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Data-driven atomistic modeling of crystalline and glassy solid-state electrolytes

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All-solid-state batteries promise safer, more stable, and higher-energy-density storage, but progress hinges on atomistic insight into solid electrolytes. Machine-learning force fields (ML-FFs) offer near-first-principles accuracy at molecular-dynamics scales, enabling simulations that are orders of magnitude larger and longer than *ab initio* approaches (e.g. density funcitonal theory). In this Review, we discuss recent ML-FF frameworks and the application of them on studying both crystalline and glassy solid electrolytes. Particually, we compare various ML-FF models and training strategies, examine transferability and uncertainty quantification, and outline best practices for data generation and validation. The applications of ML-FF on battery systems reveal advances in illustrating ionic-transport pathways, defect-mediated conduction, structure-property relationships, phase stability and transformations, and interfacial phenomena at grain boundaries and electrode|electrolyte contacts. Then we conclude with perspectives on key challenges—including long-range electrostatics, chemical reactivity, and multicomponent complexity. Together, these developments position ML-FFs to accelerate the discovery and optimization of robust, high-performance solid electrolytes for practical all-solid-state batteries.

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1. Introduction

Global decarbonization and electrification are driving a sharp rise in battery demand.¹ Conventional lithium-ion batteries with flammable liquid electrolytes face safety risks and energy density limits that restrict their broader deployment. All-solid-state batteries (ASSBs) have been proposed to address these challenges with enhanced safety and higher energy density potential.² However, broad commercialization of ASSBs has yet to be realized due to challenges in cost, cycle life, and fast

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charging.³ Many of these barriers are fundamentally rooted in materials issues—most notably the solid-state electrolyte (SSE), which strongly impacts the battery system's safety, power capability and durability. Despite the long commercial success of liquid electrolytes, practical SSEs are hindered by limited ionic conductivity, unstable and resistive electrode–electrolyte interfacial, and manufacturing scalability constraints.⁴

To address the materials limitations outlined above, we need tools that resolve the atomistic processes governing ionic transport and interfacial stability. Computational methods now play an important role in advancing the understanding and design of battery materials, ^{5–8} particularly for investigating phenomena that are difficult to access experimentally. ⁹



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Meanwhile, machine learning approaches are accelerating discovery and optimization across battery materials research. 10-13 Among these, machine learning force fields (ML-FFs) have emerged as a powerful computational framework applied broadly to inorganic SSEs systems from crystalline phases to glassy solid state electrolytes (GSEs). 13-16 ML-FFs bridge the long-standing fidelity-efficiency gap in materials simulation: classical force fields are fast but constrained by fixed functional forms and empirical parameterization, 17,18 which limits accuracy for multicomponent SSEs chemistries and their complex ion transport. This transportation is often mediated by a wide range of defects, including stacking faults, 19 Li-stuffing, 20 cation site disorder, 21,22 and anion site disorder. 23-25 Firstprinciples calculations (e.g. density functional theory, DFT) offer high accuracy, but their computational cost scales drastically with system size $(O(N^3)-O(N^7)$ in the number of electrons), restricting accessible time-scales and length-scales. In contrast, ML-FFs enable simulations that reach millions of atoms, ²⁶ and extend into microsecond regime while retaining near-DFT accuracy, making them ideal for examining long-timescale ionic transport in batteries.

In this Review, we will first discuss the major families of inorganic solid-state electrolytes and their transport and stability characteristics. Then, we summarize recent computational studies that employed ML-FFs to investigate SSE structures, ionic dynamics and electrode-electrolyte interfaces. Finally, we will present case studies illustrating how to use ML-FFs to elucidate ionic transport mechanisms, quantify interfacial processes, and map structure-property relationships, thereby informing the design of next-generation ASSBs.

2. Solid state electrolytes

Based on different criteria, SSEs can be categorized into several classification schemes. Structurally, they are commonly grouped into crystalline SSEs, amorphous glassy solid-state electrolytes, and glass-ceramic SSEs. Fig. 1 shows structural schematics of typical crystalline SSEs, including oxides, sulfides, halides, and hydrides. Certain crystalline SSEs, such as lithium germanium phosphorus sulfide (LGPS), achieve a high



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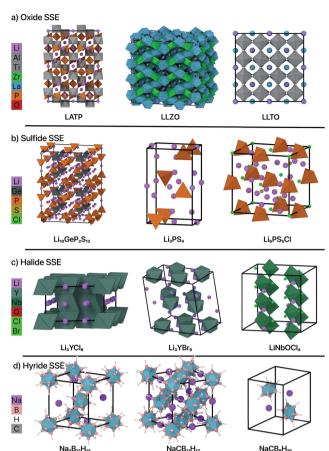


Fig. 1 Schematic structures of representative crystalline solid-state electrolytes (SSEs): (a), oxides; (b), sulfides; (c), halides; (d), hydrides. For clarity, anions at the vertices of the coordination polyhedra (O, S, and halides) are

room-temperature ionic conductivity of 12 mS cm⁻¹, but their macroscopic performance can be limited by grain boundaries resistance and anisotropy.²⁷ In contrast, GSEs are intrinsically isotropic and free of grain boundaries. 28 The ionic conductivity of a GSE is theoretically higher than that of its crystalline counterpart because of its typically larger molar volume.²⁹ In certain systems, such as NASICON-type, 30 sulfide-based, 31 and halide-based electrolytes, 32 an even higher ionic conductivity can be achieved through partial crystallization of the amorphous precursor into a glass-ceramic solid electrolyte. This enhancement arises from the formation of highly conductive nanoparticles and grain-boundaries. 33 The resulting glass-ceramic solid electrolyte combines the favorable mechanical properties of the flexible polyanion network found in glasses with the enhanced ionic conductivity provided by the crystalline parts.

In addition to the crystalline-amorphous distinction, SSE can be classified by the topology of their anion framework.³⁴ In polyhedral-network types SSE, polyhedral share corners or edges to form a continuous framework, that provides migration channels for Li⁺, Na⁺ transportation. In cluster-anion electrolytes, the anions arrange into fcc, hcp, or bcc sublattices

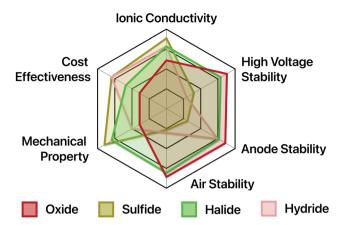


Fig. 2 Radar chart that compares the key performance of various types of solid electrolytes.

without direct interconnection between them, and alkali cations diffuse through interstitial sites.³⁴

From a compositional perspective, SSEs are categorized into oxides, sulfides, halides, and hydrides, as shown in Fig. 2. Oxides electrolytes (Fig. 1(a)) generally exhibit good compatibility with metal anodes and wide electrochemical windows, but they typically possess low ionic conductivity, higher stiffness, and often require high-temperature processing. 27,35 Sulfides (Fig. 1(b)) often exhibit high room-temperature ionic conductivity and enable low-temperature processing, yet they suffer from narrower electrochemical stability windows, reactivity with metal anodes, and sensitivity to air and moisture.^{36,37} Halides electrolytes (Fig. 1(c)) provide wide electrochemical windows, 38,39 and have shown encouraging cycling performance. For example, the recently reported LaCl3-based LiTaLaCl SSE show 81.6% capacity retention after 100 cycles against a Li metal anode. 40 Complex hydrides (Fig. 1(d)) show good thermal and electrochemical stability and favorable mechanical properties, but they are sensitive to moisture and electrode materials.41

In what follows, we focus on representative Li-based SSEs with high ionic conductivity to illustrate these classes and to establish structure–property links that guide materials selection and device design.

2.1 Crystalline solid-state electrolytes

Garnet type. Garnet-type SSE with the general formular $\text{Li}_x \text{M}_3 \text{M}_2' \text{O}_{12}$ are among the most intensively studied oxide systems. They are structurally related to the oxide-garnet framework of $\text{Ca}_3 \text{Al}_2(\text{SiO}_4)_3$, with Li replacing the Si atoms and occupying interstitial sites within the garnet lattice. Depending on the Li content x, garnet-type SSEs are often divided into subtypes: Li₃-type (Li₃La₃Te₂O₁₂), Li₅-type (Li₅La₃M₂O₁₂), Li₆-type (Li₆ALa₂M₂O₁₂), and Li₇-type (Li₇La₃Zr₂O₁₂).

Among these, $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZO) is the prototypical oxide SSEs, achieving room temperature ionic conductivity up to 10^{-3} S cm⁻¹ in optimized compositions. LLZO exists in two polymorphs: a high-conductivity cubic phase⁴² and a

low-conductivity tetragonal phase.⁴³ In the tetrahedral structure, Li ordering—including full occupation of tetrahedral sites—reduces the number of available vacancies and narrows migration pathways, yielding conductivities near 10⁻⁶ S cm⁻¹. In contrast, the cubic phase features partially occupied Li sublattices that create a three-dimensional network of accessible sites, enabling less correlated Li⁺ motion and substantially higher conductivity.^{34,44}

Perovskite type. Typical perovskite-type SSEs, most notably the $\text{Li}_{3x}\text{La}_{2/3-x}\text{TiO}_3$ (LLTO) family and compounds of the form (Li, Sr)(M, M')O₃, are derived from the ABO₃ perovskite structure, in which large A-site cations (*e.g.* La³⁺, Sr²⁺) and smaller B-site cation (*e.g.*, Ti⁴⁺, Zr⁴⁺) define the framework for fast-ion conduction.

Perovskite-type SSEs are generally A-site deficient, creating vacant A sites that enable Li⁺ migration through the interconnected BO₆ octahedral channels. Tetragonal LLTO attains room-temperature ionic conductivities on the order of 10^{-4} S cm⁻¹, with optimal compositions around x=0.11, achieving 1.3×10^{-3} S cm⁻¹. In LLTO, La³⁺ cations are unevenly distributed along the c-axis, resulting in La-rich and La-poor layers. Li⁺ ions migrate relatively freely within the ab plane, whereas transport along the c-axis is limited by temperature-dependent bottlenecks associated with this La ordering.

NASICON type. The NASICON (Na superionic conductors) family was first reported in 1976 with the general formula $Na_{1+x}Zr_2Si_xP_{3-x}O_{12}$. Lithium-based NASICON-type SSEs adopt the formula LiMM'(PO₄)₃ and typically exhibit room-temperature ionic conductivities of 10^{-4} to 10^{-3} S cm⁻¹. A prototypical member is $Li_{1+x}Al_xTi_{2-x}(PO_4)_3$ (LATP), for which optimal Al substitution (x = 0.3) yields conductivities near $\sim 10^{-3}$ S cm⁻¹ at room temperature. As

In NASICON-type SSEs, MO₆ octahedra connect with PO₄ tetrahedra through corner-sharing oxygen atoms, forming a 3D framework that facilitates alkali ion movement in LATP, aliovalent Al³⁺ substitution for Ti⁴⁺ requires charge compensation by additional Li⁺, creating preferred Li interstitial sites adjacent to AlO₆ units. Although the smaller Al³⁺ contracts the lattice, the combined effects of increased charge carrier concentration and favorable local environments enhance Li⁺ mobility and boost conductivity. ^{48–50}

LGPS type. The earliest LISICON (Li superionic conductor) materials, such as $\text{Li}_{14}\text{Zn}(\text{GeO}_4)_4$, exhibited low room-temperature ionic conductivity ($\sim 10^{-4} \text{ mS cm}^{-1}$).⁵¹ Replacing oxygen with sulfur led to thio-LISICON compositions (*e.g.* $\text{Li}_{3.25}\text{Ge}_{0.25}\text{P}_{0.75}\text{S}_4$) that improved conductivity into the 10^{-4} to 10^{-3} S cm⁻¹ range.^{52,53}

The $\rm Li_{10}GeP_2S_{12}$ (LGPS) family, first reported in 2011, achieves exceptional room-temperature conductivity of up to 12 mS cm $^{-1}$. $^{54-56}$ Unlike the orthorhombic thio-LISICON structures, LGPS adopts a $P4_2/nmc$ tetragonal structure 54 consisting of a three-dimensional framework comprising ($\rm Ge_{0.5}P_{0.5}$)S₄ tetrahedra, PS₄ tetrahedra, along with $\rm LiS_4$ tetrahedra, and $\rm LiS_6$ octahedra. $\rm Li^+$ transport proceeds through quasi-one-dimensional channels along the c axis that are interconnected

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within the ab plane, yielding effectively three-dimensional diffusion pathways.⁵⁷

Argyrodite type. The original argyrodite Li₇PS₆ exhibits low room-temperature ionic conductivity (10⁻⁶ S cm⁻¹) because its high-conductivity cubic phase is only stable at elevated temperatures (e.g. 483 K).⁵⁸ Deiseroth and co-workers overcame this limitation by substituting one sulfur with a halide X (Cl, Br, I) in the chemical unit to form Li₆PS₅X, thereby stabilizing the high-temperature cubic phase at room temperature and achieving conductivities on the order of 10⁻³ S cm^{-1,59} Subsequent studies showed that additional cation substitutions (e.g. Si, Ge) can likewise stabilize the cubic phase and other high temperature phases at ambient conditions.⁶⁰

Li₆PS₅X argyrodite-type SSEs adopts a crystal structure with space group of $F4\bar{3}m$. The anions framework forms a cubic close-packed sublattice in which PS4 tetrahedra occupy octahedral sites, while remaining S2- anions reside in tetrahedral sites. Li⁺ primarily occupies the 24g and 48h Wyckoff positions. 59,61 For Cl and Br compositions, S2- and halide anions exhibit site disorder over the 4a and 4c positions. This anion disorder and Cl⁻-induced Li⁺ vacancy formation has been identified as a key contributor to the enhanced Li⁺ conductivity of argyrodite electrolytes. 24,25,62,63

Halide type. Halide-type SSEs generally have the chemical formula LiaMXb and can be categorized by metal center into group 3 elements (e.g. Y, Sc, Er), group 13 elements (e.g. Al, Ga, In), and divalent metals.³⁸ Many group 3 and group 13 metal halides reach room-temperature ionic conductivities on the order of 10^{-3} S cm⁻¹.

In Li₃YCl₆ and Li₃YBr₆, the halide anions form close-packed sublattices (hexagonal close-packed (HCP) for Li₃YCl₆ and cubic close-packed (CCP) for Li₃YBr₆), while Li⁺ and Y³⁺ cations occupy octahedral sites. The high ionic conductivity is attributed to partially vacant octahedral sites that provide interconnected diffusion pathways. Owing to its HCP stacking, Li₃YCl₆ exhibits strongly anisotropic diffusion: concerted Li⁺ migration along the c axis dominates overall conductivity because it proceeds with lower activation barriers than diffusion in the ab plane.

LNCO type. Oxyhalide SSEs are an emerging class that seek to combine the chemical stability of oxides with the high Li⁺ mobility often found in halides. The anti-perovskite structure Li₃OCl oxyhalide reaches room-temperature conductivities of $\sim 10^{-4} \text{ S cm}^{-1}$. More recently, metal oxyhalides have shown markedly higher performance: Hu et al. reported a low-cost LiZrClO oxyhalide SSE with a room temperature conductivity of 2.42 mS cm⁻¹,⁶⁴ and the studies on LiNbOCl₄ (LNCO) and LiTaOCl₄ demonstrated ionic conductivities near 10⁻² S cm⁻¹ at room temperature.65 Their sodium counterparts can also achieve high ionic conductivities on the order of 10^{-3} S cm^{-1,66}

Despite these promising transport properties, the crystal structure of LiNbOCl₄ remains under activate debate^{65,67-69} due to poor crystallinity and low coherence length.⁶⁷ Regardless, it is commonly described as consisting 1D parallel polyanion chains built from [NbOCl₄] octahedra, with highly disordered Li⁺ ions occupying interstitial sites.^{68,69} Li⁺

transport involves two different diffusion pathways: (1) diffusion along the a-axis following the polyhedral chain; and (2) diffusion within the bc plane, with the former mechanism generally considered energetically favored.⁶⁹

Complex hydrides. Motivated by the discovery that LiBH₄ exhibits high Li⁺ conduction and can function as a solid-state electrolyte, complex hydrides have emerged as a distinct SSE family. These materials are composed of alkali and alkalineearth cations (e.g. Li⁺, Na⁺, Ca²⁺) and complex anions such as BH_4^- , AlH_4^- , and *closo*-type borate/carborate species ($B_{12}H_{12}^{2-}$, $B_{10}H_{10}^{2-}$, $CB_9H_{10}^{-}$, $CB_{11}H_{12}^{-}$).

These materials typically exhibit ordered low-temperature phases and disordered (often "plastic") high-temperature phases, the latter displaying substantially higher ionic conductivity. For example, Li₂B₁₂H₁₂ undergoes an order-disorder phase transition at \sim 615 K; in the disordered phase, rotationally mobile B₁₂H₁₂²⁻ anions occupy the fcc sublattices while Li⁺ migrates through vacancy-rich interstitial networks. Because these highly conducting disordered phases are typically unstable at room temperature, compositional strategies such as anion mixing⁷⁰⁻⁷² have been employed to stabilize them closer to practical operating conditions. In systems including $NaCB_{11}H_{12}$ - $NaCB_9H_{10}$ and $Na_2B_{12}H_{12}$ - $Na_2B_{10}H_{10}$ anion mixing has successfully suppress the order-disorder transition temperature and preserved fast-ion transport at reduced temperatures.⁷¹

2.2 Glassy solid-state electrolytes

GSEs offer unique advantages due to their amorphous structure, lower elastic moduli that mitigate stress during volume changes under charge/discharge cycles, inherent isotropic ionic conduction, the absence of grain boundaries and thus grainboundary resistance, and superior mechanical compliance.⁷³ Their grain-boundary-free microstructure also inhibits dendrite nucleation and growth, addressing a key failure mode of many crystalline electrolytes.74

Theoretically, GSEs can achieve a higher ionic conductivity than their crystalline counterparts because structural disorder provides greater free volume and less constrained diffusion pathways. In addition, the absence of grain boundaries eliminates intergranular impedance that commonly limits performance in polycrystalline solids.

Thiophosphate glass. Li₂S-P₂S₅ (LPS) glasses are among the most widely studied GSEs. The 75Li₂S·25P₂S₅ composition exhibits a room-temperature ionic conductivity of $\sim 2 \times$ 10⁻⁴ S cm⁻¹ with a Li⁺ transport number near unity.⁷⁵

Despite these advantages, LPS glass suffers from poor compatibility with Li anodes due to interfacial chemical reactivity. Several modifications strategies have been developed to address this limitation. Introducing a second network formers, such as Si, improves cycling stability by suppressing dendrite formation. 28,76 In addition, SnS2 additions enhance the air stability. 76 Furthermore, halide additions, particularly LiI, significantly enhance conductivity; for example, adding 20 mol% LiI to $70\text{Li}_2\text{S}\cdot30\text{P}_2\text{S}_5$ increases conductivity from 1.3 \times 10^{-4} to 5.6 imes 10^{-4} S cm $^{-1}$ and widens the electrochemical

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stability window.⁷⁷ Oxygen doping offers another route and typically improves chemical stability.⁷⁸

LiPON glass. Lithium phosphate oxynitride (LiPON) is another important family of the GSEs, typically achieving conductivities of $\sim 10^{-6} \text{ S cm}^{-1,79}$ which limits its application primarily to thin film batteries. Despite its lower conductivity relative to other systems, LiPON shows exceptional stability against Li metal, sustaining over 10 000 cycles with 95% capacity retention. This stability makes LiPON widely used as a protective interfacial coating on battery electrodes and electrolytes, where it provides chemical inertness and interfacial stabilization.80

Oxyhalide glass. Several oxyhalide amorphous solid electrolytes, referred to as AMCO (where A = Li/Na; M = Mg, Al, Zr, Ta, Nb, etc; C = Cl, I), have been developed in recent years, $^{64,66,81-86}$ and exhibit both high ionic conductivity and good electrochemical stability. Among these oxyhalide GSEs, systems based on earth-abundant metals such as aluminum (LiAlClO86) and zirconum (LiZrClO^{64,82}) are particularly promising. These compositions offer a cost-effective alternative to high-ionic conductivity halides (e.g. Li₃YCl₆, Li₃InCl₆), which often contain expensive rare elements. This strategy is illustrated by the work of Dai et al., who demonstrated that partial oxygen incorporation can transform the low-conductivity crystalline LiAlCl4 into a vitreous lithium-aluminum oxychloride glass with significantly enhanced ionic conductivity.86 You et al. proposed a trimer-like Si₃O₂Cl₈ structural motif for such this glass; their experimental indicate that the resulting GSE lacks ionic O-Li bonding, and that Cl⁻ anions undergo rotational motion. 85 The introducing of oxygen promote glass formation without forming non-bridging oxygen species, thereby increasing free volume and enable greater Cl- rotational dynamics, which in turn facilitate Li⁺ diffusion.⁸⁷

3. Machine learning force fields

The previous section discussed the breadth of SSEs chemistries and their rapid progress. Computational methods, particularly molecular dynamics (MD) simulations, are well suited to bridge the gap between macroscopic transport properties and atomiclevel structural features, providing multiscale insights to guide materials optimization.

Historically, computational studies of SSEs have relied primarily on density functional theory (DFT). While DFT is highly accurate, its computational cost restricts accessible length and time scales. This is a critical limitation because cation diffusion and anion dynamics in SSEs are slow processes that require long simulations, and because mesoscale features-such as interfaces with cathodes or Li metal anodes and grain boundaries within the electrolyte—can strongly influence performance. The classical force fields, which use fixed functional forms and empirical parameters, often lack the flexibility and accuracy needed across diverse SSE chemistries.88 Consequently, ML-FFs that approach DFT accuracy while retaining near-classical efficiency are particularly well suited for SSE research, enabling larger systems, longer timescales, and explicit treatment of microstructure.

Development of ML-FFs was pioneered by Behler and Parrinello, who firstly used neural networks to represent potentialenergy surfaces (PES).89 In their high-dimensional neural network potential (HDNNP), the total energy E of a system is decomposed into a sum of atomic contributions.

$$E = \sum E_i$$

Each atomic energy E_i depend on the local chemical environment of atom i. To ensure translational, rotational, and permutational invariance, atomic environments are encoded as atomcentered symmetry functions (descriptors) constructed from the atomic coordinates; these descriptors are then passed to element-specific neural networks to predict E_i .

Most ML-FFs adopt this PES-decomposition framework, obtaining forces as the negative gradient of the learned with respect to atomic positions; by contrast, approaches like symmetric gradient domain machine learning (sGDML) model learn forces directly.90 The locality assumption—that each atomic contribution depends primarily on its local environment-confers strong transferability across system sizes: models can be trained on small DFT datasets and then applied to large-scale MD simulations. However, it also requires extensive sampling of diverse local environments and can neglect long-range interactions that are important in some systems.¹⁸

Based on how the atomic energy E_i is mapped from the local environment, ML-FFs can be categorized into kernelbased, linear, and neural network-based models (Fig. 3(a)). Kernel methods represent learning as Gaussian-process or kernel-ridge regression over a similarity measure between local environments; descriptors such as smooth overlap of atomic positions (SOAP) provide rotationally invariant features. Representative models include Gassian approximation potential (GAP)91 and self-learning and adaptive database (SLAD),92 and on-the-fly training/inference is available in electronic-structure packages such as Vienna ab initio Simulation Package (VASP). 93,94 Linear models, such as moment tensor potential (MTP), express the atomic energy E_i as a linear combination of basis functions, offering computational efficiency and interpretability.⁹⁵ Neural network-based models employs deep learning architectures to map local atomic energy to E_i , enabling capture of highly non-linear PES landscapes. 17,96,97 Widely used examples include the original HDNNP89 (with implementation in aenet, 98 n2p2, 99 Amp, 100 SIMPLE-NN101), end-to-end models such as Deep Potential models 96,97,102-104 and NEP, 105,106 and graph neural network-based models such as MACE, 107 NequIP, 108 DPA-3109 and CHGNet. 110 In next section, we will discuss the commonly used ML-FF models in recent literature on SSEs studies.

3.1 Commonly used ML-FF models in SSE studies

Gaussian approximation potential (GAP). The GAP employs Gaussian process regression with carefully designed local Highlight ChemComm

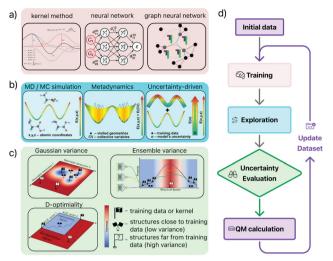


Fig. 3 Machine-learning force fields (ML-FF) and active learning. (a) Schematics of ML-FF architectures—kernel-based, neural-network, and graph neural-network models. (b) Common active-learning sampling strategies. (c) Typical uncertainty-quantification metrics. (d) A generic active-learning workflow. Kernel method illustration adapted from ref. 17. Fig. 6(B) under the terms of CC-BY-NC-ND 4.0 (https://creativecom mons.org/licenses/by-nc-nd/4.0/) from American Chemical Society, copyright 2021; neural network schematic adapted from ref. 111. Fig. 1(d) with permission from Wiley, copyright 2019; illustration of graph neural network adapted from ref. 108. Fig. 1(a) under terms of CC-BY 4.0 (https:// creativecommons.org/licenses/by/4.0/) from Springer Nature, copyright 2023. Panels b and c adapted from ref. 88. Fig. 7 and 3 under the terms of CC-BY-NC-ND 4.0 (https://creativecommons.org/licenses/by-nc-nd/4. 0/) from American Chemical Society, copyright 2024.

kernels. 17,91 In GAP, the atomic energy for a new configuration x_i is predicted as

$$E_{\rm i}(x_{\rm i}) = \sum_n K(\mathbf{d}_{\rm i}, \mathbf{d}_n)$$

where K is the kernel, **d** is descriptor, and n are the learned weights. The kernel measures the similarity (covariance) between the test descriptor \mathbf{d}_i and the *n*-th reference descriptor \mathbf{d}_n .

The SOAP descriptor¹¹² is commonly used in GAP to represent the many-body term, 113 capturing rich geometric detail while remaining smooth and differentiable. As a kernel-based ML-FF, GAP often performs strongly on small to moderate datasets relative to neural-network models. 16,17,92 Its native uncertainty estimates also enable active-learning workflows without ensemble models. However, the inference cost scales with the number of reference environments, which can limit applications to very large or highly complex systems.¹⁷

Moment rensor potential (MTP). Unlike GAP, which uses Gaussian-process kernels, the MTP represents the atomic energy E_i as a linear expansion in a systematically improvable basis constructed from moment tensors of the neighboring environment:

$$E_{\rm i} = \sum \xi_{\alpha} B_{\alpha}(\mathbf{n}_{\rm i})$$

where ξ_{α} are trainable parameters and B_{α} are the basis

functions. The basis functions B_{α} are invariant polynomials constructed from contractions of moment tensor descriptors. Similar to GAP, MTP often achieves faster training convergence from scratch than neural-network-based approaches, while retaining good accuracy and transferability. 114

Drautz later introduced the atomic cluster expansion (ACE), which expresses atomic properties (such as atom's energy) as a systematic body-order expansion using basis functions constructed from radial functions and spherical harmonics. 115 This means that contributions from one-body, two-body, three-body, and higher-order interactions are all included through a complete set of radial and angular basis functions representing the local environment. As a result, ACE offers a complete and efficient general framework for representing atomic interactions. 116 Drautz also demonstrated that many existing interatomic potentials can be viewed as special cases of the ACE formalism, 115 including the previous mentioned ML-FFs such as the MTP, SOAP-GAP, SNAP. 95,112,117 This aspect reveals ACE's broad applicability and significance in the development of ML-FFs.

Deep potential model. Handcrafted descriptors often do not generalize well across various chemical systems and typically require human intervention for specific settings and optimization. 17,97 To overcome these limitations of descriptorbased neural-network ML-FFs, Zhang et al. introduced the deep potential (DP) model as an end-to-end approach.97

In DP, the atomic energy E_i is obtained via two coupled networks: an embedding (encoding) network that maps the local atomic environment into a symmetry-preserving feature space (respecting translational, rotational, and permutational invariance), and a fitting network that converts these features into E_i . By learning the representation directly from data, DP obviates the need for hand-crafted descriptors. DP has been widely applied in SSEs studies, delivering an excellent balance between near-DFT accuracy and computational efficiency.

Neural equivariant interatomic potential (NequIP). The NequIP is an ML-FF model belong to the class of E(3)equivariant graph neural network (GNN).118,119 In GNN-based ML-FFs, a molecular or condensed-matter systems is represented as undirected graphs whose nodes are atoms and whose edges connect neighbors within a cutoff. The atomic energy E_i is obtained from the sequence of node embedding via trainable readout functions, such as,

$$E_{
m i} = \sum R_{
m t} \Big(\sigma_i^{(t)} \Big)$$

where $\sigma_i^{(t)}$ is state of each node i in layer t, R_t is the trainable readout function. In some GNN-based ML-FF architecture, an additional energy bias or scaling term $e(Z_i)$ will be introduced for each atom type. 109 Through message-passing operations, information flows along edges so the model can encode interatomic interactions.

Earlier neural network typically enforced symmetry using invariant scalar features. NequIP instead employs tensor (irreducible) features that transform equivariantly under 3D rotations and reflections, ensuring that predicted energies and

forces obey E(3) symmetry by construction. GNN-based ML-FFs such as NequIP can achieve data efficiency comparable to kernel methods while retaining neural networks' flexibility. ¹⁰⁸

GNN-based ML-FFs are considered semi-local because their layered message-passing architecture enables the capture of interactions beyond the typical cutoff distance. These models are, however, often more computationally intensive than descriptor-based or simple NN potentials due to (1) substantial communication and memory traffic from multi-hop message passing over large atomic neighborhoods, 120,121 and (2) the expensive Clebsch–Gordan tensor products required for equivariant feature coupling. Efforts to reduce this overhead include GPU-optimized implementations 22 and architectural innovations such as Allegro's strictly local message-passing scheme, which reduce the number of interactions and improves parallel scalability, 221 albeit at the cost of neglecting long-range

interactions. Alternatively, ML-FFs such as the cartesian atomic cluster expansion¹²³ (CACE) and the cartesian atomic moment potential¹²⁴ (CAMP) construct invariant features directly in the Cartesian space, eliminating the need for spherical harmonics. This Cartesian approach avoids the computationally intensive Clebsch–Gordan tensor operations, offers greater simplicity, and improved computation efficiency.¹²⁴

3.2 Active learning in ML-FFs

ML-FFs are generally unreliable under extrapolation and provide trustworthy predictions only within the domain spanned by their training data.¹⁷ As many SSE-focused ML-FFs now achieve high accuracy (*e.g.* energy errors <10 meV per atom), as shown in Table 1, performance depends increasingly on the breadth and fidelity of the training set rather than on architectural choice. Consequently, ML-FF development demands

 Table 1
 List of studies of machine learning force fields for solid-state electrolytes. AL: active learning; AIMD: ab initio molecular dynamics. Numbers in parentheses indicate GNN interaction layers

System		Year	ML-FF model	DFT level of theory	Data generation scheme	Cutoff (Å)	Energy error (meV per atom)	Force Error (eV Å ⁻¹)	Ref.
Thiophosphate	Li-P-S	2021	MTP	optB88-vdw	AIMD	5	2.07 ^a	0.09	132
			MTP	PBE	AIMD	5	3.86 ^a	0.12	133
		2024	GAP	PBE0, PBEsol, r2SCAN	AIMD	5	7.0^{b}	0.17	134
		2024	HDNNP	PBE	AIMD	7	12.7^{b}	0.24	135
			aenet	PBE	AIMD		3^b		136
	Li-Si-P-S		DeepMD		AL	6	7.57^{b}	0.12	137
	Li-P-S-B-O		MTP	PBE	AL		4.43 ^a	0.14	138
	Na-P-S			trained model			3.12^{b}		139
			DeepMD		AIMD		a		140
			MACE	PBE + D3	AIMD	$6(\times 2)$	14 ^a	0.03	141
	Na-P-S-O		DeepMD		AIMD	6	9.74^{b}_{b}	0.21	142
	Na-P-S-W		Allegro	r2SCAN	AIMD	$6.5(\times 2)$	0.38^{b}	0.03	143
LGPS-type	Li-Ge-P-S		PaiNN	PBE	AL	$5(\times 3)$	13.74^a	0.03	144
	-: (: -)		MTP	PBE	AIMD	5	2.5 ^a	0.07	133
	Li-{Ge,Si,Sn}-P-S			PBEsol,PBE	AL	6	1.33^{b}	0.08	145
LLZO-system	Li–La–Zr–O		NNP	PBE	AIMD	6	3.7^{b}	0.17	146
			NEP	PBEsol	AL	7.5	0.66^{b}	0.06	147
			DeepMD		Metadynamics	6	8.51 ^b	0.27	148
D 12:	Li-La-Zr-O-Nb		SALD	WC	AIMD	5.2917	11.7 ^a	0.26	149
Perovskite-type	Li-La-Ti-O		MTP	optB88-vdw	AIMD	5			132
MAGICON	Li-Sr-Ta-Hf-O		DeepMD		AIMD	6	ob	0.25	150
NASICON	Li-Ge-P-O		DeepMD		AL	7	5.79^{b} 1^{b}	0.26	151
KTP-type	Na-Ga-P-O-F		MTP	PBE	AL	_		0.14	152
Argyrodite	Li–P–S–Cl		MTP	PBE	AL	5	6.9 17.8^b	0.16	61
			MTP	PBE	AL	5		0.05	153
			DeepMD MTP		AL AL	8 5	1.34^{b}	0.05	63
				PBE + D3	AL AIMD	Э	7.5 ^b	0.27	154
	Li-P-S-{Cl,Br,I}		MTP MTP	optB88-vdw	AL	5	3^a	0.37	25
	Li-P-S-{Cl,B1,1} Li-P-S-Cl-O-C			optB88-vdw PBE			0.5^{a}	0.1	155 156
Halide	Li-Y-Cl		NequIP MTP		AIMD AIMD	$8(\times 4)$	1.11^{b}	0.01 0.04	132
Hanut	Li-Y-Br		MTP	optB88-vdw PBE	AIMD	_	1.11 1.05^a	0.04	
	Li-Er-Cl		MTP	PBE	AIMD	5 5	2.57^a	0.05	133 133
	Na-{Nb,Ta}-Cl		DeepMD	PBE	AIMD	8	1.6 ^a	0.03	157
	Li-Nb-Ta-Cl		GAP	PBE	AIMD	0	1.0	0.03	158
Closo-hydroborate			SLAD	PBE	AIMD	4.23	2.6 ^a	0.073	92
Cioso-nyuroborate	гі-р-п		MTP	rev-vdW-DF2,PBE,PBE-D3	AL	4.23	1 ^b	0.08	159
			DeepMD		AL	6	0.59^a	0.01	160
	Na-C-B-H		1	rev-vdW-DF2	AL	7	1.26^{b}	0.04	161
Li-N	110 O D 11		eSNAP	PBE	AL	,	0.9^a	3.77	162
T)1 1/4			GAP	PBEsol	AL AIMD	5	3.61^b	0.04	163
Li-P			MTP	PBE	AL	3	18.5^{b}	0.34	164
LIPON			NequIP	PBE	AIMD	4.3(× 6)	5.5^a	0.01	165
LII ON		2023	requir	I DE	ищи	4.3(^ 0)	3.3	0.01	103

 $[^]a$ Mean absolute error (MAE). b Root mean squared error (RMSE).

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datasets with both comprehensive configurational coverage and high-quality labels to ensure reliability across the relevant configuration space.

Training data may be drawn from existing DFT datasets—most commonly ab initio molecular dynamics (AIMD) or generated via active-learning workflows (Fig. 3(b)). AIMD is computationally expensive and can under sample rare but mechanistically important high-energy events because Boltzmann statistics overrepresent low-energy regions. Moreover, AIMD is often performed at reduced precision (e.g., coarser kpoint meshes and lower plane-wave cutoffs) relative to singlepoint calculations, which can degrade label quality. 125 Active learning offers a systematic alternative: a provisional ML-FF explores configuration space, while uncertainty or diversity criteria select configurations for DFT labeling, preserving efficiency while expanding coverage. 126

The general active learning workflow comprises four iterative stages, as shown in Fig. 3(b): training, exploring, uncertainty evaluation, and labelling. 127,128 Starting with an initial model (or ensemble) trained on an initial dataset, MD simulations use the current ML-FF to explore configuration space. When configurations exceed a predefined uncertainty threshold, they are selected for DFT calculations. The newly labelled data are then added to the training set, and the model is retrained. This approach has been formalized in automated pipelines such as DP-GEN, 128 which systematizes the DP active learning loop with minimum human intervene.

Two exploration strategies are commonly used (Fig. 3(c)): (1) conventional MD simulations or Monte Carlo (MC) sampling, typically in NPT or NVT ensembles, to sample thermodynamically accessible configurations; and (2) the metadynamics sampling, which applies bias potentials along carefully chosen collective variables (CVs) to access rare events and highenergy regions. Metadynamics offers superior exploration of rare events but requires system-specific CV selection and tuning. A recent alternative is uncertainty-driven sampling, which uses model uncertainty as a bias to steer exploration toward regions where the ML-FF is least reliable. 129

Common uncertainty metrics (Fig. 3(d)) include: (a) Gaussian Gaussian-process predictive variance, available natively in GAP-type ML-FFs; (b) D-optimality criteria, used in MTP via an extrapolation grade derived from the determinant of the information matrix; and (c) ensemble disagreement, in which multiple models with different random initializations are trained and the dispersion in predicted forces is used as the uncertainty estimate. 93,127,130 Ensemble methods are model-agnostic and generally robust, and are therefore widely adopted across ML-FF frameworks. 131

4. The application of ML-FFs in SSEs research

4.1 Identification of GSEs

Because of their amorphous character, glassy SSEs present structural challenges that require specialized

approaches for accurate atomic modeling. As shown in Fig. 4, the atomic structures can be generated either by reverse Monte Carlo (RMC) fitting to experimental data 166,167 or by meltquenching simulations. 16,168,169

The RMC method employs the Metropolis acceptance-rejection algorithm to minimize the difference between simulated and experimental features (Fig. 4(a)). However, in its basic form RMC is under-constrained and can yield chemically unreasonable networks, particularly in systems with complex bonding. A hybrid RMC-MLFF approach (RMC-DL) has been proposed to address this issue,171 using an ML-FF to assess the energetic plausibility and thereby produce more physically meaningful structures, as demonstrated by Yamada et al. 136

An alternative is to use ML-FFs directly to perform meltquench simulations (Fig. 4(b)). Due to their scalability, ML-FFs enable larger supercells and longer trajectories than AIMD while retaining near-DFT accuracy, yielding more representative amorphous networks. This strategy has been applied across multiple glassy SSE families, including halide glass, 158,172 sulfide glass, 135,137,141,142 LiPON glass, 165 amorphous LLZO systems, 148 and metal oxyhalide systems. 85-87

However, developing ML-FFs for amorphous systems is hampered by limited training data: metastable amorphous phases are largely absent from open datasets and, by extension, from pre-trained models. Recent evaluations of available pretrained ML-FFs have identified unphysical structural motifs and poor agreement with measured mechanical properties.

We recently developed ML-FF models for Li-thiophosphate GSE and Na-oxythiophosphate GSEs (Fig. 5), namely, Li₂S-SiS₂- P_2S_5 and $Na_3PS_{4-x}O_x$ systems. These ML-FFs reproduce experimental densities, structure factors, radial distribution

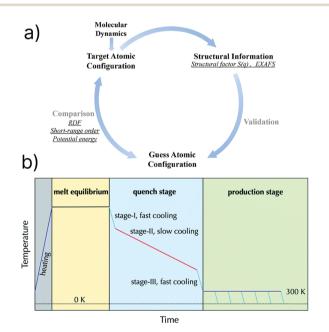


Fig. 4 Workflow of obtaining glass structures. (a) reverse Monte Carlo (RMC) method. (b) Melt-Quenching simulation. Panel (a) reprint from ref. 170 with permission from Wiley, copyright 2023; panel (b) reprint from ref. 168 with permission from The Royal Society of Chemistry, copyright 2022.

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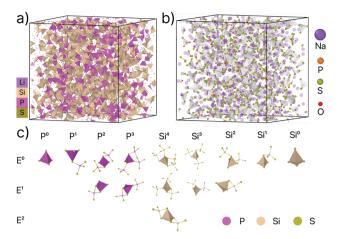


Fig. 5 Structure of obtained Li₂S–SiS₂–P₂S₅ and Na₃PS_{4–x}O_x glass through ML-FF melt-quenching simulations. (a) $60\text{Li}_2\text{S}-2\text{SiS}_2-8\text{P}_2\text{S}_5$ (b) Na₃PS_{3.85}O_{0.15}. (c) local environments of P and Si. Pictures adapted form ref. 142 with permission from American Chemical Society, copyright 2023 and ref. 137 with permission from The Royal Society of Chemistry, copyright 2024.

functions, and mechanical properties. The larger simulation cells accessible with ML-FF-driven melt-quench workflows enable robust statistics; by contrast, typical AIMD supercells (~ 100 atoms) provide insufficient sampling for reliable structural analysis.

Using these ML-FFs, we examined (1) oxygen doping in NaPSO (Na₃PS_{4-x}O_x) and (2) incorporation of a second network former in Li-thiophosphate glasses. In the Li₂S-SiS₂-P₂S₅ system, compositional scans reveal that the medium-range connectivity of short-range structural units (edge sharing, corner sharing, or isolated motifs shown in Fig. 5(a) and (c)) strongly modulates Li-ion diffusion. In Na₃PS₄- $_x$ O_x, oxygen exerts dual effects—reducing free volume *via* increased electronegativity while simultaneously enhancing network flexibility—together governing ionic transport.

4.2 Phase transition

As discussed in previous sections, many SSEs exhibit temperature-driven polymorphism, with a low-temperature phase (typically low ionic conductivity) and a high-temperature phase (typically higher conductivity). For example, LLZO adopts a high-conductivity cubic phase ($Ia\bar{3}d$) and a low-conductivity tetragonal phase (I41/acd), with a phase transition temperature around 900 K that alter Li⁺ transport by orders of magnitude. Complex hydrides, commonly transform from ordered monoclinic structures at low temperature to disordered phases at high temperature; in Na₂B₁₀H₁₀, the B₁₀H₁₀²⁻ anions undergo an order–disorder transition around \sim 373 K, enabling superionic conduction. Sulfide electrolytes (e.g., Na₃PS₄) and halide electrolytes (e.g., Li₃YCl₆) likewise display temperature-dependent phase transitions that strongly affect transport properties.

Elucidating the atomistic mechanisms of these transitions is crucial for devising strategies to stabilize high-conductivity

phases at room temperature. By delivering near-DFT accuracy at (near) classical MD cost, ML-FFs enable the long trajectories and large cells needed to capture nucleation pathways and order–disorder dynamics—capabilities beyond classical force fields and typically inaccessible to AIMD. As shown in Fig. 6(a)–(g), Shimizu *et al.* tracked the crystallization of Li_3PS_4 glass over ~ 100 ns, a timescale infeasible for AIMD. Maltsev *et al.* investigate the temperature-induces order–disorder phase transition in $\text{Li}_2\text{B}_{12}\text{H}_{12}$ (Fig. 6(h) and (i)) and $\text{LiCB}_{11}\text{H}_{12}$ and compared the effects of different exchange–correlation functionals on crystal structure and phase transition temperature.

We developed ML-FFs for the Na–C–B–H *closo*-hydroborate family and investigated the temperature-induced transition in $Na_2B_{12}H_{12}$, as shown in Fig. 6(j) and (k) The simulations reveal a martensitic, Bain-like pathway from the room-temperature monoclinic phase to a high-temperature bcc phase, accompanied by a pronounced increase in Na^+ conductivity and a reduced barrier for anion reorientation. Despite the smaller unit-cell volume of the bcc phase, the dense network of tetrahedral interstitials, together with faster anion reorientation, yields abundant, dynamically connected migration pathways that enable rapid Na^+ diffusion.

4.3 Room temperature calculation

A primary goal of computation is to quantify ionic conductivity and migration barriers at operating temperatures; reliable estimates are essential for SSE screening and optimization. Although AIMD is intrinsically accurate, it faces three major limitations: (1) short trajectories that yield large statistical uncertainties, (2) non-Arrhenius transport in many SSEs that invalidates high-temperature extrapolation, and (3) computational cost that restricts accessible length and time scales.

Statistical errors arise because AIMD runs typically span only hundreds of picoseconds, providing too few diffusion events for well-converged transport coefficients. He *et al.* showed that uncertainties in AIMD-derived transport properties can be substantial, especially for the slow diffusion characteristic of room-temperature SSEs. The Moreover, as shown in Fig. 7 and 8, the common practice of extrapolating high-T data to room temperature with an Arrhenius law is often unjustified: materials such as LGPS and Li₃YCl₆ exhibit non-Arrhenius behavior due to phase transitions, temperature-dependent diffusion mechanisms, or anion dynamics (*e.g.*, PS₄ rotational modes). In such cases, direct room-temperature calculations are preferable, as Arrhenius extrapolation can lead to errors of orders of magnitude.

4.4 Anion rotational movement

Anions play a crucial role in SSEs performance, serving both as the structural framework and as active participants in cation diffusion. In particular, the rotational dynamics of complex anions (Fig. 9) have emerged as key to understanding superionic conduction. ^{133,136,157,178–185}

We recently utilized ML-FFs to study anion dynamics in $\text{Li}_2\text{S-SiS}_2\text{-P}_2\text{S}_5$ GSE and in $\text{Na}_2\text{B}_{12}\text{H}_{12}$ SSE. For melt-quench glass models of $x\text{Li}_2\text{S-}(1-x)\text{P}_2\text{S}_5$ (x=67%, 70%, 75%), we

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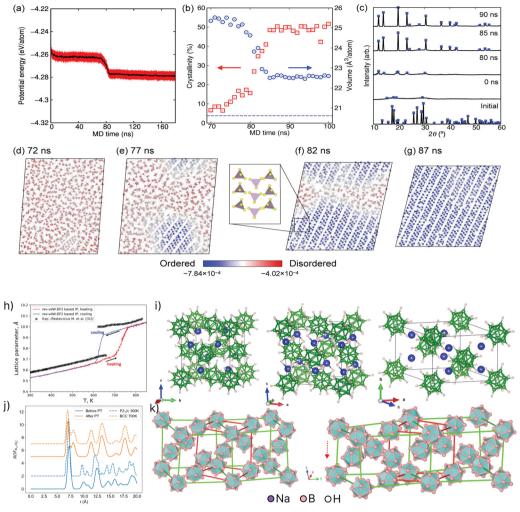


Fig. 6 Phase transition studies in SSEs. (a)-(g) Crystallization process of Li₃PS₄ GSE: (a) potential energy profile, (b) changes in crystallinity and cell volume, (c) calculated XRD pattern, (d)-(g) snapshots from 72 ns to 87 ns. (h), (i) Order-disorder transition of Li₂B₁₂H₁₂: (h) lattice constant during heating and cooling, (i) crystal structures of $\text{Li}_2\text{B}_{12}\text{H}_{12}$ cubic and monoclinic phases. (j) –(k) Phase transition in $\text{Na}_2\text{B}_{12}\text{H}_{12}$: (j) pair distribution function of $\text{Na}_2\text{B}_{12}\text{H}_{12}$ before and after phase transition, (k) illustration of Bain martensitic transition path in Na₂B₁₂H₁₂. panel a-g reprint from ref. 135 with permission form American Chemical Society, copyright 2024; panel h, i reprint from ref. 174 with permission form American Chemical Society, copyright 2023; panel j, k reprint from ref. 161 with permission form American Chemical Society, copyright 2025.

found that decreasing fractions of corner- and edge-sharing PS units, coupled with increasing isolated PS4 tetrahedra, lead to more bridging sulfur atoms, which allows for more rotational movement. In 60Li₂S-32SiS₂-8P₂S₅ GSE (Fig. 9(c)), calculations of Li⁺ diffusion and anion rotation show small-angle rotations (<20°) of PS units along the Li⁺ diffusion pathways, suggesting that enhanced anion rotational degrees of freedom can lower Li⁺ migration barriers.

We applied a similar analysis to Na₂B₁₂H₁₂. As shown in Fig. 9(d), in the low-temperature monoclinic phase, $B_{12}H_{12}^{2-}$ anions undergo slow, discrete rotations about symmetry axes (predominantly fivefold), whereas in the high-temperature bcc phase they reorient more rapidly with frequent hops between symmetry-equivalent orientations. Fitting the orientational autocorrelation to an exponential yields Arrhenius behavior (Fig. 9(e)): the reorientation rate increases by $\sim 20 \times$ in the

high-T phase, and the activation energy drops from 0.77 eV to 0.18 eV.

The paddle-wheel effect has been a subject of considerable debate in the SSEs community. 157,160,178,179 This debate stems partly from the lack of a unified definition. 178 Regardless, it is well agreed that rotational motion of anions can lower cation migration energy barriers, as illustrated in Fig. 9(a). AIMD simulations by Smith and Siegel (Fig. 9(b)) suggest that such effects can operate at room temperature in glassy electrolytes: in Li₃PS₄ glass, PS₄³⁻ tetrahedra exhibit large rotational displacements ($\sim 20-75^{\circ}$) that are temporally and spatially correlated with Li⁺ migration events. 179

However, recent ML-FF studies have deepened our understanding of anion rotation effects. Xu et al. employed MTP to investigate polyanion rotation effects across multiple SSEs systems (β -Li₃PS₄, Li₇P₃S₁₁, Li₁₀GeP₂S₁₂, Li₃ErCl₆, Li₃YBr₆) ChemComm Highlight

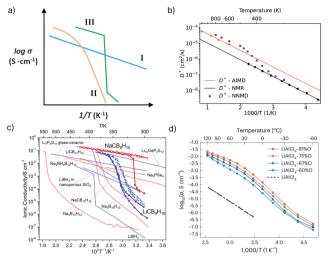


Fig. 7 Temperature-dependent ionic conductivity. (a) Schematic illustration of three types of conductivity-temperature relationships, (b)–(d) Arrhenius ionic conductivity of LGPS, closo-hydroborate SSEs, glassy LiAlCIO SSEs. Panel (a) reprinted from ref. 145 with the permission of AIP Publishing, copyright 2021; panel (b) reprinted from ref. 144 (Winter et al., 2023) under terms of CC-BY 4.0 (https://creativecommons.org/licenses/by/4.0/) from IOP Publishing, copyright 2023; panel (c) reprinted from ref. 175 with permission from Wiley, copyright 2016; panel (d) reprinted from ref. 86 with permission from Springer Nature, copyright 2023.

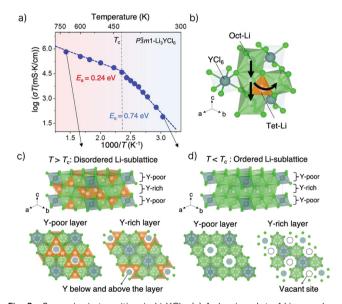


Fig. 8 Super-ionic transition in Li_3YCl_6 . (a) Arrhenius plot of Li+ conductivity, (b) Li⁺ migration pathways in Li₃YCl₆, (c) structure of partially occupied Li⁺ sites in Li₃YCl₆, (d) structure of ordered Li⁺ sublattice in Li₃YCl₆. Reprinted from ref. 176 under the terms of CC-BY-NC-ND 4.0 (https://creativecommons.org/licenses/by-nc-nd/4.0/) from Wiley, copyright 2023.

using μs -scale simulations.¹³³ Surprisingly, they found that only $\text{Li}_7 P_3 S_{11}$ shows significant polyanion rotation at room temperature, and that rotational $[PS_4]^{3-}$ groups correlate weakly and negatively with Li^+ diffusion, challenging the notion

of pervasive paddle-wheel-assisted transport in crystalline systems at ambient conditions.

The current understanding suggests that the paddle-wheel effect requires careful consideration of several factors: (1) the need for a strict definition based on quantitative metrics rather than qualitative observations, ¹⁷⁸ and (2) understanding that its importance varies significantly between different materials and temperature regimes. ML-FF simulations provide the temporal and spatial resolution needed to disentangle coupled cationanion dynamics and to assess these effects rigorously.

4.5 Interface study

ML-FFs are particularly well suited to interface studies for three reasons: (1) they enable simulations of large interfacial regions with adequate statistical sampling; (2) they access the extended time scales required to capture slow interfacial processes; and (3) they explicitly describe bond breaking and formation, allowing direct treatment of interfacial chemistry. These capabilities make ML-FFs especially valuable for elucidating grain-boundary transport, surface reactions, and solid-solid interfaces in SSEs.

Grain boundary. Grain-boundaries (GBs) effects vary across SSEs systems. In oxide-based SSEs, GBs typically impede Li⁺ transport and potentially promote dendrite growth, whereas in systems such as Li₃InCl₆ and Li₃PS₄ they often enhance Li diffusion due to local amorphization. ML-FF enables detailed, atomic-scale investigation of GBs structures and transport properties that would be prohibitively expensive with AIMD. As shown in Fig. 10, Ou *et al.* modeled Σ 3 and Σ 5 GBs in argyrodite Li₆PS₅Cl⁶¹ and showed that the opening of Licoordinated cages at GBs strongly influences Li⁺ diffusion (Fig. 10(b)). You *et al.* recently utilized an ML-FF to study the vertical and horizontal GBs in LLZO garnet-type SSEs; in that case, GB amorphization hinders Li transport but suppresses Li aggregation and inhibits dendrite formation. ¹⁸⁶

Collectively, recent computational and experimental and experimental studies have demonstrated that grain boundary engineering can be employed to optimize the performance of SSE by enhancing ionic conductivity and suppressing dendrite formation. Specific strategies include inducing amorphization and increasing vacancy concentrations at GBs. 138,187 By elucidating the atomic-level structure-transport relationships at interfaces, researchers can design targeted synthesis and post-processing approaches to minimize GB resistance. This progress presents both opportunities and challenges for ML-FF modelling. One on hand, ML-FFs enable simulations involving thousands of atoms with near-DFT accuracy. On the other hand, complex atomic environments, composed of metastable amorphous and defect-rich phases, pose significant challenges for data-driven ML-FF approaches. Addressing these challenges requires the development of ML-FF models with exceptional generalizability, supported by comprehensive datasets that capture the full spectrum of structural diversity.

SSE-surface reactions. Surface stability is crucial for SSEs processing and long-term performance. Li *et al.* used an ML-FF to study gas–solid reaction dynamics on Li₆PS₅Cl surfaces

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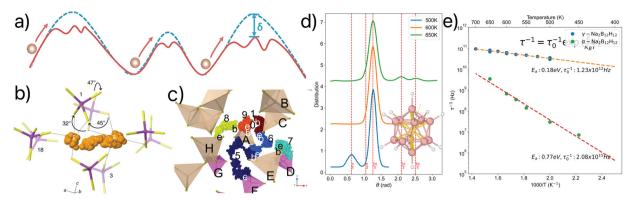


Fig. 9 Anion rotation in SSEs. (a) Illustration of the effect of anion rotation on lowering cation migration energy barrier, (b) illustration of the coupling of cation transport with the reorientation of anions in LPS glass, (c) Li⁺ diffusion trajectories in $60Li_2S-32SiS_2-8P_2S_5$ glass over 30 ps, with PS₄ (purple) and SiS₄ (brown) tetrahedra, (d) anion rotation angle and axis in low-temperature Na₂B₁₂H₁₂ phases, (e) Arrhenius plot of anion reorientational speed in high-temperature (γ) phase and low-temperature (α) Na₂B₁₂H₁₂ phase. Panel (a) adapted from abstract image of ref. 180 with permission from Elsevier, copyright 2024; panel (b) reprinted from ref. 179 (Smith *et al.*, 2020) under terms of CC-BY 4.0 (https://creativecommons.org/licenses/by/4.0/); panel (c) reprinted from ref. 137 with permission form American Chemical Society, copyright 2024; panels (d)–(e) reprinted from ref. 161 with permission form American Chemical Society, copyright 2025.

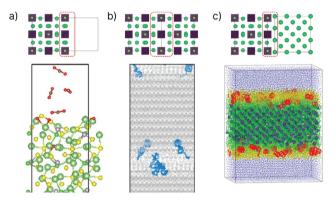


Fig. 10 Solid-state electrolyte interface schematics (top row) and atomic models in argyrodite SSEs (bottom row): (a) surface of argyrodite. (b) Argyrodite Σ 5 grain boundary model and Li diffusion trajectory (blue). (c) Argyrodite|Li metal interface after 90 ps. Top row schematics from ref. 13 with permission from Springer Nature, copyright 2025; bottom models adapted from ref. 156 with permission form American Chemical Society, copyright 2025, ref. 61 with permission form American Physical Society, copyright 2024, and ref. 153 with permission form American Chemical Society, copyright 2024, respectively.

under CO_2 and mixed CO_2/O_2 atmospheres (Fig. 10(a)). ¹⁵⁶ Their NequIP model enabled nanosecond-scale simulations that revealed detailed reaction mechanisms impossible to AIMD approaches.

The study demonstrated that in pure CO₂, the surface evolves toward Li₂CO₂S *via* C–S bond formation, whereas in CO₂/O₂ mixtures O₂-mediated pathways favor Li₂CO₃. These insights clarify how ambient gases govern surface chemistry during synthesis and operation, informing atmospheric processing conditions and protective-coating strategies. More broadly, the ability to model gas–solid reactions with near-chemical accuracy over extended timescales provides a powerful route to dissect SSE degradation mechanisms.

Metal-SSE interfaces. The solid electrolyte interphase (SEI) is a critical component of ASSBs, yet most studies lack the

atomic-level resolution necessary to elucidate reaction pathways and structural evolution. Hence, it is important to study and understand the reaction of SEI in SSEs. Using ML-FF, Ren et al. studied the SEI formation at a β -Li $_3PS_4/Li$ -meal contact at 300 K. 188 Their MD simulations reveal a 4 stage process: (1) a brief interdiffusion stage in which interfacial S and P migrate toward the Li metal while Li migrates into the electrolyte, producing an amorphous SEI with a mutual diffusion depth up to \sim 41 Å; (2) nucleation of a crystalline interphase at the interface; (3) anisotropic growth that proceeds rapidly parallel to the Li surface with comparatively slow thickening and, once the surface is covered, expansion toward the SSE; and (4) a quasi-steady-state regime in which SEI thickening slows and the structure stabilizes.

Most current ML-FFs cannot capture electrochemical charging/discharging, limiting their ability to model dendrite nucleation or SEI evolution under redox driving forces. To address this gap, Hu *et al.* introduced the DP-QEq framework, which enables constant-charge or constant-potential simulations by decomposing the total energy into a short-range part learned by the ML-FF and a long-range part treated *via* charge equilibration (QEq). This approach allows explicit control of electrochemical driving forces at SSE-electrode interfaces, enabling mechanistic studies of SEI formation, growth, and stability.

5. Future perspective and conclusion

Transferability between different studies

Transferability remains a major challenge for ML-FFs, arising from both architectural differences and heterogeneity in training data. Although numerous SSE-focused ML-FFs and large datasets now exist, models trained in one framework are rarely usable without modification in another.

Data are generated at different levels of precision, using different DFT software, exchange-correlation functionals,

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Hubbard-U corrections, and van der Waals interactions, complicating cross-study reuse. The explored configurations spaces also differ substantially. For instance, in Table 1, a ternary Li-P-S ML-FF could be trained on crystalline β-Li₃PS₄ alone or with different degrees of disorder, 133 on other Li₃PS₄ polymorphs or related thiophosphates, ¹³⁴ on decomposition products such as Li₂S and Li₃P, 145 on amorphous/glassy structures, 137 or on Li-metal interfaces. 188 The adoption of active learning in SSEs studies also depends heavily on software ecosystems, with MTP and DP showing greater usage due to their integration with established software packages, such as DP-GEN and MLIP. 120,127,128,191 Moreover, active-learning datasets are themselves conditioned by the underlying ML-FF architecture, further hindering transfer between models. 192

As a result, new systems often require training from scratch. A pragmatic path forward is a pre-trained "foundation model" plus fine-tuning: large models trained on broad datasets learn transferable atomic-environment embeddings, which can then be adapted efficiently to target chemistries with system-specific data, often achieving higher accuracy and computational efficiency. 130,193-196

From bulk to interfaces

Most ML-FF studies focus on bulk SSE materials, but there is growing need for a deeper understanding of interfaces: SSEmetal anode, SSE-high voltage cathode, and SSE-SSE interfaces. To date, relatively few ML-FF studies have examined interfaces; most target SSE-metal and SSE-SSE systems, with only limited studies on SSE-cathode interfaces.

From the ML-FF development perspective, SSE-cathode interfaces are especially challenging because they introduce additional elements and compounds. Many ML-FFs use element-specific descriptors or networks to improve accuracy, so expanding the chemical space increases data requirements and can slow inference. Moreover, transition-metal species such as Ni, Co, and Mn add complexity through variable oxidation states, strong correlation, and spin degrees of freedom. 197,198

Understanding how interfaces behave under realistic mechanical and electrochemical conditions is critical for advancing solid-state systems. This includes accounting for GPa-level stress¹⁹⁹ buildup at the interfaces and the variations in chemical potentials that occur during electrochemical cycling.⁷³ Recent advances in ML-FFs are beginning to address these challenges. These developments include models designed to capture long-range interactions (e.g., 4G-HDNNP, 200 DPLR, 201 Latent Ewald Summation 202) as well as those designed to predict additional scalar and tensor properties, such as charge states (e.g., CHGNET¹¹⁰), spin (e.g., DeepSPIN¹⁹⁷), and dynamic charge-related properties such as the Born effective charge (e.g., Equivar, 203 CACE-LR 204). While these advanced models enable more accurate and physically informed descriptions of interfacial phenomena, their performance remains fundamentally limited by the quality and diversity of their training data. This reveals the urgent need for active learning workflows and pre-trained models to efficiently sample, explore, and generalize across these highly complex systems. 196

In conclusion, ML-FFs have emerged as a powerful computational tool for studying solid-state electrolytes, bridging the gap between the first-principles accuracy and the large-scale, long-timescale simulations required to understand these complex materials. As revealed in this review, the application of ML-FFs has already provided critical atomic-level insights across a wide range of crystalline and glassy SSEs, from accurate modeling of structures and mechanical properties to the study of complex transport dynamics and reactions. The growing interest in the interfacial phenomena and studies under more realistic mechanical and electrochemical conditions demands even more accurate ML-FF models and comprehensive, diverse training datasets. Recent advances, including the development of long-range models, foundation models, and active learning workflows, enables promising pathways towards a deeper atomic-level understanding of solid electrolytes, ultimately accelerating the discovery and optimization of robust, high-performance materials for next generation allsolid-state batteries.

Conflicts of interest

There are no conflicts to declare.

Data availability

No primary research results, software or code have been included, and no new data were generated or analysed as part of this review.

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