

Progress in Electrolytes and Interfacial Regulation for Rechargeable Magnesium Batteries

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Abstract

With the rapid development of new energy systems, secondary batteries with high safety and high energy density have become key technologies for supporting smart grids and large-scale energy storage. Magnesium metal batteries are regarded as promising alternatives to lithium-based systems due to their high volumetric capacity (3833 mAh cm⁻³), abundant resources, and relatively dendrite-free deposition behavior. However, the strong solvation of Mg²⁺ leads to high desolvation energy barriers and sluggish interfacial charge-transfer kinetics. Moreover, the reductive decomposition of solvents and anions readily forms poorly Mg²⁺-conductive passivation layers on the magnesium surface, resulting in high interfacial resistance, severe deposition/stripping polarization, and reduced Coulombic efficiency.

Focusing on the critical issue of electrolyte–magnesium anode interfacial failure, this work systematically reviews the mechanisms of interfacial passivation and their effects on reversible magnesium deposition/stripping behavior. Recent advances in solvation engineering and functional additive regulation, weakly coordinating anion design, and the construction of artificial solid electrolyte interphases are highlighted. Finally, the future development of in situ interfacial characterization techniques and the synergistic design of electrolytes and artificial interphases for achieving highly stable and high-energy-density rechargeable magnesium batteries are discussed.

Keywords: electrolyte solvation structure; weakly coordinating anions (WCA); magnesium anode interface; artificial SEI

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I. Introduction

With the continuous emergence of new energy technologies, there is an urgent demand for efficient energy storage systems that can deliver electricity to users through smart grids [1–3]. Over the past decades, rechargeable lithium-ion batteries have achieved remarkable success in the battery market due to their high power output, long cycle life, and low self-discharge. However, with the increasing demand for batteries with higher energy density, the energy density of lithium-ion batteries can no longer meet practical requirements [4].

In comparison, magnesium metal used as an anode is less prone to dendrite formation and possesses a high volumetric capacity (3833 mAh cm⁻³). In addition, magnesium is abundant in natural resources and offers cost advantages [5–9], making magnesium metal batteries highly promising candidates for high-energy-density energy storage systems.

However, magnesium metal batteries currently face several challenges. On the one hand, Mg²⁺ strongly coordinates with solvent molecules, resulting in difficult desolvation and sluggish Mg²⁺ transport. When Mg²⁺ reaches the magnesium anode interface, solvent decomposition readily occurs on the magnesium surface, forming a poorly Mg²⁺-conductive interfacial passivation layer. Consequently, Mg²⁺ ions have difficulty penetrating the interphase to undergo reversible deposition/dissolution reactions at the magnesium anode [10–14], leading to high interfacial resistance and large polarization. The passivation mechanism at the magnesium anode–electrolyte interface is illustrated in Figure 1.

Although conventional chloride-containing electrolytes can alleviate surface passivation on the magnesium anode and the formed magnesium chloride species can enhance Mg²⁺ transport efficiency, these electrolytes suffer from a narrow electrochemical stability window, which limits the development of magnesium metal batteries toward higher energy density. Moreover, halides may corrode current collectors and other cell components, thereby compromising battery stability. Therefore, the development of chloride-free electrolytes that enable rapid Mg²⁺ desolvation and fast, reversible magnesium deposition/dissolution is crucial to overcoming these challenges.

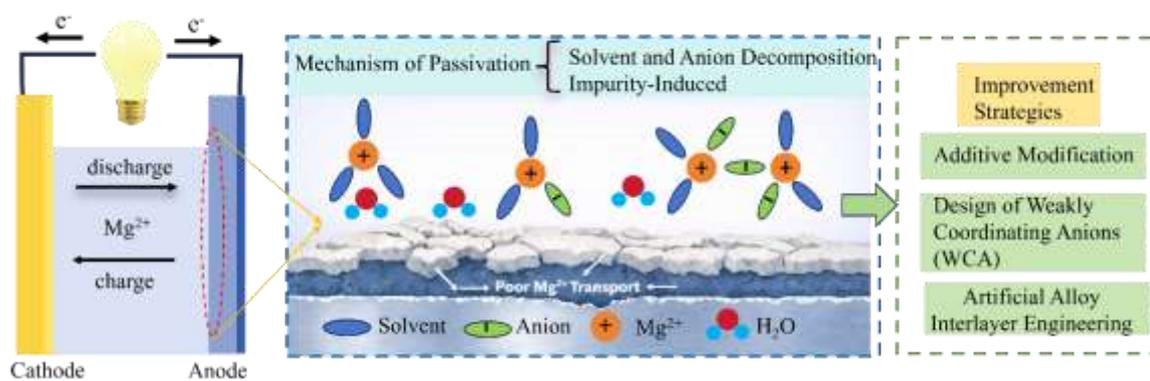


Fig. 1. Schematic illustration of rechargeable magnesium batteries: failure mechanisms and regulation strategies at the magnesium anode–electrolyte interface

At present, magnesium electrolytes mainly include organomagnesium (Grignard reagent) systems and their derivatives, magnesium aluminum chloride complex systems, and boron-based systems. Due to the highly reducing nature of the magnesium metal anode, inorganic magnesium salts containing perchlorates, hexafluorophosphates, and similar species are prone to reduction on the magnesium surface, forming electrically insulating Mg^{2+} interfacial layers [15]. In contrast, magnesium salts with bulky boron-based anions exhibit good dissociation ability and a relatively wide electrochemical stability window, enabling the formation of more Mg^{2+} -conductive interphases on the magnesium surface, and thus have attracted extensive research interest.

Various magnesium electrolytes have been developed to achieve reversible magnesium deposition/dissolution; however, their overall electrochemical performance remains unsatisfactory. The deposition current density and areal capacity of magnesium metal anodes are still limited, necessitating further improvement in the ionic conductivity of Mg^{2+} in electrolytes [16–17]. Although electrolyte component design—such as solvation structure regulation, functional additive introduction, and weakly coordinating anion construction—has partially alleviated interfacial passivation and improved deposition/stripping reversibility, the in situ solid electrolyte interphase (SEI) formed via electrolyte decomposition typically exhibits complex composition, structural heterogeneity, and continuous evolution during long-term cycling.

Such interphases inevitably contain electronically conductive or soluble components, making it difficult to maintain stable ion-transport pathways under high current densities and large areal capacities. As a result, the practical performance of magnesium metal anodes remains constrained.

To address these issues, constructing an artificial solid electrolyte interphase (artificial SEI, a-SEI) with well-defined composition and functional separation has been proposed as an effective strategy. By introducing an electronically insulating yet highly Mg^{2+} -conductive protective layer between magnesium metal and the electrolyte, direct contact between the highly reducing magnesium surface and the electrolyte can be avoided, thereby enhancing interfacial stability and prolonging cycle life.

In recent years, alloy-type [18], porous inorganic framework-type [19], and polymer/composite-type artificial SEI layers [20] have demonstrated significant advantages in suppressing side reactions, inducing uniform magnesium deposition, and improving cycling stability, gradually becoming a research hotspot in interfacial engineering for magnesium metal batteries.

This section reviews recent advances in magnesium electrolyte engineering and the construction strategies of different types of artificial SEI, as well as their regulatory mechanisms on the electrochemical behavior of magnesium metal anodes.

II. Additive-Modified Electrolytes

The solvation structure plays a critical role in determining interfacial properties. Compared with Li^+ , Mg^{2+} carries a higher charge density, leading to the formation of more stable solvation structures. Consequently, Mg^{2+} requires higher energy for desolvation. Introducing solvents with stronger chelating ability can regulate the coordination structure of Mg^{2+} , thereby tailoring SEI properties and improving deposition/stripping efficiency [21].

The group of Wang Chunsheng [22] first reported methoxypropylamine (M3) as a strong chelating solvent for magnesium batteries. M3 enters the primary solvation sheath of Mg^{2+} , reconstructs the solvation structure, and significantly reduces solvation reorganization energy. This enables low-overpotential Mg^{2+}/Mg charge transfer, suppresses electrolyte decomposition, and forms a thin and reversible interfacial layer, thereby enhancing deposition/stripping efficiency and significantly improving reversibility and cycling stability. The $\text{Mg}(\text{TFSI})_2\text{-M3/DME}$ electrolyte achieves an average Coulombic efficiency exceeding 99.5% in $\text{Mg}||\text{SS}$ cells

(Figure 2a). These chelating agents exhibit 6–41 times stronger affinity for Mg^{2+} than DME, effectively suppressing the formation of stable $[\text{Mg}(\text{DME})_3]^{2+}$ complexes. Owing to its wide anodic stability window (3.8 V vs. Mg^{2+}/Mg), the electrolyte delivers a discharge capacity of 190 mAh g^{-1} when paired with a MnO_2 cathode and maintains stable cycling for 200 cycles (Figure 2b).

In addition, additives that favor interfacial compositions conducive to Mg^{2+} transport can further enhance battery performance. The team of Sei Zhi Wei [23] introduced an organic bromide additive, 1-bromooctane (OctylBr), into the conventional $\text{Mg}(\text{HMDS})_2/\text{DME}$ electrolyte, enabling the in situ formation of a bromine-rich hybrid SEI and inducing planar magnesium deposition along the (0001) crystal plane. OctylBr does not significantly alter the bulk Mg^{2+} solvation structure but effectively suppresses passivation film formation and reduces interfacial resistance. As a result, $\text{Mg}||\text{Mg}$ symmetric cells achieve cycling lifetimes of several thousand hours, while $\text{Mg}||\text{Al}$ asymmetric cells exhibit Coulombic efficiencies approaching 99.3%. Moreover, excellent compatibility is demonstrated in full cells with Mo_6S_8 cathodes, highlighting the universal regulatory potential of organic bromide additives in magnesium electrolytes.

The Nazar group [24] reported an intrinsically stable, halide-free $\text{Mg}(\text{OTf})_2\text{-G2/TEP}$ electrolyte. Through precise tuning of contact ion pair structures, a thermodynamically and kinetically stable solvation sheath is constructed, effectively suppressing electrolyte decomposition and forming a dynamically “clean” $\text{Mg}/\text{electrolyte}$ interface during magnesium deposition. As shown in Figures 1c and 1d, unlike conventional $\text{Mg}(\text{TFSI})_2$ systems that form thick insulating interphases, this electrolyte induces the growth of dense hexagonal magnesium platelets with (0001) orientation, accompanied by only an ultrathin surface layer. Consequently, $\text{Mg}||\text{Cu}$ and $\text{Mg}||\text{Mg}$ cells maintain near-100% Coulombic efficiency ($\approx 99.8\text{--}99.96\%$) at practical current densities of $2\text{--}5 \text{ mA cm}^{-2}$ and high areal capacities up to 50 mAh cm^{-2} , and achieve stable cycling for over 2000 hours in symmetric cells (Figure 1e), even under ultrahigh areal capacity conditions. Furthermore, this electrolyte possesses an anodic stability window exceeding 4 V (vs. Mg^{2+}/Mg), enabling high-voltage organic cathodes to maintain high specific capacities over hundreds of cycles. These results demonstrate that “dynamic clean interface” engineering is an effective strategy for realizing high-energy-density rechargeable magnesium batteries.

Xiao et al. [25] systematically regulated Mg^{2+} solvation structure and interfacial chemistry by introducing imidazole-based cosolvents with different alkyl chain lengths into a chloride-free $\text{Mg}(\text{TFSI})_2\text{-DME}$ system. Among them, 1-propylimidazole (PrIm) exhibited optimal performance. PrIm enters the primary solvation sheath of Mg^{2+} and preferentially forms a dense organic-rich layer on the Mg surface, thereby suppressing DME/TFSI^- decomposition and constructing a thin, ion-conductive SEI. As a result, $\text{Mg}||\text{Mg}$ symmetric cells achieve stable cycling for over a thousand hours (Figure 1f), while $\text{Mg}||\text{Cu}$ asymmetric cells maintain Coulombic efficiencies close to 98%, along with significantly improved cycling life and rate performance in Mo_6S_8 full cells.

Overall, the solvation structure critically influences magnesium interfacial behavior. Strong chelating solvents or cosolvents (e.g., M3, PrIm) can enter the primary solvation sheath, weaken stable coordination complexes, reduce desolvation and reorganization energies, suppress electrolyte decomposition, and form thin, reversible, and ion-conductive SEI layers. These effects lower overpotential and enhance deposition/stripping efficiency and cycling stability. Interfacially oriented additives (e.g., OctylBr), although minimally affecting bulk solvation, can in situ construct bromine-rich hybrid SEI layers, reduce interfacial resistance, and induce planar magnesium deposition.

Therefore, solvation engineering and interfacial chemical additives provide effective design strategies for constructing rechargeable magnesium electrolytes with wide stability windows ($\sim 3.8\text{--}4 \text{ V}$) and high reversibility.

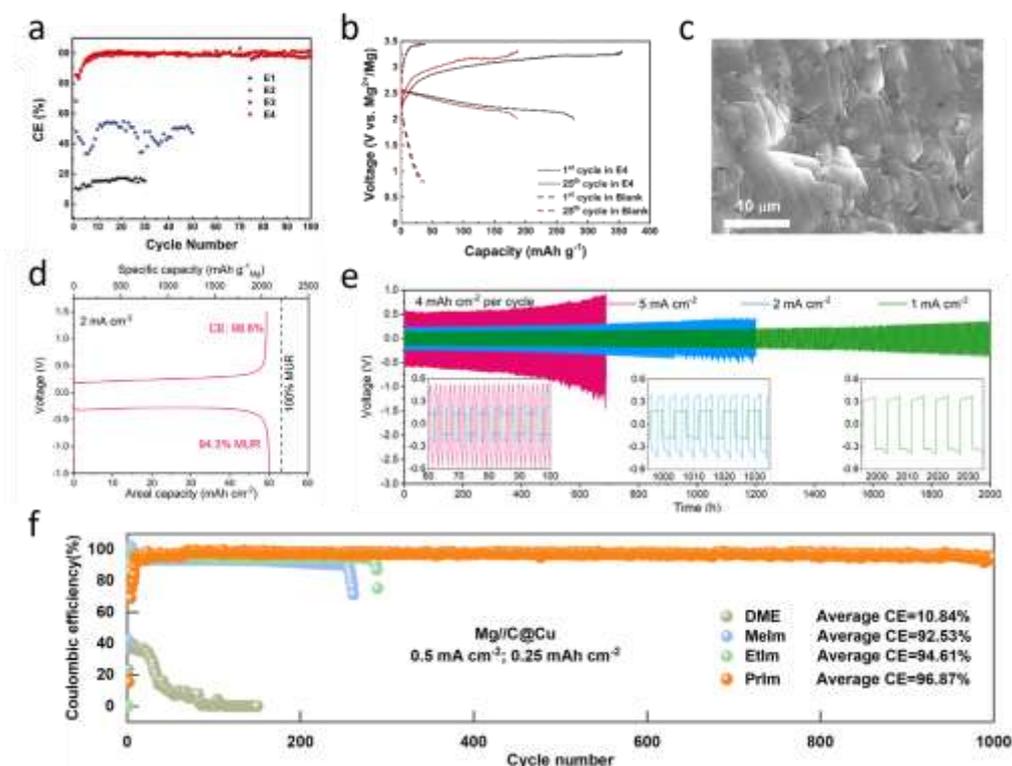


Fig. 2 (a) CEs for Mg plating and stripping in Ex in Mg||SS cells cycled at 0.1 mA cm^{-2} [22]; (b) The charged-discharge curve of the Mg_{0.15}MnO₂||Mg cell in E4 and the blank at 0.5C ($1\text{C} = 200 \text{ mA g}^{-1}$) [22]; (c) SEM images of magnesium deposition in $0.2 \text{ M Mg(OTf)}_2\text{-G2/TEP}$ at 0.5 mA cm^{-2} (0.5 mAh cm^{-2}) [24]; (d) Voltage profiles of a Mg||Cu cell at 2 mA cm^{-2} with cutoff voltages of -1.5 and 1.5 V at plating and stripping, respectively, showing the magnesium utilization rate (MUR) of 94.3% [24]; (e) Long-term stability of magnesium E/S in Mg||Mg cells at various current densities with an areal capacity of 4 mAh cm^{-2} [24]; (f) Comparison of Coulombic efficiencies (CEs) of MeIm, EtIm and PrIm co-solvents-based electrolyte solutions [25]

III. Design Of Novel Anion-Based Electrolytes

Recent mechanistic studies on anion decomposition and solvation structures have consistently demonstrated that constructing bulky weakly coordinating anions (WCAs) with highly delocalized negative charges is a key strategy for achieving wide electrochemical stability windows and non-passivating magnesium electrolytes [26–27]. As shown in Figure 3a [26], electrostatic potential distribution and molecular orbital calculations reveal that among common magnesium salt anions, species such as Ohfip⁻, B(OHFIP)₄⁻, and the cage-like CB₁₁H₁₂⁻ exhibit lower surface negative potentials and higher LUMO/HOMO energy levels. These features effectively weaken the Coulombic interaction between Mg²⁺ and the anion and suppress reductive decomposition, making them promising candidates for chloride-free, high-voltage magnesium electrolytes.

Compared with conventional Mg(TFSI)₂-based systems, halide-free magnesium electrolytes incorporating carborane and fluoroalkoxy borate/aluminate WCAs (e.g., CB₁₁H₁₂⁻, [Al(HFIP)₄]⁻, [B(HFIP)₄]⁻, FPB⁻, and [B(Otfe)₄]⁻) exhibit significant advantages in Mg²⁺-anion interactions, interfacial stability, ionic conductivity, and electrochemical stability window.

In traditional Mg(TFSI)₂ electrolytes, TFSI⁻ readily undergoes reductive decomposition on the Mg surface, forming dense passivation layers such as MgF₂, MgS, and MgO. This leads to high deposition overpotentials and severe stripping polarization. For instance, in Mg(TFSI)₂/DME, the deposition/stripping potentials reach $-0.9 / +2.1 \text{ V}$, with a Coulombic efficiency (CE) of only $\sim 40\%$.

In contrast, WCAs possess large steric volume, highly delocalized charge distribution, and extremely weak coordination with Mg²⁺, rendering them nearly non-reducible at magnesium deposition potentials. As shown in Figure 3b, Mg(CB₁₁H₁₂)₂/DG enables Mg||Mg symmetric cells to operate at 0.5 mA cm^{-2} with deposition/stripping polarization of only $-0.35 / 0.05 \text{ V}$ and a stable CE of $\sim 97\%$ [28]. As shown in Figure 3c,

Mg[Al(HFIP)₄]₂/DME achieves a deposition/stripping CE of 99.3%, reduces deposition overpotential from -0.8 V to -0.35 V, and delivers an ionic conductivity as high as 6.50 mS cm⁻¹ at 0.25 M concentration [29].

In more structurally stable perfluorinated anion systems (Figures 3d and 3e), anodic stability exceeding 4.0 V vs. Mg²⁺/Mg can be achieved. For example, 0.5 M Mg-FPB/DGM exhibits a deposition/stripping overpotential of only 197 mV and a CE of ~95%, while Mg|Mg symmetric cells maintain stable cycling for over 500 h at 0.1 mA cm⁻² [16].

Furthermore, the more cost-effective [B(OTf)₄]⁻ anion also demonstrates favorable reversible magnesium deposition in 0.75 M Mg[B(OTf)₄]₂/THF. As shown in Figure 3f, it exhibits a low deposition overpotential of ~0.2 V and a high CE of 96.7%, while Mo₆S₈ full cells retain a capacity of ~57 mAh g⁻¹ for over 200 cycles (Figure 3g) [30].

Overall, WCA-based electrolyte systems generally enable reversible magnesium deposition/stripping with low polarization (~0.2–0.4 V) and high Coulombic efficiency (>95–99%) in various ether-based solvents, while extending the anodic stability window to above 3–4 V. These electrolytes also exhibit low corrosivity toward practical current collectors such as stainless steel and aluminum, generate fewer interfacial byproducts, and possess enhanced chemical stability.

Therefore, WCA design provides an effective anion engineering principle for developing practical chloride-free, high-voltage magnesium electrolytes. However, the synthesis of these sophisticated anions often involves complex procedures and high costs, which currently limit their large-scale application.

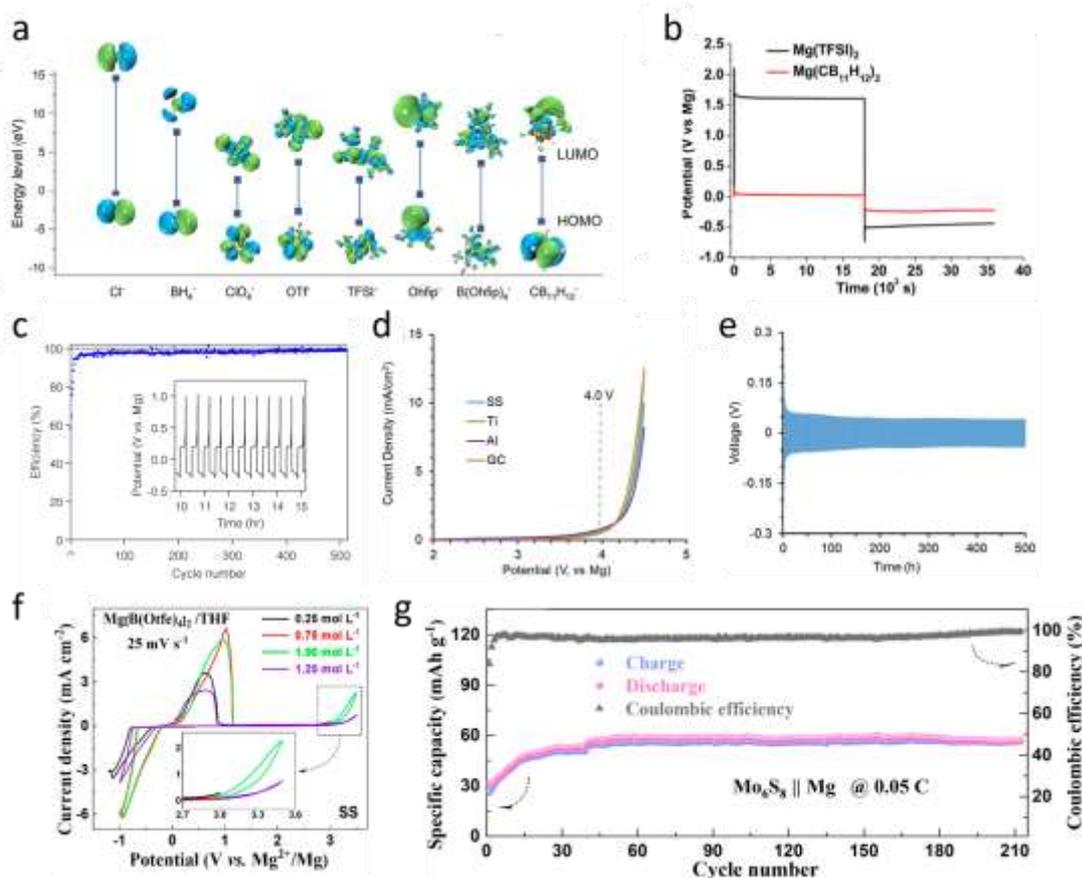


Fig.3 (a) The HOMO and LUMO energies of these anions[26]; (b) chronopotentiometry curves of Mg stripping and plating. The current density is 1 mA cm⁻² and the duration is 5 h for stripping and 5 h for plating at room temperature[28]; (c) Coulombic efficiency as a function of cycle count from a galvanostatic plating and stripping experiment. Inset shows voltage behavior during cycling[29]; (d) Linear sweep voltammetry (LSV) curves of different working electrodes show the anodic stability of the Mg-FPB electrolyte in DGM: stainless steel (SS, blue), Ti (green), Al (purple), and glassy carbon (GC, orange). Condition: Mg as reference and counter electrode, 50 mV · s⁻¹ scan rate [16];(e) Galvanostatic cycling performance of Mg |0.5m Mg-FPB|Mg symmetric cell at 0.1 mAcm⁻² [16]; (f) Cyclic voltammograms of the SS disc electrode conducted via a three-electrode electrolytic cell, the selected

concentration ranges from 0.25 to 1.20 mol L⁻¹[30]; (g) Long-term cycling performance and Coulombic efficiency at 0.05 C[30]

IV. Construction Of Artificial Protective Layers

For rechargeable magnesium batteries, an ideal solid electrolyte interphase (SEI) should possess high electronic insulation and fast Mg²⁺ conductivity. Unlike bare magnesium metal that directly contacts the electrolyte, constructing a Mg²⁺-conductive solid interphase to prevent direct contact between the highly reducing Mg surface and the electrolyte has become a widely adopted strategy for achieving reversible Mg plating/stripping.

Accordingly, various artificial SEI (a-SEI) layers have been developed, including alloy-based [31–32], porous framework-based [33], and polymer-based interphases [20]. These artificial layers enhance Mg²⁺ transport and mitigate side reactions triggered by unstable solvents or anions, thereby improving the interfacial properties of magnesium metal anodes.

The construction of alloy-based SEI layers is often straightforward, typically involving immersion of the magnesium anode in a precursor solution. The group of Zuo Pengjian [32] proposed a simple strategy to overcome the incompatibility between metallic magnesium and several organic electrolytes. By dissolving LiTFSI and AlCl₃ in TEGDME and immersing Mg foil for 24 h, an artificial SEI layer was formed in situ. The resulting a-SEI, rich in MgF₂, MgCl₂, and MgS, effectively suppresses the formation of MgCO₃ and MgO, reduces interfacial resistance and polarization, and enables stable reversible Mg deposition/stripping in various electrolytes. As shown in Figure 4a, Mg||Mg symmetric cells cycled at 0.01 mA cm⁻² operate for over 6000 h with polarization below 200 mV, whereas untreated Mg exhibits polarization exceeding 2000 mV under similar conditions.

Similarly, Cui Guanglei's group [34] formed an artificial protective layer by immersing Mg foil in BiCl₃/DME, generating a composite interphase containing Bi, Mg, Mg₃Bi₂, and MgCl₂. The insulating MgCl₂ component provides resistive properties that suppress excessive Mg²⁺ reduction at the surface and drive uniform Mg deposition beneath the protective layer. As shown in Figures 4b and 4c, Mg||Mg symmetric cells cycled at 1 mA cm⁻² for over 4000 h. The protected Mg electrode not only suppresses parasitic reactions between Mg and Mg(TFSI)₂/DME electrolyte but also mitigates uneven growth during repeated plating/stripping. In Mg–Cu₂-xS full cells, the protected Mg anode delivers significantly improved cycling performance, maintaining ~200 mAh g⁻¹ over 70 cycles at 20 mA g⁻¹.

The Nazar group [35] reported a low-cost inorganic interfacial engineering strategy for high-voltage magnesium batteries. A 3A zeolite/polymer composite film was introduced onto the Mg surface as a protective layer to suppress solvent reduction in halide-free electrolytes. The composite membrane selectively adsorbs free diglyme (G2) molecules while maintaining rapid Mg²⁺ transport along the membrane surface and through the zeolite channels, thereby enabling stable Mg plating/stripping.

As shown in Figure 4d, in Mg(B(OHfip)₄)₂/G2 electrolyte, the protected Mg anode achieves over 6000 h of dendrite-free cycling in symmetric cells with Coulombic efficiency exceeding 98%. Compatibility was further validated in Mo₃S₄ and high-voltage organic PANI full cells. The PANI||Mg full cell operates stably at a cutoff voltage of 3.5 V and high rates, delivering an energy density of ~320 Wh kg⁻¹ and a power density of 1320 W kg⁻¹. This work demonstrates that low-cost molecular sieve-based interfacial engineering is an effective strategy to enhance Mg anode stability without sacrificing voltage or rate capability.

Häcker et al. [20] reported an organic artificial SEI for Mg–S batteries by coating the Mg anode with Aquivion/PVDF, SPEEK/PVDF, or PAN-based polymer layers to suppress polysulfide-induced side reactions at the anode. These protective layers exhibit mechanical flexibility and Mg²⁺ conductivity.

Electrochemical results indicate that although coated Mg anodes show slightly higher polarization in Mg||Mg symmetric cells, they significantly improve the initial Coulombic efficiency and mitigate capacity fading in Mg–S full cells. As shown in Figure 4e, the Aquivion/PVDF-coated Mg anode retains ~310 mAh g⁻¹ after 20 cycles, substantially outperforming bare Mg. In situ optical observations and XPS analysis further confirm that the organic artificial SEI effectively repels polysulfides and alleviates self-discharge.

Overall, polymer/ionomer-based artificial SEI layers represent a viable interfacial engineering strategy to enhance the cycling stability of magnesium–sulfur batteries.

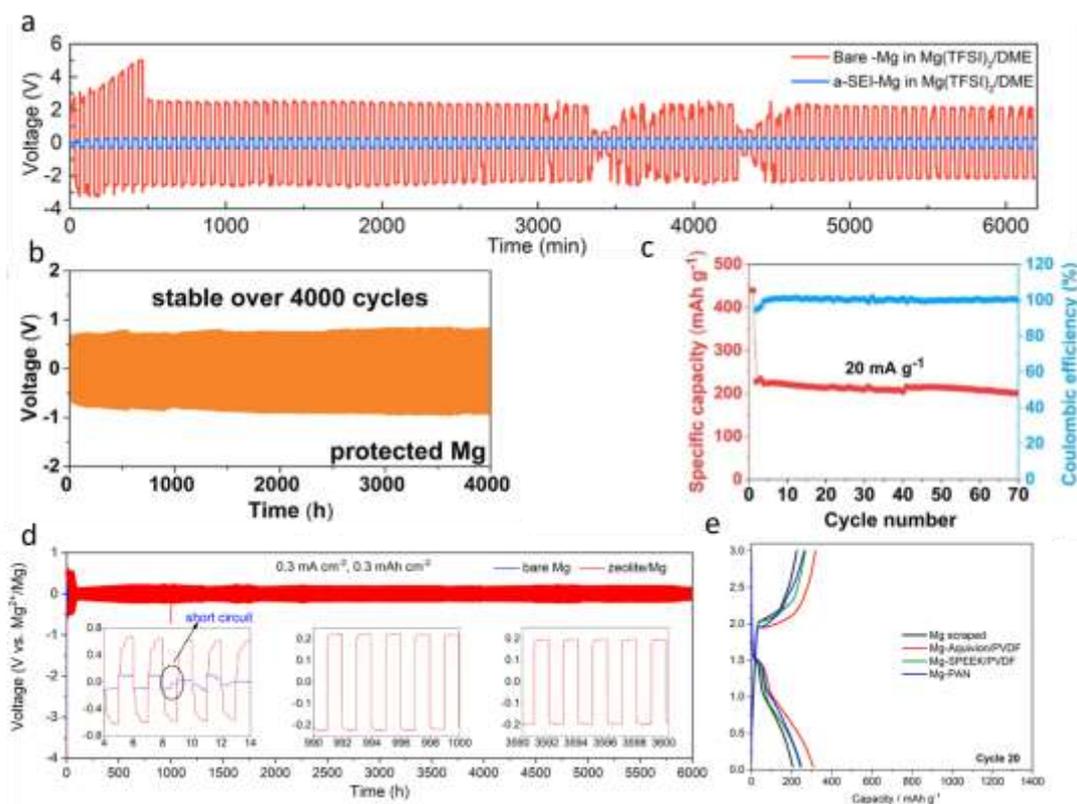


Fig. 4 (a) Voltage responses of bare-Mg/bare-Mg and a-SEI-Mg/a-SEI-Mg cells in Mg(TFSI)₂/DME electrolyte at a current density of 0.01 mA cm⁻²[32]; (b) Cycling performance of symmetric Mg–Mg cells using the pristine Mg with 1 mA cm⁻², 0.5 mAh cm⁻²[34]; (c) cycling performance of the protected Mg-Cu_{2-x}S full batteries at 20 mA g⁻¹[34]; (d) Long-term cycling of Mg||Mg and zeolite/Mg||zeolite/Mg symmetric cells. Cycling conditions are at 0.3 mA cm⁻² with a stripping/plating capacity of 0.3 mAh cm⁻² [35]; (e) Performance comparison of pristine Mg anodes and coated Mg anodes in Mg–S batteries after 20 cycles at C/10 rate[20]

V. Conclusion And Outlook

In summary, this review has focused on the critical bottleneck of electrolyte–magnesium anode interfacial failure in rechargeable magnesium batteries. We systematically summarized the formation mechanisms of interfacial passivation and their influence on magnesium deposition/stripping behavior, and highlighted recent advances in electrolyte engineering and interfacial regulation strategies.

The reductive decomposition of solvents and anions, the high desolvation energy barrier arising from strong Mg²⁺ solvation, and the electronic conductivity of interfacial films are key factors responsible for increased polarization, reduced Coulombic efficiency, and limited cycling stability of magnesium anodes. Therefore, transforming the conventional “passivation layer” into a functional interphase that is highly Mg²⁺-conductive yet electronically insulating is essential for realizing reversible magnesium metal anodes.

At the electrolyte level, the introduction of strong chelating solvents, functional additives, and multicomponent cosolvent systems can effectively reconstruct the Mg²⁺ solvation structure, lower desolvation energy barriers, and induce the formation of compositionally controllable and ion-conductive interphases. Meanwhile, weakly coordinating anions (WCAs), owing to their bulky structure, highly delocalized charge distribution, and resistance to reduction, exhibit systematic advantages in suppressing anion decomposition, widening the electrochemical stability window, and reducing deposition/stripping polarization. These features provide an important design principle for constructing chloride-free, high-voltage magnesium electrolytes.

Artificial SEI construction offers an alternative interfacial engineering approach to enhance magnesium anode stability. By performing alloy-type, porous inorganic framework-type, or polymer-based protective layers on the magnesium surface, it becomes possible to precisely regulate interfacial chemistry without relying on in situ electrolyte decomposition. Such artificial SEI layers effectively isolate the electrolyte from direct contact with metallic magnesium while providing continuous and low-impedance Mg²⁺ transport pathways. Compared with conventional in situ SEI, artificial interphases demonstrate superior stability under high current densities,

large areal capacities, and high-voltage conditions, holding significant promise for the development of high-energy-density magnesium metal batteries.

However, from a practical perspective, several challenges remain. The dynamic evolution of interphases is still insufficiently understood. Both electrolyte-derived in situ SEI and artificially constructed protective layers may undergo dissolution, reconstruction, or compositional migration during prolonged cycling, leading to interphase thickening, hindered ion transport, or electronic leakage. Furthermore, transition metal dissolution from high-voltage cathodes, as well as gas generation associated with additive or SEI precursor decomposition, may continuously disrupt interfacial integrity and deposition morphology, thereby compromising battery safety and lifetime.

Although rechargeable magnesium batteries are not yet ready for large-scale commercialization, continuous advances in weakly coordinating anion chemistry, electrolyte solvation engineering, artificial interphase construction, and advanced characterization techniques are steadily pushing magnesium metal batteries from proof-of-concept toward practical implementation. It is anticipated that, driven by the synergistic integration of theoretical modeling, materials design, and interfacial engineering, magnesium metal batteries—leveraging their intrinsic safety, resource abundance, and cost advantages—will play a significant role in next-generation sustainable energy storage technologies.

References

- [1]. L Y L, Dong H, Aurbach D, et al. Current status and future directions of multivalent metal-ion batteries [J]. *Nature Energy*, 2020, (9) : 646-656.
- [2]. Zhang F Y, B Geng H, Wei W F, et al. Challenges and recent progress in the design of advanced electrode materials for rechargeable Mg batteries [J]. *Energy Storage Mater*, 2019, (20) :118-138.
- [3]. Long J C, Liu Y, He Z, et al. Redesigning Solvation Structure toward Passivation-Free Magnesium Metal Batteries [J]. *ACS Nano*, 2024, (1823) : 15239–15248.
- [4]. Mohtadi R., Tutusaus O, Arthur T. S, et al. The metamorphosis of rechargeable magnesium batteries [J]. *Joule*, 2021,(5) : 581–617.
- [5]. ForeroSaboya J D, Tchitchekova D S, Ohansson P, et al. Interfaces and Interphases in Ca and Mg Batteries [J]. *Advanced Materials Interfaces*, 2021, (8) : 2101578.
- [6]. Fu Q, Wu X Y, Luo X L, et al. High-Voltage Aqueous Mg-Ion Batteries Enabled by Solvation Structure Reorganization [J]. *Advanced Functional Materials*, 2022, (16). 2110674.
- [7]. Yang R, Yao W J, Tang B, et al. Development and challenges of electrode materials for rechargeable Mg batteries [J]. *Energy Storage Mater*, 2021 (42) : 687-704.
- [8]. Wang Li P, Jankowski P, Njel C, et al. Dual Role of Mo6S8 in Polysulfide Conversion and Shuttle for Mg-S Batteries [J]. *Advanced Science*, 2022, (7) : e2104605.
- [9]. Liu F F, Wang T T, Liu X B, et al. Challenges and Recent Progress on Key Materials for Rechargeable Magnesium Batteries [J]. *Advanced Energy Materials*, 2020 (2) : 2000787.
- [10]. Sun Q, Luo S H, Huang R, et al. Recent progress of magnesium electrolytes for rechargeable magnesium batteries [J]. *Coordination Chemistry Reviews*, 2024, (515) : 215956.
- [11]. Song J, Sahadeo E, Noked M, et al. Mapping the Challenges of Magnesium Battery [J]. *Journal Of Physical Chemistry Letters*, 2016, (9) : 1736–1749.
- [12]. Seguin T J, Hahn N T, Zavadil K R, et al. Elucidating Non-aqueous Solvent Stability and Associated Decomposition Mechanisms for Mg Energy Storage Applications From First-Principles [J]. *Frontiers In Chemistry*, 2019, (7) : 175.
- [13]. Li Y B, Cheng M, Liu Q Q, et al. Toward High-Performance Mg/S Batteries with M4-Assisted Mg(AlCl₄)₂/PYR₁₄TFSI/DME Electrolyte and MoS₂@CMK/S Cathode [J]. *Small*, 2024, (20) : 2307396.
- [14]. Xiao J H, Zhang X X, Fan H Y, et al. Cosolvent-Assisted Formation of Charged Ion-Solvent Clusters and Solid Electrolyte Interphase for High-Performance Magnesium Metal Batteries [J]. *Advanced Energy Materials*, 2022, (12) : 2202602.
- [15]. Ren W, Wu D, Nu Li Y N, et al. An Efficient Bulky Mg[B(Otf)₄]₂ Electrolyte and Its Derivatively General Design Strategy for Rechargeable Magnesium Batteries [J]. *ACS Energy Letts*, 2021, (9) : 3212–3220.
- [16]. Luo J, Bi Y J, Zhang L P, et al. A Stable Non-Corrosive Perfluorinated Pinacolatoborate Mg Electrolyte for Rechargeable Mg Batteries [J]. *Angewandte Chemie International Edition*, 2019, (58) : 6967–6971.
- [17]. Dong H, Tutusaus O, Liang Y L, et al. High-power Mg batteries enabled by heterogeneous enolization redox chemistry and weakly coordinating electrolytes [J]. *Nature Energy*, 2020, (5) : 1043–1050.
- [18]. Li, G G, Chen, K Y., Lei, M, et al Construction of Fluoride- Rich Interphase for Sustained Magnesiophilic Site Release Toward High- Stability Chloride- Free Magnesium Metal Batteries[J] *Advanced Energy Materials*, 2024, (14) : 2401507.
- [19]. Wang, Y.Q, Cheng, F L., Huang, Y.Z, et al Vertically-oriented growth of MgMOF layer via heteroepitaxial guidance for highly stable magnesium-metal anode [J].*Energy Storage Materials*, 2023, (61) : 102911.
- [20]. Häcker J, Rommel T, Lange P, et al. Magnesium anode protection by an organic artificial solid electrolyte interphase for magnesium-sulfur batteries [J]. *ACS Applied Materials & Interfaces*, 2023, (15) : 33013-33027
- [21]. Zhao W Y, Pan Z H, Zhang Y J, et al. Tailoring Coordination in Conventional Ether- Based Electrolytes for Reversible Magnesium- Metal Anodes [J] *Angewandte Chemie International Edition*, 2022, (61) : e202205187.
- [22]. Hou S, Ji X, Gaskell K, et al. Solvation sheath reorganization enables divalent metal batteries with fast interfacial charge transfer kinetics [J]. *Science*, 2021, (374) : 172-178.
- [23]. Chinnadurai D, Kumar S, Zhang C, et al. Co- Regulating Planar Mg Deposition and Bromine- Rich Mg Anode- Electrolyte Interface by Multifunctional Organic Bromine [J]. *Additive. Advanced Energy Materials*, 2025,(15) : 2500979.
- [24]. Li C., Guha R D, House S D, et al. A dynamically bare metal interface enables reversible magnesium electrodeposition at 50 mAh cm⁻² [J]. *Joule*, 2025, (9) : 101790.
- [25]. Yang A Q, Gao X, Pei M J, et al. Synergistic Effects of Interfacial Chemistry and Ion- Solvent Interactions to Enable Reversible Magnesium Metal Anode in Chloride- Free Mg(TFSI)₂ Electrolytes [J]. *Angewandte Chemie International Edition*, 2025, (64) : e202424237.
- [26]. Long J C, He Z, Zhang G, et al. Solvation Chemistry of Nonaqueous Electrolytes for Rechargeable Magnesium Batteries [J]. *Advanced Materials*, 2025, (37) : e10488.

- [27]. Riedel S, Wang L P, Fichtner M, et al. Recent advances in electrolytes for magnesium batteries: bridging the gap between chemistry and electrochemistry [J]. *Chemistry–A European Journal*, 2024, (30) : e202402754.
- [28]. Jay R, Tomich A W, Zhang J, et al. Comparative study of Mg (CB₁₁H₁₂)₂ and Mg (TFSI)₂ at the magnesium/electrolyte interface [J]. *ACS Applied Materials & Interfaces*, 2019, (11) : 11414-11420.
- [29]. Herb J T, Nist-Lund G A, Arnold C B. A fluorinated alkoxyaluminate electrolyte for magnesium-ion batteries [J]. *ACS Energy Letters*, 2016, (1) : 1227-1232.
- [30]. Long J C, Liu Y, Zhang W W, et al. Balancing ion diffusion-reduction in chlorine-free electrolytes enables long-life Mg metal batteries [J]. *ACS Energy Letters*, 2024, (9) : 5019-5026.
- [31]. Mandai T, Tanaka U, Watanabe M. Mg–Zn–Cl-integrated functional interface for enhancing the cycle life of Mg electrodes [J]. *Energy Storage Materials*, 2024, (67) : 103302.
- [32]. Li Y Q, Zuo P J, Li R N, et al. Formation of an artificial Mg²⁺-permeable interphase on Mg anodes compatible with ether and carbonate electrolytes [J]. *ACS Applied Materials & Interfaces*, 2021, (13) : 24565-24574.
- [33]. Hu M, Li G Y, Chen K Y, et al. A gradient structured SEI enabling record-high areal capacity anode for high-rate Mg metal batteries [J]. *Chemical Engineering Journal*, 2024, (480): 148193.
- [34]. Zhao Y M, Du A B, Dong S M, et al. A bismuth-based protective layer for magnesium metal anode in noncorrosive electrolytes [J]. *ACS Energy Letters*, 2021, (6) : 2594-2601.
- [35]. Li C, Shyamsunder A, Key B, et al. Stabilizing magnesium plating by a low-cost inorganic surface membrane for high-voltage and high-power Mg batteries [J]. *Joule*, 2023, (7) : 2798-2813.