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REVIEW ARTICLE

Heteroatom-Doped Biomass-Derived Porous Carbon for Supercapacitor Applications: A Review

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Abstract

The increasing demand for high-performance energy storage technologies poses significant challenges to conventional materials, particularly regarding the need for enhanced energy and power output in symmetric supercapacitors. This review explores the potential of biomass-derived porous carbon as a solution, emphasizing the role of heteroatom doping in combating these challenges. By introducing various heteroatoms - including oxygen (O), nitrogen (N), phosphorus (P), sulfur (S), and boron (B) - into the carbon matrix, recent research has demonstrated significant improvements in material wettability, pore structure optimization, and overall electrochemical performance. Specifically, strategies utilizing dual and multi-heteroatom doping, such as N/O, P/O, S/O, N/S, N/O/S, N/O/P, N/P/S, N/P/B, O/P/S, and N/O/P/S, have been shown to generate synergistic effects that markedly enhance energy storage capacity and electrical conductivity compared to traditional single-atom doping schemes. The review provides a comprehensive evaluation of the energy storage capacity, cycling stability, and energy/power output associated with heteroatom-doped activated carbons. Key insights reveal that achieving a uniform pore distribution, along with the establishment of hierarchical connections between 2D and 3D nanostructures, is vital for optimizing electrochemical performance. Importantly, the incorporation of heteroatoms not only augments electrical conductivity but also facilitates more efficient electrochemical reactions - essential elements for the development of high-performance supercapacitors. The integration of 3D nano-hierarchical pores and multi-doping heteroatoms delivers outstanding supercapacitor performance, with a capacitance of 401 F/g and an impressive energy output of 76 W/kg in a symmetric 2-electrode configuration. This performance firmly positions it as a competitive alternative to commercial batteries. In conclusion, this review highlights the innovative structural modifications and doping strategies employed in biomass-derived carbon materials, including synthesis processes crucial for generating unique 2D and 3D architectures. By detailing these methods and their contributions to improving the energy density of mixed electrical double-layer capacitors (EDLCs) and pseudocapacitors, this review underscores the competitive potential of these biomass-derived materials when compared with traditional energy storage devices such as batteries. Future research directions could focus on optimizing these methodologies further and exploring commercial viability in large-scale applications.

Keywords: Self-doped, Co-doping, Multi-doped, Heteroatom, Biomass, Supercapacitor

Introduction

The primary barrier to the sustainable deployment of these technologies is the intermittent nature of energy availability. High-performance energy storage devices are essential to complement the renewable energy systems [1]. Batteries are currently among the most widely used commercial energy storage devices, capable of storing electrical energy through internal chemical reactions [2]. However, batteries face limitations in effectively harnessing energy from renewable sources due to unpredictable power fluctuations. The application of batteries in high-energy-demand devices can also diminish power density, thereby constraining the use in

more complex applications. Supercapacitor on the other hand, have been proposed as superior candidates for electrochemical energy storage due to the exceptionally high-power density, extended cycle life, high conductivity, excellent stability, as well as rapid and safe charging and discharging capabilities [3-5]. These attributes make supercapacitor highly versatile, with potential applications extending from simple electronic circuits to more sophisticated dynamic electrical devices, including LEDs, electric motors, crane systems, pulsed laser components, and telecommunications [6-9]. Supercapacitor are broadly categorized into 3 main types based on the operating principles specifically electric double-layer capacitors (EDLCs), pseudocapacitors, and hybrid supercapacitor [10]. EDLCs store energy by accumulating charges on the electrode surface through electrostatic forces, offering excellent charge-discharge cycle stability and suitable for applications requiring long lifetimes and high-power efficiency [11]. In general, common electrode materials for EDLCs include various forms of carbon, such as activated, nanofibers, nanotubes, and graphene [12]. Pseudocapacitors use redox (oxidation-reduction) reactions at the electrode surface for energy storage, enabling higher energy densities compared to EDLCs. Electrode materials typically comprise transition metal oxides, conductive polymers, and composites to maximize energy storage capacity [13]. However, pseudocapacitors often show lower cycle stability and shorter lifespans, which can limit suitability for long-term applications. Hybrid supercapacitor combine the electrostatic energy storage mechanism of EDLCs with the electrochemical energy storage of pseudocapacitors, aiming to enhance overall performance [14,15]. Doping electrode materials with metal elements and heteroatoms is a common strategy to increase apparent capacitance. Despite these improvements, challenges related to cyclic stability and durability persist, potentially affecting efficiency in complex and sustainable applications.

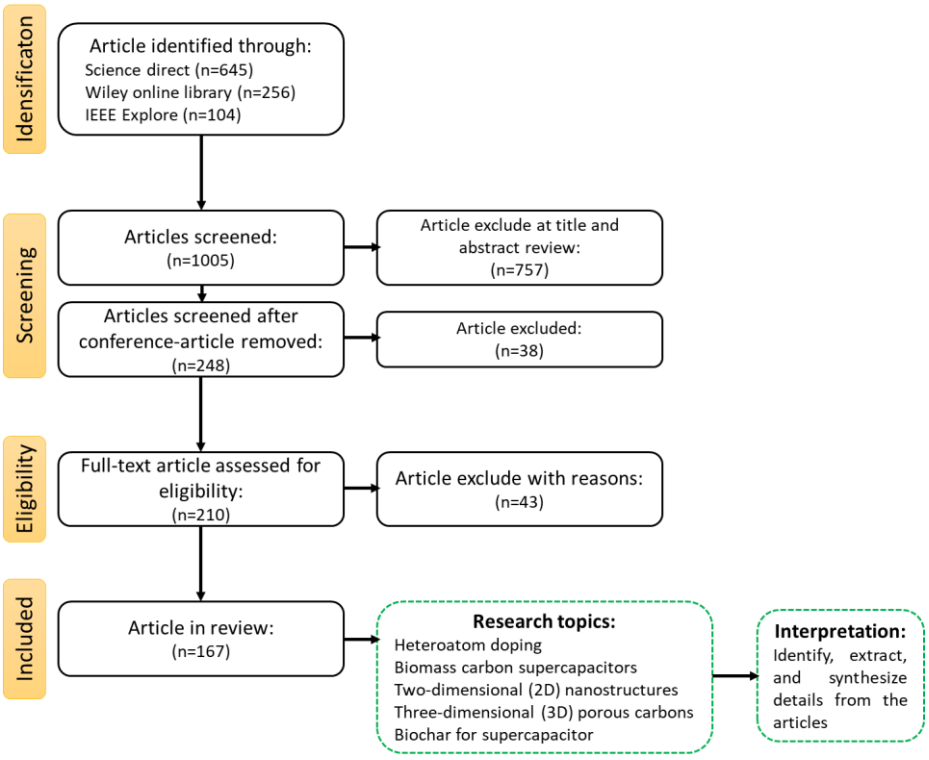

Figure 1 Prisma-flow-diagram for this article review.

The performance of supercapacitor is highly dependent on the active materials used in the electrodes. These materials can be categorized into 3 primary groups such as metal/transition metal oxides, conductive polymers, and carbon-based materials. In the context of pseudocapacitors, transition metal oxides are particularly prominent for high energy density, comparable to conventional batteries, and the ability to

undergo Faradaic redox reactions, which merge the characteristics of pseudocapacitance with those of EDLCs [16]. Conductive polymers within this category have excellent mechanical properties, well-suited for flexible supercapacitor applications. The remarkable properties of these materials have enabled the integration of supercapacitor into advanced military and space technologies [17-19]. The commercialization of electrode materials is influenced not only by electrochemical performance but also by factors such as resource availability, cost, and environmental impact. Although metal oxides are effective, the materials are often scarce, expensive, and large-scale exploitation poses environmental risks. This makes metal oxides less viable for widespread application. Conductive polymers, which are synthesized from fossil-based materials, also face limitations due to low stability, reduced conductivity during redox reactions, and the generation of toxic byproducts. The formation of an electric double-layer in supercapacitor can further reduce the conductivity of these polymers. Currently, biomass-derived activated carbon is the predominant choice for commercial supercapacitor, due to the low cost, excellent chemical stability, virtually unlimited cycle life, and reliable EDLC performance [20,21]. These attributes make activated carbon a highly competitive material for expanding the use of supercapacitor in broader and more sustainable applications. Although carbon-based EDLCs offer excellent power density, the energy density remains relatively low. Enhancing the specific energy of supercapacitor without compromising high power density is essential for expanding future application scope. Biomass-derived carbon materials have emerged as a promising alternative for supercapacitor applications due to their abundant availability, cost-effectiveness, and favorable environmental profile. These materials provide high mechanical stability and exhibit desirable electrical conductivity, both of which are critical for energy storage devices. The porous structure inherent in biomass-derived carbon increases its specific surface area, which is essential for facilitating charge storage through electrochemical double-layer capacitance (EDLC). These characteristics highlight the importance of using biomass as a sustainable precursor for carbon materials in supercapacitors. In the context of supercapacitors, carbon materials generally operate based on 2 main mechanisms: EDLC and pseudocapacitance. While traditional carbons primarily rely on the EDLC mechanism, they often suffer from limitations such as low specific capacitance and poor electron conductivity [22].

Several strategies can be used to enhance the quality of biomass-derived carbon materials, thereby improving the energy density of supercapacitor. The first strategy comprises designing biomass-derived carbon materials with hierarchical nanopore structures, particularly in 2-dimensional (2D) and 3-dimensional (3D) forms such as nanofibers, nanosheets, nanospheres, and interconnected pores [23-26]. Biomass, an abundant carbon source, offers significant potential for advanced carbon structures due to the rich lignin, cellulose, and hemicellulose content which naturally produce carbon in 2D and 3D configuration [27]. For example, 2D nanostructures such as nanofiber-nanosheets enable the development of active materials with unique electric charge transport properties, characterized by nanoscale thickness, infinite length, and well-defined layered structures [28,29]. These features are closely linked to the ability to maintain high power density in EDLCs for complex electronic device applications. Furthermore, the 3D hierarchical pore morphology plays a crucial role in providing abundant active sites and facilitating ion migration with minimal resistance in all directions [30,31]. The precise arrangement of these pores is essential for enhancing energy storage without compromising the power density of the device. However, these features decrease the material's conductivity due to the significant porosity they create, which poses a major challenge for biomass-derived carbon-based EDLCs.

Recent advances have demonstrated that incorporating heteroatoms into carbon structures can significantly enhance these materials. The introduction of doping elements such as nitrogen, sulfur, and phosphorus not only modify the electronic properties of carbon but also induces pseudocapacitance, leading to improved overall electrochemical performance of the electrodes. The presence of heteroatoms alters the surface chemistry of the carbon matrix, increasing its wettability and enabling better interactions with the electrolyte. This ultimately enhances the specific capacitance and cycling stability of supercapacitors

[32,33]. The intrinsic properties of biomass, combined with the advantages of heteroatom integration, offer a solution to the limitations of traditional carbon-based electrode materials. Among the various heteroatoms, nitrogen is particularly significant due to its capacity to enhance electron donation and facilitate additional charge storage. Nitrogen-doped carbon materials have shown impressive results regarding energy density and power output [34]. Other heteroatoms, such as oxygen, boron, phosphorus and sulfur, also contribute to performance enhancement through similar mechanisms [35]. Notably, studies have indicated that dual/triple/multi-doped biomass-derived carbon exhibits high specific capacitance values while maintaining stability over multiple cycles, making it a suitable candidate for supercapacitor electrode materials [33]. Additionally, the biomass carbonization process plays a critical role in determining the structure and functionality of the resulting carbon. Various methods, such as hydrothermal carbonization, pyrolysis, and gasification, allow for manipulation of porosity and surface characteristics. These synthetic strategies enable the development of carbon with hierarchical porous structures that maximize surface area and optimize ion diffusion pathways. This is essential for fast charge/discharge cycles in supercapacitors [36]. Furthermore, approaches to ensure the presence of doping on the carbon shell can be implemented either externally or internally, thus enhancing the pseudocapacitance effect of the electrode material.

The approaches discussed above have been widely implemented in biomass-derived carbon materials over the past 5 years, particularly in biomass sources. These include torch ginger leaves [30], aromatic evergreen laurel [37], young coconut coir fibers, pineapple leaf fibers, black tiger shrimp shells, Alpinia galanga stems, and bio-organic waste from nutmeg leaves [38] and etc. This review provides a detailed account of the contributions and synergistic effects of structural modifications and heteroatom doping approaches applied to biomass-derived carbon materials. Furthermore, this review provides a unique contribution by highlighting various methods for creating 2D and 3D surface structures, along with specific heteroatom doping on biomass-derived carbon frameworks for supercapacitor applications. In contrast to other reviews, which typically focus solely on the role of individual dopants, this review also includes detailed information about the synthesis process. The results demonstrate that the strategies significantly enhance the energy density of mixed-type EDLC and pseudocapacitor, showcasing competitive performance with other energy storage devices, such as batteries.

Methodology

review is based on articles published between 2017 to 2025, focusing on biomass-derived carbon materials applied in supercapacitors. A systematic literature search was conducted using multiple academic databases, including Google Scholar, Science direct, Wiley online library, and IEEE Xplore, to ensure a diverse range of studies were included. The keywords used in the search were “heteroatom doping,” “biomass carbon supercapacitors,” “2-dimensional (2D) nanostructures,” “3-dimensional (3D) porous carbons,” and “biochar,” facilitating a precise focus on relevant literature. This search was limited to studies published within the last 6 - 8 years to maintain relevance and currency in the field of electrochemical energy storage. In addition, we adhered to a specific timeframe for studies, which spanned from January 2017 to 2025, to guarantee a comprehensive overview of advancements made in the field. The selection of articles strived to include not only experimental studies but also review articles to gather a holistic understanding of the methodologies employed in biomass-derived carbon supercapacitor research. The inclusion criteria in this review article divide into: (i) studies focus on biomass-derived carbon materials used as electrode materials in supercapacitors. This includes various types of biomass, such as rice husk, corn starch, and pine sawdust, etc. (ii) Articles report on specific electrochemical performance metrics relevant to supercapacitors, including specific capacitance, energy density, and cyclic stability. Meanwhile, the exclusion criteria following: (i) non-peer-reviewed sources, such as conference papers and theses, were excluded to maintain academic rigor, focusing instead on journal articles with established scientific vetting; (ii) articles that did not specifically address the electrochemical performance or did not discuss heteroatom doping techniques relative to biomass-derived carbons were excluded; and (iii) studies with incomplete data or lacking empirical evidence on the performance of the supercapacitors

synthesized from biomass were disregarded, thus preserving the quality of the reviewed work. The prisma-flow-diagram for this study was illustrated in **Figure 1**. **Figure 2** shows the relationship between the selected biomass, the modifications of the carbon structure, and the heteroatom dopants that have been successfully produced. These modifications will be explored in greater detail, providing insights into how biomass can be used to create high-performance carbon materials suitable for applications in supercapacitor and other energy storage devices.

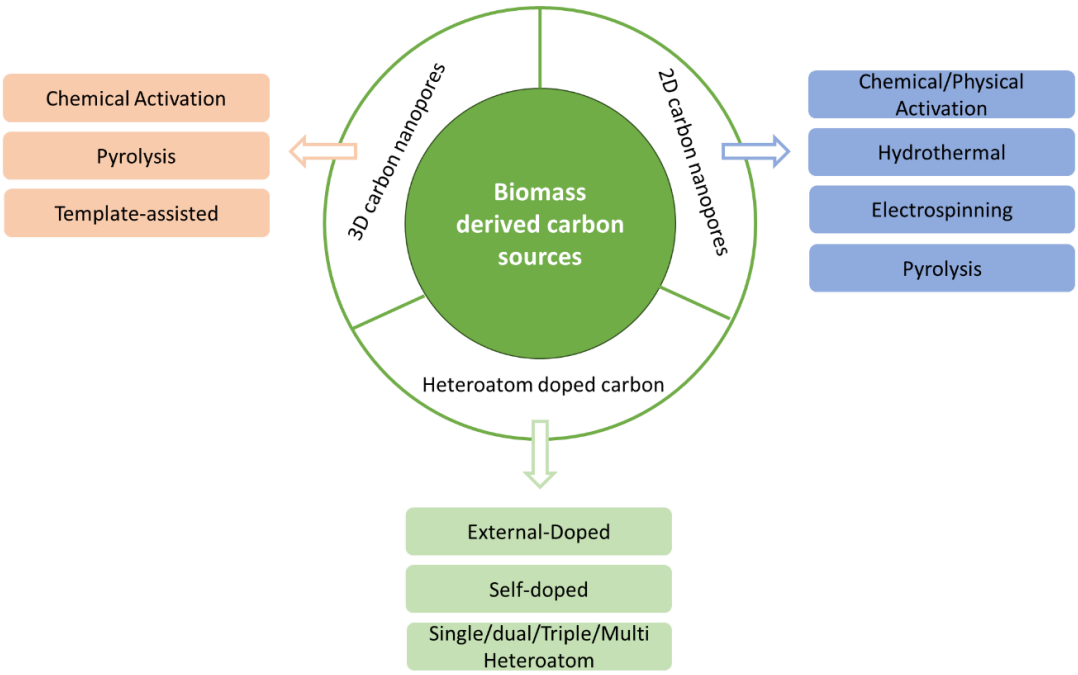

Figure 2 The biomass sources for obtained carbon structure modifications (2D and 3D), and the heteroatom dopants.

Modification of 2D and 3D porous carbon biomass-derived materials

A series of approaches have been used to engineer the surface pore structure of carbon-based materials into 2 (2D) and 3-dimensional (3D) nano-hierarchical configurations (**Tables 1** and **2**). The initial critical step is based on the selection of biomass with inherent potential for nano-hierarchical structuring. To establish the nanoporous architecture within carbon materials, multiple mechanical treatments are required. Furthermore, biomass precursors are subjected to chemical activation, typically using catalytic agents such as potassium hydroxide (KOH) and zinc chloride ($ZnCl_2$). This review focuses on the development of biomass-derived porous carbons, emphasizing the use chemical catalytic immersion as an efficient, environmentally sustainable synthesis strategy that preserves material conductivity. Additionally, the conversion of biomass into high-purity carbon is facilitated through a high-temperature pyrolysis process, which integrates carbonization and physical activation in a single step under nitrogen (N_2) and carbon dioxide (CO_2) gas atmospheres. These approaches have successfully yielded 2D and 3D nanoporous structures in selected biomass-derived carbons.

2D nanoporous carbon materials are carbon-based structures with nanometer-scale dimensions confined to a single plane in 3D space. These materials, including nanofibers, nanosheets, and nanospheres, have unique microstructures characterized by well-ordered layers, flake formations, and linear morphologies [39]. The microstructural features impart exceptional optical, mechanical, electrical, physical, and chemical properties to 2D nanoporous carbon materials, rendering them highly advantageous for supercapacitor applications [40]. The covalent bonds present on the surface of these materials confer extremely high electrical conductivity, facilitating rapid electron propagation across the plane. The layered porous architecture of 2D materials not only offers a large specific surface area with numerous active

sites for electrochemical reactions but also shortens ion transport pathways, significantly enhancing electrochemical performance. The hierarchical porous structure of 2D nanoporous carbon materials, when used as electrode materials, can accommodate volume changes during charge and discharge cycles, thereby ensuring superior long-term cycling stability.

Figure 3 (a) The flow-synthesis of carbon nanofiber derived from *Alpinia galanga* stem, (b) the structure surface porous carbon nanofiber 2D, and (c) Ragone-plot represented the energy supercapacitor [41].

In recent years, several abundant and environmentally benign biomass precursors have been successfully converted into 2D nanoporous carbon materials through efficient processing techniques (**Table 2**). For example, Alpinia galangal stem was effectively transformed into hierarchical porous carbon nanofibers through chemical activation using KOH [41], followed by high-temperature integrated pyrolysis for supercapacitor applications, as depicted in **Figure 3(a)**. The carbon material obtained demonstrated a high specific surface area of 1,065.58 m^2/g and a well-defined hierarchical nanofiber structure (**Figure 3(b)**), which provides abundant active sites and optimal pathways for ion transport at the electrode-electrolyte interface. Electrochemical performance was assessed using a 2-electrode system without binders, revealing typical cyclic and galvanostatic profiles that indicate the formation of a robust electric double layer on the electrode surface. The symmetric supercapacitor also showed a high specific capacitance of 226 F/g at a current density of 1 A/g, with a coulombic efficiency of 89 %. The energy output of the symmetric supercapacitor system was measured at 9.35 W/kg, with a power density of 96.04 W/kg (**Figure 3(c)**). However, when a 2D nanostructure is too dense, it can negatively affect porosity, blocking ions from accessing the working electrode surface during insertion and deinsertion.

Biomass-derived carbon synthesized from Indonesian green aromatic bay leaves [37] was used as precursors, with $ZnCl_2$ activator through a chemical impregnation process followed by a 1-step physical activation-carbonization strategy to produce carbon materials with 2D nanosheet structures, as shown in **Figure 4(a)**. This 2-step process effectively purges impurities from the carbon materials, while fine-tuning the ratio of activator to precursor enhances the morphology and properties of the resulting carbons. The surface modification successfully yielded 2D nanoporous structures, characterized by a high abundance of nanosheets. As illustrated in **Figure 4(b)**, the combustion and confined reaction of $ZnCl_2$ significantly altered the surface morphology of the 2D nanosheet precursors, with the walls becoming densely populated with pores, forming gauze-like structures. The numerous micro-mesopores on the nanosheet walls, generated by $ZnCl_2$ etching and the hydrolysis of lignin, cellulose, and volatiles during combustion, provided extensive transport pathways and interconnected active sites on the carbon materials. Furthermore, the obtained porosity texture, characterized by a surface area of 689 m^2/g with a distribution of 88 % micropores and 12 % mesopores, contributed to substantial improvements in

electrochemical behaviors. These improvements, beneficial for electrode materials, reduce electron transfer resistance and facilitate electrolyte diffusion to the inner surfaces of the carbon, potentially leading to enhanced electrochemical performance. When incorporated into a supercapacitor, the prepared carbon material electrodes demonstrated excellent electrochemical properties, as depicted in **Figure 4(c)**. The specific capacitance of the symmetric system was measured at 205 F/g with a current density of 1 A/g in a 1 M H_2SO_4 electrolyte. Moreover, the capacitance retention was 81.16 % at 10 A/g, with an optimal coulombic efficiency of 81.66 %. The symmetric supercapacitor device in aqueous electrolyte also achieved an energy density of 21.56 W/kg at a power density of 1.101 kW/kg.

The hierarchical connected 3D carbon structure facilitates rapid charge reversibility and establishes a sequential conductive network. This structure is closely tied to the diversity of pore types within carbon materials, which include micropores (pore width < 2 nm), mesopores (2 nm < pore width < 50 nm), and macropores (pore width > 50 nm) [42,43]. All 3 types of pores are interconnected in 3D space, allowing for comprehensive accessibility. Each pore type plays a specific role, specifically, micropores serve as the primary structure, providing abundant active sites for charge storage and the formation of electric double layers [44,45]. Mesopores help shorten the charge transport path, enhancing the accessibility and delivery of charges within the electrode [46]. Meanwhile, macropores function as reservoirs for electrolytes and improve interfacial wetting, which in turn reduces the resistance to ion transport [47]. By combining this strategy with 2D nanomodification, it is possible to create a fast-access architecture with high conductivity. A practical, economical, and sustainable approach using biomass is needed to achieve this combination.

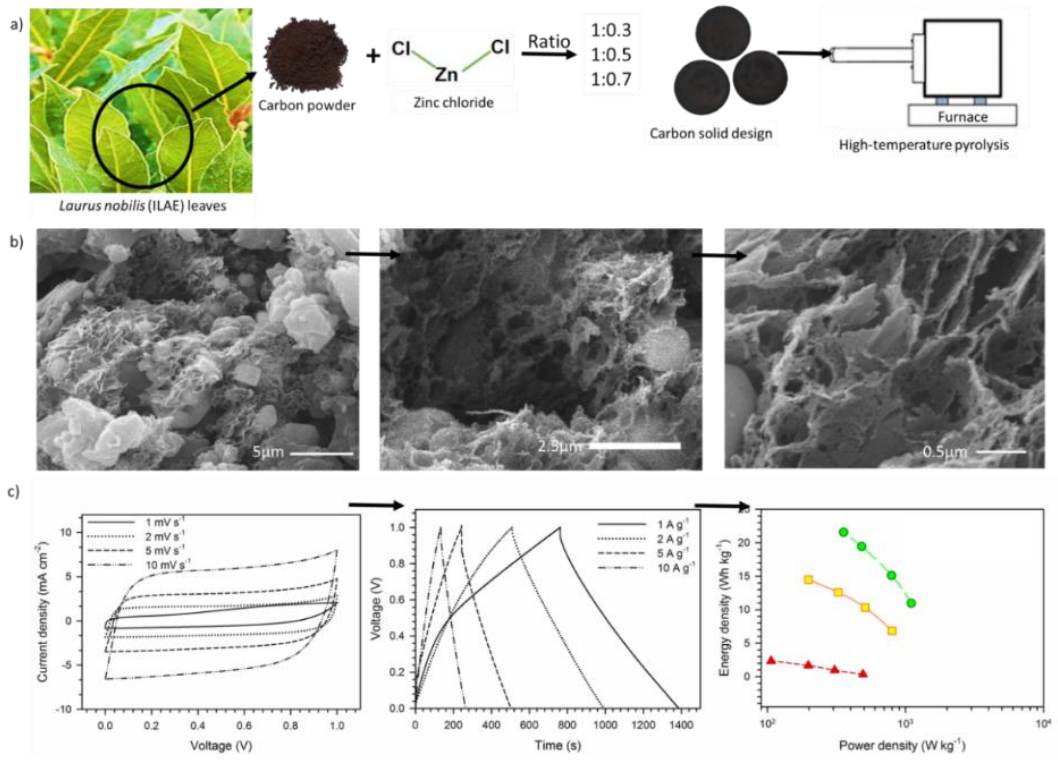

Figure 4 (a) The flow-preparation of carbon porous nanosheet 2D derived novel evergreen aromatic laurel (*Laurus Nobilis*)-based porous carbon, (b) The structure surface of carbon nanosheet 2D, and (c) electrochemical behavior of symmetric supercapacitor [37].

Table 1 Hierarchical 3D porous carbon derived biomass for supercapacitor application.

Sources	Activation technique	Morphology structure	S_{BET} (m^2/g)	Electrolyte	C_{sp} (F/g^{-1})	E_{sp} (W/kg)	P_{sp} (W/kg)	References
American poplar fruit	KOH activation	A tubular-like	942	6 M KOH	58.71	7.99	372	[48]
Porous organic polymers	KOH-CH_3 activation	Hierarchical porous	3,270	$EMIMBF_4$	184	92.2	950	[49]
Wood tar	Crab template	Hierarchical porous	2,489	6 M KOH	338.5	9.9	33.87	[50]
Grape marcs	N-doped	Hierarchical porous	2,221	1 M H_2SO_4	446.0	16.3	348.3	[51]
Waste shaddock	KOH activation	Hierarchical porous	1,265	1 M H_2SO_4	99	46.88	300	[42]
Peanut shell	N-doped	Hierarchical porous	2,014.6	6 M KOH	300.6	40.92	990	[52]
Cattail fiber	K_2CO_3 impregnation	Hierarchical porous	1,016	1 M $NaSO_2$	38.59	27.44	400	[53]
Wheat straw	KOH activation	Hierarchical porous	772	6 M KOH	226.2	-	-	[54]
Mangosteen peel	NaOH activation	Stone-like morphology	2,623	6 M KOH	114.8	51.6	12,00	[55]
Mandarin peels	KOH activation	Multiscale honeycomb	1,273	3 M KOH	341	10.92	240	[56]
Typha orientalis fiber	KOH activation	Bamboolike microfibers	2,792	6 M KOH	351	9.3	125	[57]
Laminaria japonica	KOH activation	Porous structures	1,902	6 M KOH	192	-	-	[58]
Osmanthus flower	KOH activation	3D framework	1,259	6 M KOH	255	7.95	10,000	[59]
Native European deciduous trees	Cavitation process	interconnected pores	614	1 M H_2SO_4	24	0.53	51	[60]
Chitin nanofiber	Bio-templates	hierarchically porous	876	6 M KOH	128.5	4.46	50	[61]

Table 2 nano-2D-porous carbon derived biomass for supercapacitor application.

Sources	Activation technique	Morphology structure	S_{BET} (m^2/g)	Electrolyte	C_{sp} (F/g^{-1})	E_{sp} (W/kg)	P_{sp} (W/kg)	References
Polyacrylonitrile (PAN) and novolac (NOC)	Electrospinning	Micro-meso nanofiber	1,468	2 M KOH	394	13.6	500	[62]
Prosopis Juliflora wood	1 step activation	3D hierarchical porous nanosheet	2,943	1 M $NaSO_2$	426	56.7	248.8	[63]

Sources	Activation technique	Morphology structure	S_{BET} (m^2/g)	Electrolyte	C_{sp} (F/g^{-1})	E_{sp} (W/kg)	P_{sp} (W/kg)	References
Jengkol shell	$ZnCl_2$ activation	Nanofiber	-	1 M H_2SO_4	220	50.56	110.11	[64]
Bacterial cellulose	Silica-assisted	Micro-mesopores nanofiber	624	6 M KOH	302	6.9	128.4	[65]
EDTA-Na2Zn	Template-assisted	Holey nanosheet	701.7	2 M KOH	205	17.92	500	[66]
Moringa oleifera	1-step pyrolysis	Hierarchically porous nanosheet	2,250	1 M Na_2SO_4	283	25.8	89	[67]
L-cysteine	NaCl template	Nanosheet	1,552	6 M KOH	363.1	13.4	325	[68]
Syzygium oleana leaves	1-stage integrated pyrolysis	Nanosheet	1,218	1 M H_2SO_4	188	26	96	[69]
Bamboo	Electrospinning	Nanofiber	1,386.1	6 M KOH	148	-	-	[70]
Chitosan of shrimp shell	Electrospinning	Nanofiber	564	2 M KOH	201	-	-	[71]

One effective method reported is a facile 1-way KOH impregnation strategy, which successfully synthesizes 3D hierarchical porous carbon from starfruit leaves [72]. The intense KOH reaction during high-temperature pyrolysis generates diverse pore structures with high connectivity, resulting in a surface area of 936 m^2/g and a micro:mesopore ratio of 4:1. In a 2-electrode system, this material demonstrated a capacitance of 293 F/g and output energy of 16.12 W/kg. Additionally, Ampong *et al.* (2024) synthesized hierarchical porous carbon from shea butter waste using a similar KOH activation method [73]. This remarkable structure achieved a surface area of 1233 m^2/g, which increased the output energy of the symmetric electrode by 12.6 W/kg. This device successfully powered an LED lamp for 1 h. Li *et al.* reported a significant surface area enhancement of up to 4,048 m^2/g for hierarchical porous carbon derived from lotus seedpods by using 2-stage KOH activation [74]. The presence of oxygen (O) functionalities also enhanced device performance, yielding a specific capacitance of 419 F/g at a current density of 1 A/g. These studies confirm that the KOH activation method is effective for creating 3D hierarchical structures in biomass-based carbon materials. Few other catalytic methods can compete with these results. Moreover, doping within the hierarchical structure tends to positively influence the electrochemical performance of supercapacitor.

A detailed examination of carbon synthesis through chemical activation, particularly using common activating agents like KOH and $ZnCl_2$, is essential. Understanding these activation processes is crucial for synthesizing porous carbon materials from biomass for supercapacitors. Each activation method results in distinct modifications to the carbon structure, which subsequently affects the electrochemical properties vital for supercapacitor applications. KOH activation is renowned for creating highly porous carbon structures, resulting in high carbon yields characterized by large surface areas and significant micropore volumes. This method promotes the development of a porous framework that optimizes ionic transport during electrochemical reactions, making the microporous structures beneficial for electrical double layer capacitance (EDLC). Conversely, the $ZnCl_2$ activation method typically yields hierarchical structures that combine micro- and mesoporous features. This dual porosity system greatly enhances ion diffusion compared to a purely microporous structure, leading to improved energy storage capabilities due to the easier accessibility of electrolyte ions. The carbon yields from these activation techniques can vary significantly. KOH

often produces higher yields than $ZnCl_2$ when using similar biomass precursors. This difference is attributed to KOH's stronger ability to dehydroxylate, which effectively breaks carbon-oxygen bonds and favors carbon structure formation. Studies indicate that KOH activation can attain specific surface areas (SSA) often exceeding 4,000 m^2/g, while $ZnCl_2$ activation typically achieves an SSA of around 1,000 m^2/g [75,76]. In addition, **Table 3** summarizes the comparison of the porosity, capacitance properties, and energy of biomass-derived carbon-based supercapacitors activated using KOH and $ZnCl_2$. Importantly, the pore structure generated through these activation methods directly influences the electrochemical performance of supercapacitors. KOH-activated carbons, with their dominant microporosity, maximize surface area but may restrict ion transport during rapid charge-discharge cycles, potentially resulting in lower power densities. In contrast, $ZnCl_2$-activated carbons, with their incorporation of mesopores, allow for improved ion mobility, enhancing rate capability. An example of this synthesis difference can be observed in lignin-derived carbons: KOH activation results in a structure dominated by micropores, ideal for achieving high energy densities. In comparison, $ZnCl_2$ activation produces a more complex pore network that supports better rate capabilities, optimizing both energy and power densities in supercapacitors.

Table 3 Comparing activation methods, porosity, and capacitance-energy values of carbon biomass for supercapacitor.

Carbon source	Chemical method	Porosity	Csp (F/g)	Esp (W/kg)	Ref
Coconut shell	KOH activation	2,143	317	13.5	[77]
Argan pulp for	KOH activation	3,235	460	15.98	[78]
Lotus seedpods	KOH activation	4,048	419	30.3	[75]
eucalyptus bark	KOH activation	1,719.15	483.5	21.7	[79]
loofah sponges	KOH activation	1,207.26	294	62.3	[80]
Acacia catechu Bark	$ZnCl_2$ activation	1,947	259.34	-	[81]
Coffee-Waste	$ZnCl_2$ activation	830	261	18.3	[82]
white chickpeas	$ZnCl_2$ activation	1,094.638	204.6	11.732	[83]
marigold flower	$ZnCl_2$ activation	1,274.45	179	23	[76]
Cotton	$ZnCl_2$ activation	1,990	173	12.5	[84]

Doping heteroatom in porous carbon biomass-derived materials

Biomass-based carbon is growing as a crucial component in supercapacitor development studies, with compelling advantages. The advantages include abundantly available raw material, diverse synthesis methods, and modifiable material structure with great flexibility, making biomass-based carbon an attractive option. The electrochemical properties are also promising but the performance faces challenges, particularly due to the limitations of conventional capacitive mechanisms based on electric double-layer capacitors (EDLCs) and the low output energy density in practical applications. The primary barrier to increasing the energy density of carbon-based supercapacitor is the dominance of pure physical charge storage mechanisms, which rely on conventional processes without Faradaic reactions. This limits the effectiveness of redox reactions in enhancing energy storage capacity. Although the capacitance of EDLCs is directly proportional to the electrode surface area, an excessive increase in surface area can have detrimental effects. This occurs because an increase in surface area potentially leads to a reduction in the proportion of mesopores, which in turn decreases charge carrier density. A large pore volume also hinders the efficiency of ion transfer, negatively impacting charge storage capacity. Micropores in carbon are often not fully accessible to aqueous-organic electrolytes, which limits the use of active sites during electrochemical processes.

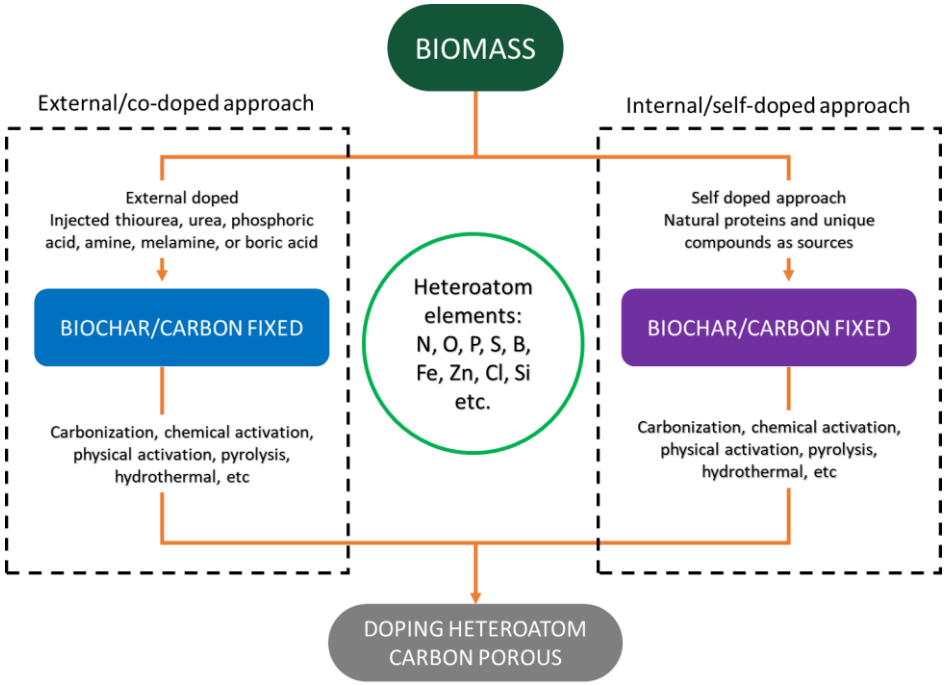

Figure 5 Biomass-based heteroatom-doped porous carbon preparation approaches.

Several reports have shown that specific capacitance tends to reach saturation at a certain surface area and exceeding this optimal level can lead to reduced specific capacitance [85,86]. Moreover, an excessive increase in surface area may also diminish the volumetric capacitance of the electrode, which ultimately has a negative impact on the high-power performance of supercapacitor [87,88]. Designing biomass-based carbon materials requires a balanced approach that considers specific surface area, pore distribution, and material structure to optimize specific capacitance while maintaining high-power performance. Strategies may include precise control of porosity properties, heteroatom doping to enhance electrode reactivity and a combination of physical and Faradaic charge storage mechanisms. One effective approach to overcoming these challenges is the introduction of heteroatoms into the carbon structure. The presence of these heteroatoms significantly influences the electronic, chemical, and mechanical properties of carbon by enhancing local reactivity [89]. Heteroatoms improve the wettability of the carbon surface, enhance electronic conductivity, and induce pseudocapacitance effects.

Commonly reported heteroatom elements include oxygen (O), nitrogen (N), sulfur (S), boron (B), phosphorus (P), silicon, and iron [90]. Porous carbons with heteroatom bonds can achieve specific capacitance enhancement through a synergy of EDLC energy storage mechanisms and pseudocapacitance behavior. The process of introducing heteroatoms into the carbon framework is known as doping, which can occur through external doping or self-doping. External doping involves adding heteroatoms to biomass carbon using agents such as thiourea, urea, phosphoric acid, amine, melamine, or boric acid [91-93]. This approach typically comprises several complex and time-consuming steps, including pre-carbonization, activation, and injection. However, it often results in well-defined improvements in wettability and high hydrophilicity, leading to significant energy output despite moderate surface areas. Self-doping is performed by directly carbonizing biomass, where the natural proteins and unique compounds present serve as a source of heteroatoms [94,95]. It yields heteroatom-doped carbon with high specific capacitance and superior rate capabilities, offering a more cost-effective, simple, and environmentally friendly alternative to external doping methods. Identifying biomass that naturally produces heteroatom bonds in the synthesized carbon framework can be challenging. The details of the external and self-doping approaches of biomass-derived carbon are illustrated in **Figure 5**.

Single-doped Heteroatom

The incorporation of heteroatom doping in porous carbon frameworks has attracted significant attention in the development of materials for supercapacitor. Heteroatom doping takes various forms, including single, dual, trinary, and multi-doped configurations, which can be produced through specific methods, as illustrated in **Figure 5**. Among these forms, single-doping with nitrogen (N) is one of the most extensively studied topics in the development of biomass-based carbon materials for supercapacitor. As shown in **Table 1**, N has several advantages over other heteroatoms including boron (B), phosphorus (P), and sulfur (S). The incorporation of N into the carbon structure can be achieved through a relatively straightforward method that is cost-effective and does not require complex additional treatments. This makes N the primary choice in the early development of biomass-based porous carbon materials. In contrast, the insertion of heteroatoms such as B, P, or S is often more challenging in biomass-based carbon and more typically applied to synthetic carbon materials, including spherical carbon derived from polymers or graphene aerogels. N contributes significantly to the enhancement of supercapacitor performance due to the unique properties. As a dopant, it improves the chemical reactivity and surface energy of the porous carbon framework without altering the fundamental characteristics [96]. The N doping process including substituting carbon atoms with N atoms, which contain 5 valence electrons compared to carbon's 4. This additional electron leads to an excess of electrons, which act as donors to the conduction band, thereby increasing the number of free electrons within the material [97]. Another advantage of N is the smaller atomic radius compared to carbon, allowing for higher dopant concentrations at the same substitution level. The higher electronegativity also enhances the adsorption of electrolyte ions, ultimately improving the material's wettability. Consequently, N serves as an ideal dopant for enhancing both the electrical conductivity and electrochemical properties of porous carbon materials.

The N doping approach for biomass-derived porous carbon materials is effectively achieved through external doping, specifically by injecting N-rich compounds during the pre-treatment phase. Li *et al.* [98] showcased the powerful combination of potassium hydroxide (KOH) activation and melamine injection to prepare N-rich carbon materials derived from lotus stalks. This method, conducted at a pyrolysis temperature of 550 °C, resulted in carbon with a remarkable porosity of 1,941 m^2/g. The presence of N heteroatoms significantly enhanced the capacitive performance, achieving an impressive 352 F/g and energy output of approximately 5.4 W/kg. Similarly, Shaku *et al.* [99] used a more advanced strategy with a higher pyrolysis temperature of 800 °C to develop N-doped carbon from Marula nutshell waste. This approach yielded a hierarchical pore structure with substantial N content, leading to an outstanding power output of 17.2 W/kg. The 3-dimensional (3D) interconnected hierarchical pore structure, combined with the faradaic reactions of N, vastly improved the performance of the working electrode. The integration of urea with KOH activation on biomass also delivers significant energy output in symmetric supercapacitor devices. In a study, rice husk waste was effectively converted into N-doped hierarchical porous carbon through the external doping strategy using urea and KOH injection [100]. Even with a surface area of only 416 m^2/g, the synergistic effects of the N-5 functional groups and a self-template 3D hierarchical structure elevated the energy density to 12.04 W/kg, with a capacitance of 242 F/g. Furthermore, volumetric scale evaluations demonstrated a remarkable volumetric capacitance of 306 F/cm^3. In another remarkable development, Gunasekaran *et al.* [101] achieved exceptional energy output results by combining 2-dimensional (2D) nanostructures with N heteroatom dopants. Carbon materials synthesized from bamboo wood waste using a KOH strategy followed by urea impregnation produced 2D nanosheet structures enriched with N co-doping. This led to an impressive energy output of 42 W/kg in a 2-electrode system.

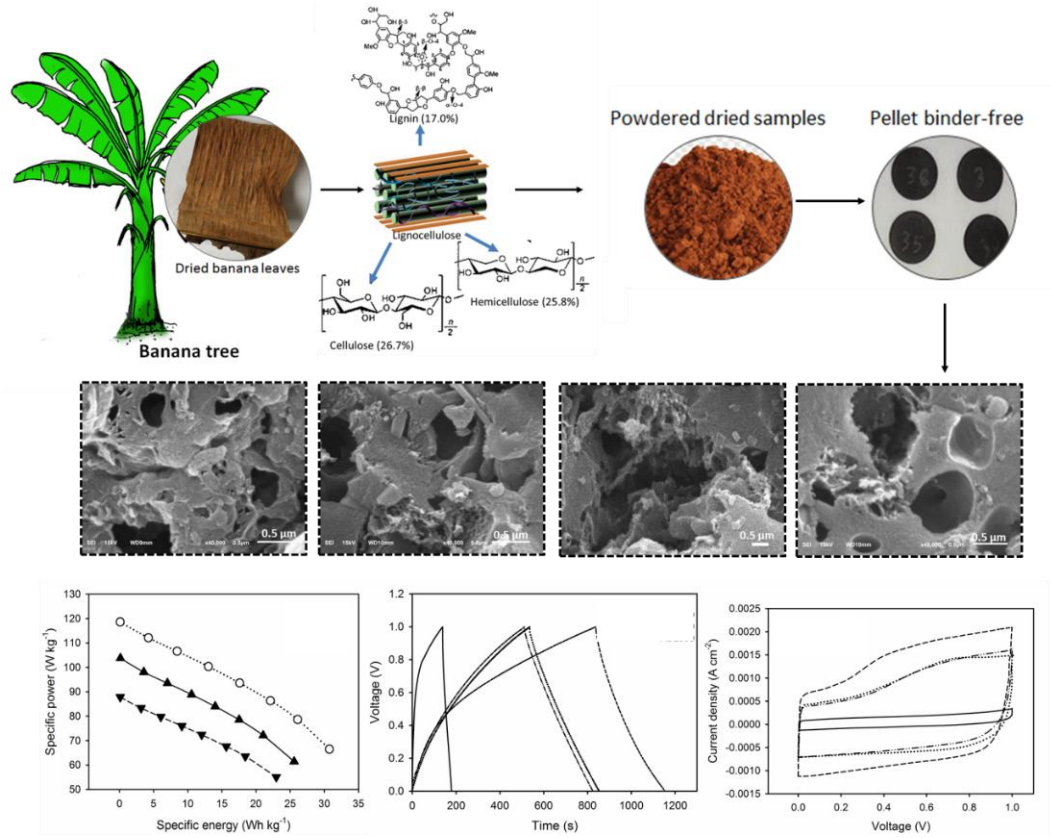

Figure 6 Banana leaves-derived carbon porous for oxygen self-doped for supercapacitor [102].

Self-doping strategies are making significant strides in introducing N heteroatoms into biomass-based carbon chains. For example, Li *et al.* [103] used Osmanthus fragrans biomass to generate N-doped carbon. The application of a KOH catalyst at a pyrolysis temperature of 800 °C resulted in an exceptional surface area exceeding 2,000 m^2/g and a capacitive performance of 351 F/g [103]. A similar method applied to eiderdown materials, which also contain high N potential, achieved a significant capacitance of 443 F/g in a 3-electrode system. However, the challenge of identifying solid biomass sources rich in N for carbon frameworks is evident. This represents a crucial opportunity for studies dedicated to enhancing the performance of biomass-based carbon materials for supercapacitor.

Oxygen (O) doping is one of the easiest strategies to apply when working with biomass-derived carbon materials. It is nearly impossible to synthesize solid biomass-derived carbons without the presence of O because the unsaturated carbon bonds tend to absorb moisture quickly. However, the content is often insufficient to make the carbon surface hydrophilic or to significantly enhance the pseudocapacitive properties [104]. Doped carbon materials generally have O levels that increase with the doping concentration within the carbon matrix. The functionalization has been extensively studied to improve the properties of carbon-based electrodes used in energy storage applications. It is important to differentiate between the terms “doping” and “functionalization.” “Doping” refers to the addition of small amounts of foreign elements (impurities) to a material to modify its properties. In contrast, “functionalization” requires adding functional groups to the surface of the material, usually through a different surface reaction process than conventional doping. This implies that the term “functionalization” is more appropriate when discussing the presence of O functional groups unless specifically addressing O doping or cyclic ether functional groups [105,106].

Common methods for hydrophilizing carbon surfaces and introducing controlled O content include chemical activation, physical activation, pyrolysis, and hydrothermal processes. Carbonyl-type oxygen groups (C=O) contribute significantly to the pseudocapacitance properties of porous carbon, as confirmed by extensive studies on various types of activated carbon. Studies

have shown a linear correlation between specific capacitance and the number of carbonyl-type surface functional groups (C=O) [107,108]. Carbonyl groups are known to provide greater pseudocapacitance than other groups, especially in acidic electrolyte media because each carbonyl can donate 2 electrons during redox reactions. Conversely, carboxylics (-COOH) are less thermally stable and do not easily undergo reversible processes, making the group less preferable than phenolic (C-OH) and carbonyl (C=O) [109]. Aside from the concentration of O and the configuration of specific functional groups, the choice of electrolyte media also plays a crucial role in determining the charge storage properties of the electrode. O functional groups tend to contribute higher pseudocapacitance in acidic media than in basic media. The redox reaction of O with acid electrolytes, such as H_2SO_4, is shown in the equations [110]:

$$O=C-O + H^+ + e^- \leftrightarrow -COOH \quad (1)$$

$$-C=O + H^+ + e^- \leftrightarrow -C-OH \quad (2)$$

Several studies have summarized the contribution of oxygen doping in enhancing the performance of biomass-derived carbon materials, which are detailed in **Table 4**. Most of these studies used a self-doping approach to prepare carbon materials, combined with chemical activation using catalysts such as KOH or zinc chloride ($ZnCl_2$). This method is based on optimizing the oxides produced during the carbon preparation from bio-organic waste. For instance, oxygen-doped carbon materials have been synthesized from banana leaf waste through a $ZnCl_2$ chemical activation process at pyrolysis temperatures ranging from 700 to 900 °C [111]. This chemical activation at varying temperatures leads to a well-defined 3D hierarchical pore structure. Increasing the temperature enables optimal etching of carbon chains, successfully creating interconnected 3D pore structures, as shown in **Figure 6**. The specific surface areas of these materials are below 1,000 m^2/g, but the electrochemical properties are remarkable. The presence of O dopants is confirmed in the carbon skeleton through carbonyl, carboxyl, and hydroxyl functional groups. Due to the synergy between the layer charge storage mechanism and the additional redox reactions of O dopants, these working electrodes demonstrate a specific capacitance of up to 401 F/g in a 2-electrode system. The output energy recorded in H_2SO_4 media reached an impressive value of 55.69 W/g.

Leaves wastes, rich in cellulose have shown the potential for O functionality in the carbon chain. Starfruit leaf was selected as the carbon source, processed using a KOH activation strategy at different concentrations [72]. The multistep synthetic route including KOH chemical impregnation, carbonization, and physical activation of starfruit leaf precursors has improved morphological structure and porosity, as shown in **Figure 7**. KOH impregnation enhanced the specific surface area by creating hierarchical porous structures on the carbon surface, clearly confirmed by the SEM images. The unique pore characteristics allow electroactive currents to flow smoothly, initiating high power for the electrode material. Furthermore, the perforated pore walls facilitate the movement of ions in a 3D space, enabling access to all possible active surfaces and promoting the formation of multiple electrical layers, which increases energy density. The reaction of KOH with the carbon chain also triggers the formation of functional groups such as carboxylate, hydroxyl, and carbonyl in the carbon material. This analysis is clearly presented in the FTIR spectra. The results confirmed that the carbon element constituted approximately 75 % of the material, while the O element accounted for around 10 - 14 %, showcasing the O functionality in the carbon derived from starfruit leaf. The synergy of the 3D hierarchical pores and O functionality led to a cyclic voltammetry (CV) profile that showed a camel-hump-like current spike (**Figure 7**). The combination of these features led to an increase in capacitive behavior to 293 F/g at a current density of 1 A/g in an acidic electrolyte. The recorded output energy was 26.5439 W/kg, achieved at a high-power density of 137.45 W/kg. Although the success of O functionalization in enhancing carbon structures is well recognized, there are significant limitations. Common issues include distributed capacitance, higher voltage drops, increased leakage current, low surface conductivity, inhibition of ion migration into micropores, and electrolyte decomposition. Corrosive oxidation also damages the carbon basal plane, leading to significant chemical and topological defects that ultimately degrade the performance of supercapacitor. Consequently, O-functionalized nanocarbon electrodes

often show poor rate capability and cycle life. To address these challenges, some studies intentionally removed O functional groups from the carbon surface to enhance electrode performance in supercapacitor applications.

Table 4 Single doped heteroatom carbon nanoporous derived biomass for supercapacitor application.

Biomass	Heteroatom	Structure	Doped strategy	Activation	Temperature pyrolysis	S_{BET} (m^2/g)	C_{sp} (F/g)	E_{sp} (W/kg)	Ref
Lotus stalks	N	Worm-like micropores	External doped with melamine	KOH	550 °C	1,941	352	4.5	[98]
Rice husks	N	3D Hierarchical pores	External doped with urea	KOH	800 °C	416	242	12.04	[100]
Bamboo wood	N	2D Nanosheet	External doped with urea	KOH	900 °C	769	296	42	[101]
Marula nutshell	N	3D Hierarchical pores	External doped with melamine	KOH	800 °C	1,427	248	17.2	[99]
Osmanthus fragrans	N	3D Hierarchical pores	Self-doped	KOH	800 °C	2,078	351	13.86	[103]
Eiderdown	N	3D Hierarchical pores	Self-doped	KOH	800 °C	3,101	443	11.9	[112]
Moringa oleifera leaves	O	Hierarchical pores 2D nanosheet	Self-doped	$ZnCl_2$	850 °C	372	201	25	[111]
Alpinia galanga stem	O	Hierarchical pores nanofiber	Self-doped	KOH	850 °C	1,065	226	9.3	[41]
Mission-grass	O	Block wall structure	Self-doped	$ZnCl_2$	850 °C	503	208	28.31	[113]
Banana leaves	O	3D Hierarchical pores	Self-doped	KOH	800 °C	860	401	55.69	[102]
Cassava peel	O	3D Hierarchical pores	Self-doped	$ZnCl_2$	850 °C	634	257	35.69	[114]
Averrhoa bilimbi leaves	O	3D Hierarchical pores	Self-doped	KOH	850 °C	936	293	26.54	[72]
Olive mill waste	Fe	3D Hierarchical pores	External doped with $Fe(NO_3)_3 \cdot 9H_2O$	KOH	800 °C	2,511	214	63	[115]

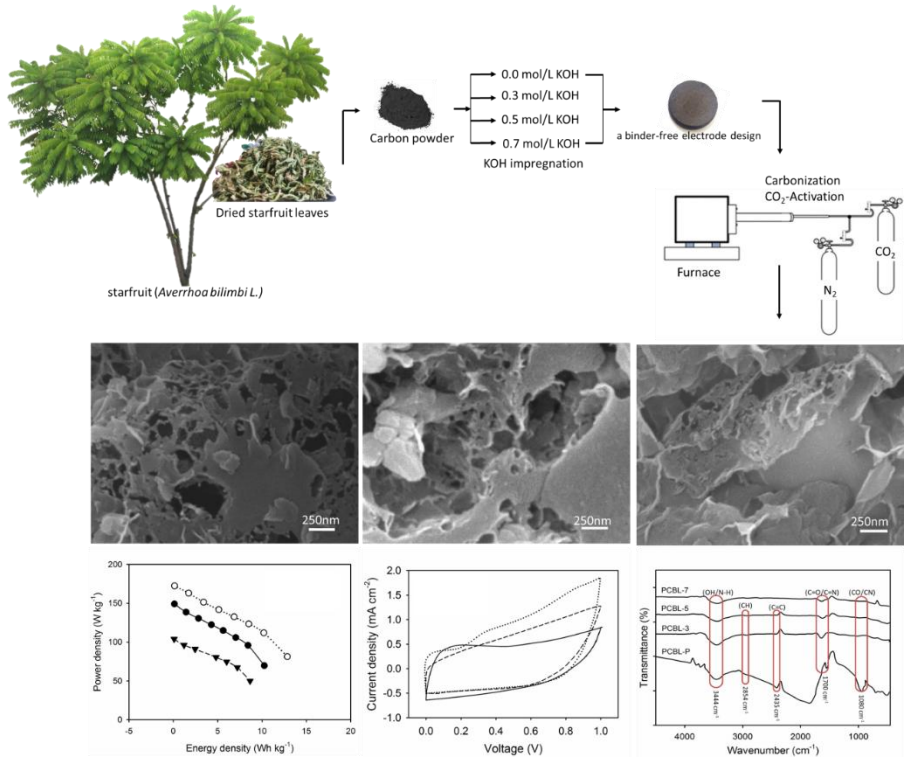

Figure 7 Starfruit leaves-derived carbon porous for oxygen self-doped for supercapacitor [72].

Dual-doped heteroatom

The electrochemical performance of carbon-based electrodes has been significantly enhanced through heteroatom doping strategies, specifically with elements such as nitrogen (N), oxygen (O), sulfur (S), boron (B), and phosphorus (P). These dopants play a crucial role in modifying the electronic and chemical properties of the carbon surface by introducing active sites that improve ion adsorption and enhance electrochemical redox reactions. The enhancement leads to substantial contributions to the apparent capacitance of the electrodes. For instance, N doping often increases the basicity of carbon, improves surface wetting by the electrolyte, and enhances electron transfer capacity due to the lone pairs of electrons present in amine or pyridine groups. In contrast, O doping typically introduces functional groups such as carbonyl or hydroxyl, which can participate in additional redox reactions and strengthen electrostatic interactions with ions in the electrolyte solution. Beyond single doping, some studies have also explored dual, trinary, and multi-doping strategies that combine 2 or more heteroatoms. These combinations produce synergistic effects that surpass the performance of single dopants. The effects stem from charge redistribution and modifications in spin density, which increase the density of electroactive sites and enhance the material electrical conductivity.

For example, the combination of N and B creates a unique electron attraction effect. B acts as an electrophilic element, increasing positive charge density, while N, a nucleophilic element, strengthens the structural stability of carbon, improving overall electrode performance [116]. The pairing of S and P not only strengthens the electronic conductivity of carbon but also expands the electrode surface area through pore expansion during synthesis. Although heteroatom doping can improve electrode performance, excessive or uncontrolled doping may damage the carbon structure, reduce the specific surface area, and inhibit ion diffusion, ultimately diminishing the specific capacitance of the material.

To address these challenges, a promising approach is by integrating heteroatom doping with nano-hierarchical pore engineering during the development of supercapacitor electrode materials. An optimal pore structure increases ion accessibility to active sites and facilitates faster ion diffusion, thereby improving energy storage efficiency. Designing a porous architecture that incorporates micro, meso, and macro pores strikes a balance between high surface area and efficient ion

transport. Micropores serve as the primary sites for electric charge storage through electrochemical capacitance, while meso and macro pores enhance ion diffusion into the electrode structure. Current studies focus on developing precise synthesis methods and controlled experimental conditions to maximize the synergistic effects of heteroatom doping and porous architecture design. This strategy aims not only to improve specific capacitance but also to ensure long-term cycling stability and high-power efficiency, making doped carbon-based supercapacitor promising candidates for future energy storage systems. Recent studies on dual heteroatom doping in biomass-derived carbon frameworks highlight their synergistic effects on structural modification, as summarized in **Table 5**.

Dual doping of B and N heteroatoms in biomass-based carbon frameworks is an effective strategy to enhance the electrochemical performance of supercapacitor. This approach takes advantage of the similarity in atomic radii between B and N, which allows for more stable intercalation within the carbon structure without causing significant distortion. The hydrothermal method has proven to be an effective technique for facilitating the diffusion of B and N atoms into the carbon matrix, thereby optimizing the material electronic configuration. Aside from improving electronic conductivity, B and N doping contributes to the formation of electrochemically active centers, which enhance both the pseudocapacitance effect and the electric double-layer capacitance.

For example, hierarchically porous carbon synthesized from sword bean biomass using a hydrothermal method in combination with pentaborate tetrahydrate impregnation demonstrates this principle [117]. The integration of external doping enables efficient attachment of B and N atoms to the carbon chain. Furthermore, chemical activation using potassium hydroxide (KOH) produces a hierarchical pore structure with a specific surface area of 2,471 m^2/g. This porous structure is crucial for supporting fast electrolyte diffusion and reducing ion transfer resistance, working in tandem with the pseudocapacitance effect of the B and N doping. With this optimal configuration, the carbon material achieved a high specific capacitance of 369 F/g at a current density of 1 A/g. In another study conducted by Zhang *et al.* [118], B- and N-doped carbon were used as electrode materials in a zinc-ion-based hybrid supercapacitor. The carbon was synthesized from coconut shell biomass through stepwise activation using urea and ammonium hydrogen borate trihydrate. The resulting material had a very high surface area of 2,770 m^2/g and a 3D hierarchical pore structure optimized for more efficient ion transport. The synergistic effects of B and N doping led to an increase in the density of surface-active centers, particularly in the form of N-based functional groups, enhancing electronic conductivity. The combination of high surface area, optimal heteroatom doping, and well-distributed pores makes this material an excellent candidate for supercapacitor cathodes, achieving an energy density of 139.46 W/kg.

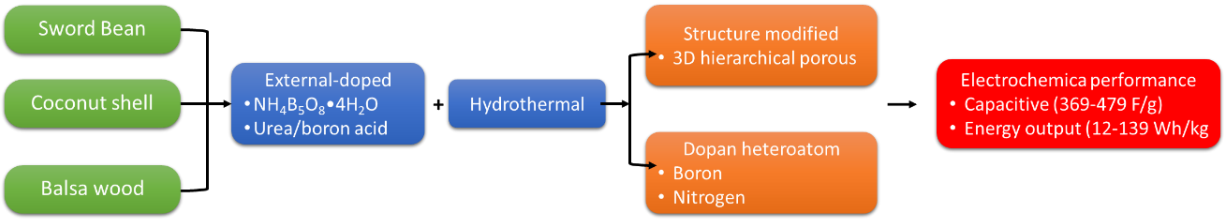

Figure 8 biomass-derived carbon porous with B/N external doped for supercapacitor.

Liu *et al.* [119] confirmed that the combination of heteroatom doping and pore structure engineering are key factors in improving the electrochemical performance of carbon-based electrodes. However, an excessive focus on 1 aspect can hinder the optimization of overall performance. The study demonstrated that effective co-doping of B and N could be combined with a trimodal pore structure through the re-engineered carbonization of natural wood. This synthesis process is related to the zinc acetate-assisted hypersaline pathway that allows simultaneous control of both the pore structure and the doping level. The resulting carbon achieved a specific surface area of 1,201 m^2/g, featuring a nanopore distribution resembling a trimodal foam.

This combination of traits facilitates a more efficient ion transfer pathway and significantly contributes to the pseudocapacitance effect induced by heteroatom doping. Consequently, the carbon-based electrode showed an impressive capacitance of 479 F/g, making it one of the highest-performing B- and N-doped carbon materials reported. The symmetric supercapacitor based on this material demonstrated high electrochemical stability, retaining over 90 % capacitance after 1,000 cycles, along with an energy density of 18.5 W/kg and a power density of 6.4 kW/kg. In summary, the strategy of dual heteroatom doping of B and N in carbon-based materials offers dual benefits including enhanced pseudocapacitance effect as well as improved ionic and electronic conductivity. This combination can be further optimized by engineering the pore structure to ensure efficient ion diffusion. Future studies should focus on understanding the doping mechanisms and the resulting electronic interactions, as well as refining the synthesis methods to improve the stability and performance of electrodes in high-energy supercapacitor applications. However, it remains challenging to find B and N elements independently doped in biomass-based carbon materials. **Figure 8** illustrates the external doping method used on biomass to achieve nitrogen and boron doping in carbon for supercapacitors.

Numerous studies decisively demonstrate that the co-doping of heteroatoms in nanoporous carbon materials produces significant synergistic effects, as shown in **Table 5**. These effects are clearly evidenced by the increase in active sites and the introduction of functional groups, which dramatically enhance the wettability of the carbon surface and improve the interaction between the electrode and electrolyte. In contrast, single heteroatom doping fails to achieve the superior electrochemical behavior associated with combined doping. Co-doping is characterized by a significant increase in structural defects, remarkable changes in specific surface area, and the formation of an optimal nanoporous structure that facilitates ion transport. Among the various heteroatoms studied, N and O play essential roles in enhancing the energy performance of biomass-based carbon supercapacitor. Both heteroatoms effectively enhance charge density, augment pseudo-capacitance contributions, and significantly improve the conductivity of carbon. The synthesis process for these advanced materials is remarkably straightforward, accomplished through co-doping or self-doping techniques that occur naturally during carbonization.

The impressive electrochemical performance of N and O-doped carbon materials derived from various biomass sources under different synthesis parameters has been extensively documented by numerous studies. For instance, Han *et al.* [120] successfully developed a carbon-heteroatom material based on grain waste from the refining industry, achieving significant advancements in supercapacitor performance. The activated carbon produced through self-doping with N and O showcases a hierarchical pore structure resulting from KOH activation at 600 °C. The combination of N (1.62 %) and O (19.27 %) in the carbon matrix greatly enhanced electrode-electrolyte interaction, yielding a specific capacitance of 345.2 F/g in a 3-electrode system. Moreover, this symmetric supercapacitor device showed remarkable cyclic stability, retaining 95.6 % of the capacitance after 5,000 cycles and achieving an outstanding energy density of 12.2 W/kg. Liu *et al.* [121] further intensified the exploration of external doping by introducing N and O heteroatoms into the carbon framework using *Perilla frutescens* biomass as a carbon precursor. Pyrolysis at 800 °C led to a 2D nanosheet structure with optimal pore connectivity. The controlled injection of urea during activation successfully yielded N (18.76 %) and O (1.70 %) heteroatoms, markedly enhancing both specific surface area and pore size distribution. This carbon-based electrode delivered an impressive specific capacitance of 287 F/cm^3 in volumetric testing, paired with an energy density of 13.9 W/kg in a symmetric supercapacitor configuration.

Chakraborty *et al.* [123] also made significant headway in modifying the nanosheet structure, successfully synthesizing N and O-doped carbon from turmeric leaf waste through KOH activation at a pyrolysis temperature of 900 °C [123]. Even with a relatively modest specific surface area of 542 m^2/g, the high heteroatom content, combined with the hierarchically interconnected nanosheet structure, produced remarkable capacitance and energy density, achieving an energy density of 39 W/kg - comparable to the performance of well-established commercial hybrid supercapacitor. Finally, Zhao *et al.* [124] applied a 1-step synthesis approach to generate N and O self-doped

carbon from soybean roots. This innovative carbonization and activation process produced a 3D hierarchical porous carbon structure characterized by a large specific surface area and optimal pore size distribution. The substantial N and O content significantly enhances conductivity and specific capacitance. Electrochemical testing confirmed a specific capacitance of 269.4 F/g, coupled with impressive capacitance retention of 97 % after 20,000 cycles. The assembled symmetric supercapacitor device operated at a voltage window of 2.0 V, achieving an energy density of 23.0 W/kg. Okonkwo *et al.* [125] also applied a similar method to castor shell and watermelon peel biomass to produce N and O self-doped carbon featuring a 3D hierarchical pore structure.

Nanofiber structures are essential for enhancing the performance of N and O-doped carbon-based supercapacitor. Recent studies clearly established that biomass from the snake plant is an excellent self-doped carbon precursor when activated with $ZnCl_2$ [122]. The naturally formed nanofiber structure, which eliminates the need for electrospinning techniques, significantly improves conductivity and charge storage capacity. Through a 1-step pyrolysis process, carbon nanofibers characterized by unique flower-like shapes and interconnected nano networks were obtained. This carbon material had a high mesopore ratio of 41 %, with impressive N (16.42 %) and O (4.35 %) content. In the symmetric supercapacitor configuration, this carbon-based electrode delivered a specific capacitance of 347 F/g and an energy density of 27.37 W/kg, clearly demonstrating the effectiveness of the nanofiber structure in enhancing the performance of solid-state supercapacitor. The process for their instant preparation technique is illustrated in **Figure 9**. Furthermore, studies on dual doping with N and O in biomass-based carbon are yielding outstanding results for developing superior electrode materials for supercapacitor. The combination of high heteroatom content, optimal pore structure, and an extensive specific surface area serves as crucial factors that drive increased capacitance and energy density in supercapacitor devices. As the current understanding of the doping mechanisms and synthesis parameters continues to advance, achieving increasingly competitive performance from biomass-derived carbon-based supercapacitor is of utmost priority. This effort is expected to provide sustainable solutions in the energy storage sector.

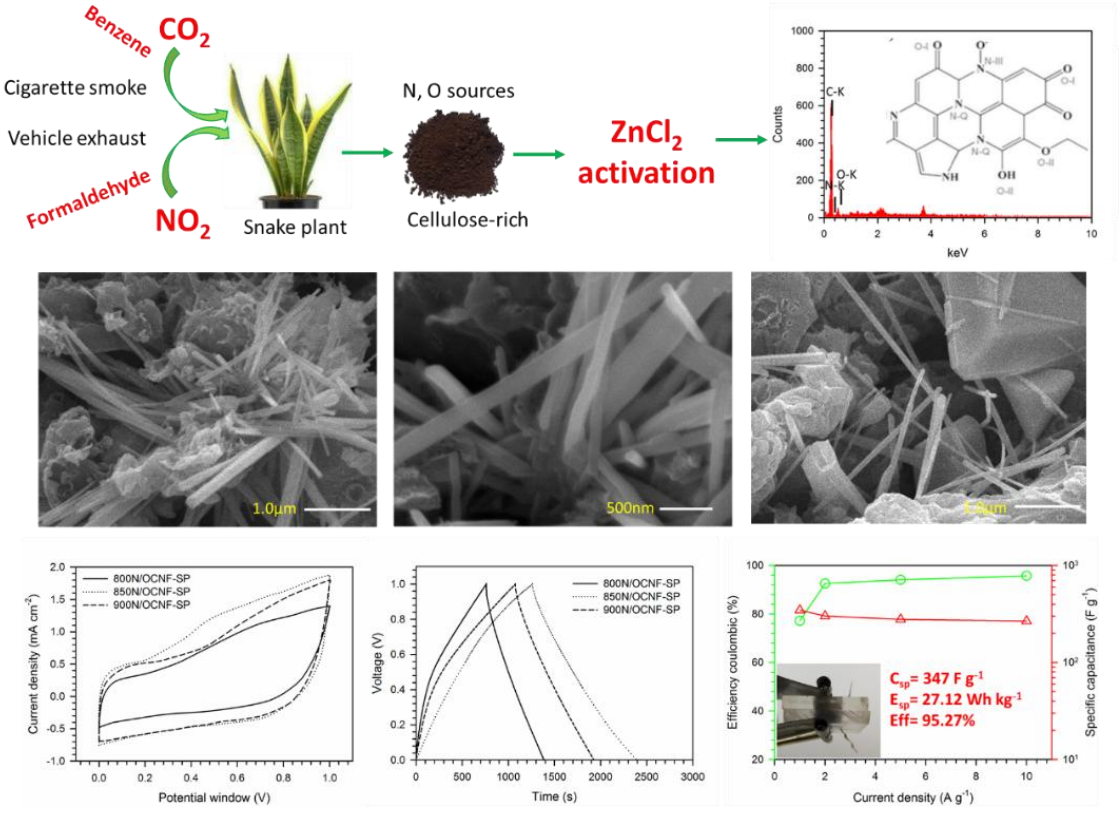

Figure 9 Snake plant-derived carbon porous with N/O-self-doped for supercapacitor [122].

N doping in carbon-based materials is a critical method for modifying electronic characteristics and significantly enhancing the electrochemical properties of supercapacitor electrodes. On the other hand, S doping offers distinct advantages due to the larger atomic size and easily polarized lone pair electrons, which greatly improve chemical reactivity and interactions with electrodes and electrolytes. The combination of N and S double doping has proven to substantially increase the number of active sites and open ionic channels in carbon materials, leading to remarkable improvements in both capacitance and electrochemical stability. In the study by Li *et al.* [126], N and S-doped carbon were synthesized from coal tar pitch using ammonium sulfate as the dopant source. The application of KOH activation produced a bubble-like carbon structure featuring a thin porous shell and optimized pore distribution. The resulting carbon demonstrated an impressive specific capacitance of 368 F/g at a current density of 0.5 A/g, along with outstanding cyclic stability. This study unequivocally established the proposed synthesis strategy as a reliable method for producing high-performance electrode materials suitable for large-scale production at reduced costs.

Rustamaji *et al.* [127] adopted a similar approach, developing N and S-doped carbon from palm oil residue using ammonium persulfate for impregnation and $CaCl_2$ for activation. The carbon material obtained showed a substantial increase in porosity, nearly doubling the initial value, and achieved a specific capacitance of 173 F/g. The study underscores the essential potential of using biomass waste to produce activated carbon containing double heteroatoms for supercapacitor applications, emphasizing both sustainability and efficiency as key benefits. Self-doping of N and S has been successfully demonstrated in various studies. For instance, Wu *et al.* [128] synthesized carbon from bacterial cellulose using the KOH activation method, resulting in a highly porous structure with a remarkable surface area of 916.4 m^2/g and interconnected pores in a 3D hierarchical arrangement. This innovative structure allowed the electrode to achieve a specific capacitance of up to 360 F/g and an energy density of 26.8 W/kg. The combination of high porosity and optimal heteroatom doping solidifies the contribution to capacitance, accelerating ion transfer kinetics and significantly enhancing overall supercapacitor performance.

Studies into S and O doping in carbon materials derived from other biomass sources have yielded compelling results. A previous study used mangosteen peel as a carbon precursor, utilizing KOH activation combined with double gas pyrolysis to produce carbon nanofibers doped with both S and O [129]. This material showed a unique wormhole-like structure with an impressive surface area of 962.4 m^2/g and an optimal micropore to mesopore ratio distribution of 1:1. High doping content of S (7.25 %) and O (20.53 %) significantly improved the electrochemical properties of the material, yielding a specific capacitance of 231 F/g at 1 A/g, a coulombic efficiency of 98 %, and low internal resistance. The significant faradaic effects, alongside a pseudocapacitance contribution of 16 %, enabled the symmetric supercapacitor device to achieve an energy density of 13.3 W/kg at a power output of 393 W/kg, as illustrated in Figure 10. These studies clearly confirm that double heteroatom doping, whether through combinations of N and S or S and O, substantially enhances the performance of biomass carbon-based supercapacitor. The doping strategy not only improves conductivity and surface charge density but also optimizes ion transfer kinetics through modifications in pore structure and enhanced active functional groups. The recent advancements in synthesis strategies demonstrate a strong potential for effectively optimizing carbon structures, as leading candidates for future advanced energy storage applications.

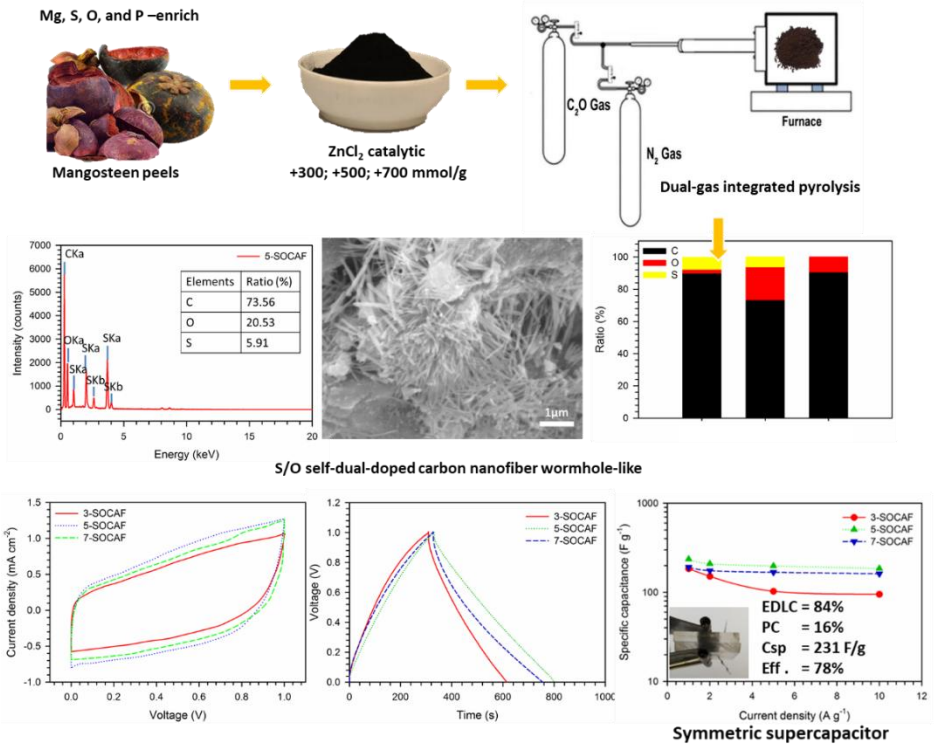

Figure 10 Mangosteen peels-derived carbon porous with S/O-self-doped for supercapacitor [129].

P and O doping in biomass carbon has been proposed as an innovative method to enhance the electrochemical performance of electrode materials used in supercapacitor. Specifically, P plays a crucial role in improving the electronic conductivity of carbon and stabilizing the porous structure. When P is incorporated into the carbon network, it creates more reactive active sites, which can increase the charge transfer rate during energy storage. P also enhances the structural stability of carbon during charge and discharge cycles, thereby extending the lifetime of supercapacitor. The interaction between P and other elements in the carbon material modifies the electron density levels, ultimately improving overall electrochemical performance. Meanwhile, O doping significantly enhances pseudo-capacitance by forming O functional groups, such as carbonyl, hydroxyl, and epoxide, on the carbon surface. These functional groups enhance the wettability of the electrode, facilitating electrolyte penetration and increasing the specific capacitance of the carbon material. O also promotes additional redox reactions that enhance charge storage capacity. The presence in the carbon structure also improves the thermal and chemical stability of the electrode material, reducing degradation during supercapacitor cycling. The synergistic interaction between P and O doping in porous carbon materials leads to increased specific capacitance and higher cycle stability compared to single doping. The combined effects create an optimal balance between enhanced electronic conductivity and pseudo-capacitance improvements. The synthesis of these materials is relatively simple, as it can be achieved through co-doping or self-doping techniques that naturally occur during carbonization.

Various studies have reported the electrochemical performance of P and O-doped carbon materials derived from different biomass sources, with variations in synthesis parameters. For instance, using banana stem biomass, fibers were extracted as precursors to create P and O self-doped carbon [130]. This process includes synthesizing these precursors into nanosheet carbon materials with a well-defined pore structure, enhanced with O and P doping. The production process includes particle crushing and high-temperature heating, with the addition of 500 mmol/g KOH as a reaction agent and carbon structure developer. The resulting material had micro-mesoporous structures, and the working electrode achieved a remarkable specific capacitance of 206.76 F/g, primarily due to double doping with O (24.15 %) and P (4.37 %), alongside contributions from

pseudocapacitance. These results illustrate the significant potential of O- and P-doped multi-heteroatom carbon nanosheets from banana stem fibers in developing high-energy density and high-power supercapacitor materials. **Figure 11** illustrates the preparation steps for dual-doped carbon from banana stem fiber, along with the resulting structural modifications and electrochemical behavior. Alternatively, an external doping approach to introduce P and O dopants into carbon materials has been reported by Chen *et al.* [131]. In this study, external doping using H_3PO_4 and hydrothermal processing converted cicada slough waste into connected micro-mesoporous carbon for supercapacitor electrodes. The carbon material was confirmed to contain a remarkable amount of P and O heteroatoms, which significantly enhanced the electrochemical performance of the supercapacitor, yielding a specific capacitance of 295 F/g and 100 % stability after 10,000 cycles.

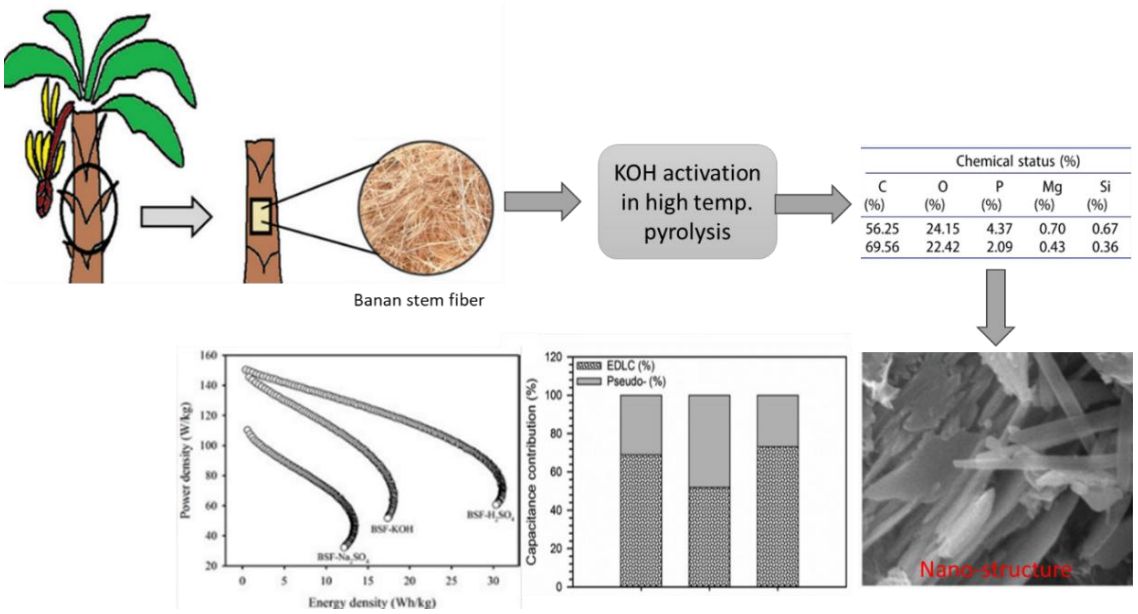

Figure 11 Banana tree fiber-derived carbon porous with P/O-self-doped for supercapacitor [130].

Table 5 Dual doped heteroatom carbon nanoporous derived biomass for supercapacitor application.

Biomass	Hetero atom	Structure	Doped strategy	Activation	Temp. pyrolysis	S_{BET} (m^2/g)	C_{sp} (F/g)	E_{sp} (W/kg)	Ref
Sword bean	B, N	3D hierarchical porous	External doped with ($NH_4B_5O_8{\cdot}4H_2O$)	Hydrothermal	500 °C	2,471	369	12	[117]
Coconut shell	B, N	3D hierarchical porous	External doped with urea and $NH_4HB_4O_7{\cdot}3H_2O$	-	800 °C	2,770	-	139.46	[118]
Balsa wood	B, N	3D hierarchical porous	External doped with urea/boron acid	Hydrothermal	500 °C	1,201	479	18.5	[119]
Perilla frutescens	N, O	2D nanosheet	External doped with urea	-	800 °C	655	270	13.9	[121]
Distiller ‘s grains	N, O	3D hierarchical porous	Self-doped	KOH activation	600 °C	1,026.77	345	12.2	[120]

Biomass	Hetero atom	Structure	Doped strategy	Activation	Temp. pyrolysis	S_{BET} (m^2/g)	C_{sp} (F/g)	E_{sp} (W/kg)	Ref
Turmeric leaves	N, O	Porous carbon nanosheet	Self-doped	KOH activation	900 °C	541	310	39.44	[123]
Soybean root	N, O	3D hierarchical porous	Self-doped	KOH activation	850 °C	1,801	269.4	23.0	[124]
Snake-plant	N, O	2D nanofiber	Self-doped	$ZnCl_2$ activation	850 °C	785.107	347	27.37	[122]
Watermelon peel	N, O	3D hierarchical porous	Self-doped	NaCl-KCl activation	800 °C	1,660	278	25.4	[132]
Castor shell	N, O	3D hierarchical porous	Self-doped	KOH activation	800 °C	1,527	365	9.14	[125]
Banana stem fiber	P, O	2D nanosheet	Self-doped	KOH activation	850 °C	326.35	206.76	31.13	[130]
Cicada slough	P, O	meso/microporous channels	External doped with H_3PO_4	Hydrothermal	500 °C	1,945	295	12.5	[131]
Coal tar pitch	N, S	Graphene like	External doped with ammonium sulfate	KOH activation	800 °C	2,326.3	368	-	[126]
Palm oil residue	N, S	-	External doped with ammonium persulfate	$CaCl_2$ activation	800 °C	440.24	173.91	5.11	[127]
Bacterial cellulose	N, S	3D hierarchical porous	Self-doped	$ZnCl_2$ activation	700 °C	916.4	360	26.8	[128]
Mangosteen peel	S, O	2D nanofiber	Self-doped	$ZnCl_2$ activation	850 °C	962.415	231	13.3	[129]
Mangosteen peel	Zn, O	2D nanorod-like	Self-doped	$ZnCl_2$ activation	850 °C	670.569	261	13.4	[133]

Triple and multi-heteroatom doping

Triple and multi-heteroatom doping leads to a more asymmetric spin and charge density on carbon atoms compared to single or dual heteroatom doping. Various studies have shown that carbon materials doped with more than 2 heteroatoms show remarkable performance in supercapacitor. Doping with high concentrations of heteroatoms such as nitrogen (N), oxygen (O), phosphorus (P), and sulfur (S) enhances wettability and conductivity while improving electrochemical performance through pseudocapacitance facilitated by Faradaic reactions. Although the strategies for external doping are complex, and finding biomass capable of producing triple or multi-doping through self-doping is challenging, the electrochemical results are impressive. This review aimed to compile relevant information to support the development of effective and efficient energy storage technology, as detailed in **Table 6**. For example, supercapacitor devices prepared using various methods applied to mixed biomass of algae and bamboo produced heteroatom dopants N, O, and S, as reported by Yue *et al.* [134]. The study outlined 3 distinct approaches to achieve heteroatom doping in carbon materials derived from mixed biomass. The first approach comprises activation using potassium acetate

(CH_3COOK), where heteroatom dopants were independently derived from spiral algae rich in N and S. Pyrolysis of the spiral algae and bamboo at 800 °C successfully introduced heteroatom functional groups into the microstructure of the carbon materials, leading to a new synthetic route. The resulting concentrations of N, O, and S were 2.03, 12.36 and 0.34 %, respectively. The activation treatment with potassium acetate at high-temperature pyrolysis produced a 3D hierarchical porous surface structure which had a surface area of 1,285.15 m^2/g, beneficial for high performance. Consequently, the gravimetric capacitance reached 320 F/g at a current density of 0.5 A/g in a 3-electrode system and 236 F/g at 0.25 A/g in a symmetrical device system. The energy output was recorded at 8.21 W/kg at an output power of 62.5 W/kg.

In a subsequent report, Yue *et al.* [135] used CH_3COOK and calcium acetate ($Ca_2(C_2H_5COO)_2$) as chemical catalysts in pyrolysis at 700 °C to obtain self-doped carbon materials composed of N, O, and S. The results showed a change in surface morphology to a nano-flower-like structure with a specific surface area of 1,171 m^2/g. The ratios of N, O, and S were found to be 2.34, 16.63 and 0.26 %, respectively. Despite the lower surface area and slightly reduced S ratio, the capacitance increased to 324 F/g at 0.5 A/g, with an energy output reaching 14.67 W/kg. This enhancement is attributed to the synergy between the nano-flower-like structure and the activity of the heteroatom functional groups, resulting in high conductivity and exceptional charge insertion accessibility in the electrode materials. Using potassium hydroxide (KOH) as a chemical catalyst on the same materials produced a remarkable surface area of 2,421.85 m^2/g along with a 3D interconnected pore-rich structure. The ratios of the dopant elements were 1.82, 10.90 and 0.23 % for N, O, and S, respectively [136]. The capacitive properties significantly improved, reaching 460 F/g, while the energy output increased to 19.09 W/kg. These 3 comprehensive reports by Yue *et al.* [136] confirm that the combination of N- and S-rich spiral algae biomass with porous bamboo is a high-potential raw material for developing environmentally friendly and sustainable heteroatom doping technology.

Other studies reported significant results, for example, Liu *et al.* [137]; Li *et al.* [138] synthesized self-doped porous carbon containing N, O, and S from lotus leaves and kelp biomass through KOH activation during pyrolysis at 800 °C. Lotus leaf biomass, which retains water, can absorb S- and N-containing compounds from water and nutrients, enabling it to present N and S functional groups in carbon materials. Through KOH activation in pyrolysis at 800 °C, the carbon material derived from lotus leaves showed a hierarchical pore structure with an exceptionally high surface area of 3,601 m^2/g, along with well-defined N, O, and S functional groups. The capacitive properties were measured at 294 F/g in a symmetrical device system. The kelp biomass showed a similar structural quality, with a surface area of 1,178 m^2/g and the presence of N, O, and S functional groups. The actual capacitive values recorded for this material were 276 F/g in a 3-electrode system and 166 F/g in a practical device. Additionally, industrial residues, especially semi-cooked materials, demonstrated potential for self-doping with N, O, and S heteroatoms for supercapacitor applications. This was achieved through activation with Na_2CO_3 during pyrolysis at a temperature of 700 °C, as detailed by Wang *et al.* [139]. Although the porosity of these materials was relatively low at 718 m^2/g due to the chosen carbon source, the capacitance properties were still competitive, reaching 170 F/g, with an output energy of 5.92 W/kg. This indicates that trial doping with heteroatoms can be effectively produced using selected biomass to develop self-doped heteroatom-based supercapacitor.

A trial study on doping using heteroatoms N, O, and P was conducted independently, using biomass from nutmeg leaves for supercapacitor applications, as shown in **Figure 12** [38]. The 1-step activation strategy was implemented using KOH as a catalyst, followed by carbonization and activation in an N_2-CO_2 gas environment. The synthesis method is shown in **Figure 8(a)**. The modification of the surface structure led to the development of dense hollow nanofibers, indicating the presence of a rich nano-hollow-fiber network, as shown in **Figure 8(b)**. This structure facilitates rapid deliverability and excellent conductivity due to density. The ratios of the dopant components N, O, and P were found to be 18.50, 17.19 and 3.07 %, respectively. The incorporation of these heteroatoms significantly contributed to reversible Faradaic reactions, providing donor electrons at the terminal/electrolyte interface and initiating induced pseudocapacitance. Moreover, these enhancements improved both the surface contribution

and ion diffusion of the materials, leading to an almost 3-fold increase in capacitive properties, reaching 235 F/g at a current of 1 A/g in a dual-electrode system, as depicted in **Figure 8(c)**. The capacitance retention was impressive, reaching 83.46 % at 10 A/g. The final electrode showed the highest recorded energy output of 32.01 W/kg, along with a maximum power output of 418.78 W/kg. These results surpass those of several previous studies using similar symmetric test systems.

An external doping approach to incorporate N, O, and P heteroatoms into porous carbon materials was reported by Wang *et al.* [140]. The study substituted N, O, and P in carbon materials derived from peach gum through external doping with $(NH_4)_2HPO_4$, while surface modification was focused on inducing $Mn(CH_3COO)_2$ during pyrolysis at 700 °C. The electrode based on a hybrid manganese combined with N, O, and P-doped carbon demonstrated a remarkable capacitance of 490 F/g and an impressive energy output of 76.87 W/kg. Furthermore, an external doping approach that comprised applying a mixture of materials has successfully demonstrated doping experiments on carbon chains derived from organic waste. For instance, external doping with porogen and H_3BO_3 was used to convert organic laver waste, resulting in the introduction of heteroatoms N, O, and B into the porous carbon framework [141]. To optimize the electrochemical performance of these carbon materials, activation was carried out using $KCl/ZnCl_2$ followed by pyrolysis at 800 °C. Consequently, the optimal carbon materials showed a 3D hierarchical pore structure, achieving a porosity of 1,514.3 m^2/g. The combination of N, O, and B doping, along with the hierarchical structure, has led to remarkable electrochemical performance. The optimal carbon-based electrode recorded a capacitance of 382 F/g in an aqueous electrolyte. The performance in a 1 M $BMIMBF_4$/AN organic electrolyte was also impressive, yielding an output energy of 51.3 W/kg. Moreover, external doping using thiourea, melamine phosphate, and boric acid introduced N, P, S, and additional N, P, and B dopants in hierarchical porous carbon materials, as detailed in studies by Yang *et al.* [93]; Yang *et al.* [142]. Additional strategies were implemented to enhance the porosity of original materials such as bamboo pulp fiber and pomelo peel through NaOH and KOH activation at high-temperature pyrolysis. The best-performing carbon electrodes showed capacitance ranging from 183 to 316 F/g, with excellent retention after 10,000 cycles.

Many studies expressed concerns that the proposed external doping methods require overly complex and costly chemical compounds, which limits practicality. Self-doping methods for introducing heteroatoms may be more favorable due to the simplicity and flexibility in preparing original materials. For instance, *Cymbopogon citratus* leaves were reported in 2023 to show a rich nanofiber structure using a KOH catalyst [143]. The carbon framework contained O, P, and S dopants in defined ratios of 8.53, 1.13 and 0.86 %, respectively. The symmetric electrode demonstrated a high capacitance of 293 F/g with an output energy of 39.21 W/kg in an aqueous H2SO4 electrolyte. The detailed Faradaic reactions occurring on the carbon electrode in this acidic environment are described in the equations.

The redox reactions of N-heteroatom in H_2SO_4 electrolyte:

$$N + H^+ + e^- \leftrightarrow N\text{-}H \quad (3)$$

$$N\text{-}O + 2H^+ + 2e^- \leftrightarrow N + H_2O \quad (4)$$

The redox reactions of P-heteroatom in H_2SO_4 electrolyte are as follows:

$$-P{=}O + 2e^- + 2H^+ \leftrightarrow -H\text{-}P\text{-}OH \quad (5)$$

The redox reactions of B-heteroatom in H_2SO_4 electrolyte are as follows:

$$*C\text{-}B\text{-}C{=}O + 2H^+ + 2e^- \leftrightarrow *C\text{-}B\text{-}CH\text{-}OH \quad (6)$$

The redox reactions of S-heteroatom in H_2SO_4 electrolyte are as follows:

$$R\text{-}(O{=}S{=}O)\text{-}R' + 2H^+ + 2e^- \leftrightarrow R\text{-}SO\text{-}R + H_2O \quad (7)$$

$$R\text{-}SO\text{-}R + 2H^+ \leftrightarrow R\text{-}SH(=OH)\text{-}R' \quad (8)$$

$$R\text{-}(O=)S\text{-}R' + H^+ + e^- \leftrightarrow R\text{-}SH(\text{-}O//H^+)\text{-}R' \quad (9)$$

Multi-doping with N, O, P, and S was reported by Lu *et al.* [144]. The study discovered a rich presence of

heteroatoms in rapeseed meal biomass, activated using KOH at 800 °C. The surface structure featured abundant 3D hierarchical pores, achieving a maximum porosity of 3,291 m^2/g. This large surface area, supported by hierarchical pores, enables the carbon material to show an exceptional dual-layer electric charge storage mechanism. The impressive Faradaic reactions of the 4 heteroatoms working simultaneously contributed significantly to the capacitive behavior of the supercapacitor, with diffusion control accounting for 38 % of the total capacitive contribution of the working electrode. Consequently, this enhanced the capacitive performance of the supercapacitor to 460 F/g in aqueous electrolyte, producing an output energy of 36.22 W/kg. The discussions above indicate that each approach to introducing heteroatoms into the carbon framework has distinct advantages and disadvantages. A measurable target is essential to ensure that the preparation approach for heteroatom-doped carbon aligns effectively with the future development of energy storage devices.

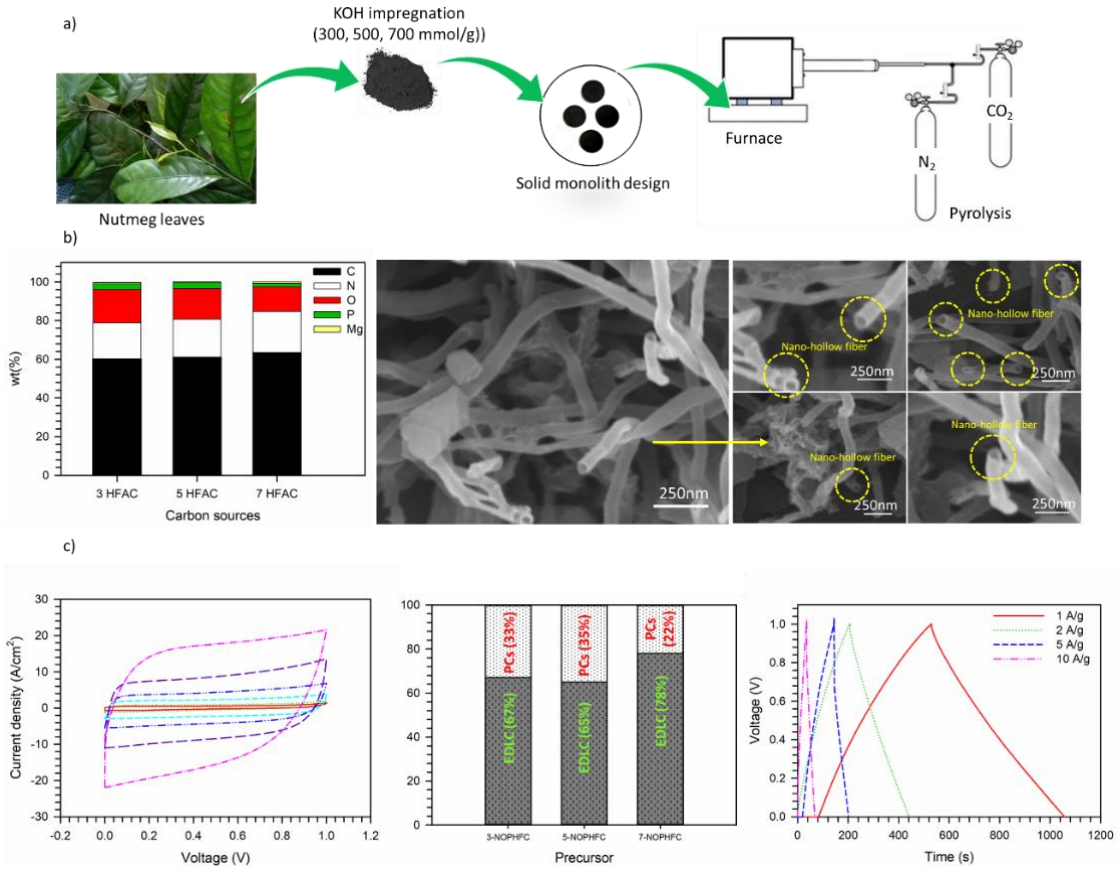

Figure 12 Nutmeg leaves-derived carbon porous with N/O/P-self-doped for supercapacitor [38].

Table 6 Multi-doped heteroatom carbon nanoporous derived biomass for supercapacitor application.

Biomass	Hetero atom	Structure	Doped strategy	Activation	Temp. pyrolysis	S_{BET} (m^2/g)	C_{sp} (F/g)	E_{sp} (W/kg)	Ref
Spiral algae and bamboo	N, O, S	3D hierarchical porous	Self-doped	CH_3COOK activation	800 °C	1,285.15	320	8.21	[134]
Spiral algae and bamboo	N, O, S	Nano-flower-like	Self-doped	CH_3COOK and $Ca_2(C_2H_5COO)_2$ activation	700 °C	1,171	324	14.67	[135]

Biomass	Hetero atom	Structure	Doped strategy	Activation	Temp. pyrolysis	S_{BET} (m^2/g)	C_{sp} (F/g)	E_{sp} (W/kg)	Ref
Spiral algae and bamboo	N, O, S	3D hierarchical porous	Self-doped	KOH activation	500 °C	2,421.85	460	19.09	[136]
Semi-coking wastewater	N, O, S	Porous material	Self-doped	Na_2CO_3 activation	700 °C	718	170	5.92	[139]
Kelp	N, O, S	3D hierarchical porous	Self-doped	KOH activation	800 °C	1,178	166	5.76	[138]
Lotus leaves	N, O, S	3D hierarchical porous	Self-doped	KOH activation	800 °C	3,601	294	-	[137]
Nutmeg leaves	N, O, P	Hollow-nanofiber	Self-doped	KOH activation	850 °C	418.39	235	32.64	[38]
Peach gum	N, O, P	3D hierarchical porous	Extternal doped with $(NH_4)_2HPO_4$	$Mn(CH_3COO)_2$ activation	700 °C	1,635	490	76.87	[140]
Rapeseed meal	N, O, P, S	3D hierarchical porous	Self-doped	KOH activationa	800 °C	3,291	416	36.22	[144]
Laver	N, O, B	3D hierarchical porous	External doped with porogen and H_3BO_3	$KCl/ZnCl_2$ activation	800 °C	1,514.3	382	29.2	[141]
Bamboo pulp fiber	N, P, S	Interconnected loose	External doped with thiourea & melamine phosphate	NaOH activation	850 °C	1,776	316	16.3	[93]
Pomelo peel	N, P, B	3D hierarchical porous	External doped with melamine, boric acid and phosphoric	KOH activation	800 °C	2,106.27	183	-	[142]
Peronema canescens leave	O, Zn, Cl	Interconnected pores	Self-doped	$ZnCl_2$ activation	850 °C	145.721	231	12.81	[145]
Cymbopogon citratus leaves	O, P, S	Nanofiber	Self-doped	H_3PO_4 activation	850 °C	629.40	239	39.21	[143]

Recent discussions on various doping strategies, including self-doping and external doping techniques, have highlighted significant improvements in electrochemical performance. A comparative analysis of these methods reveals their respective advantages and challenges, which are crucial for optimizing the characteristics of carbon electrodes while achieving high performance. Understanding the role of different heteroatoms, particularly the synergy between combinations such as nitrogen, oxygen, phosphorus, boron, and sulfur, provides valuable insights for tailoring materials used in supercapacitors. Self-doping, where heteroatoms are derived from the biomass precursor itself, typically results in the incorporation of

organic compounds rich in heteroatoms - like nitrogen, sulfur, or oxygen - through carbonization, activation, or pyrolysis processes. This method often yields heterogeneous material structures that exhibit diverse electrochemical properties due to the varying nature of the precursor materials. Advanced self-doping techniques utilize the inherent chemical composition of biomass, leading to lower environmental impacts and production costs. In contrast, external doping involves the post-synthesis introduction of heteroatoms via gas-phase infiltration or liquid-phase methods, which allows for greater control over the quantity and distribution of dopants within the carbon structure. This precision can result in homogeneously doped materials that demonstrate improved wetting, conductivity, and catalytic properties. For instance, co-doping with nitrogen and boron has shown to enhance the oxygen reduction reaction due to an increased charge density within the carbon matrix, optimizing the overall surface reactivity [146,147]. Additionally, the impact of binary or ternary heteroatom doping strategies has gained attention. Research indicates that double doping with nitrogen and oxygen can increase the number of electroactive sites, thereby improving charge storage efficiency. For example, Yue *et al.* [148] revealed that carbon co-doped with nitrogen and oxygen exhibits elevated energy levels, which favor enhanced electrochemical oxidation and facilitate better electron transfer at the electrode-electrolyte interface.

Furthermore, introducing additional heteroatoms, such as phosphorus, in ternary doping has shown to yield further benefits for overall performance. The effectiveness of certain heteroatom combinations, such as nitrogen, oxygen, and sulfur, is often linked to their electronic and chemical interactions, which significantly modify the fundamental properties of the carbon material. Nitrogen plays a crucial role by increasing charge density through the induction of positive charge, while also enhancing conductivity through effective electron donation. When nitrogen is combined with oxygen, which introduces electronegative characteristics, a balance is achieved that significantly improves the wettability and ionic conductivity of the electrochemical interface. The optimal electrochemical behavior observed in co-doped nitrogen and oxygen systems can be attributed to enhanced hydrogen bonding and altered work functions that facilitate better ion transport. Additionally, doping sulfur with nitrogen may create a more complex architecture conducive to pseudocapacitive behavior, which is a distinct feature that enhances the overall charge storage capability of the electrode. The unique electronic configuration of sulfur allows it to act as a bridge for facilitating electron transfer, while nitrogen enhances conductivity, resulting in a synergistic effect that boosts supercapacitive performance. Recent studies by Li *et al.* [149]; Chen [150] support this concept, revealing that the simultaneous presence of heteroatoms significantly outperforms singly doped materials in terms of electrochemical capacitive performance.

Finally, a comparative analysis of self-doping and external doping methods reveals that both approaches offer distinct advantages in producing high-performance biomass-derived heteroatom-doped carbon materials. The strategic incorporation of heteroatoms, particularly N, O, S significantly enhances the electrochemical properties needed for effective supercapacitor applications. Understanding the mechanisms behind these interactions provides valuable insights for future material development, aiming to optimize performance, sustainability, and commercial viability in energy storage technologies.

Biomass-based heteroatom-doped carbon in practical supercapacitors and commercial feasibility

The integration of biomass-based heteroatom-doped materials into supercapacitor technology holds significant potential for practical applications and commercial viability. These materials utilize the sustainability of biomass resources and exhibit enhanced electrochemical performance due to their engineered structures and chemical properties. Biomass-derived carbon materials, such as those produced from mushrooms and agricultural wastes, demonstrate unique porosity and surface functionalization after treatment, which greatly improve their capacitance and cycling stability. For instance, Wang and Liu [151] reported that porous carbon derived from mushrooms retained approximately 92 % of its initial specific capacitance after 10,000 charge-discharge cycles, showcasing its durability as an environmentally friendly electrode material. Additionally, Zhang *et al.* [152] found that nitrogen and oxygen codoping using tannic acid and cogongrass not only improved surface wettability but

also introduced additional electrochemically active sites, resulting in an increase in overall capacitance. These dual functions are critical for developing high-performance supercapacitors that can meet the evolving performance criteria in commercial applications.

Table 7 The key performance of practical supercapacitor based on different carbon heteroatom sources.

Carbon source	Dopant heteroatom	C_{sp} (F/g)	E_{sp} (W/kg)	P_{sp} (W/kg)	Stability performance	Ref
Bamboo	N	298	15.36	400.1	94.35 % after 10,000	[153]
Peanut shell	N, O	47	13.6	350	83 % after 10,000 cycles	[154]
Phyllanthus emblica leaves	N, O	118	42.6	726	90 % after 1000 cycles	[155]
Litchi peel	N, O		15.36	400	96.9 % after 12,000 cycles	[156]
Benincasa hispida	N, O	260.6	18.1	50	97.7 after 10,000 cycles	[157]
Urea	N, B	193	7.11	479	69 % after 5000 cycles	[158]
Paper fiber	N, S	95.3	13.24	500	91 % after 10,000 cycles	[159]
Urine cow	N, S	165	22.9	502	95.3 % after 5000 cycles	[160]
Artocarpus heterophyllus wood	N, S	262	-	-		[161]
Bagasse	N, S	217	13	100	71 % after 4000 cycles	[162]
Platycladus orientalis leaves	N, O, S	298	17.5	162.5	92 % after 10,000 cycles	[150]
Humulus scandens	N, O, S	335	129	792	78 % after 5000 cycles	[149]

Moreover, multiple heteroatom doping strategies have proven effective in optimizing the microstructure of carbon electrodes, collectively enhancing their electrochemical properties [163]. The combination of different heteroatoms creates a synergistic effect, allowing for various enhanced performances tailored to specific applications. This aligns with the findings of Chen *et al.* [32]; Yin *et al.* [164], who emphasized the role of tailored biomass-derived carbon in the future of energy storage technologies. The ability to adapt material properties through specific doping elements encourages further exploration of biomass-derived materials for customized applications in supercapacitors. The commercial feasibility of these engineered materials is supported by their low production cost and scalability. Taslim *et al.* [165] argue that the straightforward synthesis of biomass-derived activated carbon could promote its widespread adoption in the supercapacitor market. Furthermore, the structural diversity of biomass - ranging from 0D to 3D configurations - provides flexibility in design, meeting various commercial requirements for energy storage solutions [166]. A comprehensive techno-economic analysis by Taslim *et al.* [165] highlighted the advantages of using biomass, including reduced production costs and the potential for large-scale implementation, which are crucial for commercial applications. Advances in this field have shown that biomass-derived materials can achieve energy densities comparable to, or even exceeding, existing commercial alternatives. For example, Zou *et al.* [166] noted that nitrogen-doped hierarchical carbon developed from bamboo fungus exhibited significant energy densities, making it competitive in the market. In addition to the increasing emphasis on sustainability, these materials also address challenges related to resource scarcity and environmental pollution, offering not only a viable alternative but also strategic advantages in the supercapacitor industry [167]. For comparison, **Table 7** presents the key performance metrics of practical supercapacitors, illustrating their potential for product commercialization. Ultimately, the integration of biomass-based heteroatom doping in supercapacitors represents a significant advancement toward sustainable

energy storage solutions. The demonstrated electrochemical performance, combined with a cost-effective production method, positions these materials favorably for commercialization. As research continues to unveil the potential of biomass-derived carbon, its application in supercapacitors emerges as an innovative and crucial step toward sustainable energy technologies.

Challenge and perspective

Biomass-based carbon materials have attracted significant interest as potential candidates for supercapacitor applications. The primary advantage lies in the sustainable and renewable nature, making the materials an environmentally friendly option. However, the commercial application still faces several challenges, particularly in improving electrochemical performance and enhancing competitiveness compared to conventional energy storage technologies. This underscores the need for a comprehensive scientific approach to bridge the gap between laboratory-scale studies and commercial implementation. One of the major challenges in commercializing biomass carbon for supercapacitor is scaling up production. Challenges such as low energy density and limited electrical conductivity have limited the overall performance of these materials. Biomass carbon typically has lower conductivity compared to conventional carbon, negatively affecting supercapacitor performance. According to ongoing studies, the performance of biomass carbon materials remains below that of established energy storage technologies, necessitating further investigation. Innovations in doping strategies and composite formation are essential to enhance electrical conductivity without compromising other important characteristics. The porosity of biomass carbon materials is critical in determining the electrochemical performance of supercapacitor. Well-defined pore structures facilitate increased charge storage capacity and improved ion transport but creating such porous structures poses challenges. Commonly used chemical and physical activation techniques can lead to particle agglomeration during high-temperature treatment, resulting in larger and less-defined pores. Pyrolysis or gasification processes may also introduce interactions with gases such as CO_2 or steam, triggering side reactions that result in non-uniform porosity. Post-conversion treatments, such as washing and drying, also impact the integrity of the pore structure, making the optimization of these processes crucial to prevent pore collapse or clogging.

Fundamental aspects related to the electrochemical behavior of biomass carbon remain poorly understood. Currently, no comprehensive theoretical model accurately describes the carbonization process and the impact on material properties. This knowledge gap limits the ability to design materials with specific desired properties for various applications. Therefore, it is essential to develop theoretical models that can elucidate the mechanistic pathways of carbonization and the relationship between the microscopic structure of the material and the electrochemical performance. The models will provide valuable insights and aid in designing supercapacitor with more efficient and effective performance. The variation in biomass composition presents another significant challenge. Each type of biomass possesses different chemical and physical characteristics that directly influence the quality of the produced carbon. This inconsistency complicates efforts to achieve reproducible results and standardize the production process. Consequently, a systematic understanding of the relationship between biomass sources and the properties of the produced carbon is crucial. In-depth studies into how the chemical composition and morphology of biomass affect the carbonization process and the electrochemical properties of carbon will be instrumental in optimizing supercapacitor performance.

From an economic perspective, using biomass as a carbon feedstock has distinct challenges. The costs associated with preprocessing, activation, and functionalization of biomass carbon tend to be high. Developing cost-effective production processes is crucial for driving the commercialization of this material. The processes should be not only economical but also environmentally friendly and scalable. Therefore, future studies should focus on creating efficient production methods that minimize the use of hazardous chemicals and reduce energy consumption, while maintaining material quality. Biomass carbon typically has a large surface area, but the electrical conductivity remains a significant limitation. The complex structure of biomass carbon often leads to low conductivity, which impairs the performance of supercapacitor.

Using heteroatom doping techniques or forming composites presents promising solutions to limitations. Doping with elements such as nitrogen (N), phosphorus (P), and sulfur (S) has been shown to enhance electrical conductivity and electrochemical performance. Additionally, exploring other dopants such as boron (B), fluorine, silicon, calcium, magnesium, and aluminum could lead to further performance improvements. Each dopant offers unique advantages, for instance, B alters electronic properties, fluorine enhances surface characteristics, and silicon improves charge storage mechanisms through pseudocapacitance.

Optimal electrode design is also crucial for maximizing the performance of biomass carbon-based supercapacitor. Integrating these materials into existing supercapacitor architectures requires a thorough understanding of compatibility with binder materials, current collectors, and electrolyte systems. Continuous advancements in electrode fabrication techniques and material processing are essential to achieve maximum efficiency and performance. Future studies should examine how the pore structure, surface area, and electrical conductivity of biomass carbon influence the overall performance of supercapacitor devices. Exploring biomass carbon for supercapacitor technology offers a compelling approach to tackling the challenges of sustainable energy storage.

It is crucial to broaden the discussion on emerging synthesis methods, commercial challenges, and interdisciplinary collaborations. Innovative synthesis techniques are key to enhancing the performance and reducing the costs of heteroatom-doped carbon materials. Various methods have been proposed, including physicochemical activation, carbonization, pyrolysis, hydrothermal synthesis, chemical injection, external doping, and self-doping. However, the commercial challenges associated with these materials highlight the limitations of these synthesis techniques. While biomass-derived materials offer low production costs, the variability in biomass quality can impact the electrochemical properties of the final carbon products. Additionally, aligning production methods with universal industry standards and certifications is vital for achieving market acceptance. Collaborations among experts in chemistry, materials science, environmental engineering, and industry can foster the development of hybrid systems that incorporate both organic and inorganic components to enhance efficiency. There is significant potential for future work to focus on applied research, where interdisciplinary teams can collaboratively create tailored solutions to address the specific challenges in energy storage applications. This includes designing composite materials that integrate biomass-derived carbon with other functional components, which have shown improved performance metrics.

Future studies should focus on developing robust and optimized supercapacitor with performance that is comparable to or even surpasses conventional technologies. In this context, a comprehensive understanding of material degradation mechanisms, along with initiatives to improve the compatibility of biomass carbon with traditional components, is necessary. The integration of advanced computational models and simulations is also crucial to guide material design. These models can provide valuable insights into structural transformations at the atomic level during charge and discharge cycles, ultimately aiding in material optimization. Furthermore, the relationship between the porosity of biomass carbon and electrochemical performance also warrants further investigation. Hybrid activation techniques that combine physical and chemical processes can produce activated carbons with superior properties. This approach aims to optimize pore size and distribution, thereby enhancing electrochemical activity without compromising material integrity. Heteroatom doping strategies can strategically improve the electrochemical properties of biomass carbon. Although common dopants have been extensively studied, investigating less common dopants such as selenium and tellurium may offer additional benefits in terms of reactivity and surface conductivity. By focusing on these fundamental aspects, future studies can develop strategies to optimize the efficiency and longevity of biomass carbon-based supercapacitor, paving the way for successful integration into next-generation energy storage systems.

Conclusions and future outlook

In conclusion, this review comprehensively elucidates the potential of biomass-derived activated carbon as a candidate for supercapacitor applications, particularly through the incorporation of heteroatoms such as nitrogen (N), oxygen (O), phosphorus (P), sulfur

(S), and boron (B). The significant effects of these dopants on electrochemical performance underscore the importance of both the type and distribution of heteroatoms within the activated carbon matrix. Notably, the doping of N has been shown to enhance electrochemical performance by improving wettability and electron conductivity, which are critical for optimal charge transfer during supercapacitor operation. Research indicates that multi-doping strategies, which involve the combination of different heteroatoms, exhibit synergistic effects that boost the overall electrochemical performance of the materials. However, challenges in achieving precise control over the doping process complicate the characterization of the resulting properties, as variations in dopant type and concentration can lead to substantial differences in performance metrics. Further investigations are warranted to fully understand how various combinations of heteroatoms affect the electrochemical behavior of supercapacitors, particularly while addressing inherent microstructural challenges of biomass-derived carbon materials. Looking forward, it is crucial to develop advanced techniques that ensure effective doping while facilitating improved control over microstructural features, such as pore distribution. Enhanced techniques could lead to uniform pore structures that permit efficient charge and ion transport, thereby significantly improving supercapacitor performance. The shift towards symmetric supercapacitors highlights the need for innovative design strategies that integrate biomass-derived activated carbon with hierarchical pore structures 3D and nanostructure 2D to enhance both energy density and operational range. From a sustainability perspective, the use of biomass waste as a source for activated carbon not only contributes to waste reduction but also aligns with the environmental objectives of utilizing renewable resources. Abundant and low-cost biomass materials offer a promising pathway towards sustainable energy storage solutions while effectively addressing global environmental challenges. Future research should emphasize optimizing synthesis conditions for these biomass-derived materials, ensuring that production processes are efficient and environmentally sound. Establishing frameworks for assessing the environmental impact of these materials throughout their lifecycle is essential for promoting their adoption in commercial systems. Finally, the ongoing exploration of biomass-derived heteroatom-doped carbon with hierarchical pore structures holds significant promise for advancing supercapacitor technologies. Strategic future research initiatives, including improved microstructural control, optimized doping processes, and sustainable production methods, will be pivotal in maximizing the performance and environmental benefits of these advanced materials.

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