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Exploring Solvation Structure and Transport Behavior for Rational Design of Advanced Electrolytes for Next Generation of Lithium Batteries

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ABSTRACT

The efficacy of electrolytes significantly affects battery performance, leading to the development of several strategies to enhance them. Despite this, the understanding of solvation structure remains inadequate. It is imperative to understand the structure-property-performance relationship of electrolytes using diverse techniques. This review explores recent advancements in electrolyte design strategies for high capacity, high-voltage, wide-temperature, fast-charging, and safe applications. To begin, current state-of-the-art electrolyte design directions are comprehensively reviewed. Subsequently, advanced techniques and computational methods

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used to understand the solvation structure are discussed. Additionally, the importance of high-throughput screening and advanced computation of electrolytes with the help of machine learning is emphasized. Finally, future horizons for studying electrolytes are proposed, aimed at improving battery performance and promoting their application in various fields by enhancing the microscopic understanding of electrolytes.

I. INTRODUCTION

In the pursuit of a sustainable society powered by renewable energy sources, batteries play an indispensable role in stabilizing the output of stored solar and wind energy sources. A battery is a complex system comprising various components, including electrodes, electrolytes, separators, and others. Lithium batteries include lithium primary batteries (non-rechargeable), lithium-ion batteries (LIBs), and lithium metal batteries (LMBs). In this review, we will focus on the LIBs and LMBs systems. The main difference between the two types of systems is that LMBs use lithium metal as the anode material. There are already several review papers summarizing the history and future trends of lithium batteries.[1-11] While LIBs have been commercialized since 1991,[12] this system still has many issues to be addressed. LMBs with higher energy density also face numerous challenges. We need to ensure battery safety and reduce costs while improving electrochemical performance, such as achieving higher energy density, higher rates, and longer cycle life. Degradation exists in almost every component of the battery, underscoring the importance of studying electro-chemo-mechanical coupling failure mechanisms across multiple scales using a variety of techniques (**Figure 1a**).[13] On the cathode side, challenges include structural fading, particle cracking, transitional metal dissolution, gas evolution, and corrosion of the aluminum foil.[14,15] On the anode side, problems involve lithium plating/dendrite formation and volume change.[16] Electrolytes play a crucial role in transporting ions and masses while insulating electrons and stabilizing the interface

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between electrodes and electrolytes.[17] Electrolytes generally consist of salt, solvent (except for solid electrolytes), additive, and diluent. It is imperative to understand the relationship between the solvation structure, transport and redox properties of electrolytes to better screen advanced electrolytes for high capacity, high-voltage, wide-temperature, fast-charging, and non-flammable applications.

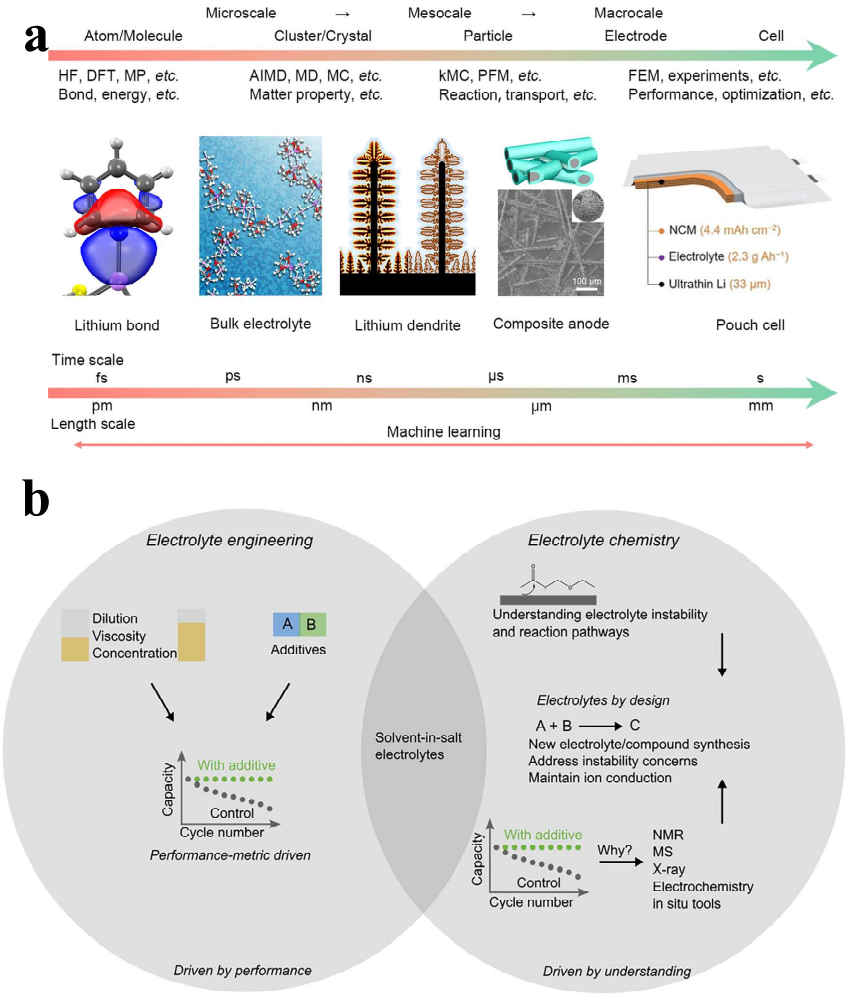

Figure 1 a) Different length and time scales for battery research.[18] Reproduced with

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permission from Chen et al., Angew. Chem., Int. Ed. Engl. 60, 24354-24366 (2021). Copyright 2021 Wiley. b) Electrolyte engineering and chemistry.[19] Reproduced with permission from Amanchukwu, Joule. 4, 281-285 (2020). Copyright 2019 Elsevier.

Two primary considerations emerge in electrolytes design strategies (**Figure 1b**).[19] One involves electrolyte engineering, propelled by performance considerations. The other revolves around electrolyte chemistry, relying on a bottom-up design strategy. Both facets play a significant role in the discovery of novel electrolytes, and obtaining accurately measured experimental parameters is critical for electrolyte design.

The investigation of the local solvation structure has been a subject of interest for several decades, given its essential role in battery performance. Studies have focused on concentration-dependent structures like solvent-separated ion pairs (SSIPs), contact ion pairs (CIPs), small aggregates (AGGs), and nanometric aggregates (n-AGGs) (**Figure 2a**).[20] Regarding cation diffusion, two primary mechanisms exist (**Figure 2b**): vehicular motion, where cations diffuse as one entity, and structural motion, involving dissociation/association exchange across different solvation shells. Often, a specific electrolyte exhibits a predominant diffusion mode, though both mechanisms may coexist.[20]

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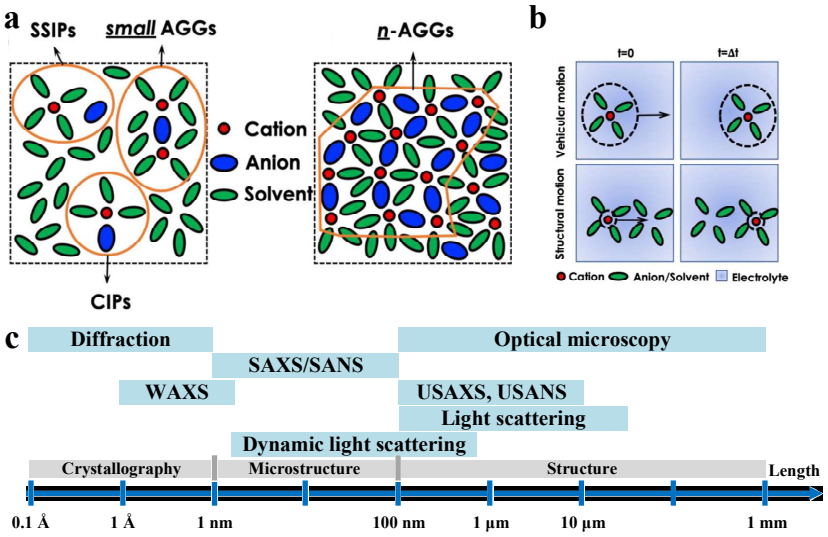

Figure 2 a) Schematic of SSIPs, CIPs, and AGGs. b) Illustration of vehicular and structural motion modes.[20] Reproduced with permission from Yu et al., ACS Energy Lett. 7, 461-470 (2022). Copyright 2022 American Chemical Society. c) Different research scales in electrolyte structure analysis.

Several experimental techniques have been developed to enhance our understanding of their structures in the field of electrolytes. Local information pertaining to intramolecular aspects of liquids can be acquired using various spectroscopic methods, e.g., nuclear magnetic resonance (NMR), vibrational spectroscopy – Raman, infrared (IR) or Fourier-transform infrared (FTIR), X-ray – pair distribution function (PDF), absorption (XAS), wide-angle (WAXS) and small-angle scattering (SAXS), and small-angle neutron scattering (SANS) (**Figure 2c**). Among these techniques, WAXS and small-angle scattering techniques, i.e., SAXS and SANS, enable the visualization of the intermolecular aspects of electrolytes, providing a global perspective (size, shape, and distance). Notably, the lithium content changes remain difficult to capture via X-ray techniques owing to the comparatively small scattering factor exhibited by lithium.[21]

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Additionally, Raman analysis may not have the ability to obtain precise quantitative information about the SSIPs, CIPs, and AGGs.[22] Therefore, it is essential to acknowledge that every analytical technique has limitations and imperfections. The combination of these techniques is considered to be a complementary approach to understanding electrolyte structures.[23] Moreover, the profound investigation of the structural and transport properties of electrolytes can be facilitated by means of computational techniques such as molecular dynamics (MD) simulation, Density functional theory (DFT) and Monte Carlo (MC) simulations,[24] and this is essential for the development of advanced electrolytes through molecular design.[25,26] However, traditional computational methods, such as classical MD and DFT, are hindered by limitations in the trade-off between accuracy and speed, making them inadequate for facilitating the molecular design process. Thus, the application of machine learning (ML) to molecular simulation has emerged as a promising approach to overcome these limitations.[27-30] On top of that, ML can also be used in electrolytes screening,[31] crack detection,[32] lifetime prediction,[33] health status prediction,[34] fast-charging protocols optimization,[35] and materials discovery.[36] The data used for training can be obtained from laboratory experiments, literatures, or datasets on open-sharing websites. Unlike traditional tedious trial-and-error methods in materials science, AI-assisted autonomous experimentation and characterization, high-performance computing (HPC), and data-driven methods can accelerate materials' discovery, synthesis, characterization, and application.

II. Electrolyte design strategies for diverse applications

Electrolytes play a crucial role in meeting the demands of a diverse range of applications, including but not limited to high capacity, high voltage, fast charging, wide temperature range, and enhanced safety such as thermal stability and non-flammability.[17,37-39] Batteries with different electrolytes exhibit distinct chemistries that influence electrochemical performance and safety (**Figure 3a**). Electrolytes encompass aqueous electrolytes, organic liquid

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electrolytes, quasi-solid electrolytes, solid-state electrolytes (SSEs), and hybrid electrolytes.[40] Aqueous electrolytes,[41] being low-cost and safe, find significant applications in large-scale grid energy storage systems. Organic liquid electrolytes,[42] with higher energy density, hold potential for use in computers, cell phones, and electric vehicles. However, their safety issues limit their further application. Organic liquid solvents generally consist of carbonate esters, ethers, and ionic liquids.[43] Severe decomposition, leading to safety concerns, is a drawback of carbonate ester-based electrolytes. Ether-based electrolytes, on the other hand, exhibit a low oxidation potential. However, the high cost of ionic liquid electrolytes limits their development. SSEs show promise in electric vehicles and even aircraft due to their high energy density and safety.[11] However, interface optimization is crucial for this system to achieve a longer cycle life. SSEs usually contain polymers, oxides, sulfides, and halides. The main challenge for polymer and oxide electrolytes is low ionic conductivity. Sulfide has the highest ionic conductivity but is moisture-sensitive and toxic. Halide electrolytes exhibit ionic conductivity close to that of sulfide electrolytes, while maintaining greater stability in ambient environments.[44-46] Quasi-solid electrolytes refer to SSEs with added small amounts of liquid electrolytes, which can solve the interfacial problems of SSEs.[47] Hybrid electrolytes can leverage the advantages of multiple electrolytes and compensate for their defects, achieving good overall performance.[48-50] The efficacy of electrolytes in these various application scenarios is contingent upon their ability to meet the specific requirements of each use case, necessitating diverse strategies to optimize performance.

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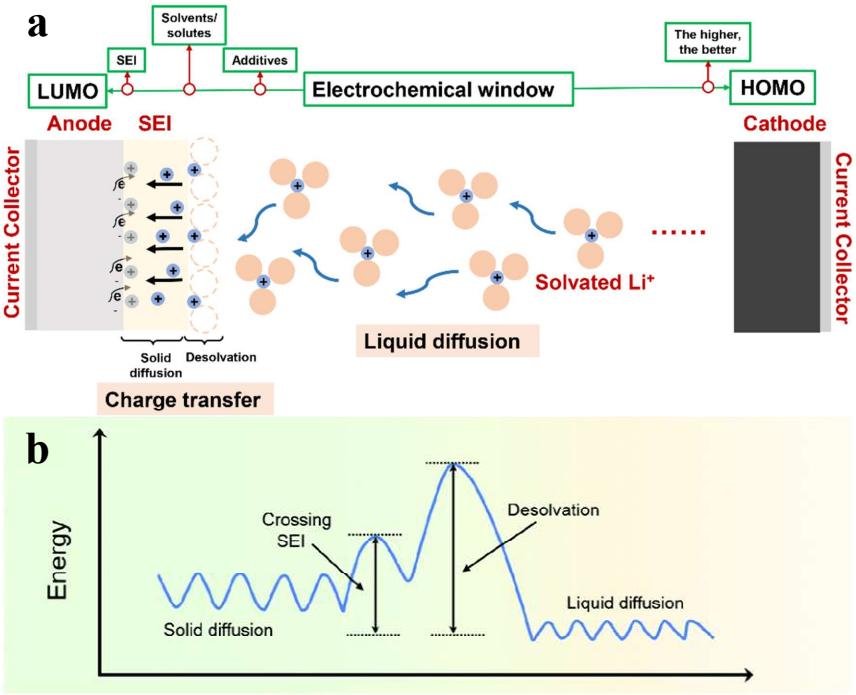

Figure 3 a) Thermodynamics and kinetics of electrolytes.[51] Reproduced with permission from Yang et al., Energy Storage Mater. 30, 113-129 (2020). Copyright 2020 Elsevier. b) The energy diagram of charging process of graphite in Lithium-ion batteries.[52] Reproduced with permission from Cai et al., Chem Soc Rev. 49, 3806-3833 (2020). Copyright 2020 Royal Society of Chemistry.

Firstly, redox flow batteries (RFBs) present a promising option for stationary energy storage devices due to their unique advantages, including decoupled energy and power, scalability, and design flexibility that can incorporate a variety of energy storage chemistries such as metal plating/stripping, solid intercalation, and photoelectrochemical reactions.[53,54] These features make RFBs a versatile option that can be extended to other energy storage and conversion systems. The performance of RFBs is highly dependent on the electrolytes' characteristics as energy carriers.[55] To achieve high-capacity electrolytes in RFBs, several strategies have been proposed, including using eutectic solvents, liquid molecules, and redox mediators.[54] These

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approaches hold significant potential for improving the performance of RFBs and expanding their applicability in various energy storage applications.

Furthermore, the augmentation of the charge cutoff voltage in batteries is a promising strategy to attain elevated energy density. However, this approach triggers the degradation of electrodes and the decomposition of electrolytes, thus impeding the practical application of high-voltage batteries.[56] To mitigate these issues, numerous approaches have been suggested to enhance the high voltage stability of batteries. These strategies encompass the development of chemically stable electrolytes, the creation of electrolytes free from ethylene carbonate (EC),[57] the construction of a robust cathode electrolyte interphase on the surface of the cathode, the utilization of preferentially oxidized additives,[58,59] such as organic molecules and salts, the introduction of functional group or diluents in ether-based solvents,[60-63] and the modulation of solvation structure through electrolyte concentration adjustment.[56,64] These strategies hold immense potential to boost the efficiency and stability of high voltage batteries, paving the way for their widespread application in various domains.

In addition, the regulation of solvation structure and the de-solvation process is crucial in achieving fast charging and optimal low-temperature operation.[65] This is attributed to de-solvation being the most substantial energy hurdle during the charge transfer process in electrolytic kinetics (**Figure 3b**).[52] The acceleration of the de-solvation process may be achieved through several approaches, including increasing the concentration of salts, fluorination, introducing fluorinated diluents/additives,[66,67] incorporating nonpolar diluents, adding strongly coordinated salt anions, and selecting robust solvated foreign cations.[68] These methods have demonstrated efficacy in promoting the de-solvation process, ultimately leading to improved fast charging and low-temperature performance. Also, introducing fluorinated diluents with a low freezing point is beneficial for low-temperature use, as it allows for tuning the Li^+-solvent-anion solvation structure.[69]

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Lastly, commercially available batteries commonly employ electrolytes containing thermally unstable and moisture-sensitive salts, such as $LiPF_6$, along with highly flammable organic solvents like carbonates and ethers, rendering batteries susceptible to thermal runaway. To mitigate the safety concerns associated with electrolytes, several approaches have been proposed. These approaches include the development of non-flammable organic electrolytes, aqueous electrolytes, eutectic solvent-based electrolytes, gel electrolytes, and solid electrolytes.[70] However, given the higher ionic conductivities, better wettability, and easier manufacturing processes associated with liquid or quasi-solid electrolytes (gel electrolytes) compared to all-solid electrolytes, the current focus should be developing non-flammable liquid electrolytes.[71] In this context, various methods have garnered significant attention. These include increasing the concentration of salts, namely high-concentrated electrolytes (HCEs) and localized HCEs (LHCEs), selecting thermally stable salts and solvents, and employing fluorinated solvents,[72,73] phosphate solvents, and fire-retardant additives.[74] Fluorinated solvents with strong carbon-fluorine bonds can improve the electrolyte's thermostability and reduce the generation of hydrogen radicals, thereby alleviating the combustion risk. Phosphate solvents possess the ability to capture hydrogen radicals and inhibit the combustion chain reaction. Furthermore, fire-retardant additives such as phosphonates and nitriles (< 10 wt%) can scavenge hydrogen radicals and terminate the radical chain reaction, thereby effectively improving the electrolyte's safety. Despite their benefits, fluorinated solvents have limited solubility, while phosphate solvents tend to decompose on the anode surface. To address these issues, it is advisable to design multi-salts and multi-solvent electrolytes and introduce multiple additives to integrate their positive effects. Such an approach can enhance the safety and electrochemical performance of the electrolyte synergistically.

In conclusion, to enhance the performance of electrolytes in batteries, numerous strategies have been explored by researchers. These include the utilization of high entropy,[75-79]

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asymmetric salt,[80-84] dual-salt,[71,85,86] dual-anion,[87-91] co-solvent,[92-95] fluorination,[96-100] fluorine-free,[101,102] HCEs,[103,104] LHCEs,[105,106] weakly solvating,[25,100,107-109] gelling,[110,111] eutectic,[112-117] and solidification tactics.[11] However, it is important to note that each of these strategies has its own set of advantages and disadvantages, and therefore, the best approach would be to combine some of these approaches to synergistically improve the properties of the electrolytes. Despite the successes achieved in battery performance, there remains a limited understanding of the altered solvation structure. Therefore, there is a pressing need to understand the solvation structure, solvation/de-solvation process, and interphase reaction at the microscopic level using in-situ/operando techniques. Advanced characterization techniques such as SAXS/SANS, WAXS, PDF, XAS, IR/FTIR, Raman and NMR, as well as AI-assisted computational methods such as DFT, MD, MC, and ML, could prove invaluable in this endeavor. Additionally, ML techniques can be employed to process experimental data and identify correlations between various factors.

III. Understanding the solvation structure and transport behavior of electrolytes

The SAXS/SANS, Raman, PDF, and MD techniques have become integral tools for investigating the microscopic characteristics of liquid electrolytes. By utilizing these techniques, it is possible to reveal important features such as solvation, molecular clusters (< 5 nm), ionic pairs, aggregates (> 5nm), and percolating networks in electrolytes.[23,118] Our group [23,119] have delivered a comprehensive review that delves into the intricacies of liquid electrolytes utilizing SAXS techniques, covering various aspects such as theory and experimental setup, data collection, processing, analysis, and in-situ/operando techniques. The authors have provided a comprehensive perspective on the application of SAXS techniques in the study of liquid electrolytes. Similarly, Hu et al.[120] presented a comprehensive review of the latest characterization techniques employed in unraveling the complex structure of electrolytes. The authors provided a detailed analysis of how these techniques have been applied to explore

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the fundamental aspects of electrolyte structures. Of course, the utilization of 2D and in-situ NMR,[121-125] in-situ electron paramagnetic resonance,[125] and operando Raman[126,127] techniques has also proven to be a powerful tool in elucidating the underlying mechanisms of working electrolytes. These in-situ/operando techniques provide us with a real-time and in-depth understanding of electrolyte evolution during cycling, thereby enabling the development of improved electrochemical systems.

Our group[128] undertaken a novel investigation into the microscopic nature of ethylene carbonate-based electrolytes via SAXS technique. The study marks the first instance in which such an inquiry has been conducted. The results indicate that EC clusters with a size of around 1 nm have been observed to form in linear carbonate, whereas no such clusters were detected in cyclic carbonate. Furthermore, it was observed that the presence of ethyl methyl carbonate (EMC) in $LiPF_6$/EC systems resulted in the formation of ~ 0.6 nm Li-PF_6 CIPs, a phenomenon that was not observed when admixing propylene carbonate (PC) in the case of $LiPF_6$/EC systems. The disparity in the structure and dielectric constant of linear and cyclic carbonates is thought to be the primary cause of these effects.

The continued study performed by our group [129] offers new insights into the solvation behavior of lithium bis(trifluoromethanesulfon)imidate (LiTFSI) in the mixture of EC with diethyl carbonate (DEC) electrolytes using SAXS and Raman spectroscopy. Specifically, we observed a flattening of the middle-q slope and the formation of a high-q peak in the SAXS data as the concentration of LiTFSI increased, which is attributed to charge ordering in saturated electrolytes (**Figure 4a, b, c, d**). Additionally, the Raman results reveal the formation of CIPs and AGGs in concentrated electrolytes, as evidenced by a decrease in band intensity of EC ring breathing and DEC bending, along with an increase in intensity of overlapped Li^+-EC and Li^+-DEC and uncoordinated DEC band, as well as a blue shift of $TFSI^-$ band (**Figure 4e, f**). Also, we[118] conducted a comprehensive investigation using SAXS/WAXS and Raman

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techniques to examine the effect of the shape of solvent molecules on charge ordering. When compared to ring-shaped molecules, linear-shaped molecules with middle q-values display a higher intensity and position of charge ordering peaks. This phenomenon is ascribed to the more prominent charge ordering and the greater distance between the charge centers of ring-shaped molecules. Thus, these findings suggest that linear-shaped molecules are more likely to exhibit charge ordering than ring-shaped molecules. Furthermore, the study suggests that short chain molecules are more prone to charge ordering than long chain molecules. These results highlight the significance of the shape and size of solvent molecules in influencing charge ordering, and could have important implications for the design of novel electrolytes with tailored properties. On top of that, we[130] carried out a thorough examination of sodium tetraphenylborate ($NaBPh_4$) in various solvents at different concentrations using a combination of SAXS, Raman, and MD methodologies. To specify, we observe SAXS peaks at high-q, middle-q, and low-q, which correspond to the shape and size of molecules or clusters, the distribution of molecules, and the existence of CIPs or AGGs, respectively. In addition, we assign the blue shift of Raman spectroscopy to an increase in solvation interaction, indicating a transformation from free-solvent to bound-solvent. We further employ MD simulation to interpret the two peaks observed in the middle-q region of the SAXS data as resulting from the short-range stacking of anion-based clusters in the concentrated $NaBPh_4$/PC electrolyte. Overall, this study underscores the value of a multi-angle approach for studying electrolytes.

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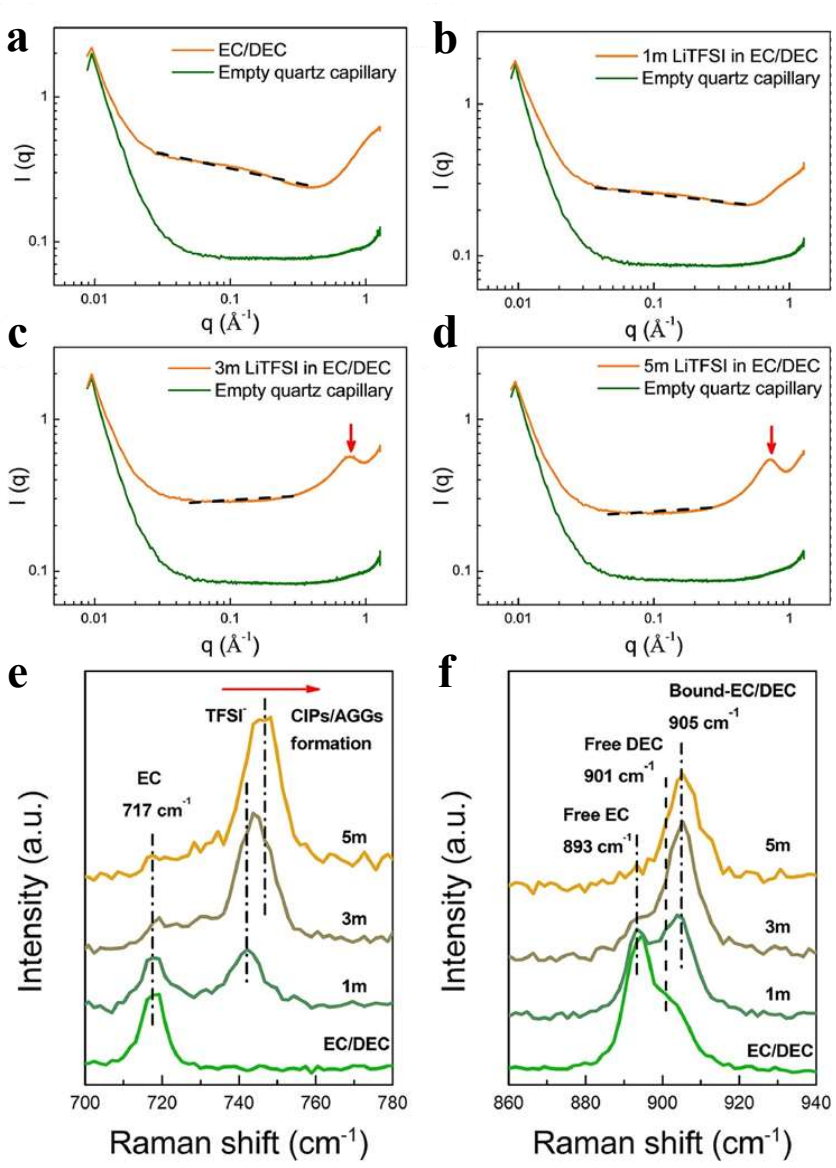

Figure 4 SAXS data of a) pure EC/DEC and LiTFSI in EC/DEC at salt concentration of b) 1 m, c) 3 m, and d) 5 m. Raman spectra of pure EC/DEC and 1 m, 3 m, 5 m LiTFSI in EC/DEC: e) 700 to 780 cm^{-1}; f) 860 to 940 cm^{-1}.[129] Reproduced with permission from Qian et

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Next, we[131] employed a rigorous investigation into the solvation structure of water in several salts featuring diverse symmetric anions at varying concentrations, utilizing SAXS/WAXS and MD simulations. We unequivocally demonstrate the existence of concentration-dependent anion solvated structures and anion networks in these aqueous solutions (**Figure 5d**). Notably, the anion network plays a critical role in contributing to the broader electrochemical stability windows.[132] Moreover, we identified intriguing linear correlations between the d-spacing of the anion solvated structure and the carbon and fluorocarbon content (**Figure 5a**), while we uncovered a concentration-dependent exponential relationship between the d-spacing of the anion solvated structure (**Figure 5b**). Recently, we[22] reported on the correlation between the solvated structure of anions and their association with SSIPs, as well as the association of anion networks with CIPs and AGGs (**Figure 5c**). Furthermore, we elucidated that the concentration-dependent viscosity behavior of electrolytes is linked to the anion-cation alternation and building blocks of ionic networks. Also, Tan et al.[133] applied a range of techniques, including SAXS/WAXS, SANS, NMR, and MD, to propose a superfluid framework for the ionic transport model of water-in-salt (WIS) electrolytes (**Figure 5e**). Their model considers the presence of water-free solid-like anion-clusters, which are interconnected with water clusters through nanoscale phase separation. Importantly, this framework elucidates the conditions that enable lithium ions to move freely, much like in an ionic atmosphere, at super high concentration. In our recent results,[134] we disclosed that an LiTFSI aqueous electrolyte, by introducing acetonitrile into water as a co-solvent, is capable of achieving an electrochemical stability window as large as the 21 m LiTFSI aqueous electrolyte window under relatively low concentrations (5 m). This capability solves the high viscosity problem associated with HCEs. These results suggest that the interplay between solvation structure and ionic network topology is a critical factor governing the behavior of electrolytes in solution.

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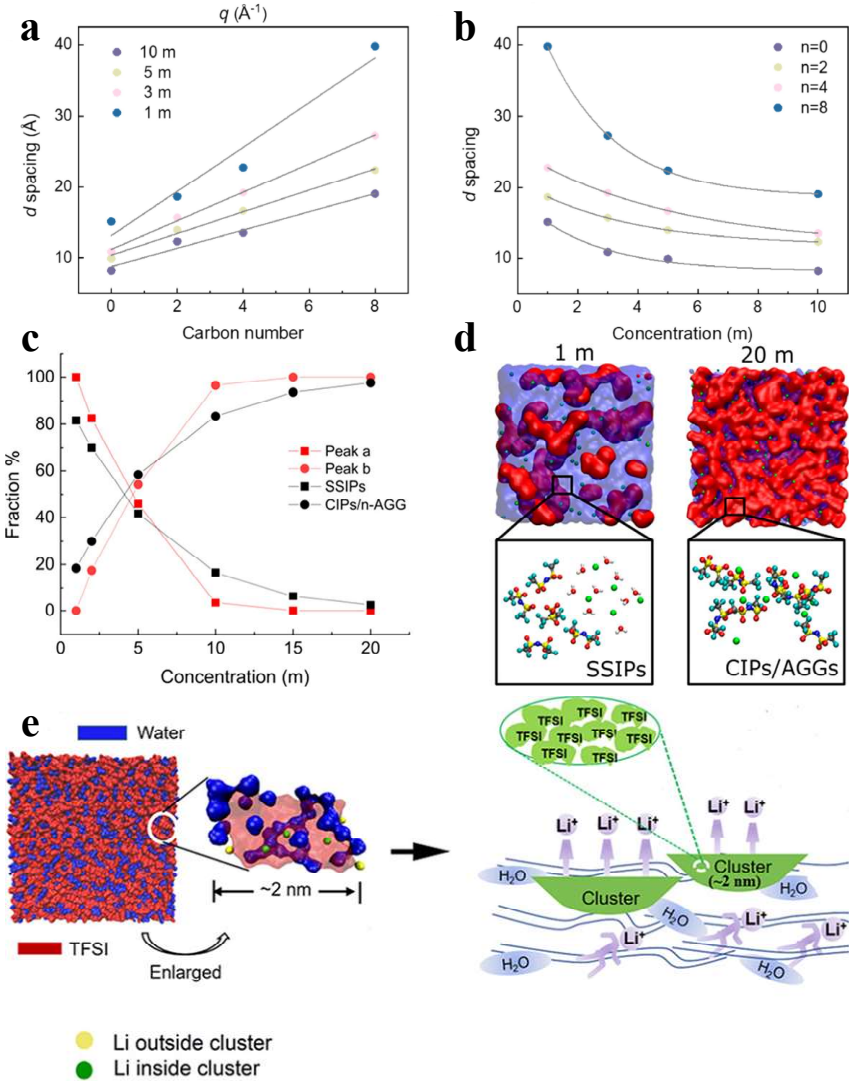

Figure 5 a) The linear relationship between carbon number and d spacing of Peak a at different concentration; b) correlation d-spacing evolution among different concentrations of four imide-based anions.[131] Reproduced with permission from Liu et al., Energy Storage Mater. 45, 696-703 (2022). Copyright 2022 Elsevier. c) The corresponding relationship between anions structure and SSIPs, CIPs, and AGGs; d) MD simulation snapshots of SSIPs, CIPs, and AGGs.[22] Reproduced with permission from Liu et al., Chem. Mater. 35, 2088-2094

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(2023). Copyright 2023 American Chemical Society. e) Illustration of free lithium-ion transport in WIS electrolytes.[133] Reproduced with permission from Tan et al., J. Phys. Chem. C. 125, 11838-11847 (2021). Copyright 2021 American Chemical Society.

Su et al.[135] executed a noteworthy investigation to systematically examine the solvation structure of LHCEs and localized saturated electrolytes (LSEs) using a variety of experimental and theoretical techniques, including Raman spectra, NMR, FTIR, and DFT calculations. The findings of their study suggest that unique solvation structures, such as CIPs and AGGs, have a profound impact on the bond length of solvent molecules and can also modify their lowest unoccupied molecular orbital – LUMO – and highest occupied molecular orbital – HOMO – levels, as well as their redox stability. Furthermore, the presence of non-solvating diluent molecules can influence the inner solvation sheath and regulate the size and distribution of solvated clusters. These results highlight the critical importance of selecting an appropriate solvent and a compatible diluent for the development of LHCEs and LSEs.

The implementation of solvation modulation has also exhibited a significant potential for optimizing the efficacy of RFBs by reducing redox potential and increasing the calendar life.[136-139] The energy density of fluid in RFBs depends on the concentration of redox-active molecules (redoxmers) that act as charge carriers in the liquid electrolyte. Shkrob and our group [140] conducted studies using SAXS and MD simulations to reveal the impact of confined charged redoxmers in crowded electrolytes on decay lifetime. The results demonstrated that aggregation behavior, prevalent in concentrated electrolytes, increased the decay lifetime, which can be modulated through structural modifications that positively affect practical applications. Similarly, Lu's group and our group.[141] used SAXS and MD simulations to show that balancing the competing forces of π-stacking and N-H bonding in crowded solutions can achieve higher redoxmers solubility, which is essential for achieving higher energy density in RFBs. Additionally, YZ and our group [142] employed a combination of SAXS, PDF, potential of mean force, MD, and DFT techniques to investigate saturated electrolytes at different states of charge.

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The results showed that the collective modes of ionic association correlated with the structural and dynamic properties of the electrolytes. As the concentration increased, the ionic association became more intense, resulting in rigid structures that caused non-linear increases in viscosity and rapid decreases in fluidity, conductivity, and diffusivity. The degree of concentration-dependent behavior differed at fully discharged and fully charged states due to additional interactions in the electrolytes. These fundamental studies provide valuable insights into the molecular design of RFB electrolytes and can aid in the practical application of RFBs.

In conclusion, the utilization of diverse experimental techniques has facilitated the elucidation of the microscopic features of liquid electrolytes across varying concentrations, electric field, temperatures, and charging rates. Notably, the size, shape, and composition of the constituent salts, solvents, additives, and diluents in the electrolytes can also modulate the solvation structure, thereby influencing the macroscopic properties and performance of the battery. While the structural analysis of electrolytes using both experimental and computational methods can be challenging, it is undoubtedly a worthwhile endeavor. These investigations hold significant promise for advancing the design of novel electrolytes, thus propelling the development of high-performance energy storage devices.

IV. AI-driven high-throughput screening and data-driven design of electrolytes

In the domain of scientific investigation, four fundamental paradigms prevail: empirical, theoretical, computational, and data-driven.[143,144] The Edisonian trial-and-error approach is no longer effective in the era of in silico materials rational design. Collaborative frameworks integrating theory and experiment now play a pivotal role in energy research.[145] Moreover, the integration of high-throughput computations and experiments with autonomous workflows, assisted by AI and machine learning (ML), is essential. The utilization of data-driven and high-throughput methods will undoubtedly expedite the design and discovery processes of battery materials and electrolytes across various time and length scales.[18,146]

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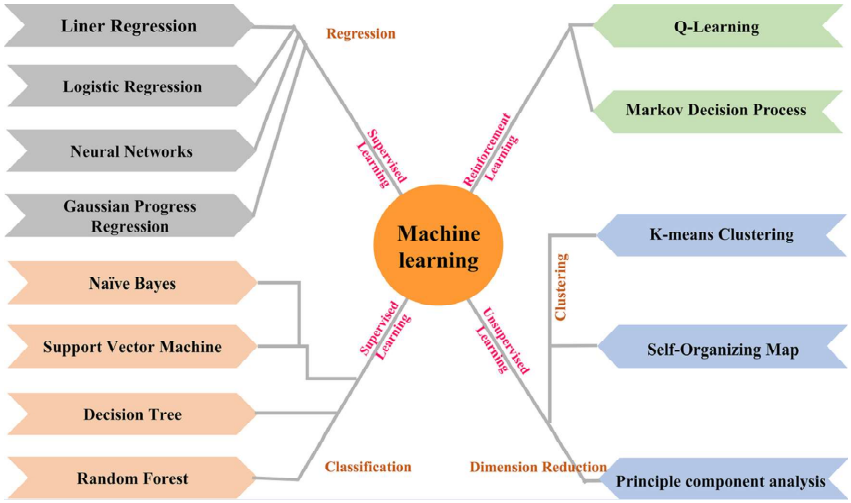

Figure 6 Common ML algorithms.[147] Reproduced with permission from Liu et al., Energy and AI. 3, 100049 (2021). Copyright 2021 Elsevier.

As a subset of AI, ML can acquire knowledge from existing data and information through the utilization of intricate algorithms and models. Liu et al. have systematically summarized the ML algorithms applied in energy research (**Figure 6**).[147] Broadly categorized, ML algorithms encompass supervised, semi-supervised, unsupervised, and deep learning techniques. Supervised ML algorithms are predominantly utilized for regression and classification tasks, whereas unsupervised ML algorithms find their applications in clustering and dimensionality reduction. Semi-supervised algorithms, however, represent a fusion of supervised and unsupervised techniques. Deep learning employs multiple neural network layers to systematically extract increasingly complex features from raw input data, outperforming other ML methods, especially as data sizes increase.[148] In general, deep learning comprises variational autoencoders (VAEs), adversarial autoencoders (AAEs), generative adversarial networks (GANs), and reinforcement learning (RL).[149] Autoencoders play a crucial role in exploring and optimizing chemical space. In contrast, RL operates within interactive

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environments, employing iterative trial-and-error methodologies guided by feedback mechanisms to enhance the learning process. The workflow for the design and discovery of battery materials and electrolytes using ML typically encompasses processes such as descriptor generation, feature engineering, model construction, prediction, and validation.[36] Finally, it is essential to differentiate between and avoid two types of overfitting: training overfitting and validation overfitting. Training overfitting arises when the model is excessively fitted to its parameters, while validation overfitting occurs when the model overfits to its hyperparameters, particularly in small datasets.[150] Considerations regarding overfitting in this context should exclude overparameterized models such as deep neural networks.

A. Autonomous search and optimization

It is essential to highlight the importance of autonomous experimentation and characterization for electrolyte discovery. Several review papers have systematically summarized the background, workflow, and trends of autonomous experimentation and characterization to accelerate discovery in the fields of biology, chemistry, and materials.[144,151-156] The Ceder Group systematically studies the autonomous synthesis and characterization of inorganic materials, integrating AI and ML into the workflow for data mining from literature (i.e., natural language processing), experimental execution, decision-making, and interpretation of results.[157-165]

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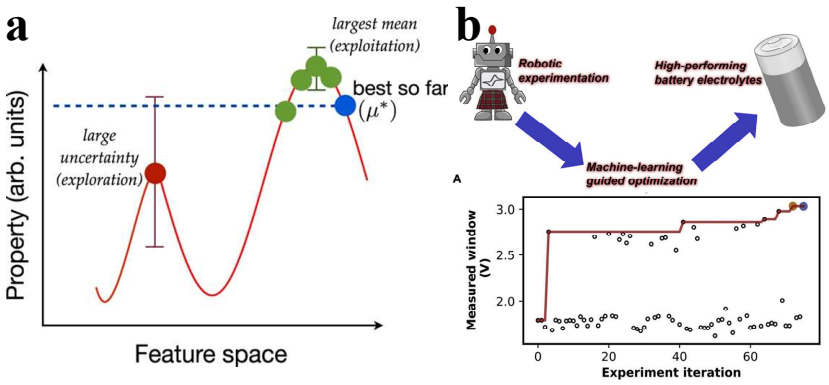

Figure 7 a) The trade-off between exploration and exploitaion.[166] Reproduced with permission from Balachandran et al., Sci Rep 6, 19660 (2016). Copyright 2016 Authors, licensed under a Creative Commons Attribution (CC BY) license. b) Results of autonomous optimization of electrolytes.[167] Reproduced with permission from Dave et al., Cell Rep. Phys. Sci. 1, 100264 (2020). Copyright 2020 Elsevier.

Benayad et al.[168] conducted a systematic review to explore the role of high-throughput experimentation and computation in accelerating the development of battery electrolytes and interfaces. Rodriguez et al.[169] presented a blueprint for a high-throughput protocol for electrolyte screening, synthesis, and characterization. Specifically, this study effectively develops deep eutectic solvents (DESs) using high-throughput automated methods and data-driven cheminformatics. The suggested workflow advocates for the use of low-cost and automatic robotic systems to integrate AI-driven algorithms, facilitating scientific exploration and optimizing electrolyte properties. In autonomous experimentation and characterization, achieving a balance between exploration and exploitation is essential in the design of experiments to effectively search for materials possessing desirable properties (**Figure 7a**).[166] In addition, hybrid learning, unlike model-based learning or data-based learning alone, possesses the capability to generate new knowledge.[170] This capability can assist researchers

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in gaining deeper insights into scientific discovery. The Viswanathan group has undertaken several projects related to the "closed-loop" optimization of electrolytes (**Figure 7b**).[167,171,172] An automated test stand is capable of mixing and characterizing both aqueous and non-aqueous electrolytes using coupled robotics and Bayesian optimization algorithms, thereby demonstrating precision and efficiency. The characterized parameters include conductivities, voltage stability window, pH levels, and electrochemical responses. The system autonomously explores two-dimensional electrolyte search spaces, successfully identifying novel blends with exceptional properties. The success of this system suggests the potential for rapid co-optimization studies in electrochemistry, especially when integrated with ML-based data assessment. Furthermore, this platform extends its utility beyond the battery community. With the exception of electrolyte formulation optimization, AI and ML can also be applied in tasks such as coin-cell mounting/disassembling and accelerated performance evaluation.[173,174]

B. AI-powered quantum chemical calculations and molecular dynamics simulations

While autonomous synthesis and characterization assisted by AI are critical for electrolyte optimization, AI can also be utilized to accelerate quantum chemical calculations and MD simulations. Within this domain, researchers need to balance the trade-off between computational efficiency and accuracy. The mainstream MD techniques, including classical MD (CMD), ab initio MD (AIMD), and machine-learning MD (MLMD), have proven to be effective supplements to experimental characterization for studying solvation structures, transport properties, and interfacial reactions of electrolytes (**Figure 8a**).[175] Unlike traditional CMD, which rely on empirical force fields, MLMD serves as a supplemental method that combines the efficiency of CMD and the accuracy of AIMD, making it capable of accelerating high-throughput MD investigations of electrolytes.[175] Multiple review papers have contributed to the exploration of ML force fields[176] and ML potentials.[27,177,178] The paper by Lee et al.[179] has shown that a machine-learned neural network force field is capable of replicating the

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structural and transport characteristics of molten salt with the precision of first principles while maintaining the computational efficiency of classical MD. Regarding electrolytes, Yao et al.[175] systematically compared how classical, ab initio, and ML−MD simulations were applied to study liquid electrolytes. Additionally, ML force fields and potentials have been utilized to improve the understanding and expedite the design of electrolytes.[180-182] The Cheng research team integrates ab initio MD, ML−MD, and free energy calculations to determine the redox potentials and elucidate the solvation structure of electrolytes (**Figure 8b**).[183,184] This effort is pivotal for clarifying and delineating the structure-properties relationship. The Grossman group's study shows that ML plays a critical role in accelerating MD simulations to understand atomic-scale dynamics in polymer electrolytes.[185,186] In one project, they employ graph dynamical networks to reveal local atomic-scale dynamics rather than global dynamics from MD simulations. In another project, autonomous coarse-grained MD (CGMD) is achieved by integrating CGMD and Bayesian optimization. This integration helps identify molecular-level correlations between lithium conductivity and material properties.

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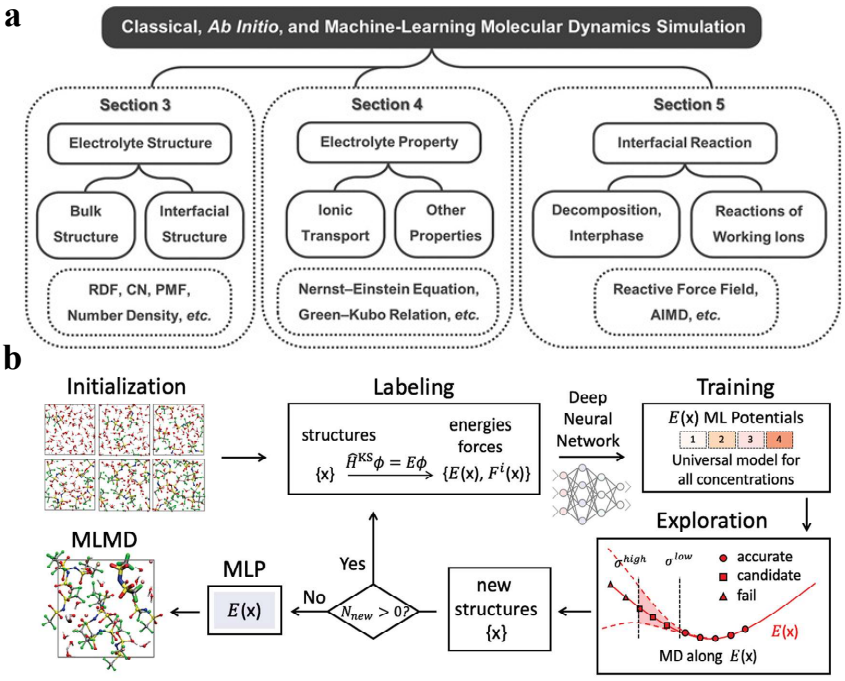

Figure 8 a) Framework of applying CMD, AIMD, and MLMD simulations in electrolytes.[175] Reproduced with permission from Yao et al., Chem Rev. 122, 10970-11021 (2022). Copyright 2022 American Chemical Society. b) Workflow of ML potential training process.[184] Reproduced with permission from Wang et al., J Am Chem Soc. 145, 4056 (2023). Copyright 2023 American Chemical Society.

C. Data-driven methods

Both experimental and computed Datasets play a critical role in the big data era, especially in the field of electrolytes, enabling us to delve deeper and gain valuable insights for the development of future energy storage systems.[187] There already exist databases of materials structures and properties,[188-191] as well as databases of batteries.[192,193] In the realm of electrolytes, AI and ML finds application in both liquid and solid electrolyte systems, enabling a deeper understanding of structural information, fundamental physicochemical properties, performance characteristics, and future developmental trends.

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In the field of the liquid electrolyte system, Gao et al.[194] constructed a database of 1399 solvent molecules using a graph-based algorithm. This provides insights into ion−solvent chemistry. Kim and colleagues[31] employed a data-driven approach, utilizing ML models that harnessed the elemental composition of electrolytes as features. Following feature reduction, they used linear regression, random forest, and bagging models to discern the significance of diminishing solvent oxygen content for achieving superior Coulombic efficiency (CE). Subsequently, these insights were applied to engineer fluorine-free electrolytes showcasing an exceptional CE of 99.70%. Wu et al.[195] explored how electrostatic potential (ESP) facilitates AI/ML in the strategic design of high-performance electrolytes, enabling the identification and screening of antisolvents, as well as solvents exhibiting robust or feeble solvating characteristics. In the realm of solid electrolytes, Forrest A. L. and colleagues[196] utilized a semi-supervised ML approach to unveil a superionic conductor within lithium batteries. They meticulously curated a set of 180 descriptors and conducted agglomeration clustering to discern underlying patterns. The efficacy of these descriptors was gauged by associating the clusters with experimental ionic conductivity data. Similar studies exploring the discovery of polymer electrolytes with the assistance of AI and ML have also been conducted.[197-200] Utilizing the nonlinear XGBoost algorithm, Wu and colleagues[201] identified the necessity of a material-specific and process-dependent approach to descriptor tuning.

V. Summary and outlook

Previously, researchers have primarily focused on electrodes. However, recent studies indicate that our community should redirect its attention towards electrolytes, an area that still holds significant untapped potential.[17] The urgent need to comprehend the structure-property-performance correlation in energy storage and conversion systems cannot be overemphasized. Varied application scenarios demand different properties and microscopic structures. However, experimental characterization tools are limited by resolution, making it difficult to decode some

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atomic or molecular-level phenomena. Fortunately, the recent advancement in computer technology, physical-chemical theory, and mathematical methods has made autonomous experimentation and high-throughput computing for atomic and molecular-level calculations and simulations possible. Consequently, a combination of experimental and computational approaches can substantially enhance the screening, prediction, and discovery of novel materials, including electrolytes. Here, we suggest considering several directions for advanced electrolyte design (**Figure 9**).

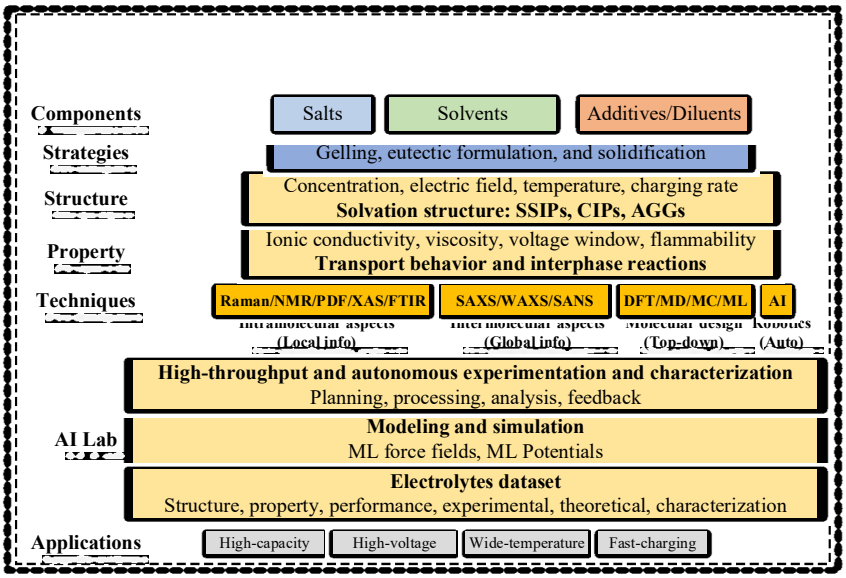

Figure 9 Characterizing and designing perspectives of advanced electrolytes.

Improved comprehension of electrolytes' solvation/de-solvation phenomena at the atomic and molecular levels is crucial for the enhanced molecular design of electrolytes. To facilitate this process, it is imperative to elucidate both the local and global aspects of electrolytes, including the solvation structure, such as SSIPs, CIPs, and AGGs, as well as the transport and interphase reactions. Each characterization technique possesses its own merits

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and limitations. While Raman spectroscopy, NMR, PDF, XAS and FTIR spectroscopy excel in investigating intramolecular aspects, SAXS/WAXS and SANS are adept at examining intermolecular aspects. Consequently, the utilization of one or multiple complementary methods can effectively unveil the complex microscopic behavior of electrolytes. Nevertheless, due to limitations in experimental resolution, a combination of computational methods such as DFT, MD, MC, and ML must be employed to simulate and model the evolutionary dynamics of electrolyte species during operation, thereby facilitating the visualization of microscopic phenomena.

Integration of various design approaches is paramount. Firstly, from a component standpoint, different salts, solvents, and additives exhibit distinct functionalities, each with advantages and limitations. Therefore, employing blended strategies such as dual/multi-salt, dual/multi-anion, dual/multi-solvent, and dual/multi-additive systems can prove beneficial. By regulating the proportion and concentration of these components, it becomes possible to achieve the superior comprehensive performance of electrolytes in batteries. Altering the composition of electrolytes influences solvation/de-solvation kinetics, consequently impacting transport and interphase reactions. Consequently, this leads to discernible changes in electrolytes' electrochemical performance. Secondly, a different perspective involves modifying the conventional liquid electrolyte form through gelling, eutectic formulation, and solidification techniques. These alternative approaches help mitigate the risks associated with the flammability and decomposition of organic solvents, which have been detrimental to battery lifetime, rate-capability, and safety.

The systematic investigation of the structure-property-performance relationship pertaining to electrolytes is imperative. It is important to note that isolated electrolytes differ from their behavior within a battery cell. Thus, we propose the utilization of in-situ/operando techniques and equipment to capture the real-time evolution of electrolytes within batteries and

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study their solvation/de-solvation behavior, reaction mechanisms, and decomposition processes. These endeavors provide invaluable insights for the design of advanced electrolytes. AI and ML can play a role in high-throughput autonomous experimentation, characterization, modeling, and simulation, as well as data-driven screening and design. However, challenges still exist in this domain. The realization of AI-enabled high-throughput autonomous experimentation and data-driven screening is contingent upon the meticulous configuration of AI systems, enabling the establishment of flexible and robust automated pipelines and seamless interconnections.[151] Endowed with both 'hands' for experimentation and theoretical modeling, as well as 'eyes' for characterization, silicon-based intelligence stands poised to revolutionize the electrolytes research. Moreover, while substantial advancements have been made in theoretical research, it is imperative to ensure the enhanced verifiability of the results.[202] Furthermore, the quality of the dataset needs to be improved, including gathering, analysis, and reporting, which are critical to the output of ML.[203]

SUPPLEMENTARY MATERIAL

There is no supplementary material for this review.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Xiaozhao Liu: Writing – original draft. Tao Li: Writing – review & editing. Volodymyr Koverga:

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Writing – revise. Hoai T. Nguyen: Writing – revise. Anh Ngo: Writing – revise

DATA AVAILABILITY

Data sharing is not applicable to this article as no new data was created or analyzed in this study.

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PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0187154

REFERENCES

[1]J. M. Tarascon and M. Armand, Nature **414** (6861), 359 (2001).

[2]F. Wu, J. Maier, and Y. Yu, Chem. Soc. Rev. **49** (5), 1569 (2020).

[3]H. Zhang, C. Li, G. G. Eshetu, S. Laruelle, S. Grugeon, K. Zaghib, C. Julien, A. Mauger, D. Guyomard, T. Rojo, N. Gisbert-Trejo, S. Passerini, X. Huang, Z. Zhou, P. Johansson, and M. Forsyth, Angew Chem Int Ed Engl **59** (2), 534 (2020).

[4]John B. Goodenough and Youngsik Kim, Chem. Mater. **22** (3), 587 (2010).

[5]J. Xie and Y. C. Lu, Nat. Commun. **11** (1), 2499 (2020).

[6]Stefania Ferrari, Marisa Falco, Ana Belén Muñoz-García, Matteo Bonomo, Sergio Brutti, Michele Pavone, and Claudio Gerbaldi, Adv. Energy Mater. **11** (43) (2021).

[7]C. P. Grey and D. S. Hall, Nat. Commun. **11** (1), 6279 (2020).

[8]J. B. Goodenough and K. S. Park, J. Am. Chem. Soc. **135** (4), 1167 (2013).

[9]Yuming Chen, Maowen Xu, Yunhui Huang, and Arumugam Manthiram, Chem **8** (2), 312 (2022).

[10]Sanjeev K. Sharma, Gaurav Sharma, Anurag Gaur, Anil Arya, Fateme Sadat Mirsafi, Reza Abolhassani, Horst-Günter Rubahn, Jong-Sung Yu, and Yogendra Kumar Mishra, Energy Adv. **1** (8), 457 (2022).

[11]Jürgen Janek and Wolfgang G. Zeier, Nat. Energy **8** (3), 230 (2023).

[12]A. Yoshino, Angew Chem Int Ed Engl **51** (24), 5798 (2012).

[13]Christoph R. Birkl, Matthew R. Roberts, Euan McTurk, Peter G. Bruce, and David A. Howey, J. Power Sources **341**, 373 (2017).

[14]Tanvir R. Tanim, Peter J. Weddle, Zhenzhen Yang, Andrew M. Colclasure, Harry Charalambous, Donal P. Finegan, Yanying Lu, Molleigh Preefer, Sangwook Kim, Jeffery M. Allen, Francois L. E. Usseglio-Viretta, Parameswara R. Chinnam, Ira Bloom, Eric J. Dufek, Kandler Smith, Guoying Chen, Kamila M. Wiaderek, Johanna Nelson Weker, and Yang Ren, Adv. Energy Mater. **12** (46), 2202795 (2022).

[15]Pin Du, Dongxu Liu, Xiang Chen, Hongwei Xie, Xin Qu, Dihua Wang, and Huayi Yin, Energy Storage Mater. **57**, 371 (2023).

10 July 2025 20:08:58

AIP Publishing Applied Physics Reviews ACCEPTED MANUSCRIPT
This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.
PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0187154

[16]Thomas Waldmann, Björn-Ingo Hogg, and Margret Wohlfahrt-Mehrens, J. Power Sources **384**, 107 (2018).

[17]Y. S. Meng, V. Srinivasan, and K. Xu, Science **378** (6624), eabq3750 (2022).

[18]X. Chen, X. Liu, X. Shen, and Q. Zhang, Angew Chem Int Ed Engl **60** (46), 24354 (2021).

[19]Chibueze V. Amanchukwu, Joule **4** (2), 281 (2020).

[20]Zhou Yu, Nitash P. Balsara, Oleg Borodin, Andrew A. Gewirth, Nathan T. Hahn, Edward J. Maginn, Kristin A. Persson, Venkat Srinivasan, Michael F. Toney, Kang Xu, Kevin R. Zavadil, Larry A. Curtiss, and Lei Cheng, ACS Energy Lett. **7** (1), 461 (2022).

[21]Craig A. Bridges, Xiao-Guang Sun, Jinkui Zhao, M. Parans Paranthaman, and Sheng Dai, The Journal of Physical Chemistry C **116** (14), 7701 (2012).

[22]Xinyi Liu, Shao-Chun Lee, Soenke Seifert, Lilin He, Changwoo Do, Randall E. Winans, Gihan Kwon, Y. Z, and Tao Li, Chem. Mater. **35** (5), 2088 (2023).

[23]Kun Qian, Randall E. Winans, and Tao Li, Adv. Energy Mater. **11** (4) (2020).

[24]Z. Deng, T. P. Mishra, E. Mahayoni, Q. Ma, A. J. K. Tieu, O. Guillon, J. N. Chotard, V. Seznec, A. K. Cheetham, C. Masquelier, G. S. Gautam, and P. Canepa, Nat. Commun. **13** (1), 4470 (2022).

[25]Y. Chen, Z. Yu, P. Rudnicki, H. Gong, Z. Huang, S. C. Kim, J. C. Lai, X. Kong, J. Qin, Y. Cui, and Z. Bao, J. Am. Chem. Soc. **143** (44), 18703 (2021).

[26]D. Bedrov, J. P. Piquemal, O. Borodin, A. D. MacKerell, Jr., B. Roux, and C. Schroder, Chem. Rev. **119** (13), 7940 (2019).

[27]V. L. Deringer, M. A. Caro, and G. Csanyi, Adv. Mater. **31** (46), e1902765 (2019).

[28]C. Schran, F. L. Thiemann, P. Rowe, E. A. Muller, O. Marsalek, and A. Michaelides, Proc Natl Acad Sci U S A **118** (38) (2021).

[29]N. R. Singstock, J. C. Ortiz-Rodriguez, J. T. Perryman, C. Sutton, J. M. Velazquez, and C. B. Musgrave, J. Am. Chem. Soc. **143** (24), 9113 (2021).

[30]F. Daeyaert, F. Ye, and M. W. Deem, Proc Natl Acad Sci U S A **116** (9), 3413 (2019).

[31]S. C. Kim, S. T. Oyakhire, C. Athanitis, J. Wang, Z. Zhang, W. Zhang, D. T. Boyle, M. S. Kim, Z. Yu, X. Gao, T. Sogade, E. Wu, J. Qin, Z. Bao, S. F. Bent, and Y. Cui, Proc Natl

10 July 2025 20:08:58

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PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0187154

Acad Sci U S A **120** (10), e2214357120 (2023).

[32]Tianyu Fu, Federico Monaco, Jizhou Li, Kai Zhang, Qingxi Yuan, Peter Cloetens, Piero Pianetta, and Yijin Liu, Adv. Funct. Mater. **32** (39), 2203070 (2022).

[33]Kristen A. Severson, Peter M. Attia, Norman Jin, Nicholas Perkins, Benben Jiang, Zi Yang, Michael H. Chen, Muratahan Aykol, Patrick K. Herring, Dimitrios Fraggedakis, Martin Z. Bazant, Stephen J. Harris, William C. Chueh, and Richard D. Braatz, Nat. Energy **4** (5), 383 (2019).

[34]Darius Roman, Saurabh Saxena, Valentin Robu, Michael Pecht, and David Flynn, Nat. Mach. Intell. **3** (5), 447 (2021).

[35]P. M. Attia, A. Grover, N. Jin, K. A. Severson, T. M. Markov, Y. H. Liao, M. H. Chen, B. Cheong, N. Perkins, Z. Yang, P. K. Herring, M. Aykol, S. J. Harris, R. D. Braatz, S. Ermon, and W. C. Chueh, Nature **578** (7795), 397 (2020).

[36]C. Lv, X. Zhou, L. Zhong, C. Yan, M. Srinivasan, Z. W. Seh, C. Liu, H. Pan, S. Li, Y. Wen, and Q. Yan, Adv. Mater. **34** (25), e2101474 (2022).

[37]K. Xu, Chem. Rev. **104** (10), 4303 (2004).

[38]K. Xu, Chem. Rev. **114** (23), 11503 (2014).

[39]M. Li, C. Wang, Z. Chen, K. Xu, and J. Lu, Chem. Rev. **120** (14), 6783 (2020).

[40]Shuang-Jie Tan, Wen-Peng Wang, Yi-Fan Tian, Sen Xin, and Yu-Guo Guo, Adv. Funct. Mater. **31** (45), 2105253 (2021).

[41]Sailin Liu, Ruizhi Zhang, Jianfeng Mao, Jodie Yuwono, Cheng Wang, Kenneth Davey, and Zaiping Guo, Appl. Phys. Rev. **10** (2), 021304 (2023).

[42]Y. K. Liu, C. Z. Zhao, J. Du, X. Q. Zhang, A. B. Chen, and Q. Zhang, Small **19** (8), e2205315 (2023).

[43]J. G. Zhang, W. Xu, J. Xiao, X. Cao, and J. Liu, Chem. Rev. **120** (24), 13312 (2020).

[44]Xiaona Li, Jianwen Liang, Xiaofei Yang, Keegan R. Adair, Changhong Wang, Feipeng Zhao, and Xueliang Sun, Energy Environ. Sci. **13** (5), 1429 (2020).

[45]Hiram Kwak, Shuo Wang, Juhyoun Park, Yunsheng Liu, Kyu Tae Kim, Yeji Choi, Yifei Mo, and Yoon Seok Jung, ACS Energy Lett. **7** (5), 1776 (2022).

10 July 2025 20:08:58

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This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.
PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0187154

[46]B. He, F. Zhang, Y. Xin, C. Xu, X. Hu, X. Wu, Y. Yang, and H. Tian, Nat. Rev. Chem. **7** (12), 826 (2023).

[47]H. Huo and J. Janek, Natl. Sci. Rev. **10** (6), nwad098 (2023).

[48]H. Liang, L. Wang, A. Wang, Y. Song, Y. Wu, Y. Yang, and X. He, Nanomicro Lett **15** (1), 42 (2023).

[49]Y. Zheng, Y. Yao, J. Ou, M. Li, D. Luo, H. Dou, Z. Li, K. Amine, A. Yu, and Z. Chen, Chem. Soc. Rev. **49** (23), 8790 (2020).

[50]Trang Thi Vu, Hyeong Jun Cheon, Seo Young Shin, Ganghoon Jeong, Eunsol Wi, and Mincheol Chang, Energy Storage Mater. **61** (2023).

[51]Huicong Yang, Juan Li, Zhenhua Sun, Ruopian Fang, Da-Wei Wang, Kuang He, Hui-Ming Cheng, and Feng Li, Energy Storage Mater. **30**, 113 (2020).

[52]W. Cai, Y. X. Yao, G. L. Zhu, C. Yan, L. L. Jiang, C. He, J. Q. Huang, and Q. Zhang, Chem. Soc. Rev. **49** (12), 3806 (2020).

[53]Minjoon Park, Jaechan Ryu, Wei Wang, Jaephil Cho, and Richland W. A. United States Pacific Northwest National Lab. Pnnl, Nat. Rev. Mater. **2** (1) (2016).

[54]L. Zhang, R. Feng, W. Wang, and G. Yu, Nat. Rev. Chem. **6** (8), 524 (2022).

[55]W. Wang and V. Sprenkle, Nat. Chem. **8** (3), 204 (2016).

[56]Hao Jia and Wu Xu, Trends Chem. **4** (7), 627 (2022).

[57]Yu Wu, Dongsheng Ren, Xiang Liu, Gui-Liang Xu, Xuning Feng, Yuejiu Zheng, Yalun Li, Min Yang, Yong Peng, Xuebing Han, Li Wang, Zonghai Chen, Yang Ren, Languang Lu, Xiangming He, Jitao Chen, Khalil Amine, and Minggao Ouyang, Adv. Energy Mater. **11** (47) (2021).

[58]Jian Xia, Remi Petibon, A. Xiao, W. M. Lamanna, and J. R. Dahn, J. Electrochem. Soc. **163** (8), A1637 (2016).

[59]Jian Xia, Remi Petibon, Deijun Xiong, Lin Ma, and J. R. Dahn, J. Power Sources **328**, 124 (2016).

[60]Dandan Chai, Haotian Yan, Xin Wang, Xiang Li, and Yongzhu Fu, Adv. Funct. Mater. (2023).

10 July 2025 20:08:58

AIP Publishing Applied Physics Reviews ACCEPTED MANUSCRIPT
This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.
PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0187154

[61]Y. F. Tian, S. J. Tan, Z. Y. Lu, D. X. Xu, H. X. Chen, C. H. Zhang, X. S. Zhang, G. Li, Y. M. Zhao, W. P. Chen, Q. Xu, R. Wen, J. Zhang, and Y. G. Guo, Angew Chem Int Ed Engl **62** (33), e202305988 (2023).

[62]Yan Zhao, Tianhong Zhou, Dominika Baster, Mario El Kazzi, Jang Wook Choi, and Ali Coskun, ACS Energy Lett. **8** (7), 3180 (2023).

[63]Xudong Peng, Tianshuai Wang, Bin Liu, Yiju Li, and Tianshou Zhao, Energy Environ. Sci. **15** (12), 5350 (2022).

[64]X. Fan and C. Wang, Chem. Soc. Rev. **50** (18), 10486 (2021).

[65]E. R. Logan and J. R. Dahn, Trends Chem. **2** (4), 354 (2020).

[66]H. Liang, Z. Ma, Y. Wang, F. Zhao, Z. Cao, L. Cavallo, Q. Li, and J. Ming, ACS Nano **17** (18), 18062 (2023).

[67]Y. Zou, Z. Ma, G. Liu, Q. Li, D. Yin, X. Shi, Z. Cao, Z. Tian, H. Kim, Y. Guo, C. Sun, L. Cavallo, L. Wang, H. N. Alshareef, Y. K. Sun, and J. Ming, Angew Chem Int Ed Engl **62** (8), e202216189 (2023).

[68]Xueying Zheng, Liqiang Huang, Xiaolu Ye, Junxi Zhang, Fengyuan Min, Wei Luo, and Yunhui Huang, Chem **7** (9), 2312 (2021).

[69]Qujiang Sun, Zhen Cao, Zheng Ma, Junli Zhang, Haoran Cheng, Xianrong Guo, Geon-Tae Park, Qian Li, Erqing Xie, Luigi Cavallo, Yang-Kook Sun, and Jun Ming, ACS Energy Lett. **7** (10), 3545 (2022).

[70]Pauline Jaumaux, Junru Wu, Devaraj Shanmukaraj, Yizhou Wang, Dong Zhou, Bing Sun, Feiyu Kang, Baohua Li, Michel Armand, and Guoxiu Wang, Adv. Funct. Mater. **31** (10) (2020).

[71]Rochelle Weber, Matthew Genovese, A. J. Louli, Samuel Hames, Cameron Martin, Ian G. Hill, and J. R. Dahn, Nat. Energy **4** (8), 683 (2019).

[72]Qujiang Sun, Zhen Cao, Zheng Ma, Junli Zhang, Wandi Wahyudi, Gang Liu, Haoran Cheng, Tao Cai, Erqing Xie, Luigi Cavallo, Qian Li, and Jun Ming, Adv. Funct. Mater. **33** (1), 2210292 (2022).

[73]Yeguo Zou, Gang Liu, Yuqi Wang, Qian Li, Zheng Ma, Dongming Yin, Yao Liang, Zhen Cao,

10 July 2025 20:08:58

AIP Publishing Applied Physics Reviews ACCEPTED MANUSCRIPT
This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.
PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0187154

Luigi Cavallo, Hun Kim, Limin Wang, Husam N. Alshareef, Yang-Kook Sun, and Jun Ming, Adv. Energy Mater. **13** (19) (2023).

[74]Gang Liu, Zhen Cao, Lin Zhou, Jiao Zhang, Qujiang Sun, Jang-Yeon Hwang, Luigi Cavallo, Limin Wang, Yang-Kook Sun, and Jun Ming, Adv. Funct. Mater. **30** (43) (2020).

[75]M. Qiu, P. Sun, K. Han, Z. Pang, J. Du, J. Li, J. Chen, Z. L. Wang, and W. Mai, Nat. Commun. **14** (1), 601 (2023).

[76]Q. Wang, C. Zhao, J. Wang, Z. Yao, S. Wang, S. G. H. Kumar, S. Ganapathy, S. Eustace, X. Bai, B. Li, and M. Wagemaker, Nat. Commun. **14** (1), 440 (2023).

[77]Q. Wang, C. Zhao, Z. Yao, J. Wang, F. Wu, S. G. H. Kumar, S. Ganapathy, S. Eustace, X. Bai, B. Li, J. Lu, and M. Wagemaker, Adv. Mater. **35** (17), e2210677 (2023).

[78]Chongyin Yang, Jiale Xia, Chunyu Cui, Travis P. Pollard, Jenel Vatamanu, Antonio Faraone, Joseph A. Dura, Madhusudan Tyagi, Alex Kattan, Elijah Thimsen, Jijian Xu, Wentao Song, Enyuan Hu, Xiao Ji, Singyuk Hou, Xiyue Zhang, Michael S. Ding, Sooyeon Hwang, Dong Su, Yang Ren, Xiao-Qing Yang, Howard Wang, Oleg Borodin, and Chunsheng Wang, Nat. Sustainability **6** (3), 325 (2023).

[79]Sang Cheol Kim, Jingyang Wang, Rong Xu, Pu Zhang, Yuelang Chen, Zhuojun Huang, Yufei Yang, Zhiao Yu, Solomon T. Oyakhire, Wenbo Zhang, Louisa C. Greenburg, Mun Sek Kim, David T. Boyle, Philaphon Sayavong, Yusheng Ye, Jian Qin, Zhenan Bao, and Yi Cui, Nat. Energy **8** (8), 814 (2023).

[80]Lequan Deng, Lutan Dong, Zhaofen Wang, Yaoyao Liu, Jun Zhan, Shuhua Wang, Ke-Peng Song, Dongqing Qi, Yuanhua Sang, Hong Liu, and Hao Chen, Adv. Energy Mater. (2023).

[81]J. Xu, V. Koverga, A. Phan, A. Min Li, N. Zhang, M. Baek, C. Jayawardana, B. L. Lucht, A. T. Ngo, and C. Wang, Adv. Mater., e2306462 (2023).

[82]Yingchun Xia, Pan Zhou, Xian Kong, Jiekang Tian, Weili Zhang, Shuaishuai Yan, Wen-hui Hou, Hang-Yu Zhou, Hao Dong, Xiaoxia Chen, Peican Wang, Ziang Xu, Lei Wan, Baoguo Wang, and Kai Liu, Nat. Energy **8** (9), 934 (2023).

[83]Rui Lin, Changming Ke, Juner Chen, Shi Liu, and Jianhui Wang, Joule **6** (2), 399 (2022).

[84]S. Yan, F. Liu, Y. Ou, H. Y. Zhou, Y. Lu, W. Hou, Q. Cao, H. Liu, P. Zhou, and K. Liu, ACS

10 July 2025 20:08:58

AIP Publishing Applied Physics Reviews ACCEPTED MANUSCRIPT
This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.
PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0187154

Nano **17** (19), 19398 (2023).

[85]G. Xu, X. Shangguan, S. Dong, X. Zhou, and G. Cui, Angew Chem Int Ed Engl **59** (9), 3400 (2020).

[86]A. J. Louli, A. Eldesoky, Rochelle Weber, M. Genovese, Matt Coon, Jack deGooyer, Zhe Deng, R. T. White, Jaehan Lee, Thomas Rodgers, R. Petibon, S. Hy, Shawn J. H. Cheng, and J. R. Dahn, Nat. Energy **5** (9), 693 (2020).

[87]Fanglin Wu, Shan Fang, Matthias Kuenzel, Angelo Mullaliu, Jae-Kwang Kim, Xinpei Gao, Thomas Diemant, Guk-Tae Kim, and Stefano Passerini, Joule **5** (8), 2177 (2021).

[88]Darren M. Driscoll, Sydney N. Lavan, Milena Zorko, Paul C. Redfern, Stefan Ilic, Garvit Agarwal, Timothy T. Fister, Rajeev S. Assary, Lei Cheng, Dusan Strmcnik, Mahalingam Balasubramanian, and Justin G. Connell, Chem **9** (7), 1955 (2023).

[89]Tao Cai, Qujiang Sun, Zhen Cao, Zheng Ma, Wandi Wahyudi, Luigi Cavallo, Qian Li, and Jun Ming, The Journal of Physical Chemistry C **126** (48), 20302 (2022).

[90]G. Liu, Z. Cao, P. Wang, Z. Ma, Y. Zou, Q. Sun, H. Cheng, L. Cavallo, S. Li, Q. Li, and J. Ming, Adv Sci (Weinh) **9** (26), e2201893 (2022).

[91]Wandi Wahyudi, Viko Ladelta, Leonidas Tsetseris, Merfat M. Alsabban, Xianrong Guo, Emre Yengel, Hendrik Faber, Begimai Adilbekova, Akmaral Seitkhan, Abdul-Hamid Emwas, Mohammed N. Hedhili, Lain-Jong Li, Vincent Tung, Nikos Hadjichristidis, Thomas D. Anthopoulos, and Jun Ming, Adv. Funct. Mater. **31** (23) (2021).

[92]F. Ming, Y. Zhu, G. Huang, A. H. Emwas, H. Liang, Y. Cui, and H. N. Alshareef, J. Am. Chem. Soc. **144** (16), 7160 (2022).

[93]D. Aurbach, E. Markevich, and G. Salitra, J. Am. Chem. Soc. **143** (50), 21161 (2021).

[94]Jing Li, Hongyang Li, Xiaowei Ma, Will Stone, Stephen Glazier, Eric Logan, Erin Marie Tonita, Kevin L. Gering, and J. R. Dahn, J. Electrochem. Soc. **165** (5), A1027 (2018).

[95]David S. Hall, Ahmed Eldesoky, E. R. Logan, Erin Marie Tonita, Xiaowei Ma, and J. R. Dahn, J. Electrochem. Soc. **165** (10), A2365 (2018).

[96]X. Cao, P. Gao, X. Ren, L. Zou, M. H. Engelhard, B. E. Matthews, J. Hu, C. Niu, D. Liu, B. W. Arey, C. Wang, J. Xiao, J. Liu, W. Xu, and J. G. Zhang, Proc Natl Acad Sci U S A **118**

10 July 2025 20:08:58

AIP Publishing Applied Physics Reviews ACCEPTED MANUSCRIPT
This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.
PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0187154

(9) (2021).

[97]Chunsheng Wang, Ying Shirley Meng, and Kang Xu, J. Electrochem. Soc. **166** (3), A5184 (2018).

[98]Q. Zhang, Nat. Nanotechnol. **13** (8), 623 (2018).

[99]Zhiao Yu, Hansen Wang, Xian Kong, William Huang, Yuchi Tsao, David G. Mackanic, Kecheng Wang, Xinchang Wang, Wenxiao Huang, Snehashis Choudhury, Yu Zheng, Chibueze V. Amanchukwu, Samantha T. Hung, Yuting Ma, Eder G. Lomeli, Jian Qin, Yi Cui, and Zhenan Bao, Nat. Energy **5** (7), 526 (2020).

[100]Zhiao Yu, Paul E. Rudnicki, Zewen Zhang, Zhuojun Huang, Hasan Celik, Solomon T. Oyakhire, Yuelang Chen, Xian Kong, Sang Cheol Kim, Xin Xiao, Hansen Wang, Yu Zheng, Gaurav A. Kamat, Mun Sek Kim, Stacey F. Bent, Jian Qin, Yi Cui, and Zhenan Bao, Nat. Energy **7** (1), 94 (2022).

[101]Ronnie Mogensen, Alexander Buckel, Simon Colbin, and Reza Younesi, Chem. Mater. **33** (4), 1130 (2021).

[102]Benjamin A. Paren, Bryce A. Thurston, Arjun Kanthawar, William J. Neary, Aaron Kendrick, Manuel Maréchal, Justin G. Kennemur, Mark J. Stevens, Amalie L. Frischknecht, and Karen I. Winey, Chem. Mater. **33** (15), 6041 (2021).

[103]C. Yang, L. Suo, O. Borodin, F. Wang, W. Sun, T. Gao, X. Fan, S. Hou, Z. Ma, K. Amine, K. Xu, and C. Wang, Proc Natl Acad Sci U S A **114** (24), 6197 (2017).

[104]Yuki Yamada, Jianhui Wang, Seongjae Ko, Eriko Watanabe, and Atsuo Yamada, Nat. Energy **4** (4), 269 (2019).

[105]H. Jia, J. M. Kim, P. Gao, Y. Xu, M. H. Engelhard, B. E. Matthews, C. Wang, and W. Xu, Angew Chem Int Ed Engl **62** (17), e202218005 (2023).

[106]Xiaodi Ren, Shuru Chen, Hongkyung Lee, Donghai Mei, Mark H. Engelhard, Sarah D. Burton, Wengao Zhao, Jianming Zheng, Qiuyan Li, Michael S. Ding, Marshall Schroeder, Judith Alvarado, Kang Xu, Y. Shirley Meng, Jun Liu, Ji-Guang Zhang, and Wu Xu, Chem **4** (8), 1877 (2018).

[107]Hui Dong, Oscar Tutusaus, Yanliang Liang, Ye Zhang, Zachary Lebens-Higgins, Wanli Yang,

10 July 2025 20:08:58

AIP Publishing Applied Physics Reviews ACCEPTED MANUSCRIPT
This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.
PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0187154

Rana Mohtadi, and Yan Yao, Nat. Energy **5** (12), 1043 (2020).

[108]X. Gao, Y. Chen, L. Johnson, and P. G. Bruce, Nat. Mater. **15** (8), 882 (2016).

[109]T. Ma, Y. Ni, Q. Wang, W. Zhang, S. Jin, S. Zheng, X. Yang, Y. Hou, Z. Tao, and J. Chen, Angew Chem Int Ed Engl **61** (39), e202207927 (2022).

[110]Pu Yang, Xiangwen Gao, Xiaolu Tian, Chengyong Shu, Yikun Yi, Pei Liu, Te Wang, Long Qu, Bingbing Tian, Mingtao Li, Wei Tang, Bolun Yang, and John B. Goodenough, ACS Energy Lett. **5** (5), 1681 (2020).

[111]Chunxiao Zhang, Zheng Lu, Miao Song, Youquan Zhang, Chuyang Jing, Libao Chen, Xiaobo Ji, and Weifeng Wei, Adv. Energy Mater. **13** (21), 2203870 (2023).

[112]C. L. Li, G. Huang, Y. Yu, Q. Xiong, J. M. Yan, and X. B. Zhang, J. Am. Chem. Soc. **144** (13), 5827 (2022).

[113]Wuhai Yang, Xiaofan Du, Jingwen Zhao, Zheng Chen, Jiajia Li, Jian Xie, Yaojian Zhang, Zili Cui, Qingyu Kong, Zhiming Zhao, Cunguo Wang, Qichun Zhang, and Guanglei Cui, Joule **4** (7), 1557 (2020).

[114]C. Zhang, H. Chen, Y. Qian, G. Dai, Y. Zhao, and G. Yu, Adv. Mater. **33** (15), e2008560 (2021).

[115]An L. Phan, Chamithri Jayawardana, Phung M. L. Le, Jiaxun Zhang, Bo Nan, Weiran Zhang, Brett L. Lucht, Singyuk Hou, and Chunsheng Wang, Adv. Funct. Mater. **33** (34), 2301177 (2023).

[116]Guangtao Cong and Yi-Chun Lu, Chem **4** (12), 2732 (2018).

[117]Tao Xuan and Liwei Wang, Energy Storage Mater. **48**, 263 (2022).

[118]Kun Qian, Soenke Seifert, Randall E. Winans, and Tao Li, Energy & Fuels **35** (23), 19849 (2021).

[119]Xinyi Liu, Lingzhe Fang, Xingyi Lyu, Randall E. Winans, and Tao Li, Chem. Mater. **35** (23), 9821 (2023).

[120]Jiangtao Hu, Yuchen Ji, Guorui Zheng, Weiyuan Huang, Yuan Lin, Luyi Yang, and Feng Pan, Aggregate **3** (1), e153 (2022).

[121]Qujiang Sun, Zhen Cao, Zheng Ma, Junli Zhang, Wandi Wahyudi, Tao Cai, Haoran Cheng,

10 July 2025 20:08:58

AIP Publishing Applied Physics Reviews ACCEPTED MANUSCRIPT
This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.
PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0187154

Qian Li, Hun Kim, Erqing Xie, Luigi Cavallo, Yang-Kook Sun, and Jun Ming, ACS Mater. Lett. **4** (11), 2233 (2022).

[122]Yuqi Wang, Zhen Cao, Zheng Ma, Gang Liu, Haoran Cheng, Yeguo Zou, Luigi Cavallo, Qian Li, and Jun Ming, ACS Energy Lett. **8** (3), 1477 (2023).

[123]W. Wahyudi, X. Guo, V. Ladelta, L. Tsetseris, M. I. Nugraha, Y. Lin, V. Tung, N. Hadjichristidis, Q. Li, K. Xu, J. Ming, and T. D. Anthopoulos, Adv Sci (Weinh) **9** (28), e2202405 (2022).

[124]E. W. Zhao, T. Liu, E. Jonsson, J. Lee, I. Temprano, R. B. Jethwa, A. Wang, H. Smith, J. Carretero-Gonzalez, Q. Song, and C. P. Grey, Nature **579** (7798), 224 (2020).

[125]E. W. Zhao, E. Jonsson, R. B. Jethwa, D. Hey, D. Lyu, A. Brookfield, P. A. A. Klusener, D. Collison, and C. P. Grey, J. Am. Chem. Soc. **143** (4), 1885 (2021).

[126]J. Fawdon, J. Ihli, F. Mantia, and M. Pasta, Nat. Commun. **12** (1), 4053 (2021).

[127]E. Miele, W. M. Dose, I. Manyakin, M. H. Frosz, Z. Ruff, M. F. L. De Volder, C. P. Grey, J. J. Baumberg, and T. G. Euser, Nat. Commun. **13** (1), 1651 (2022).

[128]Zhange Feng, Erik Sarnello, Tao Li, and Lei Cheng, J. Electrochem. Soc. **166** (2), A47 (2019).

[129]Kun Qian, Yuzi Liu, Xinwei Zhou, David J. Gosztola, Hoai Nguyen, and Tao Li, Energy Storage Mater. **41**, 222 (2021).

[130]Kun Qian, Zhou Yu, Yuzi Liu, David J. Gosztola, Randall E. Winans, Lei Cheng, and Tao Li, J. Energy Chem. **70**, 340 (2022).

[131]Xinyi Liu, Shao-Chun Lee, Soenke Seifer, Randall E. Winans, Lei Cheng, Y. Z, and Tao Li, Energy Storage Mater. **45**, 696 (2022).

[132]Xinyi Liu, Zhou Yu, Erik Sarnello, Kun Qian, Soenke Seifert, Randall E. Winans, Lei Cheng, and Tao Li, Energy Mater. Adv. **2021**, 1 (2021).

[133]Pan Tan, Jinming Yue, Yuanxi Yu, Binghang Liu, Tao Liu, Lirong Zheng, Lunhua He, Xiaohua Zhang, Liumin Suo, and Liang Hong, The Journal of Physical Chemistry C **125** (22), 11838 (2021).

[134]Xinyi Liu, Shao-Chun Lee, Soenke Seifert, Randall E. Winans, Y. Z, and Tao Li, Chem.

10 July 2025 20:08:58

AIP Publishing Applied Physics Reviews ACCEPTED MANUSCRIPT
This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.
PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0187154

Mater. **35** (16), 6415 (2023).

[135]Laisuo Su, Xunhua Zhao, Michael Yi, Harry Charalambous, Hugo Celio, Yuanyue Liu, and Arumugam Manthiram, Adv. Energy Mater. **12** (36), 2201911 (2022).

[136]K. Sharma, S. Sankarasubramanian, J. Parrondo, and V. Ramani, Proc Natl Acad Sci U S A **118** (34) (2021).

[137]Jing Yang, Hui Yan, Huanhuan Hao, Yuxi Song, Ying Li, Qinghua Liu, and Ao Tang, ACS Energy Lett. **7** (7), 2331 (2022).

[138]Kang Peng, Yuanyuan Li, Gonggen Tang, Yahua Liu, Zhengjin Yang, and Tongwen Xu, Energy Environ. Sci. **16** (2), 430 (2023).

[139]Yuyue Zhao, Zhou Yu, Lily A. Robertson, Jingjing Zhang, Zhangxing Shi, Sambasiva R. Bheemireddy, Ilya A. Shkrob, Z. Y, Tao Li, Zhengcheng Zhang, Lei Cheng, and Lu Zhang, J. Mater. Chem. A **8** (27), 13470 (2020).

[140]I. A. Shkrob, T. Li, E. Sarnello, L. A. Robertson, Y. Zhao, H. Farag, Z. Yu, J. Zhang, S. R. Bheemireddy, Y. Z, R. S. Assary, R. H. Ewoldt, L. Cheng, and L. Zhang, J. Phys. Chem. B **124** (45), 10226 (2020).

[141]Y. Zhao, E. S. Sarnello, L. A. Robertson, J. Zhang, Z. Shi, Z. Yu, S. R. Bheemireddy, Y. Z, T. Li, R. S. Assary, L. Cheng, Z. Zhang, L. Zhang, and I. A. Shkrob, J. Phys. Chem. B **124** (46), 10409 (2020).

[142]H. Farag, A. P. Kaur, L. A. Robertson, E. Sarnello, X. Liu, Y. Wang, L. Cheng, I. A. Shkrob, L. Zhang, R. H. Ewoldt, T. Li, S. A. Odom, and Z. Y, Phys. Chem. Chem. Phys. **25** (5), 4243 (2023).

[143]Ankit Agrawal and Alok Choudhary, APL Mater. **4** (5), 053208 (2016).

[144]X. Peng and X. Wang, MRS Bull. **48** (2), 179 (2023).

[145]Anubhav Jain, Yongwoo Shin, and Kristin A. Persson, Nat. Rev. Mater. **1** (1), 15004 (2016).

[146]A. Y. S. Eng, C. B. Soni, Y. Lum, E. Khoo, Z. Yao, S. K. Vineeth, V. Kumar, J. Lu, C. S. Johnson, C. Wolverton, and Z. W. Seh, Sci. Adv. **8** (19), eabm2422 (2022).

[147]Yun Liu, Oladapo Christopher Esan, Zhefei Pan, and Liang An, Energy AI **3**, 100049 (2021).

10 July 2025 20:08:58

AIP Publishing Applied Physics Reviews ACCEPTED MANUSCRIPT
This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.
PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0187154

[148]Tian Xie, Massachusetts Institute of Technology, 2020.

[149]T. Lombardo, M. Duquesnoy, H. El-Bouysidy, F. Aren, A. Gallo-Bueno, P. B. Jorgensen, A. Bhowmik, A. Demortiere, E. Ayerbe, F. Alcaide, M. Reynaud, J. Carrasco, A. Grimaud, C. Zhang, T. Vegge, P. Johansson, and A. A. Franco, Chem. Rev. **122** (12), 10899 (2022).

[150]Austin D. Sendek, Brandi Ransom, Ekin D. Cubuk, Lenson A. Pellouchoud, Jagjit Nanda, and Evan J. Reed, Adv. Energy Mater. **12** (31), 2200553 (2022).

[151]Zhichu Ren, Zekun Ren, Zhen Zhang, Tonio Buonassisi, and Ju Li, Nat. Rev. Mater. **8** (9), 563 (2023).

[152]Yunchao Xie, Kianoosh Sattari, Chi Zhang, and Jian Lin, Prog. Mater Sci. **132**, 101043 (2023).

[153]John M. Gregoire, Lan Zhou, and Joel A. Haber, Nat. Synth. **2** (6), 493 (2023).

[154]J. Yano, K. J. Gaffney, J. Gregoire, L. Hung, A. Ourmazd, J. Schrier, J. A. Sethian, and F. M. Toma, Nat. Rev. Chem. **6** (5), 357 (2022).

[155]Natalie S. Eyke, Brent A. Koscher, and Klavs F. Jensen, Trends Chem. **3** (2), 120 (2021).

[156]Milad Abolhasani and Keith A. Brown, MRS Bull. **48** (2), 134 (2023).

[157]T. He, H. Huo, C. J. Bartel, Z. Wang, K. Cruse, and G. Ceder, Sci. Adv. **9** (23), eadg8180 (2023).

[158]V. Tshitoyan, J. Dagdelen, L. Weston, A. Dunn, Z. Rong, O. Kononova, K. A. Persson, G. Ceder, and A. Jain, Nature **571** (7763), 95 (2019).

[159]Zheren Wang, Kevin Cruse, Yuxing Fei, Ann Chia, Yan Zeng, Haoyan Huo, Tanjin He, Bowen Deng, Olga Kononova, and Gerbrand Ceder, Digital Discovery **1** (3), 313 (2022).

[160]Z. Wang, O. Kononova, K. Cruse, T. He, H. Huo, Y. Fei, Y. Zeng, Y. Sun, Z. Cai, W. Sun, and G. Ceder, Sci. Data **9** (1), 231 (2022).

[161]N. J. Szymanski, Y. Zeng, H. Huo, C. J. Bartel, H. Kim, and G. Ceder, Mater. Horiz. **8** (8), 2169 (2021).

[162]H. Huo, C. J. Bartel, T. He, A. Trewartha, A. Dunn, B. Ouyang, A. Jain, and G. Ceder, Chem. Mater. **34** (16), 7323 (2022).

[163]Haoyan Huo, Ziqin Rong, Olga Kononova, Wenhao Sun, Tiago Botari, Tanjin He, Vahe

10 July 2025 20:08:58

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Tshitoyan, and Gerbrand Ceder, npj Comput. Mater. **5** (1), 62 (2019).

[164]Nathan J. Szymanski, Christopher J. Bartel, Yan Zeng, Qingsong Tu, and Gerbrand Ceder, Chem. Mater. **33** (11), 4204 (2021).

[165]N. J. Szymanski, P. Nevatia, C. J. Bartel, Y. Zeng, and G. Ceder, Nat. Commun. **14** (1), 6956 (2023).

[166]P. V. Balachandran, D. Xue, J. Theiler, J. Hogden, and T. Lookman, Sci. Rep. **6**, 19660 (2016).

[167]Adarsh Dave, Jared Mitchell, Kirthevasan Kandasamy, Han Wang, Sven Burke, Biswajit Paria, Barnabás Póczos, Jay Whitacre, and Venkatasubramanian Viswanathan, Cell Rep. Phys. Sci. **1** (12), 100264 (2020).

[168]Anass Benayad, Diddo Diddens, Andreas Heuer, Anand Narayanan Krishnamoorthy, Moumita Maiti, Frédéric Le Cras, Maxime Legallais, Fuzhan Rahmanian, Yuyoung Shin, Helge Stein, Martin Winter, Christian Wölke, Peng Yan, and Isidora Cekic-Laskovic, Adv. Energy Mater. **12** (17), 2102678 (2021).

[169]Jaime Rodriguez, Maria Politi, Stuart Adler, David Beck, and Lilo Pozzo, Mol. Syst. Des. Eng. **7** (8), 933 (2022).

[170]Kedar Hippalgaonkar, Qianxiao Li, Xiaonan Wang, John W. Fisher, James Kirkpatrick, and Tonio Buonassisi, Nat. Rev. Mater. **8** (4), 241 (2023).

[171]A. Dave, J. Mitchell, S. Burke, H. Lin, J. Whitacre, and V. Viswanathan, Nat. Commun. **13** (1), 5454 (2022).

[172]Jay F. Whitacre, Jared Mitchell, Adarsh Dave, Wei Wu, Sven Burke, and Venkatasubramanian Viswanathan, J. Electrochem. Soc. **166** (16), A4181 (2019).

[173]Per H. Svensson, Pavel Yushmanov, Aleksandar Tot, Lars Kloo, Erik Berg, and Kristina Edström, Chem. Eng. J. **455**, 140955 (2023).

[174]Jackie T. Yik, Leiting Zhang, Jens Sjölund, Xu Hou, Per H. Svensson, Kristina Edström, and Erik J. Berg, Digital Discovery **2** (3), 799 (2023).

[175]N. Yao, X. Chen, Z. H. Fu, and Q. Zhang, Chem. Rev. **122** (12), 10970 (2022).

[176]O. T. Unke, S. Chmiela, H. E. Sauceda, M. Gastegger, I. Poltavsky, K. T. Schutt, A.

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Tkatchenko, and K. R. Muller, Chem. Rev. **121** (16), 10142 (2021).

[177]P. Friederich, F. Hase, J. Proppe, and A. Aspuru-Guzik, Nat. Mater. **20** (6), 750 (2021).

[178]T. Zubatiuk and O. Isayev, Acc. Chem. Res. **54** (7), 1575 (2021).

[179]S. C. Lee, Y. Zhai, Z. Li, N. P. Walter, M. Rose, B. J. Heuser, and Y. Z, J. Phys. Chem. B **125** (37), 10562 (2021).

[180]H. Guo, Q. Wang, A. Urban, and N. Artrith, Chem. Mater. **34** (15), 6702 (2022).

[181]Ioan-Bogdan Magdău, Daniel J. Arismendi-Arrieta, Holly E. Smith, Clare P. Grey, Kersti Hermansson, and Gábor Csányi, npj Comput. Mater. **9** (1), 146 (2023).

[182]F. Li, X. Cheng, L. L. Lu, Y. C. Yin, J. D. Luo, G. Lu, Y. F. Meng, H. Mo, T. Tian, J. T. Yang, W. Wen, Z. P. Liu, G. Zhang, C. Shang, and H. B. Yao, Nano Lett. **22** (6), 2461 (2022).

[183]F. Wang and J. Cheng, Chem. Sci. **13** (39), 11570 (2022).

[184]F. Wang, Y. Sun, and J. Cheng, J. Am. Chem. Soc. **145** (7), 4056 (2023).

[185]Yanming Wang, Tian Xie, Arthur France-Lanord, Arthur Berkley, Jeremiah A. Johnson, Yang Shao-Horn, and Jeffrey C. Grossman, Chem. Mater. **32** (10), 4144 (2020).

[186]T. Xie, A. France-Lanord, Y. Wang, Y. Shao-Horn, and J. C. Grossman, Nat. Commun. **10** (1), 2667 (2019).

[187]L. Cheng, R. S. Assary, X. Qu, A. Jain, S. P. Ong, N. N. Rajput, K. Persson, and L. A. Curtiss, J. Phys. Chem. Lett. **6** (2), 283 (2015).

[188]Juan-Pablo Correa-Baena, Kedar Hippalgaonkar, Jeroen van Duren, Shaffiq Jaffer, Vijay R. Chandrasekhar, Vladan Stevanovic, Cyrus Wadia, Supratik Guha, and Tonio Buonassisi, Joule **2** (8), 1410 (2018).

[189]K. T. Butler, D. W. Davies, H. Cartwright, O. Isayev, and A. Walsh, Nature **559** (7715), 547 (2018).

[190]Chen Ling, npj Comput. Mater. **8** (1), 33 (2022).

[191]Evan Walter Clark Spotte-Smith, Orion Archer Cohen, Samuel M. Blau, Jason M. Munro, Ruoxi Yang, Rishabh D. Guha, Hetal D. Patel, Sudarshan Vijay, Patrick Huck, Ryan Kingsbury, Matthew K. Horton, and Kristin A. Persson, Digital Discovery **2** (6), 1862 (2023).

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[192]Gonçalo dos Reis, Calum Strange, Mohit Yadav, and Shawn Li, Energy AI **5**, 100081 (2021).

[193]Logan Ward, Susan Babinec, Eric J. Dufek, David A. Howey, Venkatasubramanian Viswanathan, Muratahan Aykol, David A. C. Beck, Benjamin Blaiszik, Bor-Rong Chen, George Crabtree, Simon Clark, Valerio De Angelis, Philipp Dechent, Matthieu Dubarry, Erica E. Eggleton, Donal P. Finegan, Ian Foster, Chirranjeevi Balaji Gopal, Patrick K. Herring, Victor W. Hu, Noah H. Paulson, Yuliya Preger, Dirk Uwe-Sauer, Kandler Smith, Seth W. Snyder, Shashank Sripad, Tanvir R. Tanim, and Linnette Teo, Joule **6** (10), 2253 (2022).

[194]Y. C. Gao, N. Yao, X. Chen, L. Yu, R. Zhang, and Q. Zhang, J. Am. Chem. Soc. **145** (43), 23764 (2023).

[195]Yanzhou Wu, Qiao Hu, Hongmei Liang, Aiping Wang, Hong Xu, Li Wang, and Xiangming He, Adv. Energy Mater. **13** (22), 2300259 (2023).

[196]Forrest A. L. Laskowski, Daniel B. McHaffie, and Kimberly A. See, Energy Environ. Sci. **16** (3), 1264 (2023).

[197]Z. Ahmad, T. Xie, C. Maheshwari, J. C. Grossman, and V. Viswanathan, ACS Cent. Sci. **4** (8), 996 (2018).

[198]K. Hatakeyama-Sato, T. Tezuka, M. Umeki, and K. Oyaizu, J. Am. Chem. Soc. **142** (7), 3301 (2020).

[199]T. Xie, A. France-Lanord, Y. Wang, J. Lopez, M. A. Stolberg, M. Hill, G. M. Leverick, R. Gomez-Bombarelli, J. A. Johnson, Y. Shao-Horn, and J. C. Grossman, Nat. Commun. **13** (1), 3415 (2022).

[200]G. Bradford, J. Lopez, J. Ruza, M. A. Stolberg, R. Osterude, J. A. Johnson, R. Gomez-Bombarelli, and Y. Shao-Horn, ACS Cent. Sci. **9** (2), 206 (2023).

[201]Y. J. Wu, T. Tanaka, T. Komori, M. Fujii, H. Mizuno, S. Itoh, T. Takada, E. Fujita, and Y. Xu, Sci. Technol. Adv. Mater. **21** (1), 712 (2020).

[202]Aashutosh Mistry, Ankit Verma, Shashank Sripad, Rebecca Ciez, Valentin Sulzer, Ferran Brosa Planella, Robert Timms, Yumin Zhang, Rachel Kurchin, Philipp Dechent, Weihan Li, Samuel Greenbank, Zeeshan Ahmad, Dilip Krishnamurthy, Alexis M. Fenton, Kevin Tenny,

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[203] Anthony Yu-Tung Wang, Ryan J. Murdock, Steven K. Kauwe, Anton O. Oliynyk, Aleksander Gurlo, Jakoah Brgoch, Kristin A. Persson, and Taylor D. Sparks, Chem. Mater. **32** (12), 4954 (2020).

10 July 2025 20:08:58