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Perspective

# Machine Learning Interatomic Potentials for Modeling Solid-State Batteries

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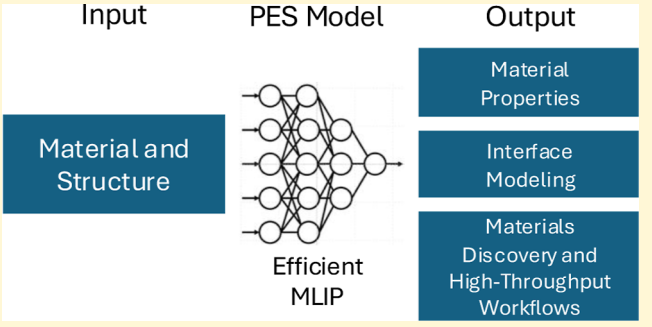
**ABSTRACT:** Atomistic simulations can provide critical insights into the fundamental behavior of battery materials. A necessary input for such simulations is a description of the potential energy surface (PES). Over the past decade, machine learning interatomic potentials (MLIPs) have emerged as a powerful approach to model the PES at near-first-principles accuracy with orders of magnitude lower computational cost. In this perspective, we examine the role of atomistic simulation in battery research, especially in the context of the capabilities afforded by MLIPs. We outline their advantages and trade-offs relative to *ab initio* methods and discuss how they enable the prediction of key intrinsic properties relevant to battery materials, especially for solid state batteries. We further explore their potential for modeling interfaces and chemical reactivity, as well as their integration into high-throughput screening and materials discovery workflows. Finally, we highlight promising applications and identify requirements for the next generation of MLIPs to advance the study of battery materials.




## 1. ATOMISTIC MODELING FOR DEVELOPING THE NEXT GENERATION OF BATTERY TECHNOLOGY

Batteries play an increasingly important role in electrification, electronic devices, and improving energy resiliency.[1] The development of all-solid-state batteries (ASSBs) shows promise for improved energy density, power density, and safety compared to conventional lithium−ion batteries.[2] In parallel, there is a growing interest in post-lithium battery technologies using cheaper and more earth-abundant charge-carrying ions, such as sodium[3] and zinc.[4]

Despite their promise, ASSBs and emerging post-lithium chemistries face significant engineering challenges. The defining component of ASSBs is the solid electrolyte, which must meet stringent criteria for room-temperature ionic conductivity (typically exceeding 0.1 mS $cm^{-1}$)[2] and electrochemical stability against both metal anodes and high-voltage cathodes.[5,6] Because few single-phase materials meet these criteria simultaneously, composite electrolytes have emerged as a viable solution, but with increased complexity as more interfaces are introduced.[7] Furthermore, the chemo-mechanical properties of the solid electrolyte play a critical role in cell cycle life. The cycle life of ASSBs remains significantly lower than their liquid-electrolyte counterparts, largely due to a failure to maintain intimate contact at solid−solid interfaces due to the volume changes that occur during electrochemical cycling.[8] This contact loss necessitates the application of external stack pressure.[2] However, the physical clamping infrastructure required to maintain this interfacial contact penalizes the energy density gained from metal electrodes at the pack level.

The vast compositional space of materials capable of addressing these issues remains largely unexplored, particularly for post-lithium alternatives. Developing strategies to synthesize these materials at an industrial scale also remains a major challenge.[9] Addressing these challenges requires a deep, atomistic understanding of ionic transport, interfacial stability, reaction pathways and many other properties across multiple length and time scales.[10]

Atomistic modeling is a powerful tool in the design and discovery of battery materials. Figure 1a provides an overview of the wide range of battery-relevant properties that can be predicted using atomistic simulations. Such capabilities enable applications such as discovering novel materials,[5,11] probing ionic transport mechanisms,[12−14] understanding interfacial and grain-boundary phenomena,[15−17] performance-guided design[18,19] and many more. However, many of the most important

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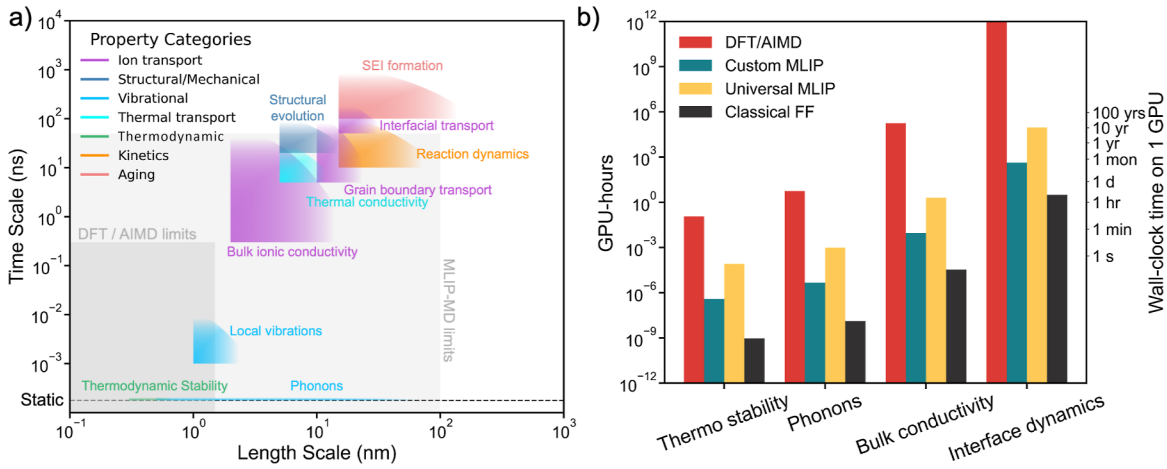


**Figure 1.** (a) Characteristic length and time scales associated with different property categories relevant to battery materials. The bottom-left corner of each shaded region estimates the minimum system size and simulation time typically required to evaluate the property, while the gradient shading indicates that extending to larger cells and longer trajectories improves statistical accuracy and realism. gray overlays show the approximate upper bounds of DFT/AIMD and MLIP-MD. (b) Estimated computational cost of representative property calculations expressed in GPU-hours (left axis, log scale) and equivalent wall-clock time on a single GPU (right axis). Costs were derived by converting each property's minimum length and time requirement into atom counts and MD steps as described in the Supporting Information. It should be noted that actual costs are dependent on chemistry, model architecture, algorithm implementation and hardware, and these numbers should be interpreted as order-of-magnitude estimates based on current technologies.

challenges in battery design occur on spatiotemporal scales that remain inaccessible to *ab initio* methods. These challenges range from accurately representing amorphous, polycrystalline, and disordered materials, to sampling long enough for rare events such as reactions and ionic conduction. This gap can be bridged with modern machine learning interatomic potentials (MLIPs), which act as fast surrogate predictors for the quantum-mechanical potential energy surface (PES), enabling atomistic modeling of battery phenomena at previously intractable scale, complexity, and near *ab initio* accuracy.

The PES maps atomic configurations ($\{\vec{\mathbf{R}}_i\}$) to a scalar energy ($E(\{\vec{\mathbf{R}}_i\})$) and its derivatives, which provide the forces on the atoms and the stress tensor. The PES can also depend on external fields such as electric fields as well as degrees of freedom such as spin, which are important when considering magnetic elements commonly used in cathode materials. Once specified, the PES enables the prediction of material properties, such as those listed in Figure 1.

Modeling the PES involves a trade-off between accuracy and computational cost. Although the underlying many-body Schrödinger equation governing the PES is known, it cannot be solved exactly for most materials. Hence, varying levels of approximation are made to achieve predictions within a reasonable cost. The optimal choice of approximation has changed over time with increasing computational power and theoretical advancements, generally enabling higher accuracy per unit of computational cost. Improvements in algorithms and hardware, including the widespread adoption of graphical processing units (GPUs), have shifted the practical boundaries of simulation. Today, three broad categories of PES models are employed in atomistic materials research: *ab initio* methods, empirical potentials, and MLIPs.

### 1.1. *Ab Initio* Methods and Empirical Potentials

*Ab initio* methods provide the highest-fidelity description of the PES, with density functional theory (DFT) serving as the most commonly used approach for atomistic modeling of battery materials. DFT captures quantum mechanical effects that empirical potentials cannot[20] and remains the method of choice for studying materials where structural or electronic properties are not well understood. However, its steep cubic computational scaling with the number of electrons restricts simulation cell sizes to $<10^3$ atoms and time scales of up to ~1 ns ($O(10^6)$ MD steps). As a result, properties that can only be reliably predicted with larger unit cells or long-time statistical sampling are beyond reach or extrapolated from idealized simulations as shown in Figure 1. Although linear scaling ($O(N)$) DFT methods are an active area of research, they have yet to replace conventional plane wave approaches for routine large-scale simulation.[21] In cases where large spatiotemporal scales are required in simulations, the classical approach is to use empirical interatomic potentials (IPs), also known as force fields. Their simpler functional forms and exceptional computational efficiency enable simulations at the largest scales,[22] but their rigid functional forms and small parameter sets often limit accuracy and transferability across chemical environments.[23] They also struggle to capture chemical reactivity, although reactive models such as ReaxFF have extended their applicability in this area.[24] Another challenge is that fitting empirical potentials is typically system-specific and nontrivial, sometimes requiring a mix of experimental and computational data. Many of the challenges of using *ab initio* methods and empirical potentials are addressed by machine learning IPs (MLIPs) which offer a favorable trade-off between accuracy and computational cost while being fit with relative ease.

### 1.2. Machine Learning Interatomic Potentials and Foundation Potentials

Trained on *ab initio* data, MLIPs use machine learning models to capture complex quantum mechanical interactions. They are orders of magnitude faster and linearly scaling with respect to the number of atoms. Simulation boxes containing ~10,000 atoms and simulation times of nanoseconds are readily accessible via MLIP-MD simulations on a single GPU.[25,26] Although MLIPs are still 1−2 orders of magnitude slower than empirical IPs,[26] their often superior accuracy and flexibility make


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| Desirable feature | Ab initio | Custom MLIP | Foundational MLIP |
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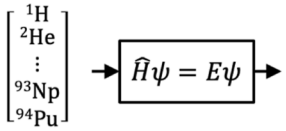
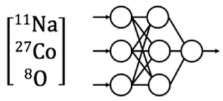
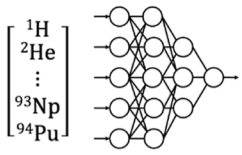
| |  |  |  |
| Ease of running calculation | Steep learning curve, requires high performance computing (HPC) | Expertise in ab initio methods and HPC to generate training data before running easily | Easy to run out-of-the-box on a laptop in most cases |
| Low training data cost | No training data necessary | Need high quality ab initio dataset and hyperparameter tuning | None, unless finetuning necessary |
| Low validation cost | Only convergence of calculation settings | Validation tests against ab initio results necessary | Validation tests against ab initio necessary |
| Low run time cost | Expensive numerical solvers | Cheap, flexible model architecture | Cheap model architecture |
| High generalizability across periodic table elements | All elements and any material class in principle | Only elements and phases in training with very poor accuracy out-of-distribution | Only elements included in training, but generally have qualitative accuracy |
| High accuracy/level of theory | Variable but always superior to MLIP trained on same settings. | Bounded by ab initio accuracy, only reliable in-distribution | Bounded by ab initio accuracy, may need finetuning for quantitative accuracy |
| Detailed electronic structure | Available from most methods | Not available | Not available |

**Figure 2.** A comparison of some different methods for modeling the potential energy surface. The darker the shading, the better the method on the corresponding feature compared to the other methods.

them powerful tools for modeling battery phenomena where these advantages outweigh the added cost, as shown in Figure 2.

MLIPs can be broadly categorized by their breadth of application.

Initially, custom MLIPs trained on a narrow chemical space were the most common. Most custom MLIPs utilize local environment descriptors with simpler ML architectures such as linear regression, kernel methods, or feedforward neural networks. Rapid improvements in local environment descriptors led to MLIPs with errors close to the noise levels of the underlying DFT data and many successful applications.[27,28] Popular MLIP architectures include the atomic cluster expansion (ACE),[29] Gaussian approximation potential (GAP),[30] moment tensor potential (MTP),[31] among others. However, a clear limitation of custom MLIPs is their poor transferability across various chemical systems.

In 2022, Chen et al. demonstrated that graph neural network architectures such as M3GNet[32] could be trained on large data sets such as the entire set of Materials Project relaxation trajectories[33] to develop MLIPs with coverage of almost the entire periodic table. Such foundation potentials (FPs), also known as universal or general-purpose MLIPs, have experienced rapid progress, both in terms of architectural and data set developments. Popular architectures include M3GNet,[32] CHGNet,[34] MACE[35] and UMA,[36] and state-of-the-art quality PES data sets include the MatPES[37] and OMat24[38] data sets for materials, OMol25 data sets for molecules,[39−42] OC22 for catalysts[36,43] and OpenPolymers26 for polymers.[44]

A key advantage of FPs is the ease with which one can run meaningful or exploratory simulations on basic hardware without high performance computing. While far more generalizable, FPs tend to have at least an order of magnitude more trainable parameters than custom MLIPs and are therefore slower and more memory intensive for inference. In addition, FPs are typically not as accurate within a small chemical space as a custom MLIP trained specifically for a material. Fine-tuning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FPs on a small data set targeted for a specific material such as a sodium oxyhalide solid electrolyte[45] or Mn-rich disordered rocksalt cathode material[46] is a popular approach to balance generalizability and accuracy in the relevant chemical space without needing as much new training data.[35] Best practices for reliable fine-tuning,[47,48] as well as methods such as model distillation that can improve efficiency, remain active areas of research.[49,50]

A different approach to MLIPs worth noting is the class of delta-learning approaches where MLIP-like models are trained to correct the predictions from cheaper potentials.[51,52] When coupled with cheap electronic structure methods such as tight-binding schemes,[53] this has advantages over pure MLIP-based PES models because one also gets electronic structure in addition to structure and energetics at lower cost than DFT.[51] Such an approach has been used to study dielectric properties of lithium-graphite intercalation compounds.[54] In addition to fitting the MLIP, the tight-binding model itself needs to be fit to DFT before use.

**1.2.1. Importance of Data Sets.** An MLIP is only as good as the underlying training and validation data, unless physical principles are built-in. In a conceptual sense, the "perfect" MLIP will reproduce the underlying *ab initio* data exactly. It is therefore important for this data to be of the highest quality and fidelity possible.

Quality is determined first by the sampling of atomic environments which must have the correct balance of generalizability and focus for the intended application. Multiple approaches exist for generating diverse MLIP data, each with different trade-offs as extensively discussed in the literature.[55,56] Two popular approaches are active learning and subsampling.

Active learning strategies, in which new training configurations are iteratively selected from the most uncertain or out-of-distribution regions of configuration space, offer a principled route to improve custom and fine-tuned MLIPs with minimal *ab initio* cost.[57,58] Uncertainty quantification methods, including committee models,[58] conformal prediction,[59,60] and Bayesian inference,[57] are increasingly integrated into MLIP workflows to flag unreliable predictions and guide adaptive retraining, which is critical for the safe deployment of MLIPs. However, the sequential nature of the active learning workflow can often be slow and costly due to repeatedly training models. Hence, selective subsampling of long trajectories given by cheaper, low fidelity simulations at different conditions to maximize diversity is another popular approach.[50] The subsampled data set is then labeled with an *ab initio* method to generate the training data without the need to run long AIMD trajectories. However, the diversity of the data in this case depends on the quality of the sampling done by the low fidelity model.

Fidelity is determined by the level of theory used in generating the *ab initio* data set and quality is further determined by the numerical convergence of that data set. The choice of exchange correlation functional and any added corrections is critical for the accuracy relative to experiment.[61] For example, choices of functional have been shown to influence predicted thermodynamic stability,[33] and ionic diffusivity,[62] among other properties. Large data sets for training FPs are being created with better functionals such as the r$^2$SCAN functional[37] for solids and the range-separated hybrid meta-GGA $\omega$B97M-V[63] for molecules[39] since the inference cost of MLIPs is independent of the functional used. In addition, the choice of numerical convergence parameters such as plane wave cutoffs and convergence of reciprocal space grids also impacts the quality of data. Kuryla et al. have shown that poor electronic convergence can lead to erroneous forces.[64] Inconsistencies in corrections such as DFT + $U$ can also lead to errors.[65] Thorough consideration of calculation parameters for training data is therefore important, especially for production simulations.

Incorporation of experimental data into MLIP training is an active area of research. Thaler et al. introduced a differentiable trajectory reweighting approach to train MLIPs from experimental data such as elastic constants and radial distribution functions.[66] Others have explored including vibrational data in the context of molecules.[67] Large, high quality experimental data sets compatible with MLIP training would significantly improve these approaches and provide valuable benchmarks, particularly to understand what physics is necessary to incorporate into MLIP architectures.[68,69]

## 2. MLIPS FOR STUDYING SOLID-STATE BATTERY PROPERTIES

In this perspective, we discuss how MLIPs can be used to model key properties relevant to ASSBs. We provide a short discussion on how MLIPs have been applied in the literature as well as a perspective on potential new applications. We first discuss intrinsic material properties of interest to battery research before discussing how MLIPs can improve our ability to model interfaces and reactions. We then discuss how MLIPs are useful in the context of accelerating materials discovery and as key components of high-throughput workflows. While our discussion focuses primarily on ASSBs, many of the concepts and methodologies are broadly applicable to other battery technologies, including post-lithium and liquid-electrolyte systems. Throughout, we speculate on the most interesting applications of MLIPs and exciting research directions in the development of the next generation of MLIPs, which we summarize in the conclusions.

MLIPs can be applied to a wide range of phenomena related to batteries. The detailed derivation of the phenomenological equations governing the relationship between various battery properties and the PES has been extensively covered in prior reviews.[10,70,71] For the sake of brevity, we will not reproduce those derivations here, and interested readers are referred to those works for details. Ultimately, an MLIP or FP is simply a surrogate for *ab initio* methods in reproducing the PES. As a conceptual framing, we will assume that an MLIP can be trained to arbitrary accuracy in reproducing the PES. In practice, MLIPs should always be validated against *ab initio* results. It should be noted that there are battery-relevant properties that are not directly accessible from the PES, such as charge distribution, magnetic moments, electronic structure, etc. Incorporation of properties beyond the PES in MLIPs is an active area of research. Although we acknowledge that the accuracy of the theoretical and statistical analysis methods is critical, we focus on situations where improved representation of the PES is advantageous, independent of the analysis.

### 2.1. Intrinsic Material Properties

**2.1.1. Phase Stability.** Thermodynamic properties provide a comprehensive map of stable and metastable phases as a function of macrovariables such as composition, temperature, pressure, etc. They serve as indicators for the synthesizability of battery materials. Reaction pathways can be elucidated by traversing the thermodynamic energy landscape, selecting reactants and intermediates on the basis of their relative stability and changes in formation energy. This is an approach


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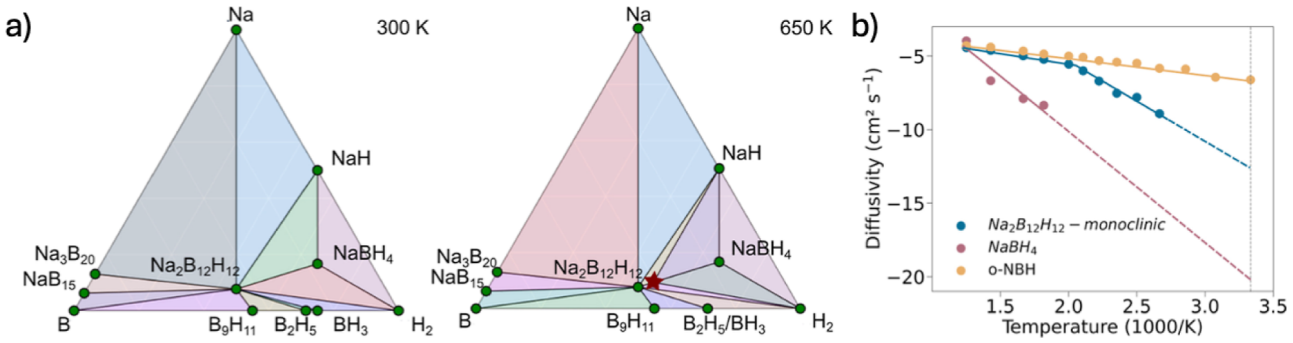


**Figure 3.** (a) MLIPs have been used to predict stability in sodium closo-hydridoborates at finite-temperature, which reveals a different phase diagram from 0 K with a new, highly conducting phase called *o*-NBH with composition $Na_3(B_{12}H_{12})(BH_4)$. Such a prediction would be challenging in DFT due to the dependence on temperature and a relatively large unit cell. (b) MLIPs were also used to characterize sodium ion transport, which involves non-Arrhenius behavior for $Na_2B_{12}H_{12}$ below 500 K which would be challenging to predict by extrapolation from high-temperature AIMD. Images adapted with permission from Oh et al.[72] Copyright 2025 Elsevier.

exemplified by the selectivity metric of McDermott et al.[73,74] Current DFT-based methods are inherently limited by the number of data points within a compositional window. In contrast, FPs can accelerate the exploration of new phases in a chemical system and evaluate reaction pathways orders of magnitude faster than DFT. MLIPs can also account for the effect of temperature on stability, accounting for vibrational entropy,[75−77] more in line with reality, at a dramatically lower computational cost than DFT.[78−80] A practical example is shown in Figure 3, where finite temperature phase diagrams from MLIP simulations were used to identify the temperature at which a novel sodium closo-hydridoborate solid electrolyte is stabilized. Simulations at increased length scales aid in designing synthesis protocols for desired phases and predicting decomposition products at interfaces, enabling phase diagram calculations involving nonstoichiometric or dilute phases, which are not feasible with DFT alone, thereby improving predictions for stable and effective battery materials. Although FPs and fine-tuned MLIPs are useful and should arguably always be used first, final phase stability predictions should always be validated against *ab initio* methods, since MLIPs have errors, especially for materials that are out-of-distribution.

**2.1.2. Ionic Conductivity.** Ionic transport underpins the performance of solid-state electrolytes and electrodes and remains a key factor determining the rate capability in batteries.[81−83] MLIPs now make it possible to simulate these complex transport environments by accessing spatial and temporal regimes far beyond the reach of ab initio methods, allowing mechanistic studies of ionic transport that were previously infeasible.[84]

In the absence of solid−solid interfaces, bulk ionic transport is often the rate-limiting step and plays a key role in determining the overall rate capability of solid-state battery materials.[82] Tracer diffusivity, which quantifies the average motion of a representative ion species at equilibrium, is a key metric for quantifying ionic mobility.[85] It is typically computed from the mean squared displacement of ions in MD simulations. AIMD is limited in accessible time and length scales,[86] leading to limited prediction accuracy due to poor statistical convergence, as shown in Figure 1a. MLIPs are orders of magnitude cheaper than *ab initio* methods, and linear scaling with respect to the number of atoms. Based on current hardware, nanosecond time scales with nanometer-sized simulation boxes with thousands of atoms on a single GPU are readily accessible, with greater length and time scales possible through parallelization. This makes MLIPs especially suited for systems with slower dynamics[87] e.g., <0.1 mS $cm^{-1}$ and/or large-scale structural complexity such as amorphous or polycrystalline materials.

For some materials where there are transitions between Arrhenius regimes, MLIPs can also enable direct simulation of the room temperature ionic conductivity,[84] such as the one shown in Figure 3b or observed in several other solid-state electrolytes.[12] Repetitions of simulations with different initial conditions and structural disorder also allow for more robust statistical error estimates on predictions of ionic conductivity.[85,87] Modeling room-temperature transport with AIMD is extremely challenging even for fast ion conductors without extrapolation using the Arrhenius relation from higher temperatures. Many solid electrolytes exhibit non-Arrhenius behavior, often associated with order−disorder transitions or the temperature-activated opening of diffusion pathways that are inactive at lower temperatures.[12,88,89] For metastable or low-melting-point materials, elevated-temperature simulations can trigger phase transitions, which also invalidates the Arrhenius extrapolation.

Tracer diffusion coefficients, in the absence of thermal, electric, and concentration gradients, are often approximated by the self-diffusion coefficient, assuming that ions diffuse independently.[87,90] However, in solids, correlated ion motion such as concerted hopping can significantly alter the true transport behavior due to different values of the Haven ratio.[91] MLIPs can access nanosecond time scales to quantify ion−ion correlations directly. This is particularly important for densely packed or disordered solids, where correlations often suppress or accelerate net ion displacement leading to a Haven ratio not equal to unity.[14,92] However, in most solids, ion−ion correlations often suppress net ionic conductivity, leading to Haven ratios below one.[93] Accurately capturing ion−ion correlations requires an explicit calculation of cross-correlation terms, which has become feasible with MLIP-accelerated simulations.[70] Furthermore, charge transfer plays an important role in determining ionic conductivity.[94] More recent MLIP frameworks incorporate charge-equilibration schemes or directly infer atomic charges from the local chemical environment. Examples of MLIP architectures include those trained to reproduce charge density or inferred atomic partial analyses,[95−97] those trained on closely related descriptors such as magnetic moments,[34] or those architectures which implicitly derive and equilibrate latent space charges.[98,99] In the near future, we anticipate that these approaches will enable more


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consistent and transferable predictions of charge-related phenomena, particularly in systems with variable oxidation states, as occurs in battery materials. For such studies, robust data sets for training and validation and benchmarks with charge information are necessary.

In addition to diffusion coefficients, activation energy barriers are used to characterize ionic mobility,[100] particularly in systems where transport can be characterized by discrete hopping events, as is the case in many electrodes and solid electrolytes. Experimentally, these barriers are often extracted from temperature-dependent electrochemical impedance spectroscopy by fitting conductivity data to an Arrhenius relation.[101] MLIPs offer a route to efficiently evaluate diffusion barriers for distinct diffusion mechanisms using path optimization techniques such as nudged elastic band[102] methods. *Ab initio* approaches can only consider a small subset of migration paths due to computational costs. MLIPs, on the other hand, enable high-throughput evaluation of a wider set of pathways and materials, as has been done for solid electrolytes.[103] This increases coverage of the energy landscape without prohibitive computational cost. However, caution must be taken as FPs are known to poorly predict out-of-equilibrium and transition states; therefore, thorough benchmarking is necessary.[104] When sufficiently long MD trajectories are available, activation energies can also be extracted from Arrhenius fits to temperature-dependent diffusivity.[12]

Enhanced sampling techniques, such as metadynamics[105] coupled with MD, further aid in resolving rare events at low temperature or that have high activation barriers. The two approaches have been combined to study ionic conductivity,[106] defect dynamics[107] and surface reactions.[108] Developing sampling techniques that complement MLIPs is therefore an exciting research direction since data sets for FPs do not explicitly sample rare events. It is therefore important to validate and fine-tune if necessary.

Already, MLIPs are giving new insights into ionic conductivity due to increased spatiotemporal scales. At the atomic scale, MLIPs are currently the most successful approach to predicting and characterizing ionic conductivity.[12] We anticipate the accuracy will improve with improved data sets based on better exchange−correlation functionals[37] and incorporation of more physics such as electric fields, long-range interactions and charge transfer. Alongside the improvement of MLIPs themselves, developments of analysis techniques that take advantage of these simulations can also give insights into ionic conduction phenomena such as correlated motion.

**2.1.3. Open Circuit Voltage.** The open circuit voltage (OCV) is a key battery parameter, particularly for cathode materials. It is often of interest to predict the voltage as a function of the state of charge, as shown in Figure 4. To do so, one has to identify the lowest energy structure at each composition and compute the corresponding free energy. DFT predictions usually only consider the enthalpy at 0 K over a limited set of compositions due to computational cost, neglecting the effect of temperature. To incorporate temperature and cover a wide range of compositions, approximations using cluster expansions[109] are often used based on parameters fitted from DFT data. With MLIPs, the free energy can also be predicted for finite temperature, including the vibrational and configurational entropy, giving more realistic results and can even be used in Monte Carlo simulations in place of cluster expansions. Furthermore, the ability to model larger unit cells allows for using a finer grid in sampling composition space,

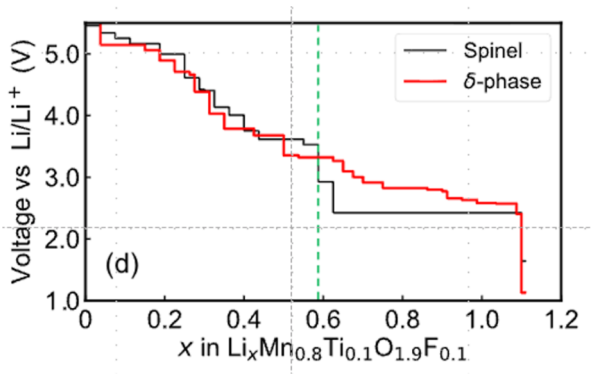


**Figure 4.** Voltage profiles for the spinel and $\delta$ phases of $Li_xMn_{0.8}Ti_{0.1}O_{1.9}F_{0.1}$ predicted by a custom CHGNet MLIP. The MLIP allowed sampling at many more values of $x$, compared to what is feasible with DFT, highlighting the large voltage drop in the spinel phase and solid solution behavior in the $\delta$ phase. Image adapted with permission from Zhong et al.[46] Copyright 2025 American Physical Society.

highlighting more features in voltage curves, which are critical diagnostic tools during battery operation. FPs can also predict voltage curves out-of-the-box for screening cathode materials before validating with DFT.

**2.1.4. Surface Stability.** Surface properties play an important role in determining the local structure of interfaces. Many chemo-mechanical and thermodynamic models consider surface energy as an important parameter, particularly for metal anode batteries.[110] The prediction of surface energies and reconstruction using DFT has been a powerful tool in studying battery materials and developing important descriptors of battery performance and has since been studied using MLIPs.[111] MLIPs open the door to study the thermodynamics and dynamics of more complex surfaces such as nanoparticles, dendrite-like nanopillars, and other morphologies.[112] Other important surface properties such as adsorption energies and surface diffusion barriers can now be applied on complex surfaces. DFT has been limited to smooth, low Miller index surfaces,[113] but real material surfaces such as electroplated materials can have complicated morphology. The large design space of facets and terminations is also challenging to explore with DFT but will be traversed quickly with foundation potentials as the appropriate data sets are developed.

**2.1.5. Mechanical Properties.** Elastic moduli such as shear and Young's modulus are parameters in chemo-mechanics models that determine stability vs dendrite formation in commonly used models such as those of Newman and Monroe and its derivatives.[114,115] Predicting the elastic moduli of bulk materials is well established and is commonly done with DFT. These estimates of elastic moduli are usually performed at 0 K and are typically within 15% of experimental values.[116] Accounting for temperature is often neglected due to the significant cost of the simulations for relatively small improvement in accuracy. However, for soft materials such as lithium and sodium and perhaps soft solid electrolytes, shear moduli can change significantly with temperature and morphology, as shown by experiments[117] and MLIP simulations.[111] In fact, sodium is expected to be in the plastic regime and exhibit creep at typical stack pressures.[110] MLIPs can therefore allow for more realistic predictions of mechanical properties and for more complicated geometries such as polycrystals with nanometer-sized grains.


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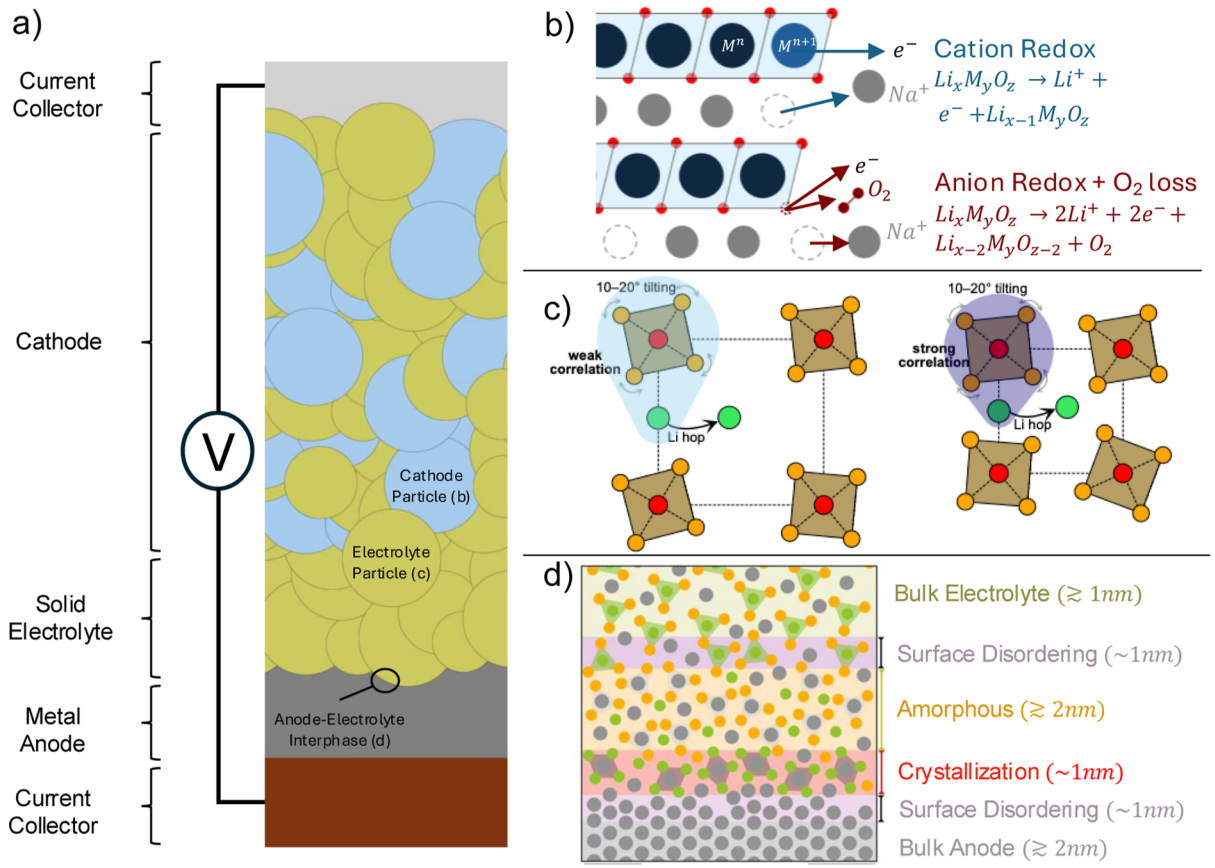


**Figure 5.** (a) Schematic of a typical all-solid-state battery and various interfaces. Depiction of (b) redox processes which consist of coupled ion motion and charge transfer locally in materials, (c) ionic motion, which can be highly correlated in superionic conductors with a variety of conduction mechanisms at play. Reproduced or adapted from.[14] Available under a CC-BY 4.0 license. Copyright 2025 Jun et al. (d) An interphase which can have complicated and dynamic local structure with amorphous, crystalline and disordered regions at various length scales.

Plasticity is a nonequilibrium property occurring over large length scales for which material history plays a significant role. It was therefore less commonly studied in atomistic models due to limited availability of interatomic potentials before MLIPs. Methods for analyzing the atomic origin of plasticity, such as stacking faults, dislocation motion and other defects, are well established and now commonly use MLIPs.[118,119] The atomic origin of plasticity, in terms of defects, bond-breaking, and atomic reconfiguration, is important in batteries,[120] particularly at heterogeneous interfaces. Contact loss and crack formation are indeed some of the major challenges in the development of all solid-state batteries with sufficient capacity retention. The application of external megapascal pressure (stack) has been demonstrated to mitigate contact loss in solid−solid interfaces, thus improving ion conduction and retention capacity.[121] However, the requirement for external pressure often leads to reduced energy density at the battery pack level.[121] An atomic level understanding and engineering can play a critical role in mitigating contact loss by guiding the design of chemically and mechanically stable interfaces.[8]

**2.1.6. Thermal Properties.** Thermal properties such as heat capacity and thermal conductivity are important for the performance and thermal management of batteries.[122,123] Heat capacity predictions are commonly used to predict phase transitions in battery materials and beyond, even using MLIPs.[88] Thermal conductivity in active materials tends to be limiting as they have higher thermal resistances compared to current collectors and other components that are typically metallic. If the heat generated during charging is not dissipated sufficiently, this can lead to degradation, and therefore designing materials with desirable thermal transport properties is of interest.[124] There also exist contradictory observations on the correlations between thermal and ion transport in solid electrolytes for which open scientific questions remain on the atomic scale.[125,126] MLIPs have been used successfully to predict thermal conductivity for solid halide electrolytes,[127] alloys,[128] phase change materials,[129] layered heterostructures[130] and molten salts.[131] Many of the methods used to predict thermal conductivities, such as Green−Kubo methods,[132,133] lattice dynamics,[134] and nonequilibrium molecular dynamics,[135] benefit greatly from increased system size and more sampling. MLIPs therefore hold advantages for heat transport predictions over other approaches. Ab initio methods based on perturbation theory can fail at finite temperatures[136] and empirical potentials fail in systems with high anharmonicity (e.g., PbTe[137]). We therefore see MLIPs as a viable approach to studying thermal properties in battery materials as more accurate models become available.

## 2.2. Interfaces

Interfaces play a major role in batteries but also tend to be the most difficult to study experimentally and theoretically. This is because the atomic structure of interfaces is often unknown, difficult to characterize experimentally and requires large length scales to model. Furthermore, the variety of interfaces shown in Figure 5 have different behaviors depending on the materials.


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MLIPs can play a major role in improving our understanding at the atomic scale, particularly for ion transport and reactions discussed in this section.

**2.2.1. Electrochemical Stability Windows.** The electrochemical stability window, which defines the voltage range over which materials at an interface remain stable against reduction or oxidation, directly affects the Coulombic efficiency and capacity fade. The thermodynamic limit to electrochemical window (excluding any kinetic effects) can be estimated using grand potential phase diagrams.[138] Although DFT has been the standard approach for mapping these windows,[10,139] MLIPs, particularly FPs, allow high-throughput screening in unexplored chemical spaces as well as accounting for the effect of temperature and dilute dopant concentrations.

A more complete understanding of stability, interfaces, and reactions needs to take into account transition states and structural changes associated with reactions. Many of these changes may not correspond to the lowest energy configurations used in DFT-based studies. MLIPs may enable accounting for these effects through carefully designed simulations that predict reaction dynamics[77] and/or kinetic barriers, e.g. using nudged elastic band approaches[102] to reactions at interfaces, going beyond purely thermodynamic predictions. This allows for more realistic and insightful modeling of degradation mechanisms, such as those occurring in sulfide or halide electrolytes during cycling.[90,140]

**2.2.2. Interfacial Transport.** Interfacial transport, which includes both grain boundary diffusion within polycrystalline solids and ion conduction across heterogeneous interfaces, plays a pivotal role in the overall performance of solid-state batteries. Even when the bulk exhibits high ionic conductivity, poor interfacial contact or sluggish transport across interfaces can become the rate-limiting step.[141] The interfaces between the solid electrolyte and the electrodes, or between grains in polycrystalline materials, often introduce resistance due to structural mismatch, electro/chemical instability, or accumulation of defects.[142]

Grain boundaries are a prototypical example of internal interfaces whose impact on transport is highly system-dependent. For example, increasing grain boundary density can enhance the ionic conductivity in $Li_3PS_4$ but reduce it in $Li_3OCl$.[143] At heterogeneous interfaces, such as electrolyte−cathode contacts, mismatches in mechanical, chemical, and electronic properties can create interphases that alter or block ion migration pathways[142] as shown in Figure 5d.

Quantitative modeling of interfaces demands large-scale simulations that capture relevant physics. Although regions affected by grain boundaries typically extend two to five nanometers, realistic modeling of coupled bulk−interface behavior, perhaps including space charge equilibration, often requires supercells spanning tens of nanometers in at least one dimension.[144] Currently, achieving such length scales is feasible only with classical potentials or MLIP-based simulations.[145] However, accurately capturing the complexity of reactions at the interface is challenging for classical potentials. MLIPs have enabled chemically accurate simulations of extended interfacial systems that were previously beyond reach. For example, Ou and Grabowski modeled ion transport in grain boundaries of $Li_6PS_5Cl$ with more than 16,000 atoms and trajectory lengths exceeding five nanoseconds.[146] Wang et al. simulated heterogeneous interfaces between Li metal and binary lithium compounds to explain the effect of the interface on Li transport ($Li_3P$, $Li_2S$ and LiCl).[15] Li et al. simulated ionic diffusion and observed the formation of an amorphous lithium metal-solid electrolyte interphase[147] as shown in Figure 5d. These simulations allow a mechanistic understanding of interfacial transport.

Accurate modeling of interfacial phenomena is challenging and there still remain a lot of challenges for MLIPs to address. A notable limitation of many existing MLIPs is their inability to dynamically assign charge. For example, Wang et al. demonstrated that moment tensor potentials could capture large-scale Li−metal/$Li_6PS_5Cl$ decomposition interfaces, but struggled to properly describe Li charge transfer across the interface, limiting their predictive accuracy for interfacial transport.[15] Recent advances in charge-aware MLIPs, such as 4G-HDNNP,[148,149] AIMNet2,[150] QET,[95] and others have significantly improved the treatment of atomic charge by incorporating environment-dependent charge equilibration, long-range electrostatics, and transformer-based architectures. Although these models represent substantial progress, fully capturing coupled ionic and electronic behavior at interfaces remains an open theoretical and computational challenge. This includes phenomena such as space charge layers.[144,151]

**2.2.3. Interfacial Reactions with Molecular Dynamics.** Synthesis conditions and solid-state reactions strongly influence the conductivity, stability, and degradation behavior of both electrodes and electrolytes.[152] Accurately capturing reactivity with classical interatomic potentials has long been a major challenge.[24] Machine-learned interatomic potentials (MLIPs) offer a promising route for studying reactions in molecular dynamics simulations. However, even the most computationally efficient potentials are generally unable to access inherently slow reaction processes.

For example, although molecular dynamics can simulate melt quenching, the quenching rates are often up to 14 orders of magnitude faster than in experiments ($\sim 10^{14}$ K/min in MD compared to ~1 K/min experimentally).[153] Similarly, direct simulation of processes such as ball milling remains difficult, despite some successful demonstrations in alloy systems.[154]

Nevertheless, MLIPs are well suited for processes in which kinetic barriers can be overcome at moderate temperatures, including exothermic reactions, diffusion, morphological evolution, grain boundary motion, and certain defect dynamics. For kinetically hindered phenomena, combining MLIPs with enhanced sampling techniques may provide a powerful approach.[155,156] At the same time, phenomena occurring over very large spatial scales, such as realistic grain sizes in some materials (>10 $\mu$m), are likely to remain beyond the reach of fully atomistic simulations.[157,158]

Currently, MLIP-MD simulations are used to investigate SEI growth, morphology, and transport properties, as well as the effects of interfacial structure and heterogeneity.[16,159,160] The reliability of MLIPs for these tasks depends on training with data that accurately represent the disordered and nonequilibrium environments of the SEI, rather than on potentials biased toward equilibrium crystalline phases. Most FPs are not specifically trained for these applications, especially due to the lack of large data sets with interfaces. Ongoing challenges include benchmarking MLIPs for SEI kinetics and structure across different chemistries, and limitations in modeling explicit charge transfer. Continued development and careful validation of MLIPs and data sets, particularly for composite, amorphous, and high-temperature systems, are crucial for predictive modeling of SEI dynamics.


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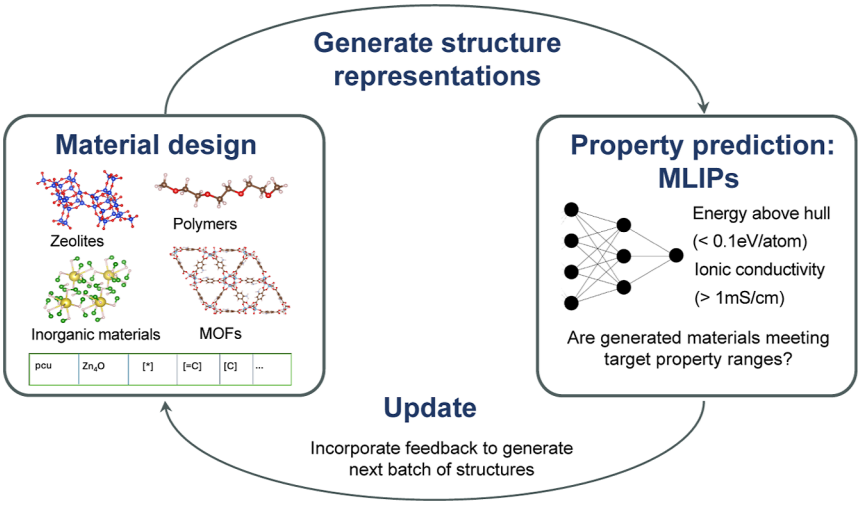


**Figure 6.** A schematic illustrating the role of MLIPs in accelerating inverse design approaches for material discovery. The "Material design" block can be trained to generate "structure representations" that are passed on to the "Property prediction" block. The MLIP-driven property prediction can then be used to "Update" and thereby "condition" the structure generation process toward materials that meet desired target ranges, for example, metal–organic frameworks (MOFs) having Na-ionic conductivities >1 mS/cm at room temperature.

**2.2.4. Redox Processes and Charge Transfer.** Redox processes or processes involving charge transfer between ionic species are at the heart of electrochemistry and involve a complex interplay between ionic and electronic transport, externally applied potentials, and the accompanying structural transformations. A better understanding of how redox processes affect degradation in cathode materials[161] is necessary for the engineering of longer-lasting batteries.

Several methods exist to describe the distribution of charge in a material including oxidation states, formal charges and partial charges. In *ab initio* methods, concrete charge accounting can only be done for the total electron charge density or more consistently using orbital basis sets rather than the most commonly used plane-wave basis sets in DFT.[20] However, there are a number of methods that are used to assign charges to ions based on predicted charge densities from DFT[162] or orbital populations.[163] These methods predict different values and are generally only used to compare changes in charge distribution. Several emerging MLIP architectures take into account these partial charges or correlated descriptors such as magnetic moments[34] or latent charges.[164] "Charge-aware" MLIPs can therefore be useful in tracking changes in the oxidation state based on the charge assignment and/or the equilibration scheme used in the training process. However, the assignment of charge using these methods may not be theoretically consistent with the underlying PES model and must be considered carefully.

**2.2.5. Phase Changes.** In some cases, electron transfer is accompanied by a corresponding structural change or the formation of gaseous compounds.[165] Examples include the proposed mechanisms of anion redox in lithium-excess cathodes, including processes like coordination changes and formation of oxygen species such as oxygen gas. The oxygen released induces local phase transformations, which leads to local stress and, subsequently, the initiation and propagation of cracks in battery materials and other degradation mechanisms.[166] MLIPs may be capable of capturing these structural changes, potentially even without explicitly modeling charge transfer, provided that the training data adequately samples the relevant regions of the potential energy surface (PES). However, the associated structural transformations typically occur on time scales far longer than those accessible to conventional molecular dynamics simulations. As a result, only processes with relatively small kinetic barriers are likely to be observed directly in MLIP-based MD simulations, while slower processes would require enhanced sampling techniques as discussed in Section 2.1.2 or combining with mesoscale models.[76,77] The equilibrium thermodynamics of these transformations have already been investigated using DFT.[161] Extending this capability to dynamic simulations would represent a major advance in the mechanistic insights that MLIPs can provide.

**2.2.6. Reactions under an External Potential.** The driving force for electrochemical reactions is an externally applied electrode potential, such as during charging. Unfortunately, accounting for an external potential remains a challenge for existing MLIP architectures and even *ab initio* methods. Implicit solvent models can be coupled with DFT to account for fixed electrode potentials, but this significantly increases computational cost and is more suitable for liquid electrolytes and molecules.[167] Robustly accounting for the external potential in redox processes remains a challenge at both the *ab initio* level and, therefore, for MLIPs. However, progress is being made; for example, Chen et al.[168] have demonstrated an MLIP trained on implicit solvent models that accounts for electrode potential and charge equilibration. Zhong et al.[169] have also argued that the electrical response can be inferred from MLIPs without training on charges or polarization or Born effective charges.

### 2.3. Materials Discovery and High-Throughput Workflows

**2.3.1. (Inverse) Design of Materials for Batteries.** A major goal of computational studies is to use physical and chemical insights about systems and processes to design and discover promising materials from a high-dimensional, near-infinite chemical space.[170] In this context, high-throughput screening studies have led to many technological breakthroughs in fields such as drug discovery,[171,172] carbon capture,[173,174] catalysis,[175,176] and others. Moreover, reverse engineering or the inverse design of materials based on specific desirable properties has opened new directions to accelerate this search for materials for a particular application.[177,178] Inverse design approaches help guide the material generation process toward the desired properties.[179,180] Generative models such as MatterGen[181] can


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possibly find new materials, but have yet to make significant advances for battery materials.

We envisage that these methodologies can prove beneficial in the discovery of materials for battery applications as well. For example, Bhowmik et al.[182] provided a possible blueprint for the inverse design of battery interfaces and interphases with desirable Solid Electrolyte Interphase (SEI) characteristics. Park et al.[183] showed that a surrogate model that predicts the property(ies) of interest in real-time can guide the metal−organic framework (MOF) generation process toward the desired direction. We extend this concept to a more generalized workflow that proposes the use of MLIPs as a property predictor for diverse classes of material, some inaccessible to DFT (Figure 6).

Generative modeling approaches (variational autoencoders,[178] diffusion models,[184] large language models[185] etc.) often require a surrogate model component to predict the property(ies) of interest in the fly. MLIP's featurization of the material design space can play a very significant role in computing target properties with reasonable accuracy for such diverse chemical spaces.[186] For example, the recently released UMA foundation potential has been used as a predictive model to discover high-performance carbon capture materials.[187,188] Fine-tuned MLIPs can then be used to more accurately predict the other properties discussed in previous sections. Rapid prediction of target properties in real-time can accelerate discovery workflows and condition them toward the high-performing regions of the material design space.[180,189]

Materials discovery is particularly valuable in material classes for which there are few known candidates and traditional enumeration approaches fail. In recent years, nanoconfined electrolytes have raised interest in the battery community due to their potential to provide electrolyte stability, optimized ion transfer, and suppressed side reactions, among other desirable properties.[156,190,191] Nanoporous materials such as zeolites, metal−organic frameworks (MOFs), covalent-organic frameworks (COFs), and carbon nanotubes provide such confinement effects through their porous networks and tunable chemistry, thus populating a vast chemical space.[174,192,193] These materials can be challenging for traditional DFT methods due to the large unit cells with several hundreds of atoms that are necessary in some cases. High-throughput screening and inverse design approaches have proven to be highly beneficial in navigating the space of nanoporous materials for diverse applications.[183,194,195] We envision such developments happening for battery applications as well, especially using FPs spanning both inorganic and organic materials. For instance, FPs such as MACE-MP-0,[35] MACE-MATPES-r2SCAN-0,[196] UMA-OMAT[36] have already been used to accurately describe complex chemical interactions of $CO_2$ with MOFs,[197] and $Li^+/Na^+$ with water-solvated zeolites.[155,156]

**2.3.2. High-throughput Screening and Autonomous Workflows.** A key practical advantage of MLIPs in materials discovery is their ability to serve as fast, accurate surrogates in high-throughput screening pipelines and autonomous workflows. Whereas DFT-based screening is limited to tens or hundreds of candidates by computational cost, MLIP-based workflows can evaluate thousands to millions of structures in the time required for a single DFT relaxation.[198] FPs are particularly well-suited for rapid first-pass screening across broad compositional spaces—for example, identifying candidate solid electrolytes in multicomponent halide or sulfide chemistries—before more expensive DFT or experimental validation is applied to the most promising candidates. Active learning workflows can then follow to finetune FPs used to predict properties of selected candidates at higher fidelity.

Such rapid feedback in guiding experiments will be a key component for autonomous laboratory platforms.[199] Autonomous laboratories represent the natural next step, coupling MLIP-driven simulation with robotic synthesis and characterization to close the loop between prediction and experiment. A persistent challenge, however, is ensuring that the structures and conditions sampled in silico are representative of those encountered during synthesis and cycling, and that MLIP uncertainty estimates are well-calibrated enough to guide these autonomous workflows reliably. Further, analysis techniques that incorporate multimodal data from experiments and simulation to reveal insights within one framework will be necessary.[200]

## 3. CONCLUSIONS

MLIPs are now a standard tool for atomistic simulation and have complementary advantages over empirical potentials and *ab initio* methods. MLIPs achieve the best trade-off for accuracy and computational cost to model the potential energy surface for large systems with $>10^3$ atoms and at nanosecond time scales. There exist many open questions, which we believe can be answered using existing MLIP capabilities, and we argue that developments in MLIP architectures will open up many more possibilities.

In materials discovery workflows, foundation potentials will allow for more rapid and reliable screening and downselection as their accuracy improves. They will be particularly advantageous in lesser explored material classes, such as materials for post-lithium batteries, nanoconfined structures, and composite materials, which are expensive for *ab initio* methods.

In property prediction, MLIPs can offer predictions that are very close to DFT predictions at orders of magnitude less cost and using basic software and hardware requirements. This should drive a paradigm shift in expectations of computational predictions toward more informative simulations accounting for more physics. For example, finite temperature phase diagrams, in addition to 0 K DFT phase diagrams, improve reliability. Directly simulating superionic conductors at room temperature with custom/fine-tuned MLIPs can be more reliable than the Arrhenius extrapolation from AIMD.

In any case, it would almost always be advantageous to first perform simulations with an appropriate FP before performing *ab initio* simulations or developing a custom MLIP. Such rapid simulation abilities can be quickly compared with those in experiment before further validation. The ability to obtain quick feedback from simulations with little lead time is a key enabler of increasingly popular autonomous laboratories.[199] However, at this point, convincing scientific arguments should still rely on *ab initio* or validated custom/fine-tuned MLIP predictions.

Coupling MLIPs with molecular dynamics, Monte Carlo, and enhanced sampling methods will allow direct observation of structural evolution at scales much closer to those of high-resolution experimental methods. This would be an exciting opportunity for the connection of experimental and computational insights. Importantly, curating appropriate computational and experimental data sets for battery applications will allow robust validation and benchmarking as new MLIPs and analysis methods that incorporate MLIPs are developed. These capabilities could enable deep insights into the complex

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structural and electronic phenomena that occur at battery interphases.

We emphasize that where *ab initio* methods are feasible, MLIP predictions should always be validated against them. Beyond accuracy, reliable deployment of MLIPs in battery research requires robust uncertainty quantification: committee models, conformal prediction, and Bayesian approaches can flag out-of-distribution predictions and guide active learning campaigns to extend model coverage efficiently. The computational efficiency, accuracy, and generalizability of FPs will continue to improve, and uncertainty quantification will be an essential component of their trustworthy use.

The ongoing development of charge-aware MLIPs and MLIPs capturing long-range interactions is a key development that will lead to a more insightful understanding of reactions and redox processes at the heart of electrochemical systems. Further developments in accounting for external influences and internal degrees of freedom, such as electric fields, electrode potentials, or spin, will be an important aspect of future generations of MLIPs. Foreseeable all-round improvements in accuracy, speed, usability, and robustness will further increase the impact of MLIPs, which is already exciting at this point.

Just as important, if not more important, now is the development of high-quality and high-fidelity data based on lessons learned in the development of existing data sets. We emphasize the importance of developing open-source data sets at the appropriate level of theory to increase fidelity, such as using better functionals rather than sticking to functionals that were traditionally chosen to make DFT more feasible for individual researchers. Furthermore, stricter and more consistent calculation settings will reduce the likelihood of numerical issues that deteriorate the quality of MLIPs.[64,65] We believe that this investment in generating better data sets will benefit the community and lead to more reliable FPs.

We have presented numerous arguments for the adoption of MLIPs in the fundamental research of battery materials. Therefore, it is pertinent for researchers to consider how MLIPs can be applied to improve battery technology, one of the most pressing technological challenges today.

## ■ ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.chemmater.6c01051.

Justification of the characteristic length and time scales in Figure 1a, computational-cost methodology for Figure 1b, and details of the literature-derived performance benchmarks (PDF)

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## Notes

The authors declare no competing financial interest.

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