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Machine learning-inspired battery material innovation

Man-Fai Ng, ^a Yongming Sun ^b and Zhi Wei Seh ^{*c}

Machine learning (ML) techniques have been a powerful tool responsible for many new discoveries in materials science in recent years. In the field of energy storage materials, particularly battery materials, ML techniques have been widely utilized to predict and discover materials' properties. In this review, we first discuss the key properties of the most common electrode and electrolyte materials. We then summarize recent progress in battery material advancement using ML techniques, through the three main strategies of direct property predictions, machine learning potentials, and inverse design. The major challenges, advantages and limitations of these techniques are also discussed. Finally, we conclude this review with a perspective on sustainable battery development using ML.

1. Introduction

Reducing global greenhouse gas emissions to limit global warming to under two degrees Celsius remains a major challenge.^{1–5} National initiatives to reduce CO₂ emissions by 2030, with a view towards achieving net zero emissions by 2050 make research and development in this area critical. The adoption of electric vehicles (EVs) is gaining major traction as

directly replacing fossil fuel vehicles removes a major source of greenhouse gases. Successful EV deployment, however, depends mainly on overcoming the 'range anxiety' issue. Developing improved battery models to monitor the state of charge and health of batteries could be one solution;^{6,7} another solution is to develop more long-lasting and sustainable batteries,⁸ in which material design plays an important role.

Batteries are complex, dynamic electrochemical systems in which the two major components (Fig. 1a) are electrodes (negative and positive) and electrolytes (liquid and solid states). In designing battery electrodes, the key parameters to consider include voltage and specific charge capacity, which contribute to the overall energy density; and volume expansions during charging and discharging, which determine the cyclability and safety issues. For electrolytes, the redox potentials and stability windows (liquid electrolytes) and ionic conductivities and

^a Institute of High Performance Computing (IHPC), Agency for Science, Technology and Research (A*STAR), 1 Fusionopolis Way, #16-16 Connexis, Singapore, 138632, Republic of Singapore. E-mail: ngmf@ihpc.a-star.edu.sg

^b Wuhan National Laboratory for Optoelectronics, Huazhong University of Science and Technology, Wuhan, 430074, China

^c Institute of Materials Research and Engineering (IMRE), Agency for Science, Technology and Research (A*STAR), 2 Fusionopolis Way, Innova #08-03, Singapore, 138634, Republic of Singapore. E-mail: sehzw@imre.a-star.edu.sg



Man-Fai Ng

Man-Fai Ng is a Senior Scientist at the A*STAR Institute of High Performance Computing in Singapore, where he has been since 2006. He received his BSc in Chemistry and PhD in Theoretical Chemistry from the University of Hong Kong in 1998 and 2003, respectively. His current research interests include machine learning and computational design of emerging materials for advanced batteries, electrocatalysis and corrosion control.



Yongming Sun

Yongming Sun received his PhD degree from Huazhong University of Science and Technology in 2012. He is currently a Professor at Wuhan National Laboratory for Optoelectronics at Huazhong University of Science and Technology. His research focuses on materials for rechargeable Li-based batteries.



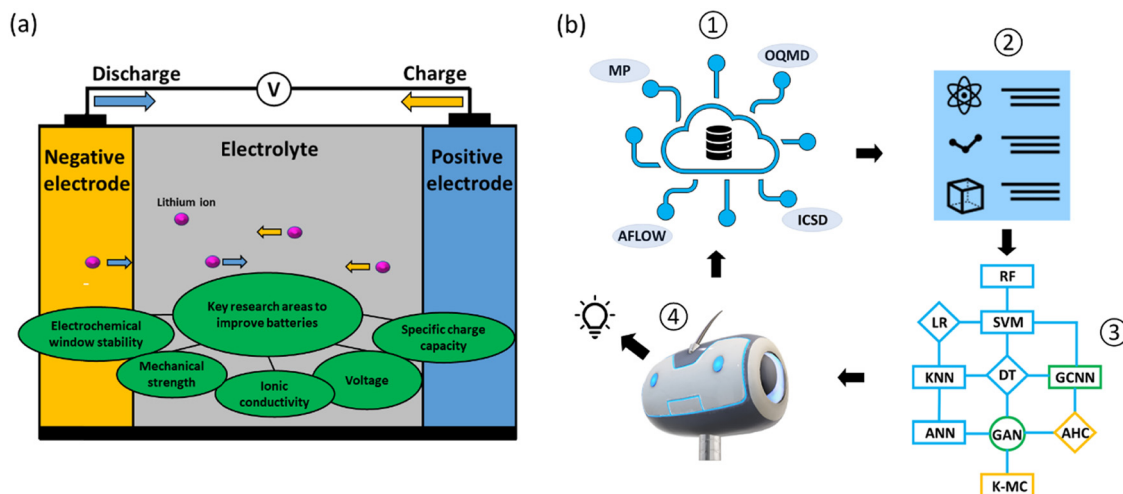


Fig. 1 (a) A schematic of a battery cell showing the major material components (electrodes and electrolyte) and the key material properties actively under research for improving the performance of batteries. (b) A general ML workflow starting with (1): preparing data from databases; (2) constructing feature vectors: feature selections for supervised learning or dimensionality reduction for unsupervised learning; (3) constructing ML algorithms: supervised, semi-supervised and unsupervised learning; (4) a predictive model to predict outputs. The predicted results might be re-fed as inputs for further ML model improvements.

mechanical strengths (solid-state electrolytes) are the key parameters. These parameters are material-dependent, and identifying better battery materials *via* material design and discovery relies on the exploration of the growing chemical spaces,⁹ making it increasingly difficult to use conventional trial-and-error discovery approaches.

Data-driven machine learning (ML) and deep learning (DL) techniques, together with high-throughput experimental and computational approaches, have recently emerged as important tools for the design and discovery of battery materials^{10–19} (Fig. 1b). The battery design problem in a data-driven approach with ML techniques essentially solves a complex function that takes into account battery formulations as the inputs and

performance measurements as the outputs. The ML techniques are thus used to optimize these correlation functions. The typical ML algorithms adopted for battery material design can be classified into three categories: supervised, semi-supervised and unsupervised learning. Supervised learning learns from labeled data. Common algorithms used are decision tree (DT), linear regression (LR), random forest (RF), support vector machine (SVM), artificial neural network (ANN) and k-nearest neighbors (KNN). Semi-supervised learning learns from both labeled and unlabeled data. Common algorithms used here are graph convolution neural network (GCNN) and generative adversarial networks (GANs). Unsupervised learning is learning without target variables and operates only with input data. Common algorithms used here are k-mean clustering and agglomerative hierarchical clustering (AHC; Fig. 1b).

Also, high quality datasets lie at the center of ML techniques. Databases are generally classified into two main types: experimental and computational. For battery material discovery, the crystal structures of inorganic and organic compounds are the most widely used datasets for screening and feature extractions. The Materials Project (MP),^{20,21} Open Quantum Materials Database (OQMD)²² and Automatic FLOW for Materials Discovery (AFLOW)²³ are open access databases for computational data of inorganic compounds, while the NIST Inorganic Crystal Structure Database (ICSD)²⁴ is the database chiefly used for experimental data. For organic compounds, the Cambridge Structural Database (CSD),²⁵ Organic Materials Database (OMDB)²⁶ and ChemSpider²⁷ are examples of commonly-used databases (Fig. 1b). These databases typically contain hundreds of thousands to millions of data entries.

In this review, we first discuss the key properties of the most common electrode and electrolyte materials. Then, we summarize recent progress on battery materials advancement using ML techniques, through three ML strategies: (1) direct property



Zhi Wei Seh

*Zhi Wei Seh is a Senior Scientist at the Institute of Materials Research and Engineering, A*STAR. He received his BS and PhD degrees in Materials Science and Engineering from Cornell University and Stanford University, respectively. His research interests lie in the design of new materials for energy storage and conversion, including advanced batteries and electrocatalysts. As a highly cited researcher in Web of Science, he is widely recognized*

for designing the first yolk-shell nanostructure in lithium-sulfur batteries, which is a licensed technology. He also pioneered the first experimental study of MXenes as electrocatalysts for hydrogen evolution and carbon dioxide reduction.



predictions; (2) machine learning potentials; and (3) inverse design. New battery materials predicted by ML are highlighted in the process. The major challenges, advantages and limitations of these techniques are also discussed. Finally, the review concludes with perspectives on the development of sustainable batteries using ML techniques.

2. Current battery materials

In this section, we briefly review the key properties of common battery electrode and electrolyte materials. Since the commercialization of lithium-ion batteries (LIBs) by SONY Corporation in 1991, their development has progressed notably, especially in terms of materials for the positive electrode. LiCoO_2 was initially used for this purpose,²⁸ while LiMn_2O_4 , LiFePO_4 and lithium nickel manganese cobalt oxide (NMC) have since been developed as more advanced electrode materials, showing improved energy density and safety. These materials have voltages ranging from 3.4 to 3.9 V and specific charge capacities ranging from 125 to 190 mA h g^{-1} .^{29,30} Despite these advancements, the quest for improving LIB technology remains.

At the same time, promising battery technologies such as sodium-ion batteries (NIBs), organic batteries, and lithium–sulfur batteries are already on the horizon. Although the typical cathode specific charge capacity for NIBs is about 150 mA h g^{-1} ,³¹ a value lower than that of LIBs, NIBs display great potential as a low-cost battery due to the abundance of sodium. The reported specific charge capacities for organic batteries are generally higher, typically at a range from 200 to 500 mA h g^{-1} , but the voltages are generally lower, at a range from 2.2 to 3.5 V when compared to advanced LIBs. Moreover, the cycling performance varies from

tens to thousands of cycles.³² Although lithium–sulfur batteries are a promising high-energy battery technology (with the first discharge specific capacity reaching 1645 mA h g^{-1} , which is close to the theoretical limit for sulfur³³), mitigating the effects of polysulfide shuttling (which can shorten battery life) remains a challenge.

The comparison of the voltage-specific charge capacity performance of common electrodes for various battery technologies is shown in Fig. 2a. The development of these battery technologies over the last 30 years and the concurrent advent of physics-based modeling have led to the accumulation of a large-volume of battery data. Many new battery electrode materials with improved performance have been predicted using data-driven approaches with ML (Fig. 2a). These predictions not only identify potential electrodes with high charge capacity or high voltage, but also provide guidelines and directions for further discovery. Voltages and specific charge capacities are the two most important parameters for battery electrodes. However, other less tangible ‘parameters’ such as adsorption strength of polysulfides on host materials and electrolyte coordination energies are also important for improving battery performance. The ML techniques can identify the effects of these parameters and more examples will be discussed in the following section.

Another important battery component is the electrolyte, especially the solid-state electrolyte, which forms a major part of a solid-state battery (SSB). SSBs can provide high volumetric energy density (an important feature for space-based applications such as EV) and are extremely safe, as they contain no flammable liquid solvents. In addition, high ionic conductivity, high mechanical strength, and a wide electrochemical stability window are some important features of solid-state electrolytes. In particular, ionic conductivity is a major parameter that is the

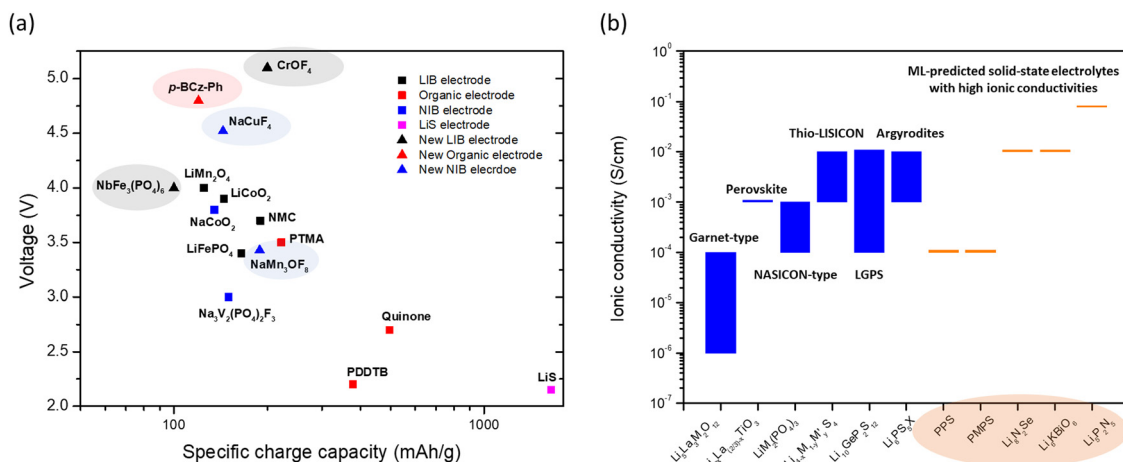


Fig. 2 (a) A plot of voltage (V) versus specific charge capacity (mA h g^{-1}) for common electrodes in lithium-ion batteries (LIB), sodium-ion batteries (NIB), organic batteries and lithium–sulphur batteries. Values for LiCoO_2 , LiMn_2O_4 and LiFePO_4 are taken from ref. 29; NMC811 from ref. 30; quinone, poly(2,2,6,6-tetramethyl-1-piperidinyloxy-4-yl methacrylate) (PTMA) and poly(1,4-di(1,3-dithiolan-2-yl)benzene) (PDDTB) from ref. 31 and the references therein; NaCoO_2 and $\text{Na}_3\text{V}_2(\text{PO}_4)_2\text{F}_3$ from ref. 32; and LiS from ref. 33. The ML-predicted new materials for the LIB electrode,⁴³ organic electrode⁴⁴ and NIB electrode⁴⁵ are highlighted. (b) A plot showing the ionic conductivities (S cm^{-1}) of common solid-state electrolytes. Values of the garnet-type ($\text{Li}_5\text{La}_3\text{M}_2\text{O}_{12}$, $\text{M} = \text{Nb}$ and Ta), perovskite, NASICON-type ($\text{LiM}_2(\text{PO}_4)_3$, $\text{M} = \text{Zr}$, Ti , Hf , Ge and Sn), thio-LISICON ($\text{Li}_{4-3}\text{M}_{1-3}\text{M}'_3\text{S}_4$, $\text{M} = \text{Si}$ and Ge , $\text{M}' = \text{P}$, Al , Zn and Ga), LGPS ($\text{Li}_{10}\text{GeP}_2\text{S}_{12}$) and argyrodites ($\text{Li}_6\text{PS}_5\text{X}$, $\text{X} = \text{Cl}$ and Br) are taken from ref. 34. The ML-predicted solid-state electrolytes^{36,37} with high ionic conductivities are highlighted.



3. Applications of ML in battery material discovery

3. Inverse design: the strategy that starts with the desired functionality of the materials and ends with specific material structures. Optimization and search methods such as genetic algorithm (GA), Bayesian optimization (BO) and particle swarm

In addition to atomistic properties, the ML techniques have also been applied to predict LIB electrode properties at mesoscales such as electrolyte infiltrations and manufacturing parameters. Shodiev *et al.*⁵¹ used an M-LP model, together with data generated by a physics-based model and X-ray images, to predict the electrode filling process in porous NMC electrodes. The developed model is

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A common practice to select ML models for electrode property predictions is to compare a few ML models and to determine the best-performing model using the prediction with the smallest root mean square error (RMSE) or highest coefficient of determination (R^2) score. Feature pairing with the ML models is also critical for accurate predictions. Among the studies outlined above, GBM, M-LP and SVM are found to be better models for electrode property predictions. The GBM algorithms have been known to be robust in describing the nonlinear correlations across a wide variable space. It has been demonstrated that the GBM model has the best prediction power for capacity predictions.⁴⁶ Moreover, the ML-P regression model is found to

Other important parameters are the mechanical properties of solid-state electrolytes and the electrode–electrolyte interface stability. Using a LightGBM model and active learning, Choi *et al.*⁴⁰ predicted the shear and bulk moduli for 2842 solid electrolyte candidates. They found that the physical properties such as volume, density, space group number, and number of atoms are important features for predicting the mechanical properties. Moreover, they found oxide structures to exhibit superior mechanical properties. Ahmad *et al.*⁴¹ used CGCNN, GBR and KRR models to predict the shear and bulk moduli of inorganic solids. They found more than 20 mechanically anisotropic interfaces between lithium metal and 4 solid-state electrolytes, which can suppress dendrite growth. Using SVM and KRR models, Liu *et al.*⁵⁹ predicted the stability of cation-doped $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ in terms of reaction energy. They found the formation energy of oxides (M_xO_y) to be the most important

Table 2 Recent advancements in battery electrolyte development

Electrolyte	Battery	ML method	Descriptor/feature/ input	Dataset	ML prediction	Accuracy	Ref.
21 new solid electrolytes	Solid-state	Log R	5 structural features	317 candidates	Ionic conductivity at room temperature	F1 score = 0.5	38, Sendek <i>et al.</i> (2019)
250 promising solid electrolytes, 26 anode coating materials	Solid-state	RF, GBDT	22 features for migration, 24 features for potentials	Screen 15 000 data	Migration barriers, oxidation and reduction potentials for Li compounds	R^2 values = 0.95, 0.92, and 0.86 for oxi. potential, red. potential, and migration barrier	39, Honrao <i>et al.</i> (2021)
New glass-type ionic conductors	Solid-state	Transfer-learned graph NN	32-dimensional vectors trained from 2000 descriptors	9600 candidates	Ionic conductivity at room temperature	MAE < 1 on a log scale	36, Hatakeyama-Sato <i>et al.</i> (2020)
16 new fast ionic conductors	Solid-state	AHC	528 representative anionic structures and their mXRDs	2986 ICSD entries	Ionic conductivity at room temperature	Number of groups = 7 (score function)	37, Zhang <i>et al.</i> (2019)
2842 solid electrolytes candidates	Solid-state	LightGBM, active learning	145 chemical, 126 structural features	Screening of 17 619 candidates	Shear and bulk moduli	R^2 value = 0.819, 0.863 for shear and bulk moduli	40, Choi <i>et al.</i> (2021)
Over 20 mechanically anisotropic interfaces	Solid-state	CGCNN, GBR, and KRR	17 descriptors	Screening 12 000 inorganic solids	Shear and bulk moduli	RMSE (log(GPa)) = 0.1268, 0.1013 for shear and bulk moduli	41, Ahmad <i>et al.</i> (2018)
18 unexplored $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ predicted	Solid-state	SVM, KRR	15 features	100 LLZOM compounds	Reaction energy	MSE = 0.04	59, Liu <i>et al.</i> (2019)
A novel aqueous electrolyte	Metal-ion	BO	Electrolyte formulations	251 electrolytes	Electrochemical stability window	N.A.	42, Dave <i>et al.</i> (2020)
Liquid electrolytes	Metal-ion	MLR, LASSO, and ES-LiR	13 ion and solvent descriptors	70 solvents, 5 cations	Coordination energies	0.016 eV	60, Ishikawa <i>et al.</i> (2019)
Electrolyte additives	Lithium-ion	GPR	Molecular structural features	149 additives	Redox potentials	MAE = 0.0522, 0.1013 for reduction, oxidation	61, Zhang <i>et al.</i> (2020)

feature, which dominates the route of the interface reactions. As such, the M–O bond strength was found to govern the interface stability of the doped $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ and 18 unexplored doped $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ were predicted and validated by calculations.

For liquid electrolytes, the electrochemical stability window and the interactions between the metal ions and solvent molecules are important parameters for a stable liquid electrolyte. Dave *et al.*⁴² combined robotics and machine learning to discover

new aqueous electrolytes by examining their electrochemical stability windows. Using a BO model, a novel dual-anion sodium electrolyte was discovered. Ishikawa *et al.*⁶⁰ used the multiple linear regression (MLR), exhaustive search with linear regression (ES-LiR), and least absolute shrinkage and selection operator (LASSO) models to predict the coordination energies between 5 types of cations and 70 solvents. They found that the ionic radius and atomic charge of oxygen atoms connected to metal

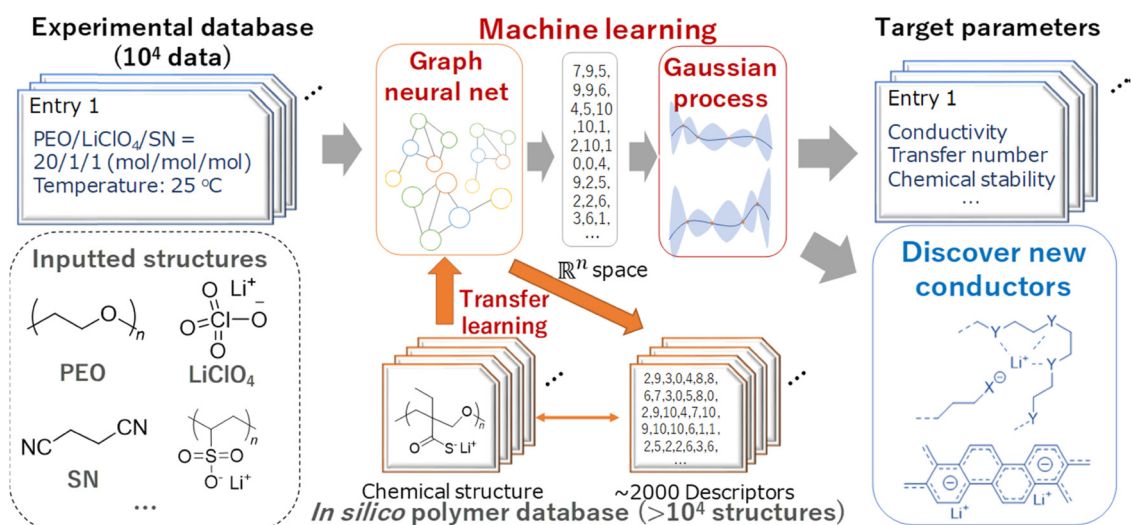


Fig. 3 An ML scheme used for new ionic conductor discovery. Reproduced with permission from ref. 36. Copyright American Chemical Society 2020.



When using the structure-property relationship for new electrode and electrolyte predictions with ML models, the features or descriptors are mainly described by the associated chemical structural properties such as the elemental properties, bonding information, and composition ratio. As such, materials defined by atoms, composition and structure (ACS)⁶² should provide crucial information for the ML predictions. Moreover, the sizes of the datasets in these works are typically from a few hundreds to tens of thousands, depending on the ML models used. Furthermore, the most common database used for screening and extracting features is the Material Project.^{20,21} The challenges for these ML approaches include the need for optimizing many model hyperparameters, the sensitivity for feature selections, the use of large datasets, and the overfitting issue.

There has been strong interest in developing MLPs for battery material discovery in recent years (Table 3). The NN potentials are one of the most used MLPs. Houchins *et al.*⁷⁶ developed NN potentials using atom-centered symmetry functions to study the NMC cathode materials. They showed that the predictions of structural properties (lithium insertion and removal) and voltage profiles of NMCs with any compositions can be conducted quickly with high fidelity. Eckhoff *et al.*⁷⁷ developed HDNN potentials to predict properties of the $\text{Li}_x\text{Mn}_2\text{O}_4$ cathode such as lattice parameters, diffusion barriers, volume changes, phase transition and phonon frequencies. They showed

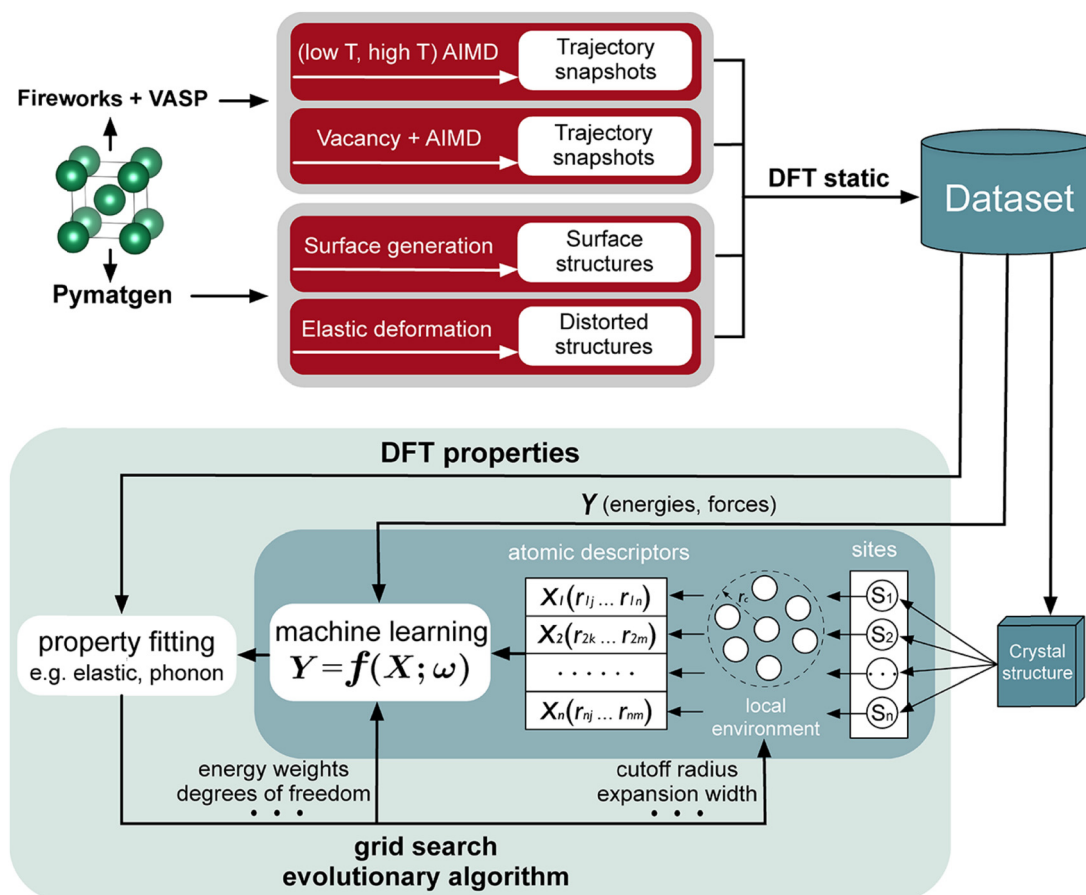


Fig. 4 A machine learning interatomic potential development workflow. Reproduced with permission from ref. 70. Copyright American Chemical Society 2020.

that the HDNN potentials can be used to represent a system with multiple oxidation states with accuracy comparable to DFT and it also enables nanosecond MD simulations. Artrith *et al.*⁷⁸ used ANN potentials and genetic algorithms to study the amorphous structures of Li_xSi as the anode for LIBs. They confirmed the predicted metastable structures with molecular dynamics heat-quench simulations and found consistency within the low-energy range.

For solid-state electrolytes, Huang *et al.*⁷⁹ developed a deep potential generator (DP-GEN) to predict the diffusion coefficients in $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$ -type solid-state electrolytes. They found that the computed data agreed well with the experimental data and they pointed out that the effect of configurational disorder is material-dependent, *i.e.*, some could obtain improved diffusion coefficients, while others may not. For liquid electrolytes, Dajnowicz *et al.*⁸⁰ used HDNN potentials, a charge recursive neural network (QRNN) architecture and DFT data to predict the thermodynamic properties of liquid carbonate solvents, such as specific heat at constant volume, density, heat of vaporization, viscosity and self-diffusivity. They found that the model is $\sim 10^4$ faster as compared to the calculations using wB97X-D/6-31G* with a single CPU (2.4 GHz). Moreover, the model can simulate MD with a simulation time of a few nanoseconds.

To further improve the accuracy of MLPs, active learning can be applied in the development of MLPs to deal with the unrealistic structures being generated during model training. Those structures can be further optimized by additional DFT calculations. Using learning-on-the-fly MTP (LOTF/MTP) potentials for MD simulations, Wang *et al.*⁸¹ discovered $\text{Li}_3\text{Sc}_2(\text{PO}_4)_3$ and $\text{Li}_3\text{B}_7\text{O}_{12}$, together with 10 other candidates, to be promising coating materials with high lithium contents and stable electrochemical windows (oxidation limit); low chemical reactivity with sulfide electrolytes and oxide cathodes; and low Li-ion migration energies. The MD can run up to 200 ns. Miwa *et al.*⁸² used a self-learning and adaptive database (SLAD) approach to develop ML potentials for simulating the ionic conductivity (σ) and activation energy (E_a) of $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$ (LGPS). The simulated results of $\sigma = 12 \text{ mS cm}^{-1}$ and $E_a = 226 \text{ meV}$ agreed well with experimental data. In addition, they found only small anisotropy effects for Li-ion diffusion in LGPS.

Hajibabaei *et al.*⁸³ developed sparse Gaussian process potentials (SGPPs) with a data-efficient on-the-fly adaptive sampling algorithm to simulate the Li diffusivity and melting/crystallization temperatures in $\text{Li}_7\text{P}_3\text{S}_{11}$. They discovered an uncharted phase with a much lower Li diffusivity that should be avoided and found that the computational cost is similar to those of the Bayesian linear regression methods. Using NN potentials and



Table 3 Recent advancements in battery materials using ML potentials

Materials	Battery	ML potentials	Descriptor/feature/input	Dataset	ML prediction	Accuracy	Ref.
LiNi _x Mn _y Co _(1-x-y) O ₂ (NMC) cathode	Lithium-ion	NNP	Atom-centered symmetry functions	12 962 points	Voltage profiles	RMSE: 3.7 meV per atom, 0.13 eV Å ⁻¹ for energy, force, Voltage: agrees with experiment	76, Houchins <i>et al.</i> (2020)
Spinel lithium manganese oxide cathode	Lithium-ion	HDNNP	Many-body atom-centered symmetry functions	15 228 Li _x Mn ₂ O ₄ structures	Lattice parameters, diffusion barrier, <i>etc.</i>	RMSE: 1.8 meV per atom, 0.108 eV/a ₀ for energy, force, Lattice: <1%, Barrier: agrees with experiment	77, Eckhoff <i>et al.</i> (2020)
Amorphous Li _x Si anode	Lithium-ion	ANN	Chebyshev descriptors	1000 first-principles calculations	Li _x Si configurations	RMSE: 7.7 meV per atom, MAE: 5.9 meV per atom	78, Artrith <i>et al.</i> (2018)
Li ₁₀ GeP ₂ S ₁₂ -type SSE	SSB	DP-GEN	Atomic environment descriptors	590 structures	Diffusion coefficient (DC)	RMSE: 2 meV per atom, 80 meV Å ⁻¹ for energy, force, DC: between 5–15 × 10 ⁻¹² m ² s ⁻¹	79, Huang <i>et al.</i> (2021)
Liquid electrolytes	Lithium-ion	HDNNP	Symmetry function	Training 360 K	Bulk thermodynamics properties	RMSE: 0.40 kcal mol ⁻¹ , 0.11 Debye, 0.47 kcal mol ⁻¹ Å ⁻¹ for energy, dipole, force	80, Dajnowicz <i>et al.</i> (2022)
Li ₃ Sc ₂ (PO ₄) ₃ and Li ₃ B ₇ O ₁₂ coating materials	SSB	LOFT/MTP	Polynomial basis	Initial set: 15ps AIMD at 1000 K	Ionic conductivity	MAE: 5.7 meV per atom, 84.3 meV Å ⁻¹ for energy, force, Ionic conductivity: MAE = 0.13 eV	81, Wang <i>et al.</i> (2020)
Li ₁₀ GeP ₂ S ₁₂	SSB	SLAD	Power spectrum	7500 local atomic geometries	Ionic conductivity at 300 K, activation energy	MAE: 7.1 × 10 ⁻⁵ Hartree per atom, 4.3 × 10 ⁻³ Hartree per Bohr, and 0.13 GPa for energy, force, and stress, Ionic conductivity: agrees with experiment	82, Miwa <i>et al.</i> (2021)
Li ₇ P ₃ S ₁₁	SSB	SGPP	SOAP	705 LCE (local chemical environment)	Li diffusivity	RMSE: 0.14 eV Å ⁻¹ for force, Li diffusivity: agrees with experiment	83, Hajibabaei <i>et al.</i> (2021)
Ether-based ionic liquid electrolytes	Metal-ion	NNP, active learning	Graph convolution	Initial 5000 geometries for training	Cation-ligand binding structures	RMSE: 0.83 kcal mol ⁻¹ , 0.5 kcal mol ⁻¹ Å ⁻¹ for energy, force	84, Wang <i>et al.</i> (2020)

active learning, Wang *et al.*⁸⁴ investigated the binding structures between cations and ether-based solvated ionic liquids for the discovery of new electrolytes. They identified candidate ether ligands for Li⁺, Mg²⁺ and Na⁺-solvated ionic liquids with higher binding affinity and electrochemical stability than the reference compounds.

The MLP approach has emerged as a promising method for battery material discovery. Different types of MLP have been developed for simulating battery material properties such as the ion diffusion barriers of electrolytes and voltage profiles of electrodes. The MLP accuracy is high with typical RMSE less than a few tenths of meV in energy and meV Å⁻¹ in force, which enables highly accurate property predictions. Moreover, new materials have been identified using the MLP approach: Li₃Sc₂(PO₄)₃ and Li₃B₇O₁₂ have found promise as coating materials for solid-state batteries using the moment tensor potentials.⁸¹ For the ML models, the neural network algorithms are the most frequently used for constructing MLPs as the NN approach is suitable for large system simulations.⁷⁷ It also has the transferability across composition space.⁸⁴ For the local atomic environment descriptors, the many-body symmetry functions by Behler and Parrinello^{71,72} are most often used. The descriptor built on the symmetry functions remains a smooth function of the atomic coordinates and can still allow atoms to go across the cutoff region during atomic simulations.

Nevertheless, challenges remain for the development of MLPs. One issue is that the long-range electrostatic effects

are usually ignored due to the local nature of MLPs. Staacke *et al.*⁸⁵ found that the description of lithium transport in the isotropic bulk Li₇P₃S₁₁ ionic conductor is valid without the long-range electrostatic terms in the MLP, but defect formation energies can only be obtained accurately with hybrid potentials and a physical model of electrostatic interactions. This points the way for future development of MLPs. Another issue is the transferability of MLPs, which depends on the extent of inclusion of relevant atomic environments in the reference/training data. Large prediction errors can result if the simulations are out of the range of validity.

3.3 Inverse design

The inverse design strategies start with the desired functionalities of materials and end in chemical spaces with the specific material structures. Ideally, one can start with a desired functionality and search for the potential components of the materials directly. However, due to the huge and growing chemical spaces, it is almost impossible to identify the governing rules or components, without the inputs from domain knowledge and optimization techniques. A good optimization technique should explore only a small amount of data, which is guided by the design principles, *e.g.*, a physics-based model that governs functions and structures.⁶²

The generic strategies for inverse design are:

1. High-throughput virtual screening: a screening scope is first defined and an ML model is employed to carry out a search



of databases. The search results often need to be verified with experiments or physics-based model simulations and domain knowledge plays a crucial role for the search success.

2. Global optimization: the approach employs optimization algorithms such as PSO, BO and GA to optimize an optimal solution for a large objective function. This approach is often employed for problems with multiple targets.

3. Generative models: this is an unsupervised learning approach in ML that can learn the input patterns and generate output with new data. Some examples are recurrent neural networks (RNNs), generative adversarial networks (GANs) and auto-encoders (AEs). Details of these strategies are well reviewed in ref. 86 and 87.

We highlight some of the recent battery material discoveries using the inverse design technique (Table 4). Sendek *et al.*⁸⁸ used a log *R* model and high-throughput screening of more than 12 000 computational structures to look for potential superionic conductors that are likely to have fast lithium conduction compared to experimental measurements. A total of 21 structures were identified, but a few are yet to be studied experimentally.

The global optimization strategy is one of the most common approaches in inverse design for battery materials. Liow *et al.*⁸⁹ used an inverse design surrogate model for correlating an experimental parameter–property relationship to predict optimal experimental synthesis parameters for NMC cathodes. They used a PSO to perform global optimizations and found that the composition of Ni, sintering temperature, cut-off voltage and charge-rate are strongly correlated with the performance of the materials. With the optimized parameters, the ML model achieved a high discharge capacity (209.5 mA h g^{−1}) and Coulombic efficiency (86%) for the batteries. Using an ANN model, Takagishi *et al.*⁹⁰ predicted the specific resistance of porous electrodes for LIBs. They found that the ML predictions agree well with the simulated results using a physico-chemical model. In addition, they performed inverse design to determine

the optimal process parameters and found that active materials with 50.4% volume fraction, binder/additives with 0.0820% volume ratio, electrolytic conductivity with 1.00 S m^{−1} and compaction pressure of 590 MPa are the key parameters for determining the total specific resistance. Li *et al.*⁹¹ used a multi-target ML model based on RF for inverse design of MXenes as high-capacity energy storage materials. They predicted the electrochemical properties with multiple targets (voltage, capacity and charge), followed by the use of the inverse design model to predict the formula of MXenes. Specifically, Li₂M₂C and Mg₂M₂C (M = Sc, Ti, Cr) were predicted to have the desired electrochemical properties.

Tagade *et al.*⁹² developed a deep learning inverse prediction framework (structure learning for attribute-driven materials design using novel conditional sampling (SLAMDUNCS)) to predict molecules with target properties. The framework was applied to predict electrolyte additive molecules with reduction potential smaller than −3.35 V against the standard hydrogen electrode. Some predicted molecules include dimethyl silyl carbonate and 2,4,6 triisopropyl-1,3,5-trioxane. In addition, using a GPR model and the active learning framework based on BO, Doan *et al.*⁹³ predicted the oxidation potentials of 1400 homobenzylic ether (HBE) molecules and identified a total of 133 molecules with the desired potentials ranging between 1.40 and 1.70 V for redox flow batteries. The schematic representation of the BO guided computational workflow for predicting HBEs with desired oxidation potentials is shown in Fig. 5.

Many new battery materials have been discovered with advancements in inverse design approaches. For instance, Li₅B₇S₁₃, Li₂B₂S₅, and Li₂GePbS₄ are among the 21 newly identified ionic conductors.⁸⁸ Also, Li₂M₂C and Mg₂M₂C (M = Sc, Ti, Cr) MXene have been suggested as promising battery electrode materials⁹¹ based on inverse design. Moreover, the inverse design approach has been used to optimize process parameters and the manufacture of battery electrodes with desired properties such as high charge capacity⁸⁹ and low resistance.⁹⁰ The neural network

Table 4 Recent advancements of battery materials using inverse design approaches

Materials	Battery	ML method	Inverse design strategy	Target	ML prediction	Accuracy	Ref.
Identified 21 promising electrolytes	SSB	Log <i>R</i>	High-throughput Screening	PLR ≥ 50%; $E_{\text{gap}} > 1$ eV; $V_{\text{ox}} \geq 4$ V, no transition metal; $E_{\text{hull}} > 0$ eV	Potential superionic structures	90%	88, Sendek <i>et al.</i> (2017)
LiNi _x -Co _{1-x-y} Mn _{1-x-y-z} O ₂ (NCM) cathode	Lithium-ion	kNN, RF, multiple imputations by chained equations (MICE)	PSO	Target discharge capacities of 150, 175, and 200 mA h g ^{−1}	Specific charge capacity, process parameters	RMSE: 8.17 mA h g ^{−1}	89, Liow <i>et al.</i> (2022)
Porous electrode	Lithium-ion	ANN	Bayesian	From 5 initial conditions tests (converged to 47 Ωm)	Specific total resistance, process parameters	R^2 : 0.827	90, Takagishi <i>et al.</i> (2019)
MXene electrodes	Metal-ion	RF	RF classifier	Target M, T, and Z (categorical descriptor)	Formula for MXenes	F1 score: 0.5833–0.9861	91, Li <i>et al.</i> (2022)
Electrolyte additives	Lithium-ion	DL	Bayesian	Target < −3.35 V against standard hydrogen electrode	Redox potential	MAE: 0.2004 V	92, Tagade <i>et al.</i> (2019)
133 molecules identified	Redox flow	GPR	Bayesian	Target 1.40 V, 1.70 V vs. NHE	Oxidation potential	RMSE: <0.15 V	93, Doan <i>et al.</i> (2020)



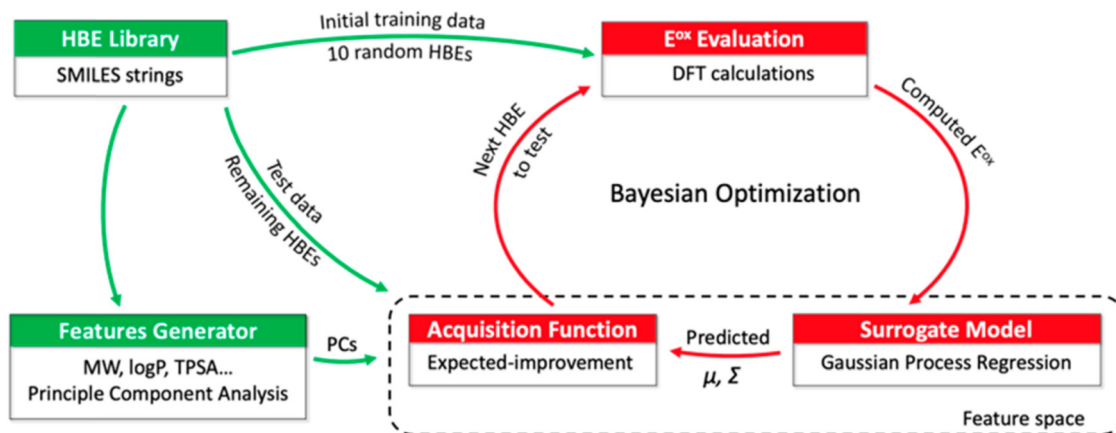


Fig. 5 A schematic representation of BO guided computational workflow for identify HBEs with desired oxidation potentials. PCs, μ and Σ are principle components, mean, and variance, respectively. Reproduced with permission from ref. 93. Copyright American Chemical Society 2020.

model is more often used in these studies while the global optimization technique (especially the Bayesian approach) is the most often used for inverse design strategies. The Bayesian optimization approach is based on Bayes Theorem that allows for a direct search of a globally optimized objective function efficiently using probabilistic principles. It is often used to tune hyperparameters of a given ML model. Nevertheless, there remain challenges for the inverse design approach as searching the huge chemical spaces is daunting. Bhowmik *et al.*⁹⁴ pointed out that extensive multi-fidelity datasets from multi-scale simulations, large-scale operando characterizations, and high-throughput syntheses need to work closely together to tackle the difficulty in solid electrolyte interphase (SEI) inverse design for batteries. The same suggested approaches should also be applied in battery electrodes and electrolyte design.

4. Outlook and conclusions

The development of ML techniques and databases has identified many novel electrode and electrolyte materials with promising properties. ML has also empowered the development of modeling techniques such as MD with faster and longer timescale simulations. In addition, the inverse design approach has become more effective with the growth of databases. These three approaches have accelerated innovation in battery materials significantly in recent years. While efforts are still needed to realize the potential shown by the predicted materials, data-driven ML approaches have been proven to be useful in the material design spaces.

Nevertheless, there remain some major challenges for the ML approaches. The most common challenge identified in the literature is data scarcity. There is a contradiction between high dimensions and low amounts of data in battery materials,¹⁰ meaning substantial dimensions are required for an accurate model but the data volume is limited, which might lead to overfitting issues and errors in predictions. Nevertheless, feature reductions and active learning are some effective approaches to mitigate such a limitation.

As opposed to the existing size of property data on organic molecules, *e.g.*, the Chemical Universal Database contains over 166 billion data entries (GDB-17),⁹⁵ and the existence of platforms for unified molecular analysis,⁹⁶ the lack of coherent big data in the battery field for ML techniques, especially for inorganic materials, remains the major limitation for accelerating the discovery of novel materials and improving battery performance. The absence of data standardization has often been raised,^{11,12,15,97} as the reported data across databases may not be in the same formats, terms and metadata schemas, especially computational data obtained from different levels of modeling and experimental data generated by different laboratories; this poses a challenge to unify coherent data for training and validation.

Nevertheless, there have been efforts towards a unified description of battery data. To address the issue, the FAIR (findability, accessibility, interoperability and reusability) data principles^{98,99} provide guidelines on scientific data management, which enable a new horizon for materials research. The FAIR data infrastructure requires in-depth description of how the data were obtained, including metadata, ontologies, and workflows. In particular, an ontology is the knowledge that can be expressed as a map of concepts and relations and expressed in machine-readable code.¹⁰⁰ For instance, the ontology is introduced to describe the production of lithium-ion batteries and support the management of data on materials and process steps.¹⁰¹ With approaches that can unify battery data and make it interoperable, this can alleviate the issue of data scarcity in battery material design.

We also notice that the majority of datasets currently available are static data; time-dependent battery data, *e.g.*, experimental battery cycling data, or computational molecular dynamics trajectory data, are rarely available but should be useful for predicting the complex dynamic behaviors in batteries. Therefore, high-volume, high-quality, unified and coherent static and dynamic datasets are essential for tackling this major challenge.

Finally, towards the development of green batteries, the concept of sustainability is crucial for the next generation of batteries.



To conclude, the present review has summarized recent ML-inspired battery electrode and electrolyte advancements. We provide insights into the discovery of these new battery materials through the ML strategies on direct property predictions, machine learning potentials and inverse design. We envision that to move towards green and more sustainable batteries, the integration of battery materials, manufacturing and product designs is essential. Data-driven machine learning is thus a promising technique to fulfil this mission.

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