

Electrodeposition of Metallic Magnesium in Ionic Liquids: A Systematic Review

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Abstract

Metallic magnesium is a strategic material with applications in mobility, energy and medicine, due to its low density, biocompatibility and use as an anode in rechargeable batteries. However, industrial production methods—such as the thermal reduction of dolomite or the electrolysis of anhydrous MgCl_2 —face environmental and operational challenges, including high temperatures, emissions, and dehydration of precursors like bischofite. In response, ionic liquids (ILs) have emerged as alternative electrolytes, offering low volatility, thermal stability and wide electrochemical windows that enable electrodeposition in water-free media. This study presents a systematic review of 32 peer-reviewed articles, applying the PRISMA 2020 methodology. The analysis is structured across three dimensions: (1) types of ILs employed, (2) operational parameters and (3) magnesium source materials. In addition to electrolyte composition, key factors such as temperature, viscosity control, precursor purity and cell architecture were identified as critical for achieving efficient and reproducible magnesium deposition. Furthermore, the use of elevated temperatures and co-solvent strategies has been shown to effectively mitigate viscosity-related transport limitations, enabling more uniform ion mobility and enhancing interfacial behavior. The use of alloy co-deposition strategies and multicomponent electrolyte systems also expands the technological potential of IL-based processes, especially for corrosion-resistant coatings or composite electrode materials. This review contributes by critically synthesizing current techniques, identifying knowledge gaps and proposing strategies for scalable, sustainable magnesium production. The findings position IL-based electrodeposition as a potential alternative for environmentally responsible metal recovery.

Keywords: magnesium; electrodeposition; ionic liquids



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1. Introduction

Magnesium is a widely distributed element in the Earth's crust, with average concentrations reaching approximately 23,300 ppm [1,2]. Its importance as a strategic material has grown considerably in recent years, driven by its potential in both structural and energy-related applications [3]. Among these, the production of metallic magnesium is of particular interest due to its use in aerospace, automotive, electronics, and biomedical sectors. Magnesium and its alloys offer outstanding characteristics—including low density, high strength-to-weight ratio, excellent biocompatibility and biodegradability—making them attractive for a wide range of high-performance applications [1]. One of the most promising emerging uses of metallic magnesium is in energy storage systems, particularly as an anode material in rechargeable batteries. Magnesium exhibits a high volumetric

capacity (3833 mAh cm^{-3}) and a relatively high standard electrode potential (-2.37 V vs. SHE), positioning it as a competitive alternative to lithium in next-generation battery technologies [2,4].

Currently, the most widely used industrial methods for producing metallic magnesium include thermal reduction and electrolysis. In thermal processes, minerals such as dolomite are reduced using ferrosilicon alloys at high temperatures, resulting in significant CO_2 and SO_x emissions. Electrochemical methods typically involve the molten salt electrolysis of anhydrous MgCl_2 at temperatures between 655°C and 660°C [1,5].

Electrochemical methods have lower environmental impacts compared to thermal processes, but they require purified raw materials.

Abundant magnesium raw materials are those obtained from natural brines or obtained as by-products of the extraction of other salts from brines (e.g., lithium and potassium). In brine exploitation, magnesium is notable for its high abundance and persistence throughout the evaporation sequence. It is within this context that a potential resource emerges: bischofite ($\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$)—a white, crystalline, and highly hygroscopic salt—formed through the evaporation of magnesium-rich residual brines [1]. Despite often being treated as a low-value material, bischofite has been identified as a promising raw material for the electrochemical production of metallic magnesium [5].

A major technical challenge in this route is obtaining high-purity, anhydrous magnesium chloride, since the dehydration of bischofite is thermodynamically complex and difficult to control [3,5]. Experimental and theoretical studies have shown that the hydrolysis of $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ is the critical step, with bischofite remaining stable in its hexahydrated form between 25°C and 117°C [1]. This stability is attributed to the distinct energy levels associated with the coordinated water molecules, which exhibit varying bond distances. Figure 1a shows a referential energy profile associated with the progressive dehydration of bischofite, highlighting the increasing energetic demand required to remove each coordinated water molecule. Figure 1b depicts the molecular structure of the salt, highlighting the Mg-O (from water) direct bonds. This molecular configuration makes the stepwise removal of water molecules increasingly energy-intensive, complicating the production of anhydrous MgCl_2 [1,3].

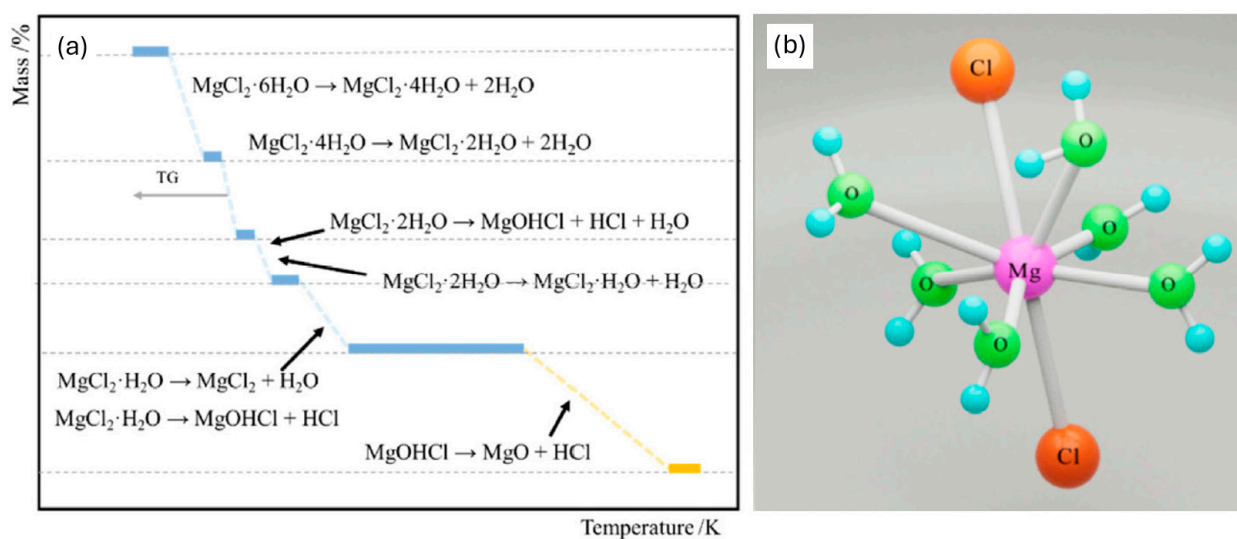


Figure 1. Schematic representations of (a) energy profiles during bischofite dehydration and (b) molecular structure of the salt. Adapted from H. Ming et al. [1].

In recent years, the drive for more sustainable and energy-efficient technologies has spurred interest in the use of ionic liquids (ILs) as alternative electrolytes in electrochemical

processes. These compounds, typically composed of organic cations and various anions, remain liquid below 100 °C, exhibit high thermal and chemical stability and possess negligible vapor pressure [4,6]. Their broad electrochemical stability window and adequate ionic conductivity make them especially suitable for metal electrodeposition, particularly of reactive metals that are difficult or impossible to deposit in conventional aqueous or organic media. Interestingly, beyond their role as electrolytes, some ILs have shown potential to facilitate the selective extraction and stabilization of Mg^{2+} ions, offering a possible advantage in processes where magnesium must be separated from complex brine compositions. This could open up new pathways for simplifying magnesium recovery, especially in systems where conventional solvent extraction or precipitation methods prove inefficient or environmentally burdensome [6,7].

Given the vast number of possible cation–anion combinations in ILs, selecting the most suitable electrolyte [4] and optimizing operational parameters [7] represent key challenges that require in-depth investigation. In response to this knowledge gap in the field of electrodeposition of magnesium using ionic liquid as electrolyte, the present review aims to contribute by systematically reviewing the scientific literature on electrodeposition technologies using ionic liquids for the recovery of metallic magnesium. The review seeks to organize and analyze existing knowledge to address the following research question: What are the recent advances in magnesium electrodeposition technologies using ionic liquids? To this end, three analytical categories are defined: (1) types of ILs employed, (2) key operational parameters and (3) raw materials used as magnesium sources. This approach is intended to provide relevant technical evidence to support the development of more efficient and sustainable magnesium production processes.

The structure of this article is as follows: Section 2 describes the methodology used for the systematic review, with emphasis on the search strategy. Section 3 presents the results, organized according to the analytical categories, and Section 4 discusses the findings, including recommendations to address identified limitations and potential directions for future research.

2. Materials and Methods

The methodology employed in this Systematic Review (SR) is based on the guidelines established by the PRISMA 2020 statement (Preferred Reporting Items for Systematic Reviews and Meta-Analyses), one of the most widely recognized and adopted methodological frameworks at the international level. This guideline provides a rigorous and transparent structure for conducting systematic reviews, ensuring the quality, reproducibility and clarity of the research process. The central purpose of this SR is to systematically and rigorously address the research questions guiding the present study, as outlined below. The present review has not been registered in PRISMA.

General Research Question (GRQ):

- GRQ: What are the recent advances in metallic magnesium electrodeposition technologies using ionic liquids?

Specific Research Questions (SRQs):

- SRQ 1: What types of ionic liquids have been used in the electrodeposition of metallic magnesium, and how do they influence deposit growth?
- SRQ 2: What operational parameters are key to improving the efficiency and quality of metallic magnesium electrodeposition?
- SRQ 3: How does the raw material used in magnesium electrodeposition influence the process, considering its purity and pretreatment?

These questions are aligned with the General Objective (GO) of this research, which is:

- GO: To describe electrodeposition technologies using ionic liquids for the recovery of metallic magnesium through a systematic review of the scientific literature.

The first step in the research process was to identify the specific knowledge domain in which the Systematic Review (SR) is situated. Figure 2 illustrates the intersection of the thematic areas that frame this analysis. This convergence helped define the research focus, design the search algorithm, and establish a robust theoretical framework for addressing the proposed questions.

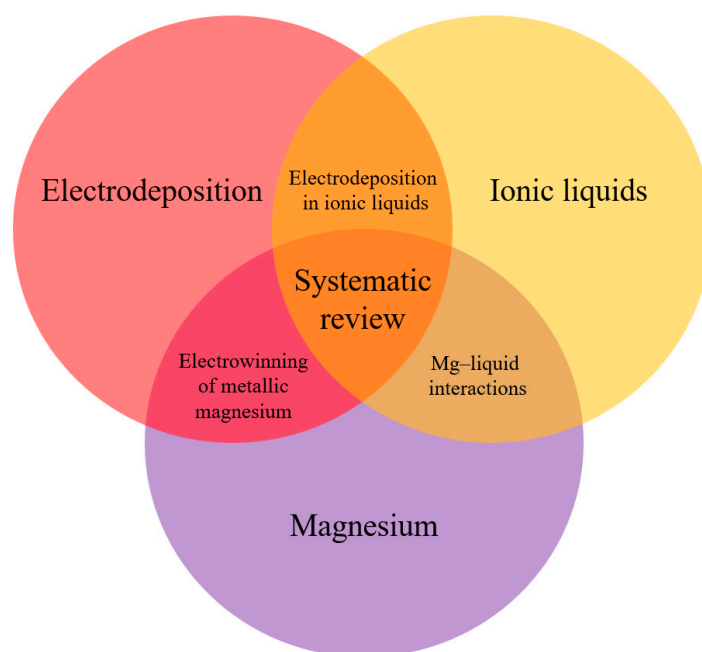


Figure 2. Venn diagram showing the convergence of thematic areas that underpin the systematic review.

The second step involved retrieving relevant scientific documents within the defined knowledge domain. The selection of articles was conducted using the Web of Science (WoS) and Scopus databases, applying the search algorithms presented in Table 1, along with the inclusion and exclusion criteria outlined in Table 2.

Table 1. Search algorithms used.

Database	Search Algorithm
WoS	electrodeposition OR electroplating OR “electrochemical deposition” OR electrodepositing OR “cathodic deposition” AND magnesium OR mg AND Ionic liquids OR ILs OR “deep eutectic solvents”
Scopus	electrodeposition OR electroplating OR “electrochemical deposition” OR electrodepositing OR “cathodic deposition” AND magnesium OR mg AND Ionic liquids OR ILs OR “deep eutectic solvents”

The selected databases were chosen to ensure the rigor and comprehensiveness of the systematic review, given their multidisciplinary scope, broad international coverage and recognition within the scientific community. Scopus, managed by Elsevier, is one of the largest abstract and citation databases worldwide. Its robust indexing system provides access to up-to-date literature and is widely used in systematic reviews due to its strong coverage in the applied sciences, engineering, and technology. Web of Science, administered by Clarivate, offers access to highly selective scientific literature, curated under strict editorial quality standards.

Table 2. Inclusion and exclusion criteria.

Dimension	Inclusion Criteria	Exclusion Criteria	Justification
Magnesium production technology	Electrodeposition OR electroplating OR “electrochemical deposition” OR electrodepositing OR “cathodic deposition”: to identify studies that use electrodeposition as a metal recovery method.	Metal production technologies other than electrodeposition.	Focuses the review on electrochemical methods relevant to the research objective.
Target metal	Magnesium OR Mg: to ensure magnesium is the metal of interest.	Studies focused on metals other than magnesium.	Ensures thematic alignment with the research question.
Electrolyte used	Ionic liquids OR ILs OR “deep eutectic solvents”: to identify the use of ionic liquids as electrolytes in magnesium electrodeposition.	Electrolytes other than ionic liquids.	Targets the specific class of electrolytes under investigation.
Accessibility	All accessible publications are included.	No exclusion criterion.	Maximizes the comprehensiveness of the review.
Language	Publications written in English are limited due to the limited number of studies in other languages.	Publications in languages other than English.	Ensures interpretability and consistency in data extraction.
Resource type	Journal articles and reviews.	All resource types other than journal articles and reviews.	Prioritizes peer-reviewed scientific literature.
Time frame	Publications from 2015 to 2025, to prioritize recent research.	Publications before 2015.	Focuses on the most current developments in the field.

Both databases were selected because they

- Ensure access to peer-reviewed publications;
- Enable replicability and traceability of the search process, in alignment with the transparency standards set by PRISMA 2020;
- Complement each other’s coverage, reduce publication bias and enhance thematic representativeness in the field under study.

The following table outlines the inclusion and exclusion criteria used in the search process. Additionally, it explains the relevance and precision of these dimensions to the research question.

Subsequently, a systematic filtering process was carried out on the collected articles, which included the removal of duplicate records and the evaluation of their relevance based on the thematic scope of this Systematic Review. The procedure began with a preliminary selection based on the article titles, aiming to identify those that directly aligned with the

defined inclusion criteria. This was followed by a detailed analysis of the abstracts, which further refined the selection and ensured the pertinence of the retained studies.

The complete search and selection process is illustrated in Figure 3, which schematically and sequentially outlines each stage of the procedure. It specifies the types of filters applied, the number of documents excluded at each phase, those that progressed to subsequent stages of review and the final set of studies included for in-depth analysis.

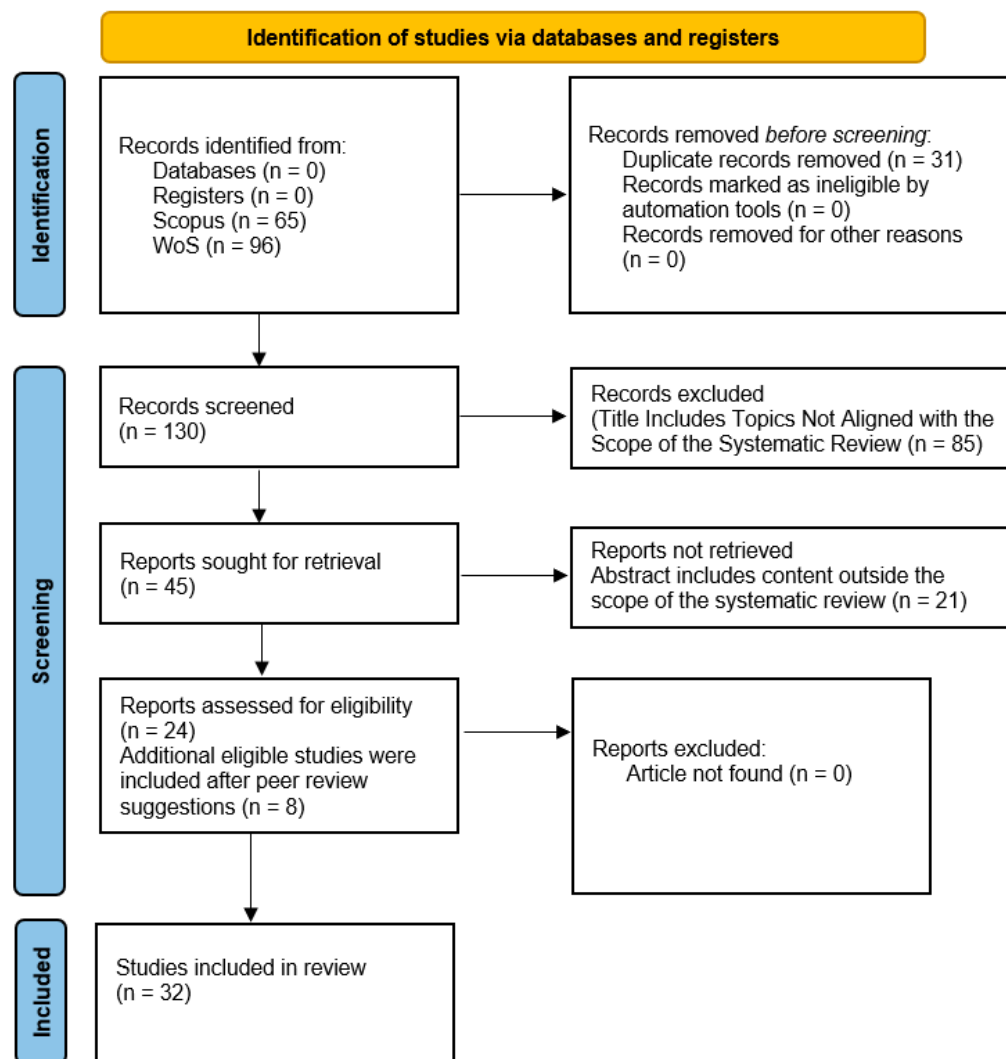


Figure 3. PRISMA 2020 flow diagram.

In addition to the systematic search using the predefined Boolean algorithm, a complementary search assisted by the ChatGPT 5 tool was carried out. This supplementary step enabled the identification of additional relevant articles that had not been retrieved with the original strategy, thereby expanding the final set of included studies. Nevertheless, the total number of selected publications remains limited, which can be explained by the emerging nature of the topic and the relatively small body of scientific literature currently available on this subject.

Based on the detailed analysis of the selected scientific articles, the following analytical categories (AC) and subcategories (SC) have been established to structure the collected information and address the research questions posed in this systematic review.

AC1: Use of Ionic Liquids as Electrolytes: This category addresses the role of ionic liquids in the electrodeposition of metallic magnesium, with particular attention to their ability to facilitate the electrochemical reactions involved. It analyzes how their properties

influence process efficiency and the quality of the resulting metallic deposit. It also examines their specific interactions with magnesium cations and other species present in the medium, considering their potential to minimize undesirable side reactions.

The review covers distinct classes of ionic liquids: conventional ionic liquids (molten salts liquid below 100 °C), functional ionic liquids (with specific functional groups), poly (ionic liquids) (polymers with ionic liquid moieties), and zwitterionic liquids (ZILs, with covalently tethered cations and anions, resulting in neutral molecules with internal charge separation).

Subcategory: Interactions with the ionic liquid

- SC1.1—Interactions with magnesium cations that promote the electrodeposition of metallic magnesium.
- SC1.2—Interactions with supplementary additives present in the medium, with potential to mitigate side reactions that may affect process efficiency.

AC2: Magnesium Electrodeposition: This category provides an in-depth exploration of the methodologies, techniques, tools, and operational parameters used in the recovery of metallic magnesium through electrodeposition processes. It includes the analysis of key variables for optimizing the efficiency and quality of the metallic deposit. It also considers the use of complementary technologies that enable more precise process monitoring and support decision-making.

Subcategory: Operational parameters in magnesium electrodeposition

- SC2.1—Design of the electrochemical cell and optimization of operational parameters to improve the efficiency of metallic magnesium electrodeposition.
- SC2.2—Monitoring and data analysis for decision-making, including sensors, automation and processing of relevant process information.

AC3: Raw Materials Used as Magnesium Source: This category analyzes the various raw materials used as magnesium sources in electrodeposition processes, with particular attention to their chemical composition, purity level and the presence of impurities that may interfere with process efficiency. It also considers the necessary pretreatment steps to ensure optimal conditions for obtaining high-quality metallic deposits.

Subcategory: Considerations regarding the raw material

- SC3.1—Chemical form and purity level, including the presence of impurities that could interfere with the electrodeposition process.
- SC3.2—Required pretreatment steps to optimize their use in the electrodeposition of metallic magnesium.

The following diagram (Figure 4) presents the analytical structure guiding this systematic review. It begins with the main research question, followed by specific sub-questions and concludes with the analytical categories and subcategories used to organize the findings.

This review covers five classes: (i) conventional ionic liquids (molten salts liquid below 100 °C), (ii) functional ionic liquids (with tailored functional groups), (iii) poly (ionic liquids) (polymers bearing IL moieties), (iv) zwitterionic liquids, ZILs (covalently tethered cation–anion within a neutral molecule), and (v) ionic liquid analogs (ILAs), including deep eutectic and chloroaluminate or solvate-type systems formed in situ (e.g., DMI–AlCl₃). Each class presents distinct trade-offs in conductivity, stability, and Mg compatibility. When individual studies assessed multiple IL formulations, we retained only the system with the highest Coulombic efficiency for Mg metal deposition to ensure comparability across the literature.

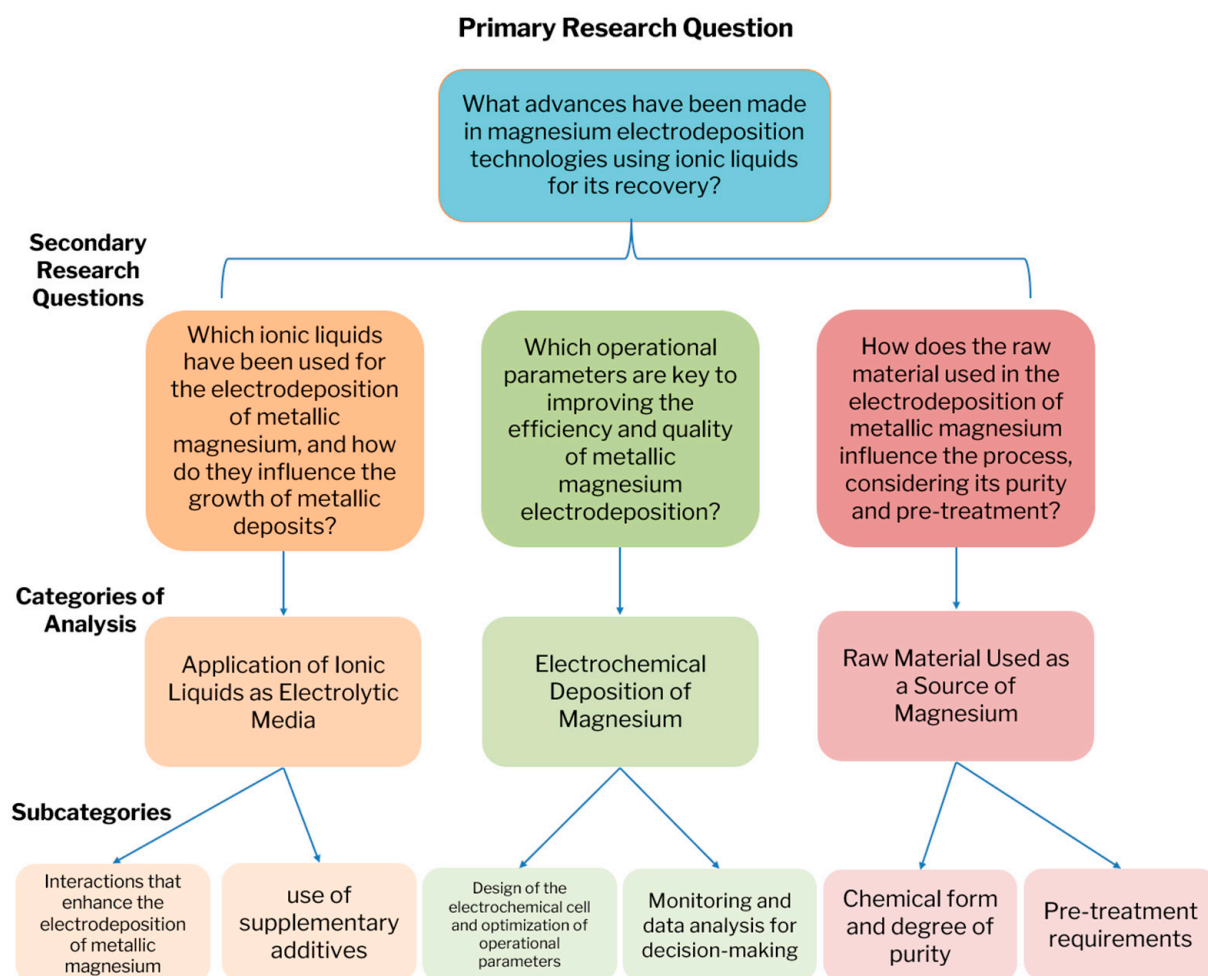


Figure 4. Diagram of analytical categories.

3. Results

The results of this systematic review are presented in three main sections. First, a bibliometric analysis was conducted to characterize the core features of the selected literature, including publication trends, authorship networks and thematic patterns. To support this analysis, metadata were automatically extracted using the DOI of each article via the Cross-Ref REST API. A custom Python 3.12 script was developed to retrieve key fields—such as title, year, authors and keywords—which were then processed using libraries such as pandas, seaborn, WordCloud, and NetworkX for visualization and pattern recognition. This approach enabled a structured, reproducible and visually coherent overview of the scientific landscape surrounding magnesium electrodeposition in ionic liquids.

Second, the findings were organized according to three predefined analytical categories: (1) types of ionic liquids employed, (2) key operational parameters and (3) raw materials used as magnesium sources. Each category was examined in depth to identify technological advances, recurring challenges and knowledge gaps. Finally, a synthesis of the most relevant insights is presented, offering a critical perspective on the current state of the field and outlining future research directions aimed at improving the efficiency and sustainability of magnesium electrodeposition processes.

3.1. Main Features of the Selected Articles

To characterize the scientific landscape surrounding magnesium electrodeposition in ionic liquids, a series of bibliometric visualizations were generated using a custom Python-based workflow. This approach enabled the automated extraction of metadata

from the selected publications, cited from reference number 8 onward, and the creation of targeted visual outputs that highlight key structural and thematic features of the literature. The visualizations include:

- (1) A word cloud generated from article titles to reveal dominant thematic terms (Figure 5).
- (2) A bar plot illustrating annual publication trends to show the temporal evolution of research activity (Figure 6).
- (3) A geographic distribution map based on the lead author's country to identify regional leadership in the field (Figure 7).
- (4) A breakdown of publications by author, highlighting individual research contributions (Figure 8).

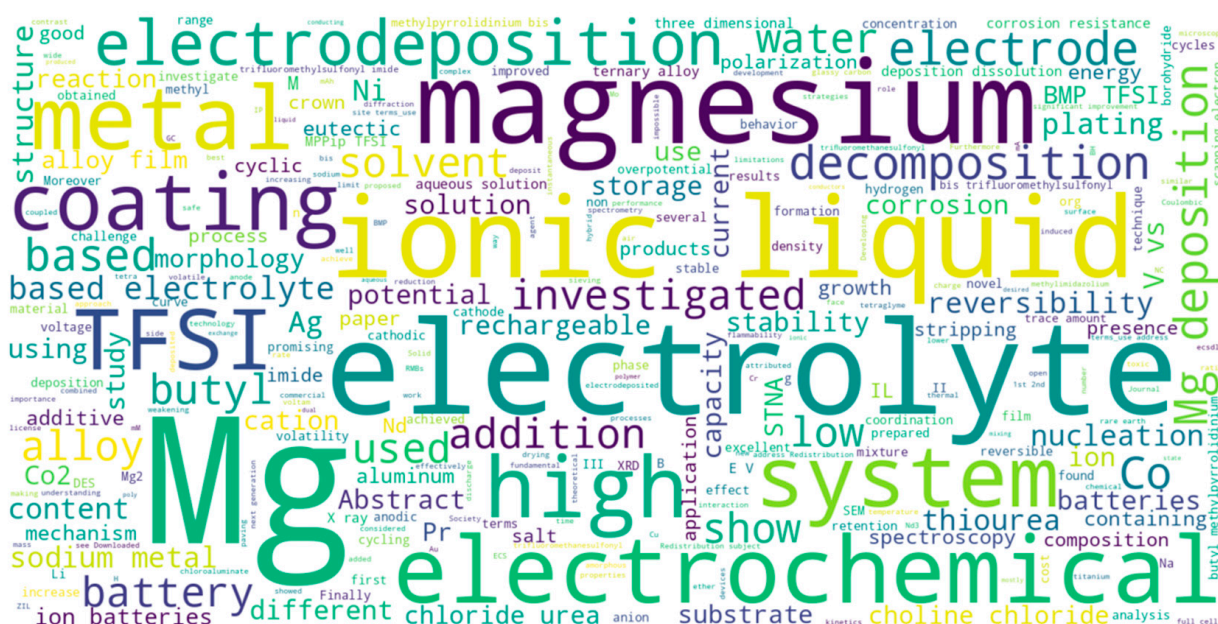


Figure 5. Word cloud generated from the titles of the selected publications.

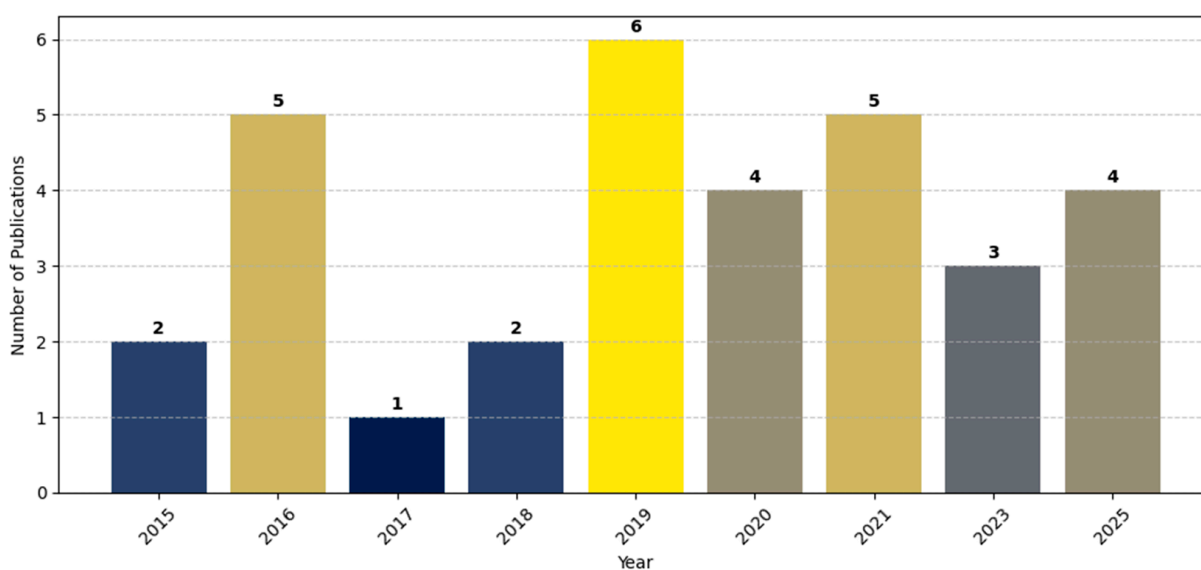


Figure 6. Number of publications by year.

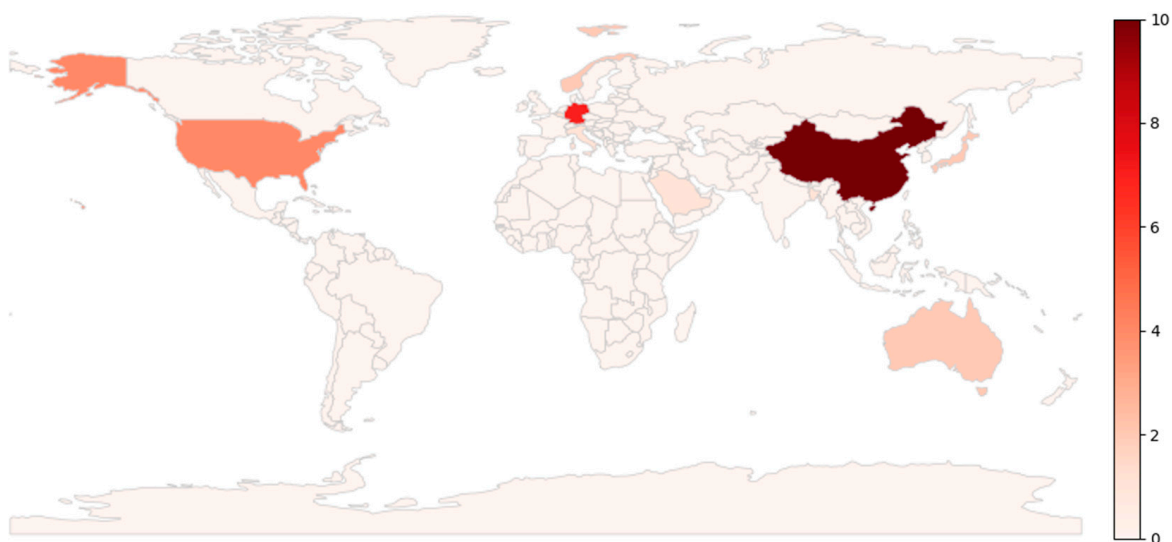


Figure 7. Lead author publications by country.

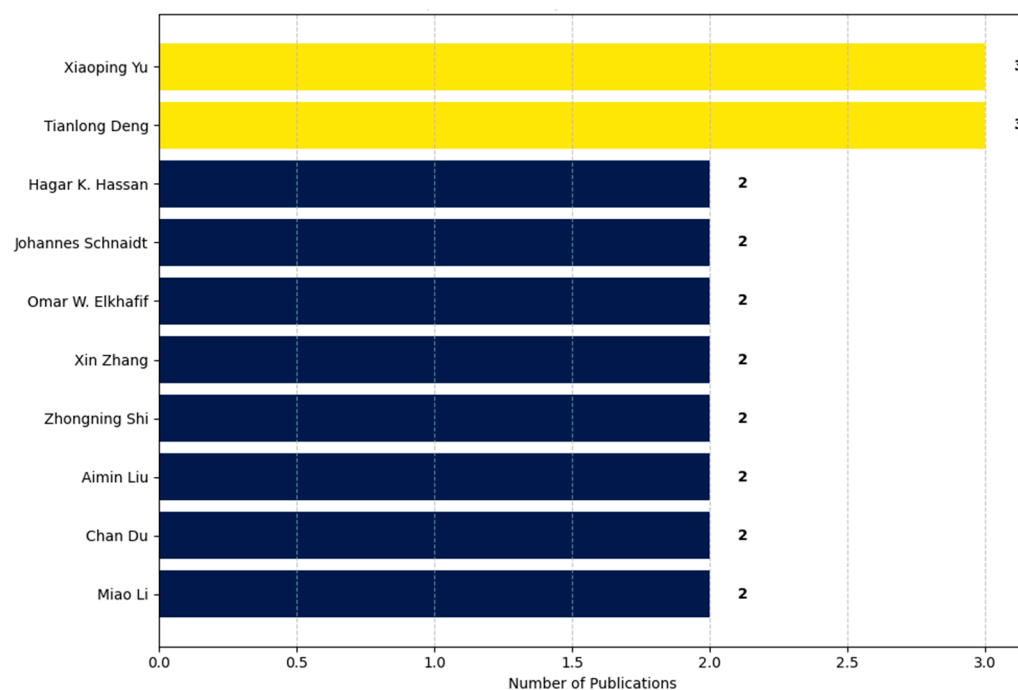


Figure 8. Top 10 most frequent authors.

Each figure was designed to uncover specific dimensions—semantic, temporal, geographic, individual and relational—while maintaining a consistent visual style for clarity and interpretability. The following sections provide a detailed analysis of each figure.

The word cloud in Figure 5 highlights the prevailing research themes within the selected literature. The most prominent terms, such as electrolyte, Mg, magnesium, ionic liquid, and electrochemical, emphasize the central role of electrolyte design and magnesium-based electrodeposition processes. Frequent appearance of words like coating, alloy, decomposition, TFSI, and urea underscores the importance of interfacial reactions, additive effects, and ionic liquid constituents. Other recurrent terms, including system, metal, battery, and stability, point to broader concerns with material performance and energy storage applications. Overall, this visualization provides a concise yet comprehensive overview of the core directions explored in magnesium electrochemistry using ionic liquids.

This bar chart (Figure 6) displays the annual distribution of the selected publications. The peak occurred in 2019, with six papers, followed by 2016 and 2021, each with five publications, and 2020 and 2025, with four publications. Moderate activity is observed in 2023 with three, while years such as 2015 and 2018 contributed only two works. The lowest output corresponds to 2017, with a single publication. Overall, this trend indicates that research on magnesium electrodeposition in ionic liquids has advanced in cycles of heightened activity, with notable surges around 2016, 2019, and 2021.

The choropleth map in Figure 7 shows the geographic distribution of the 32 selected publications by lead-author country. China leads with ten papers, followed by Germany with seven and the United States with four. Australia, Japan, and Norway each contribute two publications, while Bangladesh, Belgium, Italy, Saudi Arabia, and South Korea contribute one each. The white-to-red gradient highlights the concentration of research activity in a small number of countries, with China and Germany standing out as the main hubs.

This bar chart (Figure 8) shows the distribution of publications among the most prolific authors. Two researchers—Xiaoping Yu and Tianlong Deng—stand out with three publications each, reflecting their sustained involvement in the field. A second group, including Hagar K. Hassan, Johannes Schnaidt, Omar W. Elkhaff, Xin Zhang, Zhongning Shi, Aimin Liu, Chan Du, and Miao Li, contributed two papers each. The recurrence of these names suggests the presence of established research teams, particularly in China, that are actively advancing investigations into magnesium electrodeposition in ionic liquids. Overall, the figure highlights both the concentration of expertise in a few leading groups and the wider collaborative nature of the research landscape.

The bibliometric analysis provides a comprehensive view of the research landscape on magnesium electrodeposition in ionic liquids. The word cloud highlights a strong thematic focus on electrolytes, ionic liquids, magnesium, and electrochemical processes, reflecting the centrality of electrolyte design and interfacial mechanisms. The annual distribution of publications shows peaks in 2016, 2019, and 2021, with additional activity in 2025, indicating that research has progressed in distinct waves. Geographically, China leads with 10 publications, followed by Germany with seven and the United States with 4. At the same time, countries such as Australia, Japan, and Norway contributed two studies each. In contrast, Bangladesh, Belgium, Italy, Saudi Arabia, and South Korea contributed only one each, underscoring both a global reach and a concentration of leadership in a few regions. At the author level, most researchers contributed only once, but a small group—including Xiaoping Yu and Tianlong Deng with three publications each, and several others with two—play a central role in advancing the field, particularly within Chinese research teams. Overall, the field appears collaborative but with limited international integration, as leading contributions remain concentrated within specific national groups.

3.2. Findings per Analysis Category

Tables 1–6 are cross-referenced through Row 1, which specifies the citation number corresponding to each article. This shared identifier enables direct correlation across datasets.

3.2.1. Use of Ionic Liquids as Electrolytes

Interactions Mg-ILs

Table 3 presents a structured overview of the ionic liquids (ILs) evaluated across the selected articles. For each IL, the table includes its full chemical name, commonly used abbreviation, reported advantages and documented limitations. This comparative summary serves as a reference point for assessing the performance, suitability and recurring challenges associated with each IL in the context of magnesium electrodeposition.

Table 3. Overview of Ionic Liquids Evaluated in the Selected Studies (operative temperatures in the range 20–80 °C except reference [8], 80–160 °C).

Reference	IL Used	IL Abbreviation	Benefits	Limitations
[9]	1-butyl-3-methylimidazol-2-ylidene borane	BBH ₃ MIm	High reductive stability (−1.66 V), avoids passivation, reversible Mg deposition (~96% CE), and low viscosity	No oxidative stability > 3.56 V; sensitive to trace water; synthesis not trivial
[10]	1-butyl-1-methylpyrrolidinium bis(trifluoromethanesulfonyl)imide	Py _{r14} TFSI	High oxidative stability (~6.0 V); stabilizes electrolyte	Increases overpotential (>±1.6 V); does not form active complexes alone; low MgCl ₂ solubility
[11]	1-methyl-1-propylpiperidinium bis(trifluoromethanesulfonyl)imide	MPPip-TFSI	High oxidative stability (~4.2 V), good conductivity (8.53 mS/cm), and low volatility.	High viscosity (124 mPa·s); Requires co-solvents and additives.
[12]	1-(2-(2-(2-Methoxyethoxy)ethoxy)-ethyl)-1-methylpyrrolidinium Bis(trifluoromethylsulfonyl)imide	MPEG _x Py _r TFSI	High reductive stability; Coulombic Efficiency up to 90%; displaces TFSI [−] and BH ₄ [−] from Mg ²⁺ coordination.	Complex synthesis, moderate viscosity, lower conductivity than ether solvents.
[8]	Poly(1-butyl-3-methylimidazolium styrenesulfonyl(trifluoromethanesulfonyl)imide) Magnesium bis(styrenesulfonyl(trifluoromethanesulfonyl)imide)	BMIPSTFSI + Mg(PSTFSI) ₂	High thermal stability; plasticization improves segmental mobility.	BMI, not Mg ² , dominates transport ⁺ ; Mg deposition is limited or non-uniform.
[13]	1-butyl-1-methylpyrrolidinium bis(trifluoromethanesulfonyl)imide	BMP-TFSI	High thermal stability; well-known structure; enables systematic analysis.	Mg deposition is inhibited by strong interactions between TFSI [−] anions and Mg ²⁺ ; TFSI [−] decomposition is enhanced; low CE.
[14]	Choline chloride + urea (DES)	ChCl:Urea	Low toxicity and a good electrochemical window (~−1.4 V) enable induced co-deposition.	Mg ²⁺ cannot be reduced directly; it requires a catalytic Co ²⁺ pathway.
[15]	Aluminum chloride–1-ethyl-3-methylimidazolium chloride	AlCl ₃ –EMIC	Enables direct Al deposition; supports Al–Mg alloy formation.	Mg does not reduce alone; the incorporation of Mg limits it. solubility constraints.
[16]	1,3-Dimethyl-2-imidazolidinone–aluminum chloride	DMI–AlCl ₃ (solvate ionic system)	Forms electroactive species ([MgCl(DMI) _n] ⁺); dense deposits.	Mg co-deposition is dependent on Al; no independent Mg ²⁺ reduction occurs.

Table 3. Cont.

Reference	IL Used	IL Abbreviation	Benefits	Limitations
[17]	1-butyl-3-methylimidazolium chloride + glycerin (1:1)	BMIC/GL	Non-toxic; good thermal stability; supports Ni–Mg alloy formation.	Mg cannot be reduced alone; conductivity is limited (~0.21 mS/cm).
[18]	Choline chloride + urea (DES)	ChCl:Urea	Enables induced co-deposition of Mg via catalytic Co^{2+} .	Mg^{2+} not reducible independently in DES.
[19]	Choline chloride + urea (DES)	ChCl:Urea	Facilitates Zn plating on Mg alloys; suppresses H_2 evolution.	No Mg deposition reported.
[20]	1-(2-hydroxyethyl)pyridinium TFSI + derivatives	OH-PyrTFSI	Wide electrochemical window; strong coordination features.	No Mg deposition achieved; coordination only studied spectroscopically.
[21]	1-ethyl-3-methylimidazolium tetrafluoroborate	EMImBF ₄	Forms complex anions; enhances Mg^{2+} carrier density; 0.007 S/cm.	Mg not electroplated; coordination/mobility studied only.
[22]	Multiple TFSI-based ILs	Various	High-voltage stability (>5.5 V); good conductivity; improved cathode compatibility.	No Coulombic efficiency is reported for Mg; only the insertion of Mg^{2+} into Cathodes are evaluated.
[23]	N-methyl-N-butylpiperidinium bis(trifluoromethanesulfonyl)imide	[C ₄ mpyr][TFSI]	High conductivity (~2.8 mS/cm); excellent oxidative stability (>3 V); improves thermal properties.	The TFSI [−] anion coordinates strongly with Mg^{2+} , leading to passivation. It generates high overpotentials.
[24]	Magnesium bis(triglyme) bis(bis(trifluoromethanesulfonyl)imide) + Tetrabutylammonium chloride	Mg(G3) ₂ (Tf ₂ N) ₂ + TBACl	CE >87%; reversible stripping; conductivity enhanced with Cl [−]	Poor performance without Cl [−] ; viscosity challenges.
[25]	N-butyl-N-methylpyrrolidinium tris(pentafluoroethyl)trifluorophosphate	PyrrFAP y otros	Mg deposited with CE up to ~85%; enhanced crystallinity.	Most ILs inhibit stripping without Cl [−] or Grignard Species.
[26]	N-methyl-N-propylpiperidinium bis[(trifluoromethyl)sulfonyl]amide	PP ₁₃ TFSI	Low volatility; high oxidative stability (~+4 V); Mg deposition with Coulombic efficiency up to 95%.	Degradation observed with the G3 system; requires glyme coordination.

Table 3. Cont.

Reference	IL Used	IL Abbreviation	Benefits	Limitations
[27]	Methyltrioctylammonium neodecanoate	[A336][V10]	Extracts Mg^{2+} selectively (60.98%); facilitates direct Electrodeposition.	Requires dehydration with MgCl_2 /tamper to prevent $\text{Mg}(\text{OH})_2$ formation.
[28]	Tributylhexylphosphonium bis(trifluoromethanesulfonyl)imide	[P ₄₄₄₆][NTf ₂]	Enhances the conductivity of the extractant system; stable with diluents.	Viscosity increases above ~10% content; no direct reactivity.
[29]	N-allyl-N-methylpyrrolidinium chloride	AMPyrrCl	High CE (~100%); avoids dendrites; prevents passivation.	Low solubility in THF; unstable at excess ratios.
[30]	Methyltrioctylammonium di-(2-ethylhexyl)phosphate	[A336][P204]	High Mg^{2+} extraction rate (~83.99%); good conductivity.	High viscosity at large volumes; conductivity decreases by more than 60% with increased content.
[31]	Choline chloride + urea (0.68:1)	ChCl:Urea (DES)	Enables uniform Mg deposition via pulse technique (~20 nm layer).	Affects deposition morphology in DC mode; sensitive to water/air.
[32]	1-butyl-1-methylpyrrolidinium bis(trifluoromethylsulfonyl)imide	BMP-TFSI	Low volatility, wide electrochemical stability window. Enables Mg plating/stripping when combined with $\text{Mg}(\text{BH}_4)_2$ and 18-crown-6.	Strong Mg^{2+} -TFSI [−] coordination promotes reductive decomposition of TFSI [−] . Without additives, Mg plating/stripping is inefficient.
[33]	1-methyl-1-propylpiperidinium bis[(trifluoromethyl)sulfonyl]imide	MPPip-TFSI	Hydrophobic ILs with wide electrochemical stability windows. Good ionic conductivity, thermal stability, and non-flammability. After drying (<1 mg·L ^{−1} water), improved reversibility of Mg deposition/dissolution and reduced passivation.	Strongly hygroscopic, easily contaminated with trace water. Residual water induces passivation (MgO , $\text{Mg}(\text{OH})_2$), shifts deposition potential by ~0.9 V, lowers conductivity, and reduces stability.
	1-butyl-1-methylpyrrolidinium bis(trifluoromethylsulfonyl)imide	BMP-TFSI		
[34]	N,N-diethyl-N-methyl-N-(2-methoxyethyl)ammonium bis(trifluoromethanesulfonyl)imide	DEME-TFSI	Improves ionic conductivity. Enhances current density and reversibility of Mg deposition/dissolution. Compatible with $\text{Mg}(\text{HMDS})_2$ - MgCl_2 /THF electrolyte, preserves high anodic stability.	At high concentration, minor decomposition (TFSI [−] traces detected in deposits).

Table 3. Cont.

Reference	IL Used	IL Abbreviation	Benefits	Limitations
[35]	1-butyl-3-methylimidazolium tetrafluoroborate	[BMIM][BF ₄]	Supports reversible Mg plating/stripping when combined with Mg(CF ₃ SO ₃) ₂ ; moderate viscosity; less reductive decomposition compared to TFSI [−] systems.	Still viscous; efficiency strongly depends on co-solvent addition; hygroscopic, requires careful drying.
[36]	1-ethyl-3-methylimidazolium tetrachloroaluminate	[EMIM][AlCl ₄] or [C2mim][AlCl ₄]	Enables reversible Mg plating/stripping at room temperature; high anodic stability; Lewis-neutral chloroaluminate species are less corrosive and compatible with high-voltage cathodes.	Requires THF as co-solvent; performance strongly depends on molar ratio (1:2 is optimal).
[37]	N,N,N-tri-(2-(2-methoxyethoxy)ethyl)-N-(2-methoxyethyl)ammonium bis(trifluoromethylsulfonyl)imide	N07TFSI	Noncorrosive compared to Pyr14TFSI (avoids F and S contamination in Mg deposits). Supports reversible Mg plating/stripping with high Coulombic efficiency.	Still exhibits moderate initial Coulombic inefficiency (~70%). Long-term cycling stability needs improvement, as degradation occurs after extended cycling.
[38]	1-methyl-1-propylpiperidinium bis(trifluoromethanesulfonyl) amide	PP13TFSA	High thermal stability, wide electrochemical window, and stable up to 350 °C.	Without ether additives, Mg plating is not reversible; it is prone to TFSA [−] decomposition, leading to impurities (F, N, S) and mossy deposits.
[39]	1,3-Dimethyl-2-imidazolidinone + AlCl ₃ (chloroaluminate ionic liquid analog).	DMI	Forms a chloroaluminate IL analog; stabilizes Mg as [MgCl(DMI) ₄] ⁺ ; enables Mg co-deposition with Al.	Deposits are mixed (Al + Al ₁₂ Mg ₁₇ alloy); efficiency decreases at high MgCl ₂ concentration due to precipitation.

From the compiled table, it can be concluded that the highest efficiencies for metallic magnesium deposition are reported in systems employing ionic liquids specifically designed for plating performance. The zwitterionic IL BBH₃MIm [9] achieved ~96% Coulombic efficiency in simplified systems without co-solvents, while PP₁₃TFSI [26] and [C₄mpyr]TFSI [23] reached up to 95% and 87%, respectively, under hybrid electrolyte configurations. More recently, the borohydride-based system N07TFSI [37] demonstrated the highest efficiency reported so far, with ~98% after initial conditioning, highlighting the effectiveness of stabilizing Mg²⁺ coordination. Furthermore, the ionic liquid analog [A336][V10] [27] enabled direct electrodeposition of metallic magnesium from brine-derived organic phases, offering a distinct low-temperature recovery pathway. Collectively, these systems represent the most efficient formulations under the reported experimental conditions. However, in contrast, many conventional and functional ILs [8,10–22,24,25,28–36,38,39] continue to face challenges such as high viscosity, strong Mg²⁺–anion interactions, or incomplete stripping, which limit their overall efficiency. This suggests that the rational design of IL analogs, functionalized ILs, and zwitterionic systems remains a promising direction to optimize conductivity, stability, and reversible magnesium plating.

Interactions Mg-Electrolyte-Additives

Table 4 compiles explicit formulations tested for the direct electrodeposition of metallic magnesium. It integrates both the final electrolyte composition and the role of functional groups, along with their experimentally reported effects and interactions with Mg²⁺ ions. This dataset provides a mechanistic complement to the IL-focused matrix, highlighting how tailored combinations of ILs and additives influence deposition efficiency, reversibility, and film morphology.

Table 4. Influence of Electrolyte Composition and Functional Additives on Magnesium Electrodeposition (operative temperatures in the range 20–80 °C except reference [8] 80–160 °C).

Reference	Electrolyte Used	Additives	Effect
[9]	0.5 M Mg(BH ₄) ₂ dissolved in BBH ₃ MIm	Mg(BH ₄) ₂	Scavenges trace H ₂ O, preventing oxide formation. BBH ₃ MIm coordinates Mg ²⁺ via a zwitterionic borane site, enabling high-purity non-passivating deposition (CE ~96%).
[10]	0.25 M Mg(TFSI) ₂ + 0.5 M MgCl ₂ in DME, TEGDME or Pyr ₁₄ TFSI	DME	DME coordinates Mg ²⁺ , forming stable [Mg ₃ Cl ₄] ²⁺ ·5DME complexes. Improves ion mobility and lowers overpotential; however, the CE is not directly quantified.
[11]	0.1 M Mg(TFSI) ₂ + 0.01 M Mg(BH ₄) ₂ in MPPip-TFSI:diglyme (1:3)	Mg(BH ₄) ₂ , Diglyme	BH ₄ [−] acts as a drying agent and improves plating morphology. Diglyme enhances conductivity and solvates Mg ²⁺ through ether oxygen interactions. Together they yield CE ~80%–85%.
[12]	Mg(BH ₄) ₂ in MPEG _x PyrTFSI (up to 0.5 M)	Mg(BH ₄) ₂	BH ₄ [−] removes water; MPEG chains coordinate Mg ²⁺ , displacing TFSI [−] from the inner sphere. The chelating effect improves deposition efficiency (~90%) and suppresses passivation.
[8]	IL1/IL2 blend: Mg(PSTFSI) ₂ in BMIPSTFSI (up to 25 mol%)	None reported	PIL system transports Mg ²⁺ via polymer chains, but conductivity is low, and deposits are sparse. Mg ²⁺ mobility is limited; no reliable CE measured.
[13]	BMP-TFSI with Mg(TFSI) ₂	None reported	Mg ²⁺ strongly interacts with TFSI [−] anions, leading to IL decomposition and poor plating (55% CE). No effective coordination or film formation was observed.

Table 4. Cont.

Reference	Electrolyte Used	Additives	Effect
[14]	ChCl:urea DES with CoCl_2 , MgCl_2 , $\text{Nd}(\text{NO}_3)_3$	Thiourea	Promotes induced co-deposition of Mg–Nd via Co^{2+} catalysis. Thiourea adjusts nucleation mode and corrosion resistance. Mg does not deposit alone in the DES system.
[15]	AlCl_3 –EMImCl with MgCl_2	None reported	Mg^{2+} is incorporated via induced co-deposition with Al, forming $\text{Al}_{5.15}\text{Mg}_{3.15}$ alloy. Independent Mg plating not observed. CE ~99% for alloy process.
[16]	DMI– AlCl_3 in solvate with MgCl_2	None reported	Mg is co-deposited via alloy formation with Al. Mg^{2+} does not plate independently; deposition requires induced reduction through Al^{3+} .
[17]	BMIImCl–Glycerin eutectic with NiCl_2 and MgCl_2	None reported	Mg^{2+} is co-deposited with Ni^{2+} . Deposition depends on Ni presence; Mg incorporation confirmed via EDX, but not isolated deposition.
[18]	ChCl–Urea DES with CoCl_2 , MgCl_2 , $\text{Pr}(\text{NO}_3)_3$	None reported	Co^{2+} induces Mg and Pr deposition. Mg^{2+} does not reduce alone; alloy films confirmed by SEM/XRD. Co is catalytic for Mg inclusion.
[19]	Comparative systems using ILs and DES with Mg sources	None reported	Highlights the advantages of ILs over aqueous media for Mg^{2+} plating. Specific electrolyte not defined; no CE quantified.
[20]	Hydroxyl-functionalized pyridinium ILs with Mg^{2+}	None reported	Hydroxyl groups enhance Mg^{2+} solvation and stability; study explores Mg–IL coordination via NMR and FTIR. No plating CE reported.
[21]	EMImBF ₄ mixed with δ - MgCl_2	None reported	BF_4^- and Cl^- form complex anionic structures with Mg^{2+} , increasing conductivity and transport. Deposition behavior not demonstrated.
[22]	Hybrid electrolyte: IL + DPGDME or acetonitrile + $\text{Mg}(\text{TFSI})_2$	DPGDME, Acetonitrile	DPGDME improves Mg^{2+} coordination; acetonitrile boosts conductivity. Used for Mg^{2+} intercalation, not for direct metallic deposition.
[23]	0.3 M $\text{Mg}(\text{TFSI})_2$ in $[\text{C}_4\text{mpyr}][\text{TFSI}]$:tetraglyme (1:2 v/v)	$\text{Mg}(\text{BH}_4)_2$ (~18–19 mM)	BH_4^- removes residual water and suppresses MgO formation; glyme coordinates Mg^{2+} , improving mobility. CE ~87% with stable non-dendritic deposition confirmed by SEM/XRD.
[24]	1:1 molar $\text{Mg}(\text{G}3)_2(\text{Tf}_2\text{N})_2$ with TBACl	TBACl	Cl^- facilitates Mg^{2+} stripping and improves reversibility; the solvate structure enhances plating uniformity and transport. CE >87% observed.
[25]	90% EtMgBr/THF + 10% IL (e.g., $[\text{Py}_{14}][\text{FAP}]$)	None reported	Direct deposition from Grignard reagent; Mg^+ acts as source species, IL stabilizes redox medium and morphology. CE up to ~85%.
[26]	$\text{Mg}(\text{TFSI})_2$ + PP ₁₃ -TFSI + glymes (G2, G3, G4) in molar ratios 1:7:8	None reported	G4 provides multidentate ether coordination, yielding crystalline $\text{Mg}(\text{G}4)(\text{Tf}_2\text{N})_2$ species. Stable plating behavior confirmed via XRD. CE ~95%.
[27]	Organic phase with $[\text{A}336][\text{V}10]$ + MIBK (extractive system)	MIBK	$\text{A}336][\text{V}10]$ enables direct deposition of Mg^{2+} from brine-rich organic phase via coordination. MIBK acts as a phase separation enhancer, increasing conductivity and supporting plating without thermal dehydration. Deposition morphology confirmed by SEM/XRD; CE not quantified.
[28]	Organic phase with 5% $[\text{P}_{4446}][\text{NTf}_2]$ + 55% MIBK + 40% P204 (saponified)	MIBK	$\text{P}_{4446}][\text{NTf}_2]$ enhances conductivity (~1500×), enabling electrodeposition from high Mg/Li ratio brine. MIBK improves phase separation and supports ionic mobility in the organic phase. Mg^{2+} deposition confirmed by XRD; CE not specified.

Table 4. Cont.

Reference	Electrolyte Used	Additives	Effect
[29]	0.4 M PhMgCl + 0–0.4 M AMPyrrCl in THF (Grignard + allyl-IL system)	PhMgCl	Grignard reagent supplies Mg^+ species for direct deposition. Allyl-functionalized IL (AMPyrrCl) is part of the base electrolyte and enhances oxidative stability ($\sim +1$ V), prevents passivation, and improves plating morphology. CE is near 100% in CV tests.
[30]	Organic phase composed of 60% [A336][P204] + 40% MIBK (extracted phase, R = 10:1)	MIBK	MIBK enhances phase separation and ionic conductivity, enabling direct Mg^{2+} deposition from the brine-derived organic phase. Deposition confirmed by XRD; CE not explicitly reported.
[31]	$Mg(CH_3COO)_2$ dissolved in DMSO and mixed with choline chloride:urea DES	DMSO	DMSO enhances the solubility of $Mg(CH_3COO)_2$ and facilitates its transport within the DES matrix. Pulsed deposition yields uniform Mg nanofilms (~ 20 nm) on TiO_2 nanotubes. CE not reported.
[32]	BMP-TFSI + 0.4 M $Mg(BH_4)_2$ + 0.2 M 18-crown-6	$Mg(BH_4)_2$ and 18-crown-6	Crown ether complexes Mg^{2+} , reducing its strong interaction with TFSI $^-$. Borohydride stabilizes Mg^{2+} solvation and improves reductive stability. Together, they significantly enhance the reversibility of Mg plating/stripping.
[33]	MPPip-TFSI/tetraglyme (1:1) + 100 mM $Mg(TFSI)_2$ + 10 mM $Mg(BH_4)_2$	$Mg(BH_4)_2$	Drying decreases solution resistance, increases capacitance, suppresses passivation, and shifts Mg deposition to less negative potentials.
[34]	0.625 M $Mg(HMDS)_2$ – $MgCl_2$ in THF + DEME·TFSI	DEME·TFSI (13.3–53.2 mol%)	Linear increase in ionic conductivity with DEME·TFSI concentration. Reversible Mg deposition/dissolution. Smaller, microporous Mg deposits with higher surface area.
[35]	1 M $Mg(CF_3SO_3)_2$ in [BMIM][BF $_4$] + [PP13][TFSI]	Ether solvents	Addition of ethers decreases viscosity and increases ionic conductivity by 1–2 orders of magnitude, enabling higher reversibility and coulombic efficiency of Mg plating/stripping.
[36]	1 M $[Mg(DIPA)_2]$ + 2 M [C2mim][AlCl $_4$] + 2.5 mL anhydrous THF	THF	A 1:2 ratio shows the lowest overpotential and the highest CE. Overpotential decreased from 375 mV (1st cycle) to 225 mV (100th cycle).
[37]	$[Mg(BH_4)_2]_{0.2}[N07TFSI]_{0.8}$ and $[Mg(BH_4)_2]_{0.3}[N07TFSI]_{0.7}$	$Mg(BH_4)_2$	Displacement of TFSI $^-$ from Mg^{2+} coordination sphere. CE: 70.3% (1st cycle) $\rightarrow$ $\sim 98\%$ from 2nd cycle onward.
[38]	$Mg(TFSA)_2$ + PP13TFSA	18-crown-6 and Diglyme	Improved anti-oxidation capability up to 3.5 V vs. Mg^{2+}/Mg . Thermal stability up to 350 °C in 1:1:5 ratio solutions.
[39]	DMI–AlCl $_3$ (molar ratio 1.6) with 0.2 M $MgCl_2$	$MgCl_2$	Shifts equilibrium from $[Al_2Cl_7]^- \rightarrow [AlCl_4]^-$; generates $[MgCl(DMI)_4]^+$ as electroactive species; supports Mg co-deposition with Al, improving current density.

Among the systems evaluated, five stand out for their superior deposition efficiency, morphological control, and electrochemical stability in direct metallic magnesium plating. The zwitterionic IL BBH_3MIm [9] achieved a Coulombic efficiency of $\sim 96\%$, enabled by selective Mg^{2+} coordination via its borane moiety. In comparison, N07TFSI with $Mg(BH_4)_2$ [37] reached the highest reported value ($\sim 98\%$ after initial conditioning), highlighting the critical role of borohydride additives in moisture removal and passivation suppression. The system based on MPEGxPyrTFSI with $Mg(BH_4)_2$ [12] delivered $\sim 90\%$ efficiency, where polyether side chains displaced passivating anions and stabilized Mg^{2+} transport through multidentate coordination. Similarly, the hybrid electrolyte $[C_4mpyr][TFSI]$:tetraglyme (1:2 v/v) with $Mg(BH_4)_2$ [23] achieved $\sim 87\%$ efficiency by combining ether solvation and water scavenging, yielding smooth, dendrite-free deposits with stable cycling. In parallel, the extractive system [A336][V10] with MIBK [27] enabled direct magnesium deposition

from brine-derived organic phases, with MIBK enhancing conductivity and phase separation. Although Coulombic efficiency was not quantified, SEM and XRD confirmed the formation of metallic Mg without thermal dehydration. Collectively, these optimized formulations demonstrate the synergistic interplay between IL architecture, additive design, and moisture management in enabling high-purity, passivation-free Mg^{2+} electrochemistry.

3.2.2. Magnesium Electrodeposition

Design of the electrochemical cell

Table 5 outlines the cell configurations employed across the studies, focusing on the physical setup, electrode arrangement and operating conditions used for magnesium electrodeposition. It consolidates material choices, reference strategies and thermal parameters, allowing a comparative interpretation of how each design contributes to deposition efficiency in diverse electrolyte systems. This synthesis supports the identification of design patterns linked to high-purity magnesium films.

Table 5. Electrochemical Cell Configurations.

Reference	Cell Design	Anode	Cathode	Reference Electrode	Cell Chamber Separator	Temp. (°C)	Setup Benefits
[9]	Swagelok cell, glass fiber separator, glovebox	Mg metal	Stainless steel 316	Mg metal (pseudo-reference)	Not reported	Ambient	Sealed cell, high reversibility, no passivation
[10]	Coin and flooded PEEK cells; symmetric Mg configuration	Mg metal	Mg metal/Pt (LSV)	Mg metal (pseudo-reference)	Whatman Grade GF/A Binder-Free Glass Microfiber Filter	20–40	Enables comparison of solvents; low overpotential in DME
[11]	BOLA cell (Teflon); Mg symmetric cells	Mg metal	Pt/WS ₂ -PANI	Mg metal (pseudo-reference)	Whatman Grade GF/A Binder-Free Glass Microfiber Filter	25	Compatible with voltammetry and full cells; inert atmosphere (<0.5 ppm H ₂ O/O ₂)
[12]	Three-electrode setup in a 25 mL round-bottom flask	Mg metal	Au	Mg metal (pseudo-reference)	No separator	25	Enables clean galvanostatic deposition on gold; smooth morphology
[8]	Coin cell (2032) with asymmetric configuration	Mg foil	Cu foil with PIL coating	Mg foil (pseudo-reference)	53 μm glass microfiber spacer	80–160	Solid-state-like evaluation; enhanced thermal stability
[13]	DEMS-integrated cell without a membrane	Mg foil	GC/Au	Ag/AgCl	No separator	25	Coupled voltammetry and mass spectrometry; detailed decomposition tracking
[14]	3-electrode cell in a round-bottom flask under argon	Mg foil	Cu	Ag (pseudo-reference)	No separator	80	Induced co-deposition with Co ²⁺ and Nd ³⁺ ; thiourea adjusts nucleation
[15]	Three-compartment Pyrex glass cell	Al metal	Cu/Au	Al metal	No separator	25–70	Enables alloy formation (Al _{5.15} Mg _{3.15}); independent Mg plating not observed
[16]	Three-electrode cell	Al metal	Cu	Al metal	No separator	50	Solvate species [MgCl(DMI) _n] ⁺ formed; dense alloy films
[17]	Three-electrode cell	Graphite	GC	Ag (pseudo-reference)	No separator	80	Ni–Mg alloy formation via induced reduction; 9.6% Mg reported
[18]	Three-electrode flask	Pt/Cu	Cu	Ag (pseudo-reference)	No separator	80	Co ²⁺ induces Mg–Pr deposition; improved adhesion and homogeneity.

Table 5. Cont.

Reference	Cell Design	Anode	Cathode	Reference Electrode	Cell Chamber Separator	Temp. (°C)	Setup Benefits
[19]	Open beaker setup	Pt	Mg-RE alloy (substrate)	None used	No separator	60–90	Zn plating onto Mg alloys; low H ₂ evolution in DES.
[20]	Three-electrode flask in glovebox	Pt	GC	Ag/AgCl	No separator	25	Mg-IL coordination studied spectroscopically; no Mg ⁰ plating.
[21]	Spectroscopic setup	-	-	-	-	20–90	Ionic mobility and coordination evaluated; no metallic Mg deposition
[22]	Three-electrode cell in glovebox	Pt/C	Mg _{0.07} V ₂ O ₅ , Mg-MnO ₂ on Al	Ag/AgNO ₃	No separator	25	Mg ²⁺ intercalation explored; no direct deposition of Mg ⁰
[23]	Three-electrode dry cell	Pt	GC	Scraped Mg foil	No separator	25	~87% CE; dendrite-free Mg deposits; stable cycling over 280 cycles
[24]	Three-electrode cell	Mg foil	Pt/Au	Fc/Fc ⁺	No separator	80	Reversible plating from solvate IL; Cl [−] is critical to performance
[25]	Three-electrode cell	Mg foil	Al/Pt	Mg (pseudo-RE)/Fc/Fc ⁺	No separator	25	Grignard-based system; high Mg ⁰ purity and crystallinity with IL
[26]	Three-electrode cell in glovebox	Mg metal	Pt/Cu	Mg RE (external) + Mg pseudo-RE	No separator	30	Enables coordination studies with G4; deposition verified via crystallography; ~95% CE
[27]	Three-electrode cell; dry N ₂ atmosphere	Graphite	Ag	Mg reference in EtMgBr/THF via frit	No separator	23–25	Direct Mg ⁰ deposition from organic phase; avoids thermal dehydration; SEM/XRD confirmed
[28]	Three-electrode cell; dry N ₂ atmosphere	Graphite	Ag/Pt	Mg RE (external)	No separator	23–25	Saponified IL improves Mg extraction; high conductivity with MIBK; pure deposit confirmed.
[29]	Three-electrode cell in Ar	Mg metal	Ni/Pt/Ag	Mg metal (pseudo-reference)	No separator	25	Allyl-functionalized IL improves stability and morphology; near-100% CE with PhMgCl.
[30]	Three-electrode cell under dry N ₂	Graphite	Ag	Mg RE external in EtMgBr/THF	No separator	23–25	Strong Mg ²⁺ extraction (~83.99%); phase separation and conductivity enhanced by MIBK.
[31]	Two-electrode cell; dry box, ambient RH < 30%	Pt	TiO ₂ nanotubes on Ti foil	None	No separator	50	Pulsed electrolysis yields ~20 nm Mg nanofilm; uniform plating confirmed via HRTEM/XPS.
[32]	Three-electrode glass cell	Mg foil	W microelectrode	Mg wire	No separator, glass chamber	~25	Minimizes current density effects and highlights nucleation/overpotential.
[33]	Three-electrode BOLA-type cell (Kelf, 0.25 cm ³ volume)	Pt foil	Au(111) or Pt sheet (12 mm)	Mg wire	Two glass fiber separators (Whatman GF/B)	~25	Small volume, high stability single-crystal electrodes, reliable EIS/CV characterization.

Table 5. Cont.

Reference	Cell Design	Anode	Cathode	Reference Electrode	Cell Chamber Separator	Temp. (°C)	Setup Benefits
[34]	Coin cell CR2016 and three-electrode Swagelok-type cells	Mg foil	Mo ₆ S ₈ Chevrel phase	Mg wire	Glass fiber separator	Ambient and 55	Elevated T° improves Mg deposition kinetics and cell stability.
[35]	Three-electrode Swagelok-type and coin-cell setups	Mg foil	Mo ₆ S ₈ Chevrel phase	Mg wire	Glass fiber filters	Room temperature and 55	Coin cells enable realistic cycling tests; elevated temperatures reduce viscosity, enhancing Mg kinetics.
[36]	T-type three-electrode cell	Mg disk	Mo, graphite, or SS316	Mg strip	No separator	Ambient	Allowed evaluation of collector corrosion and overpotentials.
[37]	Three-electrode cell	Mg metal	V ₂ O ₅ aerogel	Mg ribbon	Glass fiber	20	Prototype Mg/V ₂ O ₅ cell demonstrated reversible cycling with high capacity.
[38]	Three-electrode cell	Mg plate	Pt plate	Ag wire	Porous glass separates the reference chamber	80	A three-electrode cell allowed evaluation of Mg plating reversibility and additive effects.
[39]	Three-electrode system	Al wire	Pt wire or Cu sheets	Al wire	No separator	~30	Pre-electroplating the electrolyte removes impurities, reducing overpotentials and enhancing current density for Al/Mg deposition.

Among the evaluated systems, several configurations stand out for their ability to couple high Coulombic efficiency with carefully engineered cell designs optimized for magnesium electrodeposition. The Swagelok-type setup [9] exemplifies simplicity and hermetic sealing, using Mg foil as both the counter and pseudo-reference electrode with stainless steel as the working electrode, achieving ~96% CE under ambient conditions without passivation. Likewise, the three-electrode round-bottom flask [12], with Au as the cathode and Mg foil as the pseudo-reference electrode, enabled clean galvanostatic deposition with ~90% efficiency and smooth, dendrite-free morphology. A robust three-electrode dry-cell configuration [23], employing GC as the cathode and a scraped Mg foil as the reference, demonstrated long-term cycling stability with ~87% CE and uniform deposits. Finally, the extractive system [27], based on a three-electrode cell with graphite and Ag electrodes under dry nitrogen, facilitated direct Mg plating from brine-derived organic media without thermal dehydration, as confirmed by SEM and XRD analyses.

Monitoring and data analysis

A wide range of sensor techniques was employed across studies to monitor the magnesium electrodeposition process, characterize the deposited metal layers and evaluate the physicochemical properties of the electrolytes. For electrochemical monitoring, cyclic voltammetry (CV), linear sweep voltammetry (LSV), chronoamperometry, electrochemical impedance spectroscopy (EIS) and galvanostatic cycling were used in [9–14,16,18,19,23–30,32–39], including time-resolved tools such as differential electrochemical mass spectrometry (DEMS) [13], chrono-coulometry [10] and quartz crystal microbalance (EQCM) [11,33]. Metallic deposits were analyzed via scanning and transmission electron microscopy (SEM/TEM), energy-dispersive X-ray spectroscopy (EDS/EDX), X-ray diffraction (XRD) and high-resolution techniques such as HRTEM and XPS [8,9,11,12,14–20,23–31,34–39], offering insights into morphology, composition, grain structure and phase purity. In parallel, ionic liquids and electrolytes were investigated using Raman spectroscopy, NMR (¹H, ¹¹B, ²⁷Al), FTIR, TGA, DSC, ICP-AES, dielectric spectroscopy, refractometry, SAXS/WAXS, calorimetry and high-resolution mass spectrometry

(HRMS) [8–11,13,15,17,20–22,24,25,27–31,36,38,39], enabling structural validation, thermal profiling and coordination analysis.

Automation in magnesium electrodeposition studies commonly relied on potentiostat systems capable of voltage control, pulse programming and real-time data acquisition. These instruments—frequently used in dry environments or gloveboxes [9–13,15–18,20,23–27,29,32–39]. In several cases [8,10,14,18,28–30,34,35,37,38], semi-automated batch routines were implemented to enable repeated cycling or deposition sequences with minimal manual input. A few setups integrated tools such as mass spectrometry or QCM for synchronized monitoring of current profiles [11,13,33]. Overall, these configurations reflect a balance between reproducibility and flexibility in electrolyte screening and process optimization.

3.2.3. Raw Materials Used as Magnesium Source

Chemical form, purity and required pretreatment

Table 6 presents a comparative summary of the magnesium precursor materials used in the reviewed studies, detailing their chemical nature, reported purity, and conditioning procedures before electrodeposition. It distinguishes between common laboratory reagents and brine-derived or extractive sources, highlighting how each approach influences handling protocols, drying strategies and system reproducibility. By consolidating these entries, the table offers a clear overview of how precursor selection intersects with experimental control and electrolyte design in magnesium-based systems.

Table 6. Magnesium Precursor Materials Used in Electrodeposition Systems.

Reference	Mg Source 1	Purity	Mg Source 2 (If Any)	Purity	Purification and Drying
[9]	Mg(BH ₄) ₂	Not specified; highly moisture-sensitive	-	-	Manipulated in glovebox; reacts with water as an in situ drying agent.
[10]	Mg(TFSI) ₂	99.5%	MgCl ₂	99.99%	Both dried at 110 °C for 24 h under vacuum; glovebox handling
[11]	Mg(TFSI) ₂	99.5%	Mg(BH ₄) ₂	95%	Mg(BH ₄) ₂ dried for one week in a glovebox with drying of air over a 4 Å molecular sieve.
[12]	Mg(BH ₄) ₂	Not quantified	-	-	IL dried ≥ 17 h at 80 °C under vacuum; mixed under dry conditions
[8]	Mg(PSTFSI) ₂ (polymeric)	Not specified	MgCl ₂	Unspecified	Gradual drying (RT → 160 °C); cell aged at 160 °C pre-polarization
[13]	Mg(TFSI) ₂	99.5%	Mg foil	Not specified	IL dried at 80 °C for 17 h; glovebox assembly (<25 ppm H ₂ O)
[14]	MgCl ₂	Not specified	-	-	Dried at 80 °C under argon before mixing into DES
[15]	MgCl ₂	99.9%	-	-	Mixed with acidic AlCl ₃ –EMIC IL; used directly
[16]	MgCl ₂	99.9%	-	-	Used under inert gas; no purification reported
[17]	MgCl ₂	99.99%	-	-	Dried 10 h at 120 °C; glyme dehydrated over 4 Å sieve
[18]	MgCl ₂ ·6H ₂ O	Analytical grade	-	-	Dehydrated 22 h at 160 °C under vacuum
[19]	Mg–RE alloys	Not specified	-	-	No precursor salt; Mg is the substrate
[20]	-	-	-	-	No Mg precursor used; coordination studied spectroscopically
[21]	MgCl ₂ (bound polymer)	Verified by TGA/FTIR	-	-	Spectroscopic drying; no electrodeposition

Table 6. Cont.

Reference	Mg Source 1	Purity	Mg Source 2 (If Any)	Purity	Purification and Drying
[22]	Mg(TFSI) ₂	Not specified	-	-	Titrated to <50 ppm H ₂ O; handled in glovebox
[23]	Mg(TFSI) ₂	99.5%	Mg(BH ₄) ₂	95%	Dried 48 h at 80 °C; glovebox manipulation
[24]	Mg(G3) ₂ [Tf ₂ N] ₂ (solvate IL)	Stoichiometric	-	-	Dried under vacuum; TBACl was dried for 48 h at 110 °C
[25]	EtMgBr/THF	1 M (commercial)	-	-	Used under an inert atmosphere; THF dried with sieves
[26]	Mg(TFSI) ₂	High purity, battery grade	-	-	Dried ≥ 24 h at 80 °C; glymes purified beforehand
[27]	Mg ²⁺ from brine	ICP-confirmed	MgCl ₂ (anhydrous)	High purity	Extractive phase dried with MgCl ₂ and a 4 Å sieve
[28]	Mg ²⁺ from natural brine (bischofite)	~76.87 g·L ⁻¹	MgCl ₂	Unspecified	Saponified organic phase; dried with sieves and MgCl ₂
[29]	PhMgCl (Grignard reagent)	2.0 M in THF	-	-	Used as received; handled under inert atmosphere in glovebox
[30]	EtMgBr/THF	1 M or 0.095 M	-	-	THF dried using 4 Å molecular sieves; all components manipulated in a glovebox
[31]	Mg(CH ₃ COO) ₂ (magnesium acetate)	Commercial grade	-	-	Dissolved in DMSO; dried in oven at 60 °C before use
[32]	Mg(BH ₄) ₂	≥95%	Mg foil	99.9%	Electrolytes prepared in an Ar-filled glovebox (O ₂ and H ₂ O < 1 ppm). Solids are dried under vacuum before use. IL used as received
[33]	Mg(TFSI) ₂	99.5%	Mg(BH ₄) ₂	95%	ILs dried with molecular sieves (3–4 Å) at 80 °C, vacuum (10 ⁻³ mbar, 20 h). Tetraglyme dried over 4 Å sieves for 1 week. Electrolytes prepared in glovebox (O ₂ /H ₂ O < 1 ppm).
[34]	Mg(HMDS) ₂ –MgCl ₂	Not specified	Mg metal foil and Mg wire	99.9%	Electrolytes prepared in glovebox (O ₂ , H ₂ O < 0.1 ppm). DEME·TFSI dried under vacuum (80 °C, 48 h). THF is distilled over sodium/benzophenone before use.
[35]	Mg(CF ₃ SO ₃) ₂	Not specified	Mg metal foil	99.9%	ILs dried under vacuum at 80 °C for 48 h. Ether solvents distilled over sodium/benzophenone. Electrolyte preparation inside glovebox (H ₂ O, O ₂ < 0.1 ppm).
[36]	Mg(DIPA) ₂	≥99.9%	Mg disks and strips	>99.9%	Mg foils/disks polished with P800 emery paper and rinsed with dry THF. Electrolytes were stirred for 24 h in an Ar-filled glovebox. THF anhydrous (99.9%), all operations under Ar (O ₂ /H ₂ O < 0.1 ppm).
[37]	Mg(BH ₄) ₂	95%	Mg foil	99.9%	ILs synthesized and extensively purified (anion exchange, filtration, drying under vacuum, glovebox storage). Mg(BH ₄) ₂ used as received.
[38]	Mg(TFSA) ₂	High	Metallic Mg plate	High purity	All reagents used as received; preparation in Ar atmosphere (glovebox); electrochemical measurements under Ar.
[39]	MgCl ₂	≥ 99.99%	-	-	All work under Ar in glovebox (O ₂ , H ₂ O < 0.1 ppm); electrolyte pre-electroplated before experiments to eliminate impurities.

Across the reviewed studies, magnesium precursors were sourced through two principal approaches: commercial reagents and extractive systems derived from natural brines. The majority of works [8–26,29–31] relied on laboratory-grade compounds, such as MgCl₂,

Mg(BH₄)₂, Mg(TFSI)₂, EtMgBr, and PhMgCl, often acquired from suppliers like Aldrich, Solvionic, and Sigma-Aldrich. Purity levels for these salts typically ranged between 95% and 99.99%, and handling protocols were rigorous—most reports emphasized glovebox environments, molecular sieves, and extended vacuum drying at temperatures between 80 °C and 250 °C [10–13,17,23,24,26]. A smaller subset of studies [27,28] focused on magnesium recovery from brine solutions or Salt Lake extracts, sometimes incorporating ionic liquid-assisted separation or saponification to isolate Mg²⁺. While extractive systems offered scalability and reduced reliance on synthetic precursors, they often required additional drying steps using MgCl₂ itself as a desiccant [27,28]. More recent contributions [32–39] introduced stricter purification and handling strategies, including the use of gloveboxes with O₂/H₂O levels below 0.1 ppm, molecular sieves (3–4 Å), sodium/benzophenone distillation of ether solvents, and even pre-electroplating treatments of electrolytes to eliminate residual impurities. In some cases, Mg foils and disks were polished and rinsed with dry solvents before assembly [36], underscoring a higher degree of experimental rigor.

Taken together, the studies demonstrate that a multifactorial interplay between electrolyte composition, additive selection, and electrochemical configuration governs the electrodeposition of metallic magnesium in ionic liquids. Systems integrating borohydrides, polyether chains, or tailored anion–cation pairs exhibited significant improvements in Coulombic efficiency, reversibility, and morphological control, often under ambient conditions. While no universal formulation emerged, certain IL-based electrolytes—particularly those incorporating zwitterionic structures or glyme-coordinated complexes—repeatedly achieved deposition efficiencies exceeding 90%, with smooth, dendrite-free films.

4. Discussion

Regarding ionic liquids as electrolytes, formulations such as BBH₃MIm [9], MPEG_xPyrTFSI [12], and PP₁₃TFSI [26] demonstrated tailored coordination environments that stabilize Mg²⁺ and support non-passivating, reversible deposition. Zwitterionic structures [9] and ILs with polyether functionalities [12,23] proved particularly effective in displacing passivating anions (e.g., TFSI[−]), enhancing the purity and adhesion of metallic films. Nevertheless, high-performance deposition systems relied not only on ILs themselves but also on carefully selected additives and co-solvents. Borohydride species such as Mg(BH₄)₂ acted as drying agents and improved plating morphology by eliminating trace water [9,11,12,23,33,37]. Coordinating agents such as glymes facilitated multidentate bonding with Mg²⁺, thereby increasing ionic mobility and improving Coulombic efficiency [11,23,26]. Some systems also employed chloride-containing additives (e.g., TBACl in [24]) to enhance the reversibility of the stripping process.

TFSI-based ionic liquids are among the most studied families in magnesium electrochemistry, yet their performance varies significantly. In some cases, such as BMP-TFSI [13], strong Mg²⁺–TFSI[−] interactions promoted anion decomposition and poor reversibility, limiting Coulombic efficiencies to ~55%. By contrast, when combined with coordinating glymes, as in [C₄mpyr][TFSI]:tetraglyme [23] or PP₁₃TFSI with G4 [26], glyme molecules displaced TFSI[−] from the Mg²⁺ coordination sphere, increasing ionic mobility and enabling dendrite-free plating with efficiencies up to 95%. These contrasting results demonstrate that the success of TFSI-based ILs depends less on the cation identity and more on the ability of co-solvents or additives to mitigate strong Mg²⁺–anion coordination. Tailoring IL–co-solvent pairs is therefore critical to unlock the potential of this otherwise problematic anion family.

Concerning the influence of temperature on electrolysis performance, no clear trend emerged, as operational conditions varied considerably. Temperature primarily affects electrolyte viscosity, which in turn governs ionic conductivity and electrode resistance.

Several ILs, such as MPPip-TFSI or polymeric PILs [8,11,13], exhibited elevated viscosities under ambient conditions, limiting mass transport and deposition uniformity. To overcome this, studies employed mild heating (typically 50–80 °C) or co-solvents such as diglyme or DMSO to enhance fluidity and coordination [11,23,26,31]. These findings indicate that viscosity control, whether through molecular design or operational adjustments, is essential for achieving stable and efficient Mg deposition. On the other hand, higher temperatures may favor water reduction from hydrophilic IL media, a competing reaction that requires strict moisture management.

A decisive factor emerging across the literature is the rigorous purification and drying of precursors. Many IL-based systems are highly hygroscopic, and even trace amounts of water or oxygen can drastically alter deposition behavior, shifting Mg^{2+} reduction potentials by nearly 1 V and promoting passivation. Most studies therefore relied on glovebox handling and extended drying under vacuum or molecular sieves, typically at 80–250 °C [10–13,17,23,24,26,33–36], while borohydride additives such as $\text{Mg}(\text{BH}_4)_2$ were employed as in situ drying agents [9,11,12,23,33,37]. Recent contributions [34–39] reinforce that reproducibility hinges not only on electrolyte formulation but also on the careful elimination of impurities prior to electrochemical testing, as poorly defined purification protocols make cross-comparison unreliable. Beyond drying itself, residual water exerts a critical influence on interfacial processes: even trace concentrations ($<10 \text{ mg}\cdot\text{L}^{-1}$) can shift deposition potentials, reduce double-layer capacitance, and promote the formation of passivating $\text{MgO}/\text{Mg}(\text{OH})_2$ films [33]. The extent of this effect depends strongly on cation hydrophobicity, with MPPip-TFSI tolerating drying to $<1 \text{ mg}\cdot\text{L}^{-1}$ with restored reversibility, whereas BMP-TFSI retains more water, aggravating passivation and lowering conductivity. These findings confirm that water acts not merely as a contaminant but also as a structural modifier of IL coordination and interfacial chemistry. Effective water management—through both chemical scavengers and rigorous physical drying—is therefore a prerequisite for reproducible and efficient Mg plating.

Cell architecture also influenced performance. Swagelok-type cells under inert atmospheres [9] and sealed three-electrode configurations with minimal separation [12,23] enabled efficient, dendrite-free deposition at room temperature. Prototype setups employing GC or Au substrates under tightly controlled conditions demonstrated reproducible, smooth morphologies and long-term cycling stability [12,23,26]. The possibility of integrating ionic liquid extraction with electrodeposition to recover magnesium directly from brine-derived phases, without thermal dehydration, offers a promising route for sustainable resource utilization, especially in alloy formation or extractive co-deposition schemes [27,28,30].

5. Conclusions

This systematic review consolidates current knowledge on the electrodeposition of metallic magnesium using ionic liquids, a research field that has undergone significant evolution in recent years.

To limit the analysis, the breadth of the search criteria has been sacrificed, losing data significance. However, the objective was to focus the study on obtaining practical conclusions for research into new processes for obtaining metallic magnesium as an alternative to current industrial processes, considering, as a new technological element, the use of ionic solvents.

Despite the technical depth and diversity of the reviewed studies, several limitations must be acknowledged. Approximately one-third of the articles did not report essential quantitative metrics—such as Coulombic efficiency or deposit thickness—hindering cross-comparison between systems [14,18,21,22,28,30]. Additionally, some studies focused

primarily on spectroscopic or coordination analyses without including electrochemical deposition experiments [20,21], limiting their relevance for practical evaluation. Experimental setups also varied widely: while many works employed inert atmospheres with careful control of precursor purity, others provided insufficient detail regarding atmosphere control or cell geometry [19,28], thereby compromising reproducibility. Reporting practices should therefore be harmonized, with clear descriptions of electrode setup, substrate materials, atmospheric controls, and performance metrics [13,18,21,22,28]. Such consistency would facilitate meta-analyses and cross-study benchmarking.

Even if the primary target of this review was metallic magnesium, many studies reported the co-deposition of alloys (Ni–Mg, Al–Mg, Co–Mg), especially in electrolytes where the reduction potential of Mg^{2+} alone was insufficient. For example, co-deposition with Al^{3+} in AlCl_3 –EMIMCl systems yielded Al–Mg alloys [15,16], while Ni^{2+} and Co^{2+} additives in DES or eutectic mixtures enabled Ni–Mg and Co–Mg alloy formation [14,17,18]. Alloying improved film adherence, surface uniformity, and corrosion resistance, and sometimes served as an indirect strategy to incorporate Mg when direct deposition was inefficient. These observations broaden the scope of application, particularly for coating technologies and composite materials.

Moving forward, efforts should focus on developing robust, scalable deposition systems capable of operating under ambient conditions while simultaneously deepening the mechanistic understanding of magnesium ion coordination and interfacial processes.

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Abbreviations

CE	Coulombic efficiency
DES	Deep eutectic solvent
FTIR	Fourier-transform infrared spectroscopy
IL	Ionic liquid
ILs	Ionic liquids
PIL	Poly (ionic liquid)
SEM	Scanning electron microscopy
SR	Systematic Review
XRD	X-ray diffraction

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