

Developing Classical Interatomic Potentials for Solid Electrolytes

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All-solid-state battery is one of the most promising candidates for the next stage of energy storage systems. With improved safety, increased energy density, longer cycling life, and higher power density, all-solid-state battery is considered as a potential successor and complement to the currently commercialized lithium-ion battery. A key component of all-solid-state lithium-ion battery includes the solid electrolyte materials, which are ceramic-based lithium superionic conductors. Ideal solid electrolyte materials should have high ionic conductivity (>1 mS/cm) at room temperature, which is a rare property for most lithium-containing inorganic materials. In the past few decades, extensive research efforts have been focused on the understanding, design, and development of novel solid electrolytes. However, only a handful of lithium superionic conductors were discovered such as $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$, $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$, $\text{Li}_7\text{P}_3\text{S}_{11}$, Li_3YCl_6 , Li_3YBr_6 , etc.^{1,2}

The unique diffusion property of solid electrolyte materials is governed by their crystal structures at the microscopic scale. Classical molecular dynamics (MD) simulations enable us to do in silico experiments and analyze how the local chemistry environments facilitate fast ionic diffusion at atomistic scale. Besides, MD simulations are also widely used to predict, screen, and validate novel solid electrolyte materials. In this Viewpoint, we highlight the necessity of development and evaluation of classical interatomic potentials for superionic materials. We discuss the importance of both encoding domain-specific knowledge and leveraging the power of data-driven machine learning methods in developing these interatomic potentials. In the end, we provide an outlook of how classical MD simulations may advance our understanding of solid electrolytes and pave the way for all-solid-state battery development in the future.

■ WHY DO WE NEED CLASSICAL MOLECULAR DYNAMICS SIMULATIONS FOR SOLID ELECTROLYTES?

Computation approaches, especially atomistic modeling, have shown to be powerful tools in understanding and design of solid electrolyte materials over the past decade. First-principles calculations and simulations reveal that the fast ionic diffusion in solid electrolytes is closely related to the atomistic structure and local chemical environment of the materials.^{3,4} High throughput calculations are widely deployed to perform large-scale screening of potential candidate solid electrolyte materials or their doped variants.^{4–8} Most of these studies rely heavily on ab initio molecular dynamics (AIMD) simulations to study the diffusion properties of solid electrolytes. By using density

functional theory (DFT) to handle the interatomic interactions, AIMD simulations require few empirical inputs and have been demonstrated in a wide range of material chemistries including sulfides, oxides, chlorides, and halides.^{9–13} However, a major shortcoming of AIMD simulations is their high computational cost, limiting their application within small systems (usually less than 200 atoms) and at high temperatures (usually above 500 K).¹⁴ To acquire an estimation on ionic conductivity at room temperature, extrapolation based on Arrhenius relationship from high temperature data is essential. The extrapolated ionic conductivity at room temperature usually has 1–2 orders of magnitude uncertainty range. Another commonly used approach to study the ion migration barrier is the nudged-elastic-band (NEB) method based on first-principles calculations. Despite that NEB calculations are computationally cheaper than AIMD simulations, NEB calculations require a priori knowledge of the diffusion modes and migration paths. However, many solid electrolytes exhibit quite complicated diffusion modes and may even lack well-defined lattice sites or diffusion paths,⁴ posing difficulties in extracting diffusion properties from NEB calculations (Table 1).

Developing classical interatomic potentials and performing classical MD simulations are attractive alternatives to AIMD simulations, which can potentially alleviate the system size and simulation time limits. Classical interatomic potentials are usually based on simple analytical functions, which are much computationally cheaper than ab initio force fields. Using a 200-atom structure as a benchmark system, a MD simulation step (usually 1–2 fs) based on ab initio force field costs ~ 20 s of computation time, while a classical MD step using a machine-trained interatomic potential costs ~ 2 ms of computation time, which is $\sim 10^4$ -times faster than AIMD (benchmarked on a common 64-core CPU node). The lower computational cost indicates that classical MD simulations can be performed on much larger systems (from 10^2 to 10^6 atoms) and on a much longer time scale (up to a few tens of ns). The extended temporal and spatial scales in classical MD simulations lead to multiple advantages. One can perform

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Table 1. Comparison between Commonly Used Atomistic Modeling Methods in Calculating Ionic Diffusion Properties

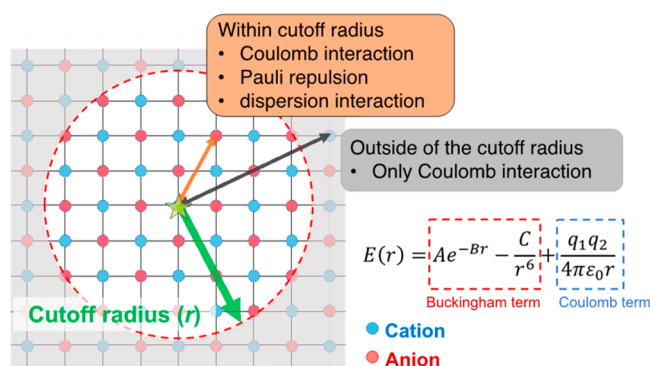
method	NEB (nudged elastic band)	MD (molecular dynamics)	
		AIMD (ab initio MD)	CMD (classical MD)
interatomic potential	DFT	DFT	empirical force field
computational cost	medium	high	low
system size	≤200 atoms	≤200 atoms	10 ² –10 ⁶ atoms
simulation time		≤100 ps–1 ns	≤10–100 ns
temperature range	0 K	usually ≥ 500 K	usually ≥ 300 K
assess room temperature diffusion properties	no	needs extrapolation	yes
migration modes	require a predefined path	applicable to various diffusion modes (no predefined migration paths needed)	
force field accuracy	good	good	poor
systematic error of results	small	small	large
statistic error of results		large	negligible
transferability	good	good	poor

classical MD simulations under lower temperature (or room temperature) directly instead of extrapolation from Arrhenius relationship. The larger scale of the simulation system leads to a decrease in statistic error and less prediction uncertainty. Large pressure fluctuations due to small system size, a notorious problem in AIMD simulations when using the *NpT* ensemble, can be suppressed in classical MD simulations.

Despite these advantages, the major bottleneck of classical MD simulations is the development of interatomic potentials. In contrast to the pseudopotentials used in DFT calculations with very good transferability, empirical interatomic potentials are usually developed case by case with little transferability and only work for specific corresponding material systems. There are few empirical interatomic potentials specifically designed for superionic conductor materials. Existing empirical interatomic potentials for solid electrolyte materials generally show poor performance when diffusion properties are calculated.

■ THE DOWNFALL OF EMPIRICAL INTERATOMIC POTENTIALS FOR SOLID ELECTROLYTES

Solid electrolytes are ionic materials with wide bandgaps and low electronic conductivity. Conventionally, the interatomic interactions in ionic materials are simplified as two-body interactions. Therefore, the empirical interatomic potentials for ionic materials adopt a pairwise form. The pairwise potentials consist of two parts: a long-range term describing the Coulomb interaction, and a short-range term describing the Pauli repulsion and dispersion interaction (Figure 1). In most cases, the Coulomb interaction term assumes nominal valence states of the ions and no empirical parameter fitting is required. The short-range term adopts a simple analytical form with only a few empirical parameters that are relatively easy to fit. Buckingham potential is one of the most widely used short-range term formalisms. Since oxide is the most extensively studied family of ionic materials and the polarization of oxygen ions is critical to multiple physical and chemical properties, core-shell models were later proposed to account for the polarizability of oxygen ions. In core-shell models, a “core”

**Figure 1.** Schematic illustration of the empirical interatomic potential for ionic materials, using Buckingham-Coulomb potential as an example.

particle and a satellite “shell” particle, which are connected by a harmonic spring, divide the charge and mass of the original atom. The relative position of a core-shell pair reflects an induced electric dipole.

Despite its simple analytical form, the conventional empirical potential captures the most important feature of interatomic interaction in ionic solids. Therefore, they show good performance in predicting many physical and chemical properties. The simple analytical form guarantees low computational cost and easy parameter fitting. For each pairwise interaction, only a few empirical parameters need to be fitted. Besides, the pairwise form implies that these interatomic potentials are transferable to some extent. For example, the empirical core-shell parameters of polarizable oxygen ions are transferred between many different oxide systems such as TiO₂, ZrO₂, SiO₂, Al₂O₃, YMnO₃, etc.^{15,16}

Despite their advantages and previous success in studying ionic materials, empirical interatomic potentials confront multiple challenges and failures in studying solid electrolytes. Previously developed empirical interatomic potentials focus extensively on oxides, while solid electrolytes have a variety of different chemistry systems. Besides oxides, solid electrolyte materials include sulfides, nitrides, halides, and hydrides, but empirical interatomic potentials developed for nonoxide systems are rare. Compared to oxide systems where there is a nearly half-century history of empirical potential development, the data and benchmark systems in nonoxide systems are scarce, posing difficulties in studying these nonoxide solid electrolytes.

Even for oxide solid electrolyte materials, empirical interatomic potentials commonly experience underfitting of the potential energy surface and suffer from large deviations when calculating diffusion properties. Unlike other oxide materials, most solid electrolyte materials have a disordered mobile ion sublattice, which correlates to a complicated potential energy surface with many local minima. The ionic diffusion process relates to the transition between these local minima. To simulate the fast ionic diffusion behavior and calculate diffusion properties, the interatomic potentials need to accurately capture the energy surface even when ions are far away from their equilibrium positions. The simple empirical potentials might be sufficient to describe the energy surface near equilibrium positions and to reflect the phonon properties. However, they often fail to capture the energy surface when ions are migrating, leading to unreliable predictions on diffusion properties (Figure 2). As an example,

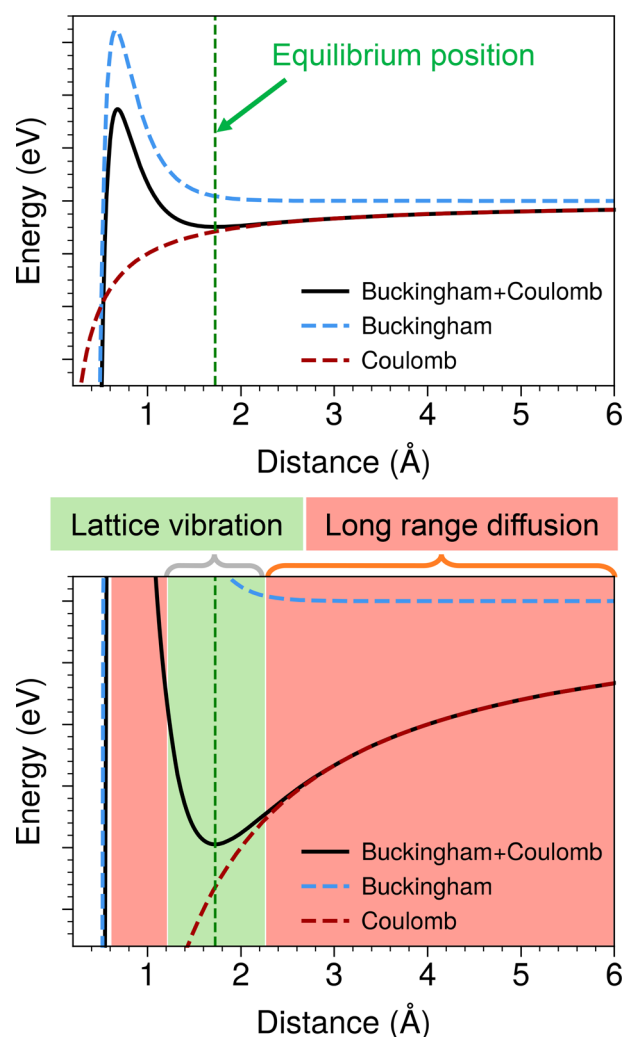


Figure 2. (a) Schematic energy contribution from the long-range Coulomb term and the short-range Buckingham term in the Buckingham-Coulomb interatomic potential. The green dashed line refers to the equilibrium interatomic distance, which is the local minimum of the Buckingham-Coulomb potential (black line). (b) An inset of the energy contribution. The energy surface near the equilibrium position mainly corresponds to lattice vibration (phonon property), while the long-range diffusion is more closely related to the region where ions are far away from equilibrium sites. The green shaded and red shaded region represents where the empirical potentials are accurate and inaccurate, respectively.

we compiled various reports from literature on diffusion properties of $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (Figure 3). The calculated activation energy from AIMD is 0.24 and 0.26 eV, which is close to the experimental measurement (0.26 eV). However, the reported activation energies from classical MD simulations are 0.14 eV, 0.19 eV, 0.48 eV, showing large deviations from AIMD or experimental results. Even the best fitted empirical potentials from our previous work, which were specifically optimized to match the experimental activation energy, still have 1 order of magnitude error on room temperature conductivity. Since the statistic error from classical MD is expected to be negligible, such deviations must originate from underfitting of the potential energy surface, which leads to inaccurate and unreliable predictions on diffusion properties of superionic conductors.¹⁷

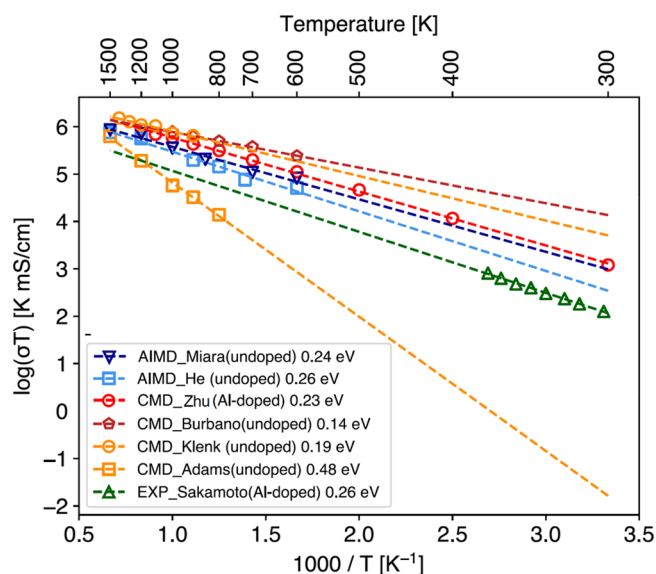


Figure 3. Arrhenius plot of ionic conductivity of $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ extracted from literature reports including experimental results (EXP),¹⁸ ab initio molecular dynamics simulation (AIMD),^{4,19} and classical molecular dynamics simulation (CMD).^{20–23} The activation energy listed in the legend is either from the corresponding literature if it is available or linear fitted if it is unavailable from literature.

In conclusion, the empirical interatomic potentials have an elegant and simple analytical form and capture most of the properties of interatomic interactions in ionic solids. However, empirical potentials suffer from underfitting when studying the diffusion properties of solid electrolytes. Few available empirical potentials exist for nonoxide solid electrolyte material systems, limiting the application of classical MD simulations in sulfides, nitrides, and halides chemistries. Besides, empirical interatomic potentials exhibit underfitting of potential energy surface where mobile ions are far away from equilibrium positions. The inaccurate description of potential energy surface at the transition states leads to failures in calculating diffusion properties.

■ THE RISE OF MACHINE TRAINED CLASSICAL INTERATOMIC POTENTIALS

In the past two and half decades, machine learning (ML) interatomic potentials have emerged as a new type of classical interatomic potentials with rapidly growing research interests. Unlike their empirical predecessors with a predetermined physical form of the atomic interactions, ML potentials use a data-driven approach and “learn” the interatomic interactions directly from the reference data. The universal approximation theorem guaranteed ML functions are able to approximate any continuous functions on a closed interval, while the interatomic potential is nothing more than a continuous function mapping atom coordinates to potential energy. In principle, ML functions are universal approximators and the performance is only limited by the quality and quantity of the training data. Therefore, a practical approach is to train ML potentials based on large data sets generated by first-principles calculations. Once the training is successfully finished, machine trained potentials can greatly accelerate MD simulations as classical interatomic potentials but maintain the accuracy level close to first-principles-based potentials. By overcoming the shortcomings of low accuracy from empirical force fields and

high computational cost from ab initio force fields, machine trained potential has gained its popularity in recent years and demonstrated itself in multiple chemistry systems.²⁴

Despite its rapid evolving, machine training potentials in ionic materials, especially for superionic conducting materials, are still preliminary. A few training frameworks and implementations were established in the last decades such as GAP, SNAP, MTP, NNP, etc.^{24,25} However, most ML potentials are trained and tested in relatively simple chemistry systems. For example, pure silicon as a benchmark model system is extensively tested by multiple machine training frameworks. Extension to systems with more than four types of atoms, which is common for solid electrolytes, is relatively rare. Besides, most of currently benchmarked systems are covalent and metallic material systems, which have very different bonding types from ionic systems. In covalent and metallic material systems, the typical cutoff radii converge well within a few Angstroms. In contrast, the dominating interaction in ionic materials is the electrostatic force, which is a long-range interaction and decays very slow with distance. The dipole and polarization effect are more important in ionic materials than metallic or covalent materials. Another unique feature of superionic conductor materials is their mobile ion sublattice. Compared to ordered solid crystalline materials, the disordered sublattice is a reflection of many local minimums of the potential energy surface. The long-range diffusion relates to the transition between these local minimum states. To correctly capture the diffusional properties, a trained ML potential should be able to capture the potential energy surface with sufficient sampling of all these local minima and the transition states. Unlike mechanical or thermal properties, which are determined by the local vibration when atoms are near equilibrium position, diffusion properties are mostly determined by the transition states. Since ion hopping is a thermally activated rare event in solids, sufficient sampling on transition states is essential for building an accurate machine learning force field. Besides, generalizability between different sized systems is required.²⁶ The training data from first-principles calculation are usually generated on small systems, and the successfully trained machine learning potential should be applicable to larger systems with accurate predictions.

CONCLUSION REMARKS: ENCODE MATERIAL-SPECIFIC PROPERTIES INTO MACHINE LEARNING POTENTIALS

Classical molecular dynamics simulation is an indispensable approach to studying solid electrolyte materials, especially the ionic diffusion properties. In this Viewpoint, we highlight the importance of developing accurate and efficient interatomic potentials, which is crucial to acquire reliable predictions from classical molecular dynamics simulations. Conventional empirical interatomic potentials have low computational cost and are relatively easy to fit. However, the accuracy of their potential energy surface is poor, leading to large deviations when calculating diffusion properties. Besides, few empirical potentials exist for nonoxide systems, posing difficulties when studying sulfide, halide, and nitrides solid electrolytes. Machine learning interatomic potentials have the capability to achieve both low computational cost and highly accurate potential energy surface. However, instead of using general machine learning potential training frameworks as a “black box”, careful considerations on material-specific properties need to be taken in the training process to ensure a successful training. For solid

electrolytes, the ionic bonding, superionic diffusion, disorder sublattice, and anion polarization are material-specific properties that need to be considered when choosing hyperparameters and reference data sets. In addition, an evaluation scheme for machine trained solid electrolyte interatomic potentials should be established to assess their performance from multiple dimensions. Here we propose to evaluate the potentials in terms of accuracy, static properties, diffusion properties, and computational cost (Figure 4). We believe that

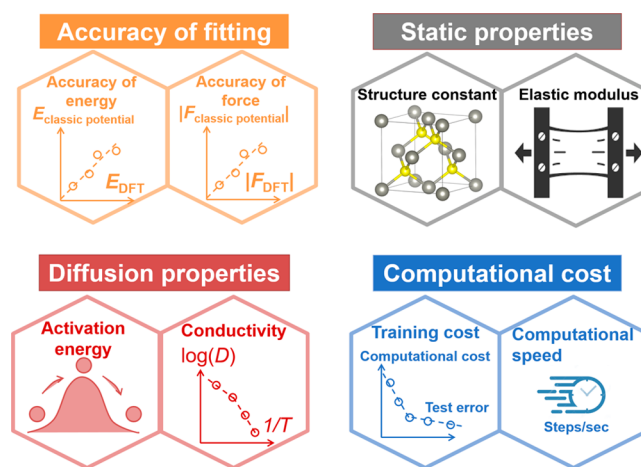


Figure 4. Proposed evaluation scheme for machine trained interatomic potentials in solid electrolyte materials systems including accuracy of fitting, static properties, diffusion properties, and computational cost.

encoding material-specific properties into the training and evaluation process of machine learning interatomic potentials is the key to building reliable classical interatomic potentials for solid electrolytes, which would be an indispensable tool in the understanding and design of solid electrolytes, paving the way to all-solid-state batteries.

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Notes

The authors declare no competing financial interest.

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Yizhou Zhu is currently an assistant professor at the School of Engineering at Westlake University. She obtained her B.S. (2011) and M.E. (2014) degree from Peking University and a Ph.D. degree from the University of Maryland (2018). She was a postdoctoral associate at the Northwestern University for three years and joined Westlake University in 2021. Her current research interest focuses on computational materials, battery materials design, and machine learning force field developments.

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