

AI-Driven Rational Design of Solid-State Electrolytes

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How To Cite: Jan, Z.; He, J.; Guo, H.; et al. AI-Driven Rational Design of Solid-State Electrolytes. *AI for Materials* **2026**, *1*(1), 12. <https://doi.org/10.53941/aimat.2026.100012>

Received: 13 May 2026

Revised: 15 July 2026

Accepted: 15 July 2026

Published: 5 August 2026

Abstract: The solid-state electrolytes (SSE) are gaining tremendous attention in designing rechargeable batteries with remarkable energy density and safety features for next-generation energy storage device applications. The rational design of SSE with promising ionic conductivity, higher electrochemical stability windows, and stable electrode-electrolyte interfaces remain a formidable challenge, traditionally hindered by trial-and-error experimentation and computationally expensive theoretical simulations. Here, we systematically review the recent breakthroughs in the artificial intelligence (AI)-driven design of SSE, spanning electrochemical stability and ionic conductivity domains, with a particular focus on how machine learning (ML) and deep learning (DL) are fundamentally transforming the discovery and optimization landscape. We critically discuss the synergy between first-principles density functional theory (DFT), molecular dynamics (MD) simulations, and advanced AI algorithms including supervised and unsupervised learning (SL, UL), graph neural networks (GNNs), and Machine Learning Interatomic Potentials (MLIP) that collectively enable accurate prediction of ionic conductivity, elucidation of ion transport mechanisms, and high-throughput screening (HTS) of vast chemical spaces. Emphasis is placed on descriptor engineering that bridges atomic-level structural features (e.g., lattice parameters, activation energies, defect chemistry) with macroscopic electrochemical performance, as well as the emerging paradigm of closed-loop, self-driving laboratories for autonomous materials discovery. Furthermore, AI-guided strategies have demonstrated remarkable interfacial ionic transport mechanism. Despite these transformative advances, persistent challenges including data scarcity, limited descriptor transferability, discrepancies between theoretical predictions and experimental realization, remain significant challenges. Looking forward, the convergence of AI with high-throughput experimentation and multiscale modeling promises to redefine SSE discovery, accelerating the deployment of high-performance all solid-state batteries (ASSBs) for sustainable energy storage.

Keywords: solid-state electrolytes; artificial intelligence; DFT approach; high-throughput screening; all solid-state batteries



1. Introduction

The global demand of energy has strengthened the search of clean, cheap and ascendable energy storage technologies [1–3]. Especially, electric vehicles (EVs) bypass fossil fuels, are manufactured on large scales, ever-increasing the demand for designing novel batteries [4,5]. Amongst the existing opportunities, rechargeable batteries are emerging as a potential solution, leveraging reversible electrochemical reactions to significantly store and release electrical energy [6–8]. The rechargeable batteries with accessibility to utilize for various applications, highlights their potential of charge storage for both short and long terms power requirements [9–11]. The main components of rechargeable batteries comprise the cathode, anode separator, electrolyte, and current accumulator. Among them, electrolytes are scalable, crucial to identify the battery performance and life cycle [12,13] offering opportunities for engineering AI based (SSE). As an ideal SSE should instantaneously possess higher ionic conductivity ($>10^{-3}$ S/cm at normal temperature) [14], negligible electronic conductivity, exhibit electrochemical stability profile >4 V [15], and stable electrode-electrolyte interfaces [16,17]. To achieve such a broad electrochemical stability, high ionic conductivity, ionic outflow, and durable solid-solid interfaces are still challenging [18,19]. To address these linked material and performance concerns, the rapid growth of scientific discovery has become an essential foundation of advancement.

The development of SSE, rely on trial-and-error synthesis and requires experimental validation, is both time-consuming and resource-intensive [20]. The integration of well-established computational approaches are reliable strategies to predict ionic conductivity and interrelated transport mechanisms [21–24]. Subsequently, ML approaches have leveraged several database platforms including Material Projects (MP) and Inorganic Crystal Structure Database (ICSD), further allowed to describe the large-scale data accumulation. They facilitate prediction of ionic conductivity, stability profile, and lattice dynamics, and thousands of candidates in a fraction of the time and processing rapid virtual screening of candidate's materials [25–27]. Nevertheless, these approaches still prior extrapolative, demonstrate connections and vigorous materials, but cannot separately design synthesis tactics or adaptively guide experiments. Recent developments are therefore shifting from predictive models to independent AI proficiency of reasoning, planning, and customize within closed discovery loops, thus boosting the route from design to real world applications. Zheng et al. and Dutra et al., have primarily focused on atomistic modelling, machine learning methodologies, and the fundamental understanding of ion transport, chemical stability, and interfacial phenomena in SSE, the present review specifically emphasizes the role of AI in enabling the rational design of SSE. In particular, AI is evolving surpassing predictive modelling towards effective materials discovery in SSE. For example, generative AI outlines incorporating generative adversarial networks and Bayesian optimization have been successfully employed to examine the SSE chemical space and recognize significant electrolyte materials with improved properties [28]. In correspondence, multiscale modelling and characterization techniques have offered potential understanding into ion transport mechanisms and interfacial processes across multiple length and time scales, thereby developing stronger structure-property relationships in SSE [29].

Commercially, EVs utilize lithium-ion batteries (LIBs) featured with liquid electrolytes, possess high ionic conductivity. However, the volatile nature, toxicity and flammability of LIBs leads to limiting their long-term reliability due to severe safety hazards [30,31]. To mitigate these risks, SSE have emerged as a potential alternative to fulfill the real-world demands for next-generation EVs and broad-scale energy storage systems, owing to their non-flammable and robust mechanical performance [32–34]. In general, SSE are classified into various categories such as polymer, inorganic (halides, oxides, sulfides) [35–37], and organic-inorganic composite types [38]. Among these, sulfide-based SSE possess room-temperature ionic conductivity (>10 S/cm), are particularly promising. However, their practical realization hindered by low interfacial stability against cathode materials, high moisture sensitivity, and considerable chemical reactivity [39,40]. To overcome these challenges, several approaches including doping optimization [41,42], interfacial engineering [43], and development of three-dimensional conductive networks [44] have been widely reported. Despite these, the commercial production of SSE demands simultaneous improvement of ionic conductivity [45], controlling interfacial side reactions [46], and fixing the large changes in electrode volume occurred during cycling [47].

To deal with these major concerns, the computational materials science has accelerated the emergence of rational design of next-generation SSE. The physical significance of the descriptors used in the ML framework. Particularly, descriptors correlated to lattice volume, bottleneck size, Li-ion migration pathways, electronegativity differences, and anion/cation chemical environments were selected because they directly influence the activation energy for ion transport and the thermodynamic stability of the solid electrolyte [48]. Previous researches have revealed that larger diffusion channels and beneficial local coordination environments can be reduce migration barriers and improve ionic conductivity, while descriptors related with bond strength, electronegativity, and formation energy relate with chemical and electrochemical stability against electrode and decomposition reactions

[49,50]. In addition, we suggest that the AI model does not simply offer empirical predictions but comprehends structure-property relationships training with these physically profound descriptors. By evaluating feature significance and descriptors trends, the model ensure which structural and chemical properties strongly effect ion transport and stability, thus linking data-driven predictions along with mechanistic understandings [48]. This elucidation improves rational design strategy by enabling specific modification of crystal chemistry and local environments to achieve enhanced electrolyte performance.

For instance, DFT explores the atomic-level insights into structural stability, understanding of electronic characteristics and ionic transport pathways [51]. In parallel, ab initio molecular dynamics (AIMD) simulations precisely evaluate the temperature-dependent ionic diffusion and thermal stability range [52–54]. Despite ensuring their reliability, these conventional quantum mechanical approaches hinder their applications in performing high-throughput screening of massive material databases due to computational cost and large time-consumption. Recently, AI including both ML and deep DL, has successfully circumvent limitations of designing novel SSE. The combined advanced data-driven AI models trained on high-quality DFT/MD datasets provides rapid predictions of physical properties of materials and screening potential SSE candidates with enhanced performance [55–57].

With these inspirations, the present review comprehensively addresses the key role of DFT computations coupled with AI approaches in rational design of SSEs with enhanced performances. Here, we briefly present the computational approaches employed in exploring SSE, assessing their current applications and limitations. Next, we outline how AI technologies effectively bridge these computational gaps, highlighting recent breakthroughs achieved via ML and DL models. Finally, we discuss the emerging paradigms by combining DFT calculations with AI workflows, highlighting the key challenges and future perspectives of adopting interdisciplinary approach to accelerate the practical deployment of ASSBs.

2. Theoretical Calculation Approaches

Recent advancement in computational material science has enabled the development of outstanding theoretical frameworks for exploring SSE materials [27,58]. In materials science, the conventional experimental techniques extensively demand physical resources and manpower to synthesize and scrutinize SSE. However, these techniques are comparatively time consuming and expensive with repeating cycles. Generally, the experimental approaches involve human resources with availability of materials for synthesis, designing and characterization of SSE, including considerable expenses and reasonable time consumption in iterative cycles. The techniques are significantly important in exploring the materials interaction and directly accessible to understand the atomic level mechanism. Nevertheless, incorporating more advance first principles studies and material simulations have further provided deep insights into structural and ion transport properties at atomic level. Prior to experimental validations, theoretical findings endorse the stability and HTS of proposed material, boost the materials discovery and novelty of SSEs with enhanced efficiency [59–61].

Furthermore, the fundamental theoretical calculations approaches are the DFT and MD calculations [62–64]. These approaches are distinct; (i) DFT calculations deep-rooted in quantum mechanical principles, ground state properties, and electronic structural features, (ii) MD calculations compute laws of atomic motion, clarifying the dynamic evolution routes of materials. The integration of these approaches establishes enrich theoretic guidance for the designing and modification of SSEs materials [65–68].

2.1. DFT and MD Approaches

The ground state geometric optimization, analysis of single-point energy and exploration of transition-state can be significantly introduced through DFT calculations [69,70]. For instance, geometrical optimization governs the stability window, the electronic properties such as band nature and charge mobility evaluated by single-point energy calculations; while the ionic diffusion with considerable reaction pathways is described by Climbing Image Nudged Elastic Band (CI-NEB) [71,72]. Additionally, phonon calculations examine the dynamic stability, which elaborate the thermal expansion and consistent heat capacity of SSEs materials [73,74]. These computational approaches, although effective, faces resources intensive and pose challenge for complex systems. In this case, ML approaches have the potential to address these challenges by facilitating data analysis, accelerating the screening of candidate materials, and developing the prediction of material properties. However, it is paramount to differentiate the corresponding contribution of NEB and AIMD in terms of ion transport in SSE. The NEB approach is employed to regulate minimum-energy migration routes and respective activation energy barriers, thereby providing promising insights into the kinetics of ion migration [75]. In contrast, AIMD simulations explicitly account for finite-temperature atomic motion and enable the direct evaluation of ion diffusion mechanisms, transport pathways, and ionic conductivity under realistic thermodynamic conditions [76]. Therefore, NEB and AIMD should

be regarded as complementary approaches, where NEB elucidates migration kinetics, while AIMD offers a comprehensive description of temperature-dependent diffusion behavior and collective ion transport phenomena in SSE.

MD is also the theoretical approach tracking microscopic routes, describing Newton's laws of motion which leads the atomic motion. This approach enables to explore the core characteristics of ionic conductivity and thermodynamic performance of SSE materials [68,77,78]. Commonly, MD comprehend classical MD, AIMD, and deep learning-driven molecular dynamics (DeepMD) [79]. Owing to computational scalability, classical MD is highly efficient for complex systems and long-term simulations. In contrast, quantum mechanical based AIMD calculations substantial for exploring complex inter-atomic electronic interactions in real time and recognition of chemical bonds. Bridging the gap between these two approaches, DeepMD uses neural networks (NNs) approach to learn potential energy surfaces from *ab initio* generated dataset, remarkably achieve classical-level computational efficiency with high quantum-level accuracy.

The selection of a suitable thermodynamic ensemble is crucial to examine ion transport mechanism of SSE. The generally implemented canonical (NVT) and isothermal-isobaric (NPT) ensembles are employed to extract the mean square displacement (MSD) of mobile ions from trajectories and reproduce operational battery environments. Subsequently, researchers computed ionic diffusion coefficients and ionic conductivity using Nernst-Einstein equation [80–82]. Moreover, evaluating the probability density distribution of migrating ions allows for the direct visualization of conduction pathways and transport mechanisms within the crystal lattice. Ultimately, multiscale computational paradigms, ranging from DFT to various MD regime, deepen the mechanistic understanding of SSE, offering a robust theoretical framework to guide materials engineering.

2.1.1. Structural Stability Analysis

The progress of advanced ASSBs is gaining tremendous attention relying on the optimization and development of SSE due to high ionic conductivity and structural stability. This discovery process further accelerated by employing theoretical methodologies. In particular, first principles study become as indispensable approach for predicting crystal structures, thermodynamic stability and electrochemical performance of promising SSE [83,84]. Generally, the halide-based SSE have shown suppressed ionic conductivity than oxide and sulfides [34,85]. However, the recent advancement in experimental and computational tools have exceptionally bridged the gap. For instance, Park et al. systematically investigated twelve Li_3MCl_6 SSE, possess large electrochemically stable window and chemical stable in both ambient humidity and against cathode materials [86]. Further, researchers have widely studied lithium ytterbium chloride (Li_3MCl_6) to understand the influence of structural stability on Li^+ migration pathways [87,88]. Li_3MCl_6 exhibits both trigonal and orthorhombic phases with electrochemical windows, as displayed in Figure 1a,b. Specifically, the orthorhombic phase possesses higher ionic conductivity coupled with superior chemical stability. Moreover, the class of halide SSE has further expanded with designing of more complex structures. Several halide systems exhibit competitive room-temperature ionic conductivity and excellent electrochemical stability. The compositionally complex $(\text{Li}, \text{Sr})(\text{Ta}, \text{Nb}, \text{Zr}, \text{Hf})\text{O}_3$ - (LSTNZH) material have enhanced performance with exceptional ionic conductivity owing to modulation of grain boundaries [89]. The results are interesting to correlate the geometry and compositional complexity in designing storage devices with enhanced proficiency.

Furthermore, compositional doping is another important strategy to optimize both interfacial stability and ionic conductivity of SSE [90–92]. By employing DFT and AIMD, the halogen doping specifically fluorine substitution in anti-perovskite ($\text{Li}_2\text{OHCl}_{0.9}\text{F}_{0.1}$) [93] SSE, has tailored the crystal framework of Li_2OHCl , as illustrated in Figure 1c,d. The fluorine partially substitutes the Cl^- atoms at A-sites introduces lattice defects, resulting in localized disorder within the lattice and phase transition from orthorhombic to cubic phase. This facilitates lowering the activation energy barrier, accelerating the Li^+ transportation across highly coordinated migration pathways the $\text{Li}_2\text{OHCl}_{0.9}\text{F}_{0.1}$ SSE. In addition, incorporating elemental dopants like Ge, Sn, and ZnO in sulfide-based electrolytes, such as $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$ (LGPS), have enhanced their chemical stability against humidity without altering the ionic conductivity [94,95]. Theoretical modeling of dopant interactions like surface substitution with Selenium (Se) in the LGPS framework, has been instrumental to gain insight into curtail moisture adsorption, hindering the release of toxic H_2S gas [21,96,97].

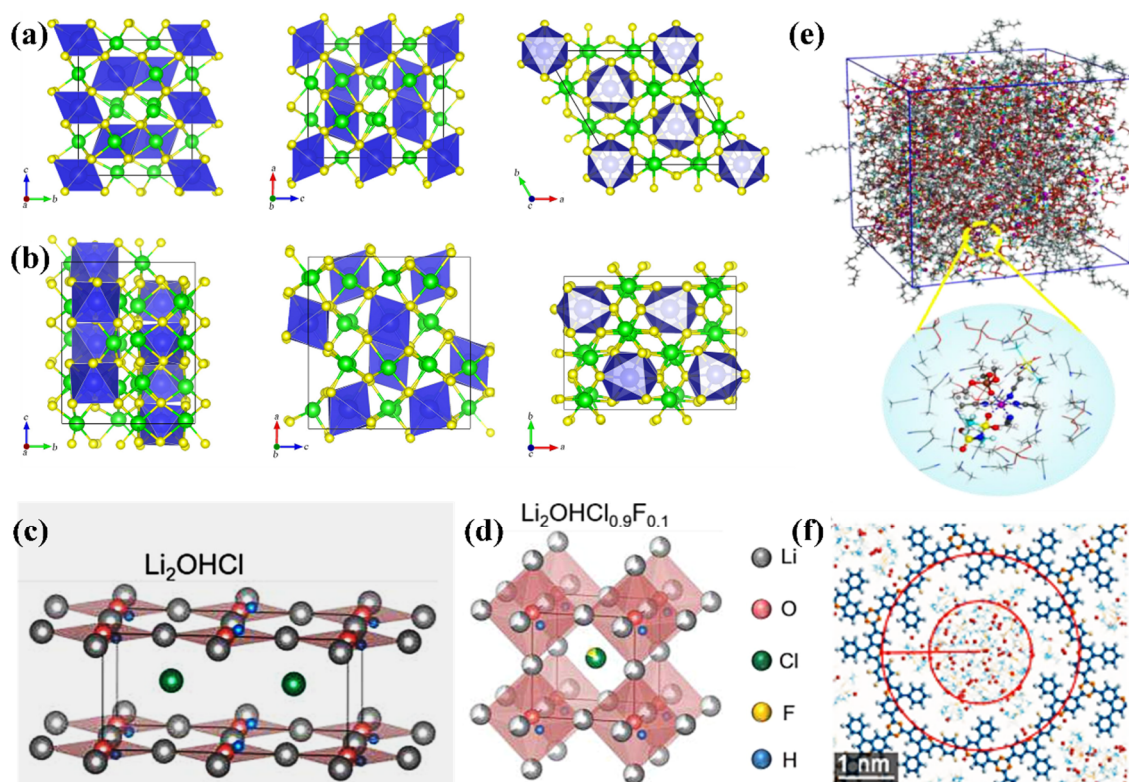


Figure 1. Crystal structures of $1 \times 1 \times 2$ supercells of (a) Trigonal phase (b) Orthorhombic phase of Li_3YbCl_6 . Reproduced with permission [87]. Copyright 2024, Elsevier. Crystal structure of (c) orthorhombic phase antiperovskite (d) cubic phase antiperovskite (where grey, red, green, yellow, and blue represents the Li, O, Cl, F, and H respectively); Noted, A-sites of corresponding antiperovskite structure are occupied by partially fluorinated Cl^- ions. Reproduced with permission [93]. Copyright 2022, Elsevier. (e) Snapshot based on molecular dynamics (MD) analysis of SSE-trimethyl phosphate electrolyte. Reproduced with permission [98]. Copyright 2023, Elsevier. (f) The geometrically optimized LiTFSI in Q-COF channels with radial distance 0.86 nm between the outer boundary (red circle) and channel center. Reproduced with permission [99]. Copyright 2021, Wiley-VCH GmbH.

In polymer and composite systems, it is essential to understand the correlation between structural optimization and nanoscale Li^+ transportation at electrode-electrolyte interface [100]. MD simulations were performed to investigate the geometrical stability of trimethyl phosphate (TMP) anchored polymer. It is obvious from Figure 1e that TMP actively promotes Li^+ solvation shell formation encapsulated by polymers and responsible for robust mechanical skeleton. This configuration leads to improving Li^+ diffusion and structural regulation, ultimately enhancing the long-term reversible cycling stability as essentially required for energy storage devices. Recently, Covalent Organic Frameworks (COFs) porous materials have extensively demonstrated in designing SSE due to their strong ionic conduction capability [101,102]. Theoretical studies (DFT coupled with MD simulations) have been mapped to explore preferred pathways of Li^+ along 1D pore walls of COFs, as shown in Figure 1f. These findings suggest that Li^+ ions preferentially localized near nitrogen (N) and fluorine (F) atoms within the COFs at 8.5 Å proximity, emphasizing the chemical affinity between COFs host and the Li^+ ions, ultimately enhancing Li^+ transport. The structural analysis and computational insights provide platforms to evaluate the interfacial stability and electrochemical performance for development of novel SSE.

2.1.2. Electronic Structure Analysis

Electronic structure analysis plays a key role in exploring the fundamental characteristics of SSE materials and their electrochemical performance. For thus, the theoretical approaches elaborate the band structure, density of state (DOS), and charge transformation analysis, provide thoughtful understandings into the electronic properties of materials and their relationships with ionic conductivity.

To analyzing the band structure of SSE materials, the $\text{NaTi}_2(\text{PO}_4)_3$ (NTP) complex performs as a demonstrative analysis [103]. DFT analysis demonstrates that the 2p states of oxygen and 3d state of titanium have major contributions in valence and conduction bands of NTP system respectively, exhibiting considerable bandgap

of (2.25 eV) shown in Figure 2a,b. These findings indicate NTP exhibit as an electronic insulator, with its narrow electronic conductivity highlighting as a substantial characteristic of SSE materials.

Furthermore, DOS provides deeper understanding into the electronic structure of materials. For example, in list of $\text{Li}_7\text{P}_3\text{S}_{11-x}\text{O}_x$ materials [104], alterations in O-doping concentration facilitates to prominent variations in DOS distributions. Particularly, in $\text{Li}_7\text{P}_3\text{S}_{10.25}\text{O}_{0.75}$, the introduction of O-atoms not only curial in the distribution of electronic state at the valence band maxima but also changes the Li^+ ion transport platform by regulating local electronic configuration. Moreover, the decreased ionic migrations barrier of (0.20 eV) mentioned in Figure 2c,d. Additionally, electrode-electrolyte interfaces are described with the help of molecular orbital energy levels and work function calculations which are essential for details analysis. In the examination of cellulose

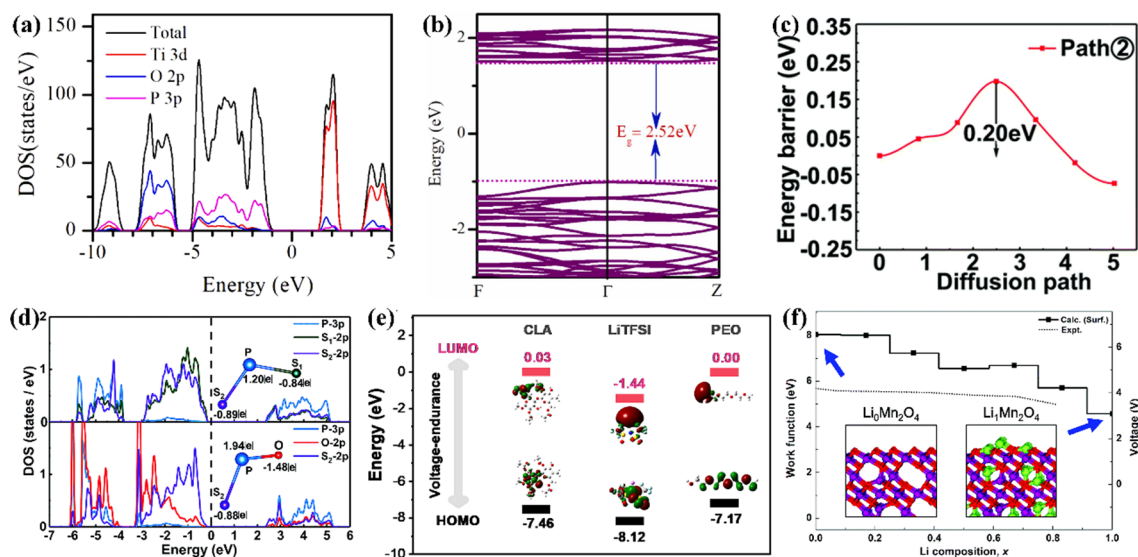


Figure 2. (a) Calculated total and partial density of states (TDOS, PDOS) of $\text{NaTi}_2(\text{PO}_4)_3$. (b) For band gap analysis, the electronic band structure is presented along high symmetry path. Reproduced with permission [103]. Copyright 2024, Elsevier Ltd. (c) Diffusion of ion against energy barriers of $\text{Li}_7\text{P}_3\text{S}_{10.25}\text{O}_{0.75}$. (d) The PDOS and Bader charges analysis for the $\text{S}_2\text{-P-S}_2$ and O-P-S_2 clusters, accordingly. Reproduced with permission [104]. Copyright 2019, the Owner Societies. (e) Comparisons of corresponding energy levels (HOMO and LUMO) with other traditional polymers. Reproduced with permission [105]. Copyright 2023, Wiley-VCH GmbH. (f) Calculated work function and potential vs. Li^+/Li across LMO surface attributed to Li^+/Li electrode. Reproduced with permission [106]. Copyright 2018, the Owner Societies.

Acetate (CLA)-containing polymer electrolytes, theoretical study demonstrate that the promoted (LUMO) at (−1.86 eV) energy level of the CLA environment suggests beneficial compatibility with Li-metal anodes illustrated in Figure 2e. The work function evaluation is also crucial for electronic structure exploration. Han et al. investigated the work function of LiMn_2O_4 cathode surfaces (with Li-ion concentration from $x = 0$ to 0.8 range) using $(3 \times 3 \times 1)$ k-point mesh integrated with the Hubbard U correction ($U = 3.9$ eV) [106]. Notably, a considerable shift into work function was identified at the electrode-electrolyte interface, see Figure 2f. The theoretical findings have demonstrated the basic physical properties of SSE, intuitively highlights the significance of atomic simulations in understanding the complex materials at atomic level.

2.1.3. Ion Transport Analysis

For SSE materials, theoretical approaches are counted as a promising tool for elaborating ion migration mechanisms, especially diffusion and conductivity performance. Designing this electrolyte with high ionic transport properties is still a challenge. For this, Li-ion transport mechanisms understanding are essential for designing new SSE materials. Encouragingly, charging and discharging process in Li-ion batteries involves the coupled transport of Li^+ ions through the electrolytes and electrons through the external circuit between the cathode and anode. This transport phenomena are driven by chemical potential gradients developed by interfacial electrochemical reactions, which cause distinct concentrations and thereby stimulate directional Li^+ diffusion and migration across the electrolyte-electrode interfaces [107–110].

The understanding of Li ion transport phenomena at microscopic level via DFT study is crucial to design novel SSE. The energy barriers, identification of possible migration pathways and analysis of intrinsic ionic

conductivity are key performance metrics demonstrated by DFT calculations. Further, transition state theory (TST) plays a primary role in modeling Li ion movement from initial to final site. This event requires specific activation energy barrier at transition state. In general, the ions favorably transport along path with minimum energy. The NEB method, extensively adopted for choosing possible ion transport path and critically determine the saddle points found in electrochemical reaction, used to demonstrate Li-ion diffusion in SSE [111,112]. To enhance the accuracy of transition-state determination, the CI-NEB approach was elaborated, in which the highest-energy image is driven toward the accurate saddle point along the migration pathway [113]. Allowing to its accuracy in tracing activation barriers, CI-NEB become a sophisticated approach for demonstrating ion transport and diffusion kinetics in SSE. Furthermore, another powerful tool for ionic migration interpretation is AIMD simulation, which offers atomistic understanding into ionic conductivity and offer guidance for the rational design of novel SSE materials (Figure 3).

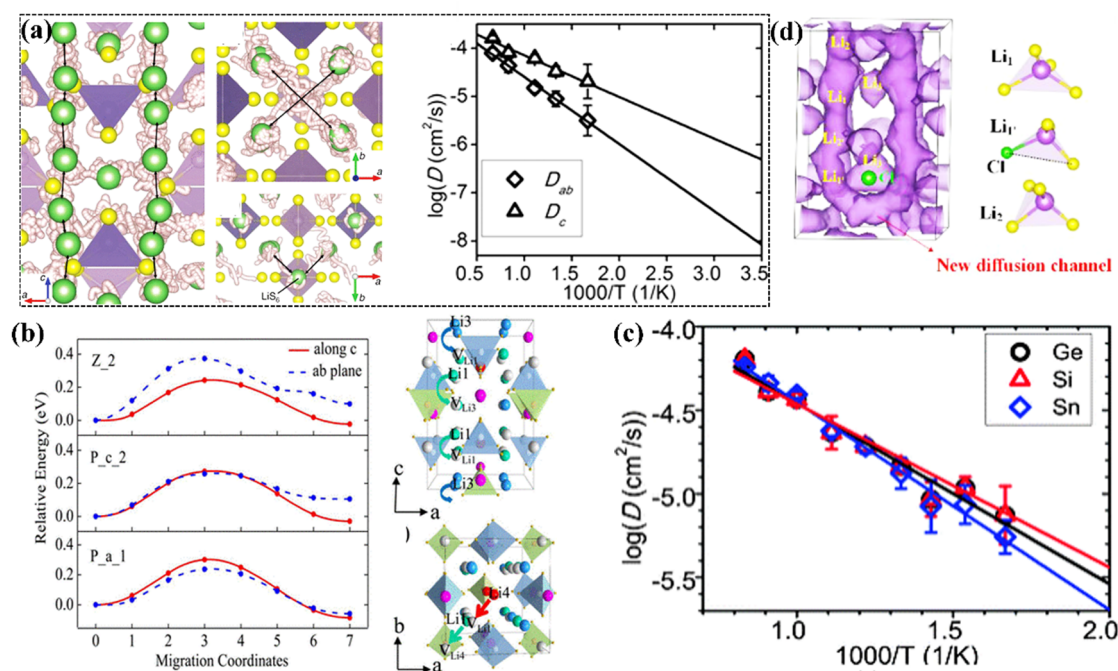


Figure 3. (a) Li-ion diffusion route in the LGPS involved of 1D, 2D diffusion and Arrhenius plots D_c , D_{ab} along c-axis and in ab-plane respectively. Reproduced with permission [114]. Copyright 2011, American Chemical Society. (b) Potential energy curves and Li ion migration pathway along the c-axis and in the ab planes for distinct structures. Reproduced with permission [115]. Copyright 2014, American Chemical Society. (c) Impact of isovalent (Si^{4+} and Sn^{4+}) cation substitution on the diffusion phenomena in the LGPS. Reproduced with permission [48]. Copyright 2013, Royal Society of Chemistry. (d) Computed AIMD simulation trajectories densities of Li ions in the Cl-doped electrolytes. Reproduced with permission [116]. Copyright 2019, Elsevier.

AIMD simulation are highly efficient in predicting the Li-ion diffusion phenomena in SSE [114]. As SSE materials are classified into various categories based on their intrinsic characteristics. It is illustrated in Figure 3a that LGPS structure can adopt 3D Li-ion transport supported by rapid 1D migration pathways along c-axis due to low activation energy and structural defects responsible for slower diffusion occur in ab-plane, maintaining the macroscopic ionic conductivity [117,118]. The migration barriers are largely influenced by microstructural modifications. For instance, microstructural alteration leads to aligning the migration barrier of ab-plane in parallel with along c-plane, see Figure 3b. DFT provides a systematic framework for understanding microstructural engineering, offering in-depth insights into the effects of elemental doping and substitution on ionic conductivity. In LGPS, theoretical findings reveal that substitution of Ge with Si or Sn results in high ionic conductivity, as displayed in Figure 3c. These findings are further confirmed by experimental validations [48,119,120]. Additionally, AIMD simulations help to visualize the Cl-doping responsible for new diffusion networks in $\text{Li}_{10}\text{SiP}_2\text{S}_{12}$ and analyzing the boosting performance, see Figure 3d. Afterwards, the extensive AIMD simulations studies were expanded to other sulfides including Li_3PS_4 , $\text{Li}_7\text{P}_3\text{S}_{11}$ and $\text{Li}_6\text{PS}_5\text{X}$ ($\text{X} = \text{Cl}, \text{Br}, \text{and I}$). Among these, Li_3PS_4 were found in three distinct structural phases (labelled as α , β , and γ - Li_3PS_4) with mutual conversion found at different temperature, result in indispensable transport properties [121–124]. It is obvious from Figure 4a, the direct-hopping of Li^+ along (001) was the most energetically favorable ion transport pathway in γ -phase, whereas the Knock-off diffusion of Li^+ along (010) had the lowest migration barrier in β -phase. Finally, CI-NEB calculations used to

demonstrate the relationship between structural geometry and barrier energy in $\text{Li}_7\text{P}_3\text{S}_{11}$ [125,126]. This highlights structural defects by Cl or F substitution and offers unique approach to optimize ionic conductivity. The reduced energy barrier for Li ionic migration on both short and long diffusion length scales are displayed in Figure 4b, resulting that the Cl and Br-replaced $\text{Li}_6\text{PS}_5\text{Cl}/\text{Li}_6\text{PS}_5\text{Br}$, such as $\text{Li}_{5.5}\text{PS}_{4.5}\text{Cl}_{1.5}$ and $\text{Li}_{5.5}\text{PS}_{4.5}\text{Br}_{1.5}$, could show enhanced Li-ion transport properties.

The ionic conductivity has closed connections with chemical configurations and microstructure of the materials, depending on the interactive variations of these consistent physical properties. Theoretical approach elucidated fundamental structure-composition-property relationships of the SSE, which are intricate to claimed via conventional experiments. NEB can demonstrate Li ion diffusion along different pathways however, AIMD simulation assists in calculating thermal structural stability but unable predict the migration path crucial for transport properties of complex configurations and establishment of novel SSE materials.

3. Artificial Intelligence Approaches

Basically, SSE materials research work has frequently depended on expensive and laborious experimental techniques. In contrast, theoretical approaches successfully incorporate experimental shortcomings. However, computational limitations persist in dealing with complex or massive materials composition. To circumvent these challenges, the AI tools relying on existing data (both experimental or DFT reported) has open up new avenue to explore significantly improved novel SSE materials [127,128].

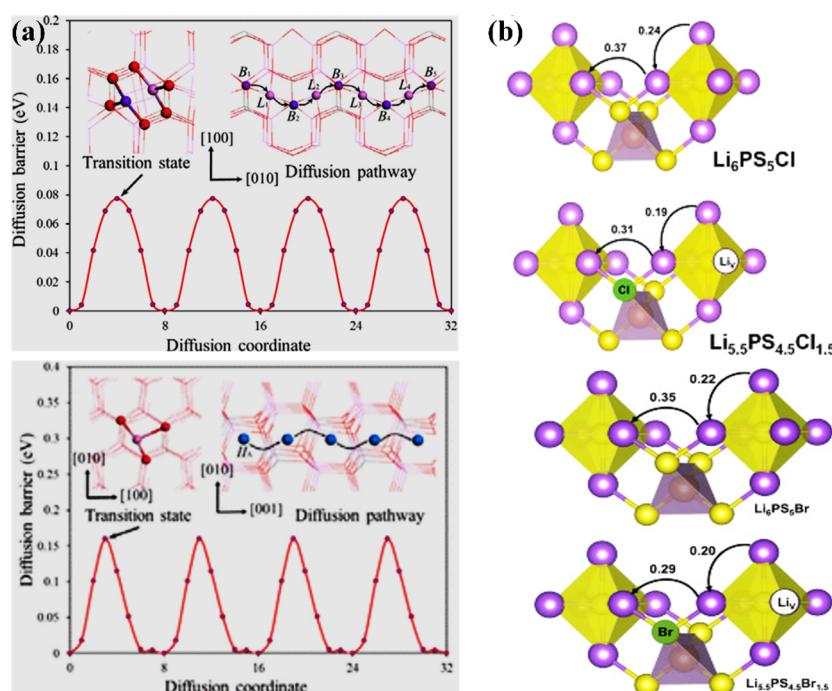


Figure 4. (a) Energy diagram, transition state structure, and migration route in the γ - and β - Li_3PS_4 . The direct hopping along (001) in the γ -phase had a diffusion barrier of 0.16 eV, whereas the knock-off diffusion along (010) in the β -phase had a diffusion barrier of 0.08 eV. Reproduced with permission [129]. Copyright 2016, American Chemical Society. (b) Energy barrier of Li ion migration in the $\text{Li}_6\text{PS}_5\text{X}$ and $\text{Li}_{5.5}\text{PS}_{4.5}\text{X}_{1.5}$ (X = Cl and Br). Reproduced with permission [130]. Copyright 2020, Elsevier. Reproduced with permission [131]. Copyright 2020, Elsevier.

AI approaches can be categorized into two classes based on their learning techniques; (i) ML, and (ii) DL. ML uses statistical algorithms to correlate structure and properties; DL extracts complex features and identifies different patterns using multi-layer NNs. These learning techniques offer new framework for designing more advance SSE materials with high energy storage capabilities.

Furthermore, the systematic summary of the recently developing AI landscape in SSE research are summarized in Table 1, including the key AI approaches, their applications, strengths, limitations, and representative studies significant to AI-driven novel design of SSE. They explore the development of AI approaches from traditional ML to graph-based learning, generative models, and closed-loop autonomous frameworks is progressively transforming SSE advancement from empirical trial-and-error approaches toward data-driven novel design and stimulated materials discovery.

3.1. Machine Learning Approach

ML approach can be divided into supervised, unsupervised, semi-supervised, and reinforcement learning respectively, based on distinct learning process [132–134]. In the domain of SSE research, first two approaches (SL and UL) are recently counted as the most beneficial approaches [132,135–137]. Although still at an early stage in SSE research, semi-supervised learning and reinforcement learning are emerging as promising approaches for accelerating materials discovery. Active-learning approaches revealed outstanding roles in generating high-quality training datasets and MLIP by iteratively choosing the most informative atomic configurations for first-principles calculations [138–140]. In parallel, RL and autonomous closed-loop frameworks are mostly being explored for materials optimization, facilitating the automated investigation of deepen structural design spaces while reducing experimental and computational expenses [141]. The improvements in AI tools are remarkably crucial for rational design of next-generation SSE. The core physical properties such as stability, mechanical strength and ionic conductivity presented in Table 1, indicating ML behaves differently from DL approach in data interpretation and feature engineering.

In SSE research, ML is promising approach for limited datasets, providing sophisticated correlations between descriptors and matrices performance, and accelerating rapid screening of materials for desired experimental validation [142–144].

SL based on labeled datasets for establishing input-output mappings as essentially required for predictive modeling [145]. In SL workflow, each entry within the training dataset entails a feature vector and its corresponding mark values. During model training, the algorithm iteratively improves its internal parameters to minimize the discrepancy between actual mark values and predicted outputs. In context of SSE, frequently employed SL methods including support vector machines (SVMs) [146], logistic Regression (LogR) [147], and Decision Tree [148], were leveraged into developing quantitative structure-property relationships.

Additionally, UL reveals intrinsic symmetry and uses unmarked data to extract geometric features. These approaches assess distance or similarities correlations among large data points to reduce dimensionality or enable clustering. It further process group sampling with relevant features and reveal primary patterns in complex datasets [133,149]. Interestingly, cluster analysis as a key UL tool has been extensively employed in studying SSE to identify novel materials class and dealing with property pattern of large databases [150,151]. Semi-supervised learning uses both labeled and unlabeled data, linking supervised and unsupervised models. This technique is more effective when encountering difficulty in acquiring labeled data [152,153]. In contrast, reinforcement learning working on iterative trial and error framework, refines decision by exploring numerous actions and improves learning following the outputs [154,155]. However, these two strategies are infancy in SSE research, rely on dataset expansion and improvement of algorithms in future [32].

Table 1. Comparison of ML and DL in AI-driven SSE design.

Feature	ML	DL
Typical algorithm	RF, SVMs, kernel ridge regression	GNNs, CNNs, RNNs
Data requirement	Small to medium datasets	Large and high-dimensional datasets
Feature engineering	Compel manually designed descriptors for example ionic radius, and lattice parameters	Automatically extract features from raw data (crystal configurations, interface patterns)
Complexity management	Captures moderate, often linear/non-linear relationships	Excels at high non-linear complex relationships
Interpretability	High-models are often interpretable	Moderate to low-models are often “black box”
Applications in SSE design	Property prediction (ionic conductivity, stability, mechanical properties), screening of known materials	Generative design of novel SSE compositions, modeling structure-properties relationships in large datasets, interface and defects analysis
Advantages	Fast, interpretable, effective for small datasets	Explore complex patterns, scalable, supports automated feature learning
Limitations	Performance limited by feature quality and dataset size	Requires large datasets and high and high computational resources

In addition, (MLIP) have nominated as a transformative theoretical framework for scrutinizing SSE, linking the venerable gap between the accuracy of first-principles calculations and the required efficiency for large-scale atomistic simulations. Furthermore, in case of potential energy surfaces based on DFT datasets, MLIP facilitate the exploration of ion transport, defect evolution, phase transitions, and electrolyte electrode interfacial phenomena over extensively extended length and time scales. Numerous MLIP frameworks have been successfully developed, such as deep potential molecular dynamics (DPMD), Gaussian Approximation potentials (GAP), Moment Tensor

Potentials (MTP), and NNPs, each offering tremendous advantages in terms of accuracy, transferability, and computational efficiency [156,157].

3.2. Deep Learning Approach

DL is a subset of ML, capable to autonomously perform extraction and interpretation of intricate features through neural networks composed of many hidden layers [158]. In contrast to traditional ML techniques, relying on manual features approach, DL architecture directly extracts features from raw data, allows modeling of complex patterns and leads to effective processing of large datasets.

For instance, neural networks including Graph Neural Networks (GNNs) and Convolutional Neural Networks (CNNs) have become promising tools in materials science. Specifically, CNNs widely used for optimizing data comprising regular grid structure such as projecting crystal structure onto voxel grids for smooth processing [159]. Interestingly, GNNs have a great attention in describing crystal structures as graphs, where atoms and their interactions are encoded as nodes and edges, thereby expelling the demand for manually designed descriptors. DL approach, in term of GNNs and CNNs, are competent of facilitating complex, non-linear relationships in broad SSE datasets. As summarized in Table 1, DL approach differ from classical ML in their stability to automatically extract features from raw data sources, including crystal graphs and interfacial characteristics. These capabilities make DL especially beneficial for reproductive design of novel SSE compositions and prediction of properties where conventional descriptors are scarce, thereby boosting rational material discovery [152]. In addition, GNNs are proficient at controlling network geometries denoting interatomic interactions, assisting direct learning of materials properties from atomic spatial geometries. Interestingly, the Crystal Graph Convolutional Neural Network (CGCNN), modified for crystalline materials, and reflecting substantial results in forecasting properties of SSE materials [143]. Advanced frameworks like M3GNet further integrate geometric and many-body interactions, ensuring more precise and transferable predictions of structure-properties relationships, ionic conductivity, and stability in SSE compared with conventional fingerprint-based ML approaches [28,160]. In addition, GNNs are proficient at controlling network geometries denoting interatomic interactions, assisting direct learning of materials properties from atomic spatial geometries. Curiously, the CGCNN, customized for crystalline materials, has considered substantial results in forecasting properties of SSE materials [143]. The progress in DL-driven potential functions offer significant improvement in MD, has revolutionized the simulation of interatomic interactions, reduced the computational cost without compromising the quantum level accuracy as essentially required for demonstration of fundamental ion transport phenomena in SSE [161].

3.3. ML-Guided SSE Design

The development and optimization of SSE for highly efficient energy storage technologies demands rapid and precise modeling techniques, to demonstrate the materials behavior under operation conditions. ML with capabilities predictive modeling of materials with tailored properties has revolutionized the discovery of materials. Researchers can train algorithms on available structural composition data, perform inverse designing of novel materials and improve the efficiency of optimization activity [162–164]. The proposed framework integrates both theory and experiments, further incorporates the rational formalism in emerging SSE.

Recently, the improvement in computational models has led to gain deeper insight into efficiency of SSE. Hamad et al. used random forest model to train 160 experimental samples with 50 distinct material features, see Figure 5a [165]. This model predicted the ionic conductivity with exceptional accuracy for Li- and Na-ion batteries by achieving coefficient of determination (R^2) as 0.97 with Root Mean Squared Error (RMSE) of 0.005 and Mean Absolute Error (MAE) of 0.007. Furthermore, LiYS₂ was identified as potential candidate by analyzing dataset of 30,000 compositions obtained from ICSD.

In a similar fashion, Mukhopadhyay et al. used eight types of algorithms to predict predicting the ionic conductivities of SSE [56]. Among these, Artificial Neural Network (ANN) provided comparatively better results and displayed outstanding accuracy along with 0 and 0.002 values of RMSE and MAE, respectively Figure 5b. By developing ML models and training based on different datasets, scientists can recognize superior SSE materials and play substantial performance for advanced energy storage technologies.

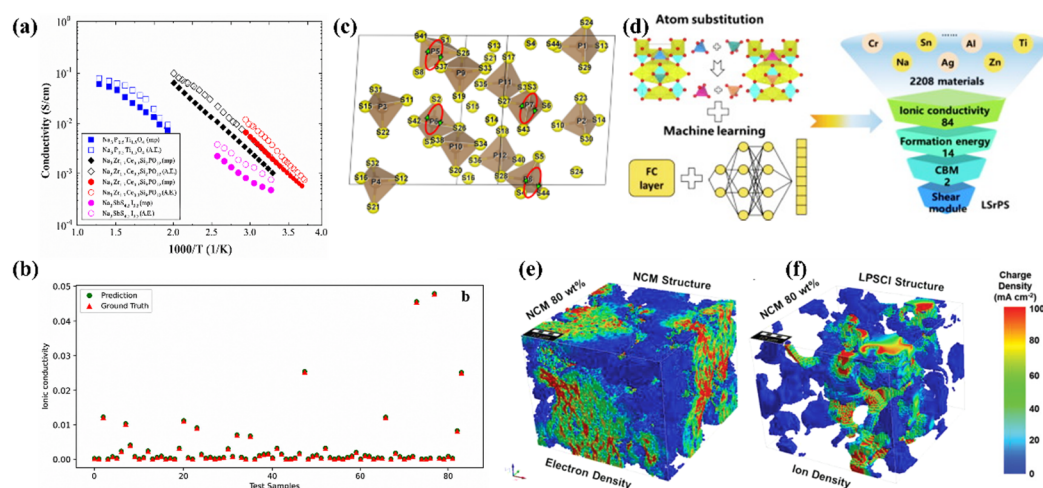


Figure 5. (a) Comparison of the experimental and predicted ionic conductivities results of four SSE as a function of temperature via RF model. Reproduced with permission [165]. Copyright 2023, Elsevier. (b) Comparison of experimental and predicted values of ionic conductivity by ANN. Reproduced with permission [56]. Copyright 2023, American Chemical Society. (c) The final structure of $\text{Li}_7\text{P}_3\text{S}_{11}$ with PS_4 polyanion in crystalline phase after (1 μs) ML–MD simulations at 300 K. Reproduced with permission [166]. Copyright 2023, Springer Nature. (d) ML–enabled fast screening framework to recognize the decent design of LGPS through element substitution. Reproduced with permission [167]. Copyright 2024, Elsevier. (e) Electron density in the electrode while (f) Li^+ density in the LPSCI structure with 80 wt% NCM electrodes. Reproduced with permission [168]. Copyright 2020, Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim.

The development of AI approaches from traditional ML to graph-based learning, generative models, and closed-loop autonomous frameworks is progressively transforming SSE advancement from empirical trial-and-error approaches toward data-driven novel design and stimulated materials discovery. In particular, the differences in model architecture, training dataset size, descriptor selection, and feature representation can influence prediction accuracy and generalization across different SSE families. Furthermore, we present Table 2 to systematically compare the model types, training data size, descriptors, target properties, and reported performance metrics from representative studies. These additions provide a more analytical perspective on the strengths, limitations, and applicability of various ML approaches for SSE research.

Table 2. Comparison of AI methodologies, representative algorithms, primary applications, key advantages of SSE and current challenges.

AI Methodology	Algorithms/Frameworks	Primary SSE Application	Key Advantages	Current Challenges	References
Supervised Machine Learning	RF, SVM	Ionic conductivity prediction, phase stability assessment, materials screening	Fast prediction efficiency and interpretability	Dependency on descriptor designing and limited extrapolation	[147,155]
Deep Learning	ANNs, CNNs, DNNs	Conductivity prediction and multi-property optimization	control complex nonlinear structure-property relationships	Requires large, high-quality datasets	[169,170]
Graph Neural Networks	CGCNN, M3GNet	Structure-aware property prediction and stability analysis	Direct learning from crystal structures without handcrafted descriptors	Computationally intensive and data requiring	[143,171]
Machine Learning Interatomic Potentials	DPMD, GAP, MTP, NNPs	Ion transport, defect evolution, interface dynamics	Near first-principles accuracy with improved efficiency	Requires widespread training datasets	[172,173]
Active Learning	Bayesian Optimization	Effective investigation of compositional space	Reduces data requirements and computational cost	Requires robust uncertainty evaluation	[174]
Generative AI	VAE, GAN, Diffusion Models	Inverse design and generation of novel SSE materials	Investigates previously unapproachable chemical spaces	Experimental validation	[175]
Closed-Loop Autonomous Discovery	AI + HTS + Automation	Autonomous SSE optimization and development	Self-improving materials discovery workflows	Limited practical implementation	[176]

In materials engineering, the well established Machine-learned potentials (MLPs) can effectively overcome the computational limitations due to classical force fields, expensive DFT computation and AIMD simulations [177,178]. For SSE, MLPs relies on high quality datasets and serves as powerful tools to treat larger systems and longer time-scale calculations with higher accuracy [179,180]. Based on MLPs, many researchers have demonstrated the behavior of Li-ion diffusion of $\text{Li}_7\text{P}_3\text{S}_{11}$, $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$, and Li_3YBr_6 [181,182]. By analyzing these parameters, researchers have demonstrated the diffusion mechanism of Li in ionic conductors, such as $\text{Li}_7\text{P}_3\text{S}_{11}$, $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$, and Li_3YBr_6 [166,183]. Moreover, the non-linear Arrhenius characteristics were demonstrated for diffusion of Li^+ ion in Li_3ErCl_6 and the polyanion rotation process of $(\text{PS}_4)^{-3}$ tetrahedra in $\text{Li}_7\text{P}_3\text{S}_{11}$ at normal temperature. This the finalized structure of $(\text{PS}_4)^{-3}$ after (1 μs) simulations at 300 K are shown in Figure 5c. Encouragingly, there is no paddle-wheel effect are observed at normal temperature according to simulations results [184], and $(\text{PS}_4)^{-3}$ group lowers the diffusion of Li-ion in $\text{Li}_7\text{P}_3\text{S}_{11}$ as demonstrated in Li argyrodite structure by BH_4 substitution [185]. The findings highlight the accuracy of MLP employed on complex systems and fundamental mechanisms to develop SSE with enhanced performance.

Although ML approaches both at molecular and atomic scale enable efficient interrogation of large datasets, facilitating the rapid screening of compositions and the identification of systems with stable structures and suggested properties. For instance, HTS has been employed to interpret correlations between electrochemical stability window, ionic conductivity and phonon band centers. In this training algorithms, the datasets consist of 53 compounds, were used to screen over a database of 14,000 materials comprising Li-ion, from MP. Only 18 suitable SSE containing oxides and halides were found promising. In addition, ML also accelerated HTS strategies for assessing elemental exchange process in SSE, being excessively time-consuming to investigate using conventional experimental and theoretical approaches. By introducing substitution in $\text{Li}_{10}\text{SrP}_2\text{S}_{12}$, has resulted in boosting the bulk modulus (10 GPa) and achieving high ionic conductivity (12.58 S/cm), as displayed in Figure 5d. This approach stems the pathway to combine physics-based model with data driven tools, allowing the possible design of electrolyte and emerging electrode materials with high efficiency and exceptional performance.

Moreover, the constraints of physics-based numerical approach can be overwhelmed using ML model-assisted digital twin techniques. Based on large datasets of structural features and concern material properties, these models authorize the valuation of structure-property relationships in real systems with complicated and non-uniform microstructures, as well as localized electrochemical process [21,186,187]. The understandings derived from digital twin modeling offer a powerful basis for the ideal design of batteries with defined topologies and the fine-tuning of their functional characteristics. For instance, a digital twin framework was currently established for all an ASSBs integrating a sulfide-based solid electrolyte [168,188]. This model enabled a comprehensive investigation of mass transport and interfacial electrochemical kinetics across a voxel-based, complex microstructure, thereby capturing phenomena occurring on micro-second timescales. Figure 5e,f presents the spatial distribution of Li^+ concentration within the solid sulfide electrolyte matrix as a function of the cathode active material loading. Under conditions of low solid sulfide electrolyte content (20 wt%) and consequently restricted ionic percolation pathways, the ion density becomes both spatially heterogenous and locally concentrated. These observations underscore the critical impact of the electrode-electrolyte blending ratio in achieving the delicate balance between electronic and ionic conductivities required for superior performance ASSBs.

3.4. Structure Property Correlations and Synthetic Optimization

The fundamental understanding of structural characteristics of SSE, the corresponding outcomes with underscoring the synthesis techniques, has been crucial for fabricating SSE materials. Adopting data-based AI approaches has significantly elucidated the correlation between structure and properties, providing comprehensions to optimize synthetic developments. Zhang et al. used UL model to generate numerical features of crystal structure by describing mXRD based features, as presented in Figure 6a,b [189]. The structural homogeneity pattern feature was successfully generated by spectral clustering and Agglomerative Hierarchical Clustering (AHC) techniques. Obviously, this strategy has reduced computational cost by highlight the role of inherent structural geometries in connection with enhanced Li-ion conductivity performance.

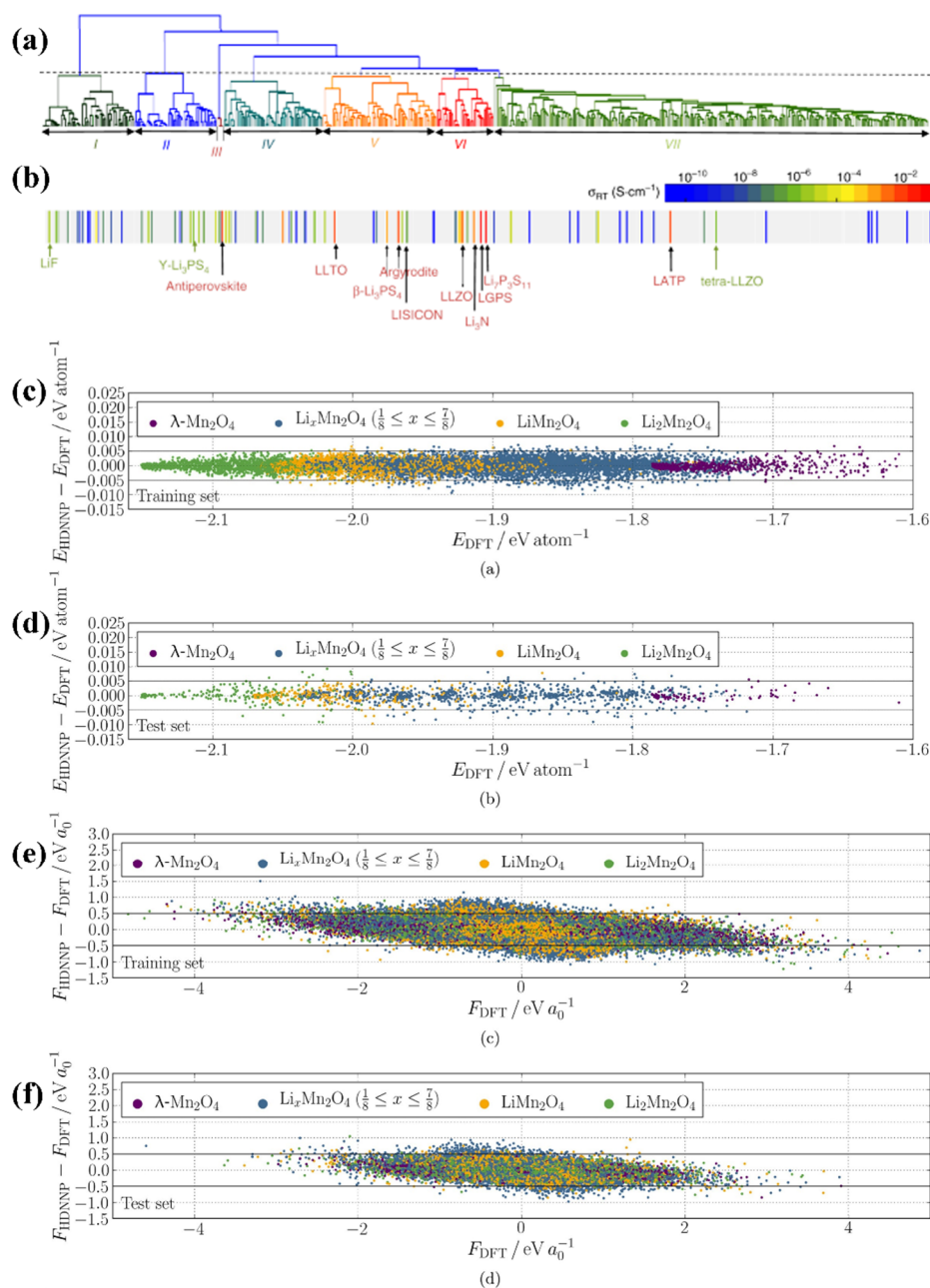


Figure 6. (a) Bottom-up diagram created through agglomerative hierarchical clustering approach. The dotted lines represent the area with understudy compounds mainly divided into (VII) groups, labeled as (I–VII) from left to right, and differentiated by various colors. (b) Mapping the dendro-gram to the conductivity demonstrates the grouping of defined solid-state Li-ion metals rods in groups (V and VI). The color bar represents the scale of σ_{RT} , while gray color denotes the conductivity was not calculated for equivalent compounds. Reproduced with permission [189]. Copyright 2020, arXiv [cond-mat. mtrl-sci]. Signed errors of the formation energies for training and test sets are illustrated in (c,d), while for the force components of training and data sets are presented in (e,f), where HDNNP as a function of the corresponding DFT results respectively. Note; the data points are colored according to the Li content of the highlighted structures. The black lines, indicating the error is less than 5 meV/atom for the energies and 0.5 eV/ a_0 for the force, accordingly. Reproduced with permission [190]. Copyright 2020, American Physical Society.

To study the phase transition of SSE materials, Eickhoff's group employed the High Dimensional Neural Network Potentials (HDNNP) technique to analyze geometrical properties of $(Li_xMn_2O_4)$ spinal system [190] Figure 6c–f. By integrating atom centered symmetry functions as signifiers, HDNNP efficiently captured complex electronic configurations features of material, incorporating the Jahn-Teller deformations due to different oxidation states. Further, the simulation has successfully demonstrated the phase transition in $Li_xMn_2O_4$ (orthorhombic to cubic) as confirmed experimentally by X-ray diffraction (XRD) technique.

Moreover, another research group of Gallo-Bueno's has used SL model and significantly updated the synthesis techniques of composite cathodes for ASSBs based on polymer electrolyte, as illustrated in Figure 7a,b. In addition, researchers used SVMs and demonstrated an impressive $R^2 = 0.95$ value for predicting active materials loading. In similar fashion, Logistic Regression (LR) model has attained accuracy up to 0.95 for electrode substance distribution. These breakthroughs in parallel with experimental validation for structural defects, caused by ball mining, have remarkably accelerated the ion migration. Consequently, the bridge between ML models and physical parameters establishes theoretical framework to refine material design and architectures. With these developments, Eickhoff and co-workers have employed weight initialization strategy for DL model to enhance the training performance of HDNNP for multi-layered and complex architectures [190]. This technique significantly improves efficiency, scrutinize prediction variances across diverse models. Furthermore, these approaches offer outlook in improving synthesis protocols and ensure geometrical integrity of novel SSE materials.

3.5. Interfacial Analysis and Dynamics Evolution

Interfacial instability remains one of the most significant barriers to the practical deployment of ASSBs, as chemical and electrochemical reactions at electrode-electrolyte interfaces can induce interphase formation, increase interfacial resistance, and accelerate capacity degradation [191]. Recent developments in AI and ML have extended their applications beyond bulk electrolyte discovery toward the prediction and engineering of stable interfaces. With the combined theoretical and experimental datasets, ML models may swiftly identify the interfacial compatibility, electrochemical stability, reaction energies etc. This can boost in exploring the feasible combination of electrode and electrolyte for SSE [192]. In addition, data-driven techniques are widely reported to identify artificial interlayers or protective coatings, suppressing the parasitic reactions across the interface and alleviating resistance during cycling. More obviously, the development of MLIP combined with MD studies have provided extensive analysis of ion transport mechanism, electrochemical degradation at the interfaces thereby, capable to gain deep mechanistic insights inclusively even not achievable with traditional computational tools. Recently, the development in AI-driven approaches have further highlighted the core descriptor related to interfacial degradation and electrochemical stability hence, accelerating the interface design engineering of ASSBs [193,194]. Together, these developments ensure that the AI-assisted interface modelling can not only promote materials screening but also bridge data-driven predictions with mechanistic insights, providing a sophisticated route towards the rational design of durable and high-performance ASSBs.

The ion transport kinetics and interfacial dynamics are key factors in practical realization of SSE. AI including NN potentials and DPMD are widely reported as transformative tools in understanding complex systems. These approaches show excellent performance to deal with complex interface models, significantly improving computational efficiency without compromising quantum level accuracy. Recently, Iwasaki employed universal neural network potentials (UNNPs) coupled with MD simulations and explored the interfacial kinetics between the garnet-type SSE $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZ) and (Li) metal and their ion. A comprehensive model composed of 1024 atoms were simulated and space-charge layer of 1 nm thickness was identified at LLZ/Li junction. The energetic analysis of Li-ion exchange further confirmed the activation barrier of 88 meV required for transporting Li-ion to electrolyte interface and conversely 159 meV to transport the metal in reverse direction, as shown in Figure 7c. These findings offer microscopic understandings of the fundamental concept of ion transport mechanism across interfacial regions.

Currently, more advanced simulation techniques adopted has significantly made progress in bulk ion diffusion analysis. Xia et al. used DPMD simulations to study Li_3OBr SSE and explored a direct linear relationship between ion mobility and defect density, as presented in Figure 7d. By introducing Schottky defect concentration of 4.2%, LiBr achieve $5.71 \times 10^{-4} \text{ S cm}^{-1}$ value of ionic conductivity at room temperature. This approach has revealed that the spatial arrangement independent of defects geometry, arranged in periodic or non-periodic pattern, has negligible influence on ion conductivity. These results indicate the transformative potential of adopting AI techniques to analysis the interfacial transport mechanism and executing dynamic simulations in studying SSE. By resolving complex molecular phenomena that avoid traditional computational approaches, the state-of-the-art AI techniques help to understand the fundamental mechanism essentially required for ion dynamics and interfacial transport phenomena in SSE. Such improvements further provide robust theoretical insights into engineering and highly efficient electrolyte materials. The progress in AI methodologies plays vital role, emerging as central tool to SSE design, has offered pathway to realized large scale viable ASSBs.

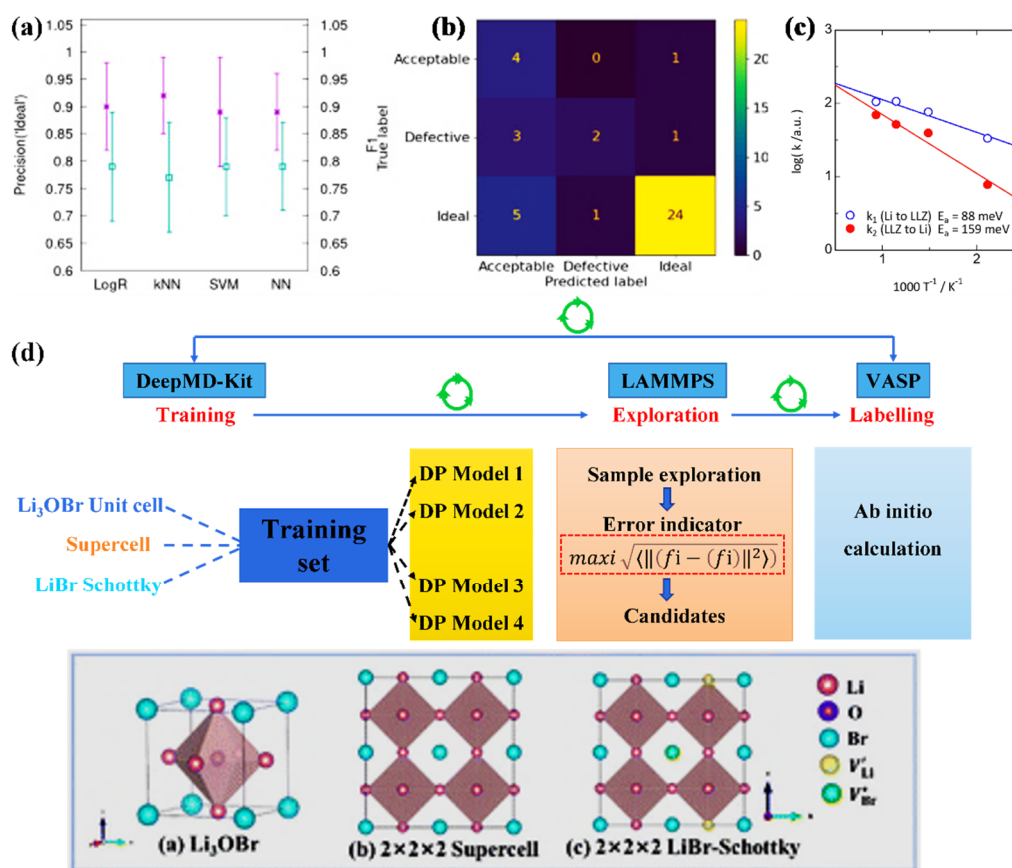


Figure 7. (a) Accuracy of the ideal class (left-side y-axis) labelled as stars and F1 on the (right-side y-axis) labelled as square for the trained multi-classification frameworks on the test set: standard deviation bars are involved for metrics and accuracy both, and the precision of the ideal class was nominated as the reference source. (b) Confusion matrix for LogR model whose hyperparameters were altered using accuracy of the ideal class as scoring function. Reproduced with permission [195]. Copyright 2024, Elsevier. (c) Arrhenius plots of rate constants for interfacial Li exchange. The blue line corresponds to the rate constant k_1 for the reaction from the LLZ phase to the Li phase, and the red line represents the rate constant k_2 for the opposite reaction; the slopes of the Arrhenius plots were used to measure the activation energies consistent to k_1 and k_2 (88 and 159 meV) accordingly. Reproduced with permission [196]. Copyright 2024, Springer Nature. (d) The workflow of DPGEN and the diagram of training models. Reproduced with permission [197]. Copyright 2024, Royal Society of Chemistry.

4. Data-Driven and Theoretical Strategies for SSE Discovery

The prolonged development of AI-driven SSE discovery depends on the availability of high-quality and manageable datasets. Recent research, such as Dynamic Database of Solid-State Electrolytes (DDSE) [198], have offering widespread experimental and computational SSE data, facilitating data-driven materials screening and ML model establishment. Nevertheless, challenges concerned to data standardization, database interpretation, integration of negative results, and continual data refinement remain to be addressed. Overcoming these challenges will be crucial for improving model reliability and accelerating next-generation AI-assisted SSE design. For instance, HTS enables systematic materials discovery by automating the recognition of target candidates from vast chemical spaces [134]. Public databases like ICSD [135], MP [136], and Material Cloud [137] offer substantial structural property data, providing a platform for HTS. By adopting standard screening criteria, scientific community may increasingly narrow down potential candidate pools to pinpoint promising target materials.

However, conventional HTS faces challenges when relying on databases like MP, AFLOWLIB, and OQMD. In materials science, the primary challenge is manifestation of incomplete datasets, where including thermal conductivity, elastic moduli, and electrochemical characteristics are frequently unfeasible. For instance, MP lacks essential descriptors like bandgaps or mechanical properties for many entries. Additionally, inconsistent data quality arises conflicting values for electrode potentials or lattice constants found between OQMD and MP can lead to unreliable inputs and inaccurate predictions. Variations in computational approaches and data standards further complicate integration. In energy storage research, timescale data often contain missing charge and

discharge cycle or irregular timestamps, reducing predictive reliability. These issues underscore the inherent limitations of public database driven HTS.

Researchers are struggling with HTS in combination with outcomes obtained using DFT and ML models. This approach overcomes the shortcomings, offers autonomous strategy to use AI for screening, has established a platform to design SSE with outstanding performance.

In this scenario, AI, DFT and HTS approaches have become emerging tools to develop novel SSE materials with enhanced functionalities. In present section, we describe the combined role of HTS, AI, DFT approaches to ensure reliability and improve the accuracy of analyzing SSE materials performance. In this context, Xie et al. used HTS technique by integrating DFT and bond valence method and proposed 329 candidates as promising SSE, as illustrated in Figure 8a,b [199]. Further, the ionic conductivity with high precision were obtained via feature extraction from structural parameters using M3GNet model. Also, BV-KMC simulation, as a competent theoretical simulation tool, provides a more updated approach than conventional DFT simulations for evaluating Li-ion migration behavior. In detailed analysis, the energy greater than hull (E_H) and bandgap (E_g), a series of materials with real-world application potential were efficiently recognized.

The primary focuses of collaborative approaches (DFT & AI) on moderating the computational costs while offering mechanistic understandings. For instance, Qi and its coworkers established the LAsou method, which combines ML with active learning algorithms, indicating in approximately a 30-fold proficiency improvement in predicting stable geometries of $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$ while revealing the physical reinforcements of geometries stability via theoretical approaches [200]. Moreover, Sendek and its coworkers gained an outstanding performance prediction by utilizing an ML model trained at microscopic level structure features, improve twice the prediction accuracy in comparison of traditional approaches Figure 8c [127]. These results facilitate the finding of novel superionic conductor materials, including $\text{Li}_5\text{B}_7\text{S}_{13}$.

Furthermore, Chen's and its groups introduced the synergistic predictive system based on the M3GNet model are illustrated in Figure 8d. Through ML models to predict substantial properties including bandgaps and stability, in combination with HTS and theoretical simulation justification, they markedly pinpointed 589,609 predicted stable materials from a pool of over thirty-two (32) thousand candidates, finally revealing eighteen (18) ideal SSE materials. Fujii [201] and his colleagues have integrated HTS with ML framework to study defect chemistry. By theoretical validation, the complex three-dimensional proton-conducting networks (Sr-doped $\text{Bi}_4\text{Ge}_3\text{O}_{12}$ and $\text{Bi}_{12}\text{SiO}_{20}$ were successfully engineered. The iterative cycle comprising three steps, ML, HTS and theoretical validation, has expedited the discovery of novel materials, further supported the accuracy and ensured reliability of the resulting predictive models. Ultimately, this combined research framework has optimized the accuracy and identified superior SSE materials, as potentially applicable to future ASSBs technology.

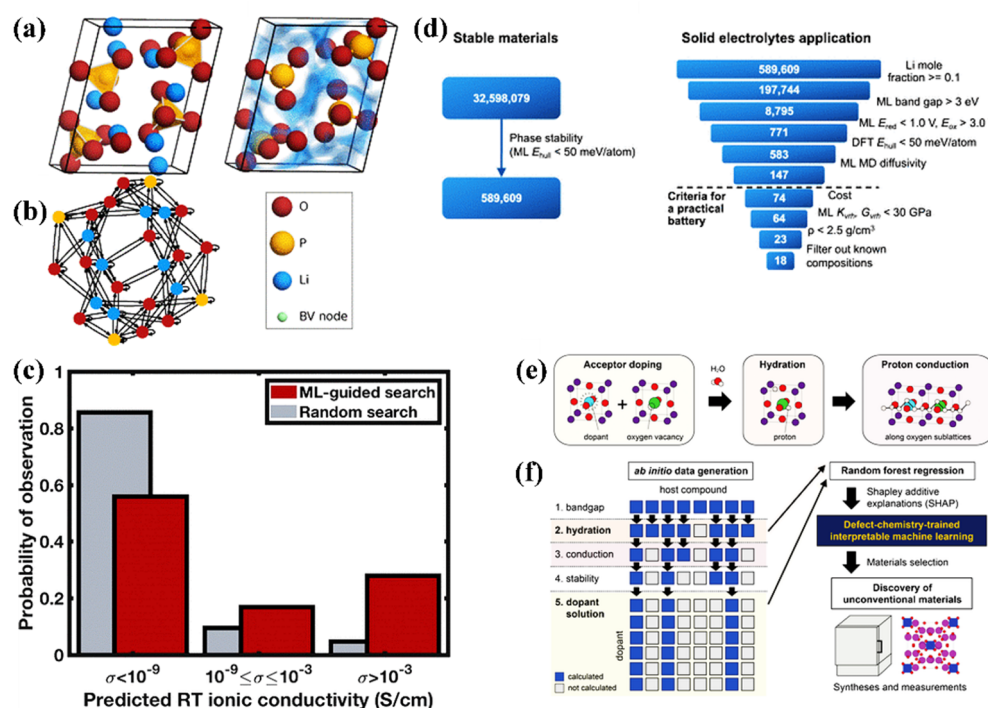


Figure 8. (a) Geometrical representation of $\text{Li}_4\text{P}_2\text{O}_7$ unit cell as SSE candidate. (b) Multigraph identifies the edges and nodes related to interatomic interactions and atoms of $\text{Li}_4\text{P}_2\text{O}_7$ crystal structure, respectively. Noted; the ionic

transport properties are evaluated using M3GNet model with multigraph serving as an input parameter. Reproduced with permission [199]. Copyright 2024, American Chemical Society. (c) Scaled distribution of predicted ambient-temperature ionic conductivities derived from a machine-learning-assisted random search for new materials. Reproduced with permission [127]. Copyright 2018, American Chemical Society. (d) Schematic of the proton conduction activation mechanism in oxides. The process begins with acceptor doping to generate oxygen vacancies, followed by hydration, where atmospheric moisture reacts with vacancies to incorporate hydroxyl protons into the lattice and eventually establishes a conductive pathway. Reproduced with permission [27]. Copyright 2024, American Chemical Society. (e) Activation process for proton conduction in oxides, divided into acceptor doping, hydration, and production of proton conduction pathway. The insertion of acceptor dopants into the host oxides induces oxygen vacancies and then subsequently hydration integrates protons into the solid by substituting these vacancies with hydroxyl groups upon exposure to moisture. (f) Integrated research pipeline for identifying novel proton-conducting oxides. The workflow encompasses ab initio calculations for hydration and dopant energetics, the development of interpretable machine learning models based on defect chemistry, and experimental validation via synthesis and characterization [201]. Copyright 2023, Wiley–VCH GmbH.

5. Conclusions

The incorporation of AI with theoretical and experimental approaches has significantly transformed the rational design of SSE, boosting discovery far beyond the limits of traditional trial-and-error independently computational approaches. We comprehensively studied the progress achieved in multiple dimensions. The DFT calculations combined with MD simulations offers foundation of demonstrating structural stability, electronic properties and ion transport phenomena [202–204]. Nevertheless, the AI approaches broadly implemented as complementary tools to traditional theoretical approaches, to accelerate the discovery and optimization of SSE. ML models including random forests, SVMs, and ANNs have revealed outstanding proficiency in predicting ionic conductivity, activation energy, and electrochemical stability directly from compositional and structural descriptors, facilitating rapid virtual screening of thousands of candidate materials. DL approaches, especially NNs and DPMD, have linked the gap between quantum mechanical accuracy and classical simulation efficiency, permitting realistic modeling of ion diffusion, interfacial dynamics, and phase transition over experimentally related timescale and system sizes. Additionally, AI-driven HTS frameworks, combined with public databases likes the MP and ICSD, have effectively identified novel superionic conductors, including sulfide, halide, and oxide-based SSE with optimized properties. Digital twin and active learning techniques have demonstrated the optimization of synthesis conditions, defect chemistries, and electrode-electrolyte interfaces, providing unprecedented understandings into structure-property relationships. Collectively, the developments in AI techniques have stimulated and underscored the optimization and discovery of next-generation SSE.

6. Challenges and Future Directions

Despite the substantial advancement, numerous critical challenges still remain that must be addressed to fully realize the transformative potential of AI-driven SSE design.

1. Data infrastructure and quality. High-quality, consistent datasets of ionic conductivity, electrochemical stability, and interfacial behavior remain scarce, especially for emerging material classes. Most ML models are trained on computationally derived properties rather than experimental measurements, leading to discrepancies when predictions are translated into real-world devices. The development of large, standardized, open-source databases that integrate experimental and high-quality computational data annotated with comprehensive metadata and uncertainty estimates will be vital for training robust and generalizable models.

2. Descriptor transferability and interpretability. Recent descriptors frequently fail to transfer across different material families (e.g., oxides vs. sulfides vs. halides) and across temperature regimes. Many models operate as black boxes, obscuring the physical mechanisms underlying their predictions. Physics-informed ML, which embeds fundamental laws such as conservation of energy, symmetry constraints, and charge balance directly into neural network architectures, promises to enhance extrapolative ability, interpretability, and scientific understanding.

3. Multi-fidelity and transfer learning. The high cost of experimental and high-accuracy computational data necessitates leveraging abundant low-fidelity data (e.g., from classical force fields or approximate DFT) to augment scarce high-fidelity measurements. Multi-fidelity and transfer learning approaches can bridge this gap, reducing reliance on costly validation while maintaining predictive accuracy.

4. Generative and inverse design. Up-to-date AI approaches primarily emphasis on screening existing candidates. Generative models including variational autoencoders, generative adversarial networks, and diffusion models provide the potential for inverse design: directly generating novel SSE compositions and structures with

targeted property profiles (e.g., specific ionic conductivity, stability window, or mechanical modulus) rather than merely ranking known materials.

5. Autonomous experimentation and closed-loop discovery. The integration of AI predictions with autonomous synthesis, characterization, and testing the so-called self-driving laboratory remains in its infancy. Scalable, fully closed-loop workflows that iteratively propose, synthesize, characterize, and improve SSE compositions without human intervention will be a game-changer. Combining robotic synthesis platforms, in situ characterization, and real-time ML feedback loops can dramatically accelerate the design-synthesis-testing cycle.

6. Digital twins and multiscale modeling. Future SSE development will benefit from digital twin technologies that incorporate atomistic simulations, continuum modeling, and experimental data into a unified framework. Such multiscale digital twins will permit real-time prediction of battery performance, degradation mechanisms, and thermal behavior under realistic operating conditions, guiding both material design and device engineering.

7. Bridge to real-world applications. Ultimately, AI-driven SSE discovery must translate into commercially feasible ASSBs. This requires not only optimizing ionic conductivity and interfacial stability but also addressing scalability, cost, manufacturability, and long-term cycle life. Despite these achievements, limitations associated with data quality and availability, model generalizability, uncertainty quantification, and real-world validation continue to hinder the realization of fully self-governing AI-driven discovery platforms. Future AI efforts should incorporate techno-economic considerations and manufacturing constraints into the design loop.

In summary, AI-driven rational design of SSE is still infancy, the trajectory is both profound and promising. By systematically addressing challenges in data quality, model interpretability, experimental validation, and autonomous workflows, and by embracing emerging paradigms such as generative design and self-driving laboratories, the research community is expecting to unlock the full potential of SSE for next-generation energy storage. This integrated, multidisciplinary approach bridging computational materials science, machine learning, synthetic chemistry, and electrochemistry will ultimately facilitate the translation of laboratory discoveries into practical, high-performance ASSBs, by contributing to sustainable and carbon-neutral future.

Author Contributions

Y.S.: conceptualization; Z.J. J.H. and Y.S.: writing—original draft preparation; all authors: investigation; Y.S.: supervision; Z.J. J.H. and Y.S.: writing—reviewing and editing. H.G.: reviewing; Q.W.: reviewing; H.U.D.: writing, reviewing, editing; G.G.: conceptualization, supervision. All authors have read and agreed to the published version of the manuscript.

Funding

This work was supported by National Key R&D Program of China (2023YFB4203900), Foundation of State Key Laboratory of Coal Conversion (Grant No. J26-27-620), Key Research and Development Program of Shaanxi Province (2025SF-YBXM-526), Shaanxi Sanqin Scholars Fund Project (2024STZZK07), Xiaomi Young Talents Program, Shaanxi Fundamental Science Research Project for Chemistry and Biology (Grant No.23JHQ078), Natural Science Basic Research Program of Shaanxi Province (No. 2024JC-TBZC-02), and the Major Science and Technology Projects of Yunnan Province (202302AB080019-4). Y. Su acknowledges the “Young Talent Support Plan” of Xi’an Jiaotong University, and the Fundamental Research Funds for the Central Universities (xtr072024025, xzy012024049 and xtr072024032). Supercomputing facilities were provided by Computing Center in Xi’an and Hefei Advanced Computing Center.

Institutional Review Board Statement

Not applicable.

Informed Consent Statement

Not applicable.

Data Availability Statement

No new data were created or analyzed in this study. Data sharing is not applicable to this article.

Conflicts of Interest

The authors declare no conflict of interest.

Use of AI and AI-Assisted Technologies

During preparation of this work, the authors used AI-assisted tool (Claude) only for language editing. The authors reviewed and edited the content as needed and take full responsibility for published article.

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