REVIEW ARTICLE

Layered Protective Interfacial Materials for Zinc Anodes in Aqueous Batteries: A Review

Yukun Fang[1], Zhicheng Zheng[1], Kaichi Chen[1], Beinuo Zhang[1], Zhongtao Chen[2], Yanmei Zheng[3,*], Pan Feng[1,*], Linfeng Hu[1,*] and Xinli Guo[1,*]

[1] State Key Laboratory of Engineering Materials for Major Infrastructure, School of Materials Science and Engineering, Southeast University, Nanjing 211189, China
[2] Rongxin (Nanjing) Energy Technology Co., Ltd., Nanjing 211100, China
[3] Jiangsu Co-Innovation Center of Efficient Processing and Utilization of Forest Resources, International Innovation Center for Forest Chemicals and Materials, College of Chemical Engineering, Nanjing Forestry University, Nanjing 210037, China

Abstract

Aqueous zinc-ion batteries (AZIBs) are promising for large-scale energy storage due to their high safety, low cost, and environmental friendliness. However, their practical application is hindered by severe parasitic side reactions and uncontrollable dendrite growth on zinc anodes, leading to rapid performance degradation. Constructing an artificial interphase layer (AIL) on the zinc surface has emerged as an effective strategy to isolate the electrolyte and regulate interfacial electrochemistry. Among various candidates, layered materials (LMs) with two-dimensional structures offer intrinsic advantages—including tunable interlayer channels, mechanical flexibility, and high surface areas—that homogenize Zn^{2+} flux and suppress dendrite growth. This review comprehensively summarizes recent progress in LMs as protective coatings for zinc anodes. We first elucidate the fundamental knowledge and core stabilization mechanisms of LM-based interphases, then survey recent advancements in LMs for AILs, covering interlayer intercalation and composite engineering. Finally, we discuss ongoing challenges and future perspectives toward practical high-zinc-utilization conditions, aiming to inspire the development of high-performance aqueous energy storage systems.

Keywords: aqueous zinc-ion batteries, zinc metal anode, layered materials, artificial interphase layer, ion-confinement effect.

1 Introduction

The escalating global energy crisis and deteriorating environmental conditions have accelerated the transition toward renewable energy sources, such as solar and wind power [1, 2]. However, the inherent intermittency of these clean energy sources

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***Corresponding authors:**
✉ Yanmei Zheng
zhengym@njfu.edu.cn
✉ Pan Feng
pan.feng@seu.edu.cn
✉ Linfeng Hu
linfenghu@seu.edu.cn
✉ Xinli Guo
guo.xinli@seu.edu.cn

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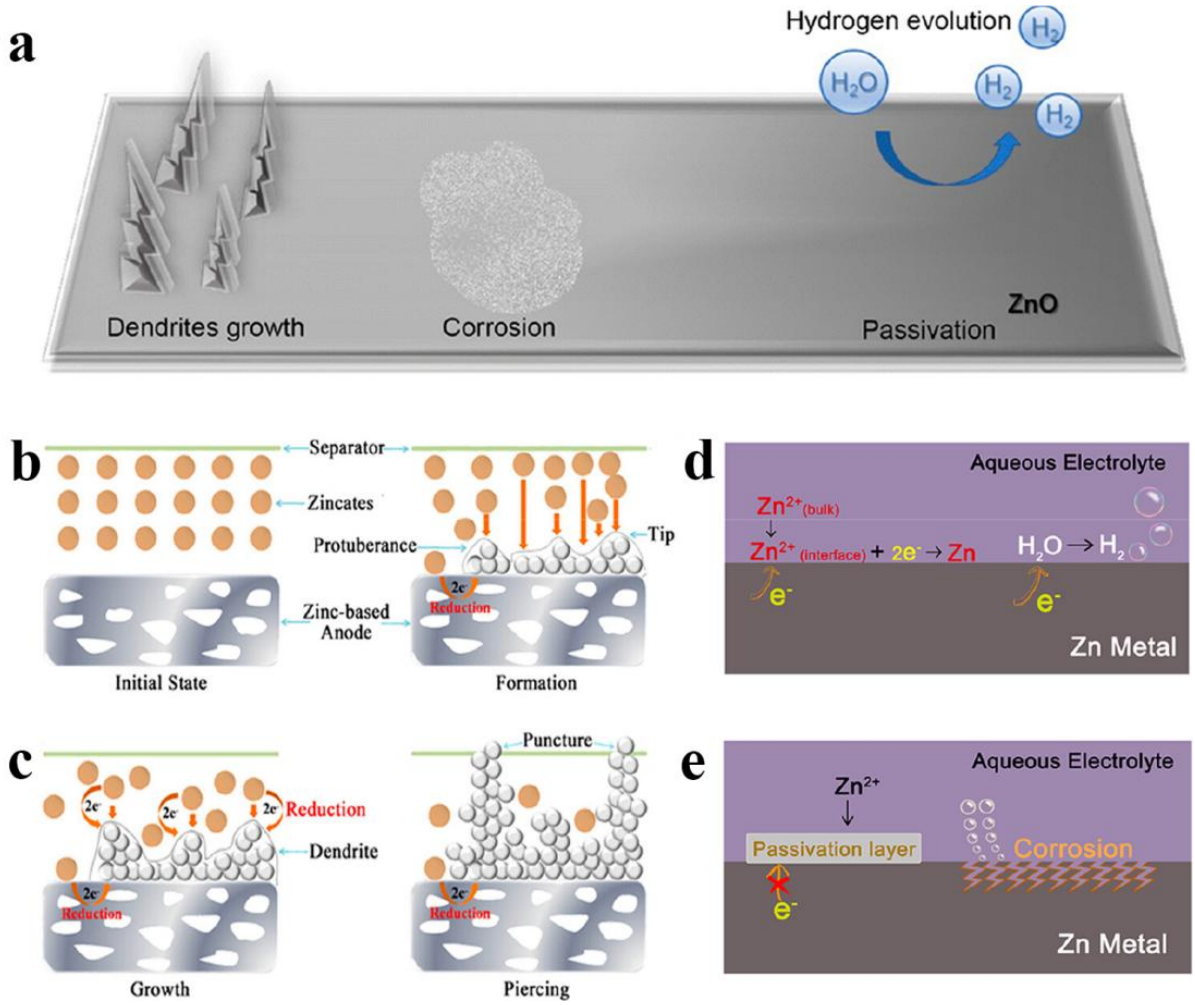

Figure 1. (a) Schematic illustration of current challenges encountered with ZMAs. (b) Formation and (c) growth processes of Zn dendrites. Schematic diagram of (d) HER and (e) corrosion and passivation. [19] Copyright 2024, American Chemical Society.

necessitates the development of large-scale and high-efficiency electrochemical energy storage (EES) systems to store surplus energy and connect the local grid for electric load, ensuring a stable and continuous power supply [3, 4]. Currently, lithium-ion batteries (LIBs) dominate the EES market due to their high energy density and long cycle life. Nevertheless, the widespread application of LIBs is increasingly hindered by significant drawbacks, including safety concerns arising from flammable organic electrolytes, the rising cost of raw materials (e.g., lithium and cobalt), and limited mineral resources [5–8]. Consequently, it is of paramount importance to explore and develop next-generation energy storage systems that prioritize safety, cost-effectiveness, and environmental sustainability to meet the burgeoning demands of the modern world [9].

Aqueous zinc-ion batteries (AZIBs) have garnered significant attention as a formidable alternative to organic-based systems since the pioneering reports on rechargeable Zn/MnO_2 chemistry [10, 11]. Unlike lithium-ion batteries that rely on toxic and flammable organic electrolytes, AZIBs utilize aqueous electrolytes, which inherently eliminate the risk of fire and explosion, thereby ensuring exceptional operational safety [12, 13]. Beyond their safety profile, zinc metal serves as an ideal anode material due to its compelling intrinsic properties, including a high theoretical gravimetric capacity (820 mAh g^{-1}), a remarkable volumetric capacity (5855 mAh cm^{-3}), and a relatively low redox potential (−0.76 V vs. SHE) [14–16]. The rapid ion-transport kinetics in aqueous media also grants AZIBs superior power density and environmental friendliness, making AZIBs an economically viable solution for large-scale grid storage [17, 18].

Despite the aforementioned merits, the practical deployment of AZIBs is severely hindered by

the intrinsic instability of the zinc metal anode (ZMA) (Figure 1(a)) [19]. During the repeated stripping/plating processes, the "tip effect" leads to localized electric field concentration, triggering the rampant growth of zinc dendrites (Figure 1(b, c)) [19–21]. These dendrites can eventually penetrate the separator, resulting in catastrophic internal short circuits. Simultaneously, the narrow electrochemical stability window of water subjects the zinc surface to severe parasitic side reactions, most notably the hydrogen evolution reaction (HER) and chemical corrosion [22–24]. The HER process not only consumes the electrolyte but also increases internal pressure and leads to the formation of insulating byproducts (e.g., $Zn_4SO_4(OH)_6{\cdot}xH_2O$), which passivate the anode surface and degrade the Coulombic efficiency [25] (Figure 1(d, e)). The typical reactions can be described as follows [19, 22, 26]:

Corrosion and HER:

$$Zn + 2H_2O \rightarrow Zn(OH)_2 + H_2 \uparrow$$

Byproduct Formation:

$$4Zn^{2+} + SO_4^{2-} + 6OH^- + xH_2O \rightarrow Zn_4SO_4(OH)_6 \cdot xH_2O \downarrow$$

To mitigate these issues, constructing an artificial **interphase layer (AIL)** has emerged as one of the most effective and straightforward strategies [27]. By introducing a protective coating—comprising inorganic materials, polymers, or composites—onto the Zn surface, one can effectively isolate the metallic zinc from the bulk electrolyte, thereby suppressing parasitic side reactions [28]. Furthermore, a well-designed AIL can homogenize the Zn^{2+} flux, regulate the nucleation overpotential [20, 29–32], and provide sufficient mechanical strength to inhibit dendrite proliferation [27], which is fundamental to achieving high zinc utilization and long-term cycling stability.

Among the diverse candidates for constructing high-performance artificial interphase layers, layered materials (LMs) have emerged as a particularly promising class of materials. LMs are generally defined as solids comprising individual sheets that are formed by various atoms linked by strong intra-layer chemical bonds while exhibiting relatively weak inter-layer forces [33, 34]. Each single layer provides a high surface-to-thickness ratio and a wealth of active sites for interfacial electrochemical processes [35]. Compared to non-layered materials (such as chaotic amorphous coatings or rigid three-dimensional bulk particulate matrices), LMs possess three unmatchable intrinsic advantages for zinc anode protection.

First, their well-defined two-dimensional interlayer galleries function as continuous, low-resistance ion-transport highways, which effectively eliminate the sluggish transport kinetics and disordered ion accumulation often observed in isotropic non-layered matrices [36, 37]. Second, the weak interlayer slip behavior endows LMs with remarkable mechanical anisotropy and flexibility; this allows the coating to intelligently accommodate the severe local volume fluctuations of the zinc anode during aggressive stripping/plating processes without cracking or delamination, a common failure mode for rigid non-layered barriers [38–40]. Third, the open and highly tailorable nature of LMs provides a versatile chemical platform where interlayer engineering (e.g., molecular intercalation, ion exchange) can precisely tune the sub-nanometer channels to achieve a strict "ion-sieving" effect [41]—allowing rapid Zn^{2+} diffusion while completely blocking bulk H_2O molecules to suppress hydrogen evolution [42, 43].

Consequently, treating layered materials as an independent, closed-loop category provides unprecedented mechanistic paradigms for interfacial design, which are fundamentally distinct from conventional non-layered optimization strategies. Up to date, literature evaluations specifically decoupled from standard organic/inorganic dichotomies to highlight the independent value of layered interphases remain highly restricted. This unique, structure-centric review uniquely satisfies this scarcity, offering a specialized and continuous conceptual thread that has been largely overlooked in broader, generalized review articles. In this review, a systematic and comprehensive summary is provided.

This review involves the recent advancements in utilizing layered materials as artificial interphase layers for aqueous zinc-ion batteries. Various layered materials and their unique stabilization mechanisms are discussed, including ion-sieving effects and electric-field homogenization. Special emphasis is placed on the research progress of layered materials and their structural modification, to illustrate how these materials address the persistent challenges of dendrite growth and side reactions. Finally, this work offers insights into the future perspectives and challenges for the practical deployment of layered-material-based zinc anodes, aiming to provide

a roadmap for the development of high-energy-density and long-life aqueous energy storage systems.

2 Layered Materials and Their Acting Mechanism

LMs represent a versatile class of building blocks for stabilizing metal anodes in aqueous systems. To gain a fundamental understanding of various layered materials, the basic knowledge and mechanisms of these materials will be detailed in the context of interfacial protection in the following chapter.

2.1 Categories and Construction

Typical LMs for ZMA surface protection are illustrated in Figure 2. Based on the nature of their interlayer bonding forces, LMs can be systematically classified into van der Waals (vdW) and ionic types, which dictate their chemical stability and exfoliation behavior [44]. Van der Waals layered materials (vdW LMs) are defined by vdW force which is weaker than chemical bond by one order of magnitude, where structurally neutral host layers are held together solely by weak dispersion forces [45–47]. Graphene is the pioneer of 2D materials and vdW LMs (which is exfoliated from bulk graphite), possessing superior electrical conductivity and mechanical robustness, making it an effective interfacial component for Zn anode protection [48]. Some vdW LMs are graphene-like materials which are arranged in a honeycomb lattice similar to graphene, including graphene oxide (GO), reduced graphene oxide (rGO), hexagonal boron nitride (h-BN), and hexagonal carbon nitride (h-CN), all of which have been actively investigated for Zn anode stabilization [49]. Other vdW LMs also belong to the huge family of vdW LMs, such as those with organic ligands, including metal organic frameworks (MOFs) and covalent organic frameworks (COFs) [50], along with 2D transition metal carbides/nitrides (MXene) and chalcogenides (e.g., MoS_2, TiS_2) [45, 47]. These systems allow for relatively straightforward mechanical or liquid exfoliation and generally exhibit chemically inert gallery environments.

Conversely, ionic LMs are defined by the ionic Coulombic interaction of interlayer, where the host layers carry permanent intrinsic charges (either positive or negative) that are electrostatically balanced by exchangeable interlayer counter-ions. These LMs are represented by layered double hydroxides (LDH), layered silicates, etc. The strong Coulombic interactions in these non-vdW ionic systems impart

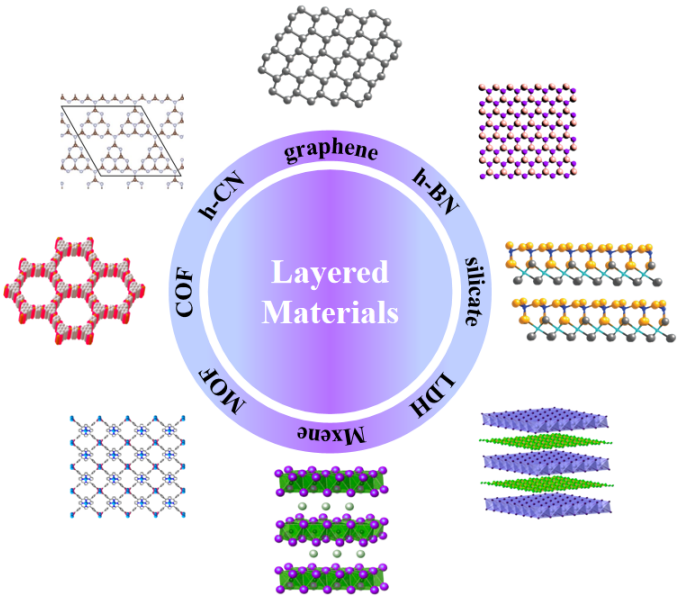

Figure 2. Schematic illustration of different kinds of typical layered materials for the protective layer of ZMA. [34, 51, 52] Copyright 2015, American Chemical Society. Copyright 2013, AAAS. Copyright 2020, Springer Nature.

robust vertical stacking stability but also open up rich host-guest chemistry, enabling remarkable ion-exchange capabilities and controllable structural swelling that are highly distinct from their vdW counterparts. While vdW layered architectures stabilize Zn anodes predominantly through precise sub-nanometer geometric confinement, highly localized atom-to-ion coordination, or tailored electronic conductivities, ionic layered systems steer interface chemistry via macro-ionic electrostatic fields, long-range Coulombic screening, and dynamic host-guest ion-exchange capabilities.

Beyond interlayer forces, LMs for Zn anode protection can also be divided into monatomic-layer and polyatomic-layer systems from the perspective of atomic configuration within the fundamental building blocks [51]. Monatomic-layer materials can be considered as the two-dimensional (2D) materials in the true sense, which consist of only a single plane of atoms in a monolayer [45, 46], including graphene and its derivatives and analogs (e.g., GO, rGO, h-BN, h-CN). In contrast, polyatomic-layer materials possess a more complex "sandwich-like" or host-guest internal structure, including MXenes, LDH, layered silicates, etc. Monatomic-layer materials offer the ultimate limit of thickness and high surface-to-volume ratios, providing a highly uniform and transparent interface for ion flux, while polyatomic-layer counterparts provide more diverse chemical sites and greater

structural tunability, allowing for sophisticated engineering of the interlaminar environment and surface functionalities to better accommodate the requirements of Zn^{2+} desolvation and deposition.

The construction methods for layered protective coatings can be categorized into ex-situ methods and in-situ methods [25, 53]. The ex-situ approach refers to introducing an external new phase, involving the pre-synthesis of layered materials irrelevant to Zn anode. The pre-synthesis method can be further divided by bottom-up and top-down strategies. The former include solution reaction (e.g. co-precipitation, hydrothermal/solvothermal treatment) or atomic/molecular layer deposition (ALD/MLD), whereas the latter include doctor blade coating, spin coating, drop coating, dip coating, sputtering and 3D printing, involving liquid exfoliation of LMs and requiring binders (e.g. PVDF, CMC, Nafion) [36, 54, 55]. In-situ construction strategies focus on the direct formation of layered structures on the Zn surface, in which the Zn foil acts as both the substrate and the zinc source, reacting with a precursor solution or atmosphere to grow a coherent protective coating. In-situ formed layers are typically synthesized via bottom-up strategies without additional binders, involving simple solution reaction, hydrothermal/solvothermal growth or electrochemical deposition, which often possess stronger adhesion and lower the risk of delamination [25, 53].

Nevertheless, their differences exist not only in interfacial adhesion, but also in dendrite suppression mechanism according to the different microstructural orientation of the layered flakes. During the blading stage, the slurry experiences strong lateral shear forces from the moving blade, which forcibly induces a horizontal alignment (parallel orientation) of the high-aspect-ratio 2D flakes relative to the substrate. This "brick-and-mortar" structure remarkably increases the tortuosity for water molecule penetration and provides robust vertical mechanical rigidity to physically block dendrite propagation [53]. Conversely, in-situ chemical or electrochemical crystalline growth typically follows substrate energy anisotropy or diffusion-controlled kinetics, often resulting in a vertical alignment or random orientation of the layered morphology [56–58]. These open vertical microchannels offer diminished activation energy barriers and faster vertical ion transport kinetics [59, 60], while their physically non-closed configuration means they may be slightly less effective in terms of providing a mechanical barrier to physically suppress dendrite penetration. Consequently, when selecting between ex-situ and in-situ construction pathways, researchers must engage in a strategic trade-off and synergistic optimization between structural controllability and interfacial integrity.

2.2 Acting Mechanism

The widespread application of LMs in stabilizing Zn anodes is rooted in their unique structural and chemical attributes. These intrinsic characteristics enable them to serve as multifunctional interphases that address multiple electrochemical challenges simultaneously. The following section details the synergistic mechanisms through which layered structures regulate ion transport and interfacial stability.

Unlike amorphous or bulk materials that require complex structural design to create certain passageway for Zn^{2+}, the intrinsic layered structure of natural materials, characterized by well-defined interlayer galleries, provides an ideal platform for precise ion transport [36]. These nano-channels possess sub-nanometer dimensions that impose severe spatial confinement on moving ions and their solvation shells, which is called "ion-confinement effect" [61, 62]. The physical/chemical environments of the channel are able to match the Zn^{2+} ionic radius (0.075 nm) or its hydrated ion diameter (0.404–0.430 nm), effectively rejecting larger interfering species (ion-sieving effect) [41, 63]. The precise control over interlayer spacing enables the modulation of the activation energy for desolvation, thereby optimizing the transition from bulk electrolyte to localized, ordered ion transport. Beyond mere physical size exclusion, ion-confinement effect involves the modulation of ionic electrochemical potentials [41]. When ion diameters are comparable to the interlayer distance, the dielectric constant of the confined solvent is significantly reduced, which profoundly weakens the solvation energy of the ions (e.g. 20.6 kJ mol^{-1} vs. 29.9 kJ mol^{-1} of bare Zn [41]). This lower energy barrier allows for faster ion movement while suppressing co-ion migration due to the overlapping of electrical double layers (EDLs) within the confined space. This "nanofluidic" behavior is essential for achieving high ion transference numbers, ensuring that the protective layer acts as a selective gate that favors the rapid transit of working ions while strictly blocking dendrite-initiating or corrosive species.

The versatility of LMs stems from their abundant

surface and interlayer active sites, which serve as sophisticated interfaces for modulating zinc ion flux and deposition behavior [36, 61]. By engineering surface functional groups and interlayer chemistry, these materials provide high-density, uniform "nucleation centers" for Zn^{2+} ions [64]. These active sites act to redistribute the local ionic concentration through tailored adsorption energies, effectively lowering the nucleation overpotential (e.g. 30.92 mV vs. 49.64 mV of bare Zn) and guiding the "bottom-up" deposition of zinc [65]. By avoiding concentrated ion flux that typically leads to uncontrolled dendrite growth, these engineered sites ensure that zinc ions are evenly distributed across the substrate, resulting in a dense, planar, and dendrite-free morphology during extended electrochemical cycling.

LMs can actively modulate the local electric field distribution through their inherent dielectric properties and tunable conductivity [36, 61]. The periodic stacking of layered sheets contributes to the homogenization of the electric field at the electrode-electrolyte interface. The uniform distribution of surface functional groups and the high aspect ratio of the layers enable an even redistribution of charge carriers. By eliminating "tip effects" and local current concentration, the stacked architecture guides a uniform ion flux across the entire electrode surface, promoting a dense and dendrite-free Zn deposition morphology.

Despite their ultra-thin geometry, LMs exhibit extraordinary mechanical strength and fracture toughness. This high mechanical robustness allows the protective layer to withstand significant volume changes and mechanical stress encountered during electrochemical cycling or harsh environmental exposure [63]. Their ability to form an impermeable "brick-and-mortar" structure effectively blocks the penetration of oxygen, moisture, and corrosive ions, acting as a physical shield that suppresses parasitic reactions such as electrochemical corrosion, hydrogen evolution reaction (HER), etc., thereby preserving the integrity of the underlying protected Zn anode [66].

3 Research Progress of Layered Materials

Due to their distinct chemical compositions and electronic structures, each class of LMs exhibits unique mechanisms in modulating Zn^{2+} solvation chemistry, guiding nucleation patterns, and providing mechanical reinforcement. This chapter proceeds to examine representative examples of LMs, with a specific focus on the correlation between their customized functionalities and the resolution of interfacial bottlenecks in aqueous Zn systems, with key electrochemical metrics summarized in Table 1.

Table 1. Summary of Electrochemical Properties of Various LMs.

LMs Types	Materials	Current density ($mA\ cm^{-2}$)	Areal capacity ($mAh\ cm^{-2}$)	Lifespan (h)	Refs.
2D Carbon Materials	NGO	5	5	300	[7]
	N-rGO	1	1	1200	[68]
	NDCN	5	1	2000	[70]
	HsGDY	2	0.5	2400	[71]
2D MOFs	2D-MOF(Mn)	4	4	2000	[72]
	NH2-MIL-125(Ti)	20	1	2800	[73]
	DZ-MOF(Zn)	5	5	5000	[56]
Mxenes	MZn-60	5	1	100	[76]
	Cu-MXene	10	1	1000	[77]
	Sn-MXene	2	0.5	2300	[78]
	MXene-mPPy	5	1	2500	[79]
LDHs	Act-LDH(CoFe)	10	5	420	[57]
	NiAl-LDH	2	1	2500	[37]
	LDH@GQDs(MgAl)	2	1	1200	[83]
	OMALDH	10	4	770	[84]
Layered Silicates	KL(Kaolin)	4.4	1.1	800	[86]
	MMT	1	0.25	1000	[26]
	Zn-Mont	2	1	700	[87]

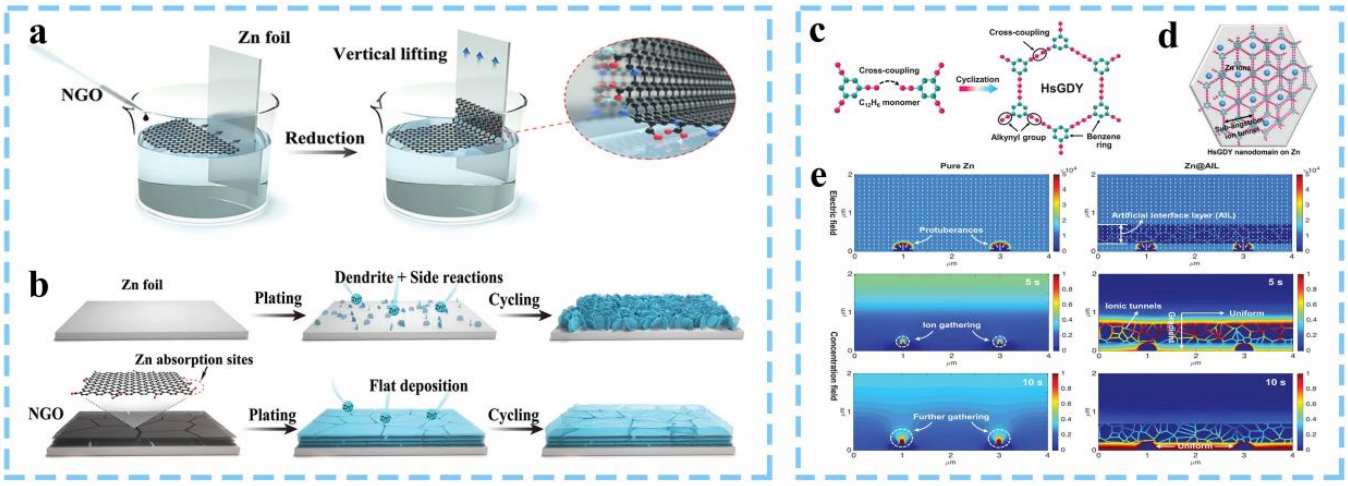

Figure 3. (a) Schematic illustration of fabricating ultrathin graphene layers on the Zn foil. (b) Schematic illustration of Zn plating on bare Zn foil (upper) and NGO@Zn electrode (lower). [7] Copyright 2021, Wiley-VCH (c) Schematic illustration of the synthesis of HsGDY. (d) Schematic illustration of the sub-ångström ion tunnel of HsGDY. (e) Dual-field simulations uncover the redistribution of Zn2+ concentration field achieved by ion-tunnel-type AIL. [71] Copyright 2020, Wiley-VCH.

3.1 2D Carbon Materials

Represented by graphene and its derivatives, 2D carbon materials have occupied a prominent position in anode protection due to their exceptional electrical conductivity, high mechanical modulus, and vast surface area [67]. These materials serve as multifunctional interphases that not only provide a robust physical barrier to suppress dendrite penetration but also act as highly conductive scaffolds to homogenize the interfacial electric field. By modulating the surface chemistry and defect density, 2D carbon layers can effectively regulate the nucleation sites and growth patterns of metal deposits. Nevertheless, their excessive electronic conductivity tends to shift the electron transfer plane to the coating's outer surface, accidentally accelerating hydrogen evolution and parasite dendrite growth atop the protective layer. Thus a balance between conductivity improving and side reaction depressing should be considered to achieve a better cycle performance.

Zhou et al. [7] successfully constructed an ultrathin (∼120 nm) artificial interface layer of nitrogen-doped graphene oxide (NGO) on zinc foil via the Langmuir-Blodgett method (Figure 3(a)). By leveraging the strong zincophilic properties of nitrogen-containing functional groups (particularly pyrrolic nitrogen) within the NGO layer, this interface effectively guides zinc ions toward (002) crystal plane directional deposition, fundamentally suppressing dendrite growth and mitigating side reactions such as the hydrogen evolution reaction and passivation (Figure 3(b)). Experimental results demonstrate that the NGO-modified zinc anode achieves stable cycling for over 300 hours at a high current density of 5 mA cm^{-2}. Furthermore, full cells paired with a $LiMn_2O_4$ cathode maintain a high energy density of 164 Wh kg^{-1} after 178 cycles, providing an effective interface engineering strategy for achieving high-energy-density and long-life aqueous zinc-ion batteries. Sun et al. [68] introduced a nitrogen-doped reduced graphene oxide (N-rGO) protective layer onto zinc anodes using a simple self-assembly method to address dendritic growth and side reactions in aqueous zinc-ion batteries. The zincophilic N-doped sites and high specific surface area of the N-rGO layer effectively lowered the Zn^{2+} nucleation overpotential and homogenized the electric field distribution, leading to uniform zinc deposition. Consequently, the N-rGO@Zn symmetric cells demonstrated exceptional long-term cycling stability (over 1200 h at 1 mA cm^{-2} and high reversibility with a Coulombic efficiency of 98.9%). Furthermore, full batteries paired with a MnVO cathode exhibited high rate capability and excellent capacity retention (97% after 2500 cycles), proving that this binder-free, self-assembled graphene interface is an effective strategy for stabilizing zinc anodes.

Nowadays, research focus has extended to 2D carbon analogs. These materials inherit the planar structural stability of graphene but introduce periodic heteroatoms (e.g., tri-s-triazine units in

g-C_3N_4) [52] or unique hybridization states (e.g., sp-sp^2 co-hybridized skeletons in GDY) [64, 69]. Such architectural modifications create "functionalized voids" and lone-pair-bearing coordination sites, which play a decisive role in precisely modulating the Zn^{2+} solvation structure and achieving highly selective ion transport at the atomic scale.

Zhu et al. [70] reported a tri-functional nitrogen-defective graphitic carbon nitride (NDCN) coating, prepared via a simple one-step calcination and spraying process, to stabilize zinc metal anodes in aqueous batteries. The NDCN layer provides abundant zincophilic sites that significantly lower the nucleation overpotential from 116.2 mV to 43.1 mV and guide zinc deposition toward a dendrite-resistant (002) crystallographic orientation. Furthermore, this protective layer effectively inhibits hydrogen evolution and side reactions, enabling NDCN@Zn symmetric cells to achieve a remarkable lifespan of over 2000 h at a high current density of 5 mA cm^{-2}. Full batteries paired with a MnO_2 cathode demonstrated superior stability, maintaining 88% capacity retention after 2300 cycles at 1 A g^{-1}. Yang et al. [71] developed a hydrogen-substituted graphdiyne (HsGDY) artificial interface layer featuring sub-ångström ion tunnels to achieve dendrite-free zinc anodes, specifically addressing the challenges under commercial-grade cathode loading conditions (Figure 3(c, d)). The HsGDY layer effectively redistributes the Zn^{2+} concentration field by spatially forcing Zn^{2+} to deviate from the irregular electric field, thereby suppressing dendrite growth and interface pulverization (Figure 3(e)). This strategy significantly extends the symmetric cell lifespan to over 2400 h and enables full batteries to maintain stability over 50,000 cycles, providing a robust solution for high-loading and long-life aqueous zinc batteries.

3.2 2D Metal Organic Frameworks (MOFs)

MOFs are distinctive crystalline porous materials characterized by a highly ordered, periodic framework of metal nodes coordinated with organic linkers, offering a unique platform with exceptionally high surface area and atomic-level structural tunability. Unlike bulkier 3D MOFs, layered MOF structures offer more accessible active sites and shortened ion transport pathways within their 2D galleries [50]. The periodic arrangement of metal nodes and organic linkers creates a well-defined ionic sieve environment, which can effectively homogenize ion flux and strip the solution shells of metal ions, thereby suppressing parasitic side reactions and promoting uniform deposition. What significantly hinders their application is the cost and scalability of their precise synthesis that limit them only to the stage of laboratory. Moreover, they often suffer from limited chemical/hydrothermal stability in aqueous electrolytes and are prone to mechanical degradation due to the rigid framework's inability to accommodate the persistent volume fluctuation of Zn metal, which impels researchers to explore the extremity of their comprehensive performance.

Cao et al. [72] developed an angstrom-level ionic sieve membrane based on a 2D-MOF(Mn) to enhance the stability and power density of aqueous zinc anodes (Figure 4(a)). Guided by DFT calculations, the 2D-MOF layer provides highly selective channels that allow the rapid passage of Zn^{2+} while effectively excluding H_2O molecules and SO_4^{2-} anions (Figure 4(b)). Compared to its 3D counterparts, the 2D-MOF exhibits superior mechanical strength, which is more conducive to suppressing dendrite growth and accommodating volume expansion. Consequently, the 2D-MOF-modified zinc symmetric cells demonstrate an exceptional lifespan of 2000 h at a high current density of 4 mA cm^{-2}. Wang et al. [73] proposed an efficient Zn^{2+} sieve strategy by employing 2D NH_2-MIL-125(Ti) nanosheets to modulate the anode chemistry in aqueous zinc-ion batteries (Figure 4(c)). The symmetric cell exhibits long-life performance under extremely high circuit density of 20 mA cm^{-2} and 50 mA cm^{-2} (Figure 4(d)). Theoretical investigations reveal that the Ti-MOF layer effectively sieves Zn^{2+} ions while blocking water molecules and SO_4^{2-} anions, thereby promoting uniform Zn deposition and alleviating the hydrogen evolution reaction (HER) and byproduct formation. Lu et al. [56] reported an in situ defect engineering strategy to construct a multifunctional defective zinc-based MOF (DZ-MOF) layer on zinc anodes to mitigate dendrite growth and side reactions. This DZ-MOF layer, characterized by upright nanosheets and anchored nanoparticles, simultaneously provides strong zincophilicity and high Zn-ion conductivity, which facilitates the desolvation process and lowers the ion migration energy barrier. Consequently, the DZ-MOF modified zinc anode exhibits exceptional cycling stability, achieving over 5000 hours of operation at 5 mA cm^{-2} and maintaining robust performance in V_2O_5-based full cells, offering a synergistic approach for high-performance aqueous

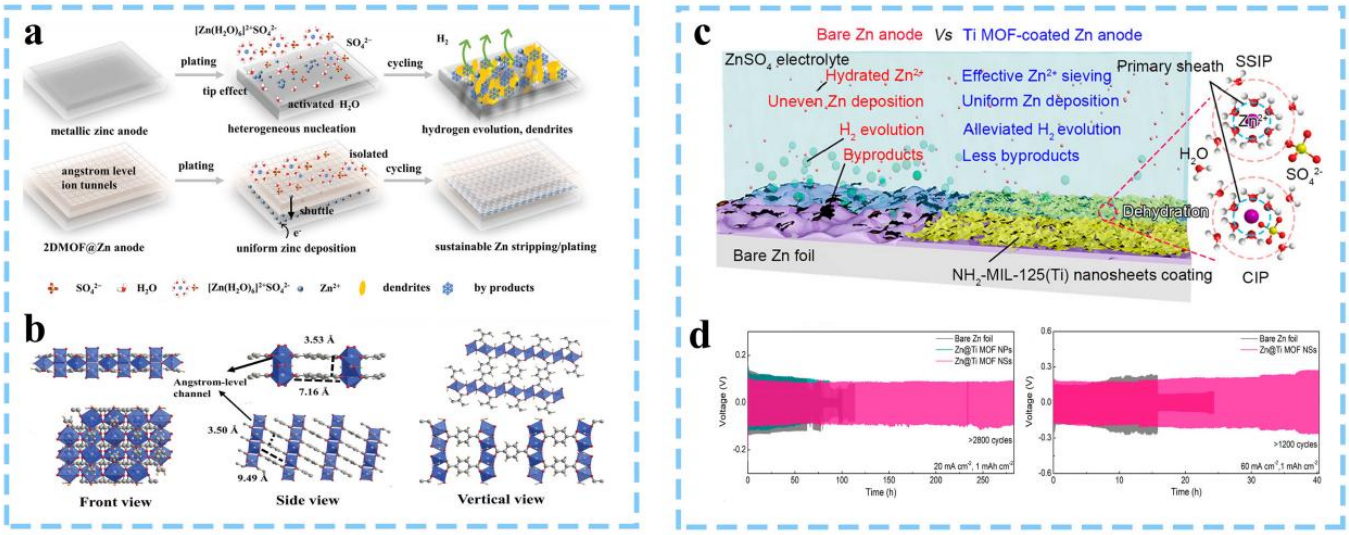

Figure 4. (a) The mechanism illustration of Zn deposition on bare Zn surface and on 2D Mn-MOF@Zn surface. (b) Crystal structure diagram of 2D Mn-MOF and 3D Mn-MOF. [72] Copyright 2023, Wiley-VCH. (c) Schematic of improving the Zn metal anode by Ti MOF Nanosheet coating. (d) cycling performance of MOF-aided symmetric cells at different current densities. [73] Copyright 2023, American Chemical Society.

zinc-ion batteries. Consequently, the MOF-coated Zn anode exhibits significantly enhanced cycling stability and reversibility, providing a profound understanding of how 2D MOF nanosheets regulate the interfacial environment for high-performance AZIBs.

3.3 MXene

MXenes are a family of 2D transition metal carbides, nitrides, or carbonitrides characterized by their high metallic conductivity and a rich variety of surface terminal groups (such as -OH, -O, and -F) [45], which collectively provide an ideal platform for fostering zincophilic interactions and ensuring rapid charge transfer. Their highly ordered layered structure provides low-resistance pathways for rapid ion diffusion, while their superior mechanical flexibility allows the protective layer to accommodate the volume fluctuations of the anode during prolonged cycling [74, 75].

The earliest application of MXene in anode protection layers was achieved by Zhang et al. [76]. They developed an in situ spontaneously reducing/assembling strategy to construct an ultrathin and uniform MXene layer on the surface of zinc anodes to address the issues of dendrite growth and side reactions (Figure 5(a, b, c)). As a result, the MXene-integrated Zn anode exhibits significantly reduced voltage hysteresis and enhanced cycling stability with a smooth dendrite-free surface (Figure 5(d)). Theoretical simulation shows this MXene coating effectively lowers the Zn nucleation energy barrier and provides a more uniformly distributed electric field through a charge redistribution effect (Figure 5(e)). When applied in zinc-ion batteries, this modification ensures high capacity retention and low polarization, offering an efficient approach for stabilizing zinc metal interfaces.

However, their highly active surface chemical states render them susceptible to irreversible oxidation in aqueous media, while their intrinsic structural flexibility often triggers severe self-restacking that blocks the engineered ion pathways. Therefore, many researchers apply different methods of structural engineering to optimize their stability during cycling process in electrolytes.

Consequently, Li et al. [77] developed a multifunctional Cu-modified $Ti_3C_2Cl_2$ MXene (Cu-MXene) protective coating via a one-step molten salt etching method to achieve highly stable zinc anodes for aqueous batteries (Figure 6(a)). The unique Cl-terminated MXene exhibits strong hydrophobicity with a contact angle of 110° that effectively isolates the zinc metal from the aqueous electrolyte, thereby suppressing hydrogen evolution and corrosion. Simultaneously, the in-situ introduced copper nanoparticles provide abundant zincophilic nucleation sites that uniformize the Zn^{2+} flux and electric field distribution, leading to dense and dendrite-free zinc deposition (Figure 6(c)). Consequently, Cu-MXene exhibited an extended cycling life of over 1000 h at 10 mA cm^{-2} in symmetric

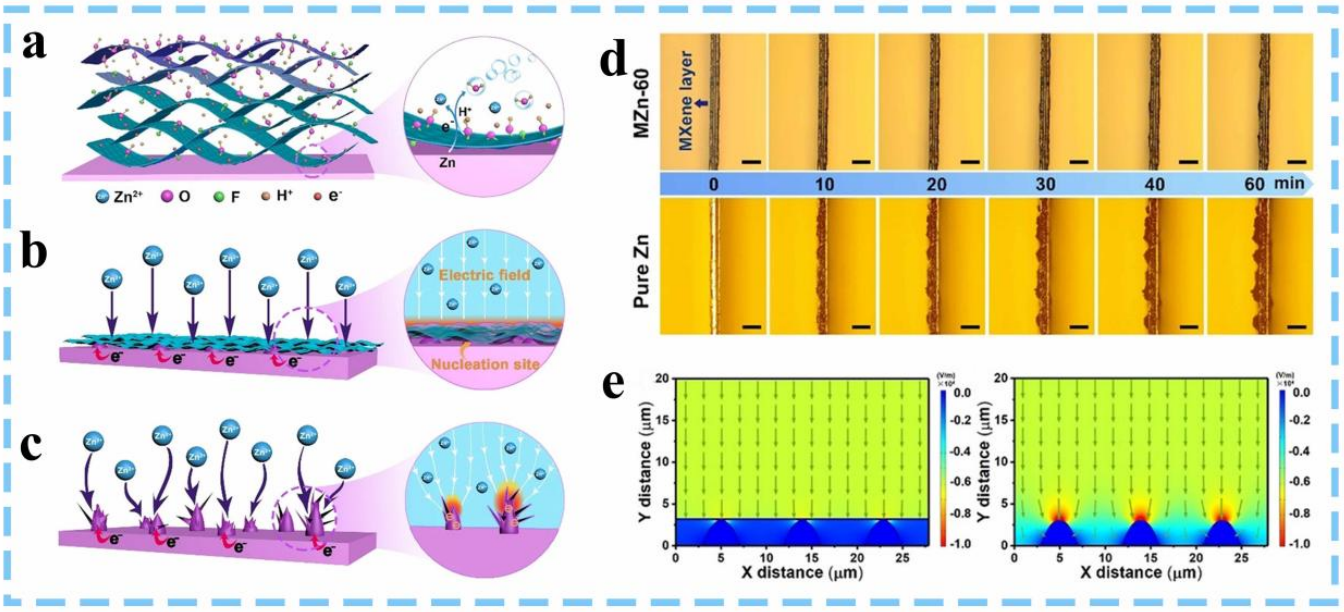

Figure 5. (a) Illustration of synchronously reducing and assembling MXene layer on the surface of Zn foil. Illustration of Zn plating behavior of (b) MXene-coated Zn, and (c) pure Zn. (d) In situ optical microscopy visualization of Zn plating on MZn-60 and pure Zn at 5 mA cm^{-2}. Scale bars: 100 μm. (e) Models of the electric field distributions for MZn-60 and pure Zn. [76] Copyright 2020, Wiley-VCH.

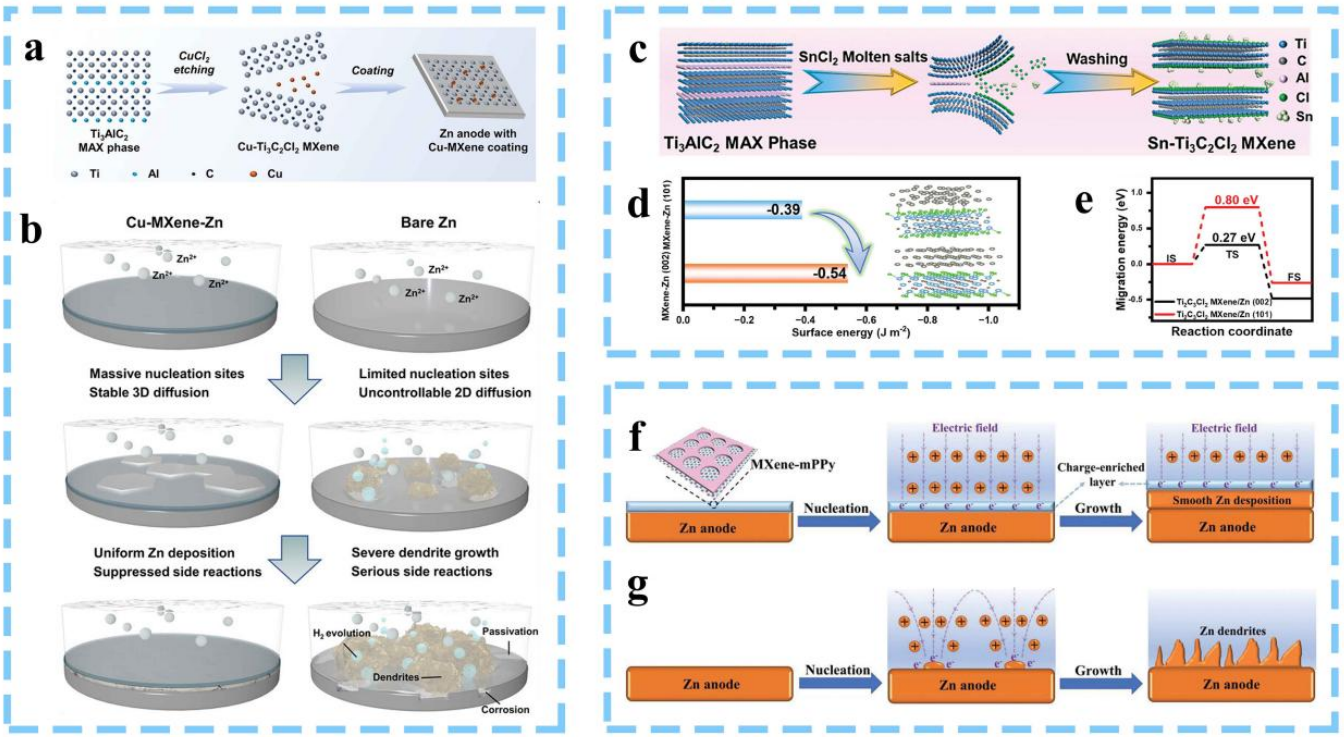

Figure 6. (a) Schematic illustration of Zn electrode with Cu-MXene coating. (b) Schematics of the Zn deposition on bare Zn and Cu-MXene-Zn surface. [77] Copyright 2023, Wiley-VCH. (c) Diagrams of the synthesis of Sn-MXene composites. (d) Calculated surface energy of the Sn-MXene-Zn (101) and (002). (e) The energy barriers of Zn^{2+} diffusion at these interfaces. [78] Copyright 2024, Wiley-VCH. Schematic illustrations of the Zn plating behaviors on (f) MXene-mPPy/Zn and (g) bare Zn. [79] Copyright 2022, Wiley-VCH.

cells and over 1000 cycles in full cells paired with a $NaV_3O_8{\cdot}7.5H_2O$ cathode. Similarly, Wang et al. [78] designed a multifunctional Sn-modified $Ti_3C_2Cl_2$ MXene (Sn-MXene) interface via the same strategy (Figure 6(c)). The Sn nanoparticles provide strong zincophilic sites that enhance Zn^{2+}

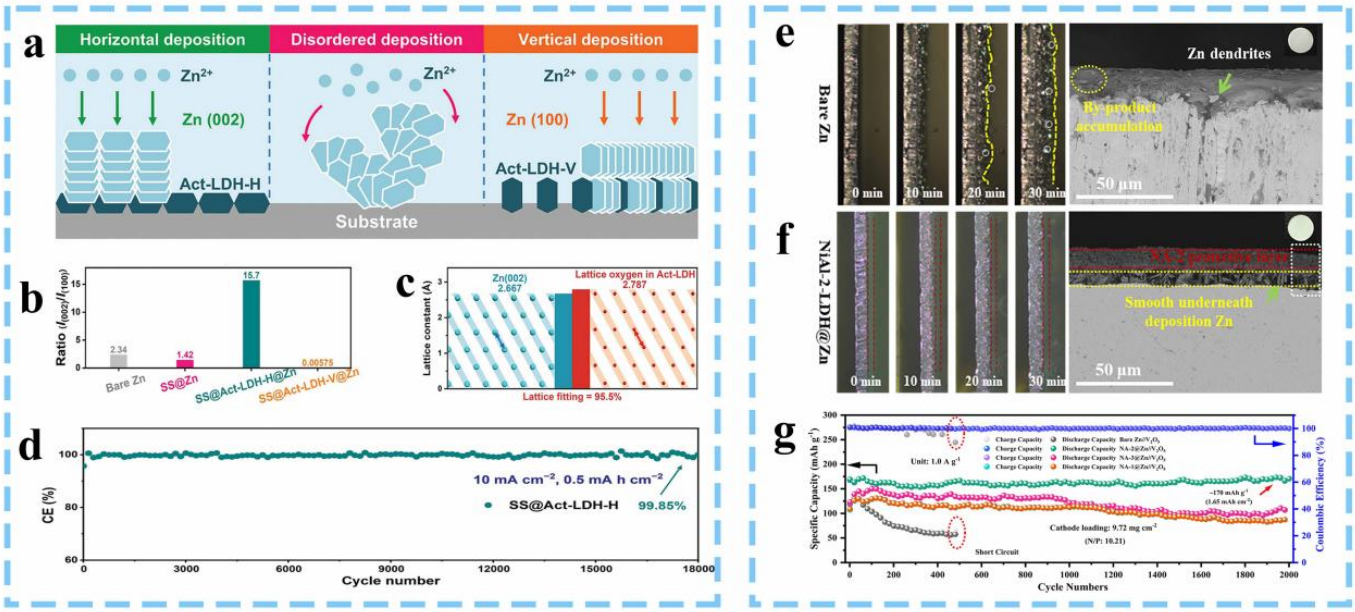

Figure 7. (a) Schematic illustration of (002), (100)-oriented Zn and unoriented Zn plating. (b) The ratio of Zn crystal planes on different plating. (c) The adsorption energy of Zn atom over LDH and Act-LDH. (d) CE plot of Act-LDH symmetric cell at 10 mA cm^{-2}, 0.5 mAh cm^{-2}. [57] Copyright 2023, Wiley-VCH. In situ optical microscopic images of Zn deposits and cross-view SEM images after 50 cycles of (e) bare Zn and (f) NiAl-LDH@Zn. (g) Long-term cycling stability comparison of Zn|V_2O_5 full cells with different anodes at 1.0 A g^{-1} with high mass loading. [37] Copyright 2023, American Chemical Society.

adsorption, while the $Ti_3C_2Cl_2$ substrate effectively lowers the surface energy of the Zn (002) plane, guiding preferential epitaxial growth along the (002) orientation (Figure 6(d,e)). Furthermore, the hydrophobic -Cl terminations of the MXene regulate the Zn^{2+} solvation structure, effectively mitigating water-induced parasitic reactions. Consequently, the Sn-MXene protected anode demonstrates significantly improved cycling stability and reversibility, offering an effective multi-interface regulation strategy for stabilizing metal anodes. Zhang et al. [79] proposed a charge-enriched strategy based on MXene-based mesoporous polypyrrole (MXene-mPPy) layers for constructing dendrite-free zinc metal anodes. This artificial interface layer exhibits an exceptional charge enrichment ability (149 F g^{-1} at 5 mV s^{-1}), which not only effectively accumulates charge but also homogenizes the distribution of the electric field and ion flux, thereby significantly suppressing zinc dendrite growth (Figure 6(f,g)). A symmetric cell using the MXene-mPPy/Zn anode achieves an ultra-long cycle life exceeding 2500 hours with excellent rate capability. When applied in MnO_2/Zn full cell, it delivers stable cycling for over 3000 cycles at a high current density of 10 A g^{-1}, with a capacity decay rate of only 0.01% per cycle.

3.4 Layered Double Hydroxides (LDHs)

LDHs are a class of ionic clays composed of positively charged brucite-like host layers and exchangeable interlayer guest anions, providing a unique ionic environment and highly tunable interlayer galleries for the precise regulation of Zn^{2+} transport in aqueous zinc battery systems [80]. This host-guest chemistry provides a highly polar environment that can effectively modulate the migration kinetics of cation fluxes. Through the strategic selection of metal cations and the intercalation of functional guest species, the interlayer spacing and electronic properties of LDHs can be tailored to create a protective barrier that simultaneously suppresses dendrite growth and inhibits electrolyte-induced corrosion.

Zhang et al. [57] reported a uniform (002)-oriented zinc metal anode achieved through a directional cation recognition and crystal assembly strategy by employing an activated LDH (Act-LDH) interface. The Act-LDH layer possesses a special vertical orientation on the surface of Zn metal which exhibits high lattice matching with the Zn (002) plane, serving as a directional recognition template to regulate the crystallographic orientation during zinc deposition (Figure 7(a)). This approach yields an ultrahigh (002)/(100) texture ratio of 15.7, effectively suppressing dendrite growth and parasitic

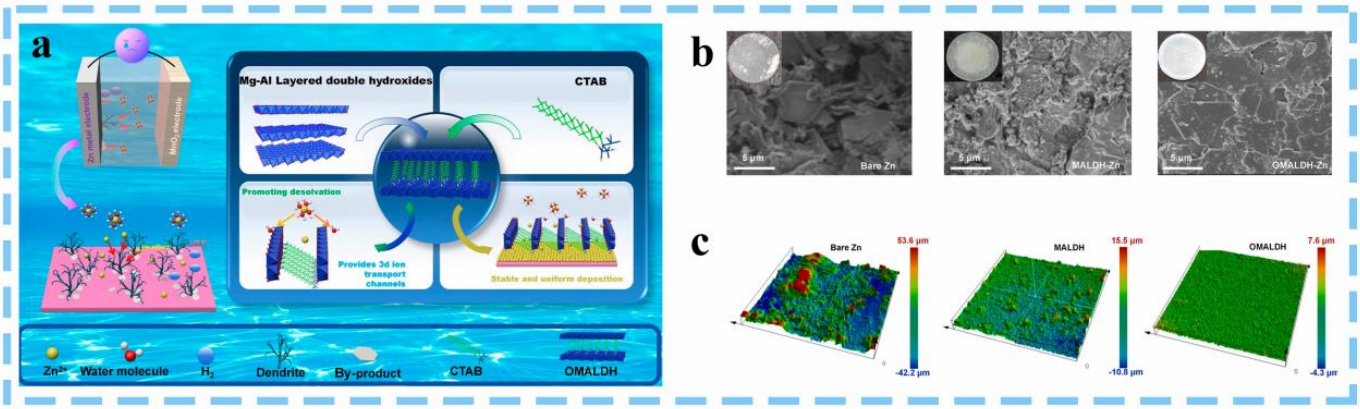

Figure 8. (a) Schematic illustration of Zn deposition on bare Zn and OMALDH-Zn electrodes. (b) SEM images, digital photos and (c) 3D CLSM images of bare Zn, MALDH-Zn, and OMALDH-Zn. [84] Copyright 2024, American Chemical Society.

side reactions. Consequently, the modified anode demonstrates an exceptional coulombic efficiency of 99.85% over 18,000 cycles and significantly enhanced stability in symmetric cells, offering a robust methodology for crystal orientation engineering in aqueous zinc batteries. Sun et al. [37] developed a hydrophilic and Zn^{2+}-conductive NiAl-LDH interface layer to enhance the reversibility and cycling stability of zinc metal anodes. The NiAl-LDH layer, characterized by its abundant hydroxyl groups and well-defined interlayer spacing, effectively homogenizes the Zn^{2+} concentration and interfacial electric field, while simultaneously serving as a physical barrier against water-induced corrosion (Figure 7(d,e)). This dual-functional interface facilitates uniform zinc deposition and suppresses dendrite growth, enabling the modified anode to achieve a high coulombic efficiency of 99.6% and stable cycling for over 2500 h at 2 mA cm^{-2}. Furthermore, the NiAl-LDH/Zn||$NH_4V_4O_{10}$ full cell demonstrates superior rate capability and long-term durability with a cathode loading of 9.72 mg cm^{-2} (Figure 7(f)).

Nevertheless, modification methods are supposed to be adopted for LDHs due to their crucial barrier of slow dissolution in mildly acidic electrolytes and high difficulty of effective processing. By employing techniques such as interlayer anion exchange, surface functionalization, and the construction of composite structures, researchers can precisely tune the d-spacing and surface polarity of LDHs [81, 82]. These modifications are designed to optimize the material's interaction with the electrolyte and the metal ions, offering a more tailored approach to addressing the complex interfacial requirements of high-performance batteries.

Liu et al. [83] developed a multifunctional hybrid interface composed of hydrophobic Mg-Al LDH and hydrophilic graphene quantum dots (GQDs) to achieve stable zinc deposition in aqueous zinc-ion batteries. The hydrophobic LDH acts as a mechanical skeleton that effectively shields the zinc anode from electrolyte corrosion, while the hydrophilic GQDs regulate the Zn^{2+} coordination environment and reduce desolvation energy to promote uniform lateral deposition along the (002) plane. Consequently, the Zn@LDH@GQDs symmetric cells demonstrated exceptional cycling stability for over 1200 h at 2 mA cm^{-2}. Furthermore, full batteries using $NH_4V_4O_{10}$ cathodes exhibited excellent stability, maintaining 2000 cycles at a high current density of 10 A g^{-1}. Wan et al. [84] developed a unique hydrophobic-ion-conducting protective layer on zinc anodes by intercalating cetyltrimethylammonium bromide (CTAB) into Mg-Al LDH (OMALDH) (Figure 8(a)). The long-chain hydrophobic cetyl groups effectively repel water molecules from the electrode surface, thereby increasing the desolvation activation energy and suppressing parasitic reactions like the hydrogen evolution reaction (HER) and corrosion. Simultaneously, the abundant zincophilic sites within the Mg-Al LDH framework regulate the nucleation behavior and promote uniform zinc deposition (Figure 8(b,c)). As a result, the OMALDH-modified zinc anode exhibits a significantly extended cycle life of over 770 h at 10 mA cm^{-2}, 4 mAh cm^{-2}. This strategy demonstrates the efficacy of organic-inorganic hybrid LDH structures in modulating both the desolvation kinetics and interfacial stability for durable aqueous zinc-ion batteries.

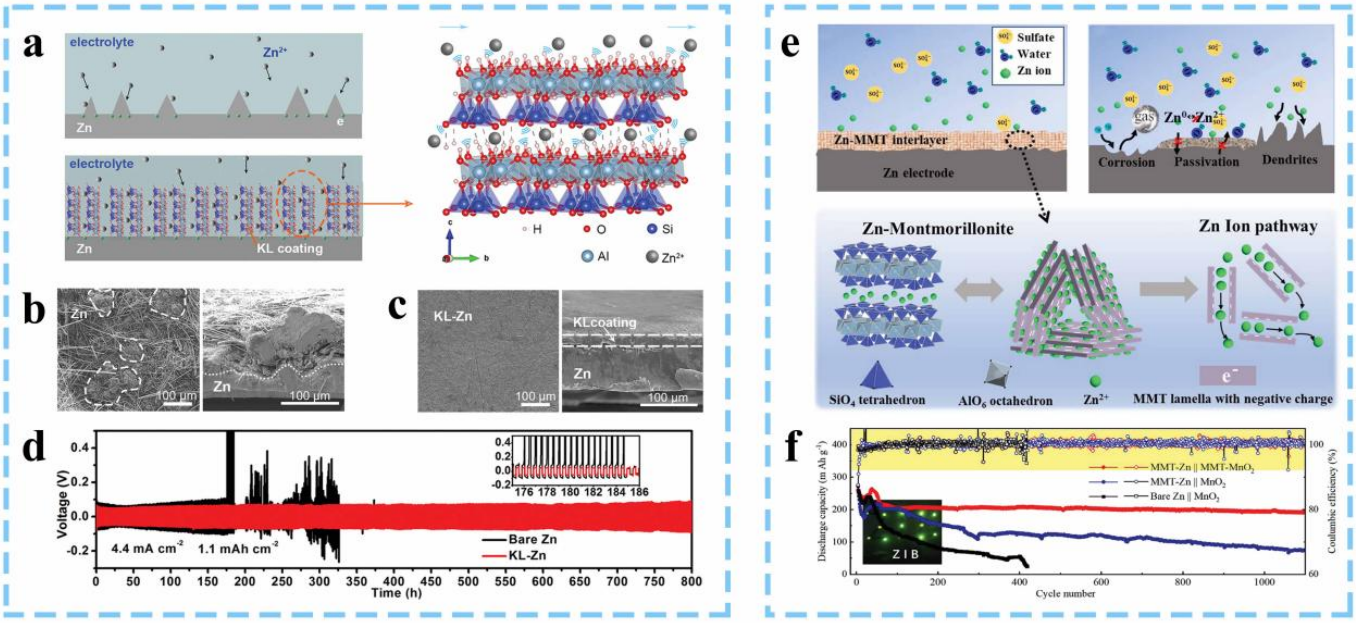

Figure 9. (a) Schematic diagrams of Zn deposition process and a detailed schematic diagram of confined Zn^{2+} transmission in kaolin. (b) SEM images of surface and cross-sectional morphology of (b) Zn and (c) KL-Zn anodes cycled with MnO_2 cathode at 0.5 A g^{-1} after 600 cycles. (d) Long-term galvanostatic cycling results of KL-Zn symmetrical cells with different electrodes at 4.4 mA cm^{-2}. [86] Copyright 2020, Wiley-VCH. (e) Schematic comparison of Zn^{2+} deposition/dissolution behaviors of Zn foil with/without MMT interlayer. (f) Long cycle performance of different MMT-Zn full cells. [26] Copyright 2021, Wiley-VCH.

3.5 Silicates

Layered silicates are mineral-based frameworks consisting of nanometer-thick tetrahedral [SiO_4] and octahedral [AlO_8 or MgO_8] sheets with inherent negative surface charges and high aspect ratios, establishing a chemically stable and tortuously-structured environment that effectively modulates ion distribution and physical barrier properties [85]. The negatively charged silicate layers, coupled with their inherent ion-exchange capacity, enable these materials to serve as effective ionic regulators at the anode interface. Their high aspect ratio and dense stacking promote the formation of a tortuous path for detrimental species while ensuring a homogenized electric field, which is instrumental in achieving long-term interfacial stability in aqueous battery systems. What layered silicates are faced with is their intrinsically low ionic conductivity and high electrical resistance that lead to severe voltage polarization and mass-transport limitations under high-rate operations, which requires further research for their wider practical application.

Deng et al. [86] proposed a sieve-functional and uniform-porous kaolin (KL) coating layer to address the dendrite growth and corrosion issues of zinc metal anodes. The KL interface layer, characterized by its intrinsic selective channels for Zn^{2+} and a uniform pore distribution (~3.0 nm), acts as an effective "ion sieve" that regulates the cation flux and ensures homogeneous zinc deposition (Figure 9(a)). This artificial interface not only facilitates dendrite-free plating but also significantly retards parasitic side reactions by isolating the zinc surface from bulk electrolyte-induced corrosion (Figure 9(b,c)). Consequently, the KL-Zn anode achieves a stable cycle life of 800 h at 1.1 mA h cm^{-2} (Figure 9(d)), and the assembled KL-Zn||MnO_2 full batteries exhibit excellent capacity retention and structural integrity. Yan et al. [26] developed a versatile zinc-based montmorillonite (MMT) interlayer to achieve an ultrafast Zn^{2+} conductor interface, effectively addressing the issues of corrosion, passivation, and dendrite growth (Figure 9(e)). The MMT-Zn coating provides a "freeway" for Zn^{2+} migration with a high transference number (t_{tr}~0.82), which significantly alleviates concentration polarization and regulates ion flux. Experimental results demonstrate that the MMT-Zn symmetric cells can maintain stable plating/stripping for over 1000 hours even at an ultrahigh capacity of 45 mA h cm^{-2} (77% depth of discharge). Furthermore, applying the MMT interlayer to the MnO_2 cathode effectively inhibits the dissolution of discharge products, enabling the MMT-Zn||MMT-MnO_2

full battery to deliver superior long-term cycling stability and high capacity retention (Figure 9(f)). Hong et al. [87] designed a fast Zn-ion conductor, Zn-intercalated montmorillonite (Zn-Mont), as an artificial solid/electrolyte interphase (SEI) to regulate the transfer and deposition behavior of Zn^{2+}. Benefiting from its unique ion-selective pathways and low migration energy barrier, the Zn-Mont coating effectively homogenizes the Zn-ion flux and accelerates the desolvation process of $[Zn(H_2O)_6]^{2+}$, thereby suppressing dendrite growth and parasitic side reactions. As a result, the Zn-Mont@Zn anode exhibits a high coulombic efficiency of 99.1% and a significantly extended lifespan of over 2000 h at 1 mA cm^{-2}. When paired with MnO_2 or V_2O_5 cathodes, the full batteries demonstrate enhanced capacity retention and rate performance, providing a promising approach for high-performance aqueous zinc-based energy storage systems.

4 Summary and Outlook

In summary, AZIBs have emerged as a promising alternative to LIBs due to their inherent safety, high theoretical capacity, and abundant raw material resources. However, the development of AZIBs is severely hindered by several challenges at the zinc anode, including dendrite growth, the hydrogen evolution reaction (HER), and passivation. Constructing an AIL has been widely adopted as a direct and effective strategy, among which layered materials exhibit unique advantages. These layers not only serve as robust physical and chemical barriers for the anode but also provide ion-transport channels, enable ion sieving, and promote uniform Zn^{2+} deposition. Consequently, this review discusses recent research progress in typical LMs used for AILs, such as 2D carbon materials, 2D MOFs, MXenes, LDHs, and layered silicates (Table 1), highlighting their significant achievements in enhancing electrochemical performance and ensuring stable cycling at high current densities.

Despite the remarkable progress of artificial interphase layers based on LMs in suppressing Zn dendrite growth and enhancing interfacial stability, several critical challenges remain before their widespread practical implementation. To further advance the industrialization of aqueous zinc-ion batteries, it is essential to deeply explore the existing limitations of AIL constructed by LMs in terms of structural stability, fabrication costs, and adaptability to real-world working conditions.

(1) Balancing structural stability with Zn^{2+} transport kinetics is central to the design of LMs interface. Volume fluctuations during cycling easily trigger structural collapse or delamination, while narrow interlaminar channels restrict rapid ion conduction and desolvation. To address these limitations, future material paradigms must focus on multi-dimensional hybridization and precise architectural regulation. Incorporating polymers or inorganic components via strong functional interactions (e.g., zincophilic sites or coordinating groups) can significantly enhance mechanical toughness and substrate adhesion. Concurrently, utilizing interlayer engineering to expand spacings or managing orientation control to induce vertical alignment can shorten diffusion pathways, achieving high ionic flux.

(2) Suppressing side reactions like hydrogen evolution and corrosion is crucial for extending cell lifespan. Future AIL constructed by LMs must transcend simple physical shielding, shifting focus toward multidimensional synergistic design with other battery components. Rather than treating anode protection as an isolated strategy, integrating it with electrolyte modulation (e.g., additive coupling) or separator modification is essential. This cross-component synergy enables in-situ defect repair, precise regulation of local solvation structures, and effective water exclusion, thereby establishing a highly efficient, system-level defense against side reactions.

(3) Providing direct evidence of the interfacial effects of layered AIL is essential for unraveling the underlying mechanisms of these layered AILs. Current research predominantly relies on ex-situ analysis, which fails to capture the real-time evolution of the interface during cycling. Future breakthroughs lie in the integration of operando characterization techniques—such as in-situ microscopy, synchrotron X-ray imaging, and in-situ Raman spectroscopy—to dynamically monitor zinc ion deposition behaviors, active water distribution, and the structural evolution of the layered materials. Concurrently, it is imperative to strengthen the synergy between experimental results and multi-scale theoretical computations (e.g., Molecular Dynamics (MD) and Density Functional Theory (DFT)) to quantitatively evaluate charge transfer, desolvation energy barriers, and interlaminar diffusion pathways at the atomic scale. This dual approach of "experiment-theory" validation will provide more precise theoretical guidance and direct scientific evidence for the rational design of high-efficiency layered interphases with tailored

functionalities.

(4) Bridging the gap between laboratory research and industrial application of layered AIL requires rigorous validation under stringent real-world operating conditions. Current evaluations often rely on coin cells with low mass loadings and excess electrolytes, which fail to capture the complexities of practical systems. Future research must shift toward performance verification under conditions of high depth of discharge (DOD), low negative-to-positive (N/P) capacity ratios, and lean electrolyte environments. Corresponding advancement strategies include: developing large-scale, continuous fabrication processes (such as roll-to-roll coating) compatible with high-loading cathodes; constructing flexible layered networks with self-healing capabilities to accommodate mechanical strains in large-format electrodes; and conducting long-term cycling assessments at the full-cell level (e.g., pouch cells) to evaluate electrochemical durability and cost-effectiveness. Only through standardized evaluation metrics that mimic practical deployment can the technical merits of layered interphases be translated into the core competitiveness of AZIBs.

Data Availability Statement

Not applicable.

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Conflicts of Interest

Zhongtao Chen is affiliated with the Rongxin (Nanjing) Energy Technology Co., Ltd., Nanjing 211100, China. The authors declare that this affiliation had no influence on the study design, data collection, analysis, interpretation, or the decision to publish. Xinli Guo served as an Editor-in-Chief, and Yanmei Zheng served as an Associate Editor of the *Journal of Advanced Materials Research* at the time of manuscript submission. To ensure the integrity of the peer-review process, neither Xinli Guo nor Yanmei Zheng was involved in the editorial handling, peer review, or decision-making process for this manuscript, which was handled independently by another editor. The remaining authors declare no conflicts of interest.

AI Use Statement

The authors declare that no generative AI was used in the preparation of this manuscript.

Ethical Approval and Consent to Participate

Not applicable.

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Yukun Fang currently studying for a master's degree in materials science and engineering from Southeast University, Nanjing 211189, China. (Email: 1500377863@qq.com)

Zhicheng Zheng received the B.S. degree in material science and engineering from Fuzhou University, Fuzhou 350108, China, in 2024. (Email: 220242761@seu.edu.cn)

Kaichi Chen received the B.S. degree in material science and engineering from Southeast University, Nanjing 211189, China, in 2024. (Email: 220246857@seu.edu.cn)

Beinuo Zhang is pursuing her Ph.D. in materials science and engineering from Southeast University, Nanjing 211189, China. Her primary research interests focus on dry electrodes and solid-state batteries. (Email: 230259241@seu.edu.cn)

Zhongtao Chen received the Ph.D. degree in materials science and engineering from Southeast University. He currently works as an engineer at Rongxin (Nanjing) Energy Technology Co., Ltd., Nanjing 211100, China. (Email: czt0425@126.com)

Yanmei Zheng is an associate professor at Nanjing Forestry University in China. She received her doctoral degree in the School of Materials Science and Engineering from Southeast University in 2023. Her current research focuses on the synthesis, performance and investigation of semiconductor photocatalysis. (Email: zhengym@njfu.edu.cn)

Pan Feng received the PhD. degree in materials science and engineering from Southeast University, Nanjing 211189, China, in 2015. She is currently a Professor at School of Materials Science and Engineering, Southeast University, Nanjing 211189, China. Her research interests include electrochemical energy storage engineering materials and organic-inorganic composite functional materials. (Email: pan.feng@seu.edu.cn)

Linfeng Hu currently working as a full professor at School of Materials Science and Engineering, Southeast University, Nanjing 211189, China. He received the PhD. degree in the National Institute of Materials Research (NIMS) in Japan. He has successively worked for the University of Aston in the UK, the Synchrotron Radiation Facility in Australia, and Fudan University in China. His research interests include polyvalent metal ion batteries, hybrid capacitors and metal-air batteries. (Email: linfenghu@seu.edu.cn)

Xinli Guo currently working at Southeast University in China as a full professor. He has worked at various research and academic institutions, including the Nanxi Mining Institute in France, the University of Seoul in South Korea, the National Institute of Materials Research (NIMS) in Japan, and the University of Osaka. He focuses on semiconductor nano-photocatalytic materials for energy conversion/storage systems. (Email: guo.xinli@seu.edu.cn)