Applied Physics Reviews

REVIEW ARTICLE | APRIL 12 2024

Exploring solvation structure and transport behavior for rational design of advanced electrolytes for next generation of lithium batteries

Special Collection: Frontiers in energy materials research: novel measurement, modelling and processing approaches

Xiaozhao Liu; Volodymyr Koverga; Hoai T. Nguyen; Anh T. Ngo; Tao Li

Check for updates

Appl. Phys. Rev. 11, 021307 (2024)
https://doi.org/10.1063/5.0187154

View Online
Export Citation

02 July 2024 12:36:38

Journal of Applied Physics
Special Topic:
Disordered Materials at the Atomic Scale
Guest Editors: Jaeyun Moon, Matteo Baggioli
Submit Today!
AIP Publishing

AIP Publishing

Applied Physics Reviews REVIEW pubs.aip.org/aip/are

Exploring solvation structure and transport behavior for rational design of advanced electrolytes for next generation of lithium batteries

Cite as: Appl. Phys. Rev. **11**, 021307 (2024); doi: 10.1063/5.0187154
Submitted: 11 November 2023 · Accepted: 22 February 2024 ·
Published Online: 12 April 2024

Xiaozhao Liu,[1] Volodymyr Koverga,[2,3] Hoai T. Nguyen,[4] Anh T. Ngo,[2,3,a)] and Tao Li[1,5,a)]

AFFILIATIONS

[1]Department of Chemistry and Biochemistry, Northern Illinois University, DeKalb, Illinois 60115, USA
[2]Department of Chemical Engineering, University of Illinois-Chicago, Chicago, Illinois 60608, USA
[3]Materials Science Division, Argonne National Laboratory, Lemont, Illinois 60439, USA
[4]Chemical Sciences and Engineering division, Argonne National Laboratory, Lemont, Illinois 60439, USA
[5]X-ray Science Division and Joint Center for Energy Storage Research, Argonne National Laboratory, Lemont, Illinois 60439, USA

Note: This paper is part of the APR Special Topic on Frontiers in energy materials research: novel measurement, modelling and processing approaches.

[a)]**Authors to whom correspondence should be addressed:** tli4@niu.edu and anhngo@uic.edu

02 July 2024 12:36:38

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 the recent advancements in electrolyte design strategies for high capacity, high-voltage, wide-temperature, fast-charging, and safe applications. To begin, the current state-of-the-art electrolyte design directions are comprehensively reviewed. Subsequently, advanced techniques and computational methods 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.

Published under an exclusive license by AIP Publishing. https://doi.org/10.1063/5.0187154

TABLE OF CONTENTS

I. INTRODUCTION 1
II. ELECTROLYTE DESIGN STRATEGIES FOR DIVERSE APPLICATIONS 4
III. UNDERSTANDING THE SOLVATION STRUCTURE AND TRANSPORT BEHAVIOR OF ELECTROLYTES 5
IV. AI-DRIVEN HIGH-THROUGHPUT SCREENING AND DATA-DRIVEN DESIGN OF ELECTROLYTES. 9
  A. Autonomous search and optimization........... 9
  B. AI-powered quantum chemical calculations and molecular dynamics simulations 10
  C. Data-driven methods 11
V. SUMMARY AND OUTLOOK 12

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 [Fig. 1(a)].[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

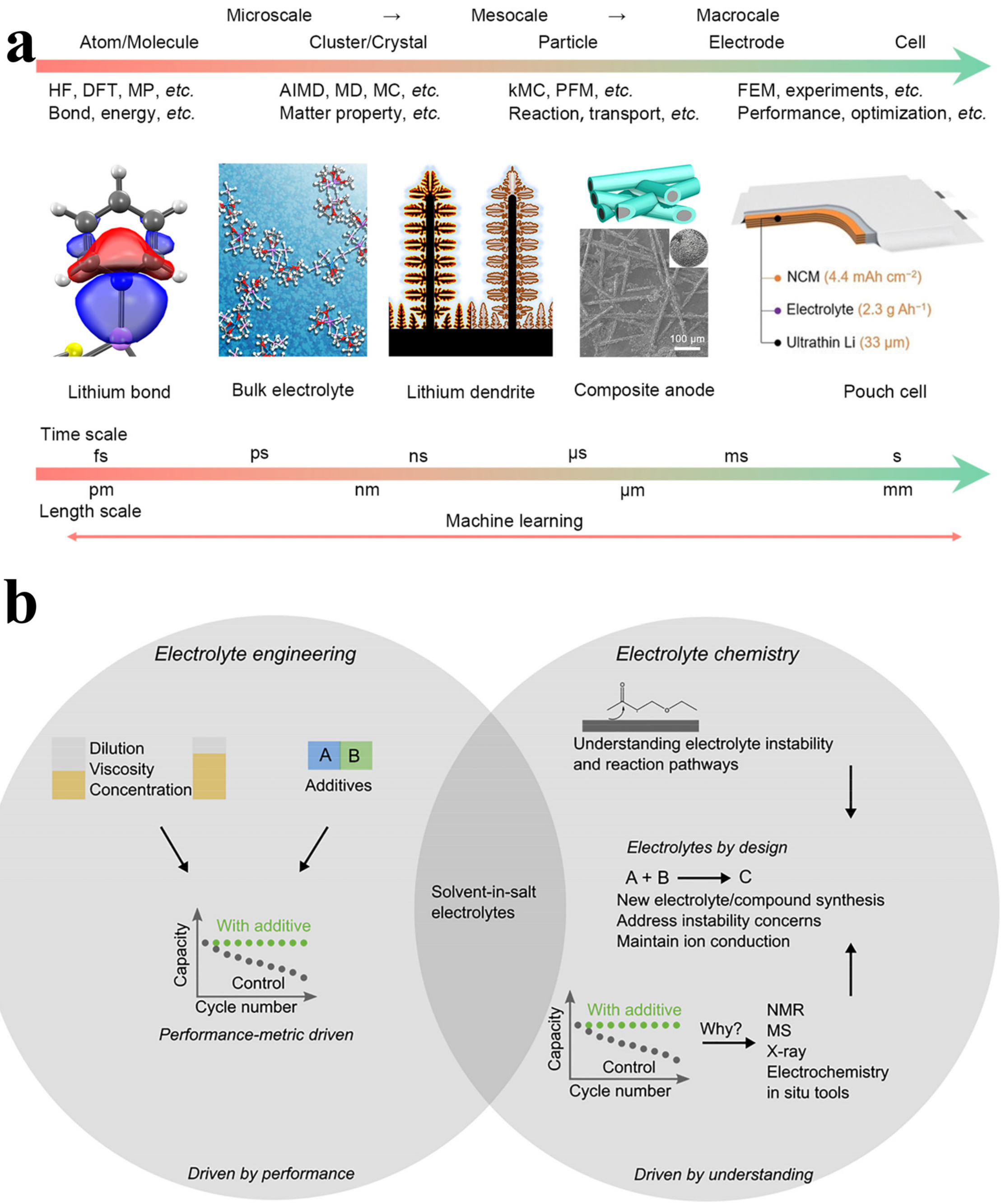

FIG. 1. (a) Different length and time scales for battery research.[18] Reproduced with permission from Chen *et al.*, Angew. Chem., Int. Ed. Engl. **60**, 24354–24366 (2021). Copyright 2021 Wiley. (b) Electrolyte engineering and chemistry.[19] Reproduced with permission from Amanchukwu, Joule **4**, 281–285 (2020). Copyright 2019 Elsevier.

02 July 2024 12:36:38

Published under an exclusive license by AIP Publishing

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.

Two primary considerations emerge in electrolytes design strategies [Fig. 1(b)].[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) [Fig. 2(a)].[20] Regarding cation diffusion, two primary mechanisms exist [Fig. 2(b)]: 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]

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) [Fig. 2(c)]. 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] 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,

02 July 2024 12:36:38

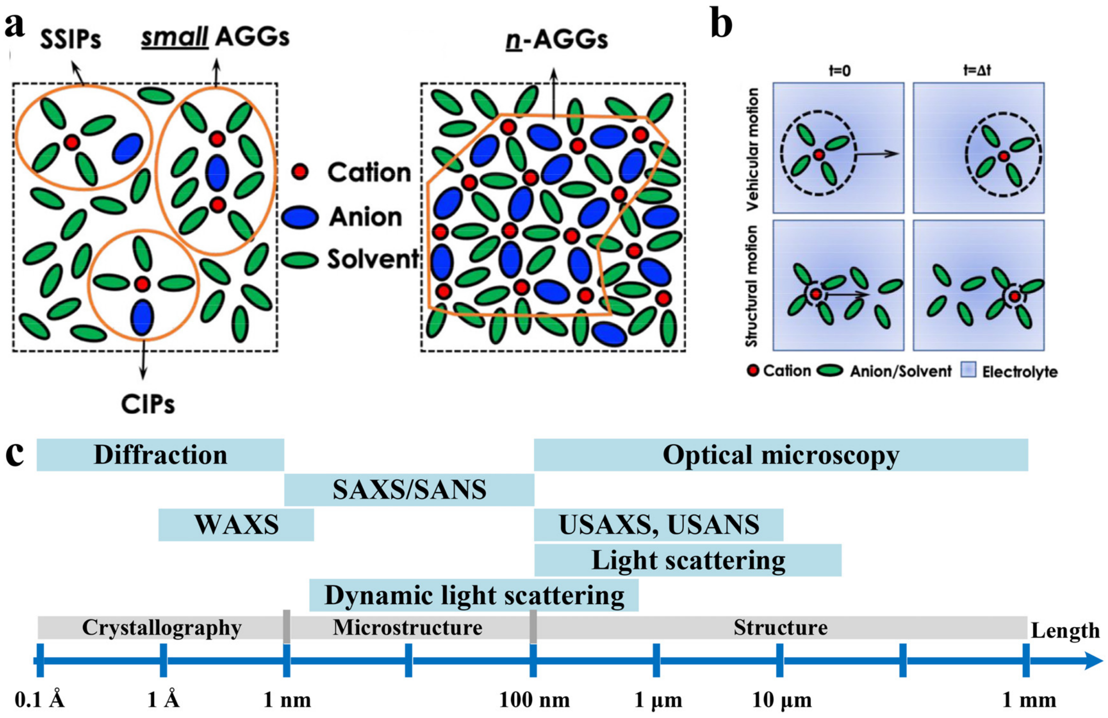

FIG. 2. (a) Schematic of SSIPs, CIPs, and AGGs. (b) Illustration of vehicular and structural motion modes.[20] Reproduced with permission from Yu *et al.*, ACS Energy Lett. **7**, 461–470 (2022). Copyright 2022 American Chemical Society. (c) Different research scales in electrolyte structure analysis.

literatures, or datasets on open-sharing websites. Unlike traditional tedious trial-and-error methods in materials science, artificial intelligence (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 [Fig. 3(a)]. Electrolytes encompass aqueous electrolytes, organic liquid 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 conditions.[44–46] Quasi-solid electrolytes refer to SSEs with added small amounts of organic liquid solvents, 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.

First, redox flow batteries (RFBs) present a promising option for stationary energy storage devices due to their unique advantages,

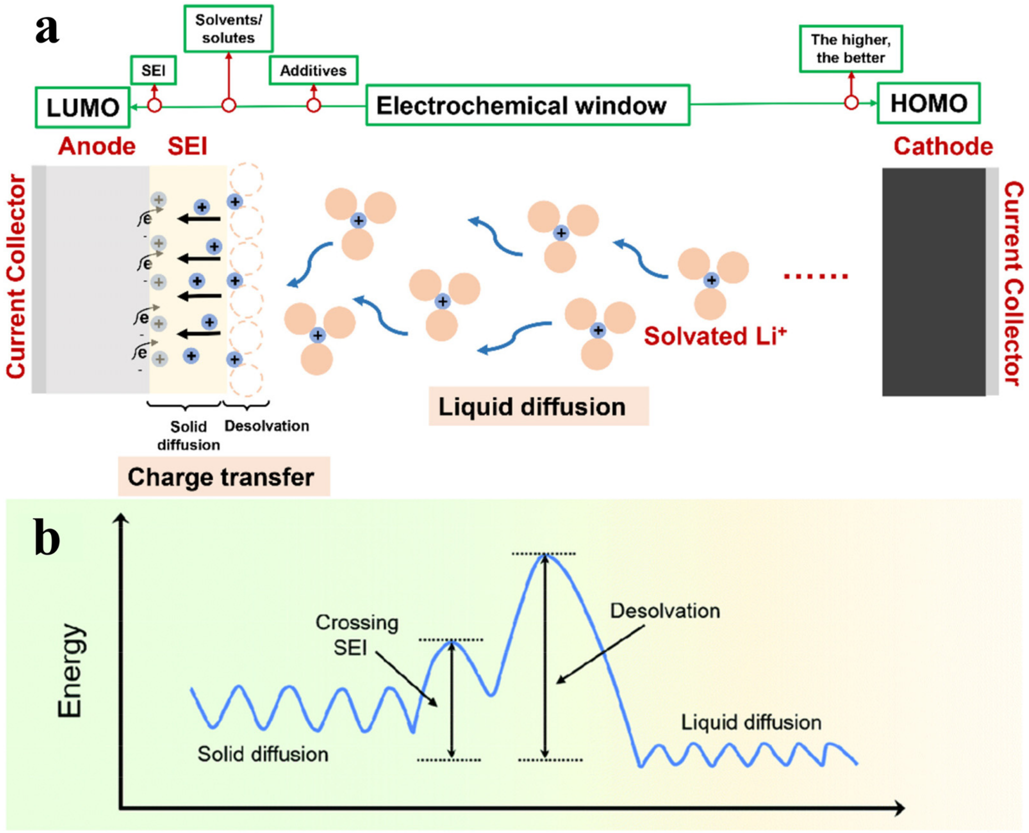

FIG. 3. (a) Thermodynamics and kinetics of electrolytes.[51] Reproduced with permission from Yang *et al.*, Energy Storage Mater. **30**, 113–129 (2020). Copyright 2020 Elsevier. (b) The energy diagram of charging process of graphite in Lithium-ion batteries.[52] Reproduced with permission from Cai *et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

02 July 2024 12:36:38

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, organic solvents, and redox mediators.[54] These 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 [Fig. 3(b)].[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 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]

Finally, 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] 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 (>5 nm), 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 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

02 July 2024 12:36:38

Published under an exclusive license by AIP Publishing

understanding of electrolyte evolution during cycling, thereby enabling the development of improved electrochemical systems.

Our group[128] has 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

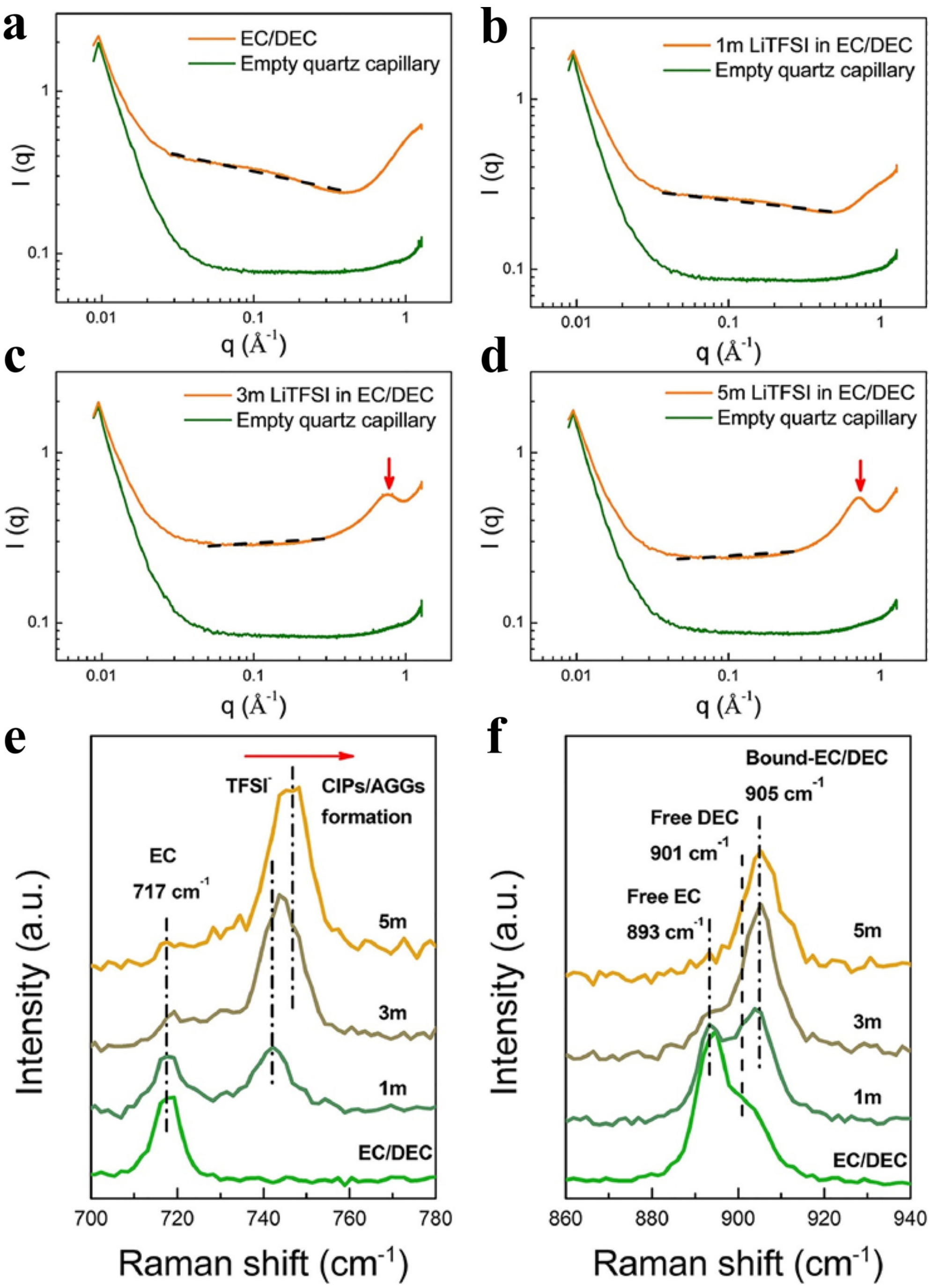

FIG. 4. SAXS data of (a) pure EC/DEC and LiTFSI in EC/DEC at salt concentration of (b) 1 m, (c) 3 m, and (d) 5 m. Raman spectra of pure EC/DEC and 1, 3, 5 m LiTFSI in EC/DEC: (e) 700–780 cm^{-1}; (f) 860–940 cm^{-1}.[129] Reproduced with permission from Qian *et al.*, Energy Storage Mater. **41**, 222–229 (2021). Copyright 2021 Elsevier.

02 July 2024 12:36:38

Published under an exclusive license by AIP Publishing

observed that the presence of ethyl methyl carbonate (EMC) in $LiPF_6$/EC systems resulted in the formation of $\sim$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 [Figs. 4(a)–4(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 [Figs. 4(e) and 4(f)]. Also, we[118] conducted a comprehensive investigation using SAXS/WAXS and Raman 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.

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 [Fig. 5(d)]. 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 [Fig. 5(a)], while we uncovered a concentration-dependent exponential relationship between the d-spacing of the anion solvated structure [Fig. 5(b)]. 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 [Fig. 5(c)]. 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 [Fig. 5(e)]. 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 superhigh 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.

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, Y Z 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. 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

02 July 2024 12:36:38

Published under an exclusive license by AIP Publishing

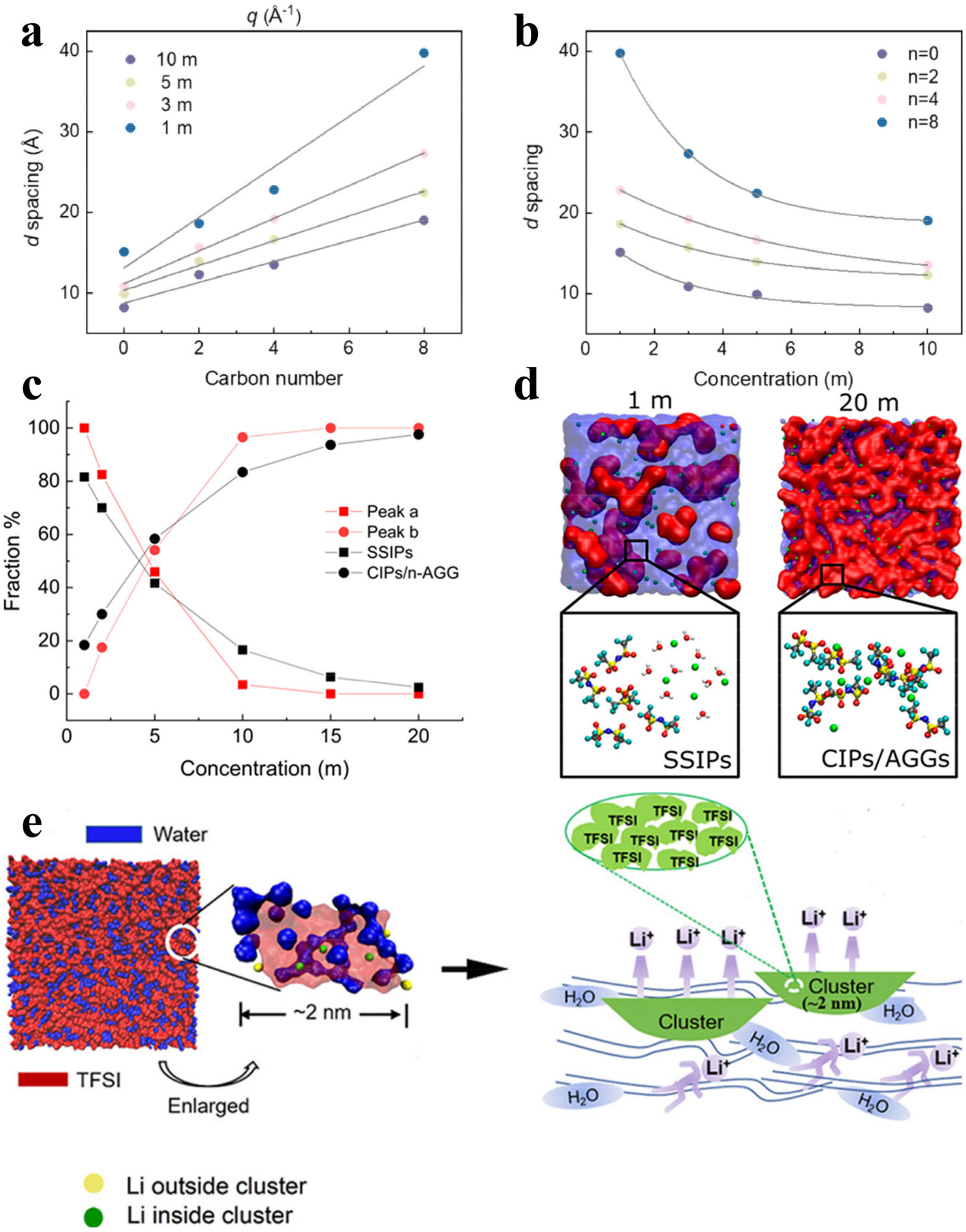

FIG. 5. (a) The linear relationship between carbon number and d spacing of Peak a at different concentration; (b) correlation d-spacing evolution among different concentrations of four imide-based anions.[131] Reproduced with permission from Liu *et al.*, Energy Storage Mater. **45**, 696–703 (2022). 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 (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.

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

02 July 2024 12:36:38

Published under an exclusive license by AIP Publishing

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 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]

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 (Fig. 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 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

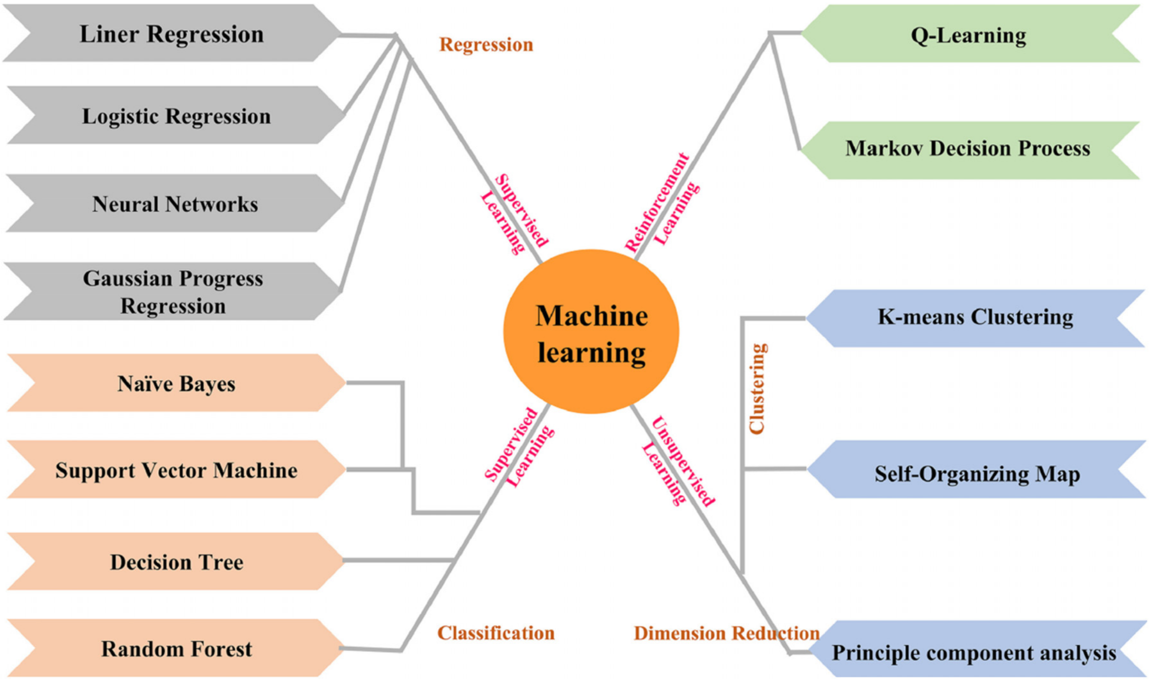

FIG. 6. Common ML algorithms.[147] Reproduced with permission from Liu *et al.*, Energy AI **3**, 100049 (2021). Copyright 2021 Elsevier.

02 July 2024 12:36:38

Published under an exclusive license by AIP Publishing

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]

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 [Fig. 7(a)].[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 in gaining deeper insights into scientific discovery. The Viswanathan group has undertaken several projects related to the "closed-loop" optimization of electrolytes [Fig. 7(b)].[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 (ML-MD), have proven to be effective supplements to experimental characterization for studying solvation structures, transport properties, and interfacial reactions of electrolytes [Fig. 8(a)].[175] Unlike traditional CMD, which rely on empirical force fields, ML-MD 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 ML neural network force field is capable of replicating the 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 AIMD, ML-MD, and free energy calculations to determine the redox potentials and elucidate the solvation structure of electrolytes [Fig. 8(b)].[183,184] This effort is pivotal for clarifying and delineating the structure–properties relationship. The Grossman

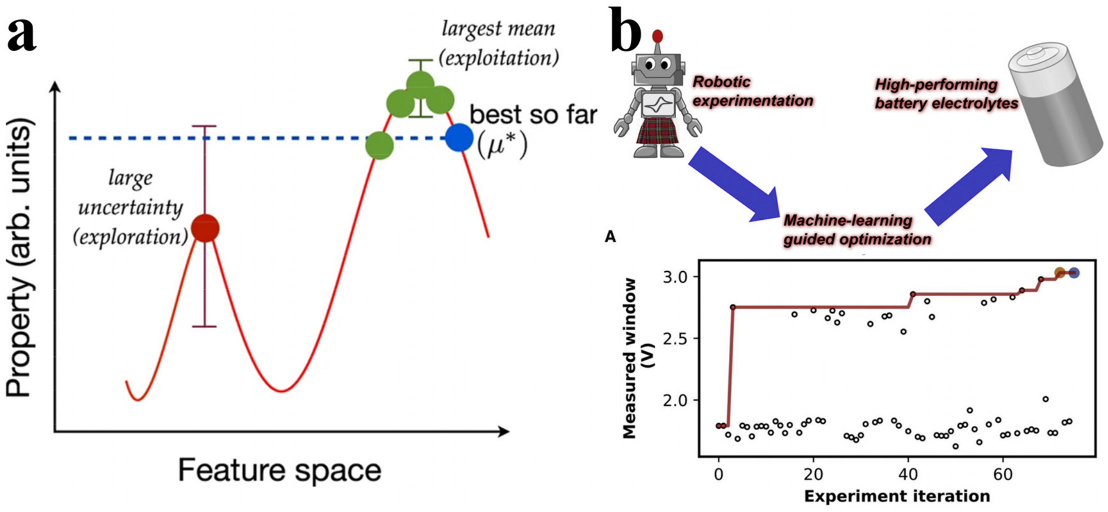

FIG. 7. (a) The trade-off between exploration and exploitaion.[166] Reproduced with permission from Balachandran *et al.*, Sci. Rep. **6**, 19660 (2016). Copyright 2016 Authors, licensed under a Creative Commons Attribution (CC BY) license. (b) Results of autonomous optimization of electrolytes.[167]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

02 July 2024 12:36:38

Published under an exclusive license by AIP Publishing

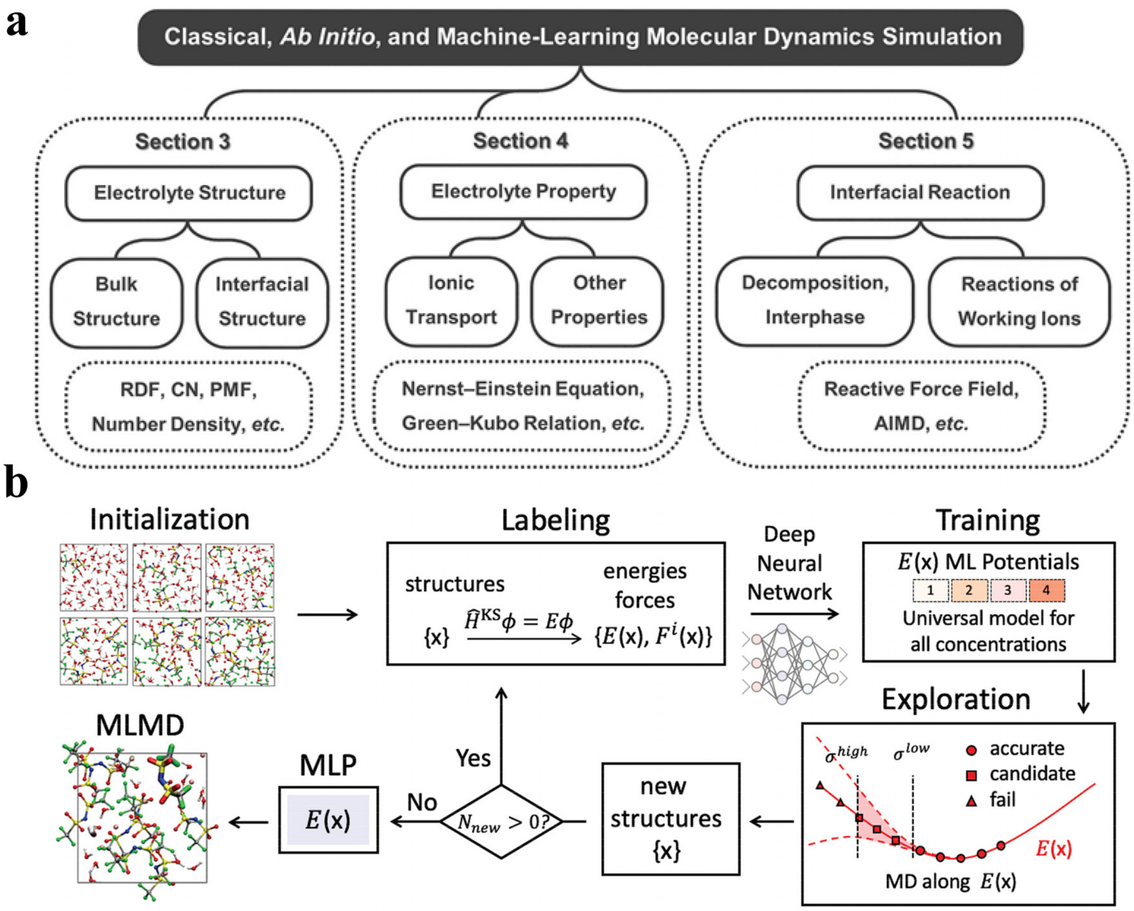

FIG. 8. (a) Framework of applying CMD, AIMD, and MLMD simulations in electrolytes.[175] Reproduced with permission from Yao *et al.*, Chem. Rev. **122**, 10970–11021 (2022). Copyright 2022 American Chemical Society. (b) Workflow of ML potential training process.[184] Reproduced with permission from Wang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

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.

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.

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 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 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

02 July 2024 12:36:38

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 toward 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 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 (Fig. 9).

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 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. First, 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. Second, 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

02 July 2024 12:36:38

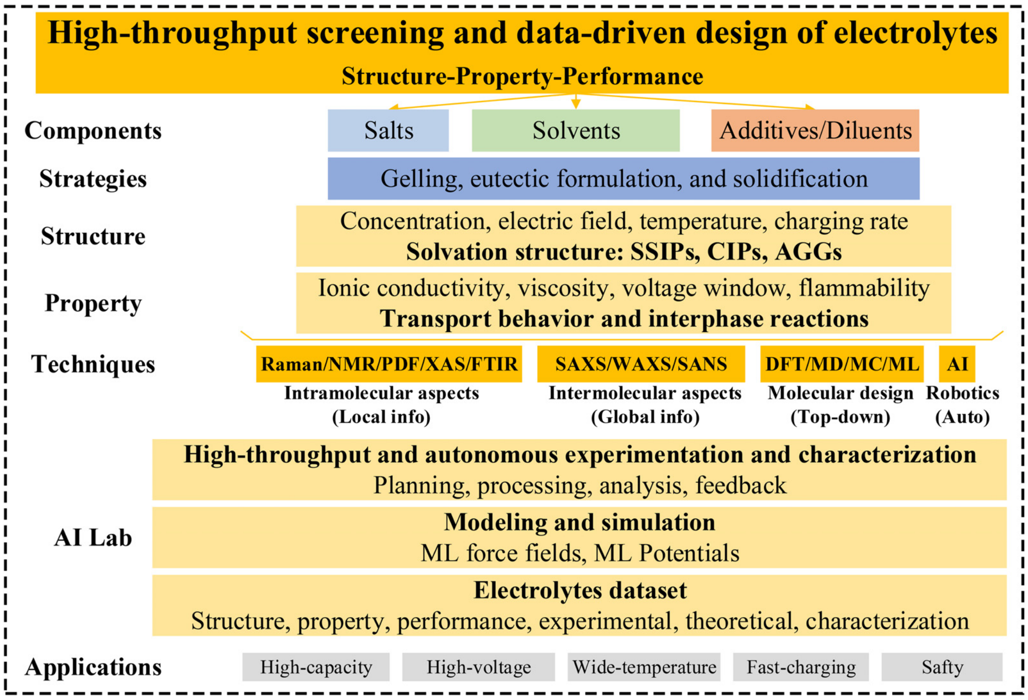

FIG. 9. Characterizing and designing perspectives of advanced electrolytes.

Published under an exclusive license by AIP Publishing

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 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]

ACKNOWLEDGMENTS

This work was supported as part of the Joint Center for Energy Storage Research, an Energy Innovation Hub funded by the U.S. Department of Energy, Office of Science, Basic Energy Sciences. T. Li is thankful for the support by U.S. National Science Foundation (Grant Nos. 2208972, 2120559, and 2323117).

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Xiaozhao Liu: Writing – original draft (equal). **Volodymyr Koverga:** Writing – review & editing (equal). **Hoai T. Nguyen:** Writing – review & editing (supporting). **Anh T. Ngo:** Writing – review & editing (equal). **Tao Li:** Conceptualization (lead); Funding acquisition (lead); Supervision (lead); Writing – review & editing (equal).

DATA AVAILABILITY

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

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] J. B. Goodenough and Y. Kim, Chem. Mater. **22**(3), 587 (2010).
[5] J. Xie and Y. C. Lu, Nat. Commun. **11**(1), 2499 (2020).
[6] S. Ferrari, M. Falco, A. B. Muñoz-García, M. Bonomo, S. Brutti, M. Pavone, and C. Gerbaldi, Adv. Energy Mater. **11**(43), 2100785 (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] Y. Chen, M. Xu, Y. Huang, and A. Manthiram, Chem **8**(2), 312 (2022).
[10] S. K. Sharma, G. Sharma, A. Gaur, A. Arya, F. S. Mirsafi, R. Abolhassani, H.-G. Rubahn, J.-S. Yu, and Y. K. Mishra, Energy Adv. **1**(8), 457 (2022).
[11] J. Janek and W. G. Zeier, Nat. Energy **8**(3), 230 (2023).
[12] A. Yoshino, Angew. Chem., Int. Ed. Engl. **51**(24), 5798 (2012).
[13] C. R. Birkl, M. R. Roberts, E. McTurk, P. G. Bruce, and D. A. Howey, J. Power Sources **341**, 373 (2017).
[14] T. R. Tanim, P. J. Weddle, Z. Yang, A. M. Colclasure, H. Charalambous, D. P. Finegan, Y. Lu, M. Preefer, S. Kim, J. M. Allen, F. L. E. Usseglio-Viretta, P. R. Chinnam, I. Bloom, E. J. Dufek, K. Smith, G. Chen, K. M. Wiaderek, J. N. Weker, and Y. Ren, Adv. Energy Mater. **12**(46), 2202795 (2022).
[15] P. Du, D. Liu, X. Chen, H. Xie, X. Qu, D. Wang, and H. Yin, Energy Storage Mater. **57**, 371 (2023).
[16] T. Waldmann, B.-I. Hogg, and M. 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] C. V. Amanchukwu, Joule **4**(2), 281 (2020).
[20] Z. Yu, N. P. Balsara, O. Borodin, A. A. Gewirth, N. T. Hahn, E. J. Maginn, K. A. Persson, V. Srinivasan, M. F. Toney, K. Xu, K. R. Zavadil, L. A. Curtiss, and L. Cheng, ACS Energy Lett. **7**(1), 461 (2022).
[21] C. A. Bridges, X.-G. Sun, J. Zhao, M. Parans Paranthaman, and S. Dai, J. Phys. Chem. C **116**(14), 7701 (2012).
[22] X. Liu, S.-C. Lee, S. Seifert, L. He, C. Do, R. E. Winans, G. Kwon, Y. Z, and T. Li, Chem. Mater. **35**(5), 2088 (2023).
[23] K. Qian, R. E. Winans, and T. Li, Adv. Energy Mater. **11**(4), 2002821 (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), e2110077118 (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. Acad. Sci. U. S. A. **120**(10), e2214357120 (2023).
[32] T. Fu, F. Monaco, J. Li, K. Zhang, Q. Yuan, P. Cloetens, P. Pianetta, and Y. Liu, Adv. Funct. Mater. **32**(39), 2203070 (2022).
[33] K. A. Severson, P. M. Attia, N. Jin, N. Perkins, B. Jiang, Z. Yang, M. H. Chen, M. Aykol, P. K. Herring, D. Fraggedakis, M. Z. Bazant, S. J. Harris, W. C. Chueh, and R. D. Braatz, Nat. Energy **4**(5), 383 (2019).
[34] D. Roman, S. Saxena, V. Robu, M. Pecht, and D. 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] S. Tan, W. Wang, Y. Tian, S. Xin, and Y. Guo, Adv. Funct. Mater. **31**(45), 2105253 (2021).
[41] S. Liu, R. Zhang, J. Mao, J. Yuwono, C. Wang, K. Davey, and Z. Guo, Appl. Phys. Rev. **10**(2), 021304 (2023).

02 July 2024 12:36:38

Published under an exclusive license by AIP Publishing

[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] X. Li, J. Liang, X. Yang, K. R. Adair, C. Wang, F. Zhao, and X. Sun, Energy Environ. Sci. **13**(5), 1429 (2020).

[45] H. Kwak, S. Wang, J. Park, Y. Liu, K. T. Kim, Y. Choi, Y. Mo, and Y. S. Jung, ACS Energy Lett. **7**(5), 1776 (2022).

[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, Nano-Micro 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] T. T. Vu, H. J. Cheon, S. Y. Shin, G. Jeong, E. Wi, and M. Chang, Energy Storage Mater. **61**, 102876 (2023).

[51] H. Yang, J. Li, Z. Sun, R. Fang, D.-W. Wang, K. He, H.-M. Cheng, and F. 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] M. Park, J. Ryu, W. Wang, and J. Cho, Nat. Rev. Mater. **2**(1), 16080 (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] H. Jia and W. Xu, Trends Chem. **4**(7), 627 (2022).

[57] Y. Wu, D. Ren, X. Liu, G. Xu, X. Feng, Y. Zheng, Y. Li, M. Yang, Y. Peng, X. Han, L. Wang, Z. Chen, Y. Ren, L. Lu, X. He, J. Chen, K. Amine, and M. Ouyang, Adv. Energy Mater. **11**(47), 2102299 (2021).

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

[59] J. Xia, R. Petibon, D. Xiong, L. Ma, and J. R. Dahn, J. Power Sources **328**, 124 (2016).

[60] D. Chai, H. Yan, X. Wang, X. Li, and Y. Fu, Adv. Funct. Mater. **34**(8), 2310516 (2023).

[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] Y. Zhao, T. Zhou, D. Baster, M. El Kazzi, J. W. Choi, and A. Coskun, ACS Energy Lett. **8**(7), 3180 (2023).

[63] X. Peng, T. Wang, B. Liu, Y. Li, and T. 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] X. Zheng, L. Huang, X. Ye, J. Zhang, F. Min, W. Luo, and Y. Huang, Chem **7**(9), 2312 (2021).

[69] Q. Sun, Z. Cao, Z. Ma, J. Zhang, H. Cheng, X. Guo, G.-T. Park, Q. Li, E. Xie, L. Cavallo, Y.-K. Sun, and J. Ming, ACS Energy Lett. **7**(10), 3545 (2022).

[70] P. Jaumaux, J. Wu, D. Shanmukaraj, Y. Wang, D. Zhou, B. Sun, F. Kang, B. Li, M. Armand, and G. Wang, Adv. Funct. Mater. **31**(10), 2008644 (2020).

[71] R. Weber, M. Genovese, A. J. Louli, S. Hames, C. Martin, I. G. Hill, and J. R. Dahn, Nat. Energy **4**(8), 683 (2019).

[72] Q. Sun, Z. Cao, Z. Ma, J. Zhang, W. Wahyudi, G. Liu, H. Cheng, T. Cai, E. Xie, L. Cavallo, Q. Li, and J. Ming, Adv. Funct. Mater. **33**(1), 2210292 (2022).

[73] Y. Zou, G. Liu, Y. Wang, Q. Li, Z. Ma, D. Yin, Y. Liang, Z. Cao, L. Cavallo, H. Kim, L. Wang, H. N. Alshareef, Y.-K. Sun, and J. Ming, Adv. Energy Mater. **13**(19), 2300443 (2023).

[74] G. Liu, Z. Cao, L. Zhou, J. Zhang, Q. Sun, J. Hwang, L. Cavallo, L. Wang, Y. Sun, and J. Ming, Adv. Funct. Mater. **30**(43), 2001934 (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] C. Yang, J. Xia, C. Cui, T. P. Pollard, J. Vatamanu, A. Faraone, J. A. Dura, M. Tyagi, A. Kattan, E. Thimsen, J. Xu, W. Song, E. Hu, X. Ji, S. Hou, X. Zhang, M. S. Ding, S. Hwang, D. Su, Y. Ren, X.-Q. Yang, H. Wang, O. Borodin, and C. Wang, Nat. Sustainability **6**(3), 325 (2023).

[79] S. C. Kim, J. Wang, R. Xu, P. Zhang, Y. Chen, Z. Huang, Y. Yang, Z. Yu, S. T. Oyakhire, W. Zhang, L. C. Greenburg, M. S. Kim, D. T. Boyle, P. Sayavong, Y. Ye, J. Qin, Z. Bao, and Y. Cui, Nat. Energy **8**(8), 814 (2023).

[80] L. Deng, L. Dong, Z. Wang, Y. Liu, J. Zhan, S. Wang, K. Song, D. Qi, Y. Sang, H. Liu, and H. Chen, Adv. Energy Mater. **14**(4), 2303652 (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. **36**(7), e2306462 (2023).

[82] Y. Xia, P. Zhou, X. Kong, J. Tian, W. Zhang, S. Yan, W-h Hou, H.-Y. Zhou, H. Dong, X. Chen, P. Wang, Z. Xu, L. Wan, B. Wang, and K. Liu, Nat. Energy **8**(9), 934 (2023).

[83] R. Lin, C. Ke, J. Chen, S. Liu, and J. 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 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, R. Weber, M. Genovese, M. Coon, J. deGooyer, Z. Deng, R. T. White, J. Lee, T. Rodgers, R. Petibon, S. Hy, S. J. H. Cheng, and J. R. Dahn, Nat. Energy **5**(9), 693 (2020).

[87] F. Wu, S. Fang, M. Kuenzel, A. Mullaliu, J.-K. Kim, X. Gao, T. Diemant, G.-T. Kim, and S. Passerini, Joule **5**(8), 2177 (2021).

[88] D. M. Driscoll, S. N. Lavan, M. Zorko, P. C. Redfern, S. Ilic, G. Agarwal, T. T. Fister, R. S. Assary, L. Cheng, D. Strmcnik, M. Balasubramanian, and J. G. Connell, Chem **9**(7), 1955 (2023).

[89] T. Cai, Q. Sun, Z. Cao, Z. Ma, W. Wahyudi, L. Cavallo, Q. Li, and J. Ming, J. Phys. Chem. 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. **9**(26), e2201893 (2022).

[91] W. Wahyudi, V. Ladelta, L. Tsetseris, M. M. Alsabban, X. Guo, E. Yengel, H. Faber, B. Adilbekova, A. Seitkhan, A. H. Emwas, M. N. Hedhili, L. J. Li, V. Tung, N. Hadjichristidis, T. D. Anthopoulos, and J. Ming, Adv. Funct. Mater. **31**(23), 2101593 (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] J. Li, H. Li, X. Ma, W. Stone, S. Glazier, E. Logan, E. M. Tonita, K. L. Gering, and J. R. Dahn, J. Electrochem. Soc. **165**(5), A1027 (2018).

[95] D. S. Hall, A. Eldesoky, E. R. Logan, E. M. Tonita, X. 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**(9), e2020357118 (2021).

[97] C. Wang, Y. S. Meng, and K. Xu, J. Electrochem. Soc. **166**(3), A5184 (2018).

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

[99] Z. Yu, H. Wang, X. Kong, W. Huang, Y. Tsao, D. G. Mackanic, K. Wang, X. Wang, W. Huang, S. Choudhury, Y. Zheng, C. V. Amanchukwu, S. T. Hung, Y. Ma, E. G. Lomeli, J. Qin, Y. Cui, and Z. Bao, Nat. Energy **5**(7), 526 (2020).

[100] Z. Yu, P. E. Rudnicki, Z. Zhang, Z. Huang, H. Celik, S. T. Oyakhire, Y. Chen, X. Kong, S. C. Kim, X. Xiao, H. Wang, Y. Zheng, G. A. Kamat, M. S. Kim, S. F. Bent, J. Qin, Y. Cui, and Z. Bao, Nat. Energy **7**(1), 94 (2022).

[101] R. Mogensen, A. Buckel, S. Colbin, and R. Younesi, Chem. Mater. **33**(4), 1130 (2021).

[102] B. A. Paren, B. A. Thurston, A. Kanthawar, W. J. Neary, A. Kendrick, M. Maréchal, J. G. Kennemur, M. J. Stevens, A. L. Frischknecht, and K. 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).

02 July 2024 12:36:38

Published under an exclusive license by AIP Publishing

[104] Y. Yamada, J. Wang, S. Ko, E. Watanabe, and A. 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] X. Ren, S. Chen, H. Lee, D. Mei, M. H. Engelhard, S. D. Burton, W. Zhao, J. Zheng, Q. Li, M. S. Ding, M. Schroeder, J. Alvarado, K. Xu, Y. S. Meng, J. Liu, J.-G. Zhang, and W. Xu, Chem **4**(8), 1877 (2018).

[107] H. Dong, O. Tutusaus, Y. Liang, Y. Zhang, Z. Lebens-Higgins, W. Yang, R. Mohtadi, and Y. 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] P. Yang, X. Gao, X. Tian, C. Shu, Y. Yi, P. Liu, T. Wang, L. Qu, B. Tian, M. Li, W. Tang, B. Yang, and J. B. Goodenough, ACS Energy Lett. **5**(5), 1681 (2020).

[111] C. Zhang, Z. Lu, M. Song, Y. Zhang, C. Jing, L. Chen, X. Ji, and W. 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] W. Yang, X. Du, J. Zhao, Z. Chen, J. Li, J. Xie, Y. Zhang, Z. Cui, Q. Kong, Z. Zhao, C. Wang, Q. Zhang, and G. 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] A. L. Phan, C. Jayawardana, P. M. L. Le, J. Zhang, B. Nan, W. Zhang, B. L. Lucht, S. Hou, and C. Wang, Adv. Funct. Mater. **33**(34), 2301177 (2023).

[116] G. Cong and Y.-C. Lu, Chem **4**(12), 2732 (2018).

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

[118] K. Qian, S. Seifert, R. E. Winans, and T. Li, Energy Fuels **35**(23), 19849 (2021).

[119] X. Liu, L. Fang, X. Lyu, R. E. Winans, and T. Li, Chem. Mater. **35**(23), 9821 (2023).

[120] J. Hu, Y. Ji, G. Zheng, W. Huang, Y. Lin, L. Yang, and F. Pan, Aggregate **3**(1), e153 (2022).

[121] Q. Sun, Z. Cao, Z. Ma, J. Zhang, W. Wahyudi, T. Cai, H. Cheng, Q. Li, H. Kim, E. Xie, L. Cavallo, Y.-K. Sun, and J. Ming, ACS Mater. Lett. **4**(11), 2233 (2022).

[122] Y. Wang, Z. Cao, Z. Ma, G. Liu, H. Cheng, Y. Zou, L. Cavallo, Q. Li, and J. 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. **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] Z. Feng, E. Sarnello, T. Li, and L. Cheng, J. Electrochem. Soc. **166**(2), A47 (2019).

[129] K. Qian, Y. Liu, X. Zhou, D. J. Gosztola, H. Nguyen, and T. Li, Energy Storage Mater. **41**, 222 (2021).

[130] K. Qian, Z. Yu, Y. Liu, D. J. Gosztola, R. E. Winans, L. Cheng, and T. Li, J. Energy Chem. **70**, 340 (2022).

[131] X. Liu, S.-C. Lee, S. Seifer, R. E. Winans, L. Cheng, Y. Z, and T. Li, Energy Storage Mater. **45**, 696 (2022).

[132] X. Liu, Z. Yu, E. Sarnello, K. Qian, S. Seifert, R. E. Winans, L. Cheng, and T. Li, Energy Mater. Adv. **2021**, 7368420.

[133] P. Tan, J. Yue, Y. Yu, B. Liu, T. Liu, L. Zheng, L. He, X. Zhang, L. Suo, and L. Hong, J. Phys. Chem. C **125**(22), 11838 (2021).

[134] X. Liu, S.-C. Lee, S. Seifert, R. E. Winans, Y. Z, and T. Li, Chem. Mater. **35**(16), 6415 (2023).

[135] L. Su, X. Zhao, M. Yi, H. Charalambous, H. Celio, Y. Liu, and A. 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), e2105889118 (2021).

[137] J. Yang, H. Yan, H. Hao, Y. Song, Y. Li, Q. Liu, and A. Tang, ACS Energy Lett. **7**(7), 2331 (2022).

[138] K. Peng, Y. Li, G. Tang, Y. Liu, Z. Yang, and T. Xu, Energy Environ. Sci. **16**(2), 430 (2023).

[139] Y. Zhao, Z. Yu, L. A. Robertson, J. Zhang, Z. Shi, S. R. Bheemireddy, I. A. Shkrob, Z. Y, T. Li, Z. Zhang, L. Cheng, and L. 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] A. Agrawal and A. Choudhary, APL Mater. **4**(5), 053208 (2016).

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

[145] A. Jain, Y. Shin, and K. 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] Y. Liu, O. Christopher Esan, Z. Pan, and L. An, Energy AI **3**, 100049 (2021).

[148] T. Xie, "Deep learning methods for the design and understanding of solid materials," Ph.D. thesis, Massachusetts Institute of Technology, 2020, https://dspace.mit.edu/handle/1721.1/129054.

[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] A. D. Sendek, B. Ransom, E. D. Cubuk, L. A. Pellouchoud, J. Nanda, and E. J. Reed, Adv. Energy Mater. **12**(31), 2200553 (2022).

[151] Z. Ren, Z. Ren, Z. Zhang, T. Buonassisi, and J. Li, Nat. Rev. Mater. **8**(9), 563 (2023).

[152] Y. Xie, K. Sattari, C. Zhang, and J. Lin, Prog. Mater. Sci. **132**, 101043 (2023).

[153] J. M. Gregoire, L. Zhou, and J. 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] N. S. Eyke, B. A. Koscher, and K. F. Jensen, Trends Chem. **3**(2), 120 (2021).

[156] M. Abolhasani and K. 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] Z. Wang, K. Cruse, Y. Fei, A. Chia, Y. Zeng, H. Huo, T. He, B. Deng, O. Kononova, and G. 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] H. Huo, Z. Rong, O. Kononova, W. Sun, T. Botari, T. He, V. Tshitoyan, and G. Ceder, npj Comput. Mater. **5**(1), 62 (2019).

[164] N. J. Szymanski, C. J. Bartel, Y. Zeng, Q. Tu, and G. 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] A. Dave, J. Mitchell, K. Kandasamy, H. Wang, S. Burke, B. Paria, B. Póczos, J. Whitacre, and V. Viswanathan, Cell Rep. Phys. Sci. **1**(12), 100264 (2020).

[168] A. Benayad, D. Diddens, A. Heuer, A. Narayanan Krishnamoorthy, M. Maiti, F. Le Cras, M. Legallais, F. Rahmanian, Y. Shin, H. Stein, M. Winter, C. Wölke, P. Yan, and I. Cekic-Laskovic, Adv. Energy Mater. **12**(17), 2102678 (2021).

[169] J. Rodriguez, M. Politi, S. Adler, D. Beck, and L. Pozzo, Mol. Syst. Des. Eng. **7**(8), 933 (2022).

02 July 2024 12:36:38

Published under an exclusive license by AIP Publishing

[170] K. Hippalgaonkar, Q. Li, X. Wang, J. W. Fisher, J. Kirkpatrick, and T. 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] J. F. Whitacre, J. Mitchell, A. Dave, W. Wu, S. Burke, and V. Viswanathan, J. Electrochem. Soc. **166**(16), A4181 (2019).

[173] P. H. Svensson, P. Yushmanov, A. Tot, L. Kloo, E. Berg, and K. Edström, Chem. Eng. J. **455**, 140955 (2023).

[174] J. T. Yik, L. Zhang, J. Sjölund, X. Hou, P. H. Svensson, K. Edström, and E. 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. 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] I.-B. Magdău, D. J. Arismendi-Arrieta, H. E. Smith, C. P. Grey, K. Hermansson, and G. 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] Y. Wang, T. Xie, A. France-Lanord, A. Berkley, J. A. Johnson, Y. Shao-Horn, and J. 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] J.-P. Correa-Baena, K. Hippalgaonkar, J. van Duren, S. Jaffer, V. R. Chandrasekhar, V. Stevanovic, C. Wadia, S. Guha, and T. 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] C. Ling, npj Comput. Mater. **8**(1), 33 (2022).

[191] E. W. C. Spotte-Smith, O. A. Cohen, S. M. Blau, J. M. Munro, R. Yang, R. D. Guha, H. D. Patel, S. Vijay, P. Huck, R. Kingsbury, M. K. Horton, and K. A. Persson, Digital Discovery **2**(6), 1862 (2023).

[192] G. dos Reis, C. Strange, M. Yadav, and S. Li, Energy AI **5**, 100081 (2021).

[193] L. Ward, S. Babinec, E. J. Dufek, D. A. Howey, V. Viswanathan, M. Aykol, D. A. C. Beck, B. Blaiszik, B.-R. Chen, G. Crabtree, S. Clark, V. De Angelis, P. Dechent, M. Dubarry, E. E. Eggleton, D. P. Finegan, I. Foster, C. B. Gopal, P. K. Herring, V. W. Hu, N. H. Paulson, Y. Preger, D. Uwe-Sauer, K. Smith, S. W. Snyder, S. Sripad, T. R. Tanim, and L. 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] Y. Wu, Q. Hu, H. Liang, A. Wang, H. Xu, L. Wang, and X. He, Adv. Energy Mater. **13**(22), 2300259 (2023).

[196] F. A. L. Laskowski, D. B. McHaffie, and K. 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] A. Mistry, A. Verma, S. Sripad, R. Ciez, V. Sulzer, F. Brosa Planella, R. Timms, Y. Zhang, R. Kurchin, P. Dechent, W. Li, S. Greenbank, Z. Ahmad, D. Krishnamurthy, A. M. Fenton, K. Tenny, P. Patel, D. Juarez Robles, P. Gasper, A. Colclasure, A. Baskin, C. D. Scown, V. R. Subramanian, E. Khoo, S. Allu, D. Howey, S. DeCaluwe, S. A. Roberts, and V. Viswanathan, ACS Energy Lett. **6**(11), 3831 (2021).

[203] A. Y.-T. Wang, R. J. Murdock, S. K. Kauwe, A. O. Oliynyk, A. Gurlo, J. Brgoch, K. A. Persson, and T. D. Sparks, Chem. Mater. **32**(12), 4954 (2020).

02 July 2024 12:36:38

Published under an exclusive license by AIP Publishing