

## Review

# Boosting the Performance of Lithium Metal Anodes with Three-Dimensional Lithium Hosts: Recent Progress and Future Perspectives

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**Abstract:** Li metal has emerged as a promising anode material for high energy density batteries, due to its low electrochemical potential and high specific capacity of 3860 mAh·g<sup>-1</sup>. These characteristics make it an attractive choice for electric vehicles and power grids. However, Li-metal batteries are plagued by dendrite issues stemming from the high reactivity of Li metal, which can ultimately result in battery failure or even safety concerns. To overcome this challenge, various strategies have been proposed to prevent dendrite formation and enhance the safety of Li-metal batteries. This review critically examines the recent progress in the development of dendrite-free Li-metal batteries, with a particular emphasis on advanced approaches of 3D Li metal host construction. Our goal is to provide a comprehensive overview of the 3D hosts for suppressing Li dendrites and to offer guidance for the future development of superior Li metal batteries.

**Keywords:** Li batteries; Li metal anode; dendrite issue; 3D hosts; 3D printing



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## 1. Introduction

The commercialization and renewal of energy storage devices has greatly promoted the developments of portable electronics and electric vehicles, which has hugely revolutionized our daily life [1–3]. Among various energy storage devices, Li ion batteries (LIBs) take a large share in the market owing to their high energy density, long cycle life and environmental friendliness [4,5]. However, the energy densities of current commercial LIBs are far below the expected value. The constant pursuit of the high-energy devices proposed demands high-capacity electrode materials. Graphite is one of the most commonly used anode materials in the market owing to its stable physical and electrochemical properties, low price and mature operation processes, but the specific capacity of a commercially used graphite anode is approaching its theoretical limit (372 mAh·g<sup>-1</sup>) [6,7]. To achieve a great enhancement in energy density, high-capacity anodes must be developed [8,9].

Li metal holds a high theoretical specific capacity of 3860 mAh·g<sup>-1</sup> and the lowest redox potential (−3.04 V vs. the standard hydrogen potential electrode) [10,11], which has been regarded as the most promising anode candidates for Li batteries. Theoretically, in case of LIBs, if a graphite anode can be successfully replaced with a Li metal anode, the energy density can be significantly improved from ~250 Wh·kg<sup>-1</sup> to ~440 Wh·kg<sup>-1</sup> [12,13]. Furthermore, metallic Li is also an indispensable anode in the Li-S and Li-O<sub>2</sub> energy storage systems, which are expected to exceed 500 Wh·kg<sup>-1</sup> in energy densities [14,15]. However, the practical application of Li metal batteries is greatly hindered by the drastic dendrite issue and large volume change in the Li anode, which seriously affects the lifespan and safety of Li metal batteries [16]. Unlike the ions' (de) intercalation energy storage mechanisms in commercial graphite anodes, Li metal anodes store energy thorough a Li plating/stripping

process [17,18]. Typically, during the charging process, Li ions gain electrons from the Li anode, are reduced into metallic Li, and simultaneously deposit onto the Li anode. During the discharging process, metallic Li loses electrons and is oxidized into Li ions, which then transfer into electrolytes and ultimately intercalate into the cathode materials. However, during practical cycling, this plating/stripping process is not quite reversible and faces great challenges. The ultrahigh reactivity of Li metal makes it prone to reacting with various liquid electrolytes, leading to the formation of a chaotic and heterogeneous solid electrolyte interphase (SEI) layer. This nonuniform SEI film induces the inhomogeneous diffusion of ions and irregular deposition of Li. Owing to the “tip effect” in which charge density tends to accumulate around sharp tips with high curvature, the subsequent Li tends to deposit on the protrusions; after long cycling, Li dendrites are formed [19]. Additionally, the uneven dissolution of Li also results in continuous electrolyte consumption and “dead Li”. These issues accelerate battery failure and pose potential safety hazards for Li metal batteries [20–22].

To address the aforementioned issues, several strategies have been reported to regulate the Li plating/stripping behaviors [23–26]. In this review, we scrutinized the approaches of dendrite-free Li metal anodes, including the modification of the anode–electrolyte interface, construction of a 3D host for Li metal deposition, and the development of rigid solid-state electrolytes, aiming to provide guidance for the development of high-energy Li metal batteries. In addition, other strategies such as modification of a separator, exploration of anode-free Li metal batteries, and external pressure are also of great significance in improving the overall performance. This mini review focuses on the design and fabrication of 3D hosts for dendrite-free anodes towards high-performance Li metal batteries. Finally, the perspectives and possible research directions towards dendrite-free Li metal anodes are also proposed.

To focus on the developments of dendrite-free Li metal anodes, in this review, we will not discuss the differences between various cathodes and electrolytes. Instead, we will provide a comprehensive overview of the common strategies that have been developed for achieving dendrite-free Li anodes.

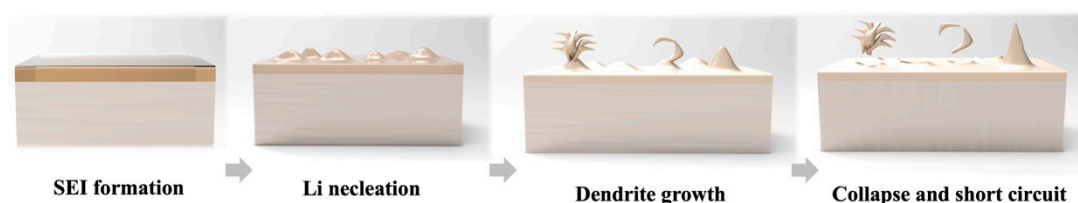
## 2. Origin of Li Dendrites

Commercial Li metal foil can be served as an anode directly, with no additional current collectors, which makes it an ideal candidate in terms of practical operation. However, the host-less Li metal faces serious challenges [27,28]. During a typical charging process, masses of solvated Li ions transfer through the electrolyte to the near-surface of the anode, where they are desolvated and reduced into Li atoms that are adsorbed onto the surface of anode. The final morphology of the electroplated Li anode is determined by the diffusion paths of these reduced Li atoms and their incorporation with metal lattice [29,30]. Conversely, during the discharging process, opposite electrochemical behaviors occur.

The electroactive Li atoms are expected to conduct plating/stripping in a two-dimensional layer-by-layer manner, resulting in a dense Li layer that provides a highly reversible anode for Li metal batteries. However, in practical occasions, the Li plating/stripping processes are often drastically uncontrollable [31–33].

Prior to normal charging/discharging processes, an insulated SEI layer is formed on the interface of the Li metal anode and the electrolyte [34–36]. However, due to the uneven electric field and unavoidable convection of cations, it is difficult to ensure homogeneous Li metal nucleation sites. Moreover, the following Li atoms are thermodynamically driven to deposit towards one dimension, resulting in the formation of dendrite whiskers rather than a two-dimensional dense layer. This is because protrusion with high curvature often gains higher current densities, causing Li atoms to deposit on the top of these protrusions and form Li dendrites [37]. Furthermore, the cation gradient in electrolyte caused by fast Li deposition will lead to local space charge near the electrode and result in branched Li deposition [38]. As a result, dendritic Li is a common occurrence in Li metal batteries, especially under high current densities [39,40]. When the Li deposition process proceeds

rapidly, the consumption rate of Li ions is much faster than the ions' diffusion rate, and the concentration gradients are enlarged and highly inhomogeneous Li deposition occurs. Due to the high reactivity of Li metal, the dendritic Li reacts with organic electrolytes and forms an insulated SEI layer, exacerbating the uneven distribution of electric field and inducing more Li dendrites, as shown in Figure 1 [41,42]. To clearly show the nucleation and growth of Li dendrites, Akihiro's group developed in-situ transmission electron microscopy (TEM) to observe this process directly, and they also provide quantitative analysis for the growth of a Li dendrite [43].



**Figure 1.** Schematic of Li dendrite growth.

The growth of Li dendrites, consumption of electrolytes, formation of an insulated SEI layer, and the uneven distribution of the electric field are all closely interconnected and mutually promoted with each other. These will lead to irreversible consumption of Li and electrolyte, as well as the internal short-circuiting, which is caused by Li dendrites, resulting in sudden capacity drop and battery failure.

Therefore, inducing smooth Li plating/stripping to inhibit Li dendrites is an effective way to realize safe and long-life Li metal batteries.

### 3. Construction of 3D Li Metal Hosts

The fabrication of conductive 3D hosts for the accommodation of host-less Li metal has been proven to be an effective strategy to address both dendrite and volume change issues [44–46]. Conductive 3D porous hosts are beneficial to induce uniform Li deposition because the increased surface area helps to decrease the current density and offer more ion diffusion pathways [47,48]. Besides, the inner space of the 3D skeleton can provide enough space for Li deposition, making it possible to achieve high Li plating capacity.

Conductive 3D hosts can be classified using the major materials of skeletons, such as conductive carbon-based and metallic-based hosts, both of which show great promise in suppressing the dendrite and volume change issues. Typically, carbon-based hosts are particularly advantageous due to their intrinsic lightweight, high conductivity, mechanical stability and the diversity of structures [47,49].

For instance, Hu's group fabricated a 3D carbon nanotube sponge (CNTS) using a typical chemical vapor deposition method. The diameter of these as-prepared CNTs ranges from 30 to 50 nm, and these self-assembled CNTs construct a 3D porous interconnected framework. Afterwards, a thin  $\text{Al}_2\text{O}_3$  layer was coated on the surface of CNTs with the help of an ALD system. The obtained 3D porous ALD-CNTs hold high specific surface area and good electrical conductivity, and have also been reported as promising hosts for uniform Li metal deposition [50]. Because 3D CNTS could provide abundant active sites for Li deposition and promote uniform charge distribution, Li dendrite growth was effectively suppressed. Compared with the  $\text{Li@Cu}$  foil counterpart, the  $\text{Li@ALD-CNTS}$  electrode delivers 2.3 times higher Coulombic efficiency after longer cycles. Xu et al. [51] reported a graphene/ $\text{MgF}_2$  framework for Li accommodation, and they prepared the freestanding  $\text{MgF}_2$  – rGO film by mixing homogeneous  $\text{MgF}_2$  + rGO solution before vacuum filtration and drying processes. After that, molten Li was absorbed into the dried composite film, forming  $\text{Mg}_x\text{Li}_y/\text{LiF-Li-rGO}$  composite anode. During cycling in both symmetric full batteries, the 3D porous rGO framework can maintain a robust structure for stable Li plating/stripping, and the well-distributed lithiophilic  $\text{MgF}_2$  sites ensure uniform Li deposition. The asymmetric cells assembled with  $\text{Mg}_x\text{Li}_y/\text{LiF-Li-rGO}$  composite anode delivers a good cycling stability of 450 h at  $1\text{ mA}\cdot\text{cm}^{-2}$  with a capacity of  $1\text{ mAh}\cdot\text{cm}^{-2}$ .

The reasonable structure design of the 3D host is crucial, but the feasibility, simplicity and economy of manufacturing processes are also non-negligible factors. Lu's group [52] deployed a one-step method to construct a 3D porous composite anode (Figure 2a). They chose copper meshes with various pore sizes and cut them into a circle shape, and then Li metal was embedded onto the copper mesh with the aid of external mechanical pressure. After this simple and cost-effective operation, the 3D Cu/Li component electrodes were successfully fabricated. Benefiting from the porous structure, this Cu/Li composite anode exhibited favored charge transfer kinetics and a smooth Li deposition process, effectively preventing dendrite growth and volume expansion. After 100 cycles, the composite anode still shows a high coulombic efficiency of 93.8%, which is much higher than that of the common anode with only 30.9% at 70 cycles.

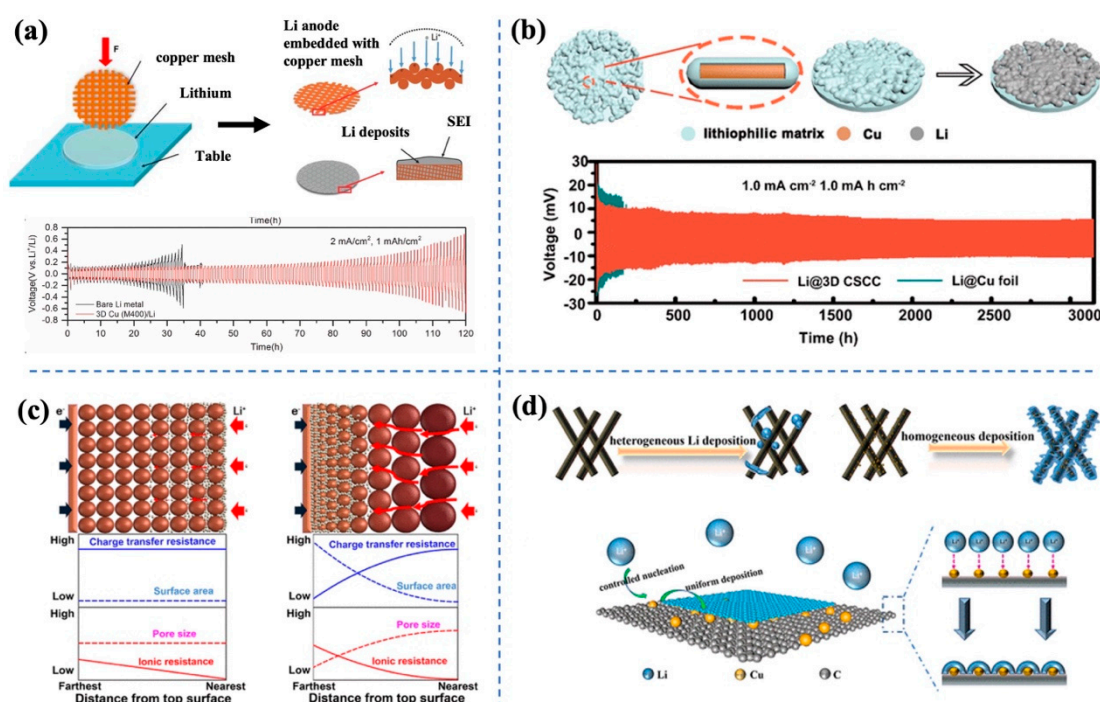
Zhang's group [53] obtained a 3D CuO and SnO<sub>2</sub> co-coated Cu host (3D CSCC) via the oxidation of commercial bronze foil by diluted aqueous ammonia solution. After holding in the ammonia solution for 36 h at 5–8 °C, the flat foil transformed into a spongy 3D structure (Figure 2b), providing an enlarged surface area and lower current density, as well as good accommodation for Li metal to release the volume change. Moreover, the modified lithophilic CuO and SnO<sub>2</sub> layer helped to lower the Li deposition overpotential and ensure homogeneous Li plating. As a result, this 3D CSCC demonstrated an excellent stability of 3000 h at 1.0 mA·cm<sup>−2</sup>.

To obtain high energy density, double-sided cathodes and anodes are commonly applied in commercial batteries. Pointing at the double-sided Li anodes for commercial Li batteries, Ma's group [54] fabricated a bidirectional porous Cu film with well-connected and tunable pores as a Li host, the through-pore structure was advantageous to the suppression of Li dendrite and the reduction of weight owing to the absence of a dense Cu layer. Benefiting from this well-designed, light-weight host, Li dendrite was released and allowed for full utilization of the anode, resulting in significant improvements in both energy density (~187.5%) and cycling life.

In addition to the above monodispersed pore frameworks, the rational design and fabrication of 3D hosts with gradient pore size can lead to better performance. Lee's group [55] explored the influence of pore size distribution on Li deposition behavior. They fabricated gradient-sized pores along the pore depth direction by sintering and sintering a commercial Cu particle. When used as a Li metal host, superconformal Li deposition was observed. Compared to the conformal Li deposition behavior in the monodisperse pore host counterpart, this superconformal deposition demonstrated superior cycling stability of 760 cycles at 2 mA·cm<sup>−2</sup> (Figure 2c). This work sheds lights on the gradient structure design and its potential in stabilizing the Li metal anode.

Zheng's group [56] synthesized a Cu-decorated carbon nanofiber (Cu-CNF) matrix through an electrospinning process in which the superior lithophilic nanocopper seeds can induce preferable Li nucleation, while the 3D host enables controlled Li growth (Figure 2d). As a result, homogeneous Li deposition was realized, and the symmetric cell assembled with Cu-CNF-scaffolded Li delivered a long cycle stability of 500 h at 5 mA·cm<sup>−2</sup>. When coupled with different cathodes including NCM811 and LFP, the half-batteries show satisfactory electrochemical performance. In contrast, a bare CNF skeleton was also prepared as a counterpart, but due to the serious mismatch of metallic Li and carbon, the nucleation and deposition of Li were uncontrollable, which also demonstrates the importance of lithophilic character for the 3D skeleton.





**Figure 2.** (a) Schematic of 3D Cu/Li component electrodes and the cycling performance of symmetric cells [52]. (b) Schematic of 3D CSCC and the cycling performance of symmetric cells [53]. (c) monodisperse pore size framework (left) and gradient pore size framework (right) [55]. (d) Schematic of Cu-CNF and Li deposition behavior [56].

#### 4. Lithiophilic Modification of Existing Substrates

In addition to the fabrication of 3D skeletons for Li accommodation, current commercial porous substrates such as carbon cloth, copper foam and nickel foam are also expected to serve as Li hosts directly. However, it is crucial for these substrates to possess a lithiophilic and smooth surface. Since the small protuberance will cause uneven current distribution and leads to inhomogeneous Li deposition, and lithiophilic surface is requested to ensure the pre-infusion of Li into the host.

Carbon cloth is a widely applied substrate in energy storage devices profiting from its 3D structure with connected networks, high conductivity, high flexibility and availability. However, it cannot be directly used as an excellent host for Li metal anode due to its lithiophobic nature and high electron conductivity. Because uncontrollable Li deposition and dendrites occur during the cycling process, this will lead to batteries' failure or even safety issues. Thus, the modification of the carbon cloth makes great sense to overcome these limitations.

Wei's group [57] developed a mixed ions/electrons conductive scaffold (MIECS) by the coating of Cu<sub>2</sub>S on the surface of carbon cloth, and the following in-situ conversion to Li<sub>2</sub>S layer with high Li ion conductivity and good Li affinity. The MIECS served as a good Li metal host by regulating Li ion flux and uniforming Li deposition, endowing the assembled batteries with excellent electrochemical performance.

Wang's group [58] decorated carbon fiber cloth with lithiophilic CuO nanoclusters through a facile hydrothermal process, as shown in Figure 3a. The ultra-stable structure allowed us to pre-store the molten Li, and facilitated homogeneous Li deposition. N, P, MnO<sub>2</sub> multidoped carbon cloth was also demonstrated as a promising host for a stable Li metal anode. The co-doped N, P and MnO<sub>2</sub> nanosheet endowed the carbon cloth with superior lithiophilicity; the molten Li was infused and evenly distributed into the host, forming an Li-based Mn@NPC-CC (Li-Mn@NPC-CC) electrode. This unique anode showed the ability to guide uniform Li plating and alleviate the volume change, and the assembled symmetric cells exhibited a prolonged lifespan of 2200 h at 1 mA·cm<sup>-1</sup> in assembled symmetric

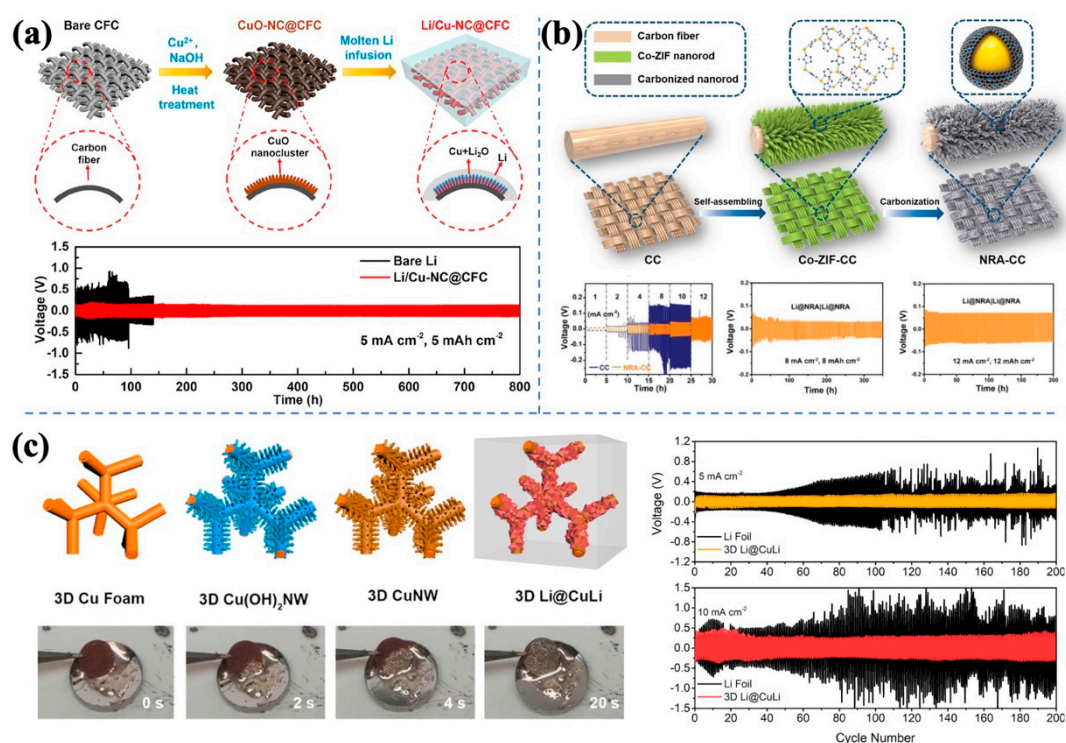
cells [59]. Chen et al. [60] reported nanoporous gold (NPG)-modified carbon fiber (CF) paper as host for the Li metal anode. First, they cleaned the carbon fiber thoroughly and then electrodeposited AuSn alloy on the surface; afterwards, the Sn component was removed by a dealloying process in mixed NaOH and H<sub>2</sub>O<sub>2</sub> solution, and the NPG@CF scaffold was constructed. When molten Li was infused into the NPG@CF scaffold, nanoporous gold converted into AuLi<sub>3</sub> and formed the Li–AuLi<sub>3</sub>@CF electrode. After all the aforementioned procedures, they assembled a coin battery with the Li–AuLi<sub>3</sub>@CF electrode and Li foil, and stripped an amount of Li from the Li–AuLi<sub>3</sub>@CF electrode to achieve the designed pre-stripping Li–AuLi<sub>3</sub>@CF electrode (PS-Li–AuLi<sub>3</sub>@CF). Owing to its superior structural stability during Li stripping/plating and the guidance for uniform Li nucleation/deposition, this PS-Li–AuLi<sub>3</sub>@CF shows better electrochemical performance than its counterparts.

Fan's group [61] decorated the carbon cloth with carbonized metal–organic framework (Co-based zeolitic imidazolate framework) nanorod arrays (Figure 3b); the product is abbreviated as NRA-CC. With this surface modification, the lithiophobic carbon cloth becomes lithiophilic. Combined with the interconnected hierarchical structure, this NRA-CC holds the ability to reduce local current density, providing abundant Li nucleation sites and accommodating volume change. Thus, this 3D NRA-CC benefits uniform Li deposition even under high current density and high areal capacity. The symmetric cells exhibited a long cycling life of over 200 h under 12 mA·cm<sup>−2</sup> and 12 mAh·cm<sup>−2</sup>. The solid-state batteries assembled with LiNi<sub>0.5</sub>Co<sub>0.2</sub>Mn<sub>0.3</sub>O<sub>2</sub> cathode, composite solid-state electrolyte (8LLZTO-2P(VDF-HFP)-6LiTFSI CSE films) and Li@NRA-CC anode showed 70% capacity retention after 150 cycles at a high rate of 0.5 C. Yu's group [62] also modified the carbon cloth with MOF-Derived Co@C; the Co-MOF on carbon cloth was obtained by simply dipping carbon cloth into the mixed solution of 2-methylimidazole and Co(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O for 4 h and the following drying process. After annealing at 600 °C in an Ar/H<sub>2</sub> atmosphere, the Co-MOF on the surface of the carbon cloth transformed into N-doped carbon nanosheet arrays with embedded Co nanoparticles (CN-Co), the final host is abbreviated as CC@CN-Co. Benefiting from the modification layer, the carbon cloth framework turns lithiophilic and becomes able to guide uniform Li nucleation/growth. As a result, symmetric cells with CC@CN-Co@Li electrodes can be stably cycled for 800 h with a low over potential of 20 mV.

Zheng's group [63] reported fluorinated carbon fibers (FCF) as the Li metal host; specifically, they chose PVDF as the fluorine source and heated at 600 °C under Ar to provide a fluorine atmosphere. Then, the carbon fibers were treated under this atmosphere for 3 h, and FCF were obtained. The strong electronegativity of F enabled the doped F atoms to enhance the Li wettability of the carbon skeleton. Soon after the molten Li infused, a thin, uniform LiF layer in-situ formed on the surface of the carbon skeleton. During the following electrochemical cycling, a robust and conformal LiF-dominated SEI layer was formed. This FCF skeleton could serve as an excellent host for Li stripping/plating, and the symmetric cells assembled with FCF-hosted Li anode delivered an extraordinary cycling stability of 100 cycles under an ultrahigh current density of 20 mA·cm<sup>−2</sup>, outperforming bare Li foils.

Cu foam is a readily available commercial 3D host with a desirable porous structure and high surface area. However, since Cu and Li are equilibrium-immiscible [64,65], it is difficult for Cu foam to induce homogeneous Li deposition. To serve as a favorable host for dense Li plating, surface modification of Cu foam is required.

Sun's group [66] modified 3D Cu foam with Cu nanowires (Figure 3c). These Cu nanowires on the surface greatly promoted the lithium affinity and allowed infusion of molten Li. Interestingly, upon introducing molten Li into the modified 3D host, Li reacted with Cu on the surface, resulting in the formation of a Cu–Li alloy. The researchers also found that Cu–Li crystals can serve as nucleation centers for Li deposition. Thus, benefiting from the unique structure with large surface area and abundant nucleation sites, Li could uniformly deposit onto the 3D host, enabling superior cycling performance. The full cells coupled with LiFePO<sub>4</sub> can be cycled stably after 400 cycles without obvious capacity decay.



**Figure 3.** (a) Schematic preparation process of Li/Cu-NC@CFC and the cycling performance [58]. (b) Schematic of NRA-CC [61]. (c) Schematic fabrication process of 3D Li@CuLi and its cycling performance at high current densities ( $5 \text{ mA} \cdot \text{cm}^{-2}$  and  $10 \text{ mA} \cdot \text{cm}^{-2}$ ) [66].

Following similar design principles, Chen's group [67] decorated the surface of Cu foam with a lithiophilic moss-like  $\text{Cu}_2\text{O}/\text{C}$  layer. Upon contacting with molten Li, the liquid Li infiltrated into the internal voids of the host soon, leading to the reduction of  $\text{Cu}_2\text{O}$  nanoparticles to nanosized Cu. The highly exposed Cu (100) and Cu (110) crystal plane facilitated this porous 3D structure's strong affinity towards Li. As a result, dense Li deposition was achieved and dendrite formation was eliminated.

Ma's group [68] modified Ni foam with electrodeposited CuO to guide Li deposition/dissolution behavior. They found that the Ni skeleton modified by CuO nanoneedle arrays played a better role in achieving uniform Li deposition at high areal capacity compared to granular CuO, which was mainly attributed to the sufficient interspace provided by the CuO nanoneedle arrays for accommodating drastic Li deposition. Benefiting from the above features, the symmetric cell shows a stable cycling performance of 1100 h at  $1 \text{ mA} \cdot \text{cm}^{-2}$  with a capacity of  $1 \text{ mAh} \cdot \text{cm}^{-2}$ . Mai et al. [69] developed a universal and scalable strategy to coat  $\text{Cu}_3\text{N}$  onto the surface of 3D metal frameworks, including Cu, Zn, Co, Ni and carbon paper. The introduction of the  $\text{Cu}_3\text{N}$  layer endowed the 3D skeletons with superior lithiophilicity and abundant Li nucleation sites, making them promising 3D hosts towards stable Li stripping/plating.

Instead of modifying the 3D skeletons before Li infusion per the aforementioned works, Song's group [70] developed a capped 3D Li matrix. They first infused the molten Li metal into Cu foam to fabricate the Li matrix; after that, (polyacrylic acid) PAA solution was dropped on the surface of the whole Li matrix, and PAA-capped Li matrix was obtained. Owing to the high ionic conductivity of LiPAA, Li ions are facilitated to transfer into the inner space of the skeleton, releasing the Li dendrite and volume change. Therefore, this PAA@Li matrix shows excellent cycling performance of 350 h at a high current density of  $5 \text{ mA} \cdot \text{cm}^{-2}$  with a high areal capacity of  $5 \text{ mAh} \cdot \text{cm}^{-2}$ .

For convenient comparison, various 3D hosts and their electrochemical behaviors have been listed in Table 1.

**Table 1.** Various 3D hosts and their electrochemical performance.

Host	Current Density ( $\text{mA} \cdot \text{cm}^{-2}$ )	Capacity ( $\text{mAh} \cdot \text{cm}^{-2}$ )	Cycling Life	Ref.
copper meshes	2	1	120 h	[52]
3D CSCC	1	1	3000 h	[53]
bidirectional porous Cu film	1	1	4000 h	[54]
gradient-sized pores	0.5	2	760 cycles	[55]
Cu-CNF matrix	5	1	500 h	[56]
Mn@NPC-CC	1	1	2200 h	[59]
3D NRA-CC	3	1	500 h	[59]
FCF	12	12	200 h	[61]
Cu nanowires decorated Cu foam	20	1	100 cycles	[63]
CuO modified Ni foam	10	1	200 h	[66]
	1	1	1100 h	[68]

## 5. Conclusions and Perspectives

Li metal has been regarded as the most promising candidate for high energy batteries. However, challenges such as Li dendrites and huge volume changes have seriously hindered its further application. Various strategies have been proposed to address these problems, with constructing 3D hosts for Li metal being an effective way. In this review, we introduce the origin of Li dendrites and scrutinize the recent progress of 3D hosts, mainly including direct fabrication of 3D hosts and the current modification of existing porous current collectors. These 3D hosts can provide a large surface area, adequate Li deposition sites and lower local current density, thus promoting homogeneous Li plating. In addition, the porous structure can accommodate Li metal and alleviate the volume change issue. However, the design principles and fabrication manipulation of 3D hosts are still vague, and their concrete influence on Li deposition behavior requires further investigation. Therefore, we propose the following perspectives with the expectation of providing guidance for the future research of Li hosts.

(1) Choice of host materials: the main component materials for Li hosts should be excellent ionic and electronic conductors, and the surface layer of the host must be lithophilic to induce uniform Li deposition.

(2) Structure design: most reports have focused solely on fabricating a 3D porous skeleton, neglecting the more specific structural design. However, for practical application, a rational design of pore size, shape of skeleton, distribution of Li deposition sites and even gradient structure can greatly benefit Li deposition behavior. For example, horizontally, homogeneous surface characteristics are expected to induce uniform Li deposition, but in the vertical direction, a gradient conductivity and gradient lithophilicity design is necessary because Li tends to deposit on the top of the host due to the short electron transfer path, rather than the inside sites.

(3) Fabrication method: to achieve the aforementioned structural design, conventional (electro)chemical or template-assisted methods have difficulties meeting the requirements. Fortunately, advanced additive manufacturing technology sheds light on fabricating rational 3D hosts, regardless of the complexity of the designed structure.

(4) In/ex-situ characterization and deposition mechanism: it has been widely demonstrated that 3D hosts can facilitate uniform Li deposition, but a deep understanding of Li deposition behavior inside the hosts remains unclear. Therefore, advanced characterization techniques should be applied to monitor the Li deposition process. This will enable a deep understanding of the relationship between Li deposition/dissolution behavior, electrode structure and the surface chemistry, ultimately providing valuable guidance for the design of 3D hosts.

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**Conflicts of Interest:** The authors declare no conflict of interest.

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