

Synergistic Strategies for High-Energy Carbon Supercapacitors: A Comprehensive Review of Nanostructure, Doping, Composite, and Electrolyte Engineering

Syed Shaheen Shah,* Manisha Das, Takaya Ogawa, Asif Ali, Laiq Zada, Sana Ullah, Zafar Said, Muhammad Usman, Adnan Younis, Md. Abdul Aziz,* and Munetaka Oyama



Carbon-based supercapacitors provide high power, fast charging, and long cycle life, yet limited energy density remains the main bottleneck. This review integrates complementary strategies that enhance energy storage without sacrificing rate capability or durability. Nanostructure engineering tunes hierarchical porosity, pore-size to ion-size matching, and surface curvature to maximize ion accessibility and shorten transport paths. Heteroatoms doping introduces fast surface redox, improves electronic structure and wettability, and stabilizes interfacial chemistry. Composite architectures that couple carbon with pseudocapacitive phases, including metal oxides, conducting polymers, MXenes, and MOF-derived materials, build percolated electron and ion pathways and mitigate mechanical degradation. Electrolyte optimization expands voltage

and kinetics through water-in-salt formulations, gel polymer and solid-state media, and bio-derived electrolytes, with attention to desolvation, viscosity, and thermal tolerance. Emphasis is placed on the electrolyte-electrode interface, including ion confinement, interphase growth, and charge compensation mechanisms. An interaction-aware framework integrates pore architecture, doping chemistry, composite selection, and electrolyte design to propose design rules and performance trade-offs. Remaining gaps include operando diagnostics at relevant length scales, scalable synthesis with narrow property variance, aging models that link microstructure to failure, and sustainability metrics. Prospects center on high-energy, durable carbon supercapacitors for electric vehicles and renewable grid buffering.

1. Introduction

In response to the significant surge in energy consumption and the global shift toward sustainable energy solutions, there has

S. S. Shah, M. Das, T. Ogawa
 Socio-Environmental Energy Science Department
 Graduate School of Energy Science
 Kyoto University
 Yoshida-honmachi, Sakyo-ku, Kyoto 606-8501, Japan
 E-mail: shah.syedshaheen.3d@kyoto-u.ac.jp

A. Ali
Department of Applied Chemistry
Graduate School of Science and Engineering
Doshisha University
Kyoto, Japan

L. Zada
International College
University of Chinese Academy of Sciences
Beijing 100049, P. R. China

S. Ullah
Institute for Fiber Engineering & Science (IFES)
Research Cluster for Social Implementation (RCSI)
Shinshu University
Tokida 3-15-1, Ueda 386-8567, Nagano, Japan

Z. Said
Mechanical and Aerospace Engineering Department
College of Engineering
United Arab Emirates University
Al Ain 15551, UAE

M. Usman, M. A. Aziz
Interdisciplinary Research Center for Hydrogen Technologies and Carbon Management (IRC-HTCM)
King Fahd University of Petroleum & Minerals
Dhahran 31261, Saudi Arabia
 E-mail: maziz@kfupm.edu.sa

A. Younis
Department of Physics
United Arab Emirates University
Al Ain 15551, UAE

M. Oyama
Department of Material Chemistry
Graduate School of Engineering
Kyoto University
Nishikyo-ku, Kyoto 615-8520, Japan

been a marked increase in the demand for energy storage systems. In energy conservation, there is a notable demand for devices that can effectively bridge power and energy densities, placing energy storage systems at the forefront of research and development in the current energy landscape. While batteries can achieve high energy densities, they often face challenges related to power density and cyclic stability.^[1,2] Supercapacitors offer a promising alternative due to their exceptional power density, rapid charge and discharge capabilities, and prolonged cycle life.^[3] Carbon-based materials are the most popular choice for supercapacitors due to their plentiful availability, affordability, customizable characteristics, and eco-friendliness.^[4] However, supercapacitors can revolutionize energy storage systems if their electrochemical performance is sufficiently enhanced.^[5–10]

Carbon-based supercapacitors leverage the unique properties of carbon materials, including their large specific surface area (SSA), excellent electrical conductivity, and chemical stability.^[6] The capacitive performance of these devices is primarily determined by how effectively the electrode material's SSA is utilized and the existence of a porous triple interface with the electrolyte.^[11] These characteristics make carbon-based materials highly attractive for supercapacitor applications; however, their natural energy-storage capabilities frequently require enhancement to meet the demands of future energy devices.^[12] This has prompted a thorough examination of methods for improving capacitance, focusing on both the materials used and the device configuration. Designing nanostructures has become a crucial strategy for enhancing the capacitance of carbon-based supercapacitors.^[13] Designing and creating carbon materials at the nanoscale enables precise management of pore-size distribution, surface structure, and the accessibility of active sites.^[14] Hierarchical porous carbons (HOPCs),^[15] graphene sheets,^[16] and carbon nanotubes (CNTs)^[17] are nanostructures that hold significant promise for enhancing ion transport and charge storage capabilities. These materials can significantly improve the electrode's SSA and facilitate swift ion movement within the electrode, which is crucial for developing high-performance supercapacitors. Furthermore, combining various carbon nanostructures can yield enhanced electrochemical advantages, positioning

nanostructure engineering as a fundamental aspect of capacitance enhancement. The heteroatom doping technique represents notable progress in improving the electrochemical performance of carbon-based supercapacitors. Adding heteroatoms, such as nitrogen, oxygen, sulfur, or phosphorus, into the carbon framework can significantly modify the material's electronic structure and surface chemistry.^[18] Dopants can create defects, enhance the conductivity, and provide additional active sites for charge storage, collectively contributing to the overall performance enhancement of the material.^[19] An example that has been thoroughly examined is nitrogen doping, which enhances the pseudocapacitive properties and wettability of carbon-based materials.^[20,21] Similarly, various heteroatoms provide unique electrochemical advantages that enable the precise optimization of performance for specific applications.^[22,23]



Syed Shaheen Shah is a program-specific assistant professor at the Graduate School of Energy Science, Kyoto University, Japan. He earned his Ph.D. in Physics from King Fahd University of Petroleum & Minerals (KFUPM), Saudi Arabia, in 2022, after completing his M.Phil. (2017) and B.S. (2015) in Physics from the University of Peshawar, Pakistan. Dr. Shah previously served as a lecturer at KFUPM (2018–2022) and as a JSPS postdoctoral fellow in the Graduate School of Engineering at Kyoto University (2022–2024). His research focuses on advanced nanomaterials for energy harvesting and storage, including supercapacitors and electrochemical sensors.



Manisha Das is a postdoctoral researcher and JSPS Fellow at Kyoto University, Japan, specializing in electrochemical energy technologies. Her research focuses on the development of advanced electrocatalysts for water splitting, fuel cells, and ammonia synthesis, aiming to create sustainable energy systems. She holds a Ph.D. from INST Mohali, India, where she engineered multifunctional nanostructured electrocatalysts for HER, OER, and ORR. Dr. Das has published extensively in reputed journals and coauthored a CRC Press book on carbonaceous energy materials. She actively mentors students, engages in interdisciplinary collaborations, and contributes to science outreach and education.



Takaya Ogawa is an associate professor in the Department of Socio-Environmental Energy Science at Kyoto University's Graduate School of Energy Science. He holds a master's in engineering from the University of Tokyo, a master's in management of technology, and a Ph.D. in science from the Tokyo Institute of Technology. His research focuses on sustainable energy solutions, including ammonia synthesis using renewable energy, proton-conducting materials, catalysts, bibliometrics, and life cycle assessment. Dr. Ogawa has held notable roles, serving as a postdoctoral fellow at MIT's Department of Chemistry (2015–2017) and Stanford University's SUNCAT Center (2017–2018). He joined Kyoto University as an assistant professor in 2018 and became an associate professor in 2023.

A notable strategy for enhancing the capacitance of carbon-based supercapacitors involves creating composites with pseudocapacitive materials. In pursuit of this goal, different carbon materials have been combined with pseudocapacitive elements, such as metal oxides,^[24] layered double hydroxides (LDHs),^[25] conducting polymers,^[26] or transition metal dichalcogenides (TMDs),^[27] to create a synergistic strategy that leverages energy storage through both electric double-layer capacitance and faradaic charge storage. The combination of high-power density in carbon materials and the high energy density of pseudocapacitive materials leads to supercapacitors demonstrating exceptional electrochemical performance. Integrating these components presents significant challenges from interface interactions and long-term stability requirements. However, the results have shown substantial improvements in capacitance



His accolades include the NF Foundation R&D Encouragement Award (2022) and the JACI Prize for Encouraging Young Researcher (2021). **Asif Ali** is currently pursuing his Ph.D. in Applied Chemistry at Doshisha University in Kyoto, Japan. He holds an MPhil in Environmental Chemistry from Abdul Wali Khan University (AWKUM), Pakistan, where he laid the groundwork for his interest in environmental and materials-based research. At Doshisha, his work spans nanoparticle synthesis, adsorption studies, and catalyst development, with publications in areas such as dyes removal and silver nanoparticle synthesis. Through his cross-disciplinary approach, he aims to address challenges at the interface of chemistry and sustainable environment.



Laiq Zada is currently pursuing a Ph.D. degree in microbiology at the Tianjin Institute of Industrial Biotechnology, Chinese Academy of Sciences (TIB-CAS), China. Mr. Zada obtained his bachelor's degree from the Department of Microbiology, Abdul Wali Khan University, Mardan, Pakistan, in 2020, and his master's degree from the Department of Microbiology, Quaid-i-Azam University, Islamabad, Pakistan, in 2023. Before joining the TIB-CAS, he was a research assistant at the National Institute of Laser and Optronics, Islamabad, Pakistan. His research interests include antibacterial materials, antimicrobial photodynamic therapy (aPDT), biocontrol agents, and plant-microbe interactions.



Sana Ullah is serving as a specially appointed assistant professor at the Institute for Fiber Engineering and Science (IFES), Shinshu University, Japan. He holds a doctoral degree in engineering (polymeric materials) from Shinshu University. Before joining IFES, he worked as a researcher at Ulm University, Germany, and the Post Lithium Storage Cluster of Excellence (POLiS), Germany, between February 2023 and December 2024. Currently, he is leading a team who focuses on researching the innovative methods for the conversion and storage of sustainable energy (green hydrogen, CO₂ reduction, sodium and potassium ion batteries).

and energy density. In recent years, considerable focus has been directed toward synergistic approaches that integrate various strategies. Utilizing nanostructure engineering, heteroatom doping, and composite formation enables the exploration of the complementary benefits of these techniques, leading to exceptional improvements in capacitance.^[28–31] Incorporating heteroatoms into carbon nanostructures has been extensively explored, allowing for the *in situ* embedding of pseudocapacitive materials within the carbon matrix. This approach has enabled the development of multifunctional and highly efficient electrodes.^[32] This approach overcomes previous drawbacks and offers a cohesive strategy for high-performance supercapacitors based on carbon materials, highlighting interdisciplinary innovations that drive progress in energy storage devices.

Electrolytes significantly influence the enhancement of the capacitance. Supercapacitor electrolytes are crucial as they facilitate the movement of ions into and out of the electroactive materials during charge/discharge cycles, influencing the overall

electrochemical properties of the device.^[8] Various advanced electrolytes, including ionic liquids (ILs),^[33] aqueous solutions,^[34] and solid-state electrolytes,^[35] have been investigated to enhance the ionic conductivity, stability, and operating potential window (OPW). Therefore, capacitance can be improved by increasing the electrode SSA and enhancing electrolyte-electrode interactions, with novel electrolytes and advanced carbon materials playing a key role in advancing supercapacitor technology.

Across the strategies reviewed here, the governing trade-off is simple: energy density scales with both capacitance and voltage ($E \propto 1/2CV^2$), while power and durability are constrained by transport and mechanical integrity. Nanostructure engineering reliably lifts EDLC contributions (typical 200–350 F g⁻¹ for high-SSA carbons; HOPC 289 F g⁻¹, 40 Wh kg⁻¹, 20 kW kg⁻¹)^[36] but can suffer from tortuous ion pathways and poor volumetric metrics when micropores dominate. Heteroatom doping introduces redox sites and lowers interfacial resistance (reported 306–660 F g⁻¹, up to 108 Wh kg⁻¹ with stable cycling),^[37,38] yet excessive dopant



Zafar Said is a distinguished associate professor at the Department of Mechanical and Aerospace Engineering at UAE University. With over 6 million AED in research funding, he has led industry-focused projects with SEWA, Tabreed, and Masdar, advancing innovations in nanofluids, energy systems, advanced thermal management, NePCM, and energy storage, and he is recognized among the world's top 2% of scientists (2020–2023) by Elsevier BV and Stanford University in the field of energy. He ranks 1st in Energy Technology by ScholarGPS for the previous 5 years. He has received multiple prestigious awards, including the 13th Future Pioneer Award in Sustainability, Beeah, Sharjah, 2025; the Best Academic Research at the 13th Dubai Award for Sustainable Transport, 2024; and First Place in Scientific Research at the Excellence and Creative Engineering Award, 2023, by the Society of Engineers, UAE. He also won the Research and Innovation Award from the UAE Ministry of Energy and Infrastructure for 2022 and 2023 and the Rising Star Science Star award by Research.com for three consecutive years. His work aligns with the UAE's Net Zero 2050 Strategy, positioning him as a key collaborator to develop sustainable energy solutions and empower future leaders in the energy sector.



Muhammad Usman is a Research Scientist-II at the Interdisciplinary Research Centre for Hydrogen Technologies and Carbon Management, King Fahd University of Petroleum and Minerals (KFUPM), Saudi Arabia. His research focuses on CO₂ capture and utilization, green hydrogen, heterogeneous catalysis, and membrane technologies. He has authored over 140 journal articles, over 30 US patents, and over 5000 citations and is recognized among the world's top 2% of scientists in chemical engineering (Stanford/Elsevier).



Adnan Younis is an associate professor in the Department of Physics at the College of Science, United Arab Emirates University (UAEU). His research expertise lies in advanced functional materials, with a particular emphasis on inorganic compounds, perovskites, 2D materials such as MXenes, and their applications in electronics, energy storage systems, thin films, and semiconductor nanostructures. He has authored over 90 peer-reviewed publications in high-impact international journals, including Advanced Materials, Advanced Functional Materials, Nano-Micro Letters, Small, and Advanced Science.



Md. Abdul Aziz received his M.Sc. in organic chemistry in 2001 from the University of Dhaka, Bangladesh. In 2009, he earned his Ph.D. in chemistry from Pusan National University, Republic of Korea. He then worked as a postdoctoral fellow in the Department of Material Chemistry, Kyoto University, from 2009 to 2011. He is now a Research Scientist-II at Interdisciplinary Research Center for Hydrogen Technologies and Carbon Management (IRC-HTCM), King Fahd University of Petroleum & Minerals, Saudi Arabia. His main research interests are preparation, immobilization, and functionalization of nanomaterials and carbonaceous materials and their application in chemical and biochemical gas sensors, water oxidation, supercapacitors, and so forth.



Munetaka Oyama received his D.Sc. degree in 1991 from the Department of Chemistry, Graduate School of Science, Kyoto University, Japan. He is now an associate professor of the Department of Material Chemistry, Graduate School of Engineering, Kyoto University, Japan. His research interests include the development of electroanalytical methods using nanomaterials. In particular, his current research topic is the application of noble metal-modified nickel electrodes prepared via galvanic replacement reactions. He has published over 250 articles in scientific journals.

content and defect density can depress graphitization and long-term stability. Composites with pseudocapacitive phases deliver the largest per-gram capacitances (e.g., $\text{Co}_3\text{S}_4/\text{rGO}$ 1442.5 F g⁻¹, 47.7 Wh kg⁻¹,^[39] NiCo-LDH/carbon felt 1661.6 F g⁻¹, 57.2 Wh kg⁻¹,^[40] but introduce challenges in interface adhesion, ion/electron percolation, and swelling over long cycles. Electrolyte design shifts the voltage ceiling and transport: aqueous systems are fast but narrow-window; organic/IL and water-in-salt widen OPW up to \approx 3–4 V and increase E markedly (micro-emulsion \approx 40 Wh kg⁻¹,^[41] water-in-salt electrolyte \approx 3.0 V),^[42] at the expense of viscosity or cost. The clearest gains arise when these levers are co-optimized, rather than applied in isolation.

While the vast literature available addresses the nanostructure design, heteroatomic doping, the compositing procedure, and the electrolyte optimization, these methods are often studied separately. There is, thus, a significant knowledge gap to bridge in the synergistic integration of processes that surpass the limits set by existing approaches in raising the energy density in carbon-based supercapacitors. This review carefully addresses the current gap by evaluating in-depth recent progress in these areas and highlighting their coherent integration. In contrast to previous reviews, this article highlights how multidisciplinary approaches synergize to promote the performance of supercapacitors. Moreover, it sheds light on the interactions between electrolytes and electrodes as crucial but often neglected elements that influence the overall capacitance enhancement. It efficiently addresses current shortcomings in meeting the increasing demand for green energy storage technologies by providing a comprehensive, multi-disciplinary blueprint for designing next-generation carbon-based supercapacitors. This review article extensively discusses synergistic strategies to enhance capacitance in carbon-based supercapacitors, with a particular emphasis on the interplay between nanostructure engineering, heteroatom doping, composite formation, and electrolyte optimization. It takes a balanced view by recognizing recent progress and the general extended implications in the field. The distinguishing feature of this work lies in its systematic examination of synergistic approaches, highlighting how the concerted application of multiple strategies can successfully overcome the conventional limitations of established methods. This review differs from previous works by providing an integrated and synergistic analysis of nanostructure engineering, heteroatom doping, composite formation, and electrolyte optimization. Rather than discussing these strategies separately, this work emphasizes how their complementary effects can lead to substantial improvements in supercapacitor performance, particularly in enhancing both energy density and cycling stability. The article opens promising avenues toward creating next-generation supercapacitors that can address the high-level requirements of modern energy storage systems.

2. Nanostructure Engineering for Enhanced Capacitance

Recent progress in nanostructure engineering has enabled the development of more refined strategies for improving the

electrochemical performance of carbon-based supercapacitors. By meticulously tailoring the nanoscale morphology and structural architecture of carbon materials, notable improvements have been realized in terms of specific capacitance, rate performance, and long-term cycling stability. Nanostructured carbon materials, in particular, offer considerable advantages, including high SSAs, well-regulated pore architectures, and efficient pathways for ion diffusion and electron transport.^[43] The significance of these characteristics lies in their ability to enhance charge storage and improve overall energy storage performance.^[14] Various dimensional nanostructures, including 0D, 1D, 2D, and 3D, offer distinct advantages that enhance the electrochemical performance.^[44] Examples of 1D nanostructures encompass CNTs and carbon nanofibers (CNFs),^[45] which offer outstanding electrical conductivity and mechanical stability, making them advantageous for durable electrode design. In contrast, 2D materials such as graphene derivatives provide significant SSA and flexibility, facilitating improved charge storage and transport.^[46] Alternatively, 3D hierarchical structures, such as aerogels and carbon foams, integrate the advantages of different dimensionalities, resulting in a synergistic improvement in capacitance.^[47] These 3D structures frequently incorporate functional networks and layered pore distributions to optimize the available SSA for ions while promoting electrolyte penetration and minimizing the resistance to ion diffusion.^[48] Figure 1 illustrates the dual solid and cavity network structure of 3D graphene, featuring an sp^2 network that enhances electronic, phononic, and mechanical transport while preventing restacking.^[49] It highlights various synthesis approaches, such as liquid-based assembly, chemical vapor deposition, and pyrolysis, alongside the applications of 3D graphene in aqueous and non-aqueous electrochemical double-layer capacitors (EDLC), leveraging its high SSA and mass transfer capabilities. A crucial aspect of nanostructure engineering is the precise control of the pore size distribution, particularly the ratio of micropores to mesopores. Micropores provide substantial SSA and enhance charge storage capabilities, whereas mesopores facilitate better electrolyte accessibility and enhance ion transport efficiency.^[50] Achieving optimal equilibrium in this balance is essential for improving electrochemical performance. Sophisticated synthesis techniques and computational tools enable the design of nanostructured carbons with optimized pore sizes, distributions, and connectivity to enhance charge storage in supercapacitors.^[12,51–53] Nanostructure engineering addresses the limitations of conventional materials, advancing global supercapacitor technology with improved performance and durability.

Nanostructure engineering has emerged as a pivotal strategy to enhance capacitance in energy storage devices, leveraging advanced material designs to optimize SSA, ion diffusion, and charge transfer. $\text{MoSe}_2/\text{polyaniline}$ (PANI) composites exhibit enhanced capacitance due to the incorporation of hollow microspheres (740 nm diameter) and synergistic effects, achieving a specific capacitance of 753.2 F g⁻¹ with a 7-fold increase in surface area (SSA) to 44.49 m² g⁻¹.^[54,55] Similarly, 3D nanostructured silver electrodes exhibit an areal capacitance of 500 mF cm⁻² and 95% retention over 2,600 cycles, attributed to reduced ion diffusion paths and pseudocapacitive contributions.^[56] Bio-inspired

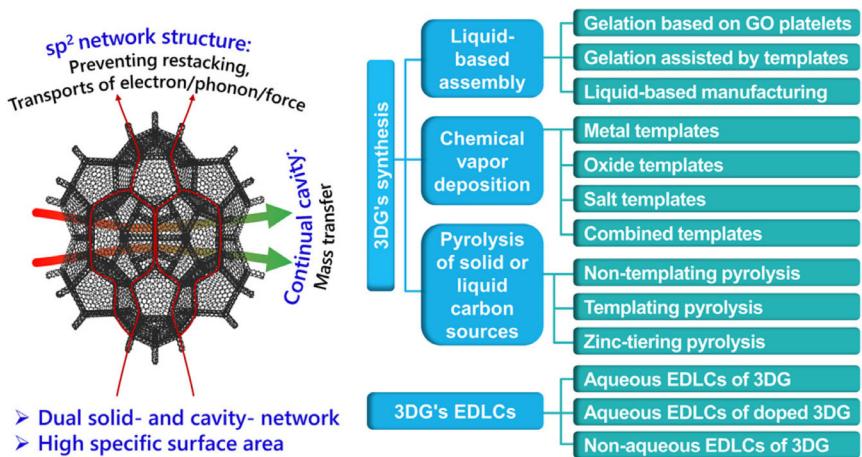


Figure 1. Structural and functional overview of 3D graphene and its synthesis methods. Reproduced with permission.^[49] Reproduced under the term CC BY 4.0. Copyright 2021; Zeng et al. Wiley.

coral reef-like NiCo-LDH on carbon felt achieves 1661.6 F g^{-1} at 1 A g^{-1} , combining vertical nanosheets for efficient ion transport.^[40] MnO₂/MnS nanocomposites exhibit a specific capacity of 305 F g^{-1} with an energy density of 5.7 Wh kg^{-1} , enhanced by defect-induced active sites.^[57] Ternary YSe₂/MoSe₂/Ni_xB heterostructures achieve a specific capacitance of 893.3 F g^{-1} and 128% capacitance retention over 5,000 cycles.^[58,59] Mn-doped MoSe₂ exhibits a 313% capacitance increase via lattice distortion.^[58] Pb_{0.97}La_{0.02}(Zr_{0.35}Sn_{0.55}Ti_{0.10})O₃ antiferroelectric capacitors achieve a 54.3% capacitance boost through electric-field-driven phase transitions, stabilizing performance.^[60] Ti₃C₂T_x nanosheets with size engineering exhibit 84% surface-controlled capacitance due to an optimized interlayer spacing.^[61] Nitrogen-doped CNFs exhibit a capacitance of $1,145 \mu\text{F}$ at 120 Hz with a phase angle of -80° , making them ideal for high-frequency applications.^[62] These advances highlight the crucial role of tailored nanostructures in optimizing capacitive performance.

Recent reviews have extensively addressed nanostructure design in carbon-based electrodes,^[63–65] emphasizing traditional porous carbon structures and their electrochemical performance. However, this review uniquely advances this discussion by explicitly highlighting hierarchical and multifunctional nanostructures, providing novel insights into the precise relationship between nanostructural features and electrochemical performance metrics. Additionally, we extensively discuss emerging techniques such as electrospinning, template-assisted synthesis, and advanced 3D graphene architectures, emphasizing their integrative potential when combined with doping and composite strategies. This area has been less comprehensively reviewed previously. Within carbon architectures, hierarchical porosity consistently outperforms single-scale porosity, as it preserves high gravimetric capacitance while sustaining high rates (EDLC plateaus $\geq 250\text{--}300 \text{ F g}^{-1}$ at modest current densities; HOPC 289 F g^{-1} , 40 Wh kg^{-1} , 20 kW kg^{-1}).^[36] Purely microporous carbons maximize SSA but raise ion-transport resistance and hurt volumetric/areal figures at practical mass loadings; purely mesoporous scaffolds improve rate but sacrifice surface-limited

storage. 3D networks (graphene foams, CNT/graphene hybrids) shorten diffusion lengths and mitigate restacking, enhancing power and cycling, yet incur lower packing density and sometimes lower absolute energy per unit volume. Synergistically pairing hierarchical carbons with moderate heteroatom contents or thin pseudocapacitive skins alleviates transport bottlenecks without collapsing pores, whereas thick active layers or over-etching/over-activation compromise conductivity and mechanical integrity.

2.1. Importance of SSA and Pore Structure

The effectiveness of carbon-based supercapacitors, especially regarding their capacitance and energy-storage potential, is greatly affected by the carbon electrodes' SSA and pore structure. An elevated SSA offers numerous active sites for ion adsorption, which directly improves the charge storage capacity, while well-defined pore structures promote efficient ion transport and accessibility. Activated carbon (AC), often derived from biomass, demonstrates the critical role of SSA, sometimes exceeding $2000 \text{ m}^2 \text{ g}^{-1}$, in boosting capacitance.^[25] Nonetheless, SSA is inadequate; the pore architecture, comprising micro-, meso-, and macropores, influences ion accessibility and affects electrochemical performance.^[66] The synergy between the SSA and pore structure is thus essential for optimizing the energy storage and efficiency of supercapacitors. **Figure 2a** depicts the hierarchical pore architecture, comprising micropores, mesopores, and macropores, alongside surface functional groups present in carbon-based materials. These structural and chemical features play a critical role in facilitating efficient ion transport, augmenting charge storage capacity, and improving the overall energy density of supercapacitors.^[67]

The SSA of carbon materials is a key determinant affecting the capacitance of supercapacitors. AC, characterized by its high SSA, is extensively used in supercapacitor applications because it offers many active sites for electrochemical reactions.^[68–70] The SSA is essential as it has a direct relationship with the charge

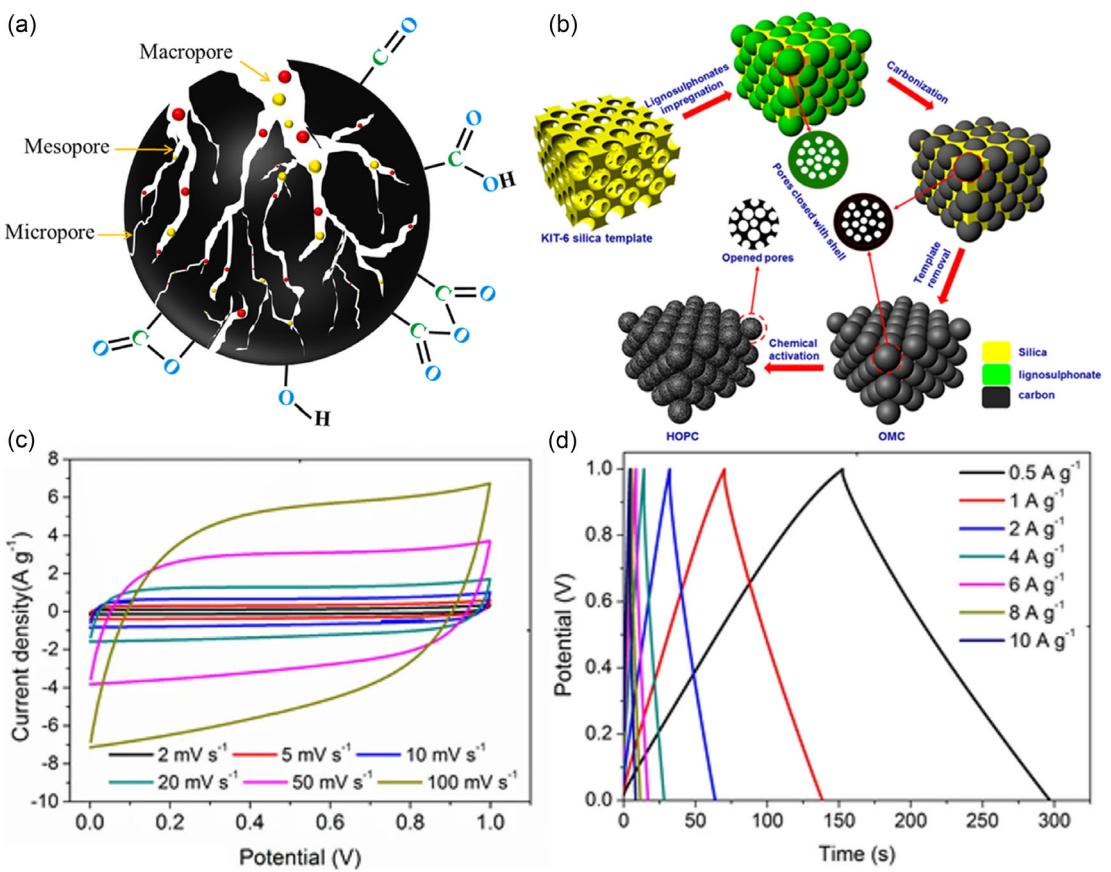


Figure 2. a) Schematic representation of hierarchical pores and functional groups optimizes ion dynamics for supercapacitors. Reproduced with permission.^[67] Reproduced under the term CC BY 4.0. Copyrights 2022; Sultana et al. Elsevier. b) Template-assisted synthesis of HOPC for efficient ion storage in supercapacitors. c) CV curves obtained at varying scan rates, and d) GCD profiles recorded at different current densities for the symmetric two-electrode supercapacitor with HOPC electrode utilizing a 6 M KOH aqueous electrolyte. Reproduced with permission.^[36] Reproduced under the term CC BY 4.0. Copyright 2020; Bai et al. Springer Nature.

storage capacity; an increased SSA facilitates the adsorption of more ions at the electrode–electrolyte interface, thus improving the overall capacitance.^[71,72] For instance, developing nitrogen-doped porous carbons has shown that increasing the SSA can significantly boost the specific capacitance owing to the additional pseudocapacitance contributed by the nitrogen functionalities.^[73,74] Moreover, the pore structure plays a vital role in the performance of supercapacitors. The size and distribution of the pores dictate how effectively ions can diffuse into the electrode material. Studies have shown that ideal pore sizes, ranging from 2.0 to 5.0 nm, are crucial for enhancing ion transport and increasing the availability of active sites for charge storage.^[75,76] This is because such pore sizes allow for efficient electrolyte infiltration while minimizing the thickness of the electrical double layer, which is critical for enhancing the capacitance.^[75] Additionally, hierarchical porous structures that combine micro- and mesopores have improved ion transport dynamics, leading to better charge–discharge performance.^[77,78]

The interplay between SSA and pore structure is also evident in the synthesis of carbon materials from biomass and other precursors. Techniques that optimize the activation process can lead to carbon materials with tailored pore structures and high SSA,

which are essential for achieving superior electrochemical performance in supercapacitors.^[79,80] For example, biomass-derived carbon has been shown to produce materials with excellent hierarchical porosity, enhancing the SSA and improving pore connectivity, facilitating better ion transport.^[81–83] Furthermore, introducing heteroatoms such as nitrogen and oxygen into the carbon framework can enhance the electrochemical properties of the electrodes. These dopants can increase the SSA and modify the pore structure, improving capacitance through additional charge storage mechanisms.^[84,85] The presence of heteroatoms can also enhance the wettability of the carbon surface, promoting better electrolyte interactions and ion accessibility.^[86,87] Bai et al. reported the use of HOPC with sodium lignosulfonate and a KIT-6 silica template.^[36] The synthesis combines hard-template methods and chemical activation with ZnCl₂, resulting in HOPC with a high SSA of 2602 m² g⁻¹ and a pore volume of 3.49 m³ g⁻¹. Figure 2b illustrates the synthesis process involving templating and activation, whereas Figure 2c,d show the electrochemical cyclic voltammetry (CV) profiles and galvanostatic charge–discharge (GCD) behavior. The HOPC demonstrated outstanding electrochemical performance as a supercapacitor electrode, delivering a specific capacitance of 289 F g⁻¹ at a current density

of 0.5 A g^{-1} , an energy density of 40 Wh kg^{-1} , and a maximum power density of 20 kW kg^{-1} . Furthermore, it exhibited robust cycling stability, retaining 86.2% of its initial capacitance after 3000 GCD cycles. The results highlight the material's nearly rectangular CV curves across scan rates, symmetrical triangular GCD profiles, and prolonged discharge time, confirming its high energy storage capability and rate stability.

Engineering the SSA and pore structure in carbon-based materials is pivotal for optimizing the electrochemical performance of supercapacitors. Hierarchical pore architectures that integrate micropores for charge storage, mesopores for ion transport, and macropores for electrolyte diffusion enable efficient ion dynamics and enhanced capacitance. Advanced synthesis methods, including templating, chemical activation, and self-assembly, allow the precise tuning of pore size, connectivity, and distribution. Meanwhile, surface modifications, such as heteroatom doping and functional group incorporation, enhance wettability, conductivity, and pseudocapacitance.^[88,89] Challenges such as balancing high SSA with conductivity and mechanical stability are addressed through synergistic design approaches, such as hierarchical porosity and surface functionalization. These advancements highlight the transformative potential of nanostructured carbon materials in supercapacitor technology, offering a viable pathway toward achieving both high energy and power densities necessary for next-generation energy storage systems.

High SSA is necessary but not sufficient: materials exceeding $2000 \text{ m}^2 \text{ g}^{-1}$ often show $200\text{--}350 \text{ F g}^{-1}$ in aqueous media, yet ion-sieving and slow electrolyte exchange emerge when micropores dominate. The HOPC ($2602 \text{ m}^2 \text{ g}^{-1}$; 289 F g^{-1} ; 40 Wh kg^{-1} ; 20 kW kg^{-1}) case^[36] illustrates that balanced micro/mesopores ($\approx 2\text{--}5 \text{ nm}$)^[75,76] deliver both capacity and rate. Doping can enhance wettability and quantum capacitance, but it may narrow effective pores or introduce tortuosity if functional groups cluster in necks; conversely, mild oxidation or N-sites strategically located at mesopore mouths can enhance accessibility. Practically, micropore volume for storage plus mesopore highways for transport is the winning combination, pushing SSA further without preserving connectivity yields diminishing returns and poorer volumetric metrics.

2.2. Strategies for Optimizing Pore Size Distribution

Optimizing the pore size distribution in nanostructured carbon materials is crucial for enhancing the capacitance and energy density of carbon-based supercapacitors. A balanced interplay of micropores, mesopores, and macropores improves ion transport, charge storage, and electrolyte accessibility, with each pore type serving distinct roles. Chemical activation using agents such as KOH,^[90] H_3PO_4 ,^[91] and ZnCl_2 ^[20] enables tailored pore engineering by adjusting the activation conditions to generate specific pore structures. For example, KOH^[90] is used to create micropores, while H_3PO_4 ^[91] is used to develop mesopores. Complementary physical activation with CO_2 or steam further refines the pore architecture,^[92] enabling the design of customized supercapacitors to meet advanced performance needs. One practical approach is synthesizing carbon materials with high SSA and a

narrow pore size distribution. The advancement of subnanometer porous carbon films demonstrates considerable potential in energy storage applications due to their capacity to offer numerous active sites for ion adsorption, a critical factor for achieving high capacitance.^[93] The synthesis method used can significantly influence the resulting pore structure, and techniques such as deep eutectic solvents have been utilized to create nitrogen-doped carbon with enhanced electrochemical performance.^[37] Moreover, incorporating heteroatoms into the carbon structure can further optimize the pore characteristics and enhance the electrochemical performance. Heteroatom doping, particularly with nitrogen, has been demonstrated to enhance the hydrophilicity of carbon materials, thereby facilitating improved ion transport and increased capacitance.^[94,95] The presence of heteroatoms can also create additional active sites, which are beneficial for charge storage mechanisms.^[96]

A key strategy involves adjusting the pore size to match the dimensions of the ions intended for storage. Research indicates that the most efficient ion adsorption occurs when the pore size is comparable to the ion size, as this maximizes electrostatic interactions and minimizes resistance during charge/discharge cycles.^[97] Therefore, precise control of the pore size distribution is essential for optimizing the performance of supercapacitors. Additionally, hierarchical porous structures have been identified as beneficial configurations for supercapacitors because they facilitate fast ion diffusion and provide a high SSA for charge storage.^[98,99] Combining different pore sizes within a single material can create a synergistic effect, thereby enhancing the overall capacitance and energy density of supercapacitors.^[100] Finally, advanced synthesis techniques, such as electrospinning and template-assisted methods, have been used to fabricate CNFs and other nanostructures with tailored pore sizes and distributions.^[101] These methods allow the creation of complex architectures that can significantly improve the electrochemical performance of supercapacitors.

Optimizing the pore size distribution in nanostructured carbon materials involves advanced synthesis methods, templating techniques, and chemical activation to enhance the performance of supercapacitors. Hard and soft templating strategies enable the precise customization of pore structures, with silica templates and block copolymers facilitating the development of hierarchical architectures.^[102,103] Integrating chemical activation, such as KOH activation, with templating techniques produces materials with extensive SSA, interconnected pores, and improved ion transport. Biomass-derived carbons offer a sustainable alternative, leveraging natural hierarchical structures for pore optimization.^[104,105] Hierarchical pore structures, which combine micropores, mesopores, and macropores, enhance charge storage, ion diffusion, and power density, underscoring the critical role of pore size tailoring in advancing carbon-based supercapacitor technologies for efficient and sustainable energy storage.

KOH activation effectively creates sub-2 nm micropores (high capacitance, lower rate), while H_3PO_4 biases toward mesopores (better rate, lower raw capacitance), and ZnCl_2 or physical activation (CO_2 /steam) offer intermediate control. Templating (hard/soft) yields narrow PSDs and ordered connectivity but adds steps

and cost; biomass routes are scalable and sustainable but require tighter control to avoid broad pore size distribution (PSDs). Quantitatively, templated/activated carbons in this work span $\approx 250\text{--}350 \text{ F g}^{-1}$ with up to 20 kW kg^{-1} power; subnanometer films can push surface utilization but risk high ESR at the device scale. The optimal PSD is electrolyte-dependent: pores approaching solvated-ion size demand partial desolvation (raising resistance), whereas slightly larger mesopores minimize barriers. Thus, PSD should be matched to ion size/viscosity and the intended OPW, not maximized in isolation.

2.3. Novel Carbon Nanostructures

The exploration of novel carbon nanostructures has significantly advanced the development of high-power supercapacitors by enhancing their capacitance, energy density, and charge–discharge rates, positioning them as essential components in energy storage systems. Among these innovations, 3D graphene structures are particularly noteworthy due to their high SSA, excellent electrical conductivity, and strong mechanical stability, which overcome lamination and agglomeration challenges in traditional graphene sheets.^[49] The interconnected porous architecture of 3D graphene facilitates efficient ion transport and electrolyte accessibility, optimizing energy storage performance.^[106] Advanced fabrication techniques, such as hydrothermal self-assembly, chemical vapor deposition, and freeze-drying, play a pivotal role in maintaining structural integrity and tailoring porosity to meet specific application requirements.^[107,108] These advancements underscore the crucial role of carbon nanostructures in electrode engineering, bridging the performance gap between traditional capacitors and batteries to achieve high-performance supercapacitors. Want et al. presented a novel synthesis of hybrid electrodes by combining porous lignin-based graphitic carbon with rhubarb, a small herbal biomolecule rich in redox centers, through ultrasonic treatment to enhance the charge storage.^[109] Figure 3a illustrates the synthesis of a sustainable biomass-based supercapacitor using lignin-derived porous carbon and rhubarb extract, integrated into a composite electrode and gel electrolyte, which emphasizes its high specific capacitance, excellent cycling stability, and eco-friendly design. The composite exhibits superior electrochemical performance as a supercapacitor electrode, attaining a specific capacitance of 250.2 F g^{-1} at a current density of 1.0 A g^{-1} , an energy density of 8.1 Wh kg^{-1} at a corresponding power density of 150.85 W kg^{-1} , and excellent cycling stability, retaining 80.9% of its initial capacitance after 3000 cycles. This innovative approach leverages redox-active biomolecules in conjunction with a gel-based electrolyte to effectively address prevailing challenges related to energy storage efficiency and environmental sustainability. Recent advancements in the fabrication of carbon-based materials, such as carbon dots and CNTs, have led to substantial improvements in specific capacitance. For instance, Phetcharee et al. reported a remarkable enhancement of $\approx 290\%$ in the specific capacitance of ZnO electrodes when integrated with carbon dots, demonstrating the effectiveness of carbon nanostructures in boosting electrochemical performance.^[110] Similarly, the incorporation of vertically oriented graphene nanosheets has been highlighted for

their significant SSA and high electrical conductivity, which are essential for efficient charge storage in supercapacitors.^[111] These structural characteristics enable more significant charge accumulation at the electrode interface, enhancing overall energy storage capacity. Moreover, engineering hybrid nanostructures, such as those combining CNTs with transition metal compounds, have been shown to yield superior electrochemical properties. For example, the work by Sreeraj et al. on S-doped VSe₂/CNTs hybrids illustrates how the synergistic effects of pseudocapacitive materials and CNTs can lead to high-performance asymmetric supercapacitors.^[112] This hybrid strategy significantly enhances specific capacitance and boosts energy density, thereby rendering these materials highly promising candidates for next-generation energy storage applications. The design and synthesis of nanostructured carbon materials also play a vital role in optimizing the electrochemical properties of supercapacitors. Liu's review on boron carbon nitride nanomaterials highlights the importance of structural design, porosity, and defect engineering in significantly enhancing the specific capacitance and energy density of supercapacitors.^[113] Introducing heteroatoms into carbon structures has been shown to improve electrochemical characteristics further, as evidenced by Mateen et al.'s findings on nitrogen-functionalized carbon materials.^[114] The adjustments enhance ion diffusion and charge transfer, which are essential for attaining high-performance supercapacitors. The development of hierarchical and porous carbon nanostructures has significantly improved the electrochemical performance of supercapacitors. Zhou et al. demonstrated that hierarchically porous carbon materials derived from metal-organic frameworks (MOFs) exhibit remarkable electrochemical stability and enhanced capacitance.^[115] The unique morphology of these materials allows for increased SSA and improved electrolyte accessibility, which are essential for maximizing the charge storage capacity.

Incorporating sophisticated carbon nanostructures, including 3D graphene architectures, CNFs, ACs, and CNTs, has significantly enhanced the efficacy of carbon-based supercapacitors. The materials demonstrate remarkable characteristics, such as elevated SSA, improved electrical conductivity, and the capacity to create novel hybrid structures. This has substantially increased specific capacitance, energy density, and charging/discharging rates.^[116,117] Hydrothermal self-assembly, electrospinning, and chemical activation enable precise control over nanostructure morphology, porosity, and surface chemistry, ensuring tailored performance for targeted applications.^[118] Furthermore, incorporating heteroatoms into carbon matrices and using hierarchical designs that synergize the strengths of different carbon nanostructures have broadened the functional scope of these materials.^[119] Such advancements optimize the electrochemical properties of supercapacitor electrodes, positioning them as pivotal components in next-generation energy storage systems. The evolution of carbon nanostructure design is increasingly aligned with the principles of sustainability and scalability, thereby addressing the growing demand for environmentally friendly energy solutions. Biomass-derived carbons, sourced from renewable precursors such as agricultural waste, provide an eco-friendly alternative to traditional materials. Advanced fabrication methods, including roll-to-roll processing and 3D printing, facilitate

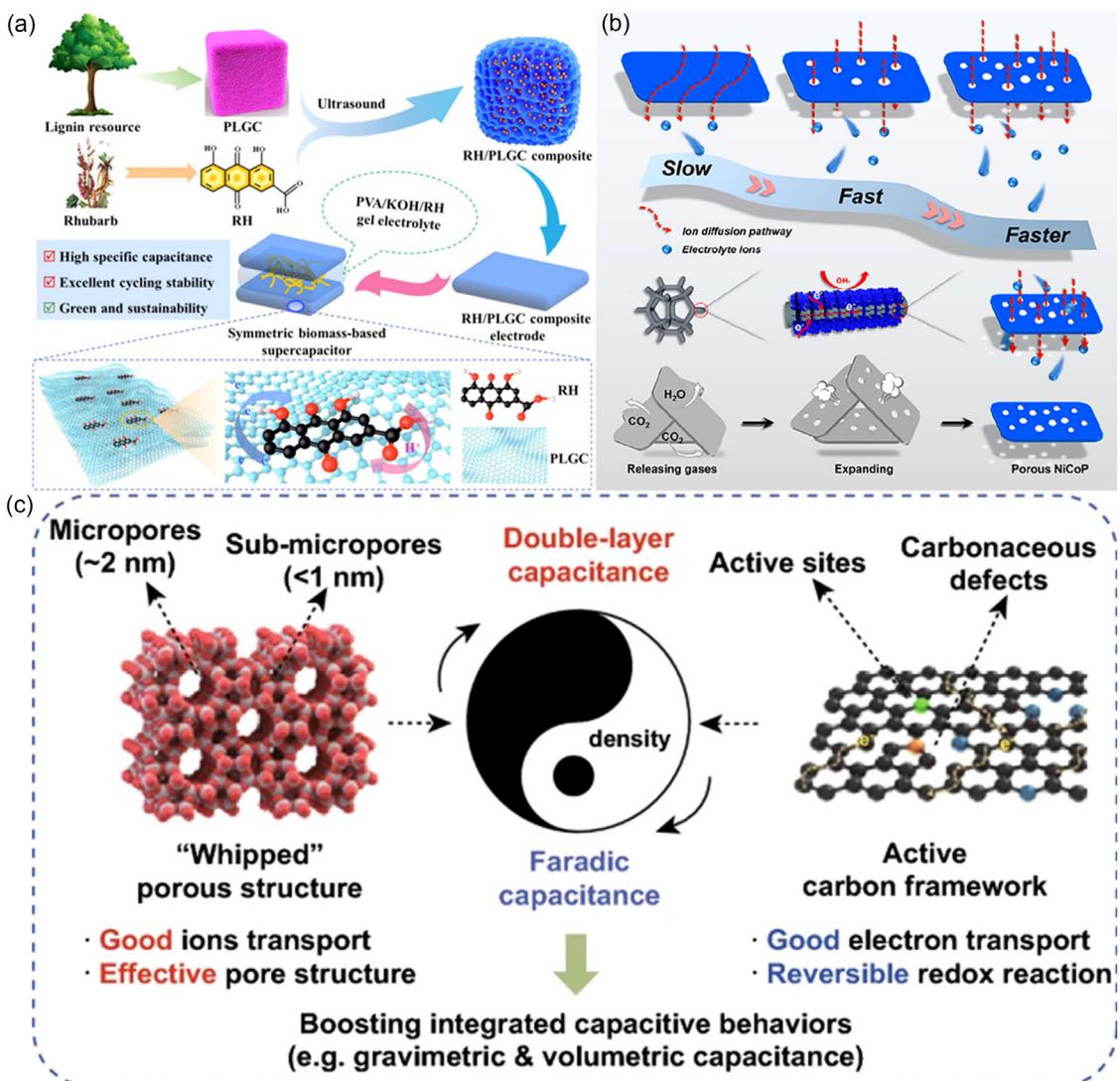


Figure 3. a) Schematic representation of a biomass-based supercapacitor using lignin-derived porous carbon and rhubarb extract for high-performance energy storage. Reproduced with permission.^[109] Copyright 2023, American Chemical Society. b) Illustration of the influence of pore distribution on electrolyte ion diffusion across the electrode surface and regulation of intrinsic pore structure formation. Reproduced with permission.^[133] Copyright 2022, American Chemical Society. c) Conceptual illustration of the dual contribution of microporous (whipped') and defect-rich carbon structures to double-layer and faradaic capacitances, respectively, promoting high-performance supercapacitors through improved ion diffusion, electron conduction, and reversible redox reactions. Reproduced with permission.^[142] Reproduced under the term CC BY 4.0. Copyright 2020, Ding et al., Springer Nature.

large-scale production.^[120–122] Modifying these materials through activation, doping, or templating has unlocked their potential for delivering enhanced electrochemical performance and versatility. Hybrid configurations, such as CNTs-graphene^[123] or AC-metal composites,^[124] have successfully mitigated challenges, further improving the cycling stability and practical applicability of carbon-based electrodes. These breakthroughs in material design, coupled with a focus on sustainable synthesis and market-ready production techniques, underscore the transformative role of advanced carbon nanostructures in shaping the future of energy storage technologies.

3D graphene, CNT scaffolds, carbon dots, and MOF-derived carbons each solve a different bottleneck. 3D graphene improves rate/cycling by preventing restacking; CNTs inject conductivity and toughness; carbon dots introduce surface redox (e.g., ≈290%

increase for ZnO/CDs);^[110] MOF-derived carbons offer high heteroatom content and porosity. However, these classes vary widely in cost, scalability, and even metric definitions (e.g., some reports use mAh g⁻¹ rather than F g⁻¹),^[115] complicating apples-to-apples comparisons. Biomass-derived hybrids (lignin + redox biomolecules: 250.2 F g⁻¹; 8.1 Wh kg⁻¹)^[109] show a pragmatic path toward sustainability but currently trail the highest-performance inorganic composites. A fair assessment, therefore, weighs not only peak values but also mass loading, electrode density, and manufacturability.

2.4. Impact on Ion Diffusion and Charge Storage

Nanostructure engineering in carbon-based supercapacitors is pivotal in optimizing ion diffusion, reducing diffusion distances, maximizing charge storage, and enhancing overall

electrochemical performance. The interplay of pore size, connectivity, and surface functionality defines ion dynamics, with hierarchically porous architectures emerging as a key strategy for improving efficiency by shortening ion diffusion pathways.^[125] Micropores primarily contribute to charge storage via electric double-layer formation, whereas mesopores and macropores alleviate ion diffusion resistance, enabling rapid ion transport even at high current densities.^[66] This hierarchical porosity, which integrates micropores for storage and meso-/macro-pores for diffusion, enables superior rate capabilities, energy density, and cycling stability, highlighting the critical role of tailored nanostructures in enhancing supercapacitor performance.

The high SSA of nanostructured carbon materials, including CNTs and graphene, presents a key benefit, enabling a greater number of electroactive sites for ion adsorption. The integration of CNTs/graphene composites has demonstrated a notable improvement in capacitance due to the synergistic interactions of these materials, facilitating enhanced ion diffusion and charge storage capabilities.^[100,126] The elevated SSA of these nanostructures facilitates enhanced electrochemical double-layer capacitance and pseudocapacitance mechanisms, which are crucial for attaining high energy and power densities in supercapacitors.^[127,128] Furthermore, the structural design of nanostructured carbon materials can be optimized to enhance ion transport pathways. For example, creating hierarchically porous structures within carbon electrodes can facilitate rapid ionic diffusion by providing interconnected channels that reduce resistance to ion movement.^[129,130] This is particularly important in supercapacitors, where the ion transport rate directly influences the charge and discharge rates. Studies have shown that materials possessing well-defined pore architectures exhibit improved ionic conductivity, which in turn contributes to enhanced capacitance and superior cycling stability.^[131,132] Zhu et al. synthesized intrinsically porous NiCoP using a hydrothermal method combined with annealing and phosphating, using NaHCO₃ as a pore-forming agent.^[133] The material demonstrated exceptional supercapacitor performance, delivering a specific capacitance of 2805 F g⁻¹ at 1 A g⁻¹, alongside a high energy density of 37.7 Wh kg⁻¹ and a power density of 8000 W kg⁻¹, as well as an impressive retention rate of 82.9% after 5000 cycles. The intrinsic porous structure significantly enhances ion diffusion, as evidenced by a diffusion coefficient of 1.79×10^{-10} m² s⁻¹ and a reduction in diffusion energy barriers. Figure 3b illustrates the simulated electrolyte concentration distributions across the NiCoP electrodes with varying pore area proportions, showing that an optimized pore distribution (50.01%) ensures efficient ion diffusion and electrode stability.

Heteroatom doping in carbon nanostructures, such as nitrogen or phosphorus, also contributes to enhanced electrochemical performance. Doping can modify the electronic properties of carbon materials, increase their conductivity, and create additional active sites for ion interactions.^[134,135] This modification enhances the overall capacitance and improves the stability of supercapacitors during cycling, as observed in studies involving nitrogen-doped carbon materials.^[136,137] Furthermore, integrating metal oxides with carbon nanostructures has been shown to enhance

capacitance through synergistic pseudocapacitive contributions. For instance, the hybridization of carbon with manganese oxide has been reported to significantly improve the charge storage capacity owing to the redox reactions occurring at the interface between carbon and the metal oxide.^[138,139] This hybrid approach leverages the high conductivity of carbon materials while utilizing the high theoretical capacitance of metal oxides, resulting in superior performance in supercapacitor applications.^[140,141] Figure 3c illustrates how the "whipped" microporous carbon architecture and the defect-rich active carbon framework synergistically deliver both double-layer and faradaic capacitances, thereby enhancing ion/electron transport and boosting gravimetric and volumetric energy-storage capabilities.^[142]

The engineering of nanostructures in carbon-based supercapacitors is pivotal for enhancing ion diffusion and charge storage efficiency. Key advancements include high-SSA materials, optimized pore structures, heteroatom doping, and hybridization with metal oxides, which synergistically improve capacitance and electrochemical performance. Functional surface groups such as hydroxyl, carboxyl, and amine groups enhance material wettability and facilitate pseudocapacitance through reversible redox processes.^[143] Heteroatom doping, incorporating elements such as N, S, or P, modifies the surface potential through localized charges, thereby promoting ionic adsorption and increasing the capacitance.^[144] Integrating 3D graphene networks and CNTs frameworks also establishes interconnected porous structures that enable effective ion transport and charge storage. These nanostructures exhibit high electrical conductivity, reduced ion diffusion distances, and compatibility with supplementary active materials, such as metal oxides or conducting polymers, further advancing the performance of supercapacitors. Electrochemical impedance spectroscopy (EIS) highlights the benefits of nanostructures in reducing ion transport resistance and improving kinetics, enabled by optimized pore size distributions and functionalized surfaces.^[145] This results in a lower charge transfer resistance, faster ion diffusion, improved power densities, and excellent cycling stability. Electrolyte properties, including ion size and conductivity, play a significant role in ion dynamics, with aqueous and IL electrolytes demonstrating distinct diffusion behaviors in micropores and mesopores. These approaches underscore the importance of tailoring pore designs, surface functionalities, and material properties, paving the way for next-generation supercapacitors with enhanced energy-storage capabilities.

Ion transport benefits when micropores (storage) are percolated by meso/macropore highways (delivery), as seen in low Warburg slopes and reduced charge-transfer resistance in EIS. Architectures with intrinsic porosity (e.g., NiCoP: 2805 F g⁻¹; 37.7 Wh kg⁻¹; $D \approx 1.79 \times 10^{-10}$ m² s⁻¹; 82.9% after 5000 cycles)^[133] showcase how diffusion barriers can be engineered down; yet extremely high gravimetric values often reflect thin, low-loading electrodes and can degrade at device-relevant areal loadings. Heteroatoms enhance quantum capacitance and wettability but may introduce localized strain that accelerates aging if not buffered by mesoporous frameworks. Hybridizing carbons with thin, conformal oxides/polymers harnesses fast Faradaic

storage, whereas thick coatings reintroduce diffusion bottlenecks; the most robust designs cap the pseudocapacitive layer thickness to maintain short paths.

Seminal surveys have typically treated each enhancement strategy for carbon supercapacitors in isolation, including nanostructure engineering,^[146] heteroatom doping,^[147] composite formation with pseudocapacitive and battery materials,^[148,149] and electrolyte choices.^[150] This siloed treatment makes it difficult to translate lab advances into coherent design rules for devices that must simultaneously maximize energy density, power density, and cycling stability. This review advances the literature by organizing these strategies into a cohesive, interaction-aware framework. We map how 1) pore size hierarchy governs ion accessibility and desolvation barriers, 2) heteroatoms tune electronic structure, quantum capacitance, and surface polarity to activate redox sites and improve wettability, and 3) pseudocapacitive phases contribute fast Faradaic storage when electronically percolated through carbon scaffolds. We emphasize cross-couplings that are usually discussed separately: for example, N/S/P doping not only introduces redox sites but also lowers interfacial resistance in oxide/polymer hybrids; meso/macroporous carbon backbones shorten diffusion paths and mitigate volumetric changes of transition metal oxides (TMOs) and conducting polymers; and pore size distributions must be matched to electrolyte ion size and solvation state to unlock wider voltage windows. A distinctive contribution of this review is the explicit, comparative treatment of the electrolyte/carbon interface. We link electrode architecture to electrolyte choices, aqueous,^[151] organic,^[152] ILs,^[153] water-in-salt systems,^[42] microemulsions,^[41] and gel polymer/solid electrolytes for flexible formats,^[154,155] and show how ion size, viscosity, and electrochemical stability window couple to subnanometer pores, heteroatom-induced polarity, and MXene/MOF/TMD composites. This interface-centric view clarifies why similar carbons can exhibit vastly different performance across electrolytes, providing practical guidelines for voltage expansion and equivalent series resistance reduction. We combined advances that earlier reviews did not cover comprehensively, including machine-learning-assisted pore/dopant optimization for O, N-codoped HOPCs,^[156] intrinsically porous phosphides with quantified diffusion metrics;^[133] ternary heterostructures (e.g., YSe₂/MoSe₂/Ni_xB) with enhanced stability;^[59] quaternary (N/P/O/S) codoping strategies;^[157] bio-derived redox electrolytes and microemulsion media that widen practical voltage while sustaining conductivity;^[41,158] and water-in-salt electrolytes that enable ≈ 3 V operation in aqueous systems.^[42] By analyzing these developments within a single framework, we drive design guidelines that cut across materials and electrolytes. In this review, we provide interaction-aware design heuristics: 1) co-optimize micropore volume for EDL storage with mesopore channels for ion transport; 2) select heteroatoms to balance conductivity (graphitic-N) and redox functionality (pyridinic/pyrrolic-N, P/S sites); 3) embed pseudocapacitive phases via conformal, nanoscale contacts to avoid kinetic bottlenecks; 4) choose electrolytes by ion size/pore size/electrochemical stability window (ESW) matching, using ILs when voltage, not only rate, limits energy density; and 5) apply

interfacial adhesion/functionalization to stabilize composites under long cycling. These cross-cutting rules, supported by the comparative tables and case studies, delineate where synergistic gains are most likely and where trade-offs remain.

3. Heteroatom-Doped Carbon

Heteroatom incorporation represents a transformative strategy for enhancing the electrochemical performance of carbon-based supercapacitors by precisely tailoring the chemical, electronic, and structural properties of electrode materials. The integration of elements such as nitrogen, boron, sulfur, and phosphorus into the carbon matrix enhances electrical conductivity, promotes pseudocapacitive effects, and increases surface wettability, thereby boosting both the electric double-layer capacitance and pseudocapacitance.^[159] Nitrogen doping, in particular, significantly improves the charge carrier density, redox activity, and wettability, while other dopants, such as boron, sulfur, and phosphorus, induce unique modifications, including band structure alterations, redox-active functionalities, and enhanced charge storage.^[160] Codoping strategies involving multiple heteroatoms have demonstrated synergistic effects, yielding superior electrochemical performance by increasing the density of active sites and optimizing charge storage mechanisms; for instance, combinations such as nitrogen-sulfur enhance conductivity and wettability, while nitrogen-phosphorus pairs boost energy and power densities.^[161] Schottky junctions in heteroatom-doped carbon enhance supercapacitor performance by facilitating rapid ion diffusion during charging and maintaining controlled discharge; however, their efficiency may depend on the material's junction properties and the uniformity of doping. Figure 4 illustrates how Schottky junctions in heteroatom-doped carbon control charge storage through fast charging (ion diffusion from semiconductor to metal) and slow discharging (ion diffusion from metal to semiconductor), thereby enhancing the performance of the supercapacitor.^[162] Advanced synthesis methods, such as chemical vapor deposition, hydrothermal techniques, and plasma-induced doping, enable precise control over dopant type, concentration, and placement. This allows for the strategic positioning of heteroatoms at edge sites or critical lattice areas, further enhancing material properties. Both theoretical and experimental studies on dopant-induced modifications in electronic structure and electrochemical behavior have guided the rational design of advanced electrode materials with improved energy and power densities. As a result, heteroatom doping has emerged as a fundamental strategy in the development of next-generation energy storage technologies.^[163]

Heteroatom-doped carbon materials have emerged as transformative electrode materials for supercapacitors, offering enhanced electrochemical performance through tailored electronic and structural modifications. Nitrogen-doped porous carbons exhibit specific capacitance up to 754.4 F g⁻¹ due to redox-active pyridinic and pyrrolic-N groups.^[163,164] Sulfur doping

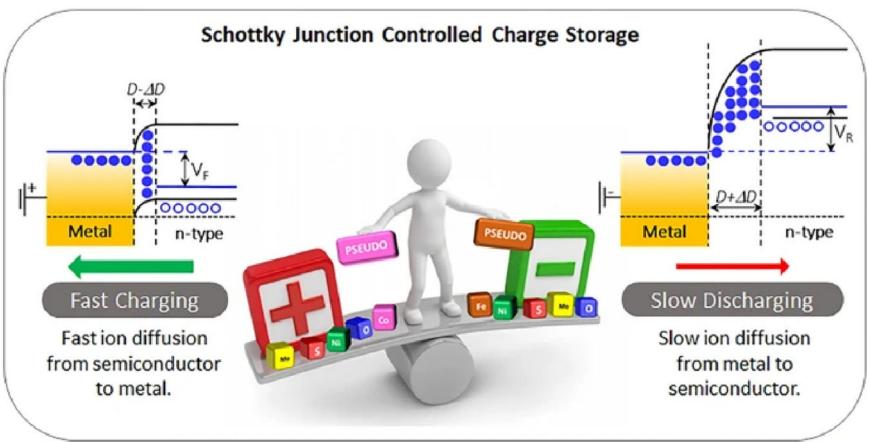


Figure 4. Schematic representation of Schottky junction-driven ion diffusion for fast charging and slow discharging using various heteroatoms. Reproduced with permission.^[162] Reproduced under the term CC BY 4.0. Copyright 2023, Seenivasan et al., Springer Nature.

induces electron-rich surfaces, thereby improving the dielectric constant and charge transfer kinetics, while phosphorus enhances pseudocapacitance through Faradaic reactions.^[52,121,165–167] N-doping reduces charge transfer resistance by 30–50% compared to undoped carbons.^[168] Oxygen functional groups (e.g., quinones) enhance wettability, increasing electrolyte accessibility.^[163] Quaternary-doped (N/P/O/S) carbons demonstrate 86% capacitance retention after 10,000 cycles due to synergistic effects.^[157] Hierarchical pore structures (micro-/mesopores) in polybenzoxazine-derived carbons achieve $1,849 \text{ m}^2 \text{ g}^{-1}$ SSA, enabling rapid ion diffusion (324 F g^{-1} at 1 A g^{-1}). Hollow carbon spheres show 241 F g^{-1} capacitance from shortened ion pathways.^[169]

While previous literature has primarily summarized the role of heteroatom doping,^[32,163,170] this review significantly differentiates itself by systematically discussing the comparative and synergistic impacts of multiheteroatom doping (codoping), clearly highlighting their complementary electrochemical and structural benefits. We further differentiate our review by integrating recent progress from computational insights and experimental validations, offering readers clear connections between doping strategies, specific heteroatom functionalities, and resultant improvements in supercapacitor performance. This integrative and comparative approach offers readers unprecedented clarity on optimizing heteroatom doping strategies. Among single dopants, N provides the most balanced gain (conductivity via graphitic-N; redox via pyridinic/pyrrolic-N), routinely lifting capacitance into the $300\text{--}700 \text{ F g}^{-1}$ range with strong retention ($\geq 94\%$ after 10,000 cycles in several cases).^[171] S and P introduce electron-rich sites and widen OPW or interlayer spacing but can destabilize the framework if concentrations are high. Codoping (e.g., N/S, N/P) achieves superior metrics (quaternary N/P/O/S: 815 F g^{-1} ; 86% after 10,000 cycles)^[157] through complementary roles, albeit at the cost of more complex synthesis and tighter process windows. The practical ceiling is set by dopant level vs. graphitization: beyond an optimal range, ESR rises and cycling suffers. Best performance arises when moderate dopant levels are embedded in hierarchically porous backbones and paired with electrolytes that exploit the induced polarity.

3.1. Mechanisms of Capacitance Enhancement Through Doping

The process of heteroatom doping plays a crucial role in improving the capacitance of carbon-based supercapacitors. This approach effectively addresses the fundamental limitations of conventional carbon materials by modifying the electronic structure and surface chemistry of the carbon framework. Elements such as nitrogen,^[20] oxygen,^[172] sulfur,^[167] and phosphorus,^[173] with their distinct electronegativity and atomic size differences relative to carbon, introduce substantial changes in the material properties, creating additional active sites for redox reactions and enabling pseudocapacitance through reversible faradaic processes. This pseudocapacitive mechanism, which complements the electric double-layer capacitance, significantly expands the overall energy storage capabilities and electrochemical properties of the electrodes, thereby making heteroatom doping an essential approach in the design of high-performance supercapacitor materials.^[163,174,175] Figure 5a illustrates the structural and electrochemical advantages of heteroatom doping (e.g., B, N, O, P, and S) in carbon-based materials, which enhances ion adsorption and charge storage in supercapacitors.^[176]

The introduction of pseudocapacitance is one of the key mechanisms through which heteroatom doping enhances capacitance. This phenomenon occurs when the introduced heteroatoms participate in redox reactions during the charge-discharge cycles. This leads to enhanced charge storage that exceeds the electric double-layer capacitance commonly associated with carbon materials. Nitrogen doping has notably enhanced the specific capacitance of carbon materials by promoting Faradaic reactions, thereby improving the overall charge storage capacity of the electrodes.^[135,177] Nitrogen enhances the wettability of the carbon surface, facilitating improved electrolyte penetration and ion transport, which is essential for achieving high-performance supercapacitors.^[178] Furthermore, the addition of heteroatoms can potentially enhance the electrical conductivity of carbon materials. This holds significant importance as increased conductivity diminishes the charge transfer resistance

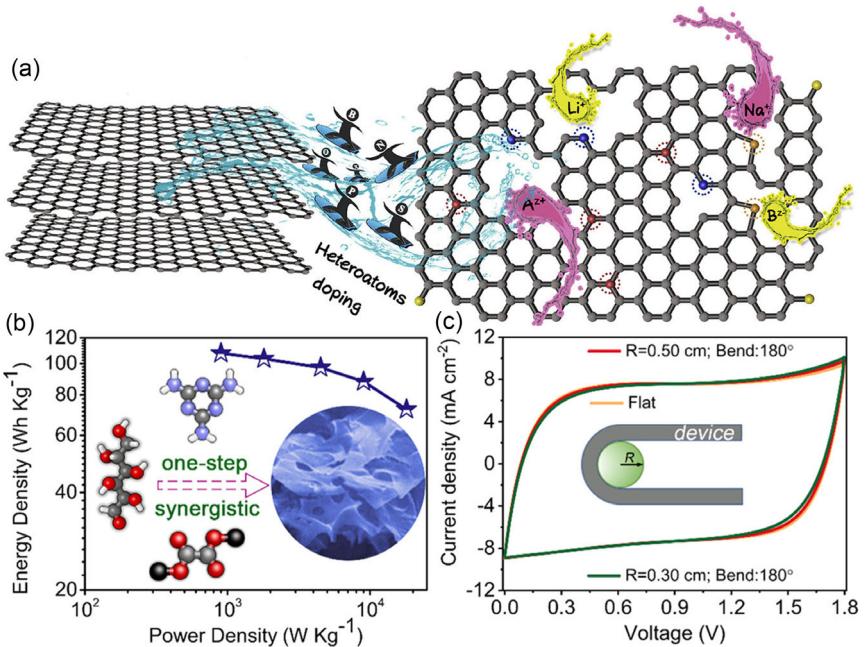


Figure 5. a) Schematic representation of heteroatom doping in carbon-boosting ion adsorption. Reproduced with permission.^[176] Copyright 2020, Elsevier. b) Synergistic porous carbon synthesis along with Ragone plot and c) flexible device performance under bending with CV curves. Reproduced with permission.^[38] Copyright 2020, American Chemical Society.

at the electrode–electrolyte interface, facilitating quicker charge and discharge rates. Research has shown that nitrogen-doped CNTs display superior electrical conductivity and better electrochemical performance when compared to their undoped versions.^[177,179] Furthermore, incorporating functional groups linked to heteroatoms can generate additional active sites for ion adsorption, thereby improving the capacitance.^[84,178] The structural features of the carbon matrix significantly influence the enhancement of capacitance. Incorporating heteroatoms can dramatically affect the porosity and SSA of carbon materials, which are essential in assessing the electrochemical performance of supercapacitors. The presence of oxygenated functional groups enhances surface wettability and promotes ion transport while also playing a role in increasing the overall SSA available for charge storage.^[84,178] The optimization of pore size and distribution via doping strategies has demonstrated an enhancement in SSA, which is directly linked to the capacitance of the supercapacitor.^[180] Incorporating doping in carbon materials leads to a notable enhancement in their performance within supercapacitors, resulting in improved specific capacitance, energy density, power density, and stability.^[181] Nitrogen-doped carbon materials have exhibited specific capacitances ranging from 306.5 to 660.1 F g⁻¹ across various current densities.^[37] The energy densities achieved can be as high as 108 Wh kg⁻¹ at a power density of 900 W kg⁻¹,^[38] while other studies report energy densities of 20.7 Wh kg⁻¹ at 300 W kg⁻¹,^[182] and 51.8 Wh kg⁻¹ at higher power densities.^[183] Figure 5b illustrates the synergistic one-step synthesis method, yielding a porous carbon material with high energy density. Figure 5c demonstrates the exceptional flexibility of the device, maintaining a consistent current density under various

bending conditions. Stability is also critical; for example, nitrogen-doped carbon electrodes retain over 94% of their initial capacitance after 10,000 cycles.^[171]

Heteroatom doping-induced structural and electronic changes, such as defect generation and alterations in the density of states at the Fermi level, improve electron conductivity, accelerate charge–discharge cycles, and boost energy and power density.^[52,121,166] Nitrogen doping, for instance, elevates the Fermi level through n-type effects,^[184] while functional groups such as pyridine and pyrrole participate in reversible redox reactions,^[185] contributing significantly to pseudocapacitance. Enhanced wettability and the introduction of redox-active sites further optimize the electrode–electrolyte interface, particularly in aqueous environments. Moreover, quantum capacitance benefits from heteroatom-induced changes in subnanometer pores, enhancing charge storage at the nanoscale.^[25] However, the performance of doped materials depends critically on the dopant species, concentration, and synthesis conditions, as excessive doping can compromise structural integrity. The interplay of these mechanisms yields synergistic improvements in energy and power densities, underscoring the importance of balanced material design in advancing next-generation supercapacitors with optimal performance metrics.

Doping adds pseudocapacitance on top of EDLC and lowers R_{ct} , but it also perturbs lattice order. Reports here span ≈ 306 – 660 F g⁻¹ for N-doped carbons,^[37] energy densities up to 108 Wh kg⁻¹,^[38] and $\geq 94\%$ retention at $10,000$ cycles,^[171] demonstrating clear benefits when redox-active sites (pyridinic/pyrrolic-N, P/S groups) coexist with adequate conductivity (graphitic-N). Overdoping, however, narrows pore necks, raises tortuosity, and can catalyze parasitic reactions, particularly in

high-voltage electrolytes. The most reliable gains are achieved by positioning dopants at edge- or defect-rich mesopores to maximize accessibility and by pairing doped carbons with electrolytes whose ion size/solvation complement the subnanometer pores where quantum capacitance is largest.

3.2. Methods and Effects of Heteroatom Doping

Introducing heteroatoms into carbon materials for supercapacitors involves incorporating elements such as nitrogen, oxygen, sulfur, or phosphorus into the carbon framework. This modification aims to improve electrical conductivity, surface wettability, and the availability of active sites for charge storage. This approach enhances the electrochemical performance of supercapacitors by elevating their specific capacitance, energy density, and cycling stability. Nitrogen-doped carbon materials have gained significant attention as promising electrode candidates for supercapacitors, primarily due to their capacity to improve electrochemical performance and energy storage efficiency markedly.^[186] Integrating nitrogen into the carbon framework leads to the formation of diverse functional groups, including pyridinic and pyrrolic nitrogen, which act as active sites for Faradaic reactions, thereby enhancing the specific capacitance of the electrodes.^[187–189] Studies have shown that nitrogen doping increases the surface wettability and conductivity of carbon materials, enhancing their interaction with electrolytes and facilitating improved charge storage capabilities.^[171,190,191] Furthermore, the hierarchical porous structure of nitrogen-doped carbons contributes to their high SSA, which is crucial for accommodating more electrolyte ions and enhancing the capacitance.^[74,192] Recent advancements in synthetic methods, including the use of biomass and waste materials, have led to the development of nitrogen-doped porous carbons with excellent electrochemical properties, demonstrating their potential as sustainable energy-storage solutions.^[164,193] Overall, nitrogen doping is a vital strategy for optimizing the performance of carbon-based supercapacitors, making them suitable for various applications in energy storage technologies.

Heteroatom doping, particularly with boron, sulfur, and phosphorus, enhances the electrochemical properties of carbon-based supercapacitors by improving the conductivity, wettability, pseudo-capacitance, and ion transport through tailored chemical and structural modifications. Doping carbon-based materials with boron, sulfur, and phosphorus significantly enhances the performance of supercapacitors. Boron doping enhances the electronic conductivity and electrochemical activity of carbon materials, resulting in increased capacitance and energy density.^[96,194] The introduction of boron creates localized defects that facilitate charge transport, thereby enhancing the overall electrochemical performance.^[195,196] On the other hand, sulfur doping enhances the wettability and conductivity of carbon electrodes, which is crucial for efficient ion transport during charge/discharge cycles.^[121,197] The larger atomic size of sulfur than carbon induces strain in the carbon matrix, which can improve the capacitive behavior through Faradaic processes.^[198] Phosphorus doping is known to widen the OPW of carbon electrodes, thereby

increasing energy density.^[199,200] Phosphorus can block active oxidation sites, further enhancing the stability and performance of supercapacitors. Collectively, these heteroatom dopants synergistically enhance the electrochemical properties of carbon-based supercapacitors, making them promising candidates for energy storage applications.^[84,201] Figure 6a highlights the critical advantages of nitrogen-doped carbon materials for supercapacitors, emphasizing their high specific capacitance, energy and power density, cycling stability, and the underlying contributions of the nitrogen-doping methods, precursors, and physicochemical structures.^[189]

The simultaneous introduction of various heteroatoms, including nitrogen-sulfur, nitrogen-boron, or nitrogen-phosphorus, significantly improves the electrochemical performance of carbon-based supercapacitors by improving their conductivity, active site density, structural stability, and energy storage capacity through complementary chemical and electronic interactions. Implementing codoping strategies in carbon materials, particularly with heteroatoms such as nitrogen and sulfur, has shown a notable improvement in the electrochemical performance of carbon-based supercapacitors. Incorporating various heteroatoms can significantly enhance specific capacitance and rate capability by augmenting the number of active sites and improving the wettability of the carbon surface, thereby facilitating ion transport.^[202–204] For instance, nitrogen and sulfur-codoped porous carbons have demonstrated superior electrochemical properties owing to the combined effects of pseudocapacitance and improved conductivity.^[197,203,204] Moreover, the structural characteristics of carbon materials, such as hierarchical porosity and SSA, play a crucial role in their performance. An enhanced SSA enables more significant charge accumulation and faster ion diffusion, which is essential to high-performance supercapacitors.^[205–207] Recent studies indicate that optimizing doping levels and porous structures can significantly enhance energy density and cycling stability.^[171,208,209] Consequently, codoping strategies provide a compelling pathway for advancing carbon-based materials tailored for energy storage applications. Figure 6b shows various natural precursors, including plant, animal, and microorganism sources, for the synthesis of heteroatom-doped carbon materials, highlighting different doping strategies (N, S, P, codoped, and triple-doped) to enhance supercapacitor performance.^[210]

Introducing heteroatoms into carbon-based supercapacitors significantly enhances their electrochemical performance, particularly in terms of specific capacitance, energy density, power density, and stability. Nitrogen-doped porous carbon has exhibited specific capacitance values as high as 379 F g^{-1} ^[211] while phosphorus-doped variants have reached 524.9 F g^{-1} .^[212] The energy density of these devices can vary significantly, with some configurations achieving up to 18.9 Wh kg^{-1} .^[211] and others around 7.8 Wh kg^{-1} .^[177] Power densities also exhibit remarkable performance, with values reaching 6.2 kW kg^{-1} .^[177] and even $4295.28 \text{ W kg}^{-1}$ in specific cases.^[213] Stability is another critical factor; supercapacitors have demonstrated over 99.5% capacitance retention after 10,000 cycles.^[177] The integration of heteroatoms into the carbon matrix enhances both electrical

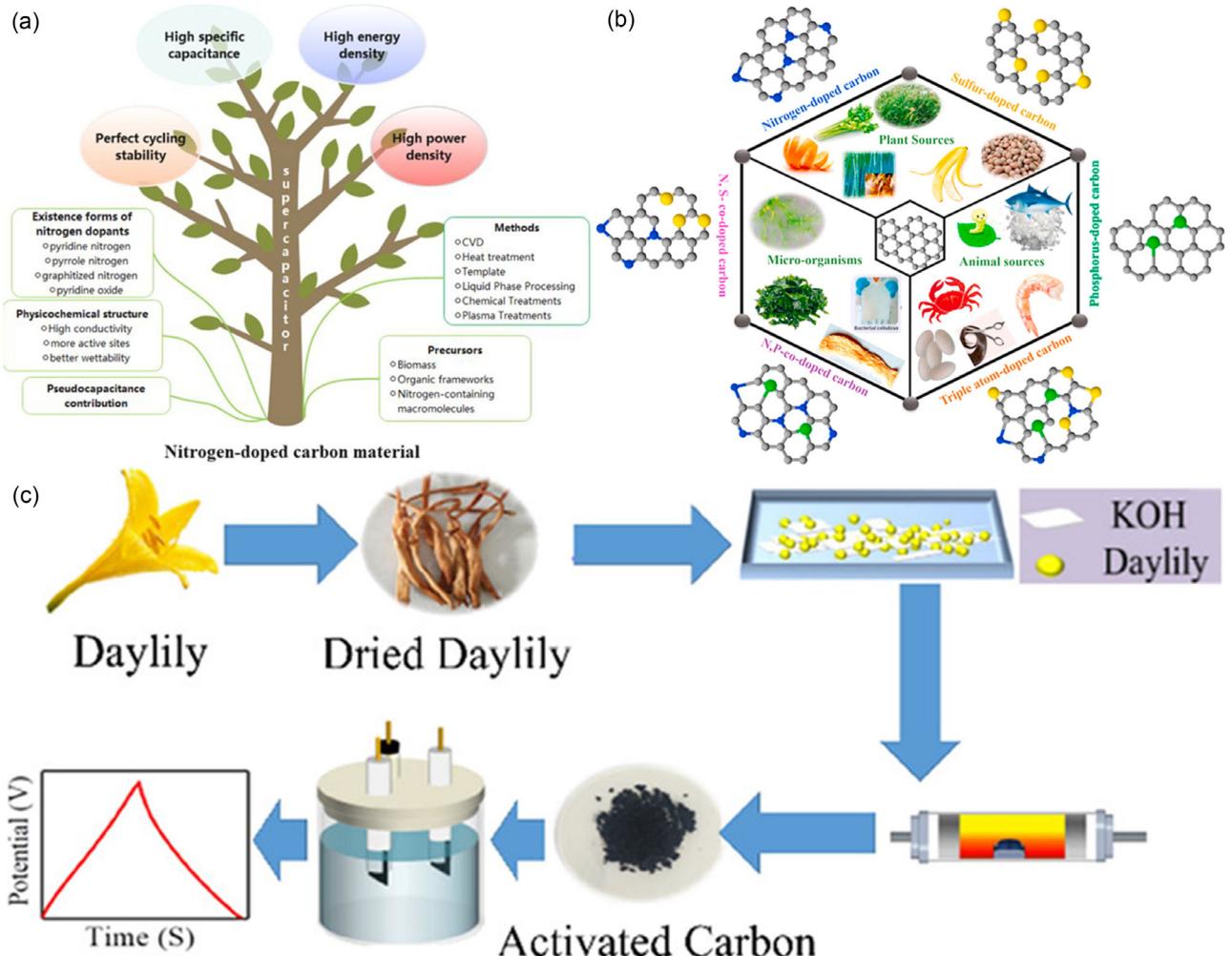


Figure 6. a) Advantages and key features of N-doped carbon materials for supercapacitors. Reproduced with permission.^[189] Copyright 2024, American Chemical Society. b) Schematic representation of natural precursors and doping strategies for heteroatom-doped carbon materials. Reproduced with permission.^[210] Copyright 2020, Elsevier. c) Synthesis of AC from daylily for energy storage. Reproduced with permission.^[214] Reproduced under the term CC BY 4.0. Copyright 2022, Jin et al., American Chemical Society.

conductivity and surface wettability, thereby facilitating faster ion transport and promoting improved electrochemical kinetics.^[214] This synergistic effect of multiple heteroatom doping is essential for optimizing the performance of carbon-based supercapacitors. Figure 6c illustrates the sustainable synthesis of AC from daylily biomass through a drying and KOH activation process, showing its application in energy storage devices with excellent electrochemical performance.^[214]

Heteroatom doping of carbon materials, particularly with nitrogen, phosphorus, boron, and sulfur, significantly enhances supercapacitor performance by modifying the electronic structure of redox-active sites, wettability, and availability. Nitrogen doping, which has been extensively explored, provides enhanced electrical conductivity (primarily via graphitic-N), pseudocapacitance from pyridinic and pyrrolic groups, and improved electrolyte accessibility due to increased surface polarity. Various doping methods, ranging from in situ pyrolysis with nitrogen-rich precursors (e.g., urea and melamine) to post-treatment with ammonia or

plasma, offer flexible control over doping levels, bonding states, and pore architectures. Similarly, phosphorus, boron, and sulfur doping can enlarge the interlayer spacing, create defects, and introduce additional functional groups that enhance the active site density, redox behavior, and overall electrochemical stability. These tailored modifications highlight the importance of carefully adjusting the dopant types and concentrations to balance conductivity, structural integrity, and capacitance. Beyond single-element doping, codoping strategies involving multiple heteroatoms (such as N-S, N-B, and N-P) offer synergistic improvements in energy and power densities. These synergisms arise from the complementary roles of each dopant; for instance, nitrogen enhances conductivity and redox activity, while sulfur introduces defects and additional active sites, or boron serves as an electron acceptor to balance the electron-donating effect of nitrogen. Advanced synthesis routes, such as chemical vapor deposition, solvothermal methods, and plasma-assisted treatments, facilitate precise control over dopant distribution and

bonding configurations, enabling hierarchical pore development and uniform defect formation. Consequently, codoped carbon materials often exhibit high specific capacitance, superior rate performance, and excellent cycling stability, making them promising candidates for next-generation supercapacitor applications.

Process choice governs the dopant type, bonding state, and pore coevolution. In situ pyrolysis with N-rich precursors is scalable and yields stable N-functionalities (N-porous carbons: 379 F g^{-1});^[211] P-doping can push higher ($\approx 524.9 \text{ F g}^{-1}$)^[212] but requires careful control to avoid brittleness. Codoped systems frequently outperform single-dopant analogs (e.g., N/S codoped or quaternary carbons) with strong stability ($\geq 99.5\%$ after 10,000 cycles in selected systems)^[157,177] and high power (kW kg^{-1} class^[174,212]).^[177,213] The trade-offs are synthetic complexity and variability in areal loading/packing density, which can inflate gravimetric figures. Standardizing reports (mass loading, density, OPW, ESR, and iR-drop corrections) is essential for fair comparison and scale-up.

4. Composite Formation with Pseudocapacitive Materials and Synergistic Strategies

A promising strategy to boost supercapacitor performance involves creating composite electrodes that integrate carbon-based matrices with pseudocapacitive materials, thus leveraging the strengths of each component. The carbon framework, often composed of graphene, CNTs, or porous carbons, functions as a conductive scaffold and provides structural integrity, whereas pseudocapacitive elements (e.g., metal oxides such as MnO_2 , RuO_2 , NiCo_2O_4 , conducting polymers such as PANI or polypyrrole (PPy), and emerging 2D materials including TMDs, MOFs, and MXenes) offer redox-active charge storage.^[14,215–220] Short charge-transport paths and uniform dispersion are achieved by ensuring intimate contact between these two phases through techniques such as in situ synthesis, electrodeposition, or controlled physical blending, facilitating swift faradaic reactions and efficient ion movement.^[215,221,222] This synergy addresses the low energy density characteristic of EDLC and the limited conductivity commonly associated with pseudocapacitive materials. Consequently, composite supercapacitors can deliver superior specific capacitance, higher energy and power densities, and robust cycling stability, positioning them as a vital class of next-generation energy storage solutions. Figure 7a illustrates the synergistic strategy of O, N codoped HOPCs derived via in situ exfoliation and pore-forming processes, emphasizing the dual functional effects, experimental validation, DFT calculations, and machine-learning-based interpretability for optimizing supercapacitor performance.^[156]

Carbon and metal oxide composites play a pivotal role in enhancing the performance of supercapacitors. The integration of carbon materials, known for their high electrical conductivity and SSA, with metal oxides, which provide pseudocapacitance, results in synergistic effects that significantly improve energy

storage capabilities. For instance, carbon components enhance the rate capability and power density of supercapacitors, whereas metal oxides contribute to high specific capacitance and energy density.^[223–225] Recent studies have demonstrated that composites, such as manganese oxide/carbon and nickel oxide/carbon, exhibit improved cycling stability and charge/discharge efficiency compared to their components.^[226,227] Additionally, incorporating conducting polymers into these composites further mitigates the limitations of metal oxides, such as their low conductivity and cycling stability.^[223,228] The development of nanostructured composites, including those derived from MOFs, has shown promising results in optimizing the electrochemical performance of supercapacitors.^[229,230] The deliberate integration of carbon materials with metal oxides in composite structures plays a pivotal role in advancing supercapacitor technology, effectively addressing key challenges associated with energy density, power output, and long-term cycling stability. Figure 7b illustrates the integration of carbon nanostructures with pseudocapacitive materials, including TMDs, MXene, conductive polymers, and MOFs/COFs, utilizing synergistic strategies to improve the supercapacitors' electrochemical performance.^[231] This composite formation enables exceptional energy and power densities, long cycling stability (up to 40,000 cycles), and high specific capacitance, making it ideal for next-generation energy-storage devices.

Carbon/conducting polymer hybrids synergistically integrate the excellent electrical conductivity and structural integrity of carbon materials with the high pseudocapacitive properties of polymers such as PANI and PPy. This combination leads to enhanced supercapacitor performance by facilitating efficient charge transport, mitigating polymer aggregation, and ensuring stable cycling. These improvements are attributed to strong interfacial interactions, such as π - π stacking, and uniform polymer deposition on the conductive carbon framework. Carbon-conducting polymer hybrids have emerged as promising materials for supercapacitor applications owing to their enhanced electrical conductivity and electrochemical performance. The integration of conductive polymers, such as PPy and PANI, with carbon-based materials, including AC and CNTs, has significantly improved the capacitance and stability of supercapacitors.^[232,233] For instance, the synergistic effect of combining MnCo_2O_4 with PPy and AC yields advanced electrode materials that demonstrate significantly enhanced energy storage performance.^[233] Furthermore, the advancement of hybrid nanocomposites, including those that integrate reduced graphene oxide (rGO) and metal oxides, has significantly improved the electrochemical performance of supercapacitors.^[234,235] The conductive pathways formed by these hybrids enable efficient charge transport, resulting in enhanced capacitance and cycling stability.^[236] Therefore, the rational design of carbon-conducting polymer hybrids is vital for the progression of supercapacitor technology, as it facilitates the achievement of higher energy densities and accelerated charge-discharge rates.

TMDs, MOFs, and MXenes, when combined with carbon-based materials, leverage their distinctive 2D structures, adjustable surface chemistries, and redox-active sites to yield high-performance supercapacitor composites characterized by efficient charge transport,

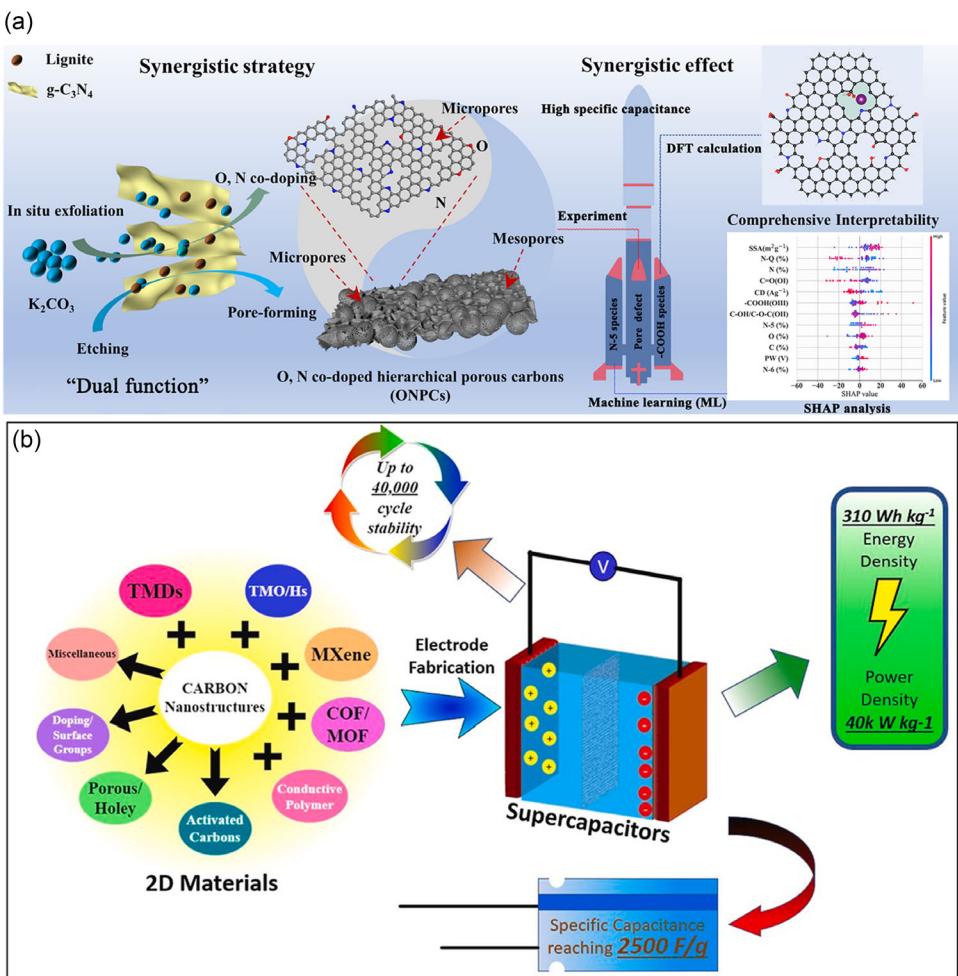


Figure 7. a) Synergistic strategy for O, N codoped porous carbons with experimental and machine learning insights. Reproduced with permission.^[156] Reproduced under the term CC BY 4.0. Copyright 2025, Liu et al., OAE Publishing Inc. b) Schematic representation of the composite formation of carbon nanostructures with pseudocapacitive materials for enhanced supercapacitor performance. Reproduced with permission.^[231] Reproduced under the term CC BY 4.0. Copyright 2023, Pathaare et al. Elsevier.

enhanced ion accessibility, and robust structural stability. MXenes and MOFs, characterized by their high electrical conductivity and significant SSA, can effectively reduce the charge–transfer resistance when combined with carbon materials, improving electrochemical conductivity and stability.^[237–240] The synergistic interactions among these materials promote the formation of hybrid architectures that significantly enhance energy storage performance by increasing the density of electrochemically active sites and optimizing ion transport pathways.^[241,242] Furthermore, integrating carbon materials, such as CNTs and graphene, into these composites significantly improves the overall electrochemical performance by providing additional conductive pathways and enhancing structural stability.^[243,244] Recent investigations have demonstrated that integrating MXenes with TMDs and MOFs significantly enhances both capacitance and energy density, positioning these composite materials as strong contenders for next-generation energy storage systems.^[232,245,246] This strategic combination of advanced material classes presents a compelling pathway toward the development of high-performance supercapacitors.

Carbon-based materials, including AC, graphene, and CNTs, have emerged as essential components for next-generation supercapacitors owing to their high electrical conductivity, structural integrity, and extensive SSAs for ion adsorption. However, hybridization with pseudocapacitive materials is crucial to boosting the energy density beyond that attainable by pure carbon (EDLC). Metal oxides (e.g., MnO₂, TiO₂, Co₃O₄), conducting polymers (e.g., PANI and PPy), and emerging materials, such as TMDs, MOFs, and MXenes, all introduce faradaic redox processes that impart higher specific capacitances. The carbon framework contributes to fast electron transport and mechanical resilience in these hybrids, while the active phases deliver substantial pseudocapacitance. By tuning synthesis parameters, optimizing material morphologies (e.g., nanowires, nanoflakes, hollow spheres), and preventing particle agglomeration, researchers can synergistically combine fast charge–discharge kinetics with elevated energy storage, thereby bridging the performance gap between traditional EDLCs and battery-type systems. In parallel, the long-term cycle stability of pseudocapacitive materials remains a critical challenge because

issues such as volume expansion, layer restacking (in TMDs and MXenes), and mechanical degradation of conducting polymers can compromise device lifetimes. Strategies to address these issues include designing nanostructured metal oxides, functionalizing carbon surfaces to improve interfacial adhesion, exploiting hierarchical core–shell or layered configurations, and carefully balancing the proportion of carbon to pseudocapacitive components. Ternary hybrids that combine carbon, metal oxides, and conducting polymers or other 2D materials (such as MXenes and TMDs) show significant promise by combining multiple redox-active phases with a robust conductive matrix, thereby improving both the power and energy characteristics. Ongoing developments in advanced synthesis methods, interface engineering, and computational modeling are expected to optimize these hybrid architectures further, laying the groundwork for future high-performance, durable supercapacitor technologies. Supercapacitors utilizing carbon and pseudocapacitive materials composites have exhibited significant advancements in their specific capacitance, energy density, power density, and stability. For instance, composites of rGO and carbon microspheres have demonstrated a specific capacitance of 403 F g^{-1} , while hybrid structures combining AC and TMOs can achieve energy densities around 29.2 Wh kg^{-1} and power densities of 8000 W kg^{-1} .^[247] Incorporating conductive polymers, such as PPy, significantly enhances the specific capacitance, often yielding values 3–7 times greater than those of pure carbon materials.^[248] Stability is also critical; for example, specific composites maintain over 100% of their initial capacitance after 4000 cycles, indicating exceptional cycling stability.^[249] These properties underscore the potential of carbon-pseudocapacitive composites in energy storage applications, making them a focal point of current research in supercapacitor technology.

Recent advancements in carbon-based supercapacitors emphasize multipronged strategies to achieve high energy and power densities, as well as excellent cycling stability. By integrating nanostructure engineering, heteroatom doping, and composite formation, researchers can overcome the shortcomings of using a single method in isolation. HOPCs are pivotal in these designs because their micropores, mesopores, and macropores facilitate fast ion transport while providing abundant active sites for charge storage. Simultaneously, heteroatom doping (e.g., nitrogen, sulfur, or phosphorus) enhances the electrical conductivity, surface polarity, and redox-active sites, thereby augmenting both the electric double-layer and pseudocapacitive behavior.^[250] The coupling of HOPCs with TMOs or conductive polymers leverages the synergistic advantages of carbon's high electrical conductivity and structural stability with the pronounced redox activity of the metal oxides or polymers. This integration results in markedly enhanced capacitance and energy density in the resulting composite materials.^[251] The careful optimization of the electrode–electrolyte interface and charge–transfer pathways is central to achieving this synergy. Chemical functionalization, interfacial adhesion strategies, and precise structural engineering, such as vertically aligned architectures or core–shell composites, have improved electron transport, wettability, and mechanical robustness.^[252] Advanced characterization tools and computational modeling offer deep insights

into these interfaces, guiding refinements in material design, pore architecture, and dopant configurations. Consequently, multistrategy approaches that blend hierarchical porosity, heteroatom doping, and high-performance composite formation represent a promising route for building next-generation supercapacitors, targeting applications ranging from portable electronics to large-scale renewable energy storage.

Recent advancements in supercapacitor technology emphasize the formation of composites with pseudocapacitive materials to synergistically enhance energy density, power density, and cycling stability. These strategies leverage the complementary properties of diverse materials, addressing limitations of individual components while optimizing electrochemical performance. Combining TMOs like MnO_2 , NiO , CoO , and RuO_2 with carbon nanomaterials improves conductivity and prevents aggregation. For example, $\text{ZnO}@\text{NiM}-\text{LDH}$ composites exhibit enhanced ion diffusion and redox activity due to their hierarchical structure.^[253] $\text{Co}_3\text{S}_4/\text{rGO}$ composites achieve a specific capacitance of 1442.5 F g^{-1} at 1 A g^{-1} , attributed to rGO preventing particle stacking and enhancing electron transport.^[39] TMO-carbon architectures (e.g., $\text{MnO}_2/\text{graphene}$) mitigate the poor electrical conductivity of pure TMOs while retaining high theoretical capacitance.^[254]

PANI and PPy are widely used for their redox activity and mechanical flexibility. CuCoS/PANI composites demonstrate 88.3% capacitance retention after 10,000 cycles, leveraging PANI's mixed ionic-electronic conductivity and CuCoS's pseudocapacitive properties.^[255] PANI-doped carbon nanohorn (CNH) hybrids achieve 762 F g^{-1} specific capacitance, outperforming pure PANI due to CNH's high SSA and charge–transfer facilitation.^[256] Integrating metal sulfides/selenides with conductive scaffolds enhances redox kinetics. VSe_2/CuS nanocomposites achieve 853.9 F g^{-1} at 1 A g^{-1} , combining VSe₂'s conductivity with CuS's pseudocapacitance.^[257] CuCoS-PANI electrodes deliver a specific capacity of 467.5 C g^{-1} , benefiting from synergistic Faradaic reactions and structural stability.^[255] MOFs combined with metal oxides or sulfides offer tailored porosity and active sites. $\text{CuCo(5-NIPA)-Nd}_2\text{O}_3$ composites achieve an energy density of 109.68 Wh kg^{-1} , leveraging MOF-derived interfaces for efficient charge storage.^[258] Hydrothermal synthesis of VSe_2/CuS creates flake-like nanostructures, increasing SSA and ion-accessible sites.^[257] Rod-like Co_3S_4 on rGO improves mechanical stability and mitigates volume expansion during cycling.^[39] Carbon-based materials provide pathways for rapid electron transfer. For instance, $\text{Co}_3\text{S}_4/\text{rGO}$ composites reduce charge–transfer resistance by 40% compared to pure Co_3S_4 . Composite formation with pseudocapacitive materials addresses key challenges in the design of supercapacitors. Synergistic strategies, including morphological tuning, conductive scaffolding, and interfacial engineering, enable breakthroughs in specific capacitance and cycling stability (retaining >88% after 10,000 cycles).

Composites deliver the highest gravimetric capacitance but are only superior when interfaces are engineered. Oxide/carbon hybrids (e.g., $\text{Co}_3\text{S}_4/\text{rGO}$: 1442.5 F g^{-1} ; 47.7 Wh kg^{-1} ; 91%/10,000 cycles)^[39] and LDH/carbon architectures (1661.6 F g^{-1} ; 57.2 Wh kg^{-1})^[40] confirm the ceiling. Conducting-polymer hybrids add faradaic storage but face swelling/creep; careful nanoscale deposition (e.g., PANI-CNH: 762 F g^{-1} ; 95%/5000 cycles)^[256] stabilizes cycling.

MXene/MOF/TMD composites reduce R_{ct} and raise active-site density but risk restacking and voltage-window constraints. Quantitatively, the biggest practical wins couple thin, conformal pseudocapacitive layers to mesoporous carbon highways, keeping

diffusion lengths short and ESR low; thick shells, agglomerates, or binder-rich films erode the very gains they promise.

Several recent reviews have explored carbon-based composites with pseudocapacitive materials.^[44,221,259] This review,

Table 1. Summary of electrode engineering strategies for carbon-based supercapacitors.

Electrode material	Morphology/structure	Key features	Specific capacitance [$F\ g^{-1}$]	Energy density [$Wh\ kg^{-1}$]	Cycle stability [% retention, no. of cycles]	References
NiCoLDH-JAC	Flake-like layered structure on porous JAC	High SSA > 2000 $m^2\ g^{-1}$, good ion access, and synergistic redox reactions from Ni and Co ions	750	100	95, 10,000	[25]
MoSe ₂ /PANI Composite	Hollow microspheres (740 nm)	Synergistic effect, increased SSA	753.2	20.1	-	[55]
3D Silver Electrode	3D nanostructure	Reduced ion diffusion, pseudocapacitive effect	0.5 $F\ cm^{-2}$	385.87 $\mu Wh\ cm^{-2}$	95, 2600	[56]
NiCo-LDH on Carbon Felt	Vertical nanosheets (coral reef-like)	Efficient ion transport	1661.6	57.2	96.7, 3000	[40]
MnO ₂ /MnS Composite	Nanorods	Defect-induced active sites	305	5.7	110, 5000	[57]
YSe ₂ /MoSe ₂ /Ni _x B	Ternary heterostructure nanosheets	Synergistic interface, enhanced stability	893.3	39.5	128.17, 5000	[59]
Ti ₃ C ₂ T _x Nanosheets	Size-engineered nanosheets	Optimized interlayer spacing	658	-	100, 2000	[61]
N-doped porous carbon	HOPC	Enhanced pseudocapacitance, improved ion transport	343	42	90.9, 10,000	[73]
HOPC from KIT-6 template	HOPC	High SSA (2602 $m^2\ g^{-1}$), large pore volume	289	40	86.2, 3000	[36]
Lignin-derived porous carbon + Rhubarb	HOPC composite	Redox-active, sustainable	250.2	8.1	80.9, 3000	[109]
ZnO + Carbon dots	ZnO enhanced with carbon nanodots	Specific capacitance increased by ≈290%	179.9	15.4	91, 10,000	[110]
S-doped VSe ₂ /CNTs	Hybrid nanostructure	Synergistic pseudocapacitance and conductivity	96 $mF\ cm^{-2}$	36.3 $\mu Wh\ cm^{-2}$	87.2, 10,000	[112]
MOF-derived HOPC	Hierarchical porous	High electrochemical stability, large capacitance	546 $mAh\ g^{-1}$	-	83, 140	[115]
CNT-graphene hybrid	Interconnected 3D porous structure	Improved conductivity and charge storage	1349.2	60.4	90.6, 10,000	[285]
NiCoP	Intrinsically porous	High energy density, excellent diffusion	2805	37.7	82.9, 50,000	[133]
Quaternary-doped carbon (N/P/O/S)	Rhombic dodecahedral shapes	Synergistic effects	815	-	86, 10,000	[157]
MnO ₂ /Co ₃ O ₄ @SA-ECNFs	Binary metal oxides uniformly wrapped on super aligned electrospun carbon nanofibers	Porous structure, binder-free, improved electrochemical performance	728	64.5	71.8, 11,000	[226]
MnCo ₂ O ₄ /PPy/AC	Ternary hybrid composite	High energy storage capability	254.98	88.12	89.68, 10,000	[233]
ZnO@NiM-LDH	LDH composite with nanorods	Enhanced ion diffusion and redox activity	45.22	16.08	75, 50,000	[253]
Co ₃ S ₄ /rGO	Nanocomposite rods shape	Reduced charge-transfer resistance, high capacitance	1442.5	47.7	91, 10,000	[39]
Carbon nanohorns/PANI	Composites with different monomer:CNH ratios; polymer covers the CNH surface	Fast charge transfer, small internal resistance, and mechanical stability	762	-	95, 5000	[256]
VSe ₂ /CuS	Spherical and flake-like morphology via wet chemical synthesis	High conductivity of VSe ₂ and pseudocapacitive behavior of CuS	853.9	-	88.3, 10,000	[257]

however, uniquely emphasizes not only state-of-the-art composite configurations but also explicitly discusses the synergistic interplay between carbon materials and advanced pseudocapacitive materials (MXenes, MOFs, and TMDs). Furthermore, this review provides a comprehensive perspective on interfacial engineering, elucidating the critical role that interfaces play in enhancing composite stability, energy density, and ion diffusion kinetics, aspects that are often overlooked or minimally discussed in previous reviews. Table 1 summarizes the electrochemical performance metrics of carbon-based supercapacitor electrodes engineered through nanostructuring, heteroatom doping, and composite formation, highlighting how each strategy contributes uniquely to enhanced capacitance, energy density, and cycling stability.

5. Role of Electrolytes in Capacitance Enhancement

The performance of carbon-based supercapacitors is governed by the properties of the electrolyte, which determine ion transport, charge storage mechanisms, and the stability of the electrode-electrolyte interface. Ion size, mobility, viscosity, electrolyte conductivity, and electrochemical stability critically influence the formation of an electric double layer and pseudocapacitive effects. Aqueous electrolytes (e.g., H_2O , KOH, H_2SO_4 , and Na_2SO_4) exhibit high ionic conductivities but are limited by their narrow OPWs, whereas organic and IL electrolytes can operate over wider OPWs yet typically display lower conductivities.^[260] Emerging options, such as water-in-salt and redox-active electrolytes, seek to combine wide OPWs with high ionic conductivity, mitigating traditional performance trade-offs. Tailoring the electrolyte composition, pore structure accessibility, and surface interactions can thus optimize the capacitance, power delivery, and energy density, underscoring the importance of a holistic electrode-electrolyte design strategy for advanced carbon-based supercapacitors.^[261] Figure 8a highlights the role of electrolyte cations in supercapacitors, emphasizing that smaller cations facilitate better performance by enhancing charge storage through efficient ion transport and adsorption.^[261] Figure 8b illustrates the enhanced proton transport and energy storage performance of supercapacitors using f-NbS₂ with SPEEK-based electrolytes, highlighting the synergistic interaction between the electrolyte and electrode material for improved energy density.^[35]

Electrolyte properties, such as ionic conductivity, ion size, viscosity, and electrochemical stability, play a crucial role in governing ion transport, charge storage mechanisms, and, consequently, the overall performance and energy density of carbon-based supercapacitors. The choice of electrolytes affects the charge-discharge kinetics, where high ionic conductivity facilitates rapid ion transport, enhancing the supercapacitor's overall capacitance and energy density.^[262–264] For instance, organic electrolytes often provide wider electrochemical stability OPWs than aqueous electrolytes, allowing for higher operating OPWs and improved energy densities.^[222,265] Moreover, the interaction between the electrolyte and carbon

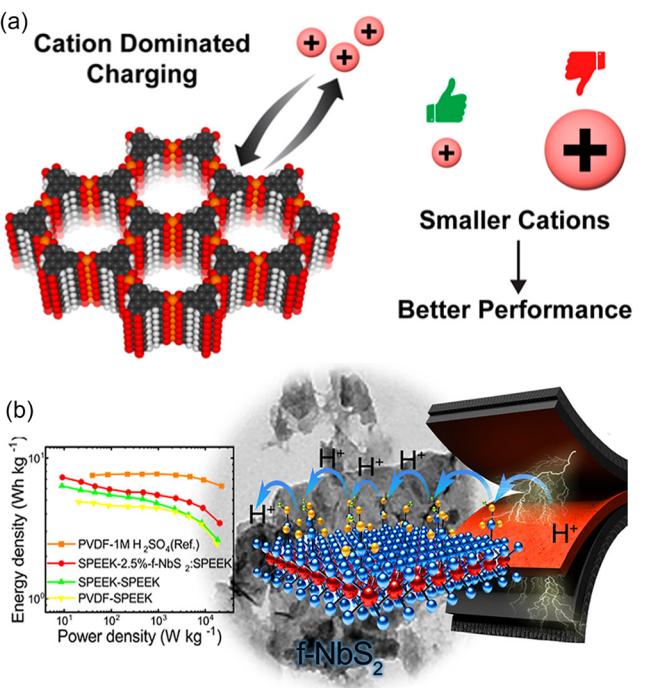


Figure 8. a) Impact of cation size on charging dynamics and performance of supercapacitors. Reproduced with permission.^[261] Reproduced under the term CC BY 4.0. Copyright 2024, Gittins., American Chemical Society. b) Enhanced proton transport and energy density with f-NbS₂ and SPEEK-based electrolytes in supercapacitors. Reproduced with permission.^[35] Reproduced under the term CC BY 4.0. Copyright 2022, Bagheri et al., American Chemical Society.

electrodes is crucial; a well-structured carbon electrode can optimize the ion adsorption and desorption processes, thereby reducing the internal resistance and improving efficiency.^[263,266] Functional groups on carbon surfaces can also enhance wettability, promoting better electrolyte penetration and ion diffusion.^[267,268] Recent advancements in hybrid and gel polymer electrolytes have shown promising results in improving the performance of supercapacitors by combining the benefits of both liquid and solid-state systems.^[154,269–271]

The selection and design of electrolytes are vital for maximizing the electrochemical performance of carbon-based supercapacitors and influencing their capacitance, energy density, and operational stability. Figure 9a illustrates the synergistic effects of the three electrolytes (LiPF_6 , KOH, and EMIMBF₄) on the electrochemical performance and structural stability of the supercapacitor electrodes, highlighting their complementary properties.^[272] The classification of electrolytes for carbon-based supercapacitors is crucial for optimizing their performance. It can be broadly categorized into aqueous, organic, and IL types, differing in OPW, ionic conductivity, and electrochemical stability, thus impacting the overall device performance. Aqueous electrolytes such as sodium sulfate are favored because of their high ionic conductivity and environmental friendliness; however, they typically operate within a limited OPW.^[262,273] On the other hand, organic electrolytes offer wider electrochemical OPWs and improved stability at elevated temperatures, making them suitable for high-energy applications.^[33,265] Gel polymer electrolytes

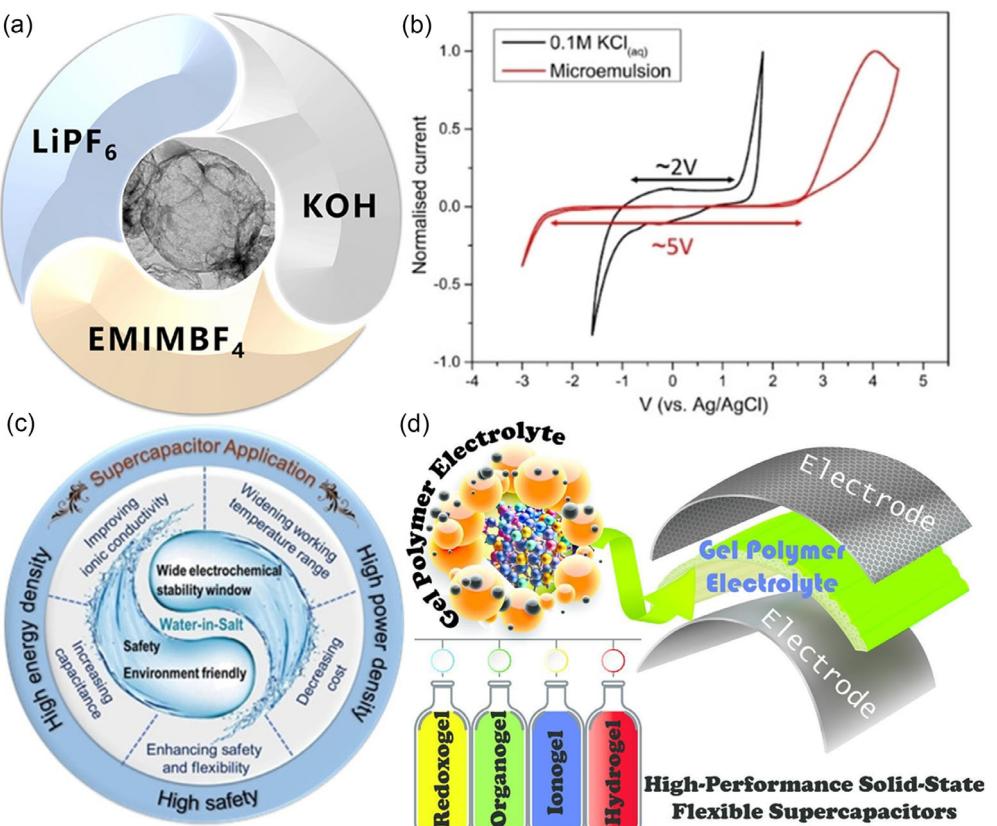


Figure 9. a) Synergistic interaction of LiPF_6 , KOH , and EMIMBF_4 electrolytes to enhance the performance of supercapacitors. Reproduced with permission.^[272] Copyright 2021, American Chemical Society. b) Comparison of the electrochemical stability of OPWs between 0.1 M KCl aqueous solution and microemulsion. Reproduced with permission.^[41] Copyright 2022, Wiley. c) Advantages of water-in-salt electrolytes for supercapacitor applications. Reproduced with permission.^[42] Copyright 2021, Wiley. d) Structural integration of gel polymer electrolytes with electrodes in high-performance flexible solid-state supercapacitors. Reproduced with permission.^[154] Copyright 2025, Elsevier.

combine the advantages of both liquid and solid electrolytes, providing enhanced safety and mechanical stability while maintaining good ionic conductivity.^[274] The choice of electrolytes has a significant impact on the energy and power densities of supercapacitors. For example, redox-active electrolytes can significantly enhance capacitance by enabling faradaic reactions at the electrode–electrolyte interface, thereby contributing to improved overall energy storage performance.^[275] Furthermore, the relationship between the electrolyte and carbon electrode material is crucial, as it affects ion diffusion and the development of an electric double layer, which is fundamental for charge storage. Therefore, recent research has focused on developing novel electrolytes that can maximize carbon-based supercapacitors' performance while addressing stability and efficiency challenges.

Carbon-based supercapacitors have shown impressive performance in various electrolyte systems. Supercapacitors with bio-based redox electrolytes have achieved a specific capacitance of 308 F g^{-1} and an energy density of 15 Wh kg^{-1} at a power density of 300 W kg^{-1} , demonstrating excellent stability with 72% capacitance retention after 10,000 cycles.^[158] In contrast, aqueous supercapacitors typically exhibit lower energy densities, around 9.2 Wh kg^{-1} , due to limited OPWs.^[272,276] However, innovative approaches, such as microemulsion electrolytes, have enabled

energy densities of up to 40 Wh kg^{-1} while maintaining a capacitance of $\approx 40 \text{ F g}^{-1}$,^[41] as shown in Figure 9b. Furthermore, using "water-in-salt" electrolytes has expanded the operating voltage to about 3.0 V, significantly enhancing energy density and power output.^[42] "Water-in-salt" electrolytes exhibit a broad electrochemical stability OPW, rapid ion transport, improved safety, cost-effectiveness, and environmental sustainability, making them highly promising for high-performance aqueous supercapacitors, as described in Figure 9c. Overall, integrating advanced electrolyte formulations and electrode materials continues to enhance the performance and stability of carbon-based supercapacitors. Dahiru et al. highlighted the role of gel polymer electrolytes, including hydrogels, ionogels, organogels, and redox-mediated gel polymer electrolytes, in advancing high-performance, flexible, and solid-state supercapacitors (Figure 9d), emphasizing their superior ionic conductivity, safety, and adaptability for next-generation energy storage solutions.^[154]

The wetting behavior, hierarchical porosity, and pore-size matching between carbon-based materials and electrolytes underpin efficient ion transport and double-layer formation. Heteroatom doping (e.g., N, O, and S) further enhances ion adsorption and introduces pseudocapacitive contributions, thereby boosting the overall capacitance. Ultimately, the

synergistic design of nanostructured carbons, specialized electrolyte formulations (including redox-active species), and interfacial engineering are crucial for achieving high-performance supercapacitors. Emerging strategies in electrolyte design emphasize redox-active species (e.g., quinones and transition metal ions), hybrid formulations combining ILs with organic or aqueous components, and bio-derived or biodegradable solvents to enhance energy density, conductivity, and environmental compatibility. By enabling faradaic charge storage and capitalizing on the broad OPWs of organic polymer-based electrolytes, these advanced electrolytes substantially enhance the overall performance and sustainability of supercapacitor systems. Electrolyte properties, encompassing ion dimensions, mobility, desolvation, OPW, ionic conductivity, and viscosity, are pivotal for supercapacitor performance, particularly in carbon-based electrodes, where the pore structure must be precisely matched to the ion size for optimal diffusion. Smaller ions exhibit higher mobility, thereby boosting micropore accessibility and capacitance; however, when pore diameters approach the size of the solvated ion, the energy-intensive desolvation process can limit power output. The ESW determines the operational voltage range and is strongly correlated with energy density. Aqueous electrolytes (e.g., sulfuric acid, potassium hydroxide, and neutral salts) offer high ionic conductivity but are limited to around 1.23 V by water decomposition. However, water-in-salt formulations have demonstrated the potential to expand this range. Organic electrolytes, typically comprising solvents such as acetonitrile or propylene carbonate, mixed with salts like tetraethylammonium

tetrafluoroborate, can reach voltages of ≈ 3 V, thereby boosting energy density; however, this comes at the expense of higher viscosity, reduced ionic conductivity, flammability, and susceptibility to thermal breakdown. ILs, known for their nonvolatility, wide ESWs (up to 4 V), and thermal stability, require careful structural modification to overcome their elevated viscosity and relatively low conductivity at room temperature. Finally, solid-state electrolytes, both polymeric and inorganic, hold promise for safer, leak-proof devices with extended longevity. However, their typically low room-temperature conductivity demands ongoing advances in composite design, plasticizers, and IL additives to realize flexible, high-performance supercapacitor systems.

Electrolytes play a crucial role in enhancing the capacitance of supercapacitors, significantly impacting their electrochemical performance. Recent studies have demonstrated that the use of different electrolyte compositions, including dual-redox electrolytes, can dramatically enhance specific capacitance, with enhancements reported to be approximately three times greater than those of traditional single-redox systems.^[277] The choice of electrolyte, particularly concentrated ionic solutions, has also been shown to improve charge transfer characteristics and stability, which is essential for the longevity and efficiency of supercapacitors.^[278] Furthermore, the use of gel electrolytes has emerged as a promising approach to address challenges related to ionic conductivity and mechanical flexibility, thereby optimizing the interfacial contact between electrodes and electrolytes to enhance overall performance.^[279,280] The integration of redox-active additives into electrolytes can improve energy storage capabilities, underscoring the importance of electrolyte

Table 2. Comparison of electrolyte systems for carbon-based supercapacitors.

Electrolyte	OPW [V]	Ionic conductivity [$S\text{ cm}^{-1}$]	Advantages	Energy density [Wh kg^{-1}]	References
PVA/H ₃ PO ₄	0–1	$2.1 \times 10^5 \text{ S m}^{-1}$	High ionic conductivity, excellent mechanical flexibility, strong electrode adhesion, easy fabrication process	$14.73 \mu\text{Wh cm}^{-2}$	[262]
KPF ₆	2.7	–	admirable stability at fast charging rate, OPW	14	[265]
EMIMBF ₄ + LiPF ₆ + KOH	0–3.0	0.6	high charge carrier, wide OPW, good thermal and chemical stability of the IL electrolyte	80.5	[272]
DEC + Gel electrolyte	1.1	6×10^{-3}	High ionic conductivity	–	[274]
Pivalic acid and ascorbic acid	1.2	–	Good columbic efficiency and high stability	15	[158]
EMIBF ₄ /PVDF	3.5	7246 S m^{-1}	Wide OPW of 3.5 V, fast rate capability, excellent stability cycles, superior conductivity	75	[286]
[DMIM][MeO(H)PO ₃] ₂	2.5	22.4×10^{-3}	Wide OPW, superior stability	38	[287]
MeEt ₃ NBF ₄ (ACN)	2.5	1.15×10^{-3}	Wide OPW, admirable stability	–	[288]
[P ₄₄₄₄][FuA]	4.6	0.032×10^{-3}	High specific capacitance, variable temperature and wide potential range, long-term cycling efficiency	29	[289]
EMIMTFSI	4	$13,000 \text{ S m}^{-1}$	Versatile even at high scan rate, fast rate performance, wide OPW	135.6	[282]
BMIMPF ₆	4	–	Wide OPW, high specific energy, stable	204	[283]
PYR ₁₄ -FSI + PIP ₁₃ -FSI	3.5	–	High OPW	–	[290]
1-ethyl-3-methylimidazoliumtetrafluoroborate (EMIMBF ₄), 1,3-dimethylimidazolium methyl phosphite [DMIM][MeO(H)PO ₃], ethylmethylimidazoliumbis(trifluoromethylsulfonyl)imide (EMIMTFSI), diethyl carbonate (DEC), potassium hexafluorophosphate (KPF ₆), N-methyl-N-propylpiperidinium bis(fluorosulfonyl)imide (PIP ₁₃ -FSI), and N-butyl-N methylpyrrolidinium bis(fluorosulfonyl)imide (PYR ₁₄ -FSI).					

design in achieving high-performance supercapacitors.^[281] This synergy between electrolyte composition and supercapacitor architecture is crucial for advancing energy storage technologies.

Electrolytes move the voltage lever and the transport lever simultaneously. Aqueous systems maximize conductivity but limit OPW ($\approx 1.0\text{--}1.23$ V); typical devices cluster around $\approx 9\text{--}15$ Wh kg $^{-1}$.^[272,276] Organic/IL systems open 2.5–4.0 V windows (e.g., EMIMTFSI up to ≈ 4 V; 135.6 Wh kg $^{-1}$,^[282] BMIMPF₆, 204 Wh kg $^{-1}$)^[283] while increasing viscosity/ESR and cost. Water-in-salt (≈ 3.0 V)^[42] and microemulsions (≈ 40 Wh kg $^{-1}$)^[41] offer appealing middle grounds. Gel and solid electrolytes trade a small transport penalty for safety, flexibility, and device integration.^[154] Performance is maximized when ion size/solvation is matched to the pore spectrum and when surface polarity (from dopants) promotes wetting. Poor matching yields high iR-drop and underutilized surface even in nominally high-SSA carbons.

Although electrolyte advancements have been summarized in multiple recent reviews,^[8,152,284] this review provides a comprehensive evaluation of both traditional and emerging electrolyte systems, uniquely highlighting the critical interplay between electrolyte characteristics and advanced electrode structures. Our review emphasizes innovative electrolyte developments, including water-in-salt electrolytes, gel polymer electrolytes, and bio-derived redox-active electrolytes, explicitly linking their performance benefits and limitations to nanostructure and composite electrodes. Furthermore, our holistic approach identifies the gaps and future directions for electrolyte development, with a particular emphasis on sustainability, scalability, and compatibility with advanced electrode designs. Table 2 compares key electrolyte systems used in carbon-based supercapacitors, detailing their OPWs, conductivity, advantages, and performance outcomes to illustrate their critical influence on device efficiency and energy storage potential.

6. Summary, Key Insights, and Future Perspectives

This review presents an in-depth assessment of the recent advances in improving carbon-based supercapacitors, focusing on nanostructure engineering, heteroatom doping, composite formation, and electrolyte optimization. Through nanostructure engineering, ion accessibility and overall capacitance are improved by tailoring pore structures and optimizing specific SSAs. Heteroatom incorporation introduces additional pseudocapacitive mechanisms, enhancing the electrodes' conductivity and wettability. Composite formation effectively combines carbon with pseudocapacitive materials, resulting in significant improvements in energy and power densities. Additionally, advancements in electrolytes have expanded OPWs, improved ion transport, and increased device stability. The review clearly outlines how the synergistic combination of these methods effectively alleviates the intrinsic deficiencies of using each approach independently, thus providing a clear direction for advancing next-generation carbon-based supercapacitor technologies. By

centering the electrolyte-electrode interface and the cross-effects among porosity, dopants, and pseudocapacitive phases, our synthesis goes beyond earlier single-strategy reviews to provide transferable, synergy-oriented design guidelines.

Despite the progress surveyed in this review, several concrete gaps still limit the translation of carbon supercapacitors to device-relevant performance. First, synthesis and electrode fabrication remain difficult to scale: many peak metrics come from thin, low-loading electrodes or multistep routes that are hard to reproduce at roll-to-roll scale; future studies should report yield, cost, and batch variability and demonstrate performance at practical areal loadings ($\geq 5\text{--}10$ mg cm $^{-2}$) and densities ($\approx 0.6\text{--}0.9$ g cm $^{-3}$) with iR-corrected Ragone plots that include inactive masses. Second, electrolyte stability at extremes is unresolved: aqueous systems are voltage-limited, whereas organic/ionic-liquid and water-in-salt formulations face viscosity, corrosion, and interphase instability, especially below -20 °C; progress will require localized high-concentration or antifreeze gel electrolytes that retain fast transport while sustaining $\geq 2.5\text{--}3.5$ V operation, selected by ion-pore size matching and validated through accelerated aging that couples high-voltage holds, temperature cycling, humidity, gas/by-product analysis, and interphase characterization on doped carbons, MXenes, and oxide/polymer skins. Third, long-term degradation in composites persists: pseudocapacitive phases swell, dissolve, restack, or lose percolation when coatings are thick or poorly anchored; durable designs should use ultrathin, conformal active layers grown *in situ* on mesoporous carbon "highways," interfacial anchoring via O/N/P sites, spacers or pillars to stabilize 2D phases, and operando diagnostics (EQCM, Raman/XRD, impedance mapping, *in situ* TEM/AFM) to link swelling, porosity evolution, and resistance growth to architecture and cycling conditions. Fourth, sustainability of materials and processing is under addressed: reliance on scarce metals, HF-based etching for some MXenes, and energy-intensive activation is at odds with scale; priorities include biomass-derived carbons, recyclable salt templating, HF-free etching or molten-salt/halide routes, water-based processing, quantified cradle-to-gate CO₂ and water use, and design for disassembly and materials recovery. Cutting across these themes, the field needs uniform reporting (areal loading, electrode density/thickness, OPW, ESR, iR-drop corrections, and full bill of materials), interface-aware design that treats the electrolyte-electrode interphase as a primary variable, and AI-assisted optimization of the coupled space of pore hierarchy, heteroatom chemistry, active-layer thickness, and electrolyte composition under manufacturing constraints. Addressing these specific gaps will convert the qualitative guidance of this review into actionable targets for high-energy, high-power, and durable carbon supercapacitors.

Recent breakthroughs in carbon-based supercapacitors have set new performance benchmarks, exemplified by state-of-the-art materials such as nitrogen-doped HOPCs, which exhibit specific capacitances exceeding 660 F g $^{-1}$ and energy densities of up to 108 Wh kg $^{-1}$. Similarly, advanced composite electrodes integrating graphene-based matrices with MXenes and TMOs have achieved energy densities exceeding 40 Wh kg $^{-1}$ and maintained capacitance retention of over 99% over tens of thousands of

cycles. These results underscore the considerable progress achieved by strategically combining multiple enhancement strategies. However, despite these remarkable advancements, critical challenges persist. Key issues such as maintaining structural stability during extensive cycling, scalable manufacturing, and achieving sustainable yet cost-effective synthesis processes remain significant barriers to widespread commercialization and practical deployment. Moreover, recent advancements in electrolyte design, particularly the development of “water-in-salt” systems and bio-derived redox-active electrolytes, have markedly expanded the OPW, thereby enabling higher operating voltages and elevating energy densities to levels comparable with those of conventional battery technologies. Nevertheless, challenges associated with electrolyte stability, ion transport efficiency at subzero temperatures, and the ecological impact of novel electrolyte components necessitate ongoing research. Overcoming these challenges necessitates an integrated research approach encompassing advanced characterization methods, computational modeling, and comprehensive life-cycle assessments, all aimed at bridging the gap between promising laboratory-scale performance and practical, real-world energy storage applications.

The extensive literature review highlights significant advancements achieved in recent years regarding carbon-based electrode materials for electrochemical supercapacitors. These advancements, notably characterized by innovations in material structures, surface chemistry, and electrolyte formulations, represent critical milestones that underscore the potential of carbon-based supercapacitors as viable high-performance energy storage solutions. Notably, the integration of hierarchical porous structures has emerged as a prominent trend, demonstrating enhanced ion transport and improved electrochemical performance. Such structures not only maximize electrode surface utilization but also effectively address the bottleneck associated with ion diffusion, a limitation frequently reported in traditional carbon electrodes. Moreover, recent studies consistently emphasize the significant role of heteroatom doping, with nitrogen-doped carbons widely recognized as particularly promising due to their improved electrical conductivity, surface wettability, and pseudocapacitive behavior. Nonetheless, recent comparative analyses reveal that codoping strategies involving combinations such as nitrogen-sulfur or nitrogen-phosphorus doping often yield even more superior performance due to their complementary electronic and chemical effects, signifying a growing trend towards multidoping strategies. Composite formation strategies represent another substantial area of advancement, wherein the combination of carbon materials with pseudocapacitive components, particularly metal oxides, MXenes, and conducting polymers, has demonstrated markedly increased capacitances and energy densities. However, despite these notable advancements, achieving robust cycling stability and maintaining the structural integrity of these composite materials over prolonged cycles remains a critical challenge, requiring further in-depth investigation.

Regarding electrolyte development, substantial efforts have been dedicated recently toward widening the OPW and

enhancing ion mobilities. Innovative electrolyte systems, such as ILs, water-in-salt solutions, and bio-derived redox-active electrolytes, show clear advantages in balancing high ionic conductivity and broader electrochemical stability. Nevertheless, the practical implementation and scalability of these electrolytes require continued optimization, particularly in addressing viscosity and cost constraints, as well as the environmental impacts associated with their broader deployment. In conclusion, the reviewed literature suggests a future where carbon-based supercapacitors will increasingly combine multiple synergistic strategies, advanced porous structures, strategic heteroatom doping, multicomponent composites, and tailored electrolyte systems to overcome existing limitations in energy density and cycling stability. Moving forward, targeted efforts towards scalable synthesis methods, eco-friendly material sourcing, and standardization of evaluation metrics will be critical to translating these laboratory-scale successes into commercially viable, sustainable energy-storage solutions. Such integrative, forward-looking approaches will ultimately determine the pace and extent of real-world implementation, as well as the broader contribution of carbon-based supercapacitors to addressing global energy challenges. While nanostructure engineering enhances surface area and ion accessibility, it often faces challenges related to ion diffusion and conductivity. Heteroatom doping can address these challenges by enhancing conductivity but may introduce instability in some instances. When these strategies are combined, their synergistic effects can significantly improve the overall performance of supercapacitors, although careful optimization is necessary to balance these trade-offs.

A pragmatic design map emerges from the data herein. To exceed $\approx 40 \text{ Wh kg}^{-1}$ while retaining kW kg^{-1} power and $\geq 90\% / 10,000$ -cycle stability, the most dependable route is as follows: hierarchical carbon (micro + meso) \rightarrow moderate N- or N/S-doping \rightarrow ultrathin pseudocapacitive skins (oxide or polymer) \rightarrow OPW-widening electrolyte (water-in-salt electrolyte or IL/organic blend). This stack directly targets C and V without sacrificing R or mechanical robustness. Two recurring pitfalls explain many underperforming devices: 1) over-activated carbons with poor connectivity and 2) thick, poorly percolated pseudocapacitive coatings. Future work should benchmark at device-relevant areal loadings and densities, report iR-corrected Ragone plots, and pair data-driven optimization with life cycle and techno-economic analyses to ensure laboratory gains translate beyond coin cells.

Future research should prioritize integrative and interdisciplinary approaches that incorporate advanced nanostructures, controlled heteroatomic doping techniques, multifunctional composites with architecturally designed structures, and custom-designed electrolyte formulations. A comprehensive exploration of electrode-electrolyte interfaces, utilizing advanced *in situ* characterization techniques and predictive computational modeling, will be central to resolving the technical hurdles. Additionally, addressing the challenges of scaling to commercial production, ensuring cost feasibility, and promoting environmental sustainability through sustainable synthesis technologies, biological sources, and eco-friendly manufacturing processes will be crucial to making production economically and practically viable.

Utilizing life-cycle assessment and eco-design principles will further encourage the creation of genuinely sustainable mass-produced supercapacitor technologies. Innovative methods that integrate various enhancement strategies signify an exciting advancement in the evolution of next-generation supercapacitors. By integrating nanostructure engineering, heteroatom doping, and composite formation, it is possible to leverage the synergistic advantages of each method to develop materials exhibiting exceptional performance characteristics. For example, hierarchical nanostructures infused with heteroatoms, in conjunction with pseudocapacitive materials, can achieve remarkable capacitance, rate capability, and cycle life. These synergistic strategies highlight the importance of a comprehensive approach to material design, where each element plays a vital role in enhancing the overall performance of the system. The importance of electrolytes in enhancing capacitance is notable, as they function as a medium for transporting and storing ions. The selection of electrolytes plays a crucial role in determining the OPW, ionic conductivity, and stability of supercapacitors. Aqueous, organic, and IL electrolytes offer unique benefits and drawbacks, with recent advancements highlighting the use of gel polymers and solid-state electrolytes for flexible and wearable applications. Optimizing supercapacitors requires careful consideration of how electrolytes interact with advanced electrode materials. Moreover, creating innovative electrolytes featuring broader OPWs and enhanced ionic mobilities offers exciting prospects for future progress.

A key future direction involves the comprehensive development of hybrid supercapacitor-battery systems, which exploit the complementary advantages of rapid charging in supercapacitors and high energy storage in batteries. Interdisciplinary research to optimize the interfacial design between these components and to develop innovative materials with self-healing and adaptive properties will significantly enhance device durability and reliability. Additionally, incorporating artificial intelligence (AI) and machine learning (ML) tools in materials discovery and electrode architecture design can rapidly accelerate the identification of optimized configurations and reduce the development cycle time. Another critical future pathway involves prioritizing scalable and sustainable manufacturing methods, such as additive manufacturing (3D printing), roll-to-roll processing, and green chemistry routes that utilize low-toxicity solvents and renewable precursors. These approaches must be paired with robust techno-economic analyses and environmental life-cycle assessments to guarantee commercial feasibility and ecological responsibility. Furthermore, standardizing performance metrics, characterization protocols, and reporting guidelines across research groups can significantly enhance reproducibility, comparability, and accelerate translation from laboratory-scale demonstrations to industrial-scale production and real-world deployment.

7. Limitations and Scope of the Present Review

This review critically evaluates the opportunities and limitations of high-energy carbon supercapacitors by examining synergistic

strategies, including nanostructuring, heteroatom doping, carbon-based composites, and electrolyte engineering, while highlighting the difficulty of isolating the influence of variables such as pore architecture, surface chemistry, wettability, and ion size. Persistent system-level constraints remain, encompassing trade-offs among energy density, power capability, and cycle life; mismatches between pore size and ion dimensions; the instability of surface functionalities under high voltage; and electrolyte windows that narrow under realistic thermal and chemical conditions. Strategy-specific shortcomings further complicate progress, with hierarchical carbons prone to pore collapse or tortuous ion pathways, heteroatom doping suffering from volatility or site blocking, composites facing interfacial resistance and mechanical mismatch, and advanced electrolytes constrained by safety, cost, or conductivity limits. Device-level and manufacturing realities are often underreported, including performance losses at practical mass loadings and electrode thicknesses, the roles of binders and conductive additives, challenges in scaling activation methods, roll-to-roll compatibility, formation protocols, and long-term aging. Environmental and supply-chain dimensions also warrant greater attention, particularly regarding the lifecycle impacts of precursors, activating agents, solvents, fluorinated species, and recycling at end-of-life. Moreover, the field would benefit from more transparent and standardized reporting practices, including full-cell data at $\geq 5\text{--}10 \text{ mg cm}^{-2}$, areal and volumetric metrics, temperature and voltage hold tests, leakage current, self-discharge, gas evolution, impedance spectra, electrolyte composition and water content, and explicit uncertainty bounds. Finally, while the scope of this synthesis is intentionally broad, it is bounded by heterogeneous testing protocols, publication bias, and a primary focus on carbon-dominant rather than hybrid or faradaic chemistries, underscoring the need for cautious interpretation of reported gains and a more rigorous framework for future research.

7.1. Cross-Cutting Limitations in Literature

A recurring limitation in the supercapacitor literature is the discrepancy between laboratory-reported performance metrics and their translation to practical, device-level relevance. Many of the highest specific capacitance values are obtained under conditions that favor inflated gravimetric results, such as three-electrode half-cell configurations, ultralow mass loadings ($<1\text{--}2 \text{ mg cm}^{-2}$), uncorrected iR-drops, or nonstandard voltage windows. These practices often produce values that cannot be directly scaled to areal (mF cm^{-2}) or volumetric (Wh L^{-1}) performance, which are more critical for real-world applications where inactive components (current collectors, separators, binders, electrolytes, and packaging) dominate mass and volume. Moreover, the absence of standardized testing protocols, ranging from differences in electrolyte composition, voltage ranges, and current densities to inconsistencies in electrode density, binder ratios, and porosity, complicates "apples-to-apples" comparisons across studies. Material characterization is likewise inconsistent: surface area and pore size are derived using different adsorbates and

analysis kernels, dopant levels are reported without systematic deconvolution of bonding states, and electrochemical impedance spectra are modeled with disparate equivalent circuits. Durability claims are further weakened by a lack of structural correlation, as gas evolution, binder migration, restacking, dissolution, and interphase growth are rarely monitored using operando or post-mortem diagnostics. The cumulative effect of these practices is that exceptional gravimetric values frequently mask trade-offs with volumetric or areal performance, obscure degradation mechanisms, and hinder the establishment of reliable benchmarks. To bridge this gap, the field must adopt rigorous, standardized, and application-relevant evaluation protocols that integrate both electrochemical and structural diagnostics, enabling more meaningful comparisons and accelerating translation from laboratory promise to device-level viability.

7.2. Strategy-Specific Limitations

Nanostructure engineering, heteroatom doping, composite formation, and electrolyte design each present distinct opportunities and inherent trade-offs for advancing electrochemical capacitors. Hierarchical porosity improves ion transport, yet excessive activation generates tortuous micropore networks that increase equivalent series resistance (ESR), reduce volumetric efficiency, and compromise electrode integrity during densification. Ultralight carbon foams and foils demonstrate remarkable rate capability but suffer from low tap density and mechanical fragility, limiting practical deployment. Heteroatom doping enhances wettability and introduces redox-active sites. However, over-doping disrupts graphitic domains, narrows pore necks, promotes parasitic reactions at elevated voltages, and complicates reproducibility due to model-dependent bonding-state quantification. The stability of dopants, particularly under aqueous or acidic conditions, remains insufficiently studied. Composite electrodes incorporating transition-metal oxides, sulfides, selenides, or conducting polymers deliver high gravimetric capacitances but frequently encounter diffusion bottlenecks, poor percolation, or interfacial delamination due to volumetric changes and mechanical stress during cycling. MXene restacking and polymer swelling further accelerate resistance growth and capacitance fade. Although advanced synthetic methods such as hydrothermal or templating approaches enable structural control, their multistep nature and reliance on cobalt/nickel chemistries raise cost and sustainability concerns. On the electrolyte front, aqueous formulations afford fast kinetics and safety but are voltage-limited, whereas organic, ionic liquid, and water-in-salt systems expand the operating potential window at the expense of ionic conductivity, viscosity, cost, low-temperature performance, and moisture stability. Gel and solid-state electrolytes improve safety and mechanical robustness but typically suffer from higher interfacial impedance and aging phenomena. Ultimately, the performance of these systems hinges on precise optimization of porosity, doping, composite interfaces, and interphase chemistry, as none of the enhancement strategies is free from intrinsic limitations that constrain scalability, reliability, or lifetime.

7.3. Device- and Manufacturing-Level Constraints

At the device level, the reliability of electrochemical energy storage systems is governed not only by intrinsic material properties but also by operational factors such as leakage current, self-discharge, gas evolution, and calendar life, yet these are rarely reported with rigor. Performance metrics often rely on ultra-thin electrodes, whereas at practical mass loadings ($\geq 5\text{--}10 \text{ mg cm}^{-2}$) the same chemistries exhibit significant rate and cycling limitations due to extended diffusion paths and increased internal resistance. Electrode calendaring, while enhancing volumetric energy density, risks mesopore collapse unless carefully co-optimized with slurry rheology, binder formulation, drying, and coating conditions. Choices between PVDF and water-borne binders, as well as processing steps, strongly influence porosity, crack formation, and electrode integrity, meaning that nanostructures successful in lab-scale films may fail under industrial roll-to-roll manufacturing. Beyond the electrode, current collectors are prone to corrosion or passivation, separators impose ionic and thermal limits, and packaging, tabs, and welds introduce uncompensated resistances, all of which complicate Ragone-based assessments by adding mass and ESR contributions that are often overlooked. Scaling from coin-cell demonstrations to pouch or cylindrical cells introduces additional design and reliability constraints, including gas management, thermal gradients, leakage dispersion, and balancing across modules. Moreover, the transition from advanced material synthesis to manufacturable electrodes highlights a persistent gap: many high-performing nanomaterials are derived from complex, low-yield processes—such as templating, chemical vapor deposition, or hydrothermal synthesis—that are difficult to scale economically. Cost, reproducibility, and manufacturing yield are further challenged by solvent management, precursor variability, residence times, and process stability. Yet, these industrially critical parameters are seldom reported alongside electrochemical performance, underscoring the disconnect between laboratory-scale studies and the realities of large-scale device manufacturing.

7.4. Environmental, Safety, and Supply-Chain Considerations

The pursuit of advanced electrode and electrolyte materials for electrochemical energy storage raises significant sustainability and safety challenges that must be addressed alongside performance gains. Activation agents, salts such as PF_6^- , and specific ionic liquid components have considerable environmental footprints, while scaling up MOF-, TMD-, and MXene-based precursors and Ni/Co-rich phases intensifies the dependence on critical minerals. Biomass-derived carbons offer a renewable alternative but demand strict control of feedstock variability and environmentally benign activation routes to be viable at scale. Hazardous processing routes, including chemical activation and etching, generate saline and alkaline effluents. Additionally, MXene syntheses often involve HF or fluoride salts, both of which pose concerns regarding toxicity and persistence. Organic electrolytes,

though effective, are flammable and toxic, while water-in-salt systems require extensive salt inventory and risk corrosion of conventional metals. Redox additive leaching, metal dissolution, and fluorinated salts further complicate end-of-life handling. Resource supply chains for cobalt, nickel, niobium, and vanadium remain geopolitically volatile and geographically concentrated, while even biomass carbons show seasonal and locational variability. Workplace hazards such as fine carbon dust and nitrogen-doping byproducts (NH_3 , NO_x) need strict occupational controls. Beyond safety, the inherently energy-intensive, high-temperature synthesis and pyrolysis steps add to the environmental burden. To ensure long-term viability, research and commercialization efforts must integrate life-cycle assessment, solvent recovery, HF-free etching, recyclable templating, and water-based processing, while prioritizing earth-abundant and sustainably sourced precursors, greener activation chemistries, and design-for-disassembly strategies. Only through such holistic approaches can claims of “green” and sustainable energy storage technologies withstand scrutiny.

7.5. Limitations of this Review

This review advances an interaction-aware framework for carbon-based supercapacitors while acknowledging several constraints inherent to synthesizing a rapidly evolving and heterogeneous field. Our approach favors breadth over exhaustive depth, emphasizing recent and representative studies over full coverage of all material families, and focusing primarily on carbon-centric electrodes and widely adopted electrolytes; pack-level considerations such as thermal management, balancing, and BMS, as well as noncarbon asymmetric partners, are discussed only briefly. Comparisons across studies are necessarily constrained by heterogeneous testing protocols, including differences in electrode loading, density, voltage window, electrolyte, and analysis methods, such that any normalization carries associated uncertainties. While we flag methodological differences and highlight best practices such as iR-corrected Ragone analysis, we refrain from reanalyzing raw datasets or refitting impedance spectra. Our synthesis is selective rather than encyclopedic, prioritizing illustrative peer-reviewed articles, and explicitly excluding patents, proprietary datasets, and the full diversity of chemistries relevant to hybrid devices. Cost and life-cycle considerations are addressed qualitatively, given the scarcity of consistent and transparent datasets. Inherent limitations of this work also include the influence of publication bias, as studies with positive or novel outcomes are more often published, potentially overstating reliability and reproducibility. Consequently, some niche but essential strategies receive less attention, and specific valuable reports are inevitably omitted. As the field continues to expand at a rapid pace, the particular data points and exemplars highlighted here will evolve. Still, the design principles extracted are intended to remain broadly instructive. Readers are encouraged to consult the most recent literature when applying these insights to research or practical device development.

7.6. Reporting and Benchmarking Recommendations

To strengthen reproducibility, comparability, and the translation of laboratory findings into scalable technologies, future studies on electrochemical capacitors should adopt a rigorous and standardized set of reporting practices. At a minimum, this should include both two- and three-electrode data, with explicit disclosure of electrode mass loading (mg cm^{-2}), thickness, density (g cm^{-3}), and component weight fractions. Device-relevant performance metrics should extend beyond gravimetric capacitance to include areal (mF cm^{-2}) and volumetric (F cm^{-3}) capacitances, full-cell Ragone plots with iR-corrected energy and power values, and electrochemical impedance spectra (R_s , R_{ct} Bode/phase) before and after cycling, along with fitting models and specified frequency ranges. Reporting should also encompass leakage current after defined voltage holds, self-discharge rates, coulombic and energy efficiency, voltage window cutoffs, temperature, separator, current collector, cell geometry, and calendaring pressures. Detailed material characterization, including porosity and pore-size distribution models, dopant contents with bonding-state analysis, and MXene surface terminations, should be disclosed alongside mechanical data for flexible devices. Stability protocols must extend beyond cycle counts to include voltage-hold tests, periodic rate checks, and post-mortem diagnostics. Benchmarking against community-standard electrolytes (e.g., 6 M KOH, 1 M TEABF₄/ACN) at practical loadings ($\geq 5 \text{ mg cm}^{-2}$) is recommended, with reporting of synthesis yield, solvent use, process flow, and, where feasible, summary life-cycle assessments to anchor claims of scalability and manufacturability. Finally, releasing raw charge–discharge, cyclic voltammetry, and impedance data with statistical analyses over multiple cells, along with interlaboratory or round-robin validation where possible, will make genuine advances transparent, reproducible, and meaningfully translatable toward commercial implementation.

8. Conclusions

Carbon-based supercapacitors hold significant promises for advancing energy storage technologies by delivering superior power densities, rapid charge–discharge capabilities, and prolonged cycle lifetimes, thereby bridging the gap between traditional capacitors and batteries. Recent advancements, systematically examined in this review, reveal that significant improvements can be achieved through synergistic strategies integrating nanostructure engineering, heteroatom doping, composite formation, and electrolyte optimization. Nanostructure engineering effectively enhances ion transport and accessibility by tuning pore architectures, resulting in increased SSAs and tailored ion diffusion pathways. Heteroatom doping introduces redox-active sites, enhances electronic conductivity, and improves wettability, thereby significantly boosting pseudocapacitance. Composites incorporating pseudocapacitive materials like TMOs, conducting polymers, and emerging 2D materials (MXenes, MOFs, and TMDs) demonstrate remarkable increases in both energy density and cycle stability. Moreover, advancements in electrolytes, including water-in-salt electrolytes, bio-derived redox electrolytes, and gel polymer

formulations, expand OPWs, enhance ionic conductivity, and improve electrochemical stability. Despite substantial progress, several critical challenges remain to be addressed. Structural degradation during extensive cycling, the limited scalability of advanced nanostructures, environmental impact considerations, and the economic feasibility of novel electrode materials and electrolytes remain significant barriers to progress. Future research should emphasize integrative and interdisciplinary approaches to overcome these limitations, incorporating advanced computational modeling, AI-driven material discovery, and sustainable manufacturing methods. Focusing on scalable synthesis processes, such as additive manufacturing and roll-to-roll production, will be critical to transitioning laboratory-scale successes to commercially viable solutions. Environmental life-cycle assessments and standardized performance metrics will also play vital roles in guiding future material design and ensuring ecological and economic sustainability. By leveraging these comprehensive strategies, carbon-based supercapacitors can evolve toward practical, efficient, and environmentally responsible energy storage solutions capable of meeting the ever-growing global energy demands, thereby significantly contributing to the sustainable energy transition.

Acknowledgements

The authors would like to acknowledge the support received from the Graduate School of Energy Science, Kyoto University, Yoshida-honmachi, Sakyo-ku, Kyoto 606-8501, Japan.

Conflict of Interest

The authors declares no conflict of interest.

Keywords: advanced electrolytes • carbon supercapacitors • heteroatom doping • hierarchical nanostructures • synergistic strategies

- [1] C. Zhang, Y.-L. Wei, P.-F. Cao, M.-C. Lin, *Renewable Sustainable Energy Rev.* **2018**, *82*, 3091.
- [2] D. Aurbach, E. Zinigrad, H. Teller, P. Dan, *J. Electrochem. Soc.* **2000**, *147*, 1274.
- [3] T. Wang, H. C. Chen, F. Yu, X. Zhao, H. Wang, *Energy Storage Mater.* **2019**, *16*, 545.
- [4] S. S. Shah, M. A. Aziz, Z. H. Yamani, *Chem. Rec.* **2022**, *22*, e202200018.
- [5] Y. Zhu, S. Murali, M. D. Stoller, K. J. Ganesh, W. Cai, P. J. Ferreira, A. Pirkle, R. M. Wallace, K. A. Cybrosz, M. Thommes, D. Su, E. A. Stach, R. S. Ruoff, *Science* **2011**, *332*, 1537.
- [6] L. L. Zhang, X. Zhao, *Chem. Soc. Rev.* **2009**, *38*, 2520.
- [7] J. Yan, Q. Wang, T. Wei, Z. Fan, *Adv. Energy Mater.* **2014**, *4*, 1300816.
- [8] T. Bhat, P. Patil, R. Rakhi, *J. Energy Storage* **2022**, *50*, 104222.
- [9] F. Béguin, V. Presser, A. Balducci, E. Frackowiak, *Adv. Mater.* **2014**, *26*, 2219.
- [10] L. Sun, K. Zhuo, Y. Chen, Q. Du, S. Zhang, J. Wang, *Adv. Funct. Mater.* **2022**, *32*, 2203611.
- [11] C. Neto, H. T. T. Pham, R. Omnée, A. Canizarès, A. Slodczyk, M. Deschamps, E. Raymundo-Piñero, *ACS Appl. Mater. Interfaces* **2022**, *14*, 44405.
- [12] X. Chen, R. Paul, L. Dai, *Natl. Sci. Rev.* **2017**, *4*, 453.
- [13] H. Jiang, P. S. Lee, C. Li, *Energy Environ. Sci.* **2013**, *6*, 41.
- [14] S. Bose, T. Kuila, A. K. Mishra, R. Rajasekar, N. H. Kim, J. H. Lee, *J. Mater. Chem.* **2012**, *22*, 767.
- [15] X. Zhang, B. Sun, X. Fan, H. Bai, P. Liang, G. Zhao, B. K. Saikia, X. Wei, *ACS Appl. Energy Mater.* **2021**, *4*, 985.
- [16] R. Kumar, S. Sahoo, E. Joanni, R. K. Singh, *J. Energy Chem.* **2022**, *74*, 252.
- [17] T. De Silva, C. Damery, R. Alkhaldi, R. Karunanithy, D. H. Gallaba, P. D. Patil, M. Wasala, P. Sivakumar, A. Migone, S. Talapatra, *ACS Appl. Mater. Interfaces* **2021**, *13*, 56004.
- [18] Y. Zheng, K. Chen, K. Jiang, F. Zhang, G. Zhu, H. Xu, *J. Energy Storage* **2022**, *56*, 105995.
- [19] X. Wang, C. Yang, J. Li, X. a. Chen, K. Yang, X. Yu, D. Lin, Q. Zhang, S. Wang, J. Wang, Z. Xia, H. Jin, *Adv. Funct. Mater.* **2021**, *31*, 2009109.
- [20] A. K. Mohamedkhair, M. A. Aziz, S. S. Shah, M. N. Shaikh, A. K. Jamil, M. A. A. Qasem, I. A. Buliyaminu, Z. H. Yamani, *Arab. J. Chem.* **2020**, *13*, 6161.
- [21] S. S. Shah, M. N. Shaikh, T. Rahman, M. I. Shams, M. A. Alfasane, S. M. Rahman, A. Raihan, S. M. A. Nayem, M. A. Aziz, *Chem. Asian J.* **2025**, *20*, e202401362.
- [22] S. S. Shah, M. A. Aziz, T. Ogawa, L. Zada, M. A. Marwat, S. M. Abdullah, A. J. Khan, M. Usman, I. Khan, Z. Said, M. Oyama, *Mater. Sci. Eng. R. Rep.* **2025**, *166*, 101041.
- [23] J. Wang, P. Wu, K. Wang, Y. Zhong, G. Sun, Y. Ji, Y. Ma, Y. Ma, *Batter. Supercaps* **2025**, *8*, e202400779.
- [24] R. Wang, X. Li, Z. Nie, Y. Zhao, H. Wang, *J. Energy Storage* **2021**, *38*, 102479.
- [25] S. S. Shah, M. A. Aziz, M. Ali, A. S. Hakeem, Z. H. Yamani, *Small* **2024**, *20*, 230665.
- [26] M. A. Aziz, S. S. Shah, Y. A. Mahnashi, W. Mahfuz, A. S. Alzahrani, A. S. Hakeem, M. N. Shaikh, *Small* **2023**, *19*, 2300258.
- [27] S. Chanarsa, N. Semakul, J. Jakmunee, P. Lamprasertkun, P.-H. Aubert, K. Ounnunkad, *J. Energy Storage* **2024**, *98*, 113083.
- [28] M. Ahmad, T. Nawaz, I. Hussain, U. Amara, X. Chen, Y. Eddahani, R. Walia, K. Zhang, *Small Methods* **2025**, *9*, 2401239.
- [29] B. Chettiannan, E. Dhandapani, G. Arumugam, R. Rajendran, M. Selvaraj, *Coord. Chem. Rev.* **2024**, *518*, 216048.
- [30] B. Gang, F. Zhang, X. Li, B. Zhai, X. Wang, Y. Song, *J. Energy Storage* **2021**, *33*, 102132.
- [31] K. Nasrin, V. Sudharshan, K. Subramani, M. Sathish, *Adv. Funct. Mater.* **2022**, *32*, 2110267.
- [32] K. Sharma, P. Kadyan, R. K. Sharma, S. Grover, *J. Energy Storage* **2024**, *100*, 113679.
- [33] M. Liu, Z. Song, D. Zhu, L. Li, L. Gan, M. Liu, *Energy Fuels* **2021**, *35*, 8443.
- [34] M. Sajjad, M. I. Khan, F. Cheng, W. Lu, *J. Energy Storage* **2021**, *40*, 102729.
- [35] A. Bagheri, S. Bellani, H. Beydaghi, M. Eredia, L. Najafi, G. Bianca, M. I. Zappia, M. Safarpour, M. Najafi, E. Mantero, Z. Sofer, G. Hou, V. Pellegrini, X. Feng, F. Bonaccorso, *ACS Nano* **2022**, *16*, 16426.
- [36] X. Bai, Z. Wang, J. Luo, W. Wu, Y. Liang, X. Tong, Z. Zhao, *Nanoscale Res. Lett.* **2020**, *15*, 88.
- [37] T. Wang, J. Guo, Y. Guo, J. Feng, D. Wu, *ACS Appl. Energy Mater.* **2021**, *4*, 2190.
- [38] X. Liu, Y. Wen, X. Chen, A. Dymerska, R. Wróbel, J. Zhu, X. Wen, Z. Liu, E. Mijowska, *ACS Appl. Energy Mater.* **2020**, *3*, 8562.
- [39] M. Zhu, X. Han, K. Zhang, H. Liu, C. Li, H. Sun, Q. Wen, J. Zhao, G. Wang, *Microstructures* **2025**, *5*, 2025040.
- [40] M. Mazloum-Ardakani, S. A. Mozaffari, B. Rasche, F. Ebrahimi, *Sci. Rep.* **2025**, *15*, 10542.
- [41] F. R. Hughson, R. Borah, T. Nann, *Batter. Supercaps* **2021**, *4*, 1122.
- [42] X. Tian, Q. Zhu, B. Xu, *ChemSusChem* **2021**, *4*, 2501.
- [43] S.-H. Park, S.-B. Yoon, H.-K. Kim, J. T. Han, H.-W. Park, J. Han, S.-M. Yun, H. G. Jeong, K. C. Roh, K.-B. Kim, *Sci. Rep.* **2014**, *4*, 6118.
- [44] S. Kumar, G. Saeed, L. Zhu, K. N. Hui, N. H. Kim, J. H. Lee, *Chem. Eng. J.* **2021**, *403*, 126352.
- [45] T. Kim, S. Subedi, B. Dahal, K. Chhetri, T. Mukhiya, A. Muthurasu, J. Gautam, P. C. Lohani, D. Acharya, I. Pathak, S.-H. Chae, T. H. Ko, H. Y. Kim, *Adv. Sci.* **2022**, *9*, 2200650.
- [46] J. Tamuly, D. Bhattacharjya, B. K. Saikia, *Energy Fuels* **2022**, *36*, 12847.
- [47] L. Liao, A. Zhang, K. Zheng, R. Liu, Y. Cheng, L. Wang, A. Li, J. Liu, *ACS Appl. Mater. Interfaces* **2021**, *13*, 28222.
- [48] Z. Yu, L. Tetard, L. Zhai, J. Thomas, *Energy Environ. Sci.* **2015**, *8*, 702.
- [49] J. Zeng, C. Xu, T. Gao, X. Jiang, X.-B. Wang, *Carbon Energy* **2021**, *3*, 193.

- [50] T. T. Salunkhe, B. Bathula, I. T. Kim, V. Thirumal, K. Yoo, *Crystals* **2024**, *14*, 482.
- [51] M. Zhi, C. Xiang, J. Li, M. Li, N. Wu, *Nanoscale* **2013**, *5*, 72.
- [52] S. S. Shah, M. A. Aziz, E. Cevik, M. Ali, S. T. Gunday, A. Bozkurt, Z. H. Yamani, *J. Energy Storage* **2022**, *56*, 105944.
- [53] M. F. Ehsan, A. Fazal, S. Hamid, M. Arfan, I. Khan, M. Usman, A. Shafiee, M. N. Ashiq, *J. Environ. Chem. Eng.* **2020**, *8*, 104556.
- [54] B. R., R. S., V. Hegde, K. H., *RSC Adv.* **2024**, *14*, 37644.
- [55] H. Zhang, G. He, D. Zheng, H. HuangFu, Y. Li, Y. Mi, M. Wu, H. Yuan, *Nanotechnology* **2024**, *34*, 415705.
- [56] A. S. González, J. García, V. Vega, R. C. Flores, V. M. Prida, *ACS Omega* **2023**, *8*, 40087.
- [57] M. Rahaman, M. R. Islam, M. R. Islam, *Nanoscale Adv.* **2024**, *6*, 4103.
- [58] S. Masanta, C. Nayak, S. Maitra, S. Rudra, D. Chowdhury, S. Raha, M. Pradhan, B. Satpati, P. Pal, A. Singha, *ACS Appl. Nano Mater.* **2023**, *6*, 5479.
- [59] R. Sukanya, R. Karthik, M. Hasan, C. Breslin, J.-J. Shim, *Chem. Eng. J.* **2023**, *473*, 145487.
- [60] T. Hu, Z. Fu, X. Liu, L. Li, C. Xu, Y. Zhou, F. Cao, J. Xia, X. Chen, G. Wang, F. Xu, *Nat. Commun.* **2024**, *15*, 9293.
- [61] H. Liu, X. Chang, L. Li, M. Zhang, *Molecules* **2025**, *30*, 241.
- [62] N. M. Santhosh, K. K. Upadhyay, G. Filipič, J. Zavašnik, M. de Fátima Montemor, U. Cvelbar, *Carbon* **2023**, *203*, 686.
- [63] Y. Wang, L. Zhang, H. Hou, W. Xu, G. Duan, S. He, K. Liu, S. Jiang, *J. Mater. Sci.* **2021**, *56*, 173.
- [64] R. Kumar, E. Joanni, S. Sahoo, J.-J. Shim, W. K. Tan, A. Matsuda, R. K. Singh, *Carbon* **2022**, *193*, 298.
- [65] G. Kothandam, G. Singh, X. Guan, J. M. Lee, K. Ramadass, S. Joseph, M. Benzigar, A. Karakoti, J. Yi, P. Kumar, A. Vinu, *Adv. Sci.* **2023**, *10*, 2301045.
- [66] R. Mendoza, J. Oliva, V. Rodriguez-Gonzalez, *Int. J. Energy Res.* **2022**, *46*, 6989.
- [67] M. Sultana, M. H. Rownok, M. Sabrin, M. H. Rahaman, S. N. Alam, *Clean. Eng. Technol.* **2022**, *6*, 100382.
- [68] S. Mishra, R. Srivastava, A. Muhammad, A. Amit, E. Chiavazzo, M. Fasano, P. Asinari, *Sci. Rep.* **2023**, *13*, 6494.
- [69] J. Fischer, K. Thümmler, S. Fischer, I. G. G. Martinez, S. Oswald, D. Mikhailova, *Energy Fuels* **2021**, *35*, 12653.
- [70] S. Li, T. Xing, Y. Wang, P. Lu, W. Kong, S. Li, X. Su, X. Wei, *Front. Energy Res.* **2021**, *9*, 680761.
- [71] L.-C. Zhang, Z.-H. He, J.-F. Hou, L.-B. Kong, *ACS Appl. Energy Mater.* **2022**, *5*, 7081.
- [72] L. Hou, C. Kong, Z. Hu, Y. Han, B. Wu, *Energy Technol.* **2022**, *10*, 2100933.
- [73] Y. Wen, L. Chi, X. Wen, X. Chen, E. Mijowska, *Adv. Electron. Mater.* **2020**, *6*, 2000450.
- [74] L. Cai, Y. Zhang, R. Ma, X. Feng, L. Yan, D. Jia, L. Ai, N. Guo, L. Wang, *Molecules* **2023**, *28*, 3660.
- [75] E. Taer, A. Apriwandi, W. Febriani, R. Taslim, *ChemistrySelect* **2022**, *7*, e202201810.
- [76] X. Zhao, C. Li, L. Sha, K. Yang, M. Gao, H. Chen, J. Jiang, *Polymers* **2022**, *14*, 3377.
- [77] S. Wang, Y. Liu, H. Wang, X. Wang, L. Li, T. Wang, *ACS Appl. Energy Mater.* **2023**, *6*, 9371.
- [78] G. Du, H. Wang, J. Liu, P. Sun, T.-h. Chen, *ACS Appl. Nano Mater.* **2022**, *5*, 13384.
- [79] R. G. Shrestha, S. Maji, L. K. Shrestha, K. Ariga, *Nanomaterials* **2020**, *10*, 639.
- [80] X. Yang, T. Lv, J. Qiu, *Small* **2023**, *19*, 2300336.
- [81] L. K. Shrestha, S. Shahi, C. L. Gnawali, M. P. Adhikari, R. Rajbhandari, B. P. Pokharel, R. Ma, R. G. Shrestha, K. Ariga, *Materials* **2022**, *15*, 8335.
- [82] S. S. Shah, M. A. Aziz, M. Usman, A. S. Hakeem, S. Ali, A. S. Alzahrani, in *Biomass-Based Supercapacitors*, Vol. 1, (Eds: M. A. Aziz, S. S. Shah), Wiley, Hoboken, NJ **2023**, pp. 435–459.
- [83] S. S. Shah, M. A. Aziz, M. Ali, M. Usman, S. Khan, F. Shehzad, S. N. A. Shah, S. Ullah, in *Biomass-Based Supercapacitors*, Vol. 1, (Eds: M. A. Aziz, S. S. Shah), Wiley, Hoboken, NJ **2023**, pp. 461–483.
- [84] N. Siraj, S. Macchi, B. C. Berry, T. Viswanathan, *Electrochim* **2020**, *1*, 410.
- [85] H. Wang, S. Wu, B. Fan, X. Liu, Y. Nie, Y. Zhou, *J. Electrochim. Soc.* **2021**, *168*, 110535.
- [86] G. Li, W. Xing, Y. Li, W. Mao, G. Du, J. Zhang, *Chemistryselect* **2020**, *5*, 2008.
- [87] F. Wu, X. Ren, F. Tian, G. Han, J. Sheng, Y. Yu, Y.-Q. Liu, W. Yang, *N. J. Chem.* **2022**, *46*, 19667.
- [88] Z. Zhai, W. Yan, L. Dong, J. Wang, C. Chen, J. Lian, X. Wang, D. Xia, J. Zhang, *Nano Energy* **2020**, *78*, 105193.
- [89] A. Afif, S. M. Rahman, A. T. Azad, J. Zaini, M. A. Islan, A. K. Azad, *J. Energy Storage* **2019**, *25*, 100852.
- [90] Y. Zhang, Y.-P. Zhao, L.-L. Qiu, J. Xiao, F.-P. Wu, J.-P. Cao, Y.-H. Bai, F.-J. Liu, *Diamond Relat. Mater.* **2022**, *129*, 109331.
- [91] Q. Qin, J. Wang, Z. Tang, Y. Jiang, L. Wang, *Ind. Crops Prod.* **2024**, *208*, 117878.
- [92] T. Prasankumar, D. Salpekar, S. Bhattacharyya, K. Manoharan, R. M. Yadav, M. A. C. Mata, K. A. Miller, R. Vajtai, S. Jose, S. Roy, *Carbon* **2022**, *199*, 249.
- [93] Z. Shu, Z. Liu, H. Cao, *Chem. Asian J.* **2020**, *15*, 2992.
- [94] Z. Dai, P. Ren, Z. Chen, Z. Guo, X. Hou, W. He, Y. Jin, F. Ren, *Adv. Mater. Interfaces* **2021**, *8*, 2101266.
- [95] M. Xing, A. Gao, Y. Liang, S. Deng, D. Shu, S. Su, F. Yi, X. Zhou, Z. Zhu, *ACS Appl. Energy Mater.* **2020**, *4*, 888.
- [96] L. Xie, F. Su, L. Xie, X. Guo, Z. Wang, Q. Kong, G. Sun, A. Ahmad, X. Li, Z. Yi, C.-M. Chen, *Mater. Chem. Front.* **2020**, *4*, 2610.
- [97] A. Kumar, H. K. Rathore, D. Sarkar, A. Shukla, *Electrochim. Sci. Adv.* **2021**, *2*, e2100187.
- [98] W.-D. Yang, J.-X. Wang, Y. T. Wu, H. W. Chang, H. H. Ko, *Materials* **2022**, *15*, 5748.
- [99] X. Wei, S. U. Shao-xing, Q. Zhang, *ChemElectroChem* **2023**, *10*, e202201166.
- [100] Y. A. Kumar, G. Koyyada, T. Ramachandran, J. H. Kim, S. Sajid, M. Moniruzzaman, S. Alzahmi, I. M. Obaidat, *Nanomaterials* **2023**, *13*, 1049.
- [101] J.-G. Kim, H.-C. Kim, N. D. Kim, M.-S. Khil, *Composites, Part B* **2020**, *186*, 107825.
- [102] P. L. Handayani, L. Nulandaya, J. Y. Cheon, T. Kim, S. I. Yoo, U. H. Choi, *Chem. Eng. J.* **2022**, *429*, 132273.
- [103] Q. Zhang, C. Deng, Z. Huang, Q. Zhang, X. Chai, D. Yi, Y. Fang, M. Wu, X. Wang, Y. Tang, Y. Wang, *Small* **2023**, *19*, 2205725.
- [104] L. Jiang, L. Sheng, Z. Fan, *Sci. China Mater.* **2018**, *61*, 133.
- [105] N. M. Keppetipola, C. Olivier, T. Toupancre, L. Cojocaru, *Sustain. Energy Fuels* **2021**, *5*, 4784.
- [106] C. Lai, Y. Guo, H. Zhao, H. Song, X. Qu, M. Huang, S. W. Hong, K. Lee, *Adv. Compos. Hybrid Mater.* **2022**, *5*, 2557.
- [107] H. Liu, Z. Sun, Y. Chen, W. Zhang, X. Chen, C.-P. Wong, *ACS Nano* **2022**, *16*, 10088.
- [108] P. Xie, W. Yuan, X. Liu, Y. Peng, Y. Yin, Y. Li, Z. Wu, *Energy Storage Mater.* **2021**, *36*, 56.
- [109] T. Wang, S. Hu, W. Yu, Y. Hu, S. Yan, M. Wang, W. Zhao, J. Xu, J. Zhang, *ACS Appl. Energy Mater.* **2023**, *6*, 2347.
- [110] K. Phetcharee, T. Jorn-am, W. Pholauyphon, T. Kwamman, N. Sirisit, J. Manyam, C. Chanthad, P. Paoprasert, *Chemistryselect* **2023**, *8*, e202301428.
- [111] P. He, S. Chen, *ChemElectroChem* **2021**, *8*, 783.
- [112] K. A. Seeraj, K. Pramoda, C. S. Rout, *J. Mater. Chem. C* **2023**, *11*, 2565.
- [113] F. Liu, X. Zhao, P. Shi, L. Li, Q. Dong, M. Tian, Y. Wu, X. Sun, *Batteries* **2023**, *9*, 396.
- [114] A. Mateen, M. Z. Ansari, Q. Abbas, A. Muneeb, A. Ahmad, S. M. Eldin, F. M. Alzahrani, N. S. Alsaiari, S. Ali, M. S. Javed, *Molecules* **2022**, *27*, 7446.
- [115] Y. Zhou, A. M. Wemyss, O. B. Brown, Q. Huang, C. Wan, *N. J. Chem.* **2020**, *44*, 4353.
- [116] S. S. Shah, F. Niaz, M. A. Ehsan, H. T. Das, M. Younas, A. S. Khan, H. U. Rahman, S. M. A. Nayem, M. Oyama, M. A. Aziz, *J. Energy Storage* **2024**, *79*, 110152.
- [117] X. Wu, R. Liu, J. Zhao, Z. Fan, *Nano Mater. Sci.* **2021**, *3*, 241.
- [118] M. Girirajan, A. K. Bojarajan, I. N. Pulidindi, K. N. Hui, S. Sangaraju, *Coord. Chem. Rev.* **2024**, *518*, 216080.
- [119] A. F. EL-Mahdy, T. C. Yu, S.-W. Kuo, *Chem. Eng. J.* **2021**, *414*, 128796.
- [120] M. A. Aziz, S. S. Shah, S. Reza, A. S. Hakeem, W. Mahfoz, *Chem. Asian J.* **2022**, *17*, e202200869.
- [121] S. S. Shah, S. M. A. Nayem, N. Sultana, A. J. S. Ahammad, M. A. Aziz, *ChemSusChem* **2022**, *15*, e202101282.
- [122] F. Niaz, S. S. Shah, K. Hayat, M. A. Aziz, G. Liu, Y. Iqbal, M. Oyama, *Ind. Crops Prod.* **2024**, *219*, 119161.
- [123] D. Rao, W. Zhang, B. Cheng, Y. Wang, C. Lei, Q. An, M. Huang, L. Mai, *Batter. Supercaps* **2024**, *7*, e202400046.
- [124] J. Liu, Z. Ding, Q. Zhou, L. Wei, Y. Yan, *Batter. Supercaps* **2023**, *6*, e202300362.

- [125] X. Zhang, H. Lin, H. Peng, W. Li, T. Wang, J. Li, Q. Xiong, Y. Liu, X. Liu, *J. Electroanal. Chem.* **2024**, *975*, 118733.
- [126] J. G. McDaniel, *J. Phys. Chem. C* **2022**, *126*, 5822.
- [127] A. Kim, G. Kalita, J. H. Kim, R. Patel, *Nanomaterials* **2021**, *11*, 3213.
- [128] T. Periyasamy, R. Atchudan, S. P. Asrafali, M. S. Moorthy, R. Vanaraj, S.-C. Kim, *Polymers* **2021**, *13*, 2048.
- [129] E. Taer, A. Apriwandi, *ChemNanoMat* **2022**, *8*, e202200217.
- [130] L. K. Shrestha, Z. Wei, S. Gokulnath, R. G. Shrestha, R. P. Singh, M. Sathish, R. Ma, J. P. Hill, J. Nakamura, K. Ariga, *Nanomaterials* **2023**, *13*, 946.
- [131] A. Hryniwcka, J. Breczko, G. Siemiaszko, A. N. Papathanassiou, K. Góra-Marek, K. A. Tarach, K. Brzeziński, A. Ilinicka, A. P. Terzyk, K. H. Markiewicz, L. Echegoyen, M. E. Płoska-Brzezińska, *Sci. Rep.* **2023**, *13*, 10737.
- [132] M. K. Gupta, Y. Kumar, S. Neeleshwar, S. K. Sharma, *Phys. Status Solidi A* **2022**, *220*, 2200451.
- [133] S. Zhu, H. Tao, Y. Liu, X. Ma, K. Wang, Y. Wang, *J. Phys. Chem. B* **2022**, *126*, 10913.
- [134] Y. C. Kang, W.-D. Yang, *Nanomaterials* **2023**, *13*, 2060.
- [135] J. Li, Z. Xia, X. Wang, C. Feng, Q. Zhang, X. a. Chen, Y. Yang, S. Wang, H. Jin, *Adv. Mater.* **2023**, *36*, 2310422.
- [136] H. Wang, Z. Zhang, Y. Li, D. Pei, S. Wan, Y. Li, H. Lu, *Acad. J. Sci. Technol.* **2023**, *4*, 78.
- [137] V. Kakani, S. Ramesh, H. M. Yadav, C. Bathula, P. K. Basivi, R. R. Palem, H. S. Kim, V. R. Pasupuletti, H. Lee, H. Kim, *Sci. Rep.* **2022**, *12*, 12951.
- [138] G. Sahoo, S. R. Polaki, P. Anees, N. G. Krishna, T. Mathews, M. Kamruddin, *ACS Appl. Energy Mater.* **2021**, *4*, 791.
- [139] J. R. Choi, J. W. Lee, G. Yang, Y.-J. Heo, S. J. Park, *Catalysts* **2020**, *10*, 256.
- [140] M. B. Wayu, *Solids* **2021**, *2*, 232.
- [141] Q. Abbas, L. Wen, A. Mateen, N. U. Hassan, A. Idrees, Z. U. Rehman, M. A. Bajaber, M. S. Javed, *Coatings* **2022**, *12*, 1101.
- [142] C. Ding, T. Liu, X. Yan, L. Huang, S. Ryu, J. Lan, Y. Yu, W.-H. Zhong, X. Yang, *Nano-Micro Lett.* **2020**, *12*, 63.
- [143] C. Qiu, L. Jiang, Y. Gao, L. Sheng, *Mater. Des.* **2023**, *230*, 111952.
- [144] X. Cai, Q. Ren, W. Sun, F. Yang, *Int. J. Energy Res.* **2021**, *45*, 21414.
- [145] V. Sunil, B. Pal, I. I. Misnon, R. Jose, *Mat. Today: Proc.* **2021**, *46*, 1588.
- [146] S. Mishra, L. Pradhan, B. K. Jena, *Energy Fuels* **2025**, *39*, 13151.
- [147] W. Li, W. Zhang, Y. Xu, G. Wang, W. Sui, T. Xu, Z. Yuan, C. Si, *Chem. Eng.* **2024**, *497*, 154829.
- [148] X. Zhu, Y. Zeng, X. Zhao, D. Liu, W. Lei, S. Lu, *EcoEnergy* **2025**, *3*, e70000.
- [149] D. R. Lobato-Peralta, P. U. Okoye, C. Alegre, *J. Power Sources* **2024**, *617*, 235140.
- [150] A. Mendhe, H. S. Panda, *Discov. Mater.* **2023**, *3*, 29.
- [151] S. Lan, C. Yu, J. Yu, X. Zhang, Y. Liu, Y. Xie, J. Wang, J. Qiu, *Small* **2025**, *21*, 2309286.
- [152] M. Saha, A. Kumar, R. Kanaoujiya, K. Behera, S. Trivedi, *Energy Fuels* **2024**, *38*, 8528.
- [153] A. P. Varghese, D. d. M. Zanata, S. Lashkari, M. Criado-González, M. Forsyth, P. C. Howlett, A. N. Rider, N. Goujon, I. Villaluenga, *Batter. Supercaps* **2025**, *8*, e202400591.
- [154] A. D. Shuaib, S. S. Shah, A. S. Alzahrani, M. A. Aziz, *J. Energy Storage* **2025**, *107*, 114851.
- [155] N. Sarfraz, N. Kanwal, M. Ali, K. Ali, A. Hasnain, M. Ashraf, M. Ayaz, J. Ifthikar, S. Ali, A. Hendi, N. Baig, M. F. Ehsan, S. S. Shah, R. Khan, I. Khan, *Energy Storage Mater.* **2024**, *71*, 103619.
- [156] H. Liu, Z. Cui, Y. Sun, Z. Qiao, Y. Zhang, Q. Bai, Y. Wang, *Energy Mater.* **2025**, *5*, 500024.
- [157] Z. Zuhra, M. Ahmad, Y. Abbas, S. Ali, S. Li, I. Hussain, G. Xie, X. Wang, *ACS Appl. Nano Mater.* **2023**, *6*, 18045.
- [158] R. Shakil, M. N. Shaikh, S. S. Shah, A. H. Reaz, C. K. Roy, A.-N. Chowdhury, M. A. Aziz, *Asian J. Org. Chem.* **2021**, *10*, 2220.
- [159] C. Cui, Y. Gao, J. Li, C. Yang, M. Liu, H. Jin, Z. Xia, L. Dai, Y. Lei, J. Wang, S. Wang, *Angew. Chem., Int. Ed.* **2020**, *59*, 7928.
- [160] F. Razmjooei, K. Singh, T. H. Kang, N. Chaudhari, J. Yuan, J.-S. Yu, *Sci. Rep.* **2017**, *7*, 10910.
- [161] R. Bahadur, B. Wijerathne, A. Vinu, *ChemSusChem* **2024**, *17*, e202400999.
- [162] S. Seenivasan, K. I. Shim, C. Lim, T. Kavinkumar, A. T. Sivagurunathan, J. W. Han, D.-H. Kim, *Nano-Micro Lett.* **2023**, *15*, 62.
- [163] X. Feng, Y. Bai, M. Liu, Y. Li, H. Yang, X. Wang, C. Wu, *Energy Environ. Sci.* **2021**, *14*, 2036.
- [164] H. K. Zafar, S. Zainab, M. Masood, M. Sohail, S. S. A. Shah, M. R. Karim, A. O'Mullane, K. Ostrivkov, G. Will, M. A. Wahab, *Chem. Rec.* **2024**, *24*, e202300161.
- [165] Y. Zhou, R. Ma, S. L. Candelaria, J. Wang, Q. Liu, E. Uchaker, P. Li, Y. Chen, G. Cao, *J. Power Sources* **2016**, *314*, 39.
- [166] S. S. Shah, M. A. Aziz, Z. H. Yamani, *Diam. Relat. Mater.* **2023**, *140*, 110450.
- [167] S. S. Shah, M. A. Aziz, M. Oyama, *Meet. Abstr.* **2024**, *MA2024-01*, 11.
- [168] H. Chen, F. Yu, G. Wang, L. Chen, B. Dai, S. Peng, *ACS Omega* **2018**, *3*, 4724.
- [169] T. Periyasamy, S. P. Asrafali, S.-C. Kim, *Polymers* **2023**, *15*, 1564.
- [170] L. Liu, W. Zhang, B. Lu, Z. Cheng, H. Cao, J. Li, Z. Fan, X. An, *Biomass Bioenergy* **2024**, *186*, 107265.
- [171] P. Wang, X. Qi, W. Zhao, Q. Meng, H. Bi, F. Huang, *Carbon Energy* **2020**, *3*, 349.
- [172] S. Lu, W. Yang, M. Zhou, L. Qiu, B. Tao, Q. Zhao, X. Wang, L. Zhang, Q. Xie, Y. Ruan, *J. Colloid Interface Sci.* **2022**, *610*, 1088.
- [173] F. Wang, J. Y. Cheong, Q. He, G. Duan, S. He, L. Zhang, Y. Zhao, I.-D. Kim, S. Jiang, *Chem. Eng. J.* **2021**, *414*, 128767.
- [174] D. Khalafallah, X. Quan, C. Ouyang, M. Zhi, Z. Hong, *Renewable Energy* **2021**, *170*, 60.
- [175] H. Wang, Y. Yuan, F. Xiong, B. Ma, J. Yang, Y. Qing, F. Chu, Y. Wu, *Chem. Eng. J.* **2023**, *476*, 146640.
- [176] Y. Yuan, Z. Chen, H. Yu, X. Zhang, T. Liu, M. Xia, R. Zheng, M. Shui, J. Shu, *Energy Storage Mater.* **2020**, *32*, 65.
- [177] R. Li, X. Hu, X. Li, R. Lu, Z. Liu, X. Wei, S. Shi, Y. Lv, H. Zhang, *N. J. Chem.* **2023**, *47*, 8759.
- [178] P. Dulyaseree, H. Sama, S. Sada, P. Ukkakimapan, V. Yordsri, V. Sattayarut, W. Wongwiriyapan, *Curr. Appl. Sci. Technol.* **2022**, *23*, 55003.
- [179] Z. Zhao, Y. Huang, J. Du, A. Chen, *Chem. Asian J.* **2023**, *18*, e202300486.
- [180] Y. B. Wang, Y. Chen, H. Zhao, L. Li, D. Ju, C. Wang, B. An, *Nanomaterials* **2022**, *12*, 3804.
- [181] B. Tong, W. Wei, Z. Wu, L. Zhao, W. Ye, J. Wang, C. Soutis, L. Mi, *ACS Appl. Energy Mater.* **2021**, *4*, 3221.
- [182] Q. Wang, J. Cheng, J. Ma, S. Cong, J. Liu, Y. Bai, H. Zhao, N. Yang, X. Wang, S. Xiong, B. Wu, K. Li, A. Zhou, *ACS Appl. Electron. Mater.* **2024**, *6*, 2594.
- [183] K. Xie, X. Liu, H. Li, L. Fang, K. Xia, D. Yang, Y. Zou, X. Zhang, *Carbon Energy* **2023**, *6*, e427.
- [184] C. Bie, H. Yu, B. Cheng, W. Ho, J. Fan, J. Yu, *Adv. Mater.* **2021**, *33*, 2003521.
- [185] Y. Ding, Y. Li, Y. Dai, X. Han, B. Xing, L. Zhu, K. Qiu, S. Wang, *Energy* **2021**, *216*, 119227.
- [186] B. Liu, R. Shi, R. Chen, C. Wang, K. Zhou, Y. Ren, Z. Zeng, Y. Liu, L. Li, *Appl. Surf. Sci.* **2021**, *538*, 147961.
- [187] X. Fu, R. Li, S. Yan, W. Yuan, Y. Zhang, W. Sun, X. Wang, *Chemistryselect* **2023**, *8*, e202301801.
- [188] M. Y. Ghobti, M. Farhadi, F. Abbasi, *ACS Omega* **2023**, *8*, 22964.
- [189] Q. Jiang, Y. Cai, X. Sang, Q. Zhang, J. Ma, X. Chen, *Energy Fuels* **2024**, *38*, 10542.
- [190] Y. Lv, J. Chen, W. Jia, X. Wu, J. Guo, L. Ding, D. Jia, F. Tong, *RSC Adv.* **2020**, *10*, 11033.
- [191] Y. Sinelnikova, A. Nizovskii, N. Uvarov, *Chim. Technol. Acta* **2023**, *10*, 15826.
- [192] L. K. Shrestha, R. G. Shrestha, R. Chaudhary, R. R. Pradhananga, B. M. Tamrakar, T. Shrestha, S. Maji, R. L. Shrestha, K. Ariga, *Nanomaterials* **2021**, *11*, 3175.
- [193] A. Pławnieć, A. Volpert, G. Dobele, A. Zhurinsh, K. Kaare, I. Kruusenberg, K. Kapräns, A. Knoks, J. Kleperis, *Sustainability* **2021**, *13*, 9237.
- [194] A. K. Thakur, K. Kurtyka, M. Majumder, X. Yang, H. Q. Ta, A. Bachmatiuk, L. Liu, B. Trzebicka, M. H. Rümmeli, *Adv. Mater. Interfaces* **2022**, *9*, 2101964.
- [195] J. Zhang, Y. Huang, C. Chen, K. She, *Energy Fuels* **2023**, *37*, 2379.
- [196] J. N. Pagaduan, A. Bhardwaj, T. Y. McCarty, S. Kraemer, C. J. Kearney, J. J. Watkins, T. Emrick, R. Katsumata, *Adv. Funct. Mater.* **2023**, *34*, 2310296.
- [197] R. Mishra, P. Panda, S. Barman, *Int. J. Energy Res.* **2021**, *46*, 2585.
- [198] S. Archana, P. C. Sharafudeen, P. Elumalai, *Energy Technol.* **2023**, *11*, 2201125.
- [199] X. Li, D. Ding, Z. Liu, L. Hui, T. Guo, T. You, Y. Cao, Y. Zhao, *ChemElectroChem* **2022**, *9*, e202200035.
- [200] A. M. Puziy, O. I. Poddubnaya, B. Gawdzik, J. M. D. Tascón, *Carbon* **2020**, *157*, 796.
- [201] L. Ma, Z. Bi, W. Zhang, Z. Zhang, Y. Xiao, H. Niu, Y. Huang, *ACS Appl. Mater. Interfaces* **2020**, *12*, 46170.
- [202] P. Li, C. Yang, C. Wu, B. Jiang, Y. Jin, W. Wu, *Nanomaterials* **2022**, *12*, 2931.
- [203] G. Nazir, A. Rehman, S. Hussain, M. Ikram, S. J. Park, *Int. J. Energy Res.* **2022**, *46*, 15602.

- [204] Y. Ye, H. Zhang, Y. Shi, Z. Wang, M. Yang, B. Liu, *Chem. Commun.* **2023**, 59, 6925.
- [205] S. Qin, P. Liu, J. Wang, C. Liu, S. Zhang, Y. Tian, F. Zhang, L. Wang, L. Cao, J. Zhang, S. Zhang, *Molecules* **2023**, 28, 6543.
- [206] X. Zhong, Q. Mao, Z.-S. Li, Z. Wu, Y. Xie, S.-H. Li, G.-C. Liang, H. Wang, *RSC Adv.* **2021**, 11, 27860.
- [207] M. Jiang, X. Yu, R. Gao, T. Yang, Z. Xu, L. Cao, *Nano* **2020**, 15, 2050096.
- [208] M. Zhou, A. M. Vassallo, J. Wu, *ACS Appl. Energy Mater.* **2020**, 3, 5993.
- [209] X. Jin, Y. Fu, Y. Li, P. Mi, Z. Liu, X. Ma, Y. Wang, F. Liu, X. Zhang, *ChemistrySelect* **2024**, 9, e202304146.
- [210] A. Gopalakrishnan, S. Badhulika, *J. Power Sources* **2020**, 480, 228830.
- [211] X. Liu, C. Yu, Z. Chen, F. Xu, W. Liao, W. Zhong, *Energy Fuels* **2021**, 35, 19801.
- [212] S. Liu, D. Shi, H. Tu, X. Yao, L. Deng, D. Xu, Y. Shao, Y. Wu, *Energy Technol.* **2021**, 9, 2100824.
- [213] G. S. S. Gopan, N. Abraham, S. Rani, D. Kurian, K. M. Alana, *ChemNanoMat* **2024**, 11, e202400505.
- [214] T. Jin, S. Jia, Q. Luo, W. Zhu, H. Lai, D. Huang, C. Wang, *Acs Omega* **2022**, 7, 37564.
- [215] S. Sahoo, R. Kumar, E. Joanni, R. K. Singh, J.-J. Shim, *J. Mater. Chem. A* **2022**, 10, 13190.
- [216] S. S. Shah, M. A. Aziz, A.-R. Al-Betar, W. Mahfuz, *Arab. J. Chem.* **2022**, 15, 104058.
- [217] A. H. Shabi, A. A. Mirghni, S. S. Shah, M. M. Mohamed, A. D. Shuaibu, A. Hussain, S. A. Ganiyu, M. A. Aziz, *J. Solid State Electrochem.* **2024**, 28, 4041.
- [218] S. S. Shah, M. A. Alfasane, I. A. Bakare, M. A. Aziz, Z. H. Yamani, *J. Energy Storage* **2020**, 30, 101562.
- [219] S. S. Shah, S. Oladejo, M. A. Ehsan, W. Iali, A. Alenaizan, M. N. Siddiqui, M. Oyama, A.-R. Al-Betar, M. A. Aziz, *Chem. Rec.* **2024**, 24, e202300105.
- [220] M. Usman, D. Li, C. Li, S. Zhang, *Sci. China Chem.* **2015**, 58, 738.
- [221] A. Borenstein, O. Hanna, R. Attias, S. Luski, T. Brousse, D. Aurbach, *J. Mater. Chem. A* **2017**, 5, 12653.
- [222] A. G. Olabi, Q. Abbas, M. A. Abdelkareem, A. H. Alami, M. Mirzaeian, E. T. Sayed, *Batteries* **2023**, 9, 19.
- [223] M. I. A. A. Maksoud, R. A. Fahim, A. E. Shalan, M. A. Elkodous, S. O. Olojede, A. I. Osman, C. Farrell, A. a. H. Al-Muhtaseb, A. S. Awed, A. Ashour, D. Rooney, *Environ. Chem. Lett.* **2020**, 19, 375.
- [224] M. Savić, A. J. Ležaić, N. Gavrilov, I. A. Pašti, B. N. Vasiljević, J. Krstić, G. Ćirić-Marjanović, *Materials* **2023**, 16, 1018.
- [225] R. Wang, Y. Hu, Z. Pan, J. Wang, *RSC Adv.* **2020**, 10, 34403.
- [226] K. Allado, M. Liu, A. Jayapalan, D. M. Arvapalli, K. Nowlin, J. Wei, *Energy Fuels* **2021**, 35, 8396.
- [227] S. C. Kishore, R. Atchudan, T. N. J. I. Edison, S. Perumal, M. Alagan, R. Vinodh, S. Manivannan, Y. R. Lee, *Energy Fuels* **2020**, 34, 14958.
- [228] K. Moorthi, S. Mohan, *Energy Fuels* **2024**, 38, 22719.
- [229] K. K. R. Reddyguntla, B. D. Kumar, *Energy Fuels* **2024**, 38, 10560.
- [230] M. Z. Iqbal, N. Amjad, M. W. Khan, *ChemElectroChem* **2022**, 9, e202200036.
- [231] Y. Pathaare, A. M. Reddy, P. Sangrulkar, B. Kandasubramanian, A. Satapathy, *Hybrid Adv.* **2023**, 3, 100041.
- [232] T. Yue, B. Y. Xia, X. Liu, Z. Wang, K. Qi, B. Y. Xia, *ChemElectroChem* **2021**, 8, 1021.
- [233] S. Kour, P. Kour, A. L. Sharma, *Nanoscale* **2024**, 16, 13627.
- [234] M. Ateş, *International Conference on Contemporary*, Vol. 1, Academic Research **2023**, 198–202.
- [235] I. J. Gómez, M. V. Sulleiro, D. Mantione, N. Alegret, *Polymers* **2021**, 13, 745.
- [236] P. Yang, J. Xie, L. Wang, X. Chen, F. Wu, Y. Huang, *Adv. Mater. Interfaces* **2020**, 7, 2000949.
- [237] X. Yang, Y. Tian, S. Li, Y. P. Wu, Q. Zhang, D. S. Li, S. Zhang, *J. Mater. Chem. A* **2022**, 10, 12225.
- [238] N. Khosroshahi, M. Bakhtian, A. Asadi, V. Safarifard, *Nano Express* **2023**, 4, 042002.
- [239] S. S. Shah, M. A. Aziz, P. I. Rasool, N. Z. K. Mohmand, A. J. Khan, H. Ullah, X. Feng, M. Oyama, *Sustain. Mater. Technol.* **2024**, 39, e00814.
- [240] S. I. Basha, S. S. Shah, A. Helal, M. A. Aziz, D.-Y. Yoo, *Case Stud. Constr. Mater.* **2024**, 21, e03586.
- [241] P. Siddhu, A. S. R. K., S. Radhakrishnan, S. M. Jeong, C. S. Rout, *Batter. Supercaps* **2025**, 8, e202400466.
- [242] D. Xu, Z. Zhang, K. Tao, L. Han, *Dalton Trans.* **2023**, 52, 2455.
- [243] J. Cherusseri, D. Pandey, J. Thomas, *Batter. Supercaps* **2020**, 3, 860.
- [244] M. Sun, W. Ye, J. Zhang, K. Zheng, *Inorganics* **2024**, 12, 112.
- [245] R. Venkatkarthick, J. Qin, T. Maiyalagan, *Int. J. Energy Res.* **2022**, 46, 22474.
- [246] R. Ahmad, N. Iqbal, T. Noor, S. K. Nemaní, L. Zhu, B. Anasori, *ACS Appl. Nano Mater.* **2023**, 7, 253.
- [247] D. V. Lam, U. N. T. Nguyen, E. Roh, W. Choi, J. H. Kim, H. Kim, S. M. Lee, *Small* **2021**, 17, 2100670.
- [248] Z. Yang, Y. Hu, Y. Chen, Q. Yang, Z. Shi, C. Xiong, *Cellulose* **2021**, 28, 7097.
- [249] X. Zhao, N. Wang, L. Li, Z. Fang, S. Tang, J. Gu, *J. Chem.* **2024**, 2024, 1.
- [250] L. Yang, Y. Yang, S. Wang, X. Guan, X. Guan, G. Wang, *Energy Fuels* **2020**, 34, 5032.
- [251] Z. Yang, M. Xiang, W. Zhu, J. Hui, H. Qin, *ACS Sustain. Chem. Eng.* **2020**, 8, 6675.
- [252] L.-Q. Mai, A. Minhas-Khan, X. Tian, K. M. Hercule, Y.-L. Zhao, X. Lin, X. Xu, *Nat. Commun.* **2013**, 4, 2923.
- [253] G. Pandey, S. Serawat, K. Awasthi, *ACS Nanosci. Au* **2024**, 4, 399.
- [254] C. V. V. M. Gopi, S. Alzahmi, M. Y. Al-Haik, Y. A. Kumar, F. Hamed, Y. Haik, I. M. Obaidat, *Mater. Today Sustain.* **2024**, 28, 100981.
- [255] H. Hassan, M. W. Iqbal, H. Alrobei, F. Riasat, A. M. Afzal, A. M. Saeedi, H. B. Albargi, A. Rehmat, *Nanoscale Adv.* **2024**, 6, 1507.
- [256] C. C. Chang, T. Imae, *ACS Sustain. Chem. Eng.* **2018**, 6, 5162.
- [257] I. Khan, D. Arif, A. U. Shah, K. Safeen, K. M. Alotaibi, B. Ali, W. M. Girma, A. Safeen, *Sci. Rep.* **2025**, 15, 19761.
- [258] A. Khizar, M. Z. Iqbal, A. Zakir, M. Shaheen, S. M. Wabaidur, M. M. Faisal, *RSC Adv.* **2024**, 14, 10120.
- [259] Z. Dong, Q. Zhou, *Nanoscale* **2025**, 17, 9786.
- [260] B. Pal, S. Yang, S. Ramesh, V. Thangadurai, R. Jose, *Nanoscale Adv.* **2019**, 1, 3807.
- [261] J. W. Gittins, K. Ge, C. J. Balhatchet, P.-L. Taberna, P. Simon, A. C. Forse, *J. Am. Chem. Soc.* **2024**, 146, 12473.
- [262] W. Wu, H. Ma, Z. Zhang, Y. Gu, J. Zhang, S. Li, R. Zhang, *J. Mater. Chem. A* **2022**, 10, 22707.
- [263] H. T. T. Thanh, M. D. Thi, T. N. Trinh, S. P. Tung, T. D. Cong, T. T. Y. Nhi, P. A. Le, D. T. M. Huong, *Vietnam J. Chem.* **2024**, 62, 54.
- [264] S. S. Shah, M. Das, T. Ogawa, *Batteries* **2025**, 11, 193.
- [265] M.-E. Yvenat, B. Chavillon, E. Mayousse, F. Perdu, P. Azaïs, *Batteries* **2022**, 8, 135.
- [266] K. Fan, X. Lei, J. Zhang, T. Yu, H. Chen, J. Liu, *Energy Technol.* **2023**, 11, 2201281.
- [267] C. K. Roy, S. S. Shah, A. H. Reaz, S. Sultana, A. N. Chowdhury, S. H. Firoz, M. H. Zahir, M. A. A. Qasem, M. A. Aziz, *Chem. Asian J.* **2021**, 16, 296.
- [268] S. Zallouz, J. M. L. Meins, C. M. Ghimbeu, *Energy Adv.* **2022**, 1, 1051.
- [269] A. D. Shuaibu, S. S. Shah, S. Alzahrani, M. A. Aziz, *J. Energy Storage* **2024**, 92, 112040.
- [270] N. Yadav, N. Yadav, S. A. Hashmi, *ACS Appl. Energy Mater.* **2021**, 4, 6635.
- [271] T. Mao, H. Chen, J. Li, F. Liu, X. Wang, S. Wang, *ACS Appl. Energy Mater.* **2020**, 3, 5163.
- [272] J. Yang, J. Liu, Y. Chen, B. Chang, X. Zhang, X. Wang, *ACS Appl. Energy Mater.* **2021**, 4, 13766.
- [273] E. Taer, H. E. Wahyuni, A. Apriwandi, R. Taslim, *J. Appl. Mater. Technol.* **2023**, 4, 31258.
- [274] M. A. Azam, T. J. Yu, T. A. T. Adaham, M. F. Aziz, A. Takasaki, *J. Teknol.* **2022**, 84, 59.
- [275] K. Nasrin, S. Gokulnath, M. Karnan, K. Subramani, M. Sathish, *Energy Fuels* **2021**, 35, 6465.
- [276] J. Yu, C. Yu, W. Guo, Z. Wang, Y. Ding, Y. Xie, K. Liu, H. Wang, X. Tan, H. Huang, J. Qiu, *Adv. Funct. Mater.* **2022**, 32, 2204609.
- [277] D. Vijayan, T. K. Kumaresan, R. Subashchandrabose, R. M. Gnanamuthu, K. Vediappan, S. S. Gunasekaran, *Ionics* **2024**, 30, 5037.
- [278] Z. A. Zafar, R. Weisser, G. Abbas, M. Silhavik, P. Kumar, J. Červenka, *ChemSusChem* **2025**, 18, e202401681.
- [279] Y. Wang, X. Zhang, P. Ji, G. Meng, J. Wu, L. Cui, Z. Liu, Y. Tai, *J. Appl. Polym. Sci.* **2024**, 141, e55727.
- [280] H. Wang, Q. Zhang, S. Chen, X. Liu, J. Liu, W. He, X. Liu, *ACS Appl. Mater. Interfaces* **2024**, 16, 56170.
- [281] V. S. Mansi, P. Dubey, A. Bakandritsos, S. Sundriyal, U. K. Tiwari, A. Deep, *Nanoscale Adv.* **2025**, 7, 2105.
- [282] C. Li, X. Zhang, K. Wang, X. Sun, G. Liu, J. Li, H. Tian, J. Li, Y. Ma, *Adv. Mater.* **2017**, 29, 1604690.

- [283] X. Wang, H. Zhou, E. Sheridan, J. C. Walmsley, D. Ren, D. Chen, *Energy Environ. Sci.* **2016**, *9*, 232.
- [284] J. Lu, J. Zhang, X. Wang, J. Zhang, Z. Tian, E. Zhu, L. Yang, X. Guan, H. Ren, J. Wu, X. Li, G. Wang, *J. Energy Storage* **2024**, *103*, 114338.
- [285] Y. Liu, N. Xin, Q. Yang, W. Shi, *J. Colloid Interface Sci.* **2021**, *583*, 288.
- [286] L. Feng, K. Wang, X. Zhang, X. Sun, C. Li, X. Ge, Y. Ma, *Adv. Funct. Mater.* **2018**, *28*, 1704463.
- [287] H. H. Rana, J. H. Park, G. S. Gund, H. S. Park, *Energy Storage Mater.* **2020**, *25*, 70.
- [288] J. Xu, N. Yuan, J. M. Razal, Y. Zheng, X. Zhou, J. Ding, K. Cho, S. Ge, R. Zhang, Y. Gogotsi, R. H. Baughman, *Energy Storage Mater.* **2019**, *22*, 323.
- [289] I. A. Khan, F. U. Shah, *ACS Sustain. Chem. Eng.* **2020**, *8*, 10212.
- [290] W.-Y. Tsai, R. Lin, S. Murali, L. L. Zhang, J. K. McDonough, R. S. Ruoff, P.-L. Taberna, Y. Gogotsi, P. Simon, *Nano Energy* **2013**, *2*, 403.

Manuscript received: July 27, 2025

Revised manuscript received: September 17, 2025

Version of record online:
