

Emerging Developments of Inorganic Magnesium Solid-State Electrolytes

Anik Brinckerhoff¹ and Hong Fang^{1,2,3,4,*}

¹Center for Computational and Integrative Biology, [Rutgers University](#), Camden, New Jersey 08103, USA

²Department of Physics, [Rutgers University-Camden](#), 530 Federal Street, Camden, New Jersey 08102, USA

³Department of Materials Science and Engineering, [Rutgers University-New Brunswick](#), Piscataway, New Jersey 08854, USA

⁴[Rutgers Climate and Energy Institute](#), New Brunswick, New Jersey 08901-2013, USA



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Usable magnesium batteries are promising alternatives to lithium technology. The currently most widely used battery type based on lithium is reaching its cost-reduction limit due to the relative rarity of the precursors and environmental side effects. While other ions, such as sodium, have also been investigated for use in batteries, magnesium stands out due to its several advantages, including high abundance and high theoretical capacity. While most of the current magnesium batteries contain liquid electrolytes, solid-state electrolytes (SSEs) are especially of interest due to their potential advantages over the liquid ones, including enhanced safety, higher energy density, wider electrochemical stability window and operative temperature range, resistance to side reactions and degradation, and simplified battery-pack design. However, Mg-based SSEs have their own difficulties, most notably the lack of ionic conductivity. Only a few Mg-ion conductors can achieve superionic conductivity at room temperature with a low activation energy. Thus, researchers continually search for inorganic solids that can conduct Mg ions better while having no other issues, such as high electronic conductivity or electrochemical instability. With a wide range of materials that have already been investigated and a vast array with potential that remains untapped, it is essential to keep updated on this rapidly expanding field. This minireview aims to identify emerging Mg-ion conductors that can serve as promising candidates for SSEs, as well as to review methods for enhancing the relevant properties of established materials. We highlight the new materials and methods investigated in the last 2–3 years that are yet to be covered in a review. An exciting new area of emerging research is highlighted, utilizing machine learning and high-throughput materials screening and calculations to identify promising inorganic Mg-ion conductors from materials databases.

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I. INTRODUCTION

The lithium battery is currently the most commercially widespread type of battery; however, it has several flaws and disadvantages [1–3]. Magnesium batteries are a promising alternative that has rapidly drawn scientific attention [4,5]. Lithium is relatively rare in the Earth's crust, whereas magnesium is 10^4 times more common [6]. As a result, it is 24 times cheaper than lithium at \$2700/ton [7]. Lithium, unlike magnesium, is also toxic and dangerous to the environment [8]. Furthermore, using

lithium batteries risks freezing, boiling, electrolyte leakage, combustion, and explosion, while magnesium is safer due to its higher melting point and atmospheric stability [9]. Magnesium anodes do not usually form dendrites during plating and stripping, unlike their lithium equivalents [10], although both high current density [11] and the use of certain electrolyte solutions may still cause dendrites to form [12]. The higher first-ionization potential of these batteries compared to lithium-ion batteries increases their stability, which is a boon to both performance and safety [13]. A low redox potential (-2.37 V vs SHE) further draws researchers towards magnesium as a potential alternative to lithium in batteries [14]. While other metals, such as sodium and potassium, are also being studied for these purposes, magnesium has a further advantage because it contains a $+2$ charge instead of $+1$, giving it an especially high specific volumetric capacity of 3833 mAh/cm³ (vs 2046 mAh/cm³ for lithium metal) [15]. All these factors indicate the great

*Contact author: hong.fang@rutgers.edu

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potential to create safer and more efficient batteries based on magnesium.

However, major obstacles remain in the way of developing magnesium batteries, especially if one uses a liquid electrolyte. Carbonate-based liquid electrolytes for magnesium batteries often form a solid-electrolyte interface (SEI) that blocks Mg-ion transport at the Mg metal anode surface. As a result, although carbonate-based electrolytes are standard for lithium-ion batteries, they become incompatible with the Mg anode. Alternative electrolytes, most commonly ethers, are used for Mg batteries instead [16,17]. However, such liquid electrolytes often suffer from corrosivity, difficulty of synthesis, or incompatibility with electrodes [18,19]. Researchers have explored Mg solid-state electrolytes that could potentially avoid SEI formation and side reactions, as well as offer a wider temperature and electrochemical range, charge and discharge more quickly, and achieve higher energy densities and longer cycle lives. However, choosing solid Mg-ion conductors instead of liquid ones as electrolytes leads to a wealth of new considerations, particularly involving the mechanisms of ion transport in solids and the large interfacial impedance or undesired interphases between electrolyte and electrode. One of the greatest challenges in making Mg solid-state electrolytes viable is that they often have high activation energies, much greater than 0.4 eV, and low ionic conductivities, which are typically far from the superionic benchmark of 10^{-4} S/cm at room temperature. The properties of many Mg solid-state electrolyte candidates have been investigated to determine if they can avoid the above-mentioned problems or, if they should fail that test, how they can be altered by methods such as doping to make them more successful. It is, therefore, essential to keep updated on this burgeoning and rapidly expanding field.

Thus, this minireview aims to identify emerging Mg-ion conductors that are promising for solid-state electrolytes and require further characterization, as well as to review potential methods for enhancing the relevant properties (e.g., room-temperature ionic conductivity) of established materials. Several reviews on Mg electrolytes have already covered a broad range of solid-state materials studied up to 2023, including inorganic, metal-organic framework, polymeric, and composite types [4,5]. Building on these comprehensive reviews, we highlight the new inorganic materials investigated over the last 2–3 years that remain to be covered. We discuss the three most common categories of Mg solid-state electrolytes: borohydrides, phosphates, and chalcogenide spinels, as well as promising candidates that fall outside these categories. The new materials are discussed alongside the key Mg-ion conductors in each category, with a critical review of their structure-property relationships and ion-diffusion mechanisms. In addition, this review discusses an emerging area of research that uses machine learning, high-throughput materials screening, and calculations to

identify candidates for Mg solid-state electrolytes from a large materials database.

Before we review each category, it is helpful to bear in mind the requirements for an ideal Mg solid-state electrolyte compared with its lithium counterpart. The ionic conductivity should reach 10^{-4} S/cm or higher at room temperature, with an activation energy below 0.4 eV [1–3,20]. It should exhibit a wide electrochemical stability window of over 4 V and, when coupled with the Mg metal anode, can form chemically and electrochemically stable interphases with high ionic conductivity [1–3,20]. Few of the known solid-state electrolytes can achieve the above, which is at the core of research in the field.

II. BOROHYDRIDES

The potential of borohydrides as Mg solid-state electrolytes was first studied by Matsuo *et al.*, who showed Mg-ion conduction in the high-temperature phase of $\text{Mg}(\text{BH}_4)_2$ using molecular dynamics simulations, based on their experimental findings for Na-ion conductors (i.e., NaBH_4 , NaNH_2 , NaAlH_4 , and Na_3AlH_6) containing BH_4^- , NH_2^- , AlH_4^- , and $[\text{AlH}_6]^{3-}$ polyatomic anions [21]. In the structure of $\text{Mg}(\text{BH}_4)_2$, every fourth BH_4^- formed a relatively small tetrahedral cage that contained Mg^{2+} inside. It was observed that the material would be more ionically conductive if there were a larger space surrounding the Mg ions, as found for the sodium and lithium borohydrides, where the cation resided in a larger octahedral cage [21]. Higashi *et al.* later demonstrated a promising Mg-ion-conducting borohydride, $\text{Mg}(\text{BH}_4)(\text{NH}_2)$, featuring a shorter Mg-Mg neighboring distance and a zigzag Mg sublattice in the channels formed by the BH_4^- and NH_2^- polyatomic anions, as shown in Fig. 1(a) [22]. Whereas $\text{Mg}(\text{BH}_4)_2$ had an ionic conductivity of only 1×10^{-9} S/cm at 150 °C, $\text{Mg}(\text{BH}_4)(\text{NH}_2)$ reached 1×10^{-6} S/cm at the same temperature, an increase of 3 orders of magnitude. It also had an activation energy of 1.31 eV, an electrochemical window of about 3 V, and a band gap of 4.9 eV, implying low electronic conductivity [22]. A cyclic voltammogram obtained with an $\text{Mg}|\text{Mg}(\text{BH}_4)(\text{NH}_2)|\text{Pt}$ cell revealed a cathodic current that matched with Mg deposition, from which Higashi *et al.* concluded that Mg^{2+} was indeed the charge carrier [22]. Ever since, this material has become an archetype borohydride to investigate. In 2019, Le Ruyet *et al.* tested various ball-milling methods for $\text{Mg}(\text{BH}_4)(\text{NH}_2)$. By milling at 400 rpm and then annealing at 120 °C for 72 h under 10 bar H_2 , a way was found to raise the ionic conductivity by 3 orders of magnitude, to 3×10^{-6} S/cm at 100 °C [24], while the activation energy remained high at 1.2–1.3 eV. Using x-ray diffraction and ^{11}B NMR spectroscopy, they found that the likely reason for the improvement in conductivity was that ball milling at higher rpm

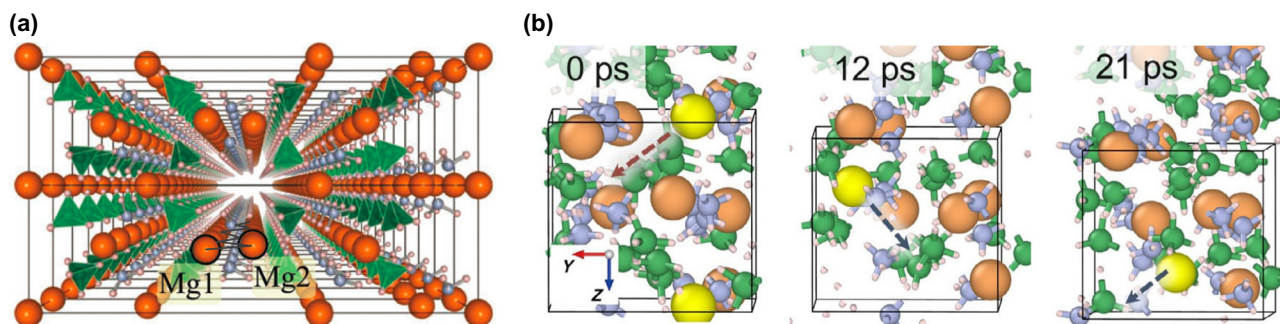


FIG. 1. (a) Ionic conducting structure of Mg(BH₄)(NH₂) featuring the zigzag Mg arrangement between Mg1 and Mg2 in brown, with BH₄ in green tetrahedra, N in light violet, and H in pink. (Reproduced from Ref. [22]. Copyright 2014 The Royal Society of Chemistry.) (b) Mg²⁺ (in yellow) diffusion steps according to the MetaD simulation from 0 to 21 ps, with B in green, N in light blue, and H in pink. (Reproduced from Ref. [23]. Copyright 2025 Wiley-VCH GmbH.)

and annealing for a longer time created an amorphous additional phase, resulting in a glass-ceramic-like composite material with increased ionic conductivity [24].

Other researchers experimented with Mg-ion conductors that contained BH₄⁻ and NH₂⁻ in different proportions. For instance, Kisu *et al.* obtained a high ionic conductivity of 1.3×10^{-5} S/cm at 30 °C from Mg(BH₄)₂(NH₃BH₃)₂ (tested with an Mo electrode); this was among the highest ever recorded at the time, with an activation energy of 1.47 eV [25]. The relatively high activation energy, combined with the high ionic conductivity, suggests that the material has a particularly large preexponential factor in the Arrhenius equation, which is consistent with the “Meyer-Neldel rule” or the “compensation law,” stating that an increasing (decreasing) activation energy often coincides with a higher (lower) preexponential factor [26], as found in various solid-state electrolytes, including the ones containing polyatomic anions (e.g., BH₄⁻, BCl₄⁻) [27–29]. However, this electrolyte had a low Coulombic efficiency of 33% in the first cycle, although this improved to 50% as the cycle number increased [25]. Mg(BH₄)₂(NH₃BH₃)₂ also stood out, exhibiting reversible Mg-ion migration that enabled Mg plating and stripping on the Mo electrode [25]. Wang *et al.* experimented with Mg(BH₄)₂ · *x*NH₃ (with *x* being 0, 1, 2, or 6) with 60 wt % TiO₂ added to increase the number of oxygen vacancies that Mg²⁺ ions could migrate through [30]. They found that only *x* = 1–2 increased the ionic conductivity, with *x* = 1.5 being the most effective, at which the ionic conductor reached a high ionic conductivity of 2.98×10^{-4} S/cm at 298 K (room temperature) with an activation energy of 0.87 eV [30]. It was also found that the Mg ions in this material first broke out of the tetrahedral cages formed by the BH₄⁻ units when a BH₄⁻ anion was captured by an oxygen vacancy and then migrated via oxygen channels between two layers of doped Ti atoms [30]. This is an unusual diffusion mechanism, as both cations and anions move, unlike

in other Mg-ion conductors like sodium superionic conductor (NASICON) types, where only the cation moves. Finally, Pang *et al.* also used Mg(BH₄)₂ · 2NH₃ to build a full cell using Mg|amorphous Mg(BH₄)₂ · 2NH₃|Mo [31]. These recent discoveries have solidified the unique properties of borohydrides, which stand out for their high ionic conductivities without electron conduction.

Another strategy some researchers have applied to borohydrides does not involve nitrogen hydride anions. Instead, these researchers use *closo*-type complex borohydrides (e.g., B₁₂H₁₂²⁻) and hydrate them with water molecules to enhance ionic conductivity. For a long time, it was unclear why hydration improved conductivity, because the low diffusion rate of Mg²⁺ made these systems difficult to study by standard molecular dynamics simulations containing inadequate ion jumps within a limited timescale. To overcome this limitation, dos Santos *et al.* studied these systems using a combination of *ab initio* metadynamics (MetaD) simulations to map the free energy landscape and global optimization based on a genetic algorithm [32]. MetaD can accelerate ionic jumps to overcome barriers in the simulation by adding bias potentials to “flood” local energy minima (e.g., lattice sites), which then enables calculation of the migration barrier height and the activation energy of the system [33,34]. MetaD simulations were conducted for MgB₁₂H₁₂ · *n*H₂O ($2 \leq n \leq 12$) and the lowest activation energies were found with $10 \leq n \leq 12$, which was due to the anion distance being large enough that ions could diffuse and form a distorted hexacoordinated octahedral complex [Mg(H₂O)₆]²⁺ [32]. At 12 water molecules, there is an experimentally determined room-temperature ionic conductivity of 6.527×10^{-5} S/cm with an activation energy of 1.12 eV. By observing which of the variables changed, along with the *n* best correlated to a low activation energy, they were able to identify three ways in which H₂O promoted the ionic conductivity: expansion of the lattice to allow for larger migration pathways, B₁₂H₁₂²⁻ anion cluster oscillation and rotation, and electronic effects

TABLE I. Typical Mg-ion conductors of each type with their available ionic conductivities and activation energies summarized [21–66]. Room temperature is abbreviated as RT.

Mg-ion conductors	Type	Ionic conductivity (S/cm)	Activation energy (eV)
Mg(BH ₄) ₂	Borohydride	1×10^{-9} (150 °C)	>3.0
Mg(BH ₄)(NH ₂)	Borohydride	1×10^{-6} (150 °C)	1.31
Mg(BH ₄)(NH ₂) (ball milled at 400 rpm, then annealed at 120 °C for 72 h under 10 bar H ₂)	Borohydride	3×10^{-6} (100 °C)	1.2–1.3
Mg(BH ₄) ₂ (NH ₃ BH ₃) ₂	Borohydride	1.3×10^{-5} (30 °C)	1.47
Mg(BH ₄) ₂ ·1.5NH ₃ with 60 wt % TiO ₂ added	Borohydride	2.98×10^{-4} (298 K)	0.87
MgB ₁₂ H ₁₂ ·nH ₂ O	Borohydride	6.527×10^{-5}	1.12
MgAl ₂ O ₄	Chalcogenide	2×10^{-8} to 2×10^{-7} (580–650 °C)	1.9
MgSc ₂ Se ₄	Chalcogenide	$2.4\text{--}5.5 \times 10^{-5}$ (RT)	0.375
MgY ₂ Se ₄	Chalcogenide	2.0×10^{-5}	0.360
MgTm ₂ Se ₄	Chalcogenide	6.5×10^{-5}	0.378
MgEr ₂ Se ₄	Chalcogenide	4.1×10^{-5}	0.375
MgSc ₂ Se ₄ (5 wt % Se in excess)	Chalcogenide	7.2×10^{-5}	—
MgSc _{1.9} Ti _{0.075} Se ₄	Chalcogenide	2.8×10^{-7}	—
MgSc _{1.8} Ti _{0.15} Se ₄	Chalcogenide	1.6×10^{-5}	—
MgYb ₂ Se ₄	Chalcogenide	$2\text{--}7 \times 10^{-5}$ (25 °C)	0.364
Mg _{0.5} Zr ₂ (PO ₄) ₃	Phosphate	1.6×10^{-8} (25 °C)	0.824
Mg _{0.7} (Zr _{0.85} Nb _{0.15}) ₄ (PO ₄) ₆	Phosphate	6.89×10^{-3} (800 °C)	1.41
Mg _{1.05} Zn _{0.4} Al _{0.3} Zr _{1.3} (PO ₄) ₃	Phosphate	3.97×10^{-4} (RT)	0.039
Mg _{0.5} Ce _{0.2} Zr _{1.8} (PO ₄) ₃	Phosphate	2×10^{-10} (25 °C)	0.307 (≥ 60 °C)
Mg _{0.6} Fe _{0.2} Zr _{1.8} (PO ₄) ₃	Phosphate	1.8×10^{-7} (25 °C)	0.141 (< 175 °C)
Mg _{0.5} Ti ₂ (PO ₄) ₃	Phosphate	$\sim 10^{-10}$	0.70
Mg _{0.5} Sn ₂ (PO ₄) ₃ 20 wt % 1M magnesium bis[(trifluoromethyl)sulfonyl]imide [Mg(TFSI) ₂] in 1-ethyl-3-methylimidazolium bis[(trifluoromethyl)sulfonyl]imide ([EMIM][TFSI])	Phosphate	1.11×10^{-4}	0.36
MgHf(WO ₄) ₃	Oxide	2.5×10^{-4} (600 °C)	0.835
0.4MgSO ₄ ·0.6Al ₂ O ₃	Oxide	1.9×10^{-6}	—
0.6β-MgSO ₄ :Mg(NO ₃) _{1.6} MgO	Oxide	$\sim 10^{-6}$ (RT)	—
0.9Mg(NO ₃) ₂ ·0.1Al ₂ O ₃	Oxide	4.0×10^{-4} (RT)	—
0.1MgCO ₃ ·0.9Al ₂ O ₃	Oxide	—	—
1.74×10^{-4} (303 K)	—	—	—
MgPON	Oxide	1.2×10^{-6} (500 °C)	1.60
Mg _{0.6} Al _{1.2} Si _{1.8} O ₆	Oxide	1.6×10^{-6} (500 °C)	1.32
Mg _{0.25} Cd _{0.75} PS ₃ ·1.9H ₂ O	Sulfide	4.81×10^{-5} (RT)	0.599
Mg _{0.2} Mn _{0.8} PS ₃ ·1.8H ₂ O	Sulfide	1.06×10^{-4} (RT)	0.557
MgAl ₂ Cl ₂ Br ₆	Halide	1.3×10^{-6} (400 K)	0.523
MgCl ₂ -GaF ₃	Halide	4.7×10^{-4}	0.406

caused by the hydration process. Building on their work, Wang *et al.* employed *ab initio* MetaD simulations, along with a large language model, to analyze many Mg borohydrides and found that the MetaD approach was effective at high-throughput characterizations of ion conductors [23]. Moreover, this approach can help to unravel novel Mg-ion diffusion mechanisms, as demonstrated in Fig. 1(b), where a complex Mg-ion migration process in Mg(BH₄)₂·2NH₃ is elucidated, involving a Mg²⁺ ion moving out of its tetrahedral arrangement towards the NH₃ cluster and then migrating through the interstitial sites that are alternately coordinating with BH₄[−] anion clusters [23]. Typical Mg-ion-conducting borohydrides and their key properties are summarized in Table I.

III. CHALCOGENIDE SPINELS

Another category of Mg-ion conductors that has recently gained much attention are chalcogenide spinels. A spinel has a general formula of AB_2X_4 , where A is a tetrahedrally coordinated cation; B is an octahedrally coordinated cation; and X is an anion, often a chalcogenide. This structure is appealing due to the large distance between the two cation types, which allows Mg ions to move within the three-dimensional channels, as shown in Fig. 2(a). Researchers often seek a spinel structure in which Mg serves as the tetrahedrally coordinated A cation. This is because Mg²⁺ favors octahedral coordination and being in a less-favored tetrahedrally coordinated state makes it

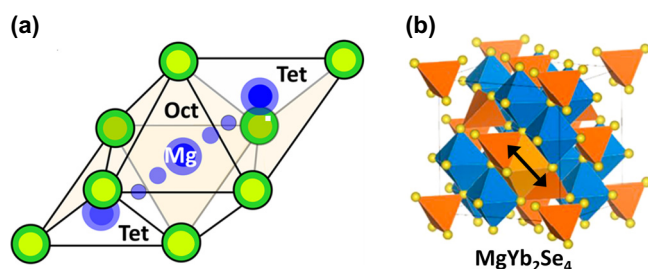


FIG. 2. (a) Crystal structure of a spinel with the chalcogenide anion in green circle and the Mg^{2+} in blue circle migrating between two tetrahedral (Tet) sites through an octahedral (Oct) site. (Reproduced from Ref. [35]. Copyright 2017 Nature Portfolio.) (b) Structure of MgYb_2Se_4 spinel exhibiting room-temperature ionic conductivity exceeding 10^{-4} S/cm, with Mg in orange, Yb in blue, and Se in yellow. Black arrow shows the Tet-Oct-Tet pathway of Mg^{2+} in the structure. (Reproduced from Ref. [42]. Copyright 2025 American Chemical Society.)

easier for Mg ions to migrate [35]. For a similar reason, selenium is preferred over sulfur for the anion, which is, in turn, preferred over oxygen; an increased volume per anion increases the polarizability, making it easier for Mg ions to migrate [35].

One early investigation of chalcogenide spinels was in Lee's 2007 thesis, which proved Mg-ion conduction in MgAl_2O_4 , while building three cells containing this compound: $\text{Al}|\text{MgAl}_2\text{O}_4|\text{Mg}$, $\text{Al}|\text{MgAl}_2\text{O}_4|\text{C}$, and $\text{Mg}|\text{MgAl}_2\text{O}_4|\text{C}$ [36]. However, the ionic conductivity of the material was low, ranging from 2×10^{-8} to 10^{-7} S/cm even at high temperatures of 580–650 °C, with a high activation energy of 1.9 eV [36]. Later, Canepa *et al.* investigated MgSc_2Se_4 , estimating its ionic conductivities from DFT-based nudged elastic band (NEB) calculations and *ab initio* molecular dynamics simulations to be 1×10^{-5} S/cm and 8.9×10^{-6} S/cm at 298 K (room temperature), respectively, with an activation energy of 0.375 eV [35]. They further found, through NEB calculations, that MgY_2Se_4 had an activation energy of 0.361 eV and MgY_2S_4 had an activation energy of 0.360 eV. While the obtained ionic conductivity was promising, they also discovered a problem that would plague chalcogenide Mg-ion conductors to this day—high electronic conductivities, as in the case of MgSc_2Se_4 , where the electronic conductivity was about 0.04% of the ionic conductivity [35]. This was despite a band gap of 2.15 eV, as determined by HSE06 calculations, which would normally suggest low electronic conductivity. Canepa *et al.* theorized that the high electronic conductivity observed in these materials was nonetheless due to either defects or secondary phases [35]. In 2023, Glaser *et al.* reevaluated the ionic conductivity of MgSc_2Se_4 because they believed Canepa *et al.* had insufficiently analyzed the impedance data; they would get an even higher ionic conductivity of $2.4\text{--}5.5 \times 10^{-5}$ S/cm at room temperature [37].

Canepa *et al.* conducted computational studies to evaluate what the most common defects were in MgSc_2S_4 , MgSc_2Se_4 , and MgIn_2S_4 , and how these defects contributed to the electronic conductivity [38]. They found that anion-poor synthesis, compared to anion-rich synthesis, led to higher electronic conductivity, due to the formation of vacancies and Mg-metal antisites (in which the other cation substituted for magnesium in the tetrahedral site), and due to there being more *n*-type than *p*-type conductivity under the anion-poor conditions [38]. Canepa *et al.* also noted that rapidly cooling the spinels after formation would “freeze in” the population of defects found at higher temperatures, and that this population bestowed a higher electronic conductivity than what was found in slower-cooling spinels. Finally, they investigated aliovalent doping as a strategy to reduce electronic conductivity while also increasing ionic conductivity [38]. Specifically, doping Sc in MgSc_2Se_4 with metals such as Bi, Nb, Ta, Ge, Sn, Pb, and Sc would not be practical due to the defect formation energy being too high, but Ce, Ti, and Zr doping proved promising, as well as halogen doping of Se [38]. Wang *et al.* carried out some of these suggestions experimentally, both synthesizing MgSc_2Se_4 with Se-rich phases and doping with Ti^{4+} and Ce^{4+} in a cell with gold electrodes, but none of these interventions substantially decreased the electronic conductivity of the spinel [39]. Kundu *et al.* later theorized that the electronic conductivity in MgSc_2Se_4 was due to a Berthelot-type conductivity mechanism, where there were nanoscale conductive free-electron-containing “pockets” within an overall low-conductivity matrix, and electrons travelled between these pockets through quantum tunneling [40]. Based on this theory, they suggested that the precursor phase of the spinel should not be ball-milled to avoid forming conductive phases of Sc/ScSe near the synthesized particle surface [40].

In 2024, Glaser *et al.* further studied other selenide spinels for their ionic and electronic conductivities [41]. They found that MgTm_2Se_4 had an ionic conductivity of 6.5×10^{-5} S/cm with an activation energy of 0.378 eV and an electronic conductivity of 3.0×10^{-7} S/cm, MgEr_2Se_4 had an ionic conductivity of 4.1×10^{-5} S/cm with an activation energy of 0.375 eV and an electronic conductivity of 2.7×10^{-6} S/cm, and MgY_2Se_4 had an ionic conductivity of 2.0×10^{-5} S/cm with an activation energy of 0.360 eV and an electronic conductivity of 4.7×10^{-6} S/cm, all at 25 °C (room temperature) [41]. Mg ions migrate in the spinel structure from initial tetrahedral sites through an empty octahedron to final tetrahedral sites [Tet-Oct-Tet, Fig. 2(a)]. The migration barrier is related to the energy difference between the tetrahedral and octahedral sites. Glaser *et al.* noted that this relationship was strengthened as the ionic radius of the non-Mg metal increased [41]. In 2025, Glaser *et al.* followed this up by looking at high-entropy

chalcogenide spinels, $\text{MgSc}_{0.4}\text{Y}_{0.4}\text{Er}_{0.4}\text{Tm}_{0.4}\text{Yb}_{0.4}\text{Se}_4$ and $\text{Mg}_{1-0.5x}\text{Sc}_2\text{Se}_{4-x}\text{Br}_x$ ($x = 0.25, 0.5, 0.75$), to see if they would have more favorable properties than the previously studied ones [42]. It was found that the electronic conductivities of these materials were significantly lower than those of the previously studied spinels, at 3.3×10^{-9} and 3.6×10^{-13} S/cm, respectively, with the latter applying for $x = 0.5$. The most promising spinel turned out to be MgYb_2Se_4 [Fig. 2(b)], which not only had an electronic conductivity of 9.8×10^{-10} S/cm (an order of magnitude lower than the previously studied structures) but also had a very high ionic conductivity of 1.4×10^{-4} S/cm with a low activation energy of 0.364 eV [42]. This was calculated by creating a C|MgYb₂Se₄|C symmetric cell [42]. Typical Mg-ion-conducting spinels and their key properties are summarized in Table I.

IV. PHOSPHATES

Phosphates are another group of Mg-ion conductors that researchers have shown interest in. The archetypical phosphate is $\text{Mg}_{0.5}\text{Zr}_2(\text{PO}_4)_3$ (MZP), which is favored due to its stable framework with many interstitial sites through which Mg^{2+} ions can fit and migrate, as shown in Fig. 3. MZP belongs to the NASICON of $\text{AM}_2(\text{PO}_4)_3$, which has proved to be successful at conducting a variety of cation types [43,44]. MZP was first discovered in 1987 by Ikeda *et al.*, who attributed to it an ionic conductivity of 2.9×10^{-5} S/cm at 400 °C and an activation energy of 0.824 eV when put in a cell with platinum anode and cathode [45]. Several researchers have subsequently doped MZP with various ions to enhance its performance. In 2000, Imanaka *et al.* doped MZP with niobium to produce a material with the formula $\text{Mg}_{1-2x}(\text{Zr}_{1-x}\text{Nb}_x)_4(\text{PO}_4)_6$, creating a cell with a platinum anode and cathode, and found the highest ionic conductivity at $x = 0.15$, 6.89×10^{-3} S/cm at 800 °C, with an activation energy of 1.41 eV [46]. Adding more Nb past this value still decreases the activation energy, although the ionic conductivity is also reduced. To achieve smaller and thus more stable unit cell dimensions, as well as promote the interstitial inclusion of Mg^{2+} ions, Anuar and Mohamed doped Zr^{4+} with smaller Zn^{2+} and Al^{3+} ions to produce a material with the formula $\text{Mg}_{0.9+0.5y}\text{Zn}_{0.4}\text{Al}_y\text{Zr}_{1.6-y}(\text{PO}_4)_3$, and they found success in that the unit cell lattice parameters a and b decreased with increasing y value [47]. Increasing y also enhanced the ionic conductivity, reaching a peak at a value approximately 3 orders of magnitude higher than that of other Mg-based ceramic electrolytes, with a maximum ionic conductivity of 3.97×10^{-4} S/cm at room temperature for $y = 0.3$, accompanied by an extremely low activation energy of 0.039 eV [47]. Given these extraordinary metrics, it is curious that there are no follow-up studies on this composition, which may be due to the ionic conductor suffering from mechanical and interfacial

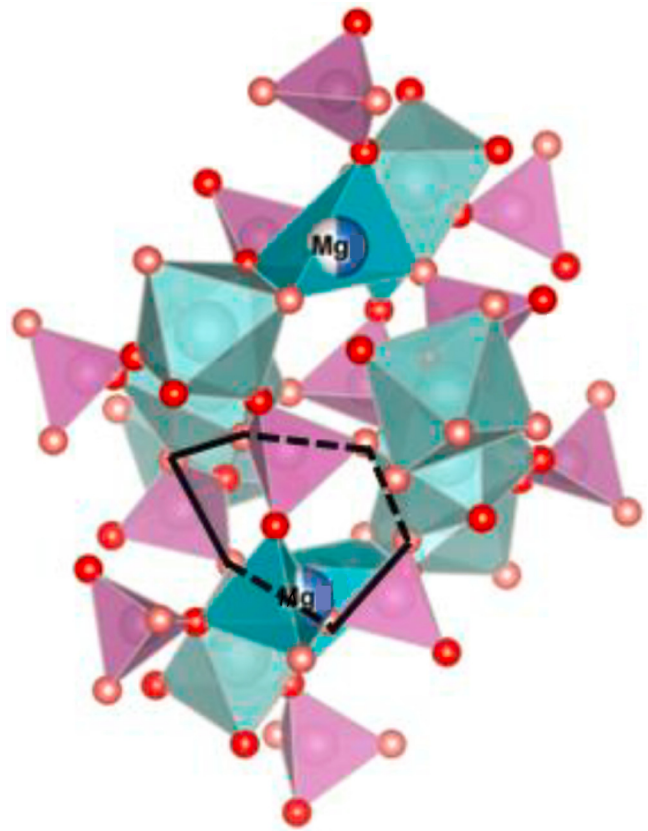


FIG. 3. NASICON structure of MZP, with tetrahedral PO_4 in magenta and octahedral ZrO_6 in green. Black contour shows the bottleneck of Mg^{2+} migration formed by edges of three PO_4 tetrahedral units and three ZrO_6 octahedral units. (Reproduced from Ref. [50]. Copyright 2020 Elsevier B.V.)

instabilities, as commonly found for NASICONs in such cases [48].

Liang *et al.* used liquid-feed flame spray pyrolysis (LF-FSP) to transform $\text{Mg}_{0.5}\text{Ce}_{0.2}\text{Zr}_{1.8}(\text{PO}_4)_3$ nanopowders into a thin film (≤ 50 μm) for use as an electrolyte [49]. They were motivated by the fact that thicker films had higher internal resistance, which led to a lower power output and decreased energy density. In addition, film thickness creates longer diffusion distances and high-concentration polarization, which causes discharge (particularly high-rate discharge) to end more quickly. LF-SFP has other important advantages compared to the previously used methods of MZP formation involving high-temperature or (non-LF-SFP) low-temperature sintering. High-temperature (above 1200 °C) sintering causes MZP to decompose; phosphorus to become volatile; and defects such as pores, cladding, and heterogeneous microstructures to form [50]. However, with typical methods of low-temperature sintering, high-density products like thin films are difficult to produce. LF-SFP is a low-temperature sintering method that allows one to produce a thin film. Liang *et al.* achieved an ionic conductivity of 2×10^{-10} S/cm at

25 °C and 1.9×10^{-7} S/cm at 200 °C with an activation energy of 0.307 eV (at temperatures ≥ 60 °C) [49]. They also produced 0.5-mm-thick pellets, which turned out to be twice as conductive as the film at 200 °C (reaching 3.8×10^{-7} S/cm) [49]. Liu *et al.* also used LF-SFP to prepare iron-doped $\text{Mg}_{0.5(1+x)}\text{Fe}_x\text{Zr}_{2-x}(\text{PO}_4)_3$ (with $x = 0.2$ leading to the optimal microstructure) thin film free of pores, with an ionic conductivity of 1.8×10^{-7} S/cm at 25 °C, an order of magnitude higher than that of pure MZP films, and a low activation energy of 0.141 eV [50].

In 2025, Wahab studied $\text{Mg}_{0.5}\text{Ti}_2(\text{PO}_4)_3$, a magnesium-titanium phosphate, instead of MZP [51]. They identified that the sintering temperature that gave the fewest impurities in the prepared material was 850 °C, achieving an ionic conductivity of 10^{-10} S/cm at room temperature and 10^{-7} S/cm at 500 °C, along with an ionic transference number of 0.98, suggesting that most charge mobility was from Mg ions and there was little electronic conductivity [51]. $\text{Mg}_{0.5}\text{Ti}_2(\text{PO}_4)_3$ exhibits an activation energy of 0.70 eV and is electrochemically stable up to 1.5 V [51]. Wei *et al.* created a hybrid solid electrolyte by adding 20 wt % magnesium ionic liquid to a NASICON-structured phosphate, $\text{Mg}_{0.5}\text{Sn}_2(\text{PO}_4)_3$, which produced a much higher room-temperature ionic conductivity of 1.11×10^{-4} S/cm with a low activation energy of 0.36 eV [52]. Typical Mg-ion-conducting phosphates and their key properties are summarized in Table I.

V. OTHER TYPES: OXIDES, SULFIDES, AND HALIDES

Besides borohydrides, chalcogenide spinels, and phosphates, researchers have studied a wide range of other inorganic Mg ionic conductors, including oxides, sulfides, and halides. In 2011, Omote *et al.* [57] prepared $\text{MgHf}(\text{WO}_4)_3$, using Mg^{2+} and Hf^{4+} as a substitute for the trivalent metal (*M*) in the ionic conductor series $M_2(\text{WO}_4)_3$ [53–56], and measured an ionic conductivity of 2.5×10^{-4} S/cm at 600 °C with an activation energy of 0.835 eV using platinum electrodes. Sulaiman *et al.* prepared a set of Mg-ion conductors based on MgSO_4 , $\text{Mg}(\text{NO}_3)_2$, and MgCO_3 systems using the sol-gel method, which could create larger surface areas and improved interfacial interactions, leading to high ionic conductivities [58]. It was found by varying the component ratios that the ionic conductivity was higher when an amorphous phase was formed in such systems. For example, they synthesized $(1-x)\text{MgSO}_4-x\text{Al}_2\text{O}_3$ ($0.1 \leq x \leq 0.6$), with the highest ionic conductivity of 5.12×10^{-6} S/cm at 303 K found at $x = 0.6$, where amorphous phase was dominant [59]. Another system of $(1-x)\beta\text{-MgSO}_4:\text{Mg}(\text{NO}_3)_{2-x}\text{MgO}$ exhibits as peak ionic conductivity of 10^{-6} S/cm at room temperature at $x = 0.4$ [58]. In these systems, disorder of the anionic sublattice of SO_4^{2-} in the amorphous state weakens the ionic bond of $\beta\text{-MgSO}_4$, facilitating Mg^{2+}

migration [58]. Even higher ionic conductivities were found in $(1-x)\text{Mg}(\text{NO}_3)_{2-x}\text{Al}_2\text{O}_3$ ($0.0 \leq x \leq 0.5$), reaching 4.0×10^{-4} S/cm and an ionic transport number of 0.85 at room temperature and at $x = 0.1$ [60]. In 2021, they synthesized $(1-x)\text{MgCO}_3-x\text{Al}_2\text{O}_3$ ($0.0 \leq x \leq 0.9$) and found that crystallinity decreased as Al_2O_3 concentration increased, with the ionic conductivity reaching a peak of 1.74×10^{-4} S/cm at 303 K for $x = 0.9$ [61]. A more amorphous state, leading to higher Mg^{2+} conductivity in oxides, was also reported by Gautam *et al.* in the context of investigating V_2O_5 as a cathode for magnesium batteries, with an immense improvement of about 7 orders of magnitude [62].

In 2023, Su *et al.* prepared magnesium phosphorus oxynitride (MgPON) by atomic layer deposition, which enabled precise thickness control and produced uniform dense films [63]. When deposited at 125 °C and added to a Pt|MgPON|Pt cell, the ionic conductivity of the resultant amorphous film reached 1.2×10^{-6} S/cm at 500 °C with an activation energy of 1.60 eV, which was a great improvement compared to the nitrogen-free MgPO equivalent [63]. In 2024, Iton *et al.* studied Mg-ion conduction in the MPS_3 -based ($M = \text{Mn}, \text{Cd}$) solid host doped with Mg in nonstoichiometric proportions [64]. It was found that adding H_2O ligands could greatly enhance Mg-ion conduction, reaching room-temperature ionic conductivities of 4.81×10^{-5} S/cm (with an activation energy of 0.599 eV) and 1.06×10^{-4} S/cm (with an activation energy of 0.557 eV) in $\text{Mg}_{0.25}\text{Cd}_{0.75}\text{PS}_3 \cdot 1.9\text{H}_2\text{O}$ and $\text{Mg}_{0.2}\text{Mn}_{0.8}\text{PS}_3 \cdot 1.8\text{H}_2\text{O}$, respectively [64]. Interestingly, the H_2O [or tetrahydrofuran (THF), acetonitrile (MeCN)] ligands enhanced the Mg-ion conductivity by both screening Mg^{2+} from electrostatic interactions and widening the interlayer spacing used as the conduction pathways, as shown in Fig. 4(a) [64].

Some halides can also serve as Mg-ion conductors. Tomita *et al.* prepared $\text{MgAl}_2\text{Cl}_{8-y}\text{Br}_y$ with varying Cl and Br ratios and, by measuring with both Mg metal and stainless-steel electrodes, found a peak ionic conductivity of 1.3×10^{-6} S/cm at 400 K (with an activation energy of 0.523 eV) at $y = 6$, corresponding to a monoclinic structure (space group $C2/c$), in which each Mg was octahedrally coordinated by six halogens, as shown in Fig. 4(b) [65]. In 2024, Yang *et al.* prepared claylike halides, $x\text{MgCl}_2\text{-GaF}_3$ ($0.75 \leq x \leq 1.25$), that had a deformable structure to maintain good electrolyte-electrode contact and reached a high ionic conductivity of 4.7×10^{-5} S/cm at room temperature at $x = 1$ with an activation energy of 0.406 eV and a low electronic conductivity of 4.01×10^{-8} S/cm [66]. They found that a full-cell battery with Mg electrodes failed after 60 h due to increasing impedance, whereas a $\text{Mo}_6\text{S}_8|\text{1MgCl}_2\text{-GaF}_3|\text{Mg}_x\text{Mo}_6\text{S}_8$ cell performed successfully [66]. Interestingly, Mg^{2+} and Cl^- exhibit correlated high mobility in such a system, with Mg^{2+} attaining the highest mobility in chlorine-rich environments with low

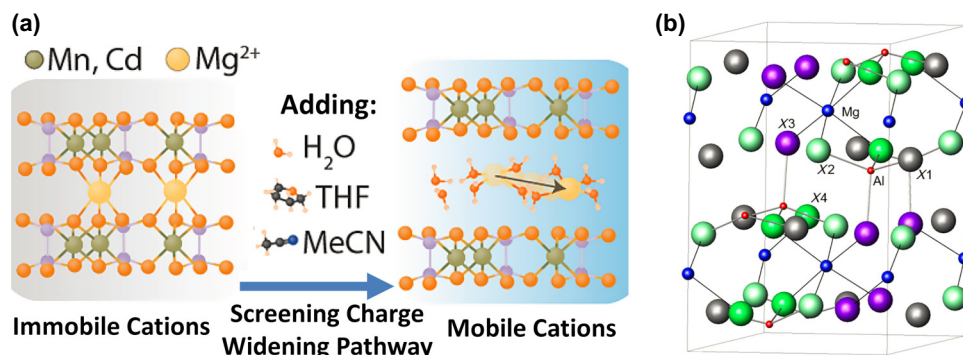


FIG. 4. (a) Adding H_2O , THF, and MeCN ligands to the Mg^{2+} -doped MPS_3 ($M = \text{Mn, Cd}$) solid host promotes Mg^{2+} mobility via both charge screening and pathway widening. (Reproduced from Ref. [64]. Copyright 2024 American Chemical Society.) (b) Structure of the $\text{MgAl}_2\text{Cl}_{8-y}\text{Br}_y$ halides, where each Mg^{2+} is octahedrally coordinated by six halogen sites ($X1-4$). (Reproduced from Ref. [65]. Copyright 2021 Elsevier B.V.)

coordination numbers [66]. Most recently, inspired by the high ionic conductivity of Li_2ZrCl_6 , Rom *et al.* prepared MgZrCl_6 via mechanochemistry [67]. However, the ionic conductivity was too low ($<10^{-8}$ S/cm) to detect, probably because the Mg ions were found in stable octahedral sites [67]. The key properties of the above-discussed ionic conductors are summarized in Table I.

VI. FINDING Mg-ION CONDUCTORS VIA HIGH-THROUGHPUT SCREENING AND MACHINE LEARNING

With a large library of Mg-containing solids to be investigated for their potential as solid-state electrolytes, one strategy that has risen to prominence is high-throughput computational screening with the aim of finding candidates with desirable properties. Takeda *et al.* [68] used a high-throughput exhaustive search of Mg oxides (which are preferred over sulfides due to their higher stability in air) using force-field molecular dynamics simulations and a percolation algorithm based on bond-valence methods [69,70], looking for candidates with low ion-migration energies. They identified μ -cordierite, $\text{Mg}_{0.6}\text{Al}_{1.2}\text{Si}_{1.8}\text{O}_6$, due to its low migration energy barrier of 0.4 eV and its components of *s*- and *p*-block elements, which would block electronic conductivity. They subsequently characterized the material using both experimental alternating-current impedance measurements and first-principles molecular dynamics simulations, finding an ionic conductivity of 1.6×10^{-6} S/cm at 500 °C with an activation energy of 1.32 eV. The activation energy obtained differed significantly from the migration energy barrier calculated using the bond-valence force field, raising the question of whether the latter method was viable for identifying ideal electrolytes. They also found that, although the AlO_4 and SiO_4 tetrahedra in $\text{Mg}_{0.6}\text{Al}_{1.2}\text{Si}_{1.8}\text{O}_6$ formed a one-dimensional tunnel containing Mg ions, Mg^{2+} migrated

via a spiral-like three-dimensional path rather than migrating through the tunnel [68].

Other researchers have focused on high-throughput screening to identify ionically conductive chalcogenide compounds. Kabanov *et al.*, starting with approximately 1500 ternary and quaternary chalcogenides in the ICSD database (some of which contain Ca, Zn, or Al instead of Mg), employed a geometrical-topological approach, where the crystal space was partitioned by an atomic Voronoi polyhedron to narrow this collection down to 104 candidates [71]. In this method, the edges and vertices of the polyhedra form a migration map for ions. They then used bond-valence-site energy analysis, another computational method based on the valence-bond model, to calculate migration energies. They accomplished this by taking 10 random configurations for each disordered structure, optimizing them, and averaging their migration energies. By setting the conditions that the migration energy barrier must be less than 0.60 eV and the global instability index must be below the threshold of 0.26, they narrowed the candidates further to 16. Finally, they used density functional theory (DFT) calculations to narrow the list to 11 chalcogenides. One Mg-containing candidate, MgEr_2Se_4 , was identified in the final list. This is a spinel that had been discovered in previous computational studies as a promising solid-state electrolyte with a relatively low migration energy of less than 0.38 eV and good phase stability with an energy-above-convex-hull below 50 meV/atom [72]. Most recently, Dheerasinghe *et al.* conducted a high-throughput screening of high-entropy oxide spinels based on their electrochemical and phase stabilities, as determined by DFT-calculated data [73]. It is found that the standard deviation of the ionic radii of the octahedral-site elements in the spinel structure and their electronegativities correlate best with the materials stabilities [73].

In addition to data-driven high-throughput screening, many researchers have also employed machine learning for the discovery of ionic conductors beyond lithium.

Dong *et al.* employed two classification models based on the “XGBoost” machine-learning algorithm [74], which combined predictions from a sequence of multiple “weak” predictive models to create a single and powerful “strong” model. In a multistep screening process, they examined garnets from a wide pool of 43 632 materials, involving machine-learning classification predictions based on the band gap and the energy-above-convex-hull data [75]. Several Mg-containing ionic conductors were discovered in the final list, including $\text{Ca}_3\text{Mg}_2\text{Fe}_3\text{O}_{12}$, $\text{La}_3\text{Mg}_2\text{V}_3\text{O}_{12}$, $\text{Nd}_3\text{Mg}_2\text{Fe}_3\text{O}_{12}$, $\text{La}_3\text{Mg}_2\text{Fe}_3\text{O}_{12}$, $\text{La}_3\text{Mg}_2\text{Ni}_3\text{O}_{12}$, $\text{Nd}_3\text{Mg}_2\text{Zn}_3\text{O}_{12}$, $\text{Ca}_3\text{Mg}_2\text{Li}_3\text{O}_{12}$, $\text{Y}_3\text{Mg}_2\text{La}_3\text{O}_{12}$, $\text{Mg}_3\text{Fe}_2\text{Nd}_3\text{O}_{12}$, $\text{Mg}_3\text{Co}_2\text{Nd}_3\text{O}_{12}$, $\text{Mg}_3\text{Ni}_2\text{Nd}_3\text{O}_{12}$, $\text{Mg}_3\text{Ni}_2\text{La}_3\text{O}_{12}$, $\text{Zn}_3\text{Mg}_2\text{La}_3\text{O}_{12}$, $\text{Mg}_3\text{Mn}_2\text{La}_3\text{O}_{12}$, $\text{Mg}_3\text{Y}_2\text{La}_3\text{O}_{12}$, $\text{Zn}_3\text{Mg}_2\text{Nd}_3\text{O}_{12}$, $\text{Mg}_3\text{Sc}_2\text{La}_3\text{O}_{12}$, $\text{Mg}_3\text{Ti}_2\text{La}_3\text{O}_{12}$, $\text{Mg}_3\text{Cu}_2\text{Nd}_3\text{O}_{12}$, $\text{Cu}_3\text{Mg}_2\text{La}_3\text{O}_{12}$, $\text{Fe}_3\text{Mg}_2\text{Nd}_3\text{O}_{12}$, $\text{Co}_3\text{Mg}_2\text{Nd}_3\text{O}_{12}$, $\text{V}_3\text{Mg}_2\text{La}_3\text{O}_{12}$, $\text{Cr}_3\text{Mg}_2\text{Nd}_3\text{O}_{12}$, $\text{Mg}_3\text{Mn}_2\text{Y}_3\text{F}_{12}$, and $\text{Mg}_3\text{Co}_2\text{Y}_3\text{F}_{12}$ [74]. Some researchers have developed new machine learning systems with the aim of making them more efficient. Wang *et al.* utilized a platform called artificial intelligence ion-migration activation energy for identifying suitable ionic conductors, which was 10^5 times faster in its predictions than other methods [76]. They took advantage of this speed to start from a formidable 144 595 compounds (not all containing Mg) and from that found 316 of the most promising, including two Mg-ion conductors, MgY_2Se_4 and Mg_2TiO_4 [76]. In 2025, Devi *et al.* made a graph neural network–based architecture that could predict migration energy, which was notably more effective than traditional machine learning methods [77]. This method obtained 88.2% precision and 77.4% sensitivity in identifying solids with a migration energy below 65 meV, and it could also distinguish multiple ion-migration pathways within a given crystal structure, which would serve as an effective tool for finding Mg-ion conductors in the future.

Finding solid-state electrolytes through high-throughput screening or machine learning methods requires a set of effective and easy-to-access parameters or features. For example, the band gap and energy-above-convex-hull (as mentioned in the above studies) are such parameters that correlate well with the electron insulation and synthesizability of a solid electrolyte. However, accurately calculating the most important property of a solid electrolyte—ionic conductivity—is expensive and time-consuming. Therefore, it is still impossible to perform high-throughput screening of ionic conductors using directly calculated DFT ionic conductivities. To overcome this, Jaafreh *et al.* proposed that the phonon density of states and, in particular, the “phonon band center,” could serve as a proxy for ionic conductivity, by showing a correlation between heightened ionic mobility and low phonon frequencies (as demonstrated in Fig. 5) in 69 Mg-containing compounds using machine learning [79]. Mg-containing materials are often brittle, which can prove a barrier to implementation as solid electrolytes (in

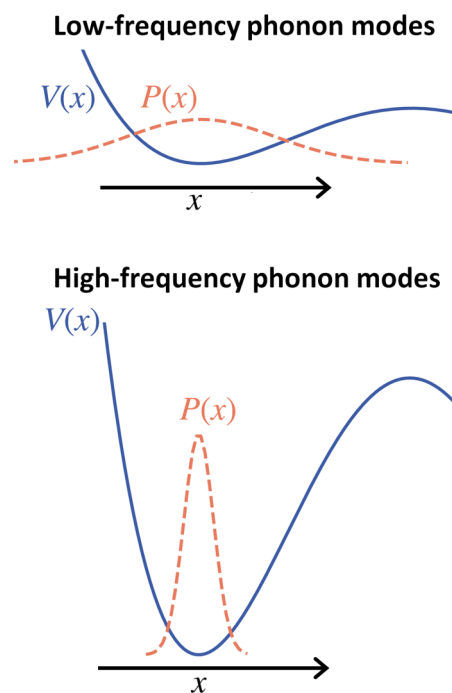


FIG. 5. Physical foundation of lower phonon frequencies correlating with lower migration barriers, where the low-frequency phonon modes show a higher probability, $P(x)$, for far-from-equilibrium displacements upon thermal excitation than high-frequency modes, due to the much stiffer potential well, $V(x)$, of the latter. (Reproduced from Ref. [78]. Copyright 2024 American Chemical Society.)

particular, their synthesis as pellets). Therefore, besides ionic conductivity, Jaafreh *et al.* also looked for materials with high ductility using the Pugh criterion in the model, reasoning that a high ductility of an electrolyte candidate meant easier synthesis from powders to pellets [79]. Greene and Siegel also studied the phonon features of multivalent ion (e.g., Mg^{2+} , Ca^{2+} , Zn^{2+} , and Al^{3+}) conductors but found that the phonon band center alone did not correlate well enough with the migration barrier of the ionic conductors to be used as an effective metric [78]. They instead created another metric called the “mean alignment-weighted frequency,” which considered both the frequencies of the phonon modes of the cations and their vibrational amplitudes towards the neighboring sites and found that this correlated with migration barriers more strongly [78]. These phonon parameters or features can be applied for the high-throughput screening of Mg solid-state electrolytes.

VII. CONCLUSION

Although several promising magnesium solid-state electrolytes have been discovered, a long journey remains ahead until any of them achieves commercial viability. There are still a few that exhibit good ionic conductivity

TABLE II. Components of solid-state full-cell batteries used for measuring the Mg^{2+} conductivity or as all-solid-state battery prototypes.

Anode	Solid-state electrolyte	Cathode	Type of electrolyte
Mg	$\text{Mg}(\text{BH}_4)(\text{NH}_2)$	Pt	Borohydride [22]
Mo	$\text{Mg}(\text{BH}_4)_2(\text{NH}_3\text{BH}_3)_2$	Mo	Borohydride [25]
Mg	$\text{Mg}(\text{BH}_4)_2 \cdot 2\text{NH}_3$	Mo	Borohydride [31]
Al	MgAl_2O_4	Mg	Chalcogenide [36]
Al	MgAl_2O_4	C	Chalcogenide [36]
Mg	MgAl_2O_4	C	Chalcogenide [36]
Au	MgSc_2Se_4	Au	Chalcogenide [39]
Au	MgY_2Se_4	Au	Chalcogenide [41]
Au	MgTm_2Se_4	Au	Chalcogenide [41]
Au	MgEr_2Se_4	Au	Chalcogenide [41]
C	MgYb_2Se_4	C	Chalcogenide [42]
Pt	$\text{Mg}_{0.5}\text{Zr}_2(\text{PO}_4)_3$	Pt	Phosphate [45]
Pt	$\text{Mg}_{1-2x}(\text{Zr}_{1-x}\text{Nb}_x)_4(\text{PO}_4)_6$	Pt	Phosphate [46]
Stainless steel	$\text{Mg}_{0.5}\text{Sn}_2(\text{PO}_4)_3$ 20 wt % 1M $\text{Mg}(\text{TFSI})_2$ in $[\text{EMIM}][\text{TFSI}]$	Stainless steel	Phosphate [52]
Pt	$\text{MgHf}(\text{WO}_4)_3$	Pt	Oxide [57]
Stainless steel	$0.6\beta\text{-MgSO}_4\text{:Mg}(\text{NO}_3)_{1.6}\text{MgO}$	Stainless steel	Oxide [58]
Stainless steel	$0.9\text{Mg}(\text{NO}_3)_2\text{-}0.1\text{Al}_2\text{O}_3$	Stainless steel	Oxide [60]
Stainless steel	$\text{MgCO}_3\text{-Al}_2\text{O}_3$	Stainless steel	Oxide [61]
Pt	MgPON	Pt	Oxide [63]
Mg	$\text{MgAl}_2\text{Cl}_2\text{Br}_6$	Mg	Halide [65]
Stainless steel	$\text{MgAl}_2\text{Cl}_2\text{Br}_6$	Stainless steel	Halide [65]
Mo_6S_8	$1\text{MgCl}_2\text{-GaF}_3$	$\text{Mg}_x\text{Mo}_6\text{S}_8$	Halide [66]

even at room temperature, and many of those that do (chalcogenide spinels in particular) have high electronic conductivities that pose an impediment. However, a greater understanding of the causes of this electronic conductivity could enable remediation of this flaw. Of the three major categories, borohydrides have the best balance of relatively high ionic conductivity and low electronic conductivity. Chalcogenide spinels, despite exhibiting decent ionic conductivity, often exhibit high electronic conductivity; however, promising work is underway to better understand this phenomenon and mitigate it. Finally, phosphates exhibit ionic conductivity that is often too low to be as appealing as that of the others, with some notable exceptions, such as the $\text{Mg}_{1.05}\text{Zn}_{0.4}\text{Al}_{0.3}\text{Zr}_{1.3}(\text{PO}_4)_3$ multi-component system (see Table I) [47], which have not been pursued further in follow-up studies. Full-cell all-solid-state Mg batteries based on these inorganic solid-state electrolytes are still in the early research stage, with only a few prototypes reported, as mentioned above and summarized in Table II, which suffer from poor ionic conductivity and high interfacial impedance [22,31,36,66].

Further chemical and mechanical engineering of these and other materials could improve the relevant properties. One effective strategy is to enhance the properties of existing Mg-ion conductors through anion or cation doping and substitution, as demonstrated in the development of Li/Na solid-state electrolytes [3]. Other promising strategies include the design of multicomponent or high-entropy materials [73]; defect and grain boundary engineering to

suppress electronic conductivity and reduce resistance; and the introduction of *ad hoc* interphases, coatings, or interlayers to stabilize the electrode-electrolyte interface and improve contact. However, researchers do not have to stop at just modifying the Mg solid-state electrolyte candidates we already have—a huge pool of candidates, mostly falling outside the three major categories, remains to be investigated. Further high-throughput computational and machine learning-based screening of this pool are necessary. By expanding the candidate pool, unraveling the mechanisms of Mg-ion diffusion and interfacial stability, and fine-tuning those we already understand, we will finally achieve the “holy grail” of a commercially viable Mg solid-state electrolyte.

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DATA AVAILABILITY

No data were created or analyzed in this study.

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