

Energy storage in supercapacitor researches: Interdisciplinary applications from molecular simulations to machine learning

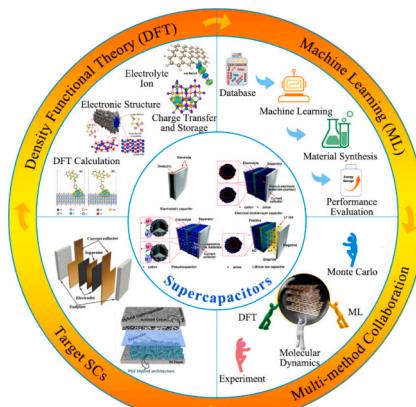
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HIGHLIGHTS

- A comprehensive overview of computational methods for SCs is provided.
- The role of both DFT and ML in SCs development and optimization is emphasized.
- The review bridges theoretical calculations with practical applications of SCs.
- Collaborative research using multiple methods for SCs is summarized.
- Future trends and challenges in the evolving field of supercapacitors are discussed.

GRAPHICAL ABSTRACT



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ABSTRACT

Sustaining scientific attention is aimed at the supercapacitors (SCs), which are significant for environmental protection and energy storage. The properties of the SCs are built on capacity, cycling stability, power and energy density, etc., in which the performances of electrode materials, interaction between electrode and electrolyte and charge transfer on the surface or interlayer of electrode vastly affect the overall abilities of SCs. In SCs research field, computational simulation applications are crucial for their simulating calculation and prediction capabilities. This review provides a comprehensive overview of the latest advancements in using density functional theory (DFT) and machine learning (ML) techniques to design and optimize SCs. We summarize the applications of DFT in understanding the electronic structure, charge storage mechanisms, and electrochemical properties of electrode materials, as well as the interactions between electrodes and electrolytes. Additionally, the role of ML in predicting SC performance, optimizing material design, and monitoring the state of health (SOH) of SC devices have been highlighted. The combination of DFT and ML offers a powerful approach to accelerate the discovery of new materials and improve the overall performance of SCs. On this basis, the integration of additional computational techniques such as molecular dynamics (MD) and Monte Carlo (MC) simulations further complements and enhances the capabilities of analysis and prediction. By integrating DFT, MD, MC simulations and

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ML, researchers can not only gain comprehensive insights into the complex behaviors of electrode materials but also significantly accelerate material screening through this synergistic computational approach.

1. Introduction

Fossil fuels belong to non-renewable energy sources that make a series of environmental pollution problems, which are aggravating the evil of the global ecological environment system change [1]. Developing advanced energy storage systems to efficiently collect renewable energy sources is an effective way, especially for the development of high-performance SCs is an important research direction to meet the future large-scale energy storage. SCs have advantages such as high-power density, excellent cycle stability, fast charging rate and relatively low production and maintenance costs, filling the gap between batteries and traditional capacitors [2,3]. As shown in Fig. 1, SC devices consist of activated materials, electrolytes, and other inactive components such as fluid collectors, conductive agents and binders, etc. [4] The electrolyte mainly plays the role in charge transfer and balance between the two electrodes, and the electrode materials are the place where the energy storage process occurs. The design and synthesis of electrode materials as well as the interaction mechanism between electrode materials and electrolyte are the main research orientations of constructing high-performance SCs.

With the improvement of computer performance and the increasing demand of material design research, computer simulation methods are emerging as a complement to basic theoretical and experimental research. Compared with experimental research methods, computer simulation technology has the advantages of low cost, excellent safety performance, high accuracy, atomic scale observation of intrinsic physical properties and chemical reactions. In recent years, the research objects based on computer simulation methods include not only atomic-scale nanomaterials but also bulk materials, which mainly include computational methods of quantum mechanics, MD simulation, ML methods, etc. Among them, the first-principles simulation method is based on the DFT of quantum mechanics, which is to solve the

Schrodinger equation with a set of approximately parameters without fitting to describe or predict the physical and chemical properties of the system. ML methods use different models to predict the electrochemical performance of SCs [5]. MD is a set of molecular simulation methods, which mainly rely on Newtonian mechanics to simulate the motion of a molecular system, and further calculate the thermodynamic quantities and other macroscopic properties of the system on this basis [6].

DFT calculation method is one of the most widely used methods to study electrode materials. For example, DFT calculations can be used to study the crystal structure of electrode materials, surface charge transport, specific capacity, interaction between electrode and electrolyte, and aging life of SCs, etc., and then interpret the experimental results from a microscopic perspective. In the 1990s, Ceder et al. made a pioneering application of DFT computing technology to design electrode materials for lithium-ion batteries [7]. Since then, DFT calculation has been applied more and more widely in the field of SCs. DFT method can calculate the electronic structure characteristics and band structure of electrode materials. Through these calculations, one can determine whether the material is conductive and further estimate its conductivity. For example, Peng et al. [8] constructed $Ti_3C_2T_x$ ($T_x = O$) electrode material and proved its excellent electrical conductivity through DFT calculation. By DFT calculations, the density of states (DOS) and the band energy of $Ti_3C_2T_x$ ($T_x = O$) verify the metal-like conductivities with exceeded states at the Fermi level (E_F) of $Ti_3C_2T_x$ ($T_x = O$) electrode. Furthermore, interaction between electrolytes and electrodes is important for electrochemical performance of SCs. For instance, Li et al. [9] obtained the adsorption energy of the material to water by means of DFT calculation, thus proving the binding site between the material and the electrolyte, and further proving the occurrence of the interface reaction between the material and the electrolyte. In recent years, the research on the energy storage mechanism of SCs by using DFT has also received extensive attention. For example, Hu et al. reported that Li^+ have a low

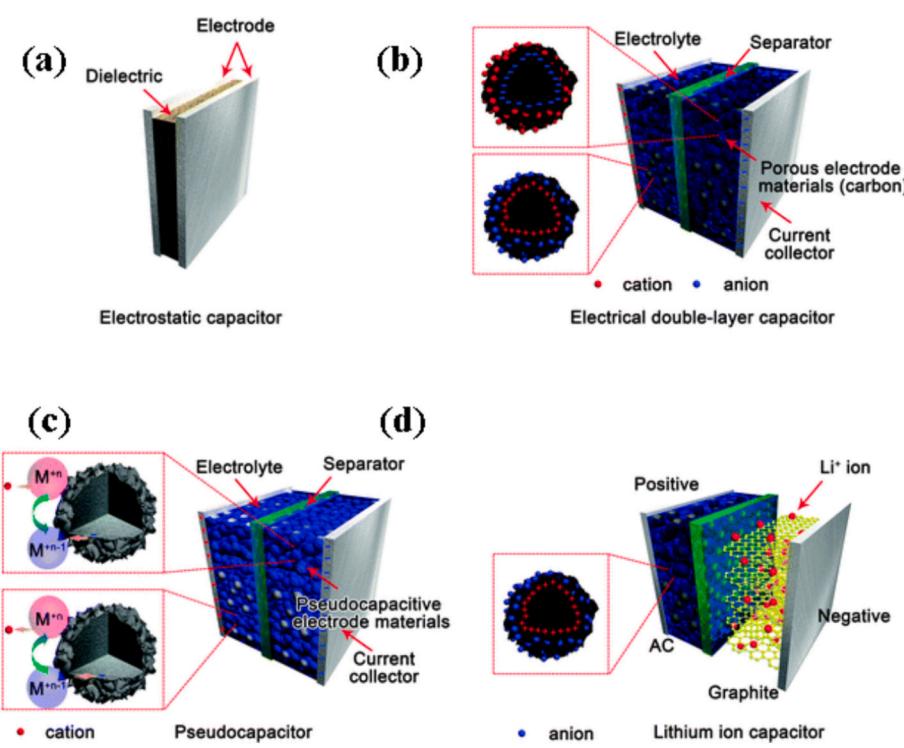


Fig. 1. (a) Static condenser. (b) Electric double layer capacitor. (c) Pseudocapacitor. (d) Hybrid SC [4]. Copyright 2015, The Royal Society of Chemistry.

diffusion barrier on (111) lattice plane of the electrode material based on the DFT calculation [10]. It can be seen from the above reports that DFT calculation is playing an increasingly important role in the field of SCs.

However, DFT often faces challenges related to computational complexity and the need for extensive datasets [11]. In contrast, ML provides an emerging data-driven alternative tool that can rapidly predict material performance based on existing data sets, identify key features affecting performance, and guide design optimization [12–14]. In the field of SCs, ML has been applied in various ways to enhance research and development. ML can create predictive models that simulate SC behavior under various operating conditions and employ techniques such as regression models, decision trees, and neural networks to predict the electrochemical performance of different materials, thereby enabling the optimization of electrode compositions and structures [15,16]. In addition, ML aids in feature importance analysis, helping researchers determine which material properties, such as surface area, porosity, and conductivity, are most critical for enhancing capacitance, energy density and lifespan. Moreover, its algorithms can analyze large datasets generated from experiments and simulations to uncover hidden patterns and correlations, allowing researchers to quickly identify promising materials and configurations, thus accelerating the material discovery process [17]. Recently, a variety of reports related to ML methods for the progress of SCs have emerged in the researches [18]. Zhou et al. [19] presented a method for predicting the remaining useful life (RUL) of SCs using a long short-term memory (LSTM) neural network, enhanced with the Adam optimization and Dropout algorithms to address overfitting. The study evaluated the impact of temperature and voltage on SC aging, demonstrating that the LSTM model achieved high prediction accuracy and robustness compared to other models like gated recurrent unit (GRU) and simple recurrent neural network (SIM RNN). Reddy et al. [20] developed an artificial neural network (ANN) model to predict the specific capacitance of carbon-based SCs by analyzing data from hundreds of research articles. They identified key input features, including specific surface area, pore size, and nitrogen / oxygen doping, and constructed 65 different ANN architectures to find the optimal model, achieving high accuracy ($R^2 > 0.99$) with a specific architecture. The findings highlight the significance of nitrogen / oxygen content and specific surface area in determining capacitance. Kaya et al. [21] proposed a data-driven research framework that combined a genetic algorithm (GA) with an ANN to optimize capacitance in carbon-based SCs. By identifying key design and operational parameters, the researchers developed an ANN model that accurately predicts capacitance performance, validated through both literature and experimental data. The hybrid ANN-GA approach not only yielded optimal parameter sets but also created design maps to illustrate the impact of tuning multiple features on capacitance. Furthermore, the integration of the Big M method enhanced the model's capability by eliminating infeasible feature combinations, making this framework a valuable tool for optimizing energy storage devices and applicable to a range of nonlinear mixed-integer problems in electrochemical energy storage.

Therefore, the ML method has been widely used in predicting the electrochemical performance and aging stability of SCs, making it a novel and effective prediction method.

In addition to the above research using a single calculation method, there are many reports involving the combination of two different calculation methods for research. Peng et al. [22] investigated the difference in capacitive properties between mesoporous carbon and microporous carbon. In this study, MD and DFT were introduced to study charge transfer and solvation of materials. Fontaine et al. [23] reported on a Faraday pseudocapacitance intermediate-thionine (Th)-in high-content LiTFSI, which was applied as a SC electronic solution to research the electrochemical storage mechanisms. The relationship between the structure and properties of the materials and the mechanism of charge transfer was studied by means of a combination of two computational methods, MD and DFT. Furthermore, by integrating ML

with traditional computational methods like DFT, researchers can adopt a more holistic approach to SC design, ultimately leading to the development of more efficient and effective energy storage solutions. Sial et al. [24] reported on a ML-based model for predicting the capacity and power density of SCs. In the meantime, by employing DFT, the electronic structure and properties of materials can be leaned deeply, thereby providing theoretical support for optimizing electrode materials. It can be seen that more and more computational methods have been applied to the research of SCs.

In recent years, we have published many reports on SCs. In 2019, the layered metal organic framework (MOF) was designed and synthesized, which was used as the electrode material for a SC device [25]. This result proves that the introduction of POM-based MOF materials into the interlayer of two-dimensional layered materials is a promising strategy to improve the electrochemical performance of SCs. In 2021, we [26] fabricated a SC with Fe-MOF and its congeneric derivatives as the negative and positive electrode, respectively. This SC shows a capacity of 224.3 C g^{-1} , at the current density of 1 A g^{-1} . In the same year, we [27] designed a battery-supercapacitor hybrid device with porous Co-MOF as the electrodes, which exhibited excellent electrochemical properties. This report verifies that porous MOF is a brilliant candidate for battery-supercapacitor hybrid devices. In 2022, we [28] designed and synthesized two isostructural iron-series-MOFs calcined derivatives and used them for the positive and negative electrode, respectively, in a SC device. This result shows that using isomorphic MOF as both positive and negative poles of a SC is a promising strategy to improve electrode matching in terms of improving the electrochemical performance of SCs. In addition, our research group [29] published a paper related to ML in 2024, which filled the gap in the toxicity prediction of silkworm using ML research and solved the previous limitation that only toxicity classification could be predicted without a specific toxicity value. However, our research on SCs still lacks some necessary theoretical calculation content. Therefore, here we review the application of theoretical calculations in the field of SCs, to facilitate the increased use of theoretical calculation methods in subsequent research. In this work, we provide a detailed review of the performance, structure, aging life and energy storage mechanism of SCs using a combination of one or several different computational methods.

2. Density functional theory for SCs

In recent years, a mass of theoretical researches has been used to forecast the highest precision and the applicability, providing support for laboratory findings, and their characteristics can substantially reduce uncertainties in models [30–32]. The well-regarded and broadly used computational quantum theory method for projecting chemistry and materials science properties is the density functional theory (DFT) computation [33,34]. Kohn proved that the ground state energy depends on the electron charge density, derived from Schrodinger's equation in the 1990s [33]. It was demonstrated that the corresponding electron cloud density distribution could be determined by settling quantitative equations for individual electrons [34].

Different computational simulation implements and methods dispose the DFT results [35]. Various software packages used for DFT calculations include Vienna Ab Initio Simulation Package (VASP) [36], DMol³ [37], Quantum Espresso Package [38] and Cambridge Serial Total Energy Package (CASTEP) [39], etc.

DFT computations offer messages on interactions between atomic nucleuses and electrons, the origin of valence chemical bonds between adjacent atoms, and enable the prediction of the chemical and physical properties of the materials. DFT calculations are widely used in many different fields including energy storage, electrocatalysis, photocatalysis, etc. DFT calculations reveal the characteristics of materials through an understanding of their structure, charges and ions transfer and interlamellar spacing, among other properties [40–45]. Moreover, DFT calculations have been applied to learn the structure of tunnels and

pores in materials, etc. [46] Researches demonstrate that DFT computations predict the performances of materials at the atomic-scale architecture [47]. DFT calculations have been used to research charge distribution in the theoretical models, such as solving the influence of ion dimension and shape [48–52] and the effect of ion transport on the characters and electrochemical properties of SCs [48,53,54]. The latter is important for energy storage in SCs [48,55]. Therefore, in recent years, many researchers have adopted DFT calculation in the field of SCs.

2.1. Application of DFT calculation in the study of supercapacitor mechanism

Firstly, the electronic structure of the electrode material, including its band structure, density of state and charge distribution, can be analyzed using DFT to understand the conductivity and charge storage capacity of the material. Secondly, the adsorption and desorption processes of electrolyte ions on the electrode surface can be simulated using DFT to reveal the interaction mechanism between ions and electrode materials, and how these processes affect the performance of SCs. Moreover, DFT calculations can be used to study the charge transfer and storage processes in the electrode material, including the charge distribution, transfer pathways and dynamic behavior, in order to deeply understand the charge storage mechanism of SCs.

2.1.1. Electronic structure of supercapacitor electrode material

The utilization of DFT computations for studying the mechanisms of SCs is very popular, and numerous studies have been verified to prove its significance for electrode materials [56–59]. The electronic structure of the electrode material, including its band structure, density of state, and charge distribution, can be analyzed using DFT to understand the conductivity and charge storage capacity of the material. The following part will focus on various electrode materials, such as transition metal carbides and nitrides (MXenes), MOFs and heterojunction materials, among others, to evaluate their properties and describe their DFT calculations used for SCs.

Recently, transition metal carbides and nitrides (MXenes), a new class of 2D layered materials, have marked an important turning point in the field of 2D layered materials since their discovery in 2011 [60]. MXenes possess characteristic structural performances, which contribute to their excellent stability and high storage capacity, attributed to their unique structure [61]. Therefore, they have been frequently utilized as electrodes for SCs [62,63]. The 2D $Ti_3C_2T_x$ MXene is a representative example of the MXene family [64,65]. The 2D layered MXene $Ti_3C_2T_x$ shows high conductivity and a regular interlayer structure, which contribute to its excellent performance as an electrode material for SCs [66,67].

Peng et al. [8] fabricated $Ti_3C_2T_x$ film using vacuum filtration, which exhibited good electrochemical performance. The binder-free structure of the $Ti_3C_2T_x$ film reduced the intrinsic resistance of the material and improved the transfer of ions and electrons. Moreover, the optimized

interlayer spacing of the $Ti_3C_2T_x$ film provided additional active sites, improving its electrochemical properties. The fabricated $Ti_3C_2T_x//NaCl-PVA//Ti_3C_2T_x$ -flexible solid-state supercapacitor (FSBSC) showed a good capacity of 112 F g^{-1} at 1 A g^{-1} and a maximum energy density of 62.3 W h kg^{-1} at 1000.8 W kg^{-1} , along with capacity retention of over 95 % after 5000 cycles. DFT calculations were executed to understand the metal-like conductivity of the $Ti_3C_2T_x$ ($T_x = O$) electrode. Fig. 2a demonstrates the crystallographic structure of pure $Ti_3C_2T_x$ ($T_x = O$). As shown in Fig. 2b-c, the density of states (DOS) and the band structure of $Ti_3C_2T_x$ ($T_x = O$) confirm its metal-like conductivity, with excess states at the Fermi level (E_F) of the $Ti_3C_2T_x$ ($T_x = O$) electrode.

To elucidate the charge storage mechanisms of modified MXenes, Chen et al. [68] systematically engineered three distinct MXene ($Ti_3C_2O_2$) architectures through strategic nitrogen doping and metal vacancy introduction: defective MXene ($Ti_{2.9}C_2O_2$), N-doped MXene ($Ti_{3-C_2O_1.9}N_{0.1}$), and N-doped MXene with metal vacancies ($Ti_{2.9}C_2O_{1.9}N_{0.1}$). These systems were investigated using DFT calculations coupled with the effective screening medium reference interaction site model (ESM-RISM) method to probe electrochemical behaviors at electrode/electrolyte interfaces. Complementing these theoretical insights, Xie et al. [69] experimentally fabricated flexible free-standing N-doped Ti_3C_2 ($Py-Ti_3C_2$) films and comprehensively characterized their charge storage mechanisms in aqueous hybrid SCs. Electrochemical analysis revealed dominant pseudocapacitive behavior, with the $Py-Ti_3C_2$ electrode exhibiting exceptional specific capacitance (in 1 M H_2SO_4). DFT calculations in conjunction with ESM-RISM and climbing image nudged elastic band methods (CI-NEB) were employed to elucidate the interfacial electrochemical phenomena.

Among tunable MOF structures, there is burgeoning interest due to their porous architecture. MOFs are frequently fabricated through coordinate bonds between metal centers and organic ligands [70]. Obviously, MOFs are taken as candidates for electrode materials in advanced energy storing devices according to the large surface area, porous structure and widespread active centers. However, conventional MOFs suffer from poor electroconductibility, due to the insulating nature of the ligands used for their synthesis, which limits their application in energy storage [71]. Moreover, DFT calculations can specifically describe the charge transfer between electrolyte and MOFs/MOF-derivatives, analyze the structure of MOFs/MOF-derivatives, and forecast their electrochemical performance [72–79].

Therefore, Ciesielski et al. [22] reported a MOF@covalent organic frameworks (COF)-7,7,8,8-tetracyanoquinodimethane (TCNQ) composite. The MOF@COF-TCNQ composite was used as an electrode in a SC device, which showed a capacity of 78.36 mF cm^{-2} . According to DFT calculations, the quantum capacity of MOF@COF composites was investigated. The DFT results confirmed that quantum capacities plays a key role in improving the electrochemical properties. The quantum capacitance of the MOF@COF composite was $26 \mu\text{F cm}^{-2}$, inferior to that of the MOF-UIO-66-NH₂-monolayer ($86 \mu\text{F cm}^{-2}$). After incorporating the TCNQ into the MOF@COF composite, the hybrid composite showed a value of $218 \mu\text{F cm}^{-2}$ (Fig. 3a-b). The introduction of TCNQ

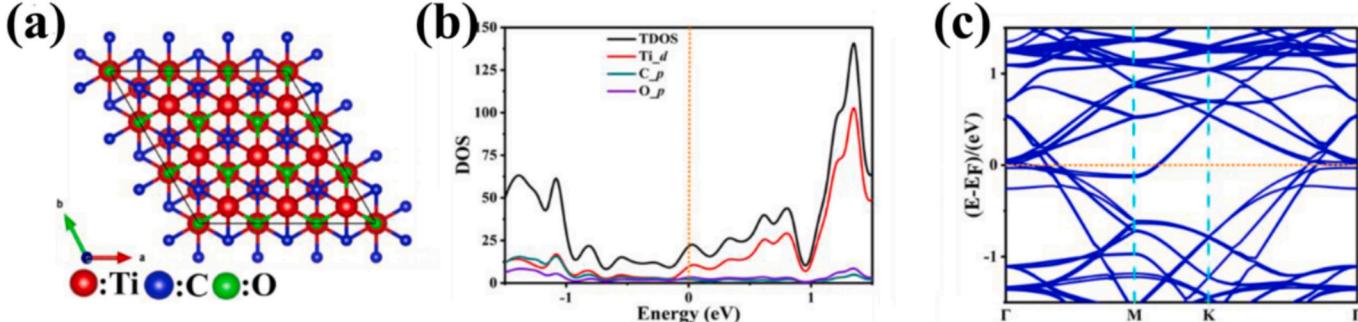


Fig. 2. (a) The crystal structure. (b-c) DOS and band structure of $Ti_3C_2T_x$ [8]. Copyright 2024, Elsevier Ltd.

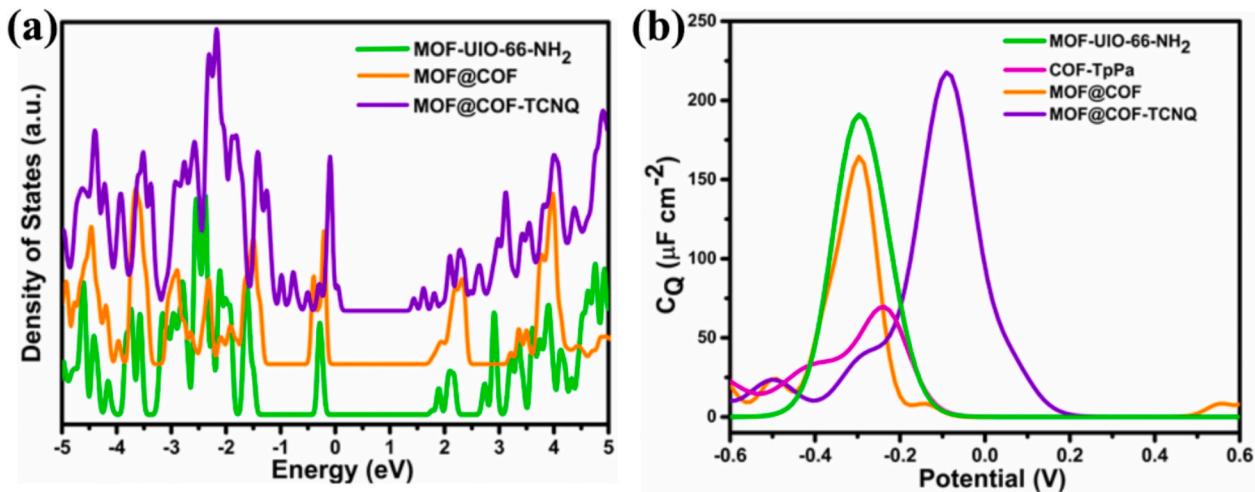


Fig. 3. Resulted DOS of (a) MOF-UIO-66-NH₂, (b) MOF@COF [22]. Copyright 2021 American Chemical Society.

molecules into the composite framework led to an improvement in quantum capacitance.

To improve the electrical conductivity of MOFs, Chen et al. [80] reported a bimetallic conductive MOFs, Co_{0.24}Ni_{0.76}-bpa-200, with a large capacity of over 1900 F g⁻¹ and a cycle retention of over 85 %. Based on the DOS results of Co-MOF, Ni-MOF and Co_{0.24}Ni_{0.76}-bpa-200, the resulting E_g (the energy gap of HOMO-LOMO) of Co_{0.24}Ni_{0.76}-bpa-200 (0.20 eV) is lower than that of Co-MOF (0.65 eV) and Ni-MOF (0.26 eV), indicating that ample charges can be excited from the valence-band maximum to the conduction band minimum in the Co_{0.24}Ni_{0.76}-bpa-200 electrode, thereby enhancing the carrier concentration. From the total DOS near the Fermi edge, the electron density of Co_{0.24}Ni_{0.76}-bpa-200 (129 eV) is almost twice that of Ni-MOF (64 eV), suggesting increased electrical conductivity [81].

The construction of heterojunction materials is a potential strategy to enhance the capacity and stability of electrode materials [82]. Heterojunction materials provide an attractive opportunity to utilize the excellent electrochemical performances at the interface, which obviously different from the dimension of several to dozens of nanometers

[83,84]. These heterojunction materials apparently promote the formation and alteration of tunable electronic structures for various applications [85]. The method for fabricating heterojunction materials, including defect engineering, interface modifications and surface modification, immediately affect the electro-capacity, cycling stability of these materials [86,87]. Therefore, manufacturing heterostructures can considerably improve ion and electron transfer, power density, etc. [88,89]

Zhang et al. [90] reported a heterojunction material (ZnIn₂S₄@CuO) possessing n-type zinc chalcogenides (Zn-In-S/C) and p-type copper oxide (CuO) as pseudocapacitive electrode materials. The symmetric SC equipment exhibited an energy density of 7 Wh kg⁻¹ at a power density of 4000 W kg⁻¹. Spin-polarized atomic simulations based on DFT were carried out to investigate the construction and the variation in the electro-performances of CuO and the heterostructure materials. The crystal structure of CuO is tetragonal, as display in Fig. 4a, and it is distinguished as a non-magnetic semiconductor, as indicated by the energy band [91]. In the meantime, the crystal structure of Zn(InS)₂ is cubic and also characterized as a non-magnetic semiconductor [92]. For

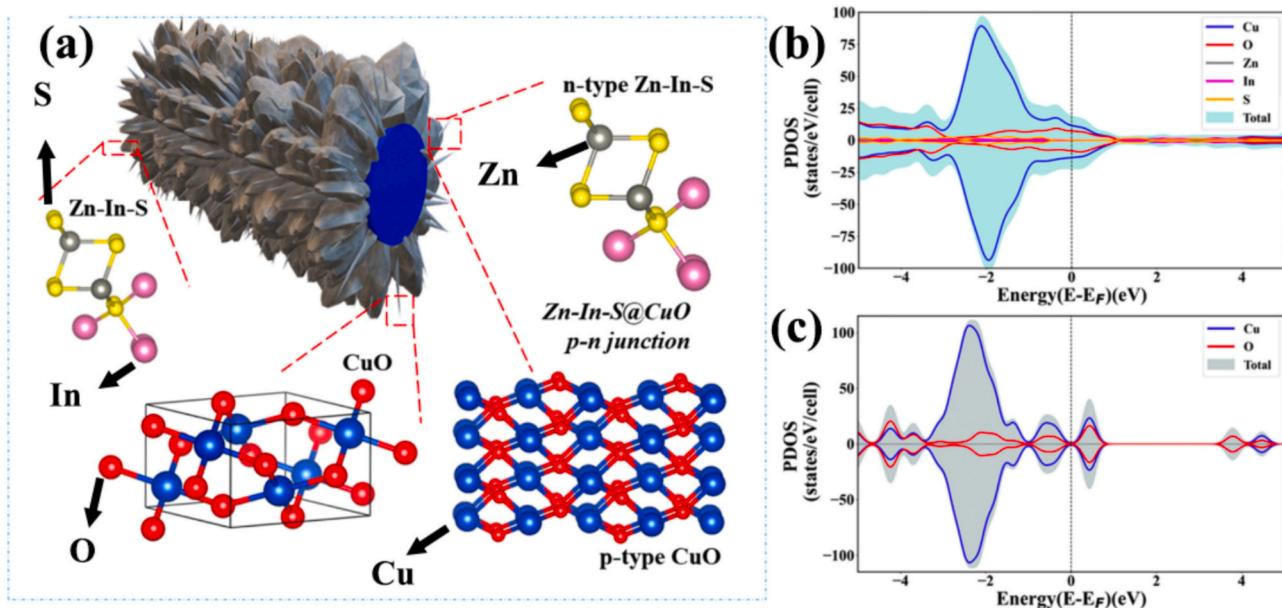


Fig. 4. (a) Schematic of ZnIn₂S₄@CuO, atomic structures of CuO, ZnIn₂S₄, and p/n junction. PDOS of (b) ZnIn₂S₄@CuO and (c) CuO [90]. Copyright 2023 American Chemical Society.

the simulating the heterojunction structure, a thick CuO plate with 4 layers was used, while a tiny cluster of ZnIn₂S₄ (Zn₂In₄S₈) was imported over the CuO layer at appropriate sites. The most stable configuration, with a high binding energy (E_b) value of -2.63 eV, was obtained. From the total density of states (TDOS) and partial density of states (PDOS) results of the ZnIn₂S₄@CuO heterostructure and CuO, as display in Fig. 4b-c, the ZnIn₂S₄@CuO heterostructure demonstrated more obvious metal-like behavior.

Zhou et al. [93] fabricated NiCo₂S₄-Ni₉S₈-C double-layered yolk-shell microspheres (NiCo₂S₄-Ni₉S₈-C DYMs). These materials contain abundant heterogeneous NiCo₂S₄-Ni₉S₈ nanoparticles (NPs). The electrochemical properties of NiCo₂S₄-Ni₉S₈-C DYMs were tested, showing a large capacity of 293.6 mA h g⁻¹ at a current density of 1 A g⁻¹ and cycling retention of over 85 % after 5000 cycles. To further investigate the effect of hetero-interfaces in NiCo₂S₄-Ni₉S₈-C DYMs, the Fermi energy (E_f) and work-functions (E_w) of the NiCo₂S₄ and Ni₉S₈ were calculated, respectively. Since the E_w of NiCo₂S₄ (6.17 eV) is smaller than that of Ni₉S₈ (6.81 eV), electrons are transferred from NiCo₂S₄ to Ni₉S₈ when an interface is formed [94].

2.1.2. Mechanism of electrolyte ion adsorption and desorption in SCs

Researching the mechanism of ion adsorption and desorption on electrode surface is important for studying the energy storage mechanism of SCs [95]. When a material is immersed in an electrolyte, an electric potential difference forms between the material and the solution, leading to electrochemical processes such as ion transfer and charge exchange at the interface. These processes deeply influence the properties of SCs. Different factors, especially the movement of ions in the solution and the material wettability, play a profound role in electrochemical performance [96]. However, some properties cannot be fully characterized experimentally, highlighting the need for theoretical explanations. Consequently, researchers have employed DFT calculations to address these problems.

The transformation from biowastes into valuable porous carbon has been broadly applied [97–101]. Activated carbon derived from biological wastes offers abundant pores, high specific surface area and good stability, making it an ideal electrode materials for SCs [102–104]. Yao et al. [105] designed an activated carbon from bamboo (named as PAB-600-350) and used it in the SC as an electrode. PAB-600-350 exhibited the capacity of 256 F g⁻¹, at the electric density of 1 A g⁻¹, and the symmetrical SCs delivered an energy density of 12.54 W h kg⁻¹ at the power density of 225 W kg⁻¹. The electrochemical properties of the material can be attributed to the reaction between the material and the interface, which DFT calculation can explain. As shown in Fig. 5a, the dipole moment of pure carbon is 0.005 Debye, but oxygen containing

groups improve the dipole moment, indicating that oxygen containing groups surface polarization. This enhanced polarization increases wettability and strengthens the interaction between materials and solutions. Fig. 5b exhibits the adsorption energy (E_{ads}) data of K⁺ on carbon models. Among them, it is obvious that the model containing carbonyl group shows the largest adsorption capacity for ions in solution. Additionally, charge delivery path ways were also deeply investigated and the differential charge density figures are shown in Fig. 5c. It is very clear that there are entirely new electron transport channels formed on the surface of the material containing carbonyl groups when K⁺ is adsorbed.

Phosphorus-doped carbon materials also show high electrochemical energy storage capacity due to improved wettability and electron transfer ability, as demonstrated by DFT calculations. Kim et al. [106] developed a SC by Ketjen black (KB) nanoparticles doped with phosphorus (P), accomplishing a capacity of 2.26 mF cm⁻² with an excellent electrochemical stability. Studies on the polarity of the material surface also rely on the corresponding electrostatic potential mappings of the DFT calculations, and the results also prove that doping of P can effectively improve the polarity of the material surface, thereby increasing its wettability. The adsorption energy (E_{ads}) results demonstrated strong interactions between the material and electrolyte ions (K⁺ and OH⁻), confirming that enhanced interfacial adsorption improves charge transport and ultimately boosts electrochemical performance of the material.

For some transition metal oxide materials, OH⁻ plays an important role in energy storage process via the redox capacity. Therefore, the transfer of OH⁻ in the interface between the material and the electrolyte is particularly important. Qin et al. [107] designed the Co-doped NiO materials ((Ni,Co)O@np-NiCo/MG), which shown a maximum capacity of over 900 F cm⁻³ and retained over 90 % capacity after 8000 cycles. As shown in Fig. 6a-b, DFT calculation shown the E_{ads} of the OH⁻ on NiO (-3.16 eV) and Co, Ni-based materials (-5.13 eV), respectively. These results confirm that stronger OH⁻ adsorption on Co, Ni-based materials, which further proves that Co-doped materials possess higher redox capacities and better electrochemical properties.

DFT calculations provide critical insights into understanding the charge transfer mechanisms and the underlying principles of fast kinetics in multi-component electrolyte systems. Through DFT simulations, researchers can systematically investigate the interactions between different components and their influence on electrochemical performance, thereby guiding the rational design of optimized electrolyte formulations. Suriyakumar et al. [108] investigated composite polymer electrolytes (CPEs) incorporated with ceramic fillers and in-situ formed fluorine-rich interphases for lithium anodes. By employing DFT

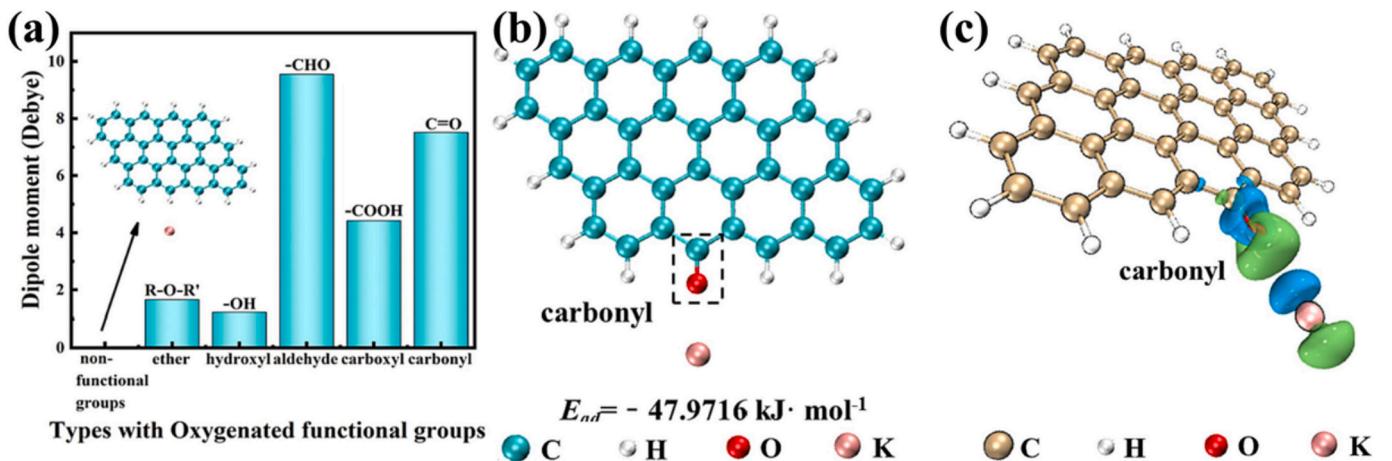


Fig. 5. (a) Dipole moments of the unoxidized surface and the other five surfaces containing ethers, hydroxyl, aldehydes, carboxyl and carbonyl groups. (b) Simulated structure of surfaces containing carbonyl groups. (c) K⁺ ion adsorption on simulated surfaces with carbonyl [105]. Copyright 2023 Elsevier B. V.

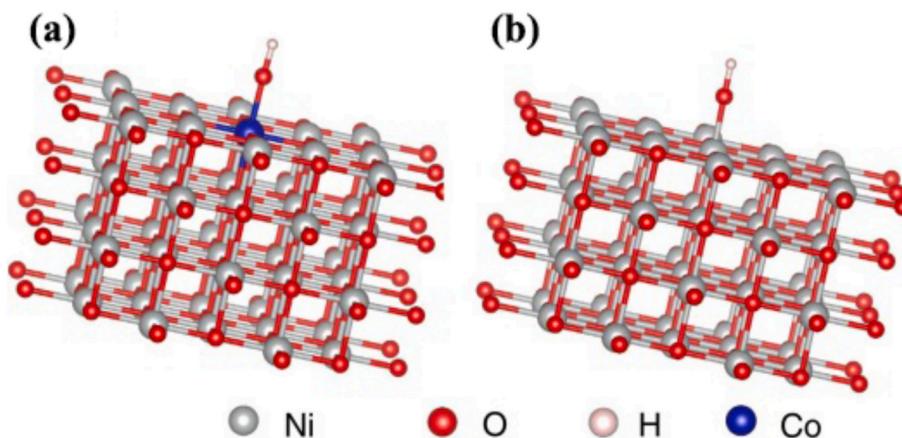


Fig. 6. The simulated structure of (a) $(\text{Ni},\text{Co})\text{O}@\text{np}-\text{NiCo}/\text{MG}$ (100) plane and (b) NiO (100) plane interacted with OH^- [107]. Copyright 2022 Elsevier B. V.

calculations to analyze the binding energies between lithium ions and ceramic-polymer interfaces as well as inter-interface interactions, they demonstrated that introducing specific additives between polymer chains could effectively weaken interchain interactions, thereby facilitating lithium ion hopping and significantly enhancing the electrochemical performance of devices. Li et al. [109] developed a ternary eutectic electrolyte composed of choline chloride, ethylene glycol, and urea for zinc-ion batteries. Complementary DFT calculations revealed that the dominant $[\text{ZnCl}_4]^{2-}$ cluster in the zinc solvation sheath exhibited the lowest plating activation energy (ΔG_p) in this electrolyte system. These results confirmed that the ternary eutectic electrolyte preferentially forms Zn^{2+} solvation structures that are thermodynamically favorable for reversible electrochemical redox reactions, consequently improving the reaction kinetics. Liu et al. [110] designed an artificial solid electrolyte interphase (SEI) consisting of gradient inorganic layers (LiF , Li_2S , and Li_2SO_3). DFT calculations of Li^+ diffusion pathways and energy barriers across various crystal surfaces demonstrated that the multi-component electrolyte configuration substantially reduced diffusion barriers. This enabled the $\text{LiF}/\text{Li}_2\text{S}$ -rich SEI to achieve exceptionally fast Li^+ transport kinetics, maintaining excellent electrochemical performance of lithium anodes even at high current densities while simultaneously improving cycle life and rate capability. Moreover, the homogeneous Li^+ distribution promoted by this SEI architecture effectively suppressed lithium dendrite growth.

These studies collectively highlight the pivotal role of DFT in advancing multi-component electrolyte research. DFT simulations not only elucidate charge transfer mechanisms at the atomic scale but also provide fundamental theoretical guidance for designing high-performance electrolytes. The integration of computational modeling with experimental approaches enables more efficient optimization of electrolyte compositions, ultimately leading to superior ion transport kinetics and enhanced electrochemical stability.

2.1.3. Charge transfer and storage mechanism in SCs

General capacitor mechanisms can be divided into two categories: electric double layer capacitor mechanisms and pseudocapacitor mechanisms. The model of electric double-layer capacitors was first constructed by Helmholtz who completely studied colloidal suspension in 1800s [111]. Different from electric double-layer capacitors, pseudocapacitors electrode materials have obvious Faradaic electrochemical properties, (i. e., oxidation-reduction reactions) during the charge and discharge process, indicating valence changes during charge and discharge process [112,113]. The pseudovapacitor electrode can achieve a larger capacity than double electric layer capacitors. Therefore, the mechanisms of storing charge in pseudocapacitors electrodes have been frequently investigated in recent years [114]. The capacities of pseudocapacitors mainly come from the redox reaction of electrodes.

These reactions are usually along with ions being adsorbed on the surface of the electrode material in electrolyte, resulting in a rapidly reversible transfer of charges and ions across the electrolyte/electrode interface [77]. In crystal structures, pseudocapacitors can also originate from the adsorption and transfer of ions and charges in channels and layers [115]. In the investigation of the charge transfer and storage mechanism of pseudocapacitors, DFT calculation is an indispensable auxiliary mean in addition to experimental analysis. By using DFT to calculate the crystal structure of the material and the adsorption energy of solution ions at different points on the material surface, combined with the DOS results adjacent the Fermi level, the electron transport path and mechanism can be obtained.

Wu et al. [116] synthesized an electrode material ($\text{CuCo}_2\text{S}_4@\text{CP}$) and firstly applied to aqueous ammonium-ion (NH_4^+) SCs (AASCs). The $\text{CuCo}_2\text{S}_4@\text{CP}$ electrode has a maximal capacity of 1512 C g^{-1} and a capacity retention of 87.74 % after 10,000 cycles. The accumulation energy mechanism between electrolyte and $\text{CuCo}_2\text{S}_4@\text{CP}$ electrode has been completely verified, and DFT calculation is significant in the research. Fig. 7a shows the structure of CuCo_2S_4 with a well-organized distribution of various atoms, constituting a frame that constructs the transport path of NH_4^+ . In Fig. 7d, the pristine CuCo_2S_4 performs a band gap of 0.5 eV. After NH_4^+ insertion (Fig. 7e), the DOS adjacent the Fermi level has a small enhancement, accompanied by a decrease in the band gap to 0.4 eV. Since Co^{3+} coordinated with S is a low spin state containing a lone pair of electrons, it is more likely to be converted to Co^{2+} when NH_4^+ is inserted. This change in the DOS adjacent the Fermi level may be related to an alteration in the valence state of the Co atom. The crystal structure and charge distribution model are shown at Fig. 7b-c, after NH_4^+ entrance into CuCo_2S_4 . Therefore, NH_4^+ transfers toward the $\text{CuCo}_2\text{S}_4@\text{CP}$ electrode during electro-discharge, gradually inserting into the layers of the material and then hydrogen-binding with adjacent S atoms.

Liu et al. [117] used the nature capsaicin-functionalized doped graphene (Cap-NG) as electrode for a symmetrical SC. The electrochemical results exhibit that the Cap-NG has a maximal capacity of 312 F g^{-1} and a retention of 102.9 % after 10,000 cycles. Additionally, DFT calculations are applied to clarify the root of charge transferring path way of Cap-NG electrode and calculate the DOS. As shown in Fig. 8a-b, Cap near the NG surface in cumulate sites with the adsorption forms (P2) and the sorptive ability 4.75 eV. The data suggest that the promising way of the charge change and transfer processes controlling by $\pi-\pi$ superposition is the adsorption way of Cap in P2 form [118]. As shown at Fig. 8c, a bigger charge repartition on NG is motivated by Cap in P2 form due to the greatly supplementary of the fatty chain involving NH_2 -group of Cap on NG with defect locations and aiming to higher electron cloud density, which benefited in charge transfer for long time. As shown at Fig. 8d, these peaks near the E_f in PDOS of O for Cap-NG are originated

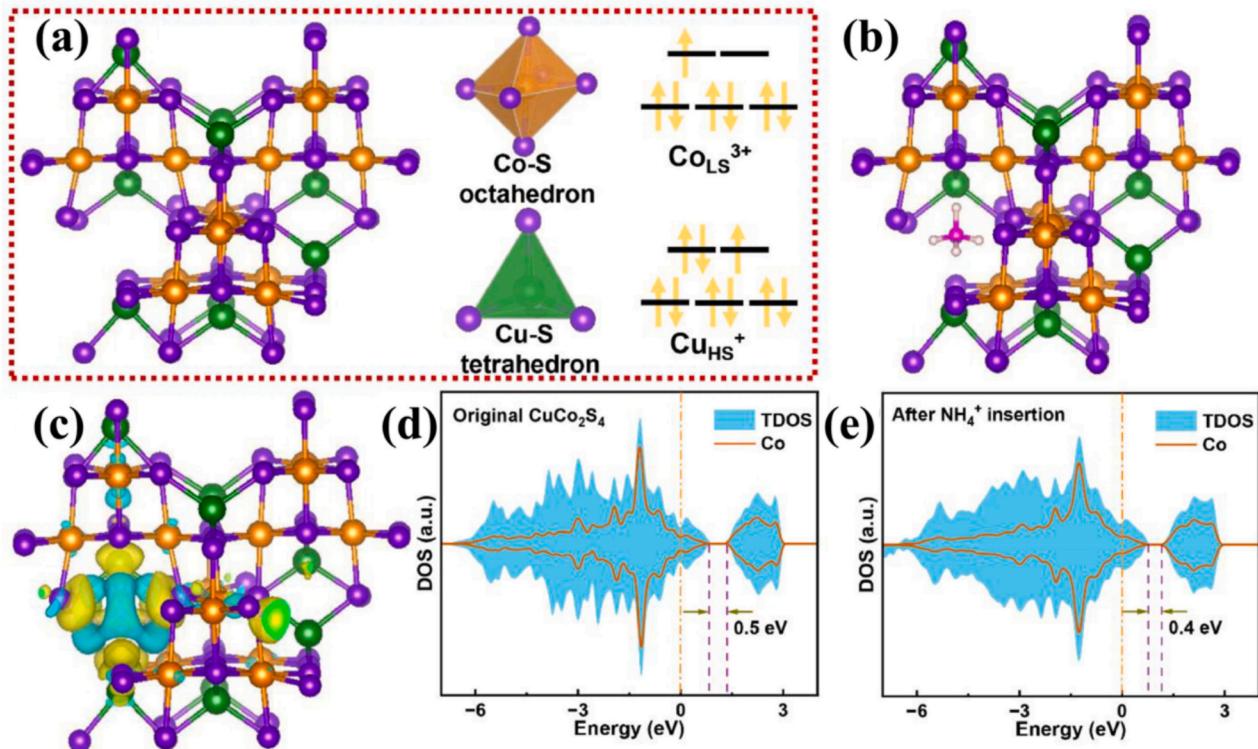


Fig. 7. (a) Schema of the crystal structure of CuCo₂S₄ and the electronic states of Co and Cu atoms. (b) Energy storage explanation of NH₄⁺ in CuCo₂S₄. (c) Charge distribution of NH₄⁺ in CuCo₂S₄. (d-e) DOS for Co atom before and after NH₄⁺ insertion [116]. Copyright 2024 Elsevier B. V.

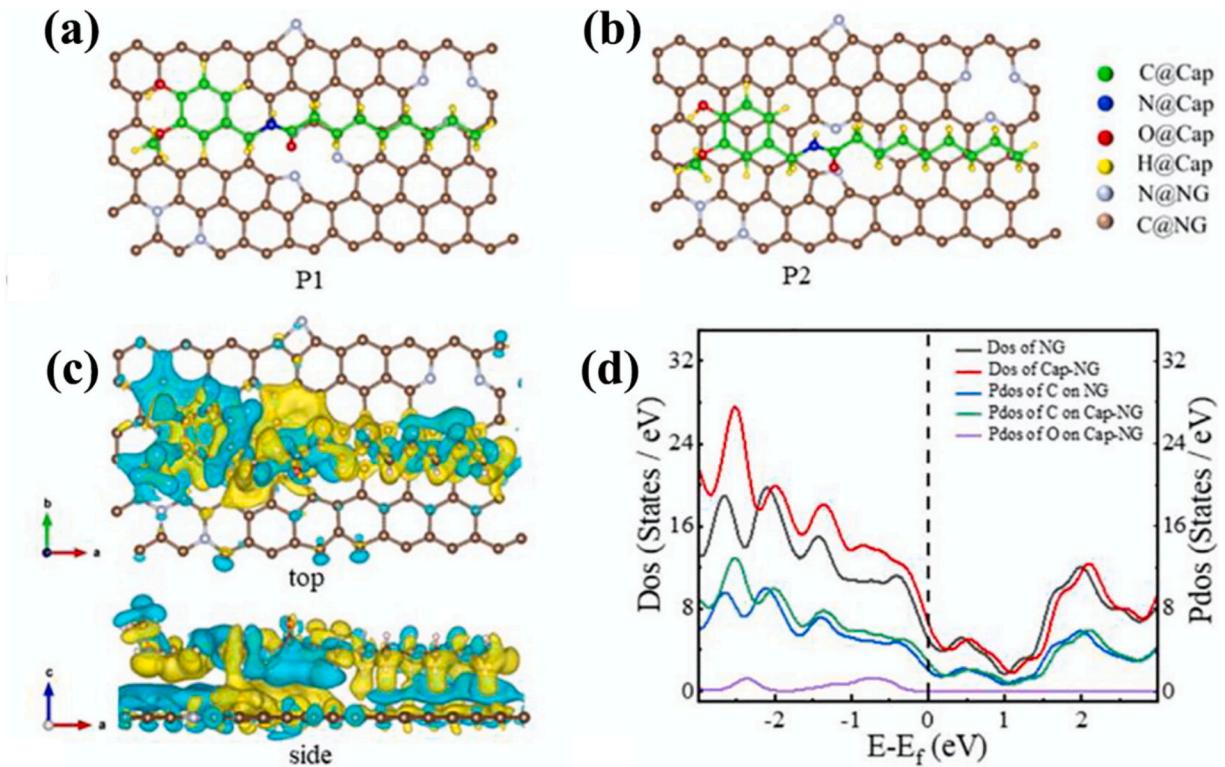


Fig. 8. (a), (b) Two possible schema of the simulated crystal structure of Cap on the NG sheet. (c) Top and bottom view of the charge density difference plot of the Cap over the NG sheet (at the mechanics of P2). (d) DOS for NG and Cap-NG; PDOS of C for NG and Cap-NG; PDOS of O for Cap-NG [118]. Copyright 2022 Elsevier B. V.

by O-2p orbital in Cap-NG, corresponding with the theory suggested beyond that oxygen-involving groups, like an access which can expedite the redox processes [119,120].

The DFT calculation results of differential charge density (CDD) can more clearly show the transport path of charge on the material surface. Zhu et al. [121] prepared $\delta\text{-MnO}_2$ -cetyltrimethylammonium bromide (CTAB) as electrode for SCs. To deeply dig the electron transfer mechanism of $\delta\text{-MnO}_2$ -CTAB materials. The CDD result indicates a charge transfer path from Na^+ to MnO_2 . Moreover, the planar average charge densities of MnO_2 and Na^+ clarify that Na misses charges and MnO_2 gains charges. Bader charge analysis exhibits that the shift from Na^+ to MnO_2 is almost 0.8 e. The minimum migration barrier energies (MBE) of Na^+ in the outside of MnO_2 is 0.04 eV, which suggests that Na^+ is likely to disperse to the surface or interlayer of MnO_2 .

Zhao et al. [122] prepared N-doped microporous carbon aerogels (NC-CMPAs) as electrode for zinc-ion hybrid SCs. For investigating the charge storage mechanism of Zn//NC-CMPAs SCs, the DFT methods were performed. For the NC-CMPAs electrodes, the Zn^{2+} binding energy changes is -2.5288 eV, indicating that the Zn^{2+} incorporation/ decoration procedure is more likely in NC-CMPAs. The charge movement can also be verified by CDD analysis upon insertion of Zn^{2+} . The charge gains and losses are occurred mainly in the Zn^{2+} and N/O sites.

Nudged elastic band (NEB) method was used to analyze ion diffusion [123]. Hu et al. [10] used a low-temperature lithiation strategy to fabricate hierachic $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ loaded on carbon cloth (LNMO@ACC). In fact, the LNMO@ACC electrode shows a capacity of almost 1400 mF cm^{-2} and cycling retention of over 80 % after 4000 circulations. The mechanism of charges and ions transfer on the lattice planes of $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ is illustrated by DFT computations. The E_{ads} of Li^+ on the cover of $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ is calculated. The E_{ads} of Li^+ stayed at $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ (400) and (111) lattice planes are -1.7 eV and -2.6 eV, respectively, which suggests that Li^+ can be conveniently adsorbed on the (111) lattice plane of $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$. Due to the NEB method, the diffusion limit of superficial Li ion is calculated, and investigate the transfer path ways of Li^+ on the $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ surface. As shown at Fig. 9a-b, the top view of the Li^+ transfer path ways on two planes of $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$. As shown in Fig. 9c, the barrier of Li^+ transfer on the (111) plane is smaller than (400) plane, verifying that Li^+ diffuse on the (111) plane more easily.

Before the last report, Hu et al. [124] prepared KCu_7S_4 /Graphene paper (GP) SCs, and revealed the specific charge transfer mechanism of KCu_7S_4 /Graphene paper (GP) SCs. Both electrochemical tests and DFT results show that the successive charge storage procedure is mainly originated by K^+ ions' adsorption and diffusion, where K^+ ions are verified to have been more positive than Li^+ ions in the pseudocapacitance reactions on the KCu_7S_4 surfaces and channels. Therefore, the mechanism of different ions at atomic scale by the DFT calculations are investigated [125,126]. And the NEB method is applied to conduct the tests of electrons' transfer in the KCu_7S_4 electrodes. The adsorption and diffusion path ways of K^+ and Li^+ are similar, both of which transfer on the surface of the channels with energy barriers of 0.3 eV and 0.73 eV,

respectively. The adsorption and diffusion path way of H^+ is relatively more complex, it can bond with S in the form of covalent bonds with strong bonding energy, and when the H-S covalent bond is broken, the activation energy of the fracture is as high as 1.5 eV. The corresponding charge path ways of the three ions are calculated by the NEB method, where the H^+ ions are more likely to insert into the KCu_7S_4 channels at the beginning, but then the capacity of H^+ ions insertion reduces fast with the addition in cycles since it has to challenge large energy barriers (41.5 eV).

Xue et al. [127] used a solid reaction with a ball milling method to synthesis channel structured Ti doped $\text{Na}_4\text{Mn}_9\text{O}_{18}$ nano-particles (TNMO-NPs) to utilize as an electrode material in the SC. DFT calculations are applied to calculate the Gibbs free energy of activation for the ions access in the electrode, and the calculations exhibit that the lowest barrier energy of ions diffusion is 0.272 eV, suggesting that the Na^+ immigrate into the electrode conveniently. For the computational calculation of the transfer procedure of Na^+ in TNMO-NPs electrochemical system, the NEB method was applied to get the lowest energy path ways [128]. Three different diffusion path ways for Na^+ have been considered, as shown in Fig. 10a. The obtained barrier energy and coordinate system of diffusion are shown in Fig. 10b. The barrier energies of path1–3 are 0.272 eV, 0.371 eV, 0.873 eV, respectively. As the results shown that path1 have minimum Na^+ diffusion energy, suggesting ions could immigrate into the electrode.

Han et al. [129] applied a wet-chemical strategy to grow nano-structured $\text{MoS}_2\text{-2H}$ phase on activated carbon cloth ($\text{MoS}_2@\text{CC}$). The $\text{MoS}_2@\text{CC}$ was used as binder-free electrodes, and an AASCs was fabricated to assess its accumulation energy property. The $\text{MoS}_2@\text{CC}$ electrode shows a maximal capacity of 1010 F g^{-1} and a cycling retention of 98 % after 10,000 cycles. The diffusion energy barrier of ion (NH_4^+) on MoS_2 surface has been learned through the NEB method. As shown in Fig. 11a, the diffusion path ways between adjacent inferior energy octahedral sites are investigated. The NH_4^+ is applied an adjacent octahedral site in the original geometry. The diffusion path ways are resulted at Fig. 11b. The path 1 exhibits a smaller energy barrier (0.8 eV). Hence, it can be resulted that the small energy barrier of path 1 contributes to the energy storage performance due to its easy embedding/ leaving into/ from the MoS_2 electrode. Furthermore, the CDDs are tested at the interlayer of MoS_2 to research the ion adsorption that happens during the intercalation procedure. As shown at Fig. 11c, the CDD results are calculated by using the total electron density of NH_4^+ , while NH_4^+ is separated at the centric site of the MoS_2 . The calculations exhibits that the NH_4^+ acts as a bridge and transfer charges through the hydrogen bond of NH_4^+ to the adjacent S of the electrode material. The strong chemical bond between NH_4^+ and MoS_2 reduces the energy barrier of the entire system. The NH_4^+ commonly known as Lewis acid, while MoS_2 act as a Lewis base hence it can get a connection with NH_4^+ [130]. The charge of the Lewis adducts is mainly concentrated on the sulfur atom bonded with NH_4^+ . This charge accumulation can be in accordance with the CDD results. The buildup suggests the advantageous entrance performances for the ions into electrode layers, enhancing the

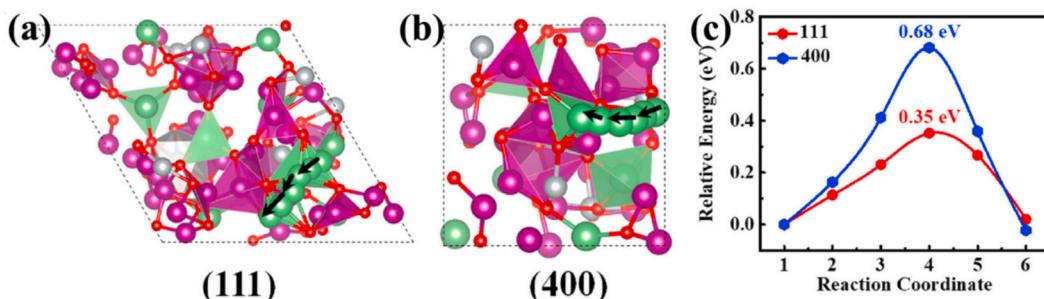


Fig. 9. (a-b) The top view of the Li^+ ions diffusion path ways on (111) and (400) facets of $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$, respectively. (c) The energy barriers on (111) and (400) facets, resulted by the NEB method [10]. Copyright 2020, Elsevier Ltd.

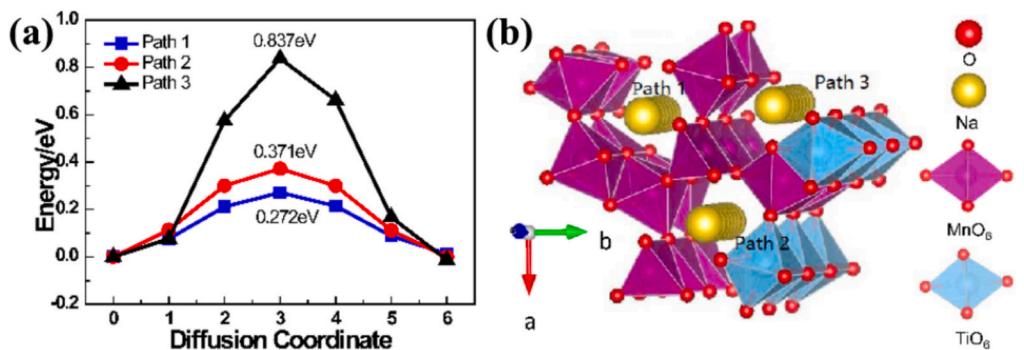


Fig. 10. (a) The simulated barrier energy and diffusion coordinate. (b) Probable diffusion path ways for Na^+ ions [127]. Copyright 2019 American Chemical Society.

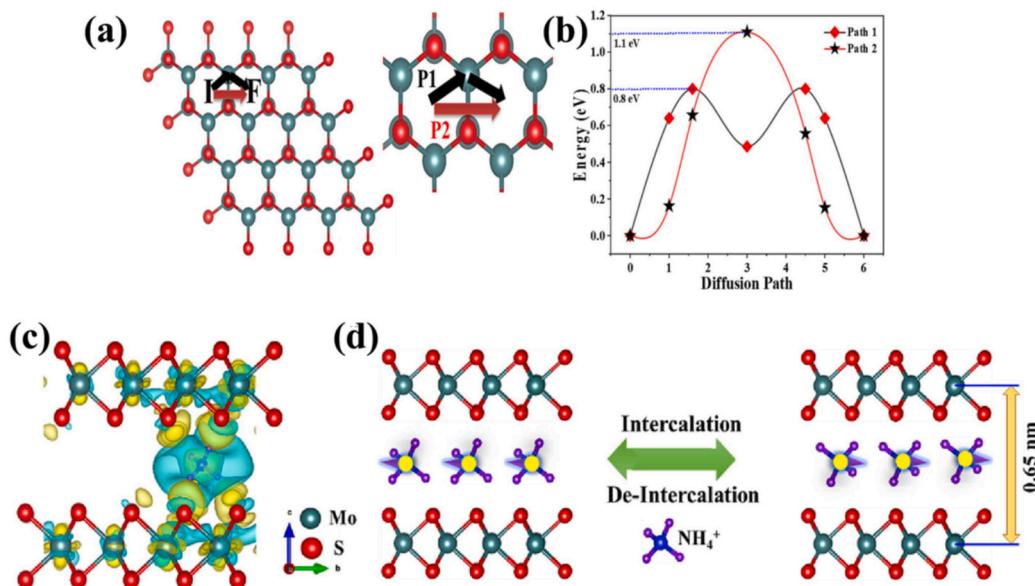


Fig. 11. (a) Transfer path way for NH_4^+ on MoS_2 . (b) Related diffusion energy barrier figure. (c) The CDDs of NH_4^+ accessed into MoS_2 structure. (d) The simulated charge storage mechanism [129]. Copyright 2022 Elsevier B. V.

electrochemical capacities of the electrode. According to these results, the prediction of the energy storage mechanism is shown in the Fig. 11d.

DFT calculations play a key role in determining the electronic structure of electrode materials in SCs. It can not only predict the electronic structure and band distribution of materials, but also analyze the active site, electron transport mechanism and material stability. Through in-depth understanding of the electronic structure of electrode materials, we can discover that the electronic properties of materials (such as band structure, state density and charge distribution, etc.) are closely related to the electrochemical properties of materials, including energy density, power density and key indicators such as cycle stability and capacitor life. These results are of great significance for designing and optimizing electrode materials and improving the performance of SCs. Therefore, the use of DFT calculation is not only a direct analysis of the properties of the material itself, but also a powerful tool for optimizing the electrochemical performance of SCs.

2.2. Application of DFT calculation in performance optimization of SCs

Firstly, the structure and composition of electrode materials are optimized by DFT calculation to improve the energy density and power density of SCs. It also explains how these optimizations affect charge storage and transfer processes to improve performance. Secondly, we can also use DFT calculation to analyze the structure change and

performance attenuation mechanism of electrode materials during charge and discharge, and guide the development of SCs with higher cycle stability and longer life.

2.2.1. Improvement of energy density and power density in SCs

Due to the faradaic pseudocapacity performances of anthraquinone molecules ($\text{C}=\text{O}$) with a good electrochemical performance, Liu et al. [131] proposed an electrode with congenerous anthraquinone molecules and $\text{Ti}_3\text{C}_2\text{T}_x$ active cores to increase the energy density of the electrode. As shown at Fig. 12, based on the DFT calculation, the charge density difference of $\text{Ti}_3\text{C}_2\text{T}_x$ was investigated to research the covalent crosslinking of organic molecules in the hybrid materials. With covalent crosslinking, the organic segment has been forecasted to conduct to a small conduction band adjacent the Fermi level, implying improvement of the electro-conductivity.

2.2.2. Improvement of cycling stability and life in SCs

In theory, the charge and discharge steps of SC should be completely reversible, that it makes them get a long recycle life. Nevertheless, in fact, because of the manifold cycles and different working environment, properties of SCs would always attenuate fleetly [132–134]. The investigation about aging mechanism should be appreciated for which could promote extending the recycle life of SCs. Some researchers use carbon-based materials as an example to investigate the mechanism of

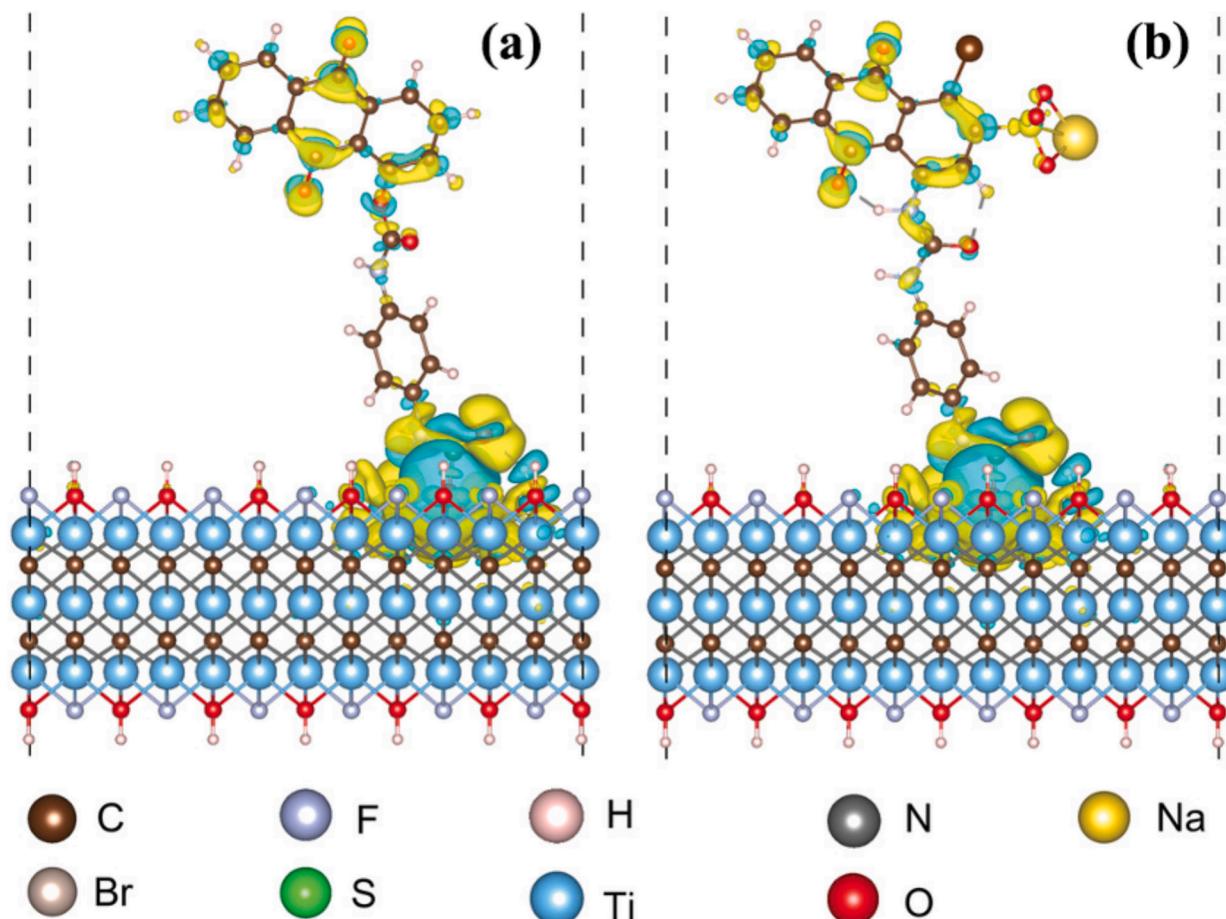


Fig. 12. Prediction of the contact between structure and electrical properties of $\text{Ti}_3\text{C}_2\text{T}_x\text{-HA}$ (a) and $\text{Ti}_3\text{C}_2\text{T}_x\text{-ABS}$ (b) by DFT calculation [131]. Copyright 2022, Wiley-VCH GmbH.

aging [135,136]. Nozu et al. reported that the closed cell of SCs could be broken by gas generation and gathering. The sources of these gases are mainly the decomposition of electrolytes and electrode by-products [137]. Moreover, Zhu et al. [138] researched electrochemical aging of commercial carbon in electrodes by various presentation methods. They suggested that the aging of carbon-based materials, specifically the blocking and collapse of pores, was the primary reason that resulted in the descending properties of carbon-based SCs. In the meantime, diverse parts of SCs are tested and a variety of instructive results along with numerous relevant characterization methods are published. The separator changed to another color and the current carrier became destroyed are found by them. Furthermore, theoretical DFT calculations has also been frequently used in the research of SC aging. Liang et al. [139] explored the aging procedure of carbon-based SCs by analyses of the primary parts in the SCs after aging process happened at disparate temperatures for a long recycle life. According to the theoretical DFT calculations, the measured size of before and after solvated BF_4^- ions were 2.86 Å and 11.11 Å, respectively. It can be seen that the aging phenomenon of SCs can be attributed to the decreased diffusion efficiency of ions and charges in holes that have collapsed or blocked. Moreover, the increase in the resistance of the entire capacitor system is also the main reason for the attenuation of capacitor performance.

To sum up, DFT calculations can predict the crystal structure of materials, which helps to design material structures with better theoretical properties. Furthermore, by calculating the band structure of the material, DFT can provide information about the conductivity, band gap width and band distribution of the material, which helps to understand and optimize the conductive properties of the electrode material. This is of great significance for improving the electrochemical properties of SCs

such as energy density and power density. Moreover, in the development process of SC materials, DFT calculation can conduct preliminary evaluation of the properties of materials before experiments, so as to screen out potential candidate materials, reduce unnecessary experimental attempts, and save time and resources. However, there are many shortcomings in DFT calculation. First of all, when DFT calculation is faced with a complex system, it usually requires a certain degree of simplification of the actual system in order to calculate. This simplification may lead to a certain deviation between the calculation results and the actual situation. Secondly, DFT calculation needs to consume a lot of computing resources and time, especially for complex systems and large-scale computing tasks. This limits the wide application of DFT in the field of SCs.

The emergence of ML can make up for the computational problems of DFT in the field of SCs. ML algorithms can be trained using high-quality data obtained from DFT calculations to build data-driven models. These models are able to quickly predict the properties of materials without having to rely on computationally intensive DFT simulations every time. For example, through the ML algorithm, electrode materials with potentially high capacitance can be quickly screened, greatly shortening the design cycle. ML can handle large-scale, multi-component material systems, and can even be combined with other computational methods (such as molecular dynamics) for multi-scale simulations, making up for the shortcomings of DFT in large systems and long-time scales. There is a huge amount of experimental and computational data in the field of SC materials, and ML can use this data to look for hidden relationships between material properties and structural parameters, and discover potential new materials.

3. Machine learning for SCs

Machine learning (ML) technology offers a transformative, data-driven approach to explore and analyze previously published research on various materials used in SCs [140]. By using advanced algorithms, researchers can identify critical features that contribute to the development of optimal materials for SCs, enhancing overall performance [141,142]. This innovative methodology not only streamlines the material discovery process but also opens avenues for various applications within the SC domain [143–148]. The subsequent sections will delve into specific methodologies and applications of ML in SC research, including the ML workflow for SC modeling, performance prediction, feature analysis and optimization, state of health (SOH) monitoring and failure control, as well as material design. Through these approaches, ML will significantly improve the efficiency and effectiveness of SC technologies.

3.1. ML workflow for SCs modeling

In the workflow for ML modeling of SCs, several key components are typically involved, including data collection, feature engineering, algorithm selection, and model evaluation [149]. Each of these elements plays a crucial role in developing effective predictive models. In the following sections, we would delve into a detailed discussion of these four components, exploring their significance and the common methods employed in each stage.

3.1.1. Data collection for SCs

Data collection is the foundational step in developing a ML model for SCs. Currently, the data sources for SC modeling are primarily categorized into three types: experimental, literature, and simulation [150–152]. Each type of data source plays a crucial role in providing comprehensive insights into the performance and behavior of SCs.

Experimental data is derived from laboratory experiments where SCs undergo various tests to measure key metrics. These metrics include capacitance (the ability to store charge), charge and discharge cycles (voltage and current profiles), equivalent series resistance (ESR, a measure of resistive losses), and cycle life (the number of charge/discharge cycles before performance degrades) [153,154]. Experimental data is essential because it provides real-world measurements that reflect the actual performance of SCs under specific conditions. The data is often collected using sophisticated laboratory equipment and is subject to rigorous validation to ensure accuracy and reliability.

In addition to experimental data, literature sources such as existing research papers, technical articles, and online databases provide valuable information on SC materials, designs, and performance metrics [155–158]. These sources help researchers to build upon previous work, identify trends, and understand the broader context of their findings. Natural Language Processing (NLP) techniques can be particularly useful in extracting and analyzing information from these literature sources, enabling researchers to efficiently summarize and synthesize large volumes of text data [159]. Literature reviews can also highlight gaps in current knowledge, guiding future research directions. By using the collective knowledge from published studies, researchers can enhance their understanding of SC behavior and improve the accuracy of their models.

Simulated data is also crucial in SC research, especially when experimental and literature data are limited. Experimental data, though accurate, often demands significant resources and time due to lengthy cycles and high material and labor costs. In contrast, simulated data can be generated swiftly and efficiently [160]. It allows us to explore various conditions that are impractical or too costly to test experimentally [161]. This not only provides detailed insights into the underlying mechanisms and potential performance enhancements of SCs but also fills the gaps left by scarce empirical data. Moreover, by offering a rich and diverse dataset, simulated data accelerates the ML process, enabling

more accurate predictions and optimizations for SC design and performance.

The integration of these diverse data sources is essential for developing robust ML models. Experimental data provides the ground truth, while literature sources offer historical context and theoretical foundations. Simulated data, on the other hand, allows for the exploration of hypothetical scenarios and the optimization of design parameters [162]. By combining these data sources, researchers can create comprehensive datasets that capture the full spectrum of SC behavior, leading to more accurate and reliable predictive models [163].

3.1.2. Feature engineering in SCs

Feature engineering plays a pivotal role in transforming raw data into meaningful representations that enhance the predictive accuracy of ML models for SC performance analysis [164,165]. This process involves systematic data preprocessing, transformation, and dimensionality reduction of datasets to capture critical relationships between material properties and energy storage capabilities.

Data preprocessing forms the foundation of feature engineering. Feature scaling techniques like standardization (zero mean, unit variance) or normalization (0–1 range) address discrepancies in feature units, such as current density and voltage, ensuring balanced model training. Common handling techniques for missing values (such as conductivity or capacitance) in SC datasets include imputation (mean, median, mode, etc.), interpolation for time-series data (e.g., charge-discharge cycles), and removal of excessively incomplete records [166,167]. Feature transformation employs nonlinear methods to SCs features. Common methods include logarithmic and square root transformations, which help stabilize variance and normalize distributions. For example, logarithmic transformations of current and voltage data can effectively address exponential growth trends often observed in charging and discharging processes, making the relationships between these variables more linear and easier for models to learn. Similarly, square root transformations can help mitigate the impact of outliers in performance metrics, such as energy density or power density.

Feature selection and dimensionality reduction simplify complex datasets to focus on performance-critical parameters. These techniques improve model performance, interpretability and computational efficiency, and help prevent overfitting. Statistical tools like Pearson correlation coefficients (PCC) quantify linear relationships between features (e.g., specific capacitance, electrolyte) [167,168]. Advanced interpretability frameworks, such as SHapley Additive exPlanations (SHAP), quantify feature contributions to predictions, guiding material optimization. In SC research, SHAP analysis reveals key factors affecting capacitance, guiding material developers in optimizing electrode design and performance [161,169]. Moreover, feature importance analysis evaluates input contributions to model predictions, identifying key performance drivers like electrode porosity and electrolyte composition. On this basis, partial correlation analysis further separates variable relationships by controlling complex factors, revealing direct dependencies between specific material properties and SC performance. Principal Component Analysis (PCA) transforms a large set of variables into a smaller number of principal components that capture the most variance in the data. By eliminating redundancy, PCA simplifies analysis and enhances visualization of high-dimensional datasets, making it particularly useful for SCs [170]. In SC research, Linear Discriminant Analysis (LDA) helps classify materials and performance metrics by maximizing separation between categories, enabling researchers to identify key features driving performance improvements. Additionally, t-Distributed Stochastic Neighbor Embedding (t-SNE) is a nonlinear dimensionality reduction technique ideal for visualizing high-dimensional data. By preserving relative distances between similar data points, t-SNE effectively clusters and reveals patterns, facilitating the identification of correlations among materials with similar performance characteristics [171].

Overall, feature engineering provides essential analytical tools for SC

research based on ML. By revealing hidden correlations between material properties (e.g., surface area, conductivity) and performance metrics, it empowers targeted design improvements to promote energy storage optimization.

3.1.3. ML algorithm selection for SCs

Choosing the right ML algorithms is crucial for effective SC modeling. For regression tasks, linear regression is suitable for modeling linear relationships, while support vector regression (SVR) excels at capturing nonlinear relationships, especially in smaller datasets [172]. For classification, support vector machines (SVM) are effective for high-dimensional data and identifying complex patterns in SC performance metrics. Decision trees (DT) offer a simple yet powerful approach by splitting data based on feature values and making decisions based on the majority class in each subset. k-Nearest neighbors (KNN) is a non-parametric method that classifies data points based on their nearest neighbors in the feature space. Ensemble methods combine multiple models to enhance performance and can be applied to both regression and classification tasks. Random Forest (RF) reduces overfitting and improves accuracy through multiple decision trees [173]. Gradient Boosting Machines (GBM) build models sequentially to minimize previous errors. XGBoost, an optimized implementation of gradient boosting, enhances performance and speed for large datasets [166,167]. AdaBoost combines weak learners to create a stronger model by focusing on errors made by previous models [174]. Bootstrap aggregating (bagging) improves stability and accuracy by training multiple models on data subsets and averaging predictions.

In addition, neural networks, particularly deep learning techniques, have shown advantages in analyzing large and complex datasets, identifying intricate patterns vital for SC research and development [174,175]. Artificial neural networks (ANNs) are universal models suitable for various predictive tasks, while feedforward neural networks (FNNs) are good for general predictive tasks such as forecasting SC performance metrics based on design parameters and operational conditions. Convolutional neural networks (CNNs) are designed for spatial data and can analyze the microstructural features of electrode materials and their impact on electrochemical performance. Recurrent neural networks (RNNs) are ideal for time-series data, such as monitoring the dynamic charging and discharging processes of SCs and analyzing temporal variations in voltage and current profiles [176].

By using these algorithms and neural network architectures, researchers can enhance the understanding and optimization of SC technologies, ultimately leading to improved performance and efficiency.

3.1.4. ML model evaluation of SCs

After training, models must be evaluated using appropriate metrics. For regression tasks, key metrics are Mean Squared Error (MSE) for prediction accuracy, Root Mean Squared Error (RMSE) for error magnitude interpretation, and R-squared (R^2) for variance explanation [172–174]. For classification tasks, key metrics typically include accuracy measuring the proportion of correct predictions, precision evaluating the ratio of true positives to all predicted positives, recall quantifying the model's ability to identify all actual positive instances, and F1-score providing a balanced measure of precision and recall [172]. Additionally, cross-validation techniques such as k-fold validation help ensure the model generalizes well to unseen data by systematically rotating training and validation subsets [177]. Hyperparameter tuning methods like grid search or random search optimize the hyperparameters of ML algorithms to enhance SC predictive performance [175]. This comprehensive evaluation approach enables researchers to develop robust models that can effectively predict SC performance characteristics such as energy density, power density and cycle life under various operating conditions, ultimately advancing energy storage technology development through optimized material selection and design configurations.

The structured ML workflow enables robust SC performance

modeling, predicting key parameters like energy density, power density and cycle life under various conditions. By optimizing designs and identifying superior materials, it accelerates the development of efficient, long-lasting energy storage systems.

3.2. Performance prediction and optimization

In recent years, the utilization of ML techniques in the field of SCs has attracted great attention, especially in predicting electrochemical performance metrics such as capacitance and longevity [178–180]. These advanced algorithms can efficiently analyze complex datasets generated during experiments and practical applications, enabling the rapid identification of optimal materials and designs. The development of accurate prediction models is essential to accelerate the development of high-performance SCs. By using ML, researchers can reveal the key relationship between various material properties and performance results, ultimately facilitating the design of more durable and efficient SCs for specific applications.

3.2.1. ML for Capacitance Prediction in SCs

Traditional methods for evaluating capacitance often involve time-consuming experimental procedures and complex electrochemical analyses [181]. However, the integration of ML algorithms offers a promising alternative, enabling researchers to analyze vast datasets and uncover hidden patterns that correlate material properties and fabrication parameters with capacitance performance [182–184]. Recent literature has reported various successful implementations of ML models, including regression techniques, neural networks and so on, to predict the specific capacitance of different SC materials [185–187]. These advancements not only enhance the efficiency of material selection but also pave the way for the design of next-generation energy storage devices.

Ghosh et al. [188] utilized a combination of ML models for value and grade prediction to assess the performance of a new material, cerium nitride oxide, in SC applications. Their model forecasted that the electrical capacity of this material was approximately 26.6 mAh g^{-1} , with a capacity retention rate exceeding 90 %. The accuracy of these predictions was verified by experimental results. Notably, this study represented the first instance of predicting and reporting the use of cerium nitride oxide as a SC electrode, which has important practical significance.

Abdi et al. [189] explored the effectiveness of modeling the capacitance of EDLC using four ML algorithms. The effects of 13 key parameters such as specific surface area, pore volume and doping elements on capacitance were analyzed by collecting the physical and chemical properties of 681 different carbon electrodes. The experimental results show that the Super Learner (SL) model performs best in terms of capacitance prediction accuracy and robustness compared to other ML models, such as Extremely Randomized Trees (Extra trees), Extreme learning machine (ELM), and Multivariate adaptive regression splines (MARS).

Tawfik et al. [154] collected data from nearly 100 published studies to focus on SCs with specific electrode types, such as mesoporous, nanoporous, microporous, and hierarchical porous carbon electrodes. These data were analyzed using ML models (Lasso regression, SVR, and ANN with different architectures) to predict the specific capacitance ($F \text{ g}^{-1}$) of the SCs (Fig. 13). Meanwhile, these models considered structural properties of the electrode material as well as various physicochemical characteristics, including the type of electrolyte, pore volume, and specific surface area. The performance of these models was evaluated based on the RMSE and the correlation between predicted and actual capacitance values. The ANN model developed in this study achieved RMSE, MAE, and R values of 30.82, 46.5624, and 0.89537, respectively, indicating highly accurate predictions compared to other models.

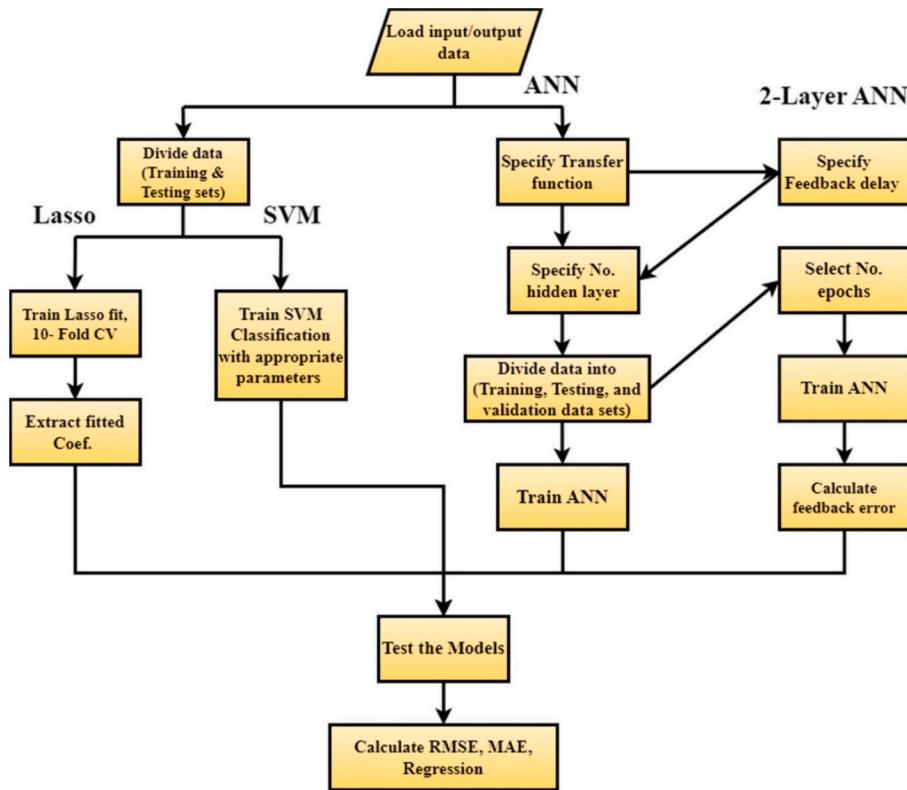


Fig. 13. ML modeling flowchart for capacitance prediction [154]. Copyright 2023, Elsevier Ltd.

3.2.2. ML for longevity prediction of SCs

The longevity prediction of SCs is a critical aspect of their development and application in energy storage systems [190]. Key factors influencing longevity include cycle life, which indicates the number of charge-discharge cycles a SC can endure before significant performance degradation; cycle stability, which assesses the retention of capacitance over repeated cycles; and RUL, which estimates the time left before the device fails or experiences substantial performance loss [191,192]. Traditional methods for evaluating these parameters can be labor-intensive and time-consuming. Recent advancements in ML have shown great promise in enhancing the accuracy of these predictions by analyzing large datasets derived from experimental results and operational conditions. These studies have employed various ML techniques, such as RF and deep learning models, to effectively predict cycle life and stability, as well as to estimate RUL based on real-time monitoring data [193–195]. These developments not only improve the understanding of SC longevity but also facilitate the design of more durable and efficient energy storage systems [196].

Ren et al. [197] explored three ML models, namely Logistic Regression (LR), PCA, and ANN, to predict the cycle life of SCs at an early age. A powerful data set was created containing 88 different SCs under various charging methods with a wide cycle life of up to 10,000 cycles. The ANN model demonstrated superior performance, achieving test errors below 10.9 % by utilizing only the initial 16 % of cycles, in contrast to 29.5 % and 18.3 % test errors from the LR and PCA models, respectively. The accuracy of the ANN can be further enhanced by increasing dataset size, reflecting its capability to handle complex systems effectively. The findings highlighted the potential of ANN for accurate predictions, while also suggesting that further advancements in ML techniques and feature selection could lead to even better performance in the future.

Nanda et al. [198] constructed a database containing 1000 data by analyzing more than 400 literatures, and applied five hybrid ML algorithms, including RF, Random Subspace (RS), Random Committee (RC),

Multiclass Classifier (MCC) and Random Tree (RT), to assist the cyclic stability prediction of SCs including grade prediction and value prediction. The analysis indicated that the RF and RS algorithms effectively predicted capacity retention while revealing key insights about cyclic stability: it improved after the complete activation of electrodes, but electrolytes exceeding 2.5 V often result in weaker stability despite higher initial capacitance. Furthermore, three-dimensional structures were shown to enhance stability during charge-discharge cycles.

Guo et al. [199] proposed a robust ML prediction model for estimating the RUL of SCs, utilizing a multi-stage (MS) approach that combines empirical mode decomposition (EMD) with a gated recurrent unit (GRU) neural network. The model leveraged time series features from the voltage curve and the residual components of the discharge capacitance curve, which show strong correlation with discharge capacity. Training was conducted using data from the first 100 cycles of discharge, and the cycling process was divided into fast decay and slow degradation stages, allowing for a modified prediction process that enhances accuracy. The proposed EMD-MS-GRU model effectively addressed limitations of traditional models, such as sensitivity to feature quality and challenges in handling long-term dependencies. In validation experiments, the model demonstrated high stability and prediction accuracy, achieving RMSE less than 0.1 % and R^2 greater than 99 %. Overall, this method highlighted the potential of artificial intelligence algorithms in energy storage systems and offered a viable solution for predicting SCs RUL.

Li et al. [200] developed a SC aging state prediction model based on heuristic Kalman filtering (HKF) algorithm optimized extreme learning machine (ELM), which effectively improved the prediction accuracy of the ELM model by optimizing the input weights and biases of the ELM model through the HKF algorithm, especially showing significant advantages in handling non-convex optimization problems (Fig. 14). Compared with traditional ELM models, PSO-ELM models, traditional Kalman models and KA-ELM models, the HKF-ELM model exhibited lower errors and faster calculation speed in predicting the capacitance

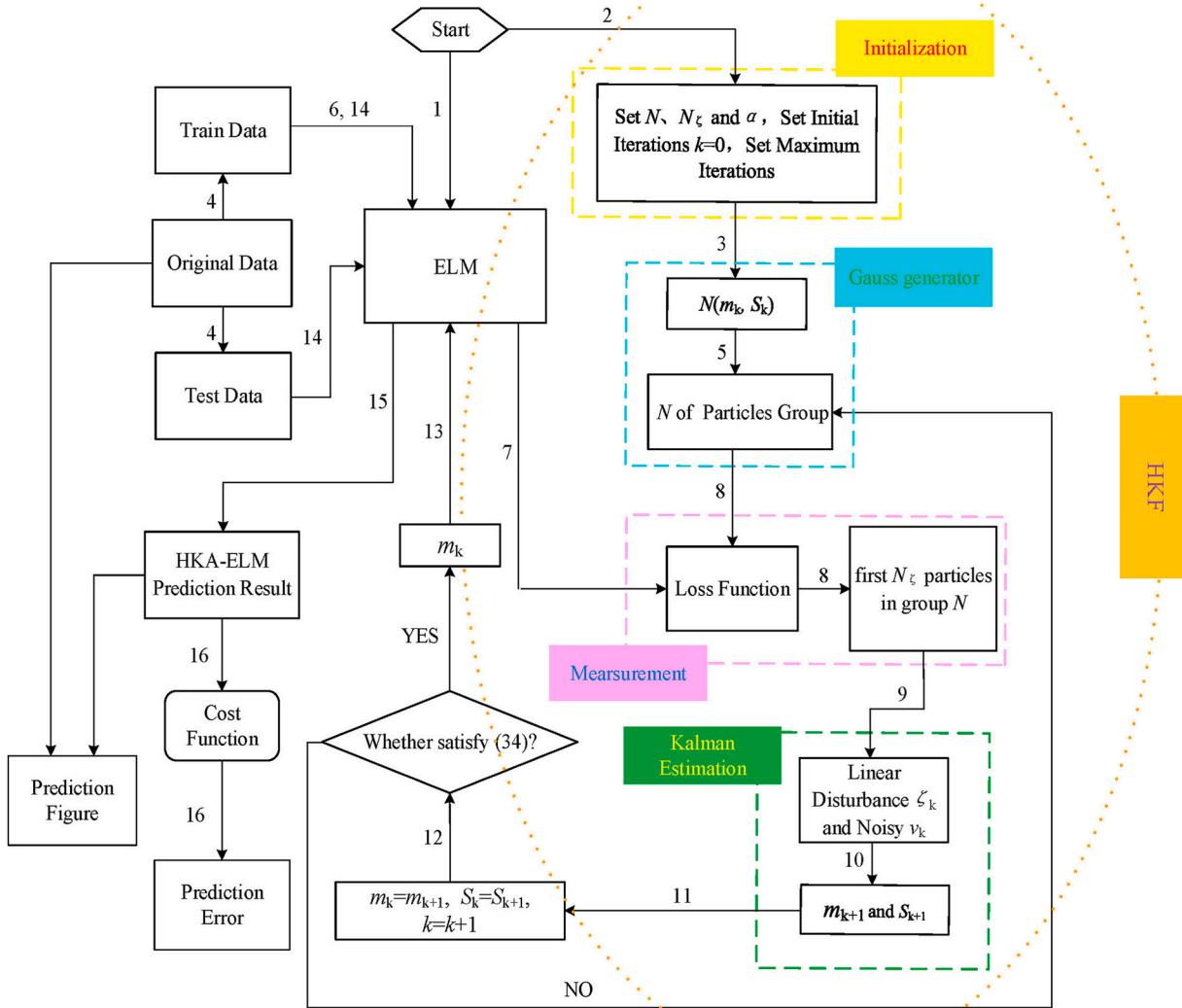


Fig. 14. Flow chart of the HKF-ELM model [200]. Copyright 2022, Elsevier Ltd.

aging state of SCs, and has good generalization ability. Experimental results showed that the HKF-ELM model can accurately predict the aging state of SCs under different temperature, voltage and current environments, which is expected to be applied to real-time aging monitoring and RUL prediction of SCs, providing technical support for ensuring the reliability of SC energy storage systems.

Performance prediction plays a crucial role in understanding how different factors influence the behavior of SCs. However, to refine these predictive models and enhance their accuracy, it is essential to delve into feature analysis and optimization. By identifying the key features that significantly impact performance, researchers can focus on the most relevant material properties and design parameters. This targeted approach not only improves the predictive models but also guides the optimization process, allowing for the development of materials and designs that maximize performance while minimizing cost and complexity. Thus, feature analysis and optimization are integral to advancing the application of ML technology in SCs through more informed and efficient design choices.

Building on this foundation, the application of ML technology in SCs extends to SOH monitoring and failure control. By employing predictive models, researchers can assess the current health of SCs in real-time, predict potential failures, and implement timely maintenance strategies, ultimately enhancing the reliability and longevity of energy storage systems.

3.2.3. Monitoring SCs with ML

Monitoring the SOH in SCs is essential for ensuring their operational efficiency and safety [201]. SOH assessment involves evaluating various parameters that indicate the internal condition of the SC, such as capacitance, internal resistance, and temperature. Traditional SOH monitoring methods often rely on manual inspections and basic performance metrics, which may fail to capture subtle changes that precede failure. In recent years, the integration of ML techniques has significantly advanced SOH monitoring and failure control strategies. By analyzing extensive datasets collected from real-time operations, ML models can identify anomalies and predict potential failures with greater accuracy. Furthermore, these models can facilitate failure control by providing timely alerts and recommendations for maintenance actions, thereby preventing catastrophic failures [202]. Recent literature has demonstrated the effectiveness of various ML techniques, including neural networks and ensemble methods, in developing predictive models that not only assess SOH but also provide actionable insights for preventive maintenance [203]. This integration of ML into SOH monitoring and failure control significantly improves the reliability and longevity of SCs in critical applications.

Li et al. [200] developed a novel HKF-ELM model, which can predict SOH of SCs in real time according to the previous charge and discharge data. Haris et al. [190] proposed a novel multi-trend learning method for predicting the SOH of SCs using non-dominated search genetic algorithm III (NSGAIII) and non-linear autoregressive network with

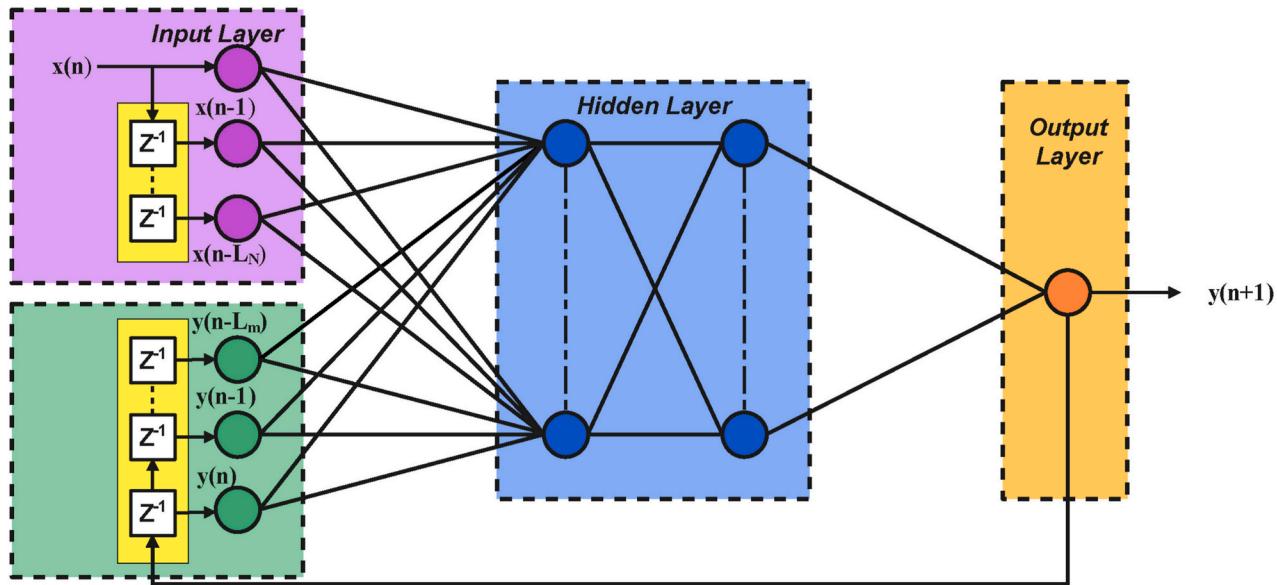


Fig. 15. Schematic diagram of the NARX neural network [200]. Copyright 2022, Elsevier Ltd.

exogenous inputs (NARX) recurrent neural network (Fig. 15). The method utilized the degradation trend data of multiple SCs under different charge-discharge currents and discharge voltage depths to train the NARX neural network model, thereby improving the model's generalization ability, reducing prediction errors, and achieving good performance on different SC datasets. Compared with traditional single-trend learning methods, this method significantly improved prediction accuracy, with an average root mean square error reduction of 49.14 %. Experimental validation confirmed the effectiveness of this approach in

enhancing the accuracy and robustness of SC SOH prediction.

Arevalo et al. [204] presented a ML-based model designed to identify photovoltaic (PV) failures using big data and simple equations while ensuring computational efficiency and precision. As shown in Fig. 16, a novel predictive power smoothing method (P-C) was introduced to alleviate power fluctuations associated with detected PV string failures by optimizing traditional algorithms such as Ramp Rate (RR) and Moving Average (MA). Laboratory experiments demonstrated that the PV monitoring system can accurately align with real-time data,

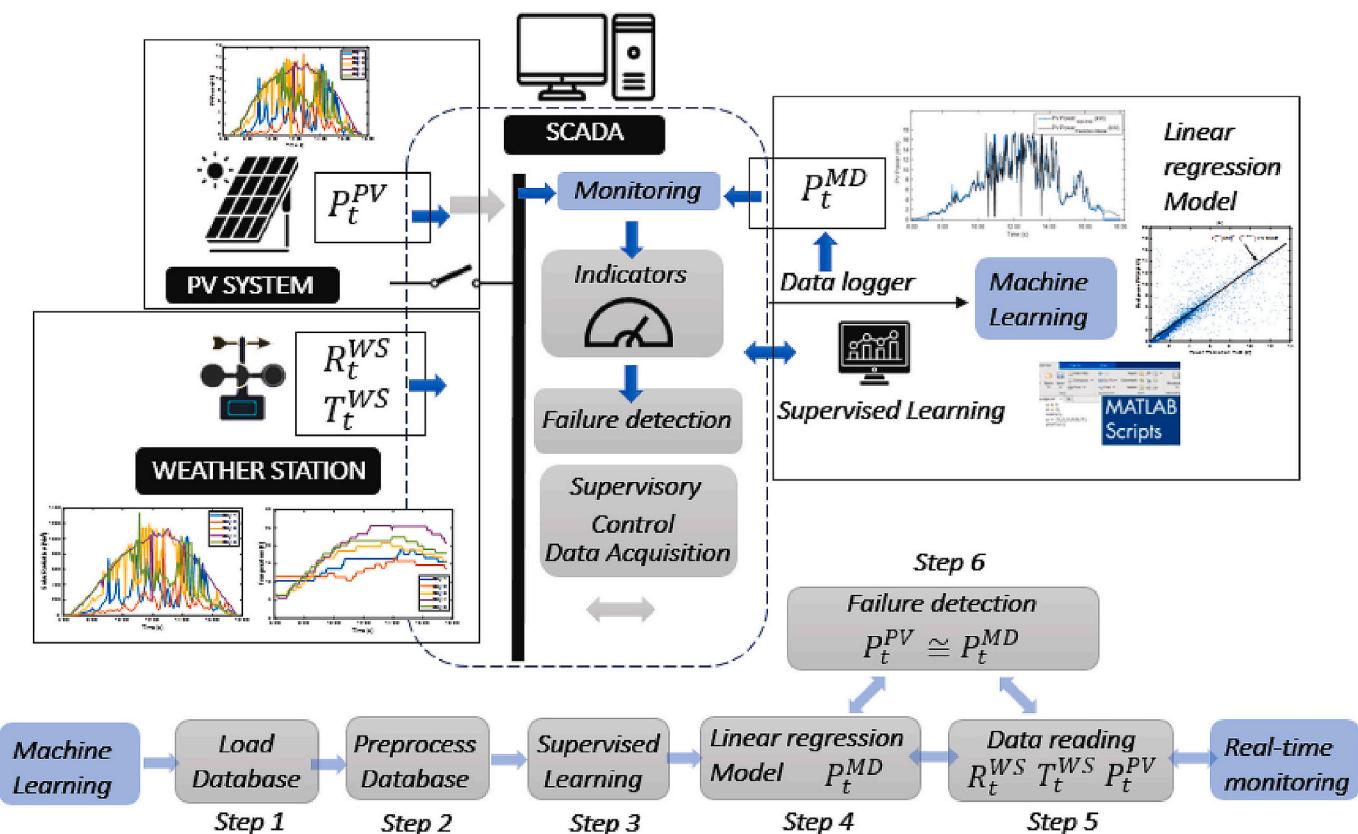


Fig. 16. Schematic diagram of ML process for PV failure detection [204]. Copyright 2023, Elsevier Ltd.

achieving an RMSE of 73.71 and an R^2 value of 0.95 for clear days. Compared to conventional methods like Moving Average, Low Pass Filter, and Ramp Rate, the P–C method yielded up to 50 % lower SC operability, due to its effective forecasting capabilities. Furthermore, the method showed resilience during fluctuations exceeding 20 % per minute, with the monitoring system recording RMSE values ranging from 0.66 to 2.47 before and after failures. Notably, the P–C method reduced computational efforts, with an initial execution time four times lower than that of the MA method, despite maintaining the same algorithmic complexity. The promising results suggested avenues for applying and validating this methodology in larger real-world scenarios to enhance its industrial applications.

These insights from SOH monitoring not only help in maintaining existing systems but also play an important part in guiding material design strategies. By using data from performance assessments and failure predictions, researchers can innovate and tailor new materials for SCs, ensuring they meet evolving performance standards and application requirements. This holistic approach connects data-driven analysis with practical advancements in material science, driving forward the next generation of SC technology.

3.2.4. ML-based feature analysis and optimization

In the research and development of SCs, understanding and optimizing various influencing factors is crucial for enhancing their performance [205,206]. With advancements in materials science and engineering technologies, researchers are employing a range of statistical and data analysis methods to unravel complex relationships. Among these techniques, PCC, PCA, feature importance analysis, and partial correlation analysis have emerged as essential tools in exploring the characteristics of SCs [207–209]. These methods not only help researchers identify and quantify the relationships between input variables and capacitance characteristics but also provide valuable insights for optimizing designs and improving performance [210]. In the following discussion, we will delve into the specific applications of these four analytical techniques in SC research and their interrelationships.

Nanda et al. [198] employed PCA and attribute prioritization to enhance value and grade prediction models for SC performance. By focusing on key features, the researchers significantly reduced computation time and improved dataset interpretability, achieving lower error metrics. Notably, Fig. 17 illustrated a decline in kappa statistics across all models, indicating that while PCA may not enhance performance due

to non-linear correlations among attributes, it still aided in improving interpretability and reducing computation time.

Sun et al. [170] explored the application of ML to clarify complex relationships in biochar preparation, and analyzed the influence of multiple factors (such as raw material characteristics, activation processes, and pore properties) on the energy storage properties of active biochar. The key parameters affecting energy storage performance were identified through PCC, feature importance and partial correlation analysis, which provided a scientific basis for future process optimization. The PCC value was calculated to assess the relationship between input variables and the output variable (specific capacitance), as well as to evaluate the collinearity among input variables. PCC helped identify which variables may have a significant impact on specific capacitance, providing preliminary statistical evidence for subsequent feature importance analysis. As shown in Fig. 18a-b, feature importance analysis evaluated the relative contribution of each input parameter to the specific capacitance of activated biochar using SHAP values. The results indicated that the activation process is the most important parameter for predicting capacitance, with the heating rate (HR) having a significant impact. A high HR value negatively affects the specific capacitance, while a low HR value has a positive effect. The characteristics of the raw materials, such as hydrogen and ash content, also play a crucial role in specific capacitance, particularly the ash content significantly influences the energy storage characteristics of biochar. Partial correlation analysis illustrated the relationships between input variables and the output variable through one-dimensional (Fig. 18c) and two-dimensional (Fig. 18d) partial dependence plots. The results showed that the pore characteristics of activated biochar (such as specific surface area (SSA) and micropore volume) are positively correlated with specific capacitance, indicating that a good pore structure can enhance electron transfer efficiency. The amount of urea added also significantly affects specific capacitance, as increased nitrogen doping contributes to improved energy storage characteristics.

In SC research, these methods can be used together to enhance analysis. For instance, PCC may be used initially to identify correlations, followed by PCA to reduce dimensionality. Feature Importance Analysis and SHAP can then provide insights into which features are most critical for capacitance, all of which can be visualized using t-SNE. By understanding the synergies among these techniques, researchers can more effectively analyze and interpret the complexities of material characteristics and their effects on sSC performance.

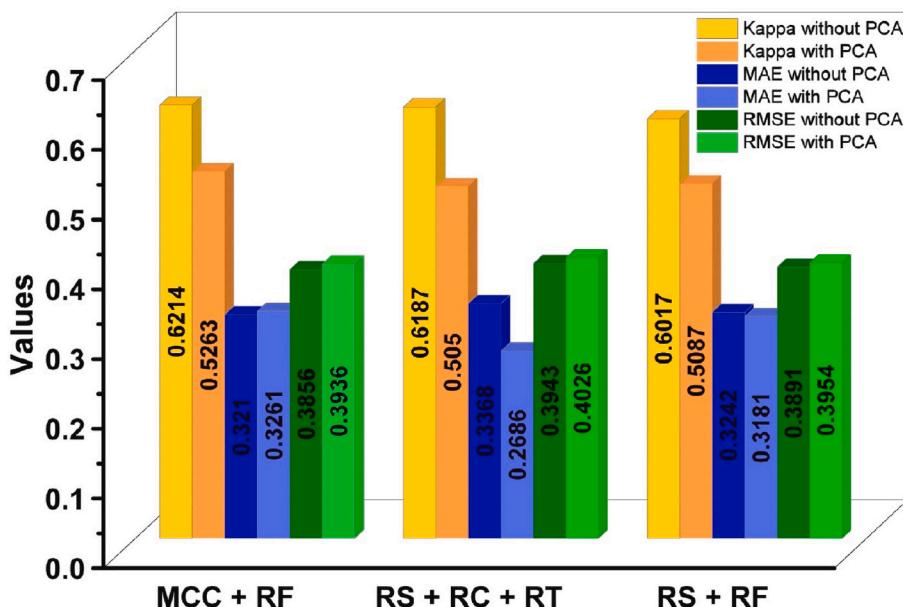


Fig. 17. The influence of PCA on the performance of grade prediction models [198]. Copyright 2022, Elsevier Ltd.

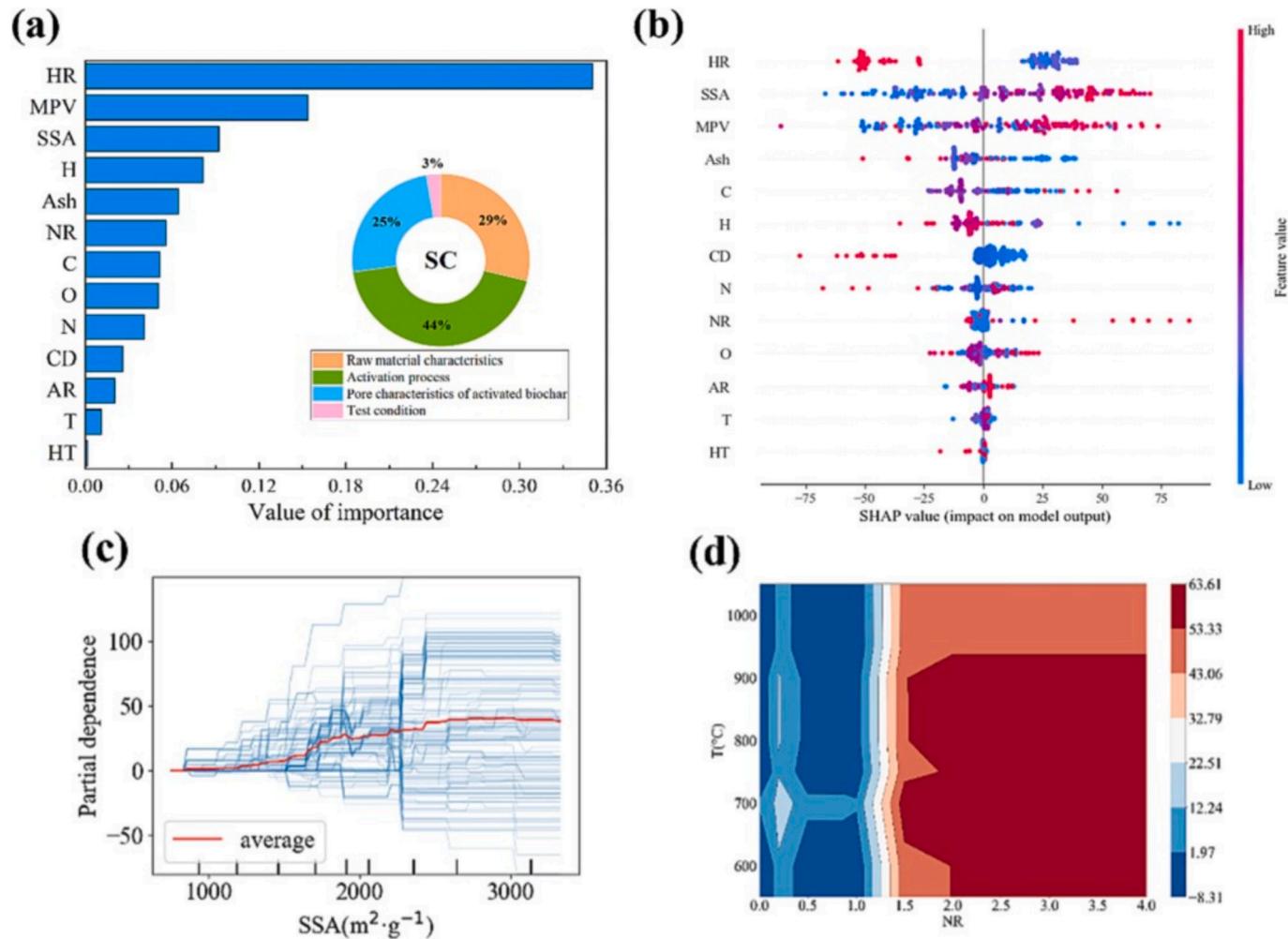


Fig. 18. Feature analysis [170]. (a) value of importance. (b) SHAP value. (c) One-dimensional partial dependence plot of SSA. (d) Two-dimensional partial dependence plot of urea-to-biochar (NR) and activation temperature (T). Copyright 2024, Elsevier Ltd.

3.2.5. Material design of SCs

The exploration of advanced materials in SC technology is the key to improve energy storage performance and efficiency [211,212]. Traditional material design methods often rely on empirical approaches, which can be labor-intensive and inefficient [213,214]. In recent years, ML has emerged as a transformative tool in the field of material design, enabling researchers to predict and optimize the properties of materials used in SCs [215,216]. ML algorithms can analyze vast datasets of material properties, structural characteristics, synthesis methods, and electrochemical performance to identify promising candidates for SC applications [180,217–219]. Recent studies have showcased the successful application of ML techniques in discovering new electrode materials, optimizing carbon structures, and enhancing the performance of hybrid SCs [220,221]. These advancements not only simplify the material discovery process, but also facilitate the design of tailored materials that meet specific performance criteria, ultimately leading to the development of next-generation SCs with improved energy density, power output, and cycle life.

Wang et al. [171] proposed a data-driven design framework for developing high-performance flexible SCs, using ML tools to identify and optimize key influencing factors for electrode materials and electrolyte selection, which can effectively shorten the design cycle for flexible energy storage devices (Fig. 19). The analysis results showed that nitrogen doping and specific surface area are crucial factors influencing the energy storage performance of electrodes. The use of ionic liquids as electrolytes ensures the safety and stability of the batteries under

extreme conditions. Additionally, the constructed ultra-thin flexible SCs maintain excellent energy storage capabilities even after rigorous electrochemical testing.

Wang et al. [222] developed a ML-assisted material discovery approach for synthesizing high-energy density aqueous SCs, utilized the ANN model to predict the upper limit of specific capacitance for oxygen-rich hyperporous carbon materials in acidic electrolytes, and a series of hyperporous carbon materials were designed and synthesized. The experimental results showed that the hyperporous carbon materials exhibited a specific capacitance of up to 610 F g^{-1} in $1 \text{ M H}_2\text{SO}_4$ solution, close to the theoretical prediction (Fig. 20a, b). Fig. 20c showed the experimental route of the target hyperporous carbons designed by the ANN model. The charge storage mechanism was revealed through step potential electrochemical spectroscopy (SPECS) and quasielastic neutron scattering (QENS) experiments, especially the heteroatom-doped mesoporous contributed the most to the total capacitance. Further, a feedback mechanism was used to incorporate experimental data into the ML model, enhancing its predictive power.

Shariq et al. [223] developed four ML models—multiple linear regression (MLR), SVR, RF, and ANN—to optimize SC electrodes made from MXene and varying graphene nanoplatelet (GNP) compositions (10–40 wt%). The objective is to identify the most accurate model for predicting optimal GNP loading to maximize electrochemical performance (Fig. 21). Characterization techniques like XRD and FE-SEM confirm improvements in conductivity and interlayer spacing with GNPs. The datasets generated from electrochemical tests train these

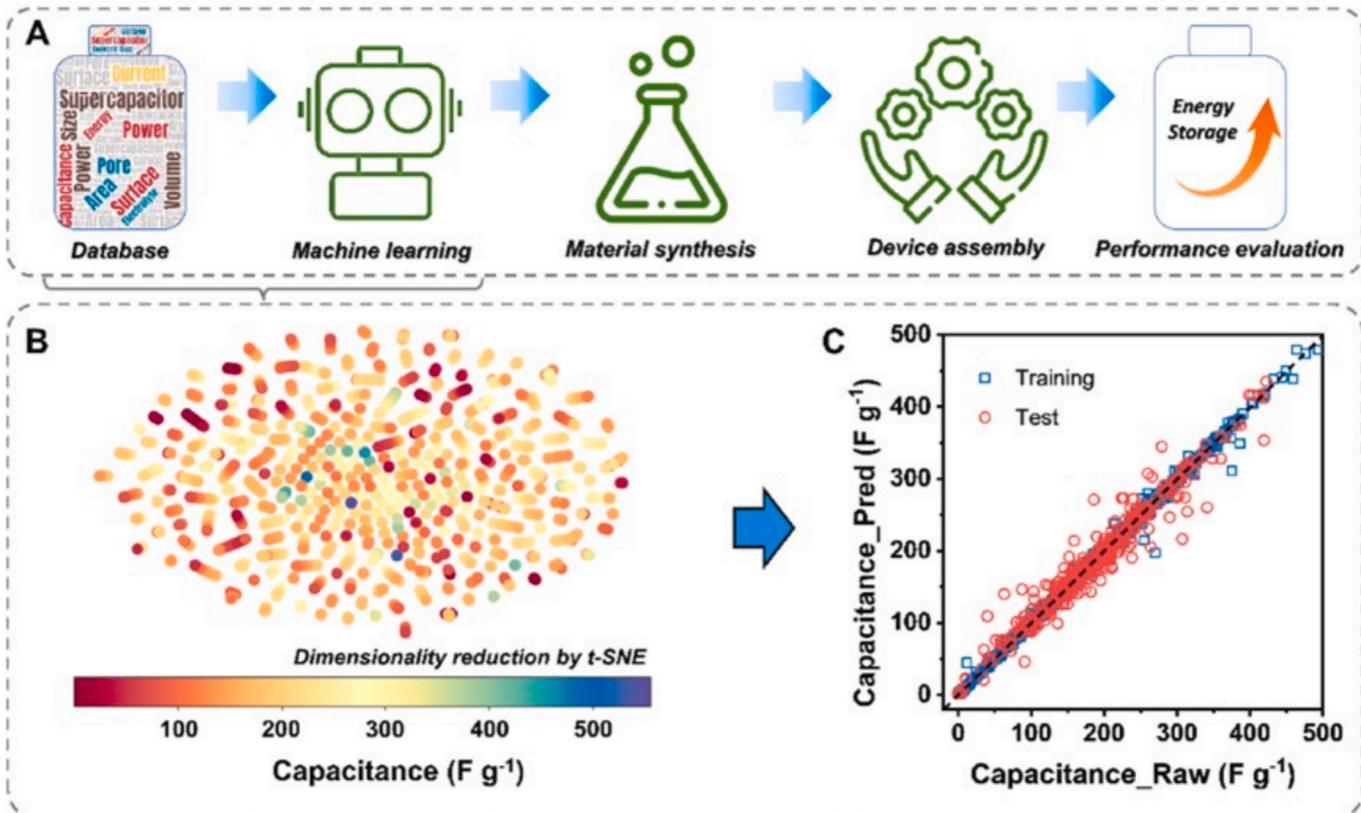


Fig. 19. Data-driven design of carbon-based materials [171]. (a) Workflow diagram of designing high-performance SCs. (b) The t-SNE algorithm for data dimension reduction. (c) The optimized selection algorithm of TPOT assisted ML. Copyright 2023, Elsevier Ltd.

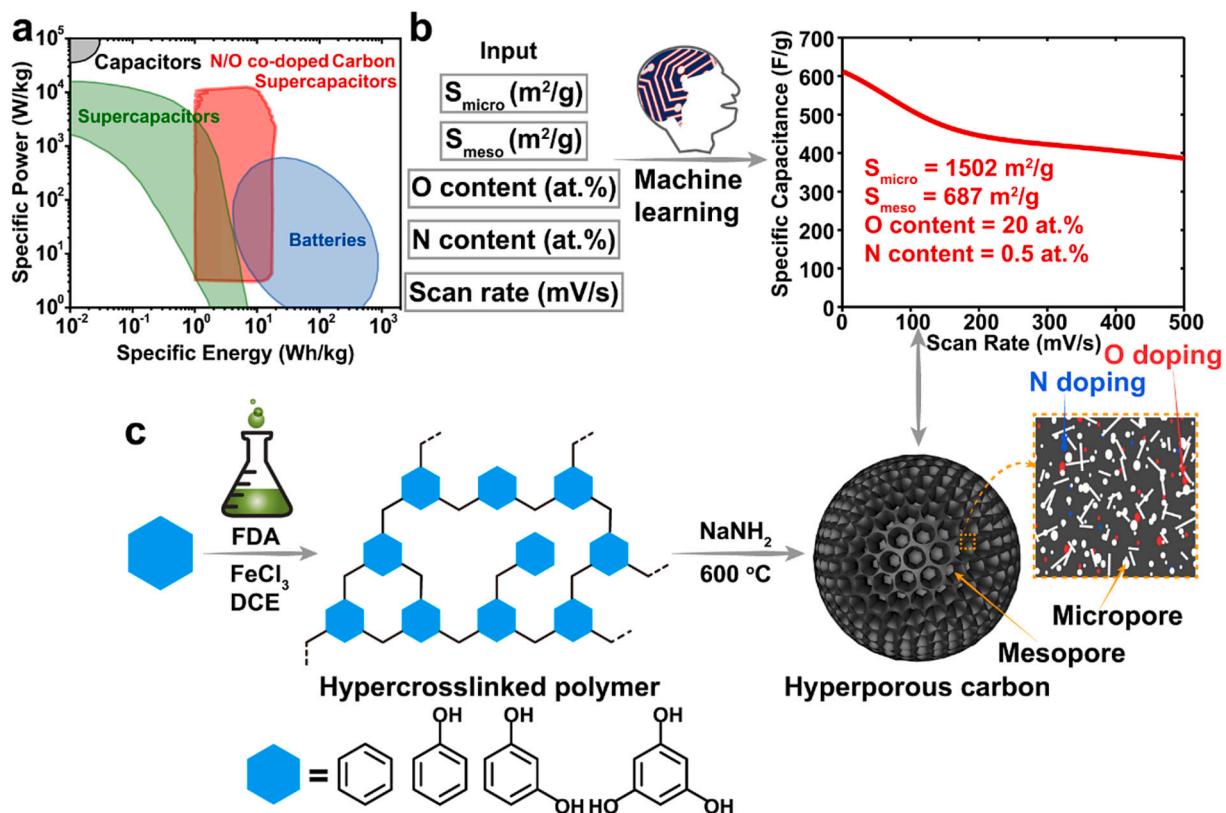


Fig. 20. ML-assisted material discovery of hyperporous carbons for SCs [222]. Copyright 2023, Springer Nature.

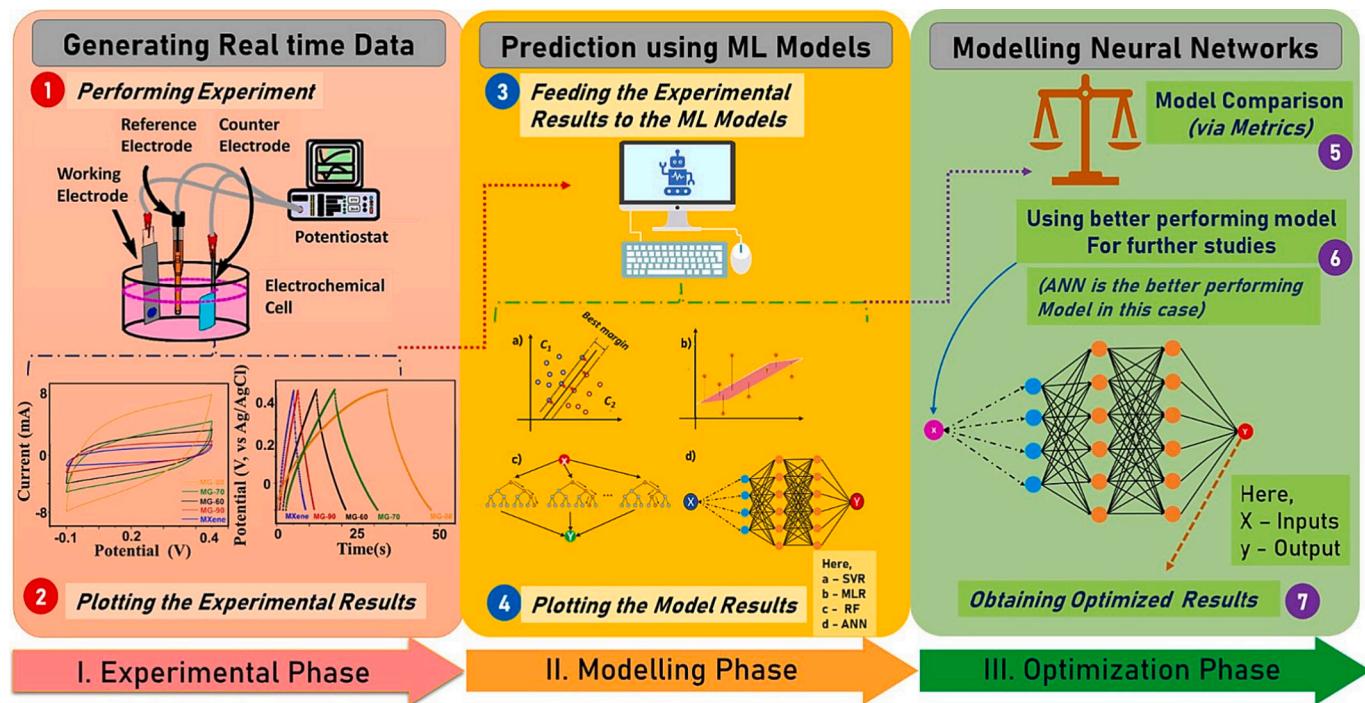


Fig. 21. ML Design of SC Electrodes with Real-Time Experimental Data [223]. Copyright 2024, Elsevier Ltd.

models, with the developed ANN model achieving a specific capacitance of 226.6 F g^{-1} for the optimal composition (20 wt% GNPs, MG-80) and accurately predicting its performance over 10,000 cycles. Moreover, the ANN considered real-time correlations between input features that influence SC electrode behavior, demonstrating its efficacy in designing high-performance electrodes.

DFT allows researchers to accurately predict the electronic properties and behavior of materials at the atomic level, while ML offers the ability to analyze large datasets and identify patterns that may not be immediately apparent. The integration of DFT and ML represents a powerful approach in the study of SCs, combining the strengths of quantum mechanical calculations with advanced data-driven techniques. This synergy not only enhances the understanding of material characteristics but also accelerates the discovery of new candidates for SC applications. Therefore, it paves the way for multi-method collaborative research on SCs, where various computational and experimental techniques are employed simultaneously. By integrating insights from DFT, ML, and other methodologies, researchers can fully investigate SC performance, optimize material selection, and develop enhanced energy storage solutions with greater efficiency.

Moreover, ML is not only confined to academic research but is also being actively integrated into industrial applications. For instance, Skeleton Technologies [224] is collaborating with Tallinn University of Technology to integrate ML and automation into energy storage production. They focus on applying Industry 4.0 techniques to optimize industrial processes for products like SuperBattery. This collaboration aims to enhance energy storage efficiency and contribute to European research. Additionally, their work on peak shaving for AI data centers could utilize ML to optimize energy management during high-demand periods. Such initiatives highlight the potential of ML in transforming the energy storage landscape through both material innovation and process optimization.

4. Multi-method collaborative research on SCs

The exploration of SCs has increasingly benefited from multi-method collaborative research, which combines various computational and

experimental techniques to enhance material discovery and optimization [225,226]. By integrating approaches such as DFT, MD, Monte Carlo (MC) simulations, and ML, researchers can gain comprehensive insights into the complex behaviors of electrode materials [227–229]. This collaborative framework not only facilitates a deeper understanding of the fundamental mechanisms governing energy storage but also accelerates the development and screening of advanced SCs with improved performance characteristics [230,231]. As the demand for efficient energy storage solutions continues to grow, multi-method research stands as a pivotal strategy in advancing the field of SC technology.

4.1. Unique contribution of integrated algorithms

The synergistic integration of DFT calculations and ML approaches leads to significant improvements in electrode performance metrics. Seo et al. [232] developed a binder-free electrode material consisting of lead vanadate (PbV_2O_6) with a unique hybrid structure grown on nickel foam. They proposed a capacitance-power prediction model based on ML and recurrent neural networks, ultimately achieving remarkable performance with a power density of 6000 W kg^{-1} and an energy density (26 Wh kg^{-1}). In comparison, similar electrode materials like NiV_2O_6 only reached 375 W kg^{-1} power density at comparable energy densities, making the PbV_2O_6 material approximately 16 times superior in power density.

While DFT provides accurate calculations, its high computational cost makes rapid screening of large material libraries impractical. The combination with ML significantly accelerates material screening and optimization processes. For instance, Wang et al. [233] employed ML models to predict band gaps of over 5000 perovskite materials, efficiently identifying six optimal candidates - demonstrating computational efficiency improvements of several orders of magnitude compared to conventional DFT calculations. Similarly, Li et al. [234] combined supervised and unsupervised learning approaches to screen 1092 two-dimensional semiconductor/metal heterostructures, successfully identifying six low-resistance materials while addressing data scarcity challenges.

This integrated approach utilizes ML for rapid preliminary screening followed by DFT validation of key candidates, dramatically reducing computational resource requirements. Moreover, the DFT-ML synergy enables more effective elucidation of structure-property relationships. Victor Fung et al. [235] developed a methodology that extracts structure-property relationships from DFT-calculated DOS data, achieving computational efficiency improvements of 10^6 compared to standard DFT approaches.

In industrial applications of novel electrode materials, DFT-assisted ML computation demonstrates substantial practical value. Wu et al. [236] developed Fe-ZIF/G micro-supercapacitors integrated into self-powered gas sensing systems, fabricated using spray-printing techniques. Furthermore, Xu et al. [237] combined DFT with deep learning to enable efficient prediction of material perturbation properties, a methodology with promising potential for extension to SC material design.

4.2. Data transfer mechanisms

In SC research, integrating various computational methods, such as DFT, ML, MD and MC, is crucial for comprehensive material analysis and

optimization. In practice, combining several of these methods (two, three, or more) requires efficient data transfer mechanisms to ensure seamless collaboration [232–239]. Generally speaking, DFT provides detailed insights into the atomic and electronic properties of materials, forming the basis for further simulations. MD captures dynamic behaviors like ion diffusion, while MC explores configurational and thermodynamic properties. By analyzing large datasets generated from DFT, MD, and MC simulations, ML models can identify unknown patterns and correlations, accelerate material screening, and optimize materials design by predicting performance metrics.

Several recent studies have demonstrated the effectiveness of multi-method collaborative research in SCs. For example, Fontaine et al. [23] combined MD and DFT simulations to explore the role of Thionine (Th) as a redox mediator in high-concentration LiTFSI SC electrolytes. MD simulations dynamically captured the diffusion behavior of Th, revealing its selective access to mesopores (3 nm) due to size exclusion in micropores (0.9 nm) and suggesting hydrogen bonding interactions with mesopore walls. DFT further explained the microscopic interactions behind these phenomena, indicating an ion association reaction between TFSI and Th due to their opposite charges, with a significant charge transfer and a dissociation energy of nearly 1 eV. This synergistic

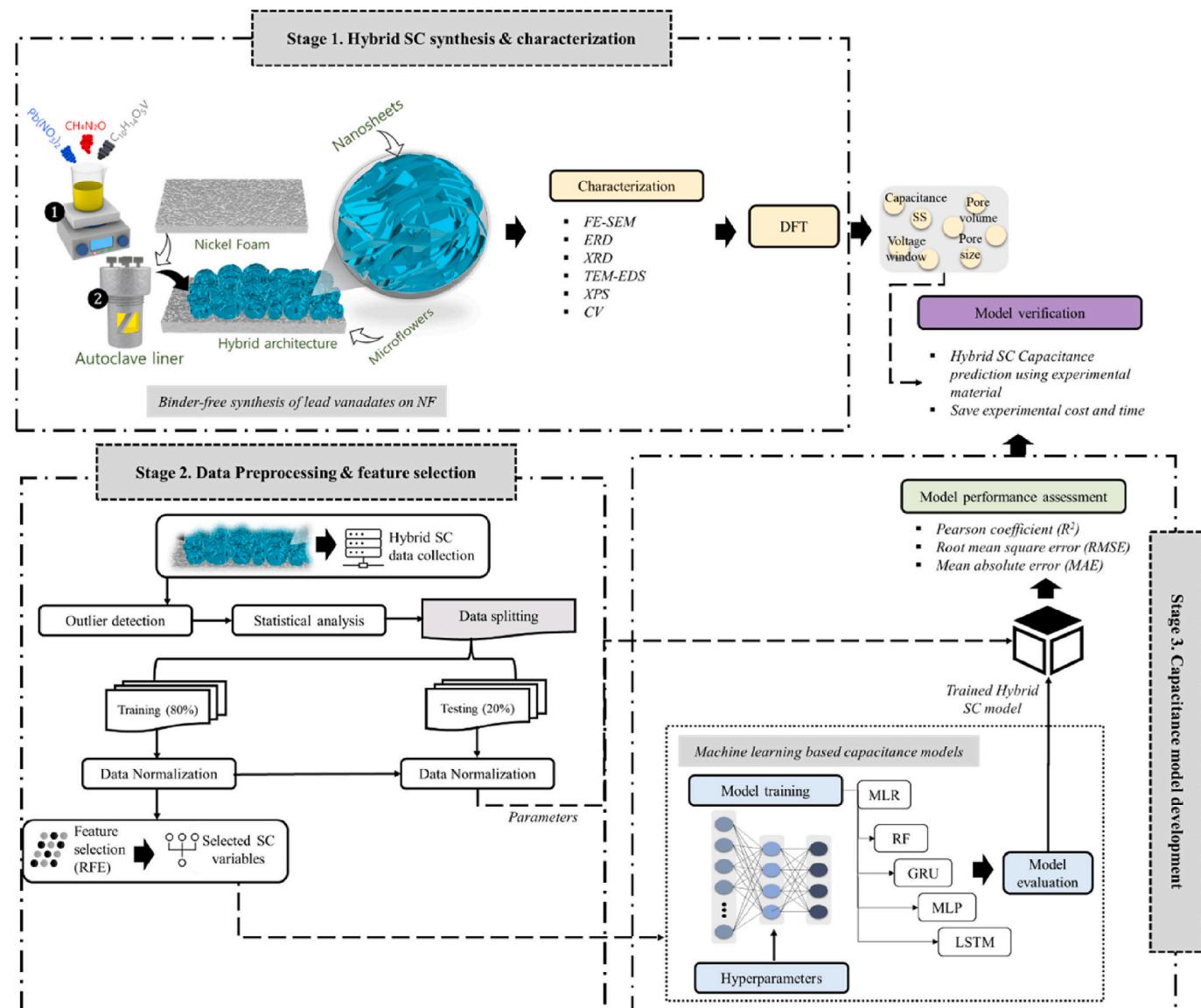


Fig. 22. Workflow diagram of the lead-vanadate hybrid SC [24]. Copyright 2024, Elsevier Ltd.

use of MD and DFT elucidated the mechanisms of Th adsorption and charging within the porous system, which is crucial for the design of advanced redox-active electrolytes for SCs.

Sial et al. [24] developed a new binder-free method for synthesizing lead vanadate (PbV) electrodes with unique morphology on nickel foam using a single-step hydrothermal method, and constructed a hybrid SCs dataset. By optimizing the crystal structure of PbV measured experimentally, DFT was employed to study the electrochemical characteristics, revealing that changes in vanadium concentration could introduce oxygen vacancies, enhance electrochemical activity, and improve electrical conductivity, thereby verifying the experimental results and guiding the development of hybrid SC devices. To predict the capacitance of PbV-based hybrid SCs, several ML methods, including MLR, random forest (RF), and multilayer perceptron (MLP), were used alongside RNN. Recursive feature elimination (RFE) was implemented for feature selection, and cross-validation was employed to assess the accuracy of the classifiers after feature selection (Fig. 22). In summary, experiments provide the data base, DFT offers theoretical validation, and ML enhances predictive capabilities. This integrated approach enables a comprehensive understanding and optimization of the SC performance of PbV hybrid architectures.

Ghosh et al. [238] integrated experimental, DFT, and ML approaches to investigate the temperature-dependent performance of cerium oxynitride-based solid-state SCs. First, they prepared CeO_xN_y electrodes and tested the SCs across a temperature range of 10 °C to 70 °C, obtaining electrochemical data that showed good charge storage capabilities and low resistance. DFT simulations were employed to elucidate the electronic structure evolution of the material with temperature, revealing significant changes such as lattice expansion, bandgap narrowing (from 2.82 eV at 0 °C to ~1.63 eV at 70 °C), and shifts in the DOS. DFT provided crucial theoretical insights into how temperature variations directly influence ionic mobility and charge storage mechanisms, thereby offering a comprehensive understanding of the material's behavior under different thermal conditions. The ML dataset was constructed by combining experimental parameters (e.g., current, temperature, voltage) and DFT-derived features (e.g., bandgap, Fermi level). DT and RF algorithms were selected to build ML models for the prediction of SCs charge-discharge behavior at high temperatures. Furthermore, a new variable, time period was introduced to model the relationship between temperature, current, and the time for charge-discharge cycles, enabling the ML models to capture both linear and exponential behaviors. The interaction between methods was crucial: experimental results validated DFT insights (e.g., thermal lattice effects on bandgap), DFT parameters enriched ML inputs, and ML predictions reduced reliance on detailed high-temperature testing. This integration of experimental data, DFT insights, and ML predictions created a comprehensive framework that not only enhances the understanding of temperature-dependent performance but also provides a robust tool for optimizing SC materials and systems.

Zhang et al. [239] conducted a comprehensive study on the capacitive charge storage mechanism in nitrogen-doped microporous carbon electrodes by integrating MD, MC, DFT, and ML methods. The workflow began with ab initio molecular dynamics (AIMD) simulations, first focusing on planar graphene/electrolyte interfaces to analyze the effects of nitrogen doping on capacitance. These simulations revealed that nitrogen doping (N₃ and N₅) is beneficial for increasing capacitance. The planar electrode simulations were unable to explain observed capacitance performance in nitrogen-doped porous carbon electrodes, necessitating further investigation into microporous structures. To address this, the researchers performed further AIMD simulations on microporous carbon electrodes, utilizing the Poreblazer software with a MC method to determine the GAP microporous carbon structure. Although the 3D microporous carbon/electrolyte interfaces were initially constructed with AIMD, they required further equilibration for accurate analysis. To overcome this limitation, they developed a ML force field to enhance the computational efficiency. This force field was trained on

data derived from DFT-based AIMD simulations, facilitating accelerated molecular dynamics (MLMD) simulations. The MLMD approach allowed for the equilibration of the interfaces at a significantly reduced computational cost while preserving the precision of ab initio methods.

The combination of AIMD, MC, DFT and ML techniques not only accelerated the simulation process but also enabled the exploration of larger and more complex systems that are difficult to calculate by traditional methods. The integrated approach provided a nuanced understanding of the electrochemical behavior of nitrogen-doped porous carbon electrodes, highlighting the pivotal role of nitrogen doping in enhancing capacitive performance and elucidating the underlying mechanisms at the molecular level.

In summary, the integration of DFT, MD, MC simulations, and ML has significantly advanced SC research. DFT provides electronic structure calculation as the basis, MD simulates dynamic behavior, MC evaluates statistical and thermodynamic properties, and ML identifies optimal configurations and predicts performance [240]. The robust data transfer mechanisms enable seamless collaboration among these methods, allowing cross-validated insights from quantum-level properties to macroscopic device performance. This multi-method collaborative approach not only accelerates the discovery and development of advanced SC materials but also ensures a more comprehensive and detailed optimization of their energy storage capabilities.

5. Conclusions and prospects

This review provides a comprehensive overview of the computational simulation methodologies applied in SCs research, which highlights the distinct applications and advantages of various computational methods, emphasizing the importance of both DFT and ML in advancing material development and performance optimization.

DFT calculations offer detailed insights into the atomic and molecular characteristics of materials. By accurately describing their electronic structure, bonding properties, and charge distribution from a quantum mechanical perspective, DFT can elucidate the electrochemical reaction mechanisms and adsorption behaviors on material surfaces. This is vital for understanding the microscopic factors influencing the performance of SC electrode materials, such as charge storage and ion adsorption processes. DFT is applicable to a wide range of materials, such as carbon-based materials (e.g., graphene), metal oxides, and MOFs, making it a universal tool in the field of SC research. Moreover, DFT can predict various physical and chemical properties of materials, including electrical conductivity, specific capacitance, adsorption energy, and diffusion coefficients. These predictions are highly valuable for designing new SC materials, as they can reduce the trial-and-error nature of experiments, thereby saving time and resources. However, DFT has limitations in calculating complex systems and dynamic processes, and it often requires simplifications that may lead to deviations from actual results. To achieve more comprehensive and accurate predictions of SC material properties, it is essential to combine DFT with other computational methods, such as ML, along with experimental validation.

ML has emerged as a powerful tool for data-driven modeling in SC research. It can analyze large datasets and identify patterns that inform predictions, optimize material design, and monitor the SOH of SC devices. The outstanding algorithms, including ANNs, SVM, and ensemble methods like XGBoost, have been effectively employed to predict performance metrics and optimize material design [241]. However, the effectiveness of ML models is heavily dependent on data quality and feature selection. Techniques such as data imputation and transfer learning can alleviate problems related to limited and incomplete datasets [242]. Data imputation fills in missing values, while transfer learning allows models to use knowledge from related tasks or domains, enhancing the robustness and accuracy of models and making them more applicable in real-world scenarios [243].

Looking forward, the interdisciplinary nature of SC research presents

immense opportunities for further innovations. The integration of DFT and ML with MD and MC simulations forms a powerful framework for advancing SC research, as they each have distinct and complementary roles. DFT provides detailed insights into electronic and geometrical properties, while ML accelerates predictions and material discovery by learning from DFT-generated data. MD captures dynamic behaviors like ion diffusion and phase transitions, and MC explores configurational space for thermodynamic properties. Combining these methods offers a comprehensive understanding of electrode materials' dynamics and charge storage mechanisms, leading to the creation of advanced SCs with improved energy storage capabilities and faster charge / discharge rates. Although the combination of multiple methods has many advantages, there are still some challenges in practical applications, particularly the trade-offs between computational cost and data quality in DFT-ML integration and the interpretability of ML models in multi-method collaborative study. Data augmentation, transfer learning and quantum-enhanced machine learning are expected to improve the quality and quantity of training data, reduce the need for extensive DFT calculations, and thereby ensure robust and efficient DFT-ML integration. To enhance ML interpretability, several strategies can be considered. For instance, feature engineering could focus on extracting physically meaningful features from different data sources and analyzing their importance to better understand their roles in model predictions. Additionally, simpler models or simplified complex models may offer clearer relationships between inputs and outputs, which could potentially improve interpretability. Visualization tools (such as PCA) and interpretability methods (like SHAP) might also be useful in explaining model behavior and feature impacts, thereby making complex data integration more understandable.

With the continuous development of the SCs field, the synergy between these computational techniques and experimental validation will undoubtedly drive further innovations, leading to the development of next-generation SCs with enhanced performance and broader applications. Future research should focus on refining these integrated approaches and exploring new computational tools to unlock even greater potential in the realm of energy storage. Overall, we believe this review will inspire new endeavors in computational simulations and drive further advancements in the design and development of high-performance SC technologies.

CRediT authorship contribution statement

Yawen Dong: Writing – original draft, Conceptualization. **Yutong Liu:** Writing – original draft, Conceptualization. **Feifei Mao:** Writing – review & editing, Supervision. **Hua Wu:** Writing – review & editing, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

Data will be made available on request.

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