

Machine Learning-Assisted Property Prediction of Solid-State Electrolyte

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Machine learning (ML) exhibits substantial potential for predicting the properties of solid-state electrolytes (SSEs). By integrating experimental or/and simulation data within ML frameworks, the discovery and development of advanced SSEs can be accelerated, ultimately facilitating their application in high-end energy storage systems. This review commences with an introduction to the background of SSEs, including their explicit definition, comprehensive classification, intrinsic physical/chemical properties, underlying mechanisms governing their conductivity, challenges, and future developments. An in-depth explanation of the ML methodology is also elucidated. Subsequently, the key factors that influence the performance of SSEs are summarized, including thermal expansion, modulus, diffusivity, ionic conductivity, reaction energy, migration barrier, band gap, and activation energy. Finally, it is offered perspectives on the design prerequisites for upcoming generations of SSEs, focusing on real-time property prediction, multi-property optimization, multiscale modeling, transfer learning, automation and high-throughput experimentation, and synergistic optimization of full battery, all of which are crucial for accelerating the progress in SSEs. This review aims to guide the design and optimization of novel SSE materials for the practical realization of efficient and reliable SSEs in energy storage technologies.

1. Introduction

Recently, there has been a surge in demand for energy storage systems, particularly in the field of advanced batteries that prioritize high energy density and safety.^[1–4] The burgeoning need for such systems is a direct result of the fast-paced advancements in portable electronic devices, electric vehicles, and renewable energy technologies. Routine liquid electrolytes used in commercial lithium-ion batteries have limitations such as flammability, leakage, and limited stability, which hinder their widespread application in these emerging technologies.^[5–7] SSEs are capable of conducting ions while maintaining a solid state, which have garnered wide attention in the field of advanced solid-state batteries due to their potential to overcome the limitations associated with liquid electrolytes. Importantly, SSEs offer several advantages

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over routine liquid electrolytes, including enhanced safety, a wider operating temperature range, and better compatibility with various electrode materials.^[8–10] These properties render them suitable for application in high-performance rechargeable ion batteries and alkali metal batteries, such as Li/Na-ion batteries (LIBs and SIBs), lithium-sulfur batteries (LSBs), and Li/Na metal batteries.^[11–13] Consequently, SSEs have emerged as a promising alternative.

Conventional approaches to material discovery and refinement are either time-consuming or/and costly, frequently hinging on trial-and-error techniques.^[14–17] This is where ML techniques come into play. Falling under the umbrella of artificial intelligence, ML grants computers the ability to extract patterns from data and autonomously make predictions or decisions, all without explicit programming.^[18–22] In the context of SSE research, ML can significantly accelerate the discovery and optimization process by analyzing large datasets and identifying patterns that are difficult for human researchers to detect.^[23–29] In recent years, the utilization of ML in SSE research has garnered considerable attention. The development of ML in SSEs has progressed from initial exploration to advanced applications, materials screening and design, structure optimization, performance prediction, interface engineering, and energy management.^[30] These advancements have significantly accelerated research and innovation in the field of SSEs. Utilizing ML model allows for the prediction of characteristics in novel SSE materials, including conductivity, stability, and compatibility with electrode materials, based on their chemical composition and structure.^[31–34] This approach has the potential to revolutionize the field of SSEs research by enabling rapid and efficient material discovery and optimization. In predicting SSE properties, various ML models are employed based on their advantages and data characteristics. Neural Networks are utilized for their prowess in modeling complex, non-linear relationships, especially in large datasets. Decision Trees and Random Forests (RF) are chosen for their interpretability and capability with both numerical and categorical data, aiding in identifying crucial factors affecting SSE properties. Support Vector Machines are preferred for high-dimensional spaces, and ideal for feature-rich datasets in SSE prediction. Linear and logistic regression models offer simplicity and effectiveness in deciphering variable relationships. Ensemble Methods, like Gradient Boosting (GB) or AdaBoost, enhance prediction accuracy by amalgamating multiple models into a more robust predictor. The selection of these models is tailored to the specific requirements of SSE property prediction, including dataset size, complexity of the feature space, and interpretability needs. For instance, Ahmad et al. conducted a study on more than 12 000 inorganic materials to identify those that can prevent dendrite growth upon interaction with a Li metal anode.^[35] ML algorithms

were employed to predict the mechanical attributes of new SSEs, allowing for a large amount of data to be gathered without time-consuming computations. Elastic constants were trained using Kernel Ridge Regression and Gradient Boosting Regressors, considering the volume of training data and the tolerance for noise. The findings show that an increase in sublattice electronegativity and volume per atom lead to a decrease in the stiffness of the material. By conducting tests and employing cross-validation, they pinpointed over 20 mechanically anisotropic interfaces between Li metal and four SSEs, showing promise in dendrite prevention. These options have both dendritic suppression and good ionic conductivity in SSEs.

Although their several promising benefits, SSEs currently still face some issues in conductivity, interface stability, mechanical performance, and screening processes. ML offers solutions by predicting conductivity, optimizing interface design, improving material properties, and accelerating material discovery, thereby addressing these issues and advancing the development of SSEs. Especially, first-principles calculations, Monte Carlo methods, and molecular dynamics simulations can contribute to ML by facilitating feature extraction, data generation, model optimization, and physical interpretation.^[36–38] These approaches expand the utilization of ML in the domains of chemistry and materials science, advancing molecular design and material discovery. D. Sendek et al. employed an ML-based predictive model to conduct a thorough exploration of material space for material selection, coupled with DFT to measure ionic conductivity.^[27] The researchers screened a vast array of materials, comprising over 12 000 candidates that had been experimentally synthesized and characterized, encompassing a wide range of structures and compositions. When contrasting the results of their ML-assisted investigation with a haphazard exploration of material space, they found that the former exhibited a 2.7-fold higher likelihood of pinpointing rapid Li ion conductors, leading to a striking 44-fold increase in Li ion conductivity. To investigate Li diffusivity within a multitude of ternary crystals as promising candidates for all-solid-state battery electrolytes, Hajibabaei et al. combined ab initio molecular dynamics (AIMD) with an on-the-fly ML approach for interatomic potential modeling.^[24] They suggested that the models crafted for these crystals could be smoothly incorporated to formulate more general and adaptable models, negating the necessity for additional training when simulating different materials. This hierarchical approach enables the representation of large-scale complexity through a combinatorial method. Additionally, an ML-driven surrogate model was designed to seek out the best garnet-type SSE candidates.^[39] Using the widely recognized $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZO) structure as a starting point, researchers made substitutions of 73 chemical elements on the La and Zr sites, yielding 5329 potential structures. To confirm the diffusion behavior of these materials classified as superionic, AIMD simulations were conducted. This thorough process led to the confirmation of ten new tetragonal-phase garnet SSEs that exhibited outstanding mechanical strength and great ionic conductivity. Despite the presence of reviews concerning ML algorithms and descriptors in the realm of SSEs, there remains a notable dearth of a comprehensive review elucidating the most recent ML advancements relevant to SSE properties.^[40–42] To this end, our review endeavors to bridge this gap by concentrating specifically on the application of ML to scrutinize the determinants of SSE

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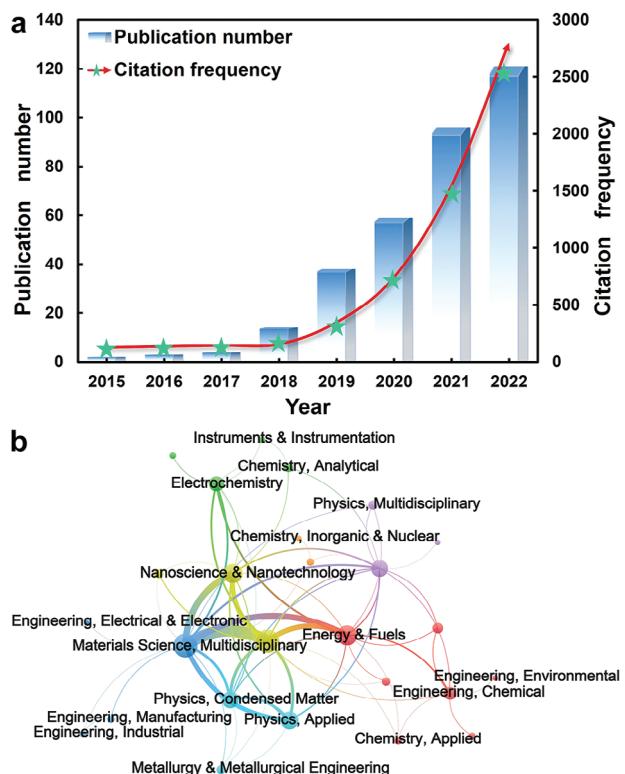


Figure 1. SSE advancements aided by ML. a) Distribution plots illustrating the number and citation frequency of relevant articles were obtained from the Web of Science database using the search terms “machine learning” and “solid-state electrolytes”. b) The corresponding distribution of different disciplines.

properties and provide a full picture of the current status and future directions in this fast-evolving field. This refined approach facilitates a comprehensive exploration of the challenges intrinsic to the development of superior SSEs.

In this review, research efforts will be directed toward the deployment of ML to predict various properties of SSEs, such as conductivity, mechanical strength, thermal stability, and interfacial stability. The SSE properties and ML procedure are presented first, and then an overview is provided on the application of ML in investigating the factors that influence SSE properties. Furthermore, the prospects and potential directions for the development of ML methods in SSE research are discussed. By synergistically harnessing the capabilities of SSEs and ML, we hope that this review will expedite the advancement of advanced energy storage systems and make substantial contributions toward achieving a sustainable and clean energy future.

2. Brief Overview of SSE and ML

By analyzing the citation and publication counts of articles, we can infer the advancement of SSEs aided by ML. As depicted in Figure 1, observing the trends in citation and publication counts over time provides valuable insights into the field’s growth and impact across various disciplines. High citations and increasing publication counts reflect a surge of interest and active research activity. Therefore, the synergistic integration of SSEs and ML

represents a pivotal advancement in the field of energy materials research. ML aids in navigating the vast and complex compositional space of SSEs, guiding researchers toward promising material candidates for further experimental validation. This dynamic interplay between SSEs and ML exemplifies the multidisciplinary nature of contemporary materials science, showcasing how ML techniques are revolutionizing the way we approach and solve complex challenges in energy storage and beyond. As research in this field continues to advance, the potential for transformative breakthroughs in sustainable energy technologies becomes increasingly promising.

2.1. The Background of SSE

SSEs play a pivotal role in a diverse range of electrochemical devices, finding extensive application in cutting-edge technologies like solid-state batteries and fuel cells.^[43] These materials serve as the critical interface between electrodes, facilitating the flow of ions in a solid state rather than through a liquid or gel electrolyte. Understanding the properties of SSEs is crucial for optimizing the performance and reliability of these devices. This section aims to provide a holistic overview of SSEs, encompassing their definition, classification, physical/chemical properties, underlying mechanisms dictating their ionic conductivity, challenges, and future development. This knowledge serves as a foundation for advancements in SSE technology, driving progress within the realm of electrochemical energy storage and conversion.

The distinct properties of SSEs have garnered substantial interest as potential alternatives to liquid electrolytes in batteries.^[44-46] (1) Ionic conductivity: SSEs demonstrate favorable ionic conductivity, facilitating the effective flow of ions within the material. This characteristic significantly impacts the overall effectiveness of the battery, as it determines the rate at which ions can travel between the electrodes, leading to improved power output.^[47] (2) Enhanced safety: SSEs offer improved safety compared to liquid electrolytes. Liquid electrolytes are prone to leakage and can be flammable, while SSEs are non-flammable and do not require the use of flammable solvents. This makes SSEs a safer choice, particularly for high-energy applications like electric vehicles.^[48] (3) Wide electrochemical stability window: SSEs have a wide electrochemical stability window, meaning they can withstand a wide range of voltage potentials without undergoing degradation. This attribute enables the utilization of high-voltage cathode materials, leading to a substantial boost in the battery’s energy density.^[49] (4) Good mechanical stability: SSEs exhibit good mechanical stability, enabling them to maintain their structural integrity even under mechanical stress. This feature plays a pivotal role in maintaining the battery’s durability and reliability over time, particularly in applications where the battery may experience physical impacts or vibrations.^[50] (5) Compatibility with multiple electrode materials: SSEs exhibit good compatibility with a broad spectrum of electrode materials, ranging from lithium metal and lithium-ion to innovative emerging options. This versatility allows for the development of batteries with different chemistries and performance characteristics.^[51] These properties contribute to higher battery efficiency and better overall performance. While SSEs offer notable advantages, they also present several challenges in practical application.^[52] These

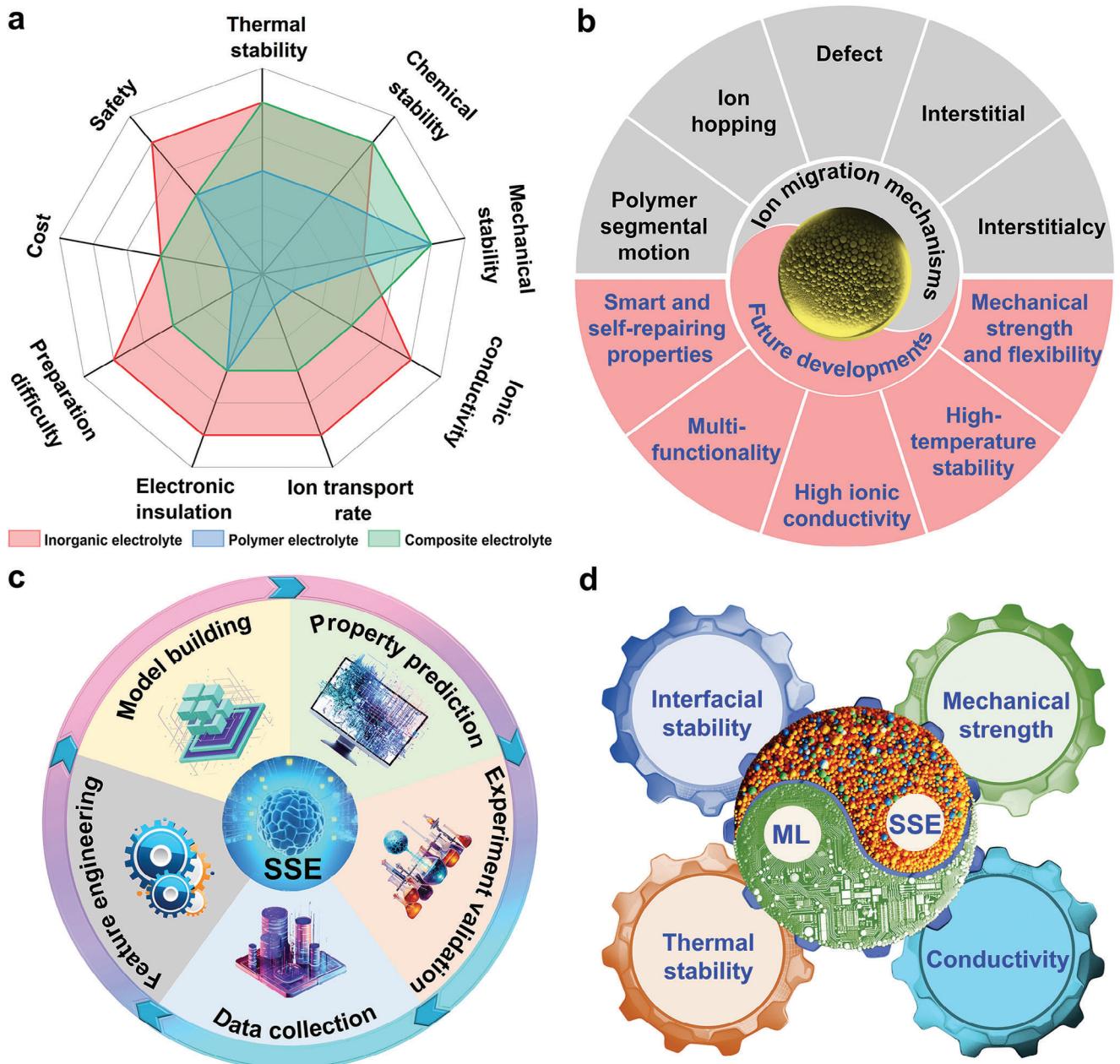


Figure 2. SSE Performance and ML prediction. a) Radar plots for the properties of varying SSEs. b) The ion migration mechanisms and future developments of SSEs. c) The process of using ML to predict the properties of SSE. d) ML-assisted prediction of SSE properties and design of SSE materials.

include the complex manufacturing process requiring precise techniques and materials, and high production costs for specific high-performance electrolytes, potentially impeding their viability in large-scale commercial applications. Furthermore, some SSE materials may undergo chemical changes over extended usage or under extreme operating conditions, impacting the stability and lifespan of the battery. Additionally, interface issues between SSEs and electrodes play a crucial role in determining the overall performance of the battery.

SSEs, functioning without reliance on a liquid or gel-like medium, fall into three primary categories: inorganic, polymer, and composite.^[53] The radar plots in Figure 2a serve as an illus-

tive tool for visually comparing the distinct characteristics of these SSE materials. Inorganic electrolytes, typically composed of inorganic compounds like oxides, sulfides, and phosphates, exhibit commendable ionic conductivity coupled with exceptional thermal stability.^[54-58] However, they are characterized by brittleness and limited flexibility.^[59] In contrast, polymer electrolytes are composed of a polymer matrix where ionic species are either dissolved or dispersed, offering excellent mechanical flexibility and the capacity for versatile shaping.^[60] Nevertheless, their ionic conductivity tends to be lower in comparison to inorganic electrolytes. Composite electrolytes, amalgamating inorganic and polymer components, strive to harness the strengths

of both categories, aiming for a synergy between high ionic conductivity and mechanical adaptability.^[61] Various factors can affect the ionic conductivity of SSEs, encompassing crystal structure, composition, and defects.^[62–64] Notably, the presence of vacancies or interstitial sites within the crystal lattice affords facilitation for ion migration, consequently resulting in heightened conductivity.^[65] Furthermore, the augmentation of ionic conductivity can be achieved through the deliberate introduction of dopant elements, which serve to introduce supplementary mobile charge carriers.^[66] Ion migration plays a crucial role in SSEs, determining their ability to effectively conduct ions and thereby impacting the performance of electrochemical devices like batteries and fuel cells. A high rate of ion migration enhances the power density and efficiency of devices, while also influencing cycle life, fast charging/discharging capabilities, and applicability in various scenarios. Therefore, a deep understanding of ion migration mechanisms is pivotal in the research and development of SSEs. The ionic conductivity in SSEs manifests through distinct ion migration contingent upon the material category (Figure 2b). Within inorganic electrolytes, ion migration operates through three prevalent mechanisms: the defect mechanism, where ions leap to adjacent vacant sites; the interstitial mechanism, facilitating ion movement within interstitial positions; and the interstitialcy mechanism, involving the displacement of neighboring lattice ions by migrating interstitial ions to alternate interstitial positions.^[67–69] Additionally, inorganic electrolytes feature grain boundaries between crystalline grains, serving as conduits for ion transport. The inclusion of grain boundaries notably amplifies the ionic conductivity within inorganic electrolytes. Polymer electrolytes, on the other hand, facilitate ion transportation either along the polymer chains or through ion hopping between distinct polymer segments.^[70] In the case of the polymer segmental motion, the mobility of polymer segments facilitates ion migration, with factors like temperature and polymer chain length exerting notable influence.^[71] Composite electrolytes, characterized by an interface between inorganic and polymer phases, promote ion transport. This interaction between inorganic and polymer phases fosters the establishment of ion-conductive pathways.^[72] For the purposeful design and optimization of their performance in a wide range of electrochemical devices, it is essential to have a thorough understanding of the different ion migration mechanisms in SSEs. Ongoing research endeavors are dedicated to elevating the conductivity of SSEs as well as exploring novel materials endowed with enhanced properties.

The future development of SSEs holds significant potential for advancements in various electrochemical devices. As shown in Figure 2b, the following are potential directions for their future development: (1) High ionic conductivity: enhancing the ability of SSEs to efficiently conduct ions is paramount for improving the performance of electrochemical devices. (2) High-temperature stability: developing SSEs with exceptional thermal stability is crucial for enabling operation at elevated temperatures, leading to increased device efficiency and energy density. (3) Smart and self-repairing properties: researching and developing SSEs with self-repairing capabilities or intelligent responsiveness will enhance device stability and longevity. (4) Reducing manufacturing costs: researchers will strive to develop more cost-effective manufacturing processes and cost-efficient SSEs to make them more competitive in commercial applications. (5)

Multifunctionality: exploring the potential for SSEs to serve multiple functions, such as simultaneous ion and electron conduction, expands their versatility and application range. Considering these directions, the future of SSEs will continue to progress toward higher efficiency, stability, environmental sustainability, and multifunctionality, providing a broader range of possibilities for the application of electrochemical devices. The collaborative efforts of researchers and ML systems hold the key to unlocking the full potential of SSEs and propelling us toward a more sustainable and energy-efficient future. We firmly believe that with the aid of ML techniques, efficient SSEs can be developed.

2.2. The Process of ML for Predicting SSE Properties

By training ML models on existing experimental data, researchers can develop predictive models that can guide the design and synthesis of new SSEs with desired properties.^[73] Figure 2c illustrates the comprehensive process for leveraging ML techniques to predict SSE properties, showcasing a systematic approach that encompasses several crucial stages. The first step, data collection, involves gathering a diverse and extensive dataset encompassing various SSE compositions, crystal structures, synthesis methods, and relevant experimental observations. This dataset serves as the foundation upon which the ML model will be trained. For ML in SSEs, identifying key data sources like scientific databases and industry reports is essential. Ensuring data quality through accuracy, completeness, and consistency is crucial for effective ML models. The volume of data, while significant for training, should be manageable. Effective data preparation techniques, such as cleaning and normalization, are necessary for usability. Utilizing open-access databases and staying updated on data collection trends can further enhance research. Including case studies exemplifies the practical application and importance of these methods.

Following data collection, the next crucial stage is feature engineering, which involves extracting meaningful and informative features from the raw dataset. Advanced techniques such as dimensionality reduction, symmetry analysis, and electronic structure calculations are employed to identify the most relevant descriptors that capture the intrinsic characteristics of SSE materials. This process plays a pivotal role in enhancing the model's ability to discern patterns and make accurate predictions. The prediction of SSEs' properties via ML hinges on both physical and chemical features. Physically, aspects like ion migration pathways and lattice dynamics determine ionic conductivity, while mechanical and thermal properties gauge stability across temperatures. Chemically, factors such as composition, electronic structure, and bonding types are crucial. These combined features significantly enhance the accuracy of ML models in assessing SSEs' properties.

Subsequently, the model-building phase entails selecting and fine-tuning appropriate ML algorithms based on the specific characteristics of the dataset and the desired predictive task. Various algorithms offer distinct advantages based on the nature of the data and the complexity of the relationships.^[74–76] Linear Regression, known for its simplicity, is particularly effective in predicting quantitative responses where a linear relationship exists between input features and outputs, such as estimating

ionic conductivity from composition and temperature in SSEs. On the other hand, Gaussian Process Regression excels in predicting complex patterns, especially valuable in cases of small datasets. It stands out by providing uncertainty measurements along with predictions, making it suitable for predicting electrolyte properties in scenarios with sparse data or for interpolating between known data points. Deep Neural Networks (DNNs), with their ability to model complex, non-linear relationships, are particularly useful for large datasets containing numerous input features. In SSEs, DNNs can be instrumental in predicting properties by analyzing a vast array of structural and chemical data. Once the ML model is trained and validated on the training dataset, it proceeds to the property prediction stage. Here, the model applies the learned relationships between features and SSE properties to make predictions for previously unseen materials. The predictive results are then subject to rigorous scrutiny to ensure their reliability and accuracy.

In ML, model evaluation metrics are crucial tools for assessing the performance of predictive models. For instance, the R-Squared (R^2) metric indicates the proportion of variance in the data that the model accounts for, reflecting how well the model fits the data. The Root Mean Square Error (RMSE) measures the error in predicting quantitative data, with lower RMSE values indicating better model fit. In classification problems, precision and recall are commonly used metrics. Precision is the ratio of correctly predicted positive observations to the total predicted positives, while recall, or sensitivity, is the ratio of correctly predicted positives to the actual positives. Additionally, the confusion matrix provides a detailed evaluation of a classification model's performance, showing true positives, false positives, true negatives, and false negatives. The Area Under the ROC Curve (AUC-ROC) is important in binary classification, assessing the model's ability to distinguish between classes, where a higher AUC indicates better performance. Finally, the Mean Absolute Error (MAE) is the average difference between predicted and actual values, with a lower MAE signifying a better-performing model. Each of these metrics has its own focus, and choosing the appropriate one based on the specific requirements and context of the ML task is essential for evaluating and improving models. Experiment validation is an indispensable step in the process. This involves comparing the ML-predicted properties with experimental data for a subset of materials from the dataset. This validation serves as a critical check on the model's predictive capabilities and helps identify any potential biases or limitations. Finally, this iterative process of data collection, feature engineering, model building, property prediction, and experiment validation is repeated and refined as new data becomes available, enabling the ML model to adapt and improve over time. This continuous feedback loop ensures that the model remains robust and reliable in the face of evolving SSE materials and experimental techniques.

Within the vast domain of SSEs, a collaborative initiative is underway to harness the potential of ML for predictive modeling across a diverse range of SSE properties. This includes, but is not limited to, conductivity, mechanical strength, thermal stability, and interfacial stability, as illustrated in Figure 2d. The upcoming work delves into these aspects, exploring the potential of ML to provide nuanced insights into SSE behavior and performance. A primary focus of the ML predictions lies in conductivity, a pivotal factor in energy storage applications. This emphasis aims to

elucidate how SSEs can augment the efficiency and overall performance of batteries and related devices. Simultaneously, the consideration of mechanical strength in the predictive models contributes to the strategic design of robust SSEs, ensuring their resilience in practical applications. Beyond these mechanical attributes, the predictions also extend to thermal stability and interfacial stability, offering a holistic perspective on SSE suitability for diverse scenarios. These insights contribute to a comprehensive understanding of SSEs, informing their potential applications in a broad spectrum of real-world settings. To achieve these objectives, a synergistic multidisciplinary approach is adopted, drawing on expertise from materials science, data science, and computational modeling. This collaborative effort is instrumental in navigating the complexities inherent in SSE research, enriching the predictive models with a diverse range of perspectives.

3. ML Applications

In recent years, ML has emerged as a potent tool for investigating the key factors that influence the properties of SSEs. By leveraging ML algorithms and techniques, researchers have managed to extract valuable knowledge regarding how various factors relate to the properties of SSEs. As shown in Figure 3, some important factors such as thermal expansion, modulus, diffusivity, ionic conductivity, reaction energy, migration barrier, band gap, and activation energy are introduced. In this intricate matrix, optimizing one property often necessitates compromises in others. For example, while enhancing ionic conductivity can inadvertently increase ion diffusivity, potentially compromising stability and safety, improving mechanical properties like modulus might reduce material flexibility. Similarly, a lower migration barrier, aimed at improving conductivity, could decrease reaction energy, thereby affecting chemical stability. Achieving an optimal balance among these factors calls for a sophisticated approach that acknowledges the interconnectedness of these properties, aiming to develop SSEs that embody an ideal mix of efficiency, stability, and durability tailored to specific applications. By harnessing the potential of ML, it becomes possible to predict these parameters and consequently forecast certain properties of SSEs.

3.1. Activation Energy

The activation energy of SSEs refers to the energy barrier that ions need to overcome when moving within the material. It holds significant importance in shaping the ionic conductivity of the electrolyte.^[77] The activation energy of SSEs depends on several factors, including the crystal structure, chemical composition, and temperature. Generally, higher activation energies result in lower ionic conductivity, as ions find it more difficult to move through the material. Therefore, reducing the activation energy is a key strategy to enhance the performance of SSEs.^[78] One application of SSEs with low activation energy is in solid-state batteries. Through the reduction of activation energy, SSEs can be tailored for utilization in solid-state batteries, fuel cells, and a range of energy storage and conversion technologies. Utilizing SSEs characterized by low activation energy can boost the battery's ionic conductivity, thereby improving both power output and overall performance.^[79]

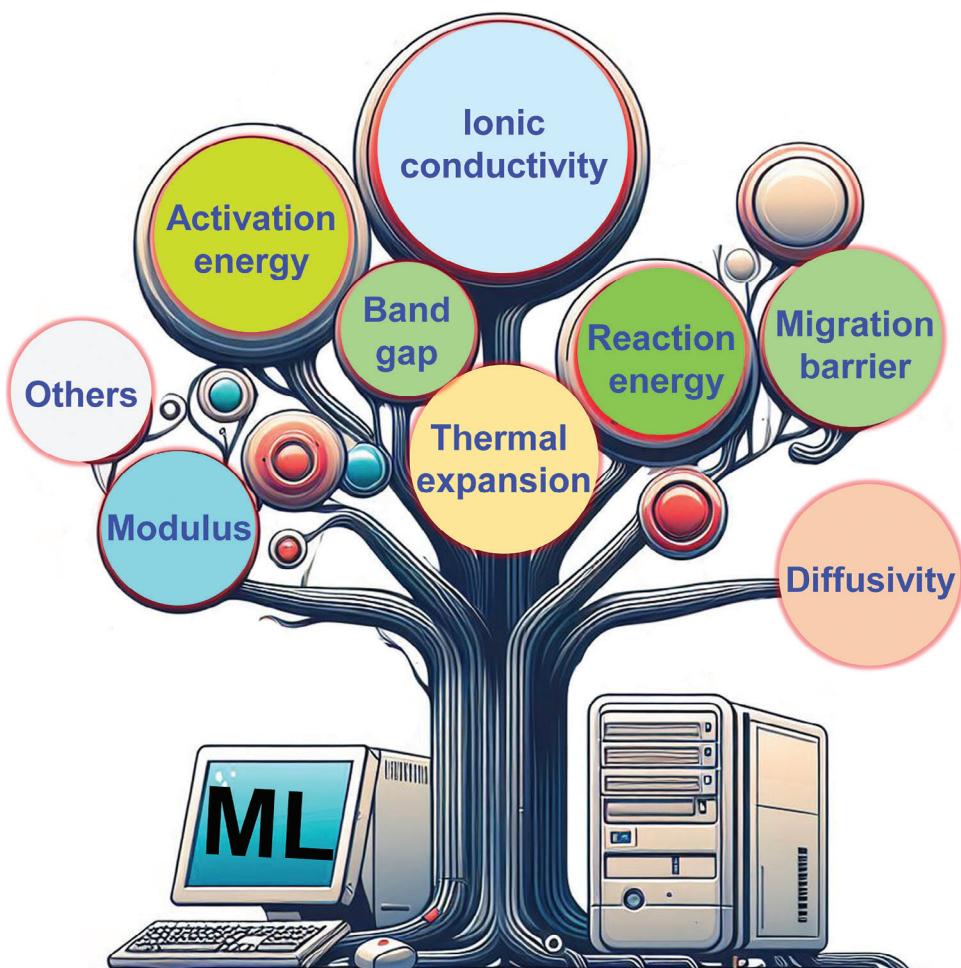


Figure 3. ML applications for studying the crucial factors that influence SSE properties.

Building upon earlier work, Zhao et al. developed a framework that utilizes a hierarchically encoded crystal structure-based (HECS) descriptor approach for ML prediction of activation energy (E_a) in SSEs.^[80] As shown in **Figure 4a**, this framework involved constructing an E_a prediction model, which was exemplified with cubic Li-argyrodites as a case study. The model displayed robust coefficients of R^2 and low RMSE values. In detail, it obtained an R^2 of 0.887 with an RMSE of 0.02 eV for the training dataset and an R^2 of 0.820 with an RMSE of 0.02 eV for the test dataset (Figure 4b). These results provide empirical support for the predictive efficacy of HECS descriptors. Additionally, variable importance in projection (VIP) scores provided insight into the integrated impact on both general and localized Li^+ conduction environments (Figure 4c). Anion size and the consequential structural changes attributed to disorder at the anion sites were particularly emphasized. The established model for predicting E_a offers strategic guidance for refining and designing new Li-argyrodites with lowered E_a . Achieving this involves expanding bottleneck dimensions, introducing site disorder, and fostering synchronized Li^+ conduction (Figure 4d). This extensive study offers crucial insights into the microscopic Li^+ conduction process, signifying substantial potential in propelling the systematic design of cutting-edge SSEs.

3.2. Band Gap

Defined as the energy difference between the valence band (the highest occupied electronic state) and the conduction band (the lowest unoccupied electronic state), the electronic band gap (E_g) encapsulates the foundational electronic features of a material.^[81] Within the array of factors influencing the electrochemical behavior of SSEs, the E_g stands out as a pivotal determinant of ion migration kinetics and the overall electrochemical performance.^[82,83] In SSEs, this parameter holds significant relevance as it directly impacts the availability of charge carriers for ion transport. A narrow E_g can facilitate facile electron excitation, promoting the movement of charge carriers and subsequently enhancing ionic conductivity. Conversely, a wider E_g restricts electron mobility, potentially impeding the ion diffusion process.^[84]

To uncover garnet-type SSEs destined for solid-state batteries, Wang and coworkers introduced an efficient target-oriented ML strategy (Figure 5a).^[85] Initially, they amassed 286 distinct garnet structures with computed E_g from the Materials Project database.^[86] Subsequently, a collection of 29 008 garnets was constructed by combining four elements derived from the known set of 286 garnets. Out of these, 7067 garnets were projected

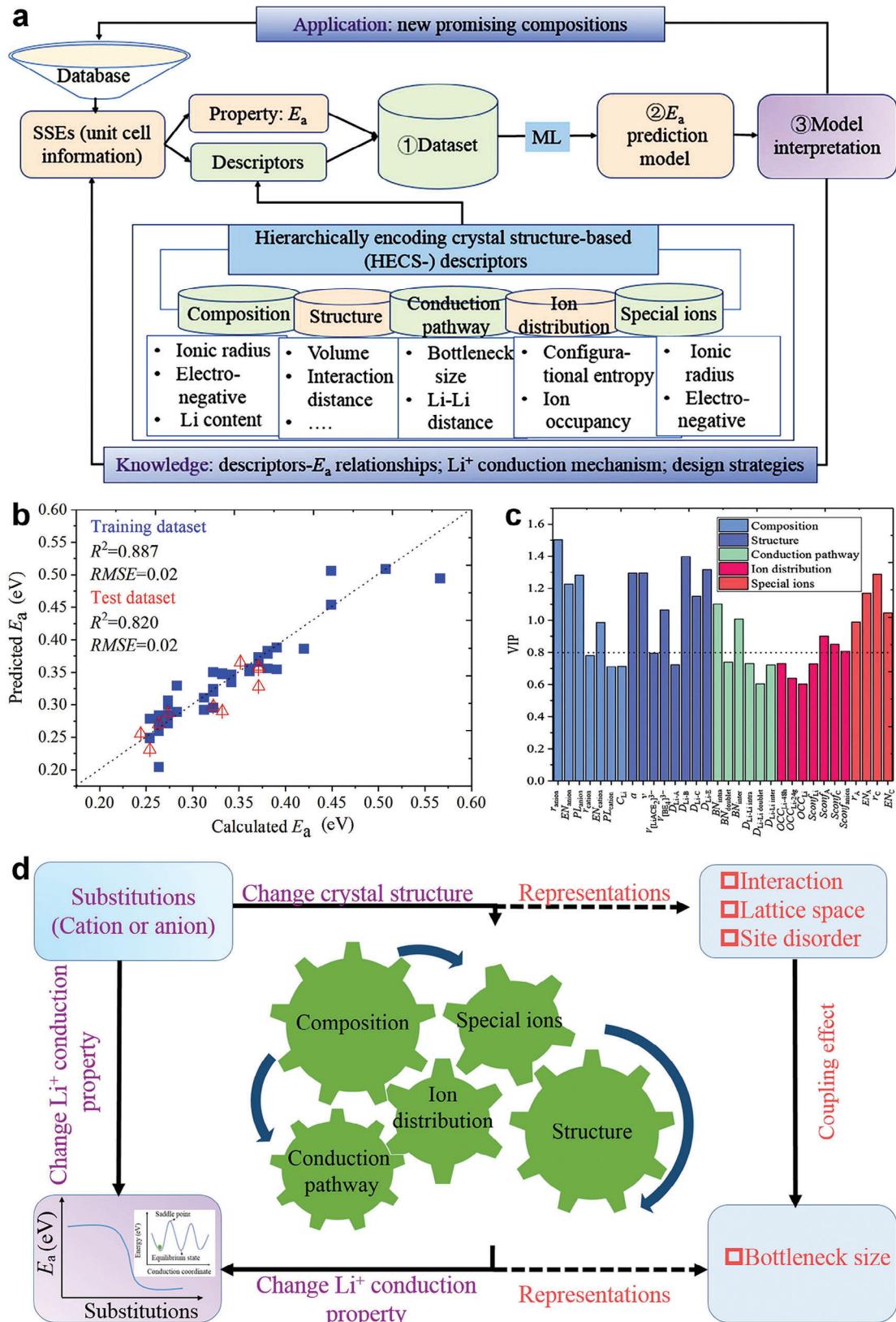


Figure 4. The activation energy predicted by ML. a) The prediction process of E_a with ML. b) The plot of calculated E_a versus predicted E_a . c) VIP scores of descriptors. d) Effects of HECS-descriptors combined on E_a and their interaction mechanism. Reproduced with permission.^[80] Copyright 2021, Elsevier B.V. and Science China Press.

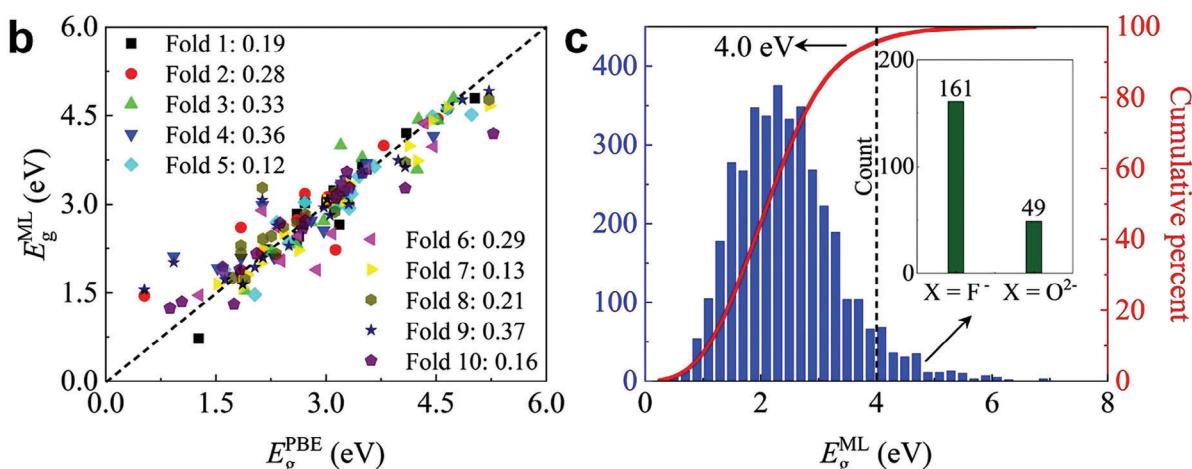
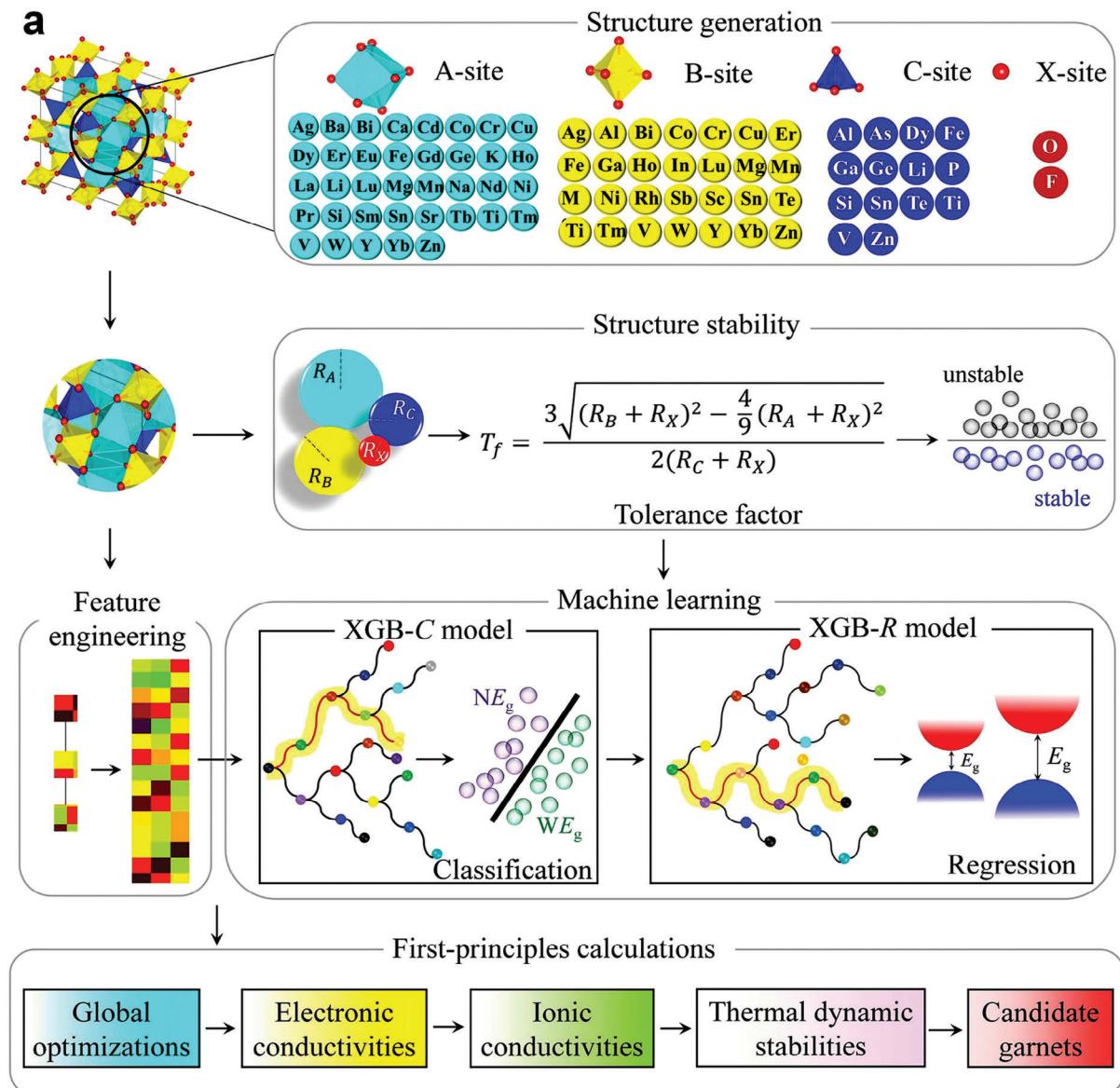


Figure 5. Utilizing ML for comprehensive design and identification of SSEs. a) The process of predicting garnet SSEs. b) E_g correlation plot with PBE and ML. c) Prediction of 3801 wider E_g garnets based on regression. Reproduced with permission.^[85] Copyright 2021, Elsevier.

to exhibit structural stability following tolerance factor filtration. Through the process of feature generation and extraction grounded on elemental characteristics, they developed two models, one for classification (XGB-C) and the other for regression (XGB-R), using the XGBoost algorithm on the dataset consisting of 286 garnets. Meanwhile, both metrics were utilized in tandem to gauge the precision of the XGB-R model. In Figure 5b, the testing results reveal the MAEs derived from tenfold cross-validations for the XGB-R model. The ML method forecasted E_g exhibited strong congruence with those derived from computations employing the Perdew–Burke–Ernzerhof (PBE) functional. Moreover, the XGB-R model demonstrated exceptional proficiency in predicting wider E_g , yielding an impressively low MAE of 0.25 eV. This resulted in a computational acceleration of ≈ 109 times when compared to ab initio calculations. In rapid succession, the XGB-R models were deployed to project characteristics for 3801 garnets. Among these, 210 novel garnets, distinguished by their diminished electronic conductivities, were singled out (Figure 5c). Subsequently, the dependable climbing image nudged elastic band technique was applied for ionic conductivity predictions,^[87] while long-range AIMD was employed for evaluating thermal dynamic stability at both room temperature and elevated temperatures. Ultimately, this endeavor yielded the discovery of 12 novel garnets boasting high ionic conductivity and commendable thermal stability at both room temperature and elevated temperatures. These garnets hold significant promise as garnet-type SSEs for solid-state lithium-ion electrolytes (Li-ion SSEs) batteries. The model's success in unveiling these 12 garnet-type SSEs from a pool of 29 008 structures led to an impressive computational reduction, shortening the garnet screening process by ≈ 95 years. This not only translates to substantial cost savings in experimental endeavors but also furnishes a distinctive perspective and methodology in the quest for advancing SSEs for SSBs, thereby propelling the progress of cutting-edge energy storage technologies.

3.3. Migration Barrier

The migration barrier refers to the energy barrier that ions must overcome during the transport within the crystalline structure of SSEs, owing to factors such as structural complexity and lattice defects.^[88] This phenomenon has a direct impact on the conductivity and cycling durability of the battery, ultimately constraining the performance of SSE batteries. Due to the complexity of the crystal structure, including variations in lattice constants, defect types, and their distribution, ions exhibit uneven diffusion rates within the electrolyte. Furthermore, lattice defects such as vacancies and impurities create traps that impede the free movement of ions, increasing the difficulty of their migration within the structure.^[89] These factors collectively contribute to severe limitations in the performance of SSE batteries regarding enhanced energy density and prolonged cycling life. To address the challenge posed by the migration barrier, researchers have employed various strategies, including optimizing crystal structures, controlling lattice defect distribution, and introducing conductive pathways to enhance the performance of electrolytes.^[90–92] Additionally, the use of novel materials and advanced fabrication techniques is considered an effective avenue for reducing the migra-

tion barrier. Through these innovative studies and technological developments, substantial advancements have been achieved in attaining both high conductivity and long-term durability in SSE batteries, laying a solid foundation for the future advancement of high-performance battery technologies.^[93]

The chemical properties such as the charge, size, and chemical stability of ions, the physical properties such as crystal structure, lattice constant, and pore structure, as well as the structural characteristics such as grain size, grain boundaries, and pore structure, all have an impact on the migration of ions in SSEs. These properties collectively influence the diffusion pathways, migration rates, local concentrations, and migration resistance of ions in SSEs. Therefore, optimizing these properties can effectively regulate the ion transport performance of SSEs. Kim and colleagues harnessed ML techniques to scrutinize how diverse chemical, physical, and structural traits impact ion mobility in the lattice matrix.^[94] They studied the anti-perovskite crystal structure and trained multiple ML algorithms for predicting ion migration barriers (Figure 6a). The training dataset consisted of over 600 barriers for the algorithms, consistently computed through DFT. This dataset encompasses 36 anti-perovskites, specifically alkali metal chalco-halides, including vacancy and interstitial migration pathways. As illustrated in Figure 6b, superfluous features were identified and eliminated to streamline models and prevent overfitting. A significant observation is that the most effective ML models make use of more concise feature sets, with five features for vacancy migration and three features for interstitial migration, in contrast to the extensive 20 feature models. Nevertheless, the earlier models exhibited enhanced accuracy. The most precise algorithm was employed to discern feature combinations that generated the most dependable predictions. For instance, as depicted in the scatter plot of Figure 6c, the 5-feature model for vacancy migration showcased an RMSE of 71 meV, in contrast to the 86 meV RMSE observed in the 20-feature model. When analyzing interstitial migration, the RMSE for a 20-feature model was 55 meV, while it was 46 meV for a 3-feature model. Anion polarizability was reasserted as an important mobility-affecting attribute in this evaluation, while shedding light on others that were not as widely acknowledged. Specifically, vacancy migration is 70% determined by lattice characteristics, including the hopping distance and channel width, while interstitial migration is $\approx 50\%$ determined by lattice characteristics. Through this analysis, critical properties of SSEs were identified to simplify the design process. However, for a broader application across various solids, it was imperative to extend these findings to other crystal structure categories. By including dynamic phenomena such as vibrations in lattices and rotations of polyanion molecules, the feature set could be made more practical. Additionally, lithium thiophosphate glasses ($\text{Li}_2\text{S}-\text{P}_2\text{S}_5$) hold considerable promise as SSEs for batteries. However, a limited comprehension of how the disordered structural attributes impact lithium transport properties has hindered the advancement of glassy electrolytes. To tackle this challenge, Chen et al. conducted simulations of glassy $\text{Li}_2\text{S}-\text{P}_2\text{S}_5$ electrolytes, introducing varying fractions of polyatomic anion clusters, specifically $\text{P}_2\text{S}_6^{4-}$, $\text{P}_2\text{S}_7^{4-}$, and PS_4^{3-} , employing classical MD.^[95] To discern alterations in ionic conductivity, they harnessed a classification-based ML metric referred to as “softness” to elucidate the underlying structural factors influencing lithium-ion mobility. The distribution of

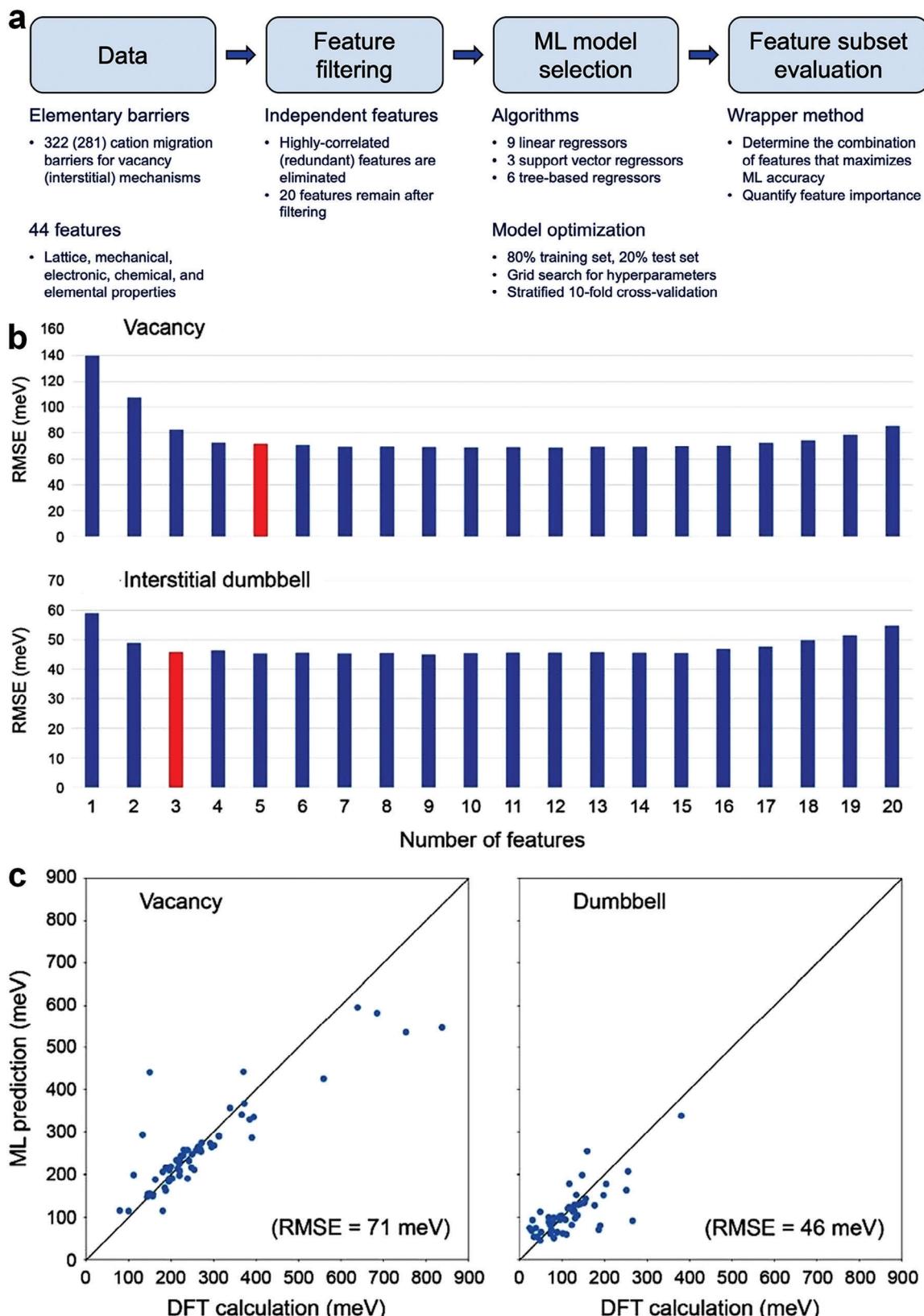


Figure 6. Uncovering the factors influencing ion mobility in anti-perovskite SSEs via ML. a) ML framework to predict ion migration barriers. b) Comparison of vacancy and interstitial dumbbell mechanisms in “adaboost + ERTR” models. c) Vacancy migration barriers and dumbbell migration barriers are predicted by different features. Reproduced with permission.^[94] Copyright 2022, The Royal Society of Chemistry.

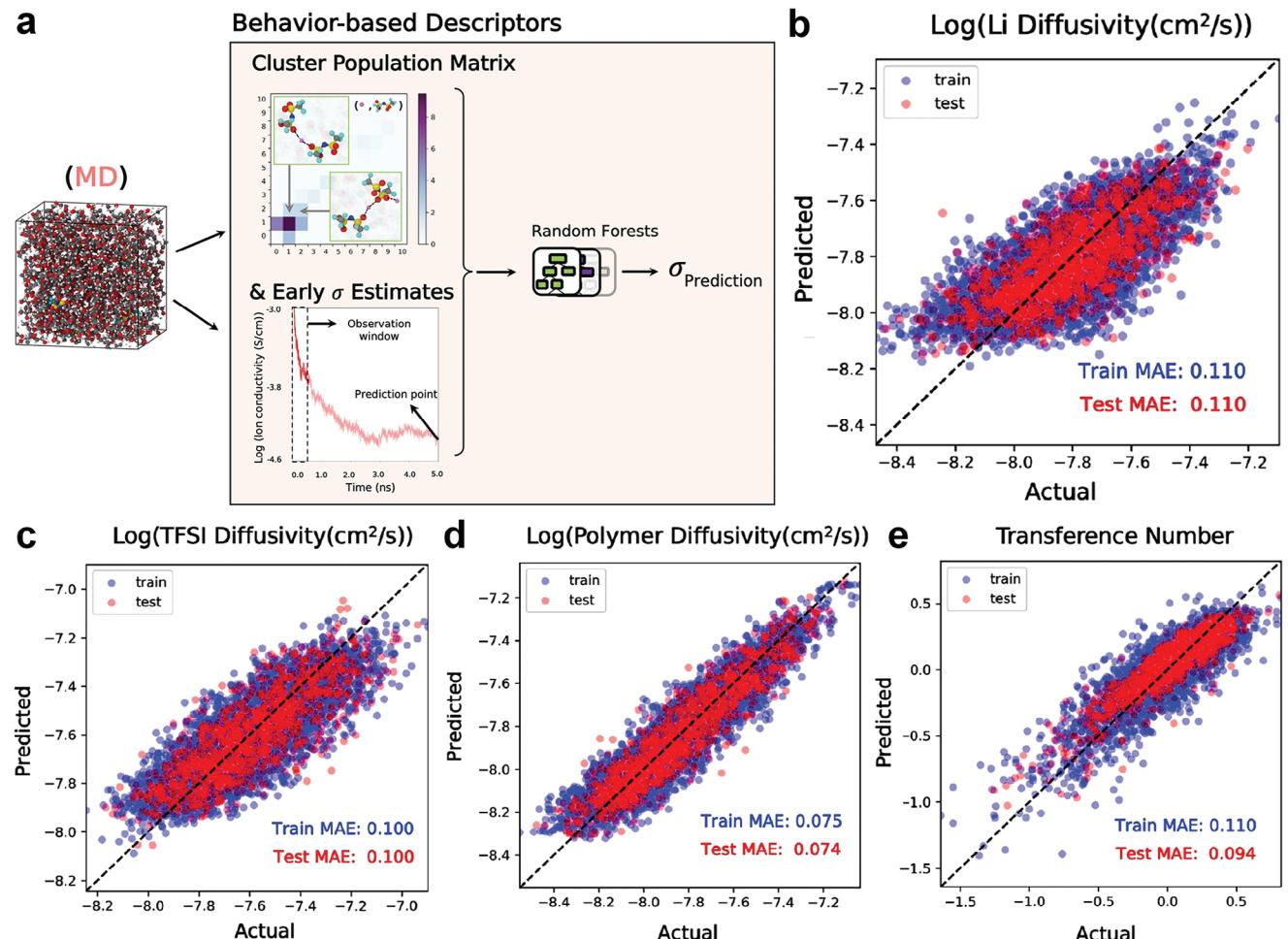


Figure 7. Ion transport properties predicted by ML. a) ML workflow. Actual and predicted plots for b) Li^+ diffusivity, c) TFSI^- diffusivity, d) polymer diffusivity, and e) transference number in the training and test sets. Reproduced with permission.^[108] Copyright 2023, American Chemical Society.

softness among lithium ions exhibited a pronounced spatial correlation: notably, lithium ions characterized as “soft” (indicating high mobility) were predominantly situated in proximity to PS_4^{3-} units, while those categorized as “hard” (reflecting low mobility) were predominantly located in the vicinity of $\text{P}_2\text{S}_6^{4-}$ units. Additionally, the study demonstrated that the migration of soft lithium ions entails surmounting a lower energy barrier compared to the barrier encountered in the case of hard lithium-ion migration.

3.4. Diffusivity

The ion diffusivity within the electrolyte lattice is a pivotal property in this domain, exerting substantial influence over the rate at which ions migrate.^[96–99] It is crucial to comprehend the intricate interplay between diffusivity and the performance of SSEs for the refinement of state-of-the-art electrochemical systems. Within the realm of SSEs, diffusivity encompasses the dynamics of ion movement, providing essential insights into the material’s capacity for ion transport.^[100,101] The relevance of this property is particularly pronounced in the crafting and optimization of solid-state batteries, fuel cells, and sensors. Efficient ion migration

is imperative for achieving high energy densities, swift charge-discharge rates, and responsive sensing capabilities.^[102–104]

MD simulations serve as valuable tools for the evaluation of solid polymer electrolytes possessing attributes suitable for utilization in Li-ion batteries.^[105–107] Nevertheless, given the extensive space of polymer designs, expediting the screening process becomes imperative, necessitating a reduction in computational time about ion transport properties in simulations. A unique set of descriptors, derived from a 0.5 ns observation window at the outset of MD simulations, was utilized and assessed by Kha-jeh et al. for predicting ionic properties (Figure 7a).^[108] These descriptors, originating from behavioral characteristics, fused the ion cluster population matrix with as-yet undiscovered ionic transport traits calculated via the cluster Nernst–Einstein equations. Diverging from alternative methods like SMILES-derived descriptors and molecular graphs, which rely exclusively on the chemical composition of the polymer, behavior-based descriptors obtained from MD simulations can capture both ion interactions and the dynamic progression of ion transport. The study exhibited that descriptors based on the initial temporal segments of simulations enhanced predictive accuracy in comparison to previous studies that solely relied on the 2D structure of polymers.

Moreover, it was found that 0.5 ns observation windows were sufficient for predicting conductivity more accurately. To examine the capacity of the ML models to apply their learned knowledge to previously unobserved data while in the training phase, a comparison of prediction performance between the training and testing datasets was conducted, as illustrated in Figure 7b–e. Standard descriptors based on behavior were employed in the training of the RF models. The visual depiction illustrated the spread of actual and forecasted values, complemented by MAEs for predictions in both the cross-validation and hold-out test sets. Notably, the MAE for predictions in the test set exhibited a level of comparability to that of the cross-validation set. Additionally, the distributions of predicted values displayed substantial similarity between the test and cross-validation sets. These observations assure that the model is expected to perform adequately, at least for datasets with a similar polymer distribution to the one in the current dataset.

3.5. Ionic Conductivity

Ionic conductivity emerges as a crucial physical property directly impacting the performance of electrolyte materials in practical devices.^[109–112] It dictates the rate at which ions traverse the electrolyte when subjected to an electric field, exerting a substantial effect on the overall performance of batteries. Ionic conductivity is a crucial parameter determining the speed of ion conduction in SSE materials when exposed to an external electric field. Elevated levels of ionic conductivity efficiently facilitate rapid ion transport, thereby enhancing the discharge performance of batteries.^[113] Moreover, ionic conductivity exerts an influence on the internal resistance of SSEs, a critical parameter in battery operation.^[114] Reduced internal resistance leads to heightened energy conversion efficiency and swifter charge–discharge rates, ultimately bolstering the overall battery performance. Additionally, the level of ionic conductivity is closely linked to the stability of SSE materials. Under harsh working conditions, such as high temperatures and voltages, specific electrolyte materials may experience structural degradation or chemical alterations, resulting in reduced ionic conductivity.^[115]

The effectiveness of ML models in predicting material properties relies heavily on the availability of high-quality data.^[116–118] Hargreaves et al. introduced a meticulously curated dataset centered on lithium-ion conductors, along with their respective conductivities determined through impedance spectroscopy.^[119] The compilation includes 820 entries sourced from 214 different references. Each entry includes details on the chemical makeup, a label given by experts based on structural characteristics, and the ionic conductivity at a particular temperature. At room temperature, there are approximately 403 distinct chemical compositions, each associated with a particular ionic conductivity value. Compounds documented in the inorganic crystal structure database correlate with the materials cataloged in this dataset. This was achieved by applying unsupervised ML techniques and employing the element movers distance metric. A classifier, built on the CrabNet architecture and trained using this dataset, is capable of determining whether a given chemical composition displays high or low ionic conductivity. This classifier provides a practical means for experimentalists to efficiently identify candidates war-

ranting further study as potential lithium-ion conductors. For instance, an ML model was used by Adhyatma et al. to classify the ionic conductivity of doped LLZO.^[120] A classifier, founded on the light GB machine algorithm, was crafted through rigorous procedures including model selection, validation, and optimization. This classifier achieved an accuracy score of 0.903 in leave-one-out cross-validation. Two pivotal factors emerged for attaining heightened ionic conductivity in doped LLZO. This investigation exemplifies the efficacy of robust data-driven approaches leveraging readily accessible features in expediting the advancement of innovative SSE design. By fortifying safety measures and allowing for greater energy densities, solid polymer electrolytes (SPEs) offer an efficient avenue for improving LIBs. Nevertheless, the ionic conductivity of SPEs is considerably lower in comparison to their liquid and solid ceramic counterparts. This restriction presents a challenge to their successful integration into operational batteries. An ML model based on chemistry-informed predictions of SPE ionic conductivity was developed by Bradford et al. to accelerate the discovery of high-ionic conductivity SPEs.^[121] This model underwent thorough training on an extensive dataset of ionic conductivity information sourced from numerous experimental publications. Their chemistry-informed model ingeniously encapsulates the Arrhenius equation, a fundamental descriptor of temperature-driven processes, within the readout layer of an advanced message-passing neural network. This augmentation in model architecture has yielded a substantial improvement in accuracy when compared to models that neglect temperature dependence. Integrating chemically informed readout layers also allows for support for various property prediction techniques based on deep learning, proving particularly advantageous in scenarios characterized by limited training data availability. By effectively utilizing this well-trained model, the researchers extrapolated ionic conductivity values for a multitude of candidate SPE formulations, thus affording the identification of promising SPE candidates. Furthermore, their model extended its utility by furnishing predictions for several distinct anions within the frameworks of poly(ethylene oxide) and poly(trimethylene carbonate), thereby underscoring its capacity to elucidate crucial descriptors governing SPE ionic conductivity. Mandal et al. introduced a combined experimental and ML methodology to facilitate the identification of glass electrolytes, simultaneously shedding light on the functional contributions of distinct glass constituents (Figure 8a).^[122] Specifically, they conducted empirical preparations and measurements of ionic conductivity for 27 compositions within the sodium aluminophosphate glass family. Subsequently, ML models were trained on this dataset to forecast ionic conductivity, demonstrating commendable concordance with the experimental findings (Figure 8b). The authors elucidated the relationship between composition and conductivity gleaned from the ML model through Shapely additive explanations (SHAP), thus unveiling the influential role of the glass components in dictating conductivity (Figure 8c–e). By capitalizing on these insights, glass formulations with enhanced conductivity characteristics were anticipated and subsequently validated through experimentation. Notably, the experimental synthesis yielded a glass composition with a remarkable conductivity value of 2.44×10^{-6} S cm⁻¹ at 373 K, the highest among all tested compositions. These results validate the findings derived from SHAP analysis and empower the refinement

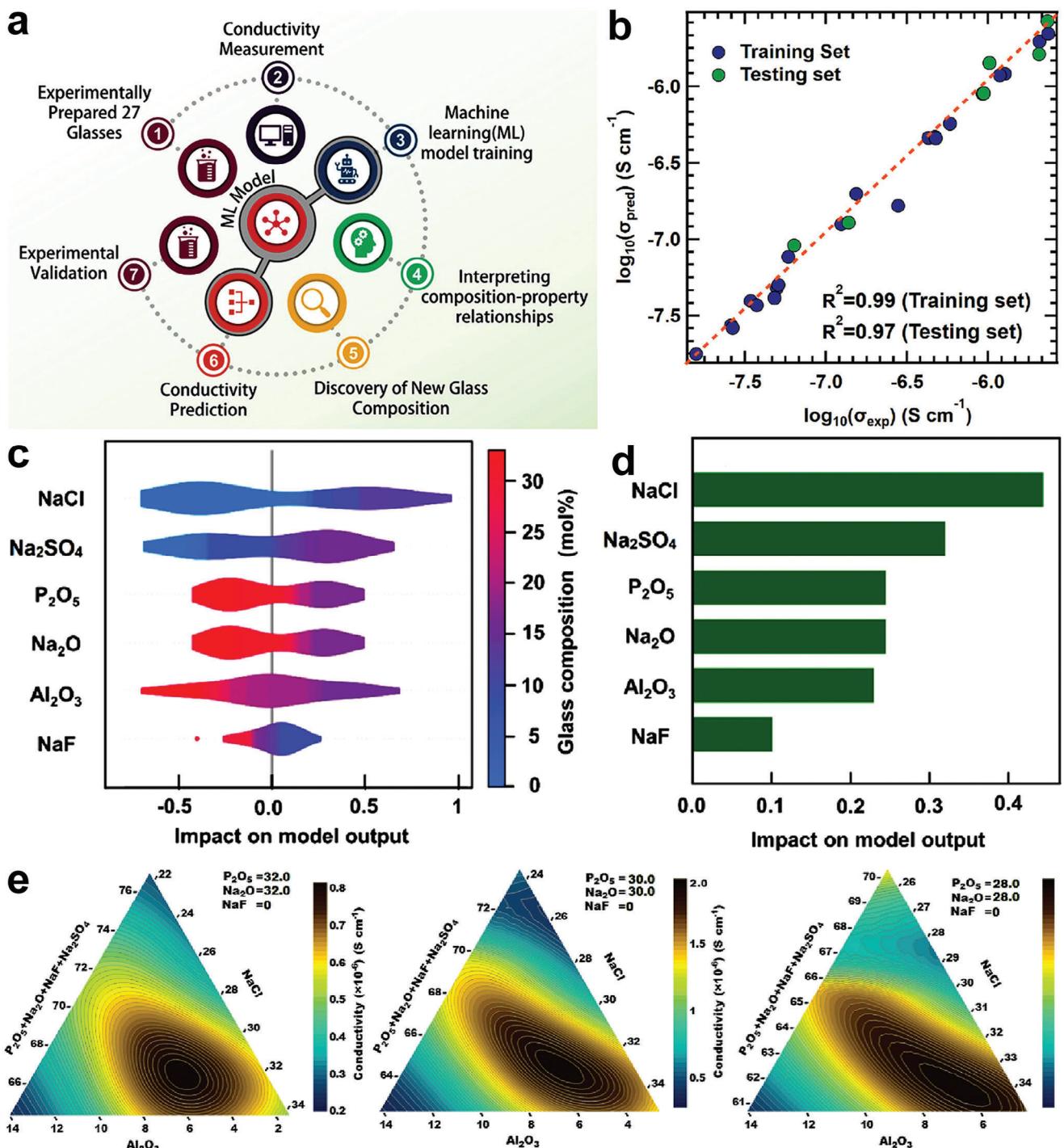


Figure 8. Design of Na-ion conducting glasses with ML. a) Conductivity prediction process using ML. b) Prediction and experiment plots for training and test sets. SHAP c) violin and d) bar plots. e) Conductivity plot for ML outputs. Reproduced with permission.^[122] Copyright 2023, American Chemical Society.

of glass compositions for practical applications. This exemplifies the notable capacity of ML tools in expediting the discovery of sodium-ion-conducting glass electrolytes.

While ML stands as an efficient approach, contemporary ML models rely on sophisticated, manually curated features derived from human intuition. This practice can introduce subjective bi-

ases, prove potentially challenging to acquire for a wide array of materials, and lead to a laborious feature selection process.^[123] In response to these challenges, Lu et al. have devised a neural network-based model that requires just three input features for each atom to forecast the presence of superionic conductivity in solid materials for SSE applications.^[124] Importantly, this

approach automatically captures any potential secondary features, significantly diminishing the necessity for human intervention during model training. Such a model exhibits the potential for broader applicability compared to manually feature-dependent models and can even be extended to forecast other material properties, all while substantially reducing complexity and training duration.

3.6. Modulus

The impact of modulus, especially the elastic or Young's modulus, on the functionality of solid-state electrolytes, stands as a pivotal facet in the material design of electrochemical devices.^[125–127] A higher elastic modulus endows the electrolyte with superior mechanical stability, conferring the ability to endure external stresses without undergoing plastic deformation or fracture. This characteristic is especially crucial in applications subjected to mechanical loading, as it ensures the structural integrity of the electrolyte over extended operational periods. However, it is noteworthy that an elevated elastic modulus may result in a modest reduction in ionic conductivity. This stems from the inherent balance between mechanical stiffness and the ability of ions to move within the crystal lattice. The stiffer lattice structure restricts ion movement, which can impede the rate of charge transfer. Therefore, striking an optimal balance between mechanical integrity and ionic transport properties becomes imperative in electrolyte material selection for specific applications. Furthermore, the modulus is a key factor in deciding the compatibility of the electrolyte with the neighboring components, particularly at interfaces. Materials with comparable moduli are more likely to form stable and well-adhered interfaces with electrodes or other adjacent materials. Achieving interfacial compatibility is critical in the design of solid-state batteries and various electrochemical devices, as it directly influences overall device performance and longevity.^[128] Additionally, the elastic modulus influences the thermal expansion behavior of the electrolyte material. A material with a higher elastic modulus tends to exhibit reduced volume changes in response to temperature fluctuations.^[129] This attribute mitigates the potential for thermal stresses, ensuring the mechanical integrity of the electrolyte even under varying thermal conditions.

Li-ion SSEs hold significant potential, but their widespread adoption faces obstacles linked to the stability of interfacial contacts and the formation of dendrites.^[130] Within this investigation, Min's research group employed an ML regression model to identify mechanically superior Li-ion SSEs from a pool of 17 619 candidates (Figure 9a).^[131] To achieve this, a surrogate model was developed, utilizing a pre-existing elasticity database for inorganic materials (Figure 9b), enabling the prediction of shear modulus (G) and bulk modulus (K) through Voigt–Reuss–Hill (VRH). Subsequently, G_{VRH} and K_{VRH} values, based on physicochemical and structural attributes, were derived. The model was subsequently employed to forecast the mechanical characteristics of prospective Li-ion SSEs, with further validation through DFT calculations. To mitigate bias, a stratified sampling approach was implemented. Specifically, training sets for the G_{VRH} model were stratified into intervals of 0, 40, 80, 120, 160, 200, and beyond, while for the K_{VRH} model, intervals of 0, 50, 100, 150, 200, 250,

and beyond were utilized. This differentiation was necessitated by the wider distribution of K_{VRH} values compared to those of the G_{VRH} model. In light of the finite resources of materials endowed with exceptional mechanical attributes, materials surpassing the thresholds of 320, 360, 400, and 480 GPa for the G_{VRH} prediction model, as well as 400, 450, 500, and 550 GPa for the K_{VRH} prediction model, were incorporated into the training set to improve prediction precision. Subsequently, their performance was assessed in comparison to the standard stratified sampling approach (Figure 9c,d). Afterward, ten prediction models were formed for each sampling approach, and the R^2 and MAE values were averaged. The results revealed that standard stratified sampling yielded the highest prediction accuracy for G_{VRH} , achieving R^2 and MAE values of 0.822 and 9.7 GPa, respectively. In contrast, for K_{VRH} , materials above 450 GPa demonstrated superior performance, attaining R^2 and MAE values of 0.892 and 14.0 GPa, respectively. Consequently, the study proceeded with the application of these models in the subsequent phase. Figure 9e,f display the scatter plots for the most precise model out of the ten predictions in each scenario, corresponding to G_{VRH} and K_{VRH} , respectively. This investigation underscores the efficacy of ML in expediting material screening for the identification of mechanically superior Li-ion SSEs, bolstered by an optimization procedure. Furthermore, the compiled material database presents crucial design insights for the progress of future-generation solid-state batteries. In addition, their research team formulated an ML-driven regression model for the prognostication of mechanical attributes in Na-SSEs.^[132] To constitute the training dataset, a compilation of 12361 materials was sourced from an extensive materials repository, with each material being meticulously characterized by its corresponding structural and chemical descriptors. The devised surrogate model displayed exceptional levels of precision, quantified at 0.72 and 0.87, alongside mean absolute deviations of 11.8 and 15.3 GPa for shear and bulk modulus, correspondingly. Subsequently, the mechanical features of 2432 Na-SSEs were systematically predicted using this model, and validated through rigorous first-principles computations. The platform proposed in this investigation stands poised to expedite the quest for Na-SSEs possessing optimal mechanical characteristics while simultaneously minimizing resource expenditure.

3.7. Thermal Expansion

Thermal expansion is a critical factor influencing the performance of SSEs. Electrolytes with well-matched thermal expansion properties can maintain structural stability under temperature variations, reducing the risk of thermal stresses and cracks.^[133–135] Conversely, mismatched thermal expansion coefficients between the electrolyte and surrounding materials can lead to structural degradation or interface delamination during device operation. Therefore, in devices like solid-state batteries, selecting electrolyte materials with appropriate thermal expansion characteristics is paramount to ensuring stable performance across varying temperature environments. Furthermore, a comprehensive understanding and control of thermal expansion provide crucial insights for optimizing the compatibility of the electrolyte with other components, such as electrodes. This ensures robust interfaces, which in turn enhance the overall performance

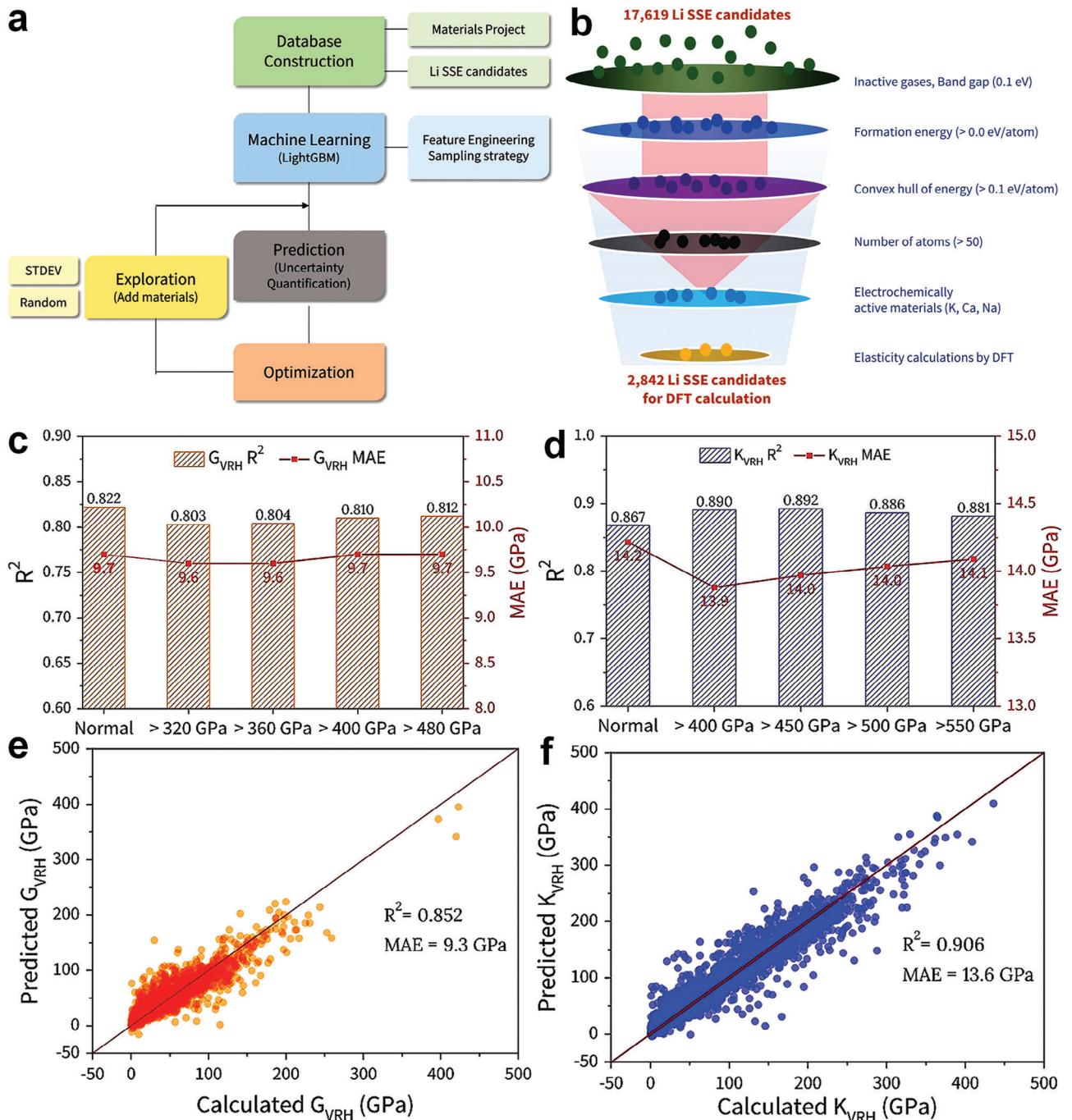


Figure 9. A data-driven approach to finding mechanically superior SSEs for LIBs. a) SSE mechanical property prediction flowchart by ML. b) Candidate selection for Li-ion SSE. Prediction accuracy varies for G_{VRH} c) and K_{VRH} d). Calculated verse predicted plots for G_{VRH} e) and K_{VRH} f). Reproduced with permission.^[131] Copyright 2021, American Chemical Society.

and reliability of the device. Additionally, the management of thermal expansion effects facilitates the design of mechanically robust and thermally stable energy storage systems, particularly in applications where temperature fluctuations are prevalent.^[136] For material science and device engineering, accounting for thermal expansion behavior is essential for achieving long-term operational stability and performance consistency. It underscores the

interdisciplinary nature of SSE research, where considerations spanning from crystallography to device integration play pivotal roles in advancing the field.

Developing thermomechanically compatible cathode and electrolyte materials is crucial for improving the performance and safety of LIBs. These materials need to withstand mechanical and thermal stresses during battery operation, including

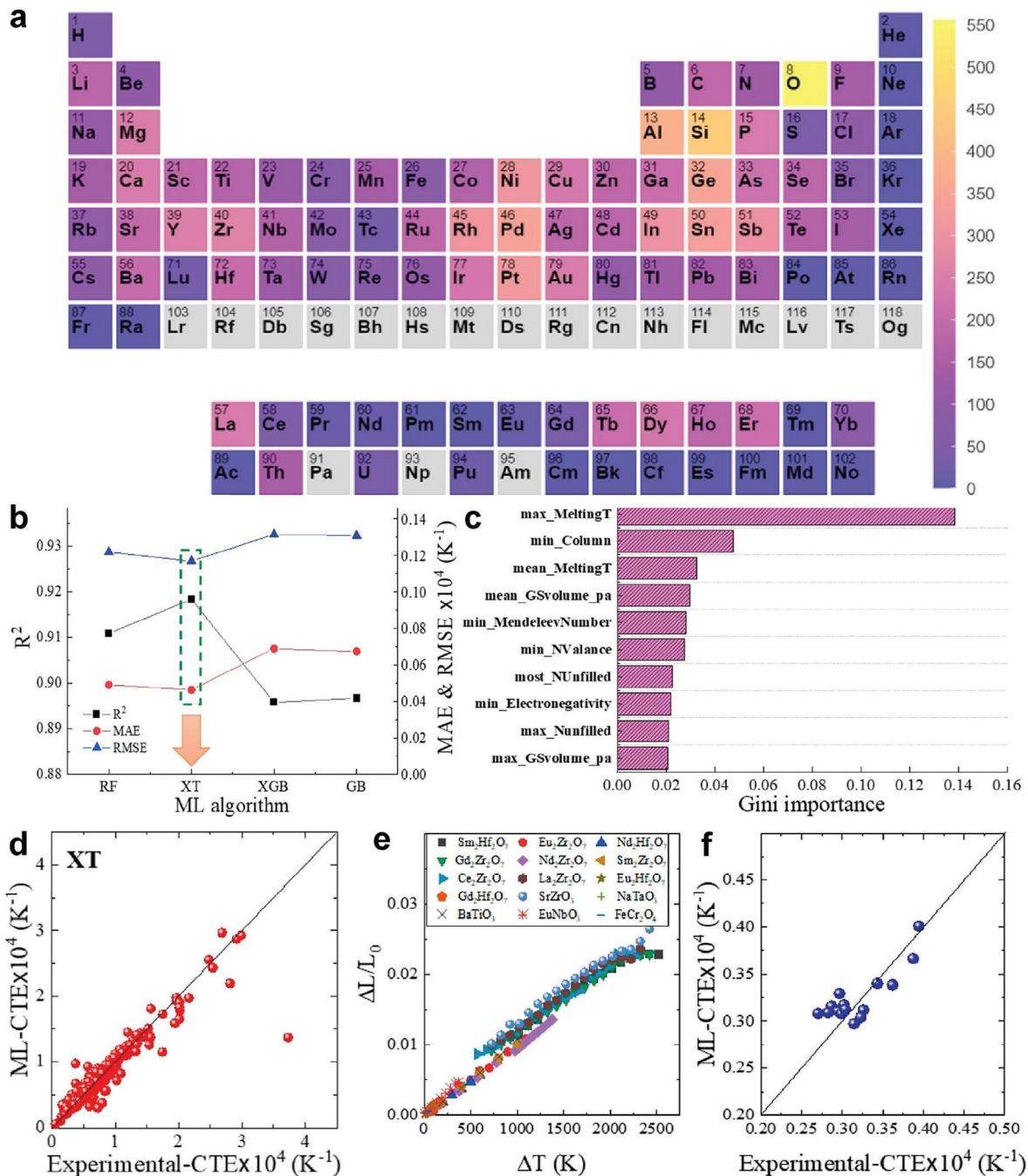


Figure 10. Developing stable solid-state LIBs with strong thermomechanical compatibility. a) Data characteristics for ML. b) Algorithm performance comparison in ML. c) Feature importance. d) The results were tested by the XT model. e,f) Model validation using XT. Reproduced with permission.^[137] Copyright 2023, Elsevier.

charging and discharging cycles, and exposure to high temperatures. Additionally, they should have good chemical compatibility to ensure long-term stability and reliability. Research and development efforts are focused on identifying and designing new materials with the desired properties, as well as optimizing manufacturing processes for successful integration into LIBs. Kumar et al. have showcased the utilization of ML techniques in predicting thermomechanically compatible cathode and elec-

trolyte materials for LIBs.^[137] A dataset of 5578 materials was used to train the ML models, with the coefficient of thermal expansion (CTE) chosen as the target property (Figure 10a). Figure 10b illustrates the R^2 , MAE, and RMSE values derived from a 10-fold cross-validation process for four models (extra trees (XT), RF, extreme gradient boosting (XGB), and GB) after hyperparameter optimization, with the XT-model demonstrating the highest level of precision. An assessment of feature

importance for the XT-model (Figure 10c) uncovered that the melting temperature exerts the most substantial influence on CTE. Due to the interdependence between melting temperatures and chemical bond strength, this observation was in accordance with the established physical principle that melting temperatures and CTE demonstrate an inverse relationship. To enhance clarity, the testing results were also broken down into a representative subset (Figure 10d). The optimized XT model underwent an initial evaluation via 10-fold cross-validation, followed by subsequent validation utilizing an experimental dataset (Figure 10e,f). After validation, CTE values for 25 819 materials were predicted using the model. The forecasted data was subsequently utilized for the assessment and screening of cathode and electrolyte materials, specifically considering their thermomechanical compatibility and CTE values.

3.8. Reaction Energy

The reaction energy, which refers to the energy involved in the interaction between the SSE and the electrodes, is a crucial factor in assessing the battery performance.^[138] It acts as a bridge between the thermodynamic stability of the interface and the kinetics of ion transport.^[139] Fundamentally, the sustainability of electrochemical processes within the battery hinges on the thermodynamic integrity of the SSE-electrode interface.^[140] An unstable interface can lead to undesirable reactions, such as electrolyte decomposition or electrode passivation, which can significantly reduce the overall efficiency and lifespan of the battery.^[141,142] On the contrary, a stable interface fosters favorable conditions for ionic conduction and electron transfer, thus contributing to enhanced battery performance. Beyond thermodynamics, the kinetics of ion transport across the SSE-electrode interface are equally pivotal. The rate at which ions can migrate between the anode and cathode influences critical parameters such as the battery's charge/discharge rate, power density, and efficiency. A high reaction energy barrier can impede ion transport kinetics, leading to sluggish charge/discharge processes and diminished power output. Conversely, a lower reaction energy barrier can facilitate rapid ion movement, enabling fast charging and discharging, which is particularly vital for applications demanding high-power bursts.^[143] A profound understanding of how reaction energy influences these crucial aspects is imperative for tailoring SSEs to satisfy the high-level demands posed by advanced energy storage technologies. By strategically adjusting the reaction energy between the SSE and electrodes, the interface becomes stable thermodynamically and efficient kinetically.^[144] This fine-tuning can yield SSEs that strike a harmonious balance between stability and reactivity, resulting in batteries that exhibit superior performance, safety, and longevity.

LSBs present a compelling option for energy storage, primarily owing to their cost-effectiveness and plentiful supply of sulfur. Nevertheless, the challenge of polysulfide shuttling and subsequent capacity degradation poses a significant barrier to commercialization.^[145–147] One potential solution lies in replacing conventional liquid electrolytes with SSEs. To address these issues, Chandrappa et al. conducted an extensive investigation into the thermodynamics and kinetics at the interface between the cathode and electrolyte within all-solid-state LSBs, employing

DFT-based calculations in conjunction with an ML interatomic potential (Figure 11a).^[148] Their results indicated that, among various SSE chemistries, sulfide-based SSEs were generally projected to exhibit the highest stability in the presence of the S₈ cathode. In contrast, other SSE chemistries were expected to be notably electrochemically unstable. In cases where alternative SSE chemistries were preferred for specific reasons, numerous binary and ternary sulfides were envisioned to serve as excellent buffer layers. Furthermore, they developed a precise moment tensor potential through an active learning approach to investigate the S₈|β-Li₃PS₄ interface. As illustrated in Figure 11b, the MAE for reaction energy (referred to as MAE_{energies}) was compared across all interfaces, with consistent alignment to the results obtained during the active learning iterations. Meanwhile, the MD simulations, involving extensive interface models, demonstrated that the Li₃PS₄(100) surface, being the most stable, shows a propensity to create interfaces with S₈. These interfaces feature 2D channels and exhibit lower activation barriers for lithium diffusion. The results offer crucial fresh perspectives on how to engineer the cathode-electrolyte interface for the forthcoming era of all-solid-state LSBs.

3.9. Other Properties

Other SSE properties, such as crystal structure, atomic structure, and reaction mechanism have also been deeply studied. In the realm of computational material discovery, high-throughput methodologies frequently encounter a challenge characterized by a combinatorial explosion.^[149] This obstacle makes it unfeasible to thoroughly explore the entire range of structural and chemical possibilities. An issue that arises is the difficulty in evaluating new compounds with conventional crystal structures. This is mainly because there is a lack of fast and reliable decision-making frameworks for accurately categorizing computed candidates as either conforming to the norm or deviating significantly (structures with excessive distortion). To address this predicament, ML-driven workflows offer a viable solution, enhancing the efficiency of geometric optimization procedures.^[150] However, the absence of appropriate pairings of geometric descriptors and exceptionally precise unsupervised models remains a significant hurdle in effectively distinguishing systems with subtle structural distinctions. Notably, Gallo-Bueno et al. identified Steinhardt order parameters as remarkably precise descriptors of the cubic argyrodite structure, facilitating the training of a variety of commonly used unsupervised outlier detection models.^[151] Most significantly, this approach empowered them to automatically classify crystal structures while maintaining control over uncertainty levels. Moreover, despite considerable work invested in producing SSEs with high ionic conductivities, gaining a complete comprehension of the ionic diffusion processes in SSEs remains elusive when employing conventional experimental and theoretical approaches. In the research carried out by Fu et al., the temperature-dependent concerted diffusion mechanism of ions within SSEs was investigated through ML and MD, utilizing Li₁₀GeP₂S₁₂ as a representative prototype.^[152] Elevated temperatures revealed diminished diffusion anisotropy, heightened disorder in Li distributions, and abbreviated residence times. Remarkably, across a wide temperature range, the

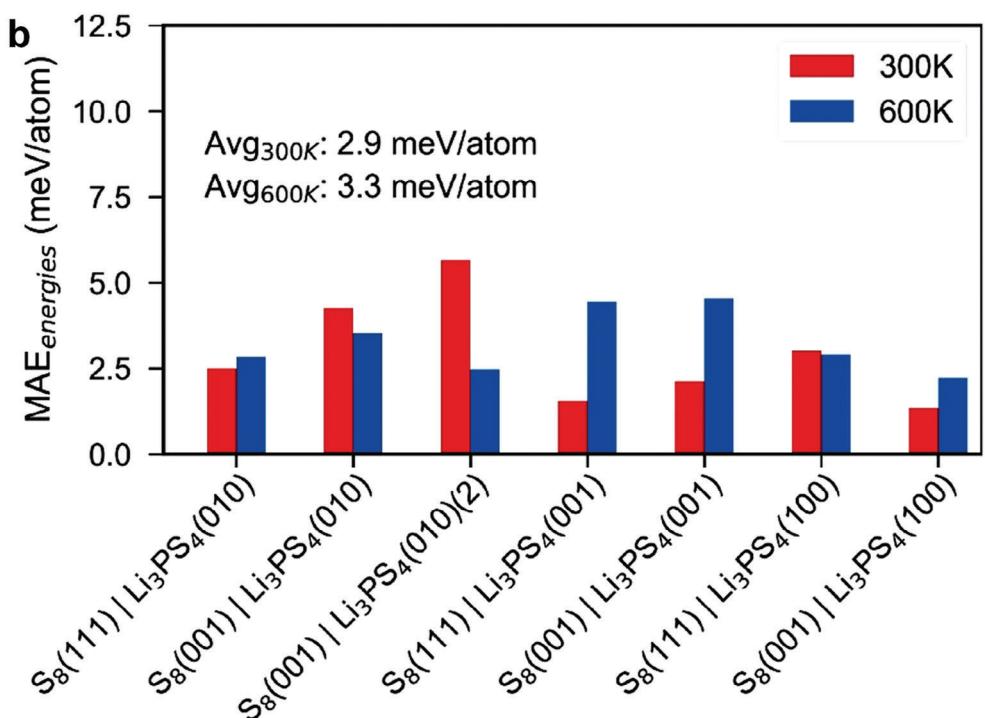
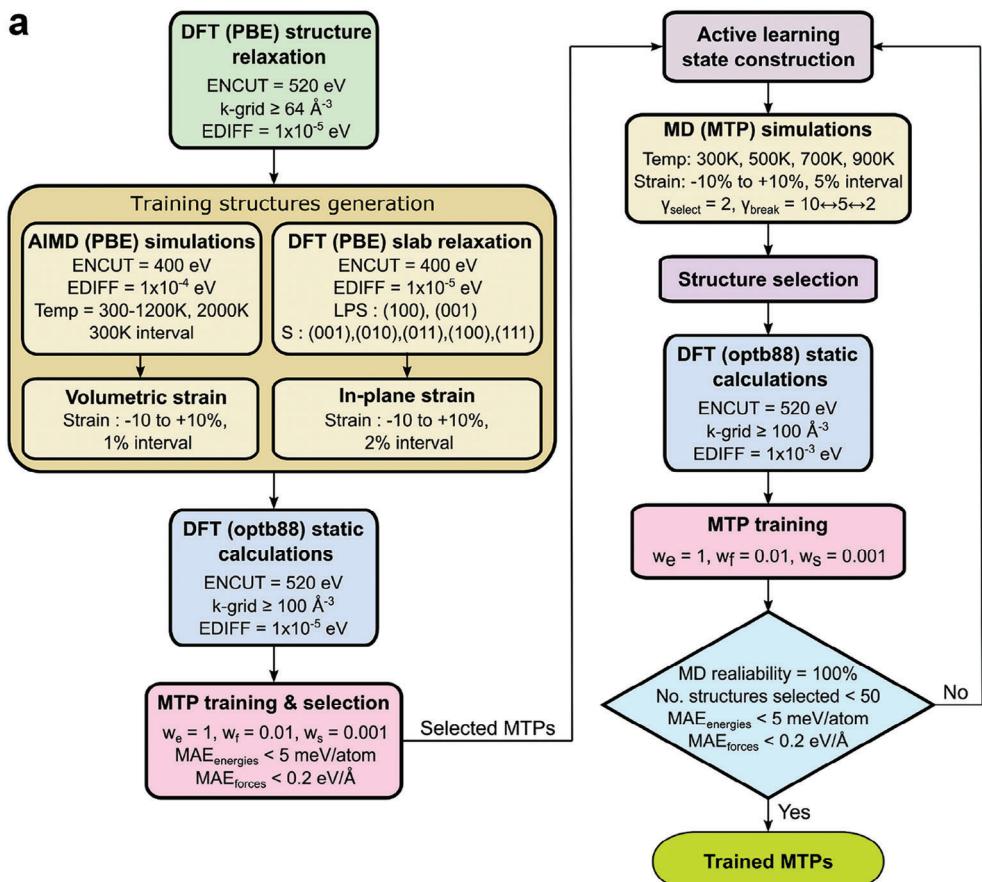


Figure 11. Cathode-electrolyte interface reaction energy prediction. a) Diagram for the passive and active learning in the advancement of interfacial MTP. b) MAE between MTP-predicted energies and DFT values at temperatures of 300 K and 600 K. Reproduced under the terms of the CC-BY 4.0 license.^[148] Copyright 2022, The Authors, published by American Chemical Society.

temperature dependency demonstrated an Arrhenius-type behavior. This was ascribed to the linear temperature-dependent modification of jump frequencies linked to different coordinated diffusion modes. The findings contribute significantly to the establishment of a theoretical framework aimed at unraveling the ionic diffusion mechanisms within SSEs, thereby enhancing our comprehension of the underlying chemical underpinnings governing temperature-dependent concerted diffusions in SSE materials. Besides the SSE materials, the SSE interphase (SEI) characterization is also very important.^[153–155] The technique of X-ray photoelectron spectroscopy (XPS) is a robust surface analysis tool extensively used in examining the SEI of lithium metal batteries.^[156] Nevertheless, utilizing the XPS technique exclusively proves inadequate in elucidating atomic structures within the deeply embedded SEI, leading to critical information gaps. In a pioneering approach, Sun et al. have introduced an artificial intelligence ab initio (AI-ai) framework, which integrates ab initio computations, reactive MD, and ML models to forecast XPS spectra associated with the SEI.^[157] Through the meticulous simulation of a localized high-concentration electrolyte system with a lithium metal anode, the hybrid ab initio reactive scheme was employed for a duration of roughly 3 ns. With the local many-body tensor representation serving as a distinctive descriptor, four ML models were applied to precisely predict core-level shifts. Remarkably, XGB proved to be superior in terms of accuracy and reduced variance when compared to the other ML models. This innovative AI-ai model facilitates the prediction of XPS spectra for a substantial ten thousand frames at a minimal computational cost.

Beyond just predicting SSE performance, ML methodologies are crucial in deciphering complex patterns in battery usage and degradation. This analytical power is essential for forecasting future performance, lifespan, and potential failures, significantly enhancing battery reliability, efficiency, and safety in diverse applications such as electric vehicles and renewable energy storage systems. Likewise, ML is transforming the field of solid material screening, particularly in the context of catalysts, by streamlining the discovery and optimization processes.^[158] Catalysts, pivotal in numerous chemical processes, have their efficiency and selectivity directly impacting process efficiency and environmental impact. ML's capability to quickly analyze extensive datasets on material properties and reaction conditions enables rapid identification of promising catalyst candidates, dramatically reducing the time and resources needed for traditional experimental procedures. The integration of ML in both battery performance prediction and material screening is not just accelerating research and development but is also opening up new possibilities in material discovery and process optimization—areas often too complex for conventional methods. This progress is set to drive major breakthroughs in energy storage, chemical manufacturing, and environmental sustainability, showcasing the transformative power of ML in these advanced applications.

4. Conclusions and Perspectives

From a technical perspective, implementing ML in materials science provides researchers with unprecedented tools and resources to gain a deep understanding of SSE properties. This will aid in rapidly screening materials, expediting the discovery and

development of new materials, thereby driving innovation within the domain of energy storage. Furthermore, as renewable energy sources experience swift growth, the demand for energy storage solutions continues to escalate. By accurately predicting and optimizing SSE performance through ML, enhancements in the efficiency and dependability of energy storage systems expand their applicability, catering to a diverse array of needs, from small-scale electronics to large-scale energy grids. Therefore, the prospects for ML in predicting SSE properties are extremely promising, with its impact extending not only to the scientific community but also making a valuable contribution to the sustainable development of the global energy sector. In the field of ML for predicting SSE properties (Figure 12), several directions can accelerate progress toward more sustainable and efficient energy storage solutions:

(1) Real-Time Property Prediction: The real-time prediction of SSE properties stands as a critical element in the ongoing quest to optimize energy storage systems. In the future, it is expected that ML models will advance to a heightened level of sophistication, enabling precise predictions of material behavior under diverse and dynamic conditions. These advanced models will likely integrate an array of dynamic factors, such as temperature, pressure, and the state of charge in batteries, allowing for a more comprehensive understanding of the complex interplay of variables influencing SSE performance. This future generation of ML models will be equipped to continuously monitor a multitude of parameters within SSEs, including chemical reactions, temperature fluctuations, voltage, and current. This dynamic monitoring capability is pivotal in the early identification of potential safety concerns, such as overheating, overcharging, or over-discharging. The ability to promptly detect abnormalities enables the implementation of timely interventions and precautionary measures, ensuring the safe and reliable operation of energy storage devices. These predictive capabilities will prove invaluable in the design and development of solid-state batteries and other energy storage devices. The enhanced performance, safety, and reliability afforded by real-time monitoring and intervention not only contribute to the longevity of the devices but also pave the way for more sustainable and efficient energy storage solutions. For instance, a model accurately estimating Li-ion battery capacity was developed, correlating capacity fade with impedance increase.^[159] This involved impedance analysis at selected frequencies and individual battery calibration to determine their capacity-impedance relationship. Tested over 300 cycles on six Li-ion polymer batteries, the model showed low average RMSE and error, demonstrating its practicality for real-world applications with online impedance measurements. As ML models continue to advance, their role in shaping the future of energy storage technology becomes increasingly prominent, offering a transformative approach to optimizing system performance and ensuring the integration of solid-state batteries into diverse applications.

(2) Multi-Property Optimization: Achieving multi-property optimization represents a significant challenge in the realm of SSE, where a delicate balance among various factors is essential to meet specific application requirements. ML emerges as a potent tool in this endeavor, leveraging advanced techniques in multi-objective optimization. These algorithms are designed to navigate the SSE properties, considering trade-offs between critical factors such as ionic conductivity, mechanical stability,

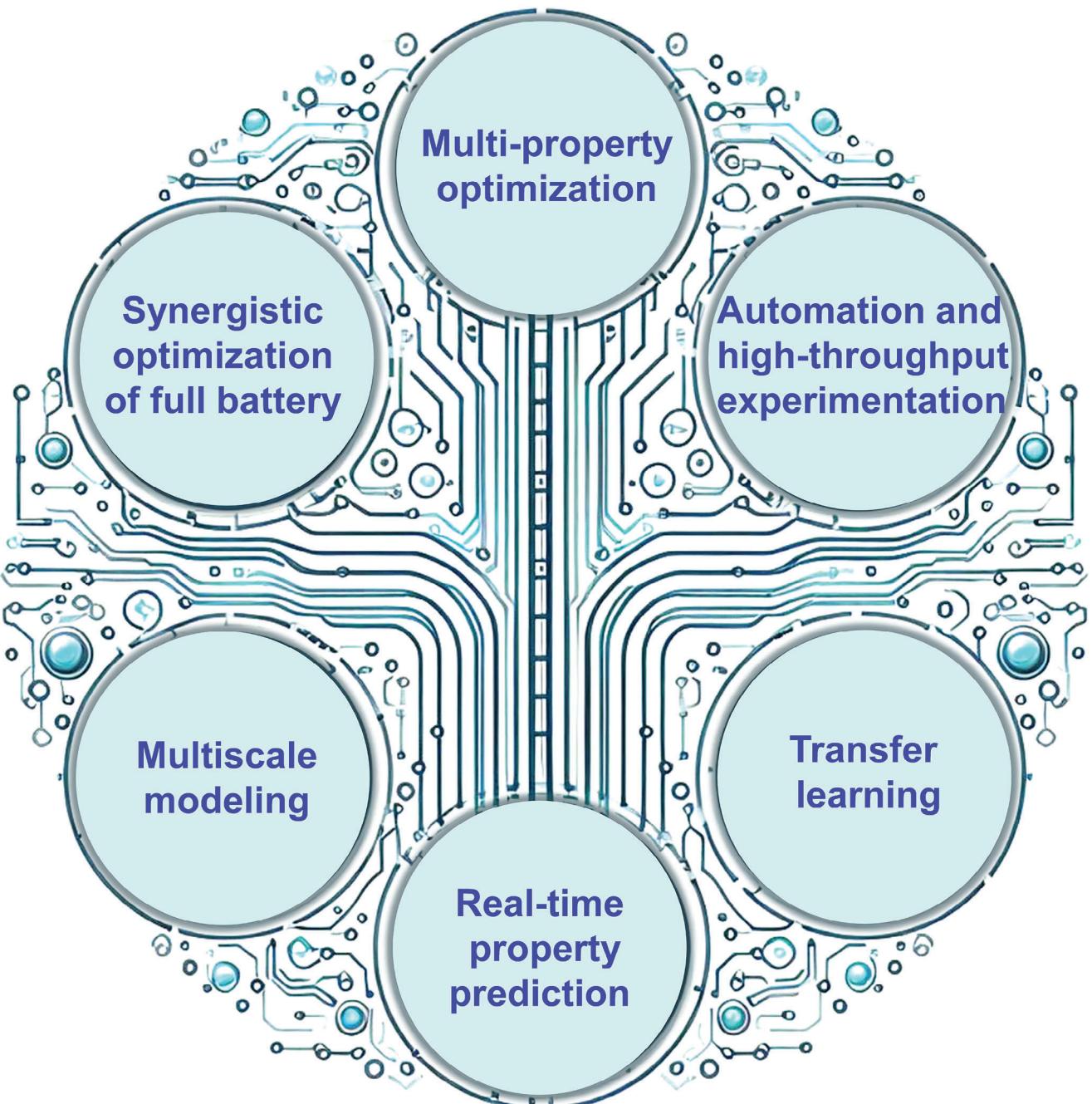


Figure 12. The study trends and future research directions of applying ML for SSEs.

thermal properties, and chemical compatibility. The implementation of multi-property optimization holds great promise for addressing the pressing need to enhance the overall performance of SSEs. By concurrently optimizing multiple properties, this approach strives to elevate the safety and cycle life of SSEs, laying the groundwork for their widespread application in high-performance energy storage devices. This holistic strategy becomes particularly crucial in surmounting the unique challenges associated with SSEs, propelling them toward practical viability in the dynamic landscape of advanced energy storage tech-

nologies. The nuanced interplay of properties, choreographed by multi-objective optimization powered by ML, not only simplifies the process of selecting SSEs but also paves the way for groundbreaking innovations in SSE design. The fine-tuning of various parameters ensures a tailored approach to addressing specific application needs, thereby broadening the scope for the development of SSEs with superior and well-balanced performance characteristics. The advancement of ML algorithms is significantly impacting the field of materials science, particularly in multi-property optimization. This innovative approach has been

applied to design multi-component Co-base superalloys with various targeted properties,^[160] paving the way for the development of next-generation SSEs.

(3) Multiscale Modeling: Multiscale modeling represents a pivotal approach in unraveling the complexities inherent in the interplay between atomic-scale intricacies and macroscopic behaviors. Navigating the complex structure and dynamics of SSEs across various length and time scales poses a formidable challenge, and the integration of ML into multiscale modeling methodologies is poised to revolutionize our comprehension of these complex systems. As technological advancements propel us toward the development of novel SSEs and applications, the synergy between ML and multiscale modeling becomes increasingly paramount. In the foreseeable future, ML is anticipated to play a progressively complementary role in enhancing the efficacy of multiscale modeling techniques. The amalgamation of these approaches is instrumental in not only deciphering the fundamental mechanisms governing SSEs but also in elucidating their transport properties and interfacial interactions. By seamlessly integrating data spanning the atomic to the macroscopic level, multiscale modeling not only provides a holistic perspective but also furnishes invaluable insights into the underlying processes that dictate SSE performance. As the fields of materials design and engineering progress, the in-depth knowledge gained from multiscale modeling, enhanced with ML, becomes essential. This integration offers a direct pathway to combine the precision of ab initio methods with the capability to investigate mechanical and failure responses of nanostructures at the continuum scale, effectively bridging these two realms.^[161] This methodological fusion empowers researchers and engineers to optimize SSEs for specific applications, tailoring their properties to meet the demands of diverse technological scenarios. In essence, the integration of multiscale modeling and ML stands out as a fundamental element in driving advancements in material science. It provides a methodical and effective strategy for deciphering the complex interplay between atomic-scale characteristics and macroscopic properties in SSEs.

(4) Transfer Learning: The versatility of SSE materials, ranging from oxides and sulfides to polymers, necessitates innovative approaches to comprehend and optimize their properties. Transfer learning, coupled with domain adaptation techniques, becomes a linchpin in achieving this transformation by facilitating the transfer of knowledge across material families. The significance of transfer learning lies in its ability to minimize the need for distinct models tailored to various material types. This reduction in model diversity translates into significant time and computational resource savings. The streamlined approach not only expedites research and development processes but also contributes to a more holistic understanding of electrolyte properties spanning a multitude of SSE materials. The efficiency gained through transfer learning thus becomes a cornerstone in advancing our comprehension of the complicated relationships between material composition and functional performance. Beyond its efficiency gains, transfer learning holds immense potential in addressing a critical challenge faced by SSEs: inadequate conductivity. Leveraging the knowledge and experience amassed from existing electrolyte materials, transfer learning accelerates the development and optimization of new materials. By applying insights garnered from

one material type to another, transfer learning serves as a catalyst in uncovering novel SSE compositions with significantly enhanced conductivity. This strategic use of transfer learning not only accelerates innovation but also positions it as a powerful tool in overcoming longstanding obstacles in achieving optimal conductivity within SSEs. For enhancing Li-ion conductivity in NASICON-type $\text{LiZr}_2(\text{PO}_4)_3$ electrolytes for solid-state batteries. Bayesian optimization, augmented by transfer learning from similar $\text{Li}_{1+x+2y}\text{Y}_x\text{Ca}_y\text{Zr}_{2-x-y}\text{P}_3\text{O}_{12}$ compositions, was employed to efficiently identify the optimal composition.^[162] This approach proved to be twice as efficient as traditional methods, indicating its potential for broader applications in material and ionic conductor optimization. Ultimately, the amalgamation of ML's flexibility and the strategic deployment of transfer learning methodologies propel SSE research into an era of unparalleled efficiency and innovation. This convergence holds the potential to usher in a promising future characterized by the development of advanced electrolyte materials boasting superior conductivity properties.

(5) Automation and High-Throughput Experimentation: The fusion of ML and laboratory automation stands at the forefront of transforming SSE research. Through the integration of ML algorithms and automated robotic systems, the pace of scientific discovery is set to reach unprecedented levels. These automated robotic systems will not only perform experiments but also conduct precise measurements, generating vast datasets at a speed that was once inconceivable through manual processes. This influx of data will form the backbone of a new era in SSE. High-throughput experimentation platforms will play a pivotal role in this transformative journey. These platforms, characterized by their ability to rapidly conduct numerous experiments simultaneously, will empower researchers to explore an expansive materials with efficiency and thoroughness. The accelerated pace of experimentation will allow scientists to delve into uncharted territories, uncovering novel materials and properties that could have remained elusive in a traditional, slower-paced research environment. The datasets generated by these high-throughput platforms will not only serve as a reservoir of raw information but will also become the training ground for advanced ML models. Through iterative learning and pattern recognition, ML algorithms will swiftly analyze and extract meaningful insights from the complex datasets, enabling the identification of promising materials with unparalleled precision. For example, an automated Thin-Layer Chromatography (TLC) system for high-throughput was reported.^[163] ML models were developed to link organic compound structures with polarity, indicated by retardation factor. These models accurately predicted retardation factor curves in different solvents, guiding purification processes and streamlining TLC data analysis. The symbiotic relationship between automation and ML will significantly expedite the entire SSE discovery process—from initial exploration and identification to synthesis and subsequent validation. As a result of this integration, the timeline for SSEs from lab-scale research to real-world applications will be drastically shortened. The accelerated discovery and validation of SSEs with desirable properties will pave the way for innovative technologies and applications across various industries, ranging from energy storage to electronics. The combination of automation and ML is not just an evolution in research methodology; it is a revolution that will reshape the field of materials

science, fostering a new era of efficiency, precision, and rapid advancement.

(6) Synergistic Optimization of Full Battery: The synergistic optimization of full battery functionality is intricately tied to the pivotal role that SSE plays in advancing the technology of full solid-state batteries. As these batteries emerge as key players in meeting the escalating demand for high-energy-density and safe energy storage solutions, the intrinsic properties of solid-state batteries assume heightened significance. The application of ML techniques in the design and development of SSE becomes a strategic imperative, demanding a nuanced consideration of the distinctive characteristics and requirements of solid-state battery technology. In this interplay between ML and SSE, the focus extends to the meticulous optimization of SSE properties. ML algorithms, when applied judiciously, prove instrumental in fine-tuning critical aspects such as ionic conductivity, mechanical stability, and compatibility with electrode materials. By navigating the sophisticated scope of material properties and design parameters, ML facilitates the crafting of SSE materials that not only meet but exceed the stringent demands of solid-state battery performance. This optimization, guided by ML-driven insights, contributes significantly to elevating the overall efficiency and reliability of solid-state batteries. For instance, a neural network potential was employed to predict both conductivity and interfacial stability in all-solid-state Li-metal batteries.^[164] The optimization resulted in batteries exhibiting low cathode/SSE internal resistance, superior average coulombic efficiency, impressive rate performance, and exceptional cycling stability. The integration of ML into the design process signifies a paradigm shift, enabling a more streamlined and efficient exploration of the expansive design space inherent to SSE materials. This, in turn, expedites the discovery and development of advanced SSE materials precisely tailored to the specific and evolving needs of solid-state battery technology. The integration of ML methodologies into SSE research not only expedites the innovation process but also guarantees a harmonious alignment of resultant materials with the ever-evolving terrain of energy storage. This synergy propels solid-state batteries to the vanguard of cutting-edge solutions for next-generation energy needs.

In conclusion, the future of ML in predicting SSE properties holds immense promise for the progress of energy storage technologies. These developments will not only drive innovation in materials science but also contribute significantly to the global transition toward sustainable and clean energy solutions.

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Conflict of Interest

The authors declare no conflict of interest.

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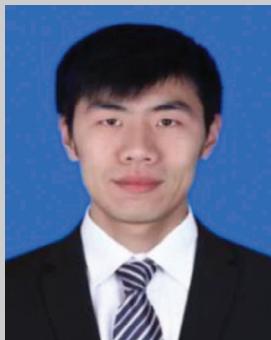
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