. Although the aforementioned strategies have demonstrated promising short-term performance enhancements, modified layers still face several problems during prolonged lithium stripping and deposition cycles. When discussing the presence of lithium carbides that cause dendrites and short circuits during lithium plating, two key studies shed light on the issue. Schmitz et al. revealed that battery-grade lithium metal often contains lithium carbide Li₂C₂ as a contaminant, detected via Raman spectroscopy and mass spectrometry both on the surface and in the bulk of lithium samples. The study shows that Li₂C₂ originates from carbon contamination during fused salt electrolysis or high-temperature purification, and its presence disrupts the formation of a uniform SEI, leading to dendritic growth. Golozar et al. further demonstrated via in situ scanning electron microscopy that carbide-based dendrites exhibit a hollow morphology and hardness greater than pure lithium, enabling them to penetrate polymer electrolytes and cause short circuits. Mass spectrometry confirms that hydrolysis of lithium metal with Li₂C₂ produces acetylene, indicating active carbide participation in interface destabilization. Collectively, these studies highlight that lithium carbides, as underestimated contaminants, compromise SEI stability and promote mechanically robust dendrites, underscoring the necessity of strict carbon contamination control in lithium production.

These problems include the repeated rupture of the SEI membrane, sharp increases in interfacial impedance, and delamination or peeling of the coating. For instance, certain alloy layers may result in uneven lithium deposition at high current densities, posing safety risks due to dendrite penetration through the separator. Crystal plane engineering requires the extremely precise control of surface roughness and lattice matching, and the stability of such interfaces under complex operating conditions (e.g., fast charging and high temperatures) remains unverified. While organic-inorganic composite coatings can improve short-term performance, organic components may undergo side reactions with the electrolyte, thereby compromising long-term cycling. Additionally, coatings reliant on adhesives risk covering active sites and increasing interfacial resistance. These drawbacks constrain the broad applicability of such strategies under long-duration cycling scenarios. Therefore, future efforts must prioritize the optimization of modified layer durability and interfacial adaptability.

Metal-based substrates remain the industrial mainstay due to their unmatched electrical conductivity and mechanical resilience, particularly suited for high-energy-density cells requiring robust current pathways. Carbon-based alternatives (graphene-enhanced meshes, carbon nanotube papers) emerge as transformative

solutions where weight reduction and electrochemical stability are prioritized, though their higher manufacturing costs currently constrain widespread adoption. Polymer-based systems (conductive polymer-coated polyimide/PET films) enable unprecedented form-factor freedom for flexible electronics, trading absolute conductivity for elasticity and corrosion resistance. Crucially, these material families exhibit synergistic relationships: metal–carbon hybrids enhance high-rate performance in aerospace applications, while metal–polymer composites serve emerging wearable technologies. As the field progresses, metal substrates continue to anchor commercial manufacturing, carbon architectures drive energy-critical innovations, and polymer systems unlock new form factors—collectively advancing current collector technology through targeted material specialization.

3. Three-Dimensional Copper-Based Collectors

Compared with two-dimensional surface modifications, 3D copper-based collectors establish a dual “structure + chemistry” regulatory framework through the synergistic design of a porous scaffold and lithiophilic interface. Abundant nucleation sites afforded by the 3D architecture enable the uniform distribution of deposition current and inhibit the growth of dendrite tips. Interconnected pores provide space to effectively buffer the volume expansion of lithium metal, with the relief rate exceeding 60%, thereby significantly enhancing the cycle life of symmetric cells.

Fan et al. addressed the issues of dendrite growth and cycling stability of lithium metal anodes, fabricating an innovative host structure. An indium layer was electroplated onto a 3D nickel foam current collector, followed by in situ alloying to form a lithiophilic $\text{In}_3\text{Li}_{13}$ interfacial layer (Figure 3a).^[44] This lithiophilic layer

offered numerous nucleation sites. Experimental data and density functional theory calculations confirmed that lithium ions preferentially deposited on the $\text{In}_3\text{Li}_{13}$ surface rather than on lithium nuclei or the nickel substrate. This mechanism facilitated dendrite-free, uniform, granular lithium deposition. The design markedly reduced lithium nucleation overpotential, improved CE (reaching 97.4% after 300 cycles at 2 mA cm^{-2}), and enhanced cycling stability. The symmetric cell exhibited low voltage hysteresis and long lifespan. The assembled $\text{Li}-\text{In}_3\text{Li}_{13}@\text{NF}/\text{LiFePO}_4$ full cell retained 82.8% of its capacity after 1000 cycles at 2C, demonstrating excellent practical potential. This study elucidates the underlying growth mechanism of lithium dendrites. This approach relies on *in situ* alloying to create lithiophilic metallic interfaces, another strategy focuses on the design of carbon-based nanostructures. Guo et al. fabricated a lightweight, 3D lithiophilic graphene aerogel current collector comprising reduced GO aerogel and silver nanowires via chemical reduction and freeze drying (Figure 3b).^[45] This structure provided uniform lithium nucleation sites and effectively suppressed dendritic growth. Compared to metal-based systems, some studies have explored organic–inorganic hybrid frameworks to address the issue of volume expansion during lithium deposition. Jia et al. synthesized a self-supporting 3D current collector (3D-1H₃N) by preparing a lithiophilic $[\text{Cu}(\text{NH}_3)_2]\text{Cl}$ compound *in situ* on the surface of a porous CuZn scaffold via a one-step method.^[46] The 3D framework accommodated volume expansion associated with lithium deposition, while the $[\text{Cu}(\text{NH}_3)_2]\text{Cl}$ phase guided the uniform nucleation and deposition of lithium within internal pores. This design effectively reduced lithium nucleation overpotential and inhibited dendrite growth. The prepared half-cell displayed a CE of 98% and maintained stable cycling for over 270 cycles at 0.5 mA cm^{-2} , while the symmetric cell operated stably for 1100 h at 0.5C. These results suggested excellent electrochemical

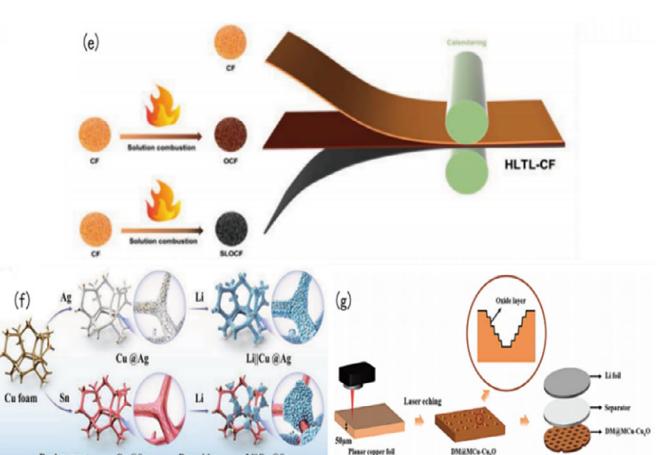
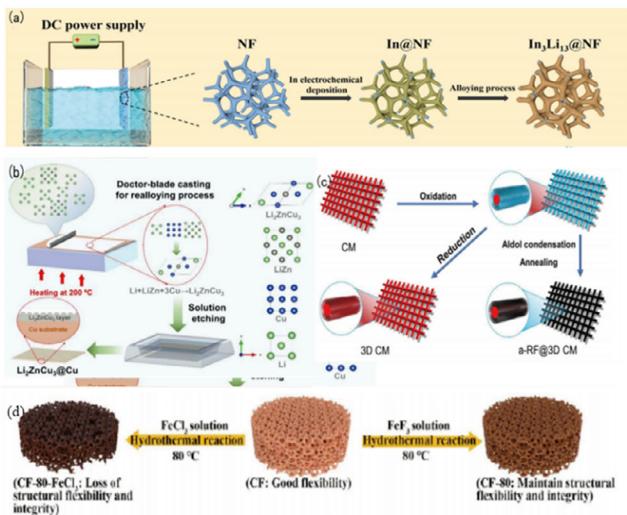


Figure 3. a) Schematic of the synthesis protocol for $\text{In}_3\text{Li}_{13}@\text{NF}$; Reproduced with permission.^[44] Copyright 2023, Elsevier. b) Schematic of the preparation of reduced GO aerogel with silver nanowires; Reproduced with permission.^[45] Copyright 2024, Multidisciplinary Digital Publishing Institute. c) Demonstration of the synthesis of 3D CM and a-RF@3D CM; Reproduced with permission.^[48] Copyright 2023, Elsevier. d) Two synthesis pathways for the Cu_2O -modified layer on CF; Reproduced with permission.^[49] Copyright 2023, Royal Society of Chemistry. e) Fabrication of hierarchically lithiophilic triple-layered CFs (HLTL-CFs); Reproduced with permission.^[52] Copyright 2024, Elsevier. f) Schematic of Cu foam modified with gold nanoparticles and a Cu–Sn alloy layer; Reproduced with permission.^[54] Copyright 2024, Elsevier. g) Formation process of MCu– Cu_xO current collector; Reproduced with permission.^[55] Copyright 2024, Elsevier.

performance and cycling stability, offering a novel perspective on current collector design for high-performance LMBs. You et al. developed a Cu₄₀@Cu₃P current collector to construct a 3D porous structure by microwave-induced oxidative etching,^[47] and then vapor phase phosphating to form a lithium-philic Cu₃P coating to regulate lithium deposition. The current collector improves the cycling stability of the anode battery at a high areal capacity of 3.4 mAh cm⁻², performs well for 100 cycles, and due to its ultra-thin (3.4 μm) and ultra-light (4.27 mg cm⁻²) characteristics, the gravimetric energy density and volumetric energy density of the battery reach 215 Wh kg⁻¹ and 540 Wh L⁻¹, respectively.

Li et al. constructed an ultra-three-dimensional lithiophilic modified layer (a-RF@3D CM), incorporating dispersed CuOx sites and oxygen-deficient carbon, via pyrolyzing terephthalaldehyde resin on the surface of a 3D copper mesh (Figure 3c).^[48] This copper-based current collector reduced lithium nucleation overpotential and suppressed dendrite growth, enabling the half-cell to cycle over 400 times with CE exceeding 98.5%. The full cell exhibited excellent cycling stability under anode-free and lithium-deficient conditions. Simpler approaches such as direct metal oxide coating have also demonstrated strong lithiophilicity. Liu et al. successfully coated a dense layer of cuprous oxide (Cu₂O) particles on 3D copper foam (CF) using a straightforward hydrothermal method, thus fabricating a lithiophilic composite current collector (CF-80) (Figure 3d).^[49] Owing to the strong lithiophilicity of Cu₂O, lithium nucleation overpotential was reduced to just 15 mV. The 3D structure further facilitated the uniform dispersion of lithium-ion deposition, substantially enhancing cycling stability. Electrochemical testing showed that, under a current density of 1 mA cm⁻² and capacity of 1 mAh cm⁻², even when capacity was doubled after 120 stable cycles, efficiency remained at 97.6%. In symmetric cells, low voltage hysteresis and extremely long cycle life were observed. The full cell (NCM-811//CF-80/Li) maintained 85.3% capacity retention after 150 cycles. Liu et al. developed a 3D, hierarchical, porous nickel foam current collector (NP-NF) via electrodeposition and de-alloying.^[50] The synergistic effect of its lithiophilic NiO surface and porous architecture facilitated uniform lithium deposition. The half-cell displayed a CE of 95% after 350 cycles. The symmetric cell exhibited low overpotential after 1000 cycles, and the full cell retained >80% of its capacity after 300 cycles at 1C, significantly enhancing the stability of the lithium metal anode. Ana C. Martinez et al. employed digital light processing (DLP) 3D printing to fabricate a copper current collector, which was then coated with electrode materials such as graphite via electrophoretic deposition.^[51] This design successfully integrated structural support and energy storage functions. Testing confirmed the application potential of this 3D-printed copper current collector in LIBs.

Peng et al. developed innovative roll-to-roll manufacturing technology to carefully fabricate CF with different lithiophilicities via a solution combustion method, followed by the formation of a unique three-level lithiophilic layer foam structure (HLTL-CF) through mechanical rolling (Figure 3e).^[52] A lithium composite electrode was subsequently constructed through lithium melt infiltration. This exquisite structure featured a bottom ultra-lithiophilic layer that effectively directed uniform lithium-ion deposition from bottom to top, maintaining internal structural

integrity; a middle lithiophilic layer actively promoted rapid ion transport to enhance charge-discharge efficiency; and a top lithiophobic copper foil was crucial in buffering volume expansion, effectively prolonging battery life. COMSOL simulations comprehensively validated the superiority of this controlled deposition mechanism. The symmetric Li@HLTL-CF cell exhibited remarkable cycling stability, maintaining over 1350 cycles at a current density as high as 20 mA cm⁻² and a practical capacity of 5 mAh cm⁻². When coupled with a high-loading LFP cathode in a full cell, a capacity retention of 86.9% was maintained even after 400 cycles at 1C. This approach significantly suppressed dendrite growth and greatly enhanced long-term cycling stability. This technological breakthrough offered an efficient and feasible solution for the large-scale production of good-performance lithium batteries, with a substantial practical value and broad industrial prospects. Parallel to multilayer structural design, Su et al. prepared a lithiophilic Cu_{1.81}S-doped carbon layer on the surface of 3D CF via chemical vapor deposition, thereby constructing a high-performance current collector.^[53] The symmetric cell incorporating this collector achieved stable cycling over 6000 h, while the full cell retained 96.4% of its capacity after 120 cycles. This structure effectively suppressed lithium dendrite growth and enhanced battery stability.

Yang et al. modified the surface of 3D CF with a lithiophilic silver/tin coating via a displacement reaction (Figure 3f), thereby fabricating a gradient current collector. The half-cell demonstrated a CE of 97.35% after 550 cycles, effectively guiding uniform lithium deposition, inhibiting dendrite growth, and improving cell stability.^[54] In contrast to surface coating methodologies, Zhang et al. constructed a multilayer microstructure on the copper foil surface using laser etching and formed a lithiophilic surface layer *in situ* (Figure 3g).^[55] This innovative dual-modification approach for the current collector substantially improved cell performance. Testing showed that the half-cell maintained a CE of >98% after over 240 cycles. Alongside microscale topographical control, Zhong et al. developed a dendritic structure on the CF surface through electrodeposition, thereby producing a dendritic CF current collector (DCFC).^[56] The geometrical confinement effect of this collector enabled uniform lithium deposition, considerably inhibiting dendrite formation and improving overall cell performance. While 3D nanostructures remain a focal point, Zou et al. fabricated a perforated copper-foil current collector via mechanical punching, considerably enhancing capacity and cycling stability in LIBs.^[57] They elucidated the structural optimization mechanism by which perforation improved cell performance. Despite the advantages conferred by 3D structures, these designs introduce new challenges in practical application. Complex fabrication processes result in a 30%–50% increase in cost compared to traditional copper foils. Furthermore, large-scale production faces difficulties in precisely controlling porosity and ensuring the uniform distribution of lithiophilic sites, which can lead to localized “hot spot” deposition. Additionally, an increased thickness of 3D current collectors (50–100 μm compared to 10–15 μm for traditional copper foils) compromises volumetric energy density to a certain extent. Their compatibility with existing roll-to-roll processing equipment also requires re-optimization. These constraints

highlight the trade-off between “structural innovation” and “engineering scalability”, underscoring the need for concurrent advances in material design and scalable manufacturing processes.

4. Other Derived Current Collectors

This category of current collectors’ centers on material innovation overcomes the limitations of conventional metallic substrates. Metal-based substrates remain the industrial mainstay due to their unmatched electrical conductivity and mechanical resilience, particularly suited for high-energy-density cells requiring robust current pathways. Carbon-based alternatives (graphene-enhanced meshes, carbon nanotube papers) emerge as transformative solutions where weight reduction and electrochemical stability are prioritized, though their higher manufacturing costs currently constrain widespread adoption.^[58] Polymer-based systems (conductive polymer-coated polyimide/PET films) enable unprecedented form-factor freedom for flexible electronics, trading absolute conductivity for elasticity and corrosion resistance. Crucially, these material families exhibit synergistic relationships: metal–carbon hybrids enhance high-rate performance in aerospace applications, while metal–polymer composites serve emerging wearable technologies. As the field progresses, metal substrates continue to anchor commercial manufacturing, carbon architectures drive energy-critical innovations, and polymer systems unlock new form factors—collectively advancing current collector technology through targeted material specialization.

Lightweight materials greatly reduce the inert mass of a cell, thereby enhancing energy density. Flexible substrates, such as carbon nanotube sheets (CNTS) and polymer films, are suitable for emerging applications including those in wearable devices and flexible electronics. Non-copper-based materials, including metallic glasses and ceramic composites, offer unique physical and chemical properties that enhance interfacial lithophilicity, inhibit galvanic corrosion, and improve mechanical stability. Some structures are also engineered to optimize ion- and electron-transport efficiencies via the use of porous architectures or conductive networks. Others employ surface functionalisation to synergise materials with lithium metal, providing a wide range of solutions for cells.

Chen et al. fabricated a lightweight, copper-coated graphene film current collector via electroplating (Figure 4a).^[59] With a surface density of 3.44 mg cm^{-2} and an electrical conductivity of $3.25 \pm 0.07 \times 10^7 \text{ S m}^{-1}$, this collector increased the gravimetric specific capacity of LIBs by 246.01% at 1C. It also demonstrated excellent rate capability and cycling stability. In addition to surface coating technologies, the adoption of 3D porous structures has been further combined with hierarchical design to enhance capacity retention performance. Huang et al. synthesized CuO microspheres on the surface of a conductive carbon fiber/Cu core–shell fiber structure to form a 3D porous composite current collector (Figure 4b).^[60] This configuration yielded a reversible capacity of 415 mAh g^{-1} after 50 cycles at 0.1C—notably outperforming 127 mAh g^{-1} achieved by a conventional copper foil. The enhancement in conductivity, capacity, and cycling stability underscored the collector’s effectiveness. Transitioning from rigid

3D composites to flexible CNT collectors via direct spinning, the focus shifts to mechanical robustness for bendable LIBs. Semin Jo et al. developed CNTS using direct spinning technology and applied them as flexible, ultra-light, and stackable 3D current collectors in LIBs.^[61] These batteries demonstrated excellent performance, and pouch cells incorporating CNTS continued to operate reliably under mechanical deformation. Seungho Lee et al. further modified CNT-based current collectors through oxidation and reduction treatments to enhance surface wettability with electrolytes and improve conductivity.^[62] To address galvanic corrosion arising from contact between traditional copper-foil current collectors and lithium metal, Li et al. proposed the use of an artificial graphite paper (GP) as an anticorrosion current collector. Owing to its low weight and excellent mechanical properties, GP effectively suppressed interfacial corrosion and maintained stable contact between lithium metal and the collector throughout cycling. Experimental results showed that batteries employing Li@GP electrodes exhibited superior electrochemical performance in the asymmetric and full-cell configurations. Notably, a 1.08-Ah NCM811||Li@GP pouch cell retained 91.6% of its capacity after 240 cycles under a low anode-to-cathode ratio ($N/p = 2$) and lean electrolyte conditions (3 g Ah^{-1}), demonstrating excellent practical potential. This study offered a simple and effective route for improving the cycling stability and energy density of LMBs, while advancing the application of carbon-based current collectors in high-energy systems.

Liu et al. fabricated a mesoporous current collector with a lithophilic copper oxide-coated surface via de-alloying copper-zirconium metallic glass (Figure 4c).^[63] This structure demonstrated the potential of metallic glasses as current collector materials in lithium batteries. To address the limited cycle life of anode-free batteries, Liu et al. proposed a surface-functionalized alloy foil current collector (Figure 4d).^[64] Through a simple wet contact reaction, the alloy foil reacted with a lithium naphthalene solution to pre-store a controllable quantity of active lithium. The resulting lithium alloy layer on the current collector surface reduced lithium nucleation overpotential, ensured uniform deposition, and continuously released active lithium during cycling to compensate for irreversible lithium loss. This strategy extended the cycle life of anode-free batteries by approximately tenfold. It was applicable to various metals such as indium, tin, and gallium, providing an effective strategy for solving lithium loss in high-energy-density systems. Surface-functionalized alloy foils focus on lithium storage via chemical reactions, whereas silver–carbon nanocomposites reduce interfacial impedance through metal–carbon synergy. Liu et al. developed a silver–carbon nanocomposite current collector, integrating silver nanoparticles with carbon materials.^[65] This configuration substantially reduced interfacial impedance and promoted uniform lithium metal deposition. The resulting battery exhibited stable cycling for over 200 cycles at a current density of 0.25 mA cm^{-2} , providing an effective strategy for improving the performance of anode-free all-solid-state lithium batteries (AFSSLBs). Qi et al. synthesized a lithophilic Ni_3S_2 nanowire array *in situ* on a porous nickel current collector via a hydrothermal method, yielding a $\text{Ni}_3\text{S}_2@\text{Ni}$ composite structure.^[66] This current collector considerably improved the lithium deposition behavior, enabling symmetric cells to cycle stably.

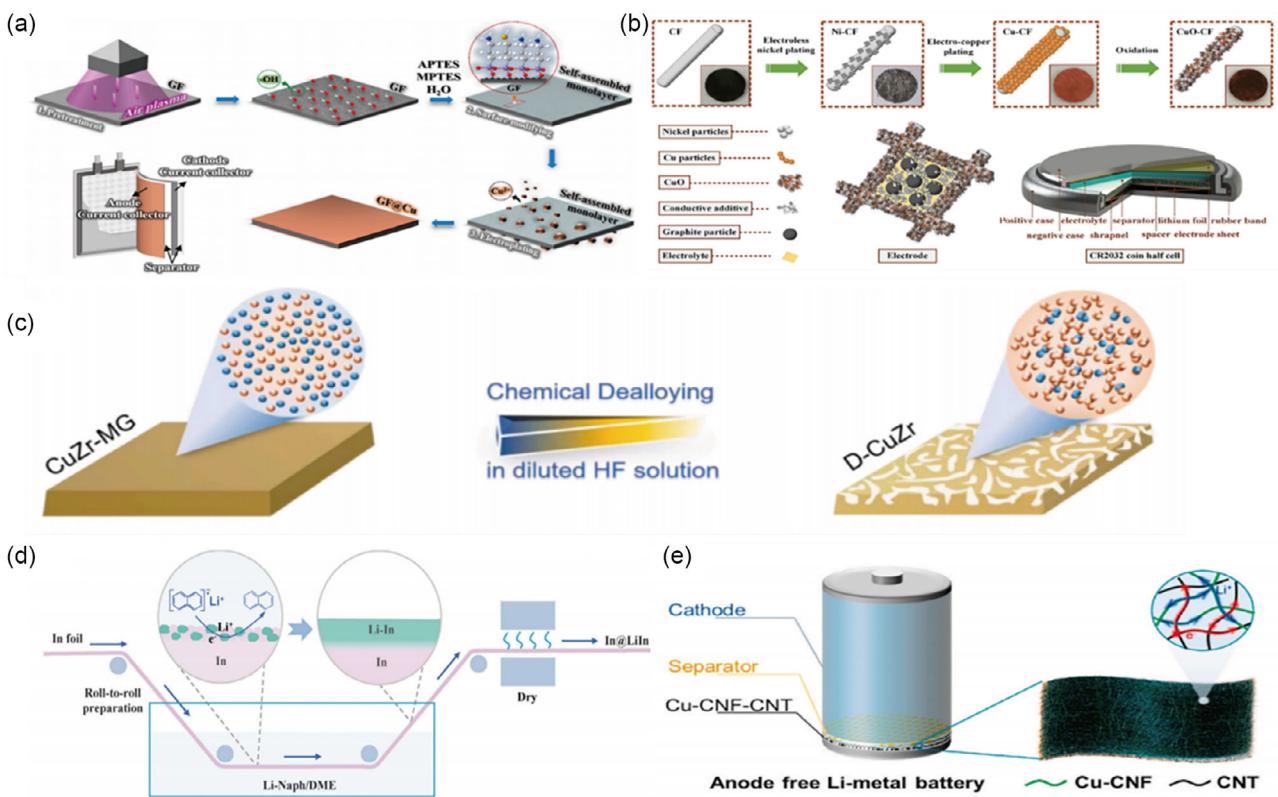


Figure 4. a) Schematic of the fabrication process for GF@Cu film; Reproduced with permission.^[59] Copyright 2023, Elsevier. b) Schematic (color online) of the preparation process for the 3D porous composite current collector; Reproduced with permission.^[60] Copyright 2018, Springer. c) Diagram of the chemical de-alloying of CuZr metallic glass (CuZr-MG); Reproduced with permission.^[63] Copyright 2023, Elsevier. d) Preparation process of surface-functionalized metal foils, where the foil is unrolled at a controlled speed into a reaction tank containing a strongly reducing lithium source; after alloying, the functionalized current collector is obtained via drying; Reproduced with permission.^[64] Copyright 2025, Wiley. e) Schematic of an AFLMB using the Cu-CNF-CNT all-fiber current collector, in which CNTs provide the electron conduction pathway and Cu-CNF fibers guide lithium-ion transport; Reproduced with permission.^[69] Copyright 2024, Elsevier.

for 900 h at 2 mA cm⁻². The corresponding full cell achieved an 80.6% capacity retention after 500 cycles, highlighting its effectiveness in stabilizing lithium metal anodes.

Mintao Wan et al. introduced a metal-free, polymer-based current collector (PE/C) comprising polyethylene and carbon black.^[67] With a thickness of just 10 µm and an areal mass of 1.03 mg cm⁻², this lightweight design promoted uniform lithium metal deposition. It increased the specific energy of LMBs to 448 Wh kg⁻¹—an 18% improvement compared to traditional copper current collectors—while also remarkably extending cycle life. Zhang et al. systematically studied current collectors for high-energy LIBs, focusing on metalized plastic composite current collectors (MPCCs).^[68] They reviewed the advantages of MPCCs—such as reduced inert weight, enhanced energy density and improved safety—using a metal–polymer–metal multilayer composite structure. Furthermore, they identified key challenges such as poor interfacial adhesion and limited electrical conductivity. Their analysis encompassed suitable polymer substrates, fabrication techniques, and modification strategies, such as improving interfacial performance through surface treatments and adhesive layer addition. They further discussed methods that enhanced electrical conductivity and welding compatibility. The study provided insights into the future application of MPCCs in

flexible, highly safe, and high-energy-density batteries, offering key guidance for the design and commercialization of next-generation current collectors. Zhong et al. developed an ultra-light, dual-conductive, all-fiber-based 3D anode current collector (Cu–CNF–CNT) by integrating CNTs with copper-coordinated cellulose nanofibers (Figure 4e).^[69] This structure combined low density, high mechanical strength, and electrolyte wettability. In AFLMBs, it increased the capacity retention rate by 33%, demonstrating significant application potential under high-energy-density scenarios. Ouyang et al. fabricated a lithophilic zinc oxide structure encapsulated within a CNT array on a carbon cloth as an anode current collector via a hydrothermal method and heat treatment.^[70] The resulting inorganic lithium-rich, mixed-conductive interface promoted uniform lithium deposition, inhibited dendrite growth, and accommodated volume changes during cycling. The half-cell achieved a CE of 98.54%.

Wu et al. investigated the reduction reactivity and SEI formation mechanisms of three polymers—poly(ethylene oxide) (PEO), poly(*ɛ*-caprolactone) (PCL), and poly(trimethylene carbonate) (PTMC)—on lithium/copper (Li/Cu) surfaces using density functional theory (DFT) and ab initio molecular dynamics simulations.^[71] They analyzed the adsorption energy, electronic structure, and charge transfer processes of these polymers on Li/Cu surfaces, revealing

that the copper current collector enhances the interaction between the polymers and lithium metal, facilitating electron transfer to the polymers and accelerating their reduction reactions. The simulations demonstrated that PCL exhibits the highest adsorption energy (-4.42 eV) on the Li/Cu surface, and its C—O bond cleavage occurs faster than on pure lithium surfaces. In contrast, PTMC decomposition does not generate CO molecules, thereby improving SEI quality. Time-dependent Bader charge analysis indicated that the copper current collector alters the charge distribution in the lithium layer, enabling the polymers to acquire more electrons and consequently influencing SEI composition and stability. This study provides theoretical insights for optimizing interfacial reactions between copper current collectors and polymer electrolytes in AFLMBs. Chen et al. developed a simple and low-cost strategy to modify copper current collectors with self-assembled γ -aminopropyltrimethoxysilane (γ -APS) films.^[72] The nanoporous polymer film promotes pebble-like lithium deposition, inhibits lithium dendrite formation, improves the lithiophilic of copper current collectors, and mitigates environmental and galvanic corrosion. The half-cell using the γ -APS-Cu current collector has an average CE of up to 99.2% over 100 cycles, a symmetrical battery life extended to 1400 h, and a full cell capacity of up to 136 mAh g^{-1} after 600 cycles at 0.5C.

Pan et al. successfully prepared a copper-coated polyimide (PI@Cu) composite current collector for LIBs by a simple electroless copper plating deposition method.^[73] The preparation process includes NaOH pretreatment of the PI film, AgNO_3 activation, and impregnation in an electroless copper bath, where a continuous copper layer is formed on both sides of the PI film at an electroless plating time of 20 min, and the current collector has a surface density of only 1.99 mg cm^{-2} , which is much lower than that of commercial copper foils, and has a rough surface with good flexibility, mechanical stability, and oxidation resistance. After applying this PI@Cu composite current collector to the lithium titanate anode, compared with the electrode using commercial copper foil current collector, it not only reduces the mass proportion of inactive components and improves the weight specific capacity based on the whole electrode mass, but also increases the interface contact area and reduces the interface resistance and internal resistance due to the rough surface of the copper layer, so that the electrochemical reaction kinetics are better, the rate performance is better, and the discharge specific capacity is higher at different current densities. This study provides a simple and effective method for the fabrication of new lightweight current collectors for high-energy-density and high-power-density LIBs.

Despite these advancements, nonmetallic and novel material-based current collectors generally face interfacial compatibility challenges. Carbon-based substrates often exhibit inadequate lithiophilicity and require surface modification to prevent uneven lithium deposition. PE/C relies on conductive additives to retain electrical conductivity but is prone to interfacial delamination due to electrolyte-induced swelling. Furthermore, materials such as metallic glasses and ceramics need complex, costly fabrication processes that are difficult to scale for industrial production. Their chemical stability or mechanical strength must be optimized, and

issues such as increased interfacial impedance and structural degradation during long-term cycling remain. These issues hinder the transition of such innovations from laboratory research to large-scale commercial implementation.

5. Modified Current Collectors for AFSSLBs

AFSSLBs represent a frontier in high-energy-density storage, yet their practical advancement is hindered by critical interfacial challenges—notably poor contact between solid electrolytes and current collectors, uncontrolled lithium dendrite growth, and unstable cycling performance. Modified current collectors have emerged as a pivotal solution to these issues, leveraging tailored surface chemistries and structural designs to regulate lithium nucleation, enhance interfacial compatibility, and suppress dendrite formation. This section highlights key strategies for engineering current collectors in AFSSLBs, emphasizing innovations that bridge material science and electrochemical performance to unlock the technology's full potential. Liu et al. developed a silver-carbon nanocomposite current collector to address the problems of contact failure and dendrite growth at the interface between electrolyte and current collector in AFSSLB.^[74] They synthesized silver nanoparticles by ligand-assisted method, mixed with conductive carbon black (Super P) to prepare a composite layer, and coated on stainless steel foil to form a current collector, which was applied to a system with $\text{Li}_6\text{PS}_5\text{Cl}$ as a solid electrolyte. The composite current collector provides abundant lipophilic sites through the uniform dispersion of silver nanoparticles, promotes the uniform nucleation and deposition of lithium, and uses silver and lithium to form an alloy to accelerate the diffusion of lithium ions and reduce dendrite growth. The test shows that the interfacial impedance is as low as $10\Omega\text{ cm}^{-1}$, and it can maintain good contact during charging and discharging. Batteries with this current collector can deposit more than $25\mu\text{m}$ of lithium without short circuits, stably deposit more than 7.0 mAh cm^{-2} of lithium at a current density of 0.25 mA cm^{-2} , achieve a capacity retention rate of 54% after 200 cycles, and an average CE of over 99.7%. Gu et al. prepared stainless steel current collectors with different surface roughnesses, including bare SS (roughness 21 nm), weakly etched SS (WESS, roughness 180 nm),^[75] and strong etched SS (SESS, roughness 536 nm), by etching method, and systematically investigated the effects of these current collectors on lithium deposition/stripping behavior in anodeless all-solid-state batteries (AFASSBs). In the liquid electrolyte system, the greater the roughness of the current collector, the better the cycling stability of lithium deposition/stripping, where SESS can stabilize the cycle for 500 h; However, in solid-state electrolyte systems, the situation is different, with moderately coarse WESS performing best because it increases the contact point with the solid-state electrolyte, providing more reaction sites and favoring uniform lithium formation. The full-cell test results showed that the WESS-based battery had an initial discharge capacity of 163 mAh g^{-1} and remained at 120 mAh g^{-1} after five cycles, with a CE of 92%, which was far better than the bare SS and SESS-based batteries. Interface analysis also

showed that WESS was able to inhibit the decomposition of the solid electrolyte and promote the uniform deposition of lithium.

Wang et al. aimed at the stability of nonanode all-solid-state lithium batteries, and improved the performance by manipulating the wettability of lithium metal on the surface of copper current collectors.^[76] They vaporized commercial copper foil to form a uniform Cu₂Te layer on the surface, and then converted it into a 1 μm thick Li₂Te layer by in situ lithium activation to construct a Li₂Te -Cu current collector, which was applied to a system with argyrodite Li₆PS₅Cl as a solid electrolyte. This current collector significantly reduces the deposition/stripping overpotential of lithium and improves CE. In the half-cell test, the cumulative thickness of lithium on the Li₂Te -Cu current collector is more than 70 μm, which is comparable to the thickness of the lithium foil counter electrode, while the unmodified copper current collector can be deposited at less than 20 μm before a short circuit occurs. It has a 99.70% CE for lithium deposition/stripping, much higher than the 98.47% for unmodified copper, and is stable for more than 380 h at 0.5 mA cm⁻². The microstructure analysis showed that the lithium deposition on the Li₂Te Li₂Te -Cu current collector was uniform and dense, without dendrites and voids, and the Li₂Te layer remained continuous and stable after peeling. Unmodified copper surfaces, in contrast, show uneven deposits, dead lithium, and dendrites protruding into the electrolyte. DFT calculations and mesoscopic simulations also confirm that the binding of Li₂Te to lithium is more conducive to the uniform nucleation of lithium atoms and promotes the

planar growth of lithium. In the full-cell test, the initial CE of the Li₂Te-Cu-based non-negative electrode-free solid-state lithium battery with NMC811 cathode was 83%, the capacity retention rate was 80% after 50 cycles, and the average CE was over 99%. However, the capacity of the unmodified copper-based battery is sharply attenuated after 10 cycles, and the performance difference is obvious. Nguyen et al. prepared an ultra-thin ZnF₂ layer by ALD on copper foil, which was electrochemically activated and converted to LiZn alloy and LiF salts to form (LiF-LiZn)/Cu current collectors.^[77] In the current collector, the LiZn alloy improves the lithiophilicity to homogeneously nucleate lithium, and LiF enhances the stability of SEI, so that the symmetrical battery can be stably cycled for more than 900 h at 3 mA cm⁻² and 3 mAh cm⁻². The capacity retention rate of the whole battery with the high-load NMC cathode is 81% for 200 cycles, and the capacity retention rate for the 50 cycles of the battery with the negative electrode is 88%. Zhang et al. designed a low Fermi level Zn-N-CNF current collector.^[78] By introducing Zn-N_x bond modification into carbon nanofibers, it restrains excessive electrolyte decomposition, promotes the formation of a thin and conductive inorganic-rich SEI, and guides planar lithium deposition. This enables highly reversible lithium plating/stripping. This current collector enables a symmetric cell to achieve stable cycling for over 6000 h at 5 mA cm⁻². Furthermore, an AFLMB maintains a capacity retention rate of 91% after 120 cycles, representing state-of-the-art performance (Table 1, 2).

Table 1. Performance comparison of recently reported works.

Material type	CE [%]	Cycle life	Advantages	Limitations	Applicable scenarios
Sn-modified layer ^[25]	98.1	800 h	Inhibit dendrite growth and deposit uniformly	It may not be uniform at high current density	High-energy density battery
Li ₂ ZnCu ₃ alloy ^[26]	99.2	200 time	Reduce nucleation overpotential	The process is rather complex.	Long-cycle demand battery
Cu _{0.64} Zn _{0.36} ^[29]	98.5	1200 h	Low surface energy, high Li adsorption energy	High-temperature process requirements	High-stability battery
In ₃ Li ₁₃ @NF ^[44]	97.4	300 time	Buffer volume expansion and uniform deposition	High cost and complex process	Batteries with high cycle stability are required
HTL-CF ^[52]	98.0	1350 time	Gradient lithium affinity, controlled deposition	The structure is complex and it is difficult to scale up	High current density battery
3D copper foam modified by Cu ₂ O ^[49]	97.6	120 time	Strong lithium affinity and low nucleation overpotential	The volume expansion buffer is limited	Medium performance requirement battery
GO@Cu-30 ^[42]	99.92	250 time	Stable SEI and low interface impedance	Organic components may have side effects	Long-cycle battery
HKUST-1 MOF ^[43]	94.1	160 h	No binder, exposed active sites; Reduce the nucleation overpotential, improve the quality of lithium deposition, inhibit dendrite growth, and ensure the long-term stable cycle of the battery	It has relatively low electrical conductivity	High-capacity retention demand battery
Fluorogold-doped titanium silicate ^[37]	96.8	600 time	Promote LiF-rich SEI	The process is complex.	Batteries with a stable SEI are required
GF@Cu ^[59]	98.8	300 time	Lightweight, high electrical conductivity	Graphene is relatively expensive	Lightweight high energy density battery
Cu-CNF-CNT ^[69]	99.0	400 time	Double conductivity, low density	Interface compatibility challenges	Flexible electronic equipment
Metallic glass ^[63]	98.2	500 time	Corrosion-resistant and with high mechanical strength	High preparation cost	Special scenarios that require high stability

Table 2. Energy density and cost comparison of recently reported works.

Material type	Energy density	Cost comparison
Traditional copper foil	Limited by the 372 mAh g ⁻¹ of the graphite anode, the energy density of the entire battery is usually less than 500 Wh kg ⁻¹	Take 6 µm copper foil as an example, the market price is ≈11,130 yuan per ton
Sn alloying (such as Li _x Sn ₂ layer) ^[25]	The energy density of the full battery is greater than 500 Wh kg ⁻¹	The cost is ≈1.5 times that of traditional copper foil
Zn alloying (such as Li ₂ ZnCu ₃ nanoarrays) ^[26]	When coupled with the LiCoO ₂ cathode (4 mAh cm ⁻²), with an N/P ratio of three, the mass energy density of the battery can reach 407.4 Wh kg ⁻¹	The cost is ≈1.8 times that of traditional copper foil
Cu _{0.64} Zn _{0.36} ^[29]	380–420 Wh kg ⁻¹	The cost is ≈2–2.5 times that of traditional copper foil
(100) Woven copper foil	165–172.5 Wh kg ⁻¹	The cost is ≈1.3 times that of traditional copper foil
MOF-CuO@ carbon coating ^[34]	450–500 Wh kg ⁻¹	The cost is ≈1.6 times that of traditional copper foil
GO@PVDF-HFP@Li-Nf composite layer ^[42]	480–520 Wh kg ⁻¹	The cost is ≈3 times that of traditional copper foil
3D copper foam (modified by Cu ₂ O) ^[49]	350–400 Wh kg ⁻¹	The preparation cost is 30% higher than that of traditional copper foil
3D copper mesh (a-RF@3D CM) ^[48]	380–420 Wh kg ⁻¹	The cost is ≈1.2 times that of traditional copper foil
HTL-CF ^[52]	420–450 Wh kg ⁻¹	The preparation cost is 40% higher than that of traditional copper foil
In ₃ Li ₁₃ @NF ^[44]	450–480 Wh kg ⁻¹	The cost is ≈2 times that of traditional copper foil
Ni ₃ S ₂ @Ni ^[66]	430–460 Wh kg ⁻¹	The cost is ≈1.8 times that of traditional copper foil
Graphene film @Cu ^[59]	448–480 Wh kg ⁻¹	The cost is ≈1.5 times that of traditional copper foil
Cu-CNF-CNT all-fiber current collector ^[69]	400–430 Wh kg ⁻¹	The cost is ≈1.7 times that of traditional copper foil
GP	380–410 Wh kg ⁻¹	The cost is ≈1.2 times that of traditional copper foil
PE/C ^[67]	300–350 Wh kg ⁻¹	The cost is basically the same as or even lower than that of traditional copper foil
MPCC ^[68]	320–350 Wh kg ⁻¹	The cost is ≈1.3 times that of traditional copper foil
CuZr metallic glass ^[63]	460–500 Wh kg ⁻¹	The cost is ≈3–5 times that of traditional copper foil
ZnO@CNT Array @ carbon cloth ^[70]	400–430 Wh kg ⁻¹	The cost is ≈1.7 times that of traditional copper foil
Composite current collector	157.5–165 Wh kg ⁻¹	The material cost is only 30% of that of traditional foil materials

6. Summary and Outlook

Amid the ongoing challenge of enhancing the energy density of LIBs, AFLMBs have achieved remarkable progress in regulating the lithium deposition behavior, suppressing dendritic growth and stabilizing the electrode interface. These advances have been realized using modification strategies such as current collector surface alloying, 3D structural engineering, and composite coating.

Nevertheless, several critical scientific challenges remain unresolved. The specific unsolved problems are as follows: insufficient long-term stability of lithiophilic interfaces (the dynamic regulation mechanism of SEI components remains unclear, and modified layers are prone to rupture, leading to abrupt impedance increase), absence of quantitative relationships between 3D structural parameters and lithium deposition uniformity, high interfacial lithium-ion migration resistance between solid-state electrolytes and modified current collectors, and material development relying on traditional trial-and-error methods.^[79] Unaddressed key points include the lack of systematic research on the multiscale synergistic regulation mechanism between nanoscale lithiophilic sites and

macroscopic porous structures, absence of compatibility studies between modified current collectors and sulfide/oxide solid-state electrolytes, and insufficiency in constructing multivariate models for current collector performance prediction using machine learning. Valuable attributes and information include core achievements of existing strategies. These include the insufficient long-term stability of lithiophilic interfaces (for example, dynamic regulation mechanisms governing the SEI components are not yet fully understood). In addition, the quantitative relationship between the 3D structural parameters—such as the pore size and surface roughness—and uniformity of lithium deposition remains unclear. Relatively high interfacial lithium-ion migration resistance between solid-state electrolytes and modified current collectors further impedes performance. Moreover, material development still heavily relies on traditional trial-and-error approaches to screen alloy compositions and coating formulations. Future research must prioritize the precise regulation of interfacial chemistry. A combined approach involving *in situ* synchrotron radiation characterization and density functional theory is necessary to comprehensively analyze phase evolution at the current collector surface during lithium

deposition and stripping. Such insights can guide the rational design of atomic-scale lithiophilic sites. Concurrently, multiscale structural optimization is necessary, particularly for the design of 3D current collectors featuring gradient lithiophilicity, and efforts should be made to quantify the correlation between the structural parameters (e.g., porosity) and cycle life. To advance compatibility with solid-state electrolytes, the development of composite interfacial layers that offer electronic and ionic conductivities is imperative to reduce interfacial impedance. Additionally, the integration of machine learning and high-throughput computational methods is expected to accelerate the identification of optimal alloy compositions and coating formulations, addressing inefficiencies inherent in traditional experimental protocols. Significantly, the large-scale development of CC is a strategic measure to comply with the vigorous development of the new energy industry, and it is of milestone significance to promote the iterative upgrading of energy storage technology. The most prominent method for large-scale development of CCs currently is template-assisted fabrication of 3D-structured collectors. In terms of manufacturing processes, the technology for preparing 3D Cu current collectors using anodic aluminum oxide (AAO) templates has high maturity. AAO templates feature highly ordered and uniform nanoporous structures, enabling precise control over the diameter and length of Cu nanowires. Through potentiostatic anodization of aluminum in acidic electrolytes, AAO templates with varying pore sizes and intervals can be produced.^[80] The three main strategies for fabricating Cu nanowire arrays—galvanostatic Cu electrodeposition after depositing a thin conductive metal layer on through-hole AAO membranes, AC Cu electrodeposition inside AAO pores without additional processing, and direct use of through-hole AAO templates as masks for Cu nanowire array electrodeposition—all exhibit excellent repeatability and stability. This allows the method to transition from laboratory-scale to industrial production, meeting the requirements for process stability and scalability in large-scale manufacturing. In terms of performance enhancement, the 3D structure provides abundant sites for lithium deposition, significantly promoting uniform lithium plating.^[81] It also effectively suppresses dendrite growth, which is crucial for improving battery safety and cycle life. In LMBs, the porous structure of 3D Cu current collectors can buffer volume changes of lithium during charge-discharge cycles, maintaining electrode structural integrity and thereby enhancing battery cycling stability. Cost-wise, AAO templates are relatively inexpensive to produce, and costs can be further reduced through process optimization. In large-scale production, AAO templates can be reused, minimizing material consumption. In contrast, some other novel current collectors rely on expensive raw materials or complex processes, leading to high costs in mass production that limit their widespread application. Additionally, this method offers good versatility. It is not only suitable for preparing 3D Cu current collectors but can also be adapted to fabricate 3D collectors of other metals or alloys by adjusting template materials and preparation conditions, catering to the needs of different battery systems.

Ultimately, by fostering comprehensive interdisciplinary collaboration across electrochemistry, material physics, and computational science, it is anticipated that original breakthroughs will

be achieved in areas such as the fundamental mechanisms of dendrite suppression, interfacial energy dissipation, and functional theoretical modeling of current collectors. These advancements will provide robust theoretical foundations and material design strategies, facilitating the real-world deployment of AFLMBs.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords: anode-free lithium metal batteries · Cu current collectors · current collector modification · lithiophilic interfaces · three-dimensional collectors

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