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Lithium Metal Anode Engineering: from Current Collectors to Hosts

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Abstract. Lithium metal anodes are regarded as the key material for next-generation high-energy-density batteries due to their ultrahigh theoretical specific capacity (3860 mAh g^{-1}) and extremely low electrochemical potential (-3.04 V vs. SHE). However, their practical application is severely restricted by lithium dendrite growth, volume fluctuation, and interfacial side reactions. This paper reviews the regulation strategies in recent years from three dimensions: current collector engineering, electrolyte engineering, and anode host engineering. Current collector engineering regulates lithium deposition through lithiophilic modification and three-dimensional structure design. Electrolyte engineering optimizes solvents, additives, and polymer electrolytes to stabilize the interface. Anode host engineering alleviates volume effects by using three-dimensional frameworks and alloying strategies. This paper aims to provide a theoretical reference for the design of high-safety and long-life lithium metal batteries.

Keywords: lithium metal anode, dendrite suppression, current collector engineering, electrolyte engineering, anode host engineering

1. Introduction

Since commercialization in the 1990s, lithium-ion batteries have achieved great success in portable electronic devices, electric vehicles and other fields. However, the energy density of traditional lithium-ion batteries based on graphite anodes is approaching the theoretical limit, which is difficult to meet the growing demand for high energy consumption. Replacing graphite anodes with lithium metal anodes has become an important direction for constructing next-generation high-energy-density batteries.

Lithium metal anodes possess an ultrahigh theoretical specific capacity (3860 mAh g^{-1}, about 10 times that of graphite) and an extremely low electrochemical potential (-3.04 V vs. SHE), and are regarded as a critical material. Nevertheless, their practical application faces severe challenges: the uncontrolled growth of lithium dendrites can pierce the separator and cause short circuits; severe volume fluctuations during cycling destroy the electrode structure and the integrity of the solid electrolyte interphase film; the high reactivity of lithium leads to unstable interfacial films, which continuously consume active lithium and electrolyte, reducing the Coulombic efficiency.

The root cause of the above problems lies in the multi-scale complexity of lithium deposition behavior, such as nucleation, growth and interface evolution. The solution is to synergistically

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regulate the deposition behavior from different levels, which can be summarized into three dimensions: current collector engineering, electrolyte engineering, and anode host engineering.

The current collector is the initial substrate for lithium nucleation. Traditional copper current collectors lack lithiophilic sites and have a high nucleation energy barrier, which easily leads to dendrites. Current collector engineering reduces the nucleation energy barrier and uniformizes the electric field through lithiophilic modification and three-dimensional structure construction, inhibiting dendrites from the source. The electrolyte is the transport medium for lithium ions, which determines the composition and stability of the interfacial film. Electrolyte engineering constructs thin, dense interfacial films with high ionic conductivity and mechanical strength by optimizing solvents, introducing additives and designing polymers, improving the reversibility of deposition/stripping. The anode host is a framework that provides physical space for lithium deposition. The three-dimensional host structure provides sufficient space to alleviate volume expansion, and guides uniform deposition through lithiophilic sites or conductive networks. Anode host engineering focuses on three-dimensional conductive frameworks, carbon-based composite materials and alloying strategies.

This paper systematically reviews the regulation strategies of lithium metal anodes from the above three dimensions: firstly, it introduces the progress of current collector engineering in regulating nucleation and deposition; secondly, it summarizes the strategies of electrolyte engineering in interface optimization and interfacial film stabilization; finally, it discusses the role of anode host engineering in regulating deposition morphology and alleviating volume changes, and prospects the future direction, so as to provide a reference for the rational design of high-safety and long-life lithium metal batteries.

2. Current collector engineering

As the initial substrate for lithium ion reduction and metallic lithium nucleation, the surface properties and structural design of current collectors have a decisive impact on lithium deposition behavior. Constructing three-dimensional conductive frameworks, introducing lithiophilic sites, and optimizing interfacial electric field distribution can effectively reduce the lithium nucleation overpotential, achieve uniform deposition, thereby inhibiting dendrite growth and improving the cycling stability of lithium metal batteries. The affinity between metallic lithium and copper is poor, resulting in a high nucleation overpotential on planar copper current collectors, and lithium ions tend to deposit preferentially in local areas to form dendrites. Therefore, lithiophilic modification of copper current collectors to induce uniform nucleation is a key strategy to suppress dendrites.

2.1 Construction of lithiophilic sites

Introducing lithiophilic sites on the surface of current collectors can significantly reduce the lithium nucleation overpotential and induce uniform nucleation. According to different mechanisms, it is mainly divided into three approaches: alloying engineering, conversion reaction engineering, and heteroatom doping engineering.

In terms of alloying engineering, Wang et al. [1] classified elements into lithiophilic elements (such as Ag, Au, Sn) with $\Delta G<0$ and non-lithiophilic elements with $\Delta G>0$ based on Gibbs free energy. The alloying process between lithiophilic elements and lithium has no nucleation barrier, and can induce preferential nucleation of lithium at low overpotential. Jung et al. [2] prepared periodic Au nanopatterns (diameter 300 nm, height 50 nm) on copper foil. Benefiting from the alloying reaction between Au and lithium, the modified layer enabled uniform lithium nucleation and

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effectively avoided dendrite growth. However, the high cost of noble metals limits their large-scale application, and exploring cheap alternatives (such as Sn, Zn) and optimizing their deposition processes are future research directions.

In terms of conversion reaction engineering, metal compounds (such as oxides, sulfides, nitrides) can react with lithium through the conversion reaction $Li^{+}+MX \rightarrow M+LiX$ (X=O, N, S, etc.) to in-situ generate metal simple substances and lithium salts. The generated metal simple substances can further form alloys with lithium, while lithium salts (such as Li_2O, LiF) have high interfacial energy, which is conducive to guiding the lateral growth of lithium. Chen et al. [3] modified a layer of silicon oxide on the copper current collector surface using laser-assisted technology, achieving a Coulombic efficiency of 99.3% and stable cycling for 250 cycles at a current density of 2.0 mA cm^{-2}. Compared with alloying engineering, conversion reaction engineering has lower material cost, but some metal compounds have poor conductivity and need to be improved by compounding conductive layers.

In terms of heteroatom doping engineering, introducing heteroatoms such as N, P, B into carbon-based materials can regulate the electronic structure of the carbon framework and enhance the adsorption capacity for lithium atoms. Ma et al. [4] co-doped N and P on carbon cloth. Density functional theory calculations showed that the binding energy between lithium atoms and carbon cloth was significantly increased from -2.7 eV to -5.2 eV, greatly enhancing the lithiophilicity of the current collector. The advantage of heteroatom doping is that it does not change the three-dimensional structure of carbon materials, and the doping is uniform and controllable, but the stability of doping sites during long-term cycling still needs further verification. Overall, the three lithiophilic site construction strategies have their own advantages and disadvantages: alloying engineering has good effect but high cost; conversion reaction has low cost but poor conductivity; heteroatom doping can regulate electronic structure but stability needs to be improved. Future research can explore the synergy of multiple strategies, such as further doping heteroatoms on the surface of metal compounds, to achieve comprehensive optimization of lithiophilicity and conductivity.

2.2 Three-dimensional carbon-based current collectors

Three-dimensional current collectors can expand the diffusion path of lithium ions from a two-dimensional plane to three-dimensional space, reduce the local current density relying on high specific surface area, and provide sufficient volume buffer space for lithium deposition. Carbon-based materials are widely studied due to their excellent conductivity, adjustable structure and low density, mainly including carbon nanotube networks, carbon cloth fiber frameworks and carbon nanofiber porous hosts.

Carbon nanotubes have high conductivity (10^{5}-10^{6} S cm^{-1}) and high mechanical strength endowed by sp^2 hybrid structure, making them ideal materials for constructing three-dimensional networks. Nilüfer et al. [5] inserted graphite particles between carbon nanotube film layers to prepare a G@CNT composite current collector. Figure 1a shows the complete preparation process of G@CNT-Li. Graphite particles not only increase the porosity of the electrode (Figure 1b-c), but also improve the lithiation capacity of the composite electrode due to their own lithium storage capacity. The material has an overpotential of only 65 mV at 0.5 mA cm^{-2} and a cycling life of more than 1400 hours; the full battery matched with the cathode still maintains a Coulombic efficiency of about 99% after 90 cycles. The carbon nanotube network can alleviate the "tip effect", confine lithium dendrites inside the three-dimensional structure, and the trapped lithium can even serve as an effective lithium source for subsequent cycling. However, the intrinsic lithiophilicity of carbon

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nanotubes is weak, and the preparation cost of high-quality CNTs is high, which needs further optimization through surface functionalization or heteroatom doping in the future.

Carbon cloth is woven by micron-sized carbon fibers, with excellent mechanical strength (tensile strength >1 GPa) and flexibility, suitable for use as self-supporting electrodes. Ouyang et al. [6] constructed carbon-coated ZnO nanotube arrays (C@ZnO NTs@CC) on carbon cloth, forming a $Li_2O/LiZn/LiC_6$ mixed ion-electron conductive interface (Figure 1d). On the one hand, the interface reduces the nucleation energy barrier by using the conversion reaction between ZnO and lithium; on the other hand, it provides a fast ion transport channel through LiZn alloy. Experiments show that lithium preferentially deposits inside the nanotubes, then fills the inter-tube gaps, forming a dense and dendrite-free morphology (Figure 1c1-h3). Compared with carbon nanotubes, carbon cloth has a lower specific surface area (about 10 $m^2\ g^{-1}$), but its mechanical strength and flexibility are more suitable for roll-to-roll processes, showing advantages in large-scale production.

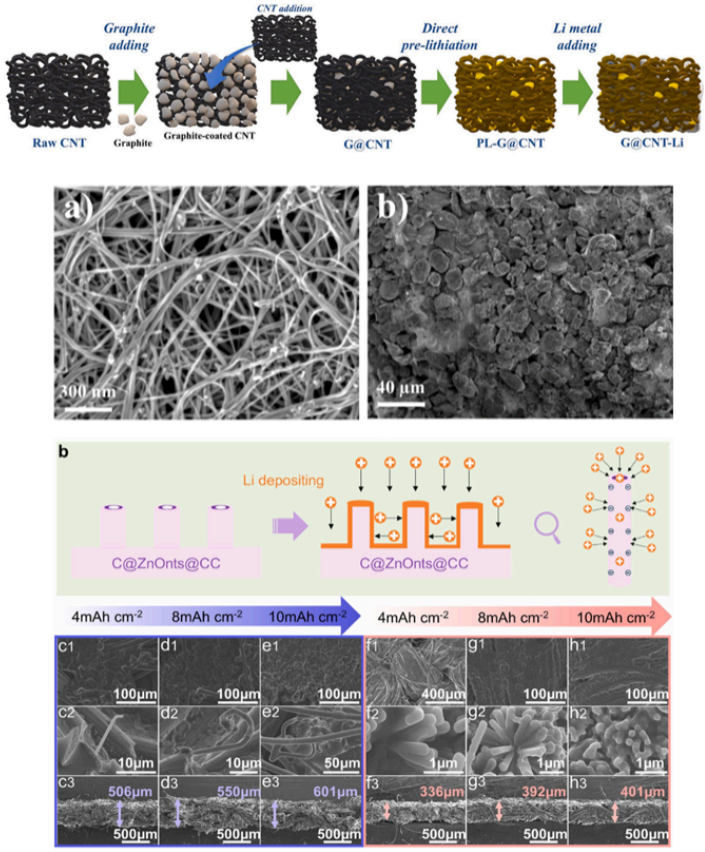

Figure 1. a. Illustration of step-by-step fabrication of G@CNT-Li; b. FE-SEM surface images of CNT film; c FE-SEM surface images of graphite-coated CNT film; dSchematic diagram of lithium deposition on C@ZnOnts@CC. The top-view SEM images of CC (c1c2, d1-d2 and e1-e2) and C@ZnOnts@CC (f1-f2, g1-g2 and h1-h2), and the cross-sectional SEM images of CC (c3 d3 and e3) and C@ZnOnts@CC (f3, g3 and h3) with areal capacity of 4 $mAh \cdot cm^{-2}$, 8 $mAh \cdot cm^{-2}$ and 10 $mAh \cdot cm^{-2}$ at 1 mA cm^{-2} respectively

Carbon nanofibers can be prepared by electrospinning combined with heat treatment, with adjustable pore structure and abundant surface defects. Wang et al. [7] prepared carbon nanofiber composite anodes with uniformly distributed ZnO nanoparticles (ZnO/CNFs) by electrospinning. ZnO grains endow carbon nanofibers with good lithiophilicity, and the interconnected three-

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dimensional structure provides abundant electrode/electrolyte interfaces, effectively reducing the local current density. The composite anode has a cycling life of 1900 hours at 0.5 mA cm^{-2}. Compared with carbon cloth, carbon nanofibers have a higher specific surface area (>200 m^2 g^{-1}), but their areal density is low, the loading capacity is limited, and the large-scale electrospinning process is difficult. The three carbon-based current collectors have their own advantages and disadvantages: carbon nanotubes are suitable for high-rate scenarios, carbon cloth for flexible devices, and carbon nanofibers for high specific surface area requirements, and the selection should be based on specific application needs.

2.3 Polymer interfacial modification layer

Polymer layers can effectively alleviate local stress during lithium deposition due to their excellent flexibility, interfacial adhesion and adjustable chemical structure, and reduce the lithium nucleation energy barrier by introducing lithiophilic functional groups (such as -C=O, -OH). Kouicem et al. [8] treated polymethyl methacrylate (PMMA) with oxygen-rich plasma to introduce active groups such as C=O and C-O on the surface, and then deposited an aluminum layer by magnetron sputtering, forming Al-O-C chemical bonds and significantly improving the adhesion between the polymer and the metal coating. The conductive protective layer can regulate the local electric field distribution, achieve uniform deposition and suppress dendrites. However, polymers are prone to swelling in electrolytes, leading to decreased interfacial bonding strength, and the long-term cycling stability of conductive polymers still needs to be improved. In addition, the thickness control of conductive polymer coatings has high requirements, and the preparation process is relatively complex. Future research can enhance the structural stability and ionic conductivity of polymer layers through cross-linking modification or compounding with inorganic nanofillers (such as SiO_2, Al_2O_3), and develop simple coating processes to reduce costs and promote their practical application in lithium metal batteries.

3. Electrolyte engineering

Electrolyte modification can significantly affect the interfacial stability and deposition behavior of lithium metal anodes. Fluorination strategy has become an important direction for electrolyte design due to its ability to enhance electrode-electrolyte interfacial stability and regulate solvation structure.

3.1 Liquid electrolyte engineering

Ether-based electrolytes have good interfacial compatibility with lithium metal anodes, but poor oxidative stability. Fluorinated ethers can improve oxidative stability, regulate solvation structure, and form LiF-rich SEI. Zhao et al. [9] designed a trifluoromethylated ether solvent TFDMP (Figure 2a). Li||Li symmetric batteries cycled for more than 1600 h in 2 mol/L LiFSI/TFDMP with a Coulombic efficiency of 99.6%; Li||NMC811 batteries had a capacity retention rate of 81% after 200 cycles. The oxygen atom of TFDMP near -CF_3 has a low electrostatic potential (-128 kJ·mol^{-1}), weakening the solvation ability, forming an anion-rich solvation structure and generating an inorganic-rich SEI [10].

Carbonate electrolytes have high oxidative stability, but ethylene carbonate (EC) is easily degraded on the lithium surface to damage the SEI. Fluorinated carbonates (FEC) can regulate the solvation structure, induce the formation of LiF-rich SEI, inhibit dendrites and enhance oxidation resistance. FEC is widely used as an additive or co-solvent. However, FEC has insufficient corrosion

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protection for nitrile solvents. Antonella et al. [9] found that soaking lithium foil in FEC to form a dense passivation layer (upper organic layer, lower inorganic layer) enabled Li||Li symmetric batteries to cycle stably for 1500 h in $LiPF_6$-ACN electrolyte (Figure 2b).

In addition, regulating lithium salt concentration, such as high-concentration electrolytes, locally high-concentration electrolytes, or introducing functional additives such as $LiNO_3$ and $CsNO_3$, can further optimize SEI components.

3.2 Polymer electrolytes

Gel polymer electrolytes (GPE) combine the high ionic conductivity of liquid electrolytes and the mechanical strength of solid polymers. A GPE-F with PVDF-HFP as matrix, [EMIM][TFSI] as ionic liquid and LASGP as filler was coated on copper foil (14 μm thick). The assembled anode-free battery achieved a Coulombic efficiency of 98.3% and voltage stability better than 5 V (Figure 2c-k). The ionic conductivity and electrochemical stability of GPE-F are better than those of GPE without fillers [11].

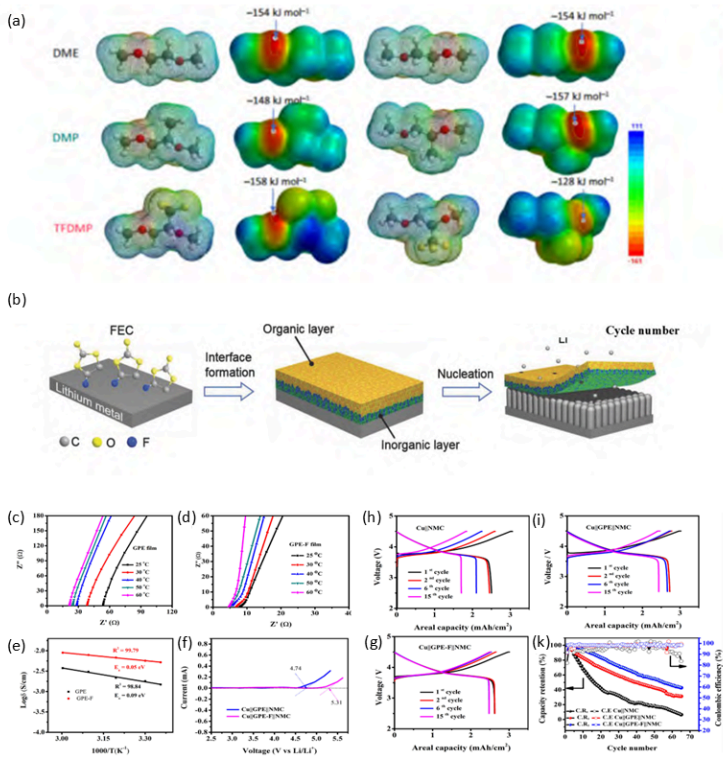

Figure 2. (a) Electrostatic potential (ESP) maps of DME, DMP and TFDMP solvents with front and back views [12], Copyright 2023, The Author(s); (b) The formation of a double-layer film on lithium metal anode after FEC treatment [13]. Copyright 2018, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim; The EIS profiles for (c) GPE film and (d) GPE-F film at various temperatures. (e) an Arrhenius plot of the GPE and GPE-F films at elevated temperature, (f) LSV curves of the GPE and GPE-F showing the electrochemical stability interval under a scan rate of 0.1mV s^{-1}; The electrochemical performance of AFLMB full cells for (h) Cu||NMC, (i) Cu||GPE||NMC, (g) Cu||GPE-F||NMC within the voltage range of 2.5–4.5V at 0.2mA cm^{-2}, (k) comparison of capacity retention and Coulombic efficiency curves for 65 cycles

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Solid polymer electrolytes (SPE) contain no liquid components, with high thermal stability and safety, but low room-temperature ionic conductivity (<10^{-5} S·cm^{-1}). Current research improves their performance through strategies such as copolymerization and introduction of fillers (LLZO, LATP).

Overall, liquid electrolytes focus on the regulation of solvation structure and interfacial chemistry, while polymer electrolytes stabilize the interface through ion transport networks. The collaborative development of the two (such as fluorinated solvent-GPE composite system, locally high-concentration gel electrolyte) is expected to promote the realization of high-safety and long-life lithium metal batteries.

4. Anode host engineering

Anode host engineering provides physical space for lithium deposition by constructing three-dimensional conductive frameworks, which effectively alleviates volume expansion, reduces local current density and guides uniform lithium deposition, serving as a key strategy to suppress dendrites and avoid "dead lithium".

4.1 Carbon-based three-dimensional hosts

Replacing planar current collectors with porous three-dimensional conductive frameworks can provide abundant lithium ion channels and uniform conductive networks, and accommodate volume changes. Zhang et al. [14] prepared composite lithium anodes by melt infusion using aluminum fluoride-doped graphene aerogel (AlF_3/GA) as the matrix (Figure 3a). AlF_3 undergoes a displacement reaction with molten lithium to in-situ generate LiF and Li-Al alloy. The gradient porous structure alleviates volume expansion, and the LiF-rich SEI improves interfacial stability. The composite anode cycled stably for 800 hours at 2 mA cm^{-2} and 2 mAh cm^{-2} with a voltage polarization of only 19 mV; the full battery matched with sulfur cathode achieved an initial specific energy of 836.62 Wh kg^{-1}.

4.2 Metal-based three-dimensional hosts

Pure lithium metal has high reactivity and poor thermal stability, and thick lithium foil (>50 μm) weakens the energy density advantage. Alloying is an effective solution. A 20 μm ultra-thin $Li_{0.7}Zn_{0.3}$ alloy prepared by melt-rolling process avoids the violent reaction between lithium and water, with excellent chemical and thermal stability. The LiZn alloy phase constructs a three-dimensional continuous framework inside the electrode, with mixed ion-electron conductivity, providing fast Li^+ channels and reducing electronic impedance. DSC tests show that the heat release of the reaction between cycled $Li_{0.7}Zn_{0.3}$ and electrolyte is reduced by 66%, and the thermal runaway trigger temperature is increased from 177.8 °C to 216.5 °C. Stable cycling for 120 times was achieved in a 53.60 Ah pouch battery with an energy density of 509.25 Wh kg^{-1} [15].

Li-Mg alloys also exhibit high ion diffusion coefficient and volume integrity. Nan et al. [16] prepared Li-Mg-20 anodes by melting method. DFT proved that its (200) crystal plane preferred orientation promotes uniform lithium deposition, and Li-Mg-20||LFP pouch batteries cycled stably for 350 cycles. The 3D Al/Mg/Li alloy framework (AM-Li-4) prepared by Li et al. [17] is composed of lithium phase, Al_4Li_9 phase and Li-Mg phase, and the symmetric battery cycled stably for 1600 h at 1 mA cm^{-2} and 1 mAh cm^{-2}.

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4.3 Functional modification of host materials

Functional modification can further improve the ion transport and deposition regulation ability of hosts. Inspired by the leaf vein network, Zhao et al. [18] constructed a patterned $Li_{0.33}La_{0.56}TiO_3$ (LLTO) nanofiber membrane as a biomimetic artificial solid electrolyte interphase (ASEI). The regular grid structure guides the uniform distribution of Li^+ flux and prevents local accumulation (Figure 3b). When LLTO contacts lithium, Ti^{4+} is reduced to Ti^{3+} and oxygen vacancies are generated, significantly reducing the Li^+ migration energy barrier (Figure 3c-e). The synergy of physical guidance and chemical vacancy mediation achieves uniform lithium deposition and dendrite suppression. BPL@Li symmetric batteries cycled stably for more than 1200 h at 1 mA cm^{-2} and 2 mAh cm^{-2}, and can operate stably for more than 70 h even under harsh conditions of 5 mA cm^{-2} and 5 mAh cm^{-2} [19]. The ASEI process is compatible with existing battery production lines and has practical application potential.

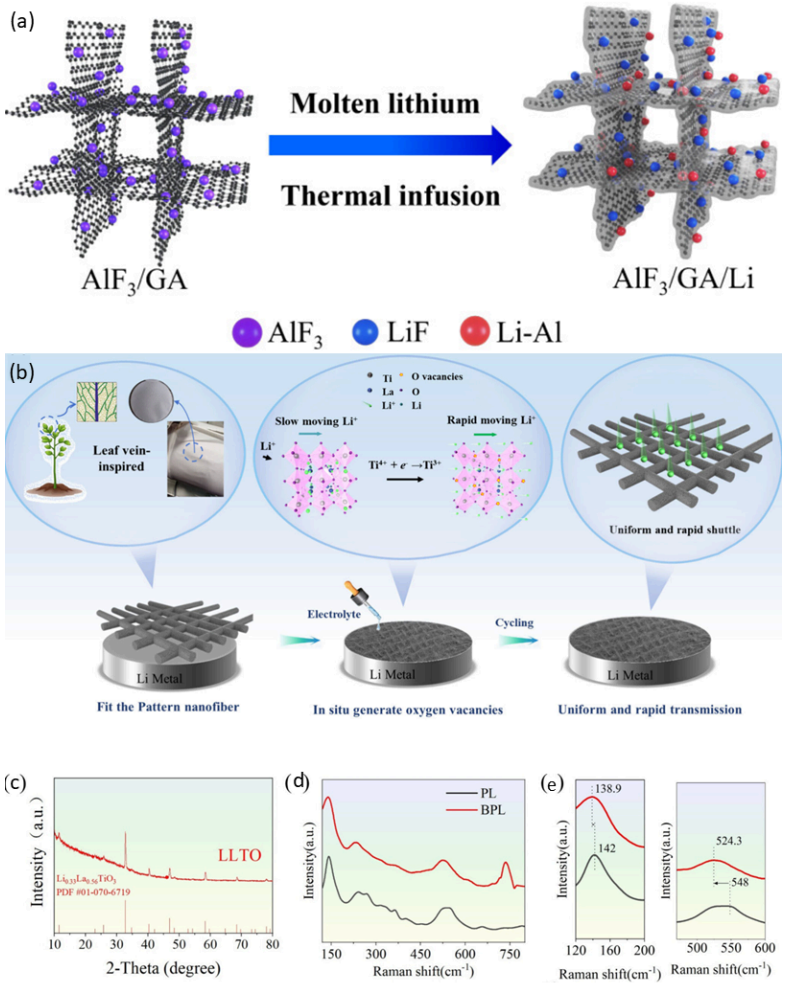

Figure 3. (a)Schematic diagram of the prepared AlF3/GA/Li composite electrode via the thermal infusion method; (b) Biomimetic design of a patterned nanofiber interphase for uniform Li flux and accelerated transport through the synergy of physical structure and oxygen vacancy channels; (c) XRD pattern of LLTO, (d) Raman spectra, and (e) corresponding enlarged details of PL and BPL. High-resolution XPS spectra of

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5. Prospect

This paper reviews the regulation strategies of lithium metal anodes from three dimensions: current collector engineering, electrolyte engineering and anode host engineering. Despite remarkable progress, their commercialization still faces challenges: the high specific surface area of three-dimensional hosts consumes active lithium, fluorinated electrolytes have high costs, and most studies stay at the coin cell level.

Future research should focus on the following directions: (1) Multi-dimensional collaborative design to realize the integration of "host-electrolyte-interface". For example, matching lithiophilic carbon cloth current collectors with fluorinated ether electrolytes is expected to achieve better interfacial stability. (2) Develop in-situ/operando characterization techniques to reveal failure mechanisms; (3) Promote practical evaluation (pouch batteries, high areal capacity, lean electrolyte); (4) Combine artificial intelligence to accelerate material screening; (5) Focus on lithium resource recovery and sustainability. Multi-disciplinary integration is expected to break through technical bottlenecks and promote the commercialization of high-safety and long-life lithium metal batteries.

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