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Machine-learning Interatomic Potentials for Interfaces in All-solid-state Batteries: Perspectives on Training Data, Model Selection, and Validation

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ABSTRACT

Interfaces play a pivotal role in dictating the performance and reliability of all-solid-state batteries (ASSBs), where complex electro-chemo-mechanical phenomena at grain boundaries (GBs) and interfaces can lead to degradation and failure. Traditional atomistic simulation methods, such as first-principles calculations and classical molecular dynamics, face limitations in modeling these interfaces due to either high computational cost or insufficient transferability to the diverse atomic environments evolving at interfaces. Machine-learning interatomic potentials (MLIPs) have emerged as a transformative approach, enabling large-scale, high-accuracy simulations of disordered and chemically complex systems by leveraging the predictability of machine learning models trained on first-principles data. Recent applications of MLIPs have demonstrated their ability to capture intricate behaviors at ASSB interfaces, including ion transport, interfacial evolution, and degradation mechanisms, with accuracy and efficiency unattainable by conventional methods. This prospective paper presents comprehensive analysis and practical guidance for MLIP development for GBs and interfaces in ASSBs, with a focus on three key pillars: data generation, model selection, and validation. We review the current state of MLIP applications for GBs and interfaces in both general and ASSB-specific materials, highlighting best practices and challenges in constructing diverse and representative datasets, choosing appropriate machine learning architectures, and rigorously validating model performance. We also discuss emerging strategies and opportunities for improved reliability and efficiency of MLIPs to simulate realistic interfaces in ASSBs.

Keywords: machine learning, simulation, energy storage, grain boundaries, interface

1. Introduction

Interfaces critically dictate the performance and durability of all-solid-state batteries (ASSBs). Electro-chemo-mechanical instabilities can occur at internal (homogeneous) interfaces—i.e., grain boundaries (GBs)—as well as external (heterogeneous) interfaces between different components in composite materials, resulting in corrosion, mechanical failure, and degradation of device performance.¹ At GBs, the higher resistance of Li transport compared to bulk grain interiors degrades rate performance. Elemental segregations at GBs change the intrinsic properties of solid electrolyte (SE) materials,² and Li metal formation and dendrite penetration can lead to capacity fade, short circuiting, and cell failure.³ SE interfaces can be (electro)chemically unstable against cathode active materials (CAMs) and Li metal anodes, resulting in decomposition and formation of interphases with increased charge transfer resistance and promoting dendrite initiation or non-uniform Li plating.¹ During cycling, electrochemical reactions are coupled with mechanical responses from CAM volume changes upon (de)lithiation, which can initiate crack formation and propagation at GBs and interfaces and lead to loss of mechanical contact and electrochemically active sites.⁴ In addition, co-sintering of cathode composite materials at elevated temperatures, which is essential to achieve intimate contact between the solid components during fabrication, can accelerate the chemical degradation at interfaces.⁵ Fundamental understanding of coupled electro-chemo-mechanical interfacial phenomena is therefore imperative for rational design and practical realization of ASSBs.

From a computational perspective, atomistic simulations based on quantum mechanical modeling and molecular dynamics (MD) have been widely used to investigate interfacial phenomena, including thermodynamic stability, dynamic evolution, and ion transport kinetics.⁶ These tools have proven useful, but each simulation method comes with unique tradeoffs for modeling interfaces. First-principles simulations carry the accuracy of quantum mechanics, but simulations are limited in length and time scales (typically ~ 100 ps and 1000 atoms) due to poor computational scaling ($\mathcal{O}(N^3)$). By contrast, complex interfaces often require thousands of atoms for proper representation, and properties such as transport can be difficult to converge in limited time. In contrast, classical MD approaches using empirical interatomic potentials can easily access large-scale simulations with millions of atoms. However, empirical potentials are typically based on pre-defined mathematical formulae fitted for well-defined chemical environments and thus suffer from transferability issues when encountering the far more diverse range of local atomic environments at interfaces. This limitation becomes especially problematic when chemical reactions, local ordering/disordering, or formation of secondary phases are involved during interfacial evolution and degradation.

To overcome these limitations, machine-learning interatomic potentials (MLIPs) have been widely adopted in the past decade.⁷ They utilize the superior predictability of machine learning (ML) regressors, including deep learning models, upon proper optimization based on training data from first-principles calculations. These models can predict energies, forces, and virial tensors of a wide variety of atomic structures with accuracy similar to a density functional theory (DFT) reference. At the same time, MLIPs enable large-scale MD simulations with significantly reduced computational cost compared to DFT. MLIPs have garnered significant attention after Behler and Parrinello (BP)'s high-dimensional neural-network potential (NNP) approach using atom-centered radial and angular symmetry functions for describing local atomic environments that satisfy translational, rotational, and permutational invariance.⁸ Thereafter, various MLIP approaches were proposed and implemented, including non-linear ML models with atomic descriptors, linear regressors with basis functions, and message passing graph neural networks (GNNs);⁹ see previous literature for a comprehensive review for different types of MLIPs.^{7,9-13} These approaches have demonstrated effectiveness in applications with varied degrees of required complexity, accuracy, and computational speed.

Energy storage materials is one field where MLIPs are being actively applied to investigate inherent properties that require large-scale atomistic modeling and long simulation times.¹³ A number of studies have utilized MLIPs to predict structural and thermodynamic properties, Li intercalation behaviors, and ion transport characteristics across a range of anode, SE, and cathode materials in ASSBs.¹⁴ Notably, most of these material systems contain a degree of disorder that significantly affects their properties or is central to their functionality. For example, Li mobilities in $\text{Li}_2\text{S-P}_2\text{S}_5$ glasses are altered by local polyanion structures with configurational disorder.¹⁵ Trends in thermodynamics and voltage profiles in CAMs are likewise affected by configurational disorder in terms of the distribution of intercalated Li ions at different charge states.¹⁶ Chemical disorder in SEs with anion anti-site defects (e.g., $\text{Li}_6\text{PS}_5\text{Cl}$) also lead to increases in ionic conductivity.¹⁷ The advantages of MLIPs for simulating such disordered systems are noteworthy. MLIPs can navigate both compositional and configurational disorders, addressing the requirement of large atomistic models to encompass local structural and chemical diversities and enabling accurate and unbiased exploration of intrinsic properties. The relative computational efficiency of MLIPs can also overcome time scale issues. Ion transport in solid-state energy storage materials can in general be slow, particularly at low temperatures; however, this is exacerbated by atomic disorder, which induces an additional dispersion of intrinsic local diffusion time scales that necessitates even longer simulations for sufficient statistical convergence. As a result, MLIPs can capture more subtle trends in transport than DFT-based approaches. For instance, non-Arrhenius behavior, which is commonly observed in many superionic conductors at low temperatures, can be accurately captured by MLIP-driven large-scale MD

simulations with enhanced statistics.¹⁸ Hence, comparisons to experiments can be improved, leading to more appropriate design principles for optimization of battery performance.

Beyond bulk disordered systems, GBs and interfaces in ASSBs present a next frontier in simulation complexity that are likewise well suited to the strengths of MLIPs.¹⁹ During battery operation, interfacial evolution driven by electro-chemo-mechanical phenomena induces spatiotemporal changes in local structures at boundary regions, characterized by varying degrees of disorder, defect formation, and heterogeneity. Such alterations can lead to side reactions, interphase formation, sluggish ion transport, mechanical fracture, and the accumulation of dead lithium. These interface phenomena in ASSBs are particularly complex because of 1) chemical complexity involving at least ternary and higher-order materials, 2) structural complexity arising from lattice disorders, distortions, and defects, 3) increased likelihood of side reactions stemming from a high-dimensional phase space with a diverse set of potential secondary phases, and 4) mechanical defects and fractures caused by changes in electrode volume during cycling. These complexities lead to a significantly wide range of spatiotemporal variations in compositions and configurations, resulting in additional requirements for the proper development of MLIPs for interfaces. Interface-ready ML models, therefore, must have sufficient expressive power to encode the vast array of diverse and reactive atomic environments, including both chemical and structural complexity. Consequently, careful strategies for data generation, model selection, and validation are essential. Especially, to ensure reliable performance, ML models need to be validated not only against energy and force errors (and also for stress and magnetic moment if trained), but also against relevant structural, vibrational, and dynamical characteristics at interfaces.

Thus far, several review and perspective papers have provided valuable summaries and opinions of the applications of MLIPs for battery materials.^{13,14,19-28} However, few have discussed in detail how MLIPs have been developed and validated and what are lessons learned from the examples, specifically for interfaces in ASSBs. The aim of this prospective paper is to offer guidance in this area, with particular emphasis on appropriate strategies and future directions in developing MLIPs suitable for GBs and interfaces in ASSBs and for computing battery-relevant properties. We provide a review of existing literature applying MLIPs for GBs and interfaces in ASSB materials (as well as general elemental and binary materials). Based on these examples, we discuss strategies for the development of reliable MLIPs along three aspects: *data generation*, *model selection*, and *validation*. Finally, we offer perspectives on further development of MLIPs, including accessing more realistic conditions, using more efficient training strategies, and improving reliability.

2. Modeling Grain Boundaries with ML Potentials

The macroscopic properties of materials are significantly governed by their microstructures, defined by the types and distributions of phases, grain size and orientation, porosity, and one-dimensional and planar defects. GBs are a key feature in ASSBs, often exhibiting high ionic resistance and serving as preferential sites for Li accumulation and dendritic growth, which can accelerate capacity fade and trigger short circuiting and cell failure.^{2,3,29-32} Here, we first review how MLIPs have been developed to study GBs and microstructures of general materials (elemental and binary compounds) by focusing on data generation, model selection, and validation, and discuss their implications for simulating GBs in ASSBs. Next, we analyze existing examples of MLIPs for GBs in ASSB materials and discuss lessons learned.

MLIPs for GBs in general materials and implications for their development. MLIPs have been successfully applied to a variety of materials, primarily focusing on elemental and binary systems, to investigate their microstructures. For example, they have been used to simulate properties at GBs, including structural and mechanical characteristics such as segregation and cracking, thermodynamic properties, thermal conductivity, as well as stacking faults and dislocations.³³⁻⁴⁶ The types of required training data depend on the target application. Purja Pun et al. simulated dislocation dynamics in bulk Al and crack growth along Al GBs.^{33,34} For this, they trained a physics-informed neural network (PINN) potential using a training dataset including surface structures, GB models, and a stacking fault model. L. Zhang et al. used structure data for surfaces, GBs, and dislocations to train various ML models, including Gaussian approximation potentials (GAP), atomic cluster expansion (ACE), moment tensor potential (MTP), and NNP, and performed simulations for dislocations and crack formation in bulk Fe (Figure 1a).³⁹ Yokoi et al. and B. Zhang et al. trained NNP models for Ge, Si, AlO₂, and CdTe and an ACE model for Ti, respectively, to simulate structural and thermodynamic properties at GBs.^{37,38,41,45,46} These studies explicitly used GB structures derived from DFT to providing detailed atomic configurations in the dataset. This approach is suitable for elemental and binary systems with relatively simple and small GBs, such as those containing 120 atoms or fewer for systems with one or two elements,^{33,34,37,38,45,46} which do not require significant computing resources for DFT calculations. For more complex systems with three elements, one study used AIMD-generated GB structures to train a NNP model (DeePMD⁴⁷) for a $\Sigma 5(120)$ symmetric-tilt GB in CdPbBr₃, investigating its geometric changes during sliding and distortion.⁴⁸ This could be feasible due to the relatively small size of the $\Sigma 5$ GB with 200 atoms and the study's focus on a single GB.

However, GBs in multicomponent compounds are chemically and structurally more complex and require larger atomic models than in elemental/binary systems and cubic perovskites, owing to less well-defined atomic lattices and a greater tendency for structural and chemical disorder. This increased complexity

leads to higher computational costs for DFT and AIMD. Moreover, the main advantage of MLIP is their ability to perform accurate large-scale MD for complex GB geometries that are beyond the practical scale limits of AIMD. Hence, MLIPs need good transferability for diverse GBs, and including GB structures

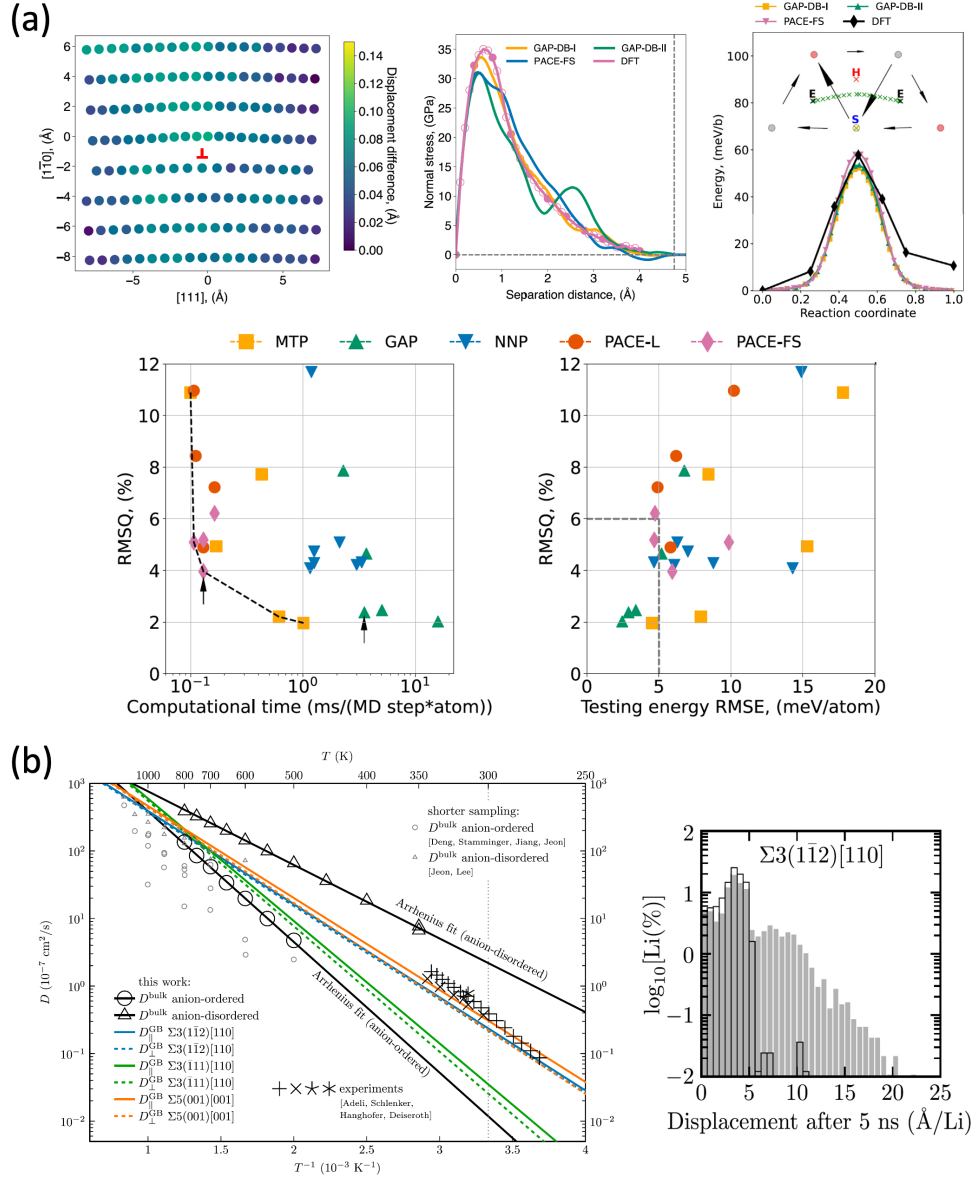


Figure 1. (a) (top) Validation of MLIP models for Fe by benchmarking the predicted dislocation (left; GAP-DB-I model), traction-separation curve for (100) plane (middle), and the Peierls barrier for a short screw dislocation (right) against DFT calculations. (bottom) Performance tests for MLIPs by comparing the root mean squared quality (RMSQ) to the computation time (left) and the root mean squared error (RMSE) for energy (right). The RMSQ is calculated by the errors for lattice parameters, elastic constants, vacation formation energies, surface energies, and stacking fault energies. Reproduced with permission from Ref.39 (L. Zhang et al.) Copyright 2024 Elsevier B.V. (b) Arrhenius plots for Li diffusion at $\Sigma 3$ and $\Sigma 5$ GBs (normal and parallel to GB plane) as well as anion-ordered and disordered bulk structures of $\text{Li}_6\text{PS}_5\text{Cl}$ compared to experimental measurements (left) and the distribution of the Li displacements in a $\Sigma 3$ GB model (gray shaded) in comparison with that in the anion-ordered bulk model (black edged) (right). Reproduced with permission from Ref.53 (Y. Ou et al.) Copyright 2024 American Physical Society.

from DFT alone cannot ensure this transferability that DFT cannot practically explore. Therefore, novel dataset generation strategies are needed to provide sufficient structural information for various GBs, enabling transferability without relying solely on DFT-derived GB structures. This aspect is especially important for developing MLIPs for GBs and interfaces of ASSBs, **which are complex in compositions and configurations with typically at least three elements.**

There are some examples of training MLIPs without explicitly feeding DFT-calculated GBs or dislocation data. Ito et al. developed a MTP model for α -Fe GBs without data for GB structures and 1D/2D defects.⁴⁰ They performed structure generation considering all crystal space groups with up to 10 atoms using the RandSpg algorithm,⁴⁹ which underwent structural variations for diverse atomic environments. The MLIP was verified by benchmarking the properties of various GBs with coincidence-site lattice (CSL) sigma numbers up to 11 against DFT, including atomic energy and force errors, GB and surface energies, 2D Peierls potential for screw dislocation, and general stacking fault energy, suggesting good transferability even without explicitly including related structures into the dataset. L. Zhang et al. found that dislocation core structures and Peierls barriers can be accurately predicted even without dislocation data because γ -surface data contains sufficient information of dislocation structures.³⁹ Likewise, Li et al. developed a spectral neighbor analysis potential (SNAP) model for the Ni-Mo system, incorporating surface, alloy, and intermetallic data but without GB data,⁴² which was used to investigate strengthening mechanisms in Ni microstructures.⁴³ Zhang and Li's studies imply that surface data may be sufficient to provide information for structural variation at GBs as well as 1D/2D microstructural defects. We presume that these training protocols can be valid for elemental and binary systems due to their structural simplicity. However, further tests may be needed to address their generalizability for ternary or quaternary compounds for ASSBs. The GB configurations of these materials are often highly disordered and evolve spatiotemporally with the extensive range of local compositional and structural variabilities. Surface and crystalline structures from all space groups alone may not be sufficient to represent the broad configurational variability at these complex GBs. Therefore, additional structural data such as amorphous/random structures as well as disordered and distorted lattices with off-stoichiometries are required to adequately capture this complexity.

Selection of a proper ML model is another important requirement to develop suitable MLIPs. One needs to consider the degree of model complexity required to correctly describe the atomic structures and potential energy surface. A previous study for benchmarking bulk elemental systems with GAP, MTP, NNP, and quadratic SNAP suggested that all methods can predict accurate energies and material properties similar to DFT calculations.¹⁰ In general, GAP shows the lowest error but with the highest computational cost and MTP shows a balanced performance between accuracy and computational cost. L.

Zhang et al. expanded this benchmarking to MLIPs for dislocations, cracks, and GBs of Fe with GAP, MTP, NNP, and ACE models and also found similar results (Figure 1a): all models show quantum-level accuracy for energies and properties, although the MTP model can be an order of magnitude faster than the GAP model with similar errors for properties, whereas the ACE model shows about twice higher error but an order of magnitude faster computation than the MTP model.³⁹ In addition, all MLIPs introduced in the aforementioned literature were developed based on relatively simple ML models utilizing local atomic descriptors and basis as input features, unlike complex state-of-the-art GNN-based models. These models were able to precisely predict the local characteristics and properties at GBs and 1D/2D mechanical defects for elemental/binary systems and demonstrated viability to model simple GBs in ternary compounds, such as a perovskite CdPbBr₃.⁴⁸ Thus, fast and accurate models like GAP, MTP, and NNP will be suitable to study the GBs of elemental and binary systems, as well as relatively simple GBs for ternary compounds.

The development of reliable MLIPs requires thorough validation. The most basic validation metrics are energy and force errors (and also stress if applicable) compared to DFT calculations using atomic snapshots from ML-driven MD (MLMD) trajectories. Benchmarking relevant properties of materials and systems against DFT/AIMD is also critical, e.g., radial distribution functions (RDFs) and Li diffusivities for studies regarding structure-transport relationship as well as vibrational density of states (vDOS) for proper dynamic behaviors of atoms,⁵⁰ since energy and force errors alone do not necessarily indicate accurate property prediction. For example, predicting thermodynamic properties requires calculating the phonon density of states (pDOS) using the dynamical matrix (i.e., Hessian matrix, which is the second-order partial derivatives of the total energy). Accurate prediction of atomic forces (the first-order partial derivatives of the total energy) does not necessarily guarantee the accuracy of the dynamical matrix and the resulting pDOS. Previous reports have been validating their MLIPs by predicting relevant properties. Purja Pun et al. benchmarked their model for Al using stacking fault, dislocation, and surface energies as well as basic properties of bulk Al against DFT and applied it to study crack propagation at Al GBs.³⁴ Yokoi et al. benchmarked their model using GB energies and partial pDOS for GB atoms and used it to predict vibrational entropies and free energies of Al GBs at finite temperatures.³⁷ L. Zhang et al. tested their model by dislocations, traction-separation curves, and the Peierls barrier to validate their models for mechanical response simulations (Figure 1a top panel).³⁹ Especially, they adopted a quality factor, which is the root-mean squared errors (RMSEs) for materials properties including surface energies, to evaluate their models (Figure 1a bottom panel). They found that the quality factor improves by lowering the energy RMSE in a relatively large energy error regime, but it is not further improved with additional decrease in energy error. These examples imply the importance of testing the trained model by

benchmarking relevant properties, including structural and vibrational characteristics, against DFT and AIMD for successful applications.

MLIPs for GBs in ASSB materials. MLIPs have been extensively utilized for bulk materials in ASSBs to study ion transport and structural characteristics in SEs.¹⁴ However, their application to GBs in ASSBs has only begun to emerge in recent years (see Table 1 for a summary of literature). It was 2019 when one of early publications appears by Deng et al. demonstrating the application of a SNAP model to GBs of Li_3N superionic conductor, showing orientation-dependent Li diffusion in GBs.⁵¹ The model was trained using unit cells with various strains and snapshots from AIMD trajectories at 400 – 1200 K. The predicted GB energies are similar to DFT results even without including surface and GB structures in the training dataset, which may be because the GB configurations are simple without disorder (twist $\Sigma 4[1000]$ and $\Sigma 7[0001]$ GBs). Sergeev et al. developed a MTP model to simulate Li whiskers formation during electrodeposition in Li metal anode, suggesting that Li deposition and diffusion proceed more rapidly along GBs than within the bulk region.⁵² They trained the model using bcc and liquid Li structures with three different densities, and benchmarked their model by comparing predicted lattice parameter, formation energies of point defects, and melting temperature against DFT results. The model was not validated by benchmarking GB-related properties against DFT. Nevertheless, the inclusion of liquid structures into the dataset, which are completely disordered structures that can provide diverse variation of local structures at GBs, may enable simulations for amorphous GBs.

Table 1. List of literature utilizing MLIPs for investigating GBs in ASSB materials.

Year	MLIP model	System and purpose	Ref.
2019	SNAP	Orientation-dependent Li diffusion in $\alpha\text{-Li}_3\text{N}$ GBs	Z. Deng ⁵¹
2022	MTP	Li whiskers formation during electrodeposition in Li metal GBs	A.V. Sergeev ⁵²
2022	NNP	Li transport at GBs and fracture/phase stability in $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$	K. Kim ⁶² , S. Yuan ⁷²
2023	MTP	Topological effects on Li transport in $\beta\text{-Li}_3\text{PS}_4$ GBs	R. Jalem ⁵⁵
2023	MTP	Diffusivities and structural evolution in $(\text{Li}_{(2/3)y}\text{Sr}_{1-y})(\text{Ta}_{0.334}\text{Nb}_{0.347}\text{Zr}_{0.211}\text{Sn}_{0.108})\text{O}_{3-\delta}$ ($y = 9/16$) GBs	S.-T. Ko ⁶⁰
2023	MTP	Li transport and structural evolution in $\text{Li}_{0.375}\text{Sr}_{0.44375}\text{Ta}_{0.75}\text{Zr}_{0.25}\text{O}_3$ GBs	T. Lee ⁶¹
2024	MTP	Li transport in $\text{Li}_6\text{PS}_5\text{Cl}$ GBs with anion disorder	Y. Ou ⁵³
2024	MTP	Li accumulation and transport in $\text{Li}_6\text{PS}_5\text{Cl}$ GBs	J.H. Kim ⁵⁴

Several studies have employed MLIPs to investigate GBs in SEs with more than two elements. Ou et al. developed a MTP model to simulate Li diffusion at GBs in sulfide $\text{Li}_6\text{PS}_5\text{Cl}$, showing that Li transport at GBs can be enhanced due to the Li-cage opening effect compared to anion-ordered bulk structure whereas it will be slower compared to anion-disordered bulk structures (Figure 1b).⁵³ They trained the MLIP using three simple GBs (two tilt $\Sigma 3$ and one twist $\Sigma 5$ GBs with the number of atoms 312 or smaller) as well as anion-ordered/disordered bulk unit cells. While they did not test the RMSEs for energies and forces for

GB models, the predicted RDF for $\Sigma 5$ GB model is comparable to that from AIMD and the calculated Li transport parameters in bulk and at GBs are in a similar range to those from experiments for polycrystalline samples. Kim et al. also developed MTP models for $\text{Li}_6\text{PS}_5\text{X}$ ($\text{X} = \text{Cl}, \text{Br}, \text{or I}$) SEs and suggested that Li accumulation occurs around $\Sigma 5$ GB up to 20 Å from the GB center and it reduces ionic diffusivity, leading to a gradual change in diffusivities from GBs to grain interior.⁵⁴ They generated the training dataset by using $\Sigma 5$ GB model and two (021) surfaces as well as amorphous models and site-disordered bulk unit cells with strains to cover diverse atomic environments. They tested MTPs for three different levels of DFT theories, i.e, PBE, PBE-D3(BJ), and optB88-vdW, and found that optB88-vdW leads to the most accurate bulk Li conductivities compared to experiment. They validated their model by benchmarking Li diffusivities at GBs against experimental value in addition to the energy and force errors. Jalem et al. developed a MTP model for $\beta\text{-Li}_3\text{PS}_4$ using an active learning (AL) method and revealed faster diffusion in GBs than bulk crystalline phase as well as topological effects on Li transport in twist/tilt GBs by analyzing Li sub-networks.⁵⁵ AL is an iterative training approach with an updated dataset in each iteration step by sampling structures uncorrelated to those in current dataset with low similarities; uncertainty quantification (UQ) methods including D-optimality criterion,⁵⁶ Mahalanobis distance,⁵⁷ atomic descriptors,⁵⁸ and cosine similarity⁵⁹ can be used to measure the similarity. The MTP model was trained using structure data for bulk cells with applied strains and six $\Sigma 3$ and $\Sigma 5$ GB models collected from AIMD up to 2000 K. The performance was validated by GB energies in addition to low energy and force MSEs for the GB models and amorphous Li_3PS_4 . The low errors for the amorphous structures may be owing to the data collection at 2000 K where the bulk unit cell and GBs can be disordered significantly and melt, providing structure information for amorphous configurations. It would enhance the transferability enabling applications of the ML model to other GBs with Σ values up to 25 and interfaces between amorphous and crystalline Li_3PS_4 phases. Perovskite oxide SEs have been also studied with MLIPs: Ko et al. and Lee et al. applied the AL method to develop MTP models for perovskite oxide SEs $(\text{Li}_{(2/3)y}\text{Sr}_{1-y})(\text{Ta}_{0.334}\text{Nb}_{0.347}\text{Zr}_{0.211}\text{Sn}_{0.108})\text{O}_{3-\delta}$ ($y = 9/16$) and $\text{Li}_{0.375}\text{Sr}_{0.44375}\text{Ta}_{0.75}\text{Zr}_{0.25}\text{O}_3$, respectively, by using $\Sigma 3$ and $\Sigma 5$ GB structures with cation disordering from Monte Carlo/MD simulations up to 2100 K during AL, and investigated the diffusivities and structural evolution at the GBs.^{60,61} The ML models were validated by energy and force errors for the GB structures from MLMD trajectories and GB energies. Lee et al. also showed that the predicted GB energies for GB models not included in the training dataset are accurate and comparable to DFT results. They applied their model to simulate a $\Sigma 51$ GB, implying good transferability for unseen GB structures during training.

The successful applications of MLIPs in these examples can be attributed to proper validation by benchmarking properties at GBs. Most studies validated ML models by comparing GB energies to DFT results, in addition to energy and force errors. However, more comprehensive tests of structural,

vibrational, and dynamical properties at GBs are still needed: among aforementioned literature, only two studies^{53,54} benchmarked RDF and/or ion transport properties (Figure 1b) against AIMD and experiments. The primary purposes of MLIPs are to investigate structural evolution and Li transport characteristics through dynamics simulations, thus comprehensive assessments of ML models will be required for reliable applications of MLIPs. The protocol for post-training evaluation of ML models can be outlined as follows: 1) RMSEs of predicted energies and forces for structures from MLMD trajectories driven by the trained model—the trajectories will serve as true “test dataset” independent from the training and validation datasets used during model training. Relying solely on low validation errors observed during training should be avoided, and mean absolute error (MAE) can omit outliers that may lead to unphysical behaviors. 2) Benchmarking basic properties against DFT/AIMD, including lattice parameters, structural characteristics (RDFs and angular distribution functions), elastic constants, and dynamical properties (e.g., pDOS and vDOS); 3) benchmarking application-specific properties against DFT/AIMD simulations and experiments, e.g., ion transport parameters, GB/interface energy, phase stability and transformation, melting, and fracture and surface energies. Since benchmarking properties of large GBs is not feasible due to the limitation in DFT scale, it will be effective and efficient to validate the ML models by benchmarking properties of distorted/disordered structures and amorphous models with a few hundreds of atoms, which can serve as proxies for local atomic configurations at GBs and interfaces.^{50,62}

Selecting ML models affects the accuracy and efficiency of MLIPs to perform MD simulations for GBs in ASSBs. Among the papers utilized MLIPs for GBs in ASSBs, most of the studies used MTP models for MLIP development,^{52-55,60} except one that used a SNAP model.⁵¹ The popularity of MTP model may be owing to the virtue of its well-balanced performance between accuracy and computational efficiency.^{10,39} A similar trend is observed in the development of MLIPs for GBs in elemental and binary systems, where GAP, MTP, NNP, and SNAP models have been utilized as discussed. It implies that these relatively simple models compared to GNN models can correctly encode local atomic configurations at GBs not only for simple and binary systems but also for complex materials in ASSBs with up to seven elements⁶⁰. There is no definitive upper limit to the number of elements that these relatively simple models can accommodate, though the computational efficiency will decline rapidly with the increase in the number of elements; in symmetry function-based NNPs, for instance, the number of descriptors increases exponentially with addition of new elements because the symmetry functions must be defined for two-body and three-body interactions between all elements.⁶³ If these models exhibit limitations in describing complex GBs, state-of-the-art GNN models such as Allegro⁶⁴ and MACE⁶⁵ will be helpful to develop MLIPs capable of encoding extremely complex configurations without limitations in the number of elements (see the Summary and Outlook section for discussions regarding GNN and foundational MLIP models).

Efficient and effective dataset generation that enables transferable MLIPs for diverse GBs is desired to explore various GBs in ASSBs. For example, $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZO) SE is characterized by no preference of GB orientations and loss of lattice coherency across GBs,⁶⁶ which require to simulate several GBs with a wide range of degree of disorder to investigate the inherent GB properties in LLZO microstructures. Interestingly, most studies included structure data for GB models into the dataset for training, which were collected either by AIMD⁵⁵ or during AL with MLMD and DFT labeling^{53,54,60}. This approach can be helpful to train a ML model with improved accuracy for simulating the same types of GBs at larger scale. Though it would not improve the transferability to simulate other GBs with different local compositions and configurations, which is necessary to explore diverse GBs. Since the system size becomes easily over 300 atoms even for simple $\Sigma 3$ and $\Sigma 5$ GBs in ternary and higher-order materials in batteries,⁵³ it is not practical to include various GB models with higher Σ values. This issue can be alleviated by using disordered/amorphous structures of bulk materials that mimic disordered GB structures. Jalem et al. might use disordered (and potentially melted) structures for training by sampling structures of Li_3PS_4 bulk and GB models from AIMD at 2000 K, which could improve the transferability to simulate more complex GBs with Σ values up to 25 (low energy and force errors were confirmed for amorphous structures).⁵⁵ Also, Sergeev et al. trained a ML model only with liquid structures that could be helpful to simulate GB models for Li metal systems.⁵² Therefore, including amorphous/disordered structures of bulk phases will improve the transferability of MLIPs for various disordered GB configurations.

Our previous work demonstrates an example of aforementioned discussions regarding data type, model selection, and validation protocol by developing a NNP model for GBs in LLZO SEs.⁶² A NNP model with BP symmetry functions⁸ was trained within the training framework within *n2p2* code⁶⁷. As proxies for diverse configurations at GBs,⁶⁸ amorphous structures with various densities were used in addition to crystalline structures to provide local atomic information at GBs, without GB data. The trained model was first tested by low energy and force errors and then benchmarked against AIMD by predicting accurate RDFs, vDOS, and elastic moduli (Figure 2a). Next, we benchmarked our model against properties critical for probing the structural and dynamical characteristics of LLZO GBs. We found that the predicted Li diffusivities in crystalline and amorphous LLZO are comparable to DFT and AIMD results (Figure 2a), and the predicted GB energy of LLZO $\Sigma 5(210)/[001]$ symmetric tilt GB (1.93 J/m^2 , 960 atoms) agrees with the DFT value (1.82 J/m^2). Also, our model is able to accurately predict tetragonal-cubic phase transformation temperature of LLZO (850 K, Figure 2a), very close to the experimental value of 918 K,⁶⁹ even though tetragonal structure data was not included in the training dataset. It would be due to the inclusion of amorphous data as well as structural similarity between cubic and tetragonal phases. Although it is not easy to directly benchmark the properties of large GB structures containing thousands

of atoms against AIMD, testing ML models on amorphous structures with a few hundred atoms by DFT can indirectly validate the capability of the MLIPs to simulate disordered GB configurations in large

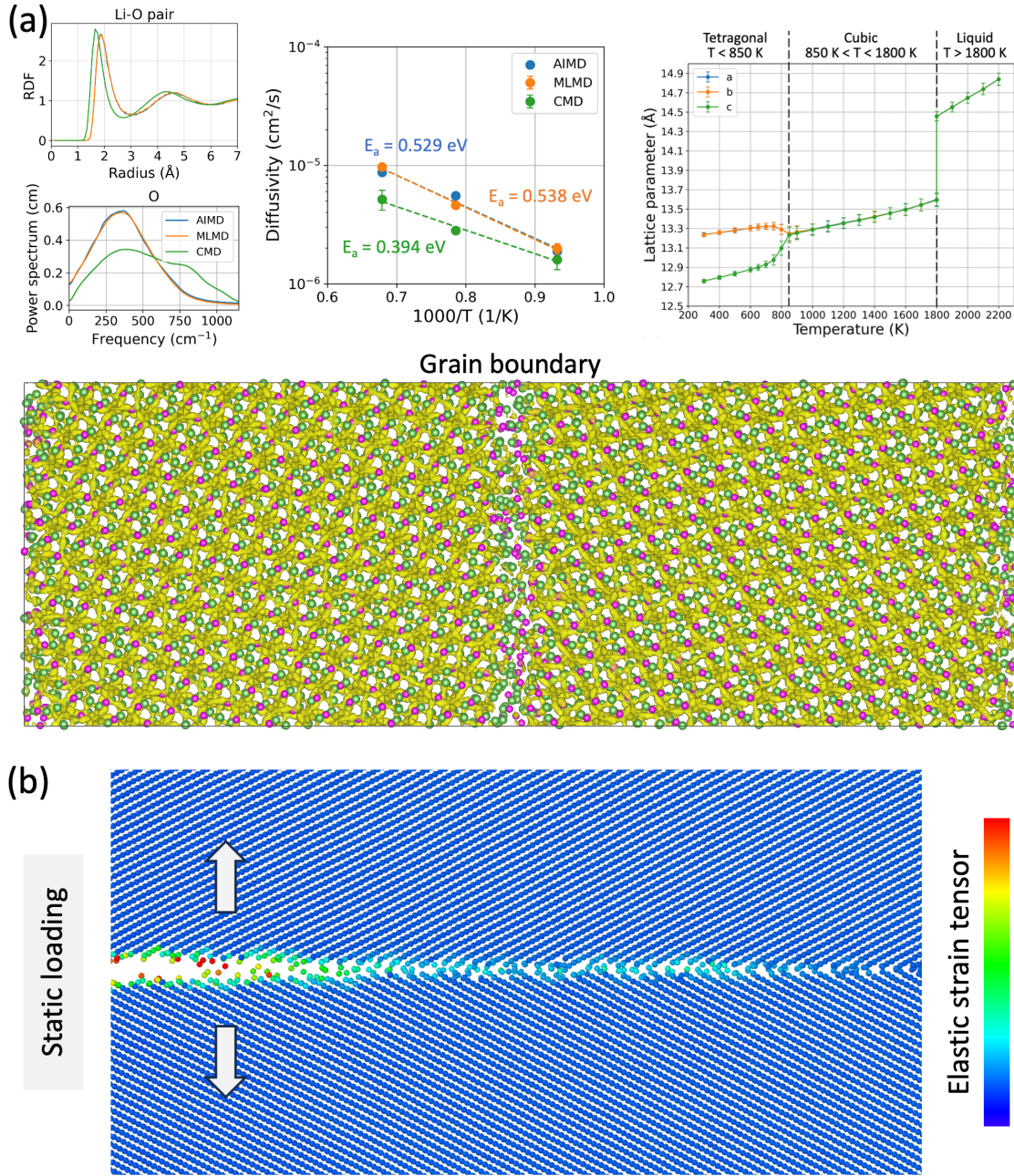


Figure 2. (a) (top) Performance test by benchmarking radial distribution functions (at 3000 K), vibrational density of states (at 3000 K), and Arrhenius plots for Li transport in amorphous LLZO against AIMD as well as tetragonal-cubic phase transformation (850 K vs. experimental measurement of 918 K⁶⁹) and melting (detail validations can be found in our previous publication). Reproduced with permission from Ref.62 (K. Kim et al.) Copyright 2022 AIP Publishing LLC. (bottom) Positional probability density (yellow isosurface) presenting trajectories of Li ions in the $\Sigma 41(3\bar{4}4)/[011]$ symmetry-tilt grain boundary model (31,488 atoms) at 1300 K by npt MD simulations for 2.5 ns, showing degraded diffusion of Li at the GB area. Magenta and green spheres represent Zr and La ions, respectively. (b) MD simulation for mechanical response at the $\Sigma 41(3\bar{4}4)/[011]$ symmetry-tilt grain boundary model (471,382 atoms) under a static tensile loading normal to GB plane, observing fracture propagation along GB plane (only the Zr sublattice is shown).

scale. Our model was able to perform nvt and npt MD at 1300 K for 20 symmetry tilt GB models with three tilt axes, CSL sigma values up to 41, and GB plane index up to 5 in a wide range of misorientation angles ($16.3 - 70.5^\circ$), which were used to extract Li transport properties at GB regions that informs transport parameters in mesoscale modeling (Figure 2a bottom panel).⁷⁰ It implies that training ML models using amorphous structures and testing it with the detailed validation protocol can develop transferable MLIPs to explore dynamic evolutions and predict accurate properties of various GBs.

In ASSBs, mechanical failure is one of key factors that critically dictate the battery performance. For example, fracture propagation at GBs lead to the loss of contact between grains and potential stress-induced transformations to unwanted phases, which results in ionic conductivity degradation.⁷¹ MLIPs can aid in accurate and efficient atomistic simulations to investigate the fracture mechanisms and mechanical response at GBs. For this, MLIPs need to be trained with surface data that will evolve during crack opening and fracture propagations with atoms exposed to vacuum. Based on our initial work,⁶² we trained the next version of MLIP with additional data for two LLZO slab models, (001) plane with Li termination and (111) plane with Zr termination obtained at temperatures ranging from 300 K to 3000 K, which cover clean, disordered, and completely amorphized surfaces as well as significant expansion with complete melting.⁷² Our MLIP shows good transferability to other surfaces that are not included in the dataset with various degree of surface disorder, e.g., (110) Li, (110) La, (110) O, and (111) Li terminations: energy and force errors for all six surface models are at most 8.4 meV/atom and 0.19 eV/Å at 2000 K. Also, inclusion of the surface data further improves the prediction of the tetragonal-cubic phase transformation temperature from 850 K⁶² to 900 K.⁷² The capability of MLIPs to predict transformations between polymorphs is important to predict chemo-mechanical degradations in ASSBs because mechanical responses and damages can induce phase transformations with undesired physico-chemical properties. Using this MLIP, we simulated LLZO's mechanical behavior, including fracture propagation at GBs (Figure 2b), to assess fracture toughness and dynamical crack growth in the bulk and GB regions.⁷² Our examples suggest that including disordered bulk and surface structures in the training data is helpful to improve the transferability of MLIPs across a wide range of atomic configurations and enable predictive analyses for chemo-mechanical response at GBs.

In summary, we analyzed existing literature that utilized MLIPs to study GBs for ASSB materials as well as elemental and binary compounds and discussed MLIP development strategies in three important aspects: data generation, model selection, and validation. It was shown that MTP and NNP models are the most popular ML frameworks with good accuracy. For reliable performance, however, thorough validations will be needed after training by benchmarking properties related to targeted MD applications against AIMD, e.g., structural, vibrational, and dynamical characteristics, since low energy and force

errors do not necessarily guarantee accurate predictions of collective and dynamical behaviors of atoms. Also, incorporating liquid-like amorphous structure into the training set is helpful to achieve good transferability across various GBs with disordering and spatiotemporal evolution, which will reduce the computational cost to directly simulate a number of GB models with AIMD simulations.

3. Modeling Interfaces with ML Potentials

Interfaces critically dictate the performance of ASSBs due to electro-chemo-mechanical instabilities that lead to non-uniform stripping and plating of operating ions, (electro)chemical reactions with formations of unwanted interphases and nucleation, and mechanical degradation with crack and fracture, resulting in dendrite growth, capacity fade, increase in resistance, and cell failure.^{1,4} Simulation capabilities to directly prove interfacial phenomena at atomistic scale are desirable to understand the fundamental mechanisms of interfacial degradations in ASSBs, which will inform design principles for interface engineering with proper inherent interfacial properties. MLIPs offer such capabilities of simulating realistic large-size atomic models with quantum-level accuracy for interfaces. To develop MLIPs, it is essential to generate datasets containing structures that capture diverse atomic environments at interfaces because chemo-mechanical instabilities can cause local structural and compositional variations, including disorders in materials and mixing between them. Also, ML model needs to be sophisticated enough to encode the complex environments. The deployment of MLIPs will proceed following proper validation of their performance. Here, we will present a few examples of MLIPs for interfaces between relatively simple metals and insulators, and then review how MLIPs have been developed to interfaces between more complex ASSB materials. We will analyze these examples in terms of data generation, model selection, and validation and discuss proper ways to develop the MLIPs for the interfaces.

MLIPs for interfaces in general materials and implications for development. MLIPs are increasingly used to model interfaces between elemental or binary metals and insulators, enabling large-scale MD simulations and improved accuracy in predicted interfacial properties, e.g., structural stability, interdiffusion process, and degradation.^{73,74} Jeong et al. developed a NNP model with the BP symmetry functions for Ni-Si interfaces to investigate the Ni-silicidation process in the fabrication of semiconductor devices for process optimization.⁷³ They used crystalline structures of Ni, Ni₂Si, NiSi, NiSi₂, and Si, their liquid/amorphous forms with complete disorder, (001) surfaces of Ni and Si, and relatively small interfaces up to 232 atoms, which were collected from AIMD up to 2000 K as well as static DFT. Thus, the training dataset covers diverse atomic environments with compositional variations (Ni_xSi_y). The authors suggested the importance of including metastable/unstable phases in the dataset: their earlier

version of MLIP leads to the formation of a less-stable NiSi phase not included in the dataset, which was discussed as an artifact, implying that out-of-equilibrium and local minima configurations on the potential energy surface (PES) are essential. Also, the authors utilized a newly developed UQ method during training, which uses replica NNPs that are trained using the predicted atomic energies by the reference NNP (i.e., output from the MLIP) and calculate the standard deviation of atomic energies, indicating higher uncertainty (deviation from a true PES) with higher standard deviation. This metric can inform which additional data will be needed for model improvement with AL. Jeong et al. also benchmarked the energy profile during interfacial disordering as well as the equilibrium volume and radial distribution functions for amorphous Ni₂Si, NiSi, and NiSi₂. These comprehensive development processes enable accurate large-scale MD simulations of interfacial evolution process with significant changes in atomic configurations. Achar et al. developed a MTP model to investigate the degradation mechanisms of Ti-Ge₂₀Se₈₀ interfaces for nonvolatile memory cells.⁷⁴ Similar to Jeong et al.⁷³, They generated structures with various compositions (TiSe₂, TiGe₂, GeSe, and GeSe₂) at high temperatures to account for interfacial disordering/amorphization and utilized D-optimality criterion⁵⁶ to measure the model uncertainty by the extrapolation grade for AL. The model was validated by the equation of state and mechanical properties of crystalline structures as well as energy and force errors for test dataset, but interfacial properties have not been validated. These works demonstrate the importance of diverse structure data and appropriate evaluation of ML model during training for the development of suitable MLIPs for interfaces. They also imply that NNP and MTP models are sophisticated enough to encode structural information at interfaces between relatively simple elemental/binary materials.

MLIPs for interfaces in ASSB materials. A review paper in 2020 noted the absence of MLIPs specifically designed to directly simulate interfaces in battery-related applications.²⁰ However, their use in

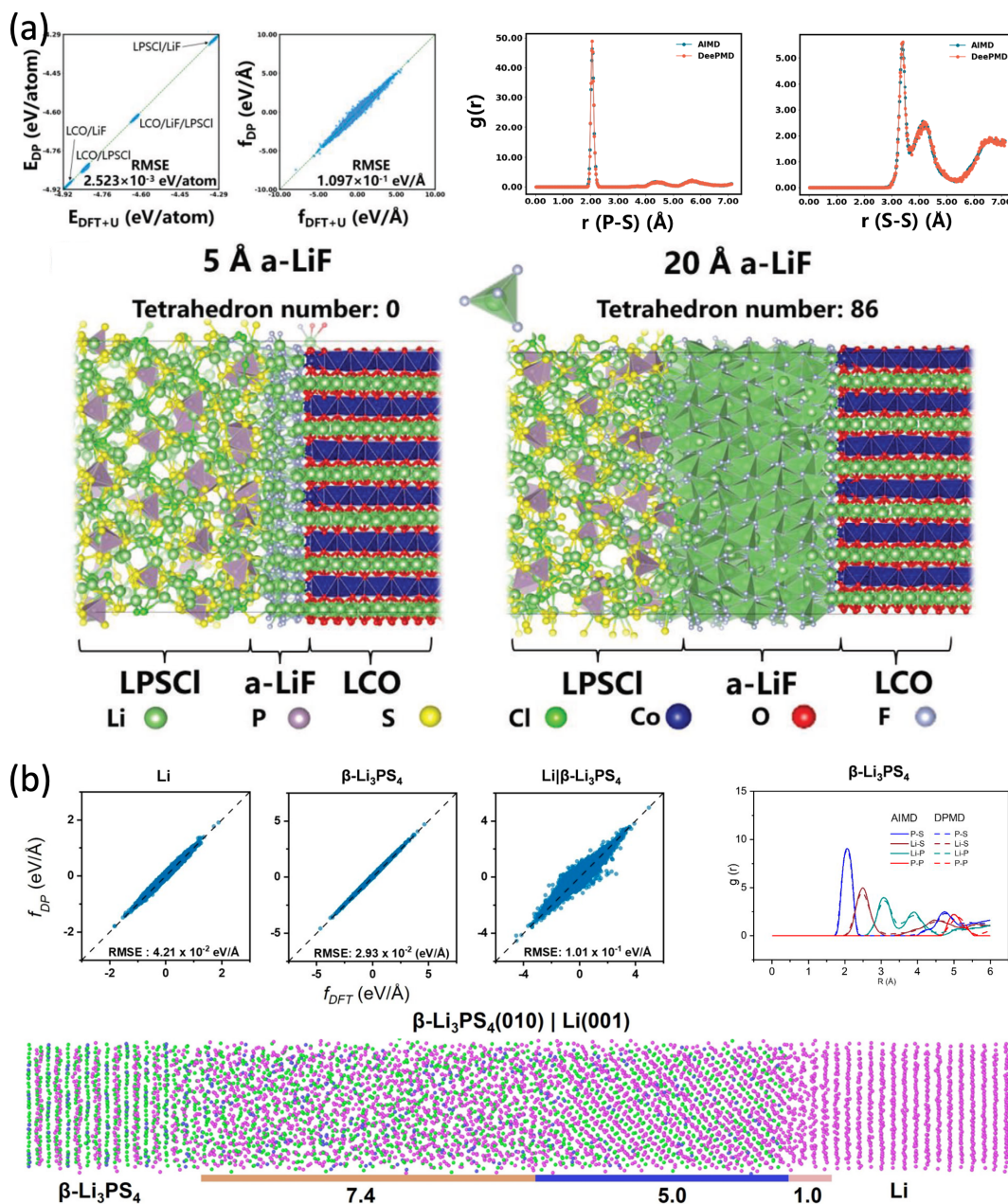
Table 2. List of literature utilizing MLIPs for investigating interfaces in ASSBs.

Year	MLIP model	System and purpose	Ref.
2022	MTP	Interface stability and Li migration channel at S ₈ /β-Li ₃ PS ₄	H. Chandrappa ⁷⁵
2022	MTP	Li transport at Li/Li ₃ P, Li/Li ₂ S, Li/LiCl	J. Wang ⁷⁷
2022	NNP	Li plating/stripping mechanisms on Cu current collector	G. Lai ^{76,79}
2023	MTP	Li transport, morphology, and thermodynamics at Li ₃ PS ₄ /Li ₃ B ₁₁ O ₁₈	C. Wang ⁷⁸
2023	NNP	Interfacial evolution and voltage profile of Li-Si anode	F. Fu ^{80,XX}
2024	NNP	Effect of LiF on interfacial stability and Li diffusion between LiCoO ₂ / Li ₆ PS ₅ Cl	T. Hu ⁸¹
2024	NNP	Interfacial evolution and interdiffusion at LiCoO ₂ /Li ₇ La ₃ Zr ₂ O ₁₂	K. Kim ⁵⁰
2024	MTP	SEI formation mechanisms at Li/Li ₆ PS ₅ Cl	G. Chaney ⁸⁴
2024	NNP	Kinetic evolution and interfacial morphology at Li/β-Li ₃ PS ₄	F. Ren ⁸²
2024	NNP	Discharge process in a full-cell configuration of Li/Li ₇ La ₃ Zr ₂ O ₁₂ /Li _x ZrO ₂	Y. You ^{85,87} D. Zhang ⁸⁸
2025	Allegro	SEI formation and effect of Li ₂ S coating at Li/Li ₆ PS ₅ Cl	X. Ding ⁹⁵

modeling interfaces in ASSBs has grown significantly in recent years (see Table 2 for a summary of literature).^{50,75-85} Chandrappa et al. trained a MTP model for cathode interfaces between S₈ cathode and β -Li₃PS₄ and exploited it to reveal that the most stable Li₃PS₄(100) surface forms interfaces featured with 2D channels and low activation energies for Li migration, which is related to the interfacial areal number density of Li.⁷⁵ They trained the MTP model using structure data for bulk β -Li₃PS₄, Li₂S, and α -S₈ from AIMD up to 2000 K and then performed AL with D-optimality⁵⁶ using seven Li₃PS₄/S₈ interfaces (less than 250 atoms) collected from MTP MD simulations with DFT referencing. The model was validated by energy and force errors and interface energies for interface models as well as properties of each material. C. Wang et al. developed a MTP model with AL for Li₃PS₄/Li₃B₁₁O₁₈ interfaces (Li₃B₁₁O₁₈ as a coating material) to study interface morphology, thermodynamic properties, and Li-ion diffusion, suggesting that the interfaces are relatively stable without blocking Li interdiffusion.⁷⁸ They sampled training data using amorphous Li₃PS₄, amorphous Li₃B₁₁O₁₈, and Li₃PS₄/Li₃B₁₁O₁₈ interfaces from AIMD up to 1000 K. The model validation was performed by energy and force errors for each material and interfaces against DFT. Hu et al. developed a DeePMD model to study the effects of amorphous LiF (a-LiF) coating at LiCoO₂/Li₆PS₅Cl interfaces, suggesting an optimal coating thickness that can suppress the interfacial side reactions while minimizing the increase in the resistance for Li transport (Figure 3a).⁸¹ They generated LiCoO₂/Li₆PS₅Cl, LiCoO₂/LiF, and LiF/Li₆PS₅Cl interfaces structures as well as a-LiF bulk structures by AIMD for initial training followed by AL for these structures and LiCoO₂/LiF/Li₆PS₅Cl interfaces. The model is validated by the energy and force errors for various interfaces and RDFs at the LiCoO₂/Li₆PS₅Cl interface. This model is impressive as it covers seven elements at various interfaces between three materials, implying that descriptor-based NNP can encode complex local configurations at disordered interfaces with an increased number of elements.

MLIPs have also been applied to interfaces between Li metal anode and SEs as well as Li-Si anode. J. Wang et al. developed MTP models for anode interfaces between Li metal and decomposed products of Li₆PS₅Cl (Li₃P, Li₂S, and LiCl) and suggested resistive nature of the solid-electrolyte interphase (SEI) layers with low Li diffusivities across the interfaces.⁷⁷ Interface models of Li(110)-Li₂S(110), Li(100)-Li₃P(001), and Li(110)-LiCl(100) (520, 406, and 439 atoms, respectively) were used to sample training data from AIMD at 1000 K in addition to each bulk material. The trained ML model was validated by benchmarking diffusivities at the interfaces against AIMD and energy and force errors for the interfaces and bulk. Later, Chaney et al. trained an MTP model to directly explore Li-Li₆PS₅Cl interfaces and suggested a two-step growth process of SEI layer, consisting of an initial electrolyte reduction followed by gradual crystallization of products.⁸⁴ The initial training dataset includes various interface models from AIMD at 298 K and is expanded during AL (the structure types for AL are not specified). The model was tested only with the energy errors for training and test datasets used during training process. Lai et al.

showed the dependence of Cu surface on Li plating/stripping mechanisms on Cu current collector for anode-free Li metal batteries by using a DeePMD model for Li-Cu interfaces, suggesting that Cu(110)



facet promotes inhomogeneous Li deposition and dendritic growth, alongside the formation of bulk Li, Cu, Li_3Cu , and LiCu_3 phases by AIMD (temperature is not mentioned).⁷⁹ They validated the model by the energy and force errors for the structures (test dataset), equation of states, and elastic properties of Li-Cu alloys, RDFs for amorphous Li_3Cu at 1500 K. They also benchmarked the adsorption energy of Li on Cu against DFT, which aligns with the purpose of the MLIP. Ren et al. developed a DeePMD model to study the kinetic evolution, interfacial morphology, and potential energy profile at the Li-metal/ β - Li_3PS_4 interfaces, suggesting the formation of crystalline Li_2S and amorphous Li_xP SEI components (Figure 3b).⁸² Initial structure data for bulk Li, bulk β - Li_3PS_4 , and Li/ β - Li_3PS_4 interfaces were sampled from AIMD at 400 K and used for the training of a preliminary model, and AL was performed by exploring the bulk and interface structures up to 1000 K and 100 bar. The validation was performed by the energy and force errors for the bulk and interface structures and RDFs only for bulk material.

Now we further analyze and discuss the aforementioned literature in terms of ML model, validation, and training data. First, it is interesting that most of existing applications of MLIPs to interfaces (and also GBs) in ASSBs have utilized MTP and descriptor-based NNP (mostly DeePMD) models. The MTP and NNP models were shown to have capabilities to simulate disordered complex interfaces with five and seven elements, respectively, along with AL with UQ measurement to cover a vast range of configurational and compositional space. It implies that relatively simpler ML models like MTP and NNP (compared to GNN-based models) can accurately perform dynamical simulations not only for relatively simple interfaces for elemental or binary materials but also for complex interfaces between multinary compounds with significant disorder, which is the same conclusion drawn from surveys of the literature applying MLIPs to GBs in ASSB materials. At the same time, we anticipate a growing number of publications utilizing GNN-based MLIPs for interfacial systems, owing to their superior ability to describe extremely complex atomic environments without a difficulty of increasing the number of elements. The choice of ML model will depend on system complexity and computational cost, as well as the availability of GPU resources required for GNN-based approaches (see the Summary and Outlook section for a discussion of GNN and foundational MLIP models).

Albeit reliable abilities of these ML frameworks, a thorough validation for ML models will be necessary with a validation protocol. Most of studies validated their models by energy and force errors for interface structures,^{75-78,81,82,84} though only some of these studies performed benchmarking tests for structural characteristics^{76,81} and static properties^{75,76} for interfaces by RDFs and interface/adsorption energies, respectively. Although most of these studies aim to predict (inter)diffusion and nucleation/interphase formation at interfaces, none have validated their models using dynamic properties—except for one, which benchmarked interfacial diffusivities against AIMD results.⁷⁷ There is no clear criteria for energy

and force errors that ensure reliable vibrational and dynamical behaviors comparable to AIMD, which can even be system dependent. Therefore, it would be advisable to benchmark MLIP models with properties relevant to applications as well as vibrational and structural characteristics for interfacial configurations, in addition to basic properties.

Transferability of MLIPs for interfaces is of importance to simulate local atomic environments with spatiotemporal evolution with a wide range of compositions and configurations. It will be helpful to include interface structures in training dataset; however, this approach is rather impractical for incorporating a wide range of interfaces with various surfaces because of the scale limitation and computational cost of AIMD. It requires practical methods to generate structures that can cover diverse atomic environment at interfaces. Utilizing amorphous mixtures between two materials with various compositions and configurations can be one of practical ways, which will serve as proxies of local structures and provide arbitrary but diverse data at the interfaces. With bulk structures for each material, the entire dataset provides information at parts of interfaces, like a “divide and conquer” strategy, and enables for ML models to describe the whole picture of interface systems. For example, to study Li-Si anode to understand detail (de)lithiation process, Fu et al. developed a DeePMD model by utilizing amorphous Li_xSi structures without interface structures, in addition to crystalline Li, Si, and Li_xSi phases with random perturbations to ionic positions and lattice vectors.^{80,83} They simulated structural changes, voltage profile, and interfacial evolution and suggested that Si flux from bulk Si to Li-Si interface dominates the interdiffusion process owing to thermodynamic preference. The model was also validated by energy and force errors and RDFs for crystalline and amorphous Li_xSi phases as well as the lattice parameters, formation energy, and voltage profile of crystalline Li_xSi . The model can perform simulations for crystalline-Si/Li and amorphous-Si/Li interfaces with low energy and force errors (7.4 meV/atom and 0.16 eV/Å) even without interface data. Also, You et al. trained a DeePMD model without interface data but using a wide range of compositions in amorphous $\text{Li}_x\text{La}_y\text{Zr}_z\text{O}_{(x+3y+4z)/2}$ in addition to tetragonal and cubic phases, surface, and GB models of LLZO as well as multiplex compounds of Li, La, Zr, and O from the Materials Project⁸⁶ database.^{87,88} The MLIP was utilized to simulate the discharge process in a full-cell configuration with interfaces between Li metal anode, LLZO SE, and Li_xZrO_2 model cathode (without electric field), suggesting that GB amorphization can mitigate Li dendrite growth in LLZO SE.⁸⁵ The model was not validated for Li/LLZO and LLZO/ Li_xZrO_2 interfaces; we presume that this MLIP could simulate the interfaces owing to diverse amorphous compositions that represents disordered local configurations at interfaces (there is no additional elements included in electrode materials) as well as information for diatomic pair potentials providing repulsion at very close distances and preventing simulation failure with unphysical atomic overlap, which is a common failure mode of MLIPs. These

studies indicate that amorphous mixtures between two materials with various stoichiometries can provide useful information for training ML models for interfaces.

In addition to these examples, our previous study suggests that a properly validated NNP model trained using dataset including amorphous mixtures between two materials exhibit good transferability to various interfaces with different planes and chemistries with increase complexity.⁵⁰ To simulate interfacial degradation between LLZO SE and LiCoO₂ (LCO) cathode and study interdiffusion across the interfaces, we developed a NNP model with BP symmetry functions (n2p2 code)^{8,67} using a dataset with 12,651 structures, including five LLZO-LCO mixture models with various stoichiometries between five elements as well as crystalline and amorphous Co-doped LLZO and LCO bulk structures from AIMD simulations up to 4000 K. The model was validated by estimating the energy and force errors (at most 28.6 meV/atom and 0.29 eV/Å for structures from MLMD simulations) and benchmarking RDFs, vDOS, and Li diffusion against AIMD at 2000 K for five LLZO-LCO mixture models not used for training as well as all structure models in the dataset (Figure 4a). Our model was also benchmarked against AIMD using various interfaces LLZO(001)/LCO(104), LLZO(001)/LCO(100), and LLZO(001)/LCO(110) with 744, 432, and 480 atoms, respectively. The results show low energy and force errors (17.9 meV/atom and 0.27 eV/Å), even as significant changes in interfacial configuration and amorphization occurred during 1 ns MLMD simulations initiated from clean, ordered interfaces. This thorough validation enables this MLIP to simulate the three interfaces, as well as LLZO(001)/LCO(003), on a large scale with approximately 20,000 atoms (Figure 4b). The simulation results revealed the propensities of interfacial degradation depending on chemical conditions with and without Li deficiency, their effects on and the mechanisms of interdiffusion of Co into LLZO, and segregation of Co ions in LLZO GBs implying the formation of Co-rich secondary phase, which supports detail understanding of experimental observation at atomic scale (Figure 4c).^{5,50} We believe that the inclusion of diverse disordered/amorphous mixtures between two materials is helpful to provide sufficient information for local atomic environments at interfaces and can improve the transferability to simulate various interface models with significant chemical and configurational variabilities.

In summary, we reviewed existing examples of MLIPs for interfaces between ASSB materials as well as elemental and binary compounds and discussed development strategies. Similar to MLIPs for GBs, MTP and NNP models have been widely used for interfaces, which are able to simulate complex atomic environments at interfaces up to five and seven elements, respectively. Efficient generations of training dataset for interface MLIPs can be achieved by utilizing amorphous structure models by mixing materials at interfaces with diverse compositions that represent spatiotemporal variation of interfacial configurations. Validations of existing models have been performed by benchmarking relevant properties

against DFT; however, few studies have validated their models using dynamic properties at interfaces or in amorphous structures. Thus, thorough validations by benchmarking both static and dynamic properties

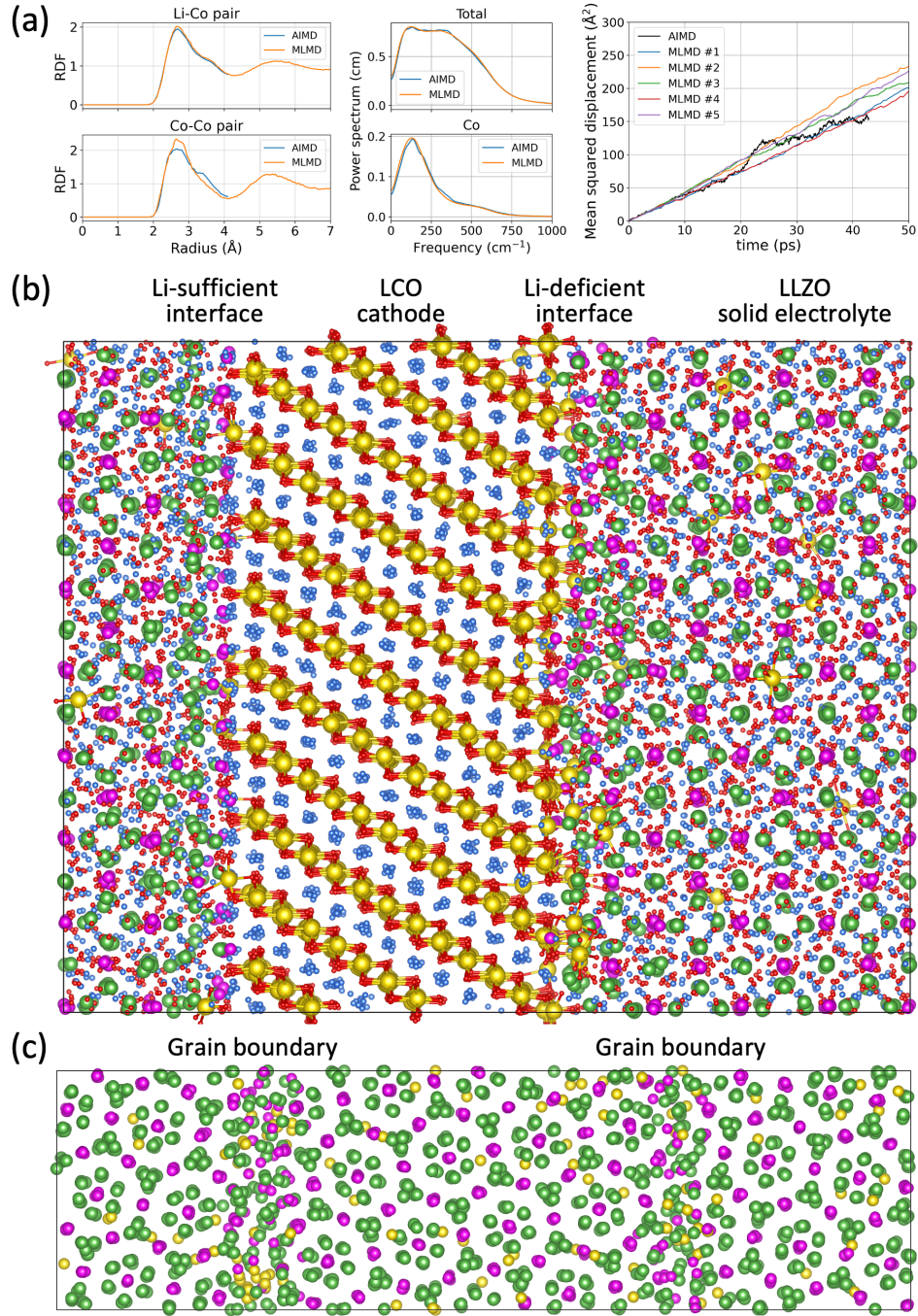


Figure 4. (a) Performance test by benchmarking radial distribution functions, vibrational density of states, and Li diffusion for an unseen LLZO|LCO mixture model at 2000 K (detail validations can be found in our previous publication)⁵⁰. (b) LLZO(001)-LCO(104) interface model after a 10 ns MLMD simulation at 1500 K. Li-sufficient and Li-deficient conditions are represented at the left and right interfaces, respectively, which result in different interfacial evolution propensities. (c) Atomic structure of $\Sigma 7(213)/[111]$ GB with Co-doped LLZO after a 10 ns MLMD simulation at 1300 K, showing Co accumulation at GB regions. Yellow spheres represent Co ions, La is green, Zr is magenta, Li is blue, and O is red. Reproduced with permission from Ref.50 (K. Kim et al.) Copyright 2024 Elsevier B.V.

would be necessary to perform reliable simulations for investigation of dynamic evolution of interfaces.

4. Summary and Outlook

The performance of ASSBs is critically influenced by the properties at GBs and interfaces. Accordingly, a deep understanding of these interfaces is essential for interface design and the optimization of cell performance. Recent advances in MLIPs have enabled comprehensive investigations of interfaces through accurate large-scale atomic simulations. Nevertheless, the development of MLIPs needs to be performed with care by rigorously validating their ability to describe interfacial phenomena, including chemical reactions and the formation of secondary phases. ML models require sufficiently diverse training data covering reactions and stable/metastable phases in a high-dimensional phase space in addition to disordered/amorphous structures with various compositions and configurations (Figure 5a). Furthermore, current MLIP implementations generally do not explicitly account for charge transfer, long-range interactions, and external fields, all of which play critical roles at electrochemical interfaces. As ML models and techniques continue to evolve rapidly, there are growing opportunities to develop MLIPs with efficient development strategies and improved reliability and simulate interfaces more realistically. These opportunities are discussed below, aligned along the three aspects of data generation, model selection, and validation, and summarized with a workflow diagram for MLIP development (Figure 5b).

- Data generation:

The quality of data with its ability to cover a wide range of diverse atomic environments is a critical factor in determining the performance of MLIPs for interfaces. Because the complete range of structure data required to accurately simulate GBs and interfaces is not fully known, it is important to conduct thorough validation and employ AL strategies for the ML model. AL schemes can supplement datasets with additional diverse structures and expand the range of amorphous/disordered structures in terms of compositional and configurational variety, which will ensure good transferability to various GBs and interfaces. Nevertheless, it remains essential to generate an initial dataset that enables preliminary models with relatively reliable performance for iterative data generation and training. As discussed, incorporating amorphous/disordered structures with varied compositions and configurations can support efficient data generation; however, producing such a wide range of structures still requires computationally expensive AIMD simulations. Recently, universal or foundational MLIP models have been developed and are publicly available, e.g., M3GNet,⁸⁹ CHGNet,⁹⁰ and MACE⁹¹ (see a full list of the foundational models (FMs) from the Matbench Discovery; <https://matbench-discovery.materialsproject.org>)⁹².

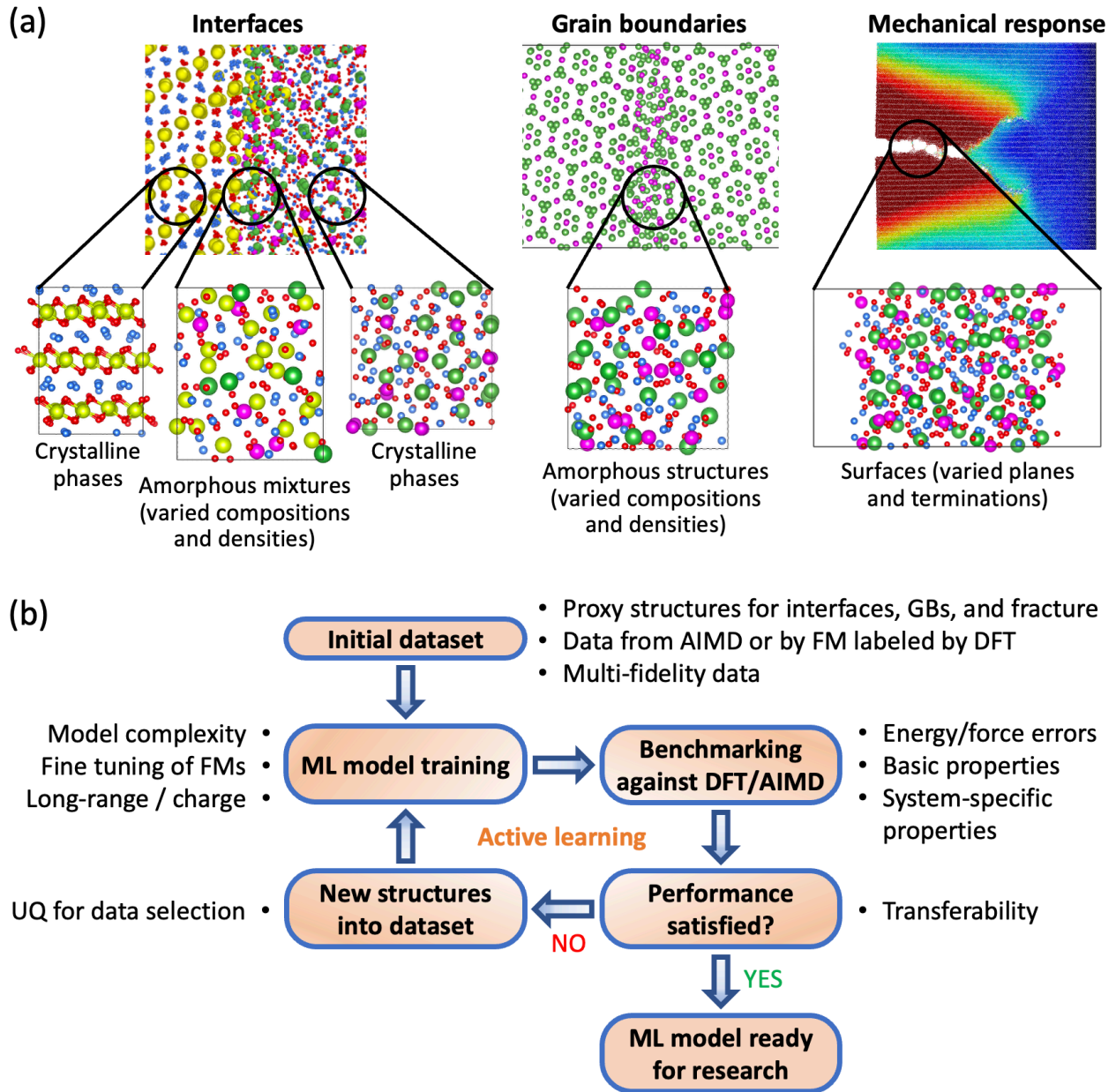


Figure 5. (a) Types of structure data for training MLIPs for interfaces, GBs, and mechanical response simulations. (b) Workflow diagram of MLIP development highlighting important aspects and potential opportunities with new evolving techniques to investigate GBs and interfaces in ASSBs.

They are pre-trained MLIPs with extensive DFT databases and can perform atomistic modeling for most elements up to atomic number 94 (plutonium). It enables the generation of reasonable amorphous/disordered and GB/interface structures without the need for costly AIMD simulations. The sampled structures can then be labeled by DFT calculations to establish high-quality training datasets. This approach would also enable efficient incorporation of higher DFT levels of theory for data generation. Typically, the generalized gradient approximation (GGA) functional is used

in DFT calculations for data generation. However, higher levels of theory, such as meta-GGA and hybrid functionals, or Hubbard-U corrections may be necessary to accurately describe electronic structures and predict properties, especially for materials and interfaces containing transition metals (TMs) with localized electrons. Long-range dispersion interactions can be significant in battery materials with ionic interacting nature and layered structures, which requires the inclusion of van der Waals (vdW) corrections or vdW non-local functionals. In addition, oxidation states of TMs varying in cathodes and at interfaces with different local environments may require spin-polarized DFT calculations to properly account for unpaired electrons. While implementing AIMD simulations with such higher levels of theory is usually not practical, single-point calculations for sampled configurations generated by foundational MLIPs can be feasible for data generation. It is remarkable that multi-fidelity and transfer learning MLIP models enable even more efficient data generation, as they require only a small portion of high-level theory data compared to lower-fidelity data.⁹³ Fine-tuning a pre-trained model only with high-fidelity data is also possible, allowing for improved accuracy while minimizing the need for extensive DFT calculations with higher level of theory.⁹⁴ These approaches will enable efficient generation of high-quality dataset and accelerate the MLIP development process with improved accuracy.

- Model selection:

Increased chemical and structural complexities at GBs and interfaces require higher model complexity of MLIPs. Although simpler models like NNP and MTP have been shown to have ability to describe complex interfaces, state-of-the-art GNN models also can be beneficial for extreme complexities with an increased number of elements and reactions forming secondary phases and nucleation, which is just beginning to emerge for simulations of ASSB interfaces. Ding et al. developed an Allegro model to investigate the formation of SEIs between Li metal anode and $\text{Li}_6\text{S}_5\text{Cl}$ and revealed a new interphase $\text{Li}_2\text{S}_{0.72}\text{P}_{0.14}\text{Cl}_{0.14}$, as well as the effects of Li_2S coating on interfacial stability and significant Li motion along the interface.⁹⁵ This GNN model is able to simulate a massive Li- $\text{Li}_6\text{S}_5\text{Cl}$ interface model up to 3.4 million atoms, showing a good scalability with system size. Therefore, GNN-based models are expected to be applied increasingly to the investigation of interfaces and GBs for ASSB materials.

As mentioned above, foundational MLIP models have been developed based on massive DFT databases. These foundational models demonstrate superior performance owing to the use of GNN-based architectures, which can handle extremely complex systems without limitations on the number of elements, while achieving high accuracy and a degree of transferability across

many chemical species.⁹¹ So, they enable researchers to perform studies for screening and validating suitability of a wide range of materials for ASSBs without training MLIPs. For example, H. Du et al. benchmarked various foundational MLIPs and found good predictabilities for elastic, thermodynamic, and transport properties of SE materials.⁹⁶ Nevertheless, the utilization of the FMs require comprehensive benchmarking tailored to their intended applications, owing to potential failure modes; they may show systematic softening⁹⁷ because of the nature of DFT database for training (e.g., Materials Project⁸⁶ and OMat24⁹⁸) with structures that are mostly populated near equilibrium configurations (although they tried to address this issue by including relaxation trajectories, perturbations in atomic structures, and 50 steps of AIMD trajectories). Also, it is also likely that this aspect increases uncertainties in performance for systems at high temperatures and with chemical and structural disorders, amorphous and metastable phases, and mechanical deformation, defects and fractures, which are inevitable to simulate GBs and interfaces in ASSBs. It will require fine-tuning of FMs with additional relevant training data to improve the accuracy and transferability to these systems. In addition, the superior ability of the FMs comes at a cost to the increase in the computational cost with parameter-rich model architectures of GNN, requiring a significant GPU and memory resources, thus benchmarking tests for scalability will be essential to simulate large interface systems with the FMs. The scalability can be improved by emerging techniques including model scaling by removing low-contribution message-passing layers as a pruning strategy for reduced complexity,⁹⁹ as well as distillation of FMs to simpler GNN and (non)linear regression models like ACE and NNP by training them with synthetic training data generated by fine-tuned FMs.¹⁰⁰

Interface modeling accounting for more realistic conditions including long-range interactions and charges under external fields will be also important in the future MLIP applications to ASSBs.

Due to the difference in chemical potentials between electrodes and SEs, interdiffusion and rearrangement of charged ions leads to the formation of a space charge layer (SCL), resulting in a potential drop and increased resistance across the interface.¹⁰¹ The interfacial configuration can also evolve through disordering and the formation of interphases driven by electro-chemo-mechanical instability, which in turn modifies the SCL; conversely, changes in the SCL can influence interfacial structure, indicating coupled effects between SCL formation and interfacial instability on interfacial resistance.¹⁰² This is one prime example of the importance of long-range electrostatic interactions in ASSBs. However, most studies to date have applied MLIPs to ASSB interfaces without accounting for long-range electrostatic (and dispersion) interactions, as well as their associated potential drop, which are critical for accurately capturing interfacial evolution

particularly at cathode interfaces where the largest potential drop occurs.¹⁰¹ Indeed, several studies have shown that long-range electrostatic interactions are especially important for chemically significant defects, structures containing sheets or chains of atoms, and composite interfaces.^{103,104} For example, Staacke et al. investigated the stability of Frenkel defects in $\text{Li}_7\text{P}_3\text{S}_{11}$ SE under electric fields using a GAP model, demonstrating that accurate stability predictions require the inclusion of long-range interactions.¹⁰⁵ Long-range interactions have been incorporated into several MLIP approaches, including NNP and equivariant graph convolutional neural networks models.^{12,106} Although not yet applied to ASSBs, a study simulated Li dendrite formation at interfaces in a Li symmetric cell with ethylene carbonate/ LiPF_6 liquid electrolyte under constant potential using a NNP model for the short range interactions combined with a charge equilibrium model for the electrostatic interactions.¹⁰⁷ In addition, spin-dependent MLIP models predicting spin states of magnetic materials are available;^{90,108} these approaches could be utilized for cathode interfaces with accurate description of spin states that may also infer the charge states of TMs. For example, the CHGNet model simulated the change in spin/charge states of Mn with ionic migration in $\text{Li}_{0.5}\text{MnO}_2$ during the orthorhombic-to-spinel phase transformation by predicting the magnetic moments accurately.⁹⁰ Incorporating long-range interactions into simulations with accurate prediction of spin/charge states at interfaces in ASSBs would enable more realistic modeling of interfacial evolution under practical electrochemical conditions and is a high priority for future development.

- Validation:

Appropriate testing of trained models is the final step in developing MLIPs for reliable application. Beyond thorough validation of structural, vibrational, and dynamic properties, it is essential to evaluate ML models using unseen data with varied compositions and configurations not used for training. In this way, one can validate transferability for chemistry and structures at interfaces with significant disorder. Transferability to larger scales is another important consideration. MLIPs are typically trained on small atomic structures from DFT and applied to much larger systems such as GBs and interfaces. For example, in NNP with BP symmetry functions, the force vector of a central atom is influenced by surrounding atoms within at most twice the cutoff radius, raising the possibility of self-interaction when the size of structures in the dataset is less than four times the cutoff (typically 20–24 Å, given a 5–6 Å cutoff).¹² Since atomic structures for training and validation are usually smaller than 20 Å due to the limitation in DFT scale, atomic environments in these datasets may differ from those in large-scale interface models during MLMD simulations, especially in disordered or amorphous systems without periodic

lattices. This could be resolved by including diverse atomic configurations with improved transferability for unseen atomic structures, but direct validation by benchmarking against DFT is not feasible for large atomic models. Instead, UQ methods like D-optimality criterion⁵⁶ and Mahalanobis distance⁵⁷ could be incorporated to ensure that atomic configurations that are observed in large-scale MLMD simulations were included in relatively small structures in the training dataset. The development of new metrics to evaluate scale-up transferability during training, not during model validation or application stage, would be an important area for future research. It is also important to consider which properties should be compared against for validation purposes, beyond forces and energies. Many processes of interest at interfaces in ASSBs are related to transport, reactivity, or deformation; these require proper sampling of transition states and dynamically averaged quantities that are further from equilibrium. It is therefore crucial to consider the properties of interest when selecting a bespoke set of benchmarks, ideally incorporating diffusional, vibrational/mechanical, and structural thermal stability criteria alongside more traditional metrics.

In summary, this prospective paper presents a thorough analysis of existing applications of MLIPs for GBs and interfaces in ASSBs, a field that has only begun to emerge in the past three to five years, discussing three key aspects: data generation, model selection, and validation. We emphasize that amorphous/disordered structures with diverse compositions and configurations can effectively represent the intrinsically complex nature of GBs and interfaces, providing valuable data for training MLIPs with enhanced transferability. This approach eliminates the need to directly include GB and interface models and reduces the computational burden of sampling structures from AIMD, thereby increasing the efficiency of MLIP development. Utilizing pre-trained foundational MLIPs and multi-fidelity training can enable efficient generation of high-quality dataset potentially with higher DFT level of theory and accelerate the MLIP development process with improved accuracy. In terms of ML models, most studies have adopted MTP or descriptor-based NNP (particularly DeePMD) to simulate GBs and interfaces. These models have demonstrated the ability to capture highly complex interfaces involving up to seven elements. At the same time, the state-of-the-art GNN-based models are just beginning to emerge for simulations of ASSB interfaces. The use of GNN-based models can be highly beneficial for simulating extremely complex atomic environments, as they offer high accuracy for systems with an increased number of elements, albeit at the cost of computational speed. **The FM models will be also beneficial to screen and validate new materials for ASSBs upon proper validation of their performance.** In addition, utilizing MLIP models incorporating long-range interactions with accurate prediction of spin/charge states can enable more realistic modeling of interfacial evolution under practical electrochemical conditions. Lastly, we strongly recommend thorough validation of ML models for their intended applications,

including assessments of application-specific properties as well as structural, vibrational, and dynamical properties. While most studies evaluate energy and force errors for GB/interface models tractable by DFT, low errors in these metrics do not necessarily ensure that MLIPs can accurately predict structural and dynamic behaviors. Testing ML models by benchmarking these properties against DFT/AIMD will ensure its performance to implement reliable dynamics simulations comparable to first-principles calculations. We also recommend evaluating the scale-up transferability to larger GB/interface models using UQ approaches. Overall, practical guidelines presented in this prospective paper will aid for future developments in MLIPs and acceleration of the rational design of interfaces for next-generation ASSBs.

REFERENCES

- 1 Banerjee, A., Wang, X., Fang, C., Wu, E. A. & Meng, Y. S. Interfaces and Interphases in All-Solid-State Batteries with Inorganic Solid Electrolytes. *Chemical Reviews* **120**, 6878-6933 (2020). <https://doi.org:10.1021/acs.chemrev.0c00101>
- 2 Cheng, L. *et al.* Effect of microstructure and surface impurity segregation on the electrical and electrochemical properties of dense Al-substituted Li₇La₃Zr₂O₁₂. *Journal of Materials Chemistry A* **2**, 172-181 (2014). <https://doi.org:10.1039/C3TA13999A>
- 3 Liu, X. *et al.* Local electronic structure variation resulting in Li ‘filament’ formation within solid electrolytes. *Nature Materials* **20**, 1485-1490 (2021). <https://doi.org:10.1038/s41563-021-01019-x>
- 4 Kalnaus, S., Dudney, N. J., Westover, A. S., Herbert, E. & Hackney, S. Solid-state batteries: The critical role of mechanics. *Science* **381**, eabg5998 (2023). <https://doi.org:10.1126/science.abg5998>
- 5 Scheld, W. S. *et al.* The Riddle of Dark LLZO: Cobalt Diffusion in Garnet Separators of Solid-State Lithium Batteries. *Advanced Functional Materials* **33**, 2302939 (2023). <https://doi.org:https://doi.org/10.1002/adfm.202302939>
- 6 Kansara, S., Kang, H., Ryu, S., Sun, H. H. & Hwang, J.-Y. Basic guidelines of first-principles calculations for suitable selection of electrochemical Li storage materials: a review. *Journal of Materials Chemistry A* **11**, 24482-24518 (2023). <https://doi.org:10.1039/D3TA05042D>
- 7 Jacobs, R. *et al.* A practical guide to machine learning interatomic potentials – Status and future. *Current Opinion in Solid State and Materials Science* **35**, 101214 (2025). <https://doi.org:https://doi.org/10.1016/j.cossms.2025.101214>
- 8 Behler, J. & Parrinello, M. Generalized Neural-Network Representation of High-Dimensional Potential-Energy Surfaces. *Physical Review Letters* **98**, 146401 (2007). <https://doi.org:10.1103/PhysRevLett.98.146401>
- 9 Behler, J. & Csányi, G. Machine learning potentials for extended systems: a perspective. *The European Physical Journal B* **94**, 142 (2021). <https://doi.org:10.1140/epjb/s10051-021-00156-1>
- 10 Zuo, Y. *et al.* Performance and Cost Assessment of Machine Learning Interatomic Potentials. *The Journal of Physical Chemistry A* **124**, 731-745 (2020). <https://doi.org:10.1021/acs.jpca.9b08723>
- 11 Unke, O. T. *et al.* Machine Learning Force Fields. *Chemical Reviews* **121**, 10142-10186 (2021). <https://doi.org:10.1021/acs.chemrev.0c01111>
- 12 Behler, J. Four Generations of High-Dimensional Neural Network Potentials. *Chemical Reviews* **121**, 10037-10072 (2021). <https://doi.org:10.1021/acs.chemrev.0c00868>
- 13 Wang, G. *et al.* Machine learning interatomic potential: Bridge the gap between small-scale models and realistic device-scale simulations. *iScience* **27**, 109673 (2024). <https://doi.org:https://doi.org/10.1016/j.isci.2024.109673>
- 14 Guo, H., Wang, Q., Stuke, A., Urban, A. & Artrith, N. Accelerated Atomistic Modeling of Solid-State Battery Materials With Machine Learning. *Frontiers in Energy Research* **9**, 265 (2021).
- 15 Ohara, K. *et al.* Structural and electronic features of binary Li₂S-P₂S₅ glasses. *Scientific Reports* **6**, 21302 (2016). <https://doi.org:10.1038/srep21302>
- 16 Houchins, G. & Viswanathan, V. An accurate machine-learning calculator for optimization of Li-ion battery cathodes. *The Journal of Chemical Physics* **153**, 054124 (2020). <https://doi.org:10.1063/5.0015872>
- 17 Lee, H.-J. *et al.* Lithium Localization by Anions in Argyrodite Solid Electrolytes from Machine-Learning-based Simulations. *Advanced Energy Materials* **14**, 2402396 (2024). <https://doi.org:https://doi.org/10.1002/aenm.202402396>
- 18 Qi, J. *et al.* Bridging the gap between simulated and experimental ionic conductivities in lithium superionic conductors. *Materials Today Physics* **21**, 100463 (2021). <https://doi.org:https://doi.org/10.1016/j.mtphys.2021.100463>
- 19 Artrith, N. Machine learning for the modeling of interfaces in energy storage and conversion materials. *Journal of Physics: Energy* **1**, 032002 (2019). <https://doi.org:10.1088/2515-7655/ab2060>
- 20 Deringer, V. L. Modelling and understanding battery materials with machine-learning-driven atomistic simulations. *Journal of Physics: Energy* **2**, 041003 (2020). <https://doi.org:10.1088/2515-7655/abb011>
- 21 Chen, X., Liu, X., Shen, X. & Zhang, Q. Applying Machine Learning to Rechargeable Batteries: From the Microscale to the Macroscale. *Angewandte Chemie International Edition* **60**, 24354-24366 (2021). <https://doi.org:https://doi.org/10.1002/anie.202107369>
- 22 Shao, Y., Knijff, L., Dietrich, F. M., Hermansson, K. & Zhang, C. Modelling Bulk Electrolytes and Electrolyte Interfaces with Atomistic Machine Learning. *Batteries & Supercaps* **4**, 585-595 (2021). <https://doi.org:https://doi.org/10.1002/batt.202000262>
- 23 Bhowmik, A. *et al.* Implications of the BATTERY 2030+ AI-Assisted Toolkit on Future Low-TRL Battery Discoveries and Chemistries. *Advanced Energy Materials* **12**, 2102698 (2022). <https://doi.org:https://doi.org/10.1002/aenm.202102698>
- 24 Diddens, D. *et al.* Modeling the Solid Electrolyte Interphase: Machine Learning as a Game Changer? *Advanced Materials Interfaces* **9**, 2101734 (2022). <https://doi.org:https://doi.org/10.1002/admi.202101734>

- 25 Ling, C. A review of the recent progress in battery informatics. *npj Computational Materials* **8**, 33 (2022).
<https://doi.org/10.1038/s41524-022-00713-x>
- 26 Euchner, H. & Groß, A. Atomistic modeling of Li- and post-Li-ion batteries. *Physical Review Materials* **6**, 040302 (2022). <https://doi.org/10.1103/PhysRevMaterials.6.040302>
- 27 Jha, S. *et al.* Machine learning-assisted materials development and device management in batteries and supercapacitors: performance comparison and challenges. *Journal of Materials Chemistry A* **11**, 3904-3936 (2023).
<https://doi.org/10.1039/D2TA07148G>
- 28 Chen, Y. *et al.* Artificial intelligence for the understanding of electrolyte chemistry and electrode interface in lithium battery. *National Science Open* **3**, 20230039 (2024). <https://doi.org/https://doi.org/10.1360/nso/20230039>
- 29 Zhang, J. *et al.* Li₁₀GeP₂S₁₂-based solid electrolyte induced by high pressure for all-solid-state batteries: A facile strategy of low-grain-boundary-resistance. *Chemical Engineering Journal* **487**, 150452 (2024).
<https://doi.org/https://doi.org/10.1016/j.ccej.2024.150452>
- 30 Hasegawa, G., Kuwata, N., Ohnishi, T. & Takada, K. Visualization and evaluation of lithium diffusion at grain boundaries in Li_{0.29}La_{0.57}TiO₃ solid electrolytes using secondary ion mass spectrometry. *Journal of Materials Chemistry A* **12**, 731-738 (2024). <https://doi.org/10.1039/D3TA05012B>
- 31 Cheng, E. J., Sharafi, A. & Sakamoto, J. Intergranular Li metal propagation through polycrystalline Li_{6.25}Al_{0.25}La₃Zr₂O₁₂ ceramic electrolyte. *Electrochimica Acta* **223**, 85-91 (2017).
<https://doi.org/https://doi.org/10.1016/j.electacta.2016.12.018>
- 32 Han, F. *et al.* High electronic conductivity as the origin of lithium dendrite formation within solid electrolytes. *Nature Energy* **4**, 187-196 (2019). <https://doi.org/10.1038/s41560-018-0312-z>
- 33 Pun, G. P. P., Batra, R., Ramprasad, R. & Mishin, Y. Physically informed artificial neural networks for atomistic modeling of materials. *Nature Communications* **10**, 2339 (2019). <https://doi.org/10.1038/s41467-019-10343-5>
- 34 Pun, G. P. P., Yamakov, V., Hickman, J., Glaessgen, E. H. & Mishin, Y. Development of a general-purpose machine-learning interatomic potential for aluminum by the physically informed neural network method. *Physical Review Materials* **4**, 113807 (2020). <https://doi.org/10.1103/PhysRevMaterials.4.113807>
- 35 Nishiyama, T., Seko, A. & Tanaka, I. Application of machine learning potentials to predict grain boundary properties in fcc elemental metals. *Physical Review Materials* **4**, 123607 (2020). <https://doi.org/10.1103/PhysRevMaterials.4.123607>
- 36 Fujii, S. & Seko, A. Structure and lattice thermal conductivity of grain boundaries in silicon by using machine learning potential and molecular dynamics. *Computational Materials Science* **204**, 111137 (2022).
<https://doi.org/https://doi.org/10.1016/j.commatsci.2021.111137>
- 37 Yokoi, T., Matsuura, M., Oshima, Y. & Matsunaga, K. Grain-boundary thermodynamics with artificial-neural-network potential: Its ability to predict the atomic structures, energetics, and lattice vibrational properties for Al. *Physical Review Materials* **7**, 053803 (2023). <https://doi.org/10.1103/PhysRevMaterials.7.053803>
- 38 Yokoi, T., Kato, H., Oshima, Y. & Matsunaga, K. Atomic structures of grain boundaries for Si and Ge: A simulated annealing method with artificial-neural-network interatomic potentials. *Journal of Physics and Chemistry of Solids* **173**, 111114 (2023). <https://doi.org/https://doi.org/10.1016/j.jpcs.2022.111114>
- 39 Zhang, L., Csányi, G., van der Giessen, E. & Maresca, F. Efficiency, accuracy, and transferability of machine learning potentials: Application to dislocations and cracks in iron. *Acta Materialia* **270**, 119788 (2024).
<https://doi.org/https://doi.org/10.1016/j.actamat.2024.119788>
- 40 Ito, K., Yokoi, T., Hyodo, K. & Mori, H. Machine learning interatomic potential with DFT accuracy for general grain boundaries in α -Fe. *npj Computational Materials* **10**, 255 (2024). <https://doi.org/10.1038/s41524-024-01451-y>
- 41 Zhang, B., Chen, E. & Asta, M. Oxygen grain-boundary segregation in HCP Ti — Computational investigations using an atomic cluster expansion potential. *Computational Materials Science* **248**, 113577 (2025).
<https://doi.org/https://doi.org/10.1016/j.commatsci.2024.113577>
- 42 Li, X.-G. *et al.* Quantum-accurate spectral neighbor analysis potential models for Ni-Mo binary alloys and fcc metals. *Physical Review B* **98**, 094104 (2018). <https://doi.org/10.1103/PhysRevB.98.094104>
- 43 Li, X.-G., Xu, S., Zhang, Q., Liu, S. & Shuai, J. Complex strengthening mechanisms in nanocrystalline Ni-Mo alloys revealed by a machine-learning interatomic potential. *Journal of Alloys and Compounds* **952**, 169964 (2023).
<https://doi.org/https://doi.org/10.1016/j.jallcom.2023.169964>
- 44 Dai, F.-Z., Wen, B., Xiang, H. & Zhou, Y. Grain boundary strengthening in ZrB₂ by segregation of W: Atomistic simulations with deep learning potential. *Journal of the European Ceramic Society* **40**, 5029-5036 (2020).
<https://doi.org/https://doi.org/10.1016/j.jeurceramsoc.2020.06.007>
- 45 Yokoi, T. *et al.* Atomic and electronic structure of grain boundaries in α -Al₂O₃: A combination of machine learning, first-principles calculation and electron microscopy. *Scripta Materialia* **229**, 115368 (2023).
<https://doi.org/https://doi.org/10.1016/j.scriptamat.2023.115368>
- 46 Yokoi, T., Adachi, K., Iwase, S. & Matsunaga, K. Accurate prediction of grain boundary structures and energetics in CdTe: a machine-learning potential approach. *Physical Chemistry Chemical Physics* **24**, 1620-1629 (2022).
<https://doi.org/10.1039/D1CP04329C>

- 47 Zhang, L., Han, J., Wang, H., Car, R. & E, W. Deep Potential Molecular Dynamics: A Scalable Model with the Accuracy of Quantum Mechanics. *Physical Review Letters* **120**, 143001 (2018). <https://doi.org/10.1103/PhysRevLett.120.143001>
- 48 Liu, D., Wu, Y., Vasenko, A. S. & Prezhdo, O. V. Grain boundary sliding and distortion on a nanosecond timescale induce trap states in CsPbBr₃: ab initio investigation with machine learning force field. *Nanoscale* **15**, 285-293 (2023). <https://doi.org/10.1039/D2NR05918E>
- 49 Avery, P. & Zurek, E. RandSpG: An open-source program for generating atomistic crystal structures with specific spacegroups. *Computer Physics Communications* **213**, 208-216 (2017). <https://doi.org/https://doi.org/10.1016/j.cpc.2016.12.005>
- 50 Kim, K. *et al.* Probing degradation at solid-state battery interfaces using machine-learning interatomic potential. *Energy Storage Materials* **73**, 103842 (2024).
- 51 Deng, Z., Chen, C., Li, X.-G. & Ong, S. P. An electrostatic spectral neighbor analysis potential for lithium nitride. *npj Computational Materials* **5**, 75 (2019). <https://doi.org/10.1038/s41524-019-0212-1>
- 52 Sergeev, A. V., Rulev, A. A., Kondratyeva, Y. O. & Yashina, L. V. Computational insight into the grain boundary structure and atomic mobility in metallic lithium. *Acta Materialia* **233**, 117988 (2022). <https://doi.org/https://doi.org/10.1016/j.actamat.2022.117988>
- 53 Ou, Y. *et al.* Atomistic modeling of bulk and grain boundary diffusion in solid electrolyte $\text{Li}_{10}\text{PS}_{12}\text{Cl}$ using machine-learning interatomic potentials. *Physical Review Materials* **8**, 115407 (2024). <https://doi.org/10.1103/PhysRevMaterials.8.115407>
- 54 Kim, J. H. *et al.* Highly reliable and large-scale simulations of promising argyrodite solid-state electrolytes using a machine-learned moment tensor potential. *Nano Energy* **124**, 109436 (2024). <https://doi.org/https://doi.org/10.1016/j.nanoen.2024.109436>
- 55 Jalem, R., Chandrappa, M. L. H., Qi, J., Tateyama, Y. & Ong, S. P. Lithium dynamics at grain boundaries of β -Li₃PS₄ solid electrolyte. *Energy Advances* **2**, 2029-2041 (2023). <https://doi.org/10.1039/D3YA00234A>
- 56 Podryabinkin, E. V. & Shapeev, A. V. Active learning of linearly parametrized interatomic potentials. *Computational Materials Science* **140**, 171-180 (2017). <https://doi.org/https://doi.org/10.1016/j.commatsci.2017.08.031>
- 57 Goryaeva, A. M. *et al.* Reinforcing materials modelling by encoding the structures of defects in crystalline solids into distortion scores. *Nature Communications* **11**, 4691 (2020). <https://doi.org/10.1038/s41467-020-18282-2>
- 58 Singraber, A., Behler, J. & Dellago, C. Library-Based LAMMPS Implementation of High-Dimensional Neural Network Potentials. *Journal of Chemical Theory and Computation* **15**, 1827-1840 (2019). <https://doi.org/10.1021/acs.jctc.8b00770>
- 59 Kim, K., Kroonblawd, M. P. & Goldman, N. Similarity Metric for Data Optimization and Efficient Training of Reactive Machine Learning Force Fields for Hydrocarbon Radiolysis. *Journal of Chemical Theory and Computation* **21**, 11079-11092 (2025). <https://doi.org/10.1021/acs.jctc.5c01301>
- 60 Ko, S.-T. *et al.* Compositionally complex perovskite oxides: Discovering a new class of solid electrolytes with interface-enabled conductivity improvements. *Matter* **6**, 2395-2418 (2023). <https://doi.org/https://doi.org/10.1016/j.matt.2023.05.035>
- 61 Lee, T. *et al.* Atomic-scale origin of the low grain-boundary resistance in perovskite solid electrolyte Li_{0.375}Sr_{0.4375}Ta_{0.75}Zr_{0.25}O₃. *Nature Communications* **14**, 1940 (2023). <https://doi.org/10.1038/s41467-023-37115-6>
- 62 Kim, K. *et al.* Flexible machine-learning interatomic potential for simulating structural disordering behavior of Li₇La₃Zr₂O₁₂ solid electrolytes. *The Journal of Chemical Physics* **156**, 221101 (2022). <https://doi.org/10.1063/5.0090341>
- 63 Bösel, L., Thürlmann, M. & Riniker, S. Machine Learning in QM/MM Molecular Dynamics Simulations of Condensed-Phase Systems. *Journal of Chemical Theory and Computation* **17**, 2641-2658 (2021). <https://doi.org/10.1021/acs.jctc.0c01112>
- 64 Musaelian, A. *et al.* Learning local equivariant representations for large-scale atomistic dynamics. *Nature Communications* **14**, 579 (2023). <https://doi.org/10.1038/s41467-023-36329-y>
- 65 Batatia, I., Kovacs, D. P., Simm, G., Ortner, C. & Csanyi, G. MACE: Higher Order Equivariant Message Passing Neural Networks for Fast and Accurate Force Fields. *Advances in Neural Information Processing Systems* **35**, 11423-11436 (2022).
- 66 Sharafi, A., Haslam, C. G., Kerns, R. D., Wolfenstine, J. & Sakamoto, J. Controlling and correlating the effect of grain size with the mechanical and electrochemical properties of Li₇La₃Zr₂O₁₂ solid-state electrolyte. *Journal of Materials Chemistry A* **5**, 21491-21504 (2017). <https://doi.org/10.1039/C7TA06790A>
- 67 Singraber, A., Morawietz, T., Behler, J. & Dellago, C. Parallel Multistream Training of High-Dimensional Neural Network Potentials. *Journal of Chemical Theory and Computation* **15**, 3075-3092 (2019). <https://doi.org/10.1021/acs.jctc.8b01092>

68 Heo, T. W. *et al.* Microstructural impacts on ionic conductivity of oxide solid electrolytes from a combined atomistic-mesoscale approach. *npj Computational Materials* **7**, 214 (2021). <https://doi.org/10.1038/s41524-021-00681-8>

69 Larraz, G., Otera, A. & Sanjuán, M. L. Cubic phases of garnet-type Li₇La₃Zr₂O₁₂: the role of hydration. *Journal of Materials Chemistry A* **1**, 11419-11428 (2013). <https://doi.org/10.1039/C3TA11996C>

70 Feng, L. *et al.* Machine-Learning-Assisted Deciphering of Microstructural Effects on Ionic Transport in Composite Materials: A Case Study of Li₇La₃Zr₂O₁₂-LiCoO₂. *Energy Storage Materials* **73**, 103776 (2024).

71 Monismith, S., Qu, J. & Dingreville, R. Grain-boundary fracture mechanisms in Li₇La₃Zr₂O₁₂ (LLZO) solid electrolytes: When phase transformation acts as a temperature-dependent toughening mechanism. *Journal of the Mechanics and Physics of Solids* **160**, 104791 (2022). <https://doi.org/10.1016/j.jmps.2022.104791>

72 Yuan, S. *et al.* Phase Instability-Coupled Fracture Behavior in Garnet LLZO Solid Electrolytes: A Machine Learning-Enabled Atomistic Study. *Submitted*. (2025).

73 Jeong, W., Yoo, D., Lee, K., Jung, J. & Han, S. Efficient Atomic-Resolution Uncertainty Estimation for Neural Network Potentials Using a Replica Ensemble. *The Journal of Physical Chemistry Letters* **11**, 6090-6096 (2020). <https://doi.org/10.1021/acs.jpclett.0c01614>

74 Achar, S. K., Schneider, J. & Stewart, D. A. Using Machine Learning Potentials to Explore Interdiffusion at Metal-Chalcogenide Interfaces. *ACS Applied Materials & Interfaces* **14**, 56963-56974 (2022). <https://doi.org/10.1021/acsami.2c16254>

75 Chandrappa, M. L. H., Qi, J., Chen, C., Banerjee, S. & Ong, S. P. Thermodynamics and Kinetics of the Cathode-Electrolyte Interface in All-Solid-State Li-S Batteries. *Journal of the American Chemical Society* **144**, 18009-18022 (2022). <https://doi.org/10.1021/jacs.2c07482>

76 Lai, G. *et al.* A Deep Neural Network Interface Potential for Li-Cu Systems. *Advanced Materials Interfaces* **9**, 2201346 (2022). <https://doi.org/10.1002/admi.202201346>

77 Wang, J., Panchal, A. A., Sai Gautam, G. & Canepa, P. The resistive nature of decomposing interfaces of solid electrolytes with alkali metal electrodes. *Journal of Materials Chemistry A* **10**, 19732-19742 (2022). <https://doi.org/10.1039/D2TA02202H>

78 Wang, C., Aykol, M. & Mueller, T. Nature of the Amorphous-Amorphous Interfaces in Solid-State Batteries Revealed Using Machine-Learned Interatomic Potentials. *Chemistry of Materials* **35**, 6346-6356 (2023). <https://doi.org/10.1021/acs.chemmater.3c00993>

79 Lai, G. *et al.* The Mechanism of Li Deposition on the Cu Substrates in the Anode-Free Li Metal Batteries. *Small* **19**, 2205416 (2023). <https://doi.org/10.1002/sml.202205416>

80 Fu, F. *et al.* Unraveling the Atomic-scale Mechanism of Phase Transformations and Structural Evolutions during (de)Lithiation in Si Anodes. *Advanced Functional Materials* **33**, 2303936 (2023). <https://doi.org/10.1002/adfm.202303936>

81 Hu, T. *et al.* Impact of Amorphous LiF Coating Layers on Cathode-Electrolyte Interfaces in Solid-State Batteries. *Advanced Functional Materials* **34**, 2402993 (2024). <https://doi.org/10.1002/adfm.202402993>

82 Ren, F. *et al.* Visualizing the SEI formation between lithium metal and solid-state electrolyte. *Energy & Environmental Science* **17**, 2743-2752 (2024). <https://doi.org/10.1039/D3EE03536K>

83 Fu, F. *et al.* What drives the heterogeneous interdiffusion in the Li-Si interfacial region of Si anodes: the Li flux or the Si flux? *npj Computational Materials* **10**, 131 (2024). <https://doi.org/10.1038/s41524-024-01327-1>

84 Chaney, G., Golov, A., van Rookeghem, A., Carrasco, J. & Mingo, N. Two-Step Growth Mechanism of the Solid Electrolyte Interphase in Argyrodyte/Li-Metal Contacts. *ACS Applied Materials & Interfaces* **16**, 24624-24630 (2024). <https://doi.org/10.1021/acsami.4c02548>

85 You, Y. *et al.* Grain boundary amorphization as a strategy to mitigate lithium dendrite growth in solid-state batteries. *Nature Communications* **16**, 4630 (2025). <https://doi.org/10.1038/s41467-025-59895-9>

86 Jain, A. *et al.* Commentary: The Materials Project: A materials genome approach to accelerating materials innovation. *APL Materials* **1**, 011002-011002 (2013). <https://doi.org/10.1063/1.4812323>

87 You, Y. *et al.* Principal component analysis enables the design of deep learning potential precisely capturing LLZO phase transitions. *npj Computational Materials* **10**, 57 (2024). <https://doi.org/10.1038/s41524-024-01240-7>

88 Zhang, D. *et al.* Exploring the Relationship between Composition and Li-Ion Conductivity in the Amorphous Li-La-Zr-O System. *ACS Materials Letters* **6**, 1849-1855 (2024). <https://doi.org/10.1021/acsmaterialslett.3c01558>

89 Chen, C. & Ong, S. P. A universal graph deep learning interatomic potential for the periodic table. *Nature Computational Science* **2**, 718-728 (2022). <https://doi.org/10.1038/s43588-022-00349-3>

90 Deng, B. *et al.* CHGNet as a pretrained universal neural network potential for charge-informed atomistic modelling. *Nature Machine Intelligence* **5**, 1031-1041 (2023). <https://doi.org/10.1038/s42256-023-00716-3>

91 Batatia, I. *et al.* A foundation model for atomistic materials chemistry. *Submitted* (<https://doi.org/10.48550/arXiv.2401.00096>).

- 92 Riebesell, J. *et al.* Matbench Discovery: A framework to evaluate machine learning crystal stability predictions. *Submitted* (<https://arxiv.org/html/2308.14920v3>) (2024).
- 93 Ko, T. W. & Ong, S. P. Data-efficient construction of high-fidelity graph deep learning interatomic potentials. *npj Computational Materials* **11**, 65 (2025). <https://doi.org/10.1038/s41524-025-01550-4>
- 94 Huang, X. *et al.* Cross-functional transferability in universal machine learning interatomic potentials. *Submitted* (<https://doi.org/10.48550/arXiv.2504.05565>) (2025).
- 95 Ding, J. *et al.* Coupled reaction and diffusion governing interface evolution in solid-state batteries. *Submitted* (<https://doi.org/10.48550/arXiv.2506.10944>) (2025).
- 96 Du, H. *et al.* Assessment and Application of Universal Machine Learning Interatomic Potentials in Solid-State Electrolyte Research. *ACS Materials Letters* **7**, 3403-3412 (2025). <https://doi.org/10.1021/acsmaterialslett.5c00336>
- 97 Deng, B. *et al.* Systematic softening in universal machine learning interatomic potentials. *npj Computational Materials* **11**, 9 (2025). <https://doi.org/10.1038/s41524-024-01500-6>
- 98 Barroso-Luque, L. *et al.* Open Materials 2024 (OMat24) Inorganic Materials Dataset and Models. *Submitted* (<https://doi.org/10.48550/arXiv.2410.12771>) (2024).
- 99 Kong, L., Shim, J., Hu, G. & Fung, V. Scalable Foundation Interatomic Potentials via Message-Passing Pruning and Graph Partitioning. *Submitted* (<https://doi.org/10.48550/arXiv.2509.21694>) (2025).
- 100 Gardner, J. L. A. *et al.* Distillation of atomistic foundation models across architectures and chemical domains. *Submitted* (<https://doi.org/10.48550/arXiv.2506.10956>) (2025).
- 101 Luntz, A. C., Voss, J. & Reuter, K. Interfacial Challenges in Solid-State Li Ion Batteries. *Journal of Physical Chemistry Letters* **6**, 4599-4604 (2015). <https://doi.org/10.1021/acs.jpclett.5b02352>
- 102 Huang, Y., Shao, B. & Han, F. Interfacial challenges in all-solid-state lithium batteries. *Current Opinion in Electrochemistry* **33**, 100933 (2022). [https://doi.org:https://doi.org/10.1016/j.coelec.2021.100933](https://doi.org/https://doi.org/10.1016/j.coelec.2021.100933)
- 103 Anstine, D. M. & Isayev, O. Machine Learning Interatomic Potentials and Long-Range Physics. *The Journal of Physical Chemistry A* **127**, 2417-2431 (2023). <https://doi.org/10.1021/acs.jpca.2c06778>
- 104 Babaei, M. & Sadeghi, A. On machine learnability of local contributions to interatomic potentials from density functional theory calculations. *Scientific Reports* **14**, 31395 (2024). <https://doi.org/10.1038/s41598-024-82990-8>
- 105 Staacke, C. G. *et al.* On the Role of Long-Range Electrostatics in Machine-Learned Interatomic Potentials for Complex Battery Materials. *ACS Applied Energy Materials* **4**, 12562-12569 (2021). <https://doi.org/10.1021/acsaem.1c02363>
- 106 Kutana, A., Shimizu, K., Watanabe, S. & Asahi, R. Representing Born effective charges with equivariant graph convolutional neural networks. *Scientific Reports* **15**, 16719 (2025). <https://doi.org/10.1038/s41598-025-01250-5>
- 107 Hu, T. *et al.* Observation of dendrite formation at Li metal-electrolyte interface by a machine-learning enhanced constant potential framework. *Nature Communications* **16**, 7379 (2025). <https://doi.org/10.1038/s41467-025-62824-5>
- 108 Xu, W. *et al.* Spin-informed universal graph neural networks for simulating magnetic ordering. *Proceedings of the National Academy of Sciences* **122**, e2422973122 (2025). <https://doi.org/10.1073/pnas.2422973122>

Author contribution

K.K.: Conceptualization, Data curation, Formal analysis, Resources, Methodology, Validation, Investigation, Visualization, Writing – original draft, Writing – review & editing. S.Y.: Methodology, Validation, Investigation, Visualization, Writing – review & editing. B.C.W.: Funding acquisition, Project administration, Supervision, Writing – review & editing. L.F.W.: Funding acquisition, Project administration, Supervision, Writing – review & editing.

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Data availability

Not applicable.

Conflict of Interest

On behalf of all authors, the corresponding author states that there is no conflict of interest.